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Study on Microstructure and Properties of Ultra-Thin Cu/Al Composite Sheets Using the Cold-Rolled Composite Method at the Microscale

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Abstract: In this paper, an ultra-thin Cu/Al composite sheet with a thickness of 0.08 mm was obtained via the cold-rolling composite method using a four-high micro-rolling mill in the laboratory. The rolling reduction of a single pass was 65%. After the annealing of the ultra-thin Cu/Al composite sheets at temperatures ranging from 350 °C to 500 °C, the interface bonding mode of the Cu/Al composite sheets changed from mechanical bonding to metallurgical bonding, and the bonding strength was significantly improved. The microhardness value at the bonding interface of the ultra-thin Cu/Al composite sheets increases with the increase in annealing temperature. When the annealing temperature is 500 °C, the maximum microhardness value at the bonding interface reached 2.0 GPa. With the increase in annealing temperature, the tensile strength and elongation of the ultra-thin Cu/Al composite sheets decreases significantly. The peel strength of the extremely thin Cu/Al composite sheets increases at first and then decreases with the increase in annealing temperature, and reached the maximum value at an annealing temperature of 400 °C. When the annealing temperature was 400 °C, the tensile and peel properties of the ultra-thin Cu/Al composite sheet reached the best state.

Keywords: ultra-thin Cu/Al composite; annealing; microstructure; mechanical properties

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1. Introduction

An ultra-thin Cu/Al composite sheet integrates the excellent properties of copper and aluminum. It not only has the good conductivity, thermal conductivity and corrosion resistance of copper and aluminum, but also greatly reduces the material cost. It has achieved the goal of saving copper with aluminum and has been widely used in packaging [1], electronics, machinery, the automobile industry and other fields [2].

The preparation technology of Cu/Al composites can be realized using many kinds of processing methods. Seifollahzadeh et al. [3] prepared Cu/Al/Ag multilayered composites using the rolling composite method. Aghajani et al. [4] conducted cooling-assisted friction stir welding on different sheets including AA3003 aluminum and A441 AISI steel. Yang et al. [5] proposed the manufacturing process of a foam aluminum sandwich (AFS), which simplified the preparation process and reduced the cost. Vajdi et al. [6] prepared TiB2–SiC–Ti (TST) ceramic composites using spark plasma sintering. Keller et al. [7] prepared architectured copper–aluminum composites using cold drawing. The cold-rolled composite method has many advantages, such as a low equipment operation cost, stable production, simple operation, etc. Yu et al. [8] prepared copper/aluminum/copper laminates with different aluminum layer thicknesses and studied the influence of the thickness effect on the deformation characteristics of copper/aluminum/copper laminates. Gao et al. [9]

successfully prepared a Cu/Al/Cu sandwich composite sheet through a rolling composite experiment. The results show that with the increase in annealing temperature, the number of intermetallic compounds formed between the composite interface increases gradually. When the annealing temperature reaches 450 °C, the mechanical properties of Cu/Al/Cu composite sheets are great. Wang et al. [10] successfully prepared Cu/Al/Cu three-layer composites through a single pass of large reduction, and they found that certain heat treatment processes can promote the formation of intermetallic compounds. Zhang et al. [11] successfully prepared Al/Cu/Ti/Cu/Al laminated composites through the cold rolling process. It was found that the size of the inner layer and the Ti layer gradually decreased with the increase in rolling reduction. The determination method of the critical reduction rate of the rolling composite sheet is explored through continuous rolling parameters such as rolling speed and reduction rate. These composite sheets prepared using cold rolling composite method are mainly for large-sized composite sheets.

In recent years, with the development of many small and micro fields such as armored cables, COB packaging, air-cooled fins, etc., higher requirements have been put forward for the thickness, shape and bonding effect of Cu/Al composite sheets. Therefore, Cu/Al composite sheets have begun to enter the field of micro-forming. However, in the field of micro-forming, due to the reduction of the overall size of the workpiece, the mechanical properties and some laws of the part in the macro-size can no longer be used to study the micro-sized parts [12]. Although the knowledge system supporting the process, part tooling and metal-forming design at the macro level is very complete, the deformation behavior and process performance affected by the size effect in micro-forming are different from those in macro rolling. Macro rolling theory can no longer be applied to the small parts prepared via micro-forming, so it is necessary to strengthen the research on the influence of process conditions on the deformation behavior of materials during the micro-forming process. Today, some scholars are beginning to pay attention to this issue. Hu et al. [13] used 1060 aluminum alloy with a thickness of 464 μm which was rolled to a constant thickness ratio by adjusting the rolling speed and other parameters, and it was found that rolling parameters and heat treatment conditions significantly affect the microstructure and structure of each thickness zone. Naseri et al. [14] rolled Al/Cu/Al sandwich composite sheets through cold rolling equipment, and summarized the characteristics, explaining that the bonding strength of the composite sheet increased with the increase in the reduction rate by changing the different reduction rates. Chu et al. [15] studied the relationship between the interfacial bonding strength and diffusion layer thickness of a cold-rolled Cu/Al composite sheet during heat treatment and they found that the diffusion layer thickness became thicker with the increase in annealing time. There was a multi-layer structure on the diffusion layer.

These studies provide guidance for the effects of various experimental parameters (rolling speed, rolling mode, reduction rate, annealing temperature) on the preparation, diffusion analysis and bonding mechanics of ultra-thin Cu/Al composite sheets. However, there are few studies on the microstructure and mechanical properties of Cu/Al composite sheets in an ultra-thin state. Ultra-thin Cu/Al composite sheets rolled by cold rolling and the composite method are adjusted to obtain great mechanical properties, and the diffusion layer with appropriate thickness is obtained after annealing to improve the bonding state between them. Therefore, the effect of annealing temperature on the interface bonding, microstructure and mechanical properties of Cu and Al at the micro scale is still a research field.

In this paper, 0.06~0.09 mm ultra-thin Cu/Al composite sheets were prepared via the cold-rolling composite method using pure Cu and pure Al sheets through a self-designed four-high laboratory micro mill. The ultra-thin Cu/Al composite sheets were regarded as the research object and the effect of annealing temperature on the interface microstructure and mechanical properties of the ultra-thin Cu/Al composite sheets was investigated.

2. Materials and Experimental Procedures

2.1. Starting Materials

In this study, pure T2 Cu sheets with 99.90% purity (chemical composition is shown in Table 1) and 1060 pure Al sheets with 99.60% purity (chemical composition is shown in Table 2) were used as experimental materials.

