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Joining alumina using an alumina/metal composite

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Abstract

Alumina/alumina (Al_2O_3/Al_2O_3) joints and alumina/aluminium metal–matrix composite (Al_2O_3/Al–MMC) joints were fabricated using an Al_2O_3–Al composite as an interlayer. The Al_2O_3/Al_2O_3 joining procedures involved the sintering of silica or silicate glass on the surface of the Al_2O_3, followed by the reactive penetration of liquid aluminium into the sintered glass to form an Al_2O_3–Al composite interlayer between the two Al_2O_3 pieces. To join Al_2O_3 to Al–MMC, after the fabrication of the Al_2O_3–Al composite layer on the surface of Al_2O_3 using the combination of sintering and reactive penetration, Al_2O_3/composite/MMC joints were fabricated by using diffusion bonding between the Al_2O_3–Al composite layer and the MMC. The microstructures, phase composition and mechanical properties of the joints were examined using scanning electron microscopy (SEM), XRD analysis and shear testing, respectively. Experimental results showed that the high porosity in the sintered glass layer led to the formation of a porous Al_2O_3–Al composite layer between two Al_2O_3 pieces. The addition of cordierite ($2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$) to the pure silica lowered the melting point of the glass and led to the formation of a dense glass layer at the surface of the Al_2O_3. Consequently, a dense composite layer was produced. Stronger joints were fabricated with shear strengths of the Al_2O_3/Al_2O_3 and Al-MMC/Al_2O_3 joints of 105.2 and 47.6 MPa respectively.

Keywords: Al_2O_3; Composite; Joining; MMC; Reactive penetration

1. Introduction

Ceramics have excellent strength, high wear and corrosion resistance at elevated temperatures, and a variety of electrical properties. The use of ceramic components for structural, electrical and electronic applications is rapidly increasing, but difficulties in machining have hindered the in cost-effective use. Hence, most applications require joining of ceramic to ceramic, or ceramic to metal for multiple functions and reduced cost and the joining technique becomes very important. Extensive research has been carried out to develop joining techniques for these materials.

Alumina ceramics not only have excellent strength, high resistance to wear and corrosion, but also exhibit high electrical insulation. Alumina is becoming important in engineering applications as both a structural and an electrically insulating material. Extensive studies have been carried out on the joining Al_2O_3 to Al_2O_3, Al_2O_3 to metals or Al_2O_3 to metal matrix composites. Most of these joints were fabricated by using an inserted metal interlayer, such as titanium,[1] aluminium,[2,3] copper,[4,5] Al-based alloy (Al–10 wt.% Mg, Al–10 wt.% Si and Al–10 wt.% Cu),[6] Ag–Cu–Ti alloy,[7] nickel,[8] Sn-based alloy/Ni-based alloy[9] and Ag–Cu–Zr alloy.[10] Due to the difference in the coefficient of thermal expansion (CTE) between Al_2O_3 and the metal, residual stresses were induced upon cooling after joining at high temperature,[11] which would cause crack formation in the joints during the service of the ceramic components. To overcome this problem caused by the residual stress, multilayer or functionally graded materials have been used for joining ceramic to ceramic or ceramic to metal.[12,13] For example, an Al_2O_3–Ni joint with an Al_2O_3–60 vol.% Ni composite interlayer has been produced using a hot pressing process.[14] However, most of these techniques involved the application of high pressure at high temperature, which limits the application of this technique for joining a wide range of ceramic components. A reactive penetration process has been developed over many years to produce Al_2O_3–Al composites in a simple and economic way.[15–18] In this process, silica or silicate

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glass was either cold pressed or sintered to produce a preform first. Then, the molten Al penetrated and reacted with the glass preform to produce Al_2O_3/Al(Si) composites. The process is simple involving a relatively low temperature process (< 1200 °C). The physical properties of the sample such as the Young's modulus, hardness are dominated by the network structured alumina, whereas the fracture toughness is improved due to the presence of the soft Al in the composites. However, fabrication of ceramic/ceramic or ceramic/metal joints using a ceramic/metal composite interlayer produced by the reactive penetration process during the joining process has not been reported. In the present study, we developed this process to join Al_2O_3 to Al_2O_3 and to join Al_2O_3 to an Al metal matrix composite. The joints show high shear strength with little change after thermal cycling up to 500 °C.

2. Experimental detail

2.1. Materials

Fused SiO_2 powder and cordierte powder were obtained from Dytech Co-operation Ltd., UK, having particle sizes of 75 μm and with a purity of 99.9%. Al powder was obtained from Aldrich Chemical Co., USA, having an average particle size of 20 μm and with a purity of +99%. Alumina pieces (99% purity) were obtained from Pi Kem Ltd., UK. The Al–30 vol.% Al_2O_3 composite (Al–MMC) was fabricated using the pressure infiltration of molten aluminium into a porous alumina preform at 600 °C.[19] Both the alumina and Al–MMC were cut into pieces of 12×12×5 mm for joining. The surfaces of the specimens were polished using 1200-grit SiC paper and then ultrasonically cleaned in acetone before joining.

2.2. Fabrication procedure

To produce a silica preform on the surface of Al_2O_3, a layer of silica glass was applied to the surface of the Al_2O_3 using screen printing of silica paste, followed by sintering. The silica paste was made by mixing silica powder and a Blythe binder (2 g powder in 1 ml binder). The binder contained 50% terpineol and 50% ethyl cellulose (John Mathew Ltd, UK). The paste was screen printed onto a surface of an Al_2O_3 piece to produce a layer with a thickness of about 200–300 μm. After drying at 150 °C in air for 3 h, the printed layer was heated at a rate of 20 °C/min to 1500 °C, remaining at this temperature for 20 min, followed by cooling to room temperature at the same rate. The surface of the sintered silica layer was polished using 1200-grit SiC paper and then ultrasonically cleaned in acetone. It should be noted that the high heating and cooling rates were used to avoid the formation of low temperature phases of SiO_2, such as α- and β-quartz and thus to obtain a higher density of the SiO_2 layer.

