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Mechanical and Microstructural Characterization of Hybrid Cu-SiC-Zn Composites Fabricated Via Friction Stir Processing

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In this work, an attempt has been made to fabricate hybrid Cu-SiC-Zn composites by friction stir processing technique. Through this investigation the different number of passes was applied to assess the effect of pass adding on the mechanical, microstructural and dislocation density behavior of the specimens. Formation of the intermetallic phases between the copper matrix and Zn particles was discovered through the processed specimens. According to the obtained results, the higher passes led to obtain more uniform dispersion of the SiC particles and intermetallic phases. This higher level of particles and intermetallic phases' distribution causes remarkable reduction of grain sizes through the composites. Dislocation density for the processed specimens was determined by using the hardness measurement method. The calculated values for the dislocation densities showed that presence of SiC particles and intermetallic phases could rise up the dislocation values. Measured microhardness values for the composites exhibit that they enhanced rather than base metal and these results were confirmed by dislocation densities values of the specimens.

Keywords: *Friction stir processing, Intermetallic phases, Dislocation density, Mechanical properties, Microstructure*

1. Introduction

Due to special mechanical and thermal properties of intermetallic materials, researchers and industries have become interested in these materials in the recent years. Industrial demand such as aerospace and aircraft for use of high strength materials at elevated temperatures, has extended researches and investigations on these materials as well. These materials are good alternatives for the conventionally used super alloys and ceramics in the aerospace industry[1]. Materials with high wear and creep resistance at high working temperatures are the main desire of aerospace industry in which, intermetallic materials can meet them[2]. Production of these materials is the purpose of a lot of researches. Metallurgical processes such as pressure die casting, laser deposition and plasma spray are common methods to achieve an intermetallic[3]. Also, mechanical alloying methods are used to produce these materials[4]. Powder metallurgy is the widely used method to create intermetallic phases[5].

Friction stir processing (FSP) is a relatively new method to produce intermetallic phases which, has get the attention of researchers. Liming Ke et al. have produced Al-Ni intermetallic phases via FSP[6]. Chuang et al.[7] used FSP to produce Mg–Al–Zn intermetallic alloys. In comparison with the powder metallurgy methods, use of FSP for alloying and production of intermetallic is more simple and economic due to elimination of the secondary operations such as powder preparation, powder compaction and sintering[7].

FSP is an advanced mechanical and solid state process which, is mainly used for processing such as alloying, grain refinement and also fabrication of surface composite and nano-composite layers in the metals[8]. Rotational and traverse speeds of tool, geometry of tool, processing passes number and features of the secondary phase (mainly a reinforcing powder) are dominant processing parameters of this method which they can determine the characteristics of the product. Therefore, the effects of these parameters on the mechanical and metallurgical properties of the FSP products were investigated in different studies. Asadi et al.[9,10] produced AZ91/SiC and AZ91/Al_2O_3 composites and nano-composites by using FSP and examined the effects of passes number, rotational and traverse speeds on the quality of the achieved products. They concluded that in the higher passes number, the reinforcing particles are distributed uniformly, grains get finer and micro hardness of the products increases significantly. Lee et al.[11] reported a hardness enhancement and also a significant improvement in the high strain rate super plasticity of the base metal by adding SiO_2 powders into AZ61Mg alloy via FSP. Barmouz et al. produced copper base composites reinforced with SiC micro particles via FSP to increase surface property of the copper. They also investigated

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the effects of passes number, rotational and traverse speeds on the strength, wear resistance, micro hardness and micro structures of the composites[12,13].

Copper and zinc have a high tendency to make copper/zinc synthesis. Therefore, addition of zinc particles in copper/SiC composites can create copper/zinc intermetallic phases and increase wettability of copper matrix with SiC particles. Therefore, to achieve these composites and intermetallic with specific mechanical and metallurgical properties, in this work, ball-milled SiC particles and zinc powders were added into a copper matrix by using the FSP method. In order to make the FSP method more reliable and also repeatable, the electrical discharge machining (EDM) was used to create holes from beginning to the end of the processing paths and the particles were filled into the holes. Measurement of hardness values was used for determination of dislocation densities in specimens so that the mechanical behavior of the products could be understood more clearly.

