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Effect of Annealing Treatment on Microstructure of Mg/Cu Diffusion Layer

Yong-qin ZHANG [1,a*],Shuang-ming DU [1,b] and Jie HU [1,c]
[1]College of Materials Science and Engineering, Xi'an University of Science and Technology, Xi'an 710054, China
[a*]18710980374@163.com[1],[b]shuangmingdu@163.com,[c]clhujie2009@163.com

Abstract. The experimental study of the "mosaic type" Mg/Cu diffusion couple in the argon protection environment was carried out. The microstructure and micro-hardness of the Mg/Cu diffusion layer were analyzed by means of SEM, EDS and micro-hardness tester. The results show that the diffusion behavior of Mg/Cu diffusion couple is influenced by annealing temperature. When the temperature located at 430~450℃, the behavior of element diffusion happened in solid phase, the thickness of diffusion layer increased slowly which was about 5~25um, and the microstructure of diffusion layer from Mg to Copper includes magnesium based solid solution /Mg_2Cu/Cu_2Mg/ copper based solid solution, the micro-hardness of the diffusion layer was significantly higher than that of the two sides, when bonded at 430℃ for 4h, the micro-hardness of the diffusion layer was highest that up to 194HV. When the temperature reached to 485℃, the thickness of diffusion layer increased significantly which was about 5.3mm because of the formation of eutectic liquid phase reaction zone, and the microstructure of the diffusion layer were magnesium based solid solution /Mg-Cu eutectic/ Cu based solid solution.

1. Introduction

The research and application of Mg/Cu binary material has already extended from sailing to the military field, such as automobile, computer, communications equipment, and other civilian products with high added value, in order to reach the goal of weight loss and product multiplicity, it is necessary to weld the two metal materials [1-3]. Mg/Cu intermetallic compound can be easily formed at the interface of the joint after Mg/Cu diffusion bonding, which seriously affect the joint mechanical properties. However, the type and thickness of these intermetallic compounds are closely related to the Mg, Cu atom diffusion behavior[4-5]. Therefore, to discuss the influence of Mg/Cu atom diffusion behavior on interface diffusion layer growth mechanism and hence infer the generation and evolution law of the intermetallic compounds is of great importance to improve the quality of joint and understand the bonding process behavior. In this paper, the experimental study of the "mosaic type" Mg/Cu diffusion couple in the argon protection environment was carried out. The microstructure and micro-hardness of the Mg/Cu diffusion layer were analyzed by means of SEM, EDS and micro-hardness tester.

* Corresponding author: 18710980374@163.com

2. Experimental Materials and Methods

The experiment using Mg wire whose thermal expansion coefficient is bigger than matrix as the "core", and copper bar whose thermal expansion coefficient is smaller than matrix as "matrix". The diameter of Mg wire is 10 mm and purity ≥ 99.99%, the diameter of copper bar is 18 mm and purity ≥ 99.99%, mosaicing10 mm diameter Mg wire in the 9.8 mm diameter hole that was drilled in the center of the copper bar, which will realize the interference fit of both and make a Mg-Cu binary diffusion couple that the interface is hook face [6-8].

The heat treatment temperature is selected according to the Mg-Cu phase diagram [9], putting prepared diffusion couple in tube furnace with argon protection and annealing heat treatment. Smoothing and polishing the sample that has been happened diffusion along the direction perpendicular to the interface, using SEM and EDS analysis organization forms and element distribution of Mg/Cu diffusion couple layer, using micro-hardness tester text the micro-hardness of diffusion layer, and revealing the influence law of diffusion temperature and time on the microstructure and mechanical properties of Mg/Cu diffusion layer, preliminarily interpreting Mg/Cu interface layer formation mechanism.

