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The effect of mechanical alloying on Al_2O_3 distribution and properties of Al_2O_3 particle reinforced Al-MMCs

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Abstract

The properties of particle-reinforced aluminum composites depend on the microstructure and evolved uniformity distribution of particles in the matrix. The distribution of Al_2O_3 particles in the matrix and microstructure properties of varying volume fraction of particles up to 15% Al_2O_3 particle-reinforced Al metal matrix composites produced by the mechanical alloying technique was investigated. Alloying was performed in a planetary ball mill using a milling time varying from 0 h to 7 h, a ball-to-powder ratio of 10:1, and a ball mill velocity of 400 rpm. The produced compositions were cold-pressed at 700 MPa with a single action and sintered at 600°C for 3 h under Argon gas atmosphere. The relative density (both pressed and sintered), porosity and hardness of composites were also examined. The results of mechanical alloying processing were investigated with scanning electron microscopy (SEM), X-ray diffraction and laser particle size analyzer. SEM observations showed that relatively homogeneous distribution of Al_2O_3 reinforcement in the matrix could be obtained by mechanical alloying after 5 h. Moreover, the variation in hardness of the composites with alumina volume fraction and milling time was observed to be a strong function of the work hardening mechanism.

Keywords: aluminum-based metal matrix composites (Al-MMCs); Al_2O_3; mechanical alloying (MA); powder metallurgy.

1. Introduction

Composites are defined as those advanced materials in which a reinforcement phase is dispersed in a matrix phase. The reinforcement can be in the form of a particulate, short fiber, or continuous fiber. There is usually a well-defined interface between the matrix and the reinforcement phases. Composites exhibit properties that are an average of the matrix and reinforcement properties. In metal matrix composites, for example, a ceramic reinforcement is generally dispersed in a metal matrix phase. Typical examples include Al-Al_2O_3, Al-SiC, Ti-SiC, Mg-Al_2O_3, etc. These composite materials combine the ductility of the metal matrix and the high stiffness of the ceramic reinforcement phase [1]. Among the advanced engineering materials for transport applications, Al-based metal matrix composites (MMCs) show the largest potential to reach this goal and to develop novel lightweight high-performance materials due to their remarkable properties, including low density, high strength and good fatigue and wear resistance [2]. MMCs can be formed from solid and molten states into forging, extrusions, sheet and plate and casting. Conventional techniques such as casting, spraying and forging have problems, e.g., reinforcement segregation, unwanted interfacial chemical reactions, higher porosity and poor interfacial bonding. Owing to the presence of alumina particles with high melting point, conventional melting and casting is not suitable for producing dispersion-strengthened Al composites [3, 4]. Alternatively, powder metallurgy ensures the fine alumina dispersoid is well distributed within the Al matrix, which eventually gives good final mechanical properties to the composite with sufficient physical properties. Mechanical alloying has also been employed to synthesize aluminum matrix composite (AMC). This method is relatively easy to produce composite powders with fine microstructure [5]. Mechanical alloying (MA), that is, a ball milling process where the powder particles are subjected to high energy impact have been recently used in the production of aluminum base matrix composite. MA enables a uniform distribution of the reinforcement particles into the aluminum matrix, the refining of the metal matrix and the fracturing of the hard reinforcement particles [6, 7].

The purposes of the present study were to produce Al-Al_2O_3 composites by MA and conventional powder metallurgy methods using Al_2O_3 particles up to 15 vol.% as reinforcement, investigate the microstructural changes in Al_2O_3 particles and research the influence of MA processing on the distribution of Al_2O_3 particle reinforcement and their corresponding properties.

2. Experimental procedure

The as-atomized Al powders (Gündoğdu Exotherm Company, Düzce, Turkey) with an average powder particle size of 377 μm and a theoretical density of 2.708 g/cm^3 were used as the matrix material and Al_2O_3 particles (Wacker Ceramic Company, München, Germany) with an average particle size of 13 μm and a density of 3.95 g/cm^3 were used as the reinforcements. The chemical composition of the as-atomized Al powder (wt.%) was Fe: 1.230, Si: 1.000, Pb: 1.000, Cu: 0.710, Zn: 0.530, Mn: 0.116, Ti: 0.071, Mg: 0.050 and Al: balance. The morphologies of both powders are shown in Figure 1A,B.

Composite powders reinforced with 5, 10 and 15 vol.% Al_2O_3 particulates, respectively, were produced by ball milling

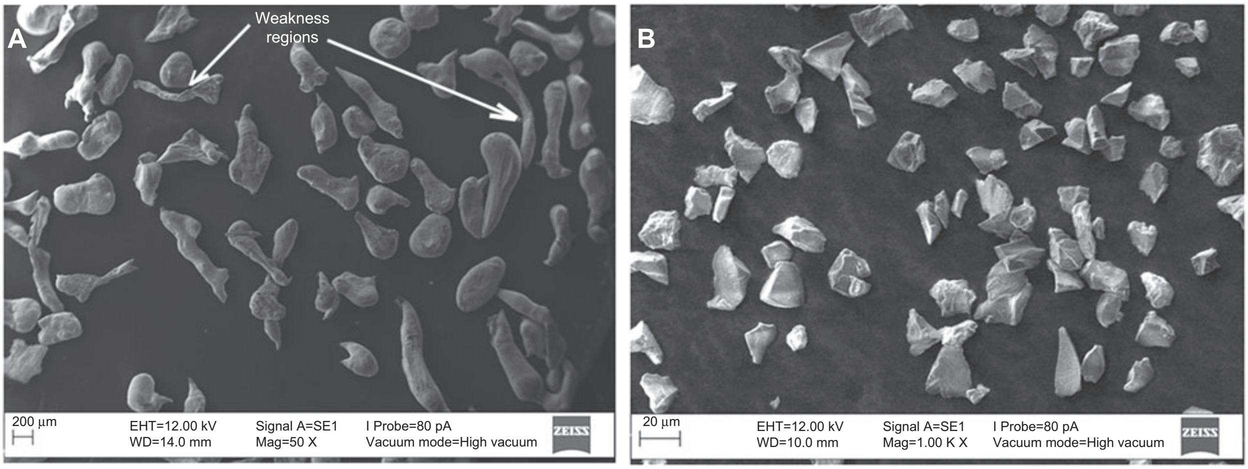

Figure 1 Morphology of as-received powders: (A) Al and (B) Al_2O_3 particles.

