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Scenario assessment of introducing carbon utilization and carbon removal technologies considering future technological transition based on renewable energy and direct air capture

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ABSTRACT

Carbon capture and utilization (CCU) and carbon dioxide removal (CDR) technologies have the potential to significantly contribute to GHG reduction. Numerous studies have evaluated the CO_2 reduction effects and economics of CCUs and CDRs; however, uncertainties in these evaluations due to various regional characterizations and future technological transitions are of high importance. In this study, four synthetic fuels (fuels produced from captured CO_2 and H_2), methanol, methane, gasoline, and diesel, were evaluated by life cycle assessment (LCA) and techno-economic assessment (TEA). Five representative countries with different regional characteristics were selected for the study. A bottom-up integrated LCA/TEA approach was used, involving detailed process simulations to avoid uncertainties and to evaluate future technological transitions based on direct air capture (DAC) and renewable energy. The overarching objective of this study was to provide a transparent framework with a common dataset generation methodology to determine the countries that have the advantages/disadvantages in synthetic fuel production and sale, considering the local operational parameters and large-scale introduction of DAC/renewable energy. Such multiple synthetic fuel analyses across various jurisdictions have not been conducted in previous studies. The results showed that the highest and lowest CO_2 reductions were achieved by diesel (average 6.33 kg-CO_2·kg^{-1}) and methanol (average 3.43 kg-CO_2·kg^{-1}), respectively, whereas the highest and lowest product costs were of gasoline (average 2.96 USD·kg^{-1}) and methanol (average 0.82 USD·kg^{-1}), respectively. The technological transition using DAC and renewable energy showed average CO_2 emission reductions of 47% (methanol), 99% (methane), 545% (gasoline), and 621% (diesel). Moreover, in the future, the lowest CO_2 emission reduction costs are expected in Germany for methanol and diesel, Australia for methane, and Canada for gasoline. These findings can contribute to improving international collaboration to promote CCU and CDR technologies.

1. Introduction

Reducing global greenhouse gas (GHG) emissions, such as CO_2, is vital for curbing global warming. In the 2015 Paris Agreement (United Nations, 2015), participating countries pledged to limit the average global temperature rise by 2 °C above pre-industrial levels (Dube et al., 2018). Nevertheless, achieving the Paris Agreement and carbon neutrality goals will be impossible, even if current efforts are continued, as CO_2 emissions in 2019 and 2020 were 35,966 Mt/y and 34,156 Mt/y, respectively (IEA, 2021). Thus, the introduction of low-carbon technologies on a large scale will be essential.

Carbon capture and utilization (CCU) technologies are potentially scalable solutions to convert CO_2 into fuel, chemical products, and minerals (Cuèllar-Franca and Azapagic, 2015; Islam et al., 2023; Pham et al., 2022; Hwa Teo et al., 2022), are currently attracting worldwide attention as technologies with the potential to significantly contribute to CO_2 reduction in the long term. To this end, various evaluations of CCU have been conducted from the perspective of energy efficiency, CO_2 reduction, and economics (Garcia-Garcia et al., 2021; Hepburn et al.,

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Abbreviations

AER	annual expense ratio
ASF	Anderson–Schulz–Flory
CAPEX	capital expenditure
CCU	carbon capture and utilization
CDR	carbon dioxide removal
DME	dimethyl ether
DEPG	dimethyl ethers of polyethylene glycol
FT	Fischer-Tropsch synthesis
GHG	greenhouse gas
IEA	International Energy Agency
LCA	life cycle assessment
$LCCO_2$	lifecycle CO_2 emission
LNG	liquid natural gas
LPG	liquid petroleum gas
MEA	monoethanolamine
MTG	methanol to gasoline
OPEX	annual operating expenditure
PSA	pressure swing absorption
RWGS	reverse water gas shift
SF	scale factor
SOEC	solid oxide electrolysis cell
TEA	techno-economic assessment
RE	renewable energy

2019; Markewitz et al., 2012; Thonemann, 2020); however, the CCU technology has not yet been implemented as a mainstream mode of CO_2 reduction at commercial stage. The latest International Energy Agency (IEA) report in 2020 indicates that only 21 CCU projects are operating worldwide, collectively capturing 40 Mt of CO_2 per year (IEA, 2020). The primary reason for the lack of mass adoption of CCUs is the need for large amounts of clean energy (Kätelhön et al., 2019). Considering the limited use of renewable energy, CCU requires analysis of the optimal CCU product in terms of CO_2 reduction potential and economic feasibility.

Currently, many evaluation studies are based on fossil fuel-derived flue gas/syngas as the source of CO_2 and hydrogen for CCU operations (Thonemann, 2020; Von der Assen et al., 2016). However, in the future, it must be considered that the use of fossil fuels will decrease to achieve a carbon-neutral global society. Therefore, to evaluate CCU introduction in the future, it is necessary to analyze optimal CCU products for CO_2 recovered from the atmosphere by carbon dioxide removal (CDR) technology, such as direct air capture (DAC) (Fuss et al., 2018; Minx et al., 2018), and carbon conversion technologies based on renewable energy.

However, these analyses are expected to vary region-wise as country specificities affect operation inputs. Depending on the CO_2 intensity and cost of grid electricity in each country, the cost of renewable energy, fuel used, CO_2 reduction effect, potential, economics, CDR technology effects, and optimal CCU product will inevitably vary. CO_2 capture and conversion technologies not only contribute to CO_2 reduction but also form the basis of important technologies regarding energy security. Briefly, countries need to supply energy to meet their own needs while aiming to meet CO_2 reduction targets. Considering the above limitations of renewable energy, international collaboration among CCUs and CDRs will be vital to achieving CO_2 reduction and energy security. Thus, the regional characteristics of CCUs and CDRs should be analyzed to accomplish significant future adoption.

The issue of the highest importance in the regional characterization of these CCUs including CDRs is the uncertainty of the assessment. There are numerous studies evaluating the CO_2 reduction effects and economics impacts of CCUs and CDRs through literature reviews (Hepburn et al., 2019; Naims, 2016; Thonemann, 2020). Although a detailed literature review is beyond the scope of this work, it is important to highlight the uncertainties resulting from various technology conditions. For instance, Hepburn et al. evaluated the cost and capacity of ten CCU pathways (Hepburn et al., 2019). Furthermore, Thonemann (2020) conducted a CCU comparative meta-analysis, while Garcia-Garcia et al. (2021) compared the results of the life cycle assessment (LCA) of various CCU products. However, it is necessary to reduce this uncertainty to the greatest degree when evaluating the regional characteristics of CCUs and CDRs and to scrutinize the operating conditions of specific technologies through process simulations rather than literature reviews. To date, although a few studies attempted to evaluate CCU and CDR via process simulations (Shi et al., 2020; Tripodi et al., 2020), there have been no examples of the selection of optimal CCU and CDR technology considering regional characteristics, even though accurate evaluations can be made for individual processes.

