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Influence of Mechanical Milling on the SiC Particulate Size in an Al-SiC Composite

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Particle reinforced aluminum-matrix composites are particularly attractive for the automobile and aircraft industries, due to their light weight, high strength, and good wear resistance. In the present work, silicon carbide (SiC) particulates have been incorporated into a pure Al matrix with the help of mechanical milling in a planetary ball-mill. Composite powders were prepared using both raw as well as premilled SiC powders. The effect of milling time on the SiC particulate size was investigated. Systematic analysis of x-ray diffraction data revealed a reinforcement particle size of about 30 nm in a composite containing 50 vol.% SiC. It has been observed that the size reduction occurs at a faster rate when indirect milling is used.

Keywords composites, milling, powder processing

1. Introduction

In the past two decades, metal-matrix composites (MMCs) have emerged as important structural materials (Ref 1-3). Attractive physical and mechanical properties can be obtained with MMCs by incorporating ceramic reinforcements in the metallic matrices. Dispersion-strengthened aluminum (Al) is one of the light MMCs that exhibit attractive properties including high specific strength and enhanced stiffness compared with conventional Al alloys (Ref 4). These Al-matrix composites generally contain 10 to 30% of the particulate reinforcements. There is, however, a tendency for segregation and nonuniformity of reinforcement particles in composites produced through liquid processing, especially when the particle size is <10 μm (Ref 2). On the other hand, particle-induced damage, which is generally associated with particle cracking (with particles >10 μm in size), has been recognized as one of the dominant mechanisms for the failure of MMCs (Ref 5, 6).

Powder processing, including mechanical alloying, has been identified as a promising technique for producing the effective dispersion of submicron particles (Ref 4, 7). Powder-processing routes also offer more flexibility in the selection of matrix alloy chemistry compared with melt-casting techniques. Mechanical milling could thus be used as a means of incorporating fine ceramic reinforcement particulate in a ductile metal matrix to produce composites. Other beneficial effects of the milling are to be able to refine the particulate size and to ensure their uniform distribution (Ref 8, 9).

The present work includes some of the results from the fabrication of Al-matrix composites that are reinforced with relatively high amounts of fine silicon carbide (SiC) particulates. The objective of the study was to follow the reinforcement particle-size reduction during the mechanical alloying process. The study covers the investigation of the composite powder particle size and shape, and the crystal size of SiC in the Al matrix.

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2. Experimental Materials and Methods

The materials used for the milling process were raw powders of Al and SiC with purity of 99.8 and 99.9%, respectively. The average particle size for the Al powder was 20 μm, and that for SiC was 2 μm. Two different sets of experiments were carried out on mechanical milling using a planetary ball mill.

2.1 Direct Milling of Powder Mixture

This is a typical milling process for the mixture of Al and SiC raw powders that is carried out directly after mixing. There is no additional step involved prior to the milling of the mixture. The composition of the mixture was 50% Al and 50% SiC by volume. Milling times of 15, 24, and 30 h were used for these samples.

2.2 Indirect Milling of Powder Mixture

The difference between indirect milling and direct milling of the powder mixture lies in the additional step carried out in the indirect milling approach. This additional step is a premilling of SiC raw powders. These premilled SiC powders are then mixed with the Al powder and are subsequently milled together to produce composite powders. The milling times for the various samples are shown in Table 1.

All of the milling experiments were carried out in a Retsch planetary ball mill with a ball-to-powder ratio of 10 to 1. The powders were placed in tungsten carbide vials having a maximum capacity of 250 mL. Spherical balls (10 mm in diameter) made of tungsten carbide were used as the grinding media. The milling was performed at a constant speed of 200 rpm, with 40 mL of ethanol added as the processing control agent. The powder particle size and morphology were studied using either a JEOL JSM 5310 (Tokyo, Japan) scanning electron microscope (SEM) or a JSM 6340F SEM equipped with a field emission electron gun.

