JOURNAL OF
EMERGING INVESTIGATORS

Article

Correlating inlet gas composition to conversion efficiency in plasma-assisted landfill gas reforming

Shihyung (William) Kim[1], Israel Torres Pineda[2]

[1] Concord Academy, Concord, Massachusetts

[2] En2Core Technology, Daejon, South Korea

SUMMARY

The escalating crisis of climate change, driven by the accumulation of greenhouse gases from human activities, demands urgent and innovative solutions to curb rising global temperatures. Plasma-based methane (CH_4) and carbon dioxide (CO_2) reforming offers a promising pathway for carbon capture and the sustainable production of hydrogen fuel and syngas components. To advance this technology, particularly in terms of energy efficiency and selectivity, it is essential to enhance the conversion efficiencies of CO_2 and CH_4. This study focused on atmospheric pressure inductively coupled plasma (ICP) in facilitating this process and focused specifically on inlet gas composition as a key parameter affecting conversion efficiency in the plasma system. We hypothesized that higher volumes of certain landfill gases (specifically CH_4, CO_2, and N_2) would increase CH_4 and CO_2 conversion efficiency due to increased reactant availability, while N_2 would modulate the reaction by acting as an inert buffer gas. A plasma gas reforming dataset, obtained during the operation of a demonstration-scale syngas/methanol production plant, indicated an average methane conversion efficiency of 95.1% and CO_2 conversion efficiency of 30.5%. By applying linear and quadratic regression models, we found that CO_2 flow significantly correlated to CO_2 conversion efficiency in a convex upward trend, characterized by a notable squared term coefficient of 9.757 ($p<0.01$). CH_4 and N_2 also were significantly correlated with the CH_4 conversion rate ($p<0.01$). These meaningful results highlight the substantial predictive strength of the models in determining conversion efficiencies based on gas variations and outline improvements that can be made to attain optimal plasma parameters.

INTRODUCTION

Methane (CH_4) and carbon dioxide (CO_2) are two of the most abundant gases in our atmosphere, and both gases can significantly impact our global climate. The release of these gases from anthropogenic sources has risen by almost six times in the last 70 years, and they have been at the epicenter of climate change debates given their high global warming potential (1). Merely reducing emissions is not a sufficient response, however, as at least 20% of CO_2 and CH_4 will remain in the atmosphere and propagate climate change for several millennia (2). Therefore, addressing the unchecked release and accumulation of these gases through carbon capture efforts will be key to mitigation efforts going into the 21st century.

In recent years, plasma, a state of matter where gas is ionized, has emerged as a promising avenue for carbon capture. Plasma-induced chemical reactions can be used to form syngas, a mixture of hydrogen and carbon monoxide, from hydrocarbons (3). During this process, electrons are excited into an ionized state to produce reactive species, or molecules possessing high reactivity due to unpaired electrons, from methane, carbon dioxide, and steam (3, 4). These reactive species include hydrogen (H_2) and carbon monoxide (CO). H_2 can act as a clean-burning, high-density fuel, serving as an alternative to fossil fuels or supplement for renewable energy systems (5, 6). This syngas can be further processed via a water-gas shift process—a chemical reaction where water vapor reacts with carbon monoxide—to produce more hydrogen (4, 7). Alternatively, the syngas can be synthesized into methanol (MeOH) or liquid hydrocarbon fuels (substances of high calorific value with wide industrial applications) (4). This repurposing of abundant gases with high global warming potential is pivotal not only for energy conservation and production but also for realizing a more circular economy where waste products are converted into valuable resources.

The above-discussed plasma gas reforming is an innovative technology with few drawbacks and excellent operational flexibility. The non-equilibrium state produced through thermal plasma, where the electron temperature is thousands of degrees higher than the gas temperature, allows H_2 to be selected reliably and efficiently (8). In this state, the electrons generated from the ionization of gas molecules in the plasma are extremely active and can easily form reactive species. Catalysts, such as nickel (Ni), may also be introduced to the system, lowering the activation energy of reactions and allowing for greater conversion efficiency and selectivity of the reforming reactions (9, 10). Plasma systems are also operationally flexible, being capable of fast initiation and deactivation; the high-voltage electric arcs necessary for plasma can be induced between two electrodes near-instantaneously, and the high-temperature reactive conditions diminish rapidly upon shutdown, needing almost no cooling period (6). Additionally, plasma systems are resilient to gas impurities, as the high-energy environment neutralizes and breaks down nearly all impure substances, making them well-suited for a variety of conditions and applications (4, 6). For example, in industrial settings where the purity of gases is not guaranteed or strictly controlled, plasma systems can bypass this entry barrier. Realizing these strengths, however, is contingent upon the development of efficient and scalable

https://doi.org/10.59720/23-252

reforming technologies.

In this study, we focused specifically on atmospheric-pressure inductively coupled plasma (ICP) as a gas reforming technology. Unlike low-pressure plasmas, atmospheric-pressure plasmas allow for operations under ambient conditions, thus making them more energy-efficient and cost-effective (11). These plasmas, energized electrically, produce reactive species capable of instigating chemical reactions at relatively lower temperatures than conventional methods. Additionally, ICP is characterized by its ability to produce plasma using an oscillating electromagnetic field. Its use of electromagnetic induction eliminates electrode-related energy losses, making it more energy-efficient than other plasma sources (9). We chose ICP for its potential in the reforming of CH_4 and CO_2, which offers an avenue for even more efficient syngas production.

