

Life cycle assessment on alternative hydrogen production for greener technology

Saw Yu Nwe¹, Aye Myat Theint Kyaw², Penjit Srinophakun², Anusith Thanapimmetha², Maythee Saisriyoot², and Nutchapon Chiarasumran^{2*}

¹ Master of Engineering Program in Sustainable Energy and Resources Engineering, Faculty of Engineering, Kasetsart University, Bangkok 10900, Thailand

² Department of Chemical Engineering, Faculty of Engineering, Kasetsart University, Bangkok 10900, Thailand

ABSTRACT

***Corresponding author:**
Nutchapon Chiarasumran
fengnpc@ku.ac.th

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Hydrogen has been recognized as an alternative energy carrier and one of the future fuels in the global transition to clean energy. Hydrogen production with net-zero-carbon emissions has been developed because steam methane reforming, as a conventional means of hydrogen production, involves carbon dioxide emissions. This study carried out a life cycle impact assessment of conventional hydrogen production and compared the outcomes to two other zero-carbon emission technologies: proton exchange membrane electrolysis and methane pyrolysis. Main inventory data collection was based on process simulation and impact assessment using the Aspen plus V12.0 and Simapro 9.1 software packages, respectively. The system boundary involved gate-to-gate consideration with electricity supply from Thailand's grid mix and a functional unit of 1,000 kg/h hydrogen production. The ReCiPe 2016 midpoint impact assessment method was used to evaluate five impact categories: global warming potential, freshwater and marine ecotoxicity, and human carcinogenic and non-carcinogenic toxicity. In terms of global warming potential, proton exchange membrane electrolysis was 1.7 times higher than steam methane reforming and 2.3 times higher than methane pyrolysis, since its primary energy is derived from fossil-based electricity. Although methane pyrolysis relied on hydrocarbon feedstock, it had the lowest environmental impact in all five impact categories investigated.

Keywords: hydrogen production; steam methane reforming; proton exchange membrane electrolysis; methane pyrolysis; Aspen Plus; life cycle assessment

1. INTRODUCTION

Climate change and global warming issues have become critical problems in this era. They are mostly caused by anthropogenic greenhouse gas (GHG) emissions from the combustion of fossil fuels such as coal, oil, and natural gas. According to Crippa et al. (2023), global GHG emissions from various sectors were only around 33,268 Mt carbon dioxide (CO₂) equivalent in 1990. However, it had

escalated to 53,786 Mt CO₂ equivalent in 2022. Long-term goals of securing carbon neutrality and net zero GHG emissions have been targeted based on the ambitious collective goals of the Paris Agreement to apply ultimate climate action on a global scale (Delbeke et al., 2019). Thailand is also committed to engage in countering climate change by establishing climate goals to achieve carbon neutrality by 2050 and net zero GHG emission by 2065 (Office of Natural Resources and Environmental Policy and

Planning, 2022). GHG emissions from industrial sources represent the largest share in Thailand, followed by the power, agricultural, and transport sectors (Crippa et al., 2023).

Hydrogen (H₂) is gaining in popularity as a source of clean energy and is considered as an important component in driving the transformation from fossil-based to renewable fuel sources. Annual worldwide hydrogen production is approximately 70 Mt, produced from three sources: natural gas (76%), coal (23%), and other (1%), according to the International Energy Agency (2019). However, currently, most hydrogen is synthesized by steam reforming using natural gas or syngas from coal gasification as feedstock. This conventional technology still emits a large amount of CO₂, which is a by-product from the water-gas shift (WGS) reaction between steam and natural gas or syngas. In tracking targeted climate change mitigation goals, it is important to examine and develop alternative CO₂-emission-free hydrogen production technologies. Water electrolysis using a proton exchange membrane (PEM) is also an attractive carbon-free hydrogen production technology, with a longer life time, lower operating temperature, and a shorter response time compared to alkaline water electrolysis (Wang et al., 2022). Another attractive technology is methane pyrolysis, in which pure methane is thermally decomposed into carbon black and hydrogen at an elevated temperature (Sánchez-Bastardo et al., 2020). The solid carbon black can be further transformed into carbon-based merchandise without any process CO₂ emission during hydrogen production (Osman et al., 2022). Some researchers have conducted life cycle assessment (LCA) to compare the carbon footprint between different hydrogen production technologies, including conventional steam methane reforming, biomass and coal gasification, water electrolysis, metal oxide cycling, methane pyrolysis, and dark fermentation of lignocellulosic biomass. For example, Mehmeti et al. (2018) and Sánchez-Bastardo et al. (2021) found that PEM electrolysis and methane pyrolysis made the lowest contributions to global warming potential (GWP) compared to other technologies. Hydrogen synthesis via water electrolysis and methane pyrolysis are of interest as potential options for comparative assessment of the different carbon footprints of various hydrogen production methods. In addition, it is necessary to investigate these technologies under operational circumstances in Thailand.

This research aimed to compare the environmental impacts of hydrogen production technologies in Thailand by applying conventional steam methane reforming, PEM electrolysis, and thermal methane pyrolysis processes to identify which had the least impacts in terms of global warming, water resource toxicity, and health. In this study, Thailand's grid mix production was proposed as the main source of energy needed for powering the electrical appliances required in the hydrogen production chain. The provision of thermal energy for the process heat was supplied from the combustion of common natural gas.

2. MATERIALS AND METHODS

Three different hydrogen production scenarios were considered in this study to perform a comparative LCA: steam methane reforming (SMR), proton exchange

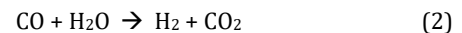
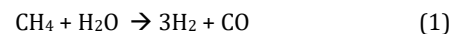
membrane electrolysis (PEM), and methane pyrolysis (MP). The research method was divided into two sections: process simulation and LCA. Foreground data for both material and energy requirements were acquired based on process simulation using the Aspen Plus V12.0 software. Natural gas and water were used as the feedstock for SMR, deionized water for PEM, and pure methane for MP, respectively. The Simapro 9.1 software was used for background data acquisition and evaluation of life cycle impacts for each hydrogen production process. For all scenarios considered, the production rate of H₂ was set at 1,000 kg/h.

