Available online at www.sciencedirect.com

ScienceDirect

journal homepage: www.elsevier.com/locate/he

Co-extruded dual-layer hollow fiber with different electrolyte structure for a high temperature micro-tubular solid oxide fuel cell

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

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

[b] *Department of Energy Engineering, Faculty of Chemical and Energy Engineering, Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia*

ARTICLE INFO

Article history:
Received 7 October 2015
Received in revised form
3 June 2016
Accepted 3 June 2016
Available online xxx

Keywords:
YSZ electrolyte
Phase-inversion
Extrusion
Sintering
SOFC
Hydrogen fuel

ABSTRACT

In this study, the phase inversion-based co-extrusion method was employed to fabricate a structural-improved electrolyte/anode dual-layer hollow fiber (HF) precursor, which was then co-sintered at 1450 °C. The electrolyte structures were thoroughly investigated by varying the loading of electrolyte material (i.e. Yttria-stabilized zirconia, YSZ) with differing particle sizes (i.e. micron, sub-micron, and nano-sized) during suspension preparation. The results showed that the most promising electrolyte layer with thin, dense, gas-tight, and defect-free properties was obtained by mixing 70% submicron-YSZ and 30% nano-YSZ in electrolyte suspension (E-0.7sub0.3nano). This electrolyte formulation co-extruded with a thick nickel-oxide-YSZ (NiO-YSZ) anode layer yielded the highest bending strength of 85 MPa, providing major mechanical strength to the HF. Besides that, the nitrogen permeability value at 2.87×10^{-6} mol m^{-2} s^{-1} Pa^{-1} suggested that the electrolyte was gas-tight, preventing fuel and oxidant transport. The fiber was then reduced to nickel (Ni)-cermet anode. It was developed to be a complete micro-tubular solid oxide fuel cell (MT-SOFC) by depositing the lanthanum strontium cobalt ferrite (LSCF)/YSZ cathode via brush painting on the dual-layer HF. The cell was fed with hydrogen gas and yielded an open-circuit voltage (OCV) as high as 1.06 V with maximum power density of 0.243 W cm^{-2}, at 875 °C. Based on this test, it was found that the electrolyte structural-modified dual-layer hollow fiber-based MT-SOFC using mixed particle sizes may result in a promising OCV. However, the relatively low value for power density may be due to a less porous anode; thus, improvements in the anode's structure are required in future research.

* *Corresponding author.*
E-mail address: hafiz@petroleum.utm.my (M.H.D. Othman).

http://dx.doi.org/10.1016/j.ijhydene.2016.06.044
0360-3199/

Please cite this article in press as: Ahmad SH, et al., Co-extruded dual-layer hollow fiber with different electrolyte structure for a high temperature micro-tubular solid oxide fuel cell, International Journal of Hydrogen Energy (2016), http://dx.doi.org/10.1016/j.ijhydene.2016.06.044

Introduction

Smaller cell diameter micro-tubular solid oxide fuel cells (MT-SOFCs) offer excellent thermal stability during rapid heat cycling, quick start-up capability, and high power output density compared to the conventional planar and tubular SOFCs [1]. These MT-SOFCs can be fabricated using dry-jet wet extrusion method [2] producing hollow fiber support. For example, preparation of multi-layers HF, i.e. dual-layer HF-based MT-SOFC, involves the single-step fabrication method using the dry-jet wet co-extrusion and co-sintering method. It combines and simplifies the fabrication of electrolyte/anode dual-layer HF, greatly reducing production time and costs [3].

In general, this single-step fabrication technique consists of three main stages: (i) formation of particle suspensions by preparing anode and electrolyte spinning suspensions; (ii) packing of particle suspensions into a dual-layer HF precursor using dry-jet wet co-extrusion; and (iii) consolidation of dual-layer hollow fiber precursor via the co-sintering process, which requires heating at high temperatures. These stages play significant roles in the production of desired hollow fiber and should be actually be implemented starting from the preliminary methods for the preparation of ceramics membranes [4].

