Int J Adv Manuf Technol (2014) 75:1331–1337
DOI 10.1007/s00170-014-6162-3

ORIGINAL ARTICLE

In situ formation of Al–Al_3Ni composites on commercially pure aluminium by friction stir processing

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Received: 20 October 2013 / Accepted: 8 July 2014 / Published online: 17 August 2014

Abstract Possibility of the formation of Al–Al_3Ni composite layers on commercial pure aluminium plates by friction stir processing (FSP) has been studied. It is believed that the hot working nature of FSP can effectively promote the exothermic reaction between Al and added Ni powder to produce Al_3Ni intermetallic compounds in the aluminium matrix. In this study, the effects of the rotational and traverse speed of the tool as well as the number of FSP passes on the in situ formation of Al_3Ni in aluminum matrix were examined. Besides, the microstructure and microhardness of the fabricated surface layers were also studied. The results showed that the ratio of tool rotational speed to traverse speed (ω/υ) is the main controlling parameter of the heat generated during FSP and hence the reaction between aluminium and nickel. Increasing the number of FSP passes also promoted the reaction between Ni and Al and improved the distribution of Al_3Ni compounds, too. The composite layer achieved by six passes of FSP showed the highest hardness, which was almost twice of that of the base metal.

Keywords Aluminium · Friction stir processing · In situ composite · Al–Ni intermetallic

1 Introduction

Applications of friction stir processing (FSP) for fabrication of composite surfaces has attracted research interest in recent years [1–3]. During this process, the rotating tool penetrates into the surface of the workpiece and traverse along a predetermined track. The friction and plastic deformation imposed by the tool heats and softens the workpiece, and the rotation of the tool promotes intermixing of material in a region in the vicinity of the interface of the tool and the workpiece. When reinforcing particles are introduced in to a hole or a groove on the surface of the workpiece in the path of the tool, they are dispersed throughout the stir zone [4, 5]. Mishra et al. [6] first demonstrated the application of FSP technology to fabricate AA5083/SiC. In their research, aluminium plates preplaced SiC particle were subjected to FSP. Al/SiC composite layers with well-distributed particles and good bonding with aluminium matrix were generated via controlling of FSP parameters. The microhardness (HV) of the fabricated surface composite was doubled with 27 vol.% of SiC particles. In other work, Dixit et al. [7] successfully dispersed NiTi compounds in Al1100 matrix via FSP. In their research, four small holes, 1.6 mm in diameter and 76 mm in length, were drilled at about 0.9 mm below the surface in Al1100 plates received in H14 condition. NiTi powders with particles in the size range of 2–193 μm were trapped inside the holes. The powder filled plates were then subjected to FSP at a tool rotational rate of 1,000 rpm and traverse speed of 25 mm/min. It was stated that the embedded particles were uniformly distributed, had strong bonding with the matrix, and no interfacial products were formed during the processing. The experimental and the modeled values showed improved mechanical properties in the prepared composite.

Besharati et al. [8] produced magnesium based metal matrix composite (MMC) by FSP. In order to produce surface composite layers, SiC particles were contrived in a groove in the middle of the specimens. Then, the SiC particles were compressed into the groove, and the next stage was plunging the tool into magnesium for stirring of the matrix and producing the composite. This study showed that using 5-μm SiC particles reduced the grain size in the stir zone from 150 to 7.17 μm, and as a result, the hardness of the stir zone increased from 63 to 96 HV.

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Huang et al. [9] fabricated bulk multi-element Mg base alloys with different fractions of AZ31 sheets and Al and Zn foils by FSP. After multi-passes FSP, some intermetallic compound phases were generated. The hardness of the multi-element Mg base alloy made by FSP reached a maximum value of nearly 400 HV, especially in the Mg37.5Al25Zn37.5 system.