2. Material and Methods

In this study, a pure copper plate with 130 mm length, 35 mm width and 5 mm thickness was used as the base metal. In order to produce surface composite layers, 5µm SiC particles were boll milled with zinc (Zn) for 2 hours in proportion of 60/40. The SiC/Zn compounds were compressed into the 1.5 mm diameter holes which are created using electro discharge machining (EDM) in a distance of 1 mm from the FSP surface in each specimen (Figure 1a and 1b). The next stage was plunging the tool by the pin into the plate for stirring and producing the composite (Figure 1c).

The FSP parameters were 1120 rpm in tool rotational speed and 40mm/min in traverse speed which was applied through the 1, 2 and 3 passes to fabricate the composites. Processing tool was tilted by an angle of 2^{o}. FSP tool was made of a hot working steel with a shoulder diameter, square pin diameter and length of 20, 5 and 3 mm, respectively. The specimens were clamped onto a thick St37 steel plate and the copper plate was fixed by four bolts.

Microstructural changes in the stirred zone (SZ) were examined using scanning electron microscopy (SEM) and the grain size was calculated using linear intercept method. FSPed surfaces were prepared by standard metallographic techniques and etched with a solution of 100 ml distillated water, 15 ml H_2O_2 and 2.5 gr $FeCl_3$. Microhardness of the specimens was measured on the cross section of the FSPed zone perpendicular to the processing direction using an indenter with a 100gr load for 30s.

The surfaces of the specimens used for indentation test were polished by alumina particles. The vickers indentation tests were applied using the loads in the range of 0.02 to 10N for dislocation density measurement.

Wide angle X-ray scattering (WAXS) analysis was performed using an X-ray diffractometer (Xpert, Philips) operating at 40 kV and 30 mA for Cu Kα radiation (λ=0.154 nm). XRD scans were performed on the cross-sections of the specimens and perpendicular to the FSP direction.

3. Results and Discussions

3.1. Microstructure

Figure 2 shows the SEM image of the 1-pass FSPed Cu-SiC/Zn composites morphology and EDX results. Formation of the intermetallic compounds could be seen in this image. The EDX results are also describing the atomic and weight percentage of the copper and zinc in the marked zones. It is indicated that the amount of copper is approximately six times of zinc and there is no sign of SiC component which could be attributed to the low level of SiC particles distribution through one pass FSP.

Figure 3 illustrates the morphology of the intermetallic compounds in 2-pass FSPed specimen using SEM image which is describing a slightly better distribution of the intermetalic phases at this specimen rather than that of one pass FSPed spacemen. The EDX results are embedded in this figure illustrating that 2-pass FSP distributes the Zn better than that of 1-pass. It is also shown that the copper

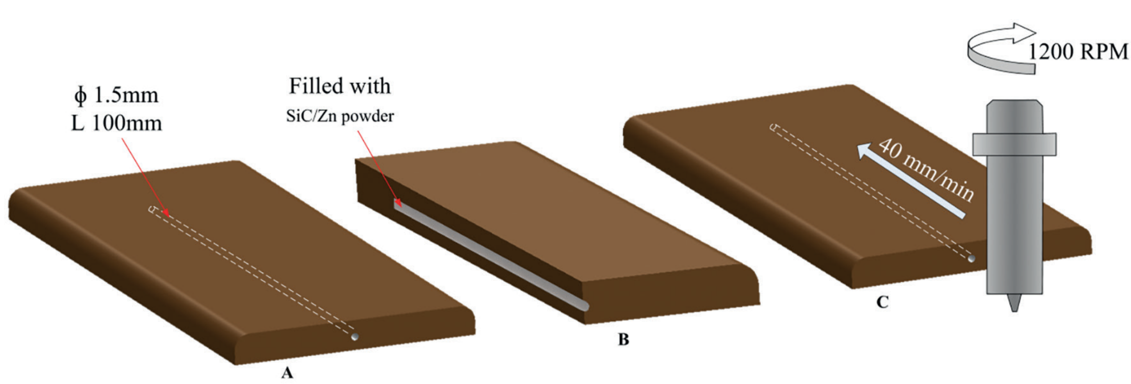


Figure 1: Schematic of the friction stir processing (a) drilling the hole in the specimen, (b) compressing the fillers into the hole and (c) plunging the tool into the plate.

