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Effects of copper addition on microstructure and strength of the hybrid laser-TIG welded joints between magnesium alloy and mild steel

Liming Liu · Xiaodong Qi

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Abstract Lap joint of magnesium alloy AZ31B to mild steel Q235 with the addition of copper interlayer by hybrid laser-TIG welding technique was investigated. The microstructure, element distribution at interfaces, and intermediate phases of joints were examined by scanning electron microscopy (SEM), electron probe micro-analyzer (EPMA), and X-ray diffraction (XRD), respectively. The results showed that intermetallic compounds Mg_2Cu with rod-like structure in the joint and equiaxed structure at interface were found, and the bonding between copper and steel was realized by mixing of copper and steel at upper margins of molten pool and a little solid solution of copper in iron at the bottom and side of molten pool. Besides, comparing with that without any interlayer, the wettability of molten magnesium alloy on steel was enhanced, which led to an intimate connection. In the end, the joining mechanism of magnesium–steel joints with copper interlayer was discussed.

Introduction

Joining Mg alloy with other materials is a trend in dissimilar materials welding field, as Mg alloys with its unique properties like the lightest structural materials, specific tensile strength, and damping capability has attracted attentions of many realms. The application of Mg alloy to electronic products, automobiles, etc. exhibits a great potential in industry [1–3]. Steel is one of the dominant materials in industry which is irreplaceable currently at least. The demand for different materials to be joined with steel has been increasing recently.

L. Liu (✉) · X. Qi
Schools of Materials Science and Engineering, Dalian University of Technology, Dalian 116024, China
e-mail: Liulm@dlut.edu.cn

The welding of dissimilar materials by FSW has attracted vast attention, as the intermetallic layer which could affect mechanical properties greatly could be well controlled. Venkateswaran et al. [4] studied that a narrow intermetallic layer was generated at the Mg/Al alloys interface by FSW, and affected the joint strength greatly. In addition, some other mechanical properties of the joint like impact values [5] could be well acquired. Lee et al. [6] reported that the strength of lap joint between dissimilar Al alloys depended mainly on the interfacial morphology by increasing tool rotation and decreasing welding speed of FSW. Thus interfacial interaction of dissimilar materials in the joint is a critical factor that determines the joint strength. Nevertheless, Mg and Fe elements are in great difference physically and mechanically such as melting points with iron 1536 °C and magnesium 650 °C, tensile strength, tensile shear strength, etc. From Mg–Fe binary phase diagram, it can be seen that the solid solubility of Mg in Fe is 0.00043 at.%, furthermore, there are not any types of intermetallic compounds between them. So the joining mode of Mg alloy to steel by FSW which is friction stir welding can only be mechanical bonding between Mg alloy and steel [7]. Our team has done previously that the lap joint of Mg alloy to steel sheets was joined through complex compounds of magnesium, iron, and oxygen [8] by hybrid laser-TIG welding. As is known to all, high-energy laser beam alone can realize high-speed welding and a great depth in materials; however, the absorption of laser energy for some materials is rather low causing great consumption of electric power. After electric arc is incorporated into the laser welding process in an appropriate way, the efficiency of laser beams was enhanced and the interaction of the arc and laser leads to improved laser welding characteristics [9]. Laser beam or arc alone usually could not yield reliable joint of Mg alloy to steel, while the hybrid processing can be done in such case that arc is used to melt one of the dissimilar materials and laser beam to create large depth on the other one. Thus the fusion welding of Mg alloy and steel can be implemented.

To obtain reliable joints, an intermediate element that can interact with both Mg and Fe elements must be imported. Therefore, Cu was selected as the intermediate element to construct a joint of Mg alloy to steel. In present report, microscopic characterization and shear strength of the joint are investigated. A lap joint configuration was adopted with Cu interlayer set between Mg alloy and steel. And the joining mechanism of the Mg and steel was also expounded.

