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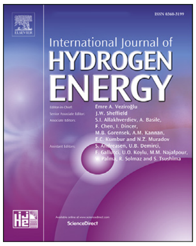


Hydrogen-rich syngas generation through microwave plasma reforming of greenhouse gases

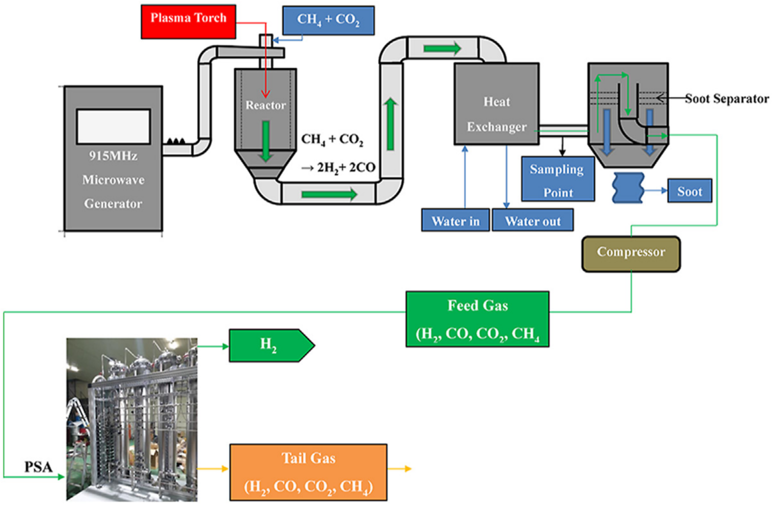
Olugbenga Akande [a,c,*], *BongJu Lee* [c,d], *Jude A. Okolie* [b],
Hugues Nkomba Museba [a]

[a] *Department of Computer Science and Electrical Engineering, Handong Global University, 558 Handong-ro, Heunghae-eup, Buk-gu, Pohang, Gyeongsangbuk-do 37554, South Korea*
[b] *Gallogly College of Engineering, University of Oklahoma, Norman, USA*
[c] *Green Science Corporation, 62, Gyesan 11-gil, Taebaek-si, Gangwon-do, 26050, South Korea*
[d] *Department of Advanced Convergence, Handong Global University, 558 Handong-ro, Heunghae-eup, Buk-gu, Pohang, Gyeongsangbuk-do 37554, South Korea*

HIGHLIGHTS

- Greenhouse gases (GHG) reforming for mass hydrogen generation.
- Investigated a 915 MHz atmospheric pressure microwave plasma torch for GHG reforming.
- Attained hydrogen production rate and energy yield of 1672 g (H_2)/h and 41.8 g (H_2)/kWh.
- CH_4 and CO_2 conversion of 95.6% and 95.4%, respectively, were realized.

GRAPHICAL ABSTRACT



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ABSTRACT

This study examined the effectiveness of a 915 MHz atmospheric pressure microwave torch for bulk syngas or hydrogen generation via noncatalytic greenhouse gas (GHG) reforming and its flexibility regarding the amount of soot formed by the process. The investigation shows that plasma-GHG reforming for mass hydrogen generation is feasible without a catalyst at high gas flow rate and microwave power.

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* *Corresponding author*. Department of Advanced Convergence, Handong Global University, 558 Handong-ro, Heunghae-eup, Buk-gu, Pohang, Gyeongsangbuk-do 37554, North Korea.
E-mail address: olu@handong.edu (O. Akande).

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The best hydrogen production rate and energy yield were 1672 g (H_2)/h and 41.8 g (H_2)/kWh, respectively, at 160 lpm gas flow rate and 40 kW microwave power at a CH_4/CO_2 ratio of 1. The conversion rates of CH_4 and CO_2 under the same conditions were 95.59% and 95.59%, respectively. The plasma reforming method employed in this study shows its excellent potential for industrial application.

Introduction

In addition to its usage in producing ammonia and nitrogenized fertilizers, hydrogen has many other applications. The uses of hydrogen include fueling hydrogen-powered factories and aircraft. It produces liquid fuels via Fischer–Tropsch synthesis (FTS) and gas-to-liquid (GTL) technology such as syngas fermentation. Hydrogen is also a precursor for dimethyl ether (DME) production [1]. Likewise, numerous processes rely on hydrogen, including petroleum refining, glass purification, semiconductor production, and the medical and pharmaceutical industries. Welding, annealing, and heat-treating metals are other uses of hydrogen [2]. The application of hydrogen extends to cooling power plant engines and hydrogenating vegetable oil's unsaturated fatty acids [3]. It is also used to power rocket launchers. The National Aeronautics and Space Administration (NASA), founded in 1958, uses the world's largest liquid hydrogen supply for rocket propulsion and fuel cell technologies [4].

The growing use of hydrogen for electricity generation, carbon-free heating, transportation, and other industrial processes is a significant step towards green reform and the hydrogen economy. Hydrogen trains, buses, cars, and forklift trucks are already operating in several countries, such as Canada and Japan [5]. Hydrogen can be produced from various feedstocks and processes involving wind, solar, and nuclear energy. However, the predominant viable feedstock for commercially viable hydrogen production in the near term is still fossil fuels [6].

Since the 18th century, industrialization and urbanization have positively impacted the global economy, but not without consequences. Owing to greenhouse gas (GHG) emissions, global warming, and climate change have reached record levels. According to current data, CO_2 emissions in 2011 were 150 times more than in 1850 [7]· Human activity has never resulted in such high levels of CO_2 emissions. The combustion of fossil fuels produces a substantial amount of CO_2. Also, CH_4 emissions are produced by agricultural operations, waste management, energy usage, and biomass burning. Anthropogenic factors in forestry and other land uses, such as deforestation, agricultural land clearing, and soil degradation, can also produce CO_2 [8].

