



Review article

Electrolyte bi-layering strategy to improve the performance of an intermediate temperature solid oxide fuel cell: A review



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HIGHLIGHTS

- Limitations of single electrolyte layered SOFC at low temperatures.
- Strategy of Bi-layering of electrolytes to overcome the issues.
- Different types bilayered electrolyte systems.
- Fabrication and performance of the bilayered cells.

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ABSTRACT

Lowering of operation temperature has become one of the primary goals of solid oxide fuel (SOFC) research as reduced temperature improves the prospects for widespread commercialization of this energy system. Reduced operational temperature also mitigates the issues associated with high temperature SOFCs and paves way not only for the large scale stationary power generation but also makes SOFCs viable for portable and transport applications. However, there are issues with electrolyte and cathode materials at low temperatures, individually as well as in association with other components, which makes the performance of the SOFCs less satisfactory than expected at lowered temperatures. Bi-layering of electrolytes and impregnation of cathodes have emerged as two important strategies to overcome these issues and achieve higher performance at low temperatures. This review article provides the perspective on the strategy of bi-layering of electrolyte to achieve the desired high performance from SOFC at low to intermediate temperatures.

1. Introduction

Development of high performance clean energy systems is critical for solving energy and environmental problems of the world. Fuel cells have been increasingly accepted as efficient chemical to electrical energy conversion systems with a low environmental impact [1]. Among the wide group of fuel cells, solid oxide fuel cells (SOFCs) are of special interest due to their all solid construction, fuel versatility and high efficiency. Based on the operation temperature range, though various SOFC temperature range definitions have been proposed, SOFCs are generally classified into high temperature SOFC (> 850 °C, HT-SOFC), intermediate temperature (650–850 °C, IT-SOFC) and low temperature SOFC (< 650 °C, LT-SOFC) [2]. State-of-the-art HT-SOFCs, built using 8 mol% yttria stabilized zirconia (YSZ) based electrolyte, $\text{La}_{1-x}\text{Sr}_x\text{MnO}_{3-\delta}$ (LSM) based cathode and Nickel/YSZ (Ni/YSZ) cermet anodes, function effectively only at high operating temperature ranges (> 800 °C) [3,4]. Low ionic conductivity of YSZ electrolytes and poor catalytic

activity of LSM electrodes at reduced temperatures demands high operating temperature (> 850 °C) for SOFCs. Such a high temperature systems are mostly appropriate for large-scale stationary applications and improved efficiency from these systems can be achieved through integration with gas turbines. However, high operating temperature imposes severe restrictions on fuel cell components, thereby hindering the development and large scale deployment of this transformative technology. High operating temperature demands that stack components to be predominantly of ceramics and balance-of-plant components to be of high temperature metal alloys. Also, high operating temperature causes considerable disadvantages, such as, short lifetime of the cell due to the thermal degradation/corrosion of the materials [5,6], side reactions occurring at the electrode/electrolyte interface, microstructural modification in electrodes [7], slow start-up and shut-down cycles, issues related to sealants, concerns about operational safety, higher systems costs, etc. These system issues dramatically limit the applicability of SOFCs in portable power and transportation

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markets. For applications, where integration with a heat engine is not an aspect, there is a trend to move towards lower operation temperatures. Lowered operational temperature reduces the corrosion rate of stack and system components, shortens the start-up time, allows for robust construction through the use of compressive seals, leads to cheaper fabrication, particularly in relation to interconnects and balance-of-plant. Reduced operating temperature of SOFCs thus unlocks a wider range of potential applications, including few that have previously been the domain of low temperature fuel cells (such as polymer electrolyte membrane (PEM) fuel cells).

Thus lowering the operating temperature of SOFCs has become one of the main SOFC research goals. Nonetheless, reduced operation temperatures are not devoid of any negative implications. Firstly, very low operational temperature ($< 650\text{ }^\circ\text{C}$) precludes the possibility of direct usage of hydrocarbons and necessitates the additional external processes for the conversion of hydrocarbon into H_2 , a penalty which causes the overall decrease in the system efficiency. In addition, thermally activated cell polarization losses increase with the decrease of temperatures and thus bringing in vast difference between the attained and theoretical efficiency. Thus there is a greater tendency to operate the cell at intermediate temperature range of ($700\text{--}750\text{ }^\circ\text{C}$), wherein there is a scope for the direct usage of hydrocarbon and use of the material (electrolyte and electrodes) with improved conductivity and catalytic activity [8]. In this context, it is interesting to note that a reasonably good performance has been demonstrated for protonic ceramic fuel cells (PCFCs) (142 mWcm^{-2} at $350\text{ }^\circ\text{C}$) in the methane fuel atmosphere. These cells have been fabricated by employing special cathodes and anodes and fabrication processes. At these temperature ranges, cheaper metallic interconnects could also be used instead of ceramic components [9].

As mentioned earlier, with decrease in temperature, electrolyte becomes the major source of polarization loss in SOFC due to increased resistivity. Fig. 1 shows the variation of conductivity of typical O^{2-} ion conducting electrolyte materials as a function of temperature. In order to compete with PEM fuel cells, desired power density from SOFC is expected to be more than 1 Wcm^{-2} ; required electrolyte area specific resistance (ASR) for the same is $< 0.1\text{ Ohmcm}^2$. For an electrolyte thickness of $10\text{ }\mu\text{m}$ (general electrolyte thickness in anode supported cell), YSZ electrolyte reaches this conductivity value (corresponding to ASR of $\sim 0.1\text{ Ohmcm}^2$) only above $750\text{ }^\circ\text{C}$. At lower temperatures, ASR value reaches prohibitively high values.