Following this, an Al paste (3 g Al powder in 1ml binder) was screen printed onto the surface of the sintered silica layer on the surface of the Al_2O_3 piece, which was followed by placing another Al_2O_3 piece on the top of the wet Al paste. After drying at 120 °C in air for 5 h, the sandwiched structure (Fig. 1a) was placed in a vacuum furnace ($p = 5\times10^{-5}$ Torr) and heated to 800–1000 °C with a heating rate of 10 °C/min, kept at the maximum temperature for 30 min, and then cooled to room temperature at the rate of 10 °C/min. When the furnace temperature reached 700 °C, a pressure of 0.68 MPa was applied to the sample in order to expel the possible molten Al present at the joint.

The method for producing a dense glass preform layer between two Al_2O_3 pieces was by sintering the glass with a composition of SiO_2–4.6 mol.% cordierite. Similarly, a mixture of SiO_2–4.6 mol.% cordierite and the binder with a ratio of 2 g powder to 1 ml binder was used to produce a glass paste. The paste was applied onto the surface of an Al_2O_3 piece and then sandwiched with another Al_2O_3 piece. After drying the paste in air at 120 °C for 5 h, the Al_2O_3/glass/Al_2O_3 joint were produced by sintering the glass at 1500 °C for 20 min. The final joint was obtained by reactive penetration of molten Al into the dense glass layer between the two Al_2O_3 pieces at 1000 °C for 5 h.

To fabricate the Al–MMC/Al_2O_3–Al/Al_2O_3 joint, firstly a dense glass layer was produced on the surface of the Al_2O_3 piece using screen printing and sintering as described above. Then the Al was reactvely penetrated into the dense glass layer to form a dense Al_2O_3–Al composite on the surface of the Al_2O_3 substrate. Finally the Al_2O_3–Al composite was joined to the Al-MMC by diffusion bonding, which was carried out at 580 °C in a vacuum for 10 min with an applied pressure of 0.68 MPa.

The cross sections of the joints were characterised using scanning electron microscopy (SEM Jeol JXA-840) coupled to an energy dispersive X-ray spectrometer (EDS). The phases in the interlayer was identified using X-ray diffraction analysis (Philips PW1140). The shear fracture strength of the joints was measured using a specially designed fixture at room temperature [Fig. 1(b)]. Samples of 5.0×5.0×10.0 mm dimension were cut for the shear testing. Three repeat shear tests were carried for the joints produced under the same joining conditions.

3. Results

3.1. Al_2O_3–Al_2O_3 joining using reactive penetration of Al into the porous silica layer

Initially the joining experiments were carried out by sintering a mixture of Al and SiO_2 powder sandwiched

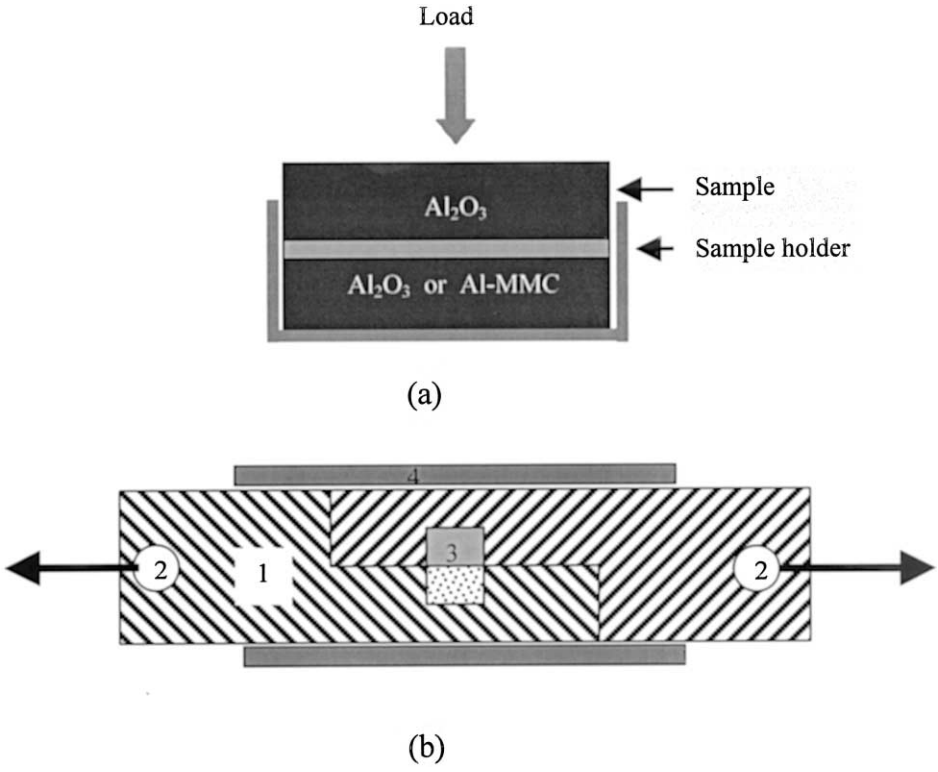

Fig. 1. (a) The sample holder and (b) shear test apparatus for the joints. The numbers shown in (b) represent: 1, for holder; 2, for bolt; 3, for sample; 4, for casing pipe.

between two Al_2O_3 pieces between 700 and 1000 °C, where the reaction occurred between the SiO_2 and Al led to the formation of very porous interlayer at these temperatures and thus no viable joint was obtained. Thereafter, sintering the SiO_2 layer on the surface of the Al_2O_3 was carried out before joining. However, it proved difficult to achieve a high density SiO_2 layer by sintering at temperatures below the melting point (1726 °C) of SiO_2, even though high heating and cooling rates were used to avoid phases transformation of SiO_2.[20] On the other hand, in order to avoid the degradation of Al_2O_3 due to its grain growth at high temperature, the SiO_2 layer at the surface of Al_2O_3 should be sintered at temperatures below 1600 °C. After sintering at 1500 °C, the porosity in the SiO_2 layer on Al_2O_3 was found to be about 20–30 vol.% measured using image analysis (Nikon light microscope with a Quantimet 520 analyser). X-ray diffraction analysis of the SiO_2 showed that the main phase in the glass layer was its high temperature cristobalite phase with a small amount of its low temperature phase, α-quartz.