2.1 Process simulation

2.1.1 Steam methane reforming

The simulated flowsheet for steam methane reforming is shown in Figure 1. The process concept was referenced from Phan et al. (2022) and the PRMHV2 property method was selected. The composition of the feed natural gas (NG) was referenced from Alizadeh et al. (2020) and is shown in Table 1. Initially, the process validation was performed based on the referenced concept with the molar ratio of feed NG for data accuracy. The validity simulation determined that an NG flow rate of 38.51 kg/h yielded a production rate of 13.18 kg/h of H₂. Therefore, it was verified that an NG feed rate of 2,921 kg/h was necessary for the production of 1,000 kg/h of H₂. The steam-to-carbon ratio was assumed to be 3:1, a value also obtained from Phan et al. (2022). The procedure involved seven primary process points: (1) feed stock preparation by preheating of NG and water; (2) the reforming operation at 909°C, in which the steam methane reforming reaction occurs; (3) a two-step WGS reaction involving high-temperature shift at 350°C and a low-temperature shift at 210°C; (4) the condensate removal process; (5) H₂ separation and purification in the pressure swing adsorption (PSA) unit; (6) H₂ compression to the storage tank; and (7) tail gas combustion with additional air for conversion into flue gas, directly emitted into the ambient air.

Nickel-based and iron-based catalysts are used commercially in the reforming and WGS reactors (Amran et al., 2017). The amounts were estimated of the nickel and iron catalysts for the production of 1,000 kg/h of H₂. Furthermore, it was assumed that instead of being dumped, the fully utilized catalysts would either be recycled or processed in alternative sectors for further usage. Process heat recovery from the flue gas was excluded in this study. The SMR and WGS reactions are represented in Equations (1) and (2) and it was assumed that no reverse reaction occurred during the hydrogen synthesis.



2.1.2 Proton exchange membrane

Both PEM and alkaline water electrolysis (AWE) are commercially matured water electrolysis technologies, while solid oxide electrolysis (SOE) is still in the early stage of development (Wang et al., 2022). AWE holds the largest market share with 61% of the global water electrolysis, while the share of PEM is 31% (Ansari et al., 2022). Nevertheless, PEM is a promising water electrolysis

technology which can improve on AWE in the decarbonization pathway with renewable energy sources since it can function well even under a fluctuating power supply (Kojima et al., 2023).

Deionized water is used for PEM water electrolysis; and Figure 2 shows the process simulation flowsheet. The simulation concept was adapted from Muazzam et al. (2022), in which the PEM cell was designed by a reactor and a separator (SEP1). The selected property method was non-random two-liquids (NRTL). Data validation for PEM was conducted using the stream results derived from the

adapted concept. Usually, the standard operating temperature for the PEM electrolysis process is less than or equal to 80°C (Wang et al., 2022). The flowsheet involves five primary process steps: (1) preheating of deionized water; (2) a PEM stack operating at 80°C; (3) cathode simulation; (4) anode simulation; and (5) H₂ compression to storage. The overall reaction of a water electrolysis process is shown in Equation (3) and only the forward reaction was considered during the simulation.

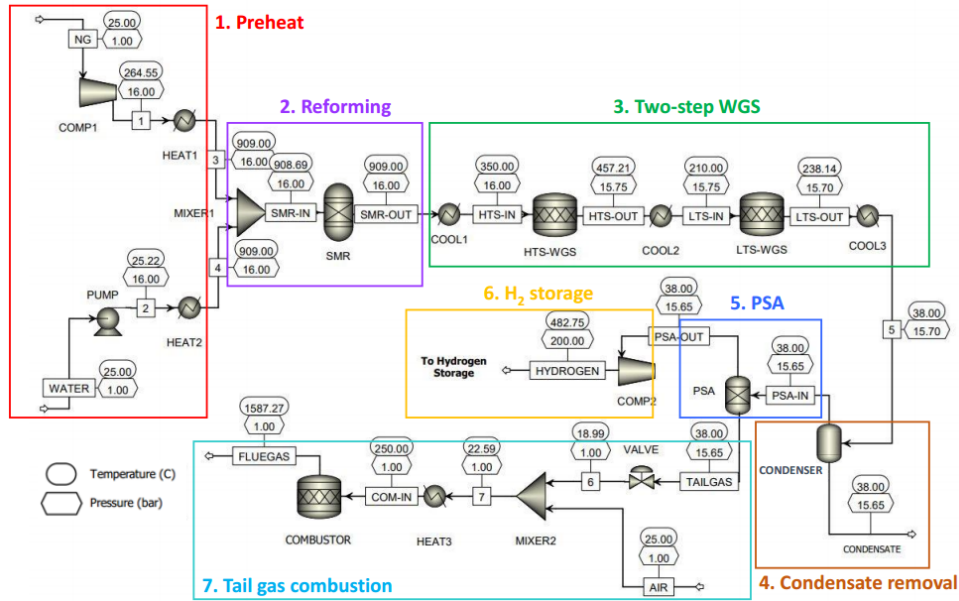


Figure 1. Flowsheet of hydrogen production process based on steam methane reforming with the capacity of 1,000 kg/h H₂