X-ray diffraction (XRD) analysis was performed using Shimadzu (Tokyo, Japan) 6000 Laboratory X diffractometer on powder samples to identify the phases and for calculating crystal size by measuring the broadening of the peaks. The peak broadening in the diffraction patterns is believed to arise from three factors (i.e., instrument effects, small crystal size, and lattice strain in the material). Thus, to enable an accurate calculation of crystal size, a correction factor needs to be involved in the peak-broadening measurements. In the present work, along with the correction factor, the Scherrer formula (as shown in Eq 1) was used for estimating the SiC crystal size (Ref 10):

$$B = \frac{0.9\lambda}{t \cos \theta} \quad \text{(Eq 1)}$$

where B is the broadening (or peak width) of the diffraction peak at half of its maximum intensity, λ is the wavelength of the radiation used, t is the crystal size, and θ is the diffraction angle. The Scherrer formula was used to obtain the crystal size of the SiC with the assumption that the nature of SiC particle is brittle (i.e., ceramic powders) and will experience minimal plastic strain during milling. Thus, peak broadening (B) is only due to small crystal size and instrument effects. To remove the instrument effects, the following relationship given by Warren (Ref 11) was used:

$$B^2 = B_M^2 - B_I^2 \quad \text{(Eq 2)}$$

In this case, B_M is the measured peak width of the diffraction peak from the milled sample and B_I is the measured peak width from an unmilled sample having grain size >1 μm. The full-width half-maximum of the intensity of the strongest peak of SiC was used in the calculations.

3. Results and Discussion

The particle size and morphology of all the composite powders was observed under the SEM, whereas crystal size was measured and structure was observed using XRD. The as-received SiC powder had an angular morphology with an average particle size of ~2 μm, as shown in Fig. 1. On the other hand, the as-received Al powder was spherical in shape, and the particle size ranged from 5 to 20 μm.

3.1 Direct Milling

Figures 2 to 4 show the SEM micrographs of composite powders obtained after various conditions of direct milling. It can be seen that the size of the composite powder particles after 15 h of milling had increased compared with those of the as-received powders, due to the agglomeration and cold welding of the ductile Al particles (Fig. 2). The particle size ranged from as little as 1 μm to >20 μm. The composite powder particles after 24 h of milling also showed a similar size range (Fig. 3). The particles, however, assumed a flattened shape and were observed to have a layered morphology. After 30 h of milling, as shown in Fig. 4, the composite powder particles were observed to have developed a more equiaxed shape.

Most of the SiC was incorporated uniformly into the Al matrix, although isolated fine particles were sometimes found adhering to the surface of composite powders. The embedded SiC particles were not discernable with the SEM. However, almost all of the energy dispersion spectrometer (EDS) spot analyses carried out on individual composite particles revealed the presence of Si and C along with Al, suggesting that they were dispersed inside the Al matrix, while being reduced in size.

3.2 Indirect Milling

In this process, the as-received SiC powder was first milled separately for varying durations of time. The particle size of the SiC powder, in this premilling process, was reduced to the nanometer scale. The premilled SiC powder was subsequently mixed with the as-received Al powder before milling of the composite mixture. Each mixture was then milled for 15 h to produce the composite powder.

The milling resulted mostly in flattened and irregularly shaped composite particles with a size range of 0.5 to 50 μm (Fig. 5, 6). The composite particles were generally composed of agglomerates of smaller particles, ranging from submicron to a few microns in size. All the particles, no matter how small or large, contained both Al and SiC, as revealed by EDS analysis in the SEM. It was observed that some composite particles appeared to have a flat surface with a layered structure, while others were rough and equiaxed. The layered structure is believed to correspond to the initial stages in the evolution of particle morphology during milling (Ref 5). Hence, this might indicate that 15 h of Al milling generates a composite particle morphology that is still in the initial stage.

Table 1 SiC reinforcement crystal size for all the samples

Sample	Premilling of SiC		SiC crystal size	Mixture composition, vol.%	Mixture milling		SiC crystal size, nm
	BPR	Time, h			BPR	Time, h	
Direct milling							
A	…	…	2 μm	50 Al: 50 SiC (raw)	10:1	15	51
B	…	…	…	…	…	24	49
C	…	…	…	…	…	30	32
Indirect milling							
D	10:1	4	60 nm	50 Al: 50 SiC (premilled)	10:1	15	41
E	10:1	8	57 nm	…	…	15	30

