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Catalytic Combustion of CH_4/Air Mixtures over Metal Foam Monoliths

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Abstract

Utilizing a catalyst in micro burners is an only possible solution to help stabilize fuel combustion. The present work focuses on investigating the catalytic combustion of methane over metal foam monolithic catalysts. The 4.8wt.% and 3.2wt.% Pd/Al_2O_3/Fe-Ni metal foams (Foam I and II) were prepared through a series of treatment of coating and impregnation. The catalytic performance of both the metal forms for methane catalytic combustion was tested in a catalytic reaction system. The results showed that Form I exhibited higher catalytic activity at low reaction temperature. The reaction temperature and flow rate of reactant mixture play an important role on the reactivity of the catalysts. The results also disclosed that the methane concentration has a weak impact on the catalytic activity, which means that the metal foams developed are effective in a wide range of fuel-air ratios.

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Keywords: Metallic foam; Monolithic catalysts; Methane catalytic combustion; Micro/Small combustor

1. Introduction

With the rapid development of micro-electromechanical systems (MEMS), the demands for miniaturized power sources are growing quickly. The high energy densities of hydrocarbon fuels make it possible for combustion-based micro power generation systems to drive the MEMS in the future. So, micro-combustors using hydrocarbons as fuel are a key component and may play a vital role in the portable production of energy. However, compared with the traditional combustors, combustion in micro-combustors becomes less steady and less efficient due to the intensified heat loss from the flame to the combustor wall, and reduced residence time. A prevalent technique to diminish the effect of the increased heat loss is to utilize a heat recirculating combustor which allows heat to be transferred from the products to the reactants. And utilizing a catalyst in a micro burner is a possible solution to help stabilize fuel combustion [1].

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Much research has been done to investigate the effects of addition of a catalyst on the extinction limits and thermal performance of small catalytic combustors [2, 3]. The catalysts utilized in those small catalytic combustors are thin plate and use the anodized alumina flat, yttria-stabilized zirconia (YSZ) felts, Pt player as catalyst supports. Recently, several kinds of metal foams, with highly porous open-cell and high-temperature resistance, were used as the base matrix material of the catalytically activate absorber in reforming of methane with carbon dioxide. But there is few application of metal foams in micro- or small- combustion. Spadaccini et al. [4] found that the addition of a porous platinum catalyst to their micro-scale gas turbine allowed for an increase in operable mass flow rates through the system, leading to an 8.5-fold increase in power density over the maximum achieved for a non-catalytic system of similar geometry.

In the present work, the Fe-Ni alloy metal (30%Ni-70%Fe) foams was first selected as a base material for the catalyst support in the continuous-flow fixed-bed reaction system. Furthermore, the method for coating open-celled metal foams with a thin layer of γ-Al_2O_3 was developed. The metal foam based monolithic catalysts were prepared and their activities of methane/air combustion were evaluated.

2. Experimental

2.1 Materials

A Fe-Ni alloy metal (30%Ni-70%Fe) foam disk was used for the preparation of the catalyst support for methane/air catalytic combustion. The disk has a diameter of 20mm, thickness of 12mm, open porosity of 95% and cells diameter of 200μm. $Pd(NO_3)_2{\cdot}2H_2O$ used for the catalyst preparation was purchased from Sinopharm Chemical Reagent Beijing Co., Ltd. AlOOH (PURAL SB powder, BET surface area 238 m^2/g) was purchased from Sasol Germany GmbH, and was used as washcoat precursors.

2.2 Preparation of the alumina sol and the metal foam monolithic catalysts

AlOOH powder (7.0 g) was mixed with 110 mL distilled water. Then, 8.37 mL of 1 M HCl was added to the solution ((H^+ (mol)/AlOOH (mol) = 0.08)). Next, the mixture was refluxed for over 5h at 85°C. Thus, a stable and homogeneous alumina sol was obtained. The metal foam monolithic catalysts are prepared in the following way: first, a A Fe-Ni alloy metal foam disk was degreased by ultrasonic in ethanol, and washed in the high-purity de-ionized water. Then, the Fe-Ni alloy metal foam disk dried in air was soaked into the alumina sol for 30 min, and then taken out and was blown by air. After that was dried at room temperature for 24 h. γ-Al_2O_3 coating on Fe-Ni alloy metal foam disk was obtained by calcining at 500°C for 4h. Secondly, $Pd(NO_3)_2$ solution was added drop by drop to as-prepared Al_2O_3/ Fe-Ni foam, and then allowed to dry for 24 h at room temperature. Finally, the Pd/Al_2O_3/ Fe-Ni foam was obtained after it was calcined at 500°C for 5 h. Two samples (metal form I and II) of different Pd loadings were obtained as specified in Table 1.