Furthermore, Kao et al. [10] used FSP to obtain in situ aluminium matrix composites of Al–Al_2Cu, Al–Al_3Ti [11], or Al–$Al_{13}Fe_4$ [12]. For this purpose, relevant powders (Al with Cu or Ti or Fe) were mixed, hot pressed, and sintered. Then, FSP was applied to produce in situ intermetallic particulate-reinforced composites. Up to now, there have not been any reports on the achievement of in situ intermetallic particulate-reinforcement composite without pre- or postheat treatment. Therefore, in the present work, the feasibility of fabrication Al_3Ni–Al composite using of friction stir processing without any pre- or postheat treatment such as sintering or annealing is demonstrated. Several Al–Al_3Ni composite layers were fabricated using multiple FSP passes by employing various rotational and traverse speeds of the tool. In addition, the effects of rotational and traverse speed on the microstructure and hardness of the fabricated layer were also studied.

2 Experimental procedures

The specimens with the dimension of 120×40×8 mm^3 were cut from a commercial pure Al plate. Grooves (100×1×3 mm^3) were cut at the center of aluminium specimens, in which nickel powder (99 % purity, average diameter <10 μm) was filled (Fig. 1b). The nominal chemical composition of the alloy is shown in Table 1.

Table 1 Chemical composition of 1100 Al alloy plate (wt%)

Si	Mg	Fe	Cu	Zn	Ti	Mn	Al
0.08	0.005	0.15	0.01	0.007	0.005	0.012	Balance

A conventional milling machine was used to conduct the FSP experiments. FSP tool was made from H13, heat treated and hardened up to 52 HRC, which had columnar shaped shoulder (ø=20 mm) and tapered pin. Figure 1a shows the schematic of the FSP tool.

In order to prevent the splashing of the powder during FSP, the groove was closed with pinless tool before FSP (Fig. 1b). The final stage was plunging of tool with pin inside the plate for producing composites (Fig. 1c). All combination of two rotational speeds of 1,600 and 2,000 rpm and two traverse speeds of 12 and 25 mm/min were applied in the experiments. The plates were subjected to two, four, and six passes of FSP. It should be noted that after each pass, the rotational direction of tool was changed. Meanwhile, each FSP pass was applied after the specimen had been cooled down to room temperature. The working parameters are listed in Table 2. To characterize the treated specimens, X-ray diffraction (Cu Kα, model Philips X'Pert pro) was used to identify the present phases in the fabricated layers. Cross-sectional microstructural observation (region A in Fig. 1c) was carried out by employing optical

