

# Formation of Intermetallic Phases in Al/Cu Compound Casting Process

## Abstract

The purpose of this article is to study the growth rate of intermetallic compounds at the welded interface of Al/Cu bimetal were produced by compound casting process. The mechanism of the intermetallic compounds (IMCs) formations, the effects of aluminum pouring temperature and copper preheating temperature on the IMCs types and thickness were investigated and Al/Cu interface microstructure, were characterized by optical microscope (OM) and electron probe micro-analyzer (EPMA). Results show that the interface is consist of three main layers, the first Layer (I) is  $\alpha$ -Al/Al<sub>7</sub>Cu eutectic structure, the second layer (II) is Al<sub>7</sub>Cu and the third layer (III) consists of the several intermetallic compounds such as AlCu, Al<sub>7</sub>Cu<sub>5</sub>, Al<sub>7</sub>Cu<sub>6</sub>, Al<sub>5</sub>Cu<sub>4</sub>. The first layer was formed by Al and Cu dissolving in liquid phase and rapid solidification, then the second layer II was formed by nucleation and growth mechanism at solid/liquid interface and finally the layer III was formed by solid-state phase diffusion. Raising the Al melt pouring temperature and preheating Cu leads to increase of the intermetallic compounds thickness at interface and consequently increases the specific electrical resistance and decreases the Al/Cu bond strength. From experiments, it is proposed that the bond strength is dominated by the thicknesses of layer II and III.

**Key word:** Compound casting, Al-Cu bimetal, intermetallic compounds, bond strength, IMCs hardness, electrical resistivity.

## 1- Introduction

Two or multi-layer metallic composites due to their striking characteristics have been developed rapidly[1].

The Al-Cu bimetal have advantages of Cu and Al in a bulk composite that offers simultaneously copper high conductivity and lower cost of aluminum at a same electrical conductivity, the bimetallic rod is 30-40% cheaper and 40-60% lighter than a copper rod [2].

The core Al clad Cu rod due to “skin effect” of Cu shell transfer main electric current through the shell surface and is used for conductor to transfer high frequency and high power electricity [3]

Different processes for fabricating Al/Cu bimetals have been applied , such as Diffusion welding [4-6], Deep drawing [7,8], Cold rolling [9-11], Extrusion welding [12-14], Explosive welding[15-17] that all processes belong to the category of Solid-Solid bonding technology.

Solid state joining processes have some limitations. In summary, difficulty in preparing composite conductors, restrictions in geometry and dimensions, long process time, high operating cost may render these solid-state process as not easy for practical and industrial applications [18].

Compound casting is defined as a process in which two metallic materials or two alloys that one in the solid state and the other in a liquid state are brought into contact with each other that belong to the category of Solid-Liquid bonding [14]. In this method, formation of metallurgical bond at the interface is under fusion and diffusion reaction. Part of elements diffusion cause to form of solid solutions and the other part cause to form a reaction zone and formation of different phases at the interface.

From the point of view of welding process, Al and Cu are incompatible metals and they have a high affinity to each other in temperature greater than 1200°C and produce brittle, low strength intermetallic phases with non-metallic bond between Al and Cu [9]. Producing of these intermetallic phases and interatomic bonds between aluminum and copper decreases the number of available free electrons and increases the electrical resistivity, moreover this phenomenon leads to reducing the bond strength and flexibility, thus controlling the intermetallic phases growth at interface to find the critical thickness of interface layer for producing optimal mechanical, physical and electrical properties is important.[15-17]. Also, in the previous studies it was found that the phase composition and microstructure of Al-Cu intermetallic phases have a significant effect on the physical and mechanical properties [9].

According to the binary phase diagram of Al/Cu shown in Figure 1, different intermetallic phases are formed depending on the temperature and chemical composition [18].

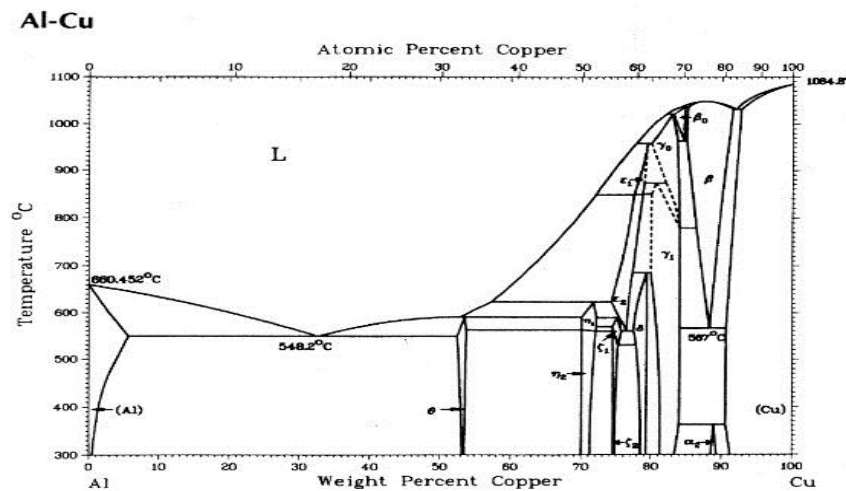


Fig. 1. Al-Cu phase diagram [18]

Neumann [20] in 1969 invented a vertical core-filling casting method to fabricate the metal cladding materials. This method is applicable to most cladding metals where the core has lower melting point.

Divandari and Vahid Golpayegani in 2009 [21] have put a copper wire into polystyrene pattern and casted Al<sub>3</sub>Si Al alloy into and showed if the matrix is Al alloy, in the small area near to the inserted Cu wire, there is a possibility of forming Al-Cu intermetallic phases. They reported that

intermetallic phases such as Al<sub>7</sub>Cu (θ), Cu<sub>3</sub>Al<sub>2</sub> (δ), AlCu (η<sub>2</sub>) and Al<sub>7</sub>Cu/Al(Cu) eutectic and Si particles was detected.

Zare et al. [11] in 2013 and other previous authors [12-13], have been reported that some intermetallic phases was characterized such as Al<sub>7</sub>Cu, AlCu, Al<sub>7</sub>Cu<sub>2</sub>, Al<sub>2</sub>Cu<sub>3</sub> and α-Al/Al<sub>7</sub>Cu.

In the present study, the effects of casting parameters such as Al casting temperature and Cu preheating temperature on the structures and intermetallic phases growth are studied. Also the effects of these factors on physical and mechanical properties of Al-Cu bimetal are discussed.

## 2- Materials and experiments

The composition and hardness of Cu and Al raw materials are measured by Optical Emission Spectroscopy (OES) and microhardness test that are summarized in Table 1.

**Table 1**

### Specification of Copper and Aluminum

Cu and Al grade	Component	Chemical Composition	Hardness(HV)
Cu-UNS C11000	Sheath	99,9% Cu, 0,01% P, 0,01% O	82
Al-UNS A91000	Core	99,0% Al, 0,05% Zn, 0,05% Mg, 0,05% Mn, 0,05% Fe, 0,05% Si	38

The copper-clad aluminum rod was produced by static casting of Al into solid Cu tube with 1 mm thickness and 200 mm length.

