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Preparation of YSZ film by EPD and its application in SOFCs

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Abstract

An electrophoretic deposition (EPD) method was applied for the preparation of yttria-stabilized zirconia (YSZ) thin film on a NiO–YSZ porous anode substrate for solid oxide fuel cell (SOFC) applications. Dense YSZ film with uniform thickness can be readily prepared with the EPD method using isopropanol as solvent and iodine as dispersant. As EPD electrode, the NiO–YSZ substrate was covered with a graphite layer to obtain enough electric conductivity. A series of experiments, such as suspension concentration, sintering shrinkage, and so on, were conducted in order to optimize the EPD technique. A planar-type SOFC was fabricated using $La_{0.4}Sr_{0.6}Co_{0.2}Fe_{0.8}O_3$ (LSCF) as the cathode and YSZ film deposited onto the NiO–YSZ anode substrate as the electrolyte. The fuel cell exhibited an open circuit voltage of about 1.0 V and a maximum power density of 440 mW/cm^2 at 900 °C. Thus, the EPD method was a suitable technology for the formation of gas-tight YSZ film.

Keywords: Electrophoretic deposition (EPD); YSZ films; SOFC

1. Introduction

Recently the research of SOFC operating at intermediate temperature becomes more popular [1–5]. Two effective methods can be used to reduce the operating temperature of SOFC from conventional range of 800–1000 °C down to intermediate temperature (500–800 °C). One is to use alternative electrolyte materials with higher ionic conductivity at lower temperature [6], and the other is to use the thin film technology of conventional electrolyte—yttrium-stabilized zirconia (YSZ). The latter is often adopted.

There are many techniques applied to produce YSZ films. In these techniques, wet processes are especially attractive because of the low cost and mass productivity [7]. Electrophoretic deposition (EPD) is a noble wet process. The EPD technique was firstly proposed by Ishihara et al. for fabrication of SOFCs [8]. In EPD process, charged particles dispersed in a stable suspension are driven by a dc electric field to move towards an oppositely charged electrode, upon which they ultimately deposit and build up a closely packed particulate layer [9]. The advantages of EPD for SOFC are listed:

(1) EPD makes it possible to fabricate the SOFC cells with any types of shapes;
(2) Both dense and porous films can be prepared by regulating deposition rates and subsequent sintering processes;
(3) It is easy to stack electrode and electrolyte layers;
(4) Co-deposition is possible to prepare composite layers such as Ni-YSZ anodes, etc.

In this paper, EPD technique was used to prepare YSZ (yttria-stabilized zirconia) film on porous anode substrate (NiO/YSZ). In this process, isopropanol was used as solvent and iodine as dispersant in YSZ suspension. The zeta potential of YSZ particles was studied by changing the amount of iodine. The effect of the amount of iodine and the concentration of YSZ suspension on the deposition weight was also studied. The prepared YSZ film and the anode substrate were then co-sintered at 1400 °C. The performance of a single SOFC, for which the YSZ film prepared by EPD process was applied, was tested. The short

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circuit current density and the maximum power density were 2.08 A/cm^2 and 440 mW/cm^2 at 900 °C, respectively.

2. Experimental

The anode materials (NiO–YSZ) were prepared by mixing YSZ powder (China building material academy, Beijing, China), NiO powder (made by ourselves) and amylums (commercial) by a weight ratio of 5:5:1. The purpose of adding amylums was to form more pores in anode. The mixture was milled in agate mortar for 1 h. Then, the mixed powders were pressed into pellets with a diameter of about 13 mm and a thickness of about 0.6 mm. The pellets were subsequently baked at 900 °C for 2 h to form the porous NiO–YSZ as substrate.

Generally, the EPD substrate should have some electronic conductivity at room temperature. However, the conductivity of NiO–YSZ is too low to be used as electrode of EPD, in this paper, a graphite film was prepared on NiO–YSZ pellets as conductive reagent, and it would be burnt out during the subsequent calcining process.

YSZ suspension was obtained by dispersing YSZ powders (TZ-8Y, Tosoh Corporation, Tokyo, Japan) and iodine into isopropanol. In a typical deposition, 0.8 g YSZ and 0.04 g I_2 were added to a 100 ml beaker with 80 ml isopropanol. The mixture was sonicated with a high intensity ultrasonic probe for 10 min to form a stable suspension.

The zeta potential of YSZ suspension was measured using a micro-electrophoretic instrument (JS94G$^+$). The experimental arrangement for EPD is schematically illustrated in Fig. 1. NiO/YSZ substrate with a layer of graphite connected with the negative output of EPD power source was hung in the deposition cell as the cathode, while a graphite disk was placed on the opposite side of the cell acting as the anode. The two electrodes were kept parallel with a distance of 6 mm. With a constant deposition voltage of 10 V, positively charged YSZ particles were electrophoretically deposited onto the cathode and YSZ film was formed in 1 h. The green YSZ film was dried in open air for about 12 h and then co-sintered with the substrate at 1400 °C for 2 h.

The sintering curves of pressed bars with powders of YSZ electrolyte and NiO–YSZ anode were measured utilizing a Netzsch DIL 402C/3/G dilatometer (Netzsch GmbH, Germany) with air purging (flow rate: 50 ml/min). In this measurement, the temperature was raised from room temperature to 500 °C at a rate of 5 K/min, and from 500 to 900 °C at 10 K/min, then being held at 900 °C for 2 h, finally raised the temperature to 1300 °C at 10 K/min and being held at 1300 °C for 2 h. The surface and cross-section of the sintered YSZ film was characterized using a SEM (Hitachi S-570 scanning electron microscope).

