Original Article

Enhanced strength and ductility of Al-SiC nanocomposites synthesized by accumulative roll bonding

Adnan I. Khdair a,b,⁎, A. Fathy c

a Mechanical Engineering Department, Faculty of Engineering, King Abdulaziz University, P.O. Box 80204, Jeddah, Saudi Arabia
b Jordan University of Science and Technology, Mech. Eng. Dept., P.O. Box 3030, Irbid 2011, Jordan
c Department of Mechanical Design and Production Engineering, Faculty of Engineering, Zagazig University, P.O. Box 44519, Zagazig, Egypt

ARTICLE INFO

Article history:
Received 19 September 2019
Accepted 30 October 2019
Available online 14 November 2019

Keywords:
Al-SiC nanocomposites
Accumulative roll bonding (ARB)
Microstructure
Microhardness
Tensile test
Fracture shape

ABSTRACT

This paper presents experimental study on improving mechanical properties of Al and Al-xSiC nanocomposites (x=1, 2 and 4%) synthesized by Accumulative Roll Bonding (ARB) technique. SEM, EDX and XRD analysis was used to characterize the structural changes in the manufactured materials while tensile and microhardness tests were used to characterize their mechanical properties. Homogeneous distribution of SiC nanoparticles was achieved after five ARB cycles. A significant strength improvement was achieved for processed Al and Al-SiC nanocomposites after five ARB cycles due to grain refinement, grain misorientation and SiC strengthening. The grain refinement and grain misorientation contributed the strength improvement by 161% and 46%, respectively, while the addition of 1% SiC nanoparticles contributed by 78% which is increased to 101% for 4% SiC. The ductility is reduced after the zero ARB cycle however it increased with increasing the number of ARB cycles reaching 8.2 and 5.8% for Al and Al-4% SiC nanocomposites, respectively, after 5 ARB cycles. The fracture shape in the ARBed samples is a combination of necking and shearing with a tendency to shearing shape for Al-SiC nanocomposites.

© 2019 The Author. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

1. Introduction

Recently, the application of metal matrix composites (MMCs) in aerospace, military, and car enterprises has grown due to their particular properties, for example, the high strength to density ratio, high elastic modulus, wear resistance, and thermal conductivity combined with excellent corrosion resistance [1–3]. Al and its alloys sheets are used to build aircraft cabins and stabilizers, car body, solar panels, curtain walls, doors and window frames and many others [4–6]. The low strength of these alloys is the main limitation of their applications. This limitation can be overcome either by changing the structure of grains or reinforcement Al alloy matrix with ceramic particles [7–9]. The combination of metal and ceramic in Al-based composites leads to more
improved properties. Aside from the fabrication strategy, the proper reinforcement is a key parameter for Al-based composites strength improvement [10–19]. In conventional MMCs, ceramic particles like Al2O3, B4C and SiC, have been generally utilized as particulate reinforcements [20–23]. Among these reinforcements, SiC particles achieved reasonable wettability with Al matrix during manufacturing process [20,21].

Several techniques have been presented in the literature to manufacture Al-based MMCs such as powder metallurgy and casting [24,25]. Despite their economic advantage, these methods have some drawbacks such as the low density of the produced composite due to the high percentage of porosity, reaction between the reinforcements and the Al matrix which result in undesired intermetallic phases, and difficulty to attain uniform conveyance of reinforcement particles [26]. As a consequence, the mechanical, thermal and electrical properties of the composites may be decreased. To overcome these restrictions, Accumulative Roll Bonding (ARB) has been developed to manufacture Al-based MMCs with improved properties. This technique own other advantage which is the significant reduction of Al grain size reaching ultrafine grains which highly improve the mechanical properties [27]. Therefore, ARB is considered as the unique solid-state process, which can be utilized for particle-reinforced metal matrix composites manufacturing with highly improved mechanical properties. In this method, reinforcement particles are added between Al strips during initial ARB cycle, after that the produced sheets are cut and stacked together and rolled. Following these procedure, uniform distribution of the particles will be achieved after certain number of ARB cycles [28,29]. Owing to its ability to reduce the grain size, it is considered as one of the severe plastic deformation methods for production of ultrafine grained and nanostructured metallic materials [30–33]. So far, this technique has been extensively applied for improvement of the strength of different metals and their alloys through grain refinement [12,34,35]. Also, enormous studies have been conducted on the manufacturing of MMCs by adding ceramic particles between metallic sheets before rolling [36–38]. It has been reported that the mechanical properties of MMCs can be improved using ARB method but considerable decrease in elongation and ductility is observed [39].

