Rapid Ultrasonic-Assisted Soldering of AZ31B Mg Alloy/6061 Al Alloy with Low-Melting-Point Sn–xZn Solders Without Flux in Air

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Abstract

A novel ultrasonic-assisted low-temperature soldering was developed to join AZ31B Mg alloy and 6061 Al alloy with a series of Sn–xZn solders. The average maximum shear strength of the joints reaches up to 87.5 MPa at soldering temperature of 300 °C under ultrasonic assistance for only 5 s using Sn–20Zn solder. The fracture path propagates completely in the soldering seam. The results indicate that the microjet generated by ultrasonic pressure in liquid solder could strike and splinter the Mg₂Sn intermetallic compounds into small pieces, which contributes to the enhancement of the joint strength. In addition, the primary Al(Zn) solid solution phase formed during cooling stage could also strengthen the joint due to the prevention of microcracks propagation.

Keywords Ultrasonic-assisted soldering · Ultrasonic effect mechanism · Microjet · Intermetallic compound distribution · Solid solution · Joint strengthening

1 Introduction

Magnesium (Mg) and aluminum (Al) alloys, compared with conventional metal materials, have some unique advantages such as low density, good high strength-to-weight ratio, dimensional stability, machinability and low

casting costs. Hence, it has gained great attention in academic research as well as industrial applications [1, 2]. Joining of Mg alloy/Al alloy dissimilar materials could not only optimize material application but also reduce the weight of the structures. Therefore, the investigation of these alloys joining is significant [3].

A variety of joining methods, such as tungsten inert gas welding (TIG) [4], friction stir welding [5], diffusion bonding [6, 7] and explosive welding [8] have been reported to join Mg and Al alloys. However, Mg–Al intermetallic compounds (Al₃Mg₂, Al₁₂Mg₁₇, etc.) will be inevitably formed in the joint because the temperature during joining is higher than that of Mg–Al intermetallic reaction point. To some extent, the microcrack initiation and propagation will appear near the continuous brittle Mg–Al intermetallic compounds (IMCs), which will seriously deteriorate the mechanical property of Mg/Al joint [9–11]. Therefore, it is necessary to avoid the formation of Mg–Al IMCs by reducing the joining temperature lower than the Mg–Al intermetallic reaction point. Compared with those methods above, brazing or soldering has many advantages in joining Mg–Al, due to its variable solders and bonding temperature selection [12–14]. Wang et al. [15] has designed a series of Sn–xZn solders for joining Mg/Al dissimilar metals. In their study, no Mg–Al IMCs

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were observed in brazing seam by using Sn–30Zn solder at 360 °C. However, the Al alloy sheet had to be pretreated by hot-dipping technique to be combined with the solders, and the soldering process had to be conducted with the protection of argon gas in a tubular resistance furnace, making this process complex and thus limiting its industrial application.

Compared with conventional vacuum brazing, ultrasonic-assisted joining methods (brazing, soldering and transient liquid phase bonding, etc.) could be carried out in air ambient, because the “ultrasonic effect” is able to remove the oxide without vacuum and flux and it also can accelerate the reaction rate effectively [16–18]. These methods have been widely studied in the lightweight materials such as Al alloy/Al matrix metal composites (MMCs) [18–20], Mg alloy/Mg MMCs [21, 22], and excellent joint properties have been achieved. Huang et al. [22] designed a novel Zn/Al/Zn compound interlayer to join SiC particles-reinforced Mg MMCs using ultrasonic-assisted transient liquid phase bonding method, where the bonding seam completely translated into full Mg solid solution and the joint strength reached up to 87.5% of base material strength. During the joining of the same material, we hope to obtain the joint microstructure more similar to the base material in order to ensure the consistency of mechanical property or other function properties (electrical, corrosion resistance, etc.) of the joints.

