Impact of hydrogen onboard storage technologies on the performance of hydrogen fuelled vehicles: A techno-economic well-to-wheel assessment

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Abstract

Hydrogen onboard storage technologies form an important factor in the overall performance of hydrogen fuelled transportation, both energetically and economically. Particularly, advanced storage options such as metal hydrides and carbon nanotubes are often hinted favourable to conventional, liquid and pressurized, storage options. This study assesses the technical and economic performance of four onboard storage technologies in a well-to-wheel assessment. It does so for centralized and onsite hydrogen production scenarios with gaseous pipeline and liquid truck distributed hydrogen. Energetically, centralized production and liquefaction outperforms onsite production and liquefaction by 20% on the average primary energy input, despite the need of pipeline or over the road distribution. The gaseous handling on the filling station performs better with an average driving energy use of $0.11\,MJ_p/km$ compared to $0.15\,MJ_p/km$ for liquid handling. The advanced onboard storage technologies perform better by 22% than the conventional storage technologies on primary energy input in the chain. The average driving cost for gaseous and liquid onboard storage are 53% above the gasoline reference cost of 21 €ct/km and the advanced onboard storage technologies as much as 98%, almost double the reference cost. The car expenditure for a fuel cell driven car compared to an internal combustion engine driven car is for a large part responsible for this difference. Advanced onboard storage technologies thus perform better than conventional onboard storage technologies energetically albeit to higher cost.
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1. Introduction

With hydrogen becoming a possible contender to replace conventional car fuels, hydrogen production, conversion and storage technologies are subject to intensive research and development (R&D). Hydrogen is considered a promising energy carrier for a number of reasons. The diverse feedstocks hydrogen can be derived from give flexibility in the choice of primary energy resources with respect to availability and price. This flexibility is also an essential characteristic in the complex transition process in which a gradual shift from conventional (fossil) feedstocks towards sustainable feedstocks for hydrogen production is foreseen [1]. In an early stage of transition,

dominated by a large share of fossil resources, carbon dioxide sequestration and storage can ensure a gradual shift to sustainable transport.

One of the major barriers for large scale use of hydrogen is the infrastructural changes needed in the transition towards a hydrogen based transportation sector. The handling of gaseous hydrogen requires a radically different design on almost all levels of the supply chain infrastructure, including; production, distribution, refuelling and the vehicle design itself. Advanced storage technologies are currently and will in the future be heavier and require more space in the vehicle than conventional storage technologies. This may limit flexibility to the vehicle design and negatively affects the fuel economy. Costs related to this radical change and to the economic competitive production of hydrogen sustainably, pose another problem. The increase in overall chain efficiency that is foreseen, presumes that a mature, proton exchange membrane (PEM), fuel cell

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Nomenclature

CNT	carbon nanotube	LH$_2$	liquid hydrogen
CPSC	compressed steel cylinder	MH	metal hydrides
FCV	fuel cell vehicles	PEM	proton exchange membrane
HFV	hydrogen fuelled vehicle	PSA	pressure swing adsorption
ICE(V)	internal combustion engine (vehicle)	R&D	research and development
		SMR	steam methane reforming

technology for vehicles is available. PEM fuel cell technologies are, however, not expected to become commercially available for vehicle applications until 2020 [2]. Other factors, not directly related to energy or cost performance, affecting the overall performance of a storage technology are a longer refuelling time and possible poor material durability, influencing, apart from cost, the reliability of the product.

The supply chain and filling station handling of hydrogen differs from conventional fuel handling in the way that hydrogen is gaseous, with a low energy density, even at very low temperatures. The gaseous state of the hydrogen requires compression steps in the different stages of the hydrogen supply chain. The pressure gradient in the total chain is a decisive aspect in the overall chain efficiency. Current hydrogen onboard storage technologies store hydrogen in compressed or liquid form. The limited energy density of these storage technologies presents barriers with respect to required tank volumes and energy requirements for refuelling. These can influence vehicle costs and thus well-to-wheel costs significantly. In the search for storage options featuring more favourable characteristics, new advanced options are being researched.

Several advanced storage technologies are under research. Current studies underline the breakthrough role that advanced storage technologies can play, if cost and energetic performance goals are met [13]. The current results of fundamental laboratory studies, however, do not yet approach the possible theoretical performance predicted and required to fulfil the claims of favourable characteristics made by the industry [6]. Because of the fundamental stage of the R&D (e.g. [3–6]), the concrete future impact potential on the well-to-wheel performance of hydrogen fuelled vehicles (HFVs) is unclear.

Chain analysis is a much used approach in the evaluation of the impact of a specific aspect on the fuel supply chain. Aspects often considered in a well-to-wheel analysis are; energy efficiency, overall costs and the environmental impact. With respect to hydrogen supply chains, numerous system studies have been executed, with different subjects of focus. In several studies (e.g. [7–9]) different modes of onboard hydrogen use are evaluated, hydrogen refuelling with compressed hydrogen storage onboard and also studies that consider fossil fuel refuelling and onboard conversion into hydrogen by means of onboard steam reforming and onboard partial oxidation of hydrogen. Others [8] study hydrogen chains and focus on the evaluation of different applications for distribution, e.g. pipeline, liquid truck and rail. Analyses with respect to storage cost at different stages of the supply chain—production plant, filling station

and onboard storage—are rather limited and not analysed in a chain approach [9,10].

The objective of this paper is to evaluate the potential future techno-economic performance of onboard storage technologies by means of a chain analysis. The analysis consists of two parts: (1) a review is made of the status of the hydrogen storage options, their performance and improvement potential. (2) The storage options are compared for different supply chains, including hydrogen production at different scales and pressures. For doing so, the energetic performances of 14 pathways are analysed, resulting in an outcome of primary energy per kilometre driven, per technology. In addition a similar approach is applied to evaluate the economic performance, resulting in the calculated costs in euro per kilometre driven, per technology. The environmental impact of the chains is focussed on the calculated energy performance and on greenhouse gas emissions (an indication for one aspect of the environmental impact). Results can help identify pathways for efficient and (more) competitive use of hydrogen for road transport in general, and the impact of advanced hydrogen storage technologies in particular.

2. Method

The onboard storage media require hydrogen in liquid or gaseous form under different pressures, depending on specifications of the onboard storage technology. The supply chains and filling station configurations are designed in such a way to facilitate these requirements. The analysis is executed using a well-to-wheel approach. The results are given in terms of primary energy[1] use per kilometre driven and related cost. First the chains are defined. In the chain configuration some aspects and parameters are distinguished, which are discussed below. Three different filling station configurations are presented.

2.1. Chain definition

Firstly, the production of hydrogen, central and decentral (onsite) steam methane reforming (SMR) and liquifaction, is analysed based on literature data. This is relevant to enable analysis with respect to scale, efficiency and cost.