Experiment procedure

The materials used in the study are AZ31B Mg alloy sheet with nominal composition of Mg–3Al–1Zn–0.2Mn–0.1Si (wt%), Q235 steel sheet with nominal composition of Fe–0.2C–0.3Si–0.7Mn (wt%), and Cu slice with 99.9% of purity. The dimensions of them are $1.7 \times 60 \times 80\ mm^3$, $1.2 \times 60 \times 80\ mm^3$, and $0.1 \times 65 \times 8\ mm^3$, respectively. Before welding the sheets were degreased and ground by acetone and abrasive cloth, respectively. Concerning the assembling methods of Mg alloy and steel sheets in a lap joint configuration, the Mg alloy sheet was placed upon the steel sheet selected in the present experiment. There are two reasons that govern why steel should be under Mg alloy sheets. Firstly, when the penetration in steel was not deep enough to reach the side of Mg alloy sheet, molten AZ31B Mg alloy by heat transfer from steel cannot be joined well with steel due to its bad wettability on steel, in addition, the coefficient of thermal expansion of AZ31B Mg alloy (2.8×10^{-5}/°C) is twice as much as that of steel (1.3×10^{-5}/°C), which may be another reason that caused them not to be joined together. Secondly, when the steel sheet was completely melted, most Mg elements of AZ31B Mg alloy were vaporized and a large number of big pores were generated at the bead of steel and a bad joint with a meaningless strength was attained. In the two processes above, an extreme amount of spatter was generated and inflicted bigger damage to the laser head of the setup. Accordingly, the Mg alloy sheet was placed on the steel sheet in a lap configuration with the dimension of 10–15 mm.

Nd: YAG laser beam is perpendicular to the plate assembled TIG torch with tilting angle α 40°, and the welding direction is parallel to the plane constructed by laser beam and the torch. Thus the hybrid heating source for the welding process is shown in upper right corner of Fig. 1.

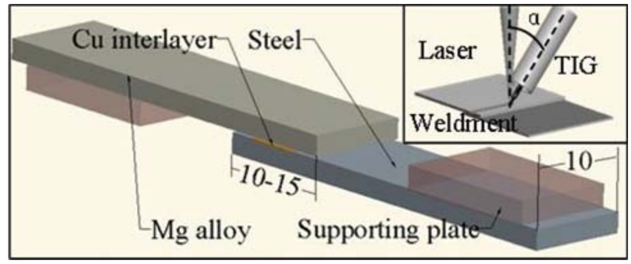


Fig. 1 Sketch of the specimen for tensile shear test (mm)

The optimum parameters in the experiment were 100 A of TIG current, 400 W of laser power, 850 mm/min of welding speed, and TIG nozzle with gas flow of 15 L/min.

Specimens in Fig. 1 machined from the weldment were subjected to tensile shear test using an electronic tension machine (css-2205, screwed driven) with a constant travel speed of 2 mm/min at room temperature. The joint strength is the average of 3–4 specimens. The supporting plates at each end of the specimen were employed to maintain the joint interface parallel to the load direction. The shear strength was calculated as follows

$$\sigma_{b\_shear} = F/S_{//},$$

where F and σ_{b_shear} are the load and the maximum shear strength, respectively; $S_{//}$ is a rectangular bonding area of the joint before tensile shear test, with its width shown in Fig. 3. Thereafter, the weld bead where fracture was located shown in Fig. 2a was analyzed by XRD test.

Macromorphology of the weld was obtained; the cross-sectioned specimens were prepared in accordance with the metallographic method, etched by picric acid for Mg alloy and Nital's (4 vol.% HNO_3 + ethanol) for steel, and the microstructure of the joint was observed by SEM equipped with EDS, the element distribution of the joint was also carried out by EPMA.

Results

The macromorphology of the weld seam and the fracture location are shown in Fig. 2. It is inevitable to produce spatters as the action of high power laser beam, which can be seen in Fig. 2a on the bead surface of Mg alloy; however, the regular ripples without pores or crack still can be seen clearly at weld seam. The fracture occurred at the interface of Cu/steel, and the Cu interlayer attached Mg alloy tightly in Fig. 2b, c. A deep penetration inflicted mainly by laser at steel side presents in circle region of Fig. 2c.