In this study, four synthetic fuels (fuels produced from captured CO_2 and H_2), methanol, methane, gasoline, and diesel, were selected as representative CCU products. This will help evaluate the most suitable products in terms of net GHG reduction and economic viability. In particular, this study evaluated the most viable synthetic fuels by selecting five representative countries with different regional characteristics. In addition, technological transitions, such as increased use of renewable energy and deployment of CDR technology to improve the efficiency of CCU pathways, can be expected in the future. To analyze this impact, "future scenarios" that adopts DAC and CO_2 conversion technologies based on renewable energy were evaluated. In comparison, a "conventional scenario" that uses CO_2 sources from fossil power plants was evaluated to assess how regional characteristics affect the optimal synthetic fuels during future technological transitions. The lack of a detailed LCA/techno- and technoeconomic assessment (TEA) studies comparing these two scenarios is hindering a more global approach to climate change solutions based on regional characteristics.

DAC has been previously compared with other CDR pathways using LCA and TEA (Erans et al., 2022), but seldom compared with flue gas-captured CCU pathways (Langie et al., 2022). Furthermore, previous studies do not provide multiple synthetic fuel analyses across various jurisdictions. More specifically, the novelty of this study is three-fold. First, a bottom-up integrated LCA/TEA approach to assess the impact of operating conditions involving in-house detailed process simulations was used. With this approach, uncertainties related to multiple data sources are minimized. Second, the proposed detailed process simulation is linked in each technology pathway to a country's assessment following large-scale deployment of the different CCU products while evaluating the impact of regional characteristics on climate change mitigation and economic viability. This will enable an improved evaluation of the role of CDR and conversion technologies in future energy transition scenarios. Third, as countries search for more effective global climate change solutions, the proposed bottom-up framework will provide a more harmonized jurisdiction-specific assessment of the best synthetic fuel vector to reduce the carbon footprint of heating and transportation. The overarching objective (hypothesis) of this study was to provide a transparent framework with a common dataset generation methodology to determine the countries that have the advantages/disadvantages in synthetic fuel production and sale, considering local operational parameters such as electricity and existing fuel costs.

2. Methods

LCA is widely employed to evaluate the environmental impact of a product system throughout its life cycle (Azapagic and Clift, 1999). In

this study, LCA and TEA were conducted for four synthetic fuels. This section describes a specific evaluation method for the CO_2 reduction effect and economic feasibility and a correlation analysis method between technological transition and regional characteristics. In this study, the intensity of CO_2 emissions and the cost of grid electricity, renewable energy, fuels, and products, as well as local taxes, were examined as regional characteristics.

2.1. Outline of lifecycle CO_2 estimation and system boundary definition

Several studies aimed at providing a harmonized LCA-based net GHG emissions accounting (Müller et al., 2020; U.S.DOE, 2022) to differentiate between actual physical emission removal from the atmosphere and avoided emission (Tanzer and Ramírez, 2019). Net emission reduction in the atmosphere, the main cause of global warming, should be the key indicator. The two main CDR pathways using conventional (monoethanolamine (MEA)) and future (DAC) carbon captures are the main building blocks of the two scenarios described in Fig. 1. The key implication of the proposed system boundary is related to the accounting approach for capturing CO_2.

This study analyzed the impact of technological transition by evaluating future scenarios that make effective use of CDR technology and renewable energy against conventional scenarios. The main difference between the two scenarios as summarized in Fig. 1 are:

- The inclusion of coal-fired power plants for emissions in the "conventional scenario" GHG accounting, whereby only 90% (Choi et al., 2019) of flue gas is captured and used downstream.
- While MEA CO_2 capture from coal-fired power plant flue gas is used in the "conventional scenarios," DAC is used in the "future scenarios."
- Reverse water gas shift synthesis (RWGS) is used for downstream conversion in "conventional scenarios," and co-electrolysis from CO_2 and water is used in "future scenarios."

This study aims at quantifying the life cycle CO_2 emissions (i.e., the amount of CO_2 emitted during the entire life cycle of the synthetic fuel, from the procurement of raw materials to product manufacturing and utilization), thereafter, referred to as $LCCO_2$. $LCCO_2$ is based on LCA defined in ISO 14040/14044 (ISO, 2006), and the system is bounded from CO_2 capture to synthetic fuel production. Therefore, it does not include CO_2 emission sources and is referred to as a "cut-off approach" in LCA. This study also quantifies net CO_2 reduction amount, synthetic fuel cost, and CO_2 reduction cost, which will be used as key indicators to compare the different CCU pathways.

The CCU pathways evaluated in this study consisted of multiple processes, such as CO_2 capture, hydrogen production, CO_2 conversion, and end-use. While considering local characteristics (e.g. energy intensity), these CCU pathways are assessed in five countries study: the United States (U.S.), Canada, Germany, Australia, and Japan. These countries were selected to include different correlations between renewable energy generation and fuel demand, as well as geopolitical conditions, such as island nations. This study also considers the introduction of CCU technologies within each country's carbon-neutral goals, under the conventional and future scenarios (Fig. 1). Specifically, the "conventional scenario" was based on fossil fuels displacement, while the "future scenario" was based on the significant introduction of CDR technology and renewable energy.

Various technologies have been proposed for CO_2 conversion to produce different CCU products (Jones and Shu, 2009; Marchese et al., 2020; Zhang et al., 2017). However, to compare the different CCU pathways in this study, it was assumed that the process conditions must be harmonized as much as possible. Therefore, the same CO_2 conversion production pathway was adapted for the production of methanol, methane, and gasoline under the conventional and future scenarios.

The process flow and system boundary of each CCU pathway are shown in Fig. 2. More detailed process flows are described below for the four synthetic fuels covered in this study.

2.2. Process simulation and optimization

To evaluate $LCCO_2$, the mass and energy balances of the CO_2 capture and conversion processes were calculated using process simulations. In particular, the process simulator PRO/II version 10.2 and ASPEN Plus version 12 were applied to quantify the material balance and energy consumption. Detailed process flow diagram of each pathway would be available upon request. Process design details for each key unit used in the different CCU pathways are provided below. The primary energy source to satisfy heat requirements in each CCU pathway was based on the energy consumption of the fuel in each country (IEA, 2021; IEA Energy and carbon tracker, 2021). Consequently, natural gas was used by Japan, the United States, and Canada, whereas coal was used by Germany and Australia.

2.2.1. CO_2 capture and separation

In the conventional scenario, the CO_2 capture from the flue gases of

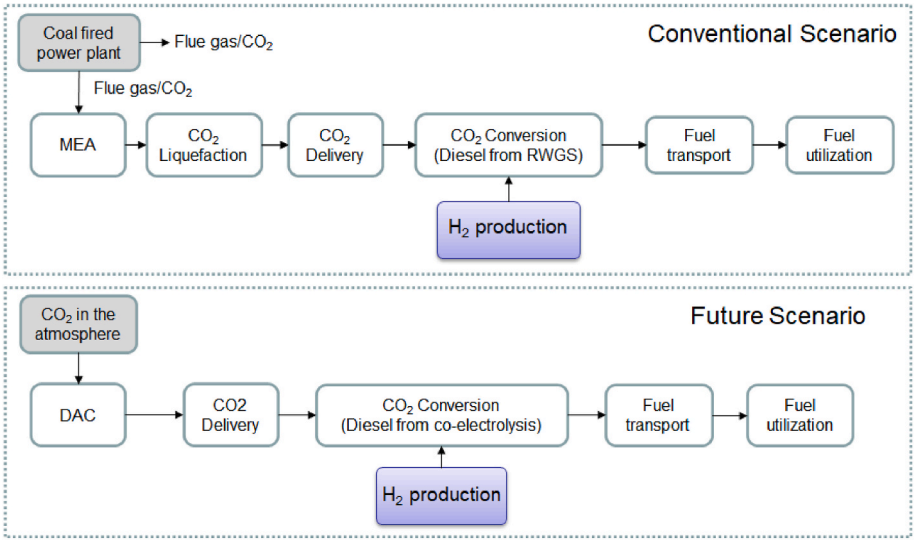

Fig. 1. Illustration of the system boundary and key process steps included in the conventional (with MEA) and future (with DAC) scenarios.