There has been plenty of research done on the optimization of the plasma parameters of energy input, residence time, input gas flow, and plasma frequency for improved conversion efficiency, where the proportion of input reactants that were transformed into target outputs, such as hydrogen, was higher (8, 9, 12). Research by Martin-del-Campo et al., in particular, provided valuable insights into gas flow and conversion efficiency (12). Their work evaluated the effects of parameters such as the CO_2/CH_4 ratio on conversion efficiency and product selectivity. At higher ratios of CO_2/ CH_4, specifically over 2.0, the production of hydrogen decreased, while the efficiency was maximized around a ratio of 1.5. Martin-del-Campo and colleagues found that this optimal point manifested due to a balance between methane dehydrogenation and the oxidizing effect of CO_2, providing a precedent for adjustments in gas composition influencing conversion efficiency (12). However, these experiments were done under idealized conditions, where fluctuations in gas compositions were made with controlled adjustments. In practice, inlet gases would be drawn from abundant sources of CH_4 and CO_2 such as waste sites, sewage systems, and agrarian settings, causing the gas composition to fluctuate significantly. The effects of unpredictable inlet gas composition are still relatively ill-studied, as most reforming setups are experimental and use precisely regulated gas mixtures (13). This gap in the existing literature warrants further investigation, which our study seeks to address.

As a consequence of gas fluctuation, thermal management problems occur. ICP reactors operate at extremely high temperatures, which results in non-uniform temperature distributions (10). This non-uniformity stems from the complex interplay between electromagnetic fields, plasma dynamics, and the thermal properties of gases that create localized hot spots, thereby affecting the overall efficiency of the conversion process. These temperature fluctuations are why current research emphasizes the importance of better understanding the relationship between inlet composition and conversion efficiency (13, 14). Despite this need, the selectivity of produced gases as they relate to a fluctuating inlet gas composition remains ill-understood.

We sought to investigate the correlation between inlet gas composition and conversion efficiency in the use case of landfill gas reforming. The variable gas composition in landfills offers rich data for analysis that corresponds well to the practical waste-to-energy application of plasma gas reforming. Furthermore, the high greenhouse gas concentration in landfills poses an ideal use case for plasma gas reforming, turning waste disposal sites into sustainable energy reservoirs. Therefore, we drew on data from a demonstration-scale landfill-to-methanol production plant located in Daegu, South Korea. The demonstration plant used landfill gas from the Bangcheon-ri Municipal Solid Waste Landfill Site, operated by the company Daesung Eco-Energy. The gas contained varying quantities of CO_2, CH_4, N_2, and steam, which were fed into an ICP reactor (**Figure 1**). In this context, we focused on understanding the effect of a variable inlet gas composition on CO_2 and CH_4 conversion efficiencies through the lens of plasma dynamics and chemical properties.

Our hypothesis was that higher volumes of certain landfill gases (specifically CH_4, CO_2, and N_2) would increase CH_4 and CO_2 conversion efficiency due to increased reactant availability, while N_2 would modulate the reaction by acting as an inert buffer gas. Our findings validate the prediction that CO_2 flow rate would increase conversion efficiency, as CO_2 volume showed a positive correlation with CO_2 conversion efficiency, but only up to a certain point, after which efficiency declined. Adversely to our prediction, higher methane flow rates tended to reduce CH_4 conversion efficiency. Additionally, nitrogen flow rates negatively impact CH_4 conversion, likely due to thermodynamic disruptions in the plasma system. This work lays the foundation for a better understanding of the impact of inlet gas composition on ICP conversion efficiency, which could ultimately lead to significant advances in the syngas industry and, therefore, our approach to climate change mitigation.

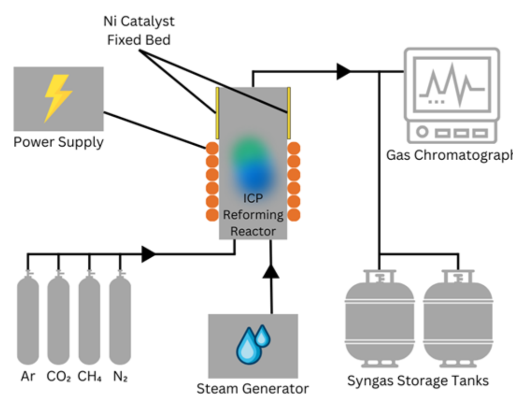

Figure 1. Setup schematic of ICP plasma simulated landfill gas reforming project. Gas cylinders are pictured in the bottom left; electricity is supplied to the reactor while steam is provided by a steam generator; fixed bed nickel catalysts assist in reaction selectivity; outlet gas flows to a gas chromatograph and is deposited in syngas storage tanks. Flow meters and oscilloscopes were used to track flow rates and energy input values. Plasma ignition was run for over 300 hours under simulated settings and gas diagnostics were taken at 20-minute intervals.