Nowadays, MMCs produced by ARB techniques attract researchers’ attentions due to the improvement of mechanical properties. For example, Yazdani et al. [40] applied this technique to produce Al-B4C composites. Their results showed a significant improvement is achieved when B4C particles are homogeneously distributed in Al matrix. Similar observations and conclusions are reached by Alizadeh et al. [41] which highlights a large dependence of mechanical properties of MMCs on the dispersion of the ceramic phase in the matrix. They utilized SiC micro-particles as reinforcement for fabrication of Al–SiC composites. They concluded that the tensile strength is increased by adding SiC particles to Al matrix.

More recently, ARB has been applied to fabricate distinctive Al-based MMCs such as: Al/SiC [42–45], Al/Al2O3 [46,47], Al/B4C [40], Al/CNT [48], Al/TO2 [49], Al/SiO2 [50], Al/W [51,52], Al/WC [53]. It has been proved that a critical reduction to the sample thickness, reduction about 50% or more in each ARB cycle, is a condition for achieving a uniform distribution of particles [54]. Moreover, some other attempts have been conducted to manufacture hybrid composites using ARB technique such as Al/Al2O3/SiC [55], Al/Al2O3/B4C [56], Al/B4C/SiC [57].

The present work focuses on the manufacturing of Al-SiC metal matrix nanocomposite using ARB technique. Three SiC volume fraction, 1, 2 and 4 vol.%, are considered and also a sample without SiC particle is manufactured with the aim of comparison. To the author best knowledge, for the first time, interplay between micro/nano-structure of the prepared samples after each ARB cycle and tensile strength, elongation, hardness and fracture shape is deeply discussed. Additionally, the different strengthening mechanisms occurs during ARB process are presented with a quantification of the contribution of each mechanism.

2. Material and experiments

Al1050 aluminum sheets and SiC particles (5 μm average particle size and 99.9% purity) were used as starting materials to prepare ultrafine grained Al and Al-SiC nanocomposites. Aluminum sheets with the dimensions of 100 mm × 50 mm × 1 mm were heated to 450 °C for 1 h in hydrogen atmosphere to eliminate the cold worked microstructure of plates just before ARB process. Before ARB process, acetone was used to degrease Al sheets to remove any contaminants over its surface. Then, a 90 mm diameter stainless steel circumferential brush with 0.3 mm wire diameter was used to scratch Al sheets at peripheral speed of 2500 rpm to achieve a good bonding. Afterward, the surfaces were cleaned by acetone again and air dried. The ARB process for manufacturing of the Al-1, 2, 4% vol. SiC composite was schematically reported in [31,32].

The ARB process comprised of two steps as appeared in Fig. 1. In the first step, after surface preparation, SiC particles were dispersed between the two Al sheets and fixed at their ends by wires. Dispersion of particles was done by using a sieve of 10 μm mesh, where firstly the SiC particles were weighed and then were poured into the sieve located on the sheets surface. The sieve was shaken and the SiC particles were poured on the sheets surface accordingly. The wire brushed surface helps to keep the reinforcement in place while rolling. The ARB process was performed using a laboratory rolling mill with 350 mm roll diameter and speed of 0.366 m/s. No lubrication was used during ARB process. At the zero cycle, the thickness reduction was 66.7% at room temperature. After that the roll bonded sheets were cut into two parts using a shearing machine. Annealing at 400 °C for 1 h for the cut sheets was applied to reduce the residual thermal stresses and improve Al sheet bonding. Finally, scratch brushing, degreasing in acetone and stacking was applied for the annealed sheets and roll bonded with 50% reduction. This procedure, cut, annealing, scratch brushing, degreasing and stacking, was repeated up to five cycles. For the production of the fine-grained Al sheets, the same procedure was applied without addition of the SiC particles between the first Al sheets. It is valuable mentioning that after each ARB cycle, the edges of the sheets were trimmed off to avoid the progression of the edge cracks.