A cross-sectioned joint was shown in Fig. 3. Some particles distributed unevenly are present and a large

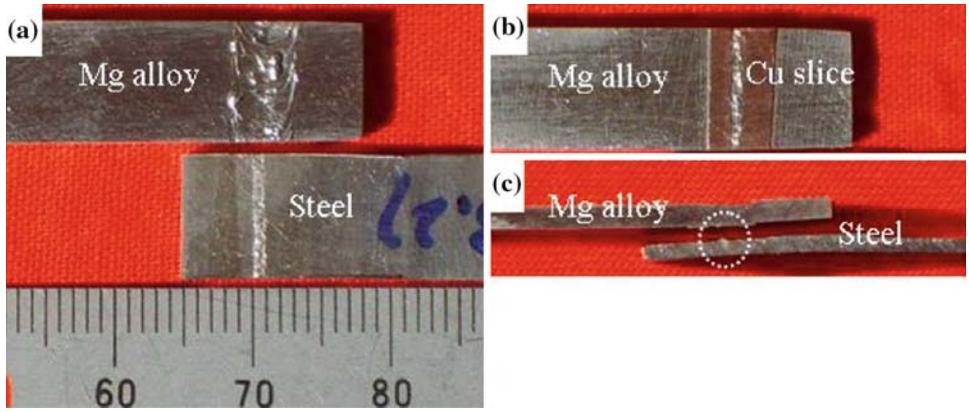


Fig. 2 Macromorphology of weld seam and failure location: **a** front face; **b** the side that Cu interlayer attached to; **c** failure location

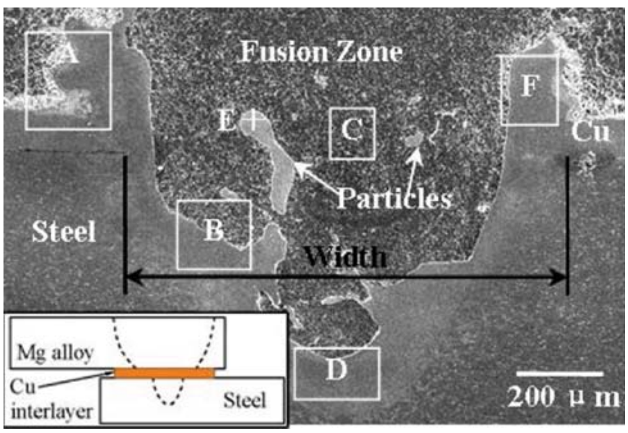


Fig. 3 Cross-section of the Cu-added joint with the sketch of relative location of three plates at lower left corner

number of white dots disperse homogeneously in fusion zone. In region A, a reaction layer attached to Cu was observed. The maximum thickness of the layer is about 40 μm, suggesting that the thickness of layer is non-uniform. EDS analysis shows that position E contains 93.98 wt% Fe and 6.02 wt% Cu, indicating that the particles are mainly the solid solution of Cu in Fe at the condition of rapid cooling in the experiment.

Figure 4 shows the interfacial microstructure of Mg alloy and Cu. In Fig. 4a, the loosen reaction layer with some equiaxed grains inside extends from Cu to Mg alloy in the form of dendritic structure and fragments further in the fusion zone. It is also observed that the reaction layer was delaminated into two layers along the boundary as shown in Fig. 4b, c; one is composed of fine particles close to Cu with larger square grains grown from, and the other exhibits equiaxed structure close to Mg alloy shown in Fig. 4c. Element distribution was conducted along the scanning line in Fig. 4c, and the profile of Mg, Cu across the interface is shown in Fig. 4e, confirming that interface was divided into two layers. Some columnar crystals shown in Fig. 4d were found, but they are not detected in regions I, II of Fig. 4a. It is noteworthy that some trivial particles in Fig. 4d and location E of Fig. 4b accumulated at the root of the columnar structure. The composition of the particles is 87.23 wt% Cu and 12.77 wt% Mg, suggesting that the fine particles are mainly supersaturated solid solution of Mg in Cu under the condition of larger cooling rate in welding process.