RESULTS

We investigated how varying flow rates of CH_4, CO_2, and N_2 into a reactor chamber affect conversion efficiency in plasma gas reforming. The ICP reactor used here was designed to simulate landfill gas quantities, so Ar, CH_4, CO_2, and N_2 gas cylinders were fed into the reactor simultaneously (**Figure 1**). Flow rates for CH_4 fluctuated between 25-30 LPM, while

https://doi.org/10.59720/23-252

those for CO_2 were around 15-20 LPM. We also employed fixed-bed nickel catalysts, a standard catalyst application method that allows plasma to be generated alongside the catalyst. To record flow rate values, we used normal gas chromatography (GC) in favor of flame ionization detector gas chromatography (FID-GC), which records trace gas quantities, to avoid including minute components that could introduce outliers or skew the results. By focusing on the more substantial gas components, the GC approach provided a clearer, more representative analysis. GC was performed in 20-minute intervals for the entire ignition, which lasted 300 hours.

We generated linear and quadratic regressions to examine relationships between gas flow rates and conversion efficiencies. A linear model was chosen because classical chemical kinetics dictate that a chemical reaction rate increases linearly with concentration up to a certain limit (3, 5). A quadratic regression model was implemented to reflect more complex curvilinear relationships, such as plasma-induced activation, where the energy from the plasma affects the reactants and intermediates in a non-linear manner. Non-linearity in plasma reactions can also be attributed to the saturation of active sites, plasma instabilities, or the formation of secondary products that either facilitate or inhibit the primary conversion process (10, 11).

First, we analyzed the relationship between CH_4 flow rate, the primary inlet component, and CH_4 conversion efficiencies to determine whether conversion efficiency increased with the quantity of the respective gas. Opposite to our hypothesis, the linear regression model indicated a significant negative correlation between CH_4 quantity and the conversion efficiency, with a coefficient of -0.384 ($t = -20.35$, $p < 0.01$), representing a significant decrease in conversion rates as the CH_4 flow rate increased (**Figure 2a**). This relationship can be attributed to the limited availability of active sites for methane's interaction with other gases, leading to a saturation point beyond which additional CH_4 does not contribute to conversion, but rather hinders it. The quadratic regression described a significant concave upward relationship characterized by a coefficient of -1.570 for the squared term ($t = -12.43$, $p < 0.01$) (**Figure 2a**). This analysis reveals a broad negative correlation between CH_4 flow rate and CH_4 efficiency.

We found similar results when we analyzed the impact of CH_4 flow rate on CO_2 conversion, which showed a slight concave downward relationship with a lower coefficient of 0.170 for the squared term ($t = -4.49$, $p < 0.01$) (**Figure 2b**). Our linear analysis followed the quadratic curve closely, showing a negative relationship between CH_4 flow rate and CO_2 conversion, with a coefficient of -0.335 ($t = -4.43$, $p < 0.01$) (**Figure 2b**). Our results suggest that increasing CH_4 flow rate is associated with a decrease in CO_2 conversion efficiency, likely because the saturation of active sites leads to a continuous decline in reaction efficiency.

Next, we evaluated the CO_2 flow rate in relation to CO2 conversion and found a significant positive linear relationship with a high coefficient value of 0.949 ($t = 8.99$, $p < 0.01$) (**Figure 3b**). These data are indicative of an increase in conversion rate with the escalation of CO_2 flow rate within the observed data range. When CO_2 is introduced into a plasma environment, it undergoes various energy-driven dissociation, ionization, and excitation reactions, facilitated by the plasma's energetic electrons (11). As more CO_2 is supplied, there are more

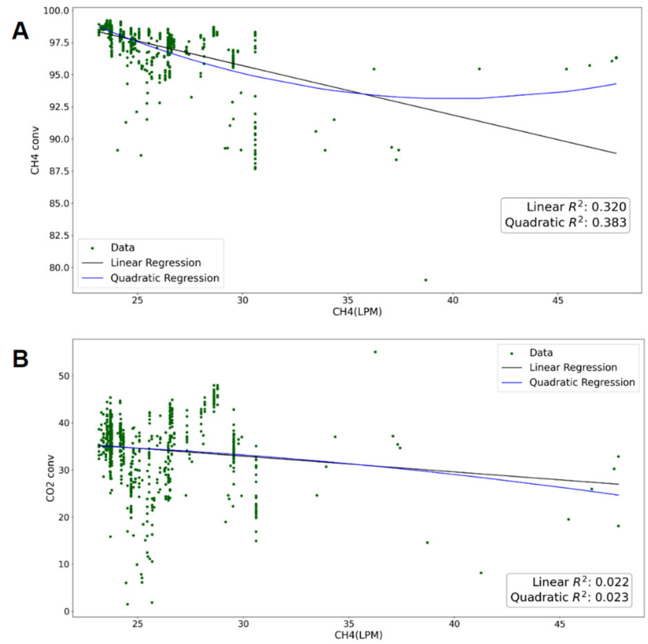

Figure 2. CH_4 inlet flow influences conversion rate of methane (CH_4) and carbon dioxide (CO_2). A) CH_4 conversion rate (%) and **B)** CO_2 conversion rate (%) as a function of CH_4 flow (LPM), fitted with linear and quadratic regression curves and corresponding R^2 values.

opportunities for energy transfer from the plasma to the CO_2 molecules, thus promoting the dissociation and subsequent reactions that contribute to increased production of syngas. Meanwhile, the quadratic regression revealed a significant convex upward trend, characterized by a notable squared term coefficient of 9.757 ($t = 15.11$, $p < 0.01$) (**Figure 3b**). This trend suggests a decline in CO_2 conversion efficiency beyond a certain threshold. In contrast, the effect on CH_4 conversion was negligible, showing only a 0.023 coefficient of the squared term ($t = -1.02$, $p < 0.01$) (**Figure 3a**). The linear analysis also showed a weak correlation, with only a -0.196 coefficient ($t = -6.08$, $p < 0.01$) (**Figure 3a**). This representation suggests that beyond a certain point, an increase in the CO_2 flow rate could potentially lead to rapid gains in CO_2 conversion efficiency but not CH_4 conversion efficiency.