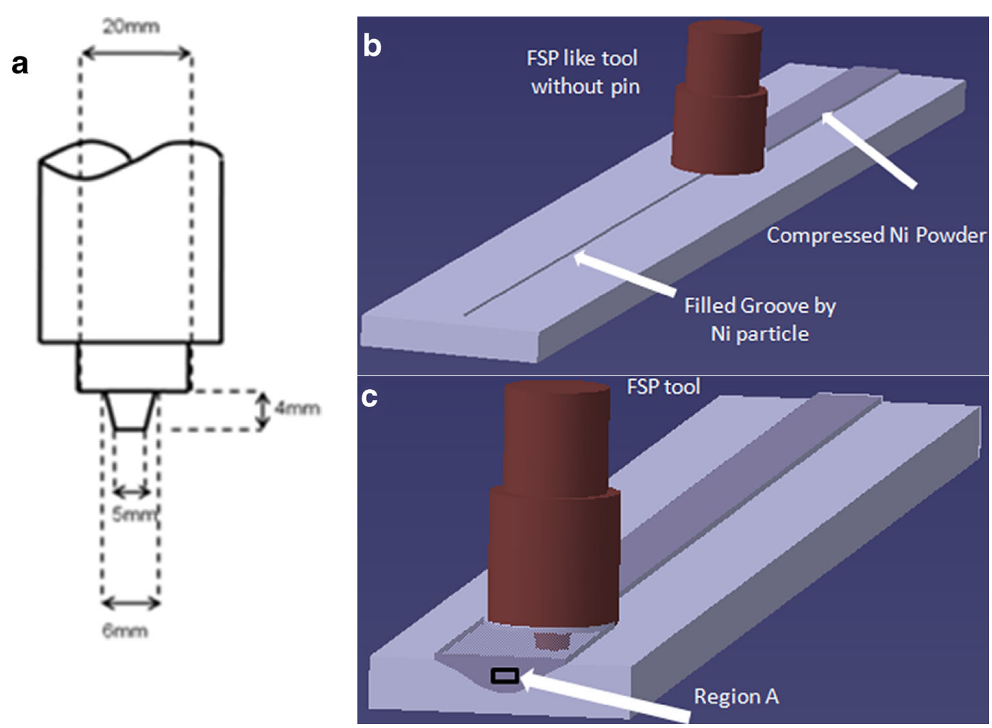


Fig. 1 Schematic of **a** the tool and **b**, **c** the friction stir processing applied. Microscopic analysis were done in the region A (stir zone)

Table 2 Working parameters used in this study

Traverse speed	25 mm/min			12 mm/min		
Rotational speed (rpm)	2 passes	4 passes	6 passes	2 passes	4 passes	6 passes
1,600	√	√	√	√	√	√
2,000	√	√	√	√	√	√

microscope (OM, model GipponGDCE-30) and scanning electron microscope (SEM, model CamScan MV2300) equipped with energy dispersive spectrometer (EDS). Microhardness was measured on the cross-section of the specimens using a Vickers microhardness tester with an applied load of 50 g for 20 s.

3 Results and discussion

3.1 XRD phase characterization

Figure 2a–d shows the XRD patterns of the treated specimens. As it is seen Al_3Ni, Al and Ni phases are detected in them.

The thermodynamic calculation results show that the Gibbs free energy of the reaction between Al and Ni to form Al_3Ni from room temperature to 773 K is negative, which thermodynamically confirms the possibilities of the occurrence of this reaction [13]. Detected Al_3Ni in XRD patterns (Fig. 2) reveals that the in situ reaction between nickel powders and aluminium matrix has been occurred.

According to literatures, the average maximum temperature on aluminium plates is a function of the pseudo-heat index w ($w=\omega^2/\upsilon$). It has been demonstrated that the general relationship between maximum welding temperature (T, °C) and FSW parameters (ω, υ) for several aluminium alloys can be explained by Eq. 1 [4],

$$T/T_m = k\left[\omega^2/\left(\upsilon \times 10^4\right)\right]^{\alpha} - \quad (1)$$

