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Diffusion bonds in copper

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The diffusion bonding of copper is studied experimentally and the results are compared with the predictions of a model of the bonding process. The dominant bonding mechanism is shown to be the power-law creep deformation of contacting surface asperities at high surface roughnesses. However, for smoother surfaces, diffusional mass transfer mechanisms become increasingly more important.

1. Introduction

Previous work of the authors identified the many possible mechanisms of diffusion bonding [1, 2] and proposed a model in which several competing mechanisms led to the formation of a bond. The model calculates the bonding rate of each mechanism. Therefore it can predict the overall bonding rate and also which of the several possible mechanisms dominates as bonding progresses. Also each mechanism is believed to affect the immediate bond interface morphology differently. This study has two purposes; first to determine the rate of bonding in copper and to compare it with the proposed model at different bonding conditions; second, to determine which, if any, of the mechanisms is principally responsible for diffusion bonding copper by examining the shape of interfacial voids present in incomplete bonds.

Copper was then chosen as a suitable experimental material for a number of reasons. First, as one of the objectives of the work was to provide evidence for the theoretical model, a requirement was the use of a material with well-documented physical and mechanical data, (diffusion coefficients, creep and plasticity terms); these are known to a sufficient degree of accuracy for pure copper and are tabulated in Table I. Second, the surface of copper is not protected by a tenacious stable oxide film which could prevent bonding; White and Allen [9] have calculated that, under normal diffusion bonding conditions, copper rapidly dissolves its own oxide. Finally, copper and its alloys can be difficult to fusion weld because of their high thermal conductivities; diffusion bonding is thus of interest as a possible practical joining technique [10].

2. The model

The model defines a perfect diffusion bond as occurring when an unbroken metal/metal contact exists along the join. Normally the faying surfaces are held apart by their roughnesses and initial contact results in the formation of a series of voids along the bonding surfaces, the scale of which is determined by the roughness. It is assumed that the rate-determining step in diffusion bonding is the removal of these voids (or a deformation of contacting asperities) by diffusion and plastic deformation so allowing an intimate contact along the bond [1, 2].

There are many possible bonding mechanisms and the model considers the effects of the following:

1. plastic deformation by yielding or creep;
2. mass transfer by diffusion through the material, along the bond interface (assumed to be a grain boundary) and along the void surface;
3. vapour-phase transport.

Each of these mechanisms is expected to alter the morphology of the bonding interface by flattening the surface asperities differently. The model groups the bonding mechanisms by deformation type, Fig. 1 outlines these deformations in terms of mass transfer around a void formed on asperity contact. Using simple models of aspterity deformation and diffusion, it is possible to build up an

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TABLE I The data required for the model

Parameter		Value for pure copper	Reference
Atomic volume	Ω (m^3)	1.18×10^{-29}	
Burgers vector	b_V (m)	2.56×10^{-10}	
Melting temperature	T_m (K)	1356	
Shear modulus at 300 K	μ_0 (MPa)	4.29×10^4	[3]
Temperature coefficient of shear modulus	$\frac{1}{\mu_0}\frac{d\mu}{dt}$ (K)	3.97×10^{-4}	
Density	ρ (kg m^{-3})	8.96×10^3	
Surface energy	γ (J m^{-2})	1.75	[4]
Bulk diffusion coefficient	D_V (m^2 sec^{-1})	6.2×10^{-5}	[5]
Bulk diffusion activation energy	Q_V (kJ mol^{-1})	207	[5]
Grain-boundary diffusion coefficient	$\delta_B D_B$ (m^3 sec^{-1})	5.12×10^{-15}	*
Grain-boundary diffusion activation energy	Q_B (kJ mol^{-1})	105	*
Surface diffusion coeff.	$\delta_S D_S$ (m^3 sec^{-1})	6.0×10^{-10}	[6]
Surface diffusion activation energy	Q_s (kJ mol^{-1})	205	[6]
P_0 for vapour pressure	(Pa)	1.26×10^5	[7]
Activation energy for vapourization	Q_{vap} (kJ mol^{-1})	324	[7]
Power-law creep constant	A	7.42×10^5	[8]
Power-law creep exponent	n_c	4.8	[8]
Yield stress	σ_y/μ_0	5×10^{-3}	

*Inferred from other fcc data [11].

iterative model of the bonding process and thus predict the relative importance of each mechanism as bonding progresses.

