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In-Situ Observation and Analysis of the Evolution of Copper Aluminum Composite Interface

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Abstract: To study the micromorphology and dynamic evolution law of copper aluminum composite interface evolution, ultra-high temperature laser Confocal microscopy (CLSM) was used to observe and analyze the evolution of copper aluminum interface in situ, and then SEM, EDS and other advanced material analysis methods were used to observe the micromorphology of the composite layer, and study the composition of the interface layer and the formation process of the copper aluminum composite interface. The results indicate that the formation of the copper aluminum composite interface layer is mainly related to the mutual diffusion of copper aluminum atoms and the interface reaction between copper and aluminum. The bonding of the copper aluminum composite interface is mainly related to the melting of the metal surface of the interface layer and the mutual diffusion of copper aluminum atoms, which is the main mechanism of the copper aluminum composite interface bonding. The intermetallic compound is mainly Al_2Cu. In situ, observation of copper aluminum composite interface shows that there is a clear and relatively flat boundary between copper and the interface layer, while the boundary between aluminum and the interface layer is not straight, which is caused by the difference in thermal expansion coefficient, Lattice constant and hardness between intermetallic compounds and matrix and between intermetallic compounds. At the same time, it was found that there is a certain relationship between the visual changes of the copper aluminum composite interface image and reaction-diffusion migration during in-situ observation using a confocal laser scanning high-temperature microscope. Moreover, under no pressure, the oxide layer and interface inclusions can seriously affect the interface bonding.

Keywords: copper aluminum composite material; interface evolution; ultrahigh temperature laser confocal microscopy; in situ observation

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1. Introduction

The copper–aluminum composite has received extensive attention in recent years, as it combines the characteristics of high conductivity and thermal conductivity of copper with the lightweight and low cost of aluminum [1–3]. It has outstanding advantages in replacing copper with aluminum, especially pure copper, and is applied in aerospace, transportation, decorative building material and other fields [4,5]. The rolling composite method [6,7] and explosive welding composite method [8,9] are widely used in producing laminated metal composites. In addition, some manufacturing methods such as electromagnetic continuous casting [10], Ultrasonic welding [11,12], friction pressure welding [13,14], and brazing [15] have also been studied, developed, and applied to a certain extent.

Studying the interface evolution law of copper aluminum composite materials is of great significance [16–19], and controlling the degree of interface reaction is the key. Copper aluminum layered composite materials have varying degrees of influence on the deformation and deep processing processes of copper aluminum composite plates due to interface layers between copper and aluminum layers [20–24]. Yasuhiro Funamizu et al. [25] studied the diffusion of copper aluminum layered composite materials and found that copper aluminum composites can form five intermetallic compounds: AlCu, Al_2Cu, Al_2Cu_3, Al_3Cu_4, and Al_4Cu_9.

For the five intermetallic compounds produced at the interface of the copper-aluminium composites, Kouters [26] et al. prepared the copper-aluminium composite by heating and thermal diffusion. After 300 °C of heat treatment, they found that the intermetallic compounds at the interface were AlCu, Al_2Cu and Al_4Cu_9. Guo Yajie et al. [27] used plasma-activated sintering technology to prepare copper and aluminum composites. They obtained results similar to Kouters et al., showing that the intermetallic compounds at the interface were AlCu, Al_2Cu and Al_4Cu_9. They also investigated why Al_2Cu forms more easily at the interface than other intermetallic compounds and the interfacial layer thickness versus the heat treatment time. They showed that Al_2Cu forms preferentially at the interface relative to other intermetallic compounds.

