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Effect of Mechanical Surface Treatment on the Bonding Mechanism and Properties of Cold-Rolled Cu/Al Clad Plate



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Abstract

In the case of valuable cold-rolled Cu/Al clad plates, billet surface treatment before rolling is a significant process that can affect the bonding efficiency and quality. While the current studies primarily focus on the influence of rolling parameters, insufficient attention has been paid to surface treatment. In this study, the effects of mechanical surface treatment on the bonding mechanism and bonding properties of cold-rolled Cu/Al clad plates were investigated. The results showed that different mechanical surface treatments have significant effects on the surface morphology, roughness, and residual stress. In addition, the effect of surface mechanical treatment on bonding quality was also observed to be critical. When the grinding direction was consistent with the rolling direction (RD), the bonding quality of the Cu/Al clad plates was significantly improved. After surface treatment along the RD for 20 s, the Cu/Al clad plates showed the highest shear strength (78 MPa), approximately four times as high as that of the unpolished samples. Simultaneously, the peel strength of this process was also significantly higher than that achieved via the other processes. Finally, on the basis of the surface morphology, roughness, and residual stress, the effect of surface treatment on the bonding mechanism and bonding properties of Cu/Al clad plates was analyzed. This study proposes a deeper understanding of the bonding behavior and bonding mechanism for cold rolled clad plates processed via mechanical surface treatment.

Keywords: Cu/Al clad plates, Cold rolling bonding, Surface treatment, Bonding mechanism, Bonding property

1 Introduction

Cu/Al clad plates not only inherit the high electrical conductivity, thermal conductivity, and formability from the substrate metals, but also possess the advantages of high corrosion resistance and precious metals conservation. They can be widely used in electronic communication, mechanical power transmission, architectural decoration, and daily necessities [1]. At present, preparation methods of Cu/Al clad plates primarily include solidsolid composite, liquid-solid composite, and liquid-liquid composite methods [2–4]. Due to the high diffusion

³ TYUT-UOW Joint Research Centre, Taiyuan University of Technology, Taiyuan 030024, China affinity between copper and aluminum, it is easy to form brittle intermetallic compounds processed above 120 °C [5]. Therefore, the most productive process for manufacturing Cu/Al clad plates is cold rolling bonding (CRB).

In recent years, many researchers have conducted indepth researches on the CRB process [6, 7]. The bonding properties can be affected by various factors, such as reduction ratio [8, 9], rolling temperature [10, 11], rolling speed [12, 13], rolling direction [14], annealing treatment [15, 16], and initial thickness of the material [14, 17]. However, little research has focused on the surface treatment before rolling. This meaningful improvement has also confirmed experimentally. Table 1 lists the bonding strengths of three surface treatment methods: mechanical treatment, shot peening, and chemical treatment.



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Table 1 Shear strength of three surface treatment methods

Surface treatment methods	Mechanical treatment	Shot peening	Chemical treatment
Shear strength (MPa)	78	51.6	35.1

It can be seen that the mechanical surface treatment method can achieve higher bonding strength. Several authors agree that the best surface treatment method involves the surfaces to be bonded being first degreased with acetone and then scratched using a wire brush [18]. It removes surface oil and provides appropriate surface roughness, which is beneficial for improving the interface bonding quality [19, 20]. Gao studied the effect of surface preparation on the bond strength of Al-St strips prepared by the CRB process [21]. The effects of belt grinding and wire brushing on the bonding strength were compared and analyzed, but the bonding mechanism was not analyzed. Therefore, there is still a lack of systematic research related to the effects of mechanical surface treatment on the bonding mechanism of clad plates. For this purpose, the effects of four different mechanical surface treatment methods on the surface microstructure and bonding properties were investigated in detail in this study, and the bonding and strengthening mechanisms were discussed.

