Iranian Journal of Materials Science & Engineering Vol. 14, No. 4, December 2017

Effect of Post-Deformation Annealing Treatment on Interface Properties and Shear Bond Strength of Al-Cu Bimetallic Rods Produced by Equal Channel Angular Pressing

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Received: July 2017 Accepted: September 2017

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DOI: 10.22068/ijmse.14.4.25

Abstract: *Effects of annealing treatment after equal channel angular pressing (ECAP) on the interface properties and shear bond strength of Al/Cu bimetallic rods were investigated. For the as-deformed samples, the one with two passes of ECAP indicated higher shear bond strength. Formation of a layer of intermetallic compounds after annealing treatment is confirmed. In general, by increasing annealing temperature, thickness of intermetallic compounds at the interface increases. Shear bond strength was initially reduced by annealing at 200, 250 and 300° C and increased at 350° C. With further increase in annealing temperature to 400° C, shear bond strength slightly decreased which is correlated to the increased thickness of the intermetallic compounds.*

Keywords: *Bimetallic Rod, Annealing; Interface; Shear Bond Strength.*

1. INTRODUCTION

In recent years, engineers have been searching for alternatives for copper for electrical applications. A potential alternative must help to maintain a reasonable electrical conductivity and high strength as well as reducing the price and weight of the product. Due to the low weight and price and good electrical conductivity of aluminum, Al/Cu bimetallic composites seem appropriate for this purpose [1]. These composites are used as transition piece in high direct-current bus systems and aerospace structural applications [2-5]. A wide range of welding techniques, e.g., diffusion bonding [6], cold rolling [6], extrusion [1], explosive welding [7] and friction stir welding [8] have been used to produce such composites.

Equal channel angular pressing (ECAP) has been recently proposed by Eivani and Karimi Taheri [9] as a new method for producing cladded rods. In ECAP, sample is inserted into a vertical channel and pressed into a continuing horizontal channel, imposing a large shear strain on the sample. In this process, the cross section of the billet is not subjected to change [10]. According to Zebardast et al. [11], strain caused by the deformation makes two mating surfaces extrude into each other. Therefore, the extruded virgin metals reach to atomic distance and cold weld forms [11].

In order to increase the ductility and relieve the residual stresses developed in Al and Cu after deformation, annealing treatment may be used [4, 11, 12]. However, due to the high activity between copper and aluminum, application of annealing treatment may result in intermetallic formation. Since intermetallic compounds are brittle and have low strength and high electrical resistivity, intermetallic formation decreases the electrical conductivity and the bond strength between Al and Cu [13]. Therefore, the post-deformation annealing must be performed at optimum time and temperature to enhance the bonding strength through the inter-diffusion of two metals and prevent formation of intermetallic compounds at the interface [4, 12, 14]. Eslami et al. [13] combined ECAP with diffusion bonding for producing Al-Cu bimetallic rods. The joint shear strength was found to increase by application of this process [13]. In this combined process, holding time and temperature should be controlled to prevent the formation of intermetallic compounds at the interface.

In the present research, effects of annealing treatment after equal channel angular pressing on the interface properties and shear bond strength of Al-Cu bimetallic rods are investigated. For this purpose, Al-Cu bimetallic rods are produced by one and two passes ECAP at room temperature. Extruded specimens are annealed at 250, 300, 350 and 400°C for one hour. The joint properties between Al and Cu in terms of formation and thickness of intermetallic compounds at the interface are studied. Variations in shear bond strength of the samples are investigated and correlated to intermetallic formation.

2. EXPERIMENTAL PROCEDURE

Commercially pure aluminum rod and copper pipe were used for producing the bimetallic rod. Aluminum samples were machined from a 20 mm thick aluminum sheet. Table 1 shows the initial dimensions of the components used for producing the composite, before which, the copper pipe and aluminum rod were annealed for an hour at 500 and 350°C, respectively.

The diameter of the channel of the ECAP die used in this study was 20 mm. The die intersecting angle was 90° with an outer curved corner angle of 22°. The die was designed using a split configuration to enable the removal of the mandrels and the extruded specimen after the process.

In order to remove surface contamination and oxide layers, mating surfaces of copper sheath and aluminum core were prepared by conventional grinding and wire brushing. Scratch brushing was performed using a steel wire brush with 0.4 mm diameter wires fixed on an electromotor with the rotation speed of 3000 rpm. Afterwards, samples were degreased by acetone and dried with compressed air. The time interval between the surface preparation and ECAP process was kept less than 2 min to avoid the formation of a thick and continuous oxide layer on the mating surfaces [13].

