

Reforming Fossil Fuel Use  
The Merits, Costs and Risks of Carbon Dioxide Capture and Storage

Kay Damen



# Reforming Fossil Fuel Use

## The Merits, Costs and Risks of Carbon Dioxide Capture and Storage

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Hervorming van fossiel brandstofgebruik  
De voordelen, kosten en risico's van afvang  
en opslag van kooldioxide

(met een samenvatting in het Nederlands)

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Da steh ich nun, ich armer Tor,  
und bin so klug als wie zuvor

– Johann Wolfgang von Goethe (Faust)

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ASU	air separation unit	LNG	liquefied natural gas
ATR	auto thermal reforming	LS	liquid steel
AU	assessment unit (of hydrocarbon reservoirs)	L/S ratio	load-to-surface ratio
AZEP	advanced zero emission power plant	MC	methane conversion
bbl	barrel (of oil)	MCA	Multi-Criteria Analysis
bcm	billion cubic meter	MCFC	molten carbonate fuel cell
BF	blast furnace	MCM	mixed conducting membrane
BOF	basic oxygen furnace	MDEA	methyl diethanolamine
BTL	biomass-to-liquids	MEA	monoethanolamine
CCS	CO <sub>2</sub> capture and storage	MOB	transport sector
CG	coal gasification	MR	membrane reformer or reactor
CHAT	cascaded humidified air turbine	NGCC	natural gas combined cycle
CHP	combined heat and power production	NL	Netherlands
CLC	chemical looping combustion	NO	Norway
COE	cost of electricity	NPV	net present value
COH	cost of hydrogen	O&M	operating and maintenance
CTL	coal-to-liquids	OCM	oxygen conducting membrane
DeNOX	NO <sub>x</sub> removal unit	PC	pulverised coal-fired power plant
DICI	direct injection compression ignition	PEMFC	proton exchange membrane fuel cell
DME	dimethyl ether	POX	partial oxidation
DR	direct reduction	PPC	process plant costs
EAF	electric arc furnace	ppm(v)	parts per million (on volume basis)
ECBM	enhanced coal bed methane recovery	PSA	pressure swing adsorption
EGR	enhanced gas recovery	RES	residential sector
EOR	enhanced oil recovery	RIS	reservoir-induced seismicity
FEP	feature event process	ROW	right of way
FCV	fuel cell vehicle	S/C ratio	steam-to-carbon ratio
FGD	flue gas desulphurisation	SCOT	Shell Claus off-gas treating process
F-T	Fischer-Tropsch	SCR	selective catalytic reduction
GE	Global economy scenario	SER	sorption enhanced reforming
GIS	Geographic Information System	SETS	sorbent energy transfer system
GHG	greenhouse gas	SEWGS	sorption enhanced water gas shift
GT	gas turbine	SMR	steam methane reforming
GTL	gas-to-liquids	SOFC	solid oxide fuel cell
GWP	global warming potential	ST	steam turbine
HAT	humid air turbine	STL	steel production
HHV	higher heating value	TCR	total capital requirement
HMCM	hydrogen mixed conducting ceramic membrane	TFC	total facilities costs
HR	hydrogen recovery	TIT	turbine inlet temperature
HRSG	heat recovery steam generator	TM	Transatlantic markets scenario
HSD	hydrogen separation device	TPC	total plant costs
ICEV	internal combustion engine vehicle	tpd	tonne per day
IDC	interest during construction	UGS	underground gas storage
IGCC	integrated gasification combined cycle	UK	United Kingdom
ITM	ion transfer membrane	UR	ultimate gas/oil recovery
LHV	lower heating value	WGS	water gas shift
		ZEC	zero emission coal



## CHAPTER I INTRODUCTION

### I TOWARDS A SUSTAINABLE ENERGY SYSTEM AND THE ROLE OF CARBON DIOXIDE CAPTURE AND STORAGE

We live in an era in which there is growing concern about climate change caused by human activities and in particular fossil energy consumption. Scientists confront us with their observations of melting glaciers, sea level rise, changing weather patterns and extinguishing species. Although the direct relation between the emission of anthropogenic greenhouse gasses (GHG) and observed climate change is difficult to prove due to the complexity of the climate system, the general scientific consensus is that global warming observed over the past few decades is for a substantial part attributable to human activities [1,2]. Because the risks of climate change could jeopardise the existence of human societies and ecosystems, an international agreement has been reached to stabilise atmospheric GHG concentrations at a level that would prevent dangerous anthropogenic interference with the climate system [3]. To keep the biophysical and socio-economic impacts, as well as the distribution of impacts within acceptable limits, it is widely agreed that the globally averaged temperature rise should not exceed 2°C above pre-industrial levels. In order to limit global warming to 2°C, the atmospheric carbon dioxide (CO<sub>2</sub>) concentration probably needs to be stabilised at a level of 450-550 ppmv<sup>1</sup> [4], and possibly even below<sup>2</sup>. Stabilisation of atmospheric CO<sub>2</sub> concentrations at 450 ppmv would require global anthropogenic CO<sub>2</sub> emissions to drop below the 1990 level within a few decades [5]. By 2050, CO<sub>2</sub> emission reductions of 50% or more may have to be realised in comparison to the 1990 level. A recent study framed the costs of achieving stabilisation between 500 and 550 ppmv at 1% of global GDP if we take action now [8]. If we fail to do so, the overall costs and risks of climate change may be equivalent to losing 5-20% of global GDP each year.

1. Parts per million by volume.

2. The temperature increase at equilibrium relative to pre-industrial levels when stabilising CO<sub>2</sub> concentrations at 450 ppmv ranges from 1.5-3.9°C and the probability to exceed 2°C would be 38% [5,6]. A recent study using climate sensitivity estimates from 11 different studies shows that the range may be 0.8-6.4°C for a stabilisation level of 450 ppmv, and the probability to exceed 2°C would be 26-78% [7].

Reduction of CO<sub>2</sub> emissions could be realised by means of a diverse portfolio of options, including:

- Reduction of energy and material demand by efficiency improvements and changes in consumption patterns
- Afforestation and reduction of deforestation
- Increased use of renewable energy sources
- Increased use of nuclear fission technology (and possibly nuclear fusion in the long term)
- Decrease of the carbon intensity of fossil fuel use by shifting towards less carbon-rich fuels
- Carbon dioxide capture and storage (CCS). CCS can be defined as the separation and capture of CO<sub>2</sub> produced at large stationary sources, followed by transport and storage in geological reservoirs, the ocean

or minerals in order to prevent its emission to the atmosphere. CCS is also referred to as CO<sub>2</sub> sequestration.

Improving energy efficiency should be the first priority in a strategy to reduce GHG emissions and our dependency on fossil fuels, with all associated negative impacts [9]. Simultaneously, the energy supply could be decarbonised by enhancing the penetration of renewable energy such as wind, solar and biomass, and nuclear energy. However, the speed and extent at which energy consumption can be reduced and renewable and nuclear capacity be installed may not be sufficient to achieve deep reductions in GHG emissions by 2050 and preserve us from severe impacts of climate change [4]. Well-known barriers in saving energy are the dispersed potential of energy efficiency measures among different sectors, low priority in firms due to the relatively low energy expenditures in many processes and activities, and ill-informed consumers and companies [10,11]. The implementation of renewable and nuclear energy is hampered by a variety of factors, most notably costs, land-use, environmental impacts and safety.

As a consequence, fossil fuels will remain the backbone of our energy supply for many decades to come [9,12]. The reserves and resources of oil and natural gas are enough to last for decades [13]; the world's proven reserves could sustain current oil production levels for 42 years and current gas production rates for 64 years [9]. Proven coal reserves are sufficient to sustain current production for 164 years [9]. Unconventional hydrocarbon resources extend these projections even further [9,13]. As coal is so abundant and can be extracted at low costs, many coal-fired power plants are and will be installed. Nobody has failed to notice the speed at which new coal-fired capacity is being installed in China, fuelled by the abundant national reserves. Also large investments in new capacity are forecasted in the OECD world, as many gigawatts of coal-fired and nuclear power generation capacity are to be replaced the coming decades [9]. Given the long lifetime of such assets, the future 'carbon' burden will be significant if we continue to use conventional fossil fuel-fired power plants, even if most efficient technology is applied.

CCS enables the continuation of the (inevitable) use of fossil fuels and its infrastructure, while simultaneously realising strong CO<sub>2</sub> emissions reduction before 2050. When applied to large-scale biomass conversion plants, even negative emissions could be achieved. In the meantime we can develop and implement energy-efficient processes, renewable and possibly nuclear energy technologies required for a sustainable energy system in the long term. Apart from electricity, hydrogen may become an important energy carrier in such a sustainable energy future. Again, CCS may fulfil a bridging function. The coming decades, hydrogen can be produced from fossil fuels with CCS at relatively low costs. In this way, we can gradually develop a hydrogen infrastructure that can be used in the long term when hydrogen produced from renewable/nuclear energy may become competitive with fossil fuel-derived hydrogen [14].

Apart from its bridging function and potential to realise large emission reductions the coming decades, there are more reasons to consider the deployment of CCS. First, inclusion of CCS in a mitigation portfolio could reduce the costs of stabilising atmospheric CO<sub>2</sub> concentrations at an acceptable level by 30% or more [15]. Second, the use of coal in combination with CCS makes it possible to enhance the security of energy supply without increasing GHG emissions, as coal reserves are distributed more evenly than oil and natural gas. In addition, oil recovery of nearly depleted reservoirs could be enhanced by injecting CO<sub>2</sub>. In the longer term, injection of CO<sub>2</sub> into nearly depleted gas reservoirs or coal seams may result in increased gas production. Like any other technology, CCS has a number of drawbacks as well. CCS differs from the other options in that it prevents CO<sub>2</sub> to be emitted to the atmosphere, instead of preventing the production of CO<sub>2</sub> itself. In that sense, CCS is often (and not unjustly) regarded as an end-of-pipe technology, and therefore a non-structural solution to the problems of fossil fuel use. To put it differently, CCS is merely a technical solution to buy time and enable clean(er) fossil fuel use by reducing GHG emissions. CCS also requires considerable amounts of energy, mainly for CO<sub>2</sub> capture and compression. As a consequence, more primary energy is needed to produce the same output. Hence, CCS induces a stronger reliance on fossil fuels, accelerating their depletion, and increases the specific emission of air pollutants, the production of waste and consumption of resources such as ammonia and limestone [16]<sup>3</sup>.

3. In practice this effect will (partly) be compensated by the fact that new efficient units with CCS will replace older and less efficient units. Some CCS technologies also enable co-capture of air pollutants such as SO<sub>2</sub> and H<sub>2</sub>S or combustion without N<sub>2</sub> resulting in low NO<sub>x</sub> levels [15].

Another issue that does not comply with the principles of sustainability is that it requires intervention in the geosphere for a period that exceeds the ‘lifetime’ of governments and companies. Apart from the liability issues that arise here, we basically burden at least some generations after us with large amounts of stored CO<sub>2</sub>. Although it is not as hazardous as nuclear waste, there are certainly risks associated with CO<sub>2</sub> storage, most notably leakage.

In conclusion, there appears to be no silver bullet to mitigate CO<sub>2</sub> emissions by now, which urges us to consider all options. The risk on large, irreversible impacts due to climate change does not allow us to be fastidious and exclude certain options on forehand. It is recommended to ‘pursue a portfolio of technologies as this will greatly reduce the risk and potentially the costs, if one or more technologies fail to make the expected progress’ [10]. CCS is a card we could play when the urgency of climate change forces us to deeply cut in GHG emissions. It is basically an intermediate solution in the evolution towards a sustainable energy system in the long term. This thesis will focus on the techno-economic potential of reforming fossil fuel use by means of carbon dioxide capture and geological storage.

## 2 TECHNICAL DESCRIPTION OF CARBON DIOXIDE CAPTURE AND STORAGE

The ccs chain consists of three main components being  $\text{CO}_2$  capture (and compression), transport and storage. Each of these components is being applied in industry, yet hardly in integrated schemes with the intention to mitigate  $\text{CO}_2$  emissions. ccs is applicable to large stationary sources, such as power plants, refineries, iron and steel mills, cement kilns, and (petro)chemical complexes.

Costs of ccs at small  $\text{CO}_2$  sources are generally considered prohibitive. Nevertheless,  $\text{CO}_2$  emissions occurring in decentralised power and heat production, the transport sector, households and the commercial sector could be reduced by application of low or zero-carbon energy carriers such as electricity, hydrogen or synthetic fuels produced from carboniferous fuels with ccs [15].

### 2.1 $\text{CO}_2$ CAPTURE

Carbon dioxide is produced by the oxidation (combustion) or transformation of carboniferous materials, and separated during natural gas processing. In some processes, among which the production of ethylene oxide by oxidising ethylene and ethanol by fermenting sugars, a pure  $\text{CO}_2$  stream is produced. However, in most processes,  $\text{CO}_2$  is present in a gas stream with other components such as  $\text{N}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{H}_2$ ,  $\text{CO}$  and/or  $\text{CH}_4$ . In order to produce a nearly pure  $\text{CO}_2$  stream required for transport and storage,  $\text{CO}_2$  needs to be separated, physically or chemically, from the gas stream. The choice of the capture technology depends on the scale of the source (volume),  $\text{CO}_2$  concentration, pressure, temperature and gas stream composition. Generally, four approaches to capture  $\text{CO}_2$  are distinguished<sup>4</sup> (see also Table 1) [15]:

1. *Capture from industrial process streams.* Today,  $\text{CO}_2$  separation is applied in several industrial processes due to product/process specifications. At refineries and petrochemical complexes, fossil fuels are converted into synthesis gas (syngas), consisting predominantly of  $\text{H}_2$  and  $\text{CO}$ . After a shift reaction, in which  $\text{CO}$  is reacted with steam to produce  $\text{CO}_2$  and more  $\text{H}_2$ ,  $\text{CO}_2$  is removed by means of physical or chemical absorption to produce  $\text{H}_2$ . Hydrogen can be used in refinery processes or for ammonia production. Alternatively,  $\text{CO}$  is shifted and  $\text{CO}_2$  removed to produce the optimal ratio  $\text{CO}:\text{H}_2$  for the production of methanol or synthetic fuels [17]. In gas processing plants,  $\text{CO}_2$  present in natural gas is removed by means of chemical absorption or membranes in order to meet transport specifications and market requirements [18].

$\text{CO}_2$  could also be captured in cement, iron and steel production [19].

Cement is produced by thermally decomposing limestone into  $\text{CaO}$  and  $\text{CO}_2$ .  $\text{CO}_2$  could be captured from the off-gas of the cement kiln.

Iron is produced by reducing ore by means of coke in a blast furnace.

In integrated steel mills, the carbon present in pig iron is subsequently oxidised in a basic oxygen furnace.  $\text{CO}_2$  can be captured from the blast furnace gas and the basic oxygen furnace gas.

4. The technologies to capture  $\text{CO}_2$  from industrial processes are similar or identical to those applied for  $\text{CO}_2$  capture from combustion processes.

2. *Post-combustion capture.* The principle of post-combustion capture is to remove  $\text{CO}_2$  from flue gas produced by the combustion of fossil fuels or biomass. The process can be applied to newly built plants or existing plants (retrofit). The leading technology in post-combustion capture is chemical absorption. This technology is already practised on a small scale to produce high-purity  $\text{CO}_2$  for urea production and the food and beverage industry [20].
3. *Pre-combustion capture.* In pre-combustion schemes, syngas is produced by reforming or partial oxidation of natural gas or by gasification of coal, oil residues or biomass.  $\text{CO}_2$  is then captured from shifted syngas, and  $\text{H}_2$  rich gas is converted in a boiler, furnace, gas turbine, engine or fuel cell. Although the complete integration of syngas production, shift,  $\text{CO}_2$  capture and combustion of  $\text{H}_2$  rich gasses is not implemented yet, syngas production and  $\text{CO}_2$  capture from shifted syngas is a common practice in the chemical industry. In addition, approximately 4  $\text{GW}_e$  of oil and coal-based integrated gasification combined cycles (IGCC) is installed worldwide [21]. Another 144  $\text{GW}_e$  is expected to be commissioned up to 2030 [9]. IGCC may become more attractive in a carbon-constrained world as the energy penalty of adding a shift reaction and subsequent  $\text{CO}_2$  capture is relatively small [15].
4. *Oxyfuel combustion.* In this process, fuel is combusted in an atmosphere of mainly oxygen, resulting in a concentrated stream of  $\text{CO}_2$  and steam, which can be separated easily by condensation. Boilers and furnaces could be retrofitted for oxyfuel combustion, although this scheme is still in the demonstration phase [15]. In the longer term, oxyfuel combustion could also be deployed in Brayton cycles, but this requires the development of new turbines [22].

TABLE I. Overview of  $\text{CO}_2$  capture options (based upon [15])

Capture option	Principle	Status
Capture from industrial process streams	$\text{CO}_2$ separation from shifted syngas ( $\text{H}_2$ ), natural gas, blast furnace gas, cement kiln off-gas	Commercially applied (syngas, natural gas) Research phase (blast furnace gas, cement kiln off-gas)
Post-combustion capture	$\text{CO}_2$ - $\text{N}_2$ separation	Applied on small scale
Pre-combustion capture	$\text{CO}_2$ - $\text{H}_2$ separation	Commercially applied in chemical industry. No integrated scheme with $\text{H}_2$ combustion
Oxyfuel combustion	$\text{O}_2$ - $\text{N}_2$ separation (to avoid $\text{N}_2$ - $\text{CO}_2$ mixing) and combustion with $\text{O}_2/\text{CO}_2$	Oxygen production: commercially applied Combustion in $\text{O}_2/\text{CO}_2$ : demonstration phase

A power plant equipped with  $\text{CO}_2$  capture (and compression) consumes about 10-40% more energy than an equivalent plant without capture [15]. The challenge in capturing  $\text{CO}_2$  lies in the development of

separation technologies with minimal energy requirements and costs. In this respect, it is important to notice that specific energy requirements of CO<sub>2</sub> separation increase with decreasing CO<sub>2</sub> partial pressure in the gas stream.

## 2.2 CO<sub>2</sub> TRANSPORT

Unless the CO<sub>2</sub> source is located on top of the storage reservoir, CO<sub>2</sub> needs to be transported. Large-scale CO<sub>2</sub> transport can be performed by pipeline or ship. In both transport modes, the density of CO<sub>2</sub> has to be increased after it has been captured, either by compression for transport using pipelines or by liquefaction for transport using ships. In pipelines, CO<sub>2</sub> is transported most economically as dense fluid at pressures of minimally 80 bar<sup>5</sup>. In the USA, the oil industry has at least 20 years of experience in operating CO<sub>2</sub> pipelines. Approximately 2500 km of pipeline, with a capacity over 40 mt CO<sub>2</sub>/yr, transports CO<sub>2</sub> from natural and anthropogenic CO<sub>2</sub> sources to oil fields for enhanced oil recovery [15]. Transport by ship appears attractive for large distances over sea. An advantage of transport by ship is that no large upfront investments are needed. In addition, ships offer a flexible CO<sub>2</sub> supply as they can collect CO<sub>2</sub> from various sources and transport it to several sinks. This may be particularly useful when a changing demand pattern for CO<sub>2</sub> exists, as in enhanced oil recovery applications. CO<sub>2</sub> transport by ship is not applied extensively; only a few small ships are in use today [15].

5. In order to avoid phase transitions, pressures should be kept well above the supercritical point (31°C, 74 bar). Above the supercritical point, CO<sub>2</sub> has the flow properties of a gas and a density close to that of liquid CO<sub>2</sub> [23].

## 2.3 GEOLOGICAL STORAGE

For underground storage of CO<sub>2</sub>, sedimentary basins<sup>6</sup> are most suitable. Each reservoir should meet several requirements to guarantee optimal and safe CO<sub>2</sub> storage [25,26]:

- The storage volume, depending on porosity and thickness, should be sufficient for the planned quantity of CO<sub>2</sub> to be injected
- Permeability should be sufficiently high to allow injection
- Generally, only reservoirs below 800 m are considered for CO<sub>2</sub> storage, because at this depth CO<sub>2</sub> is in supercritical state, where the density of CO<sub>2</sub> ensures optimal storage
- The density of supercritical CO<sub>2</sub> is 50-80% the density of water. Hence, CO<sub>2</sub> will rise up due to buoyancy effects. In order to retain CO<sub>2</sub> for a long period, possibly in the order of thousands of years, and minimise leakage to other formations, the reservoir should in general be sealed with a low-permeability cap rock (clay, salt).

6. A depression in the crust of the earth formed by plate tectonic activity in which sediments accumulate [24].

The reservoirs considered for CO<sub>2</sub> storage include:

1. *Hydrocarbon reservoirs* are generally suitable for CO<sub>2</sub> storage, because they have a proven seal that retained oil, gas and often CO<sub>2</sub> for millions of years. Due to their economic value, oil and gas fields are well studied and characterised. Therefore, relatively accurate estimations of storage capacities can be performed [15]. In many of these reservoirs, there is still some oil and/or gas present, which could

- not be produced cost-effectively by pressure depletion and water flooding. By injecting CO<sub>2</sub> into oil reservoirs, oil is mobilised through miscible or immiscible displacement, which may increase its recovery. This process is referred to as enhanced oil recovery (EOR) using CO<sub>2</sub>, a process which has been proven on commercial scale, primarily in the USA [27]. CO<sub>2</sub> injection in nearly depleted gas fields might cause some enhanced gas recovery (EGR), but this is not proven yet.
2. *Deep saline aquifers* are widely distributed carbonate and sandstone formations filled with saline or brackish water. In contrast to hydrocarbon reservoirs, aquifers generally have not proven the ability to contain large amounts of gasses. Although these formations offer an enormous storage potential, estimations on the storage potential are rather uncertain as many of these structures have not been studied so extensively as hydrocarbon reservoirs [15].
  3. *Coal seams* that are too deep to be mined are potential CO<sub>2</sub> storage reservoirs. When injecting CO<sub>2</sub> in a coal seam, CO<sub>2</sub> is adsorbed to the coal matrix, replacing coal bed methane (also known as mine gas), which could be recovered and used to offset the costs of CCS. This process, also known as enhanced coal bed methane (ECBM) recovery, has so far only been tested in demonstration projects [28,29]. Today, only conventional CBM production (by pumping formation water causing pressure depletion) has been applied successfully.

TABLE 2. Overview of geological storage options (after [15])

Storage option	Global capacity (Gt CO <sub>2</sub> )	Status of CO <sub>2</sub> injection	Pros and cons
Discovered oil and gas fields	675-900	Proven in commercial projects	<ul style="list-style-type: none"> <li>- Well-characterised reservoirs</li> <li>- Proven cap rock</li> <li>- Potential for enhanced oil/gas recovery</li> <li>- Often penetrated by a large number of wells, enhancing risk of CO<sub>2</sub> leakage</li> </ul>
Deep saline aquifers	At least 1000, but possibly up to 10 <sup>4</sup>	Proven in a commercial project <sup>a</sup>	<ul style="list-style-type: none"> <li>- Small knowledge basis</li> <li>- Potentially enormous storage capacity</li> <li>- Presence and integrity of seal uncertain</li> </ul>
Unminable coal seams	3-200	Demonstration phase	<ul style="list-style-type: none"> <li>- CO<sub>2</sub> adsorbs to coal matrix, thereby replacing CH<sub>4</sub> that can be recovered</li> <li>- Coal might swell, reducing permeability</li> <li>- Depressurisation or future mining activities may cause adsorbed CO<sub>2</sub> to be released</li> </ul>

<sup>a</sup>It is important to realise that much of the experience is acquired in the Sleipner project, where CO<sub>2</sub> is injected into an optimal aquifer given its size, permeability and extensive seal [31]. It is the question how large-scale CO<sub>2</sub> injection would evolve in more typical structures.

Geologists estimate the likely<sup>7</sup> worldwide technical storage capacity at roughly 2000 Gt CO<sub>2</sub> [15] (see Table 2). The methods to estimate these potentials are rather simple though, and for several regions in the world there is hardly any information. It is concluded that the likely storage capacity seems sufficient to cover the economic CCS potential of 220-2200 Gt CO<sub>2</sub> in scenarios to stabilise atmospheric CO<sub>2</sub> concentrations at 450-750 ppmv [15]. However, some regions may face a lack of storage capacity as reservoirs are not distributed homogeneously across the world. In addition, there are constraining factors that may reduce the technical storage capacity. Some reservoirs may not be suited from a geotechnical perspective, for instance due to low permeability, doubtful seal quality, presence of non-sealing faults or tectonic activity. The latter three may cause CO<sub>2</sub> leakage out of the reservoir, and possible seepage into the atmosphere. As many hydrocarbon fields are penetrated by exploration and production wells, some of them in bad conditions, these reservoirs may also be susceptible to leakage along the wells [30]. Finally, some fields may simply be too small for storage or are suitable for alternative applications such as underground gas storage or the production of geothermal energy.

7. Probability of 66-90%.

#### 2.4 OCEAN STORAGE

By injecting CO<sub>2</sub> into the vastness of the deep ocean, the slow natural process of dissolution is accelerated in order to reduce the peak in atmospheric CO<sub>2</sub> concentrations. CO<sub>2</sub> would be isolated from the atmosphere for centuries. There are, however, concerns with respect to the environmental and ecological impacts caused by the acidifying effect of CO<sub>2</sub> [15]. This is one of the reasons why ocean storage has not been applied or demonstrated yet; so far only laboratory experiments and modelling studies have been performed.

#### 2.5 MINERAL CARBONATION

The idea behind mineral carbonation is to increase the rate of weathering, a reaction occurring in nature. In this exothermic process, minerals such as calcium and magnesium oxide present in silicate rocks react with CO<sub>2</sub> to form (stable) carbonates. Industrial waste like slags and ashes, which contain small quantities of CaO and MgO, could also be used for this purpose, as has been shown in demonstration units [15]. The carbonate minerals can be disposed in subsurface reservoirs or be re-used, e.g. for construction purposes. As CO<sub>2</sub> is stored in solid form, there is no need for monitoring as required in geological storage. In order to increase reaction rates, the minerals are prepared or additives are used, which are rather energy-intensive processes. At present, the energy consumption of a plant that captures CO<sub>2</sub> and stores it in mineral carbonates would require 60-180% more energy than the equivalent plant without CCS [15]. Research in this area is focussing on reducing the energy use and costs by optimising reaction conditions and searching for alternatives with more favourable kinetics.

## 3 CURRENT STATUS OF CCS

The concept of CO<sub>2</sub> capture and storage into geological reservoirs as greenhouse mitigation option is relatively young. Apart from various research and demonstration projects, only a few commercial, large-scale projects with the purpose of underground CO<sub>2</sub> storage exist to date (see Table 3). Most of these projects involve CO<sub>2</sub> removal during natural gas processing, because CO<sub>2</sub> needs to be separated anyway. A number of plans have been proposed for integrated CCS schemes, including CO<sub>2</sub> capture from power or industrial plants. The projects cover a wide range of fuels (bituminous coal, lignite, natural gas and petroleum coke) and capture technologies (post-combustion capture, pre-combustion capture and oxyfuel combustion).

TABLE 3. Overview of ongoing and proposed commercial CCS projects (after [15,32] and various internet sources).

Figures for proposed projects are indicative. Tba = to be assessed

Project (developer)	Location	Start up	Injection rate (t CO <sub>2</sub> /day)	Capture	Storage
Sleipner	Norway	1996	3000	Natural gas processing	Aquifer
Weyburn	USA	2000	3-5000	Synfuel production	CO <sub>2</sub> -EOR
In Salah	Algeria	2004	3-4000	Natural gas processing	Gas field
Salt creek	USA	2004	5-6000	Natural gas processing	CO <sub>2</sub> -EOR
Snøhvit	Norway	2006	2000	Natural gas processing	Aquifer
<i>Planned/proposed projects</i>					
Gorgon	Australia	2009	~10,000	Natural gas processing	Aquifer
Centrica	UK	2009	Tba	IGCC pre-combustion (coal + petcoke?)	Tba
DF1 (Peterhead-Miller)	UK	2009	3300	NGCC pre-combustion	CO <sub>2</sub> -EOR
Mongstad	Norway	2010	Max. 3600	NGCC post-combustion	CO <sub>2</sub> -EOR
Powerfuel	UK	2010	Tba	IGCC pre-combustion (coal)	Tba
Stanwell	Australia	2010	Tba	IGCC pre-combustion (coal)	Aquifer?
Halten	Norway	2010/11	~6800	NGCC post-combustion + MeOH production	CO <sub>2</sub> -EOR
Nuon Magnum	Netherlands	2010/11 <sup>a</sup>	Tba	IGCC pre-combustion (coal, biomass, natural gas)	Tba
EON	UK	2011	Tba	IGCC pre-combustion (coal + petcoke?)	Tba
DF2 (Carson)	USA	2011	11,000	IGCC pre-combustion (petroleum coke)	CO <sub>2</sub> -EOR
Siemens	Germany	2011	Tba	IGCC pre-combustion (coal)	Tba
Scottish and Southern Energy	UK	2011/12	Tba	PC post-combustion (capture ready)	Tba
Futuregen	USA	2012	2700-5500	IGCC pre-combustion (coal)	Tba
Hypogen	EU	2012	Tba	Tba	Tba
Saskpower	Canada	2012	8000	PC oxyfuel	Tba

rwe	Germany	2014	Tba	igcc pre-combustion (coal)	Tba
rwe	UK	2016	Tba	pc post-combustion (capture ready)	Tba
Karsto GE & Polish utility	Norway	Tba	Tba	ngcc post-combustion	CO <sub>2</sub> -EOR
	Poland	Tba	Tba	igcc pre-combustion (coal)	Tba
nZEC	China	Tba	Tba	igcc pre-combustion (coal)	Tba
Hazelwood	Australia	Tba	Tba	Lignite post-combustion	Tba

<sup>a</sup> CO<sub>2</sub> capture is projected to start 2 years later

However, injection of gasses into geological formations is not new. As was mentioned earlier, the oil industry has several decades of experience with CO<sub>2</sub> injection into nearly depleted oil reservoirs to enhance oil recovery. In Canada, acid gas (a mixture of CO<sub>2</sub> and H<sub>2</sub>S) is separated from natural gas and disposed in underground formations [33]. In various countries, natural gas is injected in depleted gas fields and aquifers with the purpose of temporary or seasonal storage [34]. The technology for large-scale CO<sub>2</sub> storage has many similarities with EOR, acid gas injection and underground gas storage. We could learn from these industrial analogues and make use of the (risk) standards developed here. Finally, we should not forget the lessons provided by nature itself, as there are several natural analogues at our disposal to gain insight into the behaviour of CO<sub>2</sub> in geological formations. Natural analogues are particularly important to help understand the risks associated with CO<sub>2</sub> leakage and seepage into the atmosphere [35,36].

#### 4 CCS IN THE NETHERLANDS

This thesis is partly dedicated to the deployment of CCS in the Netherlands. Specific characteristics related to CO<sub>2</sub> sources and sinks, the potential for alternative mitigation options, as well as recent CCS developments in this country are highlighted here.

##### 4.1 GENERAL CONTEXT AND ROLE OF CCS IN REDUCING GHG EMISSIONS

In the coming decades, CCS may play a significant role in decarbonising the Dutch energy sector, as there are numerous large CO<sub>2</sub> sources and potential sinks (see Figures 1 and 2). The storage potential (>11 Gt CO<sub>2</sub>) is sufficient to cover at least a few decades up to one century of current CO<sub>2</sub> emissions produced in the energy and manufacturing industry (nearly 100 Mt/yr), depending on the availability of the Groningen gas field. The Netherlands also has decades of industrial experience with the transmission, distribution and processing of natural gas, which could be valuable in developing a CCS infrastructure.

A recent scenario analysis that assessed the potential and costs of different measures to reduce Dutch ghg emissions in 2020 illustrated the importance of CCS [37]. The general conclusion was that energy saving, CCS and nuclear energy have a large potential at relatively low

costs. It was found that a 25% reduction in 2020 with respect to the 1990 level could only be achieved with CCS. The costs to reduce ghg emissions by 6-15% below the 1990 level will be 450-1800 mt/yr higher than without CCS, as we would have to rely on more expensive energy efficiency measures and renewable energy technologies.

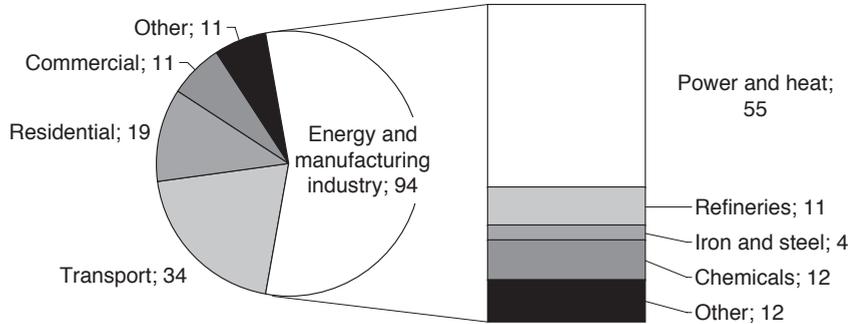


FIGURE 1. CO2 emission breakdown for the Netherlands in 2003 [38]  
All numbers are in mt/yr. Total CO2 emissions were 170 mt

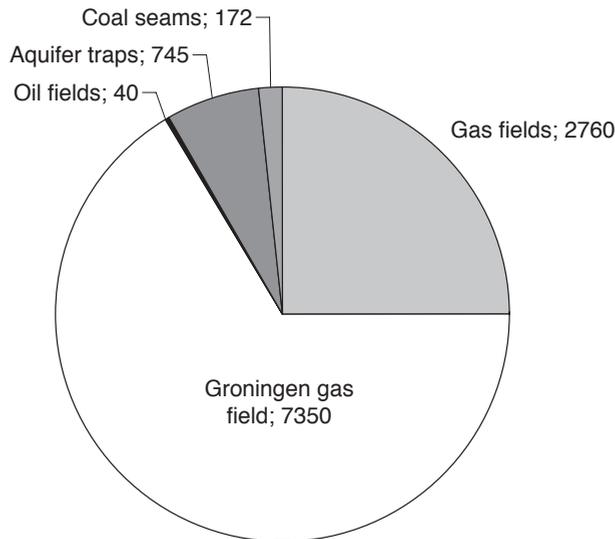


FIGURE 2. Geological storage capacity for the Netherlands in mt [39]

The official policy note on CCS composed by the Dutch Ministry of Economic Affairs in 2003 acknowledged the role of CCS for the medium and long term, but refrained from formulating a clear vision [40]. In recent years, the limitations in alternative GHG emissions and the specific advantages of CCS have become more apparent. Together with recent international developments in energy and climate policy, R&D and commercial projects, this has caused a growing interest in CCS. In a broad strategic context, CCS has been officially recognised as one of the

transition paths to reform the Dutch energy system [41,42]. The energy transition is an ambitious long-term dynamic process to realise a sustainable energy supply by 2050. One of the objectives is to reduce CO<sub>2</sub> emissions by 50% in 2050 compared to the 1990 level. The emission reduction potential of CCS by 2050 has been estimated at 25-30 Mt CO<sub>2</sub> in energy production, 4-8 Mt in the transport sector and 0(!) Mt in the industry [41]. CCS, like nuclear energy, is also considered as transitional technology in an 'acceleration package' necessary to achieve short-term CO<sub>2</sub> reduction aims. It is the question though whether CCS could be implemented in the short term, as there are still a number of issues to be resolved, which will be discussed briefly in Section 5. This thesis will look in more detail on the possibilities of CCS deployment in time.

#### 4.2 PROJECTS AND DEVELOPMENTS

In order to enhance knowledge development and streamline CCS research activities, a national research programme (CATO) was initiated in 2004, encompassing a wide range of activities related to capture, storage, mineralisation, monitoring, system integration and communication [43]. Within CATO, a pilot project is being prepared to test new technologies for post-combustion CO<sub>2</sub> capture from flue gas of a pulverised coal-fired power plant.

In 2004, Gaz de France started the injection of approximately 20 kt CO<sub>2</sub>/yr separated from natural gas into the nearly depleted K12-B field located in the North Sea [44]. The injection amount could be scaled up to approximately 0.4 Mt CO<sub>2</sub> per year. Apart from gaining experience with CO<sub>2</sub> storage, the project aims to study the feasibility of enhanced gas recovery. Although CO<sub>2</sub> injection was performed successfully, no measurable improvement in gas production of the tested compartment as a consequence of CO<sub>2</sub> injection was observed by the end of 2005 [45]. In the Energy Valley project, possibilities are investigated to strengthen the economic activities in the Northern provinces, in which energy-related activities are the main focus [46,47]. An important role is assigned to CCS as this region is characterised by a great number of gas fields that reach the end of their productive life and are ideal candidates for CO<sub>2</sub> storage.

Recently, additional funds have been allocated to realise one or more of the following demonstration projects:

- Storage of 0.6 Mt CO<sub>2</sub>/yr produced by a gasification unit at the Shell refinery into a nearby hydrocarbon reservoir. One of the candidate fields was recently rejected due to concerns over inaccessible abandoned wells.
- Construction of a 50 MW<sub>e</sub> zero emission power plant (ZEPP) using an innovative oxyfuel combustion technology. The CO<sub>2</sub> produced, approximately 0.2 Mt/yr, would be injected into a nearly depleted gas field with a potential of enhanced gas recovery.
- CO<sub>2</sub> capture from syngas slipstream of the IGCC in Buggenum. The main purpose is to investigate the impact of degradation of elements

8. Shifting syngas is current practice in the chemical industry, but the energy loss due to steam consumption is of less relevance in comparison to an IGCC unit where steam consumption is attended by a lower electricity output.

present in the syngas on the solvents needed to capture CO<sub>2</sub> [48]. Also the shift reaction could be tested and optimised<sup>8</sup>. The results of these tests will be accounted for in the design of a similar, yet larger IGCC unit with CO<sub>2</sub> capture that is currently in the planning phase (Magnum project).

The insights and experience generated in the R&D and demonstration projects previously discussed should be taken into account when realising full-scale projects in the coming years. By 2010, roughly 3 GWe of new capacity is needed to cover the increase in electricity demand and replace old units that will be decommissioned [49]. The demand for new capacity in the coming years seems too early for nuclear and too large for renewables, and therefore an important role is foreseen for fossil power plants, especially coal-fired units. Table 4 gives an overview of the plans announced for the construction of new coal-fired capacity. In all plants, the option for CO<sub>2</sub> capture by making the plants capture ready is considered [49].

TABLE 4. Planned coal-fired capacity in the Netherlands (2006)

Company	Technology	Location	In operation	Capture rate (mt/yr) <sup>a</sup>
Electrabel	600/800 MWe PC	Maasvlakte	2011	2.7-3.7
Eon	1100 MWe PC	Maasvlakte	2011	5
Nuon (Magnum)	1200 MWe IGCC	Eemshaven	2010/11	1.5
RWE	1600-2200 MWe PC	Eemshaven	2011	7.3-10.1
Total	7000-7800 MWe			17-20

<sup>a</sup> Assuming an electric efficiency of 46% and a load factor of 80% for all units. For the pc units, the CO<sub>2</sub> capture rate is 90% using MEA; no biomass co-firing is considered. For the multifuel IGCC, we assume 40% of electric output is produced from natural gas and 60% from coal/biomass [48]. From the syngas stream, 40% of the produced CO<sub>2</sub> is assumed to be captured [50].

## 5 GAPS IN KNOWLEDGE

Previous sections indicate that CCS may become a serious GHG emission reduction option in industrialised areas such as the Netherlands, provided that stable economic incentives are in place and legal and regulatory issues are resolved. In addition, a number of gaps in knowledge and uncertainties need to be addressed to facilitate decision-making on the role of CCS and make CCS an economic, reliable and safe GHG emission reduction option. Some major issues are (see also [15]):

1. *Techno-economic performance of CO<sub>2</sub> capture.* In the previous two decades, scientists and engineers identified numerous options for CO<sub>2</sub> capture and assessed their thermodynamic performance and costs. The results that were produced need to be standardised in order to compare the different options. In addition, the calculated figures need to be verified by full-scale demonstration of integrated CCS

- schemes. Commercial application will be crucial in bringing down the costs, together with R&D in innovative options that promise highly-efficient, low cost CO<sub>2</sub> capture.
2. *Source-sink matching.* For several regions in the world, Geographic Information Systems (GIS) have been applied to match CO<sub>2</sub> sinks and sources (see e.g. [51,52]). These studies show whether a region has a lack or excess in storage capacity and what infrastructure requirements are needed. Most studies, however, lack the temporal dynamics in sources and sinks. Focus is on existing CO<sub>2</sub> sources, whereas CCS will most likely be deployed to new sources that are not necessarily installed at the same locations. Also the availability of hydrocarbon reservoirs in time is generally not accounted for. Hence the challenge lies in developing more advanced approaches that incorporate dynamics in time and enable calculations on multiple sink-source combinations and networks.
  3. *Capacity and risks of geological storage.* Earth-scientists have produced a first estimation of global and regional storage capacities [15]. We do need, however, a more accurate estimation of these capacities together with more detailed assessments of the associated risks. In this way, the feasibility of CO<sub>2</sub> storage for specific reservoirs can be evaluated and the storage capacity can be better judged. As the storage capacity available for CO<sub>2</sub> may be limited due to competition with UGS and geothermal energy, the demand and potential of these alternative applications need to be analysed as well. By accounting for the risks and alternative applications, as well as economic, legal and regulatory barriers, ultimately more realistic and viable storage capacities (see [53]) can be derived.
  4. *Role of CCS in GHG emission reduction strategy.* As we are on the threshold of CCS deployment, it is about time to come up with strategies for large-scale CCS deployment as part of the transition towards a more sustainable energy system. For this purpose, we need insights into the way CCS may evolve and what its role in reducing greenhouse gasses could be in conjunction with alternative options. Such insights are of great value in the energy transition process that we described in Section 4.1.

## 6 OBJECTIVE AND SCOPE OF THIS THESIS

The objective of this thesis is to assess the technical possibilities, costs and risks of CCS systems to allow comparison, identify promising options and determine how and to what extent CCS could be deployed over time in the Netherlands.

We look upon issues from a system perspective, which means that we unify insights provided by, amongst others, chemical and mechanical engineering (capture and transport) and earth sciences (storage). Insights in capture, transport and storage are combined to investigate the implications of CCS deployment in the Netherlands, representing a

densely populated industrialised country with a large ccs potential. The analyses are restricted to geological storage of  $\text{CO}_2$ , primarily because this is the most obvious storage medium in the Netherlands. Ocean storage is not considered as it is still in research phase and the Netherlands has no access to deep waters. Storage by mineralisation and reuse of  $\text{CO}_2$  in industrial processes or horticulture are not considered either. The topics that we study are predominantly technical by nature; regulatory issues and social acceptance of ccs are outside the scope of this thesis.

In order to meet the objectives, we formulate four research questions, each of which is linked up with the four gaps in knowledge discussed in the previous section:

1. What are the technical possibilities for ccs and what is their energetic and economic performance in a complete chain from source to sink?
2. To what extent does the spatial distribution of  $\text{CO}_2$  sources and sinks match, and what are the prospects of early opportunities that combine pure  $\text{CO}_2$  sources with nearby low-cost storage reservoirs?
3. What are the main factors that may affect the storage capacity? In particular, what are the risks of geological  $\text{CO}_2$  storage for man and the environment and what is the current lack of knowledge on these risks?
4. What is the potential role that ccs may play in decarbonising the Dutch energy system over time and how is this translated in terms of  $\text{CO}_2$  avoided, costs and infrastructure requirements?

The research questions are addressed in various chapters across this thesis (see Table 5). Basically, the results of techno-economic analyses of various ccs options, providing the answers to the first research question, are presented in all chapters except 6. While the analyses presented in Chapter 2 and 3 are rather generic by nature, Chapter 4, 5 and 7 zoom in to specific technologies and case studies. The second research question is split up into two parts. The first part looks at global low-costs source-sink combinations, of which the results are discussed in Chapter 5. In the second part, which is presented in Chapter 7, we perform a more detailed source-sink matching analysis for the Netherlands. Research questions 3 and 4 are discussed in Chapter 6 and 7.

TABLE 5. Overview of research questions addressed per chapter

CHAPTER	2	3	4	5	6	7
QUESTION 1	X	X	X	X		X
QUESTION 2				X		X
QUESTION 3					X	X
QUESTION 4						X

## 7 OUTLINE OF THIS THESIS

*Chapter 2* encompasses an extensive literature review of the thermodynamic performance and costs of technologies to produce electricity and hydrogen from coal and natural gas with CO<sub>2</sub> capture. A large effort was put in standardising the data provided in numerous scientific studies in order to calculate conversion efficiencies, energy production costs and CO<sub>2</sub> mitigation costs, the parameters which served as basis for comparison.

In *Chapter 3*, the most promising technologies identified in *Chapter 2* are further assessed by means of a chain analysis, incorporating fuel extraction and transport, CO<sub>2</sub> transport and storage and energy carrier transmission, distribution and end-use.

While performing the research presented in *Chapter 2*, we found that little was known on the techno-economic performance of small-scale membrane reactors, as the majority of research is directed to large-scale CCS systems. These advanced decentralised hydrogen units promise low-cost CO<sub>2</sub> capture. Therefore, we performed a more detailed assessment of this option using Aspen<sup>plus</sup> and cost-engineering analyses, the results of which are presented in *Chapter 4*.

In *Chapter 5*, the worldwide potential for low-cost CCS opportunities has been studied. This so-called low-hanging fruit consist of pure industrial CO<sub>2</sub> sources closely located to depleted oil reservoirs and coal seams, from which additional revenues could be generated by means of EOR or ECBM. A Multi-Criteria Analysis was applied to rank the source-sink combinations, after which some of the most promising cases were analysed in more detail.

In *Chapter 6*, the focus of our research is moved to the risks of underground CO<sub>2</sub> storage. We first review the possible risks that may occur when transporting and storing CO<sub>2</sub> in geological formations, describing both the mechanisms and effects. Then, an overview is given of current (gaps in) knowledge and research areas that need to be addressed to increase our understanding in those risks.

The overall synthesis of knowledge gathered throughout this thesis is presented in *Chapter 7*. Capture and storage potentials are combined to sketch four possible pathways how CCS (r)evolution may occur in the Netherlands up to 2050. For each pathway, the implications in terms of CO<sub>2</sub> stored and avoided, costs and infrastructural requirements are assessed.

Finally, in *Chapter 8*, the results are summarised and main conclusions are drawn.

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## **CHAPTER II A COMPARISON OF ELECTRICITY AND HYDROGEN PRODUCTION SYSTEMS WITH CO<sub>2</sub> CAPTURE AND STORAGE**

### **PART A: REVIEW AND SELECTION OF PROMISING CONVERSION AND CAPTURE TECHNOLOGIES \***

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#### **ABSTRACT**

We performed a consistent comparison of state-of-the-art and advanced electricity and hydrogen production technologies with CO<sub>2</sub> capture using coal and natural gas, inspired by the large number of studies, of which the results can in fact not be compared due to specific assumptions made. After literature review, a standardisation and selection exercise has been performed to get figures on conversion efficiency, energy production costs and CO<sub>2</sub> avoidance costs of different technologies, the main parameters for comparison. In the short term, electricity can be produced with 85-90% CO<sub>2</sub> capture by means of NGCC and PC with chemical absorption and IGCC with physical absorption at 4.7-6.9 €/ct/kwh, assuming a coal and natural gas price of 1.7 and 4.7 €/GJ. CO<sub>2</sub> avoidance costs are between 15 and 50 €/t CO<sub>2</sub> for IGCC and NGCC, respectively. In the longer term, both improvements in existing conversion and capture technologies are foreseen as well as new power cycles integrating advanced turbines, fuel cells and novel (high-temperature) separation technologies. Electricity production costs might be reduced to 4.5-5.3 €/ct/kwh with advanced technologies. However, no clear ranking can be made due to large uncertainties pertaining to investment and O&M costs. Hydrogen production is more attractive for low-cost CO<sub>2</sub> capture than electricity production. Costs of large-scale hydrogen production by means of steam methane reforming and coal gasification with CO<sub>2</sub> capture from the shifted syngas are estimated at 9.5 and 7 €/GJ, respectively. Advanced autothermal reforming and coal gasification deploying ion transport membranes might further reduce production costs to 8.1 and 6.4 €/GJ. Membrane reformers enable small-scale hydrogen production at nearly 17 €/GJ with relatively low-cost CO<sub>2</sub> capture.

## I INTRODUCTION

In an energy future driven by greenhouse gas constraints and fossil fuels retaining their dominant role in the world energy supply in the coming decades, CO<sub>2</sub> capture and storage (CCS) enables the production of electricity and hydrogen with strongly reduced CO<sub>2</sub> emissions [1-4]. CCS can be defined as the separation and capture of CO<sub>2</sub><sup>1</sup> produced at stationary sources, followed by transport and storage in geological reservoirs or the ocean in order to prevent its emission to the atmosphere. CCS generally also encompasses fixation of CO<sub>2</sub> in the form of minerals or utilisation for industrial purposes. Many studies have been performed with the purpose to identify promising CCS technologies for fossil fuel conversion into electricity and (to a lesser extent) hydrogen. The level of detail and data quality of these studies varies considerably. Due to the large variety in technologies covered, methodologies used, scales considered and assumptions made on technical performance and economic factors, the results of these studies can in fact not be compared. Few studies compare series of conversion technologies with CO<sub>2</sub> capture with respect to their energetic and economic performance on a common basis, also accounting for potential developments in state-of-the-art conversion and capture technologies. David and Herzog [5], Simbeck [6], Göttlicher [7,8] and Rubin et al. [9] compared various coal and natural gas-fired power plants with CO<sub>2</sub> capture. Parsons et al. [10] focused on advanced power systems. In the recently accomplished CCP project, various advanced CCS technologies (mainly natural gas-fired) have been analysed in detail, including potential improvements in current capture technology [11]. Hydrogen production from various fossil fuel routes with CO<sub>2</sub> capture have been compared in [12-14]. However, no complete review of state-of-the-art and advanced technologies for both electricity and hydrogen production with CO<sub>2</sub> capture is available that allow for a comparison of these options on a common basis.

The main objective of this study is to make a consistent techno-economic comparison of electricity and hydrogen production technologies with CCS using coal and natural gas. Therefore, an analysis based on literature review has been performed to collect data on energetic and economic performance of new-built plants and identify promising options. We consider state-of-the-art technologies that in principle could be operated on commercial scale (not considering legal and financial barriers) in the short term (around 2010) and advanced technologies that may become mature for market introduction in the longer term (around 2030).

Options with promising prospects in terms of energy efficiency and costs are further assessed in a chain analysis, incorporating CO<sub>2</sub> transport and storage and distribution/use of energy carriers to the end-user. A chain analysis is the most appropriate methodology to compare energy production costs and CO<sub>2</sub> avoidance costs in a consistent matter, because energy and CO<sub>2</sub> distribution costs can differ strongly among

1. Thermal cracking of natural gas into carbon and hydrogen followed by storage or useful application of carbon can also be considered as CCS.

technologies and can be decisive for the performance of a technology. The analysis is performed for the Netherlands and North Sea area, where CCS may play a significant role in reducing GHG emissions, as this region has large geological storage potential and large CO<sub>2</sub> point sources.

Our analysis is presented in two chapters. This chapter (part A) summarises the review and data normalisation of electricity and hydrogen production technologies and possible combinations with CO<sub>2</sub> capture. On that basis, we discuss promising options and highlight gaps and weak spots in information, which can be useful to set R&D priorities. Chapter 3 (part B) continues with the chain analysis of promising options selected in part A.

## 2 METHODOLOGY

### 2.1 DATA STANDARDISATION AND SELECTION

An extensive literature review has been performed to assess conversion efficiency figures and costs of electricity and hydrogen production technologies with and without CO<sub>2</sub> capture. Since each study contains specific assumptions affecting conversion efficiency and capital/energy production costs, data need to be standardised in order to make a fair comparison. We performed the following procedure:

1. Normalising CO<sub>2</sub> and H<sub>2</sub> pressure. Conversion efficiency and capital cost figures are corrected for standard CO<sub>2</sub> and H<sub>2</sub> pressure of 110 and 60 bar, respectively. This is done using regression fits of compression energy versus pressure difference derived from Aspenplus flowsheet modelling software. Capital costs of compression as function of capacity are derived from [15].
2. Indexation. Costs are converted to €<sub>2003</sub> using GDP deflators [16] and annual currency exchange rates ([www.oanda.com](http://www.oanda.com))<sup>2</sup>.
3. Normalisation of capital cost figures. Total capital requirement (TCR) or turnkey project costs consists of various components [17]:
  - Total plant costs (TPC) or overnight construction costs represent the cost to erect the plant (process plant costs or PPC) plus engineering and contingencies.
  - Owners costs are the costs to develop and start up the plant (royalties, preproduction costs, inventory capital, cost of land and site preparation).
  - Interest during construction (IDC)

Not every study includes the same components. If specified, generally TPC is reported and some studies also include owners costs and IDC. For those studies that did not include contingencies and/or owners costs, we used average figures based on data for equal technologies from other studies (contingencies: 10% of PPC, owners costs: 7% of PPC). IDC was calculated considering a construction period of 4 years for coal-fired power plants and 2

2. Most data in literature are presented in US\$. We use the average exchange rate of 2003: 0.885 €/US\$.

years for gas-fired power plants and equal annual expenses. For hydrogen production, we considered a 3-year construction period for large-scale natural gas-fired plants and 4 years for gasification plants, with equal annual expenses.

4. Scaling of capital cost figures. Since capital costs depend strongly on the capacities considered, which may vary substantially among studies, we standardised TPC applying a generic scaling relation (Equation 1). A base scale of 600 MWe<sup>3</sup> is considered for central power production (with and without capture) and 20 MWe for decentralised power production with natural gas-fired fuel cells. For hydrogen production, central facilities of 1000 MWH<sub>2</sub> (LHV basis) and facilities at refuelling stations of 2 MWH<sub>2</sub> are considered.

3. This capacity represents a typical Dutch coal-fired power plant and a large gas-fired power plant.

$$\frac{Costs_A}{Costs_B} = \left( \frac{Capacity_A}{Capacity_B} \right)^R \quad \text{EQUATION 1}$$

in which R = scaling factor for different conversion technologies

After standardisation, the literature sources are selected on a number of criteria in order to get figures on conversion efficiency and costs. Obviously, preference is given to recent, high-detail and transparent studies, ideally including data on conversion and capture efficiency, investment and O&M cost. For state-of-the-art technologies often a multitude of high-quality references is available, which makes it rather difficult to make a selection on these criteria. Other criteria are then applied to get a value or range in conversion efficiency and investment costs, among which technological maturity (technologies need to be commercially available within considered timeframe) or plant specifications. Less mature, advanced concepts are generally studied in less detail. In these cases, we often have to rely on efficiency forecasts and even less certain cost projections. Investment costs for advanced technologies are generally forecasted values based on technology improvements and do not explicitly account for the effect of technological learning.

## 2.2 KEY PARAMETERS FOR COMPARISON

The different technologies with CO<sub>2</sub> capture are compared on net efficiency (LHV basis), energy production costs and CO<sub>2</sub> avoidance costs. Hydrogen production systems often comprise various energy inputs and outputs: feed, fuel, hydrogen, steam and electricity. The conversion efficiency of these systems is defined as quotient of the sum of energy outputs by the sum of energy inputs, all in primary terms:

$$\eta = \frac{E_{hydrogen} + \frac{E_{el,out}}{\eta_e} + \frac{E_{th,out}}{\eta_{th}}}{E_{feed} + E_{fuel} + \frac{E_{el,in}}{\eta_e} + \frac{E_{th,in}}{\eta_{th}}} \quad \text{EQUATION 2}$$

in which  $\eta_e$  = electric efficiency (0.5) and  $\eta_{th}$  = thermal efficiency (0.9)

Levelised energy production costs (COE and COH for electricity and hydrogen, respectively) are calculated by dividing the sum of annual capital, o&m and fuel costs by the annual energy production. For hydrogen production, also electricity costs or revenues are accounted for. Annual capital costs are derived from the total capital requirement by means of a capital recovery factor, which is a function of the discount rate and economic lifetime. o&m costs generally include costs related to maintenance, (operating) labour, supervision and administration, chemicals and consumables and waste disposal. Some studies include insurance, local taxes and by-product credits (sulphur), but these are not included here. We express o&m costs as a share of TPC. CO<sub>2</sub> avoidance costs differs from CO<sub>2</sub> capture costs as it accounts for the fact that the plant with CO<sub>2</sub> capture is less efficient (and hence produces additional CO<sub>2</sub>):

$$\text{CO}_2 \text{ avoidance costs} = \frac{COE_{cap} - COE_{ref}}{m_{CO_2,ref} - m_{CO_2,cap}} \quad \text{EQUATION 3}$$

in which  $m$  = CO<sub>2</sub> emission factor (g/kwh or g/GJ hydrogen) of capture plant (cap) and reference plant (ref). These emission factors include indirect (avoided) CO<sub>2</sub> emissions by electricity use or production.

The choice of the reference system has a significant impact on CO<sub>2</sub> avoidance costs. In Chapter 3, the impact of various reference systems is considered more extensively. In this chapter, we apply the most common approach comparing identical plants with and without CO<sub>2</sub> capture (i.e. the baseline varies per technology). This approach gives a clear insight which technologies enable inherent low-cost CO<sub>2</sub> capture.

### 3 ELECTRICITY PRODUCTION TECHNOLOGIES

There are many combinations of energy conversion technologies and CO<sub>2</sub> capture technologies conceivable (in some options CO<sub>2</sub> capture is inherent to conversion technology), which urges the need for a transparent classification. The most common classification of CO<sub>2</sub> capture strategies distinguishes post-combustion capture, pre-combustion capture and oxyfuel combustion (see Figure 1). The principle of post-combustion capture is to remove CO<sub>2</sub> from flue gas after combustion at low pressure and low CO<sub>2</sub>-content. In pre-combustion schemes, CO<sub>2</sub> is captured from shifted syngas (consisting predominantly of H<sub>2</sub> and CO<sub>2</sub> at high pressure) produced by reforming (SMR) or partial oxidation (POX) of natural gas or gasification of coal. Hydrogen rich gas is subsequently combusted in adapted turbines. In oxyfuel processes, fuel is combusted in an atmosphere of oxygen with recycled CO<sub>2</sub> and/or steam, resulting in a concentrated stream of CO<sub>2</sub> and steam, which can be separated easily by condensation. In this study, we discuss these capture options from the perspective of

power production technologies, being pulverised coal-fired power plants (PC), natural gas combined cycle (NGCC), integrated coal gasification combined cycle (IGCC) and (high-temperature) fuel cells.

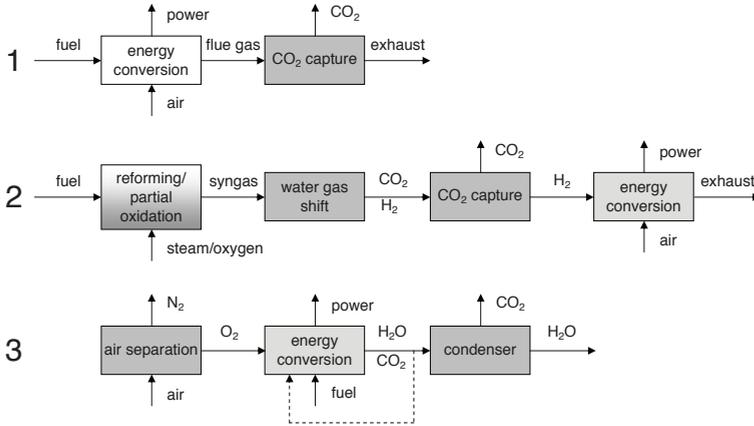


FIGURE 1. Principle of post-combustion capture (1), pre-combustion capture (2) and oxyfuel combustion (3). White components are equal to the configuration without capture, light grey indicates modifications to the configuration without capture, dark grey indicate new components

3.1 PULVERISED COAL-FIRED POWER PLANTS

In PC plants, pulverised coal is combusted with excess air in a boiler to produce high-pressure/temperature steam, which is expanded in a steam turbine to generate electricity. The efficiency strongly depends on steam pressure and temperature, which showed an increasing trend over the last decades. Supercritical plants<sup>4</sup> are state-of-the-art technology, reaching net electrical efficiencies in the range of 41 to 46% [18-20]. Developments in materials make use of higher steam pressures and temperatures to ultra super critical conditions possible. Efficiency forecasts for 2010 and 2025 are 48-50% and 50-55%, respectively [21]. CO<sub>2</sub> can be captured by means of post-combustion capture and oxyfuel combustion.

4. Steam pressure higher than critical pressure of 220 bar and steam temperatures higher than 560°C.

3.1.1 POST-COMBUSTION CAPTURE

The leading technology in post-combustion capture is chemical absorption of CO<sub>2</sub> using monoethanolamine (MEA). This technology is commercially applied to produce high-purity CO<sub>2</sub> from the exhaust of coal and gas-fired boilers, furnaces and turbines. The main applications for high-purity CO<sub>2</sub> are enhanced oil recovery, urea production and in the food/beverage industry. At present, the largest operating unit has a capacity of 800 t CO<sub>2</sub>/day [22]. To put this into perspective, a 500 MW<sub>e</sub> PC unit produces circa 8000 t CO<sub>2</sub>/day. MEA is suited for low CO<sub>2</sub> partial pressures as occurring in flue gas. First, the flue gas is cooled and impurities are removed. NO<sub>x</sub> and SO<sub>2</sub> removal to low concentrations prior to CO<sub>2</sub> capture is essential, since these components form heat

stable salts. In the absorption column,  $\text{CO}_2$  reacts chemically with the amine. The  $\text{CO}_2$ -rich absorbent is pumped to the desorber, where  $\text{CO}_2$  is released by increasing temperature up to  $120^\circ\text{C}$ , for which steam is extracted from the steam cycle. The regenerated absorbent is recycled to the absorber and  $\text{CO}_2$  is dried and compressed for transport conditions (typically between 100 and 150 bar).

The major bottleneck is the large steam requirements for  $\text{CO}_2$  regeneration (circa  $4.2 \text{ MJ/kg CO}_2$  for conventional Econamine FG process [23]) and high investment costs of the capture unit. However, existing processes are improved and new processes are researched/developed. The Econamine FG+ process, which includes a split-flow configuration, an improved solvent formulation and better heat integration, achieves significantly lower energy use [21]. A new solvent based on sterically hindered amines (KS-1) has been commercialised and adopted in an ammonia plant to capture  $\text{CO}_2$  from furnace exhaust. Recovery energy of KS-1 has been reduced to  $3.2 \text{ MJ/kg CO}_2$  [24]. Further improvements are foreseen in the Econamine and KS-1 processes (stripping steam requirements and amine loading). A recent study indicates that precipitating amino-acid salt solutions with higher  $\text{CO}_2$  loading and lower heat of desorption might reduce energy requirements to  $2.3 \text{ MJ/kg CO}_2$ , a reduction of 45% in comparison to MEA technology [25]. Investment costs of absorption units can be further reduced by further optimisation of absorber design, e.g. omitting the cooler by integrating this element in the flue gas desulphurisation unit [21]. The application of membrane contactors, serving as contact between flue gas and absorption solvent, enables the application of smaller absorption columns [26].

Alternative options to capture  $\text{CO}_2$  from flue gasses are adsorption, low temperature distillation and membranes. Energy requirements of conventional pressure (and temperature) swing physical adsorption, in which  $\text{CO}_2$  binds to a solid surface (e.g. zeolites), are prohibitively large [27]. A relatively new concept is the use of high-temperature solid compounds, which react with  $\text{CO}_2$  to form a carbonate. In a different reactor, the sorbent is regenerated, producing pure  $\text{CO}_2$ . The impact on the conversion efficiency and costs is not quantified at the time this chapter was written. Low temperature distillation uses a freezing technique to capitalise on the difference between the freezing point of  $\text{CO}_2$  and the rest of the flue gas. Göttlicher [8] indicated that  $\text{CO}_2$  avoidance costs of distillation are higher than for chemical absorption. The driving force for  $\text{CO}_2$  separation using membranes depends on the partial pressure difference of  $\text{CO}_2$  between permeate and retentate side of the membrane. Using commercially available polymeric membranes results in relatively large energy requirements and  $\text{CO}_2$  avoidance costs in comparison to chemical absorption, due to the low driving force as a consequence of the low  $\text{CO}_2$  partial pressure in flue gas [7,28,29].

### 3.1.2 OXYFUEL COMBUSTION

In a power plant with oxyfuel combustion, pulverised coal is combusted using (nearly) pure oxygen, which is produced by a cryogenic air separation unit (ASU), although new technologies such as ion transport membranes (ITM) are being worked on. The flue gas, containing primarily CO<sub>2</sub> (the rest being H<sub>2</sub>O, Ar, O<sub>2</sub>, N<sub>2</sub>, NO<sub>x</sub> and SO<sub>2</sub>) is partially recycled to the boiler to control the combustion temperature. The remaining flue gas is cooled to remove water, compressed, followed by separation of non-condensable gasses (Ar, O<sub>2</sub> and N<sub>2</sub>) from CO<sub>2</sub>, and is finally boosted to pipeline pressure. The advantage of oxyfuel combustion is that it enables nearly 100% CO<sub>2</sub> capture. In addition, the selective catalytic reduction (SCR) unit and the flue gas desulphurisation (FGD) unit might be omitted as oxyfuel combustion results in low NO<sub>x</sub> emissions and the remaining NO<sub>x</sub> and SO<sub>2</sub> present in the flue gas could in principle be left for co-storage with CO<sub>2</sub> or could be separated easily<sup>5</sup> [31,32].

Although there is experience with oxygen fuel combustion in the glass melting and the steel and aluminium industry, the concept of oxyfuel combustion has not been applied in power plants yet. So far only some demonstration tests have been/are conducted. These tests aim to study the effects on plant operation (combustion behaviour, heat transfer<sup>6</sup>) and plant maintenance (fouling, slagging, corrosion). After these issues are resolved, full-scale burners need to be demonstrated successfully. Therefore this option is not considered to be viable for implementation in the short term.

### 3.1.3 KEY PERFORMANCE DATA

Table I shows the range in efficiency and investment costs of all PC options with CO<sub>2</sub> capture. We use the efficiency and investment costs of a typical, modern supercritical coal plant for North western European conditions, integrated with the latest generation post-combustion capture process (Econamine FG+) [21]. The capture penalty of this process is optimistic in comparison to the majority of studies, in which a conventional MEA is applied. Industry projections for a 2020 plant, including improvements in conversion and capture (amine) technology, results in investment cost reductions of 20% and 39%, respectively [21]. Annual O&M costs for PC with capture by means of MEA are between 4 and 6% of TPC [18,20,21]. The table seems to indicate that the efficiency (penalty) and capital costs of PC with oxyfuel combustion are comparable to PC with post-combustion capture. Few studies were accomplished which considered both capture strategies, and these studies show considerable discrepancy in the results. Both Göttlicher [7] and Parsons et al. [10] conclude that the efficiency penalty and additional costs of oxyfuel combustion schemes are lower in comparison to post-combustion capture schemes. Simbeck analysis indicate that PC with oxyfuel combustion causes a higher efficiency penalty, but lower additional costs in comparison to post-combustion capture [6]. IEA GHG studies indicate that the efficiency penalty of both capture options is

5. NO<sub>x</sub> and SO<sub>2</sub> "Co-transport and storage" with CO<sub>2</sub> might have serious implications for transport pipelines and storage reservoirs (e.g. enhanced oil recovery) [30]. If required, NO<sub>x</sub> and SO<sub>2</sub> can be separated easily from CO<sub>2</sub> by means of an additional distillation unit in the CO<sub>2</sub> purification system [31].

6. A recent study indicated that the heat transfer within the furnace is improved as the emissivity of the flue gas in oxyfuel combustion systems is higher due to the higher CO<sub>2</sub> and H<sub>2</sub>O content, which results in a higher boiler efficiency [31].

comparable and the additional capital costs are higher for the oxyfuel combustion plant [21,31]. Hence it is unclear whether oxyfuel combustion proves to be a more cost-effective capture option than post-combustion strategies.

TABLE I. Key parameters of PC plants (using bituminous coal) with CO<sub>2</sub> capture from literature review

Conversion technology	CO <sub>2</sub> capture technology	Net electric efficiency <sup>a</sup>	Efficiency penalty	CO <sub>2</sub> capture efficiency	TCR (€/kW <sub>e</sub> ) <sup>b</sup>	Sources
(ultra) super-critical PC <sup>c</sup>	Post-combustion (amines)	30-35%	8- 13% points	85-90%	1720-2490	[6,7,9, 18,20,21]
(ultra) super-critical PC 2020	Improved post-combustion (amines)	40%	9% points	85%	1520	[21]
(ultra) super-critical PC <sup>d</sup>	Oxyfuel combustion (ASU)	33-36%	9-12% points	90-100%	1830-2220	[6,7,31]

<sup>a</sup> Including CO<sub>2</sub> compression to 110 bar.

<sup>b</sup> Investment costs have been scaled to 600 mwe assuming a scaling factor of 0.75, derived from PC investment costs of various scales based on engineering economic models to evaluate fossil fuel power plants [9].

<sup>c</sup> Many studies consider CO<sub>2</sub> capture at subcritical plants, which are not considered here as state-of-the-art PC plants in North western Europe are generally supercritical.

<sup>d</sup> Several other studies have been performed on oxyfuel combustion applied at lignite-fired power plants [33,34]. Lignite generally has a higher moisture content and lower heating value than bituminous coal, which has a significant impact on efficiency and capital costs of the power plant [34]. As our focus is on the Netherlands, where bituminous coal is used for power generation, CO<sub>2</sub> capture at lignite-fired power plants is not further considered.

### 3.2 NATURAL GAS COMBINED CYCLE

In NGCC plants, natural gas is combusted and the hot flue gas is expanded in the turbine, driving the air compressor and a generator. The heat of the gas turbine exhaust is used to raise steam in a waste heat boiler, which is expanded in a steam turbine. The efficiency depends principally on the turbine inlet temperature (TIT). The most advanced H-class turbines reach a TIT exceeding 1400°C employing closed loop steam cooling of the gas turbine first two expansion stages and steam reheating [35]. These turbines, reaching net electrical efficiencies of 60%, are currently being demonstrated, but are not expected to operate in fully commercial plants until 2008 [21]. Gas turbine technology is likely to progress further in the coming decades. Systems with higher temperature exhaust temperature enable the application of supercritical, once-through heat recovery steam generator (HRSG) units, which will increase the efficiency and reduce investment costs [21]. Other techniques (being) commercialised to increase efficiency are sequential combustion (reheat), recuperation and intercooling to reduce power demand of air compression [36]. These techniques are (partly) applied in (cascaded) humid air turbines ((C)HAT), in which compressed air is humidified in an air saturator and heated using turbine exhaust

prior to combustion. In these concepts, the HRSG and steam cycle can be omitted. A recent study shows that the efficiency of a large scale HAT cycle was slightly lower than a standard G-class turbine and investment costs are significantly higher, indicating that this cycle is not advantageous to NGCC (when not integrated in other cycles) [10]. CO<sub>2</sub> capture in a NGCC can be performed by means of post-combustion capture, pre-combustion capture and oxyfuel combustion.

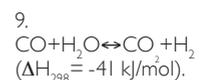
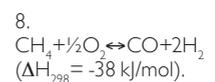
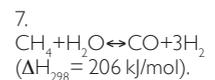
### 3.2.1 POST-COMBUSTION CAPTURE

Due to the low partial pressure of CO<sub>2</sub> in the flue gasses of a NGCC (about 0.04 bar), chemical absorption using amines (MEA) is the preferred capture technology. Again, technological options can be implemented to reduce the energy penalty caused by conventional chemical absorption, which have been studied extensively for NGCC in two recent studies [21,37]. These options can be classified into process simplification, process integration and solvent improvement. Cost reductions can be achieved by omitting the flue-gas cooler, using different (cheaper) heat exchangers and pumps and structured column packing. Process integration includes recycling part of the flue gas to the air compressor to enhance the CO<sub>2</sub> concentration and inserting the amine reboiler tube bundles directly in the HRSG [37]. Integrated designs require further developments in equipment. Flue gas recycling can be performed in existing turbines, but the combustor and fuel and control systems need some modifications [38]. The most optimistic system considered in the CCP study, in which KS-1 solvents are combined with innovative design engineering to achieve high integration between power cycle and capture unit, could be commercially introduced for large-scale application before the end of this decade if aggressively pursued [39].

Apart from absorption, also alternative technologies have been studied [37]. Adsorption processes are relatively expensive due to the low CO<sub>2</sub> loading and cryogenic CO<sub>2</sub> separation is also considered less attractive than absorption. Capital costs savings of a membrane contactor combined with KS-1 solvent integrated versus a conventional absorption and desorption unit are within the uncertainty of the estimates.

### 3.2.2 PRE-COMBUSTION CAPTURE

Pre-combustion capture systems encompass syngas production by reforming<sup>7</sup> and/or partial oxidation<sup>8</sup> of natural gas (see Section 4 for more details), subsequent water gas shift (WGS)<sup>9</sup>, separation of CO<sub>2</sub> and hydrogen by physical or chemical absorption and combustion of hydrogen in a gas turbine. The main problem with combusting hydrogen in current turbines is that it will result in increased NO<sub>x</sub> emissions due to an increased flame temperature. The current technology in gas turbines to reduce NO<sub>x</sub> emissions using premixed combustors cannot be deployed for hydrogen combustion due to its combustion characteristics [38,40]. SCR is an alternative, but costs are considered too high for reducing NO<sub>x</sub> emissions from levels that arise



when combusting pure hydrogen [40]. This makes fuel dilution with steam (from the steam cycle) or nitrogen the most feasible option. In systems where hydrogen is produced by POX using oxygen, nitrogen from the ASU can be injected in the combustor to reduce flame temperature. If nitrogen is present in the hydrogen-rich fuel (when using air as oxidizing agent), changes in the combustor may be smaller than for burning pure hydrogen [38]. A thermodynamic analysis has indicated that the efficiency of combined cycles is only moderately decreased when diluting the fuel with nitrogen or steam [40]. Various levels of process integration are possible in order to decrease energy losses and/or costs:

1. CO shift and CO<sub>2</sub> capture can be performed simultaneously in the so-called sorption enhanced water gas shift (SEWGS) process, in which a WGS catalyst and a special high temperature CO<sub>2</sub> adsorbent are combined in a reactor [41]. As CO<sub>2</sub> is removed from the gas phase, the WGS reaction is shifted to the product side. Since no cooling is required, the energy penalty for cooling/reheat is avoided and the steam is preserved in the hot hydrogen stream, contributing to power generation. This technology is expected to be commercially ready after 2010 [42].
2. In the membrane reformer (MR-CC<sup>10</sup>), steam methane reforming is integrated with a hydrogen separation membrane. The membrane, placed inside the reforming tube, continuously withdraws hydrogen from the reaction zone (retentate side). As hydrogen permeates through the membrane, the chemical equilibrium of the reforming and WGS reaction is shifted to the product side according to Le Chatelier's principle, thereby increasing methane conversion. On the permeate side, nearly pure hydrogen is collected. By using steam/nitrogen as sweep gas, the partial pressure difference across the membrane increases, thereby increasing the hydrogen flux. Simultaneously, the nitrogen or steam in the gas will depress NO<sub>x</sub> formation in the combustion chamber. The retentate gas stream leaving the reactor consists mainly of CO<sub>2</sub> and water vapour at feed pressure. CO<sub>2</sub> can be captured easily by condensation of the water vapour.

Membrane material can be metal (composite) such as Pd or Pd alloy, microporous (SiO<sub>2</sub>, C, zeolite) and dense mixed conducting membranes<sup>11</sup> (perovskites) [43,44]. Membranes with sufficient selectivity, stability and lifetime still require serious development efforts. MR technology is expected to be commercially ready after 2015 [42].

### 3.2.3 OXYFUEL COMBUSTION

In NGCC systems with oxyfuel combustion, natural gas is combusted with (nearly) pure oxygen. Cycles with a conventional ASU to produce pure oxygen can be distinguished from novel oxyfuel power cycles such as advanced zero emission power plants (AZEP) and chemical looping combustion (CLC), employing membranes and oxygen transport particles to enable stoichiometric combustion with oxygen. Various

10. This concept integrates a membrane reformer with a combined cycle. We use the abbreviation MR-CC to distinguish from a membrane reformer to produce pure hydrogen, which is not (directly) converted in a combined cycle (see Section 4.1.3).

11. Material with high ionic and electronic conductivity.

oxyfuel cycles with ASU and CO<sub>2</sub>/H<sub>2</sub>O recycling have been proposed and evaluated, among which the Graz, Water and Matiant (combined) cycle. The Graz cycle consists of a high-pressure combustor with steam injection and a recuperated gas turbine integrated with a steam cycle. Both CO<sub>2</sub> and steam are recycled to the combustion chamber [45]. The Water cycle is an oxygen-fired cycle with steam recycle, producing a high-pressure, superheated mixture of mainly H<sub>2</sub>O and CO<sub>2</sub> in a gas generator, which is then expanded in a series of (advanced) turbines. A 20 MW<sub>th</sub> gas generator fuelled with natural gas has been tested and a 500 kW<sub>e</sub> power plant is being developed by Clean Energy Systems (CES) [46,47]. In the Matiant or oxyfuel combined cycle, natural gas is combusted in an O<sub>2</sub>/CO<sub>2</sub> atmosphere. The reaction product, consisting principally of CO<sub>2</sub> and H<sub>2</sub>O, is expanded in an adapted gas turbine and the heat of the turbine exhaust is used to generate steam, which is expanded in a steam cycle [48].

Oxyfuel combustion in Brayton cycles requires gas turbines that use CO<sub>2</sub>/H<sub>2</sub>O as working fluid, which has different expansion characteristics than nitrogen used in conventional turbines. The optimal pressure ratio for CO<sub>2</sub> turbines is higher than for conventional turbines, which implies that the efficiency of a CO<sub>2</sub> turbine for typical pressure ratios nowadays is lower than the efficiency of conventional turbines [8,38,48]. Existing gas turbines cannot simply be adapted for CO<sub>2</sub> as working fluid; hence complete new gas turbines (combustor and compressor) need to be developed. Such a development is not likely to occur as long as gas turbine manufacturers do not foresee a significant market to arise [38].

#### *Advanced zero emission power plant*

The key of the AZEP concept is substitution of the conventional combustion chamber in a gas turbine by a mixed conducting membrane (MCM) reactor, which combines oxygen production, fuel combustion and heat transfer. Compressed air enters the MCM reactor, where oxygen passes through the membrane and is transported to the combustion chamber. The heat of combustion is transferred to oxygen-depleted air, which is expanded in a conventional turbine. Steam is generated from the turbine exhaust and the CO<sub>2</sub>/H<sub>2</sub>O stream generated in the combustion chamber, which is then expanded in a steam turbine. The use of a conventional turbine is essentially the advantage of AZEP over oxyfuel concepts using gas turbine with CO<sub>2</sub>/H<sub>2</sub>O as working fluid. The AZEP concept combines well with current technology, requiring minor adaptations in gas turbine (working fluid has lower oxygen content) and HRSG. Obviously, there are still technical challenges in the MCM development.

The AZEP concept is now being developed by a consortium of energy companies. The gas turbine selected for test phase is a 64 MW<sub>e</sub> GTX100, with 53% efficiency in CC mode. Also a 400 MW<sub>e</sub> v94.3A turbine with an efficiency of 57.9% has been studied, which is further considered in this study. Due to the limited temperature in the MCM reactor (circa 1200°C), TIT is lower than TIT of most advanced gas turbines, which exceed

1400°C. By installing an afterburner to increase TIT and accepting a lower CO<sub>2</sub> capture efficiency of 85%, the efficiency loss can be reduced [49].

### *Chemical looping combustion*

CLC is an oxyfuel concept, which is still in an early phase of development. The reactor technology is currently being tested in a 10 kW prototype [50]. CLC is based on fuel combustion by means of two separate reactors in order to separate nitrogen from the combustion products. In the reduction reactor<sup>12</sup>, fuel is oxidised by an oxygen carrier, generally a metal oxide such as iron/nickel oxide. The reduced metal oxide is then returned (in particulate form, using a fluidized bed concept) to the oxidation reactor, where it reacts with oxygen to close the cycle. The oxidation of the metal is highly exothermic and provides high temperature exhaust air for power generation. The metal oxide supplies heat to the endothermic reduction reaction. Both natural gas and syngas can be used as fuel, and the technology can be integrated in various power cycles. In a concept studied by Brandvoll and Bolland, a humid air turbine is used to expand (saturated) air from the oxidation reactor [51]. The metal oxide also supplies heat to the exhaust stream of the reduction reactor (CO<sub>2</sub> and H<sub>2</sub>O) in a gas-solid heat exchanger. The exhaust is expanded in a CO<sub>2</sub>/H<sub>2</sub>O turbine. Yu et al. considered the so-called sorbent energy transfer system (SETS), a CLC concept designed to interface with an existing NGCC with minimal modifications [52]. A comparative study, in which concepts with and without CO<sub>2</sub> expansion turbine have been analysed, indicates that the efficiency is similar [53]. A crucial issue for CLC systems is the maximum reactor temperature at which sorbent stability is not endangered. This temperature in turn determines TIT and thus efficiency. Brandvoll and Bolland assume TIT of 1200°C in their thermodynamic analysis [51]. Yu et al. [52] set base case reactor temperatures at 900°C, which requires supplemental gas burning to achieve the specified TIT of a F-class turbine, reducing CO<sub>2</sub> capture efficiency to 50%. Wolf assumes 1200°C to be the maximum reactor temperature, although it is uncertain whether the oxygen carrier particles can withstand such high temperatures [53].

12. Reduction/oxidation refers to the state of the oxygen carrier.

### 3.2.4 KEY PERFORMANCE DATA

Table 2 shows the range in efficiency and investment costs of all natural gas-fired electricity options with CO<sub>2</sub> capture. Many extensive studies have been performed on NGCC with post-combustion capture. The observed range in costs and efficiency data is primarily caused by turbine type and capture process considered. Annual O&M costs for NGCC with post-combustion capture lie between 3 and 6% of TPC [10,18,20,21,42].

For the short term, we use data on performance and costs from a recent, detailed study considering two GE 9FA frame turbines integrated with chemical absorption using Econamine FG+ solvent [21]. In the longer term, (advanced) post-combustion, pre-combustion and oxyfuel

combustion technology are candidates for high-efficient, low-cost electricity production with strongly reduced CO<sub>2</sub> emissions. For the post-combustion options, we consider an advanced turbine ( $\eta_e \sim 62\%$ ) integrated with further improved processes based on Fluor's Econamine FG+ [21]. Further cost reductions in that study in both NGCC and capture unit are assumed to be 20% and 32% of state-of-the-art figures, respectively. Pre-combustion capture systems comprising syngas production, WGS and CO<sub>2</sub> capture by absorption do not seem to offer efficiency and costs advantages in comparison to post-combustion schemes. However, advanced pre-combustion technologies such as MR-CC seem promising. We use efficiency and costs forecasts from a MR-CC with hydrogen mixed conducting ceramic membranes [42,43]. The oxyfuel concepts do not show a clear winning option at present due to the large range in conversion efficiency and lack of accurate costs estimations. The conversion efficiency of Matiant (oxyfuel) and Graz cycle with conventional ASU are clearly lower than AZEP and CLC. The Water cycle entails large technological uncertainty, which causes the wide range in Table 2. It may reach conversion efficiencies over 50%, provided that high temperature turbine machinery is developed. Results of thermodynamic analyses comparing various gas-fired options show that the Water cycle (with moderate assumptions on turbine development and oxygen production by ASU) does not offer higher conversion efficiency than advanced oxyfuel concepts such as AZEP and CLC, nor post and pre-combustion strategies<sup>13</sup> [10,54,55]. Parsons et al. [10] show that the Water cycle has higher investment costs than NGCC with chemical absorption, although current estimations quoted by CES are somewhat lower [56]. Since various studies indicate that the Water cycle does not seem to offer energetic nor economic advantages above NGCC with chemical absorption and efficiency and investment costs of advanced systems are highly uncertain, the Water cycle is not further considered. We consider AZEP and CLC as the most promising advanced oxyfuel technologies. For AZEP, we use conversion efficiencies calculated for the configuration with 100% CO<sub>2</sub> capture [49]. The capital costs are estimated from a cost quotation [57] and results of cost calculations [58,59]. For CLC, we assume a conversion efficiency of 51% based on [55] and use a rough cost estimation quoted in [60].

13. When comparing the Water cycle with AZEP, it would be fair to take into account the efficiency improvement in oxygen production by membranes in the Water cycle.

TABLE 2. Key parameters of natural gas-fired power plants with CO<sub>2</sub> capture from literature review

Conversion technology	CO <sub>2</sub> capture technology	Net electric efficiency <sup>a</sup>	Efficiency penalty <sup>b</sup>	CO <sub>2</sub> capture efficiency	TCR (€/kW <sub>e</sub> ) <sup>c</sup>	Sources
<i>Post-combustion capture</i>						
NGCC (F-class) <sup>d</sup>	Chemical absorption	43-50%	6-12% points	85-90%	700-1010	[9,18,20, 21,55,61]
NGCC (G/H-class) <sup>d</sup>	Chemical absorption	48-53%	5-11% points	85-90%	780-1770	[6,10,18, 21,42,48,62]
NGCC 2020	Improved chemical absorption	55%	6% points	85%	650	[21]
<i>Pre-combustion capture</i>						
SMR/POX/ATR-CC <sup>e</sup>	Chemical/physical absorption	43-50%	8-13% points	85-90%	900-1470	[6,10,20,48, 55,62-64]
ATR-CC	SEWGS	47-48%	9-10% points	90%	980-1060	[42]
MR-CC <sup>f</sup>	Hydrogen membrane	51-53%	5-6% points	100%	940	[42,55]
<i>Oxyfuel combustion</i>						
Graz cycle <sup>g</sup>	ASU	9-45%	13-19% points	100%	-	[54,55]
Water cycle <sup>h</sup>	ASU	40-62%	10-13% points	100%	820-880	[10,47,54, 55,65,66]
Oxyfuel (Matiant) CC	ASU	44-48%	11-13% points	90-100%	1060-1250	[6,31,48, 55]
AZEP <sup>i</sup>	Oxygen membrane	50-55%	2-8% points	85-100%	~700-900	[49,55, 57-59]
CLC-CC + CO <sub>2</sub> turbine <sup>j</sup>	Oxygen carrier	51-54%	3-5% points	100%	~900-1300	[51,53, 55,60]
CLC-CC <sup>k</sup>	Oxygen carrier	50-53%	3-5% points	50-100%	~500	[52,53,67]

<sup>a</sup> Including CO<sub>2</sub> compression to 110 bar.

<sup>b</sup> The efficiency penalty refers to the difference with a standard NGCC.

<sup>c</sup> As gas turbines are available in certain sizes, the scale for NGCC cycles is determined by the number of turbines installed and can, in fact, not simply be readjusted to the scale desired. However, capacities of NGCC cases studied are generally between 300 MW<sub>e</sub> (single turbine) and 800 MW<sub>e</sub> (2 turbines), which cause significant cost differences. Therefore, investment costs have been scaled to 600 MW<sub>e</sub> using a scaling factor of 0.9 derived from turnkey NGCC prices for different capacities given in [68,69].

<sup>d</sup> The range in efficiency forecasts is depending primarily on the capture process, ranging from conventional MEA (high efficiency penalty) to KS-1 process (low efficiency penalty). The highest level of integration is represented by the so-called BIT option (best integrated technology), combining process simplification and integration with use of KS-1 solvent. This results in an efficiency penalty of 5% points [42]. Note that the TCR estimate of 1770 €/kW<sub>e</sub> is atypical.

<sup>e</sup> These figures are representative for a set of pre-combustion configurations, integrating various syngas production technologies with WGS, CO<sub>2</sub> capture by means of absorption and H<sub>2</sub> combustion in various turbine classes.

<sup>f</sup> The highest efficiency is forecasted for a MR-CC with a dense hydrogen mixed conducting

ceramic membrane (HMCM) and nitrogen/steam as sweep gas (costs given in the table refer to this configuration) [43]. Simulations of a Pd based MR-CC with steam extracted from the steam turbine for sweep gas application results in a net electric efficiency of 51% [55].

- <sup>g</sup> The efficiency range in brackets is caused by the assumed condenser pressure, ranging from 0.1 to 0.9 bar [54].
- <sup>h</sup> Many Water cycle configurations have been studied. Generally, gas is first expanded in a high-pressure, high-temperature steam turbine. After expansion, the gas mixture is reheated and expanded in an intermediate and low-pressure turbine (which can be either a steam or gas turbine). The large range in efficiency is mainly caused by the assumed TIT of the turbines, which can be increased by using special alloys and steam cooling techniques applied in gas turbines. The lowest value represents a state-of-the-art Rankine power cycle with high-pressure TIT of 900°C and intermediate-pressure TIT of 1328°C [54]. The most optimistic configuration assumes a high-pressure TIT of 816°C and intermediate and low pressure TIT of 1649°C (double reheat) and expansion of high-pressure N<sub>2</sub> from the ASU [47]. Such turbines, based on turbine development goals of the US-DOE Vision 21 program, require considerable R&D efforts. The efficiency penalty and costs are representative for a configuration with single reheat and intermediate-pressure TIT of 1200-1400°C [10,55,66].
- <sup>i</sup> Represents efficiency projections of a 400 MW<sub>e</sub> system. Additional investment costs estimations range between 400 €/kW<sub>e</sub> in the short term to less than 250 €/kW<sub>e</sub> in the long term [57]. A more detailed techno-economic evaluation of 30-40 MW<sub>e</sub> simple cycle AZEP concepts shows that the specific investment costs for AZEP systems are 120-170% higher than for the reference GT [58]. The concepts with afterburner are in the low range, although CO<sub>2</sub> avoidance costs of systems with and without afterburners are very close. The costs related to CO<sub>2</sub> capture are lower for the original combined cycle configuration AZEP [58], also considering the difference in scale of the 30-40 MW<sub>e</sub> simple cycle systems and 400 MW<sub>e</sub> combined cycle system.
- <sup>j</sup> The high efficiency forecast represents a 50 MW<sub>th</sub> natural gas-fired cycle with HAT turbine and CO<sub>2</sub>/steam turbine to expand the exhaust of the reduction chamber. Crucial issues on oxygen carrier durability, mechanical properties and fuel conversion have been idealised [51]. A confidential thermo-economic analysis revealed that a similar CLC concept of 250 MW<sub>e</sub> might reach a net electric efficiency of 50% when using natural gas containing 15% nitrogen [70]. The capital costs are rough estimates derived from individual costs of reactors, compressors and turbines [60,70].
- <sup>k</sup> The net electric efficiency of the SETS cycle (=CLC-CC) ranges from 53% for a capture efficiency of 50% to 50% for 100% CO<sub>2</sub> capture [52]. Investment costs of the SETS system are estimated to be 10% higher than the total investment costs of an NGCC [67].