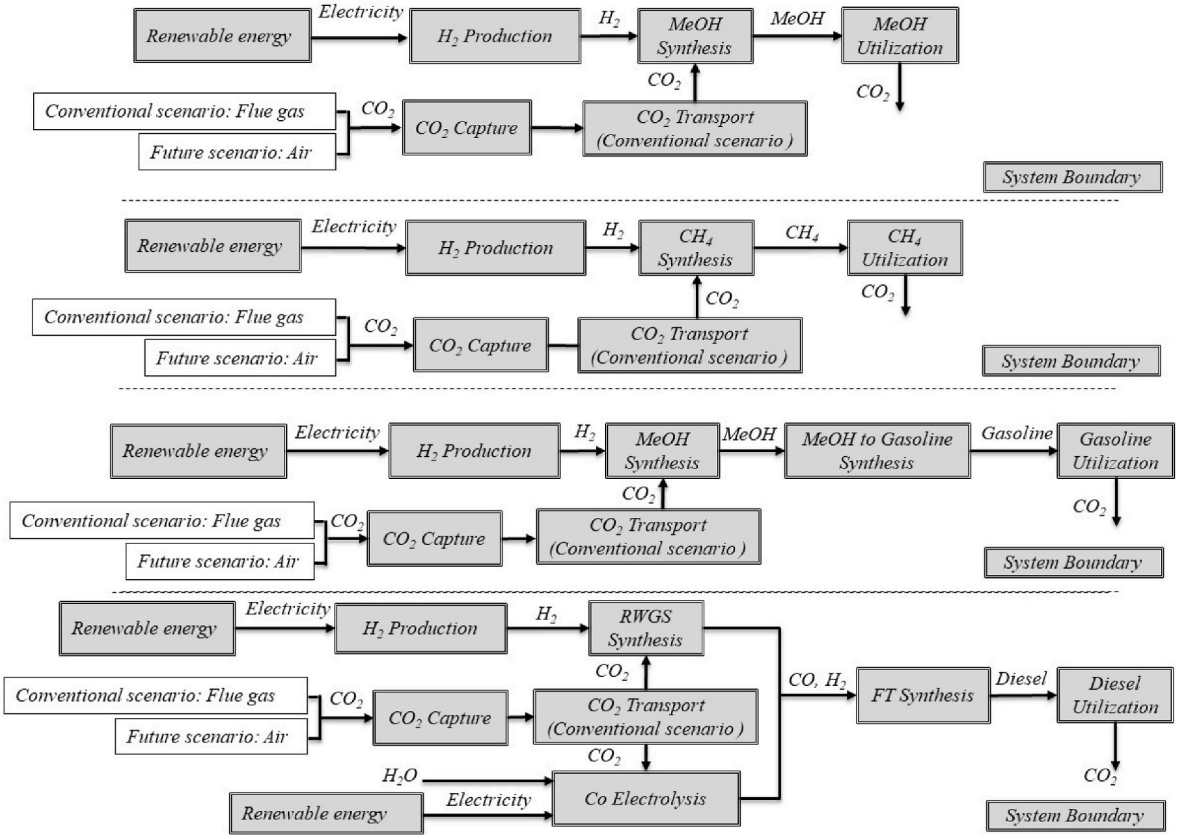

Fig. 2. High-level descriptions of process flow and system boundary for the production of synthetic fuels under Scenario 1 and Scenario 2.

coal-fired power plants, which are the main sources of CO_2 emissions in any country, was assumed, and MEA was used as the chemical absorption method. Process design and simulations were performed to capture 90% CO_2 from flue gas, as described in the literature (Choi et al., 2019; Morimoto et al., 2022). After absorption, MEA regeneration and CO_2 purification (i.e., 99% or higher purity) were conducted using a stripper column that supplied heat. The CO_2 concentration in the flue gases of coal-fired power plants was 13% (Tang et al., 2014a, 2014b), and the reboiler temperature in the stripper column was set to 118 °C.

The process simulation of CO_2 separation and recovery in DAC was carried out by referring to the KOH process developed by Carbon Engineering Ltd. (Keith et al., 2018; Kishida, 2022). In this process, CO_2 separation and recovery from the air consisted of the following four chemical reactions:

$$CO_2(g) + 2KOH \longrightarrow H_2O + K_2CO_3 - 95.8\ kJ \bullet mol^{-1} \quad (1)$$

$$K_2CO_3 + Ca(OH)_2 \longrightarrow 2KOH + CaCO_3(s) + 5.8\ kJ \bullet mol^{-1} \quad (2)$$

$$CaCO_3(s) \longrightarrow CaO(s) + CO_2(g) + 178\ kJ \bullet mol^{-1} \quad (3)$$

$$CaO(s) + H_2O(l) \longrightarrow Ca(OH)_2 - 63.9\ kJ \bullet mol^{-1} \quad (4)$$

In this DAC process, the CO_2 in the air was absorbed as K_2CO_3 by the air contactor KOH aqueous solution (Eq. (1)). The absorbent was circulated into the air contactor, where K_2CO_3 reacted with the supplied $Ca(OH)_2$, producing $CaCO_3$(s) and KOH (Eq. (2)). $CaCO_3$(s) was heated (calcined) to recover CO_2 via the combustion of methane with pure oxygen at 900 °C (Eq. (3)). Eventually, moisture and CaO from the calciner reacted to form $Ca(OH)_2$, as shown in Eq. (4). More detail process is described in Supplementary Information (SI), "Detail process description of direct air capture." and in Keith et al. (2018).

CO_2 capture requires the construction of an MEA unit adjacent to the coal-fired power plants; In the case of DAC, there was no restriction on the location. Therefore, in this study, only CO_2 captured by MEA was included in the process of compressing and liquefying the recovered CO_2 to 15.1 MPa and transporting the liquefied CO_2 by a truck using diesel fuel for 30 km.

2.2.2. H_2 production

For H_2 production, alkaline water electrolysis using electricity from renewable energy sources was assumed in both scenarios. In this study, detailed process simulations were not performed for the H_2 production process as this study focused on CO_2 separation and CO_2 conversion technologies. Therefore, energy consumption, utility, and cost were obtained from the literature (IAE, 2019; Navarro et al., 2015) on alkaline water electrolysis.