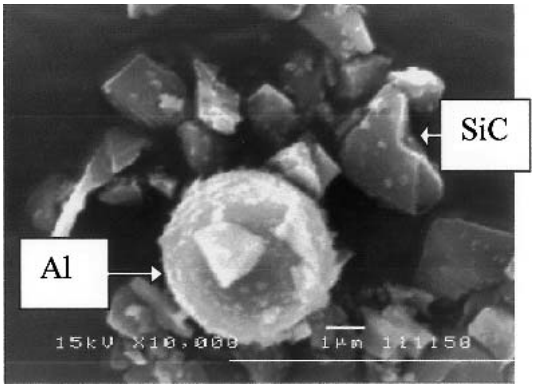

Fig. 1 Initial powder particle size and shape

3.3 Particle Size of SiC Reinforcement

The XRD peaks from the raw powder as well as the composite powder mixtures were identified as Al and αSiC (Fig. 7). These data served as a reference for the identification of the strongest peaks for each phase after milling had been performed. The SiC particle size both after premilling and after composite milling was calculated using the broadening of the diffraction peaks that are shown in Fig. 8 to 10. The calculated values for SiC crystal size are given in Table 1 along with other processing conditions.

The crystal size of SiC in the samples of direct milling compared with as-received powder shows a gradual refinement with the smallest size (32 nm) achieved after 30 h of direct milling. A similar SiC particle size was also produced using the

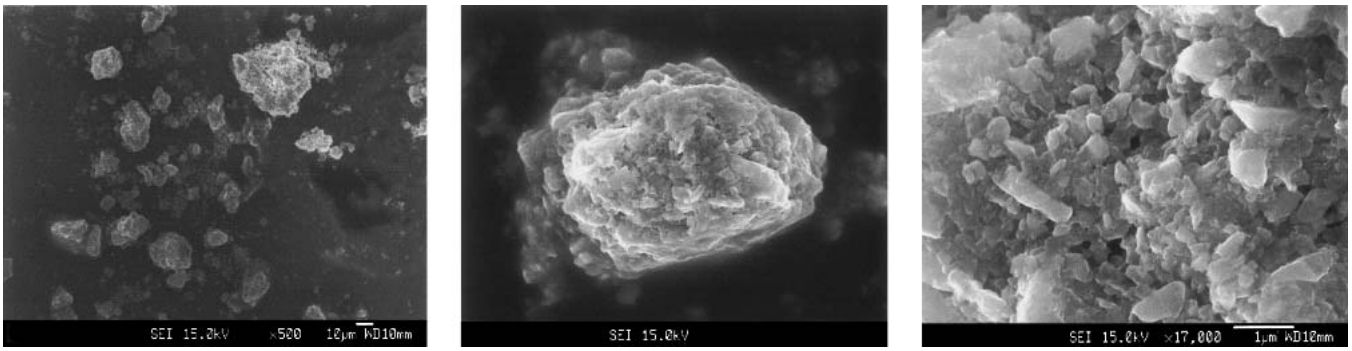

Fig. 2 Size and morphology of composite powder particles after 15 h of direct milling

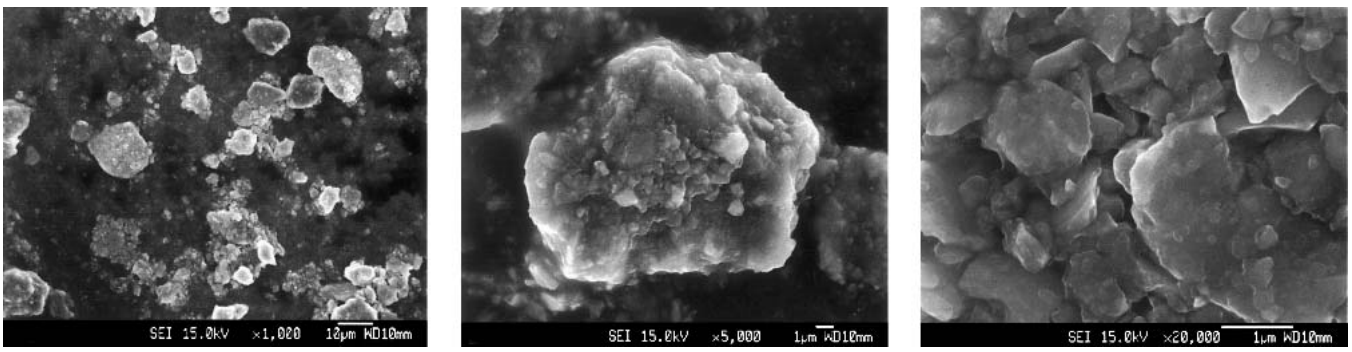

Fig. 3 Size and morphology of composite powder particles after 24 h of direct milling

