

# Counteraction of particulate segregation during transient liquid-phase bonding of aluminium-based MMC material

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Particle segregation during transient liquid-phase bonding of aluminium-based metal matrix composite material using copper filler metal was investigated. Segregation was promoted by the slow movement of the solid–liquid interface during isothermal solidification and alumina particles with diameters less than 30  $\mu\text{m}$  were segregated when the copper foil thickness exceeded 5 and 15  $\mu\text{m}$  for the base metals examined. When bonding at 853 K, the liquid widths produced using these copper foil thicknesses were almost identical to the median inter-particle spacing in the base metals investigated. When the amount of liquid formed at the bonding temperature decreased below a critical level, the test specimens broke apart immediately following the joining operation. The minimum film thickness of copper for satisfactory joint strength increased from 0.6  $\mu\text{m}$  to 2.4  $\mu\text{m}$ , when the heating rate to the bonding temperature decreased from 1  $\text{K s}^{-1}$  to 0.01  $\text{K s}^{-1}$ .

## 1. Introduction

Transient liquid-phase (TLP) bonding comprises a number of distinct stages—base metal dissolution following melting of the filler metal, isothermal solidification and homogenization. TLP bonding has been generally applied for the repair of nickel-based superalloy turbine blade components, because the final joint has mechanical properties similar to that of the base metal. However, particulate segregation has been observed during TLP bonding of aluminium-based metal matrix composite material containing alumina and silicon carbide [1, 2], particle-reinforced copper [3], and when joining yttria-bearing oxide dispersion strengthened (ODS) alloys such as MA956 [4]. Fig. 1 illustrates particulate segregation during TLP bonding of aluminium-based MMC material. The region of weakness produced by particulate segregation at the bondline region promotes preferential failure during tensile testing [2, 3] and creep rupture testing of joints in ODS material [4].

Stefanescu *et al.* [5] and Shangquan *et al.* [6] developed an analytical model to explain pushing or engulfment of insoluble particles by the advancing solid–liquid interface. They indicated that the particulate material would be pushed when the rate of movement of the solid–liquid interface was less than a critical value. The critical velocity is given by the relationship

$$V_c = \Delta\sigma a_0 / 12\eta\alpha R \quad (1)$$

where  $\Delta\sigma$  is the relative surface energy difference ( $\sigma_{ps} - (\sigma_{pl} + \sigma_{sl})$ ),  $\sigma_{ps}$  is the interfacial energy between the particle and the solid,  $\sigma_{pl}$  is the interfacial energy

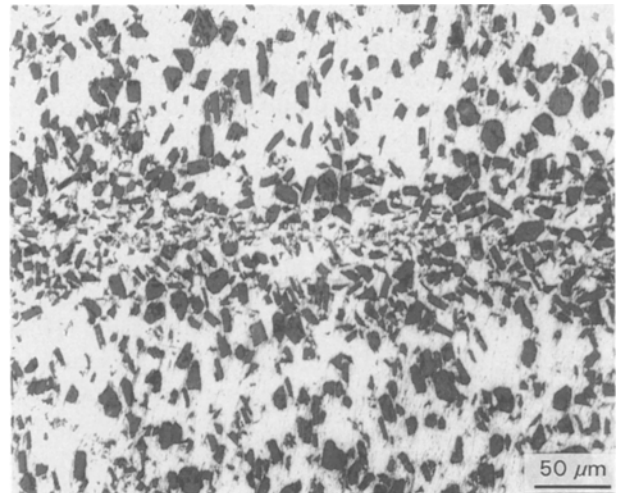


Figure 1 Segregation at the bondline during TLP bonding of aluminium-based MMC with copper foil as filler metal (for a bonding temperature of 853 K and a holding time of 6 min).

between the particle and the liquid,  $\sigma_{sl}$  is the interfacial energy between the liquid and the solid,  $a_0$  is a dimensional constant,  $\eta$  is the viscosity,  $\alpha$  is the thermal conductivity ratio of the particle and the melt, and  $R$  is the particle radius. For a given growth rate, particles having radii larger than  $R$  will be engulfed and particles with radii less than  $R$  will be pushed. In directionally-solidified Al–Mg–SiC melts, increasing the rate of interface movement from 16  $\mu\text{m s}^{-1}$  to 400  $\mu\text{m s}^{-1}$  (for a temperature gradient of 95  $^\circ\text{C cm}^{-1}$ ) and from 16  $\mu\text{m s}^{-1}$  to 40  $\mu\text{m s}^{-1}$  (for a temperature gradient of 117  $^\circ\text{C cm}^{-1}$ ) altered the behaviour from

particle pushing to engulfment [5]. The shape of the solid–liquid interface and the viscosity of the melt also play important roles. For example, in Al–Ni–SiC melts, particles were pushed when the rate of interface movement was up to  $155 \mu\text{m s}^{-1}$  and the temperature gradient was as low as  $70^\circ\text{C cm}^{-1}$ . For similar solidification conditions, particle engulfment occurred in Al–Mg–SiC melts. This difference in behaviour was attributed to the flatter solid–liquid interface profile and the lower viscosity in the Al–Ni–SiC melt [5].

