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EFFECT OF HEATING RATE ON THE PARTIAL TRANSIENT LIQUID PHASE DIFFUSION BONDED Al/Mg2Si METAL MATRIX COMPOSITE AND MAGNESIUM ALLOY (AZ91D) COUPLE

M. Mazar Atabaki*, A.H. Meisami

[1]*Institute for Materials Research, the School of Process, Environmental and Materials Engineering, Faculty of Engineering, University of Leeds, Leeds, UK*
[2]*Department of Materials Engineering, Faculty of Mechanical Engineering, Universiti Teknologi Malaysia, 81310, Malaysia*
[3]*Metallurgy & Material Dept., Golpayegan University of Technology, P.B.: 87717-65651, Golpayegan, Iran.*

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Abstract

Partial transient liquid phase diffusion bonding of aluminium metal matrix composite (Al/Mg2Si) and magnesium alloy (AZ91D) was performed using two heating rates. With an increase in heating rate from 2 to 20°C/min, Mg content in the activated bond surface decreased and the microstructure was changed. The composition and microstructure of the joined areas were examined by X-ray diffraction (XRD) and scanning electron microscope (SEM) equipped with energy dispersive X-ray spectroscopy (EDS). Kinetics of the bonding process was accelerated due to the increase of solute diffusivity through grain boundaries of the metal matrix composite. Results suggest that Mg and Si contained in the interlayer favours the partial disruption of oxide films, giving superior creep resistance at high temperature.

Keywords: Metal-matrix composite; Magnesium alloy; Transient liquid phase diffusion bonding

Introduction

In the last decade, the need for lightweight materials has led to extensive efforts in the improvement of aluminium metal matrix composites and magnesium alloys. Metal matrix composites were developed by the addition of various types of reinforcement particles to aluminium alloys with the intention of extending their applicability in electronic, aerospace, shipbuilding and automotive industries [1,2].

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* Corresponding author Mehdi Mazar Atabaki, m.mazaratabaki@gmail.com.

However, a new type of aluminium metal matrix composite (Al/Mg_2Si) showed a potential as a replacement of lightweight alloys in automobile industries [3-5]. On the other hand, the use of magnesium alloys in the automotive applications continues to grow at an unprecedented rate. Among the magnesium alloys, AZ91D is increasingly being selected to replace aluminium, zinc and plastics as manufacturers strive to reduce component weight and production costs. These materials put high demands on the joining process because of their physical properties in the commercial applications. Development of a joining process for bonding of these materials is an essential step for their widespread industrial utilization. The conventional joining techniques are limited for bonding the aluminium metal matrix composite and the magnesium alloy for many reasons. For example, the magnesium alloy shows a relatively broad melting interval and this severely increases the risk of hot-cracking during the fusion welding. Another drawback with the fusion welding of the composite and the magnesium alloy is high viscosity and poor flowability of the liquid in the welding pool. This problem leads to a great mixing of the substrates with filler metal and accelerates the formation of deleterious phases in the bond region. In the conventional fusion welding methods, rejection of the reinforcement particles in the metal matrix composite during the solidification causes microsegregation and inhomogeneous distribution of the particles in the welding pool. As a consequence, there exist many defects and porosities in the fusion zone. In addition, using solid state joining techniques like friction stir welding often ended up with segregation and fragmentation of the reinforcement particles [6-10]. Recently, partial transient liquid phase diffusion bonding technique was applied for bonding some materials in critical applications [11-14]. In this process the design of a suitable interlayer for the system is the most important parameter.

This study concentrates on an approach to join Al/Mg_2Si metal matrix composite to AZ91D by partial transient liquid phase diffusion bonding using a developed Al-base interlayer with 70µm thickness. The study coupled with creep evaluation of the bonds at high temperature.

Experimental procedure

In situ Al/Mg2Si composite was prepared using gravity casting technique by melting commercial pure Al, Mg, Si and 15wt.% Mg2Si reinforcement particles with average diameter of 20 µm. Casting was performed in an electric resistance furnace and the pouring temperature was 750°C. Degassing was conducted for 6 minutes using dry tablets containing C2Cl6. After stirring, the molten composite was poured into a cast iron mould. The solidified composite was cut into 50×50×25mm pieces by a wire cut machine. For preparing the magnesium alloy (AZ91D), commercial pure Mg, Al, Si, Zn and Mn were melted at 590°C in an electric resistance furnace and then poured into a preheated steel mould to form ingots. Subsequently, the ingots were machined into the specimens with size of 50×20×20mm. The Al-base interlayer was produced by melting the components at 710°C in alumina crucible placed in an air furnace. The molten interlayer was stirred about 10 min using a ceramic coated steel impeller. The speed of the stirrer was maintained at 400 rpm. Then the melt was poured into a cast iron mould. The Al-base interlayer was rolled to reach the thickness of 70µm. The nominal compositions of the materials are shown in Table 1.

