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Rolling strain effects on the interlaminar properties of roll bonded copper/aluminium metal laminates

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Metal laminates of copper/aluminium were prepared by roll bonding at 430°C with various rolling strains. The effect of the rolling strain on the interface development and bond strength of the laminates was examined. It was found that as the rolling strain increased the bond strength of the laminates was generally enhanced in both as-rolled and sintered conditions. Critical post-rolling sintering conditions were found to exist for achieving optimum bond strengths of the laminates. It is evident that the development of optimum strength for the laminates is related to the formation of various intermetallic phases at the interface which is in turn determined by the diffusion activity of the metallic elements in the area. The greatest strength enhancement was generally observed to develop in the 60% rolled samples, suggesting that rolling strain of the roll bonding process may impose great influence on diffusion of the metallic elements. A higher copper content, without significant Kirkendall void formation, was found to build up in the interface area of the material, leading to development of strong interfacial phases. It is believed that a higher rolling strain of the roll bonding process has provided a greater area of physical contact between the bonded metals and imposed diffusion enhancement of the metallic elements across the interface. © *2000 Kluwer Academic Publishers*

1. Introduction

Metal laminates have attract significant industrial and research interest in recent years because of their wide applications in engineering areas [1, 2]. Metal laminates can be manufactured by a number of methods and roll bonding is one of the major production methods commonly used in industry [3]. The metal laminates are first bonded through a rolling process. After the rolling stage, sintering heat treatments are usually applied to the laminates to enhance the bond strength [3]. However the complex interface development of the metal laminates in the rolling and sintering stages is still not fully understood.

Peng *et al.* [4, 5] have recently conducted a detailed study on metal laminates under different rolling and sintering conditions. It was found that the bond strength and properties of the laminates are determined by several important processing parameters including rolling temperature, rolling strain and sintering temperature and duration. In a study of the copper/aluminium laminates, Peng *et al.* [4] have identified four different types of phase development in the interface regions of the copper/aluminium laminates during sintering treatments at temperatures between 300 and 500°C. As the sintering time and/or temperature increased, copper-rich phases developed and a Cu_9Al_4 phase became dominant, leading to a high bond strength of the materials. On the other hand, as the sintering time and/or temperature increased, Kirkendall voids were found to became significant with agglomeration of voids, resulting in formation of a weak porous layer in the interface regions. The competing effects of the two interface reactions then determined the fracture behaviour and bond strength of the laminates.

In a study of the rolling temperature on the bond strength development of the metal laminates, Peng *et al.* [5] have, however, observed that the sintering behaviour of the metal laminates was greatly affected by the rolling conditions of the bonding process. Previous studies on the solid state bonding of metals [6–9] have suggested that under the combined action of pressure and heat, over short periods, the reactions between the metal laminates involve a three-stage process of (i) development of physical contact (ii) activation of the surfaces in contact and (iii) interaction between the materials being joined. While a mechanical bond usually forms between the metals during the rolling process, stages (ii) and (iii) allow for the development of a strong metallurgical bond during the post-rolling sintering treatment. It was however found that the development of an optimum metallurgical bonding during the sintering process was dependent upon to the creation of physical contact between the bonded metals [5]. With a high roll bonding temperature the interfacial oxide layer which formed in the rolling stage was retained in the sintering process. The existence of the oxide layer retarded the diffusivity of the metallic elements and affected the formation of intermetallic phases at the interface and degraded the interfacial bond strength of the laminates. On the other hand, there has been no detailed investigation reported, regarding the effect of rolling strain on the interface development of roll bonded materials. This paper reports on the results obtained in a recent study of the effect of rolling strain on the as-rolled and sintered behaviour of copper/aluminium laminates.