The rate of interface movement during the isothermal solidification stage of TLP bonding is extremely slow. For example, Kokawa *et al.* [7] found an average rate of interface movement of around  $2 \times 10^{-4} \mu\text{m s}^{-1}$  when joining coarse-grained Nickel-200 material, and Tuah-Poku and Massalski [8] indicated an average rate of interface movement of  $5 \times 10^{-4} \mu\text{m s}^{-1}$ , when silver was TLP bonded using copper filler metal. It will be shown later in this paper that the average rate of interface movement is also extremely slow (around  $1 \times 10^{-3} \mu\text{m s}^{-1}$ ) during TLP bonding of aluminium-based metal matrix composite material. It is worth emphasizing that these solidification rates are much less than the critical interface velocity values ( $16\text{--}400 \mu\text{m s}^{-1}$ ) required to push SiC particles during solidification of directionally-solidified Al–Mg–SiC melts [5].

Based on the above discussion, it is suggested that segregation will occur during the isothermal solidification stage of TLP bonding, when particles that have radii less than a critical value are pushed ahead of the moving solid–liquid interface. With this in mind, the segregation tendency will depend on the relationship between the liquid film width produced at the bonding temperature,  $T_b$ , and the particle diameter and particle spacing distributions in the metal matrix composite material. When the width of the liquid film is large enough, so that sufficient particulate material is contained in the melt, particles will be pushed ahead of the moving solid–liquid interface and segregation will occur at the bondline region. However, if the liquid width at the bonding temperature is less than some critical value, segregation will not occur.

The present paper examines the relation between segregation, liquid film width at the bonding temperature and the base metal particulate diameter and particle spacing distributions during TLP bonding of aluminium metal matrix composite material. Because the width of the liquid film at  $T_b$  depends on the width and chemical composition of the filler metal employed during TLP bonding, the present paper examines joining of aluminium-based MMC material using copper filler metals with different thicknesses. Also, during conventional TLP bonding, the heating rate to  $T_b$  is relatively slow, unless induction heating is used. Because copper diffuses into the base metal during the heating cycle from room temperature to the bonding temperature, the heating rate will have a marked influence on the content of liquid formed at  $T_b$ . With this in mind, the present paper employs numerical modelling to evaluate copper diffusion into the base metal for a range of heating rates and evaluates the

influence of heating rate on the liquid width formed at the bonding temperature.

## 2. Experimental procedure

### 2.1. Materials

The 6061 aluminium-based MMC material employed in this study contained 20 vol% alumina. Base metal A contained small particles (the average particle diameter was  $9 \mu\text{m}$ ) and base metal B contained larger particle diameters (the average particle diameter was  $27 \mu\text{m}$ ). Cylindrical test specimens, 10 mm diameter  $\times$  7.5 mm thick, were employed during TLP bonding. The copper film thicknesses were 25, 15, 10 and  $5 \mu\text{m}$ . When very thin copper films were required, these were sputtered on to the surfaces of both MMC substrates. During sputtering, the vacuum was  $10^{-5}$  torr and the sputtered film thickness ranged from 0.6– $2.4 \mu\text{m}$ .

### 2.2. Procedure

The contacting surfaces of the MMC base metal were machined and then polished using 1200 grade emery paper. Prior to bonding, the test specimens were ultrasonically cleaned in an acetone bath and the copper filler metal was inserted at the joint interface. The test assemblies were fixed in place using a specimen holder and the TLP bonding was carried out in a vertical furnace that was configured so that the test specimens could be quenched into a water bath. The vacuum was maintained at  $10^{-5}$  torr (1 torr = 133.322 Pa) and the bonding temperature ( $853 \pm 5 \text{ K}$ ) throughout.

The heating rate between room temperature and the bonding temperature (853 K) in the vertical furnace was  $5 \text{ K s}^{-1}$ . After known holding times at the bonding temperature, the test specimens were quenched into water. It has already been confirmed that the liquid width during TLP bonding can be measured using test samples water-quenched from the bonding temperature [9]. The width of the melted zone in the test samples was measured following etching in a solution of 2 ml HF, 3 ml  $\text{HNO}_3$ , 5 ml HCl in 250 ml distilled water.

The effect of heating rate on the liquid width produced at the bonding temperature was evaluated using a horizontal vacuum furnace. During testing, the heating rate from room temperature to 573 K was  $0.05 \text{ K s}^{-1}$  and the heating rate from 573 and 853 K was  $0.01 \text{ K s}^{-1}$ . This particular heating cycle was selected because the diffusion rate of copper in aluminium is extremely low at temperatures less than 573 K.

### 2.3. Particulate distribution in base metals A and B

The particulate distributions in base metals A and B were evaluated by point counting, see Fig. 2. The particle dimensions were measured using photomicrographs produced at  $\times 200$  mag. It was worth pointing out that although Fig. 2 indicates particle diameter values, this is an approximation (because both base metals A and B contained alumina particles which had

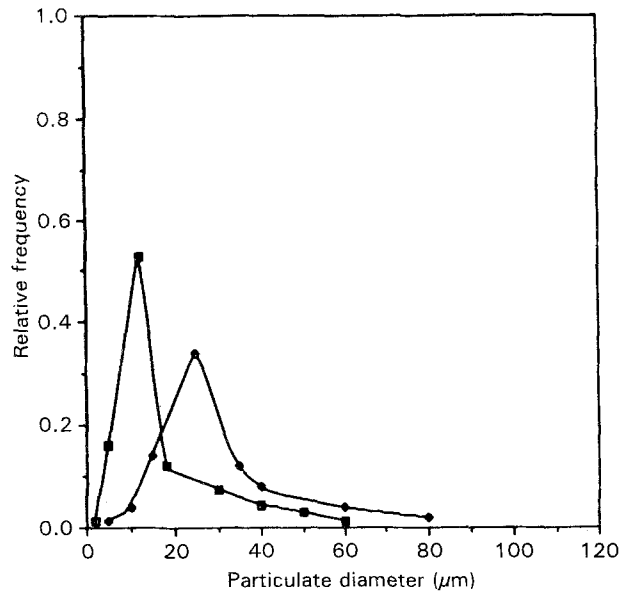


Figure 2 Particle diameters in base metals (■) A and (◆) B.