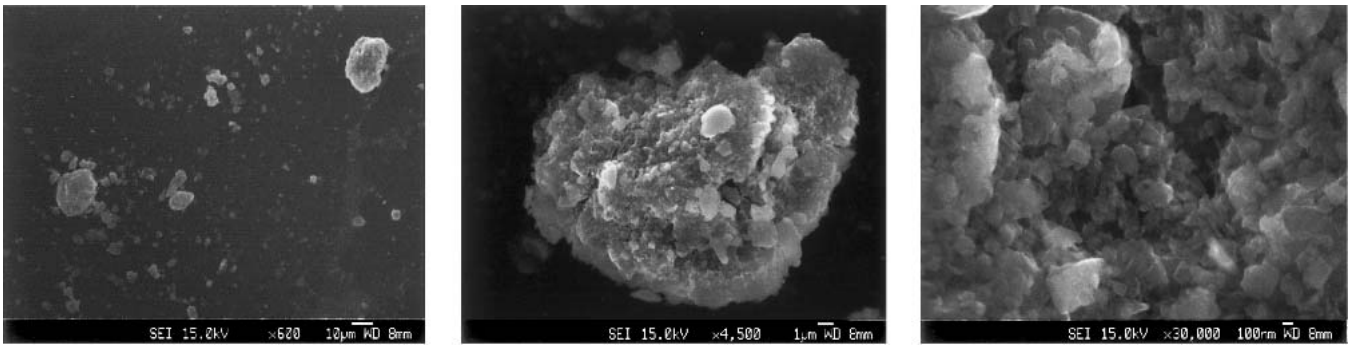

Fig. 4 Size and morphology of composite powder particles after 30 h of direct milling

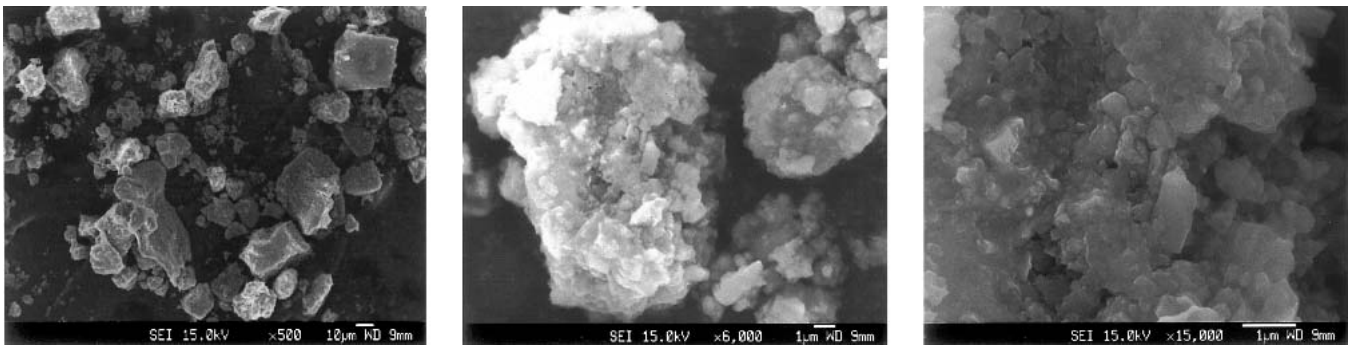

Fig. 5 Size and morphology of composite powder particles after indirect milling (4 h of premilling and 15 h of mixture milling)