Table 1. Composition of metal foam I and II

Samples	Al_2O_3 coating[a] (wt.%)	Pd[b] (wt.%)
Pd/Al_2O_3/Fe-Ni foam Ⅰ	9.9	4.8
Pd/Al_2O_3/Fe-Ni foam Ⅱ	9.9	3.2

a, based on the mass of the foam matrix; *b*, based on the mass of Al_2O_3/Fe-Ni foam.

2.3 Experimental facility and combustor

Figure 1 shows the test system. The experimental system was designed to allow for efficient data acquisition on the extinction limits and fuel conversion under heterogeneous (catalytic) operation conditions. Three mass flow controllers were used to regulate the flow rates of oxygen, nitrogen and methane in the combustor. A mixture of 21% O_2 and 79% N_2, simulating air, was used as oxidizer. The mixer was for mixing the gases sufficiently. The exhaust gases were condensed in a cooling trap connected to the outlet of the reactor. And the dry effluent gases were analyzed by gas chromatography equipment (Agilent 7890A) with a TCD detector and a FID detector to determine gas compositions. The carbon balance was calculated according to the inlet and outlet carbon content. The methane conversion (X_{CH4}) was calculated by the following equation:

$$X_{CH_4} = \frac{F_{CH_4,in} - F_{CH_4,out}}{F_{CH_4,in}} \times 100\% \quad (1)$$

Where F is molar flow rate.

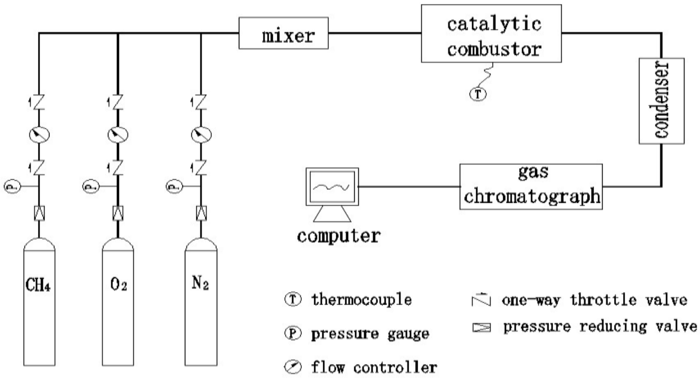

Figure 1. Test system

A single turn heat recirculating combustor was used. It was fabricated from quartz glass. The combustor was composed of two concentric tubes, the inner tube with 20mm in diameter and 180mm in length, the outer tube of a diameter 32mm. The metal foam monolithic catalyst was placed in the heat recirculating combustor. The quartz tube was heated to the given temperature (300~550°C) to carry out the catalytic (flameless) combustion. The temperature of the catalyst bed was measured using a K-type thermocouple placed at the front of the catalyst bed.

