Effects of Asymmetrical Roll Bonding on Microstructure, Chemical Phases and Property of Copper/Aluminum Clad Sheet

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Abstract

The present paper investigated the morphology and chemical phases of interfacial layer of Copper/Aluminum clad sheet by scanning electrical microscope equipped with energy dispersive X-ray detector and X-ray diffraction, also measured the mechanical property through micro-hardness test. The results are that the interfacial bonding is enhanced and the thickness of interfacial layer increases with mismatch speed ratio rising. The improved interfacial bonding can be found from the tensile fracture. The formation of intermetallic compound (IMC) is promoted by the significant element diffusion at high speed ratio. For sample annealed at 400 °C for 20 min, the formation of IMC is negligible, but the fracture lies between compounds. The micro-hardness on the interface decreases with speed ratio increasing. The study shows that the improvement of microstructure and mechanical property and formation control of IMC of Copper/Aluminum clad sheet can be achieved using asymmetrical roll bonding with high speed ratio.

Introduction

Copper/aluminum (Cu/Al) clad sheet combines the low cost, high specific conductivity and good resistance to corrosion, so has got considerable concern in automobile and electronics industry [1-2]. The cold roll bonding (CRB) is more efficient and economical than other types of processes, such as explosive welding, diffusion welding, roll bonding, and friction stir welding [3-7]. Bonding is achieved when surface deformation breaks up the contamination layers and roll pressure causes the extrusion of material through any cracks present in the fractured surface, with interfacial diffusion promoted by subsequent annealing. However, it is a great challenge to produce Cu/Al clad sheet due to the different chemical and physical characteristics of the dissimilar component metals [8-10].

It has been reported that asymmetrical roll bonding provides a cross shear deformation zone due to the displacement of neutral plane of upper and lower roll, causes a severe deformation for materials and reduces the power consumption [11-13]. some researchers have studied the cold roll bonding process, created several theories about metal bonding, and obtained some key parameters to fabricate laminated composites, including Al, Cu, steel, Al-Ti, Al-Cu, Al-Steel [2].

The formation of intermetallic compound on the interface plays an important role on the bonding and properties of compositions. Especially for the reactive metal of Cu and Al, the formation control of IMC needs much more attention to be achieved. However, the studies about asymmetrical roll bonding is few, and the mechanism of improvement of interfacial bonding, chemical phases as well as the mechanical properties by asymmetrical roll bonding have not been reported thoroughly.

In this paper, the interfacial microstructure, chemical phases and mechanical property of clad sheet produced by asymmetrical roll bonding were studied, through scanning electrical microscope (SEM), energy dispersive X-ray detector (EDX), X-ray diffraction (XRD) and micro-hardness test.

Experimental Procedure

The raw materials used in this study were commercial pure aluminum strips and pure copper strips in fully annealed condition with the specifications given in table 1. The strips were cut into specimens with width of 25 mm and length of 250 mm.

Table 1. Specifications of the component metals strips

Material	Chemical composition (wt.%)	Tensile Strength (Mpa)	Thermal Expansion (10-6/°C)	Hardness (HV)	Thickness (mm)
AA1060	99.6Al	83	23.6	23	1.0
C11000	99.9Cu, 0.04O	280	9.4	63	0.8

The CRB process was carried out using a four-high mill with diameter of both work rolls as 92 mm and a loading capacity of 20 tons. The mismatch speed ratios of lower to upper roll controlled by the transmission gear unit were set to 1.06, 1.19 and 1.31, while the speed of lower roll was constant as 21 rpm.

In order to get rid of the contaminants such as oxides, adsorbed ions, greases, and dust particles, the metal surface was degreased in acetone for 5 min, and then scratched using a rotated brush with 0.3 mm diameter stainless steel wires running at 1200 rpm. The scratching process was essential to remove surface contaminants as well as to create a work-hardened surface layer [6,13-14]. The component metals were stacked together by a soft aluminum wire in terms of copper lying underneath aluminum [15]. The stack combination was fed into the rolling mill without lubrication.

