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The Effect of a Coating Sprayed Using Supersonic Flame Coating Technology on the Mechanical Properties and Interface Structure of a Thick Steel/Aluminum Composite Plate during Hot Rolling

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Abstract: Given the characteristics of a thick steel/aluminum composite plate, such as its large thickness and the significant differences between its components, it is difficult to prepare using direct rolling. Instead, a thick steel/coating/aluminum composite plate may be successfully prepared by combining supersonic flame coating technology with a hot rolling composite process. In this study, the interface shear strength test, SEM, EDS, and other detection methods were applied to investigate the effects of the reduction rate and coating thickness on the interface structure and mechanical properties. The results show that under the condition of single-pass direct rolling, the micro-interface of steel/aluminum is improved with an increase in the reduction rate, but the bonding strength of the interface remains poor. After adding the coating, the thickness of the diffusion layer and the shear strength increase significantly. When the coating thickness is reduced to 0.1 mm, the deformation coordination and shear strength of the composite plate are further enhanced under the combined action of mechanical interlocking and metallurgical bonding. The tensile shear fracture is mainly located at the steel/coating interface. The interfacial shear strength reaches 66 MPa, which exceeds the requirements of the US military standard MIL-J-24445A (SH) for steel/aluminum shear strength. The research results thus support the use of this new method for the simple and efficient production of thick steel/aluminum composite plates.

Keywords: thick steel/coating/aluminum composite plate; supersonic flame coating technology; hot rolling composite process; microstructure; mechanical properties

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1. Introduction

A steel/aluminum composite plate has the advantages of high strength, good wear resistance, the high hardness of steel, and the excellent electrical and thermal conductivity, corrosion resistance, and low density of aluminum. Accordingly, it is widely used in ships, rail transit, aerospace, and other fields [1–3]. The surface of a new ship is mostly made of an aluminum alloy, which is connected with the steel structure of the hull through steel/aluminum composite joints, to achieve a lightweight effect and reduce the center of mass of the ship, to improve its maneuverability and stability. This kind of steel/aluminum composite joint often has thickness specifications [4,5]. A thick steel/aluminum composite plate can also be used as a steel-backed aluminum bearing, steel/aluminum composite guide rail, and so on. Given the great differences in the properties of steel and aluminum, the brazing method and explosive composite method are mainly used in the preparation of a thick steel/aluminum composite plate at this stage [6–8].

The brazing method connects the weldment firmly by heating the low-melting-point solder at the melting temperature, wetting the base metal with the liquid solder, and diffusing it with the base metal. It is necessary to strictly control the brazing temperature when preparing a steel/aluminum-clad plate via the brazing method, which mainly includes three types of welding: laser, electron beam, and arc [9–11]. Liu [12] successfully produced 6 mm 6061-T6/SUS304 butt joints using a laser hybrid metal inert gas (MIG) technique by optimizing the laser power. Yang [13] used a nanosecond laser to ablate the surface of Q355 steel, and they generated grooves with different depths at different processing times. Satisfactory Al/steel joints were formed at different groove depths. The preparation of a thick steel/aluminum composite plate lap joint via the brazing method has great advantages, but for a steel/aluminum composite plate with a large contact area, problems of an incomplete filler metal filling and an uneven brazing strength can easily arise.

The explosive composite method is a composite material processing technology that uses the huge energy generated by an explosive moment to instantaneously combine two or more similar or very different materials [5,14]. At present, thick steel/aluminum composite plates, such as steel/aluminum transition joints for ships, are mainly prepared by using the explosive composite method [15,16]. However, the pollution and noise problems caused by the explosion are serious concerns, placing high requirements on the kind of site that is suitable. Furthermore, this production method needs to be operated in the field, cannot be continuously operated, and involves a low degree of automation, meaning it is not suitable for mass production.

Given its advantages of good process control, low cost, high efficiency, high degree of automation, and continuous batch production, the rolling composite method has gradually come to be the new direction for the composite preparation of steel/aluminum plates [17,18]. Nezhad [19] analyzed the influence of the preheating temperature and reduction rate on the interface bonding quality and strength of steel/aluminum composite plates prepared using the hot rolling process with an initial thickness of 1.9 mm. Chen [20], meanwhile, successfully prepared a steel/aluminum/aluminum alloy composite plate by applying a two-pass isothermal rolling process. During the rolling process, it has been found that the steel and aluminum components can be well compounded when rolling a thin plate, but with an increase in the thicknesses of the components, the deformation and elongation of the steel will decrease, or it will even become difficult to deform. Moreover, the interface bonding strength of the composite plate is also significantly reduced [21,22]. In that context, researchers have made many attempts to improve the bonding strength of thick composite plates. Wang [23], for instance, used the corrugated cold rolling bonding method to prepare a metal composite plate. Thanks to the strong friction shear stress at the interface of the corrugated composite plate, the bonding strength of the Cu/Al composite plate was doubled. Induction heating technology has also been applied for the preparation of thick composite plates [24,25]. According to the difference in deformation resistance, the metal is heated to different temperatures at which rolling is carried out. By improving the coordinated deformation ability between metals, the bonding strength of composite plates is enhanced. In addition, by adding an intermediate transition layer or coating, the formation of brittle and hard intermetallic compounds can be avoided, effectively promoting the creation of a bond between the metals. Thus, the bond strength is improved [26,27]. Huang [21] used plasma coating technology to closely combine a coating layer with the surface of a steel plate, and then they hot rolled this with an aluminum plate to prepare a thick steel/aluminum composite plate with a total thickness of 13.5 mm.