Results from the model are displayed either as a prediction of expected fractional area bonded against bonding time (e.g. Fig. 6) or using a conventional mapping technique [11] (e.g. Fig. 3). The latter maps have axes of two process variables on which contours of the extent of bonding after a certain elapsed time are plotted, the space between the axes is mapped into regions of process conditions within which one mechanism is dominant (i.e. the mechanism which gives the greatest contribution to the bonding rate). Of course the fact that one mechanism contributes a faster individual bonding rate does not preclude the parallel operation of other mechanisms. Often more than one mechanism contributes significantly to void closure.

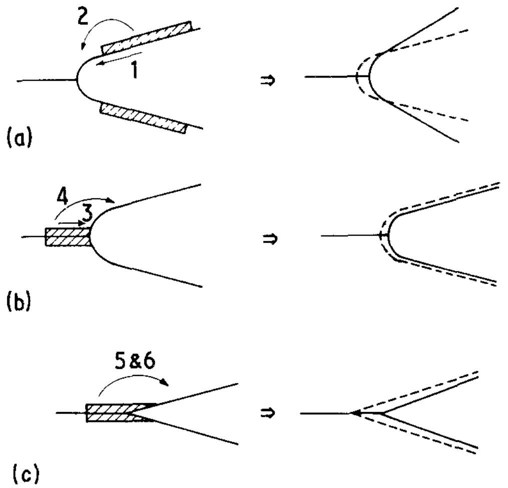

Figure 1 The effect of bonding mechanisms on void shape (as previously shown in [1]. (a) Transfer of material from void surface to growing neck by surface and bulk diffusion; (b) transfer of material from bond interface to neck by diffusion along the interface (grain boundary) and diffusion through the bulk of the material; (c) gross deformation, by plastic yielding or creep, deform the surface ridge but does not create a curved neck.

Each bonding mechanism transports material to deform the surface ridges and hence shrink the interfacial voids in a different manner (Fig. 1). This deformation will directly affect the shape of voids remaining on the interface as bonding proceeds. Thus a study of these voids might reveal whether or not one mechanism is dominant for the bonding conditions used. Naively interpreting the model, one would expect voids closed by diffusion to have rounded necks, whereas those closed by plastic flow to be sharp (Fig. 2); this is implied by the modelled mass transfer.

3. Previous work on diffusion bonding copper

A major problem in using work published before the present study is the lack of information describing the state of the surfaces prior to bonding. The diffusion bonding model that has been

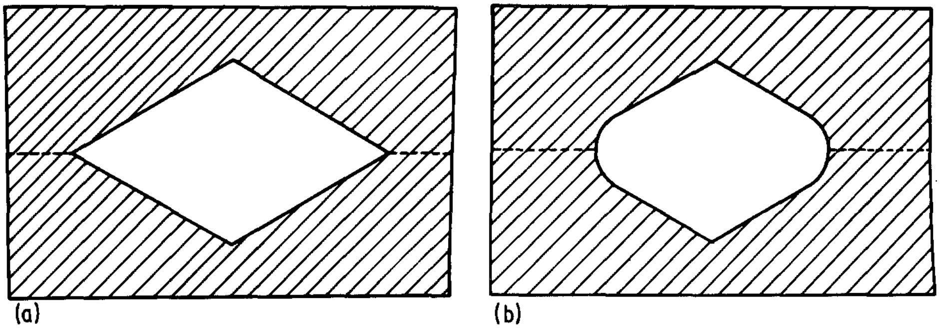

Figure 2 Idealized void shapes assuming only one mechanism of mass transport operates: (a) creep, (b) diffusion.

developed [1, 2] has predicted the crucial importance of surface roughness in determining the time required to produce a good bond. For example, Bradford and Nagpal [12] investigated the temperature dependence of diffusion bonding copper but gave only a qualitative description of their surface treatment, at best only an estimate of their surface roughnesses can be made and hence no valid comparisons with the model are possible.

Ohashi and Hashimoto have produced a comprehensive survey of diffusion bonding oxygen-free high-conductivity copper in a series of papers [13–15]. Unlike most other studies, measurement of surface roughness prior to bonding were made and the roughnesses investigated are listed in Table II. Using surfaces prepared to these roughnesses, Ohashi and Hashimoto investigated the quality of the bond as a function of temperature and bonding pressure. Consequently, their experimental investigations of the effects of temperature, pressure and surface roughness on diffusion bonding can be used to determine the applicability of the model.

The temperature dependence of the bonding process at a constant bonding load was investigated. A series of bonds were made at a pressure of 4.9 MPa and at different temperatures. After 4 min, the bonding was stopped and the fractional area bonded was then determined. Ohashi and Hashimoto's results are superimposed upon the model's prediction of temperature dependence in Fig. 3. The pressure dependence was investigated similarly at a constant temperature of 800° C with identical surfaces. Again comparisons between model and experimental results can be made (Fig. 4). Superimposed upon the maps are predictions of the dominant bonding mechanism for the appropriate bonding conditions.

