

## Single-Chamber Solid Oxide Fuel Cells at Intermediate Temperatures with Various Hydrocarbon-Air Mixtures

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The performance of a single-chamber solid oxide fuel cell (SOFC) was studied between 350 and 900°C in flowing mixtures of methane, ethane, propane, or liquefied petroleum gas and air with a fuel/air volume ratio of one, where their oxidation proceeded safely without explosion. Among all tested electrode materials, Ni-Ce<sub>0.8</sub>Sm<sub>0.2</sub>O<sub>1.9</sub> cermet and Sm<sub>0.5</sub>Sr<sub>0.5</sub>CoO<sub>3</sub> oxide functioned best as the anode and cathode, respectively, in various gas mixtures. A cell constructed from a La<sub>0.9</sub>Sr<sub>0.1</sub>Ga<sub>0.8</sub>Mg<sub>0.2</sub>O<sub>3</sub> electrolyte with the two electrodes generated >900 mV in a methane-air mixture between 600 and 800°C and in an ethane-air mixture between 450 and 650°C. A small electrode reaction resistance resulted in increasing power density with decreasing electrolyte thickness. The peak power density at 450°C increased from 34 to 101 mW cm<sup>-2</sup> with decreasing electrolyte thickness from 0.50 to 0.18 mm. The working mechanism of the single-chamber SOFC at different temperatures was also studied by measuring the catalytic activities of the two electrodes for partial oxidation of the hydrocarbons.

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There is worldwide interest in the development and commercialization of fuel cells for vehicles and portable electric devices. It is a presently accepted notion that polymer electrolyte fuel cells (PEFCs) are the only devices capable of operating at low temperatures. We believe, however, that PEFCs are not perfect from a practical point of view because they require hydrogen as the fuel, which is impractical in terms of storage and handling. An external reformer, therefore, must be used to convert more viable alcohols and hydrocarbons into hydrogen, thereby defeating their portability. There have been recent successes with solid oxide fuel cells (SOFCs) which perform well between 500 and 700°C directly using alcohols and hydrocarbons as the fuels.<sup>1-3</sup> A further reduction in the operating temperature of internal-reforming SOFCs and an enhancement of their thermal- and mechanical-shock resistance would make this technology a promising alternative to PEFCs.

A novel type of fuel cell, which is distinguished from conventional fuel cells in design and principle, has been proposed by many researchers.<sup>4-8</sup> This fuel cell consists of only one gas chamber, where both the anode and the cathode are exposed to the same mixture of fuel and air. We will use a single-chamber fuel cell (SCFC) as a notation for this type of fuel cell. Because there is no need to separate the supply of fuel and air, it is more thermally and mechanically shock resistant than conventional fuel cells. We have recently succeeded in applying this cell design to an SOFC constructed from an yttria-stabilized zirconia (YSZ) electrolyte with a Ni-based anode and a strontium-doped lanthanum manganite (LSM) cathode.<sup>9</sup> This SCFC exhibits high power density in a flowing mixture of methane and air, but it must operate at the high temperature of 950°C in order to achieve sufficient ionic conduction in the solid electrolyte.

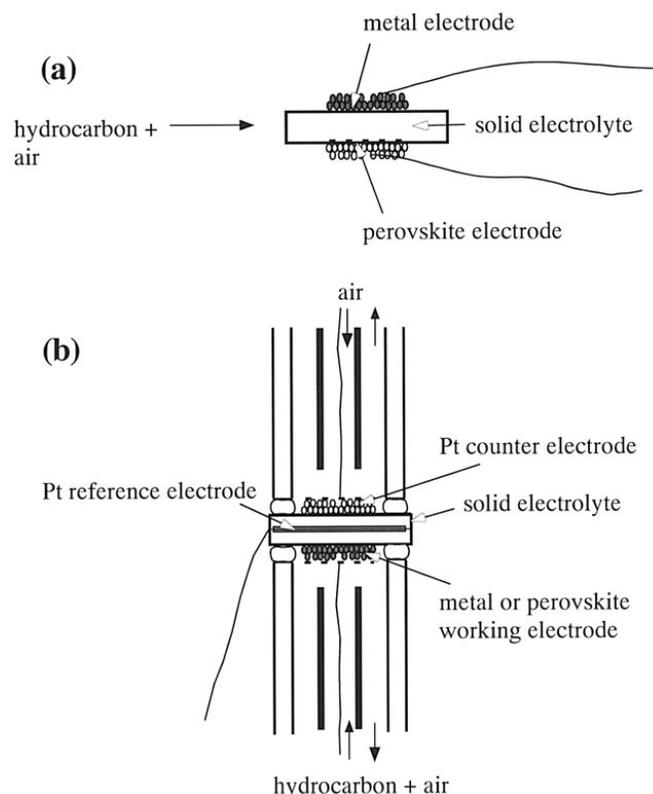
The operation of SOFCs at reduced temperatures causes excessive ohmic and polarization losses in the cell. Thus, it is necessary to use a highly conductive electrolyte together with a highly active anode and cathode. Lanthanum gallate<sup>10-14</sup> or ceria-based<sup>15-18</sup> oxides would be promising electrolytes because of their much higher ionic conductivities than that of YSZ. In addition, Ni-ceria cermets<sup>19</sup> and Co-based perovskite oxides,<sup>20,21</sup> which exhibit mixed ionic and electronic conduction under each of their respective operating conditions, have been generally regarded as suitable anodes and cathodes, respectively, at reduced temperatures.

