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**Master's Thesis**

**A study on the life cycle assessment for the calculation of greenhouse gas emissions and the evaluation of eco-friendliness in the production of clean hydrogen using biogas**

**The Graduate School  
of the University of Ulsan  
Department of Mechanical Engineering  
Azamly Mariam S I**

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**Supervisor: Prof. Ocktaeck Lim**

**A Thesis**

**Submitted to**

**The Graduate School of the University of Ulsan**

**In partial Fulfillment of the Requirements**

**for the Degree of**

**Master**

**by**

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**June 2024**


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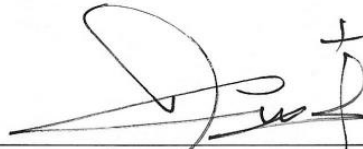
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## ABSTRACT

The increasing demand is driving a rapid development in hydrogen production. However, since the procedures involved in producing hydrogen have the potential to pollute the environment, more considerations must be taken. This work specifically looks at carbon dioxide CO<sub>2</sub> emissions associated with the production of hydrogen which is a significant contributing factor to the greenhouse effect. This study offers a comprehensive numerical analysis of the carbon dioxide emissions associated with hydrogen production from various renewable feedstocks. The study focuses on renewable feedstocks, such as landfill gas, animal and food waste, and wastewater (sludge). Robust examination yields interesting revelations on carbon dioxide emissions using GREET 2022 software. The discovery that using wastewater sludge as a feedstock result in the lowest carbon dioxide emissions (-26.83 kg CO<sub>2</sub>/kgH<sub>2</sub>) is especially notable unlike, blending above 25% with other renewable feedstocks which was noticed to raise carbon dioxide emissions correspondingly. Furthermore, it is disclosed that the utilization of landfill gas results in the maximum emissions of carbon dioxide among all different waste combinations, amounting to (3.92 kg CO<sub>2</sub>/kg H<sub>2</sub>). These results not only greatly advance the usage of renewable feedstocks but also offer insightful information to stakeholders seeking to reduce greenhouse gas emissions.

**Key words:** LCA, GREET, GHG, SMR, RNG

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## LIST OF ABBREVIATIONS

AD	Anaerobic digestion
AW	Animal Waste
CCS	Carbon capture and storage
CCU	Carbon capture and utilization
CCUS	Carbon capture, utilization and storage
CO <sub>2</sub>	Carbon dioxide
CO	Carbon monoxide
Co-Mo	Cobalt-molybdenum
Co-Ni-Mo	Cobalt-nickel-molybdenum
CHP	Combined heat and power
CuO	Copper oxide
CuS	Copper sulfide
PM10	Fine dust
GHG	Green House Gas
REET	Greenhouse gases, Regulated Emissions, and Energy use in Technologies
HDS	Hydrodesulfurization
H <sub>2</sub>	Hydrogen gas
H <sub>2</sub> S	Hydrogen sulfide
HS	Hydrogen sulfide
Fe <sub>2</sub> O <sub>3</sub>	Iron oxide
FeS	Iron sulfide
LFG	Land fill gas
LCA	Life cycle assessment
LNG	Liquified natural gas
LTS	Low-Temperature Shifter
LT-WSR	Low-temperature water gas shift reaction
MSW	Municipal waste
NG	Natural gas
NO <sub>x</sub>	Nitrogen oxides
N <sub>2</sub> O	Nitrous oxide
O <sub>2</sub>	Oxygen gas
PSA	Pressure Swing Adsorption

RNG	Renewable natural gas
SMR	Steam methane reforming
SOX	Sulfur oxides
PSA	Pressure swing adsorption
PM2.5	Ultrafine dust
VOC	Volatile organic compounds
WS	Water Sludge
WGS	Water–gas shift reaction
WTG	Well to gate
WTW	Well to Wheel
ZnO	Zinc Oxide
ZOD	Zinc Oxide Desulfurization

# 1 INTRODUCTION

## 1.1 Hydrogen Global Trend

Clean hydrogen is displacing current fossil fuels (diesel, gasoline, etc.) in transportation across the globe in order to lower greenhouse gas emissions and encourage energy conversion. Hydrogen's enormous energy storage capacity and primary application in hydrogen mobility mean that it will be a key coupling to attain carbon neutrality. Nevertheless, there is not much of a greenhouse gas reduction impact when using hydrogen for transportation.

Major nations worldwide are pledging to become carbon neutral in response to climate change, and one keyway to do this is by enacting laws governing the production, delivery, storage, and use of hydrogen—a fuel and energy source that emits no carbon—as well as other related measures. Despite the fact that hydrogen is essentially carbon-free, the majority of it is now created through the reforming reaction of natural gas and by-product gases from petrochemical and oil refining activities. By-product hydrogen and natural gas reformed hydrogen are categorized as gray hydrogen because they are not ecologically friendly, and throughout the production process, carbon dioxide emissions range from 11.39 to 15.02 kg-CO<sub>2</sub>/kg-H<sub>2</sub>[1]. Thus, the predominant hydrogen (gray) of today is a green energy source with various consequences for lowering greenhouse gas emissions worldwide.

Renewable hydrogen is also known as clean hydrogen or low-carbon hydrogen, which is smaller than the original but produces fewer greenhouse gases. Building an environmentally friendly hydrogen ecosystem that includes the production, supply, storage, and use of hydrogen using the renewable hydrogen) manufacturing technique is of great interest globally and locally

The goal of the Korean government's initiatives to supply eco-friendly fuels and become a carbon neutral nation is to broaden the basis of the clean hydrogen ecosystem. As a climate response fund project, the Ministry of Environment started a pilot project in 2022[2] to investigate the whole life cycle assessment (LCA) of hydrogen generation based on biofuel. The initiative produces clean hydrogen utilizing biogas based on organic waste resources. Promoting methodical and successful initiatives requires knowledge of the institutional and financial landscape both domestically and internationally and the development of a plan to increase local production of sustainable, clean hydrogen that is self-sufficient.

## 1.2 Clean Hydrogen Certification

IPHE[3] is the (International Partnership for Hydrogen and Fuel Cells in the Economy). Formed in 2003 to promote “international cooperation on hydrogen and fuel cell R&D, common codes and standards, and information sharing on infrastructure development”. Member countries are Australia, Austria, Belgium (new member), Brazil, Canada, Chile, China, Costa Rica, France, European Commission, Germany, Iceland, India, Italy, Japan, South Korea, Netherlands, Norway, South Africa,

Switzerland, Singapore (new member), United Arab Emirates (new member), UK, US. Among IPHE members there are two working groups that have been formed and are active in IPHE: the Education and Outreach (E&O) Working Group, and the Regulations, Codes, Standards, and Safety (RCSS) Working Group. Nevertheless, there are also individual "major countries" efforts led mostly by governments and some other times by private sectors for H2 certification as summarized in Figure 1.1(a) and 1.2(b).

The nations listed in figures 1.1(a) and 1.2(b) are members of the G7, G20, and IPHE. It was verified that 77% of IPHE member nations have clean hydrogen certification and methodology, and 55% of G20 nations have implemented clean hydrogen standards. It is evident, therefore, that every nation has a unique certification procedure for clean hydrogen, or low-carbon hydrogen.

Table 1-1 Clean hydrogen (low-carbon hydrogen) certification method by major countries (a)

Country	Standard matter	Certification Name	range	boundary	Manufacturing method	Year of announcement	implementation status	H2 purity pressure	reference value kg-CO <sub>2</sub> e/kg-H <sub>2</sub>	Biogas clean H <sub>2</sub> case
IPHE	H2	N/A	Scope 1,2,3	WTG	6 methods	2022	-	Pressure and purity corresponding to the requirements of subsequent steps	Not Specified	O
EU	H2	EU Taxonomy (government)	-	WTG	Not specific	2021	operating	-	3.0	-
		RED III (Renewable Energy Directive III) (government)	Scope 1,2,3	WTW	Water electrolysis (renewable energy power, low carbon power) Recycled carbon fuel	2023	under preparation	99% 3MPa (30bar)	3.4	-
		CertifyH (private)	-	WTG	Electrolysis (Renewable energy power, nuclear power) Natural gas CCS	2018	operating	-	4.4	-
		TUV SUD CMS 70 (private)	-	WTG	Water electrolysis (renewable energy power) Biomethane glycerin	2021	operating	-	1.1	-
UK	H2	UK Low Carbon Hydrogen Standard (Government)	Scope 1,2,3	WTG	Water electrolysis (regardless of power source) Natural gas CCS Biomass, waste	2023	under preparation	99% 3MPa (30bar)	2.4	O (0.69 kgCO <sub>2</sub> e/kg-H <sub>2</sub> )
		Renewable Transport Fuel Obligation (RTFO)	-	Well to point of delivery	Water electrolysis (renewable energy power) excluding biomass	2021	operating	-	4.0	-
France	H2	French Ordinance No. 2021-167 (government)	Scope 1,2,3	WTG	Not specific	2023	under preparation	99% 3MPa (30bar)	3.38	-
US	H2	Clean Hydrogen Production Tax Credit (IRA) (government, state)	Scope 1,2,3	WTG	Not specific	2022	under preparation	99% 3MPa (30bar)	2.5-4 1.5-2.5 0.45-1.5 <0.45	O (47 cases - California)

Table 1-2 Clean hydrogen (low-carbon hydrogen) certification method by major countries (b)

Country	Standard matter	Certification Name	range	boundary	Manufacturing method	Year of announcement	implementation status	H2 purity pressure	reference value kg-CO <sub>2</sub> e/kg-H <sub>2</sub>	Biogas clean H <sub>2</sub> case
Canada	H2	Clean Hydrogen Investment Tax Credit	Scope 1,2,3	WTG	Water electrolysis (regardless of power source) Natural gas CC	2022	Under preparation	-	2.4 0.75-2 <0.75	-
Japan	H2	Hydrogen basic strategy (government, local government)	Scope 1,2,3	WTG	Not specific	2023	under preparation	-	3.4	O (8 cases) Local government
	Ammonia	Hydrogen Basic Strategy (Government)	Scope 1,2	GTG	Not specific	2023	under preparation	-	5.1 (0.84tCO <sub>2</sub> /t NH <sub>3</sub> (excluding NG procurement process))	-
China	H2	Standard and evaluation of low-carbon hydrogen, clean hydrogen and renewable hydrogen (private)	-	WTG	Not specific	2022	Under preparation	-	Low carbon hydrogen 14.5 Clean hydrogen 4.9	O
India	H2	No. 353/35/2022-NT Ministry of New and Renewable Energy National Green Hydrogen Mission	Scope 1	WTG	Water electrolysis - biomass	2023	Under preparation	Average value of consent over the last 12 months	2.0	O

For some major countries the clean hydrogen policies will be mentioned further like the following:

1. United States(US):

Hydrogen utilizing presence is undeniable in the United States specially in California with general booming in the FCEV sales along the last decade reaching around 18000 vehicles by this year 2024 and with an escalating manner of establishing new Hydrogen units as shown in the figures below:

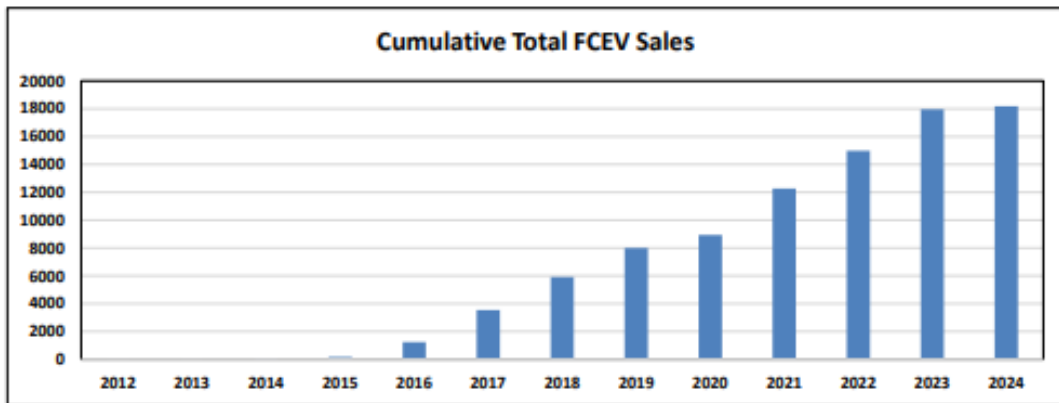


Figure 1.1 Vehicle sales data from Baum and Associates. Sales data is based on car sales sold by a dealer to a retail or fleet customer [4]

Table 1-3 Official number of hydrogens fueling units in the United States both operating and under construction [5]

Station Counts by State and Fuel Type									
State	Biodiesel	CNG	E85	Electric <sup>a</sup> (station locations   charging ports Level 1   Level 2   DC Fast)	Hydrogen <sup>b</sup> (retail   non-retail   total)	LNG	Propane <sup>c</sup> (primary   secondary   total)	Renewable <sup>d</sup> Diesel	Total <sup>d</sup>
Ohio	4	50	217	1,741   4,061 41   3,129   891	0   2   2	4	50   49   99	0	4,437
Oklahoma	18	100	101	360   1,367 6   521   840	0   0   0	0	30   79   109	0	1,695
Oregon	30	13	5	1,355   3,491 73   2,543   875	0   0   0	1	31   13   44	7	3,591
Pennsylvania	4	77	172	1,861   4,755 51   3,589   1,115	0   0   0	2	48   45   93	0	5,103
Rhode Island	1	1	0	333   898 82   734   82	0   0   0	0	3   2   5	0	905
South Carolina	33	13	58	609   1,600 15   1,074   511	0   0   0	1	25   17   42	0	1,747
South Dakota	0	0	81	104   254 2   138   114	0   0   0	0	4   11   15	0	350
Tennessee	5	21	90	962   2,380 34   1,770   576	0   0   0	4	28   26   54	0	2,554
Texas	10	96	265	3,385   9,655 86   6,880   2,689	0   0   0	15	177   179   356	0	10,397
Utah	1	41	0	910   2,344 23   1,930   391	0   0   0	0	18   21   39	0	2,425
Vermont	2	3	0	426   1,090 21   900   169	0   0   0	0	0   2   2	0	1,097
Virginia	7	24	84	1,636   4,883 218   3,464   1,201	0   1   1	1	41   36   77	0	5,077
Washington	24	23	13	2,324   6,209 163   4,803   1,243	0   1   1	1	49   21   70	0	6,341
West Virginia	1	3	36	161   470 19   299   152	0   0   0	0	6   9   15	0	525
Wisconsin	23	46	263	648   1,547 6   1,074   467	0   0   0	1	21   40   61	0	1,941
Wyoming	0	7	8	109   265 3   141   121	0   0   0	0	6   10   16	1	297
<b>Total</b>	<b>1,755</b>	<b>1,377</b>	<b>4,565</b>	<b>68,404   192,091 2,976   145,916   43,171</b>	<b>56   18   74</b>	<b>94</b>	<b>1,374   1,408   2,782</b>	<b>607</b>	<b>203,345</b>

Table 1-4 United States hydrogen status and certification and support measures[5][6]

Country	Clean hydrogen status	Certification and support measures
U.S	<p>- In the United States, hydrogen fuel cell vehicles (hydrogen vehicles) are being distributed with subsidies from the state of California and hydrogen charging stations are being installed and operated along with the distribution of hydrogen vehicles</p> <p>- Hydrogen vehicles: 18180 total and 66 buses in CA</p> <p>- Hydrogen charging stations: 55 in CA, total 74</p>	<p>Clean H2 certification system:</p> <p>- A system that certifies hydrogen as clean and provides incentives when greenhouse gas emissions are below a certain level during the process of producing or importing hydrogen (minimum standard for clean hydrogen: 4kg or less of carbon emissions per 1kg of hydrogen production)</p> <p>- If clean hydrogen is produced in accordance with the Inflation Reduction Act (IRA), tax benefits of up to \$3 per 1 kg of hydrogen and up to 30% for investment in related facilities are provided.</p>

Tax credits for clean hydrogen production in the U.S:

- Tax deduction amount calculation method (multiplication of the items below)
- kg of clean hydrogen produced by the taxpayer at a clean hydrogen production facility during the relevant tax year over a period of 10 years from the date the facility was first put into operation.
- Amount related to the hydrogen in question
- Applicable amount:

1. The amount is the same as the corresponding rate of \$0.60. If the amount determined is not a multiple of 0.1 cent, the amount is the nearest 0.1 cent.

Rounded to the nearest multiple.

Application ratio: Application ratio of clean hydrogen produced through the following process with life cycle greenhouse gas emissions:

- Carbon dioxide emissions per 1kg of hydrogen between 2.5kg and less than 4kg: 20%
- Carbon dioxide emissions per 1 kg of hydrogen: 1.5 kg or more but less than 2.5 kg: 25%
- Carbon dioxide emissions per 1 kg of hydrogen: 0.45 kg or more but less than 1.5 kg: 33.4%

Carbon dioxide emissions per 1 kg of hydrogen less than 0.45 kg: 100% [6]



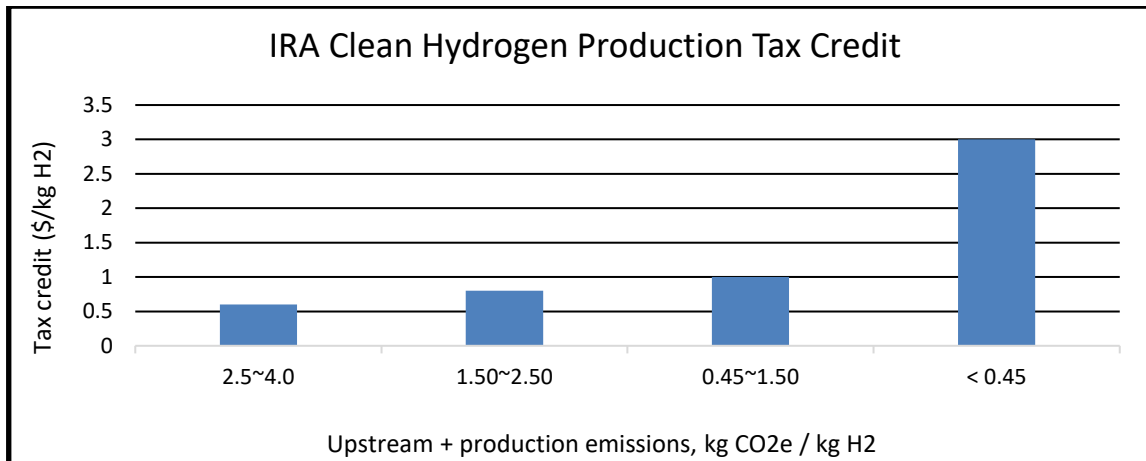


Figure 1.2 IRA Clean Hydrogen Production Tax Credit [6]

California state shows a leading role hydrogen supporting regulations with the LCFE (Low Carbon Fuel Standard) regulation, which aims to reduce greenhouse gas emissions related to transportation, was approved and implemented by the state of California in 2009, and this system is the predecessor of the U.S. federal government's clean hydrogen certification system. Major highlight is:

- Carbon Intensity (CI) value is assigned to each type of fuel, and is generally measured as carbon dioxide equivalent per energy unit (g CO<sub>2</sub>e/MJ, etc.)
- Well to Wheel boundary
- Calculate carbon intensity CI for each project through GREET 2022 version

The figure below shows the percentage decrease in the transportation fuel pool's carbon intensity (CI) for California. The LCFS aim is to establish a declining annual target, or compliance level, and work toward a 20% decrease from a baseline year of 2010 by 2030. Owing to legal issues, the compliance level was set at a 1% reduction from 2013 to 2015. Banked credits are produced by years when the market was introduced to alternative fuels in greater quantities (green line) than were required to achieve the compliance criterion (black line). Future years, such as 2020, can use banked credits to meet the need. After 2030, the program will still exist, albeit with a 20% cut[7].