for 0.5, 2, 5 and 7 h. The milling process was carried out in a planetary ball-mill (Fritshc GmbH, model 'Pulverisette Premium Line 7') at room temperature using tungsten carbide bowl and high argon atmosphere. The milling medium was tungsten carbide balls, 10 mm in diameter. The ball-to-powder weight ratio and rotational speed were 10:1 and 400 rpm, respectively. A total of 2 wt.% of methanol (Merck) was added to the ball-mill as process control agent. The milling atmosphere was argon which was purged into the bowl before milling. To prevent overheating, ball milling experiments were stopped (every 0.5 h) and then resumed when the temperature of the bowl decreased to room temperature. Powder samples were withdrawn at time intervals of 0.5, 2, 5 and 7 h for morphological, microstructural and structural analysis.

To obtain the raw material, Al and Al_2O_3 particle powders were introduced together into a cylindrical mixer. The purpose of this step which is called 'conventional mixing' throughout this article was to mix powders without changing their original characteristics. The Al powder without reinforcement was used for comparison to determine the effect of the presence of reinforcement particles on consolidation.

The as-received Al powder, the 'conventionally mixed' (CM) and the mechanically alloyed powders were uniaxially cold pressed in a cylindrical die at 700 MPa, with graphite as the die lubricant. The green compacts were sintered at 600°C for 3 h under high argon atmosphere. The sintered compacts were cooled to room temperature in the sintering furnace. The experimental density of the composite samples was measured using Archimede's method. In this technique, density was determined by measuring the difference between the samples weight in air and when it was suspended in distilled water at room temperature. The theoretical density was calculated using the mixture rule according to the volume fraction of the Al_2O_3 particles. The morphologies, distribution of the Al_2O_3 particles and the microstructures of the Al-Al_2O_3 composites were analyzed using scanning electron microscopy (SEM).

The particle size distributions were determined using laser diffraction (Malvern, Model mastersizer Hydro 2000e), connected to a computer that models the volume size distribution, D_{10}, D_{50}, D_{90} calculation automatically; three measurements were carried out for each sample. The phase identification of the products was conducted by X-ray diffraction (Rigaku Corporation, Japan) using Cu-Kα radiation.

The hardness of the samples was measured using the Brinell hardness method under a load of 31.25 kg held for 20 s. Reported hardness values were the average of five measurements.

3. Results and discussion

3.1. Particle size evaluation

The effect of milling time on the particle size of ductile-ductile and ductile-brittle powders has been studied separately by previous authors [8–13]. The effect of milling time on the average particle size of monolithic Al and Al-Al_2O_3 composite powders is shown in Figure 2. Unlike most of the

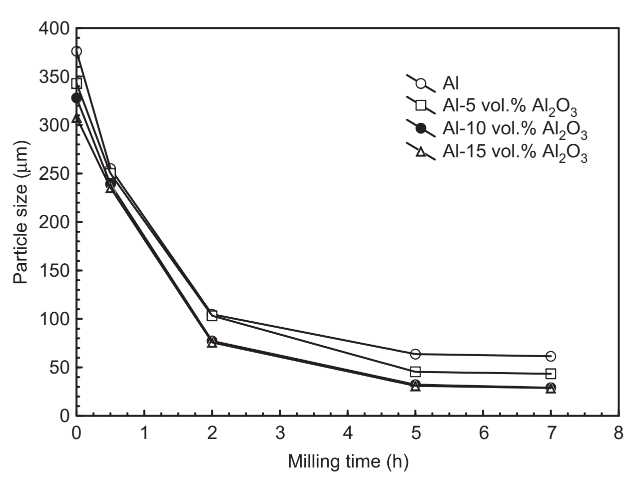

Figure 2 The effect of milling time on the average particle size of monolithic Al and Al-Al_2O_3 composite powders.

work related to MA [7, 14–16] in this study, the particle size decreased continuously. This may be attributed to the initially used Al powders having ligamental shape. These powders can be easily deformed by ball collisions and they can be easily fractured in weak regions. It should be noted that initial powder morphology is an important factor affecting the change of the particle size. The average particle size ($d_{0.5}$) of the monolithic Al and Al-Al_2O_3 composites powders decreased with increasing milling time (Figure 2).

3.2. Morphologies of powders

Figure 1A,B show the morphologies of the as-received Al and Al_2O_3 particles. The Al matrix powders were of ligamental-like shape and the Al_2O_3 particles were polygonal in shape. Mechanical milling severely deformed the powders and flattened powders were formed [12]. After 0.5 h milling, the Al powders formed into a flake-like shape (Figure 3A). Owing to the ductile nature of the Al powder, welding seems to be the dominating mechanism over this stage of milling [8, 11, 12] and thus the 0.5-h milled particles flattened shape (Figure 3A). The flatten shape-like Al powders were work hardened after 2 h milling, hence fracture mechanism was activated (Figure 3B). SEM images of milled Al powders showed that after 5 h milling, the morphology of Al powders were completely equiaxed, which was the characteristic of Al powders at steady state. Further milling up to 7 h had no effect on morphology (Figure 3C); indeed, at milling times longer than 5 h the steady state predominates.

The variation of Al-Al_2O_3 powder shape during ball milling as a function of milling time up to 7 h is shown in Figures 4–6. During the MA process, powders are subjected to ball-powder-ball collisions, which caused severe plastic deformation, cold welding and fracturing of the powders [17]. Plastic deformation and cold welding were predominant during the initial stage of ball milling, in which the deformation led to a change in particle shape and cold welding led to an increase in average particle size. With continued milling, the cold welding and fracturing events continue to take place leading to microstructural refinement. At this stage, the particles consist of convoluted lamellae (Figure 4A). Owing to the increased amount of cold working, the number of crystal defects introduced – dislocations, vacancies, grain boundaries, etc. – increases with time. The impact of the ball-ball, ball-powder, and ball-wall collisions also causes a rise in powder temperature. Work hardening occurs due to the combined effects of all these factors [17]. The addition of hard reinforcement particles would accelerate the fracture process of the matrix powders, which was reported elsewhere [14]. The increased fracturing tendency for high reinforcement was due to more collision with balls and more support from hard ceramic particles [7]. If the presence of reinforcement results in a high deformation of the metallic matrix and advances the mechanic alloying process, a higher reinforcement fraction will result in a still greater deformation of the metallic particles and will accelerate the process even more [7, 14, 18].

