

INVESTIGATING THE FORMATION OF INTERMETALLIC COMPOUNDS AND THE VARIATION OF BOND STRENGTH BETWEEN Al-Cu LAYERS AFTER ANNEALING IN PRESENCE OF NICKEL BETWEEN LAYERS

A. Shabani*, M. R. Toroghinejad and A. Shafyei

* ali.shabani@ma.iut.ac.ir

Received: September 2015

Accepted: April 2016

Department of Materials Engineering, Isfahan University of Technology, Isfahan, Iran.

Abstract: In the present study, the effect of post-rolling annealing heat treatment on the formation of intermetallic compounds between Al-Cu strips, in the presence of nickel coating on the Cu strips, was investigated. In addition, the effect of post-rolling annealing and intermetallic compounds on the bond strength of Al-Cu strips was evaluated. In order to prepare samples, Cu strips were coated with nickel by electroplating process. After surface preparing, Cu strips were placed between two Al strips and roll bonded. This method is used for producing Al-Ni-Cu composites. Then the samples were annealed at 773K for 2 h. The formation of intermetallic compounds was studied using energy dispersive spectroscopy (EDS) and X-ray diffraction (XRD). Also, in order to investigate bond strength of Al-Cu after post-rolling annealing heat treatment, samples were produced using nickel powder and nickel coating. Then bond strength of strips was investigated using peeling test. The results revealed that by post-rolling annealing of layers, the bond strength between Al-Cu strips decreases dramatically.

Keywords: Intermetallic compounds, XRD, Bond strength, CRB process

1. INTRODUCTION

Metal matrix composites (MMCs) have played a significant role in the materials used in industry for the last several decades. Al-based metal matrix composites, due to their specific properties such as low density, good electrical and thermal conductivity, wear resistance and good formability, are commonly used in aerospace, automotive and structural industries [1-3]. Although different methods are used to manufacture these composites, cold roll bonding (CRB) process has exhibited rapid growth and development in recent years [4-6].

In addition, Al intermetallic compounds are being studied as a structural material for special applications which demand light weight, high thermal stability, corrosion resistance, and good mechanical properties at high temperature [7-10]. However, the intrinsic low ductility and low fracture toughness at room temperature confine their application in engineering designs. A main method to compensate the brittleness of intermetallic compounds includes the embedding of the intermetallic phases in the ductile matrix materials in the form of particles or laminates [8].

Hereon, when a crack propagates inside the intermetallic phase and reaches the ductile metallic phase, owing to plastic deformation at the crack tip, the crack is blunted. Thus, such useful properties can practically be utilized in a structural material and the ductility of matrix material could avoid its brittle deficiency.

Cold roll bonding is a type of pressure welding or solid state welding process used to join similar and/or dissimilar metals, where bonding is established by joint plastic deformation of the metals to be bonded. CRB is a simple method of bonding and can be easily automated. This process is commonly used to produce metal composite sheets from different metals by rolling at room temperature [5].

Recently, the ARB process and subsequent annealing reactions have been used as a new production method of metal intermetallic composites (MICs) [11, 12]. In this technique, complex heat and chemical treatments, as heat treatment at controlled environment, are not required [13]. Also, simplicity, no dimensional limitations, and cheaper primary commodity are the other major advantages of the ARB process for the production of MICs. One of the most

important parameters in ARB process is bonding between layers. So in producing MICs, it seems to be needed to investigate bonding strength between layers after annealing heat treatments.

Lots of works have been performed to produce different composites using accumulative roll bonding process, such as, Al/Ti [14], Al/Cu [8], Al/Mg [15], Ti/Al/Nb [16], Cu/Zr [17] and Al/Ni [18, 19]. Additionally, some studies have been done on particle reinforced metal matrix composites, including Al/Al₂O₃ [20], Cu/Al₂O₃ [21], and Al/SiC [22]. Recently, Jamaati et al. produced a metal matrix composite (aluminum/alumina) using a coating method (anodizing) and accumulative roll bonding process. The most important advantage of coating is the producing high-strength and highly uniform composites with ultra fine grain structure [23]. In the first step of their work, the two annealed strips were prepared in terms of surface, and then the anodized strip was laid between the prepared surfaces of strips. The strips were stacked over each other, fastened at both ends, and roll-bonded. In the second step, in order to improve mechanical properties and have a uniform distribution of reinforcement particles, accumulative roll bonding was performed. It became obvious that bonding between layers in the presence of coating, plays an important role in final properties of composite.