The Paris climate agreement's goal of reducing carbon emissions by at least half by 2050 can be achieved through energy efficiency approaches, end-use electrification, and net-zero energy production. Instead of conventional fossil fuels, a "hydrogen economy" framework envisions hydrogen as a low-carbon alternative energy carrier for transportation and heating.

However, long-distance transportation, industrial operations, and residential heating pose significant challenges in transitioning to a low-carbon economy [9,10]. Consequently, low-carbon fuels like natural gas and biofuels and energy carriers like hydrogen will remain necessary in many applications. Hydrogen has many advantages over traditional electric battery storage systems, including persistent storage and usage of hydrogen on demand. Instead of using natural gas alone, hydrogen may be mixed with natural gas and used in existing power plants and household appliances. The International Energy Agency [11] reported that hydrogen fuel could decrease emissions from various sectors, including transportation worldwide.

Hydrogen derived from GHG reforming would be valuable to achieve the Paris Climate Agreement's goals of reducing carbon emissions while providing jobs and stimulating the economy.

Anthropogenic activities are the principal sources of GHG emissions (predominantly CO_2 and CH_4). Over two-thirds of all GHG emissions worldwide are made up of carbon dioxide [12]. Global warming, air pollution, severe weather, food supply instabilities, and increased forest fires are all exacerbated by GHG emissions.

Given the impact of GHG emissions on the environment, research has focused on strategies to minimize GHG emissions. Consequently, the use of CO_2 in many industries is receiving much attention. Hence, CO_2 is considered a valuable resource rather than a pollutant [13]. It is challenging to control GHG emissions in the current global energy system, but their use in synthesizing other chemicals is favorable from an environmental perspective. GHG reforming and carbon sequestration are some methods scientists have proposed to mitigate carbon emissions. However, methods that use or combine CO_2 with other compounds to produce fuels or other valuable chemicals are the most suitable to deal with the challenge of carbon emission.

GHG reforming began in 1888 and was intensively investigated in 1928 by Fischer and Tropsch [14]. The conventional GHG reforming is performed by introducing a CH_4–CO_2 mixture into a tubular fixed-bed reactor with catalysts and externally heated by natural gas combustion. Due to the reduction in CH_4 consumption while utilizing CO_2, GHG reforming has both ecological and economic benefits. Besides, because CO_2 is also a carbon source in the reforming process, the CH_4–CO_2 reaction stoichiometric saves half the methane required to produce the same CO in steam methane reforming

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(SMR) and partial oxidation (POX) processes [15]. Though the CH_4–CO_2 reforming yields a one-to-one H_2/CO ratio, modifying the H_2/CO ratio is performed by altering the CH_4/CO_2 ratio in the feed.

Plasma can effectively reform GHG in a compact reactor due to its low specific energy requirement, high temperature, and abundant reactive species. When GHG interacts with plasma, it produces syngas, a mixture of H_2 and CO. Syngas is useful in fuel cells, off-grid power plants, and other cutting-edge technologies. An intermediate step in the production of hydrogen is syngas generation. In 1986, plasma reforming of GHG was performed for the first time using a controlled arc [15]. Since then, even if energy consumption is still an issue, it has received more attention. Carbon buildup on the catalyst surface, which results in catalyst deactivation, is the fundamental hurdle to scaling up the catalytic GHG reforming process. Electron-induced chemistry and thermochemical processes can produce higher conversions and selectivity for plasma-GHG reforming than conventional catalytic reforming without a catalyst or soot formation [15].

Compared to coal or biomass gasification and natural gas reforming, hydrogen production through GHG reforming, also known as CO_2 dry reforming (CDR), can significantly reduce carbon emissions. Meanwhile, the GHG reforming reactor may be employed similarly to the steam methane reforming reactor [16]. A significant drawback of coal gasification or natural gas reforming hydrogen production methods is ample CO_2 emission. For instance, 1 ton of coal gasification produces 3.5 tons CO_2 [17]. Similarly, SMR generates approximately 10 kg CO_2 per kg H_2 [18,19].

The main methane reforming processes with conventional heating and a heterogeneous catalyst listed in Table 1 can be broken down into four stages, which are as follows:

The first and crucial stage in the process is the activation of methane, which requires a certain amount of energy to break the bonds between C-H_x or CH_x-H_x molecules. The characteristics of the catalyst surface influence the ability to do this. As the CH_x molecules become partially dissociated, they attach to the catalyst's surface, and active carbonaceous species, C*, are also present [20].

When the catalyst surface or the metal support comes into contact with core reactants such as H_2O, CO_2, or O_2, active oxygen species are generated through their activation in the second stage.

During the third stage, hydroxyl groups (OH-) are produced on the surface through a reaction involving CO_x and H radicals or surface O and H radicals. Upon binding to the surface, CH_x species react with O–H groups, forming CH_xO intermediates that ultimately decompose into CO and H_2. Typically, the catalyst-support material interface is the active location for CH_xO formation [20]

The fourth stage is the desorption of intermediates from the surface through oxidation. Oxygen species on the metal catalyst surface can react with surface groups such as CH_x, CH_xO, or CO. This can lead to the production of CO through the dissociation of CH_xO and CO surface or the Boudouard reaction (equation 1.8). Other reactions, such as the reverse water gas shift (the reverse of equation 1.6) and the CH_4 decomposition shift (equation 1.1), may also affect reforming performance and product selectivity [20]·

GHG reforming, which has significant ecological benefits has not been adopted on an industrial scale due to rapid catalyst deactivation. Hence, this investigation aims to demonstrate the possible practicality of a noncatalytic atmospheric pressure microwave plasma-GHG reforming process for the generation of hydrogen-rich syngas and hydrogen production at high absorbed microwave power (>30 kW) without the need for extra heating. Also, the study seeks to evaluate the microwaves plasma torch's potential for practical application in large-scale GHG reforming. The performance of the stainless-steel reactor employed in the study was tested through parameters such as hydrogen production rate in g

Table 1 – The main chemical reactions in methane reforming with H_2O, CO_2, and O_2 adapted from Ref. [22].