Secondly, the distribution system, gaseous pipeline distribution, liquid truck transport and filling station configurations are

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[1] *Primary energy*: Energy embodied in natural resources (e.g. coal, crude oil, sunlight, uranium) that has not undergone any anthropogenic conversions or transformations [11].
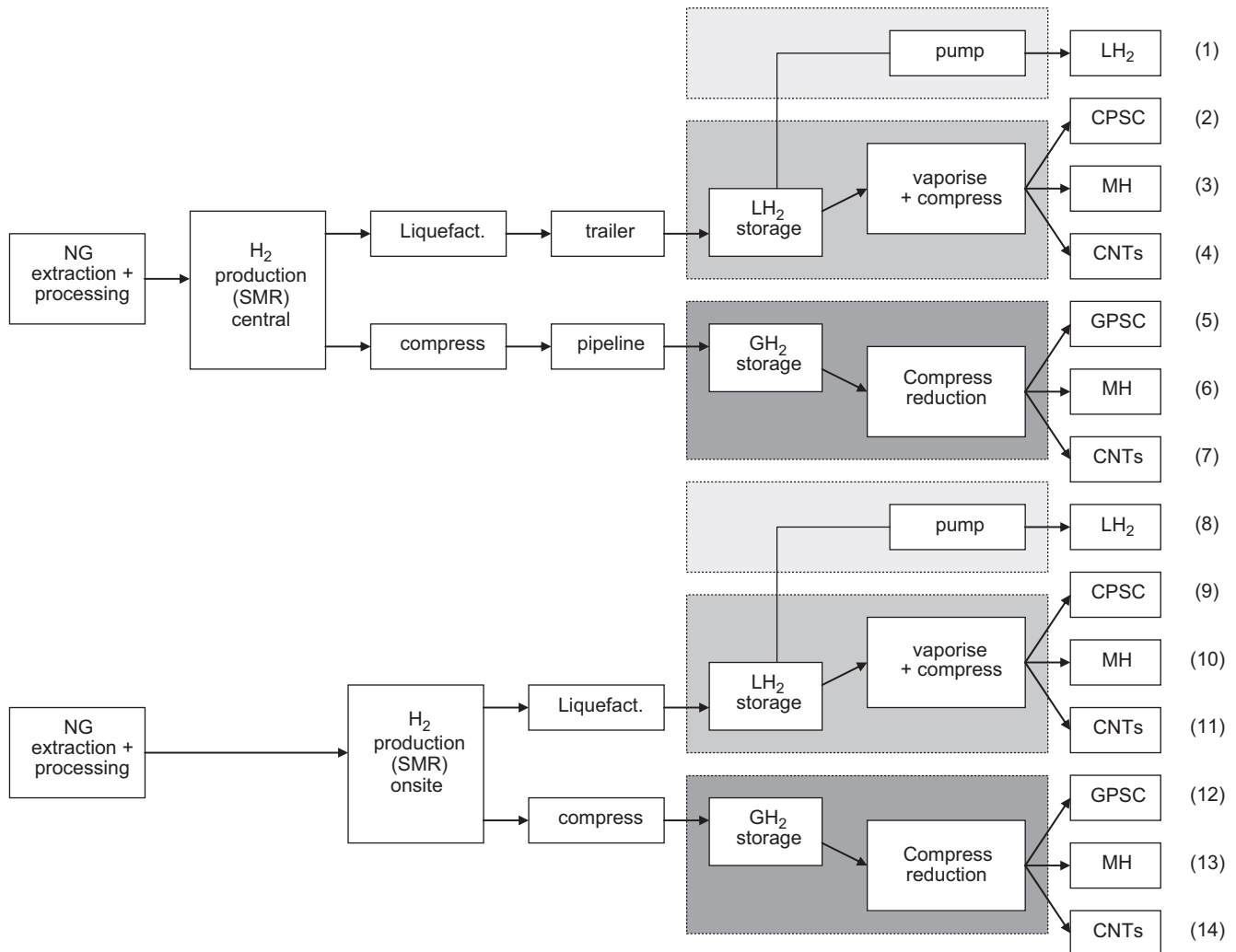
Fig. 1. Overview of assessed chains. Chain 1–7 are supplied by centrally produced and liquefied (1–4) hydrogen. Chain 8–14 use natural gas onsite to produce and liquefy (8–11) hydrogen. In the chains dark colored boxes indicate different filling station configurations. The top box represents a liquid hydrogen pump, the middle gives the configuration for a liquid supplied and gaseous dispensed system the bottom configuration shows a system for gaseous supply and dispensing.

evaluated. The evaluation comprises detailed calculation of the primary energy use and related cost of the distribution system. Three filling station configurations are considered to meet the required form and conditions of hydrogen for the onboard storage system of the vehicle. These three types comprise a gaseous supplied and two liquid supplied configurations. The vehicles included in the assessment are PEM fuel cell vehicles (FCV) and hydrogen fuelled internal combustion engine vehicles (ICEVs). Both propulsion type vehicles are assessed in a hybrid[2] form also, resulting in a total of four vehicle types assessed with respect to their fuel economy and driving properties. The resulting 14 chains are presented in Fig. 1.

Of key importance for an assessment focussed on differences between supply chains, filling station configurations, onboard storage and propulsion technologies is the inclusion of pressure

differences along the chain, and energy (heat) required to free hydrogen from the storage medium onboard the vehicle. This is explicitly taken into account in the specifications of the chains and included in the calculations.

3. Data and assumptions

3.1. Onboard storage technologies

Four hydrogen onboard storage technologies are included in the analysis. The four technologies are chosen from a range of possible storage technologies and are considered representative with respect to cost and energy efficiency performance in the well-to-wheel chain. Both the gaseous and the liquid onboard storage technologies are included in the analysis as key reference technologies. These reference technologies represent R&D efforts by some major car manufacturers (e.g. [12]). With respect to advanced options for hydrogen onboard storage, gas

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[2] PEM fuel cell operating on 100% compressed hydrogen with electric drive train with addition of a battery to meet peak demand. Hydrogen ICE with CV transmission plus battery and electric motor, running in parallel.

Table 1
Property overview of hydrogen storage systems considered in this analysis

Storage medium	Pressurized H_2	Liquid H_2	Gas on solid adsorbents	Chemical storage	Reference	
					Gasoline	Diesel
Volumetric density (kg H_2/m^3)	23.2–39.4[a]	67.7	62[e]	73–125[b]	88	134
Gravimetric density (kg H_2/kg storage medium)	2–8.5[b]	6–13[c]	0–10[a, b]	1.3–14[b]	12.6	14
Storage tank weight[f] (kg)	134–29	43–18	High–25	207–17	–	–
Typical storage pressure (MPa)	20–69	0.01–2	0.07–11.35	3–6	–	–
Typical storage temperature (K)	273–313	20.3	80–700	273–373	–	–
Leakage and boil-off (%/day)	–	0.3–0.5[d]	No leakage	No leakage	–	–
Energy required to free H_2 (MJ/kg H_2)	–	–	–	17.28[g]	–	–

[a]23.2 at 345 bar to 39.4 at 690 bar.

