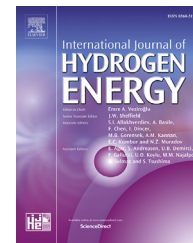


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Effect of anode calcination on the performance and redox stability of low-temperature solid oxide fuel cells prepared via impregnation

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ARTICLE INFO

Article history:

Received 9 August 2017

Received in revised form

25 September 2017

Accepted 25 October 2017

Available online 16 November 2017

Keywords:

Solid oxide fuel cells

Redox stability

LSGM

Anode

Impregnation

Electrochemistry

ABSTRACT

Development of low-temperature solid oxide fuel cells (SOFCs) is crucially important for their stable and continuous operation as a reliable power source. With Ni nanoparticles impregnated into the thick anode support, Sr- and Mg- doped LaGaO₃ (LSGM) based electrolyte has been demonstrated to be adept at reducing the ohmic resistance for high performance at low temperatures (450–650 °C). In this study, 55-μm LSGM-electrolyte-based SOFCs are prepared via the impregnation for both cathode and anode, and high performance is obtained at low temperatures. The durability and reduction-oxidation (redox) stability are examined in terms of the calcination temperature of LSGM/Ni(O) anode that determines the thermal growth of NiO crystals and interfacial reaction between NiO and LSGM. We find that the cell with anode calcined at 1100 °C does contain larger NiO grains and more secondary phases, but it shows similar power density comparing to the one calcined at 800 °C. Both cells are stable at 650 °C and no damage on the electrolyte is found during the redox cycle, but the performance of the former is more sensitive to the redox cycle due to the high ohmic resistance arisen from the possible discontinuous connection between the large Ni grains.

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Introduction

Solid oxide fuel cells (SOFCs) are electrochemical devices that convert chemical energy directly into useful electrical energy [1,2]. State-of-the-art anode-supported SOFCs involving high-

temperature sintering of NiO/yttria-stabilized ZrO₂ (YSZ) during the processing of composite electrode contains micrometer Ni(O), while the technique of impregnation can effectively preserve the nanometer size of metal catalyst by two-step sintering [3–5]. In an impregnation process, a porous

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<https://doi.org/10.1016/j.ijhydene.2017.10.142>

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scaffold of the electrolyte material, such as YSZ or Sr- and Mg- doped LaGaO₃ (LSGM), was prepared via high-temperature sintering along with the solid electrolyte and then the precursors of electrocatalysts, such as metals and oxides, is impregnated in to the well-sintered scaffold to obtain nanoscale catalyst after the second low-temperature conversion reaction [5–7]. YSZ is the widely-used electrolyte and scaffold for the impregnation process due to the chemical stability and high mechanical tolerance for high porosity. However, as the emerging SOFCs need to function ideally at low temperature (<650 °C) to reduce the degradation of structural components that would arise from the high temperature corrosion [8] or the performance deterioration due to the thermally-induced compositional and morphological variation [9,10], the oxide-ion conductivity of YSZ is insufficient to yield an ohmic resistance below 0.1 Ω cm² at temperatures below 700 °C with a thickness of 10 μm. Therefore, LSGM as an electrolyte material showing much higher conductivity than YSZ [7,11,12] and higher oxide-ion transport number than ceria under reducing atmosphere has been investigated as the scaffold material for an SOFC with nanoscale electrocatalysts for low-temperature operation [13]. With nickel impregnation into porous LSGM scaffold supporting a thin LSGM electrolyte, a power density higher than 1.2 W cm⁻² has been achieved at 600 °C [13,14], which is very promising as a low-temperature fuel cell. Ni(O) is also impregnated into a thin LSGM porous anode functional layer on a thick Sr_{0.8}La_{0.2}TiO_{3-δ} porous support for power densities higher than 1.12 W cm⁻¹ at 650 °C [15,16]. Liu et al. [17] demonstrated a thin LSGM electrolyte based cell with the impregnation of Ni and ceria into the scaffold prepared using freeze-drying tape casting and slurry drop-coating technique, and a power density of 0.99 W cm⁻² was achieved at 650 °C.

