



## A Short Review on the Proton Conducting Electrolytes for Solid Oxide Fuel Cell Applications

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### ABSTRACT

The performance of solid oxide fuel cells (SOFCs) mostly relies on the electrolyte component, which is considered the heart of the SOFC. Since intermediate-temperature SOFCs (IT-SOFCs) have become popular, more consideration has been given to the performance of the electrolyte components for proton-conducting SOFCs (H-SOFCs). Potential electrolytes for H-SOFCs must meet the three basic requirements of good sinterability, high chemical stability and high ionic conductivity. Unfortunately, studies have shown that it is very difficult to find a material that meets all three requirements simultaneously. BaZrO<sub>3</sub> and BaCeO<sub>3</sub> are two common proton conducting electrolytes used in H-SOFCs. BaZrO<sub>3</sub>-based proton conductors showed perfect chemical stability in CO<sub>2</sub> and H<sub>2</sub>O atmospheres but have poor sinterability, while BaCeO<sub>3</sub>-based proton conductors showed high proton conductivity and good sintering behaviour but could easily react with CO<sub>2</sub> and H<sub>2</sub>O atmospheres. In contrast, SrCeO<sub>3</sub> has low conductivity due to its lower rate of proton transport. Few literature reports describe efforts to improve the sintering behaviour and conductivity of electrolytes. Thus, this review highlights developing trends in electrolyte sintering behaviour and conductivity, which will support efforts to develop high-performance proton-conducting SOFCs.

#### Keywords:

Solid oxide fuel cell, Proton conducting electrolyte, Sintering behaviour

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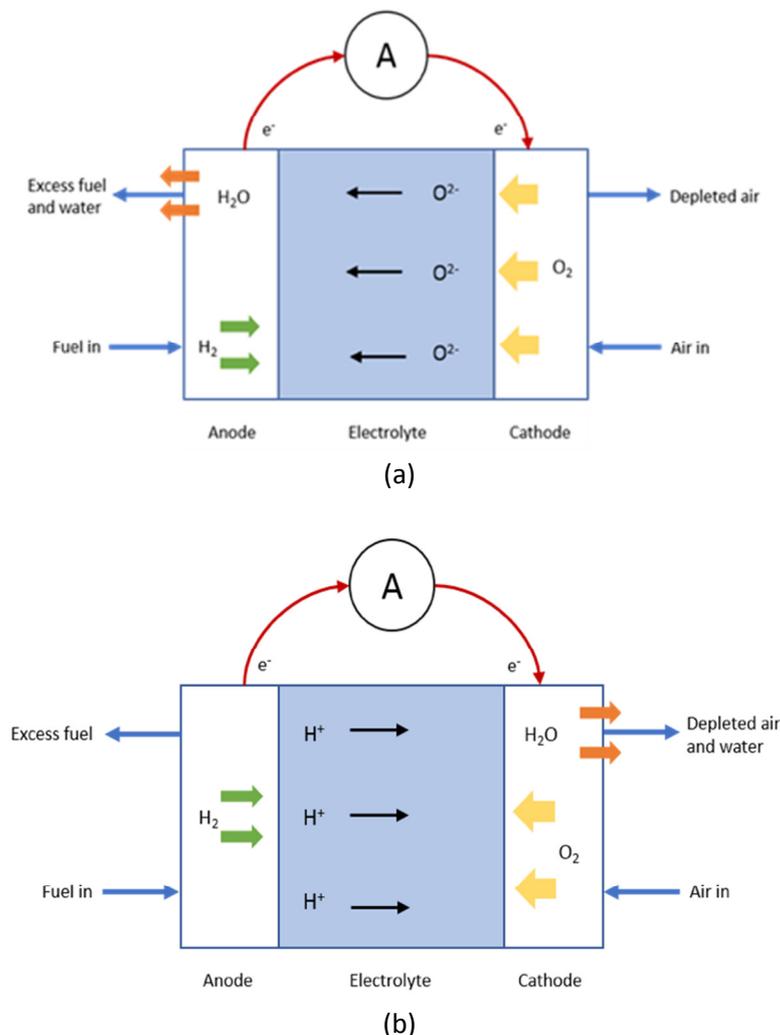
## 1. Introduction

A fuel cell is an electrochemical device that converts chemical energy from a reaction between a fuel and an oxidant into electrical energy, providing a clean, pollution-free technology that electrochemically generates electricity at high efficiencies [1]. Solid oxide fuel cells (SOFCs) can be divided into two main types based on their diffusion mechanism: oxygen ion-conducting solid oxide fuel cells (O-SOFCs) and proton-conducting solid oxide fuel cells (H-SOFCs). Both types of fuel cells strongly depend on the transport of oxide ions or protons across a ceramic with operating temperatures ranging from 800 to 1000°C for O-SOFCs and from 400 to 700°C for H-SOFCs [2–5].

