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Short communication

Preparation of $Pr_{0.35}Nd_{0.35}Sr_{0.3}MnO_{3-\delta}$/YSZ composite cathode powders for tubular solid oxide fuel cells by microwave-induced monomer gelation and gel combustion synthesis process

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Abstract

A microwave-induced monomer gelation and gel combustion synthesis process was successfully developed to synthesize well-dispersed $Pr_{0.35}Nd_{0.35}Sr_{0.3}MnO_{3-\delta}$ (PNSM)/YSZ composite cathode powders for tubular solid oxide fuel cells (SOFCs). The thermo-gravimetric (TG) analysis of as-prepared ash indicated the decomposition process of most of metal nitrates during gel combustion. The X-ray diffraction (XRD) pattern of the powders calcined at 1000 °C showed only pure PNSM and YSZ phase. Transmission electron microscopy (TEM) revealed that the morphology of powders was characterized with the YSZ particles enwrapped by fine PNSM particles so that PNSM/YSZ composite powders were much better-dispersed compared with the powders made simply by mechanical mixing process. The cell made from PNSM/YSZ composite powder showed lower cathode ohmic resistance and polarization resistance, and produced higher power density subsequently.

Keywords: Microwave-induced gelation; Combustion synthesis; $Pr_{0.35}Nd_{0.35}Sr_{0.3}MnO_{3-\delta}$/YSZ; Tubular solid oxide fuel cell

1. Introduction

The performance improvement of solid oxide fuel cells (SOFCs) has attracted much research work mainly devoted to designing and synthesizing cell materials. Compared with mature anode and electrolyte materials, cathode materials have been studied more actively. In order to avoid reaction between the La in $La_{0.7}Sr_{0.3}MnO_3$ or $La_{0.6}Sr_{0.4}CoO_3$ cathode and the Zr in YSZ electrolyte, La was substituted by Pr [1] and Nd [2], respectively. In this study, we used $Pr_{0.35}Nd_{0.35}Sr_{0.3}MnO_{3-\delta}$ as cathode material, and the characterization of it would be reported in detail in another paper. The cathode layers of anode-supported cells should exhibit high electro-catalysis and have good contact with electrolyte films. So the cathode/electrolyte composite powders are usually used to fabricate cathode layers.

A wide variety of methods have been used to synthesize cathode powders for laboratory use, such as the solid-state reaction method [2], the glycine-nitrate process [3], the citrate method [4], the combined citrate-EDTA method [5], etc. Gel-casting process has also been demonstrated as a good choice to fabricate materials for SOFCs, including anode [6], electrolyte [7,8], and cathode [9]. Among these methods, combustion synthesis is more suitable for high activity cathode powders. The powders made by combustion synthesis have low depositing density and large contacting area between particles, which would increase the porosity and electrical conductivity of cathode layers, respectively. However, the powders made by gel-casting process aggregate to some degree and are required to be ground carefully.

Composite cathode powders are usually made by the mechanical mixing of cathode and electrolyte powders [10,11]. In this study, we developed a microwave-induced monomer gelation and gel combustion process to synthesize PNSM/YSZ composite cathode powders. For comparison, we also fabricated composite cathode powders by mechanical mixing. The TG

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analysis, phase, morphology, and electrochemical properties in cell of as-prepared powders were characterized.

2. Experimental

2.1. Synthesis of PNSM/YSZ powders

First, stoichiometric Pr_6O_{11}, Nd_2O_3, $SrCO_3$, and $MnCO_3$(all in 99.9%, Sinopharm Chemical) were dissolved in calculated amount of nitrate, and formed a cation solution. Organic monomer (acrylamide, AM) and cross-linker (*N*,*N*-methylenebisacrylamide, MBAM) were dissolved in deionized water to prepare a premix solution. Secondly, commercial YSZ powder, the cation solution, and the premix solution were mixed and ball-milled for 10 h, and a suspension was obtained. Then, the beaker containing the suspension was heated by a microwave oven (MA-2270EGC, 700W) for 10 min, and the suspension was gelated and combusted subsequently. Lastly, the as-prepared ash was calcined at temperatures ranging from 700 to 1000 °C for 2 h, and formed the PNSM/YSZ composite powders. For comparison, pure PNSM powders were also synthesized by the same process and mixed with commercial YSZ powder by ball-milling in ethanol. After being dried at 80 °C, the PNSM + YSZ powders were obtained.

