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Microstructural Evolution of Al Composite Powders Reinforced by Nano and Micrometric SiC Particles During Mechanical Milling

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Abstract

Aluminum powder and 5 vol.% SiC particles with an average diameter of 50 nm are milled to produce Al-SiC nanocomposite. Al matrix composite containing 5 vol.% SiC with the average particle size of 1 μm was also produced and characterized to examine the influence of nanometric and micrometric reinforcement particles on the mechanical milling process. It was shown that the presence of hard ceramic particles leads to a faster work-hardening and promotes the onset of the fracture process of Al particles. The effect of nanometric SiC on the milling stages was found to be slightly less pronounced as compared with the micrometric SiC. The analysis of XRD patterns revealed that grain refinement occurred faster in the aluminum matrix reinforced with nanometric hard particles.

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Introduction

Development of the modern technologies creates demands for new construction materials with improved mechanical properties, higher corrosion and creep-resistance [1]. Discontinuous reinforced aluminum matrix composites are ranked as one of the most promising materials for structural applications, of which light-weight is crucial [2]. It has been reported [3-6] that by decreasing the reinforcement particle size and the matrix grain size higher mechanical strength is achieved. Smaller particles are less prone to have internal defects and thus are more difficult to be fractured [7]. Additionally, the stress concentration level on each particle is lower as there are more particles bearing applied load, leading to a decrease in the probability of fracturing. Therefore, it is of great interest to use nanoscaled ceramic particles as the reinforcement of metal matrices. Nevertheless, the fabrication of metal matrix nanocomposites (MMNCs) is a challenging task due to difficulties in distributing of the reinforcement particles throughout the metal matrix homogeneously [7]. A very high surface to volume ratio of nanoparticles induces particle agglomeration and clustering, deteriorating the mechanical properties of the composite [8-10].

A look through the literature, for example [11-13], reveals that many works have been performed to improve the distribution of micrometer reinforcement particles into metal matrices. Amongst different procedures, mechanical alloying (MA) has been successfully employed for the fabrication of metal matrix composites [14-17] and metal matrix nanocomposites [18-21]. Naser et al. [22] have shown that this method is a novel approach for dispersion strengthening of metals by nano-oxides where the internal oxidation technique is not applicable. Meanwhile, the procedure is not restricted to nano-oxides, rather than it can be applied to other systems and with any type of reinforcement [19, 23, 24]. Recently, dispersion of SiC nanoparticles in Al-5083 [24], Al-6Ti-6Nb [23], and Mg [19] matrixes by mechanical milling has been investigated. It was shown that the powder particles are trapped between the colliding balls during milling and undergo deformation, welding or fracture, depending on the mechanical behavior of powder components. Therefore, the nanometric particles are distributed throughout the matrix uniformly after a relatively short milling time.

In the present work, the structural evolution during mechanical milling of aluminum powder reinforced with 5 vol.% SiC nanoparticles was studied. The effect of the reinforcement particle size, i.e. 50 nm and 1 μm, on the morphological changes and mechanical milling stages of aluminum powder are compared. The variation in crystallite size and lattice strain versus the milling time is reported in order to highlight the role of nanometric particles on the grain refinement process of the metal matrix.

Experimental Procedure

Aluminum powder with an average particle size of 49 μm and 99.5% purity was produced by nitrogen gas atomization process. The shape of the particles is almost spherical with relatively broad particle size distribution. SiC powder with an average size of 50 nm and 1 μm was supplied by Alfa Aesar (Ward Hill, MA, USA). Figure 1 shows the morphology of SiC particles taken by electron microscopy. The micrometric particles are angular type and the nanometric particles exhibit spherical shape.

Composite mixtures comprising of 5 vol.% SiC and Al powders were prepared by 20 min blending in a Turbula T2C mixer (Basel, Switzerland). To prevent excessive cold welding during milling, 1.5 wt.% stearic acid was used as the process control agent (PCA). Mechanical milling was performed in a planetary ball mill using 200 hardened stainless steel balls of 4 g each. The ratio of ball to powder weight was 10:1. The rotational speed was controlled at 250 rpm. A high purity (>99.999%) argon atmosphere was used to prevent possible oxidation of the new surfaces created on fracturing. The milling experiments were stopped periodically every 120 min and small amount of the powder was collected for testing. In order to highlight the role of nanometric and micrometric SiC particles, the same procedure was applied for the unreinforced aluminum powder.

