and volumetric power density (W/cm^3) low; higher power density alternate tubular geometry cells are now being conceived and developed. Planar SOFCs are capable of achieving very high power densities, but require high temperature seals which is a big factor in their performance stability and useful lifetime. When fully developed and are cost-competitive, SOFCs will have widespread application in the stationary distributed power generation, and transportation market sectors. Systems based on both the tubular and the planar SOFCs are ideal power generation systems-reliable, clean, quiet, environmental friendly, and fuel conserving.

Acknowledgements

The author would like to thank his many colleagues in the SOFC community whose work is reviewed in this paper. The Pacific Northwest National Laboratory is operated by Battelle Memorial Institute for the U. S. Department of Energy under Contract No. DE-AC06-76RLO 1830.

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Journal of the Korean Ceramic Society
Vol. 42, No. 12, pp. 787~792, 2005.

Electrochemical Performance of LSCF Cathode with GDC Interlayer on ScSZ Electrolyte

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(Received November 1, 2005; Accepted November 7, 2005)

ABSTRACT

A symmetrical LSCF ($La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$)/ScSZ ($89ZrO_2$-$10Sc_2O_3$-$1CeO_2$)/LSCF electrochemical cell with a GDC (Gadolinium-Doped Ceria, $90CeO_2$-$10Gd_2O_3$) interlayer that was inserted between the LSCF cathode and ScSZ electrolyte was fabricated, and the electrochemical performance of these cells was evaluated. The GDC interlayer was deposited on a ScSZ electrolyte using a screen-printing technique. The GDC interlayer prevented the unfavorable solid-state reactions at the LSCF/ScSZ interfaces. The LSCF cathode on the GDC interlayer had excellent electrocatalytic performance even at 650°C. The Area Specific Resistance (ASR) was strongly dependent on the thickness and heat-treatment temperature of the GDC interlayer. The impedance spectra showed that the cell with a 15~27 μm thick GDC interlayer heat-treated at 1200°C had the lowest ASR.

Key words : SOFCs, LSCF, ScSZ, Buffer layer, Impedance spectrum

1. Introduction

Recently, there has been considerable interest in the field of Solid Oxide Fuel Cells (SOFCs). The distinct advantages of SOFCs over other fuel cells is that there is no need for noble metal catalysts such as platinum, all the components are solids, which offers considerable cell design flexibility and eliminates material corrosion problems, and there is the potential for cogeneration due to their high temperature operation.[1,2] However, high temperature operation at around 800~1000°C is the main disadvantage of SOFCs because of the low long-term reliability, the requirement for lanthanum chromite-based ceramic interconnectors, which are quite expensive and difficult to fabricate and machine, high temperature interdiffuion between the constituent elements, difficulties in sealing the SOFC stack, and the slow start-up response. These problems can be partly solved by lowering the operating temperature to 500~700°C.[3-5] However, both the electrocatalytic activity over oxygen reduction at the cathode and fuel (hydrogen) oxidation at the anode as well as the ionic conductivity of the electrolyte decrease significantly with decreasing SOFC operating temperature. Therefore, considerable effort has been made to improve the performance of the SOFC materials.

YSZ is widely used as a solid electrolyte material in SOFCs on account of its excellent mechanical, thermal, and chemical stability at high temperature. In order to decrease the cell resistance and the operating temperature, solid electrolytes with a higher ionic conductivity than YSZ in an intermediate temperature range need to be developed. Scandia-Stabilized Zirconia (ScSZ) is a candidate for intermediate temperature applications because it has a higher ionic conductivity and mechanical strength than YSZ.

LSCF ($La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$) is an electrode material for intermediate temperature applications owing to its high mixed conductivity, and excellent thermal and chemical compatibility to ceria-based electrolyte materials such as GDC, SDC etc.[6,7] On the other hand, the LSCF reacts easily with ZrO_2 to produce an interfacial-insulating layer ($La_2Zr_2O_7$ or $SrZrO_3$), which decreases the catalytic activity of the electrodes.[8,9] CeO_2-based electrolytes are chemically compatible with LSCF but have several disadvantages such as low mechanical strength and are reduced in the fuel atmosphere.

