# Life Cycle Assessment on Methanol, Dimethyl Ether and Formaldehyde Production for Sustainable Carbon Dioxide **Utilization Options**

Aye Myat Theint Kyaw, Janissara Tangmesang, Varisara Phuaran, Penjit Srinophakun, Anusith Thanapimmetha, Maythee Saisriyoot, and Nutchapon Chiarasumran<sup>\*</sup>

Department of Chemical Engineering, Faculty of Engineering, Kasetsart University, Bangkok, Thailand

Email: ayemyattheint.k@ku.th (A.M.T.K.); jjaniss.09@hotmail.com (J.T.); varisara.mint@gmail.com (V.P.); fengpjs@ku.ac.th (P.S.); fengjrc@ku.ac.th (A.T.); fengmts@ku.ac.th (M.S.); fengnpc@ku.ac.th (N.C.);

\*Corresponding author

Manuscript received February 1, 2024; revised March 27, 2024; accepted April 5, 2024; revised September 27, 2024

*Abstract***—The high risk of climate change impacts is a global concern due to Global Warming Potential (GWP) and greenhouse gas emission. Carbon Capture and Utilization (CCU) is a way to reduce GWP and also produce valuable products from CO2. This research conducted Life Cycle Assessment (LCA) of CO2 utilization for methanol, Dimethyl Ether (DME), and formaldehyde production from CO2 and identified a product that efficiently utilized CO2, providing valuable benefits while minimizing the overall environmental impacts. The research consisted of two parts: a model simulation of production processes and an environmental impact assessment. Model simulation covered the preparation of raw materials CO2 and hydrogen, mixing, reaction, separation of gases, and absorption, with the last stage being distillation to purify the products. All data were analyzed using a model of the processes in the Aspen Plus V12 software. After that, the mass balance and net energy were employed for the environmental impact assessment using LCA based on a functional unit of 1,000 kg CO2 feedstock and gate-to-gate approach. The results appeared that formaldehyde created the highest impacts, while DME has the lowest for all five impacts based on the same functional unit. It was also found that the impacts were mainly caused from the source of energy used in the process, which was produced from non-renewable energy source in Thailand. It can be concluded that CCU has the potential to help achieve net-zero targets and carbon neutrality, but the vital part is the energy source that should be from renewable energy.** 

*Keywords***—carbon dioxide utilization, methanol, dimethyl ether, formaldehyde, process simulation, life cycle assessment** 

## I. INTRODUCTION

Global Warming Potential (GWP) is becoming more severe, leading to widespread environmental impacts, with the main cause being the Greenhouse Gas (GHG) effect.  $CO<sub>2</sub>$  is a major source of the GHG effect. At present, all countries are seeking to develop technologies to reduce CO<sup>2</sup> emissions, mitigate the root cause of GWP, and remediate the environmental impacts. Recently, the record of  $CO<sub>2</sub>$ concentration has reached a high in the earth's atmosphere and its concentration is increasing annually. According to the United States Government Accountability Office (GAO) report in 2022, the average  $2021 \text{ CO}_2$  concentration was 12.4 percent higher than in 2000. Carbon Capture, Utilization, and Storage (CCUS) is a beneficial technology being explored to obtain a valuable product from  $CO<sub>2</sub>$ emissions. CCUS technology involves two main steps: Carbon Capture and Storage (CCS), which focuses on capturing and storing CO2, while the second step involves either storing or utilizing the captured  $CO<sub>2</sub>$ , called Carbon Capture And Utilization (CCU) [1]. In addition, CCUS can be applied to improve the circular economy and sustainable development because the waste  $CO<sub>2</sub>$  can substitute for existing  $CO<sub>2</sub>$  as a raw material and be recycled to use as feedstock for producing a valuable product  $[2]$ . The CO<sub>2</sub> can be converted into valuable products, such as methanol, to capture the highest possible value. Methanol is among the chemical commodities imported in Thailand, and it is the precursor for producing other chemicals, such as Dimethyl Ether (DME) and formaldehyde. DME is an eco-chemical compound and alternative fuel source being non-toxic, biodegradable, colorless, and non-corrosive, as well as not containing Particulate Matter (PM) and Sulfur Oxides (SOx) [3]. Likewise, Formaldehyde (FM) is an important chemical for many industries in the production and construction of building materials, textiles, sterilization products, plastics, and cosmetics [4]. Methanol production uses  $CO<sub>2</sub>$  as the main feedstock; however, it needs other chemicals and energy that create adverse environmental impacts during their production process. Furthermore, the DME and formaldehyde production processes use methanol as a main feedstock in addition to other chemicals and energy that can create environmental impacts [4–6]. Consequently, there is a need to assess the environmental impact of the production process. LCA is one of the best environmental performance tools that have been widely adopted for environmental impact assessment, based on the standardized approach specified in the ISO 14040 standards. LCA can analyze, compare, and evaluate the largest hotspot of environmental impact in every step of the production process  $[7]$ .  $CO<sub>2</sub>$  is currently being used or converted into valuable products which has over 230 million metric tons annually [1]. CCUS plays a critical role in reducing  $CO<sub>2</sub>$  levels in the atmosphere. CCUS can mitigate and temporarily store the  $CO<sub>2</sub>$  emission to the atmosphere, although electricity, hydrogen supply chain, and heavy metal contamination were the highest burdens of the CCU implementation process [8].

