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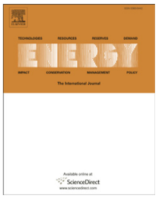

Evaluation of an integrated methane autothermal reforming and high-temperature proton exchange membrane fuel cell system

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ABSTRACT

The aim of this study was to investigate the performance and efficiency of an integrated autothermal reforming and HT-PEMFC (high-temperature proton exchange membrane fuel cell) system fueled by methane. Effect of the inclusion of a CO (carbon monoxide) removal process on the integrated HT-PEMFC system was considered. An increase in the S/C (steam-to-carbon) ratio and the reformer temperature can enhance the hydrogen fraction while the CO formation reduces with increasing S/C ratio. The fuel processor efficiency of the methane autothermal reformer with a WGS (water gas shift reactor) reactor, as the CO removal process, is higher than that without a WGS reactor. A higher fuel processor efficiency can be obtained when the feed of the autothermal reformer is preheated to the reformer temperature. Regarding the cell performance, the reformate gas from the methane reformer operated at $T_{in} = T_R$ and with a high S/C ratio is suitable for the HT-PEMFC system without a WGS reactor. When considering the HT-PEMFC system with a WGS reactor, the CO poisoning has less significant impact on the cell performance and the system can be operated over a broader range to minimize the required total active area. A WGS reactor is necessary for the methane autothermal reforming and HT-PEMFC integrated system with regard to the system efficiency.

1. Introduction

Due to environmental problems and the energy crisis, many studies have aimed to develop both effective and clean technologies. Fuel cells are one of the most attractive power generation technologies that do not release pollution. Compared to other types of fuel cells, the PEMFC (proton exchange membrane fuel cell) is suitable for portable and automotive application as well as small scale power generation, such as residential application, due to its low weight, volume and operating temperature, which provide for the shortest start-up time [1]. On-site and on-board hydrogen productions are the best choices during the development of hydrogen infrastructure and storage.

Currently, natural gas is the most common fuel employed for hydrogen production because of its cost effectiveness for industrial hydrogen. Natural gas primarily consists of methane, as well as nitrogen, CO_2, ethane, propane, butane, pentane and traces of other components. The high hydrogen-to-carbon ratio of methane results in a product with a high hydrogen concentration. The steam reforming of methane reportedly yields the highest hydrogen content of 75–78 vol.% [2]. When synthesis gas with a higher hydrogen concentration is fed to fuel cells, the efficiency of the fuel cell system is improved because the hydrogen fraction directly affects the performance of the fuel cell.

Many studies have focused on fuel processors integrated with PEMFC systems [2–5]. Eszos et al. [6] concluded that steam reforming and autothermal reforming are the most competitive fuel processing options in terms of fuel processing efficiencies and heat integration within the PEMFC systems to provide a high PEMFC system efficiency. Ouzounidou et al. [7] studied a combined methanol autothermal steam reforming and PEM fuel cell system in order to develop a model for exploring the interactions of the integrated system. In addition, Salemme et al. [8] investigated a steam reforming and autothermal reforming integrated with PEMFC system

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with various types of fuel. The methane steam reforming-based system achieved the highest efficiency. Furthermore, Specchia et al. [9] revealed that the efficiency of a steam reforming integrated with a PEMFC system was higher than that of an autothermal integrated system. However, steam reforming systems are more complex in term of heat integration, which impacts the system start-up time. The very good dynamic response of the autothermal reforming process makes it suitable for mobile applications [10,11]. Given the required fast start up and size and weight limitation for automobiles, the ATR (autothermal reforming) has an initially higher reaction rate than the steam reforming and can be operated without an external heat-supplying unit.

Because the CO (carbon monoxide) tolerance of the PEMFC is low, sophisticated CO removal units are needed to reduce the amount of CO (carbon monoxide) to less than 10 ppm. Recently, a HT-PEMFC (high-temperature proton exchange membrane fuel cell) was developed as a promising technology to handle the CO poisoning problem found in a conventional PEMFC [12,13]. When operating at a high temperature, the quantity of CO that adsorbs to the Pt catalyst in the HT-PEMFC decreases, and thus, the CO removal units of the HT-PEMFC system are less complex than those of a conventional PEMFC system [14]. Presently, the HT-PEMFC integrated with reforming processes has been widely reported. Gardemann et al. [15] studied the compact ethanol autothermal processor integrated with the HT-PEMFC system for small scale power generation. The advantages of such system are its compactness and startup reliability. It was found that heating of the shift reactor to remove CO in the fuel processor is the most time-consuming step. Wichert et al. [16] developed the LPG fuel processor integrated with an HT-PEMFC system. The results showed that main reactors of the systems can be operated with stable performance and were easy to control during long-term experiments. Arsalis et al. [17] investigated a 1 kW_e HT-PEMFC integrated with a fuel processor consisting of a methane steam reformer and a water gas shift reactor for Danish single-family households. The waste heat was applied for space heating and producing hot water. The obtained electrical efficiency of the partial (25%) and full loads was 45.4% and 38.8%, respectively. As the CO removal unit is related to the fuel processing process and the HT-PEMFC performance, the effect of its inclusion on the HT-PEMFC system efficiency should be clearly analyzed.

