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Formation of magnesium-eutectic mixture layered composite

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Abstract

The work was started from study of the structural transformations, that in elevated temperatures, take place at the magnesium-aluminium interface. It was found experimentally that at temperature of 445°C solid state diffusion is followed by liquid phase appearance at the Mg-Al interface. For process passing with the liquid phase contribution thickness **d** of the reactions products against the time **t** can be expressed by the equation $\mathbf{d=At^n}$ with the time exponent value $\mathbf{n}>1$. This process is very fast compared to solid state polyphase diffusion and can be controlled, therefore may be used for fabrication of layered composite. To obtain a composite, magnesium and aluminium sheets are formed alternately into a pocket and then heated at temperature of 445°C. Heating is prolonged for passing all the aluminium with part of the magnesium sheets into a liquid phase. During solidification liquid phase is transformed into eutectic mixture. So, composite that was formed consists of alternating magnesium and eutectic mixture layers. Using X-ray microprobe analysis and on the basis of Mg-Al binary phase diagram we were found that the eutectic mixture contains two phases: $Mg_{17}Al_{12}$ intermetallic compound and solid solution of aluminium in magnesium.

Keywords: Composites, Intermetallic phases, Magnesium

1. Introduction

It is commonly known that intermetallic phases have greater strength properties than the elemental metals from which they are formed. From the other side intermetallic phases are brittle compared to metals. Metal-intermetallics composites offer an attractive combination of properties from the both components. This is why interest in intermetallic composites has grown, especially over the past two decades.

Composites have been produced using a wide range of different methods [1]. Fabrication methods can be roughly divided into two techniques: solid state processing and liquid phase processing. Formation of intermetallic phases between two dissimilar metals at high temperature allows develop a new method of producing a layered metal-intermetallics composite. Foils of two different metals are formed alternately into a packet and heated to form intermetallic layers. The process is continued till one of the metals is fully consumed in the course of reaction. Using this method Ni-intermetallics [2-4], Fe-intermetallics [2-4], Ti-intermetallics [2-7], Ti3Al2.5V-intermetallics [8] and Cu-intermetallics [9] layered composites have been processed.

We were adopted this method for formation of magnesium layered composite. In the last years magnesium and its alloys have been a subject of great interest. This interest has been due largely to its low specific density and especially attractive specific strength of the magnesium alloys. The main disadvantage of these materials is the low elastic modulus. It appears that composite consisting of layers of magnesium (or magnesium alloy) and layers containing intermetallic phases could possess substantially higher strength and stiffness. So, we intended to obtain composite with layers of magnesium partitioned by hard layers containing intermetallic phases. Aluminium was chosen for synthesis of intermetallic phases because of low density. According to Al-Mg binary phase diagram [10] in the Al-Mg system the $Mg_{17}Al_{12}$ and Al_3Mg_2 intermetallic compounds should be expected. It was also taken into account, that aluminium is major alloying element or commonly used alloying element of the magnesium alloys.

2. Experimental procedure

The work was started from analysis of structural transformations at the Mg-Al interface. Mg-Al couples were held at a temperature of 430 ^{0}C or/and at temperature of 445 ^{0}C for different period of time. Selection of the temperatures was based on the Al-Mg binary diagram presented in Figure 1. The 430 ^{0}C is just below and 445 ^{0}C just above the eutectic temperature of 437 ^{0}C.

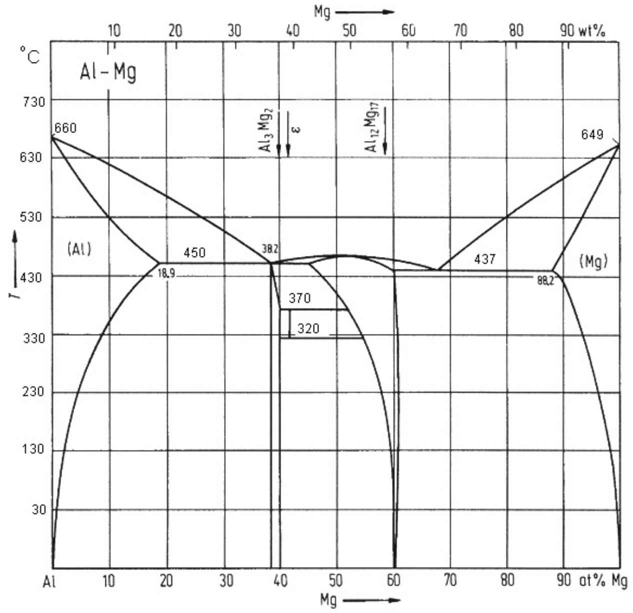

Fig. 1. Al-Mg phase diagram [10]