The particle size distribution of the milled powders was evaluated by laser particle size analyzer (Mastersizer 2000, Malvern Instruments, UK). The morphological changes of the particles during milling were studied by SEM (SEM S360, Cambridge, UK). The apparent density

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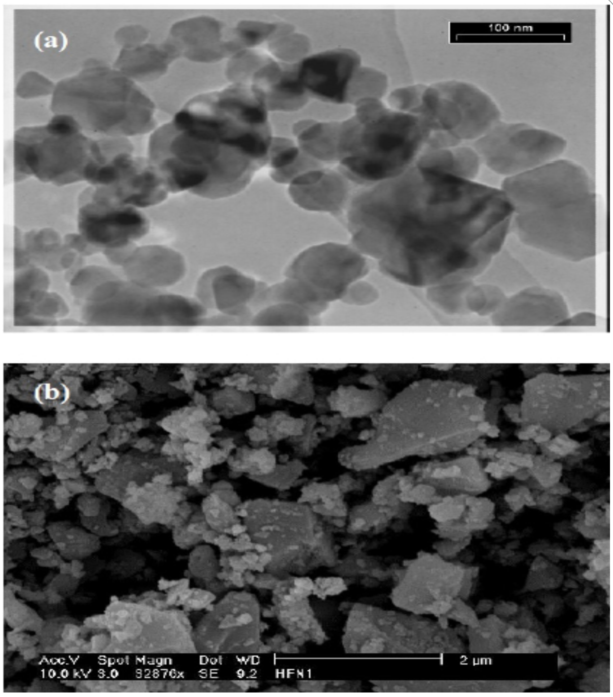

Figure 1: TEM images of nanometric SiC particles (a) and SEM image of micrometric SiC particles (b).

of powders at different stages of milling was measured according to the standard method (ASTM B417). X-ray diffraction (XRD) analysis was employed to determine the crystallite size and the lattice strain. A Philips X'Pert MPD Diffractometer with Cu Kα radiation was used. The Williamson-Hall method [25] was used for analyzing the XRD patterns. An annealed Al powder was used as standard sample for instrumental broadening correction. For microstructural study, the milled powders were cold mounted in a resin, grinded by progressive emery papers, and polished with 1 µm diamond paste. The microhardness of the milled powders was measured by Vickers method at 10 g indenting load.

Results

Morphology

SEM was used to study the structural evolution of powder particles during mechanical milling. Selected micrographs at different milling intervals are shown in Figures 2-4. As seen in Figure 2a, the Al particles exhibit a spherical shape with broad size distribution while small satellite particles attached to the large ones. After 2 h milling, the initial particles were deformed and a change from spherical to irregular shape was noticed (Figure 2b). The average size of the particles was also increased slightly. At longer milling times, the particles were flattened and flake-like particles were formed (Figure 2c and d). Micro-welding between the particles and the onset of fracture were observed at prolonged milling times.

