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Chemical compatibility between Sr-doped lanthanum manganite air electrode and AISI 441 interconnect

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ABSTRACT

Chromium poisoning of the air electrode of solid oxide fuel/electrolyzer cells is a critical issue when stainless steel with high Cr content is used as an interconnect material. The mechanism of the diffusion and deposition of the Cr species is not totally clear yet. In this work, an yttria-stabilized zirconia electrolyte (YSZ)/strontium-doped lanthanum manganite electrode (LSM)/AISI 441 alloy interconnect tri-layer structure has been fabricated by screen printing in order to simulate the working environment of a real cell. The samples are thermally treated in moist air atmosphere at 800 °C for up to 500 h. The porous LSM layer thickness has been controlled to investigate the Cr distribution in the porous layer and the diffusion behavior of the Cr species. The microstructure and elemental analysis show that Cr species diffuses into the porous LSM layer and deposits on the LSM surface. The deposition occurs simultaneously with the LSM grain growth.

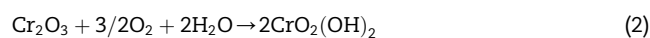
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1. Introduction

Solid oxide fuel cells (SOFCs) and solid oxide electrolyzer cells (SOECs) are electrochemical devices that produce electricity from fuel or hydrogen by splitting water [1,2]. In a SOFC/SOEC stack, an interconnect is placed between the air electrode and the fuel electrode in order to electrically connect single cells [3,4]. According to the planar stack design, the electrical resistance of the electrodes, the electrolyte, the interconnect, and their interfaces should be low enough to minimize the current loss and achieve a high energy conversion efficiency. High oxidation and corrosion resistance in oxidizing atmospheres, suitable coefficient of thermal expansion match with other cell components, good chemical compatibility with other cell materials, and long-term stability are also needed [5,6].

Conductive perovskite ceramics have been considered as air electrode materials, such as Sr-doped lanthanum manganite (LSM) and Sr-doped lanthanum ferrite [7–9]. A variety of

alloy systems have been investigated as interconnect materials, such as Fe–Cr, Fe–Ni, Ni–Cr, and Fe–Ni–Cr alloys. Ferritic stainless steel with high Cr content is widely studied as a more cost effective interconnect material [5,6,10]. The common trait of these systems is that they all form a surface layer of Cr₂O₃, or chromia, under the oxidizing atmosphere, and this layer provides high temperature oxidation resistance. However, chromia evaporates at high temperatures and forms gaseous CrO₃ species. The pressure of the gaseous CrO₃ increases with oxygen partial pressure. The moisture in the oxidizing environment can cause chromia to evaporate as well, in the form of CrO₂(OH)₂. Water content above 0.1% in air results in the partial pressure of CrO₂(OH)₂ to exceed the partial pressure of CrO₃ [11,12]:



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During the SOFC/SOEC operation, interconnect/air electrode interfacial evolution and structure/performance degradation encompass diffusion and reaction as well as the corresponding microstructure changes. The air electrode materials such as LSM show good durability in a single cell [13,14]. However, during the operation with Cr-containing metallic interconnect materials, the volatile Cr species transport to the air electrode and the air electrode/electrolyte interface. The interaction between the porous air electrode and the volatile Cr species is generally called Cr poisoning, which causes major degradation on the air electrode side [15,16]. When the porous LSM air electrode interacts with the interconnect that forms volatile Cr species, the Cr poisoning leads to a dramatic increase of the polarization resistance and a fast drop of the output voltage of the cell in several hundreds of hours [17–22]. In SOFCs, the Cr poisoning is undoubtedly the most serious problem. The atmosphere strongly affects the composition of the volatile Cr species [11,12]. The humidity of the atmosphere in the air electrode of SOFCs can lead to a large drop of the output cell voltage and degradation of the air electrode/electrolyte interface [23]. In SOECs, it is an even bigger problem because the interfacial degradation is 10 times faster than that in SOFCs due to the high moisture condition [24].

The interaction between the Cr-containing species from the interconnect and the air electrode has been investigated. However, there is considerable disagreement on the mechanism of Cr-containing species interaction with and deposition on the air electrode. Some studies suggested that the deposition of Cr species is closely related to the oxygen activity at the electrode/electrolyte interface-triple phase boundaries (TPB) [17,19]. In contrast, others believed that the interfacial degradation by Cr poisoning could be caused by (i) blocking of electrochemically active sites by electrochemical reduction of Cr-containing species, and (ii) decomposition of the air electrode by the formation of Cr-containing mixed oxides, driven by the thermodynamics without any influence of the electrical potentials [25]. There were also claims that the deposition of Cr species depends strongly on the nature of the air electrode materials and that Cr species deposition on the air electrode is most likely controlled by a non-electrochemical process [18,26,27].

