Title
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Journal
ECS Transactions, 103(1)

ISSN
1938-5862

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Publication Date
2021-07-09

DOI
10.1149/10301.0067ecst

Peer reviewed

ECS Transactions

The Electrochemical Society
Advancing solid state & electrochemical science & technology

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To cite this article: Nguyen Q. Minh *et al* 2021 *ECS Trans.* **103** 67

ECS Transactions, 103 (1) 67-71 (2021)
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Sputtered Thin-Film Solid Oxide Fuel Cells

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ABSTRACT

An all-sputter process has been developed and optimized to fabricate high-performance thin-film solid oxide fuel cells (TF-SOFCs) using conventional materials. The process involves sequential sputtering of cell components onto a porous substrate. An example structure comprises of an anode such as nickel-yttria stabilized zirconia (Ni-YSZ), an electrolyte such as YSZ, an interlayer such as gadolinium doped ceria (GDC), and then a cathode such as lanthanum strontium cobalt perovskite (LSC)-GDC. These are sputtered onto a porous substrate such as anodized aluminum oxide (AAO). Process conditions and sputtering procedures have been tailored and engineered to produce the components with desired structural characteristics, *i.e.,* a fully dense electrolyte and interlayer, and a porous anode and cathode. The porous electrodes show a columnar structure with branch-like nano-fibers. Sputtered TF-SOFCs exhibit superior performance at reduced temperatures; for example, the peak power density was about 3.0 W/cm^2 at 650°C (with H_2 for the cell Ni-YSZ/YSZ/GDC/LSC-GDC) and the peak power density was about 2.6 W/cm^2 at 700°C (with CH_4 for the cell Ni-GDC/YSZ/GDC/LSC-GDC).

Introduction

An all-sputter process has been developed and evaluated for fabrication of thin-film solid oxide fuel cells (TF-SOFCs) based on yttria stabilized zirconia (YSZ) electrolytes (1-3). The process involves sequential sputtering of the anode, electrolyte, cathode/electrolyte interlayer, and cathode films onto a porous substrate. Sputtering conditions and deposition procedures have been tailored and engineered to produce cell components and complete cells with the desired microstructural characteristics. Single cells made by this process have been characterized for their electrochemical performance at different operating temperatures and various fuel compositions.

Sputter Process

The process developed for the fabrication of TF-SOFCs is based on magnetron sputtering using an AJA Orion system (AJA International). This system has four 5-cm diameter sputter sources in the outer ring and a 7.5-cm diameter center source. All sources can use either RF or DC power supplies for deposition of insulating or conducting target materials, respectively. The 7.5-cm source is positioned at normal incidence to the substrate and the four 5-cm sources are positioned at an angle respect to the rotating substrate. The standard substrate holder diameter is 7.5 cm, which can be heated to 850°C (with full DC or RF biasing, as well as with the choice of annealing in vacuum, oxygen, or nitrogen atmospheres). The system is compatible with reactive sputtering with nitrogen or oxygen at room temperature or at elevated substrate temperatures. The system is computer-controlled, allowing automatic control of: the gas flow, sputtering power supplies, substrate temperature, and opening and closing of the shutters (on the sputtering sources) to allow the deposition or co-deposition of films and heterostructures.

The sputter process developed for fabricating TF-SOFC cells involves the following fabrication sequence: (i) the Ni-YSZ anode is first deposited onto a porous anodized aluminum oxide (AAO) substrate by co-sputtering with 7.5-cm Ni and 5-cm Y-Zr alloy targets, (ii) the YSZ electrolyte is then deposited on the anode using a 7.5-cm YSZ target, (iii) the gadolinium doped ceria (GDC) interlayer is then sputtered on the electrolyte using a 7.5-cm GDC target, and (iv) the lanthanum strontium cobalt iron perovskite (LSCF)-YSZ or lanthanum strontium cobalt perovskite (LSC)-YSZ or LSC-GDC cathode is deposited on the interlayer by co-sputtering with a 5-cm ceramic (LSCF or LSC) and a 7.5-cm alloy (Y-Zr or Gd-Ce) targets. All the sputtering processes were conducted with a target-to-substrate distance of about 4-10 cm. Process specifications have been developed and defined for the sputtering steps to make cells with desired structural characteristics and electrochemical performance. These specifications are given in Table 1. This process can be used to produce complete TF-SOFC cells in the same chamber using a single piece of equipment.