However, for the dissimilar metals joining, it is difficult to obtain the joint microstructure that is the same to any one of the base materials. It is more significative and feasible to focus on optimization of the phase morphology and the distribution of IMCs in the joint. In other words, we need to eliminate the harmful IMCs or smash them from continuous layer into small pieces to reduce the risk of brittle fracture. Fortunately, it has been reported in our previous studies that [23] high-strength Mg–Al joint was obtained using pre-coated Zn–Al solders on the Al sheet for a short time of ultrasonic assistance (3 s) at 340 °C. This research provided a novel and efficient method to join Mg/Al dissimilar metals. However, there are still some aspects needed to be further improved and studied: (1) These bonding temperatures are still relatively high which might induce the residual stress concentration and the reduction of base material strength after the soldering thermal cycle [24]; (2) The mechanism of ultrasonic effect in joining process, especially the affection of ultrasonic in distribution of IMCs morphologies, is uncertain.

Therefore, in this study, a lower melting-point Sn–xZn solder with the base element Sn and the additional element Zn with 9, 20 and 30 wt%, respectively, was designed. The Sn-based solders are suitable to join temperature-sensitive alloys having low melting point [25], and the addition of Zn can strengthen the interfacial bonding due to high

mutual solubility of Al [26]. The Al/Mg dissimilar alloys are rapidly joined with the assistance of ultrasound, and the optimized solder is selected based on the joint strength. The effects of ultrasonic assistance and joint formation mechanism were investigated based on the optimized solders by controlling varied ultrasonic assistance time.

2 Experimental

The base materials are AZ31B Mg alloy and 6061 Al alloy sheets with dimension of 45 mm × 10 mm × 3 mm. The chemical compositions of base materials are shown in Table 1. The sample surfaces were ground with SiC sand paper to 1000# and ultrasonically cleaned with acetone for 15 min before soldering. The schematic illustration of the ultrasonic-assisted soldering process is shown in Fig. 1a. Mg alloy sheet was placed on the top of Al alloy sheet with 3 mm overlapping length and 200 µm gap. The solder was placed on the edge of the overlapping zone on the bottom of Al alloy sheet. During the soldering process, the ultrasonic horn was loaded on the Al alloy sheet with a constant pressure of 0.15 MPa. The frequency and amplitude of ultrasound are 20 ± 0.1 kHz and 10 µm, respectively. High-frequency induced heating device was used to heat the soldering system, and the heating speed is $20\text{ }^{\circ}\text{C min}^{-1}$. The parameters used in the process are shown in Fig. 2. The soldering parameters are divided into two processes: (I) the samples soldered with pure Sn, Sn–9Zn, Sn–20Zn and Sn–30Zn are under the ultrasonic assistance for 5 s after heated to 260, 230, 300 and 340 °C, respectively; (II) the samples soldered with Sn–20Zn are under ultrasonic assistance for 1, 3, 5, 10 and 15 s, respectively, after heating to 300 °C.

The cross sections of the joints were cut and prepared using a standard polishing technique. The microstructures were observed using scanning electron microscopy (SEM, Quanta 200F) equipped with an energy-dispersive X-ray spectroscopy (EDS) analysis system. The phase constitutions of the joint were analyzed using a D/max 2500 X-ray diffraction (XRD). The shear strength of the joints was tested using electronic universal testing machine (ZWICK-Z020) at a loading speed of 2 mm min^{-1} . The testing schematic diagram of which is shown in Fig. 1b.

Table 1 Chemical compositions of AZ31B Mg alloy and 6061 Al alloy (wt%)

Base metal	Al	Si	Zn	Mn	Mg
AZ31	2.5–3.5	0.1	0.5–1.5	0.2–0.5	Bal.
6061	98	0.5	0.0005	0.0038	0.97

Fig. 1 Schematic diagrams of ultrasonic-assisted soldering process **a** and shear strength test **b**