The interfacial structure of Mg alloy to steel is shown in Fig. 5. It can be seen that a distinct boundary line is presented and a transitional zone (TZ) from the line to steel is presented. In order to investigate the composition of TZ, line analysis along the scanning line shown in Fig. 5b at the bottom of molten pool was carried out. The results in Fig. 5c, suggest that there are not any diffusion processes taken place in TZ between Mg and Fe elements. Nonetheless, position I of Fig. 5a contains 2.65 wt% Cu and II in Fig. 5b contains 3.53 wt% Cu according to EDS analysis, indicating that most of TZ is steel with a little limited solution of Cu in Fe in terms of Fe–Cu binary phase diagram.

Figure 6 shows the structure of fusion zone. In fact, the aforementioned white dots consisted of rod-like structure which may result from the difference of flow intensity in the weld pool [10] distributed mainly in grain boundary of Mg alloy, which can also be ascertained in Figs. 4e and 5c, as the dashed lines display the location where the Mg intensity is lower, is just where that of Cu is higher, and vice versa. The fine rod-like structure aligned tightly along the grain boundaries is a typical eutectic structure. EDS analysis shows that position A contains 50.34 wt% Mg and 49.66 wt% Cu, indicating that they may be intermetallic compound (IMC) Mg_2Cu.

In order to determine whether the IMC formed at the interfaces, XRD analysis was performed and the spectra are shown in Fig. 7. The results of Fig. 7a show that Mg_2Cu phase was formed. According to the outcomes of EDS analysis at position A in Fig. 6, the rod-like structure is Mg_2Cu phase. The position D in Fig. 4b contains 48.62 wt% Mg and 51.38 wt% Cu, indicating that these areas are mainly

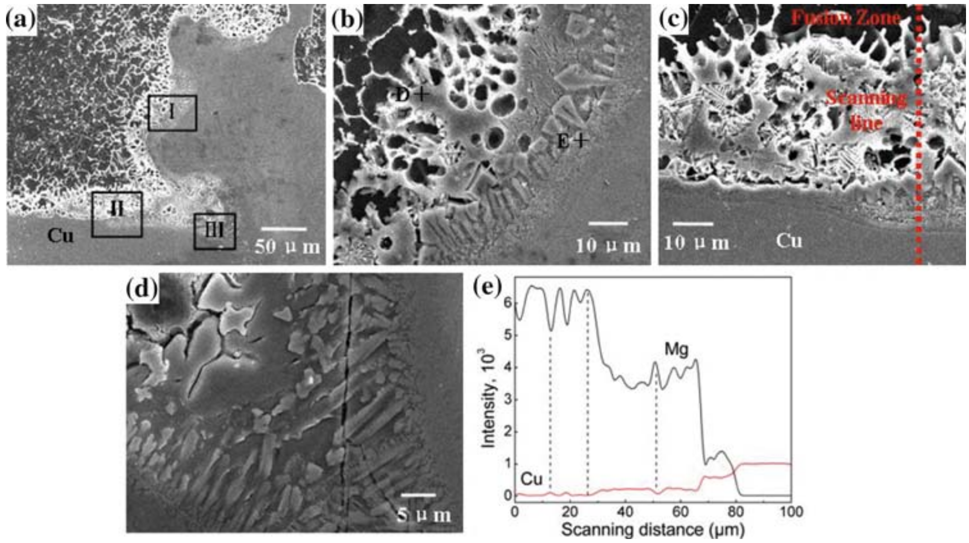


Fig. 4 Interfacial microstructure of the joint between Mg alloy and Cu interlayer and element distribution: **a** region A in Fig. 3; **b**, **c**, and **d** are regions I, II, and III from (**a**), respectively; **e** line analysis along scanning line of (**c**)