Moreover, they studied the relationship between the interface layer thickness and the heat treatment time and calculated the activation energy [28] of various intermetallic compounds. L.Y. Sheng et al. [18] prepared copper-aluminum composite plates by cold rolling method and studied the variation law of the interfacial layer thickness with the heat treatment temperature and time. They found that when the interfacial layer reaches a certain thickness, its thickness changes very little with time and temperature. Won-Bar Lee et al. [29] used friction welding to prepare copper-aluminum composite plate joints and studied the growth rules of intermetallic compounds by using thermodynamic and kinetic theories. They calculated the diffusion activation energy of the five intermetallic compounds. Studying the solid reaction process of copper and aluminum and calculated the energy required for intermetallic compound generation, Y. Tanaka et al. [30,31]. The results show that at lower heat treatment temperatures, Al_2Cu is preferentially generated at the aluminum side at the interface, followed by Al_4Cu_9, which forms AlCu between Al_2Cu and Al_4Cu_9. Different preparation processes will generally lead to a different appearance at the interface of copper aluminum composite material. These studies are important for understanding the interfacial behavior and improving the properties of copper aluminum composites.

Many scholars have extensively researched the mechanisms and interface control of different composite material production processes. In the Cu/Al interface composite mechanism study, Liu Li et al. [32] analyzed the growth of intermetallic compounds on the composite interface layer of copper/aluminum composite plates under conventional annealing and high-temperature short-term annealing conditions. They explored the effects of annealing temperature, annealing time, and other factors on the growth of intermetallic compounds. In terms of the pressureless solid-liquid composite mechanism of copper/aluminum bimetallic materials, Zhang Hong'an et al. [33] believe that the metallurgical bonding of the copper aluminum composite interface is achieved through mutual diffusion of copper aluminum atoms on the interface layer and local melting of the metal. The formation of the copper aluminum composite layer is mainly related to the reaction between copper and aluminum interfaces and the mutual diffusion of copper aluminum atoms. The bonding mechanism of a solid-liquid interface composite is divided into two parts: diffusion bonding and fusion bonding. The interface structure is generally the result of the combined action of these two binding mechanisms. Yang Qingling et al. [34] observed in situ the growth and evolution of intermetallic compounds at the interface of copper aluminum bimetallic composites based on in-situ high-resolution Transmission electron microscopy technology.

The research shows that the formation mechanism and microstructure of the interface layer of copper aluminum composites play an extremely important role in its interface control and high-end applications. The research on the Spreading activation, dissolution, reaction, and compound formation of metal atoms at the interface of copper aluminum composites using in-situ analysis is rare, lacking systematic and scientific understanding, and needs further exploration and research.

In summary, current research on the interface microstructure, composite effect, and bonding mechanism of bimetallic composite materials mainly uses SEM, TEM, EDS, and material analysis and detection instruments to conduct post-analysis and test the prepared material samples [35]. The experiment of this subject relies on the advanced ultrahigh temperature laser confocal microscope of the State Key Laboratory of Refractory Materials and Metallurgy jointly built by Wuhan University of Science and Technology. At the same time, using advanced material analysis and characterization methods such as SEM and EDS, in-situ observation of the interface evolution of copper aluminum composites and analysis of the formation of compounds by copper aluminum atom diffusion were conducted.

2. Experimental Materials and Methods

2.1. In-Situ Observation at High Temperature

The ultrahigh temperature laser Confocal microscopy produced by the Japanese Lasertech/Yonekura company is used. The equipment combines advanced technologies such as laser confocal scanning, infrared heating, and tensile compression. The maximum temperature rise and fall rate is 300 °C/min, the maximum working temperature is 1700 °C, the temperature control is accurate, and the process conditions can be changed halfway. The whole experiment process is photographed, which meets the requirements of in-situ observation of the high-temperature microstructure evolution process of materials. This technology can observe and analyze the structural changes of melting, solidification, crystallization, and phase change of metal materials in real-time and with high definition, which has certain positive significance for the study of interdiffusion between composite material matrix, solid solution formation and the formation of intermetal compounds.

