



A review on proton conducting electrolytes for clean energy and intermediate temperature-solid oxide fuel cells

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ABSTRACT

The aims of this review article is to understand the mechanism of proton-conducting solid oxide fuel cells (SOFCs) and compare it with conventional SOFC, to understand how dopants help to improve the conductivity and stability of doped materials, to investigate and analyze the effect or impact (in terms of conductivity and stability) of different types of dopants on the materials as proton-conducting electrolyte in intermediate temperature solid oxide fuel cell (IT-SOFC) and to study the experimental reviews on the methods in synthesizing the materials. Emphasis is given on the relationship between structural and mechanistic features of the crystalline materials and their ion conduction properties. This review will be focusing more on BaCeO₃ and BaZrO₃ as these electrolyte materials were in the focus of research during the past decades due to their considerable proton conductivity and stability.

1. Introduction

1.1. Motivation

Metal oxides constitute a most wonderful field of materials with an extensive variety of properties. Fossil fuels in various forms have acted as a reliable source of energy since the industrial revolution in the 18th century. Though, growing consciousness of environmental issues and inadequate source of energy resources has forced the society to look for alternative clean source. From the practical point of view, SOFCs are preferred in comparison to aqueous electrolyte fuel cell such as phosphoric acid or alkaline fuel cell [1]. In most cases SOFC runs at high temperatures e.g., $T > 600$ °C. This requires not only extra energy for heating but also stability and performance of materials at these operating temperatures need to be considered [2]. The polymer electrolyte fuel cell (PEFC) is low temperature ($T < 100$ °C) fuel cells, where Nafion (perfluoro sulfonic polymer) is used in electrolyte, require expensive platinum catalysts. Therefore, fuel cell operating at intermediate temperature range (200–600 °C) is desirable alternatives. In many cases, proton conductors show higher conductivity than oxide ion conductors especially at intermediate temperature range. However, literature data compiled by Norby [3] displays the existence of a gap within the intermediate temperature range, in which no materials

demonstrate high proton conductivity. Tapering this gap would be very advantageous from a technical point of view.

This provides a strong motivation to investigate the proton conductivity in the intermediate temperature range especially in perovskite oxides. By understanding the key factors that strongly influence the proton conductivity it should be possible to improve the conducting properties of the materials and also predict new materials which exhibit high proton conductivity.

Applications of these materials are aiming at further development of techniques leading towards the possibility of real “design” and synthesis of mainly complex oxides and to carefully study the structure and dynamics of the new materials.

1.2. Perovskites

In the 1830's, the geologist Gustav Rose had first described the perovskite and termed it after the prominent Russian mineralogist Count Lev Alekseevich von Perovskii. Perovskites are the most abundant mineral on the earth. Ideal perovskites have the general formula ABO_3 , where the *A*-site are typically larger cations than the *B*-site and analogous in size to the *O*-site anions. ABO_3 structure can be supposed to as a face centered cubic (fcc) lattice with the *A* atoms at the corners and the *O* atoms on the faces. The *B* atom finalizes the

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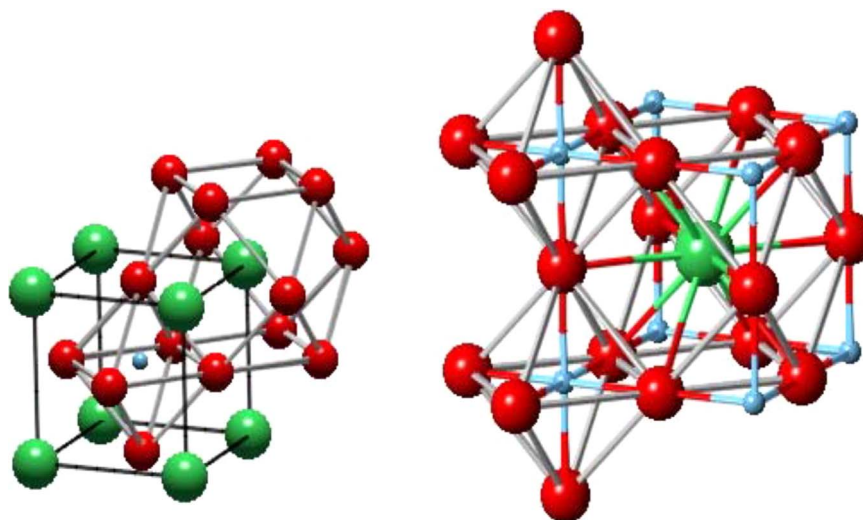


Fig. 1. A representative of the ideal ABO_3 perovskite structure (space group: $Pm-3m$) display the octahedral and cubo-octahedral (12-fold) coordination of the A and B -site cations, respectively; (a) A -cell setting (b) B -cell setting [4].

structure which is located at the center of the lattice. In this structure, the largest atom is A and therefore grows the whole size of the AO_3 (fcc) structure. The structure of perovskite comprises of a three dimensionally connected system of BO_6 octahedra, with A ions making AO_{12} cube-octahedra to fill the spaces between the octahedra. Fig. 1.1 shows the six coordinated B -site and twelve coordinated A -site in a perovskite structure. In perovskites there are three structural degrees of freedom: (a) movement of cations A and B from the centers to AO_{12} and BO_6 of their cations coordination polyhedra; (b) distortions of the anionic polyhedra coordination around A and B ions; (c) the BO_6 octahedra tilting about one, two, or three axes.