This study fabricated LSCF/ScSZ/LSCF electrochemical cells with a GDC interlayer between the ScSZ electrolyte and LSCF electrode for use in Intermediate Temperature (IT) SOFCs. The influence of the interlayer thickness and the heat-treatment temperature of the GDC interlayer on the electrochemical performance of the LSCF cathode formed on the ScSZ electrolyte were examined.

2. Experimental Procedure

Two types of symmetrical electrochemical cells were fabricated, LSCF/ScSZ/LSCF and LSCF/GDC (interlayer)/ScSZ/GDC (interlayer)/LSCF. Fig. 1 shows a schematic diagram of the cells. The GDC interlayers were deposited using a screen-printing technique on both sides of the ScSZ (89 mole % ZrO_2-10 mole % Sc_2O_3-1 mole % CeO_2, Japan Fine Ceram-

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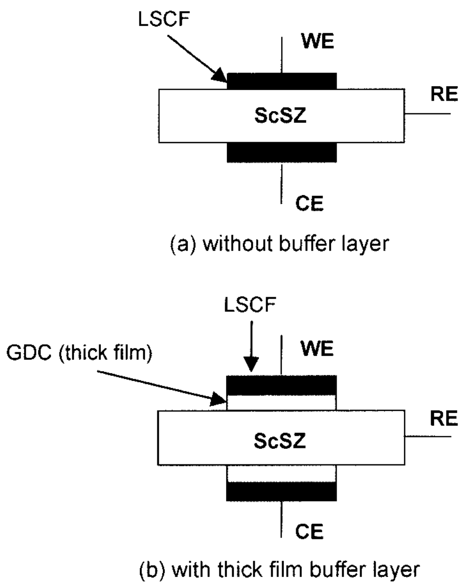

Fig. 1. Schematic diagrams of the two types of electrochemical cells used in this study.

ics Co. Ltd., Sendai, Japan) disk before applying the LSCF electrodes. The ScSZ disk was prepared by firing a green ScSZ compact at 1400°C for 3 h in air. The dimensions of the sintered ScSZ disk were 10.6 mm in diameter and 1.0 mm in thickness.

A GDC paste was prepared by mixing a commercially available GDC powder (Anan Kasei Co. Ltd., Tokyo, Japan) with an organic vehicle. The green GDC films were screen-printed onto both sides of the ScSZ electrolyte disk and heat-treated at 1000, 1100, 1200, 1300, and 1350°C for 2 h in air. The area of the GDC interlayer was 0.79 cm^2. A cathode layer of the LSCF was also deposited by screen-printing a LSCF paste, which was prepared by mixing a LSCF powder ($La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$, Japan Fine Ceramics Co. Ltd., Sendai, Japan) and an organic vehicle, on the symmetrical GDC/ScSZ/GDC cell, and heat-treating it at 1000°C for 2 h in air. The area of the LSCF cathode was 0.50 cm^2.

The performance of the LSCF cathode was examined using a three-probe AC complex impedance measurement with a steady DC current. A Pt reference electrode was prepared on the side surface of the ScSZ electrolyte disk in order to complete the three-probe measurement. The reference electrode was exposed to flowing air. AC impedance spectroscopy was measured using a frequency analyzer (Model SI 2160/1287 electrochemical interface, Solartron,

