Journal of Power Sources 250 (2014) 166–173

Contents lists available at ScienceDirect

Journal of Power Sources

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

Long-term evaluation of solid oxide fuel cell candidate materials in a 3-cell generic short stack fixture, Part II: Sealing glass stability, microstructure and interfacial reactions

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HIGHLIGHTS

- A sealing glass was demonstrated chemically compatible with YSZ after 800 °C/6000 h.
- SEM showed some corrosion of alumina protection layer; however, no chromate was formed.
- Volatility study showed two stages of weight loss behavior.

ARTICLE INFO

Article history:
Received 24 June 2013
Received in revised form
25 September 2013
Accepted 29 September 2013
Available online 13 November 2013

Keywords:
Sealing glass
AISI441
Aluminization
(Mn, Co)-spinel
SOFC

ABSTRACT

A generic solid oxide fuel cell stack test fixture was developed to evaluate candidate materials and processing methods under realistic conditions. Part II of the work examined the sealing glass stability, microstructure development, interfacial reaction, and volatility issues of a 3-cell stack with LSM-based cells. After 6000 h of testing, the refractory sealing glass YSO7 showed desirable chemical compatibility with YSZ electrolyte in that no discernable interfacial reaction was identified. In addition, no glass penetration into the thin electrolyte was observed. At the aluminized AISI441 interface, the protective alumina coating appeared to be corroded by the sealing glass. Air side interactions appeared to be more severe than fuel side interactions. Metal species such as Cr, Mn, and Fe were detected in the glass, but were limited to the vicinity of the interface. No alkaline earth chromates were found at the air side. Volatility was also studied in a similar glass and weight loss in a wet reducing environment was determined. Using the steady-state volatility data, the life time weight loss of refractory sealing glass YSO77 was estimated to be less than 0.1 wt%.

1. Introduction

Solid oxide fuel cells (SOFCs) are an emerging technology which converts chemical energy directly to electrical energy at elevated temperatures. The energy conversion employs an ionic conductor (e.g., oxygen ion conductor such as YSZ) through which fuel and oxidant can react electrochemically, instead of through conventional combustion processes. Hence SOFCs can have higher energy conversion efficiency and a CO_2-concentrated exhaust which offers a more feasible and economic gas stream for carbon capture [1,2]. Stationary and transportational applications are current potential industrial markets. Two generic designs of SOFCs, tubular and planar, [3–5] are both being considered. For planar SOFCs, the advancement of ceramic processing has led to the development of the state-of-the-art anode-supported thin (<10 μm) electrolyte cells for which the operation temperatures can be lowered from ~1000 °C to ~800 °C. As a result, metals are being considered as potential interconnect materials, instead of the fragile and difficult-to-process ceramic interconnect such as $LaCrO_3$. However, great challenges are encountered when using metallic interconnect materials in a planar-type SOFCs where multiple tens of repeating unit cells are separated by metallic interconnect plates. These challenges apply to a spectrum of SOFC materials in terms of chemical compatibility, thermal stability, structural integrity, and electrical properties. For example, hermetic and durable sealing becomes more difficult when sealing dissimilar materials like metal to ceramic as compared to ceramic to ceramic, especially in the harsh dual environment with numerous thermal cycles and long life time requirements (e.g., ~40,000 h) at elevated temperatures. Chemical compatibility is another major concern for metallic interconnects when glass sealants are used. Current glass sealants,

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0378-7753/$ – see front matter © 2013 Published by Elsevier B.V.
http://dx.doi.org/10.1016/j.jpowsour.2013.09.148

such as a crystallizable glass like G18 which turns into a glass ceramic or a crystallization-resistant glass like the compliant SCN-1 glass, all contain appreciable amount of alkaline earths such as Ba and Sr [6–8]. These alkaline earths were found to be very reactive with Cr from metallic interconnects to form $BaCrO_4$ or $SrCrO_4$, whose very high thermal expansion can cause seal failure, if no protective coating is applied to the metal [9,10]. In addition to the chemical compatibility between sealing glass and Cr-containing metals, the issue of cathode poisoning is the primary concern when using these metals, as published in previous literature [11–16]. The coating not only needs to block Cr outward diffusion but also must be electrically conductive. A novel (Mn,Co)-spinel coating was therefore developed for ferritic stainless steels such as AISI441, which is a leading candidate for metallic interconnect as well as the window frame that surrounds the fragile ceramic cell [17,18]. These candidate materials have been extensively studied on their own or in small button-sized cells, but the behavior in a realistic SOFC stack environment, however, was rarely reported. It was therefore the objective of this work to study these candidate materials in a generic stack fixture to fully assess their stabilities and interactions with mating materials for long-term operation. Part one of the work reported the design of a generic stack fixture, cell assembly, seal system, and each cell's performance [19]. Part two of the work will address the sealing glass microstructure development, materials stability, and interfacial reactions with mating materials.

