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Microstructural characteristics of joint region during diffusion-brazing of magnesium alloy and stainless steel using pure copper interlayer

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Abstract: A novel joining method, double-stage diffusion-brazing of an AZ31 magnesium alloy and a 304L austenitic stainless steel, was carried out using a pure copper interlayer. The solid-state diffusion bonding of 304L to copper was conducted at 850 °C for 20 min followed by brazing to AZ31 at 520 °C and 495 °C for various time. Microstructural characteristics of the diffusion-brazed joints were investigated in detail. A defect free interface of Fe−Cu diffusion area appeared between the Cu alloy and the 304L steel. Cu−Mg reaction products were formed between AZ31 and Cu alloys. A layered structure including AZ31/Cu−Mg compounds/Cu/Fe−Cu diffusion layer/304L was present in the joint. With time prolonging, the reduction in the width of Cu layer was balanced by the increase in the width of Cu−Mg compounds zone. Microhardness peaks in the zone between AZ31 and Cu layer were attributed to the formation of Mg−Cu compounds in this zone.

Key words: magnesium alloy; stainless steel; diffusion bonding; brazing; microstructural characteristics; dissimilar metals welding

1 Introduction

Magnesium alloys are the lightest metallic materials, have the lowest density and exhibit high specific strength, good corrosion resistance, excellent stiffness, good castability, good noise and vibration damping capacity and favorable recycling capability [1−6]. These characteristics make this alloy particularly attractive for the automotive and aerospace industries to improve fuel economy and protect the environment [3], based on the requirement for reduction in the net mass and size of components used in the transport industry. It is well known that stainless steel is one of the most common materials in the modern industry. Thus, tasks of joining magnesium alloy to others such as stainless steel must be faced. Recently, the weldability of magnesium alloys has been investigated using such methods as laser welding [7,8], resistance spot welding [9,10], ultrasonic welding [11], friction stir welding [3,12−14] and diffusion bonding [15,16]. Joining of magnesium alloy and stainless steel can enlarge the application of magnesium and make their advantages yield well. However, these dissimilar magnesium alloys and stainless steels have been difficult to join together by conventional fusion welding because of the significant differences between these two alloys in metallurgical, chemical and physical properties [17].

In view of the facts, diffusion-brazing is a double-stage joining process, which combines the beneficial features of diffusion bonding and transient liquid-phase bonding techniques. This process eliminates the adverse influence of single joining technique on both materials and thus could offer an alternative method for joining these dissimilar metals. In this study, joining of a magnesium alloy to a stainless steel was conducted by the double-stage diffusion-brazing process using a pure copper as the interlayer. Microstructural characteristics of the joint region were examined in detail.

2 Experimental

The composition of 304L stainless steel (mass fraction) was 0.03% C, 0.1% Si, 1.21% Mn, 9.5% Ni, 18% Cr and the balance Fe. The AZ31 magnesium alloy had a composition (mass fraction) of 3.0% Al, 1.0% Zn and 96% Mg.

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Optical microstructures of the 304L steel and the AZ31 alloy are given in Fig. 1. The 304L and AZ31 rods were cut into specimens of *d*16 mm×30 mm. A pure copper foil with the thickness of 100 μm was used as the interlayer. The surfaces of the sectioned coupons were ground on SiC paper to a 2000-grit finish and polished by diamond suspension of 1 μm particle size, and then ultrasonically cleaned in a acetone bath. A series of bonding tests were carried out in a Gleeble−1500D thermo-mechanical system under a pressure of 1×10^{-2} Pa. Between room and bonding temperatures, the heating rate and the cooling rate were around 5 °C/s.

