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Review Paper

Structure Formation in Anode and Its Effect on the Performance of Micro-Tubular SOFC: A Brief Review

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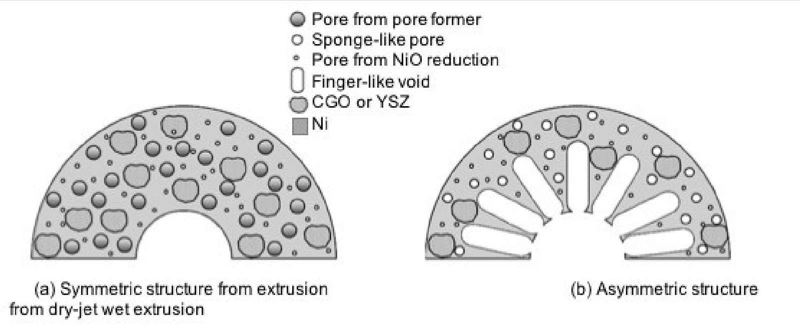

Highlights

- There is limited information on the structure of anode and its effect to the cell performance.
- Pore formation in anode and its effect to the MT-SOFC performance has been reviewed.
- Finger-like voids affected the anode's mechanical strength, permeation and conductivity.
- Pore formation in anode become a key parameter in producing high performance MT-SOFC.

Abstract

Anode-supported micro-tubular solid oxide fuel cell (SOFC) offers many advantages over the electrolyte and cathode-supported configurations in terms of simplicity, reliability, and efficiency. In such design, the anode substrates should possess a highly porous structure, provide active sites reaction as well as serving good mechanical strength. This structure is desired to provide enough fuel, which in turn increases the reaction rate and reduces the concentration polarisation. Hence, pore formation in anode become a key parameter in producing high performance micro-tubular SOFC. This review is mainly focusing on the fabrication of anode substrate for micro-tubular SOFC, the types of pores in anode structure and its effect to the cell performance.

Contents

1. Introduction

Recently, development in solid oxide fuel cells (SOFCs) are mainly focusing on the structures, designs, materials, and fabrication techniques of the components (i.e. anode, electrolyte, cathode, and interconnects) in order to meet the requirement of power production and boost the SOFCs performance in an economic way. There are two common structural designs; planar or tubular SOFCs, depending on its shape and structure. The tubular design has been introduced by extrusion method in order to prevent the thermal shock problem facing by conventional planar design. Since the power density is inversely proportional to the tubular cell diameter, Singhal and Kendall [1] had initiated an effort by introducing a smaller cell diameter known as micro-

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tubular SOFCs (MT-SOFCs). The development of this advanced cell design promotes an excellent thermal stability during rapid heat cycling, quick start-up capability, high power output density, low capital cost and portable characteristics compared to the conventional planar and tubular SOFCs [2].

In early 1990s to late 2000s, first generation of MT-SOFC developed was designed in an electrolyte-supported SOFC system with YSZ electrolyte tubes up to 5 mm in diameter. Thick electrolyte layer was first developed to serve as support layer of the cell. Indeed, support layer responsible to provide mechanical strength to the entire cell for deposition of the remaining cell layers [3]. Other promising designs include electrode-supported SOFC which uses thick anode or cathode as the supporting layer. Table 1 shows the comparison of different configurations in MT-SOFC. Anode-supported MT-SOFC [4-104] is more favourable than other type of configurations; electrolyte-supported [80,105–114] and cathode-supported [2,115–120] since it allows the deposition of thin electrolyte layer, which results to the reduction in ohmic lose and consequently, enhance cell's power density [35,83,121]. Besides, it offers many other advantages in terms of simplicity, reliability, and efficiency.

The anode will particularly serves as a "backbone" to support for the whole micro-tubular cell which gives mechanical support for the thin electrolyte and cathode layer [122]. Therefore, it is crucial to fully understand the structural properties of the anode substrates because these structural properties (i.e. particle size, porosity, pore-size distribution and composition of phases) determine the performances of SOFC system [4,11,50,123–127]. For instance, the performances represent its mechanical strength, gas permeability, tortuosity, electrical conductivity, and power-generating characteristic [128]. Therefore, this review focuses mainly on the fabrication of anode tube as a substrate layer for MT-SOFC, types of pore structure in the anode and how the pores could affect the performance of anode-supported SOFC.

