Journal of Power Sources 74 (1998) 146–150

Short communication

A study of the internal humidification of an integrated PEMFC stack

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Received 8 December 1997; accepted 24 January 1998

Abstract

An integrated proton exchange membrane fuel–cell (PEMFC) system has been developed with an internal humidifier within the stack. Research is concentrated on selecting a membrane with low cost and good water permeability because, to date, high-cost membranes (e.g., as Nafion) have been used. The gas and water permeability of several membranes were measured. A low-cost ultra filtration (UF) membrane shows better characteristics for the internal humidifier and cell performance than the others. Also, saturated water vapour permeating through the UF membrane can be supplied at the stack from the internal humidifier. The internal humidifier using UF membrane is thought to be a satisfactory humidifier for a PEMPC. © 1998 Elsevier Science S.A. All rights reserved.

Keywords: PEMFC; Internal humidification; Ultrafiltration membrane

1. Introduction

As a power generator for transportation, the proton exchange membrane fuel–cell (PEMFC) has many advantages in comparison with other fuel cells [1]. By using a solid polymer electrolyte, it is virtually uninfluenced by corrosion and can be operated at low temperature. Also, it has high power density. At present, many efforts are being made to develop power generator for electric vehicles.

The polymer electrolyte requires a supply of water to maintain proton conductivity. Water management is essential for the performance enhancement of a PEMFC stack because proton conductivity depends strongly on the hydration of the polymer [2–4]. Indeed, water must be supplied continuously to prevent drying of the membrane. In most cases, water supply has been achieved by passing both the anode and the cathode gas through external humidifiers [5]; cooling water was also supplied. Two external humidifiers and the cooling system are divided in two parts. This is an unfavourable design as it adds to the complexity of the fuel cell system due to its complexity.

To solve this problem, Nakaoka and Yoshida [6] developed an internal humidification method that employed a porous plate at the channel of cooling water. Staschewski [7] has also attempted internal humidifying. We have developed a system with a humidifier within the stack, as proposed by Ballard Power Systems, which is the world leader in such techniques. The main point of the idea is to humidify the gases by using cooling water flowing in the stack, without an external humidifier. This system, which is termed an ‘integrated stack’, has the internal humidifier using the membrane in the fuel–cell stack.

In this paper, the permeation characteristics of several membranes, used for the internal humidifier and the performance of the stack using each membrane are compared. Also, the humidification effect of the internal humidifier is investigated.

2. Experimental

The integrated fuel–cell was composed of a fuel cell stack and an internal humidifier, as shown in Fig. 1. One cooling plate is placed in the cells and a separator (a PTFE plate) is used to separate the internal humidifier from the cells. The stack has six holes for the inlet and outlet of hydrogen, oxygen and cooling water. The area of each cell is 50 cm^2. Most of previous works (e.g., Ballard systems), has employed thermoplastic gaskets for gas sealing, but the outstanding feature of this new designed stack is to use O-type rings instead of gaskets. The use of O-type rings makes it easier to assemble and disassemble the cells. Water is supplied in the stack through the water flow

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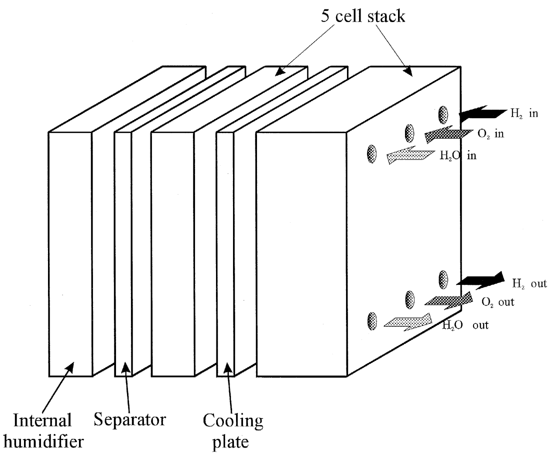

Fig. 1. Schematic of fuel cell stack with internal humidifier.

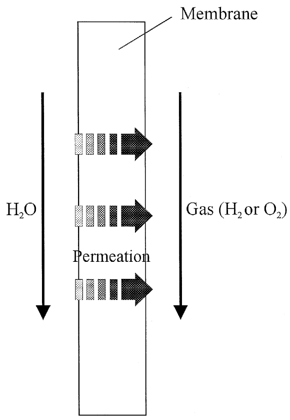

Fig. 2. Structure of internal humidifier.

channel, and serves to provide both humidification and cooling. The cooling of the stack is carried out at the water-flow channel and the cooling plate. Fig. 2 shows a schematic diagram of the structure of the internal humidifier. Water and reaction gases flow on opposite sides of the membrane and water permeates through the membrane. The reaction gases are humidified by passing through the internal humidifier.

