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Effect of intermediate nickel layer on seal strength and chemical compatibility of glass and ferritic stainless steel in oxidizing environment for solid oxide fuel cells

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ABSTRACT

The effects of intermediate nickel layer on seal strength and chemical compatibility of seal glass and interconnect materials for solid oxide fuel cells (SOFCs) were investigated. Two types of samples (metal/glass/metal sandwiches and glass coated metals) were prepared with the sheet of AISI 430 (nickel plated and uncoated) and slurry of compliant silicate sealing glass (SCN-1). The joined and coated samples were heated at 850 °C for different time durations (0.5–100 h). Tensile and impact tests were performed and SEM micrographs were used to analyze the glass/metal interaction. The results indicate that nickel plated AISI 430 shows higher adhesion strength at short durations of heating due to dendrite development at the interface. For longer durations, intermediate nickel layer leads to rapid loss of adhesion strength due to extension of unstable austenite zones but prevents the accelerated weakening near the triple-phase boundaries metal/glass/air (TPB) by compensating for absence of protective oxide layer (Cr–Mn oxide).

Introduction

Solid oxide fuel cells (SOFCs) are emerging technologies for direct conversion of fuel to electricity. SOFCs are considered as alternative systems for generating electric power due to their interesting features such as high energy-conversion efficiency, fuel flexibility, environmental safety, low noise and ability to recover exhaust heat [1–3]. One of the major challenges for commercializing SOFCs, especially planar-SOFCs, concerns sealant materials. These sealants must prevent fuel–oxidant mixing and provide electrical insulation of the stack layers for long times (5000–40,000 h) at high temperatures (between 600 °C and 1000 °C) [4,5]. Several approaches have been used to seal SOFCs including compliant (viscous) seal, compressive (soft) seal and rigidly bonded (semi-rigid) seal [6–8]. Glasses and glass-ceramics (GCs), especially the silica-based ones, have been extensively studied in all three approaches due to achievable good properties such as low electrical conductivity, compatible coefficient of thermal expansion (CTE), good adhesion and limited reactivity with SOFC components and atmospheres [6–11].

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A determining factor affecting the durability of SOFCs is the interaction of glass with interconnects. Ferritic stainless steel (FSS) is the most promising candidate for interconnect applications due to its low cost, good CTE match with electrolyte materials and high oxidation resistance [12]. Nevertheless, undesirable reactions between glass and FSS, especially near the air-side, can potentially lead to deleterious high expansion chromate formation, microstructure degradation and electrical shorting [13,14]. Therefore, it seems that further improvement in long-term stability is needed. On the other hand, chemical interactions are necessary to achieve a satisfactory glass/FSS adhesion during thermal cycling, especially below the glass transition temperature (T_g) when the glass is not soft [15–18]. For these purposes, in addition to modification of glass and FSS compositions [19–23], many efforts have been made on the modification of FSS surface including pre-oxidation [24,25], aluminizing [26–29] and applying protective coating [30–37].

Recently, nickel has been investigated as protective coating for AISI 430 and AISI 441 in order to reduce the rate of Cr evaporation and prevent the growth of semi-conductive Cr_2O_3 scale under SOFC conditions [38–41]. Also, it has been reported that Ni-oxide improves the adhesion and reduces the metal/glass interactions [30]. Although effects of intermediate nickel layer between steel and glass are very well-known in porcelain enameling industry for many years [42–44], the details of its behavior in SOFC conditions are not available.

The purpose of this study was to provide a better understanding of chemical interaction of glass/metal adhesive joints with/without intermediate nickel layer. The impact test was used to validate the low strain rate tensile test for SOFC sealing applications. Also, SEM micrographs were analyzed based on prior knowledge about the glass/metal interactions (corrosion, enameling, glass coloring and …). SCN-1 as a non-crystallizing compliant sealing glass allows us to focus on interfacial interactions for seal strength investigation.