For the release of residual deformation and enhancement of interfacial diffusion, the clad sheet was heat treated in a furnace without protective atmosphere at 400 °C for 20 min, then cooled down to ambient temperature in the furnace [3,16].

The cross-section of samples were ground and polished following the standard metallographic procedures, and etched in a solution of 5 ml HNO_3 + 3 ml HCl + 2 ml HF + 190 ml H_2O. The interfacial microstructure, peeled surface and tensile fracture of clad sheets were observed using scanning electron microscope SUPERSCAN SSX-550. The chemical compositions of clad sheets were performed using energy dispersive X-ray detector with point analysis and line analysis and X-ray diffractomer X'Pert Pro MPD-PW3040/60X. The scanning speed of XRD was at 12 degree per minute, and the radiation was Cu Kα. The Vickers micro-hardnesses of interfacial zone were measured on 402-MVD tester with 25 g load for a dwell time of 25 s.

Results and Discussion

Microstructure of interfacial zone

Figure 1 shows the interfacial microstructure of Cu/Al clad sheet roll bonded at the thickness reduction 48% using asymmetrical roll bonding with mismatch speed ratio 1.06 and 1.31. It is clear to see from figure 1(a) that the interface gap between Cu and Al layer is observable, while it vanishes and interfacial layer appears on some locations in figure 1(b).

The hardened layer caused by scratch on metal surface fractures due to the deformation in CRB process. Then a mechanical mesh can be obtained under enormous roll pressure and friction between metal surfaces. During asymmetrical roll bonding process, there exists a cross shear stress between component metals due to the different metal flow speed. The friction can enhance the surface metal to deform and cause more cracks. Thus, the mechanical mesh between Cu and Al layer can be improved and the interface achieves a tight bonding [4,8,17].

However, the mechanical bonding through mesh depending on the attractive force of metal atoms is weak. Some researchers have investigated the effect of annealing on interfacial microstructure and bonding, and considered the bonding mechanism as the Energy Theory [14,18-19].

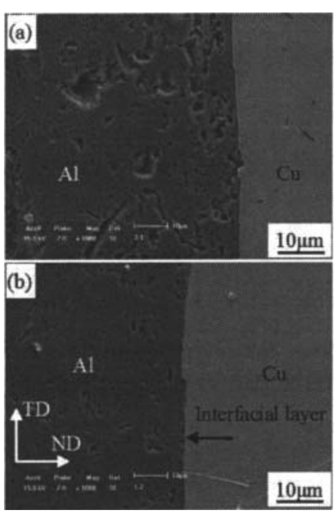

Figure 1. SEM graph of as-rolled clad sheet with different mismatch speed ratio (a) 1.06 and (b) 1.31. TD, ND represents the transverse direction and normal direction in roll process.

Figure 2 shows the interfacial microstructure of Cu/Al clad sheet annealed at 400 °C for 20 min. The interfacial layer grows significantly referring to the as-rolled clad sheet. It can be also found from figure 2(a) that some cracks exist in the interfacial zone of clad sheet roll-bonded with low mismatch ratio, nevertheless the interface in figure 2(b) bonds well and no any visible crack exists there. The crack can be attributed to the poor mechanical mesh caused by asymmetrical roll bonding with lower mismatch speed ratio. In addition, the damage of thermal stress due to the rather different thermal expansivity during annealing aggravates the interfacial bonding. In another way, the oxides formed on the interface during roll bonding and annealing process prevent the interfacial diffusion and lead to the crack of interfacial zone in figure 2(a).

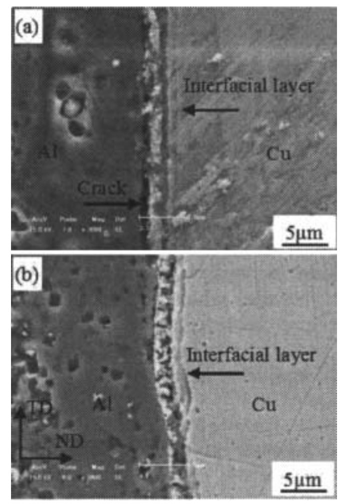

Figure 2. SEM graph of clad sheet with different mismatch speed ratio (a) 1.06 and (b) 1.31 annealed at 400 °C for 20 min.

