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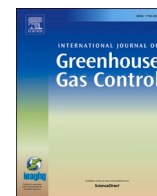
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Cross-sectoral assessment of CO₂ capture from U.S. industrial flue gases for fuels and chemicals manufacture

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ABSTRACT

Although CO₂ impacts the environment negatively, it can be a valuable resource due to its carbon content. The U.S. industry emits over 825 Mt of CO₂ annually, with an expected increase in the future. This article analyzes 27 different technology combinations for capturing and using CO₂ for industrial feedstocks, including the production of synthetic methane, methanol, and Fischer-Tropsch fuels. The study also estimates and compares the energy requirements for capturing and converting CO₂ from 16 different industrial sources, as well as the energy requirements for hydrogen production through state-of-the-art and emerging electrolyzer technologies. Additionally, the study develops a combined scenario that outlines a design for applying an inclusive approach to achieve net-zero CO₂ emissions in the industrial sector, incorporating multiple decarbonization measures. The results suggest that the use of CO₂ for methane production has the potential to replace all natural gas demands considered in the base case and combined scenarios. However, using CO₂ utilization-based Fischer-Tropsch products alone to replace naphtha feedstock and transportation fuels is not sufficient to achieve complete decarbonization in the studied end uses. CO₂ utilization-based methanol could potentially substitute for several times the current U.S. methanol production and meet the current global demand for methanol. Moreover, the study conducts an economic analysis to estimate the costs of CO₂ utilization, which vary for different industrial sectors and depend on the technologies employed. Overall, this study provides valuable information for policymakers and industry stakeholders who are striving to develop effective strategies to decarbonize the industrial sector.

1. Introduction

According to United Nations (UN), climate change is humanity's "code red" warning (UN News 2021). The Paris Agreement aims to combat climate change by limiting the rise in global average temperature to less than 2 °C, and preferably to 1.5 °C, compared to pre-industrial levels (United Nations, 2023). Implementation of the Paris accord is also crucial for achieving the UN's sustainable development goals (SDGs) (United Nations, 2023) and provides a roadmap to building climate resilience. Countries that signed the historic agreement, including the United States (U.S.), are required to develop and implement national climate action plans to reduce greenhouse gas (GHG) emissions. The industrial sector accounts for almost one-third of the global total energy demand and related emissions (IEA, 2022). However, from a climate perspective, the industrial sector is considered "hard to abate" due to its long-lived capital equipment, lack of mature alternatives for many processes, and inadequate economic incentives for

technological change.

The U.S. industrial sector emitted over 825 million metric tons (Mt) of carbon dioxide (CO₂) into the atmosphere in 2018. These emissions account for 30 % of the total U.S. energy-related CO₂ emissions (U.S. DOE 2022). As a major world player, the U.S. aims to achieve net-zero CO₂ emissions in the industrial sector by 2050 and lead the global clean energy transition. However, the U.S. faces a range of structural and technical challenges in decarbonizing its industrial sector. This is a significant issue because the U.S. has a large portfolio of domestically manufactured industrial products, and decarbonization will require a wide range of technological solutions that may have a ripple effect across the country's complex supply chains (U.S. DOE 2022). Additionally, the U.S. aims to improve its manufacturing productivity and cost competitiveness on a global scale, develop innovative products, and meet expanding societal needs while reducing its carbon footprint. In this context, the U.S. department of energy (DOE) has developed an Industrial Decarbonization Roadmap (U.S. DOE 2022) to achieve

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net-zero CO₂ emissions in the industrial sector by 2050, while enhancing innovation and competitiveness.

Carbon capture, utilization, and storage (CCUS) is identified as one of the four key decarbonization pillars in the Industrial Decarbonization Roadmap, and it is critical to achieving the target of net-zero emissions from the industrial sector by 2050. However, carbon capture is primarily a mitigation strategy for industrial processes that are hard to abate using technologies other than carbon capture (U.S. DOE 2022). Captured CO₂ can be stored in oil and gas reservoirs, deep saline formations, coal beds, etc. (Hong, 2022) and can be utilized for several applications, including enhanced oil recovery, chemical feedstocks manufacture, plastics production, and as working fluids, etc. (Zhu, 2019; Alper and Yuksel Orhan, 2017; Burton et al., 2013). However, the potential impacts of each of these pathways are not well established in the literature. CO₂ utilization can be either direct or as a feedstock for further synthesis. According to estimates, direct utilization (e.g., beverage carbonation, food packaging, industrial gas, etc.) accounts for only 9 % of the small global CO₂ utilization market, while the majority is dominated by the synthesis pathway (Billig et al., 2019). Therefore, there is a need to investigate different CO₂ synthesis pathways and quantify their impacts. Given the diversity of these pathways, it is challenging to analyze them all at once, and detailed assessments are needed in steps to quantify the potential impacts of each pathway.

The CO₂ synthesis pathway can be further divided into chemical and biological processes. The biological processes are mostly based on different types of algae and bacteria. CO₂ can be utilized in these processes to produce biomass, while some organisms, such as cyanobacteria and green algae, may also emit hydrogen during the process (Aresta et al., 2013). However, biological routes are farther from commercialization at scale compared to chemical processes and are therefore outside the scope of this study. Chemical processes for CO₂ conversion to widely used products such as methane, methanol, and liquid hydrocarbons are technologically more mature (Billig et al., 2019; Gabrielli et al., 2020; König et al., 2015). The potential conversion of CO₂ to these high-value products also competes with traditional, energy-intensive processes for the manufacturing of these products, ultimately creating new markets and driving economic growth. Several studies in the literature have emphasized the importance of CO₂ capture from industrial sources and its conversion to high-value products like fuels and chemical feedstocks for intra-sectoral and inter-sectoral utilization.

König et al. (2015) investigated CO₂ capture and utilization (CCU) for Fischer-Tropsch (FT) fuel production using fluctuating wind power as a renewable energy source. They proposed a process concept utilizing a proton exchange membrane (PEM) electrolyzer for flexible hydrogen (H₂) production and a storage unit to manage energy fluctuations. The study included an economic analysis of the electrolyzer, H₂ storage units, and CO₂ conversion plant, but did not cover the techno-economic evaluation of CO₂ capture resources and technologies. Gabrielli et al. (2020) also studied the use of CO₂ captured directly from air (direct air capture – DAC) as a feedstock, together with green H₂ supplied via a PEM electrolyzer. They studied the pros and cons of the technology and assessed its feasibility for the specific case of methanol production. Their study concluded that high electricity demand for green H₂ production and high CO₂ intensity of electricity grids are the major challenges to the pathway's large-scale application. Graf et al. (2014) evaluated the status and techno-economic potential for the conversion of captured CO₂ to synthetic natural gas or methane using green electricity and H₂.

Billig et al. (2019) studied renewable CO₂ sources and analyzed their potential utilization for methanol, FT fuels, and methane production in Germany. Their work highlighted that sustainable and economic production of H₂ is one of the limiting factors to processing sustainable CO₂ into the studied products. The article concluded that the highest H₂ demand is caused by methanation while methanol and FT fuels production show almost the same rate of H₂ demand per unit of CO₂ conversion. However, their work does not offer any economic indicators for the three utilization pathways. More recently, Zang et al. (2021)

performed enviro-economic analysis to identify opportunities in U.S. regions to inform regional and national actions for CCU development. Their work studied the cost of producing synthetic methanol and FT fuels using CO₂ captured from selective industrial and power generation processes across the U.S. They concluded that a total of approximately 1600 Mt of fossil-based and process CO₂ can be captured annually and converted into 319 and 85 billion gallons of methanol and FT fuels, respectively, with the minimum selling prices of these products estimated at 1.8 – 2.8 times the price of the same products manufactured by conventional processes. While this study is one of the most comprehensive to date, the technological scope for CO₂ capture and conversion, and H₂ production is rather limited.

It is evident from the literature review that several researchers have analyzed multiple sources for CO₂ capture, including selective industrial processes and direct air capture, and explored its potential utilization for specific fuels and chemicals manufacture. However, the literature lacks sufficient information on the techno-economic evaluation and comparison of cutting-edge technologies for capturing CO₂ from diverse sources with fluctuating CO₂ concentrations, synthesizing it into fuels and essential chemical building blocks, and producing green H₂ in the context of CO₂ capture and utilization. Given these knowledge gaps, this article aims to contribute to the timely debate on the role of carbon capture in industrial decarbonization. It offers a comprehensive analysis and quantitative comparative assessment of different technologies for CCU pathways, covering 27 unique technology combinations across 16 industrial sectors. Moreover, to reach net-zero CO₂ emissions, a combination of different decarbonization measures in all manufacturing sectors will be required. Hence, this article builds on Zuberi et al. (2023, 2024) regarding the U.S. industrial decarbonization through process heat integration and electrification and studies CCU under different implementation scenarios in the U.S. as a case study. The specific aims and objectives are as follows:

1. Investigate CO₂ capture from different U.S. industrial sources and its utilization for synthetic methane, methanol, and Fischer-Tropsch (FT) fuels' manufacture.
2. Estimate energy demand for CO₂ capture and conversion as a function of manufacturing sector-specific flue gas composition.
3. Estimate green H₂ demand for CO₂ conversion and analyze different electrolyzer technologies and the corresponding energy demand for H₂ production.
4. Investigate and compare various technology combinations and their associated costs for the studied CCU pathways across different implementation scenarios.
5. Identify the barriers and drivers to the wide-scale application of the studied technologies and provide recommendations to overcome the barriers.
6. Apply an inclusive approach to achieve net-zero CO₂ emissions in the manufacturing sector by incorporating multiple decarbonization measures.

2. Methods and materials

2.1. CO₂ emissions from U.S. industrial sources

CO₂ emissions originate from numerous sources across all sectors of the economy. Most CO₂ emissions, not only in the U.S. but globally, mainly originate from the power, transportation, and manufacturing sectors. Due to the decentralized nature of the transportation sector, it is not suitable for carbon capture (Billig et al., 2019). As renewable energy continues to increase and displace thermal electricity generation, CO₂ emissions from industry will constitute an increasingly larger portion of global and U.S. emissions. When discussing CO₂ emissions from industrial sources, a distinction can be made between combustion-related and process-related emissions. Combustion-related emissions occur when carbonaceous fuels such as coal, natural gas, and petroleum products are

Table 1
CO₂ emissions and concentrations in flue gases from the U.S. manufacturing industries in 2018.

NAICS code	Manufacturing sector	2018 fuel demand for process heat ^{a,b} PJ	2018 Wt. fuel emission factor ^b tCO ₂ /TJ	2018 Combustion-related emissions MtCO ₂	2018 Process-related emissions ^c MtCO ₂	2018 Wt. avg. CO ₂ conc. in flue gases ^d vol%
3313	Alumina & aluminum	107	53.1	5	1.7	5 ^f
327,310	Cement	230	80.9	18	39.0	20 ^g
325	Chemicals	2316 ^e	59.3	128 ^c	45.8	10 ^h
334, 335	Computers & electronics	37	53.4	2	0	6
332	Fabricated metals	88	54.3	5	0	6
311, 312	Food & beverage	784	58.6	44	0	7
321, 322	Forest products	1988	82.2	142	13	6 ⁱ
3315	Foundries	43	59.8	2	0	6
3272, 327,993	Glass	135	54.4	7	2.0	6
331,110, 3312	Iron & steel	790	99.4	74	41.3	15 ^{aj}
333	Machinery	29	55.7	2	0	6
324,110	Refineries	2995	68.9	196	0	8 ^g
326	Plastics	61	54.1	3	0	6
313–316	Textiles	36	55.2	2	0	7
336	Transport equipment	99	54.8	5	0	6
–	Balance of manufacturing	482	64.7	31	16.8	7
	All manufacturing	10,219		679	146.6	

^a Demand includes fuels for both direct process heat supply and indirect heat supply, which includes steam and hot water from combustion boilers. However, the share of fuels for combined heat and power (CHP), which is representative of on-site electricity generation (approximately 12 %), is excluded.

^b The estimated values are based on U.S. MECS datasets (US EIA, 2021). For a detailed methodology for estimating the weighted average emission factors for each sector, refer to Zuberi et al. (2022).

^c The data is acquired from the Inventory of U.S. Greenhouse Gas Emissions and Sinks (US EPA 2022) except for forest products, for which the estimate is based on discussions with experts.

^d Unless otherwise stated, the values are estimated based on the typical CO₂ concentration in flue gases from fuel-based combustion processes given by Rodin et al. (2020) and U.S. sector-specific fuel mixes adapted from MECS (US EIA, 2021). In the combined scenario (see Section 2.2), where combustion-based CO₂ emissions could potentially decrease, a slightly higher estimate for CO₂ concentration in some sectors with process emissions is assumed.

^e Fuel demand and CO₂ emissions from U.S. methanol manufacturing are excluded to avoid double-counting. Fuel demand for U.S. methanol manufacturing is estimated based on Saygin et al. (2011) and the U.S. bandwidth study for chemicals manufacturing (US, 2015).

^f Despite the significant amount of process emissions, the flue gas coming out of Hall-Héroult cells is diluted with cooling air, leading to a low CO₂ concentration, which could range between 1 and >4 vol% depending on the cell design (Wang and Song, 2020; Jilvero et al., 2014; Springer and Hasanbeigi, 2016). However, a higher value of CO₂ concentration is assumed for the sector because the NAICS code also includes processes other than smelting for primary aluminum.

^g The assumed values are directly from Rodin et al. (2020).

^h Due to relatively high process-related CO₂ emissions from several manufacturing processes that are often mixed with combustion-related emissions, a higher value for the concentration is assumed to reflect high-purity CO₂ streams.

ⁱ A small amount of process CO₂ is emitted from the lime kiln in the Kraft process for pulp manufacturing, and the amount varies from mill to mill (Miner and Upton, 2002). These process emissions have high purity; however, the low CO₂ concentration in flue gases resulting from biomass combustion could make their effect on the weighted average CO₂ concentration of the sector insignificant.

