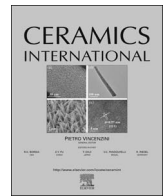




Contents lists available at ScienceDirect

Ceramics International

journal homepage: www.elsevier.com/locate/ceramint

Enhanced ionic conductivity of scandia-ceria-stabilized-zirconia (10Sc1CeSZ) electrolyte synthesized by the microwave-assisted glycine nitrate process

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ARTICLE INFO

Keywords:

- A. Microwave processing
- A. Sintering
- C. Ionic conductivity
- E. Fuel cells

ABSTRACT

Scandia-stabilized-zirconia is a potential zirconia-based electrolyte for intermediate temperature solid oxide fuel cells (IT-SOFCs). In this study, the properties of zirconia co-doped with 10 mol% Sc and 1 mol% Ce (scandia-ceria-stabilized-zirconia, 10Sc1CeSZ) electrolyte synthesized by the microwave-assisted glycine nitrate process (MW-GNP) were determined. The effects of microwave heating on the sintering temperature, microstructure, densification and ionic conductivity of the 10Sc1CeSZ electrolyte were evaluated. The phase identification, microstructure and specific surface area of the prepared powder were investigated using X-ray diffraction, transmission electron microscopy and the Brunauer-Emmett-Teller technique, respectively. Using microwave heating, a single cubic-phase powder was produced with nanosized crystallites (19.2 nm) and a high specific surface area (16 m²/g). It was found that the relative density, porosity and total ionic conductivity of the 10Sc1CeSZ electrolyte are remarkably influenced by the powder processing method and the sintering temperature. The pellet sintered at 1400 °C exhibited a maximum ionic conductivity of 0.184 S/cm at 800 °C. This is the highest conductivity value of a scandia-stabilized-zirconia based electrolyte reported in the literature for this electrolyte type. The corresponding value of the activation energy of electrical conductivity was found to be 0.94 eV in the temperature range of 500–800 °C. Overall, the use of microwave heating has successfully improved the properties of the 10Sc1CeSZ electrolyte for application in an IT-SOFC.

1. Introduction

Solid oxide fuel cells (SOFC) usually operate at very high temperatures ranging from 600 to 900 °C [1]. Reduction of the operating temperature can be achieved by developing new materials and adopting thin film techniques [2]. High ionic conductivity, low thermal expansion, negligible electronic conduction and good mechanical properties are the important factors to be considered in selecting the electrolyte for intermediate temperature SOFCs (IT-SOFCs) [3]. Stabilized zirconia such as yttria-stabilized-zirconia (YSZ) has been considered the most promising solid electrolyte material for SOFC due to its high phase stability, high ionic conductivity and low electronic conductivity

in both the oxidizing and reducing environment of an SOFC [4]. However, YSZ exhibits poor ionic conductivity at lower operating temperatures (< 700 °C). Therefore, wide attention has been focused on improving the ionic conductivity of the YSZ electrolyte [5]. One approach is the use of scandium oxide (Sc₂O₃) to stabilize ZrO₂ to improve the conductivity at lower operating temperatures [6,7]. The cubic fluorite-type phase of scandia-stabilized-zirconia (ScSZ) has been reported to be an excellent electrolyte material for IT-SOFC. However, ScSZ exhibits a phase transition from the highly conductive cubic phase to a low conductive rhombohedral or tetragonal phase at the IT-SOFC operating condition [8]. In previous studies, Al₂O₃ and the oxides of rare earth elements such as CeO₂, Sm₂O₃, Yb₂O₃, and Gd₂O₃ were used

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<http://dx.doi.org/10.1016/j.ceramint.2017.03.135>

Received 5 November 2016; Received in revised form 3 March 2017; Accepted 21 March 2017
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as dopants to mitigate the undesirable phase transition [9–12]. Several authors have reported that the phase transition of ScSZ can be prevented by doping ZrO_2 with Sc_2O_3 and CeO_2 [13,14]. According to previous reports, the 10 mol% Sc_2O_3 –1 mol% CeO_2 –89 mol% ZrO_2 composition (10Sc1CeSZ) enables the retention of the highly conductive cubic phase at the IT-SOFC operating temperature [15,16].

The performance of the 10Sc1CeSZ electrolyte can be improved by optimizing the powder characteristics, especially by adopting a suitable preparation technique. Powder properties such as particle size, surface area and purity are influenced by synthesis or preparation methods [17]. Thus, several powder synthesis methods have been applied for the synthesis of 10Sc1CeSZ electrolyte materials, such as the glycine-nitrate process [6,14], solid-state reaction [18], co-precipitation [19–21], the sol-gel method [21,22], the modified Pechini method [23] and the solid-liquid method [24]. Each method has its own respective advantages and disadvantages. Every method influences the final ionic conductivity depending on the powder properties and level of impurities. Solution combustion synthesis is an effective and rapid method to produce nanosized ceramic oxide powders at low calcination temperatures. This process involves a self-sustained combustion of a homogenous solution containing metal nitrates and fuels (urea, glycine, sucrose). The reaction temperature and reaction rate are the important parameters that govern phase formation during combustion synthesis. Microwave heating is a mature technique that has been widely used to replace the conventional heating technique in combustion synthesis methods to produce nanosized oxide powders within a shorter time [25–28]. Microwave energy generally provides uniform heating and promotes faster reaction kinetics, which consequently increases the reaction rate and homogeneity of the final product. In conventional heating, the heat is slowly transferred through the walls of the vessel to the reactants. This is a relatively slow and inefficient method for transferring energy into the reacting system. Owing to its advantages, in recent years, researchers have employed the microwave heating technique to produce materials for SOFC applications [25,29,30]. However, the solution combustion synthesis of the 10Sc1CeSZ electrolyte using microwave heating has not been previously reported, with the exception of a recent study that used the microwave-assisted hydrothermal method [28] to synthesize the identical composition.