### 2011-2023 Performance of the Low Carbon Fuel Standard

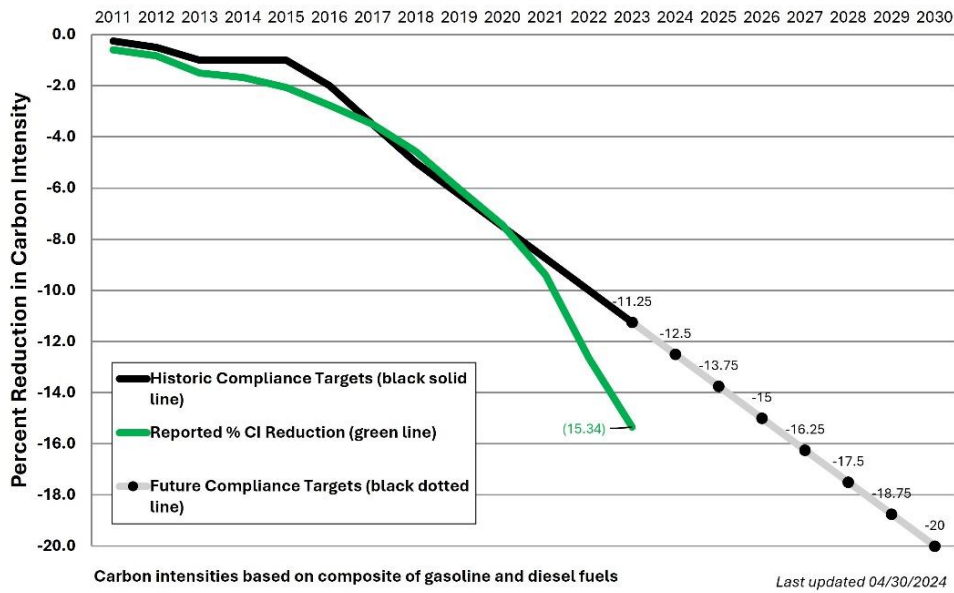


Figure 1.3 2011-2013 Performance of the Low Carbon Fuel Standard [7]

### 2023 Volume-weighted Average Carbon Intensity by Fuel Type for Non-Liquid Fuels

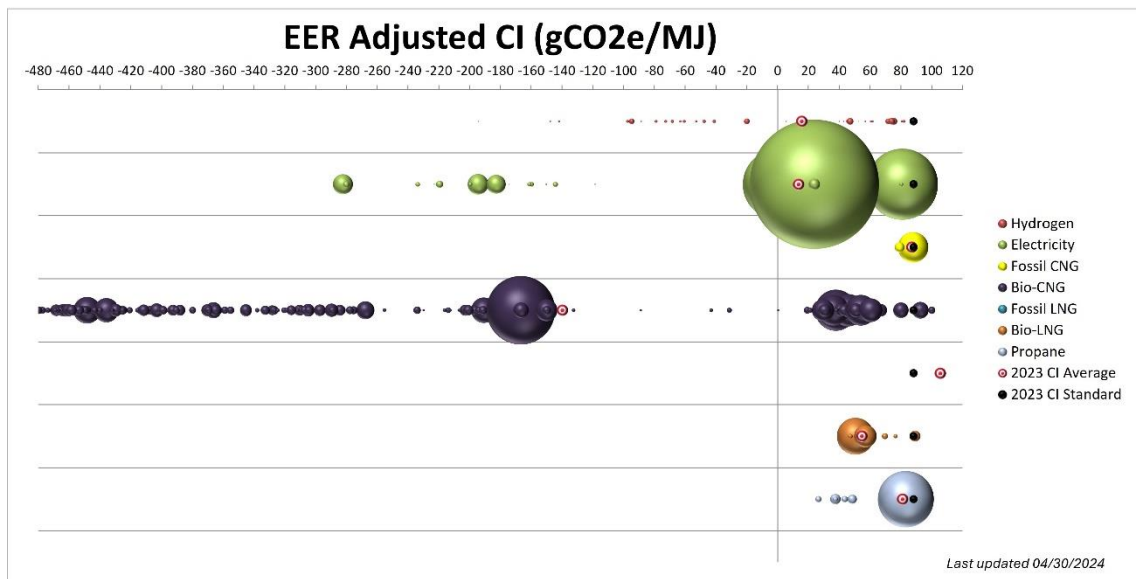


Figure 1.4 2023 Volume-weighted Average Carbon Intensity by Fuel Type for Non-Liquid Fuels[7]

## 2. Canada:

Canada as well showing efforts on enhancing hydrogen promoting in their economy by 2050.

Canada hydrogen policy similar to the United States as illustrated below:

Table 1-5 Canada hydrogen status and certification and support measures[8]

Country	Clean hydrogen status	Certification and support measures
Canada	<ul style="list-style-type: none"> <li>- Goal for realizing hydrogen economy in 2050: Increase hydrogen production by 7 times and reduce production costs by 8 times.</li> <li>- (Hydrogen production) Build a carbon capture facility to produce 20 million tons of low-carbon hydrogen per year</li> <li>- Proportion of hydrogen in national energy mix: 1.6% (2024) → 6.2% (2030) → 30% (2050)</li> <li>- (Production cost) decreased from C\$5.0~12/kg (2025) to C\$1.3~3/kg (2050)</li> <li>- (Carbon Reduction) Carbon emissions reduction expected to be 190 million tons in 2050</li> <li>- (Economic effect) 350,000 hydrogen economy jobs + creation of a market worth C\$50 billion</li> </ul>	<ul style="list-style-type: none"> <li>Clean hydrogen certification system</li> <li>Minimum standard for clean hydrogen like the United States: less than 4kg of carbon emissions per 1kg of hydrogen production.</li> <li>Clean hydrogen support plan</li> <li>production subsidies</li> <li>- Estimated life cycle emissions per kg of hydrogen produced - carbon dioxide equivalent (CO<sub>2</sub>e) of less than 0.75 kg: 40%</li> <li>- Estimated life cycle emissions per kg of hydrogen produced - 0.75kg to 2kg of carbon dioxide equivalent (CO<sub>2</sub>e): 25%</li> <li>- Expected life cycle emissions per kg of hydrogen produced - 2kg to 4kg carbon dioxide equivalent (CO<sub>2</sub>e): 15%</li> <li>- Introducing a 30% refundable investment tax credit for clean hydrogen production</li> </ul>

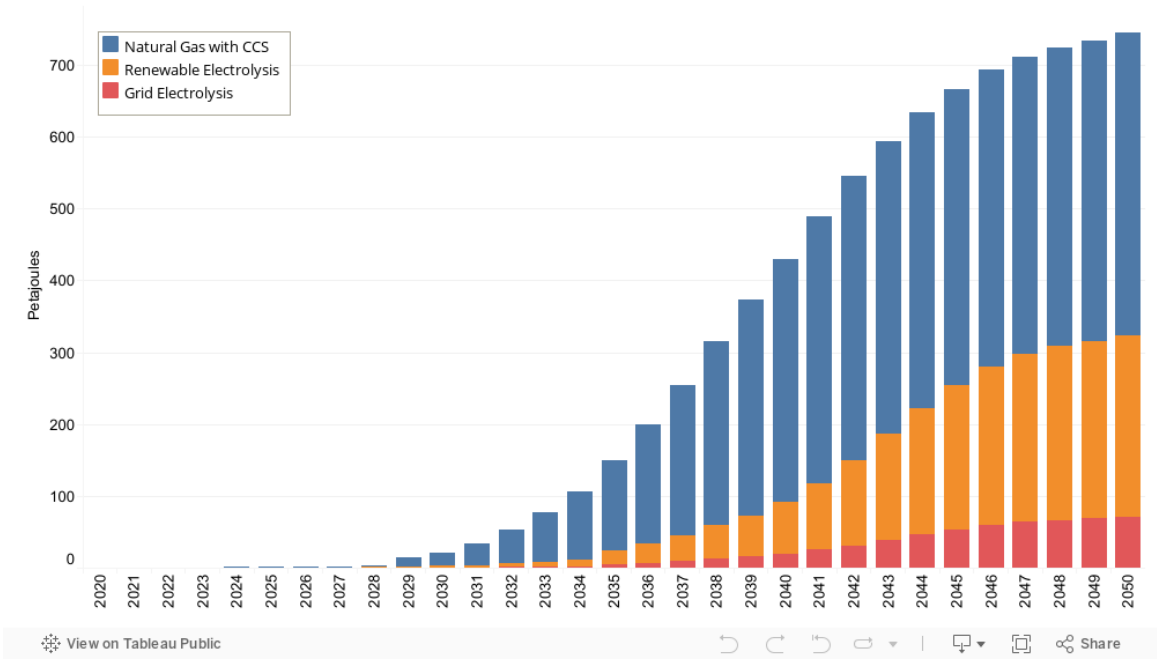


Figure 1.5 Canada fuel demand, Low carbon hydrogen production[9]

This bar graph illustrates how much electricity and natural gas are needed in grid electrolysis, renewable electrolysis, and natural gas with carbon capture and storage processes to create low-carbon hydrogen in the EF2021 Evolving Policies Scenario. The demand for natural gas increases from 19 petajoules in 2030 to 338 petajoules in 2030 and 422 petajoules in 2050 as a result of the carbon capture and storage process. From 2 petajoules in 2030 to 71 petajoules in 2040 and 252 petajoules in 2050, the amount of electricity required from renewable electrolysis grows. From 0.5 petajoules in 2030 to 20 petajoules in 2040 and 70 petajoules in 2050, the amount of electricity required for grid electrolysis rises.

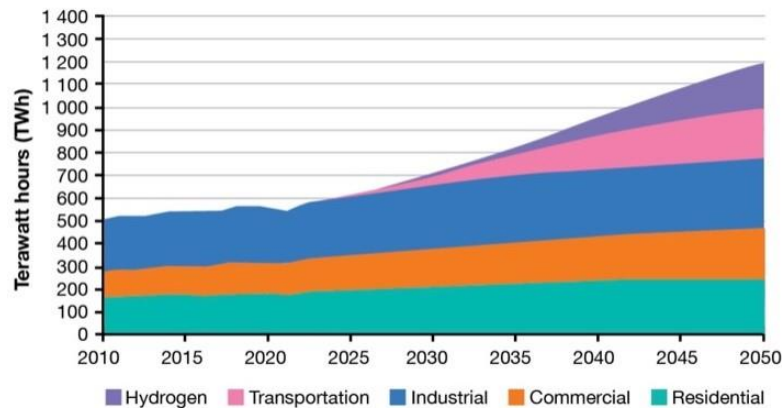


Figure 1.6 Canada 2050 Global Net-Zero Scenario[10]

### 3. Europe Union(EU):

Europe union countries are demonstrating a strong competitive action plan including current and

near future situation and beyond as presented below:

Table 1-6 Europe Union hydrogen status and certification and support measure[11]

Country	Clean hydrogen status	Certification and support measures
Europe Union	<ul style="list-style-type: none"> <li>- Most of the hydrogen currently produced is hydrogen produced based on fossil fuels, and clean hydrogen based on renewable energy is very limited.</li> <li>- Europe announced the European Hydrogen Strategy in 2020 and is promoting policies across all fields, including hydrogen production, infrastructure, and industrial application, through various initiatives and laws such as REPowerEU in 2022.</li> <li>- Hydrogen accounts for less than 2% of EU energy demand, most of which is used as a fuel in crude oil refining and production of ammonia, methanol and hydrogen peroxide.</li> <li>- The European Commission predicts that a total investment of 86 billion to 126 billion euros will be needed to build major hydrogen infrastructure, including pipelines, storage, electrolyzers, and strengthening production capabilities, by 2030.</li> <li>- For the above investment, various funds such as Horizon Europe, Innovation Fund, Cohesion Fund, and Fair Transition Fund will be invested.</li> </ul>	<ul style="list-style-type: none"> <li>- EU Commission's proposal for hydrogen definitions and standards</li> <li>- Low-carbon hydrogen: Hydrogen extracted from non-renewable energy, greenhouse gas emissions reduction standard is 70% –3.4 kg CO<sub>2</sub>e/kg H<sub>2</sub>, LHV standard</li> <li>- Defined in delegated legislation until the end of 2024</li> <li>- Delegated Act for RFNBO and RCF Greenhouse Gas Reduction Standards and Methodology</li> <li>- Must meet the 70% greenhouse gas reduction standard of 94g CO<sub>2</sub>e/MJ (10.80kg CO<sub>2</sub>e/kg H<sub>2</sub>, LHV standard), which is the standard for comparing fossil fuels.</li> <li>- To date, the clean hydrogen certification system does not recognize hydrogen of biological origin, only hydrogen of abiotic origin has been announced, and legislation is currently in progress.</li> </ul>

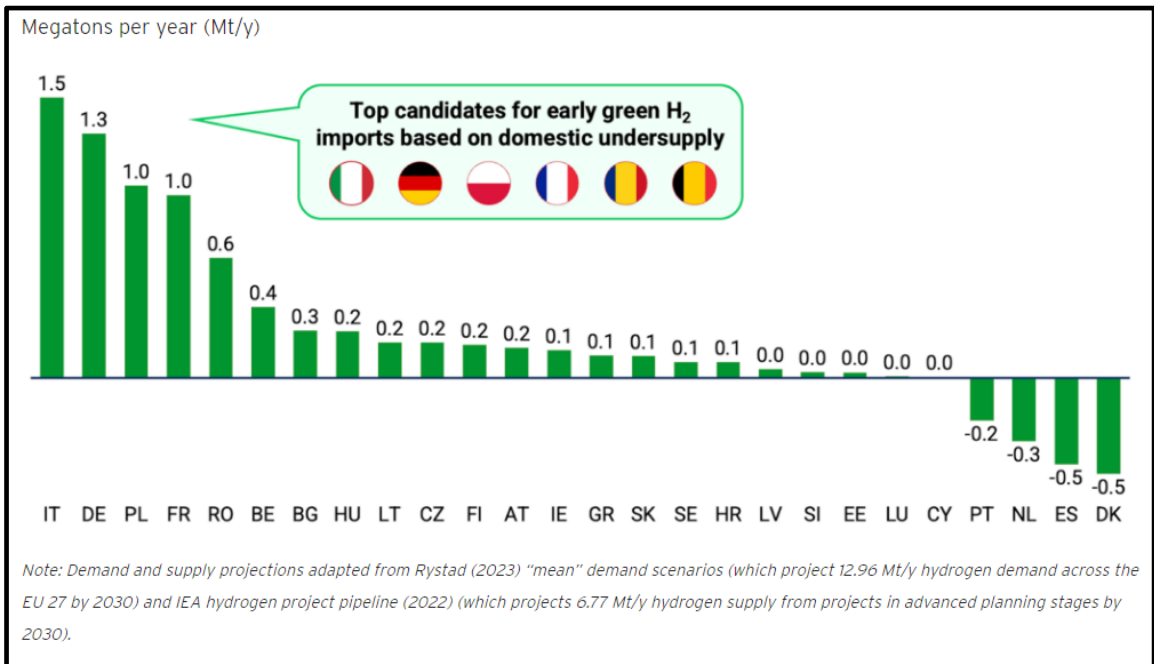


Figure 1.7 Gap between projected demand for green hydrogen in EU Member States and expected supply (based on announced projects) in 2030[12]

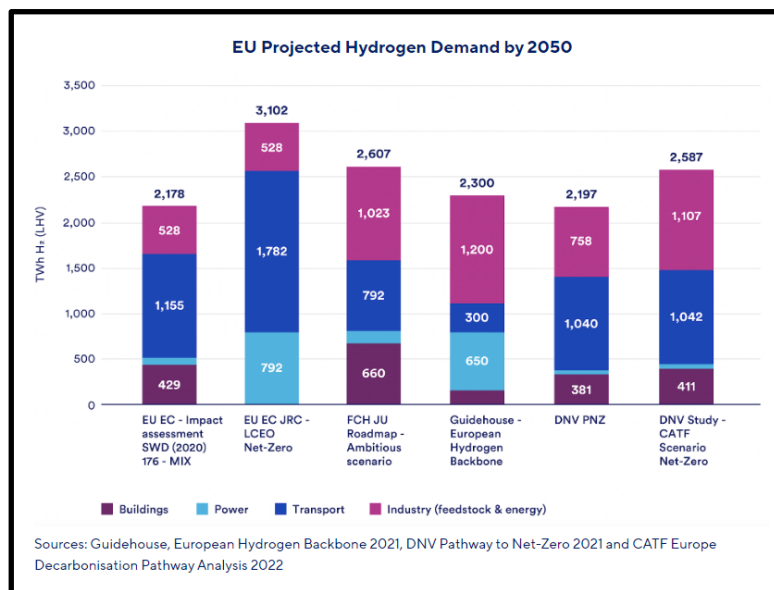


Figure 1.8 EU Projected Hydrogen Demand by 2050[13]

#### 4. United Kingdom(UK)

United Kingdom still not far away from the clean hydrogen trend with generous investments promoting the decarbonization infrastructure as shown below:

Table 1-7 UK hydrogen status and certification and support measure[14]

Country	Clean hydrogen status	Certification and support measures
United Kingdom	<p>- UKRI's Industrial Decarbonization Challenge offers £170 million matched by £261 million from industry to invest in developing industrial decarbonization infrastructure, including CCUS and low carbon hydrogen, for various sectors.</p> <p>- Scotland has the potential to contribute significantly to the UK hydrogen economy, producing industrial-scale hydrogen from offshore wind resources and CCUS. Economic analysis suggests that Scotland could deliver 21-126TWh of hydrogen per year by 2045, generating significant jobs and local economic benefits.</p>	<p>- The UK government issued “Guidance on greenhouse gas emissions and sustainability criteria under the UK Low Carbon Hydrogen Standard” on April 18/May 18, 2023. During the announcement, the threshold for low-carbon hydrogen certification was presented.</p> <p>- Critical emission intensity: 20 gCO<sub>2</sub>e/production H<sub>2</sub> MJ LHV [2.4 kg CO<sub>2</sub>e/kg H<sub>2</sub>]</p> <p>- Scope of application: Hydrogen production site (Point of production)</p>

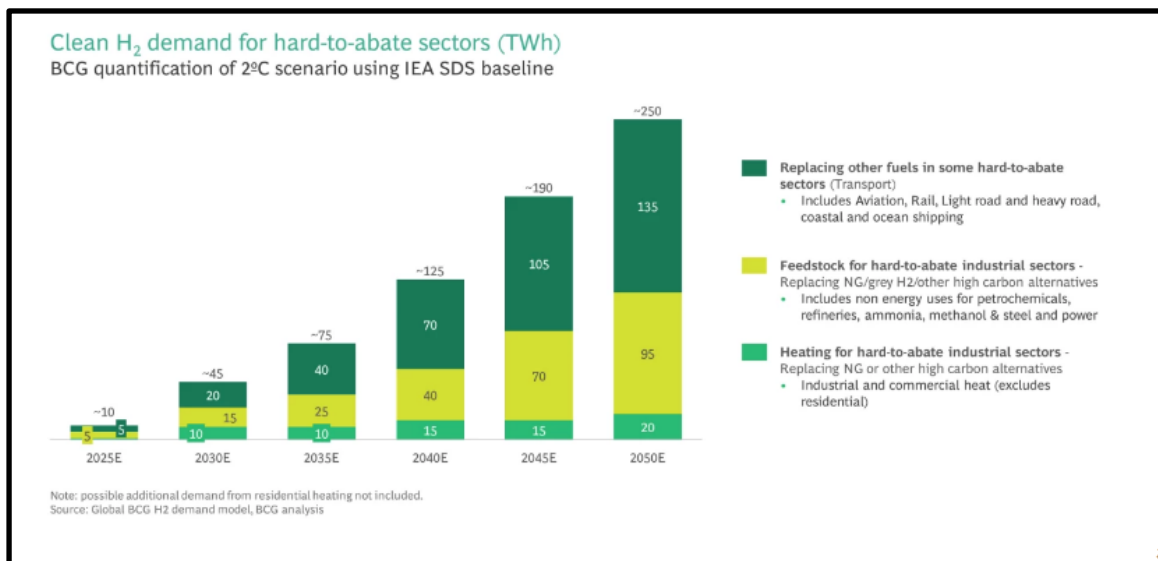


Figure 1.9 Forecast for hydrogen demand in the UK for hard-to-abate sectors[14]

