Dependency of annealing behaviour on grain size in Al–TiC composite produced by accumulative roll bonding

HAMID REZA JAFARIAN* and JAFAR HABIBI-LIVAR

School of Metallurgy and Materials Engineering, Iran University of Science and Technology (IUST), Tehran 16846-13114, Iran

*Author for correspondence (jafarian@iust.ac.ir)

MS received 2 March 2016; accepted 2 September 2016; published online 9 June 2017

Abstract. This work investigates the effect of grain size on annealing behaviour in both coarse-grained and ultrafine-grained Al–TiC composite processed by accumulative roll bonding (ARB). Microstructural analysis indicates that annealing behaviour of the specimens are essentially determined by the level of strain accumulation or number of ARB cycles, i.e., recrystallization phenomenon are accelerated by increasing the level of strain accumulation or number of ARB cycles. Microstructure analysis illustrates that annealing treatment at 200 or 250°C for the 1-cycle ARB-processed Al–TiC composite having coarse grains does not lead to the recrystallization phenomenon, indicating that only recovery appears. In contrast, partial recrystallization occurred after annealing at 250°C in the 7-cycle ARB-processed Al–TiC composite having ultrafine grains. Furthermore, annealing treatment significantly enhanced elongation for both ultrafine as well as coarse-grained Al–TiC composites in spite of the fact that yield and ultimate strength decreased during annealing treatment. The results proved that yield strength and tensile strengths for both 1-cycle and 7-cycle ARB-processed Al–TiC composites gradually decreased by annealing treatments between 200 and 300°C. In contrast, ultimate elongation drastically improved by about 100% after annealing at the aforementioned conditions.

Keywords. Metal–matrix composites; microstructure; mechanical properties; electron microscopy; accumulative roll bonding; annealing.

1. Introduction

Metal–matrix composites (MMCs) have been developed by many materials scientists owing to attractive properties such as high specific strength, stiffness and better wear resistance compared with conventional metals or alloys [1–7] since the extensive demand for high-performance and high strength to weight ratio materials for aerospace applications has rapidly grown up; among all the MMCs there is a specific attention on aluminium MMC [4,5,8,9].

Recently, there have been great attempts to fabricate ultrafine grain (UFG) MMCs [10–15]. It has been well established that ultrafine-grained materials exhibit excellent mechanical properties [16–20]. One of the promising methods to produce ultrafine-grained materials is severe plastic deformation processes such as equal channel angular pressure (ECAP) [21], high-pressure torsion (HPT) [22] and accumulative roll bonding (ARB) [23]. Since the ARB process has many advantages compared with other severe plastic deformation techniques, it has been efficiently used for fabrication of ultrafine-grained MMCs [13–15,24–28]. Recently, Jafarian *et al* [29] indicated that the ARB process can be employed to produce Al/TiC MMC successfully.

Poor elongation is one of the major limitation of ultrafine-grained materials produced by severe plastic deformation

methods [30,31]. Therefore, optimizing strength and ductility is an important issue, and subsequent annealing treatment after the ARB process is one of the promising ways to overcome this limitation. However, due to the high energy stored in ultrafine-grained materials during severe deformation process and possible formation of heterogeneous microstructure, control of annealing conditions with respect to level of strain is crucial to avoid undesirable microstructures. The aim of the present study is to clarify annealing and recrystallization behaviour for both the conventionally deformed Al–TiC composite (1-cycle ARB-processed Al–TiC composite) and the severely deformed Al–TiC composite (7-cycle ARB-processed specimen) with respect to microstructure evolution and mechanical properties.

2. Materials and methods

Commercial purity 1050 aluminium sheets with initial dimensions of 200 mm in length, 400 mm in width and 0.3 mm in thickness were used as the starting material. Table 1 represents the chemical composition and mechanical properties of the sheets. In order to fabricate MMC, nano-sized TiC particles were used as reinforcement particles.

In the present study, the ARB process was used to fabricate ultrafine-grained Al–TiC composite. A schematic illustration

Table 1. Chemical composition of grade 1050 Al (wt%).