Many researchers analyzed the environmental burden of methanol and DME production processes, although FM from carbon utilization has not been many comprehensive studies. Most research assumed that electricity came from renewable sources and neglected the consumption of electricity from the country's electricity grid mix or nonrenewable energy sources. The research would like to fill a

gap in the environmental impact analysis of valuable products of CCUS technology by using the Thailand electricity grid mix. The current research aim was to identify a product that could efficiently utilize  $CO<sub>2</sub>$ , providing valuable benefits while minimizing overall environmental impacts.

## II. LITERATURE REVIEW

# *A. Carbon Capture, Utilization and Storage (CCUS) Technologies*

characterized by the concentrated stream of  $CO<sub>2</sub>$  from fuel combustion and the implementation of point sources. The carbon capture categories are post-combustion, precombustion, oxyfuel combustion, and direct air capture. Post-combustion captured the  $CO<sub>2</sub>$  from the process flue stream after the combustion processes which has CO<sup>2</sup> concentration between 17% to 70%, and typical pressure about 1 bar. Pre-combustion is separated the  $CO<sub>2</sub>$  from syngas before the combustion which has  $CO<sub>2</sub>$  concentration between 15% to 60%, and typical pressure between 14 bar to 70 bar. Oxyfuel-combustion involved the use of pure oxygen ( $>95\%$ ) instead of air for combustion which has  $CO<sub>2</sub>$ concentration between 3% to 20%, and typical pressure about 1 bar. Direct air capture captured the  $CO<sub>2</sub>$  from the ambient air for  $CO<sub>2</sub>$  concentration up to  $0.04\%$  and typical pressure about 1 bar [9]. The carbon captured technologies are based on  $CO<sub>2</sub>$  separation such as absorption, adsorption, membranes, cryogenic distillation, calcium looping and chemical looping [10]. The carbon capture and utilization  $(CCU)$  converted the captured  $CO<sub>2</sub>$  from a product by using the conversion pathways such as mineral carbonation, hydrogenation, electrochemistry, co-polymerization, and microbial conversion [1]. Carbon capture, storage and utilization (CCUS) is ready for wider deployment in carbon emissions reduction pathways although its application is still challenging and limited due to constraints in the technology readiness level, economic feasibility, and commercial plant establishment. The captured CO<sub>2</sub> must have high purity and be readily available to the market for use in the industrial process. The captured CO2 process should be applied to minimize environmental impacts and maximize the benefits and value of  $CO<sub>2</sub>$  [1]. Environmental impacts of methanol, DME, and formaldehyde based on CCUS technology. The Carbon Capture and Storage (CCS) technology is

Methanol and DME from CO<sub>2</sub> hydrogenation reduced the GHG emissions about 82–86% and reduced fossil fuel depletion by 82–91% compared to conventional petroleumbased fuels [4]. A methanol production process that used CO2 as an energy input reduced the carbon emissions by 1.5 kgCO2 per kilogram of methanol production [5]. Comparing the production of DME and methanol based on the same feedstock, the DME impacts were higher than for methanol. The largest environmental hotspot was the fuel production process of the DME process [8]. The significant environmental impacts of methanol, DME, and FM production were GWP, ecotoxicity, human non-carcinogenic toxicity and resource scarcity associated with major pollutants of production and raw material extraction processes such as SO2, NOx, CO2, CH4, N2O emissions and particulate matter [11, 12].

