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Short communication

Joining of mullite ceramics with yttrium aluminosilicate glass interlayers

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Abstract

Pellets of yttrium aluminosilicate glass (Y_2O_3:Al_2O_3:SiO_2 = 30:20:50 mol%) powder were used as the filler interlayers (0.4 mm thick) to join two mullite substrates. The glass interlayer partially melted at joining temperature to bond the substrates and then crystallized during cooling to have better bonding strength. The results showed that joining could be performed at 1390–1420 °C for 1–5 h with applied pressure of 0.02 MPa. After joining, the thickness of glass layers varied between 250 μm and 80 μm, depending upon the temperatures. The glass interlayer crystallized into cristobalite, mullite and $Y_2Si_2O_7$. When joining mullite/3 mol%yttria–zirconia substrates using the same glass pellet, a layer of zircon/mullite was formed at the interface, indicating that reaction occurred between glass and substrates. The formation of zircon usually accompanied with cracks in the substrates. These cracks deteriorated the strength. The achievable three-point bending strengths were 139 MPa for joined mullite and 76 MPa for joined mullite/3 mol%yttria–zirconia.

Keywords: A. Joining; C. Strength; D. Mullite; D. Glass

1. Introduction

Mullite composites are candidate materials for structural applications because of prominent high-temperature mechanical properties. However, like most of structural ceramics, their low fracture toughness poses obstacles in fabrication as well as in industrial applications [1]. Because of the low fracture toughness, the tolerance of the size of processing flaws is very limited. Consequently, it is beneficial to fabricate parts in small sizes in order to reduce the sizes of possible flaw. Then, the small parts are assembled, joined/bonded into desired components. On the other hand, ceramic components with complicate shapes should be also fabricated by joining/bonding pieces to eliminate processing difficulties or to reduce machining cost [2,3]. Joining or bonding technology of the structural ceramics becomes a key factor in the industrial applications of these materials.

Joining between ceramics is not an easy task. Due to chemical inertness, low diffusion rate, high-melting temperatures of ceramics, joining methods prevailing in metals are usually not applicable in ceramics. Successful joining methods for ceramics include diffusing bonding, fusion welding and active interlayer joining [2,3]. So far, joining of ceramics with active interlayers appears to be most promising [4–7]. The keys to a good joining with interlayers are the good wettability and low mismatch of thermal expansion coefficients between the interlayers and the substrates. In view of these requirements, silicate-based glasses (e.g. SiO_2–Al_2O_3–MgO, SiO_2–CaO–Al_2O_3, SiO_2–Y_2O_3–Al_2O_3) seem to be the most suitable interlayer materials for the joining [8–13]. Nevertheless, the interlayer glass should crystallize to increase the strength at the joint after joining.

In this research, we attempted to join mullite and mullite/3 mol%yttria–zirconia pellets with yttria–aluminia–silicate (YAS, Y_2O_3–Al_2O_3–SiO_2) glass powder. The optimal joining conditions and the microstructures and strengths of the joint were characterized.

2. Experimental

Mullite pellets (denoted M) were fabricated by reaction sintering of powder mixtures of α-alumina (Baikolox CR15, Baikowski Corp., Charlotte, NC, U.S.A.) and amorphous silica, which were obtained by controlled gellation of tetraethyl orthosilicate (TEOS, Fluka Chemie AG, Switzerland) [14]. The ratio of α-alumina to amorphous silica for mullite was 3:1 by

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weight. In order to enhance subsequent reaction sintering of the pellets, 1 wt% of Y_2O_3 were added. The addition of Y_2O_3 was achieved by adding yttrium nitrate solution into the starting powders before mixing. Mullite pellets with 20 vol% of 3 mol%yttria–zirconia composites (denoted M20Z) were also prepared by similar procedures. The mullite/3 mol%yttria–zirconia (i.e., M20Z) pellets consisted of mixtures of α-alumina, amorphous silica and proper amount of 3 mol% yttria–zirconia.

The green compacts were obtained by uniaxial pressing with a WC die. The size of the compacts was 15 mm in diameter and ~3 mm in thickness. These green compacts were reaction-sintered at 1300–1500 °C for 2 h to form mullite or mullite/3 mol%yttria–zirconia for subsequent joining.

