

Article

Effect of Liquid-Solid Volume Ratio and Surface Treatment on Microstructure and Properties of Cu/Al Bimetallic Composite

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Abstract: Due to exceptional conductivity, lightweight nature, corrosion resistance, and various other advantages, Cu/Al bimetallic composites are extensively utilized in the fields of communication, new energy, electronics, and other industries. To solve the problem of poor metallurgical bonding of Cu/Al bimetallic composites caused by high-temperature oxidation of Cu, different coating thicknesses of Ni layer on Cu rods were used to fabricate the Cu/Al bimetallic composite by gravity casting. The effect of liquid–solid volume ratio and coating thickness on microstructure and properties of a Cu/Al bimetallic composite were investigated in this study. The results indicated that the transition zone width increased from 242.3 μm to 286.3 μm and shear strength increased from 17.8 MPa to 30.3 MPa with a liquid–solid volume ratio varying from 8.86 to 50. The thickness of the transition zone and shear strength increased with the coating thickness of the Ni layer varying from 1.5 μm to 3.8 μm , due to the Ni layer effectively preventing oxidation on the surface of the Cu rod and promoting the metallurgical bonding of the Cu/Al interface. The presence of a residual Ni layer in the casted material hinders the diffusion process of the Cu and Al atom. Therefore, the thickness of the transition zone and shear strength exhibited a decreasing trend as the coating thickness of the Ni layer increased from 3.8 μm to 5.9 μm . Shear fracture observation revealed that the initiation and propagation of shear cracks occurred within the transition zone of the Cu/Al bimetallic composite.

Keywords: Cu/Al bimetallic composite; liquid–solid volume ratio; coating thickness



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1. Introduction

Due to a combination of the high electrical and thermal conductivity of Cu [1] with the light weight of Al [2–6], Cu/Al bimetallic composites [7–14] were widely used in the electric power transportation industry. Cu/Al composite materials can replace copper materials in generators and aluminum materials in external power grids, as well as the contact surfaces between the two, thereby reducing the use of copper resources and reducing the accident rate in power generation and supply. Although the compound casting method has exhibited great superiority in fabricating irregular shapes of Cu/Al bimetallic composites, it still has some drawbacks in the aspects of the rapid growth of intermetallic compounds and the oxidation of the solid Cu substrate. The hard and brittle transition zone was formed at the interface of the Cu/Al bimetallic composite, which reduced the mechanical properties of the material. Therefore, optimization of the transition zone has become a hot topic in the research and development of Cu/Al bimetallic composite.

It was widely reported that the transition zone played an important role in the microstructure and mechanical properties of Cu/Al bimetallic composites [15]. There have been extensive studies on the formation mechanism of the transition zone. For example, Wang et al. [16] used synchrotron X-ray technology to study the interfacial diffusion behavior and microstructure evolution of Cu/Al bimetallics. Cu and Al first diffuse to each other and then form α -Al dendrites and intermetallic compounds (IMCs) between the matrix.

The liquid–solid ratio was one of the key factors for the formation of the transition zone in the preparation of bimetallic composite by gravity casting. For example, it has been found that the bonding quality of AZ91 and AZ31 alloys was better when the liquid–solid ratio was larger [17]. Other studies have shown the preparation of high chromium cast iron and medium carbon steel bimetals by gravity casting. With the increase of liquid–solid product ratio, the diffusion activity of elements increased, leading to the increase of the interfacial transition zone width and shear strength [18]. It has been reported that the transition zone consisted of the intermetallic compounds and the remelting zone, and the cooling rate influenced the thickness of intermetallic compounds, the microstructure of the remelting zone, and the morphology of the remelting zone/Al interface [19]. Tavasoli et al. [20] reported the effect of pouring temperature on the transition zone, and the results showed that an increase in Al melting temperature resulted in a gradual increase in the thickness of intermetallic compounds and interfaces. Chen et al. [21] found that pouring temperature, cooling mode of Cu plate surface, and starting time of forced cooling after pouring had no effect on the microstructure species of the transition zone.

In addition, during the preparation of the Cu/Al bimetallic composite by gravity casting, an oxidation reaction occurred on the surface of Cu during preheating [22], and the formation of an oxide film reduced the metallurgical bonding property of the interface between Cu and Al. Therefore, it was particularly important to cover the surface of the copper with a protective film to prevent oxidation [23]. A suitable protective film can not only prevent the surface oxidation of Cu, but also promote the metallurgical bonding between Cu and Al. Boucherit et al. [24] achieved friction stir welding of Cu/Al using a zinc interlayer and found that Zn can significantly reduce the formation of intermetallic compounds such as Al_2Cu and Al_4Cu_9 , thereby improving the shear lap tensile strength of the joint. Ye et al. [25] adopted a new Zn–Al–Si filler metal to braze Cu/Al and found that Si could inhibit the growth of intermetallic compounds, thus significantly improving the corrosion resistance of Cu/Al bimetallic materials. Breedis et al. [26] found that adding a certain amount of Ni to copper alloys can inhibit the growth rate of Cu/Al intermetallic compounds, effectively reduce the content of intermetallic compounds, improve the microstructure of copper alloys, and effectively improve its properties. It was found that by depositing Ni–P coating on a copper substrate by electroless plating, the intermediate coating acted as a protective film, which could reduce the rate of intermetallic compound generation [27]. If Ni was used as the intermediate layer during the pouring process of Al and Cu, it can be seen from the Cu–Ni binary phase diagram that due to the infinite solubility of Cu and Ni, intermetallic compounds will not be generated and copper matrix oxidation can be prevented. Liu et al. [28] reported that a uniform Ni protective layer on the electroplating of the copper matrix can prevent the surface oxidation of the copper matrix, and the Ni layer dissolved during the interfacial reaction during the pouring process, promoting the metallurgical combination of copper and aluminum.

