Microstructure of Diffusion-Bonded Mg-Ag-Al Multilayer Composite Materials

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Abstract: Mg-Al bonded composite materials expand Mg and Al alloys’ applications by combining their unique performances together. However, the formation of Mg-Al intermetallic compounds in interface zone of Mg/Al directly-bonded joint seriously obstructs its further development. To solve this problem, Mg-Ag-Al multilayer composite materials have been successfully prepared by diffusion bonding technology. The effect of key process parameter (bonding temperature) on microstructure of this material has been mainly investigated. The results show that Mg and Al were well bonded by using silver interlayer when the bonding temperature exceeded 370°C. But Mg17Al12 and Mg2Al3 compounds were formed in the interface zone at temperatures higher than 420°C. By means of controlling the bonding temperature (380 °C–420 °C), silver interlayer effectively restrained the generation of Mg-Al intermetallic compounds, and Mg-Ag intermetallic compounds \( (Mg3Ag, MgAg) \) were formed in the interface zone instead.

1. Introduction
Welding composite materials play an important role in manufacturing and construction industries for their unique performances [1, 2]. Mg/Al bonded composite materials with their excellent properties together, such as low density, high strength and high corrosion resistance have been widely used in many fields, especially in aerospace, automobile and electronic products by largely reducing the weight of the structural components and saving the costs [3, 4].

Diffusion bonding method is an efficient way to produce composite materials, which has been widely used in bonding dissimilar metals [5-7]. However the formation of brittle Mg-Al intermetallic compounds at the interface of directly-bonded Mg/Al joint has become an obstacle in its development [8, 9]. Interlayers such as Zn alloy interlayer and Ti interlayer inserted between Mg and Al have been proved to be an efficient way to solve the problem above by blocking the inter-diffusion of Mg and Al atoms [10, 11]. Silver interlayer with high ductility and good metallurgical property has been successfully used in welding similar and dissimilar metals, even ceramics [12, 13]. Bonding temperature as the most important parameter in diffusion bonding process has a great effect on the microstructure and mechanical properties of the joints [14, 15].

In the present work, Mg and Al metals have been successfully jointed at different bonding temperatures by using silver interlayers. The effect of bonding temperature on the variation in the microstructure of diffusion-bonded Mg-Ag-Al multilayer composite materials has been investigated particularly, which provides an important guidance for understanding the fundamental law of welding science deeply.

2. Experimental procedures
Pure magnesium Mg1 and commercial pure Al 1060 plates with dimension of Φ25mm×5mm were used in this work. Table 1 shows the chemical composition of the materials above. Pure silver (>
99.99%, wt %) was used as the magnetron sputtering target material.

Table 1: Chemical composition of Mg1 and 1060Al

| Materials | Mg | Al | Cu | Fe | Si | Zn | Ti | Mn | Ni |
|------------|----|----|----|----|----|----|----|----|----|
| Mg1        | Bal| 0.0056 | 0.0007 | 0.0025 | 0.012 | 0.003 | 0.005 | - | 0.0006 |
| Al 1060    | 0.02 | Bal | 0.03 | 0.05 | 0.10 | 0.02 | 0.01 | 0.005 | - |

In all cases, Mg1 and Al 1060 were ground flat by grit SiC paper to 2000# and a diamond polishing agent to 0.05 μm, and then cleaned in ethanol and acetone before being silver sputtered. Then silver interlayers were deposited onto the clean surfaces of Mg1 and Al 1060 using magnetron sputtering technology. The thickness of the silver interlayer on each side is 2.5 μm. Then these samples were fitted together and introduced into a vacuum furnace (10⁻⁴–10⁻³ Pa). To investigate the effect of bonding temperature on microstructure of the Mg-Ag-Al multilayer composite materials, samples at different bonding temperatures (370 °C–450 °C for 30 min) were prepared respectively. The load used in this investigation was 5 MPa for all conditions. The microstructure test samples were cut from the bonded materials by wire-cutting.

Microstructures at the joint interfaces were characterized by using scanning electron microscope (SEM, S-3400N HITACHI). The main element concentration profiles and composition of the reaction layers near the interfaces were analyzed by JXA-8230 electron probe micro analyzer (EPMA) equipped with Oxford Inca X-Act energy dispersive spectrometer (EDS). The appearance and phase compositions of fracture surfaces after strength tests were measured by SEM and a Rigaku Ultima X-ray diffraction system (XRD). All tests were carried out on the polished surfaces, without etching.

3. Results and discussion

The experimental results of the joints under different bonding temperatures were presented in Table 2. Several inferences were obtained as follows: (1) when the bonding temperature was lower than 380 °C, Mg and Al cannot be jointed because of the low interdiffusion speed of the atoms at low temperature (Fig. 1(a)). (2) When the bonding temperature was higher than 440 °C, the bonding of Mg and Al failed due to the melting of Mg base caused by the eutectic reaction at high temperature (Fig. 1(c)). (3) When the bonding temperature was controlled from 380 °C to 420 °C, strongly-combined Mg/Ag/Al joints were produced without the formation of Mg-Al compounds at the interfaces (Fig. 1(b)). At 430 °C and 440 °C, Mg-Al compounds occurred at interfaces, which was harmful to the strength of the joints.

