

# The carbon dioxide concentrator by using MCFC

Kimihiko Sugiura<sup>a,\*</sup>, Kenji Takei<sup>a</sup>, Kazumi Tanimoto<sup>b</sup>, Yoshinori Miyazaki<sup>b</sup>

<sup>a</sup>Osaka Prefectural College of Technology, 26-12 Saiwai, Neyagawa, Osaka 572-8572, Japan

<sup>b</sup>National Institute of Advanced Industrial Science and Technology, Kansai,  
1-8-31 Midorigaoka, Ikeda, Osaka 563-8577, Japan

## Abstract

The removal characteristics of CO<sub>2</sub> in molten carbonate fuel cell (MCFC) were elucidated by the single cell whose electrode area was 81 cm<sup>2</sup> and electrolyte was 52Li<sub>2</sub>CO<sub>3</sub>/48Na<sub>2</sub>CO<sub>3</sub>. The experimental value of the CO<sub>2</sub> removal rate from cathode to anode was almost corresponding to the theoretical value. Cell voltage dropped drastically when concentration of CO<sub>2</sub> in cathode became 15% or less. This tendency showed strikingly, as concentration of O<sub>2</sub> in cathode became lower and lower. The cell performance is influenced with the  $p_{O_2}/p_{CO_2}$  ratio of the cathode gases, because cell voltage decreases by decreasing the  $p_{O_2}/p_{CO_2}$  ratio. Therefore, when the  $p_{O_2}/p_{CO_2}$  ratio is as low as the thermal power plant, it is necessary to add air to cathode for improving the cell performance. Consequently, the reduction target of CO<sub>2</sub> in COP3 can be achieved. © 2003 Elsevier Science B.V. All rights reserved.

**Keywords:** MCFC; CO<sub>2</sub> concentrator; Thermal power plant; Electrochemistry

## 1. Introduction

According to the Kyoto protocol (COP3), Japan has to reduce the greenhouse gases by at least 6% below 1990 levels in the commitment period 2008–2012 (the reduction rate is 3.5% in the group of CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O). If the exhaust amount of the greenhouse gases increases by 2% every year, the exhaust amount of CO<sub>2</sub>, which was 314 million tons in 1996, will increase 347 million tons in 2010. Japan has to reduce the group of CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O by 24% in 2010 to achieve the target of COP-3. It is necessary to decrease 8.2% of the CO<sub>2</sub> exhaust amount in 2010, even if the exhaust amount of CO<sub>2</sub> decreases from 1996 every year according to the data of Agency of Natural Resources and Energy [1]. Therefore, Japan should make the further efforts to achieve the target. Though the CO<sub>2</sub> removal with the de-carbonizing technology by chemical absorption method have been evaluated in the thermal power plant, the problems on the cost, etc. remain.

In molten carbonate fuel cell (MCFC), CO<sub>2</sub> moves from cathode to anode in the form of the carbonate ion as an ion conductor. Therefore, MCFC can be used as the CO<sub>2</sub> removal device. The CO<sub>2</sub> removal device using MCFC (CRDM) can be classified into two modes of operation as shown in Fig. 1. One of the CRDM operation modes is the decomposition mode as shown in Fig. 1a. In this case, CO<sub>2</sub> and O<sub>2</sub> are moved to the anode side to supply the mixed gas,

which contains CO<sub>2</sub> and O<sub>2</sub> to the cathode, and to supply external energy. Therefore, the composition of anode exhaust gas is 66.7CO<sub>2</sub>/33.3O<sub>2</sub> [2]. The other CRDM operation mode is a method of using the power generation reaction of MCFC as shown in Fig. 1b [3]. In this case, the CO<sub>2</sub> purity is controlled by fuel gas utilization. For example, the CO<sub>2</sub> purity is 100%, if the fuel gas utilization is set to 100%. In addition, the energy conversion efficiency of the thermal power plant with this CRDM is higher than the former CRDM, because the electric power supply is not necessary for this CRDM. However, CO<sub>2</sub> concentration in exhaust gas from the thermal power plant is much too low in 4–8% to use exhaust gas as a cathode gas of MCFC. Therefore, it is impossible to forecast the performance of this CRDM from the current MCFC performance data.

From such a background, this paper aims to decrease the amount of CO<sub>2</sub> exhausted to the atmosphere by using exhaust gas from the thermal power plant as a cathode gas of MCFC. Therefore, the influence of CO<sub>2</sub> concentration on the cell performance was examined, and the feasibility study on the operating condition and the scale of MCFC when exhaust gas from the thermal power plant was used as a cathode gas for MCFC was done.

## 2. Experimental apparatus and procedure

Fig. 2 shows a schematic diagram of the experimental apparatus. The fuel cell is installed in the electric furnace to

\* Corresponding author. Tel.: +81-72-820-8537; fax: +81-72-821-0134.  
E-mail address: [sugiura@sys.osaka-pct.ac.jp](mailto:sugiura@sys.osaka-pct.ac.jp) (K. Sugiura).

**Nomenclature**

<i>a</i>	constant ( $\Omega \text{ cm}^2 \text{ atm}^{0.25} \text{ K}^{-1}$ )
<i>a'</i>	constant ( $\Omega \text{ cm}^2 \text{ atm}^{-0.5} \text{ K}^{-1}$ )
<i>A</i>	constant ( $\Omega \text{ cm}^2 \text{ atm}^{0.25}$ )
<i>b, b'</i>	constant ( $\Omega \text{ cm}^2 \text{ atm K}^{-1}$ )
<i>B</i>	constant ( $\Omega \text{ cm}^2$ )
<i>C</i>	constant
<i>E</i>	open-circuit-voltage (mV)
<i>I</i>	cell current density ( $\text{mA}/\text{cm}^2$ )
<i>m</i> <sub>CO<sub>2</sub></sub>	molar fraction of CO <sub>2</sub>
<i>m</i> <sub>H<sub>2</sub>O</sub>	molar fraction of H <sub>2</sub> O
<i>p</i> <sub>CO<sub>2</sub></sub>	partial pressure of CO <sub>2</sub> (atm)
<i>p</i> <sub>O<sub>2</sub></sub>	partial pressure of O <sub>2</sub> (atm)
<i>Q</i> <sub>CO<sub>2</sub></sub>	CO <sub>2</sub> removal rate ( $\text{cm}^3/\text{min}$ )
<i>R</i> <sub>A</sub>	anode reaction resistance ( $\Omega \text{ cm}^2$ )
<i>R</i> <sub>C</sub>	cathode reaction resistance ( $\Omega \text{ cm}^2$ )
<i>R</i> <sub>ir</sub>	cell resistance ( $\Omega \text{ cm}^2$ )
<i>V</i> <sub>I</sub>	flow rate of cathode exhaust gas at on-load ( $\text{cm}^3/\text{min}$ )
<i>V</i> <sub>OCV</sub>	flow rate of cathode exhaust gas at open-circuit-voltage ( $\text{cm}^3/\text{min}$ )
<i>η</i> <sub>ne</sub>	Nernst loss (mV)

maintain isothermal condition. Both anode gas and cathode gas are humidified by passing the bubbler and supplied to MCFC. Cell voltage was measured and recorded by data logger. The gas composition of anode or cathode exhaust gases was analyzed by gas chromatograph and cell resistance was measured by a milliohm meter with ac four probes. In addition, gas flow rate of anode or cathode exhaust gases was measured by precision-film-flow-meter. MCFC made by National Institute of Advanced Industrial Science and Technology, Kansai (AIST) is 81 cm<sup>2</sup> single cell; these electrodes were sintered nickel as shown in Table 1. The electrolyte eutectic is a mixture of lithium carbonate (Li<sub>2</sub>CO<sub>3</sub>),

