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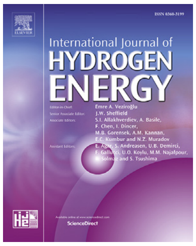

Oxidation, electrical and mechanical properties of Crofer®22 solid oxide fuel cell metallic interconnects manufactured through powder metallurgy

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ABSTRACT

This study aimed to investigate oxidation, electrical and mechanical properties of solid oxide fuel cell interconnects. To this goal, two different Crofer®22 interconnects samples were produced via different manufacturing routes (machining from bulk material, and powder metallurgy approach). The samples were characterized by scanning electron microscopy with energy dispersive spectroscopy (SEM-EDS), X-ray diffractions. Four-probe area specific resistance (ASR), bonding strength, leakage tests were also performed. The results indicated that interconnect sample manufactured through powder metallurgy approach can be a reliable alternative to the one manufactured from commercially available Crofer®22 alloy in bulk form.

Introduction

Energy demand is endlessly growing along with the increase in population and industrialization. Solid oxide fuel cells (SOFCs) are energy conversion devices that can directly convert fuel into electricity without combustion [1–3]. Due to high operation temperatures (600–1000 °C) of SOFCs, they do not need any precious catalyst and have several attractive features such as fuel flexibility, silent operation, environmental friendliness and higher efficiencies that can go up to 80–90% through co-generation [4–10]. SOFCs basically consist of membrane electrode assembly (MEA) where electrochemical reaction occurs, interconnects that provide mechanical support and collects the current produced in the cells, and sealants that prevent air/fuel leakages in between MEA and interconnects [11–14]. Interconnects are used to provide distribution of the fuel and oxidant to the electrodes and collect the current as well as provide mechanical strength

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and integrity to the SOFC stack [15–20]. Ferritic stainless steels (FSS) have become the preferred choice for the interconnect material replacing more expensive ceramic counterparts in recent years [21–25]. FSS offer several advantages over the $LaCrO_3$-based ceramic interconnects such as low material cost, good formability, higher mechanical, and electrical properties, good oxidation resistance as well as they have compatible coefficient of thermal expansion (TEC, $10.5–12.5 \times 10^{-6}$ K^{-1}) with other components of SOFC stack [22,25–27].

Metallic interconnects are manufactured using the casting-rolling-(forging) route with subsequent machining (wire erosion approach) of the semi-finished products, traditionally [5,28–31]. Powder metallurgy (P/M) is another approach to manufacture metallic interconnects and has some advantages over traditional machining such as near-net shape production, fast and high production rates, elimination or reduction of machining step and scrap material [32–34]. Glatz et al. and Antepara et al. manufactured interconnect plates used in SOFCs by powder metallurgy method. Powder metallurgy approach was found much easier than traditional method [21,35–37]. Antepara et al. manufactured interconnects with porous structure and investigated the influence of porous structure on the SOFC efficiency. Same group of researchers studied also the interconnect plates manufactured using Crofer®22, ZMG232 and FeCr (70:30) metal powders and investigated the relation between electrical conductivity and porosity [38,39]. These interconnects was found compatible with electrolyte in terms of TEC. It was also indicated that near-net shape interconnects were obtained with relatively higher efficiency [40].

There are numerous studies in the literature regarding with the oxidation behavior of metallic interconnects in recent years. Different coating techniques have been employed to increase the oxidation resistance of metallic interconnects [41–49]. Miguel-Pérez et al., investigated the formation of oxide scales on Crofer 22 APU, SS430 and Conicro 4023 W188 metallic interconnects at 800 °C for 100 and 1000 h duration. They noted that spinel $(Fe, Cr, Mn)_3O_4$ and $(Fe, Cr, Ni)_3Co_2O_4$ outer layers and a chromia (Cr_2O_3) inner layer were formed on the interconnects. Crofer 22 APU and Conicro 4023 W188 samples were determined to be more promising as metallic interconnects compared to SS430 due to their higher oxidation resistance [41]. Przybylski et al., coated a $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$ (LSCF48) film on the Crofer 22 APU steel using the screen-printing technique and compared to oxidation behavior of coated and pure Crofer 22 APU. The oxidation tests were performed in air, under isothermal and cyclic conditions at 800 °C. The results indicated that the coated composite provides resistance against oxidation [42]. In another study, Hosseini et al., coated bare and pre-oxidized (100 h at 800 °C) Crofer 22 APU samples' surfaces with $CuFe_2O_4$ spinel layers and samples were exposed to oxidation for 400 h at 800 °C in air. The coating reduced the evaporation of Cr by 92% and 83% for bare + coated and pre-oxidized + coated alloy, respectively when compared to the uncoated substrate material [44].