At present, the research on interface processing of Cu/Al composite materials mainly focuses on coating materials and coating methods, while there are few studies on the effect of coating thickness on the microstructure and properties of Cu/Al composite materials. In this study, the fabrication of Cu/Al bimetallic composite was achieved by gravity casting, and the effect of liquid–solid ratio and coating thickness on the microstructure and properties of Cu/Al bimetallic composites was discussed. The appropriate thickness of the transition zone will significantly improve the mechanical properties of the Cu/Al bimetallic composites.

2. Materials and Methods

The Cu/Al bimetallic composite was fabricated by pure copper rods and aluminum rods. In order to prevent oxidation of the copper while being kept at elevated temperature, a layer of nickel was plated on the surface of the copper rods before casting. The electroplating solution consisted of 840 g Ni_2SO_4 , 100 g $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$, and 3 L deionized water. The nickel-plating voltage was 4V, and the nickel-plating time was 10, 25, and 40 min, respectively.

The electroplating device was shown in Figure 1. Both the casting mold and nickel-plated copper rod were kept at 500 °C with the resistance furnace at the beginning of the test. The pure aluminum rod was melted and refined in a steel crucible at approximately 740 °C. The melt was left to stand at 720 °C for about 10 min to ensure the equilibrium temperature after the refining slag was skimmed. Then, the aluminum melt was cast into the steel mold equipped with the nickel-plated copper rod, and after waiting until it had completely cooled and solidified, it was removed from the mold.



Figure 1. Electroplating Operation Console.

The metallographic specimen was first polished on different grit sandpaper, then with a combination of mechanical polishing and hand polishing on a velvet polish cloth with a solution of 0.5 μm aqueous magnesium oxide. The microstructure of Cu/Al bimetallic composite was observed by AXIO-type metallographic microscopy (OM). The Ultima IV X-ray diffraction (XRD) was used to analyze the types of intermetallic compounds in the transition zone of materials, with a voltage of 35 kV and a scanning speed of $10^\circ/\text{min}$; diffraction angle range was $10^\circ \leq \theta \leq 45^\circ$. Jade 6.0 software was then used to perform phase calibration on the acquired dates. The sampling locations of hardness and shear samples were shown in Figure 2. The hardness (HV) of the Cu/Al bimetallic composite were tested on the HXD-1000TM digital microhardness tester with a load of 200 g and loading time of 15 s. The cylindrical shear specimen with diameter of 16 mm and height of 6 mm was fabricated by the electric spark machine. The shear specimen was performed on HXD-1000TM electronic universal material testing machine with the speed of 1.0mm/s. Then, the FEI Scios-2 HiVac scanning electron microscopy (SEM) was used to analyze the shear surface of the Cu/Al bimetallic composite.

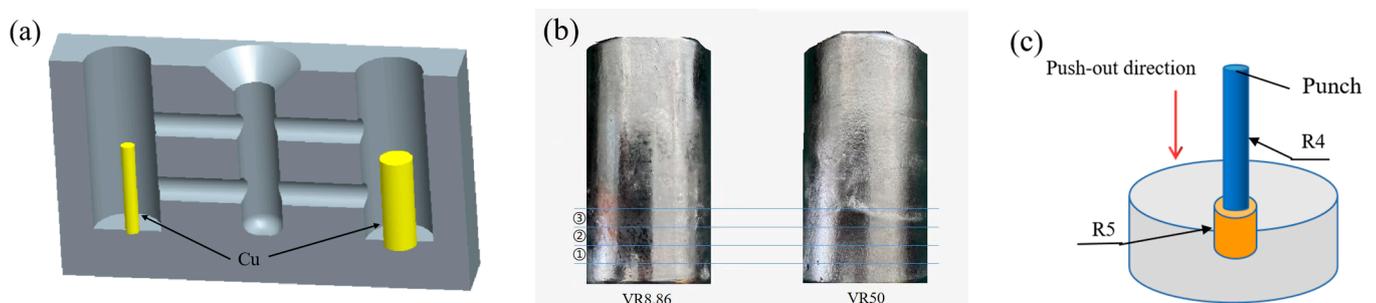


Figure 2. Casting mold drawing and shear material: (a) casting mold diagram; (b) Cu/Al bimetallic composite; (c) schematic diagram of shear strength test.

3. Results and Discussion

3.1. Effect of Liquid–Solid Volume Ratio on the Microstructure and Properties of Cu/Al Bimetallic Composite

3.1.1. Effect of Liquid–Solid Volume Ratio on the Microstructure of Cu/Al Bimetallic Composite

The Ni electroplating time for this part was 25 min. The metallographic structure of the Cu/Al bimetallic composite prepared at different liquid–solid volume ratio was shown in Figure 3. It was found that a distinct transition zone was formed at the junction of Cu and Al when casting at 720 °C. The thickness of the transition zone increases with the increase of the liquid–solid volume ratio, as the solidification time increases with the increase of the high-temperature liquid volume ratio, which means that Al and Cu atoms have a longer diffusion time. Figure 4 displayed the statistical results of dissociation thickness of the Cu and the thickness of the transition zone. With increasing the liquid–solid volume ratio of Cu and Al from 8.86 to 50, the thickness of the transition zone increased from 242.3 μm to 286.3 μm and the dissolved thickness of Cu increased from 74.3 μm to 90.3 μm . The ratio between the thickness of the transition zone and the dissolved thickness of Cu was 3.26 and 3.17 with increasing the liquid–solid volume ratio. Then, the ratio of Cu to Al in the transition zone was constant with increasing the liquid–solid volume ratio. Therefore, it is reasonable to infer that the phase composition of the transition zone did not change with the liquid–solid volume ratio, which was consistent with the literature [28].