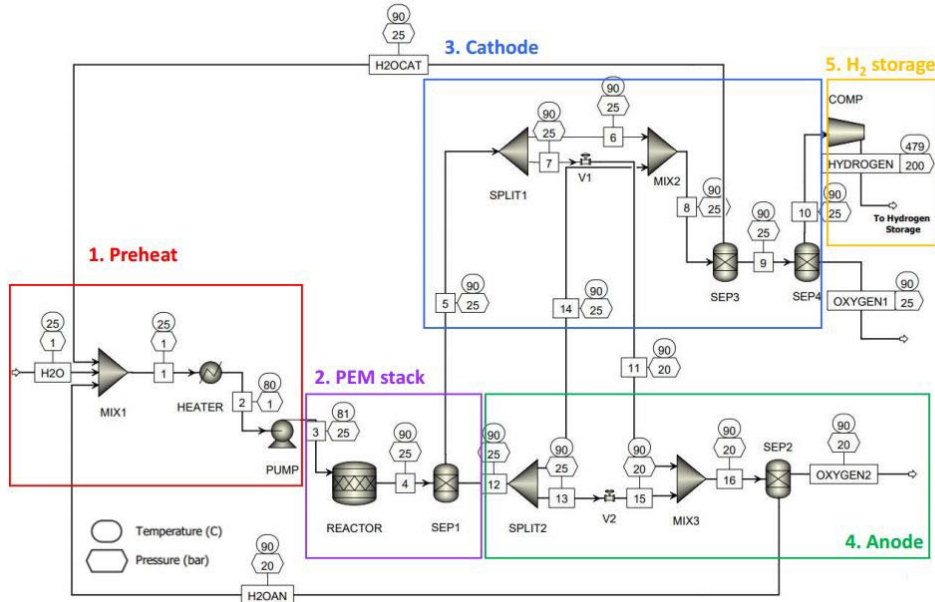


Figure 2. Flowsheet of hydrogen production process based on PEM electrolysis with the capacity of 1,000 kg/h H₂

2.1.3 Methane pyrolysis

Alternative terms for methane pyrolysis are methane cracking or natural gas pyrolysis. However, to avoid the formation of impurities during the process, it is important to feed pure methane, whether it comes from natural gas or biogas. A notable fact regarding methane pyrolysis is that it is entirely free from byproduct CO₂ emission, despite methane being a primary feedstock. Instead, the methane is broken down into hydrogen and solid black carbon. There are two types of methane pyrolysis: thermal and catalytic. In catalytic methane pyrolysis, the quality of the produced solid carbon may vary depending upon the type of catalysts use in the reactor (Noyan et al., 2023). This solid carbon can be further processed into industrial carbon-based materials, thereby enhancing the circular carbon economy. Since it does not produce any GHG, methane pyrolysis is appealing as an intermediary hydrogen production technology in the transition from a fossil-based to a renewable-based energy system.

The optimum temperature ranges are 1,100–1,600°C for thermal pyrolysis and 500–1,000°C for catalytic pyrolysis (Sánchez-Bastardo et al., 2020). Figure 3 shows the process simulation of hydrogen production based on thermal methane pyrolysis. Both the process concept and data validation were obtained from Banu et al. (2022), in which the Peng-Robinson equation of state (PENG-ROB) method was chosen for simulation. The flowsheet involves four primary steps: (1) high pressurization of methane up to 10 bar; (2) a pyrolysis reactor operating at 1,200°C; (3) H₂ separation using PSA; and (4) H₂ compression to storage. In the reactor, the methane pyrolysis reaction occurs according to the following Equation (4); as with SMR and PEM, it was assumed that only the forward reaction occurred during synthesis.

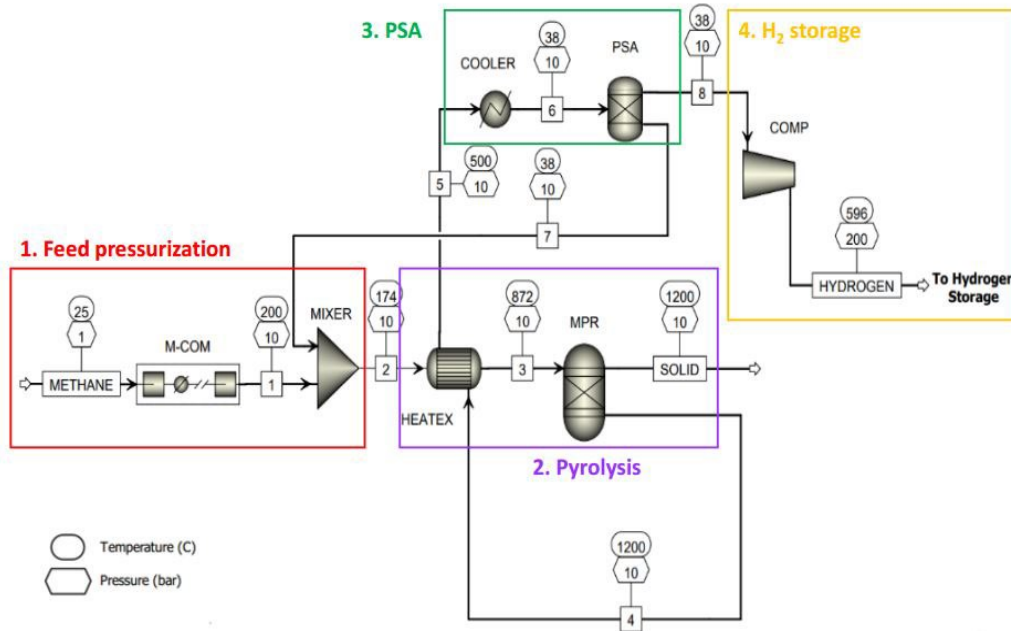


Figure 3. Flowsheet of hydrogen production process based on methane pyrolysis with the capacity of 1,000 kg/h H₂

2.2 Life cycle assessment

2.2.1 Goal and scope

Consistent with climate change mitigation targets in Thailand, the goal of the current research was to conduct a comparative life cycle impact assessment of a conventional hydrogen production technology (SMR) and of process CO₂ free hydrogen production technologies (PEM electrolysis and MP). This work used a gate-to-gate methodology and therefore mainly focused on the impacts associated with the primary mechanisms involved during hydrogen production. The system boundary started from the inputs

of feed stocks, electricity, and energy into the process and extended to the emissions from the hydrogen production units into the ambient air and water. The functional unit was 1,000 kg/h of H₂ production; Figure 4 shows the system boundary of this assessment. According to the comparison of lower heating values provided by The Engineering ToolBox (2003), it was estimated that 1,000 kg/h of H₂ could replace approximately 2,542 kg/h of NG consumption for the generation of 33,300 kWh of grid electricity.