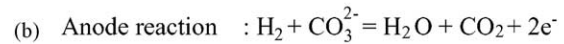
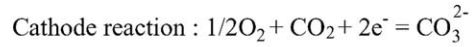
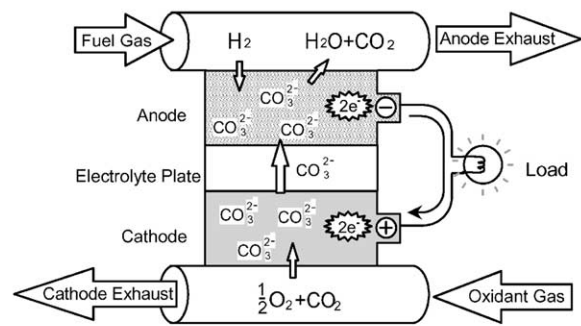
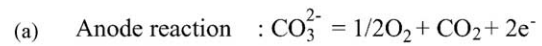
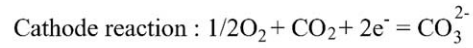
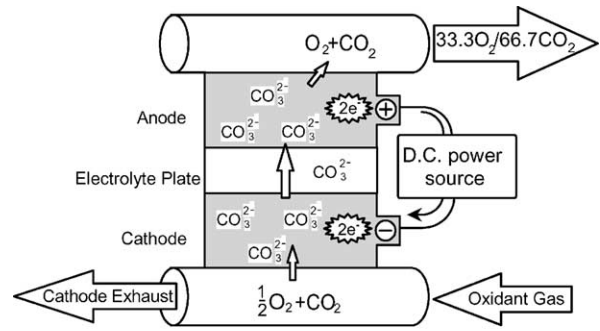


Fig. 1. Schematic diagram of CRDM method (a) decomposition and (b) generation operation.

by 52 mol%, and sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>), by 48 mol%. The electrolyte matrix is lithium aluminum oxide.

Experimental conditions are also shown in Table 1. The cell is heated to 923 K at 2 K/min of heating rate under

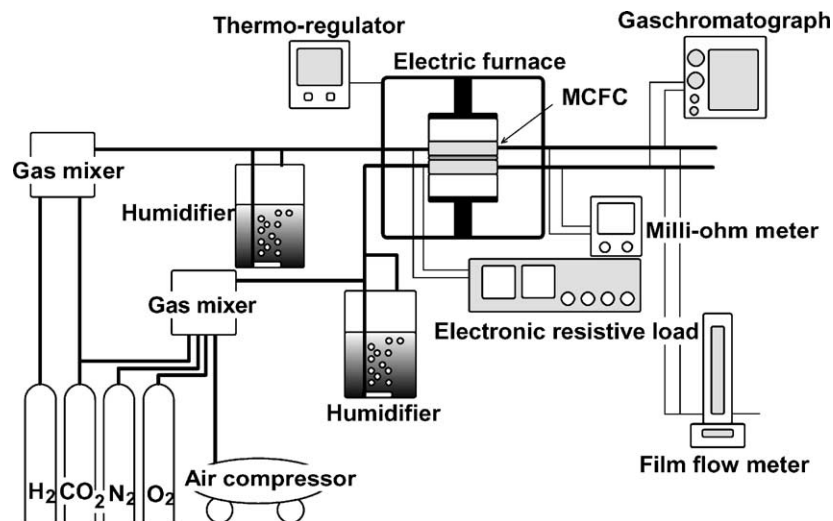


Fig. 2. Schematic diagram of experimental apparatus.

Table 1  
AISTs MCFC components and experimental conditions

	Cathode	Anode	Matrix
Materials	NiO (in situ)	Ni/Al	LiAlO <sub>2</sub>
Electrolyte		52Li <sub>2</sub> CO <sub>3</sub> /48Na <sub>2</sub> CO <sub>3</sub>	
Electrode area (cm <sup>2</sup> )		81	
Temperature (K)		923	
Pressure (MPa)		0.1	
Current density (mA/cm <sup>2</sup> )		0–150	
Gas utilization (%)		20–100	
Gas composition (%)	CO <sub>2</sub> (0.9–50) O <sub>2</sub> (0.45–40) H <sub>2</sub> O (0–50) N <sub>2</sub> (balance)	H <sub>2</sub> (70.4) CO <sub>2</sub> (17.6) H <sub>2</sub> O (12)	

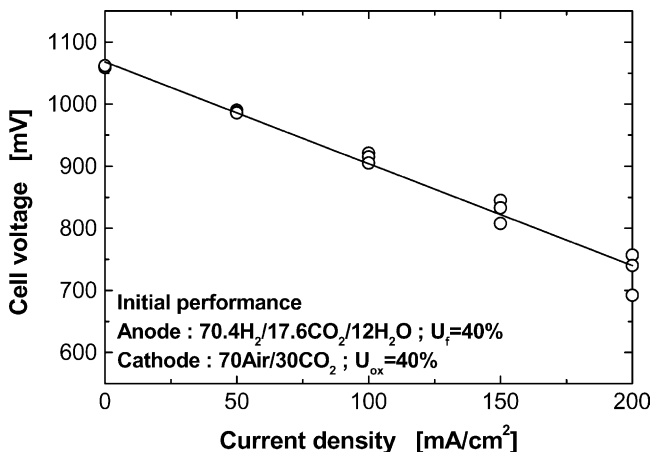


Fig. 3. Initial performance of AISTs cell.

atmospheric pressure. After the cell temperature becomes 923 K, the standard gas is supplied to each electrode. The anode standard gas is 70.4H<sub>2</sub>/17.6CO<sub>2</sub>/12H<sub>2</sub>O, and the cathode standard gas is 70air/30CO<sub>2</sub>. In order to study the effect of cathode gas composition on cell performance, CO<sub>2</sub>, O<sub>2</sub>, and H<sub>2</sub>O concentration of cathode gas were widely varied, but anode gas composition was constant. Gas utilization was varied 20–100%.

Fig. 3 shows an initial cell performance used by the experiment under the standard condition (gas composition is each standard gas composition; fuel gas utilization is 40%; oxidant gas utilization is 40%).

