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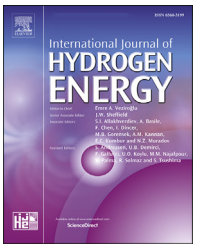

Short Communication

A novel single-step fabrication anode/electrolyte/cathode triple-layer hollow fiber micro-tubular SOFC

Siti Munira Jamil [a,b], Mohd Hafiz Dzarfan Othman [a,b,*], Mohd Hilmi Mohamed [a], Mohd Ridhwan Adam [a], Mukhlis A. Rahman [a], Juhana Jaafar [a], Ahmad Fauzi Ismail [a]

[a] Advanced Membrane Technology Research Centre, Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia

[b] Renewable Energy Research Group, Department of Energy Engineering, School of Chemical and Energy Engineering, Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia

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ABSTRACT

Micro-tubular solid oxide fuel cells (MT-SOFCs) have attracted much attention due to; higher volumetric output density, greater tolerance to cycling, quicker start-up capability and better mobility. Fabrication process of MT-SOFCs is one of the critical factor that ensures the success of a cell. There are three main components (i.e. anode, electrolyte and cathode) in the development of MT-SOFCs which are generally fabricated through separated and multiple laborious steps. This study aims to produce a novel anode/electrolyte/cathode (NiO-CGO/CGO/LSCF-CGO) triple-layer hollow fiber (TLHF) in a single-step using phase inversion based co-extrusion combined with co-sintering technique. The challenge lies within the diverse sintering behaviors among the anode, electrolyte and cathode layers at some point of the cell fabrication. In this study, the TLHF was found being able to survive the co-sintering at 1450 °C resulting a defect free precursor producing 0.48 Wcm^{-2} maximum power densities at 525 °C, which is comparable with the cell fabricated using conventional multiple-step process.

Introduction

For decades, research in solid oxide fuel cells (SOFCs) has mainly focused in improvement of the cell performance and producing more compact designs such as micro-tubular SOFCs (MT-SOFC) [1]. The MT-SOFC have recently attracted much attention because of their higher volumetric output density, better cycling tolerance, faster start up time and greater mobility [2]. The fabrication of the main and critical components of the MT-SOFCs namely; anode, electrolyte and cathode can only be attained through multiple step procedures [3]. For instance, an anode layer is firstly extruded as a support to the SOFC, subsequently by the electrolyte and

* *Corresponding author*. Advanced Membrane Technology Research Centre, Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia.
E-mail address: hafiz@petroleum.utm.my (M.H. Dzarfan Othman).

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cathode deposition. Every step requires at least one sintering step, which increases the fabrication cost [4–6]. Thus, single step fabrication that combined multiple step into one is anticipated in reducing production costs and time, and eventually the fuel cell price.

Recently, a more cost-effective fabrication technique of MT-SOFC with flexibility in quality control pioneered by Li and co-workers [7], i.e. a phase inversion-based co-extrusion process, is employed to fabricate electrolyte/anode dual-layer hollow fiber as part of intermediate temperature SOFC (IT-SOFC) [8–12]. The improvement of the dual-layer hollow fiber structures by increasing the anode effective porosity (70% void length) [7,13] and decreasing the electrolyte thickness to 10 μm [14] was done by simply altering the co-extrusion parameters without sacrificing the performance of the cell. An impressive power output, approximately 2.32 Wcm^{-2} which is almost double than the one prepared from conventional multi-step technique [15,16], was demonstrated in previous work using the co-extrusion technique [17].

One of the most successful architecture of this superior phase inversion extrusion and sintering technique is the production of asymmetric structure which consists of a combination of finger-like structure with some denser or sponge-like region [2]. Good asymmetric structure provides better fuel diffusion and a greater active zone eventually facilitating the optimal deposition of electrolyte. In the meantime, combination of co-extrusion and co-sintering indicates some of advantages for the development of anode/electrolyte/cathode TLHF in a single-step. The co-extrusion is greater attractive as it can reducing the production time and cost through the combination of three steps into one, reducing the probability of inducing defect as well as enhancing the adhesion among layers.

