Study on the interfacial bonding mechanism of Al/Mg gradient material

Luping Long, Yingbiao Peng, Bin Sun and Wensheng Liu

1 College of Metallurgical and Materials Engineering, Hunan University of Technology, Zhuzhou 412007, People’s Republic of China
2 State key laboratory of powder metallurgy, Central South University, Changsha 410083, People’s Republic of China
E-mail: pengyingbiao1987@163.com

Keywords: Impedance-graded material, magnesium (Mg), aluminum (Al), intermetallic compounds.

Abstract

Ti/Al/Mg impedance-graded material sheet is an alternative to the homogeneous single-layer sheets, which has been presented to improve shielding performance through the high efficient absorption and dissipation of fragment kinetic energy. Nevertheless, one limitation of the Ti/Al/Mg gradient material is associated with the weak Al/Mg interface. The purpose of this study is to investigate the interfacial bonding mechanism of Al/Mg gradient material interface. Microstructure characteristics at the Al/Mg interface have been investigated. It shows that an isolated island structure preferentially generates at the crystal defect as a consequence of interdiffusion, which is composed of Al3Mg2 and Al12Mg17. Then such intermetallic compounds (IMCs) grow to continuous layers with distinct convex. Finally, the interface is fully transformed into flat. A theoretical analysis and calculation have been carried out to explain the thermodynamically feasibility of IMCs. The evolution mechanism of voids at the interface is also discussed. Fractographic analysis performed on selected specimens shows typical brittle cleavage fracture characteristics, and detailed analyses reveal that the fracture failure is associated with brittle IMCs.

1. Introduction

The spacecraft can be serious damaged or even get complete wrecked by hypervelocity space debris collision. This kind of collision becomes the top threat to the safe operation of orbit spacecraft [1, 2]. A variety of high performance protective materials and structures, such as Kevlar cloth, Nextel cloth and Beta cloth [3–9], have been developed. A large number of computational and experimental works show that the Ti/Al/Mg impedance-graded material [10, 11] is a potential space debris protection material because of the higher efficient absorption and dissipation of fragment kinetic energy compared to the homogeneous single-layer sheets [12, 13].

The Ti/Al/Mg impedance-graded material with gradient density along the thickness direction can be obtained by advanced material preparation technology. The key of the preparation process is the effective metallurgical bonding of the interface. The two-step diffusion bonding process, in which metallurgical bonded Al/Mg and Ti/Al interfaces can be obtained successfully through the sufficient diffusion and reaction of the surface atoms at high temperatures, is the most commonly adopted method for preparing Ti/Al/Mg gradient material.

Compared with the Ti/Al interface, a large amount of brittle Al-Mg intermetallic compounds (IMCs) formed by the higher activity of Al and Mg atoms and subsequent diffusion reaction. Consequently, the bonding mechanism of the Al/Mg interface is more complicated. Since the growth of Al-Mg IMCs is difficult to control, the Al/Mg interface becomes a weak part of the Ti/Al/Mg gradient material. At present, the key to the development and application of Ti/Al/Mg density gradient materials lays on the quality of the Al/Mg interface.

G Mahendran [14, 15] et al have studied the influence of diffusion bonding processing parameters on the properties of the 2024Al/AZ31BMg gradient material, and also a basis for the selection of parameters was developed. M. Joseph Fernandus [16, 17] et al can effectively predict the properties of bonded Al/Mg interface by the established formula, and the surface response optimization method was used to adjust the bonding
parameters. Al/Mg gradient materials that the maximum shear strength was 18.94 MPa obtained by Liu Peng [18], used solid phase diffusion bonding technology. The diffusion behaviors and mechanical properties of the bonded Al/Mg interface were also investigated in our previous works [19, 20]. Moreover, researchers can eliminate the Al-Mg IMCs by the metal intermediate layers such as Zn and Ag [21–23], so the interface strength was increased. However, the distribution of density changed by the intermediate layer, which will directly affect the overall functional properties of the gradient material. At present, the research on diffusion bonding Al/Mg gradient materials mainly focuses on the connection process parameters, diffusion characteristics, and mechanical properties of materials. As a result, systematically studying the bonding mechanism of Al/Mg interface is the prerequisite for further improving the protective properties of gradient materials.