^j According to Wang and Song (2020), the CO₂ concentration in flue gases from primary steel manufacturing (blast furnace) could range between 20 and 27 vol%. However, a conservative estimate is assumed for the sector because the NAICS code also includes processes other than primary steel.

burned for energy applications. Process-related emissions occur from chemical reactions that are required to produce a desired product, such as the manufacturing of petrochemicals, metals, cement, lime, etc. (Bains et al., 2017). The combustion- and process-related CO₂ emissions from the manufacturing sector in 2018 are presented in Table 1. The estimates in Table 1 are based on the EIA's 2018 Manufacturing Energy Consumption Survey (MECS) (US EIA, 2021), Manufacturing Energy and Carbon Footprints (US DOE/Energetics 2022), and Inventory of U.S. Greenhouse Gas Emissions and Sinks (US EPA 2022). It should be noted that the MECS datasets are published quadrennially, and the latest datasets for 2018 were released in 2021. Furthermore, the combustion-related emissions are estimated using the weighted average emission factors calculated based on the sectoral fuel mixes reported by the MECS datasets (US EIA, 2021). Finally, CO₂ emissions from off-site energy generation facilities such as power plants are excluded in this study for reasons explained in Section 2.2.

Energy demand and the cost of CO₂ capture from various industrial sources depend on their concentration and partial pressures in flue gases. Wang and Song (2020) have presented that higher CO₂ concentration in flue gases generally correlates with higher partial pressures. In the case of pressurized combustion, the partial pressure of CO₂ is much

higher (Kothandaraman et al., 2009). It is important to note that further research and data verification are needed to establish a conclusive relationship. Moreover, higher CO₂ concentration in flue gas composition is associated with lower energy demand and reduced costs for its separation (Bains et al., 2017) (refer to Section 2.4 for details). The CO₂ concentration in the flue gas depends on the fuel type and combustion conditions, such as the amount of excess air (Ghiat and Al-Ansari, 2021). In several cases, combustion- and process-related CO₂ emissions can occur within the same manufacturing unit. When these emissions are mixed, there is the potential for higher-purity CO₂ streams, as in the case of lime or cement manufacturing (Bains et al., 2017). The average CO₂ concentrations by volume in flue gas streams coming out of different U.S. manufacturing sectors are also presented in Table 1. The CO₂ concentration levels in the U.S. cement (20 vol%), iron and steel (15 vol%), and refinery (8 vol%) sectors are assumed directly from the literature (Wang and Song, 2020; Rodin et al., 2020). Rodin et al. (2020) also provide ranges of CO₂ purities by volume from coal-, natural gas-, fuel

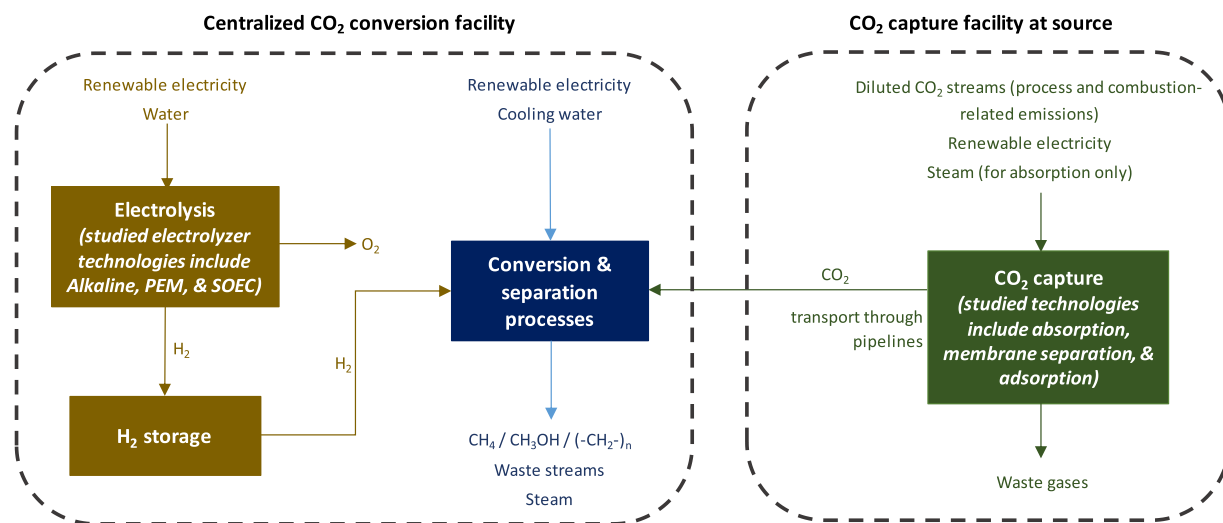


Fig. 1. System boundary and block flow diagram of the process concept.

oil-, and biomass-based combustion processes.¹ These estimates, together with the sector-specific fuel mixes, are then used to conservatively estimate the weighted average CO₂ concentration levels in the remaining U.S. manufacturing sectors, as shown in Table 1, with an exception for the chemical sector.

Considering the by-product CO₂ generated from industrial processes such as ethanol fermentation, steam-methane reforming (SMR) for ammonia, etc. (Zang et al., 2021), a high CO₂ concentration of 10 vol% in flue gases is conservatively assumed for the entire chemical sector, partly based on expert judgment. Ideally, the flue gas concentration for the overall chemical industry would be estimated as a weighted average of the concentrations for individual chemical processes. However, given that the U.S. chemical industry manufactures over 70,000 distinct chemical products (CISA, 2022), it is impossible to precisely estimate the weighted average flue gas concentration for the sector. Therefore, crude information on the major basic chemicals was leveraged to make an informed judgment on the concentration estimate. However, this approach may be considered conservative. For some chemical sub-sectors, such as ethylene and ammonia, the flue gas concentrations could be significantly higher than assumed. This would result in lower energy demand and costs for CO₂ capture. Furthermore, regardless of the initial CO₂ concentration from each sector, the captured CO₂ for utilization is assumed to have the same purity.

2.2. System boundaries and scenarios

While captured CO₂ can be stored or utilized for various applications, this study focuses specifically on the possible use of CO₂ as a base resource in combination with H₂ for the synthesis of chemical energy carriers. This pathway unlocks the possibility of manufacturing a broad range of sustainable fuels and chemicals. Due to the scope of this study, not all possibilities are considered. Instead, this study focuses on the production of methane, methanol, and liquid hydrocarbons, or Fischer-Tropsch (FT) fuels, from renewable electricity and CO₂. These three routes are among the most promising and technically matured routes for CO₂ utilization (Billig et al., 2019) (see Section 2.3 for brief process

¹ Since the carbon-neutrality of biomass fuels is debated due to concerns about the origin of biomass feedstock supply, its sustainable aspects, and whether the associated air-quality impacts from biomass utilization are tolerable, this study does not consider biomass fuels as carbon-neutral. On the contrary, capturing CO₂ emissions from biomass combustion processes can also be seen as negative emissions and/or partly as compensation for CO₂ capture inefficiencies.

descriptions). The block flow diagram, system boundary of the process concept, and studied technologies are presented in Fig. 1. The system boundary includes modeling of potential CO₂ capture facilities at sources such as manufacturing plants and centralized CO₂ conversion facilities, including on-site H₂ production. The study focuses on post-combustion CO₂ capture technologies that are discussed in Section 2.4. Pipeline transportation is considered the most cost-effective method for transporting large-volume CO₂ to nearby conversion facilities (Zang et al., 2021).

As illustrated in the figure, the conversion of CO₂ into chemical products necessitates feedstock H₂. Conventionally, H₂ is produced through steam-methane reforming (SMR), with a significant portion of its capacity allocated to refinery operations, ammonia, and methanol production. Currently, SMR with CCS, also known as blue hydrogen, is being discussed as one of the options for clean H₂ production. Retrofitting the existing SMR capacity within the refinery and chemical sectors for CO₂ capture, transport to conversion facilities, and utilization is being considered. However, for utilization purposes at the centralized facility, feedstock H₂ must be sourced from cleaner pathways, such as water electrolysis utilizing clean electricity. While future demand for the studied chemical products may involve SMR with CCS, these facilities would have their own captured CO₂ from their H₂ production units available for utilization, thus having no impact on CO₂ emissions reduction from the other existing industrial processes. Given the focus of this study on capturing and utilizing current CO₂ emissions, water electrolysis and its corresponding electrolyzer technologies, as listed in Fig. 1, are analyzed. These electrolysis technologies necessitate renewable electricity for sustainable operation. Furthermore, as fluctuating renewable electricity requires a flexible electrolyzer unit for H₂, a buffer storage unit acts as the link between the fluctuating renewable source, the electrolyzer unit, and the continuous CO₂ conversion. In other words, H₂ is stored if excess renewable electricity is available and used when the demand exceeds its production (König et al., 2015).

The energy required to operate CCUS facilities, as depicted in Fig. 1, is sourced from electricity (electrified CCU pathways; see Sections 2.3–2.5 for details). If this electricity comes from renewables (green electricity), the operation of these facilities will result in zero CO₂ emissions. This assumption is validated by the fact that the U.S. administration has set an ambitious target to generate 100 % carbon-free electricity by 2035 (The White House, 2021). Consequently, it is projected that by 2035, the U.S. national average electricity grid emission factor will be zero. It must be noted that electricity grid decarbonization, as well as the estimated CO₂ capture from the U.S. industrial sectors and its utilization for fuels and chemicals manufacture, are based

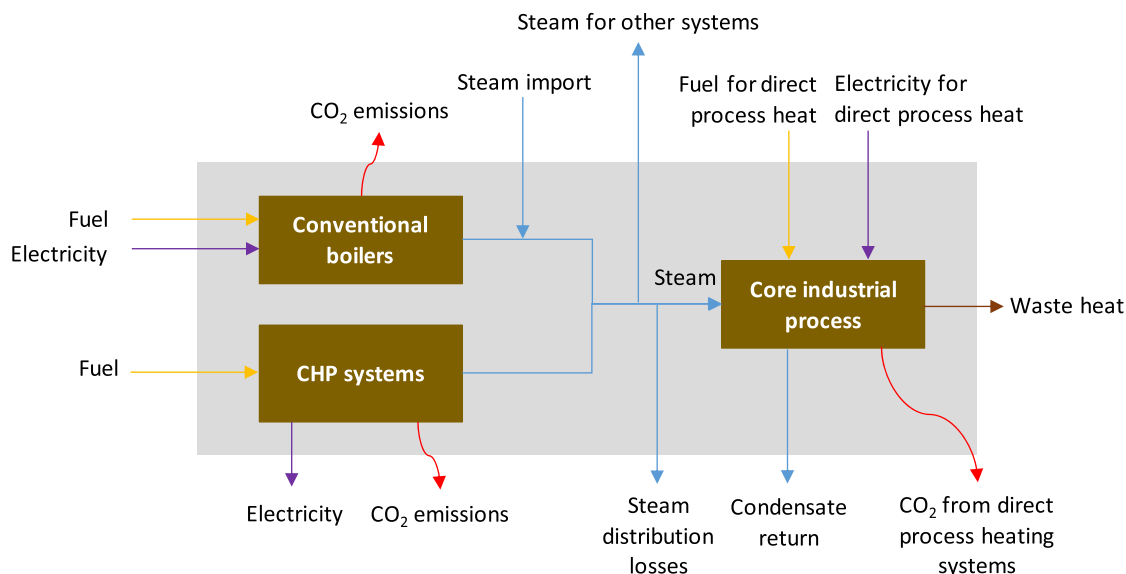


Fig. 2. CO₂ emissions from process heating systems in the *base case* scenario (flows in purple are for electricity, yellow for fuels, blue for steam, maroon for waste heat, and red for CO₂).

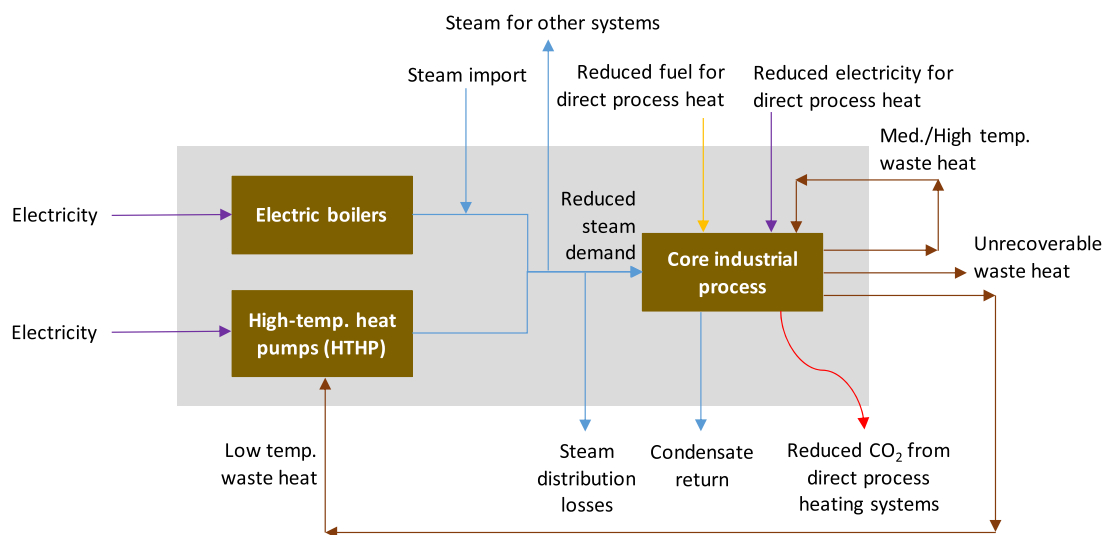


Fig. 3. CO₂ emissions from modified process heating systems in the *combined* scenario (flows in purple are for electricity, yellow for fuels, blue for steam, maroon for waste heat, and red for CO₂).

on technical potentials assuming an adoption rate of 100 %. However, the actual adoption of CCU pathways in U.S. manufacturing will be gradual over time. The temporal aspects regarding the adoption of CCU technologies are currently outside the scope of this work. Furthermore, the economic assessment includes costs estimated for CO₂ capture and transportation through pipelines, electrolyzers, H₂ storage units, and CO₂ conversion plants, encompassing auxiliary units and utilities.

