INTERNATIONAL JOURNAL OF HYDROGEN ENERGY 40 (2015) 1523–1533

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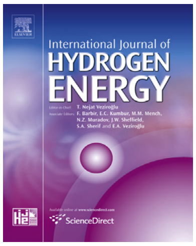

Interfacial characterization of nickel–yttria-stabilized zirconia cermet anode/interconnect joints with Ag–Pd–Ga active filler for use in solid-oxide fuel cells

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ARTICLE INFO

Article history:
Received 12 June 2014
Received in revised form
4 November 2014
Accepted 6 November 2014
Available online 12 December 2014

Keywords:
Solid oxide fuel cell (SOFC)
Interconnect
Joint strength

ABSTRACT

Nickel–yttria-stabilized zirconia cermet anode (Ni–YSZ)/interconnect joints with silver–palladium–gallium (Ag–9Pd–9Ga) active fillers are prepared by vacuum brazing. The joint structure and microstructure are analyzed by X-ray diffraction (XRD) and scanning electron microscopy coupled with energy dispersive spectroscopy (SEM/EDS), and by using an electron probe microanalyzer (EPMA). SEM observations of the joint show no cracks near the interface, confirming the compatibility of the Ag–9Pd–9Ga filler with a different anode and interconnect. The XRD pattern of the joint specimens oxidized at 800 °C for 250 h shows Cr_2O_3 and $(Mn,Cr)_3O_4$ surface layers. EPMA analysis of the cell/Ag–9Pd–9Ga/alloys joint at the cross section shows Cr, O, Fe, Zr, Ni, Ag, Pd, Ga, Y, and Mn. Overall, the results indicate that the bonding between metal and cermet is well established and the interface is smooth. By correlating the XRD and EPMA analysis results, we also analyzed the possible stages during joint oxidation. The joint strength was evaluated at 25 and 800 °C under shear and tensile loading conditions, respectively, and the brazed Ag–9Pd–9Ga sealant compared favorably with the commercially available glass-ceramic GC-9 counterpart.

Introduction

A solid oxide fuel cell (SOFC) is an electrochemical fuel cell that produces electricity directly through oxidation of a fuel. SOFCs are characterized by the use of a solid oxide as the electrolyte. In principle, a fuel cell is composed of an anode (exposed to the fuel), an electrolyte, and a cathode (exposed to the oxidant) [1]. In a conventional SOFC, the oxide-ion conducting electrolyte is a dense yttria-stabilized zirconia (YSZ) membrane sandwiched between a porous nickel–zirconia cermet anode and a doped lanthanum–manganite–perovskite cathode [2]. Among the

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http://dx.doi.org/10.1016/j.ijhydene.2014.11.028

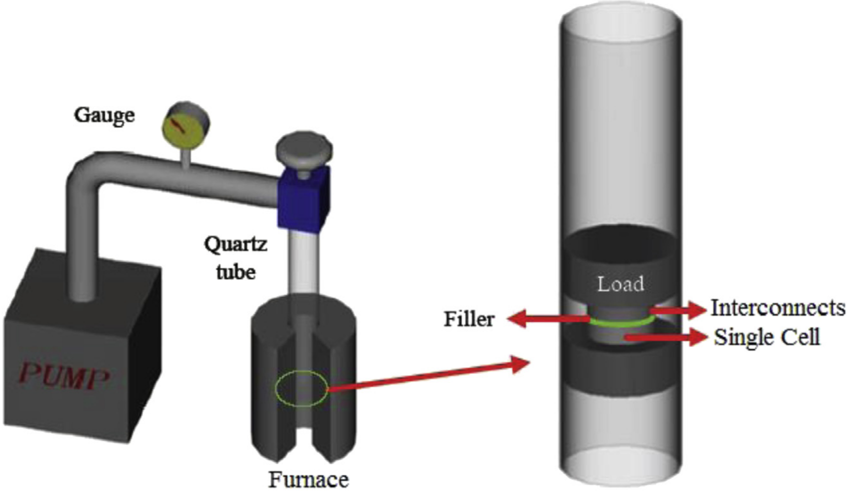

Fig. 1 – Schematic of the vacuum brazing system used in this study.

