Application of Ce-based oxides with high-sintering characteristics to electrolytes in intermediate-temperature Solid Oxide Fuel Cells

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From the viewpoint of the quick start, the use of metallic cell components, and the provision of higher terminal voltage, there is a need to investigate intermediate-temperature solid oxide fuel cells (IT-SOFC) in order to apply the IT-SOFC to the main driving source and the auxiliary power unit. Ceria-based oxides have been suggested as an alternative electrolyte in the SOFC to stabilized zirconia.

The low-temperature sinterable $Ce_{1-x}RE_xO_{2-\delta}$ powders (RE = Y, Sm, Gd) have been developed using a newly-devised heat-treatment process in the coprecipitation method [1,2]. Especially, the $Ce_{0.9}Y_{0.1}O_{1.95}$ and $Ce_{0.9}Gd_{0.1}O_{1.95}$ powders show the lowest value in the reported sintering temperatures : A relative density $\geq$ 94%, where sintered ceramics can provide gastightness, is attained for these powders after heating at temperatures $\geq$ 1050°C for 2 h.

It is well known that ceria-based oxides show the formation of oxygen vacancies in the fluorite structure and a charge compensation of the Ce^{4+} to the Ce^{3+} ions occurs in reducing atmosphere at temperatures greater than ~550°C. These cause the appearance of electronic conduction and isothermal expansion under reducing atmosphere at temperatures $\geq$ 500°C.

Figure 1 shows the open circuit voltage (OCV) of the H_2-O_2 fuel cell using the dense $Ce_{0.9}Gd_{0.1}O_{1.95}$ electrolyte and oxide-ionic transport number as a function of temperature. The dotted and broken lines show the theoretical OCV and oxide-ionic transport number, respectively. With increasing temperature, observed OCV trends to decreasing, compared to those of the theoretical ones. This might be due to an electronic conduction in the electrolyte.

Figure 2 shows the oxygen contents of $Ce_{1-x}Y_xO_{2-\delta}$ in a H_2 atmosphere, when the oxygen contents of the fluorites are assumed to be zero at 25°C. With increasing temperature, an oxygen release caused by the reduction of Ce^{4+} into Ce^{3+} ions in the fluorite structure is observed.

Figure 3 shows the differential coefficient of thermal expansion-temperature curve of $Ce_{1-x}Sm_xO_{2-\delta}$, where the values are calculated using thermal expansion slops between 2.5°C above and 2.5°C below a give temperature. It is seen that thermal expansion coefficients in the H_2 atmosphere increased remarkably at around 500 - 600°C.

The problems mentioned above are the major technology barrier to the development of IT-SOFC using ceria electrolyte. The aim of this study is to develop Ce-based oxides with high sintering characteristics, and without electronic conductivity and isothermal expansion at temperatures $\leq$ 600°C under reducing atmospheres.

References

1) E. Suda, B. Pacaud, Y. Montardi, M. Mori, M. Ozawa and Y. Takeda, *Electrochemistry*, **71** (10), p.866 (2003).
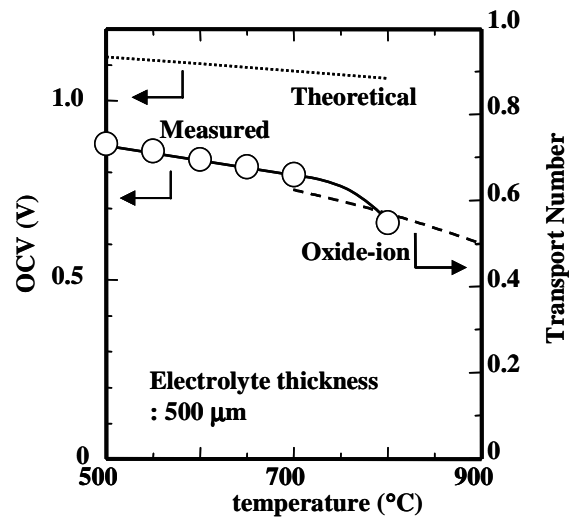2) E. Suda, B. Pacaud, Y. Montardi, M. Mori, and Y. Takeda, *J. IUMRS*, Published (2004).

Fig. 1 OCV of H_2-air fuel cell with $Ce_{0.9}Gd_{0.1}O_{1.95}$ electrolyte, and oxide-ionic transport number for $Ce_{0.9}Gd_{0.1}O_{1.95}$. H_2 gas supplied after bubbling through pure water at 10°C.
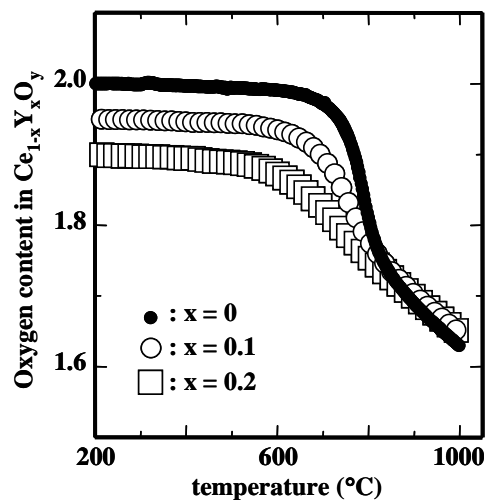

Fig. 2 Oxygen content in $Ce_{1-x}Y_xO_y$ in the H_2 atmosphere as a function of temperature.
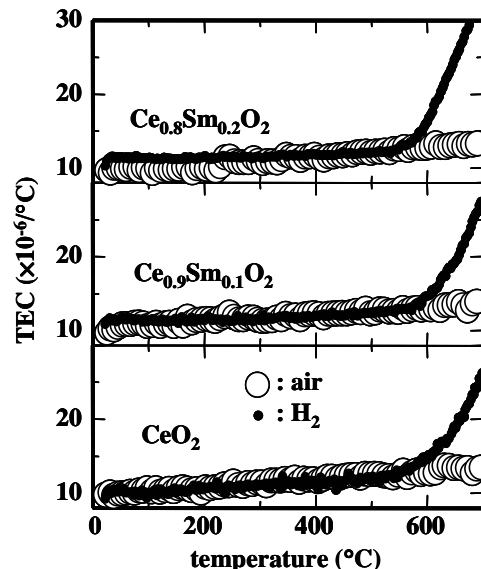

Fig. 3 Differential coefficient of thermal expansion-temperature curve of $Ce_{1-x}Sm_xO_{2-\delta}$. H_2 gas was supplied after bubbling in pure-water at 10°C.