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CCUS scenarios for the cement industry: Is CO_2 utilization feasible?

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ABSTRACT

In this work, four illustrative CO_2 capture, utilization and storage chains are investigated in order to evaluate the economic feasibility of CCUS technologies in connection to the cement industry. A CCS reference chain in which 90% of the CO_2 emissions (or 0,694 $MtCO_2/y$) are stored in a saline aquifer is first studied. Due to emissions related to energy usage in the capture, conditioning and transport processes, a total of 0,504 $MtCO_2/y$ are avoided, or 65% of the CO_2 emitted by the cement plant at a cost of 114 €/t CO_2 avoided. Then, production of ethanol, polyols or food-grade CO_2 is integrated to the chain, composing three alternative CCUS chains. These products are chosen based on an assessment of market, energy demand, and technology readiness level of technologies. For CCUS, we show that the economic feasibility is case dependent. The cost of producing blue ethanol is estimated as 656 €/t, slightly above the market value of 633 €/t. The cost per tonne of CO_2 avoided drops from 114€ (CCS) to 111€ (sugarcane-based displacement) and the amount of CO_2 avoided increases by 3%, to 0,518 $MtCO_2/y$. In the second CCUS scenario, we have evaluated the integrated production of polyols. The entire CCUS chain avoids 0,708 $MtCO_2/y$, and produces 288 kt/y of polyols, generating a profit of 18 €/t CO_2 avoided. In the third CCUS scenario, we show that the production of food-grade CO_2 is feasible as long as it is used to replace fossil-derived CO_2, with a total CO_2 avoidance of 0,504 $MtCO_2/y$ at a cost of 108 €/t. A general conclusion from this work is that the average cement plant emits much more CO_2 than can be utilized in a single CO_2 utilization plant. That is either due to market constrains or limited availability of raw materials. For the routes evaluated in this work, the fraction of the emitted CO_2 directed to the utilization plant was always below 10%. Therefore, when connected to the cement industry, utilization is not likely to be applied as a stand-alone solution, but as an integrated link in the CCUS chain.

1. Introduction

Driven by the need to limit global warming, governments' commitment to reduce carbon footprint, and the need for value creation to support carbon capture, many novel carbon capture and utilization (CCU) technologies to convert CO_2 into fuels, minerals or chemicals have been reported. In a recent roadmap, the potential of CO_2 utilization adds up to a maximum of 7 Gt of CO_2 uptake per year by 2030 [1]. As the quantity of CO_2 uptake by CCU technologies is limited by the market [2], and given that the global energy-related CO_2 emissions are estimated at 33 Gt in 2021 [3], CCU options can only be complementary to CO_2 storage, in order to achieve a significant decarbonisation through carbon capture. In fact, the IEA projections for 2070 indicate that, even in the far future with technologies for advanced synthetic fuels and chemicals fully developed and largely applied, 8% of the captured CO_2 would be utilized, with the remaining 92% being geologically stored [4].

The cement industry is one of the major sources of CO_2, corresponding to about 6–7% of global anthropogenic emissions. About 60% of these emissions come from mineral decomposition ($CaCO_3$ to CaO), and the remainder is from fuel combustion. CO_2 is therefore an inevitable by-product of the process and, in order to significantly reduce the climate impact of cement production, carbon capture is unavoidable. As consequence, the IEA points to CO_2 capture and storage (CCS) as the major contributor to emission reductions in the cement industry (56% by 2050, with up to 920 Mt of CO_2 stored per year) to be deployed from 2020 [5]. The European Cement Association (CEMBUREAU) also attributes a prominent role to CCUS in decarbonizing the industry: 33% of the emissions would be avoided by CCUS (or 280 kg CO_2/t cement) by

Abbreviations: CO2-eq, CO_2 equivalent; DME, dimethylether; MEA, monoethanolamine; t, tonnes, or 10^6 g; kt, kiltonnes, or 10^9 g; Mt, million tonnes, or 10^{12} g; y, year.

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2050 [6].

The best CCUS option for each cement plant is dependent on the plant location, as the local market demands, waste heat availability within the plant, and local availability of geological storage sites, amongst other factors, will influence the economics of the CCUS chain. In order to evaluate the feasibility of CCUS technologies in connection to the cement industry, and to understand the interaction between utilization and storage, four illustrative CO_2 capture, utilization and storage (CCUS) chains are evaluated in this work.

In the framework of the H2020 CEMCAP project [7], a reference cement plant was defined based on the best available technique standard as defined in the European BREF-Document for the manufacture of cement. For this work, this hypothetical plant is considered to be located in Belgium.

The first chain, defined as a reference case, considers CO_2 capture using an amine scrubbing system and subsequent geological storage in a deep saline aquifer (CCS). For this reference chain, it is assumed that the CO_2 is transported to a storage formation on the Dutch Continental Shelf. When combining CO_2 utilization to geological storage, the product choice will impact the amount of CO_2 that can be utilized and the economic performances of the integrated chain. Within the current work, 16 possible CO_2 utilization pathways and products are evaluated from the perspective of the market size, technology readiness level and energy demand. Based on this assessment, three alternative CCUS chains were evaluated: making a fuel (ethanol), a polymer feedstock (polyol), and food-grade CO_2. The CCUS chains are represented in Fig. 1.

The paper is structured as follow. Section 2 presents the selection of the CO_2 utilisation technologies and products. In Section 3, the considered CCUS chains are described in detail together with the methodology for evaluation. Finally, Section 4 presents the results of the assessments, while the conclusions are given in Section 5.

2. Selection of CO_2 utilization products

A total of 16 CO_2 utilization products were considered in this work, divided into 4 categories: minerals, fuels, chemicals and polymers, and CO_2 (direct use without conversion). Minerals included ground and precipitated calcium carbonate (GCC and PCC), aggregates and carbonated concrete. Fuels included methanol, dimethyl ether (DME), methane, ethanol, isopropanol and biodiesel. Chemicals and polymers included polypropylene carbonate (PPC), polyols, cyclic carbonates, and formic acid. The possibilities of direct use of CO_2 in the food and beverage market (food-grade CO_2) and in greenhouses for enhanced vegetables growth were also considered.

For each product, three criteria were evaluated: technology readiness level (TRL), CO_2 uptake potential (connected to the market size), and energy demand. For most of the products considered, various production routes are possible, with varying TRL, energy demand and associated CO_2 emissions. The approach used in this work is described next.

2.1. Technology readiness level

The TRL of the different technological routes was evaluated according to Table 1, which follows the definition of TRL given in the Horizon 2020 Work Programme [8]. Next, the TRL for producing each one of the 16 CO_2-based products is determined, considering the most mature CCU technological route. The routes are organised by TRL, with aggregates, carbonated concrete, methanol, dimethyl ether, methane, propylene carbonate, polyols and food-grade and "greenhouse-grade" CO_2 all at TRL9; ground and precipitated calcium carbonate at TRL7; ethanol and biodiesel at TRL5; and cyclic carbonates and isopropanol at TRL3, as summarised in Table 2.