Reaction	Δ_rG function ($P = 1$ bar) kJ mol^{-1}	ΔrH_{298} kJ mol^{-1}	Equations
Pyrolysis	$\Delta_rG = 21{,}960 - 26.5 \times T$	+75	(1.1)
$CH_4 \rightarrow C_{(S)} + 2H_2$			
Dry reforming	$\Delta_rG = 61{,}770 - 67.3 \times T$	+247	(1.2)
$CH_4 + CO_2 \rightleftharpoons 2CO + 2H_2$			
Partial oxidation	$\Delta_rG = -653.9 - 369 \times T$	−71	(1.3)
$2CH_4 + O_2 \rightleftharpoons 2CO + 4H_2$			
Methane combustion	$\Delta_rG = -803{,}508 + 13 \times T -$	−802.5	(1.4)
$CH_4 + 2O_2 \rightleftharpoons CO_2 + 2H_2O$	$0.018 \times T2 + 8 \times 10-6 \times T3$		
Steam reforming	$\Delta_rG = 210{,}359 - 233.9 \times T$	+206	(1.5)
$CH_4 + H_2O \rightleftharpoons CO + 3H_2$	$\Delta_rG = -39{,}802 + 37.673 \times T$	−41	(1.6)
$CO + H_2O \rightleftharpoons CO_2 + H_2$	$\Delta_rG = 170{,}557 - 196.29 \times T$	+165	(1.7)
$CH_4 + 2H_2O \rightleftharpoons CO_2 + 4H_2$			
Boudouard reaction	$\Delta_rG = 39{,}810 + 40.9 \times T$	−172	(1.8)
$2CO \rightleftharpoons C + CO_2$			
Steam reforming of carbon	$\Delta_rG = 132{,}184 - 138.8 \times T$	+131	(1.9)
$C_s + H_2O \rightleftharpoons CO + H_2$			
Partial oxidation of carbon	$\Delta_rG = -110{,}872 - 89.4 \times T$	−110.5	(1.10)
$C + O \rightleftharpoons CO$			
Carbon combustion	$\Delta_rG = -393{,}647 - 2.5 \times T$	−393.5	(1.11)
$C + O_2 \rightarrow CO_2$			

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(H_2)/h, CH_4 and CO_2 conversion degree (%), and energy yield in g (H_2)/kWh as a function of absorbed microwave power.

Plasma-GHG reforming

GHG reforming may be considered the opposite of SMR (wet reforming) and is represented by equation (1.2). GHG reforming reactions require a high process temperature (700–900 °C) and a catalyst (Ni, Co, or MO_2C) for a high conversion rate due to the highly oxidized nature and thermodynamic stability of CO_2 as well as the inert chemical tendency of CH_4. Currently, GHG reforming has not been applied on an industrial scale due to its apparent drawback; the susceptibility of the process to coking and consequent catalyst deactivation [21,22]. GHG reforming has a high degree of endothermicity. At 1 atm and below 633 °C, the thermodynamic analysis suggests that the reaction is not spontaneous. Possible intermediate processes include steam reforming, methane decomposition, reverse water–gas shift, methanation, Boudouard reaction, and carbon gasification. Table 1 lists the main chemical reactions in methane reforming with steam, carbon dioxide, and oxygen.

Plasma reactors are compact and lightweight because of their high-power density, allowing for various applications. In addition, they are compatible with various fuel types, including natural gas, heavy oil, and biofuels [23–25]. Due to its capacity to function without an electrode, microwave plasma is ideal for gaseous pollutant abatement and purification [26,27]. Furthermore, when no catalysts are used to aid the reforming process, sulfur intolerance is not a problem, and the process can function at lower temperatures than conventional reforming methods. Plasma technology is scalable, flexible, and adaptable to many environments and has good carbon capture and utilization potential.

Compared to conventional techniques, microwave plasma reforming eliminates many drawbacks, such as the high capital cost and rapid degradation of catalysts, significant environmental footprint, slow startup time, and constraints regarding hydrogen generation from heavy hydrocarbons [24]. Plasma reforming has the same general reforming reactions as conventional reforming. However, the energy and free radicals required for the reforming reaction are supplied by plasma commonly generated using electricity or heat. Additionally, they may be configured to operate at temperatures lower than conventional reforming.

Plasma reforming can improve POX, Autothermal (ATR), and SMR reactors, which now account for most hydrocarbon reforming reactors [28]. In the future, these functionalities will allow for on-demand and remote manufacturing. Examples of on-demand installations include fuel cell feeds in residences, onboard automobile exhaust treatment systems, and containerized interior air treatment facilities for large-scale industrial operations [28].