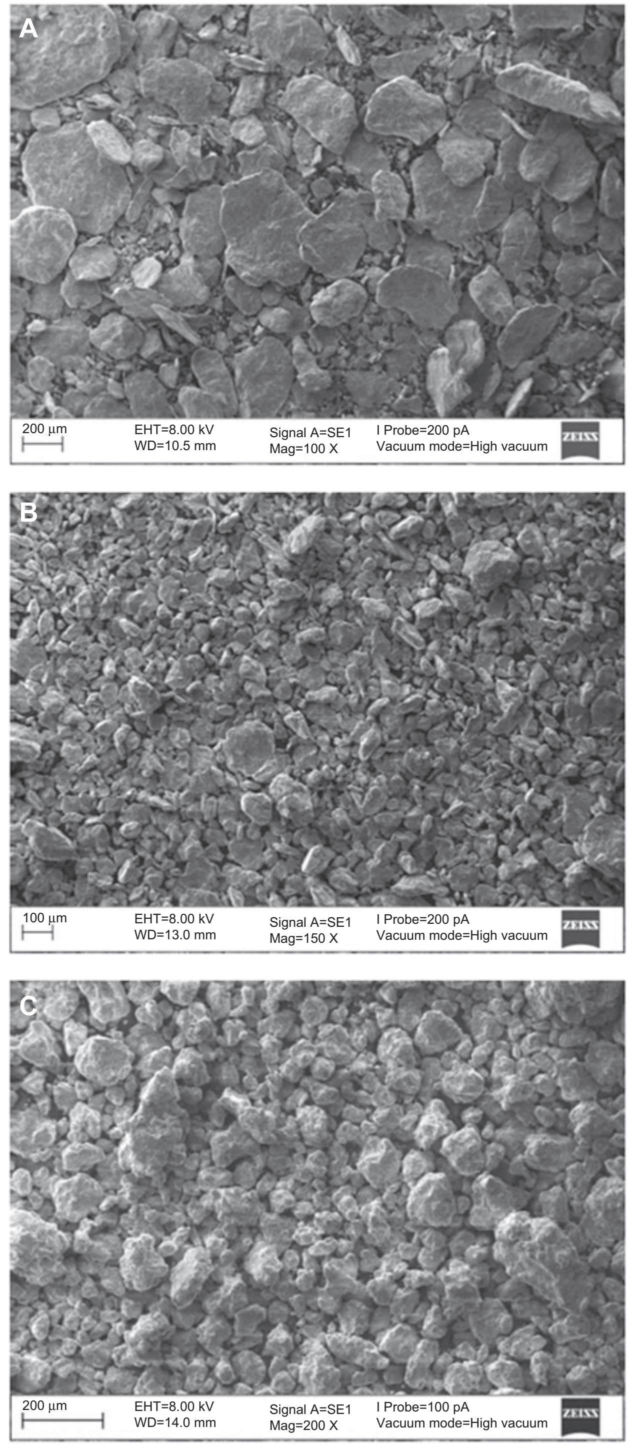

Figure 3 Morphology of the milled Al powders for: (A) 0.5 h, (B) 2 h, (C) 7 h.

The Al_2O_3 particles were increasingly embedded in the matrix powder with increasing MA time (Figure 7A,B). The Al_2O_3 particles embedded into Al powders during milling lead to their fracture toughness reduction enhancing their fracture [19]. Figure 7C,D show fracturing starting regions of Al-10 vol.% Al_2O_3 composite powder after 2 h milling time.

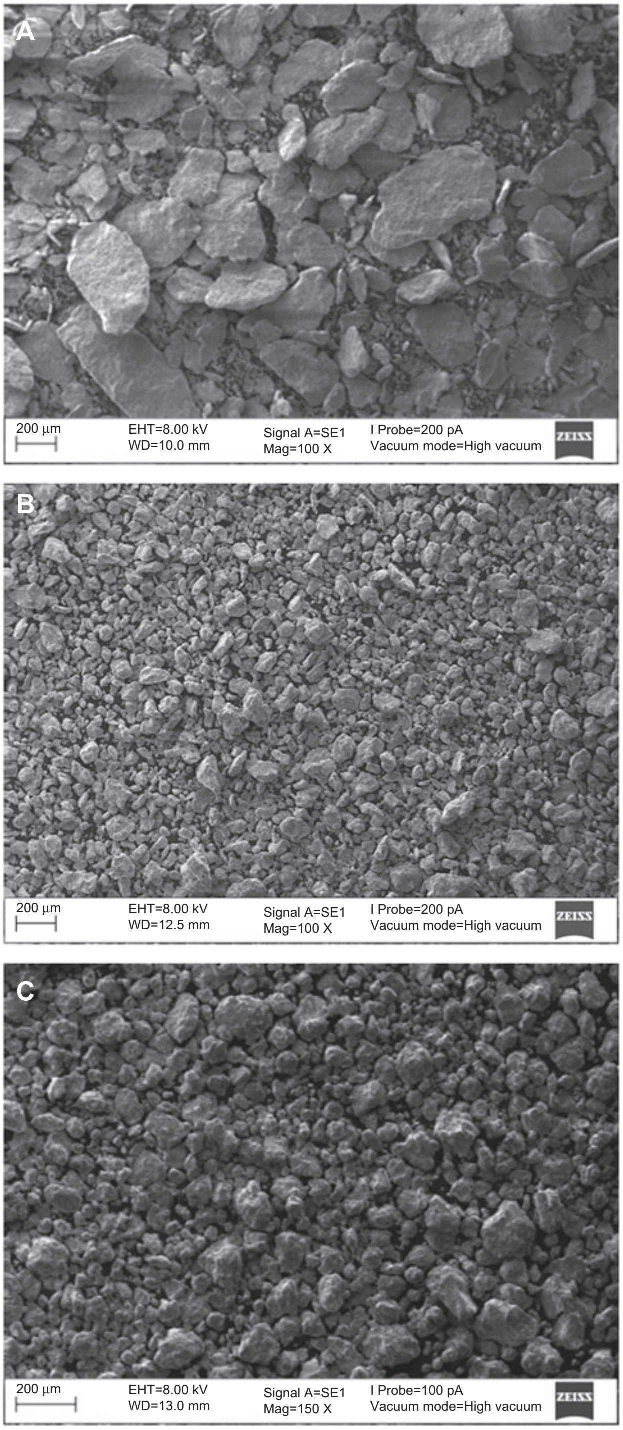

Figure 4 Morphology of Al-5 vol.% Al_2O_3 composite powder as a function of milling time: (A) 0.5 h, (B) 2 h, (C) 7 h.

