

Enhanced ceria based electrolytes by codoping samaria and scandia for intermediate temperature solid oxide fuel cells



Yun Gan ^{a, b}, Jigui Cheng ^{a, b, *}, Mingming Li ^{a, b}, Hailin Zhan ^{a, b}, Wenzhou Sun ^{a, b}

^a School of Materials Science and Engineering, Hefei University of Technology, Hefei 230009, China

^b Key Laboratory of Advanced Functional Materials and Devices, Hefei University of Technology, No. 193 Tunxi Road, Hefei, Anhui 230009, China

HIGHLIGHTS

- Codoping of Sc_2O_3 and Sm_2O_3 improves the electrical conductivity of ceria electrolyte.
- The doping amount of Sc_2O_3 also affects the electrical conductivity.
- Oxygen vacancy ordering causes the electrical conductivity drop in heavily doped ceria.

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ABSTRACT

In this paper, ceria codoped by samaria (Sm_2O_3) and scandia (Sc_2O_3) ($\text{Sc}_x\text{Sm}_{0.2-x}\text{Ce}_{0.8}\text{O}_{1.9}$, $x = 0, 0.01, 0.02, 0.04, 0.06, 0.08$, SSDCX) was prepared by the glycine-nitrate method and investigated as candidates for electrolyte materials of intermediate temperature solid oxide fuel cells (IT-SOFCs). The influence of scandia addition on the phase composition, crystalline structure, microstructure, electrical conductivity and chemical stability of the ceria based electrolytes was investigated. Cell performances with $\text{Sc}_{0.04}\text{Sm}_{0.16}\text{Ce}_{0.8}\text{O}_{1.9}$ as electrolytes were also tested and compared with those of cells with $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}$ as electrolytes. The results indicate that ceria codoped with proper amount of Sc_2O_3 and Sm_2O_3 have higher electrical conductivity than ceria only doped with Sm_2O_3 . The doping amount of Sc_2O_3 also has influence on the electrical conductivity of the SSDCX materials, and the highest electrical conductivity of $2.64 \times 10^{-2} \text{ S cm}^{-1}$ is achieved for $\text{Sc}_{0.04}\text{Sm}_{0.16}\text{Ce}_{0.8}\text{O}_{1.9}$ materials at 700°C . Single cells with $\text{Sc}_{0.04}\text{Sm}_{0.16}\text{Ce}_{0.8}\text{O}_{1.9}$ as electrolytes demonstrate a maximum power density of 225 mW cm^{-2} , and an open circuit voltage (OCV) of 0.87 V at 700°C . Codoping Sc_2O_3 and Sm_2O_3 also suppresses the reduction of ceria thus inhibits the drop of OCV under working condition at 700°C .

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1. Introduction

With an increasing demand of green energy, solid oxide fuel cells (SOFCs) have attracted more and more attention in recent years because of their ability to provide clean energy with high efficiency [1–3]. Traditional SOFCs with yttria stabilized zirconia (YSZ) as electrolytes have to operate at relatively high temperature (around $800\text{--}1000^\circ\text{C}$) to attain high outputs due to the relatively low electrical conductivity of YSZ electrolytes at low temperature. However, the high operating temperature brings about problems such as thermal degradation, thermal expansion mismatch, and

reactions between the cell components [4]. Therefore, it is necessary to reduce the operation temperature to intermediate temperature range ($600\text{--}800^\circ\text{C}$) in order to solve the above problems and enlarge the applicability of SOFCs.

Doped ceria materials, which exhibit lower electrical conduction activation energy (E_a) and higher ionic conductivity at intermediate temperature, have been widely investigated for use as electrolytes in intermediate temperature range. Much work has been done in search for suitable dopants for ceria that can improve the comprehensive properties. Among the various doped ceria, Samaria doped ceria (SDC) and gadolinium doped ceria (GDC) have been reported to have higher ionic conductivity [5–10].

Andersson et al. have concluded that the suitable dopants for ceria should have an effective atomic number between Pm (61) and Sm (62) [11]. In addition, Kim studied the influence of ionic radius and valences of dopant cations on the lattice parameters of fluorite

* Corresponding author. School of Materials Science and Engineering, Hefei University of Technology, Hefei 230009, China.

E-mail address: jgcheng63@sina.com (J. Cheng).

oxides and put forward ' r_c ' to define a critical ionic radius of the dopants which causes neither expansion nor contraction of the host lattice. The value of r_c for trivalent dopant cations of ceria is calculated to be 0.1038 nm [12]. Therefore, co-doping strategy i.e. doping of different elements with proper concentration was developed to achieve an effective ionic radius similar to r_c . Numerous investigations have been conducted and demonstrate the validity of this strategy [13–16], for example, Omar et al. enhanced ceria based electrolyte through the codoping of Lu and Nd which shows higher ionic conductivity than ceria only doped by Lu or only doped by Nd [17,18].

Scandia is known as the best dopant for zirconia based electrolyte (SSZ) which demonstrates the highest ionic conductivity among various doped zirconia [19,20]. Moreover, it is reported that SOFC with zirconia stabilized by both Scandia and Ceria as electrolyte display a performance of 911 mW cm⁻² at the current density of 1.25 A cm⁻² and 800 °C [21]. However, Scandia as dopant for ceria is reported to attain lower electrical conductivity than those obtained in other doped-ceria materials with an undefined explanation [22], and by far there are few research on ceria based electrolyte codoped by Scandia and other element. So, in this paper, ceria codoped by Sm₂O₃ and Sc₂O₃ with different doping amount of Sc₂O₃ (Sc_xSm_{0.2-x}Ce_{0.8}O_{1.9}, $x = 0, 0.01, 0.02, 0.04, 0.06, 0.08$; SSDCX) was prepared and the effects of Sc₂O₃ addition on the structures and performances of the ceria based materials were studied.