### 3. Results and discussion

#### 3.1. Removal rate of CO<sub>2</sub>

In evaluating the CO<sub>2</sub> removal experiment, the amount of CO<sub>2</sub>, which moves from the cathode to the anode, is the most important factor. Generally, the CO<sub>2</sub> gas according to the power generation of MCFC is generated at an anode and consumed at a cathode according to an electrochemical theory. Therefore, the CO<sub>2</sub> removal rate was evaluated by measuring the flow rate of cathode exhaust gas with each

current density. Here, the CO<sub>2</sub> removal rate ( $Q_{\text{CO}_2}$ ) is defined by Eq. (1),  $V_{\text{OCV}}$  means flow rate of cathode exhaust gas at open-circuit-voltage and  $V_1$  means flow rate of cathode exhaust gas at on-load

$$Q_{\text{CO}_2} = 0.667(V_{\text{OCV}} - V_1) \quad (1)$$

The experimental parameters are CO<sub>2</sub> concentration and current density. Fuel gas utilization is 75% and oxidant gas utilization is 50%. Fig. 4 shows the comparison between the experiment value and the theoretical value of the CO<sub>2</sub> removal rate. The experiment value and the theoretical value show an excellent agreement in Fig. 4. Therefore, the CO<sub>2</sub> removal rate can be obtained by calculation using the electrochemical theory.

#### 3.2. Effect of CO<sub>2</sub> concentration on cell performance

Fig. 5 shows the effect of CO<sub>2</sub> concentration on cell performance. The experimental parameters are the partial pressure ratio of CO<sub>2</sub> and O<sub>2</sub> ( $p_{\text{CO}_2}/p_{\text{O}_2}$ ), current density and CO<sub>2</sub> concentration. Here, fuel gas utilization is 40% and CO<sub>2</sub> gas utilization is 50%. Cell voltage decreases by decreasing CO<sub>2</sub> concentration; it decreases most drastically

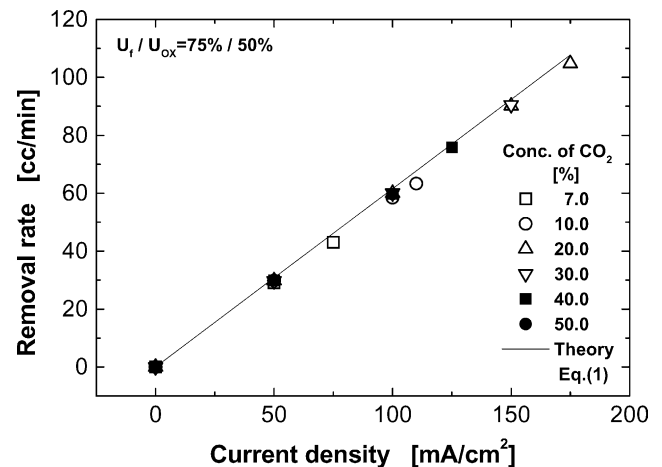


Fig. 4. Comparison between the experiment value and the theoretical value of the CO<sub>2</sub> removal rate.

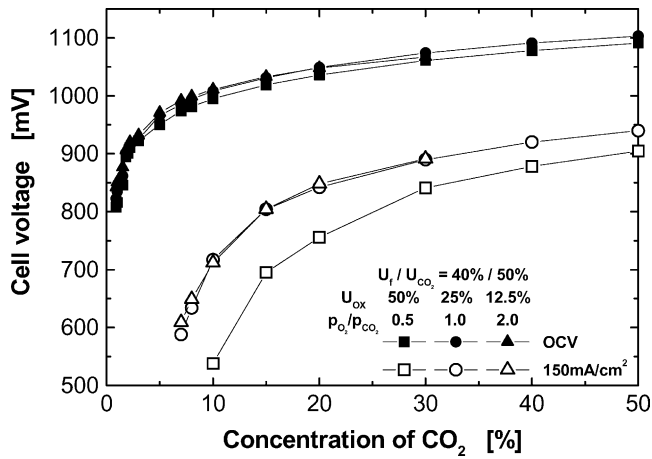


Fig. 5. Effect of CO<sub>2</sub> concentration on cell performance.

when the CO<sub>2</sub> concentration becomes 10% or less. These cell voltage drops, even considering the Nernst potential, grow by decreasing CO<sub>2</sub> concentration. Moreover, cell voltage is improved by decreasing the partial pressure ratio. Then, the effect of the partial pressure ratio on cell performance is examined as shown in Fig. 6. Here, the cell performance is rearranged by the difference between the open-circuit-voltage and the voltage at 50 mA/cm<sup>2</sup> of current density. The Nernst loss can be excluded from each voltage because cell voltage reduction caused by the Nernst loss is extremely smaller than cell voltage reduction by the change the partial pressure ratio. When the partial pressure ratio becomes one or less in the low CO<sub>2</sub> concentration region, the cell performance is drastically deteriorated. However, if the CO<sub>2</sub> concentration is high, the partial pressure ratio change does not influence cell performance. From the above-mentioned, the factor of the cell voltage drops by the low CO<sub>2</sub> concentration can be considered as follows:

- (1) These cell voltage drops are controlled by the diffusion of CO<sub>2</sub> at cathode side. As Reynolds number of

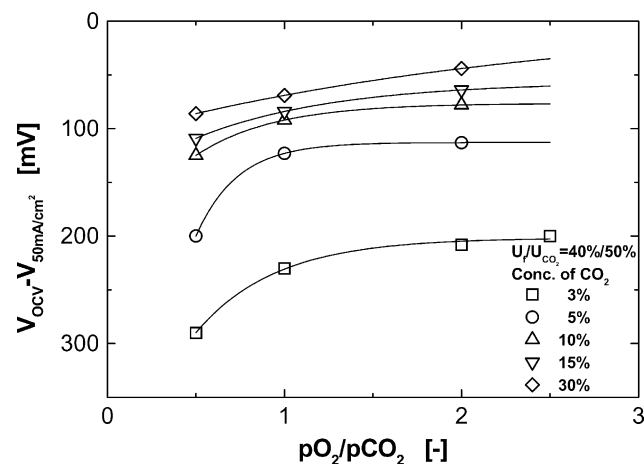


Fig. 6. Effect of the partial pressure ratio on cell performance.

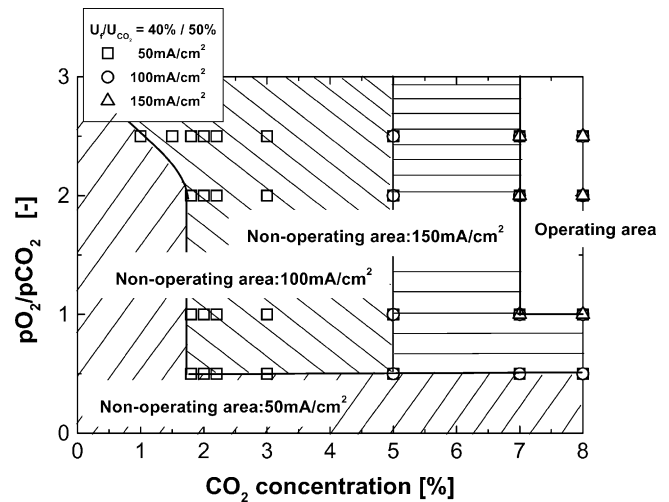


Fig. 7. Operation area of MCFC.

cathode is about 50–100, CO<sub>2</sub> cannot reach the cathode by mixing of the fluid.

