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ARTICLE

Effect of Surface Treatment on Bonding Strength of Aluminum-Magnesium Hot-Rolled Composite Plate

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Abstract: The effect of different surface treatments on the bonding strength of composite plates was investigated under the conditions of 400 °C and reduction ratio of 45%. Results show that the wire brush grinding treatment can only eliminate the oxide film on the plate surface, but it can hardly produce a hard layer on the plate surface. The bonding effect depends on the element diffusion promoted by the close contact between the metals on both sides of the interface. After anodic oxidation, there is a hard layer on the metal surface, and the hard layer broken during the rolling process forms a mechanical occlusion at the bonding interface. However, the hard layer cannot form an effective combination with the metal at the interface, and the bonding can only occur in the fresh metal bonding area at the crack of the hard layer. The acid-alkali washing treatment can completely remove the hard layer on the surface of both alloys without increasing the surface roughness of the plate, and the metal on both sides of the interface is more closely bonded during the rolling process. The optimal bonding strength can be obtained by surface treatment of acid-alkali washing for the aluminum-magnesium hot-rolled bonding.

Key words: aluminum-magnesium composite plate; surface treatment; hard layer; bonding strength

Magnesium alloys have the advantages of low density, high strength, shock absorption, and strong impact resistance, which are widely applied in the aerospace and automotive industries^[1]. However, the inferior corrosion resistance and plastic deformation of magnesium alloys restrict their applications to a certain extent^[2]. By contrast, aluminum alloys have good plasticity and corrosion resistance^[3]. Therefore, the metallurgical bonding between aluminum alloys and magnesium alloys attracts much attention for the combined advantages of the two materials and the compensation effect in the performance defects between magnesium alloys and aluminum alloys, extending the application field^[4]. The rolling bonding method has the advantages of high production efficiency and low cost, which is widely used in the production of aluminum-magnesium composite plates. However, in the rolling process, because of the large difference in plastic deformation of aluminum alloys and magnesium alloys, bonding can only be achieved under a large rolling reduction ratio or at high rolling temperatures^[5].

When the rolling temperature is higher than 500 °C, Al_2Mg_3 ^[6] and $Al_{17}Mg_{12}$ brittle phases are easily generated at the bonding interface of the aluminum-magnesium composite plate^[7]. In addition, the increasing reduction ratio may promote cracks^[8], which dissatisfy the production requirements. The defects can be avoided by lowering the rolling reduction ratio and the rolling temperature, but the bonding strength of the prepared aluminum-magnesium composite plate is generally low. Hence, the aluminum-magnesium composite plates with high bonding strength are difficult to achieve by simply adjusting the rolling process parameters. Surface treatment, as an important step before hot rolling, cannot only effectively remove impurities, such as oil stains, on the plate surface but also forms a special surface state, which directly affects the bonding quality of the composite plates^[9]. According to thin film theory, the rolling compositing process of metal mainly undergoes three steps. Firstly, the surface hard layer ruptures and explodes to reveal the underlying fresh metal, and this process is called plastic deformation. Secondly, hard layer

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fragments are embedded at the interface to form a mechanical bite. Thirdly, the fresh metal is squeezed into the crack, and the ones at the crack bottom are in contact with each other, forming metallurgical bonding^[10]. An important parameter affecting the bonding strength is the surface treatment^[11]. The main factors to achieve a good bonding between the sheets are high surface roughness of metals^[12] and the existence of desirable thin oxide layer^[13]. Therefore, in the preparation of laminated metal composite plates by rolling, wire brush grinding^[14-18] is often used to produce a hard layer on the surface for bonding, which also promotes the formation of intermetallic compounds. For example, Jamaati et al^[12] studied the effects of surface treatment, surface roughness, scratch brush parameters, and delay time between surface treatment and rolling on the bonding strength of aluminum strip. It is reported that the increase in rolling force, pressure, and surface roughness can lead to work hardening effect, thereby improving the bonding strength. Han et al^[19] found that in the Cu/Al composite plate, copper plate surface was produced by wire brush polishing, and the hard layer broken during the rolling process pierced the aluminum side. Meanwhile, the soft aluminum metal filled into the cracks of the hard layer, forming a good metallurgical bonding with the fresh metal on the copper side. The bonding strength of the composite plate was significantly improved, compared with that without hard layer. Other methods have also been used to produce hard layer on the metal surface for the quality enhancement of the rolled metal composite. For instance, Zhang et al^[20] found that different surface treatments produce different weld strengths. Mehr et al^[21] used the anodic oxidation surface treatment method to form a hard layer on the surface of aluminum plate. The hard layer broke during the rolling process and it was embedded at the bonding interface as a reinforced particle, therefore improving the bonding effect of the copper plate and aluminum plate. However, the rolling process is not applicable to all metals to produce hard layer. The existence of hard layer may influence the diffusion behavior between metals, which has a negative effect on the bonding strength of composite plates. The acid-alkali washing method can effectively remove the hardened layer on the metal surface, so the metal on both sides of bonding interface can produce enhanced diffusion behavior, thereby improving the bonding strength.

