Counteracting particulate segregation during transient liquid-phase bonding of MMC–MMC and Al2O3–MMC joints

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The microstructure and mechanical properties of MMC–MMC and Al₂O₃–MMC joints (MMC
is matel metrix composite) produced at a bonding temperature of 952 K using connecteile is metal matrix composite) produced at a bonding temperature of 853 K using copper foils ranging in thickness from 10 to 30 μ m were examined. The particle segregation tendency during transient liquid-phase (TLP) bonding of aluminium-based MMC material markedly increases when the aluminium-based composite material contains large number of small radius (less than 10 μ m) reinforcing particles. Also, the particle segregation tendency is much greater in dissimilar Al₂O₃-MMC joining since the rate of solid-liquid interface
maximum at is much along and the time required for completing the insthumal solidities movement is much slower and the time required for completing the isothermal solidification during TLP bonding is much longer. The particle segregation tendency during MMC*—*MMC and Al₂O₃–MMC bonding can be counteracted using a combination of a short (1 min)
halding time at the handing temperature (953 K) and subsequent nest unld hast tracti holding time at the bonding temperature (853 K) and subsequent post-weld heat treatment at 773 K for 4 h. This TLP-bonding*—*heat-treatment cycle removes the retained eutectic phase present at the joint centreline.

1. Introduction

Transient liquid-phase (TLP) bonding has been generally applied for the repair of nickel-based superalloy turbine blade components since the final joint mechanical properties are similar to those of the as-received base material [\[1\]](#page-4-0). However, the TLP-bonding process has an inherent problem when particle-reinforced base materials are TLP-bonded since particles contained in the liquid formed at the bonding temperature segregate to the joint centreline when the joint solidifies isothermally. Particle segregation at the centreline of completed MMC–MMC and Al₂O₃–MMC joints has been associated with preferential failure during subsequent mechanical testing [\[2](#page-4-0)*—*[6\]](#page-4-0).

It has been suggested that particle segregation during TLP bonding depends on the relationship among the liquid width formed at the bonding temperature, the particle diameter and the interparticle spacing in the as-received composite base material [\[2](#page-4-0)*—*4]. When the liquid film formed at the bonding temperature is large enough that large numbers of reinforcing particles become encapsulated, the particles are pushed ahead of the moving solid*—*liquid interface during joint solidification and a segregated layer forms at the joint centreline. However, when the liquid width is less than a critical value, particle segregation is not observed.

Zhai *et al*. [\[4\]](#page-4-0) confirmed that particle segregation could be avoided during Al_2O_3-MMC bonding when the holding time at the bonding temperature was very short (less than 5 min). However, the shear strengths of completed joints produced using short holding times were extremely poor owing to the retention of eutectic material at the centreline of completed joints. The present paper investigates the relation between particle segregation, particle size and joint shear strength properties in MMC*—*MMC and Al_2O_3-MMC joints. It is confirmed that the particle segregation tendency during TLP bonding of MMC–MMC and Al₂O₃–MMC joints can be counteracted through the combination of very short holding times at the bonding temperature with postweld heat treatment to remove retained eutectic phase at the centreline of completed joints.

2. Experimental procedure

Cylindrical sections 10 mm long and of 5 mm diameter of aluminium alloy 6061 (W6A-T6) composite base material and alumina ceramic were employed throughout. The aluminium-based composite material (MMC) contained 20 vol% Al_2O_3 particles. Two aluminium-based composite base materials were examined, namely, base material A (this is referred to as MMC1 in some figures) which contained Al_2O_3 reinforcing particles having an average diameter of $9 \mu m$ and base material B (this is referred to as MMC2 in some figures) which contained Al_2O_3 reinforcing particles having an average diameter of $28 \mu m$. The alumina ceramic contained 0.1 wt% MgO and 0.22 wt% CaO as impurities.

The contacting surfaces of the alumina and MMC base material were polished using 1200 grade emery paper and were then ultrasonically cleaned in an acetone bath prior to TLP bonding. TLP bonding was carried out using pure copper foils having thicknesses ranging from 10 to 30 μ m. All TLP-bonding trials were carried out at 853 K in a vacuum of 10^{-5} torr. The heating rate between room temperature and the bonding temperature was 5 K s^{-1} and, after a known holding time at 853 K, the test specimens were furnace cooled to room temperature. The influence of postweld heat treatment at 773 K on the shear strength of completed joints was investigated using test sections previously TLP bonded at 853 K using a holding time of 1 min.

The shear strength of completed joints was evaluated using a specially designed fixture which prevented sample rotation during mechanical testing (Fig. 1). All joints were mechanically tested at room temperature and the reported shear strength results are the average of two tests at each condition.