different types of SOFCs, planar–type SOFCs are considered the most cost-effective and mechanically robust, and they exhibit potential for higher power densities compared to other types of SOFC [3]. To obtain usable cell voltage and power, several individual cells are generally connected in series to form a "stack." The repeating unit of a planar configuration is formed by combining joint anode–electrolyte–cathode structures that are connected using a sealing material. These sealing interconnect materials form a critical component of SOFCs as they electrically and structurally connect adjacent fuel cells in a sequentially stacked SOFC [3,4]. In other words, the interconnect material in a stacked SOFC ensures electrical connection between the anode of one individual cell (repeating unit) and the cathode of the neighboring cell [2]. The reduction in SOFC operating temperature has enabled the use of oxidation/corrosion-resistant metallic alloys as interconnect materials. Further, metallic alloys are being increasingly used instead of ceramics interconnects since they offer advantages such as low cost, high toughness, and excellent manufacturability [5].

The key problem associated with the fabrication of planar SOFCs is the sealing of the electrolyte (that could be YSZ-wafer- and anode-supported, depending on the cell configuration) with the metallic interconnect, in order to obtain a hermetic (gas tight) joint. The ceramic/metal joints in SOFCs require resistance against grain coarsening and thermomechanical degradation, in addition to high strength, and good wetting properties [6,7]. To this end, diffusion bonding, friction welding, transient liquid-phase (TLP) bonding, and brazing have been commonly used for realizing ceramic/ceramic and ceramic/metal joints. Among the robust integration technologies available for joining oxides to metals, active metal brazing is the most commonly used method [8–13]. Thus far, studies have focused on joining ceramics to metals by using active brazes such as Ag-based [8–10], Cu-based [8,11], Au-based [12], and Pd-based [13] metal fillers. Nevertheless, the high operating temperatures in air and fuel environments can lead to material degradation in SOFCs, especially at the metal and ceramic joints [14]. Metallic sealants have been shown to facilitate more rigid sealing and better stress accommodation than ceramic sealants [15,16]. The main disadvantage of traditional ceramic sealants such as glass is material availability. Widely reported glass sealants are not commercially available and the specific ingredients are highly classified as propriety materials. In order to sustain high-temperature operation under dual atmospheres, metallic sealants that can afford oxidation resistance and long-term chemical stability should be judiciously selected. A number of sealant systems based on noble metals have shown remarkable properties, and they exhibit potential for large-scale applications [15,16]. Pure Ag is an attractive filler base metal because of its chemical inertness and excellent ductility, fluidity, and thermal (428 W m^{-1} K^{-1}) and electrical (6.8×10^7 S m^{-1}) conductivity. The noble metal Pd is an attractive base filler metal that is used as an alloying additive to Ag-base brazes [17]. The use of palladium and gallium allow high-temperature operation, while providing high oxidation resistance and ductility. Although the function of different seals in the SOFC could differ substantially depending upon the SOFC design, the most crucial expectation from all SOFC seals is hermeticity. Coefficient of thermal expansion (CTE) mismatch and elemental inter-diffusion between seals and cell and/or interconnect can affect hermeticity after prolonged use. Thus far, stainless steel has been the most commonly used

Table 1 – Chemical compositions of the alloys (wt%).

Alloys	Fe	Cr	Mn	Si	Cu	Al	S	P	Ti	La	Nb	Ni	Zr	C
Crofer22 APU	Bal.	24.36	0.49	0.05	0.01	0.01	0.02	≦0.05	0.06	0.08	–	0.02	–	0.01
Crofer22H	Bal.	24.18	0.39	1.54	0.02	0.06	0.01	≦0.03	0.07	0.07	0.83	0.19	–	0.02
SS430	Bal.	17.13	0.8	0.42	–	–	–	–	–	–	–	0.16	–	0.02
ZMG232L	Bal.	22.42	0.15	0.11	0.01	0.04	0.01	≦0.03	–	0.06	–	0.42	0.16	0.03