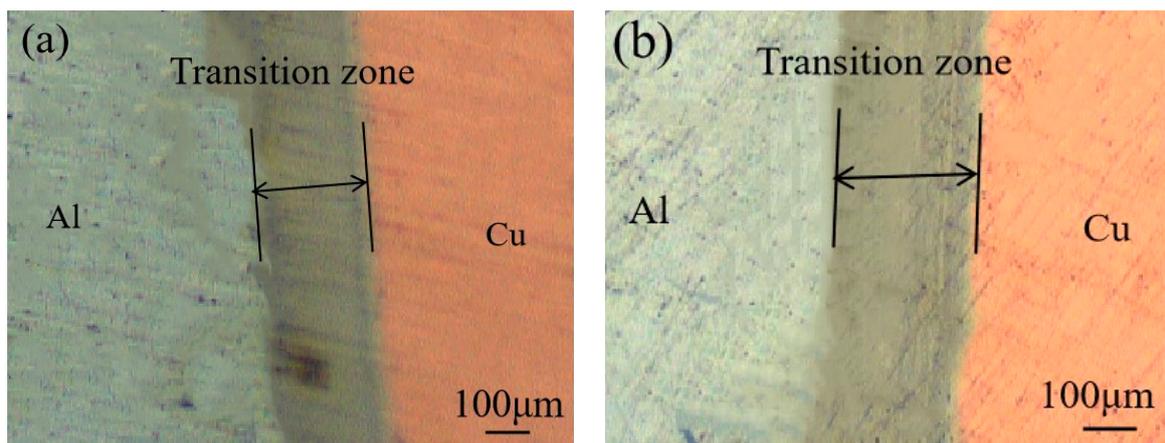


Figure 3. Metallographic structure of Cu/Al bimetallic composites prepared with different liquid–solid volume ratios: (a) VR8.86; (b) VR50.

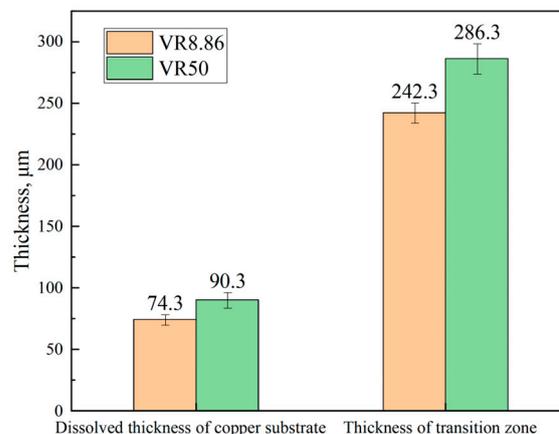


Figure 4. Thickness of the copper matrix and transition zone in composite fabricated with different liquid–solid volume ratio.

3.1.2. Effect of Liquid–Solid Volume Ratio on the Properties of Cu/Al Bimetallic Composite

(1) Shear strength

The shear strength of the Cu/Al bimetallic composite was described in Figure 5. The shear strength of the Cu/Al bimetallic composite increased with increasing the liquid–solid volume ratio. As shown in Figure 5, the shear strength was 17.8 MPa and 30.3 MPa for VR8.86 and VR50, respectively. With the increasing volume ratio from 8.86 to 50, the shear strength of the Cu/Al bimetallic composite increased 70%. As discussed above, the thickness of the transition zone was increased with the liquid–solid volume ratio. In addition, the microhardness of the phase (Al_2Cu , AlCu , and Al_4Cu_9) in the transition zone was higher than the pure Cu. This may account for the fact that the shear strength increased with the thickness of the transition zone. To further confirm this phenomenon, metallographic and SEM observation of the shear fracture of the Cu/Al bimetallic composite were performed.

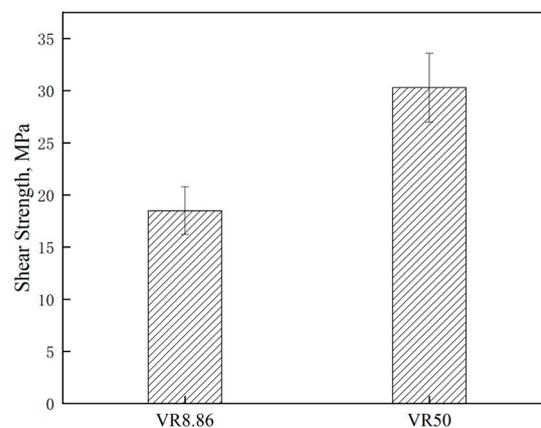


Figure 5. Shear strength of Cu/Al bimetallic composite fabricated with different liquid–solid volume ratio.

Figure 6 depicted the metallographic observation of the shear fracture of Cu/Al bimetallic composite fabricated with different liquid–solid volume ratio. It was shown that the yellow was Cu and some intermetallic compounds remained at the edge of Cu after the shear test. In addition, the content of the remaining intermetallic compounds remained at the edge of Cu increased with the thickness of the transition zone. This phenomenon indicated that the shear fracture was directly related to the transition zone.

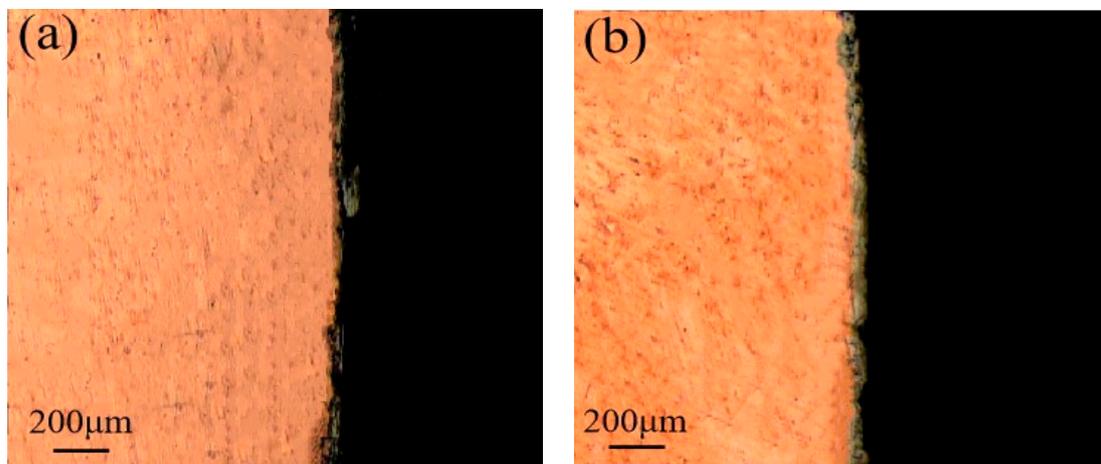


Figure 6. Metallographic observation of shear fracture of Cu/Al bimetallic composite fabricated by different liquid–solid volume ratio: (a) VR8.85; (b) VR50.

