

Proton Conduction in Perovskite Solid Electrolyte for Proton Ceramic Fuel Cell Application at Intermediate Temperatures: A Short Review

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ABSTRACT

Proton-conducting ceramics based on perovskite-type oxides have been significantly applied in a wide range of electrochemical devices such as fuel cells, hydrogen sensors, and steam electrolyzers. One of the emerging applications of these ceramic proton conductors is as an electrolyte component in a solid oxide fuel cell (SOFC), where the proton is mobilized from the anode to the cathode side via these conductors. The proton (hydrogen ion) diffusion mechanisms and activation energies (E_a) in these materials are heavily influenced by their composition, stoichiometry, and crystal structure. Hence, this review presents and discusses the mechanism of hydrogen ion movement for proton-conducting solid oxide fuel cells or known as proton ceramic fuel cells (PCFCs), based on experimental and modelling data, including the vehicular and the Grotthuss mechanisms. This review will provide a brief understanding of the connection between experimental and modelling evidence for proton mechanisms in perovskite electrolyte materials.

Keywords: Proton transport; perovskite oxide; proton-conducting ceramic; DFT

INTRODUCTION

Solid oxide fuel cells (SOFCs) have sparked much interest in renewable and hydrogen technology sectors due to their massive potential as a promising output device (Ramadhani et al. 2017). The SOFCs have established comprehensive consideration for their sustainable and clean energy resources because of their fuel flexibility, high efficiency in energy conversion and minimal release of toxic elements or gas, which has a low effect on the environment. In a fuel cell operation, the electrolyte parameters in the SOFC determine the path of the reactions, the open-circuit voltage, the cell's stability, mechanical performance, and operating temperature. Because the electrolyte is exposed to both the anode and cathode atmospheres, it should be chemically stable throughout a wide range of oxygen partial pressures.

The state-of-the-art oxide-ion SOFCs (O^{2-} -SOFCs) are composed of yttria-stabilised zirconia (YSZ) as the electrolyte material. In this thermally activated system, oxygen ions are transported from the cathode side to the anode across the electrolyte. A high operating temperature (800-1000°C) is needed to overcome the activation energy (E_a) for the electrolyte's ion migration and simultaneously improve the electrode reaction kinetics and reduce ohmic resistance. However, the high-temperature operation has

been demonstrated to cause interfacial diffusion between the electrolyte and electrode materials, a complication in materials sealing, and high polarisation resistance (Muhammed Ali et al. 2017; Zhu 2012). Nowadays, it is widely accepted that lowering the operating temperature of SOFCs to the intermediate range is the key to expediting widespread use.

Recently, proton-conducting SOFC (H^+ -SOFC) or so-called proton ceramic fuel cell (PCFC), has achieved great interest as an alternative for the O^{2-} -SOFC owing to its capability to operate at intermediate temperature (400-800 °C) while maintaining good ionic conductivity (Baral et al. 2014; Dailly et al. 2017). In H^+ -SOFC, the electrolyte materials are known as proton-conducting ceramic oxide. The growing interest in proton-conducting oxide has led to the discovery of novel materials, particularly barium-cerate oxide ($BaCeO_3$) and barium-zirconate oxide ($BaZrO_3$). Some trivalent or divalent cations are partially substituted at cerium sites in barium cerate to form $BaCe_{1-x}M_xO_{3-\delta}$ and at zirconium sites in barium zirconate to form $BaZr_{1-x}M_xO_{3-\delta}$, where M is a rare earth element, x is less than its upper limit of solid solution formation range (usually less than 0.2), and δ is the oxygen deficiency per unit formula. Doping

on BaCeO₃ and BaZrO₃ has proven to improve the ionic conductivity and stability in a fuel cell atmosphere (Baral et al. 2014; Rørvik et al. 2014).