Therefore, on the basis of the aluminum-magnesium rolling bonding process, the influence of surface treatments on the bonding strength of aluminum-magnesium composite plates was studied. Aluminum-magnesium composite plates with high bonding strength were expected to be prepared under small reduction ratio and at a suitable temperature. In this research, steel wire brush polishing, anodic oxidation, and acid-alkali washing treatments were used to form different surface states of aluminum and magnesium alloys. The aluminum-magnesium composite plate was prepared by the hot rolling bonding method. The influence of surface states on the bonding strength of the aluminum-magnesium composite plate was analyzed through bonding strength, morphology, and element distribution of the composite plates after different

surface treatments.

1 Experiment

In this study, 5052 aluminum alloy and AZ31B magnesium alloy were selected as experiment materials, and their chemical composition is shown in Table 1 and Table 2, respectively. The billet sizes of the aluminum and magnesium alloys were 100 mm×40 mm×2 mm, and the billet was symmetrically assembled in accordance with the AA5052/AZ31B/AA5052 structure.

The to-be-bonded metal surface should be treated before hot rolling to produce a completely different state. Three surface treatments were conducted. (1) Wire brush polishing treatment: the plate surface was reciprocally polished with a bowl-type wire brush using an angle grinder along the rolling direction until the adsorbed layer on the plate surface was completely removed and a bright metallic luster was exposed, forming a hardened layer on the metal surface. (2) Anodic oxidation treatment: the acid-alkali washing process was conducted to remove the original oxide film on the sheet surface. The electrolyte of the aluminum alloy in the oxidation process consisted of 20 g/L oxalic acid and 140 g/L sulfuric acid. The electrolyte of the magnesium alloy was composed of 40 g/L sodium hydroxide, 80 g/L sodium silicate, and 5 g/L sodium citrate. The anodizing voltage was 25 V, and the oxidation duration was 20 min. Afterwards, a uniformly distributed hard layer was prepared on the plate surface. (3) Acid-alkali washing treatment: the corrosion liquid for the aluminum alloy was 2vol% nitric acid solution, and the corrosion liquid for the magnesium alloy was 100 g/L sodium hydroxide solution. To avoid excessive corrosion of the plate surface during the process of acid-alkali washing, the duration of acid-alkali washing was controlled within 5 min. Owing to the existence of hard layer on the plate surface, incongruity occurred at the interface of metal and matrix during the deformation process, and the hard layer of metal surface preferentially cracked during the deformation process, thereby exposing fresh metal of the matrix. Therefore, the unidirectional tensile test of the surface-treated plate was conducted by universal testing machine, and the surface morphology of the plate after tensile deformation was observed by JSMIT500 scanning electron microscope (SEM). Then, the surface state of the plate was judged, and the unidirectional tensile rate was 5 mm/min until the plate was pulled off. Because the plastic deformation of aluminum alloy

Table 1 Chemical composition of 5052 aluminum alloy (wt%)

Mg	Si	Mn	Fe	Cu	Cr	Ni	Ti	Al
2.41	0.14	0.17	0.28	0.02	0.16	0.02	0.04	Bal.