As displayed in Figure 7, XRD results of shear fracture surface of Cu/Al bimetallic composite indicated that the Al_2Cu , Al_4Cu_9 , and AlCu phase remained at the surface of the Cu rod after the shear test. The SEM photograph of the shear fracture of the Cu/Al bimetallic composite fabricated by different liquid–solid ratio was shown in Figure 8. Combined with energy spectrum analysis and XRD results, the phase calibration of the shear fracture was illustrated in Figure 8. In summary, it was concluded that the initiation and propagation of shear cracks occurred in the transition zone of the Cu/Al bimetallic composite.

(2) Microhardness

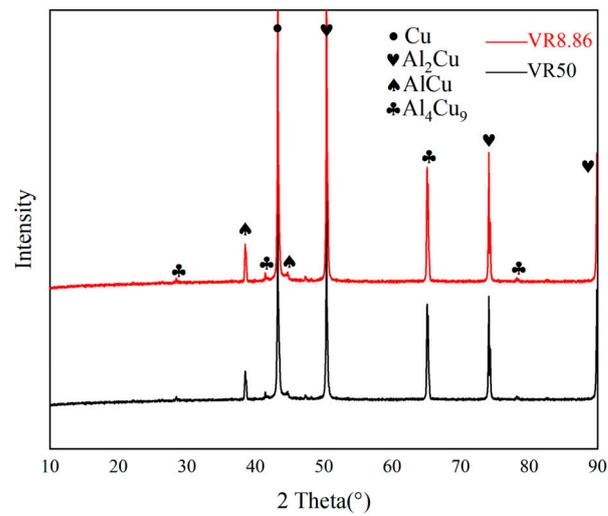


Figure 7. XRD patterns on shear fracture of Cu/Al bimetallic composite fabricated with different liquid–solid volume ratio.

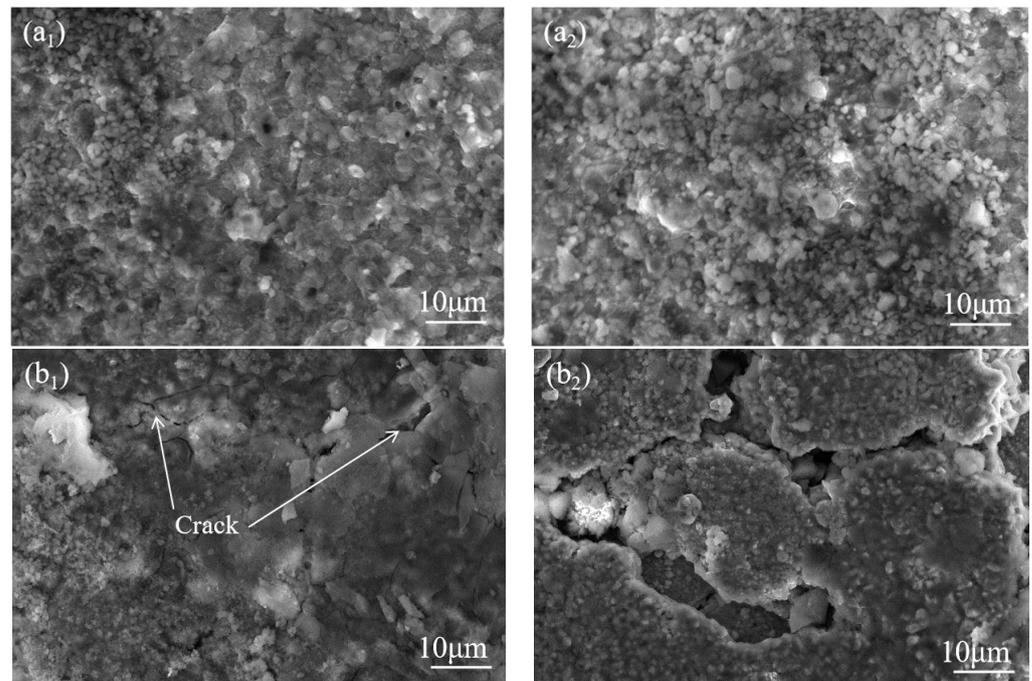


Figure 8. SEM photograph of shear fracture of Cu/Al bimetallic composite fabricated with different liquid–solid volume ratio: (a₁, a₂) VR8.86; (b₁, b₂) VR50.

The distribution of microhardness indentations and statistical results for the interfaces of Cu/Al bimetallic composite fabricated with different liquid–solid volume ratio were depicted in Figure 9, from left to right: Al, transition zone, Cu. It was reported [29] that the hardness of Al_2Cu was 400–500 HV, and the microhardness of $[\alpha(Al) + Al_2Cu]$ eutectic structure was about 150–200 HV. The microhardness of the transition zone of Cu/Al bimetallic composite was 140–180 HV, which was higher than Cu and Al. In addition, with the increase of liquid–solid volume ratio, the thickness of the intermediate transition layer increased, the content of the mesophase was higher, and the microhardness was increased.

