

Critical Review on Fabrication of Tubular Solid Oxide Fuel Cell

P. S. Rane¹, R. V. Dubey², D. B. Rawal³, M. B. Mandake⁴

^{1,2,3}U.G. Student, department of chemical engineering, BVCOENM, Maharashtra, India.

⁴Asst. Professor, Dept. of chemical Engineering, BVCOENM, Maharashtra, India.

Abstract - In this, cold isostatic pressing and sintering process has been used to fabricate YSZ electrolyte over tubular NIO-YSZ for solid oxide fuel cell purpose. Tubular cells are preferred over planar because of no sealing requirement and their ability to withstand thermal stresses. SOFC is preferred over SOEC because of its ease of work and relevant for our fabrication purpose. Electrolytic cell was prepared by taking NIO-YSZ in 1:1 ratio and after mixing, drying, CIP and sintering the tubular cell was prepared. Sintering temperature was around 13500C for fabrication purpose. Agglomerates were removed for better purpose and much finer particle were used in order to lower the temperature of the process. Heat would be obtained by coupling the process either to a high-temperature reactor (HTR) or to a geothermal source. It is possible to improve the performance of electrolysis processes by operating at a high temperature. So the electrolytic cell was prepared with the help of Nickel oxide, yttria stabilized zirconia, ethanol, para-films, zirconia balls, planetary ball mill (200-250rpm), sieving, CIP and sintering etc.

Key Words: SOFC; Tubular SOFC; Cold isostatic pressing; symmetrical cell; high-temperature reactor...

1.INTRODUCTION

Fuel cells are a radically new and fundamentally different way of making electrical power from a variety of fuels. A fuel cell is an energy conversion device that produces electricity (and heat) by electrochemical combination of a fuel with an oxidant. A fuel cell consists of two electrodes (the anode and cathode) separated by an electrolyte. Fuel (e.g., hydrogen) is fed to the anode where it is oxidized and electrons are released to the external (outer) circuit. Oxidant (e.g., oxygen) is fed to the cathode where it is reduced and electrons are accepted from the external circuit. The electron flow (from the anode to the cathode) through the external circuit produces direct-current electricity. The electrolyte conducts ions between the two electrodes. Practical fuel cells are not operated as single units rather; they are connected in electrical series to build voltage. A series of cells is referred to as a stack. A component, variously called a bipolar separator or interconnect, connects the anode of one cell to the cathode of the next cell in a stack. Fuel cell stacks can be configured in series, parallel, and both series and parallel, or as single units, depending on the particular application. The key feature of a fuel cell is its high energy conversion efficiency. Because a fuel cell converts the chemical energy of the fuel directly to electrical energy without the intermediate

of thermal energy, its conversion efficiency is not subject to the Carnot limitation. Compared with conventional methods of power generation, fuel cells offer several advantages: substantially higher conversion efficiency, modular construction, and high efficiency at part load, minimal siting restriction, potential for cogeneration, and much lower production of pollutants.

The principles of fuel cell operation were first reported by Sir William Grove in 1839 [1]. His fuel cell used dilute sulphuric acid as the electrolyte and operated at room temperature. Ceramic fuel cells came much later and began with Nernst's discovery of solid-oxide electrolytes in 1899 [2] and the operation of the first ceramic fuel cell at 1000°C by Baur and Preis in 1937 [3]. Since that time, ceramic fuel cell technology has made excellent technical progress. Multi kilowatt fuel cells, based on stabilized zirconia electrolyte, have been operated for thousands of hours and have shown excellent performance. Recently, ceramic fuel cell research and development has received much attention, reflecting widening interest in this technology.

Ceramic fuel cells have several distinct advantages over other types of fuel cells, e.g., use of nonprecious materials, no liquids involved in the fuel cell, and invariant electrolyte. The use of a solid electrolyte in ceramic fuel cells eliminates material corrosion and electrolyte management problems. Ceramic fuel cells are generally operated at high temperatures (>600°C). The high operating temperature promotes rapid reaction kinetics, allows reforming of hydrocarbon fuels within the fuel cell (internal reforming), and produces high-quality by product heat suitable for use in cogeneration or bottoming cycle. Thus, power systems based on ceramic fuel cells can be simple and are more efficient than many other technologies. Furthermore, because all the components are solid, ceramic fuel cells can be fabricated in very thin layers, and cell components can be configured into unique shapes unachievable in fuel cell system shaving a liquid electrolyte [4]. This feature permits cell designs with additional performance improvements.