The electrode and polymer electrolyte were an E-TEK electrode (0.4 mg Pt cm^{-2}) and Nafion 115 (127 μm, Dupont), respectively. The M&E assemblies were made by hot pressing at 120°C and 2 tons for 5 min.

The membrane used for the internal humidifier were Nafion 115, an ultrafiltration (UF) membrane and a reverse osmosis (RO) membrane. The membranes were classified by pore size. Nafion is a non-porous membrane, and UF and RO are asymmetric ones. Also, Nafion 115 is a little thinner than the others. The properties of each membrane are shown in Table 1.

2.1. Gas permeation test

To compare the properties of the membranes, their permeation ability was measured. The permeability of the membrane has an important effect on the performance of the internal humidifier. If the gas-permeation flux is too large, it may cause a cross-over in the stack. On the other hand, a large water permeation flux increases the conductivity of the electrolyte and enhances the cell performance. The permeation test was performed in a cell with the diameter of 3 cm over a pressure range of 1 to 3 atm and a temperature of 70°C. The membranes were prepared in both a dry and a wet state. The permeation gases were pure hydrogen and pure oxygen.

2.2. Performance test

Hydrogen and oxygen were provided to the cell through a mass-flow controller (Model 8274, Matheson) from a storage tank. Water for cooling and humidification was supplied and circulated by a small pump. The current and voltage of the stack were measured by a d.c. electronic load (HP-6050A, Hewlett Packard) connected to an IBM-PC.

3. Results and discussion

The variation with pressure of the permeation flux of hydrogen gas passing through dried UF and RO membranes is shown in Fig. 3. The hydrogen permeation flux of the UF membrane is much greater than that of the RO membrane, due to the larger pore size, and is proportional

Table 1
Membrane properties

	Nafion 115	Ultrafiltration (UF) membrane	Reverse osmosis (RO) membrane
Type	Perfluorinated sulfonic	Polysulfone	Polyamide composite
Thickness (μm)	127	160	160
Pore size	Non-porous	< 1 μm	< 10 Å
	(Ionic cluster size: 5 Å)	(MWCO: 35,000)	
Cost (US$)	320[a]	4[b]	6[b]
Company	Du Pont	UOP	UOP

[a] Per ft^2.
[b] Per ft × yd.

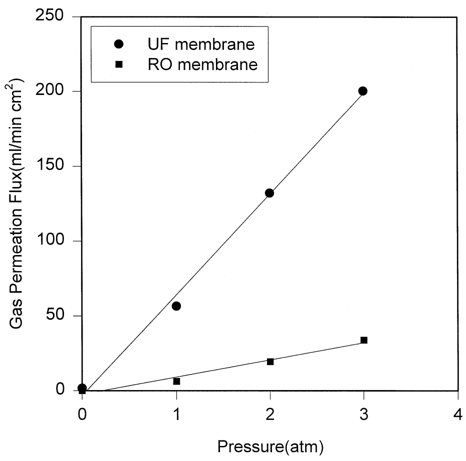

Fig. 3. Hydrogen permeation flux through dried UF and RO membranes with pressure.

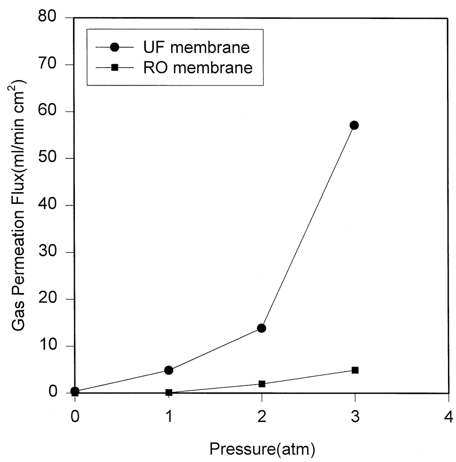

Fig. 5. Oxygen permeation flux through dried UF and RO membranes with pressure.

to the increase in pressure. The hydrogen permeation flux of the UF membrane is so large that it may cause a cross-over in the stack. The permeation flux of the fully wetted UF or RO membranes is decreased markedly compared with that of the dried counterparts, as shown in Fig. 4. This is because if the membrane is wetted fully, water molecules occupy most of the pores of the membrane and gas permeation is disturbed by tension between the water molecules. In fact, as the membrane for the internal humidifier is used in a fully wetted state, the flux of the wetted membrane is more important than that of the dried membrane. In this test, Nafion was not investigated because it is non-porous and its gas permeation flux is extremely small [8].