Finally, we examined the CH_4 and CO_2 conversion as a function of N_2 flow rate, where we found a significant negative relationship between N_2 flow and CH_4 conversion, marked by a coefficient of -0.238 ($t = -9.64$, $p < 0.01$). This showcases a decrease in CH_4 conversion efficiency as the N_2 flow rate elevates. The quadratic model portrayed a significant concave downward trajectory, with a squared term coefficient of 1.411 ($t = 7.16$, $p < 0.01$) (**Figure 4a**). A similar quadratic trajectory was observed with CO_2 conversion with a concave downward trend and a squared term coefficient of 1.794 ($t = 1.19$, $p = 0.837$) (**Figure 4b**). The increase in CH_4 conversion efficiency at the center of the quadratic curve despite a downward linear trend may result from a balance between the diluting effects of nitrogen in the gas mixture and its role in stabilizing the plasma, thereby facilitating more efficient reforming reactions at higher concentrations (15). This model elucidates that, in line with our hypothesis, N_2 flow rate may facilitate higher CH_4 conversion efficiencies at moderate levels, but also that larger concentrations reduce efficiency due to dilution.

https://doi.org/10.59720/23-252

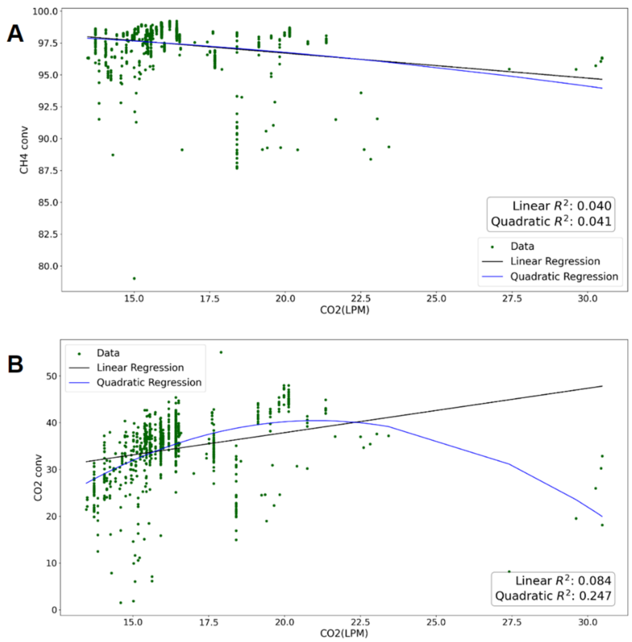

Figure 3. CO_2 inlet flow influences conversion rate of methane (CH_4) and carbon dioxide (CO_2). A) CH_4 conversion rate (%) and **B)** CO_2 conversion rate (%) as a function of CO_2 flow (LPM), fitted with linear and quadratic regression curves and corresponding R^2 values.

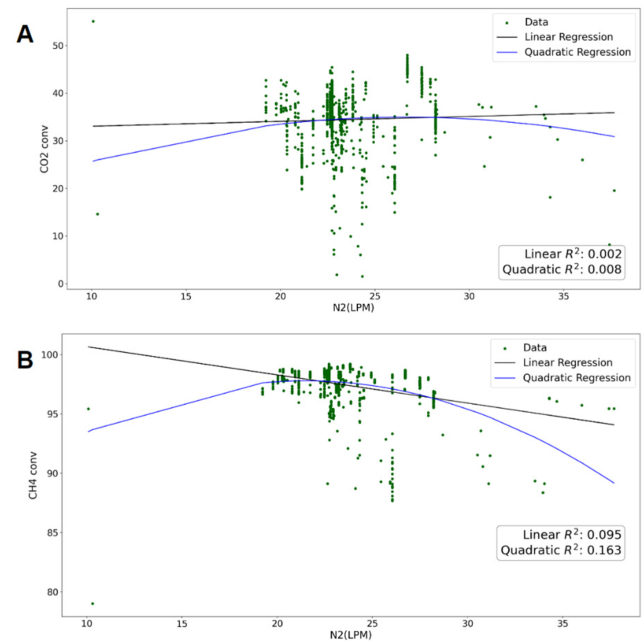

Figure 4. N_2 inlet flow influences conversion rate of methane (CH_4) and carbon dioxide (CO_2). A) CH_4 conversion rate (%) and **B)** CO_2 conversion rate (%) as a function of N_2 flow (LPM), fitted with linear and quadratic regression curves and corresponding R^2 values.

DISCUSSION

We investigated the effect of varying inlet gas compositions on the conversion efficiencies of methane (CH_4) and carbon dioxide (CO_2) in an inductively coupled plasma (ICP) system. We found that higher CH_4 flow rates decrease its conversion efficiency, while CO_2 conversion efficiency increases with CO_2 flow until it declines at higher levels. Nitrogen (N_2) negatively impacts CH_4 conversion, likely due to thermodynamic disruptions in the plasma.

