Effect of different reduction rates on the near-interfacial structure of pressed 304/Q235 composite plate

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Abstract

The 304 stainless steel/Q235 composite plate was pressed at 1200 °C using a 500-ton hydraulic testing machine at a reduction rate of 25%, 30%, 35%, and 40% respectively. The microstructure of the composite plate was analyzed by means of scanning electron microscopy (SEM) and electron backscatter diffraction (EBSD). The results showed that the black matter at the interface was the oxide of Mn and Si, and during the compression deformation process, the partial oxide film and the linear inclusions were crushed and extruded into fine particles. Small grains of different sizes appeared on the composite interface, and this deformation made the Q235 carbon steel and the 304 stainless steel on both sides of the composite interface show a coordinated deformation tendency when the reduction rate reached 35%. The whole process showed that the deformed carbon steel was first deformed, and the hard-to-deform stainless steel began to be deformed at a certain point, and then they were both further deformed together.

1. Introduction

Many manufacturing techniques have been widely used for metal joining of stainless steel clad plates [1], such as explosive cladding, hot roll bonding [2, 3], diffusion bonding, cumulative roll welding (ARB), thermocompression bonding [4, 5], etc. All of these stainless steel bonding technologies rely on fast solid-state bonds under heat and plastic deformation to achieve metal connection. Therefore, the quality of the bonding interface is a paramount issue in metal joining. Various study has been conducted on microstructure characteristics related to interface formation and corresponding bonding properties [6–8].

There is a distinct diffusion zone of alloying elements around the bonding interface of the stainless steel composite plate. The diffusion behavior of carbon leads to the formation of decarburization layer and carburized layer, which seriously affects the mechanical properties and corrosion resistance of stainless steel composite plates, while the diffusion behavior of other alloying elements (iron, nickel, and chromium) plays an important role in enhancing the toughness of and strengthening the composite interface [9, 10]. The bonding state and bonding strength of the interface are the main factors for judging the quality of stainless steel composite plates. The bonding strength and toughness of the interface determine its practicability in the subsequent processes of cutting, forging, rolling, bending, welding and stretch forming [11–13]. The interface bonding strength of stainless steel composite plates mainly depends on the rolling process parameters, such as vacuum degree, rolling temperature, reduction rate, inter-layer thickness, etc [14–16].

At present, some scholars at home and abroad have focused on the joint strength and mechanical properties of stainless steel composite plates by hot-rolled bonds. So far, studies on the microstructure of the composite bonding interface in the hot pressing process are rare. In this paper, the interface microstructure of the pressed 304/Q235 composite plate was analyzed, and the effects of reduction rates on the defects and recrystallization at the composite bonding interface was investigated.
2. Experimental procedure

2.1. Fabrication of pressed 304/Q235 composite plate

In this experiment, a double-layer 304 stainless steel/Q235 composite plate was pressed, with Q235 low carbon steel as the base layer and 304 stainless steel as the composite layer. The size of the base layer blank is 200 mm × 100 mm × 15 mm, and the size of the composite layer blank is 200 mm × 100 mm × 5 mm, and the chemical compositions of the 304 stainless steel and Q235 is shown in table 1.

Before the thermocompression bonding, a hand-held electric wheel grinder with a stainless steel wire brush was used to grind the steel plate surface, removing the foreign objects and oxides on the surface. Then, the composite plate was sequentially subject to surface treatment with acid-base washing, and the impurities such as oil and adsorbate were removed by wiping them with alcohol. Subsequently, assembly was performed, a shallow hole with a diameter of 5 mm was drilled in the Q235 low carbon steel surface for bonding, and a small hole with a diameter of 5 mm was drilled on the adjacent side to make them communicate. After the metal chips were cleaned, argon arc welding was performed for encapsulation welding, and a seamless steel pipe with an external diameter of 8 mm was welded at the small hole to create vacuum. For the composite panel, connect the seamless tube, vacuum the vacuum pump to 10⁻³ Pa, and perform heat seal for the seamless tube. The resulting composite panel is as shown in figure 1(a).

The mold and pressing pad required for pressing at different reduction rates were designed. The 304/Q235 composite slab was preheated to 1200 °C for 60 min in an electric furnace, then pressed with the mold and pressing pad in a 500-ton hydraulic testing machine of the laboratory model THP01-500 (figure 1(b)), under a reduction rate of 25%, 30%, 35%, and 40% respectively. In order to make the results more informative, three composite plates were pressed for each reduction, and composite slabs with different process parameters were recorded separately. After being pressed, the samples were cooled to room temperature. The specific slab size and press process parameters are shown in the table 2.

