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Electrochemical behaviour of a hydrogen/oxygen gas mixture at a solid polymer electrolyte interface

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The electrochemical behaviour of a hydrogen/oxygen gas mixture at platinum electrodes in contact with a solid polymer electrolyte (Nafion® 115) was studied in two modes: hydrogen enrichment and fuel cell. In the hydrogen enrichment (purification) mode, i/V(V being the applied voltage) characteristics were identified for the electrochemical cell arranged so that the gas mixture with varying ratios which contacts the anode was purified to pure hydrogen gas that was released at the cathode. In the fuel cell mode, V/i (V being the terminal voltage) characteristics were determined for the cell which utilized the gas mixture with varying ratio at the anode and pure oxygen at the cathode. The effect of the decrease in the mole fraction of hydrogen (χ_{H_2}) on the current density or the terminal voltage was particularly severe for $\chi_{H_2} < 0.6$. The drop was analysed employing terms such as oxygen reduction current, oxygen adsorption and the change of e.m.f. (electromotive force) due to the decrease in the partial pressure of the hydrogen gas.

Keywords: *energy conversion*, *hydrogen purification*, *hydrogen/oxygen gas mixture*, *PEMFC*, *solid polymer electrolyte*

1. Introduction

Solid polymer membrane fuel cells (either SPEFC or PEMFC (proton exchange membrane fuel cells)) have recently emerged as promising candidates for transportation applications [1–4] because of their high power density and simplicity of fabrication when compared to other types of fuel cells [5]. Supplying hydrogen fuel for fuel cells for transportation applications requires an on-demand generation of fuel gas due to a weight penalty for using stored fuel. Methods which are emerging for on-board generation of hydrogen gas include the reformation of fossil fuels [6] or iron sponge–water reaction [7]. In the first case impurities such as carbon monoxide in the reformed gas must be removed before they can be used in fuel cells.

Hydrogen gas can also be generated from water decomposition using external energy or metals with high oxidation potential. Hydrogen can be generated through electrical discharge using a metal/water system [8]; generation by this method accompanies oxygen coproduction. It is also known that radiolysis of water by nuclear energy generates a mixture of hydrogen and oxygen [9, p. 62]. The device allowing direct electrochemical utilization of such a gas mixture was considered by Justi *et al.* in 1956 [9, p. 374]. In their device, the radiolytically generated oxyhydrogen gas first contacts the cathode, which is active to oxygen, and then passes to the anode where separated hydrogen is oxidized. The effect of oxygen on the performance of hydrogen fuel at the anode which was in contact with the polymer electrolyte was not reported.

In addition to the case described above, a H_2/O_2 gas mixture could exist at the oxygen electrode of PEMFC system through various kinds of crossovers. A thin membrane like Nafion® 112 can cause a significant crossover of hydrogen through the membrane electrolyte to the oxygen electrode [10]. Such crossovers have been reported on other membrane electrolytes [11–14]. Another kind of crossover can be observed when a thin membrane electrolyte at a high current density develops hot-spots which convert to pinholes, eventually causing cell failure. This cell failure has become a less serious matter in recent days due to the advent of more efficient membranes such as Aciplex-S® 1104 and Dow 800.

Regardless of whether the circumstances which arise from mixing gases are intentional or accidental, information about electrochemical performance of gas mixture is valuable. It provides the basis for evaluating the power reduction of the cell utilizing the gas mixture. This can also be used for predicting cell malfunctions originating from accidental gas mixing. Electrochemical purification of the gas mixture has no practical value as a prestep for providing the pure hydrogen gas to the cell. However, information about the process provides not only the basis for evaluating its energy efficiency, but also an indirect basis for evaluating fuel cell performance. Our research was aimed at investigating the performance of the

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PEMFC utilizing H_2/O_2 gas mixtures of various ratios. The electrochemical information was obtained on two types of cell arranged in the H_2/O_2 fuel cell or in the gas enrichment mode.

