

Hydrogen production from partial oxidation of dimethyl ether using corona discharge plasma

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Abstract

Dimethyl ether (DME), with its non-toxic character, high H/C ratio and high-energy volumetric density, is an ideal resource for hydrogen production. This work presents a hydrogen production from partial oxidation of DME using corona discharge plasma performed at ambient conditions. The partial oxidation can avoid the undesired carbon deposit existing in the decomposition of DME and a stable operation has been obtained. The carbon deposit is completely avoided when the O₂/DME ratio is 1.05. Partial oxidation can also greatly improve the conversion rate and energy efficiency. The optimal O₂/DME ratio is 1.38, with which the rate of H₂ production is 1.33 times higher and the energy efficiency is 1.72 times higher than that of DME decomposition. The higher O₂/DME ratio results in complete oxidation and reduces the rate of H₂ production. AC plasma is more efficient than the DC plasma and the optimum frequency is 2 kHz. The order of desired waveform is: sinusoid > sinusoid triangular > square > ramp.

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1. Introduction

Dimethyl ether (DME) has been recently recognized as an alternative diesel fuel and potential fuel for power generation and domestic uses. DME is relatively inert, non-corrosive, and non-carcinogenic. The physical properties of DME are similar to those of liquefied petroleum gas (LPG). It can be easily liquefied, stored and transported using the well-established facilities supplying LPG. In addition, DME can be synthesized directly from syngas, which is more thermodynamically favored, compared to methanol synthesis from syngas. This makes DME production more economically favored. DME will play an important role in the future fuel supply. In fact, DME has been investigated as a potential fuel for automotive proton exchange membrane fuel cell system [1].

DME is also an alternative hydrogen carrier due to its high H/C ratio and high energy density. Hydrogen can be produced

from hydrocarbons, alcohols and biomass via catalytic conversion at elevated temperatures [1–5]. In recent years, hydrogen production from DME has attracted increasing interests [6–12]. Galvita et al. reported steam reforming of DME to hydrogen for the first time [8]. Since then studies on the conversion of DME to produce hydrogen have quickly increased. Partial oxidation and steam reforming are usually used to convert DME into hydrogen. Partial oxidation is conducted at 400–600 °C over supported Ni catalysts [6,7]. Steam reforming of DME is carried out at 200–400 °C. Multi-component catalysts like Cu, Zn, Pd and/or Pt supported on acidic oxide are often used for this reaction [8–12].

Most of the established or developing processes for hydrogen production are conducted at elevated temperature with the presence of catalysts. These processes need a start-up stage and no hydrogen is produced before the temperature reaches the defined value. In addition, the catalysts inevitably suffer from deactivation. A simple, flexible and quick-responding hydrogen production technology is more favorable for remote and domestic applications where temporary but instant hydrogen

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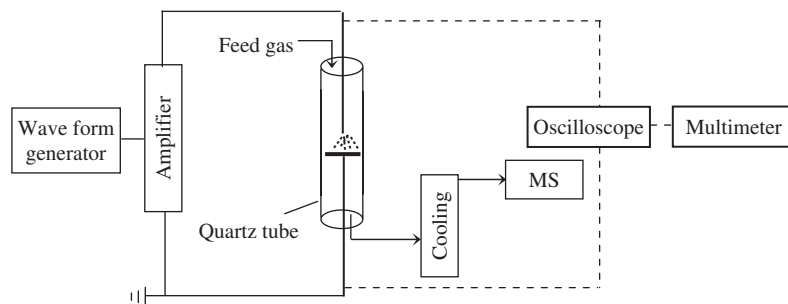


Fig. 1. Schematically representative of experimental apparatus for partial oxidation of dimethyl ether.

supplying is needed. In these regards, cold plasma could be a good choice. Exactly, cold plasmas have attracted much attention in materials, environmental and many other applications. Cold plasma is characterized by highly energetic electrons and low gas temperature. A major advantage of cold plasma is its rapid response. Plasma reactions can be carried out quickly without induction period that exists in conventional catalytic processes. The cold plasmas have been successfully utilized for the production of ozone and have been extensively investigated to convert methane into higher hydrocarbons and syngas [13–16].