This article first uses 1060 pure aluminum and C1100 Copper to make diffusion couples in a vacuum environment at low temperature and low pressure in Gleeble and follows the size specifications of the crucible for the confocal laser scanning high-temperature microscope sample stage, taking ⌀5 mm × 3 mm circular thin plate was used as the sample, For the samples, coarse and fine grinding on 400 # to 2000 # and then polished with 2.5 μm diamond grinding paste on the polishing machine. Corrode, rinse and dry with hydrofluoric acid and iron chloride solution. The sample was heated according to the process curve shown in Figure 1. The solid-liquid composite interface of the copper aluminum diffusion couple during rapid heating to 500 °C and insulation was observed in situ using a confocal laser scanning high-temperature microscope (Lasertech/Yonekura, Tokyo, Japan).

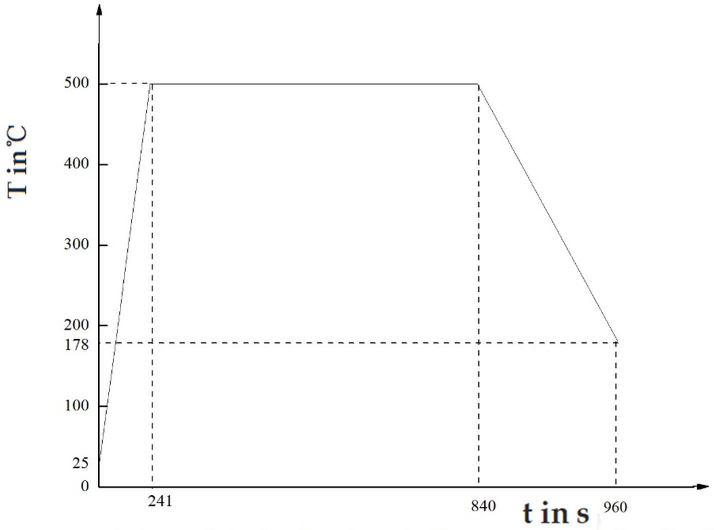


Figure 1. Copper aluminum composite in-situ observation process curve.

2.2. SEM Analyse

A field emission scanning electron microscope (model: JSM-7800F, JEOL, Tokyo, Japan) was used to scan the distribution of object and phase elements in the interface area. The acceleration voltage is 20 KV, and the working distance is 8 mm.

2.3. TEM Analyse

Transmission electron microscopy (JSM2100, JEOL, Tokyo, Japan) was used to analyze, observe, and determine the intermetallic compound composition and structure in the interface area with an accelerating voltage of 200 KV. The transmission sample was prepared by scanning electron microscope (JEOL, Tokyo, Japan) with FIB technology.

3. Experimental Verification

3.1. Analysis of In-Situ Observation Experiments

This article selects areas with good interface bonding for observation experiments, and Figure 2 shows the photograph of part of the copper-aluminum composite process under a light mirror. In Figure 2a–e are the sample interface topography at 100 °C, 200 °C, 300 °C, 400 °C, 500 °C, respectively, Figure 2f,g are the sample interface topography at 500 °C for 300 s and 599 s. Figure 2 shows that as the temperature continues to rise, copper and aluminum begin to diffuse towards each other. That is, through diffusion, copper and aluminum continuously "corrode" each other and form a clear diffusion front, as shown in Figure 2a–e.

