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Microstructure and mechanical properties of Cu/Al/Cu clad strip processed by the powder-in-tube method

H.T. Gao[a], X.H. Liu[a,b,*], J.L. Qi[a], Z.R. Ai[c,d], L.Z. Liu[d]

[a] State Key Laboratory of Rolling and Automation, Northeastern University, Shenyang, 110819, China
[b] Research Academy, Northeastern University, Shenyang, 110819, China
[c] Key Laboratory for Anisotropy and Texture of Materials, Northeastern University, Shenyang, 110819, China
[d] School of Material Science and Engineering, Northeastern University, Shenyang, 110819, China

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ABSTRACT

A Cu/Al/Cu clad strip was produced successfully by the powder-in-tube method. The interface morphology, element distribution, peeling strength and tensile strength of the clad strips after annealing and further cold rolling were investigated. The experimental results indicate that the diffusional width of the intermetallic layer will increase with the annealing temperature, but the average peeling strength of the Cu/Al/Cu clad strip rises first and then falls. Further cold rolling process can significantly reduce the thickness of the intermetallic layer and improve the average peeling strength of the Cu/Al/Cu clad strip except for those annealed at 550 °C. There exists an optimal diffusional thickness (approximately 15.4 μm) for the annealed Cu/Al/Cu clad strip processed by the powder-in-tube method. The clad strips annealed at 450 °C and further cold rolled to 0.5 mm can obtain the best mechanical performance, the peeling strength, ultimate tensile strength and elongation are 17.8 N/mm, 286 MPa and 9.4%, respectively. The peeling surfaces, peeling tests and tensile tests prove that compact and uniform intermetallic compounds can provide excellent performance for Cu/Al/Cu clad strips.

1. Introduction

Cu/Al/Cu clad strips not only inherits the excellent conductivity, thermal conductive and formability from the substrate metals but also have the advantages of attractive appearance and saving precious metals. It can be widely used in many fields, such as transfer pipes, reservoirs, heat exchangers, kitchen utensils and atomic energy applications (Jing et al., 2014). There have been many discussions about the techniques to fabricate clad strips. In the early studies, copper clad steel strips with high-dimensional accuracy and short technological process were obtained by Yu et al. (1998) through twin-roll casting technology. Gerland et al. (2000) explored the feasibility of explosive cladding of a thin Ni-film to an aluminum alloy. Abbasi et al. (2001) studied the growth rate of intermetallic compounds at the interface of cold rolling bonded Al/Cu bimetal at 250 °C and compared the results with a similar study performed on the friction welding of Al to Cu. Viala et al. (2002) manufactured bimetallic automotive components consisting of light alloys and cast iron by using the combination of Al-Fin process and gravity casting. Butt-joint welding of an aluminum alloy plate to a steel plate was successfully achieved by Kimapong and Watanabe (2004), and the effects of pin rotation speed, position of the pin axis, and pin diameter on the tensile strength and microstructure of the joint were investigated.

In recent years, in-depth, theoretical investigation on the preparation methods of clad strips is quickly increasing. Bae et al. (2011) fabricated Mg/Al clad sheets by a simultaneous casting, cladding and rolling method. Grydin et al. (2013) extended the twin-roll casting technology towards the manufacturing of aluminum–steel clad strips and analyzed the contact zone structure and properties of the flat bimetallic product. To achieve a sound metallurgical bonding between Al alloy or pure Al and low-carbon steel, Liu et al. (2014) developed an auxiliary hot-dip Zn-Al alloy process. Kim et al. (2015) investigated the effect of annealing and secondary rolling process on the evolution of interface microstructure as well as the interfacial bonding mechanisms of roll-bonded two-ply Mg/Al clad sheets. Xie et al. (2015) investigated the microstructure and mechanical properties of CP-Ti/X65 bimetallic sheets fabricated by explosive welding and hot rolling. To weld 3003 aluminum alloy (Al) to 4130 steel (Fe), Chen et al. (2016) developed the vaporizing foil actuator welding method. Li et al. (2016) investigated the effects of processing variables and heat treatments on the Al/Ti-6Al–4 V interface microstructure of bimetal clad-plate fabricated via a novel route employing friction stir lap welding. These production methods have their own application range and certain limitations, which still exist some problems in the process of industry production,

* Corresponding author at: State Key Laboratory of Rolling and Automation, Northeastern University, Shenyang, 110819, China.
E-mail addresses: liuxh@mail.neu.edu.cn, 13709822710@163.com (X.H. Liu).