#### III. MATERIALS AND METHODS

The research was separated into two parts: 1) model simulation; and 2) environmental impact assessment of products. The model simulation used the Aspen Plus V12 software to measure the mass balance and molecular balance of the inputs and outputs in all steps of the methanol, DME, and formaldehyde production processes based on CO2. Then, the data from modeling was used in the life cycle inventory (LCI) phase, with the evaluation of environmental impacts based on the LCA (gate-to-gate) approach and system boundaries, as shown in Fig. 1.



Fig. 1. System boundaries of methanol, dimethyl ether, and formaldehyde production from CO<sub>2</sub>.

## *B. Process Design and Modelling*

#### *1) Model of methanol production process from CO<sup>2</sup>*

Fig. 2 shows the methanol production process from  $CO<sub>2</sub>$ that can be divided into:

- Raw materials preheat, where the main raw material  $(CO<sub>2</sub>)$ gas) from an external source was heated 300ºC and pressurized to 50 bar.
- Reaction, with four reactions occurred in a plug flow reactor in Eqs.  $(1)–(4)$  [5]:

1)Reverse Water Gas Shift reaction

$$
CO_2 + H_2 \leftrightarrow CO + H_2O \tag{1}
$$

2)CO2 Hydrogenation

$$
CO_2 + 3H_2 \rightarrow CH_3OH + H_2O \tag{2}
$$

3) Methanol Synthesis

$$
CO + 2H_2 \rightarrow CH_3OH
$$
 (3)

4) Steam Methane Reforming

$$
CO + 3H_2 \rightarrow CH_4 + H_2O \tag{4}
$$

- Separation, where the light gases were separated in a twophase separator from mixed with methanol, water and a tiny amount of  $CO<sub>2</sub>$ . The light gases were recycled back into the production process.
- Distillation was the final step involving separation of the water from the methanol to achieve high purity methanol (up to 99%).

From this production process, light gases from the purge 1 and purge 2 steams entered the next combustion process, where they combusted with air at flow rates of 5,700 kmol/hr and 1,500 kmol/hr, respectively, in a stoichiometric reactor (RStoic) at 200°C and a pressure of 1 bar.

# *2) Model of DME production process from methanol*

Fig. 3 shows the process of producing DME from methanol, beginning with the raw material preparation process. Methanol was mixed with the methanol obtained from recycle stream resulting in a mix of methane  $(CH<sub>4</sub>)$ ,  $CO<sub>2</sub>$ , carbon monoxide (CO), and water (H<sub>2</sub>O). The gas temperature was increased using the heat exchanger (HX) and the heater (HT1) before entering the reaction process in a plug flow reactor (Rplug), where the following reaction occurred in Eq. (5):

$$
2CH3OH \rightarrow CH3OCH3 + H2O
$$
 (5)

Acid zeolite catalyst was used to accelerate the reaction and then the temperature of the resulting products (DME, water, methanol) from the reactor (PFR) were reduced using a HX and a chiller. Next, the steams entered the distillation phase to separate the DME, methanol and water using the first distillation column. First, the DME was separated, and then the water and methanol were separated using the second distillation column. After that the methanol reentered the recycle stream because a large volume of methanol mixed with water remained that was separated using the third distillation column to obtain purified methanol. The purified methanol could be reused in the recycling stream, completing the production process. Purified methanol was obtained as a co-product, with DME as the main product.

## *3) Model of formaldehyde production process from methanol*

Fig. 4 shows the model of the formaldehyde production process from methanol using silver as a catalyst. First, the raw material preparation used methanol at a flow rate of 990 kmol/hr and air at a flow rate of 421.77 kmol/h. The feedstock stream temperature was adjusted to the desired level before entering the mixing process between the two raw materials. Next, the temperature of the materials was reduced before entering the reaction process in Rplug. In the reactor, the methanol and air were converted to formaldehyde gas at 200°C. Two reactions occurred in Eq Eqs. (6, 7):

$$
CH3OH + 1/2 O2 \rightarrow CH2O + H2O
$$
 (6)

$$
CH3OH \rightarrow CH2O+H2
$$
 (7)

