MULTILAYER DIFFUSION BONDING OF POLYCRYSTALLINE Al₂O₃ TO Al ALLOYS CONTAINING Sr, Mg, AND Si

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Abstract— Multilayered ceramic-metal composites were produced by solid state diffusion bonding of polycrystalline alumina and three Al alloys containing 1 to 2% Sr, 2% Mg and Mg + Si (5057 Al alloy). The multilayered structures were fabricated at 610°C under a compressive stress of 3 MPa for 2 hours in He-5%H₂ atmosphere. The ceramic-metal interfaces were studied by SEM and TEM equipped with an energy dispersive spectrometer (EDS). Addition of 1 wt % Sr to Al promoted good adhesion, without producing any interfacial products, while the Al-2wt%Sr was poor in this regard due to the presence of dispersed Al₄Sr, which is an intrinsic phase in the Al-Sr phase system. The Al-2%Mg alloy produced a continuous film at the interface, which was characterized as being MgAl₂O₄. Al alloys containing Mg and Si (5057 commercial alloy) produced not only the spinel structure (MgAl₂O₄), but another phase rich in Mg and Si (close to Mg₂Si). These latter interfacial reaction products were not the intrinsic phases in Al-Mg alloys, but were reaction products at the Al₂O₃—Al alloy interfaces.

Keywords— Multilayered, diffusion bonding, interface, composite, alumina/Al

1. INTRODUCTION

Diffusion bonding of layered structures is a solid state process which can be utilized to connect the following materials [1]:

a) two similar materials such as metals [2-4]
b) two similar materials with the aid of an interlayer [5-6]
c) two dissimilar materials such as metal-ceramic [7-12]
d) two dissimilar materials with a different interlayer [13-16].

Among these, bonding at metal ceramic interfaces has been the subject of many theoretical [17-21] and experimental investigations during the last three decades [7-12].

During diffusion bonding several physical and metallurgical processes occur simultaneously, such as creep, diffusion, dislocation formation, recrystallization, and formation of reaction products at the interface. Physical and chemical behavior of the interfaces may be one of the types shown in Fig. 1, [19]. Bonding of the types b through e are commonly formed at the metal/ceramic interfaces depending on the materials used.

In design of a composite it is desirable to have a strong interface with proportional distribution of load between the components, while the integrity of the composite is preserved. In the ideal case, the perfect interface should be a mechanical continuum, involving coherency of the bond on the atomic level, and a chemical discontinuum, requiring the absence of any interdiffusion between the constituents. In practice, the nature of the interface in metal/ceramic composites can be categorized into three groups. First, the

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metal and ceramic phases are thermodynamically stable at the processing and service temperature and conditions, and no reaction occurs between the two components (Fig. 1, b and f). In the second group, the metal and ceramic phases are not thermodynamically stable, forming one or more reaction products at or near the interface (Fig. 1, c, d, and e). There are many examples of bond strength domination by localized chemical reactions at the interface [22-24]. In the third group, the metal and ceramic phases are separated by a thin amorphous film which is stable during the processing of the alloy [24-26]. These films may restrict formation of reaction products at the interface. The third category can be viewed as the first group containing an interlayer between the metal and ceramic which exists prior, or is produced during the composite processing.

Fig. 1. Interfacial bonding mechanisms: a) molecular entanglement, b) electrostatic attraction, c) interdiffusion of elements without any interfacial reaction, d) chemical reaction between groups A on one surface and a compatible group B on the other surface, e) chemical bonding by formation of a new compound at the interface, f) mechanical interlocking

In this work multilayered Al/Al$_2$O$_3$ composites were produced and their interfaces were studied. Polycrystalline alumina wafers were solid bonded at 610°C to four aluminum alloys. This was different from other works which used liquid Al and/or vapor deposition (CVD) to produce layered Al/Al$_2$O$_3$ composites [27-30], and the solid bonded composites at temperatures above 645°C which were close to the melting point of aluminum [31, 34].

2. EXPERIMENTAL PROCEDURES

Diffusion bonding of two materials can be achieved by applying a pressure (normally less than half the yield stress) and at a temperature well above half the melting point (in Kelvin) of the material with the lowest melting point [19].