TABLE II The roughness of the surfaces used by Ohashi and Hashimoto [15]

Roughness height (μm)	Roughness wavelength (μm)
3	60
30	250
70	500
0.1	undefined but estimated as being 10 μm

In general, the results of Ohashi and Hashimoto are in very good agreement with the model. However, a serious divergence between prediction and experimental result occurs at very low surface roughnesses. The variation of bonding time with surface roughness at 800° C is shown in Fig. 5; this shows the onset of divergence at roughnesses (mean peak to valley height of the original surface profile) below about 3 μm. It also can be seen that the divergence occurs when the predicted dominant mechanism is surface diffusion whereas when power-law creep predominates there is good agreement. The surface diffusion model is known to accurately predict the bonding of ferrous alloys [16] and so this suggests that the data rather than the model is at fault for copper. However, Ohashi and Hashimoto did not publish a roughness trace of their smoothest prepared surfaces (unlike for the three rougher cases) and a small change in roughness under these bonding conditions will produce a large change in bonding rate (Fig. 5). Thus any uncertainties as to the correct value of surface roughness will lead to a poor comparison with the predictions of the model. Therefore, either inaccurate surface diffusion data or a bad estimate of the surface roughness for the smoothest surfaces could be the reason for the lack of agreement.

4. Experimental work

A short experimental programme was carried out

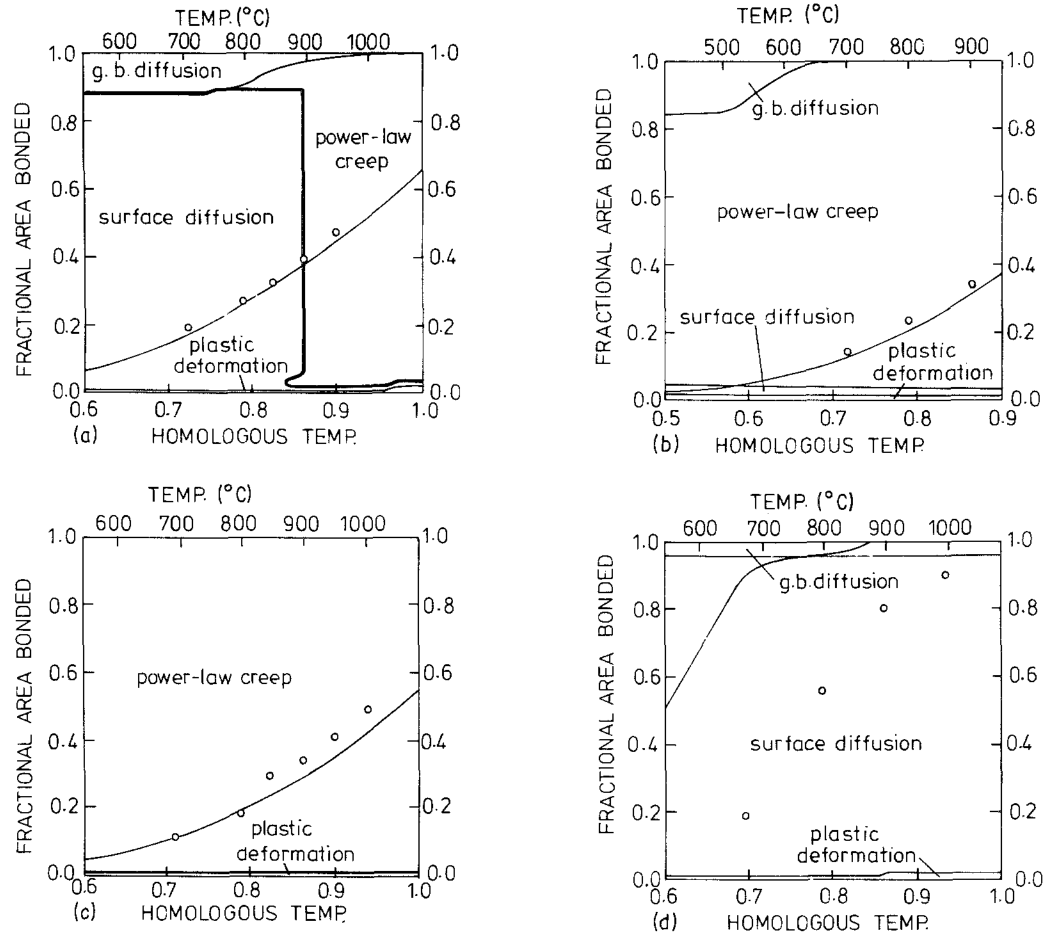

Figure 3 Comparison of the model's prediction (contours) for temperature dependence with the experimental results (points) of Ohashi and Hashimoto [13]. Conditions were: pressure 4.9 MPa, time 4 min and surface roughnesses: Height (μm)/Wavelength (μm) (a) 3.0/60; (b) 30/250; (c) 70/500; (d) 0.1/10.