- (2) The main reason for these cell voltage drops in the low CO<sub>2</sub> concentration region is the diffusion of O<sub>2</sub> to the cathode, because the cell voltage improved by decreasing the partial pressure ratio.

Therefore, the O<sub>2</sub> partial pressure should be increased to obtain a stable cell performance in the low CO<sub>2</sub> concentration region.

From these results, the CO<sub>2</sub> concentration region, where MCFC can be operated by keeping the voltage of 500 mV or more, is summarized in Fig. 7. MCFC can hardly be operated in the low CO<sub>2</sub> concentration region when the current density is increased. When CO<sub>2</sub> concentration becomes 7% or less, MCFC cannot be operated at 150 mA/cm<sup>2</sup> of current density. However, MCFC can be operated by setting the current density to 50 mA/cm<sup>2</sup> in the low CO<sub>2</sub> concentration region. If the conversion efficiency is bad even if MCFC can be operated in the low current density, it is not worth operating MCFC. Therefore, the energy conversion efficiency of MCFC in the low CO<sub>2</sub> concentration region should be examined.

Generally, efficiency is defined in the ratio of the electric power and the reactive heat of hydrogen used by the cell reaction. An increase in the flow rate means an increase in the quantity of heat to raise the gas up to the cell temperature, but this quantity of heat is not reflected in conventional efficiency. In the low CO<sub>2</sub> concentration region, it is necessary to increase the cathode flow rate to increase the partial pressure of oxygen and keep CO<sub>2</sub> utilization. Therefore, the energy conversion ratio ( $\eta_{EC}$ ) by which an increase in the cathode flow rate is reflected in efficiency is defined by Eq. (2). Fig. 8 shows the energy conversion efficiency by using Eq. (2) in 7% of CO<sub>2</sub>. Energy conversion efficiency decreases as the current density increases even if  $p_{O_2}/p_{CO_2}$  is changed. Therefore, MCFC in the low CO<sub>2</sub> concentration region should be operated

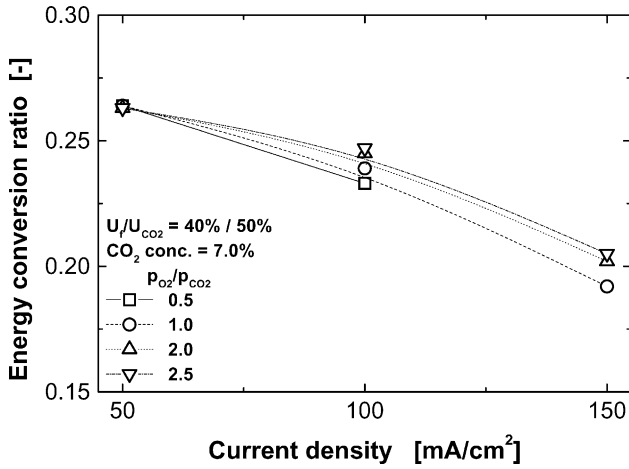


Fig. 8. Effect of operation condition on energy conversion ratio.

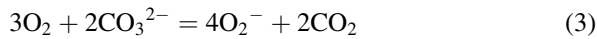
under 50 mA/cm<sup>2</sup> of current density

$$\eta_{EC} = \frac{IV}{H_{IN}^C + H_{IN}^A} \quad (2)$$

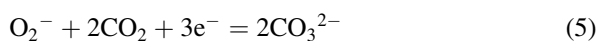
### 3.3. Influence of CO<sub>2</sub> concentration on the cathode reaction mechanism

The influence of CO<sub>2</sub> concentration on the cathode reaction mechanism is examined from the cathode reaction models, which was proposed by Nishina et al. [4]. There are two kinds of oxygen reduction mechanisms about (A) the superoxide path (SOP) and (B) the peroxide path (POP). And, the cathode reaction resistance ( $R_C$ ) in each mechanism is shown by Eqs. (6) and (10). Here,  $a$ ,  $a'$ ,  $b$  and  $b'$  are constants that put together the factors except temperature and partial pressure. Here, the values of cathode reaction resistance ( $R_C$ ) are calculated on Eq. (11) with the experimental data, which are open-circuit-voltage ( $E$ ), cell voltage ( $V$ ), and cell resistance ( $R_{ir}$ ) and cell current ( $I$ ).  $\eta_{ne}$ , which is Nernst loss, is calculated by gas compositions.  $p_{CO_2}$  and  $p_{O_2}$  are treated as average values of inlet and outlet partial pressure at the cell. The anode reaction resistance ( $R_A$ ) can be treated as constant, because the experiment makes the anode gas composition constant [5]. In addition,  $R_A$  is neglected as  $R_A$  is smaller than  $R_C$ .

(A) superoxide path (SOP)

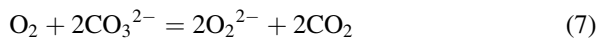


$$[O_2^-] = K_1 p_{O_2}^{0.75} p_{CO_2}^{-0.5} \quad (4)$$



$$R_C p_{CO_2} = a T p_{O_2}^{-0.75} p_{CO_2}^{1.5} + b T \quad (6)$$

(B) peroxide path (POP)



$$[O_2^{2-}] = K_2 p_{O_2}^{0.5} p_{CO_2}^{-1.0} \quad (8)$$

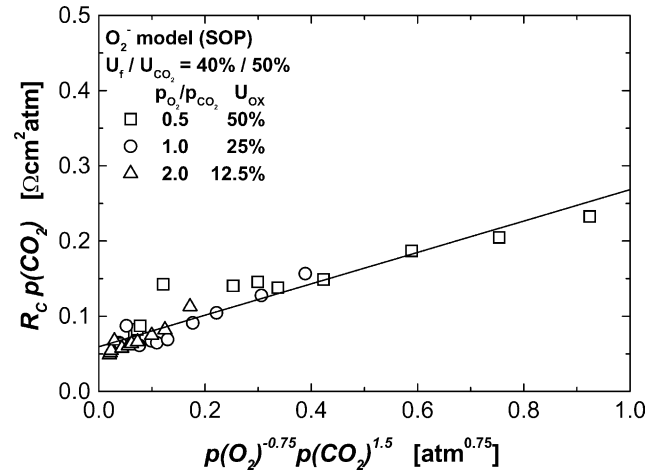
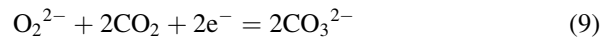