3. Experimental Result and Analysis

3.1The Micro-structure of Mg/Cu Diffusion Couple Interface Reaction Layer

Fig.1 shows the result of diffusion dissolution layer backscattered electron image and spectrum scan of Mg/Cu diffusion couple under the temperature of 430℃ for 4h. It can be seen from fig. (a) that the diffusion dissolution layer includes dark grey diffusion zone of magnesium side and light gray transition layer of copper side, the average width of diffusion dissolution layer is about 5um, the interface between diffusion zone and Mg, Cu matrix shows clear, diffusion zone structure is compact and no metallurgical defects such as crack and incomplete penetration, the formation of diffusion zone is the result of mutual diffusion between Cu and Mg. From the image of Line scan, it can be seen that Mg element concentration field rises and falls evidently near side of Mg matrix, and Cu element in the whole diffusion process is mild, has no big volatility, and shows a decreased trend from Cu side to diffusion zone. Due to the melting point of Mg is lower than that of Cu, under the same temperature, the diffusion coefficient of Cu is less than Mg, namely Cu atom diffusion rate is less than Mg atom. When a large amount of magnesium atom diffuse to the Cu substrate in the process, a small amount of copper atom diffuse to Mg, Mg-Cu solid solution or intermetallic compound can form near the diffusion interface layer. Because each phase layer corresponds to a phase, so each layer composition is homogeneous and can randomly test a point on it, according to the atomic percentage can roughly infer the corresponding compounds of the layer. Combined with the energy spectrum spot scan result of 001 and 002 two points, it can be inferred that the dark grey diffusion zone is Mg-Cu binary compound, light gray transition layer is copper based solid solution.

TABLE 1. THE ELEMENTAL COMPOSITION DETERMINATION RESULTS OF DISSOLVED DIFFUSION LAYER IN -DIFFERENT AREAS (ATOM PERCENTAGE)

elements	test position	
	1(At%)	2(At%)
Mg	10.63	77.90
Cu	89.37	22.1

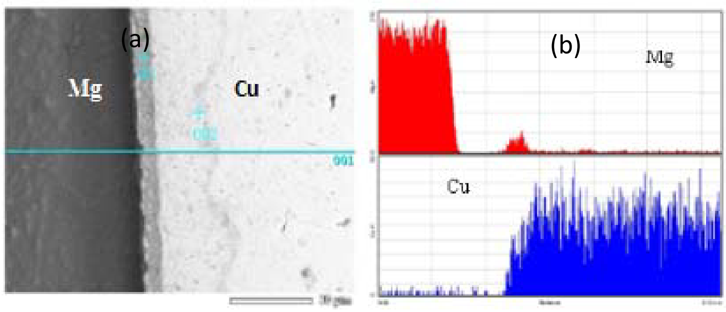

Fig.1.The backscattered electron imaging and spectroscopy scanning curve of diffusion layer of Mg/Cu diffusion couple at 430 ℃ for 4h:

(a) the back scattered electron image of diffusion layer, (b) energy spectrum scanning curve

3.2The Microstructure of Mg/Cu Diffusion Couple Interface Reaction Layer

Figure.2 is diffusion layer scanning electron microscope of Mg/Cu diffusion couple with different heating temperature and holding time. It can be seen that a diffusion layer formed after the diffusion couple was heated at 430℃ for 8h, and the average width of diffusion layers is about 10um, which is related to the mutual diffusion coefficient of Mg and Cu. The interface between diffusion zone and the matrix of magnesium and copper is still clear. When the temperature reached to 450℃, the average width of the diffusion region increased to 20um, and the center position of the diffusion region is almost continuous crack, which is due to the brittle hard compound in this zone suffered the tensile stress of the adjacent layer in the cooling process. When the temperature continued to rise to 485℃, the width of the diffusion zone sharply increased to 5300um, and it can be seen that the interface between magnesium matrix and diffusion layer was clear. Figure.2(e) is the amplification of the A part of the figure.2(d), the organization of the diffusion region was a clear lamellar structure, combined with the Mg-Cu binary phase, it can be considered as the regional organization is Mg-Cu eutectic. Figure.2(c) shows the SEM of the diffusion zone bonded at 430℃ for 12h, it can be seen that the width of the diffusion region increased, with an average width of 25um, when compared figure.2(c) with figures: 1 and 2(a), however, a continuous crack emerged in the diffusion region. This is because the thickness of the brittle hard phase formed during the diffusion process is relatively large, resulting in the increased brittleness. While the stress effect emerged in the later cooling process would lead to micro-crack and gradually transformed into a continuous crack.

