



Methane steam reforming with a novel catalytic nickel membrane for effective hydrogen production

Shin-Kun Ryi^{a,*}, Jong-Soo Park^b, Dong-Kook Kim^b, Tae-Hwan Kim^b, Sung-Hyun Kim^c

^a Department of Chemical and Biological Engineering, University of British Columbia, 2360 East Mall Vancouver, Canada V6T 1Z3

^b Korea Institute of Energy Research, 71-2, Jang-Dong, Yuseong-gu, Daejeon 305-343, South Korea

^c Department of Chemical and Biological Engineering, Korea University, 5-ka, Anam-Dong Sungbuk-Gu, Seoul 136-701, South Korea

ARTICLE INFO

Article history:

Received 7 July 2008

Received in revised form 27 March 2009

Accepted 25 April 2009

Available online 3 May 2009

Keywords:

Membrane

Methane

Steam reforming

Fuel processor

Hydrogen

ABSTRACT

This paper describes a study of methane steam reforming using a novel catalytic nickel membrane for hydrogen production. The catalytic nickel membrane was made by the uniaxial-pressing and thermal treatment of nickel powder. Since the nickel powder had catalytic activity, it was unnecessary to deposit an additional reforming catalyst on the nickel filter. The methane conversion, reformat composition, and hydrogen production rate were investigated at very high gas hourly space velocities of 22,350–100,700 h⁻¹ with a steam to carbon ratio of 3.0. When a mixture of methane and water was introduced into the catalytic nickel filter, the experimental trends exceeded the theoretical equilibrium model, because of the difference in the permeation rates of the gases, even with residence times of only ~0.036 s.

© 2009 Elsevier B.V. All rights reserved.

1. Introduction

Hydrogen is a widely used feedstock in the chemical, food, and refining industries. The demand for hydrogen is expected to drastically increase, because of its potential as a major energy source in the future [1]. Methane steam reforming is one of the most common methods of producing hydrogen. It is a reversible and endothermic process and is usually carried out at temperatures in the range of 800–1000 °C and a pressure of about 20.265×10^5 Pa (20 atm) over a nickel based catalyst [2]. Due to its heat and mass transfer problems, the conventional fixed-bed reactor has some disadvantages, such as low catalyst effectiveness factors and high temperature gradients [3].

Much effort has been made over the past few decades to improve the performance of the conventional fix-bed methane steam reforming process. Microchannel reactors (MCR) comprising micron-sized channels patterned on thin metal plates have been studied, due to their many advantages, such as their enhanced heat and mass transfer, safe control in an explosive regime, high specific-surface area, process compactness and easier scale-up without geometry changes [4,5]. However, there is still an issue which remains to be solved, namely the catalyst coating on the metal surface. Fluidized-bed reactors can improve the heat and mass transfer

in methane steam reforming [6]. Compared with the fixed-bed process, the temperature distribution in a fluidized-bed is much more uniform and the reactants come into contact with the catalyst more easily, however there is a problem of catalyst loss. Membrane reactors have been employed to improve the process performance. Due to the reversible process of the methane steam reforming, the selective and continuous removal of hydrogen from the reaction shifts it towards the product side [7–9]. However, there are temperature limitations in a membrane reactor, due to the stability of the membrane and the membrane reactor has to be enlarged to provide an area large enough to obtain a sufficient hydrogen permeation rate [8,10].

Recently, interesting studies have been reported on the use of a ceramic filter and a macroporous silicon loaded catalyst to achieve a chemical reaction with high efficiency. Engelen et al. [11] studied a catalytic filter for tar removal from biomass gasification gas. Ilcorca et al. [12] reported that they coated macroporous silicon with a Co₃O₄-ZnO catalyst by a complexation-decomposition method and carried out ethanol steam reforming.

We developed a porous nickel membrane for hydrogen separation [13], a porous nickel support for Pd-based composite membranes [14] and a nickel filter for gas cleaning [15]. Recently, Rakass et al. [16] used unsupported nickel catalysts for the in situ methane steam reforming process of solid oxide fuel cells. They used packed-bed micro reactors and the nickel catalysts had high reforming activity without coke formation at an S/C ratio of 2.0. We fabricated a catalytic nickel membrane with a nickel powder which

* Corresponding author. Tel.: +1 604 827 5563; fax: +1 604 822 6003.

E-mail addresses: pdmembrane@naver.com, sryi@chml.ubc.ca (S.-K. Ryi).

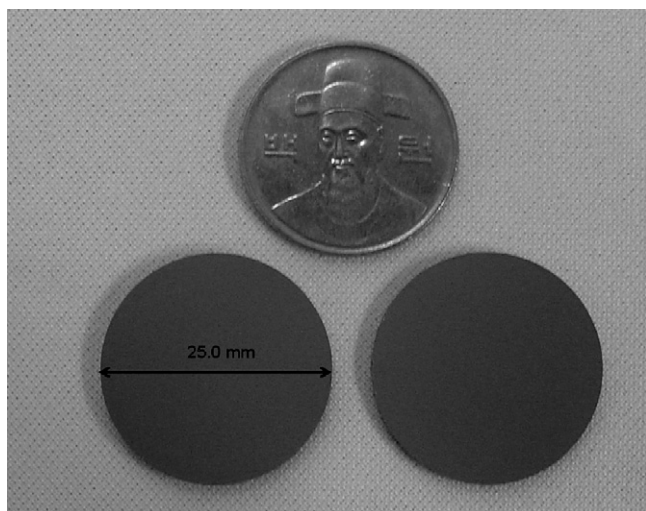


Fig. 1. Photograph of catalytic nickel membranes.

had sub-micron size pores. We used the catalytic nickel membrane for the compact methane steam reforming process with super-equilibrium conversion. The catalytic nickel membrane exhibited good heat and mass transfer properties, providing the reactor with compactness.