Here are a few recent plasma-GHG reforming investigations: Alawi and Nguyen examined dry methane reforming for syngas production in an atmospheric pressure dielectric barrier discharge (DBD) plasma reactor. The best CH_4 (81.24%) and CO_2 (48.67%) conversions were obtained using a feed gas flow rate of CH_4: CO_2: N_2 of 0.3:0.3:1 and an input power of 10 W. Maximum H_2 and CO yields of 44.13 and 36.15%, respectively, were recorded under these input conditions. Low conversion degree and capacity are the limitations of the DBD plasma [29]. Dinh D. K et al. investigated GHG reforming in two different arc plasma reactors. The study obtained optimal CH_4 and CO_2 conversions of 74% and 49%, respectively, and energy efficiency of 53% [30]. A previous study by Chun, Se Min et al. evaluated syngas production by reforming CO_2 and CH_4 in a microwave plasma (2.45 GHz) torch, using a microwave power of 6 kW. The reforming process produced CO_2 and CH_4 conversions of 68.4% and 96.8%, respectively. The resulting syngas H_2/CO ratio was 0.9:1 when the reforming gas CO_2/CH_4 ratio was 1:1 [31]. The bench-scale experiment demonstrated that a microwave torch could reform CH_4/CO_2 without a catalyst.

Similarly, a waveguide-supplied metal cylinder-based microwave plasma source (MPS) operating at atmospheric pressure was employed by Hrycak B. et al. to assess hydrogen production efficiency by reforming CO_2 and CH_4 [32]. However, the drawback was low throughput and plasma destabilization at a high flow rate; also, using nitrogen as a balancing gas did not yield any results.

Furthermore, the effects of reaction temperature, CO_2/CH_4 ratio, and concentration of the primary gas components on the rate of CH_4 conversion and the energy efficiency of plasma and La–Ni/-Al_2O_3 catalyst for the GHG reforming were tested by LI Jia-qing et al. The results show that at 450 °C, the conversion rate of CH_4 was 41.57%, at a CO_2/CH_4 ratio of 1. This investigation's best CH_4 conversion rate was 92.82% at a CO_2/CH_4 ratio of 5 [33].

Likewise, Huaqin Wang et al. considered GHG reforming using a nickel (Ni)/activated carbon (AC) catalyst [34]. The reforming process was examined under plasma without a catalyst, catalyst alone and plasma with a catalyst to study the synergistic effect of plasma and catalyst. The best result was obtained when GHG reforming was performed with plasma utilizing a Ni/AC catalyst, resulting in 64.6% CO_2 conversion and 65.5% CH_4 conversion, 27.9% H_2 yield, and 32.1% CH_4 yield. The conversion rate and throughput of GHG reforming are lower in nonthermal plasmas, even in the presence of a catalyst than in warm or thermal plasma.

Huang et al., 2022 [35] used an impregnation method to produce a variety of Ni/mV2O3V2C catalysts with varying Ni concentrations for dry methane reforming. The best Ni addition rate was 13% by weight, whereas the optimum CH_4 and CO_2 conversion rates were 94% and 89%, respectively. Also, Cu/Ni La0.75Sr0.25Cr0.5Mn0.5O3 (LSCM) catalysts were used by Liu et al., 2022 [36] for dry methane reforming. The best methane conversion obtained was 88.75%.

Furthermore, Azeem et al., 2022 [37] studied the dry reforming of methane over Ni-supported mesoporous zirconia support. 68.8% for methane and 70.2% for carbon dioxide were the best results, respectively. Despite high conversion rates in all the procedures mentioned earlier, catalyst deactivation continues to be challenging, as with all catalytic GHG reforming.

To decrease soot formation and improve the H_2/CO ratio of the generated syngas, Yun Xia et al. explored using steam and greenhouse gases in a gliding arc discharge (GAD) plasma reformer [38]. According to the study, CH_4 conversion and energy conversion efficiency of syngas production initially increased and then fell as the Steam to Carbon(S/C) molar ratio increased from 0 to 2.3; the best values obtained were 55.6% and 36%, respectively, at an S/C ratio of 0.58. Their finding suggests that H_2O in the plasma produces more *OH radicals and H atoms, which promotes CH_4 dissociation, H_2 selectivity, and H_2 generation as the H_2/CO ratio increases from 0.76 to 1. The study also found that steam considerably reduced soot generation. The CH_4/CO_2 ratio varied between 1/3 and 3/1, and a carbon balance of more than 82% and a highly adjustable H_2/CO ratio ranging between 0.4 and 3. Compared to microwave plasma, the erosion of the electrodes in plasma jets, glow discharges, and GAD is a drawback in industrial applications.

Experimental

Plasma generation was accomplished via a capacitively connected device that combines an E-bend, a rectangular waveguide (WR975), with the tapered end of the waveguide coupled to a quartz tube holder. The microwave energy was supplied from a (915 MHz, 75 kW) magnetron. The microwave power transmitted to the discharge region, referred to as the absorbed microwave power, was estimated by deducting the reflected power from the incident power. A directional coupler connected to a high-power meter was used to determine incident power and reflected power. A metal igniter ignites the microwave plasma. The reactor was preheated to 600 °C with a mixture of CH_4 and air plasma, and no balancing gas was used. Likewise, the CO_2 and CH_4 mixture was preheated by passing it to the refractory of the reactor before being introduced into the reactor through the quartz holder gas inlet as a swirling gas. The product gas was determined using a syngas analyzer (Fe Gas Analyzer from Fuji Electric Co., Ltd. Korea Republic). The experimental conditions for the Plasma-GHG reforming are as follows: The inlet CH_4/CO_2 molar ratio was 0.25–1, the microwave power was 20–45 kW, and the flow rate was 120–180 lpm. Reforming occurs in a tubular reactor with an inner diameter of 280 mm and a length of 1500 mm, respectively. The gas temperature was measured using two thermocouples along the length of the reactor. Product gas composition from the reactor was measured using a syngas analyzer. The reactor's exit gas is cooled by passing it through a heat exchanger. Soot is separated from the reformer's exit gas via the soot separator.

