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Autothermal reforming of *n*-dodecane and desulfurized Jet-A fuel for producing hydrogen-rich syngas

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ABSTRACT

Catalytic reforming is a technology to produce hydrogen and syngas from heavy hydrocarbon fuels in order to supply hydrogen to fuel cells. A lab-scale 2.5 kW_t autothermal reforming (ATR) system with a specially designed reformer and combined analysis of balance-of-plant was studied and tested in the present study. NiO–Rh based bimetallic catalysts with promoters of Ce, K, and La were used in the reformer. The performance of the reformer was studied by checking the hydrogen selectivity, CO_x selectivity, and energy conversion efficiency at various operating temperatures, steam to carbon ratios, oxygen to carbon ratios, and reactants' inlet temperatures. The experimental work firstly tested *n*-dodecane as the surrogate of Jet-A fuel to optimize operating conditions. After that, desulfurized commercial Jet-A fuel was tested at the optimized operating conditions. The design of the reformer and the catalyst are recommended for high performance Jet-A fuel reforming and hydrogen-rich syngas production.

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Introduction

The quick depletion of fossil fuel is one of the major challenges our world is facing with today. Alternative sustainable energy technologies are needed for future power generation. Developing clean and sustainable power sources for transportation sector [1] is one of the most difficult challenges in the new energy technologies. Power generation using fuel cells is a promising alternative technology which can be applied in transportation tools to reduce energy consumption. The use of fuel cells is also attractive as auxiliary electrical power units (APUs) for aircrafts, vessels, and trucks, due to the high energy conversion efficiency, and the quite low levels of

emissions [2,3]. However, the supply of hydrogen for fuel cells is a big issue, since there is no existing infrastructure for hydrogen production and storage, particularly, for transportation vehicles [4]. As a practical solution, onboard fuel processing system was proposed to convert liquid hydrocarbon fuels into hydrogen or hydrogen-rich syngas through reforming reactions. In various hydrocarbon fuels, jet fuels receive significant attention for onboard reforming because they are logistic fuels for both civilian and military aircrafts, vessels, and trucks [5]. Among different types of fuel cells, a solid oxide fuel cell (SOFC) is a promising candidate for APUs, as it can be connected to jet fuel reforming systems due to the following considerations: (a) operation temperature of SOFC (650–1100 °C) allows high temperature syngas from the

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reformer to be fed into fuel cells without cooling; (b) CO poisoning is not an issue for SOFC, and CO is one type of fuels for SOFC to produce power; (c) water-gas shift (WGS) reactor and preferential oxidation (Prox) reactor are not necessary for SOFC-based APUs, and thus the fuel reforming system is less complex and more stable [6–9].

The three main approaches for reforming of hydrocarbon fuels are steam reforming (SR), partial oxidation (POX), and autothermal reforming (ATR). There has been a number of research works studied the reforming of different fuels, such as ethanol, gasoline, diesel, and jet fuels [10–13] employing one of the above-mentioned approaches. It is understood from the literature that SR has high H_2 concentration in the reformat, but it requires lots of external heat and the system is usually bulky and heavy. POX has a compact system and the reaction is exothermic, however, the H_2/CO ratio in the reformat is relatively low. ATR is a combination of POX followed by SR and it has the most suitable characteristics for onboard fuel reforming. An ATR system has favorable H_2/CO ratio in the reformat, less coke formation tendency, no requirement of external heat source, relatively compact system size and weight, and rapid startup and dynamic responses [14–16]. Because of the advantages of ATR, it was selected as the approach of reforming in the present study. Both reforming catalyst and operation conditions can significantly influence the ATR reaction in respect to H_2 and CO concentrations, hydrogen and CO_x selectivity, as well as system energy conversion efficiency [17]. Ni-based catalysts for fuel reforming have been widely used because of the high activity and low cost. However, Ni catalysts have inherent challenges such as sulfur poisoning and coke formation [18]. Noble metals were proved to be effective in fuel reforming reactions, but the high price of noble metals is a disadvantage [19,20]. Thus bimetallic catalysts (including nickel and noble metal) have been proposed and investigated in some works for fuel reforming [21–24]. Effects of ATR operating conditions such as steam to carbon ratio (S/C) and oxygen to carbon ratio (O_2/C) were investigated both numerically and experimentally for different reformers [25–27], and it is believed that the reasonable working ranges of S/C and O_2/C are 1.25–2.5 and 0.35–0.5, respectively.