3 Results and Discussion

3.1 Soldering Seam Microstructures and Mechanical Properties with Different Zn Contents

The SEM images of the joint soldered with a series of Sn- x Zn solders for 5 s ultrasonic assistance of the soldering process I are shown in Fig. 3. The widths of joints soldered with pure Sn, Sn-9Zn, Sn-20Zn and Sn-30Zn are 200, 214, 240 and 293 μm , respectively. The increase in the joint width is mainly attributed to the increasing dissolution of Mg and Al base metal into the liquid solder. The phase existing in the pure Sn soldered joint (Fig. 3a) is mainly Sn-Mg eutectic phase with some Mg_2Sn blocky phase. There is no Al-based phase observed in the soldering seam, because Al element only has solid solubility of 1 at.% in Sn at 225 $^\circ\text{C}$ according to Al-Sn binary phase diagram [27]. This solubility is too small to form unique Al-based

phases. It has been reported that the pure Sn was used to solder 2024 Al alloy at 300 $^\circ\text{C}$ but the bonding at the interface is weak [28]. On the Mg side, there is a continuous thin layer of Mg_2Sn phase. As the 9 wt% Zn is added into the solder (Fig. 3b), the needle η -Zn phase appeared and the black spherical Al(Zn) solid solution phase emerged in the joint, which indicates that the addition of Zn promoted the dissolution of Al.

Figure 3c shows the microstructures of joint soldered with Sn-20Zn. Some blocky Mg_2Sn phases form in whole soldering seam, indicating that more Mg is dissolved and reacts with Sn. With the increase of Zn, the η -Zn phase grows up and the amount of Al(Zn) solid solution increases and migrates into the whole soldering seam. Figure 3d shows the microstructures of the joint soldered with Sn-30Zn at 340 $^\circ\text{C}$. A large amount of Mg is dissolved into the soldering seam, forming the continuous Mg_2Sn phase. The Al(Zn) solid solution further increases, and the Al grain boundaries are eroded. As shown in Fig. 4, the shear strength of the joints with pure Sn was only 44.3 MPa. As Zn content increases to 9 wt%, the shear strength of the joint increases up to 66.8 MPa. It is significant that the maximum strength of 87.5 MPa is obtained when the Sn-20Zn solder is used to joining Mg-Al at 300 $^\circ\text{C}$ without flux in air ambient. Compared with other methods reported, this joint strength of Mg-Al joints is higher and the joining temperature is much lower (joint shear strength $\sigma = 70.73$ MPa; temperature $T = 360$ $^\circ\text{C}$; Sn-30Zn; argon gas protection ambient) [15]. However, the shear strength of the joints soldered with Sn-30Zn solder drops to 56.9 MPa. Therefore, it can be concluded that the Sn-20Zn alloy is relatively much more suitable to be used to join Mg-Al structures due to the high joint strength obtained and moderate soldering temperature used.

Fig. 2 Schematic diagrams of technological process parameters for different Zn contents of Sn- x Zn ($x = 0, 9, 20, 30$, wt%) solders **a** and varied ultrasonic assistance time using Sn-20Zn solder **b**

Fig. 3 Microstructures of joint with different Zn contents: **a** pure Sn (260 °C); **b** Sn-9Zn (230 °C); **c** Sn-20Zn (300 °C); **d** Sn-30Zn (340 °C)

Fig. 4 Shear strength of Mg/Al alloy joint with a series of Sn-*x*Zn solders

3.2 Primary Analysis of Ultrasound Assistance Effect on the Soldering Seam Microstructures

In order to study the ultrasonic effect on the microstructures of the soldering seam, the soldering process II using Sn-20Zn solder was carried out under the ultrasonic assistance for 1, 3, 5, 10 and 15 s, respectively. As shown

in Fig. 5a, the major phases of soldering seam are Sn-Zn eutectic structure and η -Zn solid solution. The matrix surface oxide films and air pores are completely removed by the ultrasound assistance for 1 s. Although the breaking behavior of oxide films is not directly observed, the removal mechanism should be similar to our previous study of ultrasonic-assisted brazing for other materials [17, 21–23]. The Mg-Sn intermetallic reaction happens as soon as the liquid solder drills into the seam under the effect of ultrasonic pressure; then, a continuous Mg_2Sn layer is generated attaching to AZ31 Mg alloy.

Figure 5b shows the joint microstructure under the ultrasonic assistance for 3 s. It can be seen that the Mg_2Sn layer starts to shrink and collapse, resulting in the wavy and fragmentary shape on the interfaces. When prolonging ultrasonic assistance time to 5 s, the Mg_2Sn layer is disintegrated into much smaller fragments and homogeneously distributed in whole soldering seam as shown in Fig. 5c, which is mainly attributed to the “acoustic cavitation effect.”