While Fig. 1 presents the schematic of the process concept for CCU, it is essential to also comprehend the end-use operations from which CO₂ is primarily emitted. Fig. 2 presents a schematic of CO₂ emissions from a typical industrial process heating system. The U.S.-specific combustion-related CO₂ emissions in Table 1 are representative of this schematic. In the 'base case scenario', all the combustion- and process-related CO₂ emissions in Table 1 are considered for potential capture and utilization. However, there are different decarbonization options available to reduce the number of CO₂ emissions from manufacturing sectors, including energy efficiency improvement, electrification, process integration, etc. Ideally, CCU should be the last option when other

opportunities for limiting CO₂ emissions have been exhausted. For example, recovering waste heat from industrial processes for reuse to minimize fossil-fuel combustion could be techno-economically a more feasible option than CCUS for decarbonization. As the authors are currently exploring the electrification of industrial steam systems and heat integration within U.S. manufacturing as part of another study, this research leverages the preliminary findings from that study to examine an additional scenario, termed the 'combined scenario'. This scenario delineates a design aimed at adopting a comprehensive approach to attain net-zero CO₂ emissions in the industrial sector, integrating various decarbonization measures.

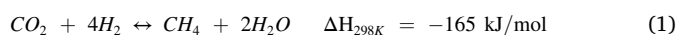
In the 'combined scenario', a reduced number of CO₂ emissions

(approximately 30 % lower combustion-related emissions than in 2018,² refer to Table 1) are available for capture. Fig. 3 presents the schematic for the combined scenario, showing resultant combustion-related CO₂ emissions after the potential for a) electrified steam supply through electric boilers and high-temperature heat pumps, and b) heat integration is exploited. The magnitude of process-related emissions is assumed to be the same in both scenarios.

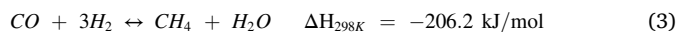
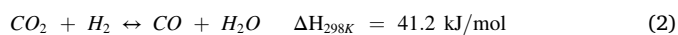
2.3. CO₂ utilization pathways for the production of carbon-rich products

2.3.1. Power-to-Gas (PtG): methane

Methane, in the form of natural gas, is widely used for heat supply, thermal power generation, and as a feedstock in the industrial sector. Natural gas is primarily extracted from subsurface rock formations via drilling (U.S. DOE March 24, 2023). However, methane gas can also be produced synthetically through the Sabatier reaction, which involves reacting CO₂ with H₂ (refer to Eq. (1)) (Bailera et al., 2017). The production of methane through the use of captured CO₂ and renewable H₂ produced via water electrolysis (Power-to-Gas – PtG) is a promising pathway for utilizing CO₂ from industrial sources. If synthetic methane is used for process heat supply in manufacturing, it could potentially close the carbon loop of an industrial plant.



The conversion of CO₂ to methane typically involves the use of nickel-based catalysts due to their good selectivity. The reaction is highly exothermic and occurs at relatively low temperatures ranging from 200 to 450 °C. It can be carried out at atmospheric pressure, although higher pressures result in a higher conversion of CO₂ (Dias and Perez-Lopez, 2021). For each cubic meter of methane produced per hour at standard temperature and pressure (STP), 1.8 kW of heat is released, which can be integrated into the plant (Rönsch et al., 2016). Other prominent reactions taking place inside the methanation reactor include the water-gas shift reaction, where CO₂ is transformed into carbon monoxide (CO) through Eq. (2), followed by hydrogenation to methane according to Eq. (3) (Dias and Perez-Lopez, 2021; Rönsch et al., 2016). The quality of the reactions taking place inside the reactor depends on the pressure, temperature, and infeed ratio of the reactants (Billig et al., 2019). Table 2 presents mass and energy flows of the methanation (or CO₂ conversion) process based on the results of chemical process simulations reported by Billig et al. (2019).



² The process heat integration potential is a conservative estimate, excluding the potential possessed in pulp and paper manufacturing. Paper manufacturing stands out due to more than two-thirds of the sector's process heat demand being in the form of steam, a significant portion of which is generated by utilizing byproduct waste, including biomass (black liquor and wood waste) (Zuberi et al., 2022, Sagues et al., 2020). The combustion of black liquor for steam production plays a crucial role in the chemical recovery and regeneration cycle. Additionally, wood waste must be combusted for energy, as it poses a potential source of methane emissions due to its high degradable organic carbon content (Pier and Kelly, 1997). These factors raise concerns and make the electrified steam supply through an industrial heat pump that utilizes low-quality plant waste heat challenging. Although (Zuberi et al., 2023) have demonstrated the potential application of heat pumps for paper drying, a more comprehensive analysis is required to precisely estimate the potential waste heat recovery and heat pump integration. Consequently, the sector-specific potential has not been studied in the combined scenario, and the CO₂ emissions (including biogenic CO₂) from the sector are considered the same in both scenarios.

Table 2

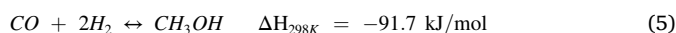
Energy and mass flows for CO₂ conversion to different products based on Billig et al. (2019); König et al. (2015).

	Product	CO ₂	H ₂	Electricity*
<i>For a kg of Methane</i>				
Mass (kg)	1.0	2.8	0.5	
Energy (MJ)	50.0		60.1	1.4
<i>For a kg of Methanol</i>				
Mass (kg)	1.0	1.4	0.2	
Energy (MJ)	19.9		22.6	1.0
<i>For a kg of FT fuels</i>				
Mass (kg)	1.0	4.2	0.5	
Energy (MJ)	44.1		64.0	2.2

* Electricity demand solely for conversion, excluding electricity demand for H₂ production and CO₂ capture.

2.3.2. Power-to-liquid (PtL): methanol

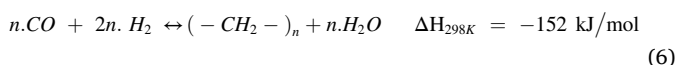
Methanol is a petrochemical mainly used as a feedstock and solvent for producing several other chemicals, including acetic acid, formaldehyde, fuel additives, and methylamines (Ghiat and Al-Ansari, 2021). The conventional method of manufacturing methanol is a chemical synthesis based on syngas (a mixture of carbon monoxide - CO and H₂), which is mainly produced through steam-methane reforming (SMR). Using renewable power to sustainably produce green H₂ via electrolysis and reacting it with captured CO₂ for methanol synthesis (Power-to-Liquid Methanol – PtL) is another manufacturing pathway and a potential solution for CO₂ utilization. CO₂ hydrogenation for methanol consists of multiple reactions occurring simultaneously, i.e., the methanol synthesis reaction based on CO₂ in Eq. (4) is commonly accompanied by the reactions in Eqs. (2) and (5) (Borisut and Nuchitprasittichai, 2019; Pontzen et al., 2011).



The synthesis reactions take place in an isothermal reactor with a Cu-Zn-Al oxide catalyst and at temperatures and pressures of 250 °C and 65–80 bar, respectively (Battaglia et al., 2021). Through liquid-gas phase separation, all unreacted CO₂ and CO are separated from methanol and redirected to the synthesis reactor (Billig et al., 2019). The heat demands for crude methanol preheating and the distillation column reboiler can be met by integrating heat from the synthesis reactor; hence, no additional thermal energy is required for the process (Atsonios et al., 2016). Table 2 also presents the specific mass and energy balances of methanol synthesis based on the process simulations given by Billig et al. (Billig et al., 2019).

2.3.3. Power-to-liquid (PtL): fischer-tropsch fuels

Fischer-tropsch (FT) synthesis is one of the most prominent routes for producing liquid hydrocarbons. FT synthesis catalytically converts a mixture of CO and H₂ (syngas) into several hydrocarbon products of variable chain length (ranging from C₅ to C₂₀), including gasoline, kerosene, and diesel fuel (Hannula et al., 2020; Dimitriou et al., 2015). Since FT synthesis is a catalytic process, the hydrocarbon products are free of sulfur and produce less soot when combusted (Dimitriou et al., 2015). The catalysts typically used for FT synthesis are Fe- or Co-based. The FT reaction is highly exothermic and can be represented by the following basic equation (König et al., 2015):



FT synthesis could employ steam-reforming of natural gas or biomass gasification. However, when CO₂ and renewable power are used to produce liquid FT fuels, the process is called Power-to-Liquid (PtL - FT fuels). All of these processes mainly differ in terms of syngas production and H₂-to-CO ratio (Billig et al., 2019). When CO₂ is used as the carbon

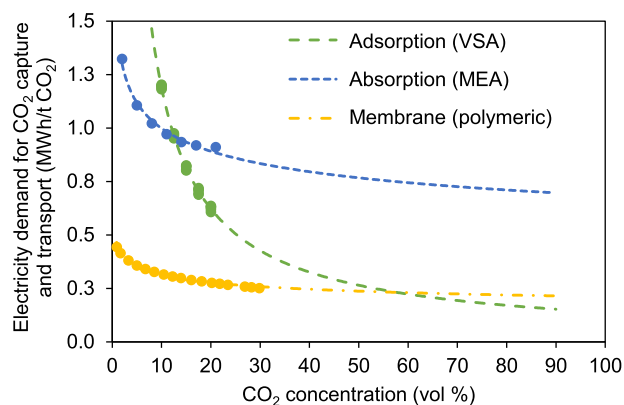


Fig. 4. Electricity demand of post-combustion CO₂ capture technologies as a function of CO₂ concentration in flue gases (dashed lines are extrapolated based on the data points acquired from the literature discussed in the relevant sections).

source, a two-step process typically occurs, combining the reverse water gas shift reaction (Eq. (2)) and FT synthesis (Eq. (6)). Since Fe-based catalysts result in lower liquid selectivity, the water gas shift reaction can be minimized when using a Co-based catalyst (Dimitriou et al., 2015). Furthermore, the FT process is generally operated at temperatures and pressures ranging from 200 to 350 °C and 20–40 bar, respectively, depending on the desired product yields. Generally, FT synthesis must be carried out at relatively low temperatures, high pressures, and high H₂-to-CO ratios to maximize FT products (Dimitriou et al., 2015). The liquid hydrocarbons (or FT fuels) are separated from unreacted feed and gaseous hydrocarbons and upgraded through hydrocracking and isomerization to yield the final products (Billig et al., 2019). König et al. (2015) studied the cooling and heating requirements using pinch analysis and demonstrated that the entire heat demand of the process can be supplied through internal heat exchange. The specific mass and energy balances of a model FT synthesis, based on the process simulated by König et al. (2015), are presented in Table 2.

2.4. CO₂ capture technologies

Three CO₂ capture systems associated with combustion processes are post-combustion, pre-combustion, and oxyfuel combustion. Post-combustion is preferred for retrofitting existing plants and has been extensively developed (Ghiat and Al-Ansari, 2021; Gruenewald and Radnjanski, 2016). However, the major challenge for post-combustion CO₂ capture is its significant parasitic energy load. Since the CO₂ concentration in flue gases is normally quite low (refer to Table 1), the energy penalty and associated costs for the capture unit to reach a high CO₂ concentration needed for transport, storage, and/or utilization are elevated (Leung et al., 2014). This study focuses on selective post-combustion technologies for CO₂ capture before transportation, including absorption (amine scrubbing - AS), membranes, and adsorption techniques

Monoethanolamine (MEA) solution is the benchmark amine due to its good CO₂ transfer rates, relatively low cost, and biodegradability (Wang and Song, 2020), and it is hence considered as the reference technology for CO₂ capture through absorption. As shown in Fig. 4, the energy demand of this technique decreases exponentially as the CO₂ concentration increases, leveling off after a certain point. The energy demand, which encompasses the stripper, gas blower, and auxiliary equipment, has been estimated using various literature sources (Husebye et al., 2012; Moullec and Kanniche, 2011; Merkel et al., 2013). The figure suggests that higher CO₂ concentration streams require less energy per unit of CO₂ captured. However, the high energy consumption associated with amine-based CO₂ capture mainly results from solvent regeneration in the solvent stripper, where a significant amount of water

Table 3

Specific electricity demands of various electrolysis technologies assumed for green H₂ production, based on (Panzone et al., 2020; Brynolf et al., 2018).

Electrolysis technology	Assumed electricity demand (kWh/kg H ₂)
Alkaline electrolysis	63.1
Proton exchange membrane (PEM) electrolysis	52.5
Solid-oxide electrolyzer cell (SOEC)	43.2

is stripped out during CO₂ release from the liquid phase (Metz et al., 2005). To release CO₂ from the solvent, steam can be generated from various sources, depending on the available heat and energy sources. These sources may include electrical or combustion boilers powered by natural gas or the recovery of heat from industrial processes. The cost of steam production is directly related to energy commodity prices and the efficiency of steam generation (Husebye et al., 2012). In this study, we consider electric boilers for steam generation due to their high efficiency (i.e., 99 % compared to around 80 % for gas boilers (Zuberi et al., 2022)) and potential environmental benefits resulting from grid decarbonization in the future.

Since polymer electrolytic membrane (PEM) offers advantages such as lower production cost, relatively high gas flux, and mechanical stability (Madejski et al., 2022), this study focuses on the use of PEM for CO₂ capture through membrane separation (MS). Similarly, due to its relatively low electricity consumption and no heat requirements (Zhao et al., 2017), vacuum swing adsorption (VSA) has been chosen as the adsorption technique for CO₂ capture in this study. Fig. 4 also shows the electricity demand of membrane separation and adsorption technique as a function of CO₂ concentration in flue gases. The energy demand for membrane separation is estimated based on the information given by Merkel et al. (2013); Han et al. (2020), while for adsorption, it is estimated as the average of different adsorbent materials reported by Zhao et al. (2018).³ Although the curve exhibits a similar trend to the absorption technique, the membrane separation method has a lower absolute energy demand, and this demand is not estimated to vary significantly for larger concentrations. Similar to absorption and membrane technologies, the energy demand for adsorption also decreases with increasing CO₂ concentration. Further discussion on the current status of technologies within each technique is provided in detail in Appendix A.

