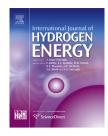


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Effect of Cr promoter on performance of steam reforming of dimethyl ether in a metal foam micro-reactor



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ABSTRACT

The CuZnAl/HZSM-5, CuZnAlCr/HZSM-5, CuZnAlZr/HZSM-5, CuZnAlCo/HZSM-5, and CuZnAlCe/HZSM-5 catalysts that were prepared by a co-precipitation method was used for hydrogen production from steam reforming of dimethyl ether (SRD) in a metal foam microreactor. These catalysts were characterized by means of XRD, TPR, SEM and BET surface areas. The results showed that promoter Cr can reduce the average pore diameter and reduction temperature of catalyst. The conversion of dimethyl ether and hydrogen yield reaches 99% and 95% respectively over CuZnAlCr/HZSM-5 catalyst under a relatively lower reaction temperature. The obtained hydrogen-riched gas is easy to purify and meet the need of polymer electrolyte membrane fuel cell. The effects of reaction temperature, space velocity and steam to DME ratio on SRD were investigated in a metal foam micro-reactor. At the conditions of T = 250 °C, the space velocity of 3884 ml/(g h), steam to DME = 5, DME conversion of >97% were obtained over the CuZnAlCr/HZSM-5 catalyst without obvious deactivation during 50 h.

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Introduction

Polymer electrolyte membrane fuel cell (PEMFC) vehicles fueled by hydrogen are one of the most promising substitutes for internal combustion engine automobiles due to their high efficiency and environmental friendly [1–4]. Dimethyl ether (DME) was proposed as a substitute and can used as a source of hydrogen rich fuel cell feed. New processes are being commercialized to produce DME is a single step via autothermal reactors and slurry phase reactors [5,6]. In addition, DME is relatively inert, non-corrosive and non-carcinogenic. The physical properties of DME are similar to those of liquefied petroleum gases (LPG). DME can be handled and transported through the LPG devices [7]. Furthermore, the steam reforming of DME (SRD) can proceed at a relative low temperature (<275 °C). SRD is a promising hydrogen source for cell vehicles.

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Steam reforming of DME proceeds via successive two-steps [8]. The first step is hydration of DME to methanol (Eq. (1)), the second step is steam reforming of methanol (Eq. (2)).

 $\label{eq:DME} DME \ hydrolysis: CH_3OCH_3 + H_2O = 2CH_3OH \ \Delta H^\circ = 37 \ kJ/mol \eqno(1)$

MeOH steam reforming (SRM) : $CH_3OH + H_2O$ = $3H_2 + CO_2 \Delta H^{\circ} = 49 \text{ kJ/mol}$ (2)

DME steam reforming :
$$CH_3OCH_3 + 3H_2O = 6H_2 + 2CO_2 \Delta H^\circ$$

= 135 kJ/mol

(3)

Usually, DME hydrolysis is catalyzed over solid-acid catalysts such as zeolites and γ -Al₂O₃ [9–12], while SRM is catalyzed over Cu- [13,14] or precious-based catalysts [15-17]. Al₂O₃ typically show an excellent DME hydrolysis activity at temperature above 300 °C due to its moderate acid amount of weak acid strength. HZSM-5 and H-mordenite have higher hydrolysis activity than alumina due to their strong acid sites. Cu-based catalyst coupled with zeolites can proceed the SRD with a total DME conversion and a low CO selectivity at lowtemperature (<275 °C). However, the coke deposition can deactivate the catalysts cause side reactions such as DME decomposition (Eq. (4)), methanol decomposition (Eq. (5)) or Boudouard reaction (Eq. (6)). Considering to get through these problems, the Cu-based catalyst and zeolites have been modified to improve the activity and stability [12,18-20]. Wang et al. [12] used the zeolites including HZSM-5, H β , HM and HY coupled with CuO/ZnO/Al₂O₃ for SRD. According to the

Nickel-Strunz classification, HZSM-5 belong to pentasil zeolite (eight 5-membered rings); HM (H-Mordenite) and HY belong to tabular zeolites (Chains of 6-membered rings); Hß (H-beta) has three-dimensional 12-membered ring pores with an interconnected channel system structure. The results showed that the acid strength and acid amount of zeolites played a significant role on SRD. The bi-functional catalyst composed of HZSM-5 and Cu/ZnO/Al₂O₃ had higher catalytic and low CO selectivity at relative low temperatures. In our work, metal oxide promoters were introduced to catalyst Cu/ ZnO/Al₂O₃/HZSM-5 for SRD in a metal foam micro-reactor. Reaction temperatures, space velocities and proportions of steam to DME were investigated in steam reforming of DME over CuZnAl/HZSM-5, CuZnAlCr/HZSM-5, CuZnAlZr/HZSM-5, CuZnAlCo/HZSM-5, and CuZnAlCe/HZSM-5 bi-function catalysts.

DME decomposition :
$$CH_3OCH_3 \rightarrow CH_4 + CO + H_2 \Delta H^\circ$$

= 1 kJ/mol (4)

 $\label{eq:MeOH} \mbox{MeOH decomoposition}: \mbox{CH}_3\mbox{OH} \rightarrow \mbox{CO} + 2\mbox{H}_2 \ \mbox{\Delta}\mbox{H}^\circ = 90 \ \mbox{kJ/mol} \end{tabular} \end{tabular} \mbox{(5)}$

Boundouard reaction :
$$2CO \rightarrow C + CO_2 \Delta H^{\circ} = -172 \text{ kJ/mol}$$
 (6)

Experimental

Fabrication and integration of micro-reactor

The SRD system consisted of vaporizer, mixer, reformer and electrical heater was sketched in Fig. 1a. The capacity can be

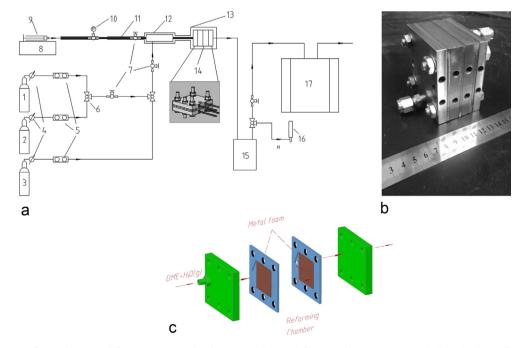


Fig. 1 – Diagram of SRD by metal foam reactor, the integrated metal foam micro-reactor and sketched configuration of the reactor 1, $H_2(5,v\%)/N_2$ 2, N_2 3, DME 4, reducing valve 5, mass flowmeter 6,three-way valve 7, stop valve 8, syringe pump 9, syringe 10, pressure gauge 11, strip heater 12, mixer 13, thermal insulating layer 14, metal foam micro-reactor 15, cold trap 16, soap film flowmeter 17, gas chromatograph.

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