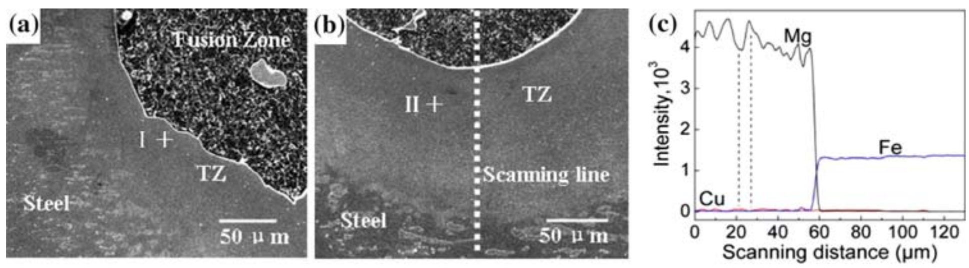


Fig. 5 Interfacial structure of the joint between Mg alloy and steel and elements distribution: **a** and **b** are from region B and D of Fig. 3, respectively; **c** line analysis along the scanning line of (**b**)

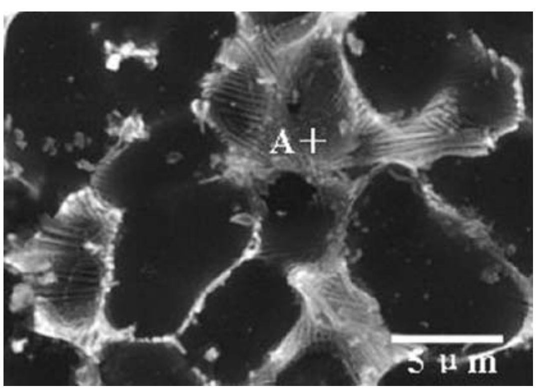


Fig. 6 Microstructure of fusion zone from region C of Fig. 3

composed of Mg_2Cu. The content of Mg_2Cu is smaller than that of Mg and Cu phase, as the intensity of it is much lower. Cu interlayer was attached to the Mg alloy sheet after tensile test; therefore it is inevitable to be examined in the spectrum. Figure 7b is the XRD pattern of steel side. The peak intensity of Fe phase is much higher than that of other phases, thus Fe is the dominant phase. Some remaining Mg and Mg_2Cu phases were also shown but not much according to their respective intensity. However, the solid solution of Cu in Fe were not found, which appears to contradict with the aforementioned EDS analysis. As it is known, the phases that are less than 5 wt% are not detectable in XRD spectrum, indicating that the content of solid solution of Cu in Fe is quite low. Thus the TZ is composed of a majority of remelted steel and a little solid solution of Cu in Fe.

Discussion

Factors affecting the joint strength

Intermetallic compounds

The joint strength of Mg alloy/steel with Cu interlayer was improved significantly, which could reach 158 nearly 100% that of base material Mg alloy, when comparing with that of direct joining of Mg alloy/steel at about 90 MPa [8]. The grains in fusion zone of the joint without any interlayer are usually fine after welding as shown in Fig. 8a; however, they become even finer as the new phase generated along the grain boundary shown in Fig. 6 with the Cu interlayer added. Firstly, plenty of Mg_2Cu phases produced in fusion zone indicates that Mg reacted with Cu, which led to the consumption of Mg element. Secondly, as Mg almost does not dissolve Cu in terms of Mg–Cu binary phase diagram, and it is hard for IMC Mg_2Cu to be found inside the crystals of Mg alloy, suggesting that the phase could only be formed at the grain boundary. Thirdly, the IMC