2.5. Green H₂ production technologies

To minimize life cycle emissions, it is imperative that H₂ is supplied to processes in a carbon-free manner. One effective method is by utilizing H₂ produced from water electrolysis using renewable electricity sources, commonly known as green or renewable H₂ (Pérez-Fortes et al., 2016). There are currently three major electrolyzer technologies for producing green H₂: alkaline electrolyzer (AE), proton exchange membrane (PEM) electrolyzer, and solid oxide electrolyzer cell (SOEC). At present, AE stands out as the most developed technology with a relatively lower level of capital investment (Panzone et al., 2020), making it the reference technology for H₂ production in this study. However, AE comes with several drawbacks, including slow start-up, susceptibility to corrosion, complex maintenance requirements, and a multitude of components (Guo et al., 2019). On the other hand, PEM and SOEC electrolysis, though less mature and costlier compared to AE, offer higher flexibility and the ability to operate at higher current densities, potentially reducing the costs of H₂ production (Panzone et al., 2020). PEM electrolysis holds promise, particularly in flexible energy storage applications, and is in the early stages of commercialization for

³ The adsorbent materials studied by Zhao et al. (2018) include activated carbons, silica gels, zeolites 5A and 13X, and metal-organic frameworks (MOF).

Table 4
Capital costs of CO₂ capture, CO₂ transport, H₂ production and conversion plants.

Plant type	Capital costs	Unit	Data source(s)
<i>CO₂ capture</i>			
Amine scrubbing (MEA)	144.2	\$/tCO ₂ capture	(Tuinier et al., 2011)
Membrane separation (PEM)	286.5	\$/tCO ₂ capture	(Tuinier et al., 2011)
Adsorption (VSA) ^a	198.0	\$/tCO ₂ capture	(Susarla et al., 2015)
<i>CO₂ compression and transport</i>			
Product CO ₂ compressor	10.8	\$/tCO ₂ capture	(Tuinier et al., 2011)
Onshore CO ₂ pipeline (25 in. pipe dia ^{b,d})	6.4	\$/tCO ₂ transport	(Morgan et al., 2018; Smith, 2021)
Onshore CO ₂ pipeline (20 in. pipe dia ^{c,d})	10.3	\$/tCO ₂ transport	(Morgan et al., 2018; Smith, 2021)
<i>H₂ production</i>			
Alkaline electrolyzer (AE)	2.6 (1100)	\$/GJ of H ₂ prod. (\$/kW _e) ^f	(Detz et al., 2018)
Polymer electrolyte membrane (PEM)	3.7 (1500)	\$/GJ of H ₂ prod. (\$/kW _e) ^f	(Detz et al., 2018)
Solid oxide electrolyzer cell (SOEC) ^e	3.3 (2000)	\$/GJ of H ₂ prod. (\$/kW _e) ^f	(Detz et al., 2018)
<i>PtX (CO₂ conversion)</i>			
Methanation	0.7 ^g	\$/GJ of CH ₄ prod.	(Kiani et al., 2021)
Methanol synthesis	1.1	\$/GJ of CH ₃ OH prod.	(Detz et al., 2018)
Fischer-Tropsch synthesis (FT)	1.0	\$/GJ of FT fuels prod.	(Detz et al., 2018)

^a The capital costs are representative of a 13X zeolite-based CO₂ adsorption system (Susarla et al., 2015).

^b Assuming 20 MtCO₂ transport within a 100-mile radius per annum in the base case scenario.

^c Assuming 10 MtCO₂ transport within a 100-mile radius per annum in the combined scenario.

^d Based on the mean capital costs of \$51,581/in-mile given by Morgan et al. (2018); Smith (2021).

^e According to Detz et al. (2018), the capital costs of SOEC technology per unit of H₂ produced are slightly lower than those of PEM technology due to SOEC's higher efficiency.

^f kW_e = kW electricity input.

^g The value is significantly more conservative than the estimates given by Graf et al. (2014); Kiani et al. (2021); Baier et al. (2018).

high-power applications (Panzone et al., 2020). Similarly, SOECs demonstrate significant potential for large-scale H₂ production and energy storage applications (Di Giorgio and Desideri, 2016). While currently utilized at kW scales, recent projects have showcased their capability to operate at an MW scale (Taibi et al., 2020). Additional generic technical details on each of these technologies are provided in Appendix B. Table 3 outlines the specific electricity demand of the three studied electrolysis technologies for producing a unit of H₂.

2.6. Chemical conversion and plant efficiencies

To assess the energetic performance of CO₂ conversion processes, this study estimates both the chemical conversion efficiency and the plant efficiency. The chemical conversion efficiency, $\eta_{CE,X}$, balances energy flows from the reactant H₂ and the chemical product X, as shown in Eq. (7) (Billig et al., 2019). The plant efficiency, η_P , balances all energy flows within a synthesis plant, including reactant and product energy flows and energy for plant operations, as shown in Eq. (8) (König et al., 2015). The energy flows of input and output streams are calculated as the product of the mass flow rate (\dot{m}) and the lower heating values (LHV) of the respective streams.

$$\eta_{CE,X} = \frac{LHV_X \times \dot{m}_X}{LHV_{H_2} \times \dot{m}_{H_2}} \quad (7)$$

Where;

$\eta_{CE,X}$ = Chemical conversion efficiency

LHV = Lower heating value (MJ/kg)

\dot{m} = Mass flow (kg per annum)

$$\eta_P = \frac{LHV_X \times \dot{m}_X}{(LHV_{H_2} \times \dot{m}_{H_2}) + P_E} \quad (8)$$

Where;

η_P = Plant efficiency

P_E = Annual energy input to conversion plants

2.7. Costs of CO₂ capture and utilization

An economic analysis of three CCU pathways is presented in this study. The cost of each pathway was calculated using Eq. (9), which sums up all cash inflows, including investments, operations and

maintenance (O&M) costs, and electricity costs, while the denominator represents the amount of CO₂ captured for utilization. Eq. (10) is used to calculate the capital recovery or annuity factor, which depends on the discount rate and project lifetime. A discount rate of 10 % (from the private perspective; see the discussion on social vs. private discount rate in Zuberi et al. (2017)), a project lifetime of 15 years (conservative estimate), and a CO₂ capture efficiency of 90 % (as assumed in many studies (Gabrielli et al., 2020; Zang et al., 2021; Boot-Handford et al., 2013; P.C. Psarras et al., 2017)) are chosen. The cost components in Eq. (9) and their corresponding assumptions are discussed in detail below.

$$C_{CCU,s} = \frac{\alpha I_s + O\&M_s + (E_{l,s} \times P_{el})}{CO_{2,s} \times \gamma} \quad (9)$$

Where;

$C_{CCU,s}$ = Costs of CO₂ capture and utilization in a sector s

α = Capital recovery factor or annuity factor (see Eq. (10))

I_s = Capital costs of CCU in a sector s (see Eq. (11) later in this section)

$O\&M_s$ = Annual operations and maintenance costs in a sector s

$E_{l,s}$ = Potential electricity demand for CCU in a sector s

P_{el} = National average electricity price for industry

$CO_{2,s}$ = Onsite CO₂ emissions in a sector s

γ = CO₂ capture rate (taken as 90 %)

$$\alpha = \frac{(1+r)^L \times r}{(1+r)^L - 1} \quad (10)$$

Where;

r = Real discount rate (taken as 10 % from the private perspective)

L = Lifetime of CCU (taken as 15 years)

The capital investment costs for the studied CCU pathways consist of the costs of CO₂ capture, transport infrastructure, H₂ production, and power-to-liquid/gas (PtX) or conversion plants. These costs are estimated using Eq. (11) and are presented in Table 4, based on literature sources. The capital investments required for CO₂ capture typically depend on the plant size. Industrial sectors with lower CO₂ emissions (e.g., food, textiles, machinery) may require higher investments for CO₂ capture due to the lack of economies of scale compared to energy-intensive sectors (e.g., cement, steel, chemicals). To estimate the size-specific capital costs of CO₂ capture technologies for industrial sectors, this study employs a cost scaling factor method. It is assumed that the

capital cost of each CO₂ capture technology in Table 4 is representative of the average-sized CO₂ capture plant per industrial facility (or establishment). The cumulative CO₂ emissions and those from each sector are provided in Table 1, while the data for the total number of establishments and those in each industrial sector are obtained from the (U.S. Census Bureau (accessed April 5, 2022) 2018).⁴ Using Eqs. (12) and 13, specific costs of each capture technology are scaled for each sector.

$$I_s = I_{cc,s} + I_T + I_H + I_{PtX} \quad (11)$$

Where;

$I_{CC,s}$ = Capital investment cost of CO₂ capture in a sector s

I_T = Capital investment cost of CO₂ transport infrastructure

I_H = Capital investment cost of an H₂ production plant

I_{PtX} = Capital investment cost of PtX (or CO₂ conversion) plant

$$I_{CC,s} = \frac{I_{CC,e,s}}{CC_{e,s}} \times CC_s \quad (12)$$

Where;

$I_{CC,e,s}$ = Capital investment cost of CO₂ capture per average-sized establishment e in a sector s

$CC_{e,s}$ = Total amount of potential CO₂ captured per average-sized establishment e in a sector s

CC_s = Total amount of potential CO₂ captured in a sector s

$$I_{cc,e,s} = I_{cc,m} \times \left(\frac{CC_{e,s}}{CC_m} \right)^k \quad (13)$$

Where;

$I_{CC,m}$ = Capital investment cost of CO₂ capture per average-sized industry establishment e

CC_m = Total amount of potential CO₂ captured per average-sized industry establishment e

k = Scaling exponent, assumed to be 0.7 based on (Chopey, 2004)

Most industrial plants are assumed to capture CO₂ and transport it to a central location for conversion (see Fig. 1). Although H₂ production and PtX plants may also benefit from economies of scale, their size is relatively independent of sector-specific CO₂ emissions due to centralized production. Therefore, it is reasonable to use the average capital costs for potential hydrogen and conversion plants presented in Table 4 for the economic analysis. Additionally, the overall capital costs are dependent on the CO₂ utilization pathway used to manufacture a specific product and the technologies adopted in the process. This study calculates investment costs for all pathways based on the literature.

CO₂ transport via pipeline is the most cost-effective mode of transport for most scenarios, except when CO₂ needs to be transported over large distances (>1000 km) or bodies of water, where transport by ship may be more economical (Smith, 2021). Transport via rail or road is only feasible for moving CO₂ on a small scale, for specialist applications (Psarras et al., 2017). The choice of transport depends on the quantity of CO₂, distance, terrain, and CO₂ stream specifications (Boot-Handford et al., 2013). This study focuses on transporting CO₂ via pipeline from the source to the conversion facility within a 100-mile radius. In the U. S., several proposed industrial CO₂ capture facilities are being evaluated for the purpose of enhanced oil recovery. Many of these capture facilities are strategically planned within a moderate distance of less than 100 miles from potential utilization sites. Consequently, the construction of these facilities will incorporate pipelines designed to transport CO₂ directly to these utilization sites (Callahan et al., 2014). While some studies have examined the possibility of longer CO₂ pipeline constructions, others have focused on the viability of a 100-mile onshore CO₂ pipeline transport for various purposes (Smith, 2021; National

Petroleum Council, 2019; Psarras et al., 2017). Although geospatial mapping of CO₂ resources is not within the scope of this study, the potential CO₂ captured from point sources is assumed to be transported within a 100-mile radius for conversion into fuels and chemical feedstocks, with costs conservatively estimated for a maximum 100-mile pipeline.

Furthermore, CO₂ is compressed to a supercritical fluid above its critical point (31.1 °C and 74 bar) before being transported through pipelines. Supercritical CO₂ is ideal for pipeline transport as it has the high density of a liquid and the favorable flow characteristics of a gas. To prevent operational issues, such as liquid slugs, pressure drops must be managed, and pipeline pressures must remain above vapor-liquid equilibrium conditions to maintain a single-dense-phase flow. Existing CO₂ pipelines operate within a pressure range of 85 to 210 bar to ensure CO₂ remains in the dense-phase state over a wide temperature range. Intermediate pumping or booster stations are necessary to maintain sufficiently high pressures. Electricity demand for CO₂ compression and transport using booster pump(s) is taken as 0.08 MWh/tCO₂ (Merkel et al., 2013) and 0.04 MWh/tCO₂ (accessed April 5, 2022; McCollum and Ogden, 2006), respectively, and it is independent of industrial flue gas compositions. For shorter distances, increasing the pipeline inlet pressure may be a viable alternative to booster stations. However, this approach requires more energy for compression and thicker-walled pipelines, making it less economical (Boot-Handford et al., 2013). The capital costs of CO₂ compression and transport through pipelines are also presented in Table 4. It should be noted that transportation costs for CO₂ have an inverse relationship with the flow rate (Zang et al., 2021). Therefore, in the combined scenario with lower flow rates, transportation costs will be higher compared to the base case scenario, as indicated in Table 4.

The annual O&M costs for CO₂ transport, H₂ production, and PtX plants are assumed to be 5 % of the total initial investment costs based on Detz et al. (2018) and Brynolf et al. (2018). However, the O&M costs for CO₂ capture using amine-based absorption, membrane, and adsorption technologies, as provided by Tuinier et al. (2011) and Susarla et al. (2015), are assumed to be higher at 25 %, 15 %, and 15 %, respectively. These costs are assumed to remain constant over the lifetime of the plants and include replacement costs for installed capital, such as sorbents and catalysts. The average electricity price assumed for the U.S. industry during the project's lifetime is \$0.08/kWh, based on the U.S. Annual Energy Outlook (AEO) (U.S. EIA, 2022). To test for price sensitivity, the study also examines a lower electricity price of \$0.06/kWh (approximately 25 % lower), based on the AEO low zero-carbon technology cost scenario in 2050.

3. Results and discussion

3.1. CO₂ utilization for fuels and chemicals manufacture

In the following subsections, the results of the three CCU pathways are presented under the 'base case scenario', where no industrial decarbonization measures have been implemented prior to CO₂ capture (refer to Fig. 2). The next section examines the results of the 'combined scenario', which investigates CCU in conjunction with electrification and heat integration (refer to Fig. 3).