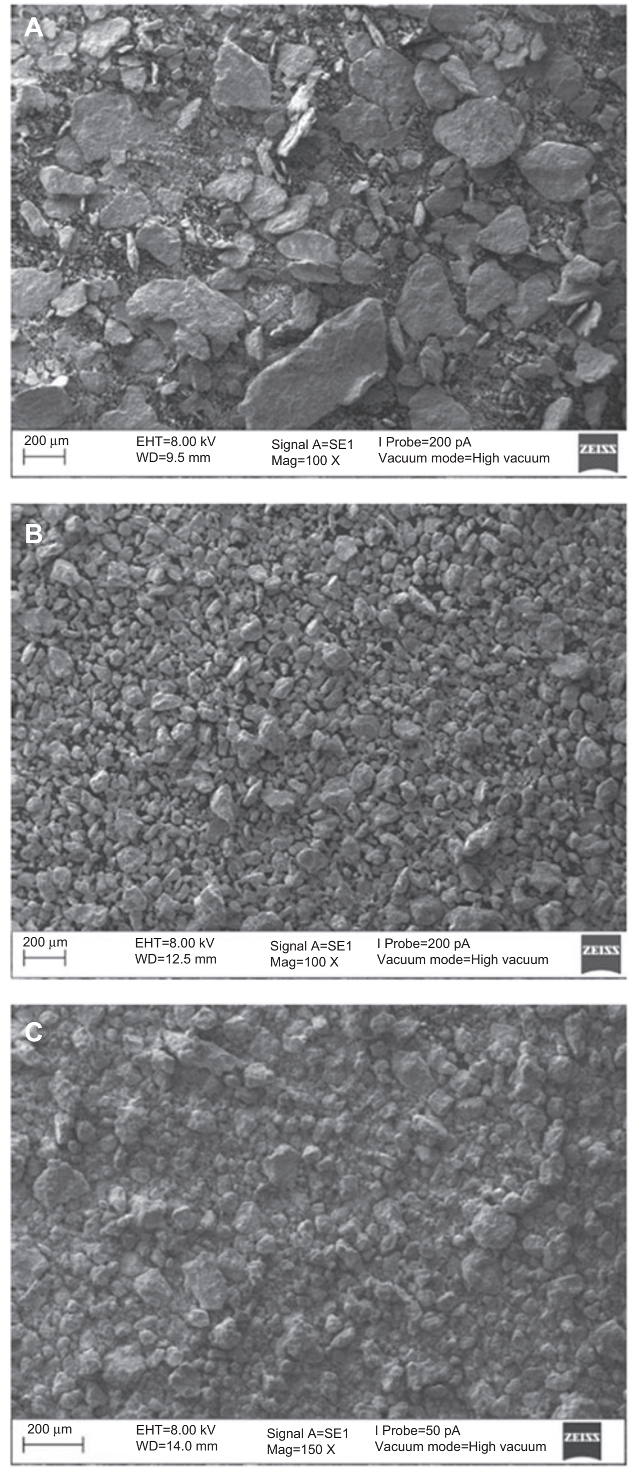

Figure 5 Morphology of Al-10 vol.% Al_2O_3 composite powder as a function of milling time: (A) 0.5 h, (B) 2 h, (C) 7 h.

3.3. X-Ray diffraction pattern

The structural evolution of mechanical alloyed composites can be seen from X-ray diffraction (XRD) spectra shown in Figure 8. Figure 8 shows XRD patterns of as-mixed and the mechanically milled Al-10 vol.% Al_2O_3 composites after 0.5, 2, 5 and 7 h milling time. As can be seen in Figure 8, the intensity of the diffraction peaks decreased with increasing milling time and became wider. It was evident from Figure 8 that there was no reaction between Al and Al_2O_3 particles for different milling times as Al and Al_2O_3 phases were still present in the powder mixtures.