2 Materials and Methods

Commercially pure copper T2 (1.5 mm thickness) and aluminum 1060 (2.5 mm thickness) cold-rolled plates were used as raw materials. Before the CRB process, the initial surface of the Cu and Al plates was ground via four different mechanical surface treatments: the parallel rolling direction grinding (PRDG) process, vertical rolling direction grinding (VRDG) process, 90° cross grinding (90° CG: VRDG + PRDG) process, and rotating wire brush grinding (RWBG) process. The first three processes were performed using a T-shaped wire brush, and the last one was performed using a bowl-shaped wire brush. The diameters of T-shaped and bowl-shaped wire brushes are 150 mm and 90 mm, respectively, and the material is copper-plated steel wire with a wire diameter of 0.3 mm. Grinding is performed by an automatic sander to ensure consistency of other process parameters. Before the grinding process, the plate height is adjusted so that the upper surface is at least 2 mm higher than the bottom end of the wire brush to ensure consistent pressure on the plate. The moving speed of the wire brush is adjusted according to the grinding time; a schematic diagram representing the grinding process is shown in Figure 1. The speed of the wire brush is 10000 r/min. Cu/



Al clad plates were prepared with a two-high laboratory rolling mill (95 mm diameters) without lubricant at room temperature. The reduction ratio is 55% and the rolling speed is 0.15 m/s.

The surface residual stress after mechanical treatment was measured using the iXRD-Portable Residual Stress Analysis System via the complete stress equation and ellipse fitting method. Cr K α and Cu K α radiations were used as the X-ray sources to measure the surface stress of Al and Cu, respectively. The roughness of the Cu and Al surface was tested using the TA620 mobile roughness meter. The residual stress and roughness measurements were repeated five times to obtain the mean value.

The tension-shear and peeling properties along the rolling direction (RD) were tested to evaluate the bonding quality with different surface treatments. The tension-shear and peeling tests were conducted according to the standards of GB/T6396-2008 and ASTM-D1876-01, respectively. Three samples for each test were prepared to obtain the average value. The tension-shear and peeling tests were carried out using an INSTRON 5969 instrument with a stretching speed of 0.2 mm/min and 10 mm/min, respectively. The fracture surface after the peeling tests was observed via scanning electron microscopy (SEM), and the element distribution was analyzed via energy dispersive spectrometry (EDS).

3 Results

3.1 Surface Characteristics

Figure 2 shows the surface morphology of Cu and Al plates with different mechanical treatments for 20 s. According to previous research, the brittle/hardening layer with many scratches can be formed on the surface after grinding with a wire brush [17]. For the VRDG and PRDG processes, the surface morphology is similar. It is obvious that the scratch direction is related to the grinding direction. The direction of the scratch resulting from the VRDG process is perpendicular to the rolling direction (Figure 2a, e), while that of the scratch resulting from the PRDG process is parallel to



the rolling direction (Figure 2b, f). Furthermore, the scratch edge appears to be defective, and the micro cracks can be clearly seen in the figures. For the 90° CG process (VRDG + PRDG), a brittle/hardening layer is formed on the treated surface, induced by the VRDG process. When the PRDG process is performed, a new brittle/hardening layer with scratches in the perpendicular direction is formed, with the original brittle/hardening layer being destroyed. This process divides the complete brittle/hardening layer into many lumps that adhere to the surface (Figure 2c, g). For the RWBG process, scratches with multiple directions are formed on the copper surface, and severe lamination and lumps of the brittle/hardening layer can be clearly observed in the Al side as shown in Figure 2h.

Figure 3 shows the surface morphology of the Cu plate after PRDG treatment for different durations. As can be seen, in the case of PRDG treatment for 10 s, the scratch with numerous lumps can be clearly observed in Figure 3a. With the increased treatment time, the edges of the scratch become clean and the defective lumps obviously decrease, as shown in Figure 3b, c. It is noteworthy that with longer treatment times (30 s), typical lamination of the hardening layer is observed, and the degree of lamination appears to be greater with an increased treatment time, as shown in Figure 3d, e. This means that the surface condition differs significantly with the treatment time, which may affect the final bonding quality.

Figure 4 shows the results of surface residual stress. The measurement obtains the results of strains based on detecting changes of the atomic plane lattice spacing, which are related to stress [22]. As can be seen, the original Al plate appears to be in an unstressed state, while the original copper surface exhibits a positive stress value, indicating significant tensile residual stress. With different mechanical treatments, both Cu and Al plates show obvious compressive residual stress (negative value). The residual stress order of different treatments on the Cu plate can be concluded as VRDG > RWBG > 90° CG > PRDG, while RWBG \approx VRDG > 90° CG > PRDG for the Al plate. Note that the samples treated by the PRDG process exhibit minimal surface compressive residual stress for both Cu and Al plates in the present study. The mean surface residual stress of PRDG treatment for Cu plate is -48 MPa, approximately one-third of that of VRDG treatment. The surface residual stress of the Al plate exhibits a similar trend. Furthermore, the surface residual stress increases gradually with the increased treatment time.