In this work, diffusion bonding of polycrystalline Al$_2$O$_3$ to some Al alloys was carried out in a hot press. Al$_2$O$_3$ wafers, 99.6% pure, both sides polished, 25×25×0.1±0.01 mm were ordered through Kyocera
Ceramic Corp. Al-%1 Sr and 2%Sr were produced in an arc furnace using an Al-%10Sr master ingot which was available commercially. An Al-2%Mg alloy was also prepared by arc melter. In addition, a sheet of 5052 Al alloy, which is a wrought Al-Si-Mg alloy, was purchased. These Al alloys were rolled to ≈ 150 μm, and the wafers were blanked to accommodate the sandwich structure of alternative layers of Al₂O₃ and one of the Al alloys. Al wafers were ultrasonically cleaned in acetone, soaked in a dilute HF-H₂O solution and cleaned in methanol prior to hot pressing.

A compression die was designed and machined from graphite (Fig. 2). The die cavity was 27×27 mm² to allow some lateral flow of Al under hot pressing.

Diffusion bonding was conducted at 10⁻⁵ torr and under various loads (1-3 tons), within a temperature range of 550-650°C for 30 to 120 minutes. Most of the samples did not bond and failed due to oxidation of Al wafers. To avoid oxidation of Al wafers, the die assembly was evacuated (10⁻⁵ torr) and then a mixture of He-%5H₂ was purged in the die before and during hot pressing. This approach at 610°C, 2 tons (~3 MPa) and 2 hours resulted in excellent adhesion of the Al-Al₂O₃ interfaces.

The thickness of the layered composite before hot pressing was ≈ 5.2 mm, and after hot pressing reduced to 4.6 mm. Further lateral flow of the metallic layers was prevented by the graphite mold.

To examine the quality of the interface adhesion, test samples 4.6×4 mm² in cross section were cut by a diamond disk for 3 point bend test. When the load was applied perpendicular to the interfaces in the laminate configuration, Al₂O₃/Al-%1Sr and Al₂O₃/5052 Al alloy composites failed by Al₂O₃ breakage without interface separation, while the Al₂O₃/Al-%2Sr and Al₂O₃/Al-2%Mg composites failed at a lower load by Al₂O₃ breakage and an appreciable amount of interface separation which could be observed by the naked eye. These effects are discussed in the next section.

Metallographic examination of the interfaces were carried out by SEM and TEM equipped with EDS. SEM samples were prepared by polishing the composites on 600 and 1000 grit emery, followed by 6 and 1 μm diamond paste. TEM samples were cut by a diamond disk perpendicular to the interfaces to a thickness of 1 mm, then embedded in crystal bond and mechanically polished to 50 μm thickness. 3 mm diameter disks were then core drilled from the 50 μm thick sheets, dimple ground from both sides to a thickness of ≈ 10 μm, and finally thinned to 1000-1500Å using an ion beam thinning machine. A major problem in TEM sample preparation was the preferential polishing and thinning of Al layers compared with Al₂O₃. This was partially solved by placing an Al₂O₃ layer in the center of the 3 mm TEM disk during core drilling and also by using sector milling during ion milling to sputter the central Al₂O₃ layer for a longer time than the two adjacent Al layers, resulting in a uniform thin central area.

3. RESULTS AND DISCUSSION

The flexural strength of bonded Al₂O₃/Al-1Sr and Al₂O₃/5052 Al multi-layer samples were ~380 MPa and 298 MPa respectively (averaged on 3 tests). Both of the above composites failed due to the fracture of the
Al₂O₃ wafers and without any apparent debonding at the metal-ceramic interface. However, Al₂O₃/Al-2 Sr samples failed at the metal-ceramic interface under relative low loads compared to the above values, while Al₂O₃/Al-2 Mg samples withstood slightly higher loads with a mixed failure mode (i.e. simultaneous debonding and fracture of Al₂O₃ wafers).

The interfacial bond strength of Al₂O₃/Al-1Sr and Al₂O₃/5052 Al were excellent based on the above results and that of Al₂O₃/Al-2Mg was adequate to withstand the harsh TEM sample preparation, especially during core drilling and dimple grinding of the 3mm TEM samples from both sides. The Al₂O₃/Al-2Sr samples disintegrated during rough polishing and slice cutting for the TEM sample preparation. These behaviors were explored and explained by metallographic examination of the interfaces in these four composites.

Figure 3 is a SEM micrograph of the Al₂O₃/Al-1Sr composite after rough polishing on 600 grit emery paper. This shows a typical acceptable appearance of the hot pressed layered composites.

![SEM micrograph of Al₂O₃/Al-1Sr composite](image1)

**Fig. 3. SEM micrograph of Al₂O₃/Al-%1Sr composite**

Figure 4a shows the same sample after fine polishing and at a higher magnification which demonstrates an interface free of void and cavity. It also shows that the alumina wafers are not fully dense and flat, which is an advantage for mechanical interlocking. A random distribution of white particles was observed inside the metallic layer and also at the interface (Fig. 4b).

