

MICROSTRUCTURE EVOLUTION OF LAMINATED MAGNESIUM – INTERMETALLICS COMPOSITE

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Abstract

Magnesium-intermetallics laminated composites have been fabricated through reactive bonding at 490 °C in vacuum using magnesium sheets and copper foils. Investigations were concerned with the structural transformations of a Mg-Cu couple boundary. Holding for only a few minutes resulted in the formation of thin layers at the investigated interface. Prolongation of the heating time lead to reactions in the liquid state and completely disappearing of copper layers. Therefore, the final microstructure consisted of alternating layers of unreacted magnesium and intermetallics. The microstructure was revealed in optical and scanning electron microscopy (SEM). The study exhibited the presence of different reaction products in the diffusion zone and their chemical compositions were determined by X-ray microprobe analysis. The occurrence of two different intermetallic compounds Cu_2Mg and CuMg_2 was predicted from the Cu-Mg binary phase diagram. The predominant part of the intermetallic layers was the hypoeutectic mixture of CuMg_2 and solid solution of copper in magnesium, since a liquid front of reaction was moving into the magnesium sheets. The microhardness of the reaction products and the elemental components was comparatively measured.

Keywords: Magnesium, copper, intermetallics, laminated composite, microstructure

1. INTRODUCTION

Magnesium and its alloys have attracted recently considerable attention, primarily because magnesium is the lightest structural metal. Low density magnesium alloys with excellent machinability and satisfactory recyclability are utilized in automotive, aerospace and electrical industries [1]. However, the low elastic modulus of the metal and its alloys has caused a limited use of these materials. A slightly higher modulus of elasticity can be obtained by introducing additional reinforcing phases into the magnesium matrix. The varied particulate-reinforced [2-4], fiber-reinforced [5-7] and laminated [8-10] magnesium composites have been fabricated. It is obvious that improvement of the composites properties depend strongly on the volume fraction, shape and morphology of reinforcing particles. It appears that significantly higher stiffness and generally higher mechanical properties e.g. improved fatigue behavior, fracture toughness, wear resistance, corrosion and wear capacity, can be obtained for laminated composites [11]. Recently, metal-intermetallic laminated (MIL) composites have attracted considerable interest. They have the potential to fulfil various functions, such as thermal management, heat exchange, vibration damping, ballistic protection and blast mitigation [12]. Their production method is based on synthesis of intermetallics at the interface between two metals. To receive a laminated composite sheets of one metal are put alternately with foils of another metal into a packet. Subsequently the packet must be heated to a temperature that is high enough to start the reaction between the metals and the process should be continued till the thinner metal is fully consumed. This method have been used to produce various MIL composites [8,12-17]. In the present study, the reaction synthesis was employed to fabricate laminated composites in vacuum using magnesium sheets and copper foils. The primary purpose of this study was to recognize the effect of high temperature on the structure and development of the interfacial zone between magnesium and copper. In order to do it, microstructural investigations of the reaction

zone was carefully performed. After that formed intermetallic phases were identified and the progress of synthesis process with prolonged time was investigated. As a result, the laminated magnesium-intermetallic composites with alternately located layers were produced and presented.

2. EXPERIMENTAL PROCEDURE

In the work, 1 mm thick sheets of magnesium and 0.15 mm thick foils of copper were used to produce laminated magnesium-intermetallic composites with controlled pressure, temperature and treating time. The room-temperature mechanical properties and chemical compositions of base metals are given in **Table 1**.

Table 1 Mechanical properties and chemical compositions of the base metals

Metal	Mechanical properties											
	Yield strength (MPa)			UTS (MPa)			Elongation (%)					
Magnesium	95			172			8					
Copper	64			211			42					
	Chemical elements (at%)											
	Cu	Mg	Al	Mn	Si	Fe	Ni	Pb	Sn	Zn	Na	Ca
Magnesium	0.045	99.79	0.03	0.03	0.02	0.02	0.001	0.001	0.01	0.04	0.003	0.01
Copper	99.99	-	-	-	-	0.001	0.001	0.001	0.001	0.001	-	-

