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New indicator for comparing the energy performance of CO_2 utilization concepts

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ABSTRACT

CO_2 utilization is increasingly considered a greenhouse gas abatement strategy alternatively to CO_2 storage. Existing indicators that assess the performance of CO_2 utilization options often provide an incomplete perspective and are unsuitable to compare different utilization options with different functionality (e.g. plastics and fuels). This study introduces a new performance indicator for CO_2 utilization options: Specific Primary Energy Consumption per unit of Fossil feedstock Replaced (SPECFER). This indicator, expressed in MJ/MJ, provides a proxy for the energy efficiency of which CO_2 conversion options can replace fossil feedstock required in conventional processes. Three CO_2 utilization case studies (CO_2 based methanol, polyols and dimethyl ether) are used to show the application and effectiveness of the SPECFER indicator. Among the case studies, only CO_2 conversion into polyol appears particularly efficient (SPECFER of 0.05 MJ/MJ), while the other options are not (SPECFER of > 1 MJ/MJ). The paper shows that the SPECFER indicator adds key insights compared to conventional indicators to the effectiveness of CO_2 utilization options and is a promising indicator complementary to CO_2 emissions reduction or life cycle greenhouse gas reduction potential. The SPECFER thus improves the understanding of the performance of CO_2 utilization and enables the possibility to distinctly compare different CO_2 converting utilization technologies.

1. Introduction

CO_2 capture and storage (CCS) is an important technology to effectively decrease greenhouse gas (GHG) emissions and mitigate climate change [1–3]. CO_2 capture and utilization (CCU) provides an additional option to CO_2 storage by aiming to use the captured CO_2 as feedstock in the production of goods (material and fuels). CCU options revolve around the direct use of CO_2 or its conversion into chemicals or materials. Examples of direct use of CO_2 include enhanced oil recovery (EOR) and direct application in e.g. the food industry [4]. CO_2 utilization options via conversion include biological conversion, mineralization and chemical conversion into chemicals, fuels or materials such as plastics [5,6]. Conversion of CO_2 requires a considerable amount of energy, due to the low thermodynamic (inert) level of the molecule. An overview of various CO_2 utilization options is presented in Fig. 1.

The concept of CO_2 utilization has been around for almost 30 years and was initially viewed upon as a promising technology alternatively to CCS (e.g. [8]). Since then, studies have indicated that the climate change reduction potential of CCU is limited compared to the potential of CCS [9], and CO_2 conversion technologies are therefore expected to play a minor role in climate change mitigation strategies [10]. However, research has also highlighted additional arguments for pursuing CCU, namely:

- To generate revenues that (partially) offset the cost of CCS [11,4,12,13].
- To reduce the net use of fossil feedstock [12,13].
- To introduce green energy (in the form of H_2 produced from renewable energy) in the fuel and chemical production chain [14–16,13].
- To stabilize electricity grids when considering a growing share of fluctuating renewable energy [17].
- To support the industry transition to a more circular (and biobased) economy [13].
- To cover the lack of geological storage potential for CCS in specific areas [18].
- To minimize public concerns regarding safety, viability and need for CO_2 storage in some jurisdictions [18].

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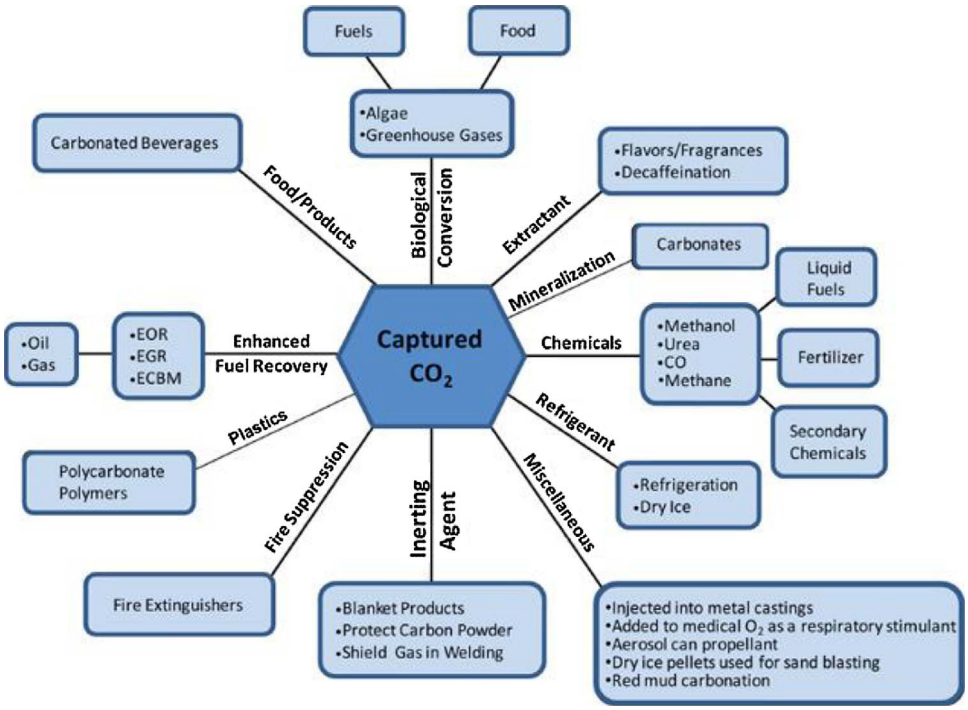

Fig. 1. Overview of various CO_2 utilization products [7].

These arguments have led to increasing interest in CCU in the recent years and its introduction as a key element in climate policy, such as in the 2016 European CO_2 abatement strategy [1]. Consequently, the term CCS is now often replaced with CO_2 capture, utilization and storage (CCUS). The increased importance of CO_2 utilization technologies in CCUS research is also highlighted by significant amount of funding that has been made available in the last five years. An example is the $6.7 million federal funding reserved by the U.S. Department of Energy (DOE) to develop CCU technologies as part of their Carbon Storage program [19] focussing on projects that will develop CCU technologies that reduce costs without generating additional GHG emissions. Another example is the funding of approximately € 100 million by the German government between 2010 and 2016 which has enabled several CCU research and development projects the last couple of years [17].

There are several options to evaluate the performance of CCU systems. Indicators that are most used are the CO_2 conversion efficiency, the energy consumption of the CO_2 utilization process [4], and climate change reduction potential [9,4,20]. The CO_2 conversion efficiency and energy consumption are used to evaluate the technical feasibility of CO_2 utilization options, but do not take into account the climate change reduction potential of these options. Determining the potential impact on climate change is crucial in the current discussion around the role of CCU [10]. Life cycle assessment (LCA) is considered best suited to assess the climate change reduction potential of CCU including the effects of all the stages in the life cycle [9,18]. GHG emissions over the total life cycle are often used as a measure for the environmental performance of CCU technologies [9,4,20]. By including the GHG emissions of material and energy inputs to the process, life cycle GHG emissions can be used to compare the climate change reduction potential of CCU alternatives with respect to CCS [9,20].

