Interfacial Characteristics for Brazing of Aluminum Matrix Composites with Al-12Si Filler Metals

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Discussions concerning the interfacial reactions and characterizations in brazing aluminum matrix composites are documented in this study. Joints of alumina particulate reinforced 6061 aluminum matrix composites were made using an Al-12 (wt pct) Si filler metal by vacuum brazing. The resulted maximum bonding strengths were 75.4, 81.5, and 71.8 MPa for 10, 15, and 20 vol pct alumina reinforcement, respectively. The microstructural examinations revealed that the bonding strength was strictly related to the reinforced alumina particles and the reaction products presented at the joint interfaces. During brazing, Mg segregated at the joining interface and alumina/6061 Al interface. Further, reactions between alumina and 6061 Al matrix resulted in the formation of Mg-rich phases, such as $MgAl_2O_4$ and MgO, near the joining interface and the alumina reinforcement. The Si in the filler material penetrated into the metal matrix composites (MMCs) matrix and segregated at the alumina/6061 Al interfaces. This phenomenon can be confirmed by a joint between two alumina bulk specimens.

I. INTRODUCTION

METAL matrix composites (MMCs) have received considerable attention as candidates for advanced industrial applications as structural materials. They are attractive materials for airframe and spacecraft structures because of their high specific strength, stiffness, thermal stability, and wear resistance.^[1] Discontinuously reinforced MMCs generally consist of dispersed ceramic reinforcement of a hard phase in metal or alloy matrix. Several types of discontinuously reinforced MMC materials are available in stock form, such as billet, rod, and tube, suitable for various secondary fabrication operations. For large potential industrial applications, such as fabricating practical complex engineering components from MMCs, appropriate bonding techniques are required. One of the most important functions of MMC joining techniques is to provide the means for economic fabrication of complex, multicomponent structures. In a previous study by Kennedy,^[2] the available capabilities of the fusion welding methods for joining MMCs are described. However, the melted zone and heat affected zone formed in the MMC matrix might cause undesirable reactions, which may be observed at the joints when fusion welding methods are used.

In the realm of metal joining, brazing technology has been the subject of extensive research over the years. There may be some interdiffusion during this process, but it usually does not change the composition significantly. On the other hand, the brazing process provides a simple means for bonding large joint areas and fabrication of complex assemblies. Therefore, the brazing method has been widely used in preference to other techniques for the large-scale joining of aluminum parts, such as automobile heat exchangers and air-conditioning condensers.^[3]

For brazing monolithic aluminum alloys, Al-12Si has

been popularly used as the filler metal, and a satisfactory bonding has been reported. However, such a prime result does not necessarily occur for the brazing of aluminum matrix composites due to the existence of ceramic reinforcement. Tillmann and Lugscheider^[4] indicated that some alloys, such as Cd-Ag, Zn-Ag, Cd-Zn, and Zn-Al, can be used as the filler metal for joining aluminum matrix composites. Suganuma et al.[5] have shown the feasibility of brazing alumina short-fiber-reinforced 6061 aluminum alloy with Al-Si filler metal and Al-Mn filler metal coated with thin Al-Si layers. The joint strength with such an Al-Si/Al-Mn/Al-Si filler metal was found to be substantially higher than that with Al-Si filler metal. Robertson et al.^[6] have shown that the soldered boron-fiber-reinforced aluminum can reach 75 MPa average shear strength. However, the interfacial phenomena correlated with the brazing mechanisms and brazing strength have seldom been discussed in the previous works.

In addition, it has been known that during brazing, the wettability of filler metal on ceramic-reinforced phase is the first issue that should be considered. Some alloying elements such as magnesium, copper, lithium, silicon, and nickel in the filler metals have been shown to possess a tendency to segregate at the joining interface, which can enhance the wettability of these filler metals.^[7] Another factor is the arrangement of reinforcements along the bonded interface. Because of the inert nature of the particulate, the particulate-particulate bonds will be weak and reduce the overall joining strength.^[5,8]

The effort in this study is concerned with the relationship between the interfacial characteristics and the bonding strength for the brazing of 10, 15, and 20 vol pct alumina-particulate-reinforced 6061 aluminum matrix composites by using a Al-12 wt pct Si filler metal.

