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Synthesis and Characterization of Mechanically Alloyed SiC_p Reinforced Al Powder

Muhammed Muaz[1,*] , Akhter Husain Ansari[2], S. K. Chouduhry[1]
[1]Mechanical Engineering Department, Indian Institute of Technology, Kanpur-208016, INDIA
[2]Department of Mechanical Engineering, ZHCET, Aligarh Muslim University, Aligarh-202002, INDIA

Abstract

The main objective of the present work is to produce composite powders of SiCp reinforced Aluminium particles by low speed ball milling using an indigenously developed multi-chamber eccentric ball mill and to study the internal structural evolution, morphological evolution, compositional evolution and homogeneity of reinforcement of SiCp in Al particles. For this purpose, the powders were subjected to X-ray Diffraction Analysis, Scanning Electron Microscopy with Energy Dispersive Spectroscopy and X-ray mapping. Effect of ball-milling time and concentration SiCp on the above characteristics has been investigated. It has been concluded that five hours of ball milling is enough to get a sample with all the particles of Al reinforced with SiCp. However, thirty hours of milling gives better internal structure with crystal size ≤ 30nm, very small particle size and round shape. Effect of mechanical alloying is more significant when concentration of SiCp is high and milling is done for longer time. Inert environment and low milling speed produce contamination free powder particles.

Keywords**:** Mechanical Alloying, Al-SiC_p Powder Composite, Ball Mill, Microstructure, Morphology.

1. INTRODUCTION

SiC reinforced Aluminium matrix composite is used in various highly sophisticated and strategic applications like aerospace, automotive etc. Aluminium is a lightweight ductile material and SiC is a brittle material. The composite has a good combination of required properties of the two phases. Methods like casting, injection moulding and powder metallurgy without mechanical alloying (MA) result in segregation of reinforcement particles, which lead to the deterioration of the properties. MA is a solid-state powder processing technique in which powder particles along with grinding media are agitated in a ball mill. During MA, the particles experience repeated flattening, cold welding, fracturing, and re-welding of welded particles. After MA, composition of individual particles becomes homogenous and same as that of the initial powder blend i.e, each powder particle becomes composite particle having homogenous distribution of reinforcement particles as compared to conventional powder metallurgy technique wherein SiC particles are embedded in bulk of Aluminium matrix after sintering. Al_4C_3 is often seen in the composite prepared by the liquid metallurgy route and is undesirable because of low strength and brittleness.

The internal structural evolution, morphological evolution, compositional evolution and homogeneity of reinforcement of SiCp in Al particles are the important characteristics of SiCp reinforced Aluminum matrix composites that affect the performance of the composites. However, very few researchers have studied all these characteristics. As discussed below, some findings are contradictory/ non-conclusive, especially, for the evolution of particle size and crystal size with milling time [1–3]. Moreover, most of the researchers used high energy and costly ball mills for MA. High-speed mills might lead to contamination/ precipitation and high temperature. L. Kollo et. al. [4] milled Al–1 vol.% nano SiC in a planetary ball mill up to 360 rpm and found that the selection of the process control agent (PCA) has a significant effect on the powder morphology. Micron sized and mm sized powders were obtained when stearic acid and heptane were used respectively as PCA. MA with 10mm dia. balls gave better hardness after hot compaction as compared to 20mm dia. balls. MA of Al (7010) and SiC particles with 2 wt. % stearic acid is done in an attritor mill by Bhaduri et al. [5] at several higher rotating speeds and ball to powder ratios (BPR). X-ray and optical micrographic techniques were used to study morphological and microstructural changes. XRD peaks' intensities were reduced with formation of equiaxed particles. MA of 7075 Al (an alloy of Al, Zn, Mg, Cu) and SiC particulate composite powders was done in a high-energy attritor mill with 2 wt. % of stearic acid used as process control agent by Sankar and Singh [6]. Particles changed from flaky to equiaxed after progressive milling of 12 h. XRD shows the presence of $MgZn_2$ and $CuAl_2$ and absence of Al_4C_3. Angers et al. [7] used a low energy tumbler ball mill to synthesize 2024 Al/SiCp composites. 5 to 35 vol. % SiC in Al was ball milled for 1 to 24 hours. Due to the usage of low energy ball mill, the risk of contamination by the balls and container material is significantly reduced. Superior mechanical properties and homogeneous distribution of SiC particles were achieved for Al-25vol% SiC composite but their ductility was decreased with increased SiCp content. Al-8 vol.% SiC particulate composite was ball milled in an attritor mill at 400 rpm by N. Zhao et. al. [8] under Argon gas and relatively homogeneous distribution of reinforcement particles in the matrix was obtained after 5 h. T. Rostamzadeh and H. R. Shahverdi [1] reported that progressive milling of Al-5vol%SiC powders in a planetary ball mill with 15:1 BPR from 0 to 25 h changes the morphology of the powder particles from flaky to near-spherical and final particle size is much larger. Al-Aqeeli et.al. [2] milled Al-5wt%SiC and Al-20wt%SiC powders in a planetary mill at 200 rpm up to 20h and reported that particle size continuously increased when 5% SiC was taken while for 20% SiC, the size initially increased and then decreased up to 20 h of milling. But the crystal size was shown to decrease continuously till 20h. He also stated that homogeneous distribution of SiC particles in the Al based matrix can be achieved effectively by the MA technique. A. Shokuhfar et. al. [3] milled Al-SiC in a high energy planetary mill and concluded that the crystal size of Al-SiC decreased during the first 12 hours of milling but for further milling up to 24 hours, the crystallite size increased. Ismayadi Ismail et. al. [9] milled some powders up to 48 h in a high energy spex shaker mill and showed that significant changes in crystal size were observed only up to 30h of milling. In the present study, an indigenous multi chamber eccentric ball mill has been designed and fabricated for mechanical alloying of Al-