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The principal components of an SOFC are two porous electrodes, an anode and cathode separated by a solid, and a fully dense electrolyte. Common anode and cathode materials in O-SOFCs are nickel cermet (Ni-YSZ) and strontium doped with lanthanum manganite (LSMO), respectively. Ytria-stabilized zirconia is the most popular electrolyte for the O-SOFC [6–9]. On the other hand, the most common anode and cathode materials in H-SOFCs are nickel oxide (NiO-BZCY) and strontium doped with lanthanum cobalt fluorite (LSCF), respectively. The best studied electrolyte for H-SOFCs is barium cerium yttrium zirconate (BZCY) [10,11]. The operating principles of conducting ion movement for the two types of SOFCs are shown in Figure 1.



**Fig. 1.** Schematic diagram showing the conducting ion movement for (a) a conventional SOFC and (b) a proton-conducting SOFC

Conventional O-SOFCs are considered to be one of the most promising fuel cells for stationary applications combining heat and power, as well as for automotive applications. Moreover, they can be operated using a large variety of fuels resistant to poisoning by CO and  $H_2S$  and do not require expensive noble metals as electrodes [4,12]. However, there are several challenges to commercializing this type of fuel cell, such as reducing the cost and increasing the reliability of the system, including the reliability of the cell-stack interactions, interconnections and seals [6,8]. Lowering the operating temperature of O-SOFCs to intermediate temperatures (400 - 800°C) where they exhibit high energy conversion efficiency and low environmental impact is currently accepted as the main strategy for SOFC commercialization. Unfortunately, this strategy results in significant

increases in the ohmic resistance of the electrolyte material and the polarization resistance of the cathode material [13,14]. Since O-SOFCs operate at high temperature, which leads to stability and reliability issues, many studies have attempted to achieve an intermediate operating temperature (400 - 700°C) by developing new electrolyte and electrode materials [15,16].

Proton conduction is based on the existence of proton defects in the oxide, which are created when oxide-containing oxygen vacancies dissociate and absorb water from the surrounding wet atmosphere [15]. This understanding led to the introduction of proton-conducting solid oxide fuel cells (H-SOFCs) by Iwahara *et al.*, [16,17] in 1981, which attracted enormous attention due their advantages over cells using oxygen ions. Since the resistance of the electrolyte layer is a major source of the overall overpotential and limits the cell efficiency, many efforts have been dedicated to its optimization. A relatively new idea is the application of highly conducting solid oxide proton-conducting membranes instead of well-established oxygen-ion-conducting membranes. This modification would present certain benefits. Hydrogen production and water gas shift reactions at the anode create protons and electrons via oxidation reactions. While protons migrate through the dense electrolyte to the interface, oxygen is simultaneously fed into the air channel and reacts with protons at the interface through a reduction reaction [17,18]. Another advantage of H-SOFCs is the production of water vapour at the cathode, as the product of the electrochemical reaction can prevent fuel dilution at the anode, thus resulting in higher OCV values and higher fuel utilization. H-SOFCs provide a wide range of advantages as a result of the cheaper interconnect materials, easier and more reliable sealing, shorter start-up and shut-down times, and a lower probability of material and performance degradation [19,20]. Additionally, the lower activation energy of proton conduction compared with that of oxygen-ion conduction implies a higher conductivity in the intermediate temperature range [21]. Complex oxides with the perovskite structure  $\text{SrCeO}_3$  exhibited proton conduction in a reducing atmosphere at high temperatures. In previous work, Zhang *et al.*, showed that the conductivity of the  $\text{SrCeO}_3$  electrolyte in hydrogen at 800°C is  $10^{-4}$  -  $10^{-3}$  S/cm. The high proton conductivity makes it a promising electrolyte material for intermediate temperature SOFCs [22].