2.1.1. Technological routes for CO_2-based products at TRL 9

Routes for aggregates and carbonated concrete production via CO_2 mineralization are at TRL 9. Carbon-negative construction aggregates are manufactured using a technology by Carbon8 since 2012 [9], whereas Solidia Cement™ and CarbonCure™ uses CO_2 curing technology to produce carbonated cement.

The company Carbon Recycling International® (CRI) is a well-known commercial example of a CCU company. CRI produces hydrogen through geothermal-powered alkaline water electrolysis, which is then reacted to produce methanol (available on the EU market under the tradename Vulcanol®). The first plant at the CRI site came online in 2011, named the George Olah Renewable Methanol Plant, producing 4000 t MeOH per year. The company recently brought a 50 kt/y facility online.

Dimethyl ether (DME) is commercially produced via methanol dehydration. Therefore, the indirect production of DME via CO_2-based methanol is also at TRL 9. Routes for direct DME production are at lower TRL.

The technology of storing electricity by producing hydrogen via water electrolysis and subsequently performing CO_2 hydrogenation to

Table 1
Technology readiness level definitions.

TRL	Definition
TRL 1	basic principles observed
TRL 2	technology concept formulated
TRL 3	experimental proof of concept
TRL 4	technology validated in lab
TRL 5	technology validated in relevant environment
TRL 6	technology demonstrated in relevant environment
TRL 7	system prototype demonstration in operational environment
TRL 8	system complete and qualified
TRL 9	actual system proven in operational environment

Source: [8].

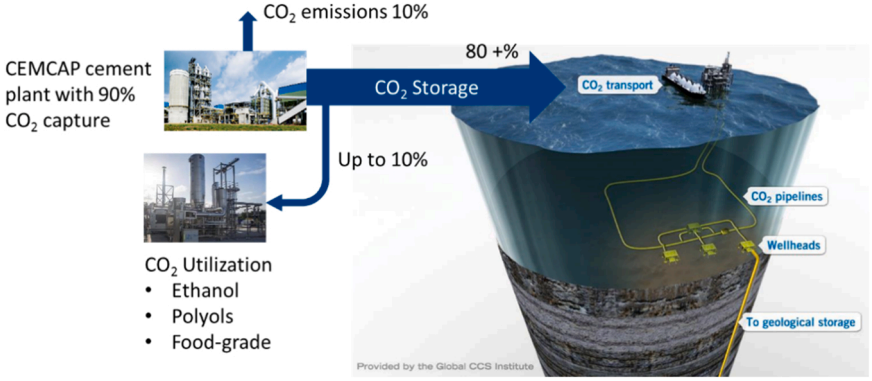

Fig. 1. Representation of CCUS chains considered in this work.

Table 2
Criteria for selection of CO_2 utilization products.

	Market size (Mt/y)	CUP (Mt/y)	TRL	C average oxidation state
$CaCO_3$ (GCC)	75	33	7	4
$CaCO_3$ (PCC)	14	6,16	7	4
Aggregates	53,200	3600	9	4
Carbonated concrete	16,500	1650	9	4
Methanol	80	110	9	-2
DME	8	7,65	9	-2
Methane	1100	3025	9	-4
Ethanol	86,8	166	5	-2
Isopropanol	2	4,33	3	-2,33
Biodiesel	20	30	5	-3,29
PPC	6	3	9	3
Polyols	10	2	9	3
Cyclic carbonates	0,1	0,04	3–4	4
Formic acid	0,7	0,67	5	2
Food-grade CO_2	17	17	9	4
CO_2 for greenhouses	5	5	9	4

methane is known as Power-to-Gas (or PtG). PtG development has progressed significantly in Europe. The Audi e-gas plant in Werlte (Germany), is the largest industrial PtG plant in the world (6 MWe). It is based in the catalytic methanation of pure hydrogen and carbon dioxide in a single isothermal fixed-bed reactor. The electrolysers are powered by an offshore wind park in the North Sea, with 4 turbines of 3,6 MW each. The high purity CO_2 is captured from a nearby biogas plant by amine scrubbing. [10].

The synthesis of some polymeric materials using CO_2 is also at TRL 9. Novomer has commercialized the production of polypropylene carbonate polyols (from propylene oxide and CO_2) for use in polyurethane hot melt adhesive applications in 2014, under the name of Converge® [11]. Also, Covestro has developed a CO_2-based process that leads to 20 wt% CO_2 content in a polyol intermediate. The polyol, cardyon™, has been used in the fabrication of flexible polyurethane foam for mattresses [12]. Belgian manufacturer Recticel launched the first products on the market end of 2016.

Food-grade CO_2 is commonly produced from captured CO_2 using available purification and liquefaction technology. In the Netherlands, captured CO_2 is also used in greenhouses, being provided via the OCAP pipeline, or liquified.

2.1.2. Technological routes for CO_2-based products at TRL 7

The Calera process, which produces calcium carbonate, is available at TRL 7 and will soon reach TRL 9. The Calera Corporation has designed, constructed and operated two pilot plants to utilize the flue gas from coal-fired power plants to produce calcium carbonate. The flue gas is contacted in a scrubber with an aqueous alkaline solution that effectively removes the CO_2 (capture step) and a calcium source that results in the formation of the calcium carbonate product. The intellectual property of Calera was acquired by Fortera, which brands their product Reactive Calcium Carbonate or RCC, and has recently teamed up with the cement company Lehigh Hanson to construct a commercial scale unit in California, USA [13].

2.1.3. Technological routes for CO_2-based products at TRL 5

Ethanol synthesis from CO_2 can be achieved based on reverse water-gas shift followed by CO hydrogenation. Despite extensive research on catalyst development [14], there are no commercially viable catalysts for the production of higher alcohols (C2-C4) from CO hydrogenation. Additionally, the presence of CO_2 can greatly change the product yield [15]. A pilot plant in Güssing, Austria, produces a mixture of methanol (45%), ethanol (14%), propanol (14%) and water (27%) from biosyngas (synthesis gas produced by steam gasification of biomass) containing 27% CO_2, 53% H_2, 14% CO and 6% CH_4 on a dry basis [16]. While CO_2 hydrogenation leads to methanol, CO hydrogenation leads to hydrocarbons and higher alcohols. [17].

The thermocatalytic conversion of hydrogen and carbon dioxide has been shown to produce 85 wt% formic acid dissolved in methanol (solvent). CO_2 and hydrogen react at around 90ºC and 100 bar in the presence of two catalysts (ruthenium- and phosphino-based catalysts), using aqueous MeOH as solvent [18].

Microalgae have a high oil content (up to 80 wt% depending on the species). Lipids, in the form of triacylglycerides (TAGs), typically provide a storage function in the cell that enables algae to endure adverse environmental conditions. About 80% of the microalgae oil content can currently be converted to biodiesel. The biodiesel production from extracted microalgae oil follows the same technology path as the commercial state-of-the-art process, but using oil produced by microalgae as a resource [19]. As compared to energy crops, microalgae are easier to cultivate and have much higher growth rates and productivity. Different microalgae species can be adapted to live in a variety of environmental conditions, thus it is theoretically possible match the local environmental conditions to the optimal microalgae growth conditions. Chlorella sp., Chlorococcum sp. and Neochlorosis oleabundans are found to be potential biodiesel feedstocks [20].