The glass interlayer for joining was made of yttria–alumina–silicate (YAS) glass powder, which were prepared by melting Y_2O_3, SiO_2 (Strem Chemical, Newburyport, MA, U.S.A.) and Al_2O_3 (Baikolox CR15) powder mixture in Pt crucibles at 1600 °C for 2 h and quenched on copper plates. The quenched glass were annealed and ground to pass 200 meshes and then ball-milled to obtain powders with an average particle size of 10.5 μm. The starting composition of the glass was Y_2O_3:SiO_2:Al_2O_3 = 50:30:20 by weight, which is close to the eutectic composition of Y_2O_3–SiO_2–Al_2O_3 ternary phase diagram. In order to increase the glass formability, 3 wt% of B_2O_3 was added in the YAS mixture. The glass powder was uniaxially dry-pressed to form pellets of 10 mm in diameter and 0.4 mm in thickness to be used as the interlayer between two substrates. The joining process was performed between 1390 °C and 1420 °C with a pressure of 0.02 MPa by placing a suitable weight on the top substrate during joining. The joining time varied from 1 h to 5 h. The heating rate was 10 °C/min before 1000 °C and 2 °C/min between 1000 °C and the joining temperature. After joining, the cooling rate was varied to investigate the microstructural evolution. Samples were furnace (fast) cooling to room temperature or cooling at 10 °C/min or 2 °C/min to 800 °C and then, furnace cooling.

The samples were characterized by XRD (D-5000, Siemens, Germany) for phase transformation of substrate and glass interlayer. Microstructures of the joint were observed in a SEM (JSM 5600, JEOL, Tokyo, Japan) and compositional analyses across the joining interfaces were performed with an energy dispersive spectroscopy (EDS), which was attached on the SEM. The strength of the joint was evaluated by three-point bending test of bars (4 mm × 4.8 mm × 36 mm), which were butt-joined with the joint region in the middle of the supporting fixture during tests (i.e., the joint region was subject to the maximum loading). Five bars were tested for each condition.

3. Results and discussion

3.1. Substrate and glass

The reaction sintering of M and M20Z was evaluated by sintering at various temperatures for 2 h. It turned out that M and M20Z could be sintered to full density and reacted to have the designed crystal phases after 1450 °C and 1500 °C, respectively. Fig. 1 shows the XRD patterns of these two samples after reaction sintering at the above-mentioned temperatures. The M sample consists of mullite phase with trace of unreacted alumina. The M20Z consists of mullite and zirconia. These samples already reached nearly zero open porosity.

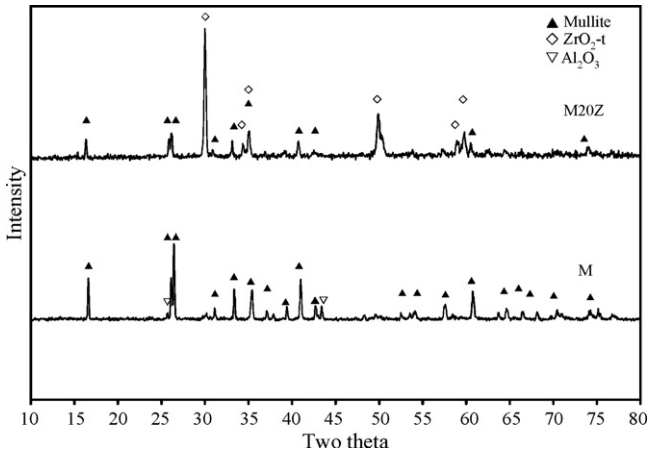

Fig. 1. XRD patterns of M and M20Z after reaction sintering at 1450 °C and 1500 °C, respectively, for 2 h.

The YAS glass was investigated for the crystallization behavior before joining. The XRD pattern of bulk YAS glass heated at 1400 °C for 2 h, which is near the joining temperature, was shown in pattern (a) in Fig. 2. The glass crystallized into mullite and yttrium silicate ($Y_2Si_2O_7$), as predicted from the phase diagram. An unidentified phase was also present. This unknown phase was likely one of the yttrium aluminate phases. The XRD patterns of the YAS interlayer after joining M sample and M20Z samples were also shown in patterns (b) and (c) in Fig. 2. It was noted that the crystallization of the glass was somewhat different once it was used as the interlayer between substrates. The glass interlayer in the joints of M samples and M20Z samples crystallized into cristobalite, mullite and yttrium silicate. The cristobalite was not found in bulk YAS glass. Furthermore, zircon ($ZrSiO_4$) was present in the joint between two M20Z substrates. This is a clear evidence of the reaction between YAS glass and the M20Z substrate.

