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Influential parameters and performance of a glass-ceramic sealant for solid oxide fuel cells

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Abstract

Glass-ceramic composites are among the few materials suitable for solid oxide fuel cell (SOFC) sealing application due to their high operating temperatures (600–850 °C). Glass-ceramics can chemically bond to both the metallic interconnector and the ceramic electrolyte and provide a gas-tight connection. However, a careful manufacturing procedure, which includes several stages, is required to obtain a gas-tight seal. In this study, the joint strength of the glass-ceramic sealant between two metallic interconnectors is experimentally investigated for different surface properties of the metallic interconnector. According to the experimental results, the optimum sintering temperature and pressure are found to be 870 °C and 0.5 kg cm^{-2}, respectively. In addition, the best bonding strength among the support materials considered is obtained for NiO. Furthermore, the sealing thickness is optimized as 0.6 mm.

Keywords: Solid oxide fuel cells; Glass-ceramic sealant; Joint strength; Sintering temperature; Interconnector coating

1. Introduction

Solid oxide fuel cells (SOFCs) are an important technology for high-efficiency electrical energy generation based on an electrochemical conversion. However, there are several technological challenges that must be solved to enable the commercialization of SOFCs and to extend their application areas. The process of sealing is one of these challenges, as a sealant for SOFCs must have the required adherence, mechanical strength, chemical stability, and electric insulation while providing a thermal expansion match with the other system elements at the operating conditions [1]. The high-temperature operation, however, leads to a significant thermal stress generation due to mismatch of the coefficient of thermal expansion (CTE) of the components as a result of the temperature gradients in the SOFC system [2–4].

Glass ceramics are widely used as a sealant for SOFCs. To develop a suitable glass-ceramic sealant, it is therefore necessary to understand the crystallization kinetics, the sealing properties and the chemical interactions when ceramic sealants come into contact with the other components of the cell under the SOFC working conditions. In recent years, numerous studies have been performed to develop a suitable glass-ceramic as a leak-free and durable sealant for SOFCs [5–12]. However, studies on the detailed investigation of the effects of the different surface conditions on the joint strength between a glass-ceramic sealant and a metallic interconnect are limited.

Smeacetto et al. [13], for example, studied the development and the performance of a glass-ceramic sealant used to join the ceramic electrolyte to the metallic interconnects for planar SOFC stacks. The glass-ceramic sealant was found to be effective for joining both Crofer 22 APU and anode-supported-electrolyte, but no data of mechanical strength were reported. Lin et al. [1] investigated the joint strength between a newly developed glass-ceramic sealant and a metallic interconnect for planar solid oxide fuel cells. A pre-oxidation treatment of the metal coupon was found to not result in a beneficial effect on the shear and tensile joint strength for all testing conditions studied.

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There are also several series of studies on the determination of the mechanical properties of glass-ceramic sealants via tensile tests [14–21]. In these studies, the tensile and the shear joint strengths between a metallic interconnect and newly developed glass-ceramic sealants were investigated. In addition, the effects of the pre-oxidation and the thermal aging on the bonding strength were investigated at both room and SOFC operation temperatures.

Therefore, in this study, the effects of the surface conditions of the metallic interconnector on the mechanical performance of commercially available glass-ceramic powders are investigated. The effects of several operational and glass forming parameters such as the temperature regime (different heating rates), the compressive force, the coating (polished, spinel, aluminized, etc.), the support materials (Yttria-stabilized zirconia (YSZ), Nickel Oxide (NiO), Scandia Stabilized Zirconia (ScSZ), and 3 mol% Y_2O_3 (3YSE)) and the sealant thickness are experimentally investigated, and the optimum parameters for better sealing are determined.