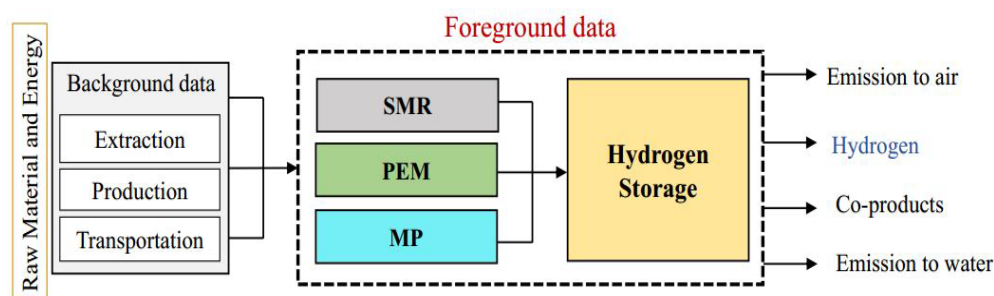


Figure 4. System boundary of LCA for hydrogen production scenarios

For SMR and PEM electrolysis, mass allocation was not taken into account, whereas it was considered in MP. The mass allocation percentages for the target product (H_2) and byproduct (solid carbon black) were 25% and 75%, respectively. The amount of solid carbon produced from 1,000 kg/h of H_2 was 2,980 kg/h, as shown in Table 4, because the carbon black from thermal methane pyrolysis can be further replaced as a carbon-based feedstock for industrial manufacturing processes, thus minimizing fossil depletion from another aspect.

2.2.2 Life cycle inventory analysis

The overall mass and energy balances of each of the H_2 production technologies for the production of 1,000 kg/h of H_2 were collected through process simulation, which could be considered as primary data collection. The ecoinvent database was selected for background data due to the wide range of chemical-related production processes. All scenarios excluded the consideration of process cooling water or cooling refrigerants requirement, since this was considered to remain within the process chain. The thermal energy required for the heaters or reactors was provided from natural gas combustion with any resultant emissions also taken into consideration, as shown in Table 4. The industrial scale electricity demand was obtained from Thailand's grid mix production and was used for the operation of pumps and compressors, where necessary. Table 4 quantifies the main inventory data for the 1,000 kg/h H_2 production, which was based on Aspen Plus simulation results.

2.2.3 Impact assessment

This study focused more on midpoint impact indicators (particularly for identifying GWP) than overall damage at the endpoint level. Therefore, the ReCiPe 2016 midpoint method was selected for conducting a comparative impact assessment of all three hydrogen production scenarios. In addition to global warming, four other prominent midpoint impact categories were considered in this assessment: freshwater ecotoxicity, marine ecotoxicity, and human carcinogenic and non-carcinogenic toxicity.

The CO_2 equivalent (CO_2 eq.) is used to express GWP, since CO_2 makes up the largest component of world greenhouse gas emissions, as well as residing in the Earth's atmosphere for a long period with an average life time of 50–200 years (Dickinson et al., 1996). The development of diseases and the incidence of malnutrition are directly impacted by global warming, thus ultimately harming human health (Huijbregts et al., 2017).

In freshwater, marine, and human beings, the unit of 1, 4-dichlorobenzene-equivalents (1, 4-DCB-eq.) unit is used to indicate the toxicity level. Freshwater and marine ecotoxicity both directly contribute to the extinction and mortality of aquatic species, eventually leading to the destruction of ecosystems (Huijbregts et al., 2017). The potential risk of human cancer and non-cancer diseases rate is monitored by the human carcinogenic and non-carcinogenic toxicity levels, which mainly cause damage to human health at the endpoint (Huijbregts et al., 2017).

3. RESULTS AND DISCUSSION

3.1 Simulation results

Tables 1–3 summarize the simulated mass composition of stream results for the SMR, PEM, and MP scenarios, respectively. Figure 5 shows the overall energy usage, material balances, and emissions from each scenario, according to the process simulation. Based on the simulated results, the inventory data for LCA is summarized in Table 4, except for the required amount of catalysts for SMR that was referenced from Amran et al. (2017). There was no catalyst requirement for hydrogen synthesis in the PEM and MP scenarios. In all cases, natural gas and air were supplied to meet the thermal energy requirement through the combustion process within heaters. Therefore, emissions from the heaters were separately presented from process emissions, in both Table 4 and Figure 5.

For SMR and MP, the electricity requirement accounted for pumps and compressors only. For PEM, electricity consumption was the highest since it was entirely based on splitting deionized water into hydrogen and oxygen via electrolysis, with the majority of electricity consumption in PEM being for the electrolysis process itself. There were no process CO_2 emissions to the air from the PEM and MP scenarios, except from the combustion emissions within heaters. The mass composition of CO_2 in each flue gas stream is depicted in Figure 5. Although there is no separate heater shown in the simulation flow sheet of MP (Figure 3), it was considered that the process heat requirement for the pyrolysis reactor was provided by NG combustion with air inside a separate furnace or heater, based on the concept from Keipi et al. (2018).

In both the SMR and PEM scenarios, no allocation was considered since the focus product was only H_2 . The generated O_2 from PEM was released into the air as emissions with no further utilization, as categorized in Table 4. In the MP scenario, carbon black was produced as

a byproduct simultaneously with hydrogen synthesis. The amount was quite large with the H₂-to-carbon ratio being 1:2.98. This solid carbon could be used as carbon-based

resource in other manufacturing industries. Therefore, mass allocation was considered in MP, unlike in the SMR and PEM scenarios.

