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Research Article

Experimental Investigation of Biogas Reforming in Gliding Arc Plasma Reactors

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Biogas is an important renewable energy source. Its utilization is restricted to vicinity of farm areas, unless pipeline networks or compression facilities are established. Alternatively, biogas may be upgraded into synthetic gas via reforming reaction. In this work, plasma assisted reforming of biogas was investigated. A laboratory gliding arc plasma setup was developed. Effects of CH_4/CO_2 ratio (1, 2.33, 9), feed flow rate (16.67–83.33 cm^3/s), power input (100–600 W), number of reactor, and air addition (0–60% v/v) on process performances in terms of yield, selectivity, conversion, and energy consumption were investigated. High power inputs and long reaction time from low flow rates, or use of two cascade reactors were found to promote dry reforming of biogas. High H_2 and CO yields can be obtained at low energy consumption. Presence of air enabled partial oxidation reforming that produced higher CH_4 conversion, compared to purely dry CO_2 reforming process.

1. Introduction

Concerns over energy demand and environmental deterioration are becoming increasingly serious due to accelerated depletion of fossil fuel reserve and exhaust gases released from burning fossil fuels. There is an urgent need to develop and secure alternative energy sources. At present, our attention is focused on biogas. Biogas is produced from anaerobic digestion of organic materials. Normal composition of biogas is about 55–70% methane, 27–44% carbon dioxide, 1% or less hydrogen, and 3% or less hydrogen sulfide, depending on raw input materials [1, 2]. Biogas is normally utilized for heating, mechanical power, and electricity generation. It is noticeable that biogas contains a significant fraction of an inert gas (CO_2), which reduces its energy content. Furthermore, utilization of biogas is rather restricted to within and around farm areas, unless pipeline networks or compression facilities are deployed. For wider utilization and application of biogas, upgrading should be undertaken. Therefore, biogas may be converted to synthetic gas, a mixture of hydrogen and carbon monoxide. Synthetic gas can be used as fuel in combustion process and fuel cells, as well as for Fischer-Tropsch synthesis of clean liquid fuels [3, 4].

There are several technologies available for synthetic gas production such as partial oxidation, steam reforming, autothermal reforming, and catalytic reforming [5–8]. However, there are limitations of such technologies due to energy requirement, yields of product, and economic cost. One possible way to overcome these limitations is by using plasma technique. Plasma is high energy phase, which can cause fast reactions [9]. Plasma can be divided into thermal and nonthermal, according to temperature and density of electrons [10, 11]. Nonthermal plasma has been used for production of synthetic gas. Advantage of nonthermal plasma relates to low temperatures (<1000 K) and low pressures (atmospheric pressure) that result in less energy consumption and minimum electrode erosion. Size and weight of nonthermal plasma reactors are relatively small and attractive for mobile applications. Gliding arc discharge plasma technology is of great interest. Gliding arc reactor consists of two or more divergent electrodes connected to AC or DC power supply. When a high voltage is applied, a relatively low current arc discharge is generated across the electrodes. It can be used to encourage reforming reaction of biogas [12]. The biogas dry reforming process can be described by an endothermic reaction (external energy is required), as shown in (1) and (2) [13, 14]. CO_2 in biogas is used to react with CH_4. Synthetic gas may be reformed in partial oxidation reaction of biogas. Equation (3) shows reaction of methane, partial oxidative, and exothermic reaction. Equations (4)–(9) show interaction of CH_4 and CO_2 with electrons in plasma cracking process after electrical breakdown:

$$CH_4 + CO_2 \longrightarrow 2CO + 2H_2 \quad \Delta H = 247\,kJ/mol, \tag{1}$$

$$CH_4 + 2CO_2 \longrightarrow 3CO + H_2 + H_2O \quad \Delta H = 288\,kJ/mol. \tag{2}$$

Partial oxidation reforming reaction is as follows:

$$CH_4 + \frac{1}{2}O_2 \longrightarrow CO + 2H_2 \quad \Delta H = -36\,kJ/mol. \tag{3}$$

Interaction of CH_4 and CO_2 with electron in plasma cracking process is as follows: [15]

$$CH_4 + e^- \longrightarrow CH_3 + H \tag{4}$$

$$2CH_4 \longrightarrow C_2H_2 + 3H_2 \tag{5}$$

$$CH_3 + H \longrightarrow CH_2 + H + H \tag{6}$$

$$CH_2 + H + H \longrightarrow CH_2 + H_2 \tag{7}$$

$$CH_2 + H_2 \longrightarrow CH + H_2 + H \tag{8}$$

$$CH + H_2 + H \longrightarrow C + H_2 + H_2 \tag{9}$$

$$CO_2 + e^- \longrightarrow CO + O. \tag{10}$$

It can be seen that major gas products are H_2 and CO. Other products of gas such as hydrocarbon (C_2H_2) are also produced, but in very small amount [16, 17]. So, in this work, only main components of synthesis gas (H_2 and CO) are considered.