At first, the inner surface of copper tube was cleaned by degreasing and 10% nitric acid solution to remove the oxide films and contamination. After preparing the surface, a Cu tube was mounted vertically as a casting mold for pouring molten Al into it. The Al melt was protected by borax flux to prevent surface oxidation. The copper tubes was preheated by resistance heating element and temperature was controlled by K-type thermocouple.

Finally molten Al was poured by static method and after 10 second the Copper-clad aluminum wire was cooled to the environment temperature by burying in wet sand.

To study of intermetallic phases growth, several experiments were carried out at Al temperatures of 700, 750, 800°C with tolerance of ±10°C and preheating Cu temperature of 20, 100, 200, 300, 400°C with tolerance of ±0°C. In the produced samples with Al temperature of 800°C and preheating temperature of Cu above 220°C, the high heat content of interface, has led to local melting, shell piercing and destruction of samples. Results of tests on the samples are given in table 2.

**Table 2**

**Obtained characteristics by changing the temperature of the molten Al and solid Cu preheated.**

Sample	Al (°C)	Cu (°C)	Intermetallic layer width (μm)	Layer III width (μm)	Specific resistance of bimetal (Ωxmm <sup>2</sup> /m)	Bond strength (N/Cm)	Phase detected by EPMA
A700C20	700	20	930	6	0,0308	120	α+θ - Al <sub>7</sub> Cu-AlCu-Al <sub>2</sub> Cu <sub>3</sub>
A700C40	700	40	3100	9,0	0,0398	26	α+θ-Al <sub>7</sub> Cu-AlCu-Al <sub>7</sub> Cu <sub>7</sub> -Al <sub>2</sub> Cu <sub>3</sub>
A700C20	700	20	900	7	0,0299	88	α+θ - Al <sub>7</sub> Cu-AlCu-Al <sub>2</sub> Cu <sub>3</sub>
A700C40	700	40	4000	12,0	0,044	14	α+θ-Al <sub>7</sub> Cu-AlCu-Al <sub>7</sub> Cu <sub>7</sub> -Al <sub>2</sub> Cu <sub>3</sub>
A800C20	800	20	980	7,0	0,0310	82	α+θ - Al <sub>7</sub> Cu-AlCu-Al <sub>2</sub> Cu <sub>3</sub>
A800C22	800	22	0100	14	0,002	11	α+θ-Al <sub>7</sub> Cu-AlCu-Al <sub>7</sub> Cu <sub>7</sub> -Al <sub>2</sub> Cu <sub>3</sub>

Thickness, types and the IMCs formation mechanism of Al-Cu interface layer were studied by optical and electron probe micro-analyzer (EPMA) with point and linear scan analysis. EPMA analysis is based on the accuracy and reliability of WDS, high vacuum and stability of electron beam with beam size of 1 to 3 microns and the tolerance of 3% that has much higher sensitivity and accuracy than using SEM/EDX to identify the phases for studying microstructure.

Microhardness test of Al-Cu intermetallic compounds was conducted with a testing load of 10g and holding time of 10s. Vickers hardness (VH) was calculated in terms of kgf/mm<sup>2</sup>

The electrical resistance of Al-Cu bimetal was measured by micro-ohmmeter Omicron resistance testing machine equipped with accuracy of 0.01 micro-ohm. A direct certain current (I=1A) is passed from the sample and the potential difference between two certain points was calculated automatically by dividing the difference of potential to passing current as relation (1)

$$R = \Delta V / I \quad (1)$$

Finally the resistivity (ρ) was calculated by the resistance (R), length (L), and thickness cross section area (s) of the sample and following relation:

$$\rho = R \cdot S / L \quad (2)$$

For evaluating the bond strength of samples, a 1000 N load cell with accuracy of 0.01 N was used.

At first two grooves with a distance of 1 cm were created in the longitudinal direction of the sample. Then bond strength was measured by mean peeling force of Cu layer from Al and was calculated using the following relation:

$$F_b = \frac{\bar{F}}{L_b} \quad (2)$$

## 2- Results and discussion

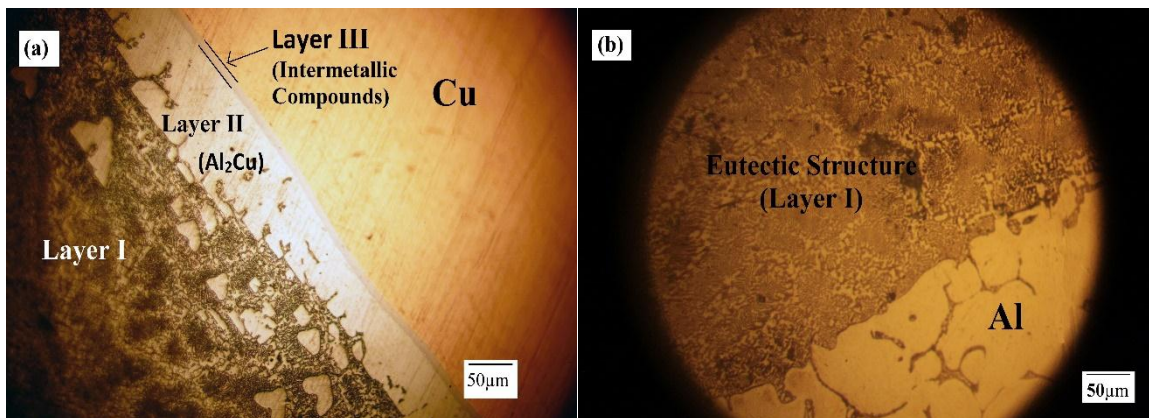
### 2-1- Microstructure

Latent heat content of liquid Al transferred to the solid-Cu, when the liquid aluminum contact with solid copper. If the heat content is enough, the Cu atoms from the inner surface of the copper tube diffused in liquid Al and forming the Al-Cu solution. In the solidification process, dissolving of cu atoms decreases gradually in Al liquid and according to the binary diagram of Al-Cu (Fig. 1), intermetallic compounds at the interface will be formed. So temperatures of molten Al and solid Cu are the main factors which are directly effective in the heat content of the interface of Al/Cu.

Optical and electron microscope observations of the Al/Cu interface in figures 2, 3, 4 and 5 show the multilayered interface between the copper sheath and aluminum core. Results show that along the radial orientation from the Al to Cu, the interface is divided into three main layers as below:

Layer I is  $\alpha$ -Al/Al<sub>2</sub>Cu ( $\alpha+\theta$ ) eutectic structure, layer II is Al<sub>2</sub>Cu and layer III consists of the several intermetallic compounds. The different IMCs in layer III have been detected by changing in main processing parameters e.g. aluminum pouring temperature and copper preheating temperature such as AlCu, Al<sub>2</sub>Cu<sub>3</sub>, Al<sub>3</sub>Cu<sub>2</sub>, Al<sub>4</sub>Cu<sub>3</sub>.

Fig 2, indicate the microstructure of the interface of Al/Cu bimetal rod in A<sub>70</sub>C<sub>30</sub> sample that has been prepared under 700°C Al and 300°C Cu, respectively. In Fig 2 the three main layers is shown separately.