2. Characteristics of an Efficient Anode Substrate for MT-SOFC

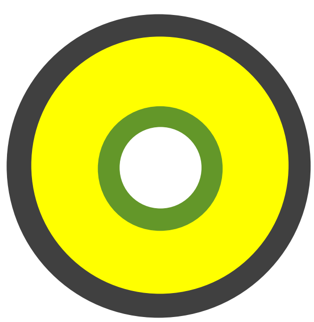
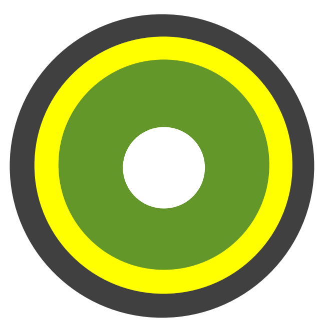
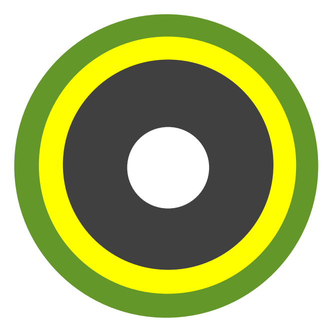
Besides acting as the mechanical support for SOFCs cell (in anode-supported case), anode tube primarily serves as a catalyst to provide active sites reaction or known as triple-phase boundaries (TPB) regions for the electrochemical oxidation of fuels. The electrochemical active sites in anode are responsible to produce numbers of ionic and electronic conduction paths besides create diffusion path for fuels and by-products (i.e. water and carbon dioxide) to reach and leave the site reactions, simultaneously [128]. Consequently, the anode substrates should possess a highly porous structure and retain its porosity after sintering. However, excessive porosity may degrade its mechanical strength [53] and its availability to provide the active TPB areas [125].

Metallic-anode material such as nickel (Ni) is commonly used as the main material for anode due to its excellent catalytic activity towards reforming and oxidation reaction [49,67,76,121-122,124,129–139]. Indeed, nickel is relatively low cost, excellent chemical stability, and high electronic conductivity. For the fabrication of anode tube as a substrate, nickel oxide (NiO) is commonly used instead of Ni, due to ease of processing and high availability, in which the sintered NiO tube will be reduced to Ni under hydrogen environment later. To date, ceramic-metallic material known as cermet is widely used as it satisfies most of the general requirements for SOFC anode. NiO is blended with electrolyte materials such as yttria-stabilized zirconia (YSZ) or cerium gadolinium oxide (CGO) in the dope suspension as the addition of electrolyte materials would extend the active zones for anode reaction, prevent the Ni from aggregation, reduce the thermal expansion coefficient mismatch and produce effective adhesion with electrolyte layer [140].

3. Fabrication Technique of Anode-Supported MT-SOFC

The quality of the anode supported cell mainly depends on its fabrication technique since the macrostructure, thickness and compound uniformity could be tailored during the fabrication process. Recently, an article that reviewed the most common and recent fabrication technique for MT-SOFCs method namely plastic mass ram extrusion and dry-jet wet extrusion has been published [141]. There are a number of similarities as well as dissimilarities between these two methods.

Table 1
Comparison of different configurations in MT-SOFCs.

MT-SOFCs Configuration	**Electrolyte-supported SOFCs**	**Electrode-Supported SOFCs**	
		Anode-supported SOFCs	**Cathode-supported SOFCs**
Schematic images of different MT-SOFCs configuration	 **Thick electrolyte**	 **Thick anode**	 **Thick cathode**
Advantages	• High mechanical robustness due to dense structures and good stability for RedOx (Reduced and Oxidation Atmosphere) cycles [142] • Gas-diffusible due thin electrodes layer [143]	• Low operating temperature (about 750 °C) and ohmic resistance due thin electrolyte layer [142] • High the electrical output due to low ohmic resistance [142] • Low materials cost since nickel (Ni) or nickel oxide (NiO) is relatively cheaper than electrolyte and cathode materials • Easy to fabricate	• Good stability under RedOx condition and low carbon deposition due thin anode [144]
Disadvantages	• High ohmic losses resulting from thick electrolyte layer [142]	• Low mechanical reliability due to porous structures and low RedOx stability [143]	• Limited study and research based on cathode-supported • Induce chemical reaction between cathode and electrolyte at high sintering temperature [145] • High polarisation resistance [145]