II. EXPERIMENTAL

The MMC materials selected for this work were the 6061 aluminum alloy reinforced with 10, 15, and 20 vol pct of α -phase alumina particulates. The average particle size was

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Manuscript submitted April 4, 1995.

Table I. Compositions and Mechanical Properties of 6061 Aluminum Alloy and Aluminum Matrix Composites in This Study

	Compositions										
Material	Element ($\times 10^{-2}$ Wt Pct)							Mechanical Properties			
	Si	Fe	Cu	Mn	Mg	Cr	Zn	Ti	Al	Tensile strength	Vicker's Hardness
6061	43	31	31	5	94	13	20	3	bal	303	55
10 vol pct MMC	57	4	24	1	105	15	4	2	bal	342	58
15 vol pct MMC	48	5	22	1	90	14	5	1	bal	362	60
20 vol pct MMC	69	4	20	1	100	15	5	1	bal	353	65



Fig. 1—Optical graph of alumina-particle reinforced 6061 aluminum matrix composites.



Fig. 2—The schematic illustration of the setup of the wetting angle test.

11.2 μ m. The chemical composition and the mechanical properties of the MMCs and the 6061 aluminum alloy matrix are shown in Table I. The as-cast ingots were extruded into rods with a diameter of 22 mm and cut into rods of 50-mm length. Both joining surfaces were polished to an arithmetical average roughness height (R_a) of 0.19 μ m. The composites contain a nominally homogeneous distribution of alumina particulates, although there are a few clustered regions. The composites also have an excellent bonding between the alumina particulates and the aluminum alloy ma-The alumina trix (Figure 1). specimens, for alumina/alumina joints, were made of A16-SG powder from Alcoa Co. (Alcoa Center, PA). The bulk alumina ceramic was fabricated by pressure slurry casting and sintered at 1600 °C for 2 hours.



G: Gage length	30.00±0.06mm
D : Diameter	6.00±0.10mm
R : Radius of fillet, mm	6mm
A: Length of reduced section, mm	36mm

Fig. 3-Dimensions of the tensile test specimen.

The Al-12 pct Si commercial foil with 0.025-mm thickness was inserted between MMCs. The filler material possesses a tensile strength of 100 MPa and a hardness of HV 40.1. All specimens were cleaned ultrasonically in acetone before the brazing treatment. The brazing was conducted in a vacuum furnace of 1×10^{-3} torr. After the furnace was heated to the test temperature and held there for 20 minutes, the joined specimens were cooled in the furnace. The brazing temperature was between 570 °C and 640 °C.

In order to evaluate the wettability of the Al-12 wt pct Si filler metal on the alumina ceramic, monolithic 6061 alloy and 6061 aluminum matrix composite surfaces, the sessile drop method was used to measure the contact angles. A furnace with a viewing window was used to obtain *in situ* wetting-angle measurements of the sessile drops (Figure 2).

The strength of the joint was measured by tensile test, which was performed with a constant crosshead speed of 2.4×10^{-4} s⁻¹ at room temperature. The tensile test specimen prepared from a brazing joint was machined to the dimensions given in Figure 3 (ASTM B557M-81). In order to ascertain reproducibility, at least three measurements were typically made for each type of sample.

Aging behavior of the brazed joints was also investigated by means of a Vicker's microhardness test. The specimen of the microhardness test was made using the Al-Si filler with 0.1-mm thickness.

Microscopic examination was performed using a scanning electron microscope (SEM). Also, the existing phases were identified using an X-ray diffractometer and an electron probe for microanalyses (EPMA).

III. RESULTS AND DISCUSSION

A. Bonding Strength of Brazed Joints

Figure 4 shows that the strength increased with increasing temperature from 570 $^{\circ}$ C to 610 $^{\circ}$ C and then decreased



Fig. 4—Temperature dependence of the bonding strength for 6061 Al-MMCs with (a) 10 vol. pct, (b) 15 vol. pct, and (c) 20 vol. pct particle alumina.

as the temperature moved beyond 610 °C. It shows that the bonding strength of the 15 pct-MMC joints is superior to that of the 10 pct- and 20 pct-MMC joints at temperatures from 590 °C to 630 °C. The maximum bonding strength of the 15 pct-MMC joints can reach 81.5 MPa at 610 °C, 20 minutes. Table II shows the average ultimate tensile stress level of the joints at room temperature for the brazing temperature 610 °C and time 20 minutes. For comparison, the tensile strength of the original base MMC materials and monolithic 6061 Al alloy are also given in Table I. This table shows that the tensile strength of the original base MMC also increased with increasing the volume fraction

