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ARTICLE

Effect of Interface on Microstructure and Mechanical Properties of Cu/Al Laminated Composite Produced by Asymmetrical Roll Bonding and Annealing

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Abstract: The interfacial microstructure and fractography of Cu/Al laminated composite fabricated by asymmetrical roll bonding and annealing were studied by scanning electron microscope. The peeling and tension tests were carried out to investigate the mechanical properties of interface. The results show that the interfacial interlayer is improved by heat treatment and the higher annealing temperature weakens the interfacial bonding. The tensile properties of laminated composites fall in between two metal components. After annealed at 340 ℃, copper substrate is nearly equal to aluminum substrate in ductility, and the crack degree of interface is lower. The mismatch elongation of metal substrates leads to the internal fracture of interlayer during tension tests. The interface plays an important role in strengthening the laminated composite as a transition between copper and aluminum.

Key words: interface; tension; laminated composite; asymmetrical roll bonding; annealing

Laminated composites have been widely used in electronics and automobile industry. Mechanical properties of homogenous material can be improved by combination of different materials[1-3]. Cu/Al laminated composite has been attracting much attention on theoretical and experimental studies due to its cost reduction, corrosion resistance and light weight compared with pure Cu[4,5]. Several techniques are utilized to fabricate laminated composite, such as thermal spraying, diffusion welding, roll bonding and explosive cladding. However, the cold roll bonding (CRB) is considered to be a more efficient and economical process[6-8]. Two component metals are deformed and their surface contamination layers are broken up. Then, underlying virgin metal is extruded through the cracks present in the fractured films causing two surfaces to bond together[9,10]. As we know, annealing treatment after CRB is an effective process for removing work hardening and results in metallurgical bonding, but causes the formation of brittle intermetallic compounds to damage the interfacial bonding, such as $CuAl_2$, CuAl and Cu_9Al_4. Consequently, it is worth studying to determine an appropriate annealing condition for metallurgical bonding without reduction in bond strength[11-14].

Nowadays, many researches have indicated that the interface provides a transition to different ductility between copper and aluminum during the tension process. Since the interface plays an important role in plastic deformation of laminated composite, it is crucial to evaluate the influence of interfacial interlayer on mechanical properties of laminated composite[15-18]. However, the research methods on homogenous material are always unsuitable for composite material. The annealing process after rolling changes the microstructure and mechanical properties of not only the metal substrates but also the interface[19-21]. The previous experimental results are always difficult to illustrate the effect of the interfacial interlayer. Thus, some new research methods should be constituted to investigate the effect of interface without consideration of metal substrates[22].

In this research, the tension tests of individual copper and aluminum clad were conducted, and the influence of interface on mechanical properties was analyzed according to

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the interfacial fracture microstructure and deformation behavior of the laminated composite and metal components. Based on the interface strengthening effect, some discussions and conclusions were obtained to make an appropriate annealing process for Cu/Al laminated composite produced by asymmetrical roll bonding.

1 Experiment

The commercially pure aluminum (A1100) and copper (C11000) strips with thickness of 1 mm, width of 25 mm and length of 130 mm were used to roll bonding. The specimens were degreased in acetone and then scratched by a steel brush to clean the surfaces. The CRB experiments were carried out using an asymmetrical four-high reversing mill with a work-roll diameter of 92 mm without any lubrication. The asymmetrical roll bonding was conducted at a speed ratio 1.2 of lower to upper roll. The rotation speed of lower work roll was set as 20 r/min. The final thickness of bonded sheet was 0.7 mm.

After rolling, some individual copper and aluminum strips needed to be prepared for comparison experiment, because only studying on tensile properties of the bonded sheet could not interpret the effect of the individual component and interfacial interlayer. In order to ensure a consistent processing condition, some bonded specimens were separated along the interface as individual strips to eliminate the interfacial interlayer. The clad sheets and separated strips were annealed at 300, 320, 340, 360 and 400 ℃ for 30 min in a resistance furnace, and then cooled down to room temperature in air.

The interfacial microstructure and fracture morphology of specimens were observed by a scanning electron microscope SUPERSCAN SSX-550. The tension and peeling tests were conducted on a SANSCMT 5000 materials testing system at room temperature. The loading direction is parallel to the roll direction. The gauge of tensile specimens is 10 mm in width and 15 mm in length. The nominal strain rate in the tensile test was set as 3.3×10^{-3} s^{-1}. Tensile strength and elongation were obtained based on the stress-strain curves by the software Origin. The clad sheets were also evaluated for their bond strength through peeling tests using the SANSCMT 5000 with a constant speed of 10 mm/min. The dimension of peeling specimens is 10 mm in width and 50 mm in length. Tensile and peeling specimens were made from the center of clad sheet along the rolling direction according to ASTM D1876-08.