2. Experimental

Metal laminates of copper/aluminium were prepared by roll bonding at 430°C, with reductions ranging from ∼20% to 60% in a single pass. The copper and aluminium strips were of initial thickness of 2.6 mm and 1.0 mm respectively. Post-rolling heat treatments were applied to the samples of 40% and 60% rolling reduction at sintering temperatures from 350°C to 500°C for various periods. The bond strengths of the laminates were determined by peel tests conducted on samples of standard dimensions of 100 (L) × 10 (W) mm. Microstructure and interface morphology of the laminates were examined using scanning electron microscopy (SEM). Composition profiles across the interface of the metal laminates were determined using energy dispersive X-ray spectroscopy (EDS) at 15 kV conducted in a scanning electron microscope. Formation of the intermetallic phases at the interface was determined by X-ray diffraction on the as-peeled samples using a Siemens D5000 X-ray diffractometer with CuK_α radiation at small scattering angles of $1°-3°$ and a scan rate of $0.02°\ \mathrm{s}^{-1}$. The divergent slit was set at 1° and the receiving slit at 0.1°.

3. Results

3.1. Bond strength development

3.1.1. As-rolled materials

Fig. 1 shows the peel strength variation of the as-rolled laminates with rolling strain. It was found that peel strength of the laminates was significantly improved with increasing rolling strain. As the rolling reduction increased from 20% to 60%, the peel strength of the as-rolled laminates, measured by the peel test, continuously increased from 11 to 59 N/cm.

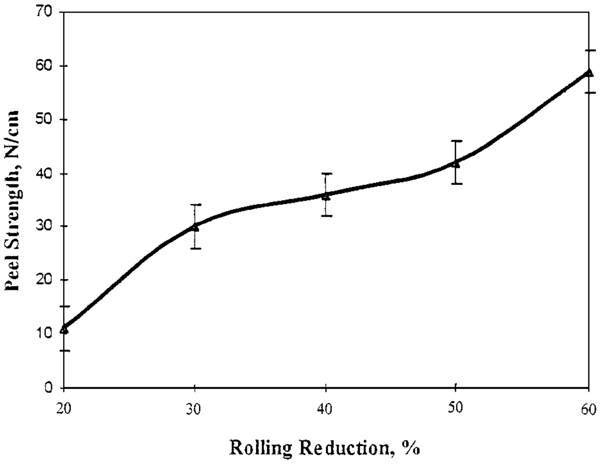


Figure 1 Variation of peel strength of as-rolled laminates with rolling reduction.

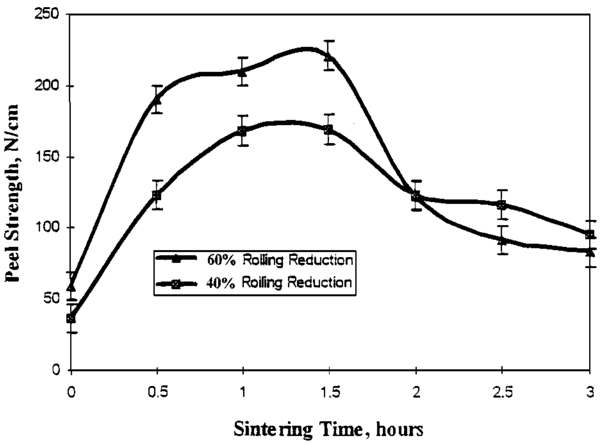


Figure 2 Variation of peel strength of the sintered laminates with sintering time at 450°C.

3.1.2. Sintered materials

Strength enhancement of the laminate materials was evident in the sintered samples. The strength enhancement was more significant in the samples of 60% rolling reduction. Fig. 2 shows the variation of peel strength of the laminates, of 40% and 60% rolling reductions, with different sintering times at 450°C. For the 60% rolled samples, as the sintering time increased, the peel strength of the laminates increased from 59 N/cm in the as-rolled condition to a maximum of ∼220 N/cm after 1.5 hours and then decreased to 83 N/cm after 3 hours. The 40% rolled samples generally exhibited substantially lower peel strengths than the 60% samples except those after a prolonged period of sintering. As the sintering time increased, the peel strength of the 40% rolled laminates increased from 36 N/cm in the as-rolled condition to a maximum of 169 N/cm after 1.5 hours and then decreased to 95 N/cm after 3 hours.

