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Preparation, structure and electrical conductivity of pyrochlore-type samarium–lanthanum zirconate ceramics

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ABSTRACT

A series of zirconate compounds with the general formula $Sm_{2-x}La_xZr_2O_7$ ($0 \leqslant x \leqslant 1.0$) were prepared by pressureless-sintering method at 1973 K for 10 h in air. The relative density, structure and electrical conductivity of $Sm_{2-x}La_xZr_2O_7$ ceramics were investigated by the Archimedes method, X-ray diffraction and impedance spectroscopy measurements. $Sm_{2-x}La_xZr_2O_7$ ($0 \leqslant x \leqslant 1.0$) ceramics exhibit a pyrochlore-type structure. The measured electrical conductivity of $Sm_{2-x}La_xZr_2O_7$ ceramics obeys the Arrhenius relation and gradually increases with increasing temperature from 673 to 1173 K. $Sm_{2-x}La_xZr_2O_7$ ceramics are oxide-ion conductors in the oxygen partial pressure range of 1.0×10^{-4} to 1.0 atm at all test temperature levels. The electrical conductivity of $Sm_{2-x}La_xZr_2O_7$ ceramics decreases with increasing lanthanum content at identical temperature levels.

1. Introduction

Solid oxide fuel cells (SOFCs) are highly efficient devices that can convert chemical energy of a fuel into electrical energy. SOFCs have drawn much attention due to environmental friendliness, and fuel flexibility including hydrogen, natural gas, petroleum gas, coal gas, biogas, etc. [1,2]. Conventional SOFCs use typical 8 mol.% Y_2O_3–ZrO_2 (YSZ) electrolytes at operating temperatures of 1173–1273 K, and anode and cathode materials are Ni–YSZ cermets and $La_{1-x}Sr_xMnO_3$ oxides, respectively [3–5]. However, the high operating temperature presents strict demands on SOFC materials and technology. When SOFCs are operated at high temperatures for a long time, unwanted chemical reactions take place at the cathode/electrolyte interface. This will generate insulating zirconates of $La_2Zr_2O_7$ and $SrZrO_3$, which have a detrimental effect on SOFCs performance [6]. Lanthanide zirconium oxides with the general formula $A_2Zr_2O_7$-type (A = lanthanides) structure show excellent electrical properties owing to high compositional diversity, structural flexibility and intrinsic concentration of oxygen vacancies [7,8]. Shinozaki et al. reported that the electrical conductivity of $Sm_2Zr_2O_7$ was comparable to those of other good oxide-ion conductors in the low-temperature region, and was approximately constant in an oxygen partial pressure range of 1.0×10^{-20} to 1 atm below 1087 K [9]. In last several decades, many efforts have been made in order to improve the electrical conductivity of solid electrolytes [10–12]. It is well known that the electrical conductivity of solid electrolytes is affected by many factors such as structure, ionic radius of doping elements, and oxygen vacancy concentration, etc. [13–15]. One strategy for the development of commercially viable SOFCs technology is to reduce the operating temperature. Reduction in cost of SOFCs is of importance for the commercialization of SOFCs. Recently, new $A_2Zr_2O_7$-type ternary oxides are of considerable scientific interest owing to variable cation radius ratios of $r(A^{3+})/r(Zr^{4+})$ [6,16–18]. A significant increase in the electrical conductivity was found by suitable substitution of isovalent rare-earth cations like Sm at the Gd site in $Gd_2Zr_2O_7$ in the temperature range of 623–873 K [16]. However, the electrical conductivity of $(Gd_{1-x}La_x)_2Zr_2O_7$ ($0 \leqslant x \leqslant 0.5$) ceramics was almost La-content independent in the temperature range of 773–1023 K [6]. For $(Sm_{1-x}Yb_x)_2Zr_2O_7$ ($0 \leqslant x \leqslant 1.0$) and $(Sm_{1-x}Y_x)_2Zr_2O_7$ ($0 \leqslant x \leqslant 0.5$) ceramics, the electrical conductivity of pyrochlore-type compositions are clearly higher than that of defect fluorite-type compositions in the temperature range of 723–1173 K [17,18]. In the present work, $Sm_{2-x}La_xZr_2O_7$ ($0 \leqslant x \leqslant 1.0$) ceramics were prepared by pressureless-sintering method at 1973 K for 10 h in air to investigate their structure and electrical conductivity.

2. Experimental

$Sm_{2-x}La_xZr_2O_7$ ($0 \leqslant x \leqslant 1.0$) ceramic powders were prepared using a standard solid state reaction of dried metallic oxides, La_2O_3, Sm_2O_3 (Grirem Advanced Materials Co. Ltd., China; purity ⩾ 99.9%), and ZrO_2 (Dongguan SG Ceramics Technology Co. Ltd., China; purity ⩾ 99.9%) powders. Stoichiometric proportions of La_2O_3, Sm_2O_3 and ZrO_2 powders were mechanically mixed in analytically pure alcohol for 24 h, dried and then heated at 1673 K for 6 h in air. In order to obtain a better homogeneity, the

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