In this study, we demonstrate that it is possible to operate a thermally and mechanically shock-resistant SOFC at reduced tempera-

tures by combining the advantages of using a highly conductive electrolyte with the single-chamber cell design. We also show that ethane, propane, and liquefied petroleum gas (LPG) can be successfully used as the fuels in the present SCFC, especially at operating temperatures below 550°C.

### Experimental

Figure 1a shows an SCFC constructed for fuel-cell tests at reduced temperatures. La<sub>0.9</sub>Sr<sub>0.1</sub>Ga<sub>0.8</sub>Mg<sub>0.2</sub>O<sub>3</sub> (LSGM),



**Figure 1.** Schematic illustrations of (a) a single-chamber cell for fuel-cell tests and (b) a two-chamber cell for polarization and catalytic activity measurement.

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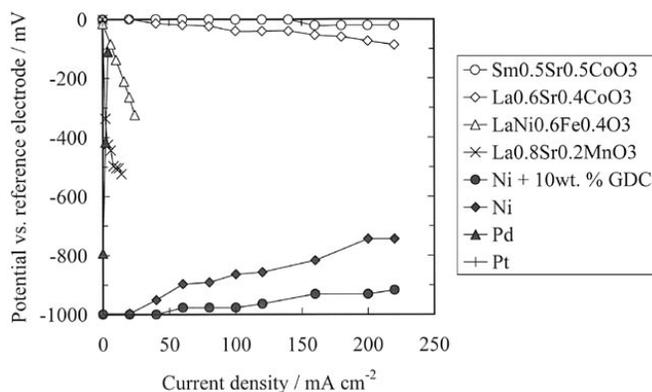
Ce<sub>0.8</sub>Sm<sub>0.2</sub>O<sub>1.9</sub> (SDC), and YSZ (8 mol % yttria) were used as the electrolytes. Both LSGM and SDC were prepared by pressing their commercial ceramic powders (Anan Kasei) hydrostatically into pellets at  $2 \times 10^3 \text{ kg cm}^{-2}$  and then sintering in air at 1500°C for 10 h. After the pellet was cut into a disk (ca. 14 mm diam, ca. 3 mm thick) with a diamond cutter, the disk surface was polished to a given thickness (0.18-0.50 mm) with an abrasive paper. The sintered YSZ disk (14 mm diam, 0.5 mm thick) was purchased from Sumitomo Osaka Cement. Unless otherwise stated, the 0.5 mm thick electrolyte was used as the test specimen.