## 2. Effect of Sintering Temperature on The Properties of Proton Conducting Electrolytes

Since Iwahara *et al.*, [23] applied  $\text{SrCeO}_3$  as a proton-conducting electrolyte for water electrolysis in 1981, additional proton conducting materials have been developed for SOFC applications, such as doped  $\text{BaZrO}_3$ ,  $\text{BaCeO}_3$ , and  $\text{SrZrO}_3$ . A candidate electrolyte material for H-SOFCs must meet the three basic requirements of good sinterability, high chemical stability and high ionic conductivity. Unfortunately, researchers have found it difficult to meet all three requirements simultaneously. For instance,  $\text{BaZrO}_3$ -based proton conductors exhibited perfect chemical stability in  $\text{CO}_2$  and  $\text{H}_2\text{O}$  atmospheres but showed poor sinterability, while  $\text{BaCeO}_3$ -based proton conductors showed high proton conductivity and good sintering behaviour but could easily react with  $\text{CO}_2$  and  $\text{H}_2\text{O}$  atmospheres, which can destroy the perovskite structure required to maintain high proton conductivity [12]. On the other hand, the conductivity of  $\text{SrCeO}_3$  is not high enough to develop a unit component due to its orthorhombic structure, which results in less proton transport compared with  $\text{BaCeO}_3$ -based compounds [11].

The sintering process used to prepare ceramics with a controlled microstructure can be adjusted to optimize the electrical conductivity of the ceramic electrolytes. Thus, the electrolyte microstructure and composition are related to the sintering conditions. Different sintering conditions can affect the microstructure of the electrolyte, including its grain size, grain boundary and relative density. The processing technique may significantly influence the ionic conductivity of

the electrolyte and help to optimize the sintering conditions of the electrolyte [24]. A large variation in the sintering temperature is necessary to produce highly dense electrolyte pellets suitable for impedance measurements used for conductivity measurement [25]. It is important to note that low sinterability may increase the grain boundary resistance and result in the degradation of the electrolyte conductivity [26]. The electrolyte sinterability is also influenced by the radius of the dopant used in the electrolyte. The concentration of oxygen vacancies in the oxide lattice and the creation of additional oxygen vacancies by the introduction of dopant in the electrolyte can facilitate oxygen-ion diffusion within the oxide lattice, subsequently improving the oxide densification process. In addition, a larger metal dopant radius can increase the free lattice volume in the electrolyte and enhance the mobility of cations, especially at the grain boundary [27].

Studies have shown that  $\text{BaCe}_{0.9}\text{Y}_{0.1}\text{O}_{2.95}$  exhibits high proton conductivity but poor stability under  $\text{CO}_2$ , while  $\text{BaZr}_{0.9}\text{Y}_{0.1}\text{O}_{2.95}$  has a high sintering temperature with poor grain boundary conductivity [28]. Electrical studies demonstrated that increasing the sintering temperature from 1150 to 1450°C leads to an increase in conductivity. This is because of the increased grain size and decreased grain boundary resistance that occurs as sintering temperature increases. However, several studies have indicated that the significant increase in the conductivity of ceramics sintered at 1550°C is due to the formation of a new phase [29]. Fully dense Y-doped  $\text{BaZrO}_3$  films were successfully obtained in which no pores could be observed from the cross-sectional view, even at a reduced temperature of 1350°C. In an exciting finding, the film appeared to be dense at the low temperature of 1300°C. In addition, the grain size in the films increased significantly with increasing sintering temperature. Previous studies encountered a variety of difficulties in obtaining dense films at such a low temperature via the traditional ceramic fabrication process without the help of sintering aids. Reducing the sintering temperature would definitely mitigate the evaporation of barium, thus improving the conductivity of Y-doped  $\text{BaZrO}_3$  films [30]. Tong *et al.*, [31] found that  $\text{BaZr}_{0.8}\text{Y}_{0.2}\text{O}_{3-\delta}$ -NiO pellet sintered at 1250°C has significant porosity with a grain size less than 1  $\mu\text{m}$ . Further increases in sintering temperature to 1450 and 1500°C resulted in the nearly complete elimination of obvious porosity, causing the grain size to increase to approximately 3  $\mu\text{m}$  and 5  $\mu\text{m}$ , respectively. Hsu *et al.*, [32] found that the higher temperature is necessary to densify  $\text{SrCe}_{1-x}\text{Zr}_x\text{O}_{3-\delta}$  ( $0.0 \leq x \leq 0.5$ ) samples. The  $\text{SrCeO}_3$  ceramics exhibited shrinkage of 15% when the samples were sintered at 1500°C for 8 h, while the  $\text{SrCe}_{0.6}\text{Zr}_{0.4}\text{O}_{3-\delta}$  pellets exhibited 9.55% shrinkage after sintering at 1500°C for 8 h. However, when the sintering temperature increased to 1600°C,  $\text{SrCe}_{1-x}\text{Zr}_x\text{O}_{3-\delta}$  ( $0.0 \leq x \leq 0.5$ ) oxides exhibited consistent shrinkage of approximately 15%. This result implied that the  $\text{SrCe}_{1-x}\text{Zr}_x\text{O}_{3-\delta}$  ( $0.0 \leq x \leq 0.5$ ) ceramics reached their final stage sintering temperature at 1600°C [32]. The electrolyte must show a high ionic conductivity and reactivity to enable a proton-conducting SOFC operating temperature of 400-700°C. Understanding the various factors that influence the ionic conductivity such as sinterability can help further to optimize the electrolyte properties [24]. Jaiswal *et al.*, found that  $\text{BaCe}_{0.7}\text{Zr}_{0.10}\text{Y}_{0.2}\text{O}_{3-\delta}$  exhibited sufficient proton conductivity and phase stability over a wide range of operating conditions [33]. However, the sinterability of  $\text{BaCe}_{0.7}\text{Zr}_{0.10}\text{Y}_{0.2}\text{O}_{3-\delta}$  with a higher cerium content remained low and required a high sintering temperature ( $\sim 1500^\circ\text{C}$ )

