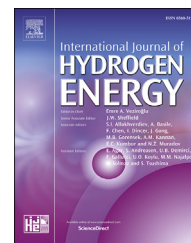


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Modeling of thermally-coupled monolithic membrane reformer for vehicular hydrogen production

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ABSTRACT

A thermally-coupled monolithic membrane reformer (TMMR) is a heat and membrane integrated reformer for portable hydrogen production. This study simulates the TMMR using Aspen Plus software to find the appropriate monolith design and operation parameters to achieve optimal energy efficiency and hydrogen production within a small volume reactor. Different types of fuels, i.e. methane, methanol and ethanol, and various operating conditions, including molar flow rate of fuel (0.01–0.05 mol/s for combustion and 0.1–0.5 mol/s for reforming) and reforming pressure (3–5 atm), were investigated via thermodynamic equilibrium analysis. When methanol and ethanol were used as feedstocks, a reverse water-gas-shift reaction occurred, resulting in a decrease of energy efficiency. While using methane as a feedstock with a specific molar flow rate of 0.03 mol/s for the combustion reaction, 0.30 mol/s for the reforming reaction, and a reforming pressure of 4 atm, significant improvement of efficiency was observed. At the same time, the performance of the TMMR design was simulated based on the surface area of different sizes and configurations of monolith, i.e. parallel and checked arrangements, by using a kinetic-based model approach. The highest efficiency achieved was from the checked arrangement of monolith with 200 cpsi of cell density, 150 mm of diameter and length, and

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exchange area of around 1.5 m². This design requires a small space of only 2.65 L in volume to provide 129 kW of hydrogen energy production with 44.0% efficiency. In conclusion, the proposed design and conditions could be a benchmark for future research on efficient compact reformers for vehicular applications.

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Introduction

Hydrogen is an energy carrier considered as an alternative fuel for renewable energy. Hydrogen fuel can potentially replace fossil fuels in vehicles leading to markedly reduced CO₂ emissions from the traditional combustion engine. The utilization of hydrogen fuel via fuel cells produces electricity for driving the vehicles and the fuel cell technology has been used recently in conventional vehicles [1–4]. As a result, the demand for hydrogen has been increasing significantly, leading to the development of the production and usage. Since the low density of hydrogen leads to an expensive transportation cost of the compressed or liquefied hydrogen fuel, on-site hydrogen production deflates the cost and increases economic feasibility [5–7], leading to an interest of portable hydrogen production using alternative fuels as a hydrogen source. Alternative fuels including methane, methanol, and ethanol have been utilized as feedstocks for hydrogen production in various industries [8,9]. These fuels can be produced from conventional industrial processes and bioprocesses such as waste fermentation, gasification and pyrolysis of biomasses [10–16]. Moreover, methane and methanol can also be produced from recycling of carbon dioxide directly [17–19], resulting in the decrease of CO₂ emissions.

In conventional processes, hydrogen production comprises three main sections, i.e. heat supply, reforming and hydrogen separation. However, the conventional processes are not suitable for a portable system because of the size and weight limitations. For this reason, micro-channel reformers have been considered especially for vehicle applications [20–22]. Micro-channel reformers have several advantages over conventional reformers such as low pressure drop, low mass and heat transfer resistance and fast start-up. Monoliths have been used in several systems [23–27] to serve as the micro-channel structure as it is commercially available. Monoliths can integrate various processes within its structure, which consequently can lead to a compact micro-channel reformer. All the three main units required in the conventional hydrogen production processes can be integrated within the monolith to form a “thermally-coupled monolithic membrane reformer (TMMR)”.

The integrated reformer has been studied separately in thermally-coupled reformers [28–30] and membrane reformers [26,31–38]. With heat being integrated, the diesel reformer in micro-scale combined with a heat exchanger of combustion gases was investigated by O’Connell et al. [28,29]. When operated at 1023 K and a steam to carbon molar ratio of 3.17, 98% of diesel was converted with successful hydrogen production for a 5-kW fuel cell. Zafir and Gavriilidis simulated two channels of thermally-coupled reformers of

methane using numerical methods [30]. It was found that the temperature gradient of the gas phase was 40 K which was significantly lower than those in conventional reformers.

The membrane integrated reformer has been investigated for a couple of decades. Membrane reactors showed remarkably high performance improvement over traditional reactors for hydrogen production [32–40]. The performance of membrane reactors depends on hydrogen separation rate increasing with the operating parameters such as temperature, pressure, sweep gas flow rate, etc. [41–43]. For the palladium membrane fabrication, Kim et al. successfully coated a palladium membrane with 8 μm of thickness within the cordierite monolith by electroless plating [26]. At the optimum operating condition, the selectivity of hydrogen per helium of 360 was achieved. Palladium membrane integrated with a water-gas-shift reactor was also investigated by Karnik et al. [31]. The membrane-supported copper catalyst yielded a hydrogen permeation flux of 5.2 mol m⁻² s⁻¹ when operated at 373 K. Additionally, a Pd-Au membrane was investigated for long term operation of methane steam reforming reaction [44]. Permeate purity of the membrane remained above 99.2% when operated continuously for 530 h. To simulate the membrane reactor, Aspen Plus using sequential modular approach was proposed [39,45,46]. In the study of Ye et al., a fluidized-bed membrane reformer was simulated via this method. Their simulation results were similar to the experimental results at various operating temperatures [39].

In this study, hydrogen production using TMMR was investigated using Aspen Plus with a sequential modular approach. The simulation was divided into two sections: thermodynamic study and TMMR design. In the thermodynamic study, methane, methanol and ethanol were used as feedstocks and the reformer was operated under various operating conditions. The TMMR design was optimized based on the surface area which depends on the monolith configuration, diameter, length, and cell density. The flow arrangement in the TMMR was also explored. The optimal design and operating conditions of TMMR as well as flow arrangement show great promise in terms of energy requirements for vehicular applications. This configuration of TMMR is proposed as a portable reformer and a guideline for TMMR fabrication in further studies.

Modeling and simulation

Model characteristics

For hydrogen utilization in vehicles, proton exchange membrane fuel cell (PEMFC) and recovering system of the process

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