Li and co-workers [18] recently employed similar approach to produce TLHF wherein the primary layers were anode and anode functional layer (AFL), whilst the outer layer was pure CGO electrolyte. In order to prepare a complete anode/electrolyte/cathode single cell, a multi-layer cathode was dip-coated on electrolyte surface with a length of 10 mm. After several modifications, the cell eventually produce 1.21 Wcm^{-2} at 600 °C using pure H_2 [19]. However, the cathode deposition step on the triple-layer hollow fiber introduces considerable boundary effects between electrolyte and cathode that subsequently increases ohmic loss.

Thus, producing a novel complete set of anode/electrolyte/cathode (NiO-CGO/CGO/LSCF-CGO) TLHF in a single-step through phase inversion based co-extrusion/co-sintering technique is motivated. In this work, three suspensions were extruded to TLHF as a complete fuel cell via this superior technique followed by co-sintering at 1450 °C and were characterize by morphological and performance tests. The development of thin cathode and electrolyte layers supported on thick anode hollow fiber was succeeded after prevailing a number of challenges.

Experimental

In this work, we demonstrated a single-step fabrication of novel anode/electrolyte/cathode TLHF. The suspensions of the anode, electrolyte and cathode were prepared separately. The compositions of each suspension were tabulated in Table 1 along with viscosity at 2 s^{-1}. Then, all of the suspensions were co-extruded concurrently through a spinneret (four-orifice design), flowing through 10 cm air gap, then subsequently to a water bath as depicted in Fig. 1. Tap water was used as the internal and external coagulants and the flow rate of internal coagulant controlled by syringe pumps with the flow rates of 7 $mLmin^{-1}$, 2 $mLmin^{-1}$ and 2 $mLmin^{-1}$, respectively. Finally, the resultant TLHF precursors were co-sintered inside a tubular furnace. Starting from room temperature, the temperature was heating up to 400 °C (5 $°Cmin^{-1}$) for 1 h, subsequently to 800 °C (5 $°Cmin^{-1}$) for 1 h and lastly to the target temperature of 1450 °C (10 $°Cmin^{-1}$) for 8 h. The temperature was then reduced to room temperature at 5 °C min^{-1}.

The properties of the fabricated TLHF were characterized by scanning electron microscope (SEM), three-point bending and gas tightness tests that were described elsewhere [23]. Using the acquired data, the hollow fiber with the desired properties was selected and subjected to performance test.

A cell reactor was arranged before conducting the MT-SOFC performance test. Cathode area was wrapped with the conductive wire which act as cathode current collector and conductive silver paste was utilized to enhance the association of the wire to the cathode surface. Another silver winding wire was embedded into the fiber's lumen to collect the current from the anode. At that point, the readied MT-SOFC reactor was set in a gas-tight alumina tube by utilizing ceramic sealing paste. The voltage-current performance of the MT-SOFCs were assessed using NOVA 1.5 software by connecting the reactor via current collection wires to a potentiostat/galvanostat (Autolab).

Performance test was conducted by flowing humidified hydrogen at 20 $mLmin^{-1}$ through anode and oxidant at 40 $mLmin^{-1}$ flowing outside the cathode. The measurement was recorded at 500 °C–600 °C. TLHF inner's diameter was used to calculate an area of 0.232 cm^2 since the region used to compute the present thickness is generally the region of the fuel electrode [10]. Similar electrochemical workstation (0.01 Hz–1 MHz) was used to assess the AC impedance spectra with 10 mV signal amplitude at 500–600 °C.

Results and discussion

Fig. 2 demonstrates the SEM cross-sectional images of the TLHF precursors. The precursor's inner layer consists of a blend of NiO-CGO anode powders and polyethersulfone (PES) polymer while the middle and outer layers consists of CGO electrolyte powder and PES and a blend of LSCF-CGO cathode powders and PES, respectively. Clearly, precursors fibers and co-sintered fibers demonstrate an asymmetric structure where the cross-area of the anode layer displays long finger-like structures beginning from the anode surface to roughly 70% of the anode layer thickness, while the other 30% comprises of sponge-like structure. The finger-like voids that notably encourage the fuel gases diffusion into the anode layer [13] were completely preserved after the co-sintering step. The development of such asymmetric structure is a

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