2.1.4. Technological routes for CO_2-based products at TRL 3

The research on the production of cyclic carbonates from CO_2 focuses on fundamental reaction aspects, such as development of catalysts and optimization of solvent matrix. As an example, the CyclicCO_2R project (2012–2016) focused on the development of a continuous process to produce cyclic carbonates from CO_2 and renewable feedstocks via two routes. In the direct route glycerol, and related diols, were reacted with CO_2 using an innovative catalyst to glycerol carbonate. In the indirect route an allyl alcohol is formed first, then epoxidized to glycidol, and then a step of cycloaddition of CO_2 leads to glycerol carbonate. It was shown that other cyclic carbonates could also be produced by the developed catalyst and process [21].

Isopropanol can be biologically produced by genetically engineered *Escherichia coli*. Laboratory tests shows it is possible to produce 81.6 mM isopropanol with a yield of 43.5% (mol/mol) in the production phase [22]. Isopropanol was also produced by *Cyanobacteria Synechococcus* in the concentration of 26.5 mg/L [19].

2.2. CO_2 uptake potential

For each product, the CO_2 uptake potential (CUP) is determined. The CUP of a certain technological route is defined by stoichiometry and the market size, according to Eq. 1.

$$CUP\left[\frac{Mt}{y}\right] = \frac{CO_2 \quad incorporated \quad [kg]}{product \quad [kg]} Market \quad size \quad \left[\frac{Mt}{y}\right] \tag{1}$$

As per Eq. (1), if 1 kg of product contains 0,4 kg of CO_2, and the product market is 2 Mt/year, then the CO_2 uptake potential is 0,8 Mt/year. It should be highlighted that the CUP metric does not consider the CO_2 emissions associated with the manufacturing processes (scopes 1 or 2). Hence, producing methanol by CO_2 hydrogenation has a fixed CUP, regardless whether renewable or fossil-derived hydrogen is used.

It should be emphasized that the CUP metric is not directly related to the potential of a certain CCU route to lower the net emissions of CO_2, which is very dependent on the technological route itself, and must be assessed by means of a life cycle analysis (LCA). Also, while CO_2-based fuels may displace conventional fossil fuel use, thus leading to CO_2 abatement, it should be assessed whether it is likely that CO_2-based fuels compete against other, more effective climate mitigation technologies and processes. This makes the cases for use of CO_2-based fuels in aviation and in road transport very distinctive. Therefore, treating all forms of CO_2 uptake technologies as de facto CO_2 abatement could have detrimental impacts on efforts to reduce emissions on the long-term

[23]. These in-depth analysis are out of the scope of the current work, but should be part of the CCUS chain definition strategy for specific (cement) plants.

Table 2 summarises the CUP for all products, based on the market assessment performed during the CEMCAP project [24]. The market size used in the assessment was based on the *current market* for the selected products. However, some of these products have a potential for a substantial market increase in the future. In particular, methanol, formic acid and DME could be largely employed as fuels, displacing fossil-based fuels by CO_2-based fuels.

2.3. Carbon atom oxidation state

The energy demand to make a product out of CO_2 is dictated by the technological route chosen. While factors such as the reaction conditions (pressure, temperature), the conversion per pass and selectivity influence the energy demand, the governing factor is thermodynamics. As an indication for the energy demand, we have chosen to look at the oxidation state of the carbon atom(s) in the CO_2-based product. The carbon atom in CO_2 is in a high oxidation state (+4), which indicates it is a highly stable molecule. Reducing the oxidation state of the carbon atom requires energy and therefore, the processes of CO_2 utilization in which such a reduction is required are typically energy intensive.

Table 2 summarises the average C oxidation state in all products. This makes evident that producing fuels out of CO_2 is energy-intensive. Most of this energy is provided by the hydrogen molecule.

2.4. Selected CO_2 utilisation products

As a result, and in consultation with the cement companies representatives participating in CEMCAP, three CO_2-based products were chosen to be further evaluated in the CCUS chains: a fuel (ethanol), a polymer feedstock (polyol), and food-grade CO_2. The choice for a fuel is guided by the need for products that have substantial markets, and the drawback of this route is the energy requirement. Polymers represent products with a small market, but relatively high prices and low energy demand, therefore increasing the probability of a positive business case. Finally, the direct use of CO_2 is a low hanging fruit, as it does not require investments in conversion.

Aggregates and carbonated concrete have a sizable carbon sequestration capacity, the production technology is at TRL9 and the energy requirement is low (therefore leading to potentially low cost). Nevertheless, these technological routes were excluded from the present analysis on the basis of the possible challenges faced by the two technologies.

Producing aggregates via CO_2 mineralization is an accelerated version of a naturally occurring process (taking minutes instead of decades). This means that the CO_2 uptake by aggregates production doesn't necessarily lead to CO_2 abatement over the product life cycle. On the other hand, avoiding mining for primary aggregates and recycling waste instead has been shown to lead to lower CO_2 emissions [25]. As for all CCU-based products, LCA is needed in order to evaluate the net effect.

Alternative cements using CO_2 to cure face different types of challenges. One of the biggest challenge is acceptance of the alternative cements and introduction to the current norms which will allow their use in construction products. Due to durability issues that are related to the lower than normal concrete pH it is expected that the acceptance will be limited to unreinforced dry cast precast elements. This can hinder the forecasted market growth. Use of the material in reinforced concrete will need further optimization of the material to resolve the lower pH issue. Other challenges include production limitations as a curing chamber is needed for the curing of the alternative concrete and changes in the quality control processes.

3. Description of CCUS chains

3.1. Reference cement plant and MEA-based capture

The reference cement plant with MEA-based capture is extracted from Roussanaly et al. [26], [27]. The reference cement plant considered in this study is a Best Available Technique (BAT) plant defined by the European Cement Research Academy (ECRA). It is based on a dry kiln process, and consists of a five-stage cyclone preheater, calciner with tertiary duct, rotary kiln and grate cooler. The plant has a representative size for a European cement plant with a capacity of 2896 tonne clinker per day. This corresponds to a capacity of ca. 1 Mt clinker per year, or 1, 36 Mt cement per year, with a run time of $>$ 330 days per year. The specific CO_2 emissions of the plant amounts to 850 kg_{CO2}/t_{clk}.

The cement plant result in a flue gas with an average yearly CO_2 content in the flue gas of 20%mol. The MEA-based solvent CO_2 capture results in work and heat consumptions of respectively 0,45 and 3,81 MJ/kg_{CO2} to achieve CO_2 capture ratio of 90%.

3.2. CCUS chain alternatives

3.2.1. The CCS reference chain

This chain evaluates CO_2 capture from the reference CEMCAP cement plant [26], [28] assumed to be located in Belgium, while an offshore site on the Dutch continental shelf is considered for the storage. After capture, the CO_2 is conditioned and transported by a stand-alone pipeline to a hub in the Rotterdam area. From this hub, the CO_2 is assumed to be transported in a shared offshore pipeline to a saline aquifer. A shared transport and storage infrastructure with an annual flow of 13,1 Mt_{CO2}/y as in the EU project COCATE [29] is considered. This chain is meant to be representative of both CCS from an inland cement plant and implementation of CCS once a strategy for joint CCS

Table 3
Description of the CCS reference case.