The principal components of a ceramic fuel cell stack are the electrolyte, the anode, the cathode, and interconnect. Each component serves several functions in the fuel cell and must meet certain requirements. Each component must have the proper stability (chemical, phase, morphological, and dimensional) in oxidizing and/or reducing environments, chemical compatibility with other components, and proper conductivity. The components for ceramic fuel cells must, in addition, have similar coefficients

of thermal expansion to avoid separation or cracking during fabrication and operation. The electrolyte and interconnect must be dense to prevent gas mixing, whereas the anode and cathode must be porous to allow gas transport to the reaction sites.

2. TYPES OF FUEL CELLS

Any gases capable of electrochemical oxidation and reduction can be used as fuel and oxidant in a fuel cell. However, hydrogen is currently the most common fuel for use in ceramic fuel cells. Hydrogen has high electrochemical reactivity and can be derived from common fuels such as hydrocarbons, alcohols, or coal. Oxygen is the most common oxidant for ceramic fuel cells since oxygen is readily and economically available from air. An electrolyte for a fuel cell based on the electrochemical combustion of is either an oxygen-ion conductor or a hydrogen-ion (proton) conductor. (Although a hydroxide-ion conductor is also possible, it will be referred to as a proton conductor in this paper for simplification reasons.) Thus, the present generation of ceramic fuel cells can be classified into two types: those based on oxygen-ion conductor and those based on proton conductor. The major difference between the two types is the side in the fuel cell in which water is produced (the oxidant side in proton-conductor fuel cells and the fuel side in oxygen-ion-conductor fuel cells). Also, certain gases, such as carbon monoxide, can be used as fuel in oxygen-ion-conductor fuel cells but not in proton-conductor fuel cells. To date, only oxides are being considered for ceramic fuel cell electrolytes. Since fuel cells are commonly identified by the type of electrolyte used, ceramic fuel cells are therefore referred to as solid-oxide fuel cells (SOFCs).

3. MATERIALS FOR CELL COMPONENTS

3.1 ELECTROLYTE

Stabilized zirconia (ZrO_2), especially Ytria-Stabilized Zirconia, is the most common electrolyte in SOFCs because the material possesses an adequate level of oxygen-ion conductivity and exhibits desirable stability in both oxidizing and reducing atmospheres. The properties of stabilized zirconia have been extensively Studied [5-7]. Several reviews on the subject are available [8-10].

ZrO_2 , in its pure form, does not serve as a good electrolyte, primarily because its ionic conductivity is too low. At room temperature, ZrO_2 has a monoclinic (m) crystal structure. The monoclinic structure changes to a tetragonal (t) form above $1170^\circ C$ and to a cubic fluorite structure above $1370^\circ C$. The tetragonal-monoclinic transformation is associated with a large volume change (3% to 5%), (Contraction on heating and expansion on cooling). The cubic phase exists up to the melting point of $2680^\circ C$. However, the addition of certain aliovalent oxides stabilizes the cubic fluorite structure of ZrO_2 from room temperature to its melting point and, at the same time, increases its

oxygen vacancy concentration. This enhances the ionic conductivity and leads to an extended oxygen partial pressure range of ionic conduction, making stabilized ZrO_2 suitable for use as an electrolyte in SOFCs. (This extended oxygen partial pressure range covers the conditions, 1 to 10^{-18} atm (105 to 13 Pa), to which a SOFC electrolyte is exposed in the fuel cell during operation.) The most commonly used stabilizing oxides or dopants are CaO, MgO, Y₂O₃, Sc₂O₃, and certain rare-earth oxides. These oxides exhibit a relatively high solubility in ZrO_2 and are able to form the fluorite structure with ZrO_2 which is stable over wide ranges of composition and temperature.