10 wt % SDC-containing Ni [Ni (SDC)] metal was used as the anode. NiO powder (Kojundo Chemical Laboratory) was mixed with the SDC powder in ethyl carbitol and then ground with a zirconia mill container and zirconia grinding balls for 1 h. Ni, Pd, and Pt metals were also used as the anodes for comparison. These pastes were smeared on one surface (0.5 cm<sup>2</sup> area) of the electrolyte disk as thinly as possible with a brush, followed by calcining in air at 950°C for 4 h. Sm<sub>0.5</sub>Sr<sub>0.5</sub>CoO<sub>3</sub> was used as the cathode. The desired amounts of Sm<sub>2</sub>O<sub>3</sub>, SrCO<sub>3</sub>, and Co<sub>3</sub>O<sub>4</sub> powders were mixed in ethanol with a mortar and a pestle and then calcined in air at 1200°C for 6 h. La<sub>0.6</sub>Sr<sub>0.4</sub>CoO<sub>3</sub>, LaNi<sub>0.6</sub>Fe<sub>0.4</sub>O<sub>3</sub>, and La<sub>0.8</sub>Sr<sub>0.2</sub>MnO<sub>3</sub> oxides were also prepared by a solid-state reaction, for comparison with the Sm<sub>0.5</sub>Sr<sub>0.5</sub>CoO<sub>3</sub> oxide. These pastes were applied on another face of the electrolyte disk in a manner similar to that stated and then calcined in air at 950°C for 4 h. A Pt wire and a Pt mesh were used as the output terminal and the electrical collector, respectively, for the Ni (SDC) anode, and a Au wire and a Au mesh were similarly used for the Sm<sub>0.5</sub>Sr<sub>0.5</sub>CoO<sub>3</sub> cathode. The cell thus fabricated was placed in an alumina tube (15 and 19 mm inner and outer diameters, respectively). Methane, ethane, propane, and LPG were mixed with air to a concentration of 18 vol % so that the oxidation would proceed safely without explosion. A mixture of ethanol and air with an ethanol concentration of 7.7 vol % was also prepared by passing air through an ethanol bubbler at 25°C. The gas mixture was supplied to the cell at flow rates of 300 mL min<sup>-1</sup> between 350 and 900°C. The fuel-cell tests were carried out by measuring the terminal voltage between the two electrodes during cell discharge using a galvanostat (Hokuto Denko HA-501) and by measuring the impedance spectrum under open-circuit conditions using an impedance analyzer (Solartron SI-1260).

Figure 1b shows a two-chamber cell constructed for the measurement of the polarization properties of the Ni (SDC) and Sm<sub>0.5</sub>Sr<sub>0.5</sub>CoO<sub>3</sub> working electrodes and their catalytic activities for the oxidation of hydrocarbons. These electrodes were deposited on the bottom surface (0.5 cm<sup>2</sup> area) of the electrolyte disk in the same manner as stated previously. Pt counter and reference electrodes were deposited by smearing the Pt paste on the top surface (0.5 cm<sup>2</sup> area) and the side surface of the electrolyte disk, respectively, and then calcining at 950°C for 1 h in air. Two gas chambers were set up by placing the cell between two alumina tubes (9 and 13 mm inner and outer diameters, respectively). Each chamber was sealed by melting a glass ring gasket at the joint of the tube and the electrolyte at 950°C. The working chamber was supplied with the same gas mixture described previously at a flow rate of 300 mL min<sup>-1</sup> between 450 and 950°C. The counter and reference electrodes were exposed to atmospheric air. The anodic or cathodic overpotential, which is the ohmic loss-free potential, of the working electrode was measured by a current-pulse generator (Nikko Keisoku NCPG-101), where the rise and fall times were 2 μs with a precision of ±0.01%. The analysis of the outlet gas from the working chamber was performed on a dry basis using on-line gas chromatography (Schimazu GC-8A). The separation of methane, oxygen, hydrogen, and carbon monoxide was performed using a molecular sieve 5A column at 50°C, and the others were analyzed using a Porapak Q column at 50°C.

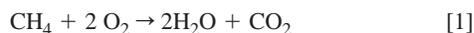
### Results and Discussion

*Cell performance in a flowing mixture of methane and air.*—Figure 2 shows the polarization curves of the four metal working electrodes in the two-chamber cell (Fig. 1b) using LSGM as the electrolyte in a flowing mixture of methane and air at 700°C. The poten-

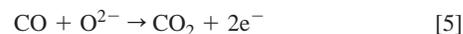
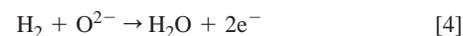


**Figure 2.** Polarization curves of different metal and perovskite oxide electrodes in a two-chamber cell: methane 52 mL min<sup>-1</sup>, oxygen 52 mL min<sup>-1</sup>, nitrogen 196 mL min<sup>-1</sup>, operating temperature 700°C. All overpotentials are free of ohmic loss.