Figure 5 shows the surface roughness of raw plates and samples with different mechanical treatments. As can be seen, the Cu and Al raw plates exhibit a relatively smooth surface with a roughness of 0.5 μ m. However, after mechanical treatment, the surface roughness increases significantly. Generally, the Al surface shows a higher roughness than that of the Cu surface, while







the surface roughness trend of the two plates appears to be coincident. The RWBG and 90° CG processes show a similar effect on roughness, which is higher than that of PRDG and VRDG processes, as shown in Figure 5. Furthermore, the surface roughness increases gradually with the increasing treatment time for certain mechanical treatments. The roughness of Cu samples treated with the PRDG process for 30 s is 2.18 μ m, approximately 118% higher than that of samples treated for 10 s. The above results indicate that the mechanical treatment method and treatment time significantly affect the surface roughness of the samples in the present study.

3.2 Tension-Shear Property

Figure 6 shows the effect of different mechanical treatments on the tension-shear property of Cu/Al clad plates. As can be seen, the Cu/Al clad plates without surface treatment exhibit poor tension-shear properties, with a break-off shear strength of 18 MPa. After mechanical surface treatment, the shear strength increases sharply. The samples treated via the PRDG process show the highest shear strength of 79.2 MPa. The shear strength order of different treatments for 20 s can be concluded as PRDG > VRDG > 90° CG > RWBG, as shown in Figure 6a. This indicates that grinding methods have a significant effect on the shear strength. Furthermore, according to the statistical results in





Figure 6b, the shear strength of Cu/Al clad plates generally increases to a peak value at 20 s and then decreases with the increase of grinding time.

3.3 Peeling Property and Fracture Characteristics

Figure 7 shows the peeling properties of Cu/Al clad plates. As can be seen, the samples without surface treatment exhibit a low peel strength at 15 N/cm. After surface mechanical treatment, the peel strength appears to increase in different degrees. The samples treated by PRDG show the peak value of peel strength as 133 N/cm, significantly higher than that of other treatments.

The peeling strength order of different treatments for 20 s can be concluded as $PRDG > VRDG > 90^{\circ} CG > RWBG$, corresponding with the tension-shear results mentioned above.

Figure 8 shows the fracture surface of the peeling test after treatment for 20 s. It can be observed that Al lumps adhered to the Cu side form ridges with certain aspect ratios (Figure 8a-d), corresponding to lateral cracks formed on the aluminum side (Figure 8e-h). The nonbonded area is relatively smooth and isolated by the Al ridges. For the VRDG process, Al ridges are short and narrow (Figure 8a). However, after PRDG processing, Al ridges appear to be wider in the rolling direction, forming a long strip-like distribution (Figure 8b). The scratch left by the wire brush can be observed on the interface, corresponding to the scratch left after the mechanical grinding process, shown in Figure 2b, f. After 90° CG processing, the peeling surface exhibits a similar phenomenon involving a long strip of Al embedded in the Cu side (Figure 8c, g). Furthermore, a number of lumps can be observed in the Cu surface, as shown in Figure 8c. According to the shape and element surface scanning analysis, the lumps form a brittle/hardening layer of Al, corresponding with the holes in the surface of the Al plate (Figure 8g). For the RWBG process, several nonbonded regions can be clearly seen in the fracture surface (Figure 8d, h), and the Al ridges are narrow and short, indicating a poor bonding quality.

Figure 9 shows the map scanning results of Cu plate in Figure 8. As can be seen, the strip embedded in the copper surface is identified as aluminum. The morphology and quantity of the Al strips can be clearly observed. In



addition, on the basis of the image processing software, the area ratio of the Al strip in the Cu surface was determined, as in Table 2. The PRDG process exhibits the largest area ratio of bonded aluminum as 44.04%, 19.5% higher than that of the RWBG process. The samples treated via VRDG and 90° CG processes show similar results (approximately 30%), corresponding to the low bonding properties. For further clarity, the correlation



 Table 2 Area ratio of aluminum on the copper side

Polishing methods	VRDG	PRDG	90° CG	RWBG
Area ratio of Al	30.76%±3.5	44.04%±4.1	31.23%±3.2	$24.54\% \pm 2.3$

analysis between peeling force and area ratio of the Al strip was conducted based on the SPSS software, as shown in Figure 9e. The allowable value for the significance coefficient p is <0.05. According to the statistical result, the value of p is 0.008, and the Pearson correlation r is 0.992. This indicates that the peeling force for the samples with different mechanical treatment is closely related to the area ratio of the Al strip.