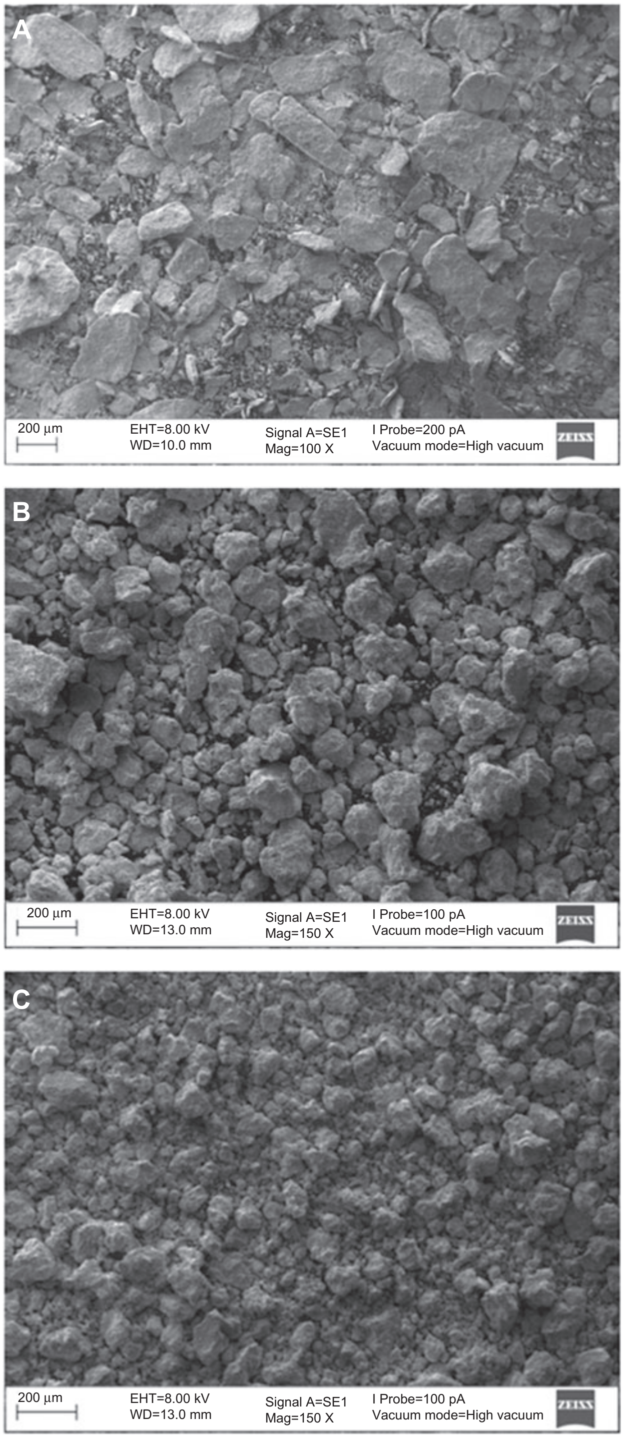

Figure 6 Morphology of Al-15 vol.% Al_2O_3 composite powder as a function of milling time: (A) 0.5 h, (B) 2 h, (C) 7 h.

3.4. Microstructure evolution

Figures 9–12 show the SEM micrographs of unreinforced Al and Al-Al_2O_3 sintered composites with varying amounts of Al_2O_3 reinforcement. Microstructure with well-defined grains and lesser amount of porosity mostly at intergranular regions can be seen for unreinforced alloy (Figure 9).

The effect of milling time on the distribution of Al_2O_3 particles was examined by SEM (Figures 10–12). Composites having Al_2O_3 up to 15 vol.% show non-uniform distribution of reinforcing phase in between the grains (0 h milling), as can be seen in Figures 10A, 11A and 12A. Increasing of Al_2O_3 reinforcement particles causes the agglomeration of Al_2O_3 reinforcement phase at grain boundaries which further restricts the interparticle contacts and densification. Non-uniform distribution of dispersoid phase and higher amount of porosity is well correlated with literature reports [2, 20, 21]. Figures 10A, 11A and 12A show the distribution of reinforcement particles obtained by a conventional mixing method (0 h milling) which is known as the routine powder metallurgy process. The reinforcement particles adhere together and a heterogeneous distribution of Al_2O_3 particles were observed that results from the large size difference between Al powder and Al_2O_3 particles.

The reinforcement particle distribution was not uniform at initial milling time (Figures 10B, 11B and 12B). It can be seen that the dispersion of reinforcement Al_2O_3 particles became more homogeneous and a smaller particle size with increasing milling time. However, the particle distribution was not uniform. However, increasing milling time caused to break the big and brittle Al_2O_3 powders and to indent them into the ductile aluminum powders. Also, with increasing milling time the distance between Al_2O_3 particles decreased gradually. After 2 h milling (Figures 10C, 11C and 12C), these particles dispersed throughout the Al matrix with a better homogeneity, although agglomeration could still be seen in some areas. A milling time longer than 5 h provided a homogeneous distribution of the reinforcement particles. Compared with 2 h MA, the distribution of Al_2O_3 in the matrix was more uniform after 5 h MA, as shown in Figures 10C, 11C and 12C and in Figures 10D, 11D and 12D. As can be seen in Figures 10C, 11C and 12C and Figures 10D, 11D and 12D, one can conclude that after 5 h, the milling is under steady-state condition and an increase in the milling time from 5 h to 7 h has no effect on microstructure. In other words, the changes of particle size and Al_2O_3 particle distribution continued until reaching a steady-state condition at this time (after 5 h). This is because the Al_2O_3 particles become fine and a higher energy is necessary to achieve a finer particle. It is worth noting that the achieved steady state is dictated by milling conditions, i.e., type and mill size, number and ball size and amount of charged powders [22].

These results demonstrated that using the MA technique to produce MMC powder produces a homogeneous reinforcement particle distribution, which cannot be achieved by using only a conventional process.

3.5. Hardness

The hardness of sintered samples vs. milling time was plotted in Figure 13. It was seen that the hardness values increased with increasing milling time and Al_2O_3 particle volume fraction. As stated earlier (from 0.5 h to 2 h), MA leads to extreme refinement of the microstructure, finally (after 5 h) resulting in finer level crystalline structure. Also, with increasing milling