3.2 ANODE

Nickel/Ytria-Zirconia Cermet: Because of the reducing conditions of the fuel gas, metals can be used as SOFC anode materials. At the $1000^\circ C$ operating temperature of YSZ-based SOFCs, suitable metals are limited mainly to nickel, cobalt, and noble metals. Nickel is most commonly used because of its low cost. To maintain the porous structure of nickel over long periods at $1000^\circ C$ and to provide other desired properties for the anode, nickel metal is often dispersed on the surface of an YSZ support. The YSZ support is added to the anode formulation in the form of powder or fiber [12]. Nickel has a higher thermal expansion coefficient than YSZ there are concerns about thermal expansion mismatch between the anode and the electrolyte. A significant degree of mismatch in thermal expansion coefficients of the SOFC component can result in large stresses, causing cracking or delamination during fabrication and operation [13]. For a thin air-fired NiO/YSZ layer (e.g., 100 pm thick), it takes only minutes to complete the NiO reduction at $1000^\circ C$. During the reduction, the conductivity of the anode generally reaches a maximum very quickly then falls off slowly until a steady state is obtained [11]. Sintering of nickel particles is a major concern in long-term operation of the SOFC anode at $1000^\circ C$. Nickel sintering results in loss of active surface area and reduced conductivity of the anode, leading to degradation of cell performance. Since nickel particles are high-surface-area solids, there will always be a thermodynamic driving force to decrease free energy.

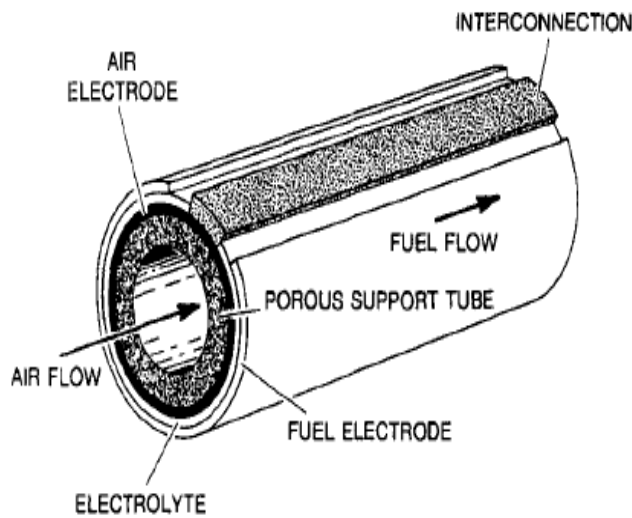


Fig.1. Sealless tubular design of solid-oxide fuel cell [14].

3.3 CATHODE

Doped Lanthanum Manganite: Because of the high operating temperature (1000°C) of the YSZ based SOFC, only noble metals or electronic conducting oxide can be used as cathode materials. Noble metals, such as platinum, palladium, or silver, are unsuitable for practical applications because of prohibitive cost. The disadvantages of most of these materials are thermal expansion mismatch, in compatibility with the electrolyte, and lack of conductivity. At present, doped lanthanum manganite (LaMnO₂) is most commonly used. LaMnO₂ is a p-type perovskite oxide. The perovskite structure of LaMnO₂ may undergo atomic distortion leading to orthorhombic or rhombohedral unit cells. Undoped LaMnO₂ is orthorhombic at room temperature [15]. At high temperatures, the material can have oxygen excess, stoichiometry, or deficiency depending on oxygen partial pressure [16-18]. For example, at 1200°C, the oxygen stoichiometry of LaMnO₃ ranged from 3.079 to 2.947 under oxygen partial pressures of 1 to 10^{-10.60} atm (10⁵ to 10^{-6.60} Pa) [16].

4. SOLID OXIDE FUEL CELL ELECTRODES REACTIONS

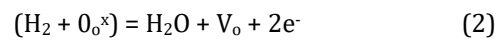
The reversible voltage E^o is the maximum voltage that can be achieved by a SOFC under specified conditions of temperature and gas composition. E^o can be calculated from the Nernst equation. The voltage of an operating cell E is always lower than E^o. As the current is drawn from the fuel cell, the cell voltage falls due to internal resistance and polarization losses. Thus, the voltage of an operating cell is given as

$$E = E^o - IR_1 - (\eta_a + \eta_c) \quad (1)$$

In the above equation, IR₁ is the internal resistance or ohmic loss (I is the cell current. R is the internal resistance of the cell) and η_a and η_c the anode and cathode polarization, respectively. Ohmic losses result from the resistance of the electrolyte and other cell components. Polarization (over potential) losses are associated with the electrochemical reactions taking place at the interface between the electrodes and the electrolyte.