2.2.3. Methanol synthesis

A direct methanol synthesis from CO_2 and H_2 by Eqs. (5) and (6) was assumed for both conventional and future scenarios as described elsewhere (Morimoto et al., 2021). In this process, CO_2 and H_2 pressures were increased to reach the reaction pressure and delivered to an adiabatic reactor after heating. The outlet gas containing methanol synthesized in the adiabatic reactor was cooled and separated into gas and liquid phases. Next, the unreacted gas was again pressurized, heated, and supplied to the isothermal reactor, to further stimulate the formation of methanol, which was cooled after the reaction and collected through the adiabatic reactor outlet. The process is described in more detail in SI, "Detail process description of methanol synthesis."

$$CO_2 + 3H_2 \rightleftharpoons CH_3OH + H_2O - 49\ kJ \bullet mol^{-1} \quad (5)$$

$$CO_2 + H_2 \rightleftharpoons CO + H_2O + 41\ kJ \bullet mol^{-1} \quad (6)$$

2.2.4. Methane synthesis

Methane production will follow the Sabatier reaction with CO_2 and H_2 as key reactants (Zhang et al., 2017). As shown in Eq. (7) and described elsewhere (Morimoto et al., 2022) methane and water are the main products in both the conventional and future scenarios. CO_2 and H_2 were heated and pressurized to the reaction temperature and pressure, respectively, and sent to the reactor. By assuming that the methanation reaction proceeds to an equilibrium state using an adiabatic reactor with PRO/II, the calculation result of the increased temperature was 649.7 °C at 1.35 MPa, as indicated by Eq. (7) below:

$$CO_2 + 4H_2 \rightleftharpoons CH_4 + 2H_2O - 164.9kJ \bullet mol^{-1} \quad (7)$$

In Eq. (7), it was assumed that steam was produced by the heat generated by the reaction, and all electricity in the process was provided by the electricity generated by the steam turbine.

2.2.5. Gasoline synthesis

Gasoline is obtained by converting methanol produced from CO_2/H_2 using the methanol to gasoline (MTG) technology. As described in the literature (Jones and Shu, 2009), methanol is pressurized, vaporized, preheated, and delivered to dimethyl ether (DME) reactor, where a part of the methanol is converted into DME and water. The outlet gas of the DME reactor was mixed with recycled gas and sent to the MTG reactor. In the MTG reactor, 100% of methanol and DME were converted to water and hydrocarbons. The MTG reactor outlet gas was cooled and separated into water, liquid hydrocarbons, and gases. Gas was recycled to control the temperature of the MTG reactor. Liquid hydrocarbons are separated into fuel gas, liquid petroleum gas (LPG), light gasoline, and heavy gasoline by deethanizers, debutanizers, and gasoline splitters. In addition, hydrogen isomerization was performed to decompose and remove Durene (1, 2, 4, 5-tetramethyl benzene) contained in the heavy gasoline.

A more detailed process is described in SI "Detail process description of gasoline synthesis."

2.2.6. Diesel synthesis

In the conventional scenario, Fischer-Tropsch (FT) synthesis was performed by the RWGS from CO_2 and H_2 to CO and H_2O, according to Eq. (8) (Marchese et al., 2020; Park et al., 2001; Zhang et al., 2018; Zhu et al., 2020).

$$CO_2 + H_2 \rightarrow CO + H_2O + 41.2kJ \bullet mol^{-1} \quad (8)$$

For diesel production by RWGS, H_2 and raw CO_2 were used at a 1:3 M ratio and pressurized to 0.5 MPa in a compressor, heated with hot flue gas from the RWGS reactor, and fed into an RWGS reactor. CO and H_2 from RWGS are then fed to the FT reaction process. The process is described in more detail in SI, "Detail process description of diesel synthesis by RWGS."

In the case of the future scenario, the reported process design (Becker et al., 2012) was used, in which a mixture of raw CO_2, unreacted CO_2 retrieved in the recovery process, H_2, and H_2O (steam) was heated in a furnace for co-electrolysis using a solid oxide electrolysis cell (SOEC) reactor.

The electrolytic reactions of CO_2 and H_2O in the SOEC reactor were converted to H_2 and CO, respectively, using Eqs. (9)–(11) in the thermodynamically and kinetically desirable temperature range of 700–900 °C on a Ni catalyst.

$$H_2O \rightarrow H_2 + \frac{1}{2}O_2 + 249kJ \bullet mol^{-1} \quad (9)$$

$$CO_2 \rightarrow CO + \frac{1}{2}O_2 + 283kJ \bullet mol^{-1} \quad (10)$$

$$H_2 + CO_2 \rightarrow H_2O + CO + 34kJ \bullet mol^{-1} \ (\text{at } 860\ ^\circ C) \quad (11)$$

CO and H_2 were fed into the FT reactor, and the reaction was carried out at a high temperature and pressure to synthesize hydrocarbons. The process is described in more detail in SI, "Detail process description of diesel synthesis by co-electrolysis."

2.3. Estimation of energy consumption and $LCCO_2$

2.3.1. Concept introduction

In this study, $LCCO_2$ was calculated for each synthetic fuel based on the embedded carbon of equipment, embedded carbon of feedstock, and energy consumption throughout the product lifecycle including utilization. Thus, $LCCO_2$ emission comprises the total amount of CO_2 emitted per ton of synthetic fuel during the production phase. In addition, emissions during the utilization phase should be included in the overall CO_2 balance. For each CCU pathway, the energy (electric power and heat) and material consumption required for all processes were assessed. Notably, neither the amount of energy consumed nor the CO_2 emissions generated during plant construction were included in the evaluation. The total lifecycle CO_2 emission volume $LCCO_2$ (kg-CO_2·(t-CH_4) $^{-1}$) was quantified using Eq. (12):

$$LCCO_2 = \sum_{i=0}^{m}\left(E\_po_i + E\_he_i + \sum_{j=0}^{n} M_{i,j}\right)C \quad (12)$$

where the power consumption of each process i (maximum, m) is E_po_i (MWhe·t^{-1}), the heat consumption is E_he_i (MJ·t^{-1}), and the consumption of material j (maximum, n) in the process i is $M_{i,j}$ (kg·t^{-1}), with a CO_2 emission intensity of C (kg-CO_2·t^{-1}).

In this study, CO_2 was captured from coal-fired power plant exhaust gas in the conventional scenario, while CO_2 was captured from the atmosphere in a future scenario. In both scenarios, synthesized fuels are burned during the use phase, and CO_2 returns to the atmosphere with one key difference. In the conventional scenario, CO_2 originated from fossil power plants, whereas in the future scenario, CO_2 originated from the atmosphere. As a consequence, in this study, after the combustion of synthetic fuel in the conventional scenario, CO_2 is emitted into the atmosphere using fossil fuels, whereas, in the future scenario, CO_2 emissions are negative as the scenario does not use fossil fuels as originally supposed. However, in practice, CO_2 captured using DAC will return to the atmosphere through the combustion of synthetic fuels in future scenarios, thus making CCU close to a carbon-neutral technology.

CO_2 reduction was obtained by subtracting the $LCCO_2$ of products manufactured by CCU from the existing products for both scenarios. In this study, $LCCO_2$ and CO_2 reductions were calculated for four synthesized fuels and five countries for the current and future scenarios, and the effects of technological transition and regional characteristics on CO_2 reduction were analyzed by comparing them.

2.3.2. Energy and material consumption

In this study, the energy and material consumption was determined by process simulation for CO_2 separation and recovery, CO_2 conversion, and CO_2 liquefaction. For other processes, data inputs are obtained from the literature. Table 1 summarizes the process simulation results for each process unit with the source of data. These data do not consider regional variations.