Similar strength development was observed in the laminates with variation in the sintering temperature. Fig. 3 shows the variation of peel strength of the laminates of 40% and 60% rolling reductions at different sintering temperatures for a period of 0.5 hour. For the 60% rolled samples, the peel strength of the samples increased from 100 N/cm at 350°C to a maximum of 216 N/cm at 400°C and decreased to a very low value of 17 N/cm at 500°C. For the 40% rolled samples, the

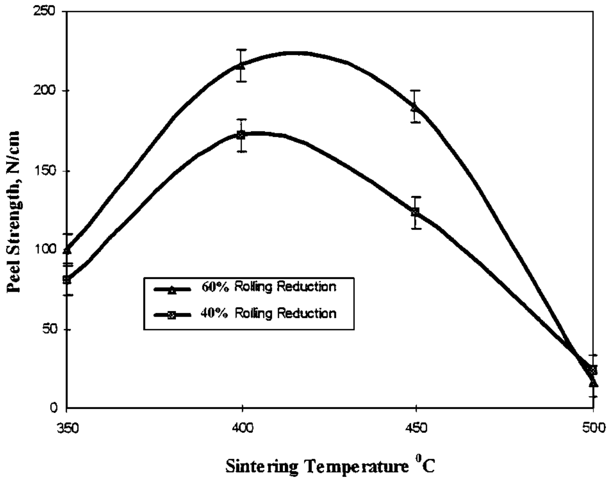


Figure 3 Variation of peel strength of the sintered laminates with sintering temperature for a period of 0.5 hour.

peel strength of the laminates increased from 81 N/cm at 350°C to a maximum of 172 N/cm at 400°C and then decreased to 24 N/cm at 500°C.