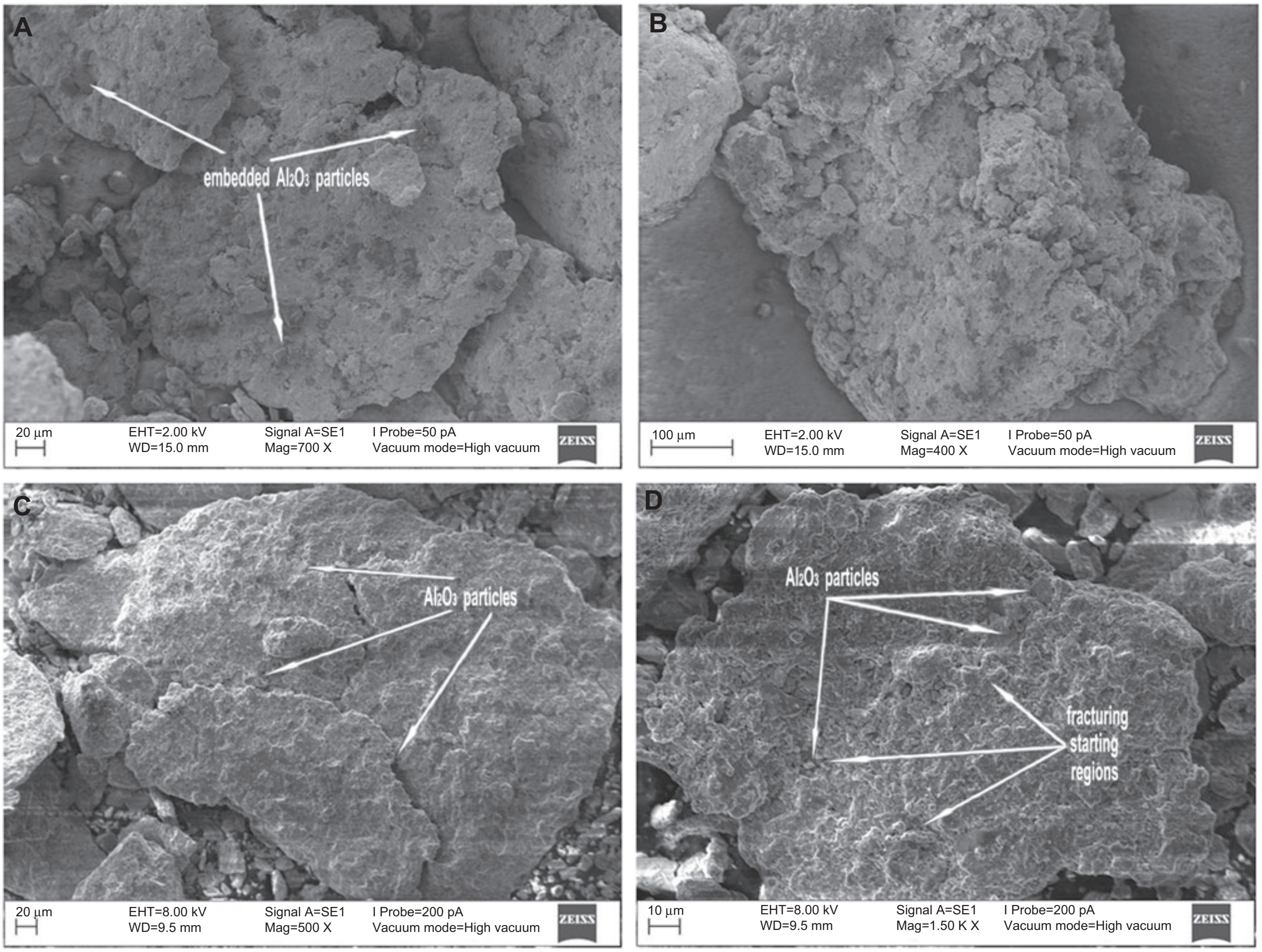

Figure 7 Several morphologies of mechanically alloyed 10 vol.% Al_2O_3 composite powders: (A) 0.5 h (embedded sample), (B) 2 h (embedded sample), (C) 0.5 h (fracturing sample), (D) 2 h (fracturing sample).

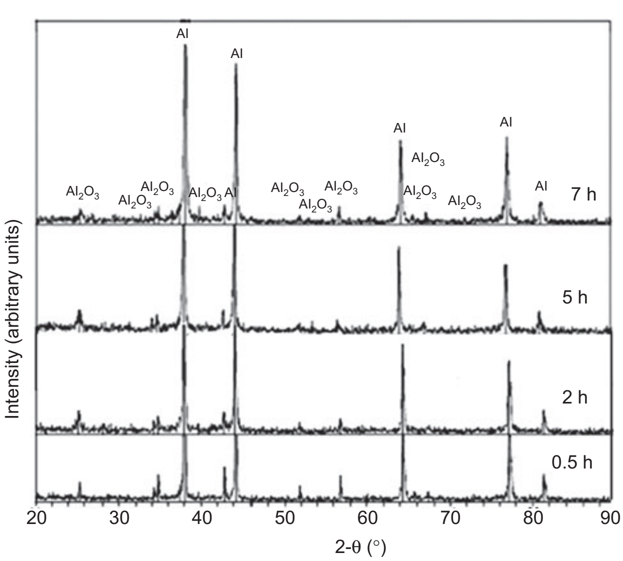

Figure 8 XRD patterns of the Al-10 vol.% Al_2O_3 composite powder after 0.5, 2, 5 and 7 h of milling.

time, more homogeneous distribution and refinement of the reinforcement were observed [11, 23, 24]. These powder characteristics should result in harder structure. It increases continuously, and then a decrease in the slope can be observed until it reaches a steady state (e.g., Figures 3–6 and Figure 2). This confirms that the dependence of hardness or work hardening is more significant. The hardness of mechanical alloyed unreinforced Al and Al-Al_2O_3 composites is affected by two factors: the first factor is the work hardening of matrix alloy due to the milling and second is the role of Al_2O_3 particles as reinforcement [10, 25].

4. Conclusions

In this work, Al_2O_3-reinforced Al base composites were synthesized by the MA process. The milling time was varied from 0.5 h to 7 h while volume fraction was varied from 0% to 15%. MA appears to be an ideal technique to produce composites in a variety of systems. The most impressive advantage of the MA method is that a uniform dispersion