2. Influence of oxygen additions to hydrogen

Application of oxyhydrogen, the stoichiometric hydrogen/oxygen gas mixture obtained from radiolysis, to a fuel cell was investigated intensively in the 1950s [9, p. 62]. The gas mixture was first taken to an oxygen electrode, consisting of a material active to oxygen such as active carbon or gold, then a major portion of the oxygen gas was selectively reduced at the electrode. This study also included the establishment of potentials at bright or platinized platinum electrode in acidic electrolyte saturated with H_2/O_2 mixtures of varying compositions. The potential behaviour is shown schematically in Fig. 1. When a platinized electrode contacts liquid electrolyte saturated with the gas mixture, the electrode acts as either the hydrogen electrode:

$$H_2 \rightarrow 2\,H^+ + 2\,e^- \qquad \text{for} \quad \chi_{H_2} > 0.67$$

or the oxygen electrode

$$O_2 + 4\,H^+ + 4\,e^- \rightarrow 2\,H_2O \quad \text{for} \quad \chi_{H_2} < 0.67$$

where χ_{H_2} denotes the mole fraction of hydrogen in the gas mixture. When bright platinum is used as the electrode instead of platinized platinum, a hydrogen/oxygen mixed potential is established for the entire mole fraction range of hydrogen. This phenomenon is ascribed to the slower surface reaction of the $H_2 + \frac{1}{2}O_2 \rightarrow H_2O$ process compared to the diffusion rate. Therefore, when such a gas mixture is used as the fuel at the anode of the fuel cell, the electrode contacting the gas mixture performs in various modes depending on the physical and chemical nature of the surface and the electrolyte. Unless the electrode catalyses the gas mixture selectively, either component of the mixture is dependently or independently consumed at the electrode, as exemplified through oxygen reduction at the anode.

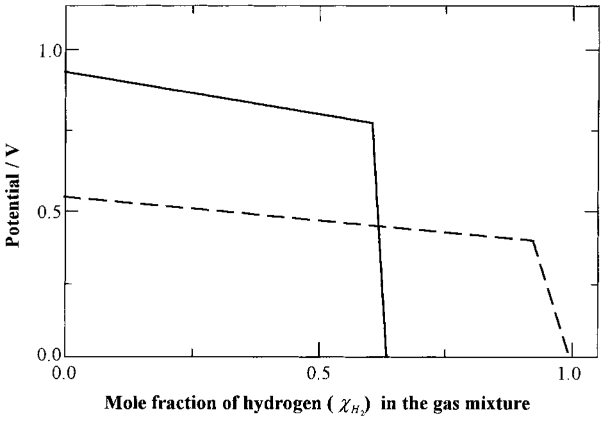

Fig. 1. Electrode potential of Pt electrode in sulfuric acid saturated with various H_2/O_2 gas mixtures: (——) platinized electrode, (- - - -) bright electrode [9]. The curves which are approximations of the actual ones shown in ref. 9 illustrate the difference between the electrodes.

It has not been reported that the above phenomenon also applies to fuel cell systems using solid polymer electrolytes. In the interface between the fuel cell anode and the solid electrolyte, chemical conditions of the solid electrolyte (such as proton activity or water content) should play a significant role in determining the potential and, thus, the electrochemical behaviour. In this study, we arranged a cell configuration in the fuel cell mode as well as in the hydrogen purifying mode. The fuel cell mode used the gas mixture at the anode side. In the purifying device, the gas mixture supplied at the anode was transformed into pure hydrogen released at the cathode as shown:

$$H_2 \rightarrow 2\,H^+ + 2\,e^- \qquad \text{at the anode}$$

$$2\,H^+ + 2\,e^- \rightarrow H_2 \qquad \text{at the cathode}$$

The schematics of the purifying arrangement and the fuel cell are shown in Fig. 2(a) and (b), respectively.

In principle, the anodic reaction mechanisms should be identical, regardless of the operational mode. In both modes, the anodic behaviour as a function of χ_{H_2} is the focal point of this investigation. When a gas mixture containing χ_{H_2} is used at the anode, a fraction of the hydrogen oxidation current is nullified by the reduction of oxygen at the anode. Oxygen can also be adsorbed on the platinum catalyst surface. This oxygen admixture at the anode may interfere with the hydrogen adsorption, causing overvoltage. Thus, the decrease in current density as χ_{H_2} decreases at a fixed applied voltage in the purifying cell may be largely attributed to oxygen reduction as well as oxygen interference with the hydrogen adsorption. This type of current decrease is defined as Δi_{OR}. Another source of current reduction is the change in e.m.f. when χ_{H_2} drops below 1.0. We call this a thermodynamic current reduction, Δi_{TH}. Thus, the total current reduction, Δi_M, due to oxygen addition is the sum of Δi_{OR} and Δi_{TH}. Such a reduction also lowers the power of the fuel cell at a given load resistance. The current reduction in the purifying device is a direct indication that the amount of purified hydrogen at a given applied voltage, V_{appl} decreases. By measuring current reduction in the purifying cell or the terminal voltage drop in the fuel cell, we can evaluate the practical value of the purifying device or the fuel cell. From such an evaluation we can predict any cell malfunctions caused by accidental gas mixing.