The temperatures of the products from PFR were reduced using a chiller before entering the process to separate the hydrogen gas and nitrogen gas from methanol, water, and

formaldehyde. In addition, the methanol was separated in a 2-phase separator. Then, the temperature was increased before entering the formaldehyde gas-absorbing process using another absorber, where the formaldehyde gas was converted into a formaldehyde solution. The solution entered the first distillation column (DST1) for the removal of the remaining methanol in the formaldehyde solution. Next, the solution entered the second distillation column for additional purification and removal of the remaining methanol to get the desired formaldehyde percentage. The final formaldehyde product had 60% purity. The model simulated the methanol obtained from the  $CO<sub>2</sub>$  off-gas stream being combusted in the RStoic reactor with the air at a flow rate of 1,200 kmol/hr, an operating temperature of 200°C, and a pressure of 1 bar.

#### *C. Life Cycle Assessment of Product*

The goal was to compare the environmental impacts of the methanol, DME, and formaldehyde production processes using the same functional  $CO<sub>2</sub>$  feedstock unit of 1,000 kg CO2. The system boundary was restricted to the production process of methanol, DME, and formaldehyde that excluded the construction, distribution, transportation, and use of the feedstock. The main CO<sub>2</sub> feedstock was assumed to have come from an external facility. In the LCI phase of LCA, the mass balance and energy consumption data were obtained from the Aspen Plus software. For life cycle impact assessment, ReCiPe is a fast method for conducts life cycle impact assessment which can provide the global scale in line with the global nature of many products life cycle. Furthermore, the hierarchies perspective is based on scientific consensus and it regards the time frame and the reasonable or probable impact mechanisms [13]. The environmental impact assessment was analyzed using the ReCiPe 2016 v1.04 midpoint, hierarchical via SimaPro software v.9.1.1.1 and Ecoinvent 3.6 as database, with gateto-gate approach.

#### IV. RESULT AND DISCUSSION

## *A. Model Simulation*

Table 1 shows the materials balance for the methanal, DME, and formaldehyde production model in the model simulation using the Aspen plus v12 software.



Fig. 2. Generalized model of methanol production proces from CO2.





Fig. 4. Generalized model of formaldehyde production process from methanol.





# *B. Life Cycle Assessment of Methanol, DME and Formaldehyde*

Table 2 shows that environmental impacts from methanol, DME and formaldehyde production process as same feedstock  $CO<sub>2</sub>$  1,000 kg for each product by gate-to-gate approach and the electricity was considered Thailand Electricity mix due to plant location in Thailand. The environmental impacts were focused on Freshwater ecotoxicity, Marine ecotoxicity, Human carcinogenic toxicity and Human non-carcinogenic toxicity caused by the highest impacts in normalization constants of ReCiPe 2016. Moreover, GWP is currently the highest global attention, the leading impacts and highest challenges in global environmental problems that is focused on this study followed by net zero emission 2050 [14].





DCB: Dichlorobenzene

## *1) Global warming potential*

Fig 5 shows that for the same functional unit of feedstock  $(1,000 \text{ kg } CO<sub>2</sub>)$ , the formaldehyde production process had the highest impacts on GWP  $(3,344 \text{ kgCO}_2)$ , followed by DME process  $(1,893 \text{ kgCO}_2 \text{eq.})$ , and methanol  $(1,797 \text{ deg})$  $kgCO<sub>2</sub>$ eq.), respectively. However, the products and coproducts from table 1 were also counted as benefit to be deducted from the GWP impact as shown in Fig 5. In case of methanol process, the main product (414.94 kg methanol) could be offset by  $282 \text{ kgCO}_2$ eq. For DME process, methanol (32.83 kg) and DME (269.54 kg) products could be offset for 405 kgCO<sub>2</sub>eq. Finally, methanol (298.6 kg) and formaldehyde (77.76 kg) products could save  $266 \text{ kgCO}_2$ eq. in the case of formaldehyde process. As the result, the total impacts on GWP for methanol, DME and formaldehyde process became  $1,515$  1,488 and  $3,077$  kgCO<sub>2</sub>eq. respectively, as shown in Fig. 5. This approach was also applied to all other LCA indicators in this work. The GWP from formaldehyde case mainly came from energy consumption in the production process of up to 2,567  $kgCO<sub>2</sub>$ eq which was directly influenced by Thailand's electricity mix due to non-renewable energy's share of 77% in the total electricity mix [15].



