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Optimum processing parameters to improve sealing performance in solid oxide fuel cells

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Abstract

Glass–ceramic composites are among the favorable candidates as a sealing material for solid oxide fuel cells (SOFC). In order to obtain a reliable, robust and hermetic sealing, the glass–ceramics must chemically bond to both the metallic interconnector and the ceramic electrolyte. A high-bonding strength and good wetting, which strongly depend on the thermal treatment, are always preferred to ensure gas-tight sealing. The thermal treatment involves three stages: binder burnout (stage-I), sintering (stage-II), and cooling (stage-III). This study investigates effects of various parameters on the sealing quality at the sintering stage. The effects of sintering temperature, clamping pressure and sealant thickness are considered. The glass–ceramic laminates are produced employing a tape casting method. The sealing quality is evaluated by measuring leakage and final macro-structure of the sealing region. It is suggested that a 900–930 °C sintering temperature and 1.5–7.6 N cm^{-2} clamping pressure ranges are better for successful sealing. The initial thickness of glass–ceramic laminates is also desired to be between 0.25–0.5 mm thickness range for both a cost-effective and reliable sealing.

Keywords: Solid oxide fuel cell; Glass–ceramic sealant; Thermal treatment; Wetting behavior

1. Introduction

Solid oxide fuel cells (SOFC) are considered to be one of the most effective electrochemical energy conversion systems due to their high energy conversion efficiency, near zero emission, ability to employ a variety of fuels, and feasibility of co-generation. Among several SOFC designs, the planar geometry is the most preferred. SOFCs have simple structural geometry, easier fabrication and lower fabrication cost. However, several technological hurdles need to be overcome for commercialization and extension of their application areas. One of the major challenges is gas-tight sealing at the high-operating temperatures of the SOFC (700–850 °C). Hermetic sealing is essential for the SOFC stack to avoid unwanted leakages (fuel and oxidant) and performance losses during the fuel cell operation.

Glass–ceramics, which have superior sealing performance compared to other sealing materials, are commonly employed in the planar SOFC. Unlike the other sealing materials, glass–ceramics can chemically bond with both electrolyte and interconnector materials and are able to soften and flow at elevated temperatures (above the SOFC operation temperature), thus, a gas-tight sealing that is required for SOFC stack operation to be achieved. In recent years, the glass–ceramic sealing materials have received much attention since a range of properties (thermo-mechanical and thermo-chemical) can be attainable through modification of the composition. By carefully selecting the glass composition, it is possible to develop compatible thermal expansion coefficient (CTE), electrically insulating, thermally and chemically stable, mechanically robust glass seals. Furthermore, the resulting glass–ceramic seals are usually easy to fabricate and cost effective [1–5].

The sealing quality of the cell is affected by not only the glass–ceramic seals' composition, but also manufacturing procedures. In order to obtain a leak-free SOFC stack, it is essential to apply a

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careful and suitable manufacturing procedure. The glass–ceramic seal production procedure mainly involves a solid loading, tape casting and thermal treatments [6–10]. In a previous study, authors investigated effects of solid loading, heat treatment temperature, sweeping gas type, and tape casting on the glass–ceramic sealing quality at the binder burnout stage [11]. After the binder burnout stage, glass–ceramic powders form a porous network structure. Since there is no force to keep the powders together, a careful sintering process is required to bond the components at elevated temperatures (850–1000 °C). At these higher temperatures, very strong bonding is formed between the interfaces of seal glass-cell components (electrolyte and interconnectors). Although the bonding strength is not directly a sign of the sealing performance of glass–ceramic seals, higher bonding strength is preferred for a hermetic sealing. The bonding strength is shown to be affected from different parameters such as ceramic composition, sealing conditions, thermal treatments, interfacial morphology affect, thermal cycle and long term operation [12–16]. Glass ceramic sintering temperature, compressive force, glass ceramic thickness also affects bonding strength [17].

Although numerous studies are available in the literature about various parameters affecting sealing and bonding strength of glass ceramics, there is no study considered glass ceramic sealing performance at the sintering stage employing industrial size of SOFC short stack. Studies usually consider button size SOFCs for experiments. However, a good contact and homogenous distribution of pressure becomes important on the sealing quality at larger sizes of SOFC stacks. Furthermore, it is hard to obtain realistic outcomes using those experimental results. Therefore, this study considers a comprehensive investigation of the effect of sintering temperature, clamping pressure, and initial glass–ceramic thickness on the sealing with an industrial size of SOFC stack (81 cm^2 active area cell).