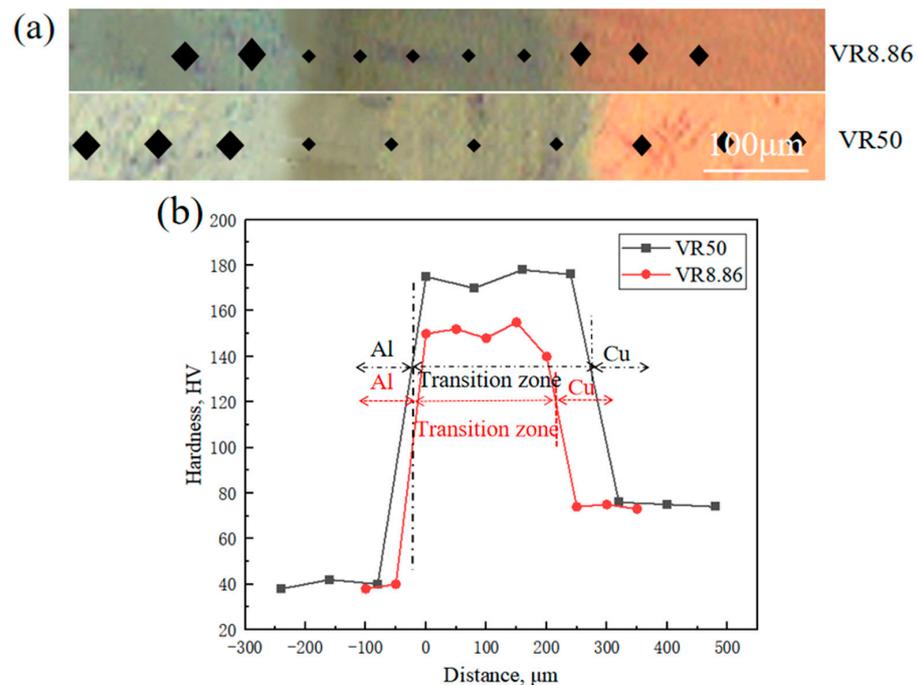


Figure 9. Microhardness of composite fabricated by different liquid–solid volume ratio: (a) indentation metallographic observation; (b) hardness distribution.

3.2. Effect of Coating Thickness on the Microstructure and Properties of Cu/Al Bimetallic Composite

3.2.1. Effect of Coating Thickness on the Microstructure of Cu/Al Bimetallic Composite

The coating thickness of the Ni layer on the Cu rod was 1.5 μm, 3.8 μm, and 5.9 μm with prolonging of the electroplating time from 10 min to 40 min, respectively. Then, the Al melt was cast into the mold equipped with an Ni-plated Cu rod, and the liquid–solid volume ratio was 50. The interfacial metallographic structure of Cu/Al bimetallic composite with different electroplating time was displayed in Figure 10. The transition zone width increased from 246 μm to 286 μm with the coating thickness varying from 1.5 μm to 3.8 μm. It was found that the thickness of the Ni plating was very uneven according to the metallographic observation of the Cu rod with Ni plating time of 10 min. Even some parts of the Ni layer had fallen off while kept at 500 °C. Then, the anti-oxidation effect of the Ni layer was sharply reduced, and part of the surface of the Cu rod was oxidized before casting the Al melt. This accounted for the smaller transition zone thickness for the electroplating time of 10 min.

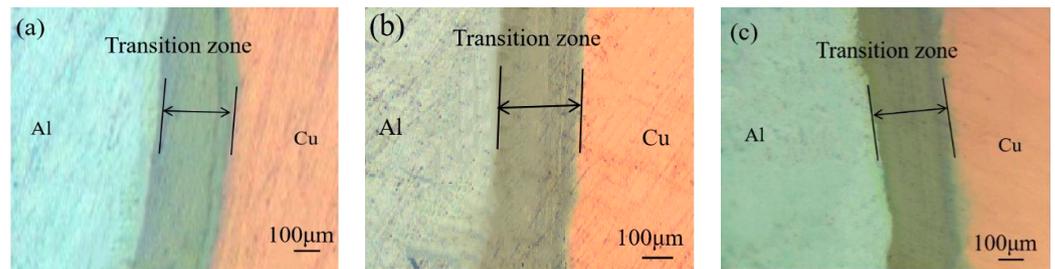


Figure 10. Interface microstructure of Cu/Al bimetallic composite with different electroplating time: (a) 10 min, (b) 25 min, and (c) 40 min.

The coating thickness and the width of the transition zone were summarized in Table 1. As shown in Table 1, the transition zone width first increased and then decreased with increasing the coating thickness of Ni from 1.5 μm to 5.9 μm . Exactly, the transition zone width decreased from 286 μm to 268 μm with the coating thickness varying from 3.8 μm to 5.9 μm . SEM image and chemical elements mapping of Al, Ni, and Cu of the Cu/Al bimetallic composite fabricated with 3.8 μm coating thickness was shown in Figure 11 Combined with Figure 10, it was inferred that the Ni layer on the Cu rod was dissolved by pouring in the high temperature Al liquid at the beginning of the casting. Subsequently, part of the Cu rod in contact with the Ni layer began to dissolve. Then, the Cu atoms diffused through the liquid Ni layer to the liquid Al, and the Al atoms diffused through the liquid Ni layer toward to the Cu side. At the same time, the Ni atoms diffused to both sides, which promoted the metallurgical combination of the Cu and Al. As the temperature dropped, different intermetallic compounds began to precipitate and transition zones gradually formed. As displayed in Figure 11, the 3.8 μm Ni layer had almost diffused out at elevated temperature. Therefore, the 3.8 μm Ni layer only partially limited the diffusion of the Cu and Al atoms, resulting in a peak thickness of the transition zone.

Table 1. Coating thickness and transition zone width of fabricated materials after different electroplating time.