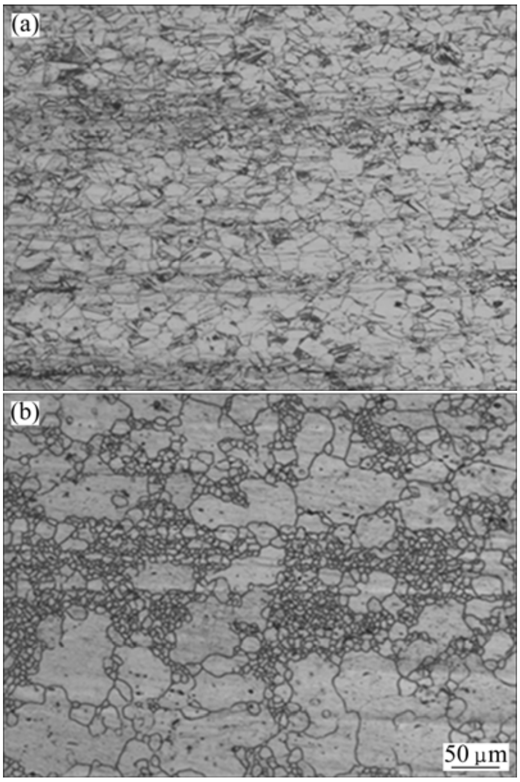

Fig. 1 Optical micrographs of base materials: (a) 304L stainless steel; (b) AZ31 magnesium alloy

The double-stage joining process was composed of higher-temperature solid-state diffusion bonding and lower-temperature transient liquid-phase bonding. The solid-state diffusion bonding of the stainless steel and the copper interlayer was performed at 850 °C using an impulse pressure of 5−20 MPa, in order to achieve a metallurgical joint at the 304L/Cu bond interface. This impulse pressuring diffusion bonding has been reported detailedly in Ref. [18]. After the copper was diffusion bonded to the surface of the stainless steel, the free surface of the copper interlayer was polished to a 1 μm finish. Subsequently, transient liquid-phase bonding of the free surface of the interlayer of the 304L/Cu specimen to the AZ31 magnesium alloy was conducted under a certain pressure of 2 MPa at double temperatures (520 °C and 495 °C) for various holding times, in order to avoid the melting of magnesium alloy at higher temperatures used for diffusion bonding and then induce the formation of a sound bond at the AZ31/Cu bond interface.

Specimens for metallographic examination were sectioned from the diffusion-brazed joints. Ground and polished cross sections were etched in an aqua regia (30% HCl + 10% HNO_3 + 10% water) and a solution of 5 g picric acid, 10 mL glacial acetic acid and 80 mL ethanol to observe the stainless steel side and the magnesium alloy side, respectively. Microstructural observations of the joints were employed by a scanning electron microscope (SEM). The chemical compositions of the micro-zones in the joints were analyzed by energy-dispersive X-ray spectroscopy (EDS). Phase structures in the joints were identified using X-ray diffraction (XRD, with Cu K_α radiation). Microhardness testing of the joint region was conducted using a hardness tester.

3 Results and discussion

3.1 Higher-temperature diffusion bonding of stainless steel to copper

To improve the bonding efficiency and the quality of the joint, joining of 304L stainless steel to copper interlayer by an impulse pressuring diffusion bonding technique was used. Since, during this bonding process, grain refinement could be induced by the compressive deformation under the pressure and the extent of contact of bonded surface can be increased by applying this impulse pressure [18].

Figure 2 illustrates a typical SEM image of the 304L/Cu specimen produced at 850 °C under an impulse pressure of 5−20 MPa with an impulse frequency of 0.5 Hz, where a line scan by EDS analysis was taken across the interface zone. It could be observed from Fig. 2(a) that the 304L/Cu interface zone is free from voids and there is no unbonded area along the interface. From Fig. 2(b), no composition platform present in the interface zone indicates that no intermetallic compounds were formed in the corresponding zone. A Fe−Cu diffusion region in the width of about 0.8 μm appeared at the interface. These results proved that under these diffusion bonding conditions, a sound metallurgical bonding at the 304L/Cu interface zone was produced.

