A Comparative Review analysis of Various Materials for Solid Oxide Fuel Cells

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Abstract

Solid State Oxide Fuel Cells (SOFC) offer clean and efficient power generation systems with good fuel flexibility, high power density, low pollution, modularity and high energy conversion efficiency. The conventional SOFCs operate at a higher temperature of about 1000 °C. This limits the use of materials to be used for their construction. Presently, the major research effort ongoing is to develop the low cost intermediate temperature (IT) SOFC. These cells work in the temperature range of 500–800 °C with power densities as high as possible. The main difficulty in developing these SOFCs is the degrading performance of fuel cells owing to lower ion conduction of the electrolyte. The selection of materials for different cell components is based on sufficient chemical and structural stability at intermediate temperatures, suitable electrical-conducting properties for various cell functions, minimal reactivity among cell components and matching thermal expansion of various components of SOFC and various designs of SOFC have been compared. Keywords: SOFC, anode, cathode, electrolyte, interconnect

I. Introduction

Solid state oxide fuel cell (SOFC) technology has made significant progress in the recent years. It promises to revolutionize electric power generation in the 21st century. The biggest advantages of SOFC are high efficiency and environmental friendliness [1]. SOFCs have several additional advantages over conventional power generation systems some of which include high power density, fuel flexibility, modularity, low emissions of CO_2 , NO_X , CO, SO_2 , etc. [2–4]. SOFCs have been developed as a practical high temperature fuel cell technology. Since there is no liquid electrolyte, therefore corrosion of its supporting materials is prevented which reduces the problem related to the electrolyte management [5].

The operating temperature of solid oxide fuel cells is above 900 0 C. Because of the high temperature operation, the exhaust heat is of good quality. The multidimensional applications like space heating, household hot water and process steam utilize the waste heat produced, during the operation of solid oxide fuel cells [13].

At the high working temperature of SOFC, the fuel cell operating environments affect the chemical stability of its cell components which may lead to corrosion of components and undesirable phase formation at interfaces between different cell components (electrolyte, anode, cathode and interconnect). To solve these problems, the working temperature of SOFC should be lowered to around 700–800 °C as reported by Badwal *et al.* [6]. If the working temperatures of SOFCs are lowered, it would lead to the development of its thermal stability and reduce startup time. As a result, they can be used as auxiliary power units for automobiles [2, 10, 16]. Many studies aim at reducing SOFC operating temperatures. But the lowering of the operation temperature leads to reduction in the ionic conduction of the electrolyte and hence the power density. To solve these problems, alternate low cost component materials and designs need to be broadly explored that remain stable and durable even at these temperatures [22].

SOFC COMPONENTS AND ITS DESIGNS

The single solid oxide fuel cell basically comprises anode, cathode, an electrolyte, interconnects and sealing materials in the solid state [6]. The ceramic oxide electrolyte shows conduction at high temperature of about 1000 °C. Figure 1 shows the working principle of SOFC. The fuel is fed to the anode where it undergoes an oxidation reaction. The electrons so released move to the external circuit. Oxygen ions are formed due to reduction at the cathode. The flow of electrons in the external circuit from the anode to the cathode leads to the production of electricity [43].

SOFC can be designed as (a) planar (b) tubular and (c) monolithic. In terms of stack design, planar and tubular design cells are more popular. The planar SOFC may be in the form of a circular disk or in a square shape.

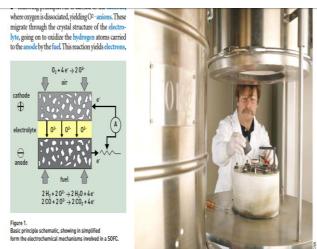
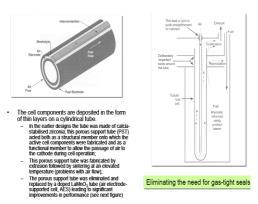


Fig. 1: Working Principle of SOFC.

The fuel is fed from the central axis in the case of circular disk or from the edges if it is of a square shape. The tubular SOFC may have varying diameter: a large diameter (> 15 mm), or much smaller diameter (< 5 mm). Figure 2a illustrates typical tubular cell bundle and Figure 2b planar cell stacks.