Cu/Al	10 min	25 min	40 min
$d_{\text{clad.}} (\mu\text{m})$	1.5 ± 0.15	3.8 ± 0.2	5.9 ± 0.3
$d_{\text{tran.}} (\mu\text{m})$	246 ± 5.4	286.3 ± 6.8	268 ± 5.5

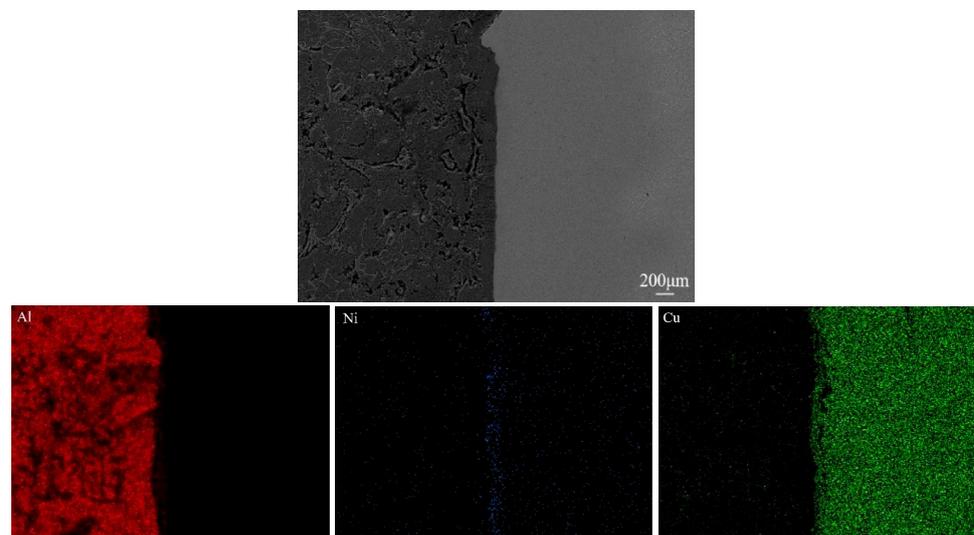


Figure 11. SEM image and chemical elements mapping of Al, Ni, and Cu of the Cu/Al bimetallic composite fabricated with 3.8 μm coating thickness.

The SEM image and chemical elements mapping of Al, Ni, and Cu of the Cu/Al bimetallic composite fabricated with 5.9 μm coating thickness was demonstrated in Figure 12. It was shown that there were still Ni layers distributed at the transition zone of the Cu/Al bimetallic composite with increasing the thickness of the Ni layer to 5.9 μm in Figure 12. The Ni layer was effectively limited the diffusion of the Cu and Al atoms at elevated temperature, which reduced the transition zone width to 268 μm .

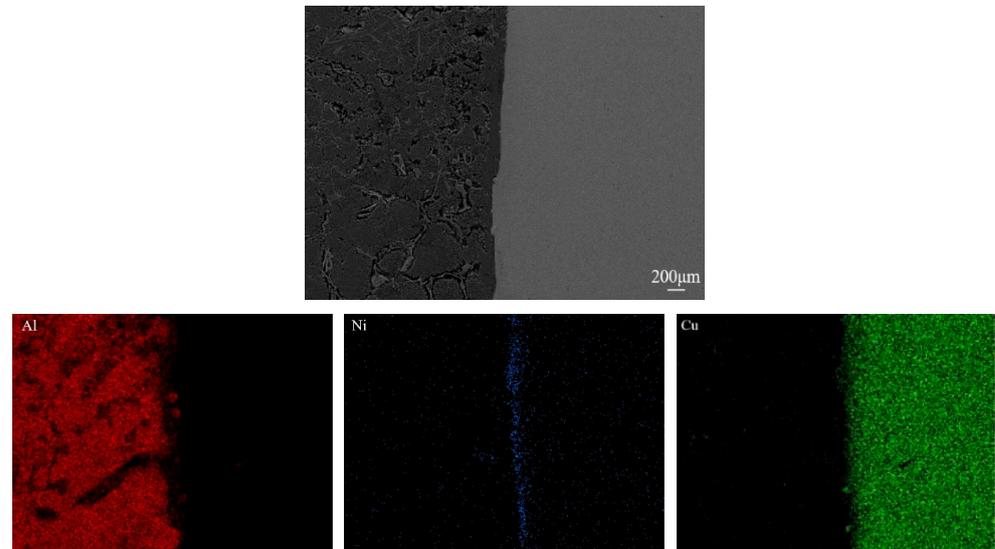


Figure 12. SEM image and chemical elements mapping of Al, Ni, and Cu of the Cu/Al bimetallic composite fabricated with 5.9 μm coating thickness.

The content of Ni in the transition zone increased with increasing the coating thickness of electroplating Ni. When the thickness of the Ni layer was 1.5 μm , the Ni layer had been diffused in the transition zone during the casting process. Therefore, the transition zone of the Cu/Al bimetallic composite with Ni layer thickness of 3.8 μm and 5.9 μm was characterized. Table 2 summarized the interface Al, Cu, and Ni elements content of Cu/Al bimetallic composite in Figures 11 and 12. The results also indicated that with increasing the thickness of the Ni layer, the diffusion of the Cu and Al was limited.

Table 2. Interface element content of Cu/Al bimetallic composite (wt, %).

Element	Time		
		25 min	40 min
Cu		49.84	44.73
Al		48.55	53.92
Ni		0.87	1.22

3.2.2. Effect of Coating Thickness on the Properties of Cu/Al Bimetallic Composite

(1) Shear Strength

The shear strength in transition zone of the Cu/Al bimetallic composite fabricated with different coating thickness was depicted in Figure 13. With increasing the coating thickness from 1.5 μm to 3.8 μm , the shear strength in transition zone increased from 18.6 MPa to 30.3 MPa, an increase of 62.9%. The shear strength decreased from 30.3 MPa to 26.7 MPa with the coating thickness varying from 3.8 μm to 5.9 μm . As discussed above, when the coating thickness was 1.5 μm , the Ni layer cannot effectively prevent the oxidation of the Cu rod, which results in poor metallurgical bonding of the Cu/Al interface. When the coating thickness was 3.8 μm , the Ni layer was effective in preventing the oxidation of the copper rod and promoting the mutual diffusion of Cu and Al atoms. Furthermore, due to the partial diffusion of the Ni layer, the binary phase structure in the transition zone

changed into the ternary phase structure, which resulted in a great improvement in the metallurgical bonding property of the Cu/Al interface. When the coating thickness was 5.9 μm , the Ni layer was not only effective in preventing the oxidation of the Cu bar, but also greatly limiting the mutual diffusion of Cu and Al, which had an adverse effect on the metallurgical bonding of Cu/Al interface.