Table 1. Mass composition of streams in SMR process simulation

Stream name	Mass flow (kg/h)	Mass composition (%)									
		CH ₄	C ₂ H ₆	C ₃ H ₈	C ₄ H ₁₀	CO ₂	N ₂	H ₂	CO	H ₂ O	O ₂
NG	2,921	62.59	11.44	8.92	13.16	2.54	1.35	-	-	-	-
WATER	8,644	-	-	-	-	-	-	-	-	100	-
1	2,921	62.59	11.44	8.92	13.16	2.54	1.35	-	-	-	-
2	8,644	-	-	-	-	-	-	-	-	100	-
3	2,921	62.59	11.44	8.92	13.16	2.54	1.35	-	-	-	-
4	8,644	-	-	-	-	-	-	-	-	100	-
SMR-IN	11,565	15.82	2.89	2.25	3.32	0.64	0.34	-	-	74.74	-
SMR-OUT	11,565	1.41	-	-	-	18.44	0.34	8.97	29.9	40.94	-
HTS-IN	11,565	1.41	-	-	-	18.44	0.34	8.97	29.9	40.94	-
HTS-OUT	11,565	1.41	-	-	-	53.67	0.34	10.59	7.47	26.52	-
LTS-IN	11,565	1.41	-	-	-	53.67	0.34	10.59	7.47	26.52	-
LTS-OUT	11,565	1.41	-	-	-	62.49	0.34	10.99	1.87	22.9	-
5	11,565	1.41	-	-	-	62.49	0.34	10.99	1.87	22.9	-
CONDENSATE	2,685	0.01	-	-	-	3.9	0.01	0.2	0.05	95.83	-
PSA-IN	8,880	1.83	-	-	-	80.2	0.44	14.25	2.42	0.86	-
TAIL GAS	7,880	2.06	-	-	-	90.37	0.5	3.37	2.73	0.97	-
PSA-OUT	1,000	-	-	-	-	-	-	100	-	-	-
HYDROGEN	1,000	-	-	-	-	-	-	100	-	-	-
AIR	15,551	-	-	-	-	-	77.8	-	-	-	22.2
6	7,880	2.06	-	-	-	90.37	0.5	3.37	2.73	0.97	-
7	23,431	0.78	-	-	-	34.17	48.56	1.28	1.03	0.38	13.8
COM-IN	23,431	0.78	-	-	-	34.17	48.56	1.28	1.03	0.38	13.8
FLUE GAS	23,431	-	-	-	-	37.93	48.56	-	-	13.51	-

Table 2. Mass composition of streams in PEM process simulation

Stream name	Mass flow (kg/h)	Mass composition (%)		
		H ₂ O	O ₂	H ₂
H ₂ O	9,030	100	-	-
1	9,039	100	-	-
2	9,039	100	-	-
3	9,039	100	-	-
4	9,039	1	88	11
5	1,010	-	-	100
6	1,000	-	-	100
7	10	-	-	100
8	1,080	-	7	93
9	1,080	-	7	93
10	1,000	-	-	100
11	10	-	-	100
12	8,029	0.1	99.9	-
13	7,949	0.1	99.9	-
14	80	0.1	99.9	-
15	7,949	0.1	99.9	-
16	7,959	0.1	99.8	0.1
H ₂ OCAT	0.09	100	-	-
H ₂ OAN	9	100	-	-
OXYGEN1	80	-	100	-
OXYGEN2	7,950	-	99.9	0.1
HYDROGEN	1,000	-	-	100

Table 3. Mass composition of streams in MP process simulation

Stream name	Mass flow (kg/h)	Mass composition (%)		
		CH ₄	H ₂	C
METHANE	3,980	100	-	-
1	3,980	100	-	-
2	4,287	97	3	-
3	4,287	97	3	-
4	1,307	15	85	-
5	1,307	15	85	-
6	1,307	15	85	-
7	307	64	36	-
8	1,000	-	100	-
SOLID	2,980	-	-	100
HYDROGEN	1,000	-	100	-

Table 4. Comparison of main inventory data for hydrogen production scenarios

Inputs	Unit	SMR	PEM	MP
<i>From nature</i>				
-Air	kg	15,551	-	-
<i>From technosphere</i>				
-Natural gas	kg	2,921	-	-
-Water	kg	8,644	9,030	-
-Methane	kg	-	-	3,980
-Nickel catalyst	kg	504	-	-
-Iron catalyst	kg	128	-	-
-Electricity	kWh	2,295.44	41,181.97	3,087.9
-Natural gas (for heaters)	kg	911.7	37.93	589.62
-Air (for heaters)	kg	19,693	819	12,735
Outputs	Unit	SMR	PEM	MP
<i>Product</i>				
-Hydrogen	kg	1,000	1,000	1,000
<i>Byproduct</i>				
-Solid carbon black	kg	-	-	2,980
<i>Emission to air</i>				
-Oxygen	kg	-	8,030	-
-Flue gas (from process)	kg	23,431	-	-
-Flue gas (from heaters)	kg	20,604.7	856.93	13,324.62
<i>Emission to water</i>				
-Condensate	kg	2,685	-	-

3.2 LCA results

Figure 6 shows the evaluated environmental impact results for the three hydrogen production scenarios using the ReCiPe 2016 midpoint method in the Simapro LCA, with these results calculated based on H₂ production per functional unit (1,000 kg/h). The impact contributions of the main processes involved in each scenario are interpreted in Figure 7, primarily related to the background materials and energy extraction processes.

3.2.1 Global warming potential

The calculated outcomes for the global warming potential from the SMR, PEM, and MP scenarios were 17,000 kg CO₂ eq., 29,500 kg CO₂ eq. and 12,800 kg CO₂ eq., respectively, as described in Figure 6a. The PEM scenario contributed the most, followed by the SMR and MP scenarios, respectively. PEM electrolysis was primarily impacted by Thailand's fossil fuel-based grid electricity production, which accounted for approximately 85% (25,260 kg CO₂

eq.) of the overall outcome. In the SMR scenario, the majority of impacts came from the process itself, accounting for about 61% (10,400 kg CO₂ eq.) of the total impact, which was primarily related to CO₂ emission from the tail gas combustion section.

In the MP scenario, the largest impact was represented by biogas production from the anaerobic digestion in the manure process, accounting for 38.67% (4,950 kg CO₂ eq.). The second-largest impact process was methane purification from biogas, contributing 31.17% (3,990 kg CO₂ eq.). In this assessment, methane was considered to be derived from biogas since there were no available inputs in the ecoinvent background database for the production of methane from natural gas. Consequently, the GWP from the MP case was the lowest, since it emitted less CO₂ in flue gas (1,617 kg/h CO₂), compared to SMR (10,405 kg/h CO₂), as shown in Figure 5. In addition, it consumed much less energy (electricity 3,087 kWh, NG 589 kg/h) than PEM (electricity 41,181 kWh, NG 38 kg/h), as described in Figure 5 and Table 4.