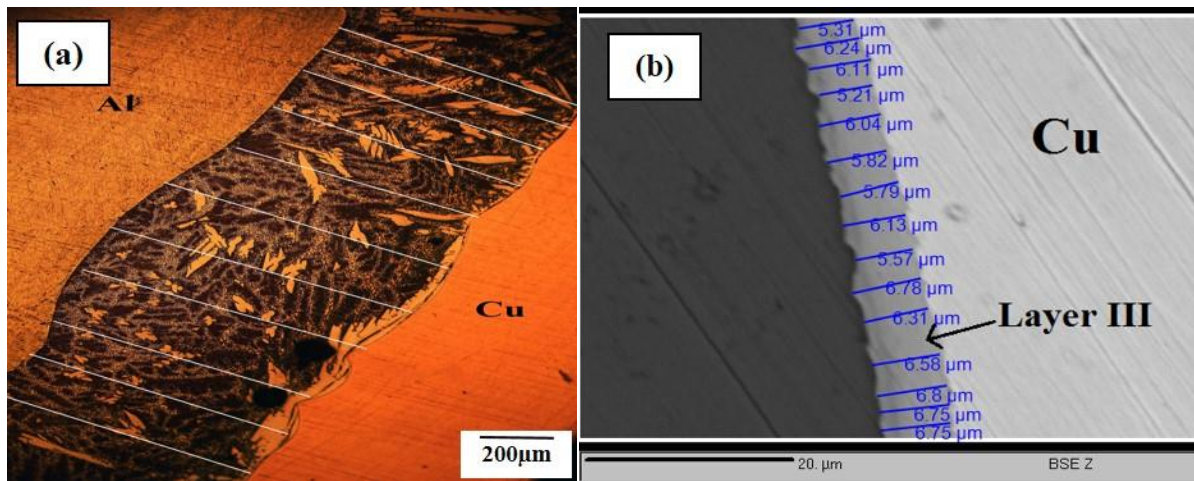


**Fig. 2. three main layers of I, II, III in sample A<sub>70</sub>·C<sub>30</sub>. a) Microstructure of Cu-side. b) Microstructure of Al-side**

**2-2- The thickness of the interface layer**

In compound casting, the interface is formed at high temperature, therefore Cu atoms can diffuse in Al melt with an appropriate atomic diffusion rate.

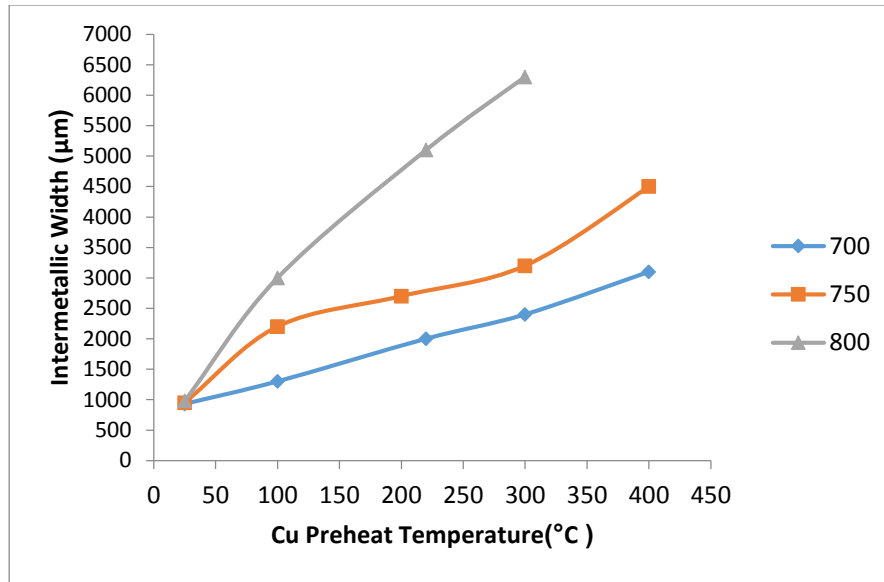
Fig 2 (a) show the interface thickness of A<sub>70</sub>·C<sub>30</sub> specimen that was casting in 700°C aluminum temperature into copper tube without any preheating. Fig 2(b), illustrate the thickness of layer III in A<sub>70</sub>·C<sub>30</sub> specimen.



**Fig. 2. a) a part of reaction layer of A<sub>70</sub>·C<sub>30</sub> with an average thickness of 602.43 μm. b) layer III of A<sub>70</sub>·C<sub>30</sub> with an average thickness of 6.146 μm**

The thickness of interface of Al-Cu bimetallic samples produced at pouring aluminum temperature of 700, 750, 800°C and preheated copper temperature at 20 to 400°C are plotted in Fig 3.

Compared in diagrams (Fig 3), samples that were casting in 800°C Al have the highest growth rate of interface thickness. With raising the preheating of copper tube to 300°C, the thickness of interface was increased to 6300 μm



**Fig. 4. The variation of thickness of interface versus preheating Cu temperature in Al-melt of 700, 750, 800°C**

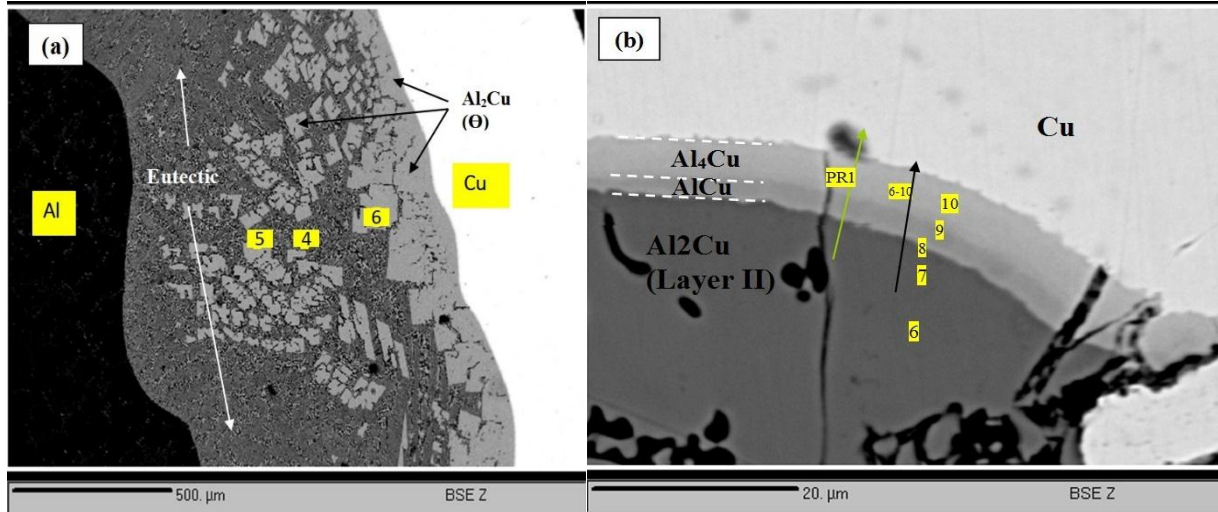
Table 3 shows the measured thickness of layer III in produced samples. According to the experiment results in table 3, it can be seen that compared to preheat Cu tube, increasing of Al pouring temperature has a less effect on the thickness growth of interface in compound casting process. Indeed latent temperature of interface is function of melt and solid components that raising each one of these factors can lead to increase of heat content of interface. Also, due to the higher thermal conductivity of Cu ( $401 \text{ W.m}^{-1}.k^{-1}$ ) than Al ( $237 \text{ W.m}^{-1}.k^{-1}$ ), increasing the Cu preheating temperature could have a greater effect on the heat content of interface. Consequently increasing the Al melt temperature without any Cu preheating, cause to growth of interface gradually. Previous studies found that the Cu atoms have a smaller radius ( $128 \text{ nm}$ ) than Al atoms ( $143 \text{ nm}$ ), then obviously, it is easier for smaller atoms (Cu) to diffuse into a region of larger atoms. On the other hand, the melting point of Cu is higher than that of Al, making it harder to break the bonds between Cu atoms than those between Al atoms, making it more difficult for Al atoms to diffuse into the Cu lattice. In contrast, the bonds in Al are weaker and forming vacancies is an easy task in comparison with Cu [14]. All mentioned factors favor the diffusion of Cu atoms into Al. thus, increasing the temperature of Cu can rouse to action more Cu atoms for diffusing in Al-Cu interface layer.