For producing the bimetallic rods, the aluminum rod was inserted into copper cylinder and deformed in ECAP up to two passes using route A. A 220 ton capacity hydraulic press with a ram speed of 2 mm/s was used. Indeed, the ram speed, i.e., the movement speed of the cross head of the press, is kept constant during the process. It should be noted that this may result in different interfacial speed at the interface between aluminum and copper which is not the subject of study in this investigation. In route A, the sample is extruded without rotation between passes. To reduce frictional effects between the samples and die walls, molybdenum disulfide (MoS2) was used as the lubricant during pressing [13]. Subsequently, extruded specimens were annealed at 250, 300, 350 and 400° C for one hour in a furnace. The interfaces of the samples were examined using Roventec Vega Tescan scanning electron microscope (SEM). Image Tools software was used for measuring the thickness of intermetallic compounds formed at the interface after annealing.

In order to measure the shear bond strength between copper sheath and aluminum core, shear bond strength tests were performed on the samples using a die designed according to ASTM F1044-87 standard [11, 13]. In each case, three samples were prepared for the test and average value was reported. The compaction force was imposed to the mandrel by using the compaction mode of SCENK TREBEL tensile strength test machine. With dividing the compaction force by the mating surface of Al/Cu the shear bond strength of the specimens were calculated. Fig. 1 shows the design and image of the die and the sample cut from the bimetallic rod.

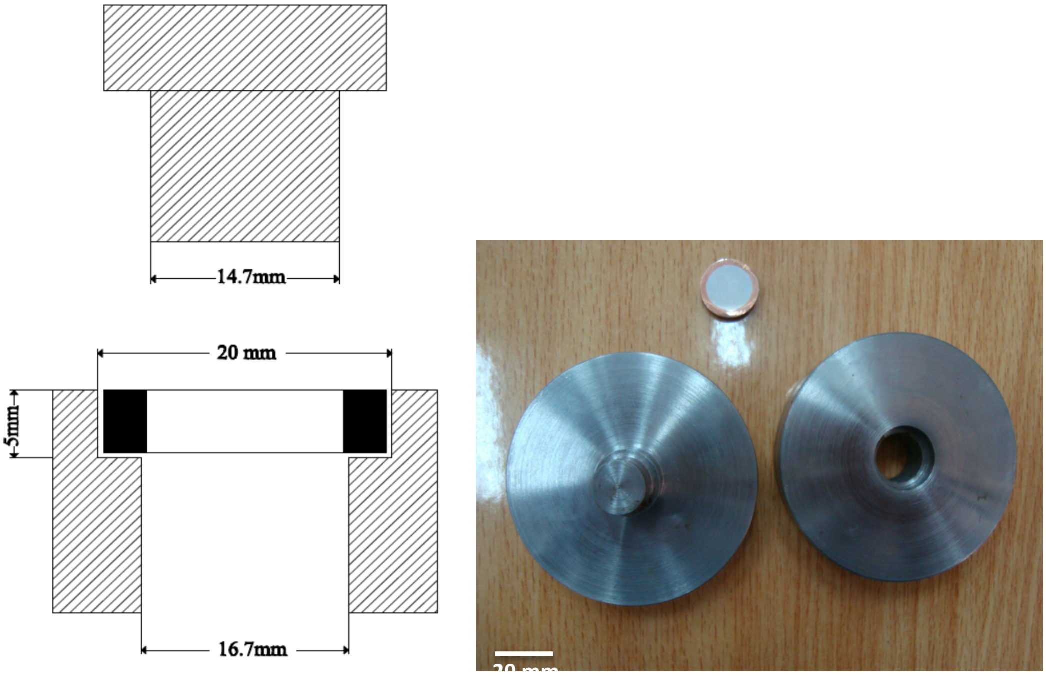
Table 1. Dimensions of the components used for producing the composites.