The acoustic pressure could be established while the ultrasonic wave transmits across the materials. Since the atoms or molecules in liquid phase or gas phase can move around with little restrain, some micro-region could be torn out, forming an instantaneous bubble when the ultrasonic rarefaction arrives as shown in Fig. 6a. Soon afterward, the bubble rapidly collapses as compression arrives. The

Fig. 5 Microstructures of joints with Sn–20Zn interlayer soldering under different ultrasonic assistance time: **a** 1 s; **b** 3 s; **c** 5 s; **d** 10 s; **e** 15 s

bubble implosion can result in a tiny hot spot. The transient temperature of the liquid surrounding the bubble can reach 1600 °C [29]. Even though the temperature of this spot is extremely high, the tiny volume and transitory duration time of the bubble itself could release heat quickly. When the bubble is closed to the solid phase surface, the bubble would be distorted from spherical to elliptic shape because of the asymmetric pressure at the solid–liquid interface as shown in Fig. 6b, c. This distortion forced the liquid phase to form the microjet, which rushes through the bubble, penetrates the opposite bubble interface and strikes solid surface violently [16, 17, 30, 31] as shown in Fig. 6d.

On the Mg alloy side interface, a homogeneous Mg_2Sn IMCs layer is formed (Fig. 7a) under the ultrasonic assistance time for 1 s and the Al alloy side interface is flat. Further increasing ultrasonic assistance time to 5 s, the Mg_2Sn IMCs layer is struck and stripped out by liquid microjet as shown in Fig. 7b, indicating that the ultrasonic wave influences the local environment at the interface between IMCs and liquid solders. In comparison with conventional brazing, the ultrasonic wave assistance could be used as an extra method to intervene the Mg–Sn IMCs formation and distribution. The microjet incises and smashes the large continuous IMCs layer into small fragments meanwhile and scatters them into the whole soldering seam. That is beneficial to the joint mechanical properties through the optimization of the brittle IMCs distribution as well as through decrease of the residual stress concentration, especially in the large surface or/and complex structure joining.

However, when applying ultrasonic assistance time for both 10 and 15 s (Fig. 5d, e), the cracked Mg_2Sn IMCs become thicken and are filled in almost the entire soldering seam, indicating that too long time of ultrasonic assistance would promote IMCs formation. As shown in Fig. 7c, an increased number of Mg_2Sn scatter into soldering seam, forming large continuous IMCs when ultrasonic assistance is applied for 15 s. Since the Sn content is sufficient, the Mg_2Sn could be rapidly formed once Mg contacts Sn. Meanwhile, the microjet constantly strips Mg_2Sn out of the Mg_2Sn layer and moves them away from Mg alloy side by the acceleration of the convection of substance. With the formation of more Mg_2Sn IMCs, a lot of space will be filled until the final recrystallization.

A few solder liquid phase and Al(Zn) solid solution are sealed into several Mg_2Sn grains, and these Mg_2Sn IMCs do not form all at once rather than recrystallization when they cool down. Owing to the ultrasonic assistance, the distribution and morphology of these IMCs could be manipulated by controlling ultrasound input time so as to avoid harmful influence of joint mechanical property. The EDS analysis results of possible phases in Fig. 7 are shown in Table 2.

On the Al alloy side interface, the Al(Zn) solid solution is formed near Al side interface as shown in Fig. 7d. A Zn diffusion layer appears on the surface of Al alloy and becomes thick with the increase of ultrasonic time (Fig. 7e). According to the Al–Zn binary phase diagram, the saturation solid solution of Al in Zn was 67 at.% at 300 °C [25]; hence, high Zn content in the solder can

Fig. 6 Diagrams of acoustic cavitation effect: **a** formation process of acoustic bubble; **b** compression process of acoustic bubble; **c** distortion process of acoustic bubble; **d** collapse process of acoustic bubble

accelerate the Al grain boundaries dissolution. On the one hand, with the erosion of Al grain boundaries, the large amount of Al is dissolved into the liquid Sn–20Zn solder at elevated temperature. The Al is supersaturated during cooling down and precipitated out as tiny and blocky phase which can be seen in Fig. 7e. On the other hand, the large Al(Zn) solid solutions are released from the Al alloy surface and migrate into the soldering seam (Fig. 7f).