An important factor that may affect these three steps of the fabrication method is the particle size of ceramic electrolyte material. Liu et al. [5] found that ceramic particle size was greatly affected the particle dispersion, suspension viscosity, porosity, and densification of fabricated fiber due to the particle packing principle, as depicted in Fig. 1. Fig. 1(a) shows that further addition of small spheres forces the large spheres apart and Fig. 1(b) shows the excessive addition of large particles leave high number of voids since the voids cannot be fully occupied with limited number of smaller particles. The most efficient packing particles can be achieved at a stage in which a sufficient proportion of small and large particles touch their three neighboring particles [4], as shown in Fig. 1(c).

In this study, the effects of ceramic particles sizes and the loading to the resultant electrolyte structures fabricated via dry-jet wet co-extrusion/co-sintering technique were extensively discussed. However, when nano size particles are used, a higher tendency of agglomeration is expected. Suspension rheology is also affected by particle size distribution. Thus excessively small or large particles do not flatten as effectively. Few attempts have been made to use mixtures of different particle size distributions, e.g. micron/submicron/nano-sized particles despite their many potential advantages [6–11]. The use of mixed particle size in preparing suspension may offer significant benefits the particle packing, where the small spheres fill all voids in large ceramic particles packing effectively [4].

However, the use of these mixed particle size has not been treated in co-extrusion/co-sintering technique literature. To the best of our knowledge, limited studies have reported the influence of mixed particle size on the preparation of dual-layer HF for MT-SOFC specifically using this phase inversion based co-extrusion-inversion based fabrication technique. Yttria-stabilized zirconia (YSZ) was chosen as the electrolyte materials and particle sizes (i.e. micron, sub-micron and nano-sized) loading of YSZ in the electrolyte suspension was varied. The most suitable electrolyte candidate to be used as micro-tubular SOFC was then reduced and multiple layers of cathode were brush painted onto the electrolyte outer layer.

Lanthanum strontium cobalt ferrite (LSCF) mixed with YSZ was chosen as cathode materials replacing the conventional lanthanum strontium manganite (LSM). Unlike LSM, LSCF offers advantages in terms of ionic-electron conductor (i.e. it is a mixed ionic electron conductor, MIEC) and long-term stability [12,13]. Finally, the results of the operation of the structural-optimized dual-layer hollow fiber-based MT-SOFC fed with hydrogen are discussed. Instead of using hydrocarbon fuel, hydrogen was utilized as an ideal fuel for SOFC. As YSZ-based MT-SOFCs are prone to carbon deposition resulted from reforming of hydrocarbon reactions fed with hydrocarbon, more research is needed to determine details of catalytic and reforming activity at the anode [14].

Experimental

Materials

Commercial nickel oxide (NiO) with particle size (d50) of 12–22 μm and different particle sizes of 8 mol% yttria-stabilised zirconia (YSZ) were purchased from NexTech Materials (USA) and used as received. Three YSZ particles sizes

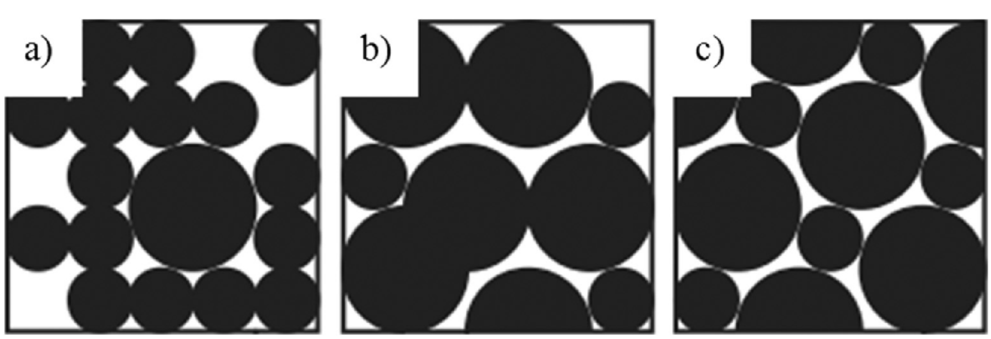

Fig. 1 – Schematic diagram of random dense packing of a) Excessive small ceramic particles; (b) Excessive large ceramic particles; and (c) Optimal packing in which the small spheres fill all voids in large ceramic particles packing [4].

Please cite this article in press as: Ahmad SH, et al., Co-extruded dual-layer hollow fiber with different electrolyte structure for a high temperature micro-tubular solid oxide fuel cell, International Journal of Hydrogen Energy (2016), http://dx.doi.org/10.1016/j.ijhydene.2016.06.044