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Biogas reforming by the honeycomb reactor for hydrogen production

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ARTICLE INFO

Article history:

Received 16 June 2015

Received in revised form

7 September 2015

Accepted 9 September 2015

Available online xxx

Keywords:

Biogas reforming

Cordierite honeycomb reactor

Ni/CeO₂

Response surface method

Hydrogen production

ABSTRACT

In this study, it was proposed that the Ni/porous-CeO₂ coated cordierite honeycomb reactor could enhance the performance of biogas reforming for hydrogen production, compared with the packed back reactor. Current results suggested that the conversion methane and yield of hydrogen using honeycomb were increased by 22.24% and 72.52% for biogas reforming and 43.12% and 41.29% for methane steam reforming, compared with packed bed reactor at 900 °C and WHSV = 5hr⁻¹. With proposed honeycomb reactor for the reaction, methane conversion in biogas reforming was low at the temperature below 500 °C and was higher than 80% at the temperature over 700 °C and WHSV = 3hr⁻¹. H₂ and CO were the major product at the higher temperature. However, conversion of methane, yield of H₂, and yield CO were lower than those in the methane steam reforming. Thus, proposed catalyst and the reactor were highly effective for the hydrogen production from the biogas reforming.

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Introduction

Biogas, methane and carbon dioxide, generated by the anaerobic digestion processes from the municipal or the agricultural waste are considered as one of the important new energy resources for the next generation. The biogas reforming and the methane steam reforming by the cordierite honeycomb reactor with impregnated Ni/CeO₂ catalyst were proposed in this study. The honeycomb catalytic reactor was commonly installed in the gasoline powered vehicles for the pollution abatement since late 1970's [1]. The honeycomb reactor was also studied in various small hydrocarbons

reforming for hydrogen production on kinetics studies [2–7] as well as the mathematical simulations [3,8,9]. With properly prepared catalyst over the surface of the honeycomb structure, the apparent activity of the catalytic reaction could be highly improved due to the low pressure-drop over the reactor.

Previous studies from our team revealed that the Ni/CeO₂ was catalytic active for the biogas reforming to produce hydrogen [10,11]. Porous CeO₂ was prepared by the reverse precipitation method to control the surface area of the CeO₂, because the activity of the catalyst was highly affected by its structure. The activities of supported Ni catalyst over CeO₂ at different Ni loadings for the biogas reforming were also

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<http://dx.doi.org/10.1016/j.ijhydene.2015.09.018>

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studied. The results elucidated that the proposed catalyst was high catalytic activity for hydrogen production and low activity for coke forming during the reforming reaction.

Besides the results from our studies, the studies of the biogas reforming, CO₂ reforming of CH₄, were reviewed by Bradford [12] according to the reaction historical background, CH₄ activation, CO₂ activation, carbon deposition, and the reaction kinetics and mechanisms of the reactions. It also elucidated that the noble metal catalysts were the most catalytic active for the reaction. Pakhare also confirmed the activity of CO₂ reforming of methane over the noble metal catalyst [13]. Basically, Ru, Rh, Pd, Ir, Pt, Re, and Au could serve as the catalyst or the promoters over different supports for the biogas-reforming reactions. Although the noble catalysts were believed to be active for the biogas reforming reaction, significant amount of the studies of dry reforming by the non-noble metal catalysts, such as Cu, Fe, Co, Ni, and W, were also catalytic active of biogas reforming for hydrogen production [12, 13]. Among all these studies, supported nickel catalyst was the most catalytically active for the CO₂ reforming of CH₄. Recently, biogas reforming for hydrogen production by the CeO₂ supported non-noble catalyst was also founded [14–19]. Bereketidou et al. suggested that 8% of Ni over 20wt% of ceria-alumina had the best activity for biogas reforming and H₂/CO ratio of 1.83 was attained [15]. Chang et al. also studied on the biogas reforming on the Ni/ZrO₂ catalyst and found that with ceria doped catalyst, the activity of the catalyst and the resistance of coke formation were enhanced [16]. Corthals et al. found that when the MgAl₂O₄ supported Ni catalyst was used, the presence of CeO₂/ZrO₂ would enhance the activity of CH₄ and CO₂ conversion and coke formation resistance [17]. Djinic et al. worked on the Ni–Co bimetallic catalyst on various ratio of ceria-alumina support. The results suggested that enhancing surface oxygen mobility over the catalyst by the support would also reduce the activity of coke formation in the biogas reforming [18]. All these studies suggested that Ni was catalytic active for the biogas reforming to produce hydrogen. With the presence of CeO₂ as the support would also enhance the surface oxygen mobility and resistance of coke formation.