5. Japan

Table 1-8 Japan hydrogen status and certification and support measure[15]

Country	Clean hydrogen status	Certification and support measures
Japan	<ul style="list-style-type: none"> <li>- As of 2030, the goal is WTG ~ 3.4kg CO<sub>2</sub>e/kg H<sub>2</sub> (equivalent to a 70% reduction in natural gas SMR)</li> <li>- Within 5 years, considering technology development in line with overseas standards and regulatory trends, CCS-new and renewable energy-resource development, overseas trends, economic feasibility, etc.</li> </ul>	<ul style="list-style-type: none"> <li>-Clean Hydrogen Certification Promotion Status:               <ul style="list-style-type: none"> <li>- Japan Hydrogen Association, “CO<sub>2</sub>-Free Hydrogen Committee” proposed with reference to international standards (IPHE) standards (November 2022)</li> <li>- The Japanese government announced that it proposed a clean hydrogen (low-carbon hydrogen) standard for the carbon intensity applied in the clean hydrogen certification standard following the calculation methodology presented by IPHE (International Partnership for Fuel cells in the Economy).</li> </ul> </li> </ul>



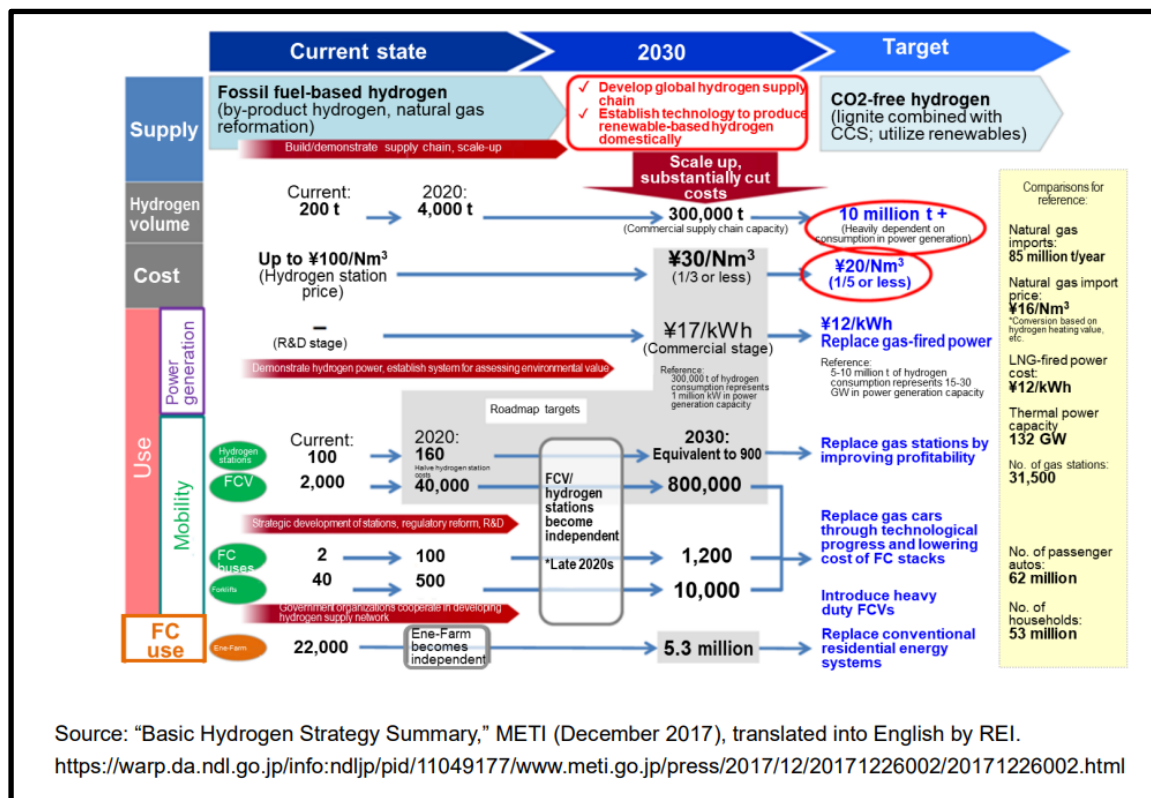


Figure 1.10 Scenario in the basic hydrogen strategy by Japan[15]

Korea as well, inspired by the international criteria for hydrogen certification, is currently developing its own. Following the figure, it can be clearly seen that each country criteria are unique however, the global clean hydrogen/low carbon hydrogen certification criteria are:

1. CO<sub>2</sub> threshold,
2. rules for emissions measurement,
3. system boundary,
4. suitable raw material production path,
5. included CO<sub>2</sub> footprint considering 1,2 and 3[16].

### 1.3 Korea Renewable Energy Production

Due to a lack of available domestic resources, about 98% of South Korea's fossil fuel demand is met by imports. South Korea depends on tanker supplies of liquefied natural gas (LNG) and crude oil to meet demand because it lacks international pipelines for either natural gas or oil. In July 2020, South Korea unveiled its Green New Deal as a component of a broader economic program. The program is to assist South Korea in reaching its targets for reducing greenhouse gas (GHG) emissions and raising the capacity of renewable energy generation. The plan also urged for increasing the use of renewable energy, improving the energy efficiency of the electrical infrastructure, and preparing

the economy for the transition to a decentralized and low-carbon energy supply. Putting money on developments in the green industry[17]. The new and renewable energy industry's sales volume in 2021 was around 29 trillion KRW, up 13.6% from the previous year, according to the Korea Energy Agency (KEA). The number of companies in related industries, such as the construction, manufacturing, power generation, and services of renewable energy products, was 110,000, up 31.7% year over year, and the number of employees reached approximately 140,000, up 19.4% year over year. These figures demonstrate the continued growth of Korea's new and renewable energy industry. These investments cover several categories including manufacturing, construction, power generation and heat supply and services[18].

According to[19] the goal of RE100, a "voluntary initiative," is to increase demand for renewable energy through industry participation, independent of national policies and laws. Furthermore, South Korea has been implementing the Renewable Portfolio Standard (RPS) system from 2012. One of the primary legislative instruments to encourage the use of renewable energy is the RPS program. Based on the New and Renewable Energy Act, it establishes an annual required renewable energy ratio for power generators and integrated energy operators that surpasses a specific threshold for generation capacity. In conclusion, the demand for renewable energy in the country is primarily driven by two factors: (1) businesses who engage in voluntary programs to get renewable energy, and (2) power providers that must fulfill their legal RPS obligations.

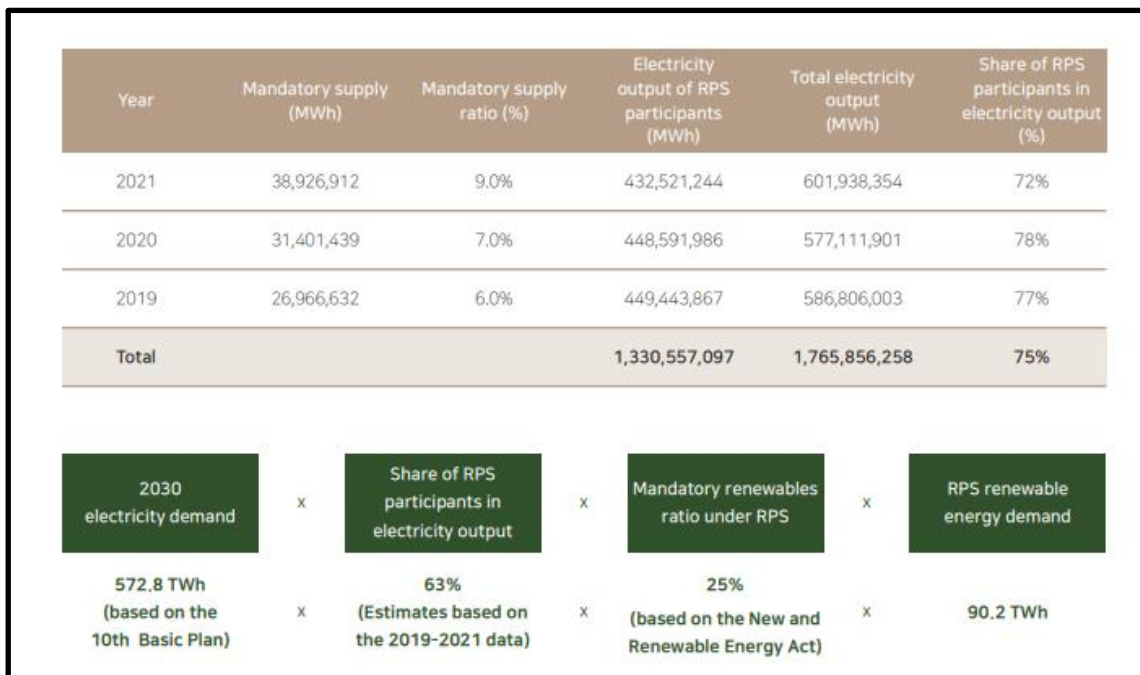


Figure 1.11 The calculation method of the share of RPS participants in electricity output and the estimated demand[19]

## 1.4 Waste in Korea

In South Korea, managing waste is minimizing waste generation and making sure that as much waste is recycled as possible. This covers handling, moving, and getting rid of the waste that has been gathered. The Environmental Protection Law (1963) and the Filth and Cleaning Law (1973) were superseded by the Waste Management Law (1986) in South Korea. With regard to South Korea's waste hierarchy, or the three "R's," this new law sought to decrease general waste. The Waste Management Law established a waste charge structure that is based on volume and is applicable to garbage generated by both industrial and residential sources (also known as municipal solid waste)[20].

According to the Ministry of Agriculture, Food and Rural Affairs in 2021, South Korea produced 52,560 tons of animal waste or livestock manure (South Korea the Ministry of Agriculture, Food and Rural Affairs, 2022)[21]. Simultaneously, food waste produced in the same year was 3,527,561 tons and total Municipal Solid Waste (MSW) is 7,032,662 tons (KOSIS, 2023)[22]. Methane emissions data in Korea were provided in 2020 by the Ministry of Environment, Greenhouse Gas Inventory, and Research Center of Korea[23]. The data indicated that 656.2 million tons of GHG emissions were caused in Korea overall. Of this total, 21.1 million tons (3.21%) are produced by the agriculture sector, while 16.7 million tons (2.54%) are garbage. CH<sub>4</sub> makes up only 27.1 million tons, or 4.13%, of South Korea's total GHG emissions, compared to CO<sub>2</sub>'s 599.8 million tons, or 91.4%, (Green Technology Development Division, 2023)[24].

More than 90% of high-carbon organic wastes, such as animal dung, sewage sludge, and food wastes, were handled in less complicated ways. But waste materials can also be used to produce biogas, which is an energy source. The Korean Ministry of Environment intends to use biomass gasification plants to recycle organic waste that has a high potential for energy production. The present number of biomass gasification facilities, 110, will be extended to 140, potentially increasing production by about 40% [23].

## 1.5 Research Background

Following Korea's hydrogen economy Roadmap: the outline's goal of producing 6.2 million fuel cell electric vehicles and rolling out at least 1,200 refilling stations by 2040[25].

More than 90% of high-carbon organic wastes, such as animal waste, sewage sludge, and food waste, were handled in less complicated ways. Nevertheless, biogas is another energy that can be produced from waste materials. The present number of biomass gasification facilities, 110, will be extended to 140, potentially increasing production by about 40% [26].

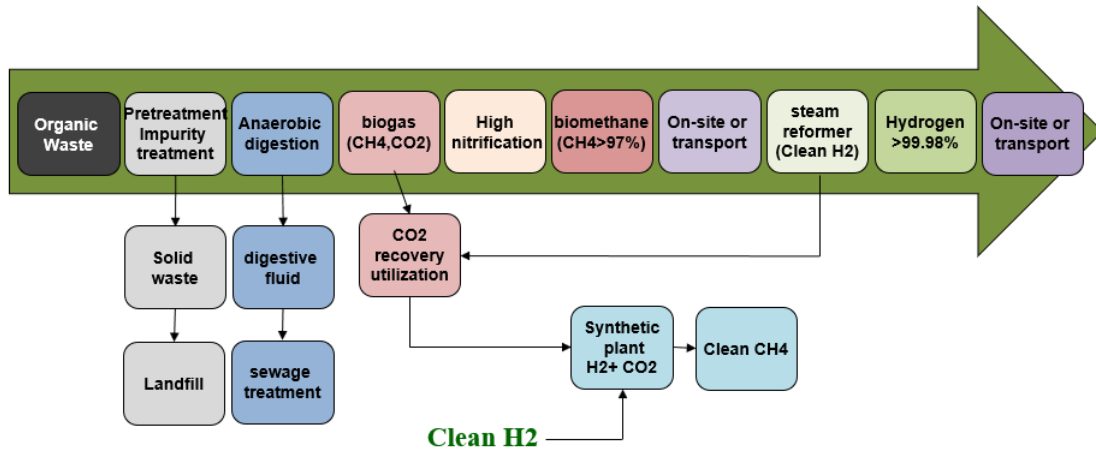


Figure 1.12 Clean hydrogen production path based on biomass

As part of a service offered by the Ministry of Environment, an assessment of greenhouse gas emissions, analyzing clean hydrogen utilizing biogas raw materials in Korea was carried out using the GREET model mentioned above. This work is implementing life-cycle assessment (LCA) which is to conduct an environmental assessment of the entire process, including production and transportation of fuel used in shipping as well as fuel used in operation. LCA was developed in the 1970s to compare and evaluate the eco-friendliness of a product and then has become global. LCA is a technique used by many companies and research institutes. The environmental problems arising from each product process can be analyzed and evaluated, and comparisons between different production operations can be made.

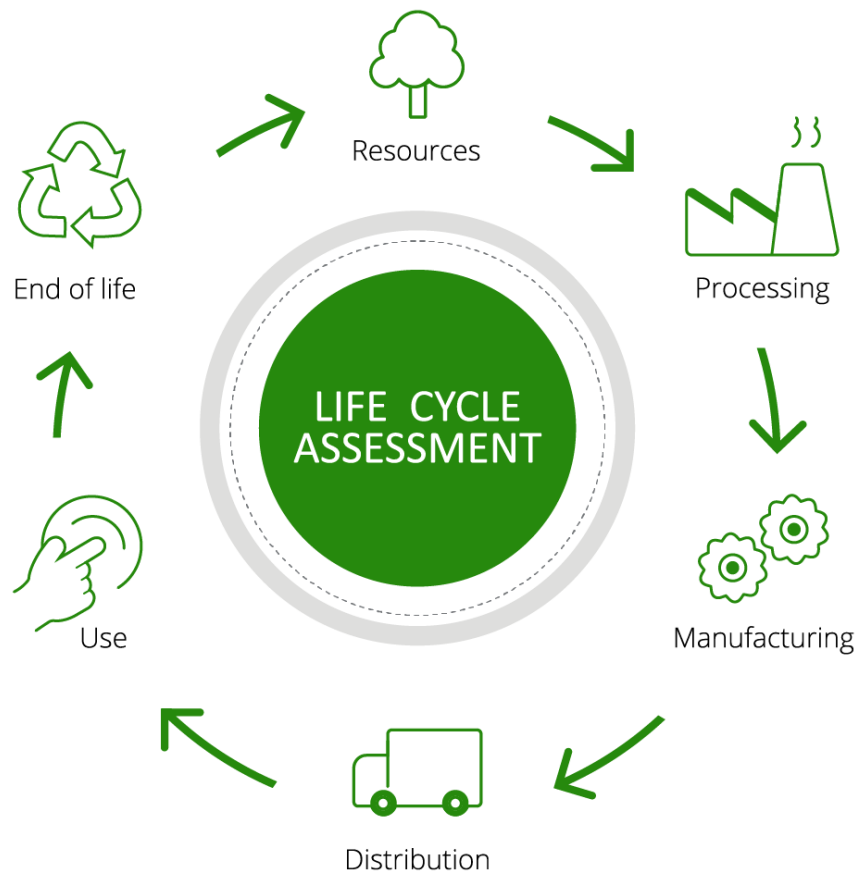


Figure 1.13 LCA diagram[27]

REET (Greenhouse gases, Regulated Emissions, and Energy use in Technologies) was developed by the Department of Energy's Argonne National Laboratory at the request of the United States Department of Energy to carry out an environmental impact assessment on the Well to Wheel (WTW) process, which involves producing fuel and using it to power vehicles.

The federal government and the state of California in the United States are using REET internationally accepted life-cycle evaluation model[28] the figure below show REET model by Argonne National Laboratory.

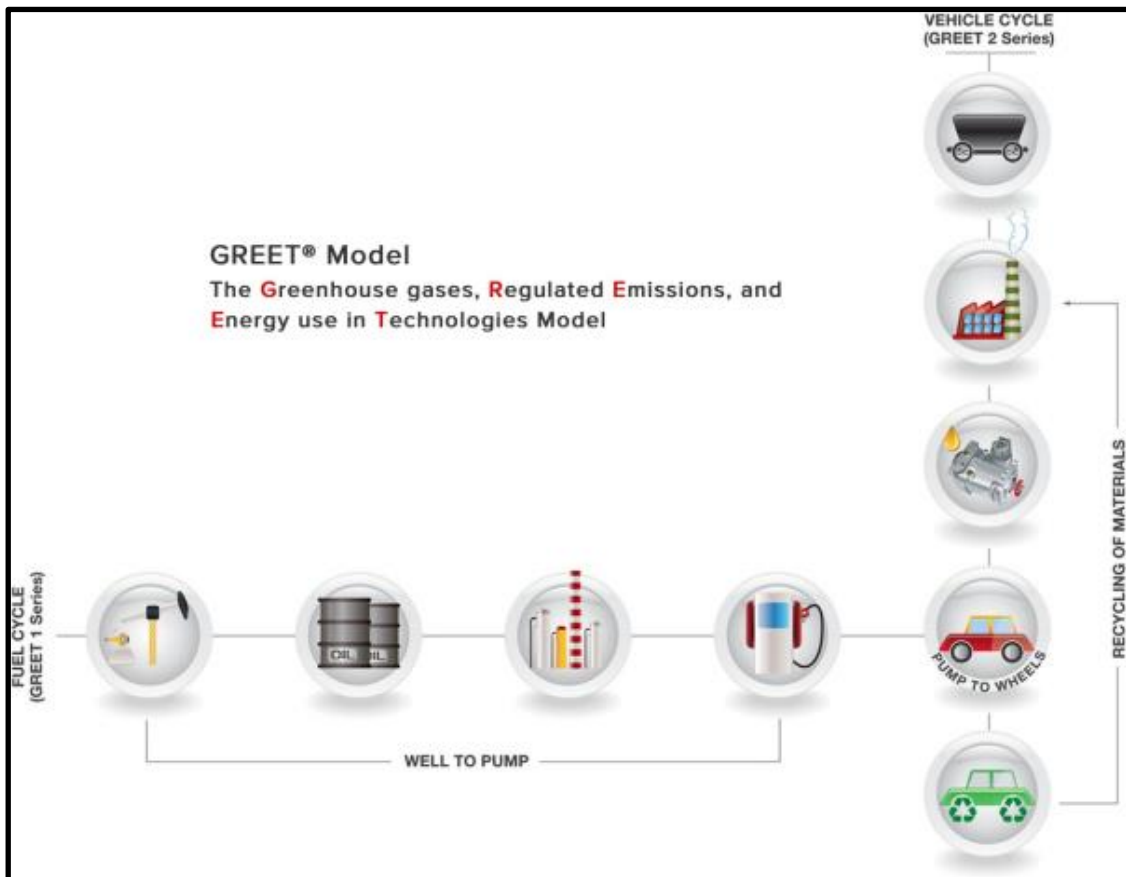


Figure 1.14 Argonne National Laboratory's GREET Model[29]

GREET is an open-source software, available online, is an Excel-based calculating simulation that can analyze the environmental impact ( $\text{CO}_2$  footprint,  $\text{kgCO}_2/\text{kgH}_2$ ) of the complete process, including the gathering of fuel raw materials, transportation, product manufacture, and ultimate consumption. On December 23, 2023, following the U.S. federal government's IRA. The California state government recognized the GREET statistics as a clean hydrogen certification program. These figures are used to quantify fuel greenhouse gas emissions from WTW in the transportation sector. Following the Department of energy[30] for any given energy and vehicle system, GREET can calculate:

- Total energy consumption (non-renewable and renewable)
- Fossil fuel energy use (petroleum, natural gas, coal)
- Greenhouse gas emissions
- Air pollutant emissions
- Water consumption.