The electricity consumption (MWhe) and heat consumption (MJ) were also included in the analysis. CO_2 emissions from renewable energy electricity were assumed to be zero (IPHE, 2022). In this study, the electricity consumed for H_2 water electrolysis and co-electrolysis for diesel synthesis was assumed to be obtained from renewable energy sources. The CO_2 emission intensity of grid power for Japan and the other countries included was obtained from the Ministry of the Environment of Japan (Ministry of the Environment, 2020) and the IEA (IEA, 2022). The CO_2 emission intensities of liquefied natural gas (Ministry of the Environment, 2020), MEA (IDEA, 2019), water (IDEA, 2019), and CH_4 mining (IDEA, 2019) were obtained from the literature.

Table 1
Energy consumption and other related values of the total system.

Process		Value	Unit	Ref.
CO_2 Capture (MEA absorption)	Heat consumption	2.58	GJ·t-CO_2^{-1}	Simulation
	Power consumption	0.02	MWhe·t-CO_2^{-1}	Tang et al. (2013)
	MEA consumption	2.40	kg·t-CO_2^{-1}	
CO_2 Capture (DAC)	Heat consumption	7.34	GJ·t-CO_2^{-1}	Simulation
	Power consumption	0.39	MWhe·t-CO_2^{-1}	
	H_2O consumption	4.62	t·t-CO_2^{-1}	
	KOH consumption	0.56	kg·t-CO_2^{-1}	
	$CaCO_3$ consumption	28.0	kg·t-CO_2^{-1}	
CO_2 Liquefaction	Power consumption	0.18	MWhe·t-CO_2^{-1}	Simulation
CO_2 Transport (truck)	Diesel fuel	0.08	L·(t·km)$^{-1}$	(IDEA, 2019)
H_2 Water electrolysis	Power consumption	47.83	MWhe·t-H_2^{-1}	Navarro et al. (2015)
MeOH Synthesis	Power consumption	0.49	MWhe·t-MeOH^{-1}	Simulation
CH_4 Synthesis	Power consumption	0.10	MWhe·t-CH_4^{-1}	
MTG Synthesis	Power consumption	2.09	MWhe·t-gasoline^{-1}	
Diesel Synthesis (RWGS and FT)	Power consumption	1.74	MWhe·t-diesel^{-1}	
Diesel Synthesis (SOEC and FT)	Power consumption	30.44	MWhe·t-diesel^{-1}	
	H_2O consumption	1.27	t·t-diesel^{-1}	

2.4. Evaluation of CO_2 reduction cost

2.4.1. Concept introduction

The CO_2 reduction cost was considered to evaluate the economic viability of synthetic fuels. In general, synthetic fuels are more expensive than existing products including methanol, natural gas, gasoline, and diesel fuel. As described above, the $LCCO_2$ of these products is expected to be lower when compared to current products. To evaluate capture cost and net emission in a single indicator, incremental cost per unit amount of CO_2 reduction (USD·tCO_2^{-1}) was estimated as shown in Eq. (13). This new indicator has been used as an economic evaluation (Tripodi et al., 2020).

$$Cost_{reduction} = \frac{Prod\_cost_{CCU} - Prod\_cost_{current}}{LCCO2_{current} - LCCO2_{CCU}} \quad (13)$$

where the $LCCO_2$ of synthetic fuels was set as $LCCO_{2CCU}$ (kg CO_2·t-products^{-1}), the cost was set as $Prod\_cost_{CCU}$ (USD·t-products^{-1}), the $LCCO_2$ of existing products was set as $LCCO_{2current}$ (kg CO_2·t-products^{-1}), and the cost was set as $Prod\_cost_{current}$ (USD·t-products^{-1}). The cost of synthetic fuels $Prod\_cost_{CCU}$ (USD·t-products^{-1}) of the entire process shown in Fig. 2 (i.e., CO_2 capture, H_2 production, energy carrier liquefaction, transportation, and CO_2 conversion) was evaluated by dividing the total annual expenditure by the amount of synthetic fuel produced.

Annual expenditure reflects the estimated capital expenditure (CAPEX) combined with the annual expense ratio (AER) and annual operating expenditure (OPEX). The CAPEX of each process was calculated based on estimates from the literature and results from the process simulation. Although various calculation methods have been proposed for the AER, in this study, it was assumed that the useful life of the equipment is 20 years (5% per year), with 1% added for maintenance and insurance for a total of 7%. As the capital scale varied depending on the study referenced, a scale factor (SF) of 0.7 (Swernath et al., 2015) was integrated into the literature-derived ratio and the capital scale developed for the processes.

The CAPEX in process i was set as $CAPEX\_CCU_i$ (USD), and the production amount from this process was set as PRD_CCUi (t·y^{-1}). The CAPEX data obtained from the literature listed in Table 2 were set as $CAPEX\_LIT_i$ (USD), and the production amount in the same literature was set as PRD_LITi (t·y^{-1}). These estimates were obtained using Eq. (14).

$$CAPEX\_CCU_i = CAPEX\_LIT_i \left(\frac{PRD\_CCU_i}{PRD\_LIT_i} \right)^{SF} \quad (14)$$

The operating expenditure, $OPEX\_CCU_i$ (USD·t-products^{-1}), was calculated based on the amount of energy and raw material consumed in the entire process, as shown in Eq. (15):

$$OPEX\_CCU_i = E\_po_i \bullet El_{price} + E_{hei} \bullet Eh_{price} + \sum_{j=0}^{n} M_{i,j} \bullet M\_price_{i,j} \quad (15)$$

where E_poi (kWhe·t-products^{-1}), E_hei (GJ·t-products^{-1}), and $M_{i,j}$ (kg·t-products^{-1}) are defined in Eq. (15). El_price (USD·kWhe^{-1}), Eh_price (USD·GJ^{-1}), and $M\_price_{i,j}$ (USD·kg^{-1}) are the prices of electricity, heat, and raw material j, respectively, used in the entire process.

When the annual synthetic fuel production amount was CCU_prod (t·y^{-1}), the $Prod\_cost_{meth}$ (USD·t-products^{-1}) of all synthetic fuels in the five countries was calculated using Eq. (16):

$$Prod\_cost_{CCU} = \sum_{i=0}^{m} \left(\frac{CAPEX\_CCU_i \bullet AER}{CCU\_prod} + OPEX\_CCU_i \right) \quad (16)$$

In this study, the cost of these synthetic fuels and the cost of CO_2 reduction were compared between the conventional and future scenarios for five countries, analyzing the impact of technological transition and regional characteristics on the CO_2 reduction effect.

2.4.2. Data input inventory

The unit prices of grid electricity and renewable energy (RE) and fossil fuel prices for 2020, and CAPEX for the five countries evaluated in this study are listed in Table 2. The existing product cost, $Prod\_cost_{current}$, is listed in SI "Existing products cost Table 2–1" For RE in the five countries, the energy source with the largest installed volume for each country (i.e., photovoltaic energy for Japan and Australia, wind energy for Germany and the U.S., and hydraulic energy for Canada) was selected (IEA, 2021; IEA Energy and carbon tracker, 2021). The fuel price was obtained from the fuel used in each country (defined in 2.2). The exchange rate was set at 114 USD/JPY.

The prices shown in Table 2 are highly uncertain, especially the electricity price, which is highly variable and has a large impact on the results. To assess this impact, sensitivity analysis was conducted for RE prices. Specifically, the costs of hydro, PV, and wind energies in 2010 were 52.3% higher on an average than those in 2020 (IRENA, 2021). Therefore, the economic feasibility was evaluated using the RE prices shown in Table 2, and prices were 52.3% higher than those values.