In this paper, the diffusion bonding technique was adopted to investigate the bonded joint of Al and Mg. The formation mechanism and microstructure characteristics of IMCs at the interface were investigated, and the element diffusion characteristics and voids evolution behaviors at the interface were also discussed.

2. Experimental procedure

Industrial aluminum and pure magnesium with dimension of 15 × 10 × 15mm was used in the study. The oxide film at the surface was removed by 2000 grit sandpaper, and then the materials were polished with Al2O3 polishing powder. Magnesium and aluminum fixed by a special fixture, which ensures its surfaces to be contacted with each other effectively. The diffusion bonding between Al and Mg was conducted in a vacuum tube furnace with TL1700 type at 1700 °C rated temperature. The effects of heat temperature at 445 °C for different holding times on Al/Mg interface were studied. The cooling process was conducted in the vacuum chamber to 100 °C.

Specimens with dimension of 10 × 10 × 10 mm, which were parallel to the load applied direction, were fabricated from the Al/Mg bonded interface by wire cut machine. The specimens were polished by 2000 grit SiC paper and Al2O3 polishing powder, and then ultrasonically cleaned in acetone. The microstructure of the Al/Mg interface was investigated by Nova Nano 230 scanning electron microscope (SEM), with secondary electron imaging, backscatter resolution 2.5 nm, resolution 1nm in high vacuum mode and resolution 1.5nm in low vacuum mode. The phase constitutions were identified using a RIGAKU RAPID II R type selected area x-ray diffractometer (XRD) with Cu-Kα target, and the minimum collimator size is 10 μm.

3. Results and discussion

3.1 Microstructure characteristics of the IMC layer

The presence of IMCs at the interface indicates that the metallurgical bonding has been obtained. The interface failure mechanisms are most attributed to the location, composition and morphology of IMCs. Therefore, the Al/Mg interfaces obtained at the bonding temperature of 445 °C for different aging-times were analyzed, as shown in figure 1. When the diffusion of components proceeds to exceed the solid solubility limit of the matrix, IMCs would preferentially nucleate at the regions with crystal defects, which further grow up as an isolated island structure appearing at the interface. As can be seen in figure 1(b), two distinct layers of island structure marked ‘Layer 1’ and ‘Layer 2’ gradually generate after diffusing at 445 °C for 10 min, indicating the formation of two different IMCs. These IMCs have been proved to be Al12Mg2 and Al12Mg17 in our previous studies [24], the IMC near the aluminum matrix is Al12Mg2, the amount of which is significantly larger than that of the IMC, Al12Mg17, near the magnesium matrix. Compared with the original interface, Al12Mg2 grows toward the aluminum matrix, while Al12Mg17 grows toward the magnesium matrix, and the interface of Al12Mg2/Al12Mg17 is located in the magnesium matrix, which also can be confirmed by the phase diagram if Al-Mg system [25]. The Gibbs energies of Al12Mg2 and Al12Mg17 phases along with temperatures between 400 °C and 500 °C are calculated at their nominal compositions based on the established thermodynamic database of the Al-Mg system by using Thermo-Calc software [26, 27]. As can be seen in figure 2, the Gibbs energy of Al12Mg17 phase shows a much larger negative value compared with that of Al12Mg2 phase, which indicates the thermodynamically feasibility of Al12Mg17 and Al12Mg2, and the Al12Mg17 has the advantages of nucleation. However, despite the
thermodynamically advantages of Al$_{12}$Mg$_{17}$ found out, the formation of IMCs is also limited by kinetic factors, such as concentration, temperature and diffusion.

Figure 1 (c) shows that IMCs have transformed from an isolated island structure into a continuous layer, and there are distinct convex at the interface extended to the substrate. Al atoms need to diffuse across the interface of Al$_3$Mg$_2$/Al toward the IMCs, while Mg atoms need to diffuse across the interface of Al$_{12}$Mg$_{17}$/Mg to the IMCs. It has been reported that Mg diffuses faster than Al$^{18}$, and therefore the concentration of Mg near the Al-matrix meets the needs for the diffusion reaction earlier. The growth rate of the IMC near Al-matrix is faster, and the proportion of Al$_3$Mg$_2$ is gradually increased, which drives the interface of Al$_3$Mg$_2$/Al to move toward the Al-matrix. Therefore, interfaces involving Al$_3$Mg$_2$ maintain a uniform straightness due to the complete growth, while the Al$_{12}$Mg$_{17}$/Mg interface exists some convex, as shown in figure 1 (d).

