

Strain Induced Double Columnar Oxide Structure as Enhanced Active Electrode for Ni-Fe Metal Supported Solid Oxide Fuel Cells

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(Ni-Fe 金属担持固体酸化物燃料電池のための活性電極としての応力を導入したダブルカラムナ－酸化物構造)

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論 文 内 容 の 要 旨

In chapter 2, a double columnar structure film consisting of $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{CoO}_{3-\delta}$ (SSC) and $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{3-\delta}$ (SDC) as an active cathode was prepared by pulsed laser deposition (PLD) on a thin $\text{La}_{0.9}\text{Sr}_{0.1}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_{3-\delta}$ /SDC bi-layer film, and the effects of SSC/SDC ratio in double columnar layer on cathodic performance were investigated. It was found that the cathodic overpotential as well as power density were strongly dependent on SSC-SDC ratio in double columnar layer. Thus, different composition of the SSC-SDC double columnar structure (SSC:SDC = 10:0, 7:3, 6:4, 5:5, 3:7, and 1:9) were deposited on thin bi-layered SDC/LSGM film using NiFe oxide anode supported cell. From the XRD and STEM-EDX, deposited SSC-SDC double columnar structure was tilted double columnar shape and consisted of SSC and SDC. In addition, tensile strain was observed on SDC in double columnar structure with lattice dislocation at the interface between SSC and SDC. The effect of the double columnar layer composition on the cathodic activity was further studied and it was found that the smallest cathodic overpotential were achieved at the columnar composition of SSC:SDC=6:4 which is the optimized composition of the double columnar layer, and extremely high power densities which is higher than 3 W/cm^2 and 0.3 W/cm^2 were achieved at 973 and 773 K. This increased power generation performance was mainly assigned to the small cathodic overpotential and decreased IR loss. The increased surface activity toward oxygen dissociation can be assigned to the tensile strain effects in SDC that increase the oxygen vacancy concentration and diffusivity of oxide ion. From XPS results,

amount of Ce^{3+} is much increased at the optimized composition of SSC:SDC=6:4 suggesting the increased amount of oxygen vacancy. It is also found that the surface segregation of Sr in SSC is prevented at the 6:4 composition double columnar because of the compressed strain in SSC column. In summary, the increased activity and reaction area in cathode were obtained by SSC-SDC (6:4) double columnar structure by nano strain effects which is highly effective for increasing cathodic activity of SOFC.

In chapter 3, in order to investigate effect of double columnar structure, different type of double columnar structure was applied between SSC cathode and LSGM film. Therefore, the cathode candidates, PBCO, PNCG, SSC, and LSF, which used in SOFC were chosen for double columnar structure to investigate effect of cathode materials in double columnar structure. Therefore, SSC-SDC, PBCO-SDC, PNCG-SDC, and LSF-SDC double columnar structure were deposited as active interlayer between SSC powder and LSGM film. From the XRD, the deposited films showed typical crystal structure of the used materials for double columnar structure with strain effect, tensile strain on SDC and compressed strain on cathode in double columnar structure. By introducing different type of double columnar structure, power generation properties of the SSC were significantly changed. SSC-SDC and PNCG-SDC interlayer showed improved power density than that of the cell without interlayer film, i.e., MPD was increased from 2.24 W/cm^2 to 2.9 W/cm^2 in SSC-SDC film, 2.24 W/cm^2 to 2.35 W/cm^2 in PNCG-SDC film at 973 K. Insertion of PNCG-SDC or SSC-SDC double columnar layer between LSGM electrolyte and SSC cathode was highly effective for decreasing cathodic overpotential resulting in the increased power density at intermediate temperature. It found that the improved cathodic activity came from suitable TEC between SSC and LSGM. Also, original cathodic catalytic properties were important parameter. Furthermore, strain effect in double columnar structure was also seen to improve cathodic activity. In conclusion, SSC-SDC or PNCG-SDC double columnar structure as active cathodic interlayer has high potential for intermediate temperature SOFCs.

In chapter 4, to improve anodic catalytic activity, metal exsolution studies have been reported in perovskite oxide structure due to stability in reduction atmosphere. Thus, new oxide structure as CuFe_2O_4 spinel structure for anode was studied. Moreover, Ni doped $\text{Cu}_{1-x}\text{Ni}_x\text{Fe}_2\text{O}_4$ ($x=0.1, 0.2, 0.3$, and

0.5; CNFO) spinel oxide was prepared by solid state reaction. Although CNFO spinel structure was totally decomposed by reduction atmosphere observed by SEM-EDX, TEM-EDX and XRD, the decomposed CNFO became metallic phase with the exsolved nano size alloyed metal particles, which are Cu doped Ni-Fe and Cu doped Fe-Ni metal, on CNFO. The common metal exsolution study have been reported on oxide structure. Thus, the exsolved metal particle on metallic anode structure was quite new concept to improve anodic properties. It seemed that Ni doped Cu-Fe metal and Ni doped Fe-Cu metal were observed in CNFO 0.1 by XRD, and CNFO 0.1 showed the highest MPD 670mW/cm^2 at 1073 K and 120mW/cm^2 at 873 K. Comparing to NiFe (9:1 wt.%) alloyed metal anode, CNFO anode showed much improved anodic overpotential. At the high temperature (1073 K), there was enough thermal energy for electrode reaction, means that potential drop was mainly depended on IR loss properties, however, by decreasing temperature, electrode overpotential became main potential drop. Thus, although MPD of CNFO was lower than that of NiFe at 1073 K, its MPD was higher blow than 973 K. Thus, the exsolved metal particle on metallic anode structure using CNFO spinel oxide can be high potential for anodic catalytic properties at inter mediate temperature.