3.1.1. Potential methane manufacture

The maximum theoretical methane generation potential can be estimated by using CO₂ sources, mass and energy balances for CO₂ methanation, and specific power requirements for different process steps, as discussed in Section 2. Specifically, for the PtG process outlined in Section 2.3.1, Fig. 5 illustrates the annual methane production potentials by sector under the base case scenario, along with the corresponding demands for H₂ and green electricity. In 2018, the estimated annual technical potential for methane production stands at 270 Mt (equivalent to 13.5 EJ), necessitating around 136 Mt of H₂ and 34 EJ of

⁴ Given the heterogeneity of certain industrial sectors, such as chemicals and food, the approach and estimated CO₂ emissions per establishment may only provide rough estimates and are subject to significant uncertainty. Therefore, any estimates for these sectors should be used with caution.

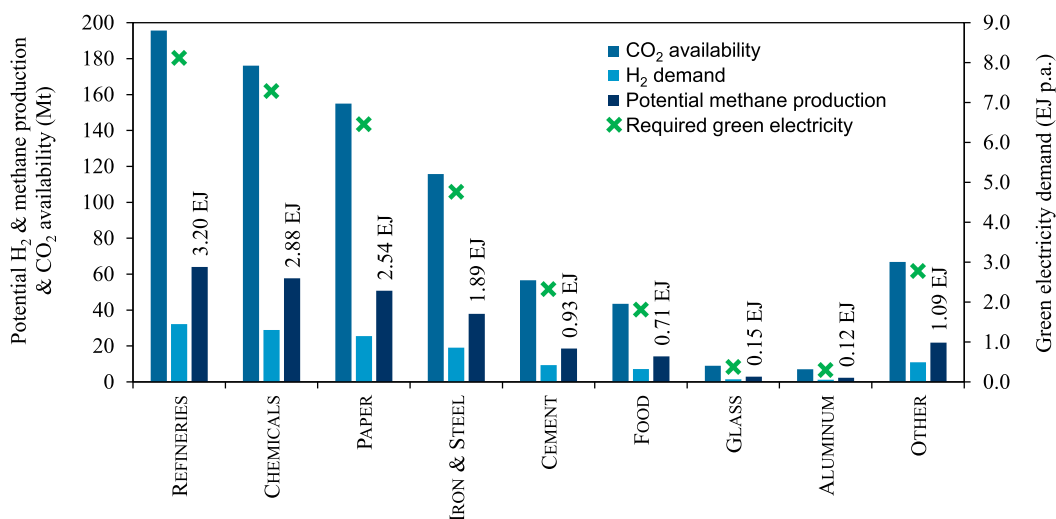


Fig. 5. Annual methane production potential in 2018 and the corresponding demand for H₂ and green electricity using the PtG process and CO₂ sources in U.S. industrial sectors (results for the reference technologies under base case scenario, refer to Sections 2.4 and 2.5; also see Appendix C of the supplementary material for combined scenario results).

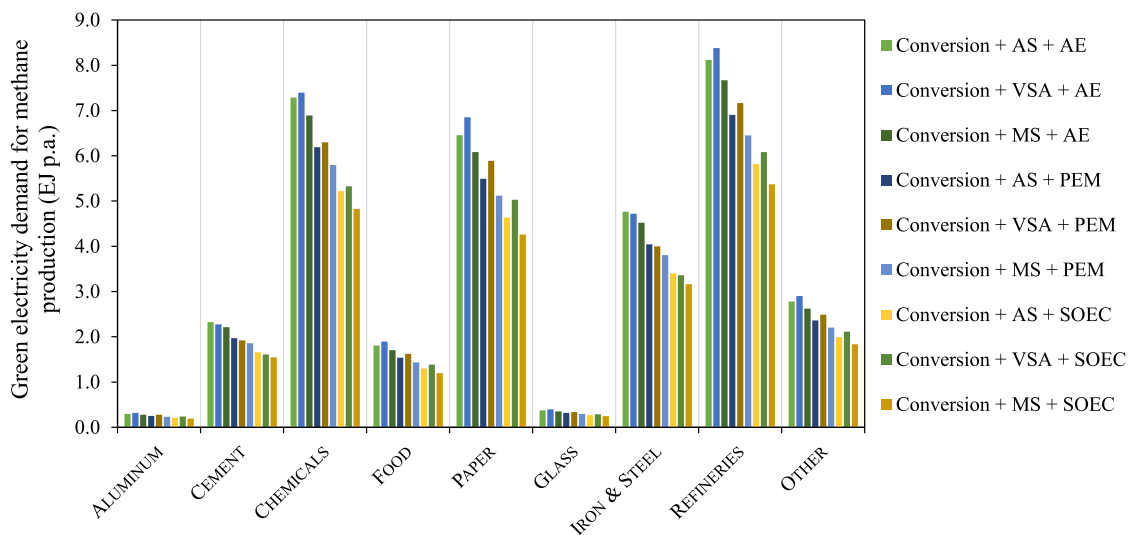


Fig. 6. Comparing green electricity demand for potential methane production in 2018 using various technology combinations (Note: AS = Amine Scrubbing, VSA = Vacuum Swing Adsorption, MS = Membrane Separation, AE = Alkaline Electrolyzer, PEM = Proton Exchange Membrane electrolyzer, SOEC = Solid Oxide Electrolyzer Cell; refer to Appendix D of the supplementary material for combined scenario results).

green electricity annually. Additionally, the figure presents that the highest annual production potential originates from refineries and the chemical industry (6.1 EJ p.a.), which require the largest quantities of H₂ (61 Mt p.a.) and green electricity (15 EJ p.a.).

Fig. 5 also illustrates the electricity demand of reference technologies, which includes CO₂ capture using amine-based absorption technique, transport of CO₂ through pipelines within a 100-mile radius, production of H₂ using alkaline electrolyzers, and catalytic conversion of CO₂ to methane, given their established technical maturity and commercial experience. However, alternative CO₂ capture and electrolyzer technologies outlined in Sections 2.4 and 2.5, respectively, are also investigated in detail. Fig. 6 provides a comparison of the overall annual potential green electricity demand for methane production in various sectors in 2018 using different technology combinations. Membrane-based CO₂ capture is observed to require the least electricity demand across all sectors, while the CO₂ concentration in flue gases determines which capture technology is the most electricity-intensive.

Vacuum swing adsorption (VSA) is found to be the most electricity-

intensive CO₂ capture technique for all industrial sectors except cement and iron and steel. Conversely, absorption technique is identified as the most electricity-intensive for these two sectors. This discrepancy is attributed to differences in the CO₂ concentration in the flue gases specific to each sector. VSA requires more electricity than amine-based absorption technique for flue gas CO₂ concentrations below 13 vol% (as illustrated in Fig. 4). Therefore, it can be concluded that the VSA (adsorption) technique is less energy-intensive than amine scrubbing (absorption) for CO₂ capture in industrial sectors such as cement (20 vol %) and iron and steel (15 vol%), which generate high-purity CO₂ streams.

The CO₂ concentration in flue gases specific to each sector does not affect the electricity demand for H₂ production and CO₂ conversion. However, it is crucial to compare the electricity demand of these process steps based on the chosen technology. To avoid overwhelming the reader with information in a single plot, Fig. 6 does not provide a breakdown of the overall electricity demand for methane manufacture. However, as an example of the cement industry, Fig. 7 illustrates the

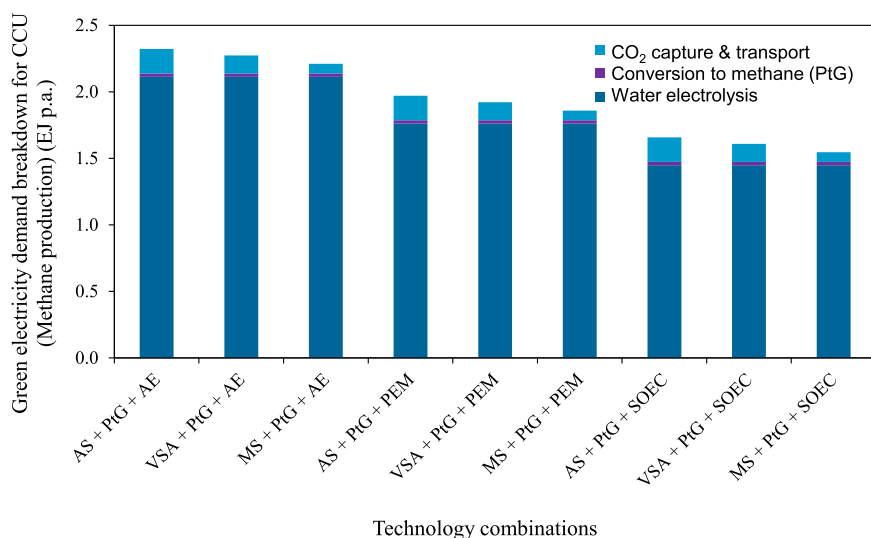


Fig. 7. Breakdown of green electricity demand for potential methane production in 2018 utilizing CO₂ from the U.S. cement sector (Note: AS = Amine Scrubbing, VSA = Vacuum Swing Adsorption, MS = Membrane Separation, AE = Alkaline Electrolyzer, PEM = Proton Exchange Membrane electrolyzer, SOEC = Solid Oxide Electrolyzer Cell).

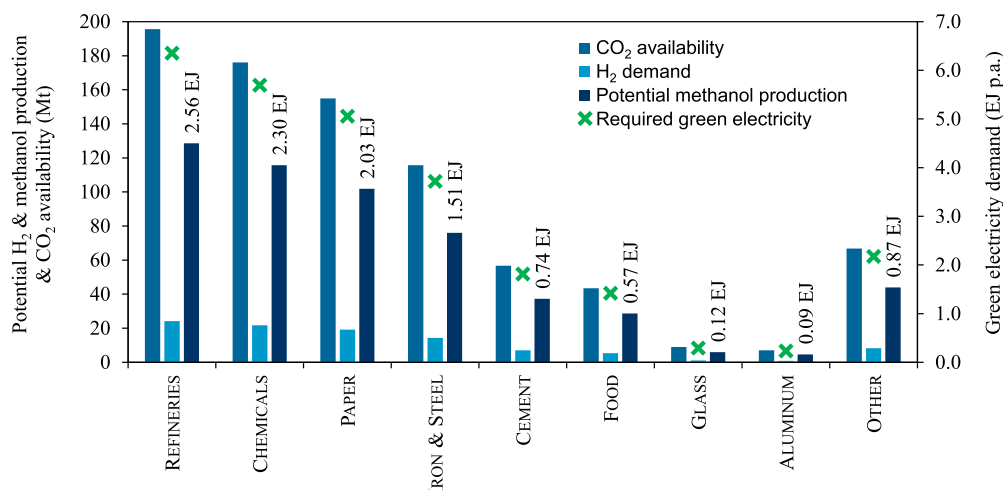


Fig. 8. Annual methanol production potential in 2018 and the corresponding demand for H₂ and green electricity using the PtL process and CO₂ sources in U.S. industrial sectors (results for the reference technologies under base case scenario, refer to Sections 2.4 and 2.5; also see Appendix C of the supplementary material for combined scenario results).

disaggregated electricity demand for each process step. Depending on the chosen technology, the contribution of electricity demand for H₂ production varies between 85 and 95 % of the overall electricity demand in PtG projects. This conclusion is consistent with the findings of Kiani et al. (2021), who also identified green H₂ production as having the largest share of energy consumption within a similar absorption-based CCU system. They estimated a slightly lower share than this study because they considered DAC for CO₂, which requires significantly more electricity due to lower CO₂ concentrations in the air. Alkaline electrolyzers are found to consume the most electricity, especially when combined with efficient membrane separation for CO₂ capture.

3.1.2. Potential methanol manufacture

Fig. 8 presents the annual methanol production potential in 2018 and the corresponding demand for H₂ and green electricity for the PtL methanol process described in Section 2.3.2. The figure demonstrates that the annual technical potential for methanol production that can be achieved today by capturing CO₂ from all industrial sectors is around 542 Mt (or 10.8 EJ), which requires approximately 102 Mt of H₂ and 27

EJ of green electricity annually. Fig. 8 shows the electricity demand of reference technologies, which includes the energy required for amine-based CO₂ capture, transportation through pipelines, alkaline electrolysis, and catalytic conversion of CO₂ to methanol.

This study also examined alternative CO₂ capture and electrolysis technologies discussed in Sections 2.4 and 2.5, respectively, in combination with the PtL methanol process. The comparison of potential green electricity demand for methanol production using different technology combinations in various sectors is consistent with the findings of methane generation and is not presented here. However, the quantitative results can be found in Appendix D of the supplementary material. Like CO₂ methanation, the process of methanol synthesis also requires significant amounts of energy, with the biggest contributor being the demand for electricity in H₂ production.

3.1.3. Potential fischer-tropsch fuels manufacture

Fig. 9 displays the annual potential for manufacturing Fischer-Tropsch (FT) fuels in 2018 and the corresponding demand for H₂ and green electricity using the PtL-FT process explained in Section 2.3.3. The

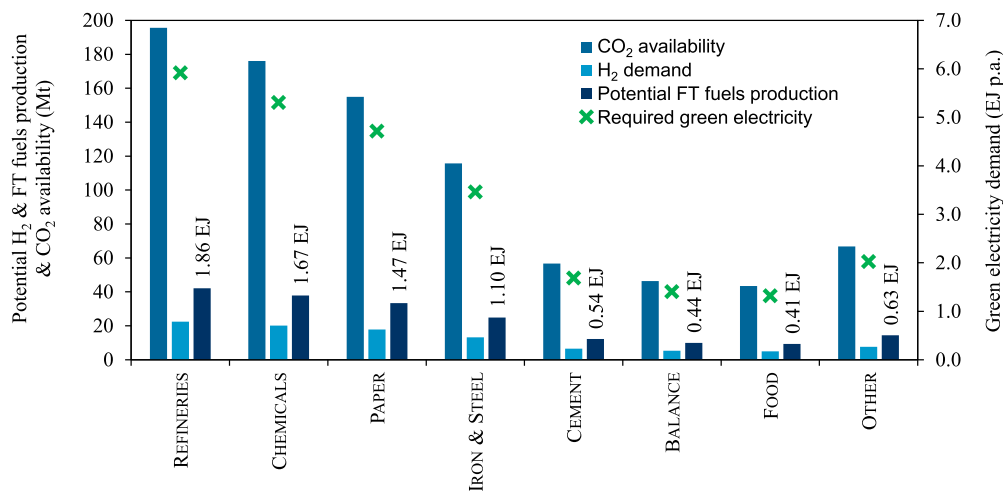


Fig. 9. Annual Fischer-Tropsch fuel production potential in 2018 and the corresponding demand for H₂ and green electricity using the PtL process and CO₂ sources in U.S. industrial sectors (results for the reference technologies under base case scenario, refer to Sections 2.4 and 2.5; also see Appendix C of the supplementary material for combined scenario results).

process simulation by König et al. (2015), which is adopted in this study, aims to create liquid hydrocarbons with a chain length between C₅ and C₂₀, with the majority of the product being within the C₅–C₁₀ range. According to the figure, the annual technical potential for FT fuel production by capturing CO₂ from U.S. manufacturing sectors is approximately 178 Mt (or 7.8 EJ), necessitating around 95 Mt of H₂ and 25 EJ of green electricity each year. The reference case electricity demand depicted in Fig. 9 includes the electricity required for amine-based CO₂ capture and transportation, alkaline electrolysis, and FT synthesis. The findings for potential green electricity demand for FT fuels’ production using different technology combinations in various sectors are also comparable to the results of methanation and methanol synthesis and are therefore not presented here. However, the quantitative outcomes are available in Appendix D of the supplementary material.

