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Short Communication

Enhancing solid oxide fuel cell performance by electrode reactivation through wet chemical treatments

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ARTICLE INFO

Article history:

Received 28 February 2016

Received in revised form

8 June 2016

Accepted 12 June 2016

Available online 27 June 2016

Keywords:

Solid oxide fuel cell

Solution chemical treatment

Electrode reactivation

Surface chemistry/morphology

ABSTRACT

The high-temperature sintering usually has adverse effects on surface chemistry/morphology of solid oxide fuel cell electrodes, hiding the intrinsic electrocatalytic activity of the materials used. Here we demonstrate the universal reactivation of the sintered electrodes using solution chemical treatments, resulting in a remarkable improvement in electrode performance. For example, the area specific resistance values were reduced by 73.0% and 29.4% for the HCl-treated strontium-doped LaMnO₃ (LSM) cathode and NaOH-treated nickel cermet anode, respectively. A 33% increase in peak power density of the cell with activated electrodes shows that activation treatment has great potential to become a strategically important process during manufacturing.

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Introduction

Solid oxide fuel cells (SOFCs) typically consisting of an ionic-conducting ceramic and two porous electrodes are electrochemical devices that directly convert chemical energy into electricity without combustion and mechanical processes [1]. The electrodes play a crucial role in the performance of the SOFCs since they provide the interface between the chemical energy associated with fuel oxidation and electrical power. The primary electrode reactions, including the oxidation of the fuel and the reduction of the oxidant, occur mainly on the surface of the electrode catalysts [2,3]. Therefore, the

electrodes with a high surface catalytic activity are of great importance to the SOFCs. Over the past decades, significant efforts have been devoted to enhancing the electrode performance, especially in the search for more efficient electrode catalyst materials. Perovskite-type Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-δ} (BSCF) and layered double perovskite LnBaCo₂O_{5+δ} (Ln: rare earth) are effective as cathode catalysts using ceria or lanthanum gallate based electrolytes [4,5]. Layered perovskite PrBaMn₂O_{5+δ} and LaCrO₃-based anodes could be alternative anode materials with high tolerance to coking and poisoning [6,7]. Another approach for enhancing the electrode performance is to design novel electrode architectures. Nano-architected electrodes prepared from ceramic

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<http://dx.doi.org/10.1016/j.ijhydene.2016.06.124>

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nanoparticles or nanofibers have large surface-to-volume ratio and enhanced surface electrocatalytic activity [8,9]. The electrode performance also can be enhanced by surface modification through a solution-based infiltration process. The discrete catalyst nanoparticles or continuous catalyst nanofilms added to the internal surfaces of the porous electrodes enhance the kinetics of the electrode processes [10,11].

At present, the high-temperature treatment steps (typically at 900–1300 °C) are still required to achieve a reasonable bonding between the electrode and electrolyte layers, and electrode particles for the charge transport in the mainstream manufacturing processes of the electrodes [12]. Such high temperature has two major adverse effects on the microstructure and performance of the electrode. First, the sintering and grain growth of the primary particles lead to a decrease in the electrode surface area [13]. Second, the cations and impurities may segregate to the surface and interfaces, blocking the surface active catalytic sites [14,15]. Therefore, it is difficult to retain the intrinsic properties of the selected material and the electrode performance may be much lower than expected. The reactivation phenomenon of the sintered electrode was first observed on LSM-based cathode after applying cathodic current/polarization potentials [16]. Based on a comparative study on the electrodes with and without dilute acid etching, Jiang et al. suggested that the surface-segregated SrO has a significant inhibiting effect on the surface exchange process of the electrode reactions and attributed the activation effect to the incorporation of SrO into LSM lattice [17]. The similar acid/alkali solution etching experiments have also been performed to investigate the effect of surface Sr segregation on the cathode performance by other researches [18,19].

Herein, we present the reactivation of the sintered SOFC electrodes and the improvement of cell performance by wet chemical treating the electrodes. The possible generality of this approach was examined through investigating the performance of several typical SOFC electrodes treated with different solutions.