Magnesium sheets and copper foils were cut into 10 mm x 10 mm square pieces. The surfaces to be joined were polished on 1200 grade abrasive SiC paper just before bonding. Any contamination on the surfaces was removed. After that sheets and foils were rinsed in water and then in ethanol. After drying rapidly, they were stacked into laminates in an alternating sequence. There were used 8 pieces of magnesium and 7 pieces of copper. A pressure of 5 MPa was employed at room temperature in a specially constructed vacuum furnace to ensure good contact between the metals. Series of attempts allowed to find that a temperature of at least 480 °C was necessary for the start and rapid development of structural processes at the interface between magnesium and copper. The temperature was increased from 20 to 420 °C at a heating rate of 0.25 °C/s. The samples were heated in vacuum of 10⁻³ Pa at 420 °C for 1 h under applied 5 MPa pressure to allow diffusion bonding of the metals. After that the packets were heated to 490 °C and held at this temperature from 5 to 60 minutes. The pressure was removed during this processing sequence with the purpose of eliminating possible expulsion of liquid phases. The temperature was then decreased slowly (cooling rate of 0.16 °C/s) to 400 °C and the pressure of 5 MPa was applied again to final consolidation of the produced laminated magnesium-intermetallic composites. After that the samples were furnace-cooled to room temperature (**Figure 1**).

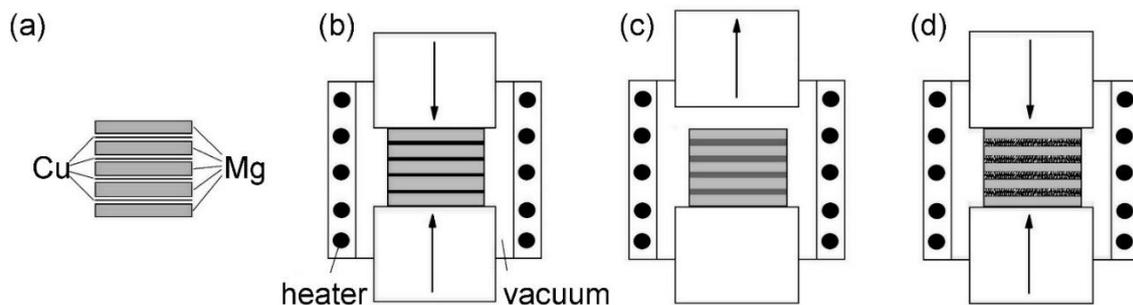


Figure 1 A schematic diagram of fabrication of magnesium-intermetallic phases composite: Mg sheets and Cu foils are stocked into a packet (a), diffusion bonding at 420 °C (b), processing with the liquid phase contribution at 490 °C (c), and the final consolidation of the laminated magnesium-intermetallic composite under pressure (d)

After fabrication, the samples were cut using diamond blade and polished applying standard metallographic techniques. Microstructural observations were performed using a JEOL JMS 5400 scanning electron microscope and a Nikon ECLIPSE MA 200 optical microscope. The chemical composition of the phases was determined by an energy dispersive spectroscopy utilizing a ISIS 300 Oxford Instruments. Before the samples were examined with the optical microscope they had been etched to reveal the structure of the intermetallic layers. Vickers (HV0.1) measurements were performed by Matsuzawa microhardness tester.

3. RESULTS AND DISCUSSION

3.1. Structural transformations at the magnesium-copper boundary during reaction

At the beginning of the structural investigations, a microstructure development due to solid state diffusion between magnesium and copper was studied intensively. The analysis of the microstructure was based on the Cu-Mg binary phase diagram (**Figure 2**) using SEM and X-ray spectroscopy.

Figure 3a shows the microstructure of the diffusion-bonded joint formed in the sample after holding for 1 h at 420 °C. **Figure 3b** shows the concentration profiles of Mg and Cu across the layers of the diffusion-bonded joint. In magnesium-copper diffusion zone four distinct layers were observed. The analysis of the microstructure based on the Cu-Mg phase diagram revealed that adjacent to copper the layer of solid solution of magnesium in copper containing 1.52 at% Mg was identified (point 1 in **Figure 3b**). This layer was followed by two single phase intermetallic layers:

Cu_2Mg (point 2 in **Figure 3b** containing 65.93 at% Cu, 34.01 at% Mg, 0.02 at% Al, 0.02 at% Mn, 0.01 at% Si and 0.01 at% Fe), and CuMg_2 (point 3 in **Figure 3b** containing 33.42 at% Cu, 66.52 at% Mg, 0.03 at% Al, 0.01 at% Mn, 0.01 at% Si and 0.01 at% Fe). Adjacent to CuTi_2 layer was the layer of solid solution of copper in magnesium (point 4 in **Figure 3b**) containing 0.03 at% Cu. The reactions at 490 °C with liquid lead to completely disappearing of copper layers. The measurements of the thickness of the reaction products showed that the reaction zone was increasing progressively with the reaction time. On the basis of measurements a relationship between the thickness (expressed in μm) of the intermetallics layer d and the holding time t (expressed in min) at the temperature of 490 °C was derived:

$$d = 0.041 t^{0.5} + 150 \quad (1)$$

The rate of synthesized layer growth with liquid phase contribution ($\text{CuMg}_2 + (\text{Mg}) \rightarrow \text{L}$) strongly exceeds the parabolic growth of intermetallic phases due to interdiffusion in the solid state.