In this paper, the formation of Al-Cu and Al-Ni intermetallic compounds between Al-Cu layers in the presence of nickel coating on Cu sheets after cold roll-bonding process was investigated. Moreover, the effects of post-rolling annealing treatment and the formation of intermetallic compounds on the bond strength of Al-Cu layers were evaluated.

2. EXPERIMENTAL PROCEDURE

2. 1. Materials

The materials included Al and Cu commercially pure strips with the specifications shown in table 1. The Strips 100 mm long, 25 mm wide, and 1 mm thick were cut from cold rolled sheets parallel to the original rolling direction. Al strips were annealed at 663 K for 2

h and Cu strips were annealed at 773 K for 2 h to ensure consistent specimen hardness. Additionally, Cu strips were coated by nickel using electroplating method. Nickel powder with the average size of less than 40 μ m also was manually dispersed between the two strips before the CRB process.

2. 2. Surface Preparation

To produce a satisfactory metallurgical bond by warm and cold roll bonding, it is essential to remove the contamination layers on the surfaces of two metals to be joined [24, 25]. In practice, metal surfaces are covered with oxide films and other surface contaminants, which inhibit bonding, at least at room temperature. The surface preparation processes, used here included the following:

1. Degreasing Al and Cu surfaces in acetone bath for at least 30 seconds.
2. Scratch-brushing the intimate surface of strips by means of a circular steel brush with 50 mm radius and 0.25 mm wire diameter. The surfaces were scratch-brushed at peripheral speed of 2000 rpm at least for 60 seconds parallel to the rolling direction.

However, degreasing in acetone bath for 30 seconds was adequate alone for the strips with nickel coating. Furthermore, the strips were rolled quickly to prevent significant re-oxidation of the prepared surfaces.

2. 3. Cold Roll Bonding (CRB) Process

After surface preparation, to avoid contaminating the surfaces, they are needed to be handled immediately and without touching. The time interval between surface preparation and rolling must be kept less than 180 s to avoid oxidation or any new contamination on the surfaces [5, 26]. Also, sheets must be fixed by steel wire at both ends. Then, three layers, consisting of two Al layers and one Cu layer, were cold roll bonded. Schematic illustration of cold roll bonding is shown in Fig. 1. Cu sheets were coated by nickel at a certain electroplating

time (120 minutes) or the same amount of nickel powder was added between layers, and then using different rolling reductions of 20 to 80% sample were made. Also, to investigate the effect of post-rolling annealing treatment, the sheets were annealed at 773 K for 2 h after adding nickel and CRB process. During bonding, a high reduction in the thickness of the materials occurs under a high pressure of the roller. This high reduction generates a great amount of heat and creates many cracks in the hard layer of the surfaces, so that virgin surfaces of the materials extrude from these cracks and cause the layers to be bonded. CRB process was done by a laboratory rolling mill, with a loading capacity of

20 tons. The roll diameter was 125mm and the rolling speed was set at 4 m/min.

2. 4. Peeling Test

The bond strength of the joined Al/Cu strips was measured using a peel test according to the ASTM-D1876-01 as shown in Fig. 2. In this test, the breaking-off forces were measured as shown in Fig. 3, and the average peel strengths were calculated according to the following equation (Ref 20): Average Peel Strength = Average Load (N) / Bond Width (mm).

Peel tests were performed using an instron tensile testing machine with a 50 kg load cell. The test was carried out using a crosshead speed of 20 mm/min.