Material	Bonding Strength (MPa)	Bonding Efficiency (Pct)	Vicker's Hardness (100 g Load)
6061 Al alloy joint	167.7	55.4	53
10 vol pct MMC joint	75.4	22.1	50
15 vol pct MMC joint	81.5	22.5	61
20 vol pct MMC joint	71.8	20.3	64

of particles and the maximum strength reached between 15 and 20 pct volume fraction of particles. Raghunathan *et al.*^[9] indicated that the existence of a critical value of the particle volume fraction for MMCs is due to the overlapping of stress-strain field between reinforced particles.

Due to the mutual interlock between the reinforcements and metal matrix, adding the reinforcements into the aluminum matrix may increase the mechanical strength of the MMCs. Therefore, the strength of the original base 10 pct-MMC is lower than the original base 15 pct-MMC and the original base 20 pct-MMC (Table I). However, the strength of the original base 20 pct-MMC is still lower than the original base 15 pct-MMC. This could be attributed to the voids and particle clustering regions increasing with increasing the volume fraction of particles.^[9]

On the contrary, it is interesting to note that the bonding strengths of the 10 pct- and 15 pct-MMC joint specimens are better than that of the 20 pct-MMC joint. This might be attributed to the particle density. The arrangement of the reinforcements aligned along the joint surface also affects the bonding strength of the MMC joint.^[8] During brazing, the alumina particles in the MMC may prevent metal-metal contact. Therefore, the bonding strength is restricted in the presence of alumina particles. This effect becomes more predominant with increasing the volume fraction of alumina. Comparison with the bonding strength of monolithic 6061 Al alloy jointed with Al-12Si filler metal shows that the 167.7 MPa strength is higher than those obtained by MMC joints (Table II). In order to clarify the reason, an alumina/alumina specimen jointed with Al-12Si filler metal at 610 °C for 20 minutes is to be designed. The average bonding strength of the alumina/alumina joint specimen is only 21.5 MPa. Obviously, the MMC bonding strength depends mainly on the joint contact of the 6061 alloy matrix/filler. Figure 5 shows that the voids occurred along the alumina particle surface at the bonding interface. This result demonstrates that the particle surfaces aligned in the planar bonding interface are potential defect sites and regions in the MMC joints. At the same time, the voids make the joint weak.^[8] Thus, if the filler metal can wet the MMC very well, the bonding strength of the MMC joints will increase. Figure 6 shows that the contact angles decrease with the increase of temperature. It can be seen that the Al-12Si filler metal wet the MMC substrate at temperatures above 610 °C for 20 minutes but that the contact angle remained about 70 deg as the temperature was further increased. Figure 7 shows that the Al-12Si filler metal also wet the monolithic 6061 alloy substrate at about 610 °C for 20 minutes. On the other hand, Figure 8 shows that the Al-12Si filler metal cannot wet the alumina substrate at temperatures from 590





Fig. 5—The voids occurred at the bonding interface for 20 pct aluminareinforced 6061 aluminum composite after brazing at 610 °C for 20 min (*a*) low-magnification view of joint. (*b*) higher-magnification of the area *A* in (a).

°C to 700 °C. Therefore, the wetting process occurs mainly through metal matrix when joining MMC.

Figure 9 shows that the fracture surface of the joint parts exhibited the features of ductile fracture. Because the strength of the MMC materials is stronger than the bonding strength of the joint interfaces, most of the joints fracture at the joint interface.

Furthermore, the joint interfacial region should be free of any brittle intermetallic phases. These attributes are salient points that the joint interfacial region will be able to effectively transfer any applied static or dynamic load from filler material to MMCs without realizing excessive microcrack evolution. These reaction products will be discussed later in this article.

B. Microstructures of the Brazed Joints

The EPMA observation (Figures 10(a), (d), and (g)) of the joint region revealed an irregular interface between the



Fig. 6—The contact angle test of the Al-12Si filler metal on MMC substrate.