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such as high energy consumption, severe pollution and spotty performance (Li et al., 2013). More importantly, the irregular edges on the clad strips, which are formed by the ductility difference between copper and aluminum, definitely reduce the metal yield.

The powder-in-tube method is usually applied to produce high-temperature superconductivity wires or strips. It can significantly improve the metal yield of clad sheets or strips with a simple process technology and low production cost. Malagoli et al. (2002) successfully fabricated long nickel-sheathed MgB_2 superconducting tapes by the powder-in-tube method. Song et al. (2002) reported the successful fabrication of single-filament composite MgB_2/stainless-steel ribbons by the powder-in-tube process. In addition, many new application areas have been explored. Yoo et al. (2013) successfully fabricated high-strength carbon nanotube/Cu composites by using powder-in-tube method and high-ratio differential speed rolling. As an attempt to recycle iron scrap, Zhang et al. (2014) produced a stainless steel/iron scrap clad plate by the powder-in-tube method and hot rolling. Gao et al. (2016) synthesized a steel clad copper rod reinforced with reduced graphene oxide (RGO) by the combination of powder-in-tube method and intermediate annealing (IA).

To the best of our knowledge, the open literature suggests that no attempt has been made to study the Cu/Al/Cu clad strip produced by the powder-in-tube method. To fill this gap, the interface and mechanical strength of Cu/Al/Cu clad strips fabricated by this novel method under different reduction rates and annealing temperatures were investigated in this paper.

2. Experimental procedures

Cu/Al/Cu clad strips were successfully fabricated by using the powder-in-tube method. To remove the oxide and other impurities, the inner wall of the copper tube was cleaned using a wire brush and acetone. This process could also increase the surface roughness and expose fresh metal, which is beneficial for the bonding of two metals. Al powder (99.99% in purity) was packed into the pure copper tubes (99.5% in purity) with an outer diameter of 10 mm and an inner diameter of 8 mm. Then, these composite tubes were sealed and cold worked into tapes (1.5 mm thickness) by flat rolling. The as-rolled tapes were cut into pieces, which were annealed in a resistance furnace, followed by further cold rolling. To explore the influence of annealing temperature and reduction rate on the microstructure and mechanical properties of the Cu/Al/Cu clad strip, these tapes were first annealed at 350 °C, 450 °C and 550 °C for 40 min, respectively. For comparison, half of these samples were subsequently rolled to a thickness of 0.5 mm. Only three rolling passes for clad strips from 10 mm to 0.5 mm are needed (10 mm–4 mm to 1.5 mm–0.5 mm). Moreover, the edges of samples are trim without obvious cracks, which can guarantee high yield of the clad strip. Fig. 1 illustrates the fabrication procedures of the Cu/Al/Cu clad strip used in this investigation.

The morphology of the bonding interface, peeling surface and tensile fracture was analyzed with a SHIMADZU SSX-550 scanning electron microscope (SEM) equipped with energy dispersive spectrometer (EDS). To investigate the element diffusion across the interface, a JEOL JXA-8530F electron probe micro-analyzer (EPMA) was used with an acceleration voltage of 20 kV and a sample current of 2×10^{-8} A. To test the bonding strength and tensile property, T-type peeling test and tensile test were carried out on a TH5000 universal testing machine under a crosshead speed of 3 mm/min at room temperature. Figs. 2 and 3 show the photographs of peeling test and tensile test, respectively.