The variation with pressure of the permeation flux of oxygen gas passing through dried UF and RO membranes is shown in Fig. 5. The permeation flux of oxygen is less than that of hydrogen because molecules of an oxygen are larger than those of hydrogen. And with hydrogen, the oxygen flux increases with pressure. The permeation flux of oxygen gas passing through wetted UF and RO membranes is shown in Fig. 6. In both cases, the flux is diminished considerably, compared with that for the corresponding dried membrane.

The membrane for the internal humidifier must have a minimum gas permeation flux because gas permeation causes a cross-over in the fuel–cell stack. From the above results, it is concluded that the gas permeation flux through UF and RO membranes is so negligibly small in the wetted state that it will not cause a cross-over.

The water permeation flux of each membrane as a function of pressure is shown in Fig. 7. The fluxes of the RO membrane and Nafion 115 with a small pore size are much smaller than that of the UF membrane and do not increase much with pressure. In a PEMFC, water is essential for proton conduction in the electrolyte. Therefore, a UF membrane is expected to enhance the cell performance because the increase of water supply in the electrolyte improves the proton conductivity of the electrolyte. In the case of the RO membrane, though the gas permeation flux

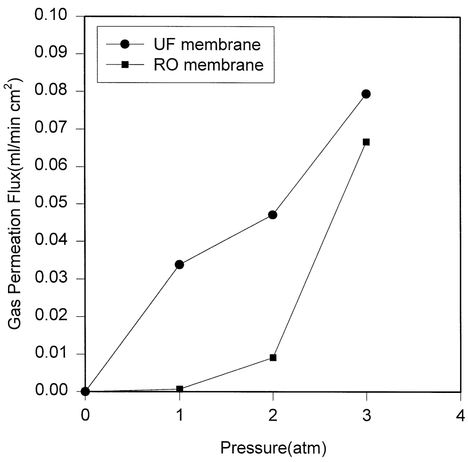

Fig. 4. Hydrogen permeation flux through wetted UF and RO membranes with pressure.

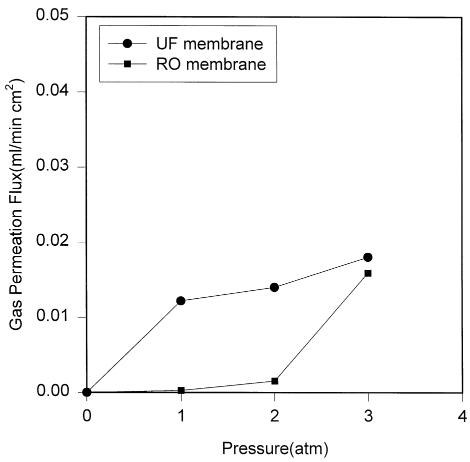

Fig. 6. Oxygen permeation flux through wetted UF and RO membranes with pressure.

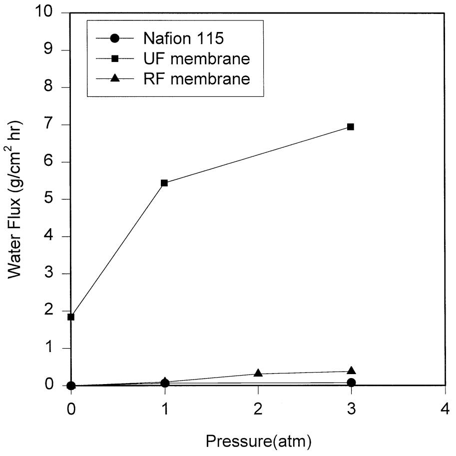

Fig. 7. Comparison of water flux through Nation 115, UF and RO membranes with pressure.