Table 1. Chemical composition of pure Cu sheet (wt.%).

Cu	Bi	Sb	As	Fe	Ni	P	Pb	Sn	S	Zn
99.90	<0.0004	<0.0010	<0.0010	0.0074	0.0013	0.0017	0.0037	0.0023	0.0015	0.0099

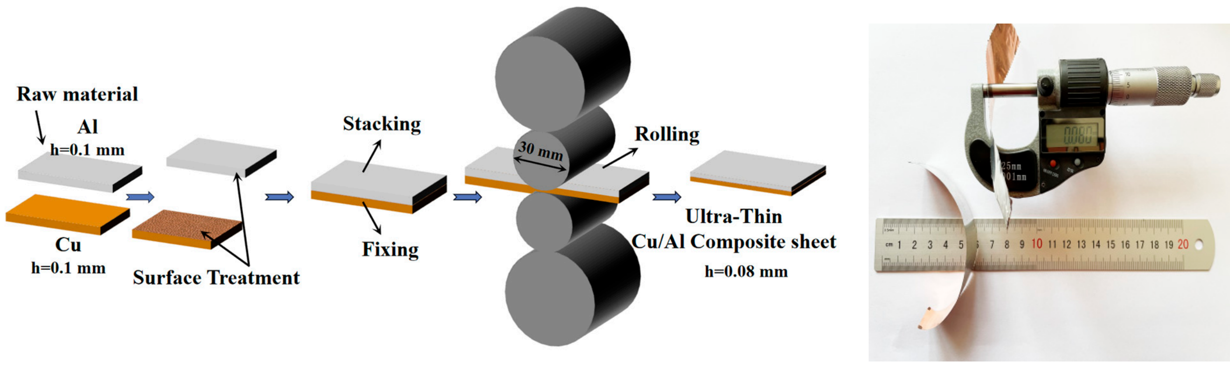
Table 2. Chemical composition of pure Al sheet (wt.%).

Al	V	Mn	Mg	Zn	Si	Ti	Fe	Cu
99.60	0.05	0.03	0.03	0.05	0.25	0.03	0.35	0.05

2.2. Experimental Method

First, T2 pure Cu layer and 1060 pure Al layer materials with an initial thickness of 0.1 mm and width of 20 mm were prepared, and then the two metal materials were cut into sheets with a length of 90 mm, width of 20 mm and thickness of 0.1 mm.

Figure 1a shows the rolling process of an ultra-thin Cu/Al composite sheet. The rolling condition was set as a 55%~70% reduction rate for single-pass rolling. The rolling speed was 7.07 mm/s. It should be noted that the surface of the roll should be cleaned with acetone before the rolling test to keep the surface of the sample prepared by rolling clean. The ultra-thin Cu/Al composite sheet of 0.06~0.09 mm was rolled using the self-designed four-high laboratory micro mill (parameters are shown in Table 3). Figure 1b shows the rolling product. The rectangular specimens of the rolled ultra-thin Cu/Al composite sheets were cut to a size of 100 mm × 20 mm and were 0.08 mm thick. The tensile specimen was cut along the rolling direction (rd) according to the inquiry m standard.



(**a**) (**b**)

Figure 1. (**a**) Rolling process of ultra-thin Cu/Al composite sheet. (**b**) Rolling products.

The annealing process was carried out in a vacuum tube furnace (GR.TF60, Shguier, Shanghai, China) under an argon atmosphere. The four annealing temperatures were set as 350 °C, 400 °C, 450 °C and 500 °C, respectively. The temperature gradient was set as 5 °C/min, the holding time after the temperature rise was 1 h, and finally the annealing experiment was completed by cooling to room temperature with the furnace.

After the annealing experiment, some ultra-thin Cu/Al composite sheets were continuously mechanically polished. The Al side was first corroded with 0.5% HF solution for about

50 s. Then, the Cu side was corroded with $NH_3 \bullet H_2O$:H_2O_2:H_2O = 5 mL:2 mL:5 mL solution for about 8 s, washed with water, and finally dried with alcohol.

Table 3. Parameters of four-high micro mill in the laboratory.

The Name of Equipment	Control Software	The Parameters of Equipment	Technical Indicators
Laboratory four-high micro-rolling mill	S7-300PLC	Rolling thickness range/mm	0.02~0.05 mm
		Maximum allowable rolling force/KN	$P \leq 500$ KN
		Rolling mill linear speed/(m/min)	$V \leq 2$ m/min
		Support roll diameter/mm	120 mm
		Working roll diameter/mm	30 mm
		Accuracy class/mm	0.005 mm

2.3. Experimental Detection

An inlay machine (XQ-2B, Hangzhou, China) was used to heat inlay the sample (150 °C). A super-depth microscope (VHX-5000, Keyence (China) Co., Ltd., Shanghai, China), scanning electron microscope (SEM) and energy dispersive spectrometer (EDS) (JSM-6480lv, Dongguan Xiemei Electronics Co., Ltd., Dongguan, China) were used to observe the interface microstructure, interface diffusion layer, tensile fracture and peel morphology of the sample. A universal tensile testing machine (WDW-300, Zhong Lu Chang, Jinan, China) was used to complete the uniaxial tensile test with a tensile speed of 1 mm/min and the peel test with a peel speed of 1 mm/min. Under the loading load of 2 mN, the hardness of the interface diffusion point and the base metal on both sides of the composite sheet was measured using nanoindentation.

3. Results and Discussion

3.1. Interface Microstructure and Diffusion Behavior of Ultra-Thin Cu/Al Composite Sheet

Figure 2 shows the grain morphology of the Cu side and Al side of the ultra-thin Cu/Al composite sheet at the four annealing temperatures of 350 °C, 400 °C, 450 °C and 500 °C. It can be seen from Figure 3 that with the annealing temperature increasing from 350 °C to 500 °C, the grain size on the copper side increases from 7 µm to 10 µm. The grain size on the aluminum side starts at 25 µm and increases to 43 µm. The reason for this phenomenon is that the position of grain boundary changes continuously with the increase in grain size, the two metal atoms continue to diffuse, and the diffusion speed becomes faster and faster. As can be seen from Figure 3, with the increase in annealing temperature, the average grain size of Cu and Al, respectively, increases from 7 µm to 22 µm and 26 µm to 43 µm. When the annealing temperature reaches 500 °C, a small amount of annealing twins also appear in the Cu side of the metal structure. As a metal with a typical face-centered cubic (FCC) crystal structure, Cu has a relatively low stacking fault energy, which leads to dislocation of Cu atoms in the process of recrystallization, thus forming annealing twins. Liang et al. [16] discovered that the Inconel 625 alloy exhibited equiaxed grains after welding thermal simulation.