Material	Outer diameter (mm)	Inner diameter (mm)	Length (mm)
Copper pipe	19.8	15.3	130
Aluminum rod	15.2	-	130

3. RESULT AND DISCUSSION

3. 1. Characterization of the Interface

Fig. 2 shows the SEM images of the joint interface of the samples after one and two passes ECAP. These images are taken from the regions



(a) **(b)**

Fig. 1- a) schematic illustration and **b)** image of the sample and the die used for measuring shear bond strength.

of full contact between the two metals. In fact, investigation of the joint interface indicated that in some regions (around 50 % of the interface), a perfect contact between the two metals occurred. However, in some other regions no contact between the two metals was formed. This is probably due to the non-uniformity of strain distribution in the transverse cross-section of the samples [15]. After the first pass of ECAP, a significant strain variation occurs between upper and lower sections of the sample [15]. The fraction of contacted regions increases from 50 to about 90 % by increasing the number of passes to two. This may be correlated to the higher strain and better homogeneity of strain caused by the deformation in the second pass that forces Al and Cu to squeeze significantly into each other and forms a serrated and wavy interface boundary as shown in Fig. 2 (b).

ECAP is known for applying a high level of strain to deforming materials. This is the case as well for the aluminum core and copper sheath and consequently a high level of residual stresses remains in the product [11]. These stresses can develop a diffusion bonding between the two metals during heat treatment. Fig. 3 shows the interface of the samples deformed for two passes after annealing at 250 and 300°C for 1hr. After annealing at 250° C, no intermediate layer is observed at the interface. In other words, no intermetallic compounds are formed after annealing at this temperature. This is in agreement with the results of Abbasi et. al. [4]. In fact, low temperature prevents the formation of intermetallic compounds at the interface. Low temperature annealing causes the Al/Cu rough interface (Fig. 2b) to become smooth (Fig. 3a). This phenomenon is probably related to the diffusion between Al and Cu [16].

With increasing annealing temperature to 300° C, an intermediate layer forms at the interface [16] (Fig. 3b). EDS analysis results of a point on this layer is shown in Fig. 4. $CuAl_2(\theta)$ is the most likely formed compound as it is the closest composition to the results of EDS analysis [16]. Magnesium content in this phase is attributed to

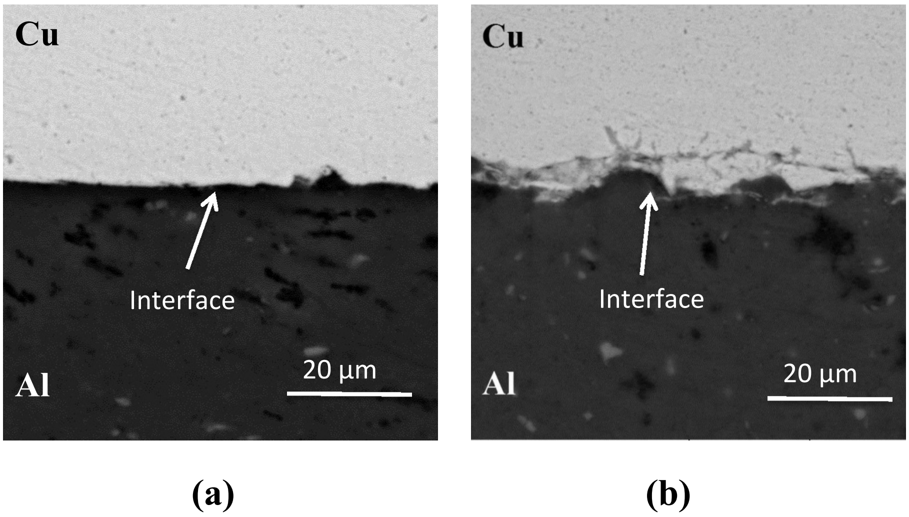


Fig. 2. SEM images of Al/Cu interface in the full contacted regions after (a) one, (b) two passes of ECAP.

magnesium impurity in aluminum that diffuses to the interface during heat treatment. According to the research conducted by Xu et al. [17], $CuAl_2$ is the first phase which forms at 300° C. This may be correlated to the lower melting point of Al in comparison with Cu and its higher diffusion coefficient at a single temperature. Therefore with the diffusion of Cu through Al, $CuAl_2$ initially forms, as it has the lowest effective heat of formation among intermetallic compounds of Al-Cu system [17, 18].