tials of the metal electrodes under open-circuit conditions were -1000 mV for Ni (SDC), -992 mV for Ni, -793 mV for Pd, and -7 mV for Pt. As can be seen from Table I, where the compositions of the outlet gases from these electrodes are summarized, methane was oxidized to hydrogen, carbon monoxide, and carbon dioxide at the first three electrodes, whereas the oxidation rate of methane was too slow to be measured at the Pt electrode. In addition, the amounts of hydrogen and carbon monoxide formed were in the order Ni (SDC) > Ni > Pd, which corresponds to the potential trend. These results indicate that the following reactions proceed at the Ni (SDC), Ni, and Pd working electrodes



(see Ref. 22)



The overpotential of these electrodes during anodic polarization decreased in the order of Pd >> Ni > Ni (SDC). In particular, the electrode-reaction resistance of the Ni (SDC) electrode, which was calculated from the slope of the polarization curve, was only 0.4 Ω cm<sup>-2</sup>.

Figure 2 also includes the polarization curves of the four perovskite oxide working electrodes under the same conditions. All these electrode potentials were a few millivolts negative under open-circuit conditions. As shown in Table I, the oxidation of methane proceeded at very slow rates at these electrodes, where a large amount of the unreacted oxygen was observed, suggesting that the following reaction determines the potentials of these electrodes



However, the cathodic overpotential of these electrodes was strongly dependent on the material: 50 Ω cm<sup>-2</sup> for La<sub>0.8</sub>Sr<sub>0.2</sub>MnO<sub>3</sub> > 12 Ω cm<sup>-2</sup> for LaNi<sub>0.6</sub>Fe<sub>0.4</sub>O<sub>3</sub> >> 0.4 Ω cm<sup>-2</sup> for La<sub>0.6</sub>Sr<sub>0.4</sub>CoO<sub>3</sub> > 0.2 Ω cm<sup>-2</sup> for Sm<sub>0.5</sub>Sr<sub>0.5</sub>CoO<sub>3</sub>. Based on the results, we concluded that the good performances of the Ni (SDC) anode and the Sm<sub>0.5</sub>Sr<sub>0.5</sub>CoO<sub>3</sub> cathode, due to their mixed ionic and electronic conduction, also apply to operation in a flowing mixture of methane and air. Therefore, we selected these electrodes for evaluating the performance of an SCFC in the subsequent experiments.

Figure 3 shows the discharge properties of the SCFCs (Fig. 1a) using the LSGM, SDC, and YSZ electrolytes in a flowing mixture of methane and air at 700°C. The cells using the LSGM and YSZ electrolytes generated electromotive forces (emf) of ~920 mV, where

**Table I. Compositions of inlet and outlet gases from working chamber under open-circuit conditions at 700°C.<sup>a</sup>**

Electrode	Inlet gas (mol %)		Outlet gas (mol %)					
	CH <sub>4</sub>	O <sub>2</sub>	CH <sub>4</sub>	O <sub>2</sub>	H <sub>2</sub>	CO	CO <sub>2</sub>	Other
Ni (SDC)	19.6	12.3	7.7	0.2	4.4	3.8	10.5	0
Ni	19.8	12.8	8.5	1.4	3	3.2	9.3	0
Pd	19.1	12.7	10	2.7	0.1	2.6	5.9	0.1
Pt	18.6	12.3	18	12.2	0	0	0.2	0
Sm <sub>0.5</sub> Sr <sub>0.5</sub> CoO <sub>3</sub>	19.3	12.2	20.3	12.2	0	0	0.1	0
La <sub>0.6</sub> Sr <sub>0.4</sub> CoO <sub>3</sub>	18.9	12.9	18.6	12.4	0	0	0.3	0
LaNi <sub>0.6</sub> Fe <sub>0.4</sub> O <sub>3</sub>	18.5	12.5	18.6	12.2	0	0	0.7	0
La <sub>0.8</sub> Sr <sub>0.2</sub> MnO <sub>3</sub>	20.4	12.3	20.2	11.8	0	0	0.1	0

<sup>a</sup> The balance gas is nitrogen.