In this study, biogas reforming over a Ni/porous-CeO₂ coated cordierite honeycomb catalytic reactor for hydrogen production was proposed. To compare the catalytic activity, the steam reforming of methane and the packed bed reactor for the reactions were also studied. The reaction temperature and the weight hourly space velocity (WHSV, hr⁻¹) were the major experimental design parameters. The second order Response Surface Method (RSM) was also adapted to elucidate the optimum reaction condition.

Materials and methods

Catalyst preparation

The Ni/ceria catalyst for this study was prepared by the reverse precipitation method [10]. The prepared powder catalyst was then wash-coated on the cordierite honeycomb-structured supporter for the kinetic study. Cerium(III) chloride heptahydrate (CeCl₃·7H₂O, Aldrich) and nickel(II) nitrate

hexahydrate (Ni(NO₃)₂·6H₂O, SHOWA) were dissolved in 50 mL of de-ionized water at the designated amount. This mother solution was mixed with the solution containing 9.4 g of hexadecyltrimethyl-ammonium bromide (CTMABr, C₁₉H₄₂BrN, Acros) in 100 mL of deionized water. The mixture was then added into 1000 mL of ammonium solution (NH₄OH, Fisher) until the pH of this solution attained 11.0. The solution was then aged at 90 °C for 90 h. The precipitate was washed, filtered, dried, and calcined at 550 °C for 5 h.

The cordierite honeycomb support was purchased from Chauher Honeycomb Ceramics Co. Ltd. (New Taipei City, Taiwan) with 10 mm in diameter and 100 mm in length. The opening pore size was 1.2 mm * 1.2 mm. The composition of honeycomb, provided by the manufacturer, was 51% of SiO₂, 34% of Al₂O₃, 13% of MgO, and 2% of cordierite. Before adding the catalyst onto the surface of the cordierite honeycomb structure, it was washed (in the following sequences) by acetone, deionized water, sodium hydroxide solution, deionized water, sulfuric acid, and deionized water to remove any residual impurities over the surface of the support. The prepared Ni/CeO₂ catalyst was then distributed in ethanol solution and transferred them onto the surface of the honeycomb support by the wash-coated method until the designated amount of metallic nickel over the catalyst surface was attained. The prepared honeycomb catalyst was then calcined at 550 °C for 5 h in air.

Characterizations

Surface area and porosity of the catalyst powder were determined using a high-resolution surface area and porosimetry analyzer (BET, Micromeritics ASAP2010). Surface morphology was determined by a cold field emission scanning electron microscope and energy dispersive spectrometer (FESEM, HITACHI S-4800). Crystallinity was determined via X-ray diffraction (XRD) (SHIMADZU XRD-6000 X-ray diffractometer), and the size of the crystallite was calculated by the Scherrer equation. The thermal stability of the prepared sample was determined by simultaneously Thermo-Gravimetric Analyzer and Differential Scanning Calorimeter (TGA-DSC, TA-Instruments, SDT-2960).

The temperature programmed reduction (TPR) experiments were done on H₂/Ar mixture (10% H₂, 30 ml/min total flow rate) from room temperature to 1000 °C (with 10 °C/min heating rate). A thermal conductivity detector (TCD) was used to monitor the effluent gas stream. The surface composition of the Ni catalyst could also be determined by this method.

Preparation of honeycomb reactor and the reaction

The prepared honeycomb catalyst was then put into a 1/2" stainless steel tube and activated by flowing 50 mL of sweeping gas with 10% of hydrogen in helium at 700 °C for two hours prior the kinetics studies.

Both biogas reforming and methane steam reforming were tested in the prepared reactor. For the biogas reforming, the molar flow ratio of CH₄, CO₂ and steam was 2.0: 1.0: 0.1. For the methane steam reforming, the molar ratio of steam and methane was controlled at 3:1. All the reactions were tested at the reaction temperature between 300 and 900 °C and the

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