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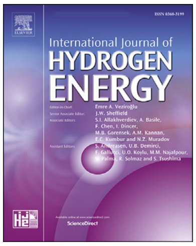

# Plasma steam methane reforming (PSMR) using a microwave torch for commercial-scale distributed hydrogen production

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## HIGHLIGHTS

- Commercial-scale distributed $H_2$ production via PSMR was experimentally tested.
- Realized high $H_2$ production rate and significant energy yield (>70 [g($H_2$)/kWh]).
- Microwave torch utilized for simultaneous PSMR and WGSR in a single reformer.
- Experimental results supported by ASPEN plus simulation.

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## ABSTRACT

Although large-scale hydrogen production through conventional steam methane reforming (SMR) is available at an affordable cost, there is a shortage of hydrogen pipeline infrastructure between production plants and fueling stations in most places where hydrogen is needed. Due to the difficulties of transporting and storing hydrogen, onsite hydrogen production plants are desirable. Microwave plasma torch-based methods are among the most promising approaches to achieving this goal.

The plasma steam methane reforming (PSMR) method discussed here has many benefits, including a high energy yield, a small carbon footprint, real-time fueling because of the short start-up time (<10 min), and the absence of expensive metal-based catalysts. Methane reforming and water gas shift reaction (WGSR) co-occur in the method advanced without a separate WGSR to achieve a high $H_2$ yield.

This study examines an experimental investigation of commercial-scale hydrogen production through PSMR utilizing a microwave torch system. The optimum results obtained showed that the hydrogen production rate was 2247 [g($H_2$)/h], and energy yield was 70 [g($H_2$)/kWh] of the absorbed microwave power. An assessment of the results indicated a similar trend to that of simulated data (ASPEN Plus). The experimental results presented in this paper demonstrate the potential of a catalyst-free PSMR for commercial-scale hydrogen production.

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## Introduction

A substantial amount of hydrogen, ammonia, methanol, acetylene, and other chemicals used in industrial processes come from sizeable central production plants through natural gas reforming or steam methane reforming [1,2]. The first step in the reforming process for hydrogen production involves manufacturing synthesis gas (syngas), a mixture of hydrogen and carbon monoxide [3], and a negligible amount of impurities.

The energy density of hydrogen is relatively high (120–142 MJ/kg); hence, hydrogen is a source of energy for fuel cell activation and heating [4]. The relevance of hydrogen as an efficient and environmentally friendly energy carrier is widely acknowledged. Presently, a considerable proportion of the world's energy supply emanates from carbon-based fuels [5]. Transitioning to a hydrogen fuel-based economy is essential for minimizing the adverse effects of fossil-fuel reliance, such as global warming, air pollution, and potential health and environmental implications. Hydrogen production through natural gas reforming with $CO_2$ capture could help to achieve this goal [6]. Several published studies have examined hydrogen production from carbon-based fuels and other feedstocks [7–9].

Diverse hydrogen production technologies exist, including coal or biomass gasification, water electrolysis, and natural gas reforming [10,11]. However, the most widely used methods for producing hydrogen are natural gas (methane) reforming, mainly steam methane reforming (SMR) with steam as an oxidizer, and dry or auto thermal reforming (ATR) with oxygen as an oxidizer [12]. High purity hydrogen production via the steam reforming of natural gas can be categorized into feedstock pretreatment, steam reforming reaction, and hydrogen purification [13]. SMR is highly endothermic and thermodynamically advantageous at high temperatures and low pressures. The process transforms steam and methane into hydrogen-rich syngas. A set of parameters, including high temperature, low pressure, and a high steam-to-methane (S/C) ratio, increases conversion while reducing soot formation. In contrast, a low S/C ratio is advantageous in terms of energy efficiency and economics. Conventional SMR occurs in externally heated packed bed reactors employing a Ni-based catalyst [14].

The coupling of SMR with non-catalytic partial oxidation (POX) in a single chamber is described as auto thermal reforming. In this process, methane interacts with oxygen and steam, and the exothermic POX reaction provides the heat necessary for the SMR. The reaction temperature and the $H_2$/CO ratio in the exhaust stream can be controlled by varying the amount of the oxidizer utilized. ATR temperature ranges between 900-1150 °C, with reformer pressures ranging between 1-80 bar [15,16]. For the POX reaction, oxygen may be provided in two forms: pure ($O_2$-blown ATR) or air (air-blown ATR).

Gasification is another means of manufacturing syngas from coal or biomass. At high temperatures and pressures, the fuel react with oxygen and steam during the gasification process. Entrained-flow gasifier is the prevalent commercial gasifier, with notable manufacturers including GE and Shell Gasifiers. Gasification temperatures range from 1350 to 1500 °C in a GE gasification plant, with pressures reaching 100 bar [17].

POX of heavy hydrocarbons such as diesel and residual oil can also produce hydrogen, and this technique can produce hydrogen-rich syngas from any compressive and pumpable hydrocarbon. The reaction proceeds once partly burned fuel combines with a stoichiometric amount of air in a reactor. POX is an exothermic reaction that progresses in the presence or absence of a catalyst. Large-scale reactors without a catalyst typically run at temperatures ranging from 1150 to 1500 °C and pressures from 25 to 80 bar. The catalytic POX reaction occurs at relatively low temperatures and pressures ranging from 780 to 900 °C and 25–35 bar [18]. Pure oxygen is utilized to avoid the requirement for downstream nitrogen processing.

