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Electrochemical performance and carbon resistance comparison between Sn, Cu, Ag, and Rh-doped Ni/ScCeSZ anode SOFCs operated by biogas

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Abstract

Ni/ScCeSZ anode-supported SOFCs were fabricated via the reverse tape casting method and an LSCF ($La_{0.6}Sr_{0.4}Cr_{0.2}Fe_{0.8}O_3$) cathode was applied. In order to prevent the chemical reaction of zirconia from the electrolyte and strontium from the cathode, a thin barrier layer of $Gd_{0.1}Ce_{0.9}O_{1.95}$ (GDC) was painted on the ScCeSZ electrolyte. On the anode side, Sn, Cu, Ag, and Rh were doped into the Ni-based cermet by infiltration. Metal nitrate hydrates were used as precursors for the dopants. The prepared cells were tested in hydrogen (H_2:N_2=3:1) and then simulated biogas (CH_4:CO_2:N_2=2:1:1) at intermediate operating temperature (600 to 750 °C). The tested cells used ambient air as the oxidant for the electrochemical reactions. The obtained cells were characterised by, scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDS), X-ray powder diffraction (XRD), and X-ray diffraction (XRD). The electrochemical performance of the prepared cells was characterised by OCV, I-V, operational stability (potentiostatic and galvanostatic), and electrochemical impedance spectroscopy (EIS).

According to our previous work, the carbon accumulation on the anode surface was successfully suppressed by infiltrating Sn, Ag, or Cu into the Ni/YSZ anode-supported cells under biogas operation at 750 °C. The addition of a small amount of Sn or Ag to the Ni/YSZ anodes can greatly improve the electrochemical performance and the operational stability of the cells operated on biogas. Due to the low melting point of Cu, the performance of Cu-Ni/YSZ anodes did not drastically improve. Hence, the Cu-doped Ni/ScCeSZ cells with LSCF cathode are expected to obtain better electrochemical results at slightly lower operating temperature.

1. Introduction

Solid oxide fuel cells (SOFCs) are considered as one of the most promising energy converting electrochemical devices because of their high efficiency of generating electricity from chemical energy. Flexible fuel choice is one of the main advantage of SOFCs because of their high operating temperature. Hence, hydrocarbons can be used as potential fuel for SOFC operation [1]. Ni cermet is the most commonly used anode material for SOFC because of its high catalytic activity towards hydrocarbon reforming. Ni is not only a good catalyst for hydrocarbon-fueled SOFCs, it also promotes carbon forming reactions [2]. Catalyst deactivation can be caused by carbon accumulation, which drastically reduce the electrochemical performance. Furthermore, carbon accumulation can even result in irreversible structural damage of the cell [3, 4]. The main carbon forming reactions in a hydrocarbon-fuelled SOFC are show as follow.

$$2CO \rightarrow CO_2 + C \quad (1)$$

$$CO + H_2 \rightarrow H_2O + C \quad (2)$$

$$C_xH_y \rightarrow xC + y/2H_2 \quad (3)$$

Eq.1 is known as carbon monoxide disproportionation occurring at low temperature (<700 °C) to produced carbon dioxide and carbon, also known as Boudouard reaction [5]. Eq.2 is the reverse reaction of 'water-gas shift' reaction, which also tends to occur at low temperatures. Hydrocarbon dissociation (Eq.3) takes place at high temperatures, which converts hydrocarbon into carbon and hydrogen. [6, 7] Typically, adding external steam or CO_2 and modifying Ni-based anode materials have been extensively investigated to alleviate carbon deposition on the anode. Nevertheless, the existence of excessive amount of H_2O or CO_2 in the fuel dilutes the fuel, resulting in reduced cell performance [2]. Therefore, doping Ni-based anode with metal catalyst or metal oxide can improve cell performance and carbon resistance without fuel dilution [6].

Many studies have reported that noble metal and/or transition metal doping can enhance electrochemical performance and/or reduce carbon deposition of SOFCs operated with hydrocarbons, such as Au [8, 9], Ag [10, 11], Rh [12], Sn [13-15], and Fe [16]. Present work provides a comparison of electrochemical performance and carbon resistance of Sn, Cu, Ag, Rh-doped Ni/ScCeSZ anode SOFCs operated with simulated biogas.

2. Experimental

Cell fabrication – Ni/ScCeSZ anode-supported SOFC button cells used in this study were fabricated using an aqueous tape casting method. Scandia/ceria-stabilised zirconia (10Sc1CeSZ, DAIICHI KIGENSO KAGAKU KOGYO CO), NiO (Green Nickel Oxide, Hart Materials), LSCF ($La_{0.6}Sr_{0.4}Cr_{0.2}Fe_{0.8}O_3$, Praxair) was used as electrolyte, anode, and cathode material, respectively. A thin barrier layer of Gadolinium-doped ceria (10GDC, Fuel Cell Materials) was applied between the ScCeSZ electrolyte and LSCF cathode to prevent chemical reaction of zirconia and strontium. Details about slurry preparation and cell fabrication process are found in our previous paper [17].

Anode modification – Ni/ScCeSZ anode modification followed the cell fabrication, using Sn, Ag, Cu, and Rh precursors, respectively. The calculated amount of precursor solution was added to the anode surface through a pipette and then dried at ambient temperature. Dopant precursors were further calcined at different temperatures (Table 1) to form metal oxides.

Table 1 – Dopant precursors and their calcination temperatures [10, 18-20].

	Sn	Ag	Cu	Rh
Precursor	$SnCl_2 \cdot 2H_2O$	$AgNO_3$	$Cu(NO_3)_2 \cdot 3H_2O$	$RhCl_3 \cdot xH_2O$
Calcination temperature (°C)	600	500	400	150

Cell testing and characterisation – Prepared cells were tested in a horizontal furnace setup with two operational mode, hydrogen (21 mL min^{-1} H_2 and 7 mL min^{-1} N_2) and biogas (14 mL min^{-1} CH_4, 7 mL min^{-1} CO_2 and 7 mL min^{-1} N_2). Cell testing was conducted at 600 to 750 °C. Silver wire (ScientificWire Company) and silver paste (DAD-87, Shanghai Research Institute of Synthetic Resins) were used as the current collector and sealant, respectively. The SOFC cathode was exposed to the ambient air to provide oxidant for the reaction. Electrochemical characterisations, such as OCV, I-V, potentiostatic, and EIS were carried out by Solartron 1470E and 1455 FRA analysers (Solartron Analytical). For AC impedance measurements, the frequency used was between 0.1 and 10^6 Hz, with a 10 mV signal amplitude. In order to investigate the surface morphology, surface element distribution, and alloy catalyst crystalline structure, SEM, EDS, and XRD is conducted, respectively. The amount of accumulated carbon on the hydrocarbon-tested cell was determined by temperature-programmed oxidation (TPO).

3. Results

In our previous work, adding 1 wt% of Sn, Ag, and Cu to Ni/YSZ anode showed improved SOFC operational stability under biogas at 750 °C. Enhanced peak power density was only obtained from the Sn-doped Ni/YSZ cell showed. Because of the low melting point of copper and silver, doped Cu and Ag might migrate or agglomerate in the anode scaffold during SOFC operation. Therefore, lower operating temperatures were used to investigate the effect of adding Cu and Ag to the Ni/ScCeSZ cells. Result will be presented at the conference.

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