3.2 Lower-temperature transient liquid-phase bonding of AZ31 alloy to 304L/Cu joint

According to the Mg−Cu binary phase diagram [19], transient liquid-phase bonding of AZ31 alloy and 304L/Cu joint was conducted under a small pressure of 2 MPa at 520 °C for various time and then at 495 °C for

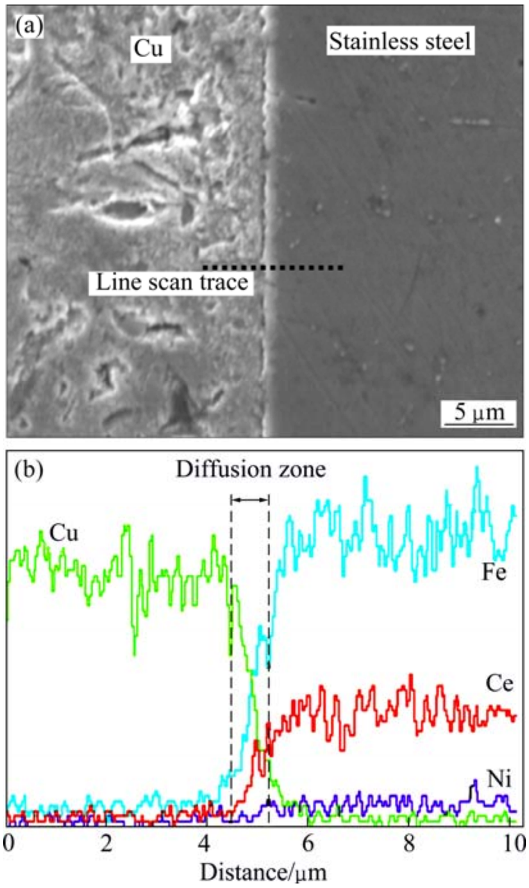

Fig. 2 304L/Cu specimen obtained at 850 °C: (a) Typical SEM image; (b) Line scanning analysis of interface zone (black dot line in Fig. 2(a) showing line scan trace)

5 min. These two temperatures were higher than the Mg−Cu eutectic temperature (487 °C) [19]. The first temperature of 520 °C was selected to enhance the interdiffusion of Mg and Cu and their reaction, and then a certain amount of liquid could be produced. The subsequent temperature of 495 °C, which is slightly lower than the first temperature, was employed to improve the extent of solidification of liquid.

Figure 3 represents typical SEM morphologies of the 304L/Cu/AZ31 joint made at 2 MPa and 520 °C for 11 min and subsequently at 495 °C for 5 min. Figure 4 presents a line scan taken across the joint shown in Fig. 3(a). Table 1 gives the chemical compositions of the zones shown in Figs. 3(b) and (c), obtained by EDS analysis. Figure 5 demonstrates the XRD pattern of the joint region. It can be seen from Fig. 3 that a metallurgical bonding was produced between 304L and AZ31 using copper interlayer and a multilayered structure appeared in the joint area. According to the results of line scan (Fig. 4), Cu−Mg compound zone was present in the bonding area and there was remaining copper with a narrow zone between stainless steel and Cu−Mg compound.