Due to solid state diffusion at the Mg-Al interface Mg and Al specimens were joined into a Mg-Al couple. The Mg-Al couples were made from 30 x 15 mm specimens that were cut from 3.0 mm thick magnesium and 0.8 mm thick aluminium sheets. The metals were of 99.9 purity. The joining surfaces were mechanically polished and then Mg-Al specimens were held at a temperature of 430 ^{0}C for 20 minutes in vacuum furnace under pressure of 5 MPa.

To investigate the structure development and the rate of growth of layer that was formed at 430 ^{0}C and at 445 ^{0}C between the metals, the Mg-Al couples were kept at these temperatures for varying periods of time and then surface cooled. After cooling, structural examinations of the samples were performed using optical microscope (Neophot 2) and electron scanning JMS 5400 microscope. Optical microscope was also used for determination of the layer thickness that with progress of the time was increased at the Mg-Al interface. The chemical composition of the phases was determined by the electron microprobe analysis using ISIS300 Oxford Instruments.

On the basis of above investigations a procedure of composite fabrication was developed. Magnesium and aluminium sheets were formed alternatively into a packet. For diffusion bonding of the Mg and Al sheets the packet was held at 430 ^{0}C in the vacuum furnace under the pressure of the 5 MPa. Then load was removed and packet was heated to 445 ^{0}C and held at this temperature for 60 minutes. Finally, at 400 ^{0}C pressure of 5 MPa was again used for consolidation of the composite. By choosing of the thickness of starting Mg and Al sheets, composites with different thickness ratio of the magnesium layers to the layers synthesised from Al and Mg metals was produced.

3. Results

Solid state diffusion produces two separate layers between Mg and Al metals. Figure 2 shows the microstructure developed after keeping the Mg-Al couple for 20 minutes at temperature of 430 ^{0}C. The chemical composition of the narrow layer (marked I): 58.9 % at. Mg and 41.1 % at. Al indicates according to Mg-Al diagram on $Mg_{17}Al_{12}$ intermetallic compound. X-ray microprobe analysis of the wider (marked II) layer: 41.0 % at. Mg and 59.0 % at. Al suggests Al_3Mg_2 phase.

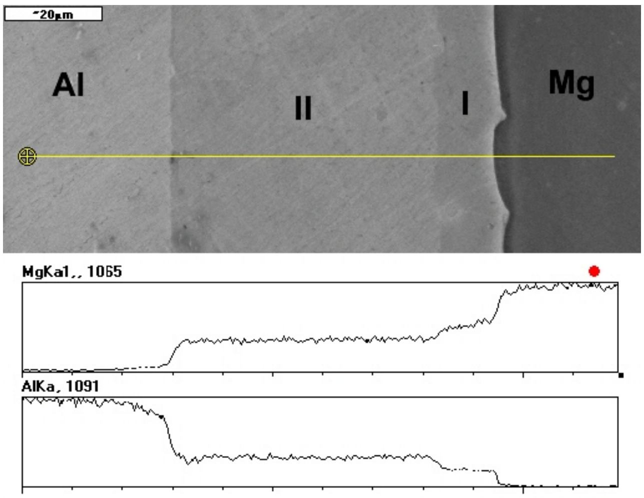

Fig. 2. Microstructure and concentration of Mg and Al profiles across the Al-Mg interface. The specimen was annealed at the temperature of 430^{o}C for 20 minutes

Measurements allow to express the thickness-time dependence for two-layered zone as $\mathbf{d = 0.019\ t^{0.36}}$. So, growth kinetics at the Mg-Al interface at the 430 ^{0}C is typical for polyphase diffusion that is commonly described using parabolic equation [11-14] $\mathbf{d = A\ t^n}$ (**d** - layer thickness, **A** - constant, **t** - time and **n** - power low exponent).

If Mg-Al couples were holding at 445 ^{0}C (just over eutectic temperature of 437 ^{0}C) liquid phase appears at the Mg-Al interface. A series of micrographs (Figure 3a through 3c) illustrates systematic increase with time of the layer formed between Mg and Al metals. Thickness of the layer can be expressed on basis of measurements by relation $\mathbf{d = 0.0093\ t^{1.34}}$ (**d** - thickness in mm, **t** - time in min.). Because the value of the time exponent is very high (**n**>1), rate of layer growth substantially exceeds a parabolic growth observed for solid state diffusion.