Hydrogen/oxygen gas easily explodes in a water-free environment if an electric spark or a catalytic surface is available. In a hydrogen–oxygen–water vapour system, water vapour significantly reduces the probability of the occurrence of an explosive hydrogen/oxygen reaction [15]. Since the gas mixture studied in this investigation was saturated with water vapour, the danger of the explosion was minimal.

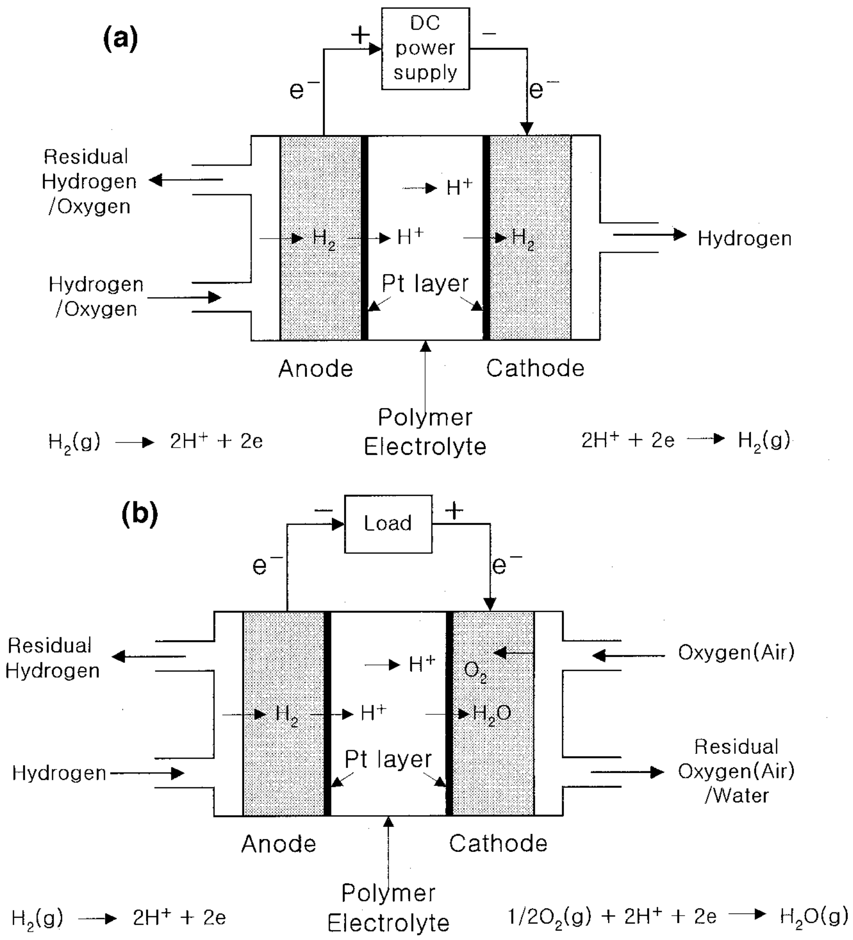

Fig. 2. (a) Schematic of a hydrogen separation device. (b) Schematic of PEMFCs (proton exchange membrane fuel cells).

3. Experimental details

Nafion® 115 and 117 were used as the proton exchange membrane. The membrane was prepared as follows. It was boiled in 3 wt % H_2O_2 solution for 1 h and then rinsed in boiling deionized water for 2 h to remove any organic compounds. It was then boiled in 0.5 M H_2SO_4 for 1 h and finally rinsed in boiling deionized water for 2 h to remove metal compounds. Commercial carbon gas diffusion electrodes (0.4 mg Pt cm^{-2}, Globe Tech. Co.) were used as both the cathode and the anode. To increase the three-dimensional contact areas between the solid electrolyte and the catalytic platinum layer, the platinum surface was impregnated with 0.8 mg cm^{-2} Nafion® (Aldrich Chemical Company). The electrodes were dried for 2 h to remove any residue solvents like isopropyl alcohol and water. The pretreated membrane and electrodes were bonded by heating and pressing at 120 °C and 70 atm for 90 s.

