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Fabrication and electrochemical performance of nickel- and gadolinium-doped ceria-infiltrated $La_{0.2}Sr_{0.8}TiO_3$ anodes for solid oxide fuel cells

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HIGHLIGHTS

- Lanthanum doped Strontium Titanate is used in an oxide-based anode backbone.
- An oxide-based anode with Ni and GDC nano particles was fabricated by infiltration.
- Ni and GDC-infiltrated anodes exhibited remarkably reduced polarization resistance.
- We investigated the role of Ni and GDC on the performance of infiltrated anodes.

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ABSTRACT

In this work, nickel and gadolinium-doped ceria (GDC)-infiltrated lanthanum strontium titanate (LST) anodes are fabricated, and their electrode performances under a hydrogen atmosphere is investigated in terms of the Ni:GDC ratios and cell operating temperature. The Ni/GDC-infiltrated LST anode exhibits excellent electrode performance in comparison with the Ni- or GDC-infiltrated anodes, which is attributed to the synergistic effect of an extended triple-phase boundary length by GDC and good catalytic activity for hydrogen oxidation because of the Ni particles. The polarization resistances (R_p) of Ni/GDC-infiltrated LST are 0.07, 0.08, and 0.12 Ω cm^2 at 800, 750, and 700 °C, respectively, which are approximately three orders of magnitude lower than that of the LST anode (68.5 Ω cm^2 at 700 °C). The effect of Ni and GDC on the electrochemical performance of LST was also investigated by using electrochemical impedance spectroscopy (EIS). The anode polarization resistance (R_p) is confirmed to be dependent on the content and dispersion state (microstructure) of the Ni and GDC nanoparticles.

1. Introduction

Ni/8 mol% yttria-stabilized zirconia (YSZ) cermet is widely employed as a solid oxide fuel cell (SOFC) anode material because of its high electrical conductivity and good catalytic activity at high temperatures under reducing conditions. However, it is particularly prone to degradation upon carbon coking, oxidation, and sulfur poisoning [1,2,3,4,5,6]. Therefore, several studies have been conducted in search of alternative SOFC anode materials. The most promising candidate materials developed to date are ceramic perovskites such as strontium titanium oxide ($SrTiO_3$). A general chemical formula for a perovskite compound is ABX_3, where A and B are two cations of different sizes, and X is an anion that forms bonds with both these cations [7,8].

Among the various ceramic perovskites, rare earth metal-doped $SrTiO_3$ are some of the most promising alternative anode materials to have been developed for SOFCs [9]. In particular, lanthanum is an appropriate donor dopant because its ionic radius is similar to that of strontium [10]. The introduction of trivalent La into the divalent Sr A-site in the $SrTiO_3$ structure and subsequent heat treatment under a reducing atmosphere leads to the reduction of Ti^{4+} to Ti^{3+}, which improves the electronic charge concentration. Therefore, La-doped $SrTiO_3$ exhibits n-type conductivity under H_2 atmosphere [11]. In addition, it has outstanding dimensional/chemical stability upon redox cycling, excellent carbon deposition resistance, and a high tolerance for sulfur in hydrocarbon fuels [12,13,14,15]. However, lanthanum strontium titanate (LST) suffers from low electrocatalytic activity with respect to hydrogen oxidation reactions. The electrochemical performances of SOFCs using the LST anode were quite poor compared to those with the Ni/YSZ anodes [10,16,17]. Thus, it is necessary to improve the catalytic activity of the LST anode. Some researchers have reported improvements in the electrochemical properties of LST anodes by infiltration or ex-solution methods [18,19,20].

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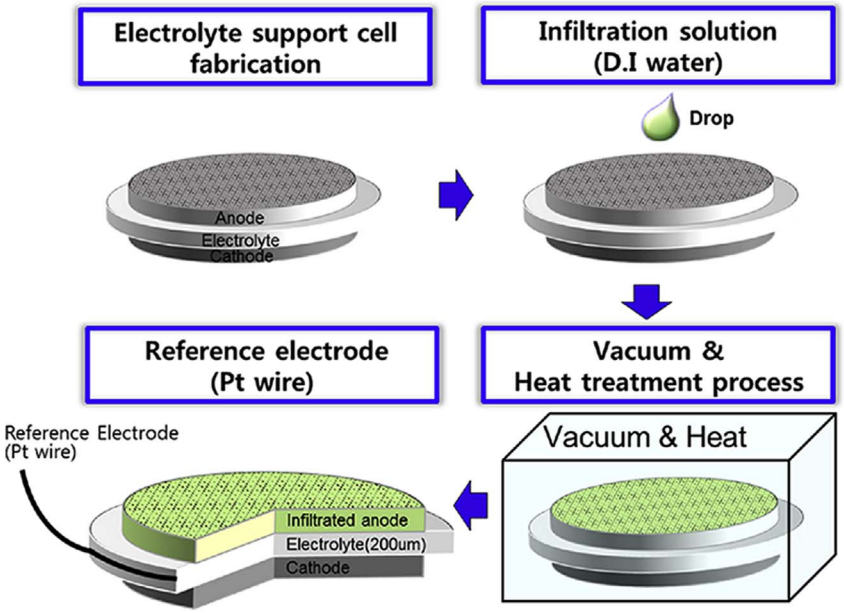

Fig. 1. Schematics of the fabrication of an electrolyte-supported cell with infiltrated anodes.

