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Visible images of the catalytic combustion of methanol in a micro-channel reactor

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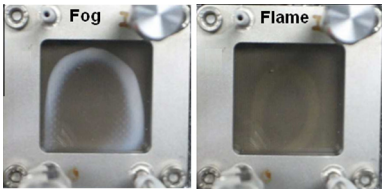
HIGHLIGHTS

- A designed micro-channel reactor allows images of fog and flames to be collected.
- The configuration of the micro-channel plate affects the flow field of reactants.
- The catalytic behavior of the catalyst changes the flow field of the reaction.
- The obtained images provide a simple method to monitor a practical flow field.

GRAPHICAL ABSTRACT



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ABSTRACT

The supply of heat to endothermic reactions through combustion reactions is the most direct and effective strategy for facilitating such reactions. The catalytic combustion of methanol at room temperature can provide broader applications of technologies that involve heat demand for chemical reactions. Hydrogen production from methanol steam reforming is a practical example. In recent years, the miniaturization of designs has become an inevitable trend due to advances in micro-electro-mechanical systems (MEMSs) technology. The micro-channel reactors prepared using MEMS technology may be a good choice for practical applications. The integration of exothermic and endothermic reactions may provide an advantage in micro-channel bed reactors. However, the progress of catalytic reactions in micro-channel reactors is not easily investigated. In this study, we designed a micro-channel reactor with a monitoring window that allowed images of fog and flames generated during the catalytic combustion of methanol. These images may advance the study of catalytic combustion on micro-channel plates. The results revealed that the configuration of the micro-channel plates affects the flow field of reactants. The catalytic activity of the catalysts also affects the gas flow field. The poor design of micro-channel configurations, which may result in a decrease in the utilization of the reaction bed area or instability of the flow field, should be avoided. The developed method provides a simple method for monitoring the practical flow field of combustion reactions in micro-channel reactors.

1. Introduction

An urgent need exists to identify new, alternative sources of energy because of limited fossil-fuel resources and because of the impact of the greenhouse effect on the Earth's climate. Proton exchange membrane fuel cells (PEMFCs) are regarded as a new, developing technology. The current practical implementations of on-board hydrogen storage systems do not meet satisfy the standards of the U.S. Department of Energy (DOE) [1]. On-site hydrogen generation has become an attractive topic for PEMFC applications. Methanol is a good candidate as a fuel for steam reforming to produce hydrogen [2].

Methanol steam reforming is an endothermic reaction. The chemical reaction is shown as follows:

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$$CH_3OH_{(l)} + H_2O_{(1)} \rightarrow 3H_{2(g)} + CO_{2(g)} \qquad \Delta H^{\circ} = 125.6\ kJ\ mol^{-1}$$

The required heat can be supplied by the catalytic combustion of methanol. The chemical reaction is shown as follows:

$$CH_3OH_{(l)} + (3/2)O_{2(g)} \rightarrow 2H_2O_{(l)} + CO_{2(g)} \qquad \Delta H^{\circ} = -726\ kJ\ mol^{-1}$$

Catalytic combustors can be operated even at room temperature [3–7]. The combustor achieves high energy efficiency due to the lack of an electronic heating system and its use of chemical energy as a heat source.

Recently, the technology of micro-electro-mechanical systems (MEMSs) has greatly advanced. A micro-channel bed is superior to a packed bed in a reactor due to the micro-channel bed's advantages of high mass transfer, high heat transfer, high surface-area-to-volume ratio and low pressure drop [8–11]. Our previous study showed that the micro-channel bed resulted in greater activity for the catalytic combustion of methanol compared with that achieved using a packed bed [6]. The literature contains some related studies about methanol steam reforming combined with the generation of heat by methanol catalytic combustion [8,11–17]. Simulation studies of reactions in microreactors have also been reported [17–23]. In addition, some studies have proposed a reactor for methanol catalytic combustion [4,6,7,14,16]. However, the literature contains no reports that discuss the progress of methanol catalytic combustion in micro-channel reactors. In our previous study, we found that the flow field of a reaction is not homogeneous in a micro-channel bed [6]. Thus, we subsequently performed this study to monitor the flow field of the reaction on the micro-channel plate for the catalytic combustion of methanol. A specific reactor was designed to capture visible images of the vapor and flame. Fog resulted from the water vapor of the products being trapped in the interface of the observation window at low temperatures. The flame images were taken at a steady state. The flow field of the reaction changed due to deactivation of the catalyst. These observations reveal the influence of the configuration of the micro-channel plate.