3. Results

It has been generally accepted that pushing or engulfment of reinforcing particles during solidification depends on the velocity of movement of the solid*—*liquid interface [7*—*[10\]](#page-4-0). When the rate of movement of the solid*—*liquid interface exceeds a critical value, the particles become engulfed. During TLP bonding, engulfment of reinforcing particles would result in a uniform distribution of particles in the solidified joint. However, the outcome of particle pushing during the isothermal solidification in TLP bonding will result in the formation of a particle segregated layer at the joint centreline. The critical rate, V_c , of solid-liquid interface movement for particle pushing and/or entrapment is determined by the relation

$$
V_{\rm c} = \frac{\Delta \sigma_0 a_0}{12 \eta \alpha R} \tag{1}
$$

where $\Delta\sigma_0$ is the interfacial energy difference between the particle and the matrix, a_0 is the atomic distance, η is the melt viscosity, *R* is the particle radius and α is the thermal conductivity. The critical velocity increases when the particle radius decreases. Consequently, the combination of fine reinforcing particles in the as-received MMC base material and a slow

rate of solid*—*liquid interface movement during the TLP-bonding operation will mean that there will be a considerable tendency for particle pushing and particle segregation at the joint centreline. The particle segregation tendency will be largest when the MMC base material contains small diameter reinforcing particles. [Fig. 2](#page-2-0) shows the calculated relation between the critical velocity, V_c , and the particle radius, *R*, when MMC–MMC and Al_2O_3 –MMC joints are TLP bonded at a temperature of 853 K. In these calculations, $\Delta \sigma_0 = 1.0 \text{ N m}^{-1}$, $a_0 = 2 \times 10^{-10} \text{ m}$, η is 0.005 Pa s and $\alpha = 0.146$ [\[7, 11\]](#page-4-0). It is readily apparent that the critical rate of solid*—*liquid interface movement becomes extremely high when the particle radius is less than $10 \mu m$.

[Fig. 3](#page-2-0) shows the relationship between the width of the particle segregated region, the particle radius and the holding time at the bonding temperature. Two MMC base materials are considered, namely, base metal A (this is MMC1 in [Fig. 3\)](#page-2-0) containing reinforcing particles with an average diameter of $9 \mu m$ and base metal B (this is MMC2 in [Fig. 3\)](#page-2-0) containing reinforcing particles with an average diameter of $28 \mu m$. The particle segregated layer in the A_2O_3 -MMC1 joint was wider than in an Al₂O₃–MMC2 joint since the MMC1 base material contained large numbers of small-diameter particles. Also, the width of the particle segregated layer formed at the centreline of the Al₂O₃–MMC1 joint exceeded that in the MMC1*—*MMC2 joint. This may be due to the much lower rate of solid*—*liquid interface movement and the much longer completion time for the isothermal solidification stage during dissimilar bonding of MMC an Al_2O_3 base materials. For example, the completion time for the isothermal solidification stage during MMC*—*MMC bonding is determined by the relation $\lceil 1, 12 \rceil$

$$
t_{\rm s} = \frac{\pi/16D_{\rm s}}{(C_{\rm F}W_0/C_{\rm al})^2}
$$
 (2)

where D_s is the diffusion coefficient of copper in aluminium, C_F is the solute content in the filler metal, W_0 is the initial width of the filler metal and $C_{\alpha l}$ is the solute concentration at the solid*—*liquid interface. Since copper cannot diffuse into the Al_2O_3 substrate during dissimilar bonding, the completion time will be four

Figure 1 Design of the shear-testing set-up. 1, casing pipe; 2, specimen; 3, holder; 4, bolt; 5, screw.

Figure 2 Relation between the critical rate of solid*—*liquid interface movement and the particle radius during TLP bonding of MMC base material (for a bonding temperature of 853 K).

Figure 3 Relation between the width of the particle segregated layer and the holding time at the bonding temperature (853 K) (when using a copper foil 30 μ m thick). ($-\Theta$), Al₂O₃–MMC1; ($-\Xi$), $MMC1-MMC2;$ (- \leftrightarrow), Al₂O₃-MMC2.

times longer than that in MMC*—*MMC bonding [\[4\]](#page-4-0), i.e.,

$$
t_{\rm s} = \frac{\pi/4D_{\rm s}}{(C_{\rm F}W_0/C_{\rm Al})^2} \tag{3}
$$

The calculated relation between the completion time and the copper foil thickness during Al_2O_3-MMC and MMC*—*MMC bonding is shown in Fig. 4. It is apparent that much longer completion times (and slower rates of solid*—*liquid interface movement) occur during Al₂O₃–MMC bonding. Consequently, there will be a very strong particle segregation tendency when MMC and Al_2O_3 base materials are TLP bonded. [Fig. 5](#page-3-0) shows particle segregation at the centreline of Al₂O₃–MMC1 and MMC1–MMC2 joints. In MMC1*—*MMC2 joints, preferential segregation of small-diameter reinforcing particles is more apparent on the MMC1 side of the joint centreline (in the base material which has the large numbers of small-diameter reinforcing particles).

Figure 4 Relation between the calculated time for completion of the isothermal solidification stage during TLP bonding and the width of the copper foil (for a bonding temperature of 853 K).