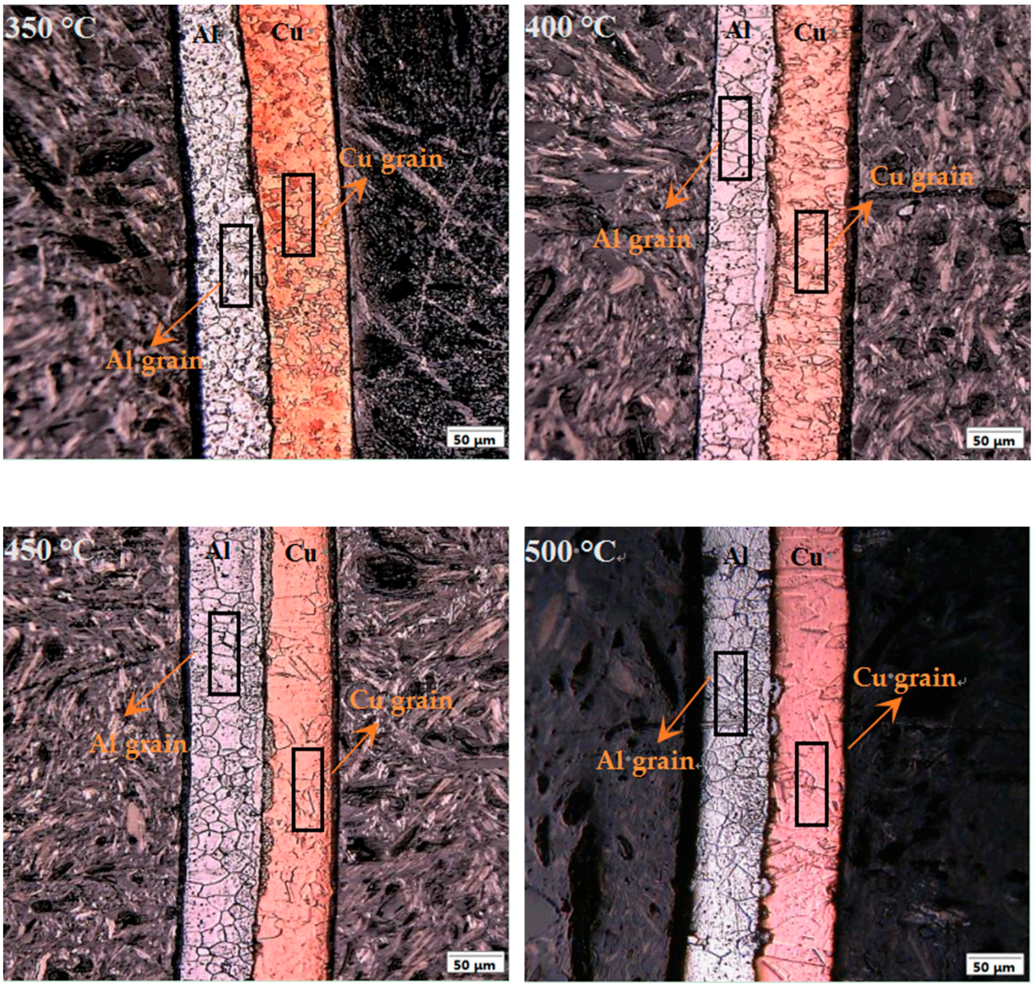


Figure 2. Microstructure on the ultra-thin Cu/Al composite sheet interface at different annealing temperatures.

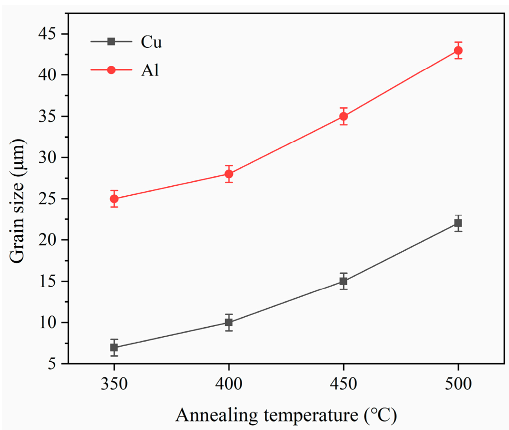


Figure 3. Effect of annealing temperature on grain size of ultra-thin Cu/Al composite sheets.

In the process of diffusion bonding of ultra-thin Cu/Al composite sheets, the thickness of the diffusion layer will seriously affect the mechanical properties of the whole composite sheet, such as tension and peeling. Li et al. [17] found that laminated composites with a great bonding interface can be prepared using the roll bonding technique. The results showed that diffusion occurred at the bonding interface, and the composite sheet with sub-micron thickness diffusion layer had high elongation [18].