both to test the accuracy of the model's predictions for copper and to determine whether one bonding mechanism was dominant at the diffusion bonding conditions chosen. The bonds fabricated were sectioned and the extent of bonding determined using optical microscopy. The sections were then examined using a scanning electron microscope (SEM) to ascertain metallographically from the void shapes whether or not there was one dominant bonding mechanism. As has been stated, each mechanism is expected to alter the shapes of the interfacial voids in a different way. In general, diffusional mechanisms transfer material to a growing neck while deformation mechanisms (such as plastic flow from yielding or creep) tend to deform the interface as a whole. Fig. 2 indicates the idealized effect of each bonding mechanism on the bond interface voids; thus it can be seen that an investigation of void morphology could help to identify the bonding mechanisms.

The bonds were made from phosphous-deoxidized copper rods (nominal composition given in Table III) of 12.5 mm diameter and 76 mm length. The surfaces to be bonded were machined flat by

TABLE III Composition of the copper used in the experimental programme

Impurity	Concentration (wt %)
Ag	0.02
As	0.04
Bi	0.03
Fe	0.03
Ni	0.01
O	0.004
P	0.04
Pb	0.01
Sn	0.01

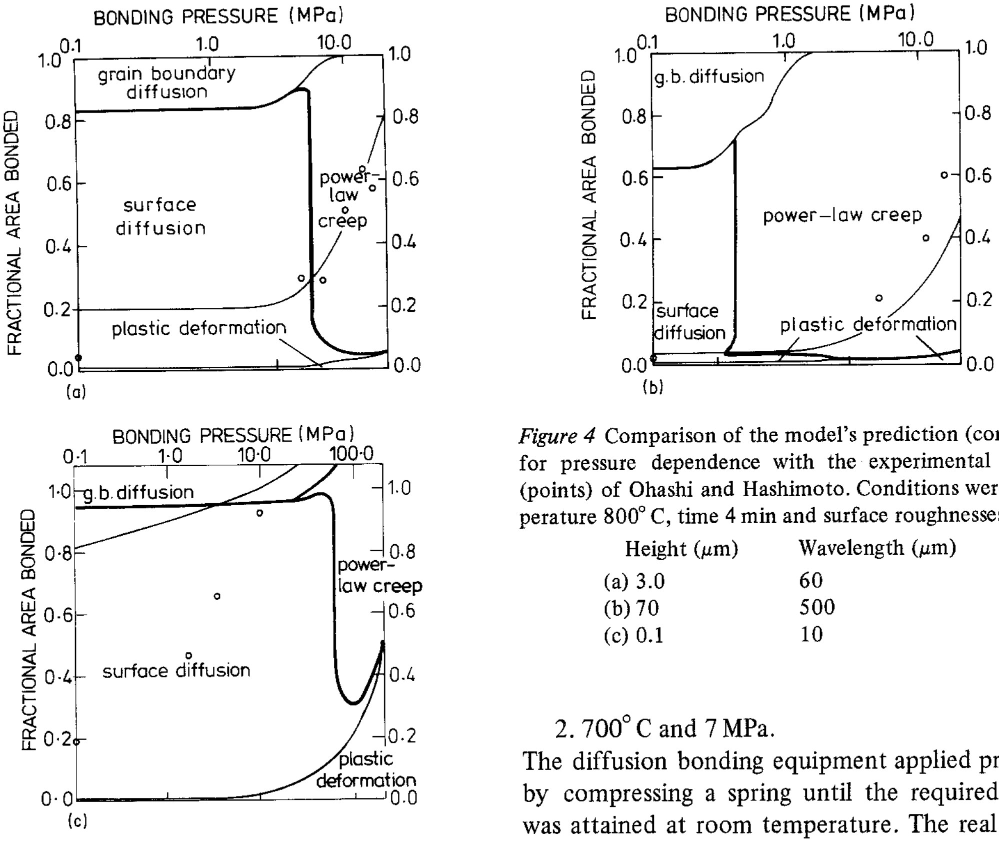

Figure 4 Comparison of the model's prediction (contours) for pressure dependence with the experimental results (points) of Ohashi and Hashimoto. Conditions were; temperature 800° C, time 4 min and surface roughnesses:

