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Transient liquid-phase bonding of alumina and metal matrix composite base materials

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Transient liquid-phase (TLP) bonding of aluminium-based metal matrix composite (MMC) and Al_2O_3 ceramic materials has been investigated, particularly the relationship between particle segregation, copper interlayer thickness, holding time and joint shear strength properties. The long completion time and the slow rate of movement of the solid–liquid interface during MMC/Al_2O_3 bonding markedly increased the likelihood of forming a particle-segregated layer at the dissimilar joint interface. Preferential failure occurred through the particle-segregated layer in dissimilar joints produced using 20 and 30 μm thick copper foils and long holding times ($\geq$20 min). When the particle-segregated layer was very thin ($<$10 μm), joint failure was determined by the residual stress distribution in the Al_2O_3/MMC joints, not by preferential fracture through the particle-segregated layer located at the bondline. Satisfactory shear strength properties were obtained when a thin (5 μm thick) copper foil was used during TLP bonding at 853 K.

1. Introduction

Transient liquid-phase (TLP) bonding has been generally applied when joining a range of metallic substrates, e.g. nickel- and iron-based superalloys, titanium alloys, stainless steel, aluminium, and aluminium- and titanium-based composites, because the resulting joints have mechanical properties similar to the as-received base materials [1]. The bonding process depends on the formation of a thin layer of low melting point eutectic or peritectic at the joint interface. The liquid film wets the contacting metallic substrates and solidifies isothermally prior to joint homogenization. For this reason, the joining process is generally considered to comprise a number of distinct stages, namely, heating to the bonding temperature, base metal dissolution, isothermal solidification and homogenization.

When aluminium-based metal-matrix composite (MMC) material is joined, the reinforcing particles are pushed ahead of the moving solid–liquid interface during the isothermal solidification stage in TLP bonding and a particle-segregated layer forms at the bondline. Preferential failure occurs through the particle-segregated layer during mechanical testing of MMC/MMC joints [2–5]. This particular problem has been counteracted by silver plating the MMC base material prior to joining, and through careful control of the copper foil thickness during TLP bonding [2–4].

Wetting of alumina by liquid aluminium has been studied by a number of investigators [6–8] and depends on a combination of different effects [8], namely: (i) a surface reaction that produces gaseous Al_2O, (ii) dehydroxylation of the alumina surface, and (iii) dissolution of some unknown phase into liquid aluminium which alters the magnitude and direction of the solid–liquid surface energy, γ_{sl}, and the magnitude of the liquid–vapour surface energy, γ_{lv}. When copper foil is employed during TLP bonding, the MMC and Al_2O_3 substrates are wetted by liquid Al–Cu eutectic. Retention of eutectic material in completed joints depends on the diffusion rate of copper in MMC base material at the bonding temperature, T_b, on the thickness of the copper foil used (on the liquid width at T_b), and on the holding time. In a similar manner, the particle segregation tendency in MMC/MMC joints depends on the liquid width produced at the bonding temperature. For example, Li *et al.* [2] confirmed that particle segregation can be avoided when the liquid width at T_b is less than the median inter-particle spacing in the composite material.

The present study investigated the relation between particle segregation, copper interlayer thickness (the liquid film width at the bonding temperature), holding time and the shear strength properties of dissimilar MMC/Al_2O_3 joints. When the width of the particle-segregated layer at the bondline exceeds 10 μm, joint failure occurs through the particle-segregated region. However, when the width of the particle-segregated layer is $\leq$10 μm, joint failure occurs in Al_2O_3 material immediately adjacent to the ceramic/metal interface, not through the particle-segregated layer.

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2. Experimental procedure

2.1. Materials

Cylindrical test samples, 10 mm diameter × 7.5 mm thick, of aluminium alloy 6061/Al_2O_3 (W6A.10A-T6) base material and Al_2O_3 ceramic were used throughout. The aluminium-based composite material contained 10 vol% Al_2O_3 particles having a mean particle diameter of 14 μm. The Al_2O_3 ceramic contained 0.1 wt % MgO and 0.11 wt % CaO as impurities. TLP bonding was carried out using pure copper foil interlayers; the range of copper foil thicknesses comprised 5, 10, 20 and 30 μm.