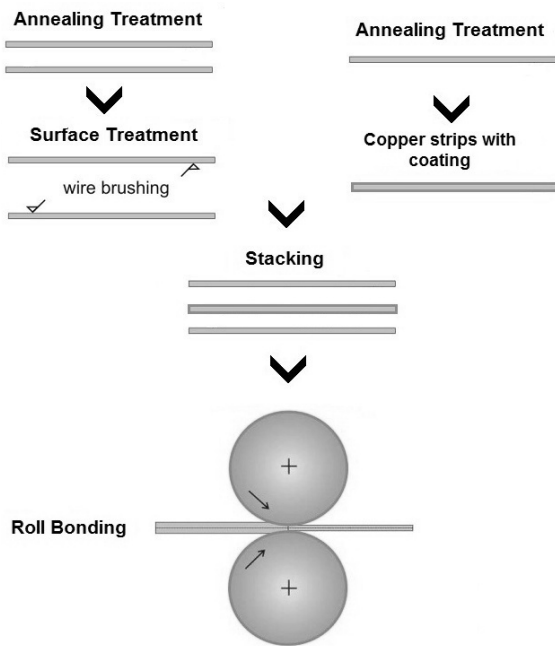


Fig. 1. Schematic illustration of CRB process for 3 layers consisting of 2 Al layers and 1 Cu layer with coating.

Table 1. Chemical composition of used materials

Al (%)	Si (ppm)	Fe (ppm)	Zn (ppm)	Ti(ppm)
99.713	730	1640	160	150
Cu (%)	Si (ppm)	Fe (ppm)	Zn(ppm)	Sn(ppm)
99.86	30	600	20	170

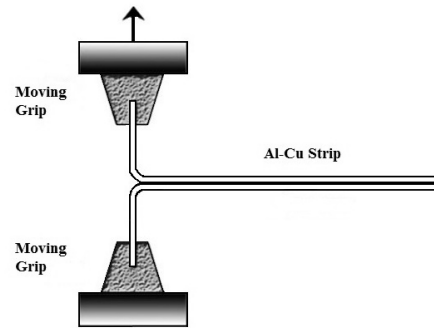


Fig. 2. Schematic illustration of peeling test fixture.

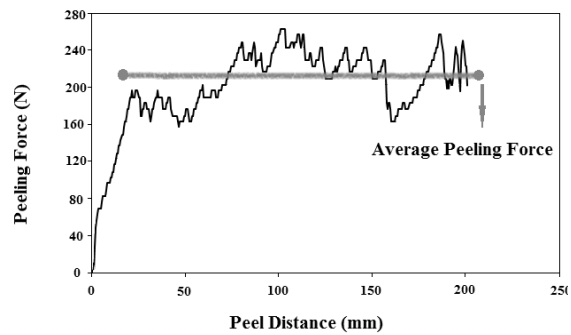


Fig. 3. Typical plot of the peeling force versus the peel distance.

Finally, in order to investigate the formation of intermetallic compounds and study their composition in the interface of Cu layers after post-rolling annealing heat treatment, scanning electron microscopy (SEM) equipped with energy dispersive spectroscopy (EDS) and X-ray diffraction (XRD) were used.

3. RESULTS AND DISCUSSION

Fig. 4 (a) and (b) demonstrate the fracture surface of Cu layer after coating, rolling, post-rolling annealing, and peeling test for two different magnitudes. The surface of a crack is shown in Fig. 4 (b). To evaluate the elements, diffusion of them and probable compounds on the surface around the crack, and also to scrutinize the intensity and atomic percentage of the elements on the surface, data were collected for four regions in the interface of Cu strips after rolling and post-heating, as shown in Fig. 4 (b). The results of the SEM/EDS analyses, corresponding to the regions in this figure with possible phases, are shown in Table 2.

This analysis showed that, for region 1, nickel was the major element with 86.88% of atomic percentage, and then Al had about 6% of atomic percentage. So, the formation of Al-Ni intermetallic compounds, in region 1, is possible. In addition, in the presence of 3.1% of Cu, the formation of intermetallic compounds between Al and Cu is possible.

In regions 2, 3 and 4, Cu was the major element and Al also had 8.6%, 33.39% and 22.84% of atomic percentage, respectively. So, in these regions the elements diffused during heat treatment and it revealed the possibility of the formation of Cu-Al intermetallic compound. Moreover, a low amount of nickel was seen in regions 3 and 4 making the formation of Al-Ni intermetallic compounds more possible.

