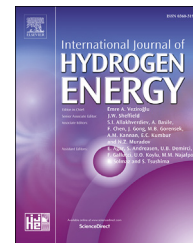




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Hydrogen production from partial oxidation of methane over dielectric barrier discharge plasma and NiO/ γ -Al₂O₃ catalyst

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ARTICLE INFO

Article history:

Received 28 March 2017

Received in revised form

31 May 2017

Accepted 2 June 2017

Available online xxx

Keywords:

Dielectric barrier discharge

Hydrogen production

Methane

Catalysis

Synergistic effect

ABSTRACT

Hydrogen production from partial oxidation of methane under the combination of dielectric barrier discharge (DBD) plasma and NiO/ γ -Al₂O₃ catalyst with cordierite honeycomb monoliths as substrate was investigated. The results showed that obvious synergistic effect was generated between DBD plasma and catalyst. Compared with the DBD plasma reactor without catalyst, the CH₄ conversion and H₂ yield increased from 60.1% and 21.3% to 83.6% and 28.4%, respectively. When the discharge power is above 70 W, the combination of DBD plasma and NiO/ γ -Al₂O₃ catalyst promotes partial oxidation of methane. The catalyst was characterized by X-ray diffraction (XRD). NiO on the surface of catalyst was reduced to Ni because of the introduction of DBD plasma. The activity of catalyst at low temperature was improved, and the generation of oiliness by-products was significantly reduced.

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Introduction

The global car ownership is increasing rapidly. The popularization of the vehicle brings convenience to people and leads to two serious problems simultaneously: oil crisis and air pollution. In addition, the energy efficiency of the conventional engines is low due to the limitation of the Carnot cycle, exacerbating the above two problems. Therefore, highly efficient and environment-friendly vehicles should be developed immediately. With the continuous development of fuel cell technology, fuel-cell vehicles have gradually become one of the most promising vehicles [1–3]. The fuel of fuel cell vehicle is hydrogen. Currently, hydrogen is mainly produced by

methane reforming [4–6]. Nevertheless, reforming of methane by traditional catalytic methods still faces two challenges. First, methane reforming by traditional catalytic methods is proceeded under high temperature (700–1000 °C) and high pressure (>10 atm), incurring high cost. Second, the performance of the catalysts is unstable at high temperature, and the formation of coke causes rapid deactivation of the catalysts, especially for non-noble metal catalysts, such as nickel-based catalysts. Therefore, new methane reforming technology should be developed to improve the activity of catalysts and the stability of catalysts at high temperatures [7–10].

In recent years, non-thermal plasma technology has attracted more and more attention from researchers for

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<http://dx.doi.org/10.1016/j.ijhydene.2017.06.008>

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converting methane into hydrogen [11–14]. In non-thermal plasma, the electrons are highly energetic (1–10 eV), which can easily break down CH₄ molecules and produce a variety of active species (excited atoms, free radicals, ions) and effectively stimulate plasma-chemical reactions. Compared with traditional catalytic methods, non-thermal plasma technology can convert methane into hydrogen at low temperatures. Some types of non-thermal plasma technology have been investigated for methane reforming including dielectric barrier discharge, glow discharge, and gliding arc discharge. Hydrogen can be generated at low temperatures by plasma reforming. However, hydrogen selectivity is low. If non-thermal plasma and heterogeneous catalyst are combined, H₂ yield and the activity of catalysts can be improved at low temperatures [15–22]. In addition, because of its uniqueness and versatility, plasma techniques also can be widely used in catalyst preparation and reaction process [23–25]. But there is no report which is related to hydrogen production from partial oxidation of methane over dielectric barrier discharge plasma and NiO/γ-Al₂O₃ catalyst using cordierite honeycomb monoliths as substrate at atmospheric pressure and ambient temperature.

In this study, hydrogen production from partial oxidation of methane was investigated using the combination of dielectric barrier discharge (DBD) plasma and NiO/γ-Al₂O₃ catalyst at atmospheric pressure and ambient temperature. Methane partial oxidation is an exothermic reaction, so its energy utilization rate is high. Parameters (discharge power, CH₄/O₂ mole ratio, and total feed flow rate) that had an effect on CH₄ conversion, product selectivity, and H₂ yield were studied. The oiliness by-products and activity of catalyst at low temperatures were also investigated.