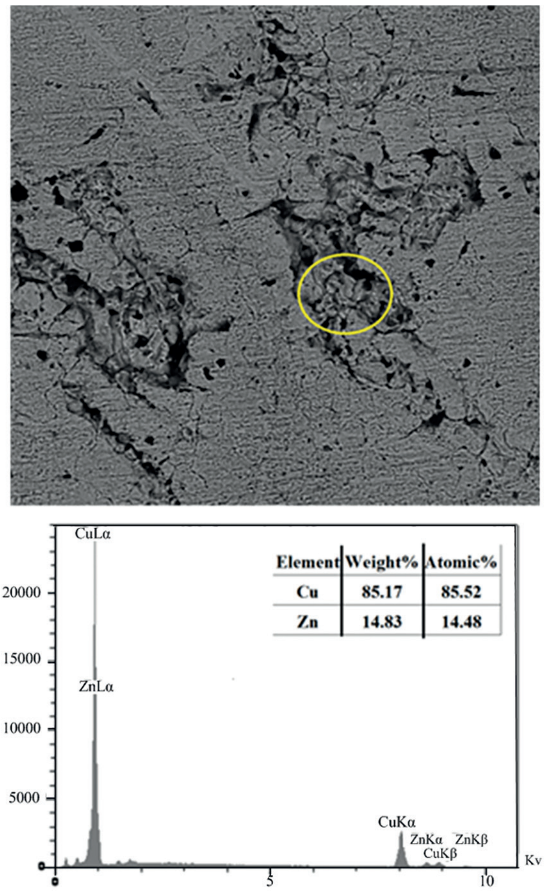


Figure 2: SEM images of the intermetallic compounds in the 1-pass FSPed specimen and EDX result.

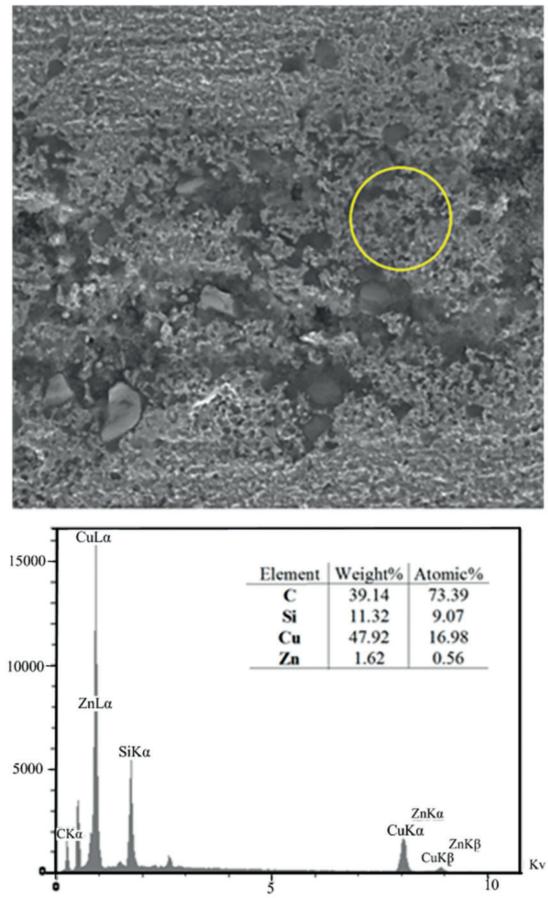


Figure 3: SEM images of the intermetallic compounds in the 2-pass FSPed specimen and EDX result.

