

Short communication

NiO/YSZ, anode-supported, thin-electrolyte, solid oxide fuel cells fabricated by gel casting

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Received 6 January 2007; accepted 19 March 2007

Available online 20 April 2007

Abstract

A simple and cost-effective gel-casting technique is developed and optimized to fabricate NiO/stabilized yttria–zirconia (YSZ) anode-supported solid oxide fuel cells (SOFCs). The effect of ammonium poly-(methacrylate) (PMAA) dispersant and pH on the zeta potential of YSZ and NiO particles and the viscosity of the NiO/YSZ slurries is studied in detail. The results show that the absolute zeta potential of YSZ and NiO particles reaches a maximum value at pH value ~ 10 and the viscosity of the NiO/YSZ slurry is lowest when the PMAA dispersant content is 1.5 wt.%. A gel-cast NiO/YSZ anode-supported button cell with a spin-coated, thin, YSZ electrolyte film ($\sim 9 \mu\text{m}$) and a $\text{La}_{0.72}\text{Sr}_{0.18}\text{MnO}_{3-\delta}$ (LSM)/YSZ composite cathode gives a peak power output of 1.07 and 0.65 W cm^{-2} at 900 and 800 °C under humidified hydrogen and air. The effect of a graphite pore-former in the gelation and microstructure of NiO/YSZ anode substrates is investigated.

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Keywords: Solid oxide fuel cells; NiO/stabilized yttria–zirconia (YSZ) anode substrates; Gel casting; Optimization; Peak power; Graphite pore-former

1. Introduction

Solid oxide fuel cells (SOFCs) have attracted considerable attention as efficient power-generating systems with very low greenhouse gas emissions and high fuel flexibility as compared with other fuel cell systems [1–3]. Currently, the commonly used materials are fully stabilized yttria–zirconia (YSZ) as the electrolyte, Ni/YSZ as the anode, doped LaMnO_3 as the cathode, and high temperature oxidation resistance alloys as the interconnect. Major technical problems facing SOFC developers include durability and thermal cycleability, corrosion of the metallic interconnect, and performance degradation. Most of these problems can be substantially reduced by lowering the operating temperature of the SOFC from 950–1000 °C to 600–800 °C.

One of the main challenges in lowering the operating temperature is the increase in the cell resistance and, consequently, losses in cell efficiency due to the rapid increase in the bulk resistivity of the electrolyte and in the electrode polarization

resistance at the electrode|electrolyte interfaces [4–7]. As the resistivity loss of the electrolyte is one of the dominant factors affecting the performance of SOFCs, extensive efforts have been made to develop an anode-supported cell structure and to reduce substantially the thickness of the electrolyte to several tens of microns [6,8–15]. There are various techniques to fabricate anode substrates that include tape-casting [16,17], die-pressing [6,9–15] and coat-mixing processes [18]. Gel casting is a novel method in fabricating complex three-dimensional ceramic parts and offers a significant advantage in terms of better homogeneity and thus higher flexural strength in comparison with the conventional forming methods [19–21]. In this process, a slurry with a high solid loading is obtained by dispersing the ceramic powders in a pre-mixed monomer and cross-linking solution. With heating or addition of a catalyst, cross-linking polymerization occurs to form a three-dimensional network structure, and the slurry is solidified in situ, to form ceramic objects of the desired shape. Recently, gel casting has gained attention as a new method in the preparation of electrode powders [22–24] and electrode-supported substrates for SOFCs [25].

This paper presents the development of NiO/YSZ anode-supported SOFCs prepared by the gel-casting method. The effect

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of ammonium poly-(methacrylate) (PMAA) dispersant on the zeta potential of YSZ and NiO particles and the viscosity of NiO/YSZ slurry is examined. The results show that gel casting is an effective method to prepare high-performance NiO/YSZ, anode-supported, thin, YSZ electrolyte cells.

2. Experimental

2.1. Preparation of NiO and YSZ powder suspension and slurry

NiO (J.T. Baker, USA) and 8 mol% Y_2O_3 -zirconia (YSZ, TOSOH, Japan) were used in this study. Before preparation of NiO/YSZ anode substrates by gel casting, the effect of dispersant and pH on the zeta potential of the NiO and YSZ particles and viscosity of NiO/YSZ slurry were investigated. A NiO suspension was prepared by mixing 0.34 g NiO in distilled water with and without the addition of ammonium poly-methacrylate dispersant (PMAA, SD-03, Jiangsu Huagui Hi-Tech Ceramics Co., China). The concentration of PMAA was 0.6 wt.% based on the solid content of NiO. A YSZ suspension was prepared in a similar manner. The total solid content in the suspension was 0.05 vol.%. The pH of the suspension was optimized by adding dilute HCl or NaOH solution. The zeta potential of NiO and YSZ particles was measured by a zeta potential analyzer (Brookhaven Instruments Corporation).