Microstructure of tensile fracture

The fractographs of tensile specimens are shown in figure 3. It can be found that the interface in figure 3(a) is divided, and the fractured surface shows some tearing microstructure, which can be attributed to the metallurgical bonding of clad sheet during roll bonding and annealing. The fracture in figure 3(b) keeps a whole bulk, and no crack forms on the interface, so the clad sheet is drawn uniformly in the tensile process.

The effect of interfacial friction to metal surface is studied in the previous investigation by some researcher [2,8,11,15,20]. Referring to the interfacial microstructure, a large amount of underlying virgin metal is extruded from the crack of metal surface, caused by the cross shear stress, and then under roll pressure they achieved the atomic bonding due to the atom attraction. During the roll bonding process, more deformation heat is got to promote the interfacial atom diffusion during annealing to obtain metallurgical bonding. Hence, the interfacial bonding of clad sheet produced by the asymmetrical roll bonding with high mismatch speed ratio is improved.

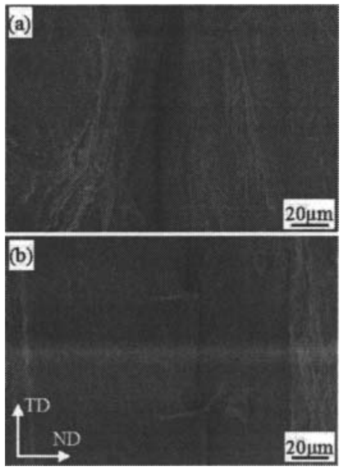

Figure 3. Tensile fractograph of Cu/Al clad sheet with different mismatch speed ratio (a) 1.06 and (b) 1.31 annealed at 350°C for 30min.

Chemical phase on the interfacial zone

The distributions of chemical composition, in the means of line scanning, on the interfacial zone of clad sheet annealed at 400 °C for 20 min are shown in figure 4. It is found the interfacial zone grows with the mismatch speed ratio increasing from 1.06 to 1.31, and the thickness of interfacial layer increases from 3 μm to 4 μm. Also, the microstructure of interfacial layer is changed, and the structure as three-ply is observed in the interfacial zone. According to the profiles of elements distribution, the central sublayer thickness can be found as an increasing trend with the rise of mismatch speed ratio.

The thickness of interfacial layer is determined by the elements diffusion on the interfacial zone. The asymmetrical roll bonding with high mismatch speed ratio provides a severe plastic deformation, and causes enormous heat during the deformation process. The atoms are therefore activated and get more enhancements in the annealing. The severe deformation promotes the interfacial diffusion and increases the interfacial layer [9,13,21].

The chemical compositions of interfacial layer of clad sheet at each sublayer are given in table 2. The proportion of main elements can be calculated, and then the typical compounds may be obtained. The prediction of intermetallic compound of clad sheets with different mismatch speed ratios is listed in table 2. The results show that the chemical composition of central sublayer has a significant change with the rise of mismatch speed ratio.

The chemical composition of interfacial zone varies with the interfacial diffusion, so some intermetallic compounds can be produced among the interfacial zone. The modification of interfacial structure can be attributed to the chemical composition. The results given in table 2 prove the modification of figure 4. The intermetallic compounds can form when the chemical composition fulfills the proportion and thermodynamic condition. Therefore, the elements diffusion promotes the formation of intermetallic compound on interface [22-24].

Figure 5 shows the results of chemical phase on peeled metal surface analyzed by XRD and software X'pert Pro. It is clear that the chemical phase of both metals surface varies with the mismatch speed ratio. On the Al peeled surface, the chemical phase is mainly the intermetallic compound $CuAl_2$, and it increases with the mismatch speed ratio increasing. In addition, there is few Cu phase on the Al peeled surface at low mismatch speed ratio. However, the number of chemical phases on Cu peeled surface rarely varies, except the appearance of few Al phase at high mismatch speed ratio.