2.2. Characterization of powders

The thermal, structural and morphological properties of as-prepared powders were characterized by several techniques.

Fig. 1. Picture of the as-prepared cell.

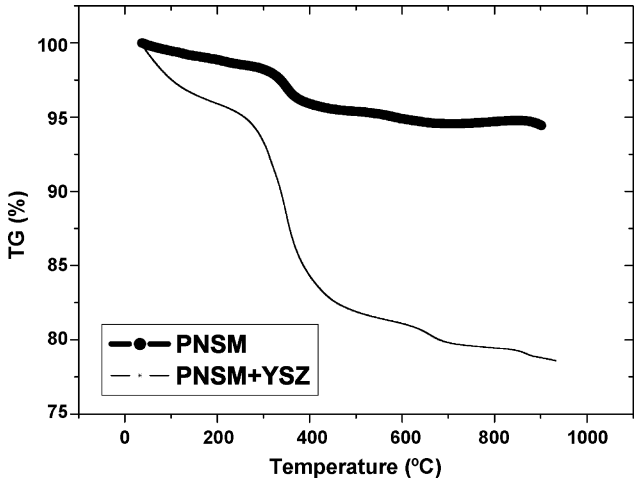

Fig. 2. TG curves of as-prepared PNSM and PNSM/YSZ ash.

The TG analysis of the powders was carried out with Perkin-Elmer Diamond TG from room temperature to 950 °C at a heating rate of 10 °C min^{-1}. The XRD of the PNSM/YSZ powders, as well as pure PNSM and pure YSZ, was carried out on a Philips X'Pert Pro Super Diffractometer with Cu Kα radiation (λ = 0.15418 nm) for phase analysis. The particle size and morphology of powders were observed by TEM (JEOL-2010).

2.3. Preparation and test of cells

The process of fabricating tubular NiO/YSZ anodes with dense YSZ films was described in previous study [12]. The PNSM/YSZ composite powder and the PNSM + YSZ powder were deposited on YSZ electrolyte films by slurry spraying, and sintered at 1100 °C for 2 h to form cells. The cells made with PNSM/YSZ composite powder and PNSM + YSZ powder were named cell A and B, respectively. Fig. 1 shows the appearance of as-prepared cell. Electrochemistry impedance spectroscopy (Chi604a, Shanghai Chenhua) was performed on the cells under

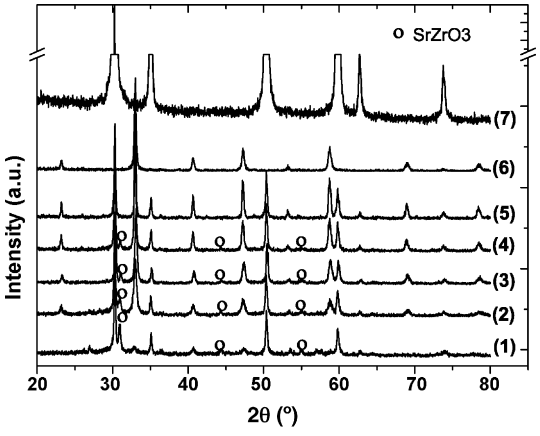

Fig. 3. XRD patterns of as-prepared powders and commercial YSZ: (1) the PNSM/YSZ powders without being calcined; calcined at: (2) 700 °C, (3) 800 °C, (4) 900 °C and (5) 1000 °C; (6) pure PNSM calcined at 800 °C; (7) commercial YSZ.