Table 1. Chemical compositions of materials (wt.%).

Material	Element Composition (wt. %)										
	Al	Mg	Si	Fe	Cu	Zn	Mn	Cr	Ni	Ti	V
Al-Mg_2Si-Metal Matrix Composite	Bal.	9.61	5.58	0.19	0.03	0.03	0.05	0.02	0.01	0.01	---
AZ91D	9.3	Bal.	0.1	---	0.025	0.45	0.15	---	0.002	---	---
Al-base interlayer	Bal.	2.40	1.75	0.70	0.35	0.25	---	0.35	---	0.55	0.01

The composition of the interlayer was chosen to prevent the formation of complicated intermetallic compounds, provide adequate mutual solubility and minimize the thermal expansion mismatch at the bond interface. The magnesium alloy comprises α-Mg grains and Mg17Al12 at inter-dendritic boundaries while the composite contains binary eutectic α-Al /Mg2Si, primary Mg2Si particles and α-Al solid solution around the primary Mg2Si particles. Before the bonding process, the mating surface of the metal matrix composite and the magnesium alloy (10×10×5 mm) were polished using 1200 grade emery paper, and ultrasonically cleaned in acetone.

The Al-base interlayer was inserted between two substrates and the specimens were assembled in a stainless steel fixture with a pressure of 0.5MPa. The bonding was carried out in a vacuum furnace maintained at 5×10-4 Pa. The bonding temperature was 400°C. Before reaching to the bonding temperature the specimens were preheated at 300°C. The key issue in this investigation was the use of different heating rates after preheating. Two heating rates (m1=2°C/min and m2=20°C/min) were set to break down the tenacious oxide layers at the faying surface of the specimens.

After the soaking time of 18 min at the bonding temperature, the specimens were cooled to service temperature (see Fig.1). The composition and microstructure of the bond regions were examined by scanning electron microscope (SEM) equipped with energy dispersive X-ray spectroscopy (EDS). Conventional compressive creep test was accomplished for the specimens with size 4×4×6 mm3. The test was performed at 350°C under vacuum with applying a constant stress of 4 MPa.

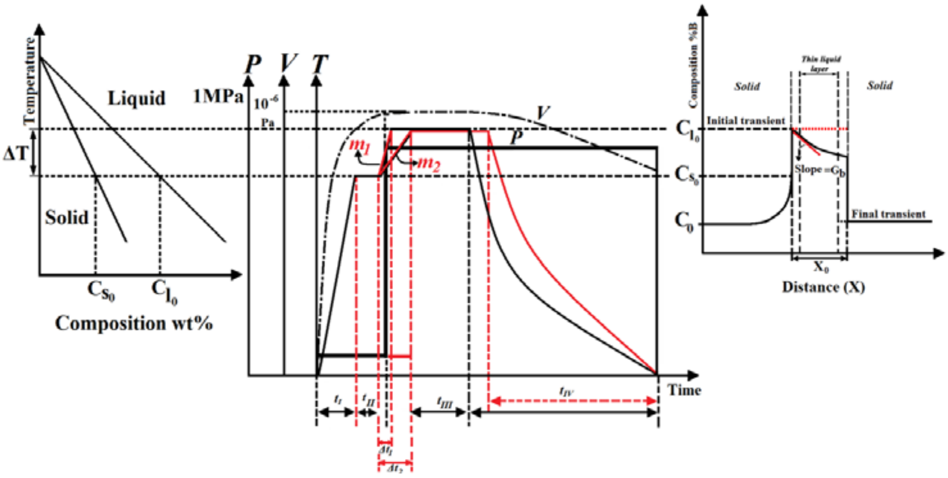
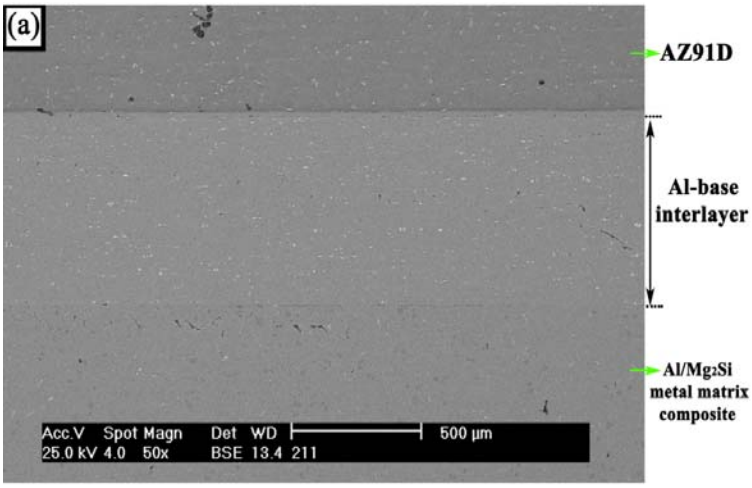