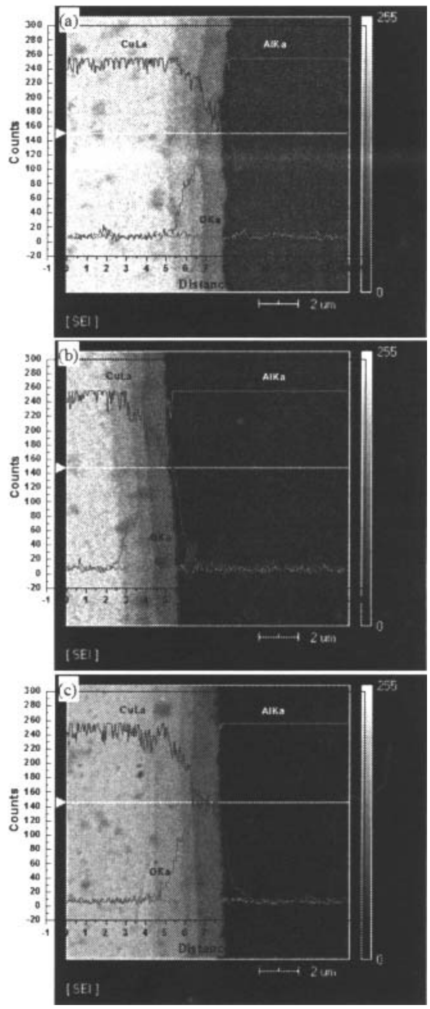

Figure 4. Distribution profile of elements of Cu/Al clad sheet with different mismatch speed ratio annealed at 400 °C for 20 min measured by EDX. (a) (b) (c) represents the mismatch speed ratio as 1.06, 1.19, 1.31, respectively.

In figure 5(a), the intermetallic compound $CuAl_2$ exists on the Al peeled surface, and increases with the mismatch speed ratio increasing. It is can be attributed to the Cu diffusion promoted by the accumulated heat during roll bonding. At low mismatch speed ratio, the element diffusion is limited and the intermetallic compound $CuAl_2$ is few. The interfacial bonding determined by diffusion is weak, thus there is some Cu phase to retain on Al surface during peeled process.

On the Cu peeled surface, the intermetallic compounds have no variation with the rise of mismatch speed ratio, and only Cu_9Al_4

can be measured through XRD. However, a few of Al phase is found to exist on the Cu surface, which can be attributed to the strong interfacial bonding due to the improved diffusion.
According to chemical phases on the peeled surface, it indicates the peeled position lies between the brittle intermetallic compound $CuAl_2$ and Cu_9Al_4. It also shows the diffusion of Cu is larger than Al during annealing [2,25-26].

Table 2. Chemical phases of interfacial layer of Cu/Al clad sheet with different speed ratio

Mismatch speed ratio	Left sublayer		Central sublayer		Right sublayer	
	Al/(Cu+Al) (at.%)	Chemical phase prediction	Al/(Cu+Al) (at.%)	Chemical phase prediction	Al/(Cu+Al) (at.%)	Chemical phase prediction
1.06	31.88	Cu_9Al_4, CuAl, α-Cu	56.82	CuAl, $CuAl_2$	72.64	$CuAl_2$, χ-Al
1.19	39.48	Cu_9Al_4, CuAl, α-Cu	54.69	CuAl, $CuAl_2$	71.82	$CuAl_2$, χ-Al
1.31	39.79	Cu_9Al_4, CuAl, α-Cu	45.43	Cu_9Al_4, $CuAl_2$	67.69	$CuAl_2$, χ-Al