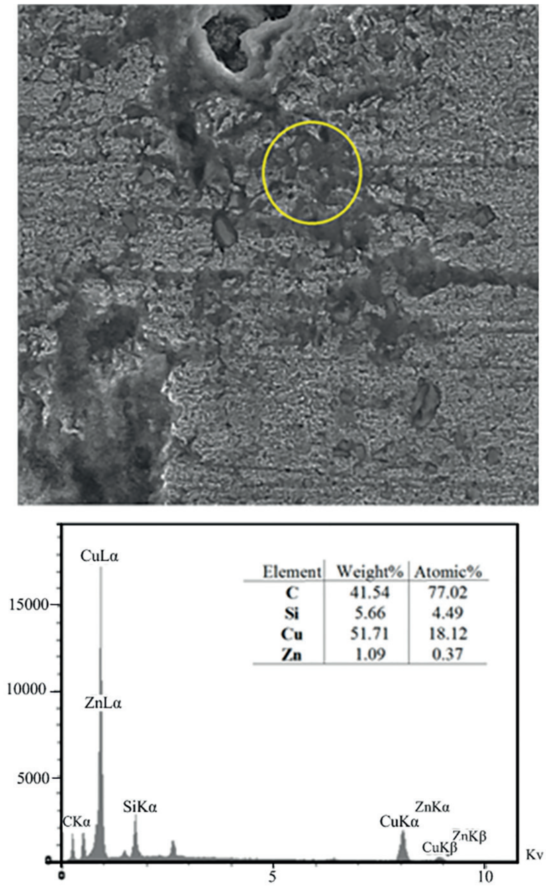
percentage is 30 times of Zn in the mentioned region. Presence of C and Si in this region shows the well separation of SiC particles in the matrix. Higher mechanical stresses and heat generation in the 2-pass FSPed specimen provides a severe material flow in the processed zone which surely distributes the different phases more efficient than 1-pass. As reported in our previous works, this phenomenon also could break down the SiC particles as a result of shear stresses[12].

Figure 4 exhibits the SEM image of the 3-pass FSPed specimen. The results are dealing with presence of intermetallic compounds in processed composite. The EDX results exhibit the increase of the copper percentage proportion to Zn to the amount of 50. This proves the represented ideas already about the severe effect of further FSP passes on the improving the distribution level of the phases. It is noticeable that Si and C are also existed in the region which is presenting the presence of SiC particles.

It is clear in the EDX results that with increase of FSP passes, the volume fraction of copper tend to be lessened in the regions revealing the fact that higher level of SiC and Zn distribution inside the matrix has been obtained.

Figure 5 shows the X ray diffraction analysis pattern for the fabricated composites. The formation of the intermetallic phases between the Cu and Zn are illustrated at this figure. Development of intermetallic phases including $CuZn_5$ and Cu_5Zn_8 inside the processed composites is confirmed by these results. As it is shown in Figure 5 in the composites fabricated by single pass FSP, only the intermetallic phase of $CuZn_5$ is created while there are exist two different intermetallic phases of $CuZn_5$ and Cu_5Zn_8 inside the 2- and 3-pass FSPed composites.

Figure 6a-d shows the SEM images of the grains size in the reference pure copper and composites fabricated by 1-, 2- and 3-pass FSP, respectively. As it is seen, the grain sizes in the composites are dramatically decreased rather than that of pure one. This is on account of the resisting effect of the SiC particles and intermetallic phases against grain boundaries migration and dynamic recrystallization. High value of heat generation in the stir zone tends to anneal and coarsen the grains, while formation of new nucleation sites as a result of dynamic recrystallization prevents the grain boundaries growth and migration. The pinning effect of SiC particles together with intermetallic phases chains hindering



Element	Weight%	Atomic%
C	41.54	77.02
Si	5.66	4.49
Cu	51.71	18.12
Zn	1.09	0.37

Figure 4: SEM images of the intermetallic compounds in the 3-pass FSPed specimen and EDX result.

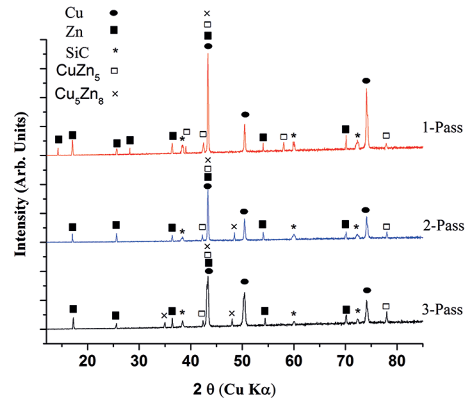


Figure 5: X ray diffraction of the Cu-Sic/Zn composites

effect act as another important parameter to control the grain size. As a result the grains sizes in the processed composites begin to reduce. It is notable that as discussed previously, the higher passes of FSP enhances the distribution level of SiC particles and intermetallic phases which is the reason for smaller grains size in the 3-pass FSPed specimens compared to that of the 2-pass one.