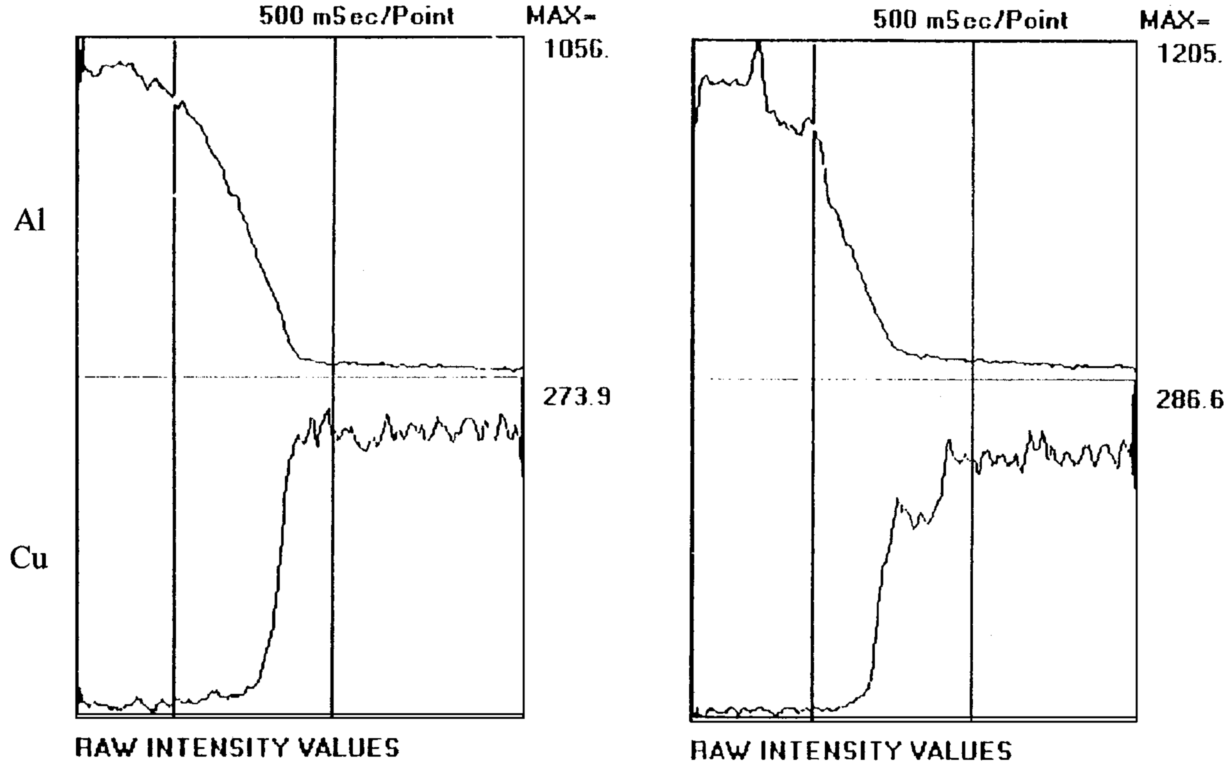
3.2. Interfacial composition profiles

The interfacial composition profiles of the sintered samples were determined using energy dispersive X-ray spectroscopy (EDS) at 15 kV conducted in a scanning electron microscope. Thickness of the interface was determined as the distance between the 5%Cu/95%Al and 95%Cu/5%Al composition boundaries in all the samples. It was found that as the sintering time and/or temperature increased, the interfacial thickness of the laminates continued to increase to high values. For sintering at 450°C, the thickness of the 60% rolled samples continuously increased from ~5 μm, in the as-rolled condition to ~30 μm after the samples were sintered for a period of 3 hours. Variation of the interfacial thickness at different sintering temperatures was also determined. For a sintering time of 0.5 hour, the thickness of the interface grew from ~8 μm at 350°C to ~30 μm at 500°C. Similar trend for interface growth was observed in the 40% rolled samples.

Variation of the interfacial composition profiles was determined as a function of increasing sintering time and/or temperature. The typical composition profiles for the as-rolled materials and sintered laminates at 450°C are shown in Figs 4–6 respectively. In the as-rolled samples, both 40% and 60% rolled laminates showed similar composition profiles of sharp concentration changes across the interface boundaries. Fig. 4, indicating that diffusion of the constituent metallic elements into the bonded area was not significant in the as-rolled materials. After sintering at 450°C for 0.5 hour, a gradual concentration gradient was found in the 40% rolled materials, Fig. 5a. The 60% rolled laminates which were of higher peel strength however showed a different composition profile with higher concentration of copper developed in a wide region close to the copper base metal, Fig. 5b. With further increases in sintering time to 3 hours, gradual changes in the concentration gradients with similar concentration intensity were observed in both 40% and 60% rolled samples, Fig. 6. These samples were of low peel strength.