With a layer of Al placed on top of the SiO_2 layer, the Al could reactively penetrate into the SiO_2 at 1000 °C in vacuum. Thus, an Al_2O_3–Al composite layer was obtained between the two Al_2O_3 pieces. Experiments showed that the reactive penetration of molten Al into the SiO_2 layer took place quickly at temperatures above 1000 °C. Fig. 2 shows cross-sections of the joints produced at (a) 850 °C and (b) 1000 °C. A layer of unreacted Al, which contains some Si precipitates, was found in both samples. At 850 °C, the reaction only occurred at the interface between the molten Al and SiO_2 without the penetration of Al into the SiO_2 [Fig. 2(a)] However, at 1000 °C, an Al_2O_3/Al(Si) composite layer was produced between the two Al_2O_3 pieces [Fig. 2(b)]. The X-ray diffraction analysis of the fracture surface for the joint produced at 1000 °C, shown in Fig. 3(a), suggested the presence of Al_2O_3, Al and Si, but no SiO_2 in the composite layer, indicating that the reaction between molten Al and SiO_2 was complete. The formation of Al_2O_3–Al composite can be described by the following reaction:

$$SiO_2 + \frac{4}{3}Al(l) = \frac{2}{3}Al_2O_3(s) + Si(s) \quad (1)$$

In this reaction, the molten Al reduces SiO_2 to produce Al_2O_3 and Si. The Si dissolved into liquid Al initially, and then precipated during cooling. Most of the Si precipitated in the Al outside the composite while a small amount of Si remained in the Al channels in the composite. This is in good agreement with the results reported previously.[16,21,22] The Si content in the unreacted Al layer outside the composite depends on the thickness of the Al and SiO_2 layers before the reaction. Due to the solubility of Si in the liquid Al–Si alloy

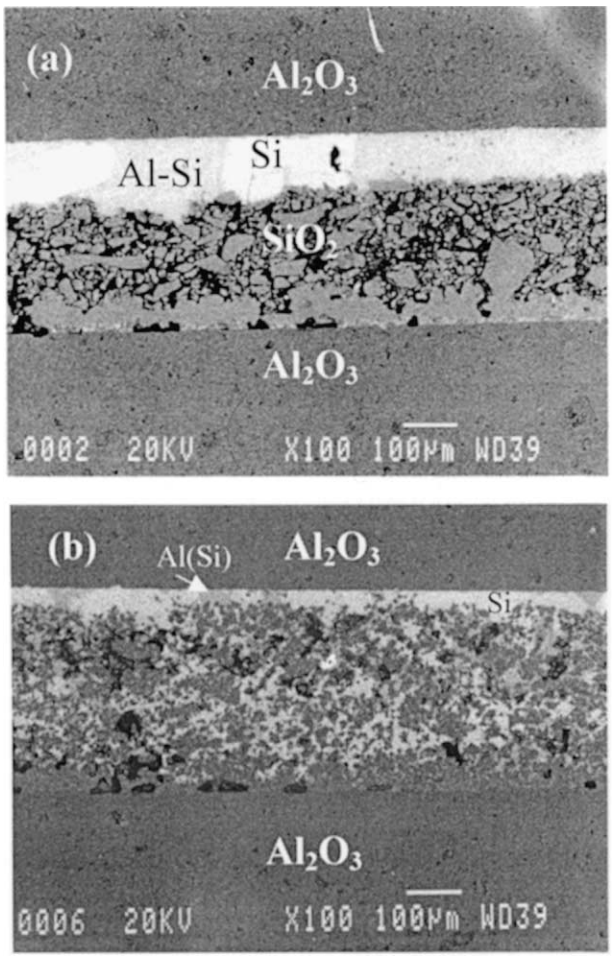

Fig. 2. Scanning electron micrographs of cross-section of the $Al_2O_3/Al–Al_2O_3/Al_2O_3$ joints fabricated from the penetration of Al into the porous SiO_2 layer at (a) 850 °C and (b) 1000 °C in a vacuum for 30 min.

at 1000 °C which is 46.0 wt.% Si and the very low solubility of Si in solid Al,[23] the Si dissolved in Al as Al alloy Al(Si) at high temperature, and then precipitated during cooling. When pressure was applied to the joint at 700 °C, the precipated Si prevented the liquid Al alloy from being expelled between the composite and Al_2O_3 (Fig. 2). As shown in Fig. 2(b), large voids in the composite layer could not be infiltrated by the molten Al during the reactive penetration process. Therefore, the formation of the composite layer was controlled mainly by the Al/SiO_2 reaction instead of by Al infiltration. In this case, the porous SiO_2 interlayer led to the formation of a porous Al_2O_2/Al composite interlayer. Therefore, the preparation of a dense SiO_2 layer on the surface of Al_2O_3 should favour the production of a dense Al_2O_3–Al composite interlayer.

Fig. 4 shows the interface between the Al_2O_3–Al composite layer and Al_2O_3 substrate, and between the Al_2O_3–Al composite layer and metal layer. The newly formed Al_2O_3 phase in the composite layer was produced based on the Al_2O_3 substrate [Fig. 4(a)]. At the interface between the metal layer and the composite layer, there no intermediate layer appeared [Fig. 4(b)]. The microstructure of the joint shows a strong bonding at these interfaces. Shear testing showed that fracture always occurred within the composite layer, (see Section 3.4). The high porosity in the Al_2O_3–Al composite interlayer led to a low shear strength of the joints.

3.2. Al_2O_3–Al_2O_3 joining using reactive penetration of Al into a dense silicate layer

In order to increase the density of the glass layer joined to the two Al_2O_3 pieces, a mixture of SiO_2–4.6