The microstructure of the prepared samples after each ARB cycle was characterized using Field Emission Scanning
During the first ARB cycles, grain elongation on the rolling direction occurs due to the rolling strain and squeezing of the material under rollers entrapping SiC particles between the elongated grains. SiC particles are found agglomerated at the early stages of ARB as shown in Fig. 2(a) and (c). Increasing the rolling strain, number of ARB cycles, the elongated grains are fractured to low angle subgrains and SiC particles are fractured to submicron size (see Fig. 2) due to its brittleness. The fractured SiC particles penetrates Al grains causing structure defects such as point defects and dislocations which increase strain hardening of the subgrains. Further ARB cycles, after five cycles, the fractured Al subgrains are deformed with large misorientation between subgrains resulting in high angle subgrains [58,59]. Additionally, owing to the high deformability of Al and the reduction of SiC particle size, plastic flow of Al matrix facilitates the distribution of SiC particles which reduce its agglomeration and hence provide more uniform microstructure as shown in Fig. 2(b) and (d) [22,35]. A higher magnification SEM micrograph of Al-4% SiC nanocomposite shows the reduction of SiC particles to nano size after five ARB cycles as shown in Fig. 2(e). The reduction of SiC particles to nano size during the process which can be in situ generated, provide advantages for ARB technique over other techniques at which the initial reinforcement size is in nano size. In such process the reinforcement clustering/agglomeration is difficult to be eliminated especially with large reinforcement volume fractions [60]. Mapping analysis of Al-4% SiC nanocomposite after five cycles is shown in Fig. 3. The figure shows the excellent distribution of the elements (Al, Si and C) without observation of any agglomeration in the sample. The EDX analysis in the same figure shows that the composition of the sample which shows that the sample contain only the three elements without observation of other contaminates or undesired elements.

Fig. 4 reveals that the Al and Al-SiC nanocomposites with different SiC content after five ARB cycles consists of only Al and SiC phases. The XRD pattern prove that no evidence of undesired phase such as Al4C3 which has negative effect on the properties of Al-based metal matrix composites [61-63]. This highlights the advantage of solid-state fabrication process over other techniques. The intensity of SiC phase increases with increasing SiC content which is logically expressed due to the higher content of SiC particles. The intensity of Al peaks is reduced with increasing SiC content due to the grain refinement mechanism during ARB process. The width of the peak is almost the same for the three composites. According to Williamson relation [64], this observation reflects negligible influence of SiC addition on the crystallite size and dislocation spacing of the produced nanocomposites. The average crystallite size of Al and Al-SiC nanocomposites containing 1, 2, and 4% SiC are 55, 53, 52 and 50 nm, respectively compared to 102 nm for the as received Al.

TEM microstructure and corresponding selected area diffraction patterns observed at the plane perpendicular to the transverse direction (TD plane) of the ARB-processed Al–SiC composite specimens of the fifth cycles have been shown in Fig. 5. As it can be seen after five ARB cycles, ultrafine grains extend in the Al matrix and also they aligned in rolling
direction. It has been found that the formation mechanism of UFG by ARB is explained in terms of grain subdivision at a submicron scale [65,66], where initial coarse grains are subdivided by deformation-induced high-angle grain boundaries.