Fig. 5. Global warming potential of methanol, DME, and formaldehyde from  $1000 \text{ kg CO}_2$  feedstock.

*2) Freshwater ecotoxicity* 



In Fig. 6, formaldehyde was the leading product in terms of freshwater ecotoxicity, with a net impact of (52 kg1,4- DCB), followed by methanol (11 kg1,4-DCB), and (DME 8) kg1,4-DCB), respectively. The result illustrated that the utilize methanol from  $CO<sub>2</sub>$  reduced the impact 23 kg1,4-DCB for methanol, utilize DME and methanol from  $CO<sub>2</sub>$ could save 28 kg1,4-DCB for DME and utilize formaldehyde and methanol from  $CO<sub>2</sub>$  could offset 21 kg1,4-DCB for formaldehyde. Electricity was the main contributor to the freshwater ecotoxicity impact, accounting

for 48% of the impact of methanol, 82% of DME, and 91% of FM, respectively. Moreover, the production processes of each product discharge water containing contaminants such as zinc, nickel, copper, and vanadium, which has led to toxicity in freshwater bodies.

#### *3) Marine ecotoxicity*

From Fig. 7, formaldehyde had the highest impact in the marine ecotoxicity, with a net impact of  $(70 \text{ kg1,4-DCB})$ , followed by methanol (15 kg1,4-DCB), and DME (12 kg 1,4-DCB), respectively. The utilize methanol from CO<sup>2</sup> could offset 29 kg1,4-DCB for methanol, utilize DME and methanol from  $CO<sub>2</sub>$  could save 36 kg1,4-DCB for DME and utilize formaldehyde and methanol from  $CO<sub>2</sub>$  could offset 27 kg1,4-DCB for formaldehyde. The result indicated that electricity was the primary contributor to marine ecotoxicity, accounting for 48% of methanol, 47% of DME, and 71% of FM, respectively. The main contamination of methanol, DME, and FM production processes were zinc, nickel, copper, and vanadium, which was the main creator of marine ecotoxicity.



Fig. 7. Marine ecotoxicity of methanol, DME, and formaldehyde by  $CO<sub>2</sub>$ 1000 kg.

#### *4) Human carcinogenic toxicity*

From Fig. 8, formaldehyde had the highest impact for human carcinogenic toxicity, producing 100 kg1,4-DCB, while utilizing methanol and formaldehyde from  $CO<sub>2</sub>$  could save 14 kg1,4-DCB that reduced the impact to 86 kg1,4- DCB. DME produced 48 kg1,4-DCB although utilize methanol and DME from CO<sub>2</sub> could offset 12 kg1,4-DCB, so that the net impact was 36 kg1,4-DCB, with methanol having the lowest impact of 44 kg1,4-DCB while utilize methanol from  $CO<sub>2</sub>$  could save 16 kg1,4-DCB that reduced the impact to 28 kg1,4-DCB.





#### *5) Human non-Carcinogenic Toxicity*

From Fig. 9, the net impact of formaldehyde was still the

highest, being 2,074 kg1,4-DCB, while methanol and formaldehyde from  $CO<sub>2</sub>$  utilization saved 608 kg1,4-DCB, reducing the impact to 1,466 kg1,4-DCB. Likewise, DME produced 988 kg1,4-DCB although methanol and DME from  $CO<sub>2</sub>$  utilization could offset 437 kg1,4-DCB, reducing the impact to 551 kg1,4-DCB. Methanol production had the lowest impact of 914 kg1,4-DCB while utilize methanol from CO2 could save 751 kg1,4-DCB, so that the net impact was 163 kg1,4-DCB. The impact of formaldehyde mainly came from the energy used in the production process.



Fig. 9. Human non-carcinogenic toxicity of methanol, DME, and formaldehyde by CO<sub>2</sub> 1000 kg.

# *C. Sensitivity Analysis of Global Warming Potential from Methanol, DME and Formaldehyde Production*

The research considered ways to reduce the environmental impacts by focusing on emission reduction, GWP reduction, and CCU implementation based on the following sensitivity analyses:

- 1) Compare the GWP of methanol, DME, and formaldehyde based on just material consumption, neglecting energy consumption.
- 2) Compare the GWP of methanol, DME, and formaldehyde based on various percentage of renewable energy usage.