Presence of oxygen in the results of EDS analysis (Fig. 5) indicates the formation of oxides at the interface after annealing. The furnace atmosphere contains oxygen. Therefore, with increasing temperature during the heat treatment, the formation of oxides at the interface, i.e., the regions that they are not in complete contact, would be expected. Since the EDS detector analysis is not reliable for the quantitative analysis of light elements, the exact amount of oxygen at the interface cannot be obtained using

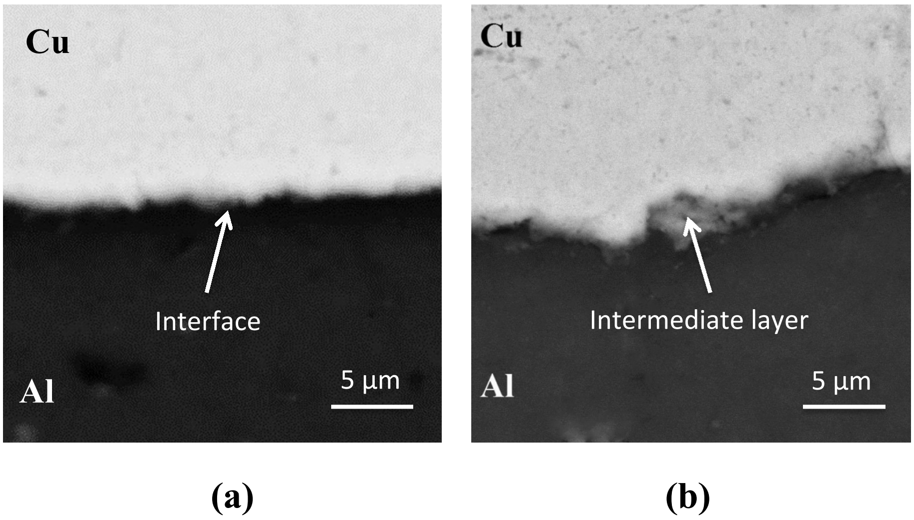
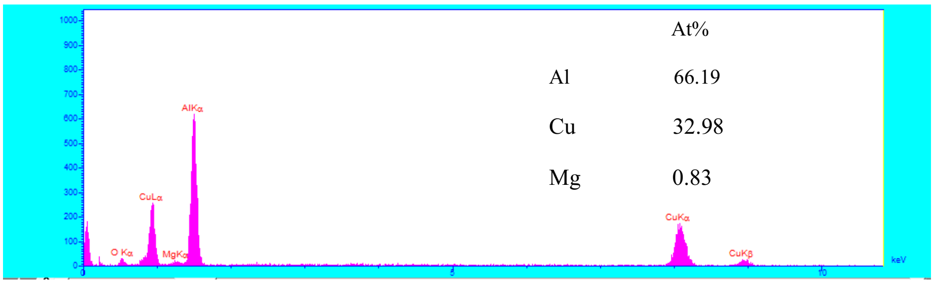


Fig. 3. Interface development after annealing at (a) 250 and (b) 300° C for 1h.



	At%
Al	66.19
Cu	32.98
Mg	0.83

Fig. 4. Results of EDS analysis of the intermediate layer formed after annealing at 300°C for 1h.

this method.

The interface of the sample deformed for two passes after annealing at 350°C for 1 h, is shown in Fig. 5. Because of the short duration of annealing, the compounds formed at the interface are thin. Moreover, due to small color differences of these intermetallic compounds, it is hard to distinguish them from one another. This is related to their little difference in average atomic number of the constituting elements of the compounds. Table 2 presents the chemical composition of the region shown by point 1 (near copper) and point 2 (near aluminum) in Fig. 5. $Cu_3Al(\xi_2)$ and $CuAl_2(\theta)$ are the closest intermetallics in chemical composition to the results of EDS analysis of points 1 and 2, respectively. Thus, as the annealing temperature increases, Cu-rich (Cu_3Al) and Al-rich compounds ($CuAl_2$) form near aluminum and near copper, respectively.

Solubility of copper in aluminum is much lower than the solubility of aluminum in copper. Therefore, it is expected that Al(Cu) solid solution saturates more rapidly which results in the formation of aluminum-rich phases such as $CuAl_2$[19]. In addition, the formation of intermetallic compounds is attributed to their formation energy. Based on the formation energy of intermetallic compounds $CuAl_2$ is the first phase that forms due to it's low formation energy [20, 21]. With further increase in temperature, the diffusion of Copper increases and intermetallic compounds with more copper content (Cu_9Al_4, CuAl, Cu_4Al_3 and Cu_3Al) are formed at the interface [2, 20, 21].