For the SMR scenario, the GWP or carbon footprint was 17,000 kg CO₂ eq. This quantity accounted for total emissions from SMR associated with raw materials (feed, fuels, electricity, and catalysts) extraction, production and transportation. In addition, it considered air and water emissions from the SMR process itself up until H₂ compression to storage. Machhammer et al. (2016) reported the carbon footprint of the SMR process was 10,790 kg CO₂ eq. However, this value only quantified the emissions associated with NG extraction, transportation, and H₂ production, while excluding emissions during the production, extraction, and transportation of raw materials, such as fuels, catalysts. In addition, the production process may have contained discrepancies as the flowsheet was not shown in the literature. Amran et al. (2017) investigated SMR based on process simulation and reported a result of 11,250 kg CO₂ eq. However, this quantity did not account for electricity consumption, H₂ compression, the tail gas combustion step, catalyst extraction and production; furthermore, steam energy was used in the process.

The carbon footprint of the PEM scenario in the current study was 29,500 kg CO₂ eq. Mehmeti et al. (2018) described the GWP from their PEM electrolysis analysis as 29,540 kg CO₂ eq. if grid electricity and deionized water were used. Machhammer et al. (2016) reported GHG emission of 16,800 kg CO₂ eq. for water electrolysis; however, the exact type of water electrolysis technology and the type of water used were not specified.

More research is still required regarding GWP and LCA of methane pyrolysis technology to facilitate meaningful comparisons. The carbon footprint of the MP scenario in the current study was 12,800 kg CO₂ eq. In contrast, Machhammer et al. (2016) reported a value of 4,370 kg CO₂ eq. for their MP scenario. However, notably, their value did not consider the thermal energy supply and emissions from the furnace of the reactor. Furthermore, instead of utilizing pure methane as a feed stock, NG extraction and production were used for the evaluation of CO₂ emission in the reference literature.

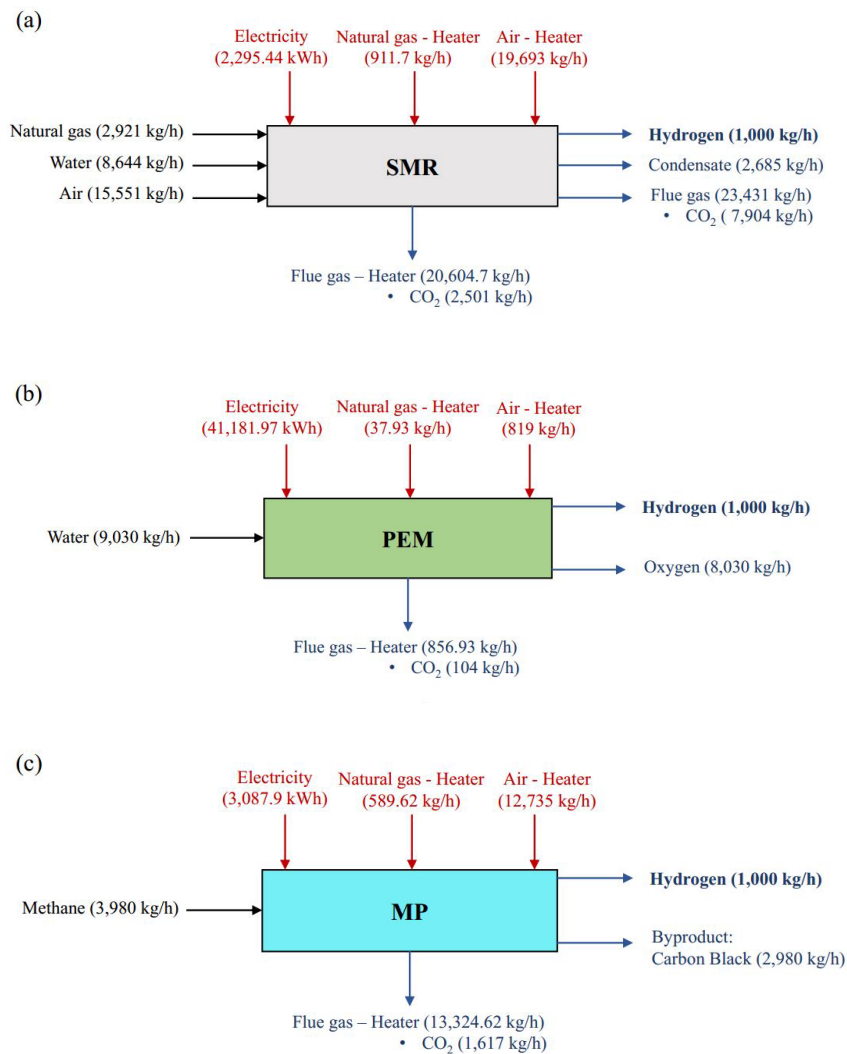


Figure 5. Simulation-based mass balances and energy usage for each hydrogen production scenario: (a) SMR, (b) PEM, and (c) MP