aspect ratios around 1.5:1). The majority of the particles in the base metal A had diameters ranging from 8–16 μm and the median particle diameter was 14 μm. The majority of the particles in base metal B had diameters ranging from 18–34 μm and the median particle diameter was 28 μm. The particle spacing in base metals A and B was estimated from micrographs (by measuring the closest distance between individual alumina particles). The particle spacing distributions in base materials A and B are shown in Fig. 3. The median particle spacing values were 26 μm (in base metal A) and 42 μm (in base metal B).

#### 2.4. Numerical modelling

Niemann and Garrett [10] employed thin copper films when joining of aluminium reinforced with continuous boron fibres and pointed out that copper diffusion into the base metal during the heating cycle had a large influence on liquid formation at the bonding temperature. However, the influence of heating rate on copper diffusion into the base metal was not calculated in their study. In the present paper, the effect of heating rate on copper diffusion into the base metal was evaluated using numerical modelling. (In these calculations it was assumed that the base metal was pure aluminium).

If the copper concentration in the base metal is  $C_0$  at the interface between the base metal and copper foil, solute diffusion depends on the relation

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} \quad (2)$$

The initial conditions are

$$c(x, 0) = 0 \quad (3)$$

and the boundary conditions are

$$c(0, t) = C_0 \quad (4)$$

$$\left(\frac{\partial c}{\partial x}\right)_{x=L} = 0 \quad (5)$$

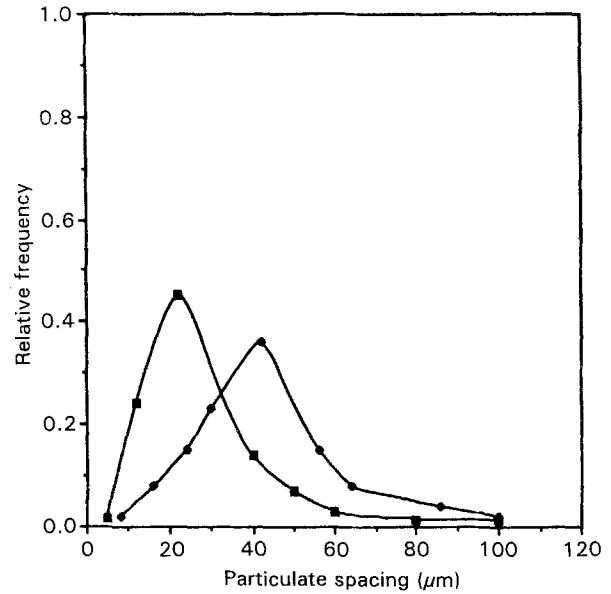


Figure 3 Particle spacing distributions in base metals (■) A and (◆) B.

where  $c$  is the solute concentration,  $D$  the interdiffusion coefficient,  $L$  the specimen length, and  $C_0$  is the solubility of copper in aluminium. Both  $D$  and  $C_0$  vary with temperature during the heating cycle, i.e.  $C_0 = 5.87 - 0.039T + 0.00007T^2$  and  $D = D_0 \exp(-Q/RT)$ , where  $D_0 = 1.8 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$ ,  $Q = 126 \text{ kJ mol}^{-1}$ ,  $T$  is the temperature (K) and  $R = 8.31 \text{ J mol}^{-1} \text{ K}^{-1}$  [11, 12].

The total amount of solute diffused into the base material during heating is given by the relation

$$m(t) = \int_0^L c(x, t) dx \quad (6)$$

and the thickness of copper consumed by this diffusion process is

$$w(t) = \frac{2m(t)}{C_F} \quad (7)$$

where  $C_F$  is the solute concentration in the filler metal.

### 3. Results

Fig. 4 shows the effect of copper foil thickness on particulate segregation at the bondline during TLP bonding of base metals A and B. Particulate segregation was observed when a 25 μm thick foil copper was used during TLP bonding of base metals A and B. In addition, particulate segregation was just visible when a 5 μm thick copper foil was employed during TLP bonding of base metal A, and when a 15 μm thick copper foil was employed during TLP bonding of base metal B (see Fig. 4). However, no bondline segregation occurred when the copper film thickness was less than 5 μm (in base metal A) and was less than 15 μm (in base metal B).

The particulate diameters in the segregated region at the joint centreline were quite different from those in base metals A and B (see Fig. 5). The segregated regions contained much smaller particles than the base metal (the diameters ranged from 5–30 μm).

