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Effect of rolling temperature on interface and bond strength development of roll bonded copper/aluminium metal laminates

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Copper/aluminium laminates were prepared by roll bonding at different temperatures between 350 and 500 °C. The effect of the roll bonding temperature on the interface reactions and bond strength development of the laminates was investigated. It was found that the bond strength of the laminates was generally enhanced with increased roll bonding temperature up to 430 °C. Optimum roll bonding conditions, in terms of maximum bond strength were identified. It is shown that the development of the optimum bonding between the metal laminates is related to the creation of physical contact between the metals in the roll bonding stage and the formation of various intermetallic phases at the interface during the subsequent sintering process. The formation of intermetallic phases is greatly affected by the diffusivity of the metallic elements across the interface. It has been identified that dissolution of the interfacial oxide layer, formed in the roll bonding stage, has a great influence on the diffusivity of metallic elements across the interface which in turn determines the bond strength development of the material.

1. Introduction

Advanced metal laminates have experienced rapid development in engineering applications in recent years [1, 2]. The materials usually possess high specific strengths, enhanced fatigue characteristics and corrosion resistance, resulting in improved service performance at reasonable manufacturing costs. Roll bonding is one of the major production methods commonly used in the manufacture of metal laminates [3]. The bond strength and properties of the laminates are generally determined by several important processing parameters such as rolling temperature, bonding strain and sintering conditions. However, the complex interface development of the metal laminates during the rolling and sintering processes is not yet fully understood.

Recent studies of copper/aluminium laminates [4, 5] show that the bonding strain and post-rolling heat treatment employed in the manufacture of the laminates substantially affect the interfacial phase development and bond strength. It was found that as the sintering temperature and duration increased, the interface continued to grow to a greater thickness. However, growth of the interface did not always enhance the strength of the laminates. A critical sintering condition was found to exist for achieving optimum bond strengths, depending on the formation of various intermetallic phases and the microstructural development at the interface. After sintering at high temperatures and/or for prolonged periods, a porous structure became dominant and led to low strengths [4]. The bonding strain used in the rolling process was found to have imposed a great control on diffusion of the metallic elements and affected the interface reaction and bond strength of the laminates in the sintering process [5]. As the rolling reduction increased, the interface reaction was enhanced to achieve higher bond strengths. The present paper reports another aspect of the study regarding the effect of roll bonding temperature on the interface reaction and bond strength of the laminates. Interface development and mechanical properties of the metal laminates prepared under different roll bonding temperatures were investigated. Relationships between interface development and oxide film dissolution were studied.

2. Experimental procedures

Metal laminates of copper/aluminium were prepared by roll bonding at 350 °C, 430 °C and 500 °C respectively with a 50% rolling reduction 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 then applied to the rolled samples at 300 to 500 °C for various periods. The bond strengths of the composites were determined by peel tests conducted on samples of dimensions 100(L) × 10(W) mm. Microstructure and interface morphology of the composites were examined using scanning electron microscopy at 15 kV. Formation of intermetallic

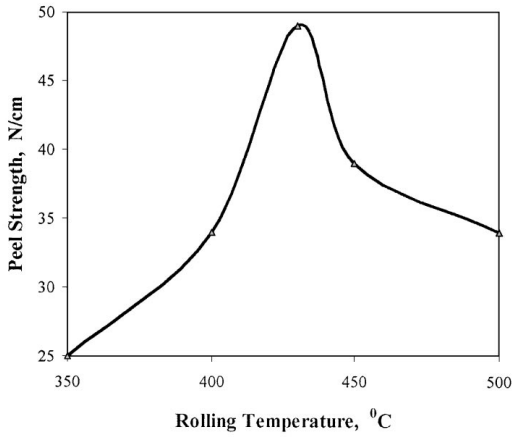


Figure 1 Variation of peel strength of as-rolled copper/aluminium metal laminates with roll bonding temperature.

phases at the interface was analyzed by X-ray diffraction on the as-peeled samples using a Siemens D5000 diffractometer with CuK_α radiation at small scattering angles of 1°–3° and a scan rate of 0.02° s^{-1}. The divergent slit was set at 1° and the receiving slit at 0.1°.

3. Results

3.1. As-bonded materials

Fig. 1 shows the variation in peel strength of the as-rolled laminates with roll bonding temperature. It was found that as the roll bonding temperature increased from 350 °C to 430 °C, the peel strength of the as-rolled laminates increased from 25 N/cm to a maximum of 49 N/cm. When the rolling temperature further increased, the bond strength of the laminates gradually dropped to 34 N/cm at 500 °C.