[b][19]—For pressurized storage two different types are distinguished; between 2 and 5 kg H_2/kg for pressurized storage in steel cylinders and between 7 and 8.5 kg H_2/kg for pressurized storage in composite material cylinders.

[c][19]—For storage and metal hydrides a wide range is given 2 (LaNi$_5$H$_7$) − 14 (LiBH$_4$ + 2H$_2$O)—for CNTs range between 0 and 3 kg H_2/kg.

[d][21] Boil of for a 50 m^3 storage dewar.

[e][22] Range given for single walled nanotubes (SWNTs).

[f]Storage tank weight is calculated using the gravimetric densities from the table and the minimum storage requirement of 2.73 kg H_2 to have a 350 km axiradius with a PEM fuel cell electromotor system with an fuel economy of 0.26 kW h/km.

[g]Storage tank weight is calculated using the gravimetric densities from the table and the minimum storage requirement of 2.73 kg H_2 to have a 350 km axiradius with a PEM fuel cell electromotor system with an fuel economy of 0.26 kW h/km.

on solid adsorbents—carbon nanotubes (CNTs)—and chemical storage—metal hydrides (MHs)—are assessed. These advanced storage options are included in the analysis, because of their favourable characteristics, such as high energy density (distinction made between volumetric[3] and gravimetric[4] storage density) and inherently safe way of storage [12]. The main challenge of MHs is their storage weight (discussed in more detail later in this paragraph). As apposed to choosing two actual storage media for the CNTs and the MH, generic characteristics representative for the advanced storage technologies are used in the analysis. The generic characteristics comprise typical pressure requirements at the refuelling station and the gravimetric and volumetric densities. The values are derived from literature ranges, presented in Table 1. The actual values used in the analysis are described further on in this paragraph.

Some additional characteristics are fuelling and storage temperature requirements, and information on the durability, which is important for the economic performance.

Pressurized hydrogen storage is most commonly used today. High pressure storage increases the energy density and is needed to meet the limited storage volume available in a vehicle. The high pressure, however, requires significant compression energy and subsequent investment and operation cost. In literature, a wide pressure range is given for pressurized storage between 345 (214 MJ/m^3) and 690 (364 MJ/m^3) bar [13]. In the analysis a storage pressure of maximum 700 bar is worked with, representing the upper end of the range.

Liquefaction of hydrogen increases the energy density by more than seven times compared to storage under 700 bar. The liquefaction process, however, is very energy consuming, influenced partly by the process scale, and can consume up to 40% of the energy content of the hydrogen [14]. Another challenge in hydrogen storage is the boil-off of hydrogen that makes long storage periods impossible.[5] The higher energy density is favourable for the volume and weight of the storage tank.

Hydrogen storage in gas on solid adsorbents presents a promising storage medium.

CNs may be a promising material for hydrogen storage, but experimental results to date have been controversial [15]. The quantity of hydrogen that can be stored per unit of volume and mass is the most important aspect for hydrogen storage. Because some experimental results concerning hydrogen storage have not been reproduced, the current status therefore is a bit unclear. For this analysis CNTs are chosen. Other gas on solid adsorbtion media include, glass microspheres and buckyballs, etc. Because of the extensiveness of different media, single walled, multiwalled, etc., and storage properties we do not present a discussion on a preferred option. Rather the aim of this study is to identify a range of characteristics on the one hand (Table 1) and choose a representative set of values from this range to execute the techo-economic analysis on the other hand.

The CNTs' storage capacity and related gravimetric density is high. Because the hydrogen is bond to the surface when stored it is expected to be inherently safe [6]. The pressure for filling is relatively low, providing an advantage compared to pressurized storage. In the analysis the pressure for filling is chosen at 75 bar, within the range 71 and 82.4 bar given by James et al. [10].

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[3] *Volumetric density*: The volumetric capacity is defined as the amount of gas adsorbed either in mass or in volume divided by the total volume occupied by the adsorbent and the adsorbed gas, i.e. the volume of the container.

[4] *Gravimetric density*: Defined as the weight percentage of the adsorbed gas to the total weight of the system, including the weight of the gas.

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[5] Due to the low temperature under which liquid hydrogen (LH$_2$) is stored and the temperature difference with ambient temperatures, the stored hydrogen will gradually boil. Because of the boiling, pressure builds up, this pressure is released by a high pressure vent. In the liquid storage tanks included in the analysis a boil-off rate of between 0.3% and 0.5%/day is used [20].
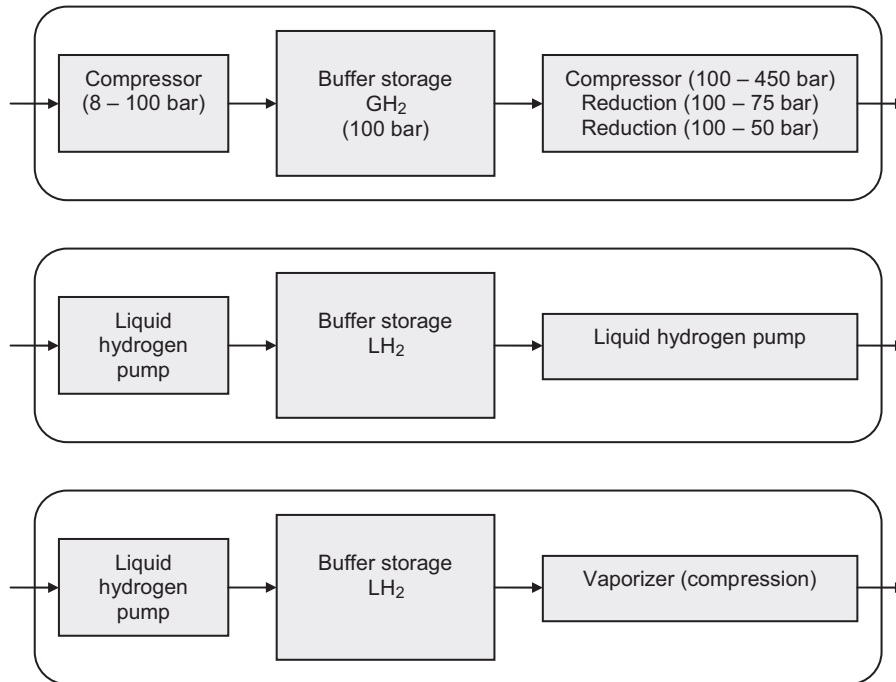
Fig. 2. Filling station configurations.

The fourth storage technology included in the analysis is a MH. A large number of metals and metal alloys can combine with hydrogen to form MHs. The hydrogen absorbing capacity is influenced by pressure and temperature. When hydrogen gas comes into contact with the metal (alloy) surface, often treated with a catalyst, the hydrogen gas dissociates in atomic hydrogen which diffuses into the metal grid. Heat is exhausted upon absorption and required to free hydrogen from the grid when desorbed [16]. Since heat is required to release the hydrogen it is assumed to be inherently safe [5]. The heat produced by the fuel cell or ICEV is used to dissociate hydrogen from the MH [17].