There have been several reports on synthetic gas production by gliding arc reactors. Steam and catalyst are normally used single gliding arc plasma reactor in reforming process. However, there appeared to be few studies using multistage reactors. Rueangjitt et al. [15] used a multistage gliding arc plasma system with four reactors connected in series to investigate combined reforming of natural gas to produce synthesis gas. Sreethawong et al. [18] used a multistage gliding arc discharge system to explore partial oxidation of methane to produce synthesis gas. Tippayawong and Inthasan [19] utilized two gliding arc plasma reactors for cracking of light tar and subsequently modified to assist in oxidative reforming of biogas [20]. Nonthermal plasma generated from gliding arc discharge has shown to have high potential in reforming reaction. However, investigation on biogas reforming in multistage plasma reactors still remains very rare. This work presents an attempt to fill this gap. The objective of this work is to parametrically investigate influential operating conditions on plasma assisted reforming of biogas.

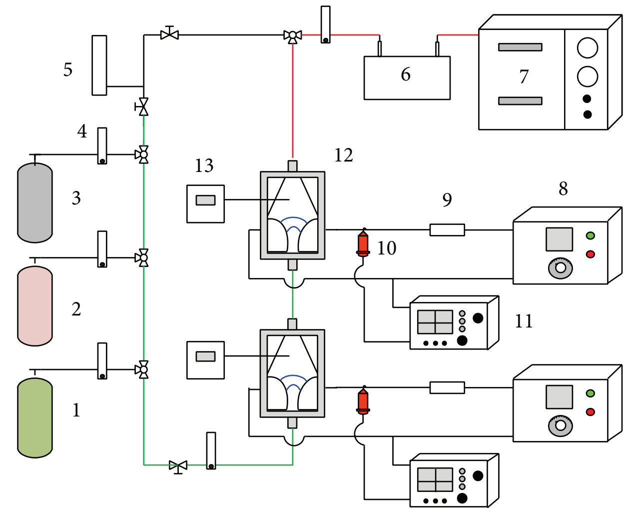

Figure 1: Experimental setup for biogas reforming: (1) CH_4 tank, (2) CO_2 tank, (3) air zero tank, (4) flow meter, (5) bubble flow meter, (6) gas filter, (7) gas chromatography, (8) power supply, (9) resistance, (10) high voltage probe, (11) oscilloscope, (12) gliding arc reactor, and (13) thermometer.

In this paper, effects of biogas flow rate, composition of biogas, power input, and air addition on production of synthetic gas using gliding arc plasma reactor were studied. A comparison between one and two reactors was evaluated. It may offer alternative route for utilization of biogas by producing high content of synthetic gas.

2. Materials and Methods

Typical composition of biogas contains 45–65% methane, 35–55% carbon dioxide, 1% hydrogen, and trace amount of hydrogen sulfide around 1000 ppm. For the current experiments, simulated biogas was generated by mixing CH_4 and CO_2 at different fractions (CH_4/CO_2 = 1, 2.33, 9), corresponding to biogas compositions found typical, and those after upgrading by removal of CO_2.

Gliding arc plasma experiments were carried out using a laboratory scale setup at atmospheric pressure. Schematic of the experimental setup specially developed is shown in Figure 1. A gliding arc plasma reactor has two knife-shaped, divergent electrodes made of stainless steel. An electrode gap distance was fixed at 4 mm. The gap distance was selected at 4 mm because it was maximum distance possible for electric jump between electrodes. Gas input was injected between electrodes via a cylindrical tube with diameter of 1 mm. The reactor was designed to be flat shaped, not cylinder shaped similar to Bo et al. [6, 21]. Flat shaped reactors have advantage over cylinder in reaction space. The setup was composed of two gliding arc plasma reactors, two power supplies, biogas feeding system, measurement probes, and control and analysis systems. Biogas feed was regulated by a flow meter (VFB-60-BV) between 16.67 and 83.33 cm^3/s. The two electrodes were connected to high voltage supplies from two AC neon transformers (LEIP EX 230A 15N)