Fig. 7—The contact angle test of the Al-12Si filler metal on monolithic 6061 alloy substrate.

filler metal and MMCs. Such an irregular interface might be caused by the partial melting of the MMC matrix accompanied by the melting of Al-Si filler metal during brazing treatment. The distribution maps of Mg and Si showed aggregations coinciding with each other in the intermediate layer (Figures 10(b), (e), and 10(h)). It should be noted that the Mg distribution in 20 pct-MMC sample is richer than that in 10 pct- and 15 pct-MMC (Figures 10(c), (f), and (i)).

From the point of view of the chemical reaction at the joint region, limited alloying is a necessary step during the brazing treatment. Figure 11(a) illustrates that the depth of penetration of the Al-12Si into the 6061 aluminum alloy near the joint interface was not uniform. Figures 11(b) and



Fig. 8—The contact angle test of the Al-12Si filler metal on alumina substrate.

(c) revealed that the Si and Mg elements still clustered along the joint interface. However, the elemental distributions of the MMC joints shown in Figures 10(b), (e), and (h) indicated that the Al-12Si alloy penetrated deeply into the composites with little distribution of the Si element. Suganuma et al.^[5] have shown that the eutectic aluminum brazing filler metal used penetrated into the aluminum base metal. However, the penetration mode of the Al-12Si filler metal into the aluminum composite differs from that into the unreinforced matrix alloy. Suganuma et al. indicated that the result can be attributed to the microchannels, which were formed in the processing history of the MMCs. On the other hand, when Si diffuses from the filler metal into the MMC, it seems to cause melting of the matrix. Sabathier et al.^[14] also indicated that the diffusion path of the monolithic alloys is significantly less than that of the MMCs.

In the present work, when Mg diffused in contact with Al₂O₃ particulate reinforcement, a layer of compound formed at the reinforcement-matrix interface, which prevented further diffusion of Mg. The joint interfaces also impeded Mg diffusion. This is why 20 pct-MMC revealed a higher distribution of Mg map than 10 pct-MMC (Figures 10(c), (f), and (i)). The Si contents in the original base MMC material and in the filler material are 0.48 to 0.69 wt pct and 12 wt pct, respectively. At the brazing temperature, the filler material melts and Si in the filler material probably diffuses rapidly into the MMC matrix; thus, the distribution of Si is not prominent in the mapping. This result coincided with Dutta's result that less Si clustered with the reinforcement addition in the MMC.^[12] However, the Al₂O₃ reinforcements in the MMC may present an obstacle when Si is diffusing. A simulated joint specimen is to be designed to reveal this phenomenon. The interfacial microstructure of the simulated specimen is shown in Figure 12. It can be seen that large quantities of Si remain in the filler metal. Therefore, it can be stated that Si diffuses under the influence of Al₂O₃ reinforcement in the MMC.



Fig. 9—SEM image of the typical fracture surfaces for the joints of aluminum matrix composites with varied reinforcement fraction after brazing at 610 °C for 20 min: (a) 10 pct, (b) 15 pct, (c) 20 pct.



Fig. 10—(*a*) EPMA line scanning and K_{α} X-ray mapping of (*b*) Si and (*c*) Mg near the joining region for 10 pct alumina-reinforced 6061 aluminum composite after brazing at 610 °C for 20 min. (*d*) EPMA line scanning and K_{α} X-ray mapping of (*e*) Si and (*f*) Mg near the joining region for 15 pct alumina-reinforced 6061 aluminum composite after brazing at 610 °C for 20 min. (*g*) EPMA line scanning and K_{α} X-ray mapping of (*h*) Si and (*i*) Mg near the joining region for 20 pct alumina-reinforced 6061 aluminum composite after brazing at 610 °C for 20 min.

C. X-Ray Analyses

Typical X-ray diffraction patterns from the fractured surfaces showed peaks originating from MgAl₂O₄, Mg₂SiO₄, Al_2O_3 , and SiO_2 (Figure 13). The peaks of Si were absent in the diffraction patterns from the fractured surfaces of the MMC joints, but the peaks from SiO_2 were found. No ad-







Fig. 10 Continued—(a) EPMA line scanning and K_{α} X-ray mapping of (b) Si and (c) Mg near the joining region for 10 pct alumina-reinforced 6061 aluminum composite after brazing at 610 °C for 20 min. (d) EPMA line scanning and K_{α} X-ray mapping of (e) Si and (f) Mg near the joining region for 15 pct alumina-reinforced 6061 aluminum composite after brazing at 610 °C for 20 min. (g) EPMA line scanning and K_{α} X-ray mapping of (h) Si and (i) Mg near the joining region for 20 pct alumina-reinforced 6061 aluminum composite after brazing at 610 °C for 20 min.

ditional diffraction peak existed that might be assigned to other Mg-Al or Mg-Si compounds.