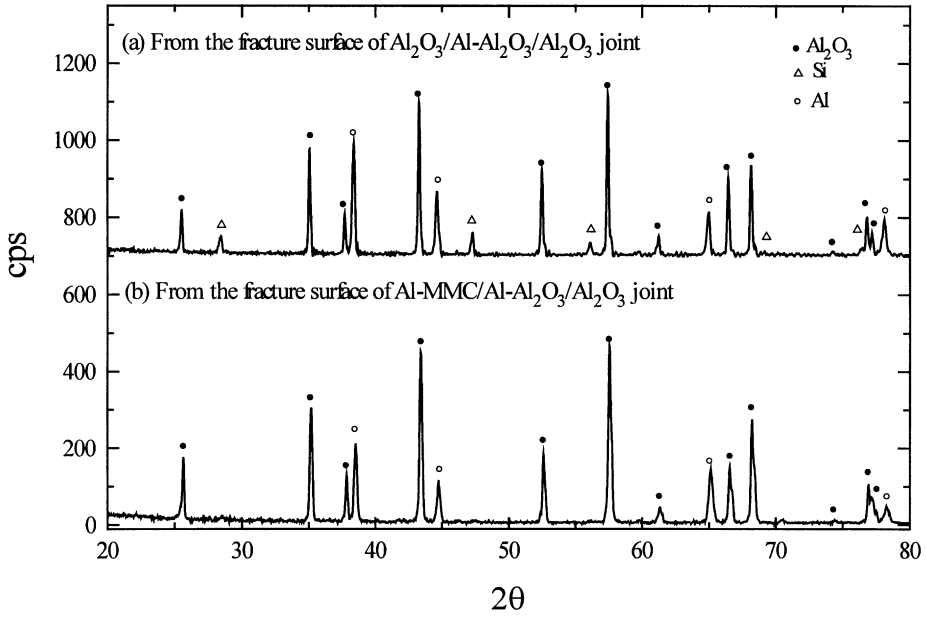

Fig. 3. X-ray diffraction analysis of the fracture surface of (a) the $Al_2O_3/Al_2O_3–Al/Al_2O_3$ and (b) Al–MMC/$Al_2O_3–Al/Al_2O_3$ joints after shear tests.

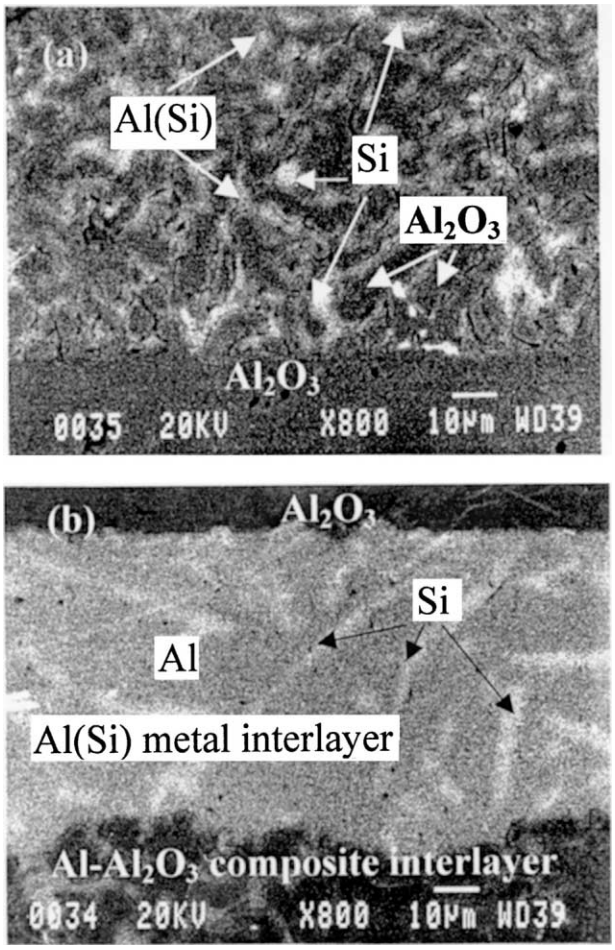

Fig. 4. Scanning electron micrographs of the interfaces (a) between the Al_2O_3–Al composite and Al_2O_3 substrate and (b) between the Al_2O_3–Al composite and metal layer.

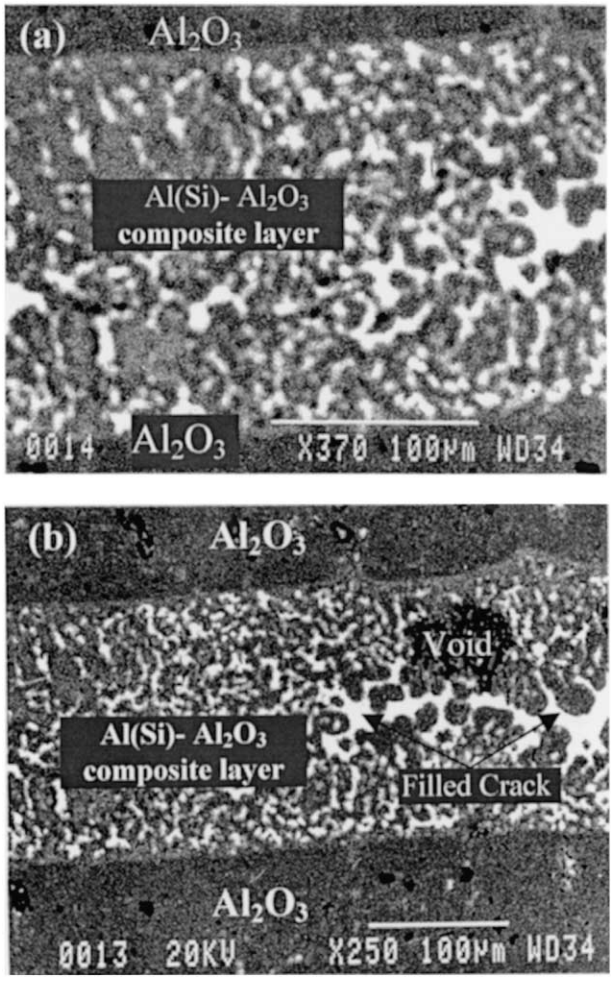

Fig. 5. Scanning electron micrographs of cross-sections of the Al_2O_3/Al_2O_3–Al/Al_2O_3 joints fabricated using a dense interlayer (a) without and (b) with a void in the joint.