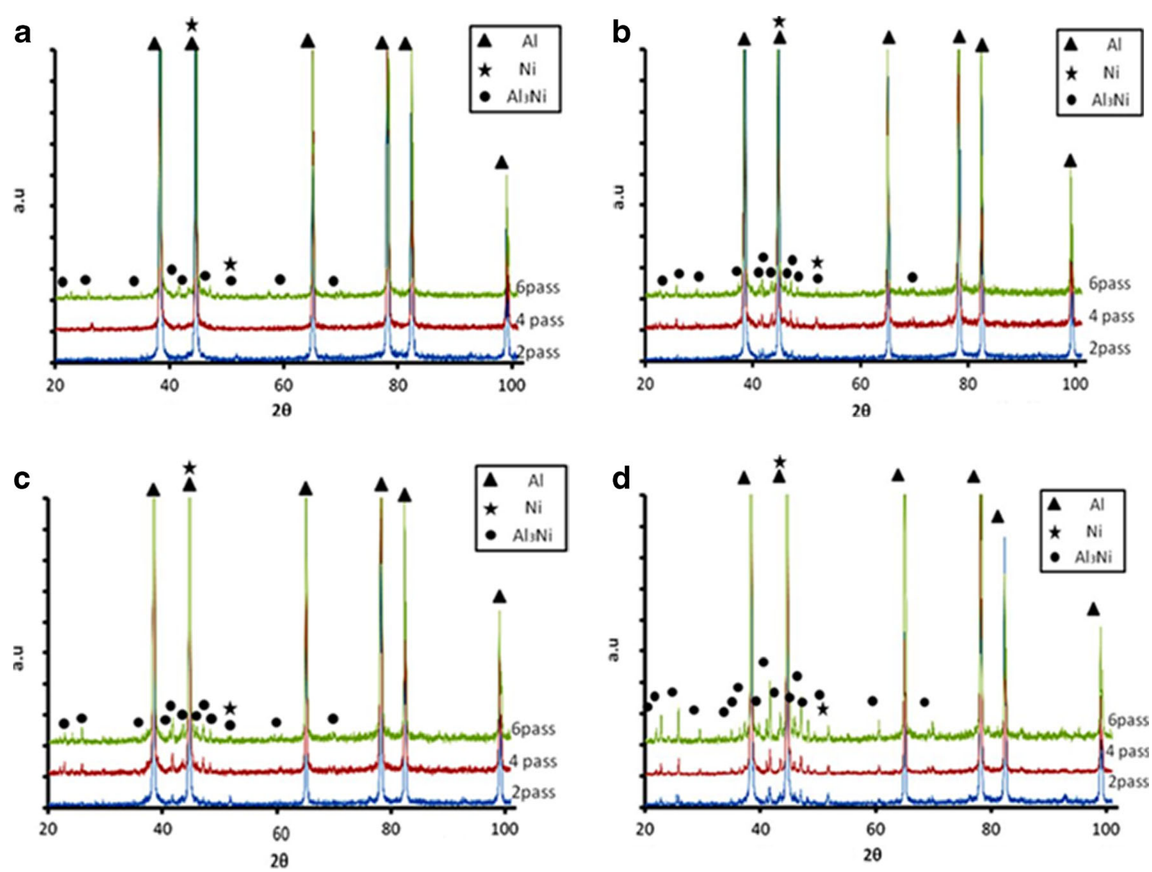


Fig. 2 **a**–**d** X-ray diffraction patterns of specimens fabricated at different rotational and traverse speeds in multiple passes at **a** 1,600 rpm and 25 mm/min, **b** 2,000 rpm and 25 mm/min, **c** 1,600 rpm and 12 mm/min, and **d** 2,000 rpm and 12 mm/min