The usefulness of the indicators listed above to compare the performance of different utilization options is however limited. The CO_2 conversion rate and energy consumption are important to assess the technical feasibility of a CCU option, however, they are not effective in comparing CCU options with different functionality (e.g. fuels vs. materials). Using life cycle GHG emissions as performance indicator to compare different CCU options is challenging, firstly, because system boundaries generally differ among the options. Secondly, the user phase of the CO_2 based product and corresponding end of life CO_2 emissions are often not included. Thirdly, CCU stores the CO_2 in the product for a limited period of time (varying from days or weeks in the case of fuels to years in the case of materials) in most cases, with the exception of options such as mineralization and EOR. The temporal nature of CO_2 storage in CO_2 utilization products makes determining the impact on climate change caused by net emission reduction difficult [18]. Finally, the potential impact on climate change strongly depends on displacement effects (for example, whether the CCU product replaces conventional production or competes with novel renewable production). Besides, using climate change mitigation as performance indicator also provides an incomplete perspective as climate change mitigation is not the main target of CCU. The concept of CO_2 utilization is that the CO_2 used in the utilization process can replace fossil based feedstock used in a conventional production process. This concept is so far insufficiently included in the available performance indicators.

The above discussion implies there is a need for a CO_2 utilization performance indicator that provides better understanding of, and allows comparison of, the performance CO_2 utilization options. Such an indicator should:

- Relate the effectiveness of replacing fossil feedstock with the energy consumption of converting the CO_2.
- Be applicable independent of the CO_2 based end product.
- Add additional insight into the performance of CO_2 utilization compared to indicators currently used in literature, such as CO_2 conversion efficiency and life cycle GHG emissions.
- Allow a comprehensive comparison of different CO_2 conversion options that produce different end-products.

This paper aims to introduce a new performance indicator for CCU technologies that meets the requirements previously listed and allows a comprehensive assessment and comparison of the vast range of CO_2 conversion options. The applicability of the indicator is showcased by applying it to three different CO_2 utilization case studies.

2. Methodology

2.1. Scope and definitions

Two quantities that can enable a comparison of CO_2 utilization options are the energy required to convert the CO_2 into the product and the fossil feedstock that is actually replaced by the CO_2. To allow comparison of different types of energy and forms of fossil feedstock, primary energy and primary fossil feedstock are a suitable starting point. Primary energy is the energy found in its original or natural form that has not been subjugated to any conversion process [21]. Primary fossil feedstock is defined as fossil feedstock in its original form found on earth, such as crude oil, natural gas or coal, that are contained in the earth's subsurface.

2.2. SPECFER indicator

The Specific Primary Energy Consumption per unit of Fossil feedstock Replaced (SPECFER) indicator combines information on the additional energy use of CO_2 utilization with the fossil feedstock that is replaced. The SPECFER indicator quantifies the efficiency of a CO_2 converting utilization process by relating the amount of primary energy that the process requires to replace a given amount of fossil based feedstock, following Eq. (1).

To calculate the fossil feedstock replaced, the utilization process needs to be compared with a reference process that produces an equal amount of the final product. This is schematically presented in Fig. 2, where (part of) the carbon content of CO_2 is utilized to manufacture the final product, thereby reducing the amount of fossil feedstock required, while additional energy input is needed for the CO_2 capture and conversion processes. The additional energy consumption and feedstock replaced can therefore be expressed as the difference between the utilization case and a reference case (Eq. (1)):

$$SPECFER = \frac{Additional\ Primary\ \ Energy\ Consumption}{Primary\ Fossil\ Feedstock\ Replaced} = \frac{\Delta E_{primary}}{\Delta F_{primary}} = \frac{\Delta E_{primary}(utilization) - \Delta E_{primary}(REF)}{\Delta F_{primary}(REF) - \Delta F_{primary}(utilization)} \quad (1)$$

Primary energy and primary fossil feedstock are used in the SPECFER calculation to enable comparison of several processes with different energy and fossil feedstock inputs. The method used to convert energy inputs into primary energy is presented in Section 2.3. The primary fossil feedstock needed for the fossil based feedstock used in a process includes all fossil inputs during the extraction, processing and transport of the feedstock. The fossil based feedstock is the form in which the feedstock is eventually used in the process under study. The method used to calculate the total primary fossil feedstock replaced is explained in Section 2.4.

2.3. Primary energy

Energy carriers, such as heat, electricity and fuels, are converted to their primary energy equivalents. This allows comparison between, and summation of, different energy carriers. In the SPECFER (see Eq. (1)), the total additional primary energy required, $\Delta E_{primary}$, is calculated using Eq. (2):

$$\Delta E_{primary} = \sum E_{primary}(Utilization) - \sum E_{primary}(REF) = \sum_i E_i{*}p_i - \sum_j E_j{*}p_j \quad (2)$$

in which, E_i is energy input i (MJ) in the utilization case, p_i the primary energy conversion factor of energy input i, E_j is energy input j (MJ) in the reference case and p_j the primary energy conversion factor of energy input j.

Fossil fuels can be used for energy generation and for non-energy purposes. Non-energy use includes the consumption of fossil fuels as feedstock in the chemical industry (e.g. the use of naphtha for olefin production) and the consumption of refinery products, coke oven products and other solid carbon for non-energy purposes (e.g. the use of lubricants for transportation) [22]. When fossil fuels are converted into a different product, it depends on the use of the final product whether the fossil fuels are used for energy or non-energy purposes. Converting fossil fuels into other fuels is an example of energy use of fossil fuels [23]. The primary energy use in the SPECFER indicator only includes energy use of energy carriers.