An unexpected result in our study was the generally low R^2 values observed in the fitted curves of the models. Since our study is fairly unique in its focus on the impact of variable inlet gas compositions in plasma gas reforming, it is unclear how these findings compare directly to other studies. Similar trends have been observed in related research, such as Martin-del-Campo, who reported challenges in achieving high R^2 values when modeling the effects of varying CO_2/CH_4 ratios on conversion efficiency in a rotating gliding arc reactor (12). They attributed this to the influence of multiple interacting plasma parameters, such as gas temperature, residence time, and the non-linear dynamics of plasma reactions, which are difficult to isolate in experimental setups (12). This aligns with our observation that other influencing factors, such as temperature, pressure, and plasma frequency, likely modulate the effects of gas flow rates on conversion efficiency, contributing to the lower R^2 values. Therefore, while the very low p-values in the models suggest a significant correlation, the effect is likely modulated by other variables beyond just gas flow rate. Overall, there is a weak linear relationship between flow rate and conversion efficiency as opposed to curvilinear relationships, which better describe a multitude of plasma variables and parameters.

We observed a decline in conversion efficiency at certain concentration thresholds, particularly within the quadratic relationship between CO_2 flow rate and CO_2 conversion efficiency. This decline suggests that the interaction dynamics between molecules undergo substantial shifts when the concentration reaches an upper threshold. This could lead to an unfavorable environment for the conversion of CO_2 to desired products. Other studies attribute this shift to the fact that CO_2 is a triatomic molecule with complex vibrational modes, meaning that its dissociation in plasma can create hot spots with a high density of reactive oxygen species, which, through various chemical reactions, alter the local temperature distribution (11). Uneven temperature distribution can affect conversion efficiency by causing suboptimal interactions between reactants and active sites and by facilitating secondary reactions that consume the desired products or generate unwanted by-products such as longer-chain hydrocarbons or ozone. The dissociation of CO_2 can also lead to a higher degree of ionization, which might significantly alter the electromagnetic field within the plasma environment. This can contribute to electromagnetic perturbations, thus affecting the stability and uniformity of the plasma (8, 11). To further increase the efficiency at higher CO_2 levels, the catalyst characteristics could be further optimized to enhance the interaction with CO_2 molecules and facilitate more effective conversion pathways.

Nitrogen's negative correlation to CH_4 conversion efficiency could be due to its influence on the thermodynamic properties of the system, specifically heat transfer dynamics, that indirectly impact reaction rates. Nitrogen, being diatomic and relatively inert, can influence the formation of localized hot spots mainly through vibrational excitation modes (15). Current research corroborates the idea that hot spots facilitate reaction pathways that are energetically unfavorable for H_2 production (16). Furthermore, nitrogen can contribute to non-uniform temperatures predominantly through collisional

https://doi.org/10.59720/23-252

processes and vibrational excitations, which can disrupt the thermal equilibrium in the plasma.

One limitation of the current study is the scarcity of data at higher gas flow rates, which primarily results from operational constraints typical in plasma gas reforming processes. High flow rates are often avoided in practice due to the risk of exceeding the plasma system's capacity for effective gas ionization and dissociation, which can lead to inefficiencies in energy use and potentially uncontrolled reaction conditions. Such operational limits are informed by foundational studies in plasma technology, which highlight the importance of maintaining flow rates that allow for optimal residence time and energy transfer for the conversion process (6, 12). Consequently, the limited dataset at these higher flow rates might skew our understanding of the plasma reforming process under these more extreme conditions. This limitation in data coverage can affect the robustness of our models, particularly in predicting the process's behavior in industrial settings where higher throughput might be desired. Given unlimited resources and time, a more robust approach would involve developing advanced plasma reactors capable of safely operating at higher flow rates. Furthermore, another limitation of this study is the lack of control over simultaneously varying gas variables during the experiments. While this approach reflects real-world conditions, it introduces complexities in isolating the specific effects of individual gases on conversion efficiency. Future investigations could aim to explore higher flow rate conditions and design experiments where variables are systematically isolated, allowing for a more precise understanding of each gas's contribution to the observed trends.

Overall, to optimize conversion rates, a deeper analysis of the reaction chemistry is needed. Identifying substances that can act as promoters to enhance catalytic activity or facilitate alternate reaction pathways with higher efficiencies might be avenues worth exploring. For instance, our results indicate that the flow rates of CH_4 and CO_2 significantly influence their conversion efficiencies, with CH_4 showing diminishing returns at higher flow rates and CO_2 exhibiting a convex upward trend before reaching a saturation point. These findings suggest that future studies should investigate the precise mechanisms behind these thresholds, such as the role of active site saturation and energy transfer efficiency in plasma environments. Once again, the focal point should be the alteration of specific variables within the reaction system while controlling for the others. Detailed studies focusing on these aspects can provide insights into the optimum conditions for achieving higher conversion efficiencies.