Further to this, supersonic flame coating technology is a kind of material surface modification and surface strengthening technology where a metal powder is heated until there are molten or semi-molten particles, and the substrate surface is impacted with a supersonic flame beam of more than 1500 m/s to form a coating [28,29]. This paper attempts to combine the supersonic flame coating technology with the hot rolling composite process. By pre-spraying a nickel–chromium layer on the surface of a steel plate, the coating layer is firmly bonded to the surface of the plate. Through hot rolling with an aluminum plate, the steel/aluminum composite is transformed into a combination of steel/coating and aluminum/coating. In doing so, the preparation method of composite plates proposed in this paper can reduce the difficulty of the rolling process and provide a new way of efficiently preparing thick steel/aluminum composite plates.

2. Experimental and Analytical Methods

2.1. Scheme for the Rolling Experiment

The component materials consisted of Q235 steel and 1060 aluminum, the compositions of which are indicated in Tables 1 and 2, respectively. A layer of 80Ni20Cr coating was sprayed onto the steel plate by using supersonic flame coating technology. The spraying thicknesses were 0.1 and 0.3 mm. The dimensions of the Q235 steel and 1060 aluminum plates were 100 mm × 60 mm, and the initial thickness was 4 mm. The rolling composite experiments were carried out on a two-high mill, and the experimental process is shown in Figure 1.

Table 1. Main compositions of Q235 steel plate (wt. %).

Elements	C	Mn	S	P	Si	Fe
Q235	0.15	0.5	0.045	0.02	0.1	balance

Table 2. Main compositions of 1060 aluminum alloy plate (wt. %).

Elements	Si	Cu	Mn	Mg	Zn	V	Ti	Al
1060	0.25	0.05	0.03	0.03	0.05	0.05	0.03	balance

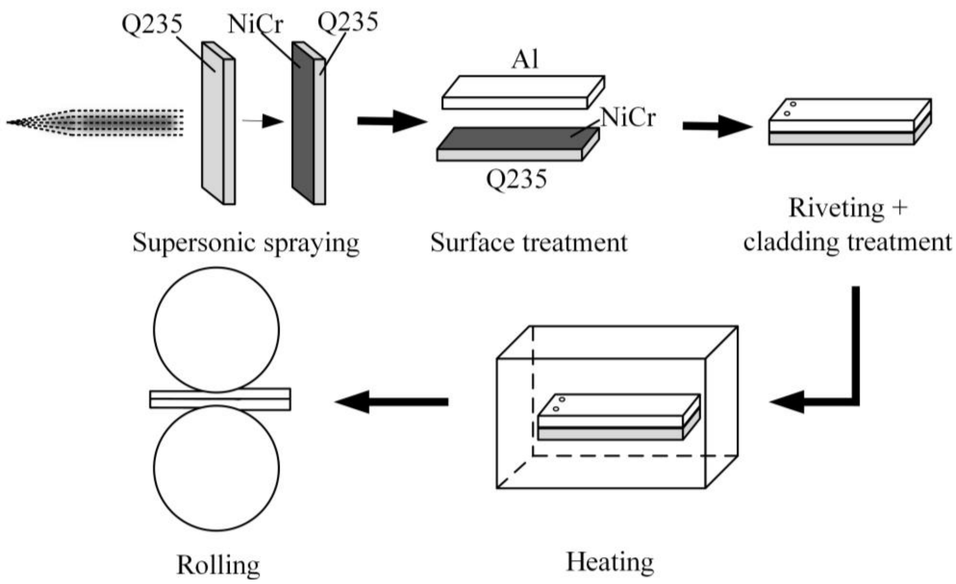

Figure 1. Steel/coating/aluminum composite plate rolling process diagram.

(1) Spray coating. The surface of the steel plate is sandblasted before spraying to remove the oxide layer from the surface of the steel plate and increase the surface roughness. Subsequently, the Ni–Cr coating is sprayed onto the surface of the steel plate by using supersonic flame spraying technology. The specific operation is as follows: first, oxygen is used as a combustion-supporting gas, and acetylene, kerosene, and other fuels burn violently and expand in the combustion chamber. A supersonic flame beam with a velocity of more than 1500 m/s is formed by Laval nozzle compression. At the same time, the sprayed powder is fed into the flame to produce molten or

semi-molten particles, which hit the surface of the steel plate at high speeds to form a Ni–Cr alloy coating.

(2) Surface treatment. The surfaces of each component metal plate are treated by removing the grease, dirt, and oxides and texturing and cleaning the bonding surface.
(3) Anti-oxidation treatment. The polished slab is wiped and cleaned with alcohol, dried, and then riveted. The riveted composite plate is wrapped in 0.02 mm aluminum foil to prevent oxidation.
(4) Preheating the plates. The front ends of component plates are riveted to ensure the stabilization of the bite stage. In addition, the component plates are covered with 0.2 mm aluminum foil and preheated at 400 °C for 15 min under a protective atmosphere.
(5) Rolling compound. A plate is taken out and sent to a two-roll mill for rolling. The roll diameter is 200 mm and the roll speed is 60 mm/s. By adjusting the size of the roll gap, rolling reduction rates of 5%, 15%, 25%, 35%, and 45% for a single pass of a steel/aluminum composite plate are achieved.

2.2. Sample Preparation and Material Characterization

For the accurate analysis of the deformation process, samples of the rolling deformation zone were acquired through an emergency stop. The bonding strength of the prepared steel/aluminum and steel/coating/Al composite plates was measured by applying a shear test, administered using an Instron 5848 tensile tester. The test was carried out at room temperature with a speed of 0.5 mm/min. To reduce the experimental error, three specimens were used for the shear test and the average number was taken as the interface bonding strength of the composite plate. The size of the shear specimen is shown in Figure 2. The model of the scanning electron microscope with SEM and EDS functions used in the experiment is FEI Scois DualBeam. A scanning electron microscope (SEM) was applied to observe the micro-morphology of the bonding interface along the rolling direction. Furthermore, the bonding interface and fracture of the sample were also observed using energy dispersive spectroscopy (EDS).