Section	Parameter	Value
Cement plant	Approximate location	Inland Belgium
	Capacity [Mt_{cement}/y]	1,36
	CO_2 emissions without CO_2 capture [Mt_{CO2}/y]	0771
	Exhaust flue gas average flow [t/h]	353,1
	Exhaust flue gas average CO_2 content [mol%]	19,8
CO_2 capture and conditioning	Type of capture technology	MEA-based
	CO_2 capture ratio [%]	90
	CO_2 captured [Mt_{CO2}/y]	0694
	Conditioning specification after capture	Pipeline
	Pressure after conditioning [bar]	150
	Temperature after conditioning [°C]	40
First transportation step	Transport scenario	Stand-alone onshore pipeline to a Dutch hub
	CO_2 transported [Mt_{CO2}/y]	0694
	Transport distance [km]	120
	Required pressure after reconditioning [bar]	200
Second transportation step	Transport scenario	Shared offshore pipeline to storage
	CO_2 transported [Mt_{CO2}/y]	13,1
	Transport distance [km]	150
	Minimum delivery pressure at storage [bar]	60
Storage	Storage type	Saline aquifer
	CO_2 stored [Mt_{CO2}/y]	13,1
	Well injectivity [Mt_{CO2}/y/well]	0,8
	Storage location	Dutch continental shelf

transport and storage infrastructure has been established.

The CCS reference case is described in detail in Table 3. The transportation distances are shown in.

Fig. 2, using Maasvlakte (nearby Rotterdam) as reference central location. This figure shows that an onshore pipeline of 120 km could connect a cement plant at the north of Belgium to a transportation hub nearby Rotterdam, whereas a shared offshore pipeline of 150 km would be able to connect the CO_2 to a saline formation, with estimated capacity between 110 and 225 Mt CO_2[30].

3.2.2. CCUS: integrated blue ethanol production

Ethanol obtained from CO_2 captured from industrial sources is referred to as *blue ethanol*, to differentiate it from *green ethanol* obtained from biomass fermentation. It should be highlighted that the differentiation is made regarding the production route, and not the quality of the final product.

The blue ethanol production route considered in the present work is based on the model presented by Atsonios et al. [31]. In a first step, CO_2 is hydrogenated to methanol, which is then dehydrated to DME. This is followed by DME carbonylation to methyl acetate, which is finally hydrogenated to ethanol. It should be highlighted that this technological route is currently at TRL 3 (i.e., the proof of principle is shown in laboratory environment), and therefore there is a high uncertainty connected to the route economic results.

The route requires renewable hydrogen to be available. For this, it is assumed that 50 MW of excess renewable electricity are available. This value represents 0,07% of the predicted 70 GW of installed wind generation capacity in the North Sea by 2030 [32]. The 50 MW can be used to produce 3,2 kt_{H2}/y (considering an efficiency of 61,6% and an availability of 40,5%), at a cost of 1,04 €/kg. This cost is obtained considering that the excess electricity is available for free, in a highly favourable scenario.

The availability of renewable hydrogen greatly limits the CO_2 utilization capacity, and consequently the ethanol throughput. Via the conversion of 23,8 kt_{CO2}/y, or 3,1% of the emissions of the CEMCAP plant, 12,5 kt/y of ethanol are produced. As 90% of the CO_2 emissions are captured, the non-utilized fraction (86,9%) must be directed to the storage site. Therefore, the proposed CCU route is not a stand-alone solution, but works as an integrated link in the CCUS chain.

3.2.3. CCUS: integrated polyol production

Propylene oxide (PO) is the main feedstock in industrial polyol manufacturing routes. A novel route, in which PO is partially replaced by CO_2 so that 20 wt% CO_2 content in the polyol product is achieved has been proposed [33]. This novel route is evaluated in the current work.

The typical size of polyols plants is around 100 kt/y, whereas the polyols market is around 10 Mt/y. Based on these market numbers, the polyol throughput is set at 288 kt/y, as proposed by Fernández-Dacosta et al. [33]. The CO_2 utilization capacity is therefore limited by the throughput of the polyol plant, which in its turn is limited by the market. The simulated polyol plant consumes 57,5 kt_{CO2}/y, which is equivalent to 7,5% of the emissions of the CEMCAP reference plant. Therefore, 82, 5% of the CO_2 emitted needs to be stored. Again, given the mismatch of scales between the cement emissions and the CO_2 utilization plant, utilization cannot be applied as a stand-alone solution, but only as an integrated link in the CCUS chain.

3.2.4. CCUS: integrated food-grade CO_2 production

Food-grade CO_2 can be used inside greenhouses to raise the atmospheric CO_2 levels to 600–1000 ppmw, in order to accelerate the plants growth. In the Netherlands, during the summer, natural gas is combusted on a large scale to provide CO_2 to greenhouses, leading to net emissions of about 7 Mt_{CO2}/y. An annual growth of 100 kt_{CO2}/y in the Dutch CO_2 market is expected up to 2020. Additionally, the food and beverage industries consume about 17 Mt_{CO2}/y worldwide [34].

The conceptual design of a plant for purifying CO_2 to food-grade quality and liquefying it are developed. The plant capacity is set as 50 kt_{CO2}/y or about 6,5% of the emitted CO_2. It is considered that the plant will serve end-users which are currently producing their own CO_2 locally. Burning natural gas to generate CO_2 is still a common practice in the horticulture industry in The Netherlands. Therefore, blue CO_2 directly replaces fossil-derived CO_2.

3.3. Parameters for techno-economic evaluation

For the CCS chain, the techno-economic performances of the CO_2 capture and conditioning units is extracted from Roussanaly et al. [26,27], while the iCCS CO_2 value chain tool developed by SINTEF Energy Research [35,36] is used to assess the costs of the CO_2 transport and storage parts of the CCS chain. While more detailed description on the modelling of the CO_2 conditioning, transport and storage in the iCCS A more detailed description of the iCCS CO_2 value chain tool and underlying assumption can be found in previously published studies [36–39].

For the CO_2 utilization plants, the investment is determined based on the available literature information. Atsonios et al. [31] determined the investment for the ethanol production plant at 170 €/ton of ethanol, and this factor is maintained in the current paper. This leads to an investment of 22,6 M€ for the utilization part of this CCUS chain. For polyol production, Fernández-Dacosta et al. [33] determined an investment of 21 M€, for a plant of the same scale as simulated in the current work. Finally, for CO_2 purification to food-grade and liquefaction, a commercial quotation was used as basis, leading to an investment cost of 16 M€. These three approaches are different, and the assumption made by Atsonios et al. [31] differ from those made by Fernández-Dacosta et al. [33]. However, given the low TRL of blue ethanol and polyol technologies, the expected accuracy in investment estimation is ± 40%. We assume that this uncertainty is larger than the differences in the baseline assumptions. For the food-grade CO_2 plant, since the investment is based on a quotation, the expected accuracy is estimated as ± 20%, which also reflects the high TRL level of this technology.