Ultimately, our hypothesis that higher volumes of certain landfill gases (CH_4, CO_2, and N_2) would increase CH_4 and CO_2 conversion efficiencies was partially confirmed. Higher CO_2 flow rates were positively correlated with CO_2 conversion efficiency up to a certain threshold, beyond which efficiency declined, likely due to saturation effects or shifts in plasma dynamics. Conversely, increased CH_4 flow rates negatively impacted its conversion efficiency, suggesting that excess methane may saturate active sites and hinder effective plasma interactions. Nitrogen's role was found to be more complex, showing both dilutive and stabilization effects at different concentrations. Our research is important in the broader context of developing sustainable and scalable technologies for carbon capture and utilization. Our work contributes to the global effort to mitigate climate change by advancing the technical understanding of plasma gas reforming and supports the transition towards a more circular economy, where waste gases can be transformed into sustainable resources. In summary, a systematic approach grounded in the principles of chemical reaction engineering and thermodynamics can provide the necessary insights and directions for enhancing the conversion efficiencies in CO_2 reforming reactions. Future work should focus on a meticulous, controlled exploration of the various factors influencing the reaction dynamics, paving the way for more efficient and optimized processes.

MATERIALS AND METHODS

Data collection

The data used in this study was collected by EN2CORE Technology, a company specializing in the development of plasma technology, at their landfill plasma reactor plant in Daegu. Flow rates for CH_4, CO_2, and N_2 were measured in liters per minute (LPM) with gas chromatography (GC), reflecting the volume of gas passing per minute. The range of flow rates was based on the operational parameters of the demonstration plant, simulating conditions similar to those expected in industrial plasma gas reforming. In addition to normal GC, data pertaining to trace inlet gases were recorded using flame ionization detector gas chromatography (FID-GC). Both were performed in 20-minute intervals for the ignition, which lasted 300 consecutive hours, for a total of 900 data points for each gas. The GC conversion efficiency was selected for analysis rather than FID-GC to exclude trace gases from the analysis.

The data collection process captured detailed records of plasma power and temperature, system pressure, and the conversion rates of CO_2 and CH_4. Temperature sensors placed at 8 cm intervals along the reactor vessel ensured a comprehensive, accurate measurement of temperature. The dataset included the calculated CH_4 conversion efficiency (**Equation 1**), CO_2 conversion efficiency (**Equation 2**), and energy conversion efficiency (**Equation 3**) obtained respectively with the following formulas:

$$Conversion_{CH_4}\,(\%) = \frac{CH_4 input(mol) - CH_4 output(mol)}{CH_4 input(mol)} \times 100 \quad \text{(Eqn. 1)}$$

$$Conversion_{CO_2}\,(\%) = \frac{CO_2 input(mol) - CO_2 output(mol)}{CO_2 input(mol)} \times 100 \quad \text{(Eqn. 2)}$$

$$ECE\,(\%) = \frac{H_2\,(mol) \cdot LHV(\frac{kJ}{mol}) + CO(mol) \cdot LHV(\frac{kJ}{mol})}{W + n_{CH_4}\ of\ consumed\ (mol) \cdot LHV(\frac{kJ}{mol})} \times 100 \quad \text{(Eqn. 3)}$$

where ECE stands for the energy conversion efficiency and LHV for the lower heating value.

Throughout the experiment, CH_4, CO_2, and N_2 gas flow rates were varied simultaneously as they were fed into the reactor, and their conversion efficiencies were recorded. The plasma frequency and energy input were the only parameters explicitly controlled and kept constant during the experiment, while the quantities of CH_4, CO_2, and N_2 fluctuated based on the conditions of the demonstration plant. Because of this setup, all gas flow rates were altered together, and no single gas component was held constant while another was varied. This approach reflects the practical conditions of landfill gas reforming, where gas compositions naturally fluctuate rather than being precisely controlled. The data gathered provides insight into the correlations between gas flow rates and conversion efficiencies under realistic operating conditions rather than controlled laboratory settings.

https://doi.org/10.59720/23-252

Analysis

Python version 3.10 was employed for data handling and statistical analysis. A variety of Python libraries were employed, including Pandas for data analysis and manipulation, NumPy for numerical and mathematical operations, Matplotlib for creating detailed visualizations, and Scikit-Learn for implementing regression models. Furthermore, the Statsmodels library was used to estimate and test statistical models, which especially aided in extracting detailed statistics from the regression models implemented. The completed Python scrip used for regression analysis and visualization is provided in the **Appendix.**

The data was imported and preprocessed using the Pandas library for efficient data manipulation. Conversion values equaling 0% or 100% were pruned as errors, and null or 0 gas flow values were pruned as well. Both linear and quadratic regression analyses were applied for predictive models. These techniques were implemented using the Scikit-learn library. The inlet gas composition values served as the independent variables, and the conversion efficiencies of CO_2 and CH_4 were the dependent variables. Ordinary least squares (OLS) regression analysis was applied to fit both linear and quadratic models. The p-values were derived from t-tests on the regression coefficients, determining whether these coefficients significantly differed from zero and thus indicating significant relationships between gas flow rates and conversion efficiencies.

To convey the outcomes visually, the regression analyses were represented through Matplotlib in scatter plots complemented by regression lines delineating both linear and quadratic trends. These plots showcased the best-fit line or curve against the actual data points. This visualization process enabled a clear and concise representation of the results, offering deep insights into the trends and patterns within the data.

