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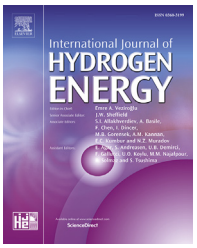

The investigation of Ag & $LaCo_{0.6}Ni_{0.4}O_{3-\delta}$ composites as cathode contact material for intermediate temperature solid oxide fuel cells

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ABSTRACT

The high interface resistance between cathodes and interconnects is a major cause for performance degradation of solid oxide fuel cells (SOFCs). Ag particles were mixed to $LaCo_{0.6}Ni_{0.4}O_{3-\delta}$ (LCN) matrix which prevented the silver densification and demonstrated porosity microstructure. The composites with different Ag content were evaluated as cathode contact materials with SUS430 alloy as interconnects. The area specific resistance (ASR) of SUS430/10%Ag & LCN/SUS430 showed the optimal performance in which the ASR was 73 mΩ cm^2 after 50 h at 750 °C and showed stable property in 10 thermal cycles from 200 °C to 750 °C. The excellent performance of 10%Ag & LCN is attributed to the high conductivity of silver, the stable microstructure of LCN and its good interface adhesion with the interconnect alloy. With 10%Ag & LCN as cathode contact materials, the power density of a single cell reached 0.623 W/cm^2 at 750 °C and the average degradation is lower than 1% in 3 thermal cycles.

Introduction

Solid oxide fuel cells (SOFCs) are attractive electric power generation system with high-energy conversion efficiency which have many advantages such as multi-fuel capability and simplicity of system design [1]. In order to achieve higher voltage, the interconnects are used to connect multiple cells in series to form a stack. The interconnects also separate the fuel on the anode side and the air on the cathode side [2].

The contacts between interconnects and the electrodes, especially with the cathode, play a significant role in the stack output performance [3,4]. As a result, researchers pay much attention to develop new materials and structures with better performance for SOFC interconnect-cathode contacts [5]. The cathode contact materials (CCM) are required to possess the following requirements: (i) high electron conductivity to minimize the resistance of the contact itself, (ii) appropriate sintering activity to ensure high mechanical strength and good bonding with the adjacent components, (iii) appropriate thermal expansion behavior and chemically compatible with neighbours or favorable reactions (iv) high thermochemical and structural stability in the oxidizing environment [6,7].

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The doped ABO_3 perovskites of different compositions (A = La, Sr; B = Ni, Fe, Mn, Co, Cu) have been extensively tested as cathode contact materials [8–10]. These materials usually show electrochemical and structure stability in oxidizing atmosphere, close coefficient of thermal expansion (CTE) and high compatibility with the neighboring materials [11]. However, these perovskites exhibit limited sintering property and thus low bond strength with interconnects and cathodes [12]. Recently developed materials M_3O_4 (M = Ni, Mn, Co, Cu, Fe, or mixture thereof) oxides with a spinel structure faced the same challenge [9]. Some efforts have been made to solve this problem, Michael C. Tucker added inorganic binder to conventional cathode contact materials to improving interface bonding [13] and B.P. McCarthy take alternating flow of air and nitrogen to get enhanced sintering of LSM-10 [6]. Noble metals such as Au, Ag and Pt have also been evaluated as contact materials in which Au showed undesired performance in the cell test and Pt showed financial disadvantage [14]. Ag is one of the most promising contact materials because of its relatively low price, high electrical conductivity, high chemical stability [15]. However, the densification of pure Ag in SOFC operating temperature limits its use as cathode contact layer [16]. Ayhan S found that addition of LSM particles to silver matrix result in more stable porosity at 800 °C for 170 h compared with Ag-YSZ composites [17]. Ag particles were also incorporated into perovskite cathodes such as BSCF, LSCF, LSM through powder mixture or infiltration which prevented the silver densification and led to a better cathode performance [18–20].

$LaCo_{0.6}Ni_{0.4}O_{3-\delta}$ (LCN) was evaluated as the cathode contact materials previously, which present higher electrical conductivity than conventional cathode materials ($La_{1-x}Sr_{x)y}MnO_{3\pm\delta}$ (LSM) and $LaNi_{0.6}Fe_{0.4}O_3$ (LNF) [8,21], close thermal expansion coefficient and compatibility with SUS430 ferritic stainless steel [22]. In order to further improving the performance of LCN perovskite, nano-sliver particles are added to perovskite matrix which are expected to combining the unique attributes from LCN and Ag. In the present study, Ag & LCN composites were test as cathode contact materials under isothermal and cyclic exposures.