![SEM micrographs of Al₂O₃/Al-%1Sr sample](image2)

**Fig. 4. a) Interface free of void of Al₂O₃/Al%-1Sr sample, b) Al₆Sr particles in the Al matrix and at the interface**
TEM observation of the same sample revealed that these particles are Al$_4$Sr, which is the intrinsic phase in the Al-Sr phase system. This phase is tetragonal with D$_{13}$ space group. Figures 5a and 5b show an Al$_4$Sr particle and its diffraction pattern in the Al matrix, while Figs. 6a and 6b show a particle and its selected area diffraction (SAD) at the interface.

A comparison of the SAD patterns of 5b and 6b reveals that the particles have the same identity and are the intrinsic phase in the binary Al-Sr system, not an interfacial reaction product. The only difference between these particles is that those inside the metallic matrix are rod shaped, while those on the interface are irregular. The SAD pattern of 6b also shows some degree of texture. It seems the particles at the interface are affected by the mismatched stresses at the interface. Nevertheless, the adhesion of Al$_2$O$_3$/Al-1Sr is almost perfect. It is in good agreement with other works (24) indicating that Sr promotes adhesion by forming an amorphous film on the interface.

Al$_2$O$_3$/Al-2Sr layered composite was hot pressed, but produced cracked interfaces (Fig. 7). This was due to the higher amount of Al$_4$Sr in the Al-Sr layer and its frequent presence at the interface. The higher %Sr made the Al-2Sr alloy more brittle to the extent that it cracked during the initial rolling process of thin sheets prior to the bonding stage.
To examine the effect of Mg, two types of Al alloys, Al-2%Mg and 5052 Al alloy (2.5 Mg-0.25Si-0.4 Fe-0.25 Cr-0.1 Cu) were selected, and their layered composites were produced. In both alloys, thin localized reaction products were produced at the interface with Al₂O₃. Fig. 8 shows the EDS X-ray map of an interface in Al₂O₃/Al-2Mg composite. This interface film was almost continuous and covered the whole interface rich in Mg.

5052 Al alloy contains not only Mg, but Si, Cu, Fe and Cr which are present in a variety of precipitates in the Al matrix (layer) of the composite (Fig. 9). Some of these precipitates are numbered on the TEM micrograph and their EDS spot analysis are shown in Figs. 10a, b, and c.

Fig. 7. SEM micrograph of Al₂O₃/Al-2% Sr showing cracks at the interface

Fig. 8. X-ray map of Al₂O₃/Al-2Mg composites showing Mg rich film at the interface

Fig. 9. TEM micrograph of 5052 Al alloy showing several precipitates
Fig. 10. a) EDS point analysis of particle 1 in Fig. 9

Fig. 10. b) EDS point analysis of particle 2 in Fig. 9

Fig. 10. c) EDS point analysis of particle 3 in Fig. 9
Figure 11 demonstrates a precipitate at the interface of Al2O3/5052 Al composite and its corresponding SAD pattern. Two types of interfacial films were identified. The corresponding SAD patterns and the EDS spectrums suggest that they are Mg2Si and MgAl2O4, Figs. 12 and 13.

Comparison of Figs. 12 and 13 with Fig. 10 reveals that none of these phases are identical, and the former were produced during hot pressing at the interface. The high peaks of Mg and Si spectrum of Figs. 12 and 13 show a migration of Mg (a strong reducing agent) and Si to the interface which produced Mg and Si rich film, and nucleated Mg2Si and the spinel MgAl2O4 phases. The spinel phase was previously observed in other interfaces such as Al-Mg alloy with Al2O3 fibers (24). The flexural test of this layered composite, explained earlier, showed an excellent adhesion of the interface containing these films, though it was less than the bond strength of Al2O3/Al-1Sr layered composites.
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Fig. 13. EDS point analysis of another area of the interface in Al₂O₃/5052 Al composite

4. CONCLUSION

1- Various multi-layer Al₂O₃/Al composites were produced by solid state bonding process.
2- The composite samples containing Al-1%Sr and 5052 Al alloy had higher bond strength than those made using Al-2%Sr and Al-2%Mg alloys.
3- The interfacial films between Al₂O₃, Al-2%Mg and 5052 Al alloys were different from the existing phases in both alloys. It was concluded that these reaction layers were merely produced by the segregation of some of Mg and Si in these Al alloys to the interface. Other elements such as Cr, Fe, Cu and Ca in the 5052 Al alloy were not observed in the interfacial films.

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