Table 1. Process Specifications of Sputter Steps in TF-SOFC Cell Fabrication (base pressure was < 5 x 10^{-7} Torr and all layers were deposited at ambient temperature).

Component	Component Material	Target	Atmosphere	Sputter Pressure	Power
Anode	Ni-YSZ	7.5cm Ni, 5cm Y-Zr alloy	Ar	30 mTorr	200 W DC for both Ni and Y-Zr
Electrolyte	YSZ	7.5cm YSZ	Ar	3 mTorr	200 W RF
Interlayer	GDC	7.5cm GDC	Ar	3 mTorr	200 W RF
Cathode	LSCF-YSZ	5cm LSCF, 7.5cm Y-Zr alloy	Ar	30 mTorr	200 W RF for LSCF, 50 W DC for Y-Zr
	LSC-GDC	5cm LSC, 7.5cm Gd-Ce alloy	Ar	30 mTorr	200 W RF for LSC, 20-40W DC for Gd-Ce

Structural Characterization

Microstructures of sputtered cell components and single cells are characterized using focused ion beam (FIB)/scanning electron microscopy (SEM)/transmission electron microscopy (TEM) techniques. Figure 1 is an example of the microstructure of cell components made by sputtering, which shows the capability of the process to produce a fully dense electrolyte and also porous anode and cathode films for TF-SOFC cells.

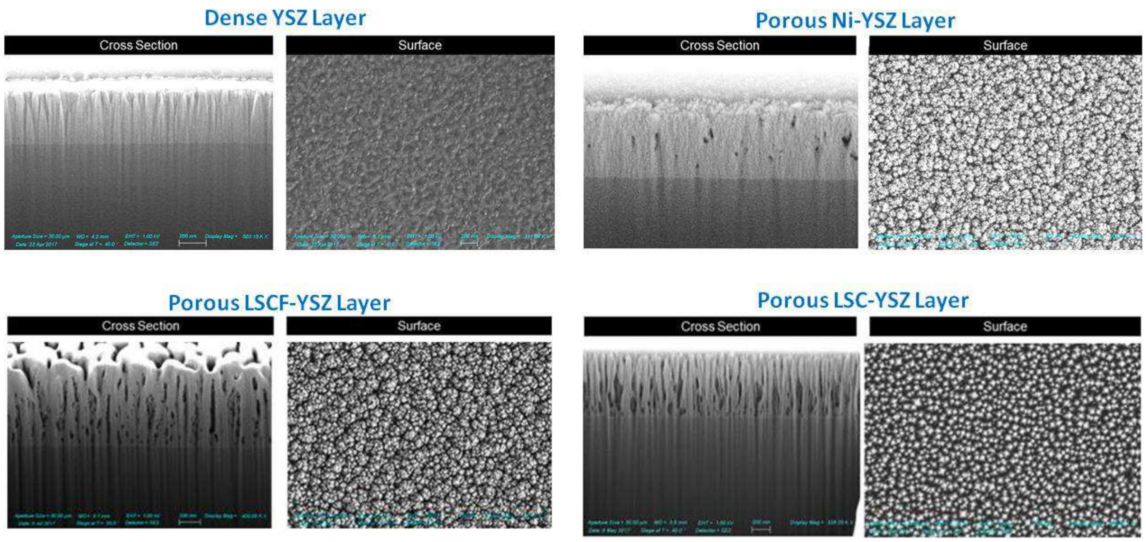

Figure 1. SEM Images that Show the Microstructure of YSZ, Ni-YSZ, LSCF-YSZ and LSC-YSZ Layers Deposited by Sputtering

Figure 2 shows as an example microstructure of TF-SOFC cells fabricated by the sputter process using the specifications given in Table 1. It can be seen from Figure 2 that the cells show cell components with the desired microstructures (i.e., fully dense electrolytes and interlayers, and porous anodes and cathodes) and well-defined interfaces between the layers. The electrodes exhibit a nanofibrous columnar structure with a diameter of less than 10 nm and relatively large openings between the columns (Figure 3). This kind of nanostructure provides a large active area and facilitates gas mass transport in and out of the electrode (resulting in reduced electrode polarization, thus high electrochemical performance. This will be discussed in the Electrochemical Performance section).