### 3-3- Intermetallic compounds

The EPMA point and linear scan analysis is used to detect of layers, phases and changing elements concentration across the intermetallic width and layers, respectively.

In layer III for all samples that were pouring with Al melt temperatures of 700, 750, 800°C without any preheating Cu tube (25°C), the EPMA analysis has identified two intermetallic compounds consist of AlCu and Al<sub>2</sub>Cu<sub>3</sub>.

Fig 10 shows the EPMA images of  $Al_{70}Cu_{30}$  and  $Al_{50}Cu_{50}$  specimens that results are summarized in table 1. In Fig 10(a), based on the EPMA analysis, moreover the sides region (layer II),  $Al_7Cu$  ( $\theta$ ) intermetallic compound could be observed in some areas as scattered islands. Fig 10(c) indicate the line scan analysis of Cu and Al elements that labeled as a PR1 line in Fig 10(b). Due to changes in concentrations gradient of Cu atoms diffusion diagrams, 4 distinct areas have been identified consist of  $Al_7Cu$ ,  $AlCu$ ,  $Al_3Cu_2$ , Cu from Al towards Cu, respectively. Thickness of layer III was obtained  $7.0 \mu m$  that  $2.3 \mu m$  is belonged to  $AlCu$  ( $\eta$ ) and  $4.7 \mu m$  is belonged to  $Al_3Cu_2$  ( $\gamma$ ).



(d)	EPMA quantitative analysis (Wt%)		
No.	Al	Cu	Phase
3	79.08	20.92	$\alpha$ - $Al_7Cu$
4	71.04	28.96	$\alpha$ - $Al_7Cu$
5	47.04	52.96	$Al_7Cu$
6	47.28	52.72	$Al_7Cu$
7	47.09	52.91	$Al_7Cu$
8	48.11	51.89	$Al_7Cu$
9	34.01	65.99	$AlCu$
10	16.06	83.94	$Al_3Cu_2$

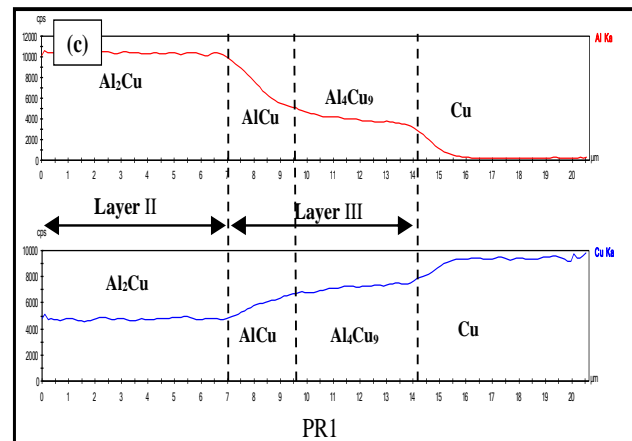


Fig. 10. a) layer I and II of  $Al_{70}Cu_{30}$  sample, b) layer II and III of  $Al_{50}Cu_{50}$  sample consist of  $Al_7Cu$ ,  $AlCu$ ,  $Al_3Cu_2$ . c) Al and Cu line scan across the PR1 line. d) Quantitative analyses of points 3 to 10 (wt%)

$Al_{70}Cu_{30}$  and  $Al_{50}Cu_{50}$  samples have been pouring with  $500^\circ C$  and  $500^\circ C$  Al melting temperature and  $400^\circ C$  preheating Cu, respectively. In this samples, intermetallic compounds of  $\alpha$ - $Al/Al_7Cu$ ,  $Al_7Cu$ ,  $AlCu$ ,  $Al_3Cu_2$  and  $Al_3Cu_2$  have been identified that summarized in table 1. Fig 11 illustrates the layer II and III in  $Al_{50}Cu_{50}$  sample. In this case, moreover  $Al_7Cu$ ,  $AlCu$  and  $Al_3Cu_2$  an intermetallic phase of  $Al_3Cu_2$  ( $\zeta$ ) was detected in point 10 (Fig 11a). This suggests, perhaps Al and Cu temperatures and interaction time in other produced samples was not appropriate for forming  $\zeta$  phase.



Fig 6(b) shows the linear scans of Al and Cu elements along the PR1. Due to low change in incline of concentration gradient, detection of phases boundaries in layer III are difficult which thicknesses of AlCu, Al7Cu4, Al7Cu7, Al3Cu4 was estimated 4,42, 2,39, 3,42, 3,70 μm, respectively. Intermetallic compounds were detected by EPMA analysis in 6 samples are shown in table 7.