3.2. Mechanical properties

Tensile stress–strain curves of Al, Al-SiC nanocomposite with different volume fraction of SiC, 1, 2 and 4% processed at
different ARB cycles are shown in Fig. 6. It is observed that all ARBed samples shows larger tensile strength than the as received Al. The improved tensile strength in the Al sample (see Fig. 6(a)) is attributed to the grain refinement in Al matrix caused by the ARB process. While in the samples contain SiC nanoparticles, two strengthening mechanism contribute the improvement of the tensile properties, grain refinement and the presence of SiC nanoparticles. These particles have higher tensile strength than Al which make them able to withstand higher tensile stresses than Al and hence improves the global strength of the produced nanocomposite. It is also observed that the failure strain is lower for ARBed samples than the as received Al due to the grain refinement and the high angle subgrains which reduce the ability of these grains to deform [67]. Fig. 7 shows the variation of ultimate strength and elongation with the number of ARB cycles for all the tested samples. For Al samples, the tensile strength is increased to 149 MPa after the first ARB cycle achieving 161% strength
improvement. This improvement is basically due to grain refinement and the formation of low angle subgrains [68]. The Al almost reach the saturation of grain subdivision after 2 or 3 ARB cycles [46]. The tensile strength is increased with increasing number of ARB cycles reaching 175 MPa for sample after five ARB cycles achieving 207% strength improvement. The main strengthening mechanism during these cycles is the increase of misorientation between grains and formation of high angle subgrains [67]. Based on these observations, we can demonstrate that the grain subdivision which occurs during the early ARB cycles contributes the major improvement of the strength (161% improvement) while the contribution of the misorientation between grains on the strength improvement is lower. The elongation of ARBed Al is highly reduced after the zero-cycle reaching 3.7% due to the higher elongation of grains in the rolling direction which eliminate the ability of grains to deform and slip on each other. Increasing the number of ARB cycles improves the elongation due to the formation of subgrains which make the slipping between subgrains more possible. Additionally, the stacking faults of Al during ARB process can contribute the elongation improvement [68,69].

When 1% SiC is added to Al, the ultimate strength is larger compared to pure ARBed Al at the same ARB cycle as shown in Fig. 7(b). For example, the tensile strength of Al-1%
SiC nanocomposite after the first ARB cycle is 187 MPa compared to 149 MPa for Al at the same cycle, achieving 25.5% increase. This increase of ultimate strength is attributed to the higher tensile strength of SiC nanoparticle which share the applied stress with Al matrix. It is valuable noting that however agglomeration is observed during the first ARB cycles (see Fig. 2(a)), the ultimate strength is increased. This highlight the minor role of SiC distribution on the strength of the ARBed composites and indicates the major role of grain refinement strengthening mechanism on the strength improvement. This is not the case for other manufacturing techniques such as powder metallurgy and casting at which the distribution of reinforcement particles in the samples plays the major role of strength improvement [1–3,8,25,70] which in some cases is difficult to be achieved [4,20]. Increasing the number of ARB cycles to five, the ultimate strength is increased to 220 MPa compared to 175 MPa for ARBed Al at the same cycle. The presence of SiC particles during ARB process facilitate the grain subdivision mechanism due to the penetration of these particle to Al grains which reduce its plasticity and hence increase its brittleness. So, the grain subdivision occurs faster in Al-SiC composites than Al. Additionally, these particles increase the tendency of subgrains to have high misorientation between them and formation of high angle subgrains and ultrafine grains. The elongation of this composite follows the same trend as observed in Al at which the elongation is highly reduced after the zero cycle and increased with the following number of cycles. The elongation of this composite is lower than the pure Al at the same ARB cycle due to the penetration of SiC nanoparticle to the Al grains which reduce its deformability. Moreover, the presence of SiC nanoparticles on the grain boundaries between Al grains reduces its ability to slip between each other. Similar observations are presented in previous research [46,47]. Increasing SiC content improves the tensile strength of Al-SiC composite reaching 233 MPa for Al-4% SiC after five ARB cycles achieving 308% strength improvement compared to the as received Al (see Table 1). Comparing the tensile strength of the prepared composites in the current study and others available in the literature for the same composites, Al-SiC, as shown in Table 1, we can demonstrate that the ARB technique provides the highest strength improvement among all the other available methods.

![Fig. 7 – Ultimate strength and elongation of the composites produced by the ARB process at various passes. (a) Al, (b) Al-1% vol. SiC, (c) Al-2% vol. SiC and (d) Al-4% vol. SiC.](image)

Table 1 – Ultimate strength of Al-SiC nanocomposite prepared by different techniques in comparison with the manufactured using ARB process in this study. The percentage of improvement is calculated based on the ultimate strength of the as received Al for each case.