2. Experimental

2.1. Materials and stack fixture

A short stack of three cells was assembled using the generic stack test fixture. A schematic drawing of the assembly is shown in Fig. 1 [19]. A commercial NiO–YSZ anode supported thin YSZ electrolyte cell of 50 mm × 50 mm × 0.5 mm with LSM–YSZ composite cathode was used. The cells were sealed onto aluminized AISI441 window frames with refractory sealing glass YSO77 and stacked together with hybrid micas as the perimeter seal. LSM and NiO were used as cathode and anode contact materials, respectively. Ni mesh was also used at the anode side as a current collector while no metallic mesh was used at the cathode side. Ferritic stainless steel AISI441 was used to fabricate the interconnect plates and window frame, and was coated on the active cathode area with Ce-modified (Mn,Co)-spinel coating. The rest of the surface area was aluminized. The details of the cell, materials, coatings, processing and heat treatment are given in Ref. [19].

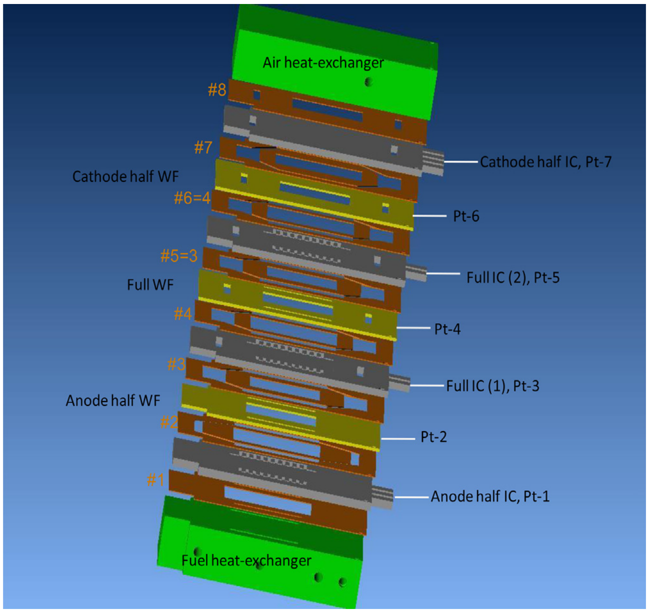

Fig. 1. Schematic drawing showing the assembly of the 3-cell short stack: three window frame plates in yellow, 4 interconnect plates in gray, 8 hybrid mica seals in brown. All were pressed between two heat exchangers for fuel and air, shown in green. WF = window frame, IC = interconnect, Pt = platinum wire (from Ref. [19]). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

2.2. Post-mortem characterization

After the long-term test, the short stack was dis-assembled. The post-mortem analysis was conducted with both optical microscopy and scanning electron microscopy with energy dispersion spectroscopy (JOEL SEM model 5900LV). Characterization with optical

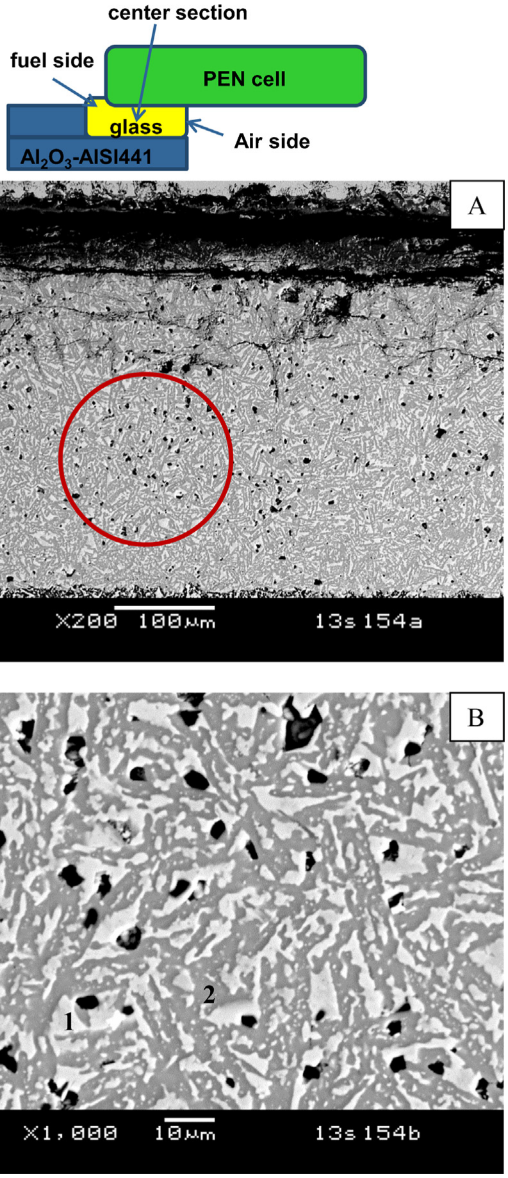

Fig. 2. Microstructure of the sealing glass YSO77 from the central section after 800 °C/6000 h test, (A) low magnification and (B) high magnification of the circled area in (A). The insert shows the location of glass for characterization. EDS of spots#1 and #2 are listed in Table 1.