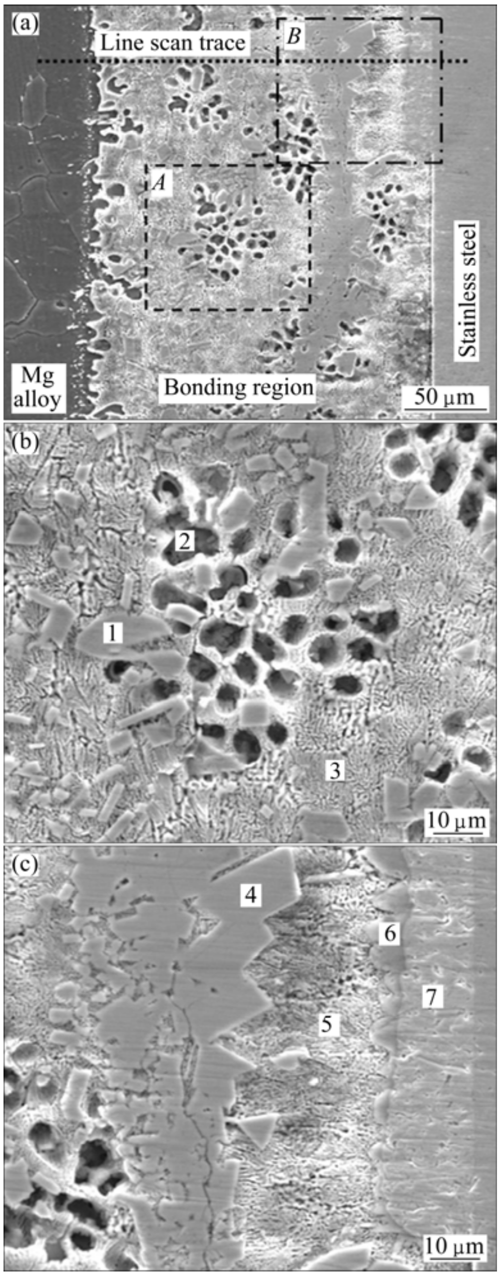

Fig. 3 SEM images of diffusion-brazed 304L/Cu/AZ31 joint produced by bonding at 850 °C and subsequent brazing at 520 °C for 11 min and then at 495 °C for 5 min: (a) Overview; (b) Magnified morphology in zone *A*; (c) Magnified morphology in zone *B*

As can be seen in Fig. 3, the joint area consists of five distinct zones. Based on Figs. 3(b), (c) and Table 1, five conclusions can be drawn. Firstly, the offwhite blocky-shaped phases (zone 1 and zone 4), containing 34.87%−36.31% Mg, 46.04%−47.80% Cu and 17.33%−17.65% Al, are suggested to be Mg−Cu compounds rich in Al, since the AZ31 alloy has some amount of Al. XRD patterns also illustrate that Al−Mg−Cu phase was detected in the joint area. Secondly, a number of Mg phases with a black circular shape (zone 2) are

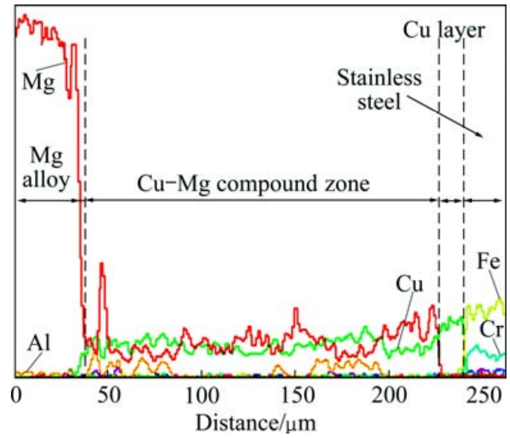

Fig. 4 Line scanning analysis of joint (black dot line in Fig. 3(a) showing line scan trace)

Table 1 Chemical composition of micro-zones within joint in Figs. 3(b) and (c), respectively

Zone	x/%				Possible phase/zone
	Mg	Al	Cu	Fe	
Zone 1	36.31	17.65	46.04	−	Al−Mg−Cu
Zone 2	98.90	−	1.10	−	Mg
Zone 3	48.39	−	51.61	−	Mg−Cu
Zone 4	34.87	17.33	47.80	−	Al−Mg−Cu
Zone 5	65.88	−	34.12	−	Cu−Mg
Zone 6	67.82	−	32.18	−	Cu−Mg
Zone 7	−	−	98.57	1.43	Cu