Only the output gas after the heat exchanger was sampled in this investigation. A water gas shift (WGS) reactor could also be used to maximize hydrogen yield. A compressor increases the gas pressure to the required level for the pressure swing adsorper (PSA), so as to enable the PSA extracts H_2 from the output gas. However, a WGS reactor and PSA were not employed in this study. The plasma-GHG reforming microwave plasma torch schematic is shown in Fig. 1.

The performance parameters of the reactor were determined as a function of the microwave power absorbed, CH_4, and CO_2 conversion degree. The hydrogen production rate g (H_2)/h represents the hydrogen produced over time (in grams/hour). The energy yield g (H_2)/kWh is the ratio of the hydrogen production rate to the absorbed microwave power or the amount of hydrogen generated per kWh of energy. Methane and carbon dioxide conversion degrees (%) are described as follows:

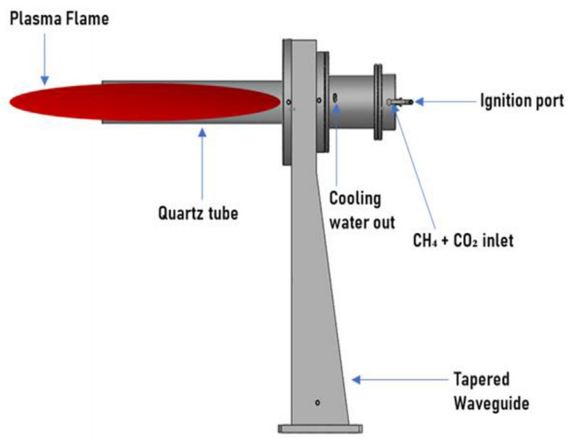

Fig. 1 – Microwave plasma torch.

$$CH_4\text{conversion } (\%) = \frac{molCH_{4converted}}{molCH_{4feed}} \text{ x } 100 \qquad (1.12)$$

$$CO_2\text{conversion } (\%) = \frac{molCO_{2converted}}{molCO_{2feed}} \text{ x } 100 \qquad (1.13)$$

Results and discussion

Absorbed microwave power effect

Figs. 2 and 3 represent the impact of absorbed microwave power on CH_4 and CO_2 conversion. The degree of CH_4 and CO_2 conversion based on absorbed microwave power at CH_4/CO_2 ratios of 0.5 and 1, and 180 lpm were examined. CH_4 conversion increased with increasing absorbed microwave power; CH_4 conversion ranged between 78.94 and 92.35% while varying absorbed microwave power between 25 and 40 kW for a CH_4/CO_2 Due to the oxygen species increase at a CH_4/CO_2 ratio of 0.5, a higher CH_4 conversion was realized at a CH_4/CO_2 ratio of 0.5 compared to a CH_4/CO_2 ratio of 1. The density of active species, such as *O, *OH, *H, and *CH_x ($x = 3,2,1$), owing to the increase in microwave energy, is likely the main reason for the greater CH_4 conversion at a lower CH_4/CO_2 [39]·

Similarly, it was observed that increasing microwave power and CH_4/CO_2 ratios affected CO_2 conversion significantly. CO_2 conversion ranged from 66.7 to 79.5% at a CH_4/CO_2 ratio of 0.5 as the absorbed microwave power was raised from 25 to 40 kW. Also, conversion ranged from 78.63% to 92.09% at a CH_4/CO_2 ratio of 1 as absorbed microwave power rose from 25 to 40 kW. The higher conversion degree of CO_2 and CH_4 with microwave power increases syngas mass yields and, thus, hydrogen. Furthermore, the results show that CO_2 conversion is less than CH_4 conversion. The lower conversion of CO_2 is owing to the GHG reforming characteristics at low plasma power, which allows for the following recombination reactions: $CO + O \rightarrow CO_2$ and $C + O_2^* \rightarrow CO_2$, resulting in a reduced CO_2 conversion. The conversion of oxygen and ions in the plasma to CO_2 shows that the reforming reaction relies heavily on microwave power regardless of CH_4/CO_2 ratio.

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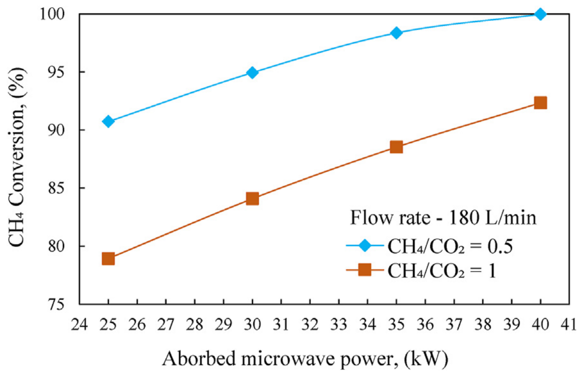

Fig. 2 – Methane conversion as a function of absorbed microwave power.

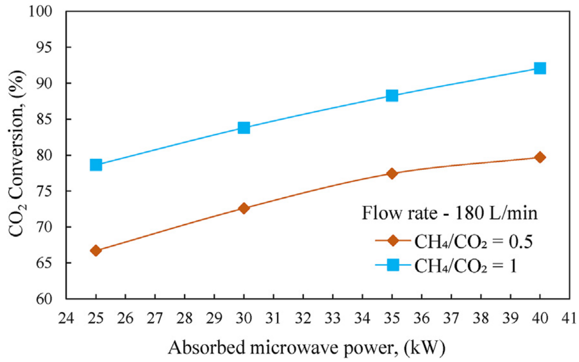

Fig. 3 – Carbon dioxide conversion as a function of absorbed microwave power.