There are two methods that can be used to calculate the primary energy equivalent of energy carriers: the partial substitution method and the physical energy content method. In the partial substitution

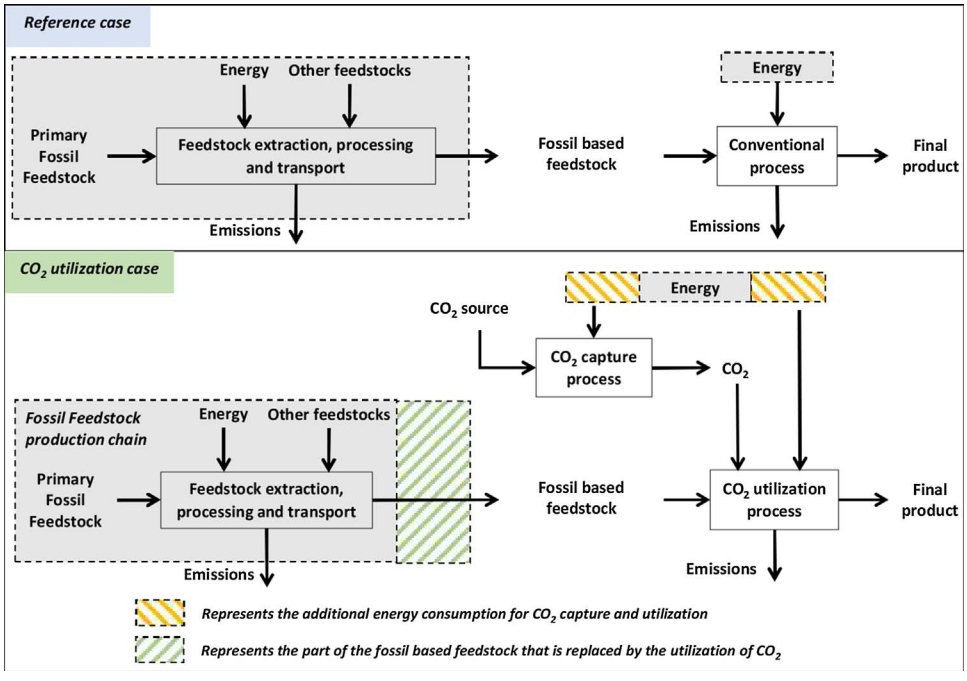

Fig. 2. Schematic overview of CO_2 utilization system (bottom) and a reference system (top) producing the same product. The fossil feedstock production chain is vastly simplified in the figure: in reality the fossil feedstock production chain consists of multiple fossil feedstocks, energy inputs and feedstock processing steps, depending on the actual feedstock used in the process.

method, the primary energy equivalent of electricity is represented by the energy amount that would be consumed to generate an equal amount of electricity in a conventional thermal power plant.[1] The physical energy content method calculates the physical energy content of the primary energy source of the energy carrier (retracing what type of primary energy was used to produce the energy carrier). The physical energy content method is considered to be the more appropriate method and is used in most international organisations [24,25] and is therefore recommended to use for SPECFER calculations. The primary energy conversion factors in Eq. (2) can be obtained from system analysis or extracted from literature sources.

2.4. Primary fossil feedstock replaced

The primary fossil feedstock replaced is the difference between the total primary fossil feedstock used in the reference case and the total primary fossil feedstock used in the utilization case (Eq. (2)). The type of fossil based feedstock can differ between the utilization case and the reference case and between different utilization cases. The total primary fossil feedstock definition includes all the fossil fuel used for energy and feedstock during the processing of the fossil based feedstock up to the system boundary of the process under study (see Fig. 2). As such, the primary fossil feedstock replaced includes both energy and non-energy use. The total primary feedstock replaced does not depend on the use of the final product, enabling the comparison of the performance of CCU options with different functionalities (e.g. fuels vs. plastics).

The primary fossil feedstock can be calculated using the mass and energy balance of the feedstock production chain by summing the fossil based feedstock and fuel inputs of all the processing steps. In this study, the primary fossil feedstock replaced, $\Delta F_{primary}$, is calculated by converting all the fossil based inputs of the utilization case and reference case into their primary fossil feedstock equivalents (in $MJ_{primary}$), following Eq. (3):

$$\begin{aligned}\Delta F_{primary} &= \sum \textit{Primary Fossil Feedstock use (REF)} \\ &\quad - \sum \textit{Primary Fossil Feedstock use (Utilization)} \\ &= \sum_j F_j{*}q_j - \sum_i F_i{*}q_i \end{aligned} \tag{3}$$

in which F_j is the amount of feedstock j (kg) in the reference case, q_j is the primary fossil feedstock required for the production of feedstock j (MJ/kg), F_i is the amount of feedstock i (kg) in the utilization case and q_i is the primary fossil feedstock required for the production of feedstock i (MJ/kg).

The amount of primary fossil feedstock required for the production of fossil based feedstocks (for example, the amount of crude needed to produce an amount of gasoline) strongly depends on the feedstock production process parameters, such as the scale of the process, the production method and the efficiency of the process. Theoretically, if such process conditions are known, the required primary fossil feedstock can be calculated for all fossil based materials. However, including all underlying processes is a time consuming exercise and potential data gaps or uncertainties can make these calculations challenging. Alternatively, other sources can be consulted to estimate the amount of primary fossil feedstock embedded in the fossil based process input materials.

An indicative way to convert fossil based feedstocks into their primary fossil feedstock equivalents is to extract cumulative energy demand (CED) values for fossil based materials from life cycle databases, such as the CED contained in the Ecoinvent database [26]. The CED measures the primary energy use throughout the life cycle of a good or a service [27]. It accounts for all primary energy withdrawn from nature, including direct energy use, indirect energy use and the energy content of feedstocks [28,29]. As such, the CED is a good proxy for the primary fossil feedstock use of (fossil based) materials. The large amount of available processes in the Ecoinvent database makes it easy to obtain CED values for almost any fossil based material and ensures that data for different materials can be obtained consistently. Using the CED for primary fossil feedstock consumption allows the SPECFER to be used as efficiency indicator, as the SPECFER is then expressed in MJ/MJ: values > 1 indicate an inefficient conversion route while values < 1 can be considered promising.

Alternatively to the CED, fossil feedstock depletion values, based on ReCiPe midpoint methodology [30], could be used to convert the fossil based feedstocks to primary fossil feedstock equivalents. The ReCiPe fossil depletion value is the amount of fossil fuel extracted, based on the CED and lower heating value [31,32]. As fossil depletion value is expressed in kg oil eq., the SPECFER would then get the unit MJ/kg oil eq. Although both methods (CED and fossil fuel depletion) are suitable for SPECFER calculations, the CED is used in this study as expressing the SPECFER in MJ/MJ enables the direct use of the SPECFER as efficiency indicator.

2.5. Energy required for CO_2 capture

In most utilization cases, CO_2 is not available and some CO_2 capture, transport and/or compression processes are required. The energy required for these processes can be included as additional primary energy demand when calculating the SPECFER. Whether it should be included depends on how the CO_2 capture energy demand is allocated. Three different options of CO_2 capture energy allocation can be distinguished:

1. CO_2 is captured independently whether the captured CO_2 is stored or utilized. The CO_2 is seen as a waste product and considered to be free of environmental impacts from life cycle perspective. The required energy for the capture of CO_2 is fully allocated to the process where the CO_2 emissions are produced (e.g. power production, steel plant).
2. CO_2 is captured to drive the CO_2 utilization process and/or the CO_2 is considered a valuable product. The amount of CO_2 capture unit is determined by the market potential of the CO_2. The CO_2 is therefore not a waste but a (sub) product and is considered to have an environmental footprint. The energy required for its capture and purification is fully allocated to the CO_2 utilization process.
3. A hybrid option in which only part of the captured CO_2 is utilized, while the rest is for instance stored. In this case, one can follow a multi-product allocation where a fraction of the energy used for the CO_2 capture is allocated to the CO_2 flow used for utilization and the rest to the main process (e.g., the power production, steel plant). The part of the CO_2 that is utilized is then considered to be a (sub) product with an environmental product whereas the part of the CO_2 that is stored is again seen as a waste product free of environmental impacts.