3.2. Dislocation density determination

Regarding the importance of mechanical properties in metals, using simple methods to find out what phenomena could govern them would be very precious. There are some valuable reports on the field of hardness indentation size effect to estimate the dislocation densities which are confirmed by transmission electron microscopy (TEM) results. According to these reports, dislocation densities act as a determinant factor in controlling the mechanical properties of metals[13,14,15,16]. Barmouz et al.[17] reported the dislocation density of FSPed pure copper via hardness measurement. They reported that 2-pass FSP reduces the dislocation density rather than that of single pass.

Considering previous reports in this field1[5,16] the hardness values increase as the indentation depth decreases. The relation between the hardness values and indentation depth is expressed as follows[16]:

$$\left(\frac{H}{H_0}\right)^2 = 1 + h^*.\left(\frac{1}{h}\right) \qquad (1)$$

where H_0 is the hardness in the limitation of infinite depth (bulk hardness) and h^* is a characteristic length. Figure 7 shows the amount of h^* in the 1-pass FSPed composite which is measured to be 2096 nm and determined by fitting the Eq. 1 to the experimental data.

The correlation between the dislocation density statically stored in the lattice (ρ_s) and the characteristic length (h^*) is expressed as follows[13,14].

$$\rho_s = \frac{3}{2}\frac{1}{f^3}\frac{\tan^2\theta}{bh^*} \qquad (2)$$

Where Θ is the angle between the surface of the plate and the indenter, b the burgers vector of the dislocation and f is a correction factor for the size of the plastic zone. In this research $\Theta = 20^\circ$, $f = 1.9$[15,16], and $b = 0.25$ nm[15,18].

Based on the above-mentioned equations, the dislocation densities for the single-pass FSPed composite is measured to be up to 5.52×10^9 cm^{-2}.

Figure 8 shows the fitting process of experimental hardness values for the composite fabricated using 2-pass FSP. As it is seen the h^* value is derived from the equation and is equal to 749.9 nm. By replacing this value into equation 2, the dislocation density is measured to be 1.54×10^{10} cm^{-2} which demonstrates a considerable growth in dislocation density of the 2-pass FSPed specimen in comparison with that of fabricated by 1-pass. The parameters which control the dislocation density in this process are heat generation, dynamic recrystallization, mechanical stresses and presence of SiC particles and intermetallic phases. Most of these factors tend to increase the dislocation density in the matrix which