3.2. Sintering heat treatment

3.2.1. Interface & bond strength development

Interface and bond strength development of the roll bonded materials was studied after sintering at 450 °C for various durations. It was found that, regardless of prior roll bonding temperature, the peel strength of the laminates increased with sintering time to maximum values at 1.5 hour and then decreased (Fig. 2). It was

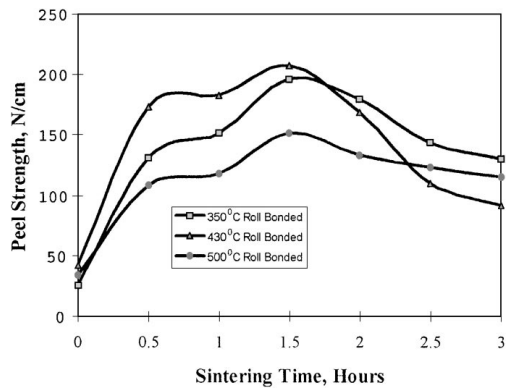
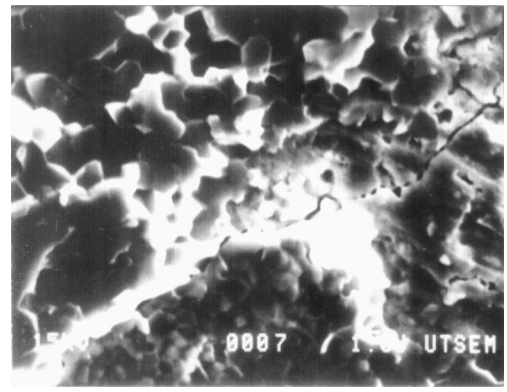
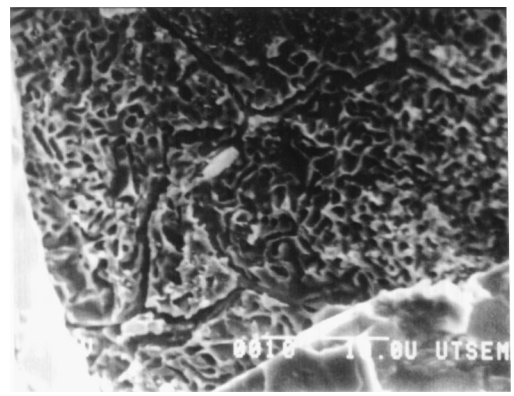


Figure 2 Variation of peel strength of roll bonded copper/aluminium laminates as a function of sintering time.



(a)



(b)

Figure 3 Scanning electron fractographs showing structural development of 430 °C roll bonded copper/aluminium laminates after sintering at (a) 450 °C for 1 hour and (b) 450 °C for 3 hours.

apparent that the 430 °C roll bonded laminates generally had higher peel strength than those bonded at 350 and 500 °C, except after prolonged sintering. As the sintering time increased, the peel strength of 430 °C roll bonded materials rose from 49 N/cm in the as-rolled condition to ∼220 N/cm after 1.5 hour and then dropped to ∼80 N/cm after 3 hours. On the other hand, the peel strength of the 350 °C roll bonded samples increased from 25 N/cm in as-rolled condition to a maximum of 196 N/cm after 1.5 hour and then decreased to 130 N/cm after 3 hours. The 500 °C roll bonded samples generally had the lowest peel strength, increasing from ∼35 N/cm in the as-rolled condition to ∼150 N/cm after 1.5 hour and dropping to 115 N/cm after 3 hours.

The interface microstructure of the laminates was studied by scanning electron microscopy. It was found that the variation in peel strength was closely related to the interface development of the bonded laminates. As the sintering time increased, the interface morphology on the fracture surface changed from a faceted structure to a porous structure after sintering for 3 hours (Fig. 3). The laminates containing a porous structure usually possessed low bond strengths. X-ray diffraction measurement showed that two dominant phases, $CuAl_2$ and Cu_9Al_4, developed in the samples under different rolling and sintering conditions.