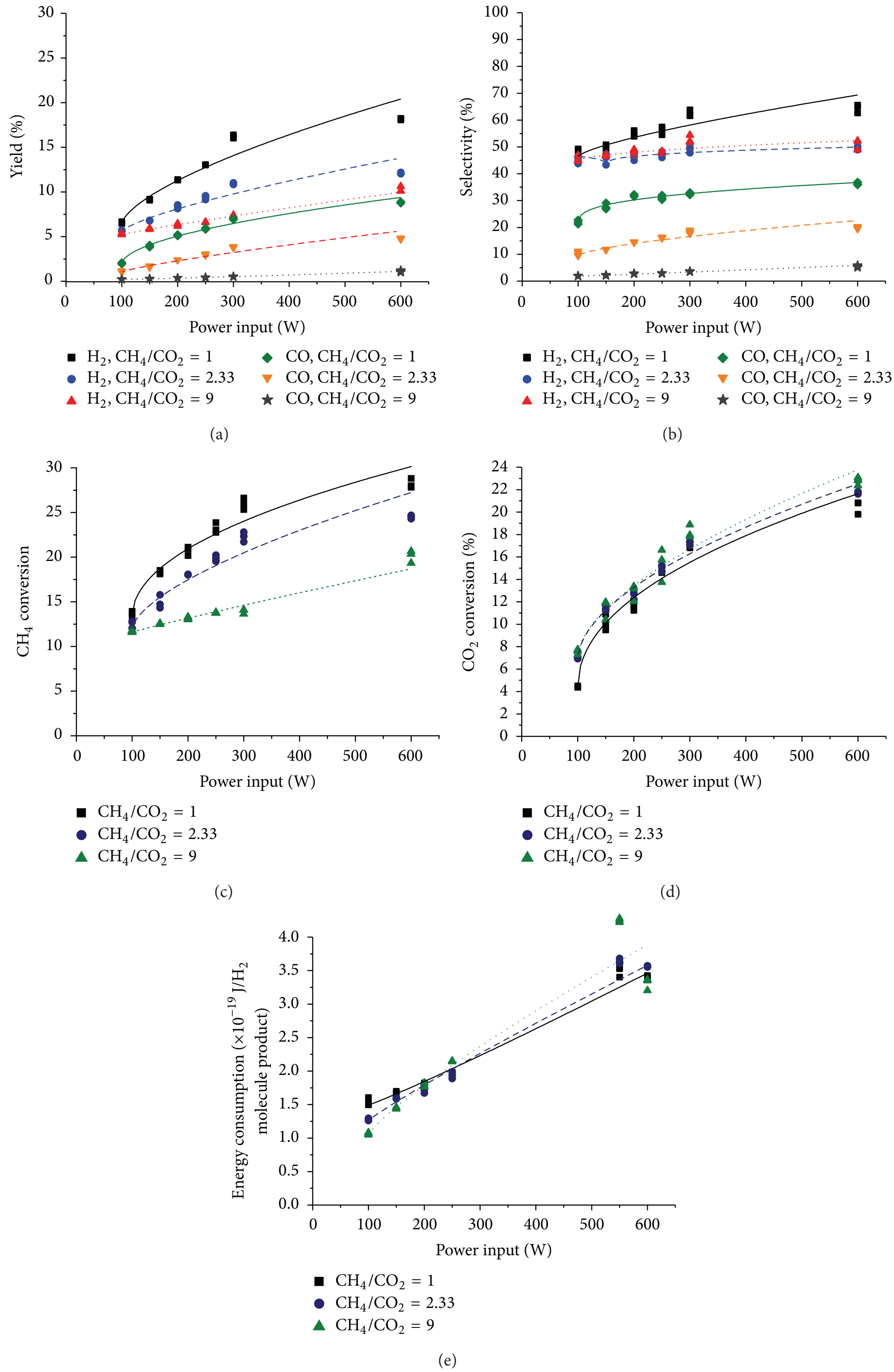

Figure 2: Effect of power input on biogas reforming performance.

with maximum voltage and current of 15 kV and 30 mA (nonbreakdown), respectively. In this work, thermocouples type K were deployed to indirectly measure plasma reaction temperature. The temperature probes were positioned downstream as close as possible to the plasma flame so that they did not interfere with the reforming reaction. Gas analysis was undertaken by a Shimadzu GC-8A gas chromatography with TCD detector.

Data analysis for performance evaluation of the reactors was as follows: [16, 21–23].

Conversion of CH_4 and CO_2:

$$CH_{4\,\text{conversion}}\,(\%) = \frac{\left[C_{CH_4,\text{in}}\right] - \left[C_{CH_4,\text{out}}\right]}{\left[C_{CH_4,\text{in}}\right]} \times 100, \quad (11)$$

where $[C_{CH_4,\text{in}}]$ is amount of CH_4 input (mol) and $[C_{CH_4,\text{out}}]$ is amount of CH_4 output (mol)

$$CO_{2\,\text{conversion}}\,(\%) = \frac{\left[C_{CO_2,\text{in}}\right] - \left[C_{CO_2,\text{out}}\right]}{\left[C_{CO_2,\text{in}}\right]} \times 100, \quad (12)$$

where $[C_{CO_2,\text{in}}]$ is amount of CO_2 output (mol) and $[C_{CO_2,\text{out}}]$ is amount of CO_2 output (mol).

Selectivity of H_2 and CO:

$$H_{2\,\text{selectivity}}\,(\%) = \frac{H_{2\,\text{produced}}}{2 \times CH_{4\,\text{converted}}} \times 100, \quad (13)$$

$$CO_{\text{selectivity}}\,(\%) = \frac{CO_{\text{produced}}}{CH_{4\,\text{converted}} + CO_{2\,\text{converted}}} \times 100. \quad (14)$$

The 2 in H_2 selectivity is the number of moles of hydrogen in methane that can produce H_2 is 2.

Yield of H_2 and CO is as follows:

$$H_{2\,\text{yield}}\,(\%) = \frac{H_{2\,\text{produced}}}{2 \times CH_{4\,\text{feed}}} \times 100, \quad (15)$$

$$CO_{\text{yield}}\,(\%) = \frac{CO_{\text{produced}}}{2 \times \left(CH_{4\,\text{feed}} + CO_{2\,\text{feed}}\right)} \times 100. \quad (16)$$

Energy consumption per H_2 molecule product (EC) is as follows:

$$\text{EC (J/Molecule)} = \frac{P \times 60}{6.02 \times 10^{23} \times \left(H_{2\,\text{produced}}\right)}, \quad (17)$$

where P is power input (W).

