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Enhanced performance of nanocrystalline Cu-doped $Pr_{0.6}Sr_{0.4}FeO_3$ as cathode for solid oxide fuel cell

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ABSTRACT

$Pr_{0.6}Sr_{0.4}FeO_3$ (PSF) and $Pr_{0.6}Sr_{0.4}Fe_{0.8}Cu_{0.2}O_3$ (PSFCu) perovskite materials were prepared by glycine nitrate method as a cathode material for intermediate temperature solid oxide fuel cells (IT-SOFC). The prepared PSF and PSFCu materials exhibited typical orthorhombic structure and highly porous morphology with high surface area (43–55 m^2/g). Upon the 20% Cu doping on the B-site of the PSF, the electrical conductivity increased from 247 S cm^{-1} (PSF cathode) to 298 S cm^{-1} and polarization resistance of symmetry cell decreased from 0.21 Ω cm^2 (PSF cathode) to 0.14 Ω cm^2 as compared to PSF cathode (0.21 Ω cm^2) at 850 °C. Cu doping in B-site of the perovskite generates the oxide ion vacancies which lead the high effective charge carriers, resulting in the enhanced electrochemical performance of PSFCu cathode.

1. Introduction

During last decade extensive researches have been carried out to find out suitable SOFC materials that can be operated at low temperature (500–800 °C). Among the various components involved in SOFC, cathode material is one of the important parts to determine the overall fuel cells performance. A mixed ionic and electronic conductor (MIEC) appears to be a promising way to decrease the cathode polarization resistance. The Fe-based perovskite compounds are one of the main materials for IT-SOFC because of their mixed ionic and electronic conductivity. Praseodymium ferrite ($PrFeO_3$) is expected to be more stable than cobaltite perovskite because Fe^{3+} ion has a stable electronic configuration $3d^5$. Co and Ni-doped $Pr_{1-x}Sr_xFeO_3$ (PSF) cathodes have shown enhanced performance with respect to electronic and ionic conductivity [1,2]. Copper oxide is also considered to possess superior electrical conductivity and high ionic conductivity. The Cu-doped lanthanum strontium ferrite showed superior kinetics for the electrochemical reduction of oxygen [3]. However, the Cu-doped composition exhibits chemical interaction with YSZ (typically above 950 °C), resulting in the precipitation of monoclinic zirconia.

Recently [4], it has been found that replacing lanthanum by smaller lanthanide elements led to reduced reactivity towards electrolyte materials, improved catalytic activity and decreased cathodic polarization [5]. Among perovskites with different rare earth cations at the A-site, Pr^{3+} incorporated materials exhibited the highest electrical conductivity with the lowest over-potential values [6]. In this paper, PSF and PSFCu perovskite as new cathode materials were synthesized and characterized on the morphological and electrical properties by fabricating the symmetric cells. Cu doping in B-site of the perovskite PSF material showed enhanced electrical conductivity and low decrease polarization resistance in cathode material.

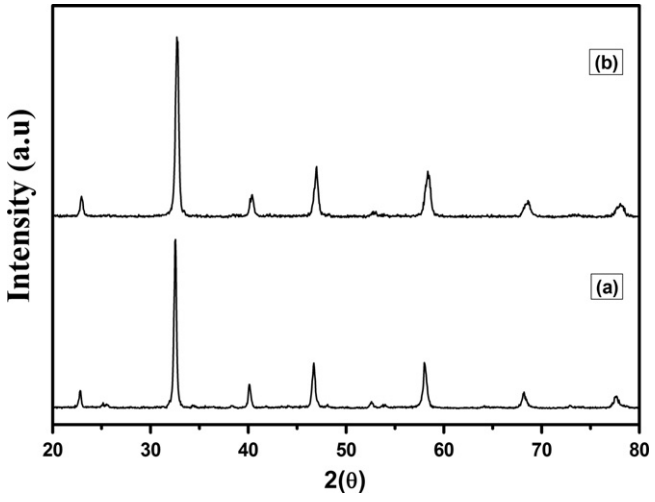

Fig. 1. The XRD patterns of PSF (a) and PSFCu and (b) synthesized at 700 °C.

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2. Experimental

$Pr_{0.6}Sr_{0.4}FeO_3$ and $Pr_{0.6}Sr_{0.4}Fe_{0.8}Cu_{0.2}O_3$ powders were prepared by a glycine nitrate process (GNP). $Pr(NO_3)_3 \cdot 6H_2O$ (Sigma–Aldrich, 99.9%), $Sr(NO_3)_2$ (Samchun chemicals, 98.5%), $Fe(NO_3)_3 \cdot 9H_2O$ (Samchun chemicals, 98.5%), and $Cu(NO_3)_2 \cdot 3H_2O$ (Sigma–Aldrich, 99.9%) were used as starting raw materials. Glycine (Junsei chemicals, 99%) was used as oxidizer and fuel. Metal nitrates (corresponding molar ratio for each metal) were dissolved in distilled water, and then Glycine (2 M) was added into the nitrate (1 M) aqueous solution under vigorous stirring. The solution mixture was heated to 200 °C on a hot plate. The viscous gel causes a combustion reaction to make a dark grey ash. Thus obtained ash was calcined at 700 °C in an air to remove the carbon residues in the ash and to form a crystalline structure. The milled powders were pressed into pallets and sintered in an air at 1200 °C for 5 h.