Fig.1. Cycle of partial transient liquid phase diffusion bonding of Al-Mg_2Si metal matrix composite to AZ91D with Al-base interlayer. In the right side, concentration profile at the beginning of the solidification stage shows a temperature gradient (G_b) across the interface.

Results and discussion

At the first stage of the process, the bonding temperature was selected under the solidus line of the Al-base interlayer. After this stage, the bonding temperature was increased to the liquidus temperature of the interlayer. Holding the substrates at the third stage (tIII) causes the formation of different phases due to the diffusion routes in the bond region. Fig.2 shows the microstructure of the substrates bonded at two heating rates. It can be seen that although slight changes occurred at the faying surface of the substrates, the bonding process does not significantly alters the microstructure of the parent alloys.



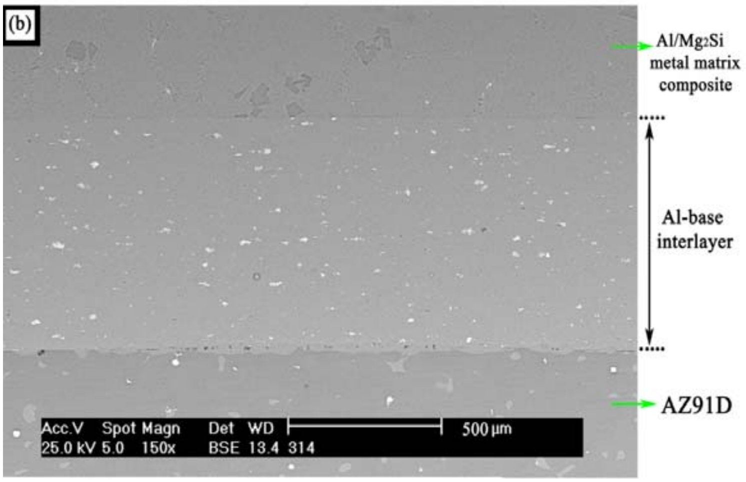


Fig.2. SEM images of joints produced at heating rates of (a) 20 and (b) 2°C/min for bonding time of 18 min. The bonding temperature was 400°C for both cases.

As it is seen, after interaction of the Al-based interlayer with the base materials, a mutual diffusion of the alloying elements occurred. This lowered the melting temperature of the contact area compared to the bulk of the interlayer. It can be observed that a small amount of dissolution has happened between the interlayer and AZ91D substrate. The interface of the composite and the Al-base interlayer was planar while a thin sinusoidal diffusion layer was observed at the faying surface of AZ91D. The joints appeared to be sound, with low porosities in both interface areas. With applying the bonding process, there was not any bulk deformation in the specimens, neither was there any aggregation of the particles at the bond interface. Some unique microstructures were formed by atomic diffusion, including partial dissolution of one of the parent alloys. The study of the microstructure shows massive precipitation of Al_4Mg inside the parent alloys. As a reason of incomplete isothermal solidification at the lower heating rate, eutectic solidification of residual liquid stimulated the formation of Al_3Mg_2, Al_2Mg_3 and Al_4Mg intermetallic compounds at the faying surface of the substrates. However, at both heating rates Al_3Mg_2 and Al_4Mg intermetallic compounds were formed. The thickness of the reaction layers was severely affected by the heating rates. In this case, considerable long-range diffusion of Al and Mg happened to nucleate equilibrium phases such as Al_3Mg_2 and Al_2Mg_3 at the interface of AZ91D and the Al-base interlayer. Fig.3 (a) and (b) illustrate the bond line at the contact surface of the base metals and interlayer. As it is seen a small fraction of the interlayer dissolves into the magnesium alloy, causing some large Al_4Mg particles trap in the liquid and enter into the bond region. Along with this result, it was observed that the reinforcement particles were not entered to the bond region giving credentials to the reliability of the process. The infiltration of the solute and its influx derived from the thin liquid layer controlled the thickness of the diffusion layer at the bonding area. The number of the reinforcement particles on the bond line was gradually reduced with decreasing the heating rate, as shown in Fig.3(c).