4.1 REACTIONS AT ANODE

The electrochemical oxidation of hydrogen on nickel anodes in contact with YSZ electrolytes has not received much attention in SOFC development probably because polarization losses at SOFC anodes are believed to be small in comparison with those from the reduction of oxygen at cathodes. The hydrogen oxidation at nickel electrodes produces water, according to the following reaction:



It has been generally assumed that the nickel metal plays the catalytic role in the oxidation. The influence of electrode materials on the electrochemical characteristics of the hydrogen reaction at 1000°C has been demonstrated [19, 20]. The catalytic effect of the electrode surface is also supported by the observation that an increase in electronic carrier concentration enhances the reaction rate [21].

5. RESULTS

1. [Yunhui Gong, Weijie Ji] Characterization of YSZ powders:

The original YSZ powder without any treatment consists of spherical granules with diameters in a range of 5–60 μm. These granules appear to be soft agglomerates of smaller primary particles as shown in the inset. The mean diameter of the original YSZ particles calculated by the SPIP software is 150 nm. These results indicate that high-energy ball milling can thoroughly break the agglomerates in the original powder [22].

2. [Yunhui Gong, Weijie Ji] Morphologies of the modified YSZ electrolytes:

The surface morphology of YSZ electrolyte is continuous and smooth. The surface microstructure of electrolyte modified by the fine YSZ powder is presented. The well-sintered parts of the inter layer indicate that the fine YSZ particles have come through the sintering and densification processes.[22]

3. [Yunhui Gong, Weijie Ji] Microstructures at the interfaces between electrolyte and cathode:

The LSM and YSZ particles connected well with each other in all the composite cathodes. However, the microstructures at the interfaces of electrolytes and cathodes are different This

interlayer seems to separate the cathode and the electrolyte, instead of increasing the effective surface area of the YSZ electrolyte, the original YSZ modified electrolyte shows no differences in the cross sectional microstructures as compared to the untreated samples [22].

4. [Chao Jin, Jiang Liu] Shrinkage and microstructure results with different pore-former:

Inorganic ceramic membrane fabricated by the phase-inversion method usually shows a typical sandwich structure, namely: the two surfaces form the dense membrane structure, the middle forms the micropore structure. Such a structure is not suitable for SOFC anode application. Therefore, in order to obtain a uniform porous structure, which is required by SOFC anode, we used pore-former with phase-inversion method and achieved a satisfying result.[23]

5. [Chao Jin, Jiang Liu] Effect of porosity on the tubular resistance:

A slow increase of the resistance with an increase of the porosity when the porosity is less than 55%. However, when the porosity is greater than 55%, the tubular resistance rapidly increases. The phenomenon can be explained as follows: in anode microstructure, pores will be present in the solid phase that is formed by the Ni and YSZ particles after the pore-formers burn out. [23]

6. [Chao Jin, Jiang Liu] Electrochemical properties analysis of tubular anode-supported SOFC's:

In our review, we also found that the power density of tubular anode-supported SOFCs decreased with an increase of length. In order to study this phenomenon, four single cells were fabricated at different locations along a 15 cm long tubular NiO-YSZ anode-support and their electrochemical properties were determined. [23]

7. [Shuying Zhen, WangSun] Characterization of NiO-3YSZ and NiO-8YSZ hollow fibers:

It can be seen that the cross section of the two hollow fiber walls both present asymmetric structure. A finger-like porous structure was formed near both surfaces of the hollow fiber wall and a small sponge-like porous structure was observing the middle. The formation of a finger-like porous structure can be explained by the viscous fingering phenomenon. [24]

8. Discharge properties:

The voltage and power density curves for MT-SOFC's with Ni-3YSZ (markedasN3Y) and Ni-8YSZ (markedasN8Y) hollow fibers as a function of current density at 800°C demonstrating maximum power densities of 0.53m and 0.67Wcm², respectively .The open circuit voltage (OCV) values of the two cells are above 1.1Vat800 °C, and close to the theoretical value calculated from the Nernst equation. This high OCV value syndicate that the electorally sufficiently dense and the cell is well sealed. [24]

9. [T. Mahata, S.R. Nair, R.K. Lenka] Fabrication of Ni-YSZ anode supported tubular SOFC.

For the fabrication of Ni-YSZ anode supported tubular SOFC the commercial YSZ powder, NiO powder and LSM powders used in this work. In this study, thin YSZ electrolyte layer is required to be integrated with tubular NiO-YSZ substrate by co-firing. The co-firing temperature has been reported to be in the temperature range of 1300-1400 OC. Powder was required to be sintered above 1400 OC. Both anode and cathode need to be porous and there should be electrical percolation through conducting phases. In this investigation, NiO and YSZ were taken in the weight ratio 1:1.

