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Performance of air-breathing direct methanol fuel cell with anion-exchange membrane

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ARTICLE INFO

Article history:

Received 22 July 2009

Received in revised form

19 September 2009

Accepted 28 October 2009

Available online 22 November 2009

Keywords:

Anion-exchange membrane

Direct methanol fuel cell

Air-breathing

Alkaline fuel cells

ABSTRACT

This report details development of an air-breathing direct methanol alkaline fuel cell with an anion-exchange membrane. The commercially available anion-exchange membrane used in the fuel cell was first electrochemically characterized by measuring its ionic conductivity, and showed a promising result of $1.0 \times 10^{-1} \text{ S cm}^{-1}$ in a 5 M KOH solution. A laboratory-scale direct methanol fuel cell using the alkaline membrane was then assembled to demonstrate the feasibility of the system. A high open-circuit voltage of 700 mV was obtained for the air-breathing alkaline membrane direct methanol fuel cell (AMDMFC), a result about 100 mV higher than that obtained for the air-breathing DMFC using a proton exchange membrane. Polarization measurement revealed that the power densities for the AMDMFC are strongly dependent on the methanol concentration and reach a maximum value of 12.8 mW cm^{-2} at 0.3 V with a 7 M methanol concentration. A durability test for the air-breathing AMDMFC was performed in chronoamperometry mode (0.3 V), and the decay rate was approximately $0.056 \text{ mA cm}^{-2} \text{ h}^{-1}$ over 160 h of operation. The cell area resistance for the air-breathing AMDMFC was around $1.3 \Omega \text{ cm}^2$ in the open-circuit voltage (OCV) mode and then is stably supported around $0.8 \Omega \text{ cm}^2$ in constant voltage (0.3 V) mode.

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1. Introduction

The Proton Exchange Membrane Fuel Cell (PEMFC) is now widely regarded as a suitable system for a range of low temperature power generation applications. PEMFCs that use an acidic membrane as a solid electrolyte may be deployed as power sources in electronic appliances (e.g., cellular phones, camcorders, and notebook computers). Among competing PEMFC designs, the direct methanol fuel cell (DMFC), which directly converts the chemical energy of methanol into electrical energy, is one of the most promising candidates for man-portable power sources. Methanol has a high theoretical energy storage capacity of 5019 Ah kg^{-1} and its liquid form

allows for more convenient storage and replenishment than other fuels. The Nafion[®] protonic membrane is a popular solid electrolyte for use in DMFCs. Despite these advantages, the development of DMFCs has been hindered by several problems: (1) slow electrode kinetics, (2) CO poisoning of the Pt catalyst at low temperature, (3) methanol crossover and (4) the high cost of the components such as noble catalysts, the membrane and so on.

It is well known that electro-oxidation of methanol in alkaline media is kinetically faster than the same process in acidic media [1–4]. Fig. 1 shows a schematic diagram of a direct methanol alkaline fuel cell (DMAFC). The electrochemical reaction of a DMAFC is as follows:

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doi:10.1016/j.ijhydene.2009.10.100

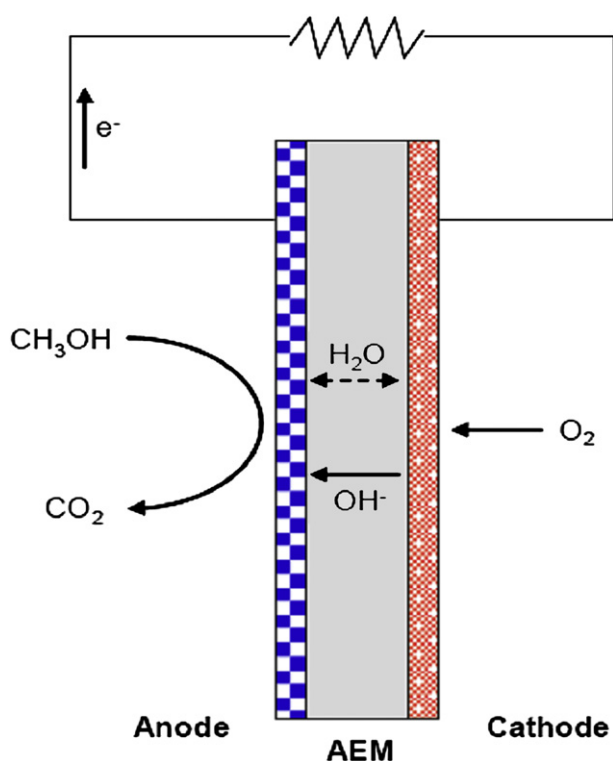


Fig. 1 – Schematic diagram of electrochemical reaction in AMDMFC.