2.2. Microstructure analysis

In order to study the microstructure, the 304 stainless steel/Q235 composite plate after rolling at different reduction rates was cut in the rolling direction into a sample with dimensions of 10 × 8 mm by the high-speed electric discharge wire-cutting machine, (figure 1(c)). In order to facilitate the observation of the bonding interface, the sample was subject to rough grinding, fine grinding and polishing. Then bonding interface was etched with a 4% nitric acid solution.

The properties of materials depend on the internal structure, and the different crystal orientation distribution will also reflect the final properties of materials. In recent years, EBSD technology has been widely...
used in the characterization of material microstructure. This technique overcomes the problems of small analysis area and poor statistics of TEM. Finally, the ZIESS SIGMA FE-SEM equipped with energy dispersive spectroscopy (EDS) and EBSD was used to analyze the microstructure of the composite interface.

3. Results and discussion

Through observation and comparison, it was seen that when the reduction rate was 25%, there were continuous dense point-like black particles on the 304 stainless steel side near the interface (figure 2). At the same time, a large amount of oxygen was found at the interface. The reason is that even if vacuum treatment was performed, a large number of oxygen atoms were adsorbed to the surface to be bonded due to its roughness. Besides, Fe, Mn, and Si elements were abundant at the interface. Fe was an element inherent in the base cladding metal. The elements of Mn and Si were spread from the 304 stainless steel and Q235 around the interface. The analysis showed that the black matter at the interface was an oxide of Mn and Si, Si and Mn easily combine with the residual O of the composite interface to form an oxide at high temperature. Li et al. [17] concluded that the selective oxidation of Al, Si, Mn, and Cr elements from the slab plate to the cladding interface resulted in oxide inclusions at the interface. Nomura et al. [18] and Wang et al. [19] reported that the cladding surface oxide inclusions were high-vacuum-degree SiO₂, MnSiO₃ and MnSiO₄. According to the internal oxidation theory, Si has the strongest affinity, which first interacts with O to form SiO₂, and then SiO₂ interacts with Mn to form MnSiO₃ and MnSiO₄. Zhu et al. [14] found that the interface oxide changed from spherical and rod-like shapes into blocks and irregular linear shapes. Nomura [18] found that Si–Mn oxides were easily formed on the steel surface containing Mn and Si, which Nomura believed was related to the sensitivity of Si and Mn to oxygen. Peng [20] reported that in the rolling process, surface oxide milling could promote strong metallurgical bonding between the two bonding surfaces. The finer the oxide, the better the bonding of the interface. Chen Jing [21] found out that there was a black strip-like inclusion of about 5 µm at the interface of the 25Cr5MoA/Q235 steel composite plate, and the formation of the inclusion was related to oxidation and diffusion of elements.
As the picture shows, there were more black particles near the interface in the 304/Q235 composite plate at the reduction rates of 25% and 30%. As the reduction rate increased to 35% or 40%, the particles became smaller and were distributed at the interface, this is because the oxide was crushed under large positive pressure. As the reduction continued to increase, when the reduction rate reached 40% (figure 2(c)), the large rolling force caused the black particles to almost disappeared, and the crushed oxide was dissolved in the composite interface, which helped to enhance the interface strength. The results show that during the process of compression deformation, part of the oxide film and linear inclusions were crushed into fine particles, and evenly distributed on the bonding interface, which is helping to improve the interface bonding strength and interface toughness.

Long Li [22] reported the $\Delta G$-T relationship curves for various oxidation reactions. When $T$ reaches 1200 °C, all chemical equations display a common phenomenon: $\Delta G < 0$ and $\text{Al}_2\text{O}_3 < \text{SiO}_2 < \text{MnO} < \text{Cr}_2\text{O}_3 < \text{FeO} < \text{Fe}_2\text{O}_4 < \text{NiO} < \text{Fe}_2\text{O}_3 < 0$, which is basically consistent with table 1. Obviously, it can be observed that the oxidation reaction proceeded spontaneously from the top to the bottom more easily, and the affinity of the corresponding material and element $O$ gradually increased. Based on the selective oxidation theory, in the preparation of stainless steel clad plates, elements with stronger affinity with element $O$ were more likely to form oxide inclusions [23].