### 3.3 INTEGRATED COAL GASIFICATION COMBINED CYCLE

An IGCC comprises gasification of solid or liquid fuels (coal, oil residues, biomass) to produce raw synthesis gas, which is then cleaned to remove contaminants such as particulate matter and sulphur. The resulting fuel gas is used to fire a combined cycle. Gasifiers applied for IGCC fuelled with (bituminous) coal are typically oxygen-blown entrained flow gasifiers, operating at pressures between 30 and 70 bar and temperatures from 1000 up to 1500°C. Several technologies are available (E-gas, Prenflo, Shell, Texaco), the main differences being the coal feed (either dry or slurry feed), number of gasifier stages (one or two) and syngas cooling (water quench or heat exchangers). IGCC is not as well developed as PC; today only four coal-fired units are in operation, although there are several other IGCC units using coke and residual oil as feedstock and more units are planned [71]. The existing coal-fired IGCC units have a capacity between 250 and 335 MW<sub>e</sub> and a net electric efficiency between 38 and 45% [72]. A major bottleneck for large-scale

commercial application is the relatively high investment costs in comparison to PC. Moreover, IGCC is a technically more complex system than PC, leading to a relatively low availability and poor operational flexibility. In order to increase plant availability to a level of 85-90%, a spare gasifier might be required, which will increase total investment costs.

CO<sub>2</sub> capture at IGCC plants can be performed by means of post-combustion capture, pre-combustion capture and oxyfuel combustion.

### 3.3.1 PRE-COMBUSTION CAPTURE

In this concept, shift reactor(s) are installed after the gas cleaning section to convert CO present in syngas into CO<sub>2</sub> and H<sub>2</sub> by adding steam. Hydrogen is mixed with nitrogen/steam and fed to an adapted gas turbine. The high partial pressure of CO<sub>2</sub> after the shift reactor makes physical absorption the most appropriate (commercially available) CO<sub>2</sub> capture technology, because this process is less energy intensive than a chemical absorption process. In physical absorption processes, CO<sub>2</sub> is recovered from the absorbent (e.g. Selexol) by reducing the pressure in flash drums, which is less energy-intensive than stripping CO<sub>2</sub> using heat as applied in chemical absorption.

Various options have been proposed to improve the energetic and/or economic performance of the capture process. Alternatives for physical absorption are SEWGS or inorganic membranes for separation of CO<sub>2</sub> (or CO) and hydrogen. Membranes could eventually be integrated with the shift reactor (water gas shift membrane reactor). Although not further considered in this study, cost reductions can be achieved by co-capture and storage of CO<sub>2</sub>/H<sub>2</sub>S (acid gas), a strategy applied in parts of North America to dispose CO<sub>2</sub> and H<sub>2</sub>S present in natural gas. In an IGCC, H<sub>2</sub>S is normally removed from the syngas prior to CO<sub>2</sub> removal and sent to a sulphur recovery system (Claus and scot unit), where it is converted into elemental sulphur. In a co-capture/storage system, the sulphur recovery unit is omitted and H<sub>2</sub>S and CO<sub>2</sub> are compressed and stored together.

### 3.3.2 POST-COMBUSTION CAPTURE AND OXYFUEL COMBUSTION

In a configuration with post combustion capture, a chemical absorption unit is installed after the gas turbine. The advantage above the pre-combustion strategy is that no shift reactor and no modifications to the gas turbine are required. The oxyfuel strategy encompasses a larger ASU, which also provides oxygen for syngas combustion. This requires the development of new turbines designed to operate on CO<sub>2</sub>/H<sub>2</sub>O as working fluid. The Water cycle fuelled with syngas (IG-Water) can be considered as an IGCC oxyfuel strategy, although this technology is strictly spoken a Rankine cycle.

### 3.3.3 KEY PERFORMANCE DATA

The key performance data for IGCC concepts with various CO<sub>2</sub> capture strategies are shown in Table 3. Various studies comparing IGCC with

post, pre and oxyfuel combustion clearly show that pre-combustion by physical absorption is the most efficient and least capital-intensive CO<sub>2</sub> capture technology [6,7,73], which will be further considered here. The large range in efficiency and capital costs for IGCC can be explained mainly by the different IGCC configurations i.e. gasifier technology (slurry/dry feed), syngas cooling (quench/heat exchangers), gas turbines (F, G and H-class), individual performance of each element and level of integration between the elements. Also the coal type (rank) has a significant impact on cold-gas efficiency and capital costs, with decreasing efficiencies and increasing costs with lower coal ranks [74]. We use a range in investment costs and performance data representing Texaco and Shell gasification technology integrated with two GE 9FA turbines<sup>14</sup> using Australian bituminous coal as feedstock [75]. The Shell technology represents high-performance, capital-intensive dry feed gasifiers with a heat recovery boiler, whereas the Texaco concept represents less efficient and less capital-intensive slurry feed gasifiers with product gas cooling by water quench<sup>15</sup>. The efficiency figures are relatively conservative in comparison to other IGCC forecasts, which might be explained by the relatively high production of ungasified carbon and the level of heat integration [75]. However, a comparison with other studies considering the same IGCC configuration (same gasifier, syngas cooling and turbines) without CO<sub>2</sub> capture revealed very similar results [74], so the data used here are considered reliable and representative.

In the longer term, significant improvements are foreseen in the gasifier, gas turbine and air separation unit. We consider an advanced IGCC envisioned for the year 2020 by Foster Wheeler, featuring dry-feed, 2-stage entrained flow gasification, an improved (H) turbine, and ion transfer membrane air separation replacing conventional, energy-intensive cryogenic ASU [75]. This configuration does not account for further improvements in CO<sub>2</sub> capture technology and might therefore prove slightly conservative. A case study on a coke gasification IGCC plant indicates that a novel cryogenic technology for CO<sub>2</sub> separation promises 16% reduction in CO<sub>2</sub> avoidance costs versus a base case with Selexol [77]. Various studies on the techno-economic impact of high-temperature membranes to separate CO<sub>2</sub> from H<sub>2</sub>, eventually integrated with water gas shift reactor, indicate that the IGCC conversion efficiency can be increased and investment costs reduced in comparison to physical absorption [7,29,78].

The Water cycle fuelled with syngas might prove a promising alternative to IGCC with pre-combustion capture. We use data for a configuration integrating an E-gas gasifier with a set of advanced turbines and single reheat. The intermediate pressure turbine has a TIT of 1427°C, the rated temperature of H-turbines [10]. Although the efficiency is somewhat conservative in comparison to other projections [46,47], we choose to use data from this study, as it contains transparent data on both efficiency and investment costs.

Annual O&M costs are between 3.5 and 5% of TPC for IGCC with physical

14. H-class turbines for hydrogen rich gas as in IGCC are not expected to be commercially available in the short term [75].

15. Shell specified 2x50% capacity gasifiers and ChevronTexaco specified 4x33% capacity gasifier in order to achieve a capacity factor of 85% [75]. In various studies on IGCC costs, Texaco-based IGCC are designed with a spare gasifier in order to achieve 90% target availability [74]. Shell's membrane cooled gasifiers have a higher availability than refractory lined Texaco gasifiers and might therefore not need a spare gasifier. In case an additional spare gasifier is required for the Shell case, TPC (€/kW<sub>e</sub>) increase with circa 10% [74]. According to calculations performed by Kreutz et al. [76] for a plant with quench cooling, COE increases with 10-12% when accounting for a spare gasifier.

absorption [10,18,20,75] and approximately 3.5% for IG-Water plants [10,46].

TABLE 3. Key parameters of IGCC plants (using bituminous coal) with CO<sub>2</sub> capture from literature review

Conversion technology	CO <sub>2</sub> capture technology	Net electric efficiency <sup>a</sup>	Efficiency penalty <sup>b</sup>	CO <sub>2</sub> capture efficiency	TCR (€/kW) <sup>c</sup>	Sources
IGCC (E-gas, Prenflo, Shell, Texaco) <sup>d</sup>	Pre-combustion (Physical absorption)	32-43%	5-9% points	85-90%	1500-2170	[6,7,9,10, 18,20,73, 75,76, 79,80]
IGCC (Prenflo)	Pre-combustion (MWGS)	40-44%	7-8% points	80%	1440-1500	[7,78]
IGCC 2020	Pre-combustion (Physical absorption)	43%	6% points	85%	1500	[75]
IGCC (Texaco)	Post-combustion (Chemical absorption)	37-38%	8-10% points	90%	2290-2430	[6,73]
IGCC (Texaco)	Oxyfuel combustion (ASU)	39%	8% points	90%	2280	[73]
IG-Water (E-gas, Texaco) <sup>e</sup>	Oxyfuel combustion (ASU)	32-55%	8% points	100%	1420-1550	[10,46, 47]

<sup>a</sup> Including CO<sub>2</sub> compression to 110 bar.

<sup>b</sup> The efficiency penalty refers to the difference with a standard IGCC.

<sup>c</sup> Due to the limited number of IGCC plants constructed, it is hard to assess a scaling factor for IGCC. We assume a scaling factor of 0.8 used by Kreutz et al. [76] to scale investment costs to 600 MWe.

<sup>d</sup> An IGCC plant with a Texaco gasifier and F-class turbine represents the lower value in the efficiency range. The upper value in efficiency range represents E-gas/Shell gasifiers integrated with G/H turbines. The efficiency penalty and additional investment costs caused by CO<sub>2</sub> capture for Texaco gasifiers are lower in comparison to Shell gasification technology, since no additional steam must be taken from the steam cycle required for CO shift, as the syngas itself contains sufficient steam [75].

<sup>e</sup> With current technology (series of conventional steam turbines), a net electric efficiency of 32% can be achieved [46]. When applying a high-pressure, high-temperature steam turbine (103 bar, 816°C) followed by an intermediate and low pressure, high-temperature turbine with relatively high TIT (26 bar, 1427°C and 3 bar, 1427°C), net efficiencies in the order of 48-51% can be reached, a decrease of 3% points in comparison to Water cycle without CO<sub>2</sub> compression. These turbines are not commercially available for power generation, but have been used in aerospace applications. The most advanced concept, assuming TIT of 1649°C based on turbine development goal of the US-DOE Vision 21 program and expansion of high-pressure nitrogen from the ASU, may reach an efficiency of 53-55% [46,47]. Further improvements can be realised by use of oxygen transport membranes. The efficiency and investment costs of the oxygen plant (representing 10-15% of IGCC investment costs) can be reduced by 37% and 35%, respectively, resulting in IGCC efficiency increase of 2% and investment cost decrease of 7% [75,81].

### 3.4 FUEL CELLS

Fuel cells convert chemical energy of a gaseous fuel directly into electricity and heat. Fuel is oxidised electrochemically, which implies lower exergy losses than direct combustion. Fuel cells consist of an electrolyte layer between anode and cathode, functioning as a membrane permeable for ions. The anode and cathode are connected to close the circuit, i.e. electrons generated at the anode flow to the cathode. Fuel cells are generally classified by the electrolyte material and operating temperature. Low-temperature fuel cells include the alkaline fuel cell (AFC), phosphoric acid fuel cell (PAFC) and proton exchange membrane fuel cell (PEMFC). Molten carbonate fuel cell (MCFC) and solid oxide fuel cell (SOFC) are high-temperature fuel cells. MCFC and SOFC are most appropriate for stationary power production (from a few hundred kW up to a few MW), due to their high electrical (and cogeneration) efficiencies<sup>16</sup> [83,84]. The efficiency of SOFC and MCFC can be further increased by integration with gas turbines (MCFC or SOFC-GT hybrids).

Various MCFC and SOFC demonstration units with typical capacities of a few hundred kW up to 1 MW have been operated successfully and more tests are planned. In 1997, a pilot project started with a 100 kW SOFC cogeneration plant in the Netherlands, which achieved 46% electrical efficiency and a 25% thermal efficiency. In California, the world's first SOFC-GT hybrid (220 kW) has demonstrated 53% electrical efficiency [84]. Several companies are at the threshold of introducing MCFC and SOFC modules at the market.

High-temperature fuel cells can be fuelled with hydrogen, syngas or natural gas. Due to the high temperature at which SOFC and MCFC are operated, reforming of natural gas and the subsequent shift can be performed in the fuel cell itself. In SOFC/MCFC configurations fuelled with natural gas or syngas, CO<sub>2</sub> capture can be performed after the fuel cell ('post-fuel cell capture'). Alternatively, hydrogen can be produced by reforming/partial oxidation of natural gas or coal gasification upstream the fuel cell<sup>17</sup>. CO<sub>2</sub> can be removed after syngas is shifted by means of physical solvents, membranes or adsorbents, also referred to the 'pre-fuel cell CO<sub>2</sub> capture' strategy. Below, we discuss various capture options accounting for the specific features of MCFC and SOFC. Note that CO<sub>2</sub> capture at fuel cells are in fact oxyfuel concepts, since fuel is converted in an atmosphere without nitrogen.

#### 3.4.1 MOLTEN CARBONATE FUEL CELL

The molten carbonate fuel cell has an electrolyte of alkali carbonates, which are fluid at the fuel cell operating temperature of ~650°C. At the cathode, O<sub>2</sub> and CO<sub>2</sub> and 2 electrons form CO<sub>3</sub><sup>2-</sup> ions. These ions are transported across the electrolyte to the anode, where they react with hydrogen to form H<sub>2</sub>O, CO<sub>2</sub> and 2 electrons. CO<sub>2</sub> is generally recycled from the anode to the cathode. A possible configuration to integrate CO<sub>2</sub> capture is to burn the anode exhaust with air, recirculate the combustion flue gas rich in CO<sub>2</sub> to the cathode and use the heat of the cathode

16. SOFC (hybrids) is the ideal technology for decentralised power (and heat) generation, due to their high efficiency at modest capacities, as a consequence of their modular design. This makes SOFC also suited for offshore power generation with CO<sub>2</sub> capture and injection into (nearly) depleted oil or gas fields or aquifers [82]. Another possibility is to install SOFC (hybrids) on top of a natural gas field, extracting gas to fuel the SOFC and injecting CO<sub>2</sub> into the gas field to enhance gas recovery [68]. In this way, the need for natural gas and CO<sub>2</sub> transport is avoided.

17. Sulphur content in syngas produced by coal gasification needs to be reduced further after the gas clean up section (by means of a ZnO bed) to acceptable levels for use in fuel cell [10].

exhaust (which contains only ~5 mol% CO<sub>2</sub>) to generate steam in a HRSG required for CO<sub>2</sub> capture by chemical absorption [85,86]. The disadvantage of this configuration is that CO<sub>2</sub> rich anode gas is diluted with mainly nitrogen. This can be avoided by installing a MCFC downstream to a gas turbine or coal-fired power plant, in which CO<sub>2</sub> from the exhaust is used as reactant at the cathode and transferred across the electrolyte, resulting in a concentrated CO<sub>2</sub> stream on the anode side [85,87]. CO<sub>2</sub> is then captured from the anode exhaust and the remaining H<sub>2</sub>, CO (and unconverted CH<sub>4</sub>) can be burned in a catalytic burner with cathode exhaust or redirected to the power cycle upstream of the MCFC. These MCFC hybrids are not further considered here as the energetic and/or economic performance of a greenfield plant has not been assessed.

### 3.4.2 SOLID OXIDE FUEL CELL

Solid oxide fuel cells have ceramic electrolytes (generally yttria-stabilised zirconia), which require temperatures around 1000°C to achieve sufficient ionic conductivity. At the cathode, oxygen atoms are ionised and transported by the electrolyte to the anode, where oxygen ions react with hydrogen. The cathode exit stream is oxygen-depleted air and the anode exit contains principally CO<sub>2</sub>, CO, H<sub>2</sub>O, and H<sub>2</sub> (when using natural gas as fuel). In many of the proposed schemes, the fuel cell is integrated with a gas turbine. In these hybrid configurations, compressed air is fed to the fuel cell cathode and the hot cathode exhaust is expanded in a gas turbine.

High CO<sub>2</sub> concentration in the anode exhaust makes SOFC an interesting target for CO<sub>2</sub> capture. If CO<sub>2</sub> capture is required, the anode and cathode exhaust need to be separated by means of a seal in order to prevent CO<sub>2</sub> rich anode exhaust to be diluted with nitrogen. CO<sub>2</sub> can be captured from the anode offgas by means of cryogenic separation, chemical absorption or shifting the anode exhaust for physical absorption. The H<sub>2</sub>(/CO) rich syngas can be recirculated to the anode inlet or can be burned with cathode outlet e.g. to drive a gas turbine. Another possibility is to oxidise the anode exhaust gas with pure oxygen in an afterburner, thereby increasing CO<sub>2</sub> concentration for capture. The afterburner can exist of a burner fuelled with pure oxygen [88], an oxygen conducting membrane (OCM) reactor [89], a second SOFC (electrochemical afterburner) [82], or a water gas shift membrane reactor (WGSMR) afterburner [90]. In an OCM afterburner, oxygen ions and electrons are simultaneously transported over the membrane, whereas in an SOFC the electrons are transported in an external circuit, thereby generating additional power.

In a WGSMR, H<sub>2</sub> in the anode exhaust permeates through the membrane to be oxidised with the cathode exhaust.

The ZEC (zero emission coal) process is a highly innovative cycle, which combines a hydrogasifier, carbonation-calcination reactors and a pressurised SOFC. Coal is gasified into a gas consisting of mainly methane, using hydrogen and water. Methane is reformed to produce

hydrogen through the use of a carbonation reaction, which converts CaO and CO<sub>2</sub> into CaCO<sub>3</sub> and supplies energy required to drive the reforming reaction. Hydrogen is partly recycled to the gasifier and the rest is used to produce electricity in an SOFC. The process heat from the SOFC is used to regenerate CaO by calcination of CaCO<sub>3</sub>, thereby producing CO<sub>2</sub> that can be stored [91]. Although very high efficiencies are claimed, this technology still requires large developments to achieve the process integration that is crucial for the technical and economical performance.

3.4.3 KEY PERFORMANCE DATA

Table 4 gives the techno-economic data of all MCFC and SOFC configurations with CO<sub>2</sub> capture. There is a large variation in both conversion efficiency and cost projections of fuel cell (hybrids). For fuel cell hybrids fired with coal, we consider the IG-SOFC-GT analysed in the NETL study [10], as this is the most extensive study with most updated cost estimations of fuel cells. Although the ZEC concept promises a high-efficient technology, we do not further include it in our analysis, as projected capital costs are still relatively high. Hence forecasted COE are not structurally lower than COE for other advanced gasification concepts (even at higher coal prices of 3 €/GJ). Natural gas-fired SOFC-GT systems are mainly foreseen for decentralised power generation. We use data for a 20 MW<sub>e</sub> system with a 2<sup>nd</sup> SOFC and afterburner assessed by Lokurlu et al. [92], the only analysis available on decentralised SOFC-GT systems with capture containing data on both efficiency and costs. Annual O&M costs for SOFC-GT systems are between 3 and 3.5% of TPC [10,79,86].

TABLE 4. Key parameters of SOFC and MCFC plants with CO<sub>2</sub> capture from literature review

Conversion technology	CO <sub>2</sub> capture technology	Net electric efficiency <sup>a</sup>	CO <sub>2</sub> capture efficiency	TCR (€/kW <sub>e</sub> ) <sup>b</sup>	Sources
<i>Fuel cells fired with coal</i>					
IG-MCFC-ST	Pre-fuel cell (shift, physical absorption)	35%	68%	3080	[86]
IG-MCFC-ST	Post-fuel cell (chemical absorption)	36%	76%	2720	[86]
IG-SOFC-GT <sup>c</sup>	Various strategies	45-50%	90%	1670-1760	[6,10]
ZEC (IG-SOFC-ST)	Pre-fuel cell (chemical adsorption)	71%	100%	1910	[91]
<i>Fuel cells fired with natural gas</i>					
MCFC-ST	Pre-fuel cell (shift, physical absorption)	49%	58%	2060	[86]
MCFC-ST	Post-fuel cell (chemical absorption)	55%	91%	1870	[86]
SOFC-GT <sup>d</sup>	Post-fuel cell (chemical absorption)	46-69%	90%	990-1600	[6,10,85,86]
SOFC-GT	Post-fuel cell (shift, physical absorption)	69%	90%	-	[85]
SOFC-GT <sup>e</sup>	Post-fuel cell (afterburner)	59-67%	80-100%	1530-1870	[55,85,92-94]

- <sup>a</sup> Including CO<sub>2</sub> compression to 110 bar.
- <sup>b</sup> The capacity of coal-fired systems is in the range 500-750 MW<sub>e</sub>, whereas natural gas-fired systems range from 10-500 MW<sub>e</sub>. For the coal-fired systems, we apply a scale factor of 0.9 (due to modular character of fuel cells, the scaling relation is expected to be less strong than IGCC). Investment costs of natural gas-fired systems are not corrected for scale, as the scaling relation for such hybrid systems over such a wide capacity range is unknown and the cost-breakdown of investment costs is usually not given.
- <sup>c</sup> Parsons et al. [10] consider a coal gasifier, which produces syngas for the SOFC anode and a gas turbine. Syngas to the gas turbine enters a hydrogen separation device (membrane), where CO is shifted and CO<sub>2</sub> removed, after which H<sub>2</sub> is sent to the gas turbine. Anode exhaust is combined with CO<sub>2</sub> from the HSD and burned in a catalytic combustor. This configuration results in 50% efficiency versus 56% without capture.
- <sup>d</sup> Recent study performed by Parsons I&TG indicated that the efficiency of a 560 MW<sub>e</sub> cascaded humidified advanced turbine-SOFC hybrid dropped from 66% without CO<sub>2</sub> capture to 46% when CO<sub>2</sub> is recovered from the flue gas by means of MEA [79], which is considerably lower than the forecasted 60% for a similar SOFC-gas turbine configuration with post-fuel cell CO<sub>2</sub> capture analysed in earlier studies [6,86] or 69% forecasted in [85]. The main difference lies in the amount of natural gas that is routed to the duct burner to generate steam for CO<sub>2</sub> regeneration, which is much higher in the Parsons study.
- <sup>e</sup> Efficiency data cover all afterburner concepts (2<sup>nd</sup> SOFC, OCM and WGS<sub>MR</sub>). A recent thermodynamic analysis of these three afterburner technologies integrated with a 20 MW<sub>e</sub> SOFC-GT revealed that applying a 2<sup>nd</sup> SOFC results in the highest efficiency. This configuration requires a larger surface area to oxidise the remaining fuel than an OCM afterburner [93]. This is confirmed by an analysis performed for a 20 MW<sub>e</sub> hybrid system, which also accounted for costs [92]. By installing an additional burner after the 2<sup>nd</sup> SOFC to increase TIT, capital costs are minimised at the expense of a lower efficiency and CO<sub>2</sub> recovery [92], resulting in lower COE in comparison to the standard configuration.

### 3.5 OVERVIEW ELECTRICITY PRODUCTION TECHNOLOGIES

Table 5 summarises key data of the most promising state-of-the art and advanced electricity options with CO<sub>2</sub> capture and compression to 110 bar.

TABLE 5. Key parameters of electricity production technologies with CO<sub>2</sub> capture

Feedstock	Conversion technology	CO <sub>2</sub> capture technology	Net electric efficiency	CO <sub>2</sub> capture efficiency	TCR (€/kw)	O&M
<i>State-of-the-art technologies</i>						
Coal	PC	Post-comb (MEA)	35%	88%	2080	5.8%
	IGCC	Pre-comb (Selexol)	32-35%	85%	1770-2170	4.8 - 5.2%
Natural gas	NGCC	Post-comb (MEA)	47%	85%	920	4.3%
<i>Advanced technologies</i>						
Coal	PC	Improved post-comb (MEA)	40%	85%	1520	6.5%
	IGCC	Pre-comb (Selexol)	43%	85%	1500	5%
	IG-Water	Oxyfuel (ASU)	41%	100%	1530	3.7%
	IG-SOFC-GT	Various (membrane/cat. combustor)	50%	90%	1760	3.3%

Feedstock	Conversion technology	CO <sub>2</sub> capture technology	Net electric efficiency	CO <sub>2</sub> capture efficiency	TCR (€/kw)	O&M
Natural gas	NGCC	Improved post-comb (MEA)	55%	85%	650	4.8%
	MR-CC	Pre-comb (HMCM)	53%	100%	940	4% <sup>a</sup>
	CLC	Oxyfuel (separate combustion)	51%	100%	900	4% <sup>b</sup>
	AZEP	Oxyfuel (oxygen membrane)	50%	100%	900	4% <sup>b</sup>
	SOFC-GT (20 MW <sub>e</sub> )	Oxyfuel (electrochemical afterburner)	59%	80%	1530	3%

<sup>a</sup> O&M costs for the MR-CC are 5.2% of TPC versus 6% for NGCC with advanced post-combustion capture [42]. We corrected the O&M costs for the MR-CC so that it corresponds in proportion to the value we adopted for advanced NGCC with post-combustion capture.

<sup>b</sup> The uncertainty in investment costs of AZEP and CLC justifies the use of a more aggregate estimate of O&M costs, which is set at 4% of TPC. Variable O&M costs for single cycle AZEP were set at 4% of TPC [58].

## 4 HYDROGEN PRODUCTION TECHNOLOGIES

Steam methane reforming is the most common and developed technology used for hydrogen production at large scales. It is likely to remain a dominant large-scale hydrogen production technology in the nearby future. Partial oxidation is the most appropriate technology to produce hydrogen from heavier feedstocks such as heavy oil residues and coal [95].

### 4.1 HYDROGEN PRODUCTION FROM NATURAL GAS

#### 4.1.1 STEAM METHANE REFORMING

In fired tubular reformers, steam and natural gas are reacted over a nickel-based catalyst to produce syngas at temperatures of about 800-900°C. In order to sustain this endothermic reaction, heat is supplied to the reforming reactor by burning part of the natural gas in a furnace. Syngas is cooled and then shifted in the wgs reactor. In older plants, CO<sub>2</sub> is subsequently removed by means of a chemical absorption unit. Modern hydrogen plants apply pressure swing adsorption (PSA) to separate hydrogen from the other components, which produces higher quality hydrogen (99.999% against 95-98% for scrubbing systems) at feedstock pressure (circa 25 bar). Purge gas from the PSA is generally redirected to the furnace. Depending on the design, steam or electricity is imported/exported. The conversion efficiency of large-scale SMR varies between 74 and 85%<sup>18</sup> [12,13,95,96].

Relatively few studies have been performed to assess efficiency loss and costs associated with CO<sub>2</sub> capture at SMR plants. CO<sub>2</sub> can be captured from the furnace off-gas by means of a MEA unit (post-combustion

18. All electricity/steam input/outputs converted into primary terms.

strategy), which would require large steam consumption due to the low CO<sub>2</sub> partial pressure. Alternatively, CO<sub>2</sub> can be captured by installing an MDEA unit between the shift reactor and the PSA unit. Although this has not been applied in hydrogen plants to date, MDEA is a proven technology. When high-purity hydrogen is not required (when applied in conventional turbines or heating purposes), the PSA unit can be omitted (representing the older hydrogen plants).

#### 4.1.2 AUTOTHERMAL REFORMING

Steam methane reforming can be combined with partial oxidation, a process referred to as autothermal reforming. Basically, steam and oxygen (or air) are injected into the reformer, causing the reforming and oxidation reaction to occur simultaneously. The exothermic POX reaction, in which the feedstock reacts (catalytically) with oxygen in sub-stoichiometric conditions, provides heat to endothermic SMR reaction. Hence, the use of ATR allows for higher CO<sub>2</sub> recovery in comparison to SMR, as CO<sub>2</sub> emissions from the furnace at a SMR plant are relatively uneconomic to capture due to the low partial pressure in furnace exhaust. Investment costs of oxygen-blown ATR are about 15-25% lower than fired tubular reformers, but the costs of oxygen supply make ATR less attractive than SMR even for large-scale plants (650 MW<sub>H<sub>2</sub></sub>) [97]. The integration of ceramic ion transport membranes for oxygen production with an ATR reactor opens new possibilities for high-efficient and low-cost hydrogen production with CO<sub>2</sub> capture in the long term [98].

#### 4.1.3 MEMBRANE REFORMING

Large-scale hydrogen production by means of membrane reforming could reduce investment cost versus conventional SMR with MDEA (by omission of shift reactor, absorption unit and reduced CO<sub>2</sub> compression requirement). However, CO<sub>2</sub> avoidance costs are nearly equal, as the MR case requires electricity import (all excess steam is required for sweep), whilst the conventional system has a large electricity export. In order to become feasible for low-cost hydrogen production, membrane costs need to be reduced by applying thinner membranes without compromising selectivity [99].

MR might prove more interesting on a small scale. A recently accomplished study demonstrates that membrane reformers enable hydrogen production at refuelling stations combined with relatively inexpensive CO<sub>2</sub> capture [100]<sup>19</sup>.

#### 4.1.4 KEY PERFORMANCE DATA

Table 6 provides the ranges in efficiency and investment costs for the different routes of hydrogen production from natural gas with CO<sub>2</sub> capture. For the short term<sup>20</sup>, we consider large-scale SMR with a chemical absorption unit (MDEA) after the shift using data from IEA GHG study [12]. This is technically feasible and more efficient and less costly than installing a MEA unit after the furnace. In the longer term, we

19. CO<sub>2</sub> capture at conventional small-scale reforming units is prohibitive due to relatively high costs of CO<sub>2</sub> capture at such scales [101].

20. Note that short-term hydrogen production is generally foreseen by means of small scale SMR or electrolysis and that CO<sub>2</sub> capture at large SMR units is foreseen in the longer term, if sufficient demand exists. The term short term in this study must be considered in terms of technological maturity

consider both large-scale ATR-ITM with a chemical/physical absorption unit after the shift [14] and small-scale MR [100].

Annual O&M costs for SMR/ATR systems with CO<sub>2</sub> capture are between 3 and 6% of TPC [12-14]. Annual O&M costs for small-scale MR are estimated at 9% of TPC, of which 5% for membrane replacement [100].

TABLE 6. Key parameters of hydrogen production from natural gas with CO<sub>2</sub> capture from literature review

Conversion technology	CO <sub>2</sub> capture technology	Fuel+feed input (GJ/GJ <sub>H<sub>2</sub></sub> )	Electricity input (GJ <sub>e</sub> /GJ <sub>H<sub>2</sub></sub> )	Conversion efficiency <sup>a</sup>	CO <sub>2</sub> capture efficiency	TCR (€/kW <sub>H<sub>2</sub></sub> ) <sup>b</sup>	Source
SMR	MEA after furnace	1.53	0.06	61%	90%	550	[98]
SMR	MDEA after shift	1.37	0	73%	85%	550	[12]
Advanced ATR-ITM	Physical solvent	1.27	0.001	79%	95%	390	[98]
		1.28	0.03	74%	90%	280	[14]
Small-scale MR <sup>c</sup>	Pd membrane	1.26	0.13	65%	70%	610	[100]

<sup>a</sup> Including CO<sub>2</sub> and H<sub>2</sub> compression to 110 and 60 bar, respectively (except MR).

<sup>b</sup> The plants considered here have a large capacity (600-2000 MW<sub>H<sub>2</sub></sub>) and are scaled to 1000 MW<sub>H<sub>2</sub></sub>. A scaling factor of 0.6 can be derived from SMR investment costs of 70 and 700 MW<sub>H<sub>2</sub></sub> [101]. We assumed a scaling factor of 0.75, which is used to scale SMR with larger capacity (>700 MW<sub>H<sub>2</sub></sub>) [14].

<sup>c</sup> Represents a 1.7 MW<sub>H<sub>2</sub></sub> membrane reactor using sweep-gas to maximise CO<sub>2</sub> concentration in the retentate flow (i.e. minimise CH<sub>4</sub> and H<sub>2</sub> concentration) to enable direct transport and storage. H<sub>2</sub> is pressurised to 480 bar (in contrast to other hydrogen production plants, which have been standardised to 60 bar) to enable fast cascade-filling into vehicle tanks at 350 bar [100].

#### 4.2 COAL GASIFICATION

Coal gasification (CG) for hydrogen production has large similarities to IGCC with CO<sub>2</sub> capture. The main difference is the addition of a PSA unit to produce high-purity hydrogen. Syngas is produced in an oxygen-blown, entrained flow gasifier, cooled and cleaned before entering the WGS reactor. H<sub>2</sub>S and CO<sub>2</sub><sup>21</sup> are removed by means of physical absorption and the clean, shifted syngas is purified in a PSA unit to deliver 99.999% pure hydrogen at feed pressure. The PSA could be omitted to produce fuel-grade hydrogen (~93%) suitable for combustion, reducing hydrogen production costs with circa 5% [76]. PSA purge gas can be used to superheat steam or can be combusted for power generation in a combined cycle or Rankine cycle (co-production systems). By lowering the steam-to-carbon ratio or directing a part of the syngas to the gas turbine (by-passing the PSA), the hydrogen-to-electricity ratio can be adapted without major thermodynamic consequences, allowing these co-production systems<sup>22</sup> to match their output to electricity and hydrogen demands [80].

In advanced coal gasification systems, ITM might be applied for oxygen production and the Selexol and PSA unit could be replaced by inorganic membranes to separate hydrogen from CO<sub>2</sub>. Both metallic (Pd and Pd alloys) and ceramic membranes are being investigated. A

21. Systems without CO<sub>2</sub> capture do not necessarily need a physical absorption unit to remove CO<sub>2</sub>, but doing so has the advantage of reducing the flow to the PSA and increasing the heating value of the purge gas to a point that makes it feasible for use in a gas turbine [80].

22. For simplicity, we allocate all CO<sub>2</sub> emissions to hydrogen production.

techno-economic study performed by Kreutz [102] assessing hydrogen production with pd alloy membranes versus Selexol and PSA indicates that cost advantages of membrane separators/reactors are moderate. Ceramic membranes have the advantage of being cheaper, but these membranes are less selective, resulting in lower hydrogen purity (>99.5%) [13]. Hydrogen of this purity cannot be applied in PEMFC, as CO poisons the catalyst. Table 7 shows the range in conversion efficiency and investment costs of hydrogen production by means of coal gasification with CO<sub>2</sub> capture. For the short term, we use the data from the analysis performed in [76,80]. This is the most detailed and transparent study, in which all optimisation strategies with commercially ready technology have been assessed. The efficiency figures are relatively high in comparison to other estimates, which is explained by the more optimistic assumptions in gasifier, power generation, PSA performance and overall integration [80]. In the longer term, overall efficiencies close to 70% can be achieved. We use the values for the advanced system with ITM [14]. As the forecasted efficiencies of a system with ceramic membranes are slightly lower and hydrogen is not sufficiently pure for use in PEM fuel cells, this configuration is not further considered here. However, it might be an interesting option to produce fuel grade hydrogen, as plants with ceramic membranes are more efficient and less capital-intensive than conventional plants with PSA and Selexol [13]. Annual O&M costs for hydrogen production by coal gasification with CO<sub>2</sub> capture are between 3.5 and 6% of TFC [13,14,76].

TABLE 7. Key parameters of hydrogen production from (bituminous) coal with CO<sub>2</sub> capture from literature review

Conversion technology	CO <sub>2</sub> capture technology	Fuel input (GJ/GJ <sub>H<sub>2</sub></sub> )	Electricity in/output (-) (GJ <sub>e</sub> /GJ <sub>H<sub>2</sub></sub> )	Conversion efficiency <sup>a</sup>	CO <sub>2</sub> capture efficiency	TCR (€/kW H <sub>2</sub> ) <sup>b</sup>	Source
CG (Texaco/ E-gas) <sup>c</sup>	Selexol	2.30	0	43%	97%	1640	[12]
		1.94	0.03	50%	92%	810	[13]
		1.48	0.11	59%	90%	790	[14]
CG (Texaco)	Pd- membrane	1.74	-0.04	62%	91%	840	[76,80]
		1.68	-0.02	62%	100%	680	[102]
CG (E-gas) <sup>d</sup>	Ceramic membrane	1.43	0.04	67%	94%	610	[13]
		1.80	-0.05	61%	94%	780	
Advanced CG <sup>e</sup>	Selexol	1.35	0.05	69%	90%	600	[14]

<sup>a</sup> Including CO<sub>2</sub> and H<sub>2</sub> compression to 110 and 60 bar respectively.

<sup>b</sup> Plants considered have capacities in the range from 430-1030 MW<sub>H<sub>2</sub></sub> and are scaled to 1000 MW<sub>H<sub>2</sub></sub>. A scaling factor of 0.8 is assumed, as applied for IGCC [76] and hydrogen production by coal gasification [14]. An economic analysis evaluating the impact of scale on investment costs of hydrogen plants by biomass gasifiers, showing large similarity to plants considered here, revealed that the scaling factors for plants larger than 400 MW<sub>th</sub> approach 0.9 [103].

<sup>c</sup> Klett et al. [13] consider a configuration with net electricity export, but do not include H<sub>2</sub> and CO<sub>2</sub> compression, so that it becomes a net importer if this is accounted for. We do not further consider these data, nor the atypical data presented in [12].

- <sup>d</sup> Hydrogen separation device operating at 600°C. Two configurations are considered. The most efficient, low-cost option configuration is designed to recover 95% of hydrogen. However, the electricity generated from the remaining purge-gas burned is not sufficient for CO<sub>2</sub> and H<sub>2</sub> compression. The sub-optimal (more conservative) represents a configuration with 80% H<sub>2</sub> recovery. The power produced from the purge gas is sufficient produced for CO<sub>2</sub> and H<sub>2</sub> compression.
- <sup>e</sup> Includes improvements in gasifier technology (high-pressure) and ITM for oxygen production.

#### 4.3 OVERVIEW HYDROGEN PRODUCTION TECHNOLOGIES

Table 8 summarises the key data of promising state-of-the-art and advanced hydrogen production options with CO<sub>2</sub> capture and compression to 110 bar and hydrogen delivered at 60 bar. We use average annual O&M of 4% of TPC for large-scale hydrogen production from coal and natural gas.

TABLE 8. Key parameters of hydrogen production technologies with CO<sub>2</sub> capture

Conversion technology	CO <sub>2</sub> capture technology	Fuel+feed input (GJ/GJ <sub>H<sub>2</sub></sub> )	Electricity in/output(-) (GJ <sub>e</sub> /GJ <sub>H<sub>2</sub></sub> )	Conversion efficiency	CO <sub>2</sub> capture efficiency	TCR (€/kW <sub>H<sub>2</sub></sub> )	O&M
<i>State-of-the-art technologies</i>							
SMR	MDEA	1.37	0	73%	85%	550	4%
CG	Selexol	1.74	-0.04	62%	91%	840	4%
<i>Advanced technologies</i>							
Adv. ATR	MDEA	1.28	0.03	74%	90%	280	4%
Adv. CG	Selexol	1.35	0.05	69%	90%	600	4%
MR (2 MW <sub>H<sub>2</sub></sub> )	Pd membrane	1.26	0.13	65%	70%	610	9%

## 5 RESULTS

### 5.1 ELECTRICITY PRODUCTION

Figure 2 shows the electricity production costs, assuming a discount rate of 10%, 20 years lifetime and a capacity factor of 85%. We used coal and natural gas price projections for 2020 for large industrial users of 1.7 and 4.7 €/GJ, respectively, including commodity, transmission and distribution costs, taxes and VAT [104]. The impact of fuel price on COE will be assessed by means of sensitivity analyses in the discussion.

We did not account for carbon taxes or credits in our calculations.

When CO<sub>2</sub> capture is applied in the short term, COE will increase strongest at PC (from 4.6 to 6.4 €/ct/kwh), followed by NGCC (from 4.2 to 5.6 €/ct/kwh) and IGCC (from 4.8 to 5.8 €/ct/kwh). Incremental COE at NGCC is lower than for PC, as less CO<sub>2</sub> is captured per kwh. More optimistic NGCC designs, in which H turbines are integrated with state-of-the-art MEA [21] and G turbines that are heavily integrated with a chemical absorption unit using KS-I solvent [37] may reach COE levels of

5.2-5.3 €/kwh. Even though IGCC technology without CO<sub>2</sub> capture is relatively capital-intensive in comparison to PC without CO<sub>2</sub> capture, CO<sub>2</sub> capture at IGCC seems to result in lower or comparable COE than PC with capture. This is due to the relatively small efficiency penalty and modest costs of physical absorption in comparison to chemical absorption. The range in IGCC represents the variation in efficiency and investment costs of Shell/Texaco gasification technology. Texaco gasifiers are less capital-intensive and less efficient than Shell gasifiers. CO<sub>2</sub> capture at systems with Texaco gasifiers is more efficient than at systems with Shell gasifiers, i.e. the energy penalty is lower, although the electric efficiency remains lower. The additional costs due to capture are also lower for Texaco systems, resulting in lower COE. For a more optimistic design of an IGCC with a Texaco gasifier and CO<sub>2</sub> capture [76,80], COE might even reach levels slightly below 5 €/kwh.

In the longer term, state-of-the-art technologies (PC, IGCC and NGCC) show potential for efficiency improvement and investment cost reduction. The investment costs of the PC/NGCC plant are assumed to decrease with 20% versus current plants and the investment costs of the capture unit are assumed to decrease with 39% for PC and 32% for NGCC after [21]. The reduction in PC/NGCC technology, but also in the capture technology, may be rather optimistic. A detailed study on the potential to reduce investment cost of current post-combustion technology for NGCC, indicated less than 20% reduction in investment costs [42]. As IGCC is a less mature technology, the potential for efficiency improvement and cost reduction is expected to be larger than for PC and NGCC.

Electricity costs for the Water cycle and SOFC-GT fuelled with syngas are comparable to those for advanced IGCC, although the cost and efficiency projections for these two technologies are relatively uncertain. Note that the most advanced developments for the Water cycle, comprising turbines with higher TIT and ITM for oxygen production were not accounted for.

Figure 2 suggests a ranking of natural gas-fired technologies in the longer term. However, there are large discrepancies and uncertainties in investment costs. First, there is no uniform basis for NGCC baseline costs. The low COE for the advanced NGCC with post-combustion capture can be explained by the optimistic costs reduction in NGCC (and capture) technology. Future cost reductions in NGCC are not explicitly considered in the studies for MR-CC and AZEP. This makes a fair comparison between advanced gas-fired technologies impossible at this stage. Efficiency forecasts of the advanced natural gas-fired technologies, which strongly affect COE, are considered more reliable.

Although the investment costs of SOFC-GT hybrids are relatively high, the calculated COE are only slightly higher than for current decentralised power generation without capture<sup>23</sup>.

23. A 20 MW<sub>e</sub> NGCC is assumed as reference with an efficiency of 40% and TCR of 800 €/kW<sub>e</sub> [69].

Figure 3 presents the amount of CO<sub>2</sub> emitted and CO<sub>2</sub> captured. The sum, the total amount of CO<sub>2</sub> produced, is a measure of the net efficiency. Generally, the amount of CO<sub>2</sub> captured and emitted for coal-

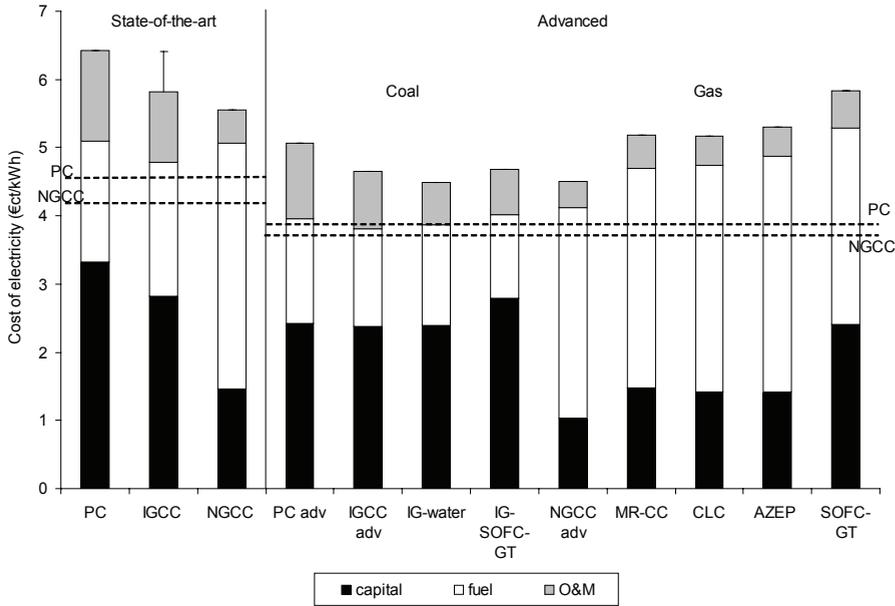


FIGURE 2. Cost of electricity (COE). The dashed horizontal lines represent COE of PC and NGCC without CO<sub>2</sub> capture

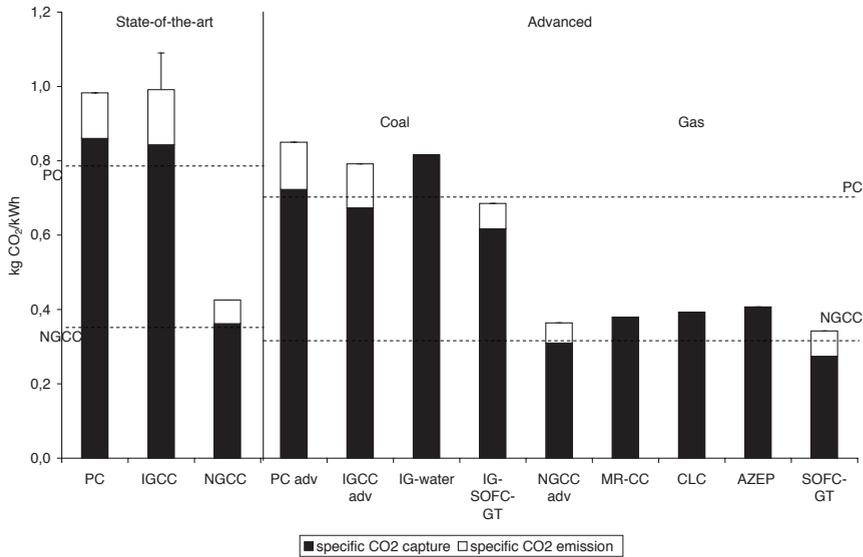


FIGURE 3. Specific CO<sub>2</sub> capture and emission rates for electricity production. The dashed horizontal lines represent emissions of PC and NGCC without CO<sub>2</sub> capture

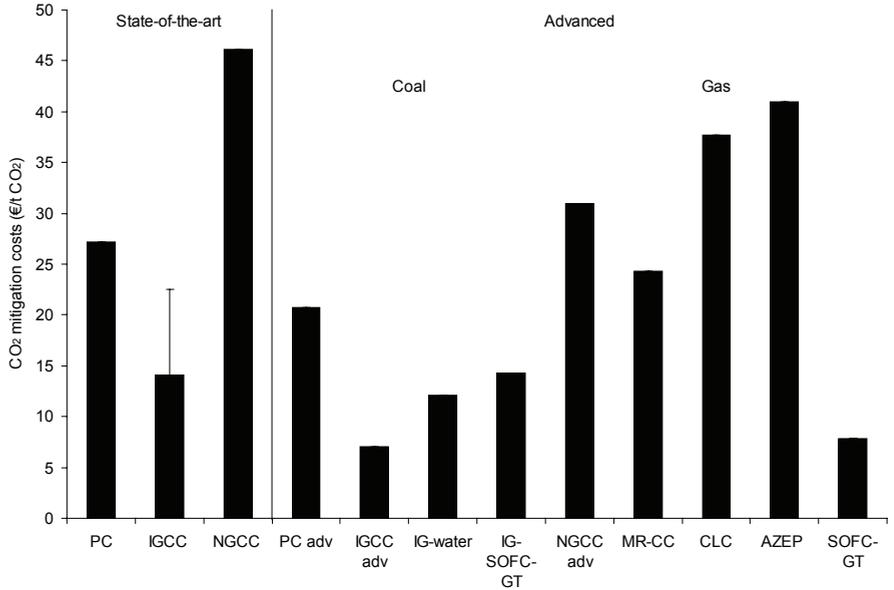


FIGURE 4. CO<sub>2</sub> avoidance costs for electricity production (reference: identical technology without CO<sub>2</sub> capture. For the advanced natural gas-fired options and IG-Water/SOFC, we use NGCC and IGCC as reference, respectively)

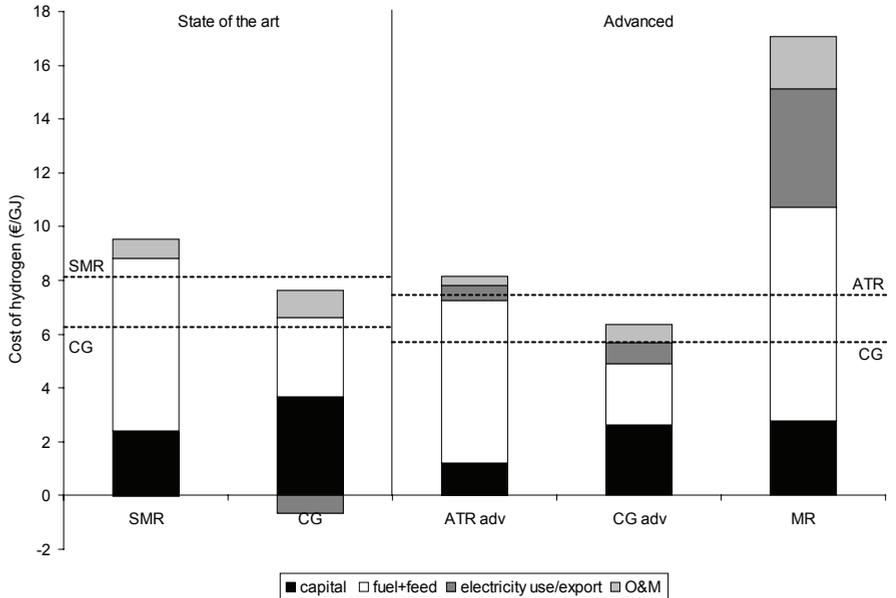


FIGURE 5. Cost of hydrogen (COH). The dashed horizontal lines represent COH of SMR/ATR and CG without CO<sub>2</sub> capture

fired power plants are more than twice as large than natural gas-fired power plants. This is explained by the higher conversion efficiency of natural gas-fired plants and lower CO<sub>2</sub> emission factor of natural gas versus coal (56 and 95 kg CO<sub>2</sub>/GJ, respectively). Total CO<sub>2</sub> produced and emitted is the highest at state-of-the-art IGCC with (conservative) electric efficiencies between 32% and 35%. The efficiencies of advanced gas-fired options are all in the same range. Note that MR-CC, CLC, AZEP (and also SOFC-GT) enable 100% capture at high efficiency<sup>24</sup>, whereas NGCC with post combustion capture achieves only 85-90% capture.

CO<sub>2</sub> avoidance costs of electricity production are presented in Figure 4. It shows that CO<sub>2</sub> avoidance costs for NGCC are higher than for PC (and IGCC), which is due to the higher volume of flue gas and lower CO<sub>2</sub> content. IGCC obviously has the lowest avoidance costs, as the additional investment cost and energy penalty are relatively modest. This technological insight is basically the same in the long term. Less energy and capital-intensive technologies are able to reduce CO<sub>2</sub> avoidance costs to levels below 20 and 40 €/t CO<sub>2</sub> for coal and gas-fired technologies, respectively. Note that the avoidance costs for AZEP and CLC might be reduced when CO<sub>2</sub> capture efficiency is reduced to 85%. SOFC-GT has even lower avoidance costs, which is primarily caused by the low conversion efficiency of the reference system.

24. By installing an afterburner, the efficiency is increased at the expense of a lower CO<sub>2</sub> recovery, with the exception of the SOFC-GT, where the efficiency is decreased.

## 5.2 HYDROGEN PRODUCTION

The costs of hydrogen production are presented in Figure 5, assuming the same economic parameters as for electricity. Natural gas prices for small industrial users (MR) are estimated at 6.3 €/GJ. Electricity price projections for 2020 of 0.06 and 0.12 €/kwh for large and small industrial users, respectively, are used (both for selling and buying) [104].

Costs of hydrogen production with CO<sub>2</sub> capture by means of state-of-the-art CG are lower than for SMR (7 versus 9.5 €/GJ<sub>H<sub>2</sub></sub>). The low costs for CG can be explained by the relatively small additional investment costs and low energy penalty and the rather optimistic assumptions in gasification, power generation, PSA and overall integration of the CG plant. For a more conservative configuration, in which electricity is imported [14], COH are significantly higher (9 €/GJ).

In the longer term, advanced ATR and CG systems might produce hydrogen with strongly reduced CO<sub>2</sub> emissions at circa 8.1 and 6.4 €/GJ, respectively. COH for small-scale MR are much higher than for large-scale SMR, although the former include costs related to hydrogen compression to 470 bar. The costs associated with additional hydrogen infrastructure (buffer storage, pipeline, compressor etc.), which are significant and should therefore be included for a fair comparison, are considered in Chapter 3. Provided that the same tariffs for natural gas and electricity could be negotiated as large-scale users, COH might drop below 13 €/GJ. Figure 6 illustrates that the amount of CO<sub>2</sub> emitted for the considered SMR plant is slightly higher than for the CG plant. This can be explained by the lower capture efficiency at SMR as CO<sub>2</sub> produced in the furnace

of the SMR is emitted and the emissions avoided by electricity export at CG (which replace CO<sub>2</sub> emissions generated by central power production). The capture efficiency of the advanced CG plant is equal to the state-of-the-art CG plant, but electricity is imported, causing additional CO<sub>2</sub> emissions. The CO<sub>2</sub> emission rate is largest for the small-scale MR, which is designed to recover only 70% of the produced CO<sub>2</sub>, and additionally, electricity is imported.

The relatively small incremental COH caused by CO<sub>2</sub> capture expresses itself in relatively low CO<sub>2</sub> avoidance costs (see Figure 7) in comparison to CO<sub>2</sub> capture at electricity production, which is explained by the fact that syngas needs to be shifted anyhow. CO<sub>2</sub> avoidance costs for CG plants are very low, since CO<sub>2</sub> is generally also separated from the shifted syngas in configurations without CO<sub>2</sub> capture. The additional investment requirements are a CO<sub>2</sub> compressor only. The ATR system with ITM would make CO<sub>2</sub> capture from hydrogen production more attractive in comparison to traditional SMR, due to the low (additional) investment costs and higher CO<sub>2</sub> recovery.

## 6 DISCUSSION AND CONCLUSION

### 6.1 ELECTRICITY PRODUCTION WITH CO<sub>2</sub> CAPTURE

Electricity production costs of NGCC with CO<sub>2</sub> capture are generally lower than for PC/IGCC with CO<sub>2</sub> capture, but CO<sub>2</sub> avoidance costs for NGCC are higher than for PC/IGCC. This outcome is confirmed by most other studies comparing PC, NGCC and IGCC, with the exception of an IEA GHG study [20], in which avoidance costs are higher for PC. Incremental COE and CO<sub>2</sub> avoidance costs at PC (and NGCC) presented in this study are relatively low in comparison to other studies, as we apply data representing improved chemical absorption technology with significantly lower energy consumption and costs in comparison to current generation MEA plants considered in most other studies. IGCC offers a more efficient and cost-effective possibility for CO<sub>2</sub> capture in comparison to PC (i.e. COE and CO<sub>2</sub> avoidance costs are lower). It should be noted that uncertainty in costs and efficiency is larger for IGCC in comparison to PC. For the former we have a limited data set for first-of-a-kind plants, whereas real costs are available for PC plants (without CO<sub>2</sub> capture).

It is unclear whether oxyfuel combustion is more cost-effective than post-combustion capture at PC plants. On a somewhat longer term, ITM technology could be deployed for oxygen production instead of the conventional ASU. This could make a difference, since the ASU is very energy-intensive and represents circa 20% of total investment costs [31]. Both PC with post-combustion capture and IGCC with pre-combustion capture still have significant improvement potential. IGCC maintains its position as low-cost CO<sub>2</sub> capture and electricity production technology in comparison to PC in the longer term. Electricity production costs in the same order as for current PC without CO<sub>2</sub> capture (circa 4.6 €/kWh)

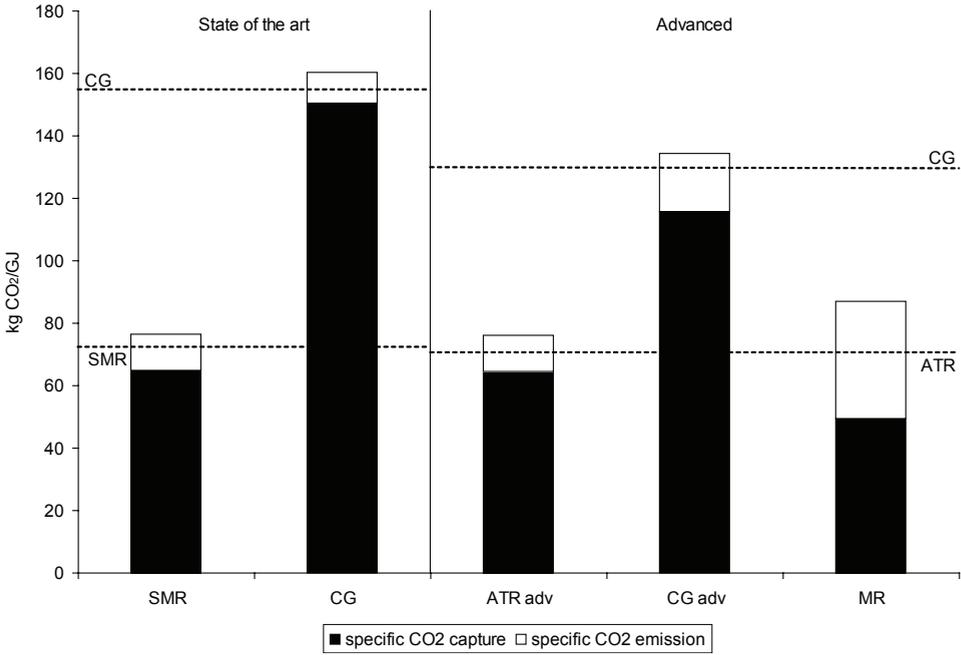


FIGURE 6. Specific CO<sub>2</sub> capture and emission rates of hydrogen production. The dashed horizontal lines represent emissions of SMR/ATR and CG without CO<sub>2</sub> capture

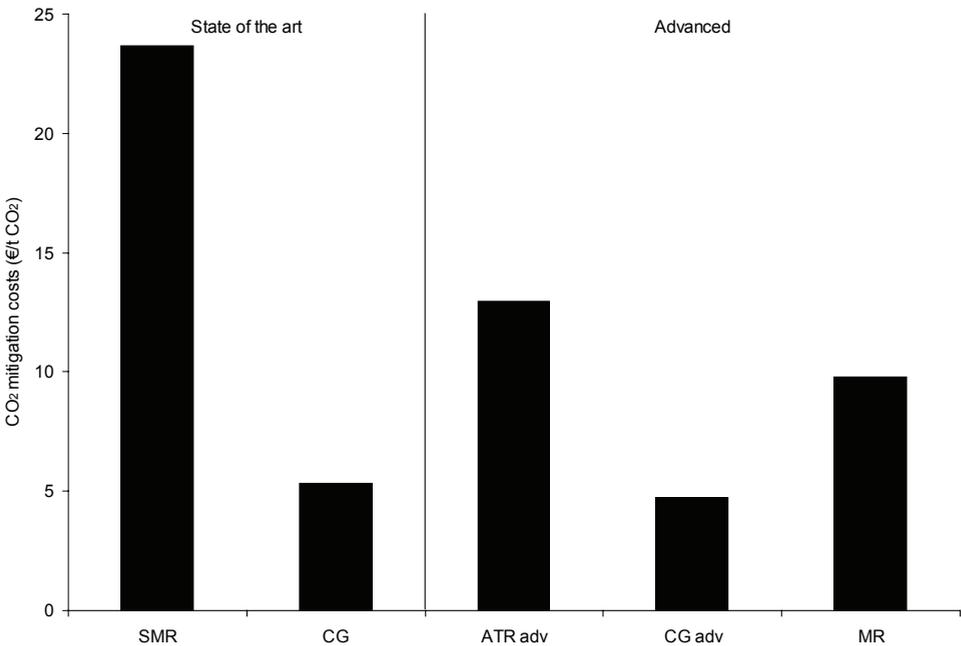


FIGURE 7. CO<sub>2</sub> avoidance costs for hydrogen production (reference: identical technology without CO<sub>2</sub> capture)

may be achievable, provided that a net electric efficiency over 43% and investment costs of 1500 €/kw prove realistic. COE of the Water cycle fuelled with syngas and IG-SOFC-GT are comparable to IGCC.

However, the technological status of these technologies is less mature. Significant R&D efforts are needed to develop and commercialise fuel cells, and turbines suited for conditions in Water cycles.

Advanced natural gas-fired options such as MR-CC, AZEP, CLC and SOFC-GT promise power generation with high efficiencies and 100% CO<sub>2</sub> capture. However, material restrictions (membranes/oxygen particles) in these concepts limit the reactor/combustion chamber temperature to a level lower than TIT of state-of-the-art gas turbines. This can be resolved by firing additional natural gas to increase TIT, thereby reducing CO<sub>2</sub> capture efficiency. The trade-off between costs and CO<sub>2</sub> capture efficiency requires further attention.

The economic performance of these advanced power production technologies is still uncertain. This is mainly due to the uncertainties pertaining to investment and O&M costs and lack of detailed cost-engineering studies for certain concepts (such as AZEP and CLC). The technologies considered contain specific elements (membranes, fuel cells, adapted turbines) for which little data are available on future commercial cost levels when integrated in a power cycle. Possibly, different/cheaper materials might be applied in future designs, which might have significant impact on performance and costs.

A recently accomplished techno-economic study evaluating amongst others advanced post and pre-combustion CO<sub>2</sub> capture at NGCC plants shows that CO<sub>2</sub> avoidance costs of MR-CC are slightly lower than NGCC with a highly-integrated post-combustion capture unit [42]. However, this study did not account for efficiency improvements and cost reductions in NGCC technology itself; only improvements in capture technology and integration were studied. An important question remains which capture strategy will benefit most strongly from autonomous developments in NGCC technology and to what extent improved turbines can be integrated in advanced power cycles with CO<sub>2</sub> capture. Therefore, more detailed techno-economic analyses of advanced natural gas-fired electricity options are required, especially for AZEP and CLC. In such analyses, efficiency and cost improvement potential in state-of-the-art and advanced conversion and capture technology should be considered. The impact of technological learning should be accounted for in cost assessments, which has not been done so far for this kind of technologies.

## 6.2 HYDROGEN PRODUCTION WITH CO<sub>2</sub> CAPTURE

CO<sub>2</sub> capture at hydrogen plants causes a relatively small increase in production costs in comparison to electricity plants, which makes hydrogen production more attractive for CO<sub>2</sub> capture. Especially coal gasification enables CO<sub>2</sub> capture with marginal additional investment costs and efficiency losses. Hydrogen production costs (with CO<sub>2</sub> capture) can be reduced further by application of ITM in advanced ATR

and CG systems. If ITM membranes prove to operate under commercial conditions, ATR would be the preferable way to produce hydrogen from natural gas, as it allows for higher recovery and less costly CO<sub>2</sub> capture. On a small scale, MR enables hydrogen production with low additional costs for CO<sub>2</sub> capture. However, significant R&D is still required to enable (mass) production of thin and stable membranes with sufficient lifetime. There are other advanced, potential low-cost hydrogen production schemes that are not further considered here mainly due to a lack of data. In sorption enhanced reforming (SER), reforming is conducted in the presence of an adsorbent that removes CO<sub>2</sub>, thereby shifting the reaction to the product site and allowing the reaction to occur at low temperatures (400-500°C) [105]. Small-scale laboratory tests demonstrated the principle. The HyGenSys process integrates a novel reformer concept with a jet engine heat generator to supply heat for the reforming reaction [106]. Plasma reforming involves natural gas heating by means of an electric arc to a temperature that a plasma state is reached, allowing the reaction to occur without catalysts [107]. This technology is still in experimental phase (in the kW range). Alternatively, natural gas can be cracked into carbon and hydrogen by means of the plasma cracking process, thereby avoiding CO<sub>2</sub> production (and hence separation). Several process variants are being developed and tested [108]. The HyPr-RING process integrates a novel gasification technology to produce hydrogen from coal with CO<sub>2</sub> capture using CaO in a single high-pressure reactor [109].

### 6.3 THE IMPACT OF FUEL PRICES

Energy production costs are strongly correlated to fuel prices, which have fluctuated significantly in the past (especially the price of natural gas, which is coupled to the oil price in the Netherlands) and are likely to increase in the future. Coal prices have been traditionally stable after 1990 at a level below 2 €/GJ, although prices exceeded 3 €/GJ in the 80'sies. Natural gas prices for large industrial users were stable at circa 3 €/GJ in the 90'sies, but increased to a level over 5 €/GJ around 2000 [110]. To account for the impact of uncertain fuel prices, we assess COE and COH for coal prices between 1 and 3 €/GJ and natural gas prices between 3 and 6 €/GJ (see Table 9). Capacity factors of NGCC plants may decrease as gas prices increase strongly, putting PC and IGCC (with CO<sub>2</sub> capture) in a more favourable perspective, as has been assessed by Rubin et al. [9]. However, in this analysis, we compare power generation on an equal load base, as the assessment of capacity factors for various technologies requires a dynamic model for the electricity sector to assess plant dispatch as a function of fuel/CO<sub>2</sub> prices and load curves.

TABLE 9. Impact of fuel prices on electricity and hydrogen production costs

Fuel price (€/GJ)	1	2	3	4	5	6
<b>COE (€/kwh)</b>						
PC	5.7	6.7	7.8	-	-	-
IGCC	5.0	6.2	7.3	-	-	-
NGCC	-	-	4.2	5.0	5.8	6.5
Adv. PC	4.4	5.3	6.2	-	-	-
Adv. IGCC	4.1	4.9	5.7	-	-	-
IG-Water	3.9	4.7	5.6	-	-	-
IG-SOFC-GT	4.2	4.9	5.6	-	-	-
Adv. NGCC	-	-	3.4	4.0	4.7	5.4
MR-CC	-	-	4.0	4.7	5.4	6.1
CLC	-	-	4.0	4.7	5.4	6.1
AZEP	-	-	4.1	4.8	5.5	6.3
SOFC-GT	-	-	4.8	5.4	6.0	6.6
<b>COH (€/GJ<sub>H<sub>2</sub></sub>)</b>						
SMR	-	-	7.2	8.6	9.9	11.3
CG	5.7	7.5	9.2			
Adv. ATR	-	-	6.0	7.2	8.5	9.8
Adv. CG	5.4	6.8	8.1			
MR (2 MW <sub>H<sub>2</sub></sub> )	-	-	12.7	13.9	15.2	16.5

The table indicates that the ranking among coal and natural-gas fired options does not change significantly with changing fuel prices, as the impact of fuel price on coal-fired options is rather limited and conversion efficiencies of the advanced gas-fired options are quite close. The break-even points of state-of-the-art NGCC with CO<sub>2</sub> capture versus PC/IGCC with CO<sub>2</sub> capture lie at a natural gas price of 5.8 €/GJ and 5 €/GJ, respectively (at a constant coal price of 1.7 €/GJ and equal capacity factors<sup>25</sup>). This would imply that CO<sub>2</sub> capture at IGCC might become competitive with the current trend of high gas prices, provided that capital cost as assumed in this study prove realistic.

Just as for electricity production, coal-based hydrogen production seems more competitive with current fuel prices and is likely to remain more competitive in the future, which is confirmed in a study performed by Williams [111].

Within the range of fuel prices studied, CO<sub>2</sub> mitigation costs of PC (and IGCC) with CO<sub>2</sub> capture remain lower than NGCC with CO<sub>2</sub> capture. Based on the results of an electricity market dispatch model, Johnson and Keith [112] conclude that CCS technologies entering the market as a consequence of high CO<sub>2</sub> prices, are characterised by higher capacity factors in comparison to base-case technologies. As a consequence, the mitigation costs of CCS technologies decreases.

25. Based on varying operating costs of technologies with and without capture as a function of fuel prices and a relation between actual capacity factors and fuel prices, Rubin et al. [9] recalculated COE. Assuming a capacity factor of 50% and 75% for NGCC and PC with CO<sub>2</sub> capture, respectively, and coal/gas price of 1.2 and 4 \$/GJ, COE of PC and NGCC with CO<sub>2</sub> capture become very close.

## 6.4 OVERALL CONCLUSION

In this study, an extensive literature review has been performed to make a comparison of coal and natural gas-fired electricity and hydrogen production technologies with CO<sub>2</sub> capture. We considered both state-of-the-art and advanced technologies and accounted for developments in existing technologies. Although the focus was on large-scale facilities, also decentralised electricity and hydrogen production technologies that enable low-cost CO<sub>2</sub> capture were considered. Energetic and economic data derived from literature were standardised and selected on a number of criteria to get figures on conversion efficiency, energy production costs and CO<sub>2</sub> avoidance costs, needed to compare the technologies in a consistent matter.

In the short term (around 2010), (ultra)supercritical PC and NGCC plants equipped with chemical absorption units and IGCC plants with a shift reactor and a physical absorption unit are the most likely candidates for electricity production with strongly reduced CO<sub>2</sub> emission (i.e. 85-90% capture efficiency). Net electric efficiencies are 30-35% for PC, 32-40% for IGCC and 43-50% for NGCC. Investment costs of 600 MWe units are 1700-2500 €/kwe for PC, 1500-2200 €/kwe for IGCC and 700-1100 €/kwe for NGCC. Calculated electricity production costs are 5.4-6.9 €/ct/kwh for PC, 4.7-6.3 €/ct/kwh for IGCC and 4.9-6.3 €/ct/kwh for NGCC, assuming a discount rate of 10%, 20 years lifetime, capacity factor of 85%, coal and natural gas prices of 1.7 and 4.7 €/GJ. CO<sub>2</sub> capture results in the lowest COE when applied to NGCC, although CO<sub>2</sub> capture at IGCC and PC could become competitive with increasingly high natural gas prices foreseen for the future. IGCC has significant lower CO<sub>2</sub> avoidance costs versus an identical plant without capture than NGCC and PC (14 €/t CO<sub>2</sub> for IGCC versus 46 €/t CO<sub>2</sub> for NGCC and 27 €/t CO<sub>2</sub> for PC).

In the longer term (2030), both new power cycles with integrated CO<sub>2</sub> capture and significant improvements in existing technology are foreseen. Advanced PC with improved post-combustion CO<sub>2</sub> capture might achieve a net electric efficiency of 40% and investment costs of 1500 €/kwe. Advanced concepts based on gasification might reach net efficiencies of 43% for IGCC (with ITM) up to 50% for IG-SOFC-GT. Investment costs are estimated between 1500 €/kwe for IGCC and 1750 €/kwe for IG-SOFC-GT. COE of advanced coal-fired power cycles, assuming identical economic assumptions as for the short term, are estimated at 4.5-4.7 €/ct/kwh for concepts deploying gasification and 5.1 €/ct/kwh for PC. CO<sub>2</sub> avoidance costs might be reduced to a level of 7 €/t CO<sub>2</sub> (for IGCC) up to 21 €/t CO<sub>2</sub> (for PC). These numbers clearly indicate that the advanced options based on coal gasification promise more efficient and cost-effective CO<sub>2</sub> capture than improved PC with post-combustion capture, despite the improvement potential for chemical absorption (and PC technology).

Advanced natural gas-fired combined cycle systems with post, pre (MR-CC) and oxyfuel (AZEP, CLC) combustion strategies may reach net electric efficiencies between 50 and 55% for CO<sub>2</sub> capture efficiencies of 85-100%. Estimated investment costs vary between 650 and 900 €/kwe,

resulting in COE between 4.5 and 5.3 €/kwh and CO<sub>2</sub> avoidance costs in the range of 25-40 €/t CO<sub>2</sub>. Decentralised SOFC-GT hybrids (20 MW<sub>e</sub>) enable power production with CO<sub>2</sub> capture at a net electric efficiency close to 60% and investment costs of 1500 €/kW<sub>e</sub>, resulting in COE of 5.8 €/kwh and CO<sub>2</sub> avoidance costs of 8 €/t CO<sub>2</sub> versus a decentralised NGCC as baseline.

In contrast to the coal-fired cases, for which pre-combustion strategies seem more promising than post-combustion strategies and oxyfuel combustion with cryogenic air separation, no winning capture strategy among advanced natural gas-fired electricity production technologies can be appointed at this stage. Mainly the uncertainty in investment costs makes it impossible to make a clear ranking of these options. It is important to realise, though, that existing technology, i.e. NGCC with post-combustion capture, still has considerable development potential and may remain competitive with advanced pre-combustion and oxyfuel combustion concepts in the coming decade. Various studies comparing advanced power cycles with state-of-the-art power cycles with post-combustion capture pass by the fact that conventional gas turbine and amine scrubbing technology will further improve.

With current technologies, hydrogen can be produced by means of large-scale SMR and CG with CO<sub>2</sub> capture from the shifted syngas, resulting in a CO<sub>2</sub> capture efficiency of 85-90%. The conversion efficiency, including electricity in/outputs in primary terms, is 73% for SMR and 59-62% for CG. Investment costs are circa 550 and 840 €/kW<sub>H<sub>2</sub></sub> for 1000 MW<sub>H<sub>2</sub></sub> SMR and CG plants, respectively. Costs of hydrogen produced by SMR are dominated by fuel and feed costs, which puts CG in more favourable perspective considering increasingly high gas prices. Hydrogen production costs for SMR are estimated at 9.5 €/GJ and an optimal designed CG plant with electricity export may reach 7 €/GJ. CO<sub>2</sub> avoidance costs versus identical plants without CO<sub>2</sub> capture are 23 and 5 €/t CO<sub>2</sub> for SMR and CG, respectively.

Advanced large-scale ATR and CG systems with ITM enable hydrogen production with 90% CO<sub>2</sub> capture at an efficiency of 73% for ATR and 69% for CG. Investment costs of these systems are estimated at nearly 300 and 600 €/kW<sub>H<sub>2</sub></sub>, resulting in COH of 8.1 and 6.4 €/GJ and CO<sub>2</sub> avoidance costs of 13 and 5 €/t CO<sub>2</sub> for ATR and CG, respectively. MR enables small-scale hydrogen production with relatively low-cost CO<sub>2</sub> capture. A 2 MW<sub>H<sub>2</sub></sub> plant may reach efficiencies of 65% and investment costs around 600 €/kW<sub>H<sub>2</sub></sub> (including hydrogen compression to 480 bar), resulting in COH of nearly 17 €/GJ, considering gas and electricity prices for small industrial users.

This study is useful in the sense that it gives a complete overview of electricity and hydrogen production technologies with CO<sub>2</sub> capture and standardised ranges in energetic and economic performance. Where possible, the causes of the variation in conversion efficiency and costs, which can be substantial for certain technologies, are explained on the

basis of specific technological or economic assumptions, which enables us to choose data according to what we consider most representative. By considering advanced technologies and improvements in current technologies, we also give insight into efficiency improvements and cost reductions that might be achievable in the longer term and technological barriers that have to be surmounted. However, the standardisation procedure has its limits. In spite of our endeavours to standardise cost and performance data, a certain variability remains due to differences in capital and labour costs, technical performance of individual components, fuel specifications, cooling water temperature etc. between the studies considered. Capital costs may vary strongly due to assumptions with respect to different cost components. Another uncertainty is what costs do refer to exactly; they can represent a plant built today, a first-of-a-kind plant built in the future or a mature advanced technology (including technological learning). Uncertainty obviously is highest in advanced concepts (e.g. CLC and AZEP). The conversion of different currencies (mainly dollars to euros) by means of GDP deflators and annual currency exchange rates introduces uncertainty as well. All these factors limit the use of the results to indicative figures for what may be achievable.

In order to compare CO<sub>2</sub> mitigation in the electricity sector versus hydrogen production and deployment, we need to consider real avoidance costs, which are directly related to the reference system. An IGCC equipped with a CO<sub>2</sub> capture unit does not necessarily replace an IGCC plant without CO<sub>2</sub> capture, as this choice is also a function of policies and market choices. 'CO<sub>2</sub> neutral' hydrogen should be compared to the fuel it substitutes, which can be either gasoline or diesel in the transport sector, natural gas in households and industry or hydrogen produced without CO<sub>2</sub> capture applied currently in the industry. In Chapter 3, we will assess the technologies considered here in a chain analysis, including the complete chain of CO<sub>2</sub> transport and storage and energy carrier distribution and end-use. Such analysis enables a comparison of CCS technologies in their complete context and also allows us to assess in which sector CO<sub>2</sub> capture proves to be most efficient, both from energetic and economic perspective.

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### CHAPTER III

## A COMPARISON OF ELECTRICITY AND HYDROGEN PRODUCTION SYSTEMS WITH CO<sub>2</sub> CAPTURE AND STORAGE

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### PART B: CHAIN ANALYSIS OF PROMISING CCS OPTIONS\*

#### ABSTRACT

Promising electricity and hydrogen production chains with CO<sub>2</sub> capture, transport and storage (CCS) and energy carrier transmission, distribution and end-use are analysed to assess (avoided) CO<sub>2</sub> emissions, energy production costs and CO<sub>2</sub> mitigation costs. For electricity chains, the performance is dominated by the impact of CO<sub>2</sub> capture, increasing electricity production costs with 10-40% up to 4.5-6.5 €/kWh. CO<sub>2</sub> transport and storage in depleted gas fields or aquifers typically add another 0.1-1 €/kWh for transport distances between 0 and 200 km. The impact of CCS on hydrogen costs is small. Production and supply costs range from circa 8 €/GJ for the minimal infrastructure variant in which hydrogen is delivered to CHP units, up to 20 €/GJ for supply to households. Hydrogen costs for the transport sector are between 14 and 16 €/GJ for advanced large-scale coal gasification units and reformers, and over 20 €/GJ for decentralised membrane reformers. Although the CO<sub>2</sub> price required to induce CCS in hydrogen production is low in comparison to most electricity production options, electricity production with CCS generally deserves preference as CO<sub>2</sub> mitigation option. Replacing natural gas or gasoline for hydrogen produced with CCS results in mitigation costs over 100 €/t CO<sub>2</sub>, whereas CO<sub>2</sub> in the power sector could be reduced for costs below 60 €/t CO<sub>2</sub> avoided.

## I INTRODUCTION

In recent years, carbon dioxide capture and storage (ccs) has received much attention for its potential to achieve major CO<sub>2</sub> reductions in a carbon constrained world. In Chapter 2, we made an inventory and techno-economic comparison of electricity and hydrogen production technologies with CO<sub>2</sub> capture to identify promising options [1]. The system boundary of this analysis was set at the production plant, i.e. only energy conversion, CO<sub>2</sub> capture and compression were considered. The complete ccs chain also encompasses fuel extraction and transport, CO<sub>2</sub> transport and storage and energy carrier transmission, distribution and end-use, causing additional CO<sub>2</sub> emissions and costs and thereby affecting the overall chain performance. As the technologies studied in [1] differ in fuel type, the amount of CO<sub>2</sub> captured and in scale, the impact of the chain elements outside the plant boundary may affect the performance of the technologies differently (see Figure 1). For this reason, the complete chain has to be considered to compare (bituminous) coal with natural gas-fired options, oxyfuel with post-combustion capture options, and central with decentralised technologies. A comparison of electricity production with ccs versus hydrogen production with ccs as competing CO<sub>2</sub> reduction options cannot be performed on plant level only. It requires the assessment of CO<sub>2</sub> mitigation costs versus a reference system, such as gasoline or natural gas in case of hydrogen. Since the distribution and end-use of hydrogen is different with respect to the fuels it substitutes, these elements have to be incorporated.

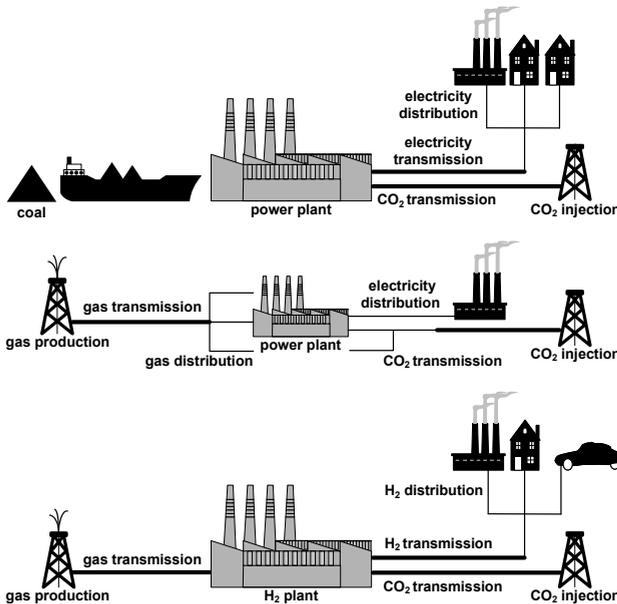


FIGURE 1. Different electricity and hydrogen production chains with ccs. (from top to bottom: central power production, decentralised power production, central hydrogen production)

Relatively few studies have been performed in which the entire chain is analysed. Most of these studies focus on large-scale combustion or gasification systems and include only a number of chain elements. ccs at decentralised units is generally considered incompatible due to economies of scale. Hendriks et al. performed a generic chain analysis for several central electricity and hydrogen chains with ccs to assess additional costs and avoided emissions versus the average park and natural gas [2]. Costs and energy use of CO<sub>2</sub> and energy transmission (excluding distribution to end-use markets) for central power and hydrogen plants have been studied in [3]. Fuel supply chains and associated costs for different hydrogen production and supply technologies have been assessed in [4]. Ogden studied large-scale hydrogen production and supply chains for the transport sector in the USA, including ccs [5]. Also various Dutch ccs chains have been analysed, in which existing electricity and industrial plants were linked to possible storage sites [6].

However, no study could be identified in which the aspects of time (short versus long term), scale (central versus decentralised units), energy transmission and distribution for different end-users and choice of the reference system are explicitly and consistently dealt with for both electricity and hydrogen systems with ccs. Therefore, in this study, promising technologies identified in Chapter 2 are further assessed in a chain analysis that accounts for these factors. Overall CO<sub>2</sub> emissions, electricity and hydrogen production costs and CO<sub>2</sub> mitigation costs of various technologies are assessed for different technological and infrastructural settings, and various reference systems. This allows us to consistently compare a wide range of ccs technologies in different contexts. By including different hydrogen end-users, the performance of hydrogen in different sectors can be compared. The chain analysis also provides insight into the economic trade-off between CO<sub>2</sub> and energy transmission. This knowledge is of importance in site selection of new plants, for the construction of new infrastructure involves large investments. Finally, the CO<sub>2</sub> emissions over the entire chain give an indication of the 'climate neutrality' of decarbonised electricity and hydrogen versus its reference as applied in [2].

The geographical focus is on the Netherlands, representing a densely populated industrialised country, where ccs would typically be deployed. ccs may play a significant role in the Netherlands in the coming decades, as this country is characterised by numerous large CO<sub>2</sub> sources and potential sinks. The Dutch CO<sub>2</sub> emission in 2003 was circa 177 mt CO<sub>2</sub>, of which approximately 100 mt emitted by the energy and manufacturing industry [7]. Power and heat production accounted for 55 mt CO<sub>2</sub>. Large point sources (>0.1 mt CO<sub>2</sub>/yr) at which CO<sub>2</sub> capture is feasible represented circa 96 mt CO<sub>2</sub> [8]. The estimated technical storage capacity of Dutch on- and offshore aquifers and gas fields exceeds 11 Gt CO<sub>2</sub> [8], implying ccs could potentially be deployed for many decades to come.

After the discussion of the main methodological issues in Section 2,

the electricity and hydrogen chains are described in Section 3. These chains are inspired by the future ‘energetic and geological map’ of the Netherlands, which gives an overview of CO<sub>2</sub> sources, hydrogen end-use markets and CO<sub>2</sub> storage reservoirs. Section 4 presents the results of the chain analysis, including a sensitivity analysis for several crucial parameters, followed by discussion and conclusions in Section 5.

## 2 CHAIN ANALYSIS

Figure 2 shows the different elements of the CCS chains. In the analysis, greenhouse gas (GHG) emissions of fossil fuel extraction, transport and distribution are accounted for. In order to capture CO<sub>2</sub>, additional fuel is required, which results in additional GHG emissions due to coal mining and natural gas extraction, as well as transport and distribution of these fuels. Electricity and hydrogen transmission and distribution is accounted for as significant energy losses and/or additional costs can occur depending on the system layout [3,5]. Hydrogen end-use is accounted for because specific hydrogen conversion technologies (fuel cells) can be fundamentally different from reference end-use technologies for conversion of hydrocarbons (turbines, boilers, internal combustion engines).

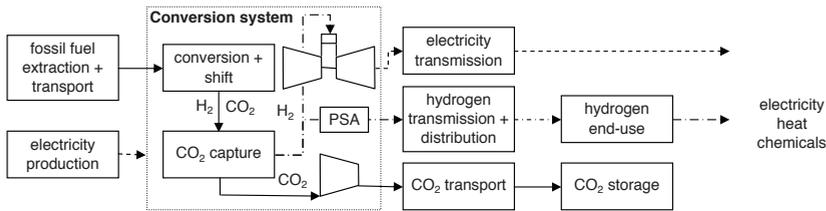


FIGURE 2. Elements in electricity and hydrogen production chains with CCS

For each chain, we set up an energy and CO<sub>2</sub> balance and calculate levelised production costs of electricity and hydrogen (COE and COH for electricity and hydrogen, respectively) and CO<sub>2</sub> mitigation costs. COE and COH are calculated by dividing the sum of annual capital and O&M costs of the conversion system and infrastructure, fuel costs and CO<sub>2</sub> storage costs by the annual energy production. Costs are converted to €<sub>2003</sub> using GDP deflators [9] and annual currency exchange rates [10]. CO<sub>2</sub> mitigation costs for electricity production with CCS are calculated using the following formula:

$$CO_2 \text{ mitigation costs} = \frac{COE_{CCS} - COE_{ref}}{m_{CO_2,ref} - m_{CO_2,CCS}} \quad \text{EQUATION 1}$$

in which COE = cost of electricity (€/kwh) and m = CO<sub>2</sub> emission factor (kg/kwh) of the CCS chain and reference chain.

The choice of the reference system has a significant impact on calculated CO<sub>2</sub> mitigation costs. In this analysis, the impact of the various reference systems will be assessed. In the most common approach as applied in Chapter 2, identical plants with and without CO<sub>2</sub> capture are compared (i.e. the baseline varies per technology). This method gives a good indication which technology inherently enables low-cost CO<sub>2</sub> capture. However, a plant with CO<sub>2</sub> capture does not necessarily replace an identical plant without capture, as the construction of a new power plant is determined by market forces and policies. In addition, the plant with low capture costs can be expensive as such. In the Netherlands, the fuel mix for power generation consists primarily of coal and natural gas, so it can be argued to consider a PC and NGCC as reference system. Hendriks et al. [2] make a distinction between a project independent approach using generic emission and cost figures, and a project specific approach, using project specific data as reference. In the former approach, the reference system could be the current Dutch power generation mix, resulting in more generalised CO<sub>2</sub> mitigation costs.

Hydrogen produced with CO<sub>2</sub> capture should be compared to the fuel it substitutes: gasoline or diesel in the transport sector, natural gas in households and industry, or hydrogen (produced without CO<sub>2</sub> capture), e.g. for chemical purposes. In the long term, hydrogen might replace cokes as reducing agent in steel production [11]. When hydrogen produced with CO<sub>2</sub> capture replaces conventionally produced hydrogen, the common formula to calculate CO<sub>2</sub> mitigation costs can be applied (Equation 1). For the cases where it substitutes gasoline, natural gas or cokes, the end-use technology should be accounted for as well (Equation 2):

$$\text{CO}_2 \text{ mitigation costs} = \frac{(COH/\eta + C + O \& M)_{H_2} - (COF/\eta + C + O \& M)_{ref}}{(m_{CO_2}/\eta)_{ref} - (m_{CO_2}/\eta)_{H_2}} \quad \text{EQUATION 2}$$

in which:

COH	= hydrogen cost (€/GJ on LHV basis)
COF	= reference fuel costs (€/GJ)
η	= end-use efficiency (functional unit/GJ)
c	= capital cost of end-use (€/functional unit)
O&M	= operating and maintenance costs (€/functional unit)
m	= CO <sub>2</sub> emission factor (kg/GJ)

A functional unit can be a kilometre, a GJ electricity/heat or a tonne of steel.

### 3 CHAIN DESCRIPTION

CCS chains, which combine specific electricity and hydrogen production technologies via specific infrastructure to specific CO<sub>2</sub> storage reservoirs and specific end-users, have both a spatial and temporal dimension. The spatial dimension encompasses the infrastructural design to connect energy extraction, conversion, and end-use markets and CO<sub>2</sub> sources with storage reservoirs. Therefore, insight into the transport costs of

primary and secondary energy carriers and CO<sub>2</sub> is required, for which quite a variation exists in literature [3,5,12-16]. For a 500 MW<sub>e</sub> NGCC, transmission costs of electricity (in €/kwh) over a certain distance are higher than for natural gas and CO<sub>2</sub> (onshore conditions) [3]. It may be more advantageous, however, to produce electricity nearby the CO<sub>2</sub> storage reservoir provided that this reservoir is closely located to the natural gas source. Such trade-offs also exist for hydrogen, natural gas and CO<sub>2</sub> transmission. It has been estimated that hydrogen will cost between 30 and 50% more to transport than an equivalent energetic quantity of natural gas [17]. Although hydrogen has a lower molecular weight and viscosity in comparison to gas, which makes hydrogen flow faster, the volumetric energy content of hydrogen is about one third of that of natural gas. The trade-off between CO<sub>2</sub> and H<sub>2</sub> transmission is amongst others depending on the fuel used to produce hydrogen and will be further studied here.

The temporal dimension is related to the timeframe considered for implementation. We distinguish chains that may be implemented on a relatively short term (2010-2015) from long-term chains (>2030), which differ in a number of aspects:

- Extent of CCS. In the short term, the construction of at most a few plants with CO<sub>2</sub> capture can be expected. In the longer term, a more significant contribution of CCS in the portfolio of CO<sub>2</sub> emission reduction options is presumed. As a guiding line, we assume 20 Mt CO<sub>2</sub> will be captured and stored annually by 2030<sup>1</sup>.
- Technologies. We consider state-of-the-art technologies for the short-term chains and more advanced technologies for the long-term chains [1]. Fuel cells for hydrogen end-use are expected to become available in the longer term. In the shorter term, hydrogen could technically be deployed for heat and power generation using boilers and turbines and for traction in internal combustion engines. However, forecasts of hydrogen demand for energy purposes in 2010-2015 appear too small to justify central hydrogen production with CCS [4,20-22].
- Storage capacity. The capacity of natural gas fields becomes gradually available in the coming decades with the depletion of these reservoirs.
- Infrastructure. In the short term, we consider dedicated CO<sub>2</sub> pipelines from power plants to storage reservoir(s). In the long term, the CO<sub>2</sub> infrastructure is likely to be expanded to a network connecting various point sources and reservoirs. Similarly, we consider a hydrogen network to connect a large plant with various end-use markets.

1. The technical potential for CCS in 2030 has been estimated at 40-60 Mt CO<sub>2</sub> [18]. A recent update studying the potential of various GHG reduction options revealed that the expected contribution of CCS in 2020 is between 0 and 15 Mt CO<sub>2</sub> avoided, depending on the emission reduction target set [19]. In order to capture and store 20 Mt CO<sub>2</sub> annually in the electricity sector, the equivalent of at least six 500 MW<sub>e</sub> state-of-the-art PC plants needs to be installed with a chemical absorption unit.