Elemental analysis only showed the diffusion and the probability of the formation of intermetallic compounds between layers. In addition, due to formation of solid solution between Cu and Ni the results of EDS could not proof the formation of intermetallic compound between layers. Therefore, to investigate the presence of intermetallic on the surface, after

post-rolling annealing, X-ray diffraction was used. The typical X-ray diffraction pattern of 1×1 cm² of the samples surfaces produced by adding nickel coating and nickel powder are shown in Fig. 5. Different compounds and elements are shown on the related peaks. Fig. 5 reveals that intermetallic compounds between Al-Cu and Al-Ni have been formed during heat treatment. Additionally, Cu and Ni elements were detected on the surface but no Al was detected as an element, which means Al on the surface only exists in the compounds, presenting another reason for the formation of intermetallic between layers.

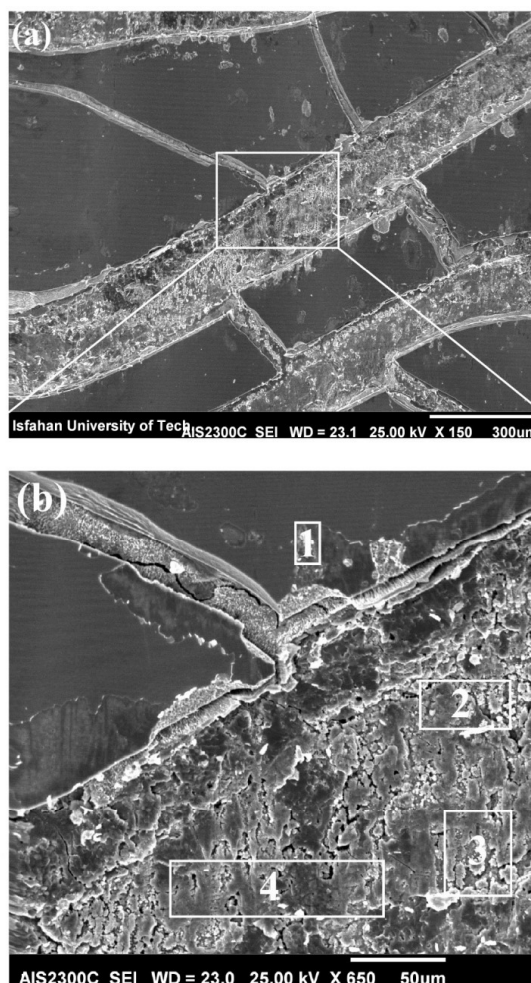


Fig. 4. (a) and (b), SEM image of a crack on the fracture surface of Cu strips after the peeling test. The images shown are for the Cu strip with 30 μ m nickel coating, after 50% reduction and post-rolling annealing for 2 h at 773K.

Table 2. SEM/EDS results corresponding to the regions in Fig. 4 (b) where takeoff angle is 35.0 and voltage is 20 kV

Points	Element	Line	Intensity (c/s)	Atomic (%)	Probable phases
1	Al	Ka	1.57	5.93	Ni, AlNi, CuAl
	Cu	Ka	1.28	3.12	
	Ni	Ka	41.21	86.88	
	O	Ka	0.45	4.07	
2	Al	Ka	0.02	8.06	Cu, CuAl
	Cu	Ka	3.67	91.94	
	Ni	Ka			
	O	Ka			
3	Al	Ka	11.34	33.39	Cu, CuAl, AlNi
	Cu	Ka	31.82	65.15	
	Ni	Ka	0.90	1.46	
	O	Ka			
4	Al	Ka	5.42	22.84	Cu, CuAl, AlNi
	Cu	Ka	22.83	65.04	
	Ni	Ka	1.21	2.72	
	O	Ka	0.83	9.40	