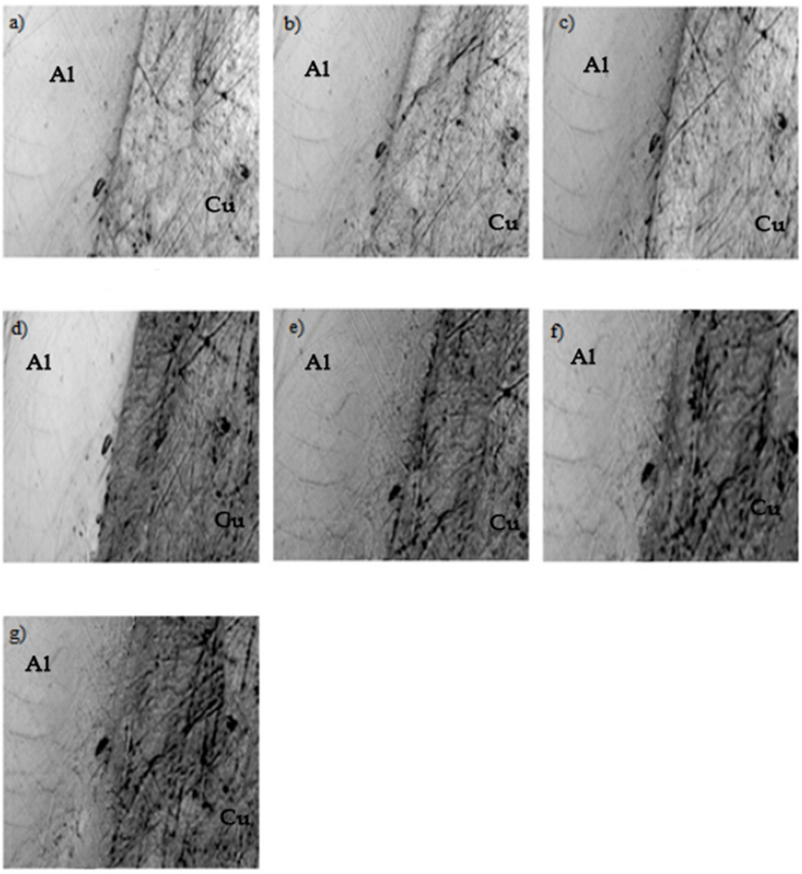


Figure 2. In situ observation images of the copper aluminum solid-liquid composite interface at different temperatures and times (**a**) 100 °C, (**b**) 200 °C, (**c**) 300 °C, (**d**) 400 °C, (**e**) 500 °C, (**f**) 500 °C (300 s), (**g**) 500 °C (599 s).

When the copper aluminum diffusion couple is heated to 500 °C, the aluminum substrate begins to soften, and black substances appear at the interface, as shown in Figure 2e.

Continuing to maintain insulation, the width of the black layer has been increasing, starting to penetrate the aluminum matrix, while the boundary line at the junction with the copper matrix is relatively regular and straight, as shown in Figure 2e–g. According to the imaging principle of confocal laser scanning high-temperature microscope, it can be concluded that there have been some changes in the height of the object surface. Therefore, it can be preliminarily determined that the black bands observed in this experiment are due to changes in surface tension and micro-volume changes caused by reaction-diffusion at the copper-aluminum composite interface. During the cooling process, black bands always exist.

3.2. SEM and EDS Analysis of Composite Interfaces

To study the microstructure and composition of the copper aluminum interface layer, the cooled diffusion couple was analyzed and characterized using a scanning electron microscope equipped with an energy-dispersive spectrometer. The results are shown in Figures 3–5. Figure 3 is the schematic diagram of the line scanning position. Figure 4 is the schematic diagram of the surface scanning area of the interface area, the distribution diagram of Cu and Al elements in the area, and Figure 5 is the result of line scanning in the interface area.