Fig. 9. Influence of CO<sub>2</sub> partial pressure on cathode reaction mechanism (superoxide model).



$$R_C p_{CO_2} = a' T p_{O_2}^{-0.5} p_{CO_2}^{2.0} + b' T \quad (10)$$

$$V = E - \eta_{ne} - (R_{ir} + R_A + R_C)I \quad (11)$$

If the partial pressure dependence of cathode reaction resistance corresponds to Eq. (6) in the SOP model, it will show the linear relation on  $p_{O_2}^{-0.75} p_{CO_2}^{1.5}$  versus  $R_C p_{CO_2}$  plots. Then, Fig. 9 shows the result of arranging the result of Fig. 5 by Eq. (6). The linearity of the cathode reaction resistance according to change in the partial pressures of CO<sub>2</sub> and O<sub>2</sub> is excellent. On the other hand, if the partial pressures dependence of cathode reaction resistance corresponds to Eq. (10) in the POP model, it will show up the linear relation on  $p_{O_2}^{-0.5} p_{CO_2}^{2.0}$  versus  $R_C p_{CO_2}$  plots. The linearity in Fig. 10 does not seem to be better than that in Fig. 9. The SOP model is applicable in the cathode reaction even by the low CO<sub>2</sub> concentration region.

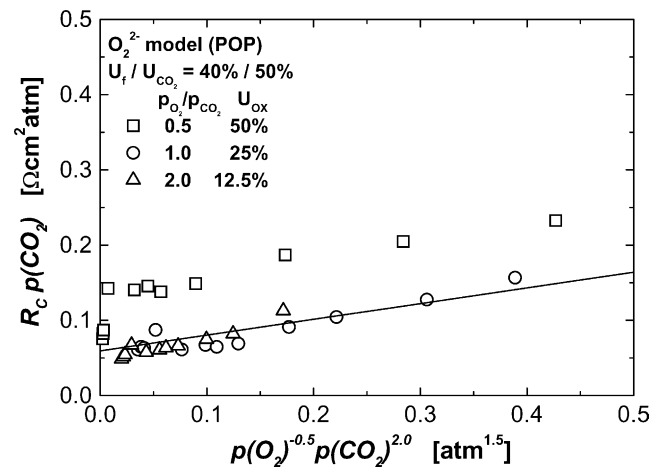


Fig. 10. Influence of CO<sub>2</sub> partial pressure on cathode reaction mechanism (peroxide model).

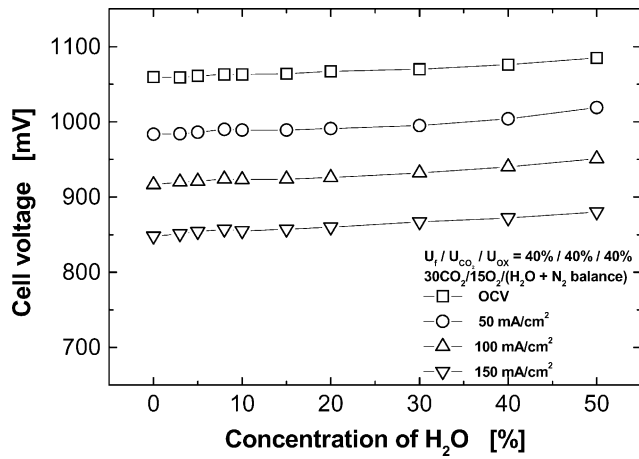


Fig. 11. Effect of H<sub>2</sub>O concentration in cathode on cell performance.

#### 3.4. Influence of H<sub>2</sub>O concentration in cathode on cell performance

The influence of the steam addition to cathode on the cell performance should be examined if the exhaust gas of the thermal power plant is used as the cathode gas of MCFC. Fig. 11 shows the influence of the steam addition to cathode on cell performance. Here, H<sub>2</sub>O concentration is balanced by N<sub>2</sub> concentration, because the CO<sub>2</sub> and O<sub>2</sub> concentrations are fixed by 30 and 15%, respectively. Cell voltage increases gradually when H<sub>2</sub>O concentration becomes 15% or more. Here, the factor of cell performance improvement by the H<sub>2</sub>O addition can be examined by elucidating H<sub>2</sub>O concentration dependency to the cathode reaction resistance that is proposed by Mugikura et al. This equation is expressed based on Eq. (5) as shown in Eq. (12) [6]. Here, *A*, *B* and *C* (=0.16) are constants that put together the factors except temperature and partial pressure. *m*(*i*) means molar fraction of *i* chemical species

$$R_C = Ap_{O_2}^{-0.75} p_{CO_2}^{0.5} + B(m_{CO_2} + Cm_{H_2O})^{-1} \quad (12)$$

If H<sub>2</sub>O concentration dependence of cathode reaction resistance corresponds to Eq. (12), the relation between RC and  $1/(m_{CO_2} + Cm_{H_2O})$  must be linearity under the condition that  $p_{O_2}$ ,  $p_{CO_2}$ ,  $m_{CO_2}$  are constants. Fig. 12 shows the effect of H<sub>2</sub>O molar fraction on the cathode reaction mechanism (SOP). Here, fuel gas utilization, CO<sub>2</sub> gas utilization, and oxygen gas utilization are all 40%,  $p_{O_2}$  is 0.15,  $p_{CO_2}$  is 0.3 and  $m_{CO_2}$  is 0.3, respectively. The linearity of the cathode reaction resistance according to change in molar fraction of H<sub>2</sub>O is excellent. Thus, the added steam decreases the cathode reaction resistance because it reacts with a superoxide as in Mugikura's proposal. Therefore, cell performance improves if the exhaust gas of the thermal power plant is supplied to cathode of MCFC directly without separating the gas and the liquid. Moreover, the system can be constructed in a simple manner because the exhaust gas does not need separating into the gas and the liquid.

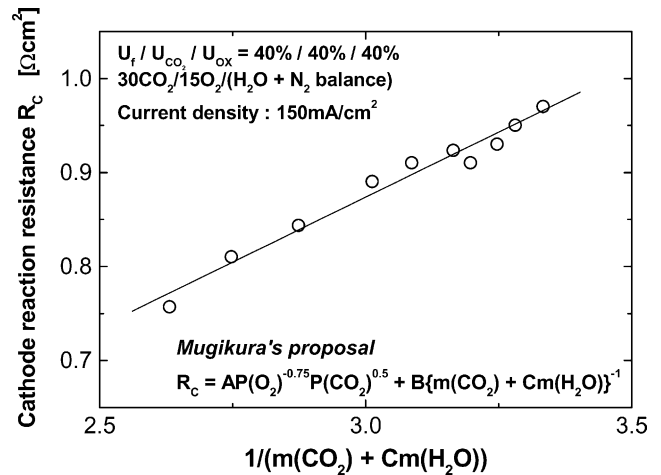


Fig. 12. Influence of H<sub>2</sub>O molar fraction on cathode reaction mechanism.