NiO powder calcined at 700 °C in air for 2 h was mixed with YSZ powder (NiO:YSZ = 65:35 by weight) in distilled water and ball-milled for 24 h. Graphite was added as a pore-former and the content was in the range of 5–15 wt.%. In the case of graphite pore-former, polyvinyl pyrrolidone (PVP, Sigma–Aldrich) was added to the slurry to promote the gelation process. The pre-mixed monomer solution was prepared from mono-functional acrylamide (AM, Sigma–Aldrich) and di-functional *N,N'*-Methylenebis-acrylamide (MBAM, Sigma–Aldrich); PMAA was added as dispersant. The NiO/YSZ/graphite mixture was then added to the pre-mixed monomer solution, followed by a planetary ball-milling for 30 min. The pH of the slurry was adjusted during the ball-milling process. The viscosity of NiO/YSZ slurry was measured by a rotary viscometer (NDJ-1 Shanghai, China).

2.2. Anode substrate and cell fabrication

Ammonium persulfate (APS, Sigma–Aldrich) and *N,N,N',N'*-tetra methyl-ethylene diamide (TEMED, Sigma–Aldrich) were added to the NiO/YSZ slurry as an initiator and a catalyst, respectively. The slurry was poured into a mould, and subsequently heated in an oven at 80 °C for 1 h. The gel was cut into discs, followed by drying at 60 °C for 24 h and at 80 °C for another 24 h. The as-prepared green anode-substrates were stored ready for use. A flow chart of the gel-casting process is given in Fig. 1.

The YSZ electrolyte thin film was prepared by a spin-coating method. A suspension of 10 wt.% YSZ in *iso*-propanol was spin coated on to the anode substrate at 1200 rpm for 15 s. The coating was dried at room temperature and the spin-coating process was repeated for several times. The YSZ-coated NiO/YSZ substrates

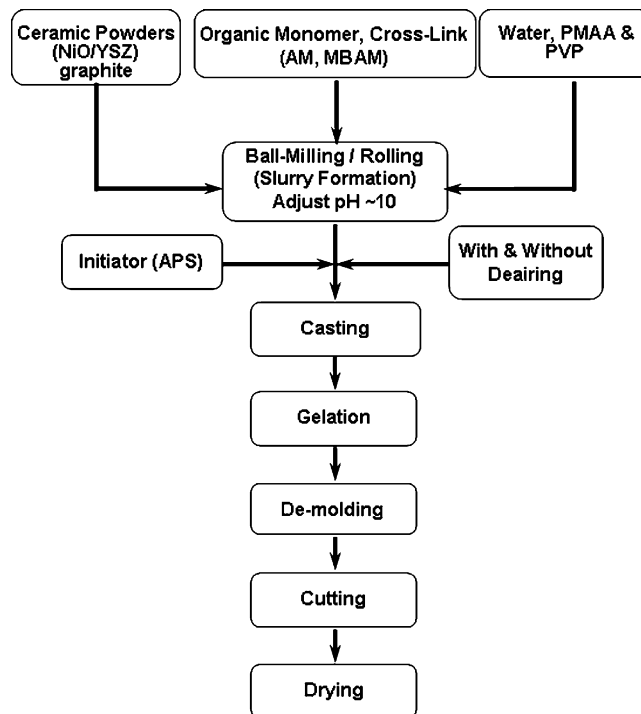


Fig. 1. Flow chart of gel-casting process for NiO/YSZ anode-substrates.

were co-sintered at 1350 °C for 4 h to form a bilayer structure with a porous anode substrate and a dense electrolyte thin film. The anode substrate was ~19 mm in diameter and ~1.2 mm in thickness.

$La_{0.72}Sr_{0.18}MnO_{3-\delta}$ (LSM) cathode was synthesized by a water-based gel-casting process [26]. A LSM/YSZ composite cathode (LSM:YSZ = 60:40, w:w) was applied to the NiO/YSZ/YSZ bi-layer substrate by means of a slurry painting method. A pure LSM layer was applied to the surface of the LSM/YSZ composite to serve as a current-collector. The cathode area was 0.5 cm². The LSM/YSZ composites cathode was sintered in air at 1150 °C for 2 h.

2.3. Cell testing

The cell performance was evaluated with a built-in-house test station. The cell was sealed between two alumina tubes with a ceramic paste. During the test, hydrogen humidified at room temperature (3% H₂O/97% H₂) was fed to the anode chamber at a flow rate of 100 sccm, while the cathode was exposed to an air flow rate of 100 sccm. Electrochemical measurements were performed by means of a Solartron 1260 frequency response analyzer in conjunction with a 1287 electrochemical interface. The overall cell impedance was measured in the frequency range of 100 kHz to 0.01 Hz with a signal amplitude of 10 mV at open-circuit over a temperature range of 600–900 °C. Newly prepared cells were activated under a constant current of 200 mA cm⁻² at 800 °C for 4 h before measurement [9]. The microstructure of the cell after the test was examined by scanning electron microscopy (SEM, Leica 360).

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