As mentioned, the ionic conductivity can be increased through doping, as the process can increase the concentration of oxygen vacancies in the electrolyte. These vacancies are critical for proton-conducting perovskites because they are required to incorporate protonic defects via dissociative water absorption (Chen et al. 2022)(Meng et al. 2019). Hydrogen transport as H² or H⁺ would also require oxygen vacancies by incorporating hydrogen into the crystal lattice. When the doped oxide is exposed to water vapour, oxygen vacancies compensate for acceptors, as shown in the reaction shown in Equation 1, where V_O, O_O, and 2OH_O represent oxygen vacancies, lattice oxygen, and hydroxide at lattice oxygen, respectively.



PEROVSKITE OXIDE ELECTROLYTE

Electrolytes with perovskite-structured oxides have an excellent potential for energy-derived devices, specifically in SOFC applications since they exhibit a wide span of electrochemical and mechanical properties. Ideally, the perovskite stoichiometry consists of ABO₃, as seen in Figure 1. The large A cation and smaller size B cation are located in the corners and centre of the cube, respectively. In this case, the oxygen is placed in the middle of each cube's twelve edges, resulting in eight corner shares (octahedra) of BO₆. Due to the different sizes of the A, B, and O ionic radii, a cubic structure is usually attainable at high temperatures (Bartel et al. 2019).

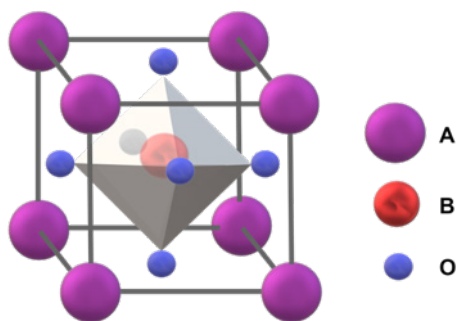


FIGURE 1. The ideal structure of perovskite oxide

In nature, proton-conducting perovskite structures do not contain protons. When the perovskite is in a dry environment, the conductance of the electron holes will prevail over the conductance of the perovskite oxide. The conductivity of this electron-hole will decrease as the proton conductivity increases when it is introduced with the proton (Rebollo et al. 2015). Because of their tiny ionic radius and substantial polarisation, proton ions cannot be separated under equilibrium conditions due to the strong covalent bonds interacting with the atmospheres (Singh et al.

2019). Under a humidified atmosphere, proton-conducting electrolytes exhibit elevated proton conductivity in the intermediate temperature range. Yang et al. investigated the proton conductivity of BaZr_{0.1}Ce_{0.7}Y_{0.1}Yb_{0.1}O_{3-δ} (BZCYYb) electrolyte material and obtained 10⁻² Scm⁻¹ at 550 °C (Yang et al. 2014). Another current perovskite material used as an electrolyte, BaSn_{0.3}Sc_{0.3}O_{3-δ} by Zvonareva et al. has obtained a reasonable proton conductivity of 10⁻³ Scm⁻¹ at 600 °C (Zvonareva et al. 2022). Similar studies on proton conductors with desirable conductivity can be found in (J. F. Basbus et al. 2016; Fabbri et al. 2008; Nasani et al. 2013). Moreover, proton-conducting electrolytes typically have a substantially lower E_a (0.4-0.6 eV) than oxygen ion conductors. As a result, proton-conducting materials are preferable to oxygen ion-conducting oxides as the electrolytes of intermediate to low temperature operation.

MECHANISM OF PROTON-CONDUCTING

Several studies have been conducted to analyse proton transport to understand the proton conduction mechanism. It is widely accepted that two different proton transport mechanisms exist: (i) vehicle mechanism and (ii) Grotthuss mechanism (Kreuer 1996).

The vehicle mechanism is typically used in high humidity and low-temperature environments. Protonic conduction can be accomplished by a proton attaching itself to an oxygen ion to form a hydroxide ion (OH⁻), which moves instead of the individual proton, as shown in Figure 2. The proton transport is ensured by the migration of the mobile group in the structure. As water molecules form a cluster (proton-conducting channel) combined with the electrolyte membrane, they operate as a vehicle for transferring protons. Hydroxide ions may have a lower activation energy of diffusion than oxygen ions due to their smaller size and can diffuse through the structure using vacancy diffusion or interstitial diffusion (Meng et al. 2019). The electroneutrality balance is maintained by the counter diffusion of unprotonated vehicles or, most probably, by oxygen vacancies: aqueous fluids, ionic liquids, and tiny molecular molecules with a low-binding energy feature this mechanism.

