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Infiltrated Pr₂NiO₄ as promising bi-electrode for symmetrical solid oxide fuel cells

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ABSTRACT

In-situ growth of nanoparticles on electrode surface for high temperature energy conversion devices is one of efficient ways for new electro-catalyst design. Here, perovskite-related Pr₂NiO₄ (PNO) has been evaluated as a novel symmetrical electrode for intermediate-temperature solid oxide fuel cells (IT-SOFCs). Nickel nanoparticles are exsolved and dispersed on the surface of PNO after exposed in H₂ at 800 °C as an efficient electrocatalyst for hydrogen oxidation reaction (HOR) as anode. The electro-activity towards oxygen reduction reaction (ORR) at cathode side is further improved by infiltration process. A synergetic effect of in-situ exsolved nanoparticles as well as infiltrated ionic conducting particles have improved HOR and ORR activity at anode and cathode side, respectively. Furthermore, symmetrical SOFC (S-SOFC) with infiltrated Pr₂NiO₄ as bi-electrode exhibits excellent short-term stability and reliable redox stability in repeated H₂/air cycles.

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Introduction

Solid oxide fuel cells (SOFCs) as an ideal power generation technology have attracted considerable attention because of their high efficiency and fuel flexibility, low emission, and high quality of exhaust heat [1,2]. Conventional SOFC is composed of a nickel cement anode, ionic or protonic conductor electrolyte, and an oxide cathode [3]. The anode and cathode were supposed to be separately fabricated, hence, inducing problems associated with component

degradation and increase of fabrication cost [4]. Herein, symmetrical solid oxide fuel cells (S-SOFCs) assembled with symmetrical electrodes have drawn increasing attention in recent years [5–10]. In this configuration, the same material is used as both anode and cathode, which demonstrated several advantages comparing to the traditional SOFCs. Firstly, such symmetrical configuration can substantially simplify the manufacture processes and save the cells fabrication cost. Moreover, carbon deposition and/or sulfur poisoning can be overcome or suppressed because of the possibility of reversing the supplied gases and oxidizing the possible carbon deposit

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and/or sulfur species, and consequently recovering the loss of output power density [12]. Additionally, the use of the same material as both anode and cathode will also minimize compatibility problems. Therefore, high-performance electrode materials are essential to simultaneously satisfy the oxygen reduction reaction (ORR) requirements for cathode and hydrogen oxidation reaction (HOR) for anode used for S-SOFCs. However, only a few materials have been proposed as symmetrical electrodes, owing to the low electrical conductivity and stability in both oxidizing and reducing environment [13–17].

Chromium based perovskite oxides were firstly proposed as symmetrical electrodes for S-SOFCs because they performed well as SOFC interconnects in both oxidizing and reducing atmospheres [18,19]. Other chromium-free perovskite oxides were also proposed as potential electrodes for S-SOFCs. For example, $\text{SrFe}_{0.75}\text{Zr}_{0.25}\text{O}_{3-\delta}$ exhibited a reasonable area-specific resistance (ASR) values of $0.10 \Omega \text{ cm}^2$ in air and $0.17 \Omega \text{ cm}^2$ in 5% H_2 -Ar at 750°C , and the S-SOFC based on $\text{SrFe}_{0.75}\text{Zr}_{0.25}\text{O}_{3-\delta}$ symmetrical electrodes and thick LSGM electrolyte delivered a maximum power density of 425 mW cm^{-2} at 800°C in wet H_2 [20]. Mo-doped $\text{Pr}_{0.6}\text{Sr}_{0.4}\text{Fe}_{0.8}\text{Ni}_{0.2}\text{O}_{3-\delta}$ was applied as bi-electrodes for S-SOFC and this fuel cell reached a power density 500 mW cm^{-2} at 800°C under wet H_2 [21]. $\text{La}_{0.4}\text{Sr}_{0.6}\text{Co}_{0.2}\text{Fe}_{0.7}\text{Nb}_{0.1}\text{O}_{3-\delta}$ (LSCFN) was reported by Xu et al. that exhibited good redox stability as both cathode and anode for S-SOFC [22]. Although these perovskite oxides have been developed as promising symmetrical electrodes, further improvement on performance is still needed for practical applications of S-SOFCs.

A_2BO_4 perovskite-related oxides have also been attracted much attention due to their unique structure, high oxygen ion diffusion and exchange properties [23,24]. The A_2BO_4 -type oxides had higher chemical stability, lower thermal expansion coefficient, and better thermally compatibility with yttrium stabilized zirconia (YSZ) or $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}$ (SDC) electrolytes than ABO_3 -type oxides [25]. In addition, the oxygen diffusion and surface exchange coefficient of A_2BO_4 -type oxides are much higher than ABO_3 -type oxides. Unlike forming interstitial oxygen at intersection point in ABO_3 , the low-order Ruddlesden-Popper compounds A_2BO_4 allows the introduction of O-interstitial defects on tetrahedral sites to generate O-vacancies on AO rock salt layer, inducing a rapid transport of oxygen ions in free space [26].