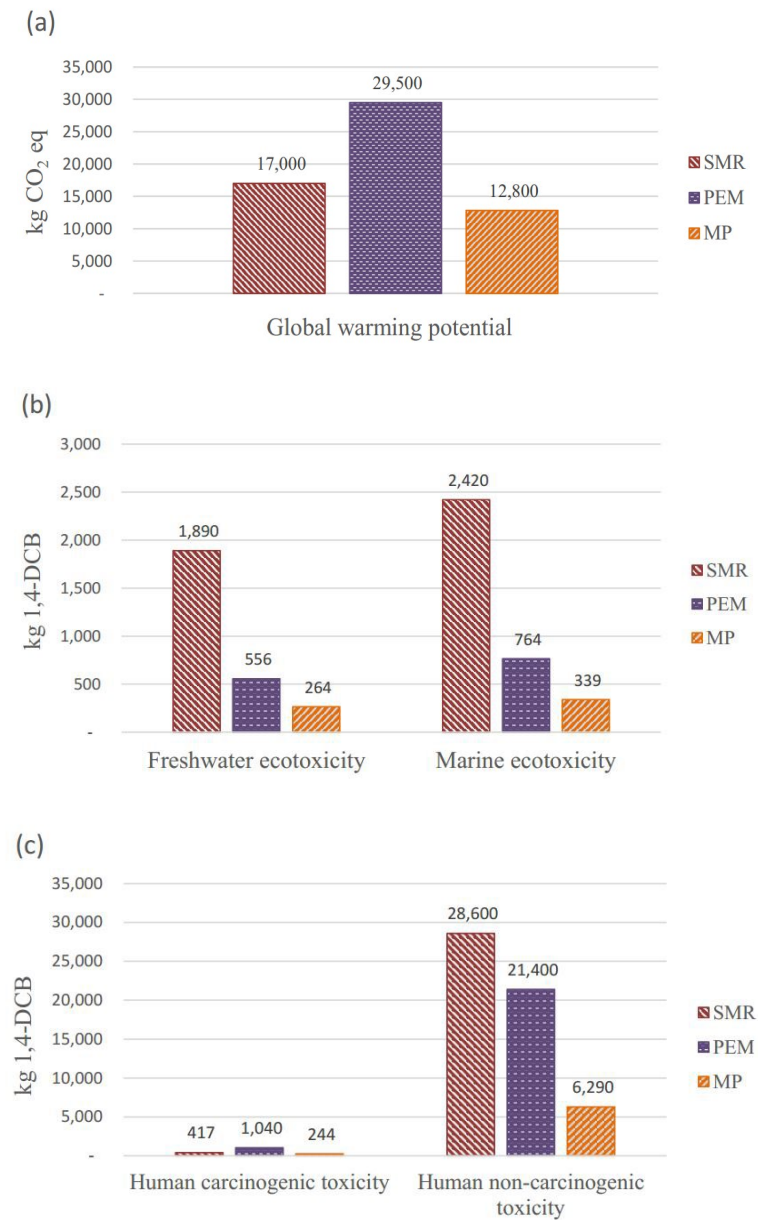


Figure 6. Characterized impact results comparison between three hydrogen production scenarios: (a) global warming potential, (b) freshwater and marine ecotoxicity, and (c) human carcinogenic and non-carcinogenic toxicity

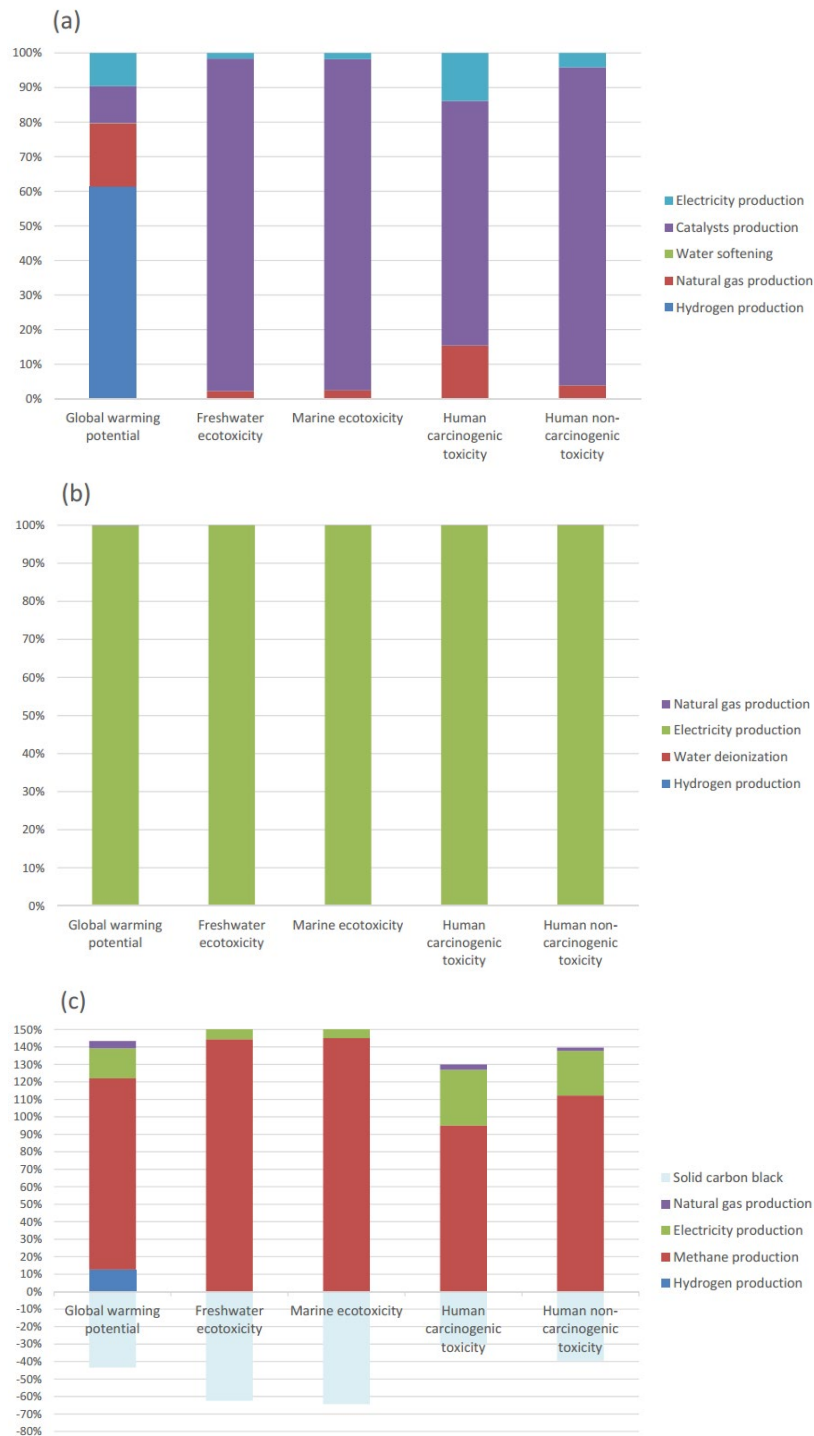


Figure 7. Process impact contribution analysis for each hydrogen production scenario: (a) SMR, (b) PEM, and (c) MP