Elements	Al	Si	Mn	Cu	Zn	Fe
Composition (wt%)	Bal.	0.25	0.02	0.05	0.05	0.3

of the ARB process is shown in figure 1. In the first step, two sides of five sheets were degreased using acetone to eliminate surface contaminations and then each side of the sheets were wire brushed to provide rough surfaces. The TiC particles were uniformly dispersed on the surface of the scratched brushed sheets. The amount of dispersed TiC particles on each sheet was 2 wt%. Later five sheets were stacked to achieve a 1.5 mm stack thickness and fastened at every edge using a copper wire to make it ready for the roll-bonding. There was one layer of powder between every two sheets of aluminium in the prepared stack. In order to achieve an appropriate bonding between strips, total reduction of the 1-cycle ARB was 60% at room temperature. Then the roll-bonded strap was cut in half and each side cleaned by acetone and wire brushed again. This process was repeated up to 7 cycles with 50% reduction in every cycle at room temperature, so that the total equivalent accumulated strain was 5.6. The roll-bonding experiment was carried out using a laboratory rolling mill with a roll diameter of 230 mm and rolling speed of 10 revolutions per minute (rpm) under dry condition. The ARB-processed specimens were annealed at 200, 250, 300 and 400°C for 30 min to obtain UFG specimens with partly recrystallized or fully recrystallized microstructures.

The microstructure of the ARB-processed specimens was characterized by scanning electron microscopy (SEM) using a field-emission type gun (Philips XL30S-FEG) equipped with an electron backscatter diffraction (EBSD) system operated at 15 kV. EBSD measurements analysis was carried out with a step size of 0.05 μm , which was much smaller than the interval of grain boundaries. The cross-section of the specimen for

EBSD observation was polished mechanically and then electrolytically in a 300 ml HNO_3 + 700 ml $\text{C}_2\text{H}_5\text{OH}$ solution at an approximate temperature of -35°C with a voltage of 20 V. The obtained EBSD data were analysed by TSL-OIM Analysis software. All the microstructures were observed from the transverse direction (TD) of the sheets.

Tensile specimens with gauge length of 25 mm, gauge width of 5 mm and thickness of 1 mm were prepared by spark wire-cutting machine with the tensile axis aligned parallel to rolling direction (RD) of the sheet. Mechanical properties were evaluated by uniaxial tensile machine at a strain rate of $8.3 \times 10^{-4} \text{ s}^{-1}$. The each reported result was the average of three test results.

3. Results and discussion

Figure 2 illustrates a SEM micrograph from TD section of the 7-cycle ARB-processed specimen. It exhibits that after 7 cycles of the ARB process, TiC particles are homogenously distributed in the matrix.

Grain boundary maps of the 1-cycle and 7-cycle ARB-processed specimens obtained from EBSD measurement and analysis are shown in figure 3. The high-angle boundaries having misorientation larger than 15° are drawn in black lines and low-angle boundaries having misorientation between 2 and 15° are drawn in red lines. In the present study, two kinds of ARB-processed specimens are subjected to study: 1-cycle ARB-processed specimen (which is equivalent to conventional rolling having coarse grains) and 7-cycle ARB-processed specimen (which is equivalent to severely deformed sample having UFGs). As indicated in figure 3a, the microstructure after 1-cycle ARB process consists of pancake shape grains and it also has very large amount of low-angle boundaries that are surrounded by high-angle boundaries. These low-angle boundaries can be attributed to the traces

Figure 1. Schematic illustration of accumulative roll bonding (ARB) process for fabrication of particle-reinforced composite.

Figure 2. Scanning electron microscope (SEM) micrographs of the 7-cycle ARB-processed Al–TiC composite indicating homogeneous distribution of TiC particles in the matrix.

of dislocations inherited by 1-cycle ARB deformation. In contrast, the microstructure of the 7-cycle ARB-processed specimen reaches the UFG regime having the mean grain size of 200 nm. Comparing the microstructure of 1-cycle ARB-processed specimen with 7-cycle ARB-processed specimen indicates that a large fraction of low-angle boundaries turned into high-angle ones as shown in figure 3. Hence, it can be said that the ARB process can be used to fabricate Al–TiC composite having ultrafine and elongated grains. It has been well indicated that the main mechanism for grain refinement during the ARB process is grain subdivision [32]. In addition, Jafarian *et al* [29] also indicated that addition of second-phase particles (TiC particles) significantly accelerates the grain refinement phenomenon due to the fact that the TiC particles are less deformable than the Al matrix, resulting in acceleration of dislocation generation or grain subdivision [29]. It should be noted the ARB process can also act as a dispensing agent of TiC particles. Hence, by increasing the number of ARB cycles, more homogenous microstructure can be obtained as seen in figure 2b. Comparing microstructures of the 1-cycle and 7-cycle ARB-processed specimens shows that elongated coarse grains with average grain size of 10 μm obtained by 1-cycle ARB process (equivalent to 50% rolling or conventional rolling) transform to more homogenous UFG microstructure having semi-equiaxed morphology with average grain size of 200 nm.

