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Strength evaluation of glass–ceramic composites containing yttria stabilized zirconia after thermal cycling

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Abstract

The effect of YSZ (yttria stabilized zirconia) on the thermal cycling stability of commercial glass–ceramics powders as a solid oxide fuel cell (SOFC) sealant material is investigated. Four different composite sealants are prepared including 1, 3, 5 and 10 wt% of YSZ with respect to the total powders. A sealant without YSZ is also prepared as the base case for comparison. The fabrication of the sealant is achieved by using cost-effective tape casting routes. The samples are then placed between two metallic interconnectors and a high temperature glass forming process is performed. The mechanical performances are measured via tensile tests before and after a number of thermal cycles. The results indicate that the fracture strength is adversely affected by the amount of YSZ loading due to reduced glass–ceramics powders in the composites. However, when the thermal cycling stability is the issue, the samples with YSZ show relatively small rate of decrease. Therefore, the addition of YSZ is found to be beneficial for improving the thermal cycle stability of glass–ceramics. The optimum YSZ content is determined to be 5 wt%, since these samples exhibit almost a stable mechanical performance after each thermal cycles considered in this study.

Keywords: Solid oxide fuel cell; Sealant; Glass–ceramic composites; Yttria stabilized zirconia; Tensile strength

1. Introduction

In order to avoid the internal combustion of the fuel and oxidant, which may lead to the degradation of the cell and stack components because of the local hot spots, low system efficiency and low performance due to reduced fuel utilization, a high temperature hermetic sealing is required for solid oxide fuel cells (SOFCs). The expected features from a seal for SOFC include excellent sealing ability, acceptable chemical and thermal stability under both reducing and oxidation atmospheres of the stack, good wetting properties to the adjacent stack components, electrical insulation, high devitrification resistance and thermal expansion close to those of other stack elements. Seals for SOFCs can be broadly divided into three groups: compliant seals, compressive seals and rigid/bonded seals [1,2]. Glass–ceramic materials belonging to rigid/bonded seals group have been investigated by several groups [3–7] since they provide a hermetic sealing ability via the chemical bonding with the other stack components, cost effective and easy production, high electrical resistivity and adjustable properties such as the thermal expansion by simply changing the composition.

Glass–ceramics are in general multicomponent oxide structures containing network formers and modifiers, intermediates and additives [8]. The main role of the glass network formers is to form the glass network while the glass network modifiers are employed generally to modify the thermal properties including the coefficient of thermal expansion (CTE). The intermediates, on the other hand, are often used to improve the devitrification resistance and to modify the viscosity. Finally, the additives are added to adjust the desired properties although they are not required components to form glass–ceramics. The composition of these oxides can be tailored in order to obtain a suitable seal for SOFC with the desired properties such as the glass transition temperature (T_g), the glass softening temperature (T_s), CTE and the devitrification resistance.

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There can be found numerous efforts in the literature on regulating these properties by changing the composition of various glass–ceramics [9–19].

Although ZrO_2 or Y_2O_3 is often employed alone in glass–ceramic composites as an additive for adjusting the viscosity and CTE and for improving the crystallization, the adhesion and the flow ability of the composite [20–22], few studies have focused on the addition of yttria stabilized zirconia (YSZ: $(Y_2O_3)_{0.08}(ZrO_2)_{0.92}$) which is the most common SOFC electrolyte material. Brochu et al. [23,24], for instance, developed glass–ceramic composites by adding micro- and nano-scale YSZ and investigated the crystallization behavior, the flow properties and the thermal expansion of the modified sealants. The reduction of viscous flow was reported depending on the volume fraction of added YSZ powders. In addition, CTE of the composite was found to decrease a value below the acceptable limit for SOFC to be operated safely. This significant amount of decrease in CTE was attributed to the formation of $BaZrO_3$ as a result of the chemical interaction between the glass–ceramic and YSZ. They also showed that the crystallization behavior of the glass–ceramic composite is independent from the addition of nm- or μm-scale YSZ.