2 Results and Discussion

2.1 Interfacial microstructure

Fig.1 shows the interfacial microstructure of Cu/Al laminated composite annealed at different temperatures for 30 min. There is no obvious diffusion layer on the interface annealed at 300 ℃. Then, a discontinuous interlayer with a certain thickness can be observed between the two metal substrates when the annealing temperature reaches 320 ℃. Moreover, a continuous and wider interlayer forms on the interface with the annealing temperature exceeding 340 ℃. According to the diffusion layer shown Fig.1d and 1e, a three-sublayer structure is presented on the interface annealed at higher temperature. The width of the interfacial interlayer increases significantly with increasing the annealing temperature. The measurement results of average interfacial width are shown in Table. 1. It is apparent that the width of the interfacial interlayer increases significantly with increasing the annealing temperature.

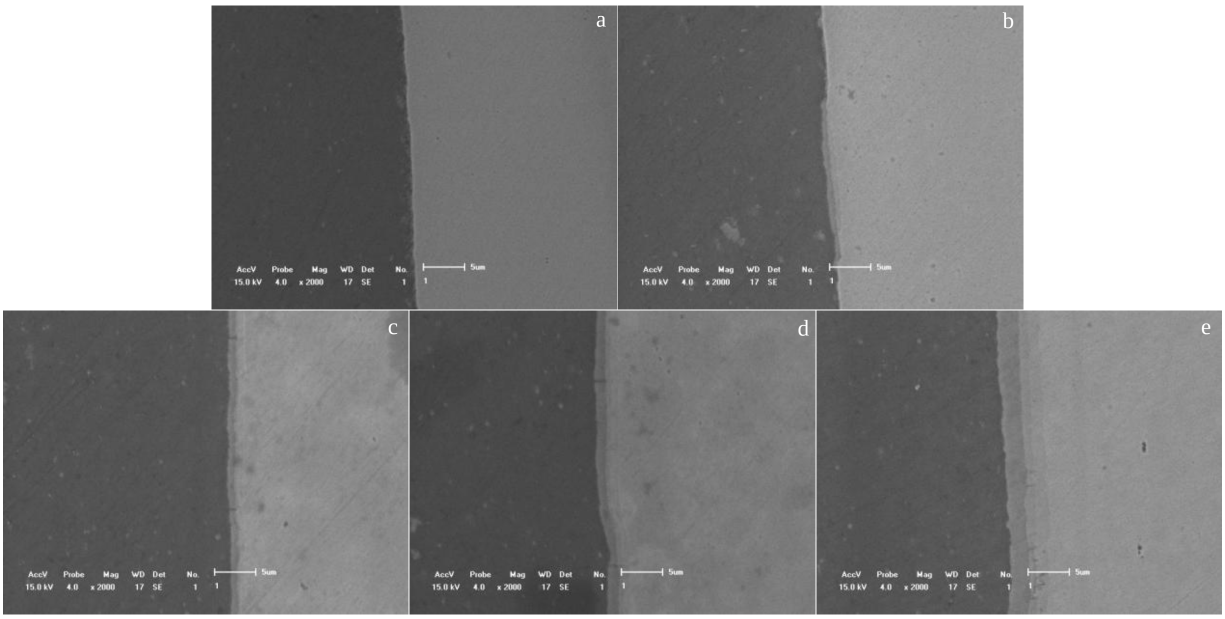

Fig.1 SEM interfacial microstructures of Cu/Al clad sheet annealed at different temperatures: (a) 300 ℃, (b) 320 ℃, (c) 340 ℃, (d) 360 ℃, and (e) 400 ℃

Table 1 Average width of interface annealed at different annealing temperatures

Annealing temperature/ ℃	300	320	340	360	400
Average width/μm	0	1.07	1.43	2.14	5.36

During asymmetrical roll bonding, the mismatch rotation provides a severe shear deformation and deformation-induced heat accumulation. The plastic flow of metal substrate along rolling direction results in fracture of the hardened layer on metal surface, and then the underlying virgin metal is extruded through the cracks. Hence, the mechanical bonding is established at the points where both component metals are extruded. Since the asymmetrical roll bonding process with a single pass and heavy reduction has been utilized, the rolled specimens need to be annealed to release the residual stress and work-hardening. In addition, the interfacial diffusion causes the establishment of metallurgical bonding and develops the interlayer. Low temperature annealing cannot provide enough diffusion activation energy to form the diffusion layer, as shown in Fig.1a. During the annealing process at 320 ℃, the thermal diffusion occurs mainly at the points of good mechanical bonding, which makes the diffusion layer discontinuous. With the annealing temperature being increased, the metal atoms near the interface obtain more energy and move to the opposite metal substrate, and then a continuous and wider interlayer covers the whole interface. The EDS results of interfacial interlayer are shown in Fig.2. It is found that copper atoms exist in the interlayer adjacent to aluminum substrate and vice versa. The thermal diffusion on interface makes the interlayer with different ratios of atom number. Hence, the interface shows the three-sublayer structure after annealing (location 1, 2, 3). Based on the previous researches, several types of intermetallic compounds form on the interfacial interlayer, which have a great influence on mechanical properties of Cu/Al clad sheet[23].

