Journal of Power Sources 239 (2013) 315–320

Contents lists available at SciVerse ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Mechanical and electrochemical performance of composite cathode contact materials for solid oxide fuel cells

Michael C. Tucker [a,*], Lutgard C. DeJonghe [b], Valerie García-Negrón [c], Rosa Trejo [c], Edgar Lara-Curzio [c]

[a] Environmental Energy Technology Division, Lawrence Berkeley National Laboratory, 1 Cyclotron Rd, Berkeley, CA 94720, USA
[b] Materials Sciences Division, Lawrence Berkeley National Laboratory, 1 Cyclotron Rd, Berkeley, CA 94720, USA
[c] Materials Science & Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6062, USA

HIGHLIGHTS

- Addition of inorganic binder or glass to SOFC contact materials enhances bonding.
- Added materials do not degrade electrochemical performance.
- 1000 h stable operation achieved.

ARTICLE INFO

Article history:
Received 21 February 2013
Received in revised form
21 March 2013
Accepted 22 March 2013
Available online 2 April 2013

Keywords:
SOFC
Cathode contact material
Adhesion
Mechanical testing

ABSTRACT

The feasibility of adding glass or inorganic binder to conventional SOFC cathode contact materials (CCM) in order to improve bonding to adjacent materials in the cell stack is assessed. Two glasses (SEM-COM SCZ-8 and Schott GM31107) and one inorganic binder (Aremco 644A) are mixed with LSM particles to produce composite CCM pastes. These are used to bond $Mn_{1.5}Co_{1.5}O_4$-coated stainless steel mesh current collectors to anode-supported button cells. The cells are operated at 800 °C for about 1000 h. The cell with SCZ-8 addition to the CCM displays quite stable operation (3.9%/1000 h degradation), whereas the other additives lead to somewhat higher degradation rate. Bonding of the CCM to coated stainless steel coupons is also assessed. Interfacial fracture toughness is determined using a four-point bend test. The fracture toughness for LSM–Schott glass (12.3 N mm^{-1}), LSM–SCZ-8 glass (6.8 N mm^{-1}) and LSM–644A binder (5.4 N mm^{-1}) are significantly improved relative to pure LSM (1.7 N mm^{-1}). Indeed, addition of binder or glass is found to improve bonding of the CCM layer without sacrificing cell performance.

1. Introduction

Assembly of solid oxide fuel cell (SOFC) stacks typically involves mechanically and electrically connecting a number of cells and interconnects in series. Connection of the cathode to the interconnect (or coating on the interconnect) is usually accomplished by compression of the stack using an external load frame, and is often aided by the use of a cathode contact material (CCM). The CCM is an electrically conductive material, and is applied as a paste or ink during stack assembly to form a continuous layer or discrete contact pads. The CCM provides electrical connection between the cathode and interconnect, and can also serve to improve in-plane conduction over the area of the cathode. Fig. 1 indicates placement of the CCM in the fuel cell stack. Often, the CCM is simply a thick layer of the electrocatalyst used in the cathode [1]. For example, a thin LSM–YSZ cathode layer optimized for electrochemical activity can be covered with a thick LSM CCM layer optimized for gas transport and electrical conductivity. A significant limitation of this approach, however, is that most cathode compositions require firing at high temperature (>1100 °C) to achieve good sintering [2]. The use of ferritic stainless steel as the interconnect material limits the firing temperature to 1000 °C or lower. In practice, therefore, using a cathode catalyst CCM in conjunction with a stainless steel interconnect results in low CCM layer strength and minimal adhesion at the CCM/interconnect or CCM/cathode interface.

Our approach to this issue is to fabricate composite mixtures of SOFC cathode material and inorganic binder or glass. Conventional CCM pastes use a cathode material to achieve both bonding and electrical contact. In contrast, the approach we take is to separate

* Corresponding author. Tel.: +1 510 486 5304; fax: +1 510 486 4881.
E-mail address: mctucker@lbl.gov (M.C. Tucker).

0378-7753/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.jpowsour.2013.03.130

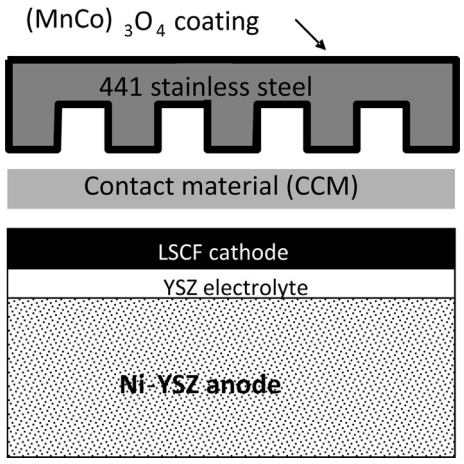

Fig. 1. Schematic representation of CCM placed between SOFC cell and coated stainless steel interconnect.