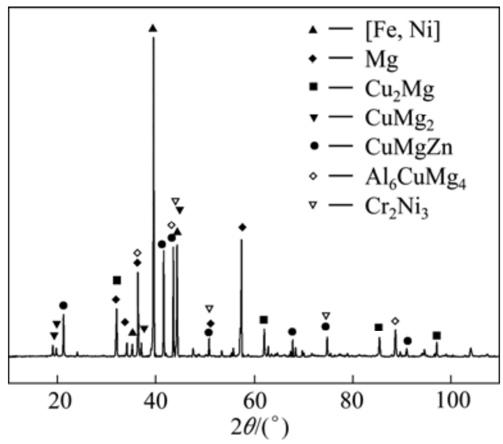

Fig. 5 XRD pattern of joint region

surrounded by the offwhite blocky-shaped phases like zones 1 and 4 and the lamellar structure like zones 3 and 5. The presence of Mg phase in the bonding region can be interpreted as follows. When the interdiffusion of Mg and Cu atoms takes place in the original interface between the AZ31 and the copper, the paths of grain boundaries are used preferentially due to notably higher diffusion rate along these boundaries. Thus, the composition of grain boundary region firstly reaches the eutectic point and then this region becomes liquid. The melting process is from the boundary area towards the grain center region. If a grain is relatively larger and time is insufficient for melting, some tiny zones of Mg are left in the grain center region. Thirdly, zones 3 and 5 with lamellar structure are rich in Cu and Mg. Based on the results of Table 1 and Fig. 5, Cu−Mg intermetallic compounds, namely Cu_2Mg and $CuMg_2$, can be thought to form due to the interdiffusion and the interaction between Mg in AZ31 and Cu in copper interlayer. Fourthly, the dendritic phases (zone 6) formed along the copper layer (zone 7) may be also considered to be Cu−Mg intermetallic compounds since zone 6 has Mg (67.82%) and Cu (32.18%). Fifthly, zone 7, which has the absolute amount of copper, could be regarded as the remaining Cu.

Figure 6 exhibits a line scan taken across the 304L/Cu/AZ31 joint produced at 520 °C for 9 min and subsequently at 495 °C for 5 min. Although the composition distribution in Fig. 6 is similar to that in Fig. 4, there is a difference in the width of each zone of the bonding region. It can be seen from Fig. 7 that as the

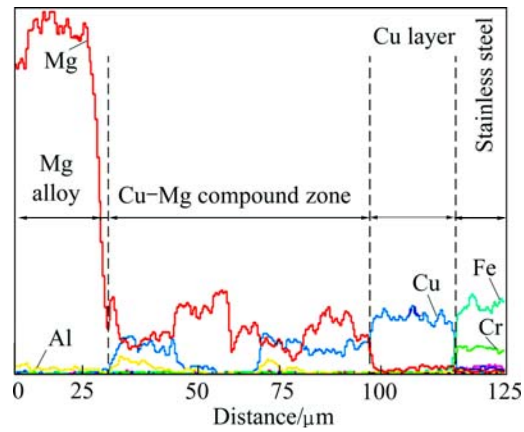

Fig. 6 Line scanning analysis of joint obtained by subsequent brazing at 520 °C for 9 min and then at 495 °C for 5 min

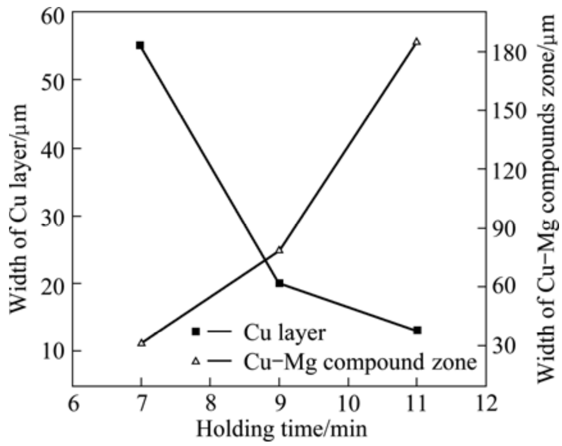

Fig. 7 Width of Cu−Mg compound zone and remaining Cu layer with brazing time

time was prolonged, the Cu layer width decreased and correspondingly the Cu−Mg compound zone width increased. Additionally, the bonding region including Cu−Mg compound zone and Cu layer was widened by increasing the time. Since an increase in the time encourages the diffusion and melting of Cu interlayer and the interaction of Cu and Mg.