The same sources ([31,33] and commercial quotation) were used to estimate the electricity, steam, cooling water and raw materials consumption for each of the routes. The operational costs, revenues and CO_2 emissions were harmonized considering the factors given in Table 4.

The CO_2 avoidance cost of each CCUS chain is calculated based on Eq. (2). The CO_2 avoidance cost is calculated on the assumption of a real discount rate of 8%[1] and an economic lifetime of 25 years [40]. In addition, investment costs assume that construction costs are shared over a three-year construction period following a 40/30/30 allocation.

$$\text{CO}_2\text{ avoidance cost} = \frac{\text{Annualised investment due to CCS implementation} + \text{Annual operating cost due to CCS implementation}}{\text{Annual amount of CO}_2\text{ emissions avoided}} \quad (2)$$

[1] This real discount rate of 8% corresponds to a nominal discount rate around 10% if an inflation rate of 2% is assumed.

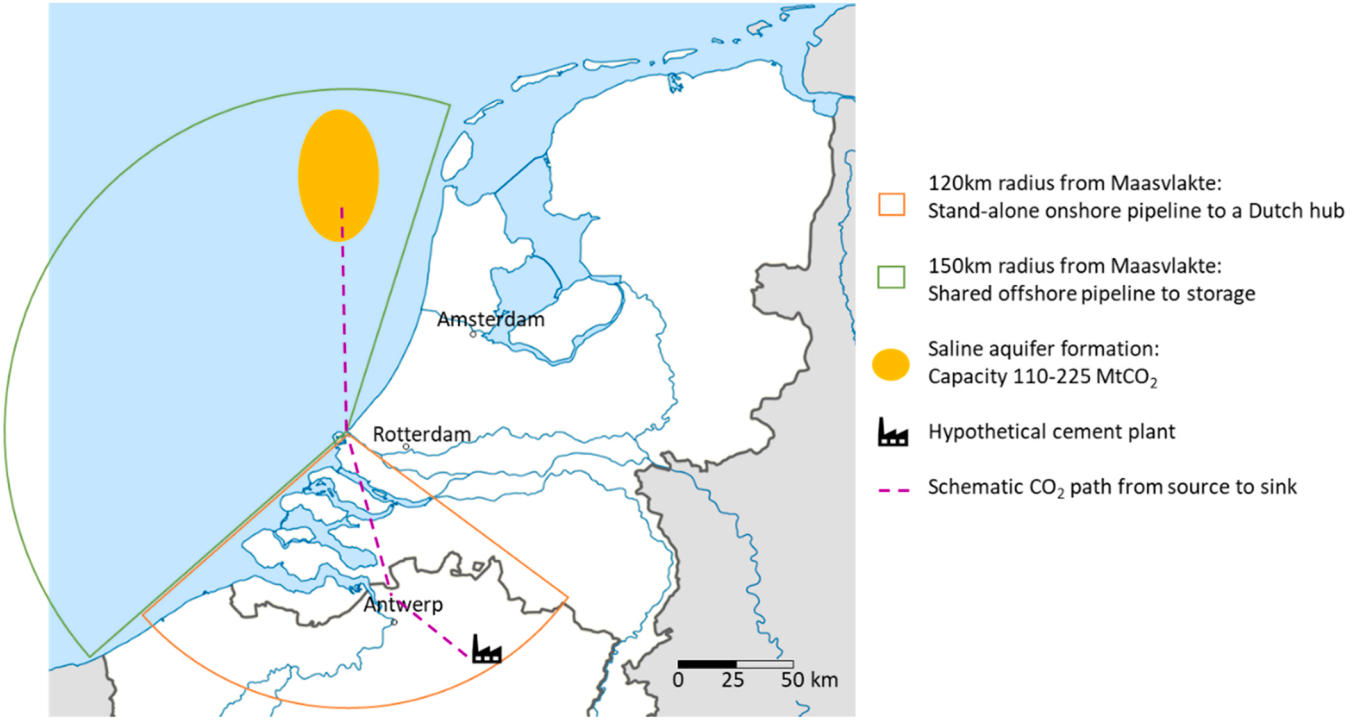

Fig. 2. Representation of the CCS reference case.

Table 4
Factors considered for techno-economic evaluation of the CO_2 utilization plants.

Utility	Price	Source
Electricity	30 €/MWh or 0 €/MWh if excess is available[a]	[36], This work
Steam (from natural gas)	9,1 €/GJ	[36]
Cooling water	0035 €/m^3	[36]
Chemical	Price	
Ethanol	633 €/t	[31]
Ethanol premium	0 €/t	This work
Polyol	1400 €/t	This work
Food-grade CO_2	80 €/t	This work
Propylene oxide	1400 €/t	[33]
Glycerol	730 €/t	[33]
Monopropylene glycol	1550 €/t	[33]
Utility/raw material	Emission factor	
Electricity	170 $kg_{CO2\text{-}eq}$/MWh	[36]
Steam (from natural gas)	56,1 $kg_{CO2\text{-}eq}$/GJ	[36]
Polypropylene oxide	4,5 $kg_{CO2\text{-}eq}$/kg	This work

[a] In the utilization cases where indicated, electricity is considered to be available at zero cost, in a simplistic, best-case scenario representation of the situation in which the amount of electricity produced surpasses the demand, leading to a temporary excess.

The total CO_2 avoided in each chain is calculated adding the mass of CO_2 geologically stored to the mass of CO_2 displacement. For instance, in case of polyols production, CO_2 partially displaces PO, which leads to actual CO_2 avoidance. The CO_2 uptake in the products is not computed as CO_2 avoidance, as this can be seen as questionable, given the relatively short lifetime of CO_2-based products, in particular fuels. To improve the estimation of the amount of CO_2 avoided, for real-life cases and particularly when considering the Emission Trading System, comprehensive Life Cycle Analyses should be performed to evaluate each CCUS chain proposal.

4. Results

4.1. CCS results

As shown in Table 3, the CO_2 capture rate is set as 90%, or 0,694 Mt_{CO2}/y. However, due to emissions related to energy usage in the capture, conditioning and transport processes, the amount of CO_2 avoided is lower than that. The quantity of CO_2 avoided is determined by the difference between the quantity of CO_2 captured and the emissions associated to each one of the processes of the chain, as summarized in Fig. 3.

In the CCS reference chain, 0,504 Mt_{CO2}/y are avoided, or 65% of the CO_2 emitted by the cement plant. The total cost of CO_2 avoided is 114 €/ton, which is mostly attributed to capture (69 €/t) and conditioning (16 €/t).

4.2. CCUS results: ethanol production

The cost of producing ethanol via DME is estimated as 656 €/ton of ethanol. This cost is only slightly above the market value of 633 €/ton. Hydrogen, even at an extremely low cost achieved by using free excess electricity, represents 41% of this total.