One of the inbuilt advantages of tubular SOFC is that the air and the fuel are naturally isolated as one end of the tube is closed; therefore, no seal is required. However, in the case of planar cell stacks, there must be a provision of a seal for the isolation of fuel and air.

Due to no seal requirement for the tubular cells, they may perform well for several years. Nevertheless, they lag behind the planar cells in respect of lower volume power density, and higher manufacturing cost [11].



Large tubular SOFC design 📑

Fig. 2a: Tubular SOFC.

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Cell and stack designs

• Planar SOFC design

 Cell componets are configured as flat plates which are connected in electrical series:

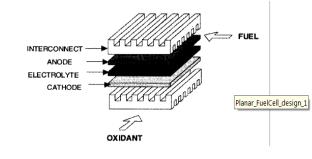


Fig. 2b: Planar SOFC.

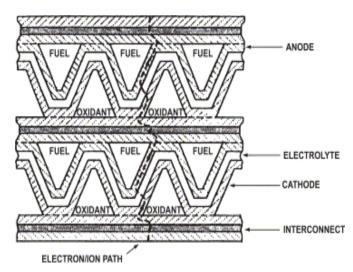


Fig. 2c: Monolithic SOFC.

A monolithic design of SOFC was first suggested in the early 1980s at Argonne National Laboratory (ANL) [26]. In this design, various components of the cell are fabricated in the form of thin layers. In the co-flow version, the cell resembles an array of adjoining fuel and oxidant channels, looking like a honeycomb. There are two types of laminated structures in a cell:

- Anode/Electrolyte/Cathode
- Anode/Interconnect/Cathode

Anode/Electrolyte/Cathode composite is uneven and is stacked alternately between flat Anode/Interconnect/Cathode (Figure 2c). While the monolithic SOFCs present the highest power density out of all the possible SOFC designs, their fabrication has proven to be a matter of concern because it deals with co-sintering of the cell components at high temperatures [12].

SOFC MATERIALS – A COMPARATIVE STUDY

By the end of 19th century, Zirconia with Yttria in solid solution (YSZ) was identified by Nernst [37]. It was used as an electrolyte in an SOFC in 1937 by Baur and Preis [38]. As high temperature was a compulsion for

successful performance of this electrolyte, a conducting oxide as cathode in contact with air was demanded. A cobalt perovskite was reported in 1966 [39], and the well-known lanthanum–strontium manganite (LSM) was introduced in the 1970s [40]. A composite of nickel and electrolyte material as anode was introduced in 1964 [41]. After a detailed study of candidate materials, Siemens Westinghouse adopted this set of materials in 1980s for the tubular cell configuration [42] and thereby proved to be a standard [28]. Table 1 shows the comparison of cell component technology for tubular SOFCs:

Comp-onent	Ca. 1965	Ca. 1975	Ca. 1998 ^a	At present
Anode	Porous Pt	• Ni/ZrO ₂ cermet ^a	 Ni/ZrO₂ cermet^b Deposit slurry, EVD fixed^c 12.5 × 10⁻⁶ cm/cm °C CTE ~150 μm thickness 20 to 40% porosity 	 Copper cermet anodes LSFCO–GDC LSCM LST
Cathode	Porous Pt	• Stabilized ZrO ₂ impregnated with praseodymium oxide and covered with SnO doped In ₂ O ₃	 Doped lanthanum magnate Extrusion, sintering ~2 mm thickness 11 × 10⁻⁶ cm/cm °C CTE from room temperature to 1000 °C 30 to 40% porosity 	 LSCF Sm_{0.5}Sr_{0.5}CoO₃ Lanthanum cobaltites
Electrolyte	Yttria stabilized ZrO ₂ 0.5 mm thickness	• Yttria stabilized ZrO ₂	 Yttria stabilized ZrO₂ (8 mol% Y₂O₃) EVD^d 10.5 × 10⁻⁶ cm/cm °C CTE from room temperature to 1000 °C 30 to 40 μm thickness 	 ZrO2 – Y2O3 LSGM SNDC
Interconnect	• Pt	• Mn doped cobalt cromite	 Doped lanthanum chromite Plasma spray 10 × 10⁻⁶ cm/cm °C CTE 	CermetsFerritic steels

 Table 1: Evolution of Fuel Cell Technology for SOFCs [44].

a – Specification for Siemens Westinghouse SOFC; $b - Y_2O_3$ stabilized ZrO_2 . c – "Fixed EVD" means additional ZrO_2 is grown by EVD to fix (attach) the nickel anode to the electrolyte. This process is expected to be replaced; d - EVD = electrochemical vapor deposition.

