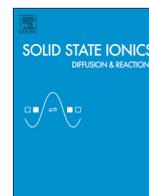




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Delamination-resistant bi-layer electrolyte for anode-supported solid oxide fuel cells

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ABSTRACT

One of the critical fuel cell degradation phenomena is ‘cell imbalance’ in a series-connected stack, which can cause abnormal operation under a negative cell voltage and consequently rapid degradation by anode interface delamination. In a previous study, the effect of electrolyte composition on the electrochemical degradation of solid oxide fuel cell (SOFC) was investigated, and it was observed that a small amount of ceria (an electronic conducting material) prevents anode delamination under abnormal (negative voltage) operation. However, the open circuit voltage (OCV) was lowered as a result of reduction of ceria. In the present study, bi-layer, YSZ (8 mol % yttria doped zirconia, a predominantly ionic conductor) at the cathode side and 8CYSZ (8 mol % ceria doped YSZ, a mixed ionic–electronic conductor) at the anode side were fabricated for anode-supported cells with a Pt probe embedded in each layer to estimate the internal oxygen chemical potential and tested under a negative voltage. The results indicated that the OCV was close to the theoretical value (similar to that of a YSZ single layer cell) and no delamination was observed under negative voltage operation (similar to the case of an 8CYSZ single-layer cell). Therefore, the bi-layer-structured electrolyte (with locally increased electronic conduction at the anode side) is effective in preventing anode/electrolyte delamination as well as maintaining open circuit voltage.

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1. Introduction

Commercially available alkaline and lithium batteries usually have a warning on their packaging saying “may explode or leak if mixed with a different battery type.” This is a warning against the phenomenon of cell imbalance. If cell imbalance exists in a series-connected battery pack, the entire battery pack as well as the imbalanced (bad) cell characterized by a higher resistance or lower capacity can be operated under a negative voltage, consequently causing damage during operation [1–6]. This cell imbalance problem is not limited to series-connected battery packs; the electrochemical degradation caused by negative voltage operation is also possible for a series-connected fuel cell stack [7,8].

In a previous study, YSZ electrolyte – anode supported cells were tested with an applied DC bias in order to study the SOFC stack failure mechanism by simulating abnormal behavior in a single cell test [7]. It was observed that the cell resistance rapidly increased when the cell was subjected to a negative voltage at a constant current. The degradation was caused by delamination, which developed along the anode interface during the negative voltage operation. When a cell is operated under a negative voltage, the ionic (I_i) and electronic current (I_e) directions through the electrolyte are the same, so that both I_i and I_e have negative signs. In such cases, a high internal p_{O_2} (partial pressure of oxygen) can be formed in the electrolyte just under the anode interface (depending upon ionic and electronic transport properties across

interfaces), and it eventually leads to physical damages (delamination) at the interface [7–12]. Note that the electronic conduction should be taken into account even in a predominately oxygen ionic conductor such as YSZ under the assumption of local equilibrium, i.e., $O^{2-} \rightleftharpoons \frac{1}{2}O_2 + 2e^-$ [9,13,14]. In terms of (electro)chemical potential, the local equilibrium can be described as

$$\nabla\varphi + \frac{1}{2e}\nabla\tilde{\mu}_{O^{2-}} = \frac{1}{4e}\nabla\mu_{O_2} \quad (1)$$

where μ_{O_2} is the chemical potential of neutral oxygen, $\tilde{\mu}_{O^{2-}}$ is the electrochemical potential of the oxygen ion, φ is the reduced (negative) electrochemical potential of the electrons or the electric potential, which is defined as $\varphi = -\frac{\mu_e}{e}$, where $\tilde{\mu}_e$ and e are the electrochemical potential of the electrons and the electronic charge, respectively.

When the electrolyte is a mixed ionic–electronic conductor (MIEC), $\nabla\varphi$ through the electrolyte is much smaller than that of a predominantly ionic conductor [15]. Accordingly, as shown in Eq. (1), $\nabla\mu_{O_2}$ must be different for an MIEC and a predominantly ionic conductor, because $\nabla\mu_{O_2}$ is linked to the corresponding $\nabla\varphi$. Electronic conduction, which is determined by the electrolyte composition, plays an important role in the electrolyte's thermodynamic stability; 8 mol% ceria (an electronic conducting material) doped in YSZ, namely 8CYSZ, improved the durability under negative voltage operation, which means that a small amount of ceria effectively prevented the development of high p_{O_2} inside the electrolyte [16]. However, the open-circuit voltage (OCV) of

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the 8CYSZ cell was lower than the theoretical value because of internal electronic leakage through the electrolyte, indicating a lower power density than that of a YSZ cell. A potential method of preventing loss of electromotive force in MIEC electrolyte cells is the use of a bi-layer structure [17,18].