The aim of this study was to analyze and design an integrated autothermal reforming and HT-PEMFC system fueled by methane. To investigate the effect of a CO removal unit, two HT-PEMFC systems are considered in this study. The first system consists of the HT-PEMFC and methane autothermal reformer without the CO removal process. The second involves a HT-PEMFC whose fuel processor consists of a methane autothermal reformer and a WGS (water gas shift reactor) to remove CO. The mathematical models of the HT-PEMFC are based on the electrochemical model and the diffusion model of the gas diffusion layer and electrolyte film layer. The effects of the operating parameters, such as the reformer temperature, inlet temperature and S/C (steam-to-carbon) ratio on the reformer efficiency, hydrogen yield and CO content are analyzed. The operating parameters considerably affect not only the system efficiency but also the complexity of the system. The optimal operating conditions of the autothermal reformer that provide a suitable product gas for HT-PEMFC are given. The efficiency and performance of the HT-PEMFC systems were analyzed by considering the required total active area of the HT-PEMFC of each designed HT-PEMFC system.

2. System description

The autothermal reforming was selected as a hydrogen production process because of its advantageous fast start up and low energy requirement. In addition, the autothermal reforming process can be operated without an external heat supply, and this condition is referred to as "the adiabatic condition". Two HT-PEMFC systems are considered here. The first involves a HT-PEMFC and a methane autothermal reformer without a CO removal process, as shown in Fig. 1. To enhance the hydrogen concentration and improve the overall system efficiency, water gas shift reactors were added to an integrated HT-PEMFC system, as presented in Fig. 2. The methane and oxidants, which are air and water, are fed to the methane autothermal reformer when the air feed content is controlled to supply the required heat of reforming via an oxidation reaction. Therefore, a burner for supplying heat to the reforming process was not added to this system. The inlet temperature of the autothermal reformer feed stream was set at the reaction temperature and standard temperature (298 K). The recovery heat from the HT-PEMFC is used to preheat the reactant of the autothermal reformer when the inlet temperature of the autothermal reformer feed stream is equal to the reaction temperature, as shown in Figs. 1(b) and 2(b). In contrast, the recovery heat from HT-PEMFC is not necessary when the inlet temperature is specified at 298 K, as shown in Figs. 1(a) and 2(a). The target power output of the HT-PEMFC systems is 50 kW for a small vehicle.

3. Modeling of HT-PEMFC system

3.1. Fuel processor

The equilibrium composition of the reformate gas obtained from the methane autothermal reforming was calculated by directly minimizing the Gibbs free energy, as shown in Eq. (1). For the methane autothermal reforming process, the gaseous components in the system are CH_4, H_2, CO, CO_2, O_2, N_2 and H_2O.

$$\min_{n_i} (G^t)_{T,P} = \sum_{i=1}^{C} n_i \bar{G}_i = \sum_{i=1}^{C} n_i \left(G_i^{\circ} + RT \ln \frac{\bar{f}_i}{f_i^{\circ}} \right) \tag{1}$$

where C is the total number of components in the reaction system, and n_i is the moles of each gaseous component. Regarding the

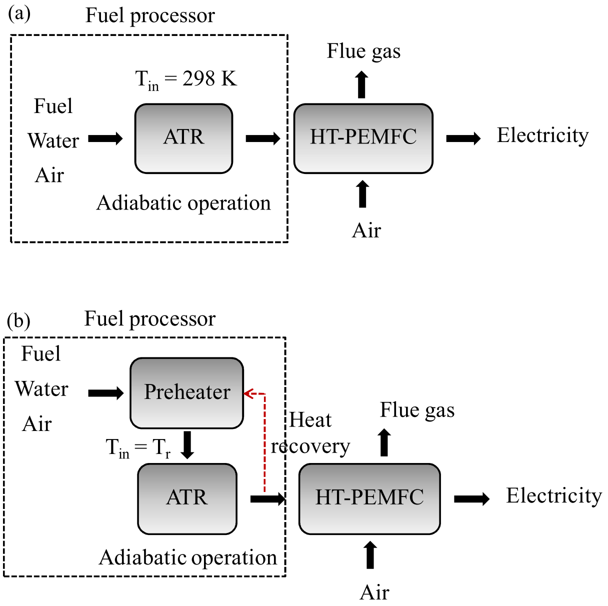

Fig. 1. Autothermal reforming integrated with HT-PEMFC system without a water gas shift reactor (case 1): (a) T_{in} = 298 K and (b) $T_{in} = T_r$.