The calorific value of ethanol is 29,7 GJ/t. In terms of energy, the ethanol production cost is 22 €/GJ. In the cement plant, coal is used as fuel, and has the price of 3 €/GJ. Therefore, substituting coal by ethanol would lead to a weaker business case. From that perspective, the produced ethanol should be sold on the market where it could substitute fuels with higher quality than coal – for instance, green ethanol.

Currently, the most cost- and CO_2-effective process for the production of green ethanol is the fermentation of sugarcane. While sugarcane growth fixates CO_2 from the atmosphere, the various steps in the production of green ethanol emit CO_2, and the net result is the emission of 3,3 t_{CO2}/t green ethanol. In case of ethanol production from wheat, the efficiency is lower, and the emissions are 3 times higher. In the current case, 12,5 kt/y of blue ethanol are produced, thus replacing the same flow of green ethanol. This replacement leads to the avoidance of 41 and 123 kt of CO_2 per year, using sugarcane and wheat as baselines,

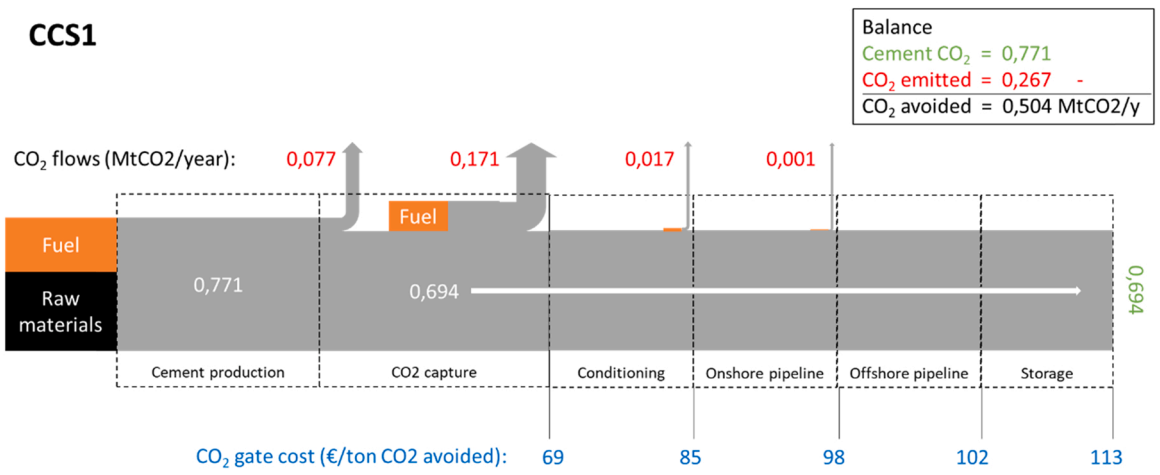

Fig. 3. CCS chain results.

respectively.

While the process of producing blue ethanol is not profitable, it contributes to increasing the total CO_2 avoidance of the CCUS chain to 0,518 Mt_{CO2}/y in the sugarcane case and 0,6 Mt_{CO2}/y in the wheat case, as compared to 0,504 Mt_{CO2}/y of the reference CCS case. In this way, the cost per tonne of CO_2 avoided drops from 114€ (CCS) to 111€ (sugarcane) or 96€ (wheat). The cost difference for sugarcane is only marginal, but in the case of wheat, it appears more relevant. It should be noted, however, that these cost differences are within the expected uncertainty level for the estimate procedure (at best, ± 40%).

This CCUS chain demonstrates the complexity involved in the CO_2 avoidance cost analysis: it must take into consideration not only the product that is formed, but also the market in which it is placed. The feasibility of integrating ethanol production to a CCS chain is therefore case dependent, and the economic evaluation must be supported by a life cycle assessment (LCA).

The results of the CCUS chain are summarized in Fig. 4 for the sugarcane case.

4.3. CCUS results: polyols production

The polyol plant CAPEX is estimated to be 21 M€, taking the work of Fernández-Daosta as basis [33]. Regarding the price of chemicals, a conservative approach is used, as both polyol and PO prices are set as 1400 €/t (zero spread). The business case of blue polyol production lies partially on the fact that the CO_2 content in the material is replacing PO. The gate cost of CO_2 after capture is 69 €/t, much lower than that of PO. Therefore, the production costs are greatly reduced.

The production of PO is carbon-intensive: 4,5 $t_{CO2\text{-}eq}$ are emitted per tonne of PO produced. Therefore, even with the partial substitution of PO by CO_2, the polyol production process is still a net CO_2 emitter if the CO_2 content in polyol is limited to 20 wt%. The route becomes a net CO_2 consumer when at least 50% of the PO is substituted by CO_2 – which is unfortunately not yet technically feasible. Yet, the production of blue polyol avoids the emission of 0,91 $t_{CO2\text{-}eq}$ per ton of PO as compared to the conventional route.

Fig. 5 shows the results of the entire CCUS chain: it avoids 0,708 Mt_{CO2}/y, and produces 288 kt/y of polyols. Due to the high value of polyols, the full chain is profitable. Even when setting the spread between the polyol and the PO prices to zero, the profit is of 43 €/ton of polyol produced, or 18 €/ton CO_2 avoided.

4.4. CCUS results: food-grade CO_2 production

Because the direct avoidance of fossil CO2 cancels out the emissions of food-grade CO2, the total CO2 avoidance of this CCUS chain is the same as that of the reference CCS: 0,504 MtCO2/year or 65% of the cement plant emissions. These results are given in Fig. 6.

The price of food-grade CO_2 is highly dependent on the location, but for Europe it can be around 80–150 €/t. In the Netherlands, CO2 delivered via a distribution pipeline to vegetable growers has a market cost of between €50–80 per tonne of CO_2, depending on transportation

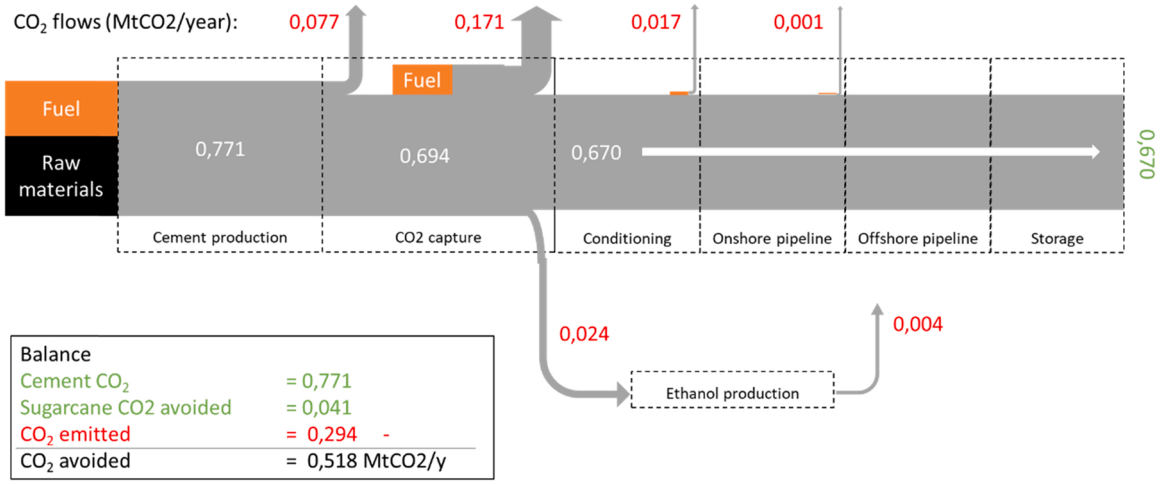

Fig. 4. CCUS results: blue ethanol production and substitution of green sugarcane-based ethanol.