2. Experimental

The micro-channel plates were constructed of SUS 304 stainless steel. The dimensions of the micro-channel plate were W 50 mm × L 50 mm × thickness 0.5 mm (the dimensions of the micro-channels were W 33 mm × L 31 mm). The micro-channels were fabricated by the wet chemical etching method. The dimensions of the single flow channel were W 500 μm × L 33 mm × D 250 μm. The dimensions of the ridge were W 100 μm × L 33 mm, and the total number of micro-channels was 56 (Fig. 1a). The flow path was established with one or two rows of cylindrical columns (magnified in Fig. 1a) to redistribute the flow field of the reactants. The Pt/Al_2O_3 catalyst that was wash-coated onto the micro-channels exhibits a darker color (Fig. 1c).

Fig. 2a shows the design of the micro-channel reactor with a glass window. It consists of upper and lower covers, a glass plate, micro-channel plates and graphite gaskets. The flows of air and liquid methanol were controlled at 825 cc/min and 9 cc/h with a flow meter and a liquid pump, respectively. The air brought liquid methanol directly into the reactor. The initial reactor temperature was set to 50 °C. Air mixed with liquid methanol passed through a gasified micro-channel plate and then flowed through the micro-channel catalyst bed. The flow path of reactants is shown in Fig. 2b. After the gas and liquid were separated by the cooling system, the gas-phase composition of the reaction products was analyzed by a nondispersive infrared (NDIR) gas analyzer.

The reactants pass through one or two rows of cylindrical columns and then flow into the micro-channel; this process is called the forward flow (Fig. 1a). The reverse flow direction in the same micro-channel plate is called the reverse flow (Fig. 1b). The images of the reaction were recorded through the glass window by video cameras. The fog was only observed again when the catalytic combustion was restarted at low temperatures. Two flow types were tested for 2 days (540 min/day).

3. Results and discussion

A window was built into the reactor to allow images to be collected during the reactor's operation. A portion of the heat of catalytic combustion was lost through the window. The heat loss led to physical poisoning of the catalysts by water produced from the catalytic combustion due to a lack of sufficient heat to vaporize water from the catalyst surface at room temperature. Thus, the reactor was initially preheated to 50 °C to prevent water poisoning. The inlet creates one or two rows of cylindrical columns, as shown in Fig. 1a. The flow fields of the reactants differ when the reactants are flowed in the forward (Fig. 1a) and reverse (Fig. 1b) directions. Fig. 3 shows the relationship between the amount of CO_2 formed and the reaction time during a two-day test period of forward-flow operation. The same trend was observed for reverse-flow operation. Less CO_2 formed during the second day of operation than during the first day of operation. The reaction area for catalytic combustion in the micro-channel plates had apparently changed. The flow field of the reaction is that of the reactant residues and the formed products. This result indicates that the flow field of the reaction may vary with increased reaction time. We investigated the flow field of the reaction by collecting images of the fog and flame. According to the images of the flame, the reverse flow produces a distinct flame during the active process. The resolution of the flame images taken during the forward flow was not sufficiently high for the production of print-quality pictures. The regular variation pictures were drawn according to the real images captured from the recorded video.

3.1. Methanol catalytic combustion during reverse flow

3.1.1. Fog images

Fig. 4 shows the images of the fog taken in the reverse-flow mode on the first day during the catalytic combustion of methanol.

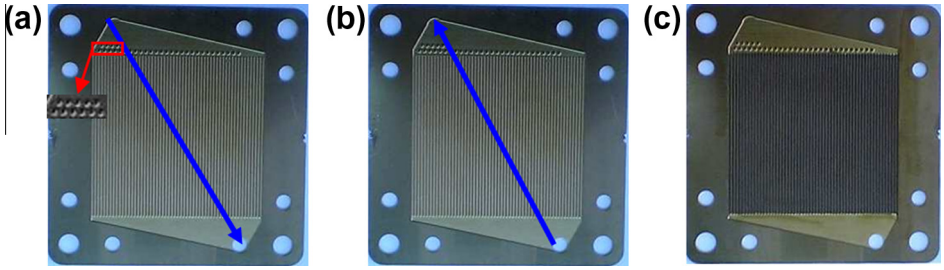

Fig. 1. Micro-channel plates (a) blank-forward flow direction; (b) blank-reverse flow direction; (c) wash-coating on catalysts.