$La_{0.4}Sr_{0.6}Co_{0.2}Fe_{0.8}O_3$ (LSCF) powder was synthesized by solid-state reaction method. Some adhesives were mixed with the powders to form slurry. The slurry was applied on the exposed surface of the YSZ film and then fired at 300 °C for 30 min to burn out the adhesives. Finally the sample was sintered at 900 °C for 5 h to obtain a positive-electrolyte-negative (PEN) component (NiO–YSZ/YSZ/LSCF).

To make a single fuel cell, a PEN component was sealed onto one end of ceramic tubes (Fig. 2) as the method mentioned in Ref. [10]. The silver layer on both electrode surfaces made from silver paste (DAD-87, Shanghai Research Institute of Synthetic Resin, Shanghai, China) was used as current collectors to collect current on both anode and cathode. Thus, a single fuel cell was prepared

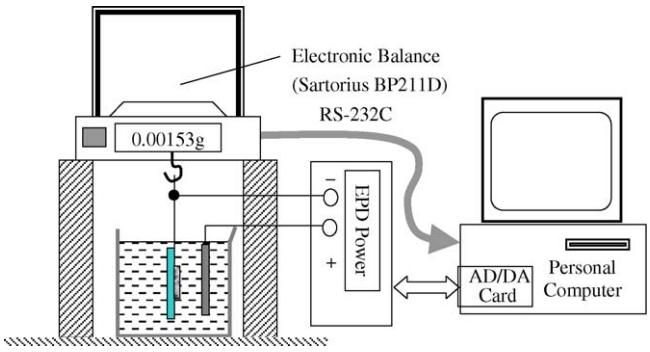

Fig. 1. Schematic diagram showing the cell configuration for EPD.

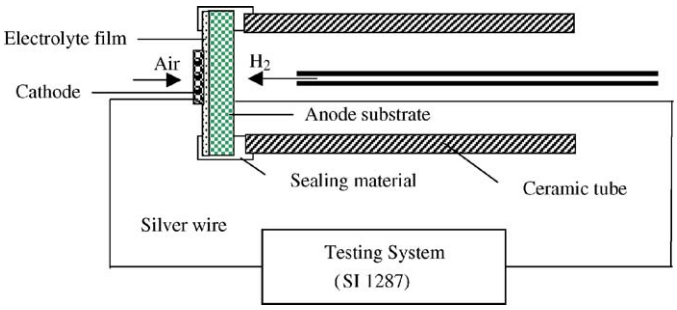

Fig. 2. Assembly and testing system of single cell.

and to be tested. In the process of fuel cell testing, the hydrogen was used as fuel and the oxygen in air was used as oxidant. The anode chamber was purged with nitrogen below 500 °C, and the nitrogen was changed to the fuel gas (H_2) at 500 °C. The performances of the single fuel cell were measured using Solartron SI 1287 electrochemical interface (Solartron Instruments, Hampshire, England).

3. Results and discussion

3.1. Electrophoretic deposition of YSZ

The thickness of prepared YSZ film was lie on the deposited weight of YSZ in EPD process. In the initial period of EPD process, the weight (w) of charged particles deposited per unit area of electrode substrate is expressed as the following equation [8], ignoring the charge carried by the free ions:

$$w = \tfrac{2}{3} C\varepsilon_0\varepsilon_r\zeta\eta^{-1}EL^{-1}t \quad (1)$$

where C represents the concentration of the suspension, ε_0 the permittivity of vacuum, ε_r the relative permittivity of the solvent, ζ the zeta potential of the particles in suspension, η the viscosity of the solvent, E the applied potential, L the distance between electrodes, and t is the deposition period. Eq. (1) suggests that zeta potential of the particles is an important parameter in EPD process. The value of zeta potential is related with the solvent and the concentration of the dispersant [11]. In this study, isopropanol was used as solvent and the iodine was used as dispersant. The zeta potential of YSZ particles in isopropanol was plotted against the amount of dispersant (iodine) in Fig. 3.

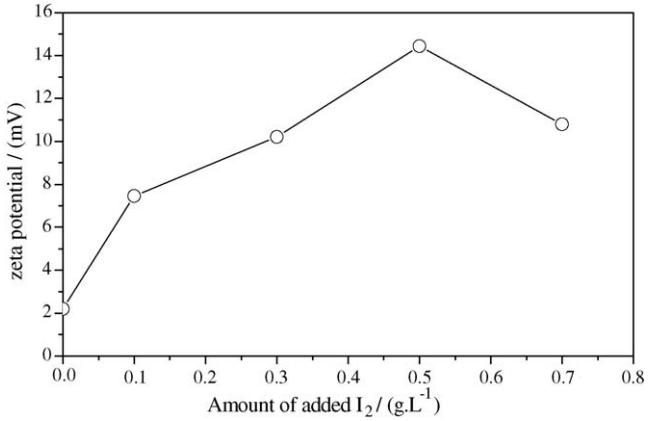

Fig. 3. Zeta potential of YSZ particle as a function of the I_2 concentration.