The convex of the Al$_{12}$Mg$_{17}$ phase penetrates into the Mg-matrix. Due to the diffusion path Al towards Mg-matrix and Mg towards Al-matrix, the concentration of Mg at the interface is higher than that inside the convex, which is opposite for Al. According to the Al-Mg phase diagram$^{25}$, new phase would nucleate and grow to aggravate the convex when the concentration of Al in front of the Al$_{12}$Mg$_{17}$/Mg is greater than its solid solubility. However, due to the relatively slow diffusion capability of Al atoms and long distance away from Al matrix, new phases do not nucleate in the front of the convex and Al dissolves in the Mg matrix. Due to the less thickness of Al$_{12}$Mg$_{17}$ at the flat area, the atomic concentration gradient is higher than that in the front of convex. Besides, the

Figure 1. SEM morphology of the Al/Mg interface at 445 °C with different holding time (a) 0 min; (b) 10 min; (c) 50 min; (d) 70 min; (e) 90 min; (f) 150 min.

Figure 2. Calculated Molar Gibbs energies of Al$_3$Mg$_2$ and Al$_{12}$Mg$_{17}$ phases based on the established thermodynamic database$^{26,27}$.
distance of diffusion is shorter at the flat area, so the growth rate of \( \text{Al}_{12}\text{Mg}_{17} \) is faster. The interface is fully transformed into flat by the sufficient heat preservation, and the thickness of reaction layer is basically identical, which is shown in figure 1(e). Figure 1(f) shows the presence of a large amount of IMCs after holding at 445 °C for 150 min Figure 3 presents the enlarged region of the rectangular area in figure 1(f). As can be seen in figure 3, some chrysanthemum eutectic structures form, and the EDS analysis indicates that the concentrations of Mg in areas 1 and 2 are 61.98 at.% and 89.69 at.%, respectively. It can be seen from the Al-Mg binary phase diagram \([25]\) that a eutectic reaction \( \text{Liquid} = \text{Al}_{12}\text{Mg}_{17} + (\text{Mg}) \) occurs at 437 °C near the Mg-matrix. After long-term heat preservation at 445 °C, the diffusion rate of Mg and Al is significantly increased as increasing amount of eutectic liquid phase forms at the interface, which results in a sharp increase of brittle IMCs and deteriorates the performance of interface. Due to the \( \text{Al}_{12}\text{Mg}_{17} \) is bcc-structure shows large lattice dislocation slip resistance and activation energy, which goes against the plastic of Al/Mg interface \([28]\).

Generally, Al and Mg atoms are interdiffused by grain boundary diffusion and bulk diffusion, and the IMCs grow up due to the diffusion reaction at the interface. The schematic diagram of structure evolution at the interface is shown in figure 4. The original interface is shown in figure 4(a), and the direction of atomic diffusion is shown as the arrows. Since the difference in the molar Gibbs energy of \( \text{Al}_3\text{Mg}_2 \) and \( \text{Al}_{12}\text{Mg}_{17} \) is small, the isolated island structure is composed of \( \text{Al}_3\text{Mg}_2 \) and \( \text{Al}_{12}\text{Mg}_{17} \) if the amount of diffused atoms and the energy that can meet the requirement, as shown in figure 4(b). Figures 4(c) and (d) show that as the increasing of holding time, the formation and disappearance of convex at the interface occur due to the unbalanced interdiffusion and the interfacial reaction.

IMCs at the interface are significantly grown after a long-term heat treatment. Due to the difference in thermal expansion coefficient between IMCs and matrixes, stress is formed at the interface. Our previous studies have shown that the maximum stress gradient is obtained at the interface of Al/\( \text{Al}_3\text{Mg}_2 \), and there are axial and radial tensile stresses inside the \( \text{Al}_3\text{Mg}_2 \) layer \([29]\). As shown in figure 4(e), because the voids are not quickly enough to grow by short-term insulation the number of voids is little even absent. Since the direction of tensile stress is consistent with the diffusion direction of Mg atoms, the diffusion of Mg atoms is accelerated by the stress gradient and tensile stress, and thus the generation of voids is exacerbated, which has also been presented in figure 4(f).