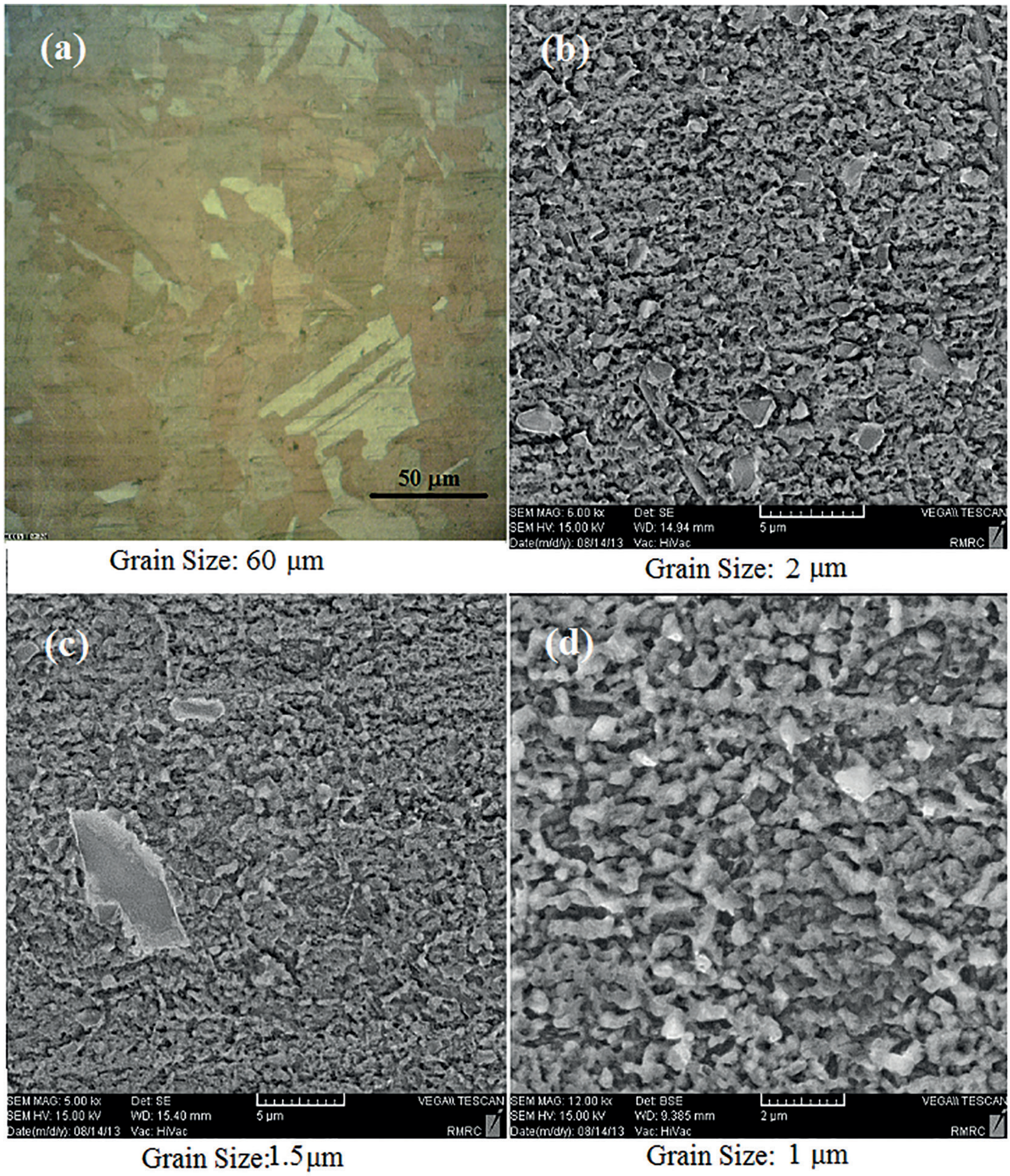


Figure 6: (a) Optical microscopy of the base metal and (b-d) SEM images of the 1-, 2- and 3-pass FSPed composites, respectively.

could be explain as follows; dynamic recrystallization forms new nucleation sites in the matrix and causes dislocation propagation; mechanical stresses resulting from severe material flow in the stir zone is the important reason to increase the dislocation density; presence of SiC particles and intermetallic compound act as the disbondings in the matrix and weaken its integrity and subsequently bring about dislocation density enlargement. The only factor contributing to decrease the dislocation density is the input heat which it did not act as a dominant factor to govern it. It is clear that abovementioned factors perform more intense in 2-pass FSPed composite rather than that of one pass and accordingly they lead to higher value of dislocation density.

Figure 9 illustrates the fitting process of experimental hardness values in order to calculation of the dislocation density in the 3-pass FSPed composite. The h^* value for this specimen is calculated in this figure and is up to 1454.33 nm. Collocation of the h^* value into equation (2) estimates the dislocation density for this specimen near to 7.96×10^{9} cm^{-2}. It is discovered that the dislocation density value in 3-pass FSPed composite decreased compared to that of 2-pass FSPed one. This phenomenon could be explained regarding the dominant effect of input heat through three passes of FSP

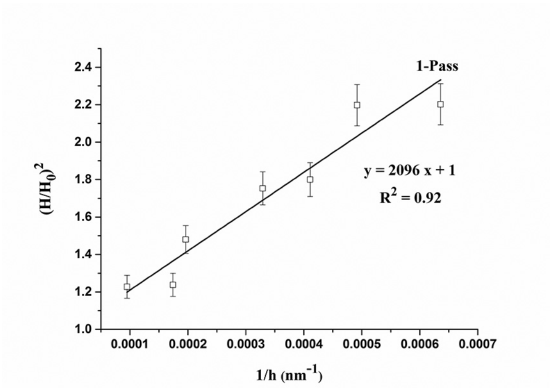


Figure 7: Fitting of Eq.(1)to the microhardness data of 1-pass FSPed composite.