First, the electricity and hydrogen production technologies are discussed, followed by the CO<sub>2</sub> storage reservoirs and the different hydrogen end-use markets. Combining the information on CO<sub>2</sub> sources, CO<sub>2</sub> sinks and H<sub>2</sub> end-use markets then allows us to design the infrastructure required to connect these elements.

TABLE I. Economic parameters and emission factors used in this analysis

Parameter <sup>a</sup>	Value	Range
Interest rate	10%	5-15%
Capacity factor	85%	60-90%
Economic lifetime for production plants (yr)	20	15-25
<i>Energy + material costs</i>		
Steam coal (€/GJ) <sup>b</sup>	1.7	1-3
Natural gas for large industrial users (€/GJ) <sup>b</sup>	4.7	3-6
Natural gas for small industrial users (€/GJ) <sup>b</sup>	5.4	4-7
Natural gas for households (€/GJ) <sup>b</sup>	8.2	7-10
Electricity for large industrial users (€/MWh) <sup>b</sup> (= average Dutch electricity costs)	50	40-70
Electricity for small industrial users (€/MWh) <sup>b</sup>	80	70-100
Electricity for households (€/MWh) <sup>b</sup>	90	80-110
Gasoline (€/GJ) <sup>c</sup>	9	5-15
Coking coal (€/GJ) <sup>d</sup>	2.1	-
Steam (€/GJ) <sup>d</sup>	5	-
Iron ore (fine) (€/t) <sup>d</sup>	18	-
Iron ore (lump) (€/t) <sup>d</sup>	24	-
Iron ore pellets (€/t) <sup>d</sup>	36	-
Scrap (€/t) <sup>d</sup>	100	-
Electrodes (€/kg) <sup>d</sup>	2.4	-
<i>GHG emission factors <sup>e</sup></i>		
Natural gas (kg CO <sub>2</sub> -eq/GJ)	56 (57)	-
Coal (kg CO <sub>2</sub> -eq/GJ)	95 (103)	-
Gasoline (kg CO <sub>2</sub> -eq/GJ)	72 (87)	-
Average Dutch electricity 2020 (kg CO <sub>2</sub> /MWh)	450	350-500

<sup>a</sup> We consider fixed costs and emission factors to increase transparency in outcome. The uncertainty in developments in fuel costs is further accounted for in the sensitivity analysis.

<sup>b</sup> Forecasts for 2020 [23,24], including commodity, transmission and distribution costs, excluding taxes and VAT. Electricity prices are used for selling and buying.

<sup>c</sup> Rotterdam gasoline price (plus distribution costs, excluding taxes and VAT) in 2003/2004, during which oil prices were around 30-35 \$/bbl [25].

<sup>d</sup> [26].

<sup>e</sup> Values in parentheses include GHG emissions of fossil fuel extraction, transport and distribution (and refining in the case of gasoline) [2,27]. Note that indirect GHG emissions of coal and natural gas use may change in the longer term as a consequence of improved mining and transport practices, longer transport distances and switch to LNG. The average emission factor of Dutch electricity production is based on a scenario forecast given in [23].

### 3.1 CO<sub>2</sub> SOURCES: ELECTRICITY AND HYDROGEN PRODUCTION TECHNOLOGIES

Key techno-economic characteristics of the technologies have been discussed in Chapter 2 [1] and are summarised in Tables 2 and 3.

Decentralised electricity and hydrogen production is generally not considered due to the relatively high costs of CCS at such scales.

Advanced concepts using fuel cells and membranes, however, offer the potential for low-cost CO<sub>2</sub> capture at relatively small scales [28].

Decentralised electricity production with CCS is of particular interest for the Netherlands given the large share of decentralised CHP units (circa 5 GW<sub>e</sub> of total installed capacity near 20 GW<sub>e</sub> in 2003) [29]. Small-scale H<sub>2</sub> production with CCS might play a role in the transition towards a hydrogen economy.

TABLE 2. Key parameters of electricity production technologies with CO<sub>2</sub> capture and compression to 110 bar

Feedstock	Conversion technology	CO <sub>2</sub> capture technology	Net electric efficiency	CO <sub>2</sub> capture efficiency	TCR (€/kW <sub>e</sub> )	O&M
<i>Reference technologies (600 MW<sub>e</sub>)</i>						
Bituminous coal	PC	-	44%	-	1500	5.2%
Natural gas	NGCC	-	56%	-	540	3.7%
<i>State-of-the-art technologies (600 MW<sub>e</sub>)</i>						
Bituminous coal	PC	Post-comb (MEA)	35%	88%	2080	5.8%
	IGCC	Pre-comb (Selexol)	32-35%	85%	1770-2170	4.8-5.2%
Natural gas	NGCC	Post-comb (MEA)	47%	85%	920	4.3%
<i>Advanced technologies (600 MW<sub>e</sub> except SOFC-GT)</i>						
Bituminous coal	Advanced PC	Improved post-comb (MEA)	40%	85%	1520	6.5%
	Advanced IGCC	Pre-comb (Selexol)	43%	85%	1500	5%
	IG-Water	Oxyfuel (ASU)	41%	100%	1530	3.7%
	IG-SOFC-GT	Various (membrane/cat. combustor)	50%	90%	1760	3.3%
Natural gas	Advanced NGCC	Improved post-comb (MEA)	55%	85%	650	4.8%
	MR-CC	Pre-comb (HMCM)	53%	100%	940	4%
	CLC	Oxyfuel (separate combustion)	51%	100%	900	4%
	AZEP	Oxyfuel (oxygen membrane)	50%	100%	900	4%
	SOFC-GT (20 MW <sub>e</sub> )	Oxyfuel (afterburner)	59%	80%	1530	3%

TABLE 3. Key parameters of hydrogen production technologies with CO<sub>2</sub> capture and compression to 110 bar

Conversion technology	CO <sub>2</sub> capture technology	Fuel+feed input (GJ/GJ <sub>H<sub>2</sub></sub> )	Electricity input (GJ <sub>e</sub> /GJ <sub>H<sub>2</sub></sub> )	Conversion efficiency	CO <sub>2</sub> capture efficiency	TCR (€/kW <sub>H<sub>2</sub></sub> )	O&M
Advanced ATR (1000 MW <sub>H<sub>2</sub></sub> ) <sup>a</sup>	MDEA	1.28	0.03	74%	90%	280	4%
Advanced CG (1000 MW <sub>H<sub>2</sub></sub> ) <sup>a</sup>	Selexol	1.35	0.05	69%	90%	600	4%
MR (2 MW <sub>H<sub>2</sub></sub> ) <sup>b</sup>	Pd membrane	1.26	0.13	65%	70%	610	9%

<sup>a</sup> Including H<sub>2</sub> compression to 60 bar.

<sup>b</sup> Estimated capacity to supply a future hydrogen refuelling station, including H<sub>2</sub> compression to 480 bar [28].

### 3.2 CO<sub>2</sub> SINKS: GEOLOGICAL RESERVOIRS

2. Storage in the deep ocean is not considered. Injection depths are at least 1000 m [30], which would require transport by ship over large distances as the North Sea is not deep enough.

3. Reservoirs should preferably offer sufficient potential to store the captured CO<sub>2</sub> of one plant over its lifetime. A 500 MW<sub>e</sub> NGCC equipped with amines captures circa 1.5 Mt CO<sub>2</sub>/yr, corresponding to 30 Mt to be stored in a 20 year lifetime. For a PC plant of equal capacity, the capture rate is more than twice as large.

Reservoirs suited for geological storage of CO<sub>2</sub> can be classified into (nearly) depleted oil and gas fields, deep saline aquifers and unminable coal seams<sup>2</sup>. In the Netherlands, gas fields offer a large storage potential of circa 10 Gt CO<sub>2</sub>, of which 7.5 Gt is represented by the Groningen gas field. Most of the reservoirs provide less than 30 Mt CO<sub>2</sub> storage capacity<sup>3</sup>, although nearly 30 reservoirs provide a storage potential between 30 and 300 Mt CO<sub>2</sub> [31]. In the coming decade various small and medium-sized gas fields become available, whereas the Groningen gas field is not expected to become available before 2050 [32]. The Dutch oil fields are less interesting for CO<sub>2</sub> storage as they represent a relatively low storage potential. The interest in enhanced oil recovery (EOR) by injecting CO<sub>2</sub> into oil fields in the North Sea [33] might offer short-term opportunities for CCS. Aquifers and coal seams are not that well studied and characterised as hydrocarbon structures, which causes a relatively large uncertainty in the storage potential. Storage in coal seams is still in an experimental phase and needs considerable testing before it might be applied commercially. In this analysis, we will therefore focus on gas fields and aquifer traps as potential CO<sub>2</sub> storage reservoirs. There is significant variation in storage cost estimates due to differences in CO<sub>2</sub> injection rate, storage capacity, reservoir type, features (pressure, thickness, permeability and depth) and location (onshore-offshore). Relations between storage costs, injection rate, reservoir type and capacity are applied to generalise storage costs (see Table 4). We distinguish storage rates between 1 and 2 Mt/yr (typical capture rate for 600 MW<sub>e</sub> NGCC or 1000 MW<sub>H<sub>2</sub></sub> SMR) and between 2 and 4 Mt/yr (typical capture rate 600 MW<sub>e</sub> PC/IGCC or 1000 MW<sub>H<sub>2</sub></sub> CG). In some specific source-sink combinations, more than one trap may be required to store all CO<sub>2</sub> in a 20-year lifetime. Storage costs may increase with a factor 2 when three gas fields are required instead of one to store 1 Mt CO<sub>2</sub>/yr [6].

TABLE 4. CO<sub>2</sub> storage costs as derived from [6]

Reservoir type	Depth (m)	Storage rate (Mt/year)	Storage cost (€/t CO <sub>2</sub> )
Aquifer onshore	1000-2500	1-2	3
		2-4	2
Aquifer offshore	1500-2500	1-2	8
		2-4	5
Gas field onshore	2500-3500	1-2	3
		2-4	2
Gas field offshore	3000-4000	1-2	10
		2-4	6

### 3.3 HYDROGEN END-USE MARKETS

In order to set up a basic design of hydrogen infrastructure, we need an indication of potentials, demand profiles and spatial distribution of future hydrogen end-use markets. In this analysis, the transport, residential and industrial sector are considered (see Textbox 1 for details).

In the transport sector, hydrogen can be used as an alternative for

gasoline and diesel in conventional vehicles with internal combustion engines (ICEV) and fuel cell vehicles (FCV). Various well-to-wheel analyses indicate that use in ICEV has no advantages from an energetic point of view [27,34], so use in ICEV is not further considered. Although FCV prototypes have been introduced into the market, significant market penetration is not expected before 2020 [4,20-22].

In the residential sector, hydrogen could gradually take over the function of natural gas and could simultaneously reduce electricity demand from the grid. It could technically be deployed in modified boilers. In the longer term, proton exchange membrane fuel cells (PEMFC) enable electricity and heat production on the level of a house/building (block) at a somewhat longer term. Hydrogen application is primarily foreseen for new build projects where an entire new infrastructure needs to be constructed<sup>4</sup>. In the short term, hydrogen demand in the residential sector is likely to be very limited [20,22].

The manufacturing industry is another interesting sector to realise large CO<sub>2</sub> emission reductions by means of CCS due to the large amount of energy consumed and its clustered nature. Hydrogen could play an important role in the decarbonisation of industrial energy use.

Nowadays, hydrogen is produced and consumed in industry on a large scale, mainly for fuel upgrading and desulphurisation at refineries, and ammonia production. In the future, the hydrogen market can be further extended by replacing fossil fuels applied in industrial CHP units, boilers and heaters<sup>5</sup>. At refineries, hydrogen production using coal or natural gas for combustion purposes is not the most obvious option, as refinery gas and heavy oil residues are often available. However, there are industries where no such energy carriers are available and natural gas is purchased for electricity and heat production. This might create some opportunities for decentralised CHP units fired with hydrogen.

A recent study on decarbonisation strategies for a number of remotely located distributed gas turbines concluded that it would be more cost-effective to produce hydrogen centrally and distribute it to each turbine than scrubbing CO<sub>2</sub> from the collected flue gasses [39].

4. The question is whether pure hydrogen can be distributed through the local natural gas grid to existing households, considering the lower energy density and material issues. Much of the distribution grid consists of polyethylene, which might be susceptible to hydrogen leakage [35].

5. Hydrogen can generally not be combusted in existing boilers and turbo-machinery. However, industrial burners using hydrogen are available [36]. Gas turbines need to be retrofitted to enable combustion of hydrogen-rich fuels [37]. In our analysis, we assume the thermal efficiency of hydrogen combustion is unchanged, although there could be improvements in efficiency [38].

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**TEXTBOX 1. Estimating hydrogen demand for the Netherlands**

In estimating hydrogen demand, we focus on the Randstad, a large urban conglomeration in the Midwestern part of the Netherlands, where major cities (accommodating over 7 million people) and industries are located.

*Transport sector*

In 2003, the fuel consumption of road transportation in the Netherlands approximated 460 PJ, causing a CO<sub>2</sub> emission of 34 Mt [7]. Projections indicate that these emissions may reach circa 45 Mt by 2020 [23]. These emissions, together with local air pollutants, can be reduced by means of hydrogen. The hydrogen demand for the Randstad area in 2030 is estimated as a function of the number of FCVs (determined by penetration level, new car sales and average car lifetime), FCV fuel economy and travel distance. Considering a fleet of 4 to 5 million light duty vehicles and assuming 5-25% of the entire light duty vehicle fleet consists of FCVs in 2030 based on penetration scenarios presented in [4,20-22], between 0.2 and 1.25 million FCVs will be on the road by that time. Given the range in fuel economy as specified in Table 5, the total hydrogen demand for the Randstad area in 2030 varies between 3 and 40 PJ/yr. We consider an average scenario assuming 4.5 million cars on the road, a FCV fraction of 20% and a fuel economy of 0.92 MJ/km, which would require an annual hydrogen supply of 13.3 PJ. The size of the individual refuelling stations, ranging from 0.1 to 8 MW<sub>H<sub>2</sub></sub> [40,41], is a function of the number of FCVs served by a refuelling station, fuel economy, travel distance and the refuelling pattern. In our analysis, we consider a uniform capacity of 2 MW<sub>H<sub>2</sub></sub> (see Table 6). This would imply that circa 250 refuelling stations need to be installed to cover the demand of 13.3 PJ/yr, which corresponds to 0.05 hydrogen refuelling station per km<sup>2</sup> on average. The average gasoline refuelling station density is estimated at 0.31 per km<sup>2</sup> using national statistics on car density and number of cars served per gasoline station [42]. This implies a coverage factor of 16%.

TABLE 5. Car characteristics [4,43-45]

Parameter	FCV	ICEV
Fuel economy (MJ/km)	0.5-1.3	1-2.8
Retail costs of drive train (€/kw <sub>peak</sub> ) <sup>a</sup>	60	25
O&M costs (€/yr)	400	400
Lifetime (yr)	15	15
Annual travel distance (km)	16,000	16,000
Hydrogen stored onboard (kg)	4.76	-

<sup>a</sup> Including transmission, fuel storage tank and motor. ICEV and FCV have a capacity of 100 and 74 kw, respectively

TABLE 6. Refuelling station features [4,40,43,46]

Parameter	Value
Station capacity (kg/d)	1440
Load factor	0.85
Peak production (kg/hr)	153
Dispenser capacity (kg/hr)	73
Tank refill (%)	80
# cars served (cars/day)	320
# cars served at peak hour (cars/hr)	40
Maximum # cars per dispenser (cars/hr)	10
# dispensers	4

TABLE 7. Cost and performance data for conventional and ‘hydrogen’ households after [49]

Parameter	Value
Heat demand <sup>a</sup>	5.3 MWh/yr
Electricity demand	3.4 MWh/yr
$\eta_{th}$ 10 kW <sub>th</sub> heater (NG/H <sub>2</sub> )	95%
$\eta_e$ PEMFC	66%
$\eta_{th}$ PEMFC	34%
Costs 10 kW <sub>th</sub> heater (NG)	720 € + 2% O&M
Costs 1 kW <sub>e</sub> nominal PEMFC <sup>b</sup>	1500 € + 2.5% O&M
Costs 8.2 kW <sub>th</sub> heater (H <sub>2</sub> )	630 € + 2% O&M
Costs 7.7 MJ <sub>th</sub> heat buffer	340 € + 2% O&M

<sup>a</sup> State-of-the-art heat demand (space and water) for houses in new residential area. In the longer term, heat demand is expected to decrease further [23], but this is not accounted for in our calculations

<sup>b</sup> Complete system costs. The value used here is an average of the USDOE target of 1500 \$/kw [4], the EU target of 2000 €/kw for micro-CHP applications in 2020 [50] and a target of 1000-1500 \$/kw for large volumes quoted in [51].

### *Residential sector*

In 2003, the Dutch residential sector consumed circa 360 PJ natural gas, producing 19 Mt CO<sub>2</sub> [7], and consumed 80 PJ electricity [47]. Considering 1,475,000 new dwellings to be constructed between 2011 and 2030 [48], we assume 200,000 dwellings constructed in this period in the Randstad area will be equipped with hydrogen infrastructure. This corresponds to circa 2.5% of the entire stock, which is within the range of estimates in other scenario studies (0.5-15% [20,22]). Based on heat and electricity demand of modern households and performance figures for micro-fuel cells for co-generation [49], we estimated hydrogen demand for ‘hydrogen districts’ comprising 5000 households. We consider a configuration comprising a (heat) load following 1 kW<sub>e</sub> PEMFC per household supplemented with a hydrogen burner and a heat buffer (see Table 7). In order to cover the specified energy demand, 10.53 MWh hydrogen is required and 1 MWh electricity is exported to the grid [49]. This would imply an annual hydrogen demand of 7.6 PJ to supply 200,000 households. The service sector, consuming circa 200 PJ fuels causing a CO<sub>2</sub> emission of 11 Mt/yr [7], would also be appropriate to

deploy hydrogen, but is not further considered here due to lack of data on heat and electricity demand patterns required to design a micro-CHP system.

### *Industrial sector*

In 2003, total fuel consumption in the Dutch industrial sector was circa 430 PJ, corresponding to a CO<sub>2</sub> emission of circa 27 Mt (excluding joint-venture CHP units, refineries and feedstock use) [7]. The chemical industry is the main energy consumer, emitting 12 Mt CO<sub>2</sub>/yr. Refineries emit another 11 Mt/yr from combustion of natural gas and oil products [7]. The fuel demand in the industry is projected to increase slightly the coming two decades [23]

Of the total industrial energy use, a significant share (circa 260 PJ) is represented by natural gas consumption in CHP units, boilers and heaters [47]. In 2003, the installed CHP capacity at refineries was about 400 MWe [52], of which the majority steam and gas turbines in the range of 10-50 MWe [53]. In the chemical industry, circa 1800 MWe is installed, of which a few very large combined cycles [52]. The capacity of CHP is expected to grow the coming decades, especially at the chemical industry. We assume a part of this capacity will be fired with hydrogen: the excess of hydrogen (production 1000 MW minus demand transport and residential sector) equals nearly 6 PJ/yr, which is sufficient to fuel four 20 MWe gas turbines with a net electric efficiency of 40%.

In the longer term, hydrogen might be applied as reducing agent in the steel industry. As the steel industry is a large CO<sub>2</sub> source in the Netherlands, emitting nearly 6 Mt in 2003 [7], we explore the opportunities for steel production using coal or natural gas with CCS. Direct reduction (DR) technology enables reduction of iron oxide with hydrogen or syngas, omitting the use of cokes in the blast furnace route to produce pig iron. Table 8 gives the basic characteristics of the reference route (blast furnace + basic oxygen furnace (BF+BOF)), and various direct reduction routes (+ electric arc furnace (EAF)) with inherent CO<sub>2</sub> removal. A more detailed description of these technologies and their energy use can be found in [54-56].

TABLE 8. Main inputs and outputs of different steel production routes [26,57]

Process	BF + BOF <sup>a</sup>	Circored + EAF <sup>b</sup>	Circofer + EAF <sup>b</sup>	Coal-based Midrex + EAF <sup>b</sup>
<i>Input</i>				
Fine ore (t)	1.2	1.4	1.4	-
Lump ore (t)	0.2	-	-	0.9
Pellets (t)	-	-	-	0.6
Scrap (t)	0.1	0.1	0.1	0.1
Coking coal (t)	0.4	-	-	-
Other coal (t)	0.1	-	0.4	0.4
Natural gas (GJ)	-	11.4	0.1	0.1
Electricity (GJ <sub>e</sub> )	0 (0.77)	2.2	2.4	3.1
Electrodes	-	2.9	2.9	2.9

Process	BF + BOF <sup>a</sup>	Circored + EAF <sup>b</sup>	Circofer + EAF <sup>b</sup>	Coal-based Midrex + EAF <sup>b</sup>
<i>Output</i>				
Liquid steel (t)	1	1	1	1
Electricity (GJ) <sup>c</sup>	-0.09 (0)	-	-	-
By-product (GJ)	-0.27	-	-	-
CO <sub>2</sub> emission (t)	1.7 (0.6)	0.4	0.5	0.5
CO <sub>2</sub> capture (t)	0 (1.1)	0.6	1.0	1.0
Costs (€/t liquid steel)				
Capital costs	32.0 (37.6)	28.0	27.1	34.3
O&M	48.2 (50.2)	49.5	52.4	46.6
Labour	27.1	17.9	19.4	17.9

<sup>a</sup> Values in parentheses refer to a configuration in which BF gas is compressed, shifted and CO<sub>2</sub> is captured and compressed.

<sup>b</sup> Including CO<sub>2</sub> capture and compression to 110 bar.

<sup>c</sup> We assume the excess BF, BOF and coke oven gas is converted into electricity to cover the internal electricity demand and eventually to export electricity to the grid.

### 3.4 INFRASTRUCTURAL REQUIREMENTS AND COSTS

#### 3.4.1 ELECTRICITY TRANSMISSION AND DISTRIBUTION

Central production units are connected to the high-voltage grid transporting electricity to the regional distribution networks. Decentralised power plants can be connected directly to the local distribution network, thereby avoiding transmission losses and costs. Typical electricity losses occurring during transmission and distribution to final consumers are estimated at 8%. Only 1% of these losses are caused by transmission, the other 7% by distribution on the medium and low-voltage grid [58]. Transmission and distribution tariffs for households are 40-60 €/MWh. Transmission tariffs for very large (50 MWe) industrial users are approximately 5 €/MWh [59]<sup>6</sup>.

#### 3.4.2 CO<sub>2</sub> TRANSMISSION

For large-scale CO<sub>2</sub> transport, pipelines are generally considered to be most suitable [14,16]. Ship transport is an alternative when the storage reservoir is located offshore, as it avoids large investments of pipeline construction and it offers flexibility in CO<sub>2</sub> purchasing and delivering. The disadvantage is the high cost for liquefaction and buffer storage, which makes ships less interesting for short transport distances. The turning point of transporting 6.2 mt CO<sub>2</sub>/yr is circa 700 km<sup>7</sup>; beyond that point ship transport becomes economically more attractive than transport by pipeline [60]. Transport costs for 20,000 t/day (equivalent of capture rate of 1000 MWe PC using MEA) over a transport distance of 1000 km are circa 13 \$/t CO<sub>2</sub> [60]. Pipeline diameters are calculated using steady-state equations applied for incompressible flow under isothermal conditions, for which the pressure drop is derived from the Fanning, or Darcy, Equation [61]:

$$\Delta p = 2 f \rho v^2 \frac{L}{D} = \frac{32 f \rho L Q^2}{\pi^2 D^5} \tag{EQUATION 3}$$

6. Transmission costs for a 500 MWe NGCC have been estimated at 1-5 €/MWh for a distance of 50-500 km [3].

7. The transport distance from Rotterdam to the Gullfaks oil field, one of the reservoirs studied for EOR, is circa 1000 km.

The friction factor ( $f$ ) is a function of the Reynolds number ( $R_e$ ) and the roughness of the pipeline inside surface, according to the Moody chart. The Reynolds number is a dimensionless quantity that indicates the type of flow:

$$R_e = \frac{\rho v D}{\mu} \quad \text{EQUATION 4}$$

For laminar flows ( $R_e < 2000$ ), the friction factor is correlated to the Reynolds number according to:

$$f = \frac{16}{R_e} \quad \text{EQUATION 5}$$

A turbulent flow ( $R_e > 3000$ ) can be either fully or partially turbulent. Various equations exist to calculate the friction factor for turbulent flows, each of them being specific for a certain pipeline and flow regime. The following generic approximation of the friction factor is used, which results in relatively conservative pipeline design [61]:

$$f = \frac{0.04}{R_e^{0.16}} \quad \text{EQUATION 6}$$

$\Delta p$  = pressure drop (Pa)  
 $f$  = friction factor  
 $v$  = average fluid velocity (m/s)  
 $Q$  = volumetric flow rate ( $\text{m}^3/\text{s}$ )  
 $L$  = pipe length (m)  
 $D$  = (internal) pipeline diameter (m)  
 $\rho$  = fluid density  
 $\mu$  = fluid viscosity

Fluid properties were derived as a function of temperature and pressure [62]. Onshore pipeline temperature is set at  $10^\circ\text{C}$ , equal to the soil temperature. Offshore pipeline temperature is circa  $6^\circ\text{C}$  [16]. The pipeline diameter is calculated iteratively until the chosen diameter meets the specified maximum pressure drop, which is set at 30 bar, assuming an inlet pressure of 110 bar and no recompression occurs along the main pipeline trajectory.  $\text{CO}_2$  pipelines are operated at high pressure to guarantee high densities for optimal pipeline utilisation. In order to avoid phase transition, pressures should be kept well above the supercritical pressure (74 bar). According to Farris [63], the minimum pressure is between 83 and 89 bar. The inlet pressure is generally set to overcome the pressure drop that occurs as a result of friction. Booster stations are required when a large pressure drop occurs, e.g. at long pipelines or hilly terrain. In order to optimise pipeline pressure and diameter, the sum of pipeline and booster station capital and o&m costs plus electricity costs for pumping should be minimised. This

‘theoretical’ optimum may differ substantially as there is a large variation in capital costs of pipelines and booster stations. The minimum distance between booster stations specified in literature varies substantially, from circa 100 to 250 km [3,16,64]. Recently, a 330 km pipeline (diameter 0.3-0.36 m) has been constructed transporting circa 5000 tonnes CO<sub>2</sub>/day overland without any booster station along the pipeline. The pressure falls from 170 to 148 bar [65]. Offshore, where booster stations are very expensive, pipeline pressure and diameters are generally higher to overcome the pressure drop.

### 3.4.3 H<sub>2</sub> TRANSMISSION AND DISTRIBUTION

Large quantities of hydrogen are generally transported in gaseous form through pipelines. Liquid and gaseous hydrogen transport by means of trucks may be an alternative for remote locations or for small-scale applications [22,40]. As transition option, hydrogen could be added to the natural gas grid up to 3% without any modifications. The hydrogen content might be increased to 25 vol% (peak concentration), which requires network upgrading and replacement of some end-use equipment [66]. The magnitude of CO<sub>2</sub> abatement (versus 100% natural gas) that can be achieved by blending 25 vol% hydrogen produced by means of SMR with CCS is small (circa 4%) [66]. A current EU project studying the possibilities of hydrogen addition to the natural gas grid shows that the margin of H<sub>2</sub> addition might be much lower [67], especially in the high pressure transmission network, which is designed for peak capacity. Reconsidering these infrastructure options and the fact that this analysis studies large quantities of hydrogen to be transported over relatively small distances, transport by dedicated pipelines seems most appropriate and is projected to give the lowest cost [12,40,68]. In the longer term, part of the natural gas grid may become obsolete when the use of hydrogen takes over as gaseous energy carrier. This may open possibilities for the conversion of parts of the existing infrastructure to transmit and distribute hydrogen. In order to design a H<sub>2</sub> infrastructure, a hydrogen demand map should be set up, for which we use estimations of potential demand and possible locations of H<sub>2</sub> production plants and end-use markets. The end-use markets vary in size and required hydrogen purity and pressure. We can distinguish high-purity (>99.99%) hydrogen for application in PEMFCs from less pure, ‘fuel grade’ (<95%) hydrogen suitable for combustion purposes. In this study, we simplify matters by considering a hydrogen grid transporting high-purity hydrogen, connecting a central hydrogen plant with refuelling stations, residential areas and industrial CHP units. Hydrogen is transported from the production unit by means of a high-pressure transmission line, after which it enters a regional medium-pressure transmission ringline, similar to the natural gas transmission system. At the city gate stations (to which CHP units are connected), hydrogen is transferred to the main distribution grid, from where it continues its route to refuelling stations and dwellings. The transport inlet pressure of the main transmission

8. Typical transport pressures of hydrogen transmission are between 10 and 30 bar [12], although pipelines have been operated up to 100 bar [17,68]. We consider a value of 60 bar, which is similar to that considered in recent studies on hydrogen transmission [40,41,69]. The high-pressure natural gas transmission system in the Netherlands is operated at 50-70 bar, after which regional medium-pressure transmission pipelines supply the gas to local gas distribution station at 20-40 bar. The local distribution grid is operated at maximally 8 bar, with the gas finally reaching the consumer at a small overpressure of 250 mbar.

line is set at 60 bar, allowing a pressure drop of 20 bar<sup>8</sup>. In the regional transmission lines, pressure is between 40 and 20 bar, after which it is further decreased to 10 bar in the main distribution pipelines. At the refuelling stations, hydrogen is compressed to 480 bar for buffer storage to enable cascade-filling, assuming hydrogen is stored onboard at 350 bar in conventional pressurised tanks [43]. In the residential sector, hydrogen is delivered near atmospheric pressure.

For pipeline design, we apply steady-state equations for an isothermal compressible flow [17]:

$$Q_b = 6.65 \left( \frac{T_b}{p_b} \right) \left( \frac{1}{f} \right)^{0.5} \left( \frac{p_1^2 - p_2^2}{ZTGL} \right)^{0.5} D^{2.5} \quad \text{EQUATION 7}$$

in which:

$Q_b$	= flow rate at base (normal) conditions (Nm <sup>3</sup> /s)
$T_b$	= temperature at base (normal) conditions (293.15 K)
$p_b$	= pressure at base (normal) conditions (101,325 Pa)
$f$	= friction factor
$p_1$	= inlet pressure
$p_2$	= outlet pressure
$Z$	= compressibility factor (1)
$T$	= average gas temperature
$G$	= gas gravity (=0.0696 for H <sub>2</sub> )
$L$	= pipe length (m)
$D$	= (internal) pipeline diameter (m)

In high-pressure gas transmission lines with moderate to high flows, the flow regime is either partially or fully turbulent. Various specific flow equations are applied in the gas industry to calculate the friction factor [17]. The Panhandle A equation is applied for partially turbulent flow conditions, medium to relatively large diameter pipelines with moderate gas flow, operating under medium to high pressure. Panhandle B and AGA fully turbulent equations are appropriate for high-flow-rate, medium to large-diameter pipelines and high-pressure systems. The pipeline variation is generally below 20% using the different equations. We applied the generic Equation 6 to calculate the friction factor, resulting in average diameters in comparison to those generated using AGA fully turbulent equation (large diameter) and Panhandle A/B (smaller diameter). This generates sufficiently reliable results given the generic nature of this study, the uncertainty in pipeline costs and the difference between internal and nominal pipe size. Average velocities calculated for the specified pressure drop, assuming no booster stations are installed along the pipeline, are generally between 5 and 20 m/s, which is reasonable for gas pipelines [17]. Given the specified pressure drop, transporting large quantities over small distances results in high average velocities that might exceed the erosional velocity [17]. Therefore, larger pipelines might be required, resulting in higher costs. Note that if the pipeline diameter is doubled, the pipeline capacity is increased by a factor  $(2)^{2.5} = 5.66$ . This illustrates

TEXTBOX 2. CO<sub>2</sub> infrastructure

Figure 3 clearly illustrates the mismatch in CO<sub>2</sub> sources and gas fields in the Netherlands; the majority of the gas fields are located in the Northern part of the country and offshore, whereas the majority of CO<sub>2</sub> sources are located in the Southern and Western part of the country. The identified aquifer traps are distributed more homogeneously. Given the potential locations of electricity and hydrogen plants and reservoirs, transport distances vary between nearly 0 and 200 km. For the short term, we assume CO<sub>2</sub> is transported in dedicated pipelines and stored in onshore gas fields located at 100 km from the plant. In the long term, a network supplying CO<sub>2</sub> to various onshore and offshore gas fields and aquifers is considered. A rough screening of the Netherlands shows that there are not enough large reservoirs nearby the sources to store 20 mt/yr in total. As the majority of reservoirs in the Netherlands are clustered in the Northern provinces and offshore, a backbone seems advantageous, to which CO<sub>2</sub> sources are connected. For the costs of transporting CO<sub>2</sub> through the trunk line, a fixed tariff of 1.3 €/t CO<sub>2</sub> is used. This tariff is based on transport costs of 20 mt CO<sub>2</sub>/yr over 200 km, including costs of the booster stations [3] and generalised costs of branches to the individual storage reservoirs. In addition, we consider a fixed storage tariff of 2 €/t CO<sub>2</sub> based on CO<sub>2</sub> injection into onshore gas fields (representing the largest storage potential).



FIGURE 3. Location of geological reservoirs and large CO<sub>2</sub> sources in the Netherlands (courtesy of TNO-NITG) and possible configuration of a trunk line as part of a CO<sub>2</sub> network

TEXTBOX 3. H<sub>2</sub> infrastructure

Figure 4 illustrates a conceptual infrastructure to deliver hydrogen from a central hydrogen production unit to various markets. For the Dutch context, the main transmission line is varied between 20 and 200 km, depending on the location of the H<sub>2</sub> production facility. The length of the regional transmission line is estimated using an idealised representation of the Randstad ( $R_1 = 40$  km,  $R_2 = 20$  km,  $R_3 = 5$  km). The length of the main distribution system is estimated using an algorithm developed by researchers of UC Davis [40], assuming 250 refuelling stations are distributed over 10 conglomerations. The length of the low-pressure distribution grid to individual dwellings is estimated using statistics on the natural gas grid. The low-pressure gas distribution network has a total length of nearly 85,000 km [66], corresponding to 12 m per household, on average.

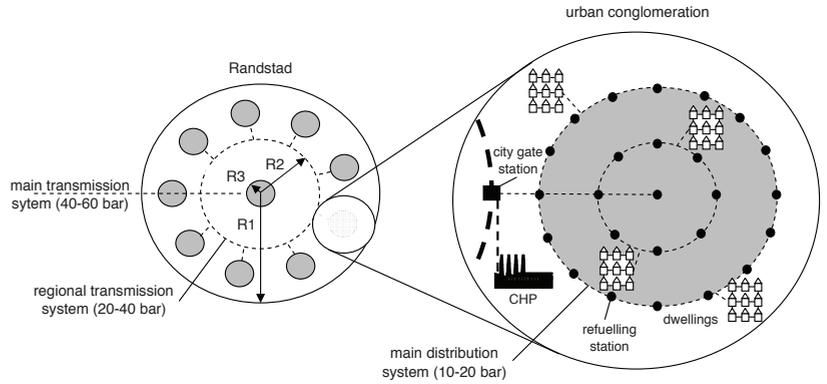


FIGURE 4. Conceptual hydrogen transmission and distribution infrastructure

the importance of considering possible future expansions (when setting up a large scale infrastructure) in the pipeline diameter selection [17].

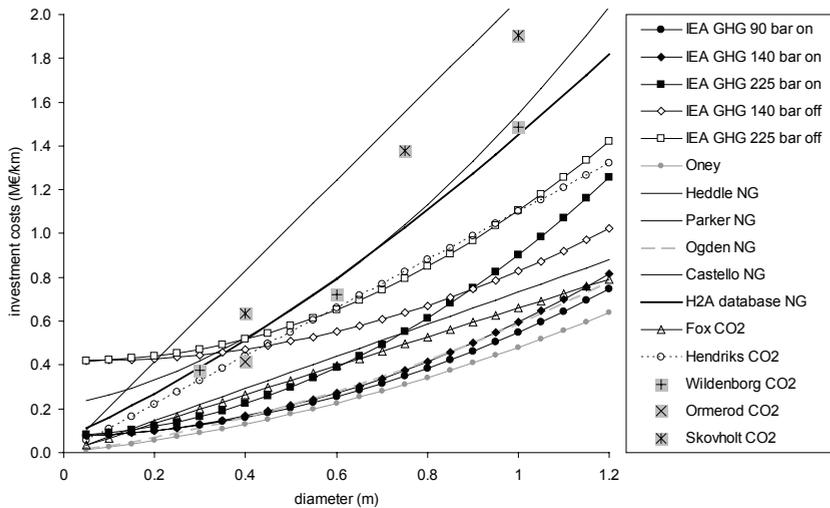


FIGURE 5. Pipeline investment cost as a function of diameter [3,6,15,16,22,40,41,71,73,74,77]

#### 3.4.4 COSTS OF CO<sub>2</sub> AND H<sub>2</sub> INFRASTRUCTURE

Pipeline costs consist of material, labour, right of way (row) and miscellaneous costs. In densely populated urban areas, pipeline construction costs are significantly higher than for rural landscapes, due to safety requirements, higher row and the large number of infrastructural crossings. Construction costs in urban terrain can be 20% higher [70] up to 10 times the costs for a pipeline installed in rural area [71]. The difference in landscape, population density, geographical location and steel prices explain the large variety in construction costs quoted in literature (see Figure 5). Many studies use data on natural gas pipelines. CO<sub>2</sub> pipelines are generally more expensive than natural gas pipelines because the former are operated at higher pressure, requiring a greater wall thickness. For H<sub>2</sub> transmission, embrittlement resistant steel pipelines are required, which are more expensive than natural gas pipelines, generally by a factor 1.3-1.5 [68,69,72]. We use the values of the IEA GHG study assuming a terrain factor of 1.2 [3], as it provides a consistent set of equations suitable for both CO<sub>2</sub> and H<sub>2</sub> pipelines for on- and offshore conditions. Note that these costs are fairly low in comparison to other studies, which may result in an underestimation of transport costs. The costs of H<sub>2</sub> distribution pipelines (and minor CO<sub>2</sub> lines connecting refuelling stations with larger trunk lines) are practically independent on pipeline diameter, as they depend primarily on the labour costs associated with pipeline installation. We use values of 500 €/m for the main distribution grid within the city and 200 €/m for the low-pressure distribution grid in new residential areas based on construction costs for small diameter pipelines [22,40,73]. We assume

pipeline O&M costs to be 2% of the investment costs, representing an average value using data from [3,74-76], and an economic lifetime of 25 years.

Figures 6 and 7 show the dependence of CO<sub>2</sub> and H<sub>2</sub> transmission costs on scale and distance. As CO<sub>2</sub> transmission costs decrease considerably with increasing flow rate, especially at larger distance, it may be more advantageous to construct a network instead of dedicated pipelines. A network consists of a large trunk line, which is connected to various CO<sub>2</sub> sources and sinks by smaller branches, similar to the existing natural gas grid. The advantage of a large backbone structure has been investigated for European conditions in [75]. The results indicate that a backbone offers reduced transport cost in comparison to dedicated pipelines when the availability of reservoirs is restricted to offshore hydrocarbon reservoirs, probably as these fields are clustered and remote from CO<sub>2</sub> sources. Dedicated pipelines would be more attractive when there are sufficient large structures nearby each source. Figure 6 also makes clear that decentralised CCS plants (20 MWe SOFC-GT and 2 MWe MR, capturing 40 and 2 kt/yr, respectively) to be connected to a CO<sub>2</sub> network need to be constructed close to a trunk line in order to keep transport costs reasonable.

Now insights can be created into the trade-off between the costs of CO<sub>2</sub> and H<sub>2</sub> transmission, which is essential for the optimisation of a hydrogen production plant location. Figure 8 shows the transmission costs of CO<sub>2</sub> and H<sub>2</sub> for a 1000 MWe plant fired with coal (CG) or natural gas (SMR). It illustrates that the costs to transport the CO<sub>2</sub> captured at a SMR unit are lower than H<sub>2</sub> transmission over the same distance. For a coal-fired unit, H<sub>2</sub> transport costs are only slightly lower in comparison to CO<sub>2</sub> transport costs. This observation is in fairly good agreement with the results from Ogden [5]. However, the additional costs to connect to the natural gas grid and the presence of coal terminals should be considered as well in choosing the plant location. Given the substantial transmission costs of natural gas [3], it may be more attractive to produce hydrogen close to the natural gas source as proposed by Mintz et al. [79], provided that CO<sub>2</sub> is captured and can be stored in nearby depleted natural gas reservoirs.

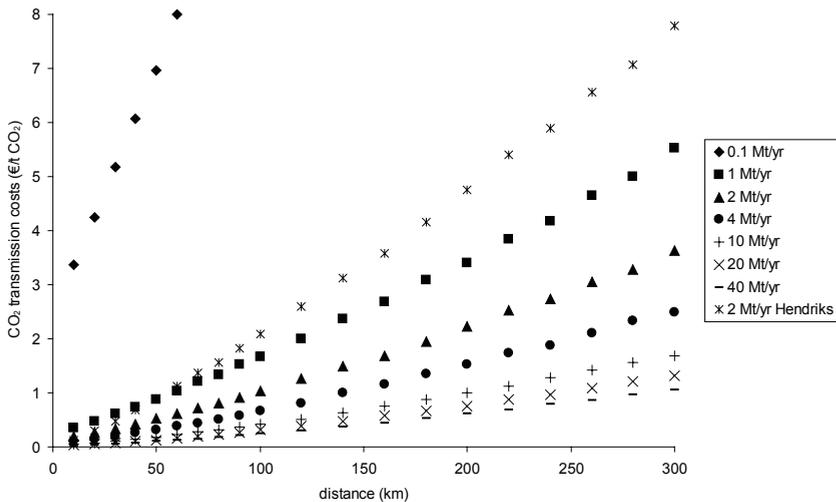


FIGURE 6. Onshore CO<sub>2</sub> transmission cost as a function of distance and mass flow rates based on IEA GHG values for cultivated land [3], excluding compression. We also included the transmission costs using the regression fit from Hendriks et al. [77], which are generally in good agreement with various industrial estimates given in [6,16,78]

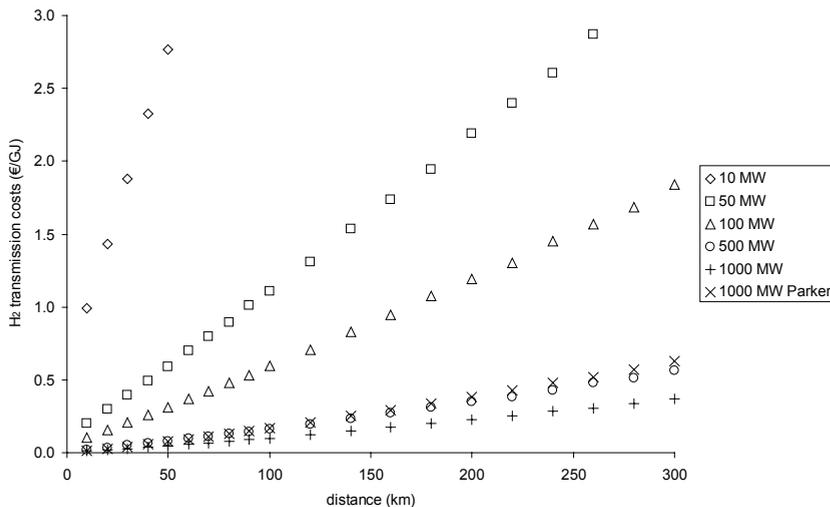


FIGURE 7. H<sub>2</sub> transmission cost as a function of distance and energy flow rates based on IEA GHG values for cultivated land [3], excluding compression. We also included the transmission costs for a 1000 MW<sub>H<sub>2</sub></sub> unit using pipeline costs derived by Parker [73]

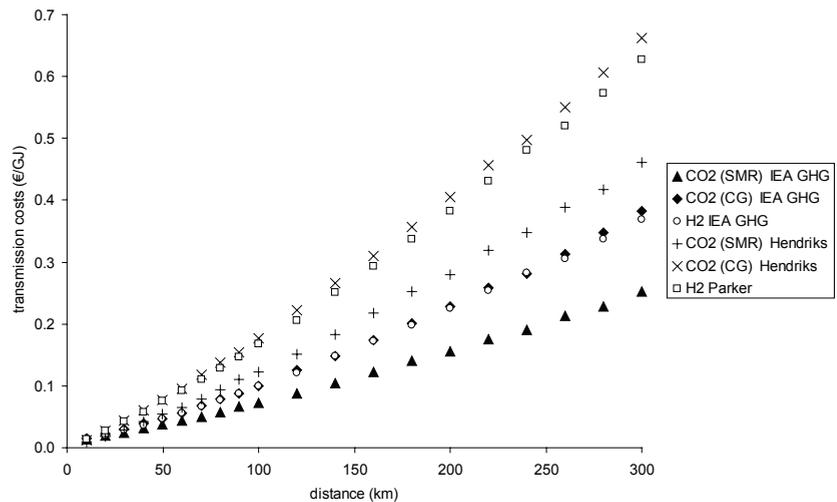


FIGURE 8. CO<sub>2</sub> and H<sub>2</sub> transmission cost of a 1000 MW H<sub>2</sub> unit using coal (CG) and natural gas (SMR), (excluding compression) using IEA GHG values [3] and regression fits derived from [73,77]

### 3.4.5 OTHER H<sub>2</sub> INFRASTRUCTURE REQUIREMENTS

Apart from pipelines, the H<sub>2</sub> infrastructure requirements consist of buffer storage, compressors and dispensers, for which the main features are summarised in Table 9.

TABLE 9. Costs hydrogen supply components

Component <sup>a</sup>	Investment costs	Base scale	Scale factor	O&M <sup>b</sup>
Storage at production unit <sup>c</sup>	10-100 M€	1.63·10 <sup>4</sup> - 1.63·10 <sup>5</sup> kg H <sub>2</sub>	1	1%
Compressor at refuelling station <sup>d</sup>	20 k€	14 kw	0.75	4%
Storage at refuelling station <sup>e</sup>	375 k€	634 kg H <sub>2</sub>	0.95	1%
Gaseous dispenser <sup>f</sup>	18.3 k€	1740 kg H <sub>2</sub> /day	1	-

<sup>a</sup> We consider 15 years lifetime for all components.

<sup>b</sup> Annual costs as share of investment costs derived from [40,80].

<sup>c</sup> In order to safeguard hydrogen supply in case of plant outages, a storage facility is needed for large plants considered in this study. Estimations for required capacity range from 0.5 to 2 days of production [40,41]. We assume a buffer capacity equivalent to 0.5 days. Although geological storage seems the least expensive option for a large unit [12], we consider above-ground storage in pressurised tanks, for which detailed industry figures are available [40].

<sup>d</sup> Representing costs to install a 9-stage reciprocating compressor for compression of maximally 4.8 kg H<sub>2</sub>/hr from 1 up to 480 bar, including drive motor and intercooling [43].

<sup>e</sup> Storage capacity at refuelling stations is required to store hydrogen produced at night time and as back-up for demand variations. The storage capacity required is estimated at 70% of the daily production capacity based on typical refueling station demand patterns and 58% hydrogen recovery with cascade storage/dispensing [43]. Costs are representative for large steel tanks suitable to store hydrogen at 480 bar [43]. Note that storage requirements may be somewhat lower when hydrogen is delivered by pipeline (as hydrogen could be stored in the pipeline).

<sup>f</sup> Derived from costs of compressed natural gas and prototype hydrogen dispenser adjusted for hydrogen specs and mass production [43]. We assume the refueling station is unattended. Another 20% is added to the capital costs of compressor, storage and dispensers to account for miscellaneous costs and contingencies [43].

The work (head) required for compression can be approximated as an isentropic process according to the following formula:

$$W = \frac{ZRT_1 k}{M(k-1)} \left[ \left( \frac{p_2}{p_1} \right)^{\frac{k-1}{k}} - 1 \right] \quad \text{EQUATION 8}$$

The compressor power, also referred to as brake horsepower or shaft power, can be derived from the isentropic head:

$$P = \frac{W\dot{m}}{\eta_{is}\eta_m} \quad \text{EQUATION 9}$$

in which:

w	= work or isentropic head (kJ/kg)
Z	= compressibility factor (set at 1 for H <sub>2</sub> )
R	= universal gas constant (8.3145 J/mol.K)
T <sub>1</sub>	= suction temperature (K)
k	= specific heat ratio (C <sub>p</sub> /C <sub>v</sub> = 1.41 for H <sub>2</sub> )
M	= molar mass (kg/kmol)
p <sub>1</sub>	= suction pressure (Pa)
p <sub>2</sub>	= discharge pressure (Pa)
P	= compressor power (kW)
$\dot{m}$	= mass flow rate (kg/s)
$\eta_{is}$	= isentropic efficiency (0.7-0.75)
$\eta_m$	= mechanical efficiency (0.99)

The choice of a compressor depends mainly on the flow rate and the differential pressure required. For compression of large, continuous quantities of H<sub>2</sub> up to 60 bar, we consider multi-stage centrifugal compressors with an isentropic efficiency of 75% [3,17,81]. Compressing relatively small quantities of H<sub>2</sub> up to 480 bar requires a multi-stage reciprocating compressor. The overall efficiency of a reciprocating compressor is circa 70% [43]. For multi-stage compressors, the power requirements may be overestimated when using Equation 9 (for a single-stage compressor), as the gas is cooled between the different stages, which reduces the work required for compression. Power requirements for multi-stage compression with N stages can be calculated from Equation 10 (derived from [82]). For H<sub>2</sub> compression, typically a compression ratio of 2 is applied [43].

$$W = \frac{ZRT_1}{M} \frac{Nk}{k-1} \left[ \left( \frac{p_2}{p_1} \right)^{\frac{k-1}{Nk}} - 1 \right] \quad \text{EQUATION 10}$$

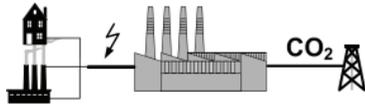
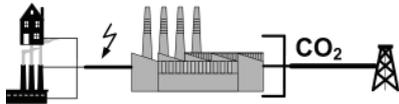
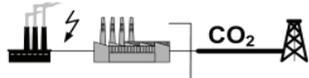
## 3.5 CCS CHAINS OVERVIEW

Tables 10 and 11 give an overview of the chains considered in this study.

TABLE 10. Electricity and hydrogen chain definitions

Chain code	Conversion + capture technology
<i>Short-term electricity</i>	
ST-E1	PC + chem. absorption (600 MW <sub>e</sub> )
ST-E2	IGCC + phys. absorption (600 MW <sub>e</sub> )
ST-E3	NGCC + chem. absorption (600 MW <sub>e</sub> )
<i>Long-term electricity</i>	
LT-E1	Advanced PC + chem. absorption (600 MW <sub>e</sub> )
LT-E2	Advanced IGCC + phys. absorption (600 MW <sub>e</sub> )
LT-E3	IG-Water (600 MW <sub>e</sub> )
LT-E4	IG-SOFC-GT + HSD (600 MW <sub>e</sub> )
LT-E5	Advanced NGCC + chem. absorption (600 MW <sub>e</sub> )
LT-E6	MR-CC (600 MW <sub>e</sub> )
LT-E7	CLC (600 MW <sub>e</sub> )
LT-E8	AZEP (600 MW <sub>e</sub> )
LT-E9	SOFC-GT + afterburner (20 MW <sub>e</sub> )
<i>Long-term hydrogen<sup>a</sup></i>	
LT-H1-MOB/RES/CHP	Advanced ATR + chem. absorption (1000 MW <sub>H<sub>2</sub></sub> )
LT-H2-MOB/RES/CHP	Advanced CG + phys. absorption (1000 MW <sub>H<sub>2</sub></sub> )
LT-H3-STL	SMR/CG + absorption + direct reduction
LT-H4-MOB	MR (2 MW <sub>H<sub>2</sub></sub> )
<sup>a</sup> MOB	= transport sector,
RES	= residential sector,
CHP	= industrial CHP,
STL	= steel production

TABLE 11. Infrastructure assumptions

Chain code	Electricity/H <sub>2</sub>	CO <sub>2</sub>
ST-E1-E3	Transmission line + distribution grid	100 km transmission line to onshore gas field
		
LT-E1-E8	Transmission line + distribution grid	10 km transmission line to CO <sub>2</sub> trunk line
		
LT-E9	Directly connected to distribution grid	50 km transmission line to CO <sub>2</sub> trunk line
		

Chain code	Electricity/H <sub>2</sub>	CO <sub>2</sub>
LT-HI-MOB	200 km transmission line + 125 km regional transmission line + 70 km high-P distribution line	10 km transmission line to CO <sub>2</sub> trunk line
LT-HI-RES	200 km transmission line + 125 km regional transmission line + 70 km high-P distribution line + 2500 km low-P distribution line	10 km transmission line to CO <sub>2</sub> trunk line
LT-HI-CHP	200 km transmission line + 125 km regional transmission line + 10 km high-P distribution line	10 km transmission line to CO <sub>2</sub> trunk line
LT-H2-MOB/ RES/CHP	20 km transmission pipeline, further infrastructure is identical to LT-HI	10 km transmission line to CO <sub>2</sub> trunk line
LT-H3-STL	-	10 km transmission line to CO <sub>2</sub> trunk line
LT-H4-MOB	-	70 km satellite line + 125 km transmission line to CO <sub>2</sub> trunk line <sup>a</sup>

<sup>a</sup> For all refuelling stations

## 4 RESULTS CHAIN ANALYSIS

### 4.1 ELECTRICITY

Figure 9 shows that the contribution of CO<sub>2</sub> transport and storage to the overall electricity costs is relatively small and does not affect the ranking among technologies. This is especially true for the large natural gas-fired technologies, due to the relatively small quantities of CO<sub>2</sub> transported and stored. Storage costs are low as we considered relatively large onshore gas fields. Offshore reservoirs might deserve

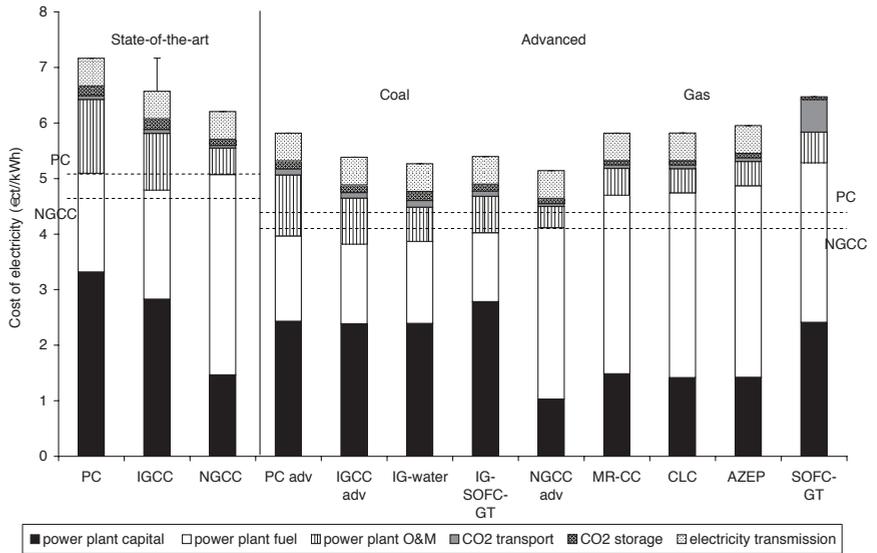


FIGURE 9. Electricity production costs including CCS and electricity transmission. The dashed horizontal lines represent COE of PC and NGCC without CO<sub>2</sub> capture

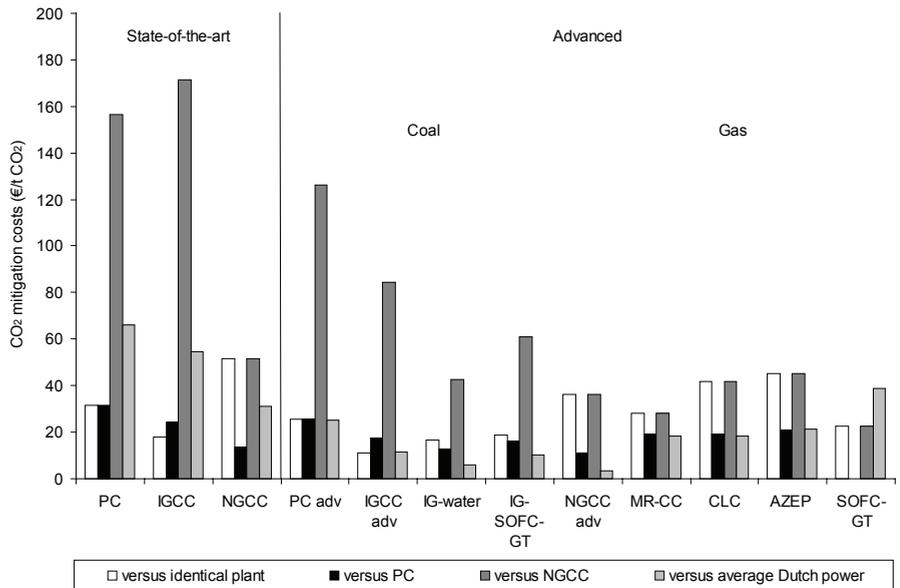


FIGURE 10. CO<sub>2</sub> mitigation costs of electricity production with CCS vis-à-vis different reference systems, including indirect emissions of fuel extraction and transport

preference, although this goes at the expense of higher transport and storage costs. Offshore transport costs for a 600 MW<sub>e</sub> PC unit over 100 km are estimated at 0.15 €/ct/kwh, circa twice the costs for onshore transport. Storage costs for this unit increase to 0.5 €/ct/kwh for a typical offshore gas field in comparison to circa 0.2 €/ct/kwh for an onshore gas field. In the longer term, transport costs per km are expected to decrease slightly due to economy of scale of the CO<sub>2</sub> network. The calculations show that COE of decentralised SOFC units with CCS are higher than COE of central units, as the advantage of omitting electricity transmission does seem to be outweighed by the additional costs of CO<sub>2</sub> transport. However, SOFC efficiency forecasts are higher than for advanced Brayton-cycles with CO<sub>2</sub> capture, which makes it a more safe technology with respect to energy price volatility.

Figure 10 illustrates why CO<sub>2</sub> mitigation costs should be used and interpreted with care, as the outcome is strongly depending on the context i.e. the choice of the reference system. When the fuel choice is fixed for a specific plant (e.g. due to present/absent infrastructure or strategic choices), one should compare a coal-fired power plant without CCS with a coal-fired unit equipped with CCS. In this setting, CO<sub>2</sub> mitigation costs of coal-fired power plants with CCS are lower than for gas-fired power plants with CCS. When replacing a mothballed PC unit for a NGCC with CCS instead of building a new PC unit, CO<sub>2</sub> mitigation costs are lower than when installing a PC unit with CCS, up to gas prices around 6.5 €/GJ. Mitigation costs will be relatively high for coal-fired units with CCS when the reference is a NGCC. This is mainly explained by the significant CO<sub>2</sub> emissions of a PC unit with CCS in comparison to an NGCC without CCS. CO<sub>2</sub> emissions of power production with CCS are between practically 0 kg CO<sub>2</sub>-eq/kwh for advanced gas-fired units up to 0.2 kg CO<sub>2</sub>-eq/kwh for a PC, of which circa 60% are direct CO<sub>2</sub> emissions and the remaining 40% are caused by GHG emissions of coal mining and transport.

#### 4.2 HYDROGEN

As illustrated by Figure 11, the additional costs of CO<sub>2</sub> transport and storage are negligible for large-scale H<sub>2</sub> production. As the additional costs of capture are modest as well [1], the carbon price required to induce CCS at advanced large-scale hydrogen plants is relatively low: 10-20 €/t (see black bars in Figure 12).

Hydrogen costs are to a large extent determined by the infrastructure requirements. For central plants supplying the transport and residential sector, costs of hydrogen infrastructure are equal or higher than the costs to produce hydrogen. It appears that the higher costs of H<sub>2</sub> production and CO<sub>2</sub> transport inherent in decentralised production units do not compensate for the omitted costs of hydrogen infrastructure in centralised chains. In addition, H<sub>2</sub> compression costs for (decentralised) MR units are significantly higher in comparison to centralised chains as H<sub>2</sub> at MR is produced at 1 bar. From that

perspective, MR units are more suitable for markets where hydrogen is required at near-atmospheric pressure (e.g. in households). Another disadvantageous factor is that natural gas and electricity costs are significantly higher for decentralised production. Assuming the refuelling stations owner enforces identical price agreements for natural gas and electricity as large industrial users, COH might be decreased to circa 18 €/GJ.

Taking a closer look at H<sub>2</sub> infrastructure makes clear that the costs of distribution are a major contributor to overall production costs for the transport and residential sector. A breakdown of H<sub>2</sub> infrastructure costs shows that especially the costs of the low-pressure distribution grid are the main cause for the high costs to deliver hydrogen to the front door of individual dwellings (80% of transmission and distribution costs). For the transport sector, also the costs of the refuelling stations are relatively high.

Although CO<sub>2</sub> emissions are greatly reduced with CCS, the remaining emissions are substantial: 10-40 kg CO<sub>2</sub>-eq/GJ in comparison to 50-120 kg CO<sub>2</sub> captured/GJ. This is part of the explanation why CO<sub>2</sub> mitigation costs, depicted in Figure 12, are relatively high for sectors where natural gas (emission factor of 57 kg CO<sub>2</sub>-eq/GJ) is the reference fuel. For the residential area, the other part of the story is obviously formed by the high costs of H<sub>2</sub> supply and PEMFC, which is not outweighed by the higher efficiency of the PEMFC. Mitigation costs in the transport sector are mainly determined by the relatively high costs of the fuel cell. This is illustrated by the costs per driven kilometre: from the 0.06 €/km for an FCV (versus 0.04 €/km for ICEV), circa 20% can be attributed to H<sub>2</sub> costs, the rest being costs of the drive train and storage tank.

In the steel industry, replacing cokes in the traditional BF route for hydrogen or syngas in DR routes results in CO<sub>2</sub> mitigation costs of 20-40 €/t CO<sub>2</sub>. Costs of CO<sub>2</sub> capture from BF gas in the traditional BF route are estimated at 15 €/t CO<sub>2</sub>, comparable to the value calculated in [83]. Figure 13 shows the production costs of various steel production technologies. Circored technology results in relatively high production costs due to the high fuel costs. For coal-based Midrex, the explanation lies mainly in higher expenses in iron ore. The costs of Circofer and the traditional BF route with CO<sub>2</sub> capture from BF gas are relatively close. Although the additional costs due to CCS is very small for Circofer (as CO<sub>2</sub> is already separated), the additional indirect emissions due to electricity requirements of EAF make that mitigation costs (versus BF without capture) appear slightly higher in comparison to capture of CO<sub>2</sub> from BF gas in the classical route. So the primary reason to switch to DR would not be CO<sub>2</sub> emission reduction, but the omission of the polluting coke oven plant.

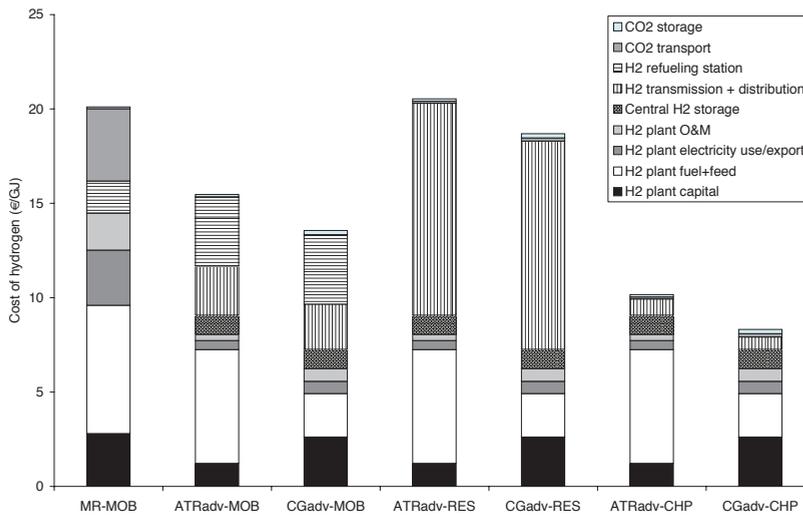


FIGURE 11. Hydrogen production costs including CCS and H<sub>2</sub> infrastructure for delivery to various end-users. (MOB = transport sector, RES = residential sector, CHP = industrial CHP). Costs of H<sub>2</sub> compression in centralised production have been accounted in the H<sub>2</sub> plant (up to 60 bar) and refuelling station (up to 480 bar). In membrane reformers, H<sub>2</sub> compression is accounted for in the production

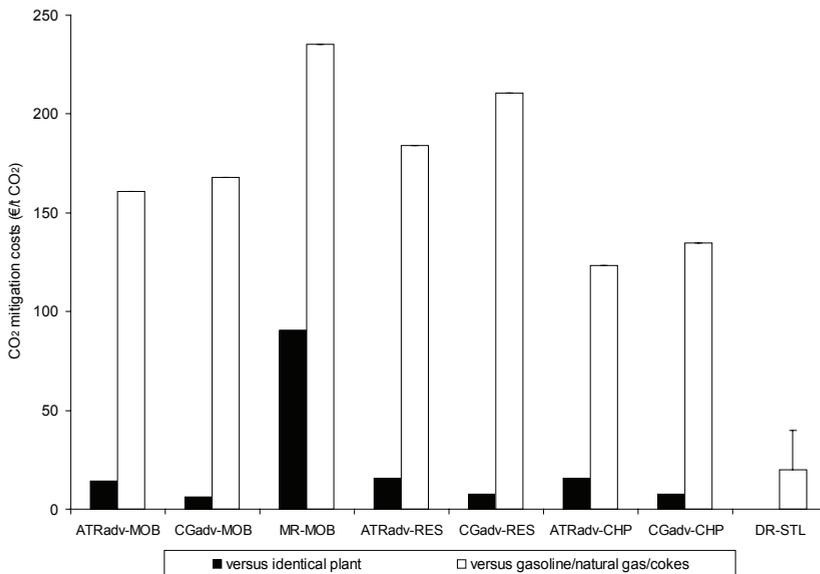


FIGURE 12. CO<sub>2</sub> mitigation costs of hydrogen application vis-à-vis different reference systems, including indirect emissions of fuel extraction and transport. MOB = transport sector, RES = residential sector, CHP = industrial CHP, DR-STL = steel production by means of direct reduction

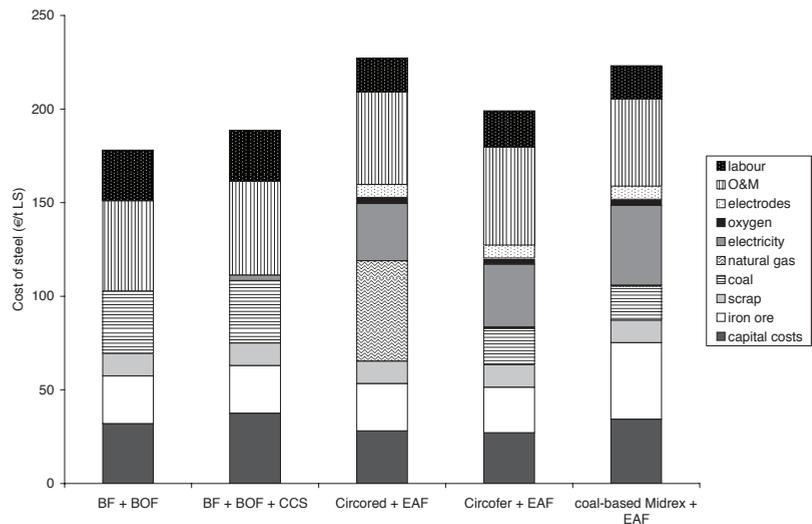


FIGURE 13. Steel production costs for a tonne of liquid steel, based on the blast furnace route (with and without CCS) and direct reduction routes (all with CCS)

#### 4.3 SENSITIVITY ANALYSIS

Figures 14 and 15 show the results of sensitivity analyses performed on several promising or likely technologies that may be implemented, encompassing the complete spectrum of technology (combustion, fuel cells, gasification), fuel (natural gas versus coal) and scale (central versus decentralised units). Apart from the basic economic variables, for which the variation is specified in Table 1, the impact of uncertainty in plant capital costs ( $\pm 30\%$  for state-of-the-art technologies and  $50\%$  for advanced technologies such as SOFC and gasification, which are not mature yet) is assessed. Note that the fuel price and capacity factor are correlated to each other, which may result in even a wider range as shown in Figure 14, especially for NGCC. A power plant dispatch model is required to study the joint impact of these parameters in more detail, which is beyond the scope of this study.

The coal-fired power plants and the SOFC-GT are, not surprisingly, more sensitive to variation in capital costs, interest rate and load factor, whereas COE of NGCC is affected especially by the fuel price. Despite the coal price increase in recent years, prices are likely to moderate the coming decades [84]. Gas prices are subject to various trends such as increased gas-to-gas competition and shift to more global markets. The exact impact in the long term is hard to establish. According to the IEA World Energy Outlook, gas prices in Europe may drop the second half of this decade and then gradually rise to around 4 €/GJ in 2030 (considering oil prices of 25-35 \$/bbl) [84]. The natural gas breakeven price at which COE of PC with CCS and NGCC with CCS are equal, assuming a coal price of 1.7 €/GJ and both are operated at equal capacity factor of 85%, lies between 6-6.8 €/GJ for CO<sub>2</sub> prices between 0 and 100 €/t CO<sub>2</sub>. Even if the capacity factor for NGCC with CCS is decreased to 60% at the

default gas price (4.7 €/GJ), COE would still be slightly lower in comparison to PC with CCS. IGCC with CCS<sup>9</sup> appears more competitive than PC with CCS, resulting in somewhat lower break-even natural gas prices.

9. Note that the IGCC in Figure 14 represents an advanced concept, which cannot be compared to state-of-the-art PC and NGCC.

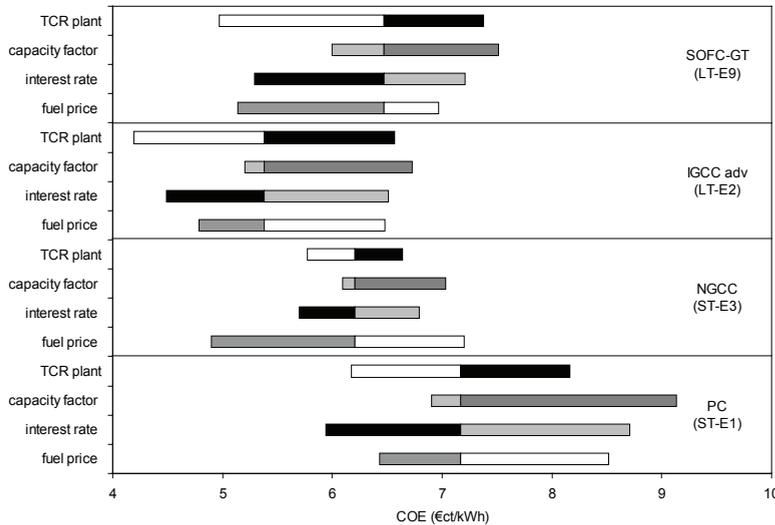


FIGURE 14. Sensitivity analyses of various electricity production chains with CCS. The chain codes (see Table 10) are given in parentheses and the variation in parameters is given in Table 1

The uncertainty in CO<sub>2</sub> transport costs hardly has any effect on the COE. Assuming structurally higher pipeline costs (up to a factor 2 in comparison to the base case) and a maximum transport length of 200 km, COE of a PC unit are increased with a mere 0.3 €/ct/kWh. The uncertainty with respect to storage costs is mainly related to reservoir characteristics. In the most pessimistic case, assuming storage costs of 10 €/t CO<sub>2</sub> for deployment of several smaller, deep offshore reservoirs with low permeability, COE for a PC are increased with nearly 1 €/ct/kWh. As illustrated by Figure 15, centralised chains with large-scale hydrogen production and CCS offer cheaper hydrogen than decentralised production by means of MR with CCS, for the range in parameters studied here. Large-scale ATR units with CCS are competitive with CG units with CCS at gas prices of 3-3.5 €/GJ, which is considerably lower than the breakeven price of their electricity counterparts (NGCC with CCS versus IGCC with CCS). COH are strongly dependent on the interest rate due to the capital-intensive infrastructure required to supply hydrogen to refuelling stations and dwellings. The uncertainty in costs of the distribution network, especially the residential hydrogen grid, has a major impact. The costs of this grid depend on local conditions and might be a factor 2.5 more expensive than our default value, resulting in COH around 33 €/GJ.

CO<sub>2</sub> mitigation costs of hydrogen application as presented in Figure 12 are to a large extent determined by the assumptions in fuel prices and end-use conversion efficiency and costs, for which large uncertainties exists. Although current oil prices (65-70 \$/bbl) are historically high, it is unclear whether such high prices will retain the coming decades. In 2004, the World Energy Outlook forecasted oil prices between 25 and 35 \$/bbl up to 2030 [84]. If investments in oil infrastructure in the Middle East are not significantly increased, oil prices might be as high as 50 \$/bbl (nearly 9 \$/GJ) in the long term [85]. Assuming gasoline costs at the refuelling station of 15 €/GJ instead of the base case value of 9 €/GJ, CO<sub>2</sub> mitigation costs would decrease to circa 80-90 €/t CO<sub>2</sub> for centralised hydrogen chains. Mitigation costs can vary with roughly 100 €/t CO<sub>2</sub> for the range of PEMFC efficiencies quoted in literature [4,43-45]. The uncertainty in fuel cell costs has an even bigger impact on the performance of hydrogen. If we consider vehicle retail prices projected for 2010 as quoted in [34], assuming fuel cell costs of 100 €/kw, mitigation costs would be more than 700 €/t CO<sub>2</sub>. Note that a significant improvement still has to be made to achieve a level of 100 €/kw given current fuel cell costs (produced in low quantities)<sup>10</sup> [4,50, 86]. Although PEMFC costs for stationary purposes are higher than mobile PEMFC costs, as the former requires a longer lifetime and the capacity is lower, it is unclear what cost difference can be expected. A lower range of 500 €/kw for stationary fuel cell systems [87] would decrease CO<sub>2</sub> mitigation costs with 25-30%. Apparently, the impact of PEMFC costs on CO<sub>2</sub> mitigation costs for stationary applications is less severe than for mobile applications; energy costs are a more important factor.

10. The cost of current fuel cell systems for mobile applications is estimated at 250-300 \$/kW at a production volume of 500,000 units per year [51].

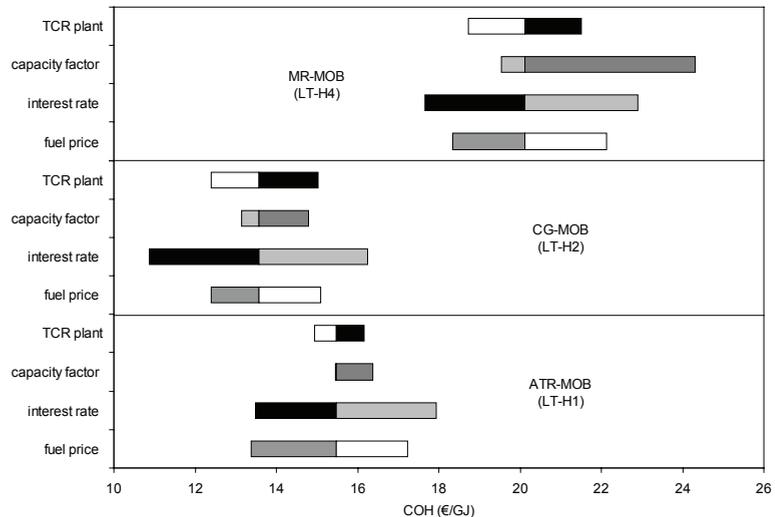


FIGURE 15. Sensitivity analyses of various hydrogen production chains with CCS supplying the transport sector (MOB). The chain codes (see Table 10) are given in parentheses and the variation in parameters is given in Table 1. The impact of variation in capacity factor includes the increased CO<sub>2</sub> and H<sub>2</sub> transmission costs when pipelines are underutilised

## 5 DISCUSSION AND CONCLUSION

A chain analysis has been performed for promising CCS options, incorporating a wide variety of technologies, infrastructural settings, hydrogen end-use markets and reference systems to study various CCS configurations under specific conditions and assumptions. The results indicate that the overall impact of CCS on CO<sub>2</sub> emissions and electricity production costs is significant. CO<sub>2</sub> emissions per kWh are reduced with 75% up to practically 100% with respect to an equivalent power plant without CCS. Note that the overall chain emission of a PC unit with CCS is still more than half the emission of a NGCC without capture. Electricity costs are increased with 25-50% and 15-35% for state-of-the-art and advanced concepts, respectively. The range of studies presented in the IPCC special report on CCS [88] shows a higher upper boundary. Some studies forecast a COE increase of 90% for PC or NGCC with post-combustion capture, transport and storage, which can mainly be explained by more conservative estimations for capital costs and/or energy use of CO<sub>2</sub> capture.

The cost increase is dominated by CO<sub>2</sub> capture and compression, representing 65-90% of the additional costs due to CCS. Only 2-11% of total production costs for central units can be attributed to CO<sub>2</sub> transport and storage. According to Rubin et al. [89], the share of CO<sub>2</sub> transport over 160 km and storage into an aquifer is 5-10% of the COE. For typical Dutch conditions (i.e. a maximum transport distance of 200 km), transport costs for a large power plant are estimated at 1-7 €/t CO<sub>2</sub> (the upper range representing offshore conditions), corresponding to 0.05-0.3 €/kWh. Although CO<sub>2</sub> transport costs are low on a kWh basis, the total investment costs to construct a CO<sub>2</sub> network are large. Considering the length of the high-pressure transmission grid for natural gas, we estimate the investments costs of a CO<sub>2</sub> network at a few billion Euros. The extent of the CO<sub>2</sub> network could be minimised when power plants with CCS are constructed in the Northern provinces, where the bulk of storage capacity is located. Only a limited capacity can be fit into the current network; investments may be required to strengthen the existing electricity grid to carry the electrons from the North down to the major electricity demand centres. More in-depth research is required to study this trade-off for optimal decision making on new plant locations.

The contribution of CO<sub>2</sub> transport is significantly higher for decentralised power generation with CCS. The general consensus is that decentralised technologies are not suited for CCS for this reason and due to concerns over economies of scale in capture technology. However, specific decentralised technologies that enable inherently low-cost CO<sub>2</sub> capture such as SOFC may produce electricity with CCS at reasonable costs, provided that the plant can be connected to a CO<sub>2</sub> trunk line located nearby. Another option would be to install the plant on top of a CO<sub>2</sub> storage reservoir. Note that storage costs will increase when CO<sub>2</sub> from a single 20 MW<sub>e</sub> distributed unit is injected into a reservoir that is characterised by a permeability that allows for much higher injection rates.

CO<sub>2</sub> storage in onshore and offshore gas fields or aquifers costs 1-10 €/t CO<sub>2</sub> at injection rates of at least 1 Mt/yr, corresponding to 0.05-0.9 €/t/kwh. In the Netherlands, natural gas fields are the primary target reservoirs for CO<sub>2</sub> storage. Whether enough suitable gas reservoirs (excluding Groningen) will be available to store large quantities in the longer term is open to question, considering future UGS demand and risk profiles of reservoirs. Possibly, aquifers and coal seams also need to be exploited.

The prospects and competition between different CCS technologies are depending on several uncertain parameters such as fuel prices, capacity factor and capital costs, for which a sensitivity analysis was performed. COE for NGCC with CCS are lower than for PC with CCS over the studied range in parameters (gas prices up to 6 €/GJ). Only combinations of high gas prices and low capacity factors favour PC with CCS. The breakeven natural gas price for IGCC with CCS is somewhat lower in comparison to PC with CCS. It should be noted that NGCC without CCS is the most competitive technology in a world where natural gas prices prevail below the range from 5.5-7 €/GJ and CO<sub>2</sub> prices of 0-55 €/t CO<sub>2</sub>. Kreutz and Williams [90] came to comparable findings, although in their analysis the perspectives for IGCC are somewhat more optimistic and NGCC with CCS enters the playfield at much higher CO<sub>2</sub> prices.

The impact of CCS on costs of centralised hydrogen production and supply is not as substantial as for electricity production; they add another 1-2 €/GJ for transport distances up to 200 km. Of the total costs of centralised H<sub>2</sub> production (between 8 and 21 €/GJ for base case assumptions on fuel price etc.), maximally 0.5 and 0.7 €/GJ are added by CO<sub>2</sub> transport and storage, respectively. Hence the carbon price required to equip hydrogen units with CCS is rather low; typically in the order of 10-20 €/t CO<sub>2</sub>. Mitigation costs for replacing natural gas or gasoline with hydrogen produced with CCS are 100-200 €/t CO<sub>2</sub>. These values should be considered with care, as they are strongly dependent on the choice of the reference system and assumptions in fuel prices and fuel cell performance and costs. Nevertheless, these costs are fairly high in comparison to mitigation costs of CCS application in the power sector, which are estimated at 10-170 €/t CO<sub>2</sub>, but in most cases below 60 €/t CO<sub>2</sub>. So hydrogen production with CCS and application for any other purpose than current (chemical) use does not deserve preference as CO<sub>2</sub> mitigation option. Hydrogen could also be adopted in the steel industry. However, CO<sub>2</sub> mitigation costs of hydrogen replacing cokes are somewhat higher in comparison to CO<sub>2</sub> capture from the blast furnace gas in the traditional BF route using cokes.

Hydrogen production and supply costs are to a large extent determined by the H<sub>2</sub> infrastructure requirements. Even for those chains with minimal infrastructure (e.g. CHP units), supply costs are significant, mainly due to the need of a centralised buffer. The base case costs to produce and deliver hydrogen to decentralised CHP units are estimated at 8-10 €/GJ (versus current natural gas price of circa 5 €/GJ). Base case

hydrogen costs for the transport sector are between 14 and 16 €/GJ for large, advanced CG/ATR units, respectively, versus gasoline costs at the refuelling station of 9 €/GJ. Approximately 50% of the costs are attributed to H<sub>2</sub> storage, transmission, distribution, compression and dispensing. Ogden [40] estimated hydrogen production and delivery costs for a similar configuration at 18 €/GJ. The difference lies mainly in the refuelling station; Ogden estimated costs of a manned refuelling station at circa 6 €/GJ (of which circa 50% labour costs) versus 3.7 €/GJ for an unmanned station considered in our study.

In a future envisioning hydrogen supply to households, total production and supply costs may even exceed 20 €/GJ (versus 8 €/GJ for natural gas), mainly due to the extensive low-pressure distribution grid. Hence H<sub>2</sub> supply to a CHP system on a block-level, from where electricity and heat are delivered to individual households, may be more attractive. In such a multi-family configuration, H<sub>2</sub> delivery costs would be around 10-12 €/GJ assuming a heat grid is present (as in many new build locations in the Netherlands). Another argument for multi-family instead of single-family PEMFC units is economies of scale of the fuel cell system, as was demonstrated by Kreutz and Ogden [91].

Apart from the costs of hydrogen supply, the construction of a dedicated hydrogen infrastructure may involve practical problems, as the Netherlands is a densely populated country with a high density of natural gas pipelines and other infrastructure. The risks associated with hydrogen transmission, distribution and end-use is of major concern, and might inhibit the use of hydrogen, especially in households and cars. From a wider perspective, the value-added of hydrogen for stationary purposes is rather poor due to the relatively high share of relatively 'clean' natural gas. For the transport sector, the problems (oil dependency, GHG emissions and problems with local air pollution) are more urgent, which may prove a strong driver for hydrogen use.

If penetration levels of hydrogen in the transport sector appear slower than projected, centralised hydrogen production with CCS may enter the playfield beyond 2030. Alternatively, decentralised MR would be an interesting technology when hydrogen demand (density) is too low to justify centralised production, although COH (20 €/GJ) are still noticeably higher than for centralised production. As the facilities to enable CCS could be added relatively simple, MR might first be operated in the non-capture mode when there is no CO<sub>2</sub> infrastructure to which it can be connected.

This chain analysis has shown the impact of different CCS chains in a non-dynamic way, in which the level of H<sub>2</sub> application and CO<sub>2</sub> reduction was fixed. A more detailed analysis is required to assess the consequences of demand and supply fluctuations on load factor, pipeline utilisation and storage requirements. Part of the fluctuations may be covered by the buffer capacity offered by the extensive pipeline network. For storage of large quantities H<sub>2</sub>, alternatives for costly steel tanks (composite tanks, geological reservoirs or liquefied hydrogen, as present in the existing hydrogen network in the Rijnmond area) should be considered.

Finally, the extent of CO<sub>2</sub> emission reduction by CCS has been set at 20 mt/yr in 2030. Further research should focus on the factors that affect CCS implementation rates in order to define scenarios that allow for a more structural assessment of the potential and costs of CCS in the coming decades.

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## CHAPTER IV

### TECHNO-ECONOMIC PROSPECTS OF SMALL-SCALE MEMBRANE REACTORS IN A FUTURE HYDROGEN-FUELLED TRANSPORTATION SECTOR \*

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#### ABSTRACT

The membrane reactor is a novel technology for the production of hydrogen from natural gas. It promises economic small-scale hydrogen production e.g. at refuelling stations and has the potential of inexpensive CO<sub>2</sub> separation. Four configurations of the membrane reactor have been modelled with Aspen<sup>Plus</sup> to determine its thermodynamic and economic prospects. Overall energy efficiency is 84%<sub>HHV</sub> without H<sub>2</sub> compression (78% with compression up to 482 bar). The modelling results also indicate that by using a sweep gas, the membrane reactor can produce a reformer exit stream consisting mainly of CO<sub>2</sub> and H<sub>2</sub>O (>90%<sub>mol</sub>) suited for CO<sub>2</sub> sequestration after water removal with an efficiency loss of only 1%<sub>pt</sub>. Reforming with a 2 MW membrane reactor (250 unit production volume) costs 14 \$/GJ<sub>H<sub>2</sub></sub> including compression, which is more expensive than conventional steam reforming + compression (12 \$/GJ). It does, however, promise a cheap method of CO<sub>2</sub> separation, 14 \$/t CO<sub>2</sub> captured, due to the high purity of the exit stream. The well-to-wheel chain of the membrane reactor has been compared to centralised steam reforming to assess the trade-off between production scale and the construction of a hydrogen and a CO<sub>2</sub> infrastructure. If the scale of centralised hydrogen production is below 40 MW, the trade-off could be favourable for the membrane reactor with small-scale CO<sub>2</sub> capture (18 \$/GJ including H<sub>2</sub> storage, dispensing and CO<sub>2</sub> sequestration for 40 MW SMR versus 19 \$/GJ for MR). The membrane reactor might become competitive with conventional steam reforming provided that thin membranes can be combined with high stability and a cheap manufacturing method for the membrane tubes. Thin membranes, industrial utility prices and larger production volumes (i.e. technological learning) might reduce the levelised hydrogen cost of the membrane reactor at the refuelling station to less than 14 \$/GJ including CO<sub>2</sub> sequestration cost, below that of large-scale H<sub>2</sub> production with CO<sub>2</sub> sequestration (~15 \$/GJ).

## I INTRODUCTION

Driven by concerns over greenhouse gas (GHG) emissions, air quality and security of energy supply, hydrogen has become a contender to replace gasoline and diesel as a future transportation fuel. Used in a fuel cell vehicle (FCV), the efficiency might be 2-3 times higher than current internal combustion engine vehicles (ICEV) whilst emitting solely water vapour. Large-scale plants using fossil feedstock are considered the most economic hydrogen production method [1,2], but major barriers are the cost of building a distribution infrastructure and the need for sufficient demand. Distributed production at refuelling stations, using the already present natural gas infrastructure, is therefore viewed as an attractive near to medium-term option. However, producing hydrogen from fossil fuels such as natural gas still causes significant GHG emissions. The capture and subsequent storage of carbon dioxide (the most important GHG) would prevent this, but at small scales (e.g. refuelling stations) this is considered too expensive with conventional reformer technologies [3]. A novel reforming technology, the membrane reactor (MR), is currently being developed and promises economic small-scale hydrogen production combined with inexpensive CO<sub>2</sub> capture because of the high concentration and pressure of the exiting gas stream [4]. This could avoid a dedicated hydrogen infrastructure, facilitate CO<sub>2</sub> capture at small scales and thus possibly contribute to a more rapid cut in GHG emissions. Since it is expected that significant development of a hydrogen transportation infrastructure will not occur within the next decade [5], the time frame of this study is the medium-term future (2015-2025). Considerable laboratory research is being conducted into this technology (see e.g. [6-9]) and two companies are seeking to commercialise it: Tokyo Gas Company (TGC) [10] and Membrane Reactor Technologies (MRT) [11]. Several experimental and theoretical studies have examined the thermodynamics of the membrane reactor [12-14,6,15,16] and have shown that the MR can reach higher fuel conversions at lower temperatures than conventional technology [17]. Additional advantages are a simple, compact design and simultaneous reaction and separation. These studies however have only been done on an experimental and theoretical level. Little detailed research has been conducted on the economic viability of the MR from a systems perspective, i.e. specifically assessing the impact of simultaneously reforming and purifying in comparison to using membranes only for purification. For example, Criscuoli et al. [18] have analysed the economics for a membrane system, but only for a water gas shift reactor. Other studies only considered large-scale applications and demonstrated considerable discrepancies ([19-21,18] and see for an extensive overview [22]). Since especially scale may have an important impact on the economics of the MR and as the technology could be an early candidate for distributed hydrogen production, small-scale MR has been analysed in more detail in this chapter. In addition, the

opportunity of possible inexpensive small-scale CO<sub>2</sub> separation has been examined. This leads us to the main questions addressed in this chapter:

- *Can the membrane reactor become a competitive alternative to conventional technology to supply H<sub>2</sub> to refuelling stations and which developments are necessary to become cost-effective?*
- *Can the membrane reactor provide an economically viable opportunity for small-scale CO<sub>2</sub> capture, suitable for subsequent storage?*

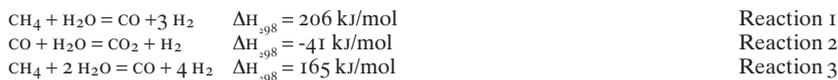
The applied methodology and structure of this chapter are as follows:

1. Technology assessment and selection of suitable configurations (Sections 2 and 3)
2. Collecting technical and economic data on all components of the system through literature and by contacting manufacturers (Section 4)
3. Modelling the membrane reactor configurations in Aspen<sup>plus</sup> to select best operating conditions, evaluate performance and assess sensitivity to different parameters (Sections 5.1 and 5.2)
4. Economic evaluation of the MR based on component cost (Section 5.3)
5. Comparing the fuel supply chain of the MR with conventional technologies regarding costs (Section 6)
6. Overall conclusions (Section 7)

## 2 TECHNOLOGY DESCRIPTION OF A MEMBRANE REACTOR

### 2.1 PRINCIPLE

The principle of the MR is based on steam methane reforming (SMR), currently the most important chemical process for the production of hydrogen or synthesis gas (mixture of mainly H<sub>2</sub> and CO) [23]. SMR consists of three reversible reactions:



The conventional large-scale process is performed in long tubes filled with nickel-based catalyst particles at pressures of 10–41 bar<sup>1</sup>, temperatures around 850°C and steam-to-carbon ratios (s/c ratio) of 2 to 4 [13,24]. The reaction is endothermic and required heat is supplied by a natural gas fuelled furnace. The total hydrogen production process consists of three steps: the steam methane reformer (reaction 1), a water gas shift reactor to enhance CO conversion (reaction 2) and hydrogen purification. Purification is generally performed with pressure swing absorption (PSA) to produce 99.99+% hydrogen, a purity level required for proton exchange membrane fuel cells. The membrane reactor can combine these three steps in one reactor (see Figure 1). In the MR, a membrane is placed inside the reforming tubes. Whilst the

1. All pressures are given in bar absolute.

steam reforming reaction takes place, hydrogen is continuously withdrawn from the reaction zone (retentate side) through the membrane (see Figure 2). Since it is an equilibrium reaction, removing one of the products will shift the equilibrium to the right-hand side according to Le Chatelier’s principle, resulting in an increase in methane conversion ( $MC = \text{methane converted/methane input}$ ). Simultaneously, very pure hydrogen is collected on the product side (permeate side) by using a membrane infinitely selective to hydrogen, i.e. no gas other than hydrogen permeates through the membrane. The amount of hydrogen removal is measured by the hydrogen recovery (HR), defined as the molar flow of hydrogen permeated through the membrane divided by the total molar flow of hydrogen produced. Since methane conversion is higher than with conventional methods, the reactor can operate at lower temperatures and still obtain similar MC.