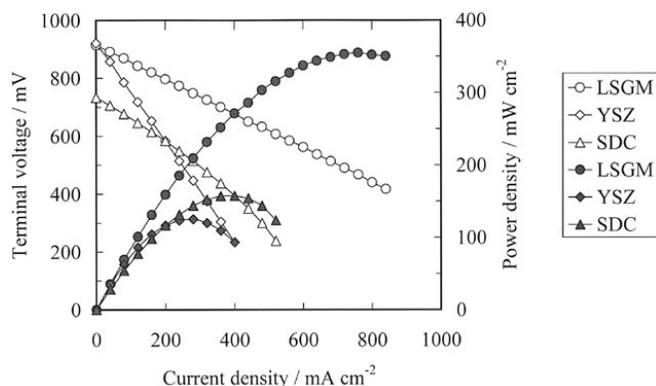
the potential of the Ni (SDC) electrode was negative vs. the Sm<sub>0.5</sub>Sr<sub>0.5</sub>CoO<sub>3</sub> electrode. This emf value was roughly in agreement with the difference in potential between the two electrodes shown in Fig. 2. The emf of the cell using the SDC electrolyte, however, was no more than 733 mV, probably due to a reduction of Ce<sup>4+</sup> to Ce<sup>3+</sup> by the hydrogen and carbon monoxide formed by Reactions 2 and 3. Although current could be reproducibly drawn from the three cells, the voltage drop during cell discharge was the least using the LSGM electrolyte. Their impedance spectra in the range of 0.1 Hz to 100 kHz under open-circuit conditions further clarified this point. The ohmic resistances of the cells using the LSGM, SDC, and YSZ electrolytes were 1.56, 1.72, and 2.90 Ω, respectively, and the electrode-reaction resistances for the LSGM, SDC, and YSZ electrolytes were 0.26, 0.53, and 1.22 Ω, respectively. The resulting peak power density of the cell using the LSGM electrolyte reached up to 355 mW cm<sup>-2</sup>, which is comparable to the peak power density of 385 mW cm<sup>-2</sup> for a conventional hydrogen-air SOFC, Ni-Ce<sub>0.9</sub>Gd<sub>0.1</sub>O<sub>1.95</sub> cermet/LSGM with a thickness of 0.2 mm/La<sub>0.6</sub>Sr<sub>0.4</sub>Fe<sub>0.8</sub>Co<sub>0.2</sub>O<sub>3</sub>, at 700°C reported by Huijismans *et al.*<sup>23</sup> Accordingly, we concluded that LSGM is the most suitable electrolyte for an SCFC.

Figure 4 shows the discharge properties of the SCFC (Fig. 1a) in a flowing mixture of methane and air between 550 and 900°C. There were two unexpected dependencies on the operating temperature; an abrupt drop of the emf to -28 mV at 550°C and a significant decrease in the peak power density at 900°C. In order to understand this behavior, the composition of the outlet gas from the Ni (SDC) or Sm<sub>0.5</sub>Sr<sub>0.5</sub>CoO<sub>3</sub> working chamber in the two-chamber cell (Fig. 1b) was measured at an open-circuit voltage between 550 and 950°C. The results are summarized in Fig. 5. In the range of 600 to 800°C, a relatively large amount of hydrogen and carbon monoxide was produced at the Ni (SDC) electrode (Fig. 5a), whereas most of the supplied oxygen remained unreacted at the Sm<sub>0.5</sub>Sr<sub>0.5</sub>CoO<sub>3</sub> elec-

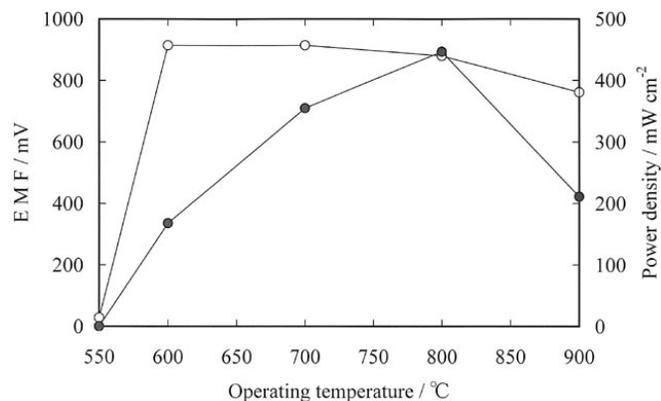
trode (Fig. 5b); therefore, the emf and the resulting power density behaved as shown in Fig. 4. At an operating temperature of 550°C, however, the Ni (SDC) electrode became abruptly inert toward Reactions 1-3 (Fig. 5a), indicating that it no longer functions as the anode. This is attributable to the very low reactivity of methane, especially at low temperatures. It is therefore necessary, for operation of the SCFC below 550°C, to use a more reactive hydrocarbon or alcohol than methane as the fuel. On the other hand, at operating temperatures above 900°C, the Sm<sub>0.5</sub>Sr<sub>0.5</sub>CoO<sub>3</sub> electrode gradually became active toward these reactions (Fig. 5b), thus causing a depression of Reaction 6. In a previous paper, an SCFC using the LSM cathode exhibited good performance even at 950°C.<sup>9</sup> It is well known that the catalytic activity of Mn-based perovskite oxides for the oxidation of hydrocarbons is lower than that of Co-based perovskite oxides,<sup>24</sup> thus making it possible for the LSM electrode to function as the cathode at such a temperature.