Height (μm)	Wavelength (μm)
(a) 3.0	60
(b) 70	500
(c) 0.1	10

off-centre lathing, which resulted in a surface with a mean peak to valley roughness height of 3 μm and a mean wavelength of 40 μm; the machined surface had a very regular profile. The bonds were made for various times at two different process conditions:

1. 550° C and 35 MPa;

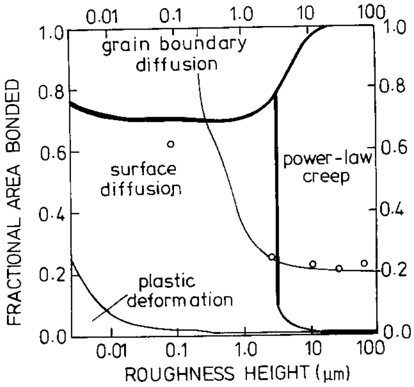

Figure 5 Variation in bonding rate with surface roughness; the model's prediction of the extent of bonding after 4 min (contour) is compared with the results (points) of Ohashi and Hashimoto. Bonding conditions; temperature 800° C, pressure 4.9 MPa, roughness wavelength is taken as ten times the given roughness height.

2. 700° C and 7 MPa.

The diffusion bonding equipment applied pressure by compressing a spring until the required force was attained at room temperature. The real bonding pressure was estimated by taking into account the effects of sample expansion and creep set-down during the bonding cycle. Fig. 6 compares the predictions of the model with the measured extent of bonding as bonding progressed. Despite the large scatter of the results (caused by the small sample cross-sectional area used for bond assessment), a reasonable fit occurs. Fig. 7 shows the evolution of the relative contributions of the three most important bonding mechanisms as bonding progresses. The 700° C bonds, which show worse agreement with the model, have a large surface diffusion contribution.

The bonded interface was examined using both optical and scanning electron microscopy. Low magnification optical micrographs (Fig. 8) show massive grain growth on either side of the interface but the interface remains a near-planar grain boundary. Higher magnification SEM micrographs (Figs. 9 to 11) reveal the residual void shapes.

For the bonds fabricated at 500° C with 35 MPa pressure, power-law creep is predicted by the model to be the dominant bonding mechanism (Fig. 7a). The void shapes observed confirm this prediction as, in general, only sharp necked voids were found. Fig. 9 shows a void after 5 min bond-