The electrochemical cell was assembled by securing Viton gaskets (or Teflon gaskets), carbon block and a copper current collector to both sides of the membrane and electrode assembly using nuts and bolts. The gas mixture which was prepared in a variety of mixing ratios, with the help of mass flow controllers (SAM SFC280E), was analysed by gas chromatography (Hewlett–Packard 5890), then fed to the anode through a humidifier which had been insulated and heated. Pure hydrogen or oxygen gas was fed to the cathode through the humidifier for the purifying or fuel cell experiments, respectively.

Finally, the line connecting the humidifiers to both the anode and cathode was heated and insulated by heating bands. The temperature of the electrochemical cell and humidifiers at the cathode and anode were set at 60, 65 and 70 °C, respectively. The pressure of the gas mixture supplied to the anode was monitored using a pressure senor (Valcom). The potential between the anode and cathode was adjusted by a d.c. power supply to allow the flow of a specific current. In all experiments, the total flow rate of the gas mixture was 300 ml min^{-1} while the pressure of the mixture was 1 atm.

4. Results and discussion

4.1. *Purifying mode*

The current in the purifying cell represents the amount of hydrogen separated. The percentage purity of the separated hydrogen gas was nearly 100% when tested using gas chromatography. Figure 3 shows current density against applied voltage (i/V_{appl}) of the purifying device using Nafion® 115. The current at a given applied voltage drops sharply as χ_{H_2} drops below 0.6. As discussed in Section 2, the electrode contacting the gas mixture could function as the oxygen electrode as χ_{H_2} falls below the stoichiometric ratio. The cell then develops an electromotive force exceeding the applied voltage from the external power source, thus preventing positive current flow through the cell. As seen in the curve for $\chi_{H_2} = 0.4$ of Fig. 3, the e.m.f. of the cell exceeds the applied voltage by such a large margin that the current reversal requires a much higher applied voltage. When χ_{H_2} exceeds 0.8, the voltage necessary to induce about 268 mA cm^{-2} of current density is 0.1 V. Figure 3 indicates that the trends in the i/V curves using Nafion® 115 and 117 are similar, showing that membrane thickness is not a prime factor in the electrochemical performance.

Figure 4 shows i/V_{appl} curves when nitrogen, instead of oxygen, is mixed with hydrogen gas in varying ratios. A monotonic reduction of current density at a given voltage with increase in the nitrogen mole fraction implies that the hydrogen partial pressure is the only factor in determining the current density. In this case, the reduction in current with the decrease of χ_{H_2} is solely attributable to the change in e.m.f. of hydrogen oxidation, since nitrogen does not interfere with the electrochemical reaction of hydrogen, that is, playing no role in current reduction. (The change in the e.m.f. means a shift of the reference point for the overpotential.) The thermodynamic current reduction, Δi_{TH}, which is solely responsible for the total current reduction is given by

$$\Delta i_{\text{TH}} = i(\chi_{H_2} = 1) - i_{\text{M}}(\chi_{H_2} \text{ in } H_2/N_2) \qquad (1)$$

where $i_{\text{M}}(\chi_{H_2})$ is the current density at an applied voltage when χ_{H_2} of the H_2/N_2 mixture is lower than 1.0.

From the characteristic values of the voltage and current shown in Figs 3 and 4, we can express the current reduction, Δi_{M}, with decrease in χ_{H_2} in the H_2/O_2 gas mixture, in terms of Δi_{OR} and Δi_{TH} as