The absorption rate of MHs is depending on the pressure and temperature under which the MH is brought in contact with the hydrogen gas, with temperature and pressure being inversely related. The pressure range lies between low pressures of 21–23 bar[6] and highs of 80 bar [18], see Table 1. As a representative pressure 50 bar is assumed.

The effect of storage weight on fuel economy is not quantitatively taken into account in this analysis, as is done in some other studies (e.g. [19]). The impact of storage weight on the performance is, however, addressed in the discussion of the results, based on analysis of a comparable study.

3.2. Filling station configurations

The filling station dimensions are derived from the supply of a large conventional filling station. The station is assumed to serve 500 cars daily. The average car has an assumed

axiradius on one tank of 350 km. With an average fuel economy for a PEM fuel cell fuelled vehicle of 1150 kg of 0.94 MJ/km[7] (1.94 MJ/km for an ICEV) [22], following the standard European driving cycle the 'New European Driving Cycle' (NEDC) [35]. The average full tank holds 2.73 kg H_2. Multiplied by 500 cars a day, which is a typical throughput for a relatively large fuelling station, the total hydrogen demand per filling station is 1366 kg H_2 per day (or 4.6 TJ H_2 per year). The figure, of 2.73 kg H_2, is also used for the driving trains with lower engine efficiencies and hence higher demand.

Three filling station configurations are considered, shown in Fig. 2. The filling stations can be supplied by gaseous or LH_2. The filling stations have to dispense the hydrogen to the four assessed onboard storage media; high pressure storage (700 bar), MH (75 bar), CNTs (50 bar) and liquid.

The hydrogen from the transportation pipeline enters the filling station at 8 bar. First, a 160 kW reciprocating compressor compresses the hydrogen to 100 bar [23] buffer storage pressure. From the buffer storage pressure, the onboard storage systems can be supplied. For the high-pressure storage, 10 42 kW hydraulic booster compressors are needed to meet the 700 bar onboard storage pressure. For the MHs (75 bar) and CNTs (50 bar) the buffer storage pressure can be reduced, requiring expanders that do not need extra energy.

The hydrogen is delivered to the filling station in liquid form by trucks. From the truck the LH_2 is pumped into the (buffer)

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[6] Hydrogen absorption of nanocrystalline Na_2LiAlH_6 without catalyst at near-equilibrium conditions.

[7] The fuel economy figures are based on CONCAWE study [22]. Two aspects are influential for the fuel economy; the vehicle characteristics and the assessed driving cycle. All simulations are based on a common, "virtual" vehicle, representing a typical European compact size 5-seater sedan; weight is 1250 kg, comparable to e.g. Volkswagen Golf. The theoretical vehicle is used as a tool for comparing the various fuels and associated technologies.

storage at the filling station. To dispense the hydrogen to the vehicle, 10 1 kW pumps [24] are used. Since no pressure differences or other conditioning has to be realized at the filling station site only energy for the pumps is needed.

Hydrogen is supplied in liquid form to the filling station buffer storage. From the liquid buffer storage the hydrogen is vaporized and pressurized to meet the required pressure for the onboard storage system. Pressurizing is done under expansion and vaporization of the liquid requiring no extra compression energy.

3.3. Supply chain processes

3.3.1. Electricity production

The electricity production generation is assumed to come from the Dutch electricity mix 2020 [25]. All processes using electricity use this electricity mix, with an overall efficiency of 55.3%.[8] The unit price for delivered electricity is assumed 0.05 €/kW h[9] [26].

3.3.2. Natural gas extraction, processing and transport

The hydrogen production chains are based on a natural gas input. It is expected that in the year 2020 a large quantity of natural gas is imported from Russia [25] and subsequently the extraction and processing is carried out there. Important processing includes the removal of heavier hydrocarbons and contaminations such as H_2S. For the processing heat is required, provided by the burning of natural gas. The extraction and processing make use of electricity, considered to come from the Dutch electricity mix.[10] The natural gas is transported and imported from Russia through a distribution pipeline. The transport over a distance of 7000 km has an overall efficiency of 97.7%, the efficiency losses are primarily due to leakage,[11] in the piping and compressor stations and the work done by natural gas fuelled compressors. From the Dutch border a 250 km pipeline transports the natural gas to the Rotterdam harbour area in the Netherlands. For transport compression, work is required delivered by a natural gas fuelled compressor with an efficiency of 33%.[12] In the pipeline transport some losses occur due to leakages. The natural gas cost are considered to be a result of the global market of oil and gas trading, not related to

specific supply chains and related process. For the calculations the unit cost for natural gas is assumed €3.35/GJ[13] [26].

3.3.3. Hydrogen production and liquefaction

Large scale hydrogen production takes place centralized, for example in the Rotterdam harbour area, using a large steam methane reformer[14,15] (SMR) of 840 MW. For hydrogen purification a pressure swing adsorption (PSA) unit is integrated to obtain a purity of 99.999% H_2, required in the PEM fuel cell systems. The overall energy efficiency of large-scale hydrogen production (Linde) is 73%. The investment costs are presented in Table 2.

Small scale hydrogen production on the filling station (onsite) make use of a small scale steam reformer of 2.7 MW with an efficiency of 69% (Haldor Topsoe).

Liquefaction of hydrogen is considered only for some chains. The liquefaction installation is assumed to be integrated in the hydrogen production facility, requiring no transport of the hydrogen gas from the production facility. A large-scale liquefaction plant of 450 MW of hydrogen throughput is used. The efficiency and related cost for the liquefaction process is especially sensitive to process scale, compared to production. The energy use for liquefaction is 0.35 MJ_e/MJ H_2 resulting in an efficiency of 61.2% on a LHV basis for centralized liquefaction. Electricity comes from the Netherlands electricity mix 2020. For onsite liquefaction this figure is down scaled [14], for a 1.7 MW plant to 0.63 MJ_e/MJ H_2, with an overall efficiency of 40.6%.

3.3.4. Hydrogen transport

The assessed case is representative for densely populated area's in western Europe. In this analysis it is assumed that the hydrogen is distributed over a distance of 100 km. For centrally produced hydrogen two modes of transportation are assessed; gaseous pipeline and liquid truck transport (Table 3). For the pipeline a large distribution pipeline, pressure 60 bar[16] over

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[8] 55.3% efficiency forecast for total electricity mix of the Netherlands for 2020 [25]. The relative shares for the different feedstocks are as follows; natural gas (84%), wind power (2%), biomass (7%), coal (hard) (5%), mineral oil (2%), hydro (0.05%). Wind power and hydro power are considered to have an efficiency of 100%.

[9] No distinction is made in the electricity unit price between small and large scale buyers, because of the difficulty to predict price differences on the basis of the amount purchased. In the sensitivity analysis, however, the effect of electricity unit price fluctuations on the total outcome is shown.