3.2. Scenario analysis

To compare the three promising CCU pathways, the sector-specific results for each sector presented in Section 3.1 (base case scenario) are aggregated at the U.S. industry level and depicted in Fig. 10 (left). The figure reveals that, for utilizing the same amount of CO₂ captured from U.S. industrial facilities, the methanation pathway has the highest H₂ demand (i.e., 0.18 kg of H₂ per kg of CO₂ utilized). The methanol and FT fuels production pathways exhibit nearly the same rate of H₂ demand (i.e., 0.14 kg and 0.13 kg of H₂ per kg CO₂ for the methanol and Fischer-

Tropsch processes, respectively). The U.S. produces more than 10 Mt of H₂ annually, with only 1 % of it produced from electrolysis (U.S. DOE, 2020). However, the current H₂ production is roughly 10 times lower than what would be needed for CO₂ conversion alone. A much bigger challenge would be to produce green H₂ on a significantly larger scale compared to today. Nevertheless, the application of CCU in combination with several other decarbonization measures could substantially reduce the potential H₂ demand while still ensuring a significant supply of sustainable fuels and feedstock. Hence, there is a need for an inclusive approach and a design framework to roadmap the decarbonization of the industrial sector.

Taking a step towards applying the aforementioned inclusive approach, the combined scenario, as explained in Section 2.2 and depicted in Fig. 3, involves integrating waste heat into industrial processes from the very outset. The primary goal is to reduce both fuel demand and CO₂ emissions in the first place. This integration involves utilizing all potential waste heat through pinch analyses and heat pump integration, leaving no waste heat to be recovered from exhausts and effluents. Without this integration, some of the waste heat would have been used for absorption-based CO₂ capture. The overarching strategy is to give priority to waste heat integration, which not only reduces CO₂ emissions but also prevents locking-in fossil-based technologies and their associated emissions. In Fig. 10 (right), the results for the combined scenario are presented, indicating the resulting CO₂ emissions after fully utilizing the potential for electrified steam supply and process heat

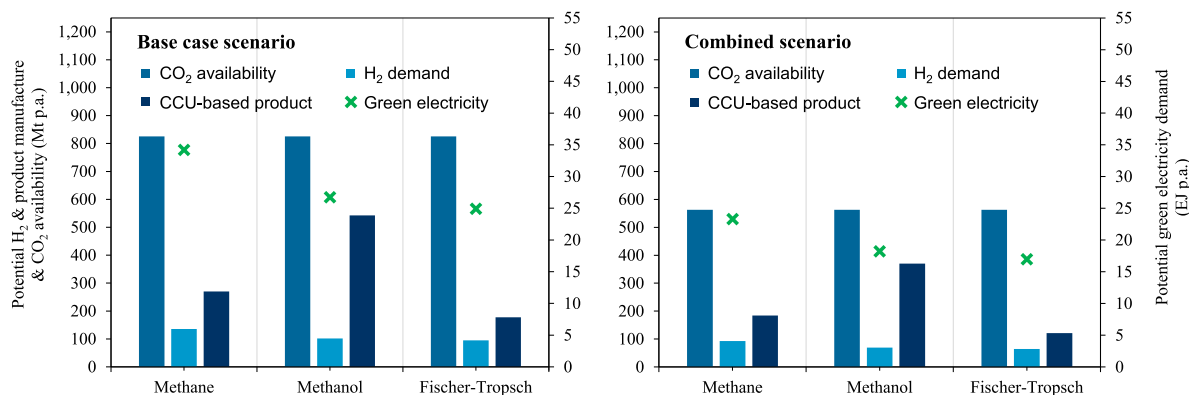


Fig. 10. Overall annual potential for CCU-based product manufacture in 2018, the corresponding demand for H₂ and green electricity, and U.S. industrial CO₂ sources under base case and combined scenarios.

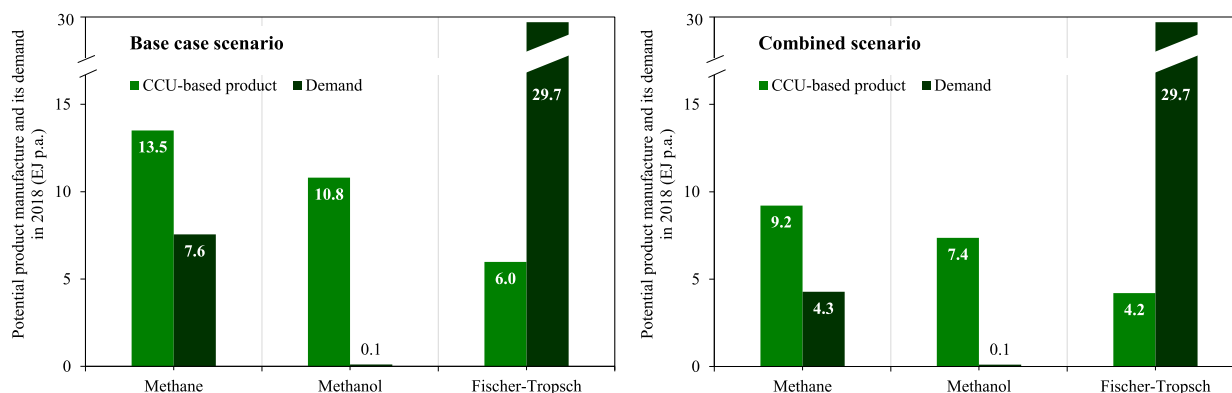


Fig. 11. Overall annual potential for CCU-based product manufacture under base case and combined scenarios, and its corresponding annual demand in the U.S. in 2018 (Note: The demand estimates for methane differ in the two scenarios because in the combined scenario, electrified steam generation and heat integration have the potential to reduce over 40 % of industrial natural gas demand. Additionally, the potentials for FT fuel production exclude refinery-based CO₂ utilization to avoid double counting.⁵¹).

integration. More comprehensive details on this topic can be found in the author's forthcoming publication (Zuberi et al., 2024), along with preliminary results presented at an international conference (Zuberi et al., 2023). The comparison shown in Fig. 10 clearly demonstrates that the annual potential for CCU-based products in the combined scenario is roughly one-third of what it is in the base case scenario in 2018.

Fig. 11 illustrates the annual U.S. demand for the studied chemical products. For simplification, methane is categorized with natural gas, which is primarily composed of methane. The annual demand for natural gas in the figure represents its usage for industrial applications and as a feedstock in the U.S. industry. Additionally, the methanol production in the U.S., as shown in the figure, accounted for only 6 % of the global demand in 2018 (P.C. Statista May 6, 2023; P.C. Statista May 6, 2023). Furthermore, the figure displays the annual demand for liquid hydrocarbons, including naphtha (utilized as a chemical feedstock), gasoline, diesel, and kerosene, in both the U.S. industry and transportation sectors. These are aggregated and presented in Fig. 11 to facilitate comparison with the potential production of FT fuel products.

Based on the estimates provided in MECS (US DOE/Energetics, 2022), the annual natural gas demand for industrial processes in the U.S. (excluding the conventional methanol industry) was 7.6 EJ in 2018. If all the available industrial CO₂ is utilized for methane production, it has the potential to replace all of the fossil-based methane demand that is considered in both scenarios. In the combined scenario, the potential production is found to be more than twice the demand for industrial applications, mainly due to significantly reduced demand for natural gas resulting from decarbonization through electrified steam generation and heat integration in the first place. However, the surplus could be utilized to reduce the remaining process heat demand, part of which is currently supplied by combusting other fossil fuels like coal and fuel oil. Alternatively, the surplus could be used to partially replace the natural gas demand in other economic sectors. However, the authors believe that electrified technologies for most heating purposes in other sectors such as residential or commercial are already market-matured. For instance, electric boilers or conventional heat pumps can replace conventional gas boilers for hot water supply in residential buildings. Hence, the impact of using synthetic methane in other economic sectors has not been studied.

The amount of methanol produced in the U.S. is approximately 5.2 Mt (equivalent to 0.1 EJ), which is much lower in energy terms than natural gas and petroleum feedstocks and fuels. Therefore, CCU-based methanol could potentially substitute for several times the U.S. production under both scenarios, especially in the base case scenario where the potential for methanol production from CO₂ is nearly 30 % higher than in the combined scenario. If all available industrial CO₂ were to be

converted to methanol, then the U.S. alone could theoretically fulfill the current global methanol demand of 92 Mt (or 1.8 EJ). Furthermore, global methanol production is projected to increase to 500 Mt (or 10 EJ) in 2050, and this increase must also be accompanied by a shift to low-carbon methanol (Saygin and Gielen, 2021; IRENA and Methanol Institute, 2021). Thus, CCU for methanol can be considered one solution, with the U.S. having the potential to become a leading producer and/or exporter of low-carbon methanol in the future.

Regarding the third utilization pathway, the substitution of naphtha feedstock and transportation fuels with FT products in the U.S. is not sufficient for completely decarbonizing the studied end uses in both scenarios. In 2018, the annual demand for naphtha feedstock, gasoline, diesel, and kerosene-based aviation fuel was approximately 30 EJ (US EPA, 2022; U.S. EIA, 2021). Therefore, even if all the available net CO₂ from the industry were converted to FT products, only a fraction, i.e., 20 % and 14 % of the targeted products' demand, could be replaced with CCU-based products under base case and combined scenarios, respectively. This implies that FT products alone cannot serve as a substitute for the studied products, and other possible substitutes will be needed to meet the targeted GHG emissions reduction. While CO₂ emissions from on-road gasoline vehicles can be mitigated through electric vehicles, FT fuels may be more effective in reducing CO₂ emissions from heavy-duty diesel vehicles and kerosene-based aviation fuels, which currently lack suitable alternative technologies for electrification. Another viable option involves exporting these cleaner fuels to nations burdened by high oil imports and downstream oil refining costs to meet their energy and feedstock demands. This export strategy has the potential to substantially reduce CO₂ emissions from oil refineries in those countries. Consequently, the U.S. refineries could earn carbon credits, enabling them to operate sustainably without necessarily cutting down on production.

Table 5
Process-specific conversion and plant efficiencies.

Performance metric	This study	(Brynolf et al., 2018)
<i>CO₂ methanation</i>		
Conversion efficiency	83 %	–
Plant efficiency	81 %	70–83 %
<i>Methanol synthesis</i>		
Conversion efficiency	88 %	–
Plant efficiency	85 %	69–89 %
<i>Fischer-Tropsch synthesis</i>		
Conversion efficiency	69 %	–
Plant efficiency	67 %	63–83 %

3.3. Performance levels

In addition to assessing the potential for producing CCU-based products, this study also evaluates the performance of the three CO₂ conversion processes using the methods outlined in Section 2.5. Table 5 presents the chemical conversion and plant efficiencies for each of the studied CO₂ utilization pathways, as well as a range of conversion and plant efficiencies reported by the review article published by Brynolf et al. (2018) for comparison. The results obtained in this study are in good agreement with the literature.

Methanol synthesis exhibits relatively high energy performance compared to the other two processes, with the amount of H₂ and electricity required to produce a unit of methanol being 62 % and 46 % lower than that needed for a unit of methane, and 65 % and 67 % lower than that required for a unit of FT fuels, respectively. The efficiencies of the FT process are found to be low due to its high demand for H₂ (0.53 kg H₂ per kg of FT products) and the significant energy demand for a relatively large number of downstream product separation and upgrading unit operations. These results will have cost implications, which are discussed in the next section.

3.4. Economic assessment

Each industrial sector incurs unique CO₂ capture costs that depend on the CO₂ concentration and flow rate (which determine the size of the plant). Fig. 12 displays the technology-specific CO₂ capture costs for each sector in the base case scenario. The CO₂ capture costs are calculated using Eq. (9), with the only difference being the consideration of specific cost components related to the CO₂ capture plant only (i.e. $I_{CC,s}$; also see Eqs. (11)–(13)). The cement and iron and steel sectors have the lowest estimated CO₂ capture costs, ranging from \$67–107/tCO₂ and \$83–121/tCO₂, respectively, due to their flue gases' higher CO₂ concentration and larger plant sizes. Similarly, the refinery and chemical sectors have relatively low CO₂ capture costs. However, in 'other' industrial sectors with smaller volume and low CO₂ purity streams from their processes, costs are higher, ranging from \$270/tCO₂ to \$320/tCO₂. Despite higher investment and operational costs, CO₂ capture through membrane technology is a lower-cost option than competing absorption and adsorption techniques due to significantly lower energy costs. These energy costs form the majority of the lifetime costs of CO₂ capture. Nonetheless, the high upfront investment required for membrane technologies poses a significant barrier.