Following figure 1.14, the purpose of GREET Excel 2 is to analyze the energy consumption and emissions related to the lifecycle of a vehicle, starting from raw material recovery and ending with disposal and recycling. Because GREET offers such a broad spectrum of vehicle technologies, it is particularly useful for comparing, assessing, and comprehending the various effects that a vehicle's lifetime might have on the energy consumption and emissions of a fuel cycle as a whole. GREET 1 and GREET 2 were designed to be used in tandem to fully model transportation and lifecycle energy emissions. While GREET 1 can be used to accurately simulate two wheels of outcomes for a fuel pathway, GREET 2 can be used to include specific vehicle factors that aren't covered in GREET 1. GREET 1 models the fuel cycle, while GREET 2 models the vehicle cycle.

Many energy feedstock sources and processes, including biomass gasification, water electrolysis, and steam methane reforming (SMR) of natural gas (NG) or biogas, can be used to create H<sub>2</sub>. Moreover, H<sub>2</sub> is a byproduct of the liquid cracking of NG and C-A. The carbon intensity (CI) of H<sub>2</sub> at various points in its value chain can vary significantly, depending on the energy source used for H<sub>2</sub> production, packaging, and transportation. Figure 1.15 shows one of the bases that this work employed for the achieving the life cycle assessment of several comprehensive hydrogen production processes. Other references were also considered as per the upstream and downstream of the process for specific waste like Lee et. al [31].

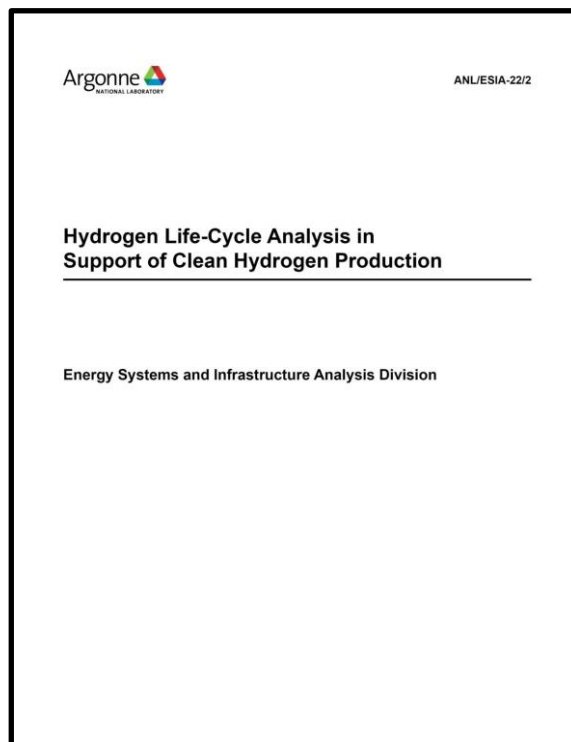


Figure 1.15 Hydrogen Life-Cycle Analysis in Support of Clean Hydrogen Production Study[32]

## 1.6 Research Objectives

The main goal of this study relies on derivation of a Korean-style hydrogen full-cycle analysis technique through the latest overseas(U.S) life-cycle analysis techniques aiming to:

- To present the life cycle resulting in GHG emissions of Hydrogen and RNG production
- To analyze the entire process of hydrogen production using SMR or LFG
- To compare various waste management techniques with RNG or Hydrogen production lines by examining the effects of selecting a combination of waste raw materials and conversion technology
- To evaluate the environmental impact of hydrogen production using SMR or LFG for domestic hydrogen production, using GREET to compare and inspect the Well to Gate (WTG) process considering:
  1. In the case of LFG gas, it is expected that a lot of greenhouse gases will be generated due to the amount of leakage found during the production and processing.
  2. Considering the energy loss due to removal of sulfur contained in LFG.
  3. Based on a conducted case study in the United States first, this work is comparing the U.S research based on SMR in accordance with domestic circumstances.
- To ensure high accuracy for the entire cycle process by calculating energy process

## 1.7 Thesis Outline

This thesis consists of five chapters: Introduction, Literature view, Methodology, Results and discussion and, Summary and conclusion respectively.



## 2 LITERATURE REVIEW

### 2.1 Problem Definition

Numerous Life cycle assessment (LCA) studies quantify the environmental effects of a single waste type while almost no LCA study has yet been conducted that thoroughly compares different waste raw materials. Comparing the findings of earlier studies is the challenge of LCA since every study has a unique set of assumptions, boundary systems, and goals.

A consistent basis allows for fair and equal comparisons when selecting different raw material/product combinations, guaranteeing that the impact of differences in results is consistent and comparable. Aside from that, careful consideration must be given to the advancement of hydrogen generation, as it is still in its infancy in South Korea.

This study illustrates the greenhouse gas emissions life cycle associated with the production of hydrogen and RNG to understand the trade-offs involved in producing RNG and hydrogen, as well as the resulting advantages for the environment.

Starting with the conversion process and finishing with the ultimate product, hydrogen, the study poses the question of whether the consequences of choosing a blend of waste raw materials allow comparison with different waste combinations of the process's resulting carbon index. A combination of four waste feedstocks is used in each step.

### 2.2 Life Cycle Assessment Principles and Frameworks

LCA evaluates a product's environmental effects at every stage of production, from the procurement of raw materials to disposal. It offers guidance on environmental improvement options and assists with decision-making. The ISO standard places a strong emphasis on [33]LCA's iterative nature, transparency, and data quality. Critical review procedures guarantee the methodological soundness and reliability of life cycle assessment research. The publication offers a thorough manual for carrying out and documenting life cycle assessments. Depending on the purpose of a given LCA study, the scope and depth of the research can vary significantly. Nonetheless, the guidelines and structure outlined in this International Standard must be adhered to in every situation.

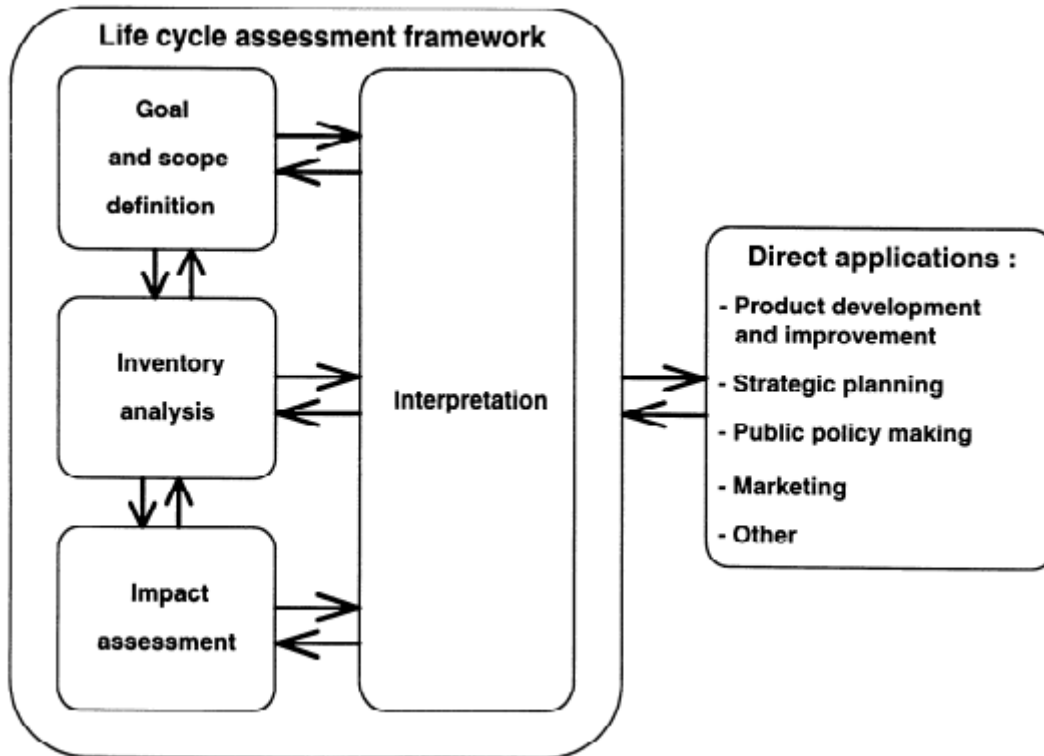


Figure 2.1 Phase of an LCA[33]

Several factors determine the system boundaries, including: the intended application of the study, the assumptions made, cut-off criteria, data and cost constraints, and the intended audience. When analyzing the life cycle assessments (LCAs) of the various hydrogen pathways, the system boundary encompasses the natural resources consumed, utilities, pertinent waste, products, and co-products. Rectangle forms represent downstream operations, while parallelogram shapes represent upstream emissions data.

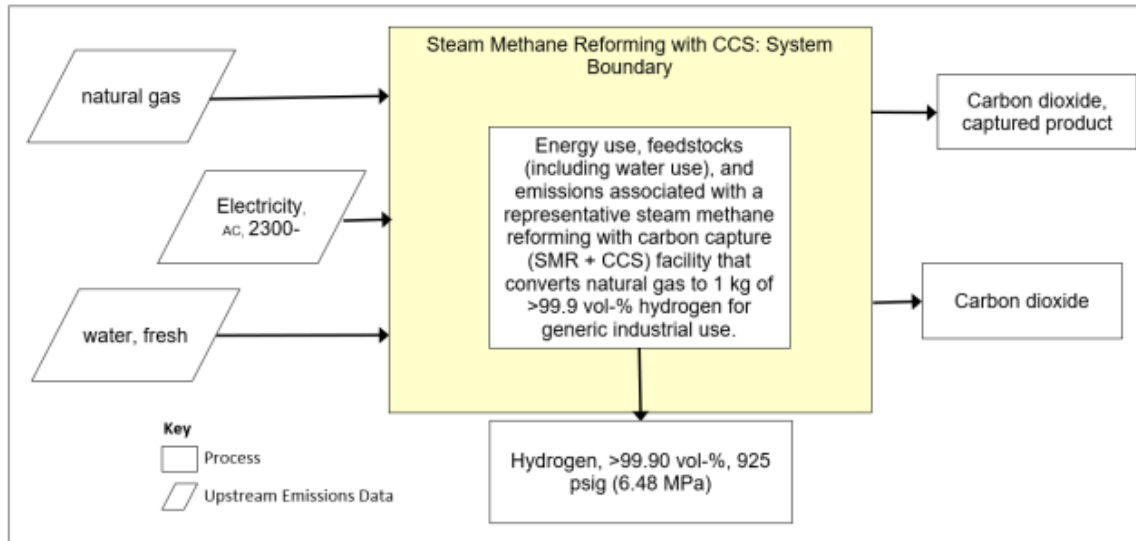


Figure 2.2 system boundary, steam methane reforming plant without CO<sub>2</sub> capture[34]

### 2.3 Life-cycle Analysis in Support of Hydrogen Production in Korea

The scope of clean hydrogen and certification information are detailed in an administrative notice of the Proposed Notification on the Operation of Clean Hydrogen Certification System (the "Proposed Notification"), which was released on December 19, 2023, by the Ministry of Trade, Industry and Energy (the "MOTIE") [35]. Prior to the November 30, 2023, amendment of the Hydrogen Economy Promotion and Hydrogen Safety Management Act (the "Hydrogen Act"), the MOTIE only provided a broad definition of "clean hydrogen certification standards" and left out more specific and technical details that should have been covered in the notification.

The Sixth Hydrogen Economy Committee meeting, which took place on December 18, 2023, also covered the operation of the Clean Hydrogen Certification System, which was mentioned in the proposed notification. The clean hydrogen greenhouse gas emissions standard under the proposed notification is "4kgCO<sub>2</sub>eq/kgH<sub>2</sub>," and it is divided into four grades according to the actual quantity of greenhouse gas emissions as follows:

Classification	Grade 1	Grade 2	Grade 3	Grade 4
Certification Standards (Emissions) (Unit: kgCO <sub>2</sub> eq/kgH <sub>2</sub> )	0.00 - 0.10	0.11 - 1.00	1.01 - 2.00	2.01 - 4.00

Figure 2.3 The clean hydrogen greenhouse gas emissions standard of the Sixth Hydrogen Economy Committee meeting[36]

Greenhouse gas emissions are further broken down for the Life Cycle Assessment (LCA) of hydrogen into three categories: direct emissions (Scope 1), indirect emissions (Scope 2), and other indirect emissions (Scope 3)[36].

## 2.4 Previous Works

In [37] order to reduce uncertainties in predicting landfill gas (LFG, primarily methane) emissions from base case landfills, a WTE life-cycle analysis (LCA) of their greenhouse gas (GHG) emissions was carried out. This study discovered that the climate and LFG management techniques have a substantial impact on landfill GHG emissions. Since LFG generation displaces regional electricity, it indirectly lowers GHG emissions in life cycle assessments (LCAs) of WTE conversion. Not all of the LFG produced along this route can be collected; some of it escapes through landfill covers and is released into the atmosphere. A fraction of the  $\text{CH}_4$  that is not collected oxidizes into  $\text{CO}_2$  as LFG passes through landfill covers. In conclusion, part of the  $\text{CH}_4$  formed from landfilled waste is burned or oxidized into  $\text{CO}_2$ , even if the  $\text{CO}_2$  generated is released without changing into other molecules, regardless of LFG collection conditions.

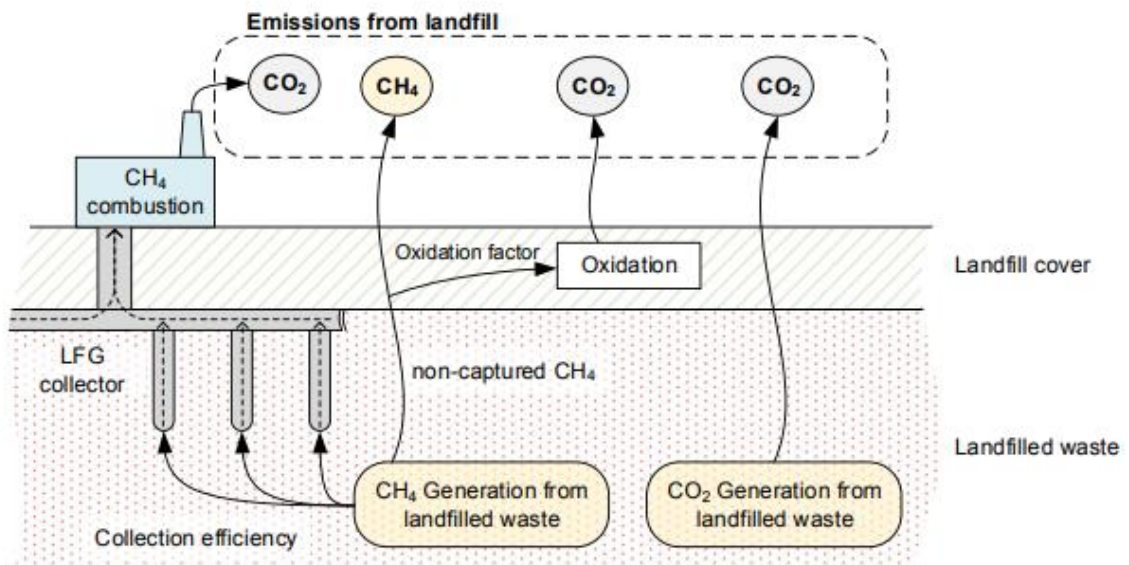


Figure 2.4 Fate of LFG emissions generated from landfilled organic waste[37]

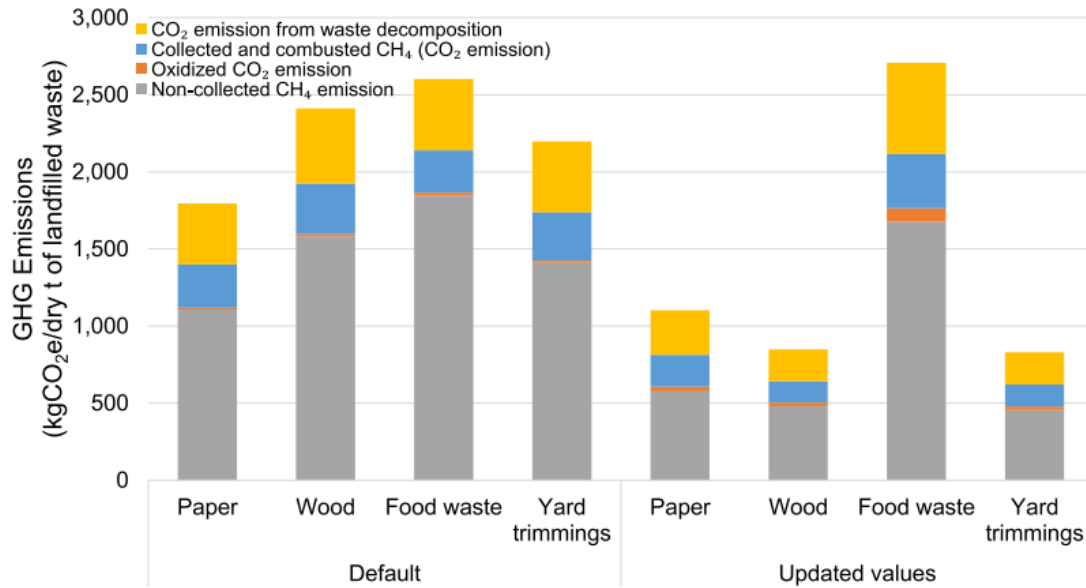


Figure 2.5 The fates of carbon from four types of landfilled wastes using default and updated parametric values for fraction of degradable materials to be decomposed and the oxidation factor of CH<sub>4</sub> from landfills[37]

As stated in [38], to ascertain if renewable gas paths satisfy the GHG reduction standards for eligibility in the REDII, the EU employs life-cycle analysis, or LCA. Due to the complexity of life cycle GHG analysis, variations in methodology, data inputs, and assumptions might determine whether a renewable gas pathway qualifies for REDII eligibility at the 50% to 80% GHG reduction threshold. In order to guarantee that policy only supports gas paths consistent with a vision of deep decarbonization, it is crucial that European policymakers employ robust life cycle assessment (LCA). The short- and long-term trends in climatic performance vary amongst paths. Specifically, the near- and long-term results of biomass gasification and electrolysis from renewable or grid electricity are not different; wastewater sludge and manure perform better in the short term; landfill gas, natural gas, and coal likely have limited potential to reduce carbon emissions in the near future. Notably, there would be a 15% increase in the greenhouse gas intensity of fossil fuel-based hydrogen in the near future.