[10] Although this obviously does not reflect the actual situation this figure is used because no detailed Russian electricity mix is known per region. Furthermore, the Russian electricity use is considered of marginal impact on the overall assessment.

[11] Although some estimates of the methane emission during production and transport of natural gas within Russia and from Russia to the EU are substantial, Lelieveld et al. [27] conclude after extensive measurements that the average leakage percentage is 0.7% (range 0.4–1.6%).

[12] Defined as primary energy in to establish the required output of work.

[13] Electricity and natural gas prices supplied from the grid are based on prices for small industrial users in the EU which correspond to the scales used in the analysis in this paper [28].

[14] Steam methane reforming (SMR): The basic SMR process uses steam ($H_2O_{(g)}$) to heat natural gas to approximately 850 °C over a nickel catalyst bed, yielding a mixture of CO and H_2O. The mixture is then cooled and catalyzed with steam again to yield pure hydrogen and CO_2 [29].

[15] A CO_2 capture unit can be integrated in the installation. With an MDEA (methyl diethanolamine) after shift CO_2 unit the capture efficiency is 85%, with an MEA (monoethanolamine) after furnace CO_2 capture unit up to 90%. The cost for the SMR process equipped with such a capture unit is 17% higher compared to a process without a capture unit [30]. The IPCC puts the cost penalty of the integration of a CO_2 capture unit in an SMR process between 18% and 33% [31]. The conversion efficiency of an SMR process equipped with an MEA unit suffers 13%$_{point}$ (from 74% to 61%) and an MDEA unit 3%$_{point}$ (from 76% to 73%) [30].

[16] 60 bar is currently used in natural gas transport pipelines. The consideration is as follows. The realized throughput depends on the pipeline diameter and the pressure under which it is transported. At higher pressures than 60 bar a thicker pipeline wall is required, resulting in higher metal cost [32]. If, however, the diameter is getting larger the material cost rises as well and it becomes practically less efficient. Because it is a consideration between pressure, wall thickness, pipeline diameter and throughput the ideal properties do not depend on which gas is transported but on the relations between these properties. Hence an optimum is achieved at 60 bar.

Table 2
Cost overview of the hydrogen production and liquefaction technologies considered

Technology	Parameter	Unit	Value	Reference
Small (onsite) reformer	Capacity	N m^3/h	1000 (=2.7 MW)	[25]
	Capacity factor	h/h	90%	
	Lifetime	years	20	
	Cost			
	Capital cost	€	2,800,000[i]	–
	O&M cost	€/a	175,000	–
	Feedstock			
	Natural gas	MJ/MJ H$_2$	1.31[a]	–
	Electricity	MJ$_e$/MJ H$_2$	0.02[b]	–
Large (central) reformer	Capacity	N m^3/h	150, 000 (=450 MW)	[25]
	Capacity factor	h/h	90%	
	Lifetime	years	20	
	Cost			
	Capital cost	€	140,000,000[i]	–
	O&M cost	€/a	700,000	–
	Feedstock			
	Natural gas	MJ/MJ H$_2$	1.35[c]	–
	Electricity	MJ$_e$/MJ H$_2$	0.01[d]	–
	Steam (output)	MJ$_{th}$/MJ H$_2$	0.31[e]	
Large centralized liquefier	Capacity	ton/h	13.6 (=450 MW)	[25]
	Capacity factor	h/h	90%	
	Lifetime	years	20	–
	Cost			
	Capital cost	€	180,000,000[g,i]	–
	O&M cost	€/kg H$_2$	0.10	–
	Feedstock			
	Electricity	MJ$_e$/MJ H$_2$	4.70[f]	–
Small on-site liquefier	Capacity	kg/h	60 (2 MW)	[25]
	Capacity factor	h/h	90%	
	Lifetime	years	20	
	Cost			
	Capital cost	€	4,504,109[h,i]	
	O&M cost	€/kg H$_2$	0.10	
	Feedstock			
	Electricity	MJ$_e$/MJ H$_2$	4.70[f]	–

[a] 420 N m^3 NG/h per 1000 N m^3 H$_2$/h.

[b] 0.07 MW h$_e$ per 1000 N m^3 H$_2$/h.

[c] 65,000 N m^3 NG/h per 150 N m^3 H$_2$/h.

[d] 5 MW h$_e$ per 150,000 N m^3 H$_2$/h.

[e] The steam output is a useful energy output and is beneficial to the overall efficiency. In terms of economics this stream is not included. 140 MW$_{th}$ per 150,000 N m^3 H$_2$/h.

[f] 1 kW h$_e$/L H$_2$.

[g] $_{,98}$ converted to €$_{,05}$.

[h] Capital cost downscaled using large liquefier data and scale factor (0.68) for general equipment assumed: $Cost_{new} = Cost_{known}(Scale_{new}/Scale_{known})^R$.

[i] The annuity factor is calculated based on the *after tax real rate of return* ($r = 10\%$), the *inflation rate* ($i = 2\%$) and the specific *lifetime* (n) of the installation considered. The lifetime is the economic lifetime. With $d = r(2i + r)$ annuity_factor $= d/1 - (1/(1 + d)^n)$.

100 km and a diameter of 50 cm is considered [26]. From the main transmission pipeline a medium pressure pipeline, pressure 8 bar, of 5 km transports and diameter of 7.5 cm the hydrogen to the filling station (buffer storage) [26]. Hydrogen-fuelled compressors, using hydrogen directly from the pipeline, with an efficiency of 30%,[17] deliver mechanical work. The mechanical work is needed to overcome friction losses.[18] No mechanical work is needed for the 5 km end delivery pipeline, since friction losses are compensated by the pressure difference.

As an alternative to gaseous hydrogen transport through pipelines, tube trailer transport can be used. To ensure comparability with pipeline distribution a transport distance of 100 km is assumed. The hydrogen is tanked into the tank trailer from the liquefaction (buffer) storage. The trailer stores approximately 50 m^3 LH$_2$ (i.e. 3389 kg H$_2$). Boil-off in a

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[17] Same efficiency as natural gas fired compressors assumed. The mechanical efficiency is assumed to be 90% the work is delivered by a hydrogen burned gas turbine or by electricity. The efficiency of 30% stated is the quotient between the work delivered and the primary feedstock input.

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[18] A simple pipeline configuration is assumed, consisting of a straight 100 km pipeline without ramifications.