Main strategy that is adopted for lowering of the SOFC operation temperature is focused around electrolyte as it is the predominant contributor of resistance. Two major routes adopted for decreasing the polarization resistance of electrolytes are:

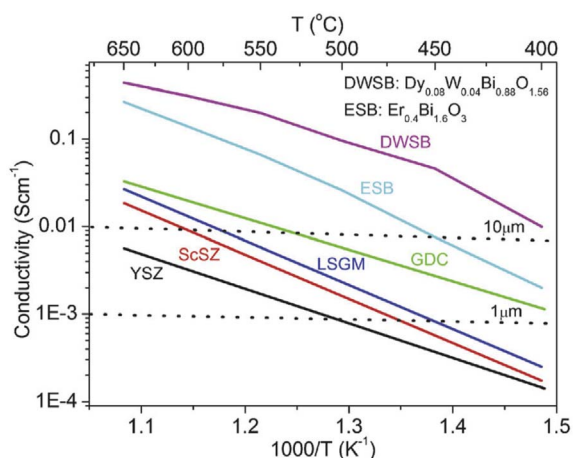


Fig. 1. Variation of conductivities of typical O^{2-} ion conducting electrolyte materials as a function of temperature [10,11].

- (i) Decreasing the dimensional thickness of the conventionally used electrolyte such as YSZ to a practically feasible thickness ($< 5\text{ }\mu\text{m}$) in the anode supported configuration by using thin film techniques such as sputtering, pulsed laser deposition (PLD), etc. [12,13]. For instance, on decreasing the YSZ electrolyte thickness to $\sim 1\text{ }\mu\text{m}$, desired electrolyte ASR value of 0.1 Ohmcm^2 can be achieved at $\sim 550\text{ }^\circ\text{C}$. By adopting thin film techniques, micro fuel cells with electrolyte thickness as low as $\sim 100\text{ nm}$ have been fabricated and good performance has been reported at temperatures as low as $400\text{ }^\circ\text{C}$ with Pt as electrodes. However, manufacturing cost of the thin film electrolyte could dominate the fabrication cost of SOFCs and unlikely that they are scalable and not so cost-effective for mass production.
- (ii) Alternatively, materials with higher O^{2-} ion conductivity such as Scandia stabilized Zirconia (ScSZ), lanthanum gallate related compounds, doped ceria compounds, etc. could be used for the fabrication of electrolyte instead of conventional YSZ. It's evident from Fig. 1 that high conductivity gadolinia doped ceria (GDC) electrolyte reaches the desired ASR value at $525\text{ }^\circ\text{C}$ for a thickness of $10\text{ }\mu\text{m}$ and doped Bi_2O_3 reaches the value at still lower temperature.

Another large contributor to the enhanced cell resistance at low temperatures is the electrode polarization; especially from the cathode side, as the kinetics associated with the oxygen reduction reaction (ORR) and charge transport are much slower at lower temperatures. In cathodes, importance is given for its nano-structuring as enhanced surface area is expected to increase the ORR kinetics. Deployment of the nano-structured composite cathode (for instance, nano-structured LSM/YSZ composite cathodes) formulations is expected to enhance the performance further. Development of alternative cathode compositions that function effectively even at lower temperatures is another alternative.

Despite of nano-structuring, catalytic activity of conventional cathode LSM, a pure electronic conductor, in which electrochemical reaction is mostly confined to the electrolyte/cathode interface, is known to decrease dramatically with decrease in temperature. Composite cathode formulations improve the performance slightly. On the other hand, owing to its high electronic and ionic conductivities as well as high catalytic activity for ORR, mixed ionic-electronic conducting (MEIC) perovskites and related structures are currently the most promising candidates for IT-SOFC cathodes [3]. In the recent past, Bismuth ruthenate based composites have also emerged as a suitable cathode for low temperature SOFC.

Among the MEIC cathodes, the $\text{La}_{1-x}\text{Sr}_x\text{Fe}_{1-y}\text{Co}_y\text{O}_{3-\delta}$ (LSCF) family of compounds has been widely used in IT-SOFCs as they exhibit high electrical ($100\text{--}1000\text{ Scm}^{-1}$) and ionic conductivity ($0.001\text{--}0.1\text{ Scm}^{-1}$) even at lower temperatures. These materials also exhibit higher O^{2-} surface exchange and diffusion rates. On using such materials as cathodes, electrochemically active region extends to a finite width from the electrode/electrolyte interface to the interior of electrode. Electrical/polarization resistance of individual electrolyte/cathode materials, which are of special interest for low and IT-SOFC applications and which are extensively used in fuel cells with bi-layered electrolytes are summarized in Table 1.

2. Characteristics of generally used electrolyte and cathode materials in bi-layered system

2.1. Zirconia-based electrolytes

Zirconia (ZrO_2) based electrolytes have good chemical and mechanical stability over a wide range of oxygen partial pressures and temperatures and hence they are the natural choice as the electrolyte materials. Among ZrO_2 based electrolytes, YSZ has been established as the preferred electrolyte candidate for HT-SOFC and ScSZ is attractive

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