Experiment

Polymeric steric entrapment precursor method was chosen to prepare the LCN powder. In the process, hydroxyl in polyvinyl alcohol (PVA) and its long chain molecular structure played an important role [23]. PVA with 1700 polymerization degree and analytically pure (Sinopharm Chemical Reagent Co. Ltd.) was dissolved in distilled water to form a 5% homogeneous mixture at 95 °C in the process the container should be sealed to prevent evaporation. Then $La(NO_3)_3{\cdot}6H_2O$, $Co(NO_3)_2{\cdot}6H_2O$ and $Ni(NO_3)_2{\cdot}6H_2O$ (Shanghai Jingchun Scientifical Co. Ltd) were added in stoichiometric proportion. Such solutions were warmed in water bath to slowly evaporate the solvent up to high viscosity solution with the temperature unchanged. The gel was then transferred to drying oven to develop porous spong-like precursor at 130 °C for 12 h. Subsequently, the precursor was grinded at room temperature and calcined in muffle roaster at 800 °C for 3 h to form the desired LCN powder. Ag powder with 0.3 μm average particle size was purchased from Shanghai Jingchun Scientifical Co. Ltd. X-ray diffractometer (XRD, PANalytical, X'Pert PRO) was employed to identify the crystal structures and field emission scanning electron microscope (FSEM, Sirion 200, FEI Corporation, Holland) was used to examine the LCN and Ag morphology.

Ag and LCN slurry was obtained by hand mixing the powders with terpilenol and ethyecellolose as organic binder using mortar and pestle. The content of Ag was 0, 10%, 20%, 30%. The well-mixed slurry was screen printed on the polished surface of two pieces of interconnect coupons with dimension of 25 × 18 × 1 mm. The coupons were then pressed together under a load of 17 psi to make a sandwiched configuration of SUS430/CCM/SUS430 which was shown in Fig. 1. The thickness of contact layer was about 70 μm. The ASR of the setup was measured by four-probe method which was presented in Fig. 2. Pt mesh and wires were used as current collector and testing leads. The long-term degradation and thermal-cycle stability of the setup are evaluated by heating it from room temperature to 750 °C with a ramping rate of 5 °C/min^{-1} and kept that temperature for 50 h. Then conducting 10 thermal cycles between 200 °C and 750 °C with a heating and cooling rate of 5 °C/min^{-1}. Powder morphology, cross-sectional microstructure and elemental analysis were examined by environment scanning electron microscope (ESEM, Quanta 200, FEI Corporation, Holland) equipped with an energy dispersive spectrometer (EDS).

The contact materials were then evaluated in a single cell stack. The cell and stack configuration are described elsewhere [24,25]. Ni paste and mesh were used as anode current collector and Ag & LCN as cathode current collector. Dry air and pure H_2 were fed to the cathode and anode respectively as the oxidant and fuel. The current-voltage performance under thermal cycle between 200 °C and 750 °C was tested to evaluate the property of the composite contact layer.

Result and discussion

Fig. 3 presents as-prepared powder morphologies of LCN and Ag. The LCN is well necked with each other which is believed to be beneficial to microstructure stabilization and the porous

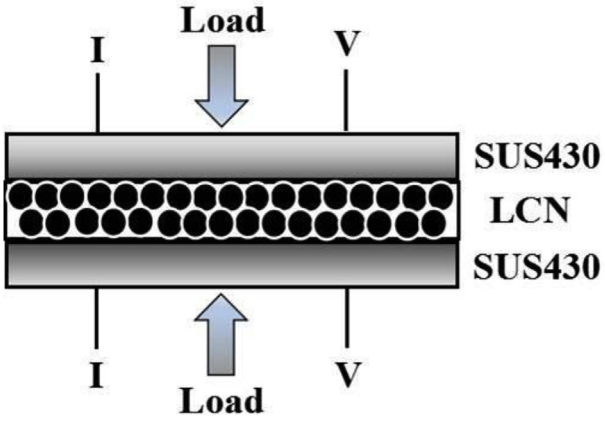

Fig. 1 – The schematic diagram of the specimen for contact ASR measurement of SUS430/LCN/SUSU430 setup.

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