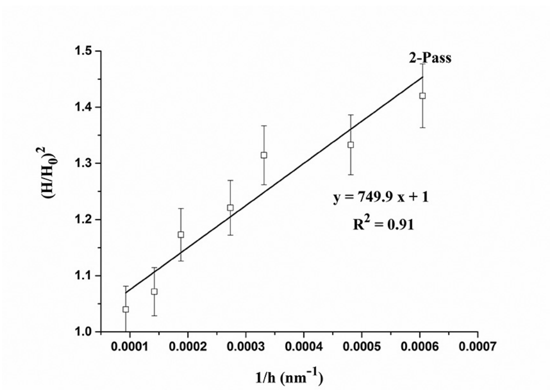


Figure 8: Fitting of Eq.(1)to the microhardness data of 2-pass FSPed composite.

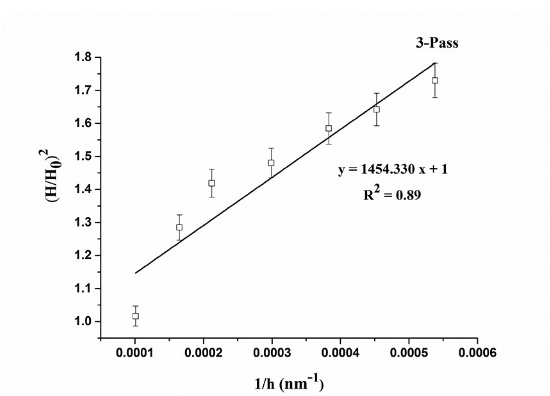


Figure 9: Fitting of Eq.(1)to the microhardness data of 3-pass FSPed composite.

which annihilates the dislocations. According to different reports of previous investigations, after the second FSP pass; the pass-adding effects on microstructure and mechanical behavior are negligible while the input heat remains similar to the previous passes. Actually, in third pass of FSP all the above-discussed congruent factors with dislocation density enhancement lose their effectiveness and input heat can play governing role in reduction of the dislocation density.

3.3. Microhardness behavior

Figure 10 shows the microhardness values of the different specimens fabricated by 1-, 2- and 3-pass FSP. As it is known the dislocation density is one of the most important factors controlling the microhardness behavior of the specimens. The microhardness results confirm the calculated dislocation values for each specimen such that higher dislocation density blocks the grain boundaries and dislocation sliding. In 1-pass FSPed specimen due to the poor dispersion of the SiC particles and intermetallic phases, the grains and dislocations have more room for sliding and motion which make the penetration of the indenter easier. In the case of 2-pass and 3-pass FSPed specimens according to the measured dislocation densities the 2-pass one has higher hardness values which is sign of accumulation of dislocation densities preventing grain boundaries motion.

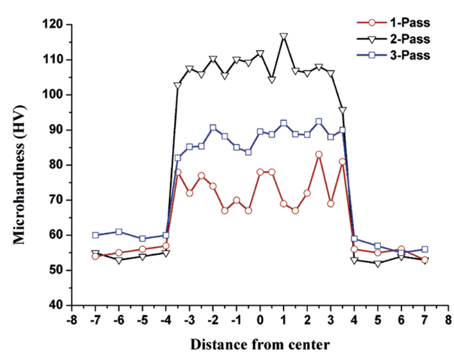


Figure 10: Microhardness values of the 1-, 2- and 3-pass FSPed composites.

4. Conclusion

The hybrid composite of Cu-SiC/Zn was fabricated and characterized successfully through this investigation. Multi pass friction stir processing was conducted on the specimens to improve the dispersion of the SiC particles and intermetallic phases into the matrix. Some of the important achievements are listed below:

1- The intermetallic phases were formed through the composite fabrication process by FSP and the dispersion of these phases was getting improved in higher passes.

2- The grain sizes of the fabricated composites were noticeably decreased rather than that of pure one and the higher passes of FSP led to more intense grain refinement.