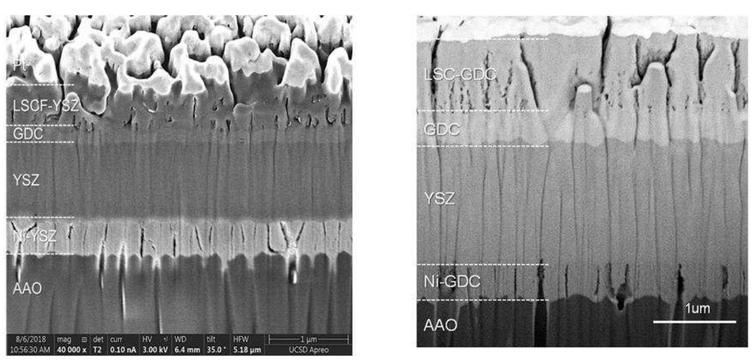

Figure 2. Typical Microstructure of TF-SOFC Cells Made by Sputter Process

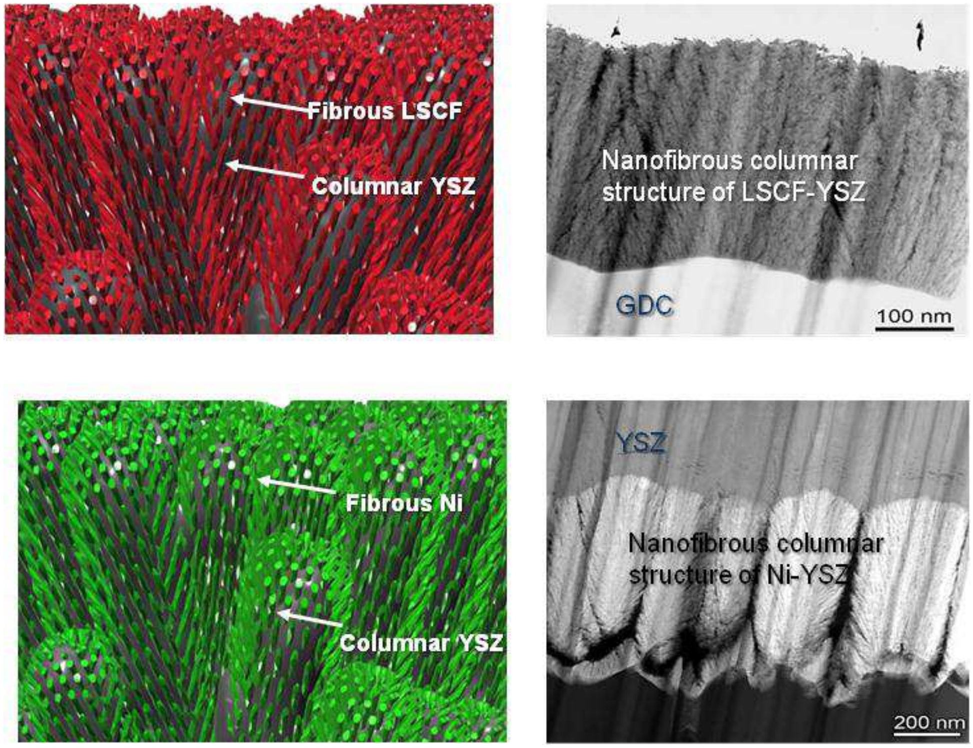

Figure 3. Electrode Microstructures (Left column: schematics, Right column: Images taken in STEM mode)

Electrochemical Performance

Sputtered TF-SOFC cells formed on AAO substrates has been evaluated for their electrochemical performance at reduced temperatures (<800°C) with different fuels (hydrogen and methane). Figure 4 shows example current/voltage (I/V) curves of two cells, Ni-YSZ/YSZ/GDC/LSCF-YSZ and Ni-YSZ/YSZ/GDC/LSC-GDC, with hydrogen fuel and air. As can be seen in Figure 4, the cells show excellent open-circuit voltage (≥ 1.0V) and superior performance at 600-650°C. For example, a peak power density of about 3.0 W/cm^2 was observed at 650°C for the cell Ni-YSZ/YSZ/GDC/LSC-GDC. With methane fuel, a cell having the configuration Ni-GDC/YSZ/GDC/LSC-GDC shows a peak power density of about 2.6 W/cm^2 at 700°C (Figure 5). These peak power densities are the highest reported for YSZ-based SOFCs at reduced temperatures.