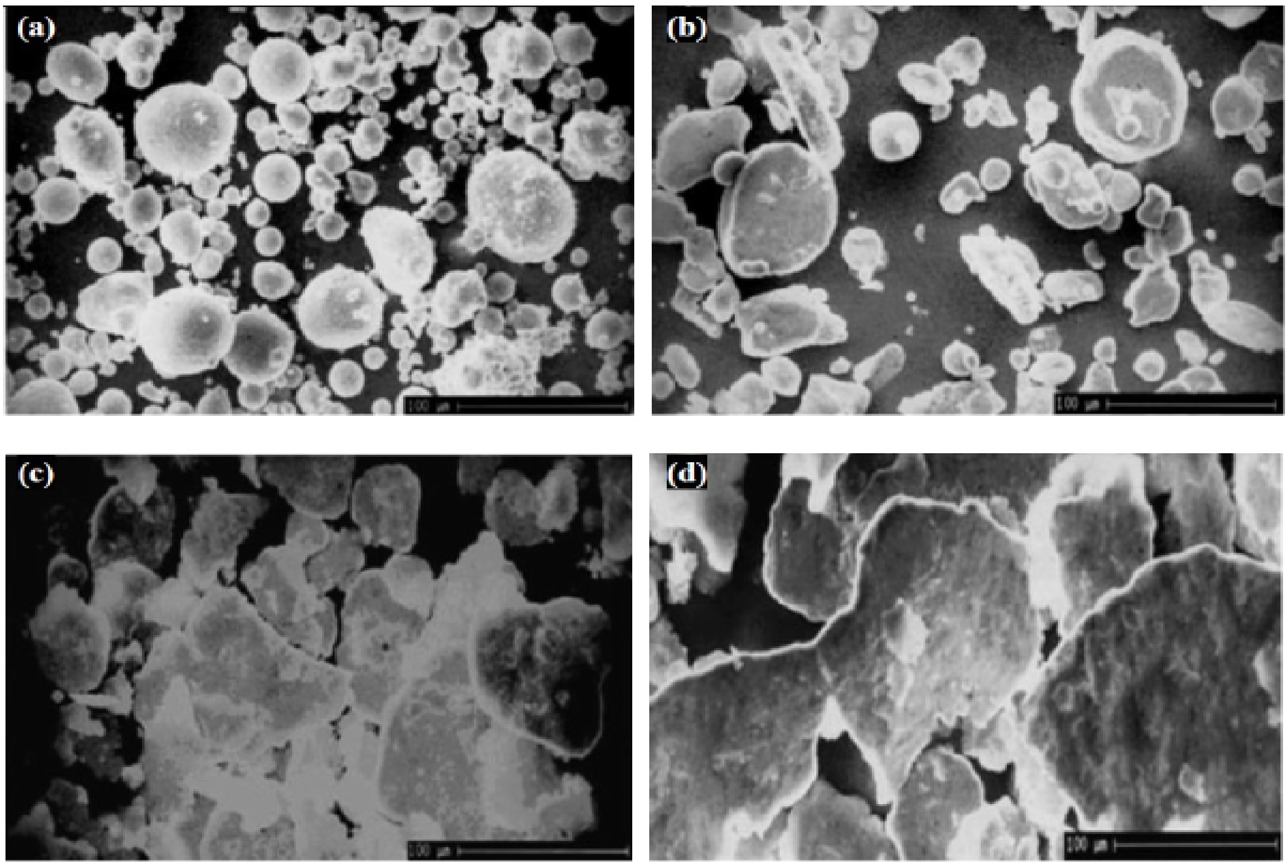

Figure 2: SEM micrographs show the morphology of as-received Al (a) and milled powder for 2h (b), 8h (c), and 18h (d).

When microscaled SiC particles were added (Figure 3), the mechanical milling stages had been influenced. At the early stage of milling (2 h), the effect of hard particles was not pronounced and conspicuous (Figure 3a). After 8 h milling (Figure 3b), the particles were appeared as irregular grains with high aspect ratio. Many small and irregular particles with relatively low aspect ratio were also seen. At longer milling times, a progressive decrease in the aspect ratio of irregular particles was noticed (Figure 3c) while some equiaxed particles were formed after 14 h (Figure 3d). As compared with the flake-like Al particles, the results indicate that welding and fracture proceed faster in the milled composite powder. Meantime, decreasing the SiC particle size from microscale to nanoscale slightly affected the deformation and fracture of the metal matrix upon milling. As Figure 4 shows, at the early stages of milling, particles have undergone deformation (Figure 4a) as similar to the microcomposite powder. The deformation of the particles became more pronounced at longer milling time (Figure 4b) and flattened particles with high aspect ratio were formed. Even after 12 h milling, the flake-like particles were seen (Figure 4c). At this milling time, the microcomposite particles were almost equiaxed whilst the aluminum particles were completely flattened. Further increase in the milling time resulted in a morphological change from laminar to almost equiaxed particles (Figure 4d).

