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Growth kinetics and thickness prediction of interfacial intermetallic compounds between solid steel and molten aluminum based on thermophysical simulation in a few seconds

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ABSTRACT

The growth kinetics of intermetallic compounds (IMCs) generated by initial interfacial reaction (interaction time < 10 s) between solid steel and molten aluminum was investigated using a thermophysical simulation method. According to the morphology evolution of the η (Fe_2Al_5) and θ (Fe_4Al_{13}) phase layers in the isothermal experiments in seconds, it is confirmed that the reaction diffusion controls the η phase growth and the θ phase precipitates from the liquid phase during cooling. The growth kinetics models of the η phase maximum and average thickness are proposed as well. The results showed that the maximum growth was governed by the interfacial reaction, whereas the average thickness was governed by a diffusion and interfacial reaction process. Considering the application of the kinetics model in welding-brazing of Fe/Al dissimilar metals, a mathematical model to predict the average thickness of η phase was established using the finite difference method in thermal cycles as well. The predicted results are consistent with the experimental results, indicating that the model is of good accuracy.

1. Introduction

In recent years, industrial production and transportation have caused severe environmental pollution. As a result, the transportation industry has been challenged to reduce fuel consumption. Therefore, the hybrid structure of lightweight alloys along with steel, such as steel-aluminum hybrid structure aiming to reduce energy consumption and air pollution, has been produced in the automotive industry [1,2]. However, the welding process is difficult due to oxidation of aluminum and the physical property difference, such as melting temperatures, thermal expansion, and conductivity. In addition, the main challenge of steel-aluminum welding is the formation of inherently brittle intermetallic compound(IMC) layers at the steel/aluminum interface, in which thickness and morphology are important in obtaining welding joint with an optimum performance. Wagner et al. gained a mechanically sound steel-aluminum joint while the IMC layers thickness was < 10 μm [3].

At present, a variety of welding methods to manufacture steel-aluminum hybrid structure have been reported, such as tungsten inert gas welding (TIG), metal inert gas Welding (MIG), cold metal transfer welding (CMT) and laser welding [4–10]. The steel welding-brazing to aluminum effectively reduced the thickness of IMCs and gained the joints with sound mechanical properties. The thickness and morphology of the IMC layers are currently controlled by arbitrary experiences, which is lack of quantitative research [11,12]. It results in unstable mechanical properties and application performance. It is not widely used in industries yet.

The mechanism of the interface reaction between solid steel and molten aluminum is the key to control the reaction process and joint property [13]. Numerous researchers have studied the interface reaction mechanism by analyzing the welding joint microstructure from the welding process. However, this method is unable to provide real-time reaction information [14]. There was also numerical simulation, but the results were not accurate due to the limitation of the calculation model [15,16].

The experiments based on aluminizing [17] or Fe/Al diffusion couples [18] deduced only the interfacial reaction mechanism in a long reaction time (a few seconds to several minutes). Previous results reported that the η phase adhering to the steel and θ phase adhering to the aluminum were found [19]. Bouché et al. have reported that the interface of η and Fe was irregular, i.e. the η was embedded in the steel substrate with a finger-like or a tongue-like shape, while other scholars have also received the similar results [17,20–22]. Most researchers believed that this phenomenon was because of the large vacancy

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concentration in the *c*-axis direction of η crystal structure, even some scholars have pointed out that in the *c*-axis direction of η lattice structure the vacancy can reach 30% [23].

The high vacancy concentration leads to the anisotropy of atom diffusion, which affects the growth rate of η in the direction perpendicular to the interface layer. Both Bouché and Shahverdi have pointed out that the interface between η/θ and θ/Al also exhibited irregular morphology. But they were flatter than η/Fe interface [24,25]. With increasing temperature and longer reaction time, free needle-like or massive θ phase appeared in the aluminum substrate. Most scholars believed that this result was due to the dissolution of Fe atom. Because the solubility of Fe atom is zero in aluminum at room temperature, Fe atom dissolves in the liquid aluminum at high temperature and crystallizes in the cooling process in the form of θ phase.

For the growth mechanism and reaction kinetic of IMCs, Dennner has reported that thickening of the IMC layers obeys a parabolic law with increasing reaction time [26]. The experiments on growth kinetics between 700 and 900 °C studied by Bouayad and Tanaka indicated that after a short period, the η layer growth obeys a parabolic law [18], while the θ layer growth is a linear law [24]. Furthermore, Bouché has found that the growth of η and θ phase was controlled by interdiffusion after a non-parabolic initial transient period. According to the parabolic law, the results of hot dip aluminization at the 800 °C showed that the thickness of the IMC layer was greater than zero when the reaction time was zero [13]. This indicated that the IMCs has a rapid growth process at the beginning of the interface reaction, and the parabolic law cannot characterize the interfacial solid steel and molten aluminum reaction mechanism at the initial stage. The interfacial reaction mechanism in the initial stage, i.e. reaction time less than 10 s, is greatly significant for understanding the reaction mechanism in the actual welding process [10,22].