Table 3
Cost factors for hydrogen pipeline transport

Technology	Parameter	Unit	Value	Reference
Large transmission pipeline	Length	km	100[a]	[31]
	Diameter	m	0.5[a]	
	Capacity	MN m^3/h	2.5[b]	
	Allocation factor	h/h	0.02%[c]	
	Lifetime	years	30[d]	–
	Cost			
	Capital cost per unit of length	€/km	1,790,000[e]	
	Total capital cost	€	3,580,000[f]	
Small delivery pipeline	Length	km	5[a]	[31]
	Diameter	m	0.075[a]	
	Capacity	kN m^3/h	6.7[b]	
	Lifetime	years	30[d]	–
	Cost			
	Capital cost per unit of length	€/km	347,550[e]	–
	Total capital cost	€	1,740,000[g]	

[a] Assumption.
[b] Calculation.
[c] Demand per filling station (1365 kg H_2/day, see calculation paragraph 2.2) divided by total capacity pipeline. This gives the proportion of the total pipeline capacity that is required for the actual demand on one filling station.
[d] Lifetime is assumed to be 30 years. This corresponds to the depreciation period more than to the actual pipeline lifetime.
[e] Calculation based on parker, 2003 total capital cost $= 4.82D - 2.48D^2$ (D is pipe diameter).
[f] Capital cost per unit length multiplied by the total length and by the allocation factor.
[g] Multiplication of the capital cost per kilometre and the length of the pipeline.

50 m^3 storage dewar is 0.4%/day (0.3–0.5%/day), half a day of storage in the truck is assumed, resulting in a boil-off of 0.2% per delivery. The truck is diesel fuelled on a 100 km (one way, 200 km return) driving cycle. The truck's weight equals 40 ton.

Concerning the cost for LH$_2$ truck transport the study of Amos [9] is used. Two factors are decisive for the cost, the quantity that is and the distance over which the hydrogen is transported. The study by Amos provides cost figures for a wide range of transport distances. The distance assessed in this paper, of 100 km, is not assessed. This cost figure therefore is obtained by linear interpolation of cost ranges for the 16 and 161 km distances. For a transport distance of 16 km the transport cost are estimated between the 0.24–1.60 $/GJ H$_2$ and for 161 km between 0.52–1.84 $/GJ H$_2$ [9]. By interpolation this becomes for a transport distance of 100 km, between the 0.40 and 1.74 $/GJ H$_2$. Per unit of mass this becomes 0.048–0.209 $/kg H$_2$.

3.3.5. Buffer storage

On the filling station site a buffer storage tank is required to meet fluctuations between demand and supply. Depending on the transported and dispensed form these can be gaseous and liquid storage tanks. The (buffer) storage dewars require space on the filling station site, cost additional energy because of compression energy needed and are subject to leakage. The leakage is minor due to limited dwelling time and a relatively large storage volume.[19] The dimensioning of buffer storage requires insight in the demand and supply fluctuations in time, and most importantly peak demand in these fluctuations.

[19] Boil off increases with a high surface to volume ratio. This ratio decreases with increasing tank size. Since the tank are relatively large, boil off is limited.

From the delivery pipeline the pressure (8 bar) is raised by a hydrogen compressor to 100 bar under which it is stored. Storage pressure is relatively low in the buffer storage, to limit energy losses due to pressure reduction for the filling pressures for MHs and CNTs. Under low-pressure conditions, required storage volume is high. A typical dimension for a one day buffer supply, for the case filling station, is 340 m^3 (1365 kg H_2 at 100 bar) demand. It is assumed that no gaseous storage facility is required because of the storage potential in the hydrogen gas supply infrastructure. For liquid storage a storage volume of one days' filling station throughput is assumed of about 21 m^3, which follows from the daily throughput calculation made in paragraph 3.2.

3.3.6. Reference scenario

For comparison a reference scenario is calculated, derived from the CONCAWE [22] study and a study by Hamelinck [33]. Three types of reference vehicles are included in the assessment; a gasoline ICE, a diesel ICE and a sugarcane ethanol ICE for the reference year 2010 [22]. The gasoline and diesel ICE chains enable comparison with the current (or near future) status of conventional propulsion technologies and fuels. The sugarcane ethanol reference is a representative cost and primary energy use bio-fuel with an average performance and therefore enables (indicative) comparison between the assessed chains and bio-fuels.

The total energy consumption from well-to-wheel is the sum of the well-to-tank pathway for gasoline and for the tank-to-wheel a fuel consumption. The well-to-tank energy consumption is the same for all three vehicle types and fuel options considered with 0.135 MJ$_p$/MJ$_f$. The tank-to-wheel energy consumption differs with 1.90 MJ$_f$/km for the gasoline ICE and

1.62 for the gasoline and bio-ethanol ICE hybrid option. The total for gasoline ICE is 2.16 MJ_p/km and for the gasoline ICE hybrid 1.76 MJ_p/km.

4. Results and discussion

4.1. Energetic performance: well-to-tank and well-to-wheel primary energy use

In Fig. 3, the energy use for fuel delivery (MJ_p/MJ_{H_2}) and for driving energy (MJ_p/km) are presented. The *fuel deliverance energy use* comprises the energy use of the supply chain from natural gas extraction, processing, distribution and hydrogen production (light grey) to, in some cases, liquefaction (darker grey), hydrogen distribution (black) and finally dispensing on the filling station to the actual storage in the onboard storage medium (white). Every stacked bar column represents the energy use, divided per process step in the chain, and per onboard storage technology (with LH_2 = liquid cylinder storage, CGH_2 = compressed gaseous hydrogen storage, MH storage and CNTs storage). The columns shown represent the result for the best performing option car transmission system (FCV hybrid), with the variation bars indicating the performance range, including all other options; FCV, ICE hybrid and ICE, the latter having the lowest efficiency. The stacked bar columns present the primary energy use for the well-to-tank and well-to-wheel. The variation bars only indicate the range that applies to the driving energy use (right) axis.

What stands out, in Fig. 3, is that the assessed chains are in the same order of magnitude as are the two fossil reference chains for diesel and gasoline. Centralized hydrogen production

and liquefaction outperforms onsite hydrogen production and liquefaction by 20% on primary energy input . The advantage of higher efficiencies for large scale processes apparently more than compensates the need for pipeline or over the road distribution. For the centralized case, the two advanced onboard storage technologies with 0.104 MJ_p/km perform 22% better as compared than to the conventional (compressed gaseous and liquid) onboard storage technologies with 0.127 MJ_p/km.

Concerning the distribution of primary energy consumption for driving, two processes consume over 80% of the energy inputs in all chains, the hydrogen production (light grey) and the liquefaction (darker grey). The energy use of hydrogen distribution (black) and the filling station handling (white) are minor contributing factors. Even the liquid truck transport, considered energy consuming, in the centralized scenario does only contribute just over 2% to the total. The filling station handling, in the centralized gaseous dispensed chains contributes to the total for 9% for the MH and CNTs and up to 21% for the compressed gaseous hydrogen storage.