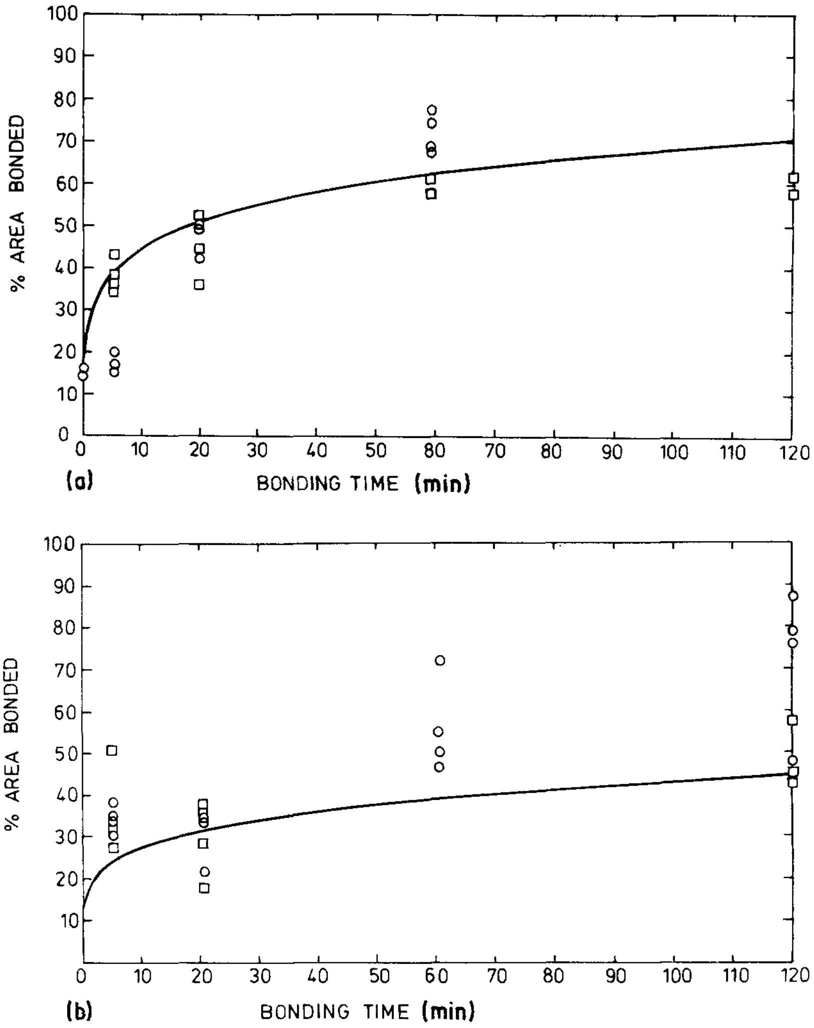

Figure 6 Model prediction compared with experimental measured bonding area. Bonding conditions; surface roughness height 3 μm, wavelength 40 μm, (a) temperature 550° C, pressure 35 MPa; (b) temperature 700° C, pressure 7 MPa.

ing, any circular neck present is of a radius too small to be resolved. The void surface is rough and if surface diffusion had been operating to any great extent some surface smoothing would have been expected. After 120 min the necks were still mostly sharp (Fig. 10a) though it was possible to find slightly rounded necks (Fig. 10b).

The bonds fabricated at 700° C and 7 MPa were expected to have a more complex void shape because the three dominant mechanisms have very similar bonding rates (Fig. 7b). The diffusional mechanisms are expected to create round smooth void shapes. However, although many rounded voids were found, it was also possible to find voids with sharp necks. After long bonding times, the sharp necked voids seemed to predominate (Figs. 11a and b) although the void surfaces are seen to be smooth indicating surface diffusion also occurred. In these bonds, it seems that diffusional mechanisms are operating in conjunction with significant amounts of creep (Fig. 7b) and hence simple deductions from the void shapes are not possible.

5. Discussion

For copper diffusion bonds, the model's predictions agree reasonably with experimental results when the dominant bonding mechanism is power-law creep deformation. However, when diffusional mechanisms become more important, particularly that of surface diffusion, deviations between prediction and results occur. As the surface diffusion