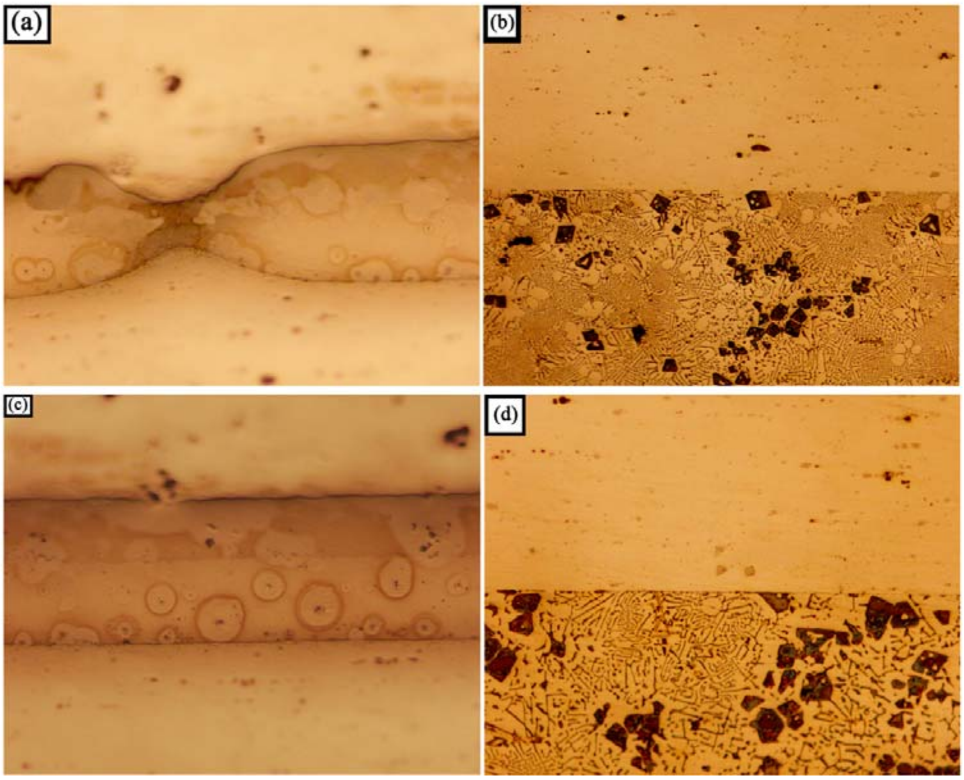
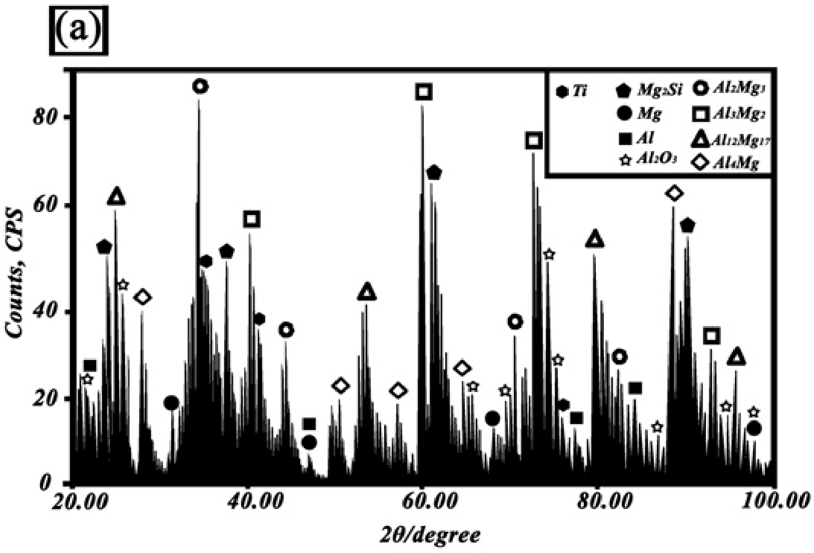


Fig.3. The microstructure near the interface of (a) AZ91D/Al-base interlayer bonded at 2°C/min; (b) Al/Mg_2Si metal matrix composite/Al-base interlayer bonded at 2°C/min; (c) AZ91D/Al-base interlayer bonded at 20°C/min and (d) Al/Mg_2Si metal matrix composite/Al-base interlayer bonded at 20°C/min.

