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Influence of deposition temperature on the microstructure of thin-film electrolyte for SOFCs with a nanoporous AAO support structure

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ABSTRACT

In this paper we investigate the influence of deposition temperature on the microstructure of yttria stabilized zirconia thin-films based on an anodized aluminum oxide (AAO) support structure. The yttria-stabilized zirconia (YSZ) electrolytes were deposited on a Pt anode/AAO support using DC magnetron reactive sputtering under deposition temperatures of RT and 500 °C. Elevating the deposition temperature led to enhanced surface mobility in the sputtered adatoms, which helped prevent pinhole generation and minimized the thickness of the electrolyte. A thin-film fuel cell with a YSZ electrolyte only 300 nm thick was successfully fabricated by elevating the deposition temperature. This cell exhibited an open circuit voltage (OCV) of 0.97 V, which is significantly higher than the OCV values of 0.3 V for a cell deposited at RT. However, in spite of the thin electrolyte, the performance of the cell deposited at the higher temperature showed limited value due to its relatively high polarization resistance. Through further investigation into the grain morphology, we verify that the increasing deposition temperature can affect not only the film density but also increase the grain size of the electrolyte, which is related to oxygen incorporation for ORR kinetics. Electrochemical impedance spectroscopy (EIS) results indicate that the grain size change caused by the elevated deposition temperature adversely affected the polarization resistance and the cell performance. These results indicate that careful adoption of elevated electrolyte deposition temperatures are required to optimize fuel cell performance.

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Introduction

Solid oxide fuel cells (SOFCs) have been intensively studied as promising energy conversion systems for their high energy conversion efficiency and environmentally friendly operation. Despite several advantages, SOFCs face a significant obstacle to commercialization due to their high operating temperature (800–1000 °C), which comes from the relatively low ionic conductivity of the solid electrolyte. The high operating temperature causes serious problems related to thermal stress and results in strict requirements for material selection. Many efforts have been made to lower the operating temperature to a low-temperature regime (300–500 °C) [1–5]. However, when the operating temperature is decreased, the yttria-stabilized zirconia (YSZ) commonly used as the solid electrolyte in SOFCs exhibits a significant decrease in ionic conductivity, which leads to increased ohmic resistance [6–8]. Also, the relatively sluggish oxygen reduction reaction (ORR) in the low operating temperature regime leads to increased charge transfer resistance at the cathode interface [9,10]. To overcome these issues at reduced temperatures, the thickness of the electrolyte should be minimized to decrease ohmic resistance and the electrochemical surface kinetics should be enhanced to reduce electrode interface resistance.

With advances in thin-film fabrication technology, various thin-film fabrication techniques such as physical vapor deposition (PVD), chemical vapor deposition (CVD), and atomic layer deposition (ALD) have been employed to fabricate thin electrolyte membranes [11–13]. In the area of thin-film SOFC, free-standing membrane-electrode-assembly (MEA) structures based on MEMS and lithography processes are employed to fabricate thin-film fuel cells with thin electrodes and electrolytes [13–18]. Dense and extremely thin electrolyte films can be fabricated in this structure, because the films are deposited on the flat and smooth surface of the supporting layer. As a result, researchers have found success in the fabrication of ultra-thin fuel-cell MEAs with electrolyte layers few tens of nanometers thick and have achieved significant enhancements in cell performance by reducing the ohmic resistance [14–17]. However, despite the high cell performance, ultra-thin free-standing fuel cells exhibit serious problems in thermomechanical stability due to their extreme thinness, thus increasing the difficulty of commercialization and fabricating large-area cells [18]. Although some research groups have successfully fabricated large-area free-standing fuel cells, they are yet to be competitive when compared to the fuel cells with supporting structures [19]. To address this weakness in thermomechanical stability, new platforms have been adopted for the anode supporting structure while maintaining the fabrication capability of thin-film MEAs, including thick, porous, anode-supported structures and anodized aluminum oxide (AAO) [20–24]. In these porous support structures, however, it is still challenging to fabricate a thin electrolyte layer with fully dense morphology due to the porous nature of the supporting structure. To address this issue, many researchers have focused on enhancing the film density of the electrolyte layer by modifying the deposition process and engineering the pore size of the supporting structure to obtain a highly dense morphology in the

electrolyte. For example, a recent report demonstrates that a dense electrolyte film was successfully achieved on a porous supporting structure by modifying the reactive gas pressure in the sputtering process [25]. Other reports describe adjusting the pore size of the support by adopting a functional layer, which has a smaller pore size than the supporting structure, allowing the fabrication of a denser electrolyte layer [20,26]. In the present study, we focus on modifying the deposition process to obtain a highly dense electrolyte layer.

Sputtering is a broadly used thin-film fabrication technique due to its relatively high deposition rate, large area coating ability, and the wide variety of materials it can deposit. These characteristics simplify the fabrication of large-area fuel cells, aiding the process to commercialization. However, the relatively low conformity of the sputtering process, which originates from the physical vapor deposition mechanism, results in thin-film electrolytes with insufficient density on the porous support structure. This low film density causes defects and voids, referred to as pinholes in thin-film electrolyte; these pinholes cause electrical shorting between electrodes and gaseous fuel permeation, leading to decreases in the open circuit voltage (OCV) and the overall cell performance [23,24]. Thicker films are therefore required in order to prevent electrolyte pinholes. Obtaining a highly dense electrolyte film with thin electrolyte is a major challenge that must be overcome to employ sputtering processes in AAO supported thin-film fuel cells.

In the sputtering process, various process parameters such as working pressure, deposition temperature, and applied bias voltage can affect the density of the deposited films [27–31]. Among these, elevating the deposition temperature is one of the most effective methods for enhancing the film density. Thornton described in detail the effects of deposition temperature on the microstructure of deposited films using a structure zone model, which classifies the possible microstructures of sputter-deposited films with variables in substrate temperature and argon gas pressure [27]. On the basis of this theory, high deposition temperatures increase the surface mobility of the adatoms and mitigate atomic level shadowing, which contributes to enhanced film density [32]. Thus, it can be expected that the film density of electrolyte layers in AAO based thin-film fuel cells can be increased and the electrolyte thickness minimized by increasing the deposition temperature. In addition, it is also speculated that the increased deposition temperature may influence the surface grain boundary of the electrolyte. In many previous reports, it is demonstrated that the surface grain boundary at the cathode side acts as a preferential pathway for oxygen incorporation and thus affects the charge transfer rate and fuel cell performance [33,34]. Accordingly, we analyze the surface grain boundary of the electrolyte layer to obtain high cell performance while optimizing the electrolyte density in order to minimize electrolyte thickness [35].

In this study, we investigate the effects of deposition temperature on the film density of the electrolyte and minimization of electrolyte thickness in AAO-supported thin-film fuel cells. The thin-film fuel cell with pinhole-free and 300-nm-thick YSZ electrolyte on the AAO supporting structure was successfully achieved by controlling the deposition temperature via the sputtering process, and the fuel cell

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