In the present study, a bi-layer-structured electrolyte, with YSZ at the cathode side and 8CYSZ at the anode side, was fabricated and investigated, with a view to improving the OCV and the electrochemical stability under negative voltage operation. In addition, a Pt probe (electron selective probe) was embedded both in the YSZ and 8CYSZ electrolytes,

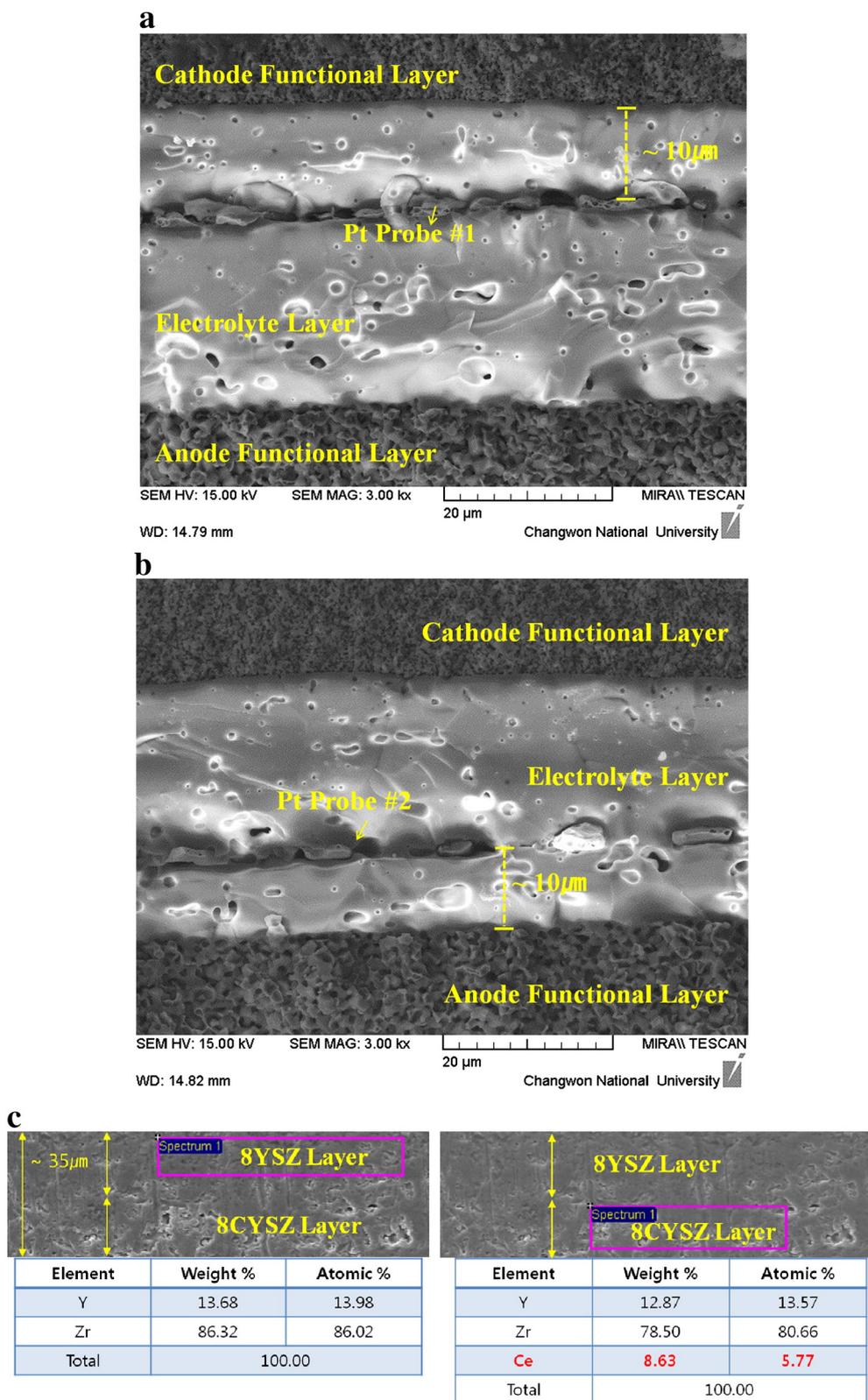


Fig. 1. SEM images of the cross section of the bi-layer electrolyte; (a) the embedded Pt probe #1 in the YSZ electrolyte $\sim 10 \mu\text{m}$ from the cathode interface; (b) the embedded Pt probe #2 in the 8CYSZ $\sim 10 \mu\text{m}$ from the anode interface; (c) the boundary between the YSZ layer ($17 \sim 18 \mu\text{m}$) and the 8CYSZ layer ($17 \sim 18 \mu\text{m}$) with EDS analysis results.

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