No diffusion layer could be seen at the interface of the composite and the Al-base interlayer while at the surface of the magnesium alloy and the interlayer a diffusion barrier was formed. Mg atoms diffused more easily into the contacting area of the reinforcement particles and the interlayer matrix at the lower heating rate. The precipitation of the rich-Mg phase inside the matrix can be explained by interdiffusion of the alloying elements. In this regard, it is very likely that Mg wet the grain boundaries once the local Mg concentration exceeds the liquidus point of the Al-Mg_2Si system. Therefore, the diffusion of Mg was controlled by both solid-state and liquid-state diffusion during the bonding process. It should be noted that a melting/resolidification mechanism was the reason for the evolution of the microstructure and solute concentration. Since the diffusion barrier was formed at the interface of the magnesium alloy and the Al-base interlayer, it was partially melted during the bonding process. When partial melting occurred, Mg diffused into the diffusion barrier. Therefore, because of the low solubility and lattice diffusivity of Mg in the matrix, it is likely that the diffusion film was supersaturated with Mg during the holding time. Consequently, there are two possible mechanisms for the Al_4Mg

precipitation within the diffusion barrier and grain boundaries of the magnesium alloy. First, in the cooling stage (t_{IV}), Mg particles precipitated out of the diffusion film and formed intermetallic compounds with Al. Second, during the bonding process, Mg precipitated from the Mg supersaturated regions. It is also possible that the nucleation of the intermetallic started from the dissolution stage (t_{II}), and the growth of the Mg precipitates followed during the cooling stage (t_{IV}). The Mg concentration in the transient liquid varied with heating rate, indicating a dynamic incident. At the contact surface of the magnesium alloy and the interlayer, competitive Mg-depletion and enrichment process occurred simultaneously. On the other hand, the isothermal solidification caused the Mg enrichment of the interlayer at the contact surface. Although liquid Mg diffused to grain boundaries of the metal matrix composite, the lateral diffusion of Mg through the grain boundaries were condensed vacancies. This mechanism was responsible for the propagation and broadening of Al_4Mg in the grain boundaries of the composite. As shown in Fig.3(c) and (d), the interface concentration of Mg at the composite side was expected to be considerably small. However, it is seen that the heating rate does not promote a wavy interface at the composite side. In contrary, the heating rate promotes occlusion of Mg-rich liquid into the composite bulk, which ended up with agglomeration of Al_4Mg at the surface of grains. These results were in agreement with diffusion bonding of Al-2124 alloy reinforced with SiCp. It was shown the presence of excess matrix dislocations in the composite causes short-circuiting paths for solute diffusion and enhancing the precipitate growth rate [15,16]. The intermetallic compounds were confirmed with the result of X-ray diffraction test. The XRD analysis was carried out using CuKα target at a working voltage 40 kV and current 100mA. The XRD results showed phase constituents for the diffusion region on both sides of the bonds (see Fig.4). In the present investigation, the interlayer width was reduced to 62 μm after 18 min of holding time at the bonding temperature. This pointed out kinetics of isothermal solidification was faster than a pure monolithic system [16].



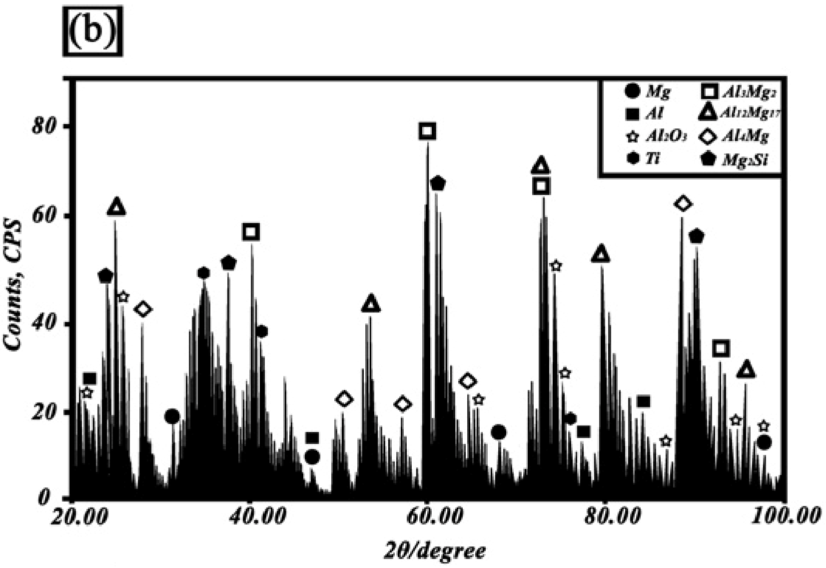


Fig.4. X-ray diffraction patterns near the interface of bonds fabricated at (a) 20°C/min and (b) 2°C/min.