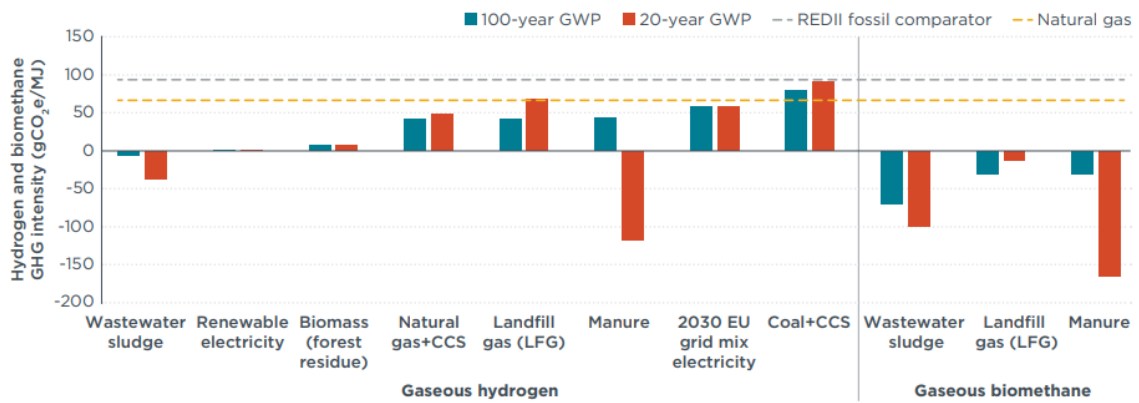


Figure 2.6 GHG intensity of gaseous hydrogen and biomethane pathways of the central case using[38]

## 2.5 GREET Software

Analyses can be performed using GREET “Greenhouse gases, Regulated Emissions, and Energy use in Technologies”, previously the “T” stands for Transportation but since now the model scope goes beyond Transportation. GREET, which was developed in Argonne National Laboratory and is sponsored by the U.S Department of Energy (Office of Energy Efficiency and Renewable Energy) in the well-known Microsoft excel platform is capable of LCA mission. For representing data a wide range of materials including collection/transportation of raw materials and operation. GREET as a comprehensive program, contains actual measurement results, not only simulation results. It also provides available data for gasoline, diesel, natural gas, electricity, and biofuel for transportation. It also allows for investigating an entire process for the energy usage along with exhaust gases and greenhouse gas emissions, GHG. Since the first version was released in 1996, it has been continuously updated reaching GREET2022 that can now let on most reliable and up-to-date data. GREET includes over 100 fuel pathways, among them electricity generated from different energy sources. The process from production to supply is illustrated in detail in which each step that occurs during calculation is easy to compare and analyze to which impact related to it.

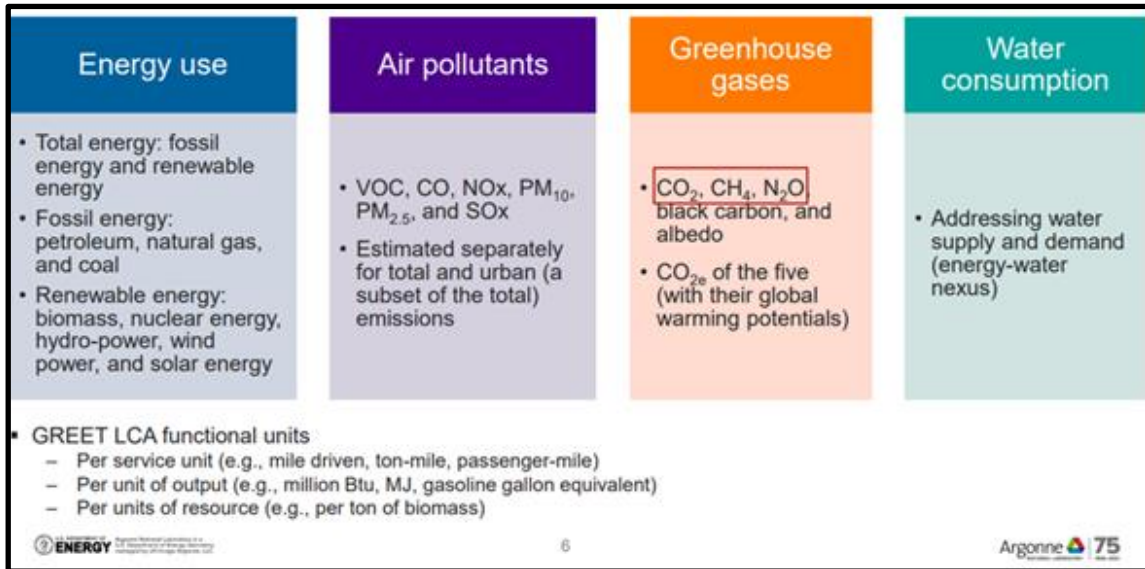


Figure 2.7 GREET sustainability matrices [32]

## 2.6 Carbon Capture, Utilization and Storage, and Sequestration

CO<sub>2</sub> is captured as part of Carbon capture, utilization and storage CCUS, usually from large point sources such as industrial sites or power plants that run on biomass or fossil fuels. When not in use immediately, the compressed CO<sub>2</sub> is either injected into deep geological formations like saline aquifers or depleted oil and gas reservoirs, or it is transported by pipeline, ship, rail, or truck for use in a variety of applications. While the technique of extracting and storing carbon dioxide from the atmosphere is known as carbon sequestration. It is one way to lessen global climate change by lowering the atmospheric concentration of carbon dioxide. Existing industrial and electricity plants can be retrofitted with CCUS to enable continuous operation. It can address pollutants in businesses that are difficult to regulate, especially heavy industries like chemicals, steel, and cement. The least-cost low-carbon hydrogen production made possible by CCUS can help decarbonize other energy system components like transportation, industry, and ships. Lastly, CO<sub>2</sub> removal from the atmosphere is a capability of CCUS that helps balance emissions that cannot be avoided or are technically challenging to reduce[39][40].

## 3 METHODOLOGY

### 3.1 Initial Conditions Parameters and Sourcing

This work utilizes the latest recent life-cycle analysis method from the United States, resulting in a Korean-style hydrogen life-cycle analysis technique based on domestic data. The clean hydrogen certification method being prepared by the Ministry of Trade, Industry and Energy [35] will basically use GREET 2022 of the Argonne National Laboratory taking into consideration that some content will be changed. However, the details of GREET 2022 have not been released to the outside, so there are very few opinions from industry.

It was confirmed in Chapter 2 that international standards for clean hydrogen certification differ by country[41]. Since Korea uses the U.S. life cycle assessment program, comparison with U.S. standards is important. In the U.S. clean hydrogen, both biogas and water electrolysis were calculated based on the standards for being recognized as clean hydrogen.

In the case of the WTG process, it is still difficult to obtain reliable data in Korea and there is still a lack of extensive data on each process. To solve this problem, it was intended to conduct comparative analysis using the results of GREET from the United States as shown below and conduct this work to apply them to the Korean situation. Data required for Well to Gate analysis are listed as:

1. Biomethane production: Energy efficiency, process fuel ratio and energy efficiency of the raw material extraction process
2. Transportation distance: Various transportation distances from fuel production sites to domestic biomethane usage sites
3. Hydrogen production plant: Energy efficiency of the hydrogen production plant, process fuel ratio, and amount of water treatment
4. CCS or CCU: Method of processing the generated CO<sub>2</sub>
5. Liquid or high-pressure storage: amount of energy used, greenhouse gas emissions, and leakage during low temperature storage.
6. Technology used: Proportion of combustion methods to create energy in each process
7. Other matters: operation of desulfurization equipment, etc.

### 3.2 RNG Pathways

#### 3.2.1 Hydrogen Production Process from Each Waste

In this study four RNG pathways are considered: Landfill Gas, Food waste, Animal waste and sludge. The figure below represents each one of these four pathways. It is noted that food waste pathways require a greater number of stages than other pathways. Animal waste and sludge pathways both demands quite the same stages after the waste source first step. The major difference between (a) pathway and others lies in the anaerobic digestion stage that happened naturally in an uncontrolled manner. Therefore, significant uncontrolled amount of natural gas will not be captured affecting the



overall GHG emissions resulting from (a). Other pathways however consist of the anaerobic digestion step preceded by waste preparing steps like sorting and shredding for the municipality food waste pathway (b). The anaerobic digestion process in (b), (c) and (d) is impacting two outcomes: the biogas and the digestate produced which will be transported to the land fill. Some resulting digestate can be used as bio-fertilizer in farming. Both (c) and (d) pathways are typically the same except for the first step which indicates the waste type. The resulting biogas is then upgraded and purified so that it can be used as a raw material to produce gaseous hydrogen for all pathways.

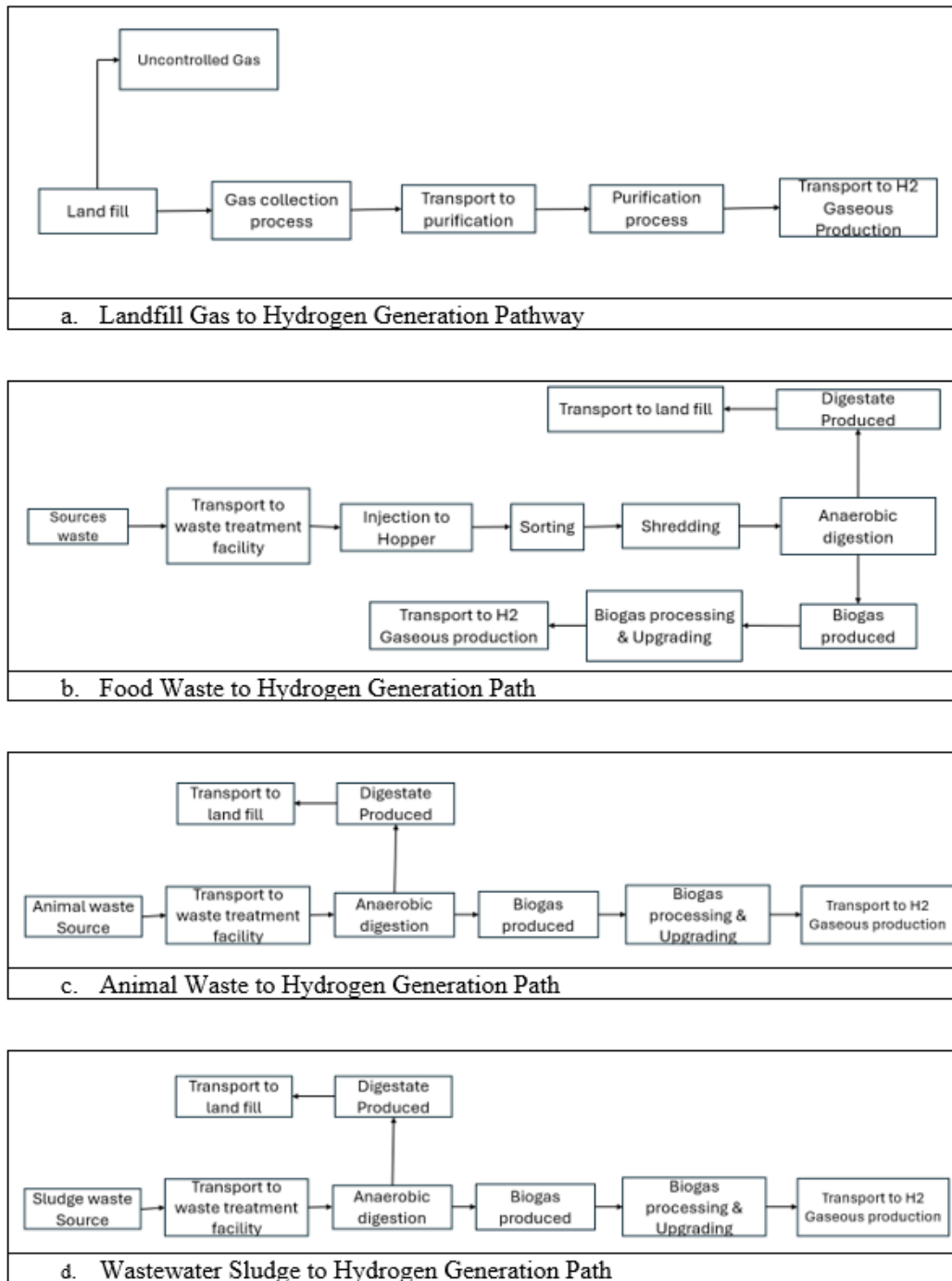


Figure 3.1 Hydrogen generation path by waste type

### 3.2.2 Initial Input Value

For this work input data were taken from a few resources that were adapted. These sources can be basically classified as sources conducted by Argonne national laboratory and others conducted by a Korean company A using A's own developed UNISIM model. These data are illustrated with, pointing to its references, in the table below:

Table 3-1 GREET model initial input value

Parameter	Unit	Value	Source
<b>Landfill Gas</b>			
Uncontrolled Emission	%	25	2010. Mitz et al. Well-to-Wheels Analysis of Landfill Gas-Based Pathways and Their Addition to the GREET Model[42].
Gas Collected	%	75	
Efficiency Natural Gas Production	%	91-97	
Generator Efficiency	%	28-44	
<b>Food Waste</b>			
Moisture Content	%	72	2011. Jeongwoo Han et al. Waste-to-Wheel Analysis of Anaerobic-Digestion-Based Renewable Natural Gas Pathways with the GREET Model[43]
Methane yield of AD	kg CH <sub>4</sub> /kg C input	0.526	
CO <sub>2</sub> Emission of AD	kg CO <sub>2</sub> /kg C input	0.863	
Volatile Solid/Total Solid	%	63	
Digestate or Solid Waste Transportation Distance	mile	40	
Carbon sequestration of digestate	%	20	
Heat Load Share by CHP	%	100	
Biogas Share to CHP	%	31	
<b>Animal Waste</b>			
Animal Waste Transportation Distance	mile	3	2011. Jeongwoo Han et al. Waste-to-Wheel Analysis of Anaerobic-Digestion-Based Renewable Natural Gas Pathways with the GREET Model [43]
Animal Waste Moisture Content	%	88	
Wet Animal Waste Input	Ton/mmBtu	1.59	
MCF of Anaerobic Digester	%	81.7	
Electricity required for AD	kWh/mmBtu biomethane	19	

Heat Required for AD	Btu/mmBtu biomethane	183,933	
MCH of AD Residue	%	0.2	
Direct N <sub>2</sub> O Emission factor	kg N in N <sub>2</sub> O/kg N	0	
<b>Wastewater Sludge</b>			
Volatile solid to total solid ratio of biosolids to digester of AD	kg VS/kg TS	0.61	2016. Uisung Lee et al. Lifecycle Analysis of Renewable Natural Gas and Hydrocarbon Fuels from Wastewater Treatment Plants's Sludge[31]
Biogas Production Rates of AD	m <sup>3</sup> biogas/kg VS destroyed	0.9	
Methane Share in Biogas of AD	% vol	65	
<b>NG Processing Assumption</b>			
CHP Generator Electrical Efficiency	%	33	Company A's SMR UNISIM Model, section 3.5,3.6
Heat Recovery Eff. from CHP Gen.	%	70	
Boiler Efficiency	%	80	
NG Processing CH <sub>4</sub> Leakage	%	2	
<b>Share of Fuel from Each Waste Assumption</b>			
Landfill Gass	%	100	GREET user input data
Biogas from AD of Food Waste	%	0	
Biogas from AD of Animal Waste	%	0	
Biogas from AD of Wastewater Sludge	%	0	

### 3.3 GREET LCA Scope and Boundaries

The GREET model, developed in Microsoft® Excel with a graphical user interface, is structured to systematically describe a variety of potential feedstocks, fuels, and conversion processes for the WTG/WTW pathways defined within the program. GREET calculates total energy generated from energy consumption sources such as fossil fuels, oil, natural gas, and coal during the LCA process, as well as three greenhouse gases (carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O)), 6 pollutants (volatile organic compounds (VOC), carbon monoxide (CO), nitrogen oxides (NO<sub>x</sub>), sulfur oxides (SO<sub>x</sub>), fine dust (PM<sub>10</sub>), and ultrafine dust (PM<sub>2.5</sub>)). Figure 3.2 shows the GREET LCA home screen in which all these ready pathways and calculations are available inputting some parameters.

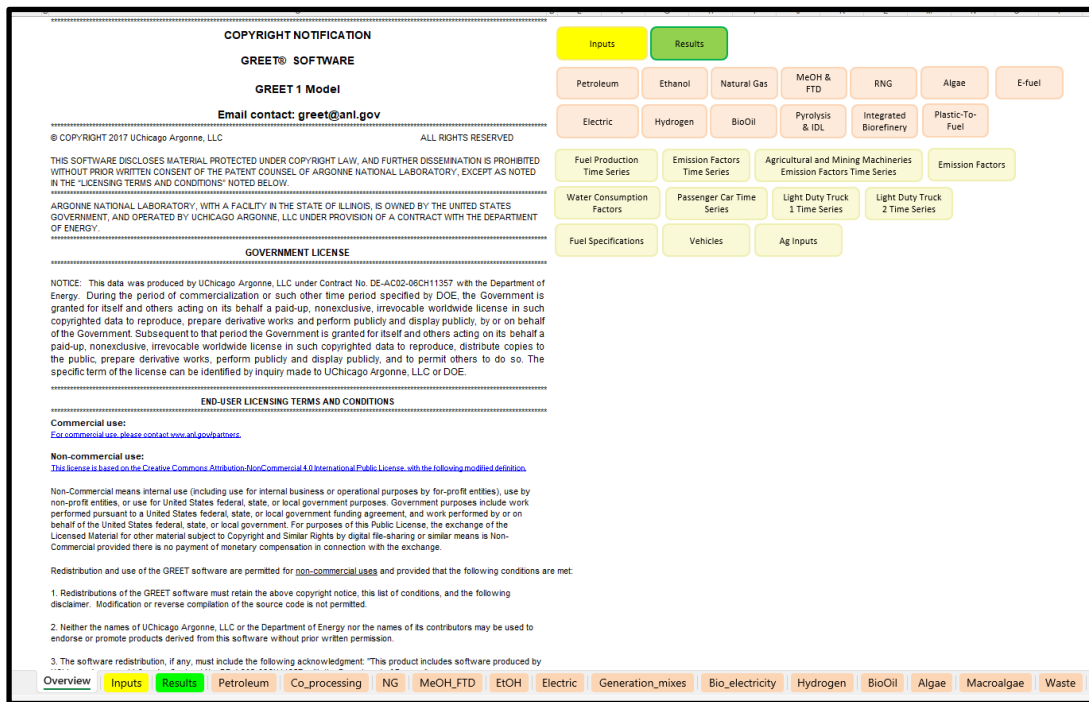


Figure 3.2 GREET LCA home screen

Understanding the impact of fuels on energy usage and emissions requires a life cycle assessment (LCA) that systematically accounts for energy use and emissions at all stages of fuel production and use. Types of processes considered in LCA include raw material acquisition, transportation and processing, product manufacturing, distribution, use, and disposal or recycling. The fuel cycle generally includes feedstock recovery and transportation, fuel production, fuel transportation and distribution, and combustion as an end use. The stages from exploration and recovery of crude oil (well) to transportation and production of products (in the case of this work, hydrogen) are collectively referred to as Well-to-Gate (WTG), and the process up to the actual use of the produced products is called Well-to-Wheel (WTW). In this study, LCA was performed within the boundary from: the raw material recovery stage for fuel (hydrogen) production corresponding to the WTG (Well-To-Gate) range to fuel production (hydrogen). The LCA path considered in this study is as shown in the figure:

