Aqueous Methane Reforming by Nonthermal Plasma

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Abstract: This work investigated the exciting possibility of using water in the liquid phase to reform methane in a nonthermal plasma system to yield hydrogen and syngas. This process, which we term aqueous reforming, produces at best results 49.9% hydrogen, 8.2% carbon monoxide, 3.7% carbon dioxide, and other useful hydrocarbons. This route shows great potential as an alternative to the energy-intensive steam methane reforming process.

1. Introduction

The reforming of methane to produce hydrogen and syngas traditionally requires high temperatures and cost-intensive processes like steam methane reforming (SMR) [1]. Nonthermal plasma proposes an auspicious alternative approach by enabling reactions under mild temperature and pressure conditions that consequently reduce process costs. The coupling of liquid water with methane gas in nonthermal plasma has been relatively unexplored. The closest study to this work was by Wang et al. [2] but involved the use of excessive power up to 900 W.

In this work, methane aqueous reforming was investigated using a different plasma regime under moderate power levels (200-400 W), with the aim of achieving industry-comparable results.

2. Methods

A nonthermal plasma reactor, as was used by Aka et al. [3], was used to discharge plasma in this two-phase system. Three different levels of applied power, gas flow rate, and liquid flow rate were used in these evaluation experiments. 200 W, 350 W, and 500 W for applied power, 50 mL/min, 300 mL/min, and 1000 mL/min for gas flow rate, and finally 25 mL/min, 100 mL/min, and 200 mL/min for liquid flow rate. For the modulations of each factor, the other two factors were held constant at their midpoints. 150 mL of distilled water was cycled through the reactor together with methane gas at the specified flow rates for each run using a peristaltic pump and a mass flow controller respectively. The gas-liquid reaction was then initiated by applying power through a variac-controlled transformer to the desired level until steady-state was reached and gas samples collected for analysis on a gas chromatograph.

3. Results and Discussion

Results from these preliminary studies revealed gas flow rate to be the most influencing factor for improved process performance. Figure 1 captures the trends in process performance parameters peculiar to traditional SMR. Lower gas flow rates were seen to promote higher conversion of methane and increased hydrogen yields. This is mostly attributed to the effect of residence time and sufficient reaction time [4]. The increased contact time between plasma-activated gas species and the liquid phase appears to enhance the conversion efficiency. At 100 mL/min gas flow rate, 350 W of applied power, and 100 mL/min liquid flow rate, the highest hydrogen yield of 49.9% was achieved, accompanied by significant CO formation (8.2%) constituting a H_2/CO ratio of 5.3.

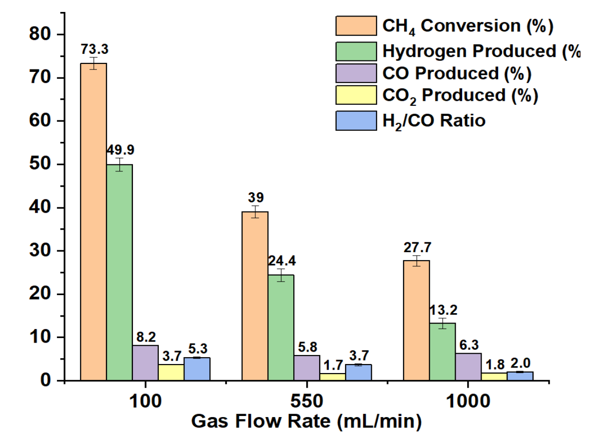

Fig. 1. Process metrics for aqueous methane reforming by plasma at varied gas flow rates

The applied power showed a direct positive correlation with product yields and methane conversion up to 400 W. The liquid flow rate also displayed an inverse relationship, with optimal performance achieved at lower flow rates.

4. Conclusion

The feasibility of aqueous methane reforming using nonthermal plasma has been demonstrated with decent performance. The process achieved promising hydrogen yields of up to 49.9% and significant syngas production with a H_2/CO ratio of 5.3 without the high temperature requirements of conventional SMR as well as without the use of expensive catalysts. These findings suggest that aqueous reforming by nonthermal plasma routes could offer an energy-efficient alternative to traditional methane reforming and hydrogen production processes.

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