3. Results and Discussion

3.1. Effect of Power Input and CH_4/CO_2 Ratio. For the effect on reforming reaction of biogas, power input was varied between 100 and 600 W, while the biogas flow rate was fixed at 16.67 cm^3/s. Composition of biogas was varied for CH_4/CO_2 between 1, 2.33, and 9. Effects of power input and CH_4/CO_2 ratio on the process performance are shown in Figure 2. Reactor temperature was about 120–250°C. For a given CH_4/CO_2 ratio, yields of H_2 and CO (Figure 2(a)) were found to increase with power input. Changes with power input were more pronounced at CH_4/CO_2 ratio of 1 than that at CH_4/CO_2 ratio of 9. For a given power input, increasing CH_4/CO_2 ratio from 1 to 9 appeared to downgrade H_2 and CO generation. CO_2 in biogas reacted with CH_4. For a dry reforming process, appropriate molar ratios between CH_4 and CO_2 would be one or two ((1) and (2)). Equation (1) was more likely to occur, compared to (2) because less energy was required. It was therefore not surprising to find that maximum yields of H_2 and CO were obtained at CH_4/CO_2 = 1. At this value, yields of H_2 and CO were observed to increase from 6.7 to 18.0% and 2.0 to 8.8%, respectively, when power input was increased from 100 to 600 W. Selectivity of H_2 and CO is presented in Figure 2(b). Similar to yield, selectivity of H_2 and CO was maximum at CH_4/CO_2 = 1. It was evident that increasing power input caused selectivity of H_2 and CO to increase from 48.7 to 65.5% and from 21.9 to 36.8%, respectively. For larger CH_4/CO_2 ratios, H_2 selectivity appeared to stay at around 45%, while CO selectivity was lower than 5% for CH_4/CO_2 = 9. As far as conversion of CH_4 and CO_2 was concerned, similar patterns with increasing input power (Figures 2(c) and 2(d)) were observed for CH_4 and CO_2 conversion. Nonetheless, it was noted that change in CH_4/CO_2 ratio affected CH_4 more than CO_2. Figure 2(e) shows that the energy consumption per H_2 molecule product increased with increasing power input. Variation in CH_4/CO_2 ratio was not found to affect energy consumption. These results were consistent with those reported in [22]. Change in power input affected all reactions in the plasma zone. Increasing power input increased electron density and electron energy which affected formation of active free radicals in inelastic collisions between molecules of CH_4 and CO_2, resulting in higher degree of combination to form synthesis gas molecules [20]. Higher power is expected to give higher conversions and yields. However, in this work, increase in power input resulted in increased coke formation in the reactor. At higher power input, coke formation became clearly visible, presenting a serious operation problem. It may be attributed to the fact that higher input power encouraged more cracking of molecules (8) that led to more carbon generation. Additionally, increased power input produces higher temperature of the electrodes' surface at which biogas may be reacted to form carbon deposit on. Coke formation may be controlled by adding oxidants such as steam, oxygen, or air to the reforming reactants.

3.2. Effect of Flow Rate. Figure 3 shows variation of the process performance parameters with feed gas flow rate. For these tests, power input was fixed at 100 W to avoid coke formation. The flow rate was varied between 16.67 and 83.33 cm^3/s. Reactor temperature was about 120–250°C. As expected, the highest yields and selectivities occurred at CH_4/CO_2 = 1. It was found that increasing flow rate resulted in reduction of H_2 and CO yields (Figure 3(a)) from 6.3 to 1.3% and from 2.0 to 0.5%, respectively. Selectivities of H_2 and CO are presented in Figure 3(b). Increase in flow rate caused a decrease in H_2 and CO selectivity from 46.5