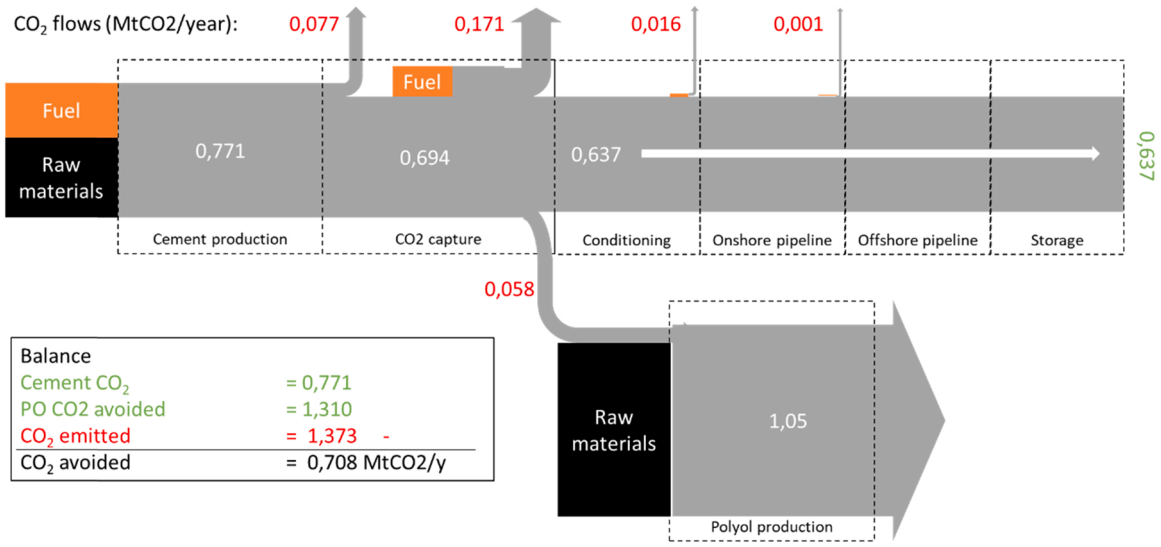

Fig. 5. CCUS results: polyol production by partial substitution of PO.

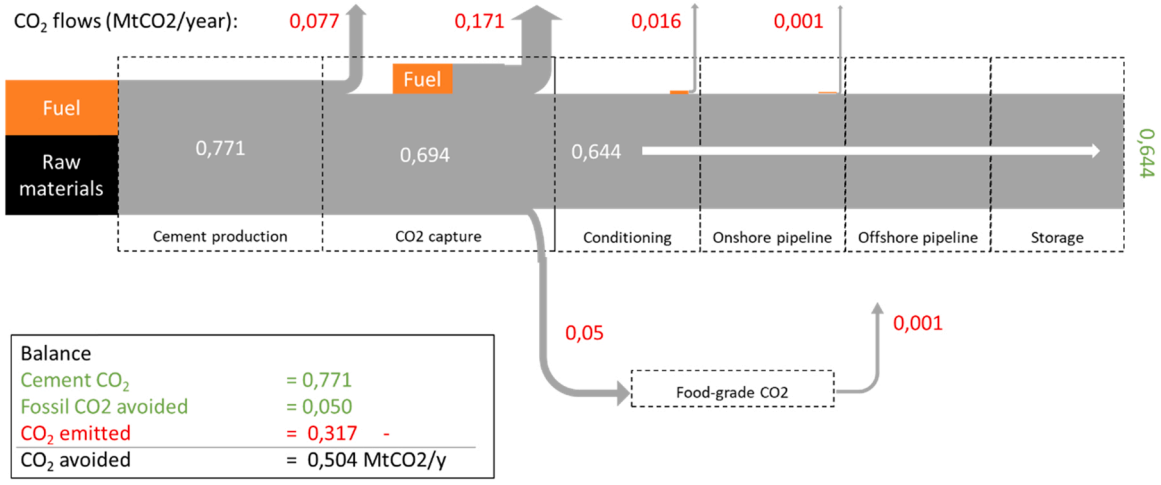

Fig. 6. CCUS results: food-grade CO_2 production with substitution of fossil-derived food-grade CO_2.

distance and greenhouse capacity [34].

Setting the price of food-grade CO_2 at 80 €/t, the total CCUS avoidance cost drops 5%, from 114 to 108 €/ton. The break-even CO_2 price – that leads to the same avoidance cost for CCUS and CCS – is 25€/t, thus below the current European price range. From this perspective, producing as much food-grade CO_2 as can be placed in the market is a viable option for lowering the integrated CCUS costs.

However, if green CO_2 is available (e.g. from fermentation), the CCUS option actually leads to a higher cost than CCS: 120 €/t CO_2 avoided, for a CO_2 market price of 80 €/t. In this case, the substitution of green CO_2 by blue CO_2 leads to lower CO_2 avoidance by the full chain – or lower sequestration efficiency – which has a detrimental effect on the avoidance cost.

5. Sensitivity analysis

The costs considered in the present analysis (see Table 4) may influence the conclusions of the present work. Therefore, we have conducted a sensitivity analysis on some of the cost factors, to understand their influence on the CCUS avoidance costs.

In a first exercise, the products' (ethanol, polyol and food-grade CO_2) prices were varied by − 50% and + 50%. The results of the ethanol and food grade cases are relatively insensitive towards product price variations, whereas for polyols, a strong sensitivity is seen in Fig. 7. However, varying the polyol price independently of the propylene oxide price is not a realistic scenario. Therefore, in a next exercise seen in Fig. 8, we have varied the spread between these two prices. There is a linear

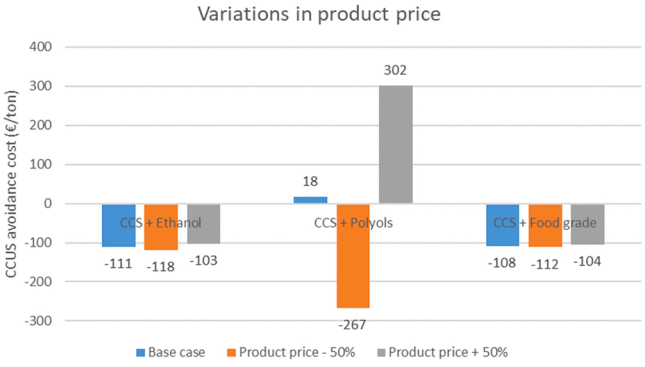

Fig. 7. Sensitivity analysis results: variations in product price. Positive values indicate profit.