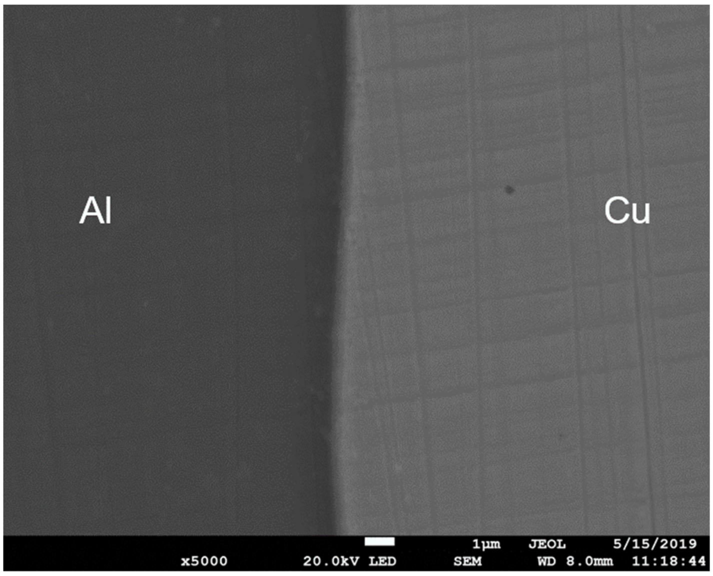
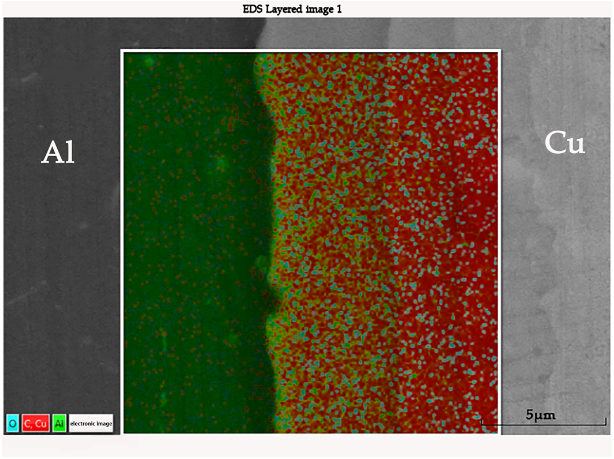
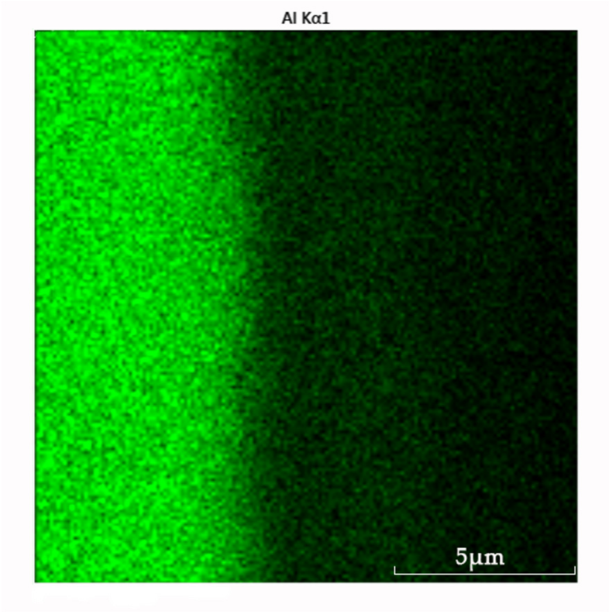


Figure 3. SEM and line scan range at the interface of the composite board.

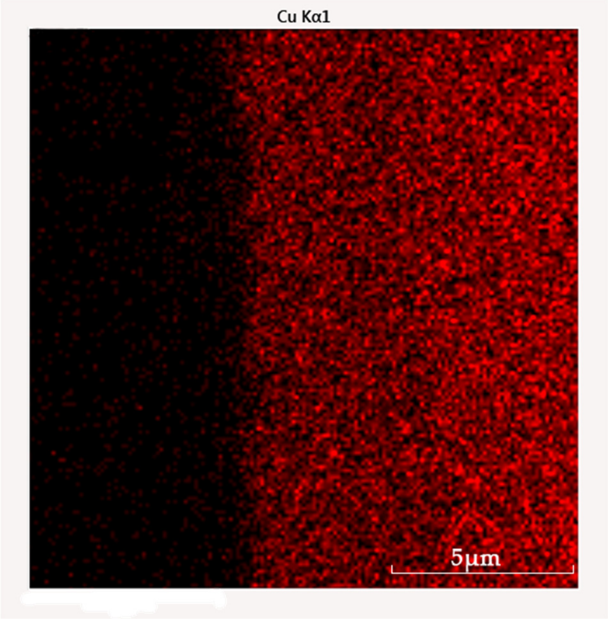


(**a**)

Figure 4. *Cont.*



(**b**)



(**c**)

Figure 4. EDS Image of Copper Aluminum Solid Liquid Interface Layer (**a**)—Copper aluminum EDS layered images; (**b**)—Al atom distribution map; (**c**)—Cu atom distribution map).