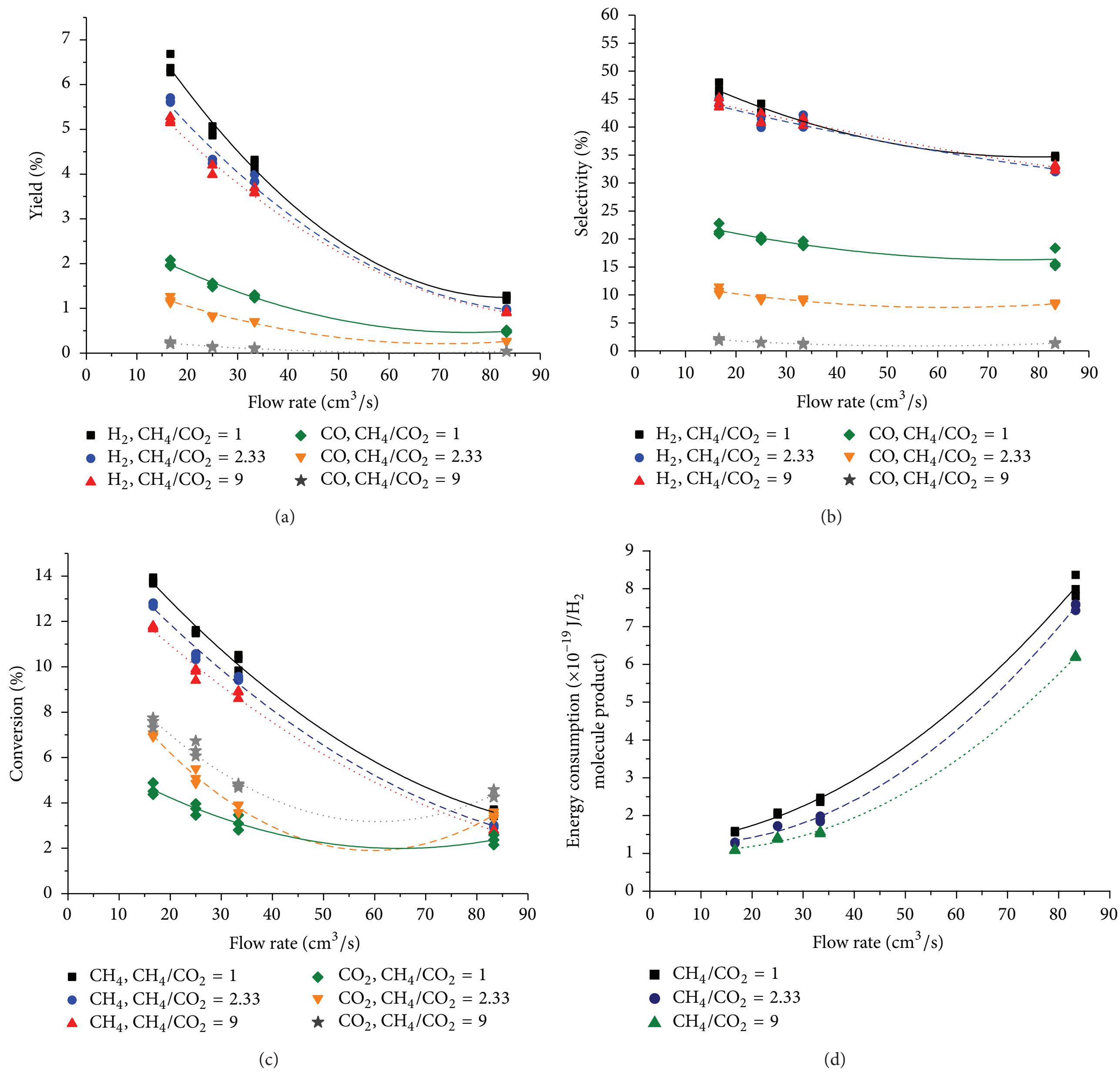

Figure 3: Effect of flow rate on biogas reforming performance.

to 36.5% and from 22.7 to 15.2%. This was largely due to reduction in residence time for the dry reforming reaction at higher flow rates. Figure 3(c) shows that increasing flow rate led to decreased CH_4 and CO_2 conversions for all CH_4/CO_2considered. Energy consumption per H_2molecule product was minimal at the smallest flow rate of 16.67 cm^3/s and the highest CH_4/CO_2 ratio of 9, shown in Figure 3(d). As the flow rate increases, yield and selectivity of H_2 and CO decreased, and energy consumption per H_2 molecule product was increased. This was anticipated because at high flow rates, it may not have enough time for CH_4 and CO_2 to react. The trend was in agreement with the literature [16, 23]. It should be noted that for the design of the present gliding arc reactor, the inner diameter of the injection nozzle was rather small (1 mm), compared to the electrode gap distance (4 mm). Larger nozzle diameter which will allow slower feed gas speed may produce better yields at high flow rates.

3.3. Effect of Air Addition. Air injection into the biogas would provide oxygen to enable partial oxidative reaction. This will be completed with CO_2 for dry reforming of CH_4 to generate H_2 and CO. For the effect of air addition, power input at 100 W and gas flow rate of 16.67 cm^3/s were chosen. Amount of