Moreover, Fig. 13 presents generalized CO₂ capture costs for industrial facilities, estimated and depicted as a function of the CO₂ concentration in industrial flue gases. The figure fits a curve to each technology- and sector-specific data point from Fig. 12, demonstrating that the relationship between specific costs and CO₂ concentration follows a similar trend to electricity demand in Fig. 4. The CO₂ capture costs estimated in this study align somewhat with the overall cost ranges for low CO₂ purity streams as reported by Brynolf et al. (2018); Panzone et al. (2020); and Zang et al. (2021) as illustrated in Table 6. The upper bound for the capture costs appears higher primarily due to the inclusion of CO₂ capture through early-stage technologies such as membrane separation and adsorption techniques. When considering only the absorption technique for CO₂ capture, the maximum costs are found to be up to \$270/tCO₂, which is relatively close to the upper bound reported

by Brynolf et al. (2018). These higher cost bounds also encompass price differences and incorporate industries such as textiles and small foundries (merged into the 'other' category as previously stated), which are typically not considered for CCUS due to their poor techno-economics, as evident. By excluding these manufacturing sectors, the upper bounds align more closely with the reported cost limits in the literature, ensuring consistency.

Transportation distance is a critical factor that affects CO₂ transport costs. In the CO₂ pipeline networks, a single spine route is assumed to handle 20 MtCO₂ and 10 MtCO₂ per annum in the base case and combined scenarios, respectively, while smaller feeder lines will connect to individual plants. The spine pipeline and CO₂ compression involve significant investments and operational costs, whereas feeder pipeline costs represent a small portion of the total transportation costs. The distance for CO₂ transport to CO₂ conversion facilities is fixed at no more than 100 miles. Consequently, the average CO₂ compression and transportation costs are estimated to be around \$12–13/tCO₂. To validate these results, various studies (Zang et al., 2021; Brynolf et al., 2018; Smith, 2021; McCollum and Ogden, 2006; Zero Emissions Platform, 2011; IEA, 2020) that reported the costs of CO₂ compression and/or compressed CO₂ transport were consulted and found to agree with the findings of this study.

The methods outlined in Section 2.7 are used to calculate the overall costs of CO₂ capture and utilization across all pathways and industrial sectors. These calculations demonstrate the costs associated with utilizing a unit of CO₂ captured from various industrial sources using different technology combinations. Fig. 14 summarizes the outcomes of the CCU cost calculations with the three left-side figures depicting the results (per-unit-CO₂ utilized basis) in the base case scenario. Depending on the technology combinations, the investments required to exploit the full potential in the base case scenario are estimated to vary between \$165 billion and \$310 billion, with 65–85 % associated with CO₂ capture and transport, 20–30 % with H₂ production, and less than 5 % with conversion equipment. On the other hand, energy costs are predominantly driven by operations related to H₂ production (over 80 %), followed by CO₂ capture and transport. Energy costs also constitute the majority of the annualized cash inflows (see Appendix E of the supplementary material for a breakdown of costs), with the costs of producing H₂ being the dominant factor, as explained in the discussion surrounding Fig. 7. Therefore, the utilization pathway requiring more electricity, primarily for H₂ production, to convert CO₂ to a chemical product has the highest cost. More specifically, for the same amount of CO₂ captured from U.S. industrial facilities, the pathway involving methanation has the highest H₂ demand, requiring 0.18 kg of H₂ per kg of CO₂ utilized. The methanol and FT fuels production pathways require similar amounts of H₂, with 0.14 kg and 0.13 kg of H₂ per kg CO₂ for the methanol and Fischer-Tropsch processes, respectively. Thus, the conversion of a unit of CO₂ to methane is found to be the most expensive, followed by methanol and FT synthesis.

It must also be noted that several studies have developed experience or learning curves to predict the decline in investment costs of various technologies. Research programs worldwide are dedicated to minimizing the costs associated with electrolytic H₂ production. The widespread deployment of electrolyzer technologies depends on the feasibility of producing H₂ in a cost-effective manner. According to the (IEA, 2018), there is a projected 65 % reduction in the capital cost of electrolyzers by 2050. However, as explained above, it is the energy costs that dominate the overall cost estimates in Fig. 14. Hence, despite assuming a 65 % lower capital cost for electrolyzers, the overall specific costs in Fig. 14 decrease by a mere 1 %. However, a price sensitivity analysis has also been conducted for 2050 to highlight the impact of energy cost reduction on the CCU costs. This work has assumed a decrease in electricity prices from \$0.08/kWh to \$0.06/kWh based on the U.S. Annual Energy Outlook's low zero-carbon technology scenario (U.S. EIA, 2022). Under this price reduction scenario, the CCU costs for different pathways, technology combinations, and sectors are estimated

⁵ CO₂ emissions from the refinery sector may persist at current levels if the demand for fuels is met by producing them in refineries through conventional processes. However, CCU for FT fuels will reduce emissions originating from refineries by decreasing the production demand for conventional refinery fuels. To prevent double counting, the potential for FT fuels in Fig. 11 excludes that from U.S. refineries. Conversely, exporting cleaner fuels could potentially earn U.S. refineries carbon credits for sustainable operation without reducing production (see later discussion in this section).

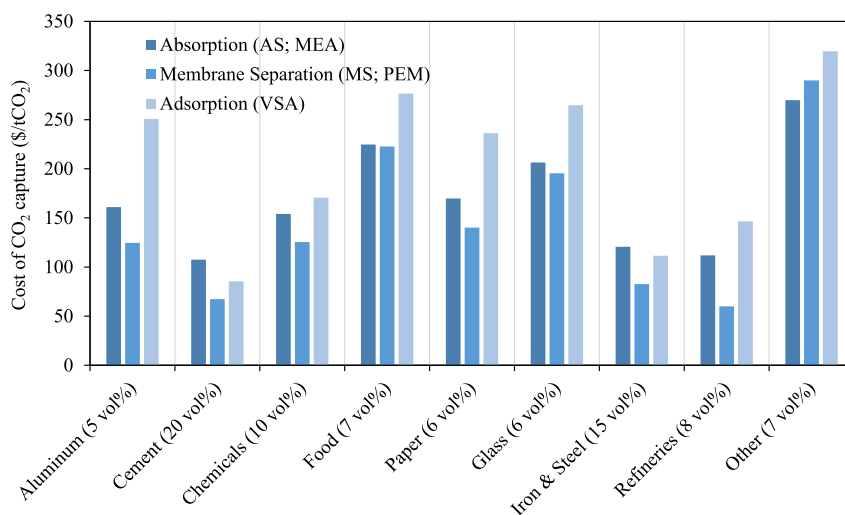


Fig. 12. Technology-specific CO₂ capture costs for different industrial sectors in the base case scenario (Unit: \$/tCO₂ captured) (Note: Sectoral weighted average flue gas CO₂ concentrations are given in parentheses).

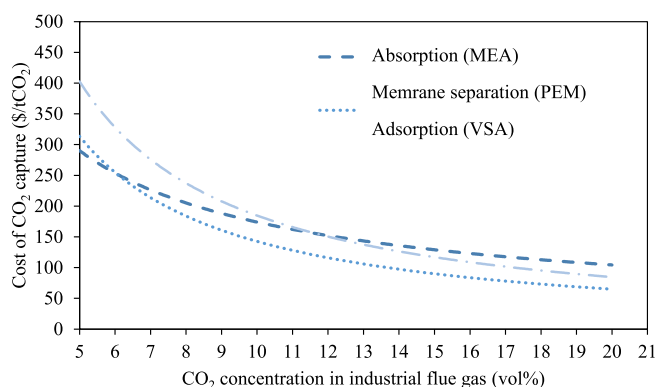


Fig. 13. Generalized CO₂ capture costs as a function of CO₂ concentration in flue gases.

Table 6

Range of CO₂ capture costs for different low-concentration CO₂ sources.

Study	CO ₂ capture costs (\$/CO ₂ captured)	
	Minimum	Maximum
This study	60	320
(Brynnolf et al., 2018) ¹	22	189
(Zang et al., 2021)	57	195
(Panzone et al., 2020) ²	60	240

¹ 1 EUR = 1.11 USD.

² Roughly estimated based on the given information.

to decrease by approximately 20 % in 2050. Appendix F of the supplementary material presents the results of the price sensitivity analysis in detail.

Moreover, analyzing the costs from a different perspective, by replacing the denominator in Eq. (9) with the amount of product manufactured, allows for expressing the costs on a per-unit-production basis, refer to Fig. 14 (right). This perspective focuses more on the production of a specific chemical through a cleaner pathway as compared to the fossil-based conventional pathway, rather than on CO₂ utilization. When considering the manufacturing of a chemical product, the FT synthesis exhibits the highest H₂ demand, requiring 0.53 kg of H₂ per kg of fuel product, followed by the methanation, which requires 0.50 kg of H₂ per kg of methane, and the methanol synthesis, which requires 0.19 kg of H₂

per kg of methanol. Therefore, from the product manufacturing perspective, producing a unit of FT fuels is the most expensive due to its low conversion and plant efficiencies (as discussed in Section 3.3). Moreover, while methane requires a higher amount of H₂ per kg compared to methanol, its relatively higher calorific value balances out the costs per unit of energy for both chemicals, as depicted in Fig. 14. Nevertheless, methanol synthesis emerges as the most favorable option from both utilization and production perspectives. However, its manufacturing potential may be constrained by future demand. This conclusion underscores the necessity for future research to identify potential end-uses that could transition to renewable methanol, such as methanol-to-olefins (MTO) and methanol-to-aromatics (MTA), rather than relying on energy-intensive steam cracking for the production of these chemicals.

In Fig. 14, it is evident that CCU costs differ based on the industrial sector. This is due to two factors: first, the difference in electricity requirements and associated costs for CO₂ capture, which are influenced by CO₂ concentration in sector-specific flue gases, and second, the size of the CO₂ capture plant, which ultimately impacts investment costs. As demonstrated in Fig. 12, the cost of CO₂ capture is comparatively economical in the cement, iron and steel, chemical, and refinery sectors, while sectors such as machinery, plastics, textiles, and electronics (combined into the 'other' category) have higher costs due to their lower magnitude of CO₂ streams and lower CO₂ concentrations in plant flue gases. As previously mentioned, H₂ constitutes the majority of CCU energy demand and costs. Its production at a centralized facility, away from the industrial CO₂ sources, makes it independent of site-specific characteristics. As a result, the overall CCU costs depicted in Fig. 14 exhibit only moderate variations. Therefore, it can be concluded that small and medium enterprises may require additional investments for CO₂ capture, which could be supported through government subsidies or tax breaks. However, from the CCU perspective, the focus should be on reducing the costs of H₂ production to make CCU projects economically viable.

Additionally, Fig. 14 illustrates the range of CCU costs for each sector, which are influenced by different technology combinations. The lower cost limits correspond to CO₂ utilization combined with membrane separation for CO₂ capture and SOEC for H₂ production. These technologies have higher efficiency and lower electricity demands compared to their alternatives, leading to lower costs despite their relatively high capital costs. The upper cost limits represent CO₂ utilization in combination with the VSA technique for CO₂ capture (except for the cement and iron and steel sectors, where amine-based absorption

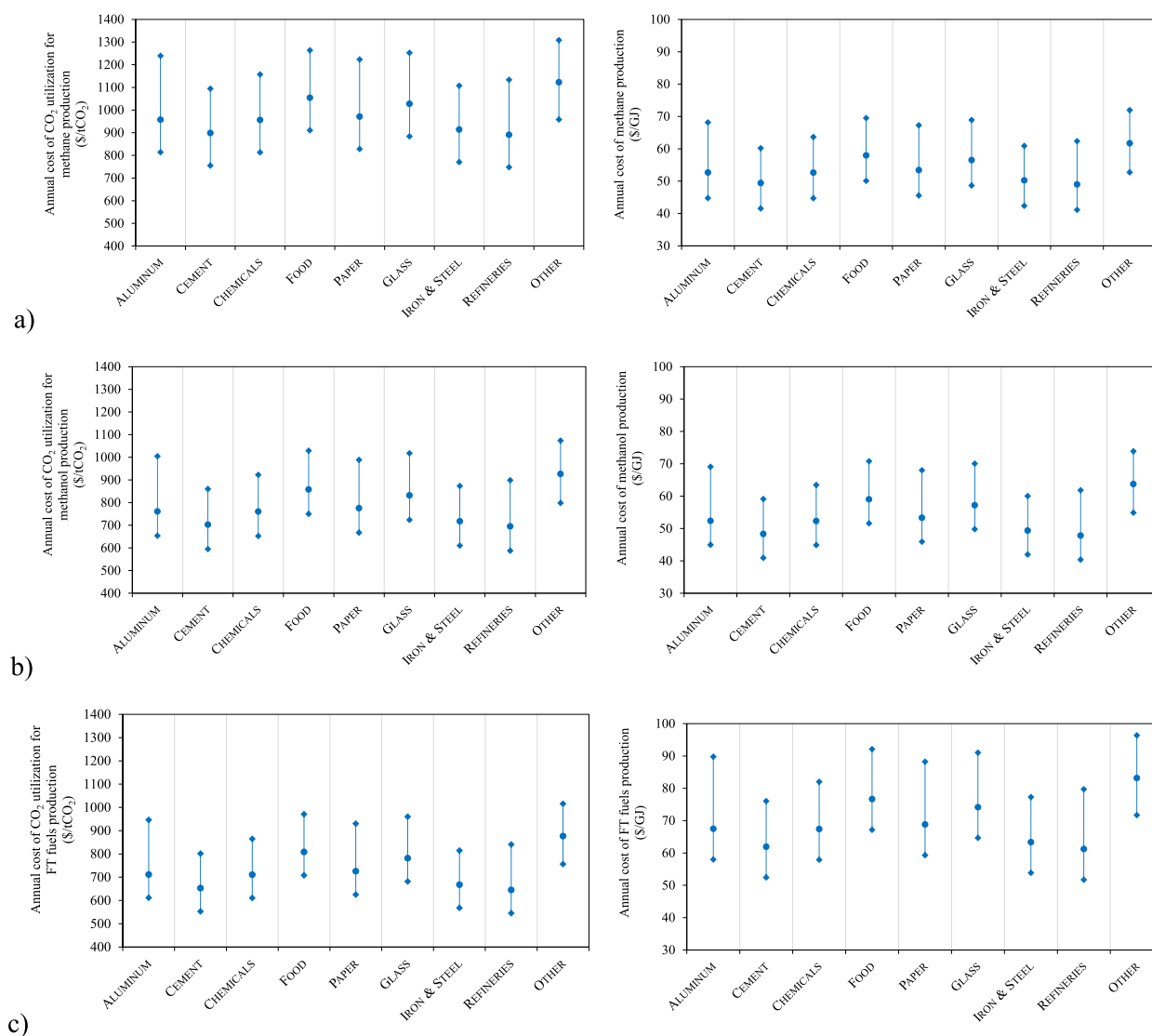


Fig. 14. CO₂ utilization costs (left) and production costs (right) of a) methane, b) methanol, and c) Fischer-Tropsch fuels manufacture using CO₂ captured from different industrial sources (results for the base case scenario; refer to Appendix F of the supplementary material for combined scenario results).