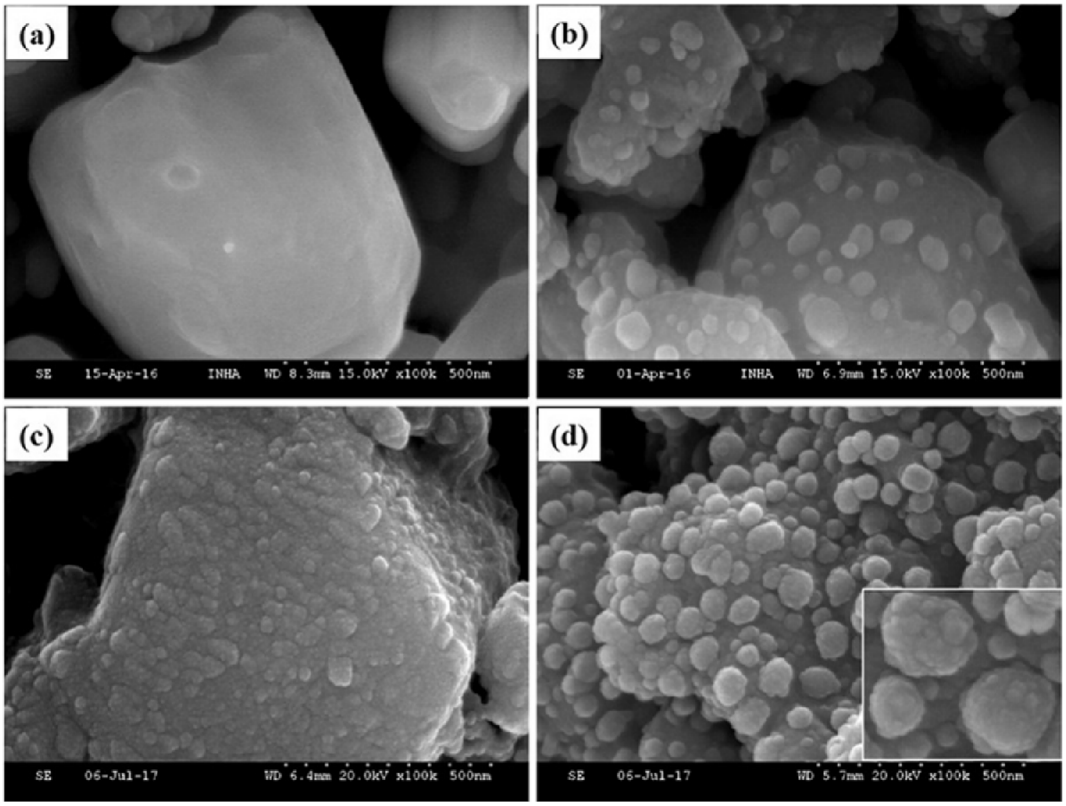

Fig. 2. SEM images of (a) un-infiltrated LST, (b) Ni-infiltrated LST, (c) GDC-infiltrated LST, and (d) Ni/GDC-infiltrated LST.

In this study, Ni and Gd-doped ceria ($Ce_{0.8}Gd_{0.2}O_{1.9}$, GDC)-infiltrated $La_{0.2}Sr_{0.8}TiO_3$ (LST) anodes were prepared in order to improve the catalytic activity of the LST-based oxide anode. In general, La doping results in an increase in the conductivity of strontium titanate. However, secondary phase ($La_2Sr_7TiO_{11}$) formation becomes more pronounced at high La doping concentrations in $SrTiO_3$ [21]. Therefore, the La doping concentration was fixed at 20 mol%. Ni and GDC nanoparticles were homogeneously dispersed in the LST backbone by infiltration. The electrochemical impedance spectra (EIS) were analyzed to understand the effects of Ni and GDC on anode performance. Furthermore, the polarization resistances (R_p) of the infiltrated LST anodes were investigated in terms of the Ni and GDC contents.

2. Experimental

The stoichiometric perovskite $La_{0.2}Sr_{0.8}TiO_3$ (LST) was synthesized via a conventional solid-state reaction (SSR) method. The starting materials were La_2O_3 (99.9%, Sigma-Aldrich, USA), $SrCO_3$ (99.9%, Sigma-Aldrich, USA), and TiO_2 (99.9%, Sigma-Aldrich, USA). These powders were planetary-milled in ethanol (94.5%, Samchun Pure Chemicals,