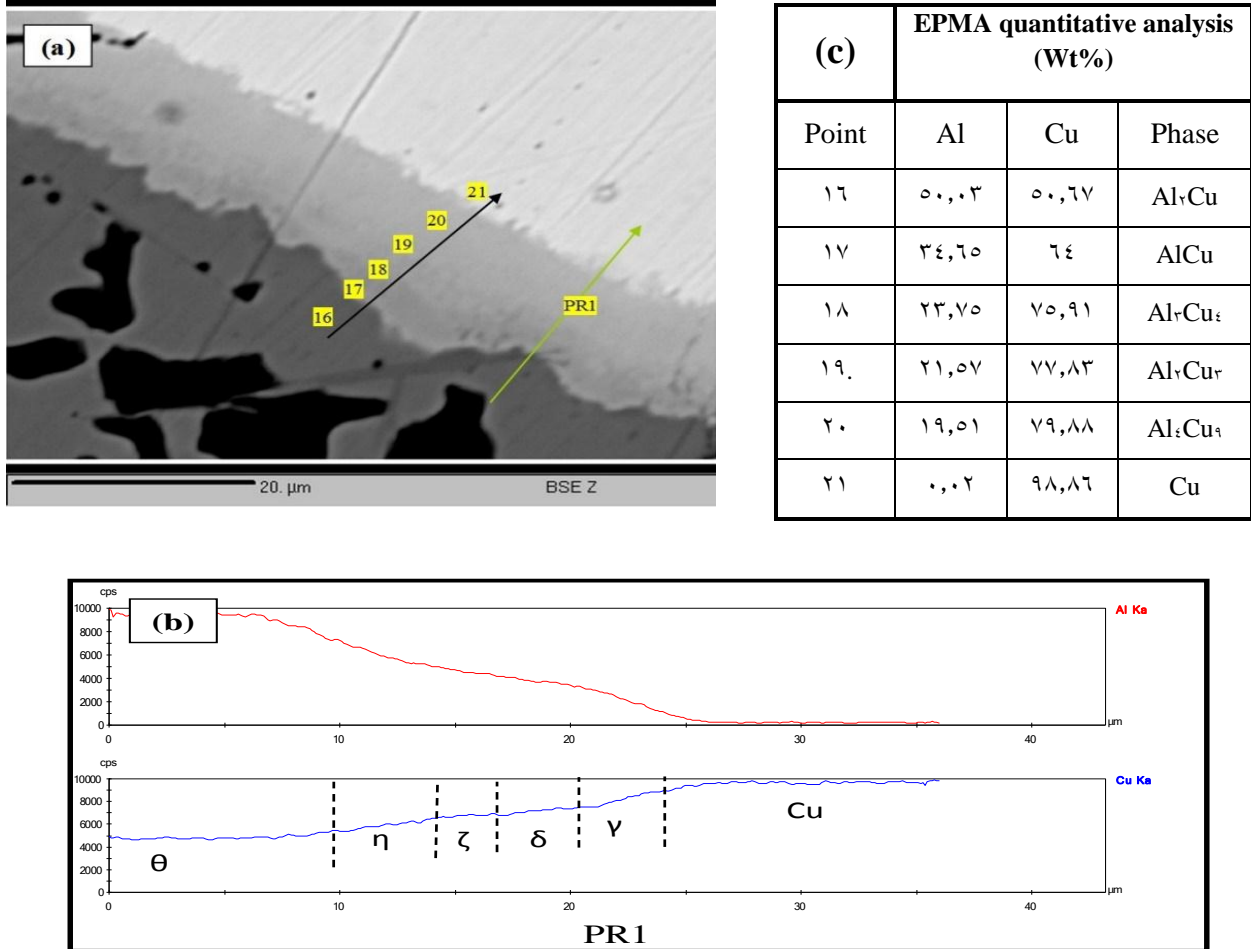


Fig.6. Microstructure of Al-Cu sample, a) EPMA image of interfacial microstructure. b) Al and Cu line scan across the PR1 c) Quantitative analyses of points labeled in part a. (wt%).

### 3-4- The formation mechanism and phase transformation of interface

According to point and linear EPMA analysis, results illustrated that Al content was reducing in the radial direction of Al core towards Cu sheathed. Also the analysis suggested that the Al content in Cu sheathed or Cu content in Al-core is low. This indicates that the diffusion reaction between aluminum and copper occurs just in the range of the interface.

#### 3-4-1- Formation of layer I:

With respect to the Al-Cu binary phase diagram, Cu can dissolve 0.10 Wt% in Al in temperature of 0.48, 7°C in an equilibrium condition and can form the solid solution, but much more Cu atoms cannot enter the solid solution, therefore begin to form intermetallic compounds.

When the temperature of interface reduces below the eutectic temperature (0.48, 7°C), in a wide concentration range of 0.10 Wt% < W<sub>cu</sub> < 0.2, 0 Wt%, Al-Cu binary alloy could form an eutectic phase consisting of  $\alpha$ -Al phase and Al<sub>3</sub>Cu phase at room temperature. It was labeled as a layer I. As known from results of analysis, layer I as a eutectic layer took place as the thickest part of interface. Thus, it can be assumed that layer I was formed through copper dissolving from the inner surface of the Cu sheath into liquid Al and then with nucleating of  $\alpha$  and  $\theta$  at interface and decreasing internal energy and entropy of the system, eutectic reaction of Al-Cu binary alloy during the cooling and solidification processes has occurred. Therefore, forming mechanism of layer I is a combined action of the melting, dissolution and solidification. Aluminum is an active metal that has a corrosive mode in liquid state, for this reason, at first, the Cu atoms in the solid phase, dissolve quickly in liquid Al and then eutectic structure ( $\alpha$ + $\theta$ ) forms by rapid solidification.

Factors such as interaction time, temperature of solid Cu and liquid Al are impressive on thickness of layer I. In other words, the higher the interface temperature is or the longer the interaction time is, the more Cu atoms dissolve into Al melt. Therefore, either raising liquid Al temperature or preheating Cu tube could cause increasing the thickness of interface and in particular, lead to increasing layer I thickness, significantly.

### 3-4-2- Formation of layer II

Considering the fact that the activation energy of Al<sub>3</sub>Cu in order to initiate crystallization between Al-Cu intermetallic compounds is at the lowest degree [11, 15]. In the meanwhile, crystallographic studies [15] could also be suggestive of sequence of phase nucleation. Thus it was understood that in general, high symmetry alloys with small unit cells will easily crystallize and nucleate, while long-range ordered with low symmetry phases are less likely to form during bonding process. Therefore, it is assumed that, the tetragonal Al<sub>3</sub>Cu phase with small unit cell and low packing factor ratio between identified phases, are anticipated to readily begin to nucleate and grow as a first solidified phase after rapid solidification of eutectic layer.

Since the layer II is thinner than its adjacent eutectic layer ( $\alpha$ -Al<sub>3</sub>Cu) and there was no distinct concentration gradient with Al-Cu ratio in linear scan (Fig 3c and Fig 3b), therefore, we can place attention to the layer II that formed through the nucleation and growth mechanism of Al<sub>3</sub>Cu phase from the inner surface of the Cu sheath to layer I just after layer I solidified. On the contrary of layer I that was formed by melting-solidification process, layer II is a diffusion-control layer that needs much more time to diffusing and growing up.

Therefore, the growth time of Al<sub>3</sub>Cu phase was determined by cooling rate, then it can be concluded that probably the higher cooling rate results in less thickness of interface.

In the all present casting samples with constant cooling process conditions to reach room temperature (embedding the copper clad aluminum into wet sand after 10 s from the end of casting process) thickness of Al-Cu (layer II) was between 10 to 200 μm approximately.

### 3-4-3- Formation of layer III

Layer III, which is distinguished in all samples, consists of several intermetallic compounds. It was very thin and has an approximately the same thickness values in all regions where is created, with straighter boundaries on both sides. There was a radial concentration gradient (Fig 9c, Fig 10b) which compared to layer II with no concentration gradient implied that the formation mechanism of these layers could be different from each other. In the linear scan of PR1 in Fig 9, from Al-core towards Cu-sheath, Cu content increased in layer III and still remained constant in layer II (Al-Cu). It is supposed that Cu atoms diffused continuously into the layer II after the Al-Cu phase solidified and leads to a solid-state phase transformation from θ phase to other intermetallic compounds such as Al<sub>2</sub>Cu, Al<sub>3</sub>Cu, Al<sub>4</sub>Cu, Al<sub>5</sub>Cu<sub>2</sub>

Therefore, it is assumed that intermetallic compounds in layer III were formed through diffusion and solid-state phase transformation. Therefore, regarding the mentioned matters, the cooling rate that here has been fixed, could be one of the controlling factor of thickness of layer III.

Because layer I was formed by eutectic reaction and α-Al-Cu phase could create simultaneously in a wide range of 0.6Wt% < W<sub>Cu</sub> < 2.0Wt%, layer I was the layer with the most thickness. The solid-state phase transformation of layer III induced by continues diffusion of Cu into Al-Cu layer after layer II solidified and since the diffusion coefficient of metal atom in solid metal is 10 to 10<sup>6</sup> orders smaller than in liquid metal [10] and the diffusion time is short, so the layer III was the smallest.