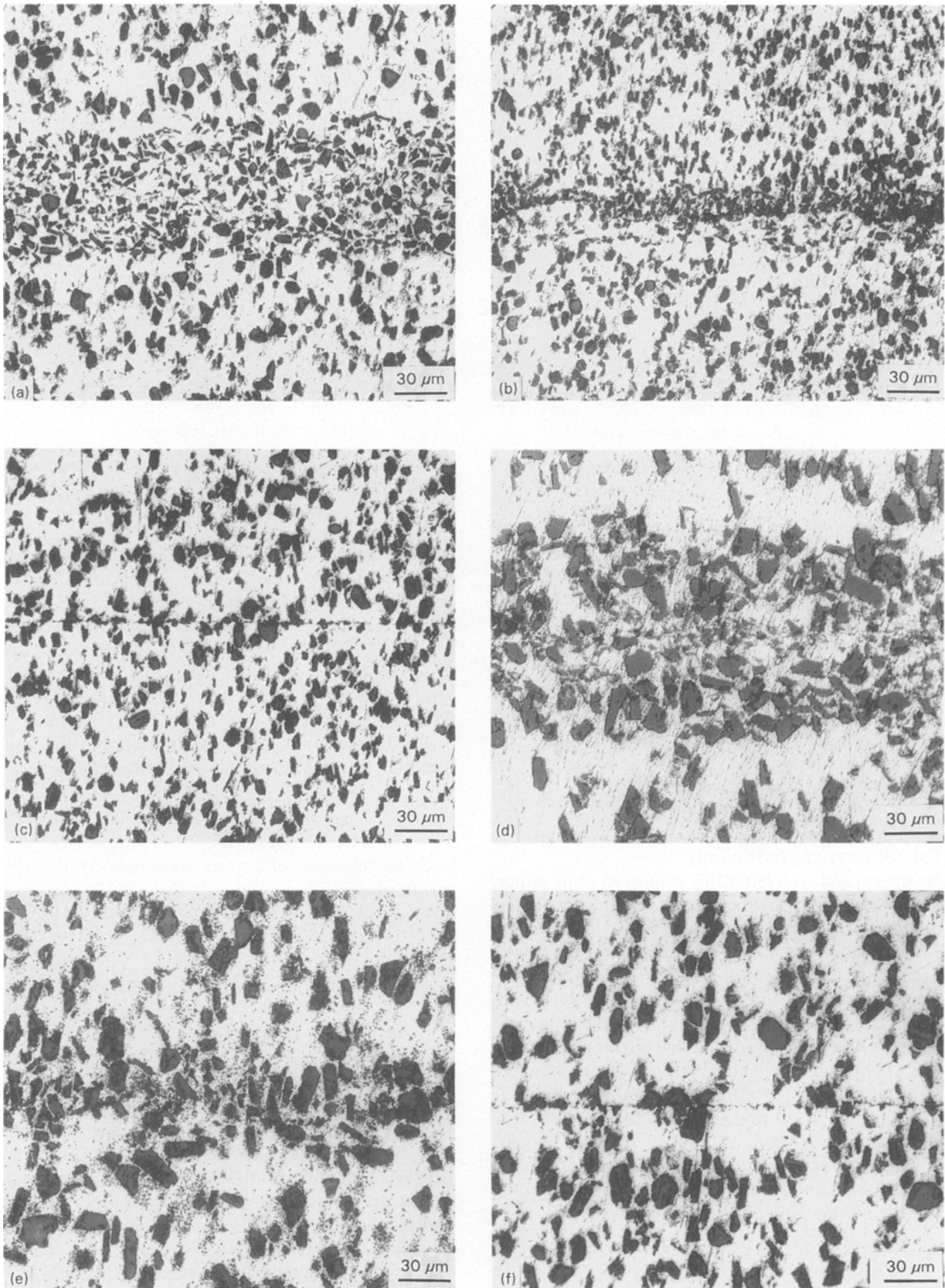


Figure 4 Effect of copper foil thickness on segregation during TLP bonding (for a heating rate of  $5 \text{ K s}^{-1}$  to the bonding temperature and a holding time of 6 min). (a–c) for base metal A, (d–f) for base metal B. Film thickness: (a)  $25 \mu\text{m}$ , (b)  $5 \mu\text{m}$ , (c)  $0.6 \mu\text{m}$ , (d)  $25 \mu\text{m}$ , (e)  $15 \mu\text{m}$ , (f)  $5 \mu\text{m}$ .

Shangguan *et al.* [6] pointed out that poor wettability between particulate material and the aluminium melt promotes clustering of particles, with gas bubbles located at the centre of each cluster. Particle clustering

is favoured because it decreases the surface energy between the particles and the matrix. In the present study, however, there was no evidence of particles clustering in segregated regions at the joint centreline.

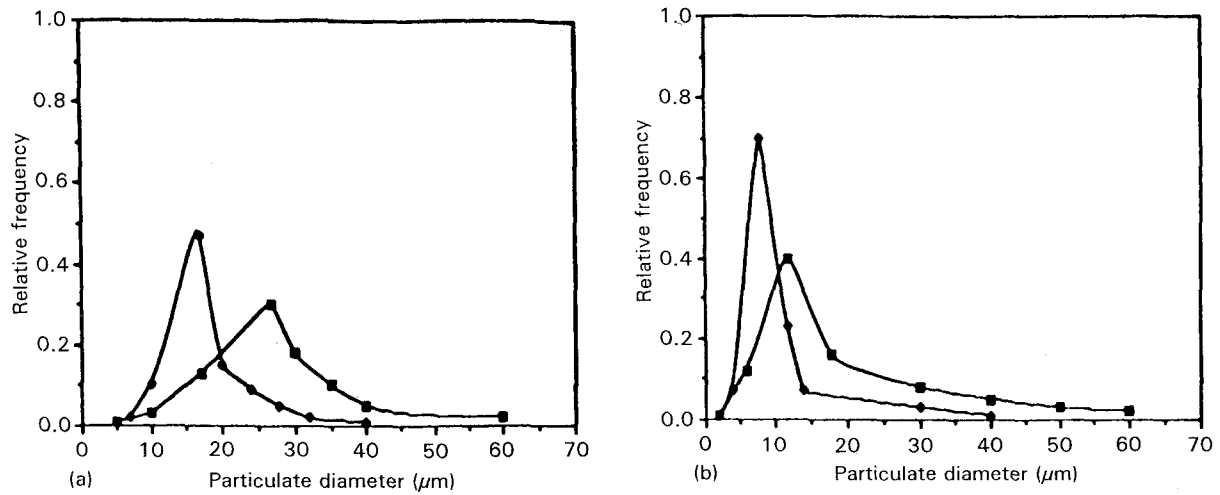


Figure 5 Particulate diameters in the segregated regions of joints (for a holding time of 6 min): (a) for base metal B, (b) for base metal A. (◆) Segregated regions.