<table>
<thead>
<tr>
<th>Material</th>
<th>Preparation method</th>
<th>Ultimate strength (MPa)</th>
<th>Percentage of improvement (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al</td>
<td>Five ARB cycles (current study)</td>
<td>175 ± 13</td>
<td>207</td>
</tr>
<tr>
<td>Al-1%SiC</td>
<td></td>
<td>220 ± 11</td>
<td>285</td>
</tr>
<tr>
<td>Al-2%SiC</td>
<td></td>
<td>229 ± 15</td>
<td>302</td>
</tr>
<tr>
<td>Al-4%SiC</td>
<td></td>
<td>233 ± 21</td>
<td>308</td>
</tr>
<tr>
<td>Al-0.3%SiC</td>
<td>Microwave sintering + extrusion [74]</td>
<td>130</td>
<td>9.2</td>
</tr>
<tr>
<td>Al-0.5%SiC</td>
<td></td>
<td>144</td>
<td>21</td>
</tr>
<tr>
<td>Al-1.5%SiC</td>
<td></td>
<td>178</td>
<td>49.5</td>
</tr>
<tr>
<td>Al-2%SiC</td>
<td>Steering [70]</td>
<td>47</td>
<td>–</td>
</tr>
<tr>
<td>Al-15%SiC</td>
<td>Liquid state mixing [71]</td>
<td>198</td>
<td>32</td>
</tr>
<tr>
<td>Al-25%SiC</td>
<td>Stir casting [72]</td>
<td>88.5</td>
<td>–</td>
</tr>
</tbody>
</table>
Fig. 8 shows the variation of microhardness of Al and Al-SiC nanocomposites with the number of ARB cycles. Generally, increasing the number of ARB cycles improves microhardness of the prepared samples. The hardness increasing rate is large at the first two cycles due to the grain refinement and strain hardening strengthening mechanisms which is in accordance with the ultimate tensile strength results (see Fig. 7). The hardness increases form 32 HV to 48 HV and 65 HV for Al and Al-4%SiC nanocomposite, respectively, after the second ARB cycle. However, from the third pass, the hardness increasing rate is very low which can be neglected in the case of Al-SiC nanocomposites. It increases form 48 HV and 65 HV after the second cycle to 53 HV and 67 HV after the fifth cycle For Al and Al-4%SiC, respectively. This is since grain refinement mechanism occurs at the first two cycles and the grain subdivision mechanism reaches the saturation [46]. It is also observed from Fig. 8 that at the early ARB cycles, 0, 1, 2 and 3, the hardness is increased with increasing SiC content. This is due to the role of SiC nanoparticles in facilitating the grain refinement and dislocation strengthening of the composites during the first ARB cycles. The hardness is increased also at the fifth ARB cycle with addition of 1% SiC, however increasing the SiC content to 2% and 4% has insignificant influence on the hardness. This can be attributed to the grain refinement saturation of Al-SiC nanocomposite with addition of 1% SiC which make the addition of more SiC nanoparticles to the composite insignificantly affect the hardness.

Based on the presented results and discussion, we can quantify the role of each of the aforementioned strengthening mechanism, grain subdivision/refinement, grain misorientation and SiC strengthening on the strength improvement of Al and Al-SiC nanocomposites. It is shown that 161% strength improvement is achieved for Al after the second ARB cycle at which the grain refinement is saturated which mean that this improvement is attributed only to the grain refinement strengthening mechanism. After five ARB cycle, the total improvement is 207% which mean that 46% of the strength improvement is attributed to grain misorientation. Addition of 1% SiC results in a total improvement after five ARB cycles of 285% which mean that the addition of 1%SiC participate in the strength improvement by 78%. Increasing the SiC content to 4% improves the strength by 308% which mean that the addition of 4% SiC results in 101% strength improvement. Therefore, we demonstrate that the major strength improvement is attributed to the grain refinement strengthening mechanism which usually occurs at the early ARB cycles of Al.

3.3. Fracture behavior

Fig. 9 shows photos of Al and Al-4%SiC nanocomposite samples after tensile test. It is known that the fracture shape of the ductile materials such as Al is necking fracture shape [73]. However, this is not the case of ARBed Al samples as shown in Fig. 9(a), after the zero-cycle, the fracture shape is combined fracture of necking and shear with angle about 28.5° with the loading axis (see Fig. 9(a) and Fig. 10(a)). As previously stated at this cycle, grain elongation occurs in the rolling direction, which reduce its ability for deformation in that direction and hence the necking behavior is eliminated. To dissipate the external tensile stresses, elongated grains start to slip over each other forming a sort of internal micropores between them. Since the micropores are generated randomly, the elongated grains are supported form one of their sides by its neighbor grain while it is free from the side connected to the micropores which make these grains subjected to combined stresses, shear and tensile stress [75]. The samples prepared after first and second cycle shows almost equal shear angle and slightly larger than the sample after the zero-cycle due to the formation. For the last three cycles, the fracture shear angle is reduced to around 21° due to the formation of high angle grains which dissipate the applied stresses in the fracture.