2.2. Procedure

The contacting surfaces of the Al_2O_3 and MMC base materials were polished using 1200 grade emery paper and were ultrasonically cleaned in an acetone bath prior to TLP bonding. The copper foil was inserted at the joint interface and TLP bonding was carried out in a 10^{-5} torr (1 torr = 133.322 Pa) vacuum at 853 K (580 °C). The heating rate between room temperature and the bonding temperature was $5\ \mathrm{K\,s^{-1}}$ and, after known holding time at 853 K, the test specimens were furnace-cooled to room temperature. The shear fracture strength of dissimilar MMC/Al_2O_3 joints was evaluated using a specially designed fixture which prevented sample rotation during testing (see Fig. 1). All joints were mechanically tested at room temperature (the reported shear strength values are the average of two tests at each condition).

3. Results and discussion

3.1. Rate of movement of the solid–liquid interface

It has been suggested that the slow rate of movement of the solid–liquid interface promotes particulate segregation during TLP bonding [2, 3]. Because copper can only diffuse into the MMC substrate during TLP bonding of dissimilar Al_2O_3 and MMC substrates, the isothermal solidification period will be longer and the rate of movement of the solid–liquid interface will be much slower than when similar MMC substrates are joined. During isothermal solidification of MMC/MMC joints, the total amount of solute, M_t, that diffuses into the base metal in time, t, depends on the relation [1, 9]

$$M_t = 2C_{\alpha L}\left(\frac{D_s t}{\pi}\right)^{0.5} \quad (1)$$

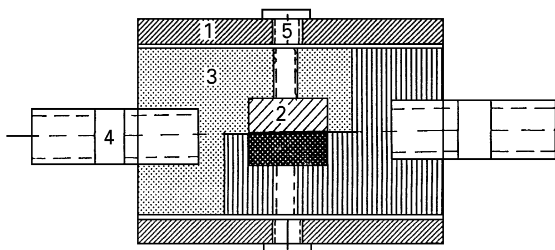


Figure 1 The shear testing fixture. (1) Casing pipe, (2) specimen, (3) holder, (4) bolt, (5) screw.

where $C_{\alpha L}$ is the solute concentration in solid at the solid–liquid interface, D_s is the diffusion coefficient of copper in aluminium, and t is the holding time. If the amount of solute diffused into the base metal during the heating and dissolution stages in TLP bonding is ignored, the total amount of solute diffused into the base metal equals the original solute content of the filler metal, i.e.

$$C_F W_0 = 4C_{\alpha L}\left(\frac{D_s t}{\pi}\right)^{0.5} \quad (2)$$

where C_F is the solute content in the filler metal and W_0 the initial width of the filler metal. The completion time, t_s, for isothermal solidification is therefore

$$t_s = \frac{\pi}{16 D_s}\left(\frac{C_F W_0}{C_{\alpha L}}\right)^2 \quad (3)$$

Because copper only diffuses into the MMC substrate during dissimilar Al_2O_3/MMC bonding, the completion time will be four times longer than that in MMC/MMC joints, i.e.

$$t_s = \frac{\pi}{4 D_s}\left(\frac{C_F W_0}{C_{\alpha L}}\right)^2 \quad (4)$$

Fig. 2 shows the calculated relation between copper foil thickness, W_0, and the completion time during dissimilar MMC/Al_2O_3 bonding based on Equation 4.