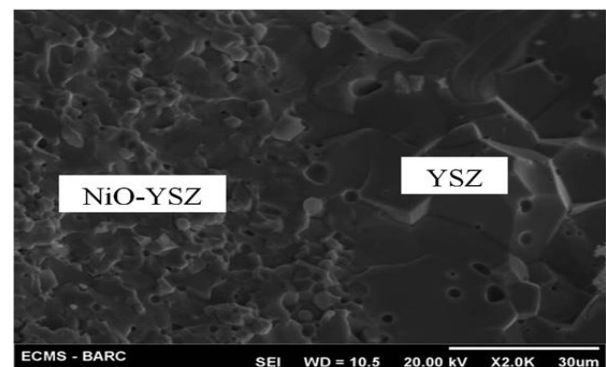


Fig. 2 Microstructure of NiO-YSZ support integrated with YSZ layer by co-pressing and co-firing. [25]

The results indicate that the pores in sintered NiO-YSZ are predominantly closed pores. Presence of these pores has been found to be beneficial in increasing the overall porosity in the reduced Ni-YSZ anode structure. The total porosity in Ni-YSZ anode structure has been estimated to be about 20%. Therefore the conductivity is increases of cell. The cathode consists two layers: one LSM-YSZ composite cathode functional layer in adherence with YSZ electrolyte layer and LSM cathode current collector layer over LSM-YSZ layer. The average pore size lies between 2 and 3 mm in both the layers. Results of impedance spectroscopy obtained with

LSMYSZ-YSZ-LSMYSZ symmetric cell at different temperatures. The electrode polarization resistances (R_p) are 2.68, 1.18 and 0.55 $\Omega \text{ cm}^2$ at 800, 850 and 900°C respectively. [25]

3. CONCLUSIONS

The surface of the YSZ electrolyte was modified with a thin YSZ inter layer. The interface modified with 1200 °C pre-sintered coarse YSZ particles had a positive effect on the electrochemical performance, which was attributed to a better electrolyte–cathode interface contact; on the contrary, the interface modified with high-energy ball milled fine YSZ particles reduced the electrolyte–cathode contact and had a negative effect on electrochemical performance.

In this work, Ni-3YSZ and Ni-8YSZ hollow fibers were prepared by phase inversion. The flexural strength, electrical conductivity, shrinkage and porosity of Ni-3YSZ and Ni-8YSZ hollow fibers were systematically investigated. Of these the Ni-3YSZ hollow fibers exhibited improved flexural strength, porosity and shrinkage properties while the electrical conductivity decreased when compared to Ni-8YSZ hollow fibers. Single cells with Ni- 3YSZ and Ni-8YSZ hollow fibers as the anode-supported were successfully fabricated resulting in maximum power densities of 0.53 and 0.67 Wcm^{-2} at 800 °C, respective

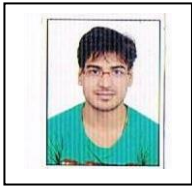
The results suggest that the electrode electrolyte interface has good adherence and reasonably low area specific resistance. Ni-YSZ-YSZ interface developed by co-pressing and co-firing at 1350 °C gives area-specific polarization resistance of 0.79 cm^2 at 900 °C in humidified H_2 atmosphere. The tubular cell generated an OCV of 0.83 V at 900 °C and a maximum power density of 62 mWcm^2 was obtained. Overall, the present study establishes that compressing and co-firing approach can be a viable technique for fabrication of Ni-YSZ anode supported tubular SOFC

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BIOGRAPHIES



1. Pratik S. Rane, U.G. Student. Diploma in Chemical engineering From SMIT Jalgaon (2014), Pursuing B.E. in Chemical (2017) B.V.C.O.E.N.M.



2. Ravi V. Dubey, U.G. Student. Diploma in Chemical engineering From BVIT Navi Mumbai (2014), Pursuing B.E. in Chemical (2017) B.V.C.O.E. Navi Mumbai



3. Dilip B. Rawal, U.G. Student. Pursuing B.E. in Chemical (2017) B.V.C.O.E.N.M



4. Manoj B. Mandake, Asst. Prof. B.V.C.O.E.N.M. (12 years of experience) B.E. Chemical (2004), M.E. Chemical (2012), From T.K.I.E.T Warananagar, Shivaji University, Kolhapur. PhD Pursuing From ICT Mumbai, Waste Water treatment.