In fact, by starting of nucleation and growth mechanism of the Al-Cu phase, diffusion of Cu atoms towards the Al side became limited. On the other word, solid Al-Cu phase is rolled as a barrier for diffusing Cu atoms into the interface layer.

Fig 10, shows the schematic mechanism and priority formation of three main layers in the interface which are summarized as follows:

First of all, at high temperature of process, Cu atoms rapidly dissolved from the inner surface of the Cu sheath into Al-melt and formed an area of Al-Cu binary alloy between the Al and Cu. After beginning the solidification process, forming θ nucleation in molten prepares the condition for eutectic solidification. At first, eutectic layer formes by rapid solidification as the thickest layer of interface. Just after that, θ phase due to the lowest activation energy between identified phases, forms on the inner surface of Cu tube by the diffusion control process that requires adequate time and temperature and grows toward the Al-core. Finally the Cu atoms continuously diffuse from surface of Cu to Al through the solidified Al-Cu phase and as a result of the interdiffusion of Cu

and Al,  $\theta$  phase transform into other intermetallic compounds such as  $\text{AlCu}$ ,  $\text{Al}_7\text{Cu}_2$ ,  $\text{Al}_3\text{Cu}_4$  by the solid-state phase transformation.

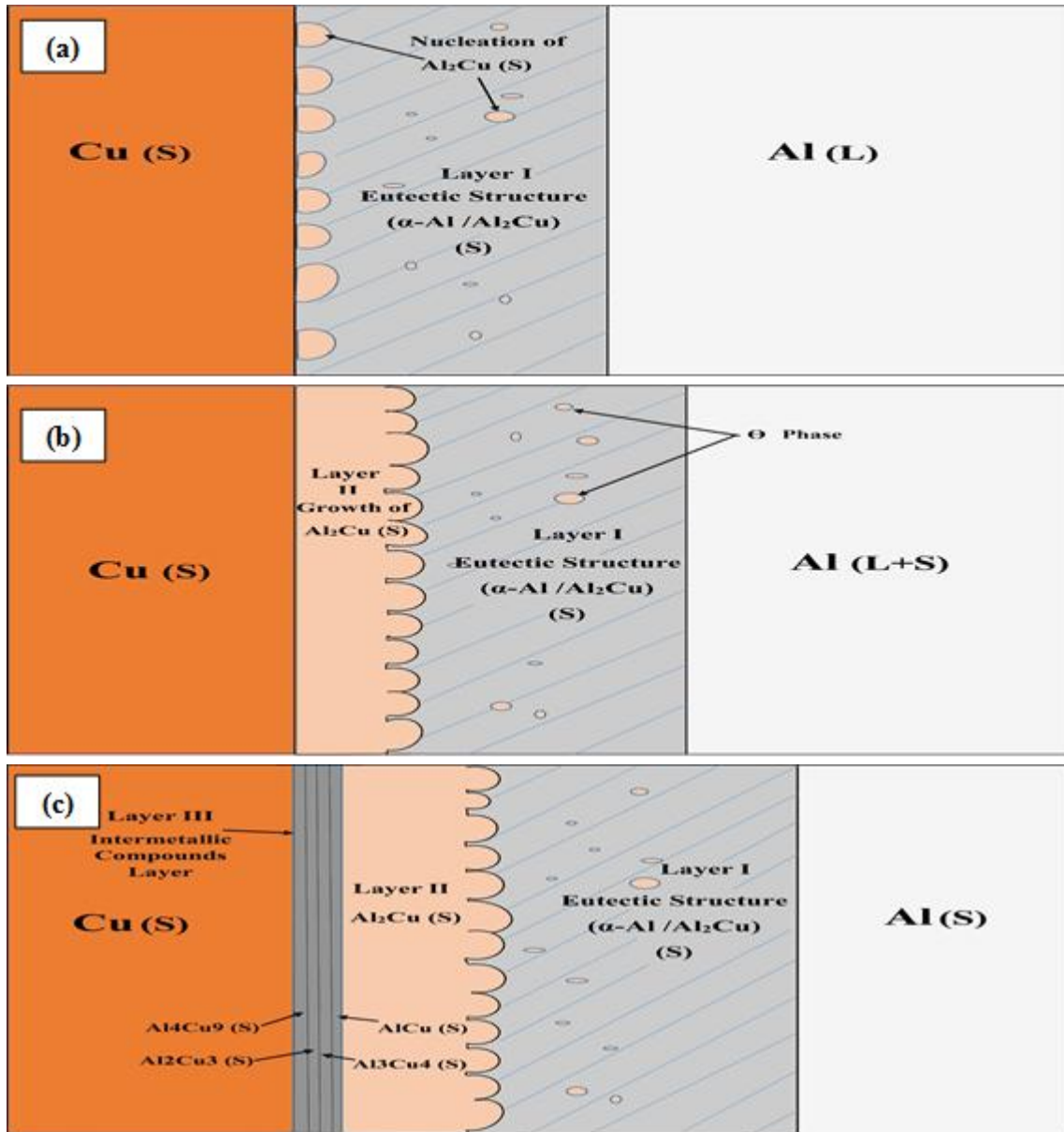


Fig.V. schematic image of the forming of interface of Al-Cu bimetal by static casting process.

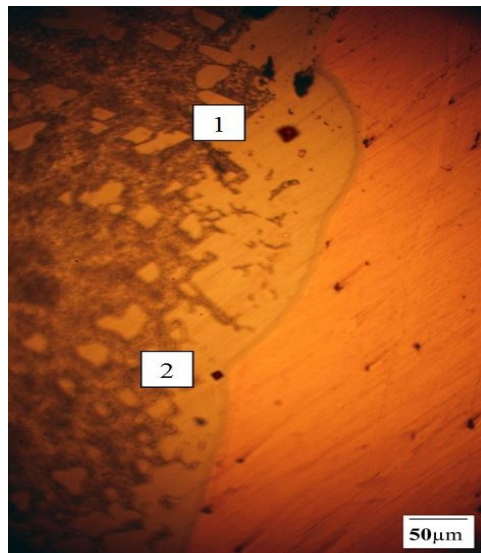
### 3.2. Microhardness

Results of hardness test illustrated that formed intermetallic compounds at interface possess higher hardness values than those of the corresponding base metals (Al and Cu). In the meanwhile, hardness values of intermetallic compounds that have been existed in layer III have the maximum values that could have a detrimental effect on the mechanical properties of Al-Cu bimetal.

Results of microhardness test and EPMA analysis at the interface of Al-Cu bimetal could be estimated the hardness values of three main layers from Al-side to Cu side respectively, as below: Vickers hardness with  $\text{Kgf/mm}^2$  unit for Al is in the range of 30 to 40, eutectic structure is in the range of 100 to 200,  $\text{Al}_2\text{Cu}$  is in the range of 400 to 500, AlCu is in the range of 800-1000, mixture of AlCu+ $\text{Al}_2\text{Cu}$  are in the range of 900 to 1000, mixture of  $\text{Al}_2\text{Cu}$ + $\text{Al}_2\text{Cu}_3$  are in the range of 1100 to 1200 and the Cu is in the range of 70 to 80.

The intermetallic compounds in layer II and III exhibit much higher hardness than that of eutectic structure, which could implies for lower fracture toughness.