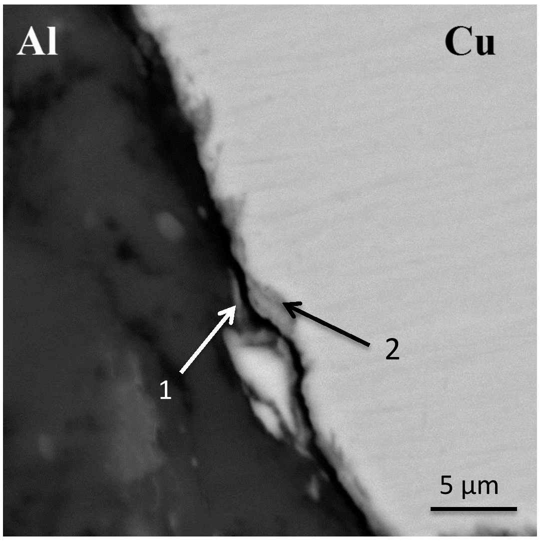


Fig. 5. Interface developed between Al and Cu after annealing at 350°C for 1h.

Table 2. Compositions of two different regions (points 1 and 2 in Fig.5) along interface (at.%).

Test position	Cu	Al	Mg	Most likely intermetallic compound
1	31.65	67.55	0.79	$CuAl_2$
2	70.51	28.46	1.03	Cu_3Al

3. 2. Effect of Annealing Temperature on the Formation of Intermetallic Compounds

In Fig. 6, the variation of the average thickness of the intermetallic layers at the interface with increasing the annealing temperature is shown. Intermetallic layers formed at the interface have low thickness and high standard deviation representing the non-uniform thickness of these layers. According to Fig. 6, no intermetallics form at the interface after annealing at 250°C. Increasing annealing temperature causes nucleation and growth of the intermetallic compounds at the interface. With further increase in temperature, the thickness of the layer formed at the interface, increases [20]. According to Lee et al. [22], by increasing annealing temperature, the thickness of intermetallic layer increases linearly which is in line with the results of current research. According to Abbasi et al. [4], increasing the thickness of intermetallic compounds at the interface increases the electrical resistivity of the bimetal. Therefore, formation of these compounds is not desirable.

3.3. Shear Bond Strength

Average shear bond strength after the first pass was 9.55 MPa. After the second pass of ECAP, shear bond strength increased to 23.57 MPa. Low bond strength indicates a weak joint between aluminum and copper after one pass ECAP. Due to the higher strain in the second pass of deformation, a noticeable increase occurs in bond strength after the second pass of ECAP. Increase in shear strength reveals an improvement in the quality of the joint after the second pass of deformation. In other words, after the second pass a good weld has been formed in about 90 % of the interface which causes significant increase in shear strength.

Fig. 7 shows the variations in shear strength in samples deformed for one and two passes and annealed at different temperatures. According to Fig. 7 (a), a notable reduction occurred in shear bond strength of the sample deformed for one pass by annealing at 250°C (p1, 250) and bond strength decreased to 4.9 MPa. As discussed earlier, no joint has been formed between aluminum and copper in about half of the interface after the first pass of ECAP. This means that at these areas oxide layers form at the mating surfaces after annealing at 250°C. An example of these oxide layers is shown in Fig. 8. Besides, according to Zebardast [11], annealing unlocks