mol% codierite ($2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$) powder was used as a joining agent, where the codierite melted incongruently with the lowest liquidus temperature of 1345 °C. The amount of liquid increased quite rapidly with increasing temperature above 1345 °C.[24] In this work, the silicate powder paste with a composition of SiO_2–4.6 mol% codierite was sandwiched between the two Al_2O_3 pieces and then sintered at 1500 °C for 20 min. After the sintering, a dense silicate layer joined the two Al_2O_3 pieces. These joints showed low shear strength (3.5–6.2 MPa) due to the low strength of the glass layer. Joints of this type were then immersed in a liquid Al bath at 1000 °C in a vacuum for 5 h. An Al_2O_3–Al composite layer with a high density between the two Al_2O_3 pieces was obtained after the reactive penetration of Al into the glass layer. Fig. 5 shows cross-sections of the Al_2O_3/Al_2O_3 joints with the dense Al_2O_3–Al composite interlayer. However, voids are present in some regions of the joints (Fig. 5b). The presence of large Al channels in the composite layer may be due to the formation of cracks or inhomogenety in the glass layer during the fabrication of Al_2O_3/glass/Al_2O_3 joints. There is no MgO found in the ceramic network, neither was Mg found in the metal channel according to EDS analysis. The ceramic network is pure alumina while the metal network contains mainly Al and a small amount of Si. A small amount of MgO (about 2 at.% Mg) was found to exist in the void region shown in Fig. 5(b). This may be due to poor penetration of Al into the void. The MgO should react with the molten Al to produce Mg which dissolved in the Al, but the level of Mg is too low to be detected. The formation of the voids may reduce the joint strength, but the low concentration of voids reduced the effect of the voids on the joint strength. During shear testing, fracture always occurred in the Al_2O_3 substrate, instead of in the composite interlayer. This indicates that a high strength of the composite layer has been achieved by using this dense glass interlayer.

3.3. MMC–Al_2O_3 joining using diffusion bonding

For MMC–Al_2O_3 joining, a dense glass layer was first produced on the surface of Al_2O_3 using screen printing

and sintering. Following this the reactive penetration of liquid Al into the glass was carried out to produce a dense Al_2O_3–Al composite layer on the surface of the Al_2O_3. After polishing the surface of the Al_2O_3–Al composite, the composite was joined to Al–MMC by diffusion bonding, which was carried out at 580 °C in a vacuum for 10 min with an applied pressure of 0.68 MPa.

Fig. 6 shows a cross-section of the Al_2O_3/composite/MMC joint. The composite interlayer shows the interpenetrating microstructure, which is similar to that in the Al_2O_3/Al_2O_3 joints. The microstructure shows that a good bond was achieved at the interfaces between the composite layer and the Al_2O_3 layer, and between the composite layer and the MMC layer. The bonding between Al_2O_3 layer and composite interlayer was achieved again by growing the newly formed Al_2O_3 phase based on the Al_2O_3 substrate. The Si precipitates in the Al network of the Al_2O_3–Al composite layer disappeared after joining. It is believed that the Si diffused into the MMC during the joining process. The Al_2O_3–Al composite layer and the MMC layer might be bonded due to the formation of the Al–Si eutectic liquid phase at the joining temperature of 580 °C, where a transient liquid-phase (TLP) formed at the temperature above the eutectic point of Al–Si alloy (577 °C).[23] XRD analysis of the fracture surface of this joint (Fig. 3b) confirms the presence of Al and Al_2O_3 phases in the composite layer.

3.4. Shear testing of the joints

The shear tests of the joints were carried out before and after thermal cycling. Three shear strength values for the Al_2O_3/Al_2O_3 joint with a porous composite interlayer were 68.0, 55.2 and 46.6 MPa. The failure always occurred inside the Al_2O_3–Al composite layer [Fig. 7(a)]. The low strength of the composite layer led to a low strength of the joint. The variation of the joint strength might be due to variation of homogeneity in the composite layer and some error in aligning the joint plane parallel to the shear direction.[25] With the fabrication of the dense composite layers, the three shear strength values of the Al_2O_3/Al_2O_3 joints became 127.2, 105.6 and 82.9 MPa. The shear strength of these joints are close to that for the alumina pieces, which was determined to be 135.5±20 MPa. The cracks always initiated at the edge of the Al_2O_3/composite interface, but propagated into the Al_2O_3 ceramic, leading to failure of the Al_2O_3 [Fig. 7(b)]. This indicates that both the bonding strength at the Al_2O_3/composite interface and the strength of the composite interlayer is higher than that of the Al_2O_3. Fig. 8 shows SEM micrographs of the fracture surfaces of the Al_2O_3/Al_2O_3 joints with the porous and the dense composite interlayer. The fracture surface of the porous joint is relatively flat showing the brittleness of the joint [Fig. 8(a)]. Large voids are present in this joint. The alumina network is coarser than that in the dense joints, an example of which is shown in Fig. 8(b). The fracture in the dense Al_2O_3/Al_2O_3 joints

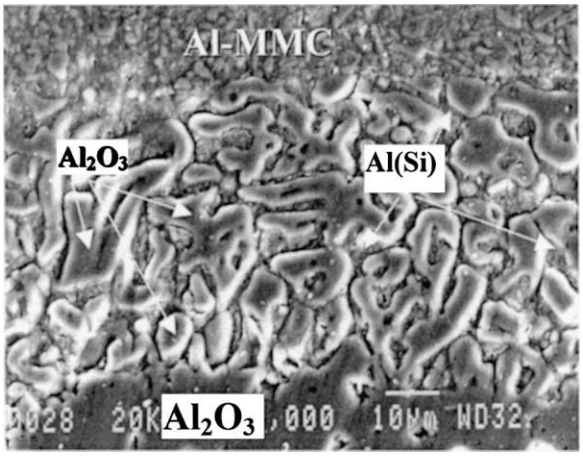

Fig. 6. Scanning electron micrograph of the cross-section of the Al–MMC/Al_2O_3–Al/Al_2O_3 joint fabricated using a dense glass interlayer.