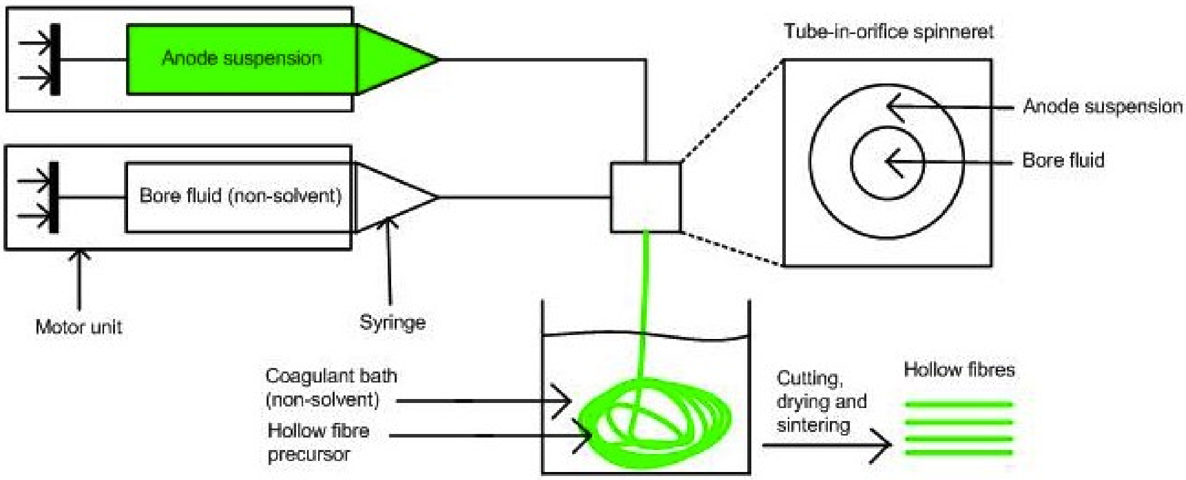


Fig. 1. Schematic diagram of hollow fibre fabrication via dry-jet wet extrusion.

In the ram extrusion method, a viscous paste is form by mixing the anode materials (NiO and YSZ) prior being transformed to support tubes using a customized die. Before proceeding to the sintering step, the support tubes are dried and cut to the 30 cm length. In contrast, the suspension of dry-jet wet extrusion method is in liquid form and the solidification of the hollow fibre occurs via phase inversion process initiated by the solvent/non-solvent exchange. Whereas in ram extrusion method, the tubes are dried immediately after extrusion before the sintering process.

The dry-jet wet extrusion is slightly more complex than that of the ram extrusion since it involves a non-solvent as coagulant. The system set-up is illustrated in Figure 1. The anode suspension is firstly prepared using anode materials (e.g. NiO and YZS), organic solvent (e.g. N-methylpyrrolidone) and polymer binder (e.g. polyethersulfone). The hollow fibre precursor formed after the internal coagulant passing through the spinneret centres. Subsequently, the solidification process via solvent/non-solvent exchange where the process called as phase-inversion [146]. Lastly, the hollow fibre is dried and cut to 30 cm before being sintered and reduced.

4. Pores in Anode Prepared by Phase Inversion

Distinctive fabrication methods would result in diverse anode structures [128]. For instance, mass ram extrusion method usually generates a symmetrical structure [34] whereas dry-jet wet extrusion technique commonly creates an asymmetric structure of anode tubes [83], as illustrated in Figure 2. Symmetric and asymmetric structures determine the homogeneous and heterogeneous morphology, respectively.

Microscopic pores that formed after NiO reduction to Ni is uniformly circulated in the anode regions which eventually improve the fuel and gas transportation in the layer [7]. Thus, several researchers had incorporated degradable pore-forming agent such as graphite [53,128] starch [125] and poly methyl methacrylate beads (PMMA) [7,52] into the anode suspension in order to induce the macro-size pore in anode substrate during the sintering process.

On the other hand, the formation of asymmetric structure through phase-inversion comprises of finger-like and sponge-like regions as shown in Figure 2 (b). The resultant structures enhances the gas diffusion as well as the flow of fuel and reaction products [83]. If the structural formation is one of the restraints of ram extrusion, the ability of dry-jet wet extrusion on generating preferred morphology can be realized by simple adjustment on additive loading in the suspension, air gap, or flow rate of internal coagulant [83,104].