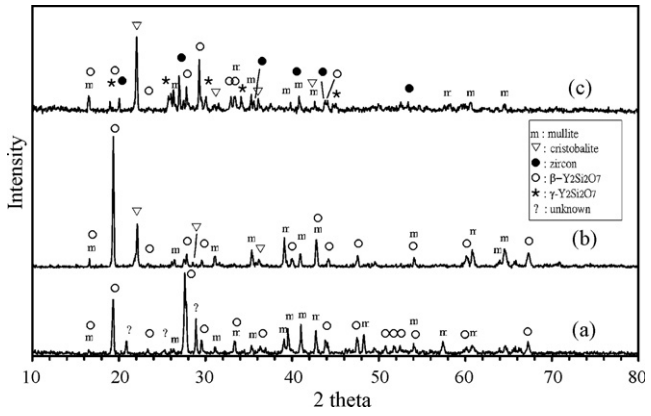

Fig. 2. XRD patterns of (a) bulk YAS glass after heated at 1400 °C for 2 h, (b) YAS glass interlayer after joining sample M, and (c) YAS glass interlayer after joining sample M20Z.

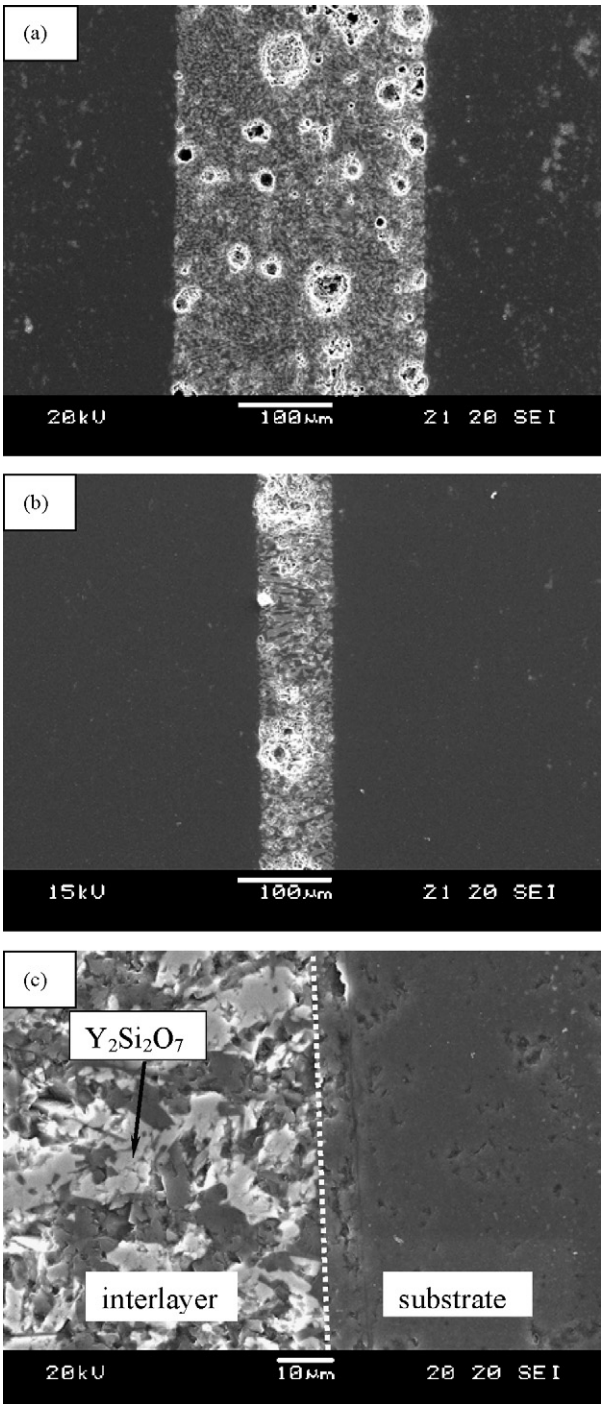

Fig. 3. SEM micrographs of the joints of sample M after joining for 2 h at (a) 1390 °C, (b) 1420 °C and (c) is a magnified view of (b) showing the microstructure at the glass/substrate interface.