Chu et al. [15] studied the relationship between the interfacial bonding strength and diffusion layer thickness of a cold-rolled Cu/Al composite sheet during heat treatment. In addition, they found that when the bonding property of a composite sheet was more than 12 μm and the thickness of the diffusion layer was greater than the critical value of 2.5 μm, the fracture mode of the interface would change from a ductile fracture to a brittle fracture. Manesh et al. [19] carried out a series of annealing experiments on a prepared ultra-thin Al alloy composite sheet, and found that when the generated interface diffusion layer is between 3 and 5 μm, the composite effect of the composite sheet will not be affected. In addition, when the thickness of diffusion layer is more than 10 μm, the bonding strength of the ultra-thin Al alloy composite sheet will be greatly reduced. Therefore, the importance of evaluating the thickness of the diffusion layer of ultra-thin Cu/Al composite sheet is emphasized. Figure 4 shows the morphology of the diffusion layer and scanning images of the element diffusion line at the interface of an ultra-thin Cu/Al composite sheet at annealing temperatures of 350 °C, 400 °C, 450 °C and 500 °C. The concentration profiles of aluminum and copper were obtained from EDS microanalysis along the lines shown on the SEM images as the Image line. The diffusion layer is formed between the composite interface of the ultra-thin Cu/Al composite sheet after annealing, because the matrix Cu and Al atoms on both sides of the cold-rolled ultra-thin Cu/Al composite sheet are heated to obtain energy and vibrate rapidly at the interface during the annealing process. In the continuous heat transfer process of annealing, Cu and Al atoms undergo atomic transition after being heated to obtain energy, and this gradually evolves into the material transfer process between the composite sheets, finally forming a diffusion layer. When the annealing temperature is 350 °C, due to the low annealing temperature at this time, no continuous and complete diffusion layer is formed, and the thickness of the diffusion layer is only 3.7 μm. The phenomenon of diffusion is not obvious. When the annealing temperature reaches 400 °C, a continuous and regular diffusion layer with a strip structure is formed at the composite interface. It can be seen that the diffusion layer displays the delamination phenomenon, and has increased from two layers to three layers. The thickness of the diffusion layer reaches 8.3 μm. However, there are some microholes in the diffusion layer. When the annealing temperature rises to 450 °C, the thickness of diffusion layer continues to increase, reaching 16.2 μm. Each layer at the interface of the diffusion layer is very clear, but there are microcracks in the formed diffusion layer. When the annealing temperature reaches 500 °C, the diffusion layer thickness is 23.3 μm. It is close to one third of the ultra-thin Cu/Al composite sheet, and there are a small number of micropore defects in the diffusion layer. During the annealing process of a Cu/Al composite sheet, with the increase in annealing temperature, the thickness of diffusion layer at the composite interface of the ultra-thin Cu/Al composite sheet gradually increases and the phenomenon of element diffusion is more and more obvious [20]. Tayyebi et al. [21] prepared Al/Cu/Mg multilayered composites via the accumulative roll bonding (ARB) process, and they found that the thickness of intermetallic compounds (Al3Mg2 and Al12Mg17) gradually increases with the increase in annealing temperature and holding time. The interface diffusion layer gradually thickens with the increase in annealing temperature, and the delamination phenomenon of the diffusion layer can be clearly seen. Excessive thickness of the diffusion layer can affect the mechanical properties of a composite sheet. Similar research results have also been observed in the study of Mao et al. [22].

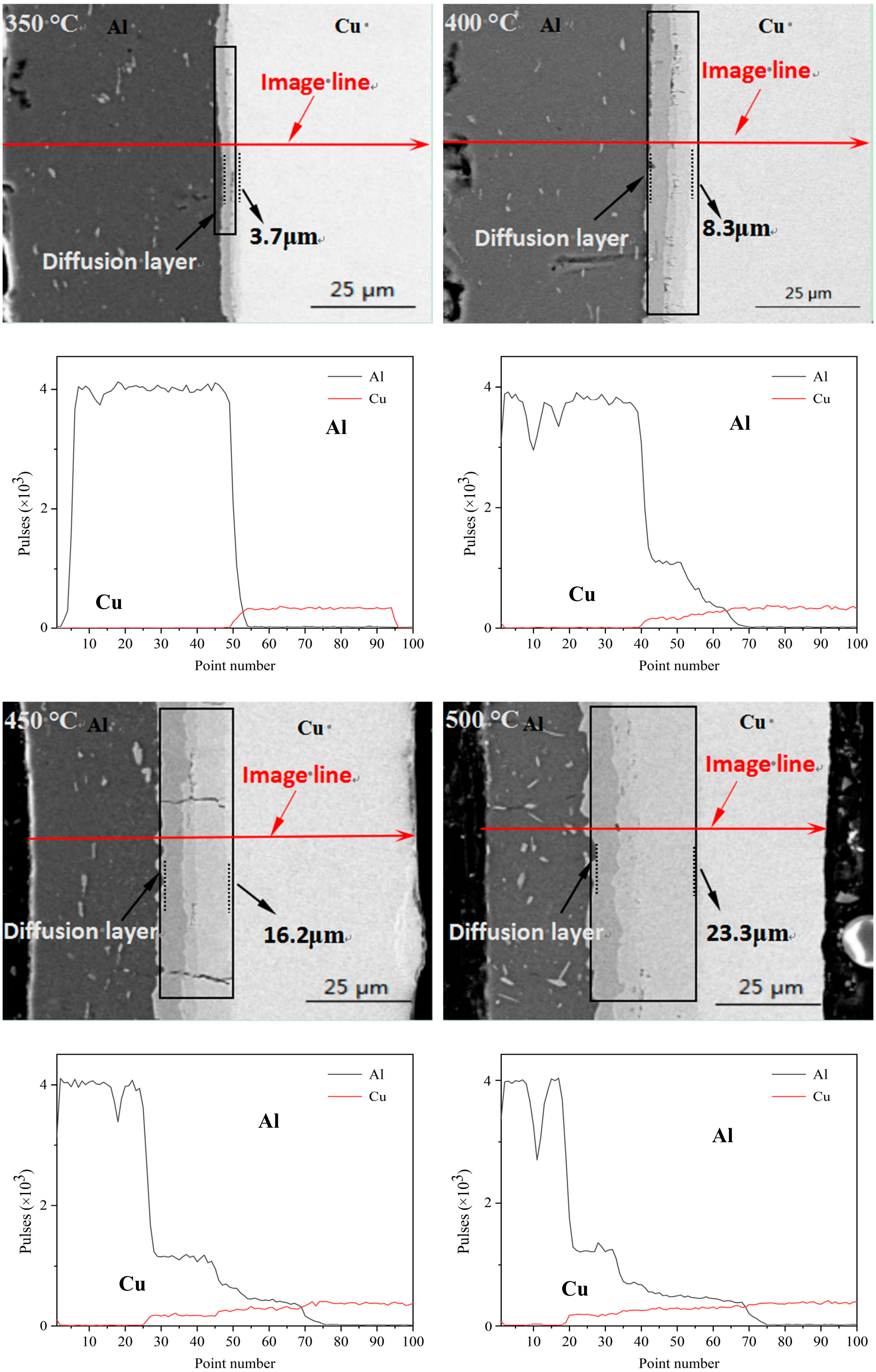


Figure 4. Scanning images of interface diffusion layer and element diffusion line of ultra-thin Cu/Al composite sheets at different annealing temperatures.