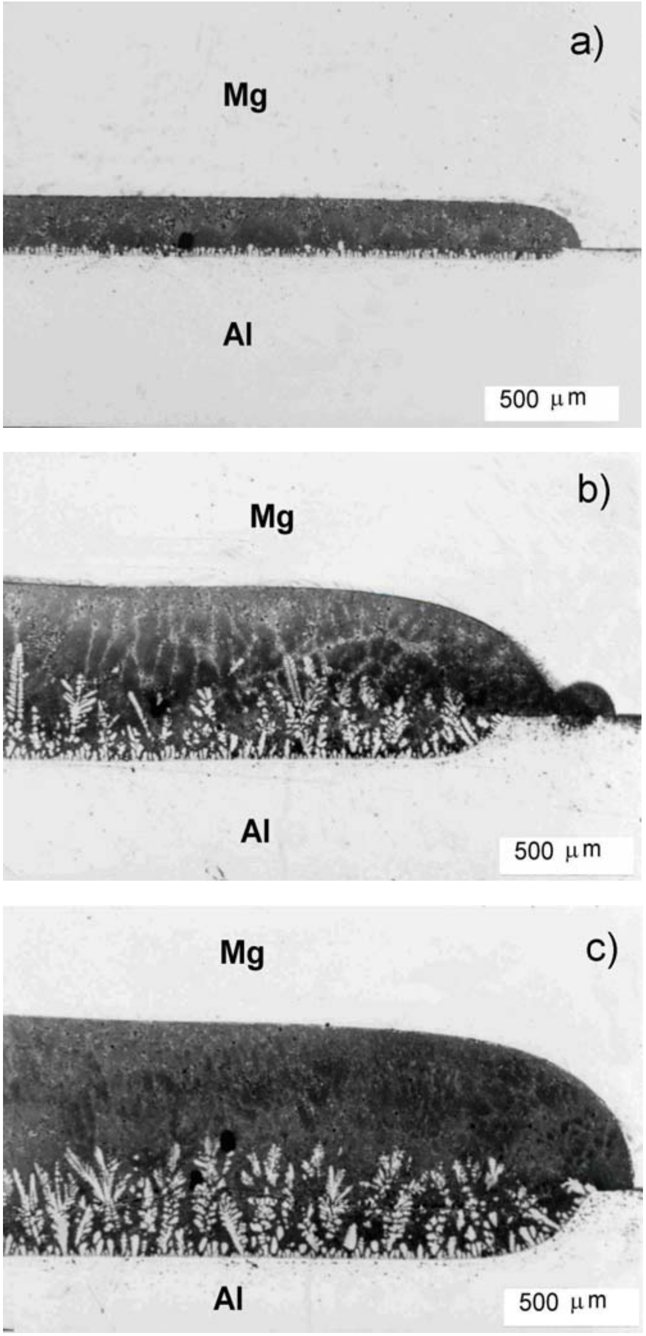

Fig. 3. Growing of the layer at the Mg-Al interface with time: 10min (a), 20min (b), 30min (c) at the temperature of 445^{0}C

Scanning electron microscopy and X-ray microprobe analysis were used for examination of the microstructure resulting from solidification. Figure 4 presents scanning electron microscope micrograph of the layer showed in the Figure 3a. Heterogeneous microstructure consists mainly of two-phase mixture. The details of the microstructure at the aluminium side and at the magnesium side are given in Figure 5.

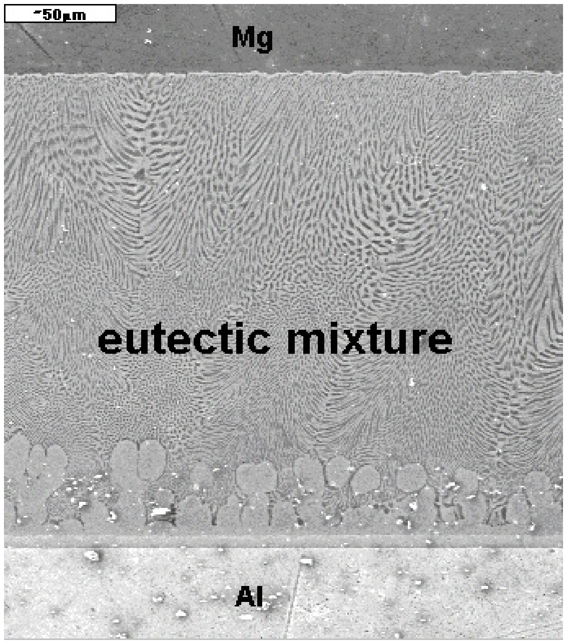

Fig. 4. Microstructure formed between Mg and Al metals after solidification of the melting zone. Mg-Al couple was annealed at 445 ^{0}C for 10 minutes and surface cooled