Further increase in the temperature gradient leads to the breaking of the tenacious oxide layers, because increase in temperature gradient promotes mass transfer of the alloying elements across the interface, which is responsible for the increase in volume fraction of the diffusion barrier. However, plastic collapse of the asperities in the faying surface leads to an intimate contact. Higher rate of diffusion provided by the higher heating rate increases the amount of intermetallic compounds at the broken oxide layers. In Fig.5, the dark phase corresponds to Mg_2Si particles, the grey phase presented eutectic structure and the white area at the surface of the grains is Al_4Mg. A homogeneous distribution and smaller size of the primary particle was observed in the composite bonded at the lower heating rate. Irregular shape primary Mg_2Si particles with a mean size of about 30µm were detected at the lower heating rate (see Fig.6). The morphology of the eutectics in the matrix changed into the rod-shaped particles due to the slow diffusion of Si atoms in the composite [18, 19].

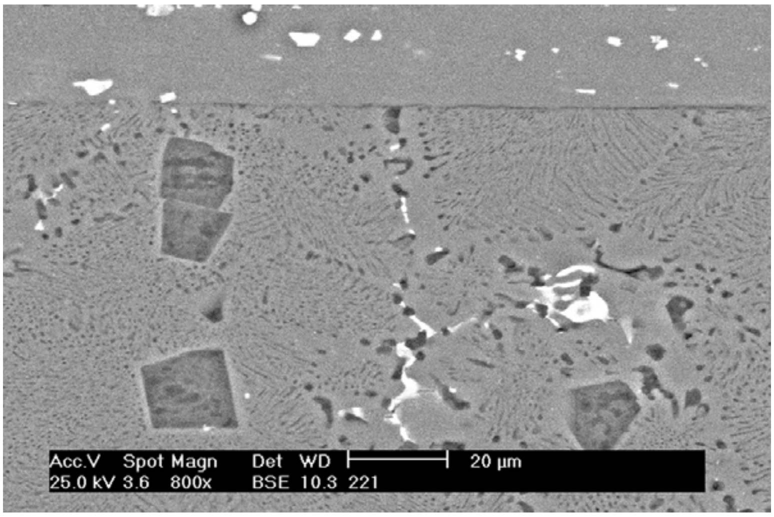


Fig.5. Microstructure of the specimens bonded at 400 °C for 18 min at the lower heating rate (see the diffusion trace of Al_4Mg from Al-base interlayer to the metal matrix composite).

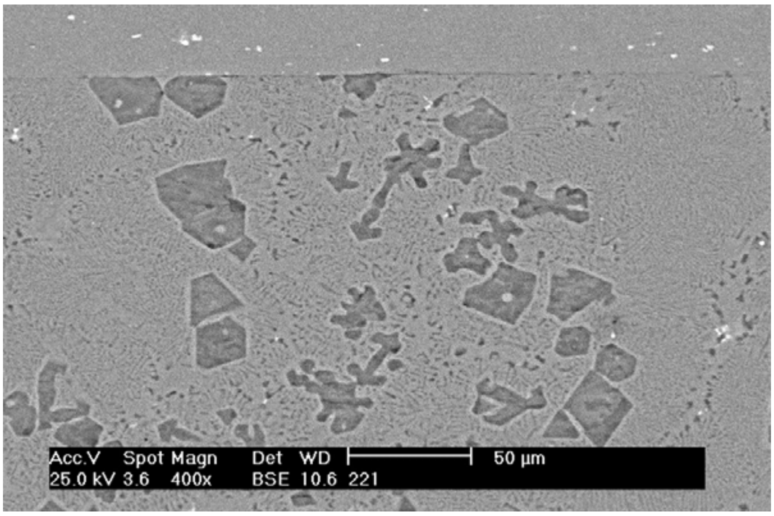


Fig.6. Irregular shaped primary Mg_2Si particles in the specimens bonded at the lower heating rate.

During the isothermal solidification, the concentration of Si atoms in the liquid layer near the surface of the solid and liquid increased with decreasing the heating rate. Thus, binary eutectic reaction was suppressed. When the heating rate increased, Al exceeded in the liquid layer. On the other side of the bonds fabricated at the lower heating rate, some petal-shape structures with networks of coarse secondary Al_4Mg particles formed a net shape fashion and spread in the AZ91D matrix (see Fig.7).