For the material price $M\_price_{i,j}$, the following values were used: 0.04 USD·kg^{-1} for steam (H_2O) (Enetrap® official website, 2022), 0.87 USD·kg^{-1} for MEA (New Energy and Industrial Technology Development Organization, 2002), 0.001 USD·kg^{-1} for industrial water (Bureau of Waterworks Tokyo Metropolitan Government, 2022), 22.81 USD·kg^{-1} for KOH (Kishida Chemical, 2022), 0.003 USD·kg^{-1} for $CaCO_3$ 35 USD·kg^{-1} (Center for Low Carbon Society Strategy, 2020).

3. Results

This section describes the $LCCO_2$ and CO_2 reduction cost assessments of synthetic fuels in conventional and future scenarios. Regional characteristics within the five countries are considered in these assessments. It also describes the correlation between CO_2 reduction potential and

Table 2
Electricity price and CAPEX.

	Value	Unit		Unit	Ref.
Electricity price			Fuel price		(Coal-Price.com, 2022; GlobalPetrolPrices.com, 2022; IRENA, 2021; Shin denryoku net, 2022)
Japan	0.18 (Grid), 0.06 (RE)	USD·kWhe^{-1}	14.00	USD·kJ^{-1}	
Canada	0.09 (Grid), 0.04 (RE)		6.39		
Germany	0.26 (Grid), 0.04 (RE)		5.00		
U.S.	0.12 (Grid), 0.04 (RE)		11.67		
Australia	0.15 (Grid), 0.06 (RE)		3.00		
CAPEX	*CAPEX_LIT$_i$*		*PRD_LITi*		
CO_2 capture (MEA absorption)	8.51×10^5	USD·(t-CO_2·h)$^{-1}$			Sano et al. (2021)
CO_2 capture (DAC)	1.61	BUSD	0.97	Mt-CO_2·y^{-1}	Simulation
CO_2 liquefaction	0.09	BUSD	1.54	Mt-CO_2·y^{-1}	Nishimura et al. (2017)
CO_2 transport	15.20	USD·t-CO_2^{-1}	0.79	Mt-CO_2·y^{-1}	(IDEA, 2019)
H_2 production	2.17	BUSD	0.23	Mt–H_2·y^{-1}	(IAE, 2019)
MeOH Synthesis	1.03	BUSD	3.50	Mt-MeOH·y^{-1}	Simulation
CH_4 Synthesis	0.27	BUSD	0.55	Mt-CH_4·y^{-1}	(IAE, 2018)
MTG Synthesis	1.31	BUSD	1.00	Mt-Gasoline·y^{-1}	Simulation
Diesel Synthesis (RWGS and FT)	5.90	BUSD	1.02	Mt-Diesel·y^{-1}	Simulation
Diesel Synthesis (Co-electrolysis and FT)	1.44	BUSD			Simulation

economic viability.

3.1. Amount of CO_2 reduction and cost of synthetic fuels

Fig. 3 summarizes the results comparing the amount of CO2 reduction and cost of synthetic fuels for the four CCU pathways in the five countries. A trend could be observed for both conventional and future scenarios.

As shown in Fig. 3, methane had the largest CO_2 emission reduction (average 2.58 kg-CO_2·kg^{-1}), and gasoline had the smallest CO_2 emission reduction (average 1.34 kg-CO_2·kg^{-1}) in the conventional scenario. Diesel had the highest product cost (average 2.70 USD·kg^{-1}), and methanol had the lowest cost (average 0.71 USD·kg^{-1}). For the future scenario, the largest and smallest CO_2 reductions were achieved for diesel (average 6.33 kg-CO_2·kg^{-1}) and methanol (average 3.43 kg-CO_2·kg^{-1}), respectively, whereas the highest product costs were spent for gasoline (average 2.96 USD·kg^{-1}) and the lowest for methanol (average 0.82 USD·kg^{-1}). By technological transition from conventional to future scenarios, the CO_2 emission reduction increased by 47% (methanol), 99% (methane), 545% (gasoline), and 621% (diesel) on average.

Methanol had the lowest product cost in both scenarios. However, differences in CO_2 reductions and costs between the conventional and future scenarios for methanol were relatively small among the products. For methane, although the product cost did not change significantly between the two scenarios, CO_2 emission reductions increased. In the case of gasoline and diesel, the CO_2 emission reductions were even greater than those for methane; consequently, diesel was the product with the highest CO_2 emission reductions in the future scenario.

The possible reasons for the above observations are that methane is the most relevant source of hydrogen among the RE sources in the conventional scenario, resulting in the largest CO_2 reduction. In the future, diesel production, which involves CO_2 co-electrolysis, will use a large amount of RE, resulting in the largest reduction in CO_2 emissions. The overall CO_2 reduction in the future scenario is larger than that in the conventional scenario, given that the DAC is used as the CO_2 capture

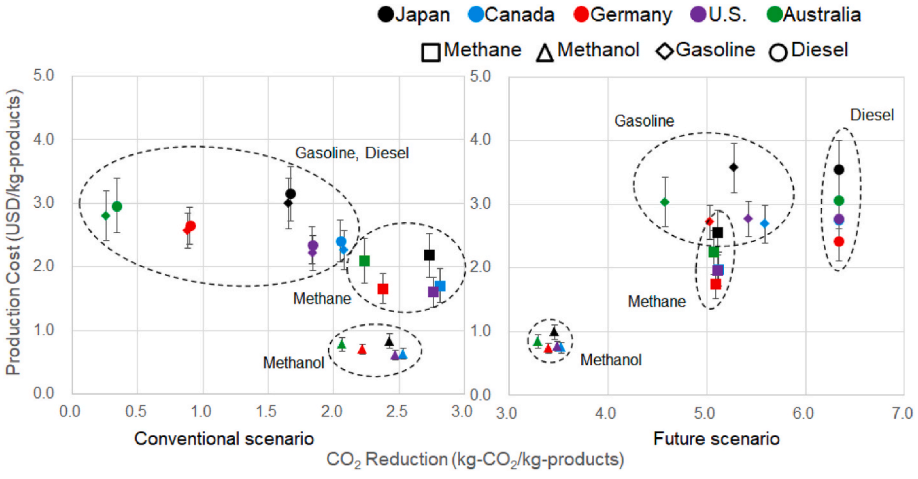

Fig. 3. CO_2 reduction and cost of synthetic fuels.

technology. This trend was clearly observed for diesel.

Regarding regional characteristics, the U.S. and Canada have relatively large CO_2 reductions for all products, whereas Australia and Germany have the lowest CO_2 emission reductions. This trend does not change significantly under either the conventional or future scenario, largely due to the single source of CO_2 emissions from the grid electricity in each country.

The production costs involve grid electricity and the unit cost of electricity from RE sources. As shown in Table 2, Japan has the highest cost in both scenarios, while the U.S. (for methane, gasoline, and diesel) and Canada (for methanol) had the lowest costs in the conventional scenario, while Germany (for methanol, methane, and diesel) and Canada (for gasoline) had the lowest costs in the future scenario. The reason for this trend is the low prices of electricity, RE, and heat in each country. Low fuel prices for CO_2 transportation, especially in the U.S., also contribute to the low synthetic fuel costs in the conventional scenario. As shown in Table 2, although the type of RE does not significantly affect the costs, it affects the scale of deployment of the CCU technology.