*Cell performance in flowing mixtures of other fuels and air.*—The discharge properties of the SCFC (Fig. 1a) were investigated in flowing mixtures of other fuels and air at 550°C. Figure 6 shows the results using ethane, propane, LPG, and ethanol as the fuels. The emf of the cell increased significantly to 1000 mV for ethane, 972 mV for propane, 931 mV for LPG, and 710 mV for ethanol, and the resulting peak power density reached 163 mW cm<sup>-2</sup> for ethane, 125 mW cm<sup>-2</sup> for propane, 94 mW cm<sup>-2</sup> for LPG, and 42 mW cm<sup>-2</sup> for ethanol. The impedance spectra of the cells using ethane and propane showed relatively small electrode-reaction resistances of 0.91 and 1.10 Ω, respectively. This suggests that the cell performance would be improved further using an even thinner LSGM film.

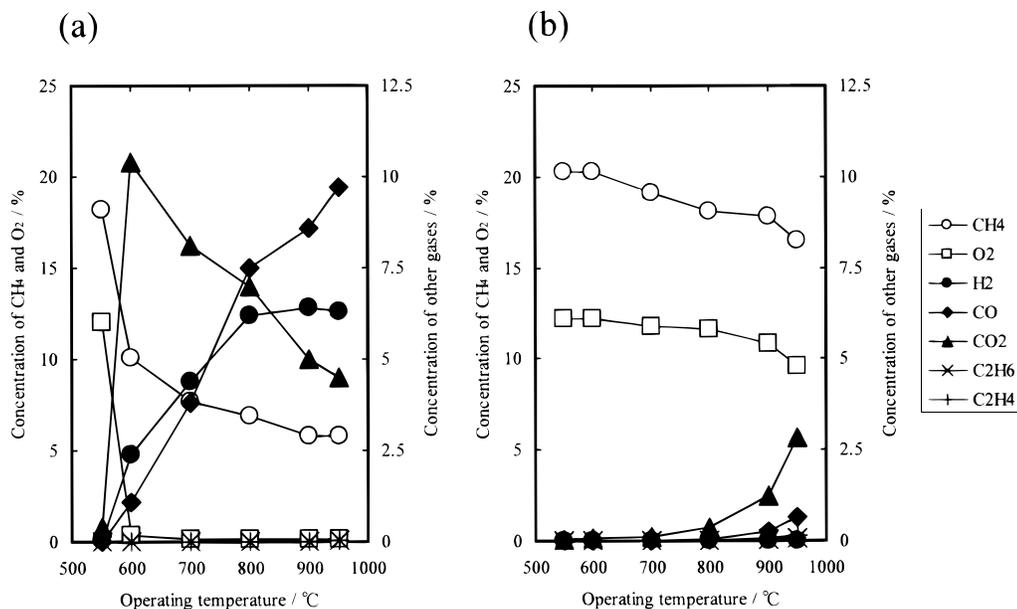
Evidence for this suggestion is provided by the discharge properties of the SCFC (Fig. 1a) using LSGM electrolytes of different thicknesses in a flowing mixture of ethane and air at 550°C shown in Fig. 7. The



**Figure 3.** Discharge properties of a single-chamber cell using LSGM, SDC, and YSZ electrolytes in a mixture of methane and air at 700°C: open symbol = terminal voltage; closed symbol = power density.

