Decreased operating temperature of solid oxide fuel cells (SOFCs) by the application of $LaGaO_3$-based oxide as electrolyte

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Application of the new oxide ion conductor of composition $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_3$ as electrolyte in solid oxide fuel cells is effective in enabling a decrease in the operating temperature.

Stabilized zirconia, particularly Y_2O_3-stabilized ZrO_2 (YSZ), is the most common electrolyte used in SOFCs.[1] However, the oxide-ion conductivity of YSZ is fairly low and, therefore, a thin electrolyte film without gas leakage, and an excessively high operating temperature (1273 K), are essential to obtain high power densities of SOFCs where YSZ is used as the electrolyte.[1] On the other hand, one of the current concerns for the development of SOFCs, is to increase the reliability of cells as electric power generation systems. A decrease in the operating temperature and use of a self supporting thick sheet for the electrolyte is the most effective method for solving these problems. Consequently, a new oxide ion conductor with high electrical conductivity is strongly required in the development of SOFCs for practical application. In our previous study, it was found that the $LaGaO_3$-based perovskite type oxide exhibits an extremely high oxide ion conductivity.[2] The application of $LaGaO_3$-based perovskite type oxide as the electrolyte of SOFCs was investigated in the present study with the aim of decreasing the operating temperature.

The perovskite type oxide of composition $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_3$, as denoted $LaGaO_3$-based oxide, was prepared according to the method reported previously.[2] Y_2O_3-stabilized ZrO_2 disks were prepared by sintering the commercial YSZ powder (Tosoh TZ-8Y) at 1773 K for 6 h. The obtained $LaGaO_3$-based oxide and YSZ disks were polished with diamond wheels to 0.5 mm in thickness. $LaCoO_3$ and $LaMnO_3$-based perovskite type oxides for the cathode were prepared by calcining the precursors at 1473 K for 6 h. The precursors of these oxides were prepared by evaporation to dryness of acetate solutions. The thus obtained cathode oxide and metal anode (commercial metal powders were used without further purification), were applied on opposite faces of $LaGaO_3$ or YSZ disks (5 mm in diameter) followed by calcining at 1173 K for 10 min. Electric power generation characteristics of single SOFCs were measured by the four-probe method using humidified hydrogen (2 vol% H_2O) as a fuel and oxygen as an oxidizing agent. It is also noted that the theoretical electromotive force estimated from the Nernst equation is 1.10 V under the present conditions.

Table 1 summarizes the effects of electrode material on the electrical power generation characteristics of SOFCs where $LaGaO_3$-based oxide was used as the electrolyte. Since hole conduction occurred to a slight extent in the $LaGaO_3$ oxide in the high oxygen partial pressure range, the open-circuit voltage of SOFCs with $LaGaO_3$-based oxide used as electrolyte was slightly lower than that estimated by the Nernst equation. However, the open-circuit voltage attained values of almost 1.10 V with every SOFC examined (Table 1). This suggests that the transference number of the oxide ions of $LaGaO_3$-based oxides was satisfactorily high in the electrolytes of the SOFCs. On the other hand, the maximum power density strongly depended on the electrode materials used (cathode and anode). The maximum power density of cells increased in the following order, Pt < $La_{0.6}Sr_{0.4}MnO_3$ < $La_{0.6}Sr_{0.4}CoO_3$ for the cathode, and Ru < Pt < Co < Ni for the anode. Although the maximum power density of the cells, where platinum was used for both cathode and anode, was as low as 0.09 W cm^{-2}, it attained a value as high as 0.71 W cm^{-2} in the cell where $La_{0.6}Sr_{0.4}CoO_3$ and Ni were used for the cathode and anode respectively. It is reported that a $LaCoO_3$ = based oxide exhibits high activity for the dissociation of oxygen molecules to oxygen ions; however, it could not be used as a cathode in a conventional cell due to its high reactivity with YSZ.[3] XRD analysis suggests that no reaction between $LaCoO_3$ and $LaGaO_3$-based oxide can be observed up to 1473 K. On the other hand, reaction between Ni and $LaGaO_3$-based perovskite type oxide was not evident by XRD in the reducing atmosphere.

Fig. 1 shows the maximum power density of the SOFCs as a function of x for $La_{1-x}Sr_xCoO_3$ cathode at 1273 K. The electrical conductivity of $LaCoO_3$ increased with increasing x,[4] and similarly for $La_{1-x}CoO_3$ the maximum conductivity also

Table 1 Effects of electrode materials on the electrical generation characteristics of SOFCs operated at 1273 K

Anode	Cathode	OCV/V	MPD/W cm^{-2}
Pt	Pt	1.098	0.09
Ni	Pt	1.099	0.15
Ni	LSM	1.098	0.31
Ni	LSC	1.099	0.71
Ru	LSC	1.051	0.01
Co	LSC	1.100	0.53

OCV open circuit voltage; MPD maximum power density; LSM, $(La_{0.6}Sr_{0.4})_{0.9}MnO_3$; LSC, $La_{0.6}Sr_{0.4}CoO_3$. Electrolyte; $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_3$ (0.5 mm thickness).