#### 3.5. Optimization of the operating conditions for CRDM

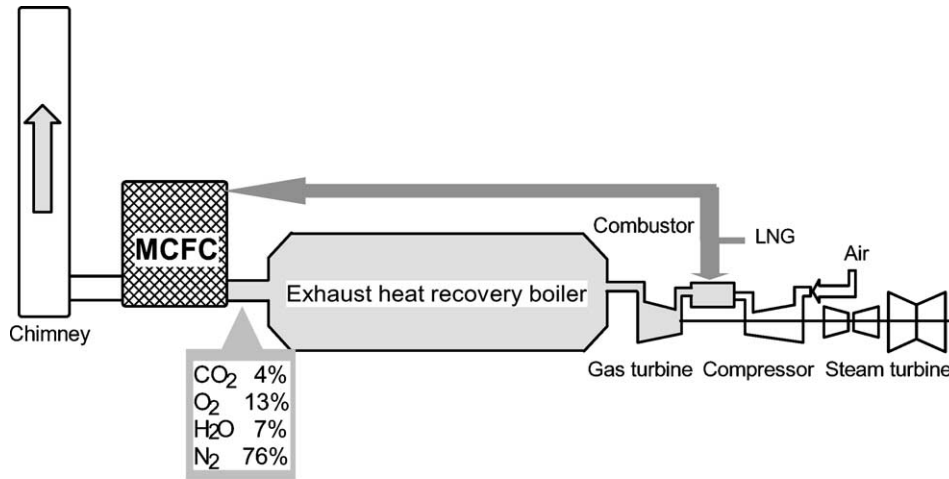
The operating conditions of MCFC have to be optimized if an exhaust gas from the thermal power plant is used as a cathode gas. The power generation method of Japan is generally divided into two systems. One is the conventional thermal power plant and another is the combined cycle system as shown in Fig. 13. Exhaust gas composition is also greatly different in these two thermal power plants. CO<sub>2</sub> concentration of these thermal power plants is too low (as low as 4–8%) to use exhaust gas as a cathode gas for MCFC. The optimum operating condition when the exhaust gas from these two kinds of thermal power plants was used as a cathode gas for MCFC made in AIST was experimentally examined. Moreover, The optimum scale of MCFC was also examined if MCFC was introduced into each thermal power plant as CRDM.

Here, the above-mentioned energy conversion efficiency and the removal efficiency were used as an index that examined the optimum conditions. The removal efficiency ( $R_{\text{eff}}$ ) is also defined by Eq. (13) to evaluate the CO<sub>2</sub> removal rate

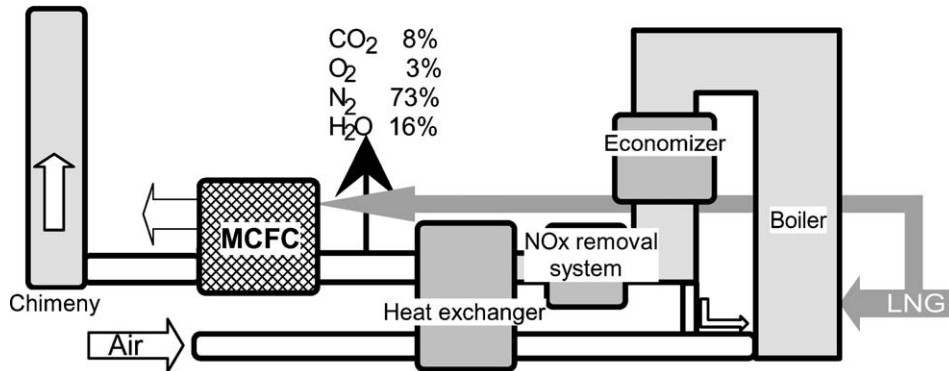
$$R_{\text{eff}} = \frac{X_{\text{IN}}^{\text{CO}_2} - X_{\text{OUT}}^{\text{CO}_2}}{X_{\text{IN}}^{\text{CO}_2}} \quad (13)$$

Here,  $X_{\text{IN}}^{\text{CO}_2}$  and  $X_{\text{OUT}}^{\text{CO}_2}$  mean CO<sub>2</sub> concentration of inlet and outlet of cathode, respectively.  $H_{\text{IN}}^{\text{C}}$ ,  $H_{\text{IN}}^{\text{A}}$ ,  $I$  and  $V$  mean the enthalpy of anode gases, the enthalpy of cathode gases, current and cell voltage, respectively.

Fig. 14 shows the cell performance of MCFC when the exhaust gas from the combined cycle system is used as the cathode gas. Here, fuel gas utilization is 40%; the partial pressure ratio of O<sub>2</sub> and CO<sub>2</sub> ( $p_{O_2}/p_{CO_2}$ ) is 3.22. Cell voltage is almost constant, though CO<sub>2</sub> utilization increases. The reason is that the partial pressure of O<sub>2</sub> is so high though CO<sub>2</sub> concentration is low. On the other hand, the removal efficiency and the energy conversion ratio increase by



(a) Combined cycle system



(b) Thermal power plant

Fig. 13. Schematic diagram of thermal power plants with MCFC.

increasing CO<sub>2</sub> utilization. The energy conversion ratio is highest when the current density is 100 mA/cm<sup>2</sup>. Therefore, the combined cycle system and MCFC can be connected directly and the optimum operating conditions are that CO<sub>2</sub> utilization is 90% and current density is 100 mA/cm<sup>2</sup> in the case of using MCFC made in AIST. For example, the case where MCFC is connected with a combined system of 236 MW as CRDM is provisionally calculated. MCFC can obtain 84 MW (0.75 V × 112,000 cells) of electric power under the optimum operating condition, if the electrode area is assumed to be 1 m<sup>2</sup>. The target of COP3, whose removal efficiency is 24%, is achieved because CO<sub>2</sub> concentration (4%) in exhaust gas from the combined cycle system decreases to 0.4% by using as cathode gas of MCFC.