It is important to be consistent in the SPECFER calculation, especially if different CO_2 utilization options are compared. If different options are assumed to utilize an equal amount of CO_2 from the same source, the relative SPECFER of the options are not influenced by how the capture energy is allocated. However, if CCU options of different sizes and/or with different CO_2 sources are considered, allocating the CO_2 capture energy to the utilization processes is recommended to ensure a fair comparison. When only part of the captured CO_2 is used for utilization, allocating a proportioned part of the CO_2 capture energy to the utilization process is recommended.

[1] This is generally applied to electricity generated by non-combustible energy resources, i.e. nuclear and non-bio renewables.

Table 1
Characteristics of example cases selected for testing the SPECFER.

Case	A [33]	B [34]	C [35]
CO_2 utilization product	Methanol	Polyols	Dimethyl Ether
CO_2 Source	Subcritical coal power plant	naphtha-based H_2 production unit at a refinery	natural gas-based H_2 production unit at a refinery
CO_2 capture method	Solvent (MEA)	Solvent (mixture of MDEA + piperazine)	Solvent (mixture of MDEA + piperazine)
CO_2 captured (kt/a)	704	552	330
CO_2 utilized (kt/a)	704	58	330
CO_2 stored (kt/a)	0	494	0
CO_2 utilized/CO_2 captured (%)	100	10.5	100
Fossil based feedstock replaced with CO_2	Natural Gas	Propylene oxide	Methanol

3. Application

3.1. Case studies

To illustrate the use of SPECFER, examples of three case studies obtained from literature are shown in this section. The case studies not only differ in the type of CO_2 utilization product, but also in the origin of the CO_2, the size of the system and the amount of CO_2 that is utilized. This showcases the applicability of SPECFER in different circumstances. The three cases are:

1 Case A – CO_2 hydrogenation into methanol using H_2 produced from renewable energy.
2 Case B – CO_2 utilization for polyol production.
3 Case C – CO_2 utilization for dimethyl ether (DME) production via dry reforming of methane.

An overview of the key characteristics of the cases is presented in Table 1.

3.1.1. Case a – CO_2 to methanol

Fig. 3 presents a simplified process overview of case A. CO_2 is captured from a coal-fired power plant via post-combustion capture using a MEA solvent. The CO_2 is then utilized via hydrogenation, based on the process reported by Van-Dal and Bouallou [33]. The hydrogen required for the hydrogenation of CO_2 is produced by electrolysis using renewable electricity from photo-voltaic solar panels. The utilized CO_2 replaces the natural gas used to produce conventional methanol in the reference case.

3.1.2. Case B – CO_2 to polyols

Fig. 4 presents the simplified process layout of case study B based on Fernandez-Dacosta et al. [34]. CO_2 is captured from a H_2 production unit at a refinery running on naphtha and utilized in a polyol production process. Only a fraction (ca. 10.5%) of the captured CO_2 can be utilized within the polyol production process, the remainder of the CO_2 is transported and stored underground. The utilized CO_2 replaces part of the energy intensive propylene oxide (PO) feedstock used for the polyol synthesis process. In the reference case, polyol is produced conventionally from PO.

3.1.3. Case C – CO_2 to dimethyl ether

Fig. 5 presents the simplified process layout of case C based on Schakel et al. [35]. CO_2 is captured from a hydrogen production unit at a refinery, and used for dry reforming of methane for the production of syngas. The syngas is then converted into dimethyl ether (DME) by direct synthesis. The utilized CO_2 replaces the methanol used for conventional production of DME in the reference case.

3.2. Primary energy input

The energy inputs for the CO_2 utilization processes were converted to primary energy inputs with the use of primary energy factors (see Section 2.3). The primary energy factors for the energy inputs used in the three cases are based on the physical energy content and are presented in Table 2. The primary energy conversion factors used in this study were obtained from the primary energy embedded values from Ecoinvent processes [26], except for the conversion value for electricity from PV. When electricity from PV is considered to be completely renewable, primary energy conversion factors of 1 or close to 1 are reported [37]. When building PV systems and infrastructure is accounted for, the primary energy conversion factor of PV is considered 1.25 [38]. In this study, the value of 1.25 was selected as primary energy factor for PV to ensure a conservative estimation of the primary energy consumption of electricity from PV.

In this study, the CO_2 capture energy of the CO_2 entering the utilization process was included in all cases to ensure a fair comparison. In case B, in which only a fraction of the captured CO_2 is utilized, mass based allocation was applied to calculate the CO_2 capture energy assigned to the amount of CO_2 utilized. Table 3 presents an overview of the energy inputs of the CO_2 utilization process and the reference case and the corresponding total primary energy of the selected case studies. The total primary energy (both including and excluding the CO_2 capture energy) was calculated using the primary energy conversion

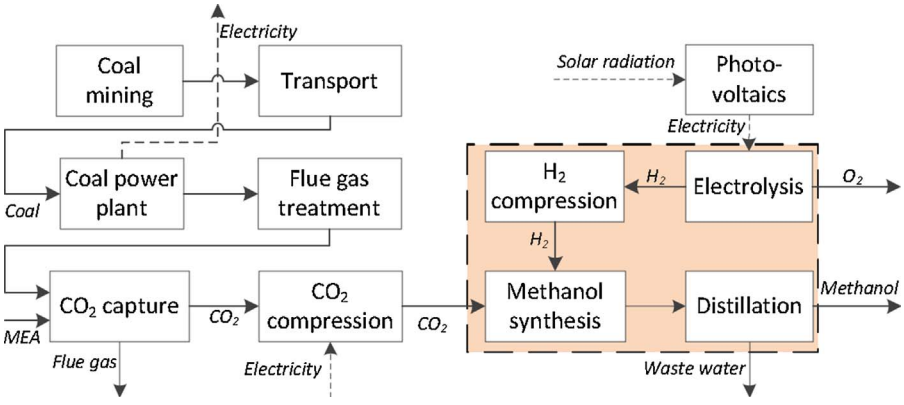

Fig. 3. Simplified process layout of case A: CO_2 hydrogenation into methanol. The coloured area represents the CO_2 utilization part of the process.