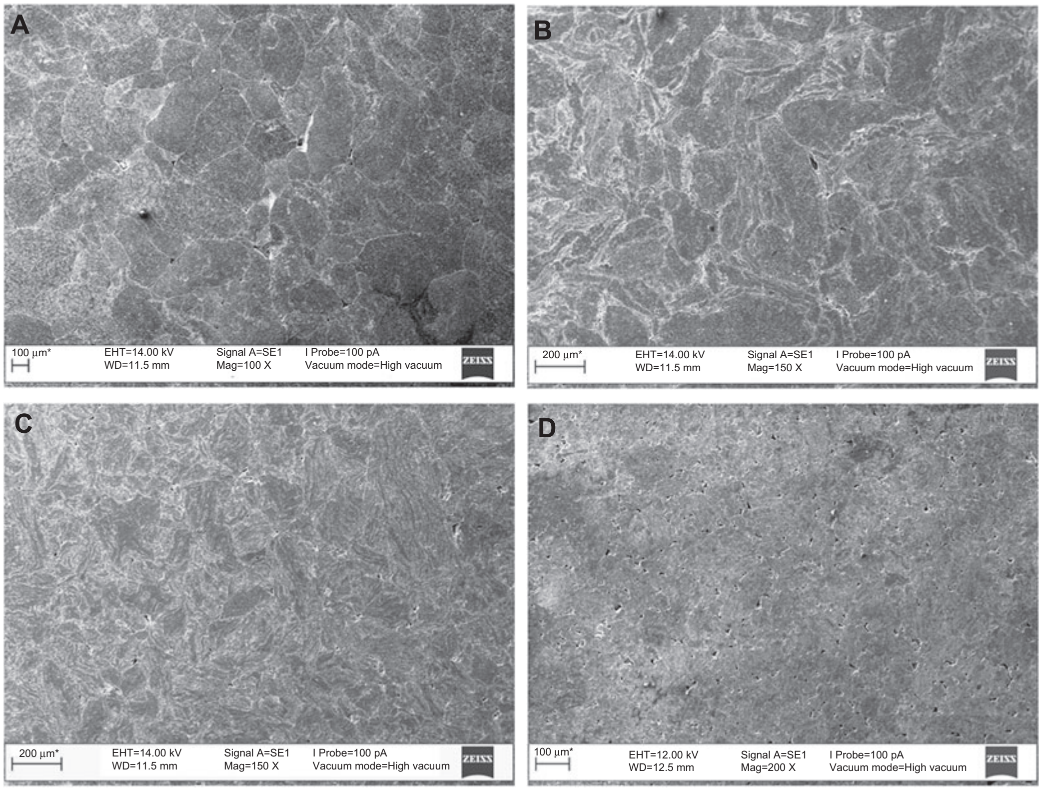

Figure 9 SEM micrographs of unreinforced Al powders after (A) 0 h, (B) 0.5 h, (C) 2 h, (D) 7 h of milling.

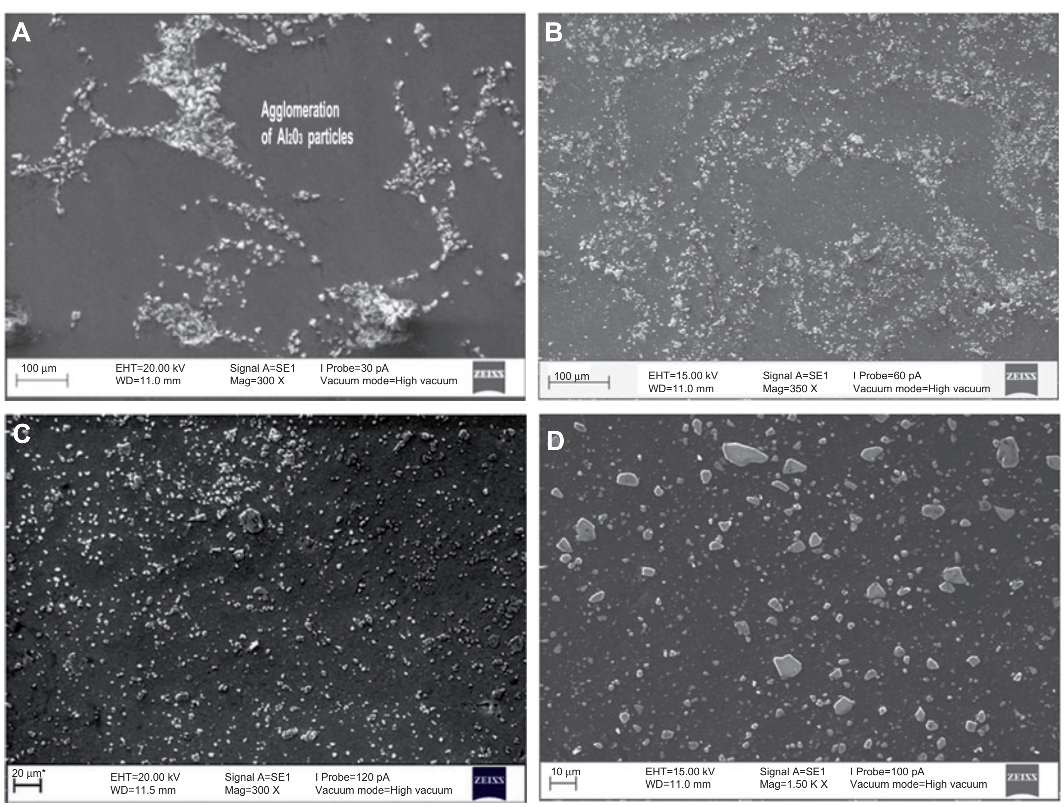

Figure 10 Distribution of Al_2O_3 particles of Al-5% vol. Al_2O_3 composites in the Al alloy matrix with milling time: (A) 0 h, (B) 0.5 h, (C) 2 h, (D) 7 h.