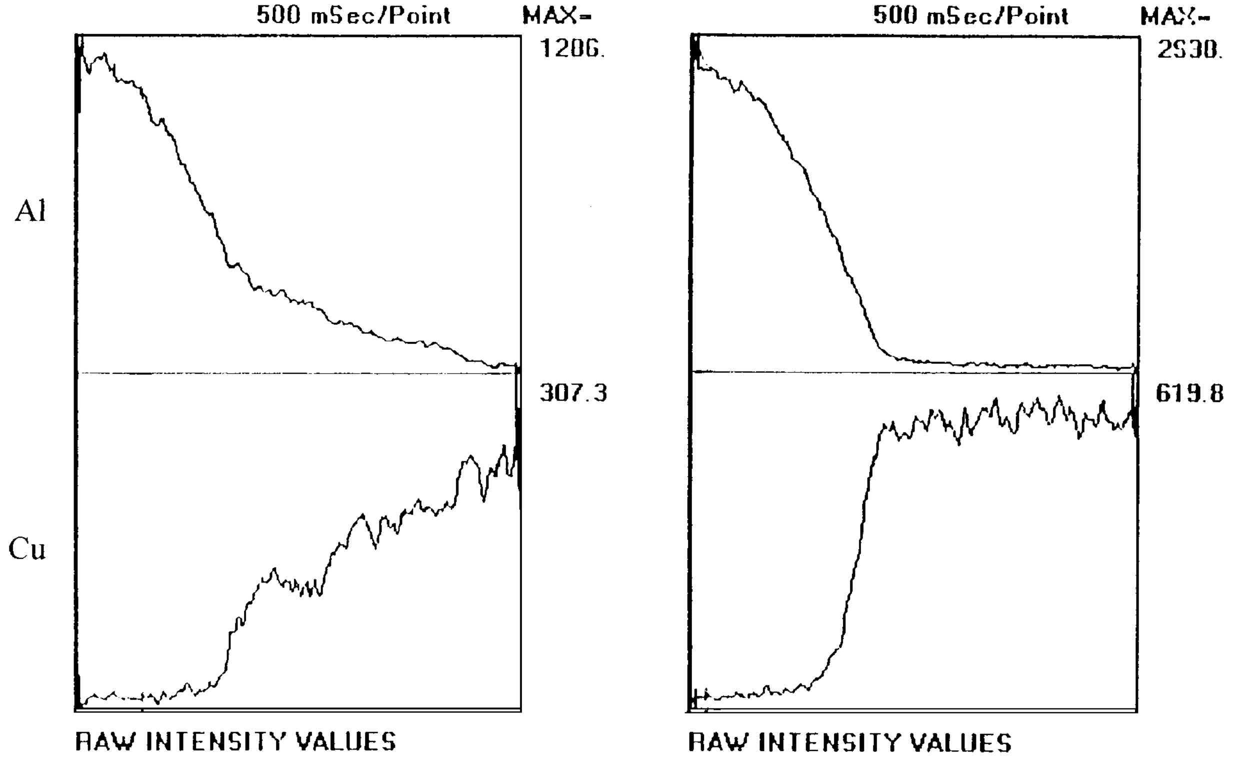
3.3. Fractography

Fracture surfaces of the peel tested samples were examined to identify origins of failure. Scanning electron microscopy of the fracture surface of as-rolled laminates are shown in Fig. 7. Faceted fracture was evident



(a) (b)

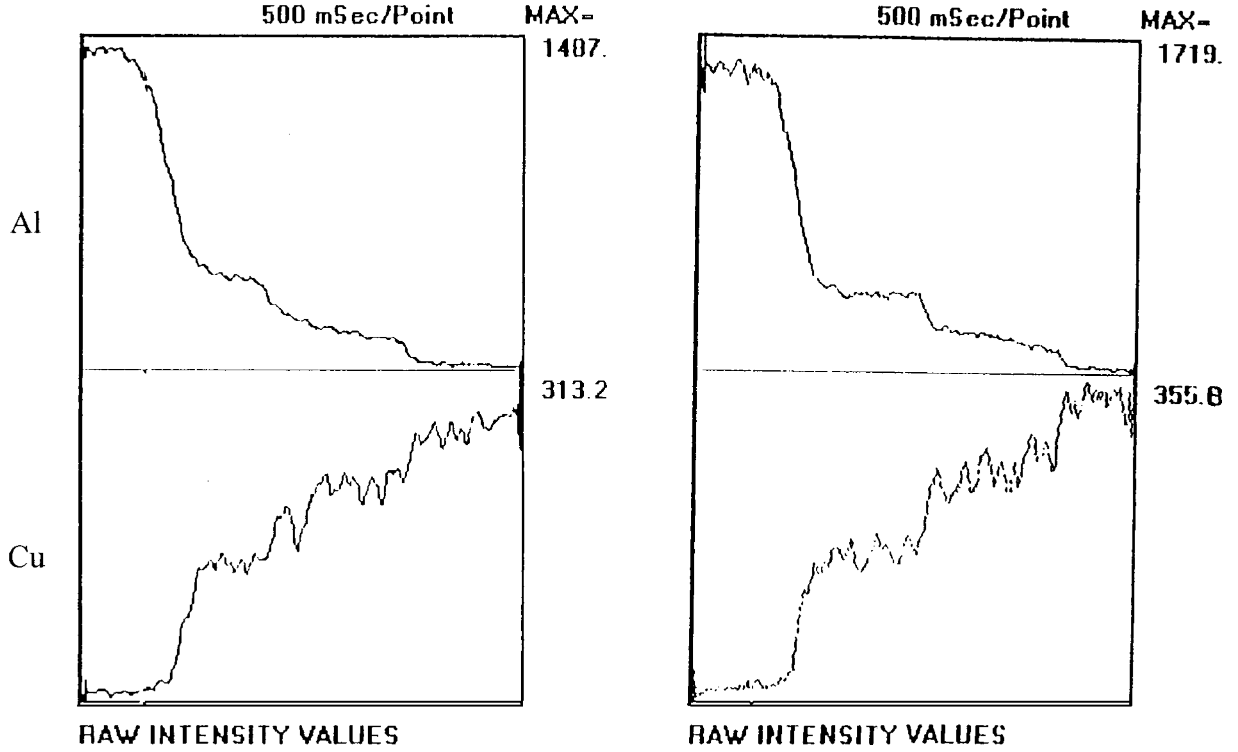
Figure 4 Interfacial composition profiles of the (a) 40% and (b) 60% as-rolled laminates.