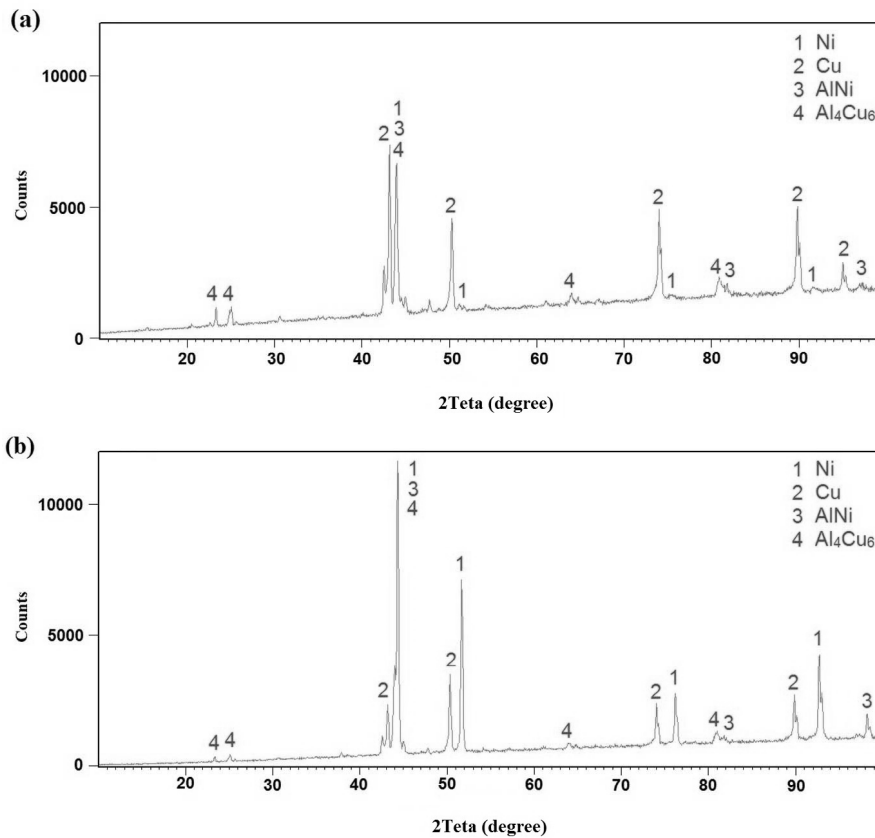


Fig. 5. Typical X-ray diffraction patterns of 1×1 cm² of the Cu fractured surface for 50% reduction and 2 hours annealing at 773 K in the presence of (a) nickel powder and (b) nickel coating.

According to the above explanations, annealing heat treatment of cold rolled Al-Cu strips with nickel coating caused the formation of Al_4Cu_6 and $AlNi$ intermetallic compounds between layers.

The effect of post-rolling annealing treatment, after CRB process, was studied by annealing of the strips at 773 K for 2 h. Figs. 6a and b display the effect of post-rolling annealing treatment of Al-Cu strips in the presence of nickel, either nickel powder or nickel coating respectively. As can be seen from Fig. 6a, annealing treatment causes 33% and 44% decrease in the bond

strength of samples with 50% and 60% reduction in thickness in nickel powdered specimens. Regarding the nickel coated ones, 62% and 60% decrease in the bond strength was observed (see Fig. 6b). To date, the post-rolling annealing treatment has been carried out for various metals [27-29]. Most researches had reported that post-rolling annealing can improve bond strength by reducing hardness of layers, improving atomic diffusion processes, and reducing residual stress between layers [27, 28]. While some other works have reported such tendency, it is also reported that, the growth of intermetallic compounds at