Gross et al. [25], who focused on the BaO–CaO–SiO_2 with various filler materials including the YSZ fibers and particles, just found the opposite. The results of the tensile strength tests carried out on circular butt joints indicated that the glass–ceramic without any filler addition provides the highest strength followed by the glass matrix with YSZ fibers and glass matric with YSZ particles. Unlike the previous studies [23,24], the loss in the mechanical strength was explained by the difference in the crystallization progress. Greven et al. [26], on the other hand, introduced multi-layer ceramic sealants concept based on again BaO–CaO–SiO_2 glass matrix. The double and triple layered sealant designs having various compositions were studied. Among them, the three layered structure composed of 20 wt% Ag reinforced layer sandwiched between two outer layers reinforced with 13 wt% YSZ was found to have the highest tensile strength value.

To the best knowledge of authors, in the relevant literature there is very little done on the addition of YSZ to the glass–ceramic sealant. Moreover, there is no comprehensive or systematic study on the thermal cycling behavior of the YSZ mixed glass–ceramic composites. Therefore, in the present study, YSZ is tested as an inhibitor for the crystallization process of glass–ceramic sealant and a reinforcement agent during thermal cycling. For this purpose, several glass–ceramic composites with various amounts of YSZ addition are fabricated via tape casting. The mechanical strengths of the samples before and after a number of thermal cycles are measured through the tensile tests by sandwiching the samples between two metallic interconnectors.

2. Experimental

2.1. Slurry preparation, tape casting and lamination

V1649 (Ceradyne Inc., Washington, USA) with 4.03 m^2/g surface area was mixed with YSZ powders ($(Y_2O_3)_{0.08}(ZrO_2)_{0.92}$) having 6–9 m^2/g surface area and 0.5–0.7 μm particle size (NexTech Materials, Ohio, USA). In order to investigate the effects of YSZ content, four different mixtures with 1–10 wt% YSZ (S05–S20) with respect to the total powders were prepared. For comparison, a tape casting slurry composed of V1649 glass–ceramic powders without YSZ addition (S00) was also prepared similarly. Alcohol based tape casting slurries of glass–ceramic composites were obtained by employing a solvent composed of a mixture of methyl ethyl ketone and ethanol (both from Sigma-Aldrich, Munich, Germany) at a weight ratio of 2:3, respectively. Next, an organic dispersant (fish oil, Sigma-Aldrich) and solvent were then added to the powders. The mixture was then ball milled for 24 h. After the addition of a plasticizer (polyethylene glycol, Sigma-Aldrich) and a binder (butvar, Sigma-Aldrich) at suitable ratios, the mixture was again subjected to another 24 h ball milling. The slurries were then tape cast on a laboratory tape casting equipment with the same blade gap of 190 μm. The detailed compositions of all slurries are listed in Table 1.

The lamination of the glass–ceramic tapes was completed after a two step pressing process. First, 15 green tapes were stacked together and laminated under 20 MPa for 4 min through a laboratory scale uniaxial press. The final lamination, on the other hand, was achieved by an isostatic press where pre-laminated glass–ceramics tapes were pressed under 50 MPa pressure for again 4 min at room temperature. The thickness of the final laminates was measured as 0.70 mm.

2.2. Mechanical characterization and joining of glass–ceramics

After completion of the lamination step, the glass–ceramic laminates were cut via laboratory scale laser cutting device into small rectangles with 4 mm × 12.5 mm outer dimensions. These sealants were then inserted between two Crofer® 22 APU plates (ThyssenKrupp VDM GmbH, Essen, Germany) which is the most widely used metallic interconnector material in SOFC applications for the tensile tests. The similar tensile measurements were also previously carried out by Lin et al. [27]. The engineering drawing of the tensile test plates having 4 mm thickness is shown in Fig. 1. A high temperature resistant SiC block was then placed on the sandwiched structures as a dead weight load (0.5 kg/cm^2) in order to improve the adhesion between the sealant and the interconnector material. This load also mimics the compressive force generally applied during the SOFC operation in order to improve the contact for better current collection. The global aspect of all test specimens after assembling is shown in Fig. 2(a). This assembled block is afterwards placed in a temperature controlled furnace. In order to improve the reliability of the tests and avoid the experimental errors

Table 1
Compositions of the tape casting slurries prepared.

Sealant name	V1649 (g)	YSZ (g)	Solvent (g)	Dispersant (g)	Binder (g)	Plasticizer (g)
S00	20	0	20	1	5	5
S01	19.8	0.2	20	1	5	5
S03	19.4	0.6	20	1	5	5
S05	19	1	20	1	5	5
S10	18	2	20	1	5	5