4 Discussion

4.1 Surface Morphology

Mohamed [21] stated that the film theory is the primary bonding mechanism due to the low rolling temperature of the CRB process. The brittle/hardening layer formed by wire brush can be broken during the rolling process. The fresh metal on both sides is subsequently extruded and made to come in contact. When the pressure is high enough, a stable bond will be established between the two metals. Unbonded regions of the brittle surface layer are confined to small isolated islands [23]. Chen [5] studied the interface fracture mechanism and believed that the combination of the Cu–Al interface also includes mechanical bonding (virgin metal joint, mechanical lock).

According to the peeling and tension-shear tests, it is obvious that samples treated by the PRDG process for 20 s exhibit the highest bonding properties. According to the observation of surface morphology, the surface scratch direction for the PRDG process is consistent with the rolling direction and the scratched edges appear to be defective (Figure 2b, f). During the rolling process, the cracks may generate from these edge-defects and continue to grow (similar to the crack propagation during the tensile test), and the brittle/hardening layer may be cut off, forming a discontinuous fresh metal area. With further rolling deformation, multiple cracks will be interconnected to form wider and longer cracks that expose more fresh metal and provide more available bonding areas. In addition, the scratch direction is parallel to the rolling direction, minimizing the flow resistance of the metal during the CRB process. As a result, it is easier to form a mechanical lock between the substrates and improve bonding quality [5]. On the contrary, for the VRDG process, the surface scratch direction is perpendicular to the rolling direction, and the metal flow and crack growth will be hindered during the rolling process, promoting the small Al strips embedded in the Cu plate and poor bonding quality.

For the 90° CG process, more block-shaped brittle/hardened layers occurred (Figure 2c, g) due to two grinding (VRDG + PRDG) treatments. Since the two grinding directions are perpendicular to each other, the microstructure and properties of Cu and Al change drastically [24]. Furthermore, a portion of the brittle/hardened layer is detached from the substrate and fixed to the surface. Under the action of significant pressure and shear forces, aluminum blocks adhere to the copper side, forming mechanical bonds with low bonding properties. Therefore, there are numerous block-shaped aluminum fragments on the fracture surface of the peeling test (Figure 8c).

For the RWBG process, a brittle/hardening layer exhibits clear lamination (Figure 2d). Even if the outermost brittle/hardening layer is broken, the brittle/hardening layer of the bottom layer may hinder the bonding of fresh metal. In addition, a brittle/hardening layer on the aluminum side is also shredded into smaller blocks (Figure 2h), and fresh aluminum metal appears not to be easily exposed, resulting in the poor bonding condition.

4.2 Surface Residual Stress

Considering the stress condition of substrates during the rolling process, the surface residual stress may also affect the bonding behavior to some degree. As shown in Figure 4, after the different mechanical grinding processes, both Cu and Al plates exhibit obvious compressive residual stress. According to the characteristic of the residual stress test conducted via X-ray diffraction, the base direction of residual stress is parallel to the rolling direction. When compressive residual stress is present, a higher applied stress strength is required to achieve a given growth rate; conversely, when tensile residual stress is present, a lower applied stress strength is required to achieve the same growth rate [25]. In other words, tensile stress would promote crack formation, whereas compressive stress may prevent crack propagation [26]. After surface mechanical treatments, the residual stress is generated on the surface, which can be represented by the initial stress intensity factor (K_{res}) . When the total stress intensity factor (K_T) is greater than or equal to the toughness of the material (K_{mat}) , a fracture occurs [27]. It can be described as:

$$K_{res} + K_{app} = K_T \ge K_{mat},\tag{1}$$

where K_{app} is the stress intensity factor produced by the application of an external force. Under the same rolling conditions, the smaller compressive residual stress is more favorable for the fracture behavior of the brittle/hardened layer. The surface of the brittle/hardening layer

after the PRDG process shows the lowest compressive residual stress, and the effect of preventing crack propagation appears to be the worst. The brittle/hardened layer is also the most susceptible for cracking during rolling. Therefore, the bonding strength is higher, which was confirmed by the peel strength results illustrated in Figure 7. Considering the positive residual stress and unstressed raw Cu and Al plate, the un-treated Cu/Al clad plate should possess the better bonding quality. However, the results of tension-shear and peeling tests appear to be inconsistent with the deduction related to the surface residual stress. This indicates that the residual stress can affect the bonding quality, but not the only factor.