The cold plasmas are also promising as a hydrogen generation technology. Water, methane and methanol have been studied as the feedstock [17–23]. Hydrogen has been produced at room temperature but the energy efficiency is very low. We have utilized corona discharge plasma (a type of cold plasmas) to decompose methanol into hydrogen with greatly improved energy efficiency [24]. This process is easily manipulated with quick response, demonstrating great potential for practical applications. Recently, we have further studied a DME decomposition using the corona discharge plasma and achieved a better result for hydrogen production [25]. However, some carbon filaments were produced as the by-products that posed a negative effect on the operation. Steam reforming or partial oxidation may be prior alternatives to solve this problem. With this regard, we have studied the plasma steam reforming of DME [26]. In this work, we aim to investigate the partial oxidation of DME to hydrogen at ambient conditions using the corona discharge plasma, in order to obtain a stable operation and to further improve the energy efficiency.

2. Experimental

Fig. 1 shows the schematic representative of the corona discharge apparatus used in this work. A quartz tube (i.d. 6 mm) was used as the tubular reactor. The corona discharge plasma was initiated between two electrodes placed in the quartz tube. The top electrode was a stainless steel pin (with a diameter of 2 mm) and the bottom electrode was a circular stainless steel disk with a diameter of 5 mm and a thickness of ca. 1.5 mm. There were four holes (i.d. 1 mm) on the disk to facilitate the flow of gas. The gap between the two electrodes was 6 mm.

The top electrode was connected with a high voltage amplifier (Trek, 20/20B) and the bottom electrode was grounded.

The discharge was generated by a DC and AC voltage supplied by a power supply unit consisting of a signal generator (HP 33120A) and the high voltage amplifier. Four types of waveforms with frequency ranging from 0 to 15 kHz were used. The input waveform was 2.0 kHz sinusoid if not specifically addressed in the discussions below. The discharge voltage and current were measured with a high voltage probe (Tektronix P6015) and a pulse current transformer (Pearson Electronics 411) and recorded with a digital oscilloscope (Tektronix 2440). The consumed power was measured with a digital multimeter (Keithley 2000). The temperature of electrode inside the reactor was measured using an infrared camera (Iacon, 100PHT) [27]. The temperature was calibrated by adjusting its emissivity until the recorded temperature equaled to the known temperature of the electrode (150 °C) that was placed inside the quartz tube and heated by an external heater. The temperature was measured when it reached a stable value (after the reaction being conducted for 10 min).

We have previously shown that the dilute gas Ar can improve the energy efficiency of plasma DME decomposition [25]. Therefore Ar was used as the dilute gas in this work. The flow rate of DME and Ar was 1.02 mmol/min (22.9 cm³/min) and 2.25 mmol/min (50.6 cm³/min), respectively. The flow rate of O₂ was controlled using a mass flow controller. The effluent gas was cooled by mixture of ice and water and then analyzed with an on-line MS (AVI-GmbH, Ominstar). The effluent flow rate was measured with a bubble meter. The gas product included H₂, CO and CO₂. Carbon deposits were measured with gravimetric analysis. The analysis showed good carbon balance in the range of 97–102%.

The conversions were defined as

$$\text{DME: } x_{\text{DME}}\% = (\text{rate of DME consumed} / \text{rate of DME introduced}) \times 100\%$$

$$\text{O}_2: x_{\text{O}_2}\% = (\text{rate of O}_2 \text{ consumed} / \text{rate of O}_2 \text{ introduced}) \times 100\%$$

The selectivities were defined as

$$\text{H}_2: S_{\text{H}_2}\% = [\text{rate of H}_2 \text{ produced} \times 2 / (\text{rate of DME consumed} \times 6)] \times 100\%$$

$$\text{CO: } S_{\text{CO}}\% = [\text{rate of CO produced} / (\text{rate of DME consumed} \times 2)] \times 100\%$$

$$\text{CO}_2: S_{\text{CO}_2}\% = [\text{rate of CO}_2 \text{ produced} / (\text{rate of DME consumed} \times 2)] \times 100\%$$

$$\text{C: } S_{\text{C}}\% = 100 - S_{\text{CO}} - S_{\text{CO}_2}$$

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