

# Tubular Type Solid Oxide Fuel Cells Using LaGaO<sub>3</sub> Electrolyte Film Prepared by Dip Coating Method on Ni-based Anode Support

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論 文 名 : Tubular Type Solid Oxide Fuel Cells Using LaGaO<sub>3</sub> Electrolyte Film

Prepared by Dip Coating Method on Ni-based Anode Support

(Niアノード基板上へのディップコート法で作製したLaGaO<sub>3</sub>電解質膜を用いる円筒型固体酸化物燃料電池)

区 分 : (甲)

### 論 文 内 容 の 要 旨

As a highly efficient power conversion device, SOFC has been attracting much interests recently. As a basic design, tubular type SOFC is more suitable and applicable for commercialization because of easy gas seal which is particularly important for reversible operation. Based on this consideration, in this study, different Ni-based (NiO-GDC, NiO-SLT and NiO-YSZ) anode support tubular cells with CMF, Ti-LDC buffer layer and LSGM electrolyte film were investigated in order to study an effective fabrication process and improve the cell performance. This thesis consisted 5 Chapters as follows:

In Chapter 1, the background and current state as well as development of tubular SOFC and SOEC was reviewed in order to clear the objective of this study. The advantages of tubular SOFC were introduced.

In Chapter 2, the fabrication process of anode supported cell with LSGM electrolyte film by dip-coating and co-sintering method was investigated with NiO-GDC anode substrate. The calcination conditions, dip-coating conditions and compositions of dip-coating slurry was optimized in order to obtain dense and uniform LSGM electrolyte film after co-sintering at 1623 K. The effect of CMF and Ti-LDC buffer layer on decrease of Ni diffusion was confirmed for preparation of NiO-GDC cell. However, small amount of Ni was still detected in the LSGM electrolyte film, in particular, at the interface of Ti-LDC and LSGM layer. The power generation property of the cell was successfully measured. OCV higher than 1 V was obtained at 973-773 K. The obtained maximum power density of NiO-GDC cell was 0.35, 0.213 and 0.116 W/cm<sup>2</sup> at 973, 873 and 773 K, respectively.

In Chapter 3, two kinds of NiO-SLT substrates were studied as the anode substrate for further preventing Ni diffusion during co-sintering. Application of SLT for anode substrate is effective for trapping Ni diffusion by formation of NiTiO<sub>3</sub>. In order to confirm the trapping mechanism of NiO with SLT by formation of NiTiO<sub>3</sub>, Ni diffusion between NiO and NiTiO<sub>3</sub> was studied. The result indicated that the vapor pressure of Ni seems to be decreased on NiTiO<sub>3</sub> than NiO. Suppression of Ni diffusion into LSGM film on NiO-SLT substrate could be explained by decrease in vapor phase diffusion of Ni by formation of NiTiO<sub>3</sub>. Therefore, much higher OCV was exhibited on the cell by using NiO-Sr<sub>0.55</sub>La<sub>0.3</sub>TiO<sub>3</sub> anode substrate. The OCV was 1.083, 1.090, and 1.039 V at 973, 873, and 773 K, respectively. The maximum power density of 0.412

$\text{W/cm}^2$  which is slightly higher than that of NiO-GDC cell was achieved at 973 K. Since the thickness of the LSGM film was 50  $\mu\text{m}$  and a low conductivity in buffer layer and SLT in substrate, large IR loss was observed, in particular, at 773 K.

In Chapter 4, NiO-YSZ substrate was considered for further increasing the power generation property. The Ni diffusion was also investigated and the result indicated that NiO-YSZ is also effective for preventing Ni diffusion compared with NiO-GDC substrate. Compared with NiO-GDC and NiO-SLT cell, 0.543  $\text{Wcm}^{-2}$  much higher maximum power density was obtain at 973 K. However, a part of cathode layer might be easily delaminated from the LSGM electrolyte film after cell test and the poor contact of the cathode could be explained by the smooth surface of the LSGM electrolyte film. Therefore, inserting a porous LSGM layer between electrolyte and cathode which may increase in contact between cathode and LSGM film was studied. Deposition of the porous LSGM layer followed by impregnation of SSC air electrode is more effective for decreasing the IR loss and cathodic overpotential. Almost the theoretical OCV of the cell was achieved and the maximum power density of NiO-YSZ cell with the porous LSGM layer was 0.782, 0.315 and 0.1  $\text{W/cm}^2$  at 973, 873 and 773 K, respectively. In addition, the electrolysis performance was also investigated by the reversible operation of SOFC. The electrolysis current density was 1.47, 0.609 and 0.129  $\text{Acm}^{-2}$  at 973, 873, and 773 K, respectively, when the potential was 1.5 V applied. Cyclic reversible operation of the cell using 30%  $\text{H}_2$ -20%  $\text{H}_2\text{O}$ -50% Ar fuel gas was successfully performed and stable SOFC/SOEC operation was observed over 30 cycles. This suggests that introduction of the surface porous LSGM layer is highly effective for increasing mechanical and chemical strength of the air electrode.

In Chapter 5, the results in this thesis were summarized and based on the obtained results obtained, future aspects are described.