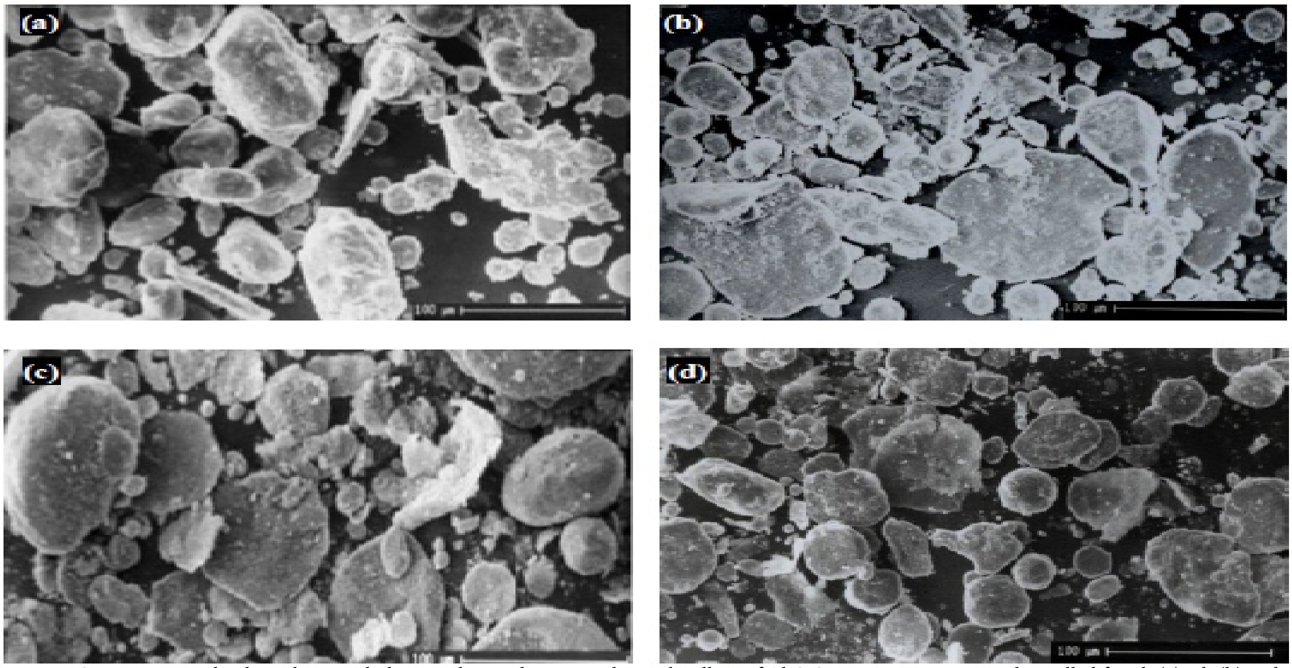

Figure 3: SEM micrographs show the morphology evolution during mechanical milling of Al-SiC microcomposite powder milled for 2h (a), 8h (b), 12h (c), and 14h (d).