3.2. Microhardness Analysis of Ultra-Thin Cu/Al Composite Sheet

In an ultra-thin Cu/Al composite sheet, the work-hardening phenomenon will obviously appear during the rolling process. There are a lot of dislocations, vacancies and other defects in the two metals, which causes a lot of energy to be stored in the composite sheet. Research shows that cold rolling can improve the microhardness of bimetal composites. For example, as described by Rahmatabadi et al. [23], the strength and microhardness values of Al5052/MgAZ31B composite materials were obtained using the roll welding process. The recovery and recrystallization phenomenon occurred during annealing, energy was released in a large amount, and the hardness of the base metals Cu and Al changed, which can reflect the overall strength change trend of the ultra-thin Cu/Al composite sheet. Figure 5a shows the microhardness measurement location.

Figure 5b shows the trend of nanoindentation hardness change at the interface of an ultra-thin Cu/Al composite sheet under four different annealing temperatures. The hardness of the bonding interface of the annealed composite sheet is significantly higher than that of the metal matrix on both sides. When the annealing temperature is 350 °C, the hardness value at the interface of the composite sheet and the hardness value at the Cu side are close to 1.2 GPa, and the hardness values of the two closely resemble one another. This is because the annealing temperature is relatively low, the compound generated at the composite interface is relatively small, and the composition is mainly a Cu solid solution, so the impact on the hardness value of the bonding interface is relatively small. The microhardness value at the composite interface of the ultra-thin Cu/Al composite sheet increases gradually with the increase in annealing temperature, which is because during the annealing process, the Cu and Al atoms at the interface of the composite sheet diffuse to form hard and brittle intermetallic compounds. In addition, the number increases continuously with the increase in annealing temperature, resulting in a significantly higher hardness value at the interface. From the SEM image in Figure 5a, it can be seen that the diffusion layer at the bonding interface of the ultra-thin Cu/Al composite sheet displays a delamination phenomenon, while in Figure 5b, at −30 µm, the hardness value of the sub diffusion layer at m reaches a peak value of 2.0 GPa at 500 °C, indicating that the hardness value of this intermetallic compound layer is the largest and has a significant impact on the bonding strength of the composite sheet.

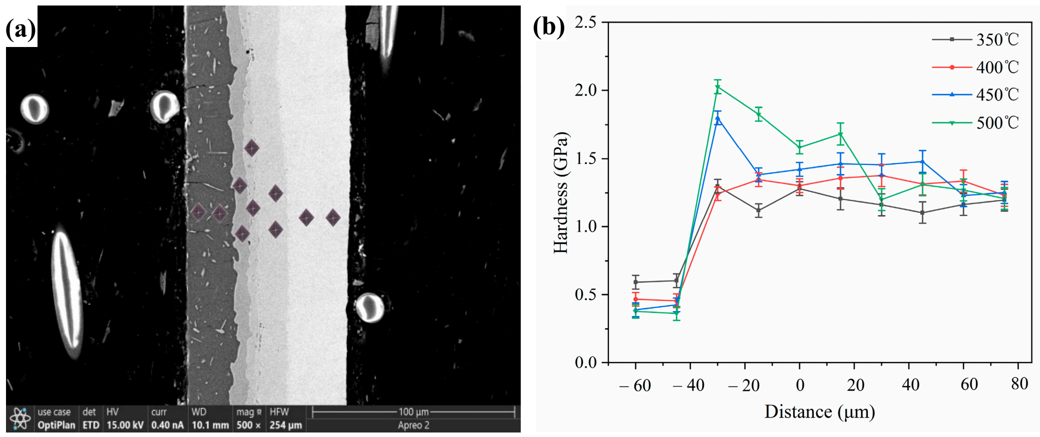


Figure 5. Variation curve of microhardness on both sides of the interface of ultra-thin Cu/Al composite sheets at different annealing temperatures. (**a**) Dotting position; (**b**) Hardness value.

3.3. Tensile Behavior of Ultra-Thin Cu/Al Composite Sheet

The tensile mechanical properties of materials, including tensile strength, elongation and yield strength, are an important factor reflecting the properties of materials. The displacement–load curve obtained from the uniaxial tensile test of ultra-thin Cu/Al composite sheet at room temperature is converted into a true stress–true strain curve, which reflects the tensile mechanical properties of the material well.

Figure 6 shows the true stress–strain curves of pure Cu and pure Al sheets and an ultra-thin Cu/Al composite sheet at an annealing temperature of 400 °C. The tensile strength and elongation of pure Al sheets are the lowest because when the annealing temperature is 400 °C, the metal Al reaches its recrystallization temperature and has completed a large degree of the recrystallization process. In addition, the grain size increases after annealing, resulting in the reduction of the tensile strength of the metal Al, which conforms to the Hall–Petch formula. The tensile strength and elongation of pure Cu sheets are higher than that of pure Al sheets. This is because the recrystallization temperature of metal Cu is relatively high at this time, but the recovery phenomenon occurs. There are a large number of strip-like grains inside the metal Cu. However, its tensile strength is smaller than that of the ultra-thin Cu/Al composite sheet. The ultra-thin Cu/Al composite sheet has the highest tensile strength and the elongation is between the Cu and Al, showing excellent tensile mechanical properties, indicating that the Cu/Al composite sheet has performance advantages over Cu and Al. Under the same annealing process, the yield strength of the ultra-thin Cu/Al composite sheet is higher than that of the two metal substrates, which directly explains the advantage of forming an intermetallic compound layer at the composite interface.

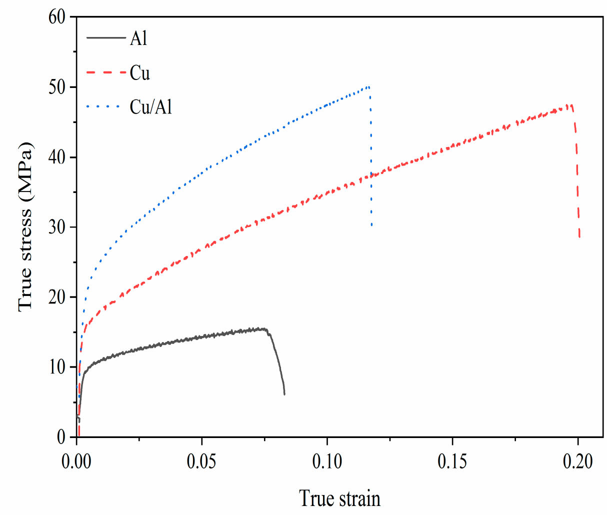


Figure 6. True stress–true strain curves of pure Cu, pure Al and ultra-thin Cu/Al composite sheets at the same annealing temperatures.