The microstructures of the 1-cycle and 7-cycle ARB-processed composites subjected to annealing treatment at 200, 250, 300 and 400°C for 30 min are shown in

figure 4. The microstructure of the 1-cycle ARB-processed composite after annealing treatment at 200°C exhibits elongated grains with many low-angle boundaries, indicates that annealing treatment at 200°C does not considerably influence the microstructure. By increasing the annealing temperature to 250°C, the fraction of low-angle boundaries significantly decreased; however, the morphology of grains remained its pancake elongated shape indicating that recovery has taken place by annealing at 250°C without any trace of recrystallization, as shown in figure 4b. The 1-cycle ARB-processed specimen after annealing at 300°C has partially recrystallized microstructure having both equiaxed and elongated grains, as shown in figure 4c. Furthermore, it should be noted that increasing the annealing temperature to 400°C results in enormous grain growth as indicated in figure 4d.

The effects of annealing treatments at 200, 250, 300 and 400°C on microstructure of the 7-cycle ARB-processed specimen are shown in figure 4e–h. It can be observed that annealing at 200°C causes a significant recovery, resulting in annihilation of a large amount of low-angle boundaries, as shown in figure 4d. The microstructure after annealing at 250°C is illustrated in figure 4e. As can be seen, the microstructure is almost recrystallized, with equiaxed morphology. By increasing the annealing treatment to 300 and 400°C, grain growth appeared in the microstructure and relatively coarse grains are obtained as illustrated in figure 4h. Comparing the microstructures after annealing treatments for the 1-cycle and 7-cycle ARB-processed specimens shows that the conventionally deformed specimen (the 1-cycle ARB-processed specimen) and severely deformed specimen (the 7-cycle ARB-processed specimen) clearly exhibited different recrystallization modes. As mentioned, the 1-cycle ARB-processed specimen did not illustrate any kind of recrystallization up to 300°C. In contrast, the 7-cycle ARB-processed specimen exhibited fully recrystallized microstructure after annealing at 250°C. This clearly shows that the severe plastic deformation can accelerate the recrystallization phenomenon by decreasing the recrystallization temperature owing to high stored energy inherited by the 7-cycle ARB process. Our results are in good agreement with the previous studies [19,20,33,34], which indicated that severe plastic deformation can accelerate recrystallization phenomenon.

From another point of view, comparing the recrystallization behaviour in severely deformed pure Al [34] and Al–TiC composite clearly demonstrates that addition of second phase reinforcement particles can increase the recrystallization temperature. This result is consistent with the result by Humphreys [35] and Rollett *et al* [36], who reported that the second-phase particles can act as a barrier for grain growth owing to the fact that they can act as pinning centres for grain boundaries migration, resulting in control of grain growth.

Previous studies concerning recrystallization behaviour of Al alloys indicated that recrystallization essentially depends on purity of the alloy, as grain boundary mobility is really sensitive to impurity in Al alloys [37]. Furthermore, Al

Figure 3. Grain boundary map of the (a) 1-cycle and (b) 7-cycle ARB-processed Al-TiC composite.

alloys have high stacking fault energy, and this hampers the dissociation of dislocations, resulting in promotion of recovery or recrystallization. Recrystallization behaviour of high-purity Al and commercially pure Al fabricated by the ARB process has been well studied by Chekhonin *et al* [38]. They found that a UFG microstructure is developed in the commercially pure aluminium, while partial discontinuous recrystallization occurs in the high-purity layers.

Change in grain size for 1-cycle and 7-cycle ARB-processed composites as a function of annealing treatment temperature is shown in figure 5. The interception method was used to measure the grain size (or interval of boundaries) along the normal direction of the sample. Here, both high-angle boundaries and low-angle boundaries were taken into account for the grain size measurement. As indicated, the grain size of the 1-cycle ARB-processed specimen increased

from 15 to 30 μm on increasing the annealing temperature with a low steep rate, but the grain size of the 7-cycle ARB-processed specimen increased from 0.2 to 35 μm with a fast steep compared with the 1-cycle ARB-processed specimen. However, it should be noted that the 7-cycle ARB-processed specimen has relatively larger grain size after annealing at 400°C compared with the 1-cycle ARB-processed specimen. This can be attributed to lower recrystallization temperature of the 7-cycle ARB-processed specimen, as already discussed.