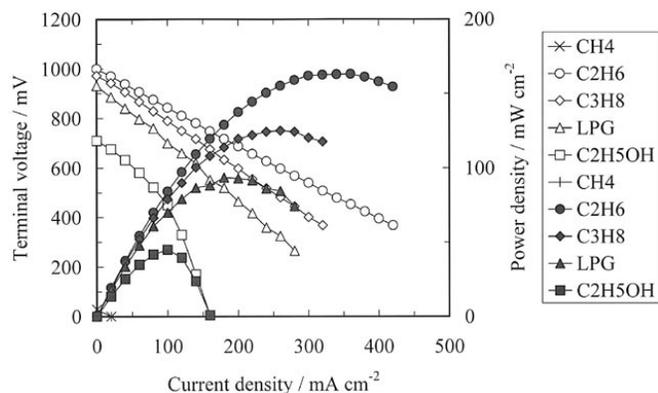


**Figure 4.** EMF and peak power density of a single-chamber cell in a mixture of methane and air at different temperatures. Open symbols indicate terminal voltage, closed symbols peak power density.

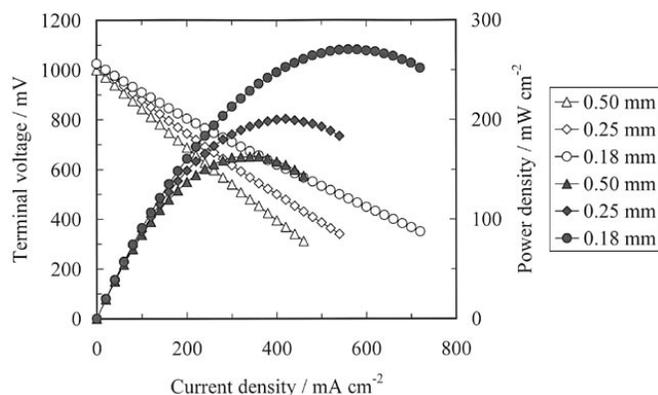


**Figure 5.** Compositions of outlet gases from (a) Ni (SDC) and (b)  $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$  working electrodes in a two-chamber cell at different temperatures; methane  $52 \text{ mL min}^{-1}$ ; oxygen  $52 \text{ mL min}^{-1}$ ; nitrogen  $196 \text{ mL min}^{-1}$ .

emf of the cell remained at almost constant values of  $\sim 1010 \text{ mV}$  for any tested electrolyte thickness, but the voltage drop during cell discharge was strongly dependent on the electrolyte thickness. As a result, the peak power density increased from  $163$  to  $270 \text{ mW cm}^{-2}$  with decreasing electrolyte thickness from  $0.50$  to  $0.18 \text{ mm}$ .



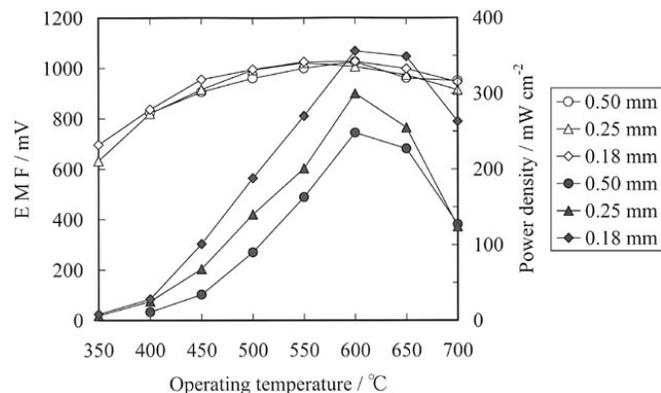
**Figure 6.** Discharge properties of a single-chamber cell in a mixture of other fuels and air at  $550^\circ\text{C}$ . Open symbols indicate terminal voltage, closed symbols power density.