2. Experimental

2.1. Sample preparation

Sc_xSm_{0.2-x}Ce_{0.8}O_{1.9} ($x = 0, 0.01, 0.02, 0.04, 0.06, 0.08$) powders were prepared by the glycine-nitrate method, in which stoichiometric amounts of Ce(NO₃)₃·6H₂O, Sm₂O₃ and Sc₂O₃ were dissolved into nitric acid [22]. By adding proper amount of glycine into the solution and stirring, a clear, transparent solution forms. The solution was then heated and a self-combustion finally takes place. The resultant precursors were further calcined at 900 °C for 3 h to remove residual organics and form a stable crystalline structure. Afterward, the obtained powders were pressed into bars (40 mm × 5 mm × 3 mm) and sintered at 1450 °C for 10 h to prepare the samples for electrical conductivity tests and the relative densities of samples were also measured.

2.2. Characterization and measurement of the SSDCX materials

X-ray diffraction (XRD, $2\theta = 3^\circ \text{ min}^{-1}$, D/MAX2500V, Rigaku Corporation, Japan) was used to examine the phase composition of the as-synthesized SSDCX powders. High-resolution transmission electron microscopy (HR-TEM, JEOL 2100F) was employed to investigate the lattice spacing and crystalline structure of the samples.

Powders with a composition of SSDC4 were pressed into green disks with a diameter of 18 mm and a thickness of 2 mm. Then the as-prepared disks were sintered at 1400, 1450 and 1500 °C for 10 h, respectively. The microstructure of the samples was observed by scanning electron microscopy (SEM, SU8020, HITACHI, Japan). The electrical conductivity of the SSDCX samples with different doping amount of Sc₂O₃ were measured in air from 400 °C to 700 °C using DC four-probe methods. X-ray photoelectron spectroscopy (XPS, Thermo ESCALAB 250) was also used to determine the valence change of Ce in SSDCX samples which were reduced in H₂ beforehand.

2.3. Preparation and test of single cells

A series of SSDCX electrolyte supports was also prepared by dry pressing the powders into pellets and sintered at 1450 °C for 10 h. The two surfaces of electrolyte were mechanically polished and ultrasonically cleaned in distilled water. NiO-SSDCX anode slurry was prepared by ball milling NiO (65 wt. %) and SSDCX powders (35 wt. %) in terpineol ethyl cellulose solution. And the cathode slurry (Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3- δ} -SSDCX) was prepared in the same way. Anode slurry was coated onto one side of the electrolyte and the opposite side was covered with cathode slurry. Then the cells were heat treated at 1000 °C for 3 h. Subsequently, Ag paste (SS-8060, Xinluyi, Shanghai, China) was painted on the surface of electrodes and connected with silver wire for current collecting. The performances of the prepared single cells were tested at 600 °C, 650 °C and 700 °C with humidified hydrogen (3% H₂O) as fuel and air as oxidant respectively.

3. Results and discussion

Fig. 1 shows XRD patterns of the SSDCX powders prepared by the glycine nitrate process. All powders have fluorite structures and the structures are similar to the patterns of pure ceria. This means that the structure of ceria does not change by codoping proper amount of Sm₂O₃ and Sc₂O₃. In addition, a small shift of the diffraction peaks to higher 2θ angles is observed as the doping amount of Sc₂O₃ increases. This demonstrates the contraction in cell parameter according to Bragg equation, which may be caused by the smaller ionic radius of Sc³⁺ (0.087 nm) than Sm³⁺ (0.1079 nm).

The lattice parameters of SSDCX powders were derived from the XRD patterns and correlated with the doping amount of Sc₂O₃ as shown in Fig. 2. The parameter values decrease from 5.438 Å to 5.417 Å as the concentration of Sc increases from zero to 0.08. Moreover, it is obvious that the dependence of the lattice parameter on the doping amount is far from linear and the quadratic coefficient is explained as the degree of oxygen vacancy ordering [23,24].

Fig. 3 shows scanning electron micrographs of the Sc_{0.04}Sm_{0.16}Ce_{0.8}O_{1.9} (SSDC4) samples sintered at different temperatures. All samples are dense with only a few closed pores

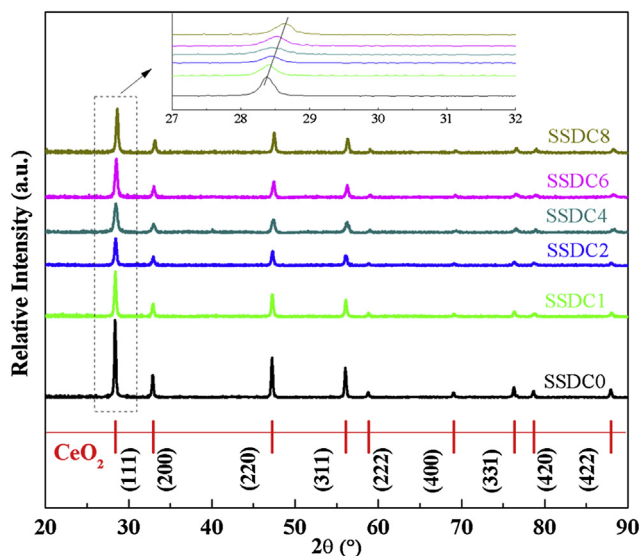


Fig. 1. XRD patterns of the prepared Sc_xSm_{0.2-x}Ce_{0.8}O_{1.9} (SSDCX) powders after calcination at 900 °C for 2 h.

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