The addition of sintering aids can often decrease the sintering temperature of proton conducting electrolytes to as low as 1350°C, enabling application to SOFCs [31]. The application of sintering aids such as  $\text{Li}_2\text{O}_3$ , LiF,  $\text{Al}_2\text{O}_3$ , and transition metal oxides such as NiO, CuO and ZnO, has been successfully demonstrated to be effective in reducing the sintering temperature of ceramic materials, including electrolyte materials, without changing their electrical properties, as reported by Fan *et al.*, [34]. Other types of sintering mixtures such as  $\text{BaCO}_3$ ,  $\text{Y}_2\text{O}_3$  and  $\text{ZrO}_2$ , which work as sintering aids, can lower the sintering temperature of BZY [35–38]. Indium dopant has also been

shown to improve the sintering activity of barium zirconate synthesized through the citric acid-nitrate gel combustion process [39]. Liu *et al.*, determined that the porosity of  $\text{SrCe}_{0.95}\text{Y}_{0.05}\text{O}_{3-\delta}$  was lower than 2.2%, and this value decreased with increasing K dopant concentration, indicating that potassium was substituted at the A-site of  $\text{SrCe}_{0.95}\text{Y}_{0.05}\text{O}_{3-\delta}$  and decreased the sintering temperature due to the lower melting point of  $\text{K}_2\text{CO}_3$  [40]. Potassium not only substituted for strontium but also acted as a sintering aid in this composition [40].

### 3. Effect of Dopants on The Properties of Proton Conducting Electrolytes

$\text{SrCeO}_3$ -based electrolyte materials were first reported to predominately show proton conduction in atmospheres containing hydrogen or steam, while  $\text{BaCeO}_3$ -based materials showed mixed ionic conduction (oxide ions and protons) at elevated temperatures. Many investigations have been conducted to improve the electrical conductivity of  $\text{SrCeO}_3$ - and  $\text{BaCeO}_3$ -based materials. Methods such as substituting isovalent and aliovalent ions at either the A-site or B-site of the  $\text{ABO}_3$  structure can result in modified point defect concentrations, which impact the electrical and proton conductivity. For example, the stability of  $\text{SrCeO}_3$  can be improved by partially substituting Ce with Zr dopant, which can increase the tolerance factor and decrease its basicity. Numerous studies have reported effects on conductivity and chemical stability when doping trivalent cations at the B-site of  $\text{SrCeO}_3$ , which generates additional oxygen vacancies [40,41]. On the other hand, Hsu *et al.*, noted that the sintering of  $\text{BaZrCe}_{0.8}\text{Y}_{0.2}\text{O}_{3-\delta}$  proton-conducting oxides decreased with increasing Zr dopant content, and the chemical stability conversely increased with increasing the Zr dopant content [32]. The combination of Sr and Zr in  $\text{BaCeO}_3$ -based material resulted in  $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{Ce}_{0.65}\text{Zr}_{0.2}\text{Y}_{0.15}\text{O}_{3-\delta}$  electrolyte with an orthorhombic structure. This material exhibited a maximum conductivity of  $2.35 \times 10^{-1}$  S/cm at  $500^\circ\text{C}$  due to its smaller lattice volume, larger grain size and lower activation energy, as reported by Sailaja *et al.*, [42]. Table 1 summarizes the conductivity of electrolytes with various dopants.