In summary, the effect of temperature on the diffusion process not only increases the thickness of the diffusion layer, but also changes the diffusion form of Mg and Cu atom, namely, from the solid diffusion to dissolved diffusion that Mg, Cu solid phase matrix atoms dissolved in the eutectic liquid phase and the diffusion in the liquid phase, the diffusion ability of the atom, and it is also beneficial to the homogenization of the composition of liquid phase. The higher the temperature, the greater the diffusion flux, diffusion reaction more intense, the larger the diffusion layer thickness. The effect of holding time on the diffusion intuitively presents the increased width of diffusion zone. With the extension of holding time, the solid phase diffusion of Mg and Cu atom is more fully, and the diffusion migration degree increases gradually, so that the thickness of the diffusion dissolution layer increases.

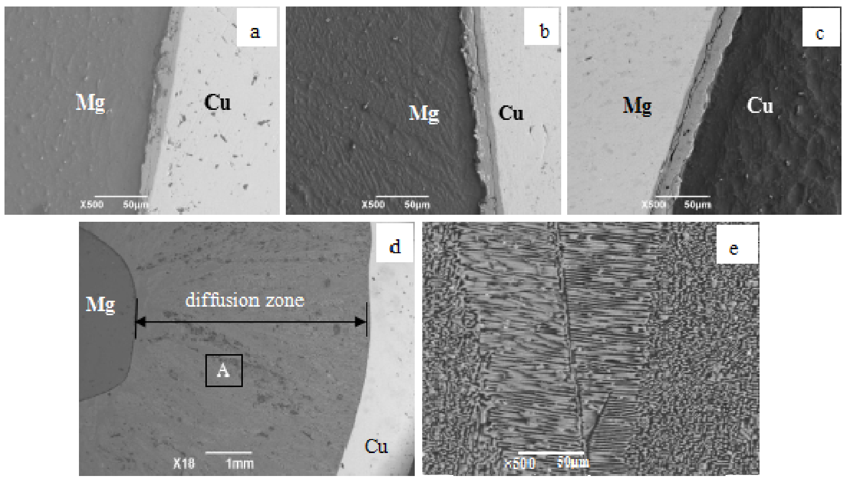

Fig.2 The diffusion layer scanning electron micrographs of Mg/Cu diffusion couple at different heating temperature and holding time:

(a) 430℃, 8h; (b) 450℃, 8h; (c) 430℃, 12h; (d) 485℃, 8h; (e) the amplification of the A part of the figure (d)

The formation of phase interfacial diffusion dissolution layer in the solid state of binary metal system is the result of the diffusion dissolution and second crystallization. Because of the difference of chemical potential, binary metals will be in a non-equilibrium state of thermodynamics when they contact, and the atom will diffuse mutually when heated under the melting point. The process that one kind metal atom diffuses into another metal or both diffuse mutually is actually the process of solid dissolution, thereby forms a solid solution. When the solid solution is over saturated, the structure is unstable, nucleation emerges and crystal grains grow into a new phase, the metal compound layer in the diffusion layer is formed by the formation of over saturated solid solution in the two groups.