Figure 7 shows the true stress–true strain curves of ultra-thin Cu/Al composite sheet at different annealing temperatures. With the increase in annealing temperature, the tensile strength and elongation of the ultra-thin Cu/Al composite sheets decreased significantly. This is because when a material with a large number of grains undergoes plastic deformation, there is also a large number of grain boundaries that hinder the strengthening effect of the dislocation movement, and the strength of the material is relatively high. However, when the total volume of the metal sheet remains unchanged and the internal grains increase, the area of the grain boundaries will decrease, and the strength of the ultra-thin Cu/Al composite sheet will significantly decrease. Therefore, in the process of annealing, the grain size of the metal matrix on both sides of the ultra-thin Cu/Al composite sheet changes, which is a process that is greatly affected by temperature, and will have a significant impact on the tensile properties of the composite sheet. When the annealing temperature is 400 °C, the tensile strength and elongation of the ultra-thin Cu/Al composite sheet reach the maximum, which are 55 MPa and 14.3%, respectively. When the annealing temperature is 450 °C, the tensile strength of the ultra-thin Cu/Al composite sheet decreases sharply. The reason for this phenomenon is that when the annealing temperature reaches 450 °C, the metal Cu is completely recrystallized, and the tensile strength of the composite sheet drops sharply. The inflection point usually occurs in the tensile curve of a metal composite sheet. When the annealing temperature reaches 500 °C, the inflection point in the tensile curve is not obvious. This is because the Cu/Al composite sheet for tensile test is very thin, so the duration of the inflection point in the curve is very short and cannot be clearly monitored. After the fracture of the Al side of the matrix, the Cu side also breaks rapidly. The main reason is that when the annealing temperature is 500 °C, a large number of hard and brittle phase compounds are formed between the metal interfaces of the composite sheet, so the element diffusion layer between the metals first breaks during the tensile process. At this time, the metals Cu and Al still have a certain plasticity. The tensile load of the tensile machine is mainly concentrated on the Cu and Al sheets, and the yield strength of the metal Al is much smaller than that of the metal Cu. After the Al side of the matrix breaks, the Cu side also breaks rapidly.

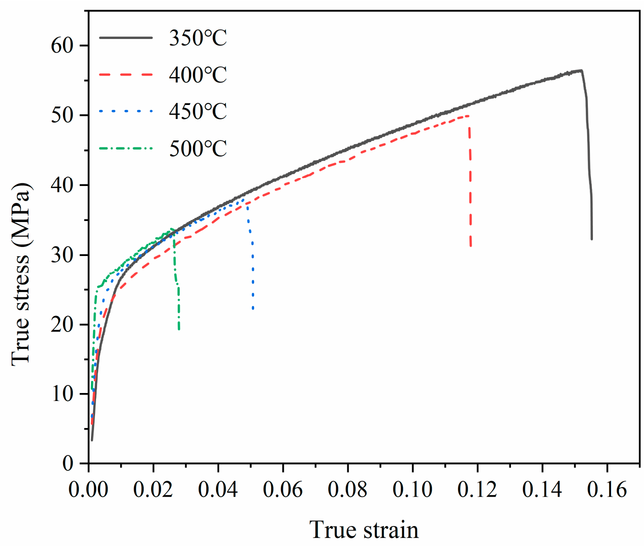


Figure 7. True stress–true strain curves of an ultra-thin Cu/Al composite sheet at different annealing temperatures.

Figure 8 shows the tensile fracture morphology of ultra-thin Cu/Al composite sheets obtained via tensile testing at room temperature after annealing at 350 °C, 400 °C, 450 °C and 500 °C. From the enlarged view on the right, it can be seen that interface pins appear at the fractures on both sides of copper and aluminum, indicating that the bonding strength of the interface is very high. When the annealing temperature increases from 350 °C to 500 °C, the delamination degree of the composite interface decreases from large to small, and a small amount of micropores appear at the fracture interface of the copper and aluminum, indicating that the fracture form at this time is a ductile fracture. The increase in annealing temperature leads to an increase in grain size and grain boundary area. The formation of large areas of grain boundaries will hinder dislocation movement, and significant stress concentration will occur at grain boundaries, leading to the formation of micropores. When the annealing temperature is 500 °C, it can be found that the tensile fracture of the interfacial diffusion layer becomes very rough, which actually achieves partial consistency between the deformation of the composite interfacial diffusion layer and the base metal.