Table 2 Chemical composition of AZ31B magnesium alloy (wt%)

Al	Zn	Mn	Si	Ca	Ni	Cu	Fe	Mg
3.85	0.955	0.413	0.167	0.017	0.0039	0.0062	0.0048	Bal.

is better than that of magnesium alloy, the deformation degree of the aluminum alloy was greater during the stretching process. Fig. 1 shows the tensile morphologies of the aluminum alloy surface after different surface treatments. As shown in Fig. 1a and 1d, no obvious cracks can be observed on the aluminum alloy surface after wire brush grinding or acid-alkali washing treatment. This result indicates that the deformation between the aluminum alloy surface and the substrate is coordinated during the tensile deformation process. Thus, it is inferred that there is no obvious hard layer on the surface of aluminum alloy after the wire brush grinding treatment or acid-alkali washing treatment. The tensile surface morphologies of the aluminum alloy after anodic oxidation treatment present dense and uniform cracks, indicating that incongruity occurs between the surface of aluminum alloy and matrix during the tensile deformation process. Additionally, an obvious hard layer is assumed to exist and evenly distributed on the surface of aluminum alloy after anodic oxidation treatment.

2 Results and Discussion

2.1 Composite plate preparation and bonding strength

The aluminum and magnesium alloys after surface treatments were symmetrically assembled in the form of AA5052/AZ31B/AA5052 structure. Both ends of the billet were fixed with rivets and aluminum wire to prevent the misalignment between the plates during rolling process. Afterwards, the billet was placed in a heating furnace. When the billet temperature reached the rolling temperature, the billet was placed in a two-roll mill for rolling experiments,

and the roll size of the two-high rolling mill was $\Phi 200 \text{ mm} \times 200 \text{ mm}$. The rolling temperature was $450 \text{ }^\circ\text{C}$, and the rolling reduction ratio was 45%. The bonding strength of the aluminum-magnesium composite plate was evaluated via tensile-shear test with miniature control electron universal testing machine at tensile rate of 0.5 mm/min . Three tensile-shear specimens were tested under each condition. The schematic diagram of tensile-shear specimen is shown in Fig. 2, and the cutting direction was parallel to the rolling direction. Fig. 3 shows the bonding strength of aluminum-magnesium composite plate after different surface treatments. The bonding strength of the aluminum-magnesium composite plate after anodic oxidation treatment is the lowest. By contrast, the bonding strength of the aluminum-magnesium composite plate after wire brush grinding and acid-alkali washing can reach 74.8 and 83.2 MPa, respectively. From the analysis of surface state of the composite plate, the plate surface has an obvious hard layer after the anodic oxidation treatment. On the contrary, the plate surfaces after wire brush grinding and acid-alkali washing do not have hard layer. Owing to the weak work hardening ability of aluminum and magnesium alloys, the wire brush grinding treatment can only eliminate the oxide film on the plate surface. Therefore, hard layer barely exists on the plate surface, and mechanical occlusion cannot be realized at the bonding interface during the rolling bonding process. According to the change in the bonding strength of the composite plate, the removal of hard layer from the plate surface is conducive to the enhancement in bonding strength of the aluminum-magnesium composite plates.