Interconnects should have good electricity conductivity and low resistivity as it is responsible from collecting the current in cell stack. There are various electrical conductivity measurement techniques employed in literature including two point-four wire probe and four point probe methods [8,44,47,50–63]. Tondo et al., coated AISI 430 stainless steel interconnects with different coatings (Y_2O_3, Y_2O_3/Co_3O_4 and Y_2O_3/Au composite films) and ASR values of coated samples were measured after oxidization in air at 800 °C up to 500 h using four-point method. Results showed that ASR values of smaller than 100 mΩ cm^2 were obtained for all samples and indicated that ASR values below the conventional acceptability limit for SOFC applications of 100 mΩ cm^2 [50]. Safikhani and Aminfard, studied Fe-22Cr-0.5Mn FSS interconnect with addition of Ti and W at different contents. Electrical and oxidation behaviors of those samples were investigated and ASR measurements were performed using four-probe method. Results showed that the F.Ti1W2 sample with 3.98% wt of W and 0.23% wt of Ti is the best composition among the tested samples as the highest oxidation resistance and the lowest electrical resistance were obtained for this sample [52]. In another study by Molin et al., the surface of Crofer 22 APU was coated with $Mn_{1.5}Co_{1.5}O_4$ coating by three methods (electrophoretic deposition (EPD), thermal co-evaporation and RF magnetron sputtering). They electrically tested the coated samples for 5000 h at 800 °C and determined ASR values. After 5000 h oxidation, ASR values were determined to be in the 22–35–45 mΩ cm^2 range for the EPD, RF sputtering and thermal co-evaporation methods, respectively. EPD method was noted to provide the best protection against Cr diffusion, contributing to the lowest ASR value and lowest increase rate [60].

Glass-ceramic sealants which bond with metallic interconnects are widely used for preventing the gas leakage. Due to their distinct thermal expansion coefficient (TEC) values, thermal stresses and cracks are generated during start-up and shutdown, and these consequently may lead to gaps at contacting surfaces, and failures. Contact loss at contacting surfaces causes gas leakages and performance loss in the cell. Therefore, an interconnect should have the sufficient strength not only to provide structural integrity but also to prevent from gas leakage. Tensile test method is the widely used to determine the bonding strength in literature [64–71]. Lin et al., developed a glass-ceramic sealant (GC-9: $BaO\text{-}B_2O_3\text{-}Al_2O_3\text{-}SiO_2$) and studied joint strength of Crofer 22 H/GC-9/Crofer 22 H sandwich specimens using tensile test method [65,67]. Wang et al., composed a LSM/YSZ/LSM cell to reveal a correlation between the adhesion strength and the ohmic resistance at the electrolyte-cathode interface. The interfacial adhesion strength was measured by means of tensile test and the results showed that the ohmic area specific resistance decreased gradually with the increase of the adhesion strength at the interfaces [70]. The Weibull analysis is a common way to estimate the failure probability of materials [72]. This method and reliability curves are often used to define bonding strength results [69,73–79].

Sealing is one of the most important parameters influencing the fuel cell performance. The most common method is to determine the pressure drop between the gas inlet and outlet is leakage test [71,81–89]. The leakage tests are mostly performed at low temperatures and pressures. Zhang et al., carried out the leakage test, starting at 10 kPa pressure level and continued until 0,5 kPa at temperature range of 100–600 °C [81]. In another study by Le et al., leakage tests

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