

FABRICATION AND CHARACTERIZATION OF ANODE SUPPORTED TUBULAR SOFCs WITH ZIRCONIA BASED ELECTROLYTE FOR REDUCED TEMPERATURE OPERATION

Tuong Lan Nguyen, Takeo Honda, Toru Kato,
 *Mitsunobu Shiono, *Akihisa Kobayashi, *Kan Hosoda,
 *Zifu Cai and *Masayuki Dokiya
 Electrotechnical Laboratory, National Institute of
 Advanced Industrial Science and Technology
 1-1-1, Umezono, Tsukuba, Ibaraki 305-8568 Japan
 *Yokohama National University,
 79-7 Tokiwadai, Hodogaya, Yokohama 240-8501, Japan

As aiming at the target of applying SOFCs to compact-size cogeneration unit and automobile power unit, reduced temperature SOFCs have been widely studied. The problems of R&D of reducing the operating temperature for SOFCs include the improvement of cell performance, reduction of manufacture cost, and scale up the capability of production. In order to overcome such above problems, we approach to fabricate anode supported thin electrolyte film tubular SOFCs by a wet co-fire process. Advantages of Y_2O_3 (3 mole %) and Sc_2O_3 (10 mole %) stabilized ZrO_2 electrolyte (3YSZ, SSZ) thin film, $(La_{0.6}Sr_{0.4})CoO_3$ (LSC) cathode were combined to improve the performance at reduced temperature. Ni (45 vol. %) cermet substrate tube was firstly fabricated by tape cast method. Electrolyte film was coated upon the substrate by a modified slurry dip coating technique, and then co-fired at $1300^\circ C$ for 2 h in air. $(CeO_2)_{0.9}(Gd_2O_3)_{0.1}$ (GDC) interlayer was deposited upon the electrolyte film before LSC was applied for preventing the reaction between stabilized ZrO_2 and LSC. Performance characteristics of fabricated cell were evaluated between 600 and $1000^\circ C$. Based on these data, the problems of materials and fabrications can be clarified for realizing of this type of cells.

Photograph of co-fired SOFC is shown in Fig.1. Sample (A) has a basic structure of NiO:YSZ/YSZ/GDC/LSC with a thick Ni cermet substrate (ca. 1mm). Sample (B) has a corrugated configuration. This allowed reducing the thickness of the substrate to ca. $150\mu m$. Sample (C) was co-fired with a Ni ferrite tube. This confirms the possibility of development of metallic substrate supported thin electrolyte film by a cost effective way than an expensive plasma spray.

Fig.2 shows SEM observation for cross section of co-fired sample. Dens, thin and uniform YSZ film stuck well to the substrate and GDC layer. In this case, the thickness of YSZ, GDC, and LSC layers were ca. 10, 3, and $20\mu m$, respectively.

In order to obtain the highest cathodic activity of LSC, in this case, we have investigated a most suitable condition for fabrication of GDC interlayer layer. The interfacial conductivity (σ_E) of LSC cathode increased as raising the fired temperature of GDC layer: $\sigma_{E,1320} \geq \sigma_{E,1200} > \sigma_{E,1100} > \sigma_{E,1000}$. However, in order to suppress reactions between GDC and stabilized ZrO_2 , it is preferred to fire the GDC at as low as possible temperature. Therefore, GDC interlayer was fire at $1200^\circ C$ before apply LSC cathode.

Primary cell performance tests were undertaken on the NiO:YSZ/YSZ/GDC/LSC cell (sample A in Fig.1) with the cathode effective area of $5.6cm^2$. 3% H_2O humidified H_2 (15ml/min/ cm^2) and air were used. The current-voltage and power output are shown in Fig. 3. OCV values in agreement with theory indicated that the electrolyte film

was perfectly fabricated. Maximum power densities were ca. $170 mW/cm^2$ ($600^\circ C$), $195 mW/cm^2$ ($650^\circ C$), $220mW/cm^2$ ($700^\circ C$), $220 mW/cm^2$ ($750^\circ C$), $230 mW/cm^2$ ($800^\circ C$), and $225 mW/cm^2$ ($850^\circ C$). In this case, the power densities did not increase so much when raising the operating temperature. The complex impedance measurements under open circuit reveal that the bulk resistances (R_b) that included electrolyte and substrate were larger than expected. As GDC interlayer was used, R_b can be enlarged due to the formation of insulator phases at 3YSZ/GDC interface. The electrode resistances were nearly equal with those of LSC/GDC case. The diffusion resistances governed the performance between 700 - $850^\circ C$.