2.2 Interfacial bond strength and peeled surface

The peeling tests were carried out to evaluate the interfacial bond strength. The results of peeling tests are shown in Fig.3. It is apparent that the peeling strength first increases and then decreases with the rise of annealing temperature. In addition, the peeling strength reaches the maximum value of 19.42 N/mm at 320 ℃. According to the microstructures shown in Fig.1a and 1b, the enhancement of peeling strength from 300 ℃ to 320 ℃ can be attributed to the formation of metallurgical bonding. On the basis of the previous researches, the intermetallic compounds generated on the interlayer damage the interfacial bonding during annealing at high temperature. These phases make the interface brittle and easy to fracture due to their low ductility. Hence, the peeling strength significantly decreases when the annealing temperature exceeds 340 ℃.

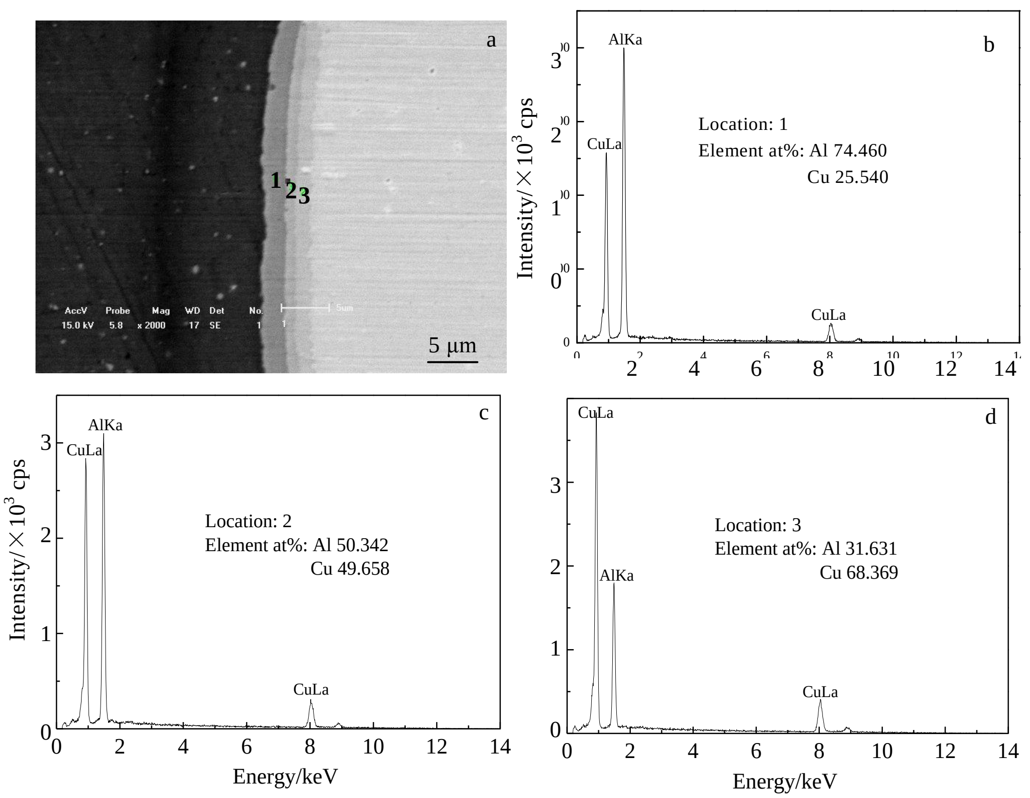

Fig.2 SEM interfacial microstructure (a) and EDS analysis results (b~d) of the interface annealed at 400 ℃ for 0.5 h

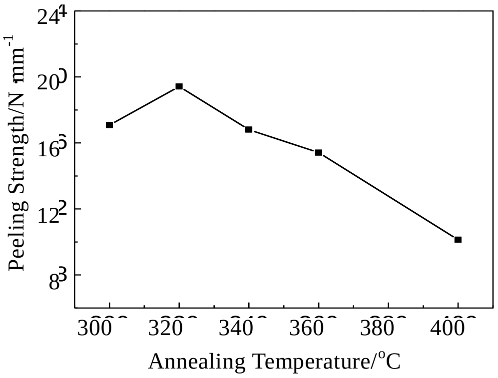

Fig.3 Variation of peeling strength of Cu/Al clad sheets with different annealing temperatures

The micromorphology of peeled surfaces annealed at different temperatures is shown in Fig.4. These SEM images can evaluate and verify the above results in peeling tests. It is known that the interfacial bonding mainly depends on fracture of metal surface and extrusion of underlying virgin metal during roll bonding. In peeling tests, the interface is peeled off under normal stress. The ductile fracture happens at the bonded area, and dimples form on the peeled surface. The interface with higher bond strength possesses more and larger dimples after being broken up as shown in Fig.4c and 4d. However, when the annealing temperature reaches 340 ℃, it can be found there are many cracks and few dimples on the peeled surface. Owing to the intermetallic compounds present in the interlayer, the fracture mechanism of peeled interface is mainly brittle fracture, which causes reduction in bond strength.