Figure 3 shows the effect of ethanol loading in the dope suspension as the fabrication parameters on anode asymmetric structure [55], where characterized by the finger-like voids length. The sample name written in the SEM images was based on how much ethanol was added into the suspension. For example, E-0 represents 0 wt.% of ethanol while E-15 was for 15 wt.% of ethanol. As can be seen, addition of ethanol (from 0 to 20 wt.%) in anode inner suspension had resulted to the reduction in the finger-like voids length from 85 to 50% of anode thickness. This can be explained by the rise in the suspension viscosity when ethanol content is increased, which then will accelerate the precipitation rate during phase inversion process and thus resulting in shorter finger-like voids.

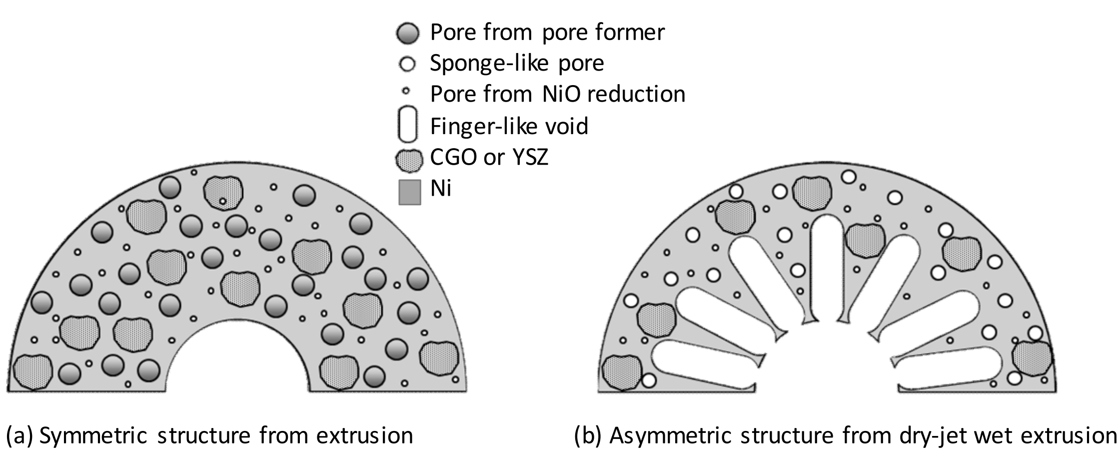


Fig. 2. Schematic diagrams of different structures of MT-SOFCs; (a) Symmetric structure from ram extrusion and, (b) Asymmetric structure from dry-jet wet extrusion [147].

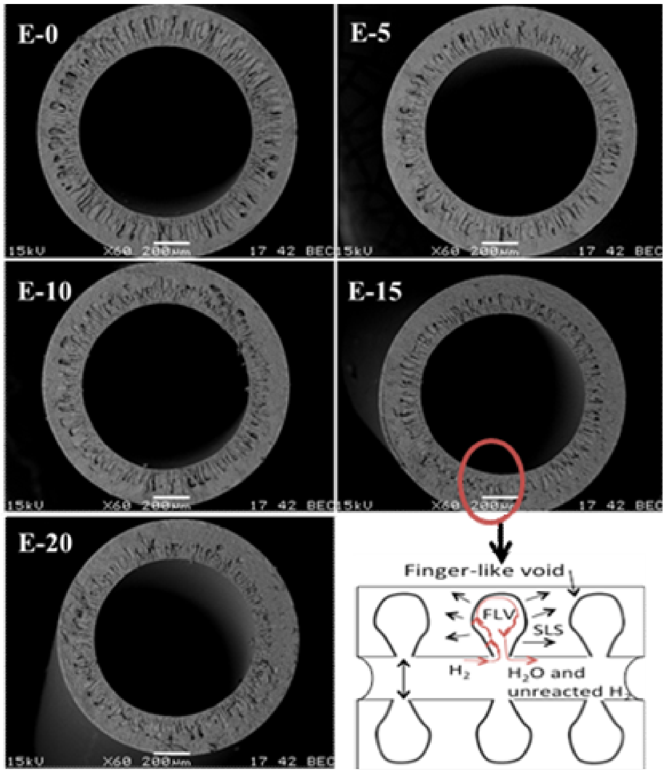


Fig. 3. SEM images of cross section of Ni-CGO anode with different anode structure [55].