(a)

(b)

Figure 5 Interfacial composition profiles of the (a) 40% and (b) 60% rolled laminates after sintering at 450°C for 0.5 hour.

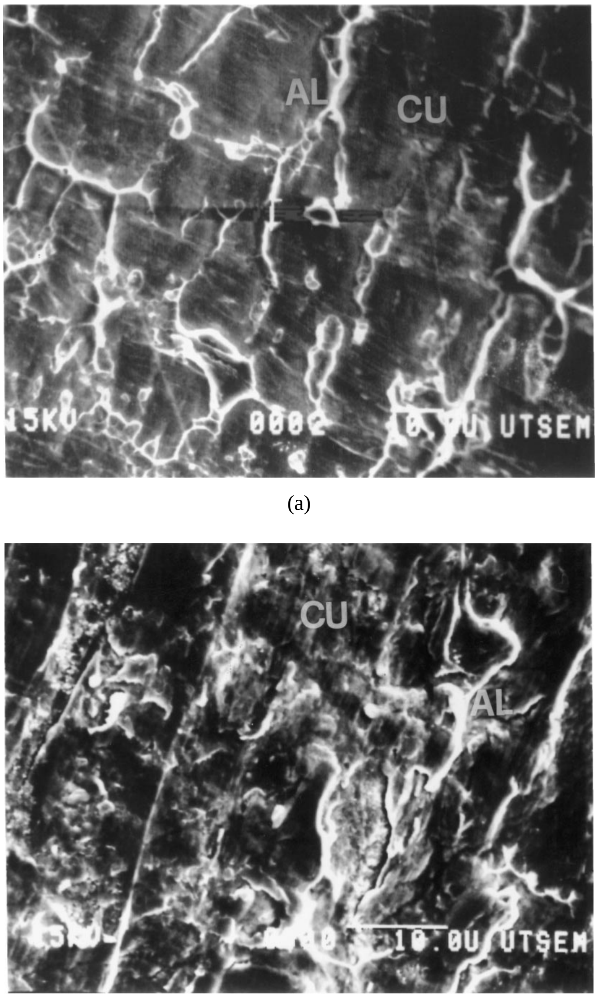


(a)

(b)

Figure 6 Interfacial composition profiles of the (a) 40% and (b) 60% rolled laminates after sintering at 450°C for 3.0 hours.

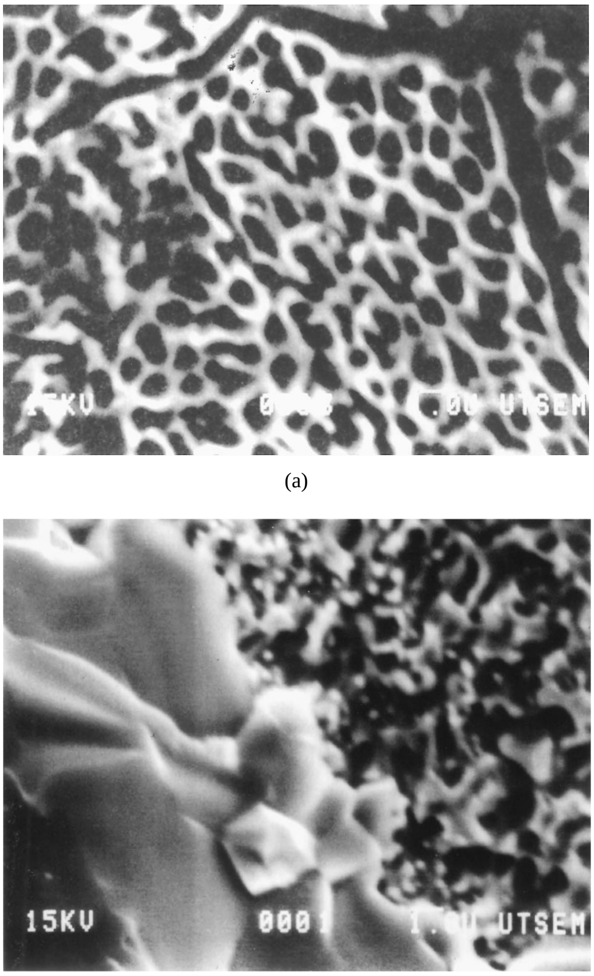
in both the 40% (Fig. 7a) and 60% (Fig. 7b) rolled samples. However whilst a network structure of aluminium was found to adhere to the copper in the 60% rolled samples, only a small amount of aluminium was evident in the 40% rolled laminates. Scanning electron microscopy of the sintered samples of 40% and 60% rolling reductions are respectively shown in Figs 8 and 9. A porous structure consisting of many voids was observed in the samples of lower peel strength in both 40% (Fig. 8a) and 60% (Fig. 9a) rolled laminates after sintering at high temperatures and/or prolonged durations. For the stronger laminates sintered at optimum conditions, cleavage facets were usually dominant with features as those shown in Figs 8b and 9b.