Most of the perovskites are considered to be ionic compounds, and the ions comprising them can be regarded to a first approximation as spheres with ionic radii r (Å). In the real structures, because of differences in the sizes of the O , A and B ions, the Goldschmidt tolerance factor (t) might be a guide as to whether a given composition will adopt the perovskite structure at a particular temperature and pressure. For ideal perovskites, the tolerance factor is derived as, $t = (r_A + r_O) / [\sqrt{2}(r_B + r_O)]$. For ideal structures t is unity, although diverse perovskites structures of lower symmetry can exist for values of t greater than 0.8.

Importantly, the value of t is not an unambiguous guide to structure type, and is certainly not an indicator of the space group that a particular perovskite will adopt at 300 K and atmospheric pressure. This is due to the factors other than ion size, e.g., interactions between metal and metal, degree of covalence, John-Teller and lone pair effects play a role in defining the space group or structure adopted [4].

1.3. Proton conductors

The proton conduction plays a vital role in heterogeneous technological applications. For example, Indium doped CaZrO_3 is used commercially as proton conducting electrolyte in hydrogen sensors in liquid metal [5,6]. Other areas of applications are in hydrogen pumps, hydrogen gas controllers, steam electrolyzers, fuel cells and hydrogenation/dehydrogenation of organic compounds [7].

A proton is a hydrogen ion (H^+) having no electron in its shell which distinguishes it from all other chemical species. It is an elementary particle which is quite stable (mean life time 2.1×10^{29} years) being positively charged of equal magnitude of an electron (1.6022×10^{-19} C). The mass of it is 1.6726×10^{-27} kg, 1837 times higher than that of an electron. The field of proton conductors or more precisely the effect of hydrogen on physical and chemical properties has emerged more than 60 years ago. In 1954 J. Volger, reported on some

ferromagnetic oxidic compounds of manganese with perovskite structure [8], whilst in 1956 Thomas and Lander reported the formation of hydroxyl groups also in zinc oxide. In 1958, J. B. Goodenough showed an interpretation of the magnetic properties of the perovskite type mixed crystals $\text{La}_{1-x}\text{Sr}_x\text{CoO}_{3-\delta}$ [9] and Rudolph measured the electrical properties in strontium-barium oxide and concluded that the protons acted as positive charge carriers. In 1965 G. Blasse explained new compounds with perovskite like structures [10]. Mobile protonic species in perovskite oxides (BaTiO_3) was first reported by Pope and Simkovich [11] in 1974. Later many researchers have investigated the proton conducting properties in different perovskite oxides [12–14]. There has been a strong interest in characterizing proton conducting perovskite structured materials by a range of theoretical and experimental techniques.

The promising devices that have high efficiency cogeneration are solid oxide fuel cells (SOFC) with temperature characteristically vary from 700 to 1000 °C. The goal of dropping the functioning temperature of solid oxide fuel cell has been an anxiety with the development and optimization of microstructures and cell materials. The benefits of falling the temperature are not only the kind robust but at the same time the cost of the total system reduces. The electrolyte that based on proton conducting oxide permits a significant decline in operating temperature in the range of 400–700 °C has been studied that proton conducting solid oxide fuel cells (PC-SOFC), creating high conductivity and stability. Yttrium doped barium cerate and barium zirconate proton conducting electrolytes have established that they can produce protons efficiently and the charge carriers that are the ions which dominate the conductivity [15,16]. However owing to particular conditions concerning the conductivity and stability of these materials, scientists are still making advance experiments on the doped materials competes with each other and the effect of different dopants the best proton conductors in solid oxide fuel cells.

1.4. Fuel cells

An energy conversion device that transforms the chemical energy of a fuel directly to electrical energy and heat, from the chemical reaction between fuel (H^+) and oxidizing agent (O_2), without the need for direct combustion as an intermediate step, is known as the fuel cell. It provides much advanced alteration efficiencies than conventional thermo-mechanical approaches. Analogous to the batteries, the working principles of fuel cells are electrochemical combination of reactants to produce electricity. In fuel cell an oxidant gas (e.g., oxygen in the air) and a mixture of a gaseous fuel (e.g., hydrocarbon or hydrogen fuels)

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