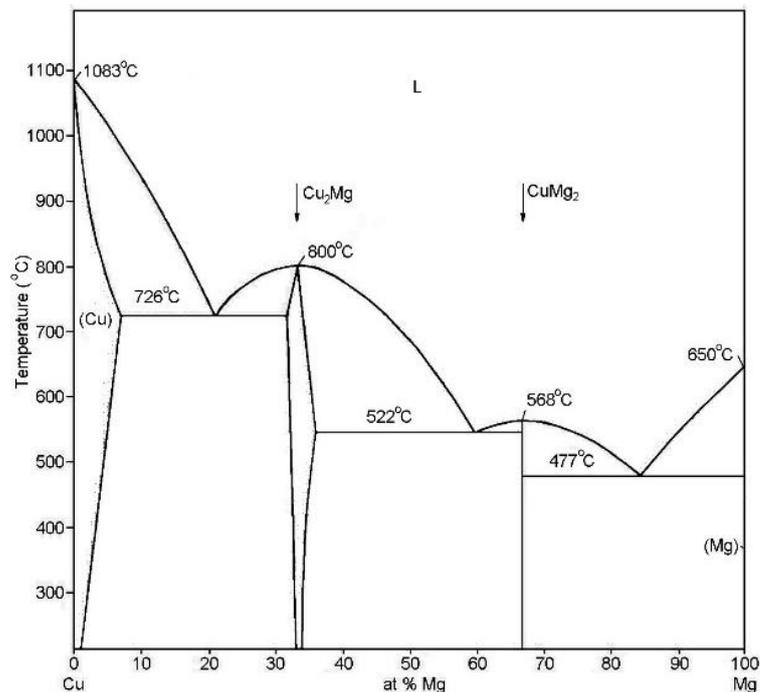


Figure 2 Cu-Mg phase diagram [18]

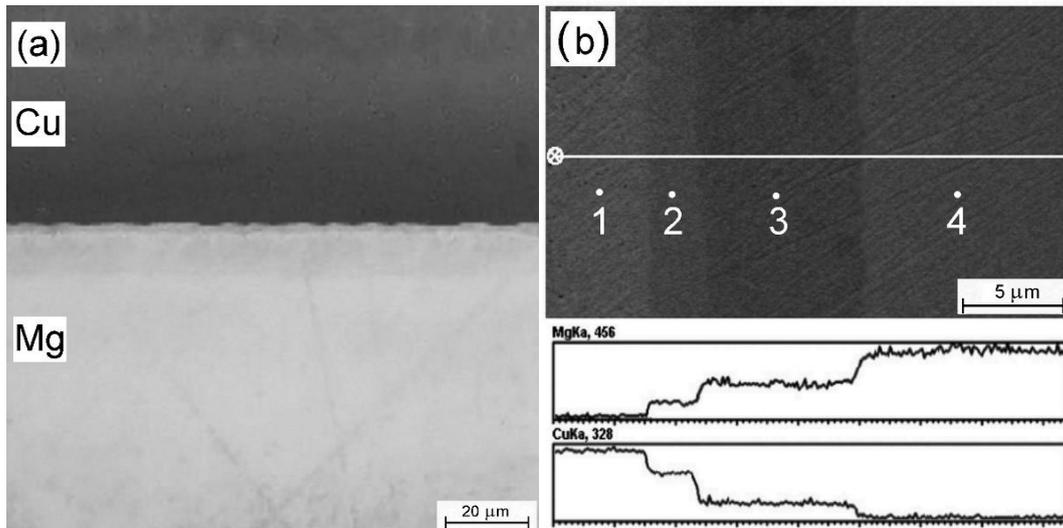


Figure 3 Optical micrograph showing the diffusion joint developed at 420 °C for 1 h (a) and SEM micrograph with the concentration profiles of Mg and Cu across the layers of the diffusion-bonded joint (b)

The final microstructure consisted of alternating layers of mixture of intermetallics + (Mg) and unreacted magnesium (**Figure 4a**). **Figure 4b** shows the microstructure of the layer formed after heat treatment at 490 °C for 1 h.