The rate of solid–liquid interface movement during MMC/Al_2O_3 bonding can be estimated using a simple one-dimensional model by assuming that migration of the liquid–solid interface is governed by diffusion in the solid phase and that diffusion in the liquid can be neglected [10]. The interface displacement, Y_0, varies with the square-root of the holding time, t, at the bonding temperature

$$Y_0 = Y - Y_{max} = 2\beta t^{1/2} \quad (5)$$

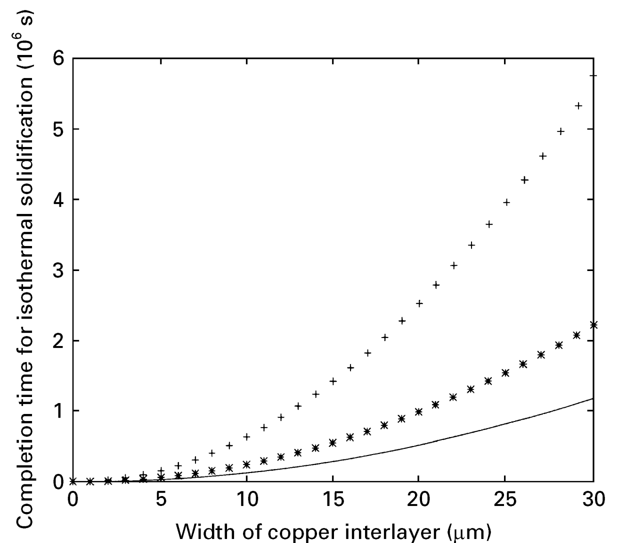


Figure 2 Calculated relation between the completion time for isothermal solidification and the width of the copper interlayer, for bonding temperatures of (+) 823 K, (*) 853 K and (—) 883 K.

where Y_{max} is the maximum value of displacement Y_0, and β is a rate constant which can be calculated numerically using the relation

$$(C_{L\alpha} - C_{\alpha L})\beta\pi^{1/2} + \frac{D_s^{0.5}[(C_{\alpha L} - C_M)]}{1 - \mathrm{erf}(\beta/D_s^{1/2})}\exp\left(-\frac{\beta^2}{D_s}\right) = 0 \quad (6)$$

where $C_{L\alpha}$ is the solute concentration in liquid at the solid–liquid interface, C_M is the original solute concentration (0 wt %). The rate constant, β, depends on the $C_{L\alpha}$ and $C_{\alpha L}$ values and on the diffusivity of copper in the aluminium-based composite material at T_b. Because copper can only diffuse into the MMC substrate, the rate of solid–liquid interface movement during MMC/Al_2O_3 bonding is given by

$$\frac{dY_0}{dt} = \frac{\beta}{2t^{1/2}} \quad (7)$$

The calculated relation between the holding time at T_b and the rate of movement of the solid–liquid interface during MMC/Al_2O_3 bonding using copper foil is shown in Fig. 3. In this calculation, $D_s(m^2\,s^{-1}) = 1.8 \times 10^{-5}\ \exp[(1.26 \times 10^{-5}/(8.31T_b)]$ [11], $C_{L\alpha} = 0.445(933 - T_b) - 1.361 \times 10^{-3}(933 - T_b)^2$ and $C_{\alpha L} = 0.05(933 - T_b)$ [12] at a bonding temperature of 853 K.

The long completion times and the particularly slow rate of movement of the solid–liquid interface during dissimilar MMC/Al_2O_3 bonding (see Figs 2 and 3) confirm that there will be a strong particle segregation tendency when these dissimilar substrates are TLP-bonded [2]. This readily explains the presence of particle-segregated regions in joints produced using 10, 20 and 30 μm foils and holding times ≥ 20 min (see Fig. 4). Also, the solidification rate decreases markedly towards the end of the isothermal solidification stage (see Fig. 3). Because the critical diameter, d_{crit}, for particle segregation increases when the solidification rate decreases [2, 13], larger diameter particles will be pushed ahead of the moving solid–liquid interface at this point in the bonding operation.