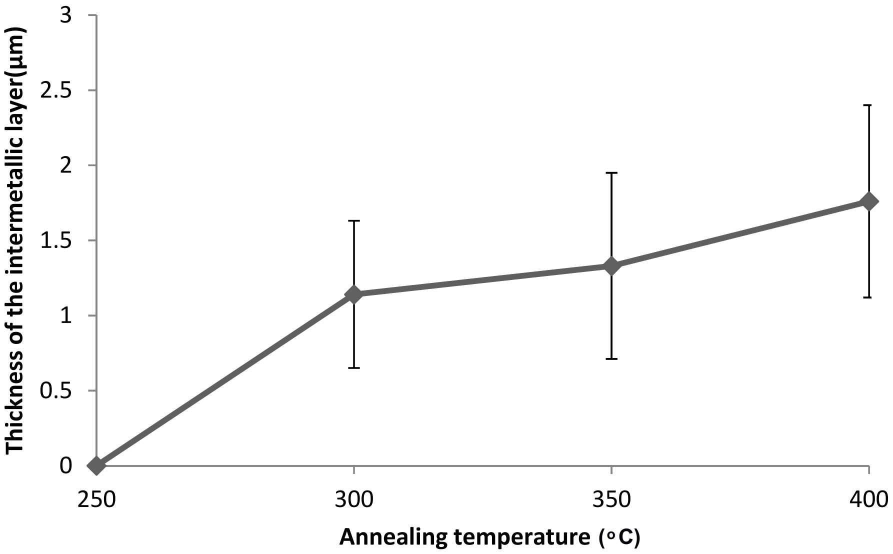
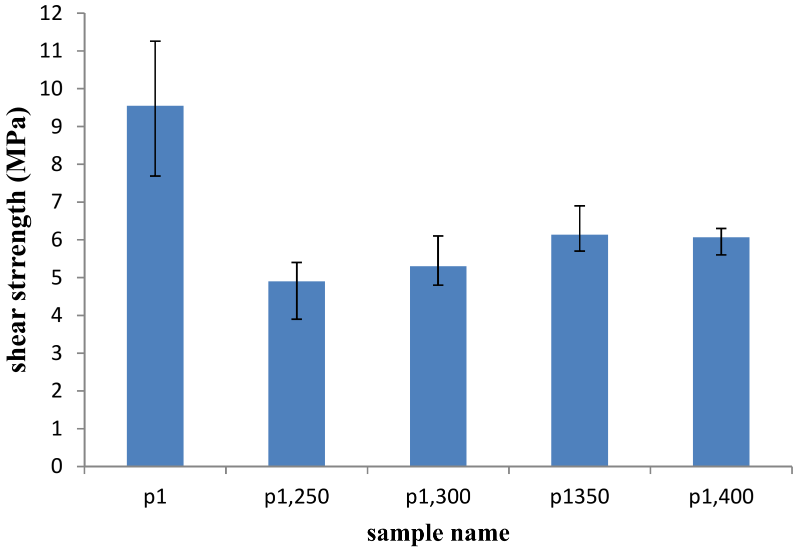


Fig. 6. Variation of the average thickness of the intermetallic layers with increasing annealing temperature. Samples are annealed for 1hour.



(a)

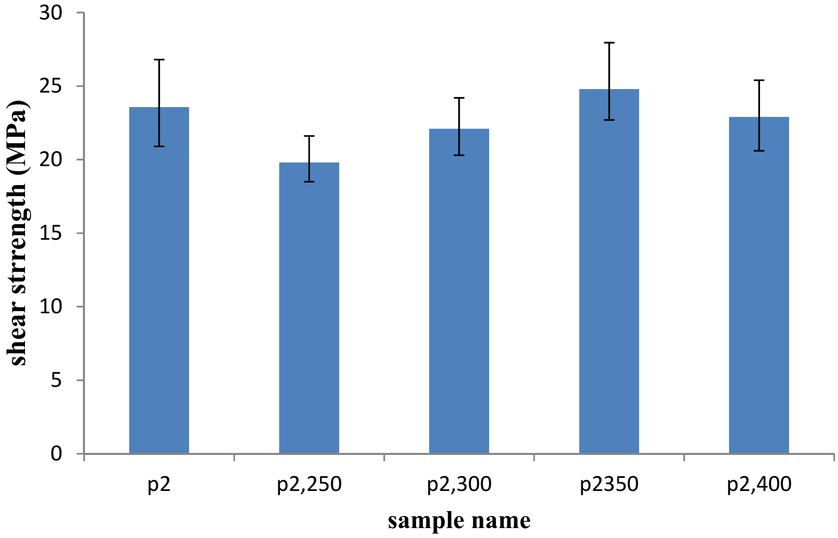


Fig. 7. Dependence of the joint shear strength of the samples on annealing temperature. The samples are deformed for (a) one and (b) two passes of pass.

the mechanical locks developed at the interface after the first pass of ECAP. Therefore, the shear strength of the annealed specimen can be considered as the metallurgical share of the strength between the copper sheath and aluminum core [11]. Consequently, elimination of mechanical locks and formation of oxide layers after annealing lead to notable reduction in shear strength of the specimens deformed for one pass. Slight increase in shear strength in higher annealing temperatures may be correlated to the increase in diffusion rate and occurrence of

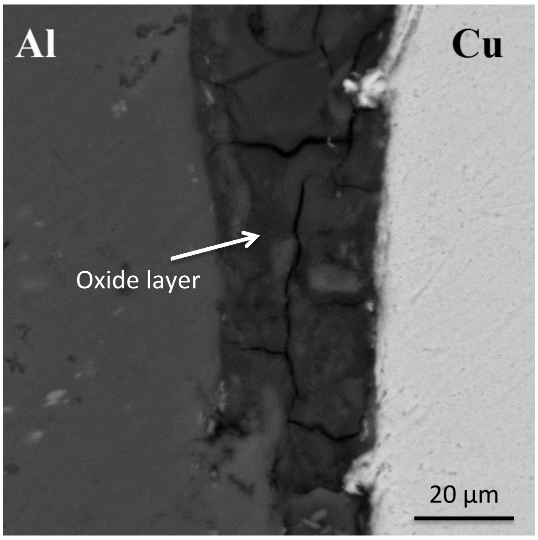


Fig. 8. Oxide layer formed at the interface of the sample deformed for one pass after annealing at 250 °C.

diffusion bonding.

