

Effects of GDC interlayer on performance of low-temperature SOFCs

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Abstract

To bridge the sintering temperature gap between the electrolyte and the cathode, low-temperature anode supported solid oxide fuel cells (SOFCs) with various thickness of electrolyte interlayer were fabricated and investigated. The porous thin interlayer was dip-coated and fired onto the dense $\text{Ce}_{0.9}\text{Gd}_{0.1}\text{O}_{1.95}$ (GDC) electrolyte surface. With humidified hydrogen as the fuel and air as the oxidant, the single cell with a 0.15 μm interlayer achieved the maximum power density (MPD) of 0.9 W cm^{-2} at 600 °C. The higher open circuit voltages (OCVs) (>0.9 V at 600 °C) were obtained in this study. The impedance results showed that the porous interlayer not only improved the interfacial contact between electrolyte and cathode, but also increased electrochemically active surface area. The cathode/electrolyte polarization resistance exhibited minimum when a 0.15 μm interlayer was added. The apparent activation energies derived from the Arrhenius plots of interfacial polarization resistances were about the same when the added interlayer was thinner, which indicated that the reaction mechanism did not change. However, the corresponding values were higher as the thick interlayer was introduced, which could be ascribed to the retarded oxygen ion transfer in the added porous layer. The cell area specific resistance (ASR) obtained by linear fitting I - V plot in the region of 0.6–0.7 V was higher than the ohmic resistance tested at OCV condition, and it was potentially attributed to the increased oxygen partial pressure at the anode as well as the contribution from polarization resistance, i.e. polarization of mass transport.

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1. Introduction

In the recent years, a number of investigations on solid oxide fuel cells (SOFCs) have been devoted to reduce the operation temperature. Low-temperature operation implies higher Nernst voltage. It allows the use of cheap stainless steel for the bipolar plates and the balance of the plant, together with the use of compliant high-temperature gaskets rather than rigid glass-based seals. Doped ceria, such as gadolinia doped ceria (GDC) and samaria doped ceria (SDC) are therefore adopted as the electrolyte [1–4], and highly active perovskites, such as $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$ (BSCF), are used as the cathode materials [5–7].

The recent noticeable development on anode supported SOFC with ceria-based electrolyte is that the high power densities, e.g. 1.010 and 1.329 W cm^{-2} , are obtained at a relatively

low temperature of 600 °C [7,8]. However, many obstacles still exist and should be overcome. The main disadvantages of doped $\text{CeO}_{2-\delta}$ ceramics are associated with a relatively easy reducibility at low oxygen partial pressures, which accelerated electronic transport and possible mechanical decomposition under large oxygen chemical potential gradients typical of SOFC operation [4,9]. Therefore the open circuit voltages (OCVs) are lower for the internal short circuit and gas leakage of the electrolyte. Some OCV values reported in literatures are summarized in Table 1. All the cells were tested with humidified hydrogen as the fuel and air as the oxidant. From the data in Table 1, it can be seen that the higher OCVs (>0.9 V at 600 °C) are obtained only when the electrolyte is thick or the fuel cell geometric area is small. Nevertheless, for the cells which are more practical, in which the electrolyte thickness is 20–35 μm and the geometric area is 0.25–1.2 cm^2 , the OCVs are lower. In addition, the stability of the cell is also negatively affected by the reducibility of the ceria-based ceramics in the anode substrate and electrolyte.

Another problem for the ceria-based electrolyte SOFCs pertains to the cathode/electrolyte interface. Presently, anode-

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Table 1
OCVs of single cells with ceria-based electrolyte in literatures

Electrolyte	Cathode	Cell geometric area (cm ²)	OCV (V) at 600 °C	OCV (V) at 500 °C	Refs.
10 μm Ce _{0.9} Gd _{0.1} O _{1.95}	Ba _{0.5} Sr _{0.5} Co _{0.8} Fe _{0.2} O ₃	0.005	0.903	0.984	[8]
10 μm Sm _{0.2} Ce _{0.8} O _{1.9}	Sm _{0.5} Sr _{0.5} CoO ₃ -SDC	1.35	0.835	0.873	[10]
20 μm Sm _{0.15} Ce _{0.85} O ₂	Ba _{0.5} Sr _{0.5} Co _{0.8} Fe _{0.2} O ₃	-	~0.85	~0.91	[7]
20 μm Sm _{0.2} Ce _{0.8} O _{1.9}	La _{0.6} Sr _{0.4} Co _{0.2} Fe _{0.8} O ₃	0.5	~0.85	~0.9	[11]
18–20 μm Sm _{0.2} Ce _{0.8} O _{1.9}	Sm _{0.5} Sr _{0.5} CoO ₃ -SDC	1.33	0.879	0.945	[12]
26 μm Ce _{0.9} Gd _{0.1} O _{1.95}	Sm _{0.5} Sr _{0.5} CoO ₃ -SDC	-	~0.83	~0.9	[13]
30 μm Sm _{0.2} Ce _{0.8} O _{1.9}	Sm _{0.5} Sr _{0.5} CoO ₃ -SDC	-	~0.86	~0.92	[14]
35 μm Sm _{0.2} Ce _{0.8} O _{1.9}	Sm _{0.5} Sr _{0.5} CoO ₃ -SDC	0.25	~0.83	~0.92	[15]
210 μm Ce _{0.9} Gd _{0.1} O _{1.95}	Sm _{0.5} Sr _{0.5} CoO ₃ -SDC	0.25	~0.95	~1.0	[16]
400 μm Sm _{0.2} Ce _{0.8} O _{1.9}	Sm _{0.5} Sr _{0.5} CoO ₃ -SDC	0.8	~0.91	~0.93	[17]