As seen from the discussion, a fundamental issue is still in heated debate for the interconnect/air electrode interface: the location of the chemical reactions. The primary reason for the disagreement is that to date there is no detailed knowledge of Cr species transport at the interface, interfacial reactions, and the corresponding electrode microstructure evolution. The accelerated interconnect/air electrode degradation in moist atmosphere and its relation to the reactions above need to be understood. Currently there is a lack of material design, testing, and characterization efforts to systematically answer these questions.

In this work, $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ (LSM) powder was made into air electrode and assembled with the AISI 441 alloy interconnect and yttria-stabilized zirconia (YSZ) electrolyte to simulate the air electrode side of SOFCs/SOECs. The microstructure and elemental distribution are investigated to evaluate the Cr poisoning of the porous air electrode layer. Focused ion beam (FIB) was used to assist the sample preparation for microstructure and surface analyses. The elemental analysis was

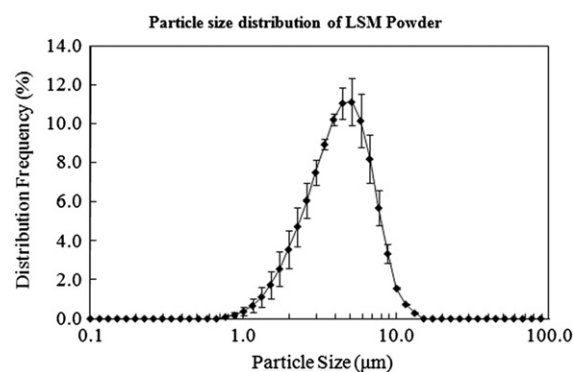


Fig. 1 – Particle size distribution of the LSM powder.

carried out by energy dispersive spectroscopy (EDS). The EDS results were corrected to minimize the interference of multiple elements. The effects of LSM electrode thickness and thermal treatment time on the Cr poisoning were investigated.

2. Experimental procedures

2.1. Sample preparation

LSM powder was prepared by conventional solid state reaction method [28]. SrCO_3 (99.9%, Sigma Aldrich, St. Louis, MO), La_2O_3 (99.98%, Alfa Aesar, Ward Hill, MA), and MnCO_3 (99.9%, Alfa Aesar, Ward Hill, MA) at designed composition ratios were mixed in a ball mill for overnight. The mixed oxide and carbonates were calcined in a box furnace (Lindberg, Model No. 51314, Watertown, WI) at 1200°C for 20 h. The LSM powder was ground in a ball mill for 72 h after the calcination. The particle size of the LSM powder was measured by a laser light scattering analyzer (LA-750, Horiba Ltd., Japan). The particle size followed a normal distribution curve. The mean particle diameter of the LSM powder was $2.92\ \mu\text{m}$ (based on three measurements, Fig. 1).

AISI 441 ferritic stainless steel samples (ATI Allegheny Ludlum Corporation, Brackenridge, PA) were prepared as rectangular substrates ($25.4 \times 25.4\ \text{mm}$, thickness 2.08 mm). They were polished to optical finish to remove the oxidized layer, if any, and to obtain a scratch free flat surface. The

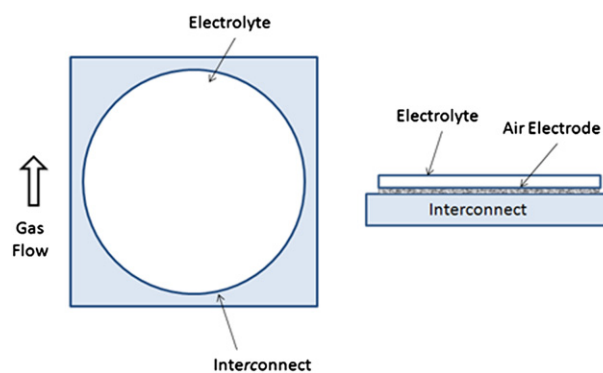


Fig. 2 – Configuration of the YSZ/LSM/AISI 441 tri-layer.

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