3.2. Microstructure of joints

Fig. 3(a–c) shows the SEM micrographs of the joints of sample M after joining at 1390 °C and 1420 °C for 2 h. The thickness of the glass interlayer decreased from 250 μm to 80 μm as the joining temperatures increased from 1390 °C to 1420 °C. At lower joining temperatures (1390 °C) when the glass did not sinter/melt well, many pores were left. On the other hand, the glass flowed easily and drained out and caused large pores in the interlayer when the joining temperature was higher than 1420 °C. Therefore, the suitable joining temperatures were in the range of 1400–1420 °C. After joining at 1420 °C and slowly cooled (2 °C/min), the glass interlayer crystallized into cristobalite, mullite and $Y_2Si_2O_7$ as revealed from XRD analyses of the interlayer in the samples in Fig. 2. The microstructure of the interlayer is shown in Fig. 3(c), which shows bright $Y_2Si_2O_7$ and gray mullite. The interface between the interlayer and the substrate was free of reaction layers.

In contrast to the ‘‘clean’’ interfacial zone in joined sample M, YAS glass would react with M20Z substrate and forming an interfacial layer. Fig. 4(a and b) shows the microstructures of the interlayer between M20Z substrates. The interfacial layer in M20Z appeared dark in the SEM. Investigations with EDS revealed that the reaction layers consisted of mullite and zircon. This is consistent with the XRD pattern (c) in Fig. 2. Zircon was derived from reaction between 3 mol%yttria–zirconia in the M20Z substrate and SiO_2 in the YAS glass. After the depletion of zirconia, the reaction zone became regions of mullite without 3 mol%yttria–zirconia.

3.3. Crystallization processes of glass after joining

The glass melted to join the substrates at joining temperatures. After the joining, the glass crystallized during

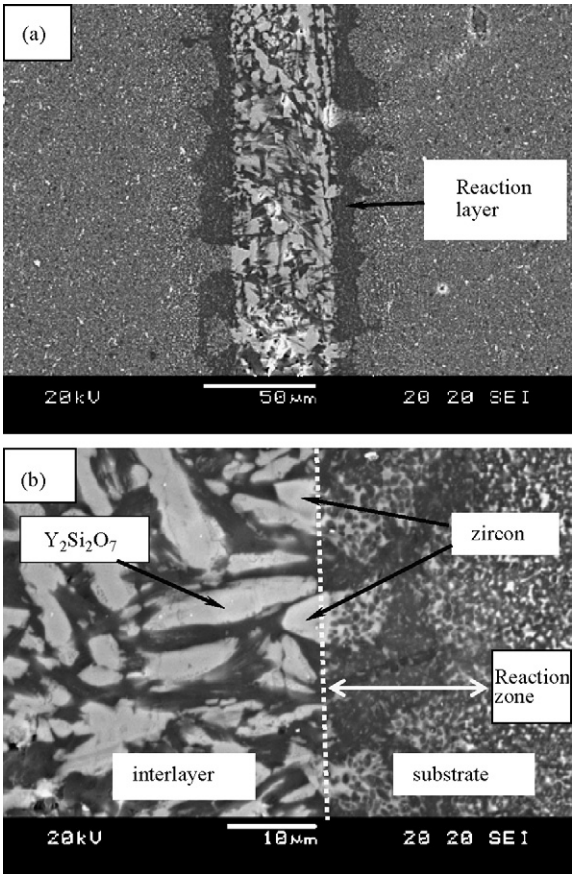

Fig. 4. SEM micrographs of the joints of sample M20Z after joining at 1420 °C for 3 h. (a) A low magnification view and (b) a higher magnification view, showing the microstructure at the glass/substrate interface.

cooling. The cooling rate would affect the crystallization of the glass. By changing the cooling rates, the crystallization sequence of the interlayer glass could be revealed. Fig. 5 (a–c) shows the microstructures of the glass interlayer cooled at different rates after joining. The fast cooling rate (furnace cooling) froze most glass. Only early crystallized phases were present. These included equiaxed cristobalite throughout the glass and elongated mullite and $Y_2Si_2O_7$ at the glass/substrate interface. Columnar mullite and $Y_2Si_2O_7$ crystallized at the interfaces and grew toward the center. Slower cooling rate resulted in coarser columnar structure and fewer equiaxed cristobalite.