3.2.2. The effect of sintering temperature

Fig. 4 shows the variation of peel strength after sintering from 350 °C to 500 °C for 0.5 hour. As the sintering

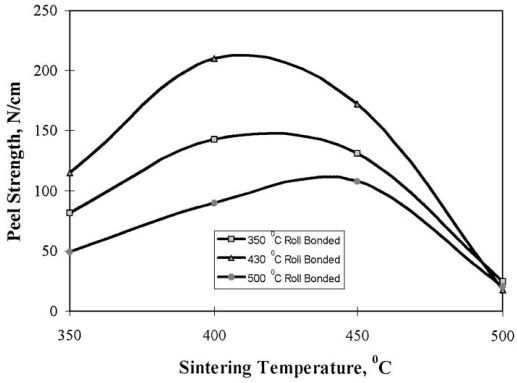


Figure 4 Variation of peel strength of roll bonded copper/aluminium laminates with sintering temperature.

temperature increased from 350 °C, the peel strength of the laminates generally increased to higher values at 400∼450 °C and then decreased substantially after sintering at 500 °C. It was found that as the sintering temperature increased, the peel strength of the 430 °C roll bonded materials slightly increased from ∼110 N/cm at 350 °C to ∼215 N/cm at 400 °C and then substantially decreased to ∼20 N/cm at 500 °C. On the other hand, the peel strength of the 350 °C roll bonded laminates increased from ∼80 N/cm at 350 °C to ∼145 N/cm at 400 °C and dropped to ∼20 N/cm at 500 °C. Lower peel strengths were generally observed in the 500 °C roll bonded material. Peel strength of the 500 °C roll bonded laminates increased from ∼50 N/cm at 350 °C to ∼80 N/cm at 400 °C and then dropped to ∼20 N/cm at 500 °C.

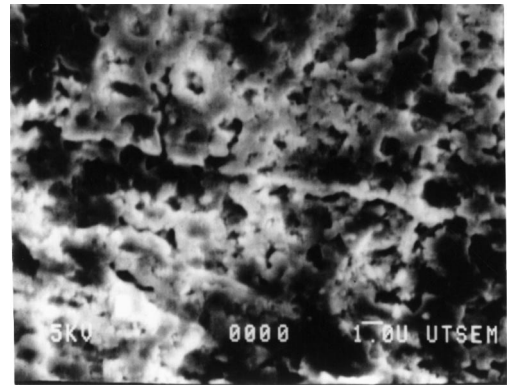
Fractographic examination of the as-peeled samples revealed that the observed low bond strength of the laminate materials was related to the interface microstructures developed at different sintering temperatures. A faceted fracture surface was usually observed in the samples sintered at 400 °C (Fig. 5a), whilst a porous fracture surface became evident in the samples sintered at 500 °C (Fig. 5b).

3.2.3. Dissolution of oxide layer

It was found that the roll bonding temperature greatly affected the formation and dissolution of the interfacial oxide layer. As the roll bonding temperature increased, the thickness of oxide layer formed in the rolling process at the interface generally increased (Fig. 6). After sintering at 450 °C for 1.0 hour, scanning electron micrography of the interface microstructure showed that the oxide layers of the 350 °C and 430 °C roll bonded samples were dissolved whilst the oxide of the 500 °C bonded samples was somehow retained (Fig. 7), indicating that the oxide formed at a high roll bonding temperature was not completely dissolved in the sintering process.

4. Discussion

The effect of roll bonding temperature on the interface and bond strength development of the copper/aluminium laminates was investigated in the



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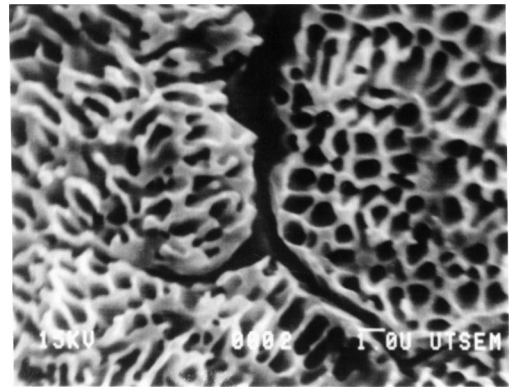
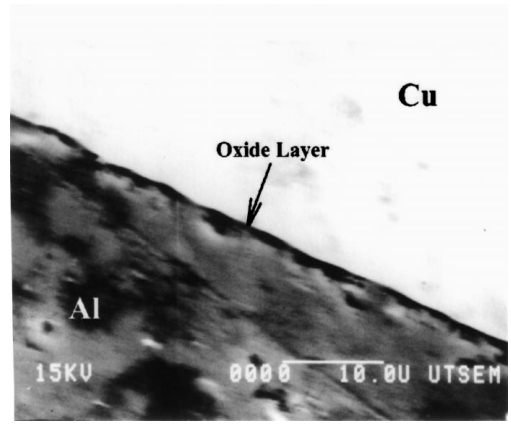


(b)

Figure 5 Scanning electron fractographs of 430 °C roll bonded copper/aluminium laminates showing structural development after sintering at (a) 400 °C for 0.5 hour and (b) 500 °C for 0.5 hours.