The products identified at the interphases could result from the following chemical reactions:

(a) Si + O ₂ \rightarrow SiO ₂	$\Delta G^{0} = -214,760 + 41 T cal$
(b) $\text{SiO}_{2(s)}$ + 4/3 Al \rightarrow 2/3 Al ₂ O ₃ + Si _(s)	$\Delta G^{0} = -53,000 + 11 T cal$
(c) $\text{SiO}_{2(s)} + 2 \text{ Mg} \rightarrow 2 \text{ MgO}_{(s)} + \text{Si}_{(s)}$	$\Delta G^{0} = -69,000 + 31 T cal$

From the preceding expressions for Gibb's energies of reaction, we may say that the silicon is first oxidized to SiO_2 under the vacuum of 10^{-3} and that the reduction of SiO₂ may be accomplished either by magnesium or by aluminum during the brazing treatment. After the following reactions have occurred, MgAl₂O₄ and Mg₂SiO₄ form. The negative ΔG° value for the formation of reaction products indicates that these are stable phases at the brazing temperature (T = 580 °C to 640 °C).

(d) $2SiO_{2(s)} + Mg + 2 AL \rightarrow MgAL_2O_{4(s)} + 2Si_{(s)}$ (e) $3Mg_{(s)} + Al_2O_{3(s)} \rightarrow 3MgO_{(s)} + 2Al_{(s)}$ (f) $3Mg_{(s)} + 4Al_2O_{3(s)} \rightarrow 3MgAl_2O_{4(s)} + 2Al_{(s)}$ (g) $\operatorname{MgO}_{(s)} + \operatorname{Al}_2O_{3(s)} \rightarrow \operatorname{MgAl}_2O_{4(s)}$ (h) $\operatorname{2MgO}_{(s)} + \operatorname{SiO}_{2(s)} \rightarrow \operatorname{Mg2SiO}_{4(s)}$

Reaction (f) is believed to be the most likely mechanism for the formation of the MgAl₂O₄ layer at the interface.^[15] Generally, the joint bonding strength was presumed to be the sum of the chemical bonding strength at the interface and the mechanical interlocking strength of the interface. The reaction between the MMC and the filler metal occurred strongly, and the contribution of chemical bonding to the joint strength was greater than that of mechanical interlocking. The bonding strength depended mainly on the properties of the reaction products. When the brazing temperature was too low and the holding time too short, the interfacial reaction was insufficient. On the contrary, the interface might increase the amounts of the brittle product. The bonding strengths of both states degenerate. In this investigation, we discovered that 610 °C and 20 minutes are the optimum brazing parameters.

D. Heat Treatment of the Brazed Joints

Another factor that affects the composites is precipitation. Many studies of the heat-treatment response of MMCs have been reported,^[10] and, in general, an enhancement of the aging kinetics has been observed. Solute segregation associated with interfaces may be present in the composite.[11] Dutta et al.[12] and Suresh et al.[13] have indicated that increasing the volume fraction of reinforcements will increase the dislocation densities in the matrix. The higher dislocation density in composites could also increase the solute diffusivity, and the enhanced diffusivity of Mg atoms has been suggested as the reason for the higher growth rate of precipitates in 6061-Al₂O₃ composites.^[12] During brazing, magnesium diffused into the reinforcement at the Al₂O₃-Al interface by pipe diffusion along the dislocation and formed MgAl₂O₄, which resulted in Mg depletion from the matrix.

Dutta et al.^[12] has confirmed that Al₂O₂-reinforced aluminum-matrix composite and aluminum alloy have the same precipitation sequence. The precipitation sequence is



(a)



(b)



(c)

Fig. 11—(a) SEM image and K_{α} X-ray mapping of (b) Si and (c) Mg near the joining region for 6061 aluminum alloy joint after brazing at 610 °C for 20 min.

documented as follows: spherical GP zones $\rightarrow \beta$ " needles $\rightarrow \beta$ ' rods $\rightarrow \beta$ -Mg₂Si plates, and the amount of silicon clustering was found to decrease with increasing reinforcement addition.