*Author to whom correspondence should be made, Email: muaz@iitk.ac.in

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SiCp powders [10] at a low but proper speed. Effects of milling time and weight percentage of SiC on evolution of particle morphology, homogeneity of distribution of reinforcement particles in the matrix and internal structure of composite particles produced through MA are studied. In order to determine particle size and shape evolution under different operating conditions, SEM technique has been used. For determining homogeneity of distribution, EDS and X-ray mapping has been used while crystal size determination has been done by XRD technique. This study allows the authors to draw concrete conclusions about the effective usage of a low energy ball mill.

2. EXPERIMENTATION

2.1. Experimental Setup

A multi-chamber eccentric ball mill was designed and fabricated for the synthesis of SiC reinforced Aluminum powders through mechanical alloying. The ball mill was designed according to the experimental requirements where one sample of 10 g powder was to be synthesized in each chamber with a ball to powder ratio (BPR) of 20:1. Each chamber was designed in such a manner that total volume of the balls and the powdered sample remained 35% of volume of each chamber. Another criterion for the design of the ball mill was its critical speed that depended on diameter of cylindrical vial as well as diameter of grinding media. Critical rotational speed of the ball mill calculated by equation (1) was obtained as 135.25 rpm;

$$\mathrm{Nc} = \frac{60}{2\pi}\sqrt{\frac{2g}{D-d}} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

N_c = Critical Rotational Speed, rpm
D = Inner diameter of vial, m
d = Diameter of ball, m
g = Acceleration due to gravity, m/s^2

Three chambers were introduced in this ball mill so that more than one sample at different conditions could be processed at a time. The experimental investigations can be conveniently controlled and economically performed with increased efficiency if conducted in this ball mill. As this ball mill is simply a horizontal ball mill with some design modifications, its cost is very low as compared to other advanced ball mills like attitor mill etc. Pictorial view of the in-house fabricated ball mill set-up is shown in Fig. 1. An application has been filed for issue of Indian patent on 05/05/2015 (File No. 1243/DEL/2015) and published in the journal of patent office on 26/06/2015 [10]. Details of the design of the newly developed ball mill cannot be explained further because issue of final certificate for the grant of the patent is still awaited.

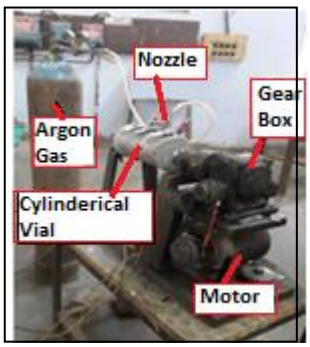

Fig. 1 Ball Mill Setup

2.2. Experimental Conditions

Aluminum (fine powder) of 99.7% purity and 48 μm average particle size was used as base material while silicon carbide particles (400 mesh) was used as reinforcing material. The powders were purchased from Otto Chemie Ltd. Mumbai, India. SEM images of as received powders are shown in Fig. 2. In the experiments, 5wt. % and 15wt. % SiC was mixed with Al powder and then milled for 0h, 5h, 15h and 30h. Process parameters and their levels were selected on the basis of literature survey. About 10 grams of SiC reinforced aluminum powder mixture was used along with stainless steel balls each of which has a diameter of 12.2 mm. The ball to powder ratio was kept fixed as 20:1 throughout the experiments. To avoid any unwanted and excessive cold welding of powder particles amongst themselves, on to the internal surfaces of the milling vial and to the surface of the grinding medium during milling, 1% by weight stearic acid was used as the process control agent. Inert atmosphere was provided into the ball mill chambers by using Argon gas. The mill was rotated at a constant speed of 100 rpm, which was nearly 75% of the critical speed.