$$\Delta i_{\text{M}} = \Delta i_{\text{OR}} + \Delta i_{\text{TH}} \qquad (2)$$

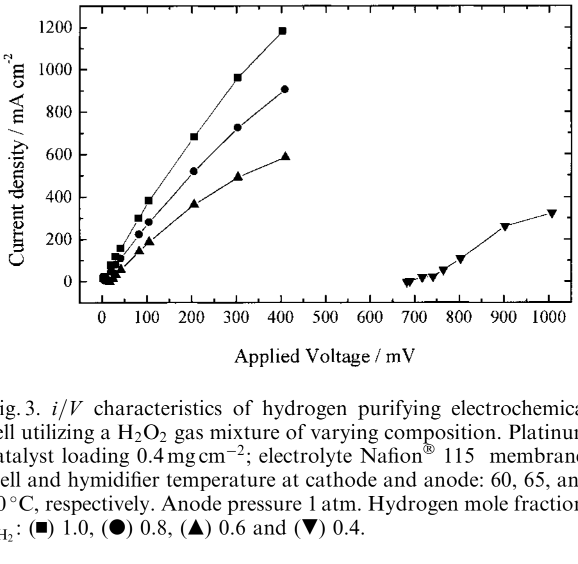

Fig. 3. i/V characteristics of hydrogen purifying electrochemical cell utilizing a H_2O_2 gas mixture of varying composition. Platinum catalyst loading 0.4 mg cm^{-2}; electrolyte Nafion® 115 membrane. Cell and hymidifier temperature at cathode and anode: 60, 65, and 70 °C, respectively. Anode pressure 1 atm. Hydrogen mole fraction, χ_{H_2}: (■) 1.0, (●) 0.8, (▲) 0.6 and (▼) 0.4.

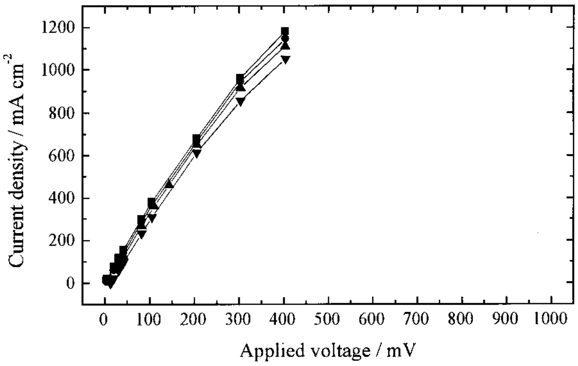

Fig. 4. i/V characteristics of purifying cell utilizing a H_2/N_2 gas mixture of varying composition. Experimental conditions as for Fig. 3. Hydrogen mole fraction χ_{H_2}: (■) 1.0, (●) 0.8, (▲) 0.6 and (▼) 0.4.

where Δi_{OR} is the current reduction due to oxygen involvement at the anode. The current reduction Δi_{M} at a given applied voltage is given by

$$\Delta i_{\text{M}} = i(\chi_{H_2} = 1) - i_{\text{M}}(\chi_{H_2} \text{ in } H_2/O_2) \qquad (3)$$

where $i_{\text{M}}(\chi_{H_2})$ is the current density at the applied voltage when χ_{H_2} of the H_2/O_2 mixture is lower than 1.0. Since Δi_{M} and Δi_{TH} are easily measured from Figs 3 and 4, Δi_{OR} is obtained from Equation 2. These values are given in Table 1. Table 1 shows that Δi_{OR} becomes the major contributor to Δi_{M} at a higher applied voltage. In other words, as the voltage increases, the overall interference of oxygen, mainly in the form of an oxygen reduction current, is the major contributor to current drop. As the mole fraction of oxygen increases twice from $\chi_{H_2} = 0.8$ to $\chi_{H_2} = 0.6, \Delta i_{\text{TH}}$, also increases by a factor of two. This result is consistent with the definition of Δi_{TH}.

The reduction in current density of the cell implies a reduction in purification efficiency. This is defined as

Reduction in efficiency

$$= \frac{\text{Power required for purification of the mixture of } \chi_{H_2}}{\text{Power required for purification of pure } H_2}$$

$$= \frac{V_{\text{appl}} \times [i(\chi_{H_2} = 1.0) - \Delta i_{\text{M}}]}{V_{\text{appl}} \times i(\chi_{H_2} = 1.0)}$$

$$= \frac{i(\chi_{H_2} = 1.0) - \Delta i_{\text{M}}}{i(\chi_{H_2} = 1.0)} \qquad (4)$$

The reduction in efficiency at several V_{appl} values is given in Table 2.