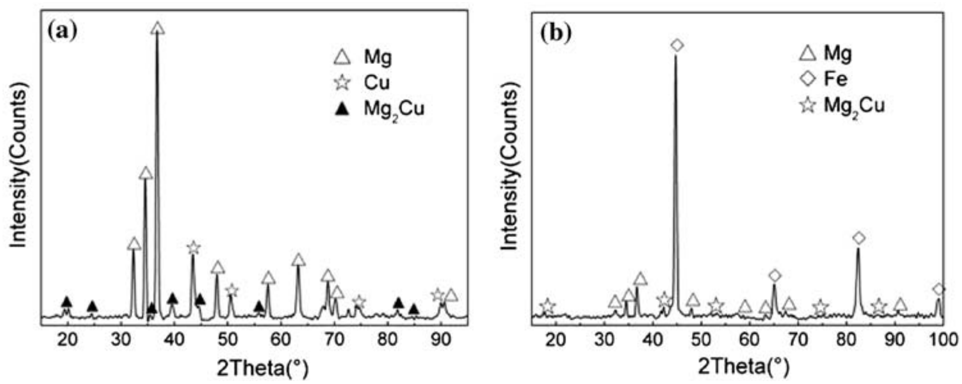


Fig. 7 XRD spectra from welding seam: **a** Mg alloy with Cu interlayer; **b** steel side with fusion zone remained

morphology with thin rod-like structure aligned continuously may block movements of intergranular when undergoing tensile test. Thereby it can be seen that the intermetallic phase plays a key role in strengthening the joint strength, in other words, Cu can be seen as a reinforcing element [11, 12].

Transitional zone

At the interface of Mg/steel, a transitional zone (TZ) that is composed of a little solid solution of Cu in Fe [13] is formed. TZ is a key bridge for the bonding of Cu and steel. Firstly, the TZ was attached to the steel intimately at the bottom of molten pool as there is no distinct boundary between them. Secondly, the TZ shows a good mixture of Cu in Fe at both upper ends of molten pool. For instance, in Fig. 9 one of the ends of TZ shows a nice mixture without any defect. In addition, the solid solution of Cu in Fe can provide a strengthening effect [14] for the steel in TZ. Therefore, the steel and Cu were bonded successfully through TZ.

Wettability of Mg alloy

Mg element was not found in TZ according to EDS analysis and elements distribution of Fig. 5c, which means that the diffusion process of Mg element into TZ did not happen. Although Cu existed both in fusion zone and TZ, there was not any IMC layers formed like that in Fig. 4 distributed at fusion zone/TZ interfaces. However, it can also be seen that the fusion zone bonded with the TZ intimately without any gaps. Such type of bonding may correlate to the wettability of molten Mg alloy on steel during solidification.

A sessile drop method was conducted to investigate the wettability of molten Mg alloy AZ31B on steel, approximately 15 wt% of Cu which was estimated according to that melted in fusion zone was added into AZ31B Mg alloy. The results are shown in Fig. 10 that wetting angle $\theta_1 < \theta_0$ evidently. The wetting angle is often used to describe wettability of liquid and solid phase[15], and the lower the wetting angle, the better the wettability of molten Mg alloy. In addition, the bonding force between the liquid and the solid phase is the work of adhesion W_a that is defined as [15]

$$W_a = \gamma_{l-g}(1 + \cos\theta).$$

Thus the bonding force can be expressed in terms of wetting angle θ. And the bonding force of Mg alloy with 15% of Cu would be higher than that of direct joining. The results of wetting angle were verified by Ref. [16] that Cu can well wet on steel, the addition to Mg alloy of Cu improved the wettability of molten Mg alloy [12] on steel. Finally, an intimate bonding between fusion zone and TZ shown in Fig. 3 was then attained.