2.3. Weighing of Powders

A precise weighing balance (Digital Weighing balance: Readability: 0.01 mg; Make: Precisa; Model No.:ES225SM-DRSCS) was used for accurately weighing of powders.

2.4. Mixing of Powders

Mixing of powders of Al, SiC and Stearic acid in the required percentages was done in a vibratory mixer (FRITSCH Pulvrisette MM-1552) for 10 minutes. Since no grinding media was taken into the mixer, therefore mechanical alloying does not take place during this mixing process.

2.5. Mechanical Alloying of Powders

The mixture of the powders taken from the FRISCH mixer along with stainless steel balls was then filled in the vial of the ball mill. When the vial of the mill rotated at a speed less than its critical speed, the balls first moved upward along with the vial wall and after moving to the proper height, the charge fell down like a projectile. The powder particles were crushed down due to high impact effects of the balls. Additionally, the rolling and sliding played grinding role in the milling process. Mechanical alloying of powders in this ball mill mainly relies on the impact of grinding media. SiC particles being harder in nature got embedded into the Al particles which are relatively soft. During MA, the powder particles experience repeated flattening, cold welding, fracturing, and re-welding of welded particles. After MA, each powder particle becomes composite particle having homogenous distribution of SiC particles in Al matrix. In order to avoid temperature rise in the cylindrical vial, rotation of the ball mill was halted periodically after an interval of 5h. The process is called mechanical alloying even though no solid solution occurs between the phases as in metallurgical alloying. Only embedding of reinforcing particles occurs in the matrix particles. So, by mechanical alloying only composite particles will be obtained.

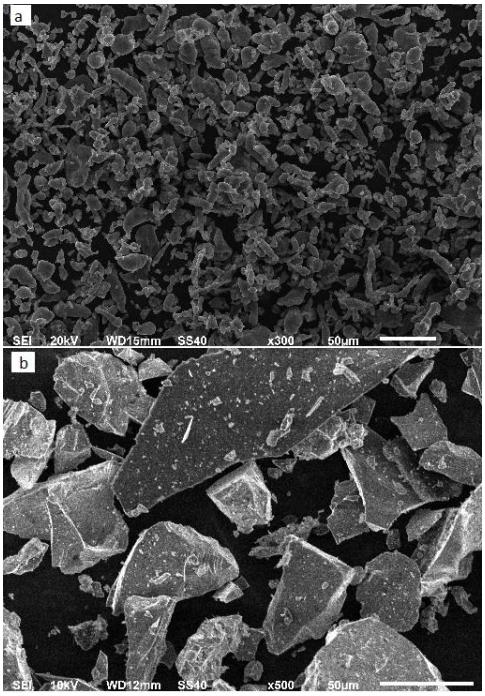

Fig. 2 SEM image of as received (a) Al powder (b) SiC powder

2.6. Characterisation of synthesised samples

The synthesised powder was finally characterized by an X-ray diffractometer (LAbX; XRD 6100, Schimadzu) using a monochromatized X-ray beam with nickel-filtered Cu-Kα radiation in the 2θ range of 10°–80° at a continuous scan mode and a scanning speed of 6 deg/min. The X-rays used for this purpose were generated at voltage of 40 kV and a current of 30 mA. SEM (JSM-6510 LV, JEOL, Tokyo, Japan) was used to study morphology and distribution of particles. An Oxford EDS System (Oxford Instruments plc, Abingdon, UK) attached to the SEM was used for EDS to determine the chemical composition of the powders and for X-ray mapping to determine the elemental homogeneity of the powders. For SEM, EDS and mapping, very small amount of powder was taken and placed on carbon tapes.

3. RESULTS AND DISCUSSION

In this section effects of SiC content and milling time on particle shape, size, homogeneity and crystal size are discussed in details.