In MMC/MMC joints, TLP bonding using a 5 μm thick copper foil produces a liquid film width of 30 μm at 853 K [2]. Few alumina particles are contained within this thin liquid film and can be pushed ahead of the moving solid–liquid interface during the isothermal solidification stage in TLP bonding. In a similar manner, the thin liquid film produced during MMC/Al_2O_3 bonding using 5 μm thick copper foil readily explains the absence of particle-segregated layers in completed joints. When thicker copper foils are used, the width of the liquid film formed at T_b markedly increases and completed joints exhibit particle segregation at the bondline (see Figs 4 and 5). Because particle segregated layers will only become apparent when the liquid width at T_b decreases significantly, dissimilar joints produced using a short holding time (5 min) are free of particle segregation (see Fig. 4).

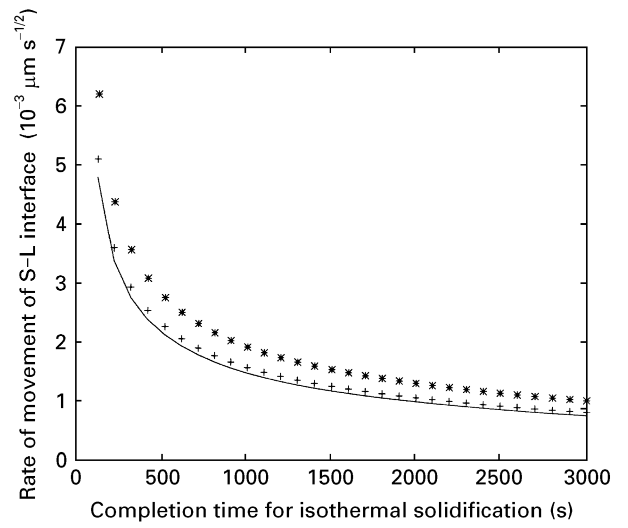


Figure 3 Calculated relation between the rate of movement of the solid–liquid interface and the completion time during isothermal solidification of MMC/Al_2O_3 joints, for bonding temperatures of (—) 823 K, (+) 853 K and (*) 883 K.

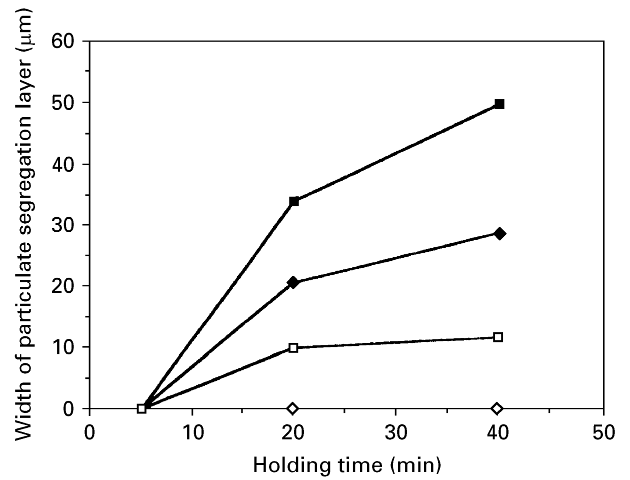
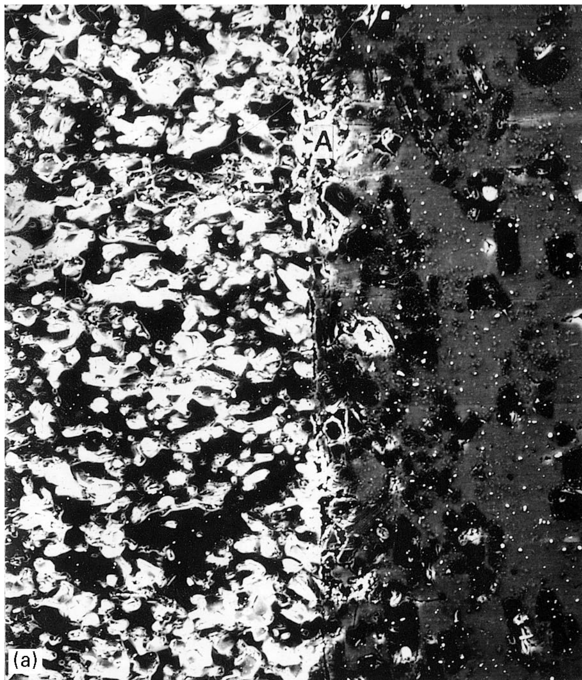