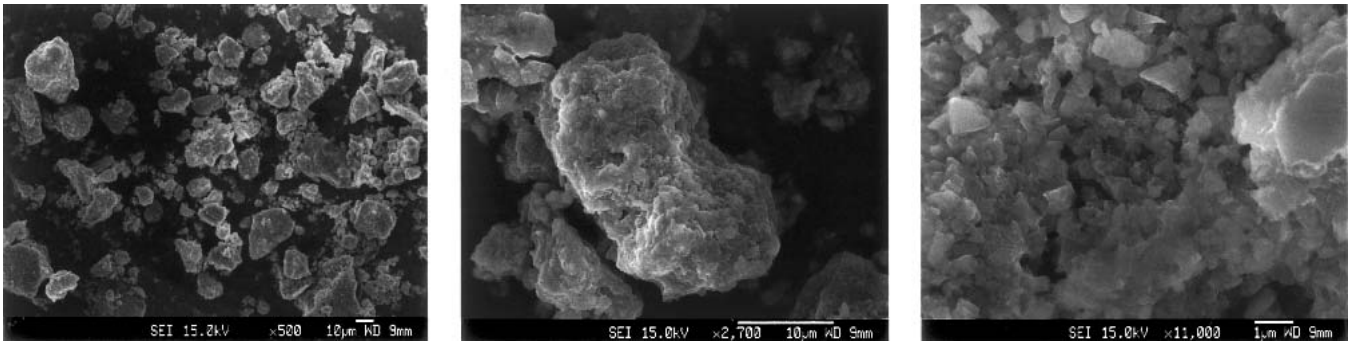

Fig. 6 Size and morphology of composite powder particles after indirect milling (8 h of premilling and 15 h of mixture milling)

indirect-milling method with a total milling time (premilling + mixture milling) of 23 h.

3.4 Evolution of Particle Morphology During Milling

Ball milling generally consists of two processes: one is the relative rolling and friction between the balls and the inner surface of the vial, and the other is the collision between the balls, and also between the balls and the inner wall of the vial. The ratio of rolling and friction to collision depends mostly on the speed of the rotation. When a low speed is used, rolling and friction dominate. Consequently, flattening of the particles may result. On the other hand, higher speeds will promote the collisions causing the powder particles to cold-weld, resulting in efficient mechanical alloying. For soft materials like Al, the powder particles may even be cold-welded to the balls and to the inner wall of the vial (Ref 5). Composite particles larger than those in the original powder would usually be produced under such conditions, which is also evident from the present experimental results.

Mechanical alloying is believed to occur in four stages, producing different particle morphologies during each stage. In the initial stage (short milling duration), when the materials are generally soft, flattening due to compressive stress with no cold welding is likely to occur. The intermediate stage results in the significant cold welding of particles, leading to the formation of a layered structure. In the third stage, considerable refinement and reduction in particle size occurs. The completion stage is where the powder particles exhibit an extremely deformed structure and the lamellae within the particles are no longer easily resolvable (Ref 5).

In the current study, all of the composite powders that had been milled for 15 h (with both indirect and direct milling) showed that some of the particles had the flat and layered surfaces. However, some particles also had a very rough and equiaxed shape. After prolonged milling, the particles hardened with the lamellae further refined, resulting in a decrease in interlamellar spacing. It is thus believed that the composite powder milled for 15 h is at the beginning of the second stage. For 24 and 30 h of direct milling, the powder surface and shape became rough and equiaxed, so that flattened surfaces were not generally observed. This could be attributed largely to the induced brittleness resulting from the incorporation of fine SiC particles into the ductile Al matrix. This feature was more prominent for 30 h of milling, indicating the final and completion stages.

From the data tabulated in Table 1, it can be seen that the final crystal sizes of premilling SiC after 8 h of indirect milling are almost similar (~60 nm) to SiC crystal size after 15 h of milling with direct milling. In other words, indirect milling can produce a similar crystal size for SiC within a shorter time compared with direct milling. This indicates that the milling of pure SiC powder alone will result in faster size reduction than milling a mixture of Al and SiC powders. This is because more impact energy can be transferred to brittle particles when the soft and ductile particles are not present.