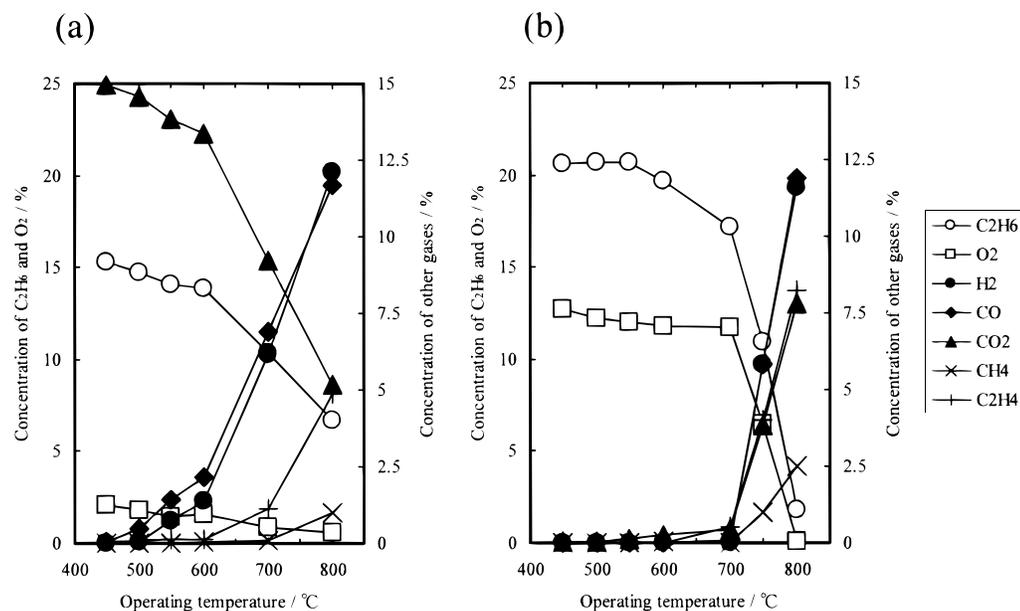
**Figure 7.** Discharge properties of a single-chamber cell using different thicknesses of LSGM electrolyte in a mixture of ethane and air at  $550^\circ\text{C}$ . Open symbols indicate terminal voltage, closed symbols power density.

Figure 8 shows the discharge properties of the same SCFC as in Fig. 7 in a flowing mixture of ethane and air between  $350$  and  $700^\circ\text{C}$ . All these cells exhibited maxima in peak power density at  $600^\circ\text{C}$ , which was lower than that observed using methane by  $200^\circ\text{C}$ . This is discussed later in detail. More important was the increasing peak power density with decreasing electrolyte thickness even at a temperature as low as  $450^\circ\text{C}$ , suggesting that the present SCFC may be utilized as a practical and efficient power generator under such conditions, provided that a several micrometer thick LSGM film can be fabricated in a manner similar to  $\text{YSZ}^{1,3}$  and  $\text{ceria}^2$  films. For example, a peak power density of *ca.*  $300 \text{ mW cm}^{-2}$  would be obtained using a  $10 \mu\text{m}$  thick electrolyte, assuming that the emf is maintained at  $955 \text{ mV}$ .

The compositions of the outlet gases from the Ni (SDC) and  $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$  working chambers (Fig. 1b) are summarized in Fig. 9. The catalytic activity of the  $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$  electrode increased abruptly above  $700^\circ\text{C}$ , where oxygen was almost completely consumed by reacting with ethane. A decrease in the emf and an increase in the cathodic overpotential would therefore occur simultaneously under such conditions, thereby resulting in the significant decrease in power density shown in Fig. 8. On the other hand, the catalytic activity of the Ni (SDC) electrode gradually became lower as the temperature decreased. From the behavior of the cell using methane as the fuel shown in Fig. 4, one can predict that a cell using



**Figure 8.** EMF and peak power density of a single-chamber cell in a mixture of ethane and air at different temperatures. Open symbols indicate terminal voltage, closed symbols peak power density.



**Figure 9.** Compositions of outlet gases from (a) Ni (SDC) and (b) Sm<sub>0.5</sub>Sr<sub>0.5</sub>CoO<sub>3</sub> working electrodes in a two-chamber cell at different temperatures: ethane 52 mL min<sup>-1</sup>; oxygen 52 mL min<sup>-1</sup>; and nitrogen 196 mL min<sup>-1</sup>.

ethane as the fuel would be unable to generate a large emf below 350°C. However, one can also expect that propane and LPG would be preferred fuels under such conditions because of their higher reactivities than those of ethane and methane.

### Conclusions

Conventional SOFC have serious problems for transportation applications due to their operation at high temperatures and their low thermal and mechanical shock resistance. These problems could be solved to a considerable extent using a highly conductive solid electrolyte, such as LSGM, together with a single-chamber cell design, where there is no need to separate the supply of fuel and air. An important feature of this approach is the use of readily available hydrocarbons, such as ethane, propane, and LPG, as fuels. The fabrication of a thin electrolyte film would further enhance the position of the present SOFC as the preferred technology as a power source for transportation.

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