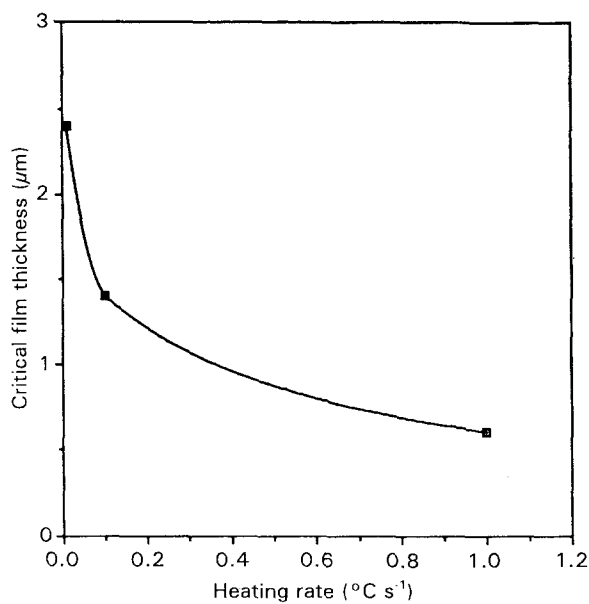


Figure 6 Effect of heating rate on the minimum thickness of copper required for satisfactory joint strength (for a holding time of 18 min). (■) Base metal A.

Although copper foil thicknesses less than 5  $\mu\text{m}$  and less than 15  $\mu\text{m}$  prevented particulate segregation during TLP bonding of base metals A and B, respectively, a minimum copper foil thickness (liquid width at the bonding temperature) was necessary to ensure satisfactory joint strength during the TLP bonding operation. When the copper film at the joint interface was too thin, the test joints had negligible strength and broke apart when they cooled to room temperature. Fig. 6 shows the effect of heating rate on the minimum copper foil thickness that produces satisfactory joint strength during TLP bonding of base metal A. When the heating rate increased from  $0.01 \text{ K s}^{-1}$  to  $1.0 \text{ K s}^{-1}$ , the required copper foil thickness decreased from  $2.4 \mu\text{m}$  to  $0.6 \mu\text{m}$ .

Fig. 7 shows the influence of holding time on the liquid width produced at the bonding temperature (for different copper filler thicknesses). It is worth pointing out that owing to difficulties in estimating the liquid

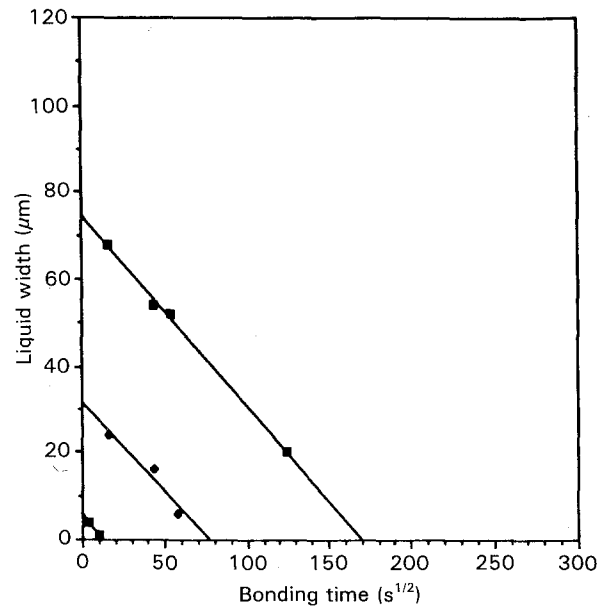


Figure 7 Liquid width/holding time relations for different copper film thicknesses (■) 25  $\mu\text{m}$ , (◆) 5  $\mu\text{m}$ , (●) 0.6  $\mu\text{m}$  (for a heating rate of  $5 \text{ K s}^{-1}$  to the bonding temperature).

width at the end of the base metal dissolution period of TLP bonding, the results in Fig. 7 consider a holding time of 6 min at 853 K. Fig. 8 shows the influence of heating rate on the liquid width produced at 853 K. Smaller liquid widths were produced when the heating rate decreased from  $5 \text{ K s}^{-1}$  to  $0.01 \text{ K s}^{-1}$ .

The results of the numerical calculations are shown in Figs 9 and 10. Fig. 9 shows the effect of heating rate (from  $0.01$ – $1.0 \text{ K s}^{-1}$ ) on the copper concentration profiles in the 6061 base metal. It is worth noting that these calculations only consider the range from room temperature to 821 K (the eutectic temperature in the aluminium-rich region of the Al–Cu binary phase diagram). The amount (thickness) of copper that diffuses into the base metal during the heating cycle from root temperature to 821 K is shown in Fig. 10. As expected, slowing the heating rate from  $1 \text{ K s}^{-1}$  to  $0.01 \text{ K s}^{-1}$  increased the amount of copper that diffused into the 6061 base metal.