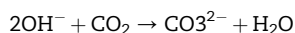
Anode : $\text{CH}_3\text{OH} + 6\text{OH}^- \rightarrow \text{CO}_2 + 5\text{H}_2\text{O} + 6\text{e}^-$ $E_a^\circ = -0.81 \text{ V}$ (vs. NHE)

Cathode : $2/3\text{O}_2 + 3\text{H}_2\text{O} \rightarrow 6\text{OH}^-$ $E_c^\circ = 0.402 \text{ V}$ (vs. NHE)

Overall : $\text{CH}_3\text{OH} + 2/3\text{O}_2 \rightarrow \text{CO}_2 + 2\text{H}_2\text{O}$ $E^\circ = 1.21 \text{ V}$

One of the advantages of alkaline fuel cells is the use of non-precious metals such as silver catalysts [5] and perovskite type oxides [6], as these catalysts are inexpensive and tolerant to methanol crossover. In addition, the ion transport within the membrane in an operating DMAFC moves from the cathode to the anode, which is opposite that in proton conducting systems as shown in Fig. 1. As such, the direction of the electro-osmotic drag is reversed, reducing the methanol crossover. If the methanol crossover could be eliminated, it would be feasible to use undiluted methanol for high energy density, and it would also be possible to use thinner membranes with lower resistance, thus further improving the cell performance. As a result, DMFCs that use alkaline medium as electrolyte have been recently considered as an alternative approach.

One problem with alkaline fuel cells is the formation of carbonate in the alkaline aqueous solution due to CO_2 from air or the oxidation product of the



This reaction has the effect of reducing the number of hydroxyl ions available for reaction at the electrodes. This

conversion from hydroxyl to carbonate also reduces the ionic conductivity of the electrolyte solution. If the advantages of alkaline media in a fuel cell are to be made use of, alkaline membranes should be used instead of alkaline aqueous solutions. Indeed, a number of studies on alkaline membrane direct methanol fuel cells (AMDMFCs) have been reported [7–12]. Wang et al. elucidated the thermodynamic disadvantages and kinetic advantages of the AMDMFC with theoretically calculated results and experimental data [13]. Yu et al. explored the feasibility of applying an anion membrane in DMFCs and performed an extensive investigation of the electrochemical performance of the membrane electrode assembly (MEA) in a DMFC [14]. Matsuoka et al. reported the demonstration of AMDMFCs at low temperature (323 K) and concluded that ethylene glycol showed the best performance among the tested alcohols for use as an anodic fuel.

In this study, we examine the feasibility of an air-breathing AMDMFC under ambient conditions and investigate the effect of methanol concentration on cell performance. Air-breathing fuel cells using an anion-exchange membrane (AEM) have rarely been reported in the literature. A fuel cell designed to breathe air without the necessity for ancillary devices such as air pumps or fans can offer several advantages, including the opportunity to eliminate parasitic power loss and to use simpler structures and more compact system designs than are possible with active DMFCs. In turn, these advantages can potentially result in higher reliability, lower cost, higher fuel utilization, and higher energy density.

2. Experimental procedure

2.1. Pretreatment of OH-form anion membrane

The anion-exchange membrane and ionomer for an AMDMFC are key technologies for such cells to achieve practical performance as fuel cells. For an anion-exchange membrane, our test used an A-010 ion exchange membrane (OH^- type, TOKUYAMA Corp.) that is a strongly basic anion permeable membrane with a thickness of 41 μm . The ionic form of the anionic membrane as shipped was a Cl^- form, which was then changed to an OH^- form as follows. The Cl^- form membrane was rinsed several times with Millipore water, and then immersed in 500 ml of 1 M potassium hydroxide (KOH) solution at 313 K for 2 h and then at 298 K for 24 h to exchange Cl^- with OH^- . The membrane was then rinsed and stored in Millipore water for later use.

The potentiometric method was applied to measure the membrane ionic conductivity and the anionic membrane was cut into $2 \text{ cm}^2 \times 2 \text{ cm}^2$ samples [11]. The experimental cell used to measure ionic conductivity was composed of two compartments, and each one had two openings to use one Pt electrode and one Hg–HgO reference electrode. Water tightness between each compartment and the membrane was secured using two gaskets. The KOH solution concentration in the experimental cell was varied between 1 M and 10 M. The membrane ionic conductivity was determined by two successive measurements of the potential difference between the reference electrodes, the first without a membrane ($R = R_{\text{KOH}}$), and the second with a membrane between the two

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