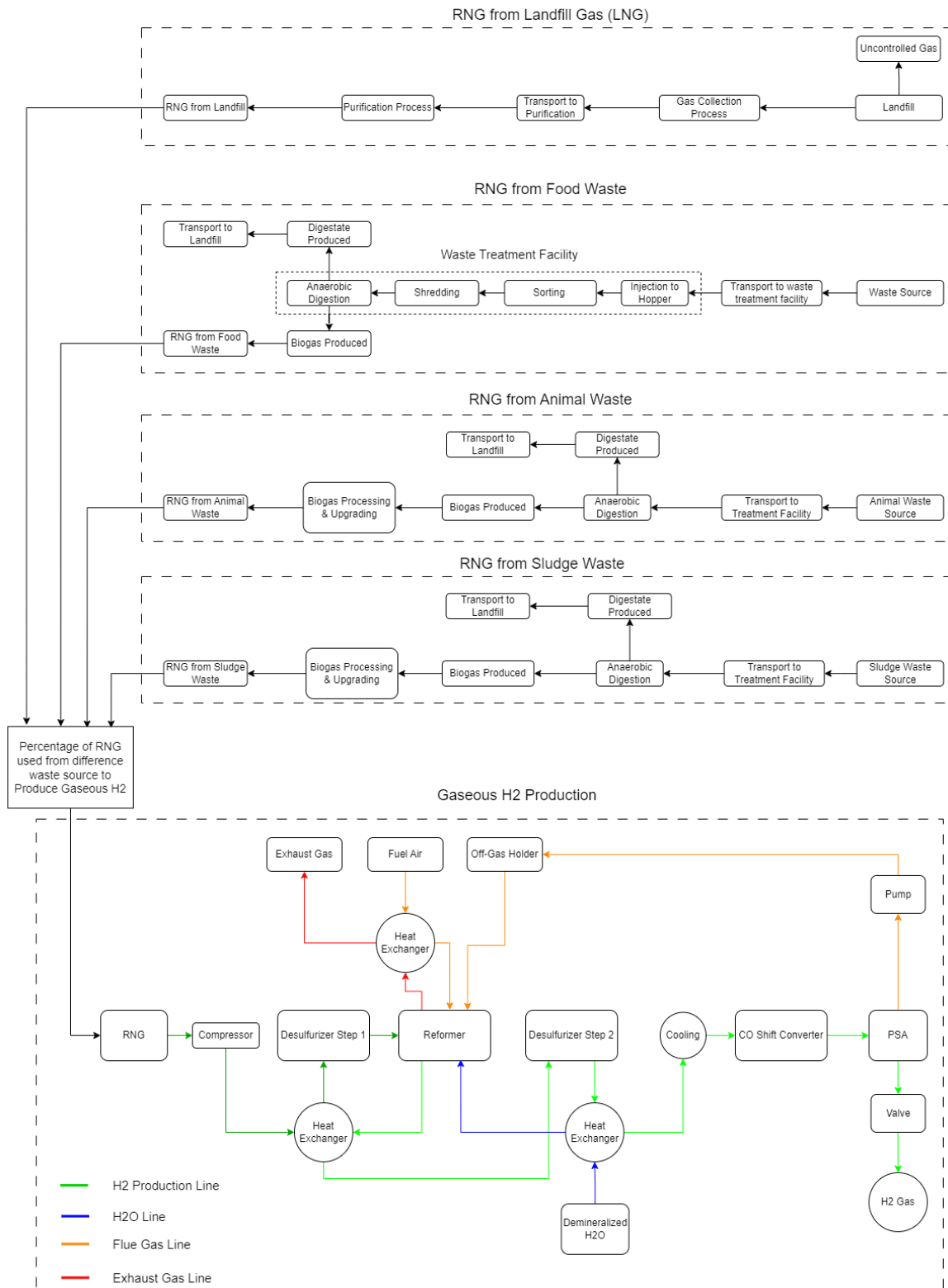


Figure 3.3 Full LCA Process Pathway to Clean Hydrogen Production

### 3.4 GREET Simulation

Referring to [31] and [43] the LFG pathways are built in GREET simulation while AD pathways

for biomass are still remaining. The amount of bio-methane produced from a given feedstock can be given as:

$$CH_{4,Manure} = \sum_S \sum_k B_0 \times MCF_{S,k} \times MS_{S,k} \quad (3.1)$$

Where:

$CH_{4,Manure}$  is the amount of  $CH_4$  produced in  $ft^3/lb$  of volatile solid (VS)

$B_0$  is the maximum methane-producing capacity for manure of a given livestock type in  $ft^3/lb$  of VS

$MCF_{S,k}$  is the methane conversion factor (MCF) for each manure management technology  $S$  by climate region  $k$  in %

$MS_{S,k}$  is the manure share (MS) handled by manure management technology  $S$  by climate region  $k$  in %

MCF(%) OF AD are estimated as:

$$MCF (\%) = \frac{B_0 - [Total CH_4 Emissions]}{B_0} \quad (3.2)$$

For this work the feedstock is considered to be in the same processing place which implies no energy added for feedstock transportation.

Following [31] the total thermal energy requirements can be expressed as:

$$\begin{aligned} E_{Thermal} &= E_{Thermal} + E_{Marginal} = E_{Base} + c_{AD} \cdot \Delta T \\ &= E_{Base} = +(\alpha + \beta \times [sludge\ intake]^{-0.34}) \times (T_{operating} - T_{amb}) \quad (3.3) \end{aligned}$$

Where:  $E_{Thermal}$ ,  $E_{Base}$ ,  $E_{Marginal}$  are total, base and marginal thermal energy requirements respectively

$c_{AD}$  represents the AD specific thermal energy requirement in terms of  $MJ/kg \cdot C^\circ$

$\alpha$  and  $\beta$  are constants as a function of the amount of sludge intake in  $m^3/day$ .

$\Delta T$  represents the difference between the operating temperature  $T_{operating}$  and the ambient temperature  $T_{amb}$

Finally, Total energy can be expressed as:

$$E_{(total)} = E_{(feedstock\ supply)} + E_{(energy\ supply)} \quad (3.4)$$

Where:

$E_{(total)}$  total energy used in feedstock and central plants RNG to produce gaseous H<sub>2</sub>

$E_{(feedstock\ supply)}$  Emission in Feedstock and central plants RNG to produce gaseous H<sub>2</sub>

$E_{(energy\ supply)}$  energy used to operate the plants RNG to produce gaseous H<sub>2</sub>

The process consists of two stages using two GREET-2022 models. Firstly, producing Biogas (Landfill Gas) then, Biomethane/RNG (Renewable Natural Gas) . The parameters for each process are illustrated in the tables below and were later specified through the Korean company A's UNISIM model.

Table 3-2 Parameters required for initial setting in GREET-2022 based SMR modeling

<b>INPUT on SMR System</b>		
<b>No</b>	<b>Process</b>	<b>Input</b>
<b>1. Input Materials</b>		
1	Gas Input	energy/mass/volume
<b>2 &amp; 3 Gaseous H<sub>2</sub> Production from RNG SMR with Steam Export</b>		
1	Water Process	Galon
2	Electricity Use	Btu
3	Gaseous Hydrogen Production in SMR	mmBtu
4	Non-Combustion emission	%
5	Natural Gas combusted in SMR	%
6	CO2 Balance - RNG	%
7	Steam	mm Btu
8	Gaseous Hydrogen loses	%
9	CO2 Captured on SMR	%
<b>4. Option: H<sub>2</sub> production from NG SMR w/CCS or wo/CCS</b>		
1	Gaseous Hydrogen	mmBtu
2	Gaseous Hydrogen loses	%
<b>H<sub>2</sub> production total</b>		
1	Gaseous Hydrogen	mass/volume

Table 3-3 Parameters required for initial setting in GREET-2022 based RNG modeling

<b>Biogas Production</b>		
<b>No</b>	<b>Process</b>	<b>Name of Input Parameter</b>
1	Gas Collection	Total Production
		Electricity source
		Efficiency
2	Purification	Technology used
		Electricity source
		Purification Process Pathway
		Total energy Used
3	Gas Transportation	Total Production
		Distance
		Transportation Mode
		Fuel Share
		Urban Share

Table 3-4 Parameters required for initial setting in GREET-2022 based biogas to RNG modeling

<b>Gass Collection Process</b>		
Emissions	Value	Unit
CO2 Total	0.2257	kg
CH4 Total	-0.0316	kg
Total Energy Used	1055	MJ
Biogas	1055	MJ
<b>Purification Process</b>		
Emissions		
CO2 Total	0.2451	g
CH4 Total	0.3642	g
Total Energy Used	1200	kJ
Biogas	1200	kJ
<b>Total Process (Include Transportation)</b>		
Emissions	Value	Unit
CO2 Total	1.9266	g
CH4 Total	0.4481	g
Total Energy Used	1235	kJ
Biogas	1204	kJ



Table 3-5 Energy required for SMR reforming equipment, energy production, product content, and greenhouse gas emissions

Division	Anaerobic digestion	High Nitrifier	Reformer	
Configuration	Anaerobic digestion tank	membrane method	SMR (hydrogen extractor), 300Nm <sup>3</sup> /hr× 2units	
Energy required	Electricity cost 200 million/year, unit price of electricity (17 years period, 90KRW/kWh) converted to 253.7 kWh	180 kWh	Power	Heat
			172.7 kWh (86.35kWh × 2units)	Biomethane 4~6Nm <sup>3</sup> /hr utilization
Energy production	Biogas 8800m <sup>3</sup> /day	Biomethane 5252 Nm <sup>3</sup> /day	H <sub>2</sub> 1280kg/day (=640kg/day× 2units)	
Product content	60% methane	97% methane water	99.995% and above of H <sub>2</sub>	
GHG (KgCO <sub>2</sub> /Kg-H <sub>2</sub> )	2.17	1.54	1.48	
	Total of 5.19kgCO <sub>2</sub> /kg-H <sub>2</sub>			

Source of required energy must be revealed, which is written based on data on power generation by power generation source in Korea as shown in the figure below:

Electricity generation by source, Korea, 2000-2022

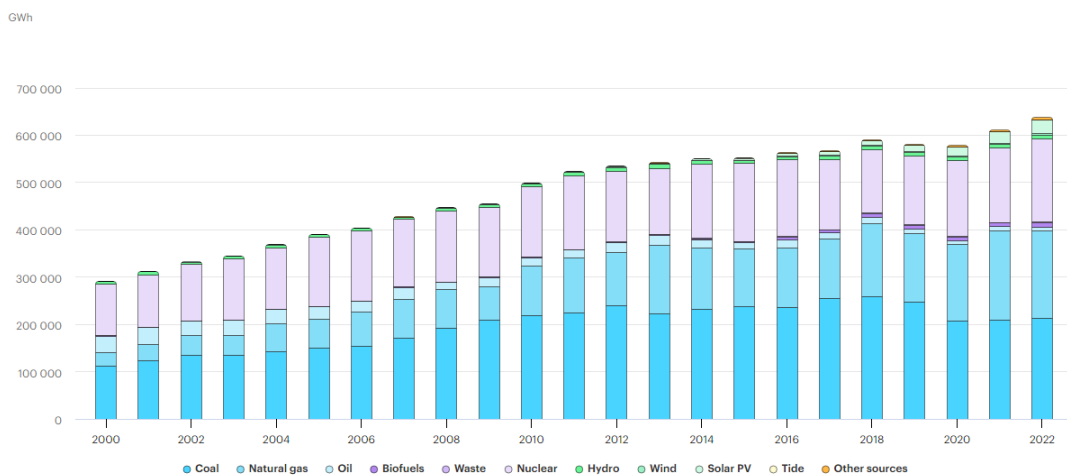


Figure 3.4 Power generation by power generation source in Korea [44]

Table 3-6 GREET initial value input - energy source and power generation amount

Electricity	Source	Security- policy
INPUT	Amount(GWh)	Percentage(%)
Coal	208,112	34.41961391
Natural Gas	188,191	31.12488257
Oil	7,517	1.243235555
BioFuels	7,732	1.278794374
Nuclear	158,015	26.13407825
Hydro	6,737	1.114231466
Wind	3,167	0.523789677
Solar PV	23,591	3.901712116
Other	1,570	0.259662075
Total	604,632	

The downstream process used in GREET simulation performed in this work consists of a few steps:

1. When producing biomethane, removing unnecessary elements from raw gas.
2. Assigning electricity production rate by domestic energy source [%]
3. Assigning 8,800 m<sup>3</sup>Amount of water used for SMR use of biomethane [Liter]
4. Neglecting environmental load resulting from facility construction
5. Considering CCU

To apply calculations in GREET, the value corresponding to “Share of Fuels from Each Waste” required to be entered. In this study, the WTG process produces clean hydrogen from landfill gas, so “Landfill Gas” is set to 100%, and all other energy sources are set to 0%.

1) Scenario Control and Key Input Parameters		Home	Inputs	Results	
1.1) Share of Fuels from Each Waste					
	NG as Intermediate	Fuel	MeOH	CNG	LNG
Landfill Gas		100%	100%	100%	100%
Bio-gas from AD of Animal Waste		0%	0%	0%	0%
Bio-gas from AD of Wastewater Sludge		0%	0%	0%	0%
Bio-gas from AD of MSW		0%	0%	0%	0%

Figure 3.5 Selection of raw material sources for hydrogen production

Considering the SMR system of domestic company A, it is necessary to enter the information below as a “User Defined” value in order to calculate GREET based on domestic actual information. Other information was unreported like:

1. Without steam or electricity export

2. With steam or electricity export (excluding energy in co-products)
3. With steam or electricity export; not electricity export credit; Btu of steam per mmBtu of H<sub>2</sub> produced.
4. With steam or electricity export; net electricity export credit; kWh per mmBtu of H<sub>2</sub> produced.

Table 3-7 Parameters required for GREET calculation

	Source		User Defined	
	Gaseous H <sub>2</sub>	H <sub>2</sub> Production for L <sub>2</sub> H <sub>2</sub>	Gaseous H <sub>2</sub>	H <sub>2</sub> Production for L <sub>2</sub> H <sub>2</sub>
	NETL Model			
Without steam or electricity export	71.8%	71.8%	71.8%	71.8%
With steam or electricity export (excluding energy in co-products)	71.8%	71.8%	71.8%	71.8%
With steam or electricity export; net steam export credit: Btu of steam per mmBtu of H <sub>2</sub> produced	213,343	213,343	213,343	213,343
With steam or electricity export; net electricity export credit: kWh per mmBtu of H <sub>2</sub> produced			14.01	14.01

Remarking that there is no numerical information for the mentioned 4 points as of now, the U.S data set as the (default) value in GREET was used.

The user for GREET simulation simply can implemented following an open-source YouTube video for instance following the source[45]:

The simulation procedure is including:

- 1) Entering the initial input value in the “RNG” sheet in the GREET1\_2022.xlsm file. At this time, the initial value is basically a value that corresponds to the American standards, so it is necessary to find and enter an appropriate value for the purpose of this study. If there is no appropriate data, the initial value already entered in the GREET as (default) is applied.
- 2) In the “Inputs” sheet of the same Excel file, some initial values were entered to suit the purpose of this study.
- 3) After completing the above procedure, the work needs to be properly saved by selecting the appropriate options of “Target Year for Simulation” and “Hydrogen Production Technologies” in the “H2\_User\_Input” sheet of the “GREET\_H2.xlms” file, then selecting the related “Process Inputs” and “Process Outputs” by entering data and performing calculations.
- 4) Finally, the result can be checked corresponding to Carbon Index (CO<sub>2</sub>-kg/H<sub>2</sub>-kg)

### 3.5 UNISIM Hydrogen Production Simulation

In this work, the LCA simulation model using UNISIM was operated, which refers to company A's technology holdings. After the high nitrification process, hydrogen is produced by reacting bio-methane with water or oxygen, using methane gas with a purity of 95% or more as a raw material through a catalytic reactor equipment capable of extracting hydrogen with a purity of 99.995% or higher. The figures below show the schematic as well as in UNISM model diagrams.

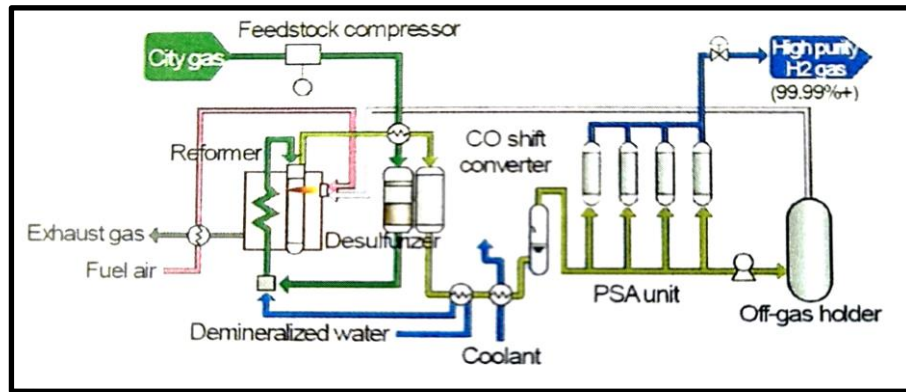
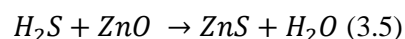


Figure 3.6 Company A's (H<sub>2</sub> extractor manufacturer) reforming technology process chart

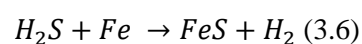
Tracking the figure 3.8 UNISIM basic processes (procedures) of the simulation can be summarized as:

- First: RNG (Renewable natural gas, assumed to be pure methane at this stage) is compressed and heated in Compressor-2, undergoes a desulfurization process in Desulfurizer-2, and then flows into Mixer-2.
- Simultaneously with stage 1, desalinated water (Water 1-2) is pumped and passes through HEATER 2-2, becomes vapor, and is supplied to Mixer-2.
- RNG and steam mixed in Mixer-2 are reheated by Heater to Rfm-2 and then moved to the reformer.
- The product in the reformer is heated again and moves to the Low-Temperature Shifter (LTS-Shift-2)
- The gas cooled by C-102-2 is supplied to the PSA device for hydrogen generation, and part of the gas is sent to Off Gas Holder-2 as a balancer. Here, the tail gas moves back to the combustor, where additional fuel and air are added to provide heat and energy for sending to the reformer.

The key equipment in UNISIM LCA modeling is Desulfurizer, Reformer, LTS (Low-Temperature Shifter), and PSA (Pressure Swing Adsorption). When using a desulfurizer to remove hydrogen sulfide (H<sub>2</sub>S) from a gas stream, the main reactions involved are:



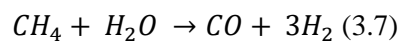
When using an iron-based catalyst to desulfurize hydrogen sulfide (H<sub>2</sub>S), the main reactions are as follows:



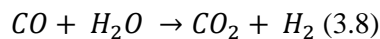
In the Desulfurizer, the desulfurization mechanism is determined depending on the type of catalyst:

1. Activated carbon-based catalyst: Activated carbon can be used as a catalyst in some desulfurization processes. Activated carbon absorbs sulfur compounds and helps remove them from the gas stream.
2. Copper-based catalysts: Copper-based catalysts such as copper oxide (CuO) or copper sulfide (CuS) can be used for certain desulfurization reactions. For example, in the Claus process for sulfur recovery, a copper catalyst catalyzes the conversion of hydrogen sulfide (HS) to sulfur.
3. Cobalt-based catalysts: Cobalt-based catalysts are used in some desulfurization processes, such as hydrodesulfurization (HDS) in petroleum refining. In HDS, cobalt-molybdenum (Co-Mo) or cobalt-nickel-molybdenum (Co-Ni-Mo) catalysts are used to remove sulfur compounds from hydrocarbons.
4. Zinc Oxide (ZnO) Catalyst: ZnO is a common catalyst used for desulfurization in processes such as the “Zinc Oxide Desulfurization” (ZOD) process, in which ZnO reacts with hydrogen sulfide to form zinc sulfide and water.
5. Iron-based catalysts: Iron-based catalysts such as iron oxide (Fe<sub>2</sub>O<sub>3</sub>) or iron sulfide (FeS) can be used for certain desulfurization reactions. Iron-based catalysts are used in a variety of processes, including iron sponge desulfurization and some hydrotreating reactions.
6. Biological catalysts (Biocatalysts): In the bio desulfurization process, certain microorganisms serve as catalysts to selectively remove sulfur compounds from fuel or gas streams.

