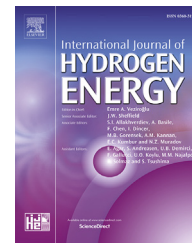




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Experimental investigation of CO tolerance in high temperature PEM fuel cells

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ABSTRACT

In the present work, the effect of operating a high temperature proton exchange membrane fuel cell (HT-PEMFC) with different reactant gases has been investigated throughout performance tests. Also, the effects of temperature on the performance of a HT-PEMFC were analyzed at varying temperatures, ranging from 140 °C to 200 °C. Increasing the operating temperature of the cell increases the performance of the HT-PEMFC. The optimum operating temperature was determined to be 160 °C due to the deformations occurring in the cell components at high working temperatures. To investigate the effects of CO on the performance of HT-PEMFC, the CO concentration ranged from 1 to 5 vol %. The current density at 0.6 V decreases from 0.33 A/cm² for H₂ to 0.31 A/cm² for H₂ containing 1 vol % CO, to 0.29 A/cm² for 3 vol % CO, and 0.25 A/cm² for 5 vol % CO, respectively. The experimental results show that the presence of 25 vol % CO₂ or N₂ has only a dilution effect and therefore, there is a minor impact on the HT-PEMFC performance. However, the addition of CO to H₂/N₂ or H₂/CO₂ mixtures increased the performance loss. After long-term performance test for 500 h, the observed voltage drop at constant current density was obtained as ~14.8% for H₂/CO₂/CO (75/22/3) mixture. The overall results suggest that the anode side gas mixture with up to 5 vol % CO can be supplied to the HT-PEMFC stack directly from the reformer.

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Introduction

Proton exchange membrane fuel cell (PEMFC) is considered as a promising clean energy conversion technology due to the high efficiency, high power density, low CO₂ emissions and fast start-up. PEMFC is classified according to working temperature as low temperature and high temperature PEMFC. Typical low temperature (LT-PEMFC) PEMFC using per-fluorosulfonic acid membrane and the working temperature is limited up to 70–80 °C [1]. Operational challenges faced by LT-

PEMFC include delicate water management and low tolerances to impurities. In addition, low operating temperature causes mass transfer limitations as it causes liquid water to accumulate at high current densities. Increasing the PEMFC working temperature above 100 °C can increase the tolerance to impurities and overcome the difficulties in water management [2,3]. High temperature PEMFC (HT-PEMFC) is gaining more attention of PEMFC research groups due to their ability to work without moisture, better fuel impurities tolerance, fast reaction kinetics and combined heat and power generation systems (CHP) applications [4–6].

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PEMFC also need to deal with the drawbacks of the cost and durability for commercialization. Because of its very low tolerance towards impurities in the fuel, the LT-PEMFC requires pure H₂ or proper fine cleaning steps after the reformer to operate reformat gases. However, the use of LT-PEMFC is limited due to the high cost of producing high purity H₂ gas or additional purification systems added to system [7,8]. Today, most of the H₂ production is produced by natural gas or methane vapor reforming. The HT-PEMFC systems can be used with reformer systems due to the high CO and impurity tolerance [9,10]. The use of H₂ produced by inexpensive industrial processes such as the steam reforming of natural gas with steam will facilitate the commercialization of PEMFCs and make it easier for them to enter daily life [11]. However, as a result of steam reforming, impurities such as CO₂, CO, CH₄ and N₂ and NH₃ can be present in the produced H₂ rich reformat. Generally, steam reforming is used to produce H₂-rich reformat gas which contains 40–70% H₂, 15 to 25 vol % CO₂ and 1 to 3 vol % CO and a small amount of H₂O vapor and N₂. A typical CO content in reformat gases is mainly dependent on the type of thermochemical process, fuel, catalyst, temperature and pressure. Generally, CO content could be varied from 1 to 5% in 230–350 °C and up to 10% for H₂ production in the industry [12]. It is important to know the effects of these impurities on PEMFC performance for the design and production of fuel cell based power generation systems [13,14].