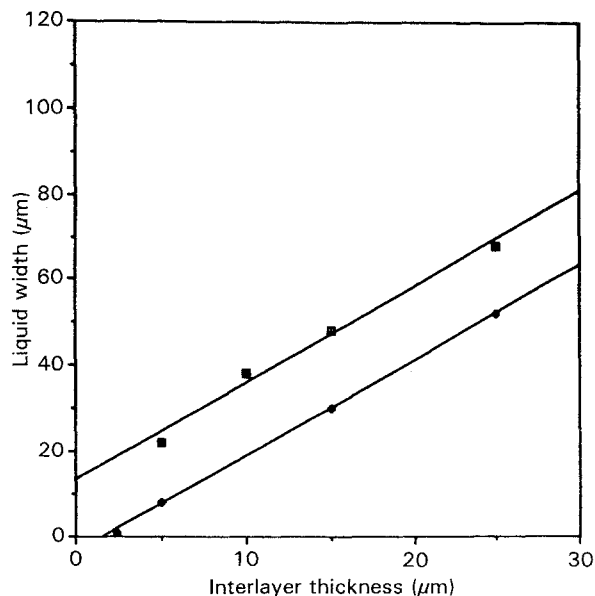


Figure 8 Effect of heating rate on the liquid width produced at the bonding temperature (for base metal A and a holding time of 6 min). (■)  $5 \text{ K s}^{-1}$ , (◆)  $0.01 \text{ K s}^{-1}$ .

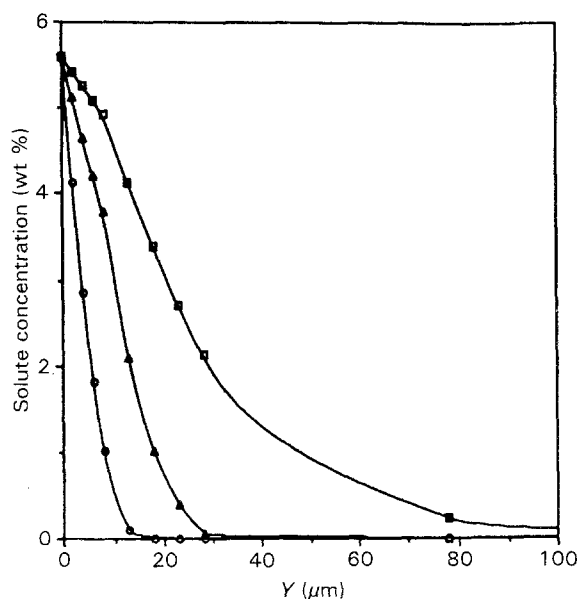


Figure 9 Solute concentration profiles in base metal for different heating rates from room temperature to the eutectic temperature. Heating rate: (□)  $0.01 \text{ K s}^{-1}$ , (▲)  $0.1 \text{ K s}^{-1}$ , (○)  $1.0 \text{ K s}^{-1}$ .

#### 4. Discussion

Fig. 5 confirms that the particle diameters in the segregated zone at the bondline were much smaller than those in base metals A and B. The particle diameters in the segregated region ranged from 5–30  $\mu\text{m}$ , and there was no evidence of particle agglomeration in any test sample. The difference in particle diameters in the segregated region and in base metals A and B can be explained if it is assumed that particles are pushed ahead of the migrating solid–liquid interface during the isothermal solidification stage of TLP bonding. As pointed out earlier, Stefanescu *et al.* [5] and Shangguan *et al.* [6] indicated that particles are pushed when the rate of solid–liquid interface migration is less than a critical value, given by Equation 1. This relation predicts that particles larger than radius,  $R$ , will be engulfed and

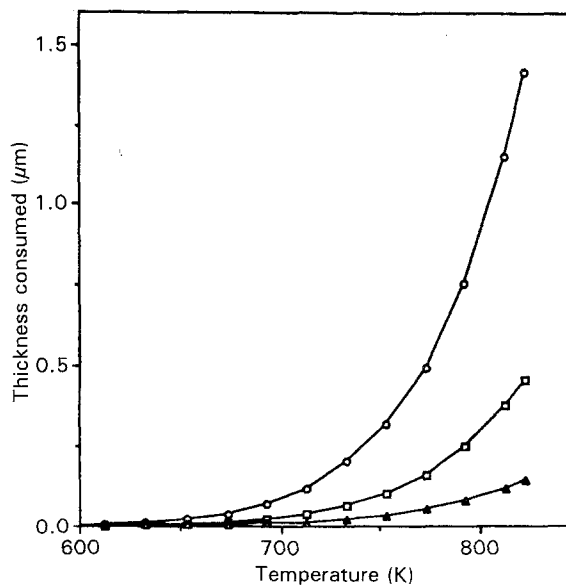


Figure 10 Effect of heating rate (from room temperature to the eutectic temperature) on the thickness of copper diffused into the base metal. Heating rate: (○)  $0.01 \text{ K s}^{-1}$ , (□)  $0.1 \text{ K s}^{-1}$ , (▲)  $1.0 \text{ K s}^{-1}$ .

that particles having radii less than  $R$  will be pushed by the migrating solid–liquid interface. However, because some alumina particles were located immediately adjacent to the joint interface prior to the TLP bonding operation, not all the particles in the segregated region result from pushing ahead of the migrating solid–liquid interface. If it is arbitrarily assumed that only 95% of the alumina particles in the segregated region result from particle pushing, the critical particle diameters for segregation during TLP bonding of base metals A and B are approximately 30 and 29  $\mu\text{m}$ , respectively. Bearing this in mind, particulate segregation would not have been a problem during TLP bonding if the composite base material only contained alumina particles with diameters exceeding 30  $\mu\text{m}$ .