2. Experimental

2.1. Glass seal preparation

Commercial glass–ceramic powders (SCZ-8, SEM-COM Company, Inc., Ohio, USA) were used for all tests because of their suitable sealing temperature for SOFC applications (around 800 °C) [18]. The glass–ceramic powders were mixed with an organic dispersant (fish oil, Sigma-Aldrich, Munich, Germany), solvent (ethyl alcohol, Sigma-Aldrich), plasticizer (polyethylene glycol, Sigma-Aldrich) and binder (Butvar, Sigma-Aldrich). The mixture was ball milled (about 24 h) to obtain homogeneous slurry. The slurry was tape casted with a blade gap of 200 μm (Keko Equipment Ltd., CAM-L252TB, Zuzemberk, Slovenia). Then they were stacked and laminated under isostatic pressure. A range of sample thicknesses (1 mm, 0.5 mm, 0.25 mm, 0.2 mm) were obtained after isostatic pressure. Then the glass–ceramic samples were cut into square window frames (11.5 cm × 11.5 cm and 10 cm × 10 cm) to fit into SOFC test cells by a laser cutter (Versa Laser,VSL2.3, Australia). The ready-to-use glass–ceramic laminates and other short stack components are shown in Fig. 1.

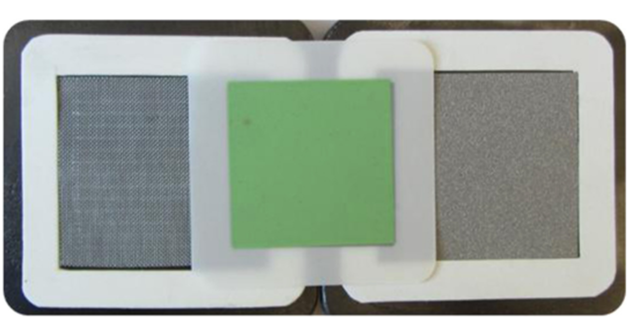

Fig. 1. The tape casted glass ceramic sheets and short stack components.

2.2. Experimental setup and testing

To evaluate the sealing performance of glass–ceramic samples, real SOFC operation conditions were set up. Two glass–ceramic sealants for anode and cathode sides, electrolyte supported SOFC cell [19–21] which has 81 cm^2 (9 cm × 9 cm) active area and 132.25 cm^2 (11.5 cm × 11.5 cm), were employed. Anode (porous Nickel) and cathode (Crofer) meshes were used for homogeneous current collection and current distribution in the anode and cathode sides, respectively. All components were stacked between two metallic interconnectors which were made of a special high corrosion resistant alloy Crofer22APU (Thyssen Krupp, Germany). The details of the short stack configuration are shown in Fig. 2.

The short stack is placed in the furnace as shown in Fig. 3. The furnace has a push rod capability which can apply a load ranging from 0–1400 N. The clamping pressure was defined as clamping force over total area of short stack surface (11.5 × 11.5 cm = 132.5 cm^2). Fig. 3 also shows the leakage measurement test systems. Two different leakage measurement test systems were employed in this study. One of the test systems, which is shown in Fig. 3a, was used to measure the leakage from the sealing regions (external leakage). Two precise flow meters (Alicat Scientific Incorporated, Tucson, USA) with an accuracy of 0.1 ml min^{-1} (sccm) were used to measure the leakage rate with N_2 inert gas. Glass–ceramic sealing performance was determined measuring inlet and outlet flow rate. The constant value of N_2 inert gas (100 ml min^{-1}) was supplied to the short stack inlet, and the short stack outlet was measured simultaneously. The difference of flow rate between inlet and outlet indicates the leakage of the test stack

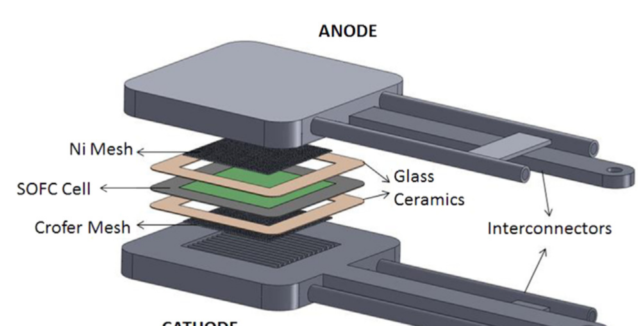

Fig. 2. SOFC short stack test fixture.