Experimental section

Catalyst preparation

The NiO/γ-Al₂O₃ catalyst was prepared by traditional impregnation method using cordierite honeycomb monoliths as substrate. The cordierite honeycomb monoliths were impregnated in a solid content alumina sol (about 10%–15%) prepared by mixing deionized water and pseudo boehmite for 30 min. The obtained catalyst was dried at 100 °C for 2 h and calcined at 600 °C for 5 h after being taken from the sol, and the redundant sol in the pores was blown off with an auralave. The above steps were repeated four times to make the loading of γ-Al₂O₃ at 10%–15%. The catalyst was impregnated in solution of 1 mol/L nickel nitrate for 2 h, dried at 100 °C for 4 h, and calcined at 500 °C for 5 h. The method was repeated several times to make the loading of NiO around 10%. The geometrical parameters of the cordierite honeycomb monoliths are shown in Table 1.

Catalyst characterization

X-ray diffraction (XRD) patterns of the catalyst samples were recorded by a Rigaku D/max 2500 diffractometer using a Ni-

Table 1 – Geometrical parameters.

Parameter	Range
Pore density (cps)	300
External diameter (mm)	25
Inner diameter (mm)	8
Channel shape	Triangle

filtered Cu-Kα radiation, which was operated at 40 kV, 40 mA, a step size of 0.18°, and rate of 10°/min from 2θ = 5°–80°. The catalyst samples also were tested by a Nexus 670 Fourier Transform Infrared Spectroscopy (FTIR) using a MCT detector and KBr beam splitter, which was operated at the resolution 4 cm⁻¹, the scan number 32, the scope of testing 400–4000 cm⁻¹.

Experimental apparatus

A schematic of the experimental setup is shown in Fig. 1. The feed of methane and air was controlled by S49-32B/MT mass flow controllers and flowed into the DBD plasma reactor after mixing in a three-way valve. The flow rate of methane and air was displayed on the Flow Readout Box (FRB, MT50-3J). After reaction, the output gases were analyzed by a gas chromatography (GC-9900). The GC contains two columns, which are filled with TDX-01 (1.5 m long) and GDX-105 (2 m long), respectively. Helium was used as carrier gas.

The DBD plasma reactor is shown in Fig. 2. The reactor consisted of two coaxial quartz tubes whose inner and outer diameters were 6 mm, 26 mm and 8 mm, 30 mm, respectively. A stainless steel bar of 5 mm diameter served as high voltage electrode was inserted in the inner tube, and a copper mesh electrode serving as grounding electrode was wrapped around the outer tube. The length of the discharge zone was 10 cm, resulting in the reaction volume of approximately 56.6 cm³. The power was supplied by an AC power supply. Waveform of discharge voltage and current was detected by a four-channel digital oscilloscope (Tektronics, TDS2014C). The discharge power (P) is defined as

$$P = \text{Frequency} \times \int_0^{\tau} i(t)u(t)dt \quad (1)$$

CH₄ conversion (X_{CH₄}), H₂ selectivity (S_{H₂}), CO selectivity (S_{CO}), and H₂ yield (Y_{H₂}) are defined as

$$X_{\text{CH}_4} (\%) = \frac{\text{moles of CH}_4 \text{ converted}}{\text{moles of CH}_4 \text{ input}} \times 100 \quad (2)$$

$$S_{\text{H}_2} (\%) = \frac{\text{moles of H}_2 \text{ produced}}{2 \times \text{moles of CH}_4 \text{ converted}} \times 100 \quad (3)$$

$$S_{\text{CO}} (\%) = \frac{\text{moles of CO produced}}{\text{moles of CH}_4 \text{ converted}} \times 100 \quad (4)$$

$$Y_{\text{H}_2} (\%) = \frac{\text{moles of H}_2 \text{ produced}}{2 \times \text{moles of CH}_4 \text{ input}} \times 100 \quad (5)$$

The range of the parameters is listed in Table 2.

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