A gap shown in Fig. 8b which was a direct joint between Mg alloy and steel without any interlayer suggested that the

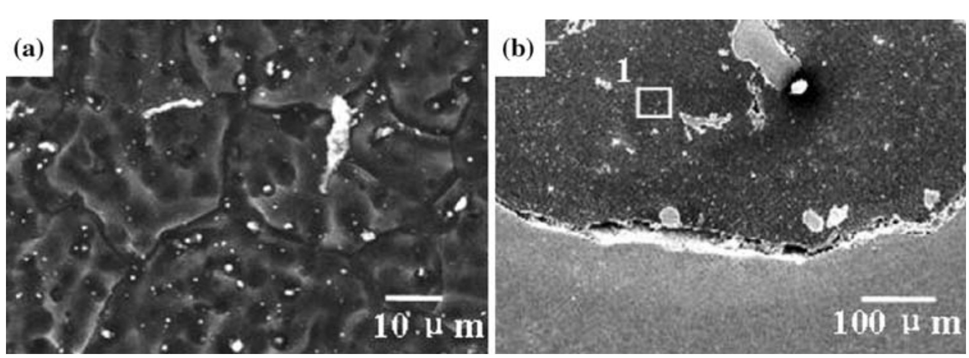


Fig. 8 Microstructure of the joint without interlayers: **a** fusion zone from region 1 of (**b**); **b** bottom of molten pool

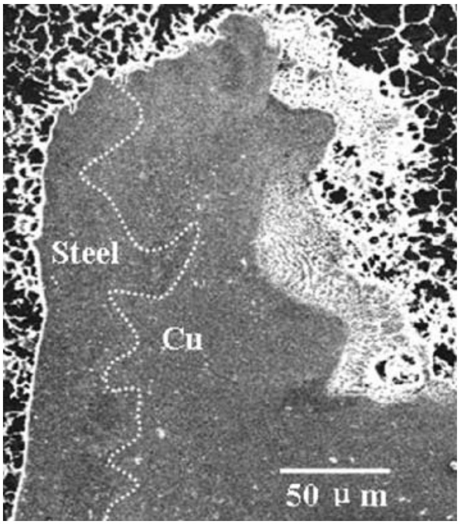


Fig. 9 Mixing area from region F of Fig. 3, the *dotted line* represents the mixing line

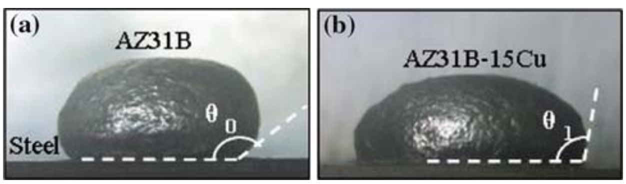


Fig. 10 Wetting angles of different materials: **a** an AZ31B drop alone, **b** the drop of AZ31B with Cu. These drops were cooled naturally to simulate the conditions of welding process as possible as they can after stable drops were attained, and were shielded by the atmosphere of argon gas

wettability of molten Mg alloy was so low that molten Mg alloy cannot nucleate well on the steel surface during solidified process, and thus the bonding force was lower than that of Cu-added joints. Accordingly, the wettability of molten Mg alloy on steel is a key factor that determines the bonding strength between fusion zone and TZ.

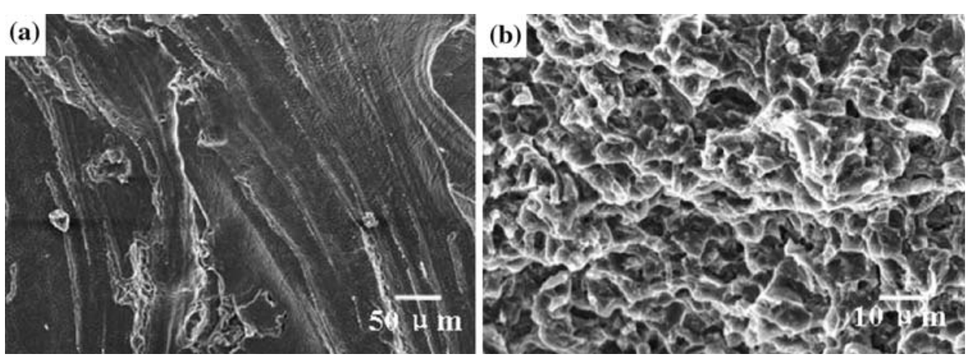
Phases formation in the joint microstructure