3. Results and discussion

3.1. Microstructure of bonding interface

Fig. 4(a)–(c) shows the SEM images close to the bonding interface of the samples (1.5 mm thickness) annealed at different temperatures. Cu and Al are tightly bonded and obvious interfacial intermetallic layers can be observed. In addition, the thickness of the reaction layer will continue to increase with the rise of heat treatment temperature. At a low annealing temperature, most of the interfacial atoms are bound by surface energy. Thus, interface diffusion is faintly characterized by a narrow intermetallic layer (Fig. 4(a)). When the annealing temperature rises, thermal energy obtained by copper atoms and aluminum atoms can exceed the diffusion barrier and promote the formation of more intermetallic compounds. The range of atomic diffusion is enlarged, and a wider diffusion layer is achieved (Fig. 4(b) and (c)). It is interesting that the bonding interface after annealing becomes wavelike and the wave amplitude will become larger with the increase of annealing temperature (Fig. 4(a)–(c)).

The bonding interfaces of the 350 °C, 450 °C and 550 °C annealed clad strips suffering 95% cold rolling reduction (0.5 mm thickness) are shown in Fig. 4(d) to (f), respectively. Compared with the interfaces of the annealed clad strips, the diffusion layer after further cold rolling becomes thin and discontinuous (Fig. 4(d)–(f)), which results from the cracking of brittle diffusional layer. Especially, for the 550 °C annealed clad strip with a thick diffusional layer, further cold rolling even causes visible diffusional layer fracture as a result of the large rolling force penetrating the thickness of the clad strip. Some fragments of intermetallic compounds are squeezed into the aluminum substrate, as shown in Fig. 4(f).

3.2. Element diffusion

Fig. 5 shows the line scans across the copper/aluminum interface. In the annealed clad strips, the diffusional width increases from 5.8 μm at 350 °C (Fig. 5(a)) to 35.1 μm at 550 °C (Fig. 5(c)). After further cold rolling, the diffusional width at the bonding interface significantly decreases, from 2.6 μm for the 350 °C annealed strip (Fig. 5(d)) to 8.3 μm for the 550 °C annealed strip (Fig. 5(f)). Moreover, it can be observed that the degree of atomic diffusion at the bonding interface for Cu is stronger than that for Al. For the intermediate annealing process, the diffusion coefficient of Cu on Al is higher than that of Al on Cu. For example, D_{Cu-Al} is 9.2×10^{-21} m^2/s and D_{Al-Cu} is 3.4×10^{-21} m^2/s for T = 110 °C (Ene et al., 2007). Besides, this difference will become more marked when the temperature increases (Shackelford and Alexander, 2001). As for the further cold rolling process, the lattice constants for Cu and Al are $\alpha_{Cu} = 0.3615$ nm and $\alpha_{Al} = 0.4082$ nm, respectively. Thus, Cu atoms enter the interstitial site of Al lattice easier than that for Al atoms, which has been proven by the accumulative roll bonding process of multi-layered Al/Cu composites (Eizadjou et al., 2008).

To make the difference of inter-diffusion zone more apparent, the width values under different conditions are plotted in Fig. 6. It is interesting that the higher intermediate annealing temperature, the larger the reduction of diffusional width that is achieved. The reductions of diffusional width for the clad strips annealed at different temperatures are 3.2 μm (350 °C), 8.5 μm (450 °C) and 25.8 μm (550 °C), respectively. After high-temperature (above 300 °C) annealing, the interfacial reaction layers have been reported to consist of several layers (Kim and Hong, 2013a,b), including Cu_9Al_4, CuAl (Cu_4Al_3) and $CuAl_2$. All of these chemical compounds are brittle and of low plasticity. The varieties and changes of the Cu/Al compound on the peeling surfaces of the clad strips will be discussed in the following sections in detail.

3.3. Peeling strength

The peeling strength curves and the average peeling strength of the Cu/Al/Cu clad strips after annealing and further cold rolling are shown in Fig. 7. The experimental results show that the Cu/Al/Cu clad strips processed by the powder-in-tube method have a relatively steady performance (Fig. 7(a)). With the increase of annealing temperature, the average peeling strength of the clad strip (1.5 mm thickness) increases