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Numerical simulation and experimental study of hydrogen production from dimethyl ether steam reforming in a micro-reactor

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ABSTRACT

To enhance the heat and mass transfer during dimethyl ether (DME) steam reforming, a micro-reactor with catalyst coated on nickel foam support was designed and fabricated. A two-dimensional numerical model with SIMPLE algorithm and finite volume method was used to investigate 1) the fluid flow, 2) the heat transfer and 3) chemical reactions consist of DME hydrolysis, methanol steam reforming, methanol decomposition and water gas shift reactions. Both the numerical and the experimental results showed that the DME conversion in the micro-reactor is higher than that in the fixed bed reactor. The numerical study also showed that the velocity and the temperature distribution were more uniform in the micro-reactor. Wall temperature, porosity and steam/DME ratio have been investigated in order to optimize the process in the micro-reactor. The wall temperature of 270 °C and the steam/DME feed ratio of 5 were recommended. Meanwhile the results indicate that a larger porosity will give a higher DME conversion and CO concentration.

Introduction

Hydrogen energy is one of the most promising new energies. The hydrogen fuel cell has been regarded as the best way to use the hydrogen energy for its high energy efficiency and environmentally friendly. The idea of the distributed hydrogen production by the fuel cells has attracted much attention in recent years [1,2]. Several hydrocarbon fuels have been studied as the sources for the hydrogen production such as methane, methanol and ethanol. Compared with those hydrocarbon sources, dimethyl ether (DME) is an ideal fuel as a hydrogen carrier because of its high H/C ratio and high energy density. Furthermore, DME is inert, non-carcinogenic, non-mutagenic, non-corrosive, virtually non-toxic and has similar physical properties with the liquefied petroleum gases (LPG) that can be stored and handled conveniently [3].

Hydrogen production from DME steam reforming (DME SR) is a two-step process. The first step is the conversion of DME to methanol by catalyzed hydrolysis over solid-acid catalysts such as zeolites and γ-Al_2O_3. The second process is the

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methanol steam reforming (MSR) over the $CuO/ZnO/Al_2O_3$ catalyst [4,5]. Semelsberger et al. [6] examined the hydrolysis of dimethyl ether to methanol using various acid catalysts and showed that the Brønsted acid site is an important factor for the DME hydrolysis. Feng et al. [7] mixed the $CuO/ZnO/Al_2O_3/ZrO_2$ catalyst with a series of solid acids and studied the influences of the reaction temperature, space velocity and feed molar ratio on the DME steam reforming. Li et al. [8] investigated the structure and the performance of the $CuO/ZnO/Al_2O_3/Cr_2O_3$ catalyst, which was prepared by the co-precipitation coupling with mechanical mixing method. Catalysts have attracted much attention but there are few reports about the numerical simulation on DME SR. Numerical simulation is a method which can simulate the reaction process to find the best reaction conditions and optimize the structure of the reactor. Li et al. [9] used a steady-state, laminar, two-dimensional axis-symmertric model to investigate the temperature gradient and hydrogen composition of steam reforming of DME in a processor packed with a $CuO/ZnO/Al_2O_3/ZrO_2$ + ZSM5 catalyst numerically. Chein et al. [10] established a model coupled with steam reforming and catalytic combustion in the annulus reactors to study the influence of methanol to air ratio, reaction temperature and heat transfer on the methanol steam reforming. Perng et al. [11] developed a three-dimensional numerical model to investigate the effects of geometrical and thermo-fluid parameters on the CO and CO_2 products as well as the heat and mass transfer in a cylindrical methanol steam reformer with a constant-volume catalyst bed. Badra et al. [12] proposed a method for recovering the wet-based mole fractions on experimental study and simulated the catalytic combustion of DME and other fuels at different T_{jet}, φ, and *Re* using FLUENT-13.