Next, the cell performance of MCFC when MCFC is connected with the conventional thermal power plant is shown in Fig. 15. This case cannot achieve the target of

COP3 because MCFC cannot obtain cell voltage of 500 mV or more under this gas composition even if CO<sub>2</sub> gas utilization is decreased. The reason for bad cell performance is that the  $p_{O_2}/p_{CO_2}$  is low such as 0.37. From the above mentioned, the  $p_{O_2}/p_{CO_2}$  of cathode gas has to be increased by adding air to achieve good cell performance. Fig. 16 shows the effect of additive ratio of air on cell performance. Here, CO<sub>2</sub> utilization keeps at 40% even if air is added to cathode gas. Fuel gas utilization is 40%. Cell voltage increases as the additive ratio of air increases, and it is saturated when the additive ratio of air becomes 20% or more. The removal efficiency is roughly constant because CO<sub>2</sub> utilization stays at 40%. On the other hand, the energy conversion ratio gets the maximum value when the additive ratio of air is 20% and current density is 100 mA/cm<sup>2</sup> in the case of using MCFC made in AIST. The  $p_{O_2}/p_{CO_2}$  at the maximum value of energy conversion ratio is 0.9. Therefore, it is necessary to increase the

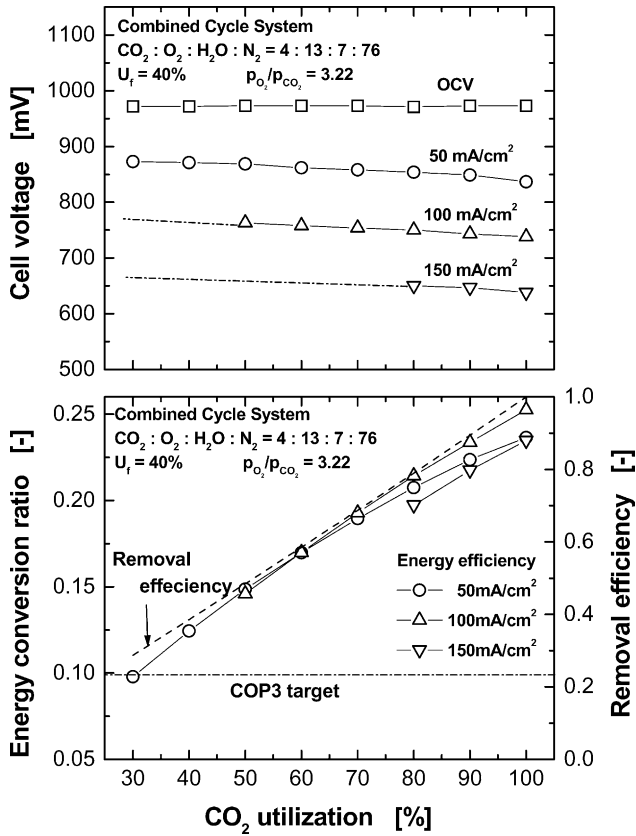


Fig. 14. Cell performance of MCFC when the exhaust gas from the combined cycle system is used as a cathode gas.

partial pressure of oxygen to obtain a good performance of MCFC in low CO<sub>2</sub> concentration region such as the exhaust gas composition of the thermal power plant. For example, the case where MCFC is connected with the conventional thermal power plant of 600 MW as CRDM is provisionally calculated. MCFC can obtain 94.5 MW (0.75 V × 126,000 cells) of electric power under the condition that CO<sub>2</sub> utilization is 40% and current density is 100 mA/cm<sup>2</sup>, if the electrode area is assumed to be 1 m<sup>2</sup>. The target of COP3 is achieved because CO<sub>2</sub> concentration (6.7%) in exhaust gas from the conventional thermal power plant decreases to 4.2% by using as a cathode gas of MCFC.

Therefore, MCFC can be connected directly to the combined cycle system. Moreover, MCFC and the conventional thermal power plant can be connected by adding 20% air.

### 3.6. Examination of purity of CO<sub>2</sub> taken out of MCFC

The discharge of CO<sub>2</sub> from the thermal power plant could be decreased by connecting MCFC with the thermal power plant. If fuel gas utilization of MCFC was too, high purity CO<sub>2</sub> can be obtained by separating the gas and the liquid of the anode gases. Therefore, the influence of fuel gas utilization on cell performance should be evaluated under the optimum conditions of the cathode gas in each thermal power plant. Fig. 17 shows the effect of fuel gas utilization

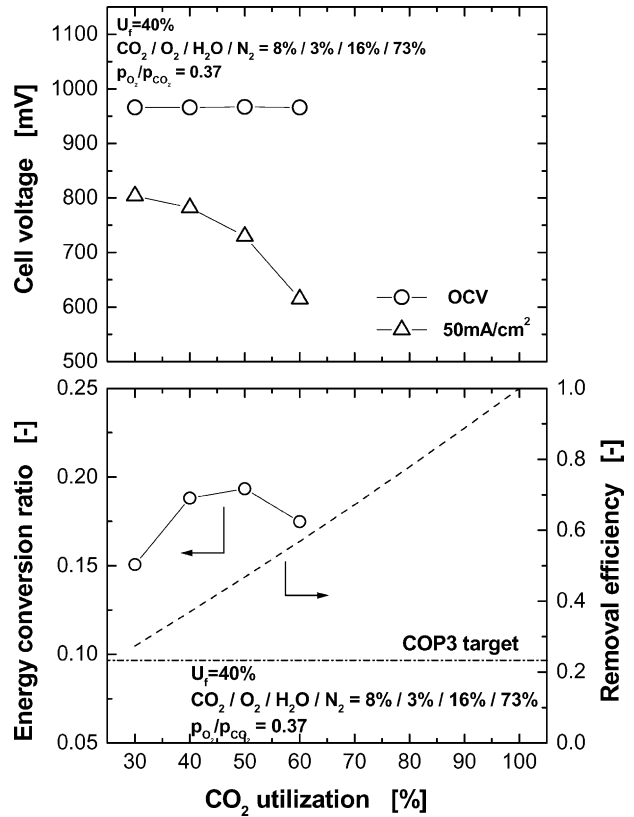


Fig. 15. Cell performance of MCFC when the exhaust gas from the conventional thermal power plant is used as a cathode gas.

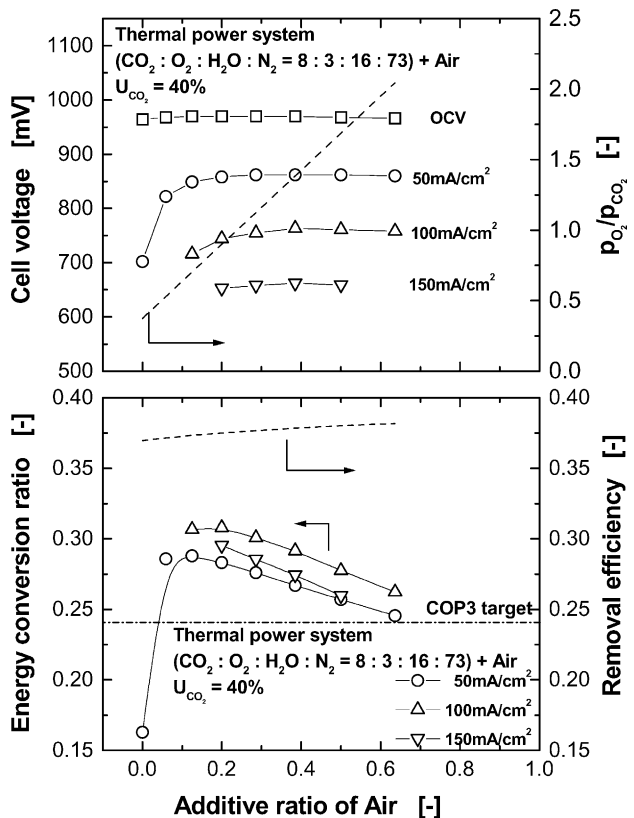


Fig. 16. Effect of the additive rate of air on cell performance.