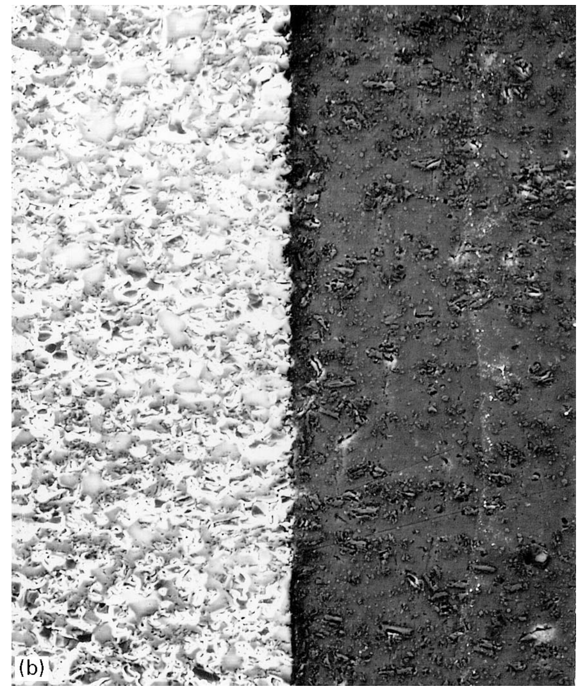
Figure 4 Relation between the width of the particle-segregated layer and the holding time at 853 K, for (◇) 5, (□) 10, (◆) 20 and (■) 30 μm thick copper foils.

3.2. Joint shear strength properties

Fig. 6 shows the average joint shear strengths produced using 5, 10, 20 and 30 μm thick copper foils. Joint strength increased significantly when thin copper foils were applied (the highest shear strength properties were produced using 5 μm thick copper foil. There was no evidence of particle segregation or retained eutectic material on the fracture surfaces of joints produced using 5 μm thick copper foil. In contrast, preferential failure occurred through the particle-segregated layer in joints produced using 20 and 30 μm thick copper foils and long holding times ($\geq$ 20 min) (see Fig. 7).

Joints made using a 10 μm thick copper foil and a holding time of 20 min had similar shear strengths to those produced using 5 μm thick copper interlayers





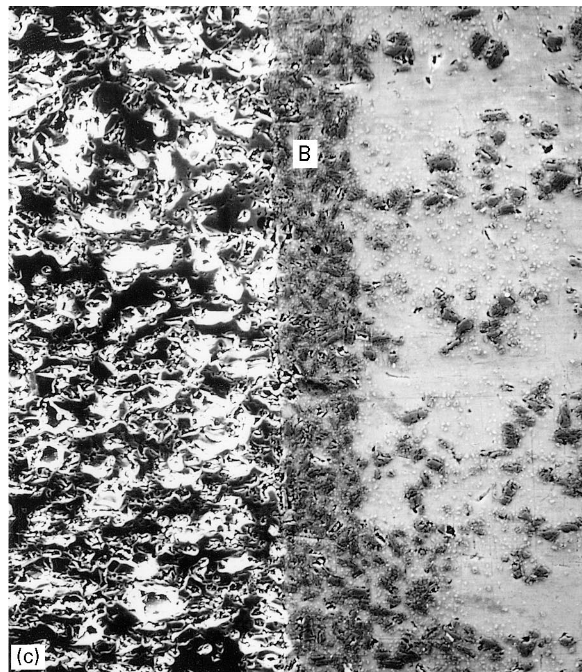


Figure 5 Dissimilar joint microstructures, mag × 220. (a) Retention of eutectic material (A) in a joint completed using a 30 μm thick copper foil and a holding time of 5 min at 853 K. (b) Particle segregation (B) in a joint completed using a 30 μm thick copper foil and a holding time of 40 min at 853 K. (c) Joint microstructure produced using a 5 μm thick copper foil and a holding time of 40 min at 853 K.