electrolyte assembly is generally sintered at a temperature over 1350 °C for ensuring a dense electrolyte film. However, the newly developed cathode materials, such as BSCF and Sm_{0.5}Sr_{0.5}CoO₃ (SSC), having a low melting temperature and a high reactivity with electrolyte, have to be fired onto the ceria-based electrolyte at a temperature below 1000 °C, even for a composite cathode [11,13,18]. Large sintering temperature gap between the electrolyte and the cathode may result in inferior cathode/electrolyte interfaces and consequently reduce triple-phase boundaries (TPBs). Thus further modification of ceria-based electrolytes is required in order to improve their performance and stability.

The “triple access” of gas, electrons, and oxygen ions to active catalyst sites, namely the TPB, ensures the occurrence of the electrochemical reaction, thus the extension of TPB becomes a determining factor in improving cell performance. TPBs strongly depend on the microstructure and morphology of the electrolyte/electrode interfaces, which in turn are determined by the materials properties and fabrication techniques. At the beginning, the extension of TPBs can be realized by adopting a composite cathode and the fine control of whole cell fabrication process [19,20]. Later, the method of adding an interlayer between the electrolyte and electrode was introduced. Effects of doped ceria interlayer on the cell performance were carried out by Barnett et al. [21–23]. A significant improvement in performance was observed when (CeO₂)_{0.84}(Y₂O₃)_{0.16} (YDC) was introduced at the La_{0.8}Sr_{0.2}MnO₃–(Y₂O₃)_{0.08}(ZrO₂)_{0.92} (YSZ)/YSZ and NiO–YSZ/YSZ interfaces, which resulted from the increased surface reactions. Ivers-Tiffée et al. [24,25] had performed the study of enlarging the electrochemical active surface by screen printing individual (Y₂O₃)_{0.1}(ZrO₂)_{0.9} (10YSZ) particles onto La_{0.75}Sr_{0.2}MnO₃/(Y₂O₃)_{0.08}(ZrO₂)_{0.92} (8YSZ) interfaces and developed a finite element model (FEM) for calculating and predicting the electrical performance of a SOFC with a three-dimensional interface between cathode and electrolyte. In a recent work of our group [26], the La_{0.8}Sr_{0.2}Mn_{1.1}O₃–YSZ/YSZ interface was modified with a thin layer of (CeO₂)_{0.01}(Sc₂O₃)_{0.10}(ZrO₂)_{0.89} (1Ce10ScZr) electrolyte, in view that 1Ce10ScZr displayed much higher oxygen ionic conductivity than YSZ and the two electrolyte showed good chemical compatibility and similar thermal expansion coefficients. With elicitation of those studies, the addition

of a high oxide conductivity and ceria containing interlayer is an effective way for the fine tuning of the interfaces and cell performance.

In order to bridge the sintering gap between the electrolyte and cathode and improve their interface microstructures, a porous GDC interlayer will be introduced with a sintering temperature between those for the electrolyte and cathode in this paper. Low-temperature solid oxide fuel cells with various thickness of GDC interlayer will be fabricated, and the effect of the GDC interlayer on the cell performance will be investigated.

2. Experimental

The home-made Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3–δ} (BSCF) and Gd_{0.1}Ce_{0.9}O_{1.95} (GDC) powders were synthesized by sol–gel process.

Anode-supported thin electrolyte bilayer assemblies were fabricated using a dual dry-pressing method. The anode, formed from a 50:50 wt.% mixture of NiO and GDC powders was dry-pressed into a pellet, and then an electrolyte powder was distributed on the anode surface and co-pressed with the anode using a uniaxial die-press (Ø25 mm). The resultant bilayer was sintered at 1420 °C for 4 h in air. The GDC electrolyte was coated with the GDC slurry onto its surface, and fired between 1150 and 1350 °C for 2 h in air. The interlayer thickness was calculated from the loading amount and density of GDC on the electrolyte, which was a parameter of loading amount per unit area. The cells with an interlayer thickness of 0.06, 0.15, 0.3, 1.5 and 3 μm were fabricated. A 30 wt.% GDC-containing BSCF cathode (area 0.5 cm²) was slurry coated on GDC film and fired at 950 °C for 2 h. In this study, the cells modified with the interlayer thickness of 0.06, 0.15, 0.3, 1.5, 3 μm were identified respectively as C1, C2, C3, C4 and C5 thereafter, and the unmodified cell was C0. The GDC thickness was calculated according to the compact film and only denoted the quantity of the GDC slurry.

Two gas chambers were set up by placing the cell between two alumina tubes. Silver meshes were used as current collectors and spring-pressed against the anode and cathode. After the in situ reduction of the NiO anode in flowing H₂ at 700 °C for several hours, the cell performance was measured at 600–500 °C by

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