Based on these advantages, the present study aimed to investigate the effect of the microwave-assisted glycine nitrate process (MW-GNP) on the phase formation, microstructure, surface area and ionic conductivity of 10Sc1CeSZ. This study also aimed to evaluate the influence of the sintering temperature on the microstructure, density and electrochemical properties of 10Sc1CeSZ electrolytes. The grain size of the pellets sintered at various temperatures ranging from 1300 °C to 1500 °C was observed by field-emission scanning electron microscopy (FESEM). Electrochemical impedance spectroscopy (EIS) was also applied to investigate the ionic conductivity of the electrolytes from 400 °C to 800 °C.

2. Experimental procedure

2.1. Synthesis of materials

Zirconia co-doped with 10 mol% Sc and 1 mol% Ce (scandia-ceria-stabilized-zirconia, 10Sc1CeSZ) electrolyte was synthesized by the microwave-assisted glycine nitrate process (MW-GNP). For the synthesis of 10Sc1CeSZ powder, the starting materials were scandium oxide (Sc_2O_3 , Aldrich), nitric acid (HNO_3 , Friendemann Schmidt), zirconium (IV) oxynitrate hydrate ($ZrO(NO_3)_2 \cdot H_2O$, Acros Organics), cerium(III) nitrate hexahydrate ($Ce(NO_3)_3 \cdot 6H_2O$, Aldrich) and glycine ($C_2H_5NO_2$, Genemark). The stoichiometric amount of scandium oxide was first dissolved in a hot nitric acid solution under stirring on the hot plate to form a clear solution of scandium nitrate. The stoichiometric amount of zirconium (IV) oxynitrate hydrate and cerium (III) nitrate hexahydrate

were then dissolved in deionized water and mixed with the prepared scandium nitrate solution. After that, glycine as a fuel was added to the resulting solution with a glycine to nitrate ratio of 1:2. The solution was continuously stirred until the solution become viscous. The viscous solution was introduced to the microwave with a power of 80%, and self-combustion occurred in the microwave oven (SHARP, 2450 MHz, 900 W). Finally, the ash was calcined at 700 °C for 2 h in a muffle furnace to obtain 10Sc1CeSZ oxide powder with the desired composition.

2.2. Powder and electrochemical characterization

The prepared 10Sc1CeSZ electrolyte powder was physically characterized. The thermal decomposition behaviour of the uncalcined 10Sc1CeSZ was investigated via thermogravimetry (TGA) and differential thermal (DTA) analysis (PerkinElmer, STA 6000, USA) by ramping the sample from 30 °C to 800 °C with a heating rate of 10 °C/min. The phase characterization, theoretical density, and crystallite size of the calcined powder were determined by X-ray diffraction (XRD) using an X-ray diffractometer (Shimadzu XRD-6000, D8-Advance, Bruker, Germany) with $CuK\alpha$ ($\lambda=0.15418$ nm) radiation and 2θ ranging from 20° to 90°. Energy-dispersive spectroscopy (EDS) was conducted to determine the elemental composition of the powders and samples were coated with a thin layer of gold. The microstructure and particle size of the prepared powders were also observed through transmission electron microscopy (TEM; Philips CM12, OR, USA) at 80 kV. The specific surface areas of the calcined powders were measured with a surface area analyser (Micromeritics, ASAP 2020, Georgia, USA) by applying the Brunauer–Emmett–Teller technique with nitrogen gas as the adsorbate.

The 10Sc1CeSZ powder was then cold-pressed at 48 MPa into cylindrical pellets with a 25 mm diameter and 1.3 mm thickness using a uniaxial die-press. The as-prepared pellets were then sintered at 1300 °C, 1400 °C or 1500 °C for 5 h in air. Field emission scanning electron microscopy (FESEM) (Zeiss Supra-55VP) was performed to examine the microstructure, particle size, shape, and morphology of the sintered pellets. The density of a pellet was determined by the Archimedes method using deionized water at room temperature. The conductivities of the sintered pellets were measured from 500 °C to 800 °C using impedance spectroscopy. Silver paste was applied to both sides of the sintered samples and heated at 800 °C for 1 h. The effective working area of the pellet was 1 cm². The silver electrode area and thickness of the pellet were used to determine the final conductivity. The measurement was conducted at 50 °C intervals with a flow of compressed air at 100 ml/min. The test was performed in potentiostatic mode using an Autolab PGSTAT302N coupled with a frequency response analyser (Autolab 302, Eco Chemie, Netherlands) over the frequency range from 10⁶ Hz to 0.1 Hz under a low-amplitude sinusoidal voltage of 10 mV. Data acquired from the impedance testing were analysed using NOVA software (Version 1.11). AC-impedance diagrams were plotted in Z' (real) versus Z'' (imaginary) to measure the conductivity of the pellets sintered at different temperatures.

3. Result and discussion

3.1. Thermogravimetry and differential thermal analysis

TGA and DTA data for the 10Sc1CeSZ synthesized by the microwave-assisted glycine nitrate process (MW-GNP) is presented in Fig. 1. The TGA was conducted to examine the thermal behaviour of the 10Sc1CeSZ powder. Three stages of significant weight loss at temperatures of ~200 °C, 200–350 °C and 600 °C were observed. The first stage of weight loss in the TGA occurred below 200 °C, indicating the desorption of water or evaporation of moisture from the powder. The second stage of weight loss occurred in the temperature range of 200–350 °C. This major weight loss could be associated with the decom-

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