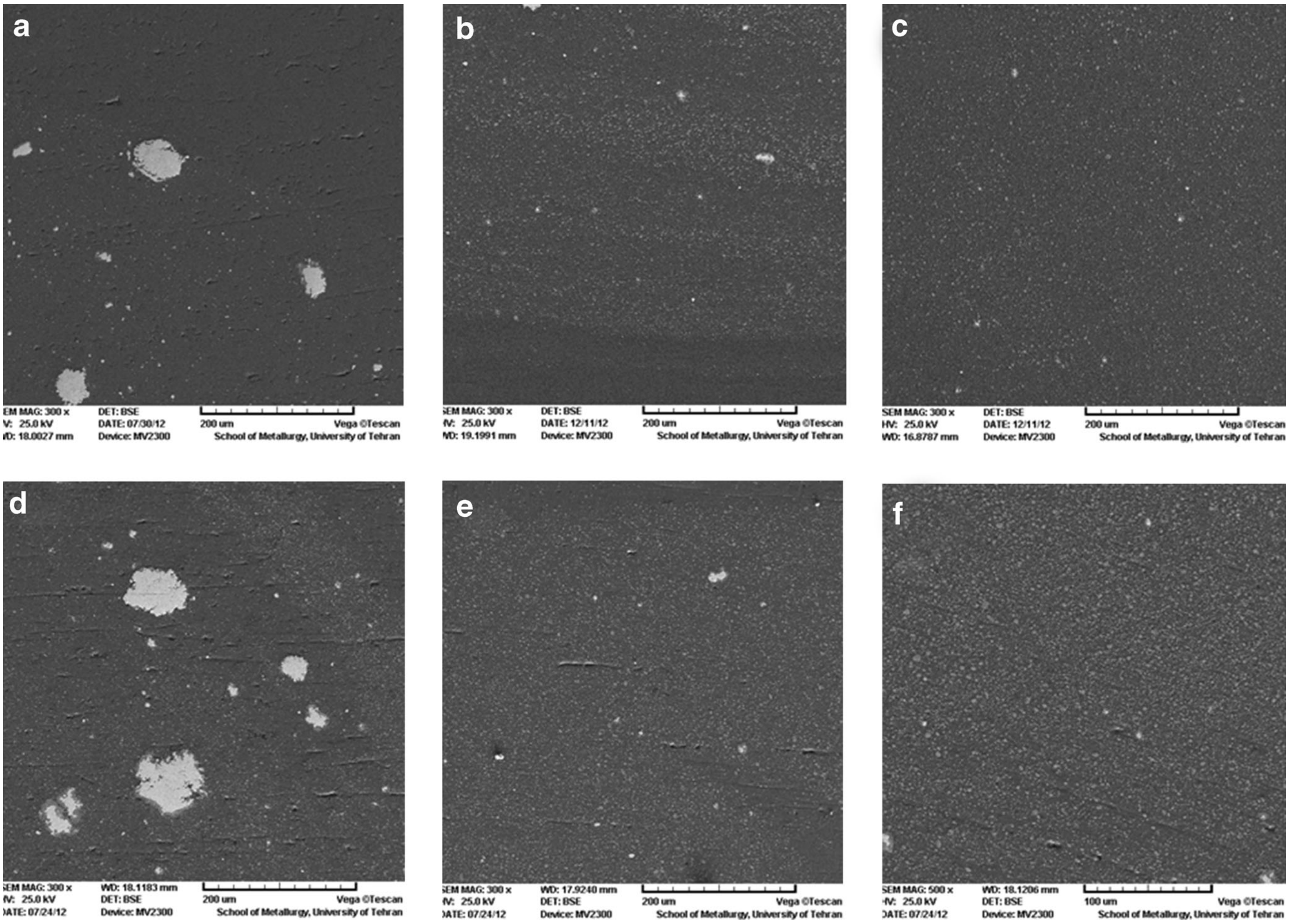
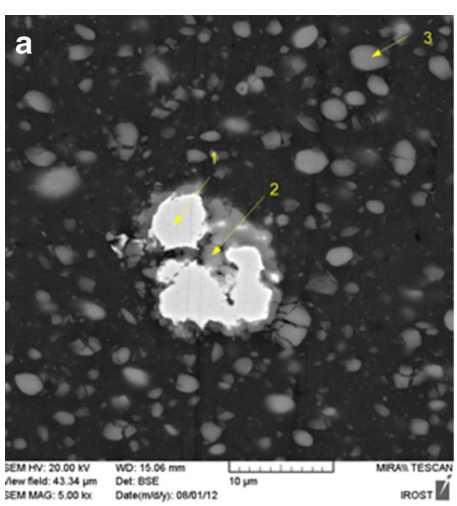


Fig. 3 SEM micrographs of the specimens at 1,600 rpm and 12 mm/min: **a** 2 passes, **b** 4 passes, and **c** 6 passes at 2,000 rpm and 12 mm/min, **d** 2 passes, **e** 4 passes, and **f** 6 passes

where the exponent α is between 0.04 and 0.06, the constant k is between 0.65 and 0.75, and T_m (°C) is the melting point of the substrate [4]. The characteristic peaks of Al_3Ni were noticed at ω/υ ratio higher than 80 for two passes of FSP (Fig. 2b). Hence, it can be concluded that the ratio lower than 80 are not high enough to support required heat input for aluminium nickel reaction. These peaks were better developed as the FSP applied passes increased from two to six for all conditions (Fig. 2a–d).

Figs. 4 **a**, **b**. BSE micrograph (**a**) and the relevant EDS analysis (**b**) of the specimen produced at 2,000 rpm 12 mm/min after 2 pass. (**a**) The white marked as 1 is nickel and the light gray phase marked as 2 is Al_3Ni and dark matrix is Al