As shown in Fig. 4, the hydrogen production rate increased with rising absorbed microwave power. The CH_4/CO_2 ratio minimally influences the hydrogen production rate at absorbed microwave power below 35 kW for CH_4/CO_2 ratios 0.5 and 1. The optimal hydrogen production rate was recorded at 40 kW, and the CH_4/CO_2 ratio of 1 was 1649 g (H_2)/h. It is important to state that the intensity of microwave energy as absorbed microwave power increases can enhance the collision of gas molecules and active species, which catalyzes the reaction.

Meanwhile, energy yield increased with increasing absorbed microwave power at a CH_4/CO_2 ratio of 0.5 from 36.4 g (H_2)/kWh at 25 kW to 39.4 g (H_2)/kWh at 35 kW; however, beyond 35 kW, energy yield decreased to 37.4 g (H_2)/kWh at 40 kW. Meanwhile, energy yield improved with increasing microwave power from 25 to 40 kW, resulting in an energy yield of 36.48–41.22 g (H_2)/kWh at CH_4/CO_2 of 1. The energy yield as a function of absorbed microwave power at different CH_4/CO_2 ratios suggests that both absorbed microwave power and CH_4/CO_2 ratio account for the energy yield of the plasma-GHG reforming process.

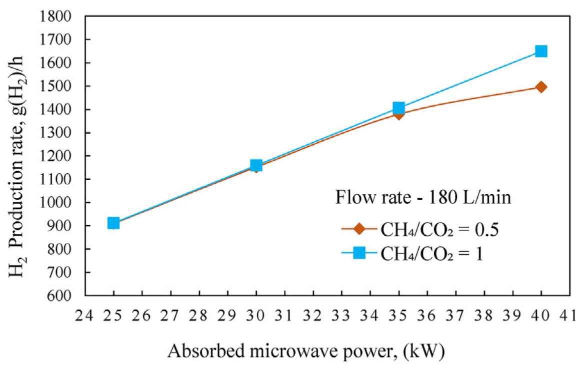

Fig. 4 – Hydrogen production rate as a function of absorbed microwave power.

CH_4/CO_2 ratio effect

The conversion degree of CH_4 and CO_2 are shown in Figs. 5 and 6 based on the following experimental conditions: CH_4 and CO_2 total flow rate was 180 lpm, at an absorbed microwave power of 30 kW and 40 kW, respectively, while varying CH_4/CO_2 ratio of 0.25–1. CH_4 conversion decreased from 99.99% to 84.08% as CH_4/CO_2 ratio varies from 0.25 to 1 at 30 kW. Comparably, CH_4 conversion decreased from 99.99 to 92.4% at 40 kW. Under the stated conditions, increasing CH_4/CO_2 ratio decreases CH_4 conversion. Thus, implying that a greater CO_2 concentration in the CH_4/CO_2 ratio is favorable to CH_4 conversion.

However, under the same conditions as CH_4 conversion, CO_2 conversion increased from 56.9 to 83.8% at 30 kW, and CH_4/CO_2 ratios of 0.25–1, respectively. At 40 kW, CO_2 conversion increased from 56.9 to 92.1% at CH_4/CO_2 ratios of 0.25 and 1, respectively. As the CH_4/CO_2 ratio rises, the CH_4 conversion

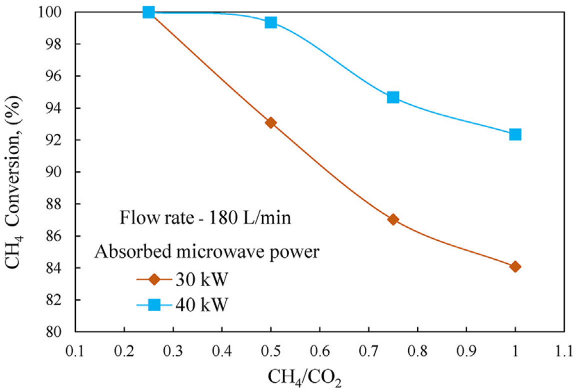

Fig. 5 – Methane conversion as a function of CH_4/CO_2 ratio.

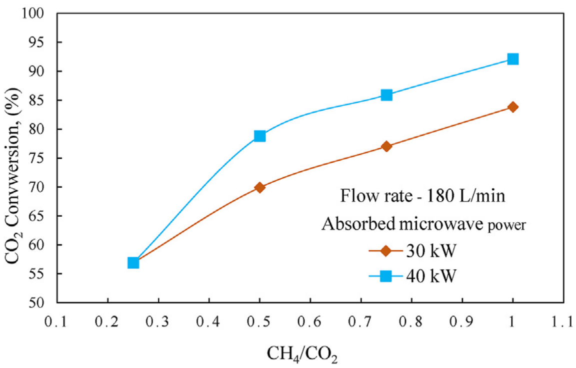

Fig. 6 – Carbon dioxide conversion as a function of CH_4/CO_2 ratio.

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degree decreases, and the CO_2 conversion degree increases; this shows that the ratio of the exit gas from plasma-GHG reforming can be varied to suit industrial purposes by adjusting the CH_4/CO_2 ratio as appropriate. Additionally, these findings imply that adding other hydrocarbon materials might cause the CO_2 microwave plasma torch to produce new materials while reducing the amount of CO_2.

Figs. 7 and 8 represent the hydrogen production rate and energy yield as a function of the CH_4/CO_2 ratio. The total flow rate was 180 lpm, CH_4/CO_2 ratio was varied from 0.25 to 1, and absorbed microwave power of 30 and 40 kW. At 40 kW and a CH_4/CO_2 ratio of 0.25–1, the hydrogen production rate rose from 936 to 1649 g (H_2)/h. It was lower at 30 kW; the hydrogen production rate increased from 936 to 1159 g (H_2)/h. Likewise, energy yield increased from 31.2 to 38.63 g (H_2)/kWh at 30 kW as CH_4/CO_2 ratio increased from 0.25 to 1. Meanwhile, at 40 kW, the energy yield increased from 23.4 to 41.23 g (H_2)/kWh.