Table 2: Experimental conditions and the interfacial compound results

| No. | Bonding process parameters | Bonding condition | Compounds at the interface |
|-----|----------------------------|-------------------|---------------------------|
|     | Temperature/°C | Time/min | Pressure/MPa | Mg-Ag compounds | Mg-Al compounds |
| 1   | 370 | 30 | 5 | Failed | — |
| 2   | 380 | 30 | 5 | Completed | Yes | No |
| 3   | 390 | 30 | 5 | Completed | Yes | No |
| 4   | 400 | 30 | 5 | Completed | Yes | No |
| 5   | 410 | 30 | 5 | Completed | Yes | No |
| 6   | 420 | 30 | 5 | Completed | Yes | No |
| 7   | 430 | 30 | 5 | Completed | Yes | Yes |
| 8   | 440 | 30 | 5 | Completed | NO | Yes |
| 9   | 450 | 30 | 5 | Failed | — | — |

Figure 1. Photographs of the joints bonded at (a) 370°C, (b) 400 °C, and (c) 450 °C.
A diffusion transition zone was formed between Mg and Al base metals during diffusion bonding process. The interfacial microstructures of the Mg-Ag-Al joints bonded at 400 °C and 420 °C for 30 min were investigated by SEM, shown in Fig. 1. It can be seen that Mg and Al have been successfully jointed without porosity, crack under current technological conditions. Fig. 2(a) shows that two reaction layers different from the parent metals were formed at the joint interface, which demonstrates that the activity of Ag is high enough to form the reaction product layers. The reaction phases marked by layers A and B were identified according to the quantitative analyses of the compositions by EPMA. Mg–Ag intermetallic compounds Mg₃Ag (Mg 74.93, Al 25.07, at. %) and MgAg (Mg 50.68, Al 49.32, at. %) were the phases of layers A and B respectively. At Mg–Ag–Al interface, the interdiffusion between Mg and Ag resulted in the formation of the layered structures (layers A, B). No Mg–Al or Ag–Ag compounds were found at the interface. When the bonding temperature rose to 420 °C, the microstructure of the joint interface changed a lot compared with that at 400 °C. Fig. 2(b) shows that a three-layered structure was observed instead of two layers. The phase compositions of layers C and D marked in Fig. 1(b) were still the Mg–Ag compound: Mg₃Ag, MgAg respectively. However, the thickness of layer C increased largely with the rising temperature compared with that of layer A at 400 °C, which is the consequence of the grain growth. On the contrary, layer MgAg (D) reduced for the continuous diffusion of Mg and Ag. The phase of the newly-grown layer E close to Al base was classified as the Mg–Ag–Al ternary compound: AgMg₆Al₄ (Mg 53.59, Al 36.46, Ag 9.65, at. %). The formation of AgMg₆Al₄ means that Mg has successfully transferred into Al side overcoming the blocking effect of silver interlayer at the current process. Mg–Al compounds can be avoided when the bonding temperature increases beyond 420 °C. So the optimized bonding temperature plays an important role in controlling the interfacial phase composition of the joints.

The line and mapping distributions of the main element Mg, Ag and Al cross the interfacial zone of the joints bonded at 410 °C for 30 min were measured by EPMA, presented in Fig. 3. Fig. 3(b) and (c) show that the amount of Mg decreased from Mg base to Al base. The distribution of Ag concentrated into two parts at the interface which was in line with the two-layered structure in Fig. 3(a). The interface contained almost no Al because of the low interdiffusion of Ag and Al at 410 °C. The generation of the thin layer close Al base resulted in the diffusion of Mg, Ag and Al. According to the data of the distribution of Al, it can be observed that silver interlayers play an important role in avoiding the formation of Mg–Al intermetallic compounds with high brittleness and hardness by preventing Al atoms diffusing from Al base to Mg base.
In order to understand better the changes at the interface of the joints, the interfacial microhardness of the Mg-Ag-Al joints bonded at 410 °C for 30 min was characterized by Vickers hardness, shown in Fig. 4. The average hardness value of Mg and Al base were about 45 HV and 75 HV respectively. The hardness value in the diffusion zone increased largely with a maximum value of 240 HV due to the formation of Mg-Ag compounds, which was considered to be a disadvantage to the joints. The higher hardness means the lower ductility and ability to resist deformation of the joints. The reduction of the joint interface hardness becomes a way worthy of consideration to improve the bonding condition of the joints in subsequent studies.

Figure 4. Vickers hardness of the Mg-Ag-Al joints bonded at 410 °C for 30 min.

4. Conclusions

Mg-Ag-Al multilayer composite materials have been successfully prepared without the formation of brittle Mg-Al intermetallic compounds at the interfaces at 380 °C-420 °C for 30 min by diffusion bonding technology. Mg3Ag close to Mg base and MgAg close to Al base are the main phases at the interface of Mg-Ag-Al joints. The addition of silver interlay has blocked the interdiffusion of Mg and Al atoms resulting in the absence of the Mg-Al compounds by controlling the bonding temperature. The microhardness of the joint interfaces increased largely compared with the value of Mg and Al parent metals due to the formation of Mg-Ag intermetallic compounds.

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