The usage of SMR, ATR, and POX for hydrogen production has some benefits and drawbacks: SMR offers the highest hydrogen production rate, but it requires meticulous thermal control and cannot be used for all feedstocks. ATR has the advantage of providing both easy thermal control and quick response; nevertheless, ATR requires careful design to maintain the exothermic and endothermic process stability during starting up and while altering the system's flow rate. As a result, the control mechanism is quite sensitive. Meanwhile, POX offers a rapid and dynamic response with less stringent thermal management because the reaction is exothermic; however, the method produces the least amount of hydrogen [19]. Thus, SMR is the most cost-effective and dominant method for producing hydrogen owing to its high yield. Major methane conversion processes have been summarized in Table 1 [20].

Studies suggest that the SMR enthalpy of reaction increases as reaction temperature increases, while Gibb's energy reduces with increasing temperature [21]. The equilibrium of equation R5 is temperature-dependent but independent of pressure; a high carbon monoxide conversion occurs at low temperatures and relatively high steam concentration, but a practical reaction rate requires relatively high temperatures. The SMR process uses a water gas shift reaction (WGSR) to reduce carbon monoxide while increasing hydrogen yield. The WGSR is a two-step process that combines a high-temperature shift (HTS) at 320–420 °C and a low-temperature shift (LTS) at 200–250 °C connected in series to take advantage of both the thermodynamics and kinetics of the reaction [22]. The dual step WGSR requires a different catalyst for each step and thus makes the design of a compact system challenging.

In SMR, the methane pyrolysis step is the rate-determining step [23]. Furthermore, at a high temperature (>800 °C) within a methane reformer, steam concentration lacks a significant effect on the reaction rate. Therefore, the temperature slope along the length of a plasma reformer gives rise to diverse thermal regions within the reformer, and the temperature profile of the plasma torch makes simultaneous methane reforming and WGSR conducive [24].

Steam methane reforming (SMR) reaction mechanisms and pathways are well understood, yet they remain a debatable topic of discussion. Most researchers suggest that the reaction mechanism (equation R4 and R5) indicates that methane reforming is the initial step accompanied by WGSR,

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**Table 1 – Methane conversion processes.**

| Process | $\Delta G^0$ (298)/kJ $mol^{-1}$ (T = Temperature) | Energy Input/mol$H_2$ | $CO_2/H_2$(kg/kg) | Reaction No. |
|---|---|---|---|---|
| **Pyrolysis** | | | | |
| $CH_4 \rightarrow C_{(S)} + 2H_2$ | 75–0.08 T | 37.5 | | R1 |
| **Dry Reforming** | | | | |
| $CH_4 + CO_2 \rightarrow 2CO + 2H_2$ | 247–0.26 T | 123.5 | 5.5 | R2 |
| **Partial Oxidation** | | | | |
| $2CH_4 + O_2 \rightarrow 2CO + 4H_2$ | −77.2 + 0.3 T | −18 | 7.3 | R3 |
| **Steam Reforming** | | | | |
| $CH_4 + H_2O \rightarrow CO + 3H_2$ | 206–0.22 T | 68 | | R4 |
| $CO + H_2O \rightarrow CO_2 + H_2$ | −41 + 0.04 T | −41 | | R5 |
| $CH_4 + 2H_2O \rightarrow CO_2 + 4H_2$ | 165–0.17 T | 41.2 | 5.5 | R6 |

which results in the overall reforming reaction (Equation R6). The overall reforming reaction is also likely to be the first step, followed by the reverse WGSR to obtain the "complete" SMR reaction [24,25].

Thermal, nonthermal, and warm plasma have recently found application in hydrogen production experiments. In thermal plasma, the temperature of the charged (electrons and ions) and neutrals (atoms and molecules) species are in thermodynamic equilibrium. In contrast, nonthermal plasma is partially ionized gases with electron temperature far above ions and neutrals [26,27]. Warm plasma is known as transitional plasma because they share thermal and nonthermal plasma characteristics, allowing them to operate at a moderate power density while sustaining a reasonably high gas temperature (~3000 K). Consequently, relatively high-energy electrons and excited ions, atoms, or molecules permit effective and selective chemical transition [28].

Plasma can be activated at lower or higher working gas pressures. Hence, the exploitation of plasma such as dielectric barrier discharge (DBD) [29–31], corona discharge [32], and microwave discharge for hydrogen production by reforming hydrocarbons in laboratory-scale experiments [33–35]. The general reforming reactions in plasma reforming are the same as in conventional reforming; however, plasma supplies the energetic species and radicals necessary for the reforming process, typically generated from electricity.

High-energy electrons generated by nonthermal plasma expedite chemical processes that lead to hydrocarbon reforming. Nonthermal plasmas' significant benefits are their high reactivity and selectivity, low-energy consumption, and ability to function at low temperatures. Catalysts are usually required in nonthermal plasma to speed up the conversion process. In a discharge region, non-equilibrium plasma is intrinsically heterogeneous, resulting in a reduced reaction area [36]. As a result, conversion rates and throughput are low.

Nonthermal plasma and warm plasma are compact and fast, making them ideal for distributed hydrogen production. In terms of the rate and efficiency of hydrogen production, nonthermal plasma such as DBD and corona discharge are inferior to warm plasma such as microwave discharge and gliding arc discharge [37]. Consequently, the application of warm plasma for methane reforming is becoming increasingly popular.