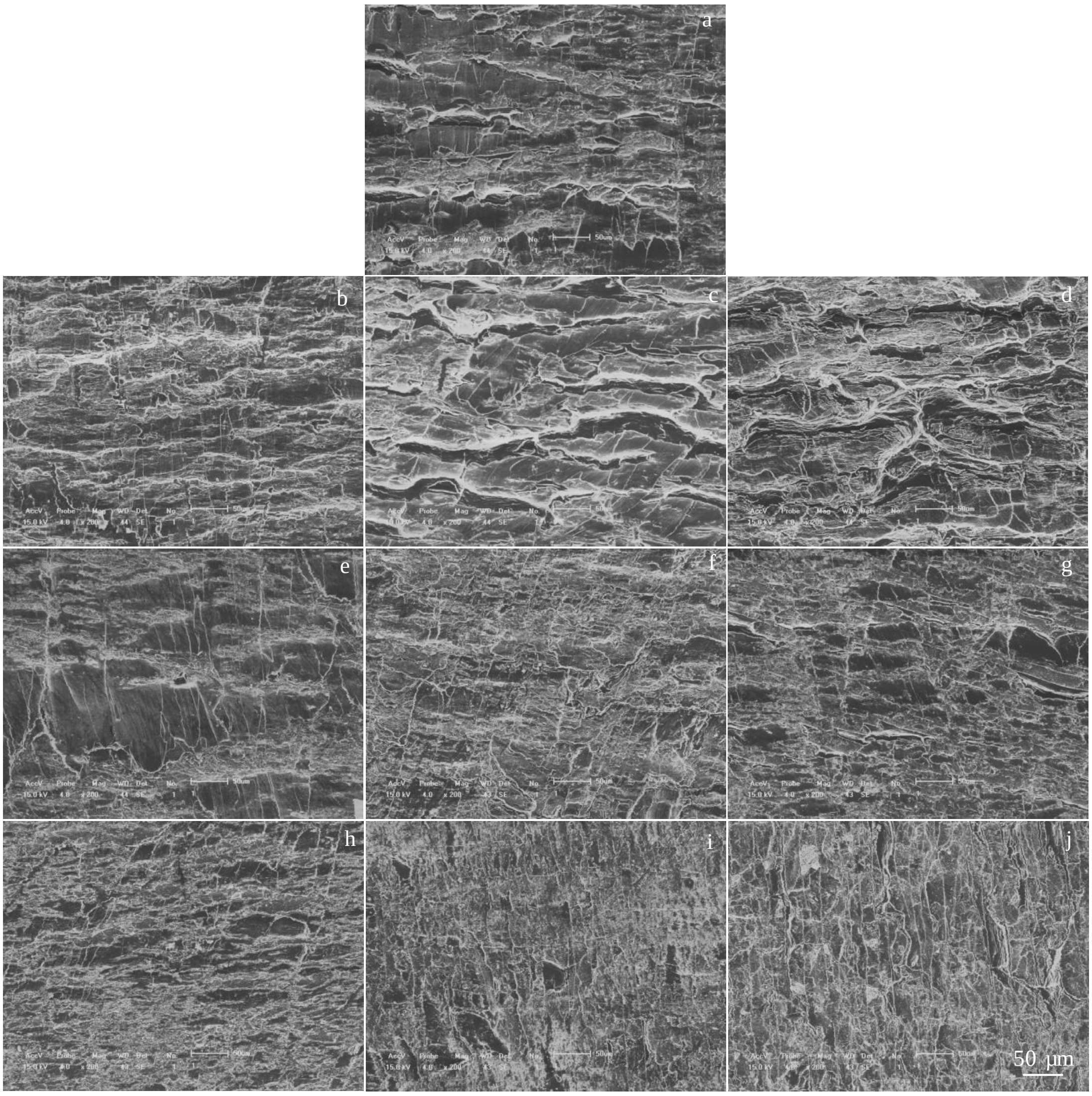

Fig.4 SEM microstructures of peeled surface of Cu/Al clad sheet annealed at different temperatures: (a) 300 ℃ Cu, (b) 300 ℃ Al, (c) 320 ℃ Cu, (d) 320 ℃ Al, (e) 340 ℃ Cu, (f) 340 ℃ Al, (g) 360 ℃ Cu, (h) 360 ℃ Al, (i) 400 ℃ Cu, and (j) 400 ℃ Al