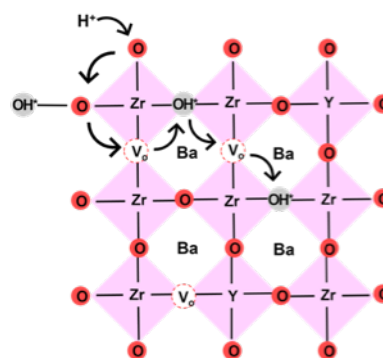


FIGURE 2. Representation of proton mobility inside a perovskite structured BZY proton conductor via the vehicle mechanism (adapted from (Regalado Vera et al. 2021)).

The Grotthuss mechanism is fairly discussed in the literature concerning proton mobility inside a perovskite-structured proton conductor. These studies have better-understood proton transfer mechanisms and practical screening strategies for identifying perovskites with improved material properties for H⁺-SOFC development. An improved understanding of proton transport materials and mechanisms will benefit energy conversion and storage technologies based on perovskite proton conductor materials (Duan et al. 2020).

In the Grotthuss mechanism, the proton is the only migratory species while the oxygen is confined around its crystalline orientation. Through this mechanism, it can be concluded that protons move through (i) proton incorporation, (ii) rotational diffusion, (iii) transfer to neighbouring oxygen, (iv) bending and (v) elongation of the B–O bond. During operation, a proton jumps from a stationary oxygen atom to a neighbouring oxygen atom after it rotates to the correct position between the two oxygen atoms, as depicted in Figure 3. Its hopping activation energy depends on the oxygen-oxygen (O–O) distance, and lower activation energy signifies that smaller energy is required for the proton to jump to the neighbouring oxygen site (De Souza & Muccillo 2010). Basbus et al. determined the protonic jump length to be 3.1 Å between oxygen sites for the BaCe_{0.4}Zr_{0.4}Y_{0.2}O_{3-δ} (Juan F. Basbus et al. 2020). It is postulated that a perovskite oxide with a cubic structure achieves higher protonic conductivity since the distance between the neighbouring oxygen sites is short. Furthermore, it is thought that the rate-limiting phase is proton transfer between two neighbouring lattice oxygens, which is dependent on the formation and breaking of hydrogen bonds. Thus, Grotthuss mechanism is predominant in perovskite oxide under a hydrogen-containing atmosphere (Ingenmey et al. 2018), as evidenced in these manuscripts (Ji et al. 2021; Zhou et al. 2019).

MECHANISM OF PROTON-CONDUCTING STUDIED USING DENSITY FUNCTIONAL THEORY (DFT)

Density Functional Theory (DFT) simulations can predict the behaviour and electrochemical properties of proton conductors at the atomic and molecular level, consequently simulating the proton transfer pathway in proton conductor electrolyte materials that is difficult to observe experimentally. In the literature, the application of DFT in the theoretical study of the proton transfer mechanism for H⁺-SOFC has been demonstrated and validated with experimental data and compared to other reported works. DFT is based on first principles equations and quantum theory with various software availability (Jing & Aluru 2020; Priya & Aluru 2021; Tao et al. 2021).

DFT software is an important tool in designing a proton conductor with high protonic conductivity, a highly required property of electrolyte material for PCFC. For example, Vienna Ab-initio Simulation Package (VASP) software has been used to analyse the proton diffusion mechanism of Y-doped BaZrO₃ (BZY) (Du et al. 2020; Priya & Aluru

2021). The computer programme Crystal09 has been utilised to explore the impact of dopant nature on structures and lattice dynamics of proton-conducting BaZrO₃ (Zeudmi Sahraoui & Mineva 2013).

One of the main electrochemical properties of SOFC that can be predicted using DFT is the energy barriers to proton migration. Theoretical work on the mechanism of proton conduction across the bulk and along the surfaces of BaZrO₃ was studied by Kim et al. (J. S. Kim & Kim 2017). It was reported that surface resistances could be higher than grain boundary resistance studied in bulk samples, so studying proton conduction on the surfaces is beneficial for H⁺-SOFC development. They reported that the proton in BaO-terminated has difficulty travelling along and across the surface; in contrast, the proton in ZrO₂-terminated can move along the surface quickly. The result was consistent with other reported works (D. H. Kim et al. 2012).