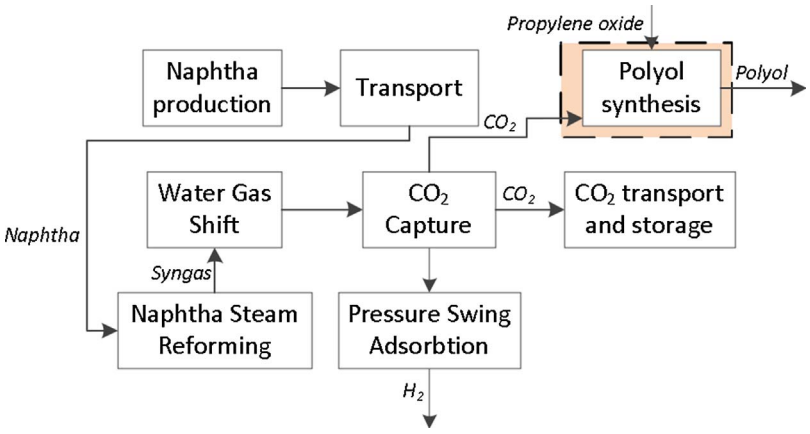

Fig. 4. Simplified process layout of case B: CO_2 utilization for polyol production. The coloured area represents the CO_2 utilization part of the process.

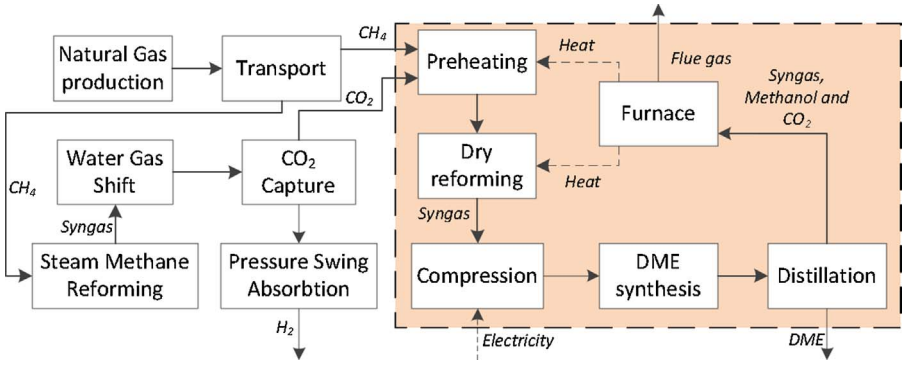

Fig. 5. Simplified process layout of case C: the production of DME via dry reforming of methane [35]. The coloured area represents the CO_2 utilization part of the process.

Table 2
Overview of primary energy conversion factors for various energy carriers.

Energy inputs	Primary energy conversion factors reported in literature	Selected primary energy conversion factor in this study
Natural Gas	1.02–1.25[a,b]	1.04[b]
Naphtha	1.08–1.24[a,b]	1.08[b]
Electricity (generic, Europe)	2.49–2.92[b,c]	2.55[b]
Electricity from Photovoltaics (PV)	1.00–1.25[c,d]	1.25[d]
Steam	1.13[b]	1.13[b]

[a] [36].
[b] Primary Energy embedded (MJ/MJ) [26].
[c] [37].
[d] [38].

factors (Table 2).

In case A, CO_2 is converted into methanol using H_2 produced from PV electricity.[2] The CO_2 capture energy includes the consumption of steam and electricity. Energy inputs for the CO_2 utilization process comprise electricity consumption during the process itself (mainly for H_2 compression), and PV electricity consumption for the production of H_2. The latter one is the most dominant energy input, accounting for 84% of the total primary energy consumption of the process, including CO_2 capture, of the utilization case. Energy inputs of conventional production of methanol comprise electricity and natural gas. In total, the CO_2 utilization process consumes 6.4 times more primary energy (7.1 if CO_2 capture energy is included) than the reference process.

In case B, the use of CO_2 instead of propylene oxide slightly increases the electricity and steam demand of the polyol production process. These additional energy inputs are however minor compared to the energy required for CO_2 capture of 3.18 MJ/kg CO_2 (0.74 MJ/kg polyol produced). In total, the CO_2 utilization process increases the primary energy demand with a factor of 1.9 (7.1 if CO_2 capture energy is included).

In case C, CO_2 is converted into DME via dry reforming of natural gas. Electricity is required predominately for the compression of syngas after the reforming step and natural gas is used to provide the heat for the dry reforming reaction. A small amount of electricity and some natural gas are energy inputs in the conventional DME production process. As such, the primary energy consumption of the utilization process is substantively exceeding the consumption of the reference process.

3.3. Primary fossil feedstock replaced

The primary fossil feedstock required for the fossil inputs in the various cases was calculated with the use of cumulative energy demand (CED) values obtained from the Ecoinvent database [26]. The CED values for the fossil feedstocks applicable to the cases in this study are presented in Table 4. The CED of methanol is smaller than that of natural gas, even though methanol is (conventionally) produced from natural gas, because the molar weight of methanol (32.0 g/mol) is larger than that of methane (16.0 g/mol): 1 mol of methane produces approximately 1 mol of methanol, so 1 kg of methane converts into approximately 2 kg of methanol. The rather high CED of propylene oxide is the result of the energy intensity of the feedstock.

Table 5 presents the fossil based feedstock and the corresponding total primary fossil feedstock of the CO_2 utilization process and reference case of the selected case studies. The total primary fossil feedstock was calculated using the CED values of the fossil based feedstocks (Table 4).

In case A, no fossil based feedstock is used in the utilization process. The reference case considers conventional production of methanol in which natural gas is used both as energy input and as fossil feedstock.

[2] It is not considered whether sufficient PV is available for the production of H_2.

Table 3
Energy inputs of the selected cases.

Energy inputs	CO_2 utilization	Reference case
Case A. CO_2 to methanol		
Steam CO_2 capture (MJ/kg CO_2)	3.19[a]	–
Electricity CO_2 capture (MJ/kg CO_2)	0.16[a]	–
Electricity methanol production (kWh/kg methanol)	0.36[a]	0.074[a]
PV electricity H_2 production (kWh/kg methanol)	10.88[a]	–
Natural gas methanol production (kg/kg methanol)	–	0.14[b]
Total primary energy excluding CO_2 capture (MJ/kg methanol)	52.28	8.20
Total primary energy including CO_2 capture (MJ/kg methanol)	58.24	8.20
Case B. CO_2 to polyols		
Steam CO_2 capture (MJ/kg CO_2)	1.92[a]	–
Electricity CO_2 capture (MJ/kg CO_2)	0.38[c]	–
Electricity polyol production (kWh/kg polyol)	0.014[c]	0.010[c]
Steam polyol production (MJ/kg polyol)	0.14[c]	0.05[c]
Total primary energy excluding CO_2 capture (MJ/kg polyol)	0.27	0.14
Total primary energy including CO_2 capture (MJ/kg polyol)	0.99	0.14
Case C. CO_2 to DME		
Steam CO_2 capture (MJ/kg CO_2)	2.19[d]	–
Electricity CO_2 capture (MJ/kg CO_2)	0.05[a]	
Natural gas DME production (kg/kg DME)	0.21[c]	–
Electricity DME production (MWh/kg DME)	0.81[a]	$1.13*10^{-3}$[e]
Total primary energy excluding CO_2 capture (MJ/kg DME)	18.9	0.01
Total primary energy including CO_2 capture (MJ/kg DME)	23.5	0.01

[a] [33].
[b] Based on global average methanol production process [26].
[c] [34].
[d] [39].
[e] [35].