4.2. *Fuel cell mode*

Figure 5 shows the terminal voltage against current density (V_{terminal}/i) of the fuel cell as it consumes the H_2/O_2 gas mixture at the anode and pure oxygen at the cathode. The trend in the V/i curves shows similar behaviour to that in the purifying device in that, in both cases, the curve for $\chi_{H_2} = 0.4$ is strikingly isolated from the rest. When χ_{H_2} is 0.4, the electrode contacting the H_2/O_2 gas mixture no longer utilizes

Table 1. Reduction in current density (Δi_M) of hydrogen purifying device as a function of applied voltage and mole fraction of hydrogen

Δi_{OR} and Δi_{TH} denote the reduction in current density due to oxygen involvement and due to thermodynamic origin, respectively

V_{appl} /V	$\chi_{H_2} = 1$	$\chi_{H_2} = 0.8$			$\chi_{H_2} = 0.6$		
	i/mA cm^{-2}	Δi_M	Δi_{OR}	Δi_{TH}	Δi_M	Δi_{OR}	Δi_{TH}
0.006	22	17	4	13	–	–	–
0.02	77	32	16	16	73	42	31
0.031	117	38	22	16	86	54	32
0.042	156	47	31	16	100	68	32
0.082	299	79	64	15	158	124	34
0.105	383	102	83	19	195	157	38
0.205	680	162	147	15	316	286	30
0.303	961	236	219	17	471	426	45
0.403	1180	286	249	37	600	531	69

Table 2. Reduction in efficiency of the hydrogen purifying device as a function of applied voltage and hydrogen mole fraction

Reduction units are as percentages

V_{appl}/V	$\chi_{H_2} = 1.0$	$\chi_{H_2} = 0.8$	$\chi_{H_2} = 0.6$
0.006	0	77	–
0.02	0	42	95
0.031	0	32	74
0.042	0	30	64
0.082	0	26	53
0.105	0	27	51
0.205	0	24	46
0.303	0	25	49
0.403	0	24	51

the hydrogen component since the electrode becomes an oxygen electrode.

It is possible to assemble V_{terminal}/i curves of the fuel cell from the i/V_{appl} curves of the purifying device. To do this, we require the V/i curve of the fuel cell using pure H_2. The V_{terminal} of the fuel cell using pure hydrogen is given by

$$V_{\text{terminal}}(\chi_{H_2} = 1.0) = V_{OC} - (\eta_{O_2} + \eta_{R_e}) \qquad (5)$$

where V_{OC}, η_{O_2} and η_{R_e} denote the open cell voltage, the overvoltage of the oxygen electrode (cathode) and the ohmic drop across the membrane electrolyte iR_e, respectively, assuming that the anode overpotential is negligible. The ohmic drop should be close to the applied voltage required to the reaction of the purifying device using pure hydrogen. Since V_{OC} represents the terminal voltage at $i = 0$, η_{O_2} of Equation 5 can be computed as a function of i. Now the terminal voltage of the fuel cell using the H_2/O_2 gas mixture is given by

$$V_{\text{terminal}}(\chi_{H_2}) = V_{OC} - (\eta_{O_2} + \eta_{R_e} + \eta_A) \qquad (6)$$

where η_A denotes the overvoltage of the anode in contact with the gas mixture of χ_{H_2}. The sum of η_{R_e} and η_A when current i flows through a load is the same as the applied voltage of the purifying device generating current i, both arrangements utilizing gas of the same χ_{H_2}. Thus, the assembled V_{terminal} of χ_{H_2} is given by

$$V_{\text{terminal}}(\chi_{H_2}) = V_{\text{terminal}}(\chi_{H_2} = 1.0) + V_{\text{appl}}(\chi_{H_2} = 1.0) - V_{\text{appl}}(\chi_{H_2}) \qquad (7)$$

The V/i curve of the fuel cell using $\chi_{H_2} = 0.8$ is assembled using Equation 7. When compared to the experimental result seen in Fig. 6, the assembled curve has a significantly higher current density than the measured one, implying that the anodic behaviour of the purifying device may be different from that of the fuel cell at higher current densities. The origin of such a deviation will be the subject of future investigations.