Figure 3 demonstrates the interface line scan results with various reduction rates by SEM. The Fe, Cr and Ni elements varied greatly, due to the large difference in the Fe, Cr and Ni elements in 304 and Q235 (304 stainless steel contains a large amount of Cr, Ni, and the Fe content is relatively low). Due to the energy spectrum’s insensitivity to light elements such as C, the C existence on both sides of the interface was not apparent. Both Cr and Ni in the 304 stainless steel diffused from the original interface to the carbon steel side, whereas the diffusion distance of Cr was longer than that of Ni. The Cr- and Ni-enriched diffusion layer was formed by the diffusion of the interface elements. The diffusion of Cr and Ni resulted in the formation of a Cr and Ni layer with a width of around 15 $\mu$m on the carbon steel side. At different reduction rates, there was no significant difference in the thickness of the diffusion layer due to the short compression deformation and air cooling duration.

The morphology of the TD-ND bonding interface was observed through EBSD and the microstructure evolution was analyzed for the 304/Q235 composites under various rolling reduction rates. In order to analyze the interfacial structure of the hot-rolled 304/Q235 composite plate, the inverse pole figure (IPF) + grain boundary diagram, grain boundary size and recrystallization characteristics of the interface region were studied. Moreover, the formation mechanism of the hot-rolled 304/Q235 composite was described.

Figure 4 shows the IPF + grain boundary, KAM and recrystallization map of 304 stainless steel/Q235 composite plate interface at different reduction rates selected from the planar TD-ND direction. The upper layer...
is Q235 carbon steel and the lower layer is 304 stainless steel. Between these two layers, there is a bonding interface. It was observed that the Q235 grain size was as fine as the axis, while the 304 stainless steel grain was coarse and the grain boundary was flat. The interface between them was flat and there was no obvious preferred direction.

In the heating furnace, keep it at 1200 °C for 60 min, the 304 stainless steel and Q235 carbon steel grains all grew. For the difference in deformation resistance between the 304 stainless steel and the Q235 carbon steel (Q235 carbon steel deformation resistance is small), in the deformation process, the carbon steel deformed first, and the degree of deformation was large, resulting in recrystallization and grain refinement. Under the reduction rate of 25%, the average grain size of the Q235 carbon steel was 26.2 μm. Under 30%, the recrystallization degree of the carbon steel was high, the grains were further refined, and the average grain size reached 23.7 μm. At the same time, the grain refinement of the 304 stainless steel was not obvious, and only on the composite interface, the number of fine crystal grains increased, indicating that the recrystallization degree at the interface increased. Under the subsequent 35% reduction rate, the Q235 carbon steel grain size increased to a certain extent, with an average grain size of 13.8 μm, while the 304 stainless steel grains began to be refined. This means that with the increase of the reduction rate, the Q235 carbon steel hardened, and its deformation resistance increased, while the deformation of the stainless steel increased, and its grains began to be refined. Under the reduction rate of 40%, the deformation of both the carbon steel and the stainless steel increased, and the grains on both sides of the interface were further refined. The whole process was like this: the easy-to-deform carbon steel was deformed first, the hard-to-deform stainless steel began to be deformed at a certain point, and then they were both further deformed together.

In polycrystals, the plastic deformation of a grain cannot be independent and will inevitably cause coordination and deformation of other grains around it. Similarly, in a composite material, the deformation of one component material causes the coordinated deformation of the other material at the interface. Q235 carbon steel and 304 stainless steel can be coordinated and deformed through the composite interface when deformed by external pressure. It can be seen from the figure 5 that the small crystal grains with different sizes appear on

![Figure 4. IPF + grain boundary, KAM and recrystallization map of 304 stainless steel/Q235 composite plate interface at different reduction rates](image-url)
When the reduction rate reached 35%, the Q235 carbon steel and the 304 stainless steel on both sides of the composite interface showed a coordinated deformation trend.

The local kernel average misorientation (KAM) refers to the average value of the orientation difference between the center point of the core and all the points closest to it [24]. The misorientation map reflects the deformation state inside the grain, and the misorientation imaging method is a straightforward and visible plastic deformation method [25]. Therefore, the higher the local KAM value, the greater the deformation degree of the crystal torsion and the larger the corresponding strain amount. Conversely, when the strain amount is small, the local KAM is also low.