4.2. Cost analysis: well-to-tank and well-to-wheel cost performance

In Fig. 4 the *fuel delivered and dispensed cost* (well-to-tank) and the *driving cost* (well-to-wheel) are presented. The *fuel delivered and dispensed cost*, shown in the left hand graph, comprise the cost for natural gas extraction, processing, distribution and hydrogen production (light grey), for liquefaction (white), the hydrogen distribution (darker grey) and the filling station dispensing (black). The driving cost, shown in the right hand
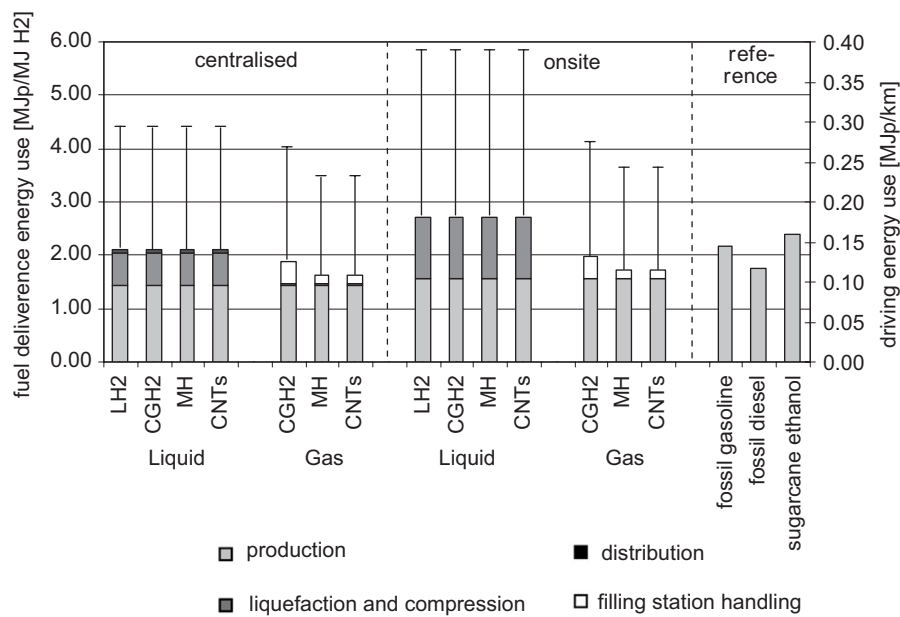

Fig. 3. Overview of the energetic performance. The figure shows the energetic performance of the 14 assessed chains and three reference chains for the well-to-tank (left axis) and the well-to-wheel (right axis). The bars indicate the difference between the best (PEM HFV hybrid) and worst performing (ICE FCV) option.
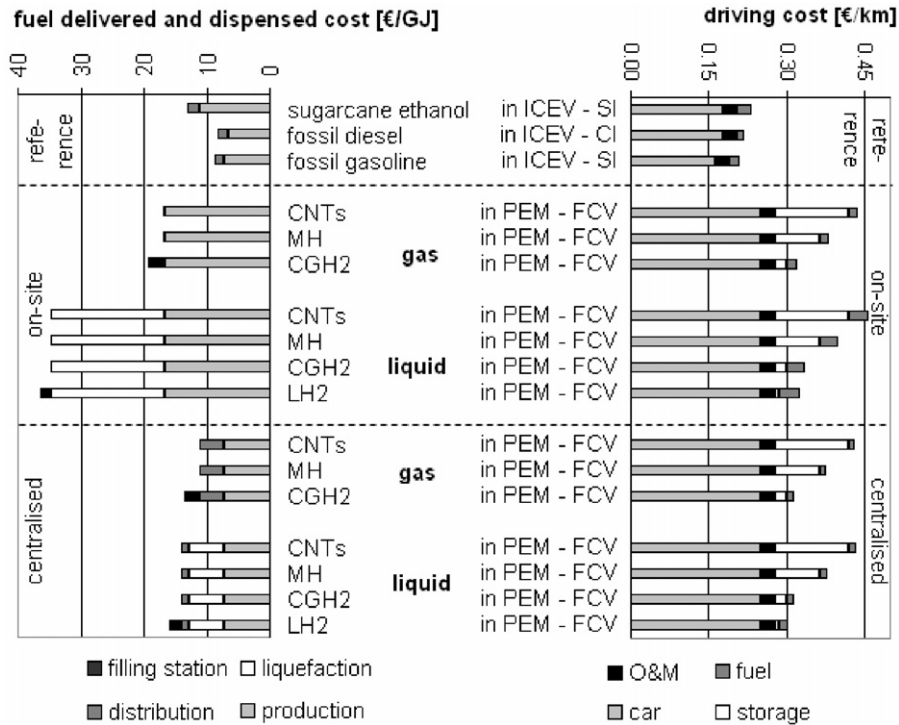
Fig. 4. Delivered and dispensed cost (left), and marginal driving cost (right) for the assessed chains and three reference combinations. The chains are divided in centralized and on-site produced and liquefied hydrogen. For all vehicles the following assumptions are made; average annual mileage 16,000 km, lifetime 10 years (economic depreciation period). PEM − FCV = proton exchange membrane − fuel cell vehicle. ICEV = internal combustion engine vehicle, SI = spark ignition and CI = compression ignition.

graph, comprise the cost for the car expenditure (light grey), the Operation and Maintenance (black), the hydrogen onboard storage medium cost (white) and the delivered and dispensed fuel cost (darker grey). The latter figure (darker grey in the right hand graph) is the total of the results in the left hand graph, hence clarifying the share of the well-to-tank cost share in the total driving cost.

Focussing on the well-to-tank (left hand axis) cost first, it shows that the cost for production and liquefaction of hydrogen on the filling station (onsite) are higher than centralized production and liquefaction. In the case of liquefaction, the cost is more than double onsite compared to centralized production and liquefaction. Furthermore, the fuel production for hydrogen is in all cases substantially higher than the two fossil and the bio-fuel reference cases. The distribution of the respective processes and their related cost, it is apparent that the production and the liquefaction incorporate the majority of the cost. The distribution cost are higher for pipeline distribution, with 28.5% of the least energy consuming chains, than are the distribution cost for liquid truck transport, comprising only 7.7% of the cost. For the filling station handling the high pressure storage does contribute to the total considerably, with 18.4% of the centralized pipeline distributed chain, especially due to the relatively high booster compressor cost.

The driving cost, shown in the right hand graph, shows the total well-to-tank cost as the fuel cost (darker grey), the storage cost (white), the operation and maintenance cost and the car expenditure. Car expenditure shows to be the decisive

cost factor in the driving cost of a car, whether conventional (ICE run on fossil or bio-fuels) or fuel cell driven. The fuel cell car costs are 41% higher than the compression ignition ICE and 54% than spark ignition car cost.

The focus of the study was the impact of onboard storage technologies on the performance. Concerning the economic performance, the two advanced onboard storage options appear to have high costs, of 8.7 €c/km for MH and 14.4 €c/km for CNTs, on top of the already higher FCV powered cars cost of 27.5 €c/km (including O&M cost). The fossil gasoline reference case the car expenditure cost, including O&M, are 18.5 €c/km. The well-to-wheel cost for the fuel only play a marginal role in the per kilometre cost, placing the main focus on the cars capital expenditure and storage material cost.