The macrostructures of the hollow fibre such as the finger-like void length and the anode thickness are normally controlled during the dope preparation and extrusion process. On the contrary, the microstructure of anode can be tailored during high temperature sintering process. The particles rearrangements during sintering will led the particles bind together and induces pore channels along the grain boundaries. Then, the pores remain shrinking besides some of them pinch off and isolated at the grain boundaries as the temperature rises. Thus, some of sponge-like pore become smaller and probably eliminated during the sintering process depends on the temperature applied for the sintering. However, macrovoids as finger-like structure are still retained during sintering [148].

After sintering, the reduction of nickel oxide (NiO) phases to Ni phases is performed and this process further promotes the micropores formation due to the removal of oxygen atoms from the NiO crystal structure [149]. Therefore, it can be concluded that three types of pore is expected to form in the anode substrate prepared by the dry-jet wet extrusion technique, which are (a) finger-like voids, (b) sponge-like pores that survived from the sintering process and (c) the pores that created during the reduction of NiO [76].

5. Effect of pores to the fuel cell performance

Finger-like voids and sponge-like regions plays an important role to the anode support. In general, finger-like voids provide less resistance route for the fuel gas and products transportation, while the sponge-like structure provides a large number of TPB for the electrochemical reactions [126] and also give the major support to the hollow fibre [7].

In terms of fuel cell performance, it has been proven that the larger entrance pore size at the finger-like voids in the anode regions together with longer finger-like voids would reduce the fuel gas diffusion resistance into the finger-like voids zone [55]. It can be clearly seen in Figure 4 that the anode structure affected the maximum power density of the fabricated cells. The maximum power density increases with the increases of finger-like void lengths. The highest value was recorded at 2.32 Wcm^{-2} for the cell with 70% finger-like voids lengths (E-5). However, at about 85% finger-like voids thickness (E-20), the number of TPB region and anode conductivity are significantly reduced which eventually impairs the overall cell performance.

The improvement in the power density by the extension of finger-like length can be explained by the micro-view illustration as shown in Figure 3. Micro-channels provided by the finger-like voids structure in anode region helps to improve the fuel mixing and diffusion in the anode TPB region. Bigger finger-like voids entrance pore is commonly results from longer finger-like voids which gives excellent fuel gas diffusion into the finger-like voids region [104]. Moreover, vortex-flow inside the macro-voids induced by the conical shape of finger-like voids as proposed by Rahman et al. [150] [151]. Hence, the existence of longer voids could improve fuel mixing and subsequently offers better and greater uniform fuel gas distribution throughout the TPB in sponge-like region.

6. Conclusion and Future Direction

According to the interpretations, the anode structure was found to be greatly influenced by the fabrication methods. The anode support prepared by ram extrusion commonly gives symmetric structure while dry-jet wet extrusion technique typically induces the formation of the asymmetric structure which consists of finger-like voids and sponge-like region. Finger-like voids provide a less resistance route for fuel gas and the products transportation whereas sponge-like structure offers a large number of TPB for electrochemical reactions as well as mechanical strength to the hollow fibre. Moreover, dry-jet wet extrusion technique allows adjustments on extrusion parameters which eventually produce controlled structures in anode. Despite the fact that the anode hollow fibre with longer finger-like voids length and high porosity structure give higher gas permeation, it can strongly reduce the substrate's mechanical strength, destroying the TPB areas and ultimately affects the overall performance. Thus, the anode structure is one of the key parameter that has to be considered in order to fabricate a high performance cell.

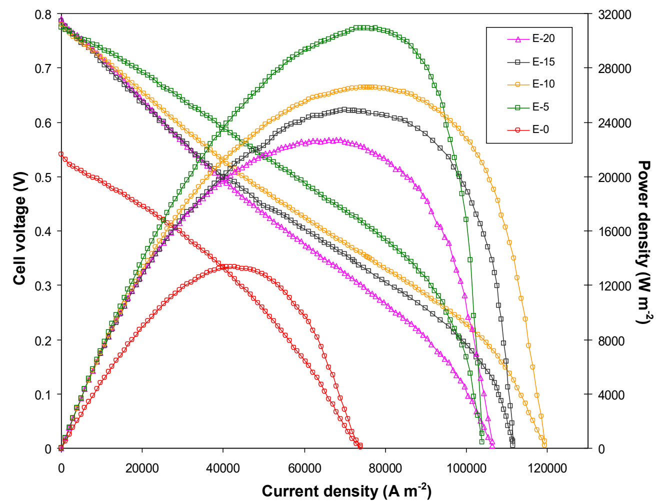


Fig. 4. MT- SOFCs performance with different anode structures [55].

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