The plasma-GHG reforming process operated smoothly throughout the experiment. Below the 0.75 CH_4/CO_2 ratio and the earlier stated experimental conditions, the amount of soot produced during the reforming process was negligible; however, with a CH_4/CO_2 ratio of 1 or greater, soot was observed in the reactor, which a soot separator can handle. Little or no soot production makes gas cleaning easier while preventing soot from covering the microwave torch quartz tube, which can diminish system efficiency. The CH_4/CO_2 ratio significantly influences the amount of soot produced in the reforming process. However, under the tested conditions, the soot produced was insufficient to destabilize or extinguish the plasma flame.

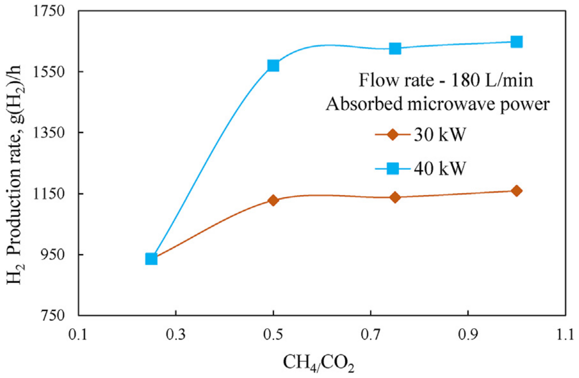

Fig. 7 – Hydrogen production rate as a function of CH_4/CO_2 ratio.

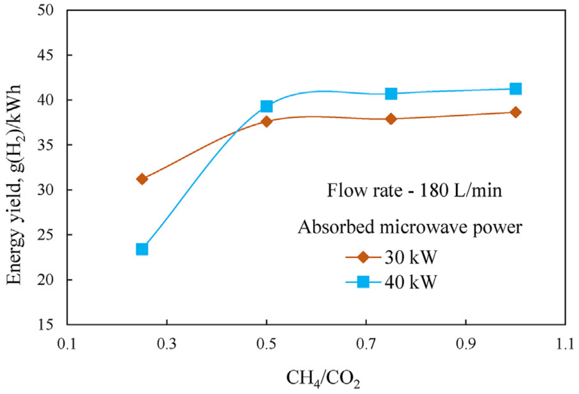

Fig. 8 – Energy yield as a function of CH_4/CO_2 ratio.

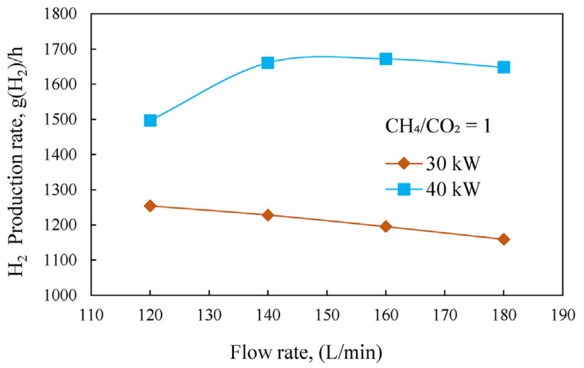

Fig. 9 – Hydrogen production rate as a function of flow rate.

Soot formation occurs predominantly due to a shift in the reaction mechanism when the CH_4/CO_2 ratio changes. When the CH_4/CO_2 ratio is less than 0.75, per the following reaction: $2CH_4 + 3CO_2 \rightarrow 5CO + 4H_2 + O$, the reaction mechanism suggests the presence of excess oxygen; the reaction route varies as the concentration of oxygen in the reaction decreases due to a higher CH_4/CO_2 ratio as indicated by the subsequent reaction: $3CH_4 + 2CO_2 \rightarrow 4CO + 6H_2 + C_{(S)}$.

Flow rate effect

The effect of the total gas flow rate on CH_4, CO_2 conversion degree, hydrogen production rate, and energy yield based on a CH_4/CO_2 ratio of 1, absorbed microwave power of 30 and 40 kW, and flow rate from 120 to 180 lpm is discussed in this section. CH_4 and CO_2 conversion decreased as the gas flow rate increased. The highest CH_4 and CO_2 conversion of 99.99% and 99.98% were obtained at 120 lpm and 40 kW. Furthermore, the hydrogen production rate and energy yield decreased with increasing gas flow rate. As depicted in Fig. 9, at 30 kW hydrogen production rate decreased from 1254 to 1159 g (H_2)/h, matching a flow rate of 120 and 180 lpm, respectively. However, at 40 kW, the hydrogen production rate initially increased with an increased gas flow rate from 1497 g (H_2)/h at 120 lpm to 1672 g (H_2)/h at 160 lpm but decreased to 1648 g (H_2)/h at 180 lpm at CH_4/CO_2 ratio of 1. Accordingly, the energy yield

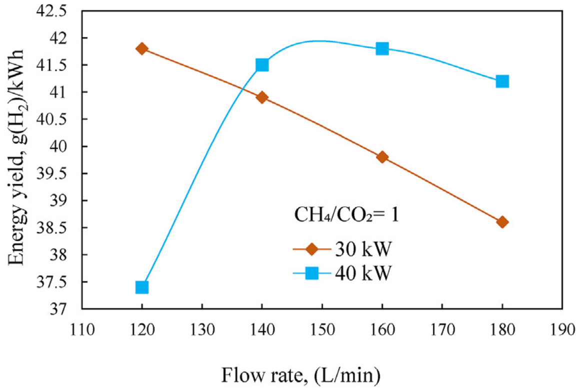

Fig. 10 – Energy yield as a function of flow rate.