b EDS analysis

region	Al(%at)	Ni(%at)	Phases
1	1	99	Ni
2	76	24	Al_3Ni
3	79	21	Al_3Ni

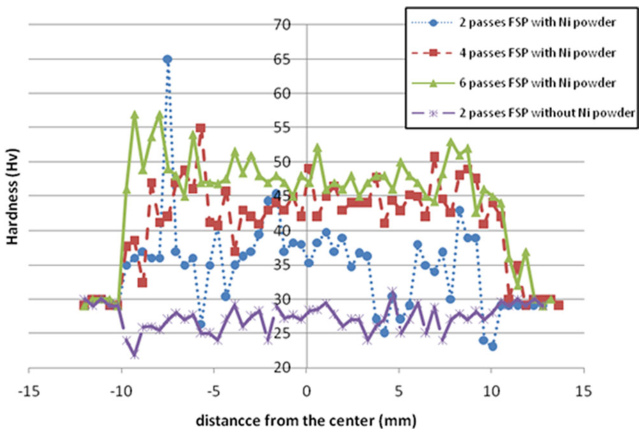


Fig. 5 Transverse microhardness profiles of the specimens with and without addition of nickel powders

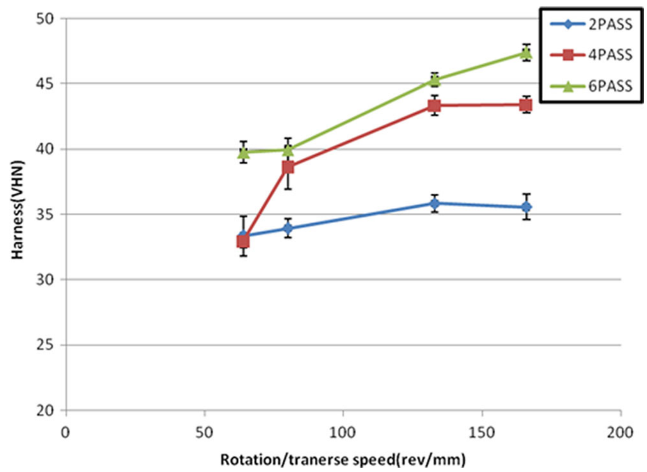


Fig. 7 Variations of hardness in relation to pseudo-heat index

Hence, by increasing the ratio of rotational speed to traverse speed (ω/υ), the amount of friction and plastic deformation is increased and so heat input and plastic deformation. Meanwhile, by providing proper conditions for diffusion, i.e., increasing temperature, diffusion route (dislocation), and also contact surfaces between Ni particles and aluminum plate, formation of intermetallic compounds is encouraged. It was also noted that the reaction between Al and Ni was not completed even after six FSP passes because nickel powder still existed in the stirred zone.

4 Microstructural analysis

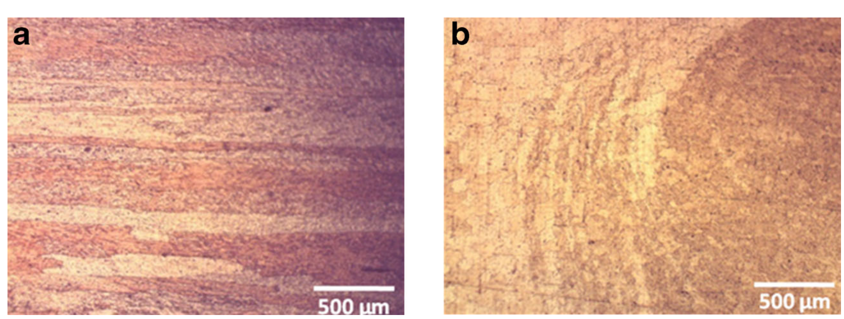
Figure 3a–c shows the SEM micrographs of the specimens with the ω/υ ratio equal to 133.3. It is exhibited in Fig. 3a that by two passes of FSP, it was not possible to achieve a homogenized composite and agglomeration of primary nickel powders was observed after two passes. However, by increasing the number of FSP passes, the produced intermetallics were distributed more uniformly (Fig. 3c)

In Fig. 3d–f, SEM micrographs of specimens with the ω/υ ratio equal to 166.6 are exhibited. By comparing Fig. 3d and f, it can be concluded that by increasing rotational speed, the cluster size of nickel powders decreases and the distribution of in situ intermetallics becomes more homogenized, due to increasing of heat input and material flow in the stir zone. Other researchers have also confirmed the homogeneity improvement by increasing the ω/υ ratio as well as increasing the number of FSP passes [14, 15].

