

Effect of flow rate, flow direction, and silica addition on the performance of membrane and the corrosion behavior of Pt–Ru/C catalyst in PEMFC



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ABSTRACT

In order to evaluate the performance of Proton Exchange Membrane Fuel Cell (PEMFC) with an active area of 25 cm², several variables were studied (flow pattern, flow rate, degradation in the Pt–Ru/C catalyst). Polarization curves revealed that the electrochemical reaction on both sides of the fuel cell is under the effect of mass transfer and the values of the cell voltage, current density, and power density were inversely proportional with the increase of the hydrogen and oxygen flow rates for both flow patterns (counter current and cocurrent) and clear corrosion phenomenon was observed on the surface of the Pt–Ru/C catalyst before and after exposure to the hydrogen and oxygen. On the other hand and in order to increase the performance of Nafion membrane, SiO₂ particles were introduced to the Nafion polymeric matrix using sol–gel method to form composites. The surface morphology of the Nafion–SiO₂ composite membrane was investigated and compared with the existing commercial Nafion membrane. It was found that water uptake of the Nafion–silica composite membrane as a function of temperature is higher than that of the existing commercial Nafion membrane.

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1. Introduction

In a typical PEM fuel cells, the membrane is sandwiched between two catalyzed electrodes to transport the protons, support the anode and cathode catalyst layers, and more importantly, separate the oxidizing (air) and reducing (hydrogen) environments on the cathode and anode sides, respectively. Therefore, the requirements for an excellent membrane are manifold and stringent, including high protonic conductivity, gas permeability, thermal and chemical stability, and so on. The most commonly used and promising membranes for PEM fuel cells are perfluorosulfonic acid (PFSA) membranes such as Nafion[®] (DupontTM). To prevent mechanical failure of the membrane, the Membrane Electrode Assembly (MEA) and flow field structure must be carefully designed to avoid local drying of the membrane, especially at the reactant inlet area. The membranes developed so far can be classified into three groups: (1) modified Nafion membranes, which are swelled with nonvolatile solvents or incorporate hydrophilic oxides and solid inorganic proton conductors; (2) alternative sulfonated polymers and their composite membranes, such as SPSF, SPEEK, PBI, and PVDF; and (3) acid–base polymer membranes, such as phosphoric acid-doped Nafion[®]–PBI composite membranes.

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With respect to the chemical and electrochemical degradation of the membrane, developing membranes that are chemically stable against peroxy radicals has drawn particular attention, Wu et al. [1].

Sridhar et al. [2], mentioned that low humidification greatly enhances membrane degradation during operation as well as under OCV. This behavior seems peculiar, because gas permeation through a Nafion membrane decreases with decreasing gas humidification. Basu [3], showed that Nafion membranes thickness ranges between (25 and 250 μm). Buchi [4], emphasized that corrosion of carbon supports may cause the electrical isolation and aggregation of platinum nanoparticles, causing a decrease in the ECA in the catalyst layers. Zhang [5], mentioned that the Gas Diffusion Layer (GDL) together with flow fields and current collectors, are designed to achieve high performance from the operation of the PEM fuel cell. Basu et al. [6], showed that Pt/Ru(40%:20% by wt.)/C and Pt-black could be used as a catalyst to prepare anode and cathode and carbon paper is used as substrate for the catalyst powder. Chul et al. [7], showed that Ti-felt with different structural properties (porosity and fiber diameter) and PTFE content were prepared for use as GDLs of the oxygen electrode, and the relation between the properties of the GDL and the fuel cell performance was examined for both fuel cell and electrolysis operation modes.

Hongtan and Andrew [8] showed that the difference in local electrical resistance under the land and channel is large enough to be a major cause for the observed local current density

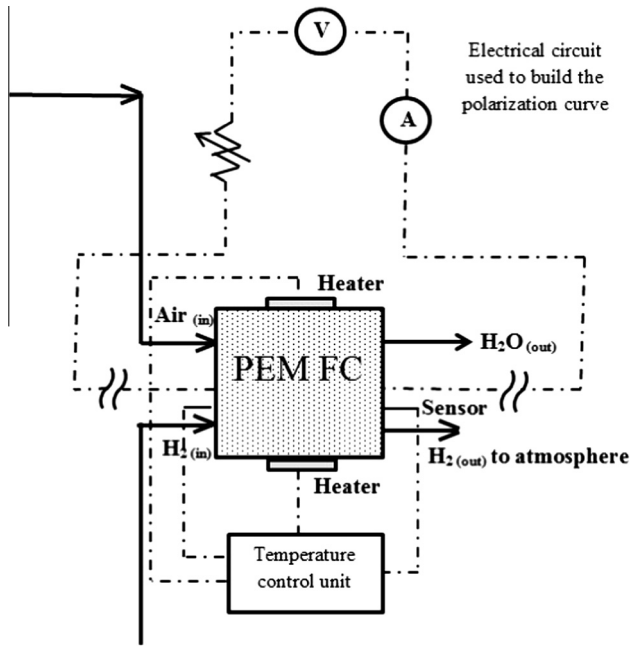


Fig. 1. PEM fuel cell setup.

