



A simplified approach to predict performance degradation of a solid oxide fuel cell anode



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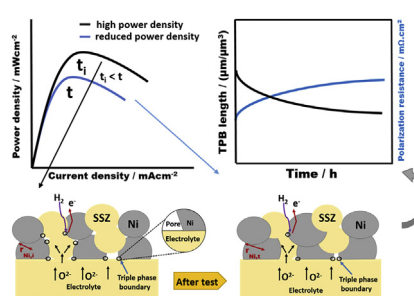
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HIGHLIGHTS

- Life-time prediction model of SOFC anode is developed and experimentally validated.
- Performance degradation of SOFC due to Ni grain growth is predicted.
- Ni particles growth and TPB length reduction fits well with the prediction model.
- Results of study discriminate degradation associated with Ni particles coarsening.

GRAPHICAL ABSTRACT



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ABSTRACT

The agglomeration of nickel (Ni) particles in a Ni-cermet anode is a significant degradation phenomenon for solid oxide fuel cells (SOFCs). This work aims to predict the performance degradation of SOFCs due to Ni grain growth by using a simplified approach. Accelerated aging of Ni-scandia stabilized zirconia (SSZ) as an SOFC anode is carried out at 900 °C and subsequent microstructural evolution is investigated every 100 h up to 1000 h using scanning electron microscopy (SEM). The resulting morphological changes are quantified using a two-dimensional image analysis technique that yields the particle size, phase proportion, and triple phase boundary (TPB) point distribution. The electrochemical properties of an anode-supported SOFC are characterized using electrochemical impedance spectroscopy (EIS). The changes of particle size and TPB length in the anode as a function of time are in excellent agreement with the power-law coarsening model. This model is further combined with an electrochemical model to predict the changes in the anode polarization resistance. The predicted polarization resistances are in good agreement with the experimentally obtained values. This model for prediction of anode lifetime provides deep insight into the time-dependent Ni agglomeration behavior and its impact on the electrochemical performance degradation of the SOFC anode.

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1. Introduction

Solid oxide fuel cells (SOFCs) are highly efficient electrochemical energy conversion devices that offer advantages of high electrical efficiency, low emissions, and fuel flexibility and are expected to fulfill the needs of electrical power generation in stationary and transportation applications [1–3]. However, long-term stability is a major hurdle impeding the practical applicability of SOFC technology. Numerous degradation mechanisms may cause performance loss, such as sintering of SOFC electrode materials during long-term operation at high temperature, chemical instability between components of the SOFC, reduction of the ionic conductivity of the electrolyte due to phase changes, and diffusion of impurities [4–9].

An SOFC unit cell is typically composed of a nickel 8mol.% yttria-stabilized zirconia (Ni-8YSZ) cermet anode, an 8YSZ electrolyte, and a lanthanum strontium manganite (LSM)-YSZ/lanthanum strontium cobalt ferrite (LSCF)-YSZ composite cathode [2,10]. The anode facilitates a fuel oxidation reaction whereas the cathode is responsible for the reduction of oxygen into oxygen ions, which diffuse to the anode through the electrolyte. The electrochemical reaction of the SOFC occurs at the TPB where the ion conducting phase, electron-conducting phase, and pore phase are simultaneously in contact [11,12]. The TPB area plays a significant role in controlling the electrochemical reaction of the SOFC. Microstructure stability and optimized TPB area are desired properties for the SOFC to work efficiently during long-term operation [13]. High-temperature operation for a longer time causes a TPB reduction due to microstructural changes such as particle coarsening and densification, which consequently leads to performance degradation. This phenomenon is specifically evident in the anode due to sintering of Ni at high temperatures [4,14–17]. Numerous experimental investigations have correlated the effect of Ni particle coarsening on the performance degradation of SOFCs. Faes et al. [15] studied the microstructure evolution of anode-supported SOFCs by using SEM imaging and revealed that Ni coarsening in a Ni-YSZ anode was responsible for 18–41% of total performance degradation. Similarly, Tanasini et al. [16] investigated the durability of Ni-YSZ anode-supported SOFCs and showed that performance degradation was strictly related to the Ni particle coarsening in the anode. The coarsening of Ni particles in the anode directly correlated with the corresponding reduction of TPB length [15]. From this relationship, it can be comprehended that the electrochemical performance of the anode is degraded primarily due to a reduction of TPB length resulting from the coarsening of Ni particles [15,16]. Furthermore, other studies [18,19] have experimentally demonstrated that the electrical conductivity of the Ni-YSZ anode decreases due to the evolution of a Ni-phase with exposure time. The coarsening of Ni particles is usually attributed to an Ostwald ripening process [20,21]. The potential underlying mechanisms for Ostwald ripening are the transport of volatile Ni species by gas phase and diffusion of vacancies due to the difference in grain size and surface curvatures [22].