From the area with a good interface bonding effect, it can be seen that there is a clear diffusion layer between copper and aluminum metals, which is different from the metal matrix on both sides. The diffusion layer close to the copper substrate has a relatively uniform composition and narrow thickness, while the diffusion layer close to the aluminum substrate has an uneven composition and wider thickness. The boundary between the copper aluminum composite layer and copper is a relatively flat and clear straight line, while the boundary adjacent to the aluminum side is not straight. On the contrary, it presents an irregular shape. This is because the hardness and thermal expansion coefficient of copper and aluminum are different, and the crystal structure of copper and aluminum is different, resulting in the mismatch of the Lattice constant. Based on this, internal stress will be generated during the growth process of intermetallic compounds and increase with the growth of intermetallic compounds, resulting in the uneven performance of the copper-aluminum boundary at the interface [36]. In addition, in terms of diffusion direction, due to the differences in crystallographic structures between copper and aluminum atoms, the difference in diffusion speed of copper in aluminum and aluminum in copper also leads to an uneven interface. This is consistent with the interface line between the black band and the metal substrate on both sides during the in-situ observation experiment. The line scan image is shown in Figure 3. The line scan results show that from the aluminum matrix to the copper matrix, the atomic concentration of aluminum gradually decreases from the highest value through the composite interface and reaches the lowest value in the copper matrix. In the same direction, the change in copper atomic concentration is opposite to that of aluminum. From the horizontal axis of the line scan image, it can be seen that the thickness of the bonding layer is consistent with the diffusion width of copper aluminum atoms, indicating that the metallurgical bonding layer formed by the mutual diffusion of copper aluminum atoms is the copper aluminum interface layer.

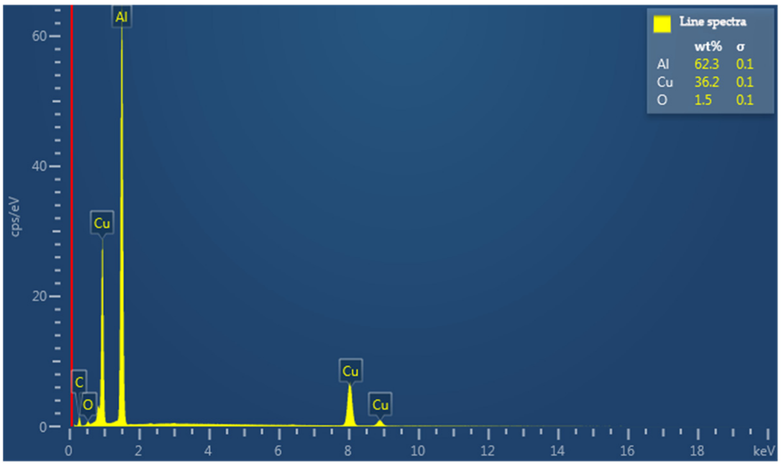


Figure 5. EDS Component Analysis of Interface Layer.

To further determine the diffusion and phase types of copper and aluminum atoms in the interface layer, an energy spectrum analysis was conducted on the interface layer and its adjacent areas. The EDS image of the copper aluminum solid-liquid interface layer (Figure 4) showed that the copper aluminum interface experienced a more intense interface reaction and diffusion behavior after the aluminum matrix entered the liquid phase. There were no aluminum atoms far from the interface on the copper side and no copper atoms far from the aluminum side. There are fewer copper and aluminum atoms only in the vicinity of the interface layer, indicating that the diffusion of copper and aluminum elements is only in the interface layer, and it can be seen that the diffusion depth of copper atoms in the matrix aluminum is significantly greater than that of aluminum atoms in the copper matrix. It can be determined that the black belt is mainly an intermediate product generated by the reaction-diffusion at the copper aluminum composite interface, and according to the EDS composition analysis results of the interface layer (Figure 5), Al_2Cu, a copper aluminum compound, appears in the interface layer, indicating that the main component of the composite layer is Al_2Cu.