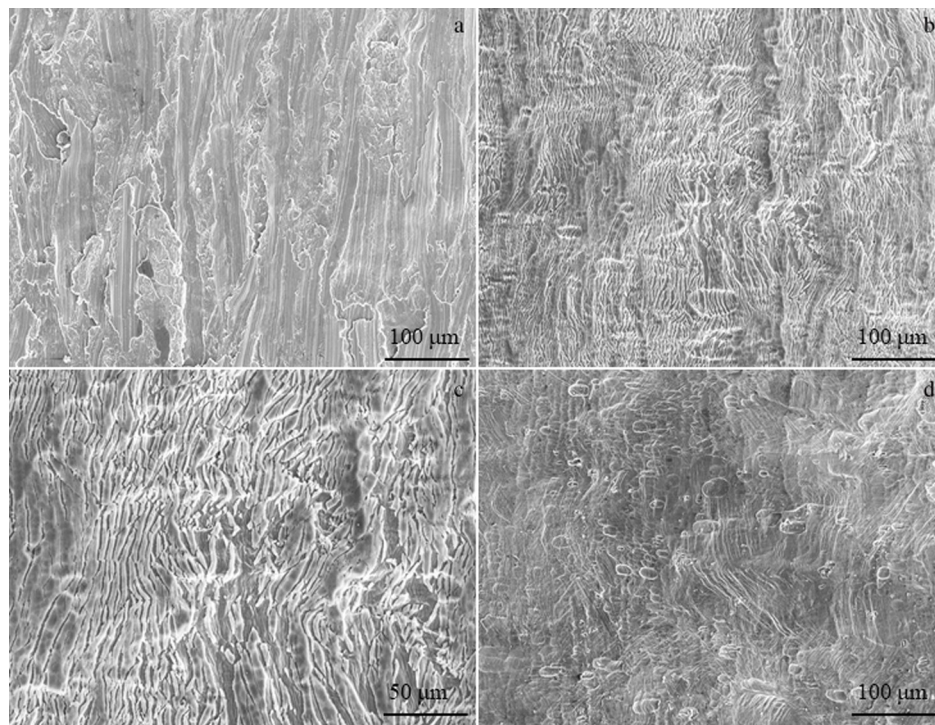


Fig. 1 Tensile surface morphologies of aluminum alloy after different surface treatments: (a) wire brush grinding; (b–c) anodic oxidation; (d) acid-alkali washing

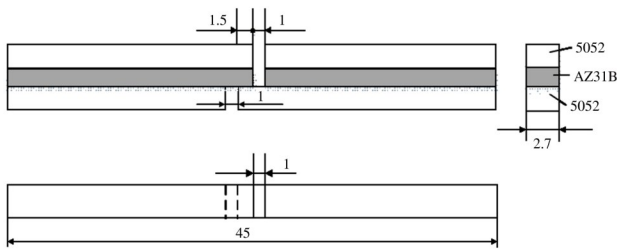


Fig.2 Schematic diagram of tensile-shear specimen

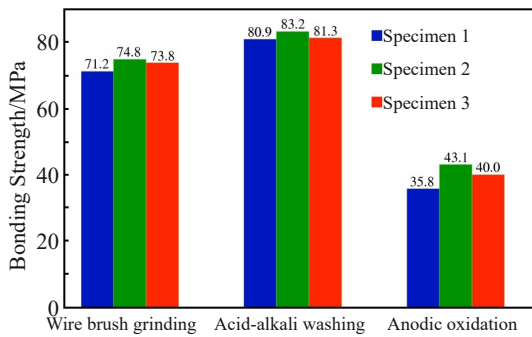


Fig.3 Bonding strength of aluminum-magnesium composite plates after different surface treatments

2.2 Morphology of composite plate

The morphologies of the bonding interface and the tensile-shear fracture of the aluminum-magnesium composite plates after different surface treatments are shown in Fig. 4. The element diffusion at the bonding interface and in the tensile-shear fracture were detected by X-ray spectroscopy (XPS). As shown in Fig. 4b – 4c, the bonding interface curve of the

composite plate is in a discontinuous state after the anodic oxidation treatment, and the hard layer fragments are scattered and embedded at the bonding interface, presenting obvious mechanical occlusion characteristics. However, there is a clear gap between the hard layer fragment and the magnesium side in the hard layer area, which indicates the unbonded state. In other areas, the aluminum side and the magnesium side are closely bonded. After wire brush grinding and acid-alkali washing for the elimination of hard layer, the interface curves of the composite plates remain continuous, and no obvious gaps, holes, or other defects can be found at the bonding interface, as shown in Fig.4a and 4d. The contact between the aluminum side and the magnesium side shows good bonding characteristics. The bonding interface of the composite plate after acid-alkali washing treatment is smoother than that after wire brush grinding, and the contact between the aluminum side and the magnesium side is more adequate. Hence, the bonding strength of the composite plate after acid-alkali washing treatment is slightly higher than that after wire brush grinding treatment.