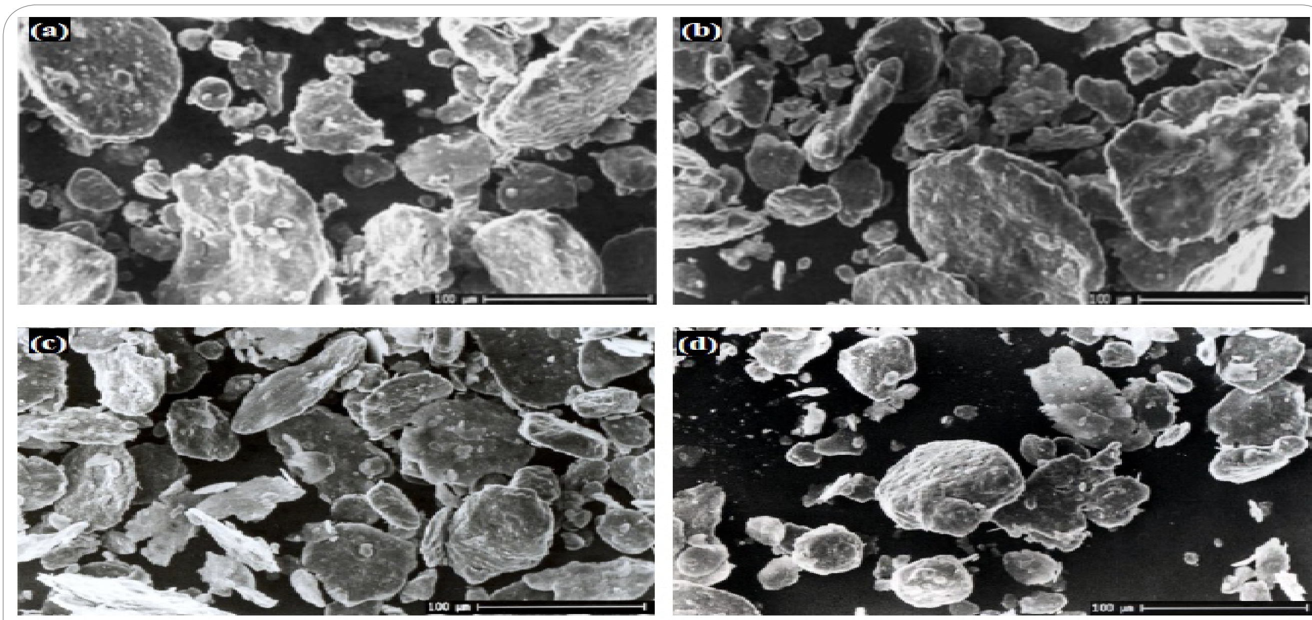

Figure 4: SEM micrographs show changes in morphology of Al-SiC nanocomposite powder milled for 2h (a), 8h (b), 12h (c), and 16h (d).

Particle size distribution

Figure 5 shows the particle size distribution of mechanically milled Al and Al-SiC composite powders. The particle size distribution of un-milled Al powder is included in the graph for comparison. The comparative data of the particle size distribution are summarized in Table 1. All the examined powders exhibit a symmetric log-normal size distribution. The higher average particle size of milled Al powder as compared with the gas atomized one is noticeable. In opposite to this observation, the particle size characteristics of the composite powder containing micrometric reinforcement particles are similar to those of the un-milled Al (Table 1). It appears that the addition of SiC particles affected the fracture of Al particles during mechanical milling, because finer and more equiaxed particles were formed at a given milling time (Figures 3 and 4). A symmetrical Gaussian bell-shaped distribution in the particle size distribution also indicates the equilibrium between fracture and welding, typical of the final stage of mechanical milling [15]. It is also important to notice the difference between the milled composite powders containing micrometric and nanometric reinforcement particles. As it is seen, the average particle size of the nanocomposite powder is higher than that of the microcomposite. The size distribution is also broader. Figure 6 shows the variation of apparent density of powders as a function of milling time. One can notice the higher density of composite powders compared with the unreinforced Al. The lower density of Al powder is related to the flake-like morphology of its particles. In contrast, the irregular shape particles of the composite powders induce higher packing density. It is also noteworthy that, the apparent density of Al–SiC microcomposite is higher than that of the nanocomposite. This observation highlights the effect of reinforcement particle size on the fracture and welding stages occurring upon milling.

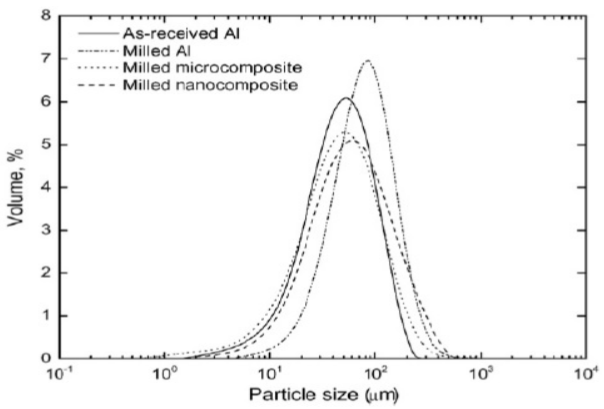

Figure 5: Particles size distribution of milled Al and Al–SiC composite powders for 16 h. The particle distribution of as-received Al powder is shown for comparison.

Materials	D_{10}	D_{50}	D_{90}	D_{90}-D_{10}
Un-milled Al	16	49	117	101
Milled Al	35	88	193	158
Al-SiC (1μm)	12	43	122	110
Al-SiC (50 nm)	19	62	190	171

Table 1: Particle size characteristics of gas atomized aluminum and milled powders[a].
[a]D_n is the particle size (mm) at the n % of the cumulative curve.