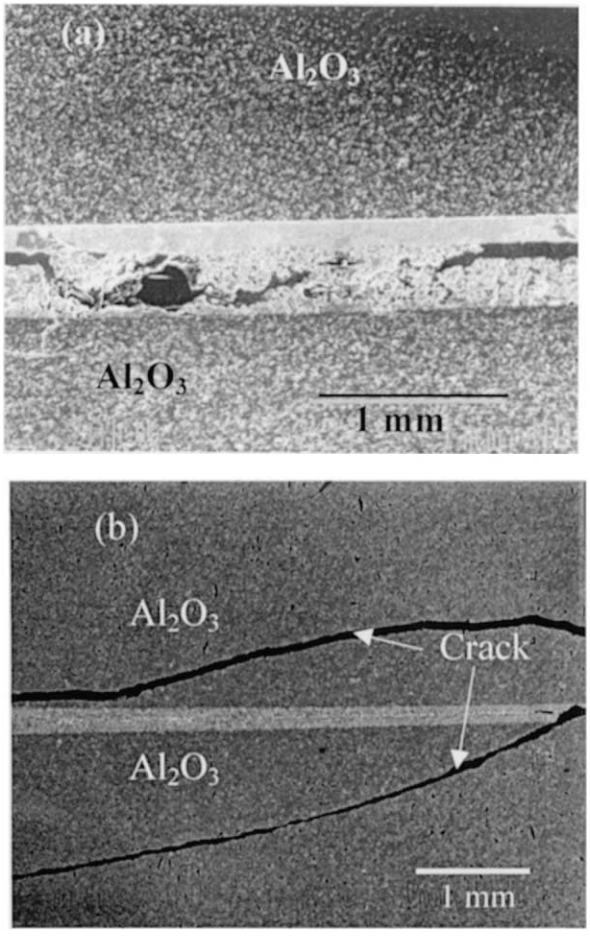

Fig. 7. Scanning electron micrographs of cross-sections of Al_2O_3/Al_2O_3–Al/Al_2O_3 joints with (a) a porous and (b) a dense composite interlayer after shear testing.

started from the interface and then propagated into the Al_2O_3 (Fig. 8b). It is well established that the maximum stress is present at the edge of the metal/ceramic interface after joining.[26] However, the newly formed composite achieved a strong bonding between the Al_2O_3 substrate and composite interlayer. The soft Al phase in the composite enhanced the toughness of the composite interlayer. Therefore the crack initiated at the edge of the interface and then propagated into the Al_2O_3 substrate. Here, the variation of shear strength might be due to the variation of the strength in the Al_2O_3 and a possible error in aligning the joint plane parallel to the shear direction.

The shear strengths of three MMC/Al_2O_3–Al/Al_2O_3 joints are 56.2, 46.9 and 39.7 MPa. There is no evidence to show segregation of Al_2O_3 particles in the MMC during joining (Fig. 9). Fracture always occurred at the interface between the MMC and the composite interlayer. The low strength of the joints may be due to the weak bond at the MMC/composite interface. The interface was formed due to the Al–Si eutectic reaction between the Al in the MMC and the Si from the composite interlayer. The flat surface after the fracture at the interface suggests that the interfacial bonding is weak (Fig. 103).

The Al_2O_3–Al_2O_3 joints with a dense composite interlayer and the MMC–Al_2O_3 joints survived the thermal cycling up to 500 °C with heating and cooling rates of 30 °C/min. The microstructure and shear strength were not significantly changed due to this thermal treatment. After the thermal cycling, the shear strength of the three Al_2O_3–Al_2O_3 joints with dense composite interlayers were 118.0, 95.6 and 83.8 MPa. The shear strengths of the MMC–Al_2O_3 joints were 55.2, 43.7 and 29.9 MPa. This indicates that the joints have good thermal shock resistance by introducing the composite interlayer. Further study needs to be carried out to examine the effect of long term thermal cycling on joint strength.

The soft Al network in the composite can blunt the propagation of the crack in the composite interlayer. The

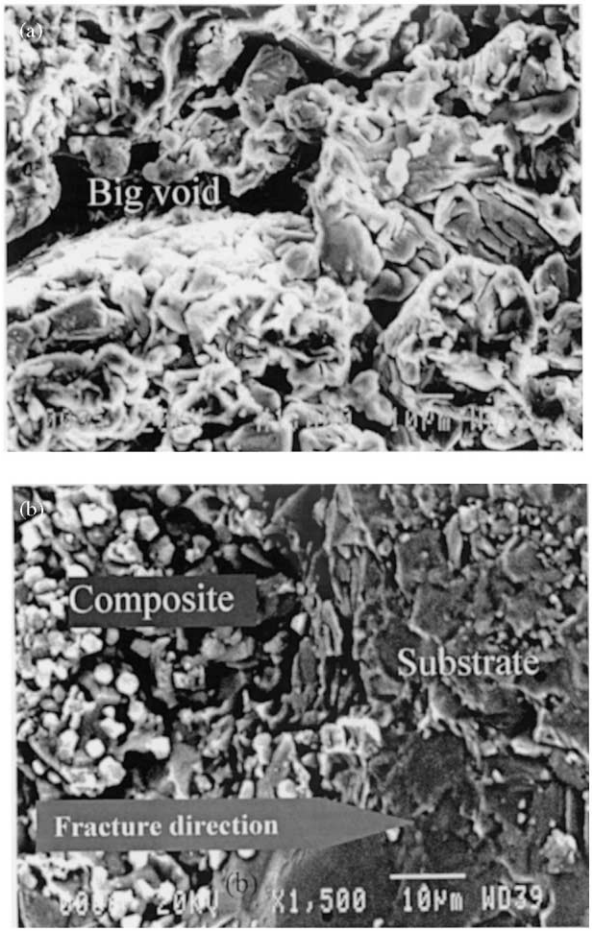

Fig. 8. Scanning electron micrographs of fracture surfaces of Al_2O_3/Al_2O_3–Al/Al_2O_3 joints with (a) a porous and (b) a dense composite interlayer after shear testing.

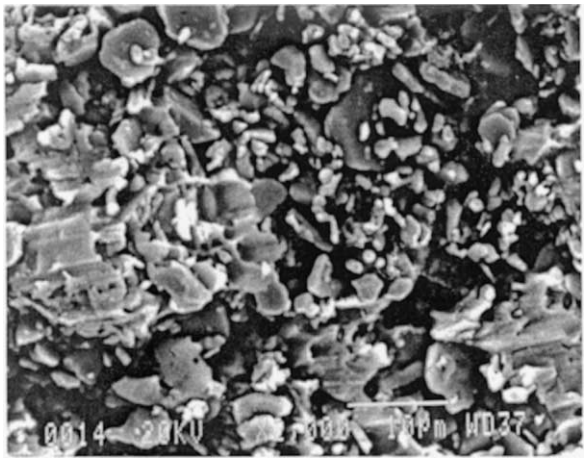

Fig. 9. Scanning electron micrograph of fracture surfaces of a Al–MMC/Al_2O_3–Al/Al_2O_3 joint with a dense composite interlayer after shear testing.