Engineering stress-strain curves of the 1-cycle and 7-cycle ARB-processed specimens and then annealed at 200, 250 and 300°C are shown in figure 6. As seen, it can be said that in both the 1-cycle and 7-cycle ARB-processed specimens, the yield and ultimate tensile strengths are decreased by annealing treatment and the ultimate elongation is increased significantly. Changes in yield and ultimate tensile strengths as well as ultimate elongation obtained from the

Figure 4. Grain boundary map of the 1-cycle ARB-processed Al–TiC composite annealed at (a) 200, (b) 250, (c) 300 and (d) 400°C. Grain boundary map of the 7-cycle ARB-processed Al–TiC composite annealed at (e) 200, (f) 250, (g) 300 and (h) 400°C.

engineering stress–strain curves for the 1-cycle and 7-cycle ARB-processed specimens are shown in figure 7. As indicated, yield strength for the 1-cycle ARB-processed specimen declined from 178 to 113 MPa by annealing treatments at

temperatures between 200 and 300°C. In contrast, the ultimate elongation increased from 4 to 8.2% after the same aforementioned annealing treatments. The results indicate that the 7-cycle ARB-processed specimen follows the same

Figure 5. Change in matrix grain size (boundary interval along normal direction) as a function of annealing temperature.

tendency of changes in yield and ultimate tensile strengths as that of the 1-cycle ARB-processed specimen. However, it can be observed that yield and ultimate strength decrease by

annealing is more accelerated in the 7-cycle ARB-processed specimen, which can be attributed to the quick recrystallization in the 7-cycle ARB-processed specimen, as shown in figure 4e–h.

The results from the tensile tests also indicated that in both 1-cycle and 7-cycle ARB-processed specimens, drastic decrease in yield and ultimate strengths appeared after annealing at 300°C compared with annealing at 200 and 250°C. This can be attributed to significant grain growth by annealing at 300°C, as shown in figure 4. As mentioned, in 1-cycle ARB-processed specimen, there is no trace of recrystallization phenomenon, indicating that only recovery takes place on annealing at 200 and 250°C and the smooth decrease in strengths come from decrease in dislocation density by recovery. However, fully recrystallized microstructure could be obtained by annealing at 300°C. As a result, a drastic drop in yield and tensile strengths appeared after annealing at 300°C.

It is well known that the stress–strain curve follows Holloman equation $\sigma = k\varepsilon^n$, where σ is true stress, ε is true strain and n is strain hardening rate [39]. In order to study the effect of the number of ARB cycles and subsequent

Figure 6. (a) Engineering stress–strain curves of the 1-cycle ARB-processed Al–TiC composite annealed at 200, 250 and 300°C. (b) Engineering stress–strain curves of the 7-cycle ARB-processed Al–TiC composite annealed at 200, 250 and 300°C.

Figure 7. Relationship between annealing temperature and yield strength and total elongation for the (a) 1-cycle and (b) 7-cycle ARB-processed Al–TiC composites.

Figure 8. Change in strain hardening rate value in specimens subjected to 1-cycle and 7-cycle ARB process and subsequent annealing.

annealing on strain hardening rate, n -values for 1-cycle and 7-cycle ARB-processed and subsequently annealed specimens are calculated and shown in figure 8. It is clear that the n -value for the 1-cycle ARB-processed specimen is relatively smaller than that of the 7-cycle ARB-processed one. This is consistent with early plastic instability on increasing the level of strain accumulation or ARB cycles that has been reported before [23,34]. In addition, annealing treatment results in enhancement of n -value due to recovery and decrease in dislocation density for both 1-cycle and 7-cycle ARB-processed specimens as indicated in figure 4.

4. Conclusion

In this work, the effect of annealing treatment on microstructure and mechanical properties was investigated in both conventionally and severely deformed Al–TiC composite fabricated by 1-cycle ARB process having coarse grains and 7-cycle ARB process having UFGs. The main conclusions drawn from the present study can be summarized as follows:

- i. Microstructure analysis illustrated that after annealing treatment at 200 and 250°C for the 1-cycle ARB-processed Al–TiC composite, the specimens do not show the recrystallization phenomenon, indicating that only recovery appeared. In contrast, partial recrystallization happened after annealing at 250°C in the 7-cycle ARB-processed Al–TiC composite. This indicates that recrystallization is essentially determined by level of accumulative strain.

- ii. The yield strength and tensile strength for both 1-cycle and 7-cycle ARB-processed Al–TiC composites gradually decreased by annealing treatments between 200 and 300°C. In contrast, ultimate elongation drastically improved by about 100% after annealing at the aforementioned conditions. Decrease of the strengths can be attributed to the recovery and recrystallization during annealing treatments.

Acknowledgements

This study was financially supported by the Grant-in-Aid through Iran National Science Foundation (INSF), Iran (Contract No. 91002129), and the support is gratefully appreciated. We are also really thankful to Prof. Nobuhiro Tsuji, Kyoto University, for fruitful discussion and providing EBSD analysis equipment.