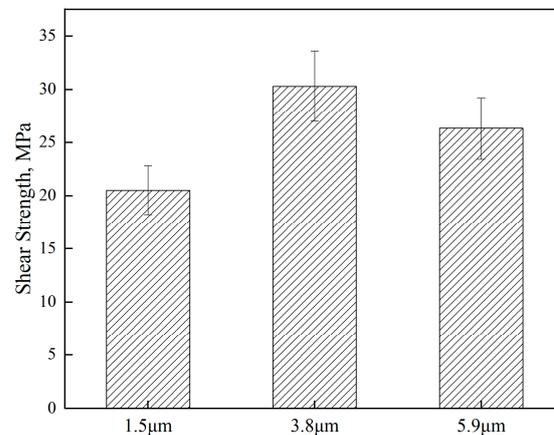


Figure 13. Shear strength of Cu/Al bimetallic composite fabricated with different coating thickness.

As illustrated in Figure 14, the phase composition of the transition zone changed with increasing the coating thickness of the Ni layer. For example, the diffraction of AlCu phase decreased with increasing the coating thickness of the Ni layer, indicating that the proportion of AlCu phase in the transition zone decreased. The diffraction of Al_3Ni_2 phase was found with the coating thickness of the Ni layer reached to 5.9 μm . Above all, the XRD results were consistent with the analysis of the influence of the Ni layer on the diffusion of Cu and Al.

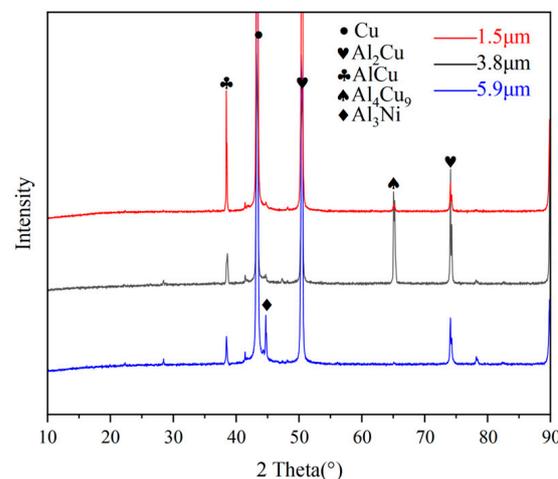


Figure 14. XRD results of shear fracture surface of Cu/Al bimetallic composite fabricated with different coating thickness.

Figure 15 depicted metallographic observations of shear fracture in the transition zone (Cu side) of Cu/Al bimetallic composite fabricated with different coating thickness. As shown in Figure 14, the content of residual intermetallic compounds at the Cu side increased with increasing the coating thickness of the Ni layer.

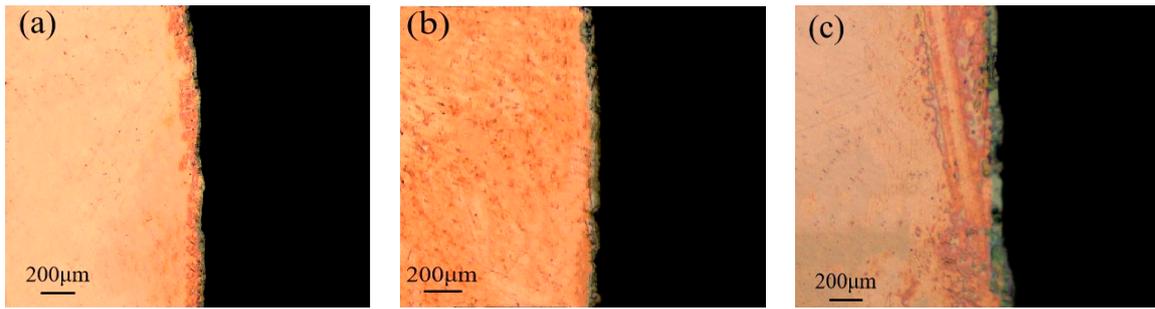


Figure 15. Shear fracture morphology of Cu/Al bimetallic composite fabricated with different coating thickness: (a) 1.5 μm ; (b) 3.8 μm ; (c) 5.9 μm .

The SEM morphology of the shear surface of the Cu/Al bimetallic composite fabricated with different coating thickness was displayed in Figure 16. Foam-like fluffy holes and cracks were found on the shear fracture surface in Figure 16a, which was consistent with poor metallurgical bonding due to surface oxidation of the Cu rod. As shown in Figure 16b, the foam-like fluffy holes disappeared and the length of cracks become shorter, indicating better shear strength than the former. As for Figure 16c, the Ni layer of 5.9 μm limited the diffusion of Al atom to the Cu side, resulting in a relatively low content of intermetallic compounds in the Cu side of the transition zone. Therefore, the morphology resembling a slip band appeared on the dissociation surface.