Improvements for deposition of GDC layer and control of porosity of anode substrate are key factor for improving the performance of co-fired SOFC at reduced temperature.

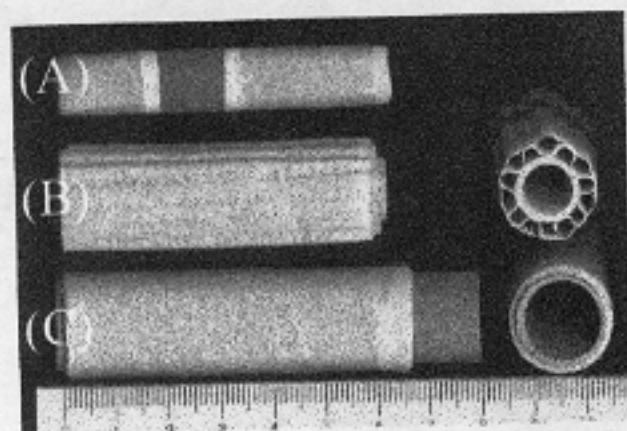


Fig. 1 Photograph of co-fired anode substrate tubular NiO-3YSZ/3YSZ, or SSZ/GDC/LSC SOFCs

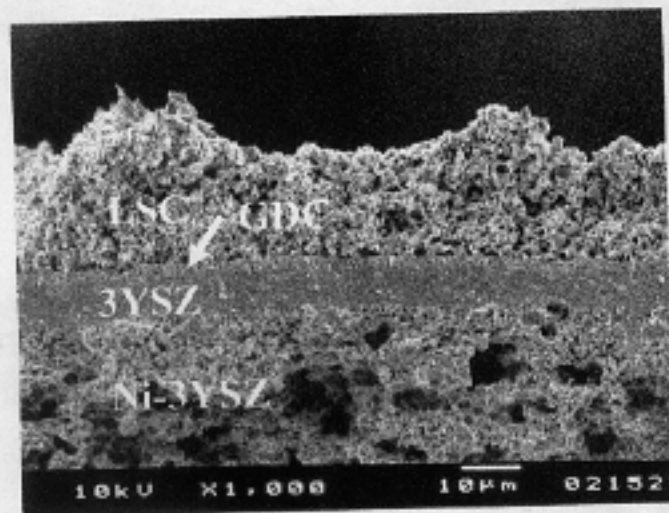


Fig.2 Cross section view of a co-fired cell

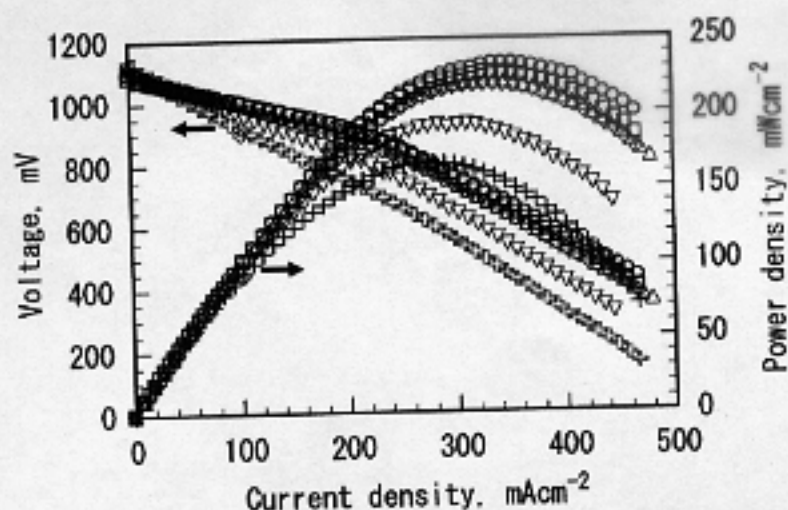


Fig. 3 Cell voltage (left) and power density (right) as function of current densities for a co-fired anode substrate tubular 3YSZ SOFC at temperatures from 600 to $850^\circ C$ ($600^\circ C$: +, $650^\circ C$: ∇ , $700^\circ C$: \diamond , $750^\circ C$: \triangle , $800^\circ C$: \circ , and $850^\circ C$: \square)