In an atmospheric pressure microwave plasma torch, microwave provides the energy required to activate plasma in gases [38]. With the injection of steam as an oxidizer into the system, hydrogen, hydroxyl, oxygen radicals, and electrons are generated to provide a favorable environment for both reductive and oxidative processes to take place [39]. In comparison to other plasma sources, atmospheric pressure microwave plasma sources (MPS) are more effective for energy transmission. An appropriately designed MPS can transfer one hundred percent microwave power to the plasma torch [40,41]. The presence of reactive species in microwave plasma accelerates the reaction rate; thus, costly metal-based catalysts prone to impurities-induced deactivation can be avoided [42].

The benefits of microwave plasma, such as fast response time from electricity to torch [43] and its ability to operate without an electrode, make it useful for gaseous pollutant abatement and gas purification [44]. Due to numerous reactive species in the microwave plasma, compact reformers may be developed [45]. Additionally, sulfur tolerance is high when no catalysts are employed to aid the reforming process, and they can operate at lower temperatures than conventional reforming methods [46]. For instance, using $Ni/Al_2O_3$ as a catalyst, Wang et al. (2009) described an atmospheric pressure microwave plasma-assisted SMR process designed primarily for $H_2$ production. The highest methane conversion obtained at steam to methane ratio (S/C) = 1 was 91.6%, and the best obtained selectivity of $H_2$ was 95.2% at inlet S/C = 3 and 1 kW, $[CH_4]_{in}$ = 5%, and 12 slpm flow rate [47]. According to Wang et al. the drawback of the bench-scale experiment was catalyst deactivation due to the generation of a large amount of carbon powder (40–70 nm).

Similarly, D.H. Choi et al. (2015) researched steam methane reforming (SMR) using a microwave plasma torch (2.45 GHz). The parameters tested were 3 kW microwave power, S/C = 3–9, and steam flow rate of 27 lpm based on different reactor and nozzle configurations, including cylinder-type, shoppe-type nozzle, and reverse vortex flow. In the study, the hydrogen production rate of 71.3 (vol. %) and 95.3% methane conversion were the best-recorded values for the reverse vortex flow reactor. They found that configurations of the torch nozzle and gas inlet produced different results. D.H. Choi et al. (2015) demonstrated that microwave plasma might be used successfully for PSMR without adding a catalyst [48].

D. Czylkowski et al. (2016) verified the efficacy of PSMR with $CO_2$ in a waveguide-supplied metal cylinder-based microwave plasma (2.45 GHz) source operating at atmospheric pressure

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without using a catalyst, the highest absorbed microwave power was 6 kW, and the working gas flow rate ($CH_4 + CO_2 + H_2O$) was up to 9000 NL/h. The best-recorded values for hydrogen production rate and energy yield were 192 [g($H_2$)/h], and 42.9 [g($H_2$)/kWh], [49].

The method advanced by D. Czylkowski et al. (2016) supports previous investigations that show the capability of the PSMR to function reliably at high gas flow rates.

Hydrogen production technologies are steadily approaching cost and performance parity with battery and other fossil-fuel technologies, therefore, likely to disrupt the energy industry [50]. However, the adoption of these technologies has been somewhat sluggish, owing to the high initial costs of new infrastructure for accessing hydrogen supplies, storage in the medium and long term, and transportation over long distances [51]. Furthermore, these technologies operate at high pressure and relatively high temperature, along with the need for costly metal-based catalysts [52]. Meanwhile, a significant drawback to natural gas or methane reforming is rapid catalyst deactivation, usually caused by coke deposition and catalyst sintering [53], necessitating frequent catalyst replacement.

As mentioned earlier, there are diverse methods of producing hydrogen; however, most hydrogen processing methods need significant development to increase efficiency, lower capital costs, enhance reliability, and increase flexibility. Because these processes use expensive metal-based catalysts and operate at high pressure (≥100 bar) and process temperature (≥1500 °C), substantial capital investment and running costs are needed [54], which means that only large-scale hydrogen production is profitable.

This paper presents the result of an experimental investigation of PSMR utilizing a microwave torch for distributed hydrogen production, as shown in Fig. 1. The study aims to demonstrate the effectiveness of a compact, catalyst-free, atmospheric pressure microwave PSMR for commercial-scale hydrogen production by assessing the hydrogen production efficiency of the system based on parameters, such as hydrogen production rate [g($H_2$)/h], methane conversion degree (%), energy yield [g($H_2$)/kWh], and the volume of hydrogen concentration in the outlet gas (%).

## Experimental

The experimental setup for hydrogen production through PSMR in an atmospheric pressure microwave plasma is shown schematically in Figs. 1–3. The ASPEN plus flow diagram used for the process flow simulation is as depicted in Fig. 4.

The non-electrode microwave plasma system employed in the investigation consists of a microwave generator system (915 MHz, 75 kW), a WR975 rectangular waveguide of internal dimensions 247.65 mm × 123.825 mm, which include a directional coupler, and a 3-stub tuner for the microwave plasma source (MPS) impedance matching. The discharge cylindrical quartz tube with the inner and outer diameters of 79 mm, and 85 mm, respectively. And a digital mass flow meter to control the gas supply to the reactor. The absorbed microwave power ($P_A$), which refers to microwave power supplied to the discharge area, was estimated by subtracting reflected power ($P_R$) from incident power ($P_I$). A directional coupler with a bolometric head and HP power meter to