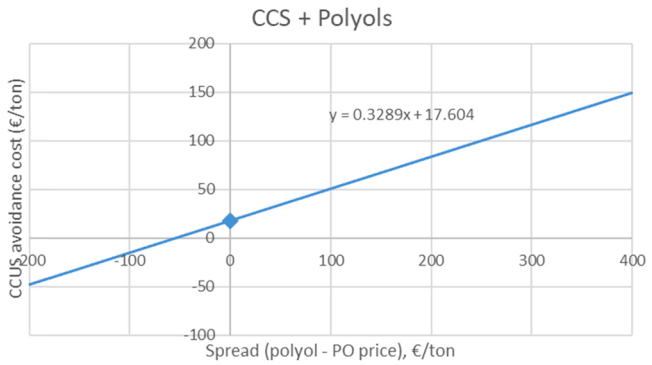

Fig. 8. Sensitivity analysis results: varying the spread between polyol and PO prices The base case is indicated with a diamond. Positive values indicate profit.

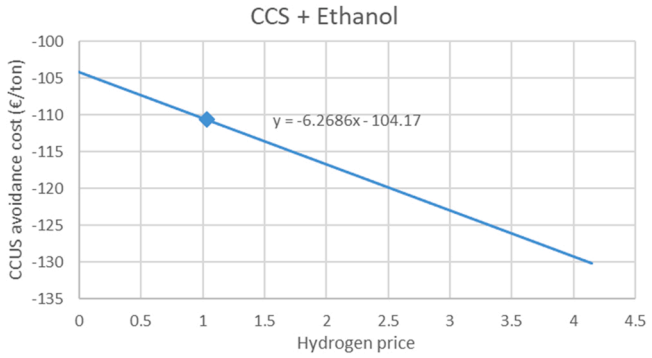

Fig. 9. Sensitivity analysis results: varying the hydrogen price. The base case is indicated with a diamond.

relationship between the CCUS avoidance cost and the spread, with the break-even point of this scenario appearing at a negative spread of -53 €/ton. This means that this scenario would lead to a profit so long as this price difference is above this limit.

For the ethanol case, the most relevant cost factor is the green hydrogen price. The base case, as discussed, assumes free excess electricity, leading to a very low hydrogen price of 1.04 €/kg. There is a linear relationship as the hydrogen cost and the total avoidance cost of the CCUS route, as can be seen in Fig. 9. For the CCUS route to have a cost benefit as compared to the CCS-only route, the hydrogen price needs to be below 1.5 €/ton.

While the products price and raw materials costs affect the utilization step of the CCUS chain, other important factors that may affect the results are related to the cost assumptions for CO_2 capture and conditioning, transport and storage. These may be related to the technology choice for capture and conditioning, the transportation distance and pressure, the geology of the storage site, etc. To represent all these uncertainties, the total cots of each chain element (capture, conditioning, transport and storage) were varied by + /− 50%. The results can be seen in Fig. 10. While all elements play a role in the total result, the costs of capture are the most relevant to the end result of the CCUS chains. For instance, when the capture costs are increased by 50%, the CCS+polyols case no longer leads to a profit, but to a cost of ca. 8 €/ton CO_2 avoided. In any case, this is still a much better result than the CCS scenario, with an overall cost of 150 €/ton CO_2 avoided in this high CO_2 capture cost scenario.

In general, while the sensitivity analysis helps putting some of the conclusions in perspective, it does not change the main outcome of the economic assessments performed in this work: the cost benefit of incorporating utilization in CCS chains, thus leading to CCUS chains, is dependent on the product in question. Cost competitive solutions may arise when CO_2 displaces other more expensive and CO_2-intensive

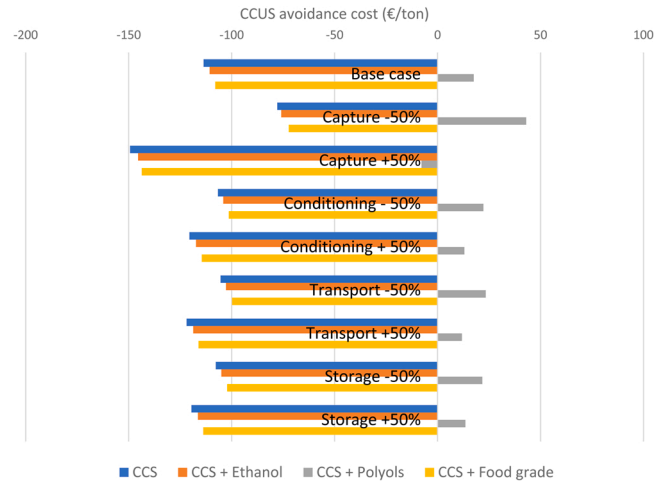

Fig. 10. Sensitivity analysis: elements of the CCS chain. Positive values indicate profit.

molecules, as demonstrated by the polyols case. However, for ethanol and food grade CO_2, the economic benefit is not necessarily a given, and will be rather limited. Thus, the rationale for most CO_2 utilization cases probably will not come from an economy perspective, but from the need to recarbonize the (petro)chemical industry, by substituting fossil-based carbon by "circular economy carbon". Therefore, captured CO_2 from cement plants (or other fossil-based sources) is unlikely to become a raw material in used abundantly in the future.

6. Conclusions

Already in the title of this paper we have posed a question regarding the feasibility of integrating CO_2 utilization to CCS chains in the cement industry. By evaluating the CCUS chains proposed, the answer to that is: "it depends". We show, for the fuel case, that the characteristics of the product that is displaced has a great influence on the avoidance cost: while producing blue ethanol to displace sugarcane ethanol seems unfeasible, the displacement of wheat-based ethanol leads to an improved business case as compared to the reference CCS case. It should be highlighted that these results consider the use of free excess electricity, which is an optimistic scenario.

In the second CCUS chain, we have evaluated the integrated production of polyols. This case leads to a profitable operation, because CO_2 replaces an expensive chemical as a raw material, and lowers the CO_2 emissions of the chain while doing so. While the polyols market is limited as compared to the total amount of CO_2 to be avoided by the cement industry as a whole, this CCUS case could be feasible for some cement plants. Moreover, the polyols case may be representative of other high added value products, such as other polymer precursors or cyclic carbonates.

In the third CCUS chain, we show that the production of food-grade CO_2 is feasible as long as it is used to replace fossil-derived CO_2 produced especially to be used in the food and beverage industries or in greenhouses. If CO_2 from other sources is available – such as green CO_2 from fermentation, then this CCUS chain leads to higher CO_2 avoidance costs as compared to the reference CCS chain.

A general conclusion from this work is that the average cement plant emits much more CO_2 than can be utilized in a single CO_2 utilization plant. That may be due to market constrains, as in the cases of polyols and food-grade CO_2, or low availability of raw materials, as in the case of ethanol and fuels in general (which require renewable hydrogen). For the routes evaluated in this work, the fraction of the emitted CO_2 directed to the utilization plant was always below 10%. Therefore, when connected to the cement industry, utilization cannot be applied as a stand-alone solution, but as an integrated link in CCUS chains.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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