The effect of two protons interacting on proton migration in bulk BaZrO₃ with four possible proton structural configurations is explored using density functional by Kim et al. (D. H. Kim et al. 2012). One of their objectives is to fill the gap of previous DFT studies on bulk BaZrO₃ that only used a single proton for energy barrier calculations. DFT can simulate various proton transfer pathways for differently designed molecule structures and calculate the energy barrier. Kim et al. (D. H. Kim et al. 2012) found that the most energetically stable structure was the asymmetrically distorted structure with the shortest distance between the two protons, which is difficult and costly to visualise and understand using experiments. When a proton migrates near another, the energy barrier occurs in the simple pathway, as reported by Kim et al. (D. H. Kim et al. 2012). Their simulations are compared with experimental energy barriers measured using an impedance spectroscopy analyser, and by using proton pairs to simulate the energy barrier of proton migration in their study, the difference between theoretical and experimental data has been significantly reduced compared to when using only one proton.

In a study by Jing et al. (Jing & Aluru 2020), the effect of A ion vacancy on the mechanism of proton transfer and hydroxide ion rotation in perovskite oxide is difficult to observe using experiment was explored theoretically using DFT. The results of this DFT study revealed that the A-site ion in a perovskite oxide could lower the energy barrier for local lattice deformations, which aids in hydroxide ion rotation and proton transfer, as illustrated in Jing et al. (Jing & Aluru 2020). This simulated energy barrier was found in agreement with the previously published value (Jing & Aluru 2020).

However, the reliability of DFT still depends on the usage of validated and verified experimental data as the input value before DFT simulations. The energy barrier values measured from experiments, for example, using A.C. impedance study, nuclear magnetic resonance (NMR) experiments and refined crystallography data, are compulsory for the validity of DFT simulations of the proton transfer mechanism of H⁺-SOFC.

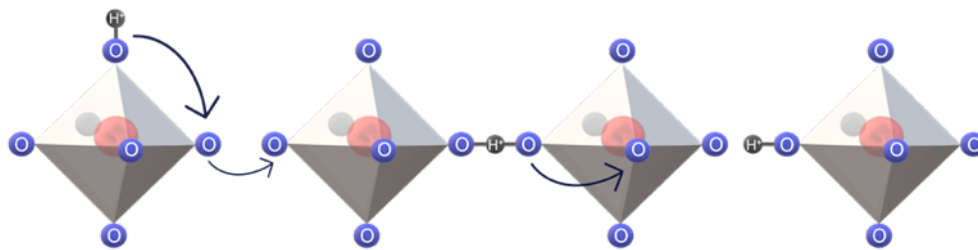


FIGURE 3. Representation of proton mobility via the Grotthuss mechanism inside a perovskite-structured proton conductor (adapted from (De Souza & Muccillo, 2010)).

CONCLUSION

In solid oxide fuel cells (SOFCs), the proton-conducting ceramics based on perovskite-type oxides are used as an electrolyte component. The proton (hydrogen ion) diffusion mechanisms and activation energies (E_a) in these materials are profoundly influenced by their composition, stoichiometry, and crystal structure. These mechanisms are hypothesised through experimental works and proved through DFT investigations for the proton-conducting perovskite oxides. It can be concluded that most of the DFT studies for proton conductors reviewed here focus on proton conductor doped and undoped BaZrO_3 . There is a massive opportunity for DFT to be further implemented as a cost-and-time-saving preliminary design tool for optimising other advanced proton conductor electrolytes such as Y-doped $\text{Ba}(\text{Ce},\text{Zr})\text{O}_3$ (BCZY) and Y- and Yb-doped $\text{Ba}(\text{Ce},\text{Zr})\text{O}_3$ (BCZYb) for further development of H^+ -SOFC.

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DECLARATION OF COMPETING INTEREST

None

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