It can be seen from the analysis results in Figure 6 that the interface contains C, O, and other impurity elements, which may be due to some inclusions left on the interface when making the sample. Due to the influence of inclusions and without pressure, copper and aluminum cannot be fully wetted, which seriously hinders and affects the interfacial reaction-diffusion. Numerous studies have found that applying a certain amount of pressure to the solid-liquid interface helps to wet between metals. At high temperatures, the aluminum liquid will react with oxides to generate other substances, leading to a reaction-diffusion between the aluminum liquid and copper matrix, thereby achieving interface bonding.

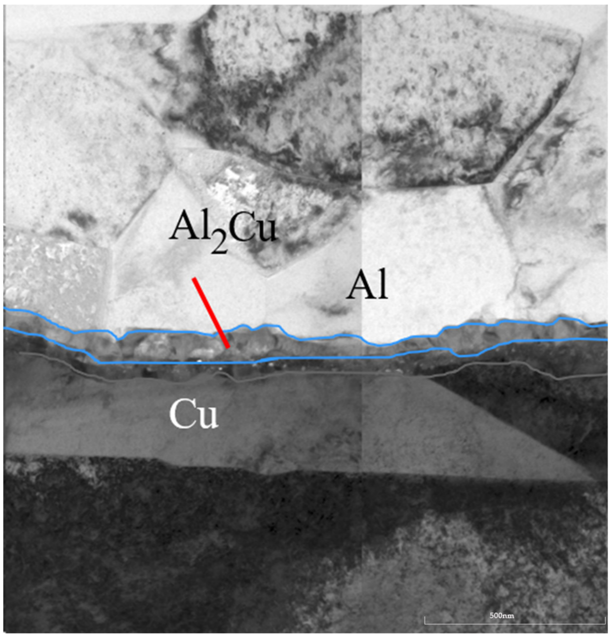


Figure 6. The TEM image at the interface of the composite plate.

3.3. *TEM Analysis of the Intermetallic Compounds in the Interface Zone*

Al_2Cu Through the TEM image (Figure 6), It can be seen that there is a layer of intermetallic compound between copper and aluminum through the Al_2Cu PDF card (JCPDS652695,14/mcm scarce group, a = 6.066 nm) and Figure 7 Medium diffraction pattern computational analysis identified this layer of intermetallic compound Al_2Cu.

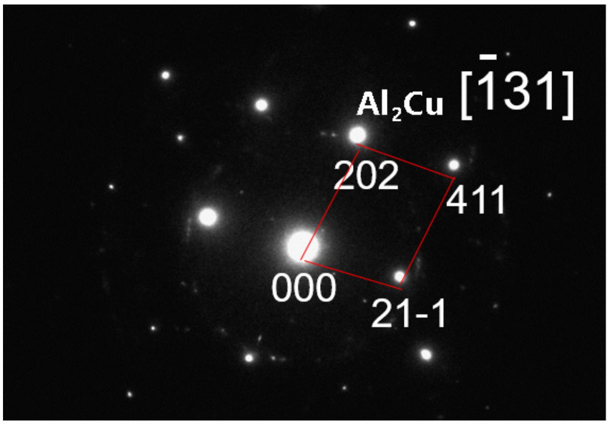


Figure 7. Electron diffraction pattern at the interface of the composite plate.

3.4. *Analysis of Interface Bonding Mechanism of Copper Aluminum Solid Liquid Composite*

Literature studies have shown that achieving metallurgical bonding at the copper aluminum composite interface is achieved through the combined action of interface fusion, interface diffusion, and interface reaction [36–38].

Interface fusion: Under high-temperature conditions, the solubility of each metal in another metal will continuously increase, and with time, copper atoms will continuously dissolve in the aluminum liquid, resulting in interface fusion. As time increases, the copper atoms will continuously diffuse from the copper to the aluminum part, exchanging positions with the aluminum atoms. This diffusion process leads to a rearrangement of the atoms at the interface, resulting in a continuous lattice structure between the two metals and a stronger intermetallic atomic binding at the interface.