As shown in Fig. 7 (b), the shear bond strength of the specimen deformed for two passes (p2) reduced to 19.8 MPa after annealing at 250° C (p1, 250) for one hour. This reduction is most likely due to the elimination of mechanical locks after annealing. Due to the improvement in joining of aluminum and copper after the second pass of ECAP, reduction in the shear strength of the specimen deformed for two passes (p2) after annealing at 250 °C (p1,250), is significantly less than the sample deformed for one pass. Thus, after the second pass metallurgical bonds are more extensively formed with respect to that after the first pass. On the other hand, after the second pass of ECAP, a good joint forms between aluminum and copper and the residual stresses develop at the interface due to the severe plastic deformation. Therefore, diffusion bonding occurs between the two metals during heat treatment. With increasing annealing temperature to 350 °C, shear strength reaches 24.8 MPa. This is correlated to the increase in the diffusion rate of copper and aluminum with increasing annealing temperature.

By increasing annealing temperature to 400 °C, shear bond strength decreases slightly. The interdiffusion of Al and Cu leads to the formation of intermetallic compounds at the interface. According to Fig. 6, the thickness of the intermetallic compounds at the interface increases with increasing annealing temperature. Based on a study conducted by Abbasi et al. [11], increasing the width of intermetallic layer more than a critical value causes a reduction in bond strength [11]. Slight decrease in bond strength after annealing at 400 °C may be attributed to the increased amount of intermetallic compounds at the interface. Moreover, formation of oxide compounds with increasing annealing temperature increases [13]. Thickening of the oxide and intermetallic phases weakens the metallurgical bonding between aluminum and copper and reduces shear bond strength. Since such phases are brittle and have covalence bond [4]. In other words, the formation of intermetallic compounds at the interface is destructive to shear bond strength and electrical conductivity of the bimetal [4].

4. CONCLUSIONS

In this study, influences of annealing treatment after equal channel angular pressing on interface properties and shear bond strength of Al-Cu bimetallic rods was investigated. Al-Cu bimetallic rods were fabricated by one and two passes ECAP and annealed at different temperatures. According to the results of this investigation, the following conclusions are made:

1. After two passes ECAP, shear bond strength between aluminum and copper significantly increased from 9.55 to 23.57 MPa.
2. By annealing the samples after the second pass, non-uniform intermetallic layer formed at the interface and Al-rich and Cu-rich intermetallic layers formed near Al and Cu interface, respectively. Thicknesses of the layers increased linearly with increasing annealing temperature.
3. By annealing the specimens after the first pass of ECAP, shear bond strength significantly decreased. With annealing the sample produced by two passes ECAP,

diffusion bonding occurs between aluminum and copper. Consequently, shear strength increased to 24.8 MPa after annealing at 350° C for an hour. With further increase in annealing temperature, shear bond strength decreased.