Higher magnification SEM micrograph in Fig. 4a reveals gray areas around white nickel cluster that according to EDS analysis (Fig. 4b) appears to be Al_3Ni. EDS analysis identified the brightest phase as Ni, the dark gray matrix as Al, and the light gray areas (which have surrounded Ni powder or dispersed in the Al matrix) as a Al–Ni compound. According to the XRD and EDS results, this compound appears to be Al_3Ni intermetallic.

FSP produces intense plastic deformation with a strain rate of 10° to 10^3 s^{-1} and a strain up to ~40, which result in the significant break up of primary nickel powders, material mixing, and better diffusion of elements [3]. In addition, the high density of dislocation is introduced at the interface of nickel powder and the matrix, which can provide many paths for diffusion of alloying elements in the matrix. Furthermore, the reaction between nickel and aluminium is an exothermic reaction, which can promote the diffusion of elements. As a result, these conditions can increase possibility of achieving in situ compounds via FSP [16]. Applying further FSP passes can enhance the contact area of Al–Ni interface by shearing

Fig. 6 Optical micrograph of **a** base metal and **b** specimen at two passes without Ni added



the metal powders and breaking the intermetallic layer surrounding the large nickel powders as shown in Fig. 3c and f, which promote the reaction.

4.1 Hardness evaluation

The hardness of the base material after FSP slightly decreased (Fig. 5) in spite of grain refinement achieved by the process, which is shown in Fig. 6. This is due to the fact that, during FSP of aluminum, the temperature can raise to about 0.6 to 0.9 T_m, which can have annealing effects and reduce the density of dislocation as well as residual compressive stress of initially rolled aluminium sheets.

Therefore, during FSP of aluminium without nickel powder addition, there is a competition between the two aforementioned phenomena that affect hardness vice versa. However, in the case of FSP with the presence of nickel powders, the strengthening mechanisms are different. There are four possible strengthening mechanisms in MMCs [15, 17]: grain boundary strengthening (Hall–Petch relationship), dislocation pinning due to reinforcement particles (Orowan theory), dislocation induced due to the difference in coefficient of thermal expansion (CTE) between matrix and reinforcement material, and work hardening due to the strain misfit between the elastic reinforcing particle and the plastic matrix. The distribution of fine particles within the grain interior of aluminium matrix can contribute significantly to strengthen the composite by the Orowan mechanism. According to Fig. 5, by comparing the hardness profiles, it can be concluded that by increasing the number of FSP passes, the fluctuations of hardness values in composite specimens reduce and hardness values increase, which is due to the better distribution of the Al_3Ni compounds confirmed by Fig. 3c and f. Finer intermetallic compounds can have more effective role in the pinning of grain boundaries and enhance the hardness, based on Hall–Petch rule. Furthermore, altering the tool rotation, frequently changes advancing and retreating sides of material through the processing track and hence improving material mixing, which enhances hardness uniformity.

Figure 7 shows the hardness values versus pseudo-heat index for composites produced by FSP process. As it is shown in Fig. 7, with increasing the pseudo-heat index, the microhardness is increased, too, which can be related to the formation of more intermetallic compounds. As stated before, another factor that affected hardness is improving particle distribution by increasing FSP pass number, which is confirmed by SEM observation (Fig. 3). The highest microhardness value (47 $HV_{0.05}$) that achieved by six passes of FSP, is 162 % higher than that of the base metal (29 $HV_{0.05}$). Similar hardness value is also reported for $Al+Al_3Ni$ alloy, elsewhere [18].