Cathode

The cathode is an electrode of SOFC where electrochemical reduction of oxygen takes place. The basic requirement for cathode can be summarized as:

- Comparatively simple fabrication method and low cost;
- Sufficient porosity to allow the diffusion of oxygen and high catalytic activity to facilitate oxygen reduction;
- Chemical compatibility with other cell components under working conditions;
- Matching thermal expansion coefficients (TEC) and large triple phase boundary (TPB);
- High electronic and ionic conductivity [4, 12, 13].

Ralph *et al.* reported that at high temperatures, LSM proves to be the best option as cathode material for YSZ based SOFCs. At low temperatures, LSF and layered structured material YBaCuO are the reasonable cathode materials for YSZ based SOFCs. For Ceria based electrolytes, $Gd_{0.8}Sr_{0.2}CoO_3$ is the best candidate at low temperature. Also BSCaCuO was tried for the first time showing better areal resistance and low activation energy [26].

MvEvoy *et al.* mentioned that mixed conductors like lanthanum strontium cobaltite are preferred to LSM due to the formation of an insulating interphase of lanthanum Zirconate [28]. Badwal *et al.* reported that Co or Ni may replace Mn providing faster kinetics at low temperatures for zirconia based electrolytes in spite of strong reactivity. LaSrCoO₃ or LaSrCoFeO₃ electrodes and the intermediate phase formation by thin layers of doped ceria prove to be promising options for Ce based electrolytes [6]. Haile *et al.* emphasized the search for transition metal perovskites in order to replace LSM at high temperatures like lanthanum cobaltites but they are also found unsuitable for YSZ based SOFCs at elevated temperatures. Recently, LSCF and Sm_{0.5}Sr_{0.5}CoO₃ have emerged as reasonable cathode candidates at low temperatures [33]. Singhal *et al.* showed that doped lanthanum manganite satisfies good qualities of a cathode at elevated temperatures. At lower temperatures, polarization losses pose a great problem towards cathode stability. So, the concept of composite cathode was introduced [11]. Taroco *et al.* recently mentioned that the choice of electrolyte decides the choice of cathode. The concentration of dopants also affects the electronic conductivity as well as TEC of the materials. Doping with Sr on the A-site and with Fe and Co on the B-site in the peroveskite increases both the ionic and electronic conductivities [29]. Also LSCF cathodes are fairly compatible with GDC electrolyte [3].

Electrolyte Materials

The electrolyte is the part of the cell allowing the conduction of ions between the cathode and anode. The basic performance requirements are [4, 12]:

• The oxide-ion conductivity should be more than 10^{-2} Scm⁻¹ at the working temperature and minimum electronic conduction;

- High density so that the gas impermeability is promoted;
- Oxygen partial pressure and thermodynamically stable over a long range of temperature and moderate cost;
- TEC matching with cell components and the concept of thin layers (less than 30 μm);

• For fracture resistance greater than 400 MPa, mechanical properties should be appropriate at room temperature;

The most explored and developed electrolytes have been the zirconia based ceramic materials at the elevated temperatures [3]. According to Bujalski *et al.*, zirconia doped with 8 to 10 mole% Yttria (YSZ)) is still the most effective electrolyte for the high temperature SOFC as zirconia is highly stable in both the reducing and oxidizing environments that are experienced at the anode and cathode, respectively and the ionic conductivity of YSZ (0.02 Scm^{-1} at 800 °C and 0.1 Scm^{-1} at 1000 °C) is comparable with that of liquid electrolytes and it can be made very thin (25–50 µm) ensuring that the ohmic loss in the SOFC is comparable with other fuel cell types. Many other electrolytes such as Bi₂O₃, CeO₂ and Ta₂O₅ have also been investigated with mixed success at high temperatures [31].