The main reforming reaction of SMR includes steam reforming of methane (CH<sub>4</sub>) to produce hydrogen (H<sub>2</sub>) and carbon monoxide (CO), and the related reactions are as follows.



The Low-Temperature Shifter (LTS) Water–gas shift reaction (WGS) can be expressed as:



At this point, certain procedures need to be assigned like selection of the type of catalyst to be applied to the reformer and initial input conditions for normal operation of the desulfurizer and PSA. Furthermore, PSA Column type needs to be appointed to activate the function of PSA Unit-2. After resolving the said issues, the initial input value is to be inserted for each part that makes up the simulation.

For this work, based on the schematic diagram of the SMR system actually operated by domestic company A, UNISIM model modified according to A’s existing SMR simulation modeling as per the description below:

Initially, the mole fraction input value for simulation refers to data held by domestic company A. The main components of this simulation model are:

1. Multi-stage compressor for RNG (Renewable Natural Gas) processing
2. Heating system for water stream treatment
3. Main reformer that converts methane gas and water vapor into synthesis gas
4. Low-temperature water gas shift reaction (LT-WSR) that converts synthesis gas into carbon dioxide and hydrogen
5. PSA splitter (PSA) that separates hydrogen and tail gas
6. Water recycling treatment to supply additional water to the water circulation cycle

Table 3-8 Initial data input (RNG and water (Demi-water) temperature, pressure, supply mass flow rate, supply molar flow data)

No	Stream	Temperature (Celsius)		Pressure (Mpa.G)	Mass flowrate (kg/h)	Molar flowrate (kmol/h)					
1	Feed Gas	30		0.2	91.8	5.01					
2	Water(Demi-water)	26		1.5	262.1	14.55					
Molar composition(%)											
No	Stream	CH <sub>4</sub>	CO <sub>2</sub>	H <sub>2</sub> O	N <sub>2</sub>	CO	H <sub>2</sub>	O <sub>2</sub>	C <sub>2</sub> H <sub>6</sub>	C <sub>3</sub> H <sub>8</sub>	i-C <sub>4</sub> H <sub>10</sub>
1	Feed Gas	89.4	0	0	0	0	0	0	5.8	3.7	1.1
2	Water(Demi-water)	0	0	100	0	0	0	0	0	0	0

### 3.5.1 Multi-Stage Compressor System Modeling

A multi-stage compression system, as a sub-modeling of SMR simulation modeling, is developed considering multi-stage compression equipment applied in the actual SMR industry. Through this system, RNG repeats “compression-maintaining appropriate temperature-intermediate cooling-compression” until it reaches the target temperature and pressure required for simulation operation.

### 3.5.2 Heating System Modeling

As a sub-modeling of SMR simulation modeling, a multi-stage heating system is responsible for the process of converting water initially supplied in liquid form into steam. The main reformer reaction operates on the principle that water vapor reacts with methane in the reformer tank, and at this time, a system is used to gradually vaporize water using several heaters to maximize reactivity within the reformer.

For this sub model as well, the initial input values of pressure, temperature, and molar flow are defined based on company A data. The figures below show the physical properties of the initially supplied water, and the water vapor evaporated through the heating process. Remarking that the same

physical properties are set for both of them, confirming that all water initially supplied in the simulation is completely vaporized.

### 3.5.3 Reformer System Modeling

The high-temperature, high-pressure RNG and steam formed in the previous step are introduced into the reformer. Moreover, Mixed steam gas generated within the reformer is supplied to the LT-WSR stem. At this time, methane gas generated through heavy-hydrocarbon reactions between ethane, propane, and isobutane contained in RNG is additionally included in the system. Ethane, propane, and isobutane used in this reaction account for 17.94% of the total RNG, and methane is additionally produced due to this heavy hydrocarbon reaction, and its proportion is approximately 12% of the supplied RNG.

The important parts of the reformer system are indicated as (a) Gas-Water Input part and (b) reformed gas part. Data on RNG gas, steam, and reformed gas corresponding to the two points can be checked as follows.

Table 3-9 Company A's data used for initial reformer setup

No	Input Parameters	Unit	Value				
1	Feed Gas mass flow	Kg/h	91.8				
2	CH <sub>4</sub> conversion ratio	%	68~70				
3	CO conversion to LT-WSR	%	90~91				
4	H <sub>2</sub> separation efficiency in pressure swing adsorber	%	85				
No	steam	Molar composition %					
		CH <sub>4</sub>	CO <sub>2</sub>	H <sub>2</sub> O	N <sub>2</sub>	CO	H <sub>2</sub>
1	Feeds gas	89.4	0	0	0	0	0
2	Water (Demi-water)	0	0	100	0	0	0
3	Reformer-inlet	22.31	0.62	71.95	0	0.04	2.47
4	Reformer-outlet	4.89	6.38	28.79	0	9.34	50.6

### 3.5.4 LT-WSR System Modeling

LT-WSR is a chemical reaction that occurs after the first SMR reaction during the hydrogen and synthesis gas production process. The main purpose of LT-WSR is to convert carbon monoxide and water vapor contained in the reformed gas generated after the SMR reaction into carbon dioxide and hydrogen.

Table 3-10 Company A data used for initial setup of LT-WSR

No	Input Parameters	Unit	Value				
1	Feed Gas mass flow	Kg/h	91.8				
2	CH <sub>4</sub> conversion ratio	%	68~70				
3	CO conversion to LT-WSR	%	90~91				
4	H <sub>2</sub> separation efficiency in pressure swing adsorber	%	85				
No	steam	Molar composition %					
		CH <sub>4</sub>	CO <sub>2</sub>	H <sub>2</sub> O	N <sub>2</sub>	CO	H <sub>2</sub>
1	Feeds gas	89.4	0	0	0	0	0
2	Water (Demi-water)	0	0	100	0	0	0
3	Reformer-inlet	22.31	0.62	71.95	0	0.04	2.47
4	Reformer-outlet	4.89	6.38	28.79	0	9.34	50.6
5	LT-WSR inlet	4.89	6.38	28.79	0	9.34	50.6
6	LT-WSR outlet	4.89	14.81	20.36	0	0.9	59.04

Likewise, if a simulation is performed with the carbon monoxide conversion rate set to 91% among the basic conditions provided by Company A, the carbon monoxide input into the LT-WSR process is reduced by 91%.

### 3.5.5 Condenser – Water Downstream Recycling Modeling

The condenser modeling performs the function of separating and removing moisture contained in the gas collected in the condenser tank, which makes it possible to maintain the dryness of hydrogen gas. The moisture separated in this process is recycled to the heating system and reused to generate steam necessary for RNG reforming. Furthermore, reformed gas stored in the condenser tank (Cond. Vap.) in (c) flows into the PSA and is then used as input gas to extract hydrogen.

### 3.5.6 PSA System Modeling

The pressure swing adsorption (PSA) system is responsible for extracting and separating hydrogen from the reformed gas supplied from the condenser, and the gas composition present at its inlet and outlet. The function of PSA in SMR is to selectively separate hydrogen from reformed gas containing carbon dioxide, carbon monoxide, and trace amounts of other impurities. The reformed gas remaining after hydrogen is extracted can be reused as flue gas and tail gas, of which the tail gas is used to remove carbon dioxide in the SMR process or to a reformer.

The hydrogen of approximately 94.23% purity is extracted from the PSA discharge point. The reason the mole fraction of hydrogen is not 1.0 is because some impurities (carbon monoxide, steam, carbon dioxide) are present. In relation to the purity of hydrogen, some problems are occurring in the UNISIM simulation, restricting achieving the target hydrogen purity value of 99.999%, but additional verification is required as of now. Before performing the simulation, the hydrogen purity target value is set to 99.99% and then the calculation is performed. In the process of performing real-



time calculations of the SMR process, this target value is modified by the program itself and automatically adjusted downward to approximately 92%. As the results derived from each element that constitutes SMR modeling interact in real time, it is necessary to determine at what point the hydrogen purity target value was adjusted downward and what other factors prevent hydrogen with a purity of 94.23% or higher from being produced. Not enough technical information is available as of now. More clues need to be added to obtain the purity of the hydrogen produced above the target value of 99.999% like instructions on PSA types on UNISIM that have the same or most similar structure and characteristics to the PSA system in use in actual industry. Information on the PSA colume type selected in conjunction with the above PSA type is also needed. Data on post-treatment systems that can increase the purity of hydrogen extracted from PSA. It is believed that LCA analysis of the production of hydrogen with a purity of 99.999% or higher will be possible through advanced research on SMR simulation modeling considering these mentioned matters.

### **3.6 UNISM Model Results**

To utilize reformed gas other than hydrogen as flue gas and tail gas, an off-gas holder is added at the rear of the PSA system. There is a need to devise a method to discharge the flue gases outside the system and recirculate the tail gases so that they can be recycled in the internal processes of the reformer. As a measure to increase the purity of hydrogen extracted from PSA, it is necessary to introduce a 5-stage PSA colume and perform simulation. Advanced research on UNISIM simulation modeling is needed through comparative review of results with actual systems or big data to prove the validity of simulation results applying the above future research contents. In order to prove the validity of the simulation configuration by matching domestic company A's SMR facility and the modeling structure for UNISIM simulation as much as possible, it is necessary to modify the compression system and heating system modeling in the existing modeling. It is necessary to upgrade SMR simulation modeling and set parameters considering domestic circumstances. SMR simulation modeling modification parts are Water splitter, Heat exchanger, Desulfurizer colume, PSA colume.

Suggested modification can be summarized as:

1. Water splitter: In Company A's SMR design, the water flow is divided into two by the water splitter. The only difference between these two flows is the molar flow rate. The molar flow rate of water supplied before the water splitter is 14.55 kgmol/h, and after that it is divided into 4.69 kgmol/h and 9.86 kgmol/h, respectively.
2. Heat exchanger: In the existing SMR modeling, SMR modeling was performed using three general heaters, but it was replaced with a heat exchanger in consideration of Company A's SMR design.
3. Desulfurizer colume: Company A does not implement a separate reaction in the UNISIM simulation because the desulfurization process is a catalytic adsorption method, but

desulfurizer modeling was added to match the actual operating system.

4. PSA colume: Modifying the modeling so that hydrogen can be separated from the gas supplied to the PSA colume. At this time, the amount of H<sub>2</sub> produced from the PSA colume is defined to be the same as the amount of H<sub>2</sub> produced in Company A's data.

In addition, the following considerations are needed regarding PSA colume.

1. Company A is conducting simulations on PSA colume using Aspen's adsorption modeling for SMR simulation, as shown in Figure 3.2
2. The simulation in this service was performed based on UNISIM, and there is a possibility that there may be modeling differences between “Aspen”, the plant simulation used by Company A, thus more research is needed to be conducted to replicate Company A's modeling as much as possible.

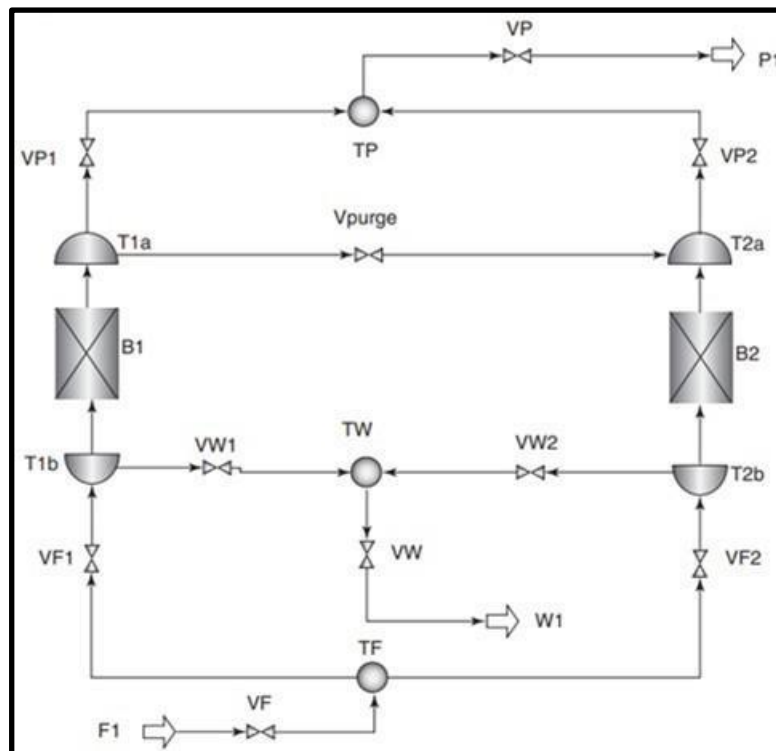


Figure 3.7 PSA workshop; Aspen adsorption modeling for air separation

## 4 RESULTS AND DISCUSSION

### 4.1 GHG from Each Waste

Reflecting domestic circumstances, three types of raw materials for RNG production were considered: AW (Animal Waste), WS (Water Sludge), and MSW (municipal waste). To determine the effect of raw material components on the Carbon Index value, a GREET calculation is performed by substituting the following conditions.

Table 4-1 Source of exhaust gas generation and environmental impact

Parameter (grams/mmBtu of Fuel Throughput)	Pathway	Value
CH <sub>4</sub>	LNG	348.298
	Food Waste	-05,961.794
	Animal Waste	-4,288.551
	Wastewater Sludge	-1,059.511
N <sub>2</sub> O	LNG	-1.302
	Food Waste	-6.378
	Animal Waste	-12.059
	Wastewater Sludge	-24.918
CO <sub>2</sub>	LNG	258
	Food Waste	54,368
	Animal Waste	24,441
	Wastewater Sludge	-70,341
GHGs (CO <sub>2</sub> , CH <sub>4</sub> , N <sub>2</sub> O)	LNG	10,144
	Food Waste	-108,994
	Animal Waste	-94,859
	Wastewater Sludge	-107,478

Table 4-2 Analysis of environmental impact trends of hydrogen production according to raw material type and composition

Division	Biogas raw materials				Hydrogen production (WTG) GHG	Analysis of environmental impact trends according to raw material composition
	Landfill Gas (LFG, %)	Animal Waste (AW, %)	Water Sludge (WS, %)	Municipal Solid Waste (MSW, %)	Carbon Index (kg_CO <sub>2</sub> /kg_H <sub>2</sub> )	
<b>AW100</b>	<b>0</b>	<b>100</b>	<b>0</b>	<b>0</b>	<b>-21.86</b>	<b>13<sup>th</sup> place</b>
AW75-WS25	0	75	25	0	-23.10	11 <sup>th</sup> place
AW50-WS50	0	50	50	0	-24.34	9 <sup>th</sup> place
AW25-WS75	0	25	75	0	-25.59	5 <sup>th</sup> place
<b>WS100</b>	<b>0</b>	<b>0</b>	<b>100</b>	<b>0</b>	<b>-26.83</b>	<b>1<sup>st</sup> place</b>
<b>WS75-SW25</b>	<b>0</b>	<b>0</b>	<b>75</b>	<b>25</b>	<b>-26.47</b>	<b>2<sup>nd</sup> place</b>
<b>WS50-SW50</b>	<b>0</b>	<b>0</b>	<b>50</b>	<b>50</b>	<b>-26.12</b>	<b>3<sup>rd</sup> place</b>
WS25-SW75	0	0	25	75	-25.76	4 <sup>th</sup> place
MSW100	0	0	0	100	-25.41	6 <sup>th</sup> place
MSW75-W25	0	75	0	25	-22.75	12 <sup>th</sup> place
MSW50-W50	0	50	0	50	-23.63	10 <sup>th</sup> place
MSW25-W75	0	25	0	75	-24.52	8 <sup>th</sup> place
AW33-S33-MSW34	0	33	33	34	-24.86	7 <sup>th</sup> place
<b>LFG100 (reference)</b>	<b>100</b>	<b>0</b>	<b>0</b>	<b>0</b>	<b>3.92</b>	<b>14<sup>th</sup> place</b>

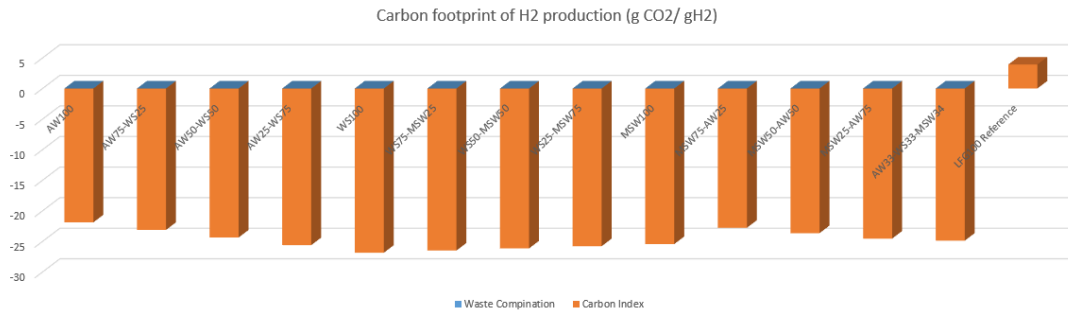


Figure 4.1 Carbon footprint of H2 Production (gCO<sub>2</sub>/gH<sub>2</sub>)

All Carbon Indexes shown in Table 4.1 are calculated based on WTG, which occurs when carrying out the entire process from the collection of waste (LFG, AW, WS, MSW), that were used as raw materials, to the production of hydrogen. Table 4.1 represents the amount of CO<sub>2</sub> produced.

Among the four types of waste considered to perform the GREET simulation, LFG was confirmed to have the greatest impact on GHG generation as it produces the highest CO<sub>2</sub> compared to other cases. This is because when LFG is used as a raw material component in the SMR process, it shows the highest CO<sub>2</sub>, CH<sub>4</sub> (used for combustion or power generation), and GHGs emissions compared to other raw material components, and this can be confirmed through GREET results as follows:

	Landfill Gas to NG as Intermediate Fuel	Animal Waste to NG as Intermediate Fuel	Wastewater Sludge to NG as Intermediate Fuel	MSW to NG as Intermediate Fuel
	Total	Total	Total	Total
Loss factor	1.000	1.000	1.000	1.000
Total energy	0	7,234	-1,454,860	-330,248
Fossil fuels	0	7,185	-1,283,472	-127,376
Coal	0	8	-40,027	-1,077
Natural gas	0	776	-1,090,303	-141,437
Petroleum	0	6,400	-153,142	15,139
Water consumption	0.000	0.143	-282.943	-11.136
VOC	-28.216	-7.921	-119.872	-36.322
CO	-42.331	12.913	-117.725	-24.592
NOx	-13.685	15.737	-168.732	-12.424
PM10	-5.406	0.528	-37.277	-6.081
PM2.5	-5.406	0.496	-30.376	-5.488
SOx	-0.274	0.107	-275.777	-21.968
BC	-5.730	-1.691	-6.942	-4.083
OC	0.136	1.498	-1.273	0.764
CH4	384,298	-4,288,557	-1,038,548	-5,961,233
N2O	-1,302	-12,059	-24,559	-6,369
CO2	258	24,437	-54,460	54,792
CO2 (w/ C in VOC & CO)	104	24,433	-55,019	54,640
GHGs	10,231	-95,624	-90,922	-109,691

Figure 4.2 Environmental impact figures according to the type of raw materials used to produce clean H<sub>2</sub>

From Figure 4.1 there are certain remarks:

- 1- The results that should be focused on in this data are the levels of CH<sub>4</sub>, CO<sub>2</sub>, and GHGs, which are closely related to the greenhouse effect. If the component is released to the outside (analysis boundary area) during the collection process of each raw material (LFG, AW, WS,

MSW), the value becomes positive “+”. However, if the waste is collected inside the analysis boundary without being released to the outside, it will be negative“-”.