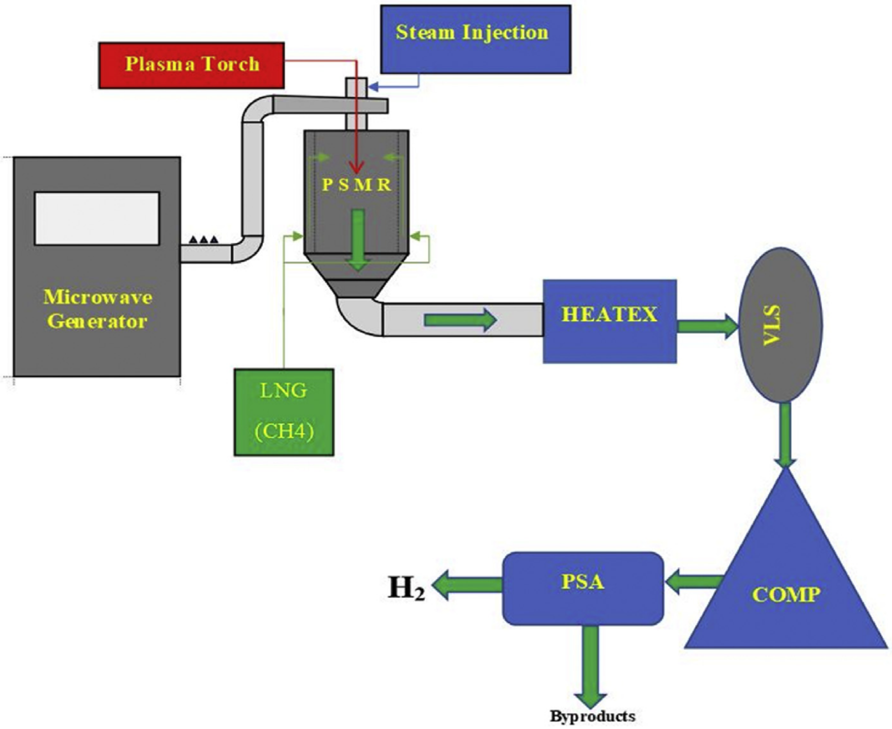

**Fig. 1 – Schematic diagram of microwave PSMR: HEATEX – heat exchanger, VLS – vapor, liquid – separator, COMP – compressor, PSA – pressure swing adsorption.**

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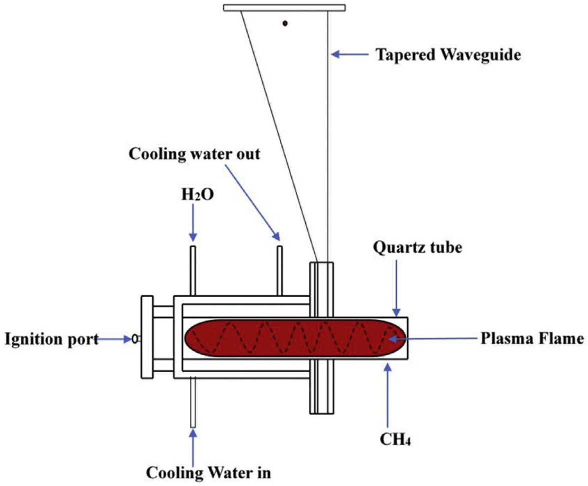

**Fig. 2 – Schematic diagram of microwave plasma torch used for the PSMR process.**

**Fig. 3 – Picture of PSMR experimental setup.**

measure $P_I$ and PR. For plasma generation, a capacitively coupled system connects the microwave generator to the reactor through an E-bend and the tapered end of the waveguide. A metal igniter was employed to initiate the microwave breakdown and plasma ignition.

The steam generator supplied steam as swirling gas through the quartz holder gas inlet to the reactor. The use of a vortex flowmeter enabled steam control. $CH_4$ gas (99.9%) was introduced into the reformer gas inlet, as shown in Fig. 1. The product gases were characterized and measured with a syngas analyzer (Fe Syngas Analyzer).

The experimental conditions were as follows: the inlet $H_2O/CH_4$ molar ratio was 3–6, microwave power varied from 15 to 50 kW, and the total flow rate was 240–420 lpm. The reforming operation takes place in a plug flow reactor, and the inner diameter and length of the reactor are 0.28 m and 2.27 m, respectively. Furthermore, to monitor the temperature along the length of the reactor (hereafter referred to as reformer), thermocouples were used.

The reformer's exit gas is routed through a heat exchanger (HEATEX) to recover energy and gas cooling. And to separate $H_2O$ from the reformer outlet gas, the vapor-liquid-separator (VLS) is utilized. The purpose of the pressure swing adsorption (PSA) system is to extract $H_2$ from the output gas, and the function of the compressor (COMP) is to elevate the gas pressure to an appropriate level for the PSA to operate. The experimental results provided only analyzed the output gas from the reformer and heat exchanger. Fig. 5 show a block diagram of the PSMR.

## Results and discussion

### *Hydrogen production efficiency of PSMR*

The efficiency of hydrogen production through PSMR reported in this paper is defined by the following parameters; The hydrogen production rate $g(H_2)/h$, which shows the mass (in grams) of hydrogen produced over time (per hour). The energy yield $[g(H_2)/kWh]$ is the ratio of hydrogen production rate to absorbed microwave power or the ratio of hydrogen mass-produced with the energy of 1 kWh. A syngas analyzer facilitated the measurement of product gas. The parameters were determined as a function of absorbed microwave power, $H_2O_{(g)}$, and $CH_4$ flow rate. S/C ratio is defined as the steam to methane ratio. And methane conversion degree (in %) is described as $1 - (CH_{4(in)} - CH_{4(out)}/CH_{4(in)}$, where: $CH_{4(in)}$ is initial $CH_4$, and $CH_{4(out)}$ is unconverted $CH_4$.