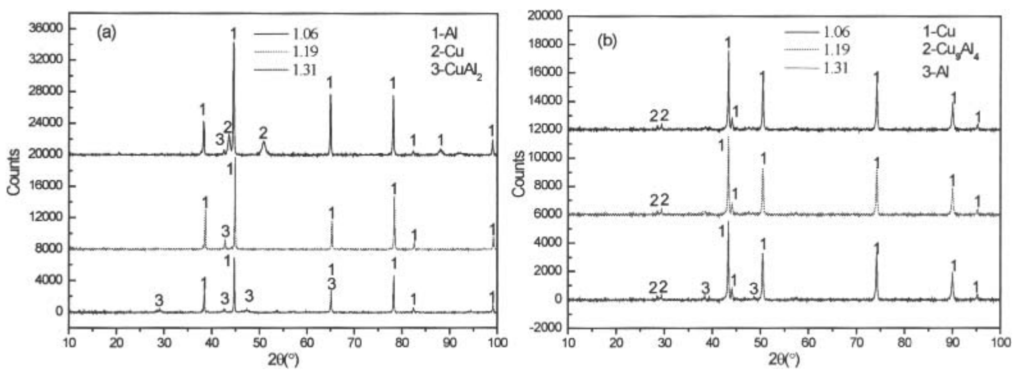

Figure 5. Chemical phases of peeled surface of Cu/Al clad sheet with different speed ratio annealed at 400 oC for 20 min measured by XRD. (a) presents the peeled Al surface, and (b) for the peeled Cu surface.

Mechanical property of clad sheet

In figure 6, the Vickers micro-hardness of interfacial zone is given to show the effect of chemical composition, or even intermetallic compound of Cu_xAl_y. The profiles show that the micro-hardness of interfacial zone is higher than parent metals, which can be attributed to the severe deformation of metal surface and composition change of interfacial zone.
A lot of dislocations generate in the Cu component due to the severe plastic deformation, while for the aluminum with high stacking faulty energy the dislocations are mostly recovered during annealing [4,8,10,27]. Therefore, in figure 6 the micro-hardness of copper layer is much higher than aluminum layer.
According to figure 6, the rise of mismatch speed ratio leads the micro-hardness of interfacial zone to decrease. It is caused by the dynamic recovery during annealing process enhanced by the larger deformation energy of asymmetrical roll bonding.
The intermetallic compounds often have the enormous hardness and brittleness at ambient temperature. From figure 6, it can be found that intermetallic compound rarely has an effect to the micro-hardness of interfacial zone.
The micro-hardness results shows that the clad sheets using asymmetrical roll bonding and low temperature annealing have a good interface without severe brittleness.

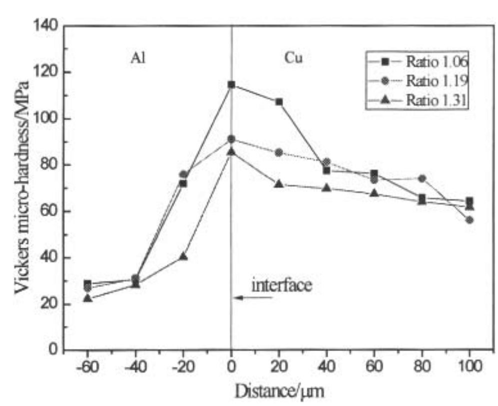

Figure 6. Vickers micro-hardness across the interface of clad sheet with different mismatch speed ratio annealed at 400 °C for 20 min.

Conclusions

(1) The interfacial microstructure of clad sheet is improved due to the severe deformation of metal surface and considerable diffusion during the asymmetrical roll bonding and annealing. The thickness of interfacial layer for annealed clad sheet with mismatch speed ratio 1.31 is ~4 μm, while that with speed ratio

1.06 is ~3 μm. The tensile fracture shows an improvement of interfacial bonding due to the severe deformation on the interface with high mismatch speed ratio.
(2) The structure and chemical phase in the interfacial layer varied with the mismatch speed ratio, which is attributed to the promotion of deformation heat.
(3) The intermetallic compounds $CuAl_2$ and Cu_9Al_4 exist on the peeled Al and Cu surface, respectively, and increase with the rise of mismatch speed ratio. The peeled position lies between the two intermetallic compounds.
(4) The Vickers micro-hardness of interfacial zone is higher than component metals, while it decreases with the rise of mismatch speed ratio due to the recovery caused by generated heat on interface.

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