Table 4
Cumulative energy demand for the fossil feedstocks considered in this study [26].

Feedstock	Cumulative energy demand (MJ/kg)
Natural gas	59.5
Methanol	36.1
Propylene oxide	114.0

Table 5
Fossil feedstock inputs of the selected cases.

Fossil based feedstock	CO_2 utilization	Reference case
Case A. CO_2 to methanol		
Natural gas methanol production (kg/kg methanol)	–	0.50[a]
Total primary fossil feedstock (MJ/kg methanol)	–	33.3
Case B. CO_2 to polyols		
Propylene oxide (kg/kg polyol)	0.81[a]	0.97[b]
Total primary fossil feedstock (MJ/kg polyol)	92.3	110.6
Case C. CO_2 to DME		
Natural gas (kg/kg DME)	0.43[a]	–
Methanol (kg/kg DME)	–	1.40[c]
Total primary fossil feedstock (MJ/kg DME)	28.6	50.5

[a] Based on global average methanol production process [26].
[b] [34].
[c] [35].

Table 6
CO_2 balance and SPECFER values for all cases under study.

		Case A (methanol)	Case B (polyol)	Case C (DME)
CO_2 used in utilization process (kg CO_2/kg product)		1.48	0.23	1.76
Net CO_2 conversion (%)		93.4	100	9.1
Net CO_2 utilized (kg CO_2/kg product)		1.38[a]	0.23	0.16
Δ Life cycle GHG emissions reduction (%)		Not reported	23[b]	8[c]
Δ Primary Energy (MJ/kg product)		57.6	0.85	24.4
Primary Fossil feedstock replaced (MJ/kg product)		33.3	18.2	21.9
SPECFER values				
SPECFER Including CO_2 capture energy	MJ/MJ	1.50	0.05	1.07
SPECFER excluding CO_2 capture energy	MJ/MJ	1.32	0.01	0.86

[a] Only the carbon content of the CO_2 is contained in the end-product (the oxygen is converted into water). As such, more than 1 kg of CO_2 is utilized for the production of 1 kg of methanol.
[b] Life cycle GHG emissions reduction of the entire system which includes the storage of 90% of the captured CO_2. However, slightly higher GHG emission reduction is reported for the part of the CO_2 that is utilized due to the replacing of propylene oxide (which has a very energy intensive production process) [34].
[c] [35].

All the fossil feedstock in the reference case is thus considered to be replaced in the utilization case. The captured CO_2 from the refinery in case B reduces the propylene oxide demand for the polyol production by 17%. In case C, conventional production of DME occurs via methanol dehydration, and methanol is considered the fossil feedstock replaced by the utilization case. The utilization process however uses natural gas as feedstock. In total, primary fossil feedstock consumption is reduced by 43%.

3.4. SPECFER

The change in primary energy consumption and the change in primary fossil feedstock consumption presented in the previous sections were used to calculate the SPECFER. Table 6 presents conventional performance indicators such as the CO_2 balance, CO_2 conversion rate and life cycle GHG emissions reduction, together with the total Δ Primary Energy, the primary fossil feedstock replaced and the calculated SPECFER values for all the cases considered in this study. Additional SPECFER values excluding the CO_2 capture energy and varying some of the key conversion factors are included to show the sensitivity to these parameters.

In Case A, the largest amount of CO_2 is utilized out of all the cases, mainly because there are no direct CO_2 emissions during the utilization process. On the other hand, the extensive energy inputs lead to the highest SPECFER in this case, indicating that this CO_2 conversion route is inefficient in terms of the amount of energy input needed to replace a unit of fossil feedstock. Approximately 50% more primary energy is used in this process than the avoided cumulative energy demand of the fossil feedstock that is replaced. Even when the CO_2 capture energy is not included, the SPECFER still clearly shows the energy inefficiency of this proposed utilization route. The high SPECFER value is dominated by the electricity consumption of the H_2 production. Although this electricity is produced from renewable PV, the selected primary energy factor of PV of 1.25 $MJ_{primary}$/MJ is still considerable despite being significantly lower than the selected primary energy factor of regular electricity (2.55 $MJ_{primary}$/MJ).

Case B clearly shows the lowest SPECFER value of 0.05 MJ/MJ of the options assessed in this study for two reasons: Firstly, the energy requirements of the utilization process are very low compared to the

other cases and compared to the CO_2 capture energy requirement. Secondly, the CED of the propylene oxide (114.0 MJ/kg) that is replaced is considerably higher than CEDs of the replaced fossil feedstocks in the other cases (i.e. 66.7 MJ/kg for natural gas and 36.1 MJ/kg for methanol). When the energy consumption of CO_2 capture is excluded, the SPECFER value would even be substantially lower (0.01 MJ/MJ).

In Case C the SPECFER is in between the values of case A and C, and the value of 1.07 MJ/MJ indicates that slightly more primary energy is used than CED avoided by the fossil feedstock replaced. The most important energy input in this case is the electricity used for the compression of syngas required for the direct DME synthesis. Grid electricity is assumed to be used in this process and a cleaner electricity production method with a lower corresponding primary energy factor could significantly lower the SPECFER value, similarly to case A. The CO_2 conversion and the amount of CO_2 utilized in the product are relatively small in this case, as a result of direct CO_2 formation (and emission) during the DME synthesis process. When CO_2 capture energy is excluded from this case, the SPECFER value would drop below 1 (0.86) MJ/MJ.

The results in Table 6 show that the SPECFER indicator complements conventional performance indicators regarding the effectiveness of a CO_2 utilization process: when only looking at the amount of CO_2 utilized and the CO_2 conversion efficiency, case A (CO_2 conversion into methanol) seems to be the most favourable of the investigated case studies. The application of the SPECFER however, shows that a case such as case A, in which a substantial amount of energy is required, is infeasible from the perspective of energy efficiency. Of the cases under study, only case B (CO_2 conversion into polyol) stands out as an effective CO_2 conversion and fossil feedstock replacement route.