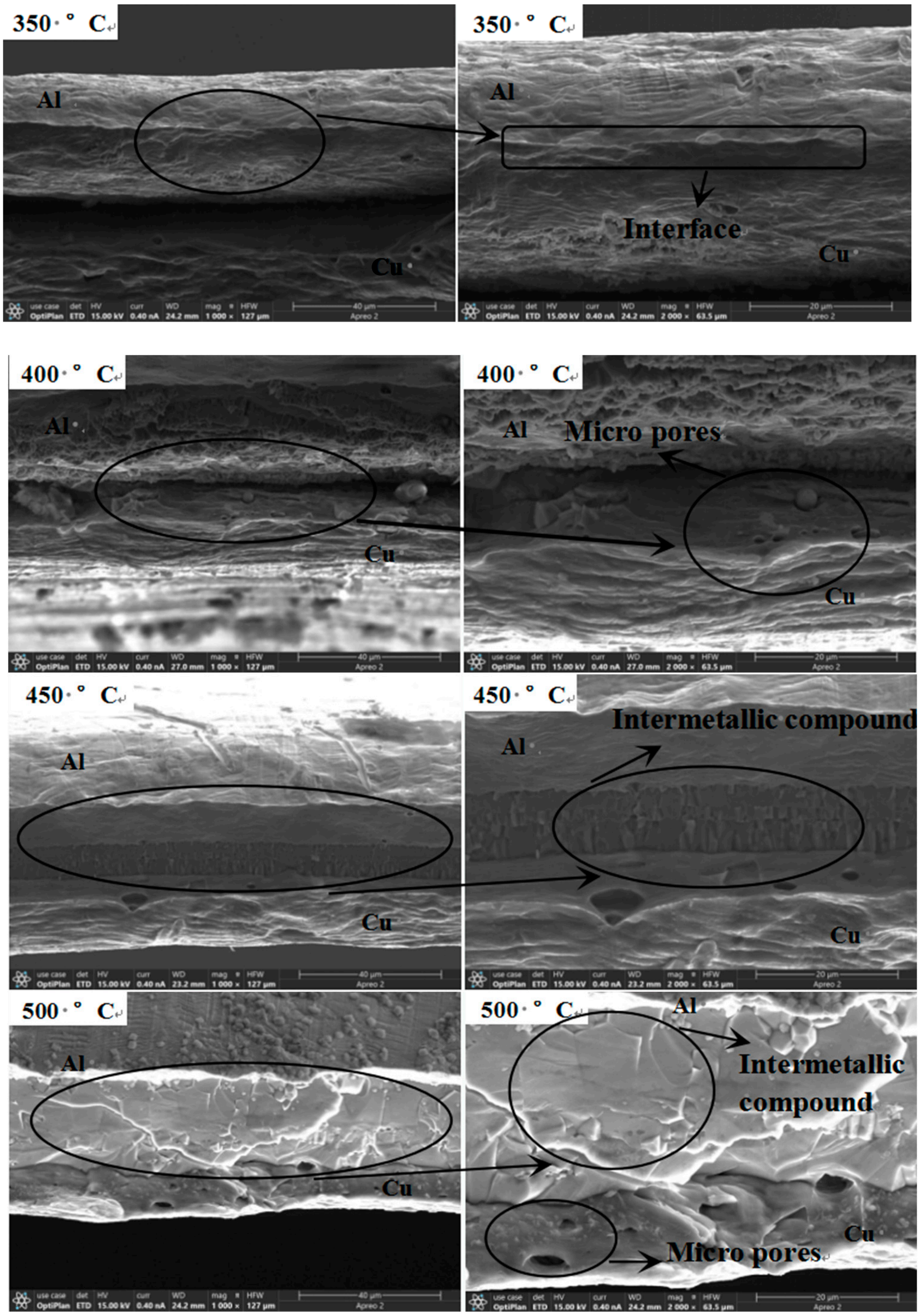
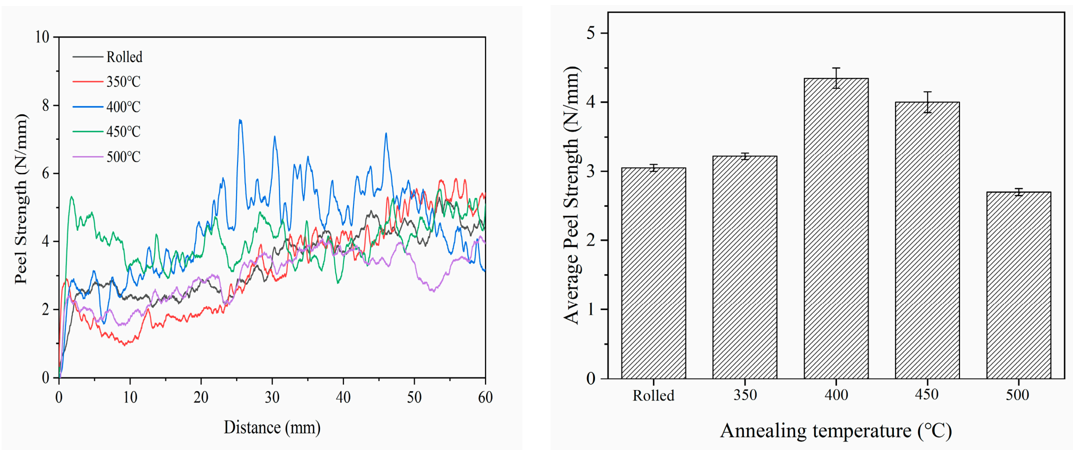


Figure 8. Tensile fracture morphology of an ultra-thin Cu/Al composite sheet at different annealing temperatures.

3.4. Stripping of Ultra-Thin Cu/Al Composite Sheet

Gao et al. [9] successfully prepared a Cu/Al/Cu three-layer composite strip and carried out a peeling test on the rolled piece after annealing. The results showed that the bonding interface diffusion layer increased with the increase in annealing temperature, and the average peeling strength of the Cu/Al/Cu composite strip increased first and then decreased. Figure 9a shows the peel strength of the ultra-thin Cu/Al composite sheet at different annealing temperatures. It can be seen from the trend of the peeling curve under four annealing temperatures that the peeling strength of the ultra-thin Cu/Al composite sheet is not a stable value but fluctuates back and forth between certain values, indicating that the bonding state at the interface of the composite sheet is irregular and discontinuous. It can actually be attributed to two causes: first, due to the large plastic deformation caused by the huge extrusion pressure on the metal Cu and Al of the composite sheet during the rolling process, the oxide film on the composite surface will be broken, and fresh metal will be exposed to each other to different degrees, so that the composite sheet will not combine evenly. Secondly, the compounds generated during annealing are not regular and continuous, and the formation of different amounts of compounds at each section of the interface results in an uneven combination of composite sheets. With the increase in annealing temperature, the peel strength of the ultra-thin Cu/Al composite sheet increases at first and then decreases. When the annealing temperature is between 350 and 400 °C, the peel strength of the ultra-thin Cu/Al composite sheet gradually increases. This is because when the annealing temperature is 350 °C, the compounds generated between the bonding interfaces of the ultra-thin Cu/Al composite sheet are very few and discontinuous. The bonding of the composite sheet is mainly mechanical bonding between metals, so the peel strength is not high.

Figure 9b shows the average peel strength of the ultra-thin Cu/Al composite sheet at different annealing temperatures. The average peel strength of the composite sheets increases first and then decreases with the increase in annealing temperature. When the annealing temperature reaches 400 °C, the diffusion of Cu and Al elements between the composite interface is sufficient, and the peel strength is significantly increased, reaching the maximum peel strength of the ultra-thin Cu/Al composite sheet, and the bonding performance is also in the best state. However, when the annealing temperature continues to rise to more than 450 °C, the peel strength decreases sharply. This is because the solid solution formed by the diffusion of Cu and Al atoms between the composite interface is supersaturated, and the type and quantity of hard and brittle-phase compounds increase sharply. The peel strength is very sensitive to the microstructure of the composite interface; a large number of hard and brittle-phase compounds that are not conducive to the bonding of the composite sheet seriously weaken the bonding performance of the composite sheet. When the annealing temperature reaches 500 °C, the peel strength between the interfaces cannot reach even one third of the peel strength of the composite sheet under the annealing temperature of 400 °C. During the peeling process, the hard and brittle-phase compounds at the interface of the composite sheet easily fracture under the tensile force, and the Cu/Al composite sheet gradually detaches, resulting in a sharp drop in the average peel strength of the composite sheet.