2.3 Tensile properties

The tension tests of the clad sheets and individual components were conducted to investigate the effect of interface on tensile deformation. It can be found that two component metals fracture at the same time, though there is a significant difference in ductility between Cu and Al. In other words, when Cu and Al strips are bonded together and then annealed for 30 min, their elongations to failure are almost equal due to connection function of the interface. The tensile failure of the clad sheet doesn't lead to the complete separation of two metal layers. During the tension test, Cu/Al clad sheet is extended as a whole structure. Thus, there is an interaction between two component metals through the interface.

Tensile strength and elongation of the clad sheet and individual components at different annealing temperatures are shown in Fig.5. The engineering stress-strain curves are illustrated in Fig.6. It can be seen that the tensile strength of the composite and individual components is decreased and the elongation is enhanced with increasing the annealing temperature from 300 ℃ to 400 ℃. Based on the mixing rule, tensile properties of Cu/Al clad sheet are between those of copper and aluminum component under the same heat treatment condition. Normally, copper is higher and aluminum is lower than the clad sheet in tensile strength. Furthermore, aluminum substrate is bigger than copper substrate in elongation at low annealing temperature, and yet becomes smaller with the annealing temperature increasing. It should be noted that the clad sheet and its two components are extended by a similar distance at 340 ℃, though the elongation of copper component is slightly larger.

Since the roll bonding process provides a great pressure, copper and aluminum strips both possess severe working hardening. The difference in recrystallization temperature leads to the disparity of plastic recovery between two component metals. Aluminum substrate regains its plastic property first during annealing because of its lower melting point. When the annealing temperature reaches 360 ℃, the recrystallization occurs in copper substrate, which possesses better ductility than aluminum. The mismatch ductility between copper and aluminum substrate makes the interfacial interlayer under shear stress along the tensile axis. However, the interface can coordinate deformation behavior of either component metal. Due to the transition of interface, the ductility of the component metal with a smaller elongation is improved, and the other one is constrained. Thus, the two metal layers deform plastically as a whole structure and fracture simultaneously in tension tests. Hence, the interlayer plays a key role in keeping the clad sheet integrity.

2.4 Fracture analysis

Fig.7 represents the fracture microstructure of the clad sheet perpendicular to the tensile axis, and the one parallel to the tensile axis is shown in Fig.8. The fracture characteristic of the interlayer was analyzed through the crack degree between two metal layers. It is apparent that the clad sheet annealed at 320 ℃ or 340 ℃ has a smaller crack than others after tensile failure. The interfacial microstructure shown in Fig.9 was measured at a distance of 1 mm from the tensile fracture. It can be seen that a number of transverse microcracks distribute through the diffusion layer along the tensile direction, as illustrated in Fig. 9c, 9d and 9e. Moreover, the microcracks can also be observed within the interlayer annealed at 360 ℃ and 400 ℃.

The component metals and the interlayer were extended synchronously in tension tests. The interlayer is more brittle and gets damaged first under a shear stress caused by the mismatch elongation between copper and aluminum. However, two metal layers are still bonded together through the cracked interlayer before fracture. And then, the interface separation occurs with the local necking of metal substrates. Thus, strengthening the interfacial bonding can restrain the interface breakage, as shown in Fig.7b and Fig.8b. Otherwise, the crack degree of the interface is determined by not only the bond strength but also the elongation difference between two component metals. Owing to similar ductility, the extending of two metal layers is mainly synchronized, and the stress concentration within the interlayer is effectively re-

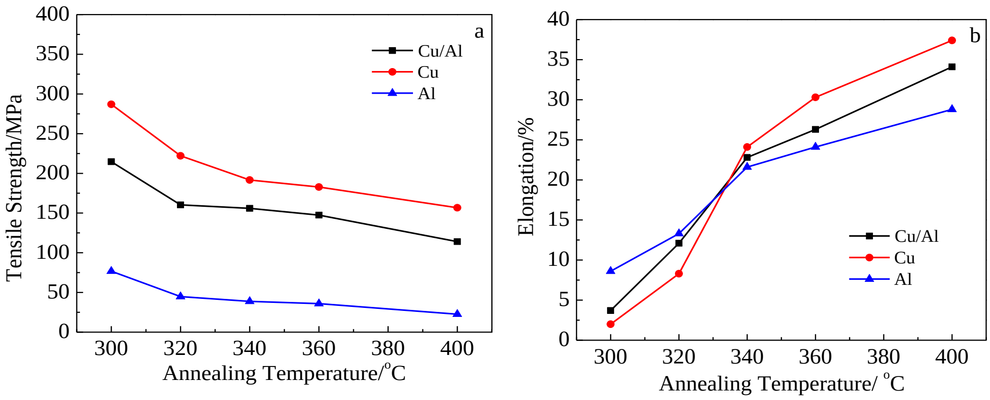

Fig.5 Variation of tensile properties of clad sheet at different annealing temperatures: (a) tensile strength and (b) elongation