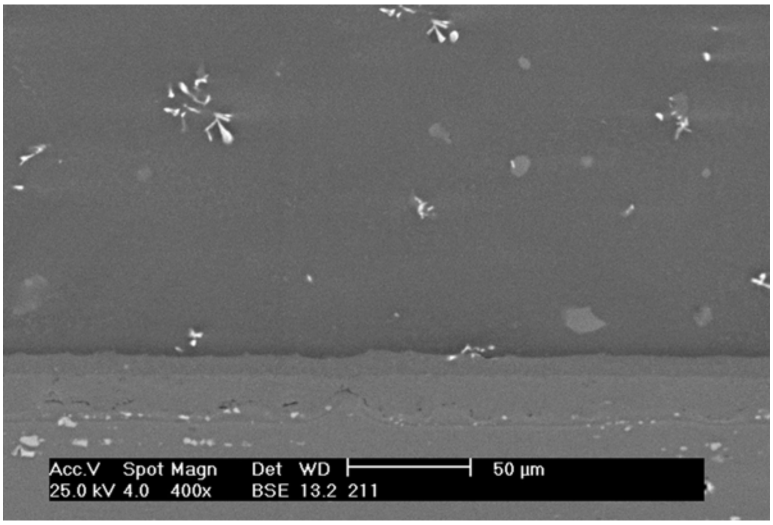


Fig.7. Microstructure of AZ91D and Al-base interlayer at the lower heating rate.

The change in composition with continuing the diffusion of Mg leads to the formation of eutectic liquid from the intermetallic phases, forming the diffusion barrier at the bond interface. From Fig.8, it can be seen that there are three diffusion layers at the surface of the magnesium alloy as marked A, B and C. The mean concentration of the alloying elements at the bond area listed in Table 2.

Table.2. Chemical composition of the bond zone in specimens joined at 400°C for 18 min with applying heating rate of 2°C/min.

Element (wt.%)	Zone		
	A	B	C
Al	Bal.	Bal.	Bal.
Mg	65	58	42
Si	1. 05	0.76	1.6
Fe	0.30	0.50	0.51
Cu	0.20	0.04	0.10
Zn	0.03	0.03	0.02
Mn	0.01	0.001	0.010
Cr	0.001	0.001	0.001
Ni	0.04	0.21	0.12
Ti	0.15	0.15	0.21
V	0.001	0.001	0.001
O	0.17	0.25	0.41

At the lower heating rate, in direct contact with the magnesium alloy, the first thin layer (less than 5 µm thick) with a single-phase appearance was formed. Based on EDS analysis (Table 2), the first layer is Al_2Mg_3 with some Si in the solid solution. The second layer (less than 10 µm thick) exhibits $Al_{12}Mg_{17}$. As shown in Table 2, probability of forming $Al_{12}Mg_{17}$ diffusion layer is confirmed. In this layer some Si is diffused in the solid solution. The third layer is thinner than the previous one (about 3 µm thick) consisting of two different phases.

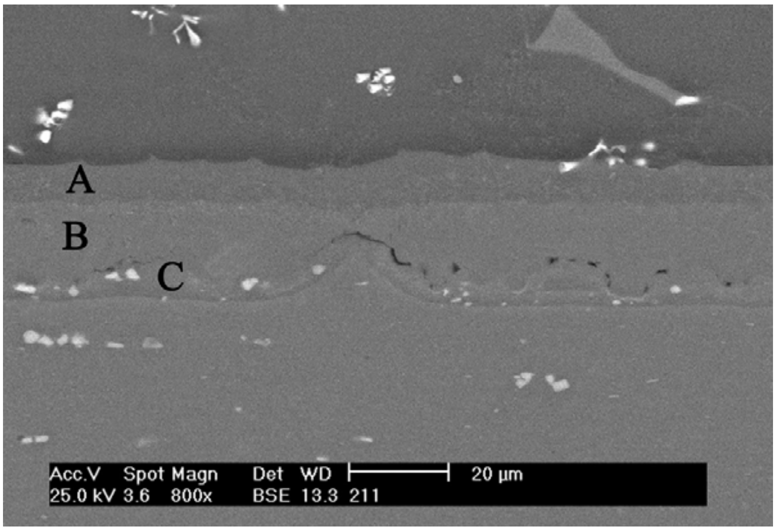


Fig.8. Microstructure across the bonds in AZ91D/Al-base interlayer side fabricated at the lower heating rate.