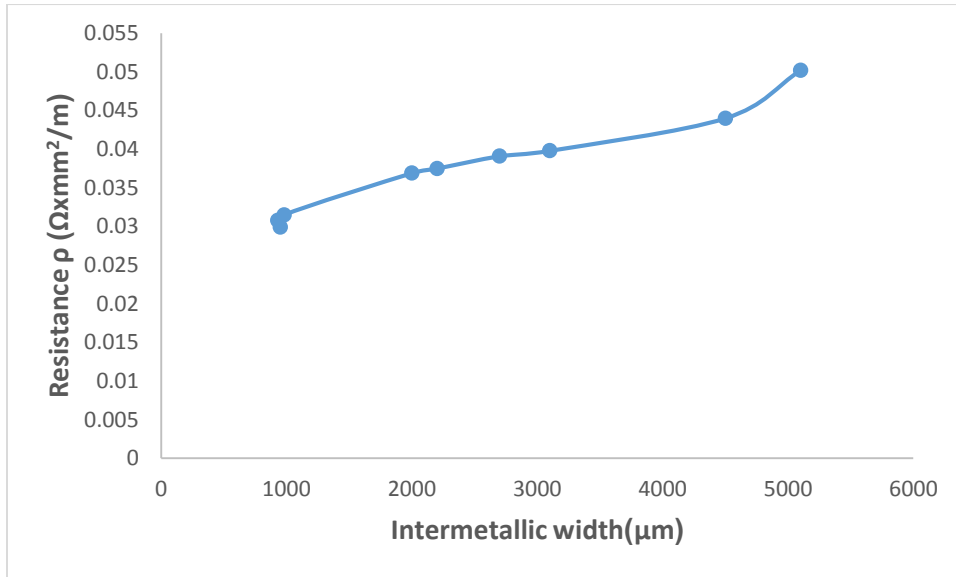
Due to the higher hardness and brittle properties of these pure intermetallic compounds compare to eutectic and base metals, layer II and III imply weaker plasticity and elasticity abilities. Fig 1, illustrate the hardness impression in the sample  $\text{Al}_{100}\text{Cu}_{20}$ . Regarding the impression of indenter, the points of 1 and 2 indicate the hardness values of the  $\theta$  ( $44\text{ HV}$ ) and  $\eta$  ( $99\text{ HV}$ ), respectively.



**Fig. 1. Microhardness test impression in  $\text{Al}_{100}\text{Cu}_{20}$  sample, point 1 in  $\theta$  phase area insert, point 2 in  $\eta$  area insert**

### 3-6- Resistance of bimetal samples

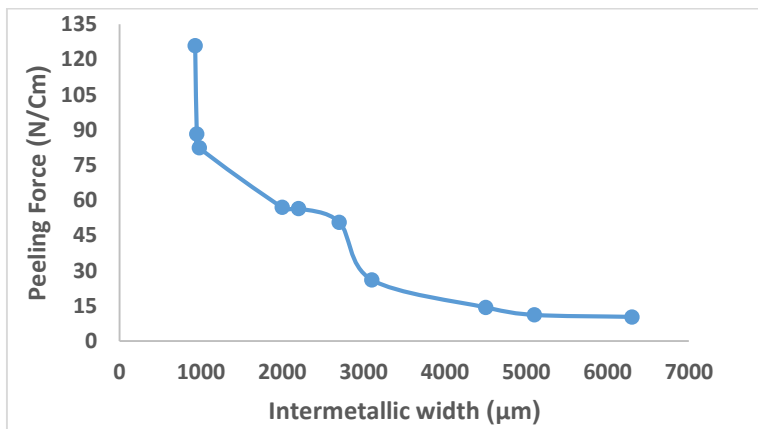
The results of change in resistance of the samples with increasing intermetallic thickness are presented in Table 3 and plotted in Fig 9. Referring to this figure, conductivity decreases and specific resistivity increases with increasing the intermetallic width.



**Fig. 9. The variation of specific electrical resistivity versus thickness of interface.**

### 3-4- Bond strength

Bond peeling strength of specimens could be seen in Table 3. The variation of bond peeling strength versus thickness of intermetallic layer is plotted in Fig 10. As can be seen, the strength of bond is decreased with increasing the thickness of interface.



**Fig. 10.**

**Fig. 10. The variation of bond strength versus thickness of interface.**

When the interfacial temperature fell below the eutectic temperature of Al-Cu alloy [ $541^{\circ}\text{C}$ ],  $\alpha\text{-Al/Al}_2\text{Cu}$  phase is formed. According to the obtained lower microhardness of eutectic structure than the other phases, it can be seen although eutectic phases ( $\alpha\text{-Al/Al}_2\text{Cu}$ ) is the thickest layer in the interface, but it contained a large amount of  $\alpha\text{-Al}$  phase besides  $\text{Al}_2\text{Cu}$  that has an appropriate plasticity. This helps layer I to raise resistance of plastic deformation. Since both layer II and III are composed of pure intermetallic compounds, the variation of the thicknesses would significantly affect the interfacial mechanical properties.

From this point of view, it seems that the bond strength is under control of thicknesses of layer II and III. Also it seems, layer I does not have a significant influence on the bond strength.

The Al-Cu binary diagram showed that chemical reaction between Al and Cu can easily produce intermetallic compounds with a nonmetallic covalence bond. For instance, sample A<sup>1</sup>•C<sup>2</sup>• with most number of intermetallic phases has the lowest bond strength in Table 2.

Therefore, phase composition and microstructure of interface, are the main factors that have been affected on the bond strength as well.

#### 4- Conclusions

In this work, the microstructure, the forming mechanism, IMCs microhardness, electrical resistance and the bonding strength of the interface of copper clad aluminum rods produced by static compound casting were studied, respectively. The effects of processing parameters, e.g. aluminum pouring temperature and copper preheating temperature on the above-mentioned cases were analyzed in details.

- 1- Compared to Al-melt pouring temperature, preheating the Cu tubes have much more influence on the formation of intermetallic thickness and types.
- 2- The interface of Al-Cu bimetal produced by compound casting process was composed of three main layers that from Al-Core towards Cu-sheath consists of Layer I where is  $\alpha\text{-Al/Al}_2\text{Cu}$  eutectic structure as the thickest layer, layer II is  $\text{Al}_2\text{Cu}$  and layer III consists of the several pure intermetallic compounds as the thinnest layer such as  $\text{AlCu}$ ,  $\text{Al}_2\text{Cu}_3$ ,  $\text{Al}_3\text{Cu}_2$ ,  $\text{Al}_4\text{Cu}_3$  respectively, while in the solid state welding processes have observed just in layer II and III. The layer I was formed by dissolving and rapid solidification below the temperature of  $541^{\circ}\text{C}$  as the first layer, then the layer II due to the lowest activation energy than the other identified phases was formed on the inner surface of Cu tube by the diffusion, nucleation and growth mechanism as the second layer. Finally the layer III was formed by solid-state phase diffusion transformation as the third layer.