4. Discussion

The Specific Primary Energy Consumption per unit of Fossil feedstock Replaced (SPECFER) was proposed as a new indicator to assess and compare the performance of CO_2 utilization options, and its application and usefulness using three examples was showcased. The indicator was set up to be comprehensive and easily applicable, but as a result also faces some limitations due to simplifications. Besides, the methods used in the indicator are subject to certain uncertainties that might affect the accuracy and reliability of the indicator. In this section, the most important limitations and uncertainties will be identified and discussed.

The selection of system boundaries plays an important role in the application of the SPECFER. Within the system boundaries (process under study), the use of fossil based material as feedstock and as energy input is clearly distinguished. The conversion of fossil based feedstock to primary fossil feedstock uses data regarding the fossil feedstock production chain, outside the system boundaries. In this chain, the use of intermediate fossil fuels (energy vs. non-energy) is not specified and all the fossil fuels used contribute to the total primary fossil feedstock consumption, regardless of their use. Therefore, it is important to select the system boundaries so that all energy inputs and fossil feedstock inputs of the processes under study are included and consistently identified.

The SPECFER indicator is an energy efficiency indicator not related to CO_2 emissions. As such, expected environmental performance (in terms of greenhouse gas reduction) improvement of using renewable forms of energy for the conversion of CO_2 are not addressed and the use of renewable forms of energy also contributes to the SPECFER. This is exemplified in case A (CO_2 into methanol), where the consumption of electricity from PV lead to the highest SPECFER value of the addressed case studies. One could argue that the use of renewable energy should not be taken into account as environmental consequences are limited compared to use of conventional forms of energy. However, as a society we need to decide how to use the available renewable energy most efficiently. Therefore, it is important to select processes which can improve energy efficiency, regardless of the source of energy, which is specifically what the SPECFER indicator does. Nevertheless, to generate a complete perspective of the performance of the CO_2 utilization process, it is recommended to always assess the SPECFER in combination with environmental performance indicators, such as climate change reduction potential.

A key limitation of the SPECFER indicator is that only direct energy and fossil feedstock inputs are included. Non-fossil based materials, such as catalysts, solvents and chemicals are excluded from the definition, and as such also their indirect energy consumption and/or fossil based feedstock use. As a result, the SPECFER of utilization cases that increase the use of non-fossil based materials compared to the reference cases might be optimistic with respect to the actual performance of these processes.

4.1. Uncertainty analysis

Uncertainties in SPECFER are directly associated with uncertainties in input parameters, the selection of reference cases and system boundaries. Performing an uncertainty analysis should therefore be part of the analysis to ensure a sound interpretation, communication, and use of SPECFER results. Aspects that play a key role in the SPECFER calculation and affect the uncertainty of the results are:

- Allocation of CO_2 capture energy to the utilization process.
- Mass and energy balance of the CO_2 utilization process.
- Mass and energy balance of the reference process.
- Primary energy conversion factors.
- CED factors of fossil based feedstocks.

With respect to allocation, the SPECFER results in Table 6 already showed that allocating (all or part of the) the CO_2 capture energy to the utilization process has a significant impact on the results. Including the CO_2 capture energy is recommended to ensure a fair comparison of different CO_2 utilization options. In cases were only part of the captured CO_2 is utilized, allocation based on the mass fraction (CO_2 utilized/CO_2 captured) is proposed.

The mass and energy balances of the CO_2 utilization and reference processes also play a large role, because they determine the amount of energy and fossil based feedstock that is used for the calculation of the SPECFER. The uncertainties associated with these values depend on the quality of the process model or literature data used to obtain these values. Those can be assessed using methods such as sensitivity analysis, but also using qualitative uncertainty assessment to cover weaknesses in e.g. data sources or methodologies (a good example is the use of pedigree analysis (see e.g. [40,41])

Variability in the applied primary energy and CED factors can also cause uncertainty in the SPECFER values. Although primary energy factors are extensively documented and narrow ranges are reported (see Table 2), values can strongly depend on the geographical location and temporal scope and be, in practice, located outside the reported ranges. The conversion of fossil based feedstock to primary fossil feedstock was done using cumulative energy demand values associated to the feedstocks used in this study. These values were taken from corresponding processes in the Ecoinvent life cycle database [26]. Although values were selected for the best matching processes, CED factors can easily change by up to 25% if alternative processes are selected.

A sensitivity analysis was performed to show the effect of variances in the above-listed aspects on the SPECFER results of the case studies. Variances of ± 25% were considered for the mass and energy inputs and the CED factors. The primary energy factors were varied according to the ranges reported in literature (Table 2). For case B, the sensitivity analysis included the effect of allocating the complete amount of the CO_2 capture energy to the utilization process, instead of the 10.5% of the capture energy that was originally included.