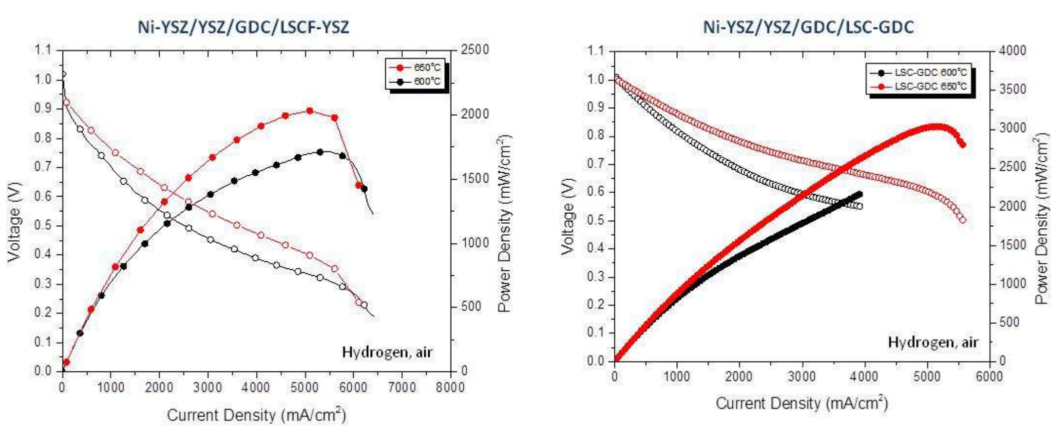

Figure 4. I/V Curves of Sputtered TF-SOFCs with Hydrogen Fuel at Reduced Temperatures

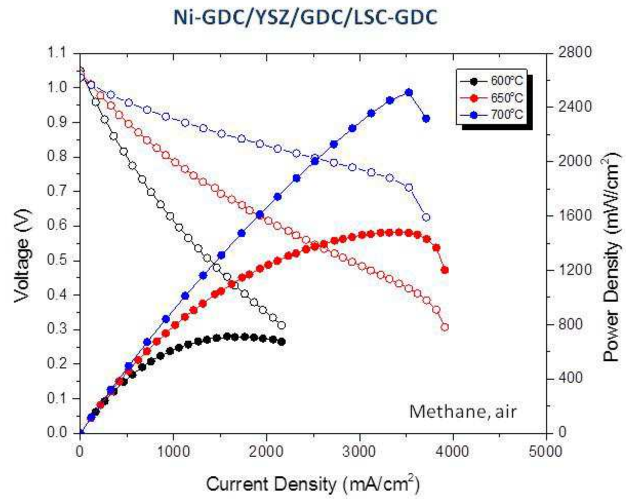

Figure 5. I/V Curves of TF-SOFC with methane Fuel at Reduced Temperatures

Summary

An all-sputtered process has been developed for fabrication of YSZ-based TF-SOFCs. Single cells fabricated by this process have shown required microstructural characteristics (fully dense electrolytes, fully dense interlayers, porous anodes and porous cathodes). The porous electrodes exhibit a nanofibrous columnar structure and relatively large openings between the columns. Electrochemical testing of sputtered TF-SOFCs has shown excellent open-circuit voltages (≥ 1.0 V) and record power densities with different fuels (e.g., about 3.0 W/cm^2 at 650°C with hydrogen fuel and about 2.6 W/cm^2 at 700°C with methane fuel).

Acknowledgment

This work is funded by the U.S. Department of Energy/National Energy Technology Laboratory (DOE/NETL) under the Cooperative Agreement DE-FE0026211.

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