References

- [1] Ibrahim I, Mohamed F and Lavernia E 1991 *J. Mater. Sci.* **26** 1137
- [2] Mortensen A 1991 *Mater. Sci. Eng. A* **135** 1
- [3] Banks-Sills L, Leiderman V and Fang D 1997 *Composites Part B: Eng.* **28** 465
- [4] Karantzalis A, Wyatt S and Kennedy A 1997 *Mater. Sci. Eng. A* **237** 200
- [5] Manoharan M and Gupta M 1999 *Composites Part B: Eng.* **30** 107
- [6] Cantor B, Dunne F P and Stone I C 2010 *Metal and ceramic matrix composites* (FL, USA: CRC Press)

- [7] Yazdani A, Salahinejad E, Moradgholi J and Hosseini M 2011 *J. Alloys Compd.* **509** 9562
- [8] Nair S, Tien J and Bates R 1985 *Int. Met. Rev.* **30** 275
- [9] Turnbull A 1992 *Br. Corros. J.* **27** 27
- [10] Alizadeh M and Paydar M 2010 *J. Alloys Compd.* **492** 231
- [11] Salimi S, Izadi H and Gerlich A 2011 *J. Mater. Sci.* **46** 409
- [12] Alizadeh M, Paydar M, Terada D and Tsuji N 2012 *Mater. Sci. Eng. A* **540** 13
- [13] Alizadeh M, Paydar M and Sharifian Jazi F 2013 *Composites Part B: Eng.* **44** 339
- [14] Alizadeh M, Ghaffari M and Amini R 2014 *Composites Part B: Eng.* **58** 438
- [15] Reihanian M, Hadadian F K and Paydar M 2014 *Mater. Sci. Eng. A* **607** 188
- [16] Valiev R Z, Korznikov A and Mulyukov R 1993 *Mater. Sci. Eng. A* **168** 141
- [17] Valiev R, Korznikov A and Mulyukov R 1993 *Mater. Sci. Eng. A* **168** 141
- [18] Valiev R Z 1997 *Mater. Sci. Eng. A* **234** 59
- [19] Tsuji N, Ito Y, Saito Y and Minamino Y 2002 *Scr. Mater.* **47** 893
- [20] Jafarian H and Eivani A 2014 *J. Mater. Sci.* **49** 6570
- [21] Iwahashi Y, Wang J, Horita Z, Nemoto M and Langdon T G 1996 *Scr. Mater.* **35** 143
- [22] Zhilyaev A P and Langdon T G 2008 *Prog. Mater. Sci.* **53** 893
- [23] Saito Y, Utsunomiya H, Tsuji N and Sakai T 1999 *Acta Mater.* **47** 579
- [24] Rezayat M, Akbarzadeh A and Owhadi A 2012 *Metall. Mater. Trans. A* **43** 2085
- [25] Jamaati R, Toroghinejad M R, Dutkiewicz J and Szpunar J A 2012 *Mater. Des.* **35** 37
- [26] Dehkordi H F, Toroghinejad M R and Raeissi K 2013 *Mater. Sci. Eng. A: Struct. Mater.: Properties Microstruct. Process.* **585** 460
- [27] Jamaati R, Naseri M and Toroghinejad M R 2014 *Mater. Des.* **59** 540
- [28] Mehr V Y, Rezaeian A and Toroghinejad M R 2015 *Mater. Des.*
- [29] Jafarian H, Habibi-Livar J and Razavi S H 2015 *Composites Part B: Eng.* **77** 84
- [30] Ma E 2003 *Scr. Mater.* **49** 663
- [31] Koch C 2003 *Scr. Mater.* **49** 657
- [32] Hughes D and Hansen N 1997 *Acta Mater.* **45** 3871
- [33] Park K-T, Kwon H-J, Kim W-J and Kim Y S 2001 *Mater. Sci. Eng. A* **316** 145
- [34] Kamikawa N, Tsuji N, Huang X and Hansen N 2006 *Acta Mater.* **54** 3055
- [35] Humphreys F 1977 *Acta Metall.* **25** 1323
- [36] Rollett A, Humphreys F, Rohrer G S and Hatherly M 2004 *Recrystallization and related annealing phenomena* (Amsterdam, The Netherlands: Elsevier)
- [37] Kassner M E, McQueen H J, Pollard J, Evangelista E and Cerri E 1994 *Scr. Metall. Mater.* **31** 1331
- [38] Chekhonin P *et al* 2012 *Acta Mater.* **60** 4661
- [39] Dieter G E 2015 *Mechanical metallurgy* 3rd edn (New York: McGraw-Hill)