ACKNOWLEDGMENTS

We thank Dr. Yoon Seong Lee (EN2CORE Technology) for providing the plasma reforming dataset and for describing information on its circumstances and setup. We also thank Dr. Hong Young Chang (Korea Advanced Institute for Science and Technology) for his insights into academia and the research process.

Received: October 1, 2023
Accepted: April 6, 2024
Published: June 28, 2025

REFERENCES

1. Ritchie, Hannah, and Max Roser. “CO2 Emissions.” *Our World in Data*, 11 May 2020, www.ourworldindata.org/co2-emissions. Accessed 13 Aug. 2023.
2. Moore, Lisa. “Greenhouse Gases: How Long Will They Last?” *Environmental Defense Fund*, 26 Feb. 2008, blogs.edf.org/climate411/2008/02/26/ghg_lifetimes/. Accessed 20 Aug. 2023.
3. Xia, Yun, et al. “Combined steam and CO2 reforming of CH4 for syngas production in a gliding arc discharge.” *Journal of CO2 Utilization*, vol. 37, April 2020, pp. 248-259. https://doi.org/10.1016/j.jcou.2019.12.016.
4. Feng, Jiayu, et al. “Plasma-Assisted Reforming of Methane.” *Advanced Science*, vol. 9, no. 34, 17 October 2022. https://doi.org/10.1002/advs.202203221.
5. Momirlan, Magdalena, and Veziroglu, T.N. “The properties of hydrogen as fuel tomorrow in sustainable energy system for a cleaner planet.” *International Journal of Hydrogen Energy*, vol. 30, no. 7, 7 Dec. 2004, pp. 795-802. https://doi.org/10.1016/j.ijhydene.2004.10.011.
6. Budhraja, Neeraj. “Plasma reforming for hydrogen production: Pathways, reactors and storage.” *International Journal of Hydrogen Energy*, vol. 48, no. 7, 22 Jan. 2023, pp. 2467-2482. https://doi.org/10.1016/j.ijhydene.2022.10.143.
7. Chen, Wei-Hsin, and Chia-Yang Chen. “Water gas shift reaction for hydrogen production and carbon dioxide capture: A review.” *Applied Energy*, vol. 258, 15 Jan. 2020. https://doi.org/10.1016/j.apenergy.2019.114078.
8. Zheng, Hongtao, et al. “Kinetic Study of Nonequilibrium Plasma-Assisted Methane Steam Reforming.” *Mathematical Problems in Engineering*, vol. 2014, no. 938618, 28 Sept 2014. https://doi.org/10.1155/2014/938618.
9. Elaissi, Samira, et al. “Energy Efficiency Enhancement of Inductively Coupled Plasma Torch: Computational Study.” *Materials*, vol. 15, no. 15, 28 July 2022, pp. 5213. https://doi.org/10.3390/ma15155213.
10. Khoja, Asif Hussain, et al. “Hydrogen Production from Methane Cracking in Dielectric Barrier Discharge Catalytic Plasma Reactor Using a Nanocatalyst.” *Energies*, vol. 13, no. 22, 13 Nov. 2020, pp. 5921. https://doi.org/10.3390/en13225921.
11. Sun, Hojoong, et al. “Experiments and modeling of atmospheric pressure microwave plasma reforming of a methane-carbon dioxide mixture.” *Journal of CO2 Utilization*, vol. 46, no. 101464, Apr. 2021. https://doi.org/10.1016/j.jcou.2021.101464.
12. Martin-del-Campo, Jennifer, et al. “Influence of Operating Parameters on Plasma-Assisted Dry Reforming of Methane in a Rotating Gliding Arc Reactor.” *Plasma Chemistry and Plasma Processing*, July 2020. https://doi.org/10.1007/s11090-020-10074-2.
13. Song, Hohyung, et al. “Study on CH4 dry reforming process by high power inductively coupled plasma torch at atmospheric pressure.” *Current Applied Physics*, vol 20, no. 7, Jan. 2020, pp. 196-204. https://doi.org/10.1016/j.cap.2019.10.023.
14. Snoeckx, Ramses, et al. “Plasma-based dry reforming: improving the conversion and energy efficiency in a dielectric barrier discharge.” *RSC Advances*, vol. 38, 19 Mar. 2015. https://doi.org/10.1039/C5RA01100K.
15. Zhang, Hao, et al. “Rotating gliding arc assisted methane decomposition in nitrogen for hydrogen production.” *International Journal of Hydrogen Energy*, vol 39, no. 24, 13 Aug. 2014, pp. 12620-12635. https://doi.org/10.1016/j.ijhydene.2014.06.047.
16. Alphen, et al. “Effect of N2 on CO2-CH4 conversion in a gliding arc plasmatron: Can this major component in industrial emissions improve the energy efficiency?” *Journal of CO2 Utilization*, vol 54, Dec. 2021. https://doi.org/10.1016/j.jcou.2021.101767.