In this present article, because the brazing temperature is close to the melting temperature of the 6061 aluminum matrix, the precipitation in the 6061 aluminum matrix may be resolved. In order to understand the effect of precipitation





Fig. 12—(a) EPMA line scanning and K_{α} X-ray mapping of (b) Si for joint of monolithic alumina bulk using Al-12(wt pct)Si filler metal (brazing at 610 °C for 20 min).



Fig. 13—The typical X-ray diffraction pattern from the fracture surface of the joined 6061 MMC (10, 15, and 20 pct joints after brazing at temperatures from 580 °C to 640 °C for 20 min).

in the MMC and MMC joints, the brazed joints are aged at 180 °C, 20 hours after the brazing process is conducted.



Fig. 14—Temperature dependence of the bonding strength for 6061 Al-MMCs joints after the aging treatment.



Fig. 15—A schematic of the specimen geometry associated with the Vicker's hardness test.

The joint temperature dependence of the tensile properties of these joints is shown in Figure 14. The bonding strength of the 10 pct reinforcement joints after aging treatment is superior to that of the 10 pct reinforcement joints without aging treatment. On the other hand, the strength of the 15 and 20 pct reinforcement joints after aging treatment is almost the same as that for joints without aging treatment. This may be attributed to the fact that the MMC strengthened aging occurs chiefly in the metal matrix. Therefore, the heat treatment may obviously increase the bonding strength of the lower alumina content MMC joints.

Some studies on hardness measurements of particulatereinforced 6061 aluminum MMCs were performed in the literature.^[16,17] In these studies, the matrix strength has been considered a primary parameter influencing the strength of composites. In our case of MMC joints, the different joining parameters may introduce further degrees of complication. Figure 15 displays the specimen geometry associated with the Vicker's hardness test. Table III shows the Vicker's hardness value across the joint interface. The hardness of region B (filler metal region) in the MMC joint is lower than that of the original unbrazed filler metal. It may be attributed to the fact that the Si element diffuses into the MMC matrix. The Vicker's hardness of the 10 pct

Table III.	Vicker's Hardness of the Brazed Joints (Brazing
	Temperature 610 °C for 20 minutes)

		The Average Hardness (HV) Load 100 g		
Treatment	Materials	A Region	<i>B</i> Region	C Region
Joint without aging after brazing	joint of 6061 Al alloy joint of 10 pct MMC joint of 15 pct MMC joint of 20 pct MMC	46.6 52.1 60.5 63.6	42.5 40.1 38.7 38.6	45.3 51.8 61.1 64.7
Joint with aging after brazing	joint of 6061 Al alloy joint of 10 pct MMC joint of 15 pct MMC joint of 20 pct MMC	51.0 59.0 60.2 62.1	35.0 35.9 38.6 41.5	52.0 58.7 58.7 65.0

reinforcement joints after aging treatment is superior to that of the 10 pct reinforcement joints without aging treatment. This result is concurrent with the tensile strength result. However, the Vicker's hardness values of the 15 and 20 pct reinforcement joints after aging treatment are almost the same as those for joints without aging treatment.

IV. CONCLUSIONS

- 1. The joint pair of MMC and MMC was fabricated using Al-12Si foil as an insert material. For the three tested materials, 10, 15, and 20 vol pct alumina-reinforced 6061 composites, the 15 vol pct particle-alumina-reinforced composites show the highest bonding strength. The ideal fabricating conditions are a brazing temperature of 610 °C for a period of 20 minutes. The joints have the average bonding strength level of 81.5 MPa.
- 2. The elemental distributions of the joint region have shown that the joint interface and the alumina particles both present obstacles when the elements Mg and Si are diffusing. A probable reaction controlling factor is the transportation of reaction elements through the materials and the reaction products.

- 3. The X-ray diffraction measurements indicated that MgO, SiO₂, Mg₂SiO₄, and MgAl₂O₄ are formed at the joining interface.
- 4. The bonding strength of the joints were influenced both by particle-particle and patricle-matrix interactions at the bonded interfaces and by the products of the reaction between filler material and MMCs on plastic deformation in tension.
- 5. The lower alumina content MMC joints increased their bonding strength after the aging treatment.

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