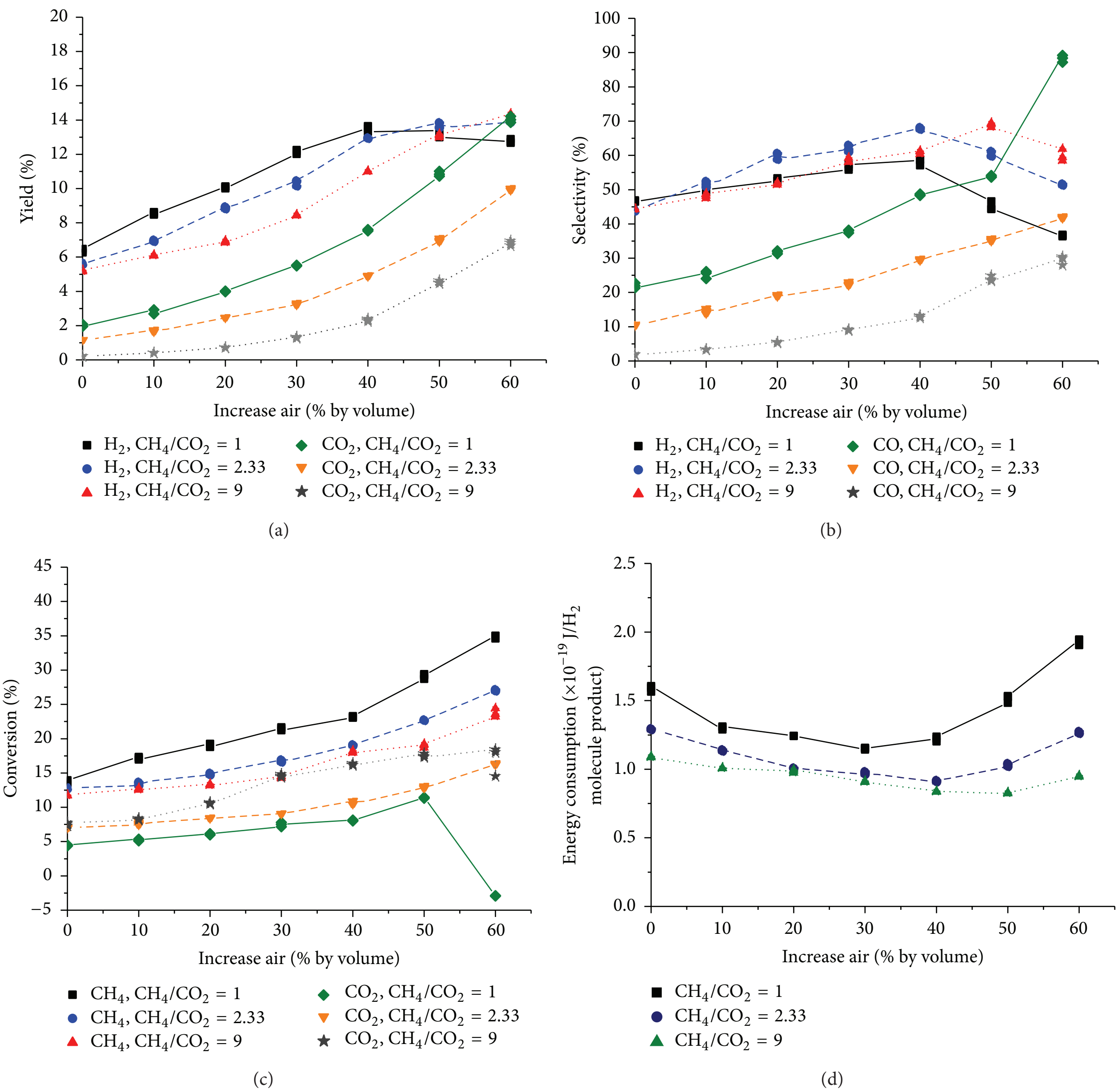

FIGURE 4: Effect of air addition on biogas reforming performance.

air in the gas feed was varied between 0 and 60% v/v. Reactor temperature was about 120–380°C, higher than previous cases of purely endothermic reaction. It was shown in Figure 4 that the presence of oxygen in air promoted reforming of biogas. Figures 4(a) and 4(b) show that H_2 yield and selectivity were increased with increasing air, as long as there was plenty of CH_4, and short of competing CO_2. These were the case for CH_4/CO_2ratios of 2.33 and 9. At CH_4/CO_2 = 1, drops in H_2 yield and selectivity after air was supplied beyond 40% were evident. These decreases of H_2 yield and selectivity may have contributed to the fact that the reaction was approaching substoichiometric combustion. CO generation became more favorable than H_2. It was clear that at higher air supply rates (>40%), changes in yield and selectivity of CO were opposite to those of H_2. This was confirmed by a negative conversion of CO_2, shown in Figure 4(c) at air supply of 60%, showing that combustion took place. Figure 4(d) shows that less energy was consumed at high CH_4/CO_2ratios. This was contributed to the fact that greater amount of air was available for an exothermic partial oxidation reaction, in line with the published literature [16, 24, 25].

3.4. Effect of Number of Reactor. In this work, the setup with two cascading reactors was also tested against the single gliding arc plasma reactor. The reacting flow may be