Considering the transition from conventional to future scenarios, scaling-up technology, changes in product demand, and risks related to public acceptance must be considered. Previous studies on methane have shown that the effect of technology scale-up can reduce the CO_2 reduction cost by 15%–31% (Morimoto et al., 2022).

Fig. 4 shows the results of the calculations for the CO_2 reduction cost breakdowns for the four synthetic fuels within each of the five countries based on the data shown in Table 2 (the year 2020). This figure helps to compare the evolution from the current to future scenarios.

As shown in Fig. 4, the CO_2 reduction costs were negative in both scenarios for German methane production, as synthetic fuels are less expensive than existing products.

The lowest CO_2 reduction cost was methanol in both scenarios, with an average of 0.11 USD·kg^{-1} in the conventional scenario and 0.10 USD·kg^{-1} in the future scenario. This indicates that for methanol there is little difference between the two scenarios. On the other hand, the highest CO_2 reduction cost was for gasoline, with an average of 1.43 USD·kg^{-1} and 0.24 USD·kg^{-1} in the conventional and future scenarios, respectively. Changing from the conventional to the future scenario lowers the CO_2 reduction costs by 1.18 USD·kg^{-1} for gasoline and 0.14 USD·kg^{-1} for diesel fuel, with gasoline having the largest impact. The average CO_2 reduction cost for methane was 0.24 USD·kg^{-1} under the conventional scenario and 0.18 USD·kg^{-1} under the future scenario, indicating that a change in scenario lowers the CO_2 reduction cost by just 0.07 USD·kg^{-1}.

The overall reason for the higher CO_2 reduction cost for gasoline is that the difference in cost between the synthetic fuels and existing products is small relative to the amount of CO_2 reduction. This is especially true for Australian gasoline in the conventional scenario and Japanese diesel fuel in the future scenario. Regarding the comparison between conventional and future scenarios, although Fig. 3 shows that diesel fuel had the largest CO_2 reduction, Fig. 4 shows that gasoline had the largest CO_2 reduction costs. This is because CO_2 reductions will be larger as SOEC is used for diesel fuel synthesis in future scenarios, but the CO_2 reduction costs will not decrease significantly due to increased equipment costs.

Regarding regional characteristics, the lowest CO_2 reduction costs were found in Germany for methanol, Japan for methane, and Canada for gasoline and diesel in the conventional scenario. In the future scenario, the lowest CO_2 emission reduction costs are observed in Germany for methanol and diesel, Australia for methane, and Canada for gasoline. The cost of products in Germany is the highest among the five countries, as shown in Fig. 3; however, the cost of existing products is also elevated, resulting in the lowest CO_2 reduction costs for methanol and diesel fuel. The results shown in Figs. 3 and 4 indicate that in the future scenario, Germany has an advantage in producing methanol and diesel fuels, whereas Canada has an advantage in producing gasoline.

These results are mostly due to the difference in the cost of synthetic fuels relative to each country's product costs and taxes. As shown in Table 2–1 (SI), the level of taxation on each product varies significantly between the five countries. This study showed that the impact of taxes is particularly significant for gasoline and diesel fuels. Especially in Germany, it was also observed that the CO_2 reduction cost was negative in

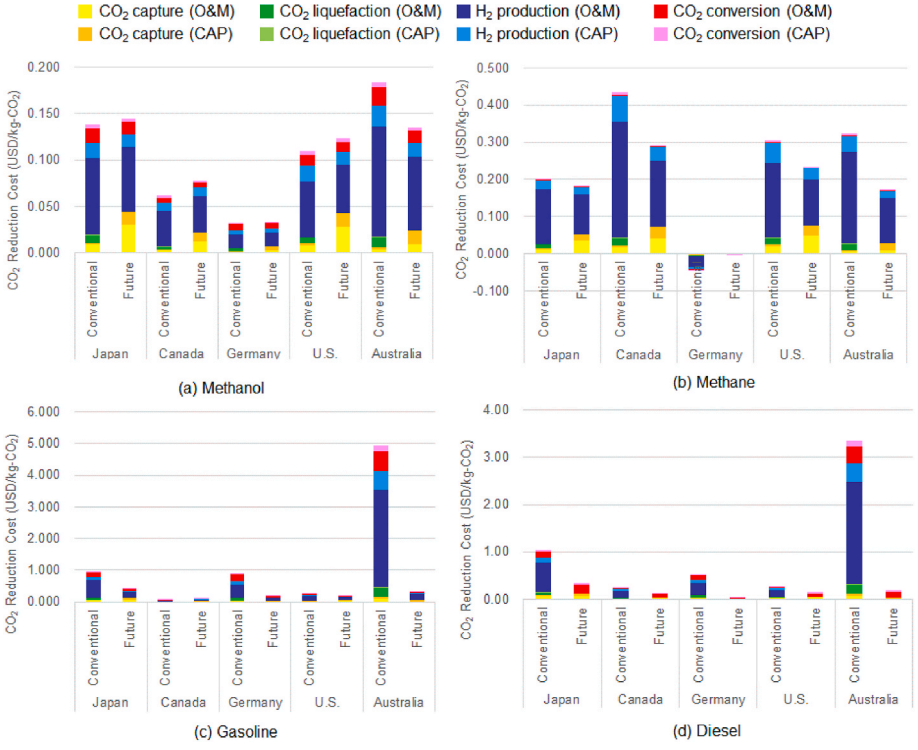

Fig. 4. CO_2 reduction costs of each synthetic fuel.

methane owing to high taxes.

CO_2 recovery costs for different CDR technologies have been examined by Minx et al. (2018). For example, the CO_2 recovery cost of bioenergy with carbon capture and storage is 0.06–0.16 USD·kg^{-1} whereas for enhanced weathering, it is 0.02–0.42 USD·kg^{-1}. As these costs are lower than those of DACs, the combination of CDR technologies other than DACs and CCUs may result in even lower CO_2 reduction costs than the present results.

3.2. CO_2 reduction potential and cost-effectiveness analysis

Fig. 5 shows the results of the analysis of the CO_2 reduction cost (the bottom of the error bar indicates the value in 2020, as shown in Fig. 4) and the potential CO_2 reduction in synthetic fuels in conventional and future scenarios. Note that the negative values in Fig. 4 are not shown.

In any country, the most effective synthetic fuels are those with the lowest CO_2 reduction cost and the highest potential.

As shown in Fig. 5, the most effective synthetic fuel in the conventional scenario was methane. At the same time, it was also important to note that U.S. gasoline and diesel fuel had a highly similar CO_2 reduction cost and greater potential than methane in Japan, Canada, Germany, and Australia.

The tendencies indicated in the conventional scenario shifted to future scenarios. For example, in the U.S., methane had a CO_2 reduction potential of 3193.4 Mt y^{-1} at a CO_2 reduction cost of 0.24 USD·kg^{-1}, while gasoline and diesel fuel had a CO_2 reduction potential of 1839.5 Mt·y-1 and 1290.4 Mt y^{-1} at even lower costs. Similarly, in Canada, methane has a CO_2 reduction potential of 402.4 Mt y^{-1} at a CO_2 reduction cost of 0.29 USD·kg^{-1}, whereas gasoline and diesel have a CO_2 reduction potential of 193.2 Mt y^{-1} and 143.7 Mt y^{-1} at considerably lower costs. In future scenarios in Japan, Germany, and Australia, methane will be the most effective synthetic fuel.