$\text{Pr}_2\text{NiO}_{4+\delta}$ perovskite-related oxides exhibited excellent performance when applied as cathode for intermediate temperature SOFCs. Chronoes et al. [27] revealed that the oxygen diffusion in $\text{Pr}_2\text{NiO}_{4+\delta}$ is highly anisotropic, occurring almost entirely via an interstitially mechanism in A–B plane. Partial Sr-substituted Pr_2NiO_4 ($\text{Pr}_{2-x}\text{Sr}_x\text{NiO}_4$) [28] was investigated as SOFCs cathode to stabilize the structure and increase oxygen non-stoichiometry of $\text{Pr}_2\text{NiO}_{4+\delta}$. Infiltration method has been applied to improve oxygen reduction reaction (ORR) activity of $\text{Pr}_2\text{NiO}_{4+\delta}$ electrodes. The infiltration solution typically containing metal nitrates is infiltrated into a porous $\text{Pr}_2\text{NiO}_{4+\delta}$ “scaffold” and dried. After a calcination, desired oxide phase is decorated on the surface of the electrode scaffold [29]. Further improvement on ORR activity of $\text{Pr}_2\text{NiO}_{4+\delta}$ was achieved via an infiltration method on porous LSGM scaffolds, and such a composite cathode with ~14 vol% $\text{Pr}_2\text{NiO}_{4+\delta}$ loading showed a

polarization resistance as low as $0.11 \Omega \text{ cm}^2$ at 650°C [30]. In addition, $\text{Pr}_4\text{Ni}_3\text{O}_{10+\delta}$ has been developed as a promising Ruddlesden-Popper oxides electrode with the polarization resistance as low as $0.16 \Omega \text{ cm}^2$ at 600°C in air [31]. Although several $\text{Pr}_2\text{NiO}_{4+\delta}$ perovskite-related oxides have been reported as promising cathode materials, to date, there are no reports showing it can be effectively applied as anode for SOFCs in this strategy.

Herein, we developed Pr_2NiO_4 (PNO) based composite materials as alternative electrode materials for symmetrical SOFCs via infiltration and in-situ reduction processes. In this in-situ reduction method, the transition-metal atoms were incorporated into a host perovskite lattice, and exsolved at the surface as metallic nano-particles under reducing conditions [32–34]. The nano-particles are uniformly distributed on the surface of host perovskite electrodes, consequently leading to a significant improvement on the electro-catalytic activity toward hydrogen oxidation reaction [35,36].

The crystal structure and morphology of PNO before and after a reduction in H_2 atmosphere were investigated. Ni nanoparticles were reduced and dispersed on the porous scaffold of the reduced PNO (r -PNO), which overwhelmingly improved catalytic activity towards H_2 fuel oxidation. Furthermore, $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}$ (SDC) was infiltrated to PNO scaffold to further improve ORR activity of cathode for S-SOFCs. Promising power output and stability were obtained in the S-SOFCs based on the infiltrated PNO as both anode and cathode.

Experimental

Sample preparation

Pr_2NiO_4 (PNO) powders were synthesized via glycine-nitrate combustion method. Stoichiometric amount of $\text{Pr}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (99.5%), $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (99.5%) and glycine were dissolved in deionized water according to the stoichiometry. The molar ratio of glycine to metal ions in the final solution was 3:1. Then the solution was heated up to 250°C in air until a combustion reaction finished. The as-synthesized powders were then calcined at 1200°C for 4 h in air.

The $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}$ (SDC) powders for electrolyte was synthesized using a glycine – nitrate process (GNP) as described above, using stoichiometric $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (99.5%) and $\text{Sm}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (99.5%) as raw materials. The combusted powders were then calcined at 900°C for 2 h in air to obtain pure SDC powders. The SDC powders were ground with 5 wt% polyvinyl alcohol (PVA) in a mortar and pressed into pellets ($\phi 15 \text{ mm}$) under 200 MPa and then sintered at 1400°C for 4 h in air.

The PNO and SDC powders were mixed with PVA and dry-pressed under 200 MPa followed by sintered at 1200°C for 4 h in air to form dense PNO-SDC bars ($5 \times 5 \times 60 \text{ mm}^3$). A pure PNO bar were also prepared for comparison. The obtained bar samples for conductivity testing possess an average relative density about 96% as determined with Archimedes method.

Cell fabrication

Electrolyte-supported symmetric cells for impedance measurements were prepared by screen printing. The thickness of

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