The voids at the interface are shown in figure 5. The actual contact areas of Al/Mg are decreased by the voids, and stress accumulates at the voids to make the void a source of cracks, the three-point bending strength decrease from the maximum (36.3 MPa) to 19.4 MPa that shows in our earlier research \([20]\). Moreover, the layered gradient structure is interrupted at the voids, which changes the propagation and dissipation paths of the shock wave. Therefore, protection performance of the gradient material is extremely deteriorated by the voids, and the key to suppressing voids is that the growth of IMCs has been strictly controlled.

### 3.2 Properties of the Al/Mg interface

The fracture surface is shown in figure 6, and the results show that the fracture present a distinct granular shape with typical brittle cleavage fracture characteristics. There are many pores formed in the Al-side
fracture after the extraction of the IMC, while the Mg-side fracture has apparent tears. The Al-side and Mg-side fracture components are 58.91 at% Al, 41.09 at% Mg, and 57.71 at% Al, 42.29 at% Mg respectively, which indicates that the fracture location is in the IMC layer. Stress is deflected along the interface of Al/Al3Mg2 and Mg/Al12Mg17. Due to the matrixes are strongly metallurgical bonded with IMC layers, the interface does not fail. Therefore, the stress further transmits to the IMC layers and causes the brittle cleavage fracture.

The XRD patterns of the Al/Mg fracture are shown in figure 7. As can be seen in figure 7, both sides of the fracture are mainly Al3Mg2 diffraction peaks with few matrix and Al12Mg17 diffraction peaks. It can be deduced that the fracture occurs in the Al3Mg2 phase layer near the aluminum matrix, which is consistent with the EDS results. Since the growth rate of Al3Mg2 phase in the interfacial transition zone is faster than that of Al12Mg17, the Al3Mg2 phase is easily become a crack source and the crack spreads to the aluminum matrix. The secondary void caused by the uneven
diffusion of Al and Mg atoms at the interface that reduces the toughness of IMC layer, moreover, microcracks formed by the large number of pores can be developed into a continuous crack, which makes the interface performance drops sharply. So the interface fracture failure location is inside the Al$_3$Mg$_2$ layer influenced by the growth of IMCs and atomic diffusion.

4. Conclusions

Ti/Al/Mg impedance-graded material sheet can significant improve the protective capability, which can be successfully developed by two-step vacuum diffusion bonding. Al/Mg interface as the weak part of Ti/Al/Mg impedance-graded material has been investigated. It was found that Gibbs energies of Al$_3$Mg$_2$ and Al$_{12}$Mg$_{17}$ phases along with temperatures were calculated indicates the thermodynamically feasibility, and thus new phases nucleated by the unstable Al-Mg supersaturated solid solutions, which grew up as isolated islands composed of Al$_3$Mg$_2$ and Al$_{12}$Mg$_{17}$. These isolated islands transfer to a continuous layer with distinct convex at the interface with the substrate, and the thickness of layers were basically identical with the bonding time increasing. But further bonding the large amount of eutectic liquid phase formed at the interface and thus a sharp increase of brittle IMCs. The voids in the Al$_3$Mg$_2$ mainly influenced by stress and diffusion that causes cracks, which was extremely deteriorated to the protection performance of the gradient material, and also caused fracture failure.
Acknowledgments

The research reported in this paper was financially supported by the scientific research project from Hunan province education department (No. 18C0536).