Due to the major effect on HT-PEMFC performance at low temperatures CO is the most studied impurity in the literature [15–17]. CO competes with H₂ for the adsorption regions of the Pt catalyst and blocks the catalytic active sites because Pt is more absorptive on the catalyst surface. As a result, even a very small amount of CO content may prevent H₂ from effectively reaching the Pt catalyst surface. The effect of CO poisoning on the anode side could be immensely reduced by HT-PEMFC operations because the kinetics of CO desorption from the Pt catalyst particles is favored with the increasing temperature [18,19]. Although HT-PEMFCs show superior CO tolerance, the combined effects of CO poisoning and hydrogen dilution could deteriorate the performance and durability of HT-PEMFCs.

Many researchers have investigated CO poisoning effect on HT-PEMFC with different working conditions. Different approaches have been applied, such as changing the parameters such as CO concentration, catalyst structure [20] and operating temperature [21] to reduce the adverse effects of CO use on PEMFC [22]. Reduction of CO poisoning by increasing the working temperature is one of the most used methods among these approaches. Li et al. investigated the CO effect at a temperature range of 125–200 °C and determined that the CO tolerance increased with increasing temperature [23]. However, increasing the working temperature can damage the cell components because of the electrochemical potential, high water partial pressure and increased carbon corrosion. Therefore, it is necessary to determine the optimum temperature at which the highest performance can be achieved [24]. The effects of CO and CO₂ on the HT-PEMFC performance have been extensively reviewed in the literature. However, there are limited studies on the dual effect of CO₂ and CO on HT-PEMFC. Andreasen et al. investigated HT-PEMFC

performance with CO and CO₂ using electrochemical impedance spectroscopy [25]. The HT-PEMFC performance was examined subject to some of the poisoning effects experienced when running on a reformat gas.

In this study, poisoning effects of impurities in the anode gas stream on HT-PEMFCs were investigated at various concentrations of H₂, CO₂, N₂ and CO. Firstly, the operating temperature at which the highest performance was achieved without damaging the cell components was determined by performing HT-PEMFC tests at the range of 140–200 °C. Then to simulate poisoning environment, the HT-PEMFC was fed with different concentrations of CO (1–5 vol %) in H₂ gas stream in the same conditions as H₂. Additionally, we investigated the effects of CO₂ and N₂ on HT-PEMFC performances. There is also a limited number of studies on the effects of the anode-side reformat gas feeding on the long-term durability of HT-PEMFC. Long term durability test of HT-PEMFC was performed with H₂/CO₂/CO reformat gas mixture with 75/22/3 vol ratio. The results of this research in the future could be the basis for the researcher to design and control HT-PEMFC systems.

Experimental

Materials

The PBI membrane was prepared by a solution casting method according to our previous study [26]. *N*-*N* dimethylacetamide (CH₃C(O)N(CH₃)₂ DMAc), polyvinyl difluoride (PVDF) and phosphoric acid (H₃PO₄, 85 wt %) were used as received. The commercially available Gas Diffusion Layer (GDL, Freudenberg-H2315C2) was obtained from Freudenberg (Germany). The Pt supported carbon black (Pt/C, 70 wt % of Pt) catalyst was purchased from Tanaka Kikinok. All gases having high purity (>99.98%) were purchased from Linde. Millipore water was used wherever required. All chemicals and solvents were high-grade reagents and used without further purification.

Method

The acid doping was carried out by immersing the PBI membranes in H₃PO₄ at room temperature for at least 1 week to maintain proton conductivity. Generally, PBI polymer is a non-conductive aromatic heterocyclic polymer in pure form. When PBI is doped with H₃PO₄, it shows high proton conductivity. The active sites in PBI imidazole rings interact with the H₃PO₄ and 2 mol of H₃PO₄ is attached per PBI unit. Fig. 1 shows the proton conduction mechanism in PBI membrane network. Generally, high H₃PO₄ doping levels increase the PBI proton conductivity but excess of H₃PO₄ in the PBI might affect the mechanical properties of the PBI membrane [27]. The doping level acquired by the PBI membrane was 11 H₃PO₄ per polymer repeat unit.

For the preparation of the MEA, non-woven carbon cloth coated with a carbon microporous layer was used as the GDL. The Pt/C catalyst was used as a both anode and cathode catalyst with 1.0 mg/cm². During long-term operation of H₃PO₄ doped PBI membranes often lose proton conductivity

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