the functions of electrical contact and mechanical (or chemical) bonding: LSM is the electrical conductor, and the binder or glass serves to improve bonding. In our previous work, we screened a wide variety of glass and binder candidates, and the reader is referred there for a more complete discussion of the candidates and screening methods [3,4]. The most promising composite additives were determined to be Aremco 644A inorganic binder, and SEM-COM SCZ8 and Schott GM31107 glasses. Composites of LSM and these additives were demonstrated to improve bonding to LSCF and $Mn_{1.5}Co_{1.5}O_4$ (MCO)-coated 441 stainless steel at room temperature without sacrificing performance when using a Pt mesh current collector. In this work, we determine mechanical integrity and electrochemical performance under more realistic conditions. Several test methods exist to evaluate the adhesion and fracture toughness of interfaces [5]. In this work we have used a 4-point bend test method initially proposed by Charalambides et al. [6] and further developed by Hofinger et al. [7] because it requires a relatively simple test configuration and specimen geometry. Interfacial fracture toughness of the composites bonded to MCO-coated steel is determined, and electrochemical performance and stability are assessed in the presence of Cr-containing MCO-coated 441 steel current collectors.

2. Experimental methods

2.1. Materials

LSM powder was purchased from Praxair. Glass powders were purchased from SEM-COM (SCZ-8) and Schott (GM31107). Aqueous inorganic binder solution was purchased from Aremco (644A). Pastes were formulated and applied as described elsewhere [3,4]. MCO-coated 441 meshes were prepared by rolling thick 441 coupons (Allegheny Ludlum) down to about 150 μm thick. Perforations were introduced by chemical etching (Italix Company, Inc.), after which both sides were coated with MCO (PNNL), see Fig. 2. Finally, the meshes were flattened and preoxidized by placing between alumina plates and heating to 1000 °C for 1 h.

2.2. Cell testing

Anode supported button cells with LSCF cathode and GDC barrier layer (MSRI) were used. The cathode was coated with CCM paste and an MCO-coated mesh was applied. The glass-containing CCM layers were then sintered at 1000 °C in air for 2 h. The

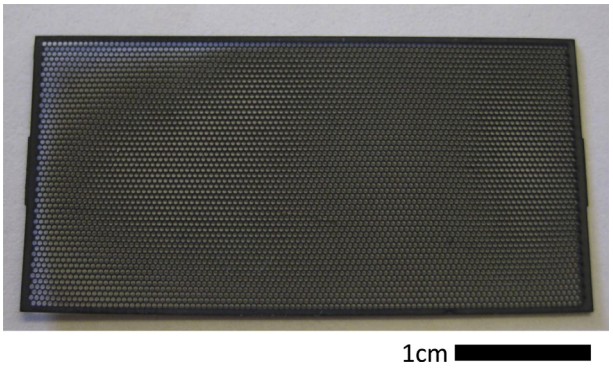

Fig. 2. Photograph of MCO-coated 441 stainless steel mesh prepared by chemical etching.

inorganic binder-containing CCM layer was cured according to the manufacturers' instructions (90° and 360 °C), and heated to 800 °C for 2 h. One cell with Pt mesh attached with Pt paste (Heraeus) to the bare LSCF cathode was prepared as a baseline. The cells were mounted to alumina tubes with Aremco 552 sealant and tested at 800 °C with 97%H_2/3%H_2O fuel and fresh air supplied to the cathode as shown in Fig. 3. The fresh air supply was intended to minimize any interaction between the sealant and Cr-containing mesh, as reported in Ref. [5]. After obtaining initial impedance spectra, the cells were operated at 300 mA cm^{-2} for up to 1000 h. DC current was applied in a 4-probe configuration using a Biologic VMP3 potentiostat.

2.3. SEM

Fracture surfaces of the cells were imaged with SEM and EDS (Hitachi S4300SE/N) after electrochemical operation.

2.4. Mechanical analysis

Specimens for interfacial fracture toughness were prepared from coupons of MCO-coated 441 steel. The coupons were coated with CCM paste and assembled as shown in Fig. 4. The pastes were cured or fired as described above. Dead weight was applied during the heating steps to ensure good contact between the coupons. The specimens were then subjected to 4-point bend testing at the Oak Ridge National Laboratory High Temperature Materials Laboratory using an electromechanical testing machine and a SiC fixture with a support span of 20 mm and a loading span of 10 mm. The diameter

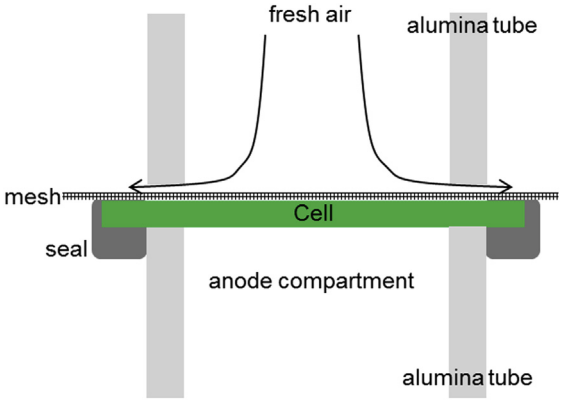

Fig. 3. Schematic of cell testing rig.