In summary, the majority of the existing growth model of IMCs are based on the isothermal interface reaction of solid steel and molten aluminum. But it actually depends on the thermal cycle during the welding-brazing process and is sensitive to the thermal cycle. The current research methods for the thermal cycle of the interfacial reaction of solid steel and molten aluminum have several limitations:

(1) Although the temperature of interfacial layers could be measured directly by thermocouple to determine the thermal cycle of the welding-brazing [27,28], it was rough to investigate accurately the interfacial structure under the corresponding the thermal cycle due to the instability of the welding process, the inhomogeneous temperature field and the influence of the thermocouple size. The thickness prediction of the IMC layers is also significantly different from the experimental results [28].
(2) Studies by numerical simulations are also used to predict IMCs thickness under the thermal cycle. But a large error between the results of simulation and experiment tests was inevitable because of the dependence of the model of the numerical simulation. This method also cannot accurately study the solid/liquid interfacial reaction mechanism and predict the thickness of IMCs [29]. According to the existing experiments and theory, the prediction error of the IMCs thickness could be very large. Moreover, the microstructure of the IMCs cannot be precisely controlled to develop the potentiality of welding-brazing.

In this paper, the interface reaction between solid steel and molten aluminum in a few seconds was investigated by a Gleeble1500 thermophysical simulation technique. The morphology and thickness of IMCs in the isothermal interfacial reaction were observed, and the formation mechanism of IMCs was analyzed. The kinetic parameters of η growth were calculated. The prediction model of the η phase thickness in different thermal cycles was also verified by the experimental results.

2. Experimental and Methods

2.1. Materials Preparation

The materials used in the experiments were 1 mm thick Q235 steel sheets (0.19 wt% C, 0.80 wt% Mn, 0.04 wt% Si, 0.33 wt% Al, balanced Fe) and aluminum blocks with a purity of 99.99 wt%. A commercial rod of Q235 steel with a diameter of 10 mm was cut into a length of 90 mm.

Before the experiment, the grease and residues on the keyways and the surface of Q235 steel sheet A (6 mm × 5 mm × 1 mm) and B were removed with acetone to avoid affecting the experimental results. Aluminum blocks were placed into the 10% NaOH solution for 3 min to remove oxides and grease on the surface, then placed into 10% HNO_3 solution for about 10 s. After chemical polishing, they were cleaned with deionized water and were dried with a hair dryer. The suspension of NOCOLOK flux in alcohol was uniformly coated on the surface of Q235 steel sheet A and two aluminum blocks (the coating thickness was about 0.5 mm).

The steel sheet A was immediately sandwiched between the two freshly prepared aluminum blocks in the keyway of the rod. Then, the keyway was sealed with a steel sheet B to prevent the molten aluminum from splashing and to isolate the air. The assemblage of aluminum blocks, steel sheet, and the keyway is an interference fit. When the experimental samples were machined, dimensional deviation of the keyway is lower, and the aluminum blocks and steel sheets are an upper deviation. The sample dimensions and assemblage of steel sheets, aluminum blocks, and steel rod are shown in Fig. 1.

2.2. Thermophysical Simulation

In the test, the K-type thermocouples were welded directly at the bottom center of the keyway, and the distance between the positive and negative poles was about 1.0 mm. The Gleeble-1500 thermophysical simulator is able to heat the sample to a high temperature in a very short time. However, the superheat and the heating time, which is required for melting the aluminum blocks, also increase with the increase of heating rate.

In order to achieve synchronization of preset temperature and melting temperature of aluminum, the specimens are preheated to 600 °C at a rate of 60 °C/s holding for 5 s. The peak temperature varies from 700 to 900 °C at a fast rate and holding time is for 0–9 s, then the sample is quenched to room temperature by the automatic water spray device immediately. A typical temperature curve of the experiment is shown in Fig. 2.

The peak temperature of the experiment is set at 700, 750, 800, 850 and 900 °C, holding for 0, 1, 3, 5, 7, and 9 s, respectively. Although

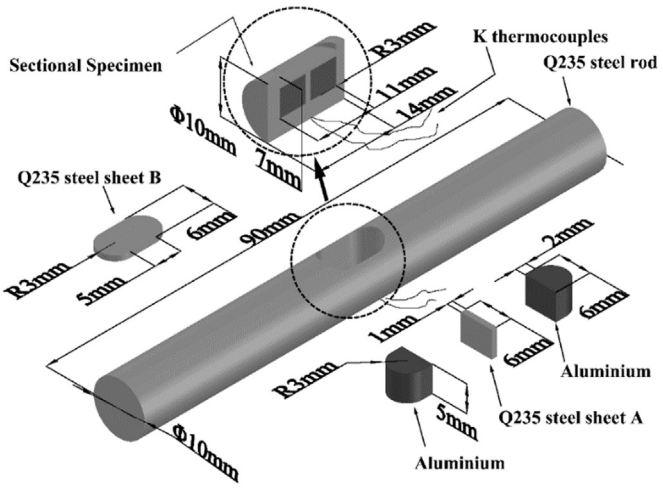

Fig. 1. Diagram of sample dimensions and assemblage of experimental materials.