For further analysis, element line scanning was conducted at the fresh metal bonding area of the composite plate, namely the black lines in Fig. 4, and the results are shown in Fig. 5. Oxygen can barely be detected at the bonding interface of the composite plate after anodic oxidation treatment. By contrast, the oxygen element at the bonding interface of the composite plate after wire brush grinding and acid-alkali washing is more obvious. The element diffusion curves at the bonding interface of the aluminum-magnesium composite panel after different surface treatments are roughly X-shaped, indicating that there is no aluminum-magnesium compound at the bonding interface. Nevertheless, the width of the mutual

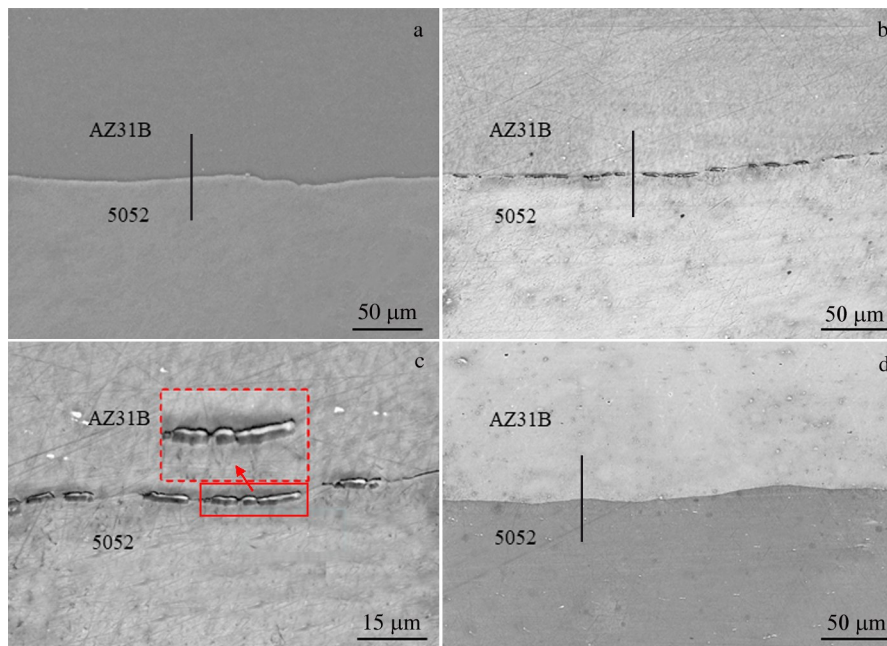


Fig.4 Bonding interface morphologies of aluminum-magnesium composite plates after different surface treatments: (a) wire brush grinding; (b–c) anodic oxidation; (d) acid-alkali washing

diffusion area at the bonding interface of aluminum-magnesium composite plate after anodic oxidation is only 3.8 μm , which is lower than that after wire brush grinding and acid-alkali washing. According to the morphology analysis of the bonding interface of composite plate after different surface treatments, the existence of hard layer on the plate surface will hinder the direct contact between the aluminum alloy and magnesium alloy during the rolling process. The mutual diffusion of aluminum and magnesium mainly occurs in the crack area of the hard layer. Under the conditions of wire brush grinding and acid-alkali washing, the surfaces of aluminum alloy and magnesium alloy can be in direct contact because there is no obvious hard layer on the plate surface. Moreover, the element diffusion occurs at the contact position during the rolling process, so the diffusion degree at the composite plate interface is high without hard layer. Fig. 6 shows the fracture morphologies and corresponding element

distribution maps of aluminum-magnesium composite plate after different surface treatments. It can be seen that the tensile-shear fracture of the composite plate after different surface treatments mainly presents the characteristics of metal on the magnesium side adhering to the aluminum side.