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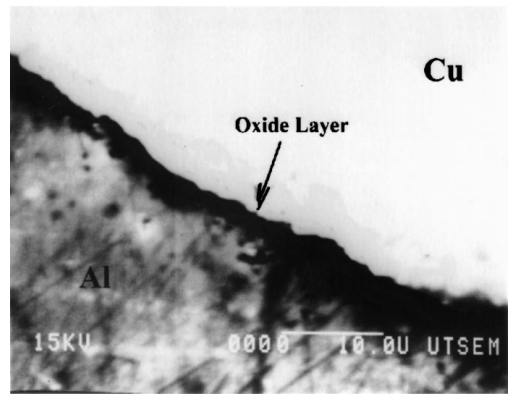
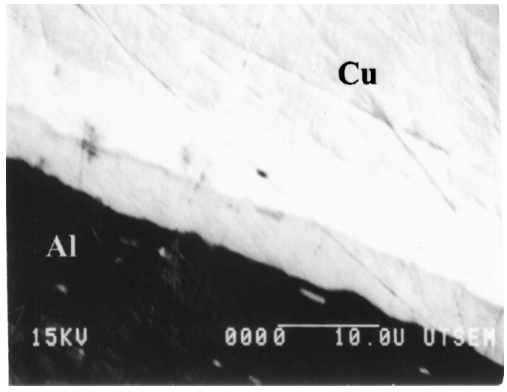


(b)

Figure 6 Scanning electron micrographs showing oxide layer formation at the interface of the as-rolled laminates bonded at (a) 430 °C and (b) 500 °C.



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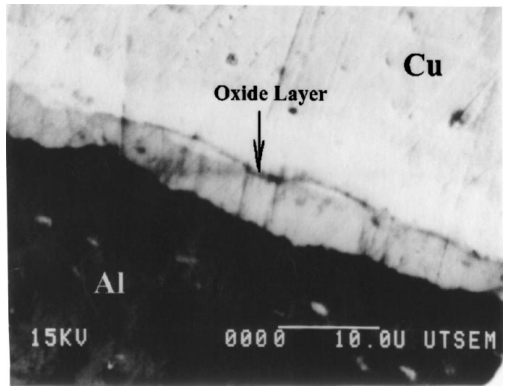

(b)

Figure 7 Scanning electron micrographs showing morphology of the oxide layer after sintering at 450 °C for 1 hour in the copper/aluminium laminates roll bonded at (a) 430 °C and (b) 500 °C.

present study. It was found that the rolling temperature not only affected the as-rolled bond strength but also the bond strength after sintering. An optimum rolling condition at 430°C was identified to achieve the maximum bond strength of the material, indicating a strict process control is necessary in the manufacture of metal laminates.

Previous studies on solid state bonding of metals [6–9] suggest that under the combined action of pressure and heat over short periods, the reactions between the metal laminates involve a three-stage process of (1) development of physical contact, (2) activation of the surfaces in contact and (3) interaction within the materials being joined. It is believed that a similar principle can be applied to the roll bonding process and a strong mechanical bonding is the major contribution to a higher bond strength of the as-rolled metal laminates. It has been also suggested [9] that the attractive force between metallic atoms is proportional to the inverse of their separation. In order to obtain a strong mechanical bonding between the metals, it is necessary to bring the two metal surfaces into close physical contact within the range of mutually attractive forces, to establish a strong mechanical bonding. In the warm roll bonding stage of the present study, the metals will react with atmospheric oxygen to form an oxide film on the metal surface which restricts the physical contact of the two surfaces. On the other hand, plastic deformation would fragment the surface oxide film and develop physical contact between the metal surfaces. The formation and fragmentation of the surface oxide film is indeed a dynamic process occurring in the rolling stage. Shorshorov & Krasulin [7] in a calculation of the pressure parameter, reported that bonding of the material did not happen over the entire surface of the metals to be bonded. The concentration of activation centers of bond was dependent upon the area of physical contact at the interface of the metals. As the roll bonding temperature increases, the tendency of oxide formation on the metal surfaces will increase, reducing the physical contact area between the two metals and weakening the bonding of the laminates. However, an increase in the rolling temperature may lead to a greater degree of recovery and recrystallization of the metals and soften the material. As the metals become softer, the effect of deformation increases, resulting in fragmentation of the oxide layer and formation of a stronger bond between the metals. With these two opposing effects, bond strength of the as-rolled copper/aluminium metal laminates had reached a maximum value at a rolling temperature of around 430°C.