(a)

(b)

Figure 7 Scanning electron micrographs showing fracture morphology of the (a) 40% and (b) 60% as-rolled laminates.



(a)

(b)

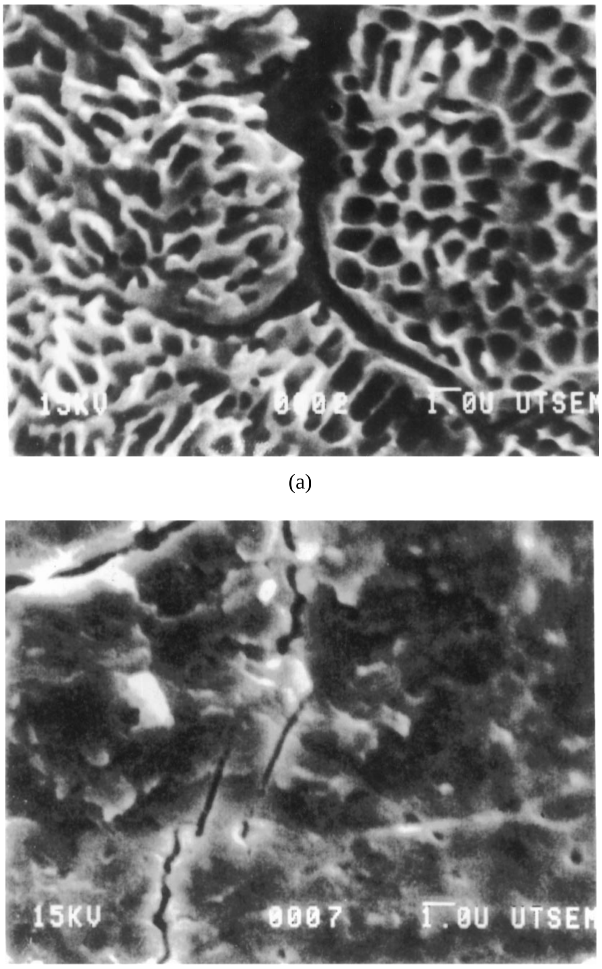
Figure 8 Scanning electron micrographs showing fracture morphology of the 40% rolled laminates after sintering at (a) 500°C and (b) 450°C for 0.5 hour.

However, voids were observed to develop in some of the 40% rolled samples, Fig. 8b whilst cleavage facets covered most areas of the 60% rolled laminates, Fig. 9b. X-ray diffraction measurement further confirmed different intermetallic phases existed in the various samples. Cu_9Al_4-γ phase was found to be a dominant phase in the samples of the higher peel strength.