The elemental analysis performed by the X-ray spectroscope was made for areas marked A, B and C in the Figure 5a and Figure 5b. The chemical compositions of the layer marked A (in the neighbourhood of aluminium): 60.7 % at. Al and 31.3 % at. Mg suggests Al_3Mg_2 intermetallic compound. For dendrites (area marked B) it was found: 63.0 % at. Mg and 37.0 % at. Al. The Mg : Al ratio do not very differ from $Mg_{17}Al_{12}$ intermetallic compound. Chemical composition of the region marked C was 87.6 % at. Mg and 12.4 % at. Al. It suggests solid solution aluminium in magnesium. For eutectic structure X-ray radiation was simultaneously emitted by the two phases. On the basis of Mg-Al diagram and taking also into accound the results of X-ray analysis (79.7 % at. Mg and 20.3 % at. Al for dark phase and 66.7 % at. Mg and 33.5 % at. Al for bright phase) we are suggesting that heterogeneous structure is the eutecic mixture composed of $Mg_{17}Al_{12}$ intermetallic compound and solid solution of aluminium in magnesium.

Process of structural transformations with the liquid phase contribution that taking place at the Mg-Al interface can be useful for obtaining of layered composite. In a such case the reaction between alternately placed magnesium and aluminium sheets must lead to complete consumption of the aluminium sheets. Aluminium with a part of the magnesium sheets are transformed into the layers of composite consisting of eutectic mixture. Figure 6 presents scheme of the procedure used for the composite fabrication. Microstructure evolution for the early stage of the composite formation is shown in Figure 7.

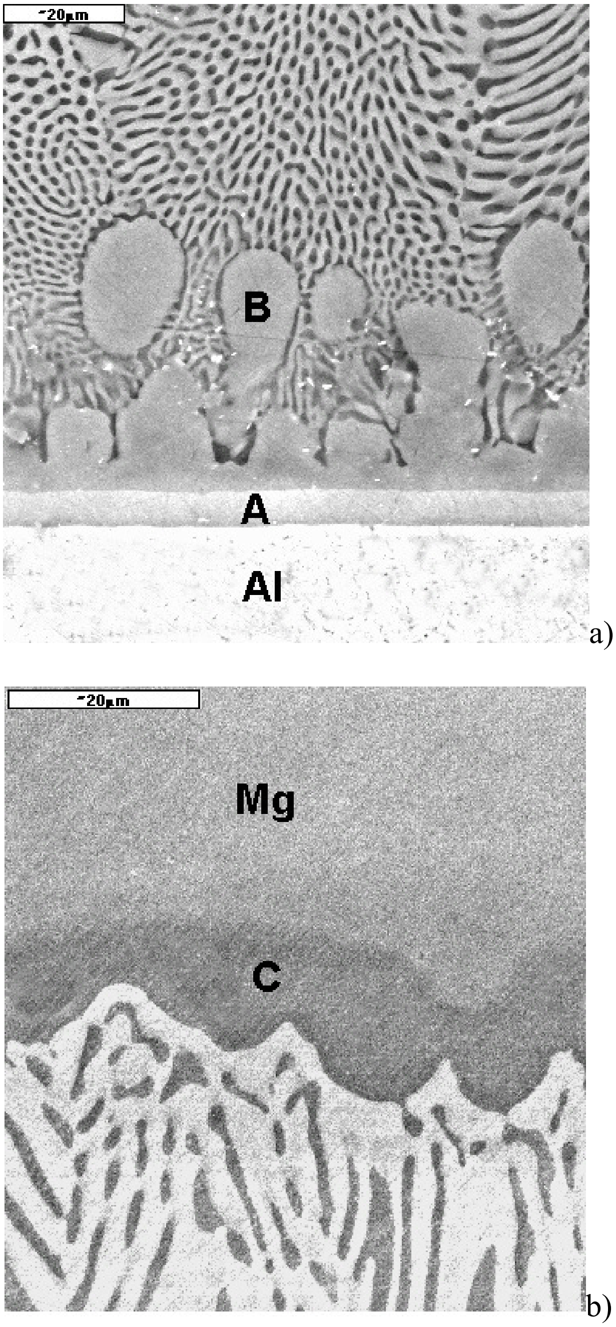

Fig. 5. Microstructure formed in the vicinity of the aluminium (a) and magnesium (b) substrates