### *Effect of absorbed microwave power*

The influence of absorbed microwave power on methane conversion is shown in Fig. 6. For this reaction, the S/C ratio was set at 3, with $CH_4$ and $H_2O_{(g)}$ at 60 lpm and 180 lpm, respectively. We varied microwave power from 15 to 50 kW. Predictably, owing to the endothermic nature of the SMR, methane conversion increased with absorbed microwave power, from 74.6% at 17 kW to 85.7% at 27 kW and then 97.3% at 37 kW, a further methane conversion of 97.7% at 47 kW. But beyond 47 kW, there was no other methane conversion without increasing the flow rate of reactants.

The lack of additional methane conversion above 47 kW under the stated conditions that the process has reached saturation. However, the experimental values of methane conversion were lower than the simulation results for the same conditions. It is possible to improve the experimental results by slightly lowering the temperature gradient from 500 °C–650 °C along the reformer's length and enhancing gas mixing. Thermodynamics does not suggest high methane conversion for steam methane reforming at ambient temperature and atmospheric pressure. Hence, plasma characteristics, particularly radicals, electron energy, and density, determine methane conversion's thermodynamic limitations.

As depicted in Fig. 7, we evaluated hydrogen production rate as a function of absorbed microwave power with microwave power varying between 20-50 kW under the following conditions: the flow rate of $CH_4$, and $H_2O_{(g)}$ was set at 60 lpm and 180 lpm, respectively. The hydrogen production rate increased

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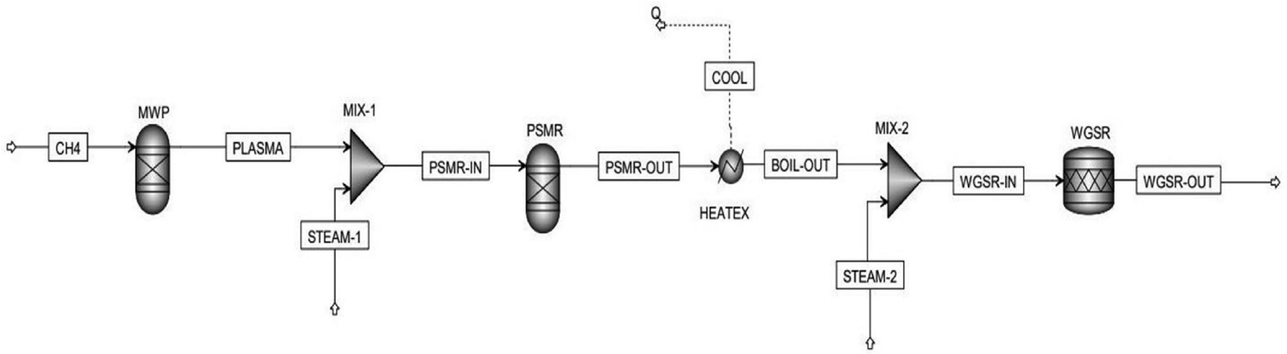