3.3 Joint Mechanical Properties and Fracture Behavior

The fracture paths of the joints under varied ultrasonic assistance time are mainly classified into two types: (1) fracture into Mg_2Sn IMCs layer and (2) fracture into soldering seam center. As can be seen in Fig. 8a, c, the joints under ultrasonic assistance for 1 and 15 s completely fracture within the Mg_2Sn layer, indicating that these brittle IMCs are the weak region. The large area of Mg_2Sn IMCs and secondary microcracks can be observed on the fracture

Fig. 7 Magnified microstructures of joints at Mg **a–c** and Al **d–f** alloys side interface with ultrasonic assistance time of 1 s **a, d**, 5 s **b, e** and 15 s **c, f**

Table 2 EDS results taken from possible phases (at.%)

Possible phase	Mg	Al	Sn	Zn
Sn	4.0	4.2	88.9	2.9
η -Zn	1.09	1.91	3.43	93.57
Mg ₂ Sn	66.9	1.7	27.9	3.5
Al(Zn) precipitated phase	0.8	87.9	00.4	10.6
Al(Zn) solid solution phase	0.88	92.23	1.2	6.22

surface as shown in Fig. 9a, b, e, f, which indicates that these IMCs are characteristically brittle in the Mg–Sn binary system. The average shear strengths of the joints under ultrasonic assistance for 1 and 15 s are 73.6 and 54.2 MPa (Fig. 10a), respectively. The XRD pattern (Fig. 10b) shows that all the fracture surfaces contain Mg₂Sn IMCs. The large residual stress exists since the

expansion coefficient of IMCs is much smaller than that of the base materials (Al: $23 \mu\text{m m}^{-1} \text{K}^{-1}$, Mg: $25 \mu\text{m m}^{-1} \text{K}^{-1}$) [11]. Thus, when cooling to room temperature, the interface is imposed a tensile stress between Mg₂Sn and Mg alloys. These brittle IMCs are torn out by the other side of parent material.

However, as shown in Fig. 8b, the fracture path of the joint under ultrasonic assistance for 5 s completely propagates across the soldering seam center, which could be ascribed to the translation of Mg₂Sn IMCs morphology from continuous layer into irregular blocky phase. The average shear strength of the joint reaches up to 87.5 MPa. As shown in Fig. 9c, d, the fracture surface is mainly consisted of the soldering interlayer microstructures and partial Mg₂Sn IMCs as well as a small number of Al(Zn) solid solution. It can be concluded that the distribution and morphology of Mg₂Sn IMCs determine the mechanical property and fracture behavior of the joints. The continuous

Fig. 8 Joint fracture paths under varied ultrasonic assistance time: **a** 1 s; **b** 5 s; **c** 15 s

Fig. 9 Secondary electron **a, c, e** and backscattering electron **b, d, f** images of Sn-20Zn soldered joint fracture surface with ultrasonic assistance time of 1 s **a, b**, 5 s **c, d**; 15 s **e, f**

layer of IMCs is corresponded to lower strength and brittle fracture, while the dispersive blocky IMCs decreasing the stress concentration are corresponded to the higher strength and plasticity fracture.

Another joint strength reinforcement factor is derived from the Al(Zn) phases, which are mainly existed as two forms as shown in Fig. 11a: (1) the tiny and blocky Al(Zn) precipitated phase and (2) the large spherical Al(Zn) solid solution. The Al(Zn) precipitated phase could strengthen the soldering seam. It has been reported by Li et al. [28] that with applying ultrasonic time for 60 s, some Al precipitated particles could migrate from the Al alloy substrate

into the bonding seam; hence, the shear strength of joint could be improved from ~ 40 MPa to ~ 60 MPa with the increase of Al particles. Furthermore, the large spherical Al(Zn) solid solution plays an important role in the reinforcement of strengthening phase interface. As shown in Fig. 11b, the Al(Zn) solid solution in the brittle Mg_2Sn IMCs deflects and prevents the propagation of microcracks across the phase interface. It might be attributed to the hindering of the mobility of dislocations and the releasing of stress concentration at the crack tip by the Al(Zn) solid solution. Zhang and Urbassek [32] studied the dislocation-based strengthening mechanisms of spherical

Fig. 10 **a** Joint shear strength and **b** XRD patterns of joint fracture surface with different ultrasonic assistance time

Fig. 11 Morphologies of Al(Zn) phase distribution **a** and microcrack propagation behavior **b**

reinforcement and pointed out that: (1) The reinforcement influences the direction of motion of the dislocations; (2) the interface swallows part of the dislocations that approach the interface.