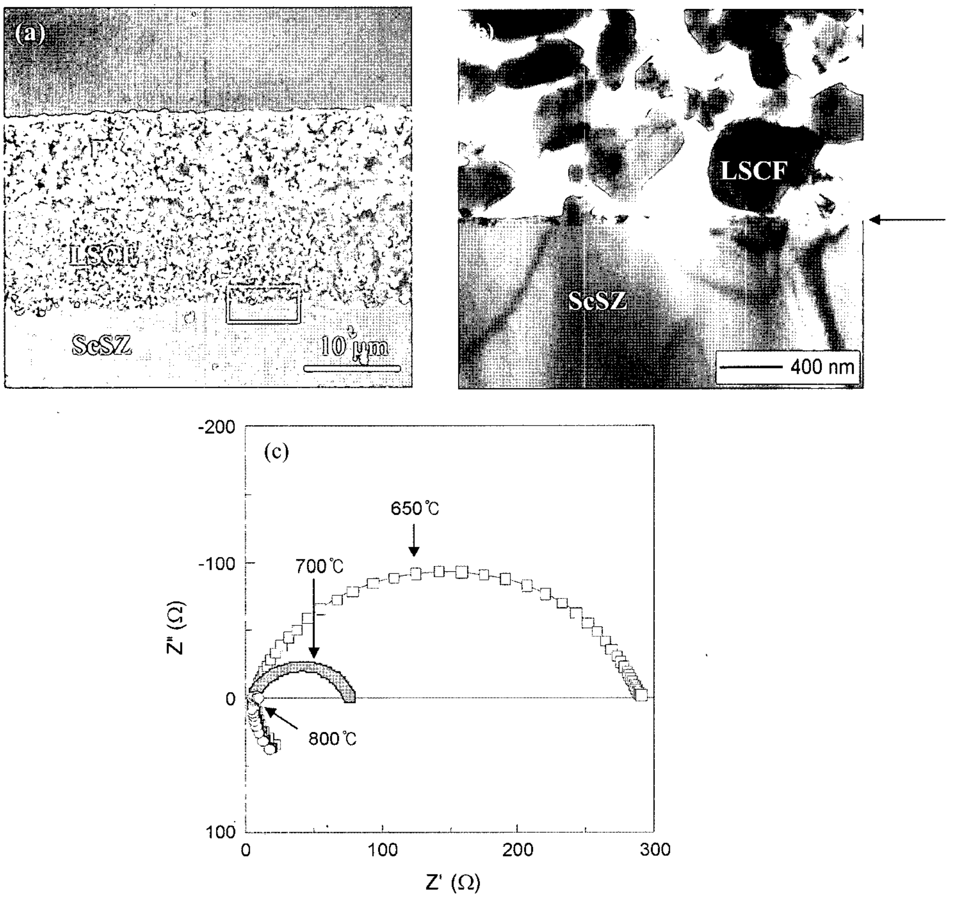

Fig. 2. SEM images showing a cross-section of the LSCF/ScSZ/LSCF cell: low magnification (a) and high magnification (b) and AC impedance spectra of LSCF/ScSZ/LSCF cell.

Farnborough, UK) in an air atmosphere at 650, 700, and 800°C. The frequency range was 1 MHz to 0.01 Hz. The amplitude of the signal to the electrochemical cells was 20 mV.

The microstructures of the GDC buffer layers and the interfaces between the LSCF and ScSZ were observed by scanning electron microscopy (SEM, JSM-6320FK, JEOL Co. Ltd., Tokyo, Japan) and transmission electron microscopy (TEM, Hitachi H-9000UHR III, Hitachi Co. Ltd., Tokyo, Japan), respectively. The crystalline phase was determined by X-Ray Diffraction (XRD, RU-200B, Rigaku Co. Ltd., Tokyo, Japan) using Ni-filtered CuKα radiation.

3. Results and Discussion

Fig. 2 shows the SEM and TEM images of the cross-section and AC impedance spectra of the LCF/ScSZ/LSCF cell measured at 650, 700, and 800°C in air. The polarization resistance of the LSCF cathode increased with decreasing the cell operating temperature. For example, the total resistance of the cell without the GDC interlayer was approximately 300 $\Omega \cdot cm^2$ at 650°C. This supports the presence of an interfacial-insulating phase at the ScSZ/LSCF interface, which significantly reduces the area of the triple-phase boundary, LSCF/ScSZ/O_2 in the cathode. Fig. 2(b) shows presence of an interfacial layer, demonstrating that the thin layer with a thickness of approximately 30 nm formed on the ScSZ electrolyte. On the other hand, the insulating phase does not appear to have caused an increase in the ohmic resistance of the cell because the layer is sufficiently thin i.e. the ohmic resistance of the LSCF/ScSZ/LSCF cell was very small (<3.2 $W \cdot cm^2$).

