



Impact of nanostructured anode on low-temperature performance of thin-film-based anode-supported solid oxide fuel cells



Jung Hoon Park^{a, b}, Seung Min Han^b, Kyung Joong Yoon^{a, c}, Hyungchul Kim^{a, c}, Jongsup Hong^a, Byung-Kook Kim^a, Jong-Ho Lee^{a, c}, Ji-Won Son^{a, c, *}

^a High-temperature Energy Materials Research Center, Korea Institute of Science and Technology, Seoul 02792, South Korea

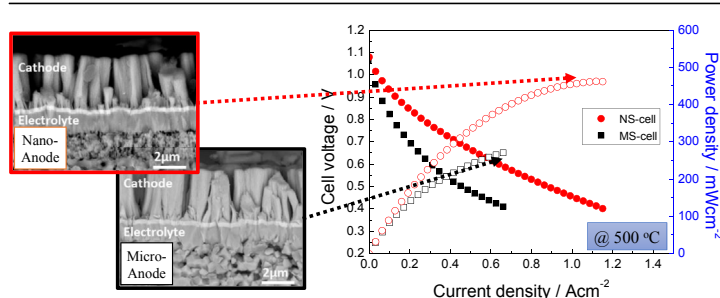
^b Graduate School of EEWS, Korea Advanced Institute of Science and Technology (KAIST), Daejeon 34141, South Korea

^c Nanomaterials Science and Engineering, Korea University of Science and Technology, KIST Campus, Seoul 02792, South Korea

HIGHLIGHTS

- The impact of a nanostructured Ni-YSZ anode on LT-SOFC is evaluated.
- The full SOFCs with and without the nanostructured anode are compared.
- The full-cell test revealed the contribution of the nanostructured anode at LT.
- The TPB increase by nanostructured anode improved charge transfer at the anode.
- Nanostructured anodes secure both the structural integrity and LT-performance.

GRAPHICAL ABSTRACT



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ABSTRACT

The impact of a nanostructured Ni-yttria-stabilized zirconia (Ni-YSZ) anode on low-temperature solid oxide fuel cell (LT-SOFC) performance is investigated. By modifying processing techniques for the anode support, anode-supported SOFCs based on thin-film (~1 μm) electrolytes (TF-SOFCs) with and without the nanostructured Ni-YSZ (grain size ~100 nm) anode are fabricated and a direct comparison of the TF-SOFCs to reveal the role of the nanostructured anode at low temperature is made. The cell performance of the nanostructured Ni-YSZ anode significantly increases as compared to that of the cell without it, especially at low temperatures (500 °C). The electrochemical analyses confirm that increasing the triple-phase boundary (TPB) density near the electrolyte and anode interface by the particle-size reduction of the anode increases the number of sites available for charge transfer. Thus, the nanostructured anode not only secures the structural integrity of the thin-film components over it, it is also essential for lowering the operating temperature of the TF-SOFC. Although it is widely considered that the cathode is the main factor that determines the performance of LT-SOFCs, this study directly proves that anode performance also significantly affects the low-temperature performance.

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1. Introduction

Solid oxide fuel cells (SOFCs) are promising next-generation power sources because they have higher efficiency than other competing energy conversion technologies [1–3]. However, the

* Corresponding author. High-temperature Energy Materials Research Center, Korea Institute of Science and Technology, Hwarangno 14-gil 5, Seongbuk-gu, Seoul 02792, South Korea.

E-mail address: jwson@kist.re.kr (J.-W. Son).

high operating temperature of SOFCs (≥ 700 – 800 °C) has limited the development and deployment of this technology because it induces fast degradation and poor reliability. Therefore, lowering the operating temperature is a major issue in SOFC research, and it has been actively studied over the past decade [3–6]. Reduced operating temperatures can prevent unwanted chemical reactions between the cell components and thermal degradation. It also enables the use of less expensive materials, e.g. stainless steel interconnectors. Low-operating-temperature SOFCs (LT-SOFCs) can be applied both as portable power sources and in automotive applications because the system size can be reduced [7–10]. However, the electrode activity of SOFCs, which is thermally activated, deteriorates significantly with decreasing operating temperature. Therefore, the performance of SOFCs is predominantly determined by polarization losses induced by the electrode reaction at low temperatures [6].

The electrode performance of SOFCs is strongly affected by its microstructural characteristics, such as porosity, grain size, and connectivity. Therefore, the relation between microstructural characteristics and the electrochemical performance of an electrode has been actively studied [11–17]. In terms of the grain size of the electrode, nanostructured electrodes have been applied intensively, and significantly, improved performances have been reported. This is because the rate of charge transfer in the electrode is influenced by the particle size and surface area of the catalyst. Thus, nanostructured electrodes have an increased rate of electrode reaction, which effectively reduces the polarization resistance at the electrodes at low temperatures [6,13,18,19]. However, the particle size obtained from conventional powder processing is on the order of micrometers because of the size of the raw powder and coarsening during high-temperature sintering. Therefore, novel nanotechniques, such as sputtering, pulsed-laser deposition (PLD), spray pyrolysis, metal organic deposition, and so on, have been employed to realize nanostructured SOFC electrodes [20–23].