https://doi.org/10.59720/23-252

https://doi.org/10.59720/23-252

APPENDIX

```
import pandas as pd
import numpy as np
import matplotlib.pyplot as plt
import statsmodels.api as sm

from sklearn.linear_model import LinearRegression
from sklearn.preprocessing import PolynomialFeatures

# Load data
data = pd.read_excel('data.xlsx', usecols=["Time", "CH4(LPM)", "CO2(LPM)", "N2(LPM)", "CH4 conv", "CO2 conv", "Efficiency"])

# Prune data points based on the conversion rate
data = data[data['CH4 conv'] > 0]
data = data[data['CO2 conv'] > 0]
data = data[data['CH4 conv'] < 100]
data = data[data['CO2 conv'] < 100]

# Function to generate regression equation string
def generate_equation(reg, reg_type):
    if reg_type == "linear":
        return f'y = {reg.coef_[0]:.3f}x + {reg.intercept_:.3f}'
    elif reg_type == "quadratic":
        return f'y = {reg.coef_[1]:.3f}x^2 + {reg.coef_[0]:.3f}x + {reg.intercept_:.3f}'
    return ""

# Function for regression and plotting
def regression_and_plot(x_col, y_col, subplot_index, title):
    x_data = data[x_col].values.reshape(-1,1)
    y_data = data[y_col].values

    # Linear Regression
    reg = LinearRegression().fit(x_data, y_data)
    pred_linear = reg.predict(x_data)
    r2_linear = reg.score(x_data, y_data)  # Calculate R^2 for linear regression

    # Quadratic Regression
    poly = PolynomialFeatures(degree=2)
    x_poly = poly.fit_transform(x_data)
    reg_quad = LinearRegression().fit(x_poly, y_data)
    pred_quad = reg_quad.predict(x_poly)
    r2_quad = reg_quad.score(x_poly, y_data)  # Calculate R^2 for quadratic regression

    # Sort x_data and corresponding predictions for a smoother plot
    sorted_indices = np.argsort(x_data, axis=0).flatten()
    x_sorted = x_data[sorted_indices]
    pred_quad_sorted = pred_quad[sorted_indices]

    # Subplot
    plt.subplot(1, 1, subplot_index)  # Adjust for a 2x3 grid
    plt.scatter(x_data, y_data, s=20, color='darkgreen', label='Data')
    plt.plot(x_data, pred_linear, color='black', label='Linear Regression')
    plt.plot(x_sorted, pred_quad_sorted, color='blue', label='Quadratic Regression')
    plt.xlabel(x_col, fontsize=20)
    plt.ylabel(y_col, fontsize=20)
    plt.legend(fontsize=20)
    plt.xticks(fontsize=18)
    plt.yticks(fontsize=18)
    plt.title(title)
```

https://doi.org/10.59720/23-252

```
    x_text = max(x_data) * 1.01  # Adjust the 0.8 as needed to move text left or right
    y_text = min(y_data) * 1.05  # Adjust the 1.1 as needed to move text up or down

    plt.text(x_text, y_text, f'Linear $R^2$: {r2_linear:.3f}\nQuadratic $R^2$: {r2_quad:.3f}', fontsize=24,
             verticalalignment='bottom', horizontalalignment='right',
             bbox=dict(boxstyle='round', facecolor='white', alpha=0.5))

    # Print regression equations to the command window
    print(f"Linear regression equation for {title}: {generate_equation(reg, 'linear')}")
    print(f"Quadratic regression equation for {title}: {generate_equation(reg_quad, 'quadratic')}")

    # Extract stats for linear regression
    coef_linear, t_values_linear, p_values_linear = fit_regression_and_extract_stats(x_data, y_data, 'linear')
    print(f"Linear regression for {title}:")
    print(f"Coefficients: {coef_linear}")
    print(f"t-values: {t_values_linear}")
    print(f"p-values: {p_values_linear}")

    # Extract stats for quadratic regression
    coef_quad, t_values_quad, p_values_quad = fit_regression_and_extract_stats(x_data, y_data, 'quadratic')
    print(f"\nQuadratic regression for {title}:")
    print(f"Coefficients: {coef_quad}")
    print(f"t-values: {t_values_quad}")
    print(f"p-values: {p_values_quad}")


# Function to fit regression and extract stats
def fit_regression_and_extract_stats(x_data, y_data, reg_type):
    if reg_type == "linear":
        x_data = sm.add_constant(x_data)
        model = sm.OLS(y_data, x_data).fit()
    elif reg_type == "quadratic":
        x_data = sm.add_constant(x_data)
        poly = PolynomialFeatures(degree=2)
        x_data_poly = poly.fit_transform(x_data)
        model = sm.OLS(y_data, x_data_poly).fit()
    else:
        raise ValueError("Unsupported regression type")

    coef = model.params
    t_values = model.tvalues
    p_values = model.pvalues

    return coef, t_values, p_values

# Visualization
plt.figure(figsize=(15, 18))

# Plots
regression_and_plot("CH4(LPM)", "CH4 conv", 1, 'Regression: CH4(LPM) vs CH4 Conversion')
regression_and_plot("CH4(LPM)", "CO2 conv", 1, 'Regression: CH4(LPM) vs CO2 Conversion')
regression_and_plot("CO2(LPM)", "CH4 conv", 1, 'Regression: CO2(LPM) vs CH4 Conversion')
regression_and_plot("CO2(LPM)", "CO2 conv", 1, 'Regression: CO2(LPM) vs CO2 Conversion')
regression_and_plot("N2(LPM)", "CH4 conv", 1, 'Regression: N2(LPM) vs CH4 Conversion')
regression_and_plot("N2(LPM)", "CO2 conv", 1, 'Regression: N2(LPM) vs CO2 Conversion')

plt.tight_layout()
plt.show()
```