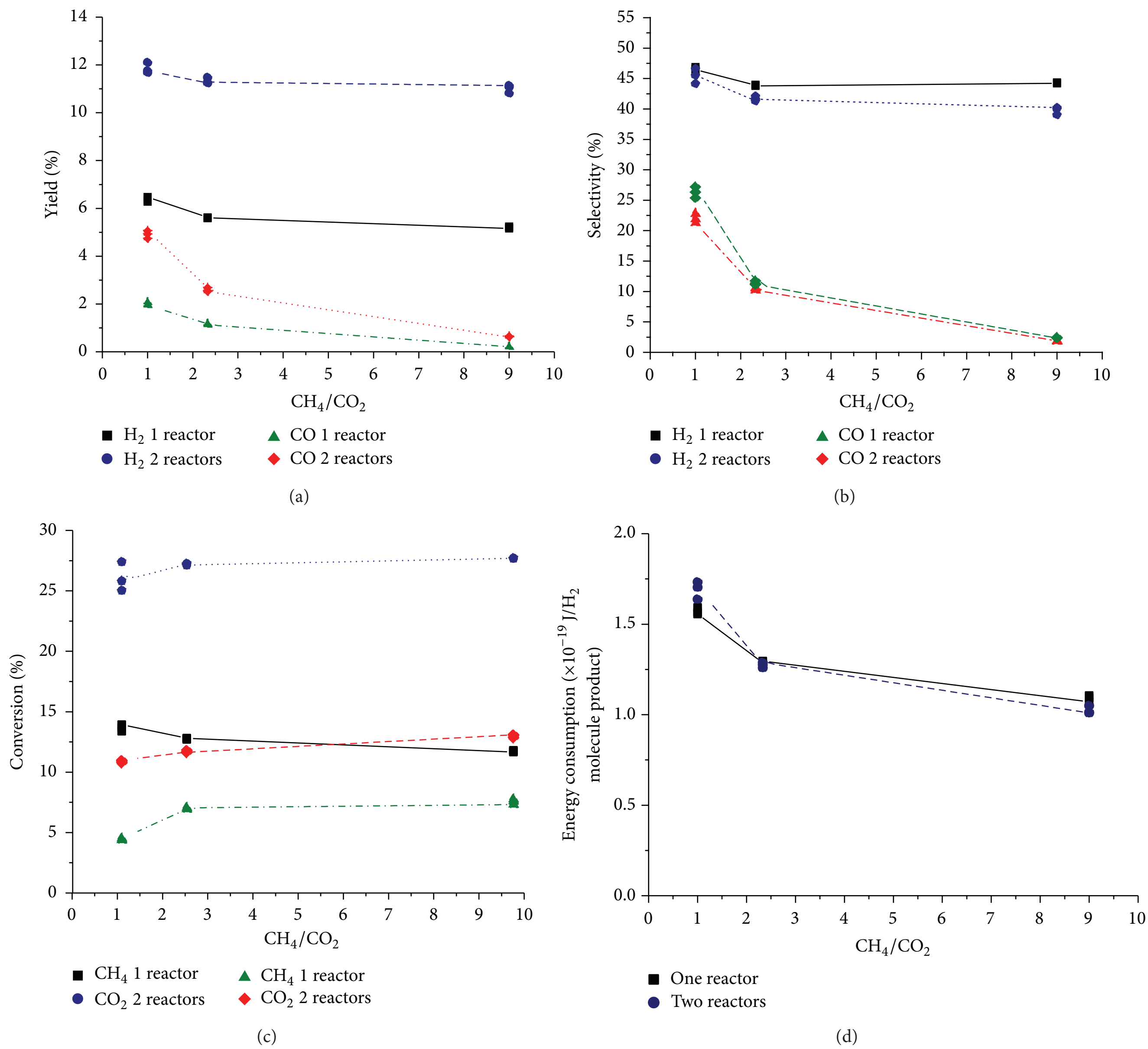

Figure 5: Effect of number of reactor on biogas reforming performance.

viewed to have longer residence time for reforming reaction. Flow rate and power input were fixed at 16.67 cm^3/s and 100 W, respectively. Reactor temperature was in similar range of about 120–250°C. Results are shown in Figure 5. Qualitatively, the two cascading reactors produced similar trends of yield, selectivity, and conversion with respect to the effect of increasing CH_4/CO_2 ratio. But, quantitatively, the two cascading reactors were found to have higher yields and conversions than the single reactor for all CH_4/CO_2 ratios considered, while selectivity in the single and two reactors was in similar magnitude. At $CH_4/CO_2 = 1$, H_2 and CO yields in the two reactors were about twice as those found in the single reactor, shown in Figures 5(a) and 5(c). At present, it was not conclusive why energy consumption found in the single and the two reactors (Figure 5(d)) was not significantly different. Further investigation may be needed to clarify this issue.

Table 1 summarizes the optimum conditions found in this work. Composition of biogas with $CH_4/CO_2 = 1$ proved to be the best in generating the highest yields of H_2 and CO. At this ratio, it was very attractive since it represented actual biogas composition in farms without upgrading. With the two gliding arc plasma reactors in cascade, higher residence time for reforming reaction may be achieved. This resulted in better performance in terms of yields and conversion. The two cascaded reactors setup appeared to consume slightly higher energy input. As far as air addition was concerned, it was clear that partial oxidation process offered alternative route in

Table 1: Performance of plasma assisted reforming of biogas.

	Flow rate (cm^3/s)	Air (% v/v)	Yield (%)		Conversion (%)		Energy consumption ($\times 10^{-19}$ J/H_2 molecule product)
			H_2	CO	CH_4	CO_2	
Single reactor							
$CH_4/CO_2 = 1$	16.67	—	6.3	1.9	13.6	4.87	1.56
$CH_4/CO_2 = 2.33$	16.67	—	5.6	1.1	12.6	6.9	1.28
$CH_4/CO_2 = 9$	16.67	—	5.1	0.2	11.7	7.5	1.07
Single reactor							
$CH_4/CO_2 = 1$	16.67	40	14.7	7.5	23.1	8.1	1.22
$CH_4/CO_2 = 2.33$	16.67	50	13.5	6.9	22.6	12.8	1.01
$CH_4/CO_2 = 9$	16.67	60	14.0	6.8	23.6	14.5	0.94
Two reactors							
$CH_4/CO_2 = 1$	16.67	—	11.7	4.9	25.8	10.7	1.70
$CH_4/CO_2 = 2.33$	16.67	—	11.2	2.5	27.2	11.6	1.25
$CH_4/CO_2 = 9$	16.67	—	11.0	0.6	27.6	13.0	1.01

reforming of biogas. It was shown to have the highest yields of H_2 and CO obtained, as well as the lowest energy input for all conditions considered in this work. In comparison with reported work by Sreethawong et al. [18] for similar conditions (multistage gliding arc plasma reactor, CH_4/O_2= 3, feed flow rate of 2.5 cm^3/s), this work proved to have similar CH_4 conversion, but with much less energy consumption per H_2 molecule product.