Badwal *et al.* mentioned that the materials typically used in SOFC include zirconia, ceria and perovskite like lanthanum gallate doped at both A and B sites. These perovskites are prone to changes in their stoichiometry due to which many impurity phases like La₂Zr₂O₇ and SrZrO₃ are formed. To eliminate this, thin layers of doped ceria are proposed to be introduced between Zr based electrolytes and La(Co, Fe)O₃. But the contact needs low temperature operation for better performance of the cell [6]. Taroco *et al.* reported that doped CeO₂ and doped lanthanum gallate are two candidates in intermediate temperature SOFCs (600–800 °C). Doped ceria offers ionic conductivities about one order of magnitude greater than stabilized zirconia under similar temperature conditions. The Ce_{0.9}Gd_{0.1}O_{1.95} (CGO) composition is reliable for IT-SOFC applications more as it has high ionic conductivity at 500 °C. In case of gadolinium doped ceria (GDC), at elevated temperatures, Ce⁴⁺ converts to Ce³⁺ under the reducing anode atmosphere. This leads to faster consumption of fuel leading to deteriorating cell performance [14, 17]. To avoid the problem, a thin film of YSZ between the Ce electrolyte and the anode is inserted [9, 20]. The electronic conductivity is lower at less temperatures around 500 °C and it has been concluded that this temperature could be an optimal one for ceria based fuel cells [21].

The compositions like $La_{1x}Sr_xGa_{1-y}Mg_yO_{3-\delta}$ (LSGM) have presented high ionic conductivities in oxidizing and reducing atmospheres [3]. But such ceramics show instability under reducing atmospheres and also Ga losses are experienced, thereby leading to new phases formation [21, 30]. These reasons limit the use of doped lanthanum gallate to be used as electrolyte in SOFC.

The comparison of various electrolyte materials has been depicted in Table 2 in terms of their ionic conductivities and thermal expansion coefficients:

Composition	or at obv C (Sem)	
$(Y_2O_3)_{0.08}(ZrO_2)_{0.92}$	10.5	0.03
$(Sc_2O_3)_{0.08}(ZrO_2)_{0.92}$	10.7	0.13
$Ce_{0.8}Gd_{0.2}O_{1.9}$	12.5	0.053
Ce _{0.8} Sm _{0.2} O _{1.9}	12.2	0.095
$La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{2.85}$	10.7	0.1

 Table 2: Ionic Conductivity and TEC of Suggested Electrolyte Materials in Air at 800 °C [29].

 Composition

 gi at 800 °C (Scm⁻¹)

 TEC (x10⁻⁶ K⁻¹)

Other options for use in IT-SOFC are salt-oxide composites and NANOCOFC materials (nanocomposites for advanced fuel cell technology) [17, 18]. Ralph *et al.* investigated the high temperature ceramic electrolyte material, Bi_2O_3 which has the highest ionic conductivity because its crystal structure is quite spacious. All the bismuth based electrolytes have the problem that Bi^{3+} reduces to Bi metal that proves fatal to the electrolyte at high oxygen partial pressures [32]. The stability of low temperature candidate LSGM with some fuels is yet unknown. So, YSZ proves to be the best electrolyte material at high temperatures, but at low temperatures, doped ceria remains the only option [26]. Steele *et al.* reported that the specific conductivity of a solid electrolyte varies as the reciprocal of temperature, e.g., the attainment of target value of ionic conductivity of YSZ at about 700 °C predicts its suitability to be used as a promising material at high temperatures, whereas

CGO attains the same at about 500 °C that makes it suitable to be used at low temperatures successfully. They added that when the thin electrolyte films are used, the operating temperature of SOFC may be lowered [5]. Singhal *et al.* concluded that to make the stabilized zirconia operational in the low temperature regime, either the thickness of YSZ electrolyte may be decreased or Y may be replaced with other acceptors, e.g., scandia doping, but the high cost of scandium puts a question mark. Also at high temperatures above about 600 °C, ceria based electrolytes possess electronic conductivity which may deteriorate the performance of the cell because of problems associated with the PEN (positive electrode-electrolyte-negative electrolyte) structure [14].