**Fig. 4 – Aspen plus process flow diagram.**

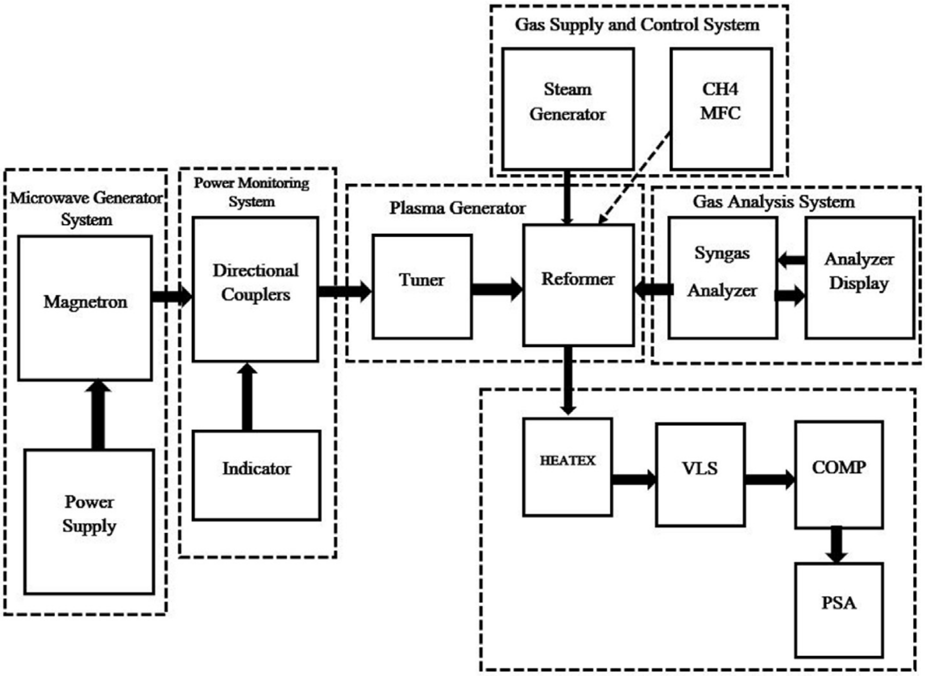

**Fig. 5 – PSMR block diagram.**

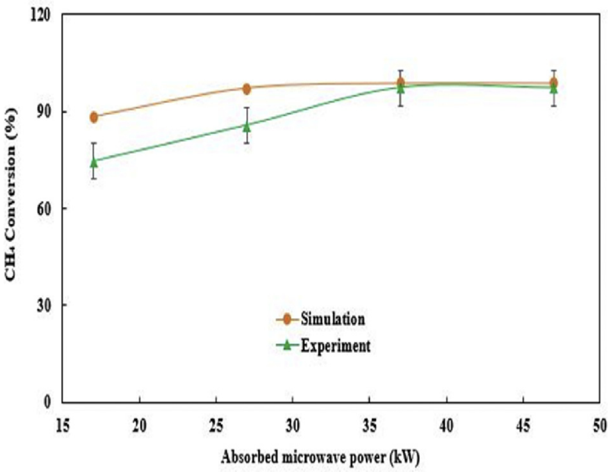

**Fig. 6 – Methane Conversion as a function of Absorbed Microwave Power.**

from 1532 to 2247 g($H_2$)/h, with absorbed microwave power rising from 27 to 32 kW. Further increase in the absorbed microwave power did not result in a higher $H_2$ yield. However, simulation results produce a higher $H_2$ yield than the experimental result under the same conditions. By optimizing the system's operating conditions, it is possible to improve $H_2$ yield. The increase in hydrogen production rate with increasing microwave power is attributable to the intensification of the electric field around the discharge zone, induced by higher absorbed microwave power, followed by a rise in gas temperature due to heat transfer and reactivity of the energetic species.

The increase in heat transfer to the surrounding gases due to the increase in absorbed microwave power ensures the maximum electron and energy density necessary for the forward path of the reforming reaction. The result reveals that higher absorbed microwave power is thermodynamically beneficial for hydrogen production rate and methane conversion. There is reasonable agreement at a higher power

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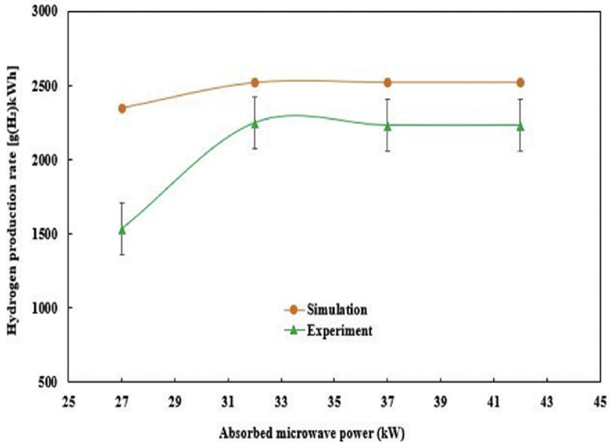

**Fig. 7 – Hydrogen Production rate as a function of absorbed microwave power.**

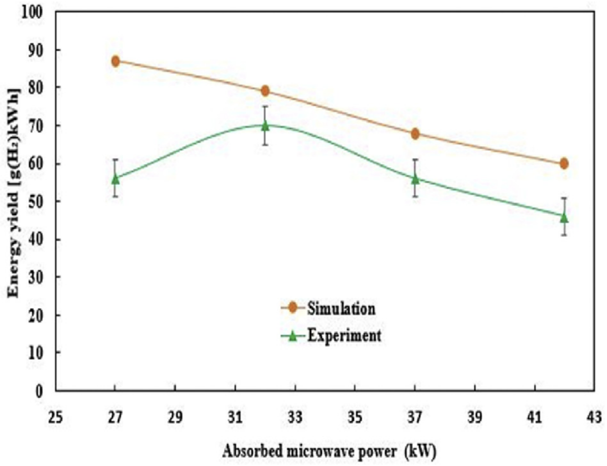

**Fig. 8 – Energy yield as a function of absorbed microwave power.**

between experimental results and the simulation values, which could improve with a better steam quality.

The results obtained for hydrogen production rate as a function of absorbed microwave power compared with simulation values show some disparity, as seen in Fig. 7. A slight reduction in the temperature slope from 500 °C–650 °C along the length of the reformer, as well as improved gas mixing in the plasma discharge zone, would almost certainly yield a better result.

Fig. 8 illustrates the effect of various absorbed microwave power on energy yield. The experimental conditions for energy yield as a function of absorbed microwave power are the same as in Fig. 7. In contrast to the hydrogen production rate and methane conversion, the energy yield decreased with increasing absorbed microwave power. Initially, energy yield increased from 56 [g($H_2$)/kWh] to 70 [g($H_2$)/kWh] at 27 kW and 32 kW respectively, then it decreased to 54 [g($H_2$)/kWh] and 46 [g($H_2$)/kWh] at 37 kW and 42 kW respectively. The result shows that the system's energy yield depended on absorbed microwave power and gas flow rate. The energy yield decreased accordingly as the absorbed microwave power increased with constant methane and steam flow rate. Thus, we could deduce an appropriate or optimal value for the absorbed microwave power regarding hydrogen production rate.

### *Effect of steam/methane ratio*

The impact of S/C on methane conversion is represented in Fig. 9. For this experiment, the absorbed microwave power was 32 kW, $CH_4$ was 60 lpm, and $H_2O_{(g)}$ was varied between 180 and 360 lpm, respectively. The result shows conversion of methane varied with increasing S/C ratio, rising noticeably from 96.6% at S/C = 3–98.8% at S/C = 4 and slightly increasing to 99.8% at S/C = 5. The S/C ratio significantly influences methane conversion, which increases as the S/C ratio increases from 3 to 5. The results reveal that under the specified conditions, methane conversion was nearly 100%. Simulation values under the same conditions exhibit a similar pattern, with higher Methane conversion at S/C = 3 and 4. Though SMR stoichiometry reactions show that 1 mol of $H_2O_{(g)}$ is needed for 1 mol of $CH_4$, in practice, the reaction is carried out with a higher S/C ratio, in the range 2.5–3 to avoid carbon formation [51].