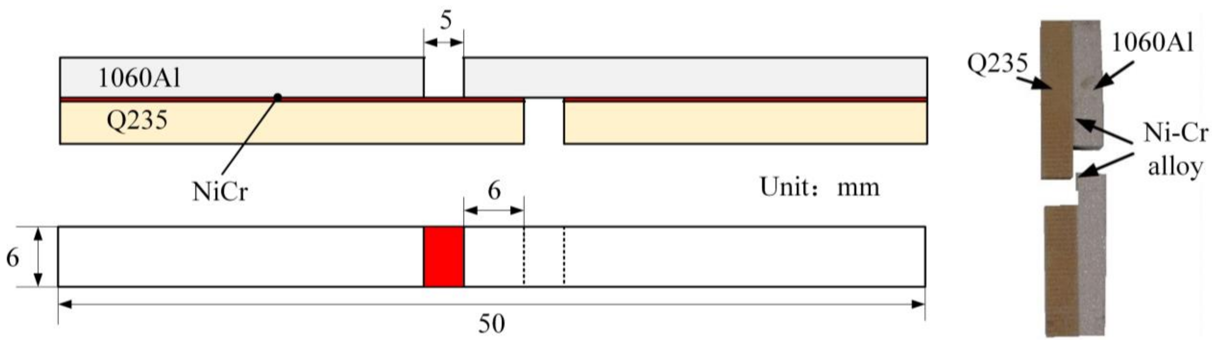

Figure 2. The shear specimen size of the composite plate.

3. Results and Discussion

3.1. Analysis of the Warm Rolling Experimental Results for a Thick Steel/Aluminum Composite Plate

A rolling experiment of a steel and aluminum plate with an initial thickness of 4 mm, rolling temperature of 400 °C, and reduction rate of 45% was carried out. It can be seen from Figure 3a that there was no macroscopic deformation or thinning of the steel plate. The deformation was mainly concentrated on the aluminum metal, and the deformation coordination of the composite plate was poor. Figure 3c gives a metallographic photograph of the interface of the steel/aluminum composite plate under different reduction rates in the deformation zone. When the reduction rate is 15%, a large number of point and strip component metals appear at the interface, and the gap between the components is obvious. When the reduction rate is 25%, the discrete component metals at the interface decrease and the particles become smaller. When the reduction rate is 35%, the metal components at the interface largely disappear, but the gap at the interface is obvious and there is no mechanical occlusion. When the reduction rate increases to 45%, there is relatively close bonding at the interface, and some positions appear to be embedded in one another, but the overall interface composite effect is poor.

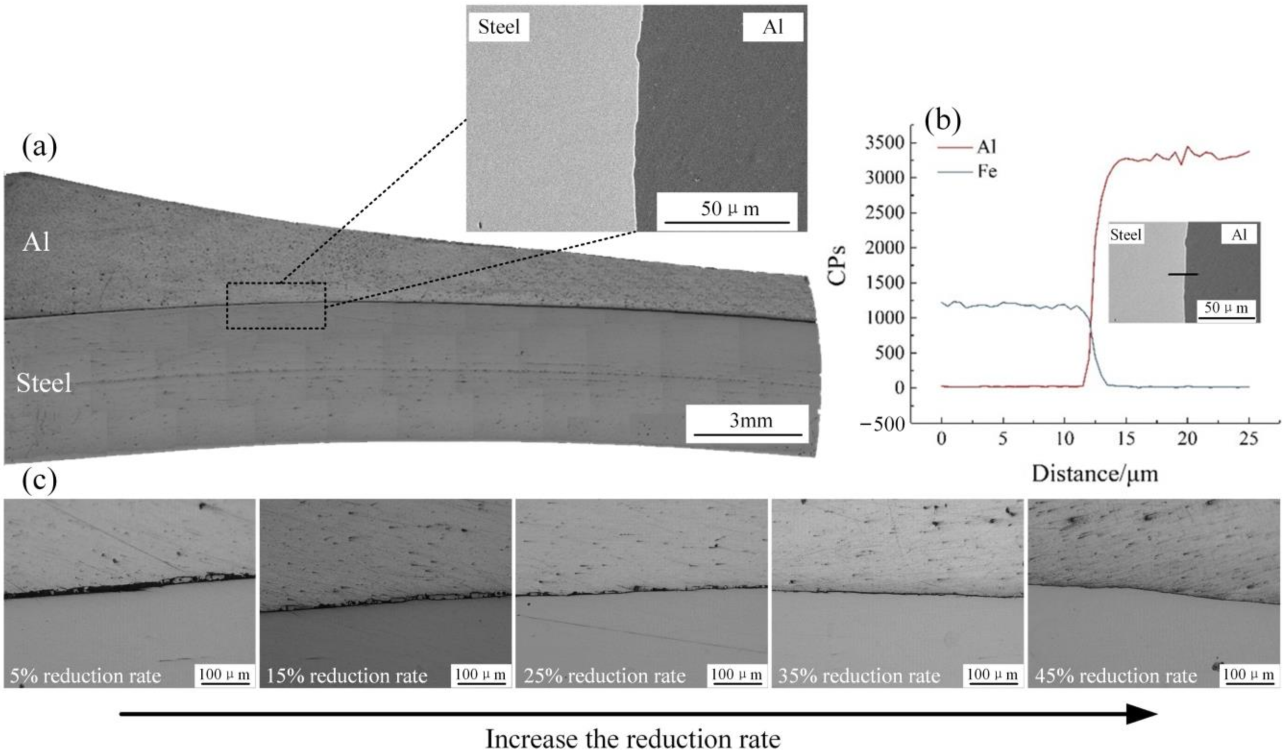

Figure 3. Experimental results of hot rolling of steel/aluminum composite plate: (**a**) metallographic photos of deformation zone of steel/aluminum composite plate at 45% reduction rate; (**b**) EDS of steel/aluminum composite plate interface at 45% rolling reduction rate; (**c**) metallography of deformation zone of composite plate under different reduction rates.