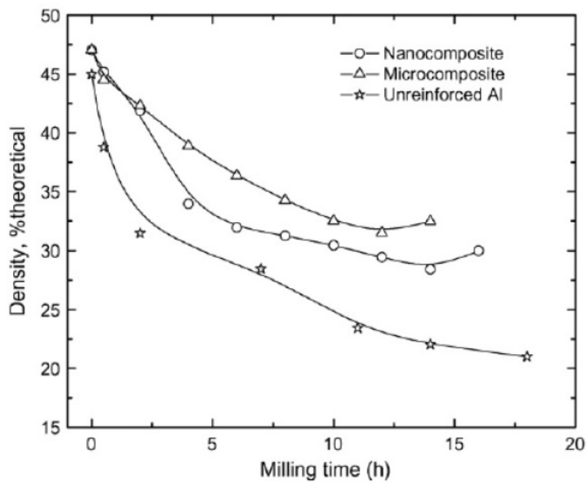

Figure 6: Apparent density (relative to bulk aluminum) of milled powders as a function of milling time.

Microstructure

The microstructure of milled composite powders is shown in Figure 7. The cellular structure of the particles elongated along the deformation axis is visible. The elongated cellular structure of nanocomposite is finer than that of the microcomposite. Although the SiC particles were distributed almost uniformly throughout the Al matrix, the micrometric particles have been refined slightly. The hardness variation versus the milling time is shown in Figure 8. As it is seen, a significant change in the hardness occurred at the early stages of MA. Also, the hardness of nanocomposite powder is higher than that of microcomposite. In order to evaluate the effect of MA on the grain refinement of the Al matrix depending on the reinforcement particles size, XRD analysis was performed. Figure 9 shows the XRD patterns of the Al-SiC composite powders at different milling stages. Peak line broadening represents a decrease in the crystallite size and accumulation of lattice strain. It was found that the effect of nanometric SiC on the peak broadening was more pronounced than that of the microscaled particles. The analysis of the XRD peaks was performed via the Williamson-Hall method according to the following equation [26]:

$$\beta_s \cos\theta = \frac{k\lambda}{d} + 2\varepsilon \sin\theta \qquad (1)$$

where β_s is the sample broadening in radians, θ the position of peak maximum, K the Scherrer constant (K = 0.9 [26]), λ the X-ray wavelength (λ_{Cu} = 0.15406 nm), d the crystallite size, and ε is an approximate upper limit of the lattice distortion. The instrument broadening (βi) was removed using the following equation according to Gaussian-Gaussian relationship by an annealed Al powder:

$$\beta_s^2 = \beta_e^2 - \beta_i^2 \qquad (2)$$

where β_e is the FWHM of the measured XRD peak. The results of XRD analysis are reported in Table 2. It is seen that the crystallite size of Al matrix decreases with increasing the milling time. Furthermore, the grain refinement of the metal matrix occurs faster in the case of nanometric particles whilst the accumulated lattice strain is lower.

Discussion

It is known that during mechanical milling, plastic deformation, welding, and fracture of the particles are dominant mechanisms

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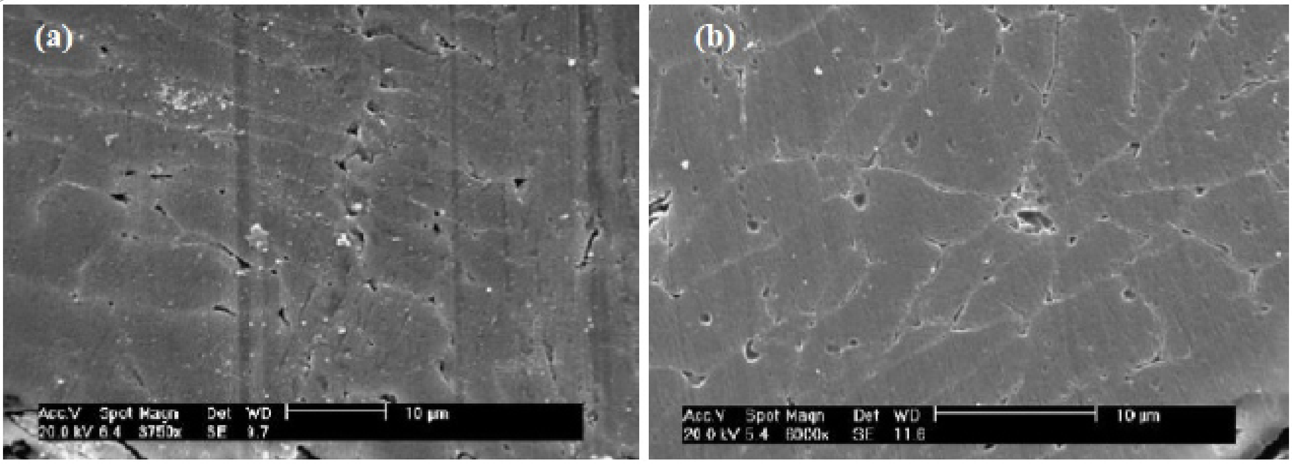

Figure 7: Optical micrographs show the microstructure of mechanically milled Al-SiC nanocomposite (a) and microcomposite (b) powders.