As shown in Fig. 6a, obvious bulging areas and flat areas can be observed on the fracture area at the aluminum side of the composite plate after wire brush grinding. Fig. 6b indicates that the element distributed in the bulging part is mainly magnesium. The bonding of the aluminum-magnesium composite plate after wire brush grinding occurs on the entire contact surface of the aluminum and magnesium alloys. Fig. 6e depicts that there are a large number of blocky protrusions and gaps on the fracture area at aluminum side of the composite plate after anodic oxidation treatment. According to Fig. 6f, the element distributed in the protrusion part is mainly magnesium, indicating that the tear behavior

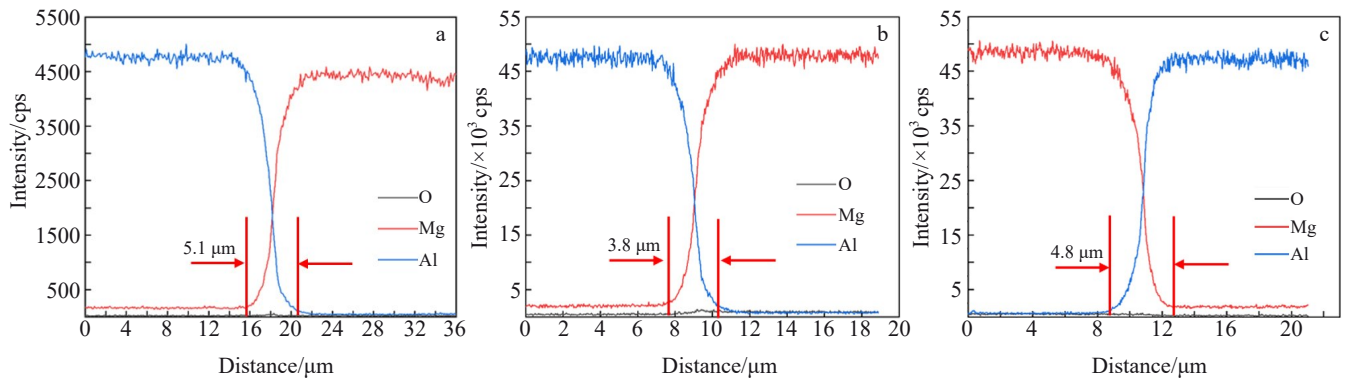


Fig.5 Element line scanning results of fresh metal bonding area of aluminum-magnesium composite plates after different surface treatments: (a) wire brush grinding; (b) anodic oxidation; (c) acid-alkali washing

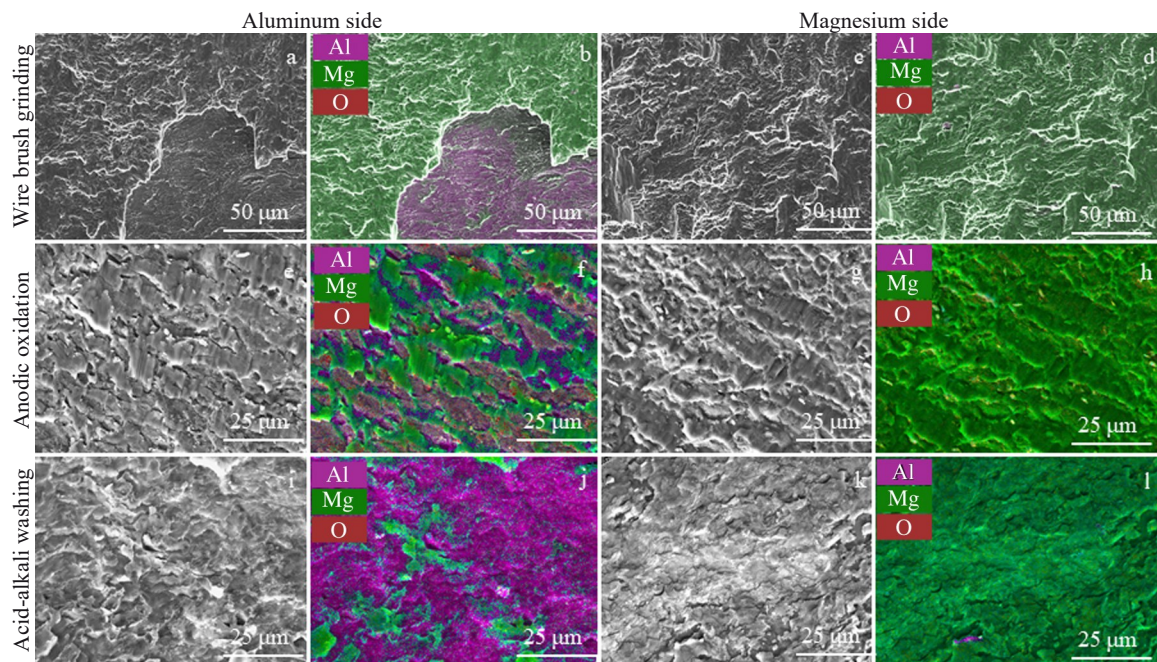


Fig.6 Fracture morphologies (a, c, e, g, i, k) and corresponding element distribution maps (b, d, f, h, j, l) at aluminum side and magnesium side of aluminum-magnesium composite plates after different surface treatments