3. Results and discussion

3.1 Activity tests

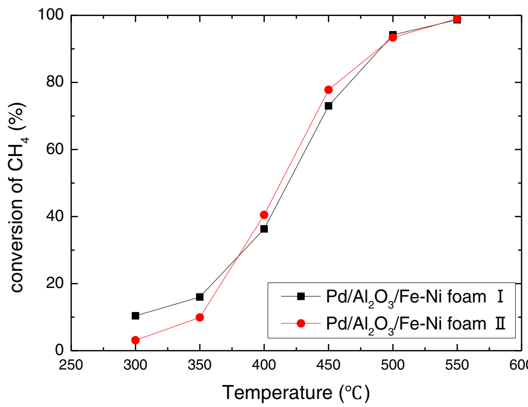

Figure 2. Methane conversion of metal foam I and II at different temperatures

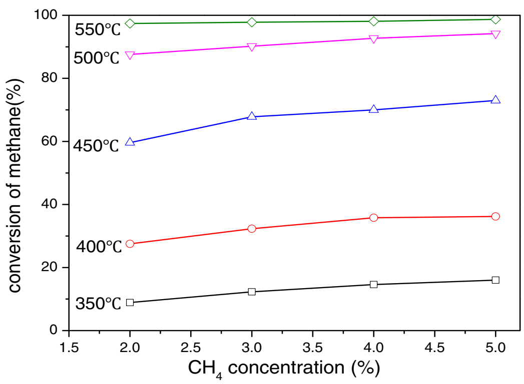

Figure 3. Influences of CH_4 concentration on the actibity of themetal foam I (the flow rate of CH_4-air mixture was set at 50ml/min)

In a catalytic combustion experimental system, the catalytic performance of the metal foam Ⅰ and II for CH_4 combustion were first tested at a flow rate of 50 mL/min of the fuel-air mixture with CH_4 concentration of 2.5%. As shown in Figure 2, for higher temperatures, the methane conversion of the metal foam II is slightly higher than that of the foam I and the conversion is as high as 99% at 550°C.

However, when the temperature is lower than 400°C, then, the methane conversion of the metal foam I is higher than that of the metal foam II. The results indicated that, with the Pd loading tested, the loading has a very weak influence on the reactivity of the catalysts.

Figure 3 depicts the influences of the CH_4 concentration on the activities of the metal foam I for methane combustion. From this figure, one can see that the reaction temperature has a very strong influence on the CH_4 conversion, and the CH_4 concentration has only a limited effect, although the conversion did increase slightly with it. This proves that the metal foam monolith catalysts developed in this paper can work effectively in a wide range of fuel-air ratios.

The influences of the CH_4-air mixture flow rate on the activity of the metal foam I were also obtained and presented in Figure 4 at different temperatures. As one can see, the flow rate did produce a considerable effect on the CH_4 conversion. For example, at the temperature of 550°C, as the reactant mixture flow rate was increased from 100 mL/min to 200 mL/min, the conversion decreased from 76% to 62%. However, it should be noted, this influence is weakened as the reaction temperature is reduced. For example, within the same flow rate change, at 400°C, the conversion is only reduced from 11% to 8%. This indicated that, for a given flow rate, the monolith catalyst should be carefully designed and verified.

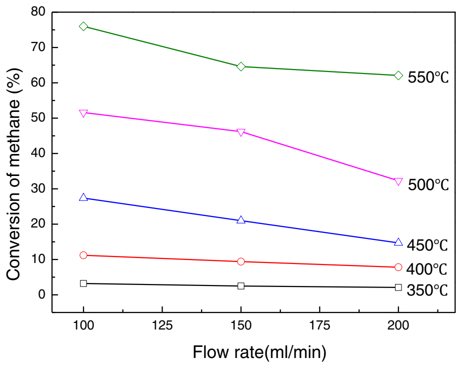

Figure 4. Influences of flowrate of the CH_4-air mixture on the conversion of metal foam I at different temperatures (CH_4 concentration is 5%)

4. Conclusion

The metal foam monolithic catalysts prepared by applying Pd/Al_2O_3 catalyst on Fe-Ni alloy metal foam were tested in the methane catalytic combustion. Compared to the Foam II, Foam I exhibited relatively higher catalytic activity for CH_4 catalytic combustion of methane at low reaction temperature. And the experimental results also show, within the range tested, the Pd loading has only a limited influence on the activity for the CH_4 combustion at high temperature. This is perhaps bucause that the Al_2O_3 coating played a more important role on the reactivity of the resulted catalysts. Our limited experimental results showed that the metal foam monolith catalyst can work in a wide range of CH_4-air ratio and the conversion can be as high as 99%. Although, the effects of reaction temperature, methane concentration, and flow rate of reactant mixture have been experimentally investigated, the long-term stability of Pd/Al_2O_3/Fe-Ni foam needs to be further verified and studied.

Acknowledgments

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