3.2.2 Freshwater and marine ecotoxicity

The characterized sequential structure of impact contribution was identical for both freshwater and marine ecotoxicity, as shown in Figure 6b, with the SMR scenario being the major contributor, followed by PEM electrolysis, and methane pyrolysis, respectively, in both cases. The results for freshwater ecotoxicity for the SMR, PEM, and MP scenarios were 1,890 kg 1,4-DCB, 556 kg 1,4-DCB, and 264 kg 1,4-DCB, respectively. These results indicate that the SMR scenario was approximately 3.4 times larger than

the PEM scenario and 7.16 times larger than the MP scenario for freshwater ecotoxicity. The primary process in SMR was driven by sulfidic tailings from nickel mine operations, which accounted for nearly 88% (1,660 kg 1,4-DCB) of the overall impact. In the PEM scenario, a major portion of the environmental impacts resulted from grid electricity generation. Approximately 74% (411.3 kg of 1,4-DCB), originated from the leakage of spoils resulting from lignite and hard coal mining activities. The contamination of surface freshwater resulting from the

landfilling of residuals remaining during lignite, hard coal, and copper mining operations for electricity production was a key factor contributing to approximately 55% (145.9 kg of 1,4-DCB) of the total impact in MP. In all three scenarios, the level of freshwater ecotoxicity was primarily attributed to mining activities, which are integral to the grid electricity generation powering the H₂ production processes.

The marine ecotoxicity results for the SMR, PEM, and MP scenarios were 2,420 kg 1,4-DCB, 764 kg 1,4-DCB, and 339 kg 1,4-DCB, respectively, as shown in Figure 6 (b). The structure of the proportionate characterized results from freshwater ecotoxicity were relatively equal to those from marine ecotoxicity, with the SMR scenario having the greatest impact, being approximately 3.2 times larger than for the PEM scenario and 7.14 times larger than for the MP scenario. The treatment process for the sulfidic tailings from the nickel mining operation made the largest contribution to the marine ecotoxicity category in the SMR scenario with 86% (2,087 kg 1,4 DCB) of the total impact.

Therefore, MP had the lowest impact on the freshwater and marine toxicity aspects compared to SMR and PEM. The MP did not require any catalyst for H₂ synthesis, thus avoiding tailings inflow from the mining operation of catalysts (nickel and iron) to the surface and ground water, unlike for SMR. In PEM, water is a primary raw material; 9,030 kg/h of deionized water was consumed to produce 1,000 kg/h of H₂, thereby affecting aquatic ecosystems.

3.2.3 Human carcinogenic and non-carcinogenic toxicity

The PEM electrolysis scenario dominated in its impact on human carcinogenic toxicity compared to the other two scenarios, as shown in Figure 6c. The human carcinogenic toxicity levels for the SMR, PEM, and MP scenarios were 417 kg 1,4-DCB, 1,040 kg 1,4-DCB, and 244 kg 1,4-DCB, respectively. The primary processes contributing to human carcinogenic toxicity in the PEM scenario were attributed to the impact of lignite and hardcore spoil leachate outflows, resulting from mining operations for electricity production in coal-fired power plants. These processes accounted for approximately 76% (790 kg 1,4-DCB) of the overall toxicity.

The SMR scenario represented the highest impact contribution to human non-carcinogenic toxicity, followed by the PEM and MP scenarios, respectively, as shown in Figure 6 (c). The human carcinogenic toxicity levels for the SMR, PEM, and MP scenarios were 28,600 kg 1,4-DCB, 21,400 kg 1,4-DCB, and 6,290 kg 1,4-DCB, respectively. For the SMR scenario, the primary source contribution to its high level was the toxic metals that were typically produced and remained in large quantities from the nickel mining operation, attributing nearly 80.9% (23,140 kg 1,4-DCB) of the entire impact. Groundwater contamination from lignite, coal mining, and lignite ash treatment for coal-fired electricity production were critical in the PEM scenario accounting for 80.7% (17,270 kg 1,4-DCB) of the overall impact. The main cause in the MP scenario was similar to that for the PEM scenario, with around 48% (3,040 kg 1,4-DCB) of the impact mostly contributed by wastes generated from the lignite and hard coal mining processes.

These results suggest that MP had the least impact in terms of human health because it mostly eliminated emissions from the upstream processes. MP consumed less electricity (3,087 kWh) compared to PEM (41,181 kWh)

and also was exempt from any catalyst extraction processes, unlike SMR that included nickel (504 kg) and iron (128 kg) extraction. In addition, the methane utilized in MP was produced from biogas, making it a more sustainable raw material than the natural gas used in SMR.

4. CONCLUSION

This study undertook a comparative environmental impact assessment of three hydrogen production technologies to provide alternative options throughout the industrial decarbonization pathway in Thailand. The assessment was performed using Aspen Plus process modelling and the Simapro software for LCA. Steam methane reforming, a conventional worldwide hydrogen production technology, was compared to two competitive low carbon emission hydrogen productions: PEM electrolysis and methane pyrolysis. In addition to global warming potential, this analysis considered freshwater, marine, human carcinogenic, and non-carcinogenic toxicity midpoint impact categories.

The global warming potential of PEM electrolysis was approximately 1.7 times higher than for the conventional steam methane reforming (since the electricity was mainly derived from the country's grid mix production) and 2.3 times larger than for methane pyrolysis. However, the effect of PEM electrolysis on the levels of freshwater, marine, and human non-carcinogenic toxicity were still lower than for SMR out of five midpoint impact categories.

The results indicated that methane pyrolysis was an interesting alternative hydrogen production method, producing the lowest environmental damage in all five impact categories. Regardless of the grid electricity demand, the global warming potential of methane pyrolysis was nearly 25% and 57% lower than for SMR and PEM electrolysis, respectively. Furthermore, solid black carbon from methane pyrolysis could help to reduce fossil resources scarcity by promoting a circular carbon economy. Even though methane was a primary feedstock in the process, it can be considered as a bridging hydrogen synthesis technology, especially during the period when instantaneous phasing out of fossil fuel consumption is not feasible.

In contrary to traditional steam methane reforming, neither PEM electrolysis nor methane pyrolysis generated process CO₂ during hydrogen production except from the emission from the production of background raw material and electricity. PEM electrolysis could serve as a long-term hydrogen production technology if it were implemented along with renewable energies. Although currently, methane pyrolysis has not yet been fully commercialized (Schneider et al., 2020), it can be considered as a potential attractive medium-term hydrogen production technology with low carbon emission during the shift from fossil fuels to the era of renewable energies.

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