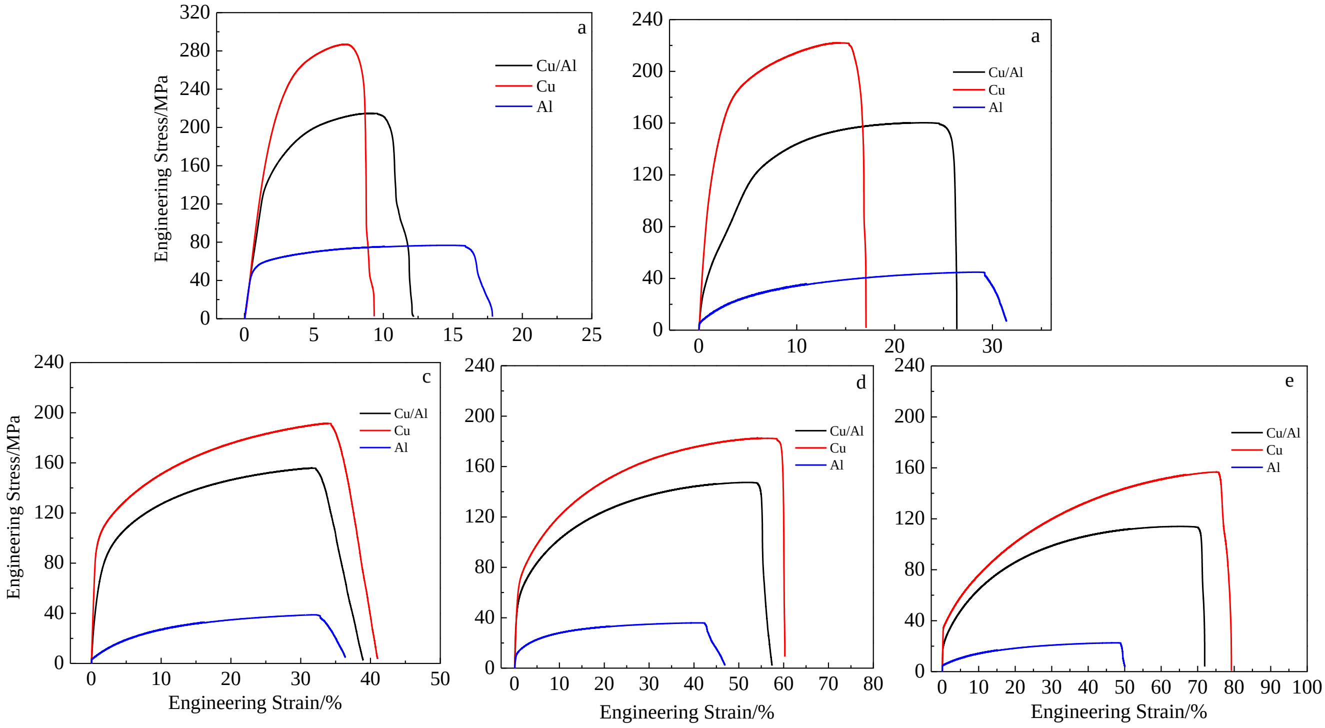

Fig.6 Engineering stress-strain curves of clad sheet and individual components at different annealing temperatures in tension tests: (a) 300 ℃, (b) 320 ℃, (c) 340 ℃, (d) 360 ℃, and (e) 400 ℃