When the reduction rate is 45%, the gap at the interface of the two metals largely disappears, but the composite effect of the interface is poor. The reason for this phenomenon is the deformation coordination of the two metals. According to the mechanical meshing theory and film theory, the generation of fresh metal at the interface during the rolling process of a steel/aluminum composite plate plays a crucial role in the bonding strength of the interface. In terms of the macroscopic morphology, the steel plate has not been deformed or thinned, which limits the generation of cracks on the steel side of the interface and the extrusion of fresh metal. Figure 3b shows the SEM morphology and EDS line scanning results of the interface of the steel/aluminum composite plate. At this time, an element diffusion area of about 2 μm is generated at the interface of the steel/aluminum composite plate. Figure 4 presents the results for the evolution of the shear strength of the composite plate at different rolling reduction rates. It can be seen from the figure that the shear strength of the interface increases gradually with the increase in the rolling reduction rate. However, the interfacial shear strength of the composite plate at a 45% reduction rate is only about 21 MPa, which is lower than the 55 MPa steel–aluminum shear strength standard of the American military standard MIL-J-24445A (SH) [30].

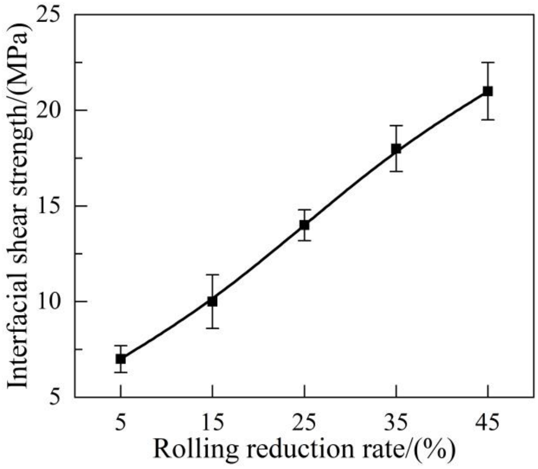

Figure 4. The influence of the rolling reduction rate on the interfacial shear strength of the steel/aluminum composite plate.

3.2. The Effect of a Coating on the Bonding Strength of the Steel/Aluminum Composite Plate

Since a steel plate is challenging to deform and the interface cannot be effectively compounded during the conventional rolling process of a thick steel/aluminum composite plate, a method of supersonic flame coating + hot rolling is proposed to prepare a thick steel/aluminum composite plate. Figure 5 gives the SEM and EDS results for the steel/coating interface in the initial unrolled state. From the SEM images, it can be seen that when the Ni–Cr coating is sprayed on the steel surface, the bonding surface in some parts shows an obvious occlusion phenomenon, and there is a tightly bonded area, but in other places, there are cracks and pores at the coating/steel interface and in the coating. This is an inevitable phenomenon when applying supersonic flame coating technology, which occurs when the Ni–Cr powder with a higher temperature is sprayed on the colder steel plate. Solidification occurs quickly and the fluidity is poor, so it cannot fill all of the tiny depressions on the surface of the substrate. However, it can be seen from the diagram that the defects are relatively small and can be reduced by adding a subsequent rolling process. EDS line scanning was carried out on the well-bonded area. Through the line scanning analysis, we found that the Ni, Cr, and Fe elements showed transitional changes at the interface, and the thickness of the diffusion layer was about 1.8 μm. This indicates that at the interface, mutual diffusion occurred between the steel substrate and the Ni–Cr coating elements, and that the bonding modes were mechanical and metallurgical.

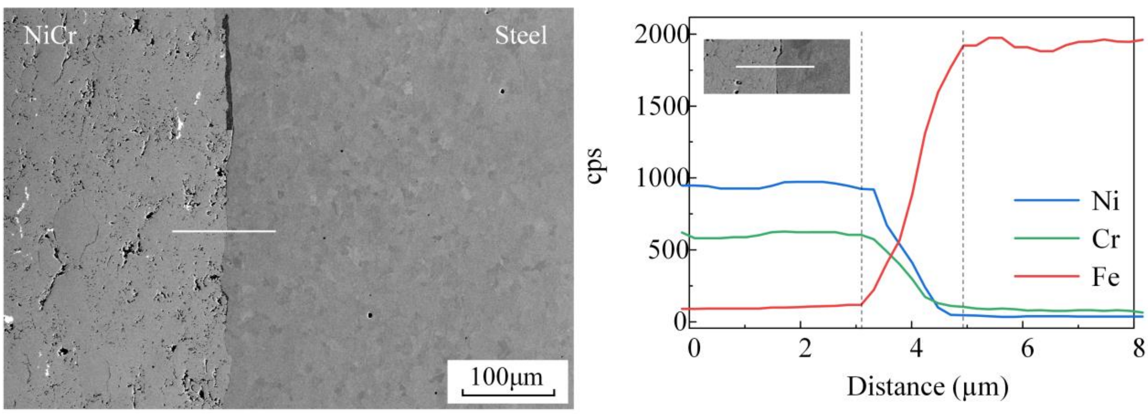

Figure 5. SEM and EDS of the initial interface of steel/coating.

Figure 6a,b show the SEM morphology of the steel/coating/aluminum composite plate interface after rolling at a 45% reduction rate when the coating thickness was 0.1 mm.