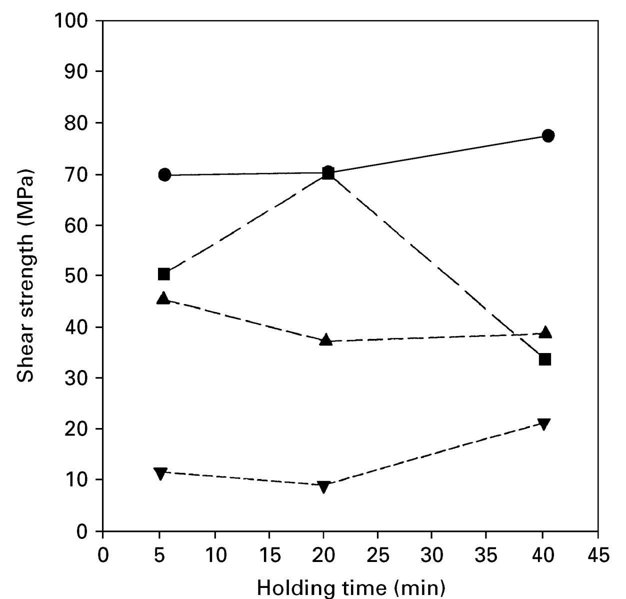


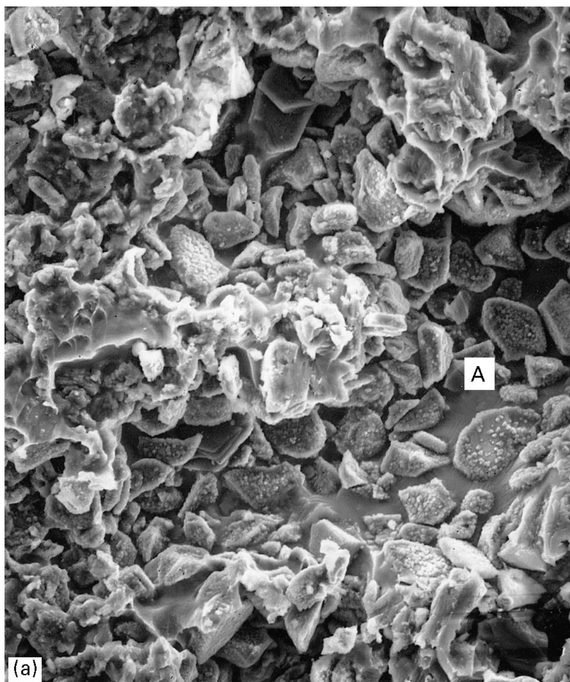
Figure 6 Average shear strength of MMC/Al_2O_3 joints produced using (●) 5, (■) 10, (▲) 20 and (▼) 30 μm thick copper foils, for a bonding temperature of 853 K.