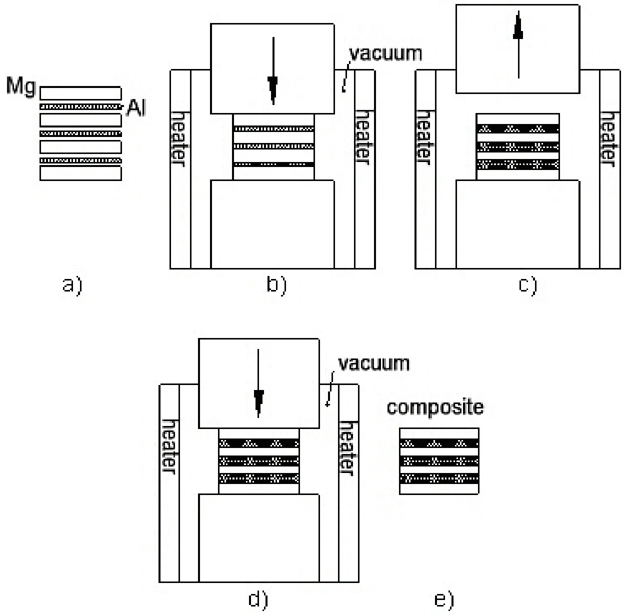

Fig. 6. Schematic presentation of the process used to form of the composite: a) Mg and Al sheets are stocked into a packet, b) diffusion bonding of the sheets, c) process with the liquid phase contribution, d) consolidation of the composite under pressure, e) magnesium-eutectic mixture composite

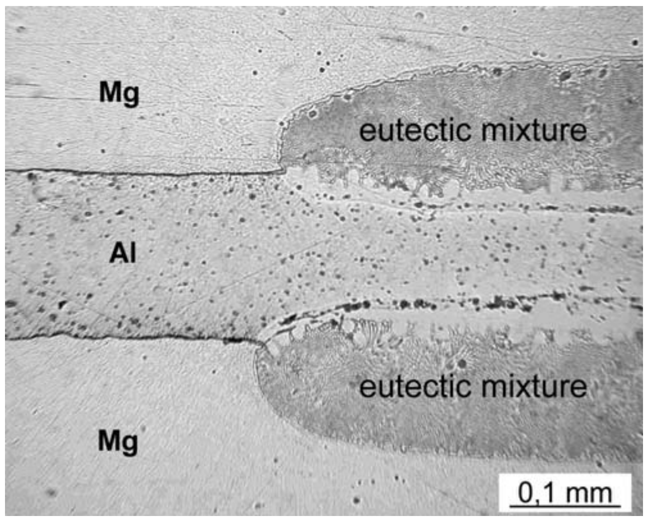

Fig. 7. Transformation of Mg and Al substrates into layer of composite containing eutectic mixture

Solidification produces microstructure that with concentration of Mg and Al plot is presented in Figure 8. The dominant part of the structure is the two-phase mixture. The chemical composition of the bright phase: 63.0 % at. Mg and 37.0 % at. Al is very close to equilibrium phase $Mg_{17}Al_{12}$. In the case of the dark phase result of X-ray microanalysis indicate on solid solution of aluminium in magnesium. This result suggests that structure formed during solidification is the eutectic mixture of the $Mg_{17}Al_{12}$ intermetallic compound and solid solution of aluminium in magnesium. Adjacent to magnesium homogenous zones marked δ in Figure 8 contain magnesium and aluminium. Concentration of aluminium is continuously changing from 11,7% at. to 1,7% at. at the magnesium side. This result indicates on solid solution of aluminium in magnesium.

By choosing the thickness of the Mg and Al sheets any ratio of Mg layers to layers containing eutectic mixture in the composite can be obtaining. Table 1 lists thickness of the starting sheets and final thickness of the composite layers.