Figures 4(e)–(h) shows the local KAM distribution of the 304 stainless steel/Q235 composite plate at different reduction rates. With the reduction rate increased from 25% to 35%, the KAM value on the Q235 carbon steel side was small, mainly due to the recrystallization which resulted in a low degree of crystal torsional deformation. However, on the 304 stainless steel side, the KAM value was high, and as the reduction rate increased, the KAM value gradually increased. The composite interface of the 304 stainless steel and the Q235 carbon steel had a high KAM value. Under the reduction rate of 40%, both the 304 stainless steel and the Q235 carbon steel possessed high KAM values. The greater the deformation, the higher the mean local KAM value, and the high KAM values were mainly distributed on the composite interface and the local grain boundary of the stainless steel.

In the EBSD test, the blue areas represented the recrystallization and dislocation soft regions, and the dislocation density was very low; the yellow areas represented the sub-grain regions with low orientation differences. The red area represented a high deformation area with a high dislocation density. These three differences are the orientation difference angles (1°–7.5°).

As shown in figures 4(i)–(l), the recrystallization distribution map of the 304 stainless steel/Q235 composite plate at different reduction rates. With the reduction rate increased from 25% to 35%, the blue area on the Q235 carbon steel side was large, due to the recrystallization and therefore low misorientation within the crystals. As the reduction rate increased, the recrystallized region represented by blue kept increasing. At the same time, the yellow subgrain region gradually reduced from large to small. This indicated that the recrystallization refinement grains increased. However, on the 304 stainless steel side, the yellow subgrain region and the red deformation region appeared. As the reduction rate increased, the red deformation region increased as well. When the reduction rate reached 40%, the red deformation regions showed a pattern of a large number of scattered small regions, and the local blue recrystallization regions appeared. At the same time, on the Q235 carbon steel side, the blue recrystallization regions reduced, and the local red deformation regions appeared.

On the composite interface between the 304 stainless steel and the Q235 carbon steel, a large number of red deformation regions and blue recrystallization regions appeared, and as the reduction rate increased, both regions increased. Recrystallization on the bonding interface facilitated the diffusion of elements within the interface and formed a strong metallurgical bond. Main recrystallization occurred, and the internal orientation of the grains was small.

As shown in figure 5, the statistics are the percentage of recrystallization, substructure, and deformed structure on the side of the composite panel interface for Q235 carbon steel and 304 stainless steel in the test area. The recrystallization content of carbon steel increased with the reduction rate from 63% to 79.4%, while at the 40% reduction rate, it decreased to 33.7%. At this time, the deformation substructure suddenly increased to 52.3%, and the deformation area increased 14.1%. In the stainless steel layer, as the reduction rate increased, the recrystallization fraction was very small, the deformation area showed an increasing trend, and the substructure area showed a downward trend. The deformation resistance of carbon steel was lower than that of stainless steel.

With the increase of the deformation reduction rate, the deformation of carbon steel in the composite plate was large. With the increase of the reduction rate to 40%, the carbon steel with low deformation resistance, due to the
effect of deformation hardening, the deformation resistance increased, the recrystallization fraction decreased, and the substructure increased. At this time, 304 stainless steel assumes enhanced deformation and local grain refinement.

4. Conclusion

In this paper, the effect of reduction on the microstructure of the 304/Q235 pressed composite plate was analyzed:

(1) There were more black particles near the interface of the 304/Q235 composite plate at the reduction rate of 25% and 30%. As the reduction rate increased to 35% or 40%, the particles became smaller and were distributed at the interface. The diffusion of Cr and Ni resulted in the formation of a Cr and Ni layer with a width of around 15 μm on the carbon steel side. At different reduction rates, there was no significant difference in the thickness of the diffusion layer.

(2) Under the reduction rate of 25%, the average grain size of the Q235 carbon steel was 26.2 μm. Under 30%, the recrystallization degree of the carbon steel was high, the grains were further refined, and the average grain size reached 23.7 μm. At the same time, the grain refinement of the 304 stainless steel was not obvious, and only on the composite interface, the number of fine crystal grains increased, indicating that the recrystallization degree at the interface increased.

(3) Under the subsequent 35% reduction rate, the Q235 carbon steel grains size increased to a certain extent, with an average grain size of 13.8 μm, while the 304 stainless steel grains began to be refined. Under the reduction rate of 40%, the deformation of both the carbon steel and the stainless steel increased, and the grains on both sides of the interface were further refined. The whole process was: the deformed carbon steel was deformed first, and the hard-to-deform stainless steel began to be deformed at a certain point, and then they were both further deformed together.

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