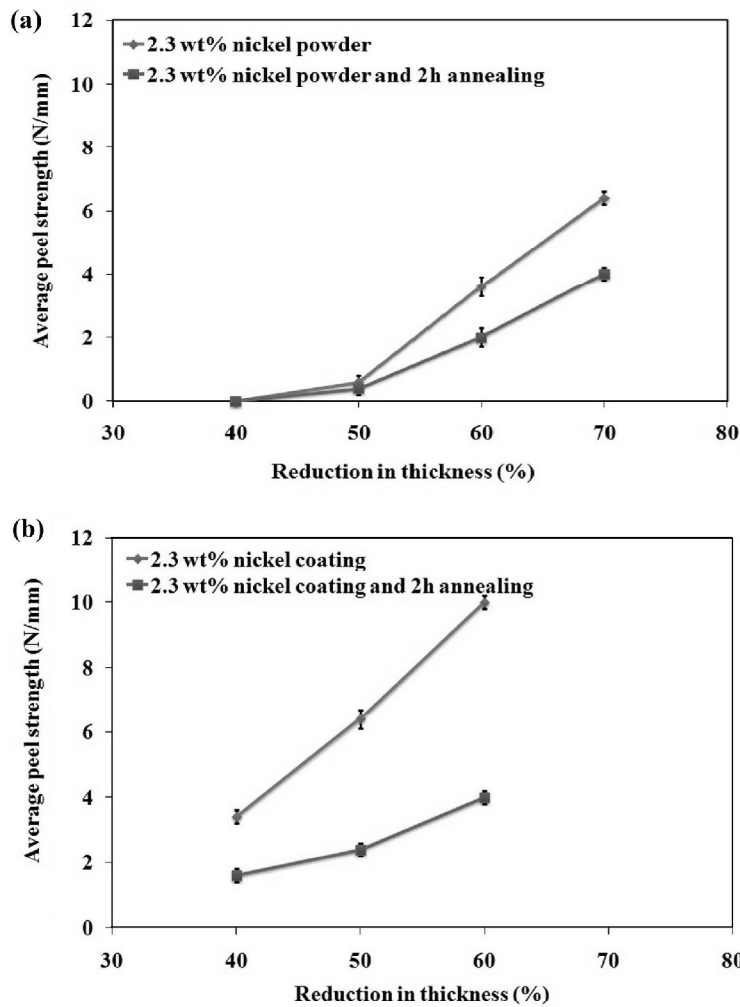


Fig. 6. Variation of average peel strength of Al-Cu strips versus reduction in thickness before and after 2h annealing at 773 K for samples (a) in the presence of 2.3 wt% nickel powder between layers and (b) in the presence of 2.3 wt% nickel coating between layers.

the interface of cold roll-bonded layers after annealing treatment reduces the bond strength of the strips [29-33]. Intermetallics have a complex crystal structure and are brittle due to their partial ionic-covalent bonding [34]. In MMCs, in presence of intermetallic compounds, the cracks initiate at these inclusions. This is followed by a region of stable crack propagation and then, by fast-fracture. Besides, the defects act as stress concentrators and increase the local stress intensity in the material, promoting easy crack nucleation [34]. So easy crack nucleation followed by fast-fracture in the presence of intermetallic compounds reduces the bond strength of the layers [26].

It can be said that the difference in the reduction of bond strength in the two types of samples discussed in this paper is most likely to be related to the different amount of intermetallics formed during annealing treatment. XRD analysis displayed in Fig. 5 confirms that annealing treatment caused formation of AlNi and Al₄Cu₆ intermetallic compounds between layers.

In addition to that, these figures reveal that intermetallic compounds can be detected better in nickel-coated samples rather than in powdered ones. Furthermore, it is obvious that in the presence of nickel powder, Cu is detected more clearly and nickel powder is not as apparently detected as nickel coating. This can be explained by the more uniform distribution of a nickel coating compared to a nickel powder. Nickel coating covers the surface completely while, nickel powder may be agglomerated in different regions of surfaces. As a side effect of this, the formation of intermetallics during annealing treatment is more likely, in the presence of nickel coating. On top of that, because of the fewer amounts of bonding area between Al and Cu in nickel powdered specimens, formation of intermetallics between Al and Cu is more unlikely, in good consistency with the statement cited above.

Briefly, it seems that annealing heat treatment of Al-Cu strips, in the presence of nickel coating and nickel powder between layers at 773 K for 2 hours, caused the formation of intermetallic compounds between layers and because

intermetallics are brittle compounds, and they are stress concentration regions, so the nucleation and propagation of cracks near this compound during peeling test is fast and so quality of bonds between layers decrease. Consequently, the presence of intermetallics between Al-Cu layers significantly decreases bond strength

To sum up when strips are coated with nickel, more intermetallic compounds forms between layers, causing more reduction in the bond strength of Al-Cu strips.