- ۳- From EPMA analysis, it is observed that in diffusion-control layer III, at first, AlCu and Al<sub>2</sub>Cu<sub>5</sub> were formed and then with increasing of preheated Cu temperature and heat content of interface, Al<sub>7</sub>Cu<sub>2</sub> and Al<sub>7</sub>Cu<sub>4</sub> were formed.
- ۴- Vickers microhardness for Al is in the range of ۳۵ to ۴۵, eutectic structure is in the range of ۱۵۰ to ۲۵۰, Al<sub>7</sub>Cu is in the range of ۴۰۰ to ۵۰۰, AlCu is in the range of ۸۰۰-۸۵۰, mixture of AlCu+Al<sub>7</sub>Cu<sub>2</sub> are in the range of ۹۶۰ to ۱۰۰۰, mixture of Al<sub>7</sub>Cu<sub>2</sub>+Al<sub>7</sub>Cu<sub>4</sub> are in the range of ۱۱۰۰ to ۱۲۰۰ and the Cu is in the range of ۷۵ to ۸۵ Kgf/mm<sup>۲</sup>
- ۵- The conductivity was decreased and electrical resistivity was increased by increasing the thickness of interface.
- ۶- It seems, the bond strength is dominated by the thicknesses of layer II and III, due to IMCs higher microhardness values than the layer I.

## References

- ۱- Eizadjou, M., Kazemi Talachi, A., Danesh Manesh, H., Shakur Shahabi, H., Janghorban, K. " Investigation of Structure and Mechanical Properties of Multi-Layered Al/Cu Composite Produced by Accumulative Roll Bonding (ARB) Process", Composites Science and Technology, Vol. ۶۸, pp.۲۰۰۳-۲۰۰۹, ۲۰۰۸.
- ۲- Khosravifard, A., Ebrahimi, R., "Investigation of Parameters Affecting Interface Strength in Al/Cu Clad Bimetal Rod Extrusion Process", Materials and Design, Vol. ۳۱, pp.۴۹۳-۴۹۹, ۲۰۱۰.
- ۳- J. Xie, C. Wu, X. Liu, "A novel forming process of copper cladding aluminum composite materials with core-filling continuous casting" Materials Science Forum, Vol. ۵۳۹-۵۴۳ (۲۰۰۷) pp ۹۵۶-۹۶۱
- ۴- Liu H, Ke F.J., Pan H, Acta Physica Sinica, Vol.۵۶(۱), pp.۴۰۷ [in Chinese], ۲۰۰۷.
- ۵- Owczarski W.A., Paulonis D.F., Welding Journal, Vol.۶۲, pp.۲۲, ۱۹۸۱.
- ۶- Lee, K.S., Kwon, Y.N., "Solid-State Bonding between Al and Cu by Vacuum Hot Pressing", Transactions of Nonferrous Metals Society of China, Vol.۲۳, pp.۳۴۱-۳۴۶, ۲۰۱۳.
- ۷- Hug, E., Bellido, N. , " Brittleness Study of Intermetallic (Cu, Al) Layers in Copper-Clad Aluminum Thin Wires", Materials Science and Engineering A, Vol. ۵۲۸ , pp.۷۱۰۳-۷۱۰۶, ۲۰۱۱.
- ۸- Lee, S., Lee, M.G. , Lee, S.P., Lee, G.A., Kim, Y.B, Lee, J.S, Bae, D.S, " Effect of Bonding Interface on Delamination Behavior of Drawn Cu/Al Bar Clad Material", Transactions of Nonferrous Metals Society of China, Vol.۲۲, pp.۳۶۴۵-۳۶۴۹, ۲۰۱۲.
- ۹- Abbasi M, Karimitaheri A, Salehi M.T. "Growth Rate of Intermetallic Compounds in Al/Cu Bimetal Produced by Cold Roll Welding Process ", Journal of Alloys and Compounds, Vol.۳۱۹, pp.۲۳۳-۲۴۱, ۲۰۰۱.
- ۱۰- Sheng, L.Y., Yang, F., Xi, T.F., Lai, C., Ye., H.Q., "Influence of Heat Treatment on Interface of Cu/Al Bimetal Composite Fabricated by Cold Rolling", Composites, Vol. ۴۲, Part B, pp.۱۴۶۸-۱۴۷۳, ۲۰۱۱.
- ۱۱- Chen, Hwang, "Effect of Annealing on the Interfacial Structure of Aluminum-Copper Joints", Materials Transactions, Vol. ۴۸, No. ۷, pp. ۱۹۳۸ - ۱۹۴۷, ۲۰۰۷.
- ۱۲- Avitzur, B., "Handbook of Metal Forming Processes", Wiley, New York, ۱۹۸۳.



- 13- Kazanowski, P., Epler, M.E., Misiolok, W.Z., "Bimetal Rod Extrusion-Process and Product Optimization", Journal of Materials Science and Engineering A, Vol.379, pp.170-180, 2004.
- 14- Luo, J.T., Zhao, S.J., Zhang, C.X., "Casting-Cold Extrusion of Al/Cu Clad Composite by Copper Tubes with Different Sketch Sections," Journal of Central South University of Technology, Vol.19, pp.882-886, 2012.
- 15- Mamalis, A.G., Szalay, A., Vaxevanidis, N.M., Manolagos, D.E. "Fabrication of Bimetallic Rods by Explosive Cladding and Warm Extrusion", Journal of Materials Processing Technology, Vol.83, pp.48-53, 1998.
- 16- Bereski, S., Stradomeski, Z., "Quality of Bimetallic Al-Cu Joint after Explosive Cladding", Journal of achievement in materials, Vol.22, pp.73-76, 2007.
- 17- Zare, G.R., Divandari, M., Arabi, H., "Investigation on Interface of Al/Cu Couples in Compound Casting", Materials Science and Technology, Vol. 29, pp.190-196, 2013.
- 18- Papis, K.J.M., Hallstedt, B., Loeffler, J.F., Uggowitzner, P.J., "Interface Formation in Aluminium-Aluminium Compound Casting" Acta Materialia, Vol. 56, pp. 3743, 2008.
- 19- ASM metals handbook. "Alloy Phase Diagram", Vol. 3. Ohio, ASM International; 1992
- 20- Neumann, N.F., US Patent, 3421579, 1969.
- 21- Divandari, M., Vahid Golpayegani, A.R., "Study of Al/Cu Rich Phases Formed in Al-Cu Alloy by Inserting Cu Wire in Pattern in LFC Process", Materials and Design, Vol. 30, pp.3279-3285, 2009.
- 22- Moreno, D., Garrett, J., Embury, J.D., "A Technique for Rapid Characterization of Intermetallics and Interfaces", Intermetallics, Vol.7, pp-1001-1009, 1999.
- 23- Liang, H., Xue, Z., Wu, C., Liu, Q., Wu, Y., "Research on Continuous Core-Filling Casting Forming Process of Copper-Clad Aluminum Bimetal Composite Material", Acta Metallurgica Sinica, Vol.23 No.3 pp206-211 June 2010.
- 24- Chen, S., Ke, F., Zhou, M., Bai, Y., " Atomistic Investigation of the Effects of Temperature and Surface Roughness on Diffusion Bonding Between Cu and Al" Acta Materialia, Vol.55, pp.3179-3185, 2007.
- 25- Xu, H., Liu, C., Silberschmidt, V.V., Pramana, S.S., White, T.J., Chen, Z., Acoff, V.L., "Behavior of Aluminum Oxide, Intermetallics and Voids in Cu-Al Wire Bonds", Acta Materialia, Vol.59, pp.5771-5773, 2011.