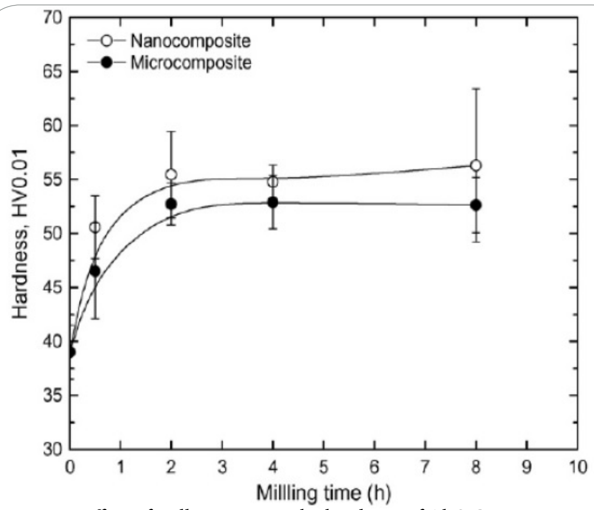

Figure 8: Effect of milling time on the hardness of Al-SiC composite powders.

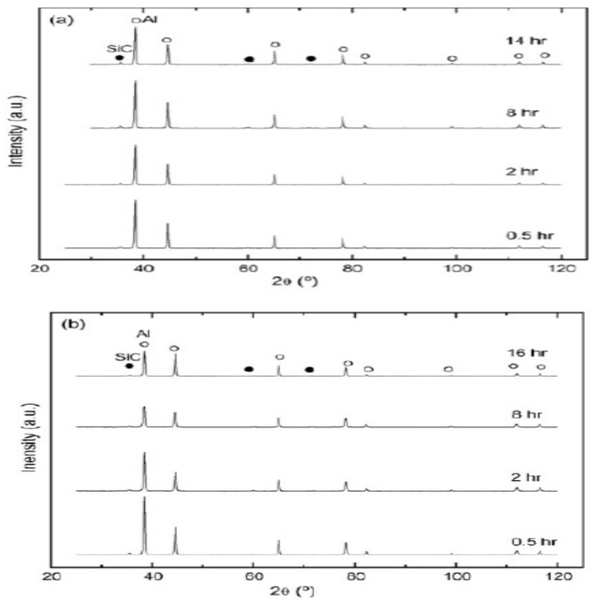

Figure 9: XRD pattern of milled Al-SiC microcomposite (a) and nanocomposite (b) at various times.

Sample	Milling Time (h)	Crystallite Size (nm)	Lattice strain(%)
Al	16	273	0.16
Al-siC(50nm)	2	173	0.1
	8	154	0.09
	16	138	0.09
Al-SiC(1µm)	2	462	0.18
	8	173	0.13
	14	138	0.1

Table 2: Crystallite size and lattice strain of Al-SiC composite powder at different milling times.

that influence the characteristics of milled powders [27]. SEM study of the milled Al powder determined the formation of flaky-like particles during MA (Figure 2). Here, the average particle size was found to be significantly higher than that of the gas atomized powder (Table 1). This observation indicates that the deformation and welding of particles are still the predominant mechanisms of milling. In contrast to this finding, a change in the morphology of Al particles from flake-like to almost equiaxed shape was noticed when SiC particles were added and a prolonged milling time was applied (Figures 3 and 4). The presence of hard ceramic particles influences the milling stages through three main phenomena including (i) local deformation in the vicinity of hard particles, (ii) work-hardening rate of the matrix, and (iii) fracture toughness of the particles. All these impact the deformation and fracture of the particles upon milling and thus the overall MA process. For instance, Figure 10a shows an optical micrograph taken from a particle of milled Al-SiC (1 µm) for 12 h. The ceramic particles are mostly located at or near the interfacial boundaries of the welded aluminum particle. Although the morphology of the particle does not exhibit flake-shape like milled Al, the orientated interfacial boundaries indicate that the steady-state condition had not been reached. Figure 10b shows a SEM picture taken from a particle of milled Al-SiC (50 nm) for 16 h. Relatively homogenous distribution of the reinforcement particles in the matrix is seen. According to the particle size distribution (Table 1) and the variation of apparent density as a function of milling time (Figure 6), it can be deduced that the size of reinforcement particles affects the fracture process. Different sources such as formation of agglomerates of ultrafine particles and the shape-effect of the reinforcement particles (Figure 1) may contribute in difference between the mechanical milling stages of nanocomposites and microcomposites. Anyway, the remarkable difference between the behaviors of two composites was observed at the early stages of milling. Although more detailed characterizations are required to identify the mechanism, it is suggestible that the agglomeration of nanometric reinforcement particles is important parameter which affects the overall milling process.