(2) Microhardness

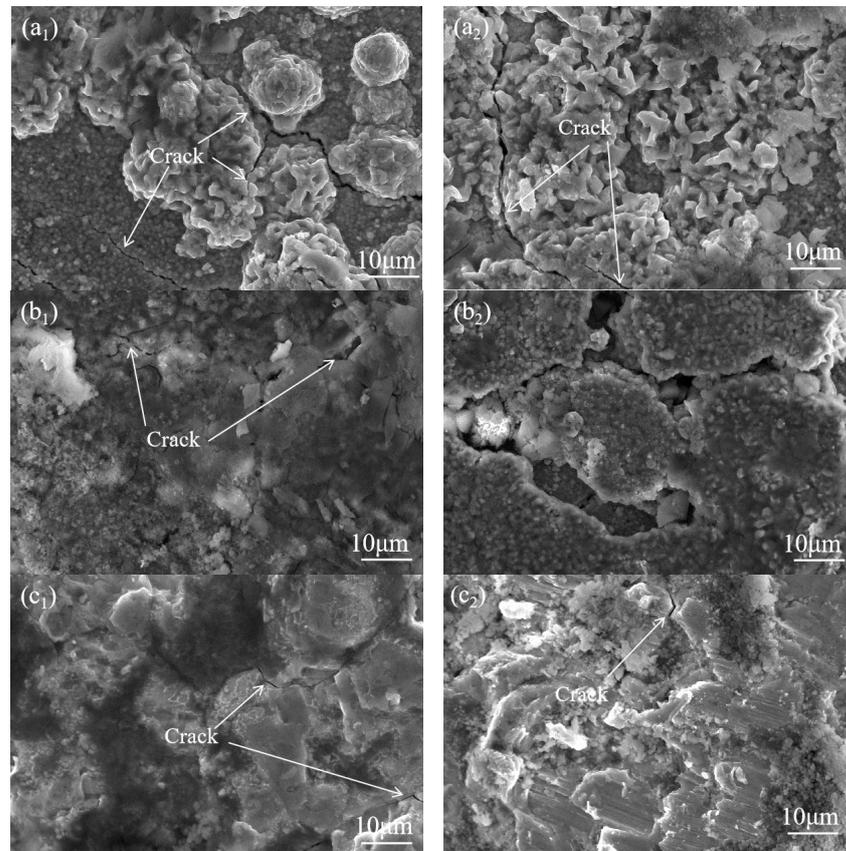


Figure 16. SEM morphology of shear surface of Cu/Al bimetallic composite fabricated with different coating thickness: (a₁,a₂) 1.5 μm ; (b₁,b₂) 3.8 μm ; (c₁,c₂) 5.9 μm .

Observations and statistical results of the interfacial microhardness of Cu/Al bimetallic composite fabricated with different coating thickness were described in Figure 17. The results showed that the micro-hardness of the Cu/Al bimetallic composite demonstrated a slight decreasing trend after a small increase, while the hardness of the interfacial compounds did not increase significantly. Ni elements were dissolved and diffused to the matrix under different electroplating times and different coating thickness, and the hardness of the electroplated Ni layer is 160–180 HV, which was close to the microhardness of the transition zone without the coating Ni layer. Therefore, it can be inferred that the Ni layer thickness has little effect on the microhardness of Cu/Al bimetallic composites.

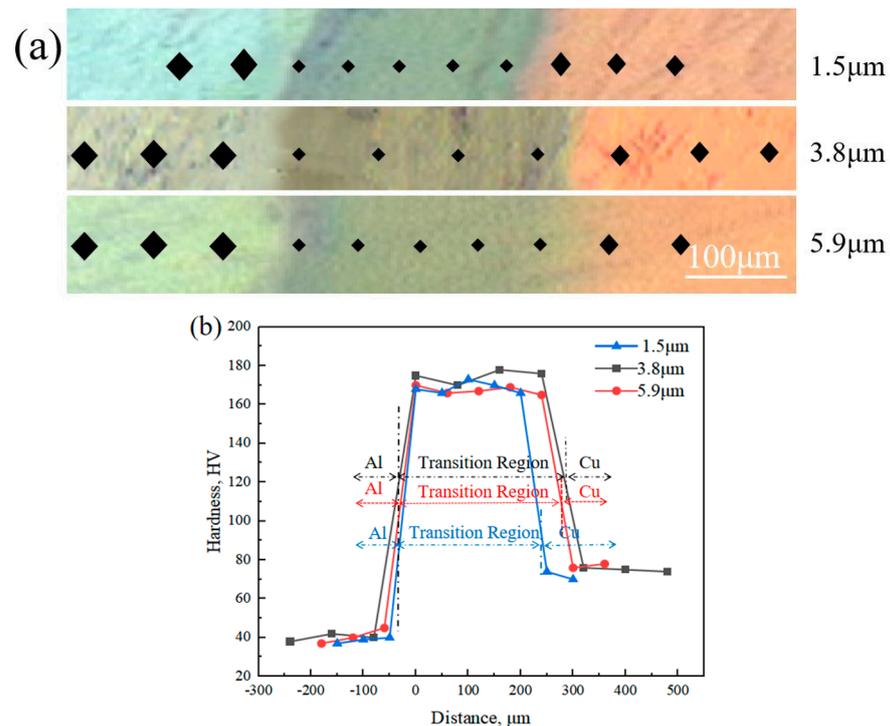


Figure 17. Microhardness of Cu/Al bimetallic composite with different coating thickness: (a) Indentation metallographic observation; (b) Hardness distribution.

4. Conclusions

The Cu/Al bimetallic composite was successfully fabricated by gravity casting. The effect of surface treatment and liquid–solid volume ratio on the microstructure and properties of the Cu/Al bimetallic composite were studied. The thickness of the transition zone was directly related to the mechanical properties of the Cu/Al bimetallic composite. Therefore, the quantitative regulation method of the thickness of the transition zone may become a new research hotspot in Cu/Al bimetallic composite fabrication. The following results and conclusions can be drawn:

- (1) The thickness of transition zone, shear strength, and microhardness of transition zone increased with increasing the liquid–solid volume ratio of Cu/Al bimetallic composite fabricated by gravity casting.
- (2) The thickness of transition zone and shear strength increased with the coating thickness of the Ni layer varied from 1.5 μm to 3.8 μm, due to the Ni layer effectively preventing oxidation on the surface of the Cu rod and promoting the metallurgical bonding of Cu/Al interface.
- (3) The thickness of the transition zone and shear strength decreased with increasing the coating thickness of Ni layer from 3.8 μm to 5.9 μm, due to the thick Ni layer limiting the diffusion of Cu and Al atoms.

- (4) The initiation and propagation of shear cracks occurred in the transition zone of Cu/Al bimetallic composite.

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