The continuous dark phase seems to be Al_3Mg_2 and the bright inclusions consist metallic Mg with about 24 wt.% Al and 0.9 wt.% Si. Formation of the diffusion zones can be described by dissolving the magnesium alloy in the thin liquid interlayer. When the activity of Mg in the liquid is sufficiently high near the AZ91D surface, the compound Al_4Mg nucleates on the magnesium alloy and rapidly forms a continuous reaction layer of small crystals that isolates Mg from the liquid. Other intermetallic compounds can then nucleate and grow by solid state diffusion between Mg and the first layer. It is obvious that the layers tend to grow by solid state volume diffusion. At the similar time, Mg and Si were dissolved in the thin liquid layer which was far from being saturated in Si. In such an occurrence where growth by solid state diffusion and dissolution are competing, the reaction zone rapidly attains an irregular thickness. This describes the increase of reaction zone thickness by growing solid-state diffusion. When the diffusion zone is fully saturated in Mg, dissolution of this element does not proceed and the different layers of the reaction zone can grow by solid state diffusion of Mg and Si atoms for all diffusion layers. As Al_4Mg precipitation (white phase) proceeds at a fast rate, the liquid in contact with the outer layer is depleted in Si. After that, Al_4Mg precipitates on the grain boundaries from the partial liquid. At this stage, further precipitation of Al_4Mg in the grains implies the solid state diffusion of Mg through the diffusion layer. The thickness of the reaction zone continues to increase, implying long-range crossed diffusion of Al, Mg and Si in the solid substrates. The chemical alteration of the first layer into Al_2Mg_3 at the side of AZ91D is the consequence of further solid state diffusion. Fig.9 shows the effect of heating rate on the creep strain of the bonds. The curves pointed out the strain rate decreases as a function of time. It is clear that creep resistance of the specimens is reduced with increasing the heating rate. This creep behaviour at the lower heating rate can be attributed to the type of intermetallics formed during the bonding process. The reason for the lower creep resistance is both thicker diffusion layer and formation of Al_2Mg_3 at the surface of AZ91D. This indicates that the intermetallic compounds control high temperature deformation.

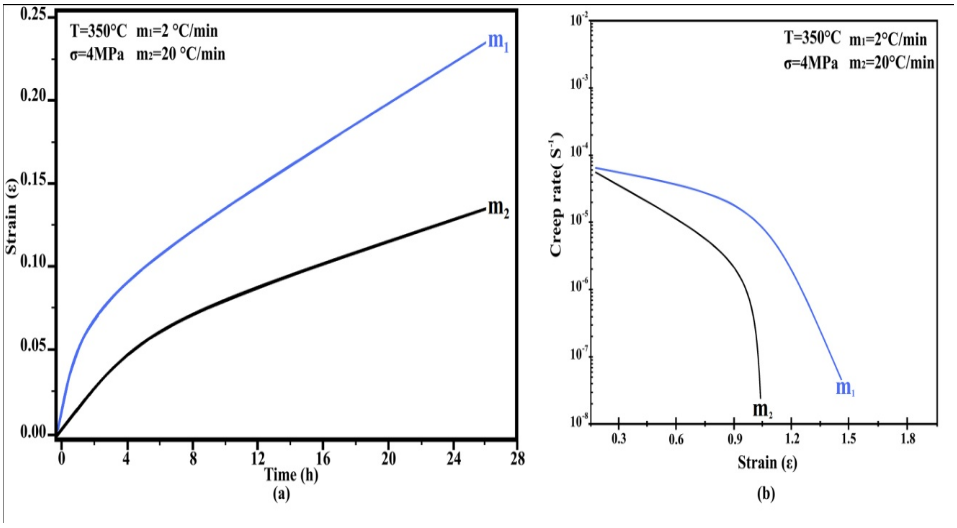


Fig.9. Response of the bonds in creep test at high temperature shows (a) ***c****reep strain as a function of time and (b) rate as a function of creep strain.*

Conclusions

Partial transient liquid phase diffusion bonding was applied to join magnesium alloy (AZ91D) and Al-Mg_2Si metal matrix composite using Al-base interlayer. The characterization of the joints reveals the following results:

1) At the lower heating rate, when AZ91D was in contact with the Al-base interlayer, three distinct diffusion layers (2-10 μm in thickness) in the sequence of $Al_2Mg_3/Al_{12}Mg_{17}/Al_3Mg_2$ were formed. The diffusion layers for the specimens bonded at the lower heating rate only consist of Al_3Mg_2 and $Al_{12}Mg_{17}$. This is explained by the fast dissolution rate of Mg in the thin liquid layer.

2) EDS results and XRD analysis show that new phases formed across the interfacial layers are $A_{l4}Mg$, $A_{l12}Mg_{17}$, $A_{l3}Mg_2$ and $A_{l2}Mg_3$. The only difference between the intermetallic compounds in the specimens bonded at two heating rates was a change in their thickness and appearance. At the lower heating rate the planar interface at the side of AZ91D altered to a non-planar interface.

3) It is found that lower heating rate provided better creep resistance at high temperature, indicating the significant effect of intermetallics on the creep properties of the bonds.

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