It can be seen from Figure 6b that the thickness of the Ni–Cr coating after rolling was about 0.078 mm, the coating had barely any extension, and a brittle fracture formed with a block morphology. On the one hand, this shows that during the rolling process, the steel plate was subjected to rolling action to produce surface expansion. Unlike steel with ductility, the coating sprayed on the steel produced cracks due to its brittle characteristics. As the rolling proceeded, the coating broke under large deformation, and the cracks perpendicular to the rolling direction increased and gradually widened. During the rolling process, the fresh metal on the surface of the aluminum side was squeezed into the cracks between the steel/coating, forming mechanical interlocking. Under the action of a continuous increase in rolling pressure, the plastic deformation of the metal was further increased. A large amount of fresh metal produced by steel and aluminum plates was squeezed out into the open space formed by the fracture of the brittle coating, and full contact was achieved through strong contact pressure. Atomic bonding occurred at the contact interface of the composite plates, so metallurgical bonding was established. On the other hand, it can be seen that the aluminum/coating interface was tightly bonded, and the metals were embedded in each other. The gap at the coating/steel interface was also reduced under the action of rolling force, and the interface became more closely bonded. Under the combined action of these two processes, the bonding strength of the composite plate was greatly improved and the internal defects of the coating were eliminated.

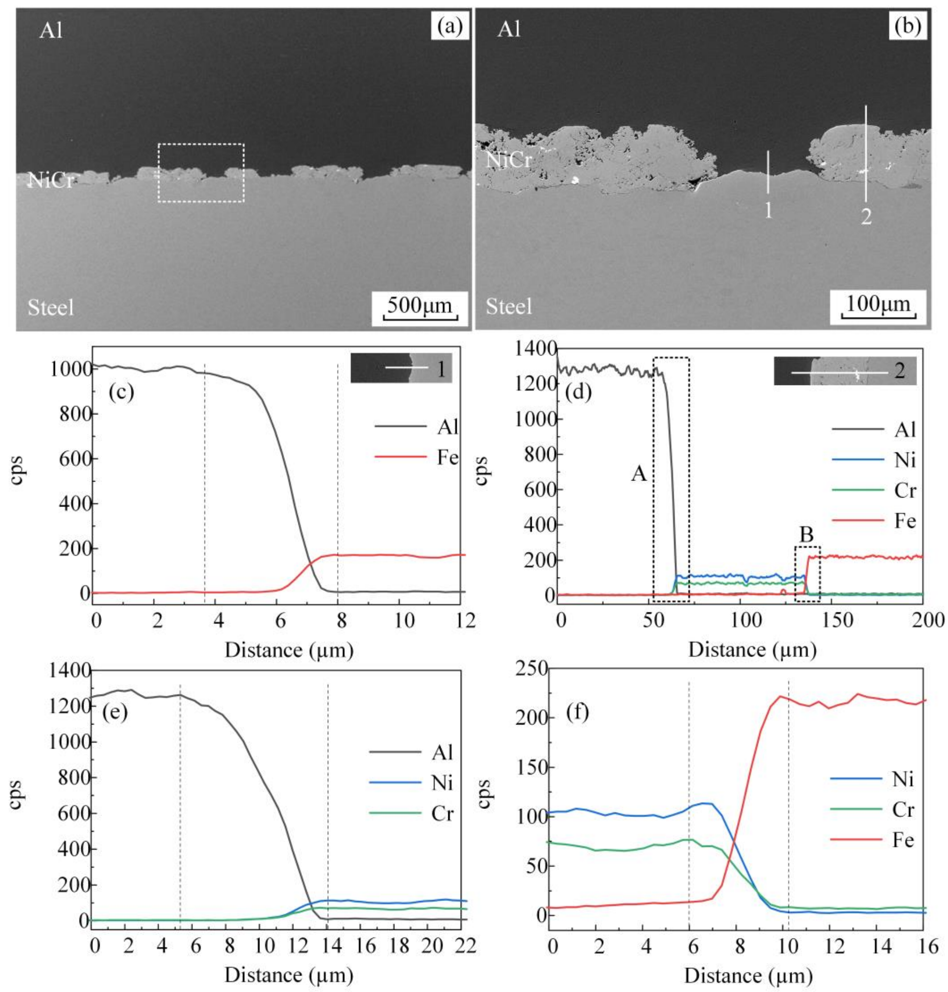

Figure 6. SEM morphology and EDS scanning results of steel/coating/aluminum composite plate interface when coating thickness is 0.1 mm: (**a**) SEM morphology of composite plate interface at 45% reduction rate; (**b**) local enlargement of the morphology in the white frame; (**c**) EDS results of region 1; (**d**) EDS results of region 2; (**e**) amplified diagram of zone A; (**f**) amplified diagram of region B.

An EDS line scan was performed on regions 1 and 2 in Figure 6b, and the results are shown in Figure 6c,d, respectively. The results show a distribution of transitional elements at the interface of the composite plate, resulting in metallurgical bonding. Element diffusion regions of 4.3 µm, 8.8 µm, and 4.2 µm were generated at the steel/aluminum contact interface, the aluminum/coating interface, and the steel/coating interface, respectively. Compared to the unrolled coating/steel interface (see Figure 5), the thickness of the diffusion layer increased by about 130%, indicating that the element further diffused after the hot rolling experiment at 400 °C.

Figure 7 shows the shear fracture of the steel/coating/aluminum composite plate when the coating thickness was 0.1 mm. According to the results of EDS surface scanning, there were a large number of Al and Fe elements in the fracture depression on the steel side, corresponding to the direct contact area of steel and aluminum metal at the interface, and there were greater Ni and Cr contents in the convex area. The fracture on the aluminum side contained many deep dimples, with high Al contents, while there were greater Ni and Cr contents in other areas. It can be seen that there were many direct contact areas between steel and aluminum during this process, and that the coating and aluminum were strongly bonded, with the fracture mainly forming at the steel/coating interface. However, there was good metallurgical bonding between the coating and the steel substrate, so the bonding strength of the composite plate was greatly improved. The thinness of the coating also reduced its internal defects, which greatly improved the overall bonding strength of the composite plate. The dimples of the fracture were deep and had obvious ductile fracture characteristics.