In this paper, a steady numerical model of DME SR was established and the performances of the $CuO/ZnO/Al_2O_3$ + ZSM5 catalyst under different conditions were studied. The performance of DME SR in the fixed bed was compared to that in the micro-reactor. The effects of wall temperature, the steam to DME ratio and the porosity were investigated in the micro-reactor with catalyst coated on nickel foam support.

Mathematical model and control equations

Mathematical model

The mathematical model of the reforming process in the reactor can be described by the conservation equations of mass, momentum, energy and species transport. The model should be simplified for the calculation under the following assumptions.

(1) All species of gases in the reactor are ideal gases.
(2) The catalyst bed can be regarded as a porous media.
(3) Due to the Reynolds number is lower than 10, the flow field can described as laminar and incompressible.
(4) The reactor temperature should not be high, so the thermal radiation and the heat conduction in the mixture gases can be ignored.

Based on the above assumptions, the equations of mass, momentum, energy and species in the porous region can be written as follows.

$$\text{Continuity equation}: \nabla\cdot(\varepsilon\rho\vec{v}) = 0 \tag{1}$$

$$\text{Momentum equation}: \frac{1}{\varepsilon^2}\nabla\cdot(\rho\vec{v}\,\vec{v}) = \nabla\cdot\left\{-pI+\frac{\mu_m}{\varepsilon}\left[\nabla\vec{v}+(\nabla\vec{v})^T\right] - \frac{2\mu_m}{3}I\nabla\cdot\vec{v}\right\}+S_i \tag{2}$$

$$\text{Energy equation}: \nabla\cdot(\varepsilon\rho c_p T) = \nabla\cdot(\lambda_e\nabla T)+Q_c \tag{3}$$

$$\text{Component transport equation}: \nabla\cdot\left\{\varepsilon\rho\vec{v}m_i-\rho m_i\sum_{j=1}^{N}\left[D_{ij}\left(\nabla x_i+(x_i-m_i)\frac{\nabla p}{p}\right)-D_i^T\frac{\nabla T}{T}\right]\right\} = r_i \tag{4}$$

In these equations ε is the porosity of the catalyst. λ_e is the effective thermal conductivity of the catalyst bed [13].

Physical model

In the fixed bed reactor the $CuO/ZnO/Al_2O_3$ + ZSM5 catalyst is loaded in the middle of the tube as shown in Fig. 1(a), while in the micro-reactor the catalyst is supported on the nickel foam with 100 pores per inch. To establish the physical model of the micro-reactor, the irregular pores must be simplified. In this study, The nickel foam was regarded as straight channels with viscous resistance and inertial resistance as shown in Fig. 1(b). A 2-D model was established which is similar to the model proposed by Suh et al. [14]. The parameters of the physical model are shown in Table 1.

Chemical reaction model

The reaction mechanisms of methanol decomposition and methanol steam reforming have been studied, but the report on the integrated process of DME SR is rare. Since DME SR is a

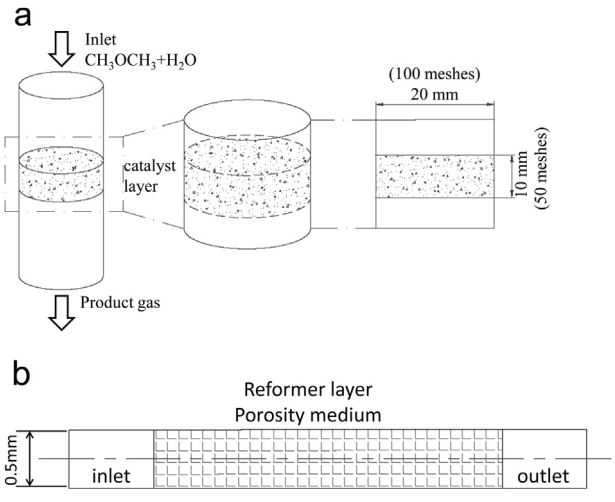

Fig. 1 – The geometrical model of (a) the fixed bed reactor and (b) the Ni foam micro-reactor.