#### 4.1. Effect of bonding temperature on particulate segregation

The influence of bonding temperature on the segregation tendency during TLP bonding of aluminium-based composite material can be estimated as follows. During TLP bonding, the rate of solid–liquid interface movement continuously changes during the isothermal stage of the joining process. However, a rate constant,  $\beta$ , can be derived if the interface displacement,  $\Delta X$ , is plotted against the square-root of the holding time,  $t$ , at the bonding temperature, i.e.

$$\Delta X = 2\beta t^{1/2} \quad (9)$$

The rate constant,  $\beta$ , is unaffected by the thickness of the filler metal employed during TLP bonding (see Fig. 7) and can be calculated numerically using the relation [13]

$$(C_{La} - C_{aL})\beta\pi^{1/2} + \frac{D^{1/2}(C_{aL} - C_M)}{1 - \text{erf}(\beta/D^{1/2})} \times \exp\left(-\frac{\beta^2}{D}\right) = 0 \quad (10)$$

where  $C_M$  is the original solute concentration (0 wt %),  $D$  is the solute diffusion coefficient,  $C_{L\alpha}$  is the solute concentration in liquid at the solute–liquid interface and  $C_{\alpha L}$  is the solute concentration in solid at the solid–liquid interface.

Because copper foils were used during TLP bonding, the rate constant,  $\beta$ , depends on the diffusivity of copper in the base metal, and on the  $C_{La}$  and  $C_{aL}$  values at the bonding temperature (which can be obtained from the Al–Cu binary phase diagram [12]). In the present study,  $D$  ( $\text{m}^2 \text{s}^{-1}$ ) =  $1.8 \times 10^{-5} \exp[(1.26 \times 10^5 / (8.31 T_b))] [11]$ ,  $C_{L\alpha} = 0.445 (933 - T_b) - 1.361 \times 10^{-3} (933 - T_b)^2$  and  $C_{\alpha L} = 0.05 (933 - T_b)$ .

If the bonding temperature is reduced from 853 K to 823 K (immediately above the eutectic temperature), the solute diffusion coefficient and rate constant values will be decreased and the completion time required for isothermal solidification will be longer. In contrast, increasing the bonding temperature from 853 K to 883 K will increase the solute diffusion coefficient and the rate constant,  $\beta$ , and decrease the time required for the isothermal solidification.

It would be expected from Equation 1 that a change in solidification rate will affect the particle diameter range segregated at the joint centreline region and the critical particle diameter,  $d_{crit}$ , for segregation. For example, decreasing the solidification rate should promote segregation of larger diameter particles during TLP bonding. However, the change in the rate constant,  $\beta$ , produced by a decrease in bonding temperature from 853 K to 823 K is relatively small. For example, the calculated rate constants are  $0.048 \mu\text{m s}^{-1/2}$  (at a bonding temperature of 823 K) and  $0.052 \mu\text{m s}^{-1/2}$  (at a bonding temperature of 853 K). It follows that the particle diameters segregated at the joint centreline and the critical particle diameter for segregation will be largely unaffected by a decrease in bonding temperature from 853 K to 823 K.

In contrast, increasing the bonding temperature from 853 K to 883 K produces a substantial increase in the rate constant (from  $0.052 \mu\text{m s}^{-1/2}$  to  $0.062 \mu\text{m s}^{-1/2}$ ). However, in spite of this change in the rate constant, the average rate of movement of the solid–liquid interface during TLP bonding is very slow compared to the values needed to produce a change from particle pushing to particle engulfment in directionally-solidified melts. For example, in a directionally-solidified Al–Mg–SiC melt, Stefanescu *et al.* [5] found that increasing the rate of interface movement from  $16 \mu\text{m s}^{-1}$  to  $400 \mu\text{m s}^{-1}$  (for a temperature gradient of  $95^\circ\text{C cm}^{-1}$ ) and from  $16 \mu\text{m s}^{-1}$  to  $40 \mu\text{m s}^{-1}$  (for a temperature gradient of  $117^\circ\text{C cm}^{-1}$ ) altered the behaviour from particle pushing to engulfment. Consequently, it is unlikely that increasing the bonding temperature from 853 K to 883 K will influence the particle diameters segregated at the joint centreline and the critical particle diameter for segregation.

#### 4.2. Effect of base metal grain size on particulate segregation

Base metal grain size affects the solidification rate during TLP bonding. For example, Kokawa *et al.* [7]

observed that the rate constant increased when fine grain size base metal was employed during TLP bonding of Nickel 200 base metal. For example, the rate constant increased from  $0.046 \mu\text{m s}^{-1/2}$  to  $0.074 \mu\text{m s}^{-1/2}$  when the base metal grain size decreased from greater than 4 mm to 40  $\mu\text{m}$  during TLP bonding of Nickel 200 base metal [7]. The solidification rate increased because liquid penetration at grain-boundary regions increased the solid–liquid interfacial area and due to increased solute diffusion at grain-boundary regions [7, 9, 13, 14]. However, the average rate of solid–liquid interface movement is still extremely slow when the fine-grained base metal is TLP bonded. As mentioned above, it is much less than that required to produce a change from particle pushing to particle entrapment in a directionally-solidified Al–Mg–Si melt [5]. Consequently, it is unlikely that decreasing the base metal grain size will affect the particle diameters segregated at the joint centreline and the critical particle diameter for segregation.