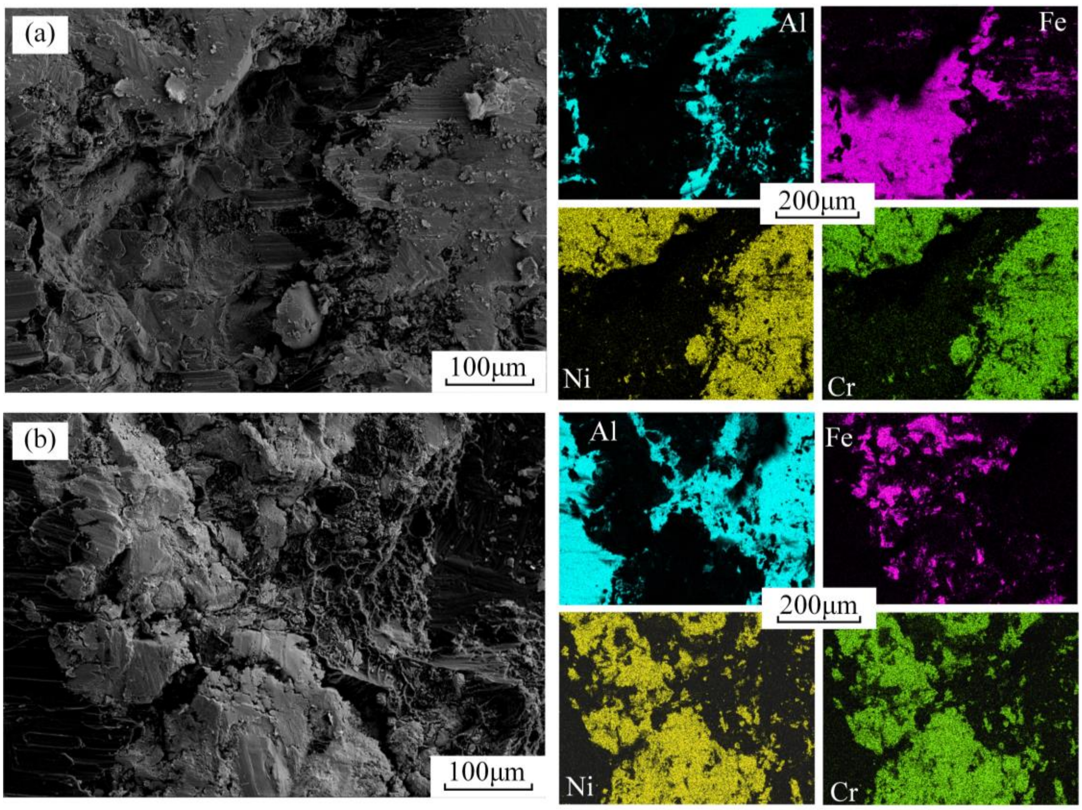

Figure 7. The fracture of the steel/coating/aluminum composite plate with a coating thickness of 0.1 mm: (**a**) the fracture of the steel side; (**b**) the fracture of the Al side.

Figure 8a,b illustrate the SEM morphology of the steel/coating/aluminum composite plate interface after rolling at a 45% reduction rate when the coating thickness was 0.3 mm. It can be seen from Figure 8b that the thickness of the Ni–Cr coating after rolling was about 0.267 mm and the fresh metal on the aluminum side of certain cracks was not completely squeezed in. Compared with Figure 6b, it can be seen that with the increase in spraying thickness, the pores and cracks inside the coating also increased. At the same reduction rate, when the coating thickness was 0.3 mm, the direct contact area of steel and aluminum and the area where the coating broke into blocks both shrank, which reduced the mechanical occlusion area, so the interface bonding strength lowered.