The fracture shape of Al-4%SiC nanocomposites follows globally the same response as Al with combination of necking and shear. However, the necking action is slightly reduced as shown in Fig. 10(b) and shear angle is slightly larger than Al as shown in Fig. 9(b). This larger shear angle is due to the lower ductility of Al-4%SiC nanocomposites than Al (see Fig. 7) which reduce the tendency of the material for necking and hence the applied stresses are dissipated in shear failure with high angles. Additionally, the presence of SiC nanoparticles between Al grains reduces the slipping action between Al grains and facilitate the creation of cracks at the interface between SiC and Al. It is observed that the fracture surface of Al-4%SiC nanocomposites at the first ARB cycles is hierarchical surface due to the agglomeration of SiC nanoparticles in these composites which make the initiation of cracks much easier at the agglomerated parts than the homogeneously distributed ones. For the sample after the fifth cycle, the fracture surface is almost the same as Al.

During tensile test, dimples appear due to the plastic deformation of the matrix material [76,77]. It is evident from the microstructure of the fracture surface that the depth and the width of dimples is larger for Al as shown in Fig. 10(a) due to its higher ductility after five ARB cycles (see Fig. 7). However, the width and depth of dimples is small for Al-4%SiC nanocomposites due to the presence of SiC nanoparticles at the grain boundaries, at which microcracks occur due to separation at the interface between them.
Fig. 9 – Photos of fractured samples of (a) Al and (b) Al-4%SiC nanocomposite at different ARB cycles.

Fig. 10 – SEM micrograph of the fracture surface of (a) Al and (b) Al-4%SiC nanocomposite after five ARB cycles.
4. Conclusions

In the present work, SiC (1, 2 and 4%) particles reinforced Al5051 matrix nanocomposite were successfully manufactured using accumulated roll bonding technique. SEM, EDX and XRD analysis were used to characterize the morphological and structure changes of the prepared composites during the ARB process. After two ARB cycles, the tensile strength of Al increased to 149 MPa compared to 57 MPa for the as received Al achieving 161% strength improvement. While after five ARB cycles, the tensile strength increased to 175 MPa. Addition of 1%SiC nanoparticles improved the tensile strength to 220 MPa after five cycle. Increasing SiC nanoparticles to 4% increases the tensile strength to 233 MPa achieving 308% strength improvement. Three main strengthening mechanisms govern the strengthening of Al and Al-SiC nanocomposites named as grain refinement, grain misorientation and SiC strengthening. Grain refinement occurs during the early ARB cycles which contribute the strength improvement by 161%, while grain misorientation contribute only 46% on strength improvement. SiC strengthening contribute by 78% of strength improvement of nanocomposite contain 1% SiC while this value is increased to 101% improvement with increasing SiC content to 4%. The elongation of the ARBed samples is smaller than the as received Al due to the work hardening effect caused by grain refinement. Increasing the number of ARB cycle improves the elongation of Al and Al-SiC nanocomposites reaching 8.2 and 5.8% for Al and Al-4% SiC nanocomposites, respectively, after 5 ARB cycles compared to 3.7 and 3.2% for the same materials after the zero-cycle. The hardness of Al and Al-SiC nanocomposites followed more/less the same trend of the strength improvement. The fracture shape of ARBed Al and Al-SiC nanocomposites under tensile stress is a combination of necking and shearing. The shearing angle is reduced by increasing the number of ARB cycles.

Conflicts of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This project was funded by the Deanship of Scientific Research (DSR), King Abdulaziz University, Jeddah, under grant No. (D-099-135-1440). The authors, therefore, gratefully acknowledge the DSR technical and financial support.

References


[57] Naseri M, Hassani A, Tajally M. An alternative method for manufacturing Al/B4C/SiC hybrid composite strips by cross