The formation of an interfacial insulation phase was also confirmed by XRD. Fig. 3 shows the XRD profiles taken from the LSCF-ScSZ powder mixtures that were heat-treated at various temperatures. Even at 800°C, an interfacial phase exists, and an increase in the heat-treatment temperature resulted in the development of more interfacial phases, which consisted mainly of $SrZrO_3$ and $La_2Zr_2O_7$ phases.[9]

Fig. 4 shows cross-section SEM images of the LCF/ScSZ/LSCF cell with the GDC interlayer. GDC thick films were

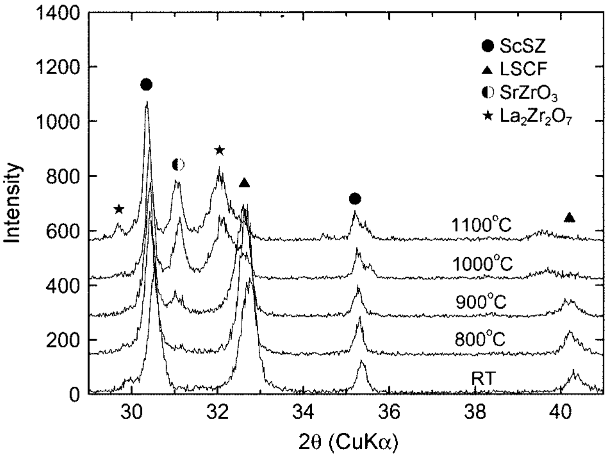

Fig. 3. XRD patterns of the LSCF-ScSZ powder mixtures heat-treated at various temperatures.

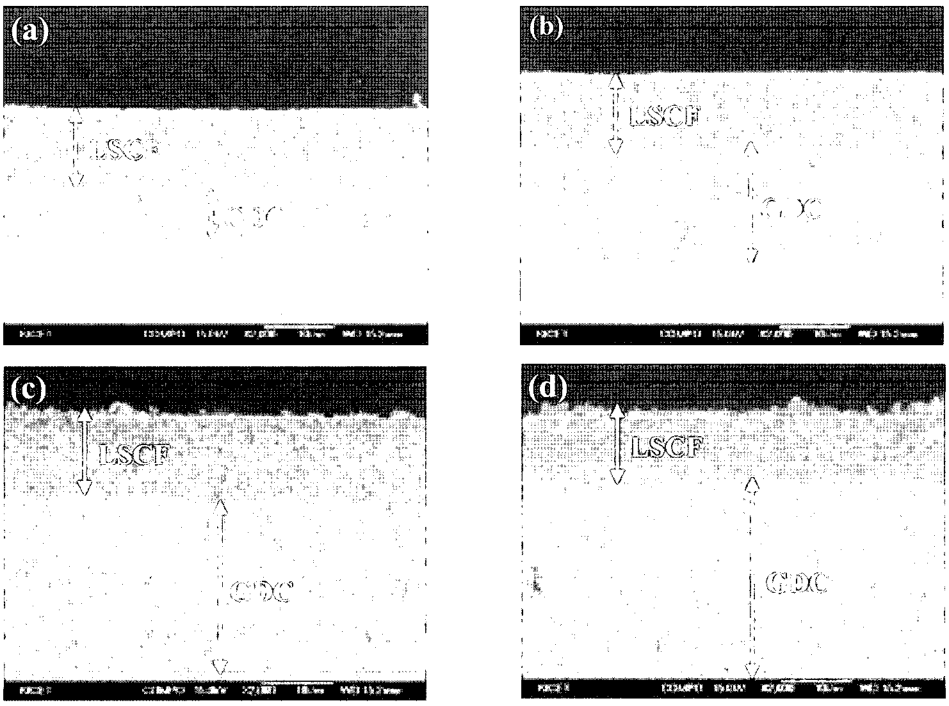

Fig. 4. SEM images showing the cross-section of the LSCF/ScSZ/LSCF cell with a GDC buffer layer of thickness; (a) 7 μm, (b) 15 μm, (c) 25 μm, and (d) 28 μm.

successfully deposited on the ScSZ electrolyte. The GDC interlayer and ScSZ electrolyte as well as the LSCF cathode and GDC interlayer exhibited good interfacial contact, and the GDC adhered well to the electrolyte with no signs of delamination. The thickness of the GDC interlayer could be increased from 7 μm (a) to 28 μm (d) by repeating the screen-printing process. The LSCF cathode was sufficiently porous and had a fine microstructure.