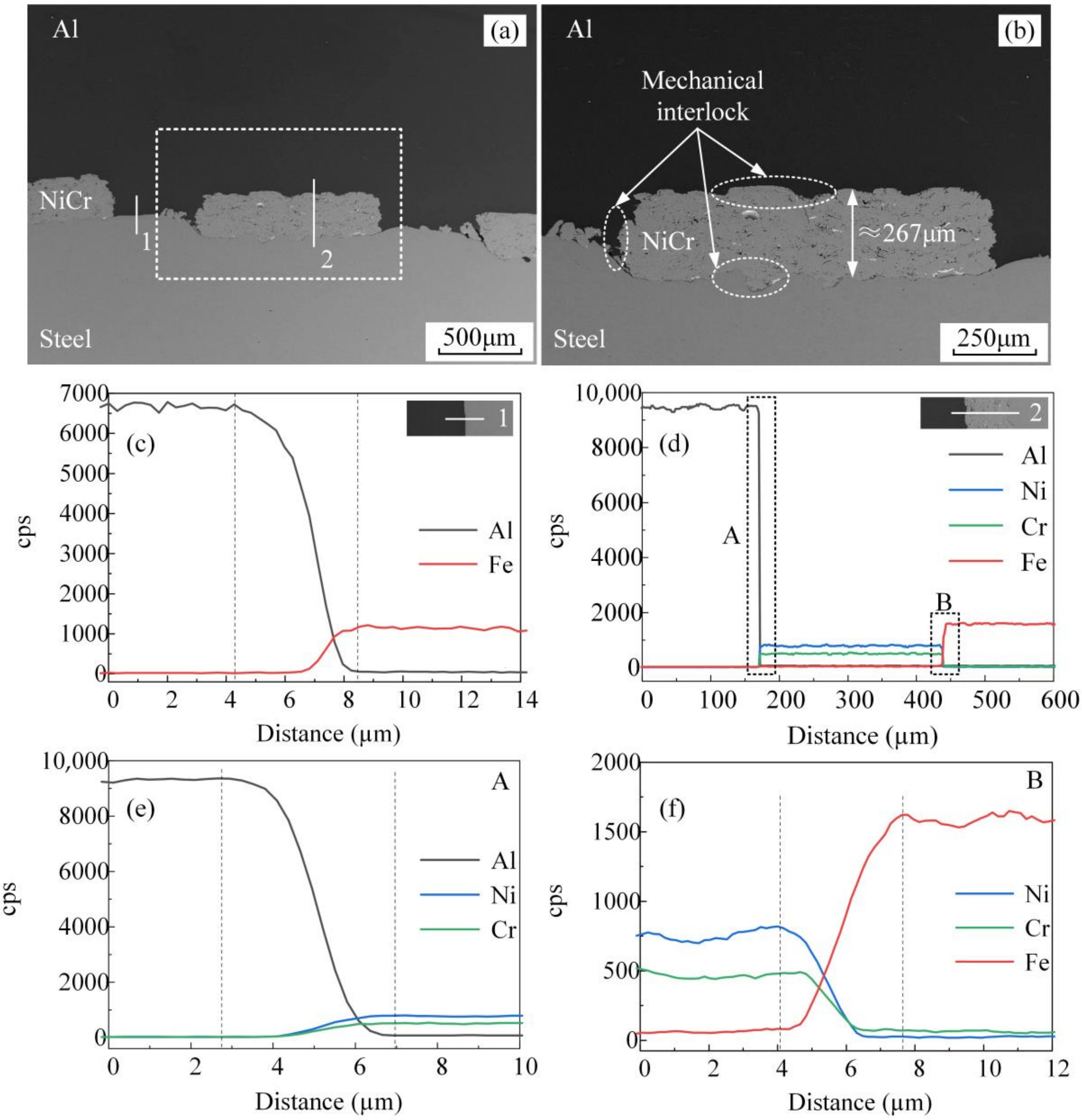

Figure 8. SEM morphology and EDS scanning results of steel/coating/aluminum composite plate interface when coating thickness is 0.3 mm: (**a**) SEM morphology of steel/coating/aluminum composite plate interface at 45% reduction rate; (**b**) local enlargement of morphology in white frame; (**c**) EDS results of region 1; (**d**) EDS results of region 2; (**e**) amplified diagram of zone A; (**f**) amplified diagram of region B.

The EDS results for the interface of the steel/coating/aluminum composite plate after rolling are shown in Figure 8c–f. Element diffusion regions of 4.2 μm, 4.2 μm, and 3.6 μm were generated at the steel/aluminum contact interface, the aluminum/coating interface, and the steel/coating interface, respectively. Compared to Figure 6c, the thickness of the diffusion layer in the direct contact area of steel and aluminum was less affected by the thickness of the coating. However, with the increase in the coating thickness, the thickness of the diffusion layer at the aluminum/coating interface and the coating/steel interface decreased, and the thickness of the aluminum/coating decreased significantly.

Comparative results of the shear strengths of the composite plates with different coating thicknesses and different thinning amounts of aluminum and steel at the outlet are shown in Figure 9. When the coating was not added, the shear strength of the composite plate was low, at only 21 MPa, and the ratio of the thinning amount of steel to aluminum was about 1:3, showing serious deformation incompatibility and poor interface bonding strength. After spraying a coating with a thickness of 0.1 mm, the shear strength of the composite plate was about 213% higher than that before adding the coating, reaching 65.8 MPa. The thinning ratio of steel to aluminum was also reduced to less than 1:1.5, achieving a good bonding effect. However, when the coating thickness was 0.3 mm, the shear strength of the composite plate decreased by about 17%, and the deformation coordination of steel and aluminum decreased. Therefore, we surmise that a thin coating is more helpful to increase the bonding strength of the composite plate.

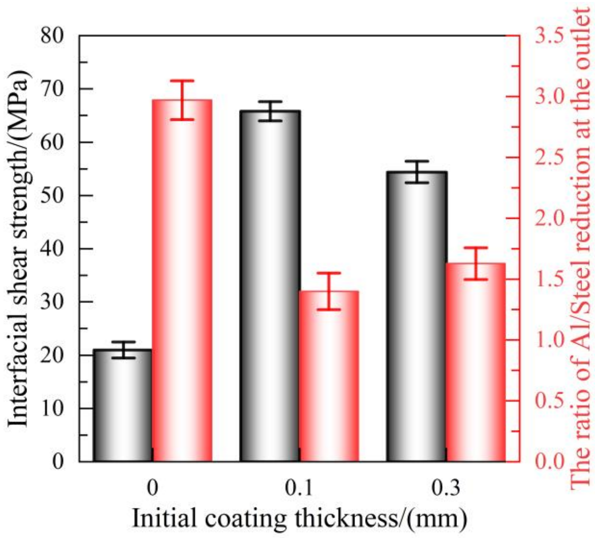

Figure 9. Comparison results of shear strength and thinning of composite plates with different coating thicknesses.