- 2- Generally, LFG contains 45-50% of CH<sub>4</sub>, 35-40% of CO<sub>2</sub>, 10-15% of N<sub>2</sub>, and trace amounts of H<sub>2</sub>, O<sub>2</sub>, H<sub>2</sub>S, NH<sub>3</sub>, etc. A large amount of CH<sub>4</sub> is removed in the process of collecting LFG. Because it is released to the outside and mixed in the atmosphere. The environmental impact value of CH<sub>4</sub> is very high compared to AW, WS, and MSW, and this can be equally applied and interpreted to GHGs.
- 3- In particular, when LFG is used as a raw material, it shows a significant CH<sub>4</sub> impact. CH<sub>4</sub> has a relatively high GHG impact compared to CO<sub>2</sub>, at least 21 times and up to 80 times depending on the standard, so AW, WS, and MSW are used as raw materials. This is compatible with Shin et. al conclusion in [46]. Compared to the case considered, the Carbon Index shows a relatively high value of 3.92 kgCO<sub>2</sub>/kgH<sub>2</sub>.
- 4- Compared to LFG, in the case of AW and MSW, the CO<sub>2</sub> impact is high, but the CH<sub>4</sub> impact is very low with a negative value, so it is possible to predict that the overall GHGs impact of AW and MSW will be significantly lower than that of LFG.
- 5- In the case of WS, because the impact of CH<sub>4</sub>, CO<sub>2</sub>, and GHGs is lowest, it recorded the lowest Carbon Index value of -26.83 kgCO<sub>2</sub>/kgH<sub>2</sub> compared to other single raw materials and mixed fuels.
- 6- Comparing the environmental impact of each raw material based on the above results, it can be observed that the Carbon Index is low in the order of WS > MSW > AW > LFG.
- 7- In addition, based on the fact that cases using WS as the main raw material such as WS100, WS75-MSW25, and WS50-MSW50 have a lower environmental impact compared to other cases, clean hydrogen production using WS is the most environmentally friendly method.

3) Summary of Energy Consumption, Water Consumption, and Emissions: Btu or Gallons or Grams per n				
	Landfill Gas to NG as Intermediate Fuel	Animal Waste to NG as Intermediate Fuel	Wastewater Sludge to NG as Intermediate Fuel	MSW to NG as Intermediate Fuel
	Total	Total	Total	Total
Loss factor	1.000	1.000	1.000	1.000
Total energy	0	7,274	-1,610,701	-334,415
Fossil fuels	0	7,240	-1,497,057	-133,086
Coal	0	88	-343,734	-9,197
Natural gas	0	748	-981,573	-138,530
Petroleum	0	6,405	-171,750	14,641
Water consumption	0.000	0.138	-260.888	-10.547
VOC	-28.216	-7.920	-121.699	-36.371
CO	-42.331	12.915	-124.994	-24.786
NOx	-13.685	15.743	-192.574	-13.062
PM10	-5.406	0.529	-42.214	-6.213
PM2.5	-5.406	0.497	-32.545	-5.546
SOx	-0.274	0.115	-308.544	-22.844
BC	-5.730	-1.691	-7.041	-4.086
OC	0.136	1.498	-1.304	0.763
CH4	384.298	-4,288.548	-1,070.949	-5,962.100
N2O	-1.302	-12.059	-25.201	-6.386
CO2	258	24,444	-79,579	54,121
CO2 (w/ C in VOC & CO)	104	24,439	-80.154	53,968
GHGs	10,231	-95,617	-117,131	-110,393

Figure 4.3 GHGs emissions by energy raw material (unit is automatically converted according to user selection among mmBtu, Btu, gal, g)

The “Negative” in GHGs calculations can mean that an action or technology has a positive impact on reducing GHG emissions or protecting the environment, indicating that active use of raw materials from various sources other than landfill gas can help mitigate climate change. Therefore, it is appropriate to understand that negative numbers in GHGs calculations reflect the positive impact of specific actions or technologies on reducing GHGs emissions, rather than model errors or problems. In the context of GHG emissions and climate change response, smaller GHG values are interpreted to have a positive impact on the environment and climate change mitigation efforts, because smaller GHG values mean less GHGs are emitted into the atmosphere. This means that it can reduce the goods that must be input to respond to global warming and climate change. Landfill gas is noticed to have a greater impact on greenhouse gas (GHG) emissions than other emission sources due to several specific factors below that affect the production of this gas and its global warming potential.

1. Gas composition: Landfill gas mainly consists of methane (CH<sub>4</sub>) and carbon dioxide (CO<sub>2</sub>). Despite its short half-life, methane is a much more powerful GHG in retaining heat in the atmosphere than carbon dioxide in the short term (approximately 30 times more powerful greenhouse gas effect) which as stated by EPA United States Environmental Protection Agency in [47]. Therefore, the amount of methane released into the atmosphere has a greater impact on global warming than the same amount of carbon dioxide.
2. Anaerobic decomposition process: In landfills, organic waste decomposes under anaerobic conditions, that is, in the absence of oxygen. Microorganisms activated under these conditions produce methane as a by-product, and a significant amount of methane is produced during this decomposition process.
3. Decomposition rate: The decomposition rate of organic waste in landfills can be quite high, with a clear trend towards this, especially in the first few years after disposal. This releases significant amounts of methane into the atmosphere.
4. Uncontrollable situation: Applicable to cases where there is no effective gas collection system.

#### **4.2 Hydrogen Production (w/o CSS)**

In the case of the US-based calculation and the Korean-based calculation, the basic outcomes such as the performance of the reforming facility (energy required, energy production) and the source of the required energy referenced in each country are different, so it can be verified that the Carbon Index produces different values. However, it is common to see the Carbon Index trend according to the presence or absence of CCS (Carbon Capture Storage). The Carbon Index in cases where CCS is included is about 1/3 smaller than in the case without CCS this goes smooth with [46]. In addition, since the difference in Carbon Index values is not that large, then it could be possible to derive sufficiently acceptable results with similar trends as above, if some values in the GREET calculation

to be developed based on the U.S. will be reset to suit domestic circumstances. Based on the figure 4.4 of this work results and figure 4.3 of the IEA report, the range of Carbon Index in the absence of CCS (95% capture) is 1.0~4.7 kgCO<sub>2</sub>-eq/kgH<sub>2</sub>, and this calculated value falls within this range, so the simulation The results can be judged to meet IEA standards.

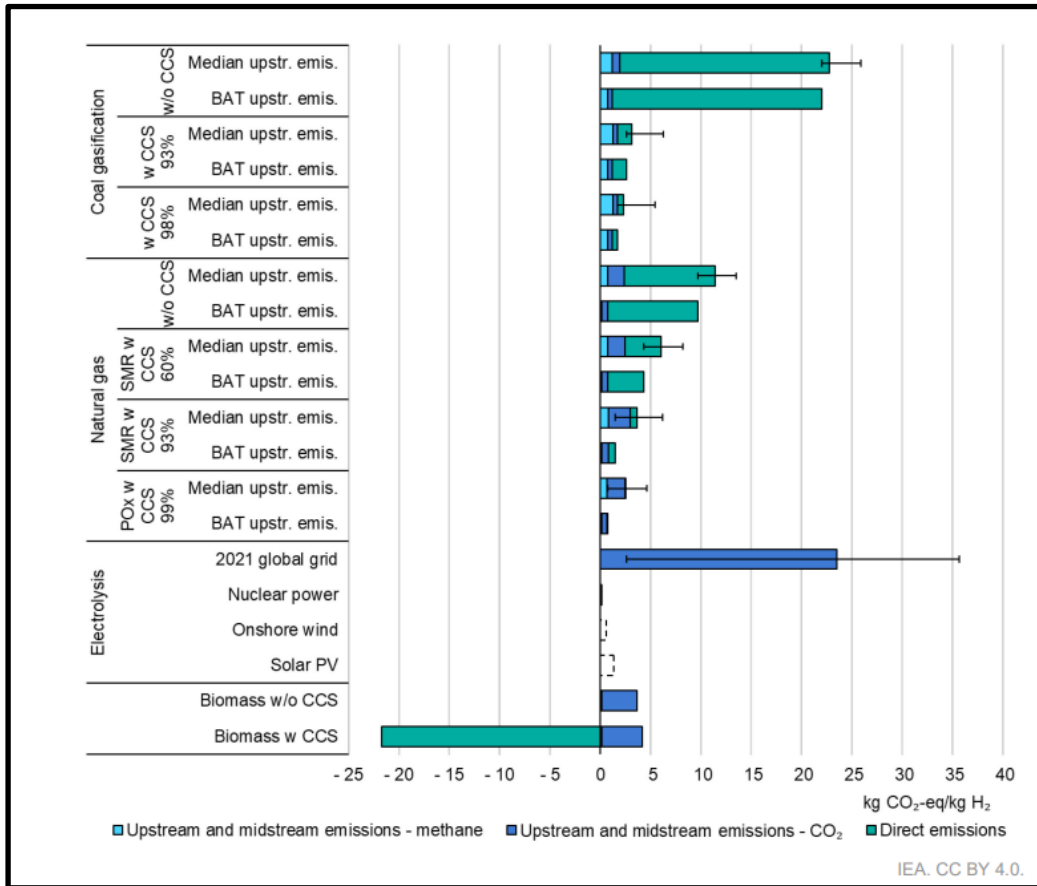


Figure 4.4 IEA report about the range of Carbon Index for different fuels[44]

The results of GREET calculation under the above conditions can be summarized as follows:

WITH GREET H2 US Electricity		
Value	Unit	
1.26	kg CO <sub>2</sub> /kgH <sub>2</sub>	With CCS
3.86	kg CO <sub>2</sub> /kgH <sub>2</sub>	Without CCS
WITH GREET H2 Korean Electricity		
Value	Unit	
1.31	kg CO <sub>2</sub> /kgH <sub>2</sub>	With CCS
3.92	kg CO <sub>2</sub> /kgH <sub>2</sub>	Without CCS

Figure 4.5 GREET calculation results (comparison of US and Korean standard calculation results)



### 4.3 Hydrogen Production Waste

Comparison of the US standard hydrogen production process (w/o CCS) and domestic company A's hydrogen production process (w/o CCS) yields that although the SMR system of the National Energy Technology Laboratory in the United States and the SMR system of Company A in Korea have similar configurations, but it also shows the following differences:

1. Domestic company A uses a two-stage desulfurizer.
2. Domestic Company A does not use Performer
3. At Domestic Company A, the flue gas generated from PSA is stored in an off-gas holder and then recirculated to the reformer.
4. It is possible to implement domestic company A's SMR in GREET, but it is necessary to consider the following modeling configuration differences.
  - a) In GREET 3-step, WSR is applied.
  - b) In GREET, the flue gas generated from PSA is supplied directly to the reformer without passing through the off-gas holder.

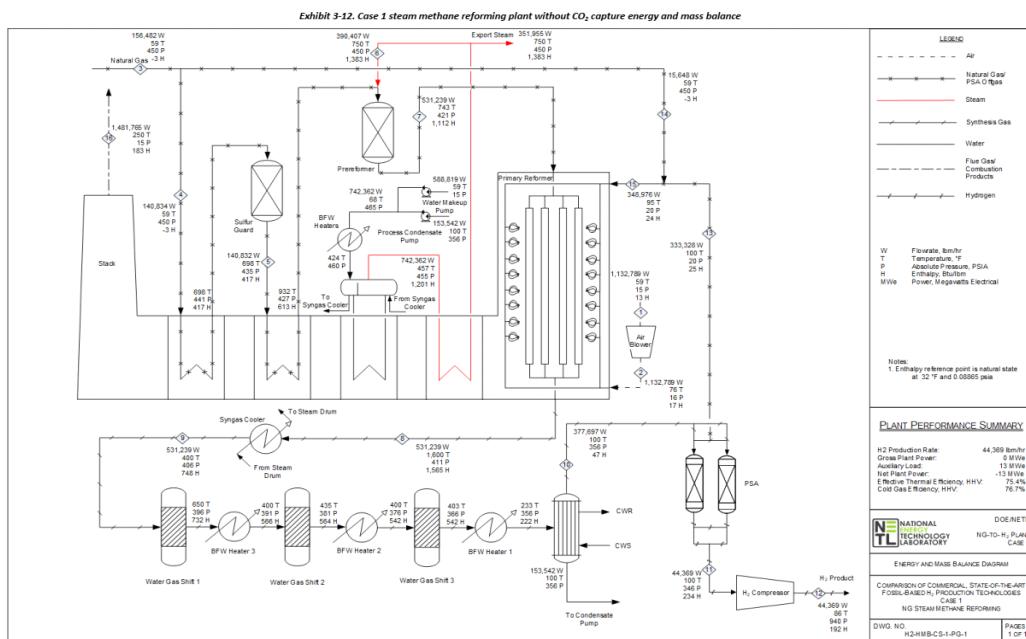


Figure 4.6 Schematic diagram of the SMR plant (w/o CCS) at the U.S. National Energy Technology Laboratory [34]

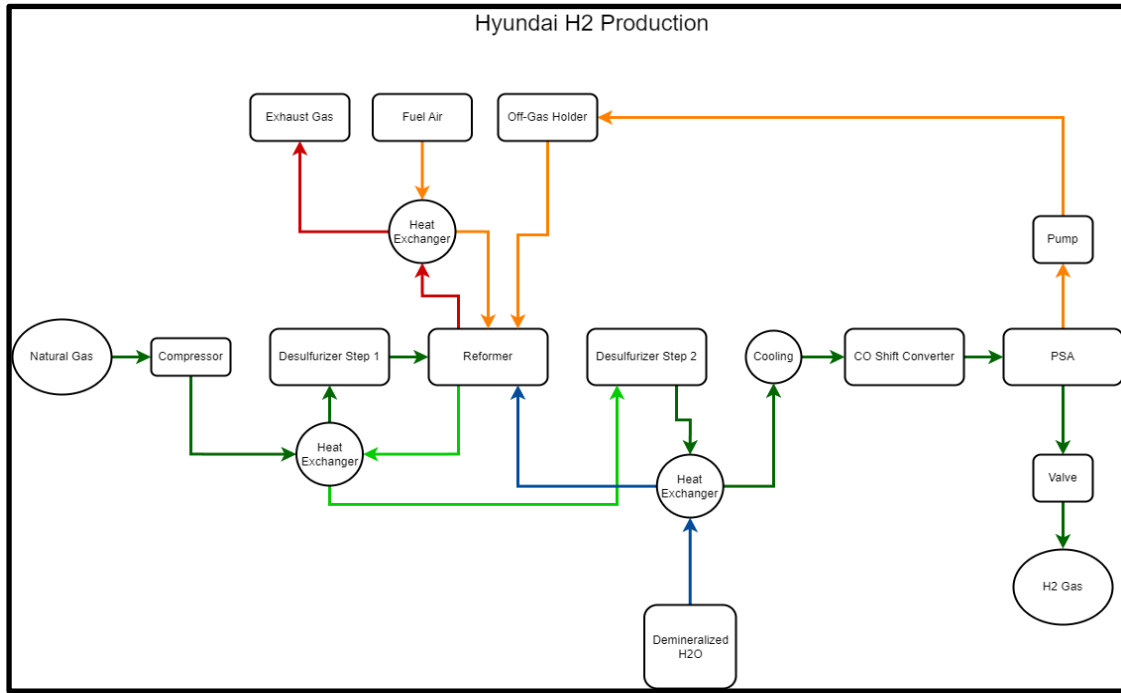


Figure 4.7 Domestic Company A SMR (w/o CCS) configuration diagram

Considering the diversification of production raw materials for methane, which is the raw material for hydrogen generation. It was attempted to apply various initial raw material compositions as follows:

1.1) Share of Fuels from Each Waste				
	NG as Intermediate	MeOH	CNG	LNG
Landfill Gas	97%	100%	100%	100%
Bio-gas from AD of Animal Waste	1%	0%	0%	0%
Bio-gas from AD of Wastewater Sludge	1%	0%	0%	0%
Bio-gas from AD of MSW	1%	0%	0%	0%

Figure 4.8 Input initial conditions for parameter study considering diversification of methane production raw materials (example)

GREET calculation is performed by applying the fuel value of “NG as Intermediate” above as follows:

Table 4-3 Landfill gas-based and other waste mixing ratio

Name	Landfill Gas (LFG, %)	Animal Waste (AW, %)	Water Sludge (WS, %)	Municipal Solid Waste (MSW, %)
LFG 97	97	1	1	1
LFG 96 A	96	2	1	1
LFG 96 B	96	1	2	1
LFG 96 C	96	1	1	2
LFG 95	95	2	2	1

Table 4-4 Carbon Index by waste mixing ratio

<b>LF 97 (LFG 97 AW 1 WS 1 MSW 1)</b>		
Value	Unit	
0.54	kg CO2/kgH2	With CCS
1.31	kg CO2/kgH2	Without CCS
<b>LF 96 A (LFG 96 AW2 WS 1 MSW 1)</b>		
Value	Unit	
0.31	kg CO2/kgH2	With CCS
2.92	kg CO2/kgH2	Without CCS
<b>LF 96 B (LFG 96 AW1 WS 2 MSW 1)</b>		
Value	Unit	
0.14	kg CO2/kgH2	With CCS
2.75	kg CO2/kgH2	Without CCS
<b>LF 96 C (LFG 96 AW1 WS 1 MSW 2)</b>		
Value	Unit	
0.16	kg CO2/kgH2	With CCS
2.75	kg CO2/kgH2	Without CCS
<b>LF 95 (LFG 95 AW 2 WS 2 MSW 1)</b>		
Value	Unit	
-0.12	kg CO2/kgH2	With CCS
2.49	kg CO2/kgH2	Without CCS

Compared to the previous calculation results that only considered LFG, the Carbon Index is significantly lower when AW, WS, and MSW are considered as methane generation raw materials, which means that the GHGs factor value for AW, WS, and MSW set on GREET is judged to be a negative number because it shows a significantly lower value compared to LFG.

## 5 SUMMARY AND CONCLUSION

The growing demand for energy is driving an acceleration of the development of hydrogen generation. Still, greater caution must be exercised because the processes involved in producing hydrogen have the potential to be environmentally harmful. In particular, the carbon dioxide footprint is the subject of this work since it is connected to hydrogen production, an overlooked contributing factor to the greenhouse effect. A comprehensive numerical analysis of the carbon dioxide footprint associated with manufacturing hydrogen from various renewable feedstocks is presented in this paper. The study focuses on renewable feedstocks, such as wastewater (SW), animal and food waste (AW), landfill gas (LFG), and food waste (MSW). After careful examination, Argonne National Labs' GREET 2022 software yields interesting results regarding carbon dioxide footprint. Based on domestic data and compared to a study referenced in the United States, a total of 14 raw material combinations (landfill gas, livestock manure biogas, and sewage sludge biogas) are assumed with different mixing ratios, resulting in the following conclusions:

1. Cases where sewage sludge biogas (WS) was used as the primary raw material, such as WS100, WS75-MSW25, and WS50-MSW50, had lower greenhouse gas emissions than other cases. This suggests that WS100 would be the most environmentally friendly method for producing clean hydrogen, with a  $-26.83 \text{ kgCO}_2/\text{kgH}_2$  Carbon Index.
2. Of the 14 combinations of  $+3.92 \text{ kgCO}_2/\text{kgH}_2$ , the Carbon Index has the highest value when using 100LFG in this assessment.
3. The Carbon Index for the GREET Hydrogen production LCA WTG assessment is approximately one-third lower in cases with CCS included than in the case without CCS, which is  $3.92 \text{ kgCO}_2/\text{kgH}_2$ .

This LCA's GHG emissions results from this work were comparable to the US governments and Argonne National Laboratory's data on greenhouse gas emissions from hydrogen production based on biogas. Moreover, the conducted outcome of Carbon Index expected for producing  $\text{H}_2$  from biogas in the absence of CCS (95% capture) falls within the range of  $1.0\sim 4.7 \text{ kgCO}_2\text{-eq}/\text{kgH}_2$ , satisfying the requirements mentioned in the Korea government IEA report in Figure 4.4.

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