3.1. Effect of post-weld heat treatment

It has been suggested that particle segregation results from the slow rate of solid*—*liquid interface movement and the formation of a large liquid width at the bonding temperature. Also, when the liquid width at the bonding temperature is less than a critical value, particle segregation is not observed in MMC*—*MMC joints [\[2, 4\].](#page-4-0) It is apparent from Fig. 3 that particle segregation is not observed in MMC*—*MMC and dissimilar MMC–Al₂O₃ joints produced using a 5 min holding period at the bonding temperature. However, such joints comprise retained Al*—*Cu eutectic phase at the joint centreline and have extremely poor shear strength properties [\[4\].](#page-4-0)

[Fig. 6](#page-3-0) shows the effect of post-weld heat treatment on the mechanical properties (shear strength) of MMC2–MMC2 and dissimilar Al₂O₃–MMC2 joints produced using a holding time of 1 min at the bonding temperature (853 K). The joint shear strength increased markedly when long holding times were applied during post-weld heat treatment. In Al_2O_3-MMC2 joints made using copper foils 10 and 20 μm thick, the highest joint shear strength properties were produced after a post-weld heat-treatment time of 4 h at 773 K. In a similar manner, the shear strength properties of MMC2*—*MMC2 joints made using a copper foil $30 \mu m$ thick were markedly improved following heat treatment at 773 K for 4 h. However, the shear strength of Al_2O_3-MMC2 joints produced using a copper foil $30 \mu m$ thick were still poor even after post-weld heat treatment at 773 for 4 h (see Fig. 6).

[Fig. 7](#page-3-0) shows the relation between the width of the retained eutectic phase at the joint centreline and the holding time at the heat-treatment temperature (773 K). Eutectic material was still retained at the centreline of A_2O_3 -MMC₂ joints made using a copper foil 30 μ m thick following heat treatment for 4 h at 773 K. This readily explains the poor shear strength properties of Al_2O_3 –MMC joints produced using a copper foil $30 \mu m$ thick [\(see Fig. 6\).](#page-3-0) [Figure 8](#page-4-0)

Figure 5 Particle segregation during TLP bonding of (a) dissimilar AI_2O_3 –MMC1 and (b) similar MMC2–MMC1 joints (for a bonding temperature of 853 K, a holding time of 40 min at the bonding temperature and a copper foil thickness of 30 μ m). (Magnification, 198 \times .)

Figure 6 Relation between the joint shear strength and the holding time during post-weld heat treatment. $(-\Theta^{-})$, Al₂O₃-MMC2 joint made using copper foil 30 μ m thick; $(- \Box -)$, Al₂O₃-MMC2 foint made using copper foil 20 μm thick; $(-\diamondsuit -)$, Al₂O₃-MMC2 joint made using copper foil 10 μm thick; (−Δ−), MMC2−MMC2 joint made using copper foil 30 μ m thick. (All joints were produced using a 1 min holding time at 853 K.)

shows the joint centreline microstructures in MMC2–MMC2 and Al₂O₃–MMC2 joints produced using a combination of a 1 min holding time at the bonding temperature (853 K) and post-weld heat treatment at 773 K for 4 h. It is readily apparent that this combination of bonding time at 853 K and postweld heat treatment at 773 K produced joints free of particle segregation and retained eutectic material.

4. Conclusions

The microstructure and mechanical properties of $MMC-MMC$ and Al_2O_3-MMC joints produced at a bonding temperature of 853 K using copper foils ranging in thickness from 10 to 30 μ m were investigated. The following conclusions were reached.

Figure 7 Relation between the width of the retained eutectic phase and the holding time during post-weld heat treatment at 773 K. $(-\ominus -)$, Al₂O₃-MMC2 joint made using copper foil 10 μ m thick; $(-\Box -)$, Al₂O₃–MMC2 joint made using copper foil 20 μ m thick; $(-\diamondsuit$ —), Al₂O₃–MMC2 joint made using copper foil 30 µm thick; $(-$ △–), MMC2–MMC2 joint made using copper foil 10 µm thick. (All joints were produced using a 1 min bonding time at 853 K.)

1. The particle segregation tendency during TLP bonding of aluminium-based MMC material is markedly increased when the aluminium-based composite material contains large number of small-radius (less than $10 \mu m$ reinforcing particles. Since the copper solute cannot diffuse into the Al_2O_3 ceramic during the TLP-bonding operation, the particle segregation tendency is much greater during dissimilar Al2 O3 *—*MMC joining (since the rate of solid*—*liquid interface movement is much slower and the completion time required for completing the isothermal solidification stage is much longer).

2. The particle segregation tendency during MMC–MMC and Al₂O₃–MMC bonding can be counteracted using a combination of a short (1 min)

Figure 8 Joint centreline microstructure produced in (a) an Al₂O₃–MMC2 joint prior to heat treatment, (b) an MMC–MMC2 joint made prior to heat treatment, (c) an Al₂O₃–MMC2 joint following heat treatment at 773 K for 4 h and (d) an MMC2–MMC2 joint following heat treatment at 773 K for 4 h. (All joints were produced using copper foil 20 μ m thick and a bonding time of 1 min at 853 K.) (Magnifications: (a), (b), (d) $198 \times$; (c) $99 \times$.)

holding time at the bonding temperature (853 K) and subsequent post-weld heat treatment at 773 K for 4 h. Post-weld heat treatment removes the eutectic phase retained at the joint centreline.

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