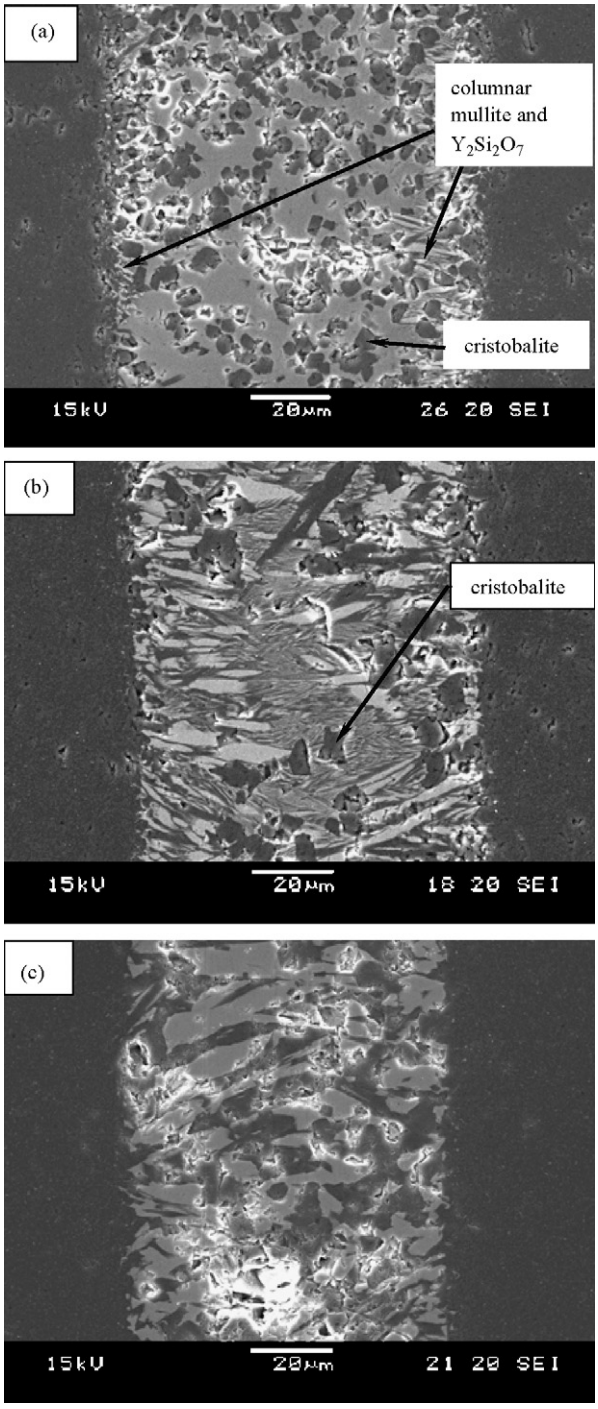

Fig. 5. Microstructures of the glass interlayer cooled at different rates after joining. (a) Furnace cooling to room temperature, (b) cooling at 10 °C/min to 800 °C followed by furnace cooling and (c) cooling at 2 °C/min to 800 °C followed by furnace cooling.

Table 1
The three-point bending strengths of the substrates and joined samples

Material	Strength (MPa)
M substrate	315 ± 40
Joint of M substrate	139 ± 19
M 20Z substrate	305 ± 27
Joint of M20Z substrate	76 ± 12

3.4. Mechanical strength

The strengths of the substrates and joined samples were evaluated with three-point bending tests. Table 1 listed the strength of each sample. After complete sintering/densification, the strength of both substrates could reach over 300 MPa. However, the strengths of joints were drastically reduced. The joint in sample M could reach 139 MPa when the sample was joined at 1410 °C for 2 h. The joint in sample M20Z was only 76 MPa. This significant reduction in the strength of the joints in sample M20Z was supposed to be due to the existence of interfacial reaction zone. In the reaction zone, zircon had relatively large thermal expansion coefficient. This could result in large residual stress near the interface and reduce the strength of the joints. In SEM cracks along the interfacial reaction zone could occasionally be observed.

4. Conclusions

Pellets of yttrium aluminosilicate glass (Y_2O_3:Al_2O_3:-SiO_2 = 30:20:50 mol%) powder were used as the filler interlayers (0.4 mm thick) to join two mullite substrates or two mullite/3 mol%yttria–zirconia substrates. The glass interlayer partially melted at joining temperature to bond the substrates and then crystallized during cooling. Appropriate joining conditions were: joining temperature at 1390–1420 °C for 1–5 h with applied pressure of 0.02 MPa. After joining, the thickness of glass layers varied between 250 μm and 80 μm, depending upon the temperatures. The glass interlayer crystallized into cristobalite, mullite and $Y_2Si_2O_7$. While the interface between the interlayer and mullite substrates were ''clean'', an interfacial reaction zone existed at the interface of interlayer and mullite/3 mol%yttria–zirconia substrates. The reaction zone consisted of zircon and mullite. The formation of zircon occasionally accompanied with cracks near the interface and deteriorated the strength. The achievable three-point bending strengths were 139 MPa for joined mullite and 76 MPa for joined mullite/3 mol%yttria–zirconia.

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