# *1) Compare global warming potential of methanol, DME, and formaldehyde based on just material consumption*

From Fig. 10, formaldehyde produced 777 kgCO<sub>2</sub>eq. while utilize methanol and formaldehyde from  $CO<sub>2</sub>$  could offset 266 kgCO<sub>2</sub>eq. that reduce the GWP to 510 kgCO<sub>2</sub>eq. Methanol produced  $766 \text{ kgCO}_2$ eq. although utilize methanol from  $CO<sub>2</sub>$  could save 282 kg $CO<sub>2</sub>$ eq. that reduce GWP to 484 kgCO<sub>2</sub>eq. DME produced 762 kgCO<sub>2</sub>eq., while methanol and DME from  $CO<sub>2</sub>$  utilization could offset 405 kgCO<sub>2</sub>eq that reduce the GWP impact to  $357 \text{ kgCO}_2$ eq. The GWP of

formaldehyde came from hydrogen consumption (360  $kgCO<sub>2</sub>$ eq.), and GWP of methanol mainly came from hydrogen consumption (363 kgCO<sub>2</sub>eq.).



Fig. 10. Global warming potential of methanol, DME, and formaldehyde by CO2 feedstock 1000 kg (consider just material consumption).

# *2) Compare global warming potential of methanol, DME, and formaldehyde by renewable energy usage percentage*

A range in renewable energy percentages (10%, 20%, 30%, 40%, 50% and 100%) was assumed for the GWP evaluation process. Renewable energy at 10% represents 10% renewable energy (Electricity, biomass, at power plant, in the Ecoinvent database), with the balance of 90% coming from the Thailand country electricity mix. From Fig. 11 and Table 3, the renewable energy percentage dominated the GWP impact reduction process. These results confirmed that environmentally friendly and CCUS implementation should use renewable energy instead of non-renewable energy sources. Furthermore, the methanol utilized from  $CO<sub>2</sub>$  could offset the environmental impacts, indicating that  $CO<sub>2</sub>$ utilization was beneficial in environmental impact reduction and emission reduction pathways.



Fig. 11. Global warming potential of methanol, DME, and formaldehyde by CO2 feedstock 1000 kg (Renewable energy percentage variable).

Table 3. Global warming potential of product based on variable renewable energy percentage for energy consumption (kg CO<sub>2</sub>eq./1,000 kg CO<sub>2</sub> feedstock)

FM	DME	Methanol	<b>Renewable Energy %</b>		
2,837	1,382	1,418	$10\%$ RE <sup>a</sup> + 90% EM <sup>b</sup>		
2,596	1,276	1,322	20% RE <sup>a</sup> + 80% electricity mix <sup>b</sup>		
2,356	1,170	1,225	$30\%$ RE <sup>a</sup> + 70% electricity mix <sup>b</sup>		
2,115	.064	1,128	40% RE <sup>a</sup> + 60% electricity mix <sup>b</sup>		
1,875	958	1,032	50% RE <sup>a</sup> + 50% electricity mix <sup>b</sup>		
672	428	549	$100\%$ RE <sup>a</sup> + 0% electricity mix <sup>b</sup>		

 $a =$  Renewable energy,  $b =$  Thailand country electricity mix

## *3) Environmental impacts compare between the current study and existing literature*

The direct comparison on the LCA results with other works were slightly difficult according to the different LCA approaches, database used, simulation methods, and the functional units. Mahabir *et al.* [11] investigated the megamethanol production from carbon utilization which used their electricity sources, a functional unit of 1 ton methanol production, cradle-to-gate approach, and SimaPro, ReCiPe 2016 (H). Puhar *et al.* [12] determined the environmental impacts of formaldehyde production via methanol stream reforming by using OpenLCA software which is cradle-togate approach and functional unit as production of 1 kg product. Rigamonti *et al.* [16] considered the functional unit as 1,000 kg treatment of process gases, environmental impact was evaluated by the Environmental Footprint (EF) method. To ensure comparability, all results were standardized to the same functional unit for each study, except for variations in the LCA approach and simulation method. Table 4 shows the environmental impact comparison between the existing literature and the current study.