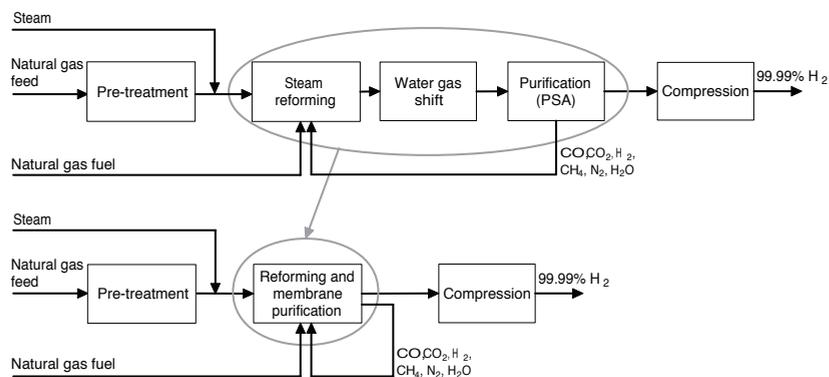


FIGURE 1. Process scheme for the SMR and MR

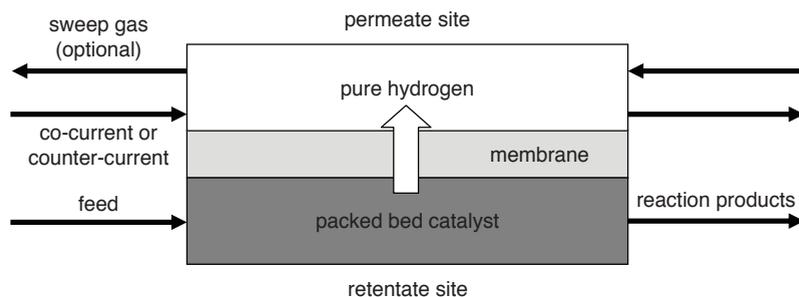


FIGURE 2. Principle of the membrane reactor [8]

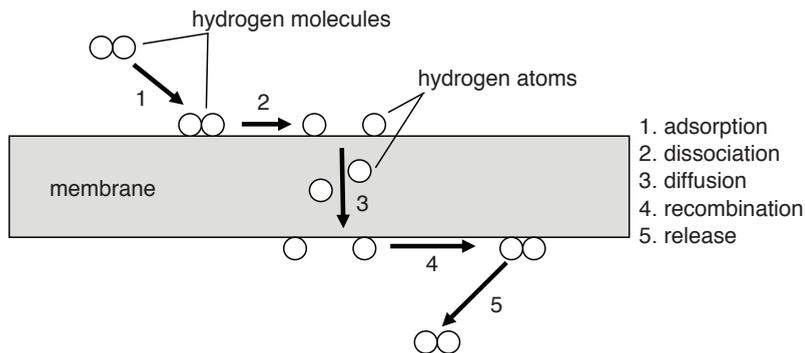


FIGURE 3. Hydrogen permeation for a metallic membrane [9]

2.2 MEMBRANE PERFORMANCE

Palladium is the most widely used membrane material for hydrogen permeation as it is infinitely selective to hydrogen and has demonstrated high permeability. It is usually alloyed with a metal to increase permeability and reduce the effect of hydrogen embrittlement [9,25]. Silver is currently the most popular alloy. Palladium-silver membranes have proven their suitability for hydrogen permeation in numerous experimental studies (see e.g. [26]) and the fabrication technology is well established. By supporting the metal membrane with porous ceramic substrates (Al<sub>2</sub>O<sub>3</sub>), the thickness can be reduced to only several micrometers without compromising stability. Membranes of about 20-50 μm thick are commercially available [19]. Thinner membranes (2-5 μm) are being produced, but this is as yet only in laboratory settings [27-29]. The main challenge remains producing these membranes without any defects [4]. In this study we assume a 20 μm Pd<sub>23</sub>Ag membrane (23 vol% silver) supported by a ceramic substrate and infinitely selective to hydrogen.

Hydrogen atoms permeate through a metal membrane by a solution-diffusion mechanism, following Sievert’s law (see Figure 3) [30]. Under normal conditions, the rate determining step is the diffusion through the membrane as governed by Equation 1.

$$Q_{H_2} = \frac{K_0 \cdot A}{l} \cdot [(p_r)^{0.5} - (p_p)^{0.5}] \tag{Equation 1}$$

Where

- Q<sub>H<sub>2</sub></sub> = hydrogen permeation rate (mol/h)
- K<sub>0</sub> = membrane permeability (mol m/m<sup>2</sup> h Pa<sup>0.5</sup>)
- A = membrane area (m<sup>2</sup>)
- l = membrane thickness (m)
- p<sub>r</sub> = partial pressure on the retentate side (Pa)
- p<sub>p</sub> = partial pressure on the permeate side (Pa)
- δ = driving force (= p<sub>r</sub><sup>0.5</sup>-p<sub>p</sub><sup>0.5</sup>)

It is important to note that the hydrogen flux is inversely proportional to the membrane thickness. According to a review by Dittmeyer et al. [8], membrane permeability for palladium-based membranes of 1.6·10<sup>-8</sup> to

$2.25 \cdot 10^{-5} \text{ mol m/m}^2 \text{ h Pa}^{0.5}$  have been attained in laboratories. We assume a state-of-the-art membrane with a permeability of  $2.25 \cdot 10^{-5} \text{ mol m/m}^2 \text{ h Pa}^{0.5}$ . The driving force is provided by the partial pressure difference of hydrogen between the two sides. To obtain an adequate partial pressure difference, either a high pressure on the retentate side or a low partial hydrogen pressure on the permeate side is required. The latter can be attained by creating a vacuum on the permeate side or by using a sweep gas to lower the hydrogen concentration. Possible inert sweep gases are steam and nitrogen of which steam is most favoured due to its availability and easy separability from hydrogen (assumed in this study). The sweep gas can either flow co-current or counter-current, i.e. flowing in the same direction as the feed flow or in the reverse direction. Previous studies have shown that a counter-current sweep gas is more economic [4,31].

Membrane performance is negatively affected by the presence of other gases and thermal cycling (repeated cycles of cooling and heating the reformer). The presence of sulphur and gases such as nitrogen, steam, CO and CO<sub>2</sub> can decrease the permeability of the membrane significantly. The negative effect on the permeation rate is caused by the competitive adsorption of these compounds on the hydrogen adsorption sites on the membrane [32]. By increasing the temperature to more than 500°C, this effect can be greatly reduced [33]. Hence, for temperatures at which the membrane reactor normally operates (500-600°C), these inhibition effects will not have a serious influence and no negative implications are assumed in our further analysis.

At temperatures around 300°C and in the presence of hydrogen, palladium has a transition between two different lattice phases. By continuous heating and cooling, this transition causes membrane stress resulting in micro-cracks and embrittlement of the membrane [24].

The lifetime of the membrane is therefore strongly dependent on the amount of thermal cycling that it experiences. The negative effect can be partly countered by heating and cooling in the presence of an inert gas such as nitrogen and is hence essential if many start-ups and shut-downs are expected. This adds cost and complexity to the system and may not be sufficient to fully prevent hydrogen embrittlement [34].

Although optimisation of these issues is not investigated here, we assume the amount of thermal cycling should be minimised to ensure a long membrane lifetime (no inert gas is used). Considering the application in the transportation sector, this implies that demand-following operation is not preferred and the focus should be on providing sufficient storage capacity to enable constant operation.

### 2.3 KEY REACTOR OPERATING PARAMETERS

Several operating issues have a significant influence on the thermodynamic and economic performance of the MR and make optimisation of the MR a complex problem. The most important process parameters determining its performance (and costs) are temperature, pressure, steam-to-carbon ratio, load-to-surface ratio<sup>2</sup>

2. The L/S ratio is the molar flow of natural gas feed relative to the membrane surface area.

3.  
The sweep ratio is the molar flow of sweep divided by that of the natural gas feed.

(L/s ratio) and, when a sweep is used, the sweep ratio<sup>3</sup>.

In a conventional SMR, full methane conversion is reached at about 900°C [21]. Hence, at these temperatures no improvement is made with a membrane reactor. The biggest thermodynamic and economic advantages are gained from low temperature operation, 500-600°C. A minimum of 500°C is required to ensure sufficient methane conversion [13,35]. By reducing the temperature, efficiency can be improved and fuel consumption lowered as less heat is required for the reaction.

In a conventional SMR, the thermodynamic equilibrium shifts to the feed side when reaction pressure is increased, lowering methane conversion. Experimental and modelling results indicate that this effect could be the opposite for the MR, given that the permeation capacity of the membrane is sufficient and enough hydrogen is removed from the reaction site [12,21,36]. An increased reaction pressure causes a higher hydrogen flux through the membrane, more than offsetting the negative thermodynamic effect of pressure on the steam reforming reaction.

Thus, higher methane conversion could be attained with higher pressures. High permeation rates reduce membrane area, but also imply increased cost for compressors, pumps and electricity. The optimal value is probably between 10 and 30 bar [14].

An excess of steam is added to the reaction to improve conversion and prevent coke formation. Coke formation is a pervasive problem in steam reforming. The solid reaction product can deactivate the catalyst by depositing on active sites, blocking catalyst pores or causing the metal to separate from the support. It can also block the space, leading to increased bed pressure and eventually obstructing the total flow [37]. Allowing the reaction to proceed to equilibrium at relatively low temperatures and with enough steam, coke formation can be suppressed. The s/c ratio for conventional steam reformers is 2.7-3.2 and is sufficient to prevent this [23]. For the MR, the optimal s/c ratio is in the same range. Although higher s/c ratios increase conversion, the presence of additional steam implies lower hydrogen partial pressure and thus increases membrane surface requirement. In addition, more steam production is needed.

The nominal L/s ratio needs to be designed such that at normal operating conditions costs are as low as possible. Obviously, this depends on several parameters. The higher the L/s ratio, the less membrane surface is required and thus both capital and operational cost are reduced. On the other hand, a lower L/s ratio ensures higher hydrogen recovery and higher methane conversion.

Whether a sweep should be used is determined by another trade-off.

By using a sweep gas the reactor can operate at lower pressures and still obtain a good driving force [12,38]. Disadvantages are that the sweep needs to be produced, elevated to the reaction temperature and afterwards separated from the hydrogen. The sweep ratio is an important parameter: a higher sweep ratio decreases hydrogen partial pressure on the permeate side, resulting in higher hydrogen recovery due to the increased partial pressure difference. Yet, using more sweep

gas requires more heat for steam generation and increases capital cost as additional equipment is required.

#### 2.4 CO<sub>2</sub> SEPARATION

In a conventional SMR, reaction products are usually sent to a PSA unit where hydrogen is separated from the remaining gases. The remaining stream still contains methane, hydrogen and carbon monoxide and is thus redirected to the furnace to raise heat for the endothermic reaction. This can also be done for the MR without the need for the PSA unit. However, if almost complete methane conversion and hydrogen recovery can be attained, the retentate stream will mainly consist of carbon dioxide and steam. Condensation of the steam leaves a concentrated CO<sub>2</sub> stream at reaction pressure and would give the prospect of an inexpensive method for CO<sub>2</sub> capture. Since CO<sub>2</sub> is best transported in a supercritical state (80-100 bar [39]), the high exit pressure will also minimise compression energy. It should be emphasised that only capture of CO<sub>2</sub> present in the retentate stream is economically viable. CO<sub>2</sub> is also produced in the furnace, but the concentration in the flue gases is too low (13%) to enable economic recovery at these scales.

According to two modelling studies [4,19], the retentate stream is almost 100% CO<sub>2</sub> after water removal. This can only be achieved using a counter-current sweep gas to dilute the hydrogen in the permeate stream [40]. If no sweep gas is utilised, approximately 5% of the hydrogen cannot be recovered [26] since a minimum partial pressure difference is required to force the hydrogen through the membrane. Other literature estimates report lower CO<sub>2</sub> concentrations, but these do not use a sweep gas and have not been optimised for CO<sub>2</sub> capture [6,26,41].

In practice, even with a sweep gas it may not be feasible to obtain such high purity CO<sub>2</sub>, due to the optimal requirements of full methane conversion and hydrogen recovery [42,43]. To the authors' knowledge no experimental results are available on the optimisation of the MR for CO<sub>2</sub> capture. Due to this uncertainty, we consider two possibilities: either the CO<sub>2</sub> is pure enough (after water removal) to enable direct transportation or significant levels of contaminants remain in the retentate stream and need to be removed prior to compression and transportation. In the latter case a separation technology is therefore necessary. Considering the scale of the system and the fact that the retentate stream has a relatively high CO<sub>2</sub> partial pressure, cryogenic separation is the preferred technology according to two manufacturers of CO<sub>2</sub> separation systems [44].

Cryogenic separation uses a freezing technique to capitalise on the difference between the freezing point of CO<sub>2</sub> and other gases [45]. The process involves many different steps. First, the off-gas is cooled to about 30°C to condense most of the water, which is removed by a water knock-out tank. The stream is then compressed to 35 bar and cooled to offset the heating of compression. More water is removed in a second

tank, followed by a dehydration step to bring the water content to ppm levels. This is necessary to avoid corrosion and hydrate precipitation in pipelines further downstream [46]. In a refrigeration loop the CO<sub>2</sub> is condensed at -40°C to remove most of the non-condensable gases such as H<sub>2</sub>, CO, CH<sub>4</sub> and N<sub>2</sub> from the mixture. The off-gas can then be sent to the furnace for combustion<sup>4</sup>. With this technology a CO<sub>2</sub> recovery of 75-85% of the carbon dioxide present in the retentate gas can be achieved [47].

4. In practice, about 20% of the methane and 8% of the carbon monoxide present in the retentate stream cannot be separated and is lost [47]. This could be improved by further optimisation.

### 3 SYSTEMS DESCRIPTION

Following the issues described in the previous section, several systems for the membrane reactor can be imagined. Previous studies (see e.g. [4,6,12,19,20]) have mainly considered two configurations, with or without the use of a sweep gas. Although the thermodynamic trade-off between the two configurations has been investigated, the economic trade-off has not been dealt with. In this study, both the non-sweep and sweep configurations are investigated. The retentate stream from the reformer is redirected to the furnace for heat production.

To evaluate the possibility of clean fossil fuel use for hydrogen production, i.e. continued consumption of fossil fuels with CO<sub>2</sub> capture and storage, two additional configurations are studied. The first uses a sweep gas and immediately produces a pure CO<sub>2</sub> stream ready for compression and transportation (ideal). The second is in principle the first configuration (non-sweep), but then supplemented by a cryogenic separation unit to purify the CO<sub>2</sub> (non-ideal). Union Engineering, a manufacturer of CO<sub>2</sub> separation units, has supplied us with a detailed flowsheet of a cryogenic unit specifically based on our situation. Their modelling approach is in accordance with our methodology and is therefore applicable to this study.

This leaves us with four configurations:

- |               |  |
|---------------|--|
| 1. Non sweep: | MR without sweep gas   |
| 2. Sweep:     | MR with sweep gas  |
| 3. Non-ideal: | MR without sweep gas and with cryogenic CO <sub>2</sub> separation |
| 4. Ideal:     | MR with sweep gas and integrated CO <sub>2</sub> capture           |

Figure 4 shows a representation of the system considered in this study. It is based on recent patents [48-50] and Myers et al. [24] and is specifically configured as a small-scale reformer at refuelling stations. We assume the configuration to be suitable for capacities of 0.2 MW to several MW<sup>5</sup>. For large-scale applications, a different configuration may be required. Several aspects are specific for this configuration. The water is taken from the city water supply. Ions such as calcium and chloride present in the water need to be removed by a deionisation tank as they cause serious fouling and corrosion problems and can poison the catalysts [24]. Sulphur and sulphur compounds in the natural gas feed (for reforming) also poison the catalyst by chemically bonding to active sites

5. Unless stated otherwise, MW is based on the HHV of hydrogen produced.

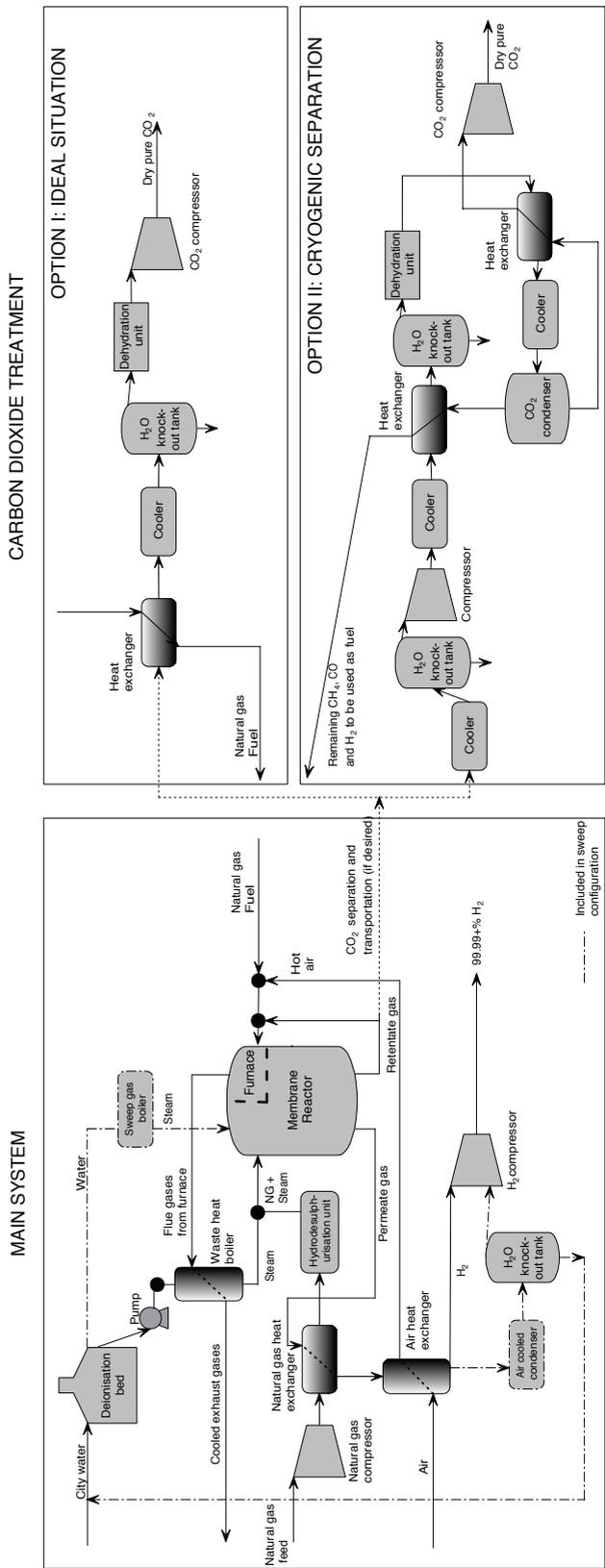


FIGURE 4. Generic system flowsheet

and are therefore removed by a hydrodesulphurisation unit (HDS) coupled with a zinc oxide bed. This step is not required when natural gas is used as fuel in the furnace. Gaseous hydrogen is considered for the system analysis as this currently appears to be the industry's preference [51]. The hydrogen is compressed to 482 bar to enable fast cascade-filling into vehicle tanks at 350 bar (see for a comprehensive explanation: Thomas et al. [52]).

## 4 METHODOLOGY AND SYSTEM ASSUMPTIONS

### 4.1. THERMODYNAMIC MODELLING ASSUMPTIONS AND MODEL VALIDATION

#### 4.1.1 GENERAL

The selected configurations are modelled using the process simulation software Aspen<sup>plus</sup>. Chemical equilibrium approximation is used for the calculations. An excess of highly active catalyst is generally used to guarantee that equilibrium is reached and that kinetic factors play a subordinate role in limiting the reaction [53-55]. For the MR the situation is different since hydrogen is simultaneously withdrawn from the reaction site. Adris et al. [7] have concluded that kinetic properties have limited influence for conditions studied in this analysis, and other studies [37,54,56,57] have demonstrated that the removal of hydrogen through Pd/Ag membranes is generally slow compared to the catalytic reaction rate (as long as no extremely thin membranes are used). Consequently, the gas composition near the membrane equals the gas composition in the catalytic bed [53,54].

In our study we have assumed an excess of catalyst to be available to ensure that equilibrium is reached. The computational effort can therefore be simplified as for kinetic calculations detailed knowledge is required on the reaction rate and its dependence on e.g. temperature, activation energy and pressure. The kinetic properties of conventional steam reforming have been researched extensively, but to the authors' knowledge no study has yet investigated the precise impact of kinetics for the MR. The sources above suggest that the impact is not necessarily significant provided that the membrane is not too thin. It would therefore be interesting to study at which permeation rate and to what degree kinetics starts to significantly influence the reaction.

#### 4.1.2 MEMBRANE REACTOR

The membrane reactor is simulated with a short-cut method since no membrane reactor module is available in the Aspen unit models. A recycling loop is introduced to capture the intricate relationship between hydrogen removal and the shift in equilibrium. The hydrogen permeation is a complex function of the partial pressure difference across the membrane and is therefore modelled with a Fortran subroutine (see Equation 1). By assuming a certain permeability, membrane thickness and membrane area, the permeation rate can

be calculated with an iterative procedure. This modelling approach does not capture the change in flux along the length of the membrane tube, the average flux is therefore approximated by the logarithmic mean of the flux at the inlet and outlet of the reactor, following [24]<sup>6</sup>. This then determines methane conversion and hydrogen recovery.

#### 4.1.3 MODEL VALIDATION

The modelling method is simple and does not represent the system in all its complexities. For the objective of our study, it is crucial that the most important output conditions (HR and MC) determining the economic viability of the technology are accurately calculated at specified input conditions (of which the most important are: driving force across the membrane, load to surface ratio and membrane permeability). Model validation is therefore done with the comprehensive mathematical model of Lin et al. [14]. With the same operating conditions<sup>7</sup>, a MC of 71.4% and a HR of 68.5% are obtained. According to their extensive mathematical model the results should be a MC of 70% and a HR of 68%. These results indicate that methane conversion and hydrogen recovery are modelled correctly, possibly somewhat on the high side.

Since this comparison does not imply that the modelling is also done correctly under different conditions we have further verified the model by comparing our main results with those of more detailed modelling and experimental studies that have included kinetics and variation along the length of the reactor. It should be emphasised that it is difficult to make a detailed comparison with other studies as different assumptions have been made on membrane type, membrane permeability, retentate and permeate partial pressure, load to surface ratio, use of sweep gas, sweep gas flow rate, etc. Different assumptions for these variables can significantly change the main output conditions of the MR as shown in Figure 5.

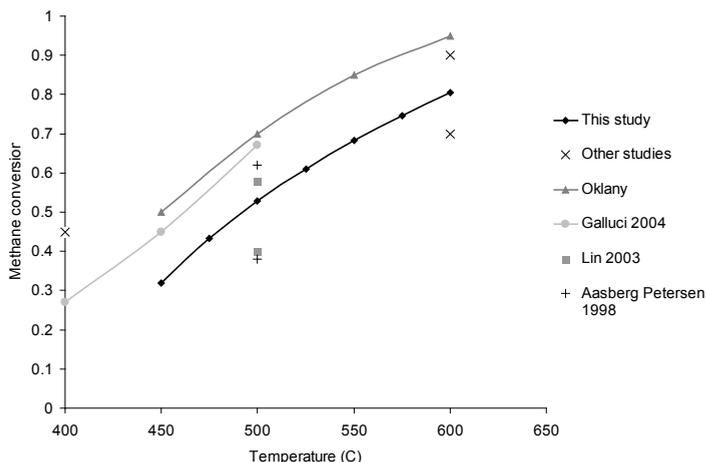


FIGURE 5. Methane conversion dependence on temperature (other studies are: [12,13])

6. The driving force  $\delta$  (see Equation 1) is then calculated with the following formula:

$$\delta = \frac{\delta_{in} - \delta_{out}}{\ln(\frac{\delta_{in}}{\delta_{out}})}$$

where  $\delta_{in}$  = driving force at input end of tube ( $\text{Pa}^{0.5}$ ) and  $\delta_{out}$  = driving force at exit end ( $\text{Pa}^{0.5}$ ).

$\delta_{in} = (p_{r,in})^{0.5} - (p_{p,in})^{0.5}$   
 where  $p_{r,in}$  = partial pressure on the retentate side at the input end (Pa) and  $p_{p,in}$  = partial pressure on the permeate side at the input end (Pa).  
 $\delta_{out} = (p_{r,out})^{0.5} - (p_{p,out})^{0.5}$   
 where  $p_{r,out}$  = partial pressure on the retentate side at the exit end (Pa) and  $p_{p,out}$  = partial pressure on the permeate side at the exit end (Pa).

7. Load to surface ratio of  $0.05 \text{ kmol/h m}^2$ , permeability of  $0.7 \text{ mol m}^{-2} \text{ h Pa}^{0.5}$ ,  $500^\circ\text{C}$ , reaction pressure 20 bar and permeate pressure 1 bar.

The values calculated by the model and the dependency of  $mc$  to temperature seem to coincide well with more detailed studies. The differences can mainly be explained by membrane permeability, use of a sweep gas and partial pressure difference between the two sides of the membrane. The temperature dependence is similar to that found by Oklany [12], but is lower due to the lower sweep to feed ratio. The model also shows a correct response to the effect of other operating conditions such as pressure and steam-to-carbon ratio (not shown). In conclusion, the obtained values for methane conversion and hydrogen recovery for specified conditions are broadly in accordance with literature. This comparative analysis indicates that the modelling approach is sufficiently accurate for preliminary efficiency calculations and to assess the economic viability of the total system (see for example also the approach taken by Jordal in [58]). It should, however, be stressed that for an exact assessment of the thermodynamics of the MR and determining the optimal operating conditions for different configurations, more elaborate models are essential such as those already developed by e.g. Adris et al. [7].

#### 4.1.4 MODELLING ASSUMPTIONS

All modelling assumptions are given in Table 1. The energy input to the system is minimised by utilising the heat of the output streams with several heat exchangers. The main intention is to select a realistic configuration in which most of the heat can be re-used without the cost for additional heat exchangers exceeding the cost savings of reduced fuel input. Heat recovery could be optimised with a pinch analysis, but that is beyond the scope of this study and in addition may give too ideal results.

TABLE 1. Unit and stream modelling assumptions used in Aspen<sup>plus</sup> simulation <sup>a</sup>

##### General

Heat Exchangers	$\Delta p/p = 2\%$ <sup>b</sup> Minimum $\Delta T = 15^\circ\text{C}$ (gas-liquid) or $30^\circ\text{C}$ (gas-gas) <sup>c</sup> Heat at temperatures lower than $100^\circ\text{C}$ is not used <sup>d</sup>
Water pump <sup>d</sup>	Centrifugal pump $\eta = 0.9$ , $\eta_{\text{driver}} = 1$
Natural gas compressor	Multi-stage compressor with intercooling, 2-3 stages Outlet temperature does not exceed $150^\circ\text{C}$ <sup>e</sup> Maximum compression ratio is three $\eta_{\text{polytropic}} = 0.84$ <sup>f</sup> $\eta_{\text{mechanic}} = 1$
Hydrogen compressor	Multi-stage compressor with intercooling, 9 stages Maximum compression ratio is two <sup>g</sup> Outlet temperature does not exceed $135^\circ\text{C}$ <sup>h</sup> Outlet pressure is 482 bar $\eta_{\text{isentropic}} = 0.78$ <sup>i</sup> $\eta_{\text{mechanic}} = 1$
Carbon dioxide compressor	Multi-stage compressor with intercooling, 5 stages Intercooling at $32^\circ\text{C}$ <sup>j</sup> Outlet pressure is 100 bar $\eta_{\text{isentropic}} = 0.78$ <sup>k</sup> $\eta_{\text{mechanic}} = 1$
Condenser	Air-cooled

*Pre-treatment*

HDS/Guard bed unit	Natural gas needs to be preheated to 380°C <sup>l</sup> Modelled as a valve for pressure drop <sup>m</sup> $\Delta p = 1\%$ <sup>n</sup>
--------------------	--

*Reforming*

Reactor	Gibbs free energy minimisation reactor Reactor operates at $T = 500\text{--}600^\circ\text{C}$ Pressure drop is modelled by valve in retentate stream, $\Delta p = -0.5\text{ bar}$ <sup>o</sup> Steam-to-carbon ratio = 3
Membrane	Modelled as separator. Separates amount of hydrogen depending on permeation rate Hydrogen at 1 bar Membrane material = Pd <sub>23</sub> Ag with porous ceramic support Membrane thickness = 20 $\mu\text{m}$ Permeability = $2.25 \cdot 10^{-5}\text{ mol m/m}^2\text{ h Pa}^{0.5}$
Furnace <sup>p</sup>	Natural gas furnace provides heat for reaction, fuelled by retentate/purge gas. If this is not sufficient extra natural gas fuel is taken from the grid. Modelled as stoichiometric reactor. $T = 20^\circ\text{C}$ more than $T$ reactor. $\Delta p = -0.5\text{ bar}$ Fractional conversion = 100% No radiative heat loss Twice the stoichiometric amount of air

*Purification*

Water knock-out tank	Modelled as two outlet flash drum $Q = 0\text{ W}$ <sup>q</sup> $T_{\text{in}} = 30^\circ\text{C}$ <sup>r</sup> $\Delta p = -0.01\text{ bar}$ (1%) Over 99% of water is separated
Dehydration	Modelled as separator 100% water removal

*Stream composition (volume %)*

Natural gas <sup>s</sup>	$\text{CH}_4 = 81.1\%$ $\text{C}_2\text{H}_6 = 3.5\%$ $\text{C}_3\text{H}_8 = 0.6\%$ $\text{C}_4\text{H}_{10} = 0.5\%$ $\text{CO}_2 = 1.4\%$ $\text{N}_2 = 12.9\%$  $\text{LHV} = 32.42\text{ MJ/Nm}^3$ $\text{HHV} = 35.91\text{ MJ/Nm}^3$ $T = 15^\circ\text{C}$ , $p = 1.02\text{ bar}$
Air	$\text{N}_2 = 77.29\%$ $\text{O}_2 = 20.75\%$ $\text{H}_2\text{O} = 1.01\%$ $\text{Ar} = 0.92\%$ $\text{CO}_2 = 0.03\%$ $T = 15^\circ\text{C}$ , $p = 1\text{ bar}$
Water/Sweep gas	$\text{H}_2\text{O} = 100\%$ $T = 15^\circ\text{C}$ , $p = 1\text{ bar}$

<sup>a</sup> The simulation has been done using the Peng-Robinson equation-of-state.<sup>b</sup> [86].<sup>c</sup> [60].<sup>d</sup> Aspen default values [98].<sup>e</sup> [99].<sup>f</sup> [99].<sup>g</sup> According to guidelines of the American Petroleum institute, quoted by Myers et al.

[24]. The low compression ratio per stage reduces the hydrogen discharge temperature and increases the piston ring lifetime.

<sup>b</sup> According to guidelines of the American Petroleum institute, quoted by Myers et al.

[24].

<sup>i</sup> [60]; reciprocating compressors with compression ratios of two reach adiabatic efficiencies of 78% [99].

<sup>j</sup> [100].

<sup>k</sup> [60].

<sup>l</sup> [101].

<sup>m</sup> To convert sulphur compounds, 2-5% of the hydrogen in the product stream is recycled to ensure complete conversion [101]. The actual amount of hydrogen consumed in the process is insignificant and has therefore been neglected [102].

<sup>n</sup> [60].

<sup>o</sup> [86].

<sup>p</sup> [86].

<sup>q</sup> [60].

<sup>r</sup> [103].

<sup>s</sup> Average natural gas composition in the Netherlands for the period 19-11-2002 to 20-11-2003 [81].

8.  
All currencies in  
US\$2003.

#### 4.2. ECONOMIC ASSUMPTIONS<sup>8</sup>

Results from the Aspen modelling are directly coupled to a spreadsheet model to evaluate the economics of the different concepts on the basis of cost data at component level. The methodology is similar to previous analyses conducted [59,60]. It should be noted that preliminary feasibility analyses such as these typically have an accuracy of ± 30% due to limited cost data and design detail [61]. The component cost figures, as shown in Table 2, are obtained from literature sources, vendor quotes and personal communication with experts. Note that these values are only valid for specific scales; to calculate the cost at other capacities (as derived from the Aspen modelling) it is common practice in engineering economics to make use of the following scaling equation (R = scaling factor) [62].

$$\frac{Costs_A}{Costs_B} = \left( \frac{Capacity_A}{Capacity_B} \right)^R \tag{Equation 2}$$

The cost data are mainly derived from a detailed analysis recently conducted by Directed Technologies Inc. [24]. These have been generated for a small-scale steam reformer (0.2 MW) according to the Design for Manufacturing and Assembly methodology (DFMA). This approach is extensively used by industry (e.g. Ford Motor Company) for product cost estimation and determines the total component cost using a bottom-up method based on material and manufacturing cost (see for a detailed explanation [24]). Component costs have been calculated for a production volume of 250 units. This is justified as (presuming a hydrogen economy becomes a reality) many small-scale reformers will be necessary to meet demand<sup>9</sup> and it is expected that they will be pre-assembled at a production plant. These values only represent the free on board (FOB) cost. Hence, for total investment expenditure of a refuelling station, additional costs are required for system assembly at the production plant and for installation at the refuelling station (on-site) (see Table 3). These fractions are mainly taken from the above-mentioned study for small-scale reformers [24].

9.  
To put these figures into perspective: the Netherlands currently has about 4150 refuelling stations [63].

TABLE 2. Capital cost of system components <sup>a</sup>

Component	Base investment cost (000\$)	Base Scale	Scale factor
<i>Pre-reformer</i>			
Natural gas compressor	3.3	5 kw	0.82 <sup>b</sup>
Natural gas feed system	0.2	5 kw	0.82
Heat exchangers (steel) <sup>c</sup>	7.8	2 m <sup>2</sup>	0.59 <sup>c</sup>
Heat exchangers (stainless-steel) <sup>d</sup>	15.5	2 m <sup>2</sup>	0.59
Desulphurisation unit	4.8	0.79 kmol CH <sub>4</sub> /h	0.6 <sup>b</sup>
Water purification	2.1	90 l H <sub>2</sub> O/h	0.68 <sup>e</sup>
Water pump	1.2	90 l H <sub>2</sub> O/h	0.7 <sup>f</sup>
Waste-heat boiler	19.2	90 l H <sub>2</sub> O/h	0.67
Sweep gas boiler	19.2	90 l H <sub>2</sub> O/h	0.67
<i>Reformer</i>			
Combustion chamber (furnace)	2.1	4.79 kg H <sub>2</sub> /h	0.78 <sup>g</sup>
Membrane reactor (without membrane tubes) <sup>h</sup>	14.1	4.79 kg H <sub>2</sub> /h	0.7 <sup>i</sup>
<i>Post-reformer</i>			
Hydrogen compressor	22.0	4.79 kg H <sub>2</sub> /h	0.82
Condenser and water knock-out	4.9	4.79 kg H <sub>2</sub> /h	0.68
Carbon dioxide compressor <sup>j</sup>	3000	6 MW	0.7
Dehydration equipment <sup>k</sup>	35.0	1180 m <sup>3</sup> /h	0.7
Cryogenic CO <sub>2</sub> separation unit <sup>l</sup>	475.0	0.6 t CO <sub>2</sub> /h	0.7

<sup>a</sup> If not indicated, values are taken from Myers et al. [24]; Values have been adjusted for inflation using the GDP Deflator Inflation Index.

<sup>b</sup> [105].

<sup>c</sup> [106]; steel tube-shell heat exchanger.

<sup>d</sup> [106]; stainless steel tube-shell heat exchanger. If sweep gas is used, this type is required to prevent corrosion.

<sup>e</sup> Average value for general and heat exchange equipment [105].

<sup>f</sup> This is for a carbon steel positive displacement pump [105].

<sup>g</sup> Average of the values 0.7-0.85, quoted from different sources by Remer et al. [105].

<sup>h</sup> This includes the reactor core itself, the inlet and outlet headers and the assembly of the reactor. Additional expenditure to prevent the negative effect of thermal cycling has not been taken into account. SS316 is used instead of Haynes556.

<sup>i</sup> [105]; this is only for reactors in general, not specifically for the membrane reactor.

Average of the values 0.65-0.74.

<sup>j</sup> Atlas Copco [107].

<sup>k</sup> Taken from [108]; capital cost data are for a dehydration unit operating at 31 bar and 10°C. Cost for the glycol unit is 20,000 \$; increased with 75% to account for installation and engineering.

<sup>l</sup> Cost estimate from Union Engineering [47] is 450,000 - 500,000 \$ for this system; the average was taken. This represents installed cost.

TABLE 3. Installation factors

<i>Total facilities cost</i>	TFC
Instrumentation and control	13%
Structural support	5%
Assembly	5%
Piping	2%
<i>Total system assembly<sup>a</sup></i>	25%
<i>Total plant cost (TPC)</i>	1.25·TFC
Site preparation	0.5%
Assembly on-site	10%
Tax, insurance and freight	2%
Engineering	5%
Contingency	10%

Fees, overhead and profit	10%
Start-up cost	5%
<hr/>	
<i>Total installation<sup>b</sup></i>	42.5%
<i>Total capex</i>	1.425·TPC

<sup>a</sup> These figures are based on Myers et al. [24], but instead of the actual cost, the percentage of the TFC is taken to enable calculation at different capacities.

<sup>b</sup> Based on Perry et al. [104] and Myers et al. [24].

Due to the experimental status of the MR no cost estimates were found for the membrane reformer itself. It is assumed that the basic reactor configuration resembles a normal SMR, but since operating temperature is lower less expensive materials can be used. For example, an SMR requires high quality steel alloys such as Haynes 556 [24], whereas the MR can suffice with stainless steel types such as 304 or 316 SS [64], which cost a factor ten less [65]. The membrane tubes are an additional cost factor, which is dependent on membrane surface, raw material price and production cost. Johnson Matthey, one of the leading producers of palladium membrane tubes, have stated that at current prices raw material constitutes 40% and tube manufacturing 60% of total production cost<sup>10</sup> [34]. The exact cost of tube production also depends on the type of support. A porous metallic support is more expensive than ceramic, but has the advantage of simple connections and easy noticeable cracks. According to expert knowledge, our assumption is on the high side of costs for ceramic support (including intermediate layers) and slightly on the low side for metallic support [66]. Production cost could be significantly reduced when machine assembly becomes an option. The annual investment cost is calculated by multiplying the total Capex with an annuity factor<sup>11</sup>. The economic parameters used are given in Table 4. Other costs such as raw materials and utilities form the annual operational expenditure. It is assumed that besides maintenance tasks the reformer can operate unattended and labour costs can therefore be omitted. Property taxes, insurance and o&m are taken as a percentage of the total installed investment. Electricity and natural gas prices supplied from the grid are based on prices for small industrial users in the EU according to the annual natural gas and electricity consumption of the reformer [67].

Other operational expenditure consists of the cost to replace catalysts, deionisation bed resin, ZnO adsorbent and membrane tubes. The cost of membrane tube replacement is a rather uncertain factor. Demonstration projects have shown lifetimes ranging from several thousand hours [68] up to more than 15 months [69] after which the membrane was rendered unusable. This was caused by leaks in the membrane due to insufficient mechanical stability. It is expected that long-term performance stability will be improved considerably. As we are concerned with a medium-term estimate, we follow the economic feasibility analysis of Criscuoli et al. [18] and assume a membrane lifetime of three years (21,000 hrs operation). The membrane material itself is not modified by operation and can therefore be recycled (already done by MRT [69]). When replacing the membrane tubes we assume it is feasible to recover 60% of the palladium<sup>12</sup> [70] and we assume the costs to be negligible [64]. As a

10. Membrane tube cost are thus calculated by multiplying the required surface area with raw material cost and then multiplied by a factor of 2.5 to account for tube manufacturing (1.5·raw material cost).

11.

$$\alpha = \frac{d}{1 - \frac{1}{(1+d)^n}}$$

with ( $\alpha$ =annuity factor,  $d$ =after tax rate of return,  $r$ =after tax real rate of return,  $i$ =inflation,  $n$ =lifetime).

12.

Confidential data showed that spent material can be sold back to the market at competitive prices.

comparison, the average recycling efficiency of palladium and platinum in the chemical and petrochemical industry is 76% [71].

TABLE 4. Cost parameters

<i>Investment</i>	
Lifetime	15 yrs
After-tax real rate of return	10%
Inflation rate	2%
Annuity factor	15.27%
<hr/>	
<i>Operational expenditure</i>	
Annual load	80%
O&M	4%
Property taxes	2%
Insurance	1.5%
Natural gas price	4.18 \$/GJ
Electricity price	0.07 \$/kwh
Palladium price	6752 \$/kg
Silver price	203 \$/kg
Water price	0.282 \$/m <sup>3</sup>
<hr/>	
<i>Replacements</i>	
Hydrosulphurisation catalyst	35.8 \$/l
Zinc oxide adsorbent	16.6 \$/l
Deionisation-bed resin	1500 \$/yr
Reforming catalyst	1113 \$/kg

#### 4.3. OPTIMAL PROCESS CONDITIONS

Since many parameters affect the thermodynamic and economic performance of the MR, an extensive analysis is performed to determine the most favourable conditions at which the MR should operate. For the aim of our study, we are mainly concerned with the most economically viable option. The optimal conditions are therefore those with the lowest hydrogen production cost and consequently do not have to coincide with the best thermodynamic conditions.

As the Aspen results are directly coupled to a spreadsheet calculating the economics, the effect of a change in process conditions on the hydrogen production cost can immediately be evaluated. By changing one parameter at a time, the conditions at which the hydrogen cost is lowest can be determined. Table 5 shows the range of values that is investigated in this study. In the case of the ideal configuration with integrated CO<sub>2</sub> capture, the optimal conditions not only need to ensure the production of hydrogen at lowest cost, but also need to comply with the fact that the retentate stream should contain as little contaminants as possible.

TABLE 5. Range of operating conditions investigated in this study

Process conditions	Investigated range <sup>a</sup>
Temperature (°C)	475 – 600
Pressure (bar)	2.5 – 60
Steam/carbon ratio (-)	2.5 – 3.5
Load/surface ratio (kmol/h m <sup>2</sup> )	0.03 – 0.50
Sweep gas ratio (-)	0.25 – 5

<sup>a</sup> Derived from literature values: [4,6,7,8,12,13,14,15,16,19,21,32,35,38,56,72,73,74,75,76,77]. The possible negative influence of operating at a slightly higher temperature and higher pressure, such as faster membrane degradation, and possibly the use of different materials has not been taken into account.

## 5 SYSTEM CALCULATION RESULTS

### 5.1 RESULTS OPTIMAL PROCESS CONDITIONS

The process parameters with the lowest hydrogen production cost are shown in Table 6 for all configurations. Although the conditions correspond well to other experimental and modelling studies<sup>13</sup> (as explained in Section 2.3) there could be small variations due to the specific modelling method and assumptions. Differences in the latter are caused by local conditions such as natural gas and electricity prices, specific configurations chosen related to heat production, heat transfer, reformer geometry or membrane deterioration. The optimal conditions for the non-ideal configuration are equal to those for the non-sweep since these give the lowest overall hydrogen production cost and a high purity retentate stream is not required due to the cryogenic CO<sub>2</sub> separation unit.

As expected, the sweep configuration can operate at lower temperatures and lower pressures than the non-sweep and ideal configuration since the driving force across the membrane is increased by the sweep flow. A higher sweep gas ratio than the optimum (0.5) results in higher hydrogen recoveries and methane conversion, but this does not outweigh the higher cost of steam generation and water removal. This is also the case for the configuration with integrated CO<sub>2</sub> capture (ideal) in which a higher temperature and a higher pressure is more economic than an increase in the sweep flow rate.

Table 6 also shows the retentate gas composition for the selected systems. The modelling results for the ideal configuration confirm the high purity of CO<sub>2</sub> in the retentate gas. The high nitrogen concentration is a direct consequence of the high nitrogen content of Dutch natural gas. It is expected that this does not pose any problems as the mixture of natural gas and nitrogen is already distributed in pipelines. The latter argument also applies to the small methane and CO concentration. In contrast, hydrogen has negative implications as it results in pipeline embrittlement. Research has recently been conducted into the effect of a methane/hydrogen mixture on natural gas pipelines and preliminary results have shown that a concentration of up to three volume percent is possible without any additional investment in the network [78]. Taking these issues into account, we can conclude that it is very likely that the retentate stream can be immediately transported.

For a 2 MW hydrogen production plant, the amount of CO<sub>2</sub> in the exhaust gas is approximately 2.68 kmol/h compared to 6.45 kmol/h in the retentate stream. This means that a mere 71% of the carbon dioxide produced in the process can be recovered for storage purposes. Moreover, recovery efficiency with separation technologies is only

13.  
[21,12,4,72,16,14,13,73,  
8,74,6,75,35,56,38,76,  
32,77,15,7,19]

about 80-90% and therefore, assuming 80% recovery, only 57% of the total CO<sub>2</sub> produced in the process can be captured.

TABLE 6. Optimal conditions for the membrane reactor

Process conditions	Optimal conditions		
	Sweep	Non-sweep/Non-ideal	Ideal
Temperature (°C)	550	575	600
Pressure (bar)	15	25	25
Steam/carbon ratio (-)	3	3	3
Load/surface ratio (kmol/h m <sup>2</sup> )	0.083	0.094	0.083
Sweep gas ratio (-)	0.5	-	0.5
Retentate gas (%mol)			
CH <sub>4</sub>	3.2%	4.5%	3.6·10 <sup>-4</sup> %
H <sub>2</sub>	6.3%	6.6%	1.0%
H <sub>2</sub> O	39.1%	40.7%	35.6%
CO <sub>2</sub>	42.8%	39.6%	55.0%
CO	1.9%	2.1%	0.6%
N <sub>2</sub>	6.6%	6.4%	7.8%

## 5.2 THERMODYNAMIC PERFORMANCE

Table 7 summarises the thermodynamic performance of the MR. The figures are equal for the capacity range studied (0.2-10 MW) as nothing is assumed to change in the configuration (although heat losses may be lower and compression more efficient at larger scales). The obtained results of around 89-92% methane conversion and 96% hydrogen recovery<sup>14</sup> are on the high-end of values given in literature. A total efficiency<sup>15</sup> of about 77.5% (HHV) for the configurations without CO<sub>2</sub> capture can be attained. Practical results with the MR are obtained from Membrane Reactor Technologies, 82% HHV [11] and Tokyo Gas Company, 70% [79]. The efficiency value of MRT includes hydrogen compression, which compares to 81.6% for the modelled MR with the same compression requirement, almost equal to the value given by MRT. The efficiency of a conventional SMR (without H<sub>2</sub> compression) is in the range of 75-80% [24,80]. The total efficiency of the MR without any hydrogen compression, 83.9%, is thus higher than conventional technology.

The total efficiency for the sweep configuration is 0.5%pt lower than non-sweep. The additional energy required for steam generation is thus not completely compensated by the higher MC and the lower temperature and pressure at which the system operates. For the ideal configuration, the efficiency loss is due to the different conditions at which the reactor needs to operate, 0.2%pt, and CO<sub>2</sub> compression, 0.3%pt. For the cryogenic option, no adjustment is required for the configuration, but efficiency loss is a consequence of the electricity needed for cooling, pumping and compression.

Efficiency increase could be established in a number of ways. Firstly, optimising heat flows can induce a small improvement. Secondly, the compressor performance can be improved (10% less electricity consumption results in a 0.5%pt efficiency increase). This will not be substantial as assumed performance is already optimistic and no

14. The HR mentioned here is higher than in Section 4, because of different definitions (see Section 2.1 for the definition applied to results in this chapter).

15. Defined as hydrogen energy output divided by energy input (fuel, feed and electricity).

16. The pressure of the grid to which many industrial users are connected [81].

significant advances are expected. Thirdly, natural gas supplied at 16 bar<sup>16</sup> instead of 1.02 bar reduces compressor power such that the total efficiency is raised with 1%pt. Fourthly, efficiency improvements could be realised by a higher membrane flux and larger membrane surface. The first can be achieved by higher permeability and is thus mainly dependent on further advances in membrane technology, but membrane assumptions are quite optimistic so significant improvement is not expected. Larger membrane surface would also cause an efficiency increase, but this would obviously have a negative impact on hydrogen production cost.

TABLE 7. Thermodynamic performance of the membrane reactor

	Literature range	Results Non-sweep	Sweep	Non-ideal	Ideal
Methane conversion (MC)	45%-99% <sup>a</sup>	88.7%	92.2%	88.7%	99.9%
Hydrogen recovery (HR)	70%-99%	95.8%	96.3%	95.8%	99.0%
<i>Mass balance (kmol/h)</i>					
Natural gas feed	-	8.2	7.8	8.2	7.1
Natural gas fuel	-	1.61.7	1.6	2.8	
Water	-	20.0	16.1	20.0	14.6
Air	-	53.7	48.2	53.7	56.4
Hydrogen produced	-	25.2	25.2	25.2	25.2
Retentate gas	-	16.5	15.3	16.5	12.2
Exhaust gas	-	71.2	64.6	71.2	59.3
<i>Energy balance</i>					
Total HHV efficiency	70%-82% <sup>b</sup>	77.8%	77.3%	76.5%	76.9%
Total HHV efficiency - primary energy <sup>c</sup>	-	70.5%	70.3%	68.1%	69.6%
<i>Internal energy use</i>					
Fuel use in furnace (GJ <sub>fuel</sub> /GJ <sub>H<sub>2</sub></sub> )	-	0.20	0.20	0.20	0.33
Electricity use (GJ <sub>e</sub> /GJ <sub>H<sub>2</sub></sub> )	-	0.11	0.11	0.13	0.11
Electricity use primary (GJ <sub>ep</sub> /GJ <sub>H<sub>2</sub></sub> )	-	0.24	0.24	0.29	0.24

<sup>a</sup> [6,12,13,14,16,20,40,79,109,110,111,112].

<sup>b</sup> [11,79]; without hydrogen compression.

<sup>c</sup> Electricity input calculated as primary energy using an efficiency of the grid of 45% [94].

17. According to the number of cars in the Netherlands: 7.7 million and the number of refuelling stations: 4153 [63], 1856 cars are supported by an average refuelling station. Assuming an annual travel distance per car of 16,100 km/yr [63], annual operation of 80% and fuel economy of 0.98 km/MJH<sub>2</sub>(HHV) (=80 mpge) [24], a production of 1.2 MW hydrogen would be required. Currently, only a fuel economy of 0.55 km/MJ has been achieved for fuel cell vehicles [82], which would require a production of 2.2 MW of hydrogen. Other studies have used capacities of up to 4 MW [83], mainly focussed on the US. Considering these different values we assume 2 MW as an intermediate capacity.

### 5.3 ECONOMIC PERFORMANCE

Results of the economic evaluation of a 2 MW capacity membrane reactor are given in Table 8 for all systems studied. This capacity represents the volume of hydrogen that would be required for a future hydrogen refuelling station<sup>17</sup>.

The results clearly illustrate that the capital expenditure for the different concepts increases as system complexity becomes greater. The non-ideal configuration is almost 30% more expensive than the non-sweep. The same holds for the annual cost, albeit in a lesser extent. The final hydrogen cost for the sweep configuration is slightly higher, 14.1 \$/GJ compared to 13.5 \$/GJ, illustrating that lower temperature and pressure do not outweigh extra costs related to the operation of a sweep gas.

TABLE 8. Economic analysis for the systems considered (capacity of 2 MW)

(000\$)	Non-sweep	Sweep	Non-ideal	Ideal
<i>Capital expenditure (capex)</i>				
Membrane tubes	232	251	232	235
Reformer vessel	89	89	89	89
Combustion chamber	14	14	14	14
NG feed system	1	1	1	1
NG compressor	16	11	16	12
NG heater	2	4	2	3
Air heater	6	13	6	12
Boiler	48	47	48	44
Desulphurisation unit	19	19	19	18
Water purification	5	3	5	5
Water pump	3	3	3	3
Condenser/Water knock-out	-	25	-	25
Condenser/Water knock-out	-	-	-	7
Hydrogen compressor	152	153	152	153
Sweep boiler	-	17	-	16
Dehydration equipment	-	-	-	1
CO <sub>2</sub> compressor	-	-	-	16
Additional heat exchangers	-	-	-	5
Cryogenic separation unit	-	-	164	-
<i>Total facilities cost (TFC)</i>	587	648	751	657
<i>Total plant cost (TPC)</i>	734	810	939	821
<i>Total capex</i>	1046	1155	1338	1170
<i>Annual cost</i>				
o&m	42	46	54	47
Methane fuel	42	56	42	72
Methane feed	203	194	203	176
Electricity	107	104	129	110
Water	1	<1	12	1
HDS catalyst replacement	10	10	10	9
Resin replacement	15	14	15	13
Membrane replacement	59	64	59	60
Reforming catalyst	5	5	5	5
Annual insurance and tax	37	40	47	41
Annual capital cost	162	179	207	181
<i>Total annual cost</i>	683	712	782	714
<i>Hydrogen cost</i>				
\$/kg	1.9	2.0	2.2	2.0
\$/GJ (HHV)	13.5	14.1	15.5	13.9
CO <sub>2</sub> cost (\$/t CO <sub>2</sub> separated)	-	-	49	14

The results clearly illustrate that the capital expenditure for the different concepts increases as system complexity becomes greater. The non-ideal configuration is almost 30% more expensive than the non-sweep. The same holds for the annual cost, albeit in a lesser extent. The final hydrogen cost for the sweep configuration is slightly higher, 14.1 \$/GJ compared to 13.5 \$/GJ, illustrating that lower temperature and pressure do not outweigh extra costs related to the operation of a sweep gas. When CO<sub>2</sub> capture is included, the hydrogen cost increases to 13.9 \$/GJ and 15.5 \$/GJ for the ideal and non-ideal situation respectively. Thus, the constraint of high purity CO<sub>2</sub> adds only 4% to the hydrogen cost in the

ideal situation and 14% in the case of a separation unit. If the hydrogen production cost is required to remain the same as for the non-sweep configuration (cheapest option), cost per tonne of CO<sub>2</sub> separated can be calculated. For the ideal configuration this is 14 \$/t CO<sub>2</sub>, three times less than with cryogenic separation, 49 \$/t CO<sub>2</sub>, and less than separation cost for large-scale (1000 MW) steam reformers: 21 \$/t CO<sub>2</sub> [84].

As illustrated by Figure 6, mainly the reformer (including membrane tubes) and hydrogen compressor determine the total capital cost and therefore a large part of the final hydrogen cost since capital recovery is responsible for about 22% of annual cost. The initial membrane tubes account for over one-third of capital expenditure and their replacement for 8% of annual cost. A cost reduction in membrane raw material or tube production can therefore have a significant effect on the final hydrogen cost. Natural gas and electricity consumption also influence cost noticeably; these inputs make up 34% (7% fuel and 27% feed) and 18% of annual costs, respectively.

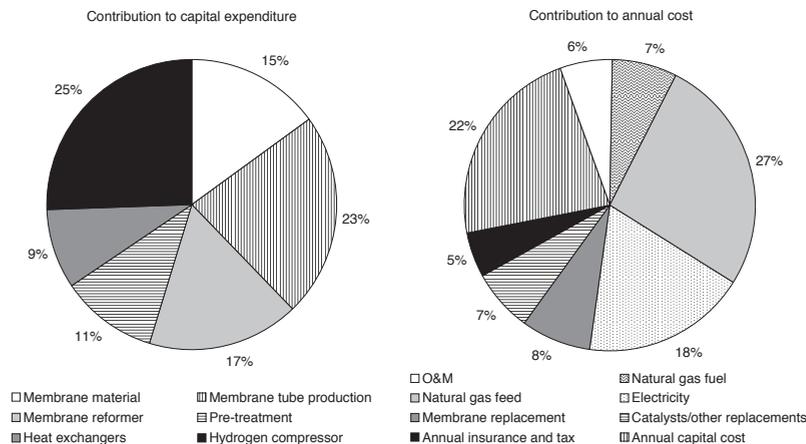


FIGURE 6. Contribution to capital expenditure and annual cost (2 MW non-sweep)

### 5.3.I TECHNOLOGICAL LEARNING

The capital cost of the membrane reactor could decline in the future due to technological learning. The effect of technological learning is commonly represented by experience curves<sup>18</sup>, which describe how capital costs decline with cumulative production. The learning rate (LR) is the characteristic parameter, describing the percentage reduction in unit price with every doubling of cumulative production. The cost reduction is a result of factors including: economies of scale due to upscaling of units, economies of scale due to the production of more identical units at one plant (mass production), process improvements, learning-by-doing, and reduction in raw material costs [85]. Figure 7 shows the effect of system upscaling on hydrogen cost as determined by the Aspen study. Although a reduction is certainly

18. An experience curve is generally described as:  $C_{cum} = C_0 \cdot CUM^b$  where  $C_{cum}$ =cost per unit,  $C_0$ =initial cost of unit,  $CUM$ =cumulative production over time and b is the experience index. The learning rate is defined as  $1-2^b$  [85].

present, the effect of upscaling is less than for conventional reforming owing to the modular nature of the MR. If the scaling equation is used (see Section 4.2) to calculate capital cost for other sizes, a scaling factor of 0.85 is obtained for the total system<sup>19</sup>. The configuration studied is for small-scale, but it is uncertain up to which capacity this scaling factor is applicable.

19. Compare this to scaling factors for SMR commonly used in literature: 0.6 [60] and 0.7 [86].

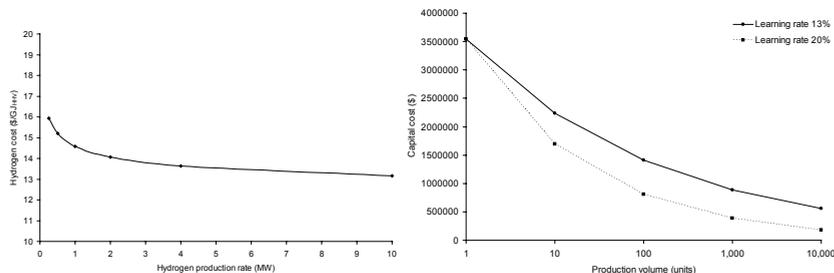


FIGURE 7. Effect of upscaling (left) and technological learning (right) of the MR (2 MW non-sweep)

To a certain extent, the effect of mass production is already incorporated in the prices since a production volume of 250 units is assumed. However, given that the technology is in an early stage of development, process improvement and learning-by-doing play a significant role as well. If these effects are included we can construct an experience curve as illustrated in Figure 7, using two learning rates: 13% and 20%. The first is representative for technologies such as wind turbines [87], whilst the latter is more applicable to modular systems such as PV solar panels [88]. No LR is available on hydrogen reforming technologies and it is therefore uncertain which would be most appropriate. Since the hydrogen reformer market can be regarded as a global market, cumulative production corresponds to global production. Considering that there are more than 4000 refuelling stations in the Netherlands alone, even with a low LR the potential exists for significant cost reductions (i.e. assuming hydrogen takes off as a transportation fuel). For instance, with a LR of 13% and only two doublings of cumulative production (250 to 1000), hydrogen cost can be reduced with 9% to 13 \$/GJ (2 MW non-sweep). One should note that this reduction is also possible for conventional technology, but as the technology is already more advanced the learning rate is less and further doublings require a larger market.

### 5.3.2 COMPARISON WITH SMR

The economic results cannot be compared to other estimates as almost none are available. Preliminary analyses of an MR have been reported by Aasberg-Petersen et al. [21], Roy et al. [20] and Middleton et al. [19], but these are unspecific and only consider large-scale membrane reformers and are therefore unsuitable for comparison. A good comparison however can be made with a conventional steam reformer.

20.  
 Load factor: 8585 hrs,  
 lifetime 10 yrs,  
 electricity price: 0.075  
 \$/kWh, gas price: 0.19  
 \$/m<sup>3</sup> and peak  
 capacity: 115 kg H<sub>2</sub>/h.  
 Storage, dispenser and  
 miscellaneous cost for  
 the refuelling station  
 have also been taken  
 equal. Hydrogen is  
 compressed to 482  
 bar.

The economic evaluation performed by Myers et al. [24] is chosen since component cost data used here is largely taken from that report. The same capacity and economic parameters are assumed<sup>20</sup>. The cost represents the wholesale hydrogen cost at a refuelling station, i.e. including hydrogen storage and dispensing cost as reported in [24]. Figure 8 illustrates that, within the error margin, the final hydrogen cost for the MR is competitive with the SMR and potentially even cheaper, mainly due to the higher efficiency of the reformer and the use of less expensive steel. It should be emphasised that this is for a very small-scale (0.2 MW). For larger scales, hydrogen production cost for a MR will be higher than for a SMR as upscaling has a more beneficial effect on the SMR.

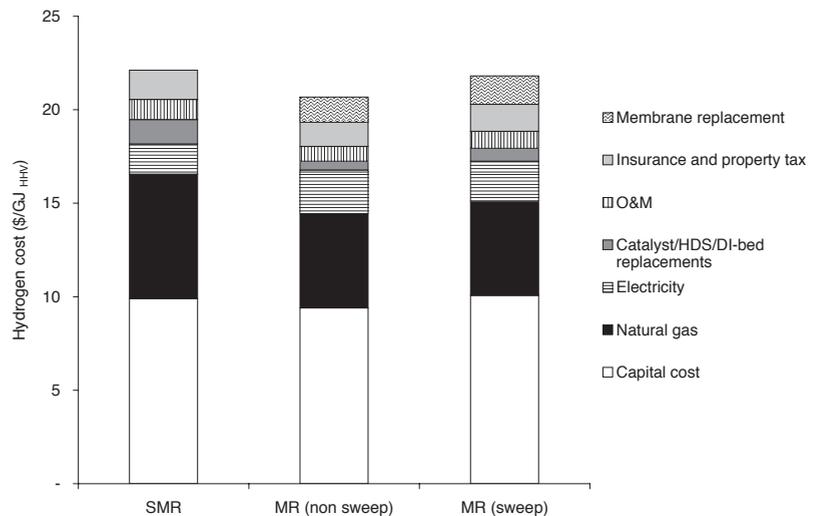


FIGURE 8. Hydrogen cost comparison with SMR (0.2 MW)

### 5.3.3 MONTE CARLO ANALYSIS

To evaluate the effect of potential uncertainties in the main assumptions a Monte Carlo analysis has been performed. The parameters are varied with ranges given in Table 9 according to a triangular distribution. According to the analysis, the mean hydrogen costs for the non-sweep configuration without CO<sub>2</sub> capture is around 14.5 \$/GJ and the median around 14.0 \$/GJ with a standard deviation of 3.1 \$/GJ. A certainty level of 55% was calculated for the range between 12 and 16 \$/GJ. Given the uncertainty in the assumptions, the most probable value is higher than the default result, which should be borne in mind when evaluating the MR against competitive alternatives.

Figure 9 shows that the cost is most sensitive to membrane thickness. Reducing membrane thickness to 5 μm lowers hydrogen cost to under 12 \$/GJ, but it can easily approach 20 \$/GJ when a thickness larger than 30 μm is used. With a membrane lifetime currently obtained by MRT (only 15 months), cost would reach 16 \$/GJ and the impact of a lower load factor illustrates the importance of matching reformer capacity

and expected demand. Related to a refuelling station it is hence of utmost importance to continuously operate the reformer and have sufficient storage capacity.

The effect of the palladium price should also be emphasised due to the extreme volatility of its market price. In the last few decades, it has fluctuated from 1600 to 32,000 \$/kg [89], even more than the range assumed here, which already results in a deviation of 2 \$/GJ to the default value. Natural gas and electricity prices are assumed for small-scale industrial users. Hence, long-term contracts with suppliers for more than just one refuelling station could greatly decrease production cost. However, it can be doubted whether natural gas prices drop again to levels below 4 \$/GJ considering recent increase in oil and hence gas prices (even for large industrial users).

TABLE 9. Ranges used for Monte Carlo analysis

	Default value	Range
Membrane thickness	20 μm	5-35 μm
Natural gas price	0.15 \$/m <sup>3</sup>	0.07-0.23 \$/m <sup>3</sup>
Palladium price	6752 \$/kg	2888 -10,615 \$/kg
Electricity price	0.07 \$/kwh	0.03-0.11 \$/kwh
Load factor	80% (7008 hrs)	67-93%
Lifetime membranes	3 yr	1.3-4.7 yr
Membrane production cost	1.5 · raw material cost	0.9 – 2.1 · raw material cost
Discount rate	13%	8-18%

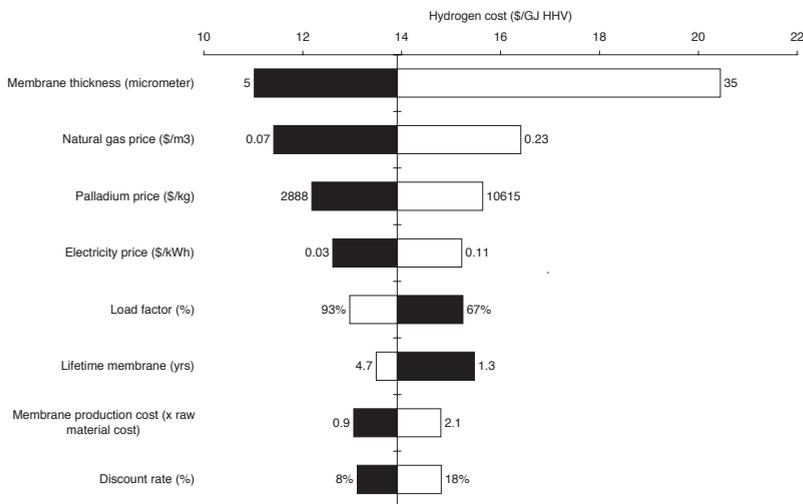


FIGURE 9. Sensitivity analysis non-sweep MR (2 MW) - parameter ranges are given in the figure

## 6 FUEL SUPPLY CHAIN ANALYSES

### 6.1 METHODOLOGY AND ASSUMPTIONS

In this section, the membrane reactor is evaluated in relation to other natural gas-fired technologies for a future hydrogen transportation infrastructure. The calculations in this section are mainly to provide an initial insight into small-scale CO<sub>2</sub> capture and to examine the trade-off between the various fuel supply chains. The results should therefore be regarded as a first indication of the total fuel supply costs.

To enable a correct comparison between the different technologies, they need to be investigated on a well-to-wheel basis. That is, the total fuel supply chain from extracting the primary fuel to the utilisation of the hydrogen in a vehicle needs to be taken into account.

For the energy requirements, a spreadsheet model developed at Imperial College London is used [90]. The model can simulate any technically feasible hydrogen infrastructure with the infrastructure configuration and demand being defined by the user. The energy requirement per infrastructure component is an output and is subsequently used in another spreadsheet model to estimate the total costs required for installing and operating the infrastructure.

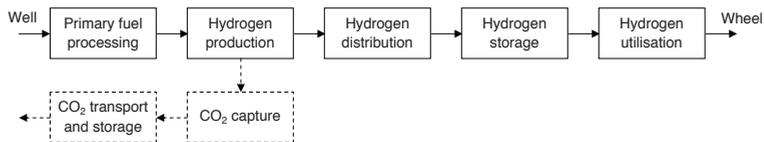


FIGURE 10. Well-to-wheel chain including CO<sub>2</sub> sequestration

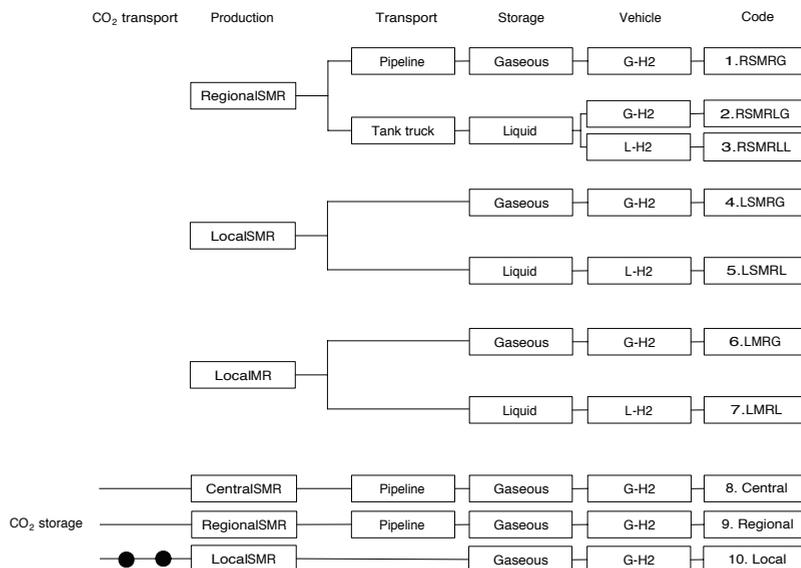


FIGURE 11. Overview fuel supply chains

Figure 10 gives a representation of the well-to-wheel chain for the hydrogen transportation sector and Figure 11 shows all chains investigated. The primary fuel (in our case natural gas) is extracted, processed and distributed to a reformer where hydrogen is produced. Three reformer capacities are considered: a central unit of 400 MW, a regional unit of 40 MW and a local unit at a refuelling station of 2 MW. The hydrogen can be produced as a gas (G-H<sub>2</sub>) or as a liquid (L-H<sub>2</sub>). Hydrogen produced at centralised reformers (40 and 400 MW) needs to be distributed to the refuelling stations, either in gaseous form through pipelines or as a liquid with tank trucks. Subsequently, the hydrogen is stored and dispensed to a vehicle, running on gaseous or liquid hydrogen.

The production of hydrogen can also be done with CO<sub>2</sub> capture, followed by transport and storage of the CO<sub>2</sub>. For the small-scale SMR, cost of CO<sub>2</sub> sequestration is prohibitive [3] and is therefore omitted. The same applies to the non-ideal MR configuration (with cryogenic separation) as cost is more than three times that of the ideal configuration. Since sequestration cost is equal for both states of hydrogen, only gaseous is chosen. The option is therefore researched for the MR (2 MW ideal) and for the regional and central SMR (40 and 400 MW).

The distribution infrastructure is shown in Table 10 with all branches originating at the reformer. The infrastructure is based on hydrogen supply to 200 refuelling stations of 2 MW hydrogen demand each and distances are based on the Netherlands. The calculations for the required pipeline diameter are done using steady-state flow equations, given a certain input pressure and pressure drop. The calculated pipeline diameters correspond well with the values given in Ogden [92] and MIT [93].

Regarding the carbon dioxide infrastructure, it is assumed that a large-scale CO<sub>2</sub> grid is already in place, as illustrated in Figure 12. The volumetric flow assumed through the third branch is therefore much bigger than when only hydrogen production plants are considered. We assumed a wellhead pressure of 80 bar. Hence, compression at the plant is done up to 90 or 100 bar depending on the distance that needs to be covered. Table 11 shows the costs and energy requirements of the various components for the fuel supply chain analysis. The lifetime of all components is set at 15 years, except for transport components (pipelines 22 yrs and trucks 10 yrs [95]). In agreement with other infrastructure estimates, costs do not include land cost and labour at the refuelling station. Consistent with the market, utility prices for centralised production are lower than for decentralised. For large-scale production 3.3 \$/GJ natural gas and 0.05 \$/kWh electricity are assumed.

TABLE IO. Pipeline infrastructure assumptions

Scale	H <sub>2</sub> infrastructure		CO <sub>2</sub> infrastructure		
	Regional	Central	Local	Regional	Central
<i>1<sup>st</sup> branch</i>					
Input pressure (bar)	60	70 <sup>a</sup>	100	100	90
Flow	-	400 MW	2.2 kt CO <sub>2</sub> /yr	-	-
Distance (km)	-	50	10	-	-
Number of pipelines	-	1	200	-	-
Pressure drop (bar)	-	10	1	-	-
Diameter (m)	-	0.25	0.03	-	-
<i>2<sup>nd</sup> branch</i>					
Flow	40 MW	40 MW	44 kt CO <sub>2</sub> /yr	44 kt CO <sub>2</sub> /yr	-
Distance (km)	30	30	30	30	-
Number of pipelines	10	10	10	10	-
Pressure drop (bar)	10	10	5	5	-
Diameter (m)	0.10	0.10	0.08	0.08	-
<i>3<sup>rd</sup> branch</i>					
	To refuelling station		To storage site		
Flow	2 MW	2 MW	5 Mt CO <sub>2</sub> /yr	5 Mt CO <sub>2</sub> /yr	5 Mt CO <sub>2</sub> /yr
Distance (km)	10	10	100	100	100
Number of pipelines	200	200	1	1	1
Pressure drop (bar)	1	1	10	10	10
Diameter (m)	0.05	0.05	0.50	0.50	0.50

<sup>a</sup> After [113]; For the regional reformer, input pressure has been assumed 60 bar to ensure the same output pressure at the refuelling station as for the central reformer.

TABLE II. Parameters for the fuel supply chain equipment

Component	Efficiency or Energy consumption	Base cost (000\$)	Base Scale	Scale factor or Engineering equation	O&M <sup>a</sup>
<i>Reforming</i>					
Regional/central SMR plant	85% <sup>b</sup>	13,844	24.4 tpd	455,820-x + 2,721,841 <sup>c</sup>	4% <sup>d</sup>
Local SMR plant	80% <sup>e</sup>	233 <sup>f</sup>	0.12 tpd	0.7 <sup>g</sup>	4%
Local MR plant	82% <sup>h</sup>	829 <sup>i</sup>	0.97 tpd	0.85 <sup>j</sup>	4% <sup>k</sup>
<i>Compression</i>					
Hydrogen compressor	78% <sup>l</sup>	30 <sup>m</sup>	10 kw	0.6	4%
<i>Liquefaction</i>					
Central hydrogen liquefier	0.848 kWh/Nm <sup>3</sup> H <sub>2</sub> <sup>n</sup>	33,432	24.4 tpd	-1315-x <sup>2</sup> + 1,146,035-x + 5,500,940 <sup>o</sup>	3%
On-site hydrogen liquefier	0.848 kWh/Nm <sup>3</sup> H <sub>2</sub>	4455 <sup>p</sup>	4.5 tpd	0.68 <sup>q</sup>	3%
<i>Storage</i>					
Gaseous storage <sup>r</sup>	-	66 <sup>s</sup>	79 kg	0.65 <sup>t</sup>	2%
Liquid storage <sup>u</sup>	-	103	3.1 t	17,221-x + 49,932 <sup>v</sup>	2%
<i>Dispensing</i>					
On-site pump/vaporiser <sup>w</sup>	0.063 kWh/Nm <sup>3</sup> H <sub>2</sub> <sup>x</sup>	171	1.2 tpd	-8670-x <sup>2</sup> + 128,085-x + 29,566 <sup>y</sup>	3%

Gaseous dispenser	- <sup>z</sup>	137	1.2 tpd	99,652·x + 17,785 <sup>aa</sup>	4%
Liquid dispenser	-	127	1.2 tpd	93,880·x + 14,339 <sup>bb</sup>	4%
<i>Transport</i>					
Pipelines	-	-	-	0.827·1.5· diameter(m)· length (km) <sup>cc</sup>	2.1%
Truck transport <sup>dd</sup>	0.39 l diesel/ km <sup>ee</sup>	545	1 truck	-	2%
<i>CO<sub>2</sub> capture and storage<sup>ff</sup></i>					
CO <sub>2</sub> capture (chemical absorption) <sup>gg</sup>	90% CO <sub>2</sub> capture efficiency <sup>hh</sup>	27,270 <sup>ii</sup>	42 t CO <sub>2</sub> /h	0.6 <sup>jj</sup>	
CO <sub>2</sub> compressor	78% <sup>kk</sup>	3000 <sup>ll</sup>	6 MW	0.67 <sup>mm</sup>	4%
CO <sub>2</sub> pipelines	-	-	-	0.827· diameter(m)· length (km) <sup>nn</sup>	2.1% <sup>oo</sup>
CO <sub>2</sub> storage <sup>pp</sup>	-	3.47	\$/t CO <sub>2</sub>	-	-
<i>Refuelling station</i>	-	1500 <sup>qq</sup>	-	-	-

<sup>a</sup> Unless stated otherwise, all o&m percentages are taken from [102]. Values are given in percentages of total capital investment.

<sup>b</sup> At present, the efficiency of large-scale reformers can vary from 76-85%<sub>HHV</sub> [23,114,115] depending on whether steam export is taken into account. The highest value is taken representing efficiency gains expected for the medium term. The output pressure of H<sub>2</sub> after the reformer and prior to the compressor is assumed 25 bar [23,116,117].

<sup>c</sup> The engineering equation is a linear fit to costs given by [24,118,119,120]. This equation is practically identical to a different engineering equation for large-scale SMR quoted by [121].

<sup>d</sup> This includes costs for labour, chemicals etc. [102].

<sup>e</sup> The efficiency of small-scale reformers ranges currently from 70 to 80%<sub>HHV</sub> [24,114,80]. The highest value is also taken here representing efficiency gains expected for the medium term. The output pressure of H<sub>2</sub> after the reformer and prior to the compressor is assumed 9 bar, after [24,110]. It is more beneficial to operate a small-scale SMR at lower pressure than a large-scale SMR.

<sup>f</sup> Taken from [24]. This estimate has also been based on a production volume of 250 units and can thus be compared to the MR. The cost is for the reformer increased with 42.5% to account for installation, contingency and tax & insurance.

<sup>g</sup> [86].

<sup>h</sup> Taken from the Aspen analysis, efficiency excluding compression (see Section 5.2). Hydrogen outlet pressure is 1 bar prior to compression.

<sup>i</sup> Total capex determined by the economic analysis in Section 5.3, excluding hydrogen compressor.

<sup>j</sup> Calculated in Section 5.3.

<sup>k</sup> Normal o&m is 4%. An extra 5% needs to be included for membrane replacement.

<sup>l</sup> [60,99]

<sup>m</sup> Confidential data for a multi-stage compressor. Cost given together with a scaling factor of 0.6 [105] gives a good fit to multi-stage compressor costs mentioned in other studies [1,24,122]. Cost reduction of 40% is assumed since production volume is 250 units.

<sup>n</sup> Liquefying plants are available in capacities of 1 to 60 tpd, supplied by e.g. Linde Gas and Air Liquide [123]. The energy required for liquefaction has a range of 10.1-15 kWh/kg hydrogen produced [1,124]. The lowest estimate is assumed to represent efficiency in the medium term (10.1 kWh/kg = 0.8484 kWh/Nm<sup>3</sup>).

<sup>o</sup> Polynomial fit to cost data taken from [1,102,119,123].

<sup>p</sup> Cost estimate for the smallest capacity liquefier has been chosen [123]. For consistency reasons the capital cost of the small liquefaction unit has also been calculated for a production volume of 250 units with a learning rate of 86%. Methodology is described in [119] and the learning rate is equal to other renewable energy technologies [24].

<sup>q</sup> Average scaling factor for general equipment assumed [105].

<sup>r</sup> Gaseous storage at 482 bar at the refuelling station. Storage capacity required is 70% of daily demand assuming a daily demand profile as given in [119] and that 58% of the hydrogen cannot be recovered with cascade dispensing [52]. Storage at the refuelling station after pipeline transport may be less as it is immediately taken from the grid. At

- the central plant storage capacity is 0.5 times the daily production [125].
- <sup>s</sup> [24]. The cost may be reduced considerably when composite materials are utilised.
- <sup>t</sup> Scaling factor for horizontal pressurised storage tanks [105].
- <sup>u</sup> At the centralised plant: 0.5 times the daily production [113]; Liquid H<sub>2</sub> boil-off is assumed 0.03%/day [123]. At the refuelling station, storage is one time the daily demand to enable one daily truck delivery.
- <sup>v</sup> Engineering equation is a linear fit to cost data reported by [95,102,118,126].
- <sup>w</sup> Nominal capacity of all dispensers is 0.25 tpd.
- <sup>x</sup> [127].
- <sup>y</sup> Engineering equation is a polynomial fit to cost data reported by [1,118,126].
- <sup>z</sup> Gaseous cascade dispensing and liquid dispensing requires very little energy and has therefore been neglected.
- <sup>aa</sup> Equation is a linear fit to data from [24,118,119,126]; There is remarkable agreement between the various literature estimates.
- <sup>bb</sup> Linear fit to data from [118,119,126].
- <sup>cc</sup> Pipeline cost is taken from [93]. This estimate is based on empirical data of more than ten years for the capital cost of natural gas pipelines. It is then multiplied by a factor of 1.5 to account for hydrogen instead of natural gas, following [92]. Hydrogen pipelines need higher quality material to prevent hydrogen embrittlement.
- <sup>dd</sup> Truck transport is calculated by round-trip, i.e. two-way to deliver one load. Total truck and o&m cost taken from [95]. Diesel price is 0.8 \$/l (EU average 2001-2003 [96]), drivers wage is 31.5 \$/h and average speed is 40 km/h [95].
- <sup>ee</sup> Diesel consumption taken from [95]. Trucks have a capacity of 56,520 liters of L-H<sub>2</sub> [128] and the boil-off assumed is 0.4 %/h [123]. During transfer of the hydrogen from tank to truck and vice versa some hydrogen may be lost as well [95], but this has been neglected.
- <sup>ff</sup> All cost for CO<sub>2</sub> sequestration are given in t CO<sub>2</sub> captured.
- <sup>gg</sup> The separation technology assumed is a chemical absorption unit using monoethanolamine (MEA) due to the low partial pressure of CO<sub>2</sub>. Amines are the most suitable for CO<sub>2</sub> removal at SMR [129,130].
- <sup>hh</sup> Capture efficiency can range from 90-98% [131]. Higher efficiency comes at a considerable higher cost; the lowest value has therefore been used.
- <sup>ii</sup> Value taken from [132] assuming off-gas with a CO<sub>2</sub> concentration of 13% [61]. The operational cost has also been taken from this source: 19.9 \$/t CO<sub>2</sub>. The formula given by Chapel coincides with the value given by Hendriks [131].
- <sup>jj</sup> A scale factor of 0.5-0.6 is reported in [132]; the highest value is assumed here.
- <sup>kk</sup> The compression energy required is calculated with the conventional engineering equation as given in [61].
- <sup>ll</sup> [107].
- <sup>mm</sup> [133].
- <sup>nn</sup> [93].
- <sup>oo</sup> Based on [134]; o&m cost consists of pipeline pigging, anti-corrosive measures, cathodic protection, monitoring the control equipment and odorisation.
- <sup>pp</sup> The cost for CO<sub>2</sub> storage has been assumed equal for all scenarios. This cost strongly depends on the geological site; here the cost at an onshore aquifer of 2 km depth has been taken from Hendriks et al. [135].
- <sup>qq</sup> Capital cost of a 2.4 tpd refuelling station is 1.5 million \$ [113]. This is divided into 80% compression and storage and 20% construction and other miscellaneous necessities. To account for the latter, 300,000 \$ (20% of \$ 1.5 million) is added to the cost of the centralised production chains. For the reformer in the decentralised chains this has already been included. To account for installation and unforeseen costs 20% has been added to the capital cost of storage, compressor and dispenser [24].



FIGURE 12. Potential configuration of a large-scale CO<sub>2</sub> network [91]

## 6.2 RESULTS FUEL SUPPLY CHAINS

The wholesale hydrogen cost for all chains, including those with CO<sub>2</sub> sequestration, is given in Figure 13. Liquid hydrogen is more expensive compared to gaseous hydrogen owing to the large contribution of liquefaction for both energy use and capital. Although centralised steam reforming is much cheaper than on-site reforming (8 \$/GJ versus 13 \$/GJ), the additional investment in distribution infrastructure increases total hydrogen cost for regional production (RSMRG) to a level close to that of local (LSMRG). While this indicates that on-site SMR could approach the cost of centralised, one should bear in mind that this is strongly dependent on the assumption of mass production. It is currently based on 250 units, but at higher production volumes the cost difference will be more in favour of small-scale reforming (see also Section 5.3). Recalling the number of refuelling stations in the Netherlands (4200), the latter situation seems more likely and implies that on-site steam reforming could potentially be as cost-effective as centralised to supply hydrogen to refuelling stations.

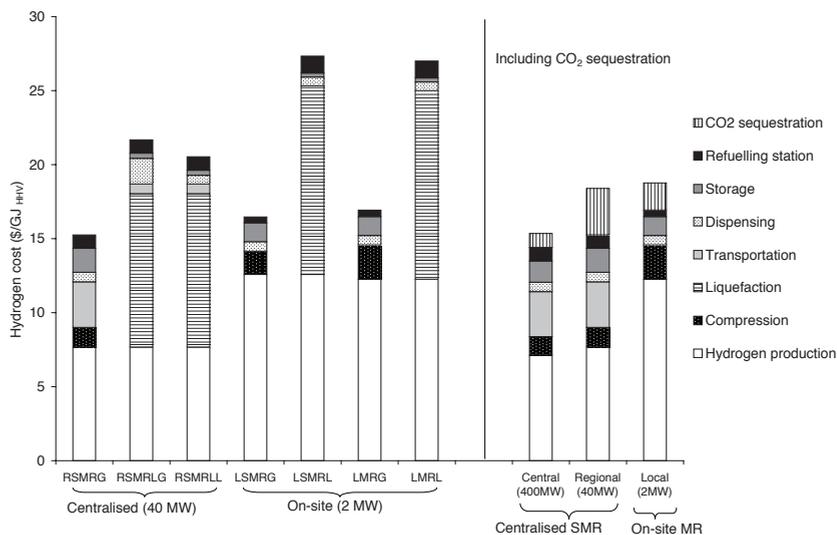
Owing to the high cost of reforming and compression, the membrane reactor (LMRG) gives a higher hydrogen cost than the SMR: 17 \$/GJ versus 15 \$/GJ (regional) and 16 \$/GJ (local). It was already indicated that upscaling has a more beneficial effect on the SMR than on the MR. Hence, the higher capacity assumed here makes the SMR slightly less expensive than the MR. By reducing membrane thickness to less than 10 micrometer, cost will

approach that of regional steam reforming (15 \$/GJ).

Including CO<sub>2</sub> sequestration in the hydrogen cost, the final cost for the MR (19 \$/GJ) nears that of the regional SMR (18 \$/GJ). Figure 14 shows the disaggregated cost of CO<sub>2</sub> capture, transport and storage. It demonstrates the low cost of CO<sub>2</sub> capture for the MR in comparison to conventional technology. Whereas the capture cost for the local scenario is less, CO<sub>2</sub> transport needs additional investment since a network from the stations to the grid is necessary. Especially the smallest capacity pipelines (1<sup>st</sup> branch) contribute considerably (45%) to the total cost and is the main cause that sequestration cost (46 \$/t CO<sub>2</sub>) exceeds that of the central scenario (42 \$/t CO<sub>2</sub>).

If we consider a best-case scenario in which thin membranes (several μm) and industrial utility prices are assumed<sup>21</sup>, first estimates of hydrogen cost are circa 14 \$/GJ including CO<sub>2</sub> sequestration and can decrease even further with higher production capacity. This is less than large-scale hydrogen production with CO<sub>2</sub> sequestration (15 \$/GJ) and would make small-scale CO<sub>2</sub> separation with membrane reactors an attractive infrastructure option.

21. Equal to large-scale production.



22. Based on a production price of 0.30 \$/l for gasoline and 0.305 \$/l for diesel in the EU 2001-2003. Energy content is 34.5 MJ/l gasoline and 38.5 MJ/l diesel.

23. For the ideal configuration 33.7 kg CO<sub>2</sub>/GJH<sub>2</sub> is emitted (heat production and electricity generation). An emission factor of 560 g CO<sub>2</sub>/kWh<sub>e</sub> is assumed for electricity generation [97].

FIGURE 13. Levelised hydrogen cost (CO<sub>2</sub> sequestration includes CO<sub>2</sub> capture, transport and storage)

To assess the advantage of the MR for emission reduction, an indication of the CO<sub>2</sub> mitigation costs are given. We assume a ‘business-as-usual’ scenario in which gasoline continues to be used as a fuel in (advanced) internal combustion engines vehicles. The CO<sub>2</sub> mitigation costs are then calculated by dividing the cost difference per km with the difference in CO<sub>2</sub> emissions per km between a hydrogen FCV and a gasoline ICEV. Without taxes or profit, gasoline costs 9 \$/GJ at a refuelling station in the EU<sup>22</sup> [96] and hydrogen 18 \$/GJ for the MR including CO<sub>2</sub> sequestration<sup>23</sup>.

For advanced ICEVs the fuel economy is 1.1-1.6 MJ/km and for FCVs fuelled with hydrogen 0.6-1.8 MJ/km<sup>24</sup>. Taking the most efficient values this results in a fuel cost of 1 \$ct/km for gasoline and 1.1 \$ct/km for hydrogen. Including investment for the car (see for more detail: [97]), costs are 16.4 \$ct/km for gasoline and 18.1 \$ct/km for hydrogen. With a reference emission for future ICEVs of 100 g CO<sub>2</sub>/km [97] and emissions for hydrogen production with the MR of 20 g CO<sub>2</sub>/km, final CO<sub>2</sub> mitigation cost is approximately 200 \$/t CO<sub>2</sub> avoided. Considering the current CO<sub>2</sub> emission market price of about 20 \$/t CO<sub>2</sub>, hydrogen production using a MR in combination with CO<sub>2</sub> sequestration is an expensive option for CO<sub>2</sub> emission reduction.

24. 80 mpge is equivalent to 1 MJ/km [24]. 1.8 MJ/km is the fuel economy of a current fuel cell vehicle [82].