In this paper, a lab-scale 2.5 kW_t autothermal reforming system with a new reformer design and novel catalyst was experimentally studied. The used bimetallic catalyst is NiO–Rh which is mixed with promoters of CeO_2 , K_2O , and La_2O_3 , and was prepared in-house for experimental test. Using *n*-dodecane as a surrogate of Jet-A fuel, the effects of operating conditions such as reformer temperature, S/C, and O_2/C were experimentally investigated in respect to hydrogen selectivity, CO_x selectivity, and the energy conversion efficiency. Coke formation was suppressed by reducing the pre-heating temperature of the fuel. After the optimized operating conditions were determined, desulfurized commercial Jet-A fuel was tested in the ATR system.

Experimental work

Catalyst preparation

The proposed catalytic reforming section is made of a cordierite monolith substrate coated with layers including an

alumina washcoated support layer, promoters of CeO_2 , K_2O , and La_2O_3 , and active materials of NiO and Rh. The promoters and active materials are dispersed onto the support and substrate by the washcoating method [19,28–30]. The substrate used as shown in Fig. 1 is a cordierite monolith with 400 cpsi (cells per square inch) and a wall thickness of 0.25 mm, in length of 60 mm, and a diameter of 40 mm. Monolithic structure substrate is better than a packed bed because of its large open frontal area (OFA) and straight parallel channels which ensure low flow resistance. Cordierite is a commonly used ceramic material for monolithic substrates due to its low thermal expansion coefficient in the magnitude of $10^{-6} K^{-1}$, high mechanical strength up to 20.7 MPa, and high melting point over 1300 °C [14]. A cordierite monolith usually has small surface area of less than 1 m²/g. However, a large surface area is important for getting a good dispersion of the active materials (noble metal) on the surface of monolith. To overcome this problem, alumina with high surface area is deposited onto the monolith walls as the catalyst support layer. Large-pore pseudo-boehmite (70 wt% Al_2O_3 , surface area >300 m²/g, pore volume 0.8–1.0 cm³/g) is the source of alumina for the current catalyst. Ceria, potassium oxide, and lanthana are added into the catalyst as promoters. Ceria is added to increase the sulfur resilience capability of the catalyst, as recommended by Qi et al. [19] that Ce could form a stable sulfide in the temperature range of ATR to serve as a sulfur sink. It can maintain and improve the stabilization of metal dispersion and also favors carbon removal as well [31]. Potassium oxide is proved to be the most effective promoter in suppressing coke formation in ATR. The addition of potassium not only lowers the coking tendencies of alumina supports, but also prevents nickel from catalyzing the decomposition reactions [32]. Lanthana is added to improve the catalytic activity of nickel. In reforming reactions, the addition of a small amount of La could possibly increase the nickel catalyst activity significantly [33]. Lanthana is also known to prevent the temperature induced phase transitions of alumina [34].

Before washcoating of the alumina support layer, the cordierite monolith was first washed by 10% nitric acid and then calcined at 815 °C for 2 h. Micromeritics TriStar II 3020

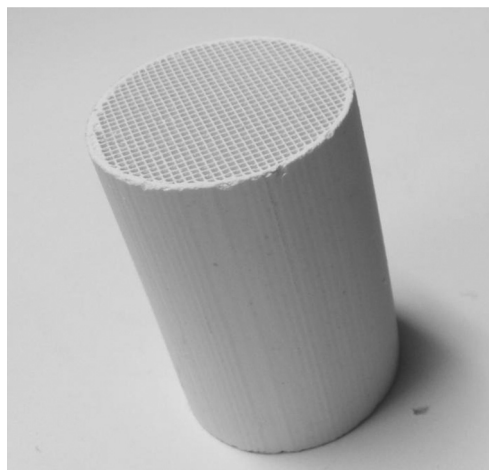


Fig. 1 – Cordierite monolith with honeycomb structure.

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