4. CONCLUSIONS

The formation of intermetallic compounds was investigated between Al-Cu layers, in the presence of nickel coating and nickel powder between Al-Cu sheets, after CRB process, and annealing heat treatment. Also, the effect of post-rolling annealing and the formation of intermetallics between Al-Cu, in the presence of nickel coating and nickel powder on the bond strength of layers were evaluated. The conclusions drawn from the results can be summarized as follows:

1. Annealing heat treatment of Al-Cu-Ni cause the formation of intermetallics between layers.
2. 2 hours of annealing Al-Cu layers, in the presence of nickel coating on Cu, cause the formation of Al₄Cu₆ and AlNi intermetallic compounds.
3. The decrease in bonding ability of Al-Cu strips, after post-rolling annealing, was related to the formation of intermetallic compounds between two layers.
4. More intermetallics compounds forms during annealing in nickel coated composites rather than nickel powdered ones, thus inducing a weakening of the first ones.

REFERENCES

1. Prasad, S. V. and Asthana, R., "Aluminum metal – matrix composites for automotive applications". tribological considerations, 2004, 17, 445-453.

2. Suresh, S. and Mortensen, A., "Functionally graded metals and metal-ceramic composites", *International Materials Reviews*, 42, 1997, 85-116.
3. Shabani, A., Toroghinejad, M. R. and Shafyei, A., "Fabrication of Al/Ni/Cu composite by accumulative roll bonding and electroplating processes and investigation of its microstructure and mechanical properties". *Materials Science & Engineering A*, 2012, 558, 386-393.
4. Vaidyanath, L. R., Nicholas, M. G. and Milner, D. R., "Pressure welding by rolling". *British Welding Journal*, 1959, 6, 13-28.
5. Li, L., Nagai, K. and Yin, F., "Progress in cold roll bonding of metals". *Science and Technology of Advanced Materials*, 2008, 9, 023001-023011.
6. Pan, D., Gao, K. and Yu, J., "Cold roll bonding of bimetallic sheet and strips". *Materials Science and Technology*, 1989, 5, 934-939.
7. Saito, Y., Tsuji, N., Utsunomiya, H., Sakai, T. and Hong, R. G., "Ultra-fine grained bulk aluminium produced by accumulative roll-bonding (ARB) process". *Scripta Materialia*, 1998, 39, 1221-1227.
8. Eizadjou, M., Talachi, A. K., Manesh, H. D., Shahabi, H. S. and 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*, 2008, 68, 2003-2009.
9. Eizadjou, M., Manesh, H. D. and Janghorban, K., "Microstructure and mechanical properties of ultra-fine grains (UFGs) aluminum strips produced by ARB process". *Journal of Alloys and Compounds* 2009, 474, 406-415.
10. Tsuji, N., Ito, Y., Saito, Y. and Minamino, Y., "Strength and ductility of ultrafine grained aluminum and iron produced by ARB and annealing". *Scripta Materialia*, 2002, 47, 893-899.
11. Inoue, H., Ishio, M. and Takasugi, T., "Texture of TiNi shape memory alloy sheets produced by roll-bonding and solid phase reaction from elementary". *Acta Materialia*, 2003, 51, 6373-6383.
12. Ding, H. S., Lee, J. M., Lee, B. R., Kang, S. B. and Nam, T. H., "Processing and microstructure of TiNi SMA strips prepared by cold roll-bonding and annealing of multilayer". *Materials Science and Engineering A*, 2005, 408, 182-189.
13. Hebert, R. J. and Perepezko, J. H., "Deformation-induced synthesis and structural transformations of metallic multilayers". *Scripta Materialia*, 2004, 50, 807-812.
14. Yang, D., Cizek, P., Hodgson, P. and Wen, C. E., "Ultrafine equiaxed-grain Ti/Al composite produced by accumulative roll bonding". *Scripta Materialia*, 2010, 62, 321-332.
15. Wu, K., Chang, H., Maawad, E., Gan, W. M., Brokmeier, H. G. and Zheng, M. Y., "Microstructure and mechanical properties of the Mg/Al laminated composite fabricated by accumulative roll bonding (ARB)". *Materials Science and Engineering A*, 2010, 527, 3073-3078.
16. Zhang, R. and Acoff, V. L., "Processing sheet materials by accumulative roll bonding and reaction annealing from Ti/Al/Nb elemental foils". *Materials Science and Engineering A*, 2007, 463, 67-73.
17. Suna, Y. F., Tsuji, N., Fujii, H. and Li, F. S., "Cu/Zr nanoscaled multi-stacks fabricated by accumulative roll bonding". *Journal of Alloys and Compounds* 2010, 504, 443-447.
18. Guanghui, M., Jung-Moo, L., Suk-Bong, K. and Hyung-Wook, K., "Evolution of microstructure for multilayered Al/Ni composites by accumulative roll bonding process". *Materials Letters*, 2006, 60, 3255-3259.
19. Mozaffari, A., Manesh, H. D. and Janghorban, K., "Evaluation of mechanical properties and structure of multilayered Al/Ni composites produced by accumulative roll bonding (ARB) process". *Journal of Alloys and Compounds*, 2010, 489, 103-109.
20. Jamaati, R. and Toroghinejad, M. R., "Manufacturing of high-strength aluminum/alumina composite by accumulative roll bonding". *Materials Science and Engineering A*, 2010, 527, 4146-4151.
21. Jamaati, R. and Toroghinejad, M. R., "Application of ARB process for manufacturing high-strength, finely dispersed and highly uniform Cu/Al₂O₃ composite". *Materials Science and Engineering A*, 2010, 527, 7430-