ORCID iDs

Luping Long https://orcid.org/0000-0001-6056-0999

References

[1] Gong Z Zh, Xu K B, Mu Y Q and Cao Y 2014 The space debris environment and the active removal techniques Spacecraft environment engineering. 31 129–35
[2] Zhang Y, Shortle J and Sherry L 2015 Methodology for collision risk assessment of an airspace flow corridor concept Reliab. Eng. Sys. Saf. 142 444–55
[3] Cherniav A and Telichev I 2014 Numerical simulation of impact damage induced by orbital debris on shielded wall of composite overwrapped pressure vessel Appl. Compos. Mater. 21 861–84
[4] Reinking M et al 2007 Method and shield structure against flying bodies and shock waves: EP EP1766319 A1
[5] Fahrenthold E P 2017 Computational design of metal–fabric orbital debris shielding J. Spacecraft Rockets. 54 1–8
[6] Fahrenthold E P and Park Y K 2015 Simulation of foam–impact effects on the space shuttle thermal protection system J. Spacecraft Rockets. 42 201–7
[7] Yang Y and Xu F 2015 Experimental and numerical investigation on hypervelocity impact response of 2D plain-woven C/SiC composite J. Mech. Sci. Technol. 29 11–6
[8] Ramadhan A A, Talib A R A, Rafie A S M and Zahari R 2013 High velocity impact response of Kevlar–29/epoxy and 6061-T6aluminum laminated panels Mater. Des. 43 307–21
[9] Zhang X T, Liu T, Li X G and Jia G H 2016 Hypervelocity impact performance of aluminiumegg-box panel enhanced Whipple shield Acta Astronaut. 119 48–59
[10] Zhang P L et al 2019 Study of the shielding performance of a Whipple shield enhanced by Ti-Al-nylon impedance-graded materials Int. J. of Impact Eng. 124 23–30
[11] Zhang P L et al 2019 Comparison of shielding performance of Al/Mg impedance-graded material-enhanced and aluminum Whipple shields Int. J. of Impact Eng. 126 101–8
[12] Hui D and Dutta P K 2011 A new concept of shock mitigation by impedance-graded materials Compos. Part B Eng. 42 2181–4
[13] Huang X, Liang Z, Liu Z D, Zhang H S and Dai L H 2012 Amorphous alloy reinforced whipple shield structure Int. J. Impact Eng. 41 1–10
[14] Mahendran G, Babu S and Balasubramanian V 2010 Analyzing the effect of diffusion bonding process parameters on bond characteristics of Mg/Al dissimilar joints J. Mater. Eng. Perform. 19 657–65
[15] Mahendran G, Balasubramanian V and Senthilvelan T 2009 Developing diffusion bonding windows for joining AZ31B magnesium–AA2024 aluminium alloys Mater. Design. 30 1240–4
[16] Fernandes M J, Senthilkumar T and Balasubramanian V 2011 Developing Temperature–Time and Pressure–Time diagrams for diffusion bonding AZ80 magnesium and AA6061 aluminium alloys Mater. Design. 32 1651–6
[17] Fernandes M J, Senthilkumar T and Balasubramanian V 2012 Optimising diffusion bonding parameters to maximize the strength of AA6061aluminium and AZ31B magnesium alloy joints Mater. Design. 33 31–41
[18] Liu P 2006 Microstructure and element diffusion of Mg/Al active dissimilar metal welding interface Shandong University
[19] Long L P, Sh. Liu W, Zh Y and Ma L W 2017 Microstructure and diffusion behaviors of the diffusion bonded Mg/Al joint High Temp. Mat. Pr-ir. 36 897–903
[20] Ma Y Zh, Wu L, Long L P., Sh Liu W and Ch L 2017 Microstructure and mechanic property of Mg/Al joints obtained by vacuum diffusion bonding The Chinese Journal of Nonferrous Metals. 6 1083–90
[21] Wang Y Y, Luo G Q and Li L J 2014 Formation of intermetallic compounds in Mg–Ag–Al joints during diffusion bonding J. Mater. Sci. 49 7298–308
[22] Wang Z, Qu W and Zhuang H 2016 Development of a Mg-Al filler metal for brazing magnesium alloy AZ31B Mater. Lett. 182 75–7
[23] Liu L, Zhang Z and Fei L 2013 Effect of addition of Ce in Sn–30Zn solder on the structure and properties of the Mg/Al brazed joint J. Mater. Sci. 48 2030–7
[24] Sh Liu W, Long L P, Ma Y Zh and Wu L 2015 Microstructure evolution and mechanical properties of Mg/Al diffusion bonded joints J. Alloy. Compd. 643 34–9
[25] Murray J L 1990 Binary Alloy Phase Diagrams 2nd edn., ed Thaddeus B. Massalski (Materials Park, OHL: ASM INTERNATIONAL) 1, 169–71
[26] Zhong Y, Yang M and Liu Z K 2005 Contribution of first-principles energetics to Al–Mg thermodynamic modeling Computer Coupling of Phase Diagrams & Thermochemistry. 29 303–11
[27] Liang P et al 1998 Z. Metallkd. 89 536–40
[28] Zhang D X 1990 Discuss in the brittleness of intermetallic compounds and solutions Rare Metal Mat. Eng. 6 27–31
[29] Long L P, Sh Liu W, Ma Y Zh and Wu L W 2016 Evolution of voids in Mg/Al diffusion bonding process High Temp. Mat. Pr-ir. 36 985–92