4.3 Surface Roughness

During the CRB process, the plasticity and deformation resistance of copper and aluminum are different, resulting in different flow rates of the two metals. On the interface, there is a relative displacement of the two metals. Therefore, roughness is another important factor affecting the bonding quality of metal clad plates [28].

As can be seen from Figure 5, the roughness increases with the increase of grinding time, and the shear strength also increases (Figure 6b) as the grinding time increases from 10 s to 20 s. This can be related to mean contact pressure. Mean contact pressure (P) of multilayer strip rolling can be calculated using the following formulation [14]:

$$P = \frac{F}{WL},\tag{2}$$

where *F*, *W*, and *L* are the rolling force, the strip width, and the length of roll contact arc, respectively. There is not enough friction at the interface as well as the increase of deformation inhomogeneity between layers due to low roughness. The metal tends to slide out along the smooth interface during the cold rolling process, which causes the bonding point to move toward the roll exit [29]. Under constant total deformation, increasing the roughness between the two metals is similar to increasing the friction coefficient between strips and rolls. An increase in the coefficient of friction results in a decrease in the contact length (L) of the strip as well as an increase in the average contact pressure (P), in Eq. (2) [23]. In addition, increasing the roughness may also result in an increase in the relative bonding length (by increasing the coefficient of friction of the rolled strip) to increase the application time of mean contact pressure on the cold bonding. At the same time, the increase in relative bond length is accompanied by movement of the bonding point towards the roll inlet [29], resulting in an increase in bonding strength. Therefore, the bond strength is enhanced by increasing surface roughness and the average peel strength [30].

Note that the shear strength is reduced when the grinding time exceeds 20 s (Figure 6b), which is inconsistent with the above discussion. Therefore, the bonding behavior should be considered by combining the above factors.

Furthermore, the elements analysis of the Cu surface after grinding for 10 s and 30 s along the rolling direction are investigated in Figure 10. The EDS results in Figure 10b and c show that the oxygen content is low, which indicates that the surface comprises primarily fresh copper after grinding for 10 s. Figure 10e, f show the line scan results after grinding for 30 s. It can be seen that the oxygen content increases sharply at the edge of the sheet-shaped brittle/hardening layer, which implies that a longer mechanical treatment time may generate too much heat, resulting in local surface oxidation and obstructions to the combination of the clad plates. Meanwhile, the surface morphology changes significantly during surface treatment. As shown in Figure 3, it can be observed that after grinding for 30 s, significant lamination of the brittle/hardening layer occurs. This lamination primarily resulted from the destruction of the brittle/ hardening layer formed in the earlier stage, and consisted of oxide and impurity, impeding the complete bonding of Cu and Al substrates.

As a result, with longer treatment time, the increase of residual stress and local surface oxidation may deteriorate the bonding behavior, counteracting the positive influence of surface roughness. Therefore, the bonding mechanism can be illustrated from the aspect of surface morphology, residual stress, and roughness, as shown in Figure 11. The optimal bonding properties of samples after PRDG treatment for 20 s should result in the highest synergistic effect of the above factors.

5 Conclusions

- After surface mechanical treatments, scratches and lumps form on the surface, and the surface stress condition and roughness are changed significantly. With the increase of treatment time, the surface compressive residual stress and roughness clearly increase.
- (2) Among the four mechanical grinding methods, PRDG process exhibits an outstanding influence on the bonding properties. After surface treatment along RD for 20 s, the Cu/Al clad plate demonstrates the highest shear strength (78 MPa).
- (3) The scratches parallel to the RD can reduce the flow resistance of the metal and promote breakage of the brittle/hardening layer formed during the surface treatment, contributing to the mechanical bonding.





The surface compressive residual stress induced by surface treatment may consume the rolling force, impeding the rolling bonding. Whereas, significant roughness may accelerate the rolling bonding due to the positive effect on the contact length and contact pressure during rolling.

(4) The optimal bonding properties of samples after PRDG treatment for 20 s should result in the highest synergistic effect of surface morphology, residual stress, and roughness.

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Authors' Contributions

JH, QH and AIP was in charge of the whole trial; HN, TW and SL wrote the manuscript; ZR and YJ assisted with sampling and laboratory analyses. All authors read and approved the final manuscript.

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Competing Interests

The authors declare no competing financial interests.

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