Table 4. Global warming potential, Freshwater and marine water ecotoxicity, human carcinogenic and non-carcinogenic toxicity comparison between the current study and existing literature

	Mahabir <i>et</i> al. [1]	Puhar et <i>al.</i> [12]	Rigamonti & Brivio [16]	<b>Base case</b>	100% <b>RE</b>		
Global warming potential (GWP) (kgCO2eq/kg product)							
Methanol	0.28		0.49	3.65	1.32		
<b>DME</b>				5.52	1.59		
Formaldehyde		1.37		39.58	8.64		
Freshwater ecotoxicity (kg1,4-DCB/kg product)							
Methanol			$-0.19$	0.001	$-0.0002$		
<b>DME</b>				0.002	$-0.0002$		
Formaldehyde				0.020	$-0.0008$		
Marine water ecotoxicity (kg1,4-DCB/kg product)							
Methanol			$-0.01$	0.036	$-0.0493$		
<b>DME</b>				0.045	$-0.0993$		
Formaldehyde				0.901	$-0.2364$		
Human carcinogenic toxicity (kg1,4-DCB/kg product)							
Methanol			$-2.68E-09$	0.068	$-0.0128$		
<b>DME</b>				0.135	$-0.0007$		
Formaldehyde		0.15		1.112	0.0409		
Human non-carcinogenic toxicity (kg1,4-DCB/kg product)							
Methanol	0.31	0.39	$-2.92E-08$	0.393	$-1.4817$		
<b>DME</b>		0.15		2.043	$-1.1240$		
Formaldehyde				18.858	$-6.0607$		

From Figs. 12, 13, and 14, comparison between each impact indicator and the study (base case), the study's results are much higher than the other studies because other studies considered the electricity came from 100% renewable energy (hydro, wind, and solar) and Mahabir *et al.*  (2021) used their electricity that could reduce their environmental impacts. In this result, the consideration focused on actual conditions that examined the impacts of valuable product production from carbon utilization that energy source was directly used in the Thailand grid mix. In comparison between each impact indicator, the study's result (100% renewable energy usage) was slightly higher than other studies due to the electricity source coming from biomass power plants, and the large amount of energy consumption of each production process. Particularly, plant efficiency, material and energy consumption, and energy sources are the main drivers for reducing emissions and developing net zero emissions and carbon neutrality [17]. The research revealed that CCUS can play a crucial role in achieving the net zero target and carbon neutrality, however, CCU commercial plant needs technology development and thorough assessments, including economic, environmental, and technological considerations.



Fig. 12. Global warming potential results comparison with existing literature



Fig. 13. Freshwater ecotoxicity and marine ecotoxicity results comparison with existing literature.



Fig. 14. Human carcinogenic and Human non-carcinogenic toxicity results comparison with existing literature.

## V. CONCLUSION

The developed model achieved 99% methanol production, 99% dimethyl ether production and 60% formaldehyde production using  $CO<sub>2</sub>$  as the main raw material. The feedstock  $CO<sub>2</sub>$  gas at 1,000 kg/hr yielded 414.94 kg/hr of methanol, 269.54 kg/hr of dimethyl ether and 77.76 kg/hr of formaldehyde.

The life cycle assessment of the production processes for methanol, dimethyl ether and formaldehyde from  $CO<sub>2</sub>$  gas demonstrated that formaldehyde production from 1,000 kg CO2 as feedstock produced the highest environmental impacts in all five indicators, whereas DME production from the same feedstock caused the least environmental <sup>1</sup> impacts on all studied impacts categories. The results were directly related to the energy consumption since the formaldehyde consumed the largest amount of electricity according to the purification process, especially from the distillation units.

This result was confirmed by the first part of the

sensitivity analysis, which indicated that all products could save net GWP up to  $1,000-2,500$  kgCO<sub>2</sub>eq. based on  $1,000$ kg CO<sub>2</sub> feedstock when neglecting energy consumption. Therefore, the second part of the sensitivity analysis proposed to increase the percentage of renewable energy from biomass used as energy source in Thailand. It was found that the higher percentage of renewable energy usage, the lower net GWP obtained. These results can lead to the summary that the environmental-friendly product production with CCU implementation can help reducing the environmental impacts, especially on GWP, but it need to employ renewable energy as energy source. Further technology development and thorough assessments, particularly economic, environmental, and technological considerations together, are still necessary.

#### CONFLICT OF INTEREST

The authors declare no conflict of interest.