4. Conclusion

In this study, gliding arc plasma was utilized to reform biogas into synthesis gas. Investigations were carried out for the effects of biogas composition (CH_4/CO_2), power input, biogas flow rate, presence of air, and number of reactors on yield and selectivity of H_2 and CO, conversion of CH_4 and CO_2, and energy consumption. CO_2 and CH_4 in biogas were reacted under a dry reforming process. As anticipated, $CH_4/CO_2 = 1$ showed maximum reforming performance. High power input and lower flow rate were observed to enhance reforming reaction. Adding air into biogas was found to encourage partial oxidation that would compete with CO_2 in reforming CH_4 and generating H_2 and CO. A setup with the two cascade reactors was shown to have higher yields and conversions than the single reactor. Energy consumed was reported to be lower than that from previously published work.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgments

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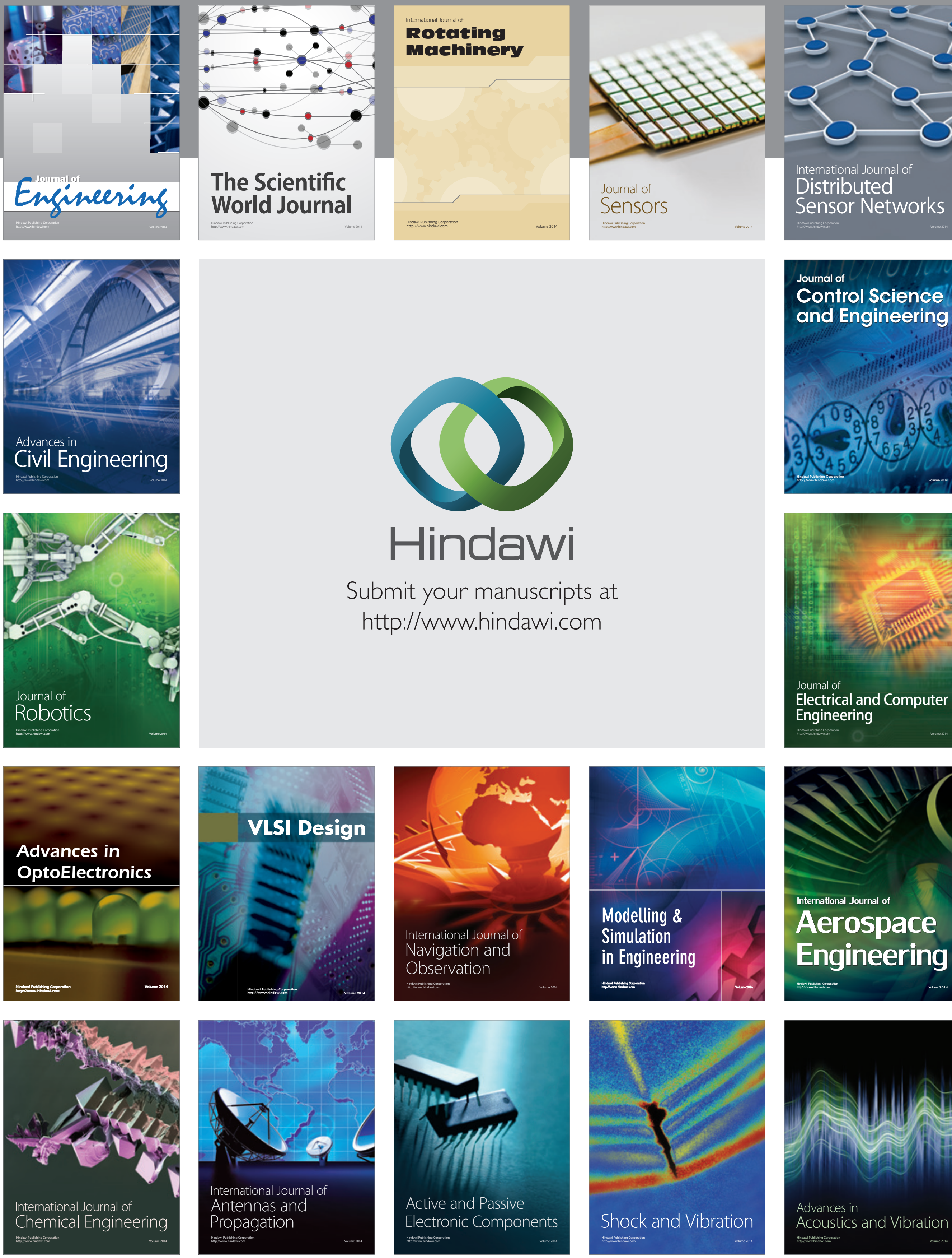
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