3- Dislocation density of the 2-pass FSPed composites was increased rather than that of 1-pass one, whereas 3-pass FSP diminished the dislocation density up to the value between 1-pass and 2-pass FSPed specimens.

4- Microhardness values of the processed composites were considerably enhanced rather than that of pure copper and their dislocation densities values were the dominant factor to control the microhardness behavior.

5. References

1. Koch CC. Intermetallic matrix composites prepared by mechanical alloying–a review. *Materials Science and Engineering: A*. 1998;244(1):39-48.

2. Antolak-Dudka A, Krasnowski M, Kulik T. Nanocrystalline Ni_3Al intermetallic produced by hot-pressing consolidation of mechanically alloyed powders. *Intermetallics*. 2013;42:41-44.

3. Joshi V, Srivastava A, Shivpuri R. Intermetallic formation and its relation to interface mass loss and tribology in die casting dies. *Wear*. 2004;256(11-12):1232-1235.

4. Yamauchi A, Yoshimi K, Kurokawa K, Hanada S. Synthesis of Mo–Si–B *in situ* composites by mechanical alloying. *Journal of Alloys and Compounds*. 2007;434-435:420-423.

5. Verdian MM, Raeissi K, Salehi M. Processing and electrochemical characterization of Ni_2Si intermetallic compound produced by vacuum sintering. *Vacuum*. 2013;90:1-5.

6. Ke L, Huang C, Xing L, Huang K. Al–Ni intermetallic composites produced in situ by friction stir processing. *Journal of Alloys and Compounds*. 2010;503(2):494-499.

7. Chuang CH, Huang JC, Hsieh PJ. Using friction stir processing to fabricate MgAlZn intermetallic alloys. *Scripta Materialia*. 2005;53(12):1455-1460.

8. Faraji G, Asadi P. Characterization of AZ91/alumina nanocomposite produced by FSP. *Materials Science and Engineering: A*. 2011;528(6):2431-2440.

9. Asadi P, Besharati Givi MK, Abrinia K, Taherishargh M, Salekrostam R. Effects of SiC Particle Size and Process Parameters on the Microstructure and Hardness of AZ91/SiC Composite Layer Fabricated by FSP. *Journal of Materials Engineering and Performance*. 2011;20(9):1554-1562.

10. Lee CJ, Huang JC, Hsieh PJ. Mg based nano-composites fabricated by friction stir processing. *Scripta Materialia*. 2006;54(7):1415-1420.

11. Barmouz M, Besharati Givi MK, Seyfi J. On the role of processing parameters inproducing Cu/SiC metal matrix composites via friction stir processing: Investigating microstructure, microhardness, wear and tensile behavior. *Materials Characterization*. 2011;62(1):108-117.

12. Barmouz M, Besharati Givi MK. Fabrication of in situ Cu/SiC composites using multi-pass friction stir processing: Evaluation of microstructural, porosity, mechanical and electrical behavior. *Composites Part A: Applied Science and Manufacturing*. 2011;42(10):1445-1453.

13. Liu Y, Ngan AHW. Depth dependence of hardness in copper single crystals measured by nanoindentation. *Scripta Materialia*. 2001;44(2):237-241.

14. Swadener JG, Misra A, Hoagland RG, Nastasi M. A mechanistic description of combined hardening and size effects. *Scripta Materialia*. 2002;47(5):343-348.

15. Graça S, Colaço R, Vilar R. Indentation size effect in nickel and cobalt laser clad coatings. *Surface and Coatings Technology*. 2007;202(3):538-548.

16. Graça S, Colaço R, Carvalho PA, Vilar R. Determination of dislocation density from hardness measurements in metals. *Materials Letters*. 2008;62(23):3812-3814.

17. Barmouz M, Abrinia K, Khosravi J. Using hardness measurement for dislocation densities determination in FSPed metal in order to evaluation of strain rate effect on the tensile behavior. *Materials Science and Engineering: A*. 2013;559:917-919.

18. Csiszár G, Misra A, Ungár T. Burgers vector types and the dislocation structures in sputter-deposited Cu–Nb multilayers. *Materials Science and Engineering: A*. 2011;528(22-23):6887-6895.