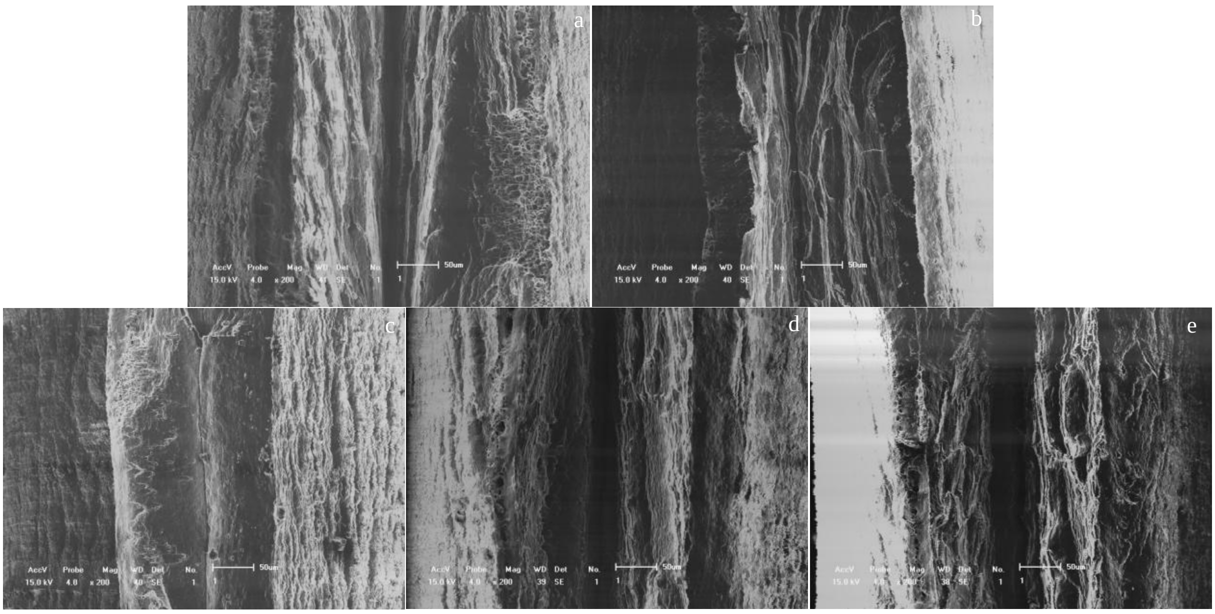

Fig.7 SEM fractographs of clad sheet at different annealing temperatures vertical to tensile axis: (a) 300 ℃, (b) 320 ℃, (c) 340 ℃, (d) 360 ℃, and (e) 400 ℃

lieved. Hence, the interface annealed at 340 ℃ gets more slightly damaged and its crack degree is less than another one, as shown in Fig.7c and Fig.8c.

According to the previous analysis, the interface breakage can be ascribed to plastic deformation of the composite and mismatch elongation between two metal layers during the tensile process. As shown in Fig.8, the interfacial microstructures near the tensile fracture are different in crack

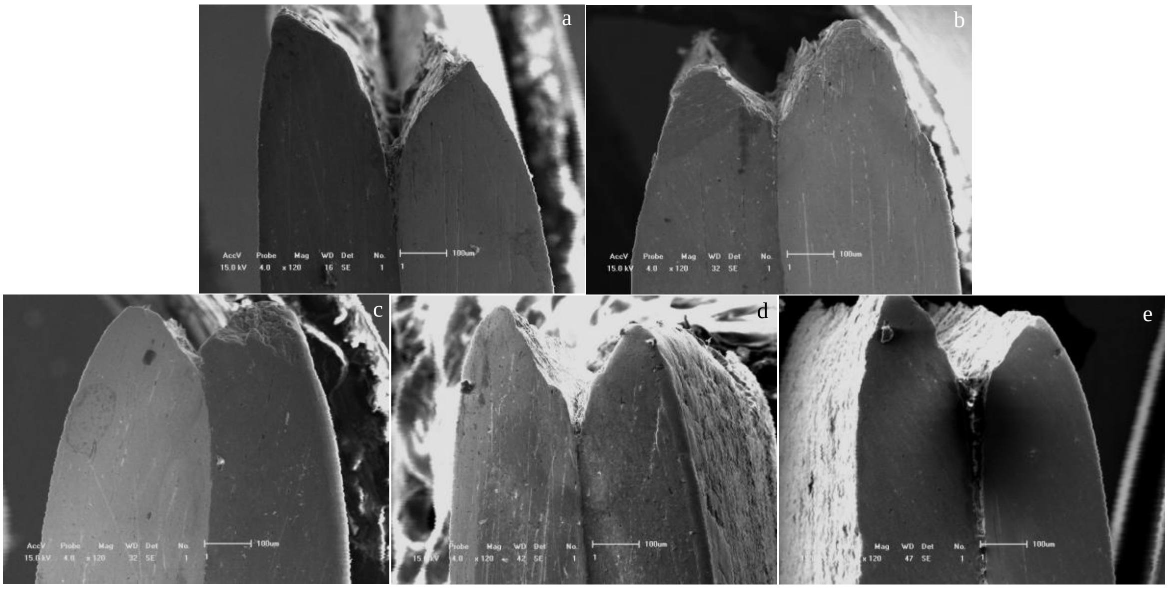

Fig.8 SEM fractographs of clad sheet at different annealing temperatures along tensile axis: (a) 300 ℃, (b) 320 ℃, (c) 340 ℃, (d) 360 ℃, and (e) 400 ℃