Several cells with different GDC film thicknesses were fabricated to determine the effect of the GDC interlayer thickness on the cathode performance of the LSCF/ScSZ/LSCF cells with the interlayer film. Fig. 5 shows the impedance spectrum of the LSCF/ScSZ/LSCF cells with the GDC buffer layer at 700°C. The GDC interlayer was heat-treated at 1200°C for 2 h. The CeO_2-based electrolyte is a potential candidate as a diffusion preventing interlayer because it has no constitution element that can react with the LSCF cathode, and shows excellent oxygen ion conductivity at the intermediate temperature range. A comparison of Fig. 5 with Fig. 2 shows that the polarization resistance of the LSCF cathode with the GDC interlayer decreased significantly, indicating that the GDC interlayer plays an important role in blocking the unwanted inter-diffusion between the LSCF and ScSZ electrolyte.

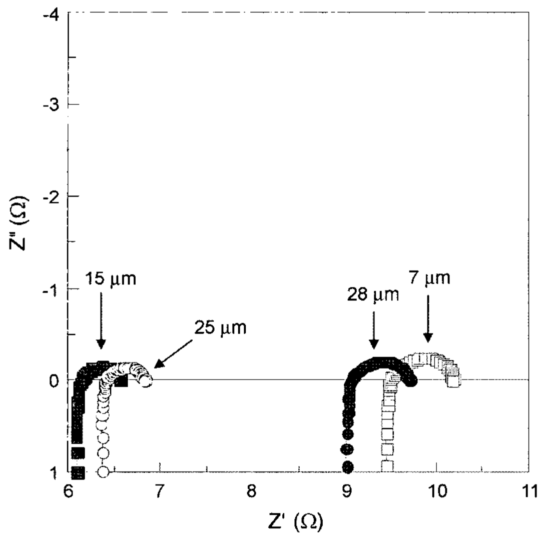

Fig. 5. AC impedance spectra of LSCF/ScSZ/LSCF cells with a GDC buffer layer. The operating temperature is 700°C.

As shown in Fig. 5, the LSCF/ScSZ/LSCF cells with a 15 and 25 μm GDC interlayer showed better cathode performance, particularly the ohmic resistance, than the cells with the 7 and 28 μm interlayer. This means that a GDC layer <15 μm is not sufficient to block the interfacial reaction between the LSCF and ScSZ. Since the ionic conductivity of the ScSZ is approximately 0.04 S/cm at 700°C, the observed ohmic resistances (6 to 6.5 Ω) of the cell with the 15 and 25 μm GDC interlayer is consistent with the reported values. It appears that the contribution of the GDC interlayer to the total ohmic resistance of the cell is low because the GDC interlayer was thin compared with the ScSZ electrolyte (approximately 1 mm). Therefore, it is unclear why the cells with a 28 μm GDC interlayer showed higher ohmic resistance than the other cells.

Fig. 6 shows SEM images of surfaces of the GDC interlayer that had been heat-treated at different temperatures.