For a practical SOFC aiming to work for forty-thousand hours, the stability is important for a constant output of electricity [18,19] even if some unintentional interruptions of fuel could happen [20,21]. The interfacial reaction between NiO and LSGM under firing at elevated temperature (above 1200 °C) is reported to cause the formation of a LaSrGa(Ni)O_{4-δ}-type perovskite or other insulating phases [22], resulting in uncertainty on the performance of Ni(O)-impregnated anode. The employment of impregnation technique could reduce this interaction between NiO and LSGM since only a low-temperature sintering is required for the formation of Ni(O) and, under an anodic current in reducing atmosphere, the presence of NiO is restricted to the very surface of the Ni particles. The degradation of electrode with nanoscale electrocatalysts was attributed to the growth of particles that decreased the triple phase boundary where reduction or oxidation reactions took place [20]. The addition of TiO₂, CeO₂ and Al₂O₃ on YSZ/Ni(O) scaffold has been investigated as sintering inhibitors to reduce the grain growth of the nickel based composite anode [23,24]. Likewise, the interfacial reaction between NiO and LSGM under oxidizing condition could cause the formation stable oxides as a sintering inhibitor, but very few studies has been done on the impact of interfacial reaction on the performance of LSGM/Ni(O) anode prepared via impregnation. In the meantime, the sintering temperature of NiO/LSGM in air will change the grain size of NiO (and the resultant Ni) besides the extent of interaction between NiO

and LSGM. The solid-state reaction has been reported to be a problem for the Ni(O) based anode on an LSGM electrolyte due to the solid state reaction between them to form SrLaGa₃O₇ and LaNiO₃ on the interface under sintering at 1400 °C or 1200 °C for the conventional processing of anode support fuel cells [25,26]. The interaction between LSGM and NiO at lower temperatures could be inhibited but still needs to be investigated as Liu et al. [27] found extra peaks for the NiO/LSGM electrode fired even at 700 °C after the impregnation process, and it should be noted that the chemical stability of LSGM electrolyte/scaffold with NiO is dependent on the synthesis route and sintering temperature of the scaffold [12]. Moreover, in the conventional YSZ/Ni(O) composite electrode, the presence of nickel causes the undesired characteristics due to the 30% volume change if it is oxidized [28,29], called the “redox” instability. The volume change during the redox cycle damages the structural integrity of the fuel cell [30] and degrades the performance [31], but the redox stability of LSGM/Ni(O) prepared via infiltration has not been studied yet. In this study, we studied the effect of sintering temperature of LSGM/Ni(O) electrode after impregnation on the performance and redox stability of fuel cell with LSGM electrolyte.

Experimental

Stoichiometric amounts of Ga₂O₃, La₂O₃, SrCO₃ and MgO were mixed and fired at 1350 °C for 10 h with an intermediate calcination and grinding at 1100 °C for 10 h to obtain the monophasic La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{3-δ} (LSGM). The hand-ground powders of LSGM were refined by high-energy ball-milling at 1000 rpm for 30 min to reduce the particle size to less than 2 μm. The conductivity of LSGM was measured on pelletized sample (1 cm in diameter and 2 mm in thickness) of the refined powders sintered at 1400 °C for 5 h. Tape casting and lamination were used to obtain the tri-layer green tape. In order to obtain porosity for impregnation, 50 wt % graphite as pore former was mixed with LSGM for the slurry that was sequentially cast on the tape for dense electrolyte. The thickness of each layer was controlled by the gap between the doctor blade and the substrate. Two green tapes containing the bi-layer structure for electrolyte and porous layer were laminated together with the green tapes for electrolyte facing each other to give the tri-layer structure green tape [4]. The final scaffold was obtained after a firing at 1400 °C for 3 h and the effective area for the impregnation is 1.1 cm². The porosity and thickness of the trilayer were measured using scanning electron microscopy (SEM) on the sample mounted in resin epoxy and the images were processed and analyzed using ImageJ software (National Institute of Health, USA) [32].

The aqueous solution of nickel nitrate (1 M) with equal molar amounts of citrate acid was impregnated into the porous structure and calcined at 450 °C for the decomposition of nitrate and organics. Multiple impregnation steps were employed to increase the loading of nickel oxide. After the weight of NiO reached 10 wt % of the electrode, the impregnated scaffold was fired at 800 °C or 1100 °C for 4 h, followed by the impregnation of the cathode for La_{0.8}Sr_{0.2}CoO_{3-δ} (LSC) or Sm_{0.5}Sr_{0.5}CoO_{3-δ} (SSC). The solution for the LSC or SSC was prepared via dissolving the respective nitrate species and

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