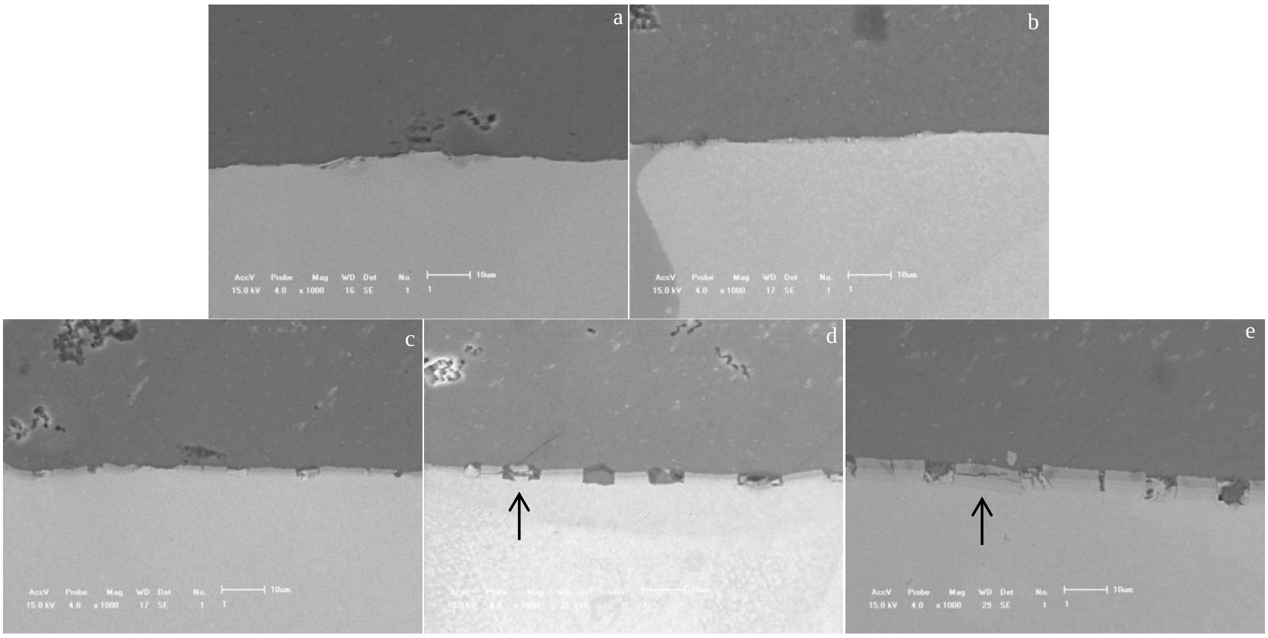

Fig.9 Interfacial microstructures of clad sheet near tensile fracture at different annealing temperatures: (a) 300 ℃, (b) 320 ℃, (c) 340 ℃, (d) 360 ℃, and (e) 400 ℃

morphology after annealing at different temperatures. The microcracks within the interlayer annealed at 320 ℃ or 340 ℃ are mainly transverse through the interface along the tensile direction. However, when the annealing temperature exceeds 360 ℃, several microcracks distribute longitudinally within the interlayer and cause layered fracture as indicated by arrows in Fig.9d and 9e. Since the annealing process at high temperature results in a decreasing bond strength, the interfacial transition is weakened. Then, the shear stress within the interlayer caused by mismatch elongation leads to layered fracture along the interface.

3 Conclusions

1) The interfacial interlayer of Cu/Al laminated composite fabricated by asymmetrical roll bonding and annealing is improved by thermal diffusion during annealing. The

diffusion layer annealed at 320 ℃ is discontinuous, and the interlayer with a uniform thickness forms when the annealing temperature reaches 340 ℃.

2) The peeling strength of the clad sheet annealed at 320 ℃ is 19.42 N/mm and higher than that of others. Moreover, annealing at high temperature damages the interfacial bonding.

3) The ultimate tensile strength and elongation of the clad sheet are intermediate between individual copper and aluminum layer. The elongation of the clad sheet annealed at 340 ℃ is almost equal to that of component metals.

4) Lower crack degree after tensile fracture can be attributed to good interfacial bonding and similar ductility between metal layers. The plastic deformation of metal substrates causes transverse cracks, and mismatch elongation of component metals causes interlamination fracture within the interlayer near tensile fracture.

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界面对铜/铝异步轧制层状复合材料组织与性能的影响

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摘　要：利用异步轧制复合技术和退火工业制备铜/铝层状复合材料，利用扫描电子显微镜观察界面微观组织和拉伸断口形貌，进行剥离和拉伸实验研究界面的力学性能。结果表明，热处理过程促进了界面层的形成，而较高的退火温度破坏了界面结合。层状复合材料的拉伸性能介于两组元金属之间。经 340 ℃退火后，铜基体的延伸性能与铝基体接近，并且界面开裂程度较低。在拉伸过程中，两金属基体延伸率不同，导致界面发生内部断裂。界面作为铜、铝之间的过渡层，在强化复合材料方面起到重要作用。

关键词：界面；拉伸；层状复合材料；异步轧制复合；退火

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