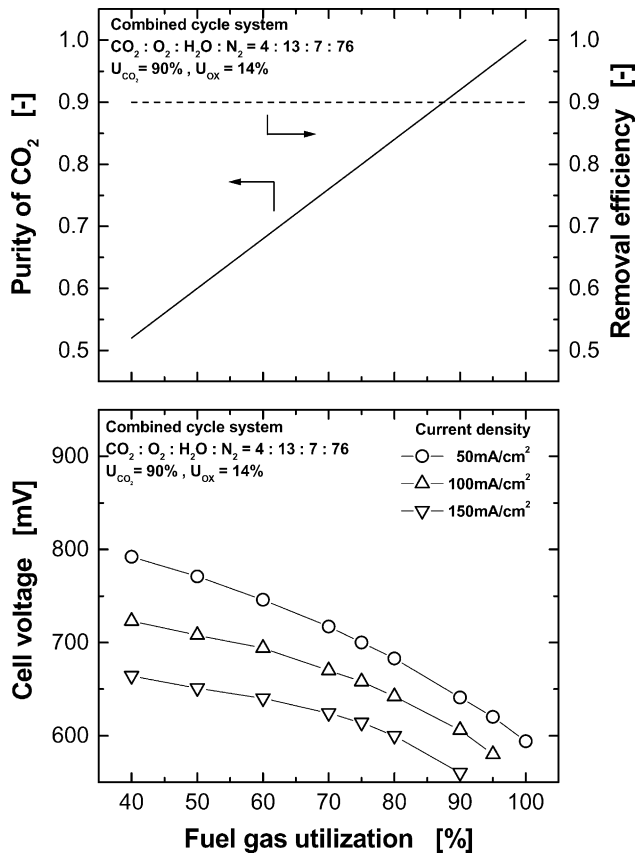


Fig. 17. Effect of fuel gas utilization on cell performance (combined cycle system and MCFC).

on cell performance in the case of using the exhaust gas of the combined cycle system as cathode gas. Here, the removal efficiency is constant even if fuel gas utilization increases because cathode gas composition and CO<sub>2</sub> gas utilization (90% from Fig. 14) are constant. H<sub>2</sub> concentration of anode outlet decreases by increasing fuel gas utilization. Moreover, the cell voltage deteriorates drastically by increasing fuel gas utilization. However, MCFC can be operated on 100% of fuel gas utilization and cell voltage is about 600 mV under current density of 50 mA/cm<sup>2</sup>. Therefore, MCFC can obtain 33.6 MW (0.60 V × 112,000 cells; electrode area: 1 m<sup>2</sup>) of electric power and pure CO<sub>2</sub> under this conditions.

Fig. 18 shows the effect of fuel gas utilization on cell performance in the case of using the exhaust gas of the conventional thermal power plant as cathode gas. This system can obtain cell voltage at high current density, though the removal efficiency is lower than the combined cycle system with MCFC. MCFC can obtain 38.4 MW (0.61 V × 126,000 cells; electrode area: 1 m<sup>2</sup>) of electric power and pure CO<sub>2</sub> when fuel gas utilization is 100% and the current density is 50 mA/cm<sup>2</sup>.

From the above-mentioned, as these CRDM systems can remove and/or concentrate CO<sub>2</sub>, connecting MCFC with each thermal power plant is a powerful method to achieve the target of COP3.

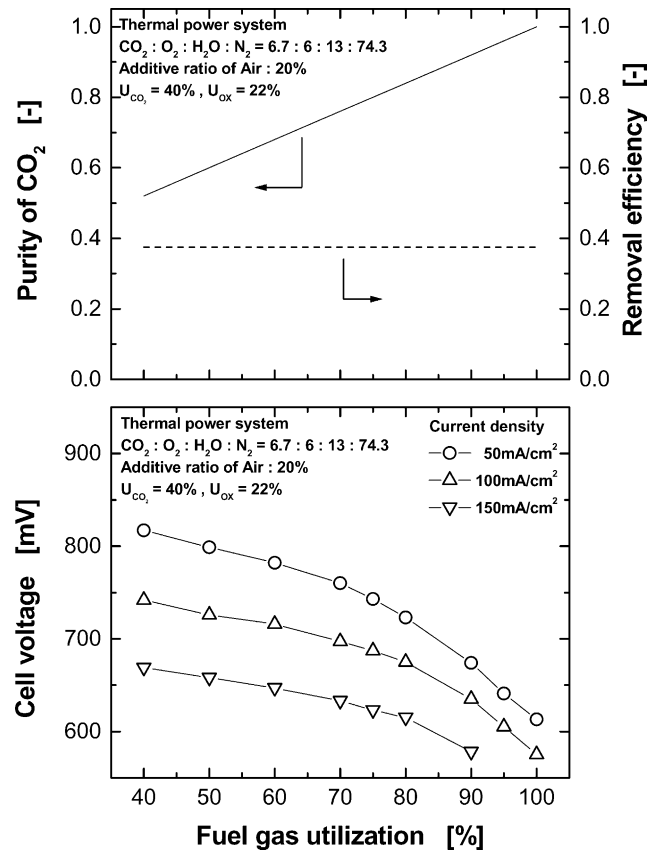


Fig. 18. Effect of fuel gas utilization on cell performance (conventional thermal power plant and MCFC).

#### 4. Conclusion

This study aimed to decrease the CO<sub>2</sub> emission to atmosphere by using exhaust gas from the thermal power plant as a cathode gas for MCFC and to find the best operating conditions of MCFC. The obtained results in this study are summarized as follows:

- (1) The experimental value of the CO<sub>2</sub> removal rate from cathode to anode is almost corresponds to the theoretical value.
- (2) Cell voltage decreases by decreasing the  $p_{O_2}/p_{CO_2}$  ratio.
- (3) Oxygen partial pressure should be increased if MCFC operates under low CO<sub>2</sub> concentration.
- (4) The cathode reaction mechanism, which is the superoxide model, is not influenced by the low CO<sub>2</sub> partial pressure.
- (5) Although the detailed reaction mechanism was not clarified, the steam addition is likely to promote the dissolution rate of CO<sub>2</sub> to molten salts. Cell performance improves if the exhaust gas of the thermal power plant is supplied to cathode of MCFC directly without separating the gas and the liquid.
- (6) The combined cycle system and MCFC can be connected directly. The target of COP3 is achieved

- because CO<sub>2</sub> of 4% in exhaust gas decreases to 0.4%.
- (7) The conventional thermal power plant and MCFC can be connected by adding 20% air. The target of COP3 is achieved because CO<sub>2</sub> of 8% in exhaust gas decreases to 4.2%.
- (8) These CRDM systems can produce pure CO<sub>2</sub> because MCFC can be operated on 100% fuel gas utilization.

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