(**a**) (**b**)

Figure 9. Variation curve of ultra-thin Cu/Al composite sheets' peel force at different annealing temperatures. (**a**) Peel strength value; (**b**) Average peel strength value.

3.5. Composition of Diffusion Layer of Ultra-Thin Cu/Al Composite Sheet

Figure 10 shows the X-ray diffraction patterns at the interface diffusion layer of the ultra-thin Cu/Al composite sheets at the four annealing temperatures of 350 °C, 400 °C, 450 °C and 500 °C. From the figure, it can be seen that three intermetallic compounds, Al_2Cu, AlCu and Al_4Cu_9, were generated from the aluminum side to the copper side at annealing temperatures ranging from 350 °C to 500 °C. With the increase in annealing temperature, more and more intermetallic compounds were generated at the bonding interface. The generation of a small amount of intermetallic compounds improves the peel strength of ultra-thin Cu/Al composite sheets. Therefore, when the annealing temperature increases from 350 to 400 °C, the peel strength increases. The generation of a large number of hard, brittle-phase compounds greatly reduces the peel strength and tensile strength of ultra-thin Cu/Al composite sheets, which is an important reason for the significant decrease in peel strength and tensile strength of the composite sheets at 500 °C.

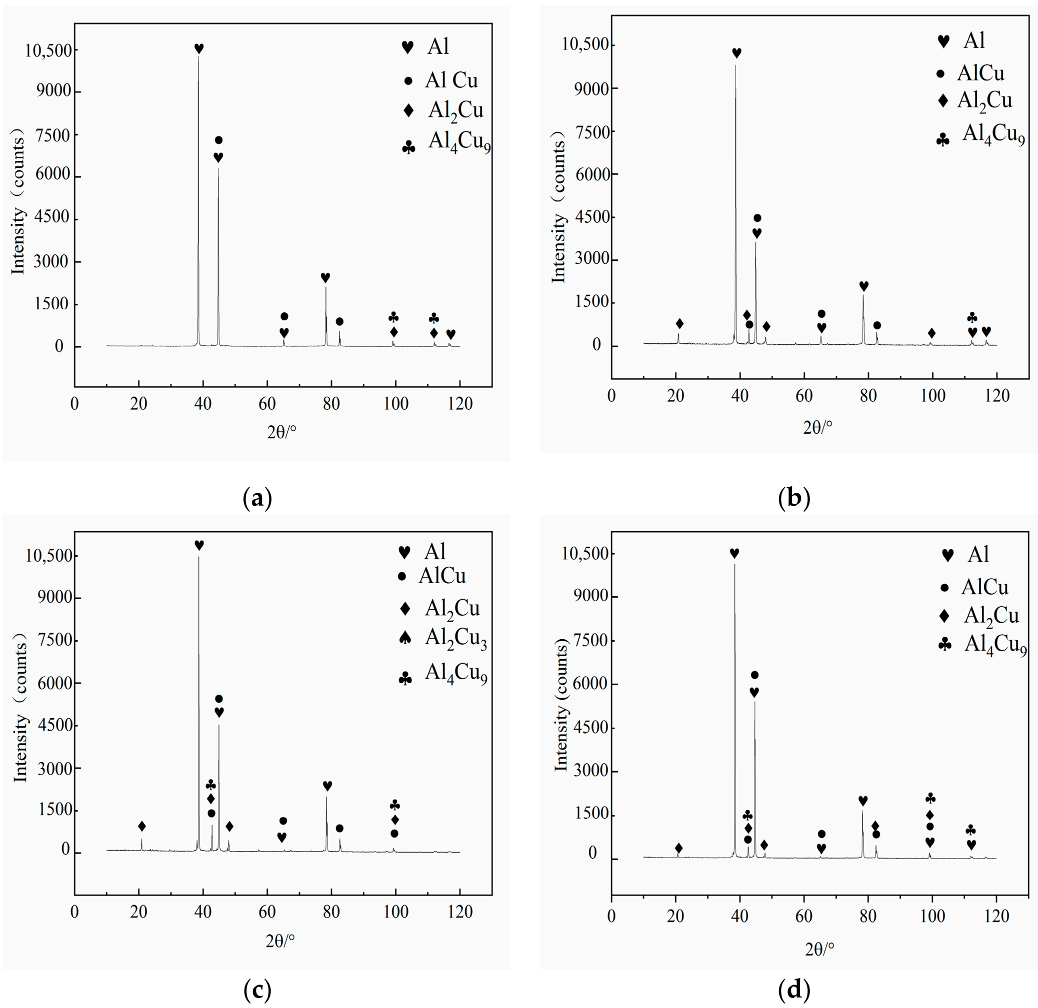


Figure 10. The X-ray diffraction patterns at the interface diffusion layer of ultra-thin Cu/Al composite sheets at four annealing temperatures: (**a**) 350 °C; (**b**) 400 °C; (**c**) 450 °C; (**d**) 500 °C.

4. Conclusions

In this paper, ultra-thin Cu/Al composite sheets were successfully prepared using the cold rolling composite method, and the effect of an annealing temperature of 350~500 °C on the microstructure and mechanical properties of the ultra-thin Cu/Al composite sheet was studied. The research conclusions are as follows:

1. The four-high micro-rolling mill designed in the laboratory can produce 0.06~0.09 mm ultra-thin and well-shaped Cu/Al composite sheets using single-pass rolling with a large rolling reduction rate of 55%~70%.
2. The thickness of the interface diffusion layer of the ultra-thin Cu/Al composite sheet after annealing treatment is higher; it is 3.7 μm at 350 °C and increases to 23.3 μm at 500 °C μm. The microhardness value at the bonding interface of the composite sheet gradually increases with the increase in annealing temperature. The formation of intermetallic compounds at the interface greatly improves the hardness value at the bonding interface of ultra-thin Cu/Al composite sheets.
3. As the annealing temperature increases, the tensile strength and elongation of the ultra-thin Cu/Al composite sheets decrease due to the combined effects of increasing the grain size of copper and aluminum and the thickening of the diffusion layer. The peel strength of the composite sheet first increases and then decreases as the annealing temperature increases. When the annealing temperature reaches 400 °C, the peel strength reaches a peak, indicating that intermetallic compounds Al_2Cu, AlCu and Al_4Cu_9 seriously weaken the bonding strength of the composite sheet. When the annealing temperature is 400 °C, the ultra-thin Cu/Al composite sheet reaches the optimal bonding state.

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