- 7435.
22. Alizadeh, M. and Paydar, M. h., "Fabrication of nanostructure Al/SiCp composite by accumulative roll-bonding (ARB) process". *Journal of Alloys and Compounds*, 2010, 492, 231-235.
 23. Jamaati, R. and Toroghinejad, M. R., "High-strength and highly-uniform composite produced by anodizing and accumulative roll bonding processes". *Materials and Design*, 2010, 31, 4816-4822.
 24. Mohamed, H. A. and Washburn, J., "Mechanism of solid state pressure welding". *Welding Journal*, 1975, 302-310.
 25. Shabani, A. and Toroghinejad, M. R., Investigation of the Microstructure and the Mechanical Properties of Cu-NiC Composite Produced by Accumulative Roll Bonding and Coating Processes. *Journal of Materials Engineering and Performance*, 2015, 24, 4746-4754.
 26. Shabani, A., Toroghinejad, M. R. and Shafyei, A., "Effect of post-rolling annealing treatment and thickness of nickel coating on the bond strength of Al-Cu strips in cold roll bonding process". *Materials and Design*, 2012, 40, 212-220.
 27. Manesh, H. D. and Taheri, A. K., "The effect of annealing treatment on Mechanical properties of aluminum clad steel sheet". *Materials and Design*, 2003, 24, 617-622.
 28. Jamaati, R. and Toroghinejad, M. R., "Effect of Al₂O₃ nano-particles on the bond strength in CRB process". *Materials Science and Engineering A*, 2010, 527, 4858-4863.
 29. Abbasi, M., Taheri, A. K. and Salehi, M. T., "Growth rate of intermetallic compounds in Al/Cu bimetal produced by cold roll welding process". *Journal of Alloys and Compounds*, 2001, 319, 233-241.
 30. Won-Bae, L., Kuek-Saeng, B. and Seung-Boo, J., "Effects of intermetallic compound on the electrical and mechanical properties of friction welded Cu/Al bimetallic joints during annealing". *Journal of Alloys and Compounds*, 2005, 390, 212-219.
 31. Koberna, M. and Fiala, J., "Intermetallic phases in cold-welded Al-Cu joints". *Materials Science and Engineering A*, 1992, 159, 231-236
 32. Heness, G., Wuhner, R. and Yeung, W. Y., "Interfacial strength development of roll-bonded aluminium/copper metal laminates". *Materials Science and Engineering A*, 2008, 483-484, 740-742.
 33. Hyoung-Joon, K., Yeon, L. J., Kyung-Wook, P., Kwang-Won, K., hee, W. J., Sihyun, C., Jin, L., Jung-Tak, M. and Yong-Jin, P., "Effects of Cu/Al Intermetallic Compound (IMC) on Copper Wire and Aluminum Pad Bondability". *IEEE Transaction on Components and Packaging Technologies*, 26, 2003, 367-374.
 34. Chawla, N. and Chawla, K. K., "Metal Matrix Composites", *springer science + business media, Inc., New York*, 2006.