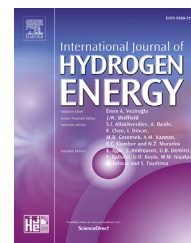


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Highly dense and chemically stable proton conducting electrolyte sintered at 1200 °C

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ABSTRACT

The BaCe_{0.7}Zr_{0.1}Y_{0.2-x}Zn_xO_{3-δ} (x = 0.05, 0.10, 0.15, 0.20) has been synthesized by the conventional solid state reaction method for application in protonic solid oxide fuel cell. The phase purity and lattice parameters of the materials have been studied by the room temperature X-ray diffraction (XRD). Scanning electron microscopy (SEM) has been done for check the morphology and grain growth of the samples. The chemical and mechanical stabilities have been done using thermogravimetric analysis (TGA) in pure CO₂ environment and thermomechanical analysis (TMA) in Argon atmosphere. The XRD of the materials show the orthorhombic crystal symmetry with Pbnm space group. The SEM images of the pellets show that the samples sintered at 1200 °C are highly dense. The XRD after TGA in CO₂ and thermal expansion measurements confirm the stability. The particles of the samples are in micrometer ranges and increasing Zn content decreases the size. The conductivity measurements have been done in 5% H₂ with Ar in dry and wet atmospheres. All the materials show high proton conductivity in the intermediate temperature range (400–700 °C). The maximum proton conductivity was found to be 1.0 × 10⁻² S cm⁻¹ at 700 °C in wet atmosphere for x = 0.10. From our study, 10 wt % of Zn seems to be optimum at the B-site of the perovskite structure. All the properties studied here suggest it can be a promising candidate of electrolyte for IT-SOFCs.

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Introduction

Fuel cells have very good impact to resolve the present energy demands in the world. Solid oxide fuel cells (SOFCs) operate at intermediate temperature have attracted much interest worldwide because of their affordability, prospective long term stability and wide range of applications (including accommodation and transports) [1–3]. In the modern ages, the proton conducting electrolytes get a great enhancement in SOFCs technologies operating in intermediate temperatures. For an extensive range of technological applications such as batteries, gas and humidity sensors, separators, supercapacitors, fuel cells, hydrogenation or dehydrogenation of hydrocarbon electrolyzers, there are increasing interests in proton-conducting electrolytes for SOFCs [1–7].

At a temperature below 600 °C, doped ceria based materials potentially meet the utmost necessities for fuel-cell operation, appears to be the best available electrolyte materials [4,8,9]. Though, comprising the protonic conductivity and indeterminate perfunctory of reliability below the fuel-cell working conditions some significant issues are still remain challenges. The electrolyte operates at low temperature show very potential candidate because of their low activation energy (AE) of protons conduction. Many perovskite-type oxides show elevated proton conductivity in reducing atmosphere [10–13] but the compromise between conductivity and chemical stability is the most substantial challenges for those type of materials [14–16].

The barium cerates (BaCeO_3) based materials exhibit mixed ionic (protons and oxide ions) conduction [17,18], as electrolytes. These materials show good sintering behavior and elevated ionic (proton) conductivity [11,19], however they are unstable under steam and CO_2 conditions lead to destroying the perovskite structure [20,21]. On the other hand the zirconates (BaZrO_3) do not sinter easily, and the maximum conductivities are only found when sintering occurs at or approaching 1700 °C [22]. Even after sintering at very high temperatures, the very poor grain-boundary conductivity of these materials is a main problem; these materials do not provide appropriate performance for practical application. Although, the crystal component is difficult to resolve in polycrystalline samples, it does seem that this is only improved when heated at very high temperatures, above of 1600 °C. This appears to indicate that reaching of the utmost conductivities is influenced by some form of phase transformation or segregation or, as suggested by Snijkers et al. [23] by a slow kinetic process of water absorption.

The formation of solid solutions between BaCeO_3 and BaZrO_3 are comparably easily occurring. It is also possible to form a solid solution by replacing a desired fraction of Ce in BaCeO_3 with Zr and other element that exhibit both sufficient proton conductivity as well as adequate chemical and thermal stability over a wider variety of conditions appropriate to fuel cell operation [10,16,21,24–28]. It was found that doped zirconates gives improved chemical stability but reduced protonic conductivity compared to the doped cerates. To accomplish elevated ionic conductivity as well as sufficient chemical and thermal stability, the stability of doped barium cerates can be improved by doping of Zr at the B-site. But still it is difficult to

develop the electrolyte material with low sintering temperature and high density which is very essential in SOFCs operation. The effect of Ba non-stoichiometry and Cr deposition, poisoning were studied on the Zr, Y co-doped BaCeO_3 based proton conductors and showed that the Ba non-stoichiometry influenced the electrical conductivity, especially with respect to grain boundary resistance and the Cr poisoning effect of the electrochemical performance of BZCY electrolyte [29,30].

Despite the fact that the sintering temperature was still very high [31], recently the partial substitution of Zr^{+4} cation into Ce^{+4} cation was reported as an electrolyte material and it was found the improved chemical stability [10,32]. It was a great challenge to process electrolyte material at low temperature to get desired result such as high conductivity, high density and chemical stability. At high temperature sintering with long time heat treatment can be predicted the barium oxide evaporation and thereby decrease the conductivity, as has been detected in BaCeO_3 [32]. But in the solid state synthesis using the barium excess of ≥ 10 mol% led to a higher conductivity [33].

It is found among the perovskite oxides that doped barium cerates exhibit mixed oxide and proton ion conductivity [34] upon exposure to stream atmospheres. However, it is difficult to get high density of doped barium cerate or zirconate materials at a sintering temperature below 1400 °C. Getting theoretical density above 90% of the material we need to sinter at high temperature (above 1400 °C) where Ba could evaporate to a certain degree [22,35]. To decrease the sintering temperature of ceramics materials, applying a sintering aid is one of the most effective ways [36,37]. In the previous years, to improve the sinterability of ceramic proton conductors, researchers have tried with different additives. The effect of MO_x metal oxides (where $M = \text{Ti, Fe, Co, Ni, Cu, Zn}$) on the densification behavior found that the addition of small amounts of these metal oxides could lower the sintering temperature by 150–250 °C [37]. Further studies showed that the Cu and Zn containing materials have significantly higher conductivity than the others in both moistened air and humidified hydrogen atmospheres. However, adding of CuO led to the formation of a $\text{Ba}_2\text{YCu}_3\text{O}_x$ impurity phase [38]. On the other hand, under a reduced atmosphere the decrease of CuO to metallic copper is a large anxiety in practical fuel cell applications. Therefore, as a sintering aid ZnO in doped BaCeO_3 - BaZrO_3 , is the most encouraging metallic oxide. Formerly, the sintering behavior of proton conductors with ZnO as a sintering aid was investigated by many researchers [37–42]. Most of those researchers usually examined the sintering behavior of thick pellets [37–42] but applying such electrolytes in real fuel cell fabrication was rare.

In order to solve the density and high sintering temperature issues, Zn was introduced into Y and Zr co-doped BaCeO_3 [38–40]. In 2005, Balibo and Haile first reported that ZnO is an excellent sintering aid for yttrium-doped BaZrO_3 , lowering the densification temperature from 1700 °C to 1300 °C [38]. In 2006, Tao and Irvine found that the introduction of small amount of Zn^{2+} substituted for Y^{3+} at B site into the perovskite structure allows a reduction in high sintering temperatures and a notable progress in the stability, relative density and conductivity [40]. Slodczyk and Sharp showed that Zn doped Barium Cerium Zirconium Yttrium oxide (BCZYZn) has been

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