occurs firstly at the magnesium side, and it gradually extends to the aluminum side in the tensile process. By contrast, the element distributed at the crack is mainly oxygen, implying that the crack consists of hard layer. Because there is no obvious magnesium distribution at the crack, no obvious bonding exists between the hard layer and the magnesium side in this area. The bonding of aluminum-magnesium composite plates after anodic oxidation is assumed to occur mainly in the fresh metal bonding zone, whereas the aluminum and magnesium sides are still unbonded in the hard layer area. As shown in Fig.6i, no large area of protrusions and cracks can be observed on the fracture area at aluminum side of the composite plate after acid-alkali washing treatment. However, Fig.6j shows that the fracture area at aluminum side of the composite plate is covered with sheet-distributed magnesium. The bonding of aluminum-magnesium composite plates after acid-alkali washing treatment is supposed to occur on the entire contact surface of the aluminum and magnesium alloys. Although the existence of hard layer on the plate surface promotes mechanical occlusion of aluminum and magnesium alloys during the rolling process, it also reduces the metallurgical bonding area between the aluminum and magnesium alloys. With the wire brush grinding and acid-alkali washing treatments, the plate surface does not have hard layer, so the contact between the surfaces of aluminum and magnesium alloys occurs directly during the rolling process. Thus, good metallurgical bonding forms at the contact position, and the bonding strength of the composite plate greatly increases.

3 Conclusions

1) Owing to the weak work hardening ability of aluminum and magnesium alloys, the wire brush grinding treatment can only eliminate the oxide film on the plate surface. Hard layer rarely exists on the surface after wire brush grinding, and mechanical occlusion cannot be realized at the bonding interface during the rolling bonding process. The bonding effect depends on the element diffusion, which is generated by close contact between the metals. Once the metals on both sides have good contact in the entire interface, high bonding strength can be achieved.

2) After the anodic oxidation treatment, there is an obvious hard layer on the metal surface, which is broken during the rolling process and forms obvious mechanical bite at the bonding interface. However, the broken hard layer cannot form an effective combination with the metals at the interface, and bonding can only occur in the fresh metal bonding area at the cracks of hard layer. Consequently, the bonding strength of the interface is low after the anodic oxidation treatment.

3) The acid-alkali washing treatment can completely remove the hard layer on the surface of aluminum and

magnesium alloys. Moreover, it will not increase the surface roughness of the plate, compared with the wire brush grinding treatment. Therefore, the metals have better bonding effect in the rolling process. Thus, the acid-alkali washing is a suitable surface treatment method for hot rolling of aluminum and magnesium composites to achieve high bonding strength.

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表面处理方式对铝镁热轧复合板结合强度的影响

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摘要: 在400 °C和45%轧制压下率条件下, 研究了不同表面处理方式对复合板结合强度的影响。结果表明, 钢丝刷打磨处理只能消除板材表面的氧化膜, 很难在板材表面产生硬质层, 复合效果取决于界面两侧金属紧密接触产生的元素扩散。阳极氧化后, 金属表面会有一层硬质层, 轧制过程中破碎的硬质层会在结合界面形成机械咬合。但是, 硬质层无法与界面处的金属形成有效结合, 只能在硬质层裂缝处的新鲜金属结合区发生结合。酸碱洗处理可以完全去除2种合金表面的硬质层, 且不会增加板材的表面粗糙度, 界面两侧的金属在轧制过程中结合更紧密。采用酸碱洗作为铝镁热轧结合的表面处理方法, 可以获得最佳的结合强度。

关键词: 铝镁复合板; 表面处理; 硬质层; 结合强度

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