3.4 Model of Ultrasonic Assistance Effect on Joint Formation

Based on the microstructural observation and analysis, the mechanism of ultrasonic assistance effect on joint formation is summarized in the schematic diagram as shown in Fig. 12. The soldering temperature is constant lower than eutectic reaction points of both Al–Mg and Mg–Zn. The intermetallic reaction only exists in Mg–Sn binary system. The Sn–Zn eutectic phase still remains in abundance after soldering process according to actual microstructure morphology. As shown in Fig. 12a, a continuous layer of Mg₂Sn IMCs is immediately formed once the liquid solder drills into the soldering gap at the initial stage of ultrasonic assistance for 1 s. With Sn content assuming, the residual

Sn–Zn eutectic phase separated out clubbed η-Zn solid solution and translated into tiny lamellar Sn–Zn eutectic phase when cooling down, the reaction formula is as follows:

where I–Sn–Zn_{eutectic phase} represents supersaturated Zn content Sn–Zn eutectic phase and II–Sn–Zn_{eutectic phase} represents tiny lamellar Sn–Zn eutectic phase after separating out η-Zn.

The continuous Mg₂Sn IMCs layer should be thickened as a result of constant sufficient diffusion [8, 11]. However, with prolonging ultrasonic assistance, these layered IMCs are struck by the microjet and splintered into small pieces scattering in liquid solder. The Al alloy side interface becomes wavy because of the erosion of grain boundary, which is ascribed to a certain amount of Zn content as

Fig. 12 Joint formation model of ultrasonic-assisted soldering: **a** initial state of solders and base materials; **b** disintegration of layered Mg_2Sn IMCs by microjet; **c** dispersive distribution of blocky Mg_2Sn IMCs under ultrasound; **d** secondary crystallization of Mg_2Sn IMCs

shown in Fig. 12b, c. Therefore, the tiny and blocky $\text{Al}(\text{Zn})$ precipitated phases separate out when cooling down. Meanwhile, the large amount of $\text{Al}(\text{Zn})$ solid solutions are released out of the Al alloy surface, which are migrated to the whole soldering seam by ultrasonic convection. The ultrasonic effect changes the local environment of the interface between Mg alloy solders and Al alloy solders, which not only refines and scatters brittle Mg_2Sn IMCs but also optimizes the Al dissolution.

Further increasing the time of ultrasonic assistance, the Mg_2Sn IMCs are constantly splintered out. However, since the Sn content is sufficient, the Mg_2Sn IMCs are formed rapidly once Mg alloy contacts the solder. If the Mg_2Sn IMCs formation and transition process proceed too long, it could finally fill in the soldering seam. The large amount of Mg_2Sn IMCs will recrystallize forming a net structure (Fig. 12d) and deteriorate the joint strength.

4 Conclusion

The dissimilar AZ31B Mg alloy and 6061 Al alloy were rapidly soldered with $\text{Sn}-x\text{Zn}$ solders at low soldering temperature under the ultrasonic assistance. The Mg_2Sn IMCs layer, formed on the Mg side interface, is the weak region of the joints. The $\text{Al}(\text{Zn})$ solid solution formed on the Al alloy side interface results in a reliable bonding. With the increase of ultrasonic assistance time, the Mg_2Sn IMCs morphologies changed from continuous layer to dispersing block layer, which enhances the mechanical properties of the joints. The maximum strength of the joints reaches up to 87.5 MPa when they are soldered at temperature of 300 °C under ultrasonic assistance for only 5 s using $\text{Sn}-20\text{Zn}$ solder.

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