Table 7
Summary of costs of CO₂ capture and utilization from different perspectives.

CCU pathway	This study		(Brynnolf et al., 2018)	(Zang et al., 2021)
	\$/tCO ₂ utilized	\$/GJ product	\$/GJ product	\$/GJ product
CO ₂ methanation	748–1462	41–80	3–178	–
Methanol synthesis	587–1227	40–84	17–111	~17–20
Fischer-Tropsch synthesis	545–1169	52–111	13–972	~51–66

for CO₂ capture was found to be the most expensive) and alkaline electrolysis for H₂ production. Although SOEC is cost-competitive due to significant efficiency gains, its scalability presents a significant implementation barrier. Conversely, alkaline electrolyzers and amine-based absorption techniques are more established technologies, but their costs are high, ultimately limiting their widespread implementation. The mid-range costs, as indicated in Fig. 14, represent the combination of CO₂ conversion, membrane separation, and PEM electrolysis. Both membrane separation and PEM electrolysis technologies offer a good

trade-off between high efficiency and low costs, making them suitable options that require more detailed feasibility assessments.

Table 7 provides a summary of the CCU cost results, which are compared with other literature sources and found to be consistent with other studies. In 2018, the prices of gasoline, diesel, and aviation fuels in the U.S. were approximately \$25/GJ, \$24/GJ, and \$17/GJ, respectively (U.S. EIA, 2021). This study finds that the minimum production costs for FT products are 2–3 times higher than the prices of relevant fuels in 2018. Additionally, the production costs of synthetic methane and methanol are estimated to be 10 times and 2 times higher than the average industrial natural gas and fossil-based methanol price of \$4/GJ (U.S. EIA, 2021) and \$20/GJ (Intratec May 8, 2023) in 2018, respectively. The comparison reveals that all CCU-based chemical products are presently not economically feasible and require R&D support to achieve cost reduction. Specifically, synthetic methane derived from CO₂ is deemed excessively costly from the production perspective, rendering it an unfeasible option at this time.

3.5. Implications, limitations and future work

The concept of carbon capture refers to the process of capturing, storing, and/or utilizing CO₂ emissions from various sources, including

fossil fuels. However, the implementation of CO₂ capture technologies may lead to a potential lock-in of current fossil-based technologies, meaning that they could continue to dominate the energy sector despite their negative impact on the environment. CCUS should be considered as a last resort, to be employed only when other decarbonization options, for example, direct electrification of process heat, have been exhausted in the efforts to limit CO₂ emissions. It is important to acknowledge this potential drawback and ensure that efforts to reduce CO₂ emissions also include a transition towards cleaner and more sustainable energy sources. To address this issue, this study has taken an inclusive approach by considering various aspects of industrial decarbonization simultaneously and developed a combined decarbonization scenario. However, the combined scenario currently only considers electrified steam generation and process heat integration. It is therefore crucial to integrate CCUS strategies within a broader framework that prioritizes the application of technologies specific to other decarbonization measures over CCUS. For example, electric resistance and electromagnetic heating are among the most promising technologies for electrifying the process heat supply on a large scale, and their working principles are cross-cutting in nature. However, the potential applications of these technologies in industrial processes depend on several factors, and the industry has only scratched the surface of their true potential. Additional assessments of emerging technologies for reducing CO₂ emissions are hence needed for integration into the broader industrial decarbonization strategies.

CCU for chemicals production, such as methane, methanol, and FT products, through catalytic conversion, has been explored to reduce CO₂ emissions and promote sustainability in manufacturing. By capturing CO₂ emissions from industrial processes and utilizing them as fuel for process heat (e.g., synthetic methane), CO₂ can be effectively recycled, which would otherwise be released into the atmosphere as GHG emissions. This end-use application may help to close the carbon loop and reduce the overall carbon footprint. If CO₂-based fuels are used as transportation fuel, due to the decentralized nature of the transportation sector, it may not be possible to capture CO₂, and it will eventually end up in the atmosphere with a delay. However, these synthetic fuels will still diminish CO₂ emissions currently originating from the conventional production of these fuels. For example, all of the current U.S. conventional methanol manufacturing and its corresponding emissions can be displaced, and similarly, part of the refinery products can be substituted by FT fuels. Furthermore, many of the studied chemical feedstocks used in plastics manufacturing end up being landfilled at the end of their life in the U.S., with more than 90 % of them being discarded in this manner (Scown, 2022). Hence, captured CO₂ that is locked in plastics could remain underground for a long period if landfilled. However, all of the aforementioned end-use applications of CCU-based products require detailed carbon accounting, and future research should perform detailed life-cycle assessments for all the utilization pathways, either separately or in combination, after prioritizing the specific chemical needs.

Methane production from CO₂ is poised to undergo gradual improvements in both efficiency and scalability, driven by advancements in catalysts and reactor designs. These enhancements hold the promise of making the process economically feasible. Similarly, breakthroughs in catalyst development and process integration and optimization have the potential to significantly boost the efficiency and economic viability of methanol production from CO₂. Advancements in FT synthesis technology and reactor design offer opportunities to improve the efficiency and selectivity of the process, thereby making CO₂ utilization economically viable. To further bolster the sustainability of these pathways, integration with renewable energy sources for H₂ production, as emphasized in this study, is essential. Moreover, the successful development and application of these CCU pathways hinge on robust regulations governing CO₂ emissions. Establishing a regulatory framework and policies are crucial for initiating and sustaining widespread CCU deployment where necessary. Potential policy mechanisms could include implementing a sustainable price on CO₂ emissions through a CO₂ levy or establishing an effective emissions trading system, both of

which would incentivize CCU deployment as needed. However, the success of CCU projects also relies on the availability of CO₂ transport and storage infrastructure. Meeting the CCU needs of various industries necessitates a coordinated effort to expand transport lines and enhance the existing CO₂ pipeline network. By addressing these key components and fostering collaboration between industry, government, and regulatory bodies, the potential for effective and scalable CO₂ utilization can be realized.

It must also be noted that the conversion of CO₂ to studied chemical products would require a substantial amount of renewable H₂ as feedstock. While green H₂ has been studied primarily in the context of CCU, it also presents a solution as a fuel option, providing zero-carbon process heat through direct combustion or conversion to heat and electricity via fuel cell CHP. Numerous studies have explored its potential and the barriers it faces in industrial process heating applications. Primarily envisioned for medium-temperature processes like steam generation, drying, and distillation, green H₂ holds promise. However, its adoption encounters formidable challenges, notably competition from low-cost natural gas. Some argue that green H₂, due to its high energy costs and concerns about water stress, is too valuable to be simply used as fuel. Economic constraints and policy uncertainties, particularly regarding carbon pricing, significantly influence the feasibility of incorporating clean H₂ into energy systems. Despite these hurdles, future efforts should focus on assessing and comparing the techno-economic feasibility and environmental benefits of alternative applications for green H₂. Exploring the potential utilization of green H₂ as a feedstock for CCU or direct combustion for decarbonization can facilitate a deeper understanding of its role in transitioning towards a sustainable energy future.

The techno-economic assessment shows that electricity costs make up the majority of CCU application expenses, primarily driven by operations related to H₂ production, followed by CO₂ capture and transport. To implement all electrified CCU pathways studied on a large scale, significantly more electricity than the current grids can handle would be required. The development and expansion of renewable-based grids would be necessary to ensure that the burden of CO₂ emissions is not shifted to the power sector. However, these analyses are beyond the scope of this study and must be conducted in the future to evaluate the aforementioned aspects. This includes spatiotemporal analyses to assess the magnitude and access to local renewable resources for power generation. Additionally, detailed policy analysis is necessary to incentivize and facilitate CCU project implementation. Future work must devise policy interventions and evaluate them to show their potential impacts, which different stakeholders may consider when developing their climate change mitigation strategies.

Finally, some of the previous studies in literature have conducted similar analyses as in this study, however, they have been limited in their scope regarding the pathways studied, technology resolution, system boundaries, and sectors. In contrast, this study is one of the most comprehensive on the topic, examining 27 different technology combinations across 16 U.S. industrial sectors. This study covers the entire process from CO₂ capture to the manufacturing of end products and considers economies of scale and CO₂ concentration, which is a novel contribution. However, despite this comprehensive assessment, there are some data uncertainties. Transparency is lacking in many relevant studies regarding the methodology and underlying data used in their respective cost-benefit analyses for CCU measures. While the capital costs used are collected from recent studies, this study did not correct for inflation due to a lack of detailed information, and these costs are assumed to be valid today. There is also ambiguity surrounding the O&M costs and lifetimes of the studied technologies acquired from the literature. Therefore, the cost estimates in this study are uncertain and should be used with caution, subject to revision in the future based on empirical evidence.

4. Conclusions

The article presents a comprehensive analysis and quantitative assessment of various technologies for CO₂ capture and utilization. The study investigates the potential for CO₂ capture from different U.S. industrial sources and its conversion into synthetic methane (Power-to-Gas) and methanol and Fischer-Tropsch (FT) fuels (Power-to-Liquid). To estimate the energy demand for CO₂ capture and conversion, this study considers the CO₂ concentration in flue gases from each industrial sector and various technology combinations. Additionally, this study estimates the demand for green H₂ for CO₂ conversion and analyzes different electrolyzer technologies and their corresponding energy demands for H₂ production.

Although, in the base case scenario, this study considers all CO₂ emissions from the U.S. industrial sectors for capture and conversion to the aforementioned products, a combination of multiple decarbonization measures will be necessary to achieve net-zero CO₂ emissions in the industrial sector. Thus, a combined scenario has been developed to apply an inclusive approach, considering various aspects of industrial decarbonization simultaneously. This includes the electrification of steam generation systems and energy efficiency achieved through excess heat recovery and integration. The potential for CCU-based products in the combined scenario is estimated to be roughly half of that in the base case scenario. The results show that if all industrial CO₂ is utilized for methane production, it has the potential to replace all natural gas demand considered in both scenarios, with a surplus available to reduce demand for natural gas in other sectors.

However, using CCU-based FT products to replace naphtha feedstock and transportation fuels alone is not sufficient to achieve complete decarbonization in the studied end uses, with only a fraction of the targeted products demand replaceable with CCU-based products under the base case and combined scenarios. The study suggests that while electric vehicles can reduce CO₂ emissions from on-road gasoline vehicles, FT fuels may be more effective in reducing emissions from heavy-duty diesel vehicles and aviation fuels. Alternatively, exporting the clean FT fuels to countries burdened by high oil imports to meet their energy and feedstock needs could enable the U.S. to earn carbon credits, facilitating sustainable refinery operations without necessarily cutting down on production. Finally, CCU-based methanol could potentially substitute for several times the current U.S. methanol production and fulfill the current global demand for methanol, making the U.S. a leading exporter of low-carbon methanol. Methanol synthesis is also estimated to have relatively high energy performance compared to the other two processes.

This study also conducts an economic analysis to estimate the costs of CCU, which vary across different industrial sectors and are contingent upon the technologies employed. Notably, the cement and iron and steel sectors are projected to exhibit the lowest CO₂ capture costs due to their elevated CO₂ concentration in flue gases and larger plant sizes. A key finding of this analysis is that a significant portion of annualized cash inflows is driven by energy costs, particularly in the production of H₂. Pathways that necessitate more electricity, particularly for H₂ production to convert CO₂ into chemical products, incur higher costs. Consequently, the overall CCU costs are observed to be highest for CO₂ methanation, as it requires the most electricity, primarily for H₂ production, to convert a unit of CO₂ to a chemical product. Despite lower capital costs, the overall specific costs register only a marginal decrease. However, a price sensitivity analysis for 2050 suggests a notable impact on CCU costs with a reduction in electricity prices. Methanol synthesis emerges as an attractive option from both CO₂ utilization and production perspectives, though its manufacturing potential may be constrained by future demand considerations.

The ranges of CCU costs for each industrial sector are further determined and are influenced by different technology combinations. Lower cost limits are associated with CO₂ utilization combined with membrane separation for CO₂ capture and solid oxide electrolyzer cell

(SOEC) for H₂ production, while the upper cost limits are largely represented by CO₂ utilization with the vacuum swing adsorption (VSA) technique for CO₂ capture and alkaline electrolysis for H₂ production. While SOEC is cost-competitive, its scalability is a significant barrier to implementation. Alkaline electrolysis and amine-based absorption techniques are commercially mature technologies but expensive, while membrane separation and proton exchange membrane (PEM) electrolysis offer a good trade-off between high efficiency and low costs, making them suitable for further detailed feasibility assessments. Overall, the study provides valuable insights into the techno-economic potential of CCU technologies and their limitations, as well as recommendations for future research and policy development.

CRediT authorship contribution statement

M. Jibrán S. Zuberi: Methodology, Resources, Visualization, Conceptualization, Data curation, Formal analysis, Writing – original draft. **Arman Shehabi:** Funding acquisition, Methodology, Project administration, Resources, Supervision, Visualization, Writing – review & editing. **Prakash Rao:** Resources, Supervision, Visualization, Writing – review & editing, Funding acquisition, Methodology, Project administration.

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.

Data availability

Data will be made available on request.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.ijggc.2024.104137](https://doi.org/10.1016/j.ijggc.2024.104137).

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