Experimental

Materials and sample preparation

Commercial alkali silicate glass (SCN-1, Par-e-tavous, Khorasan-e-razavi, Iran) was used. This silicate glass contains alkaline earth elements, mainly in the form of BaO (8.23 mol%) and CaO (3.34 mol%), alkalis of K_2O (10.0 mol%) and Na_2O (7.3 mol%), Al_2O_3 (2.8 mol%), and some impurities (less than 1%) of Fe, Mg and Ti with the balance of SiO_2. T_g, softening point (T_d), and CTE were about 470 °C, 550 °C and $11 \times 10^{-6}\ °C^{-1}$, respectively. The details of glass composition and its thermal behavior with AISI 441 are available in the literature [17,28,33,45].

AISI 430 is a commercial ferritic stainless steel (Hardox, Oxelösund, Sweden) containing Cr (17.5 wt%), Ni (0.13 wt%), C (0.05 wt%), Mn (0.25 wt%), Cu (0.13 wt%) and Si (0.15 wt%) with the balance of Fe. In this study, AISI 430 sheets with a thickness of 0.5 mm were used in two states of Ni-plated and uncoated which are specified here by N and S, respectively. N sheets were prepared as follows; AISI 430 was abraded with #1200 grit abrasive paper, the surfaces were degreased with acetone and activated in Woods nickel strike solution and then nickel was electroplated using Watts solution until obtaining a thickness of around 10 μm. Each step was followed by water rinsing to remove chemical residues. The interactions between SOFC atmospheres and FSS have an important role in damage growth near the triple-phase boundaries metal/glass/air (TPB). The coating with thickness of 10 μm is chosen because it has been reported that FSS with 8–10 μm thick nickel coating performs better under SOFC atmospheres [38,41].

The glass was applied onto the AISI 430 surface by using a slurry method. The slurry was a mixture of glass powder and minor additives (borax, sodium nitrate, kaolin, and silica) dispersed in deionized water. Two types of samples were prepared: glass coated AISI 430 sheets with dimensions of $6 \times 6\ cm^2$, for impact tests and AISI 430/glass/AISI 430 sandwiches (joined samples) with dimensions of $1 \times 1\ cm^2$ for tensile tests. The thickness of glass in type one and two after joining was 0.4 ± 0.1 mm and 0.5 ± 0.05 mm, respectively. After applying the slurry, the samples were dried at 70 °C for 15 min. The dried samples were then thermally treated at 850 °C for different time durations (0.5–100 h) in air. The heating and cooling rates were about 9 °C/min and 2 °C/min, respectively. The code of the samples was based on the surface condition of AISI 430 sheet and heat treatment duration; For example, N1 was assigned to a sample with Ni-plated sheet which was heat treated for 1 h.

Mechanical testing and microstructural characterization

Impact test was performed in accordance with a modification of EN 10209 Annex D method in ambient conditions [46]. In this method, a punch with hemispherical tip hits glass coated sheet and the adhesion strength is evaluated based on the destroyed surface appearance. For quantifying the results, the percent of bare areas which is inversely proportional to the adhesion level was calculated with the aid of the ImageJ software (version 1.46) [47]. For seal strength tests, the joined samples were glued to two aluminum test fixtures by cyanoacrylate (as shown in Fig. 1). The fixture had a self-alignment joint to minimize bending or twisting during tensile testing. Detailed information about test principles and equipment is described in Refs. [24,48]. The assembly was then tested in uniaxial tension with a cross-head speed of 0.5 mm min^{-1} in ambient conditions. For each condition, 4 samples were tested, the outlier was discarded, and the average strength was determined. Some of the samples were also mounted in epoxy and then sectioned and polished for interfacial characterization using optical (Olympus BX51M) and scanning electron microscopes (SEM VEGA\\TESCAN-XMU and FE-SEM TESCAN MIRA3 LM).

Results and discussion

Seal strength

Fig. 2 shows the room temperature seal strengths of AISI 430/glass/AISI 430 joined samples as a function of heating duration. Fracture surface analysis revealed that crack propagation

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