The formation of Mg_2Cu phase is a dominant intermediate phase of fusion zone and is a key factor in the improvement of joint shear strength. The Mg–Cu phase diagram shows that there is also another IMC $MgCu_2$. However, XRD spectrum demonstrates that Mg_2Cu was the dominant phase in the present experiment. Firstly, the energy provided by hybrid laser-TIG is sufficient to melt Cu and Mg; secondly, molten Mg alloy accounts for approximately 90 wt% in present experiment, comparing with molten Cu about 10 wt% in the joint, which is in favor of the formation of Mg_2Cu [17]. In addition, Hong et al. [18] reported that the Mg_2Cu phase is the only product without IMC $MgCu_2$ during the rapid cooling process. Hassan et al. [19] also mentioned that Mg_2Cu was the only IMC in Mg17.2Cu (wt%) alloy. According to thermodynamic calculations of standard free energy of formation on the two phases [20], the possibility for the reaction of Mg_2Cu product is much higher than that of $MgCu_2$ phase in the present composition range of 10–20 wt% Cu. Therefore, in the present welding process, Mg_2Cu forms preferentially in the exothermic reaction $Mg + Cu \rightarrow Mg_2Cu + Q$, where Q is the released heat that may contribute to the reduction of temperature difference on which the formation of columnar particles depends in Fig. 4d. Besides, fractography is shown in Fig. 11. The fracture surface of the direct joint with river-shape pattern and smooth surfaces in Fig. 11a indicates that they are completely brittle. While the surface in Fig. 11b displays some dimple features and a large number of torn arises, suggesting that the IMC Mg_2Cu in the Cu-added joint played a significant role in strengthening the microstructure of the joint, and thus improved the joint strength. Consequently, the addition of Cu contributes to the formation of reinforced phase and improves the microstructure in the joint.

Joining mechanism of Mg alloy and steel with Cu interlayer

The IMC Mg_2Cu as the dominant phase forms both at interface of Mg alloy/Cu and in fusion zone. A successful joining of Mg alloy and Cu were obtained. The transitional zone (TZ) containing a little solid solution at the bottom of molten pool and physical mixture of steel and Cu at upper

Fig. 11 Fracture surfaces of **a** direct joint and **b** Cu-added joint. Fracture of direct joint came from the location where the gaps at the interfaces shown in Fig. 8b, and the Cu-added joint fracture is from across fusion zone shown in *circle* region of Fig. 2c



margins of the pool marks a validated joining of steel and Cu. The joining mechanism is proposed below. Under the action of hybrid laser-TIG welding process, molten Mg alloy reacts with Cu to form Mg_2Cu phase in both Mg alloy/Cu interlayer interfaces and fusion zone, and wets on steel well due to the addition of Cu; meanwhile, a little of molten Cu dissolves into molten Fe at the bottom and side of molten pool, and the molten steel at upper margins of molten pool blends with Cu interlayer substantially.

Conclusions

Lap joint of Mg alloy AZ31B to mild steel Q235 with Cu interlayer can be successfully constructed by hybrid laser-TIG welding technique. The maximum shear strength of the joint can achieve almost 100% that of Mg alloy AZ31B, comparing with that of direct joint without any interlayer. The Mg_2Cu phases generated and precipitated in grain boundaries with fine rod-like structure not only refine grains but strengthen the grain boundaries. The transitional zone (TZ) is consisted of two parts, one is the majority of remelted steel including a little solid solution of Cu in Fe distributed along the edge of the molten pool at steel side, and the other is the mixture of Cu and Fe on the upper margins of the pool. Such distribution of TZ makes sure the contact of Cu with steel. The reason that the fusion zone attached to the TZ tightly is that the addition of Cu improves the wettability and facilitates its nucleation on steel. Consequently, it can be said that the addition of Cu interlayer not only joins Mg alloy and steel together, but contributes to the improvement of strength effectively.

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