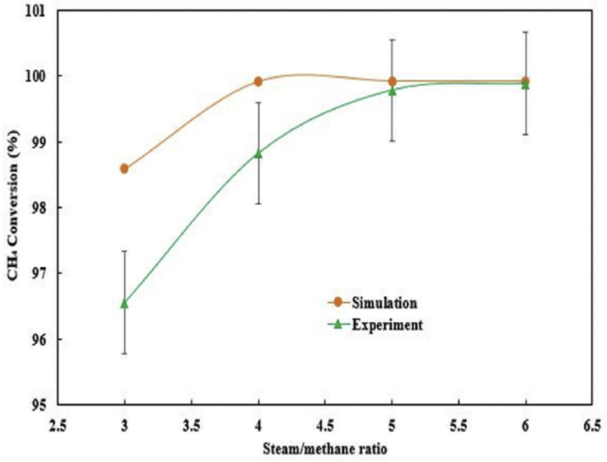

**Fig. 9 – Methane Conversion as a function of Steam/Methane Ratio.**

Hydrogen production via microwave steam plasma requires a stable plasma flame. We observed a steady plasma flame in the quartz discharge tube under all experimental settings specified in this study. An evaluation of the result shows that the S/C ratio is a crucial factor influencing methane conversion, as methane conversion increases with increasing S/C ratio. However, beyond S/C = 4, methane conversion showed no significant conversion; this implies a reduction in the energetic parameters in the discharge zone. With a higher S/C ratio, there is a higher increase in electric field in the reaction zone, which results in the extension of the plasma flame and collision time. The PSMR's steady operation at high gas flow rate proves the system's capacity for large-scale methane reforming. The optimal results obtained for the energetic parameters of hydrogen production through plasma steam methane reforming, namely, hydrogen production rate and energy yield, are 2247 [g($H_2$)/h] and 70 [g($H_2$)/kWh] respectively. We repeated the experiment several times to ensure the results were reliable. The energy yield in this study was calculated based on the

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absorbed microwave power by the reforming process rather than the overall electrical power used. Apart from the plasmatron with a catalyst and methane cracking with the metal cylinder-based microwave plasma source, the PSMR described in this study recorded one of the best results among the listed methane reforming methods in Table 2. Table 2 shows a comparison of energy yield and hydrogen production rate of various hydrogen production methods.

The impact of steam to methane ratio (S/C) on methane conversion is highlighted in Fig. 9. For this experiment, the absorbed microwave power was 32 kW, $CH_4$ and $H_2O_{(g)}$ were 60 lpm and 180 lpm, respectively. The result shows conversion of methane varied with increasing S/C ratio, rising noticeably from 96.6% at S/C = 3–98.8% at S/C = 4 and slightly increasing to 99.8% at S/C = 5. The S/C ratio significantly influences methane conversion, which increases as the S/C ratio increases from 3 to 5. The results reveal that under the specified conditions, methane conversion was nearly 100%. Simulation values under the same conditions exhibit a similar pattern, with higher Methane conversion at S/C = 3 and 4. Though SMR stoichiometry reactions show that 1 mol of $H_2O_{(g)}$ is needed for 1 mol of $CH_4$, in practice, the reaction is carried out with a higher S/C ratio, in the range 2.5–3 to avoid carbon formation [55].

Hydrogen production via microwave steam plasma requires a stable plasma flame. We observed a steady plasma flame in the quartz discharge area for all experimental settings specified in this study. An evaluation of the result shows that the S/C ratio is a crucial factor influencing methane conversion, as methane conversion increases with increasing

**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 |
|---|---|---|---|---|
| Conventional steam reforming of methane (Catalyst) | $CH_4 + H_20 +$ air | | 60 Established industrial process | [55] |
| Metal Cylinder based MPS | $CH_4 + N_2$ | | 85 | [56] |
| Metal Cylinder based MPS | $CH_4 + CO_2 + H_2O$ | 180 | 42.9 | [33] |
| Metal Cylinder based MPS | $CH_4 + CO_2$ | 89 | 26 | [57] |
| Dielectric Barrier Discharge | $CH_4 +$ air | | 6.7 | [31] |
| Microwave (2.45 GHz) Plasma | $CH_4 + CO_2$ | 240 | 41.4 | [58] |
| Gliding arc | $CH_4 + H_20 +$ air | | 40 | [59] |
| Electron Beam Radiolysis | $CH_4 + H_2O$ | | 3.6 | [60] |
| Plasmatron with Catalyst | $CH_4 + H_20 +$ air | | 225 | [61] |
| Microwave(915 MHz) Plasma without catalyst | $CH_4 + H_2O$ | 2247 | 70 | This work |