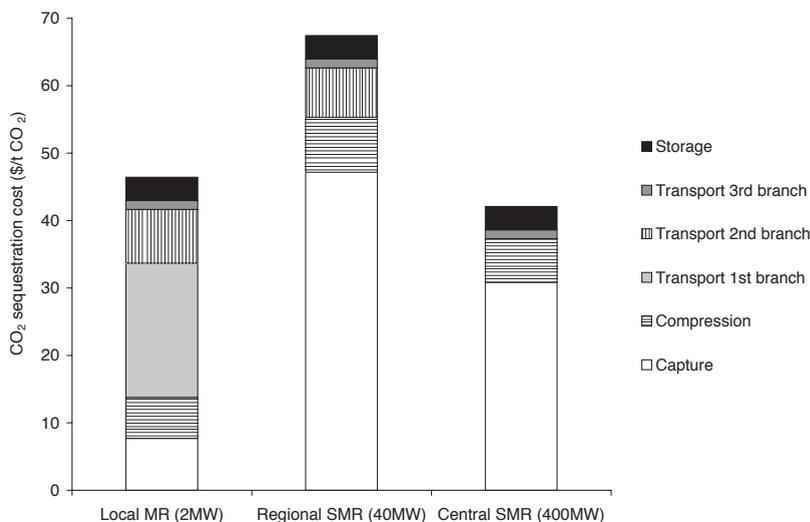


FIGURE 14. CO<sub>2</sub> sequestration cost

## 7 CONCLUSIONS

The small-scale membrane reactor has been evaluated in relation to its future economic and thermodynamic potential including its prospect of inexpensive CO<sub>2</sub> separation. The studied configurations for the reformer have been specifically designed as on-site hydrogen production facilities, envisaged at refuelling stations in a future transportation infrastructure.

The main promises of the membrane reactor such as higher methane conversion at lower temperature and higher total efficiency compared to conventional steam reforming are confirmed. The highest efficiency attained with the MR is 84%<sub>HHV</sub> without hydrogen compression and 78% with compression (482 bar) for the non-sweep configuration. The sweep configuration is 0.5%<sub>pt</sub> less efficient. Optimal heat integration, higher natural gas feed pressure and improved membrane performance may give further efficiency improvement.

Hydrogen production cost with a MR is competitive with conventional steam reforming for a 0.2 MW plant, but gets more expensive at larger capacities. This is mainly caused by the smaller benefit of upscaling for the MR due to its modular nature. For a 2 MW MR hydrogen cost is 14 \$/GJ with the sweep configuration being a few percent more expensive than non-sweep. The inherent uncertainty in the input assumptions showed that there is a large range of values possible with the most likely value around 14.5 \$/GJ. Reducing membrane thickness is one of the best opportunities of making the technology competitive, but since this may also change the reaction dynamics we can only give a rough estimate. With a thickness of several micrometers, already realised in laboratory settings, production cost is lowered to less than 12 \$/GJ. This would also reduce the risk related to the price volatility of palladium. Technological learning can contribute to further cost reductions as market volumes are expected to reach considerable quantities if hydrogen really takes off as a transportation fuel.

Several uncertainties are crucial with respect to the competitiveness of the MR. Assumptions on membrane permeability and lifetime remain to be proven in actual demonstration projects. Especially the effect of thermal cycling on the lifetime is important if one considers its application as an on-site reformer. Production technology of the membrane tubes is another fundamental issue that needs to be solved. At present, assembly of the membrane tubes is done by hand and constitutes an important part of the capital and annual cost. An inexpensive production method enabling machine assembly, and thus the prospect of mass production, and low gas prices are indispensable to lower costs. If these criteria are met, the MR may become a competitive alternative to the SMR.

Of the ten different fuel supply chains analysed, centralised steam reforming is most economically viable, but on-site steam reforming with gaseous hydrogen as a product might be a competitive alternative. The final hydrogen cost is 15 \$/GJ for a 40 MW SMR compared to 16 \$/GJ for a 2 MW SMR and 17 \$/GJ for a 2 MW MR (all gaseous H<sub>2</sub>).

The modelling results also revealed that the MR has the potential of relatively inexpensive carbon dioxide capture. That is, as long as a minimum concentration of contaminants in the gas stream (consisting mainly of CO<sub>2</sub>) does not pose any problems to the pipeline distribution network (ideal). If the exit gas is not pure enough for pipeline transport a cryogenic separation unit is added to purify the CO<sub>2</sub> stream. Including CO<sub>2</sub> separation and compression, total system efficiency is lowered by a little over 1%pt, which is a low penalty in comparison to other systems with CO<sub>2</sub> capture being studied. Cost for a small-scale MR with cryogenic separation is expensive (almost 50 \$/t CO<sub>2</sub>), but the economic feasibility of the ideal situation is promising: circa 14 \$/t CO<sub>2</sub> captured. For small-scale CO<sub>2</sub> capture, however, an expensive infrastructure is required to transport CO<sub>2</sub> to a storage site, resulting in total sequestration costs of 46 \$/t CO<sub>2</sub> for the MR, compared to 65 \$/t CO<sub>2</sub> for a 40 MW centralised plant. For the latter a less extensive CO<sub>2</sub> infrastructure is necessary, but CO<sub>2</sub>

capture is more expensive. At larger plants this advantage disappears as both hydrogen production cost and CO<sub>2</sub> sequestration cost are considerably reduced: hydrogen cost (including CO<sub>2</sub> sequestration) is around 15 \$/GJ for a 400 MW SMR versus 18 \$/GJ for a 40 MW SMR and 19 \$/GJ for a 2 MW MR. Regarding the trade-off between a H<sub>2</sub> infrastructure and a CO<sub>2</sub> infrastructure, it therefore strongly depends on the scale of centralised production whether CO<sub>2</sub> sequestration with on-site membrane reactors should be considered.

In the 'best-case' scenario for MR, which incorporates thin membranes, industrial utility prices and high production volumes, total cost for the MR can be reduced to less than 14 \$/GJ (including CO<sub>2</sub> sequestration), thus making this scenario more attractive than large-scale production. However, total cost of CO<sub>2</sub> sequestration would be 200 \$/t CO<sub>2</sub> avoided. Finally, it should be noted that the modelling approach for the reactor is simplified and does not take into account reaction kinetics. Comparison of the results with other studies do indicate that the approach is sufficiently accurate for our techno-economic calculations, but as it does not represent the technology in all of its complexities, the results should be interpreted accordingly and as such only give an indication of the potential of the technology. Especially for thin membranes the accuracy of the calculations is reduced, since thermodynamic and kinetic conditions may be slightly altered. It is therefore recommended that a more detailed model is developed to determine the possible impact of our assumptions.

This study has identified the following areas in need of further research:

- The performance of the membrane itself is a crucial factor. More RD&D is needed to enable the production of a membrane that combines a thin layer, high flux and suitable stability in all operating conditions. It should also facilitate a cheap manufacturing method with the potential of mass production. Membrane materials other than palladium-based such as zeolites and amorphous silica could provide a cheaper and more stable alternative and should be investigated.
- The stability, reliability and lifetime of the reactor under normal operating conditions and with numerous start-up and shut-down cycles need to be improved. Especially the lifetime of the membrane tubes should be given extra attention.
- A more detailed modelling method is required to further optimise operating conditions.
- Experimental data should be gathered to verify the technical feasibility of CO<sub>2</sub> separation with the MR.
- The relation between hydrogen storage, production and demand patterns should be evaluated.
- More in-depth analysis of the trade-off between size of production facility and infrastructure requirements.

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## CHAPTER V

### IDENTIFICATION OF EARLY OPPORTUNITIES FOR CO<sub>2</sub> SEQUESTRATION - WORLDWIDE SCREENING FOR CO<sub>2</sub>-EOR AND CO<sub>2</sub>-ECBM PROJECTS \*

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#### ABSTRACT

A study has been performed to identify potential worldwide opportunities for early application of CO<sub>2</sub> sequestration. An early opportunity is defined as a high purity CO<sub>2</sub> point source, which can provide CO<sub>2</sub> at low costs to oil or coal fields, where the CO<sub>2</sub> is sequestered, and simultaneously enhance oil production or coal bed methane production. A Geographic Information System (GIS) was used to combine worldwide CO<sub>2</sub> point sources and oil and coal fields. This resulted in 429 potential source-oil field and 79 source-coal field combinations. A Multi-Criteria Analysis (MCA), in which technical and socio-economic criteria are taken into account, was applied to rank the source-reservoir combinations generated by the GIS exercise. Some of the most promising cases were considered in more detail to select four illustrative cases for further study: two potential enhanced oil recovery (EOR) projects and two potential enhanced coal bed methane recovery (ECBM) projects. Case 1 consists of a hydrogen plant in Saudi Arabia, which could sequester 0.26 mt/yr CO<sub>2</sub> in a depleted oil reservoir at a net saving of approximately 3 €/t CO<sub>2</sub>. EOR case 2 is a hydrogen plant in California, USA, which has to be retrofitted in order to generate a pure CO<sub>2</sub> stream. Approximately 0.28 mt CO<sub>2</sub> could be stored annually. Mitigation costs have been estimated at 9 to 19 €/t CO<sub>2</sub>, depending on the availability of steam for CO<sub>2</sub> regeneration. In case 3 and 4, circa 0.68 and 0.29 mt CO<sub>2</sub> from ammonia plants in China and Canada could be sequestered annually in coal fields for ECBM production at approximately 5 and 6 €/t CO<sub>2</sub>, respectively.

## I. INTRODUCTION

In the coming decades, the role of fossil fuels in world energy supply is expected to remain dominant. Careful analysis of the long-term availability of oil and gas reservoirs indicate that available resources could meet the world's energy demand for another 50-100 years at low cost levels, and possibly longer. Coal resources are so abundant that they could last for centuries [1]. Simultaneously, many countries have committed themselves to reduce their CO<sub>2</sub> emissions. However, the market penetration of renewables is slower than was hoped for and improvements in energy efficiency in general have not offset the increasing demand for energy due to economic development. A possible solution is foreseen in the so-called 'clean use' of fossil fuels, in which the majority of CO<sub>2</sub> produced during the use of fossil fuels is captured and stored in a geological reservoir or the ocean. The process of CO<sub>2</sub> capture and storage is also referred to as carbon dioxide removal (CDR) or CO<sub>2</sub> sequestration.

A CO<sub>2</sub> sequestration scheme involves three distinct components: capture of the emitted CO<sub>2</sub> from the source followed by compression and dehydration, transportation to the storage site and injection and storage of the CO<sub>2</sub> in the geological reservoir. One of the key barriers to the introduction of CO<sub>2</sub> sequestration technology is the cost associated with CO<sub>2</sub> capture. Most CO<sub>2</sub> point sources considered to supply CO<sub>2</sub> (industrial and electricity plants) generally emit diluted CO<sub>2</sub> streams. The costs of CO<sub>2</sub> capture at gas and coal-fired power plants has been studied extensively, e.g. in [2-5]. Typical cost ranges for CO<sub>2</sub> removal from the exhaust gas of power plants with amines (state-of-the-art technology) are in the range of 40-60 \$/t CO<sub>2</sub> avoided for pulverised coal plants (PC) and 30-70 \$/t CO<sub>2</sub> avoided for natural gas combined cycles (NGCC). However, capture costs can be minimised by utilising exhaust gas streams with high purity CO<sub>2</sub>, which are emitted by several industrial processes. Plants using these processes are the most favourable options to supply low-cost CO<sub>2</sub> in the short term.

The contribution of transport to the total CO<sub>2</sub> sequestration costs is generally low. Transport costs vary between 1 and 3 \$/t CO<sub>2</sub> per 100 km of pipeline [6]. However, for high purity sources the contribution of transport costs will become more significant in the economics of the whole scheme. In order to minimise transport costs, the reservoir should be close to the source.

Storage costs depend on the reservoir type and local geological conditions. For aquifers and gas reservoirs (on- and offshore) storage costs vary between approximately 1 and 15 \$/t CO<sub>2</sub> [7]. In certain reservoirs, storage can be realised at small (or even negative) cost. This opportunity exists where oil and gas recovery is enhanced by the injection of CO<sub>2</sub> into oil/gas reservoirs or deep unminable coal seams, thereby generating oil/gas revenues, which can offset CO<sub>2</sub> sequestration costs. The injection of CO<sub>2</sub> (mainly from natural geological reservoirs) into oil reservoirs which are close to the end of their economic lifetime to enhance oil recovery

(CO<sub>2</sub>-EOR), is successfully applied in several ongoing commercial projects, mainly in the USA [8]. CO<sub>2</sub> injection into deep unminable coal seams to store CO<sub>2</sub> and simultaneously produce coal bed methane (CBM) held in the coal bed is referred to as enhanced coal bed methane production with CO<sub>2</sub> (CO<sub>2</sub>-ECBM). The exploitation of CO<sub>2</sub>-ECBM is being tested in projects in the USA and Canada and will be tested in Poland by the end of 2003, but is not yet commercially proven [9,10].

The objective of this study is to identify potential worldwide opportunities for early implementation of CO<sub>2</sub> sequestration. Therefore, combinations of high purity CO<sub>2</sub> sources with closely located, low-cost storage options such as CO<sub>2</sub>-EOR and CO<sub>2</sub>-ECBM are identified for which CO<sub>2</sub> mitigation costs are minimal (or even negative), making these potential attractive projects for investors. Such CO<sub>2</sub> sequestration projects are important to gain practical experience in the short term and could pave the way for cost reductions and consequently other applications of this option.

In order to identify those early opportunities, the following steps were performed:

1. A Geographic Information System (GIS) has been used to link worldwide high-purity CO<sub>2</sub> point sources to oil and gas reservoirs. This step has been discussed extensively in [11].
2. Subsequently, a Multi-Criteria Analysis (MCA) was applied to rank the source-reservoir combinations generated by the GIS exercise to allow the selection of four promising cases studies, which serve as illustrative examples. Generic source, reservoir and country-specific features were accounted for in the MCA.
3. A simple mass and energy balance was set up for each case to estimate the quantity of CO<sub>2</sub> that could theoretically be stored and the amount of additional oil/gas that could be recovered.
4. A simple economic analysis was performed to give insight into compression, transport and storage costs, as well as oil/gas revenues, from which CO<sub>2</sub> mitigation costs could be estimated.

Several studies have been completed or are planned to identify the most economic sequestration schemes focussing on a specific country, source or reservoir. A GIS-based tool for CO<sub>2</sub> source and reservoir data in Europe, the USA and Canada has been used to examine opportunities for capturing and sequestering current and future CO<sub>2</sub> emissions [12-14]. Kuuskraa et al. [15] developed a model to select the most economic sequestration site for power and industrial plants in the USA. This study differs from the previously mentioned studies in the fact that high-purity sources and sequestration sites that offer value-added opportunities in the form of oil or gas revenues are explicitly considered as potential sequestration projects. The approach in our study is similar with respect to source-sink matching, but is carried out on a global scale. A key additional step in this analysis is the application of a MCA, in which various non-technical aspects are taken into account, in order to identify early opportunities in very different contexts worldwide.

## 2 WORLDWIDE CO<sub>2</sub> SOURCES AND OIL AND COAL FIELDS

### 2.1 CO<sub>2</sub> SOURCES

A recent study made an inventory of all large anthropogenic CO<sub>2</sub> point sources (industrial and power plants) worldwide [16]. The total emission in this inventory equals 13.5 Gt CO<sub>2</sub> per year, corresponding to 60% of the estimated 1997 total anthropogenic emission worldwide (excluding emissions caused by deforestation) of approximately 22.6 Gt CO<sub>2</sub> [17]. All sources were imported into ArcView, a Geographic Information System. In this study, only those sources that produce a waste stream of nearly pure CO<sub>2</sub> (hydrogen<sup>1</sup>, ammonia<sup>2</sup> and ethylene-oxide plants) with a minimum emission of 0.1 mt/year<sup>3</sup> were selected. The resulting set of so-called 'pre-selected sources' consisted of 198 sources (see Figure 1).

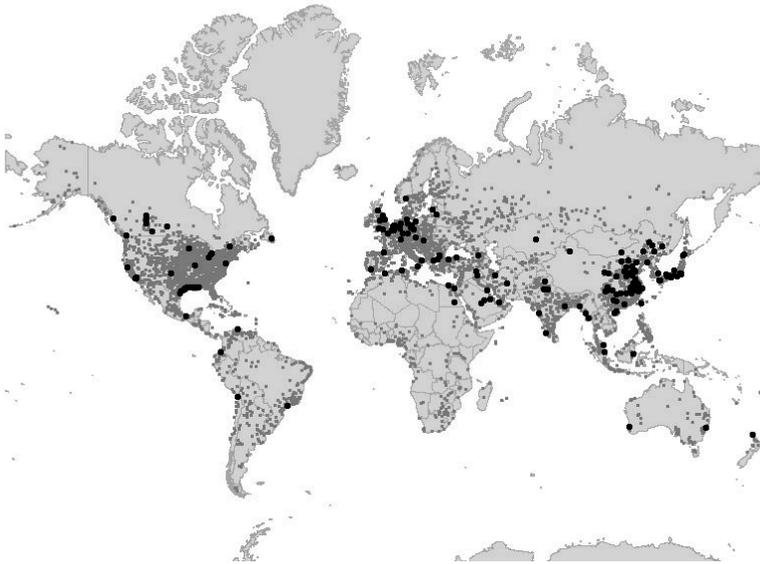


FIGURE 1. Geographical distribution of CO<sub>2</sub> sources [16]. The 'pre-selected sources', including hydrogen, ammonia and ethylene-oxide production plants, are indicated in black

### 2.2 OIL OCCURRENCES

The main sources of information on global oil occurrences used in this study are the Digital Data Series of the U.S. Geological Survey covering their latest petroleum assessments [18,19] as can be seen in Figure 2. Oil resources were evaluated at the level of the Assessment Unit (AU), which is a mappable volume of rock that encompasses reservoirs (discovered and undiscovered), which share similar geologic traits and socio-economic factors [18]. An AU has typical dimensions of thousands square kilometres. Within the AUs, areas with known oil accumulations were digitised.

For the calculation of the enhanced oil yield and volumes of CO<sub>2</sub> that

1. Hydrogen is generally produced by converting natural gas into CO<sub>2</sub> and H<sub>2</sub> by steam reforming, after which CO<sub>2</sub> is separated. At older hydrogen plants, CO<sub>2</sub> is removed by a scrubbing process using Benfield, Sulfinol, MEA or similar absorption materials, thereby generating an almost pure CO<sub>2</sub> stream. At more modern hydrogen plants, a pressure swing absorption (PSA) unit is used to produce H<sub>2</sub>, thereby generating a gas stream containing CO<sub>2</sub> (about 50%), unrecovered H<sub>2</sub>, CO, CH<sub>4</sub> and nitrogen. This stream is recycled to the reformer as fuel to reduce the natural gas input for steam reforming. The CO<sub>2</sub> concentration in the exhaust gas is low (in the order of 10-20%) and therefore, strictly speaking these units do not meet the high-purity criterion.

2. CO<sub>2</sub> emissions are corrected for CO<sub>2</sub> use for urea production. Some operators also sell a part of produced CO<sub>2</sub> for other commercial purposes, but this is not taken into consideration.

3. To obtain a minimum scale of operation, in order to keep transport costs at an acceptable level.

can potentially be stored during EOR-operations, the method used by Stevens et al. [8] was followed, with some adaptations [11,20]. In this method, the volume of extra oil due to CO<sub>2</sub> injection and the volume of CO<sub>2</sub> that can be stored are correlated to the contact factor (between CO<sub>2</sub> and oil), reservoir depth, oil viscosity (API gravity) and (net) CO<sub>2</sub>:oil ratio. The data provided by USGS allow the application of uncertainty quantification by Monte Carlo analysis. By means of this analysis, the expected extra oil produced and amount of CO<sub>2</sub> that can be stored in an oil reservoir are predicted in a probabilistic manner, accounting for variations in contact factor, depth, API and CO<sub>2</sub>:oil ratio.



FIGURE 2. Worldwide areas with known oil occurrences [18,19]

### 2.3 COAL OCCURRENCES

The most important data sources used to construct a global map of coal occurrences were the earlier IEA studies [9,21] and the IGCP study on world coal fields [22] (see Figure 3). Although this inventory is probably not complete, it covers the major coal producing countries of the world. For each selected coal seam, the amount of producible CBM and the amount of storable CO<sub>2</sub> was calculated per square kilometre, assuming homogeneous deposits throughout the investigated area [11,20]. Again, Monte Carlo analysis was applied to quantify the uncertainties in the calculated volumes, among which cumulative thickness of coal, coal density, gas content, completion factor, recovery factor and exchange ratio. A conservative exchange ratio (from a sequestration point of view) of CO<sub>2</sub>:CH<sub>4</sub> was used in the calculations, varying between 1 and 2.5.

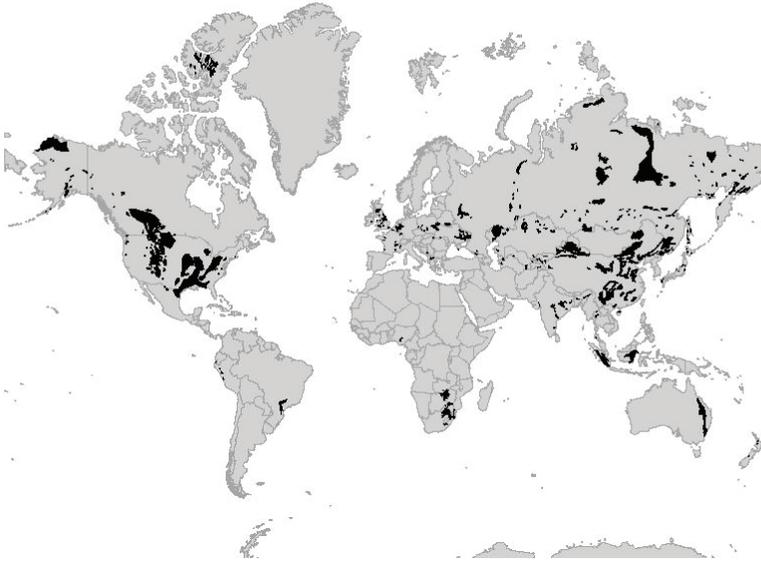


FIGURE 3. Global coal occurrences [9,21,22]

#### 2.4 COMBINING SOURCES AND RESERVOIRS

Potential early opportunities can be identified by scanning for source-reservoir combinations located within a limited distance from each other in the GIS programme. Imposing a maximum distance of 100 km to limit transport costs results in 64 CO<sub>2</sub> point sources located within this distance from oil accumulations. In total, 420 source-oil field combinations could be established, since many of the sources could be linked to more than one AU. For ECBM, 59 CO<sub>2</sub> point sources are located within 100 km distance from coal accumulations, from which 79 combinations could be made in total. The most promising combinations of CO<sub>2</sub> source and reservoir were selected on the basis of a rough estimation of the ratio between oil or gas revenues and expected drilling costs<sup>4</sup>. In doing so, it is possible that some reservoirs are excluded, which are most suited from a geological and technical point of view. However, within this study it is not realistic to consider all selected oil and coal fields in such a level of detail.

### 3 SELECTION OF EARLY OPPORTUNITIES FOR CO<sub>2</sub> SEQUESTRATION

#### 3.1 MULTI-CRITERIA ANALYSIS

Apart from the reservoir characteristics and technical features of the CO<sub>2</sub> source, there are other relevant aspects, which determine the feasibility of a CO<sub>2</sub> sequestration project. In order to account for various relevant aspects in a systematic way, a Multi-Criteria Analysis can be applied. This method, frequently used to support policy decisions, enables to evaluate different alternatives. By means of an MCA, the alternatives (source-reservoir combinations) can be ranked, considering different

4. Oil and gas revenues are calculated by multiplying the extra oil/gas yield with an oil/gas price of 15 \$/bbl and 2.6 \$/GJ, respectively. The drilling costs for oil fields are calculated by multiplying the number of wells required to store the cumulative CO<sub>2</sub> production in the project lifetime by the depth of the reservoir and drilling costs of 700 €/m [23]. The drilling costs for coal fields are calculated from the number injection and production wells and the investment costs for a production and injection well of 0.5 M€ and 0.3 M€, respectively [24].

5.  
The sum of the weight factors should always be 1.

criteria and priorities. There are several MCA methods, of which the weighted summation method is applied in this study. In this method, each criterion is first standardised, converting all criteria in values between 0 and 1, in which 0 is the worst value and 1 the best value. The standardised criteria are then multiplied by a weight factor<sup>5</sup>, representing the contribution of that criterion to the total score, which is the sum of the products of criteria and weight factors. Both the criteria and weight factors were generated by consultation between several experts on reservoirs and CO<sub>2</sub> sequestration technology, although certain subjectivity is inherent to the choice of criteria and weight factors. Therefore, three weight factor sets were applied. The first set puts emphasis on the techno-economic criteria, whereas in the second set, the socio-economic criteria are more strongly represented. The third weight factor set can be considered as an average set. Cases with a high total score for each weight factor set can be classified as promising early opportunity.

The various criteria accounted for in this study are given in Table 1. The ratio P90:P10 reflects the distribution in the enhanced oil or gas yields from the Monte Carlo calculations. The oil viscosity is accounted for, since the oil market value depends on it [8]. The vicinity of other sources, which could supply CO<sub>2</sub> to the same reservoir, is included, because this might reduce costs as more sources can make use of the same pipeline infrastructure. The population density is a relevant factor in the project implementation, since pipeline construction costs increase in densely populated areas. The government quality is the sum of aggregate governance indicators of a country, among which political stability, government effectiveness, regulatory control, rule of law and control of corruption [25]. For each country, these indicators are presented on a scale from -2.5 (worst) to 2.5 (best). The sovereign long-term foreign currency rating classifies a country on the quality of their financial system (investment climate). In countries with a bad rating of this kind, private investors will find it hard to obtain foreign finance for possible CO<sub>2</sub> sequestration projects. In this study, the country ratings of Standard & Poor's are used [26]. This rating agency classifies long-term foreign currency rating of a country in letter codes with decreasing quality from AAA to CCC<sup>6</sup>. The replicability indicates whether the source, resource and location are representative as a typical CO<sub>2</sub>-EOR/ECBM case. Atypical cases (e.g. extremely poor socio-economic conditions or extremely high oil reserves) are less interesting, since the potential that a similar case will be realised somewhere else is small.

6.  
Each code is converted into a value between 0 and 1 according to the scheme below:

Code	Value
AAA	1
A-	0.76
BB-	0.4
AA+	0.91
BBB+	0.69
B+	0.27
AA	0.89
BBB	0.67
B	0.22
AA-	0.87
BBB-	0.62
B-	0.18
A+	0.8
BB+	0.49
CCC+	0.04
A	0.78
BB	0.44
CCC	0

The MCA was deployed for the 420 EOR and 79 ECBM source-reservoir combinations that resulted from the first steps of this analysis (see Section 2.4). This resulted in a ranking of source-reservoir combinations, which was used to compose a top-15 for each weight factor set. These short lists of possible projects for both EOR and ECBM are presented in Figure 4 in an aggregate way.

TABLE I. MCA criteria and weight factors used for CO<sub>2</sub>-EOR and CO<sub>2</sub>-ECBM (in brackets). Data on depth of the coal fields were not available

Criteria	Values/ units	Weight factor set 1 (techno- economic)	Weight factor set 2 (socio- economic)	Weight factor set 3 (average)
Transport distance source-field	1 (0-50 km) 0.5 (51-100 km)	0.1 (0.1)	0.05 (0.05)	0.075 (0.075)
Probable enhanced oil/gas yield (p50)	mbbl (mm <sup>3</sup> )	0.25 (0.35)	0.05 (0.05)	0.2 (0.25)
Ratio P90:P10	-	0.075 (0.1)	0.025 (0.025)	0.05 (0.05)
API gravity oil	31-41°	0.05	0.025	0.05
Drilling depth	m	0.15	0.05	0.1
More sources, which can store CO <sub>2</sub> in same resource?	0 (no) 1 (yes)	0.05 (0.05)	0.025 (0.025)	0.025 (0.025)
CO <sub>2</sub> emission source	Mt/yr	0.2 (0.3)	0.05 (0.05)	0.175 (0.2)
Population density	men/km <sup>2</sup>	0.025 (0.025)	0.175 (0.2)	0.05 (0.1)
Government quality	-2.5 - 2.5	0.025 (0.025)	0.2 (0.25)	0.1 (0.125)
Sovereign long-term foreign currency rating	AAA-CCC	0.05 (0.025)	0.3 (0.3)	0.15 (0.15)
Replicability	0 (no) 1 (yes)	0.025 (0.025)	0.05 (0.05)	0.025 (0.025)

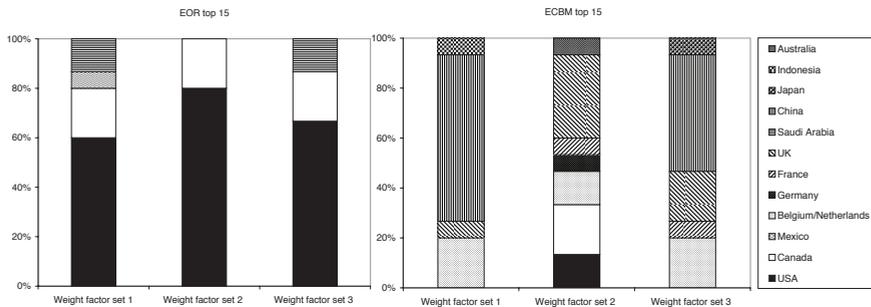


FIGURE 4. Distribution of top 15 promising sources for CO<sub>2</sub>-EOR and CO<sub>2</sub>-ECBM (short list). The presence of many potentially promising EOR projects in the USA can be explained by the fact that relatively many oil occurrences were defined due to the use of more detailed units to describe oil occurrences in the USA

### 3.2 SELECTION OF CASE STUDIES

The source-reservoir combinations from the short list were reviewed to select four illustrative case studies. The choice of the four case studies from this top 15 was undertaken in close collaboration with IEA Greenhouse Gas R&D Programme. In this selection procedure, more site-specific information was added, amongst others by consulting local experts. The characteristics of several reservoirs were checked to make sure whether they are indeed appropriate for CO<sub>2</sub> storage and oil/gas production. Where possible, coal occurrences were checked for CBM and mining activities. In cases where mining activities exist in the area or are planned for the future, the coal occurrence was not considered appropriate for CO<sub>2</sub>-ECBM. Apart from technical and feasibility requirements, the case studies should represent a broad range of

geographical/social conditions as this study aims to illustrate early opportunity projects worldwide.

From the EOR cases, it appeared that the Canadian oil reservoirs that occur in the Michigan basin are less suitable, because there is not much capacity for CO<sub>2</sub> storage on the Canadian side of the basin [27]. This restricts the choice to Saudi Arabia and the USA. The cases found in the USA represent 3 different regions: Texas, Louisiana and California. There are already ongoing CO<sub>2</sub>-EOR projects in both Texas and Louisiana [8]. When an early opportunity is considered as an example project, the sites in California are more interesting. Benson [28] studied the options for CO<sub>2</sub> sequestration in California by considering the distribution of CO<sub>2</sub> emissions from fossil fuel power plants and their proximity to active or depleted oil/gas reservoirs. In a study of Kuuskraa et al. [15], a case study is mentioned examining CO<sub>2</sub> sequestration opportunities for a series of refinery-based hydrogen plants in the West Coast area.

From the Chinese ECBM cases, a coal basin in the province of Shaanxi located near the Qinshui basin offers the best prospects for short-term ECBM recovery. Although not as extensively drilled as the neighbouring province (Shanxi), the area is one of nine coal bed methane blocks approved for exploitation through foreign co-operation by the Chinese government [9]. A study in which the potential and feasibility of CO<sub>2</sub>-ECBM at several coal basins worldwide is determined points out that most (if not all) of the European coal basins are geologically less suited to CBM development [9]. Based upon this conclusion, it was decided that the European reservoirs from the top 15 would not be included in the list of cases to be studied in more detail. The Indonesian coal field located in Kalimantan has been evaluated as a potential reservoir for CO<sub>2</sub>-ECBM [9]. Although it is one of Indonesia's most prospective coal basins for CO<sub>2</sub>-ECBM, a key barrier is the absence of a gas market and related infrastructure. Moreover, the socio-economic indicators for this site give a low score<sup>7</sup>, and therefore this region is not included. The next potential candidate is a Canadian (re)source in Alberta, although its techno-economic score is quite average. Canada has a large CBM potential, but so far this has not been exploited. In 1998-1999, a single well micro-pilot ECBM test was carried out successfully at the Fenn Bigg Valley, Alberta, Canada [29]. Despite modest exploration effort to date, the CBM potential of the Western Canada basin is still considered to be attractive [9], so this source-reservoir combination was decided to be eligible as case study.

7. Countries with a rating lower than BBB- are considered not to be eligible for pilot projects [20].

### 3.3 THE CASE STUDIES

The four selected source-reservoir combinations are given in Table 2. It is important to realise that these are illustrative case studies. Various site-specific aspects need to be considered in order to assure that both source and reservoir are indeed suited for CO<sub>2</sub> sequestration. One relevant uncertainty is the production phase in which each individual oil field within an assessment unit or play actually is. This information

cannot be obtained from the data used in this study. It might be that a certain oil field is still in the primary production phase, making CO<sub>2</sub>-EOR unnecessary. A more thorough analysis of especially the reservoirs would be required to carry out a more detailed feasibility study.

TABLE 2. Case studies selected from the short list of promising source-reservoir combinations

Parameter	1	2	3	4
Type of plant	Hydrogen	Hydrogen	Ammonia	Ammonia
Country	Saudi Arabia	USA	China	Canada
CO <sub>2</sub> emission (kt/yr)	260	281	677	293
Type of hydrocarbon recovery	CO <sub>2</sub> -EOR	CO <sub>2</sub> -EOR	CO <sub>2</sub> -ECBM	CO <sub>2</sub> -ECBM

*Case 1* is located in the city of Al-Jubail in the Eastern Province in Saudi Arabia. Circa 0.26 mt pure CO<sub>2</sub> stream is generated annually at a steam methane reforming (SMR) hydrogen plant from the Petromin-Shell refinery, which could be injected into oil fields of the Central Arch Horst-Block Anticlinal Oil and Gas AU (part of the Greater Ghawar Uplift Province). The average depth of this AU is 2000 m. Relatively little oil has been produced until now (the oil maturity is 24<sup>8</sup>) and few oil fields are exploited in this AU. The probable enhanced oil yield of one oil field within this AU is 11.67 mbbbl and the probable CO<sub>2</sub> storage capacity is 5.21 mt<sup>9</sup>. It is assumed that the cumulative CO<sub>2</sub> production in 15 years (the assumed project lifetime) can be sequestered in one oil field by 16 injection wells<sup>10</sup>.

*Case 2* is an Atlantic Richfield SMR hydrogen plant located in the city of Carson, California, USA. Recently, the company added a PSA downstream unit [31]. This implies that CO<sub>2</sub> (0.28 mt/yr) will be available at a concentration of only 12% (since the purge gas is combusted in the furnace to provide heat to the reformer) instead of 100% as was reported in the database. Since more and more hydrogen (and ammonia) plants in the future will have PSA systems and generate diluted CO<sub>2</sub> streams [16,32], it was decided to examine the performance and costs of CO<sub>2</sub> removal needed to generate a pure CO<sub>2</sub> stream in such a system<sup>11</sup>.

CO<sub>2</sub> produced at this plant can be injected into the Whittier Fault Zone and Fullerton Embayment, which comprise of the Los Angeles Basin. The average depth of this AU is on average 1676 m and it contains relatively heavy oil (average API gravity 27<sup>o</sup>). No data are available on the cumulative oil production. Relatively few oil fields (13) have been discovered in this AU. The probable enhanced oil yield of an oil field within this AU is 2.58 mbbbl and the probable CO<sub>2</sub> storage capacity is 1.11 mt. In order to store 15 years cumulative CO<sub>2</sub> production (4.2 mt), at least 4 oil fields and 18 injection wells are required.

*Case 3* is a fertilizer plant belonging to Chemical Industry Group, Ltd.

8. The oil maturity is a number between 0 and 100. In fields with high oil maturity, the proven oil reserves have been exploited to a large extent and undiscovered reserves are expected to be small.

9. These numbers refer to an oil field that can be encountered in this AU.

10. The number of oil fields is estimated by dividing the cumulative CO<sub>2</sub> production and the probable CO<sub>2</sub> storage capacity per oil field (CO<sub>2</sub>-50) from the Monte Carlo calculations. The number of injection wells is calculated by dividing the estimated daily oil production with the product of the average oil production rates per well (circa 46 bbl per day) and average production to injection well ratio (circa 2.8) from various CO<sub>2</sub>-EOR operations in the USA [8,30].

11. Strictly considered, plants with a PSA unit cannot be classified as early opportunity. On the other hand, these plants offer opportunities to capture CO<sub>2</sub> from relatively high-quality streams (read high CO<sub>2</sub> partial pressure) in the hydrogen production process. Another argument to consider this configuration is that it is one of the potential hydrogen production routes with CO<sub>2</sub> capture, which might play an important role in future hydrogen economies as envisioned by several energy specialists and market players [33].

located in the province of Shaanxi, in the north of China. The plant emits 0.68 mt of pure CO<sub>2</sub> per year, which can be injected into the Eastern piedmont of Taihang Mountains Coal basin. In [9], this basin is referred to as Taixing Shandou, which is very close to the Qinshui basin. The probable enhanced gas yield is about 118 mm<sup>3</sup>/km<sup>2</sup> and the probable CO<sub>2</sub> storage capacity is 0.38 mt/km<sup>2</sup>. The area required to sequester the cumulative CO<sub>2</sub> production is estimated at 36 km<sup>2</sup>. Assuming that 6x6 units of 1000x1000 m are realised, the number of production and injection wells that have to be constructed are 49 and 36, respectively.

*Case 4* is an ammonia plant located near the city of Medicine Hat in Alberta, Canada, operated by Canadian Fertilizer's Ltd. CO<sub>2</sub> emitted at this plant (0.29 mt/yr) could be injected into a coal seam in the Plains Region, which is around 1000 m deep at the location of the source. The probable enhanced gas yield is 44 mm<sup>3</sup>/km<sup>2</sup> and the probable CO<sub>2</sub> storage capacity is 0.14 mt/km<sup>2</sup>. The area required to sequester the cumulative CO<sub>2</sub> production is estimated at 42 km<sup>2</sup> (7x6 km), so 56 production and 42 injection wells have to be constructed.

#### 4 TECHNICAL PERFORMANCE OF THE CASE STUDIES

##### 4.1 CO<sub>2</sub> SUPPLY

It is assumed that the sources will supply a constant CO<sub>2</sub> stream in the next 15 years, which is taken as the economic lifetime for an early opportunity project. In reality, the demand for CO<sub>2</sub> is changing in time. When starting a project, relatively much CO<sub>2</sub> is required to mobilise the oil. Once CO<sub>2</sub> breakthrough occurs and the produced is recycled, the demand for CO<sub>2</sub> will decrease. In case of a constant CO<sub>2</sub> stream, a strategy is needed to deal with this problem. A possible solution would be to start the project with a relatively low injection rate (equal to CO<sub>2</sub> supply of source), so that it will take longer for CO<sub>2</sub> to break through. In time, the injection rate is increased when recycling rate is increased. This will have consequences for the oil production in time, but this impact is not accounted for in this study.

To meet the transport specifications, CO<sub>2</sub> should be compressed to 80 bar, dried and cooled. All sources in the case studies are located within 50 km of the reservoir.

In case study 2, an additional capture unit has to be installed to generate a pure CO<sub>2</sub> stream. In a study by Katofsky [34], it is suggested to install two PSA units in series. The first removes all CO<sub>2</sub> and H<sub>2</sub>O and the second unit removes all residual gases. In [33], a hydrogen production scheme for the foreseeable future producing 100% CO<sub>2</sub> is presented, consisting of a SMR plant and a capture unit using a solvent between the water gas shift reactor and the PSA unit. In this study, it was decided to consider an absorption unit with mono ethanolamine (MEA) as solvent, which is

commercially proven and for which more comprehensive data for costs and energy use are available. The main disadvantage of amines is the high steam consumption needed to regenerate CO<sub>2</sub> from the solvent. At refineries, there might be an excess of low/medium pressure steam, which could be deployed to regenerate, dry and cool CO<sub>2</sub>. The steam reforming process itself generates an excess of mainly high-pressure steam. The steam export for the production of 1000 Nm<sup>3</sup> H<sub>2</sub> by SMR is typically 1.1 t (75 bar/350°C) [32], corresponding to circa 0.25 GJ/GJ<sub>H<sub>2</sub></sub>. This corresponds to circa twice the steam demand to capture CO<sub>2</sub> from the hydrogen plant of case study 2 in an absorption unit, assuming a steam demand of 4 MJ/kg CO<sub>2</sub>.

#### 4.2 CO<sub>2</sub>-EOR

CO<sub>2</sub>-EOR or carbon dioxide flooding can be applied at oil fields where oil has already been produced with conventional extraction technologies (primary and secondary oil recovery)<sup>12</sup>. CO<sub>2</sub> is injected in the oil field, alternated with water injection. In the reservoir, CO<sub>2</sub> mobilizes oil by miscible and immiscible displacement [8]. Part of the CO<sub>2</sub> is sequestered in immobile oil and empty pores. At the production well, oil, water, natural gas and CO<sub>2</sub> are produced. These components are separated and the CO<sub>2</sub> is compressed and recycled to the injection well. It is important to realise that in existing commercial CO<sub>2</sub>-EOR projects, the main purpose is to maximise oil recovery with a minimum quantity of CO<sub>2</sub>, which might be contradictory with the purpose to maximise CO<sub>2</sub> sequestration.

From the cumulative CO<sub>2</sub> emission and the probable CO<sub>2</sub>:EOR ratio (0.45 and 0.43 t/bbl for case 1 and 2, respectively), the total enhanced oil production is calculated. A production profile is required to determine the enhanced oil yield in time and to estimate the CO<sub>2</sub> recycle rate. However, CO<sub>2</sub>-EOR production curves are scarce and site-specific. A constant oil production (and CO<sub>2</sub> injection/recycle<sup>13</sup>) rate in time is assumed for the economic calculations, similar to the schematic oil production profile given for a group of smaller fields [35]. It is assumed 50% of the injected CO<sub>2</sub> is purchased from the pipeline and 50% by recycling of CO<sub>2</sub> that has broken through, based upon similar values observed in the Wasson-Denver Field [8].

#### 4.3 CO<sub>2</sub>-ECBM

Coal bed methane is generally recovered by drilling production wells to pump water out of the coal seam, reducing the reservoir pressure. Consequently, methane will desorb from the coal matrix, diffuse to the cleats and flow with the water to the production well. By injecting CO<sub>2</sub>, theoretically, methane recovery can be increased, although this technology is not proven yet. For CO<sub>2</sub>-ECBM, a fine network of injection and production wells has to be constructed. A 5-spot cubical set-up of 1000 by 1000 m with 4 production wells and 1 injection well in the middle is assumed to be the optimal well spacing [9,24]. Injection and production start at the same time and last for 15 years, after which the

12. Injecting CO<sub>2</sub> during the early stages of exploitation (primary production phase) could improve oil recovery, while simultaneously maximising CO<sub>2</sub> storage. This concept requires detailed evaluation [8].

13. In [35], the recycling rate is linearly depending on the oil production. With an increasing oil production in time, also the CO<sub>2</sub> recycling rate will increase. Therefore, the recycling compressor needs to be designed for the maximum recycle ratio. In this study, a constant oil production and, consequently, a constant recycle rate is assumed, on which the compressor is designed.

production well is closed to sequester the CO<sub>2</sub> permanently. ECBM revenues as a function of time were calculated by means of a multiphase reservoir model [24]. The model calculates the gas production as a function of a (constant) CO<sub>2</sub> injection rate and a default amount of gas present in the coal layer. The production profile (see Figure 5) is adjusted to the probable methane content (CH<sub>4</sub>-50) of the considered coal occurrence and the emission rate of the source. The exploitation area is calculated from the cumulative CO<sub>2</sub> production and the probable CO<sub>2</sub> storage potential per km<sup>2</sup> (CO<sub>2</sub>-50). A correction factor is applied for the lower CO<sub>2</sub>:CH<sub>4</sub> exchange ratio when injecting for 15 years in comparison to the injection period of 42 years assumed in the reservoir model<sup>14</sup>. Finally, it is checked whether the selected coal field is large enough to ensure sequestration of the produced CO<sub>2</sub> in the source lifetime.

14. In the Monte Carlo calculations of the CO<sub>2</sub> storage potential, an exchange ratio for a completely produced field is used (between 1 and 2.5). However, the exchange ratio is lower in the first years, since the peak of methane production is in this period. This should be accounted for when injecting for only 15 years. The model predicts net exchange ratio CO<sub>2</sub>:CH<sub>4</sub> of 1.21 and 1.64 for 15 and 42 (complete field production) years injection period, respectively.

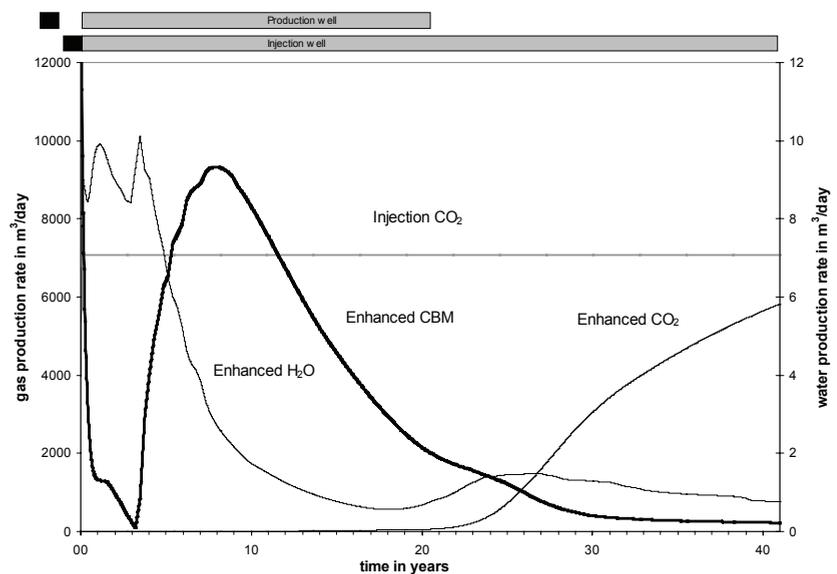


FIGURE 5. Gas and water flows for Dutch coal used as generic production profile for this study (Stars model, 1000×1000 m<sup>2</sup>, 410 m inseam, skin -3, CO<sub>2</sub> injection from first year) [24]. The enhanced CO<sub>2</sub> line indicates the CO<sub>2</sub> breakthrough in case the production well is not closed

#### 4.4 MASS AND ENERGY BALANCE

Table 3 shows the mass flows of CO<sub>2</sub> and oil/gas production in the project lifetime. CO<sub>2</sub> emissions caused by energy generation for capture, compression, drying and cooling of the CO<sub>2</sub> stream are given in Table 4. It is assumed that the oil and gas produced by CO<sub>2</sub>-EOR and CO<sub>2</sub>-ECBM replaces oil and gas produced in a conventional way (without CO<sub>2</sub> storage), so CO<sub>2</sub> emitted during the combustion of the enhanced oil and gas yield is not accounted for.

TABLE 3. Mass balance over the lifetime of the selected cases (15 years)

	EOR-1 Saudi Arabia	EOR-2 USA	ECBM-3 China	ECBM-4 Canada
Cumulative CO <sub>2</sub> emission source (Mt)	3.90	4.22	10.16	4.40
Total CO <sub>2</sub> injection (Mt)	7.80	8.44	10.16	4.40
Total CO <sub>2</sub> recycled (Mt)	3.90	4.22	-	-
CO <sub>2</sub> emissions (losses) (Mt) <sup>a</sup>	-0.20	-0.21	-	-
CO <sub>2</sub> emission electricity production (Mt) <sup>b</sup>	-0.36	-0.91	-0.48	-0.21
Net CO <sub>2</sub> sequestration (Mt)	3.34	3.10	9.67	4.19
Total oil/gas production (PJ) <sup>c</sup>	47	58	149	66

<sup>a</sup> Emissions from unidentified and abandoned wells are assumed to be 5% of net purchases in case of CO<sub>2</sub>-EOR [8]. For CO<sub>2</sub>-ECBM, emissions due to leakage are assumed to be 0.

<sup>b</sup> Calculated from the annual steam and electricity requirements to capture, compress, dry and cool CO<sub>2</sub> for transport, see Table 4.

<sup>c</sup> Assuming a LHV of 42 MJ/kg for crude oil and 35.8 MJ/m<sup>3</sup> for CBM gas.

TABLE 4. Annual steam (TJ/yr) and electricity (GWh/yr) requirements and related CO<sub>2</sub> emissions

	EOR-1 Saudi Arabia	EOR-2 USA	ECBM-3 China	ECBM-4 Canada
<i>Energy requirements</i>				
Steam for CO <sub>2</sub> capture <sup>a</sup>	-	1060	-	-
Electricity for CO <sub>2</sub> capture <sup>a</sup>	-	12.8	-	-
Electricity for compression at source <sup>b</sup>	27.8	30.1	72.4	31.3
Steam for drying <sup>c</sup>	2.1	2.3	5.4	2.3
Electricity for cooling <sup>d</sup>	0.6	0.6	1.5	0.7
Electricity for compression in field (recycling)	27.8	30.1	-	-
Total electricity requirements <sup>e</sup>	56	141 (74)	74	32
CO <sub>2</sub> emission (Mt/yr) <sup>f</sup>	0.02	0.06 (0.03)	0.03	0.01
Total CO <sub>2</sub> emission project lifetime (Mt)	0.36	0.91 (0.47)	0.48	0.21

<sup>a</sup> In order to recover 90% of the CO<sub>2</sub> with amines (Econamine FG process), between 3.7 and 4.9 MJ/kg CO<sub>2</sub> of low-pressure steam at 192 and 182°C is required, respectively [2]. Well-designed Econamine FG plants have been proven to use less than 4.2 MJ/kg CO<sub>2</sub> [36]. The CO<sub>2</sub> recovery energy of the KS-1 solvent (a new developed amine based solvent) can even reach a level of 3 MJ/kg CO<sub>2</sub> for CO<sub>2</sub> concentrations higher than 10% [37]. In this study, a conservative value of 4 MJ/kg CO<sub>2</sub> is assumed. The electric power consumption for flue gas and stack gas blowers in an electricity plant (173 kJ<sub>e</sub>/kg CO<sub>2</sub> recovered [38]) is used to estimate electricity consumption of CO<sub>2</sub> recovery.

<sup>b</sup> Compression is performed using a multistage compressor with a cooling step after each stage. The required shaft power to compress CO<sub>2</sub> from 1 to 80 bar is 385 kJ<sub>e</sub>/kg CO<sub>2</sub> [7].

<sup>c</sup> Drying requires 8 kJ steam/kg CO<sub>2</sub> [39].

<sup>d</sup> Cooling requires 8 kJ<sub>e</sub>/kg CO<sub>2</sub> [39].

<sup>e</sup> When there is no excess of low-pressure steam available for CO<sub>2</sub> recovery and drying, it is assumed that it is extracted from a steam turbine, which is often present at/near industrial areas to provide heat and power. The decrease in electricity production as a consequence of steam extraction is calculated similar as in [2] and added to the electricity requirements (for CO<sub>2</sub> capture, compression and cooling). The values between brackets represent the required electricity when there is an excess of low-pressure steam available to fulfil 100% of the steam requirements for CO<sub>2</sub> regeneration and drying.

<sup>f</sup> Assuming a CO<sub>2</sub> emission factor for a large gas-fired combined cycle of 0.43 kg/kWh.

## 5 ECONOMICS

### 5.1 CO<sub>2</sub> SUPPLY COSTS

CO<sub>2</sub> supply costs consist of compression, drying, cooling and transport costs and eventually CO<sub>2</sub> capture costs. Capital costs of a multistage compressor with a capacity of 250 t CO<sub>2</sub>/hr amount to 23 M€ to compress CO<sub>2</sub> from 1 to 80 bar [7]. A scale factor of 0.75 is used to correct capital costs for the required capacity [24]. Investment costs for onshore pipelines are 1040 €/km·mm<sup>15</sup> with a diameter between 0.1 and 0.6 m, specially designed for CO<sub>2</sub> transport [40]. Since pipeline construction costs depend on the terrain where the pipeline is constructed, capital costs are corrected using a terrain factor given in a report on CO<sub>2</sub> transmission [40]. O&M costs are a function of the pipeline diameter and length. Since the country where the project is realised has an impact on the capital and O&M costs, these costs are corrected for using a location factor given in [40], ranging from 1.2 for the UK to 0.7 for South Africa, Russia, China, India and Central Asia.

For the calculation of capture costs in case study 2, cost figures of the commercially applied Econamine FG Process given in [2] are used. Since the availability of waste steam is site and plant specific, it was decided to calculate capture costs for the case where steam needs to be extracted from a steam turbine (resulting in lower electricity production) and the case where the excess of steam is sufficient to cover the total heat requirements of the capture process.

### 5.2 COSTS AND REVENUES OF CO<sub>2</sub>-EOR

In the economic analysis of CO<sub>2</sub>-EOR, only injection well drilling costs are accounted for, since production wells are already present in oil fields under tertiary phase production. Well installation costs of 700 €/m were used after [23]. This is an average value from several literature sources for different well depths. Operating and maintenance costs of 3.24 €/bbl (800-1500 m), 4.06 €/bbl (1500-2500 m) and 4.86 €/bbl (>2500 m) reported in [8] are used. These costs are assumed to include costs for oil/water lift, separation of oil and CO<sub>2</sub>, compression and recycling of CO<sub>2</sub>, water cleaning & disposal and the relatively small costs of monitoring to ensure long-term sequestration.

A conservative oil price of 17.5 €/bbl is assumed for both cases, based on a value reported in [8]. Oil prices can fluctuate strongly; between 1990 and 2003, crude oil prices varied between 10 and 35 US\$/bbl [41]. We will address the impact of such fluctuations on CO<sub>2</sub> mitigation costs in the sensitivity analyses.

### 5.3 COSTS AND REVENUES OF CO<sub>2</sub>-ECBM

Costs for well drilling, production facilities, field infrastructure and operation were taken from a cost-benefit analysis of CBM recovery in Australia and New Zealand [42] and are presented in Table 5. Total well costs to drill CBM production wells and CO<sub>2</sub> injection wells were based on anticipated costs for a large-scale project. It is assumed that the

15. Investment costs of a pipeline depend on the length (km) and diameter (mm).

construction of the injection and production well starts 1 and 2 years before the actual injection starts.

For the Chinese case study, a natural gas price of 1.7 €/GJ is assumed, equal to the gas price used in the economic analysis of an ECBM project in the Qinshui basin in China [9]. For the Canadian case study, a gas price of 3 €/GJ is assumed.

TABLE 5. Cost data for CO<sub>2</sub>-ECBM from [42]. Costs are given for production and injection wells. Costs for injection wells are given in brackets

<i>Drilling and completion costs China (k€/well)</i>	
Drilling and completion costs <sup>a</sup>	379 (262)
Hydraulic fracturing	105
Inseam drilling (410 m) <sup>b</sup>	463
<i>Drilling and completion costs Canada (k€/well)</i>	
Drilling and completion costs <sup>a, c</sup>	200 (200)
Inseam drilling (410 m) <sup>b</sup>	463
<i>Production facilities and field infrastructure costs (k€/well) <sup>d</sup></i>	
Flowlines	87 (17)
Gas gathering lines	44 (44)
Gas delivery line	7 (7)
Satellite stations	44 (11)
Gas treatment and compression facilities	70
Engineering, administration and management	23 (7)
<i>Operating costs</i>	
Fixed costs (€/well-month)	1053
Variable costs	
Compression (€/10 <sup>3</sup> m <sup>3</sup> CH <sub>4</sub> )	4.15
Gas treatment (€/10 <sup>3</sup> m <sup>3</sup> CH <sub>4</sub> )	2.07
Water disposal (€/m <sup>3</sup> )	1.1

<sup>a</sup> It is assumed that these costs include costs for geological and geophysical expenditure.

<sup>b</sup> Inseam drilling costs of 1.13 k€/m as assumed in [24] are used in this study.

<sup>c</sup> Drilling, completion and stimulation (hydraulic fracturing) costs for several Canadian shallow gas wells have been determined by Massarotto [42]. The range lies between 80 and 240 k€/well, depending on the depth of the gas reservoirs (from 550-1330m) and whether well stimulation is applied. By extrapolating those costs for 1000 m, a value of 200 k€/well was found, which will be used for both production and injection wells. The drilling, completion and stimulation costs in Canada reported in [42] are very low. This is due to the scale of the operations in the area (over 1000 wells per year in low pressure gas reservoirs), the incentives of competition between service companies and contractors, and time to continuously improve all facets of design and operations [42]. So for a relatively small ECBM project, those costs will be higher, which is accounted for in the sensitivity analysis.

<sup>d</sup> The production facilities and field infrastructure requirements are based on a CBM producing well spacing of 64 ha (800x800 m), which dictates the costs for flowlines, satellite separation and gathering lines. The assumed well spacing in this study is 100 ha and therefore, a correction factor is applied to estimate the costs of these facilities.

#### 5.4 CO<sub>2</sub> MITIGATION COSTS

Based on CO<sub>2</sub> supply and storage costs and oil/gas revenues, CO<sub>2</sub> mitigation costs are calculated for each case study (see Table 6). The net present value (NPV) method is applied, since costs and revenues are not distributed equally in time, assuming an interest rate of 10%.

TABLE 6. Economics of case studies. The values in brackets represent the costs when the refinery has an excess of low pressure steam to cover 100% of steam requirements for CO<sub>2</sub> regeneration. The price of electricity is set at 0.06 €/kwh

Parameter	EOR-1 Saudi Arabia	EOR-2 USA	ECBM-3 China	ECBM-4 Canada
<i>Costs</i>				
Capture costs (€/t CO <sub>2</sub> removed)	-	36.5 (21.7) <sup>a</sup>	-	-
Compression costs (€/t CO <sub>2</sub> )	10.3	10.2	9.2	9.8
Transport costs (€/t CO <sub>2</sub> )	4.1	4.6	1.8	4.4
Total supply costs (€/t CO <sub>2</sub> )	14.4	51.3 (36.4)	11	14.2
PV CO <sub>2</sub> supply (M€)	26	100 (71)	52	29
<i>EOR</i>				
Injection well investment costs (M€)	20.2	21.6	-	-
Operating costs (M€/yr)	2.1	2.6	-	-
<i>ECBM</i>				
Injection wells investment costs (M€)	-	-	60	52.7
O&M costs injection wells (k€/yr)	-	-	617	705
Production well investment costs (M€)	-	-	16.3	12
O&M costs production wells (k€/yr)	-	-	453	529
PV storage costs (M€)	33	38	95	78
PV CO <sub>2</sub> supply and storage (M€)	59	137 (108)	146	106
<i>Revenues</i>				
PV oil/gas yield (M€)	-71	-79	-100	-80
CO <sub>2</sub> mitigation costs (€/t) <sup>b</sup>	-3	19 (9)	5	6

<sup>a</sup> The investment cost for a train size of 42 tonne CO<sub>2</sub> per hour for flue gases with a CO<sub>2</sub> concentration of 8% is 31.5 M€. The gas stream from the water gas shift reactor contains a higher CO<sub>2</sub> concentration; we estimated it at 20% based on the stoichiometric ratio of the overall reaction. The investment costs of the absorber are inversely proportional to the CO<sub>2</sub> concentration in the flue gas when these range from 4 to 8%, but we assumed this relation can be applied as well for CO<sub>2</sub> concentration of 20%. A scaling factor of 0.8 is used to correct the investment costs for the scale of the absorber. Annual O&M costs are assumed to be 3.6% of the investment plus the cost for purchase of chemicals and disposal of chemicals. MEA is partly entrained in the gas phase, resulting in a consumption of 2.9 €/t CO<sub>2</sub> recovered, assuming no SO<sub>2</sub> in the gas stream [2]. Since natural gas undergoes desulphurisation before reforming, the SO<sub>2</sub> content is expected to be negligible. Used MEA can be disposed of by combustion in refuse incinerators, which costs around 0.29 €/t CO<sub>2</sub> recovered [2].

<sup>b</sup> These costs are different than the costs presented in the PEACS study [20], because several adaptations and improvements in the cost calculations have been made (e.g. capture costs for case 2 are accounted for).

It can be concluded carefully that CO<sub>2</sub>-EOR is economically feasible for the assumed conditions, when the CO<sub>2</sub> stream is pure (as in case study 1). The economics for case study 2, a hydrogen plant with a PSA unit in which CO<sub>2</sub> must be captured, are less attractive. Capture costs can be reduced significantly when there is an excess of waste steam at the refinery to cover the steam requirements for CO<sub>2</sub> regeneration. The costs of case study 1 and 2 should be considered as indicative values, because of the rather simplistic assumption on the oil production profile and uncertainties in costs related to capture and storage. However, the values do not seem unreasonable when comparing them to a storage cost curve for CO<sub>2</sub>-EOR given in [8], which shows that about 120 Gt can

be sequestered in depleted oil fields and CO<sub>2</sub>-EOR projects in oil fields at net savings between -25 and 0 \$/t CO<sub>2</sub>, assuming CO<sub>2</sub> supply costs are 0. If the CO<sub>2</sub> supply costs are assumed to be 0, mitigation costs of case 1 are about -11 €/t CO<sub>2</sub>.

Mitigation costs of the CO<sub>2</sub>-ECBM case studies appeared to be higher than for the CO<sub>2</sub>-EOR case studies, which can mainly be attributed to the large investment costs of the production wells (see Figure 6). The economics of worldwide CO<sub>2</sub> sequestration using ECBM presented in [9] show that the CO<sub>2</sub> sequestration potential which is profitable, is relatively small. In order to become economically feasible, the project developer needs an incentive, i.e. in the form of carbon credits, CO<sub>2</sub> tax or an incentive for production of climate neutral gas or electricity (when electricity is generated from CBM).

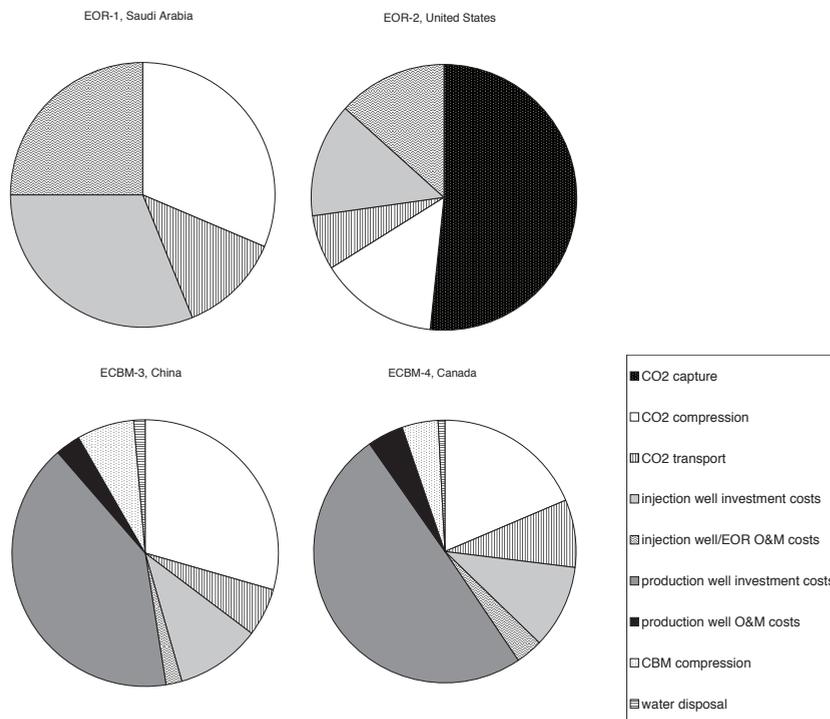


FIGURE 6. Present value cost breakdown of the considered case studies

## 5.5 SENSITIVITY ANALYSIS

A sensitivity analysis was performed to determine the impact of the variation in several parameters on mitigation costs in case study 2 (CO<sub>2</sub>-EOR in California) and case study 4 (CO<sub>2</sub>-ECBM in Canada) (see Figure 7 and 8). The economic feasibility of the CO<sub>2</sub>-EOR project depends strongly on the oil price. At high oil prices (around 40 \$/bbl), it might even be feasible to capture CO<sub>2</sub> from refineries (with PSA) for CO<sub>2</sub>-EOR purposes. The mitigation costs for the CO<sub>2</sub>-ECBM case study are sensitive to the interest rate and the investment costs of the wells, given the large capital

investment associated with the construction of the dense well network. Figure 8 also illustrates that the prospects for CO<sub>2</sub>-ECBM can be improved greatly when gas prices increase, which is not unlikely for China in the foreseeable future.

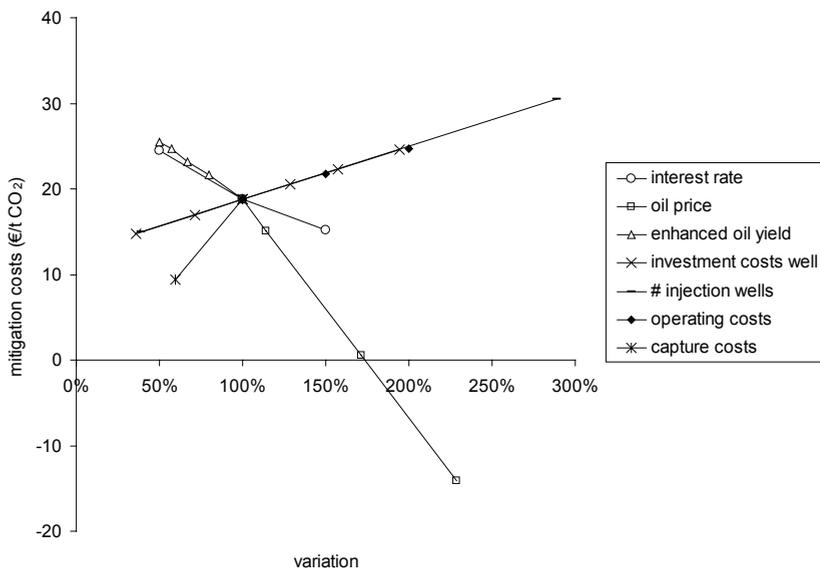


FIGURE 7. Spider diagram EOR-2, USA

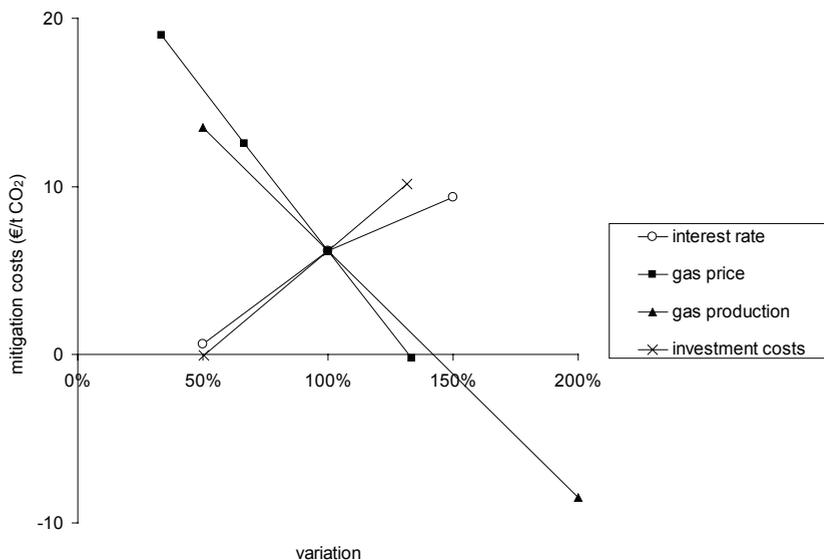


FIGURE 8. Spider diagram ECBM-4, Canada

6 DISCUSSION AND CONCLUSIONS

The objective of this study was to provide an overview of potential

worldwide early opportunities for CO<sub>2</sub> sequestration. The combination of a GIS tool to deal with large databases of CO<sub>2</sub> sources and reservoirs and a Multi-Criteria Analysis as a screening tool to identify promising combinations is useful for this purpose. The results can be used for selecting regions for more detailed data collection.

The analysis shows that many potentially promising EOR projects are located in the USA, which can be explained by the use of more detailed units to define oil occurrences in the USA, resulting in relatively many oil occurrences. Other promising source-reservoir combinations are located in Saudi Arabia, Canada and Mexico.

The most promising ECBM cases consist of Chinese, European, a few Canadian, a Japanese, Australian and an Indonesian (re)source. Especially China shows a high techno-economic potential for ECBM projects.

From these potential early opportunities, four illustrative case studies were considered in more detail. Case 1 consists of a hydrogen plant in Saudi Arabia, which could sequester 0.26 Mt/yr CO<sub>2</sub> in a depleted oil field at net savings of approximately 3 €/t CO<sub>2</sub>. EOR case 2 is a hydrogen plant in California, USA, which has to be retrofitted in order to generate a pure CO<sub>2</sub> stream. Approximately 0.28 Mt CO<sub>2</sub> could be stored annually at 9-19 €/t CO<sub>2</sub>, depending on the availability of steam for CO<sub>2</sub> regeneration. In case 3 and 4, circa 0.68 and 0.29 Mt CO<sub>2</sub> from ammonia plants in China and Canada could be sequestered annually in coal fields at approximately 5 and 6 €/t CO<sub>2</sub>, respectively. Due to the lack of specific data and the somewhat generic data and assumptions used for the economic analysis, the mitigation costs should be considered as indications for those cases. The main inaccuracies in the cost figures are well drilling and operating costs and the oil production profile. In order to perform a more accurate calculation of mitigation costs for the CO<sub>2</sub>-EOR cases, a more realistic model to determine the oil production profile and CO<sub>2</sub> recycling and injection rate is required. Finally, it should be kept in mind that ECBM still has to prove itself commercially, making it hard to draw firm conclusions on ECBM economics.

The analysis performed in this study must be considered as a quick scanning tool with best available data to provide a first overview of promising cases. The method has limitations, which are mainly caused by the lack of detailed, site-specific information and by the subjectivity of the criteria applied in the MCA, and related weight factors.

The lack of more detailed information played a critical role in the choice and further analysis of the selected cases. The databases used in the analysis do not always contain all relevant information required to generate reliable results. The CO<sub>2</sub> source database is very comprehensive, but an important feature such as start-up year of a plant (to get an indication whether the plant will not be shut down or retrofitted within several years) is often missing and reported technologies are not always up-to-date. The retrofit and technology issue is relevant for hydrogen and ammonia plants, since older units are generally being upgraded with a PSA unit, generating a diluted CO<sub>2</sub>

stream. The economic performance (with respect to CO<sub>2</sub> sequestration) of those configurations is considerably lower than conventional units in which a pure CO<sub>2</sub> stream is produced and therefore, these facilities are less suited as early opportunity project. The reservoir databases contain many oil and coal fields, but with the available information, it cannot be assessed in detail whether a certain reservoir is suited for CO<sub>2</sub> sequestration. A specific oil field might be in its primary production phase making CO<sub>2</sub>-EOR unrealistic or the permeability of a coal seam could be so low that CO<sub>2</sub> injection is not feasible. Since several relevant features of source and reservoir are not included in the databases, the method used in this study bears the risk that the selected cases are not feasible for CO<sub>2</sub> sequestration. Consequently, the selected cases should be considered as illustrative potential early opportunities. It is recommended that the databases are updated and more specific information on the selected sources and reservoirs is added to assess whether source and reservoir conditions are indeed suited for short-term CO<sub>2</sub> sequestration. Sources should be checked (by contacting plant managers) to ensure a pure CO<sub>2</sub> supply in the project lifetime could be guaranteed. The selected reservoirs should be screened in more detail to assess whether they are appropriate storage media and enable enhanced hydrocarbon recovery.

The MCA contains some subjective elements, among which the choice of the criteria that should be included in the analysis ('what are decisive factors?'). There are other criteria/aspects that should play a role in judging whether a project is feasible, such as the assurance of an extensive, competent cap rock, identification of existing wells, ease of permitting [15], environmental impacts and the local oil and gas market. Important reservoir screening criteria for CO<sub>2</sub>-ECBM not included are reservoir homogeneity, minimal faulting/folding, optimal depth range (300-1500 m), concentrated coal geometry, adequate permeability and the existence of already established commercial CBM production [9]. Also the sequence of importance ('is the potential oil yield more important than the drilling depth?') and the assessment of the values of each weight factor remains a rather arbitrary matter. In order to minimise the subjectivity in the weight factor assessment, we performed the analysis with different weight factor sets. By comparing the outcome of each weight factor set, cases with a high overall, technical and socio-economic score can be analysed. It appeared that the outcome is not altered heavily when the sequence/value of the weight factors are varied; the ranking will change, but the most promising cases remain present in the higher regions of the ranking.

There are other choices made in this study, which might exclude promising early opportunities, among which:

- Gas processing plants are not included, because the CO<sub>2</sub> concentration in the waste steam is not reported in the database used in this study. Gas processing plants generally emit a large amount of almost pure CO<sub>2</sub> and are therefore important sources for EOR operations and ongoing/planned sequestration projects.

–Gas fields are not accounted for in this study. Although economically not as promising as CO<sub>2</sub>-EOR, CO<sub>2</sub> storage in disused gas fields should be considered in more detail, considering the relatively high storage potential estimated at 800 Gt CO<sub>2</sub> and relatively low storage costs. Some 100 Gt CO<sub>2</sub> can be stored at 7 \$/t and a further 575 Gt at costs of 10-17 \$/t [8].

The method that has been developed allows for easy updates (with other sources such as natural gas processing plants, reservoirs and MCA criteria) and addition of site-specific information, which therefore provide us with a dynamic screening tool to assess global CO<sub>2</sub> sequestration options.

Finally, it is important to realise that only about 6% of total global industrial emissions (including power plants) are caused by ammonia, hydrogen and ethylene oxide production plants [16]. The potential contribution of CO<sub>2</sub> sequestration from high-purity sources to total CO<sub>2</sub> reductions is therefore limited. The bulk of CO<sub>2</sub> reductions should be realised at power plants, involving higher capture costs.

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## CHAPTER VI

### HEALTH, SAFETY AND ENVIRONMENTAL RISKS OF UNDERGROUND CO<sub>2</sub> STORAGE - OVERVIEW OF MECHANISMS AND CURRENT KNOWLEDGE \*

\*  
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#### ABSTRACT

CO<sub>2</sub> capture and storage (CCS) in geological reservoirs may be part of a strategy to reduce global anthropogenic CO<sub>2</sub> emissions. Insight into the risks associated with underground CO<sub>2</sub> storage is needed to ensure that it can be applied as safe and effective greenhouse gas mitigation option. This chapter aims to give an overview of the current (gaps in) knowledge of risks associated with underground CO<sub>2</sub> storage and research areas that need to be addressed to increase our understanding in those risks. Risks caused by a failure in surface installations are understood and can be minimised by risk abatement technologies and safety measures. The risks caused by underground CO<sub>2</sub> storage (CO<sub>2</sub> and CH<sub>4</sub> leakage, seismicity, ground movement and brine displacement) are less well understood. The main R&D objective is to determine the processes controlling leakage through/along wells, faults and fractures to assess leakage rates and to assess the effects on (marine) ecosystems. Although R&D activities currently being undertaken are working on these issues, it is expected that further demonstration projects and experimental work is needed to provide data for more thorough risk assessment.

## I INTRODUCTION

In order to stabilise the concentration of greenhouse gasses in the atmosphere, many countries have committed themselves to reduce their greenhouse gas emissions. These emissions are dominated by CO<sub>2</sub> and to a large extent related to fossil fuel use. Reduction of CO<sub>2</sub> emissions can be realised by means of a diverse portfolio of options covering energy and material efficiency improvements, afforestation, increased use of renewable and nuclear energy and decrease of the carbon intensity of fossil fuels. The latter option comprises a shift from coal towards gas and carbon dioxide removal, in which CO<sub>2</sub> emitted at stationary sources is captured and stored in geological reservoirs or the ocean. It is becoming clear that energy and material efficiency improvements and the increased use of renewable energy sources cannot achieve the emission reductions required to reach long-term atmospheric stabilisation targets below 550 ppmv CO<sub>2</sub> [1]. The use of nuclear energy meets public resistance in many countries. Given the large amounts of fossil fuels (especially coal) that can be extracted at low costs, cleaner use of fossil fuels by capturing and storing CO<sub>2</sub> is considered to be a potential element of a strategy to substantially reduce global anthropogenic CO<sub>2</sub> emissions in the coming decades [1-4]. The technical potential or capacity (see Table 1) is sufficient to store worldwide emissions for several decades up to several hundred years.

TABLE I. Geological reservoir types and estimated global storage capacity [5-8]

Reservoir type	Global capacity (Gt CO <sub>2</sub> )
Depleted oil and gas fields <sup>a</sup>	920
Deep saline aquifers	240-10,000
Unminable coal seams <sup>b</sup>	40-270

<sup>a</sup> Including oil and gas fields approaching the end of their economically productive life (by primary and secondary production). In these reservoirs, injection of CO<sub>2</sub> may enhance the oil/gas yield (enhanced oil/gas recovery or EOR/EGR).

<sup>b</sup> In these reservoirs, injection of CO<sub>2</sub> can result in the production of coal bed methane (enhanced coal bed methane recovery or ECBM).

A key factor affecting the implementation of CCS is the risks associated with underground CO<sub>2</sub> storage. Gaining a better understanding and quantification of these risks is needed to ensure that they will comply with safety standards (also after injection has been completed). Risk assessment is a first step in a strategy to set up management and control measures to minimise risks of underground CO<sub>2</sub> storage. Also, it helps to facilitate the formulation of standards and regulatory frameworks required for large-scale application of CCS. To date, a wide variety of activities studying the risks of underground CO<sub>2</sub> storage has been completed and is being performed. The risks associated with underground CO<sub>2</sub> storage have been discussed extensively in an EU study on underground disposal of CO<sub>2</sub> [9]. However, in the meantime new insights have been obtained into the risks of CCS.

The objective of this study is to give an overview of the knowledge and especially the gaps in knowledge with regard to health, safety and environmental risks of CCS, useful for policymakers to prioritise R&D, set standards and define strategies. The overview is based on a review of scientific literature and information gained from R&D projects, supported by expert consultation.

Health, safety and environmental risks can be caused by operation of surface and injection installations and by storage of CO<sub>2</sub> in a geological reservoir. CO<sub>2</sub> capture and compression are commonly applied technologies in industry to produce high-purity CO<sub>2</sub> for various industrial applications (enhanced oil recovery, carbonisation of beverages, cooling, drinking water treatment, welding, foam production). The associated risks are considered to be acceptable according current industry standards and therefore not discussed here. The risks associated with surface and injection installations are discussed very briefly. We will focus on the risks associated with CO<sub>2</sub> storage in geological formations. The risks associated with CO<sub>2</sub> storage in the ocean are not considered.

The core of this chapter describes the (gaps in) knowledge on the risks that may occur when transporting, injecting and storing CO<sub>2</sub> in geological formations (Section 2 and 3). Section 4 discusses the results of various risk assessments. The knowledge that can be obtained from experience with industrial and natural analogues, which show similarities with underground CO<sub>2</sub> storage, is considered in Section 5. This knowledge is used to get insight into factors and processes that may be (ir)relevant to the risks of underground CO<sub>2</sub> storage. In Section 6, knowledge gaps are summarised and compared to an overview of ongoing R&D activities. Subsequently, research areas and priorities can be formulated to reduce gaps in knowledge.

## 2 RISKS ASSOCIATED WITH SURFACE AND INJECTION INSTALLATIONS

1. Although CO<sub>2</sub> transport generally occurs by high-pressure pipelines, transport by tankers might be a viable option for offshore reservoirs.

2. Note that these pipelines are mainly sited in areas of low to medium population density.

Surface and injection installations comprise a transmission pipeline<sup>1</sup>, (booster stations), CO<sub>2</sub> delivery station, a pipeline distribution network, injection well(s) and a monitoring system. When hydrocarbons are produced simultaneously, the system also comprises production wells and surface facilities to produce, clean, compress and transport the extracted hydrocarbons.

CO<sub>2</sub> produced from natural CO<sub>2</sub> fields or captured from industrial facilities is transported and injected on a commercial scale for enhanced oil recovery (CO<sub>2</sub>-EOR), principally in the USA. Worldwide, approximately 3100 km of pipeline exists with a capacity of circa 45 mt CO<sub>2</sub>/yr [10]. The major risk associated with pipeline transport is a pipeline failure, which can be either a (pin)hole or rupture, resulting in CO<sub>2</sub> release. The accident record for CO<sub>2</sub> pipelines in the USA shows 10 accidents from 1990 to 2001 without any injuries or fatalities<sup>2</sup>,

corresponding to a frequency of  $3.2 \cdot 10^{-4}$  incidents per km per year [10]. Statistics of incidents with natural gas and hazardous liquid pipelines between 1986 and 2001 in the USA show a frequency of  $1.7 \cdot 10^{-4}$  and  $8.2 \cdot 10^{-4}$  per km per year, respectively [10]. Since CO<sub>2</sub> is not explosive or inflammable, the consequences in case of leakage are expected to be less severe than for natural gas. However, in contrast to natural gas, which is dispersed quickly into the air, CO<sub>2</sub> might cause dangerous situations when it is able to accumulate in confined spaces, as it is denser than air (see Section 3.1.2). The possible consequences of a rupture of a buried pipeline transporting 250 t liquid CO<sub>2</sub>/hr at 60 bar have been analysed in [11]. The results of the model indicate that the safety distances to the pipe at which concentrations of minimal 5% will occur for 60 seconds, lie between 600 and 150 m, depending on the distance between safety valves.

The major risk associated with injection is a well failure, which may result in escape of CO<sub>2</sub> that will migrate upwards due to its relatively low density in comparison to water. The likelihood of a sudden escape of all CO<sub>2</sub> stored in an underground reservoir is very small due to the limited capacity of the injection system [9]. In the majority of well failures, an amount equal to the content of the well tubing will be released. In normal cases, this leak will be detected by the monitoring system, resulting in the closure of the back flow preventer and the emergency shutdown valve at the well head [9].

Failure of the back-flow preventer or packer may result in a well blowout [9]. A blowout is an uncontrolled flow of reservoir fluids (which can be CO<sub>2</sub>, but also salt water, oil, gas or a mixture of these) into the well bore to the surface. Apart from CO<sub>2</sub> release, the potential consequences are casualties among operators and economic damage caused by explosion or fire when upcoming hydrocarbons are ignited or by parts of the well, which can be launched by the pressure release. The frequency of blowouts from offshore gas wells has been estimated at  $1 \cdot 10^{-4}$  per well year, based on a database of blowouts in the Gulf of Mexico and the North Sea between 1980 and 1996 [12]. Other estimates based on both oil and gas wells give a frequency of  $3 \cdot 10^{-4}$  per well year [13].

Summarising, there is a lot of industrial experience with extraction, processing, transport and injection of CO<sub>2</sub>. Additionally, the experience with hydrocarbons and other chemicals is partly applicable to CO<sub>2</sub> transport and injection. Industrial experience with CO<sub>2</sub> and other gases shows that the risks from industrial facilities are manageable using standard engineering controls and procedures [14].

### 3 RISKS ASSOCIATED WITH CO<sub>2</sub> STORAGE IN GEOLOGICAL RESERVOIRS

The risks of CO<sub>2</sub> storage in a geological reservoir can be divided into 5 categories (see Figure 1):

- *CO<sub>2</sub> leakage*: CO<sub>2</sub> migration out of the reservoir to other formations, from where it may escape into the atmosphere.
- *CH<sub>4</sub> leakage*: CO<sub>2</sub> injection might cause CH<sub>4</sub> present in the reservoir to migrate out of the reservoir to other formations and possibly into the atmosphere.
- *Seismicity*: The occurrence of (micro) earth tremors caused by CO<sub>2</sub> injection.
- *Ground movement*: Subsidence or uplift of the earth surface as a consequence of pressure changes induced by CO<sub>2</sub> injection.
- *Displacement of brine*: Flow of brine to other formations (possibly sweet water formations) caused by injection of CO<sub>2</sub> in open aquifers.

We will mainly focus on the risk of CO<sub>2</sub> leakage, for which the mechanisms, local and global effects will be discussed extensively in Sections 3.1.1 to 3.1.3. The mechanisms and effects of the other risks are discussed in less detail.

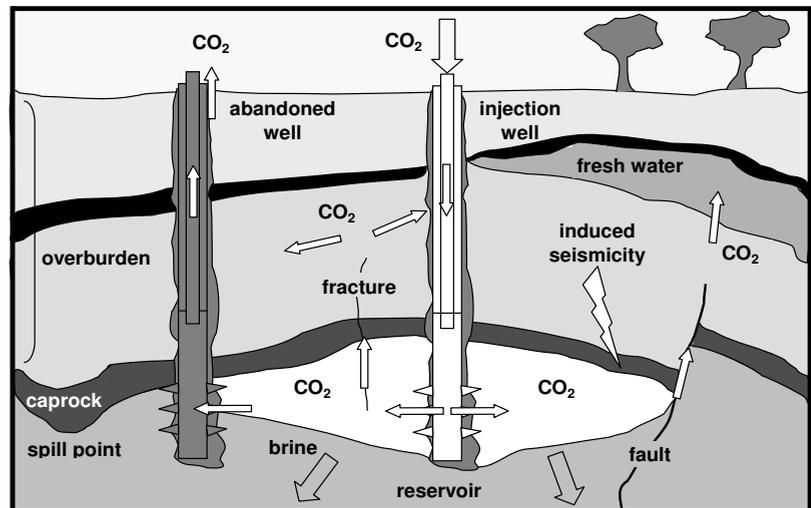


FIGURE 1. Risks of underground CO<sub>2</sub> storage. White arrows represent CO<sub>2</sub> flows (along fractures, abandoned wells and faults). Grey arrows represent brine displacement as a consequence of CO<sub>2</sub> injection

### 3.1 CO<sub>2</sub> LEAKAGE

When CO<sub>2</sub> is injected into geological reservoirs, it might potentially migrate out of the reservoir through the subsurface, migrate laterally in overburden formations and finally leak into the atmosphere/biosphere. The potential for leakage will depend on well and cap rock (seal) integrity and the trapping mechanism. CO<sub>2</sub> can be retained in reservoirs by means of the following trapping mechanisms [15-18]:

- When injecting CO<sub>2</sub> in a hydrocarbon reservoir, gaseous or supercritical CO<sub>2</sub> will rise up due to buoyancy effects. The presence of geological traps such as a low-permeable cap rock will prevent the CO<sub>2</sub> to migrate further; CO<sub>2</sub> will accumulate under the cap rock.
- CO<sub>2</sub> injected in deep saline aquifers might take thousands to millions

of years to migrate from injection point to the surface due to the extremely low flow rates encountered in these formations (hydrodynamic trapping).

- CO<sub>2</sub> can be trapped in the pore space by capillary forces (residual gas trapping).
- Since CO<sub>2</sub> is highly soluble in water and also dissolves in oil, solubility trapping is an important trapping mechanism. When injecting CO<sub>2</sub> into an aquifer, CO<sub>2</sub> will mainly be present as supercritical fluid before it fully dissolves. Model calculations of CO<sub>2</sub> injection in an Australian formation indicate that complete dissolution is expected to take place on a time scale ranging from 10,000 to 100,000 years [19]. Simulations of CO<sub>2</sub> injection into the Utsira formation at the Sleipner site<sup>3</sup> suggest that CO<sub>2</sub> will be dissolved completely after 5000 to 50,000 years [20]. When the CO<sub>2</sub> is completely dissolved, leakage is no longer possible, since free CO<sub>2</sub> is not present anymore, provided that no CO<sub>2</sub> is released as a consequence of pressure and temperature changes in the reservoir.
- Dissolved CO<sub>2</sub> can react with silicates and carbonates to form bicarbonate or carbonate ions (ionic trapping).
- CO<sub>2</sub> can also react with minerals and organic matter present in the geologic formations to become part of the solid matrix, also referred to as mineral trapping. This is the most secure form of trapping. However, the extent to which injected CO<sub>2</sub> reacts with minerals present in either sandstone or carbonate reservoirs is considered to be low. Reservoir simulations of aquifers similar to the Utsira formation at Sleipner revealed that less than 1% precipitates as carbonate minerals after 20 years [21]. In the longer term, limited additional reaction is expected [13].
- In coal seams, CO<sub>2</sub> will be trapped by adsorption to the coal surface, displacing adsorbed methane, and by physical trapping in the cleats within the coal. Due to adsorption to the coal surface, less 'free' CO<sub>2</sub> is present. Consequently, the risk of leakage in coal seams is expected to be smaller than for hydrocarbon reservoirs and deep saline aquifers, where CO<sub>2</sub> is predominantly present in free state in the first phase after injection.

3. The Sleipner project is the first commercial-scale CO<sub>2</sub> storage project. Since 1996, annually circa 1 million tonnes of CO<sub>2</sub> removed from natural gas is injected into the Utsira formation, a saline aquifer 1000 m below the floor of the North Sea. A total of 20 Mt is expected to be stored over the project lifetime.

The permeability of the overburden, the formations above the target reservoir, is another critical factor for leakage, since it determines the retention time of CO<sub>2</sub> in the subsurface. Simulation of CO<sub>2</sub> diffusion through the 700 m overburden above the Utsira formation at Sleipner indicates that it will take more than 500,000 years for CO<sub>2</sub> to reach the sea floor [20]. A model simulating the release of CO<sub>2</sub> from an aquifer at 1000 m depth in the Netherlands indicated a breakthrough time of CO<sub>2</sub> to reach the surface of about 5500 years assuming a permeability of 1 mDarcy [9].

### 3.1.1 MECHANISMS OF CO<sub>2</sub> LEAKAGE

#### *Depleted oil and gas fields*

Hydrocarbon reservoirs, which generally have been well researched, are considered to be safe sinks for CO<sub>2</sub> storage, since these media have held oil/gas for geological timescales (i.e. millions of years) without major incidents of sudden migration. Many gas reservoirs are holding significant quantities of CO<sub>2</sub> as well, giving further confidence that CO<sub>2</sub> can be stored safely without large releases of CO<sub>2</sub>. However, there is a risk that CO<sub>2</sub> escapes from the reservoir through or along wells or by means of a cap rock failure. CO<sub>2</sub> might also escape via spill points<sup>4</sup> or dissolve in fluid flows in the reservoir rock beneath the CO<sub>2</sub> accumulation to surrounding formations.

4. The structurally lowest point in a hydrocarbon reservoir (see Figure 1). Once a reservoir has been filled to its spill point, CO<sub>2</sub> or hydrocarbons will spill or leak out.