These aforementioned studies [15–18] provide insight into the progression of SOFC degradation by Ni particle coarsening. However, very limited work [15,23] on predicting and correlating the impact of microstructure evolution on the electrochemical behavior of the SOFC anode has been performed. Holzer et al. [22] comprehensively summarized the time-based Ni grain growth data from the literature, but the data fail to provide a clear understanding of the Ni grain growth mechanism. The data in the literature indicate insufficient analyzed data points over time and different Ni particle coarsening behaviors. The apparently contradicting findings could be due to different experimental conditions, methodological uncertainty, and the underlying geometrical model for particle size quantification. Vaßen et al. [24] and Gao et al. [25] have considered the surface diffusion driven Ni particle agglomeration as Ni growth mechanism. However, practically, the anode materials are composite of Ni and YSZ particles and thus the Ni coarsening is hindered by YSZ skeleton. Faes et al. [15] have developed

capacitor charging model in which maximum Ni particle size can be obtained only after the long-term operation test which is generally an unknown value. The degradation associated with the Ni particles coarsening cannot be predicted precisely from the unknown value.

Most of the mathematical works in the literature are focused on the Ni coarsening phenomena. However, to date, no study has successfully and systematically quantified Ni coarsening over time and correlated it with the degradation of electrode performance. The main reason for this could be that microstructure studies become more difficult and cannot be performed in-situ under particular experimental conditions. Therefore, there is a strong need to develop lifetime prediction models and tools for evaluating the Ni coarsening mechanism in porous SOFC anodes which consider the Ni coarsening by YSZ interaction and correlate the Ni agglomeration, TPB length and performance degradation described by electrochemical polarization resistance.

The present study is devoted to the prediction and systematic investigation of the performance degradation due to Ni coarsening in an SOFC anode by using a simplified mathematical and experimental approach. An anode-supported flat-tubular type SOFC is considered in this study because this advanced design is the most widely commercialized [26–29] among various types [30] due to its advantages of high volumetric power density, easy gas-tight sealing, and ability to withstand thermal cycles [27,31]. The material studied is a Ni-scandia ceria stabilized zirconia (SSZ) anode functional layer (AFL) in a Ni-8YSZ anode-supported flat-tubular SOFC. The image analysis technique was used to analyze 2D backscattered electron (BSE) images of the AFL after every 100 h of operation at 900 °C. Image analysis is a simple and effective technique compared to other complex 3D techniques used in various studies [17,18,23] for particle size quantification. The obtained quantitative results are compared with the electrochemical characterization performed on a full cell utilizing EIS and equivalent circuit fitting to isolate the AFL components of the impedance responses. Mathematical correlations are developed for the prediction of Ni particle coarsening and TPB length reduction and fitted to the experimental data. The quantitative results provide a systematic and thorough understanding of the degradation phenomenon in Ni-based cermet anodes and its impact on the performance degradation of the SOFC.

2. Experimental

2.1. Fabrication of anode-supported flat-tubular SOFC

NiO (J. T. Baker), 8 mol.% Y₂O₃-stabilized-ZrO₂ (8YSZ; LTC) and carbon black powder were well mixed by a ball milling process for fabrication of a porous anode support. The flat-tubular anode support was fabricated by extruding a kneaded mixture of NiO-8YSZ powder with carbon black pore former, distilled water, organic binder, and plasticizer. The width and thickness of the extruded anode support were 43 and 4 mm, respectively. The extruded anode support was dried at 80 °C under 90% relative humidity (Jeio Tech Temperature Humidity Chamber) for three days and then pre-sintered at increasing temperature of 350, 750, and 1100 °C for 3 h at each step, respectively. An AFL composed of 50 wt% NiO (J. T. Baker) and 50 wt% (Sc₂O₃)_{0.10}(CeO₂)_{0.01}(ZrO₂)_{0.89} (SSZ; Fuel Cell Materials, USA) was coated on the anode support by a dip coating process followed by pre-sintering at 1000 °C for 3 h. Bilayer electrolytes of SSZ and Gd_{0.10}Ce_{0.90}O_{1.95} (GDC; Fuel Cell Materials, USA) were coated by a vacuum slurry coating process [32] and pre-sintering was performed at 1000 °C for 3 h for each electrolyte. Dual-layer Sr_{0.7}La_{0.2}TiO₃ (SLT) and La_{0.8}Sr_{0.2}MnO₃ (LSM) (HuBen Tech) dense ceramic interconnects were consecutively screen-printed onto the anode support. The anode support, AFL, electrolytes, and interconnects were co-sintered at 1400 °C for 5 h in air. An electrolyte and interconnect leak test was performed by supplying air in a gauge pressure range of 0.5–1.0 atm. A composite cathode paste of 50 wt% (La_{0.60}Sr_{0.40})_{0.95}Co_{0.20}Fe_{0.80}O_{3-x} (LSCF) and 50 wt% GDC (Fuel Cell Materials, USA) was coated onto the sintered

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