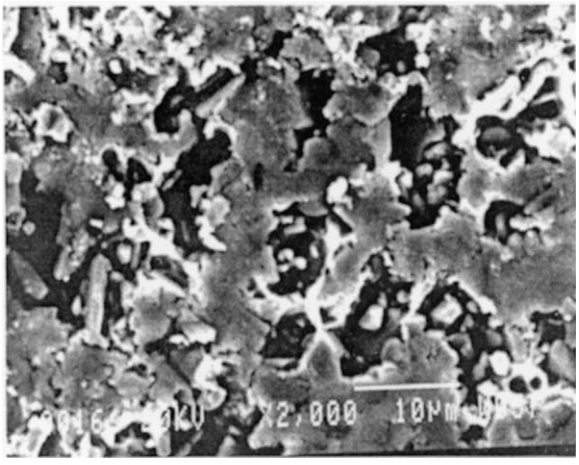

Fig. 10. Fracture surface of the Al–MMC/Al_2O_3–Al/Al_2O_3 joint showing the flat surface of the composite.

composite interlayer can also reduce the residual stresses in the joint by reducing the coefficient of thermal expansion of the interlayer compared with that of a metal interlayer. It is suggested that the stronger joint would be would be achieved with a stronger alumina substrate.

4. Discussion

Extensive research has been carried out to study the formation of ceramic–metal composites by reactive metal penetration.[15–18,21,22] The preform ceramics studied include pure silica, pure mullite and mullite-glass composites. The reaction products can be Al_2O_3/Al, Al_2O_3–Al(Si) and Al_2O_3–Al(Si)–Si composites, depending on the reaction conditions and starting materials. In the present study, the reactive penetration of molten Al into a SiO_2 layer took place at 1000 °C in vacuum and produced an Al_2O_3–Al(Si)–Si composite layer between two Al_2O_3 pieces. From the reaction described in Eq. (1), the volume fraction of Al_2O_3 in the Al_2O_3–Al composite layer can be calculated, assuming that the preform layer has full density. The weight of Al_2O_3 (W_2) formed from the reaction between SiO_2 (W_1) and Al should be $W_2 = 2W_1M_2/3M_1$, where M_1 (60.09) and M_2 (101.98) are the molecular masses of SiO_2 and Al_2O_3 respectively. The volumes for the SiO_2 (W_1) and Al_2O_3 (W_2) are $V_1 = W_1/\rho_1$ and $V_2 = W_2/\rho_2$ respectively, where ρ_1 (2.15) and ρ_2 (3.97) are the densities for SiO_2 and Al_2O_3 respectively. Assuming the volume of the composite product is same as that of the preform based on the results reported in,[15] the volume fraction of Al_2O_3 in the composite should be:

$$V(Al_2O_3) = \frac{V_2}{V_1} = \frac{W_2\rho_1}{W_1\rho_2} = \frac{2M_2\rho_1}{3M_1\rho_2} = \frac{2\times 101.98\times 2.15}{3\times 60.09\times 3.97} = 0.613$$

Although the SiO_2–4.6 mol.% cordierite mixture contains MgO, the molar fraction of SiO_2 is 98.0 mol.%. Therefore the volume fraction of Al_2O_3 in the composite interlayer produced from this perform should be close to 61.3%.

The ceramic network in the composite controls many of its physical properties such as hardness, modulus, and expansion coefficient, while the interpenetrating ductile Al phase contributes to an improvement in composite fracture toughness. The composite fracture toughness increases with increasing Al content.[27] The use of a composite layer in the Al_2O_3–Al_2O_3 and Al_2O_3–MMC joints should reduce the residual stresses in the joints, compared with joints which use a metal interlayer. The composite containing Al should have a coefficient of thermal expansion (CTE) between the CTE of Al_2O_3 and CTE of many metals.[28] Such an interlayer is especially beneficial for joining Al_2O_3 to MMC by in producing a functionally graded structure between the Al_2O_3 and Al–MMC. In addition, this reactive penetration technique may be used to produce Ni_3Al–Al_2O_3 and Ti_3Al–Al_2O_3 composite layers at the ceramic-metal joints for high temperature applications.

The best shear strength of the Al_2O_3/Al_2O_3 joint achieved was 127.2 MPa, which is comparable to the maximum shear strength for the joint of Al_2O_3/Al_2O_3 (170 MPa) by using an inserted Ag–Cu–Ti alloy foil.[7] It is suggested that the shear strength of the joint produced in this study can be enhanced by using stronger alumina pieces and by further optimising the fabrication conditions. The shear strengths of the Al_2O_3/MMC and MMC/MMC joints with inserted Cu interlayer reported by Zhai and co-worker[9,29] are from 10 to 70 MPa, depended on the thickness of the Cu foils used (5–30 μm). These joints were formed using to the eutectic reaction between Cu and Al. Thicker Cu foil lead to segregation of ceramic particles and resulted in a lowering of the strength. In the present study, no segregation has been found in the Al–MMC/Al_2O_3 joints. The lower strength (47.6 MPa) of the Al–MMC/Al_2O_3 joint in this work may be due to the weak bonding between the composite layer and the MMC, which is caused by the uneven distribution of the Si phase in the metal channels of the Al_2O_3–Al composite interlayer. This might be improved by inserting a Cu or Al–Si foil between the Al–MMC and Al_2O_3–Al composite.

5. Conclusions

A reactive penetration technique has been used to produce an Al_2O_3–Al composite interlayer between Al_2O_3 and Al_2O_3, and between Al_2O_3 and Al–Al_2O_3 metal matrix composite, where Al_2O_3/Al–Al_2O_3/Al_2O_3 joints and Al–MMC/Al–Al_2O_3/Al_2O_3 joints were produced. The production of strong joints required the fabrication of a dense glass layer on the surface of Al_2O_3, which was produced by sintering a SiO_2–4.6 mol.% cordierite glass layer at 1500 °C. The reactive penetration of Al into the dense glass layer took place in a vacuum at 1000 °C to produce an Al_2O_3–Al composite. The shear strength of the Al_2O_3/Al–Al_2O_3/Al_2O_3 joints and Al–MMC/Al–Al_2O_3/Al_2O_3 joints, produced using this method, were 105.2 ± 22 and 47.6 ± 8.6 MPa respectively.