5 Conclusions

In this study $Al–Al_3Ni$ composite layer was successfully fabricated in situ via FSP of commercial pure aluminium plates preplaced with Ni powder. The followings were concluded:

1. FSP resulted in severe plastic deformation together with heat generation in the treated layer resulting in reaction between Ni powders and aluminium causing in situ formation of Al_3Ni.
2. Increasing the rotational speed or decreasing the traverse speed of the FSP tool improved distribution of Al_3Ni compounds in aluminium.
3. Increasing the number of FSP passes and changing the direction of tool rotation after each pass enhanced the amount and uniformed distribution of Al_3Ni compounds in the treated layer.
4. Hardness increased significantly by increasing the number of FSP passes due to their refining effect and uniformity enhancement of Al_3Ni particles in the treated layer.
5. The average hardness of the composites after six passes of FSP (2,000 rpm, 12 mm/min) was 47 Vickers that was more than 60 % higher than that of the based aluminium.

Acknowledgment The authors appreciate the support from University of Tehran under the research scheme no. 8107024/1/01.

References

1. Wang W, Shi Q, Liu P, Li HK, Li T (2009) A novel way to produce bulk SiCp reinforced aluminum metal matrix composites by friction stir processing. Mater Process Technol 209:2099–2103
2. Hodder KJ, Izadi H, McDonald G, Gerlich P (2012) Fabrication of aluminum–alumina metal matrix composites via cold gas dynamic spraying at low pressure followed by friction stir processing. Mater Sci Eng A 556:114–121
3. Ma ZY (2008) Friction stir processing technology: a review. Metall Mater Trans A 39:642–658
4. Mishra RS, Ma ZY (2005) Friction stir welding and processing. Mater Sci Eng R 50:1–78
5. Mazaheri Y, Karimzadeh F, Enayati MH (2011) A novel technique for development of A356/Al2O3 surface nanocomposite by friction stir processing. Mater Process Technol 211:1614–1619
6. Mishra RS, Ma ZY, Charit I (2003) Friction stir processing: a novel technique for fabrication of surface composite. Mater Sci Eng A 341:307–310
7. Dixit M, Newkirk JW, Mishra RS (2007) Properties of friction stir-processed Al 1100–NiTi composite. Scr Mater 56:541–544
8. Asadi P, Faraji G, Besharati Givi MK (2010) Producing of AZ91/SiC composites by friction sir processing (FSP). Int J Adv Manuf Technol 51:247–260
9. Huang JC, Chuang CH, Hsieh PJ (2005) Using friction stir processing to fabricate MgAlZn intermetallic alloys. Scripta Mater 53:1455–1460
10. Hsu CJ, Kao PW, Ho NJ (2005) Ultrafine-grained Al–Al2Cu composite produced in situ by friction stir processing. Scripta Mater 53:341–345

11. Hsu CJ, Chang CY, Kao PW, Ho NJ, Chang CP (2006) Al–Al3Ti nanocomposites produced in situ by friction stir processing. Acta Mater 54:5241–5249
12. Lee IS, Kao PW, Ho NJ (2008) Microstructure and mechanical properties of Al–Fe in situ nanocomposite produced by friction stir processing. Intermetallics 16:1104–1108
13. Chen HL, Doernberg E, Svoboda P, Schmid-Fetzer R (2011) Thermodynamics of the Al3Ni phase and revision of the Al–Ni system. Thermochim Acta 512:189–195
14. Azizieh M, Kokabi AH, Abachi P (2011) Effect of rotational speed and probe profile on microstructure and hardness of AZ31/Al2O3 nanocomposites fabricated by friction stir processing. Mater Des 32: 2034–2041
15. Barmouz MM, Givi MKB (2011) Fabrication of in situ Cu/SiC composites using multi-pass friction stir processing: Evaluation of microstructural, porosity, mechanical and electrical behavior. Compos Part A : Applies Sci Manuf 42:1445–1453
16. Gan YX, Solomon D, Reinbolt M (2010) Friction stir processing of particle reinforced composite materials. Materials 3:329–350
17. Shafiei-Zarghani A, Kashani-Bozorg SF, Zarei-Hanzaki A (2009) Microstructures and mechanical properties of Al/Al2O3 surface nano-composite layer produced by friction stir processing. Mater Sci Eng A 500:84–91
18. Compton DN, Cornish LA, Witcomb MJ (2001) The effect of microstructure on hardness measurements in aluminium-rich corner of the Al–Ni–Cr system. J Alloy Compd 318:372–378