2. Experimental

2.1. Fabrication of catalytic nickel membrane

We used nickel powder purchased from Sigma–Aldrich. It has a purity of 99.7% and an average particle size of 3 μm with a spherical but undulated shape. In order to increase its thermal stability, the nickel powder was coated with aluminum nitrate solution by the incipient wetness impregnation method. An appropriate amount of aluminum nitrate ($\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, Aldrich) was dissolved in deionized water, and the solution added to the dried nickel powder

to obtain 0.1 wt.% Al. The alumina modified nickel powder was compressed without a binder in a metal cylindrical mold with a diameter of 25.4 mm under a pressure of 165 MPa. 4 g of the nickel powder was placed in the metal mold and then brushed with a plate type metal stick to make the surface level. The metal mold filled with nickel powder was pressed with a homemade press described in our previous study [15]. The press linearly pressed the metal mold to the set point and then held it at the set pressure for 10 s. The compressed nickel membrane was further treated at 950 $^\circ\text{C}$ under hydrogen for 2 h. The prepared nickel membrane is shown in Fig. 1. Its diameter and thickness were 25.0 and 1.55 mm, respectively. SEM analysis was used to characterize the surface of the nickel membrane. The pore size distribution, average pore size, total pore volume and porosity of the prepared catalytic nickel membrane were measured by a mercury porosimeter (Micromeritics, Autopore χ 9500).

2.2. Gas permeation measurements

Gas permeation tests were conducted using hydrogen, carbon monoxide, carbon dioxide and methane at a room temperature with a membrane assembly, as shown in the sub-figure in Fig. 2. The effective surface area of the assembled catalytic nickel membrane was about 3.1 cm^2 . The permeation apparatus consists of a membrane assembly, a pressure indicator, mass flow controllers and a digital bubble flow meter. The gases were introduced by a mass flow controller (MFC, Brooks 5850 E series) and the pressure was detected by a digital pressure regulator (Alicat, PC-100PSIG-D). The gas permeation flux was measured by a digital soap-bubble flow meter (SENSIDYNE, Gilibrator 2). The ideal selectivities were defined as the ratio of the permeation flux of hydrogen to that of the other gases at the same pressure difference.

2.3. Methane steam reforming

Methane and liquid water were supplied to the nickel membrane for methane steam reforming. Methane steam reforming tests were carried out with a steam to carbon (S/C) ratio of 3.0.

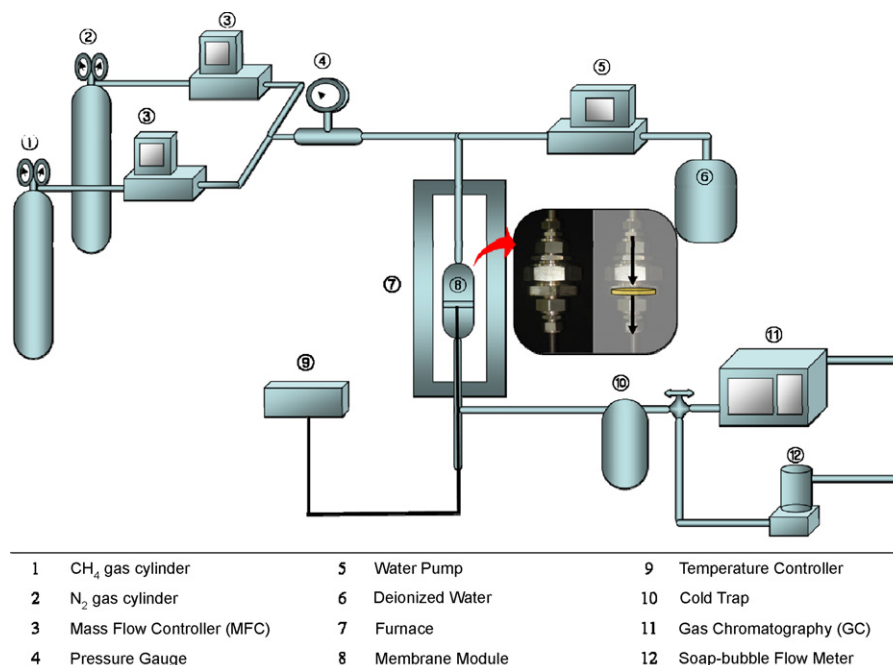


Fig. 2. Schematic of methane steam reforming experimental set-up.

Download English Version:

<https://daneshyari.com/en/article/637048>

Download Persian Version:

<https://daneshyari.com/article/637048>

[Daneshyari.com](https://daneshyari.com)