4. Discussion

Results of the present study show the significant effects of rolling strain on the peel strength of the copper/aluminium laminates. As the rolling strain increased, the bond strength of the laminates was generally enhanced in both as-rolled and sintered conditions. It is apparent that the increase of bond strength of the as-rolled laminates with higher rolling reductions is mainly attributable to the development of a stronger mechanical bonding imposed by higher deformation. Fig. 7b shows much greater physical contact and bonding area between aluminium and copper in the 60% as-rolled materials. On the other hand, the mechanism of strength enhancement of the sintered samples and the effect of deformation strain on diffusion of the metallic elements are found to be much complicated. As the sintering temperature and/or duration increased, the peel strength of both the 40% and 60% rolled laminates initially rose to some maximum values and then dropped to low values after sintering at high temperatures for prolonged periods. Nevertheless a stronger strength enhancement generally developed in the 60% rolled samples under the optimum sintering conditions.

Information about the effect of deformation on diffusivity of metallic elements and strength enhancement in roll bonded laminates is very scarce. However, in pressure welding processes, it has been suggested that several possible causes of diffusion enhancement may occur [10, 11]. These are related to (i) the generation and migration of excess defects during deformation, (ii) short-circuiting along static or moving dislocations or grain boundaries created in the deformation and (iii) short-circuiting along cracks created during deformation. In a study of cold-pressuring aluminium cylinders, Erdmann-Jesnitzer [12] has also reported that the activation energy for diffusion of aluminium elements decreased with increasing deformation. In the present study, it is difficult to identify which of the above mechanisms had occurred in the laminate materials. However it is apparent that diffusion enhancement occurred in the sintered samples of higher rolling deformation. Different composition profiles were found to develop in the interface of the 40% and 60% rolled materials under



(a)

(b)

Figure 9 Scanning electron micrographs showing fracture morphology of the 60% rolled laminates after sintering at (a) 500°C and (b) 450°C for 0.5 hour.

optimum sintering conditions. In the sintered samples that had been rolled 60%, it was evident that copper atoms had diffused more actively into the interface area of the laminates, creating a wide interfacial region of high copper content. With more favourable formation energy, a high content of copper will generally result in formation of the copper-rich Cu_9Al_4 phase [13, 14]. The Cu_9Al_4 phase possesses a much high strength than the aluminium-rich phases as observed [4]. Increasing roll bonding deformation therefore enhanced the interfacial diffusion of the copper atoms in favour of the formation of the copper-rich phases. High peel strengths were achieved in these samples.

A high diffusion rate of copper elements will also accelerate the formation of the Kirkendall voids, eventually leading to degradation of the interfacial bonding of the metal laminates [4]. With more active diffusion of the copper atoms, the sintered samples of the 60% rolled material are expected to have more significant Kirkendall void formation. Scanning electron microscopy on the fracture surface of the tested samples however showed this not to be the case. Under optimum sintering conditions, void formation was more evident in some volumes of the 40% rolled laminates, Fig. 8b, and cleavage facets were generally observed in the samples of 60% roll reduction, Fig. 9b. With lower concentrations of copper as shown in the interfacial composition profiles, it is believed that diffusion of copper atoms might actually be localised in the limited contact positions of the bonded materials in the 40% rolled laminates and created higher void volumes in these areas. Scanning electron microscopy of the as-rolled materials showed that much less physical contact areas were created in the 40% rolled laminates. On the other hand, with a greater contact area between the metals which promoted formation of the high strength copper-rich interfacial phases, a high strength enhancement was maintained in the sintered samples of the 60% rolled materials.

5. Conclusions

Effect of rolling strain of the roll bonding process on the interface development and bond strength of copper/aluminium metal laminates was examined. It was found that as the rolling strain increased, the bond strength of the laminates was generally enhanced in both the as-rolled and sintered conditions. Critical sintering conditions were established for achieving optimum bond strengths of the laminates. It is evident that development of the optimum strengths of the laminates is related to the formation of copper-rich phases at the interface which is in turn determined by the diffusion activity of metallic elements in the area. A stronger strength enhancement was generally observed in the laminates of higher rolling strains under an optimum sintering condition. A higher copper content, but without significant void formation, was built up in the interface area of the material, leading to development of a strong metallurgical bond between the metals. It is believed that a higher rolling strain during the roll bonding process has provided a greater area of contact between the bonded metals and imposed diffusion enhancement of the metallic elements across the interface.

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