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Table 2 – Comparison of energy yield and hydrogen production rate.

Production Method	Initial Composition	Production Rate g (H_2)/h	Energy Yield g (H_2)/kWh	References
Microwave (915 MHz) Plasma without catalyst	$CH_4 + H_2O$	2247	70	[40]
Metal Cylinder based MPS	$CH_4 + CO_2 + H_2O$	180	42.9	[41]
Conventional steam reforming of methane (Catalyst)	$CH_4 + H_20 + air$		60 Established industrial process	[42]
Metal Cylinder based MPS	$CH_4 + N_2$		85	[43]
Microwave (2.45 GHz) Plasma	$CH_4 + CO_2$	240	41.4	[44]
Metal Cylinder based MPS	$CH_4 + CO_2$	89	26	[44]
Dielectric Barrier Discharge	$CH_4 + air$		6.7	[45]
Gliding arc	$CH_4 + H_20 + air$		40	[46]
Electron Beam Radiolysis	$CH_4 + H_2O$		3.6	[47]
Microwave (2.45 GHz) Plasma With catalyst	$CH_4 + CO_2$	177	59.1	[48]
Microwave(915 MHz) Plasma without catalyst	$CH_4 + CO_2$	1672	41.8	This work

Table 3 – Comparison of 1 kg hydrogen production cost for SMR versus Plasma reforming.

Categories	SMR		Plasma-GHG reforming	
	Cost ($)	Percent (%)	Cost ($)	Percent (%)
Natural gas/CO_2	2.86	52.80%	2.86	58.08%
Water	0.07	1.30%	0.07	1.46%
Electricity	0.25	4.60%	0.37	7.60%
Labor	0.31	5.70%	0.18	3.58%
maintenance	0.31	5.70%	0.14	2.87%
Depreciation	1.62	29.90%	1.3	26.41%
Sum	5.42	100.00%	4.92	100.00%

Hydrogen station grade (100 kg/day) would cost 7.5 USD/kg.
Source (SMR cost): JNK Heater Co., Ltd, South Korea.

followed a similar trajectory by increasing from 37.4 g (H_2)/kWh at 120 lpm to 41.8 g (H_2)/kWh at 160 lpm before decreasing to 41.2 g (H_2)/kWh at 180 lpm as illustrated in Fig. 10.

Plasma flame elongation due to flow rate increase suggests that more residence time for gas to remain around the flame is offered, which explains the higher conversion due to the flow rate increase at 40 kW. However, it is noteworthy that the hydrogen production rate and energy yield depended on a synergy between the flow rate and absorbed microwave power rather than the flow rate alone. The highest hydrogen production rate and energy yield achieved were 1672 g (H_2)/h and 41.8 [g (H_2)/kWh] at a total gas flow rate of 160 lpm, 40 kW absorbed microwave power, and CH_4/CO_2 ratio of 1. Generally, GHG conversion was observed from temperatures $\geq 350 < 800$ °C. The results of this investigation suggest that plasma-GHG reforming for the mass production of hydrogen is viable.

Conclusion

The 915 MHz microwave plasma source was used to produce a non-electrode microwave plasma for GHG reforming in a tubular reformer at atmospheric pressure without a catalyst. The experimental results show that CH_4 and CO_2 decomposition form carbon powder (soot), which is observable from the reformer's viewport. The exit gas from plasma-GHG reforming includes H_2, CO, unconverted CO_2, CH_4, and soot generated based on the CH_4/CO_2 ratio.

The influence of absorbed microwave power on the conversion of CH_4 and CO_2, hydrogen production rate, and energy yield were examined. The study found that absorbed microwave power significantly affected the conversion degree of all tested parameters for the CH_4/CO_2 ratio and total flow rate. The CH_4/CO_2 ratio also substantially affected CH_4 conversion, producing a better result at a ratio of 0.5 than 1. In the case of CO_2 conversion, the converse was true. There was less CO_2 conversion when the CH_4/CO_2 ratio was 0.5 than when the CH_4/CO_2 ratio was 1. The study found that CH_4/CO_2 ratio significantly affects soot formation.

Also, at a gas flow rate of 160 lpm, 40 kW absorbed microwave power, and a CH_4/CO_2 ratio of 1, the best hydrogen production rate and energy yield achieved was 1672 g (H_2)/h and 41.8 g (H_2)/kWh, respectively. At the same time, CH_4 and CO_2 conversion under the same experimental conditions were 95.59% and 95.4%, respectively. The experimental results presented in this work demonstrate that plasma-GHG reforming for mass hydrogen production is feasible at high gas flow rates and microwave power.

In reference [48]' the presence of a catalyst increased hydrogen production rate and energy yield. Although the lack of a catalyst resulted in a decreased hydrogen production rate and energy yield in this work, the method is not susceptible to catalyst deactivation, even at high power. Table 2 compares the existing work's hydrogen production rates and energy yields.

The plasma reforming method in this study shows significant potential regarding its industrial applicability because catalyst deactivation can be substantially reduced by adjusting the inlet CH_4/CO_2 ratio. Due to its small equipment footprint, the microwave plasma reformer can be used in

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various applications, including onboard and compact systems.

The costs of producing 1 kg of hydrogen using the plasma reformer and SMR are contrasted in Table 3. Although the plasma reforming method employed in this study appears marginally less expensive, utilizing a microwave plasma torch still has significant room for improvement. Future studies related to this work will concentrate on extracting CO from the exit gas to have an almost carbon-neutral fuel for gas engines.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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