The structure and phase stability of the materials were investigated by X-ray diffraction (XRD) analysis using a Rigaku D/Max diffractometer with Cu Kα radiation. The morphology and microstructure of the sintered electrodes were examined by scanning electron microscope (SEM, JEOL JSM-6400) at 20 KV accelerating voltage. The electrical conductivities of the PSF and PSFC materials were measured using a standard four probe DC

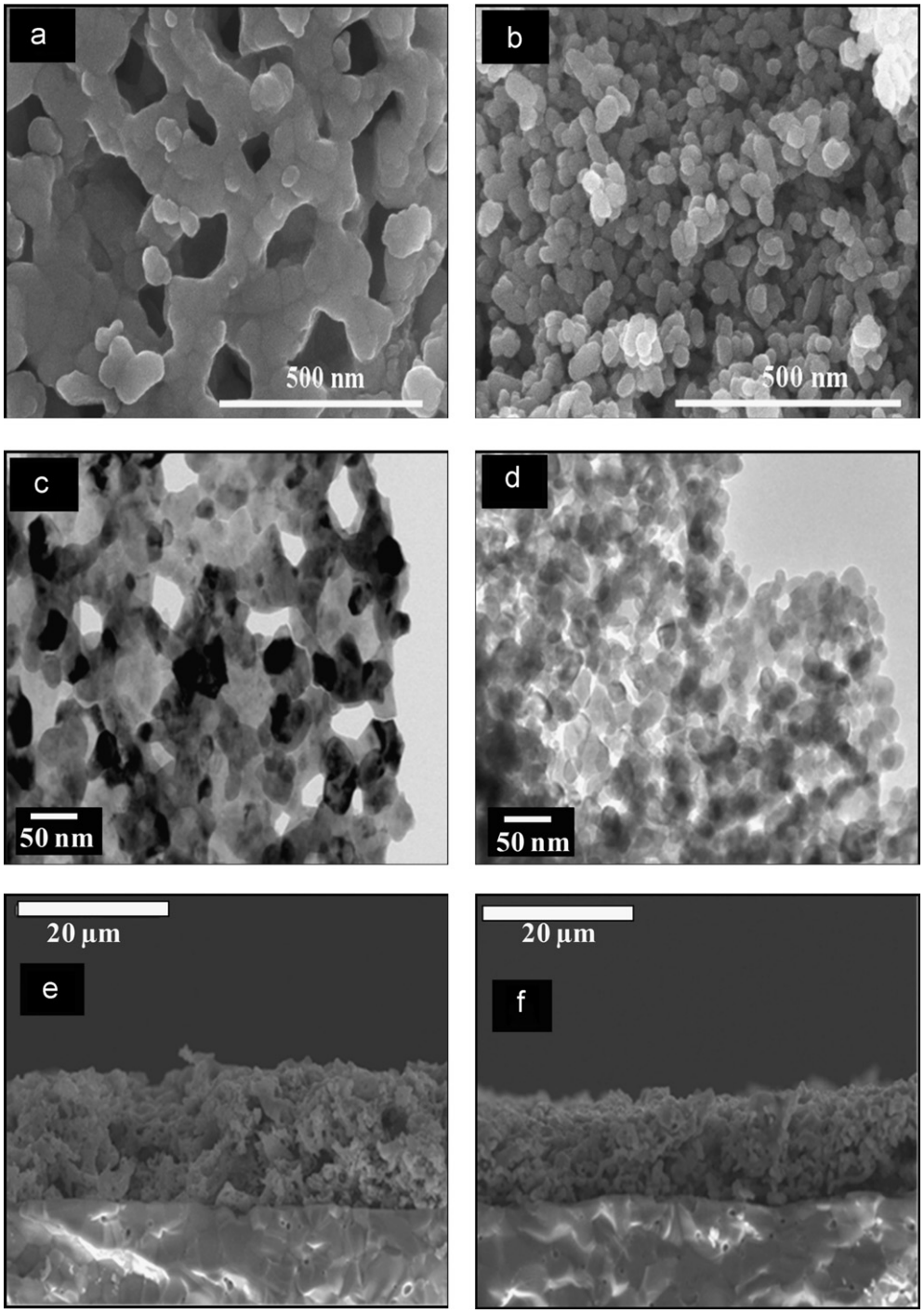

Fig. 2. SEM (a and d) and TEM (b and e) images of the powders synthesized at 700 °C for PSF (a and b) and PSFCu (d and e). Cross-section view of SDC-PSF (c) and SDC-PSFC (f) interface.