CO<sub>2</sub> leakage through or along wells after the injection phase can be caused by casing or cementation defects due to improper design or construction, corrosion of the casing and deterioration of cement plugs by CO<sub>2</sub> and/or brine. Abandoned wells can be an important migration pathway, since depleted oil/gas reservoirs are generally 'punctured' by a large number of non-operative wells, some of them in bad condition. Especially unidentified and poorly (improperly plugged) abandoned wells are potential point sources. Moreover, control and maintenance of abandoned wells is a complex matter in several regions in the world. Diffusion of CO<sub>2</sub> through the cement or steel casing is a process, which will progress very slowly (in the order of 20 cm in 100 years) [22]. However, it is uncertain how the well bore integrity (the cement and casing) is affected by CO<sub>2</sub> and brine considering a storage timescale of 100's to 10,000's of years. Degradation may affect the permeability of cement, which might increase leakage rates in time. Over long time scales, wells may thus serve as preferential leakage pathways and may therefore represent a significant (long-term) risk [23].

In order to assess potential leakage of a certain reservoir, detailed information must be available on the number, type and age of wells, completion technique and type of materials used. This information will not always be available in some regions, making it hard to get a quantitative estimation of leakage potential.

A cap rock failure is a generic term for various mechanisms described below:

- Capillary leakage occurs when the pressure difference of fluid phase and the water phase in the pores adjacent to the cap rock is higher than the capillary entry pressure of the cap rock. Since the capillary entry pressure of a cap rock has generally been sufficient to retain hydrocarbons and the capillary entry pressure can be measured, capillary leakage of CO<sub>2</sub> is not considered to be a problem [24].
- Diffusion of CO<sub>2</sub> through the cap rock is expected to be a very slow process, but can be the controlling mechanism for leakage in the long term [24].
- CO<sub>2</sub> might leak through man-made fractures, also referred to as hydraulic fracturing. Fractures can be created by over pressuring the

reservoir. Fractures could be sealed in time by precipitation of newly formed minerals, but could also be re-opened as a consequence of new changes in stresses during storage of CO<sub>2</sub> [24]. Also earlier production/injection processes to exploit hydrocarbon reservoirs may have created fractures. In the selection of a suitable reservoir, the impact of primary, secondary and tertiary recovery processes<sup>5</sup> on the hydraulic integrity of the bounding seals should therefore be assessed [24].

In order to prevent fracturing, the maximum injection pressure should always be kept below the level at which the cap rock may shear (fracture pressure) [25]. The risk of leakage through fracturing is low as long as the storage pressure does not exceed the initial reservoir pressure. However, there is a certain level of overpressure, at which CO<sub>2</sub> can be safely contained. This 'safety factor' depends on the stress state of the cap rock, which depends on depth, pore pressure, rock properties and sedimentary and tectonic history. The maximum injection pressure can be predicted by determining the in-situ stress profile [9].

- Dilatant<sup>6</sup> shear formation and fracturing may occur in cap rocks, which can ultimately create preferential flow paths and increasing the cap rock permeability and consequently the risk of CO<sub>2</sub> leakage. However, shear deformation can also result in a reduced permeability [24].
- High-permeability zones might exist or be formed by reaction of CO<sub>2</sub> with the cap rock, causing the cap rock to dissolve. CO<sub>2</sub> can also dehydrate clay shales in the cap rock, thereby increasing its permeability.
- CO<sub>2</sub> might leak through open (non-sealing) faults, which extend into the cap rock. The risk of leakage along faults can be minimised by performing a detailed analysis of the geological setting of the reservoir prior to injection and selecting only those reservoirs with no/minimal faulting.
- Seismic disturbances might cause cap rock failure [26].

Of these mechanisms, leakage along or through wells, faults and fractures are generally considered to be the most important leakage pathways. However, there is still a lack of understanding in the physics of CO<sub>2</sub> leakage (i.e. the processes that control leakage) through wells and faults. Also, there is a lack of data to close the knowledge gap [27].

### *Deep saline aquifers*

Leakage from deep saline aquifers basically occurs via the same mechanisms as discussed above. A major difference with hydrocarbon reservoirs is that aquifers generally do not have cap rocks or seals that have stood the test of time in retaining gasses. Since deep saline aquifers are not of economical interest such as hydrocarbon-reservoirs, the number of wells penetrating aquifers, and consequently the potential for CO<sub>2</sub> leakage through/along wells is lower in comparison to

5. After primary recovery (pressure depletion) and secondary recovery (waterflooding, injection of water), the oil recovery can be increased by means of tertiary recovery techniques such as CO<sub>2</sub> or thermal-EOR

6. Dilatancy is the increase in the volume of rocks as a result of deformation.

hydrocarbon fields.

Another difference with hydrocarbon reservoirs is the fact that CO<sub>2</sub> storage in an aquifer will induce a (temporary) pressure increase in the reservoir, because the space to store CO<sub>2</sub> only becomes available as a result of compression of the fluids and rock in the reservoir, or displacement of formation water into adjacent formations or to the surface [9]. This pressure increase might trigger fracture zones, which might end up in CO<sub>2</sub> migrating upwards [25]. Deep saline aquifers have not been researched that well as hydrocarbon reservoirs. An extensive research programme, the completed SACS project followed by the ongoing CO<sub>2</sub>STORE project, has been set up to study and monitor CO<sub>2</sub> injected in the Utsira aquifer at Sleipner. Seismic surveys that have been acquired so far (latest in 2002) do not indicate any CO<sub>2</sub> leakage to levels shallower than the Utsira Sand [28]. Diffusion through the cap rock is expected to be the main leakage transport mechanism, although this is expected to be very slow; modelling indicates that this process will take millions of years [20]. Reactive transport model simulations of Sleipner indicate that after 120 years, mineral precipitation caused by CO<sub>2</sub> will have decreased the porosity and permeability of the cap rock base from 5% to 2.3% and from 3 to 0.3 mDarcy, respectively [21]. These results suggest that the sealing properties of the cap rock are enhanced by CO<sub>2</sub> storage.

#### *Unminable coal seams*

Coal seams are unique in the sense that injected CO<sub>2</sub> is to a large extent adsorbed to the coal matrix, replacing coal bed methane, as CO<sub>2</sub> is more strongly adsorbed to coal than methane. In general, it is argued that if coal seams have held methane for millions of years, it will probably retain CO<sub>2</sub> for similar timescales.

However, there are still several aspects to be studied on the interaction between CO<sub>2</sub> and coal seams. Especially the chemical reactions and physical processes that could occur during CO<sub>2</sub> injection into coal seams and their impact on the integrity of the coal seams are not well understood. One of these reactions is swelling of the coal matrix when injecting CO<sub>2</sub>, which might cause a reduction in the permeability. Swelling might also induce stresses on the overlying and underlying rock strata in non ideal coal seams (thin, low permeable and highly faulted), that could cause faulting and possible migration pathways out of the coal seam [29].

#### 3.1.2 LOCAL EFFECTS OF CO<sub>2</sub> LEAKAGE

CO<sub>2</sub> might cause health effects when exposed to humans, animals and ecosystems at elevated concentrations. Health effects of elevated CO<sub>2</sub> concentrations on human beings and animals are well understood. Prolonged exposure to high CO<sub>2</sub> levels, above 20–30%, will cause death by suffocation to humans and the majority of air-breathing animals [14]. Deaths from catastrophic releases of CO<sub>2</sub> are known from industrial accidents and natural disasters in volcanic areas. The sudden release of

0.24 mt CO<sub>2</sub> from Lake Nyos (Cameroon) in 1986 caused the deaths of at least 1700 people and thousands of animals. The most widely accepted hypothesis to explain the sudden release is an overturn of the deep lake as the bottom part became oversaturated with CO<sub>2</sub>, caused by a slow leak of CO<sub>2</sub> from magmatic sources into the deep lake waters [30].

Although a spontaneous release as occurred at Lake Nyos is no analogue for CO<sub>2</sub> leakage from a geological reservoir, a similar situation could occur in which anthropogenic CO<sub>2</sub> leaking from a geological reservoir accumulates in a deep lake. This can be prevented by selecting reservoirs without any lakes in vicinity or with proven connection to lakes. If leaking CO<sub>2</sub> nonetheless accumulates in a lake, the hazard can be mitigated by degassing the lake [14].

The incident at Lake Nyos illustrates that the health hazard caused by CO<sub>2</sub> releases depends also on the nature of release and not only on the total volume released. Since CO<sub>2</sub> is heavier than air, leakage of relatively small quantities of CO<sub>2</sub> poses a lethal threat when CO<sub>2</sub> is able to accumulate in confined spaces such as valleys or cellars. The topography around Lake Nyos played a crucial role in the disaster, since it provides ideal conditions for the emitted CO<sub>2</sub> cloud to remain concentrated rather than disperse [30]. Atmospheric dispersion modelling indicates that small, isolated leaks (~0.1-100 g/m<sup>2</sup>/day) will quickly disperse under typical topographic conditions and wind velocities, not posing lethal threats [27].

Obviously, the potential impact on human beings of a sudden release of CO<sub>2</sub> from an offshore reservoir will be lower than from an onshore reservoir. From that perspective, offshore reservoirs deserve preference in selecting reservoirs for storage practices.

In general, the environmental and ecological effects are less well understood as health effects on humans, although natural CO<sub>2</sub> leaks have provided insight into the relation between leakage rates and environmental/ecological impacts.

Fresh, potable groundwater, located in the top 100-200 m of the subsurface, could be contaminated by leakage of CO<sub>2</sub>. Even small CO<sub>2</sub> leaks may possibly cause significant deteriorations in the quality of potable groundwater. An increase in CO<sub>2</sub> concentration might cause a decrease in pH to a level of 4-5, which might cause calcium dissolution, increase in the hardness of water and change in the concentration of trace elements [9]. A model simulating the pH change and the enhanced dissolution of trace metals caused by CO<sub>2</sub> dissolution shows that in poorly buffered aquifers, trace metals can be released (by dissolution/desorption) to levels that exceed drinking water standards [31].

However, mineral dissolution kinetics, an important parameter affecting trace metal concentration, have not been characterised completely yet. Also surface water could be contaminated by leakage, which could affect aquatic ecosystems by decreasing the pH, especially in stagnant or stably stratified waters [14].

Elevation of CO<sub>2</sub> concentrations in the soil due to leakage is likely to lower the soil pH, and adversely impact the chemistry of nutrients, redox

sensitive elements and trace metals, as well as plant growth [26]. Plants usually have a higher resistance against CO<sub>2</sub> than mammals, but persistent leaks could suppress respiration in the root zone. Tree kills associated with soil gas concentrations in the range of 20 to 30% CO<sub>2</sub> have been observed at Mammoth Mountain, California, where volcanic outgassing of CO<sub>2</sub> (0.13-0.44 Mt CO<sub>2</sub>/yr) has been occurring since at least 1990 [14].

The effects of CO<sub>2</sub> on subsurface organisms dwelling in deep geologic formations and the effects on marine ecosystems are not well known [14,27]. Various studies [32,33] and research projects have been/are conducted in which the impact of pH decrease caused by CO<sub>2</sub> injection in the ocean on marine ecosystems (plankton) were/are studied. However, there is a large difference between injection of relatively large quantities CO<sub>2</sub> in the ocean and small leaks of CO<sub>2</sub> from offshore reservoirs to the seafloor.

### 3.1.3 GLOBAL EFFECTS OF CO<sub>2</sub> LEAKAGE

From a global perspective, leakage of CO<sub>2</sub> from reservoirs would make CO<sub>2</sub> storage less effective as mitigation option. The crucial question is what leakage rates are acceptable to assure stabilisation of atmospheric greenhouse concentrations in the coming century is not endangered. Obviously, the acceptable leakage rate depends on stabilisation targets and the extent and timing of CO<sub>2</sub> storage.

Let us assume 1000 GtC will be stored between now and 2300. In order to stabilise greenhouse gas emissions at a level of 450-750 ppm, annual anthropogenic greenhouse gas emissions must be reduced to circa 2 to 4 GtC per year in 2300, with a faster cut in emissions for the more stringent stabilisation concentrations [34]. If we assume only 1 to 10% of an allowable emission of 3 GtC per year on average may be caused by leakage from underground reservoirs, the maximum long-term leakage rate would be circa 0.003-0.03%/yr.

Various studies have been performed in which acceptable leakage rates have been assessed by means of modelling. Hawkins [35] calculated the emission caused by leakage in a scenario in which the CO<sub>2</sub> reduction required to reach a stabilization level of 450 ppm from the IPCC IS92a reference case emissions to 2100 is completely covered by underground CO<sub>2</sub> storage (total storage of circa 800 GtC). The results show that with a 99.9% storage retention (0.1% annual leakage rate), emissions from leaks rise to the total allowable emission rate by 2200. According to Hepple and Benson [36], leakage rates must be less than 0.01% per year for stabilisation targets of 350, 450 and 550 ppm CO<sub>2</sub>, and be less than 0.1% per year to meet stabilisation targets of 650 and 750 ppm. The average total amount of carbon stored to 2300 ranged from 930 to 2490 GtC, depending on the stabilisation target. Lindeberg [20] used a more realistic model to calculate required average residence time of CO<sub>2</sub> in geological reservoirs, in which geological and physical features are accounted for. According to their calculations, an average residence time of at least 10,000 years is required, corresponding to an average

annual leakage rate of 0.01%. Although there is a certain range in the acceptable leakage rate (caused mainly by different assumptions on reference scenario and extent of CO<sub>2</sub> storage), most authors seem to agree that the mean annual leakage rate should not exceed 0.1%.

### 3.2 CH<sub>4</sub> LEAKAGE

The injection of CO<sub>2</sub> in depleted hydrocarbon reservoirs, coal beds and deep saline aquifers might result in leakage of methane and light alkanes, which is ubiquitous in the former two reservoirs and moderately common in deep saline aquifers [37]. An important feature of CH<sub>4</sub> is that it is more mobile than supercritical CO<sub>2</sub>.

Like CO<sub>2</sub> leakage, CH<sub>4</sub> leakage may have both local and global impacts. On a local scale, CH<sub>4</sub> may affect shallow water quality and poses a lethal threat when accumulating in confined spaces such as basements. Since the global warming potential (GWP) of methane is circa 23 times that of CO<sub>2</sub> [38], CH<sub>4</sub> leakage is an important factor to be assessed in order to verify the effectiveness as greenhouse gas mitigation option.

### 3.3 INDUCED SEISMICITY

The injection of large amounts of fluid into a reservoir increases the pore pressure and thereby modifies its mechanical state (existing underground stress fields). This might induce fracturing or activate faults, such that micro-seismicity and even damaging earth tremors might occur [9]. Potential effects of reservoir-induced seismicity (RIS) are damage to the cap rock and wells, which might cause CO<sub>2</sub> leakage, and damage to buildings and infrastructure.

Oil and gas reservoirs may be sources of reservoir-induced seismicity, generally when fluids are extracted, causing pressure changes in the reservoir [9]. Reservoir-induced seismicity has also been observed in liquid waste injection and underground gas storage (UGS) in geological formations [14,25], as will be described in Sections 5.3 and 5.5.

Generally, there are few cases of seismic activity in industrial analogues reported in literature and large quantities of brine, liquid waste, natural gas and CO<sub>2</sub> have been injected so far, which suggests that the probability of seismicity is low. Nevertheless, seismicity must be carefully monitored.

The risk of seismicity caused by CO<sub>2</sub> storage can be minimised by controlling the injection pressure. In situ stresses and pore fluid pressures need to be determined in order to assess the maximum injection pressure and identify faults that have high potential for reactivation [39]. The problem of seismicity might be more serious when CO<sub>2</sub> is injected into a reservoir in tectonically active regions (with high density of active faults), which can be found in e.g. Japan and California [40].

### 3.4 GROUND MOVEMENT

It is possible that the earth's surface will sink or rise because of man-made pressure changes, which might cause damage to buildings and infrastructure and might also trigger seismicity. Several cases of subsidence in history (mainly during exploitation of oil and gas fields) are known and well documented (e.g. Groningen gas field). In general, the mechanism of subsidence is well understood, but prediction is considered to be difficult [9].

It is not envisaged that uplift will take place in a CO<sub>2</sub> reservoir as long as the maximum storage pressure is kept below the geostatic pressure. However, in a reservoir that is under high tectonic stresses, any significant reduction of the grain pressure (pressure acting between individual rock particles) may trigger faults. This may lead to uplifting or down-faulting of the surface [9]. Subsidence can also be caused by a chemical reaction between CO<sub>2</sub> dissolved in brine and the reservoir rock, which may cause dissolution of the reservoir rock (chemical compaction). Consequently, the reservoir may cave in under the weight of the overburden formation. Chemical compaction or dissolution of the reservoir rock will particularly be a matter of concern in carbonate rocks with a high porosity [9].

### 3.5 DISPLACEMENT OF BRINE

The injection of CO<sub>2</sub> in aquifers might cause displacement of saline groundwater (brine). This may cause undesirable effects such as a rise of the water table (which could have negative impact on land quality and use) and an increase in salinity of sweet water reservoirs used for drinking water extraction and irrigation. The fate of brine displaced by the injected CO<sub>2</sub> and the risks it entails remains uncertain [14].

## 4 RISK ASSESSMENT STUDIES

A risk assessment has been performed for a conceptual CO<sub>2</sub> surface and injection system, using industry records on frequencies of leakage occurrences. Results indicated leakage rates below 0.03% of the annual CO<sub>2</sub> storage rate. It was concluded that multiple fatality risks are very unlikely. The risk of fatality for individuals may exceed typical risk criteria for industrial facilities for some modules, but can fall within acceptable limits with additional (technical) measures [27].

Risk assessment case studies have been performed for two oil fields: the Forties field in the North Sea and the Weyburn field in Canada<sup>7</sup>. At the Forties field, the cap rock is not faulted and there is limited fluid flow in the field, so the risk of leakage through the cap rock and via the underlying aquifer is considered to be negligible. However, the long-term impact of CO<sub>2</sub> on the cap rock integrity and leakage through or along wells have not been assessed in detail [27].

Both deterministic and probabilistic modelling has been performed to assess the long-term migration of CO<sub>2</sub> in the Weyburn oil field. The

7. The Forties oil field is considered for CO<sub>2</sub>-EOR. In the Weyburn project, circa 5000 t CO<sub>2</sub>/day is injected into the Weyburn oil fields in Canada in order to boost oil production and store CO<sub>2</sub> since October 2000.

deterministic approach indicates that the total amount of CO<sub>2</sub> removed from the EOR area 5000 years after the end of injection is 26.8% of the initial CO<sub>2</sub> in place at the end of EOR. The majority (18.2%) moves into the geosphere below, 8.6% migrates laterally outside the EOR area and only 0.02% diffuses through the cap rock. The maximum cumulative leakage through/along abandoned wells (circa 1000) corresponds to 0.14% of the initial CO<sub>2</sub> in place at the end of the EOR operation. However, better insight is needed in long-term degradation characteristics of cement and casings [41].

The probabilistic performance assessment has not been completely developed, but results of benchmarking with a reservoir simulation are consistent. The analysis indicates an average cumulative CO<sub>2</sub> release to the biosphere of 0.2% of the initial CO<sub>2</sub> in place. The analysis shows there is a 95% probability that 98.7 to 99.5% of the initial CO<sub>2</sub> in place will remain stored for 5000 years [41].

The models use a base scenario describing the expected future system development and leakage pathways, in which the cap rock integrity is not impaired. Geomechanical performance assessments show that the cap rock integrity has been maintained during historical injection/production and will be maintained given current CO<sub>2</sub> injection pressures. This assessment also indicates that salt dissolution by CO<sub>2</sub> will probably have minimal influence on seal integrity [41].

Also a more generic risk assessment methodology has been developed, in which leakage from a 'typical' deep saline aquifer has been modelled to estimate leakage rates from wellhead and cap rock failure [26].

Results indicate that leakage through a failed cap rock poses the highest risk to all environmental media. The calculated flux from a continuous fracture corresponds to a leakage rate of 0.1% of the total volume stored per year. Leakage rates through permeable zones in the cap rock are estimated at 0.05% of the total volume scored per year. Spatial frequency of cap rock failures within the area of review was estimated at 0.01 for both a fractured cap rock and high-permeability zones, assuming that 1% of the cap rock area spread over an area of review of 50 km radius is fractured and another 1% is highly permeable.

Although the estimated frequency of  $2 \cdot 10^{-5}$  for a major wellhead failure, based on statistics of UGS accidents in the USA and Canada, is much lower, the consequences (CO<sub>2</sub> flux) of such event are larger [26].

Obviously, cap rock failure is strongly dependent upon the site-specific geological characteristics and should be evaluated based on a geological assessment. Also failure rates of wells exposed to CO<sub>2</sub> are expected to be different than the statistical average from wells applied in UGS, as CO<sub>2</sub> is corrosive and reactive. Additionally, the frequency of well failures might be much higher in other regions in the world.

## 5 INDUSTRIAL AND NATURAL ANALOGUES FOR UNDERGROUND CO<sub>2</sub> STORAGE

Industrial analogues for underground CO<sub>2</sub> storage can be found in enhanced oil recovery with CO<sub>2</sub>, acid gas injection, disposal of industrial and nuclear waste in underground reservoirs and underground storage of natural gas. At least some of these analogues are common practices in several countries, for which extensive risk assessments have been performed. Natural analogues include reservoirs where CO<sub>2</sub> has been successfully trapped for geological timescales and reservoirs where CO<sub>2</sub> is migrating to the surface.

Underground CO<sub>2</sub> storage differs from industrial and natural analogues in various aspects (in compound stored, quantities stored and timeframe considered for storage). Nevertheless, there are strong similarities, which make analogues valuable to get insights that might increase our understanding into the risks of underground CO<sub>2</sub> storage. Industrial analogues might also provide useful insights into risk assessment and management (i.e. monitoring) and mitigation strategies for geologic storage of CO<sub>2</sub> [14].

### 5.1 CO<sub>2</sub> ENHANCED OIL RECOVERY

CO<sub>2</sub>-EOR is applied on a commercial scale in various countries to enhance the lifetime of depleted oil fields [7,42]. In the USA, circa 70 projects are in operation, injecting over 100,000 t/day [43]. CO<sub>2</sub> injection (often alternated with water injection) can achieve enhanced oil recovery by mobilising the oil through miscible or immiscible displacement. At the production well, oil, water, CO<sub>2</sub> and natural gas are produced and separated, after which CO<sub>2</sub> is recycled to the injection well. Only a part of the injected CO<sub>2</sub> is stored by dissolution in immobile oil. Although the purpose of CO<sub>2</sub>-EOR is primarily oil production and not CO<sub>2</sub> storage, CO<sub>2</sub>-EOR practices could enable us to study the behaviour of CO<sub>2</sub> in the reservoir and the risks of leakage. Monitoring CO<sub>2</sub> in the reservoir might increase our insight into the storage of CO<sub>2</sub> in immobile oil and leakage through abandoned wells and via fractures and faults extending into the cap rock. Unfortunately, CO<sub>2</sub> storage characteristics in the EOR industry have not been well documented [7,42], the Weyburn project being the first with a monitoring programme.

From EOR operative experience in the USA, it has been concluded that seals are maintaining their integrity and retaining CO<sub>2</sub> in place. No significant leakages have occurred during CO<sub>2</sub> injection period, although several operators mentioned that CO<sub>2</sub> migrated through fractures or via flanks of the structure to zones that are in communication with the injection zone [43]. Soil gas measurements at the Rangely Weber oil field, where CO<sub>2</sub> is injected to enhance oil recovery, have been performed driven by concerns of leakage as a consequence of reservoir over pressuring. These measurements indicate annual fluxes of maximally 3800 t CO<sub>2</sub> (0.13 g CO<sub>2</sub>/day/m<sup>2</sup>) and 400 t of thermogenic CH<sub>4</sub> (=25,300 t CO<sub>2</sub> equivalents) originating from deep sources over an area of 78 km<sup>2</sup> [37]. The CO<sub>2</sub> flux corresponds to

approximately 0.1% of the annual injection and 0.02% of the cumulative CO<sub>2</sub> storage<sup>8</sup>. These observations are reason to more carefully assess the impact of reservoir over pressuring as a consequence of CO<sub>2</sub> injection.

## 5.2 ACID GAS INJECTION

Oil and natural gas generally contain varying amounts of hydrogen sulphide (H<sub>2</sub>S), a toxic gas, and CO<sub>2</sub>, acidic components that have to be removed before the product is sent to the market. After the acid gases have been removed by absorption, H<sub>2</sub>S can be converted into elemental sulphur and CO<sub>2</sub> vented to the atmosphere. Alternatively, H<sub>2</sub>S can be flared (causing SO<sub>2</sub> emission) or the gases (containing between 15 and 98% CO<sub>2</sub>) can be re-injected into a geological formation. In western Canada, increasingly more oil and gas producers are turning to acid gas re-injection. Although the purpose of acid gas injection is to dispose of H<sub>2</sub>S, significant quantities of CO<sub>2</sub> are injected simultaneously, because it is not economic to separate the gases [44].

There are currently 39 acid gas injection projects operating in Canada. In the period 1989-2002, close to 1.5 Mt CO<sub>2</sub> and 1 Mt H<sub>2</sub>S have been successfully injected into depleted hydrocarbon reservoirs and saline aquifers. In the USA, another 16 acid gas injection operations exist [44]. The young history indicates that acid gas injection is mature from an engineering point of view, but the fate of acid gas in the reservoirs has not been monitored yet, which is crucial to get insight into subsurface behaviour and risk of leakage. The acid gas injection operations provide a unique, commercial scale analogue for CO<sub>2</sub> geological storage, since CO<sub>2</sub> is injected in similar formations and conditions as considered for underground CO<sub>2</sub> storage, also with the purpose of permanent storage (in contrast to CO<sub>2</sub>-EOR). Monitoring injected acid gas might increase the insight on long-term containment of CO<sub>2</sub> and leakage by cap rock and well failures. In addition, information on reservoir characteristics of acid gas injection operations can be used to screen and identify sites for underground CO<sub>2</sub> storage.

## 5.3 UNDERGROUND DISPOSAL OF INDUSTRIAL WASTE

The technology of deep well injection of hazardous industrial liquid wastes has many similarities to the technology of CO<sub>2</sub> storage in deep saline aquifers. Many of the formations currently used for deep well disposal of industrial waste are also suitable candidates for CO<sub>2</sub> storage [14]. The risks involved in underground disposal of industrial waste also play a role in underground CO<sub>2</sub> storage. Examples of seismicity have been observed at injection sites of industrial waste [9]. The re-injection of liquid waste in the Rocky Mountain Arsenal (USA) well caused several earthquakes ranging between 0.5 and 5.3 on the Richter scale. Also a blowout of liquid waste has been reported [14]. Early performance of underground disposal of industrial waste in the USA (before the introduction of more stringent regulations) showed many examples of well failures and contamination of drinking water aquifers.

8. Injection started in 1986. Since then, 57 Mt has been injected and 22.3 Mt CO<sub>2</sub> has been stored. Annual injection rate in 1998 was circa 3 Mt, which increased slightly in the period 1998-2003 [7,42].

Failures were attributed to poor characterisation of the confining units, improper well completion techniques, use of well construction materials that were incompatible with the waste streams leading to corrosion, inconsistent or inadequate monitoring, and leakage through abandoned wells [14]. Recently, the Chemical Manufacturers Association undertook a probabilistic risk assessment of component failure of a hazardous waste well system and showed that failure of any of the system components under current regulations was in most cases much less than  $10^{-6}$  per year [14]. However, the chemical and physical features of  $\text{CO}_2$  are different from industrial waste such as industrial organic waste or brine water co-produced with oil/gas production. The density of  $\text{CO}_2$  is lower than that of liquid waste and therefore, buoyancy forces will tend to drive  $\text{CO}_2$  upward, whereas injected fluid wastes tend to migrate away from the injection well with little buoyant force driving it up or down [14]. The effects caused by leakage are comparable neither. This makes the use of experience on underground disposal of industrial waste for underground  $\text{CO}_2$  storage rather limited.

#### 5.4 UNDERGROUND DISPOSAL OF NUCLEAR WASTE

Like  $\text{CO}_2$  storage, safe nuclear waste disposal requires understanding the complex, coupled physical-chemical-mechanical processes that will occur over periods of hundreds to thousands of years [14]. Underground disposal of nuclear waste differs in many aspects (physical/chemical features, effects, disposal method and media) from geological  $\text{CO}_2$  storage. The lessons to be learned from underground disposal of nuclear waste should be found in the area of risk assessment methodology, monitoring, and public outreach (specifically what went wrong in this process).

The FEP methodology developed in the nuclear waste area might be a suitable framework to identify and evaluate the (long-term) risks associated with underground  $\text{CO}_2$  storage [14]. The FEP framework is a procedure to identify, classify and screen all relevant Features, Events, and Processes that may cause or affect risks. Features refer to geologic features, such as stratigraphic layering and faults or fracture zones. Events refer to occurrences such as changes in precipitation fluxes, seismic activities, and mining enterprises. Processes refer to physical/chemical and other processes active at the site such as buoyancy flow of variable-density fluids and chemical sorption. By combining critical FEPS (selected by expert opinion on basis of probability and consequence), scenarios are constructed and selected for performance assessment (by modelling). These scenarios describe possible future evolutions or states of the storage facility [45]. Within the IEA GHG Weyburn  $\text{CO}_2$  monitoring and storage project and the CCP project, this method is adapted for the purposes of  $\text{CO}_2$  storage [45].

## 5.5 UNDERGROUND STORAGE OF NATURAL GAS

Underground natural gas storage in depleted oil and gas fields and in aquifers is applied to help meet cyclic seasonal and/or daily demands for gas. Generally, UGS has been applied safely and efficiently, although there have been a number of documented cases where leakage has occurred [14]. Nine reservoirs of the circa 600 storage reservoirs operated in the United States, Canada and Europe have experienced leakage. Five cases were due to defective wells (casing corrosion and improperly plugged wells), three cases were due to cap rock failure and one case was due to inaccurate reservoir selection (too shallow) [46]. One of the main causes of leakage is that many UGS projects are operated at overpressure [14].

In order to reduce the risks of leakage in UGS projects, reservoirs should be characterised extensively, wells should be well constructed, monitored and maintained, over pressuring should be avoided and abandoned wells in the area should be located and plugged. In case of a leaking well, the well can generally be repaired or plugged. In case of leakage related to high storage pressure or a cap rock failure, the pressure in the reservoir needs to be reduced [14]. In all reported cases of leakage, remediation procedures were and continue to be effective [46].

There are also a limited number of cases of reservoir-induced seismicity caused by UGS; in Germiny (France), gas storage in an aquifer caused light earth shocks with a maximum of 1.5 on the Richter scale. However, investigations have shown the risk of earth tremors in case of gas storage in empty gas fields to be small, even at an over-pressure of 10% above initial reservoir pressure [25].

UGS experience is useful for underground CO<sub>2</sub> storage (i.e. increase our knowledge on leakage mechanisms and pathways) as UGS is in many ways analogous to CO<sub>2</sub> storage. Equal storage reservoirs and injection technologies are applied and natural gas behaves similarly to CO<sub>2</sub> (it is less dense than water and tends to rise to the top of the storage structure). However, CO<sub>2</sub> is denser and more viscous (and thus less mobile), reactive (in particular when dissolved in water) and not explosive nor flammable. Moreover, the duration of CO<sub>2</sub> storage is longer than it is for UGS and much larger volumes are involved. These differences deserve special attention when assessing risks of CO<sub>2</sub> storage using insights obtained from UGS.

## 5.6 NATURAL ANALOGUES

CO<sub>2</sub> reservoirs and CO<sub>2</sub> containing oil and gas fields<sup>9</sup> are natural analogues where CO<sub>2</sub> has been contained for geologic timescales. No catastrophic CO<sub>2</sub> releases from CO<sub>2</sub> and hydrocarbon reservoirs have been reported in literature, although all reservoirs are thought to leak over geologic time [14].

CO<sub>2</sub> containing reservoirs are ideal to assess long-term effects of underground CO<sub>2</sub> storage (e.g. on cap rock integrity). Within the GEODISC project, the Ladbroke Grove gas field in South Australia has

<sup>9</sup>. One in ten gas fields contain 1-5% CO<sub>2</sub> and one in hundred contain on average 50% CO<sub>2</sub>, some fields even exceeding 80% [47].

been studied. CO<sub>2</sub> originating from nearby volcanoes has migrated into the reservoir between 1 million and 4500 years ago. Mineralogical analysis has revealed that some of the CO<sub>2</sub> has been permanently stored by mineralisation due to the high amount of reactive minerals present in the reservoir, although the majority of CO<sub>2</sub> is stored in gaseous and aqueous phases [48].

At sites where CO<sub>2</sub> is actively leaking, leakage rates can be assessed by soil gas and flux measurements and also the impacts of leakage can be determined. At Mammoth Mountain in California, volcanic outgassing of CO<sub>2</sub> occurs through faults and fractures. The leakage rate varies between 25 and 7000 g CO<sub>2</sub>/day/m<sup>2</sup> [13]. Within NASCENT, various leaking and non-leaking CO<sub>2</sub> reservoirs have been studied to identify leaking conditions and consequences. At Mátraderecske, a village in Hungary, the average CO<sub>2</sub> flux is 240 to 480 g CO<sub>2</sub>/day/m<sup>2</sup>, with a maximum at 19,200 g CO<sub>2</sub>/day/m<sup>2</sup> along faults [49]. In Ciampino, Italy, fluxes of 700 g CO<sub>2</sub>/day/m<sup>2</sup> have been measured. Such high fluxes can result in lethal concentrations in basements if not properly ventilated. Also high soil gas concentrations might occur, resulting in tree-crop death. At some CO<sub>2</sub> producing fields, there is evidence of groundwater pollution due to increased Ca<sup>2+</sup>, Mg<sup>2+</sup>, HCO<sub>3</sub><sup>-</sup> concentration and total hardness [27]. These natural analogues indicate that CO<sub>2</sub> migration to the surface occurs predominantly through faults and fractures. However, studies of fractures within NASCENT have indicated that open fractures through which CO<sub>2</sub> leaked might have been sealed in time by formation of carbonates. The role of fractures in controlling leakage is not completely clear yet [27].

## 6 R&D TOPICS

The previous sections have revealed a number of gaps in knowledge with regard the risks of underground CO<sub>2</sub> storage that require further research efforts:

- Processes that control leakage through or along wells should be determined. Long-term degradation characteristics of cement and casings need to be studied to estimate CO<sub>2</sub> fluxes and failure rates of abandoned wells.
- Processes that control leakage through faults and fractures need to be determined. Sealing of fractures by mineral precipitation needs further attention.
- Geochemical impact of CO<sub>2</sub> injection on reservoir rock and cap rock integrity.
- Chemical reactions and physical processes that could occur as a result of CO<sub>2</sub> injection in coal seams.
- Effects of leakage on (marine) ecosystems.

Data and insight into processes that control leakage and field data are needed to assess realistic leakage rates (fluxes) in order to validate the

values calculated by analytical and numerical models. One might increase insight on processes controlling leakage by performing leakage experiments, in which CO<sub>2</sub> is injected in reservoirs with proven leakage pathways such as non-sealing faults.

Well failure and the impact of CO<sub>2</sub> on cement and casings are being studied within various projects (see Table 2). A well bore integrity model is being developed and refined and laboratory work is performed in which various cement types are exposed to CO<sub>2</sub> under different pressures [27].

A number of research activities are underway to study the role of faults in natural CO<sub>2</sub> reservoirs and the geochemical impact of CO<sub>2</sub> on cap rock performance [27]. A core sample from the cap rock above the Utsira formation in the North Sea has been taken for analysis and experiments to assess the impact of CO<sub>2</sub> [13]. Field measurements are necessary to determine the large-scale features of the cap rock that could not be tested on laboratory samples, such as continuity and the presence of faults or fractures [14].

Various R&D programmes are dedicated to study the behaviour and impact of CO<sub>2</sub> on coal seams.

Although there is some knowledge on the relation between CO<sub>2</sub> leakage and death of animals and trees, the impact of CO<sub>2</sub> on whole ecosystems, especially offshore, is not well understood. As many storage reservoirs are located offshore, it is important to study the impacts of CO<sub>2</sub> leakage on marine ecosystems (which might affect fish populations). Although no specific project is dedicated to the impact of CO<sub>2</sub> leaks on marine ecosystems, there are several research projects on ocean storage studying the impact of large-scale CO<sub>2</sub> on marine organisms, which might increase our insights in this area.

Monitoring will play an important role in studying these research topics. It can be used to get insight into a wide range of parameters such as CO<sub>2</sub> behaviour, reservoir and cap rock performance, migration pathways, solubility, geochemical interactions (among which mineral trapping), groundwater and soil quality, ecosystem impacts and micro-seismicity associated with CO<sub>2</sub> injection [50].

Seismic methods are highly developed and can cover a large area with high resolution. The migration of CO<sub>2</sub> injected into the Utsira aquifer under the North Sea is monitored by means of time-lapse seismic surveys. It appeared that the overall effect of the accumulated CO<sub>2</sub> on the seismic signal is significant, making time-lapse seismic surveying a highly suitable geophysical technique for monitoring CO<sub>2</sub> injection into a saline aquifer [51]. One of the shortcomings of geophysical techniques is the difficulty in quantifying the amount of CO<sub>2</sub> that is present and the rate of leakage that might occur. Myer et al. [52] studied the resolution of seismic monitoring and concluded that a plume of circa 20,000 t CO<sub>2</sub> at 2000 m depth may be detectable. Other work suggests that the detectable volume of CO<sub>2</sub> would be much smaller [50].

By combining geophysical measurements with other techniques, such as formation pressure measurements and reservoir simulation, it will be

possible to obtain more quantitative estimates of leakage rates [50]. This will require additional research efforts and field-testing. Various research programmes are running to optimise existing monitoring techniques. More research is also required on techniques for monitoring the integrity (cement plugs, corrosion of casing) of abandoned wells. Methods for monitoring these aspects are studied within the CCP project [45].

Finally, besides the required R&D to increase our insight into risks itself, also a common risk assessment methodology able to assess long-term effects of underground CO<sub>2</sub> storage should be further developed.

TABLE 2. Summary of major R&D projects on risks of underground CO<sub>2</sub> storage

Project	Funding source(s)	Systems	Country	Project aims (related to risks of underground CO <sub>2</sub> storage)	Completion
CO <sub>2</sub> capture project (CCP) (SMV team)	- Industry sources - European Commission - US DOE - Klimatek	CO <sub>2</sub> -EOR/ EGR/ECBM aquifers	USA and Europe	· Develop, evaluate and apply tools and methodologies for risk assessment, risk mitigation and risk remediation, long-term monitoring and verification of CO <sub>2</sub> movement in geological formations · Assess sealing capacity of tubulars & cement by experiments	2004
GEODISC	- The Australian Greenhouse Office - Industry sources	CO <sub>2</sub> -EOR Aquifers Natural analogues	Australia	· Increase understanding of CO <sub>2</sub> -water systems and interaction between fluids and reservoir rocks and cap rock by experiments · Monitor CO <sub>2</sub> injection via modelling of seismic characteristics · Assess and quantify risks associated with CO <sub>2</sub> injection · Develop enhanced understanding of CO <sub>2</sub> trapping through study of natural analogues	2003
RITE R&D projects for geological sequestration of CO <sub>2</sub>	Not available	Aquifers (on & offshore)	Japan	· Increase understanding of CO <sub>2</sub> behaviour and interactions in an aquifer by means of experiments, models and injection tests · Evaluate monitoring methods for the assessment of environmental impact and safety	2005

Project	Funding source(s)	Systems	Country	Project aims (related to risks of underground CO <sub>2</sub> storage)	Completion
NACS	- US DOE - Industry sources	Natural analogues	USA	Study CO <sub>2</sub> trapping to evaluate the safety and security of geological storage processes	2004/2005
NASCENT	- European Commission - Industry sources	Natural analogues	Europe	Address key issues associated with geological CO <sub>2</sub> storage that include long-term safety, stability of storage underground, and potential environmental effects of leakage: · Interaction between CO <sub>2</sub> -water and reservoir rocks and cap rock (geochemistry) · Geomechanical testing and gas migration studies in low permeability cap rocks · Identification of pathways through soil gas surveys for CO <sub>2</sub> and associated tracer gases · Perform geochemical analyses of carbonated waters to assess the effects of CO <sub>2</sub> on groundwater	2004
SACS/ CO <sub>2</sub> STORE	- European Commission - National authorities- Industry sources	Aquifer offshore	Norway	· Monitor the injected CO <sub>2</sub> using repeat seismic monitoring to study the behaviour of CO <sub>2</sub> in the reservoir · Validate geophysical modelling	SACS 2002 CO <sub>2</sub> STORE 2006
GEO-SEQ	- US DOE - Industry sources	CO <sub>2</sub> -EOR/ EGR/ECBM aquifers	USA	Optimise a set of monitoring technologies ready for full-scale field demonstration in oil, gas, brine, and coal bed formations	2004
Weyburn CO <sub>2</sub> Monitoring Project	Various governmental and industry sources	CO <sub>2</sub> -EOR	Canada	· Assess geochemical impacts on the formation's CO <sub>2</sub> storage integrity and capacity · Monitor the movement of various fluids within the reservoir · Characterise fluid and phase behaviour to establish the mechanisms that govern the distribution and displacement of the CO <sub>2</sub> -rich fluids · Develop better monitoring tools and techniques	2004

## 7 CONCLUSIONS AND RECOMMENDATIONS

The (gaps in) knowledge of risks associated with underground CO<sub>2</sub> storage and research areas that require more attention are summarised in Table 3. The risks associated with pipeline transport and surface and injection facilities of CO<sub>2</sub> are known and can be minimised by risk abatement technologies and safety measures.

Although industrial and natural analogues suggest that CO<sub>2</sub> can be stored safely in geological reservoirs for thousands to millions of years, various issues need to be studied in more detail to assess (long-term) risks of underground CO<sub>2</sub> storage. Generally, there is a lack of knowledge and data to quantify the processes controlling/causing risks, which can partly be explained by the fact that underground CO<sub>2</sub> storage is a relatively young area. Most of the (demonstration) projects are still in their early stages. Another complicating factor is that underground storage encompasses long-term effects, which are difficult to assess by means of CO<sub>2</sub> injection operations or laboratory experiments.

Leakage of CO<sub>2</sub> from the reservoir is the main R&D issue. Leakage through or along wells, faults and fractures is generally considered to be the most important leakage pathways. The potential for leakage will depend on trapping mechanism and well, cap rock and overburden integrity. The type of reservoir in which CO<sub>2</sub> is stored is another important factor for leakage. Hydrocarbon fields are generally well studied and considered to be safe reservoirs for CO<sub>2</sub> storage, since they have held oil, gas and often CO<sub>2</sub> for millions of years without catastrophic leakage. However, these reservoirs are generally punctured by a large number of abandoned wells, some in bad condition, offering a potential leakage pathway. Deep saline aquifers and unminable coal seams have not been studied that comprehensively. Especially aquifers need to be studied in more detail as these reservoirs represent an enormous CO<sub>2</sub> storage potential. The risk of leakage might be higher than for hydrocarbon fields, as the cap rock integrity has generally not been proven. Coal seams generally have held coal bed methane for million of years and, moreover, CO<sub>2</sub> is adsorbed more strongly than methane, so the risk of CO<sub>2</sub> leakage is expected to be low. One of the principal objectives in future R&D is to determine the processes that control leakage through/along wells, faults and fractures to assess (a range of) leakage rates for various geological reservoirs. Research topics include the quantification of degradation of cement and casing by CO<sub>2</sub> and the role of fault sealing by mineral precipitation. Although experiments and models suggest that the geochemical impact of CO<sub>2</sub> on the cap rock integrity is minimal or even positive, further work is required in this area. The leakage rate at its turn is required to quantify the effects on human beings, ecosystems and groundwater quality. The effects of elevated CO<sub>2</sub> concentrations on human beings, animals and even for some biota are understood, but the effects on (marine) ecosystems need further research.

TABLE 3. Overview of (gaps in) knowledge and R&D issues related to risks of underground CO<sub>2</sub> storage

Risk	Chance	Potential consequences/effects	Major R&D topics
<i>Pipeline, surface and injection facilities</i>			
Pipeline failure	Frequency minor incident in order of 10 <sup>-4</sup> per km per year	CO <sub>2</sub> escape (effects see 'CO <sub>2</sub> leakage')	(1)
Surface equipment failure	Can be estimated from industrial experience with CO <sub>2</sub> and other gases	CO <sub>2</sub> escape (effects see 'CO <sub>2</sub> leakage')	(1)
Well failure (during injection)	· Can be estimated from CO <sub>2</sub> -EOR, acid gas injection and UGS experience · Frequency well blowout offshore gas fields estimated at 10 <sup>-4</sup> per well per year	- CO <sub>2</sub> escape (effects see 'CO <sub>2</sub> leakage') - Well blowout might cause casualties among operators	(1)
<i>Underground storage</i>			
CO <sub>2</sub> and CH <sub>4</sub> leakage	· Frequency well failure (after injection) unknown · Frequency cap rock failure is unknown (site/reservoir specific) · Chance of CO <sub>2</sub> leakage is generally expected to be lowest for coal seams and highest for deep saline aquifers	· Health hazard to people and animals (understood) · (Marine) ecosystem impact (not understood) · Affect soil and groundwater quality (understood) · Make CO <sub>2</sub> storage ineffective as mitigation option	· Determine processes that control leakage through wells, faults and fractures to assess leakage rate · Impact of CO <sub>2</sub> on well (cement, casing) and cap rock integrity require more research · Effects on (marine) ecosystems need to be studied · Hydrocarbon reservoirs relatively well studied. Deep saline aquifers (which have not always proven cap rock) and coal seams (chemical and physical processes) require more research.
Seismicity	Unknown. The frequency can be reduced by controlling injection pressure	Damage to buildings and infrastructure · Cap rock damage, which might cause CO <sub>2</sub> leakage	· Impact induced seismicity on cap rock integrity
Ground movement	Unknown	· Damage to buildings and infrastructure · Seismicity	(1)
Displacement of brine	Unknown	· Rise water table · Increase salinity drinking water resources	(1)

(1) These risks are generally understood or considered to be minimal/controllable with risk abatement technologies and safety procedures. Therefore, research topics for these risks are not discussed in literature.

Various research programmes and projects on geological CO<sub>2</sub> storage exist, in which risks are being studied. Most of these programmes are still running. Research items include possible migration pathways of CO<sub>2</sub> and interaction between CO<sub>2</sub> and the reservoir, cap rock and well bore. Quite a variety of reservoirs are being studied: deep saline aquifers, hydrocarbon reservoirs and coal seams in the USA, Europe, Australia and Japan. There are several ongoing programmes studying industrial and natural analogues. Industrial analogues where CO<sub>2</sub> is injected (CO<sub>2</sub>-EOR and acid gas injection) offer the opportunity to study the behaviour of CO<sub>2</sub> in geological reservoirs. Natural analogues are useful in providing a better understanding of leakage through faults and fractures, long-term physical and chemical interactions between CO<sub>2</sub> and the reservoir/cap rocks and effects on groundwater and ecosystems. These R&D programmes cover the major research issues with the exception of impacts on marine ecosystems. However, it is expected that more work is to be done as understanding the processes controlling leakage requires many field and experimental data. Additionally, risks strongly depend on reservoir and other site-specific conditions (cap rock, stratigraphic layers overburden, onshore/offshore, presence of water resources, ecosystems), for which a large variety exists. The site-specific nature of risks requires that a variety of pilot and demonstration storage projects be carried out, monitored and assessed.

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## CHAPTER VII

### **PATHWAYS TOWARDS LARGE-SCALE IMPLEMENTATION OF CO<sub>2</sub> CAPTURE AND STORAGE: A CASE STUDY FOR THE NETHERLANDS \***

\*  
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#### ABSTRACT

In this chapter, we sketch four possible pathways how ccs (r)evolution may occur in the Netherlands, after which the implications in terms of CO<sub>2</sub> stored and avoided, costs and infrastructural requirements are quantified. ccs may play a significant role in decarbonising the Dutch energy and industrial sector, which currently emits nearly 100 mt CO<sub>2</sub>/yr. We found that 15 mt CO<sub>2</sub> could be avoided annually by 2020, provided some of the larger gas fields that are forecasted to become available the coming decade could be used for CO<sub>2</sub> storage. Halfway this century, the mitigation potential of ccs in the power sector, industry and transport fuel production is estimated at maximally 80-110 mt CO<sub>2</sub>/yr, of which 60-80 mt CO<sub>2</sub>/yr may be avoided at costs between 10 and 40 €/t CO<sub>2</sub>, including transport and storage. Avoiding 30-60 mt CO<sub>2</sub>/yr by means of ccs is considered realistic given the storage potential represented by Dutch gas fields, although it requires planning to assure that domestic storage capacity could be used for CO<sub>2</sub> storage. In an aggressive climate policy aiming for 50-80% CO<sub>2</sub> reduction by 2050 versus 1990, avoiding another 50 mt CO<sub>2</sub>/yr may be possible provided that nearly all capture opportunities that occur are taken. Storing such large amounts of CO<sub>2</sub> would only be possible if one of the mega structures, either the Groningen gas field, large reservoirs in the Bunter Sandstone formation or the Utsira formation in the North Sea, will become available.

## I INTRODUCTION

Carbon dioxide capture and storage (ccs) is expected to become a serious CO<sub>2</sub> emission reduction technology in the Netherlands. The annual Dutch CO<sub>2</sub> emission is nearly 180 mt CO<sub>2</sub> at present, of which approximately 100 mt CO<sub>2</sub>/yr emitted by the energy and manufacturing industry [1]. As we are on the threshold of its introduction, it is about time to come up with strategies for large-scale ccs deployment as part of the transition towards a more sustainable energy system. For this purpose, we need insights into the way ccs may evolve and what its role in reducing greenhouse gasses (GHG) could be. More specifically, the capture potential in time, and the extent and pace at which this could be deployed given available storage capacity, needs to be studied in more detail.

Recently, the contribution of ccs in reducing Dutch GHG emissions has been forecasted at 0-15 mt CO<sub>2</sub>/yr avoided in 2020, depending on the emission reduction target set [2]. Crucial in this analysis was the assumption on the maximal ccs contribution, which was loosely based on the expected storage capacity that could be operational in 2020. Although useful in comparing different GHG options to compose a technology portfolio, the analysis does not shed light on the road towards that 15 mt CO<sub>2</sub> emission reduction in 2020, nor does it consider the road ahead. As most scenario studies indicate that ccs deployment really takes off beyond 2020 if a climate policy is in place [3-5], the analysis should at least cover the period up to 2050 to understand the potential development of ccs.

Rather than studying the competition between ccs and alternative GHG mitigation options as performed in [2] and as investigated in more detail for the power sector in concurrent research [6], we will focus on the prerequisites, bottlenecks and consequences of different ccs pathways for the Netherlands. The former approach would answer the question how much ccs can be expected in a portfolio of GHG mitigation options. Our analysis tries to answer the question how different futures for ccs deployment may look like. The main objective of this study is to set up ccs pathways that may unfold and assess the implications in terms of CO<sub>2</sub> avoided and stored, costs and infrastructural requirements. We cover a potentially wide range of ccs implementation and timing, as many of the factors determining the ccs potential are uncertain. The pathways are created by combining information on CO<sub>2</sub> capture potential in different sectors and the capacity and availability of geological reservoirs. The temporal and spatial aspects of the energy system and geological reservoirs are explicitly addressed in this analysis. So far studies have focussed either on the dynamics in the power sector (e.g. [7,8]) or the spatial matching of existing sources and sinks [9,10]. The timeframes in which hydrocarbon reservoirs become available are generally not considered. Source-sink matching has been performed for current Dutch sources [11], but the developments and capital stock turnover in the energy sector and availability of sinks in time was not incorporated.

The Netherlands may become an electricity exporting country, considering its strategic location on the sea (coal logistics, cooling water) and good infrastructure (natural gas and electricity transmission lines). An additional reason that may become important is the ample storage options in the Netherlands and the North Sea, and the limited storage capacity in neighbouring countries such as Belgium, Germany and France [12].

Composing CCS pathways serves multiple purposes. First, they illustrate how much CO<sub>2</sub> could and may be avoided in time, at what costs, and for how long we could continue relying on this option. This is valuable information for decision makers who need to develop strategies to reduce GHG emissions. Second, analysing the requirements and bottlenecks of possible CCS futures could also make clear what short-term actions may be needed to realise long-term goals. Third, an integrated assessment of this kind may reveal synergies in the form of common infrastructure to transport CO<sub>2</sub> from various sources.

The analysis we present is techno-economic by nature, as it deals with power and industrial plants, infrastructure and reservoirs, with a focus on the medium to longer term. We do not explicitly investigate actions to set off CCS the coming years. Neither do we study institutional, legal and social barriers that may inhibit its deployment.

## 2 METHODOLOGY

In setting up different pathways, we consider the following factors as being the most decisive for the future of CCS. The CO<sub>2</sub> infrastructure is not considered as driving factor for the deployment of CCS, but as an outcome.

- *Climate policy scenario.* The CO<sub>2</sub> reduction aimed for as function of time is a crucial factor in the role of CCS, as was demonstrated in [2]. The pathways studied here differ in the reduction targets set for different timeframes (2020 and 2050).
- *Baseline scenario.* The CO<sub>2</sub> capture potential is determined by the development in energy and material demand and technological characteristics and dynamics in the energy and industrial sector (vintage structure, fuel mix etc.).
- *Capacity and availability of CO<sub>2</sub> storage reservoirs.* The geological capacity available for CO<sub>2</sub> storage is rather uncertain due to geotechnical issues and competition with other applications such as underground gas storage (UGS). The storage capacity and availability is therefore varied among different pathways.
- *CO<sub>2</sub> capture options and costs.* CO<sub>2</sub> capture can be realised in different sectors by retrofitting plants or by installing completely new units with integrated capture technology. Progress in capture and conversion technologies is accounted for by differentiating between state-of-the-art technologies and more advanced technologies that are expected to become available in the longer term.

We set emission reduction targets (climate policy scenarios) versus baseline scenarios and translate the potential role of CCS into pathways. Each pathway represents a so-called ‘wedge’ (as proposed in [13]) in filling the gap between baseline and climate policy scenario. This methodology is in many aspects similar to a scenario analysis performed for the UK [14]. The difference lies in the assessment of the future role of CCS; in the UK study a varying share of CCS is assumed, after which each scenario is qualitatively evaluated by means of a Multi-Criteria Analysis. In our analysis, the potential for CO<sub>2</sub> capture and storage are used to estimate the role of CCS, after which infrastructural requirements and costs of different pathways are computed using a spreadsheet model. We distinguish the steps described below in setting up CCS pathways (see also Figure 1). Steps 1-3 form the basis of the CCS pathways, whereas the actual synthesis is done in steps 4-6.

1. Choosing baseline scenarios and climate policy scenarios. Climate policy scenarios consist of emission reduction targets and trajectories towards these targets (gradual versus instantaneous). We select four combinations of baseline and climate policy scenarios, each representing a specific pathway.
2. Evaluating geological storage capacity that becomes available in time, which set the physical boundaries for CCS. On that basis, storage supply curves are created, expressing storage potential versus storage costs, accounting for the availability of different reservoir types in different timeframes.
3. Assessing CO<sub>2</sub> capture potential in different sectors, which is mainly determined by the developments in the baseline scenario (energy demand, capital stock turnover etc). CO<sub>2</sub> mitigation supply curves are composed for each decade showing the CO<sub>2</sub> capture potential versus costs in time.
4. Deriving CCS contributions (in terms of CO<sub>2</sub> avoided) by combining step 1, 2 and 3. In this assessment, three situations can be distinguished:
  - a. Storage limitation: The CCS wedge is limited by the geological storage capacity and/or availability.
  - b. Capture limitation: In a scenario where practically the entire technical CO<sub>2</sub> storage potential would be available, CCS penetration is limited by the opportunities for CO<sub>2</sub> capture in the different sectors.
  - c. No limitation: When both the capture and storage potential are larger than the required emission reduction, we assume a maximum CCS contribution. This value can either be based on other limitations (e.g. rate at which new infrastructure can be realised), or be a fixed value (a maximum of 50% of the gap) when there are practically no limits conceivable.
5. Matching sources and sinks given their spatial distribution and temporal availability. For this aim, we compose a supply curve, in which CO<sub>2</sub> transport and storage costs are calculated for each CO<sub>2</sub> source following the CO<sub>2</sub> mitigation supply curve from step 3, up to the CCS contribution assessed in step 4.
6. Composing CCS pathways. Step 4 and 5 are repeated for each decade,

accounting for the storage capacity being reduced by the extent at which CCS has been applied in previous time periods. In this iterative process, we incorporate future developments in CO<sub>2</sub> capture capacity with a perfect foresight of 25 years in deciding on storage capacity to be exploited and infrastructure to be constructed.

7. Calculating the implications. Finally, the amount of CO<sub>2</sub> stored and avoided in time, investment costs of the entire system and CO<sub>2</sub> mitigation costs are calculated for the various pathways. We use the present value method to calculate the economic performance of the CCS systems, as fuel prices, electricity production and hence CO<sub>2</sub> production vary in time and investments of the CCS system are incurred at different timeframes. For electricity and fuel production and use, we use Equations 1 and 2, as the reference system may be a different technology<sup>1</sup>. For industrial processes, where the reference system is simply the plant without capture, we use Equation 3.

1. An IGCC with CO<sub>2</sub> capture may replace a PC unit without capture. In the transport sector, hydrogen could replace gasoline, which involves different vehicle costs and tank-to-wheel efficiencies.

$$COE = \frac{\sum_{t=0}^T \frac{C_t}{(1+r)^t}}{\sum_{t=0}^T \frac{E_t}{(1+r)^t}} \tag{Equation 1}$$

$$MC = \frac{(COE / \eta_{end-use} + C_{end-use})_{CCS} - (COE / \eta_{end-use} + C_{end-use})_{ref}}{(m_{CO_2} / \eta_{end-use})_{ref} - (m_{CO_2} / \eta_{end-use})_{CCS}} \tag{Equation 2}$$

$$MC = \frac{\sum_{t=0}^T \frac{C_{t,CCS}}{(1+r)^t}}{\sum_{t=0}^T \frac{M_{t,avoided}}{(1+r)^t}} \tag{Equation 3}$$

- COE = production cost of energy carrier (€/GJ)
- C<sub>t</sub> = costs in year t (€)
- E<sub>t</sub> = energy production in year t (GJ)
- T = project lifetime (yr)
- r = discount rate
- MC = mitigation costs (€/t CO<sub>2</sub>)
- η<sub>end-use</sub> = end-use efficiency (functional unit/GJ)
- C<sub>end-use</sub> = end-use costs (€/functional unit)
- m = CO<sub>2</sub> emission factor (t/GJ) of CCS chain and reference chain
- M<sub>t,avoided</sub> = avoided CO<sub>2</sub> emission in year t (t CO<sub>2</sub>)

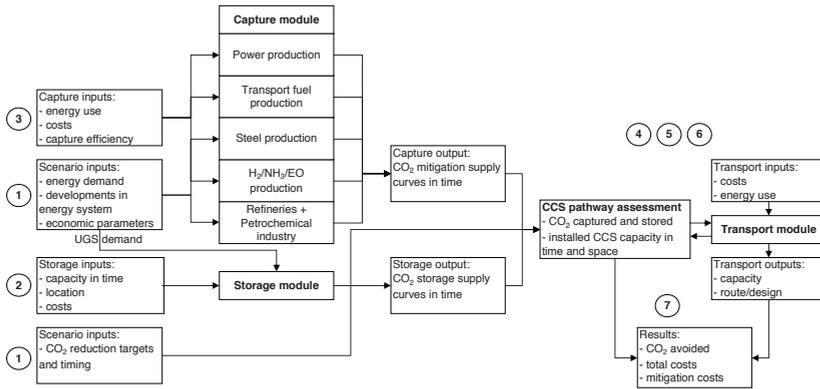


FIGURE I. Sequence of and relations between different modules in estimating the role of CCS

### 3 DEVELOPING CCS PATHWAYS

#### 3.1 BASELINE AND CLIMATE POLICY SCENARIOS

The four combinations of baseline and climate policy scenarios, each representing a specific pathway, are summarised and depicted in Table 1 and Figure 2. The emission reductions levels are based upon ambitions formulated by the European Union, which are between 15-30% by 2020 and as much as 50-80% by 2050 in comparison to the 1990 level [15,16]. Pathway 1 strives for an extreme cut in emission reduction starting right away. In the second pathway, CO<sub>2</sub> reduction is postponed to the period beyond 2030, after which we need to accelerate CO<sub>2</sub> emission reductions to achieve the targets set for 2050. In pathway 3, CO<sub>2</sub> emissions are to be stabilised at the 2010 level by means of national actions, assuming a large share of emission reduction is achieved by investments abroad. Pathway 4 is a more average and gradual, but still ambitious, scenario.

For the baselines, we adopt the Global Economy (GE) scenario and the Transatlantic Market (TM) scenarios. These scenarios have been composed by various Dutch planning agencies to study the impact of various trends in the Netherlands on the physical environment up to 2040 [17,18]. GE is characterized by global free trade and a strong orientation towards private responsibility, resulting in a relatively high economic growth of 2.6% annually up to 2040. The TM scenario has a somewhat lower economic growth (1.9% per year). EU member states focus on national interest (instead of international cooperation as in GE) and current trade blocks are maintained. The absence of trade relations with main gas-exporting countries results in high gas prices after 2020. In both scenarios, energy and environmental policies that are in place today, among which the EU emission trading system, are assumed to be continued up to 2020, after which they expire. The high economic growth in the GE scenario results in strong increase in energy demand (+55% in 2040 versus 2002), net import of electricity and a preference

for coal and natural gas-fired power generation. In the TM scenario, the energy demand in 2040 is 40% higher in comparison to 2002 and electricity exports exceed imports. Electricity supply is mainly based on coal and gas, and on nuclear energy in the long term [18].

TABLE I. Pathways for different CO<sub>2</sub> emission reduction scenarios. Values in parentheses are cumulative reductions

Pathway	Baseline reduction 2020	Emission reduction 2020	CO <sub>2</sub> reduction 2050	Emission reduction 2050	CO <sub>2</sub>
1. 'deep reduction' GE		30% reduction vs. 1990	95 Mt CO <sub>2</sub> /yr <sup>a</sup> (740 Mt)	80% reduction vs. 1990	300 Mt CO <sub>2</sub> /yr (6800 Mt)
2. 'postponed action'	GE	no action	0 Mt CO <sub>2</sub> /yr (0 Mt)	50% reduction vs. 1990	250 Mt CO <sub>2</sub> /yr (2600 Mt)
3. 'action abroad'	TM	stabilisation 2010	20 Mt CO <sub>2</sub> /yr (110 Mt)	stabilisation vs. 2010	65 Mt CO <sub>2</sub> /yr (1500 Mt)
4. 'ambitious'	TM	15% reduction vs. 1990	65 Mt CO <sub>2</sub> /yr (500 Mt)	50% reduction vs. 1990	165 Mt CO <sub>2</sub> /yr (4100 Mt)

<sup>a</sup> This would imply realisation of the complete technical reduction potential [2].

2. Bradshaw et al. [19] define three categories, being theoretical, realistic and viable capacity. The theoretical capacity assumes that the entire reservoir formation (pore volumes, water) is accessible to store CO<sub>2</sub>. The realistic capacity applies a range of technical cut-off limits such as quality of the reservoir and seal, and whether there may be other competing interests that could be compromised by injection of CO<sub>2</sub>. Finally, the viable capacity also considers economic, legal and regulatory barriers to CO<sub>2</sub> geological storage. The figures used in our report are somewhere in between the theoretical and realistic capacity and are hereby referred to as technical storage capacity.

3.2

CO<sub>2</sub> STORAGE

Table 2 shows the technical capacity<sup>2</sup> for CO<sub>2</sub> storage in the Netherlands and the continental shelf. Gas fields represent the major storage potential. The Dutch oil fields represent a relatively low storage potential and, therefore, not further considered. Aquifers and coal seams are not that well studied and characterised as hydrocarbon

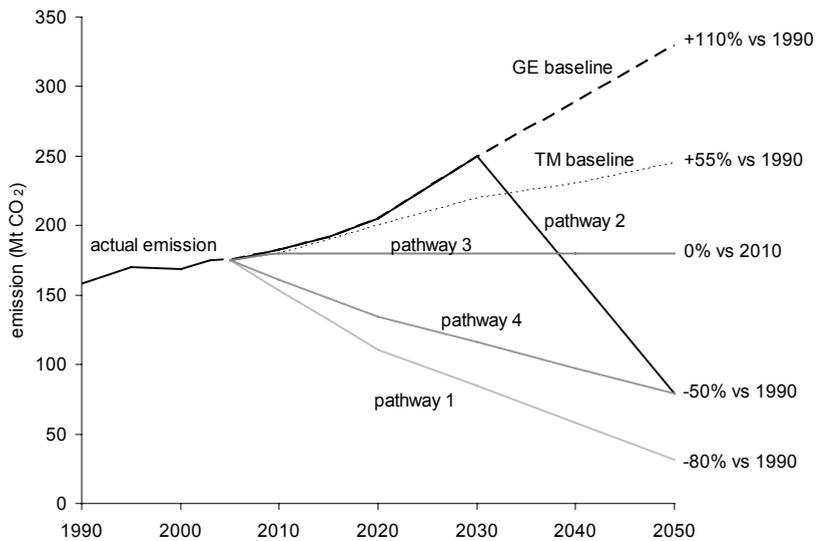


FIGURE 2. Emission reduction pathways investigated in this study versus baseline scenarios developed for the Netherlands in [18]

structures, which causes a relatively large uncertainty in the capacity figures. In Table 2 only aquifer traps (geological structures that physically retain buoyant fluids) are included. Possibly other parts of the water bearing layers may be used for CO<sub>2</sub> storage as well. The gross storage potential of the entire Dutch aquifer formations, assuming 2% storage efficiency, is estimated at roughly 10 and 6 Gt CO<sub>2</sub> for onshore and offshore formations, respectively [20,21]. These figures are highly uncertain, as there is little geological data available, and the storage potential depends on local pressure, temperature and aquifer salinity [22]. In addition, the seal quality for some formations is questionable and the permeability in some parts of the aquifer may be too low for sufficient injectivity [21].

TABLE 2. Technical CO<sub>2</sub> storage potential in the Netherlands

Reservoir	Storage capacity (Mt CO <sub>2</sub> )	Source
Gas fields <sup>a</sup>		
Groningen gas field	7350	[23]
Other gas fields onshore	1600	[23] + confidential data
Other gas fields offshore	1150	[23]
Oil fields <sup>b</sup>	40	[23]
Coal seams <sup>c</sup>	170 (40-600)	[24]
Aquifer trap prospects		
Onshore <sup>d</sup>	400	[23]
Offshore <sup>e</sup>	350 (90 – 1100)	[21]

<sup>a</sup> The figures in [23] refer to reserves with a storage capacity larger than 4 Mt CO<sub>2</sub>, assuming 100% of the volume recoverable hydrocarbons could be replaced with CO<sub>2</sub> up to hydrostatic pressure. Undiscovered fields are estimated to add another 50 bcm the coming 10 years. The expectation is that future discoveries, given the mature stage of exploration in the Netherlands, will generally be smaller structures on average [25]. As small fields are less suitable for CCS, we do not make a large error by excluding them. The uncertainty margin in the estimation of the onshore gas fields is ± 20%.

<sup>b</sup> Plans exist to take the Schoonebeek field, representing a storage capacity of 20 Mt CO<sub>2</sub>, back into production using horizontal wells and steam injection [26], which seem more attractive techniques than CO<sub>2</sub>-EOR. The other fields are generally too small for CO<sub>2</sub> storage [27].

<sup>c</sup> Represents coal seams above 1500 m depth. Between 1500 and 2000 m, there is another 260-2260 Mt storage capacity, but the exploitation of coal seams in this interval is currently technically and economically questionable [24].

<sup>d</sup> Total onshore capacity including traps with a storage capacity larger than 5 Mt CO<sub>2</sub> identified in Permian, Triassic, Late Jurassic/Early Cretaceous and Tertiary aquifers. The storage potential of aquifer traps with a minimal storage capacity of 10 Mt is estimated at nearly 300 Mt. The storage efficiency is a function of the trap type: 2% for closed traps, 6% for open traps and 4% for unknown traps.

<sup>e</sup> The range represents uncertainty in trap-density (1-10%) and storage efficiency (2-6%).

Some critical remarks should be made with respect to the values presented in Table 2. The technical storage capacity of hydrocarbon fields and coal seams is determined on the basis of ultimate gas/oil recovery (UR). The UR calculations in [23] were calculated indirectly using field area and cumulative production figures. A comparison with confidential UR figures for various regions showed that the total onshore capacity estimations were nearly identical, although field/region estimations may deviate significantly. In addition, the method to convert UR values to CO<sub>2</sub> storage potentials is not standardised yet. For

gas fields,  $UR$  is generally divided by the gas expansion factor and multiplied with the  $CO_2$  density, assuming all gas can be replaced by  $CO_2$ . This method may lead to an overestimation in storage capacity, as it does not account for the fact that the pore pressure is reduced during gas production. As a consequence, irreversible compaction and reduction of the matrix porosity may occur [27]. In water driven reservoirs, part of the pore space will be occupied by water that cannot be replaced completely by  $CO_2$ . There is, however, no or small water influx into many Dutch gas fields [28,29]. The storage potential is also affected by the maximum injection pressure. Most calculations assume pressure build-up in the reservoir is limited to the initial reservoir pressure. If the injection pressure can be raised to the leak off pressure, the storage capacity can increase to more than five fold the storage capacity at initial reservoir pressure for some reservoirs [30]. Breunese [25] on the other hand argues that  $CO_2$  may not be injected to the original reservoir pressure.

In addition, a variety of technical, legal, social and/or economic reasons may reduce the capacity available for  $CO_2$  storage. Apart from those reservoirs that may simply not be suited due to unfavourable characteristics (e.g. low permeability, doubtful seal quality, complex reservoirs such as the limestone formations in the eastern part of the Netherlands [31]), a number of factors that may inhibit the  $CO_2$  storage can be distinguished:

3. Production wells are generally designed for lifetime of oil or gas production. When using these wells for  $CO_2$  storage, possibly parts of the well have to be replaced in order to make these elements resistant to corrosion and high injection pressures. Platforms may be reused when they are large enough and the costs to maintain them are not prohibitive. In this analysis, reuse of wells and production platforms is not further considered.

4.  $CO_2$  injected into a producing field may mix with the gas and may cause breakthrough at the producing wells, thereby degenerating the natural gas resources. On the other hand, the recovery of gas may be enhanced, although this technique is not proven yet [25]

- *Size constraints.* Small reservoirs are not preferred from an economic point of view. Generally, the lower limit is set around a few up to 10 mt  $CO_2$  [9,10,21]. Reservoirs should preferably offer sufficient potential to store the captured  $CO_2$  of one plant over its operational lifetime. Most of the Dutch gas fields are rather small (below 5 bcm, equivalent to roughly 10 mt  $CO_2$ ). Approximately 30 gas fields have an estimated  $UR$  larger than 10 bcm [25,27]. Only 15 aquifer traps were identified with a storage capacity above 10 mt  $CO_2$  [23].
- *Timing.* Figure 3 shows that most gas fields become available gradually in the coming two decades. Possibly, the lifetime of gas fields may be extended a few years with rising gas prices. The Groningen gas field is not expected to become available before 2040, and possibly (far) beyond 2050, provided that significant investments are made [32]. The pattern at which gas fields become available dictates the ‘window of opportunity’ for  $CO_2$  storage. Ideally,  $CO_2$  injection into gas reservoirs starts immediately after the production of gas has ceased, in order to subdue changes in reservoir features, minimise water influx and allow for possible reuse of infrastructure (wells, pipelines, platforms<sup>3</sup>) and knowledge of the reservoir. Possibly,  $CO_2$  injection may even start in the tail of the gas production curve<sup>4</sup>. Reservoirs that were abandoned earlier may be taken into use again for  $CO_2$  storage, but at higher costs in comparison to immediate reuse. Aquifer traps are not characterised sufficiently yet. Therefore, it is assumed that these formations may be used for  $CO_2$  storage beyond

2010. Large-scale storage in coal seams is not expected to be feasible before 2020.

- *Alternative applications.* Reservoirs may be reserved for other functions such as underground natural gas storage, geothermal energy (aquifers) or possibly hydrogen storage in the longer term. In addition, plans may exist to construct new buildings or infrastructure on onshore fields. An extension of current UGS capacity can be expected due to declining and less flexible domestic gas supplies and increased reliance on inflexible gas import. With the decreasing pressure of the Groningen gas field, it will become more difficult to match supply and demand, which enhances the demand for UGS to cover demand fluctuations and provide seasonal flexibility [33]. Gas fields are the most cost-effective reservoirs for this purpose [34]. The Netherlands, with its abundance of gas fields, may possibly serve as a gas hub, providing storage facilities for surrounding countries with a lack of (suitable) gas reservoirs. If the Netherlands will develop itself as a gas hub, the role of the Groningen gas field as seasonal balancer must be taken over by additional UGS capacity. In such a scenario, we estimate an additional 100-150 bcm (total gas volume) may be required the next 20 years, based on storage requirements forecasted in [25,34]. This corresponds to 13-20% of the UR of all onshore gas fields minus the Groningen gas field. Fields of at least 10 bcm are preferable for both UGS (seasonal storage, not peak shaving facilities) and CCS, for which only a limited number (~30) are available [25].

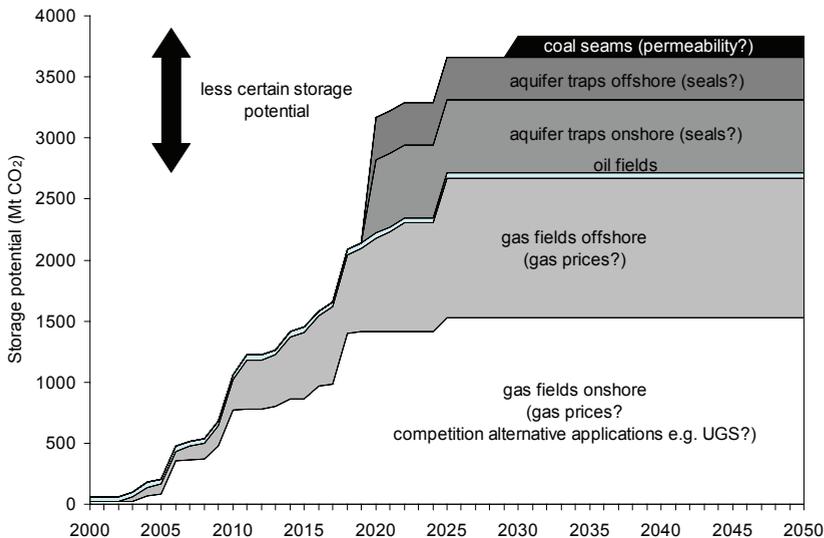


FIGURE 3. Storage availability in time (excluding current UGS reservoirs and Groningen gas field). The time line for the gas fields is derived from publicly available figures [23]. With less certain storage capacity, we refer to uncertainty in reservoir capacity, storage security and technical feasibility

Figure 4 illustrates the spatial mismatch between (current) CO<sub>2</sub> sources and sinks. The majority of the gas fields are located in the northern part of the country and the offshore continental shelf, whereas most large CO<sub>2</sub> sources are located in the western part of the country. The coals seams are predominantly located in the southern and eastern part of the country, whereas the aquifers are distributed more homogeneously. In the IJmond and Rijnmond area (region I and II), the joint geological capacity of aquifers and gas fields will be sufficient to store the lifetime CO<sub>2</sub> emissions of a single large coal-fired power plant. Given the presence of many sources and the interest to construct new power plants in the Rijnmond region, also the gas fields located offshore may have to be deployed. Most of the storage capacity represented by offshore gas fields is located in the KL quadrant. CO<sub>2</sub> produced in Zeeland (region III) could be transported to the Rijnmond area, piped offshore, or stored in the deep coal seams that can be found in this region, if this option would be feasible. Sources in Limburg (region IV) may have to rely on its coal seams or the aquifers located somewhat more north. Eemshaven, located in the Eemsmund region (VI), is being considered for the construction of new power plants. This region, together with regions V and VII, offers a large storage potential in gas fields.

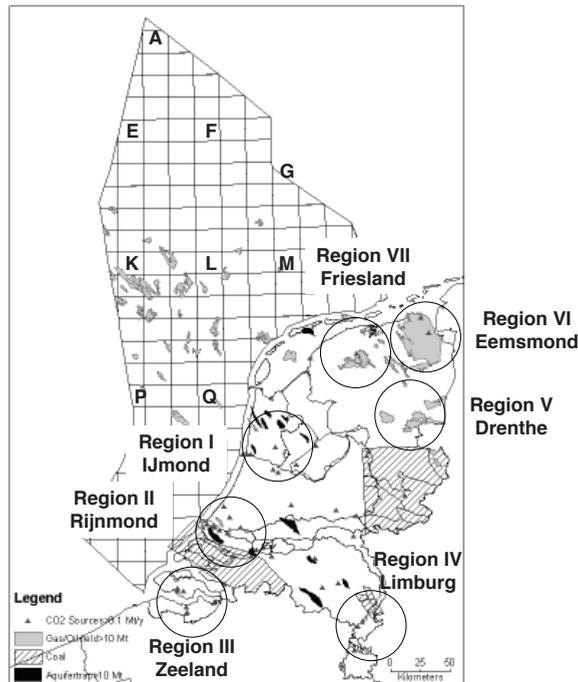


FIGURE 4. Location of geological reservoirs and large CO<sub>2</sub> sources in the Netherlands (derived from data of TNO-NITG). We defined 7 regions where CO<sub>2</sub> sources and/or CO<sub>2</sub> sinks are located. The Groningen field is the enormous structure in region VI. Note that this map is a snapshot; future CO<sub>2</sub> sources that will be equipped with CO<sub>2</sub> capture may be located in areas where few emissions are occurring now

Although the Dutch reservoirs offer a large technical storage potential, the limiting factors mentioned above may force us to look beyond national boundaries. There are a number of very large structures located in the North Sea, which may be preferred above the relatively smaller fields located in the Netherlands. The southern North Sea basin of the UK offers over 14 Gt CO<sub>2</sub> of storage capacity in closed structures in aquifers and nearly 3 Gt of CO<sub>2</sub> in gas fields [35]. The Leman field, a large gas field located at approximately 150 km from the Dutch shore in the UK part of the North Sea, offers a storage potential of nearly 1 Gt CO<sub>2</sub>. Given the production figures and the remaining reserves to be produced, the field will become available for storage around 2020 [9]. The Utsira formation in the Norwegian part of the North Sea, in which CO<sub>2</sub> from the Sleipner platform is injected, is of such dimensions that it could theoretically store 42 Gt CO<sub>2</sub>, of which 850 Mt in traps [36]. Another interesting opportunity is enhanced oil recovery using CO<sub>2</sub> (CO<sub>2</sub>-EOR) for nearly depleted oil fields in the North Sea. The UK sector offers a sequestration potential of 450-950 Mt CO<sub>2</sub> [37]. As many of the fields are approaching the end of production, there is a sharp decrease in potential after 2010 and another after 2016. It would require an enormous effort to have sufficient CO<sub>2</sub> capture and transport capacity online by 2010 in order to supply such large amounts of CO<sub>2</sub>. For the fields in the Norwegian sector, the window of opportunity is less critical; most CO<sub>2</sub> would be required between 2015 and 2025 [38]. In order to realise the EOR potential of 20 fields suitable for CO<sub>2</sub> injection, between 500 and 750 Mt CO<sub>2</sub> would be required, equivalent to 17-25 Mt/yr over a period of 30 years [38]. Holt et al. [39] studied a scenario in which 67 Mt CO<sub>2</sub> would be injected annually over a 40 year-period starting in 2010. Approximately half would be injected in Norwegian oil fields, representing a storage potential of 1100-1500 Mt CO<sub>2</sub>, and the rest would be stored in aquifers. Given the location of these reservoirs, sources in the UK and the few sources in Norway are most likely targets to provide CO<sub>2</sub>. Therefore, the North Sea EOR opportunities are not incorporated in this analysis.

In conclusion, the deployment of storage capacity depends on a number of uncertainties, which we captured in the form of scenarios:

- Availability of the Groningen gas field. Assuming this reservoir becomes available for CO<sub>2</sub> storage, it is the question when injection could be started. In a risk-averse strategy, injection is started after depletion. Depletion occurs most rapidly in the GE baseline, somewhere between 2040 and 2050 [18]. In the TM scenario, with lower energy consumption and a strong focus on energy security, depletion occurs somewhat later. However, its lifetime may be extended even longer by strategies imposing a cap on exploitation rates, or because improved techniques that increase recovery.
- Availability of the UK offshore gas fields. There are various power plants in the regions around the southern North Sea sector that may become future CO<sub>2</sub> suppliers. We consider a scenario in which part of

- these fields becomes available for ‘Dutch CO<sub>2</sub>’.
- A third, more remote reservoir is the Utsira formation in the Norwegian part of the North Sea, which might become a central storage hub for Northwest European CO<sub>2</sub>. This option would only make sense if a large-scale infrastructure is being constructed.
  - Competition with alternative applications, most importantly UGS. In the GE scenario, the Netherlands may develop itself as international gas hub, ‘confiscating’ many onshore gas fields for UGS.
  - In a scenario where emphasis is put on risk minimisation (risk averse strategy), storage is most likely to occur in (offshore) gas fields, for which storage security can be guaranteed with more confidence.
  - Profitability of enhanced hydrocarbon recovery. In a scenario with high energy prices and/or where energy security is one of the main priorities in energy policy, CO<sub>2</sub> storage with oil/gas revenues generated by EOR, EGR or ECBM may be the preferred option for storage. However, ECBM still requires significant development and must be considered as an uncertain storage option at this moment. EGR is not considered due to the uncertainty about its feasibility.

TABLE 3. Storage deployment in different pathways

Pathway	1. ‘deep reduction’	2. ‘postponed action’	3. ‘action abroad’	4. ‘ambitious’
Spatial constraints	Onshore + offshore NL, UK	Onshore + offshore NL, UK, NO	Offshore NL, UK	Onshore + offshore North Sea NL
2010-2020	Gas fields Aquifer traps	-	Offshore gas fields	Aquifer traps
2020-2030	Gas fields Aquifer (traps)	-	Offshore gas fields	Gas fields Aquifer (traps)
2030-2040	Gas fields Groningen field Aquifer (traps)	Aquifer (traps)	Offshore gas fields	Gas fields Coal seams Aquifer (traps)
2040-2050	Groningen field Aquifer (traps)	Aquifer (traps)	Offshore gas fields	Coal seams Aquifer (traps)

Table 3 gives an overview of the reservoirs we consider to be available for CO<sub>2</sub> storage in different pathways and periods. In pathway 1, basically all Dutch reservoirs are available, apart from some gas fields deployed for UGS. We assume CO<sub>2</sub> injection into the Groningen field could be initiated in the tail of production curve around 2030. In pathway 2, the window-of-opportunity for hydrocarbon fields has closed and/or reservoirs are being deployed for UGS, hence only aquifers are available. Energy security and risk minimisation are the key drivers in pathway 3. This will induce storage in offshore depleted hydrocarbon reservoirs, as gas fields not yet fully depleted are considered valuable assets (not to be ‘polluted’ with CO<sub>2</sub>) and the risks of leakage at onshore fields may be considered unacceptably high. In this scenario, storage in

aquifer traps is no option due to concerns about leakage. In pathway 4, focus is on national actions. We assume that the high gas prices cause the lifetime of gas fields to be extended by 5-10 years and induce CO<sub>2</sub> storage in the Dutch coal seams in a later stage.

3.3 CO<sub>2</sub> CAPTURE IN DIFFERENT SECTORS

Apart from the power sector, a number of industrial CO<sub>2</sub> sources and future transport fuel production are included in this analysis. Only sources emitting more than 0.1 Mt CO<sub>2</sub>/yr are accounted for. We do not include CO<sub>2</sub> capture from industrial CHP units, as the majority are decentralised sources emitting only a few kt/yr.

3.3.1 POWER SECTOR

In order to study the opportunities for CCS in the power sector, we first consider developments in electricity production as occur in the baseline scenarios. Several power plants have undergone or may undergo lifetime extension programmes and various utilities have announced plans for the construction of new power plants. The coming 5 years approximately 9 GWe of capacity may be commissioned, consisting mainly of large coal and gas-fired units. Up to 2020, the development of electricity supply in the GE and TM scenarios is nearly identical; coal, renewables and also decentralised CHP are the main grow markets. Beyond 2020, the baselines diverge. In GE mainly coal and gas-fired capacity is installed and also the share of decentralised CHP is increasing (see Figure 5). In TM, 6 GWe of nuclear capacity is installed in addition to coal-fired capacity. Electricity production by means of decentralised CHP is decreasing due to unfavourable electricity and gas prices. The market for renewable energy collapses after 2020 in both GE and TM, as climate policy is not continued and incentives for green electricity are abolished [18].

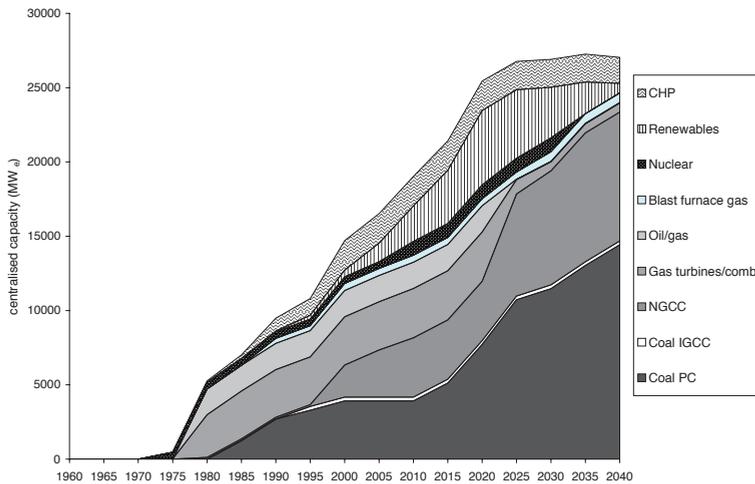


FIGURE 5. Electric generation capacity in the GE baseline scenario derived from [18] (excluding decentralised CHP units, which represent approximately 25% of the current installed capacity)

In studying CO<sub>2</sub> capture in the power sector, it may be useful to distinguish capture at existing plants, plants built in the short term (2007-2015) and plants that will be built in the longer term (beyond 2015).

Existing plants could be retrofitted with CO<sub>2</sub> capture technology. Retrofitting may become necessary when deep emission reductions are required, given the current trend for lifetime extension of existing assets, especially coal-fired units. There are several drawbacks and barriers that might inhibit retrofitting, among which the strong increase in electricity production costs and reduction in power output, which has to be compensated by additional capacity<sup>5</sup>. Moreover, at many power plant locations there may not be enough space for absorption and regeneration columns, compressors and eventually cooling towers, for example because DeNO<sub>x</sub> and FGD units were installed in a later stage. Also, the FGD unit may have to be upgraded to specifications of the solvent. In our study, it is assumed that retrofitting existing PC units is possible in pathway 1.

Plants that are built in the short term should be flexible in reducing their CO<sub>2</sub> output in order to anticipate uncertain climate policy. For coal-fired units that may be constructed the coming years, strategies are being considered to enable CO<sub>2</sub> emission reduction by either co-firing biomass and/or making the units 'capture ready'; i.e. designing the plants in a way that allows for cost-effective add-on of CO<sub>2</sub> capture later on. In practice, this means that additional space is reserved for CO<sub>2</sub> capture components, compression and cooling capacity. Also the vicinity of storage reservoirs may be considered in plant siting. For PC units, pre-investments could be made in the steam turbine to allow extraction for CO<sub>2</sub> regeneration and a high efficiency (or upgradable) FGD [40], if this is not required by environmental legislation. For IGCC units, the coal handling units, gasifier, ASU and downstream equipment could be oversized in order to prevent turbine derating when retrofitting in a later stage [41]. The turbine choice is of great importance. Also high pressure gasifiers may be selected and even a shift unit may be included in the base case design without CO<sub>2</sub> capture [42]. As the need for CO<sub>2</sub> capture is uncertain, and also the development in capture technologies cannot be foreseen, pre-investments are expected to be modest. Bohm et al. [43] concluded that the gains of retrofitting an IGCC with pre-investment are modest in comparison to retrofitting a standard IGCC. Fossil power plants that will be built in the longer term are assumed to be equipped with CO<sub>2</sub> capture, provided that more stringent climate policy has solidified, and sufficient certainty has been created to enable such large investments. Table 4 contains the main input parameters for the selection of technologies considered in this analysis.

5. We assume additional IGCC capacity with CO<sub>2</sub> capture is constructed to compensate for the loss in power output.

TABLE 4. Forecasts of electric efficiency and capital costs (€<sub>2000</sub>, year 0 in the model) of power plants in time

Technology <sup>a</sup>	2010–2020		2020–2030		2030–2050	
	$\eta_e$ (LHV)	TCR(€/kW <sub>e</sub> )	$\eta_e$ (LHV)	TCR(€/kW <sub>e</sub> )	$\eta_e$ (LHV)	TCR(€/kW <sub>e</sub> )
PC	46%	1200	49%	1100	52%	1050
IGCC	46%	1500	50%	1300	54%	1200
NGCC	58%	500	60%	450	63%	450
PC + CCS <sup>b</sup>	36%	1800	40%	1600	44%	1500
IGCC + CCS <sup>c</sup>	38%	1900	43%	1600	48%	1400
NGCC + CCS <sup>d</sup>	49%	850	52%	700	56%	650
PC capture ready retrofit <sup>e</sup>	36%	700	37%	700	38%	700
IGCC capture ready retrofit <sup>f</sup>	38%	500	39%	500	40%	500
PC retrofit 1 <sup>g</sup>	29%	900	30%	900	Na	Na
PC retrofit 2 <sup>h</sup>	32%	850	33%	850	Na	Na

<sup>a</sup> Figures are derived from [44-49]. All plants with CO<sub>2</sub> capture include compression to 110 bar. O&M costs are set at 4% of TCR for all configurations except for PC + CCS and PC retrofit at 5%. Lifetime of coal and gas-fired power plants is 45 and 30 years, respectively. Costs of retrofitting represent additional costs per kwe after retrofitting. Downtime due to retrofitting is not taken into account.

<sup>b</sup> Advanced supercritical PC + post-combustion capture (90% CO<sub>2</sub> capture efficiency).

<sup>c</sup> Dry-feed gasifier + pre-combustion capture (90% CO<sub>2</sub> capture efficiency).

<sup>d</sup> Brayton cycle + post-combustion capture (90% CO<sub>2</sub> capture efficiency).

<sup>e</sup> Advanced supercritical PC + post-combustion capture add-on (90% CO<sub>2</sub> capture efficiency).

<sup>f</sup> Dry feed gasifier + pre-combustion capture add-on (90% CO<sub>2</sub> capture efficiency).

<sup>g</sup> Subcritical PC + post-combustion capture add-on (90% capture efficiency), including refurbishment costs of 150 €/kwe to extend the plant lifetime by 20 years [46]. Repowering to advanced supercritical conditions reduces the energy penalty (and hence power output) and additional costs [46,49], but this option is not considered here.

<sup>h</sup> Supercritical PC + post-combustion capture add-on (90% CO<sub>2</sub> capture efficiency).