3.3 Microhardness profile of diffusion-brazed joint

Hardness profile is a good indicator of bonding microstructure and can be utilized to assess the influence of precipitates and compounds on mechanical properties. Microhardness profile as a function of distance is shown in Fig. 8. The microhardness peak of the bonding region present in the zone between the Cu layer and the AZ31 can be attributed to the microstructure of this zone because the hard, brittle Cu−Mg intermetallic compounds are formed. By contrast, AZ31 and pure copper exhibited lower hardness values, whereas 304L had a higher hardness.

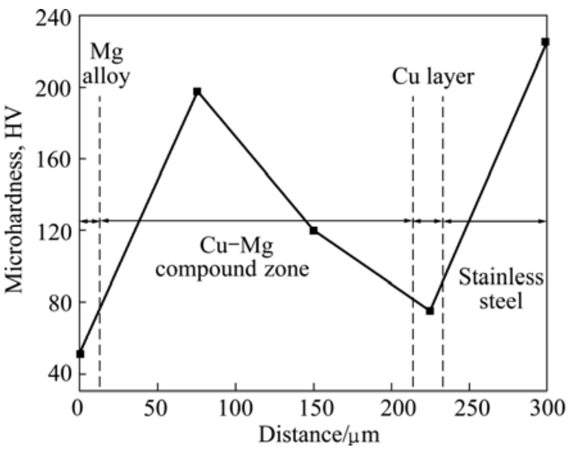

Fig. 8 Microhardness profile as function of distance for joint made by bonding at 850 °C and subsequent brazing at 520 °C for 11 min and then at 495 °C for 5 min

4 Conclusions

1) A metallurgical joint of the AZ31 and the 304L was produced by this novel joining process using the Cu foil. The interface of Fe−Cu diffusion area between the Cu alloy and the 304L steel was free from defects. The Cu−Mg intermetallic compounds were formed between the AZ31 and the Cu layer. A multi-layer structure containing AZ31/Cu−Mg compound/Cu/Fe−Cu diffusion layer/304L appeared in the joint region.

2) By increasing the time, the width of Cu layer reduced and correspondingly that of Cu−Mg compounds zone increased, and furthermore, the bonding region including Cu−Mg compounds zone and Cu layer was widened.

3) The formation of Mg−Cu intermetallic compounds was responsible for the microhardness peaks in the transition zone between the AZ31 and the Cu layer.

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纯铜作中间层的镁合金与不锈钢扩散−钎焊接头区的微观结构特征

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摘 要：以纯铜作中间层采用一种新型的两步式扩散−钎焊方法对 AZ31 镁合金和 304L 奥氏体不锈钢进行连接。304L 与铜的固态扩散连接在 850 °C 下进行 20 min，随后与镁合金在 520 °C 和 495 °C 进行不同时间的钎焊。对扩散−钎焊接头区的微观结构特征进行研究。在铜与 304L 钢之间形成没有缺陷存在的 Fe−Cu 扩散界面。在 AZ31 和铜之间形成 Cu−Mg 反应物。在接头处出现包含 AZ31/Cu−Mg 化合物/Cu/Fe−Cu 扩散层/304L 的层状结构。随着时间的延长，铜层的宽度降低，而 Cu−Mg 化合物层的宽度增加。形成的 Mg−Cu 化合物使 AZ31 和铜层之间的区域出现显微硬度的峰值。

关键词：镁合金；不锈钢；扩散连接；钎焊；微观结构特征；异种金属焊接

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