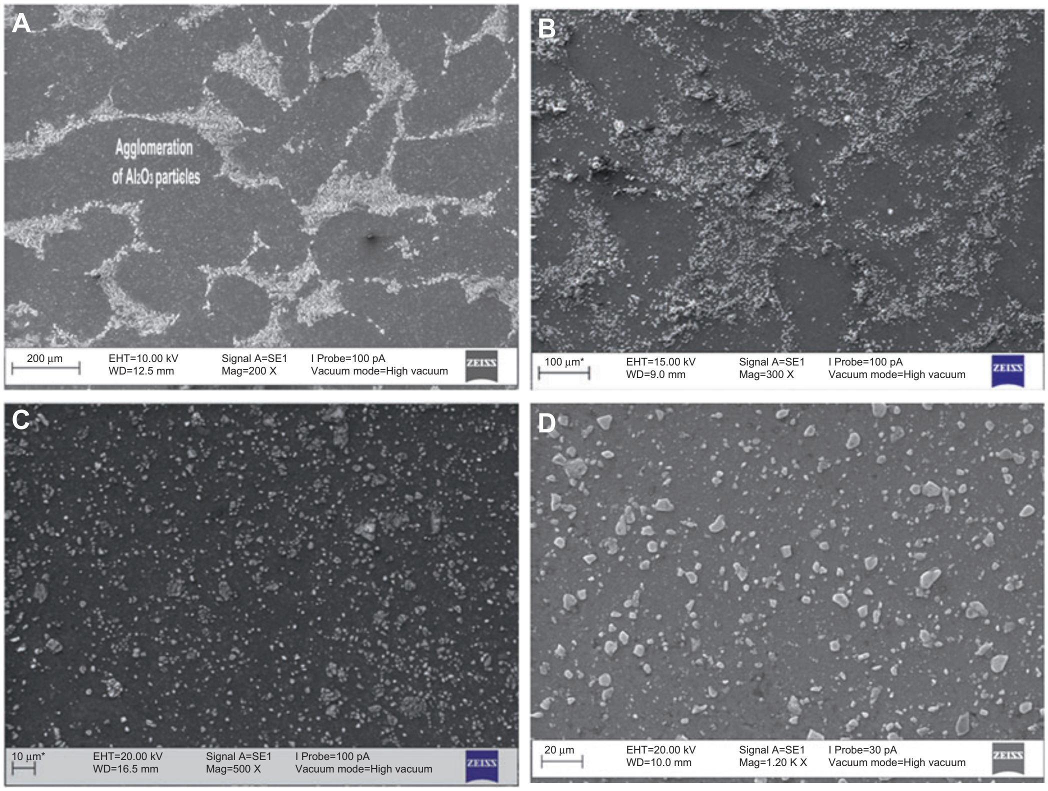

Figure 11 Distribution of Al_2O_3 particles of Al-10% vol. Al_2O_3 composites in the Al matrix with milling time: (A) 0 h, (B) 0.5 h, (C) 2 h, (D) 7 h.

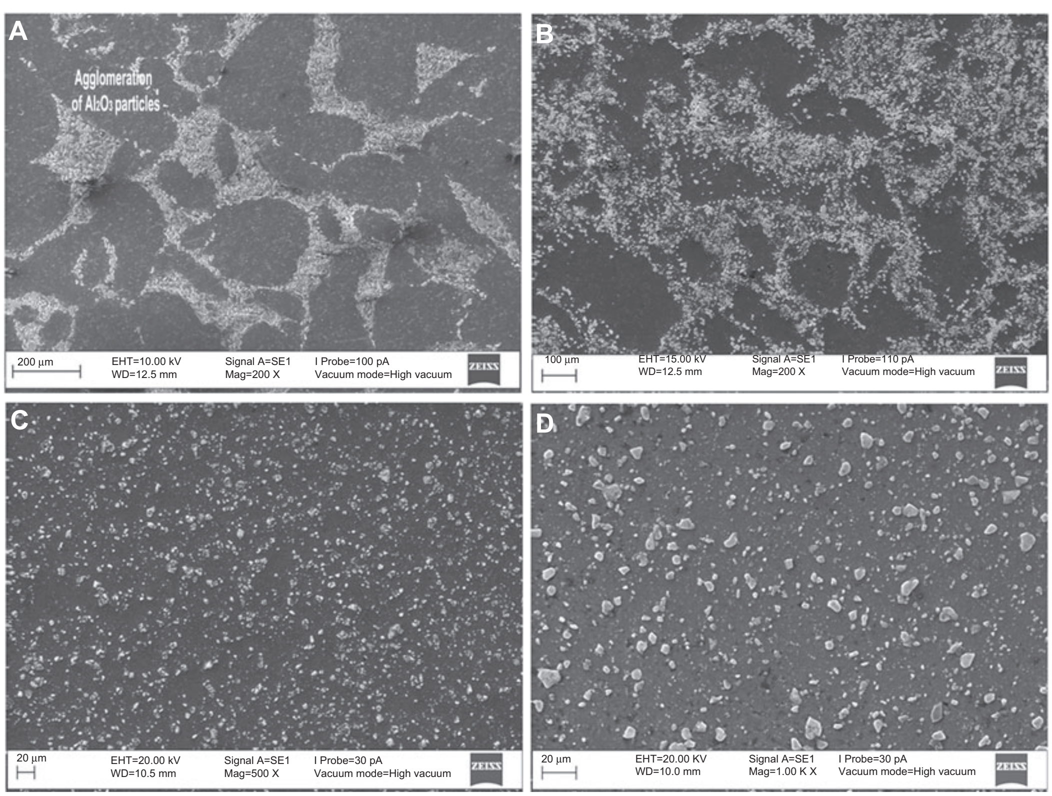

Figure 12 Distribution of Al_2O_3 particles of Al-15% vol. Al_2O_3 composites in the Al matrix with milling time: (A) 0 h, (B) 0.5 h, (C) 2 h, (D) 7 h.

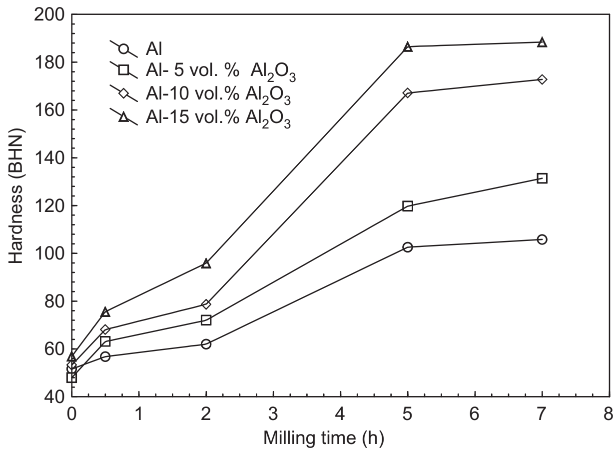

Figure 13 Hardness of the Al and composites vs. milling time at 700 MPa.

can be succeeded by optimizing the process parameters. It was observed that higher hardness is obtained when volume fraction (reinforcement ratio) was increased. The results of the macro-hardness measurements for Al-Al_2O_3 composites showed an important increase in hardness compared with the unreinforced sample. Moreover, the average particle size, powder morphology and hardness of both series of materials (monolithic Al and Al-Al_2O_3 composites) with the function of reinforcement and function of milling time was investigated. It was found that the addition of hard reinforcement in the case of composite powder showed great influence on the morphological and microstructural characteristics of MMCs.

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