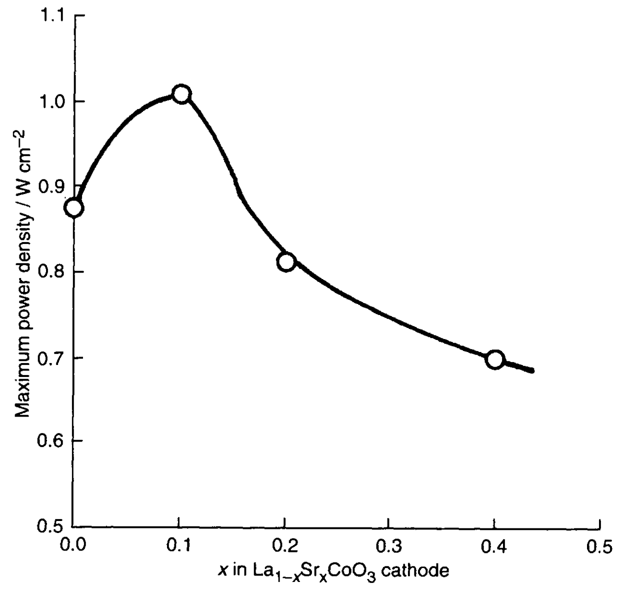

Fig. 1 Maximum power densities of the SOFCs as a function of x in the $La_{1-x}Sr_xCoO_3$ cathode at 1273 K

increased with increasing x and attained a maximum at $x = 0.1$. For the SOFC consisting of $La_{0.9}Sr_{0.1}Ga_{0.2}Mg_{0.2}O_3$, Ni and $La_{0.9}Sr_{0.1}CoO_3$ as the electrolyte, anode and cathode, respectively, the maximum power density was as high as 1.01 W cm^{-2} at 1273 K despite the low thickness (0.5 mm) of the electrolyte plate.

Fig. 2 shows the temperature dependence of the maximum power density of the SOFC consisting of $LaGaO_3$-based oxide and YSZ for H_2–O_2 and that of the $LaGaO_3$-based oxide in H_2–air. Although the maximum power densities of cells are not directly comparable, the cell performance of YSZ in Fig. 2 is typical[1] for H_2–O_2 cells. While the power density and the current density at short circuit decreased with decreasing operating temperature on each cell, these parameters for SOFCs containing $LaGaO_3$-based oxide electrolyte (with O_2 as oxidant) still attained values of 0.17 W cm^{-2} and 0.7 A cm^{-2}, respectively, at an operating temperature as low as 973 K. In agreement with the lower apparent activation energy for the electrical conductivity of $LaGaO_3$-based oxide relative to YSZ, the difference between the maximum power density of cells based on $LaGaO_3$ and YSZ increased with decreasing operating temperature. The attained maximum power density of cells with $LaGaO_3$-based oxide electrolyte is three times larger than that of YSZ at 1273 K, and is an order of magnitude larger at 973 K. Moreover, the electrical power can be generated in these SOFCs even at 873 K. Considering the thickness (0.5 mm) of the electrolyte and comparison of the power generation characteristics of cells of YSZ electrolyte, it is clear that the power generation characteristics of the $LaGaO_3$-based oxide cells were extremely high. Furthermore, except at 1273 K, decreases in the power density of cells of $LaGaO_3$-based oxide were negligibly small on changing the oxidant from oxygen to air. High power density of SOFC at low temperature seems to be due to the high oxide ion conductivity of $LaGaO_3$-based perovskite and the high activity of $La_{0.9}Sr_{0.1}CoO_3$ cathode for the dissociation of oxygen molecules to oxide ions.

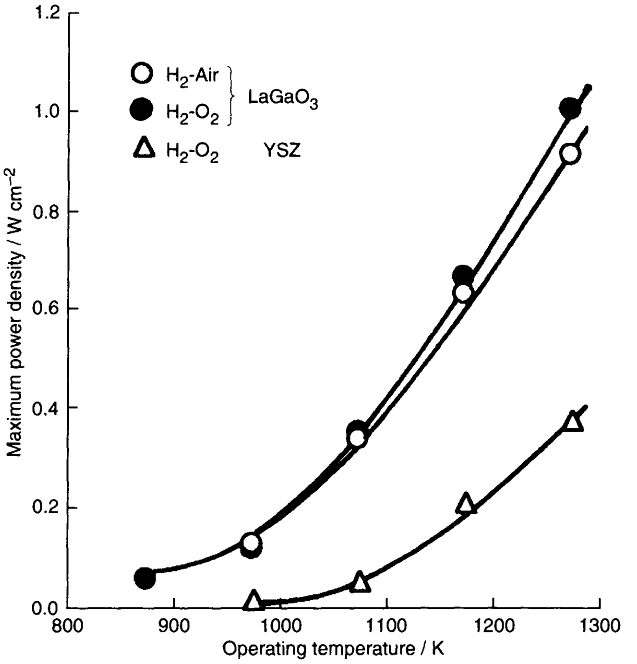

Fig. 2 Maximum power densities of SOFCs, where $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_3$ and YSZ were used as electrolyte, as a function of operating temperature

In conclusion, the new $LaGaO_3$-based perovskite type oxide conductor is a promising material to facilitate a decrease of operating temperatures of SOFCs.

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