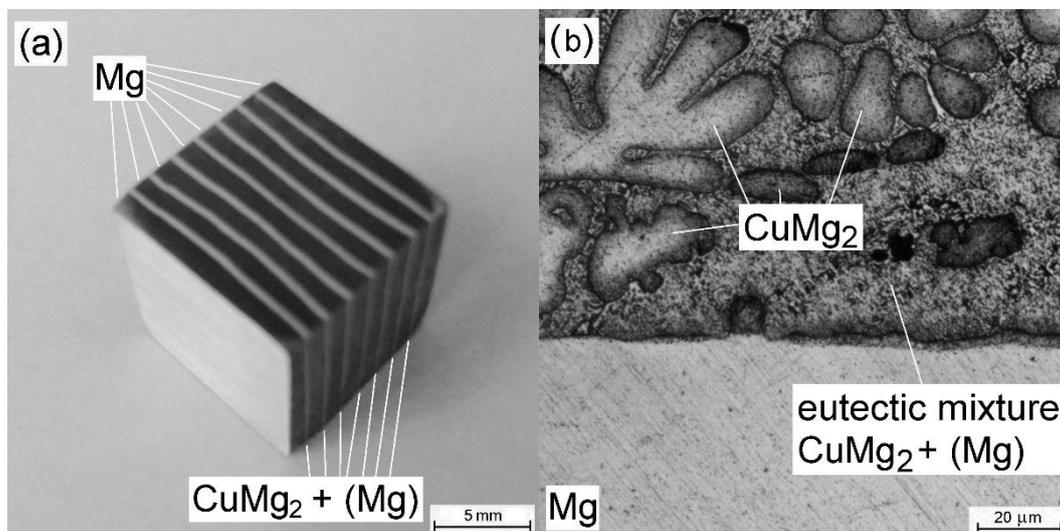


Figure 4 Micrographs showing the fabricated laminated composite (a) and a typical microstructure of the layer formed at 490 °C (b)

The X-ray analysis shown that intermetallic layers after treating for 1 h at the temperature of 490 °C were only composed of two phases: CuMg_2 (containing 33.27 at% Cu, 66.61 at% Mg, 0.04 at% Al, 0.02 at% Mn, 0.01 at% Si and 0.01 at% Fe) and solid solution of copper in magnesium. The intermetallic layers were in fact layers of hypoeutectic alloy containing crystals of CuMg_2 and eutectic mixture of CuMg_2 and (Mg) (**Figure 4b**). The results clearly show that the structure resulting from the solidification of a liquid layer contains phases that are all enriched in magnesium. It is evident that the front of reaction zone at Cu-Mg couple migrated into the magnesium sheet, as it shown in **Figure 3**. In spite of complete copper dissolution due to reaction with the liquid phase contribution, more volume of magnesium than copper were consumed. Therefore, the predominant part of the intermetallic layers was CuMg_2 phase, since a liquid front of reaction was moving into

magnesium. This is also the reason why there is no Cu_2Mg phase in the formed composite. It reacted with magnesium and formed a CuMg_2 phase according to the formula: $\text{Cu}_2\text{Mg} + \text{Mg} \rightarrow \text{CuMg}_2$. The process of producing laminated composites with the use of other base metals was similar [8-17], and only other types of intermetallic phases were obtained.

3.2. Microhardness measurements

Microhardness measurements were performed for basic metals as well as for formed intermetallics and eutectic mixture. Results of microhardness measurements are given in **Table 2**.

Table 2 Results of hardness measurements

Magnesium	Copper	CuMg_2	Eutectic mixture $\text{CuMg}_2 + (\text{Mg})$
55-62 HV0.1	95-105 HV0.1	320-353 HV0.1	235-286 HV0.1

The maximum hardness values in the range of 320 to 353 HV were achieved for relatively large CuMg_2 intermetallic phase crystals that were present in hypereutectic alloy. In the formed layers, comprising the eutectic mixture of CuMg_2 and (Mg), hardness was lesser and contained in the range of 235 to 286 HV. In comparison, for metals used to produce the laminated composites: magnesium and copper the values of hardness were from three to five times lesser. Similar results were obtained by Dziadoń et al. [8] for layered Mg-eutectic composites synthesized using Mg and Al foils.

4. CONCLUSION

In a consequence of reaction occurring between magnesium sheets and copper foils in vacuum at 420 °C the diffusion-bonded joint is formed containing four distinct layers: solid solution of magnesium in copper, Cu_2Mg , CuMg_2 , and solid solution of copper in magnesium. When temperature is raised to 490 °C, a laminated magnesium-intermetallic composite can be formed. It is evident from metallographic examinations that the predominant part of intermetallics is synthesized in the region passing from a liquid state to a solid state. The reaction zone contains intermetallic compound CuMg_2 , but the predominant part of the formed layers is the eutectic mixture of CuMg_2 and solid solution of copper in magnesium, since a liquid front of reaction is moving into the magnesium sheets. The formed layers are approximately from 5 to 6 times harder than layers of magnesium, and the maximum hardness values in the range of 320 to 353 HV can be achieved for the CuMg_2 intermetallic phase.

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