REFERENCES

1. Khosravifard, A. and Ebrahimi, R., “Investigation of Parameters Affecting Interface Strength in Al/Cu Clad Bimetal Rod Extrusion Process”. Mater. & Des., 2010, 31, 493-499.
2. Eslami, P., Taheri, A. K. and Zebardast, M., “A Comparison Between Cold-Welded and Diffusion-Bonded Al/Cu Bimetallic Rods Produced by ECAE Process”. J. Mater. Eng. Perf., 2013, 22, 3014-3023.
3. Guo, Y., Qiao, G., Jian, W. and Zhi, X., “Microstructure and Tensile Behavior of Cu–Al multi-Layered Composites Prepared by Plasma Activated Sintering”. Mater. Sci. Eng. A., 2010, 527, 5234-5240.
4. Abbasi, M., Karimi Taheri, A. and Salehi, M., “Growth Rate of Intermetallic Compounds in Al/Cu Bimetal Produced by Cold Roll Welding Process. J. Alloys Compd., 2001, 319, 233-241.
5. Sapanathan, T., Khoddam, S. and Zahiri, S. H., “Spiral Extrusion of Aluminum/Copper Composite for Future Manufacturing of Hybrid Rods: A study of Bond Strength and Interfacial Characteristics”. J. Alloys Compd., 2013, 571, 85-92.
6. Chen, S., Ke, F., Zhou, M. and Bai, Y., “Atomistic Investigation of the Effects of Temperature and Surface Roughness on Diffusion Bonding between Cu and Al”. Acta Mater., 2007, 55, 3169-3175.
7. Acarer, M., “Electrical, Corrosion, and Mechanical Properties of Aluminum-Copper Joints Produced by Explosive Welding”. J. Mater. Eng. Perf., 2012, 21, 2375-2379.
8. Abdollah-Zadeh, A., Saeid, T. and Sazgari, B., “Microstructural and Mechanical Properties of Friction Stir Welded Aluminum/Copper Lap Joints”. J. Alloys Compd., 2008, 460, 535-538.
9. Eivani, A. and Taheri, A. K., “A New Method for Producing Bimetallic Rods”. Mater. Let., 2007, 61, 4110-4113.
10. Tolaminejad, B., Karimi Taheri, A., Shahmiri, M. and Arabi, H., “Development of Crystallographic Texture and Grain Refinement in the Aluminum Layer of Cu-Al-Cu Tri-Layer Composite Deformed by Equal Channel Angular Extrusion”. Int. J. Mod. Phys., 2012, 5, 325-334.
11. Zebardast, M. and Karimi Taheri, A., “The Cold Welding of Copper to Aluminum using Equal Channel Angular Extrusion (ECAE) Process. J. Mater. Proc. Technol., 2011, 211, 1034-1043.
12. Shabani, A., Toroghinejad, M. R., Shafyei, A., “Investigating the formation of intermetallic compounds and the variation of bond strength between Al-Cu layers after annealing in presence of nickel between layers”, Iran. J. Mater. Sci. Eng., 2016, 13, 35-43.
13. Eslami, P., Karimi Taheri, A., “An Investigation on Diffusion Bonding of Aluminum to Copper using Equal Channel Angular Extrusion Process”. Mater. Let., 2011, 65, 1862-1864.
14. Lee, J., Bae, D., Chung, W., Kim, K., Lee, J. and Cho, Y., “Effects of Annealing on the Mechanical and Interface Properties of Stainless Steel/Aluminum/Copper Clad-Metal Sheets”. J. Mater. Proc. Technol., 2007, 187, 546-549.
15. Kim, W., Namgung, J. and Kim, J., “Analysis of Strain Uniformity during Multi-Pressing in Equal Channel Angular Extrusion. Scr. Mater., 2005, 53, 293-298.
16. Sheng, L., Yang, F., Xi, T., Lai, C. and Ye, H., “Influence of Heat Treatment on Interface of Cu/Al Bimetal Composite Fabricated by Cold Rolling”. Comp. Part B: Eng., 2011, 42, 1468-1473.
17. Xu, H., Liu, C., Silberschmidt, V., Pramana, S., White, T., Chen, Z. and Acoff, V., “Behavior of Aluminum Oxide, Intermetallics and Voids in Cu–Al Wire Bonds”. Acta Mater., 2011, 59, 5661-5673.
18. Chen, C. Y. and Hwang, W. S., “Effect of Annealing on the Interfacial Structure of Aluminum-Copper joints”. Mater. Trans., 2007, 48, 1938-1947.
19. Guo, Y., Liu, G., Jin, H., Shi, Z. and Qiao, G., “Intermetallic Phase Formation in Diffusion-Bonded Cu/Al Laminates”. J. Mater. Sci., 2011, 46, 2467-2473.

20. Chen, C. Y., Chen, H. L. and Hwang, W. S., "Influence of Interfacial Structure Development on the Fracture Mechanism and Bond Strength of Aluminum/Copper Bimetal Plate". Mater. Trans., 2006, 47, 1232-1239.
21. Jiang, H., Dai, J., Tong, H., Ding, B., Song, Q., Hu, Z., "Interfacial Reactions on Annealing Cu/Al Multilayer Thin Films", J. Applied Phys., 1993, 74, 6165-6169.
22. Lee, W. B., Bang, K. S. and Jung, S. B., "Effects of Intermetallic Compound on the Electrical and Mechanical Properties of Friction Welded Cu/Al Bimetallic Joints during Annealing". J. Alloys Compd., 2005, 390, 212-219.