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References

1. Kato, H., Imai, M. and Yoshikawa, K., Influence of welding conditions and residual-stresses on strength of Al_2O_3/Ti diffusion welds. *Mater. Sci. Technol.*, 1990, **6**, 641.
2. Watanabe, A., Kato, H. and Yoshikawa, K., Properties of alumina/Al/alumina joints produced in air atmosphere. *Mater. Sci. Technol.*, 1993, **9**, 155.
3. Kato, H. and Chen, Y., Influence of pressure on bonding of alumina with molten aluminium as insert. *Mater. Sci. Technol.*, 1995, **11**, 1287.
4. Urena, A., Gomez de Salazar, J. M. and Quinones, J., Diffusion bonding of alumina to steel using soft copper interlayer. *J. Mater. Sci.*, 1992, **27**, 599.
5. Zhai, Y., North, T. H. and Serrato Rodrigues, J., Transient liquid-phase bonding of alumina and metal matrix composite base materials. *J. Mater. Sci.*, 1997, **32**(6), 1393.
6. Kobashi, M., Ninomiya, T., Kanetake, N. and Choh, T., Effect of alloying elements in the brazing sheet on the bonding strength between Al_2O_3 and aluminum. *Scripta Mater.*, 1996, **34**, 415.
7. Hao, H., Jin, Z. and Wang, X., The influence of brazing condition on joint strength in Al_2O_3/Al_2O_3 bonding. *J. Mater. Sci.*, 1994, **29**, 5041.
8. Esposito, L., Bellosi, A., Guicciardi, S. and De Portu, G., Solid state bonding of Al_2O_3 with Cu, Ni and Fe: characteristics and properties. *J. Mater. Sci.*, 1998, **33**, 1827.
9. Zhai, Y. and North, T. H., Transient liquid-phase insert metal bonding of Al_2O_3 and AISI 304 stainless steel. *J. Mater. Sci.*, 1997, **32**, 1399.
10. Yoo, Y. C., Kim, J. H. and Park, K., Microstructure and bond strength of Ni–Cr steel/Al_2O_3 joints brazed with Ag–Cu–Zr alloys containing Sn or Al. *Mater. Sci. Technol.*, 1999, **15**, 1331.
11. Nicholas, M. G., Mortimer, D. A., Jones, L. M. and Crispin, R. M., Some observation on the wetting and bonding of nitride ceramics. *J. Mater. Sci.*, 1990, **25**, 2679.
12. Holt, J. B., Koizumi, M., Hirai, T. and Munir, Z. A., ed., *Ceramic Transations,* Vol. 34*, Functionally Gradient Materials*. American Ceramic Society, Westerville, OH, 1993.
13. Ilschner, B. and Cherradi, N., ed., *Proceedings of the 3rd International Symposium on Structure and Functionally Gradient Materials*. Presses polytechniques et Universitaires Romandes, Lausanne, Swizerland, 1995.
14. Rabin, B. H., Williamson, R. L. and Bruck, H. A., Residual strains in an Al_2O_3–Ni joint bonded with a composite interlayer: Experimental measurements and FEM analyses. *J. Am. Ceram. Soc.*, 1998, **81**, 1541.
15. Liu, W. and Koster, U., Fabrication of ceramic/metal composites by reduction of glass SiO_2 preforms in molten metals and alloys. *J. Mater. Sci. Letters*, 1996, **15**, 2188.
16. Saiz, E. and Tomsia, A. P., Kinetics of metal-ceramic composite formation by reactive penetration of silicates with molten aluminum. *J. Am. Ceram. Soc.*, 1998, **81**(9), 2381.
17. Yoshikawa, N., Funahashi, S., Kikuchi, A. and Taniguchi, S., Variations in processing, microstructure and properties of Al/Al_2O_3 composites fabricated by displacement reaction. *Int. J. Mater. Prod. Technol.*, 2001, **16**, 32.
18. Yoshikawa, N., Funahashi, S., Taniguchi, S. and Kikuchi, A., Fabrication and mechanical properties of Al/Al_2O_3 composite bodies by reactive infiltration of molten Al into SiO_2 preform. *J. Mater. Res.*, 2000, **15**(11), 2314.
19. Zhou, Z., Peng, H. X., Fan, Z. and Li, D. X., MMCs with controlled non-uniform distribution of submicrometre Al_2O_3 particles in 6061 aluminium alloy matrix. *Mater. Sci. Technol.*, 2000, **16**(7–8), 908.
20. Hlavac, J., *The Technology of Glass and Ceramics*. Elsevier, New York, 1983.
21. Loehman, R. E., Ewsuk, K. and Tomsia, A. P., Synthesis of Al_2O_3–Al composites by reactive metal penetration. *J. Am. Ceram. Soc.*, 1996, **79**(1), 27.
22. Gao, Y., Jia, J., Loehman, R. E., Ewsuk, K. G. and Fahrenholtz, W. G., Microstructure and composition of Al–Al_2O_3 composites made by reactive metal penetration. *J. Mater. Sci.*, 1996, **31**(15), 4025.
23. Smithells, C. J., *Metals Reference Book*, 5th edn. Butterworth, London, 1976.
24. Kingery, W. D., Bowen, H. K. and Uhlmann, D. R., *Introduction to Ceramics*, 2nd edn. John Wiley, New York, 1976.
25. Durov, A. V., Kostjuk, B. D., Shevchenko, A. V. and Naidich, Y. V., Joining of zirconia to metal with Cu–Ga–Ti and Cu–Sn–Pb–Ti fillers. *Mater. Sci. Eng. A*, 2000, **290**(1–2), 186.
26. Elssner, G., Bond strength of ceramic-metal joints. In *Joining of Ceramic*, ed. M. G. Nicholas. Chapman and Hall, London, 1990, pp. 137.
27. Loehman, R. E., Ewsuk, K. G., Fahrenholtz, W. F. and Lakshman, B. B., Ceramic-metal composite formation by reactive metal penetration. *Key Eng. Mater.*, 1997, **127**(Part 1 and 2), 431.
28. Ewsuk, K. G., Glass, S. J., Loehman, R. E., Tomsia, A. P. and Fahrenholtz, W. G., Microstructure and properties of Al_2O_3–Al(Si) and Al_2O_3–Al(Si)–Si composites formed by in situ reaction of Al with aluminosilicate ceramics. *Metall. Mater. Trans. A*, 1996, **27**(8), 2122.
29. Zhai, Y. and North, T. H., Counteracting particulate segregation during transient liquid-phase bonding of MMC–MMC and Al_2O_3–MMC joints. *J. Mater. Sci.*, 1997, **32**, 5571.