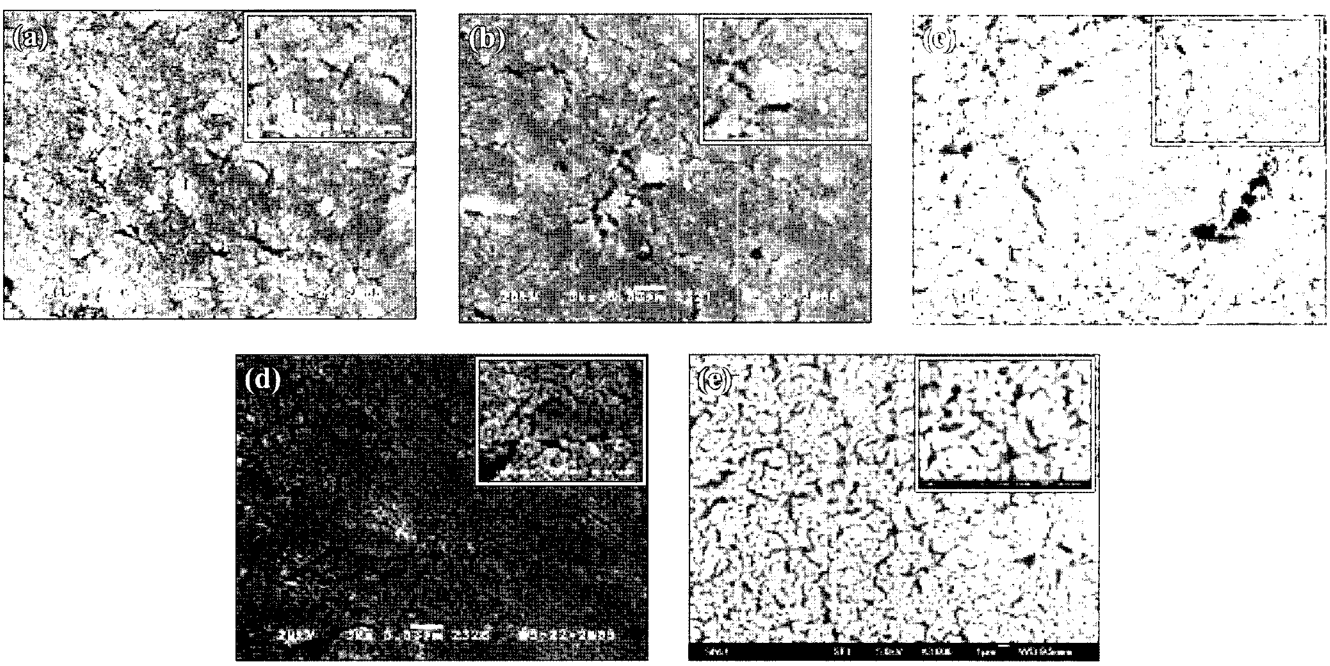

Fig. 6. SEM photographs showing the microstructure of a GDC buffer layer of the LSCF/ScSZ/LSCF cells heat-treated at (a) 1000°C, (b) 1100°C, (c) 1200°C, (d) 1300°C, and (e) 1350°C for 2 h.

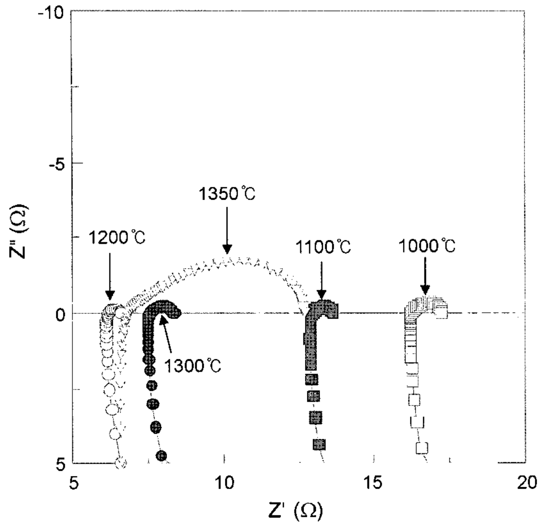

Fig. 7. AC impedance spectra of LSCF/ScSZ/LSCF cells with a GDC buffer layer heat-treated at different temperatures. The operating temperature was 700°C.

The GDC interlayer was not dense and there were some cracks and voids in the microstructure. However, it appears that such inhomogeneities did not significantly deteriorate the performance of the cells. As the heat-treatment temperature was increased, the GDC particles sintered coalesced and the size of the GDC particles and intergranular pores increased gradually. The grain size of the GDC was approximately 200 and 500 nm in the1200°C- and 1350°C-sintered samples, respectively.

Fig. 7 shows the AC impedance spectra of the LSCF/ScSZ/LSCF cells with the GDC interlayer sintered at different temperatures. The LSCF cathode was sintered at 1000°C for 2 h. Generally, the intercept of the impedance semicircle with the real axis at high frequencies (the left intercept of the semicircle) corresponds to the ohmic resistance (R_{ohmic}) of the electrochemical cell, which mainly includes the bulk resistance of the electrolyte and the contact resistance between the electrolyte and the electrode. On the other hand, the intercept of the semicircle with the real axis at low frequencies (right intercept of the semicircle) indicates the total resistance (R_{total}) of the electrochemical cell. The polarization resistance or reaction resistance (R_{pol}) was determined by subtracting the ohmic resistance from the total resistance.