**Table 1**  
 Sintering temperature and total conductivity of selected materials

Electrolyte material	Total conductivity ( $\text{S cm}^{-1}$ )	Sintering temperature ( $^\circ\text{C}$ )	Ref
$\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Ce}_{0.6}\text{Zr}_{0.2}\text{Gd}_{0.1}\text{Y}_{0.1}\text{O}_{3-\delta}$	$\sim 8.00 \times 10^{-3}$	1400	[19]
$\text{SrCe}_{0.95}\text{Gd}_{0.05}\text{O}_{2.975}$	$8.14 \times 10^{-6}$	1450	[21]
$\text{BaCe}_{0.95}\text{Gd}_{0.05}\text{O}_{2.975}$	$1.87 \times 10^{-4}$	1450	[21]
$\text{BaZr}_{0.8}\text{Y}_{0.2}\text{O}_{3-\delta}$	$1.60 \times 10^{-3}$	1350	[30]
$\text{BaCe}_{0.7}\text{Zr}_{0.1}\text{Y}_{0.2}\text{O}_{3-\delta}$	$1.02 \times 10^{-2}$	1400	[34]
$\text{BaZr}_{0.8}\text{In}_{0.2}\text{O}_{3-\delta}$	$4.57 \times 10^{-4}$	1600	[39]
$\text{Sr}_{0.95}\text{K}_{0.05}\text{Ce}_{0.95}\text{Y}_{0.05}\text{O}_{3-\delta}$	$8.00 \times 10^{-3}$	1550	[40]
$\text{Ba}_{0.8}\text{Sr}_{0.2}\text{Ce}_{0.65}\text{Zr}_{0.2}\text{Y}_{0.15}\text{O}_{3-\delta}$	$2.35 \times 10^{-1}$	1300	[42]
$\text{BaZr}_{0.3}\text{Ce}_{0.5}\text{Y}_{0.2}\text{O}_{3-\delta} - 2 \text{ mol\% Bi}_2\text{O}_3$	$5.26 \times 10^{-3}$	1400	[43]
$\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Ce}_{0.6}\text{Zr}_{0.2}\text{Gd}_{0.1}\text{Y}_{0.1}\text{O}_{3-\delta}$	$3.00 \times 10^{-3}$	1450	[44]
$\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Ce}_{0.5}\text{Zr}_{0.35}\text{Y}_{0.1}\text{Sm}_{0.05}\text{O}_{3-\delta}$	$2.39 \times 10^{-3}$	1450	[45]
$\text{Sr}(\text{Ce}_{0.6}\text{Zr}_{0.4})_{0.9}\text{Y}_{0.1}\text{O}_{3-\delta}$	$5.37 \times 10^{-3}$	1350	[46]
$\text{SrCe}_{0.9}\text{Yb}_{0.1}\text{O}_{3-\delta}-(\text{Li/K})_2\text{CO}_3$	$1.00 \times 10^{-1}$	1500	[47]
$\text{BaZr}_{0.8}\text{Y}_{0.2}\text{O}_{3-\delta}$	$7.90 \times 10^{-3}$	1450	[48]

### 4. Conclusion

Solid oxide fuel cells (SOFCs) are the most widely used fuel cells because of their high energy conversion efficiency and low negative impact on the environment. Current research trends show that improvement of SOFC performance relies on the electrolyte component, especially for

intermediate-temperature operation. Sintering temperature plays a role in determining the properties of the proton-conducting SOFC electrolyte, such as the electrical conductivity and microstructure. A large number of electrolyte materials have been examined for their ability to improve sinterability. Moreover, substituting new materials as dopants into the electrolyte can also affect the performance of the electrolyte. Thus, this review focused on the improvement of the electrolyte properties in terms of sintering behaviour and conductivity, which will further enhance the performance of proton-conducting solid oxide fuel cells, supporting their eventual commercialization.

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