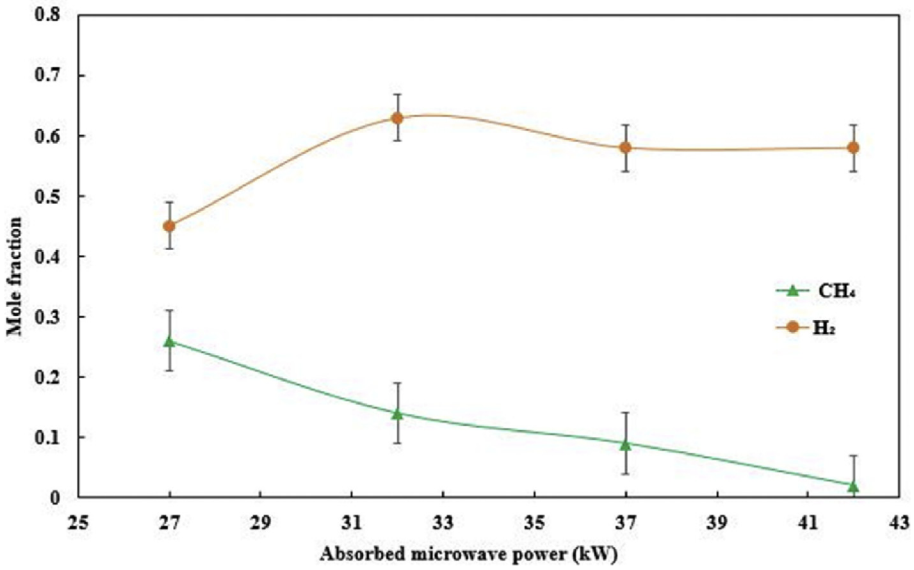

Fig. 10 – Mole fraction of $CH_4$ and $H_2$ as a function of absorbed microwave power.

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S/C ratio. However, beyond S/C = 4, methane conversion showed no significant conversion; this implies a reduction in the energetic parameters in the discharge zone. With a higher S/C ratio, there is a higher increase in electric field in the reaction zone, which results in the extension of the plasma flame and collision time. We did not observe the formation of soot throughout the experiment for the tested parameters. The best composition of the outlet gas by mole per cent obtained from our investigation is as follows: $CH_4$ - 3.4%, $H_2$ - 63.4%, CO -10.4%, $CO_2$ - 6.3%, $H_2O_{(g)}$ −16.5% at methane and steam flow rate of 62 lpm and 180 lpm respectively, and 32 kW microwave power. Methane conversion degree varied from 74.6% to 99.8%, depending on the absorbed microwave power and steam to methane ratio.

Methane conversion increased with absorbed microwave power as well as increasing flow rate before the system reached saturation. We varied the S/C ratio to maximize hydrogen yield in the reported experiments. Because the reactants can be altered to determine the reformer's outlet gas composition, the PSMR is versatile and can produce syngas of a variety of $H_2$/CO ratios suitable as a precursor for Fischer-Tropsch synthesis and other synthesis processes. Fig. 10 shows the mole fraction of $CH_4$ and $H_2$ as a function of absorbed microwave power.

## Conclusion

The renewed interest in hydrogen as an environmentally benign source of energy, as well as the need for distributed hydrogen production systems, motivated us to develop and test the PSMR to validate its capability for commercial-scale distributed hydrogen production. In this study, the results of the PSMR process for hydrogen production performed with a microwave steam plasma torch was presented. The experimental results for the energetic parameters of hydrogen production, precisely the hydrogen production rate and energy yield of 2247 [g($H_2$)/h] and 70 [g($H_2$)/kWh] respectively, were the optimum values realized.

The PSMR experimental results indicate that at an absorbed microwave power of 32 kW, methane conversion increased with increasing the S/C ratio. Methane conversion also increased with absorbed microwave power; as absorbed microwave power increased from 17 kW to 47 kW, methane conversion increased from 74.6% to 97.7% at a fixed S/C ratio. Similarly, as absorbed microwave power increased from 27 to 32 kW, the hydrogen production rate increased from 1532 to 2247 g($H_2$)/h.

Furthermore, energy yield as a function of absorbed microwave power decreased from 70 [g($H_2$)/kWh] to 46 [g($H_2$)/kWh] as absorbed power increased from 32 to 42 kW, respectively. The best $H_2$ - 63.4%, CO - 10.4%, $CO_2$ - 6.3%, $H_2O$(g) - 16.5%. Based on the results obtained, a high gas flow rate and a high hydrogen generation rate were possible without a catalyst and additional heating via the PSMR. Figs. 6–9 shows that the experimental results showed good alignment with the ASPEN Plus simulation values. In analyzing the output gas composition except for hydrogen, the other species like carbon dioxide, carbon monoxide, methane, and other gases are considered byproducts. Pressure Swing Adsorption (PSA) is utilized to recover pure hydrogen; however, we did not use it in the presented work. The PSMR may recycle unconverted methane. The need to slightly reduce the temperature gradient along the length of the reformer is vital to enhance the efficiency of the system. Since we could leverage the abundant reactive species in plasma, the hydrogen production method described here did not require a catalyst.

The experimental results advanced in this investigation signify that the PSMR is a ready technology for distributed hydrogen production. The method has many advantages, including high energy efficiency, low carbon emissions, and fast start-up time. Due to the various thermal regions in the system, methane reforming and WGSR co-occur in the reformer; hence, a separate WGSR unit may be redundant. Furthermore, operating the reformer without soot makes handling impurities simpler since a gas soot separator is unnecessary.

We anticipate that the technology would provide a cost-effective way to produce hydrogen in a decentralized manner. PSMR can be developed for small and large-scale applications owing to its high-power density capability, and it is suitable for onboard and compact applications. It is worth mentioning that this technology could be the first of its kind for commercial hydrogen production. In addition, the presented method surpasses the US Department of Energy's target of 60 [g($H_2$)/kWh] energy yield for distributed hydrogen production by 2020. Hydrogen production from methane or natural gas and other fuels can be done efficiently through PSMR technology.

## 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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