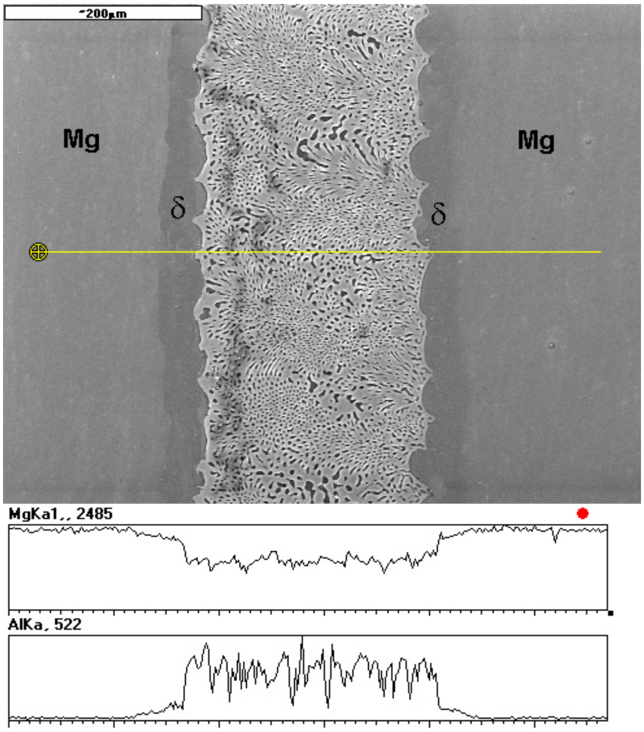

Fig. 8. Microstructure and concentration of the Mg and Al profiles across the composite layer

Table 1.
Thickness of starting metal sheets and final layers in fabricated composites

Starting Mg sheet thickness [mm]	Starting Al sheet thickness [mm]	Final Mg layer thickness in the composite [mm]	Final eutectic mixture layer thickness in the composite [mm]	Ratio of the Mg layers to the layers of eutectic mixture
1,45	0,06	1,25	0,25	5
1,4	0,1	0,9	0,4	2,3
1,35	0,15	0,7	0,6	1,2
1,3	0,2	0,5	0,8	0,6
1,25	0,25	0,3	1	0,3

It should be added that the structure formed at the Mg-Al interface is independent of the thickness ratio of the starting magnesium and aluminium sheets. An example of the magnesium-eutectic mixture composite presents Figure 9.

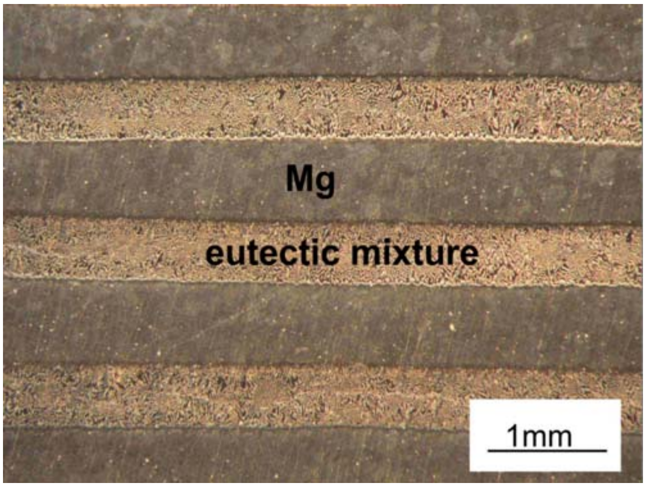

Fig. 9. Microstructure of the layered composite with the 1,2:1 thickness ratio of the magnesium layers to layers of the eutectic mixture

4. Conclusions

1. Due to solid state diffusion layers of $Mg_{17}Al_{12}$ and Al_3Mg_2 intermetallics compounds are formed at the Al-Mg interface.
2. The fast growing of the layer formed between magnesium and aluminium metals was observed with the liquid phase appearance at the Mg-Al interface. The dominant part of the layer is the eutectic mixture composed of $Mg_{17}Al_{12}$ intermetallic compound and solid solution of aluminium in magnesium.
3. It has been shown that magnesium-eutectic mixture layered composite can be produced from the magnesium and aluminium sheets formed alternately into a packed and heated to temperature of 445 ^{0}C.
4. By varying the thickness of the starting magnesium and aluminium sheets any ratio of the thickness of composite layers can be obtained.

Acknowledgements

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