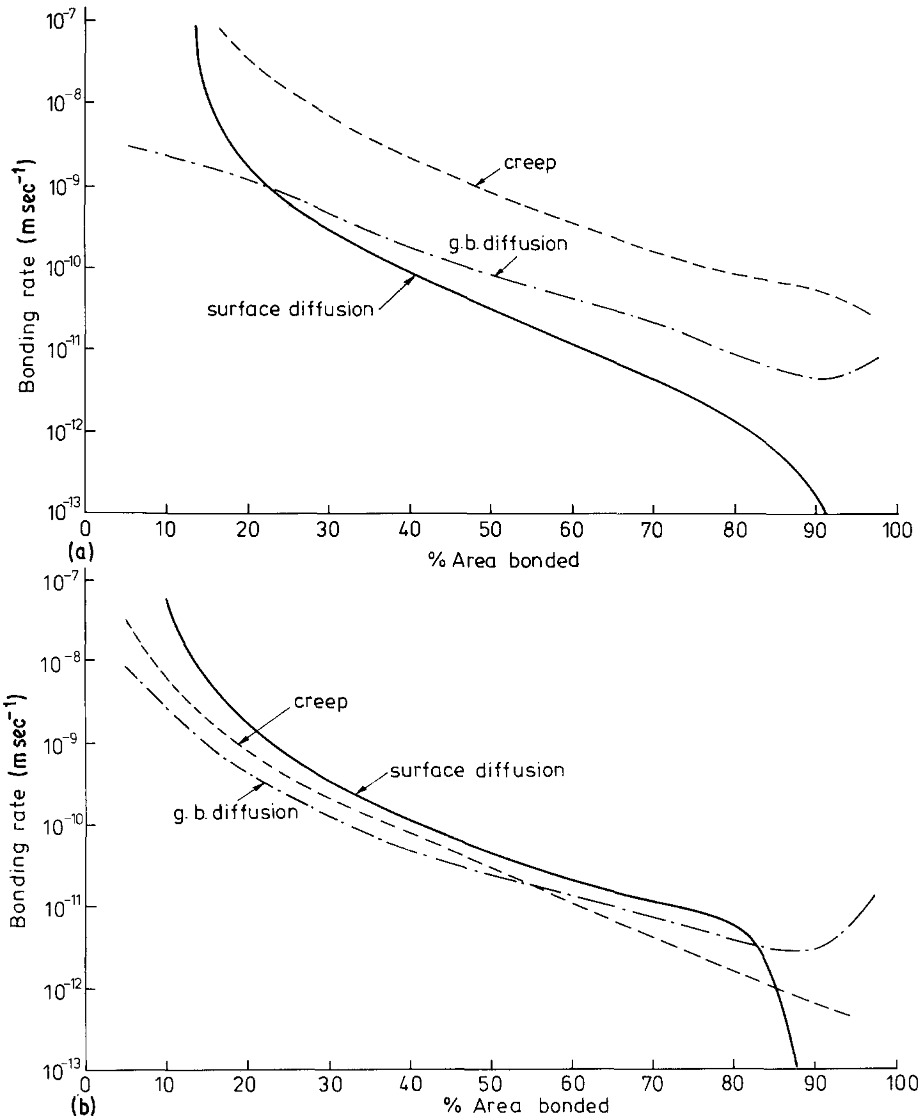

Figure 7 Relative contribution to the bonding rate for each of the three dominant bonding mechanisms. Bonding conditions are as given in Fig. 6: (a) 35 MPa bond, (b) 7 MPa bond.

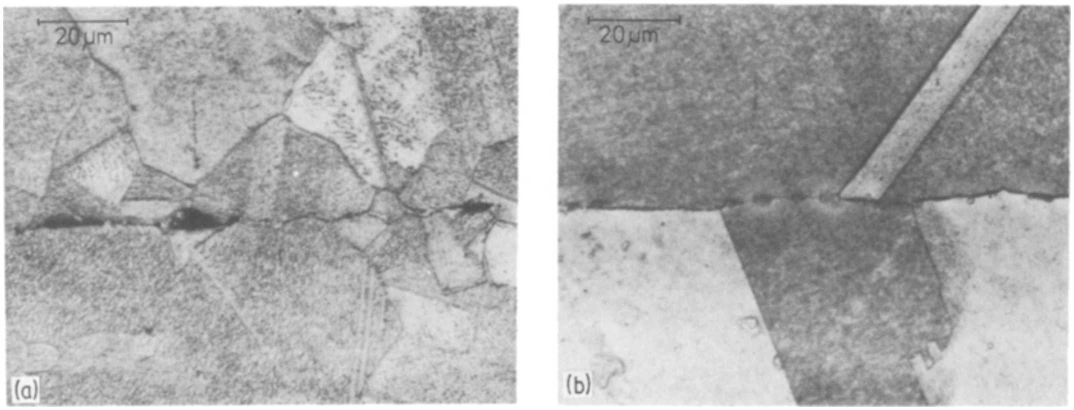

Figure 8 Optical micrograph of bond interface in copper after 120 min at: (a) 550° C and 35 MPa; (b) 700° C and 7 MPa.

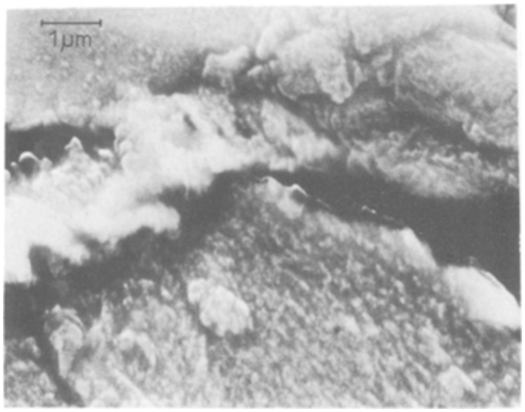

Figure 9 Scanning electron micrograph of interface void after 5 min at 550° C and 35 MPa.

model works well for bonds in pure iron and some steels [16] under conditions of surface diffusion dominance, an error of the model's formulation is not suspected. The differences between model and the bonds made by Ohashi and Hashimoto at low surface roughnesses [13–15] might be attributed to poorly documented surface conditions and therefore are not considered serious. More relevant divergences from prediction are those found with the bonds fabricated by the authors at 700° C, when three bonding mechanisms are operating at very similar rates. The computer model considers each mechanism as completely independent of the effect of others. However, a brief examination of the driving forces of each mechanism indicates that this assumption may not be so. Consider the concurrent operation of creep and diffusional mass transfer. Fig. 1c illustrates the expected action of creep, the asperity is squashed down and out. If diffusional mechanisms are also operating there would be a circular neck (the mass sink) at the edge of the void where the asperities were in contact. The net effect of the creep would be to squash the neck and so reduce its radius. All diffusional mechanisms are driven by a gradient in the local chemical potential, which is dependent on the neck radius as defined by the Joule–Thompson effect. Hence, if the neck radius is reduced by the action of creep, the driving force for diffusion will be greater than expected by the action of diffusion alone. The copper bonds fabricated at 700° C are expected to have creep and diffusion mechanisms of about the same rate (Fig. 7), in these bonds a faster than expected bonding occurred which could perhaps be explained by creep deformation increasing the driving force for diffusion.