Here the power of the fuel cell utilizing a gas mixture of χ_{H_2} is compared to the cell using pure hydrogen, while assuming the load resistance R is the same. Now, the power ratio is defined by

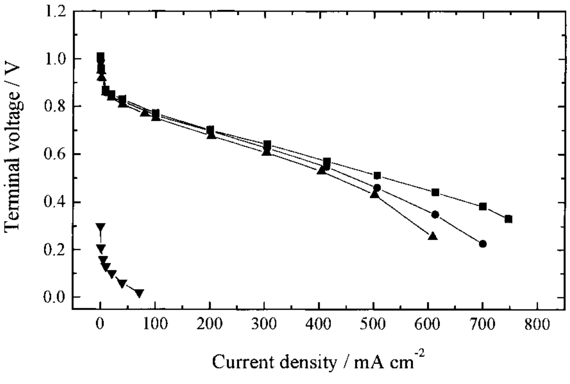

Fig. 5. V_{terminal}/i characteristics of fuel cell utilizing the H_2/O_2 gas mixture at anode and pure O_2 at cathode. Other conditions are the same as those in Fig. 3. Hydrogen mole fraction χ_{H_2}: (■) 1.0, (●) 0.8, (▲) 0.6 and (▼) 0.4.

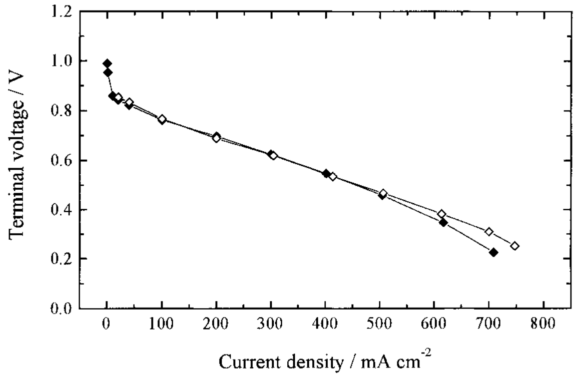

Fig. 6. Assembled (open symbol) and measured (solid symbol) V/i curves of the fuel cell utilizing the gas mixture of $\chi_{H_2} = 0.8$.

Table 3. Power ratio of fuel cell at various levels of load resistance

χ_{H_2}	*Load resistance*/Ω cm^2				
	0.44	0.72	1.38	3.50	20.75
1	1.00	1.00	1.00	1.00	1.00
0.8	0.79	0.85	0.93	0.99	0.99
0.6	0.65	0.76	0.90	0.98	0.98
0.4	0.01	0.01	0.01	0.02	0.04

Power ratio of the fuel cell

$$= \frac{\text{Power of the cell using the gas mixture of } \chi_{H_2}}{\text{Power of the cell using pure } H_2} = \frac{V_{\text{terminal}}(\chi_{H_2}) \times i(\chi_{H_2})}{V_{\text{terminal}}(\chi_{H_2} = 1.0) \times i(\chi_{H_2} = 1.0)} = \frac{i^2(\chi_{H_2})\, R}{i^2(\chi_{H_2} = 1.0)\, R} = \frac{i^2(\chi_{H_2})}{i^2(\chi_{H_2} = 1.0)} \quad (8)$$

The ratios at several R values are given in Table 3.

When the fuel cell consumes pure hydrogen gas for fuel, its performance is 200 mA cm^{-2}, 0.7 V at 3.5 Ω cm^2 electric load (typically 0.7 V is the target potential of PEMFC). When the cell utilizes a gas mixture of $\chi_{H_2} = 0.8$, the power reduction is only 1% at the same load. However, the power of the fuel cell is reduced by more than 20% when the load is decreased to 0.44 Ω cm^2.

5. Conclusion

The performance of a PEMFC or a hydrogen purifying device utilizing a hydrogen/oxygen gas mixture was evaluated using the V_{terminal}/i or i/V_{appl} characteristics, respectively. For both modes, the functional transformation of the anode from a hydrogen electrode to an oxygen electrode was not as sharp as that manifested in the electrode involving liquid electrolyte saturated with the gas mixture. The efficiency of each mode as determined by the current for hydrogen separation or the terminal voltage was severely reduced as χ_{H_2} became less than 0.6. The coupling of oxygen reduction with hydrogen oxidation at the anode was the prime contribution for the current reduction at a higher applied voltage or a lower χ_{H_2}. The power reduction of the fuel cell using the gas mixture was found to be moderate for the range $\chi_{H_2} = 1.0 \sim 0.6$. The power reduction of the cell using gas of $\chi_{H_2} = 0.8$ was less than 21% for the better portion of the current density.

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