Interface diffusion: Copper and aluminum atoms can undergo finite interdissolution in a solid state and infinite interdissolution in a liquid state. Under high temperatures, copper and aluminum atoms at the interface will get enough energy so that copper atoms can diffuse to the aluminum matrix and dissolve in aluminum liquid to form a Solid solution with aluminum as the solvent and copper as the solute. Aluminum atoms also diffuse to the copper matrix and dissolve in the copper matrix to form a Solid solution with copper as the solvent and aluminum as the solute. Copper and aluminum atoms penetrate and diffuse with each other, forming a certain thickness of the interface layer, thereby achieving metallurgical bonding. Meanwhile, the mutual diffusion of copper and aluminum atoms provides certain conditions for the formation of metal compound layers.

Interface reaction: according to the results of scanning electron microscope, energy spectrum analysis and in-situ observation, at high temperatures, when the aluminum matrix on the interface melts, Metallic bonding will be formed between copper atoms and aluminum atoms with sufficient energy. After copper and aluminum form a Solid solution, when the concentration reaches a certain range, intermetallic compounds will be formed, such as the intermediate product Al_2Cu, to achieve copper-aluminum metallurgical bonding. At the same time, copper atoms dissolved in aluminum liquid will also precipitate Al_2Cu during the solidification process. At 500 °C, Al_2Cu is first formed between copper and aluminum, followed by Al_4Cu_9 [39,40]. In addition, the formation energy of Al_4Cu_9 is 0.83 eV, which is higher than the formation energy of Al_2Cu, which is 0.78 eV [27]. Al_2Cu is formed earlier. At the initial stage of copper aluminum contact, copper and aluminum diffuse each other under thermal activation to form Cu (Al) and Al (Cu) Solid solution. According to the copper aluminum binary phase diagram, within this temperature range, the solubility of copper Solid solution formed in aluminum is about 3%.

In comparison, the solubility of aluminum in copper is as high as 19%. The diffusion rate of copper in aluminum is (6.5×10^{-5} m^2/s) is much faster than the diffusion rate of aluminum in copper (4.5×10^{-6} m^2/s) [41], the Al (Cu) Solid solution is saturated first than the Cu (Al) Solid solution, and Al_2Cu is formed at the side near the aluminum in the interface zone. The theoretical atomic concentration percentages of copper and aluminum atoms in the interface layer are 66% and 33%. With the continuous casting and rolling process, the Al_2Cu phase grows up in the direction perpendicular to the interface and finally spreads out [42].

4. Conclusions

Observe the copper aluminum composite interface in situ. At the copper aluminum composite site, the interface layer differs from the metal matrix on both sides. The boundary between copper and the interface layer is obvious and straight. Due to the difference in thermal expansion coefficient, Lattice constant and hardness between the matrix and intermetallic compounds, the boundary between aluminum and the interface layer is uneven and irregular.

The bonding of copper aluminum composite interfaces requires a combination of interface fusion, interface diffusion, and interface reaction. Local fusion and high-temperature reaction-diffusion are the main mechanisms of interface bonding in copper aluminum composites.

Under no pressure, the oxide layer and interface inclusions can seriously affect and weaken the bonding effect of the interface.

Through SEM and EDS analysis of the diffused copper aluminum material, it can be concluded that after heating up, the copper aluminum atoms diffuse with each other under temperature. The diffusion depth of copper atoms in the aluminum matrix is significantly greater than that of aluminum atoms in the copper matrix. During the diffusion process, a solid solution is formed, and as the diffusion progresses, the concentration of the solid solution gradually increases. When the equilibrium concentration is exceeded, a supersaturated solid solution is formed, and copper and aluminum diffuse to form intermetallic compounds at the junction after heating. According to energy spectrum analysis, it is preliminarily determined as Al_2Cu. Further TEM analysis of intermetallic compounds in the interface zone of copper aluminum composite materials determined that the intermetallic compound is Al_2Cu.

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