Hydrogen production by methane reforming based on micro-gap discharge

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Abstract. Based on micro-gap strong ionization discharge, this paper presents a study of hydrogen production by methane reforming at room temperature and atmospheric pressure without catalyst. Influence rules of conversion of methane and production of hydrogen were studied by changing discharge power and feed gas flow rate. Results show that when the discharge power was about 341 W, the discharge gap was 0.47 mm and the flow rate of feed gas was 100 mL min⁻¹, the conversion of methane and yield of hydrogen reached optimization. The conversion rate of methane and the highest yield of hydrogen were 68.14 % and 51.34 %, respectively.

1. Introduction
Methane and natural gas reforming are widely applied in industry to obtain hydrogen. Hydrogen is definitely a clean energy source, and fuel-cell technology has been investigated extensively and vigorously to practically utilize fuel cell vehicles and cogeneration system. Conventional technologies of hydrogen production, i.e. coal gasification, hydrocarbon reforming and water electrolysis, are too expensive or not applicable for specific applications, due to technical reasons. Thus, new methods are under development, like steam reforming [1], biological and plasma methods. Recently, developed micro-gap discharge operated at atmospheric pressure [2-7] seems to have a high potential for hydrogen production via hydrocarbon reforming.

In this paper, a method for production of hydrogen via methane reforming by the use of an atmospheric pressure micro-gap discharge reactor is presented.

2. Experiment setup and method
The plasma reaction chamber of hydrogen produced by methane was shown in figure 1. The plasma reactor is rectangular, and its dimensions are 260 mm (long) × 130 mm (wide) × 35 mm (thick). The thinner dielectric layer of α-Al₂O₃ is on the both sides of discharge electrodes, the dimensions of the dielectric layer are 175 mm (long) × 80 mm (wide) × 0.64 mm (thick), and the discharge gap is 0.47 mm. The dielectric constant and the electric insulation intensity are 10 and 350 kV cm⁻¹, respectively. The high voltage electrode is made from silver, grounding electrodes on both sides of which are made from aluminium. With this thinner dielectric layer, the high energy electrons can be obtained

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further to form the strong electric-field discharge. A YDKS-WK power supply was used in the experiment, whose frequency was 9.85 kHz.

In this study, the energy efficiency of the methane conversion and the production of compounds were evaluated using the specific input energy (SIE) which could be determined as:

\[
SIE = \frac{P_0}{v}
\]  

(1)

Where \(P_0\) was discharge power and \(v\) was flow rate of feed gas. The discharge power in micro-gap was measured and calculated using the method of charge-voltage graphics [7]. The generation rate \(r\) could be expressed as:

\[
r = \frac{\text{yield} \times v}{22.4(\text{L} \cdot \text{mol}^{-1})}
\]  

(2)

The electric field intensity \(E_g\) could be calculated by

\[
E_g = \frac{V_{pp} \varepsilon_d}{2l_d \varepsilon_g + l_g \varepsilon_d}
\]  

(3)

Where \(V_{pp}\) was the voltage peak value, \(\varepsilon_d\) was the dielectric constant. \(l_d\) is the gap of discharge, \(l_d\) is the dielectric thickness. The electron power is in direct proportion to the square of \(E_g\). The electron energy is restricted by the electric field intensity of critical breakdown, the gas density or the gas pressure.

![Figure 1. Structural diagram of plasma reaction chamber.](image)

### 3. Experiment result and discussions

**Table 1. Influence of discharge power for hydrogen generation and methane conversion.**

| Discharge power (W) | H\(_2\) yield (v/v, %) | Methane conversion (v/v, %) | H\(_2\) Generation rate (\(\mu\)mol min\(^{-1}\)) | \(E_g\) (kV mm\(^{-1}\)) | SIE (kJ L\(^{-1}\)) |
|---------------------|------------------------|-----------------------------|---------------------------------|-------------------------|---------------------|
| 143.8               | 20.44                  | 30.0                        | 1825                            | 11.37                   | 43.13               |
| 153.4               | 25.13                  | 34.2                        | 2244                            | 15.49                   | 46.01               |
| 226.0               | 29.20                  | 40.0                        | 2607                            | 17.56                   | 67.81               |
| 297.8               | 30.65                  | 41.9                        | 2737                            | 19.73                   | 88.28               |
| 352.7               | 32.16                  | 45.1                        | 2871                            | 20.70                   | 105.81              |

The experiments were carried out under the conditions of the room temperature, the 105 Pa pressure, without any catalyst. The mixed gases of CH\(_4)/O\(_2\)=95/5 (v/v) were introduced into the reactor at the
flow rate of 200 mL min\(^{-1}\). Table 1 shows the effects of discharge power for hydrogen generation and methane conversion. The SIE was 43.13 kJ L\(^{-1}\) under the 143.8 W discharge power. According to equation (1) and (3) the SIE and electric field intensity increased with the improvement of discharge power, the electron density of discharge gap discharge and electron collision frequency increased accordingly. Therefore an increase in discharge power can help to improve the chances of methane and electron collisions, the methane conversion rates and the hydrogen yield.

![Graph](image1)

**Figure 2.** Influence of discharge power for H\(_2\) yield and conversion of CH\(_4\).

![Graph](image2)

**Figure 3.** Influence of flow rate of feed gas power for H\(_2\) yield and conversion of CH\(_4\).

As shown in figure 2 and figure 3, both discharge power and flow rate of feed gas had great effects on the experimental results. The yield of H\(_2\) was 51.34 % (v/v) and the conversion of CH\(_4\) was 68.14 % (v/v) when the flow rate of feed gas was 100 mL min\(^{-1}\). Here excitation voltage and discharge power were 2.8 kV and 341 W respectively. In figure 3, when the excitation voltage was unchanged, the change of electric field intensity and discharge power were not obvious with the improvement of flow rate of feed gas, while the SIE decreased obviously. Hence, the yield of H\(_2\) and the conversion of CH\(_4\) decreased.

![Graph](image3)

**Figure 4.** Voltage-charge Lissajous graphics.

![Graph](image4)

**Figure 5.** Voltage & Current oscillograms.
Figure 4 and figure 5 show the voltage-charge Lissajous graphics and the Voltage & Current waveform which were used for measurement and calculation of discharge power. It can be inferred that YDKS-WK power supply provides a high degree of electric field intensity and power, since a large number of CH₄ molecules were ionized and dissociated to produce H₂.

4. Conclusions
This paper presents a method of methane conversion to hydrogen. Influence rules of conversion of methane and production of hydrogen were studied by changing discharge power and feed gas flow rate. As shown in results, with the improvement of discharge power, electric field intensity increases, hence, the conversion rate of methane and hydrogen yield increase. As the feed gas flow rate increases, specific input energy diminished, and the conversion rate of methane and hydrogen concentration reduce. Under optimal conditions conversion rate of methane and hydrogen yield reached 68.14 % and 51.34 % respectively.

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