differences. Kap-Seung et al. [9], studied the geometrical characterization of the serpentine flow-field in order to enhance the performance of PEMFC in relation to pressure drop, discharge of condensed water, maximization of cell voltage, and uniformity of current density over the entire surface area. Luis et al. [10], studied the influence of the relative entry positions of hydrogen and oxygen on the distribution of gases. Santarelli and Torchio [11] studied the effect of six operating conditions on the performance of single PEMFC: cell temperature; anode flow temperature in saturation and dry conditions; cathode flow temperature in saturation and dry conditions; and the reactants pressure while Hsieh et al. [12] studied the effect of cell temperature, gas humidification, cell operating pressure and reactant gas flow rate with interdigitated flow fields. On the other hand Miaomiao et al. [13], studied the water uptake and swelling ratio, thermal and chemical stability, mechanical properties, proton conductivity. Jin et al. [14] studied the polarization curves of a single PEMFC having a Nafion membrane fed with H₂/O₂ with relative humidity (RH) of 35%, 70% and 100% at cell temperatures ranging from 65 °C to 120 °C at back pressure of 0 atm and 1 atm, respectively. Afshari and Jazayeri [15] developed a fully numerical model of two dimensional, non-isothermal, electrochemical-transport to investigate simultaneous water, heat

transport phenomena and their effects on PEM cell performance. Tian et al. [16] studied the carbon loading, PTFE content and species, sintering time, temperature and pore formers.

It is worth to mention that the current work is considered novel in comparison with the revised literature in terms of the studied parameters which play a vital role in the performance of the PEM fuel. In other words the work is based on studying the functionality of Nafion membrane and the corrosion behavior of the Pt–Ru/C catalyst due to hydrogen and oxygen as a function of the flow rates (5, 10 and 15 ml/min) and its directions (i.e. Counter and Cocurrent) in serpentine flow field at constant temperature (80 °C) in addition to studying the surface morphology and water uptake of the casted [Nafion, Nafion–SiO₂ composite] in comparison with the commercial membranes at various temperatures (i.e. 20, 35, 50, 65, 80 °C).

2. Experimental

2.1. Materials

The catalysts used to prepare the electrodes (cathode and anode) were Pt–Ru (40%, 20% by wt.)/C. Carbon black and carbon paper were used as a substrate for the mentioned catalyst. Nafion solution was used as a binder. Hydrogen with purity of 99.999% was used as a fuel. Air was supplied to the fuel cell as source of oxygen. Nafion dispersion was used to cast the proton exchange membrane in addition to commercial Nafion membranes. Sulphuric acid was used for activating both casted and commercial membranes. Isopropanol was used as diluent. Silicon dioxide was used to improve the water uptake of the casted Nafion membrane.

2.2. Membrane preparation

The Nafion [perfluorosulfonic acid]–SiO₂ membrane was casted from Nafion dispersion containing 4% wt. Nafion ionomer using sol-gel method, Alvarez et al. [17]. The isopropanol and Nafion dispersion were mixed in a 1:3 volume ratio, and then 1 mg of SiO₂ was added to selected samples. These samples were kept in an oven for 7 h at 100 °C until all solvent was evaporated and the polymeric ionomer forms a solid polymer membrane known as Nafion–SiO₂ composite membrane. Then the formulated membrane film was treated for 1 h in 1 M H₂SO₄ at 80 °C. Finally, it was rinsed in boiling distilled water for 1 h. The thickness of the resulted membrane was 103.2 μm.

2.3. Preparation of anode and cathode

Electrode of PEMFC should be porous to enable hydrogen and oxygen from air to diffuse through the anode and cathode active

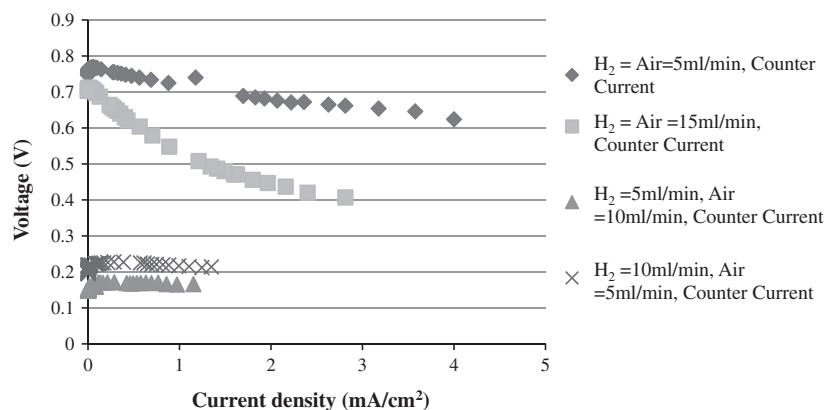


Fig. 2. PEMFC performance (operating conditions: counter current flow, 80 °C).

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