4.3. The PEM fuel cell and ICE hybrid

The effect of vehicle mass on the fuel economy can be of influence on the performance of the energetic and economic parameters. Because drive cycle modelling is beyond the scope of this assessment we limit ourselves to some reviewing comments on reference results on vehicle mass influence on the fuel economy. Of great importance in providing an estimate on the influence of vehicle weight on the fuel economy is the required axiradius and hence the amount of hydrogen that is to be stored onboard the vehicle. With the assumed axiradius requirement and related onboard storage capacity the effect on the fuel economy will be the highest for MHs with a negative impact of

about 2.9%[20] (from 0.104 to 0.107 MJ_p/km). The effect of energy required to free hydrogen from the storage medium only arises with the MH. For the performance it is important to assess if extra energy (heat) is required to free the hydrogen from the storage medium. The PEM fuel cell delivers a heat surplus during operation, whether this is enough for the adsorption of the MHs depends on the MH and its related desorption energy requirement. If a low or medium MH is assessed the surplus heat from the PEM fuel cell stack is sufficient for desorption of the hydrogen so that no extra energy is needed [10]. The characteristics chosen for the MH correspond to the low and medium MHs, hence the condition of desorption by surplus PEM fuel cell heat also applies to the researched case.

5. Conclusions

The focus of this research is to assess four onboard storage technologies with respect to their energetic and economic well-to-wheel performance and by doing so being able to identify the outlook for each assessed hydrogen onboard storage technology. The energy well-to-wheel performance of the assessed chains show results which are in the same order of magnitude of the fossil and bio-based reference cases, at least for the best performing PEM-FC hybrid engine technology. The energy performance range is within 0.10 and 0.20 MJ_p/km, the gasoline reference case at 0.14 MJ_p/km. This indicates that in terms of primary energy, the assessed hydrogen pathways do not show a significant advantage over fossil or bio-based fuel use. Although not specifically taken into account in the analysis; based on literature data an estimate of the environmental performance is made. From the European commission Concawe study [22] the greenhouse gas emission factors,[21] in grams of CO_2 equivalent per energy of fuel, give an indication for the environmental impact performance. From the energy analysis' results it is clear that pressurization of the hydrogen consumes the majority of the electricity, which is reflected in a high emission factor[22] of 98 g $CO_{2-eq.}$/MJ_{H_2}. For the liquefied hydro-

genchains this figure is, due to the extra energy consuming liquefaction process, even higher at 130 $CO_{2-eq.}$/MJ_{H_2}. Even if CCS is applied (on the gaseous chain) the emission factor amounts 35 $CO_{2-eq.}$/MJ_{H_2}. Compared to the fossil gasoline emission factor of 12.5 $CO_{2-eq.}$/MJ_{fuel} all hydrogen chains perform worse than the fossil Ref. [22]. Comparing the hydrogen storage technologies relative to each other, advanced storage technologies perform about 22% better on energy use than do conventional onboard storage technologies for the centralized chains and the gasoline reference chain. The least performing chains, using ICE non-hybrid drive train technology, perform poor on a (primary) mega joule per kilometre basis. Especially the chains using liquefied hydrogen use more than double the primary energy per kilometre than the reference chains. The advanced onboard storage technologies perform best, in the case of centralized hydrogen production even slightly better than the fossil and bio-based reference chains. An obvious advantage of hydrogen use as an energy carrier in transportation applications is the possibility of CO_2 capture at production. Although an energy penalty has to be paid for CO_2 capture, it can play a key role in mitigating transport related emissions, because over 85% of the emitted carbon dioxide can be captured [30].

Centralized produced hydrogen delivered to the fuelling station, with an average delivered energy use of 13.4 €/GJ, outperforms on-site production with an average of 27.7 €/GJ. Of the total cost, hydrogen production and liquefaction are by far the decisive cost factors. Hydrogen transportation, either in liquefied form by truck or gaseous by pipeline, only marginally contribute to the overall cost (Table 3). Indeed production, and when applicable, liquefaction account for more than half of the total cost on a €/GJ basis. In all but the centralized produced and pipeline distributed chains, the production and liquefaction account for more than 80% of the total hydrogen cost. In production and delivery this puts the focus on production rather than deliverance to improve cost performance. For the total driving cost, three other aspects are included in the analysis; car expenditure, operation & maintenance and, the focus of the paper, the storage tank cost. In the fossil and bio-based references cases car expenditure and O&M account for the bulk of the driving cost, with 16 €c/km for gasoline and 18 €c/km for diesel over 80% of the total cost per kilometre. For the assessed hydrogen chains car expenditure and O&M costs are higher compared to conventional ICEVS, because a higher cost for HFVs using PEM FC. Car expenditure for PEM FCVs is 25 €c/km with O&M expenses of 10% of the capital expenditure per year. Also the fuel production and deliverance of hydrogen is slightly higher than for the reference cases in the total driving cost. The production and deliverance, however, do not, with 2 €c/km, account for more than 10% of the total driving cost and in the most costly hydrogen chain do not account for more than 3%. The two advanced onboard storage technologies cost, contribute heavily to the total driving cost with 8.7 €c/km for MH and 14.4 €c/km for CNTs. The traditional hydrogen storage options, liquid and pressurized, contribute marginally to the total driving cost, with only 1 €c/km for gaseous and 2.3 €c/km for liquid storage. From this we conclude that at this stage advanced storage options lack the economic perspective,

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[20] The vehicle weight of the reference vehicle is 1150 kg. Required axiradius is 350 km, resulting in a required onboard storage capacity of 2.73 kg H_2. For a MH with a gravimetric density of 5% this is 54.6 kg MH rounded up to 60 kg for casing, etc. According to the influence of total vehicle weight on fuel economy by Allison and Cole [34] an increase of the vehicle weight of 10% has a 5.5% lower fuel economy.

[21] All emission factors are taken from the Concawe study [22]. Emission factors are estimated from the graphs and should therefore be considered estimates.

[22] The electricity production related emission factor is based on a European average derived resources mix. Consisting of the sources; nuclear (37.5%), Coal (22.4%), Oil (9.6%), Gas (15.5%), Hydro (12.4%), Wind (0.4%), Waste (1.8%) and other renewables (0.3%) [22]. In the calculations for the energy performance, the electricity resource mix for the Netherlands of 2020 is used differing in composition from the European mix as described here. The electricity mix of the Netherlands of 2020 has an average emission factor of 351 g CO_{2-eq}/kW h, compared to 430 g CO_{2-eq}/kW h used in the Concawe study for the European average. Although the European average has a large non-CO_2 production capacity (due to nuclear and hydro), the Netherlands mix has a lower emission factor, most notably because of the large share of natural gas. This on the one hand means one should exercise caution when comparing energy and emission results on the other hand it exemplifies how emissions depend on the used electricity fuel mix.

to develop into a competitive alternative to either the traditional hydrogen storage options or to the reference cases. The car expenditure and the O&M cost contribute to the total driving cost heavily, especially for PEM fuel cell driven vehicles. The advanced storage cost add a significant additional cost that, when compared to the total reference cost, can at present be a show-stopper. For the future it clearly emphasizes the importance of reducing cost of HFVs and advanced storage technology cost in order to make them a competitive alternative.

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