These problems are solved at low temperatures successfully. They also listed some of the drawbacks of LSGM as an electrolyte, viz., its stability and the cost of constituents. Even then LSMG is being used as a promising electrolyte at Mitsubishi Materials Corporation, Japan [11]. Haile *et al.* raised a question on the long term feasibility of ceria based SOFCs because of its high electronic conductivity though it has good ion transport properties, and the internal stress resulting from the chemical expansion of ceria during reduction processes. They added that LSGM is restricted to a temperature range of about 700–1000 °C. To avoid the reactivity of lanthanum gallate with nickel, the buffer layers of ceria have been used between the anode and the electrolyte [33]. Chao *et al.* reported that to develop IT-SOFC, either alternate materials having higher ionic conductivity like LSGM having maximum power density of 0.612 W/cm² at 500 °C and Sm_{0.075}Nd_{0.075}Ce_{0.85}O_{2-δ} (SNDC) having the maximum power density 0.32 W/cm² at 500 °C [27] but less stable as compared to Yttria-stabilized zirconia (YSZ) or to reduce electrolyte thickness so as to the improve the fuel cell performance at low temperatures [22].

They also designed a corrugated membrane of thin film electrolyte by using the technique of nanosphere lithography and the polarization and ohmic losses were reduced by atomic layer deposition (ALD) at low temperatures. The hexagonal-pyramid array nano-structure was observed for the resulting micro-SOFC electrolyte membrane and a power density of 1.34 W/cm²at 500 °C was obtained [22]. Ramesh *et al.* demonstrated that co-doped ceria electrolytes offer high ionic conductivity for SOFCs at the intermediate temperatures [34].

Anode Materials

The anode is required to provide reaction sites for the electrochemical oxidation of the fuel gas. An appropriate anode has:

- A TEC same as that of the adjoining components and chemical compatibility with them;
- Fine particle size and the capacity to avoid coke deposition;
- Large triple phase boundary and high porosity (20–40%);
- High electrochemical activity for the oxidation of the selected fuel gas;
- Good electronic and ionic conductivity [12, 15].

Singhal *et al.* investigated that Ni-YSZ remains the best choice as anode material despite its high catalytic activity that limits its use for higher hydrocarbons where alternative materials like ceria or strontium titanate and copper cermet anodes for IT-SOFC may be used. Also ceramic anodes like doped perovskites along with ceria have been suggested despite some drawbacks [11]. Steele *et al.* reported that the use of nickel metal may lead to a deleterious volume reaction due to the formation of NiO. So redox stable and electronic conducting oxides may replace Ni or alternative anodes for anodic oxidation of hydrocarbons directly may be used at intermediate temperatures [14]. Badwal *et al.* in 2001 reported that the stability and performance of anode at the operating temperatures is greatly influenced by microstructure of the cermet. Alternative materials need to be explored to avoid the deteriorating anode performance due to possible Ni particles agglomeration and Ni dewetting of zirconia [6]. Haile *et al.* investigated that it is compulsory to limit the concentration of Ni to avoid coking and ceria must be incorporated into the anode cermet for better anode performance [33].

Taroco *et al.* demonstrated that Ni to YSZ volume ratio affects the conductivity of the material. The Ni/YSZ composite is not a good option for ethanol, methanol like fuel based SOFCs, because of its low acceptance to sulfur and particulate matter deposition. To avoid this the operating temperature can be reduced or by the choice of alternative anode materials like Cu-anodes alloyed with a another metal like nickel or new perovskite materials like LSFCO–GDC and LSCM or LST (for IT-SOFC) [3]. Ringuede *et al.* showed that SOFCs which utilize Ni–YSZ cermet anodes are prone to sulphur poisoning as low as 2 ppm H₂S at 1273 K. In their work it was found that the performance loss can be reversed at H₂S concentrations to less than 15 ppm [23]. Ralph *et al.* reported that Ni-YSZ cermet is the most commonly used anode material that need proper equipment to remove sulphur from the fuels to avoid the poisoning of Ni sites. Due to percolation threshold of YSZ, CGO and CSO are being investigated as cermets with Ni microstructure [26]. McEvoy *et al.* explained the ongoing efforts to replace Ni with Cu or Co. The studies are on to investigate oxide based anodes such as reduced ceria or electronically conducting pervoskites [28].

Interconnect

Interconnect is used to connect two fuel cells together so as to produce sufficient current and voltage. An interconnect must have the following characteristics:

- 100% electrical conductivity.
- No reaction with other cell components.
- Zero porosity.
- Thermal expansion compatibility.