#### 4.3. Filler metal thickness and particulate segregation

Segregation was not observed when copper foil thicknesses less than 5  $\mu\text{m}$  (in base metal A) and less than 15  $\mu\text{m}$  (in base metal B) were employed during TLP-bonding. For a heating rate of  $5 \text{K s}^{-1}$  to the bonding temperature, the liquid film widths produced using these foil thicknesses were approximately 25 and 45  $\mu\text{m}$  (see Fig. 8). It is worth noting that these liquid film widths were similar to the median particle spacings in base metals A and B. It follows that the particle spacing in base metals A and B has a critical influence on the particulate segregation tendency. For example, Fig. 3 suggests that few particles will be contained in a liquid width of 25  $\mu\text{m}$ , when base metal B is TLP bonded using a 5  $\mu\text{m}$  thick copper foil. The same argument can be applied if base metal A is TLP bonded using a copper foil thickness much less than 5  $\mu\text{m}$ .

#### 4.4. Effect of heating rate

The liquid width at the bonding temperature decreased when a slower heating rate was employed (see Fig. 8). The numerical calculations confirm that more copper diffuses into the base metal, when the heating rate decreases from  $1.0 \text{K s}^{-1}$  to  $0.01 \text{K s}^{-1}$  (see Figs 9 and 10). However, these calculations only consider the heating range between room temperature and the eutectic temperature (821 K) and the bonding temperature employed in the present study was 853 K. Nakagawa *et al.* [15] have already pointed out that the use of a thin filler metal and a slow heating rate ( $0.05$ – $0.1 \text{K s}^{-1}$ ) can promote solidification when the specimen temperature increases from the filler metal melting point to the bonding temperature. If a similar effect occurs during TLP bonding of aluminium-based composite material, solidification in the temperature range between 821 and 853 K will decrease the liquid width produced at the bonding temperature. It follows that the smaller liquid widths produced as a result of

slow heating to 853 K (in Fig. 8) may result from a combination of increased copper diffusion into the base metal (in the temperature range between room temperature and 821 K) and solidification (in the temperature range between 821 and 853 K).

When the amount of liquid formed at the bonding temperature decreased below a critical level, the test assemblies broke apart at room temperature following the joining operation. The copper film thickness that produced satisfactory bond strength increased from 0.6  $\mu\text{m}$  to 2.4  $\mu\text{m}$  when the heating rate decreased from 1  $\text{K s}^{-1}$  to 0.01  $\text{K s}^{-1}$  (see Fig. 6). This change in film thickness corresponds well with the calculated results presented in Fig. 10 (the calculated thickness of copper that diffuses into the base metal during slow heating from room temperature to the eutectic temperature is around 1–1.5  $\mu\text{m}$ ). In effect, satisfactory joint formation depends on sufficient copper being available for liquid formation at the bonding temperature (853 K). In this connection, liquid formation at the joint interface has already been associated with increased joint strength during diffusion bonding of Al–Cu–Mg alloy material to titanium [16]. Enjo *et al.* [16] related bonding temperature with joint strength and found that there was negligible joint strength at bonding temperatures less than 790 K (the solidus temperature of their Al–Cu–Mg alloy). However, joint strength increased rapidly when the volume fraction of liquid produced at the bondline increased to around 6% and the highest joint tensile strength in Al–Cu–Mg alloy/titanium joints was attained using a bonding temperature of 840 K.

## 5. Conclusions

Particulate segregation during TLP-bonding of aluminium-based metal matrix composite material has been examined. The principal conclusions of this study are as follows.

1. The extremely slow rate of movement of the solid–liquid interface during the isothermal solidification stage of TLP bonding promotes particulate segregation. The particle-size distribution in the segregated region of completed joints is quite different from that in the base metal (particle diameters less than 30  $\mu\text{m}$  are preferentially segregated). Particle diameters segregated at the joint centreline and the critical particle diameter for segregation will not be affected by bonding temperature selection (because the average rate of movement of the solid–liquid interface is extremely slow during the TLP bonding process). In addition, the particle diameters segregated at the joint centreline and the critical particle diameter for segregation will not be affected by the grain size of the base material that is TLP bonded.

2. The segregation tendency markedly depends on the particle spacing in the composite material. In the present study, segregation was observed when the copper foil thickness (the liquid width at the bonding temperature) exceeded critical values. During TLP bonding at 853 K, the critical copper foil thicknesses were 5  $\mu\text{m}$  (in base metal A) and 15  $\mu\text{m}$  (in base metal

- B). The liquid widths at  $T_b$ , produced using these foil thicknesses were similar to the median particle spacing values in the base metals examined.

3. When the amount of liquid formed at the bonding temperature decreased below a critical level, the test samples broke apart at room temperature following the joining operation. It follows that satisfactory joint formation depends on sufficient copper being available for liquid formation at the bonding temperature (853 K). The minimum thickness of copper for satisfactory joint strength increased from 0.6  $\mu\text{m}$  to 2.4  $\mu\text{m}$  when the heating rate decreased from 1  $\text{K s}^{-1}$  to 0.01  $\text{K s}^{-1}$ .

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