To delineate the effects of indirect and direct milling on powder mixtures, a comparison between composite powders

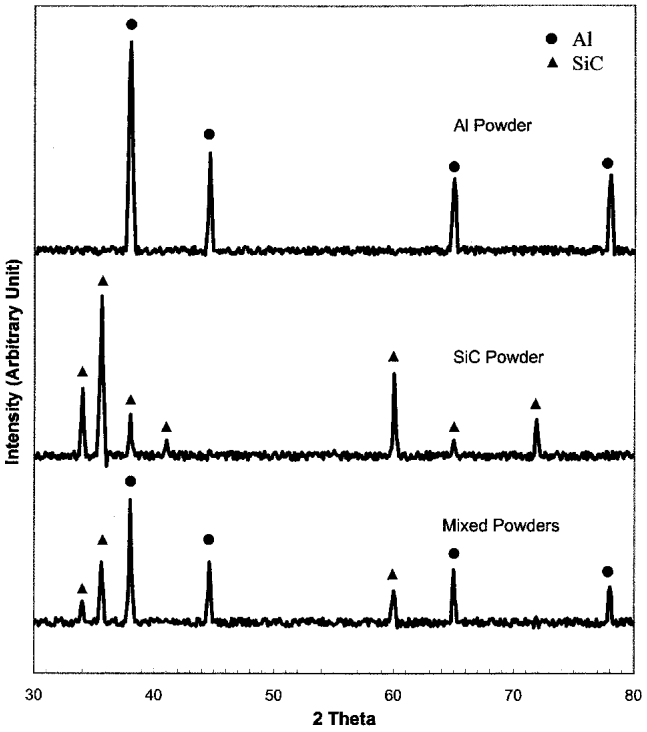

Fig. 7 XRD patterns of as-received Al, as-received SiC, and a mechanical mixture of both

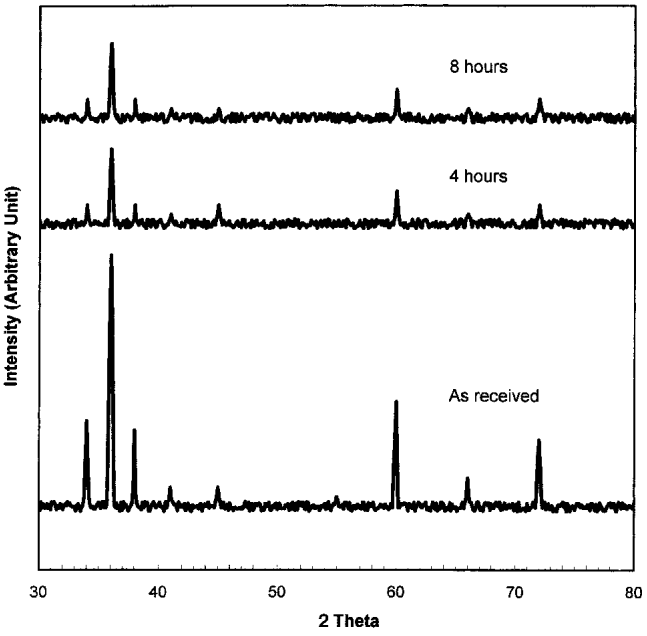

Fig. 8 XRD patterns of as-received and premilled SiC powders showing all the αSiC peaks

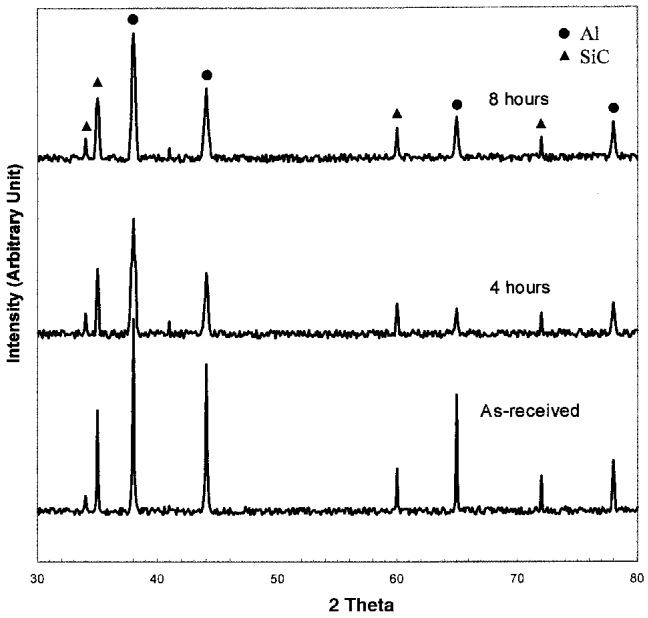

Fig. 9 XRD patterns of the as-received powder mixture and the composite powder after indirect milling

