



# High performance zirconia-bismuth oxide nanocomposite electrolytes for lower temperature solid oxide fuel cells



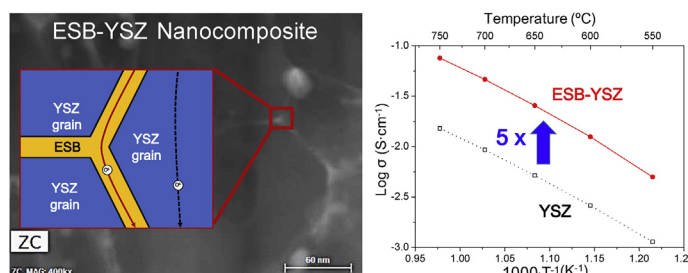
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## HIGHLIGHTS

- A novel nanostructured ESB-YSZ composite electrolyte is developed.
- The nanocomposite electrolyte is sintered as low as 800 °C.
- ESB-YSZ exhibits 5 times higher ionic conductivity than that of YSZ.
- The high ionic conductivity is maintained for ~600 h, demonstrating high stability.
- Oxygen reduction reaction with ESB-YSZ is effectively enhanced by ~70% compared to YSZ.

## GRAPHICAL ABSTRACT



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## ABSTRACT

We develop a novel nanocomposite electrolyte, consisting of yttria-stabilized zirconia (YSZ) and erbia-stabilized bismuth oxide (ESB). The 20 mol% ESB-incorporated YSZ composite (20ESB-YSZ) achieves the high density (>97%) at the low sintering temperature of 800 °C. The microstructural analysis of 20ESB-YSZ reveals the characteristic nanocomposite structure of the highly percolated ESB phase at the YSZ grain boundaries (a few ~ nm thick). The ionic conductivity of 20ESB-YSZ is increased by 5 times compared to that of the conventional YSZ due to the fast oxygen ion transport along the ESB phase. Moreover, this high conductivity is maintained up to 580 h, indicating high stability of the ESB-YSZ nanocomposite. In addition, the oxygen reduction reaction at the composite electrolyte/cathode interface is effectively enhanced (~70%) at the temperature below 650 °C, mainly due to the fast dissociative oxygen adsorption on the ESB surface as well as the rapid oxygen ion incorporation into the ESB lattice. Thus, we believe this ESB-YSZ nanocomposite is a promising electrolyte for high performance solid oxide fuel cells at reduced temperatures.

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## 1. Introduction

Solid oxide fuel cells (SOFC) are the most efficient technologies to directly convert stored chemical energy to usable electricity via an electrochemical reaction [1]. Moreover, their unique fuel

flexibility to operate on conventional hydrocarbon fuels as well as hydrogen potentially provides both current and future energy solutions [2,3]. An SOFC makes up of two porous electrodes (cathode and anode) separated by a solid oxygen ion conducting electrolyte. At the cathode, oxygen molecules are reduced into oxygen ions and incorporated into the electrolyte lattice. At the anode, oxygen ions, which diffuse through the solid electrolyte, react with hydrogen gas molecules, producing electricity with H<sub>2</sub>O (and CO<sub>2</sub> for hydrocarbon fuels) and heat as by-products.

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The yttria-stabilized zirconia (YSZ) is the most popular choice as a solid electrolyte for use in solid oxide fuel cells (SOFCs) due to its reasonably high ionic conductivity as well as high chemical/mechanical durability (>10,000 h) [4].

Over the last two decades, the SOFC R&D has been focused on lowering the operating temperature below 700 °C, which can allow us to effectively reduce the system cost, and improve the long-term stability of SOFC stacks and the balance-of-plants (BOPs) [3,5,6]. At reduced operating temperatures, however, the ohmic resistance of SOFCs is exponentially increased due to the thermally activated nature of oxygen ion migration through the YSZ electrolyte [7].

Moreover, it has been repeatedly reported that zirconia-based solid electrolytes have high reactivity with high performance SOFC components, including doped ceria (e.g., Gd doped ceria, GDC) electrolytes [8,9] and cobaltite-based perovskite cathodes such as  $\text{La}_{1-x}\text{Sr}_x\text{Co}_{1-y}\text{Fe}_y\text{O}_{3-\delta}$  (LSCF) and  $\text{Ba}_{1-x}\text{Sr}_x\text{Co}_{1-y}\text{Fe}_y\text{O}_{3-\delta}$  (BSCF), due to the high processing temperature (>1400 °C) [10,11].

To address above issues, it has been reported that low concentrations (<5 mol%) of sintering agents, such as  $\text{Co}_3\text{O}_4$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{Al}_2\text{O}_3$ , and  $\text{Bi}_2\text{O}_3$ , are effective to lower the sintering temperature of zirconia-based electrolytes by 100–350 °C along with the partial liquid sintering mechanism [12–15]. However, most of the sintering aids are sacrificed or form solid solutions with the host elements during the high temperature process (>1000 °C), leading to the deleterious effects on the ionic conductivity primarily caused by increase in the grain boundary conductivity.