3.3. Bonding Mechanism of the Steel/Coating/Aluminum-Clad Plate during Rolling

The hot rolling composite method involves heating the sheet to be composited, and then the fresh metal produced by the interface rupture is brought in close contact with the huge rolling force of the rolling mill, resulting in an atomic reaction, so that a certain strength of metallurgical bonding forms at the interface [31]. The high temperature in the rolling process of the hot rolling composite method will lead to a phase transformation of the metal, a change in its microstructure, and the formation of brittle compounds between metals, such as $FeAl_2$, $FeAl_3$, etc. These brittle compounds are the main factors that cause a decrease in bonding performance [32,33]. To resolve the problems above, the method of inserting an intermediate layer or coating between steel and aluminum is usually adopted [34]. As a kind of nickel-based superalloy, the 80Ni20Cr coating is known for its good bonding with the matrix material. Arbo [35] found that the bonding strength of a steel/aluminum composite plate with a nickel interlayer was improved after a rolling heat treatment. The Ni–Cr coating also had good high-temperature oxidation resistance, which meant that adverse effects of the oxide layer on the bonding strength of the steel/aluminum composite plate could be avoided [36,37]. However, there was a significant decrease in the plasticity of the Ni–Cr coating in the medium temperature range. Therefore, the Ni–Cr coating exhibited brittle characteristics at 400 °C. The bonding mechanism of the steel/coating/aluminum composite plate during the rolling process is shown in Figure 10. In the early stage of rolling, the composite plate produces plastic deformation. Then, during the rolling process, as the steel extends along the rolling direction, the brittle Ni–Cr coating on the steel plate breaks and extends in the form of cracks. With the large reduction in its thickness, the fresh metal on the aluminum side is squeezed into the cracks under the action of the rolling force, resulting in some mechanical occlusion. Following this, with a further increase in plastic deformation, the cracks of the coating continue to expand, and the fresh metals produced by steel and aluminum make direct contact in the coating gap. When the rolling process is complete, the coating is broken and has formed a block shape, and the interface between the coating and the steel and aluminum is more closely bonded, with mechanical interlocking having formed in many places. Subsequently, the fresh metals of steel and aluminum come in full contact, and the direct contact area is further expanded, forming strong metallurgical bonding, which greatly improves the bonding strength of the whole composite plate. Moreover, when the coating thickness is appropriately reduced, the internal defects in the coating are lessened, resulting in a further increase in the overall bonding strength of the composite plate.

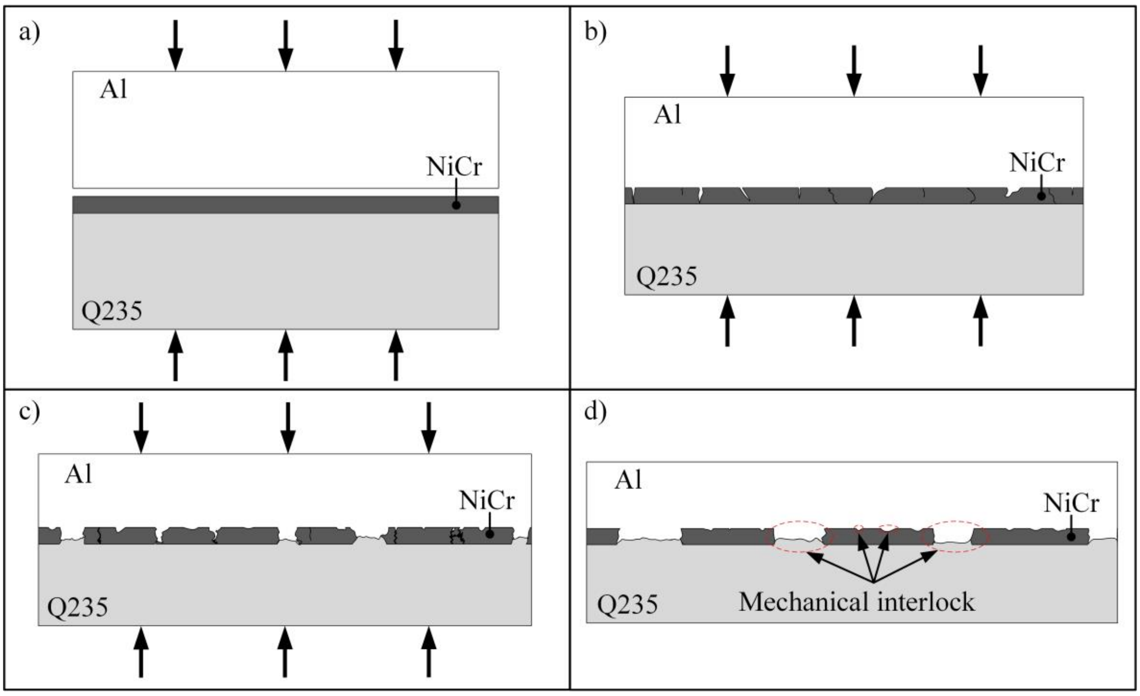

Figure 10. A schematic diagram of the bonding mechanism of the steel/coating/aluminum rolled composite plate: (**a**) the initial stage of rolling; (**b**) the coating began to break; (**c**) coating fracture and fresh interface exposure; (**d**) metallurgical bonding and occlusion strengthening.

4. Conclusions

1. Due to the poor deformation coordination in the hot rolling of an uncoated steel/aluminum composite plate, the bonding strength of the composite plate was low and the diffusion layer only measured at 2 μm. However, a thick steel/coating/aluminum composite plate was prepared via supersonic flame coating and a hot rolling composite method, where the bonding strength of the composite plate was significantly enhanced and the diffusion layer increased to more than 3.6 μm.

2. The Ni–Cr coating was gradually broken into a block shape during the rolling process. The coating and the steel and aluminum metal interface became more closely bonded, and mechanical interlocking and metallurgical bonding formed in many places, thus effectively improving the bonding strength of the composite plate.

3. By reducing the thickness of the coating, the defects were further reduced and the bonding strength of the composite plate was improved. At the same time, the thickness of the element diffusion layer at the interface increased, especially on the aluminum side. The

coating became more closely combined with aluminum, and the fracture mainly occurred at the steel/coating interface. At the same time, the block fracture area of the coating expanded, and the mechanical interlocking area increased. The shear strength of the 0.1 mm thin-coating composite plate reached 66 MPa, exceeding the requirements of the US military standard MIL-J24445A (SH) for steel/aluminum shear strength. Nonetheless, it should be noted that due to the influence of multiple factors such as the coating type, coating process, and rolling schedule, the characteristics of the steel/aluminum composite interface will change. To determine the scope of these changes, we will conduct a detailed process analysis in the future.

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Data Availability Statement: The data presented in this study are available on request from the corresponding author. The data are not publicly available due to privacy.

Conflicts of Interest: The authors declare no conflict of interest.

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