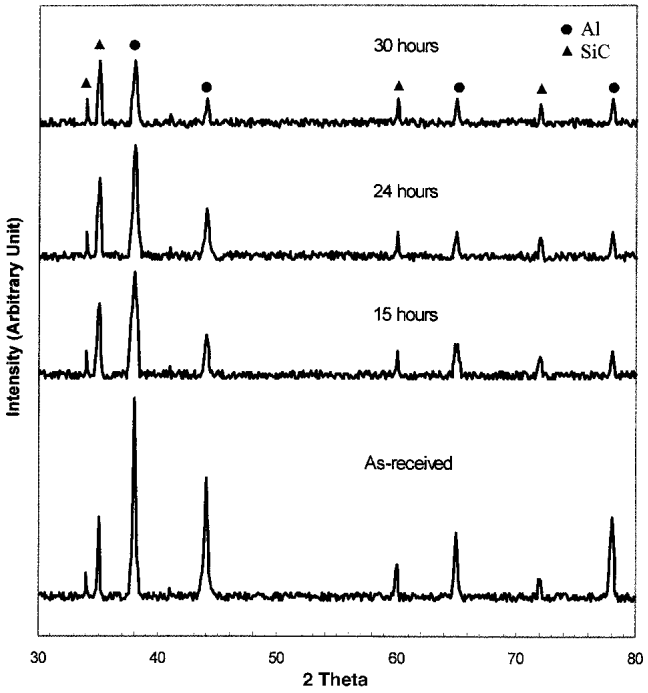

Fig. 10 XRD patterns of the as-received powder mixture and the composite powder after direct milling

from samples B and E was made. It can be seen that 23 h of indirect milling produces a smaller SiC crystal size compared to that with direct milling. Therefore, it may be deduced that the indirect-milling method results in more efficient crystal size reduction. Milling for longer times, for instance in sample C (a powder mixture direct-milled for 30 h), has produced an even smaller final SiC crystal size of 32 nm. There is, however, a limit to particle size reduction in such milling processes. This is because the breakup of ceramic particles by fracturing into smaller size occurs only to the point at which the fracture strength of the small particles become equal to or greater than the stress caused by the collisions (Ref 12).

It can also be noticed in Fig. 10 that, for the as-received powder mixture, a large difference in the diffraction peak intensity exists for Al and SiC (i.e., 2θ at ~35° and ~38°, respectively). However, this difference decreases as the powder mixture is milled for longer times. The peak intensity of both the phases becomes almost equal for sample C (30 h of direct milling). This can be explained in terms of the deformation-induced strain and its accumulation in the Al matrix. This process leads to subgrain formation in the Al matrix, effectively resulting in a smaller grain size. This effect is not seen in the indirectly milled samples, in which the mixture undergoes less deformation, and, hence, less strain is induced in the Al matrix.

For both the direct-milling and indirect-milling methods, 15 h of milling resulted in a particle morphology showing the initial stages of evolution (i.e., a layered structure). After 30 h of milling (using the direct-milling method), however, the composite particle morphology had reached the final stage (or steady state). Comparing the apparent SiC particle for the longest milling time for both methods finds them to be similar in SiC size (~30 nm). The difference only lies in the morphology of the composite particles. The composite particles are flat and layered when indirectly milled and have a rough and equiaxed shape when directly milled. This is also evident in the diffraction peaks where the intensity of the strongest Al and SiC peaks are almost identical compared with the short milling durations where the intensity of the Al peak is much higher than that for SiC peaks. This is an indication that more work hardening is being induced into Al through prolonged milling.

4. Summary

Mechanical milling is an effective way of producing composites with nanoscale reinforcement dispersions. A composite powder (Al/SiC) with a high volume fraction of reinforcement has been successfully prepared. The incorporated SiC reinforcement particles were reduced in size to ~30 nm using both direct and indirect milling.

Direct milling was found to be more effective in terms of SiC embedment in the Al matrix and resulted in a more homogeneous composite material. On the other hand, SiC particle size reduction took place at a faster rate when using indirect milling. However, indirect milling is less efficient for SiC embedment, and longer milling times were needed to induce the same degree of Al deformation to achieve steady-state composite particle formation.

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