Experimental

The $\text{La}_{0.75}\text{Sr}_{0.25}\text{Cr}_{0.5}\text{Mn}_{0.5}\text{O}_{3-\delta}$ (LSCM) has been prepared by solid-state reaction using La_2O_3 , SrCO_3 , Cr_2O_3 and Mn_2O_3 . The stoichiometric quantities of reactants were ball-milled with ethanol for 8 h, and then calcined in air at 1400 °C for 24 h. Commercial powders of YSZ, NiO, and LSM were also selected as electrode materials. The symmetric cells used for electrode polarization resistance measurements. The YSZ substrates with a thickness of ~1 mm were prepared by die-pressing YSZ powder (TERIO Co., China) and sintering at 1500 °C for 2 h in air. To prepare the electrode, the electrode powders were ball-milled with a terpeneol-based organic vehicle to form electrode paste. The paste was then screen-printed onto both sides of the YSZ substrate, followed by sintering typically at 1000–1300 °C. The sintered symmetric cells were dipped into various aqueous solutions for electrode treatments, thoroughly washed by de-ionized water and then dried at 100 °C

for 2 h. Details of the electrodes and treating parameters are shown in Table S1. The electrolyte-supported single cells were adopted for the fuel cell test. The YSZ electrolyte was 20 mm in diameter and 0.2 mm in thickness. The anode NiO-YSZ slurry was screen-printed onto one side of the electrolyte and sintered at 1300 °C for 2 h. Then, the cathode LSM-YSZ ink was screen-printed onto another side and fired at 1100 °C for 2 h. The single cells also were dipped into aqueous solutions for electrode treatments.

The electrode polarization resistance of the symmetric cells was investigated by Electrochemical Impedance Spectra (EIS) measurement in a two-electrode configuration under symmetric atmospheres (wet H_2 for anode and air for cathode) using CHI 604e electrochemical workstation. EIS data were recorded in the frequency range of 100 KHz to 0.1 Hz (or 0.01 Hz) using AC signal of 10 mV under open circuit voltage (OCV) condition. The morphology and elemental composition of the electrode particle surfaces were examined by a field-emission scanning electron microscope (Ultra55 FE-SEM, ZEISS) equipped with energy-dispersive X-ray spectrometer (EDS, Oxford). The single cell was tested using 50 mL min^{-1} humidified hydrogen as the fuel and ambient static air as the oxidant. The current–voltage curves of the cells were monitored using a Keithley 2420 digital source meter. The impedance spectra of fuel cell were recorded in the frequency range from 100 KHz to 0.1 Hz with an AC amplitude of 10 mV at open circuit.

Results and discussion

The area specific resistances (ASRs) of several typical electrodes obtained from the impedance spectra on the symmetric cells at 800 °C are tabulated in Table 1. As can be seen, although the wet chemical treatment parameters, such as composition and concentration of the solution and treatment time, have not been systematically optimized, the ASR values for most of the treated electrodes are generally on a downward trend. The catalytic activity of these treated electrodes is obviously enhanced. For each type electrode, there are one or more solutions which can be used to treat the electrode for enhancing the electrode performance. Therefore, the improvement of the catalytic property may be a generally existent phenomenon for the sintered SOFC electrodes treated with chemical solutions. It should also be noted that an inappropriate treatment may result in an increase of the electrode ASR. For example, the ASR for LSCM anode increases by 53.1% after treating with H_2O_2 . The concentration of the solution and treatment time may play crucial roles in the electrode reactivation. Usually, it takes a long time to achieve a satisfactory treating result with low concentration solutions. Increasing the solution concentration is able to shorten the required treating time. However, lowering the concentration of the treating solution can improve the controllability of the treatment process, resulting in a better treatment effect. Further study is needed to optimize the wet chemical treatment parameters, such as composition and concentration of the solution and treatment time.

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