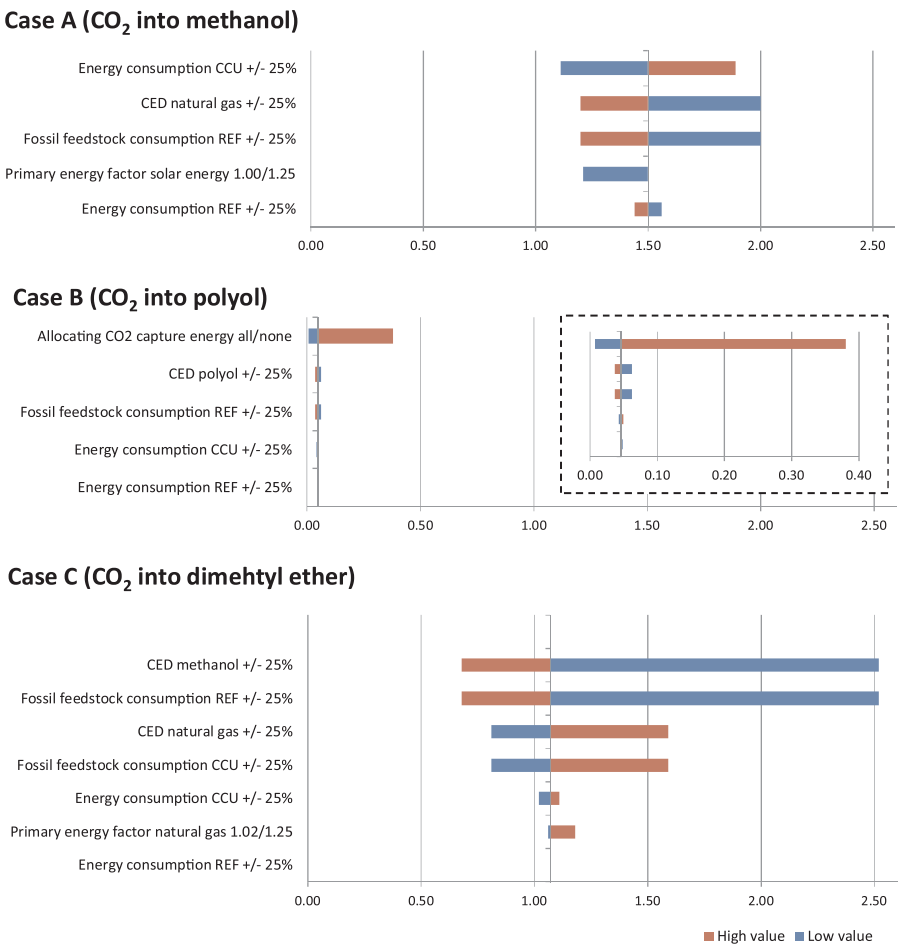

Fig. 6. Sensitivity analyses for the different cases. Tornado diagrams are shown for each case with the basic SPECFER value (y-axis) and the effect of modifying key parameters on the SPECFER values. The box within the graph of case B shows a zoom-in of the sensitivities of that case.

Fig. 6 presents the results of the sensitivity analysis using tornado diagrams, where the range in the SPECFER values is shown for each of the parameters varied. Note that for all cases, as expected, increasing the parameters that affect the primary energy or fossil feedstock consumption of the utilization process causes a rise in the SPECFER values, while increasing the parameters that affect the consumption of the reference case lowers the SPECFER values.

In case A, the SPECFER value is most sensitive to the energy consumption of the utilization process and the parameters that affect the primary fossil feedstock consumption of the reference case (fossil feedstock consumption REF and CED natural gas). Changes in these parameters do not lead to a SPECFER value lower than 1 MJ/MJ. On the other hand, SPECFER values of up to 2 MJ/MJ are reached when the primary fossil feedstock consumption of the reference case is reduced. The energy consumption of the reference case has limited effect on the SPECFER value, as the energy consumption of conventional methanol production is very low compared to the energy consumption of the utilization process.

In case B, the SPECFER value changes by up to 32% with a 25% variance in the energy or fossil feedstock consumption. The energy consumption of both the utilization process and the reference process is marginal compared to the CO_2 capture energy, which dominates the SPECFER result. This is also highlighted in The largest effect when varying the input parameters on the SPECFER, is shown in case C. The SPECFER is most sensitive to parameters that affect the fossil based feedstock consumption. A substantial amount of fossil feedstock (natural gas) is used in this CO_2 conversion process. Consequently, a 25% variation in fossil feedstock consumption in either the utilization case or reference case changes the amount of primary fossil feedstock replaced by more than 25%. A decrease in fossil feedstock consumption in the utilization process or an increase in the reference process could therefore lower the SPECFER to below 1 MJ/MJ. On the other hand, the SPECFER can be significantly increased to values exceeding 2.5 MJ/MJ as a result of lower fossil feedstock consumption in the reference case. Because the ranges of the SPECFER values of case C show overlap with the SPECFER ranges of case A, it is not possible to conclude whether case C performs better than case A, despite the significant difference in the base value. However, the sensitivity analysis supports the conclusion that case B is the most efficient utilization process among the three cases.

The sensitivity analysis was performed by making changes as one-at–the time and did not consider simultaneous changes in multiple parameters, which could result in a propagation of uncertainties in the SPECFER values and thereby lead to values that exceed the SPECFER ranges presented. Furthermore, the sensitivity analysis did not stress the importance of selecting and assessing the reference system. The selection of a proper reference system can be challenging, especially when it is not straightforward which products or processes are replaced by the CO_2 utilization option.

5. Conclusion

The assessment and comparison of the performance of CO_2 utilization technologies is a complex exercise due to the different system boundaries and functionality of the final CO_2-based products. Indicators that ore often used to assess the performance, such as the CO_2 conversion efficiency and reduction of greenhouse gas emissions, provide useful but limited insights and are ineffective in providing a comprehensive comparison of the performance of different utilization options.

In this paper, a new indicator, the Specific Primary Energy

Consumption per unit of Fossil feedstock Replaced (SPECFER) was introduced to assess and compare the performance of CO_2 conversion options. This indicator relates the additional energy consumption of CO_2 conversion processes with the amount of fossil feedstock that is avoided due to the use of CO_2, and can be used as a proxy for the efficiency of a CO_2 conversion technology. The key advantage of the SPECFER indicator is that it can be applied independently of the final CO_2-based product, and therefore can be used to compare the performance of utilization technologies with different end-products. These characteristics make the SPECFER a useful alternative to existing technical indicators such as CO_2 conversion efficiency.

The application of the SPECFER indicator was tested by examples of three different utilization cases. Results show that the indicator added the following insights to the performance of CO_2 utilization systems:

- A high CO_2 conversion efficiency does not necessarily improve the performance of a system. When a substantial additional energy amount is required to convert the CO_2, this is not always compensated by associated fossil feedstock savings.
- Examples of the selected cases show that only the performance of the small-scale application of CO_2 conversion into polyols was efficient. The energy inputs required to drive large-scale utilization processes converting CO_2 into dimethyl ether and methanol appear too significant compared to the actual fossil feedstock savings of these cases.
- The use of renewable energy, even though accompanied by much lower primary energy factors than fossil energy, can significantly contribute to the SPECFER and make a CO_2 conversion process inefficient. As energy efficiency indicator, the SPECFER can therefore help to identify what utilization processes are best suited to effectively use (intermittent) renewable energy.

The reliability of applying the SPECFER indicator, especially if different CO_2 utilization options are compared, strongly depends on how consistently the methodology is applied. It is particularly important to:

- Select correct reference systems.
- Apply system boundaries consistently.
- Include all energy and fossil based inputs.
- Correctly distinguish energy and fossil feedstock inputs and avoid double counting.
- Consistently allocate the energy required for the capture of CO_2 to the utilization process.

Even when the methodology is correctly and consistently applied, uncertainties in the SPECFER results still exist. The conversion of energy inputs to primary energy consumption and fossil feedstock to primary fossil feedstock depend on the used primary energy factors and cumulative energy demand values. The associated uncertainties in these values can be significant, as in many cases average values, simplifications and/or proxies are used to calculate these figures. Therefore, it is recommended to include uncertainty assessment when conducting a comparative assessment following the SPECFER methodology.

The SPECFER indicator gains adds key insights into the energy efficiency of CO_2 utilization options that convert the CO_2 to products, but does not assess the effectiveness to abate greenhouse gas emissions of that option. Therefore, it is recommended to use the SPECFER in combination with a GHG reduction or climate change mitigation measure to assess the complete perspective of the performance of CO_2 utilization. The application of SPECFER can then improve the understanding of CO_2 conversion technologies and help to successfully compare technologies with different end-product functionality.

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