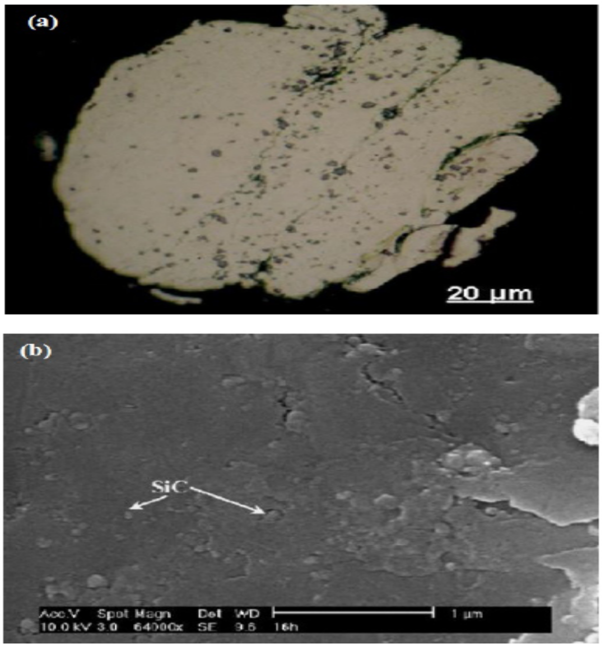

Figure 10: Optical (a) and SEM (b) micrograph shows the distribution of SiC particles in the Al matrix after 14h mechanical milling.

Results of structural study by XRD analysis revealed that, the crystallite size of aluminum matrix in the composite powders is smaller than that of the unreinforced aluminum (Table 2). Also, lower crystallite size was obtained when nanometric particles were used. The acceleration of the grain refinement process by adding SiC particles can be attributed to the generation of a high dislocation activity caused by the interaction between the hard particles and dislocations [28]. Because SiC particles are hard and non-deformable, they can hinder the dislocations movement, leading to an increase in the dislocation density. When the hard particles are small enough (<1µm), the Orowan bowing mechanism leads to dislocation multiplication [29]. The increased dislocation density accelerates the grain refining progress. Furthermore, the mean free path between the particles decreases as they become smaller, which results in more interaction between dislocations and the obstacles. Hence, the grain refining process in the nanocomposite powder is accelerated.

Conclusion

Morphological changes during processing of Al-5 vol.% SiC nanocomposite powder by high-energy mechanical milling were investigated. The effect of reinforcement particles on the milling stages was studied. The findings can be summarized as followings:

- The size of reinforcement particles influences the milling stages of Al-SiC powder mixture during co-milling.
- Morphological study by SEM as well as the apparent density measurement and particle size analysis indicated that the aluminum powder containing nanometric SiC particles approached a steady-state condition in a longer time compared with micrometric particles. This is possibly due to the presence of agglomerates of ultrafine particles at the early stages of milling.
- A faster grain refinement of the Al matrix containing nanometric particles compared with micrometric particles was noticed during co-milling. This finding suggests that the plastic deformation of the metal matrix is affected by the size of reinforcement particles.
- The hardness of milled Al-SiC powder mixture increases abruptly with increasing the milling time and approaches a plateau at the early stages of the processing. The hardness of powder particles reinforced with nanometric particles was found to be higher than those with micrometric particles, particularly at the early stages of milling.

Competing Interests

The authors declare that they have no competing interests exit.

Author Contributions

All the authors substantially contributed to the study conception and design as well as the acquisition and interpretation of the data and drafting the manuscript.

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