These evaluation results were verified by comparison with published data (Keith et al., 2018) as shown in Table 1 and by comparison of trends with published data for synthetic fuels (Kannangara et al., 2022).

4. Discussion and future work

In this study, $LCCO_2$, CO_2 reduction amount, synthetic fuel cost, CO_2 reduction cost, total CO_2 reduction potential, and their trade-offs were analyzed for four synthetic fuels in five countries. In particular, the impact of regional characteristics was analyzed by clarifying the relationship between the amount of CO_2 reduction and economic feasibility in the current and future scenarios. The impact of technological transition in the capture (from MEA to DAC) and the conversion (from thermal to electrochemical) phases have been also assessed.

As shown in Figs. 3–5, the country with the highest economic CO_2 reduction for each of the four synthetic fuels was identified by analyzing the following factors: the unit price of electricity and heat, product prices, taxes, and CO_2 transportation costs. Country-specific tax policy significantly impacted the introduction of synthetic fuels, as quantitatively illustrated in this study. Therefore, improving the existing tax policy is an effective means to enable the deployment of CCU solutions at a larger scale. For example, it would be effective to introduce policies such as tax incentives for CCU products or expanding carbon credits.

When comparing conventional and future scenarios, it is clear that introducing CDR technologies, such as DAC and CO_2 conversion technologies that use RE, can significantly enhance the effectiveness of CCU solutions. However, implementing this strategy would require a significant increase in RE capacity.

This work showed that reducing cost and GHG of H_2 production step. Even with the assumption of zero carbon footprint of renewable, H_2 production is the most significant contributor. As a potential suggestion for future work, the impact of advanced H_2 production technology must be considered. Another option for future work, is to consider technologies using direct thermochemical (Rumayor et al., 2019) and electrochemical (Dominguez-Ramos and Irabien, 2019) CO_2 conversion processes without the use. As shown previously, biocarbonate technologies provide a pathway to reduce cost and GHG emission for CO_2-to--diesel (M. Kannangara et al., 2022). However, comparing the results of this study with past reports raises some challenges due to differences in terms of methodology, system boundary, and data inputs. For example, electricity carbon intensity which could vary by multiple factors will have a significant impact on both $LCCO_2$ and the cost of the CCU product.

The CCU products with the highest economic CO_2 reduction potential vary between countries and scenarios. Figs. 3 and 4 show that in the conventional scenario, the most effective way to reduce CO_2 emissions is to produce gasoline in Canada and sell it in Japan. In the future, the most effective way to reduce emissions is to produce gasoline in Germany and sell it in Japan. Further case studies, including supply chain studies and other large CO_2-emitting countries such as China, are necessary to make a more detailed evaluation of the establishment of such international collaborations. The methodologies in this study can be applied to any country by preparing the economic data shown in Table 2 and Table 2–1 (in SI).

Moreover, economic evaluation in this study is based only on cost analysis which is not reliable for comparing different technological chains. In future studies, income from the sale of electricity must be considered which could change the entire comparison results.

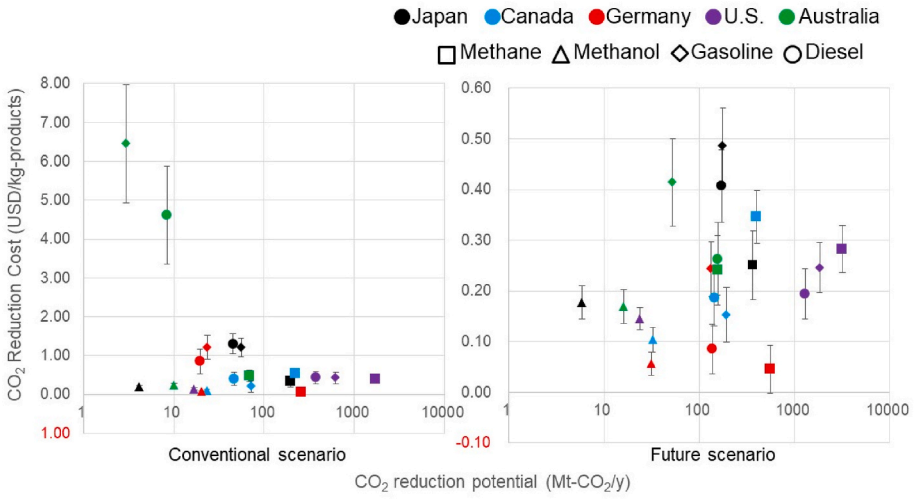

Fig. 5. CO_2 reduction costs and potential annual CO_2 reduction from each synthetic fuel.

The mass deployment of CCUs and CDRs requires large amounts of clean energy; therefore, international collaboration is quintessential. Thus, the results of this study are expected to contribute not only to global CO_2 reduction but also to international cooperation to secure each country's energy demands.

5. Conclusions

This study evaluated the net CO_2 reduction in the atmosphere and their economic viability among the four representative CCU products in five different countries. The overarching objective of this study was to provide a transparent framework with a common dataset generation methodology to determine the countries that have the advantages/disadvantages in synthetic fuel production and sale, considering the local operational parameters such as electricity and existing fuel costs. The novelty of this study is that detailed process modeling was combined with cost and GHG emission estimation for multiple synthetic fuel analyses across various jurisdictions.

Two carbon capture technologies and two conversion technology groups have been assessed for large-scale deployment. This study also assessed the impact of regional characteristics on the introduction of CCU, RE, and CDR technologies. In conclusion, methanol showed the lowest CO_2 reduction cost in the future scenario as its CO_2 reduction potential is low. Diesel fuel showed the next lowest CO_2 reduction cost, which has a high potential, indicating that it is an effective CCU pathway. Gasoline has the largest impact on the technological transition from the conventional to the future scenario. Analysis of regional characteristics showed that Germany and Canada have economic advantages, whereas the U.S. has advantages in terms of local market and export demand. The trends observed for diesel fuel in the present study were similar to those of other studies.

This study has several limitations that should be recognized. As this study focuses on process simulation for key data input generation, detailed process design and economic evaluation of current and future hydrogen production technologies were not included. CCUs and CDRs comprise multiple technologies, including CO_2 capture and conversion technology as well as hydrogen production technology. System optimization integrating these elements must consider the respective supply chains. In addition, the present study estimated the CO_2 recovery from DAC as negative as it reduces atmospheric CO_2. There are no shared guidelines on how to calculate CO_2 reduction from the DAC; thus, drawing such guidelines is necessary. To implement CCU, RE, and CDR technologies on a large scale, efficient management and cost-effective system design are crucial in the future. Moreover, the number of countries for the case study should be increased to enhance international collaboration.

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CRediT authorship contribution statement

Shinichirou Morimoto: Conceptualization, Formal analysis, Funding acquisition, and, Investigation, Methodology, Project administration, Supervision, Validation, Visualization, and, Writing – original draft, Writing – review & editing. **Naomi Kitagawa:** Data curation, Formal analysis. **Farid Bensebaa:** Methodology, Supervision, Validation, Visualization, and, Writing – original draft, Writing – review & editing. **Amit Kumar:** Methodology, Writing – review & editing. **Sho Kataoka:** Resources, and, Software. **Satoshi Taniguchi:** Resources, and, Software.

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

A part of the data is available on request.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2023.136763.

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