In general, nanostructured cathodes have been intensively studied for low-temperature operation because it is known that the polarization loss at the cathode is the most significant factor in the poor low-temperature performance of SOFCs [24–26]. Nonetheless, the loss at the anode at low temperatures (≤ 600 °C) is predicted to become as significant as that at the cathode [27]. Therefore, it can be said that studies on nanostructured anodes are as important as those on the cathode for low-temperature operation. However, it is more difficult to fabricate nanostructured anodes because they mainly use metals as catalysts. Metal-phase agglomeration at SOFC operating temperatures eventually leads to serious problems such as the reduction of the triple-phase boundary (TPB) length and the loss of conductivity and thus to degradation of the cell stability [28,29]. In spite of the difficulty, in our previous study, a very uniform nanostructured Ni–yttria-stabilized zirconia (Ni–YSZ) anode with fine grain sizes of approximately 100 nm was successfully fabricated by using PLD [30]. The nanostructured layer was designed to suppress Ni agglomeration and to support thin-film electrolytes with a thickness of approximately 1 μm or less. Based on three-dimensional (3D) tomographic analyses, a recent paper reported that this nanostructured Ni–YSZ had a much higher TPB density, reaching almost 20 times that of conventional Ni–YSZ anodes [13].

Still, it is difficult to experimentally discern the impact of the nanostructured anode functional layer (nano-AFL) on full-cell performance for two reasons. One is that the nano-AFL is so thin (2–3 μm) that the catalyst amount in the layer is minute in comparison to that of the whole anode support, and thus it is difficult to exclude the participation of the top portion of the anode support in the anode reaction. If we can build thin-film electrolytes and cathodes directly over the anode support without the nano-AFL,

then we can compare the properties of the cells with and without the nano-AFL and elucidate the impact of nano-AFL, at least qualitatively. However, due to the surface conditions of powder-processed anode supports, such as a large pore size, high porosity, etc., it has been extremely difficult to build thin electrolytes directly over anode supports [30]. This is the second reason why it has been difficult to demonstrate the impact of the nano-AFL on the performance of LT-SOFCs with thin-film electrolytes (TF-SOFCs) experimentally.

However, by modifying the processing technique of the anode supports, we have been successful in obtaining a tape-cast anode support with satisfactory surface conditions by inducing more shrinkage in the bottom anode tape layer by means of pore formers. Now it is possible to build TF-SOFCs with and without nano-AFL and to elucidate the effect of the nanoscale grain size of the AFL more precisely, especially by electrochemical analyses. In this study, therefore, we investigated the electrochemical properties of TF-SOFCs at low temperatures as a function of the grain size of the Ni–YSZ composite anode. The output power and impedance spectra at various cell operating conditions were analyzed to compare the electrode reaction mechanism as affected by the grain size of the anode in the full-cell test.

2. Experimental

We prepared two different unit cells, one with an anode with a nano-scale AFL and one with an anode of micron-scale grain sizes. The cell with the nano-scale Ni–YSZ AFL is denoted as the NS-cell and that without the nano-AFL is denoted as the MS-cell hereafter. A 2 cm \times 2 cm and 1-mm-thick NiO–YSZ (NiO/8YSZ = 56:44 wt%) anode support was fabricated by tape casting. 150- μm -thick NiO–YSZ tapes with poly(methyl methacrylate) (PMMA) as a pore-forming agent and 30- μm -thick NiO–YSZ tapes without PMMA were also fabricated by tape casting. Then, seven layers of the 150- μm -thick tape and one layer of the 30- μm -thick tape were laminated at 75 °C under a uniaxial pressure of 15 MPa. The laminated substrate was sintered at 1300 °C for 4 h to fabricate a completely rigid substrate. For the NS-cell, a 2- μm -thick NiO–YSZ AFL was deposited onto the tape-cast anode support by PLD at 700 °C and an ambient oxygen pressure (P_{amb}) of 6.67 Pa. The support with the deposited nano-AFL was post-annealed at 1200 °C in air for 1 h [30]. This induces of the grain growth of NiO–YSZ from several nm to around 200 nm level and it would stabilize the nano NiO–YSZ structure by strengthening the oxide network and preventing uncontrollable Ni agglomeration [30–32]. Then, a 1- μm -thick YSZ electrolyte and 200-nm-thick gadolinia-doped ceria (GDC) buffer layers were deposited using PLD at a substrate temperature of 700 °C and a P_{amb} of 6.67 Pa. A 3- μm -thick lanthanum strontium cobaltite (LSC) layer with a 1 cm \times 1 cm area was deposited onto the GDC by PLD at room temperature and a P_{amb} of 13.3 Pa, followed by post-annealing at 650 °C in air for 1 h to crystallize the cathode. The detailed physical and electrochemical properties regarding the cathode by this process are reported elsewhere [33–36]. The MS-cell was also prepared in an anode-supported configuration, the same as the NS-cell except for the nano-AFL (Fig. 1). The electrolyte, buffer, and cathodes were directly formed on the tape-cast anode support under identical fabrication conditions as those of the NS-cell.

Electrochemical characterization was performed by the full-cell measurement. During the fuel cell operation, air and humidified H₂ (3% H₂O) were used as the oxidant and fuel, respectively. A detailed cell test configuration is described in our previous study [37]. The cell operating temperature varied from 650 to 500 °C at intervals of 50 °C, and the electrochemical impedance spectra (EIS) and current–voltage–power (I – V – P) curves were obtained at each

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