6. Conclusions

The reliability of a theoretical model for diffusion bonding has been investigated using single-phase copper specimens. The performance of the model is encouraging at low temperatures and pressures for coarse surface roughnesses. The prediction that, under these conditions, the dominant mechanism will be power-law creep appears to be borne out by an investigation of void morphology. However, at higher temperatures and with very smooth initial surfaces, the model's performance is poorer. When the dominant mechanism is surface diffusion, uncertainty appears to be greatest. The surface diffusion model seems to work well for other metals [16] and is, therefore, not the most probable cause of error. More likely, the surface diffusion data are incorrect, possibly due to con-

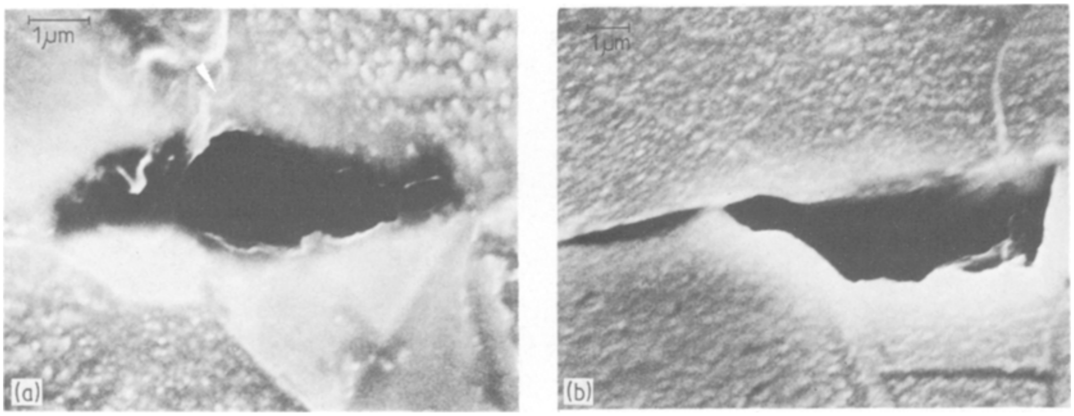

Figure 10 Scanning electron micrograph of interface voids after 120 min at 550° C and 35 MPa: (a) rounded; (b) sharp.

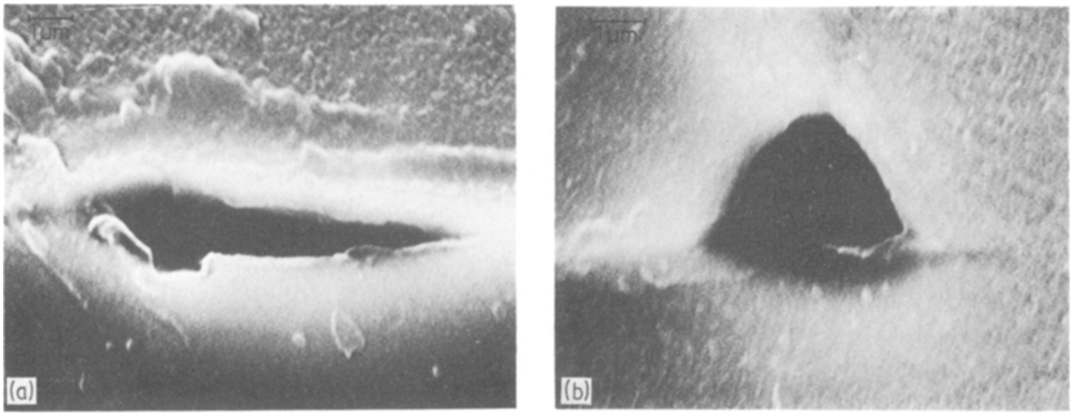

Figure 11 Scanning electron micrograph of interface voids after 120 min at 700° C and 7 MPa: (a) sharp necked; (b) slightly rounded.

tamination of the bonding surfaces. One further possibility, which is suggested from the bonds fabricated at 700° C, is that the various models for mass transfer mechanisms, which have been derived in isolation, do not adequately describe what happens when several mechanisms are operating at similar bonding rates.

Despite these reservations, this study has shown the usefulness of the model [1] as an aid to understanding the diffusion bonding process.

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