Lanthanum chromite is the most commonly used material for interconnect of ceramic based SOFCs because it can withstand even unfavorable working conditions though it suffers from the problem of fabrication according to Ovenstone et al. [36]. Matasuzaki et al. reported that at elevated temperatures of about 900-1000 °C, interconnects like Inconel600 (a nickel based alloy) may be used [25]. McEvoy et al. suggested the optimum design target temperature of ferritic steel interconnector at about 700 °C [28]. Ralph et al. declared that ferritic steels may be efficiently used at or below 800 °C as they are cheaply and easily available. Also, the oxide scale problem can be overcome by either alloying the ferritic steel alloys by Ni or Co ions, or by coating the surface with a conductive ceramic [26]. Singhal et al. mentioned that at the high temperatures above 800 °C, doped lanthanum chromite ceramic is found to satisfy above requirements of the interconnect, but at temperatures less than 800 °C metallic alloys may be used efficiently in terms of low cost and easy manufacture, e.g., ferritic stainless steels like crofer22 and ZMG232. To solve the problems of Cr evaporation, electrical resistance and corrosion when exposed, surface coatings can be applied over the metallic alloy interconnects [11]. Badwal et al. suggested thick impermeable conductive and protective coatings having good chemical compatibility, and low Cr and oxygen ion diffusion coefficients on the metallic interconnect materials [6]. Tietz et al. studied the effect of applying the protective layers of LaCrO₃ in lowering the chromium vapor deposition on the cathode. Gettering may also help in neutralizing the Cr atoms in the coated layers.Cr₅FelY₂O₃. Chromium based interconnect has shown this recent development [9].

Effect of Processing on the Properties of Component Materials

Ovenstone et al. prepared three perovskite ceramics Ca doped lanthanum chromite, strontium doped lanthanum chromite and strontium doped lanthanum manganite by emulsion processing. EDX has proved that unsinterable powder may be formed due to small stoichiometric changes [36]. Tietz et al. reported that cost, automation, precision of various techniques and reproducibility are the factors that determine the fabrication processes. Various methods like tape casting, screen printing, warm processing, wet powder spraying and tape calendaring, laser ablation, colloidal deposition, CVD, electrophoresis or multiple spin coating may be used [9]. Suciu et al. tried a new modified sol-gel method for preparing 7SvSZ and 11ScSZ nanoparticles to be used as electrolytes by using sucrose and pectin. Their conductivities were found to vary at temperatures lower than 400 °C [35]. Bujalski et al. discussed a number of techniques for making thin (5–20 μm) YSZ electrolytes, viz., sputtering, dip coating, spin coating, spray pyrolysis, electrophoretic deposition, slip casting, plasma spraying, electrostatic assisted vapor deposition, vacuum evaporation, laser spraying, transfer printing, sedimentation method, and plasma metal organic chemical vapor deposition [31]. Boldrini et al. prepared La_{0.80}Sr_{0.20}Ga_{0.83}Mg_{0.17}O_{2.815} by two methods – a conventional and a microwave assisted sol-gel Pechini method. MWA-SGP method showed ionic conductivity values higher than those checked for SGP samples. This clearly explains the vital role of microwave processing on lesser time and costs, but it also improves the electrolyte properties [24]. Chao et al. reported that MEMS allows the fabrication of very thin electrolytes. Various techniques like physical vapor deposition and atomic layer deposition were used to reduce the YSZ electrolyte thickness to tens of nanometers [22].

Various processing techniques for different fuel cell types are suggested in Table 3:

Design	Fabrication method electrolyte	Electrode	Interconnect
Tubular Concept	CVD/EVD, plasmaspraying	Slurry coating, plasma spraying, CVD/EVD	EVD, plasma spraying
Monolithic	Calendar rolling, laminating, co-sintering	Calendar rolling, laminating, co-sintering	Calendar rolling, laminating, co-sintering
Planar	Tape casting, calendar rolling	Screen printing, slurry coating	Ceramic or metal processing

II. CONCLUSIONS

The work of SOFCs acknowledges and emphasizes man's endeavor to adapt to reduce the cost, to generate power densities as high as possible and for the long term durability and reliability. For these newer areas of exploration, a number of materials have been explored and tested that can replace the conventional

materials without deteriorating the fuel cell performance. Apart from the composition, optimization of synthesis process is further needed in the microstructure in order to fabricate the SOFCs cost effectively.

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