Thus, the rational tailoring of zirconia-based electrolytes to achieve both the higher ionic conductivity as well as the lower processing temperature is essentially important for lower temperature (LT)-SOFC applications. In this respect, we designed a novel nanostructured bismuth oxide (e.g., Er-stabilized  $\text{Bi}_2\text{O}_3$ , ESB) and zirconia (e.g., YSZ) composite electrolyte, resulting in the enhanced performance. Due to the highly functional ESB phase [16,17], such as low sintering temperature (i.e., ~800 °C for ESB vs. ~1500 °C for YSZ) and high ionic conductivity, we can expect

two major benefits from this novel dual-phase electrolyte, as conceptually illustrated in Fig. 1. First, during the heating of the dual-phase mixture, ESB can easily diffuse and surround YSZ particles due to the low melting temperature of  $\text{Bi}_2\text{O}_3$  (~825 °C) and act as ‘glue’ between YSZ grains, thus enhancing the sintering process at much lower temperature (Fig. 1(a)). Moreover, when the ESB phase is properly connected in 3-D in YSZ grain boundaries, the oxygen ion transport can be expedited along it, thus increasing the ionic conductivity of this nanocomposite structure (Fig. 1(b)).

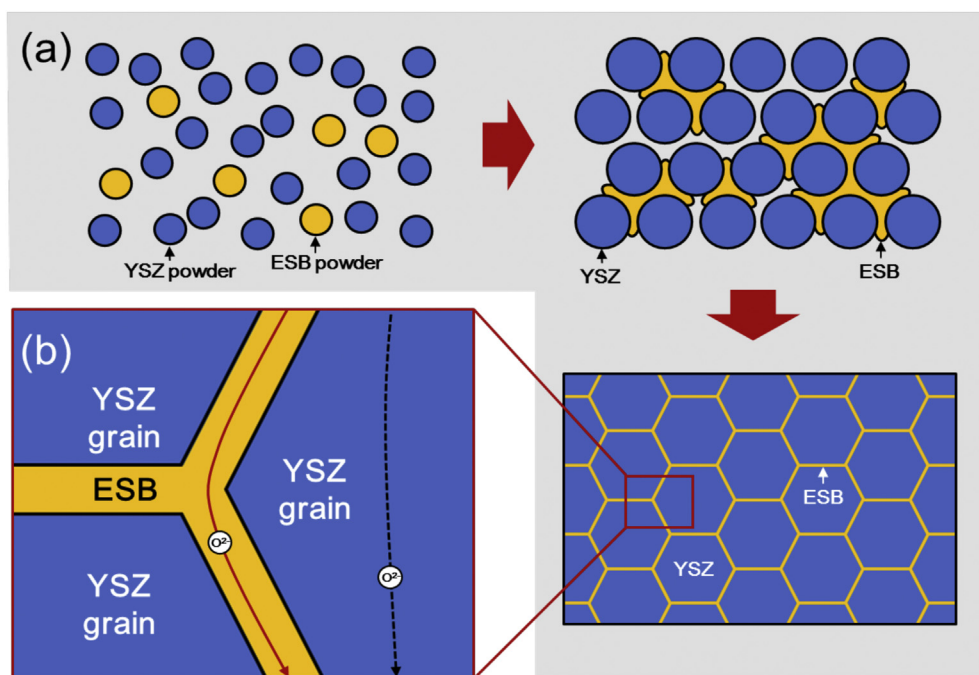
However, in order to realize this concept, the ESB phase should be well-distributed in a few nanometer thick YSZ grain boundaries throughout the electrolyte bulk. Besides, ESB should not be reacted with YSZ nor sacrificed during the high temperature sintering process, although there is great difference in the sintering temperature (~600 °C) between two phases [17].

Herein, we developed ESB-YSZ nanocomposite electrolytes with superior sinterability as well as high oxygen ion conductivity. The sintering behavior and electrochemical performances of the ESB-YSZ nanocomposites were systematically investigated over a wide composition range along with microstructural analysis. In order to demonstrate the feasibility of them as SOFC electrolytes, the long term durability of the ionic conductivity as well as the redox stability of the ESB-YSZ electrolytes were studied. In addition, the effect of ESB incorporation into YSZ on oxygen reduction reaction (ORR) at the cathode/electrolyte interface was examined.

## 2. Experimental

### 2.1. Sample preparation

ESB (20 mol% erbia) powders were synthesized via a conventional solid-state reaction method. A stoichiometric mixture of  $\text{Bi}_2\text{O}_3$  (99.9995% pure, Alfa Aesar) and  $\text{Er}_2\text{O}_3$  (99.99% pure, Alfa Aesar) were ball-milled with zirconia ball media for 24 h. After milling, the mixed powders were calcined at 800 °C for 16 h and



**Fig. 1.** Schematic of the proposed design of the nanostructured ESB-YSZ composite electrolyte. (a) A sintering process of the ESB and YSZ, dual-phase mixture upon heating and the final microstructure of the ESB-YSZ nanocomposite electrolyte after sintering. (b) The magnified nanostructure of the ESB-YSZ electrolyte with the fast oxygen transport path through the ESB phase (solid line) at YSZ grain boundaries compared to the slow oxygen conduction path through YSZ grains (dotted line).

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