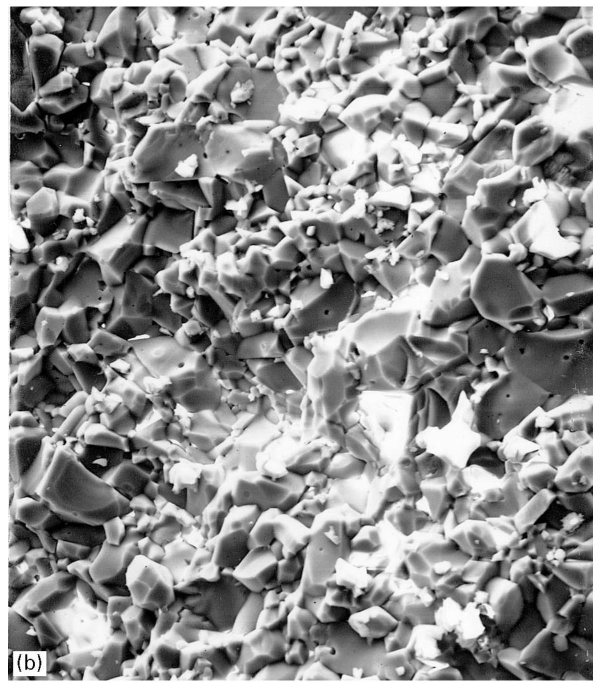
(see Fig. 6). Although these joints contained thin 10 μm wide particle-segregated layers at the bondline (see Fig. 4), there was no evidence of particle segregation or of retained eutectic material on the fractured test samples (see Fig. 7c). In these joints, the mode of joint failure was consistent with preferential fracture through alumina material immediately adjacent to the bondline. Because the thermal expansion coefficients of the Al_2O_3 and MMC substrates are markedly different, large tensile residual stresses are generated at the toe region of the dissimilar joints and this promotes joint failure through ceramic material immediately adjacent to the bondline [14]. Consequently, it is suggested that, when the particle-segregated layer is very thin (< 10 μm), failure is determined by the residual stress distribution in Al_2O_3/MMC joints, not by preferential fracture through the particle-segregated layer located at the bondline.

4. Conclusions

TLP-bonding of MMC and alumina ceramic substrates was investigated. The following conclusions were reached.

1. The completion time is much longer and the rate of movement of the solid–liquid interface is much slower in dissimilar MMC/Al_2O_3 joints because copper can only diffuse into the aluminium-based composite. For this reason, the particle-segregation tendency is markedly increased when these dissimilar substrates are TLP-bonded.





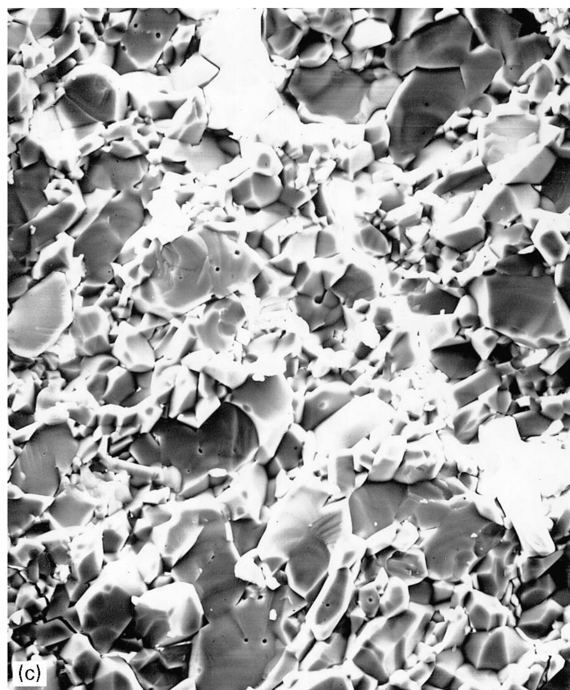


Figure 7 Fracture surface morphologies, mag × 600. (a) Alumina particle segregation (A) on the fracture surface of the joint produced using a 30 μm thick copper foil and a holding time of 40 min. (b) Fracture in ceramic material immediately adjacent to the bondline, in the joint produced using a 5 μm thick copper foil and a holding time of 20 min. (c) Fracture in ceramic material immediately adjacent to the bondline, in the joint produced using a 10 μm thick copper foil and a holding time of 20 min.

2. During TLP bonding at 853 K, the highest joint shear strength properties are produced using 5 μm thick copper foil. When the width of the particle-segregated layer at the joint interface exceeds 10 μm, this region acts as a site for preferential failure during mechanical testing. However, when a thin (< 10 μm thick) particle-segregated layer is formed at the bondline joint, failure occurs in Al_2O_3 material immediately adjacent to the bondline.

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