Ideally, an electricity market dispatch model should be applied to study the penetration of CCS in the power sector as such models can incorporate the effects and dynamics of CO<sub>2</sub>/fuel prices, sunk capital and plant dispatch [7]. Such an analysis is currently being performed for the Netherlands by van den Broek et al. [6]. Instead, we adopt a more heuristic approach to forecast the potential role of CCS in the power sector, making use of insights generated by such studies. We compose CO<sub>2</sub> mitigation supply curves by means of a simple plant-level analysis<sup>6</sup>, assuming the power plants with capture in the climate policy scenarios operate in the same load as the plants they replace in the baseline scenario. In the GE and TM scenario, PC plants are generally operating in base load and NGCC plants, with the exception of a few units, in intermediate and peak load. For the gas prices prevailing in the GE and TM baseline, an IGCC with CO<sub>2</sub> capture would be the most cost-effective mitigation option to replace PC units, representing the majority of new installed capacity in the baseline scenarios. NGCC with CCS may be deployed in intermediate load at sufficiently high CO<sub>2</sub> prices, which may occur in scenarios in which large emission reductions are required (such as pathway 1). As coal-fired plants are replaced for coal-fired plants with CCS and ditto for gas-fired plants, the ratio coal to gas does not change significantly over time. In reality, a switch to more gas-fired capacity

6. For each plant that will be constructed in the baseline, we calculate the mitigation costs to install a plant with CO<sub>2</sub> capture instead. Likewise, retrofit costs for existing units are computed.

without CO<sub>2</sub> capture may occur in some pathways. Figure 6 shows NGCC without CO<sub>2</sub> capture would be most economic for gas prices below 5.5-7 €/GJ and CO<sub>2</sub> prices up to 40-55 €/t CO<sub>2</sub>. Dispatch models indeed show that with rising CO<sub>2</sub> prices, NGCC will be dispatched more often and new capacity will be predominantly NGCC [5,7]. At a certain CO<sub>2</sub> price, coal-fired units with CCS become the preferable option for base load generation above NGCC. This would imply that the opportunities for CCS are postponed. Obviously, the construction of new NGCC capacity as a first reaction to rising CO<sub>2</sub> prices strongly depends on expectations on future gas prices. We ignore this temporary gas ‘revival’ and assume utilities invest in IGCC with CCS straight away. The possible early retirement of capital stock, which may occur under very stringent emission reduction pathways, is not accounted for.

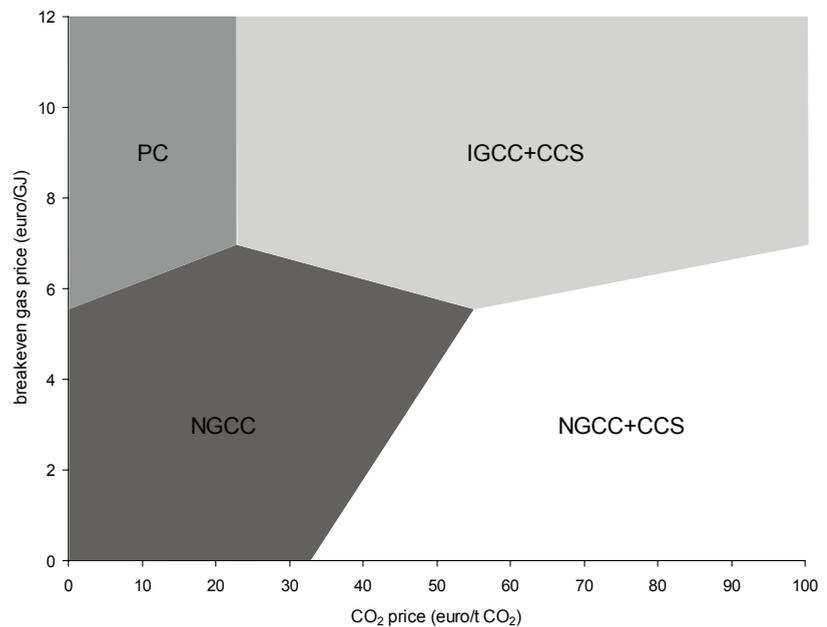


FIGURE 6. Competition between coal and gas-fired technologies as a function of gas and CO<sub>2</sub> price, assuming a coal price of 1.7 €/GJ. The breakeven gas price is defined as the gas price where the electricity production costs of two competing options are equal. Values include compression, 100 km transport and storage in an onshore gas field. We consider PC as the preferred technology in a world without climate policy, whereas IGCC with CCS is considered to be the preferred technology in a carbon constrained world

Now that we have insight into the capture potential in time and the basic dynamics that determine under which gas and CO<sub>2</sub> prices CCS technologies enter the market, the locations where new capacity can be installed are determined. Locations where old plants are being decommissioned will be used for new plants, as the construction of (especially coal-fired) power plants is to be preferred at existing sites due to legislation, social acceptance, logistics and available infrastructure.

Locations being considered for new large power plants are Eemshaven, the Rijnmond area and the Vlissingen area in Zeeland. Another interesting location might be on the shore of the IJmond region. We assume that all new capacity will be constructed in one of these locations.

### 3.3.2 INDUSTRIAL SOURCES

We consider industrial processes that generate relatively small quantities of pure CO<sub>2</sub>, the early opportunities, and processes generating large quantities of CO<sub>2</sub> concentrated at a single site as occur in the steel industry, petrochemical industry and refineries (see Table 5). CO<sub>2</sub> emissions from those industrial sources are set constant within the time period considered, assuming the market for these products remains stable or the growth is offset by energy efficiency measures.

TABLE 5. Overview industrial CO<sub>2</sub> sources, including capture costs and energy use [11,50-53]

Source	Region	CO <sub>2</sub> emission (Mt/yr) <sup>a</sup>	CO <sub>2</sub> purity	TCR (M€/kg CO <sub>2</sub> /s) <sup>b</sup>	Heat requirements (kJ/kg CO <sub>2</sub> )	Electricity requirements (kJ <sub>e</sub> /kg CO <sub>2</sub> ) <sup>b</sup>
Ammonia plant 1	IV	0.5	~100%	1.3	0	410
Ammonia plant 2	III	0.8	~100%	1	0	410
Hydrogen plant 1	II	0.6	~100%	1.2	0	410
Hydrogen plant 2	II	0.1	~100%	3	0	410
Ethylene oxide plant 1	II	0.13	~100%	2.6	0	410
Ethylene oxide plant 2	III	0.06	~100%	4	0	410
Gas processing plant 1	K	0.4	~100%	1.4	0	410
Ethylene plant 1	II	1.4	~12%	5	3000	470
Ethylene plant 2	III	2.7	~12%	3.1	3000	470
Ethylene plant 3	IV	1.7	~12%	4.2	3000	470
Steel plant 1	I	3.7 <sup>c</sup>	~20%	0.7	0	620
Refineries 1-4	II	6.6 <sup>d</sup>	7-13%	3.6-8.6	3200	480
Refinery 5	III	1.0	7-13%	6.7	3200	480

<sup>a</sup> Estimated CO<sub>2</sub> emission available for storage.

<sup>b</sup> Includes capture and compression to 110 bar.

<sup>c</sup> Only CO<sub>2</sub> produced in blast furnaces, i.e. the carbon input minus carbon incorporated in pig iron (~4%), is considered for capture. Based on the emission statistics for 2003 [1], we estimate approximately 9.2 Mt CO<sub>2</sub> was produced in blast furnaces and nearly 1 Mt in the basic oxygen furnace. A large share of BF gas is sold to the power sector, where it causes a CO<sub>2</sub> emission of 5.5 Mt. Note that BOF gas, consisting of approximately 55-80% CO and 10-18% CO<sub>2</sub> [54] may also be suited for CO<sub>2</sub> capture after shifting.

<sup>d</sup> Estimated emissions from boilers and heaters, derived from the Dutch energy balances [55]. CO<sub>2</sub> emissions are allocated to individual refineries on the basis of crude oil throughput.

We adopt the relations between costs and energy use of post-combustion CO<sub>2</sub> capture and CO<sub>2</sub> flow and concentration as applied in [56], for all sectors except iron and steel production. The high partial pressure of CO and CO<sub>2</sub> in blast furnace gas makes pre-combustion capture (after CO shift) more cost-effective than post-combustion capture. We use the costs and energy use quoted in [57].

### 3.3.3 PRODUCTION OF ALTERNATIVE FUELS

In the longer term, new opportunities for CCS may arise when a market

evolves for alternative fuels for the transport, residential and commercial sector. These sectors could be decarbonised by application of hydrogen or synfuels (Fischer-Tropsch diesel, methanol, DME) produced from coal (CTL), gas (GTL) and biomass (BTL) with CCS. Alternatively, cars may be driven electrically (using electricity produced with CCS), but this option is not considered here.

As the future of hydrogen is very much depending on the successful introduction of fuel cells, which is uncertain<sup>7</sup>, we incorporate a H<sub>2</sub> scenario in pathway 2 only. In the other pathways, we consider F-T diesel produced by gasification of coal and biomass with CCS as nearly climate-neutral alternative to clean hydrogen<sup>8</sup>. In contrast to hydrogen, the successful introduction of F-T diesel is not determined by the progress in immature technologies, as the fuel is compatible with existing vehicles and infrastructure. Its success will primarily be driven by oil prices, and the creation of large biomass markets and supply chains in case of BTL. F-T diesel produced from coal/biomass will become competitive with conventional diesel somewhere at 65-70 \$/bbl at zero carbon prices, and around 45 \$/bbl at carbon prices near 30 \$/t CO<sub>2</sub> [58,59]. In absence of scenario studies on F-T diesel penetration in time, we assume half the current diesel market could be replaced by synthetic diesel halfway this century.

In the H<sub>2</sub> variant, the demand is computed as a function of FCV penetration, for which we adopt the values proposed in the Hyways study [60]. In this scenario, 4 million FCVs are on the road by 2050, representing 30% of the entire fleet. In the first years, when the demand is still small, the general consensus is that H<sub>2</sub> will be produced predominantly by distributed SMR units and/or truck delivery of merchant H<sub>2</sub>. At a certain moment in time, the demand is sufficiently high to make a transition towards centralised hydrogen production with CCS. Combining the insights on transition dynamics published by Yang and Ogden [61] and the adopted hydrogen demand scenario [60] for the Dutch context suggests that the optimal transition year towards large-scale production may be around 2030. We assume reformers are installed to cover the growth in H<sub>2</sub> demand around 2030, after which coal gasification units are installed and operated in base load (see Table 6).

TABLE 6. Efficiencies and costs of transport fuel production and use [44,58,62]

Fuel	H <sub>2</sub>	H <sub>2</sub>	F-T diesel
Technology	SMR + CCS	CG + CCS	CBTL + CCS
Capacity (MW <sub>fuel</sub> )	1000	1000	1000
Efficiency (GJ <sub>fuel</sub> /GJ <sub>input</sub> )	73%	57%	55%
Electricity output (GJ <sub>e</sub> /GJ <sub>fuel</sub> )	0	0.04	0.09
TCR (€/kW <sub>fuel</sub> )	550	840	1113
O&M (% TCR/yr)	4	4	4
Car <sup>a</sup>		FCV	hybrid DICI
Fuel economy (MJ/km)		0.94	1.66
Costs (€/km)		0.32	0.28
Direct CO <sub>2</sub> emissions (g CO <sub>2</sub> /km)		0	100

<sup>a</sup> Reference system for H<sub>2</sub> and F-T diesel cars is a hybrid ICEV on gasoline and diesel, respectively, with fuel economy of 1.63 and 1.46 MJ/km and costs of 0.19 and 0.20 €/km.

7. In the energy futures sketched for the Netherlands up to 2040, the use of hydrogen is not accounted for due to large uncertainties in technological breakthrough of fuel cell technology [17].

8. F-T diesel production by coal gasification with CCS, but without biomass co-gasification, has well-to-wheel carbon emissions close to oil-derived diesel [58,59] and is therefore not considered.

3.3.4 CO<sub>2</sub> MITIGATION SUPPLY CURVES

Figures 7 and 8 show the mitigation potential for CO<sub>2</sub> capture in different sectors in time. Note that the curves for each timeframe include plants equipped with CO<sub>2</sub> capture in earlier timeframes that are still operative. In all pathways, the bulk of CO<sub>2</sub> emissions in the power and industrial sector could be captured at costs below 50 €/t CO<sub>2</sub> avoided. In 2050, roughly 60-80 Mt could be reduced at costs below 20 €/t CO<sub>2</sub> avoided, primarily from coal gasification systems to produce electricity or synthetic fuels, and from blast furnaces. In pathway 1, there is a substantially higher capture potential in comparison to pathway 3 and 4. This can be attributed to retrofit options and the fact that more opportunities arise for CO<sub>2</sub> capture at coal-fired and to a smaller extent gas-fired capacity. The costs to retrofit existing PC units vary between 35 and 100 €/t CO<sub>2</sub>, depending strongly on the age of the unit and the year of retrofitting. Pathway 2 is nearly identical to pathway 1 (and therefore not represented), with the exception that the retrofit option for existing plants has passed. In pathways 3 and 4, the installation of new nuclear capacity limits the application of CCS. As a consequence of nuclear capacity that becomes operative after 2030, several coal-fired plants are operated in intermediate load, driving mitigation costs up. However, the general trend is that costs to capture CO<sub>2</sub> decreases in time due to the application of improved technologies.

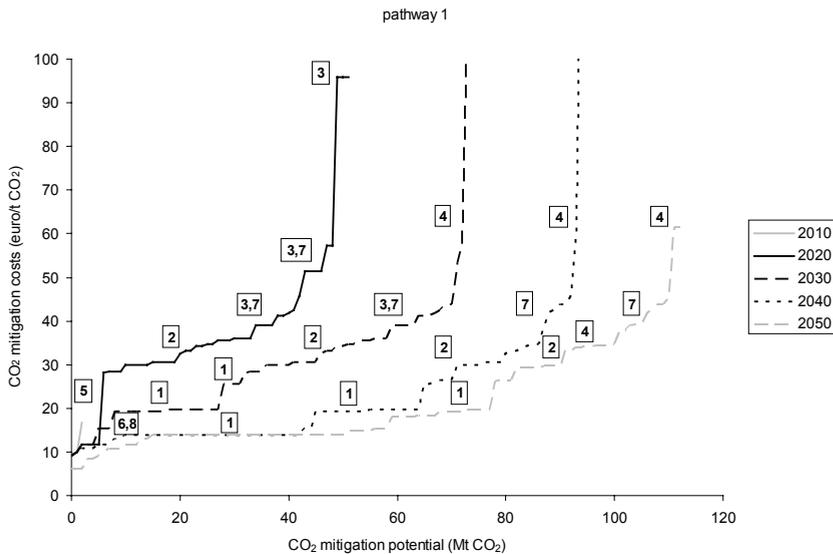


FIGURE 7. CO<sub>2</sub> mitigation supply curve for pathway 1 as function of time, excluding transport and storage costs

- 1 = IGCC + CCS
- 2 = PC + CCS
- 3 = PC retrofit
- 4 = NGCC + CCS
- 5 = H<sub>2</sub>/NH<sub>3</sub>/EO production
- 6 = steel production
- 7 = refineries/ethylene production
- 8 = FT diesel production and use

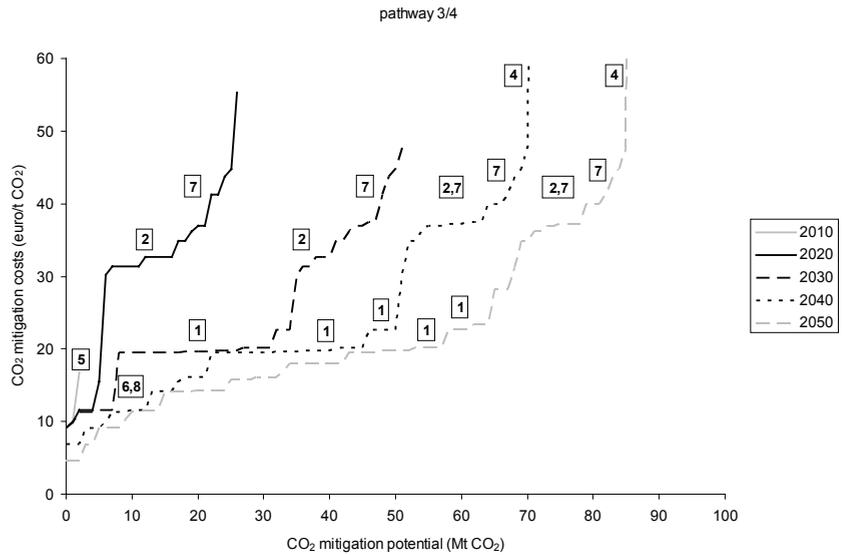


FIGURE 8. CO<sub>2</sub> mitigation supply curve for pathway 3 and 4 as function of time, excluding transport and storage costs

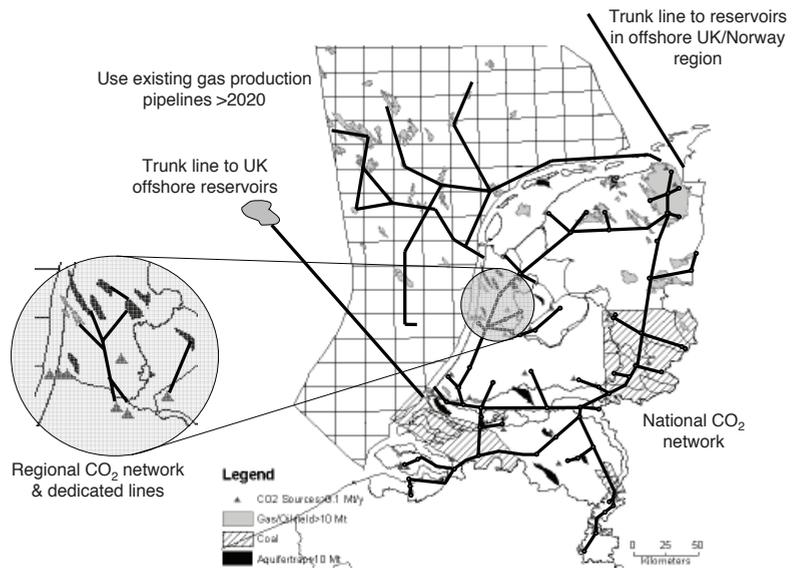


FIGURE 9. Conceptual CO<sub>2</sub> transport configurations

3.4 CO<sub>2</sub> TRANSPORT

CO<sub>2</sub> infrastructure could consist of direct source-sink pipelines or a CO<sub>2</sub> network (see Figure 9). A network, connecting multiple sources with multiple sinks or multiple sources with one sink and vice versa, may be suitable when there are no sufficiently large structures nearby sources. In pathways with clear and ambitious long-term emission reduction targets, in which CCS is most likely to contribute substantially, the construction of a CO<sub>2</sub> network connecting various (future) sources and

sinks may be preferable above dedicated lines. In less ambitious futures with regard to CCS deployment, the gradual build up of capacity is more likely, in which individual plants are directly connected to (nearby) sinks. In some regions, the challenge lies in connecting clusters of small and medium-sized fields. Alternatively, an export terminal could be constructed, from where CO<sub>2</sub> is transported via a large trunk line to one of the large structures in the North Sea. Although not explicitly considered in this study, parts of the existing offshore natural gas infrastructure may be reused for CO<sub>2</sub> transport and injection.

### 3.5 ASSESSING CCS CONTRIBUTION IN DIFFERENT PATHWAYS

Now that we have insight into the storage capacity and the capture potential up to 2050, we combine those pieces of information to compose different CCS pathways<sup>9</sup>, the results of which are summarised in Table 7. The table shows the potential contribution of CCS to achieve the emission reduction targets specified in the different pathways. In order to realise the deep emission reduction in pathway 1, all coal-fired power plants to be constructed in the future are equipped with CO<sub>2</sub> capture. In addition, the most efficient and youngest existing PC units are retrofitted prior to 2020 to achieve the target of 30% reduction versus the 1990 level. Beyond 2020, CCS is also implemented in NGCC units operating in intermediate load and in other sectors (industry and transport), in order to contribute maximally to the 80% emission reduction target in 2050. Still, a large share of the required reductions has to be realised by alternative GHG mitigation options. In pathway 2, the opportunity to retrofit existing PC units is missed. In addition to new-built CCS plants, power plants constructed before 2030 are retrofitted in order to realise the aggressive pathway beyond 2030. By taking nearly all capture opportunities in pathway 1 and 2, roughly 40% of the required emission reduction could be achieved by means of CCS by 2050. In absolute terms, no large CCS penetration occurs in pathway 3, due to the relatively low emission reduction required (domestically), and the fact that only offshore gas fields are available. Only the least costly mitigation options, as occur in blast furnaces and IGCC units dispatched at full load that are located in coastal regions and can be linked to relatively large reservoirs, are implemented. Pathway 4 is in many aspects similar to pathway 1, though existing PC plants are not retrofitted, CCS is implemented less rigorously, and fewer reservoirs are available. Occasionally, plants are faced with a lack of storage capacity, causing some missed opportunities or delay of CO<sub>2</sub> capture.

9. Note that the transport and storage costs, which are computed for each source as will be explained in the next section, also determine CCS deployment.

TABLE 7. Total annual CO<sub>2</sub> emission reductions and CCS contribution in parentheses (mt CO<sub>2</sub>) in different pathways

Pathway	2020		2050	
	Reduction	CO <sub>2</sub> capture at	Reduction	CO <sub>2</sub> capture at
1 'deep reduction'	95 (31)	- Power plants (new-built PC + IGCC, retrofit existing PC) - Blast furnaces - H <sub>2</sub> plant (refinery) - NH <sub>3</sub> plants	300 (112)	- Power plants (new built IGCC + NGCC) - Blast furnaces - Refineries - Steam crackers - F-T diesel plants
2 'postponed action'	0 (0)	Na	250 (108)	- Power plants (new-built IGCC + NGCC, retrofit PC) - Blast furnaces - Refineries - Steam crackers - H <sub>2</sub> plants (transport fuel)
3 'action abroad'	20 (3)	- Blast furnaces	65 (27)	- Power plants (new-built IGCC) - Blast furnaces
4 'ambitious'	65 (15)	- Power plants (new-built PC + IGCC) - Blast furnaces - H <sub>2</sub> plant (refinery)	165 (68)	- Power plants (new-built IGCC) - Blast furnaces - Refineries - Steam crackers - F-T diesel plants

### 3.6 COMPOSING CO<sub>2</sub> TRANSPORT AND STORAGE SUPPLY CURVES:

#### SOURCE SINK MATCHING

Source sink matching is performed by means of an economic optimisation procedure, in which the costs of transport and storage are computed for specific source-sink combinations. Each source is connected to the reservoir for which the total costs of transport and storage are minimal, following the 'who first comes, first gets' principle. A reservoir should in principle have sufficient capacity to store the plant emission for at least 25 years. After this period, the remaining capacity is calculated to decide on the storage strategy for the next period. As the storage capacity of many gas fields and aquifers is lower than 25 years of CO<sub>2</sub> production of large industrial or power plants, we also consider clusters of fields.

For each reservoir (cluster), the number of wells and investment costs (wells, site development, surface facilities and monitoring equipment) are computed using data from [9]. Pipeline dimensions are calculated according the method described in Chapter 3. The choice between a dedicated source-sink line and a trunk line connecting various sinks and sources is based upon NPV minimisation computations. The results of the source-sink matching are summarised in the CO<sub>2</sub> transport and storage supply curves in Figure 10.

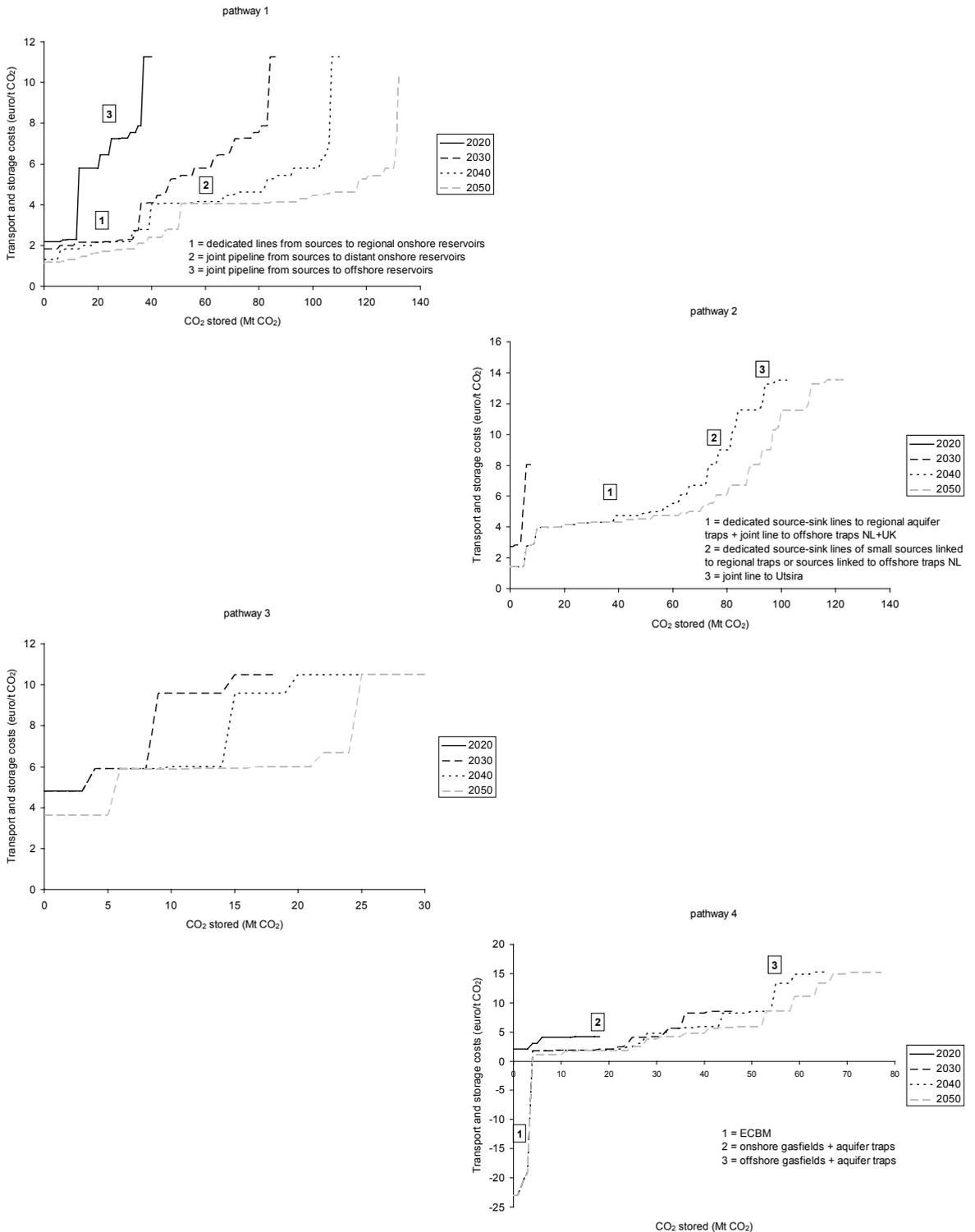


FIGURE 10. CO<sub>2</sub> transport and storage costs in different timeframes. Beware that the scales are different

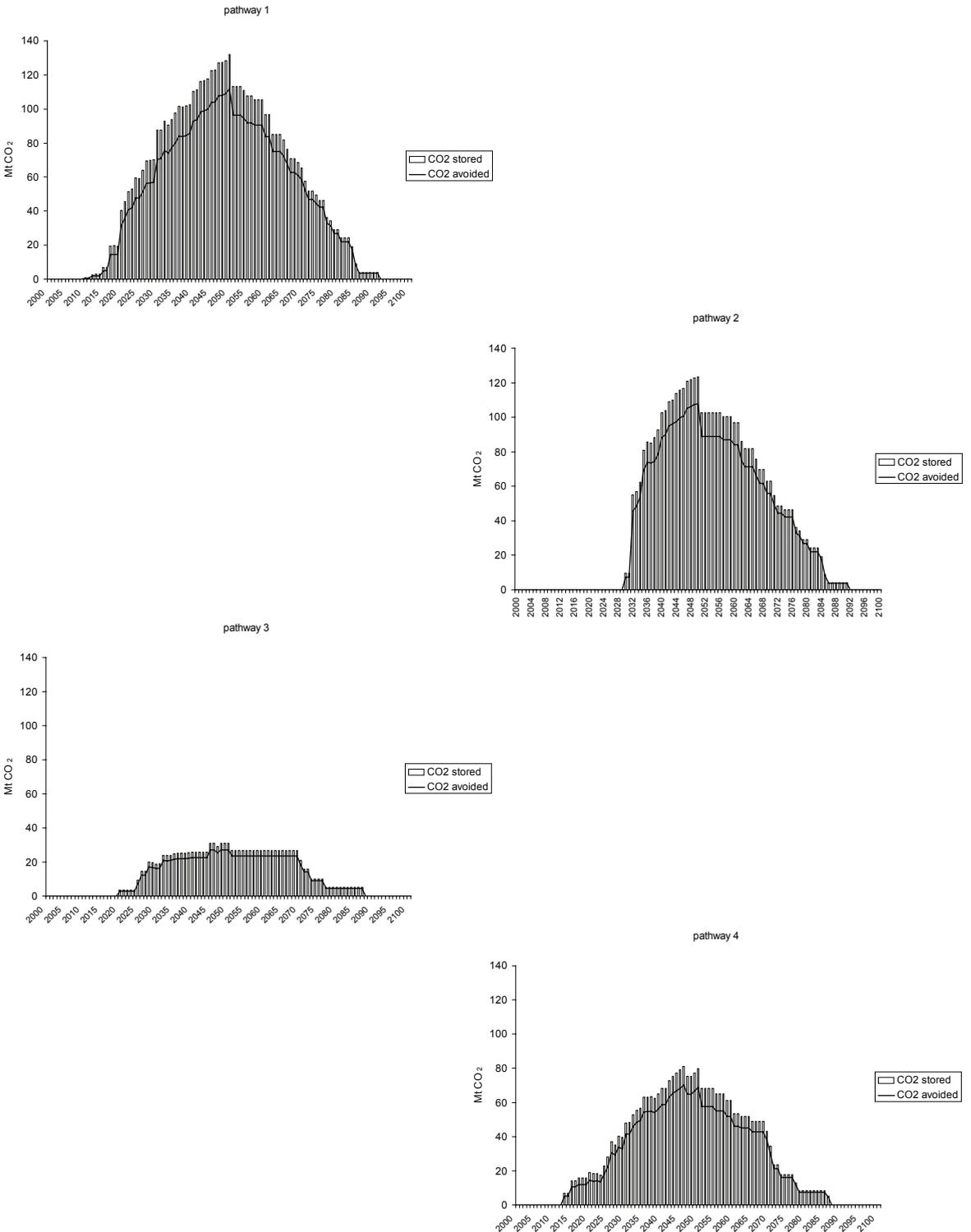


FIGURE 11. Annual quantities of CO<sub>2</sub> stored and avoided in time, including all plants equipped with CCS constructed prior to 2050. Beyond 2050, there are still a number of (mainly coal-fired) plants in operation for a few decades due to the long lifetime of these units

A general trend that can be observed is the decreasing costs in time, which is partly due to the fact that drilling costs (representing a large share of storage costs) are assumed to be reduced by 30% between now and 2020 [9]. Transport costs are modest in most pathways, which can be explained by relatively small transport distances, large CO<sub>2</sub> flows and the scale advantage of trunk lines. The need to retrofit PC units and the construction of new power plants with CO<sub>2</sub> capture in the Rijnmond area around 2020 in pathway 1, in combination with the lack of regional storage capacity, makes it attractive to construct a large trunk line to offshore reservoirs. Ten years later, a trunk line of similar dimensions as current gas transmission lines is constructed to transport CO<sub>2</sub> from sources located in the southwest to the Groningen gas field. Similarly, the clear turning point in climate policy in pathway 2 causes many plants to be retrofitted in a short period. Together with the lack of large aquifer traps onshore, this opens up possibilities for 'joint' transport. Two large trunk lines are constructed to large offshore structures. One connects the sources in the Rijnmond area to the Bunter Sandstone traps in the UK sector of the North Sea (to which also traps in Dutch part of the North Sea can be connected). The second line transports CO<sub>2</sub> from the Eemshaven region to the Utsira formation. As the construction of these trunk lines takes several years, many plants will go on stream somewhere between 2032 and 2035. In pathway 3 and 4, high transport and storage costs and the lack of regional storage availability, respectively, prevent or postpone the application of CCS at several, notably small, sources. In spite of the pure CO<sub>2</sub> available at early opportunities, transport and storage costs make overall costs too high. Possibly, synergy could be found in transporting CO<sub>2</sub> from the early opportunities and the steam crackers to aquifer traps in Brabant or the Rijnmond. In pathway 4, we observe that various plants store CO<sub>2</sub> in a cluster of offshore depleted gas fields using a dedicated line, which results in relatively high costs. Around 2030, plants will compete for storage in the coal seams in Limburg and eastern part of the country. Transport and storage costs may become negative when storing in coal seams, because gas prices (and hence coal bed methane revenues) prevailing in this scenario increase strongly beyond 2020.

#### 4 RESULTS

Combining the information generated in the previous sections enables us to present the quantities of CO<sub>2</sub> stored and avoided in time for the different pathways, as illustrated in Figure 11. In pathway 1, representing the most ambitious CCS scenario, nearly 3 Gt CO<sub>2</sub> is stored by 2050. The cumulative CO<sub>2</sub> production of all sources that go on stream before 2050 equals nearly 40% of the Dutch technical storage potential. Figure 12 shows that if all CO<sub>2</sub> would be stored in Dutch reservoirs that become available between 2005 and 2030, assuming every gas field and aquifer trap will be available and suitable for CO<sub>2</sub> storage, injection

could be continued to roughly 2050. As this condition is unlikely to be met, we can conclude that such a scenario would only be possible if one of the mega structures, in this case the Groningen gas field, becomes available prior to 2050. In pathway 2, the Groningen gas field and other gas fields are not available, enforcing  $\text{CO}_2$  storage in aquifers. Roughly 96% of the nearly 2 gt  $\text{CO}_2$  captured between 2030 and 2050 is stored in the rich Bunter Sandstone formation in the UK part of the North Sea and the vastness of the Utsira formation. This illustrates the need to look beyond the national borders when large ccs deployment rates are strived for and gas fields are not available in such high numbers as in pathway 1. In pathway 4, about 1.7 gt is stored more homogeneously in Dutch reservoirs up to 2050, requiring about 73% of the total storage potential (excluding the Groningen gas field).

It could be argued whether the peak of ccs will occur in 2050. Given fossil fuel reserves, the peak may be extended some decades, which is also observed in certain scenarios presented in [63]. If we assume that the storage rates in 2050 are maintained up to 2100, the cumulative storage would lie between 2.2 and nearly 10 gt by that time, representing roughly 20-85% of the entire Dutch technical storage potential, including Groningen. Excluding Groningen and reservoirs abroad, we would run out of technical storage capacity somewhere between 2050 and 2075 if storage rates above 80 mt/yr are continued beyond 2050. In a more modest scenario where storage rates are gradually increased and then kept constant at 30 mt/yr, we could continue storing  $\text{CO}_2$  to at least 2100.

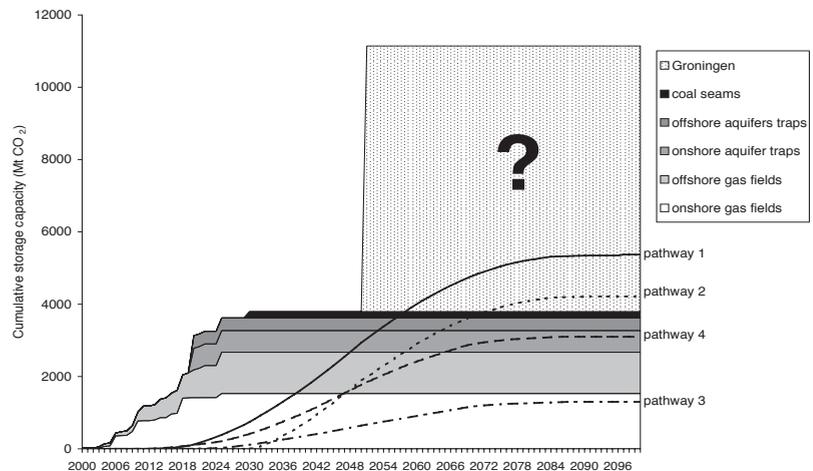


FIGURE 12. Storage availability in time versus cumulative  $\text{CO}_2$  storage

The realised ccs potential and the overall  $\text{CO}_2$  mitigation costs, including transport and storage, are given in Figure 13. By 2020, up to 30 mt  $\text{CO}_2$  could be reduced at marginal costs of roughly 50 €/t  $\text{CO}_2$  in a scenario where retrofitting the most efficient pc units would be feasible and a part of the storage capacity in the British part of the North Sea is

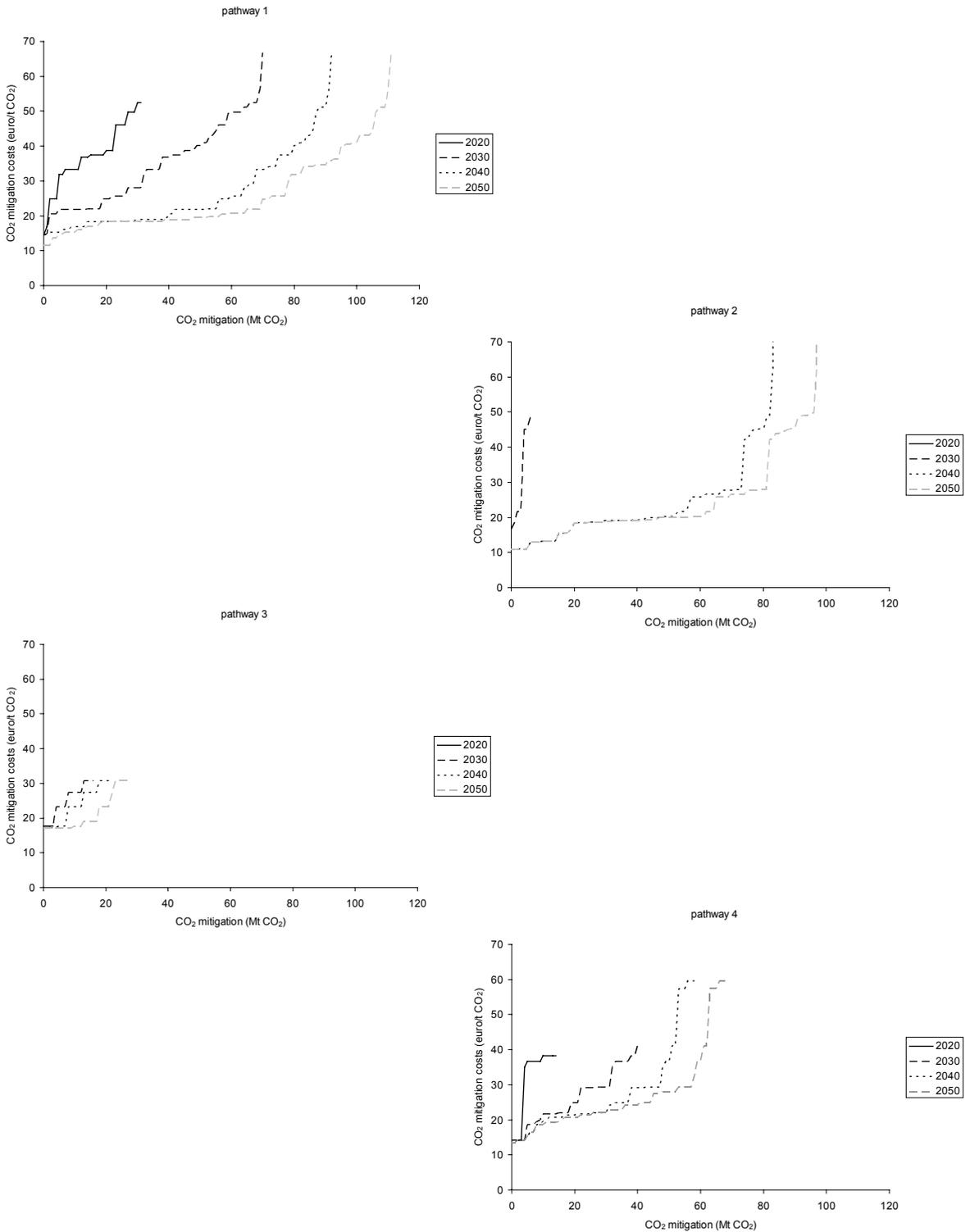


FIGURE 13. CO<sub>2</sub> mitigation costs of realised CO<sub>2</sub> reductions in time, including CO<sub>2</sub> transport and storage

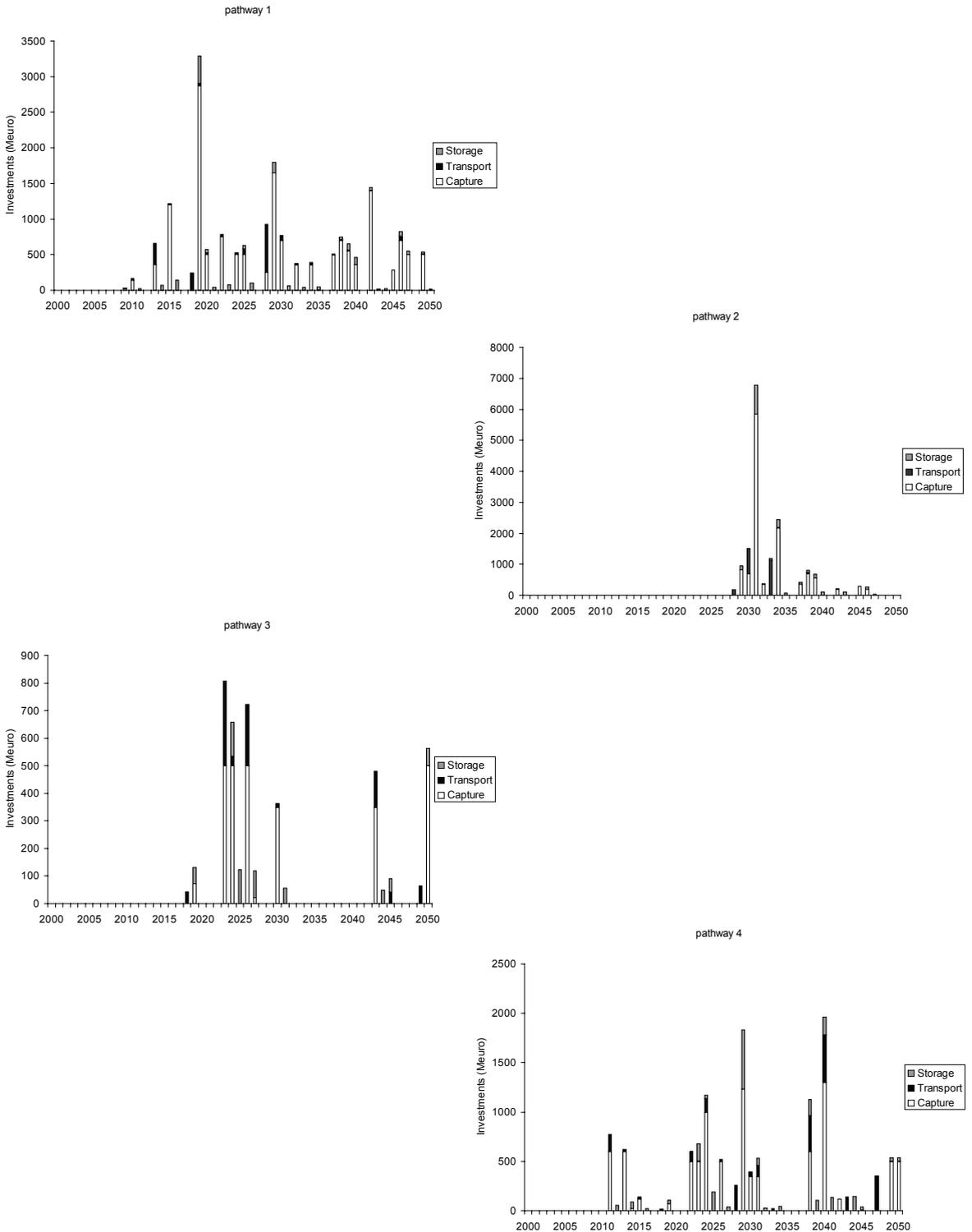


FIGURE 14. Investments patterns of CO<sub>2</sub> capture (excluding power plants), transport and storage (Euro<sub>2000</sub>). Beware that the scales are different

available. Excluding the retrofit options, 15 mt CO<sub>2</sub> could be avoided at costs below 40 €/t CO<sub>2</sub> by capturing CO<sub>2</sub> from blast furnaces and new-built coal-fired power plants and storing it in Dutch gas fields and aquifer traps. Halfway this century, about 60 mt CO<sub>2</sub> can be reduced at costs below 40 €/t CO<sub>2</sub>, and in pathways 1 and 2 even below 20 €/t CO<sub>2</sub>. The majority of this potential is represented by the many new coal-fired power plants to be installed. By 2020, electricity production from plants equipped with CO<sub>2</sub> capture lies between 0% and 35% of total electricity supply. Thirty years later, this number has increased to 20% in pathway 3 up to 70% in pathway 1. Note that the costs to avoid the first 20 mt in pathway 2 are somewhat lower in comparison to pathway 1 as the costs to retrofit new-built plants in a later stage are lower than constructing a plant with CO<sub>2</sub> capture right away. This is due to the fact that investments in capture equipment are postponed and hence the additional costs of retrofitting are modest in real terms. On the other hand, costs in pathway 2 are expected to be higher in comparison to pathway 1, where cost reductions could be realised in the period between 2010 and 2030.

In the course of time, costs are reduced because of advances in more efficient capture and conversion technologies. In pathway 4, however, the cost reduction of CO<sub>2</sub> capture in time is partly outweighed by the combined effect of lower dispatch and the increased costs to transport and store CO<sub>2</sub> in relatively smaller reservoirs.

In the upper regions of the chart, above 40 €/t CO<sub>2</sub>, we encounter new-built PC and NGCC units with CCS operated in intermediate load, and boilers, heaters (at refineries) and steam crackers retrofitted with a post-combustion capture unit. In pathway 2, the marginal costs may rise up to 300 €/t CO<sub>2</sub> for the last mega tonnes avoided in the transport sector by the application of clean hydrogen.

In order to understand what such CCS deployment rates means financially, the additional investments in capture and compression units, pipelines and storage facilities over time are presented in Figure 14. In overall terms, total CCS investments up to 2050 have been framed at 6 to 23 billion Euros. The share among capture, transport and storage varies between 85%, 7% and 8% for pathway 1 and 71%, 14% and 15% in pathway 4, respectively. Not surprisingly, the investment pattern is quite diverse; in pathway 1, a relatively homogenous distribution is observed, with an occasional peak due to massive retrofitting (2020) or the construction of a trunk line. Pathway 2 shows a concentration of investments around 2030, also due to retrofits and trunk line investments. Pathway 4 is characterised by a peak in investments in 2030, which can be explained by the numerous wells that need to be drilled to inject CO<sub>2</sub> into the coal seams. Both pathway 3 and 4 have in common that there are periods of a few years up to 10 years in which hardly any investment occur. This feature can be explained by the vintage structure of the power plants, assumed lifetimes and growth in energy demand in the TM baseline.

A trunk line is attractive when several plants in a specific region go on stream in the same period and the storage capacity in vicinity is limited. The vintage structure and the locations of new plants are decisive, as well as the timing and extent of emission reduction. The ambitious goals in pathway 1, the sudden need for action in pathway 2 in combination with a lack of storage capacity onshore, and the need to go offshore in pathway 3 make the construction of trunk lines attractive. In pathway 1 and 4, a 250 km trunk line is constructed between the Rijnmond area and the gas fields in the northern provinces with a diameter of at least 40", costing circa 350 to 600 m€. A 250 km trunk line to the southern North Sea basin of the UK (via the reservoirs in the Dutch part of the continental shelf) is another interesting trajectory, as the power stations in the Rijnmond area that are constructed and/or retrofitted the next decade will begin producing CO<sub>2</sub> when the bulk of the gas fields in that region are not yet available. The capacity we forecasted is between 10 and 20 mt/yr (26-36"), depending on the need to retrofit PC units. A pipeline of such dimensions is framed at roughly 200-300 m€. In pathway 2, a 750 km 42" trunk line is constructed to transport over 20 mt CO<sub>2</sub> from the Eemshaven region to the Utsira formation, costing over 1 billion Euros. This option should be considered if ambitious CO<sub>2</sub> emission reduction targets are strived for, while the storage potential in gas fields is scarcely available for CCS. As the marginal transport costs per tonne of CO<sub>2</sub> are modest beyond such scale, it may be beneficial to construct a larger pipeline that also allows power plants in Northwest Germany to make use of it.

## 5 DISCUSSION

In this section, we will elaborate on the findings produced in the different pathways and the uncertainties that we need to face in developing strategies for CCS deployment. Some critical remarks are justified on the cost figures in this analysis, both on the values and on their interpretation.

### 5.1 CO<sub>2</sub> CAPTURE POTENTIAL AND DEPLOYMENT

There are various factors we did not account for that may alter, either positively or negatively, the capture potential we have reported (see Table 8).

TABLE 8. Impacts of assumptions and uncertainties on capture potential.  
The quantified impacts represent upper boundaries

Assumption/uncertainty	Timing impact	Indicative impact on capture potential
<i>Factors that decrease CO<sub>2</sub> capture potential</i>		
Neglecting the potential of renewables <sup>a</sup>	Beyond 2020	5-20 mt CO <sub>2</sub> /yr
Assuming activity level in the carbon-constrained scenarios is equal to that in the baseline scenarios <sup>b</sup>	Short + long term	Potentially large
Ignoring coal to gas of switching <sup>c</sup>	Short - medium term (postponing CCS)	5-25 mt CO <sub>2</sub> /yr on average
<i>Factors that increase CO<sub>2</sub> capture potential</i>		
Fix CO <sub>2</sub> emissions refineries industry in time <sup>d</sup>	Short + long term	2-8 mt CO <sub>2</sub> /yr
Fix CO <sub>2</sub> emissions petrochemical industry in time <sup>e</sup>	Short + long term	10-18 mt CO <sub>2</sub> /yr by 2050
Fix CO <sub>2</sub> emissions iron and steel production in time <sup>f</sup>	Short + long term	10 mt CO <sub>2</sub> /yr by 2050
Retirement of inefficient PC units <sup>g</sup>	Short - medium term (<2030)	9 mt CO <sub>2</sub> /yr
Capture potential alternative fuel production <sup>h</sup>	Short + long term	Unknown

<sup>a</sup> The baseline scenarios we adopted are characterised by a gradual phase-out of renewables as incentives are abolished; by 2020, 30 TWh is produced from renewable resources, which is reduced to 3 TWh in 2050 [18]. However, the competitiveness of renewable energy technologies depends on the assumed learning rate (see e.g. [64]), feedstock prices in case of biomass and oil prices, which are relatively low in the GE/TM baseline scenarios (20-30 \$/bbl). In the high oil price variant of GE, where the oil price rises to 45 \$/bbl, the contribution of renewable energy is 3 TWh as well [18]. Assuming high learning rates and currently prevailing oil prices will be structural, some renewable energy sources may be competitive with fossil energy. As a minimum, we assume the current onshore wind capacity is maintained (at the expense of coal-fired capacity with CCS) and as maximum, the forecasted production from renewables in 2020 in the GE scenario is maintained.

<sup>b</sup> By implementing climate policy, the starting points of the baseline scenario will actually change, e.g. the demand for energy is likely to decrease due to efficiency measures and consumption patterns, thereby reducing CCS potential.

<sup>c</sup> Johnson and Keith [7] calculate that approximately 20% of the emissions are reduced by carbon-ordered dispatch and increased use of gas-fired capacity for gas prices starting at 4.2 \$/GJ (comparable to prices in GE/TM) before CCS enters the market. This corresponds to a cumulative reduction up to 2050 of 300 up to 1000 mt CO<sub>2</sub>. It is the question though whether utilities will choose for more gas-fired capacity given the risk of increasing gas prices and the possibility that a strict climate policy is implemented (increasing gas prices even further), which would result in a high-cost technology path [7]. In addition, the Dutch power sector is already relying heavily upon natural gas. As utilities in the Netherlands strive for portfolio diversification, a situation in which the balance would turn towards more gas is unlikely, also considering the fact that this implies a stronger dependency on gas exporting countries the coming decades.

<sup>d</sup> The crude oil throughput grows from approximately 55 Mt now to 93 Mt in GE and 70 Mt in TM by 2040 [18]. Current emissions are approximately 11 Mt CO<sub>2</sub> [1]. Note that the hydrogen demand at refineries is projected to increase, due to the shift to heavier crudes, the growth in demand and stricter sulphur standards for transport fuels. Merchant hydrogen, generally produced by means of relatively small-scale SMR or ATR units with a PSA for hydrogen purification at which CO<sub>2</sub> capture is not attractive, may cover the gap in first instance. Alternatively, the growth in H<sub>2</sub> demand could be covered by gasification of oil residues with CCS.

<sup>e</sup> In GE and TM the (physical) growth in the chemical industry is 2.5%/yr [18]. We assume no efficiency improvements, a current ethylene and ammonia production of 3.7 and 2.7 mt/yr estimated from current production capacities and a CO<sub>2</sub> emission factor of 1.7 t CO<sub>2</sub>/t ethylene and 1 t (net) CO<sub>2</sub>/NH<sub>3</sub> [65]. Note that ammonia plants might be operated in lower load or even shutdown in the longer term due to increasing natural gas prices, which represents the lower value presented in the table.

- <sup>f</sup> In GE and TM the (physical) growth in the iron and steel industry is 1.3-1.4%/yr [18]. We assume no efficiency improvement, a current steel production of 6.6 mt/yr and a CO<sub>2</sub> emission factor of 1.7 CO<sub>2</sub>/t steel [1]. Note that we restricted our analysis to blast furnace gas used on-site. Roughly 10 mt CO<sub>2</sub> is produced in the blast furnaces and basic oxygen furnace, of which 5.5 mt is emitted in the nearby located power plants, the rest being emitted within the boundaries of the steel plant. As CO<sub>2</sub> is present at high partial pressure and produced on a single site, these streams appear interesting targets for low-cost CO<sub>2</sub> capture by means of water-gas shift and physical absorption. In addition, it enables fuel upgrading, i.e. separating the remaining N<sub>2</sub> and H<sub>2</sub> after CO<sub>2</sub> removal, resulting in increased conversion efficiencies [57].
- <sup>e</sup> Assuming the least 3 inefficient PC units would be retired. Wise and Dooley [8] illustrate that a cut to one-third of current emissions by 2050 induces the retirement of the least efficient coal capacity by 65%. These units would mainly be replaced by IGCC with CCS. Note that the PC units considered in [8] are less efficient than the least efficient PC units in the Netherlands.
- <sup>b</sup> Currently prevailing oil prices, which varied between 55 and 70 \$/bbl when this research was performed, are structurally higher than forecasted oil prices in the GE and TM scenarios used in this analysis. In scenarios with structural oil prices of 60-70 \$/bbl, F-T diesel produced from coal and biomass with CCS is calculated to be nearly competitive with fossil diesel even without a carbon price [58].

With the exception of pathway 3, the CCS pathways in this analysis represent radical scenarios (that we created on purpose to investigate the boundaries of CCS). According to the IEA World Energy Outlook, CCS will start playing a role in their most ambitious scenario to cap CO<sub>2</sub> emissions in 2030 at today's level (identical to pathway 3) [66]. In this scenario, the share of coal-fired plants with CCS in global power generation would reach 8% by 2030, which is even lower than the value of 13% we calculated in pathway 3.

As noted in the introduction, an analysis on options to reduce Dutch GHG emissions by 2020 [2] forecasted a maximum CCS contribution of 15 mt, of which approximately 5 mt from industrial processes emitting pure CO<sub>2</sub> and 10 mt from large-scale industrial CHP units. Our study makes clear that this 5 mt is an overestimation; we estimate approximately 2.5 mt CO<sub>2</sub>/yr could be avoided by CCS at pure sources. However, there are no proven reservoirs in vicinity of the ammonia plants and transport & storage costs to more remote reservoirs are relatively high as volumes are modest. In addition, we doubt that CHP units are the first targets for CCS; all signs indicate that the deployment is foreseen at coal-fired power plants first.

For the year 2050, the Dutch emission reduction potential of CCS has been estimated at 25-30 mt CO<sub>2</sub> in energy production, 4-8 mt in the transport sector and 0(!) mt in industry [67]. The figures we produced indicate that the potential in the power sector and industry is probably much higher. The figure of 25-30 mt is representative for a scenario where the storage potential is limited to Dutch gas fields excluding the Groningen gas field.

## 5.2 TRANSPORT COSTS

For pipeline costs, we used generic figures for onshore and offshore conditions, without accounting for the specific Dutch context, which is characterised by a relatively high population density. In addition, various natural obstructions such as rivers may have to be crossed.

These factors may add significantly to transport costs and need to be investigated, specifically for the trajectory between the Rijnmond area and the northern provinces. In recent years, industry has also experienced an increase in steel and labour costs, which will certainly affect the costs of pipelines and wells.

Transport (and storage) costs may be reduced, either by optimising the spatial planning of new power plants or by reusing infrastructure. The Eemshaven region is an interesting new location for power plants due to the vicinity of many gas fields, among which a few relatively large reservoirs. Obviously, installing plants in the north will sooner or later require investments in power transmission capacity, although another 2 GW<sub>e</sub> can be connected to the transmission grid in the northern provinces at present [68]. We identified some sink-source combinations in which reuse may be worthwhile considering, such as the steel plant in the IJmond region storing CO<sub>2</sub> in one of the gas fields in the P or K quadrant. Possibly also smaller pipelines may be reused in the concept where gas fields are used as buffer, as ‘branches’ of the main trunk line heading for the oil fields up north.

### 5.3 CAPTURE COSTS

Capture costs should be considered as indicative figures, as we could not account for the specific conditions of individual plants. To take the example of refineries, CO<sub>2</sub> has to be captured from flue gas of various boilers fired with different fuels. As no data on the individual sources were available, we estimated CO<sub>2</sub> emissions for the entire refinery and used generic or average figures on flue gas concentration and waste heat availability. The latter is an important factor for the economic feasibility of post-combustion capture in the industrial sector; mitigation costs may increase with more than 10 €/t CO<sub>2</sub> when no waste heat is available. Shell has calculated that the costs to avoid 1 Mt of CO<sub>2</sub> by means of capture from a stack at their refinery in Pernis, assuming all energy required for the reboiler is produced in a gas-fired boiler, would be 70 €/t CO<sub>2</sub> [69]. Mitigation costs we calculated for similar conditions are approximately 60 €/t CO<sub>2</sub>, indicating costs may have been underestimated in some specific cases.

Capture costs at IGCC units are rather low as well. In the scenarios we considered, many IGCC units (with CO<sub>2</sub> capture) have a load factor near 90%, a value that is still not realised with today’s IGCC unit in Buggenum (without capture) [70]. It can therefore be argued that such high loads in combination with high efficiencies are somewhat optimistic considering their operational characteristics. As we have seen in the TM scenario, several coal-fired power plants will be dispatched less often when nuclear comes in and the role of natural gas is diminishing. Therefore, the flexibility in changing load factor at IGCC units and the impact of CO<sub>2</sub> capture in this should be analysed in more detail.

### 5.4 OVERALL MITIGATION COSTS

In conclusion, the costs associated with all elements from the CCS chain

may well be optimistic. A comparison with values reported in literature seems to support this notification; costs are on the low side of the range reported in the IPCC special report on CCS [63]. This is due to a combination of future performance and cost figures of capture installations we assumed and the relatively low costs for CO<sub>2</sub> transport and storage in the Netherlands.

The trend in mitigation costs as can be seen in Figure 13 illustrates that the costs will decrease in time due to the fact that more efficient and less costly technologies will be installed by then, which might be interpreted as a plea for a wait-and-see policy. This would, however, be a wrong interpretation of the results, as costs reductions need to be achieved partly by learning-by-doing. In order to reach the long-term costs as shown in the figure, CCS capacity should be installed the coming decades to gain operational experience and enable cost reduction. We have not specifically accounted for the effect of learning as a function of installed capacity, as this would require an estimation of worldwide installed CCS capacity.

As we have warned for in Chapter 3, CO<sub>2</sub> mitigation costs should be interpreted with great care, as the chosen reference system is decisive. In pathway 2, the mitigation costs of replacing fossil gasoline by clean H<sub>2</sub>, produced from fossil fuels with CCS, are relatively high in comparison to CCS in the power and industrial sector. This seems to suggest that decarbonising the transport sector by means of clean hydrogen is low on the priority list of mitigation options. Two important notes are justified here. First, the costs strongly depend on FCV costs and efficiency, which makes the mitigation costs rather uncertain. Secondly, if these fuels are being introduced for other reasons than CO<sub>2</sub> emission reduction only, such as concerns over supply security and local air pollution, the additional costs to capture CO<sub>2</sub> are modest (around 10-25 €/t CO<sub>2</sub>) [44,59].

## 6 CONCLUSION

CCS has the potential to become an important mitigation option in curbing Dutch CO<sub>2</sub> emissions, which currently approach 180 Mt CO<sub>2</sub>/yr. This study shows what the role of CCS could be in time given the CO<sub>2</sub> capture and storage potential, and uncertainties involved. We found that maximally 50 Mt CO<sub>2</sub>/yr could be avoided in 2020 by capturing CO<sub>2</sub> from various sources at costs between 10 and 100 €/t CO<sub>2</sub> (excluding transport and storage), versus a baseline scenario characterised by relatively large economic growth and strong increase in energy demand. Only sources emitting at least 0.1 Mt CO<sub>2</sub>/yr were included in the analysis, being power plants, blast furnaces, boilers and heaters in refineries, steam crackers, reformers and gasifiers. The potential includes about 20 Mt CO<sub>2</sub>/yr that may be avoided by retrofitting existing PC units, which may be an important back-stop option to achieve a target of 30% CO<sub>2</sub> reduction in 2020 versus 1990. However, there are

some technical drawbacks (e.g. lack of space, reduction in power output) and costs are rather high: 40-50 €/t CO<sub>2</sub> avoided for the most efficient PC units. By 2050, the forecasted capture potential is 80-110 Mt CO<sub>2</sub> avoided/yr, of which 60-80 Mt CO<sub>2</sub>/yr may be realised below 20 €/t CO<sub>2</sub> (excluding transport and storage). From the different sectors contributing to this potential, the power sector offers the biggest opportunities for CO<sub>2</sub> capture. The mitigation potential is estimated at 11-14 Mt CO<sub>2</sub>/yr by 2020 (excluding retrofit) and 60-84 Mt CO<sub>2</sub>/yr by 2050, assuming electricity supply increases from 95 TWh in 2005 up to 210 TWh in 2050. Industrial sources add another 16 Mt CO<sub>2</sub>/yr, of which 2.5 Mt/yr in pure CO<sub>2</sub> sources, at least 3 Mt/yr in steel production, 4.5 Mt/yr in ethylene production and 6 Mt/yr in boilers and heaters at refineries. The development of a market for alternative fuels produced via syngas production with CCS creates an opportunity to decarbonise the transport sector. The reduction potential for F-T diesel and H<sub>2</sub> has been estimated very roughly at 10 Mt CO<sub>2</sub>. Most economic capture options exist in the production of power by means of IGCC, the production of hydrogen, ammonia, ethylene oxide, steel and F-T diesel, and gas processing. It is recommended to perform a more detailed study on the prospects for CCS in the steel industry, representing one of the largest CO<sub>2</sub> point sources in the Netherlands. Also additional research is required to get insight into the emission reduction potential of F-T diesel produced by coal and biomass gasification with CCS.

In our analysis, the actual deployment of the capture potential is determined by the emission reduction targets and the geological capacity available for CO<sub>2</sub> storage. The uncertainty in these two decisive factors has been dealt with by performing scenario analyses. We sketched four CCS deployment pathways that allow us to get an idea on what emission reductions are reasonably achievable by means of CCS, and what may be possible if we 'pull out all the stops'. We found that 15 Mt CO<sub>2</sub> could be avoided annually by 2020 when some of the larger Dutch gas fields that are forecasted to become available the coming decade could be used for CO<sub>2</sub> storage. If, in addition, part of the existing PC units or large industrial boilers and furnaces is retrofitted with CO<sub>2</sub> capture, we may have to rely on the large reservoirs in the UK part of the North Sea. Alternatively, clusters of relatively small gas fields could be used, which could add 15 €/t CO<sub>2</sub> for transport and storage.

If the Netherlands focuses on national opportunities and does not consider storage options abroad, 30 Mt CO<sub>2</sub>/yr could be avoided by 2050 with relatively small efforts. Provided a good planning is being set up to ensure the domestic storage potential that will become available could be used for CO<sub>2</sub> storage, we may even avoid up to 60 Mt CO<sub>2</sub>/yr in 2050. In aggressive climate policies aiming for 50-80% reduction in CO<sub>2</sub> emissions by 2050 versus 1990, avoiding another 50 Mt CO<sub>2</sub>/yr may be possible provided that nearly all capture opportunities that occur are taken. Storing such large amounts of CO<sub>2</sub> would, however, only be possible if one of the mega structures, either the Groningen gas field,

large reservoirs in the Bunter Sandstone formation or the Utsira formation in the North Sea, would become available. Another option could be to store CO<sub>2</sub> in aquifer formations that are not structurally trapped. However, the availability of Groningen is highly uncertain, the availability of the UK reservoirs is depending on the application of CCS in the UK and exploiting Utsira requires a costly offshore CO<sub>2</sub> infrastructure. Excluding Groningen, the reservoirs abroad and factors limiting the storage potential, the Netherlands would run out of storage capacity sometime between 2050 and 2075 for storage rates above 80 mt/yr. For more modest storage rates of 30 mt CO<sub>2</sub>/yr, we could continue storing up to the year 2100.

Gas fields seem the most appropriate candidates for CO<sub>2</sub> storage given their large and, generally, secure capacity. Over 70% of the potential in gas fields is represented by the Groningen gas field, which will not be available prior to 2040 and possibly (far) beyond 2050. Insight into the feasibility of different applications and injection strategies prior to depletion are indispensable in judging what the large storage capacity of Groningen could mean for the future of CCS. The remaining 30% is represented by average and small-sized fields. Not all of these reservoirs will be available due to competition with alternative applications, most notably UGS, and due to geological conditions that complicate storage or imply a high risk of leakage. Many gas fields will be abandoned before 2025 given current production projections, followed by a gap of at least two decades before the Groningen field might become available for CO<sub>2</sub> storage. Large-scale deployment of CCS is not due to start before 2012. A future is conceivable in which plant owners will face a lack of suitable storage reservoirs in vicinity when the reservoir used in the first half of the plant lifetime is abandoned. In the best case, several small structures or more remote traps will have to be exploited, resulting in higher transport and storage costs. In the worst case, plants will have to be operated without capture. In order to guarantee sufficient storage capacity in the spring of a potential CCS era - sometime between 2020 and 2040 - a strategy must be developed to bridge this gap and assure the possibility of CO<sub>2</sub> storage in a later stage.

In conclusion, the challenge for the coming years is to find suitable (clusters of) reservoirs that will become available at the right time, have sufficient storage capacity and will not be used for other purposes. If it appears that natural gas reservoirs cannot provide sufficient storage potential (at the right time), the Netherlands may have to rely on aquifers and, possibly, coal seams. However, the uncertainty in capacity and the technical/economic feasibility of CO<sub>2</sub> storage do not make these options a very safe bet for large-scale CCS deployment. Therefore, more work is required in geological characterisation and mapping of aquifers and their traps, and demonstrating ECBM.

Throughout this chapter we have mainly discussed CO<sub>2</sub> capture and storage potentials. The actual deployment for CCS, however, is to a large

extent driven by national and international policy choices. When the EU and the Netherlands postpone climate policy and the need for far-reaching mitigation will become apparent in a few decades, overall costs may increase as the opportunities of technological learning, CO<sub>2</sub>-EOR at the North Sea and infrastructure reuse are missed, and possibly more expensive reservoirs will have to be used. In a future where the concerns about climate change force us to do everything we possibly can beyond 2010, we probably have to transport CO<sub>2</sub> beyond the Dutch part of the continental shelf. This would require an internationally coordinated action to plan the CO<sub>2</sub> streams and realise the construction of an offshore CO<sub>2</sub> infrastructure. As neighbouring countries with a lack of storage capacity will have to rely on reservoirs located at the Dutch shore and North Sea, we need to extend our view beyond national borders and think within a Northwest European context.

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## CHAPTER VIII

### SUMMARY AND CONCLUSIONS

#### I INTRODUCTION

The sense of urgency in achieving large reductions in anthropogenic CO<sub>2</sub> emissions, driven by concerns over the impacts of human induced climate change, has increased the interest in carbon dioxide capture and storage (CCS). CCS can be defined as the separation and capture of CO<sub>2</sub> produced at large stationary sources, followed by transport and storage in geological reservoirs, the ocean or minerals in order to prevent its emission to the atmosphere. It is generally regarded as an intermediate solution in the evolution towards a sustainable energy system in the long term, which is based on more durable solutions. CCS enables to continue the inevitable use of fossil fuels and its infrastructure, while simultaneously realising strong CO<sub>2</sub> emissions reduction. The application of CCS will continue the use of coal, whose abundant reserves are larger and distributed more evenly across the globe in comparison to oil and natural gas. Hydrocarbon recovery could be increased by injecting CO<sub>2</sub> into nearly depleted oil fields, and may be increased by CO<sub>2</sub> injection into gas fields or coal seams. In this way, CCS could be of great value in unifying two key priorities in the energy policy of many nations: reducing both greenhouse gas (GHG) emissions and the vulnerability of energy supplies caused by the dependence on oil and natural gas imports. CCS could also fulfil a bridging function in developing a hydrogen infrastructure as hydrogen could be produced from fossil fuels with CCS at relatively low costs.

There are, however, a number of drawbacks and fundamental arguments against the application of CCS. First, CCS is an end-of-pipe technology as it prevents CO<sub>2</sub> to be emitted into the atmosphere instead of preventing its production. Therefore, it is a non-structural solution to the problems of climate change caused by fossil fuel use. Second, the energy consumption and costs of CCS are significant. As more primary energy is required to produce the same output, CCS induces a stronger reliance on fossil fuels with all associated externalities. Third, CO<sub>2</sub> needs to be stored at least hundreds and possibly thousands of years, which involves certain risks, most notably CO<sub>2</sub> leakage.

The concept of CO<sub>2</sub> capture and storage into geological reservoirs as greenhouse mitigation option is relatively young. Only few commercial, large-scale projects with the purpose of underground CO<sub>2</sub> storage exist, most of which involve CO<sub>2</sub> removal during natural gas processing. The techniques used for CCS, however, are not new. The oil and chemical industry has many years of experience with CO<sub>2</sub> separation, treatment, transport and injection into geological formations. The challenge in the coming years is to scale up the different components and integrate them into CCS chains, thereby making use of current experience.

There are, however, still a number of scientific challenges ahead of us. Some of the major issues that need to be addressed are:

1. *Techno-economic performance of CO<sub>2</sub> capture.* Numerous options for CO<sub>2</sub> capture have been identified and assessed on their thermodynamic performance and costs. The results need to be standardised in order to allow a comparison of the different options. In addition, the calculated figures need to be verified by full-scale demonstration of integrated CCS schemes. Commercial application will be crucial in bringing down the costs, together with R&D in innovative options that promise highly-efficient, low cost CO<sub>2</sub> capture in the longer term.
2. *Source-sink matching.* In the coming years, detailed regional assessments should be performed to match CO<sub>2</sub> sources and sinks. More advanced models are needed that incorporate developments in time (both sources and sinks) and enable calculations on multiple sink-source combinations and networks.
3. *Capacity and risks of geological storage.* We do need more accurate estimations of global and regional storage capacities together with more detailed assessments of the risks of underground CO<sub>2</sub> storage. In this way, the feasibility of CO<sub>2</sub> storage for specific reservoirs can be evaluated and the storage capacity can be better judged. Also the demand and potential of alternative applications, such as underground gas storage (UGS), need to be assessed. By accounting for the risks and alternative applications, as well as economic, legal and regulatory barriers, ultimately more realistic and viable storage capacities can be derived.
4. *Role of CCS in GHG emission reduction strategy.* As we are on the threshold of CCS deployment, it is about time to come up with strategies for large-scale CCS deployment as part of the transition towards a more sustainable energy system. For this purpose, we need insights into the way CCS may evolve and what its role in reducing greenhouse gasses could be in conjunction with alternative options.

## 2 OBJECTIVE AND SCOPE OF THIS THESIS

The objective of this thesis is to assess the technical possibilities, costs and risks of CCS systems to allow comparison, identify promising options and determine how and to what extent CCS could be deployed over time in the Netherlands. In the coming decades, CCS may play a significant role in decarbonising the Dutch energy and industrial sector, as there are numerous large CO<sub>2</sub> sources and potential sinks. The Netherlands also has decades of industrial experience with the transmission, distribution and processing of gasses, especially natural gas, which could be of great value in developing a CO<sub>2</sub> infrastructure. We focus on geological storage of CO<sub>2</sub>, because this is the most obvious storage medium in the Netherlands. We look upon issues from a system perspective, which means that we unify insights provided by, amongst

others, chemical and mechanical engineering (capture and transport) and earth sciences (storage). In order to meet the objective, we formulated the following research questions:

1. What are the technical possibilities for CCS and what is their energetic and economic performance in a complete chain from source to sink?
2. To what extent does the spatial distribution of CO<sub>2</sub> sources and sinks match, and what are the prospects of early opportunities that combine pure CO<sub>2</sub> sources with nearby low-cost storage reservoirs?
3. What are the main factors that may affect the storage capacity? In particular, what are the risks of geological CO<sub>2</sub> storage for man and the environment and what is the current lack of knowledge on these risks?
4. What is the potential role that CCS may play in decarbonising the Dutch energy system over time and how is this translated in terms of CO<sub>2</sub> avoided, costs and infrastructure requirements?

We will first present the results of the individual chapters of this thesis, after which we relate these results to the research questions.

### 3 SUMMARY OF THE RESULTS

1.

All conversion efficiencies are based on LHV. Electricity inputs/outputs in H<sub>2</sub> production are converted into primary energy assuming an electric efficiency of 50%. Plants with CO<sub>2</sub> capture include CO<sub>2</sub> compression to 110 bar. Economic figures are presented in Euro<sub>2003</sub>. Unless otherwise stated, we assumed a discount rate of 10%, an economic lifetime of 20 years for power and industrial plants, capacity factor of 85%, coal and natural gas price of 1.7 and 4.7 €/GJ, respectively.

2.

The original results from Chapter 4, which were presented in US\$ on HHV basis, are converted to € on LHV basis. Note that 1 US\$/GJ<sub>HHV</sub> is roughly 1 €/GJ<sub>LHV</sub>. See also footnote 3.

In *Chapter 2*, we performed a comprehensive inventory and comparison of state-of-the-art and advanced electricity and hydrogen production technologies with CO<sub>2</sub> capture using coal and natural gas, inspired by the large number of studies found in literature. The results of these individual studies cannot be compared due to specific assumptions made. Data from the various studies were therefore standardised in order to calculate conversion efficiencies, energy production costs and CO<sub>2</sub> mitigation costs. This standardisation comprised normalisation of efficiencies with respect to CO<sub>2</sub>/H<sub>2</sub> pressure, normalisation and scaling of capital cost figures, indexation, and the use of uniform economic assumptions<sup>1,2</sup>.

In the short term, (ultra)supercritical PC and NGCC plants equipped with chemical absorption units and IGCC plants with a shift reactor and a physical absorption unit are the most likely candidates for electricity production with strongly reduced CO<sub>2</sub> emissions. Table 1 shows the costs and efficiency figures obtained for different power plants.

In the longer term, both improvements in existing conversion and capture technologies are foreseen as well as new power cycles integrating advanced turbines, fuel cells and novel (high-temperature) separation technologies. Advanced natural gas-fired combined cycle systems can be combined with post-combustion capture, pre-combustion capture using membrane reformers and oxyfuel combustion.

TABLE I. Range in costs and efficiency of central electricity production with CO<sub>2</sub> capture. Values in parentheses are the ones used for further analysis. Mitigation costs have been calculated versus an identical plant without capture

Parameter	State-of-the-art conversion and capture technologies			Advanced conversion and capture technologies		
	PC	IGCC	NGCC	PC	IGCC	NGCC
Net electric efficiency (%)	30-35 (35)	32-40 (32-35)	43-50 (47)	40%	43%	50-55%
Total capital requirement (€/kwe)	1700-2500 (2080)	1500-2200 (1770-2170)	700-1100 (920)	1500	1500-1750	650-900
CO <sub>2</sub> capture efficiency (%)	85-90	85-90	85-90	85-90	85-100	85-100
Electricity production costs (€/kwh)	5.4-6.9 (6.4)	4.7-6.3 (5.8-6.4)	4.9-6.3 (5.6)	5.1	4.5-4.7	4.5-5.3
CO <sub>2</sub> mitigation costs (€/t CO <sub>2</sub> )	27-38 (27)	12-28 (14-23)	32-77 (46)	21	7-14	24-41

Oxyfuel combustion cycles are distinguished on the way oxygen is produced, either using membranes (AZEP), oxygen transport particles (chemical looping combustion) or a conventional cryogenic air separation unit (e.g. Water cycle). Decentralised SOFC-GT hybrids (20 MW<sub>e</sub>) may enable power production with CO<sub>2</sub> capture at a net electric efficiency close to 60% and investment costs of 1500 €/kwe, resulting in COE of 5.8 €/kwh.

The investment costs of advanced technologies, however, are highly uncertain and hence no clear ranking or winning capture strategy can be appointed at this stage. It is important to realise though that NGCC with post-combustion capture still has considerable development potential and may be competitive with advanced pre-combustion and oxyfuel combustion concepts in the coming decade. Various studies comparing advanced power cycles with state-of-the-art power cycles with post-combustion capture pass by the fact that conventional gas turbine and amine scrubbing technology will further improve.

Hydrogen can be produced by means of large-scale steam methane reforming (SMR) or coal gasification (CG) with CO<sub>2</sub> capture from the shifted syngas, resulting in a CO<sub>2</sub> capture efficiency of 85-90%. The conversion efficiency is 73% for SMR and 59-62% for CG, including H<sub>2</sub> compression to 60 bar for pipeline transport. Investment costs are approximately 550 and 840 €/kW<sub>H<sub>2</sub></sub> for 1000 MW<sub>H<sub>2</sub></sub> SMR and CG plants, resulting in production costs of 9.5 and 7 €/GJ, respectively. Advanced autothermal reforming and coal gasification deploying ion transport membranes may reduce production costs to 8.1 and 6.4 €/GJ. Hydrogen production is more attractive for low-cost CO<sub>2</sub> capture than electricity production, as CO<sub>2</sub> needs to be captured anyway, resulting in CO<sub>2</sub> avoidance costs of 5 and 23 €/t CO<sub>2</sub> for CG and SMR, respectively.

Membrane reformers, which are studied in more detail in Chapter 4, enable small-scale hydrogen production at nearly 17 €/GJ with relatively low-cost CO<sub>2</sub> capture.

A similar overview of technologies has been given in the IPCC special report on carbon dioxide capture and storage, although we included more advanced technologies and performed the standardisation procedure, which makes the numbers more consistent. Nevertheless, the numbers that we produced are generally in agreement with the range presented in the IPCC special report. Several values used in our analysis are somewhat more optimistic though, which could be explained by more progressive estimations on the reduction of costs and/or energy use of CO<sub>2</sub> capture. Some studies referred to in the IPCC special report consider rather inefficient base cases. As the relative impact of CCS decreases with improved base-case efficiencies, CCS should be integrated with state-of-the-art energy production technologies.

In *Chapter 3*, the most promising technologies identified in *Chapter 2* are further assessed by means of a chain analysis, incorporating fuel extraction and transport, CO<sub>2</sub> transport and storage and energy carrier transmission, distribution and end-use. The motivation to perform a chain analysis is twofold. First, the technologies studied in *Chapter 2* differ in the fuel type, amount of CO<sub>2</sub> captured and scale, so the impact of the chain elements outside the plant boundary will affect the performance of the technologies differently. Second, a comparison of electricity production with CCS versus hydrogen production with CCS as competing CO<sub>2</sub> reduction options requires an assessment of CO<sub>2</sub> mitigation costs versus the energy carrier that is replaced. Since the distribution and end-use of hydrogen are different with respect to the fuels (gasoline, natural gas) it substitutes, these elements are incorporated.

For electricity chains, the performance is dominated by the impact of CO<sub>2</sub> capture, increasing electricity production costs with 10-40% up to 4.5-6.5 €/kWh. For large-scale power plants, CO<sub>2</sub> transport costs are 1-7 €/t CO<sub>2</sub> for distances up to 200 km (the upper range representing offshore conditions), adding another 0.05-0.3 €/kWh. CO<sub>2</sub> storage in onshore and offshore gas fields or aquifers may cost 1-10 €/t CO<sub>2</sub>, corresponding to 0.05-0.9 €/kWh.

The impact of CCS on large-scale hydrogen production is even smaller; it adds 1-2 €/GJ for transport distances up to 200 km. Long-term production and supply costs range from approximately 8 €/GJ for the minimal infrastructure variant in which hydrogen is delivered to CHP units, up to 20 €/GJ for supply to households. Hydrogen costs for the transport sector are between 14 and 16 €/GJ for advanced large-scale autothermal reforming or coal gasification. Approximately 50% of the costs are attributed to H<sub>2</sub> storage, transmission, distribution, compression and dispensing. The costs of H<sub>2</sub> transmission and distribution could be avoided by decentralised membrane reformers, although decentralised production costs are higher. When CO<sub>2</sub> is captured as well, a relatively expensive CO<sub>2</sub> infrastructure is needed, increasing overall production costs using decentralised membrane

reformers at refuelling stations to approximately 20 €/GJ.

Although the CO<sub>2</sub> price required to induce CCS in hydrogen production is low in comparison to most electricity production options, electricity production with CCS is generally preferred as CO<sub>2</sub> mitigation option. Replacing natural gas or gasoline for hydrogen produced with CCS results in mitigation costs over 100 €/t CO<sub>2</sub> avoided, whereas CO<sub>2</sub> in the power sector could be reduced for costs below 60 €/t CO<sub>2</sub> avoided. Note that the mitigation costs of using hydrogen produced with CCS are very uncertain, as they are strongly dependent on the choice of the reference system and assumptions in fuel prices and fuel cell performance and costs. Therefore, CO<sub>2</sub> mitigation costs presented in the literature are to be interpreted with great care. Mitigation costs are often calculated versus the identical system without CCS, like we did in Chapter 2. This method gives a good indication which technology inherently enables low-cost CO<sub>2</sub> capture. However, a plant with CO<sub>2</sub> capture does not necessarily replace an identical plant without capture. In Chapter 3, we have quantified the impact of choosing different reference systems, both for electricity and hydrogen.

Chapter 4<sup>3</sup> presents the results of a more detailed assessment of membrane reformers (or membrane reactor, MR), a novel technology for the production of hydrogen from natural gas. It promises economic small-scale hydrogen production at refuelling stations and has the potential of inexpensive CO<sub>2</sub> separation, because the retentate (or reformat) stream consist mainly (>90%<sub>mol</sub>) of carbon dioxide and steam, which can be separated easily by condensation. Four configurations of the membrane reactor have been modelled with Aspen<sup>plus</sup> to determine the thermodynamic performance. We included a MR with and without sweep gas (steam), a MR without sweep gas and with cryogenic CO<sub>2</sub> separation, and a MR with sweep gas and integrated CO<sub>2</sub> capture. In the latter, we assumed that a minimum concentration of contaminants in the retentate stream does not pose any problems to pipeline transport. If the gas stream is not pure enough for pipeline transport, a cryogenic separation unit is added to purify the CO<sub>2</sub> stream. The overall energy efficiency of the non-sweep configuration is 78% without H<sub>2</sub> compression and nearly 67% with compression up to 480 bar. The sweep configuration is 0.5%<sub>pt</sub> less efficient. Assuming the retentate stream is pure enough for transport, the overall conversion efficiency of a system including CO<sub>2</sub> separation and compression is reduced by only 1%<sub>pt</sub>. Hydrogen production costs for a 2 MW<sub>H<sub>2</sub></sub> membrane reformer are approximately 14 €/GJ<sub>H<sub>2</sub></sub> including H<sub>2</sub> compression to 480 bar, assuming a production volume of 250 units and a gas price of approximately 4 €/GJ. The costs of CO<sub>2</sub> capture are framed at 12 €/t CO<sub>2</sub> for a configuration in which steam is condensed and CO<sub>2</sub> is compressed. When a cryogenic separation must be added to purify the CO<sub>2</sub> stream, capture costs are beyond 40 €/t CO<sub>2</sub>.

A Monte Carlo analysis was performed to determine the impact of uncertain input parameters. The mean hydrogen costs for the non-

3. The analysis in Chapter 4 was performed using other assumptions on certain input parameters than those adopted in Chapter 2. After we finished the analysis in Chapter 4, we used the efficiency and capital and O&M costs to recalculate hydrogen production costs using the assumptions applied in Chapter 2 (see footnote 1). The performance of membrane reactors in the entire chain, including CO<sub>2</sub> and H<sub>2</sub> infrastructure, has been investigated (relatively simply) in Chapter 4. In Chapter 3, we performed a more detailed analysis of the CO<sub>2</sub> and H<sub>2</sub> infrastructure.

sweep configuration without CO<sub>2</sub> capture is around 14 €/GJ with a standard deviation of 3 €/GJ. Production costs are most sensitive to membrane thickness. Reducing membrane thickness to 5 μm lowers hydrogen cost to under 12 €/GJ. Membrane reactors might become competitive with conventional steam reforming provided that thin membranes can be combined with high stability and a cheap manufacturing method for the membrane tubes. Also larger production volumes (i.e. technological learning) could reduce the levelised hydrogen cost of the membrane reactor.

In order to assess the trade-off between production scale and the construction of a CO<sub>2</sub> and H<sub>2</sub> infrastructure, a complete chain analysis of the membrane reactor versus central steam reforming has been performed. Large-scale H<sub>2</sub> production seems the most economic, because the scale advantages of H<sub>2</sub> production outweigh the additional costs of H<sub>2</sub> infrastructure. Hydrogen production costs including CCS was calculated at 15 €/GJ for a 400 MW<sub>H<sub>2</sub></sub> plant, and nearly 19 €/GJ for a 2 MW<sub>H<sub>2</sub></sub> MR.

In *Chapter 5*, the worldwide potential for low-cost CCS opportunities has been identified. These early opportunities consist of high-purity industrial CO<sub>2</sub> sources closely located to (nearly) depleted hydrocarbon reservoirs or coal seams in which CO<sub>2</sub> could be stored and additional revenues could be generated by means of enhanced oil recovery (EOR) or enhanced coal bed methane recovery (ECBM). A Geographic Information System (GIS) was used to combine worldwide CO<sub>2</sub> point sources with oil fields and coal seams. This resulted in 429 source-oil field and 79 source-coal seam combinations. A Multi-Criteria Analysis (MCA), in which technical and economic criteria are taken into account, was applied to rank the source-reservoir combinations generated by the GIS exercise. We identified potentially promising source-oil field combinations located in the USA, Saudi Arabia, Canada and Mexico. Promising ECBM cases are located in China, Europe, Canada, Japan, Australia and Indonesia.

The most promising opportunities were considered in more detail to select four illustrative cases for further study: two potential EOR projects and two potential ECBM projects. Case 1 consists of a hydrogen plant in Saudi Arabia, which could sequester 0.26 Mt/yr CO<sub>2</sub> in a depleted oil reservoir at practically zero costs. EOR case 2 is a hydrogen plant in California, USA, from which 0.28 Mt CO<sub>2</sub> could be stored annually. Mitigation costs have been estimated at 10-20 €/t CO<sub>2</sub>, depending on the availability of steam for CO<sub>2</sub> regeneration. Case 3 and 4 are ammonia plants from which 0.3 and 0.7 Mt CO<sub>2</sub> may be stored annually at costs around at 5 €/t CO<sub>2</sub> into coal seams in China and Canada, respectively. The cost-effectiveness of the EOR cases is strongly depending on the oil price. By the time this study was performed, relatively conservative oil prices were used; we used a value near 20 \$/bbl.

It was beyond the scope of the study to assess whether the selected reservoirs are indeed suitable for CO<sub>2</sub>-EOR and ECBM, which requires

detailed geological assessments. Consequently, the selected cases should be considered as potential early opportunities. Due to the lack of specific data and the somewhat generic data used for the economic analysis, the mitigation costs should be considered as indicative values that could be somewhat on the low side.

The source database also contains certain gaps. Natural gas processing plants and bioethanol<sup>4</sup> were not included. Start-up years are often missing and reported CO<sub>2</sub> emissions and technologies are not always up-to-date. Older steam reformers producing H<sub>2</sub> are generally being upgraded with a PSA unit, thereby generating a diluted CO<sub>2</sub> stream as the PSA purge gas is combusted in the furnace to provide heat to the reformer. Hydrogen demand at refineries is projected to increase, due to the shift to heavier crudes, the growth in demand and stricter sulphur standards for transport fuels. A part of this demand growth will be supplied by gasification of oil residues, which offers low-cost CCS opportunities par excellence.

4. In the production of ethanol by fermenting sugars, a pure CO<sub>2</sub> stream is generated.

In *Chapter 6*, we move the focus of our research to the risks of underground CO<sub>2</sub> storage. This chapter comprises an overview of health, safety and environmental risks, current gaps in knowledge and research areas that need to be addressed. Insight into these risks is needed to ensure that CCS can be applied as a safe and effective greenhouse mitigation option. Safety is an important issue when transporting and injecting CO<sub>2</sub>, as it could be released by a pipeline failure or well failure/blow-out. Although CO<sub>2</sub> is not explosive or inflammable, prolonged exposure to elevated CO<sub>2</sub> concentrations may cause damage, and eventually death by suffocation to humans and animals. The risks associated with pipeline transport and surface and injection facilities of CO<sub>2</sub> can be quantified and minimised by risk abatement technologies and safety measures as being applied in the oil and gas industry. Less is known on the risks of underground CO<sub>2</sub> storage. CO<sub>2</sub> injection into geological reservoirs may cause CO<sub>2</sub> and CH<sub>4</sub> leakage, seismicity, ground movement, and the displacement of brine. Leakage of CO<sub>2</sub> from the reservoir and seepage into the atmosphere is the main R&D issue at present. It may have both local and global impacts. Locally, CO<sub>2</sub> leaks may cause deterioration of the quality of potable groundwater and negatively affect organisms and ecosystems. From a global perspective, leakage of CO<sub>2</sub> from reservoirs would make CO<sub>2</sub> storage less effective as mitigation option. Leakage through or along wells, faults and fractures are generally considered to be the most important pathways. The potential for leakage depends on the trapping mechanism and well, cap rock and overburden integrity. The type of reservoir in which CO<sub>2</sub> is stored is another important factor for leakage. Hydrocarbon fields are generally well studied and considered to be safe reservoirs for CO<sub>2</sub> storage, since they have held oil, gas and often CO<sub>2</sub> for millions of years without catastrophic leakage. However, these reservoirs are generally penetrated by a large number of abandoned wells, some in bad condition, offering a potential leakage pathway. The major concern for

storage in aquifers is the cap rock integrity, which has generally not been proven. As CO<sub>2</sub> is adsorbed stronger to coal than methane, the risk of CO<sub>2</sub> leakage from coal seams is expected to be low as long as no depressurisation occurs.