The polarization resistance of the LSCF/ScSZ/LSCF cells with the GDC interlayer that had been sintered at 1000 to 1300°C was similar. In addition, the ohmic resistance decreased with increasing GDC sintering temperature. This means that the electrochemical performance is improved by sintering the GDC interlayer at higher temperatures but <1350°C. The high polarization resistance of the 1350°C-sintered sample appears to be due to the interfacial reaction between the GDC and ScSZ. Some reports have suggested an interdiffusion phenomenon of Zr^{4+} and/or Sc^{3+} in ScSZ and GDC.[10-12] They confirmed that ZrO_2-Sc_2O_3-CeO_2-$GdO_{1.5}$ solid solutions were formed at the ScSZ/GDC interfaces.

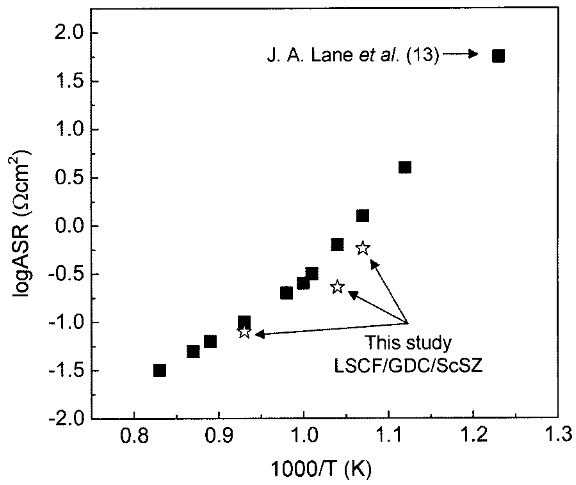

Fig. 8. Area Specific Resistances (ASR) of the LSCF/ScSZ/LSCF cells with a GDC interlayer.

However, the ohmic resistance decreased with increasing sintering temperature up to 1200°C and became saturated or slightly increased thereafter. As mentioned above, the ohmic resistance includes not only the bulk resistances of the ScSZ disk and GDC interlayer but also the contact resistance of the interface between the ScSZ disk and GDC interlayer. Since the thickness of the ScSZ disk and GDC interlayer was 1 mm and 25 μm, respectively, the bulk resistance of the LSCF/ScSZ/LSCF with the GDC interlayer depends on the resistance of the ScSZ disk. All the cells shown in Fig. 7 had the same ScSZ disk. Therefore, the bulk resistance corresponding to the ScSZ and GDC should be the same. Overall, the decrease in the ohmic resistance from 1000 to 1200°C might be associated with the reduction in contact resistance, which was induced by sintering the GDC interlayer at high temperatures. Fig. 8 shows the Area Specific Resistance (ASR) of the LSCF/ScSZ/LSCF with the GDC interlayer together with the previous result.[13] The ASR of the cell prepared in this study was 0.07, 0.24, and 0.56 $\Omega\cdot cm^2$ at 800, 700, and 650°C, respectively.

4. Conclusion

The LSCF cathode and ScSZ electrolyte produced an interfacial-insulating layer, which is responsible for the high polarization resistance of the LSCF/ScSZ/LSCF cell. A GDC buffer layer designed to protect the LSCF cathode from the reaction with the ScSZ electrolyte was fabricated using a simple screen-printing technique. The screen-printed GDC interlayer prevented the interfacial reaction resulting in a low polarization resistance. The thickness and heat-treatment temperature are important for reducing the resistance of the LSCF cathode on the ScSZ electrolyte. A 15 to 25 μm thick GDC interlayer and a heat-treatment temperature of 1200°C were found to be the optimal conditions for the high performance LSCF/ScSZ/LSCF electrochemical cell.

Acknowledgements

This work was supported by Inha University Research Grant (INHA-31578).

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