In the manufacture of metal laminates, a sintering process is generally employed to enhance the bonding of the material. A metallurgical bond develops at the interface of the metal laminates in the sintering treatment. Recent studies of aluminium/copper metal laminates by the authors [4, 5] show that three major diffusion controlled processes, involving phase transformation, the Kirkendall effect of void formation and the dissolution of the interfacial oxide layer have occurred during the sintering process. In a sintering treatment, both copper and aluminium atoms will be thermally activated. As the diffusivity of copper in aluminium is greater than that of aluminium in copper [10], a composition gradient of the metallic elements will be created and various phases will therefore form in different regions across the interface. According to the phase equilibrium diagram of the copper-aluminium system [11], eight types of intermetallic phases may possibly form within the composition range developed in the present materials. X-ray diffraction measurements conducted in the related studies [4, 5] show that the $CuAl_2$ and Cu_9Al_4 were the dominant phases detected on the fracture surface of the as-peeled laminates and trace phases of Cu_3Al and Cu_4Al were also occasionally observed in some of the samples. It was also evident that with increasing sintering time, diffusion of metallic elements was enhanced, resulting in formation of copper-rich phases. The copper-rich Cu_9Al_4 phase was found to be of much higher hardness which would contribute to a higher peel strength of the metal laminates. Countering this, the Kirkendall effect of void formation was promoted with prolonged sintering, which then weakened the bond strength of the material. As a consequence, the peel strength of the metal laminates generally rose to maximum values with increasing sintering time and then dropped to low values after prolonged sintering.

The results of the present study show general agreement with the above investigation but the laminates of 430°C roll bonding generally developed optimum peel strengths in the sintering treatment, and those bonded at 500°C possessed much lower strengths. It is believed

that dissolution of the interfacial oxide layer may have affected the interface and bond strength development of the sintered samples. In examination of the interfacial oxide layer, it was found that formation of oxide always developed at the interface area of the metal laminates in the roll bonding stage regardless of the rolling temperature. However, as the rolling temperature increased, the thickness of the oxide layer generally increased and reached a maximum at 500°C. In the subsequent sintering treatment, dissolution of the interfacial oxide layer was evident in the samples roll bonded at lower temperatures. The oxide was decomposed in the samples bonded at 350 and 430°C after sintering for sufficiently long periods. The barrier to the diffusion of metallic elements was thence removed, allowing for formation of strong metallurgical bonding at the interface of the laminates. The results indeed agree with the study of decomposition of surface films by Tylecote and Wynne [12], who reported on the decomposition of oxide films at elevated temperatures. Peshkov *et al*. [13] further suggested that the kinetics of activation and the bonding of the contact surfaces depended on the factors of dissolution of oxides on the metal surface and development of physical contact between the surfaces. A stronger mechanical bond in the as-rolled material and the dissolution of the oxide layer in the sintering process therefore contribute to the much higher bond strengths of the 430°C roll bonded laminates. On the other hand, with a thick layer of oxide at the interface in the 500°C roll bonded material, remnants of oxide are always observed to remain, suggesting that the oxide layer formed at 500°C could not be completely decomposed under a normal sintering treatment. The existence of the oxide layer in the 500°C bonded material in turn retards the diffusivity of the metallic elements and affects the formation of intermetallic phases at the interface. Strength enhancement of the 500°C roll bonded laminates by the sintering process therefore was significantly reduced.

5. Conclusions

Interface reactions and bond strength development of the copper/aluminium laminates prepared by roll bonding at temperatures ranging from 350 to 500°C were investigated in the present study. It was found that the rolling temperature had not only affected the bond strength of the as-rolled material but also that of the sintered laminates. Optimum rolling conditions at 430°C were identified for achieving maximum bond strengths of the metal laminates. The peel strength of the as-rolled laminates increased with the roll bonding temperature and achieved a maximum at 430°C. In the subsequent sintering treatment, the peel strength of the laminates generally increased with sintering time to a maximum and then decreased after prolonged sintering. Nevertheless, the 430°C roll bonded laminates always possessed much higher peel strength against the others and those roll bonded at 500°C had much lower strengths. The development of bond strength in the metal laminates was found to be closely related to the microstructure and intermetallic phase formation at the interface. It is believed that development of optimum bonding between the metal laminates is related to creation of physical contact between the metal laminates in the roll bonding stage and formation of strong intermetallic phases at the interface during the subsequent sintering process. Dissolution of the interfacial oxide layer in the sintering process is identified as having a great influence on the diffusivity of metallic elements across the interface and thus the bond strength development of the material.

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