## AUTHOR CONTRIBUTIONS

Evaluation of the environmental impacts assessment (LCA), writing, and preparation were performed by Kyaw. Model simulation was applied by Tangmesang and Phuarun. Srinophakun, Thanapimmetha, and Saisriyoot provided supplementary advice. Chiarasumran conducted the conceptualization, supervision, resources management, validation, editing and writing manuscript. All authors approved the final version.

## FUNDING

This research was funded by Faculty of Engineering, Kasetsart University, Bangkok, Thailand.

#### ACKNOWLEDGMENT

The authors wish to thank the Department of Chemical Engineering, Faculty of Engineering, Kasetsart University for providing the research facilities.

#### **REFERENCES**

- [1] United States Government Accountability Office, *Technology Assessment Decarbonization Status, Challenges, and Policy Options for Carbon Capture*, Utilization and Storage, U.S, 2022, pp. 51–52.
- [2] National Academies of Sciences, *Engineering and Medicine, Gaseous Carbon Waste Streams Utilization: Status and Research Needs*. Washington. DC.: The National Academies Press, 2019, ch. 3, p. 40.
- [3] G. Leonzio, "State of art and perspectives about the production of methanol, dimethyl ether and syngas by carbon dioxide hydrogenation," *Journal of CO2 utilization*, vol. 27, pp. 326-354, 2018.<br>[4] I.Ruwida,
- "Optimization of formaldehyde production from methanol," MS. thesis, Dept. of Chemical Eng and Applied Chemistry, Atilim Univ, Turkey, 2018.
- [5] Cordero-Lanzac et al., "A techno-economic and life cycle assessment for the production of green methanol from carbon dioxide: catalyst and process bottlenecks," *Journal of Energy Chemistry*, vol. 68 pp 255-266, 2022.
- [6] Asadi and Farahani, "Optimization of dimethyl ether production process based on sustainability criteria using a homotopy continuation method," *Computer and Chemical Engineering*, vol. 115, pp. 161– 178, 2018.
- [7] M. Finkbeiner, "The international standards as the constitution of life cycle assessment: The ISO 14040 series and its offspring," *Springer*, pp. 85–106, 2014.
- [8] Matzen and Demirel, "Methanol and dimethyl ether from renewable hydrogen and carbon dioxide: Alternative fuels production and lifecycle assessment," *Journal of Cleaner Production*, vol. 139, pp. 1068–1077, 2016.
- [9] A. Allangawi *et al*, "Carbon Capture Materials in Post-Combustion: Adsorption and Absorption-Based Processes," *Journal of Carbon Research*, vol. 9, no. 1, p. 1, 2023.
- [10] B. Dziejarski, R. Krzyżyńska, and K. Andersson, "Current status of carbon capture, utilization, and storage technologies in the global economy: A survey of technical assessment," *Fuel,* Elsevier, vol. 342, p. 127776, 2023.
- [11] J. Mahabir *et al.*, "What is required for resource-circular CO<sub>2</sub> utilization within Mega-Methanol (MM) production?" *Journal of CO<sup>2</sup> Utilization*, vol. 45, p. 101451, 2021.
- [12] J. Puhar *et al.*, "Reduction of cost, energy and emissions of the formalin production process via methane steam reforming," *Systems*, vol. 9, 2021. DOI: 10.3390/systems9010005
- [13] M. A. Huijbregts *et al.*, "ReCiPe2016: a harmonized life cycle impact assessment method at midpoint and endpoint level," *The International Journal of Life Cycle Assessment*, vol. 22, pp. 138-147, 2017.
- [14] International Energy Agency, *Global Energy and Climate Model Documentation-2023*, France, 2023.
- [15] International Trade Administration, *Thailand Country Commercial Guide*, 2024.
- [16] L. Rigamonti and E. Brivio, "Life cycle assessment of methanol production by a carbon capture and utilization technology applied to steel mill gases," *International Journal of Greenhouse Gas Control*, vol. 115, p. 103616, 2022.
- [17] Ministry of Natural Resources and Environment, "Thailand's longterm low greenhouse gas emission development strategy (revised version)," *Office of Natural Resources and Environmental Policy and Planning*, Bangkok, 2022.

Copyright © 2024 by the authors. This is an open access article distributed under the Creative Commons Attribution License which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited  $(CC BY 4.0)$ .