The self-diffusion activation energy of most metals generally has the following relations:$Q \approx 0.144T_m$, the melting point of copper is much higher than Mg, so the self-diffusion activation energy of Cu is much higher than magnesium, under the condition of heat activating the solid phase with same temperature, the Magnesium crystals in Mg wire are more prone to collapse, but the Cu atom almost not diffuses, so the edge of diffusion is always at the end of the Cu. According to Mg-Cu binary phase, when the temperature is below 500℃, the Mg has a solubility of 5% in Cu, while Cu doesn't dissolve in Mg, Mg atom migrates to Cu block and enters into Cu matrix, and thus forms a solid solution layer with Cu as the solvent. Because of the solubility of Mg in Cu is very low, prolongs the holding time, when Mg atom concentration is low, the solid solution formed at the Mg/Cu interface is saturated. Then, in the defect region, Mg-Cu intermetallic compound crystal nucleus whose Mg atom fraction is relatively lower firstly formed, the crystal nucleus grows along the Mg/Cu, and gradually forms instable CuMgx intermetallic phase, when further extends the holding time, the Cu atom in parent material crosses the unstable reaction layer, even diffuses into the Mg matrix and generates reaction generates a CuxMg intermetallic compound, when holding time reaches to a critical value, the interface reaction layer is mainly composed of Mg_2Cu and Cu_2Mg.

3.3Micro-hardness of Mg/Cu Diffusion Couple Interface Reaction Zone

When bonded at 430℃ for 4h, 8h and 12h, the micro-hardness test of Mg/Cu diffusion couple sample was carried out by using micro-hardness tester. Taking 7 points along Cu side to Mg, and the micro-hardness values are shown in Fig.3.

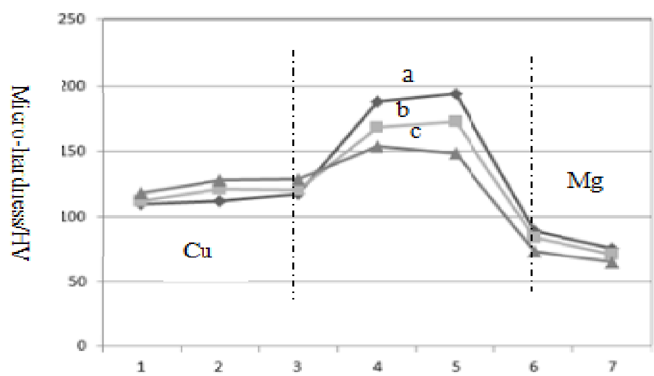

Fig.3. The microhardness of interface laye under different holding time at 430 ℃: (a) 4h; (b) 8h; (c) 12h.

It can be seen from Fig.3 that the micro-hardness of Mg and Cu matrix side are uniform, the micro-hardness value of Cu matrix side is slightly higher than Mg matrix, and joint diffusion zone hardness value is significantly higher than Mg, Cu matrix micro-hardness value. This is due to Mg atom and Cu atom formed a large number of solid solution in the diffusion region, which strengthened the organization structure of the diffusion zone and improved the micro-hardness of the microstructure. As the holding time increased, the solid solution gradually converted to Mg-Cu intermetallic compound and enriched in the Mg matrix, which is why the maximum micro-hardness close to the Mg matrix. Comparing the micro-hardness of diffusion zone with different holding time, it can be found that the shorter the holding time, the higher the micro-hardness of the diffusion region. This is because as the holding time prolonged, Mg and Cu atom diffused thoroughly, the junction structure tended to be more uniform, the organization structure was more stable, the micro-hardness of joint relative to diffusion zone whose microstructure is more complex was slightly smaller.

Conclusions

(1) Under the same holding time, heating temperature had a significant effect on the diffusion behavior of Mg/Cu diffusion couple interface element. At 430 ~450℃, Mg, Cu atom diffused in the solid phase, and the diffusion layer thickness was about 5~25um, the

thickness of diffusion layer increased slowly as the temperature increased; At 485℃, the thickness of the interface reaction layer increased sharply, and the 5.3mm eutectic liquid phase reaction zone formed in Mg/Cu diffusion region.

(2) At the same temperature, as the holding time extended, the thickness of Mg/Cu diffusion couple reaction layer was greater, and the structure of diffusion region from Mg to Cu included Magnesium based solid solution/Mg_2Cu/Cu_2Mg/copper based solid solution. The micro-hardness of interface reaction zone was significantly higher than that of the two sides, the highest micro-hardness value that up to 194HV was obtained at 430℃ for 4h, the micro-hardness value with holding time of 8h was lower, and 12h was lowest.

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