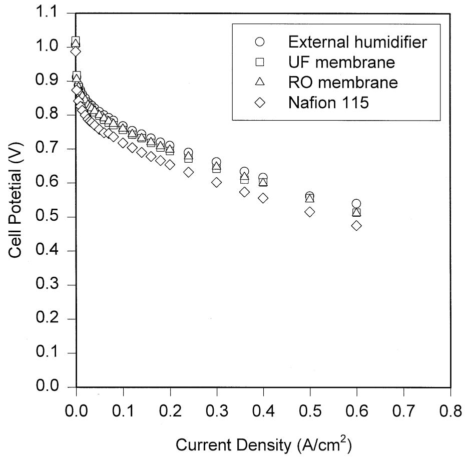

Fig. 9. Comparison of cell performance for a stack with an external humidifier or with an internal humidifier using Nafion 115, UF, RO membranes at 50°C.

is small, water permeation flux is too small. Thus, the RO membrane is not suitable for the internal humidifier.

The variation of water flux through the UF membrane with temperature reveals an interesting result, as presented in Fig. 8. It displays temperature behaviour that is similar to a saturated steam pressure curve. This implies that the water through the UF membrane is in a vapour state rather than in a liquid one. Such water vapour is not expected to hinder the passage of reaction gases to the active sites of the catalyst. Therefore, an internal humidifier using a UF membrane is expected to operate in a satisfactorily manner.

The performance of the five-cell stack with an internal humidifier using each membrane is presented in Fig. 9. The performance with an internal humidifier is no better than that of a stack with an external humidifier, but the rapid decrease of cell performance due to cross-over does not occur. With an internal humidifier, the UF membrane is better than Nafion 115. This is because the water permeation flux of Nafion 115 is too small to provide sufficient water to the cells. This suggests the possibility for employing a UF membrane in the internal humidifier, instead of Nafion 115 which has a high cost. On the other hand, the RO membrane displays almost the same performance as the UF membrane in spite of its low water permeation flux. As shown in Fig. 10, however, when the stack is operated at a high current density, the cell voltage drops sharply. It is considered that the RO membrane and Nafion 115 cannot provide adequate water for the electrode reaction at the operating condition of high current density, and thus the voltage decreases. Therefore, the RO membrane and Nafion 115 that have a poor water permeation flux have a lower stability than the UF membrane at high current density.

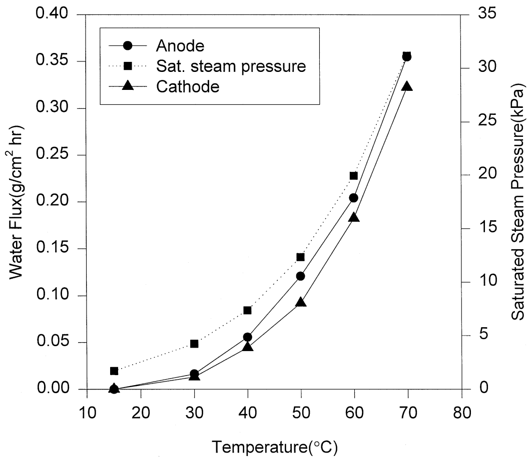

Fig. 8. Comparison of water fluxes through UF membrane and saturated steam pressure with cell temperature at anode and cathode (Area: 50 cm^2, H_2 flow rate: 400 ml min^{-1}).

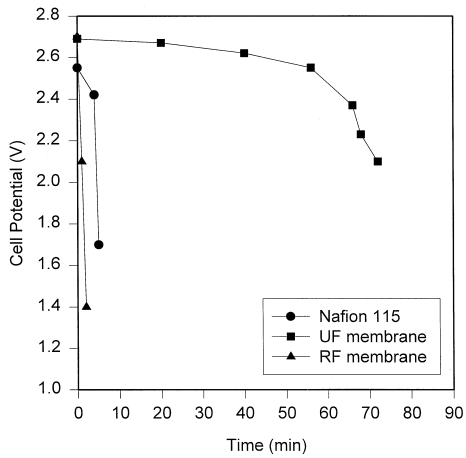

Fig. 10. Cell potential drop with time after applying constant high current density (600 mA cm^{-2}) to a five-cell stack.

4. Conclusions

A PEMFC system with a humidifier within the stack, termed as ‘integrated stack’, has been developed. As present membranes (e.g., Nafion) have high cost and poor water permeability, an attempt has been made to find an alternative membrane with low cost and good water permeability. It is found that there are no major difference in cell performance for a stack using an internal humidifier or an external one. A low-cost, UF membrane exhibits better characteristics for the internal humidifier and cell performance than the other membranes. Also, saturated water vapour permeating through UF membrane can be supplied at the stack from the internal humidifier.

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