2. Experimental section

2.1. Fabrication of the glass-ceramic sealant

Commercially available glass-ceramic powders (Viox-V1649, Seattle, USA) were mixed with an organic dispersant (fish oil, Sigma-Aldrich, Munich, Germany) and a solvent (ethyl alcohol, Sigma-Aldrich). After ball milling for approximately 12 h, a certain amount of plasticizer (polyethylene glycol, Sigma-Aldrich) and binder (Butvar, Sigma-Aldrich) was added to the mixture. The composition of glass-ceramic powders is mainly B_2O_3, SrO, and La_2O_3. The slurry was ball milled again for another 12 h. Next, the slurry was tape cast with a blade gap of 200 μm via laboratory-scale tape casting equipment (Keko Equipment Ltd., CAM-L252TB, Zuzemberk, Slovenia). A sufficient amount of glass-ceramic tapes were stacked together and laminated under an isostatic pressure of 40 MPa for 4 min. The final thickness was adjusted to 0.6 mm for all cases considered. The laminates were then cut into small rectangles (12.5 mm × 4.8 mm) using a laser cutter (Versa Laser, VSL2.3, Australia).

2.2. Experimental setup and testing

The glass-ceramic laminates were placed between two metallic interconnectors (Crofer 22 APU), as shown in Fig. 1. To investigate the joint strength between the glass-ceramic sealant and the SOFC elements, various support materials were also inserted between two glass-ceramic laminates (Fig. 1). The formation of the glass-ceramic sealant was achieved after sintering between high-temperature-resistant bricks. Thirty samples were sintered at the same time to reduce the experimental errors. Thus, 30 tensile samples were prepared and tested for each case. The sintering test setup is shown in Fig. 2. The setup was designed such that the burn-out products can be safely removed without damaging the support bricks. A dead weight load was also placed on top of the tensile apparatus to improve the joining process. To distribute the load homogenously, 2-mm-thick ceramic fabric was used between the dead weight load and the samples. The effect of the magnitude of the dead weight load on the joining strength was also considered in the study. The samples were then sintered according to the heating regime given in Fig. 3 and then cooled down to room temperature at a cooling rate of 2 °C min^{-1} [22].

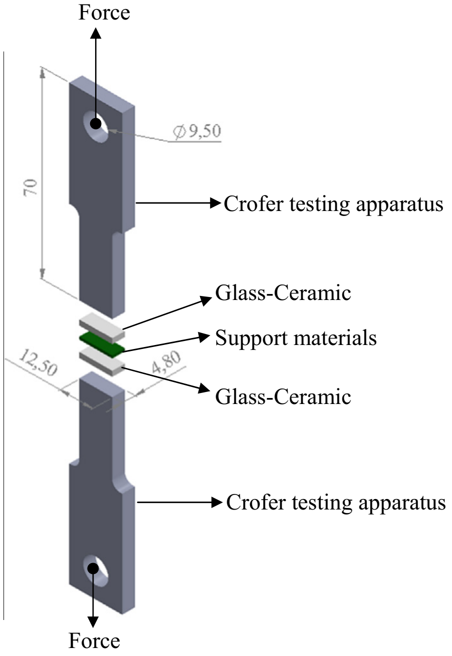

Fig. 1. Tensile testing apparatus (unit-mm).

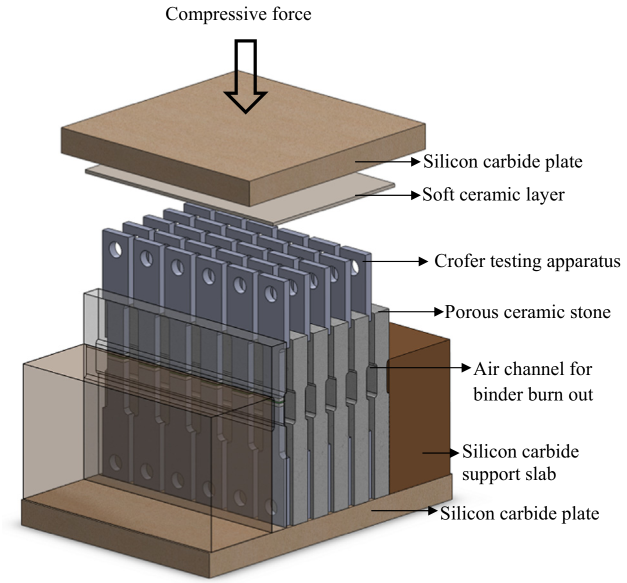

Fig. 2. Glass-ceramic bonding and sintering method with testing apparatus.

After the joining process, which took approximately 30 h, the mechanical strengths of the samples were measured via a tensile test machine (Shimadzu Autograph AG-IS, Kyoto, Japan) after placing the samples onto the hook assembly for