Although the mechanisms of leakage are basically understood, quantification of the risks is still a major challenge. Natural and industrial analogues as well as models of CO<sub>2</sub> transport in geological formations seem to suggest that the chance is rather small that a large fraction of CO<sub>2</sub> will leak from a well-selected reservoir and be released into the atmosphere. However, more R&D is required to quantify the processes that control leakage through or along wells, faults and fractures to assess CO<sub>2</sub> leakage rates for various geological conditions. Research topics include the quantification of degradation of cement and casing by (long-term) exposure to CO<sub>2</sub> and the role of fault sealing by mineral precipitation. Although experiments and models suggest that the geochemical impact of CO<sub>2</sub> on the cap rock integrity is minimal or even positive, further work is required in this area. Knowledge on potential leakage rates is required to quantify the effects on human beings, ecosystems and groundwater quality. The effects of elevated CO<sub>2</sub> concentrations on human beings, animals and some biota are understood, but the effects on (marine) ecosystems need further research. Natural analogues are useful in providing a better understanding of the environmental effects of leakage, as well as long-term impacts of the interaction between CO<sub>2</sub> and the reservoir/cap rocks. Industrial analogues such as CO<sub>2</sub>-EOR and acid gas injection offer the opportunity to study the behaviour of CO<sub>2</sub> in geological reservoirs and the frequency and rate of CO<sub>2</sub> leakage along wells, fractures and through cap rocks.

The synthesis of knowledge gathered throughout this thesis is presented in *Chapter 7*. We studied what the role of CCS could be in curbing Dutch CO<sub>2</sub> emissions, which currently approach 180 mt CO<sub>2</sub>/yr. We first analysed the capture potential in different sectors, which indicated that maximally 50 mt CO<sub>2</sub>/yr could be reduced in 2020 by capturing CO<sub>2</sub> from power plants, blast furnaces, boilers and heaters in refineries, steam crackers, reformers and gasifiers. Mitigation costs (excluding transport and storage) vary between 10 €/t CO<sub>2</sub> for pure industrial CO<sub>2</sub> sources up to 100 €/t CO<sub>2</sub> for retrofitting old PC units. Halfway this century, the mitigation potential of CCS is estimated at maximally 80-110 mt CO<sub>2</sub>/yr, of which 60-80 mt CO<sub>2</sub>/yr may be realised below 20 €/t CO<sub>2</sub> (excluding transport and storage). The power sector offers the biggest opportunities for CCS; its mitigation potential has been estimated at 60-84 mt CO<sub>2</sub>/yr by 2050, assuming electricity supply is doubled by 2050. Industrial sources add another 16 mt CO<sub>2</sub>/yr. The development of a market for alternative fuels produced via syngas production with CCS creates an opportunity to decarbonise the transport sector. The reduction potential by 2050 has been estimated very roughly at 10 mt CO<sub>2</sub>/yr for F-T diesel or H<sub>2</sub> production with CCS.

In our analysis, the actual deployment of the capture potential is determined by the emission reduction targets and the geological capacity available for CO<sub>2</sub> storage. The uncertainty in these two decisive factors has been dealt with by performing scenario analyses. Four possible pathways have been sketched to study how CCS (r)evolution may occur in the Netherlands up to 2050. We estimate that avoiding up to 15 mt CO<sub>2</sub>/yr would be feasible in 2020 if some of the larger gas fields that are forecasted to become available the coming decade could be used for CO<sub>2</sub> storage. If larger reductions are required, existing PC units or large industrial boilers and furnaces could be retrofitted with CO<sub>2</sub> capture, which would require that part of the large reservoirs in the North Sea become available for CO<sub>2</sub> storage. Alternatively, we have to rely on clusters of relatively small gas fields, which could add 15 €/t CO<sub>2</sub> for transport and storage.

If the Netherlands focuses on national opportunities and does not consider storage options abroad, 30 mt CO<sub>2</sub>/yr could be avoided by 2050 with relatively small efforts. Provided a good planning is being set up to ensure the domestic storage potential that will become available could be used for CO<sub>2</sub> storage, we may even avoid up to 60 mt CO<sub>2</sub>/yr in 2050. In aggressive climate policies aiming for 50-80% CO<sub>2</sub> reduction by 2050 versus 1990, avoiding another 50 mt CO<sub>2</sub>/yr may be possible provided that nearly all capture opportunities that occur are taken. Storing such large amounts of CO<sub>2</sub> would, however, only be possible if one of the mega structures, either the Groningen gas field, large reservoirs in the Bunter Sandstone formation or the Utsira formation in the North Sea, would become available. Another option could be to store CO<sub>2</sub> in aquifer formations that are not structurally trapped. However, the availability of Groningen is highly uncertain, the availability of the UK reservoirs is depending on the application of CCS in the UK, and exploiting Utsira seems only realistic when an offshore CO<sub>2</sub> infrastructure is constructed. Excluding Groningen, the reservoirs abroad and factors limiting the storage potential, the Netherlands would run out of storage capacity sometime between 2050 and 2075 for storage rates above 80 mt CO<sub>2</sub>/yr. For more modest storage rates of 30 mt CO<sub>2</sub>/yr, we could continue storing up to the year 2100.

Gas fields seem the most appropriate candidates for CO<sub>2</sub> storage given their large and, generally, secure storage capacity. Over 70% of the potential in gas fields is represented by the Groningen gas field, which will not be available prior to 2040 and possibly (far) beyond 2050. The remaining 30% is represented by average and small-sized fields. Many of these reservoirs will be abandoned before 2025 given current production projections, and not all of them will be available for CCS. If it appears that natural gas reservoirs cannot provide sufficient storage potential at the right time, we might have to rely on aquifers and, possibly, coal seams. However, the uncertainty in capacity and the technical/economic feasibility of CO<sub>2</sub> storage do not make these options a very safe bet for large-scale CCS deployment.

#### 4 OVERALL CONCLUSIONS

Although answers to the four research questions can be found in the results presented in the individual chapters, we will summarise the overall findings per research question from a somewhat broader perspective.

**RESEARCH QUESTION 1:** What are the technical possibilities for CCS and what is their energetic and economic performance in a complete chain from source to sink?

The analysis described in Chapters 2 and 3 shows the range in conversion efficiencies and costs of various state-of-the-art technologies to produce electricity and hydrogen with CCS, and what advanced options may achieve in the long run. The electric efficiencies of state-of-the-art-technologies with CO<sub>2</sub> capture are estimated at 32-35% for coal-fired plants and 47% for gas-fired plants. Electricity production costs and CO<sub>2</sub> mitigation costs versus an identical plant without capture are calculated at 5-7 €/kWh and 15-50 €/t CO<sub>2</sub> (excluding transport and storage). Overall, the efficiencies could be improved with 30-40% in the longer term, and mitigation costs could eventually be reduced to 10-40 €/t CO<sub>2</sub>. These figures give an indication of the CO<sub>2</sub> price needed to induce CCS and could be a basis to develop policies to cover the gap between the electricity production costs with CCS and the electricity market price.

One of the major challenges for energy companies in the coming years is to optimise their portfolio in a world with uncertain natural gas and CO<sub>2</sub> prices. Our analysis indicates that for base load operation, NGCC without CO<sub>2</sub> capture would be most economic for gas prices below 5.5-7 €/GJ and CO<sub>2</sub> prices up to 40-55 €/t CO<sub>2</sub>. At higher gas prices and CO<sub>2</sub> prices above 15-25 €/GJ, IGCC with CO<sub>2</sub> capture promises the lowest electricity production costs.

A number of plans have been proposed to construct power plants with integrated CO<sub>2</sub> capture beyond 2010. These projects, located in North America, Europe and Australia, cover a wide range of fuels and capture technologies, illustrating the diversity of CCS routes followed at present. Several utility companies are considering the construction of an IGCC plant with pre-combustion capture. IGCC technology is still relatively immature and capital-intensive in comparison to conventional PC units and has not achieved the high availability factors of PC units yet. Much of the experience with gasification is in the (petro)chemical industry. Consequently, utility companies opting for proven and safe technology prefer PC units with post-combustion capture. Other companies believe in the prospects of oxyfuel combustion. Although there is still improvement potential in post-combustion capture and oxyfuel combustion (solvents with lower regeneration energy, O<sub>2</sub> production using membranes), our techno-economic analysis indicates that pre-combustion capture seems most promising for (bituminous)

coal-fired power plants. The reduction potential in capital costs of IGCC is significant, in contrast to PC plants, where more expensive materials are required for higher steam temperatures to achieve higher conversion efficiencies. In addition, gasification offers the possibility of poly-generation of energy carriers (electricity, hydrogen and synthetic fuels) produced from multiple feedstocks (coal, oil residues and biomass). More insight is needed into the operational impact of using different fuels and varying load factors on the CO<sub>2</sub> capture plant. In addition, various full-scale (multi-fuel) IGCC projects have to be implemented to gain operational experience and prove equal reliability as PC.

Our analysis also incorporated various capture options for NGCC units, but we could not ‘appoint’ the most promising option at this stage, mainly due to uncertainty in capital costs.

The IPCC special report on carbon dioxide capture and storage focuses primarily on large-scale sources, as capturing CO<sub>2</sub> directly from small sources is expected to be more difficult and expensive. Our analysis adds to current knowledge by giving an overview of potential efficiencies and costs of decentralised electricity and hydrogen production with CCS by means of SOFC and MR, respectively. Although energy production costs are indeed higher, they may not be prohibitive in all cases. As SOFC and MR enable low-cost CO<sub>2</sub> capture, electricity or hydrogen could be produced with CCS at reasonable costs, provided that plants can be connected to a nearby CO<sub>2</sub> trunk line or a nearby CO<sub>2</sub> storage reservoir to minimise transport distances. SOFC technology is of particular interest for the Netherlands, which is characterised by a relatively high share of decentralised CHP in industry and the presence of many nearly depleted gas fields that could be used for storage. Decentralised MR would be an interesting technology when hydrogen demand (density) is too low to justify centralised production. Promising regions for the application of MR are relatively densely populated industrialised regions, preferably with a highly developed natural gas infrastructure. As the facilities to enable CCS could be added relatively simply, MR could first be operated in the non-capture mode when there is no CO<sub>2</sub> infrastructure to which it can be connected.

**RESEARCH QUESTION 2:** To what extent does the spatial distribution of CO<sub>2</sub> sources and sinks match, and what are the prospects of early opportunities that combine pure CO<sub>2</sub> sources with nearby low-cost storage reservoirs?

This question has been addressed on different geographic levels. In Chapter 5, we have discussed the prospects of *worldwide* early opportunities for CCS. In Chapter 7, detailed source-sink matching was performed on a *national* level (the Netherlands).

The results in Chapter 5 reveal that there are hundreds of industrial sources that produce nearly pure CO<sub>2</sub>, which could be stored in depleted

oil or coal fields within 100 km distance, with a potential for EOR and ECBM. Nevertheless, the current emission of these sources is only a small fraction of total emissions from industry and the energy sector. Consequently, capturing and storing CO<sub>2</sub> from these sources would provide only a small contribution to large CO<sub>2</sub> emission reductions. Still, the application of early opportunities is important for the further development of CCS, as it yields operational experience and confidence in the technology. In addition, net mitigation costs of many potential EOR cases are likely to be negative with structural oil prices near today's 60 \$/bbl. In contrast to EOR, which is applied commercially, ECBM is still in demonstration phase. Especially China shows a high techno-economic potential for ECBM projects. Many low-cost capture opportunities are expected to arise here in the coming decades with the construction of plants producing chemicals, electricity, hydrogen and low-carbon energy carriers by means of (coal) gasification. This trend towards gasification is likely to occur in other regions of the world as well. Gasification plants could become the new low-cost CCS opportunities of tomorrow. The potential that gasification promises in terms of CO<sub>2</sub> mitigation outnumbers the potential of current early opportunities, as the energy carriers produced from syngas could replace fossil fuels currently applied in the transport, residential and commercial sector.

Zooming into the Netherlands, roughly 2.5 mt pure CO<sub>2</sub> is emitted annually at present. Approximately 1 mt CO<sub>2</sub>/yr, produced by a hydrogen plant and a natural gas processing plant, could be stored in nearly depleted hydrocarbon reservoirs. The remaining CO<sub>2</sub> is produced by ammonia and ethylene oxide plants. The ammonia plants have no access to hydrocarbon reservoirs in vicinity and transport to more remote reservoirs is costly because of the relatively small volumes produced. One ammonia plant is located on top of a coal seam, which might be deployed for ECBM in the longer term. The ethylene oxide plants are generally too small and hence too expensive for CCS. The spatial distribution of current CO<sub>2</sub> sources and storage reservoirs does not match very well. Most sources are located in the Western part of the Netherlands, whereas most gas fields are located offshore or in the Northern part of the country. Transport distances, however, are modest (<300 km). CO<sub>2</sub> could also be transported to some of the larger reservoirs beyond the Dutch continental shelf, because storage costs are lower in comparison to the relatively small reservoirs at the Dutch continental shelf. In scenarios requiring deep CO<sub>2</sub> emission reductions, the Netherlands may have to rely on storage reservoirs abroad. Interesting candidates are the large offshore reservoirs in the British part of the Bunter Sandstone formation or the Utsira formation in the Norwegian part of the continental shelf, assuming the Groningen gas field will not be available for storage.

**RESEARCH QUESTION 3:** What are the main factors that may affect the storage capacity? In particular, what are the risks of geological CO<sub>2</sub> storage for man and the environment and what is the current lack of knowledge on these risks?

Current expectations on the prospects of ccs are based upon theoretical and/or technical estimates of geological storage capacity. For the Netherlands, with its abundance of onshore gas fields situated in a central position of the Northwest European gas market, underground gas storage may become a serious ‘competitor’ with underground CO<sub>2</sub> storage. In the long run, geological reservoirs may also be applied for hydrogen storage. As a first approximation, 100-150 bcm storage volume may be required for UGS in the next two decades to develop the Netherlands into an international gas hub. This corresponds to 13-20% of the storage capacity of all onshore gas fields minus the Groningen gas field. Even more capacity may be required beyond 2025, when many gas fields will be depleted and the reliance on imports will have further increased. Our scenarios indicate that ccs may require between practically 0% up to 80% of the Dutch technical storage potential in onshore gas fields by 2050. The upper boundary represents a scenario in which emission reductions by means of ccs have reached nearly 70 mt CO<sub>2</sub>/yr by 2050, and CO<sub>2</sub> storage is confined to Dutch reservoirs. This kind of considerations (see also Chapter 7 and research question 4) should be accounted for when developing policies and strategies for energy supplies and GHG mitigation such as being developed in the Dutch energy transition process. A vision in which the Netherlands will develop itself as gas and CO<sub>2</sub> hub in Northwest Europe requires optimal tuning of demand and supply of nearly depleted gas fields.

The storage capacity of underground formations may be further reduced by an array of geological and geotechnical considerations, in particular the risk of CO<sub>2</sub> leakage. Leaking reservoirs may have negative effects on man and the environment and reduce the effectiveness of the storage operation. In this way, leakage may eventually undermine the public acceptance of ccs. Although the mechanisms of CO<sub>2</sub> leakage are fairly well understood, hardly any quantitative information on leakage rates was available when our research was performed. With the publication of the IPCC special report on carbon dioxide capture and storage, earth scientists were challenged to provide such figures, which resulted in the following statement:

*‘Observations from engineered and natural analogues as well as models suggest that the fraction retained in appropriately selected and managed geological reservoirs is very likely<sup>5</sup> to exceed 99% over 100 years and is likely<sup>6</sup> to exceed 99% over 1000 years’.*

The term ‘appropriately selected’ suggests that there are reservoirs for which the risk of CO<sub>2</sub> leakage would be higher. It would involve large costs and/or monitoring efforts to exploit them, which make them less suitable or unsuitable for CO<sub>2</sub> storage. This brings us to the crucial

5. Probability of 90-99%.

6. Probability of 66-90%.

question to what extent the technical potential is reduced by risk concerns and other geotechnical considerations. In order to answer this question, basically three challenges are to be surmounted by (geo)scientists:

1. More work is to be done to quantify the processes controlling leakage and determine potential leakage rates. The site-specific nature of risks associated with geological CO<sub>2</sub> storage requires that a variety of pilot and demonstration storage projects is carried out and carefully monitored.
2. We need more robust geological and geotechnical assessments of the storage capacity, preferably on reservoir level. For the Netherlands, an assessment of some of the major reservoirs, in particular the Annerveen field, and their feasibility for CCS (and UGS), would be a good start.
3. A screening tool needs to be developed to classify the storage capacity into suitable and less suitable reservoirs. Although first attempts in that direction are already made, more work is required, also accounting for the insights generated in point 1.

In conclusion, it is recommended to increase our efforts in quantifying the impact of factors that constrain the technical storage potential, i.e. geotechnical features, the implications of risk standards and competition with alternative applications. We need go one step beyond the existing estimations on technical storage capacities and derive realistic and viable storage capacities. Such figures are the key to more solid assessments of the future role of CCS.

**RESEARCH QUESTION 4:** What is the potential role that CCS may play in decarbonising the Dutch energy system over time and how is this translated in terms of CO<sub>2</sub> avoided, costs and infrastructure requirements?

Large-scale deployment of CCS is not due to start before 2012 given the uncertainty in climate policy, the commissioning of new plants, the time needed to demonstrate CCS and to construct CO<sub>2</sub> infrastructure. In the coming years, we could start injecting approximately 1 Mt CO<sub>2</sub>/yr produced in H<sub>2</sub> production and gas processing. This figure could be increased gradually to 12-15 Mt CO<sub>2</sub>/yr avoided in 2020 by capturing CO<sub>2</sub> from new coal-fired power plants put into operation after 2010. CCS penetration may be increased more radically by retrofitting PC units and industrial boilers and furnaces, at higher costs though. The emission reduction potential of CCS in 2050 could potentially reach figures above 100 Mt CO<sub>2</sub>/yr in a baseline scenario where CO<sub>2</sub> emissions grow to 250-330 Mt CO<sub>2</sub>/yr. This is much higher than the CCS potential of 40-60 Mt CO<sub>2</sub>/yr quoted in various Dutch policy studies. We estimate that roughly 60-80 Mt CO<sub>2</sub>/yr could be avoided by that time against mitigation costs between 10 and 40 €/t CO<sub>2</sub>, including transport and storage. It can therefore be concluded that CCS represents a large and rather low-cost

mitigation option to reduce Dutch CO<sub>2</sub> emissions.

However, truly realising this contribution at these costs will require long-term climate policy and a clear and internationally oriented vision on the organisation of CCS deployment in the coming decades. The challenge for the coming years is to find suitable (clusters of) reservoirs that will become available at the right time, have sufficient storage capacity and will not be used for other purposes. As many reservoirs will be abandoned before 2025, a strategy must be developed to assure sufficient storage capacity will be available in the spring of a potential CCS era, sometime between 2020 and 2040. Even though we could potentially store 60-80 Mt CO<sub>2</sub>/yr for many decades given the domestic technical storage potential, also reservoirs beyond the Dutch continental shelf may have to be deployed. This would require an internationally coordinated action to plan the CO<sub>2</sub> streams and the construction of an offshore CO<sub>2</sub> infrastructure. In addition, neighboring countries such as Belgium and Germany have a lack of storage capacity and may look for reservoirs located at the Dutch shore and the North Sea. The vision in which the Netherlands develop itself as CO<sub>2</sub> hub hinges upon the post-Kyoto emission reduction targets set for the EU, developments in the energy and industrial sector and the demand for UGS in Northwest Europe. In conclusion, the Netherlands needs to extend its view beyond national borders and think within a Northwest European context.

If, on the other hand, the EU and the Netherlands postpone climate policy, overall costs may increase as the opportunities for domestic low-cost storage, CO<sub>2</sub>-EOR at the North Sea and infrastructure reuse are or could be missed. Moreover, it can be argued whether the cost level of 10-40 €/t CO<sub>2</sub> can be achieved without CCS capacity being installed the coming decades to gain operational experience and enable cost reductions by means of technological learning.

As distances between sources and sinks are generally low in the Netherlands, transport costs do not add strongly to mitigation costs, provided that the volumes involved are not too low (<1 Mt/yr). Depending on timing, required emission reduction and location of sources and sinks, either direct source-sink pipelines or large trunk lines of similar dimensions as in current natural gas transmission, connecting (clusters of) various sources and sinks, need to be constructed. Upfront investments vary between 10<sup>7</sup> and 10<sup>9</sup> Euro for small local pipelines and long-distance offshore trunk lines, respectively. Government support or participation may be indispensable in financing large (trans-boundary) trunk lines.

## 5 RECOMMENDATIONS FOR FURTHER RESEARCH

Throughout this thesis we came across different topics that may be worth investigating in more detail, of which some are summarised below:

- Opportunities, costs and technical implications of CO<sub>2</sub> capture in steel production.
- Potential of synfuel production by means of coal and biomass gasification in combination with CCS.
- Performance, costs and flexibility of multi-fuel poly-generation IGCC units with CO<sub>2</sub> capture.
- Prospects of decentralised technologies that promise low costs CO<sub>2</sub> capture such as SOFC, AZEP and ZEPP, in particular for industrial CHP applications.
- The potential role of central and decentralised hydrogen production from fossil fuels in the transition towards a hydrogen economy.
- Potential cost reductions of CCS by means of technological learning (both capital and operational costs).
- Specific costs of CO<sub>2</sub> trunk lines for a number of trajectories (Eemshaven-Annerven, Rijnmond-Groningen, Rijnmond-southern North Sea (UK) via the PKL quadrants, Rijnmond/Eemshaven-Utsira via the British and/or Norwegian oil fields).
- Feasibility of reusing the gas infrastructure, also as part of a larger CO<sub>2</sub> network to connect sinks in the British and Norwegian part of the continental shelf.
- More detailed mapping and geological characterisation of aquifers and their traps, especially for the Dutch part of the continental shelf. A pilot project may increase our insights in the injectivity, storage efficiency and sealing capacity of aquifer traps.
- Feasibility of different injection strategies in the Groningen gas field prior to depletion to judge what its enormous storage capacity could mean for the future of CCS.

Finally, we plea to perform a detailed feasibility study to evaluate the possibilities of CCS for the Rijnmond region, where many industrial activities are centralised, various new coal-fired power plants are being planned, several gas and oil fields are located and the storage potential offshore is within reach. Topics to be addressed are co-siting, the potential to utilise low temperature steam available in industry for CO<sub>2</sub> regeneration, the benefits of a large trunk line to offshore reservoir(s) and detailed geological characterisation of potential reservoirs for CO<sub>2</sub> storage.



## SAMENVATTING EN CONCLUSIES

### I INLEIDING

Het besef van noodzaak tot ingrijpende reducties in antropogene emissies van koolstofdioxide ( $\text{CO}_2$ ), voortkomend uit de zorgen over de effecten van het door de mens veroorzaakte broeikas-effect, heeft de laatste jaren geleid tot een groeiende interesse in  $\text{CO}_2$  afvang en opslag. Deze technologie wordt afgekort met ccs (Carbon Capture and Storage) en wordt in het Nederlands ook wel aangeduid als schoon fossiel. Het omvat de afvang van  $\text{CO}_2$  dat wordt geproduceerd door grote puntbronnen, waarna de  $\text{CO}_2$  wordt getransporteerd en opgeslagen in geologische reservoirs, de oceaan, of vaste mineralen, met als doel om de emissie van  $\text{CO}_2$  naar de atmosfeer te voorkomen. ccs wordt doorgaans beschouwd als een tijdelijke oplossing op de weg naar een duurzame energie voorziening. Het stelt ons in staat om het onvermijdelijke fossiele brandstofverbruik voort te zetten en tegelijkertijd de  $\text{CO}_2$  emissies die hierdoor ontstaan te reduceren. De toepassing van ccs zal gepaard gaan met een grote rol voor kolen, waarvan de wereldwijde reserves groter en meer heterogeen verdeeld zijn in vergelijking met olie en aardgas. Door de injectie van  $\text{CO}_2$  in olievelden kan extra olie worden gewonnen.  $\text{CO}_2$  injectie in lege gasvelden of kolenvelden zou mogelijk tot extra methaanproductie kunnen leiden. Op die manier kan ccs twee prioriteiten in energiebeleid verenigen, namelijk het reduceren van  $\text{CO}_2$  emissies alsmede het verminderen van de afhankelijkheid van olie en aardgas uit instabiele regio's. Daarnaast kan ccs een brugfunctie vervullen in het ontwikkelen van een waterstofinfrastructuur, omdat waterstof geproduceerd kan worden uit fossiele brandstoffen tegen relatief lage kosten in vergelijking met waterstof uit hernieuwbare elektriciteit.

Natuurlijk zijn er ook een aantal nadelen verbonden aan de toepassing van ccs. Een veelgehoord tegenargument is het feit dat ccs een zogenaamde 'end-of-pipe' technologie is, omdat de productie van  $\text{CO}_2$  niet wordt voorkomen. Daarom wordt ccs beschouwd als een niet-structurele oplossing voor de klimaatproblemen veroorzaakt door fossiel energieverbruik. Ook zijn het energieverbruik en de kosten van ccs significant. Omdat er meer primaire energie nodig is om dezelfde output te genereren, leidt ccs tot een grotere afhankelijkheid van fossiele brandstoffen. Tenslotte moet het afgevangen  $\text{CO}_2$  tenminste honderden en mogelijk duizenden jaren worden opgeslagen, wat bepaalde risico's met zich meebrengt, waaronder lekkage van  $\text{CO}_2$  uit reservoirs.

Op dit moment worden er slechts een klein aantal commerciële, grootschalige ccs projecten uitgevoerd. Het merendeel van de projecten bestaat uit gaswinning waarbij de  $\text{CO}_2$  in het aardgas wordt afgescheiden

en opgeslagen in geologische formaties. De technologieën die nodig zijn voor CCS worden echter al toegepast; de olie en chemische industrie heeft vele jaren ervaring met CO<sub>2</sub> scheiding, behandeling, transport en injectie in ondergrondse reservoirs. De uitdaging voor de komende jaren is om de verschillende CCS componenten op te schalen en te integreren, daarbij gebruik makende van de opgedane industriële ervaring.

Er zijn echter nog een aantal wetenschappelijke uitdagingen die ons wachten, waarvan er een aantal hieronder zijn beschreven:

1. *Rendementen en kosten van CO<sub>2</sub> afvang.* Er zijn vele opties om CO<sub>2</sub> af te vangen. De meeste zijn geëvalueerd op basis van hun thermodynamische rendement en kosten. De resultaten van deze studies moeten gestandaardiseerd worden om de verschillende opties op een consistente wijze te kunnen vergelijken. Daarnaast moeten de resultaten van de berekeningen geverifieerd worden door middel van grootschalige demonstratieprojecten. Commerciële toepassing is bovendien cruciaal in het reduceren van de kosten, naast R&D in innovatieve technologieën die op termijn efficiënte en goedkope CO<sub>2</sub> afvang mogelijk maken.
2. *Matchen van CO<sub>2</sub> bronnen en reservoirs.* De komende jaren zijn meer gedetailleerde regionale analyses nodig om CO<sub>2</sub> bronnen en potentiële opslagreservoirs op elkaar af te stemmen (te 'matchen'). Daarvoor zijn modellen nodig die het mogelijk maken om de ontwikkelingen van zowel bronnen als reservoirs in tijd te analyseren, en tevens te rekenen aan meervoudige bron-reservoir combinaties en CO<sub>2</sub> netwerken.
3. *Capaciteit en risico's van ondergrondse CO<sub>2</sub> opslag.* Er is behoefte aan meer gedetailleerde schattingen van mondiale en regionale opslagcapaciteiten en studies naar de risico's van ondergrondse CO<sub>2</sub> opslag. Op die manier kan de geschiktheid van reservoirs voor CO<sub>2</sub> opslag worden vastgesteld en kunnen de opslagcapaciteiten beter op waarde worden ingeschat. Ook de vraag naar en het potentieel van alternatieve toepassingen voor geologische reservoirs, waaronder ondergrondse opslag van aardgas, moeten worden bepaald. Indien de risico's, alternatieve toepassingen en economische en wettelijke barrières worden meegenomen, kunnen uiteindelijk meer realistische schattingen van opslagcapaciteiten worden gemaakt.
4. *Rol van CCS in een strategie om broeikasgasemissies te reduceren.* Nu we op de drempel van daadwerkelijke toepassing van CCS staan is het zaak om strategieën te ontwikkelen voor grootschalige CCS implementatie als integraal onderdeel van een transitie naar een duurzame energievoorziening. Hiertoe zijn inzichten nodig in de manier waarop CCS kan evolueren en wat haar rol kan zijn in samenhang met andere opties voor CO<sub>2</sub> emissiereductie.

## 2 DOELSTELLINGEN VAN DIT PROEFSCHRIFT

Het doel van dit proefschrift is het analyseren van de technische mogelijkheden, kosten en risico's van CCS systemen om aldus opties te vergelijken, veelbelovende technologieën te identificeren en een beeld te krijgen van de rol die CCS de komende decaden in Nederland kan vervullen in de reductie van CO<sub>2</sub> emissies. De perspectieven voor CCS in Nederland zijn veelbelovend, gezien de aanwezigheid van grote CO<sub>2</sub> puntbronnen en talloze geologische reservoirs. Bovendien is er jarenlange industriële ervaring met de transmissie, distributie en behandeling van gassen, waaronder aardgas, welke van groot belang kan zijn voor het ontwikkelen van een CO<sub>2</sub> infrastructuur.

In dit proefschrift wordt louter CO<sub>2</sub> opslag in geologische reservoirs beschouwd. De insteek is om de analyses vanuit een systeemperspectief uit te voeren. Dit betekent dat inzichten van verschillende disciplines, in het bijzonder chemische technologie en aardwetenschappen, worden gecombineerd. Om bovenstaande doelstellingen te beantwoorden, zijn de volgende onderzoeksvragen geformuleerd:

1. Wat zijn de technische mogelijkheden voor CCS en wat zijn de energetische rendementen en kosten van deze opties in een keten van bron tot reservoir?
2. In welke mate komt de ruimtelijke verdeling van CO<sub>2</sub> bronnen en reservoirs overeen, en wat zijn de mogelijkheden om pure CO<sub>2</sub> bronnen te combineren met nabij gelegen reservoirs die goedkope CO<sub>2</sub> opslag mogelijk maken?
3. Wat zijn de voornaamste factoren die de opslagcapaciteit beïnvloeden? Meer in het bijzonder, wat zijn de risico's van ondergrondse CO<sub>2</sub> opslag voor de mens en het milieu en wat zijn de huidige hiaten in kennis van deze risico's?
4. Wat is de potentiële rol die CCS zou kunnen vervullen in het 'ontkolen' van de Nederlandse energievoorziening in de tijd en wat betekent dit in termen van vermeden CO<sub>2</sub> emissies, kosten en benodigde infrastructuur?

Eerst zullen de resultaten van de individuele hoofdstukken van dit proefschrift worden gepresenteerd, waarna we in algemene zin antwoord geven op de hierboven geformuleerde onderzoeksvragen.

## 3 SAMENVATTING VAN DE RESULTATEN

*Hoofdstuk 2* beschrijft de resultaten van een uitgebreide inventarisatie en vergelijking van huidige en toekomstige technologieën voor elektriciteits- en waterstofproductie met CO<sub>2</sub> afvang op basis van (bitumineuze) kolen en aardgas. De oorsprong van deze analyse ligt in het feit dat de resultaten van de verschillende studies in de literatuur niet vergeleken kunnen worden vanwege specifieke aannames die per studie verschillen. Om een vergelijking mogelijk te maken zijn de data

uit de verschillende studies gestandaardiseerd zodat de conversierendementen, energieproductiekosten en CO<sub>2</sub> mitigatiekosten kunnen worden berekend. Deze standaardisatie omvat het normaliseren van rendementen naar CO<sub>2</sub>/H<sub>2</sub> druk, het normaliseren en ‘schalen’ van kapitaalkosten, inflatiecorrectie, en het gebruik van uniforme economische veronderstellingen<sup>1,2</sup>.

Op de korte termijn kan elektriciteit worden gegenereerd met sterk verminderde CO<sub>2</sub> emissies door (ultra)superkritische poederkoolcentrales (PC) en aardgas gestookte stoom- en gasturbine (STEG) eenheden te integreren met chemische absorptie om CO<sub>2</sub> uit de rookgassen te verwijderen. Een veelbelovend alternatief is een centrale op basis van kolenvergassing met een stoom- en gasturbine (KV-STEG) in combinatie met een shift reactor en fysische absorptie van de gevormde CO<sub>2</sub>. Tabel I toont de kosten en rendementen voor de verschillende technologieën.

TABEL I. Kosten en rendementen van centrale elektriciteitsproductie met CO<sub>2</sub> afvang. De waarden tussen haakjes zijn gebruikt voor de verdere analyse. Mitigatiekosten zijn berekend versus een identieke conversietechnologie zonder CO<sub>2</sub> afvang

Parameter	Huidige conversie en afvang technologieën			Geavanceerde conversie en afvang technologieën		
	PC	KV-STEG	STEG	PC	KV-STEG	STEG
Netto elektrisch rendement (%)	30-35 (35)	32-40 (32-35)	43-50 (47)	40%	43%	50-55%
Totale kapitaalkosten €/kwe	1700-2500 (2080)	1500-2200 (1770-2170)	700-1100 (920)	1500	1500-1750	650-900
CO <sub>2</sub> afvang efficiëntie (%)	85-90	85-90	85-90	85-90	85-100	85-100
Elektriciteitsproductie kosten (€/kwh)	5,4-6,9 (6,4)	4,7-6,3 (5,8-6,4)	4,9-6,3 (5,6)	5,1	4,5-4,7	4,5-5,3
CO <sub>2</sub> mitigatiekosten (€/t CO <sub>2</sub> )	27-38 (27)	12-28 (14-23)	32-77 (46)	21	7-14	24-41

Op de lange termijn worden zowel verbeteringen in huidige technologieën voorzien evenals nieuwe concepten waarin geavanceerde gasturbines, brandstofcellen en nieuwe (hoge temperatuur) scheidingstechnieken worden geïntegreerd. Geavanceerde STEG eenheden kunnen worden gecombineerd met systemen waarbij CO<sub>2</sub> wordt afgevangen na de verbranding (‘post-combustion’), voor de verbranding (‘pre-combustion’) en door middel van zuurstofrijke verbranding (‘oxyfuel combustion’). Systemen op basis van ‘oxyfuel combustion’ worden onderscheiden op basis van de manier waarop zuurstof wordt geproduceerd. Dit kan met behulp van membranen, zuurstof transporterende deeltjes of conventionele cryogene distillatie. Decentrale brandstofcel-gasturbine hybrides (SOFC-GT) zouden in de toekomst elektriciteit kunnen produceren met CO<sub>2</sub> afvang tegen 5,8 €/t kwh uitgaande van een netto elektrisch rendement van circa 60% en kapitaalkosten rond de 1500 €/kwe.

1. Alle conversierendementen zijn gebaseerd op de onderste of netto verbrandingswaarde (LHV). Elektriciteit input en output bij waterstofproductie worden omgezet in primaire energie op basis van een elektrisch rendement van 50%. Alle centrales met CO<sub>2</sub> afvang omvatten CO<sub>2</sub> compressie tot 110 bar. Kosten en prijzen worden gepresenteerd in Euro<sub>2003</sub>. Tenzij anders vermeld rekenen we met de volgende aannames: een disconteringsvoet van 10%, een economische levensduur van 20 jaar, een capaciteitsfactor van 85%, kolen en aardgas prijzen van 1,7 en 4,7 €/Gj.

2. De originele resultaten uit hoofdstuk 4, die zijn gepresenteerd in Dollars en op basis van bovenste calorische waarde (HHV), zijn geconverteerd naar Euro en LHV. 1 US\$/Gj<sub>HHV</sub> is ongeveer 1 €/Gj<sub>LHV</sub>. Zie ook voetnoot 3.

De kapitaalkosten van geavanceerde technologieën zijn echter hoogst onzeker, waardoor het niet mogelijk is om een winnende technologie aan te wijzen. Het is echter belangrijk te beseffen dat een STEG centrale met CO<sub>2</sub> afvang uit de rookgassen nog een aanzienlijk verbeterpotentieel heeft en mogelijk competitief kan zijn met moderne 'pre-combustion' en 'oxyfuel combustion' systemen. Verschillende studies in de literatuur vergelijken geavanceerde, toekomstige technieken met huidige technieken voor elektriciteitsproductie en CO<sub>2</sub> afvang uit rookgassen. Daarmee wordt voorbij gegaan aan het feit dat conventionele gasturbines en chemische absorptietechnieken zich verder ontwikkelen in de tijd.

Waterstof kan worden geproduceerd door stoom reforming (SMR) of kolenvergassing (KV), waarna de CO in het synthesegas kan worden omgezet in CO<sub>2</sub> en H<sub>2</sub>, en de CO<sub>2</sub> kan worden afgevangen. Het omzettingsrendement is ongeveer 73% voor SMR en 59-62% voor KV, inclusief H<sub>2</sub> compressie tot 60 bar voor transport. Kapitaalkosten zijn geschat op circa 550 en 840 €/kW<sub>H<sub>2</sub></sub> voor een 1000 MW<sub>H<sub>2</sub></sub> SMR en KV, wat resulteert in productiekosten van respectievelijk 9,5 en 7 €/GJ waterstof. Geavanceerde 'autothermal reforming' en kolenvergassing waarbij gebruik wordt gemaakt van ion transport membranen voor zuurstofproductie zouden productiekosten kunnen verlagen tot respectievelijk 8,1 en 6,4 €/GJ. De additionele kosten van CO<sub>2</sub> afvang bij waterstofproductie zijn lager dan bij elektriciteitsproductie, omdat CO<sub>2</sub> sowieso moet worden verwijderd om waterstof te produceren. Dit leidt tot kosten van 5 en 23 € per ton vermeden CO<sub>2</sub> voor KV en SMR. Membraanreformers, welke in meer detail zijn bestudeerd in hoofdstuk 4, kunnen worden gebruikt voor decentrale waterstofproductie tegen kosten van circa 17 €/GJ. Bovendien kan tegen relatief lage kosten CO<sub>2</sub> worden afgevangen.

Een vergelijkbaar overzicht van technologieën is gepresenteerd in het 'IPCC special report on carbon dioxide capture and storage', hoewel het overzicht in dit proefschrift meer geavanceerde technieken bevat. Bovendien hebben wij een standaardisatie procedure toegepast, waardoor de berekende waarden beter vergelijkbaar zijn. Desalniettemin zijn onze resultaten in overeenstemming met de resultaten gepresenteerd in het IPCC special report. Sommige waarden die zijn gebruikt in de analyses zijn wat aan de optimistische kant, wat kan worden verklaard door het feit dat de meest progressieve technologieën met de laagste kosten en/of energieverbruik voor CO<sub>2</sub> afvang zijn beschouwd. Sommige studies die in het IPCC special report worden aangehaald beschouwen geen state-of-the-art referentie technologieën. Aangezien de kosten en het energieverbruik van CCS afnemen met een stijgend energetisch rendement van de centrale dient CCS bij voorkeur te worden toegepast in combinatie met de meest efficiënte conversietechnologieën.

In *hoofdstuk 3* worden veelbelovende technologieën die in hoofdstuk 2 zijn geïdentificeerd verder onderzocht in een ketenanalyse. Hierin worden de winning en transport van primaire energiedragers, CO<sub>2</sub> transport en opslag, en de transmissie, distributie en eindgebruik van secundaire energiedragers meegenomen. Er zijn een aantal redenen om een ketenanalyse uit te voeren. Ten eerste verschillen de technologieën die zijn geanalyseerd in hoofdstuk 2 in capaciteit en hoeveelheid CO<sub>2</sub> die wordt afgevangen, waardoor ook de kosten van de elementen buiten de centrale nogal kunnen verschillen. Ten tweede vereist een vergelijking van elektriciteitsproductie met CCS versus waterstofproductie met CCS als concurrerende opties een beoordeling van de CO<sub>2</sub> mitigatiekosten ten opzichte van de energiedrager die wordt vervangen. Aangezien de distributie en het eindgebruik van waterstof verschillen in vergelijking met benzine en aardgas dienen deze ketenelementen te worden beschouwd in de analyse.

Voor elektriciteitsketens worden de kosten en rendementen met name beïnvloed door het afvangen van CO<sub>2</sub>; de productiekosten stijgen hierdoor met 10-40% tot 4,5-6,5 €/kWh. Voor grootschalige elektriciteitscentrales bedragen de kosten voor CO<sub>2</sub> transport 1-7 € per ton CO<sub>2</sub> voor afstanden tot 200 km, waardoor productiekosten toenemen met 0,05-0,3 €/kWh. CO<sub>2</sub> opslag in gasvelden of aquifers kost ongeveer 1-10 € per ton CO<sub>2</sub> wat overeenkomt met 0,05-0,9 €/kWh. Indien CCS wordt toegepast in combinatie met grootschalige waterstofproductie stijgen de kosten met 1-2 €/GJ voor transportafstanden tot 200 km. De lange-termijn kosten voor de productie en distributie van waterstof variëren van 8 €/GJ voor een configuratie waarin waterstof met beperkte infrastructuur wordt geleverd aan decentrale WKK-eenheden tot 20 €/GJ voor levering aan huishoudens. Waterstofkosten voor de transportsector worden geschat op 14-16 €/GJ uitgaande van geavanceerde, grootschalige reformers of kolenvergassers. Grofweg de helft van deze kosten kan worden toegeschreven aan de opslag, transmissie, distributie en compressie van waterstof, en de brandstofpompen. Door waterstof te produceren met decentrale membraanreformers worden de kosten van waterstoftransmissie en -distributie vermeden. Echter, de productiekosten van decentrale eenheden zijn hoger. Wanneer ook de CO<sub>2</sub> wordt afgevangen is bovendien een relatief dure CO<sub>2</sub> infrastructuur nodig, waardoor de totale kosten van waterstof geproduceerd met decentrale membraanreformers op ongeveer 20 €/GJ worden geschat. Ofschoon de CO<sub>2</sub> prijs die nodig is om CCS te implementeren in waterstofproductie uit fossiele energiedragers laag is in vergelijking met elektriciteitsproductie, verdient elektriciteitsproductie met CCS de voorkeur als CO<sub>2</sub> mitigatieoptie. De kosten om aardgas of benzine te vervangen voor waterstof geproduceerd uit fossiele energiedragers met CCS liggen boven de 100 € per ton vermeden CO<sub>2</sub>. Daarentegen kan CO<sub>2</sub> in de elektriciteitssector worden gereduceerd door CCS tegen kosten onder de 60 € per ton vermeden CO<sub>2</sub>. De mitigatiekosten van waterstoftoepassing zijn echter erg onzeker, omdat deze voornamelijk

worden bepaald door de keuze van het referentiesysteem en de aannames in brandstofprijzen en het rendement en de kosten van brandstofcellen. Daarom dienen CO<sub>2</sub> mitigatiekosten die in de literatuur worden gepresenteerd kritisch te worden beschouwd. Deze kosten worden meestal berekend ten opzichte van het identieke systeem zonder CCS, zoals wij hebben toegepast in hoofdstuk 2. Deze methode geeft een goede indicatie welke technologie inherent goedkope CO<sub>2</sub> afvang mogelijk maakt. Een centrale met CO<sub>2</sub> afvang vervangt echter niet altijd een centrale gebaseerd op identieke technologie zonder CO<sub>2</sub> afvang. In dit hoofdstuk hebben wij de invloed van de keuze van verschillende referentiesystemen gekwantificeerd voor zowel elektriciteit als waterstof.

3. De analyse in hoofdstuk 4 is uitgevoerd met andere aannames voor bepaalde input parameters dan in hoofdstuk 2. Nadat we de analyses in hoofdstuk 4 hadden afgerond, hebben we de rendementen, kapitaal en onderhoudskosten gebruikt om de waterstofproductiekosten te herberekenen met de aannames uit hoofdstuk 2 (zie voetnoot 1). De ketenanalyse zoals die in hoofdstuk 4 is uitgevoerd is vrij eenvoudig wat betreft de CO<sub>2</sub> en H<sub>2</sub> infrastructuur. Daarom hebben we in hoofdstuk 3 een meer gedetailleerde analyse van de CO<sub>2</sub> en H<sub>2</sub> infrastructuur gedaan.

*Hoofdstuk 4*<sup>3</sup> presenteert de resultaten van een gedetailleerde analyse van membraanreformers (MR), een nieuwe technologie om waterstof te produceren uit aardgas. Deze technologie kan op kleine schaal worden toegepast, bijvoorbeeld voor tankstations. Omdat het reformaat, de gasstroom die uit de reformer komt nadat waterstof is afgescheiden, voornamelijk uit koolstofdioxide en stoom bestaat, kan CO<sub>2</sub> in principe tegen lage kosten worden afgevangen door stoom te condenseren. In onze analyse zijn vier MR configuraties gemodelleerd met Aspen<sup>plus</sup>: een MR met en zonder 'sweep-gas' (stoom), een MR met sweep-gas en cryogene CO<sub>2</sub> scheiding en een MR zonder sweep-gas en geïntegreerde CO<sub>2</sub> afvang. In de laatste configuratie nemen we aan dat de overige gassen aanwezig in het reformaat geen problemen veroorzaken voor transport middels een pijpleiding. Indien het reformaat niet puur genoeg is voor transport (nadat stoom is verwijderd) is een cryogene installatie nodig om de CO<sub>2</sub> stroom te zuiveren. Het energetische rendement van de configuratie zonder sweep-gas en CO<sub>2</sub> afvang is 78% zonder H<sub>2</sub> compressie en ongeveer 67% inclusief compressie tot 480 bar. De configuratie met sweep-gas is 0,5%<sub>pt</sub> minder efficiënt. Het rendement daalt met slechts 1%<sub>pt</sub> in de configuratie waarin stoom wordt gecondenseerd en de CO<sub>2</sub> wordt gecomprimeerd voor transport. De kosten van waterstofproductie voor een 2 MW<sub>H<sub>2</sub></sub> membraanreformer zijn ongeveer 14 €/GJ<sub>H<sub>2</sub></sub> inclusief H<sub>2</sub> compressie tot 480 bar, uitgaande van een MR productievolume van 250 eenheden en een aardgasprijs van circa 4 €/GJ. De kosten van CO<sub>2</sub> afvang en compressie zijn ingeschat op 12 € per ton CO<sub>2</sub>. Als een cryogene scheiding nodig is om de CO<sub>2</sub> stroom te zuiveren stijgen de afvangkosten tot boven de 40 € per ton CO<sub>2</sub>. Een Monte Carlo analyse is toegepast om de invloed van onzekere input parameters te kwantificeren. De gemiddelde waterstofkosten voor de configuratie zonder sweep-gas en CO<sub>2</sub> afvang zijn ongeveer 14 €/GJ met een standaarddeviatie van 3 €/GJ. De productiekosten worden het sterkst beïnvloed door de dikte van de membranen. Wanneer membranen kunnen worden geproduceerd van 5 µm kunnen de waterstofkosten mogelijk worden gereduceerd tot minder dan 12 €/GJ. Membraanreformers zouden concurrerend kunnen worden met conventionele stoom reformers als dunne en stabiele membranen

op een goedkope wijze kunnen worden geproduceerd. Verdere kostenreducties zijn mogelijk door het productievolume te verhogen en door technologisch leren.

Om de 'trade-off' tussen de productiecapaciteit en de kosten die de constructie van een CO<sub>2</sub> and H<sub>2</sub> infrastructuur met zich meebrengen te onderzoeken hebben we een ketenanalyse uitgevoerd voor zowel een decentrale MR en een grootschalige SMR. Hieruit blijkt dat grootschalige reformers tot de laagste waterstofkosten leiden, omdat de schaalvoordelen van waterstofproductie afwegen tegen de additionele kosten van een H<sub>2</sub> infrastructuur. Waterstofproductie en -distributie kosten inclusief CCS worden geschat op 15 €/GJ voor een 400 MW<sub>H<sub>2</sub></sub> SMR, en 19 €/GJ voor een 2 MW<sub>H<sub>2</sub></sub> MR.

*Hoofdstuk 5* gaat in op het wereldwijde potentieel van CCS mogelijkheden die tegen relatief lage kosten kunnen worden geïmplementeerd. Deze mogelijkheden bestaan uit industriële bronnen waarbij pure CO<sub>2</sub> vrijkomt dat tegen lage kosten kan worden afgevangen, getransporteerd en opgeslagen in nabij gelegen olievelden of kolenlagen. CO<sub>2</sub> injectie in olievelden en kolenlagen kan leiden tot extra olie of methaan productie. Deze processen worden aangeduid als 'enhanced oil recovery' (EOR) en 'enhanced coal bed methane recovery' (ECBM). In deze analyse is een Geografisch Informatie Systeem (GIS) gebruikt om de mondiale CO<sub>2</sub> puntbronnen te combineren met dit type reservoirs. Dit resulteerde in 429 potentiële combinaties van puntbronnen en olievelden en 79 combinaties van puntbronnen en kolenlagen. Daarna is een Multicriteria Analyse (MCA) toegepast om de verschillende combinaties te ordenen. In deze analyse zijn zowel technische als economische criteria meegenomen. Veelbelovende combinaties van pure CO<sub>2</sub> bronnen en olie reservoirs die zijn geïdentificeerd liggen in the Verenigde Staten, Saudi-Arabië, Canada en Mexico. Mogelijk veelbelovende combinaties van pure CO<sub>2</sub> bronnen en kolenlagen liggen in China, Europa, Canada, Japan, Australië and Indonesië. Uit deze combinaties zijn vier veelbelovende cases geselecteerd die verder zijn onderzocht. De eerste case studie bestaat uit een waterstoffabriek in Saudi-Arabië die jaarlijks ongeveer 0,26 mt CO<sub>2</sub> produceert dat kan worden opgeslagen in een olieveld. In dit geval compenseren de extra olie-inkomsten door CO<sub>2</sub> injectie de kosten voor transport en opslag. De tweede case studie bestaat eveneens uit een waterstoffabriek, ditmaal in Californië, waar jaarlijks ongeveer 0,28 mt CO<sub>2</sub> beschikbaar is voor opslag. De mitigatiekosten zijn geschat op 10-20 € per ton vermeden CO<sub>2</sub>, afhankelijk van de beschikbaarheid van stoom benodigd voor CO<sub>2</sub> regeneratie. Case studie 3 en 4 zijn ammoniak fabrieken in China and Canada waar jaarlijks ongeveer 0,3 en 0,7 mt CO<sub>2</sub> kan worden opgeslagen in kolenlagen tegen kosten van circa 5 € per ton CO<sub>2</sub>. De kosteneffectiviteit van de eerste twee 'EOR' cases zijn afhankelijk van de gebruikte oliepijzen. In de tijd dat deze studie werd uitgevoerd, werd met relatief lage oliepijzen gerekend; de resultaten zijn gebaseerd op een prijs van 20 \$ per vat.

In onze analyse is niet onderzocht of de geselecteerde reservoirs ook daadwerkelijk geschikt zijn voor CO<sub>2</sub> injectie en EOR of ECBM. Hiervoor zijn gedetailleerde geologische studies nodig. De geselecteerde cases dienen daarom als mogelijk interessante projecten worden beschouwd. Vanwege het gebrek aan specifieke data en de generieke informatie die is gebruikt om de economische analyses uit te voeren zijn de berekende mitigatiekosten slechts indicatieve schattingen, die mogelijk aan de lage kant zijn.

De database met daarin de wereldwijde CO<sub>2</sub> bronnen bevat ook een aantal hiaten. Ten eerste zijn de installaties waar aardgas van CO<sub>2</sub> wordt ontdaan en bio-ethanol<sup>4</sup> fabrieken, die eveneens een pure CO<sub>2</sub> stroom genereren, niet meegenomen. Ten tweede is het jaar waarin eenheden in bedrijf zijn genomen vaak niet vermeld en zijn de gerapporteerde CO<sub>2</sub> emissies en toegepaste technologieën niet altijd up-to-date. In veel oudere reformers die waterstof produceren wordt de chemische absorptie eenheid vervangen voor een 'pressure swing adsorber' (PSA). Aangezien de CO<sub>2</sub> rijke gasstroom uit de PSA wordt verbrand in het fornuis om de reformer van warmte te voorzien ontstaat er een verdunde CO<sub>2</sub> stroom. Anderzijds kan de productie van 'pure CO<sub>2</sub>' toenemen doordat de vraag naar waterstof op raffinaderijen zal stijgen. Deze stijging wordt veroorzaakt door een verschuiving naar zwaardere aardolie, een groeiende brandstofvraag en strengere eisen voor het zwavelgehalte in transportbrandstoffen. Een deel van deze groei zal worden voorzien door het vergassen van olieresiduen, waar CO<sub>2</sub> tegen lage kosten kan worden afgevangen.

4. Bij de productie van ethanol door het vergisten van suikers wordt een pure CO<sub>2</sub> stroom geproduceerd.

In *hoofdstuk 6* geven we een overzicht van de gezondheids-, veiligheids- en milieurisico's van ondergrondse CO<sub>2</sub> opslag, de huidige hiaten in kennis en de R&D gebieden die in dit verband meer aandacht vereisen. Inzicht in deze risico's is noodzakelijk om te verzekeren dat CCS op een veilige en effectieve manier kan worden toegepast. Veiligheid is een belangrijk punt van aandacht voor het transporteren en injecteren van CO<sub>2</sub>. Hoewel CO<sub>2</sub> niet explosief of ontvlambaar is, kan blootstelling aan verhoogde CO<sub>2</sub> concentraties bij mensen en dieren leiden tot schade en eventueel tot verstikking. De risico's veroorzaakt door pijpleidingen en injectiefaciliteiten voor CO<sub>2</sub> kunnen worden berekend en geminimaliseerd door middel van technologieën en veiligheidsmaatregelen die al worden toegepast in de olie- en gas industrie.

Er is echter minder bekend over de risico's van ondergrondse CO<sub>2</sub> opslag. Het injecteren van CO<sub>2</sub> in geologische reservoirs kan mogelijk leiden tot CO<sub>2</sub> en CH<sub>4</sub> lekkage, seismische activiteit, bodemdaling of stijging, en het verplaatsen van grondwater. Er wordt momenteel veel onderzoek gedaan naar lekkage van CO<sub>2</sub> uit geologische reservoirs. Op een lokale schaal kan lekkage leiden tot een verslechterde kwaliteit van (drinkbaar) grondwater en kan het schade aan organismen en ecosystemen veroorzaken. Vanuit een globaal perspectief reduceert CO<sub>2</sub> lekkage de effectiviteit van CCS als mitigatieoptie. De belangrijkste paden waarlangs CO<sub>2</sub> uit een reservoir kan ontsnappen zijn putten,

breuken en scheuren in de afsluitende laag. De kans op CO<sub>2</sub> lekkage is afhankelijk van de manier waarop CO<sub>2</sub> wordt opgeslagen (in vrije vorm, opgelost, gemineraliseerd, geadsorbeerd) en van de integriteit van de putten, afsluitende laag en de geologische formaties boven het reservoir. Ook het reservoirtype waarin CO<sub>2</sub> wordt opgeslagen is van groot belang. Olie- en gasvelden zijn goed bestudeerd en worden in het algemeen beschouwd als veilige reservoirs voor CO<sub>2</sub> opslag, omdat ze koolwaterstoffen en vaak ook CO<sub>2</sub> voor duizenden tot miljoenen jaren hebben ‘vastgehouden’ zonder catastrofale lekkage. Een nadeel is echter dat olie- en gasvelden vaak zijn aangeboord door vele exploratie- en productieputten, waarvan een aantal in slechte staat verkeert. De voornaamste zorg voor CO<sub>2</sub> opslag in aquifers is de integriteit van de afsluitende laag. Het risico van CO<sub>2</sub> lekkage uit kolenlagen is naar verwachting laag (zolang de druk in de kolen niet wordt verlaagd), omdat CO<sub>2</sub> sterker aan de kolenmatrix wordt geadsorbeerd dan methaan.

Hoewel de mechanismen van CO<sub>2</sub> lekkage bekend zijn, is het kwantificeren van de risico's nog een grote uitdaging. Zowel natuurlijke en industriële analogieën, als modellen die de verplaatsing van CO<sub>2</sub> in geologische reservoirs berekenen suggereren dat de kans klein is dat er een grote fractie van CO<sub>2</sub> zal ontsnappen, mits het reservoir zorgvuldig is geselecteerd. Er is echter meer R&D nodig om de processen te kwantificeren die CO<sub>2</sub> lekkage langs of door putten, breuken en scheuren controleren, zodat CO<sub>2</sub> fluxen kunnen worden berekend voor verschillende geologische condities. Belangrijke aandachtsvelden zijn de degradatie van cement en de casing als gevolg van (lange-termijn) blootstelling aan CO<sub>2</sub>, en de rol die de neerslag van mineralen speelt in het afsluiten van breuken. Hoewel experimenten and modellen suggereren dat de geochemische impact van CO<sub>2</sub> op de afsluitende laag beperkt is of zelfs positief, vereist dit gebied meer aandacht. Er is ook meer onderzoek nodig om de effecten van CO<sub>2</sub> op (mariene) ecosystemen in kaart te brengen en te kwantificeren. Natuurlijke analogieën zijn erg waardevol om beter inzicht te krijgen in de milieueffecten van CO<sub>2</sub> lekkage en het lange-termijn effect van de interactie tussen CO<sub>2</sub> en het reservoir en de afsluitende laag. Industriële analogieën zoals de injectie van CO<sub>2</sub> om extra olie te produceren (EOR) en de injectie van ‘acid gas’ (een mengsel van CO<sub>2</sub> en H<sub>2</sub>S) bieden de mogelijkheid om het gedrag van CO<sub>2</sub> in geologische reservoirs te bestuderen. Bovendien geven deze analogieën mogelijk informatie over de frequentie en mate van CO<sub>2</sub> lekkage.

*Hoofdstuk 7* is feitelijk een synthese van vergaarde kennis op het gebied van CO<sub>2</sub> afvang, transport en opslag, toegepast op de Nederlandse situatie. Het doel is om de rol van CCS te onderzoeken in het reduceren van CO<sub>2</sub> emissies, welke op dit moment ongeveer 180 mt per jaar bedragen. Om dit doel te bereiken is eerst het CO<sub>2</sub> afvangpotentieel in verschillende sectoren onderzocht. Hieruit blijkt dat in 2020 maximaal 50 mt CO<sub>2</sub>/jr zou kunnen worden gereduceerd door CO<sub>2</sub> af te vangen uit

(rook)gassen geproduceerd door elektriciteitscentrales, hoogovens, boilers en fornuizen op raffinaderijen, stoomkrakers, reformers en vergassers. De mitigatiekosten (exclusief transport en opslag) variëren van 10 € per ton vermeden CO<sub>2</sub> voor industriële bronnen die pure CO<sub>2</sub> produceren tot 100 € per ton vermeden CO<sub>2</sub> voor het retrofitten van de oudste poederkoolcentrales. Halverwege deze eeuw wordt het mitigatiepotentieel voor CO<sub>2</sub> afvang geschat op maximaal 80-110 Mt CO<sub>2</sub>/jr, waarvan 60-80 Mt CO<sub>2</sub>/jr mogelijk kan worden gerealiseerd onder de 20 € per ton vermeden CO<sub>2</sub> (exclusief transport en opslag). De elektriciteitssector biedt het grootste afvangpotentieel: wij schatten in dat circa 60-84 Mt CO<sub>2</sub>/jr kan worden vermeden in 2050 voor scenario's waarin de elektriciteitsproductie tegen die tijd is verdubbeld. Industriële bronnen voegen nog eens 16 Mt CO<sub>2</sub>/jr toe. Daarnaast creëert de ontwikkeling van een markt voor alternatieve transportbrandstoffen geproduceerd via vergassing of reforming nieuwe CCS mogelijkheden. Het reductiepotentieel in de transportsector voor 2050 is geschat op grofweg 10 Mt CO<sub>2</sub>/jr voor Fischer-Tropsch diesel of H<sub>2</sub> productie met CCS.

In onze analyse wordt de daadwerkelijke realisatie van het afvangpotentieel bepaald door emissiereductie doelstellingen en de geologische capaciteit voor CO<sub>2</sub> opslag. De onzekerheid in deze twee bepalende factoren wordt ondervangen door het toepassen van scenarioanalyses. Daartoe zijn vier paden geschetst hoe de (r)evolutie van CCS in Nederland mogelijkerwijs kan verlopen tot het jaar 2050. Op basis van deze paden schatten wij in dat het mogelijk is om 15 Mt CO<sub>2</sub>/jr te reduceren met CCS in 2020, mits een aantal van de wat grotere gasvelden die in de komende 10 jaar vrijkomen voor CO<sub>2</sub> opslag kunnen worden gebruikt. Als grotere reducties vereist zijn kunnen bestaande poederkoolcentrales en industriële boilers en fornuizen worden uitgerust met CO<sub>2</sub> afvang. Dit vereist dat een deel van de grotere reservoirs in de Noordzee vrijkomt voor CO<sub>2</sub> opslag. Anders moeten clusters van relatief kleine gasvelden worden ontsloten, wat leidt tot relatief hoge kosten van ongeveer 15 € per ton CO<sub>2</sub> voor transport en opslag. Indien Nederland enkel haar aandacht vestigt op binnenlandse mogelijkheden kan het waarschijnlijk zonder veel problemen 30 Mt CO<sub>2</sub>/jr reduceren in 2050. Het is zelfs mogelijk om 60 Mt CO<sub>2</sub>/jr te vermijden in 2050, mits een goede planning wordt opgezet om te verzekeren dat de Nederlandse reservoirs die geleidelijk aan vrijkomen kunnen worden aangewend voor CO<sub>2</sub> opslag. Wanneer 50-80% CO<sub>2</sub> emissiereductie moet zijn bewerkstelligd in 2050 (t.o.v. 1990) zou eventueel 50 Mt CO<sub>2</sub>/jr extra kunnen worden gereduceerd met CCS, mits alle afvangmogelijkheden die wij hebben bestudeerd worden gerealiseerd. Het opslaan van zulke enorme hoeveelheden CO<sub>2</sub> is alleen mogelijk als een aantal van de megastructuren vrijkomen voor CO<sub>2</sub> opslag: het Groningen veld, grote reservoirs in de Bunter formatie of de Utsira formatie in het Britse respectievelijk Noorse deel van de Noordzee. Een andere mogelijkheid is om CO<sub>2</sub> op te slaan in delen van aquifers die niet structureel worden afgesloten. Aan al deze opslagmogelijkheden

kleven onzekerheden. Zo is het de vraag wanneer het Groningen veld precies vrijkomt. De beschikbaarheid van de reservoirs in het Britse deel van de Noordzee is afhankelijk van de ontwikkelingen rondom ccs in het Verenigd Koninkrijk. CO<sub>2</sub> opslag in de Utsira formatie is enkel realistisch als er een (kostbare) offshore CO<sub>2</sub> infrastructuur wordt gebouwd. Als we het Groningen veld, de buitenlandse reservoirs en alle factoren die het technische opslagpotentieel reduceren buiten beschouwing laten zal er een tekort aan opslagcapaciteit ontstaan ergens tussen 2050 en 2075 wanneer we CO<sub>2</sub> opslag opvoeren tot boven de 80 mt CO<sub>2</sub>/jr. Wanneer Nederland maximaal 30 mt CO<sub>2</sub> per jaar opslaat kan ccs in principe worden voortgezet tot na 2100, ervan uitgaande dat alle vrijkomende gasvelden ook geschikt zijn voor CO<sub>2</sub> opslag.

Gasvelden zijn in principe de meest geschikte en voor de hand liggende reservoirs voor CO<sub>2</sub> opslag gezien hun grote, en relatief 'veilige', opslagcapaciteit. Het Groningen veld levert ongeveer 70% van de opslagcapaciteit. Dit veld komt naar verwachting pas na 2040 en mogelijk zelfs (ver) na 2050 vrij. De overige 30% bestaat uit kleinere velden, die de komende 20 jaar vrijkomen. Lang niet al deze velden zijn geschikt of beschikbaar voor ccs. Er kunnen zich situaties voordoen dat gasvelden niet genoeg opslagcapaciteit bieden op het juiste moment, waardoor een beroep moet worden gedaan op aquifers en/of kolenlagen. Het is echter riskant om op deze structuren te rekenen als onderdeel van een strategie van grootschalige ccs implementatie gezien de onzekerheid in opslagcapaciteit en de haalbaarheid van CO<sub>2</sub> opslag in deze reservoirs.

#### 4 ALGEMENE CONCLUSIES

Hoewel de antwoorden op de vier onderzoeksvragen te vinden zijn in de hierboven beschreven resultaten geven we in deze paragraaf een reflectie op de algemene bevindingen per vraag.

**ONDERZOEKSVRAAG I:** Wat zijn de technische mogelijkheden voor ccs en wat zijn de energetische rendementen en kosten van deze opties in een keten van bron tot reservoir?

De analyses beschreven in hoofdstuk 2 en 3 geven inzicht in de rendementen en kosten van huidige en toekomstige technologieën voor de productie van elektriciteit en waterstof met ccs. De elektrische rendementen van huidige conversietechnologieën met CO<sub>2</sub> afvang en compressie zijn ingeschat op 32-35% voor kolengestookte centrales en circa 47% voor gasgestookte centrales. Onze berekeningen laten zien dat elektriciteitsproductiekosten voor deze technologieën rond de 5-7 €/ct/kwh liggen. CO<sub>2</sub> mitigatiekosten ten opzichte van een identieke centrale zonder CO<sub>2</sub> afvang zijn geschat op 15-50 € per ton vermeden CO<sub>2</sub> (exclusief transport en opslag). Elektrische rendementen kunnen op de

lange termijn worden verbeterd met circa 30-40% en mitigatiekosten zouden gereduceerd kunnen worden tot 10-40 € per ton vermeden CO<sub>2</sub>. Deze getallen geven een indicatie van de benodigde CO<sub>2</sub> prijs of onrendabele top om CCS van de grond te krijgen.

Een van de belangrijkste uitdagingen voor energiebedrijven is het optimaliseren van hun portfolio gegeven onzekere gas en CO<sub>2</sub> prijzen. Onze analyses laten zien dat basislast elektriciteit tegen de laagste kosten kan worden gegenereerd met een STEG zonder CO<sub>2</sub> afvang voor gasprijzen lager dan 5,5-7 €/GJ en CO<sub>2</sub> prijzen tot 40-55 € per ton CO<sub>2</sub>. Een KV-STEG met CO<sub>2</sub> afvang is het meest concurrerend wanneer gasprijzen deze grens overschrijden en CO<sub>2</sub> prijzen stijgen boven de 15-25 € per ton. In 2006 zijn een aantal plannen aangekondigd voor de bouw van elektriciteitscentrales met geïntegreerde CO<sub>2</sub> afvang in Noord Amerika, Europa en Australië. Deze projecten, met een geplande inbedrijfsname tussen 2010 en 2015, tonen een verscheidenheid aan keuzes met betrekking tot brandstof en technologie. Een aantal bedrijven lijken te kiezen voor een KV-STEG centrale met 'pre-combustion capture'. KV-STEG is een relatief weinig toegepaste en kapitaalintensieve technologie in vergelijking met conventionele poederkoolcentrales. Bovendien hebben KV-STEG eenheden nog niet een dergelijk hoge beschikbaarheid als poederkoolcentrales en is veel van de ervaring met vergassingstechnologie geconcentreerd in de (petro)chemische industrie. Om deze redenen zijn veel elektriciteitsbedrijven geneigd om te kiezen voor bewezen techniek in de vorm van een poederkoolcentrale met een scrubber om CO<sub>2</sub> uit het rookgas te verwijderen. Andere elektriciteitsbedrijven lijken meer heil te zien in verbranding van (bruin)kolen met pure zuurstof. Er is nog een aanzienlijk verbeterpotentieel in deze afvangtechnieken door toepassing van absorptiemiddelen met een lagere regeneratie energie en zuurstofproductie met membranen. Desalniettemin suggereren onze analyses dat vergassing in combinatie met zogenaamde 'pre-combustion capture' de efficiëntste en goedkoopste technologie is om elektriciteit te produceren uit bitumineuze kolen met sterk gereduceerde CO<sub>2</sub> emissies. De kapitaalkosten van een KV-STEG kunnen nog significant worden gereduceerd. Daarentegen zijn voor de bouw van de modernste poederkoolcentrales dure materialen nodig om hogere stoomtemperaturen en daarmee hogere rendementen te realiseren. Een ander voordeel van vergassingstechnologie is de flexibiliteit om meerdere secundaire energiedragers (elektriciteit, waterstof en synthetische brandstoffen) te produceren uit verschillende primaire energiedragers (kolen, olieresiduen en biomassa). Er is echter meer inzicht nodig in de invloed van verschillende brandstoffen en variërende capaciteitsfactoren op de bedrijfsvoering in combinatie met CO<sub>2</sub> afvang. Ook dienen er een aantal grootschalige KV-STEG projecten worden gerealiseerd om operationele ervaring op te doen. Een belangrijk punt is het verbeteren van de beschikbaarheid van KV-STEG eenheden tot een niveau vergelijkbaar als dat van poederkoolcentrales. In onze analyse zijn ook verschillende CO<sub>2</sub> afvangtechnieken voor

aardgasgestookte STEG eenheden geanalyseerd. Hier kwam echter geen voorkeur uit voor een bepaalde techniek, voornamelijk vanwege de onzekerheid in kapitaalkosten.

Het IPCC special report on carbon dioxide capture and storage richt zich voornamelijk op grote CO<sub>2</sub> bronnen, omdat directe CO<sub>2</sub> afvang bij kleine bronnen naar verwachting gecompliceerder en duurder is. Hoewel onze analyses ook vooral gericht zijn op grote bronnen hebben we daarnaast de rendementen en kosten onderzocht van decentrale elektriciteitsproductie met behulp van SOFC brandstofcellen en waterstofproductie door toepassing van membraanreformers gecombineerd met CCS. De energieproductiekosten van kleinschalige systemen zijn inderdaad hoger in vergelijking met grootschalige systemen, maar toch kunnen deze technologieën interessant zijn in een bepaalde context. Omdat CO<sub>2</sub> tegen relatief lage additionele kosten kan worden afgevangen, zijn de productiekosten van elektriciteit en waterstof nog enigszins acceptabel indien een cluster van eenheden kan worden aangesloten op een nabij gelegen CO<sub>2</sub> pijpleiding of opslagreservoir. SOFC is een interessante technologie voor Nederland gezien het hoge aandeel van decentrale industriële WKK eenheden en de aanwezigheid van vele gasvelden die mogelijk voor CO<sub>2</sub> opslag kunnen worden gebruikt. Membraanreformers zijn interessant in het begin van een waterstof transitie, wanneer de vraag(dichtheid) nog te laag is voor productie in grootschalige eenheden, waarvoor een relatief dure infrastructuur nodig is. Dichtbevolkte industriële gebieden met een goed ontwikkelde aardgas infrastructuur lijken de meest geschikte niche. Aangezien de faciliteiten die nodig zijn om CO<sub>2</sub> af te vangen relatief eenvoudig bij zijn te plaatsen kan de membraanreformer in eerste instantie zonder afvang draaien als er nog geen CO<sub>2</sub> infrastructuur is om op aan te sluiten.

**ONDERZOEKSVRAAG 2:** In welke mate komt de ruimtelijke verdeling van CO<sub>2</sub> bronnen en reservoirs overeen, en wat zijn de mogelijkheden om pure CO<sub>2</sub> bronnen te combineren met nabij gelegen reservoirs die goedkope CO<sub>2</sub> opslag mogelijk maken?

Deze vraag is op verschillende geografische niveaus beantwoord. In hoofdstuk 5 zijn we ingegaan op *wereldwijde* bron-reservoir combinaties die tegen lage kosten zouden kunnen worden gerealiseerd. In hoofdstuk 7 is een gedetailleerde ‘matching’ van CO<sub>2</sub> bronnen en reservoirs uitgevoerd voor *Nederland*.

De resultaten van hoofdstuk 5 laten zien dat er honderden industriële bronnen zijn die nagenoeg pure CO<sub>2</sub> produceren dat kan worden opgeslagen in olievelden of kolenlagen binnen een straal van 100 km. Het is belangrijk om te beseffen dat de emissie van deze bronnen slechts een kleine fractie is van de mondiale CO<sub>2</sub> emissie afkomstig van de industrie- en energiesector. Toch is de toepassing van deze mogelijkheden belangrijk voor de verdere ontwikkeling van CCS

doordat er operationele ervaring wordt opgedaan waardoor zowel bedrijven als publiek vertrouwen in de techniek kunnen krijgen. Bovendien zijn de mitigatiekosten in het geval van de EOR cases, waarbij extra olie wordt geproduceerd door CO<sub>2</sub> injectie, laag of mogelijk negatief afhankelijk van de olieprijzen. Hetzelfde geldt ook voor CO<sub>2</sub> injectie in kolenlagen dat extra methaan kan genereren (ECBM). In tegenstelling tot EOR, wat al op commerciële basis wordt toegepast, is ECBM echter nog in de demonstratie fase. China heeft een enorm technisch potentieel voor ECBM. Daarnaast zullen er komende decennia waarschijnlijk vele vergassingsinstallaties worden gebouwd om elektriciteit, chemicaliën en brandstoffen te produceren, waar CO<sub>2</sub> afscheiding noodzakelijk is of tegen relatief lage kosten is te realiseren. Deze trend (naar vergassing) zal naar alle waarschijnlijkheid ook plaatsvinden in andere delen van de wereld. Zo zal er een nieuwe generatie goedkope CCS mogelijkheden ontstaan. Het CO<sub>2</sub> reductiepotentieel dat hiermee ontstaat is in principe groter dan het potentieel dat de huidige pure CO<sub>2</sub> bronnen bieden, omdat de geproduceerde energiedragers in zowel de transport, huishoudelijke als commerciële sector kunnen worden toegepast.

In Nederland wordt op dit moment jaarlijks ongeveer 2,5 mt pure CO<sub>2</sub> uitgestoten. Een waterstoffabriek en een offshore installatie die aardgas verwerkt produceren jaarlijks samen 1 mt CO<sub>2</sub> dat kan worden opgeslagen in (bijna) uitgeputte olie- of gasvelden. De overige CO<sub>2</sub> wordt geëmitteerd bij de productie van ammoniak en ethyleenoxide. Er zijn echter geen olie- of gasvelden in de buurt van de ammoniakfabrieken en CO<sub>2</sub> transport naar verder gelegen reservoirs is duur vanwege de relatief kleine volumes. Eén fabriek ligt vrijwel boven een aantal kolenlagen; mogelijk kan CO<sub>2</sub> in deze lagen worden geïnjecteerd als deze techniek is bewezen. De hoeveelheden CO<sub>2</sub> die vrijkomen tijdens ethyleenoxide productie zijn erg klein waardoor CCS relatief duur zou worden. In het algemeen komt de ruimtelijke verdeling van de CO<sub>2</sub> bronnen en reservoirs in Nederland niet goed overeen. De meeste bronnen liggen in het westen, terwijl de meeste gasvelden in het noorden of offshore liggen. De transportafstanden zijn echter relatief klein; de maximale afstand tussen bron en reservoir is circa 300 km. Een deel van de geproduceerde CO<sub>2</sub> kan wellicht worden getransporteerd naar de grote reservoirs buiten het Nederlandse deel van het continentale plat, omdat de opslagkosten voor deze reservoirs lager zijn dan voor de relatief kleine reservoirs in Nederland. Onze scenarioanalyses laten zien dat CO<sub>2</sub> opslag buiten Nederland waarschijnlijk wordt wanneer grote CO<sub>2</sub> emissiereducties worden nagestreefd, ervan uitgaande dat het Groningen veld voorlopig niet beschikbaar is.

**ONDERZOEKSVRAAG 3:** Wat zijn de voornaamste factoren die de opslagcapaciteit beïnvloeden? Meer in het bijzonder, wat zijn de risico's van ondergrondse CO<sub>2</sub> opslag voor de mens en het milieu en wat zijn de huidige hiaten in de kennis van deze risico's?

De huidige verwachtingen over de bijdrage die CCS kan leveren aan het terugdringen van de CO<sub>2</sub> uitstoot zijn deels gebaseerd op schattingen van theoretische en/of technische opslagcapaciteiten. Op basis van de overvloed aan opslagcapaciteit in gasvelden concluderen velen dat CCS een belangrijke rol zou kunnen spelen in Nederland. Tegelijkertijd zijn deze gasvelden, mede dankzij hun locatie in Noordwest Europa, ideale kandidaten voor ondergrondse gasopslag (UGS). Er kan dus concurrentie optreden tussen UGS en CCS, en op de lange termijn kan ook waterstofopslag in ondergrondse reservoirs een rol gaan spelen. In de komende 20 jaar is mogelijk 100-150 miljard kubieke meter aan opslagcapaciteit nodig voor UGS indien Nederland zich ontwikkelt tot een internationale gas 'rotonde'. Dit komt overeen met 13 tot 20% van de geschatte opslagcapaciteit van alle gasvelden op land, exclusief het Groningen veld. Onze scenariostudies geven aan dat CCS in 2050 maximaal 80% van het Nederlandse opslagpotentieel in gasvelden op land zal 'claimen'. Dit maximum kan worden bereikt in een scenario waar in 2050 jaarlijks circa 70 Mt CO<sub>2</sub> wordt vermeden met CCS en waar CO<sub>2</sub> opslag beperkt is tot Nederlandse reservoirs.

Dit soort overwegingen dienen meegenomen te worden in het ontwikkelen van beleid en strategieën rondom energiezekerheid en CO<sub>2</sub> emissiereductie zoals dat gestalte krijgt in het energietransitie proces dat in Nederland gaande is. Indien Nederland zich wil ontwikkelen als internationaal aardgas en CO<sub>2</sub> knooppunt in Noordwest Europa, is het noodzakelijk om de vraag naar, en de beschikbaarheid van gasvelden optimaal op elkaar af te stemmen.

De CO<sub>2</sub> opslagcapaciteit van ondergrondse reservoirs kan verder worden beperkt door een reeks van geologische en geotechnische factoren, waaronder het risico op CO<sub>2</sub> lekkage. Lekkende reservoirs leiden mogelijk tot schadelijke effecten voor mens en milieu en reduceren de opslageffectiviteit. Aldus kan lekkage ook de publieke acceptatie van CCS ondermijnen. Hoewel de mechanismen waarop CO<sub>2</sub> kan ontsnappen bekend zijn is er nauwelijks kwantitatieve informatie over lekkage. Met het verschijnen van het IPCC special report on carbon dioxide capture and storage werden aardwetenschappers uitgedaagd om een schatting te geven van de kans op CO<sub>2</sub> lekkage. Dit leidde tot de volgende (vertaalde) uitspraak:

*'Waarnemingen van industriële en natuurlijke analogieën en modellen suggereren dat de fractie CO<sub>2</sub> die in het opslagreservoir blijft na 100 jaar zeer waarschijnlijk<sup>5</sup> groter is dan 99%, en na 1000 jaar waarschijnlijk<sup>6</sup> meer dan 99%, mits de reservoirs zorgvuldig worden geselecteerd en gemanaged'.*

De toevoeging 'zorgvuldig geselecteerd' impliceert dat er reservoirs zijn

5. Waarschijnlijkheid van 90-99%.

6. Waarschijnlijkheid van 66-90%.

waar het risico op CO<sub>2</sub> lekkage groter is. In zulke gevallen zou het extra kosten en/of monitoring vergen, waardoor ze minder of helemaal niet geschikt zijn voor CO<sub>2</sub> opslag. Dit brengt ons tot de vraag in welke mate het technische opslagpotentieel wordt gereduceerd door normen betreffende het toelaatbare risico van CO<sub>2</sub> lekkage en andere geotechnische factoren. Om deze vraag te beantwoorden dienen aardwetenschappers de volgende acties te ondernemen:

1. De processen die lekkage controleren en CO<sub>2</sub> fluxen bepalen moeten beter worden bestudeerd en gekwantificeerd. Aangezien de risico's op ontsnapping van CO<sub>2</sub> erg locatie en reservoir specifiek zijn, is er een verscheidenheid aan demonstratie opslagprojecten nodig met intensieve monitoring.
2. Er zijn meer gedetailleerde geologische en geotechnische studies nodig naar de opslagmogelijkheden van individuele reservoirs. Voor Nederland zouden ten eerste een aantal van de grotere reservoirs, waaronder Annerveen, beoordeeld moeten worden op hun geschiktheid voor CCS en UGS.
3. Er dient een raamwerk te worden ontwikkeld om de opslagcapaciteit te screenen en te classificeren in (op het eerste gezicht) geschikte en minder geschikte reservoirs. Een aantal aanzetten hiertoe zijn gemaakt, maar er is nog meer werk nodig. Hierin dienen de inzichten die onder punt 1 zijn verkregen te worden meegenomen.

Concluderend kunnen we stellen dat er meer werk moet worden verricht om te kwantificeren in hoeverre bovengenoemde factoren het opslagpotentieel reduceren. In feite moeten we een stap verder gaan dan de bestaande schattingen van het theoretische en/of technische opslagpotentieel en de daadwerkelijk beschikbare opslagcapaciteit bepalen. Wanneer zulke data beschikbaar komen kunnen de mogelijkheden voor CCS met meer zekerheid vastgesteld worden.

**ONDERZOEKSVRAAG 4:** Wat is de potentiële rol die CCS zou kunnen vervullen in het 'ontkolen' van de Nederlandse energievoorziening in de tijd en wat betekent dit in termen van vermeden CO<sub>2</sub> emissies, kosten en benodigde infrastructuur?

Grootschalige toepassing van CCS is niet te verwachten voor 2012 gezien de onzekerheid in internationaal klimaatbeleid, het in bedrijf nemen van nieuwe centrales en de tijd die nodig is om CCS op commerciële schaal te demonstreren en een CO<sub>2</sub> infrastructuur aan te leggen. In de komende jaren kan CCS worden geïnitieerd met de opslag van ongeveer 1 Mt CO<sub>2</sub>/jr geproduceerd in de productie van waterstof en de zuivering van aardgas. In 2020 kan de uitstoot jaarlijks met circa 12-15 Mt CO<sub>2</sub> gereduceerd worden door CO<sub>2</sub> afvang te realiseren bij nog te bouwen kolengestookte elektriciteitscentrales die na 2010 in bedrijf worden genomen. Indien grotere reducties noodzakelijk zijn kunnen bestaande poederkoolcentrales en industriële boilers en fornuizen worden uitgerust met CO<sub>2</sub> afvanginstallaties, ofschoon de kosten van deze optie

relatief hoog zijn in vergelijking met CO<sub>2</sub> afvang bij nieuw te bouwen centrales. In 2050 schatten wij het CCS reductiepotentieel in op meer dan 100 Mt CO<sub>2</sub>/jr uitgaande van een scenario waarin de energievraag en CO<sub>2</sub> emissies sterk toenemen. Deze schatting is aanzienlijk hoger dan de waarde van 40-60 Mt CO<sub>2</sub>/jr die wordt vermeld in een aantal Nederlandse beleidsstudies. Op basis van onze scenarioanalyses schatten wij dat in 2050 ongeveer 60-80 Mt CO<sub>2</sub>/jr zou kunnen worden gereduceerd tegen kosten van 10 tot 40 € per ton vermeden CO<sub>2</sub>, inclusief transport en opslag. Ter vergelijking: de CO<sub>2</sub> emissies in Nederland stijgen van circa 180 Mt CO<sub>2</sub>/jr nu naar 250-330 Mt CO<sub>2</sub>/jr in 2050 voor de referentiescenario's zonder klimaatbeleid. Op basis hiervan concluderen wij dat CCS een grote bijdrage kan leveren in het reduceren van de CO<sub>2</sub> uitstoot in Nederland tegen relatief lage kosten. Daadwerkelijke realisatie van het bovengenoemde potentieel tegen de genoemde kosten vergt echter lange-termijn klimaatbeleid en een heldere en internationaal georiënteerde visie op de organisatie en implementatie van CCS in de komende decaden. De komende jaren dienen geschikte (clusters van) reservoirs te worden geïdentificeerd die op het juiste tijdstip vrijkomen, genoeg opslagcapaciteit hebben en niet voor andere toepassingen in aanmerking komen. Gezien het feit dat veel reservoirs voor 2025 zijn uitgeproduceerd, moet er een strategie ontwikkeld worden om genoeg opslagcapaciteit te garanderen mocht CCS een vlucht gaan nemen tussen 2020 and 2040. Uitgaande van het technische opslagpotentieel in Nederland zou het in principe mogelijk kunnen zijn om 60-80 Mt CO<sub>2</sub>/jr voor tientallen jaren op te slaan. Toch is het goed mogelijk dat ook reservoirs buiten het Nederlands deel van het continentale plat worden gebruikt om 'Nederlands' CO<sub>2</sub> op te slaan. In een dergelijk scenario is een internationaal gecoördineerde aanpak nodig om de CO<sub>2</sub> stromen en de beschikbare reservoirs te matchen. Ook de constructie van een offshore CO<sub>2</sub> infrastructuur is mogelijk een internationale actie. Aangezien België en Duitsland bovendien een gebrek aan opslagcapaciteit hebben en mogelijk dus toegang zoeken tot reservoirs in Nederland en de Noordzee is het van belang dat Nederland een CCS strategie ontwikkelt waarin de Noordwest Europese context wordt meegenomen. Realisatie van het toekomstbeeld waarin Nederland CO<sub>2</sub> transporteert en opslaat voor de ons omringende landen, zoals wel eens geopperd in beleidsdiscussies, is sterk afhankelijk van post-Kyoto doelstellingen, ontwikkelingen in de industrie en energiesector en de vraag naar UGS in Noordwest Europa. Als de EU en Nederland echter een streng klimaatbeleid uitstellen kunnen de kosten van CCS toenemen, omdat mogelijkheden voor opslag in bepaalde reservoirs, CO<sub>2</sub>-EOR op de Noordzee en hergebruik van bestaande infrastructuur niet meer zijn te verzilveren. Bovendien is het sterk de vraag of een kostenniveau van 10-40 € per ton vermeden CO<sub>2</sub> realistisch is indien er geen of nauwelijks CCS capaciteit wordt gebouwd in de komende decennia, wat van groot belang is om kosten te verminderen door technologisch leren.

Omdat de afstanden tussen CO<sub>2</sub> bronnen en reservoirs in Nederland niet erg groot zijn dragen transportkosten in het algemeen nauwelijks bij aan de mitigatiekosten, mits de volumes niet te klein zijn (<1 mt/jr). De keuze tussen een één-op-één pijpleiding of een pijpleiding die meerdere bronnen en reservoirs verbindt is afhankelijk van de timing, de locatie van bronnen en reservoirs en de doelstellingen voor emissiereductie. Investeringskosten voor CO<sub>2</sub> transport liggen in de ordegrootte van 10<sup>7</sup>–10<sup>9</sup> Euro. De ondergrens is representatief voor kleine lokale pijpleidingen en de bovengrens voor lange-afstand offshore pijpleidingen van vergelijkbare afmetingen als huidige aardgas transmissieleidingen. Overheidssteun is mogelijk noodzakelijk om CO<sub>2</sub> leidingen (die eventueel de landgrens overschrijden) te financieren.

## 5 AANBEVELINGEN VOOR VERDER ONDERZOEK

In dit proefschrift zijn we op een aantal onderwerpen gestuit die mogelijk interessant zijn voor verder onderzoek. Een aantal is hieronder puntsgewijs samengevat:

- De mogelijkheden, kosten en technische implicaties van CO<sub>2</sub> afvang in combinatie met de productie van staal.
- Potentieel van de productie van synthetische brandstoffen door middel van kolen en biomassavergassing gekoppeld met CCS.
- Rendement, kosten en flexibiliteit van ‘multi-fuel poly-generation’ (d.w.z. verschillende primaire energie inputs en secundaire energie outputs) centrales gebaseerd op vergassingstechnologie met CO<sub>2</sub> afvang.
- Vooruitzichten van technologieën voor decentrale elektriciteitsproductie in combinatie met CCS, o.a. voor industriële WKK toepassingen. Technieken die nader onderzoek vergen zijn brandstofcellen (SOFC) en gasturbines waarin de brandstof met pure zuurstof wordt verbrand (AZEP en ZEPP)
- De potentiële rol van centrale en decentrale waterstofproductie uit fossiele brandstoffen in de transitie naar een waterstofeconomie.
- Reducties in kosten van CCS door technologisch leren (zowel kapitaal als operationele kosten).
- Specifieke kosten van CO<sub>2</sub> transmissieleidingen voor een aantal trajecten (Eemshaven-Annerven, Rijnmond-Groningen, Rijnmond-zuidelijke deel van de Britse Noordzee via de PKL kwadranten, Rijnmond/Eemshaven-Utsira via de Britse en/of Noorse olievelden).
- Haalbaarheid van het hergebruik van offshore gasinfrastructuur, waaronder de mogelijkheid om delen hiervan in te zetten in een CO<sub>2</sub> netwerk om reservoirs in het Nederlandse, Britse en Noorse deel van het continentale plat voor CO<sub>2</sub> opslag te ontsluiten.
- Het in kaart brengen en karakteriseren van aquifers en de daarin liggende traps (gesloten structuren), in het bijzonder voor het

Nederlandse deel van het continentale plat. Een proefproject is zinvol om CO<sub>2</sub> opslag in een aquifer trap te demonstreren en om inzicht te krijgen in de injectiviteit, de opslag efficiëntie (de fractie van het porievolume dat gevuld kan worden met CO<sub>2</sub>) en de integriteit van de afsluitende laag.

- Haalbaarheid van verschillende injectiestrategieën in het Groningen veld voordat het uitgeproduceerd is, zodat beoordeeld kan worden in hoeverre dit enorme reservoir een rol kan spelen in de toepassing van CCS.

Tenslotte pleiten we ervoor om een haalbaarheidsstudie uit te voeren naar de mogelijkheden voor CCS in de Rijnmond regio. In dit gebied zijn vele industriële activiteiten geconcentreerd en zijn een aantal nieuwe kolengestookte elektriciteitscentrales gepland. Ook zijn er een aantal olie- en gasvelden in dit gebied en het enorme potentieel in gasvelden op de Noordzee is relatief dichtbij. Specifieke aandachtspunten zijn co-siting (het delen en gebruikmaken van energie, ruimte en infrastructuur door meerdere bedrijven) en het potentieel om lage temperatuur stoom te gebruiken voor CO<sub>2</sub> regeneratie. Potentiële reservoirs moeten in detail worden onderzocht op de haalbaarheid van CO<sub>2</sub> opslag, net als de mogelijkheden voor de aanleg van een pijpleiding om verschillende CO<sub>2</sub> bronnen te verbinden met offshore reservoirs.





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