3.1. Morphological Evolution

Drastic changes in the shape of particles made it clear that MA could be successfully done in a simple ball mill setup with some modifications in the design but at an appropriate low speed. High energy and costly ball mills with high milling speed are not necessarily required for MA. Fig. 3 shows strong evidence of the fact that in the initial stages of the ball milling process, rate of flattening was lesser than the rate of fracturing and vice versa. Just after 5 hours of milling, all particles attained flake like structure (Fig. 3b) but roundness in shape was increased with increase in milling time. Size reduction observed after 15 hours of milling was small which signified the higher rate of cold welding as compared to the rate of fracturing of particles. Therefore, milling should be continued further to get equilibrium state. After 30 hours of milling, almost all particles became small and round (Fig. 3d). For the case of 5wt%SiC, more crushing and flattening was observed in the present study after 15 hours of milling as compared to that of Al Aqeeli et al.'s findings [2] after 20 hours. T. Rostamzadeh and H. R. Shahverdi [1] milled Al-5vol%SiC powders up to 25 hrs even then final particle size is much larger. These were the reasons why the authors of the present study extended the milling time from 15 hours to 30 hours directly and as was evident from Fig. 3(d), overall particle size was reduced to such a great extent that further reduction could not be expected. The small particles signified that the rate of cold welding and the rate of fracturing became equal and hence it can be concluded that 30 hours of milling is sufficient for proper MA of Al-SiC powders. Therefore, as a whole, particle size of Al-SiC powders first increased until 5 hours of milling and then decreased continuously until 30 hours. After thirty hours, no reduction in size is anticipated.

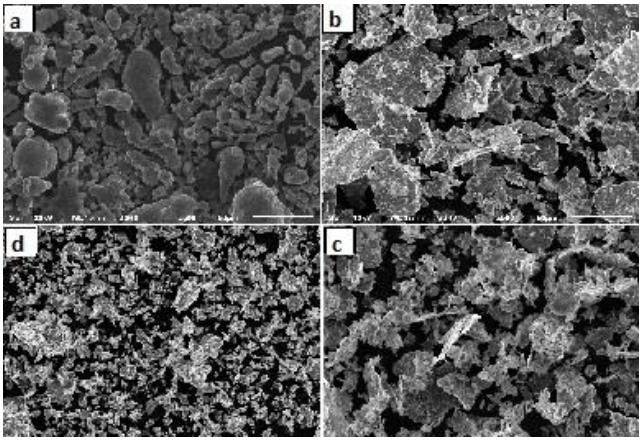

Fig. 3 SEM images for Al-05wt%SiC composite powders (a) before milling (b) after 5 hours of milling (c) after 15 hours of milling (d) after 30 hours of milling

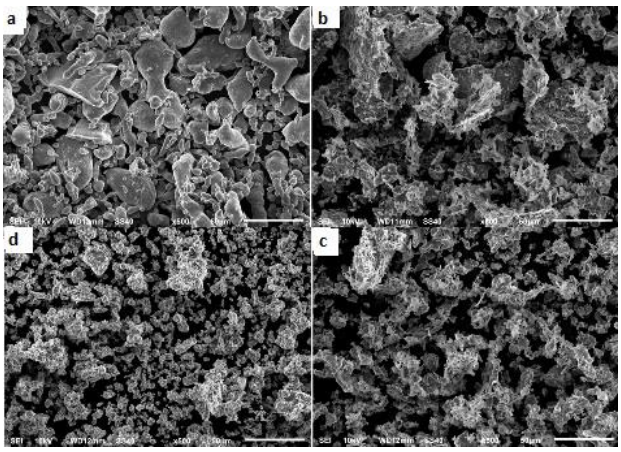

Fig. 4 SEM images for Al-15wt%SiC composite powders (a) before milling (b) after 5 hours of milling (c) after 15 hours of milling (d) after 30 hours of milling

Although the same scenario of cold welding and fracturing was observed for both percentages of SiC content, as depicted from Fig. 3 and Fig. 4 small particles with round shape were obtained when concentration of reinforcement material was increased. This phenomenon occurred due to the ductile nature of Al and brittle nature of SiC particles. SiC particles got embedded into Aluminium particles and layered structures were obtained. These mechanically alloyed particles became brittle and their tendency to fracture was enhanced with increasing SiC content.

Roughness of the surface of the particles increased with increasing milling time for both of the percentages of SiC as shown in Fig. 3 and Fig. 4. Better compatibility was expected with various metallic and polymeric matrices in case of enhanced surface roughness. Therefore, it was encouraging to get the mechanically alloyed particles with enhanced surface roughness. This was supported by other researchers also [11, 12].

3.2. Particle Composition

EDS images (Fig. 5) verified the contamination free milling of powders in the ball mill. This was achieved due to the following two reasons:

i. Inert environment of Argon gas.
ii. Low milling speed.

The temperature rise was low during low speed ball milling which reduces the chance of any phase transformation during the process. Inert environment of an inert gas eliminated the chance of oxide and nitride formation during the MA of composites as also reported elsewhere [2, 12, 13].

X-Ray maps for the powders only mixed for 10 minutes (without ball milling) verified the non-uniform distribution of SiC particles in the sample of un-milled Al-5wt%SiC. Maps for milled powders provided strong evidence for the homogeneity of the two phases of the composite powders. SiC particles got embedded into the Aluminium particles and ensured the presence of SiC particles attached with every Aluminium particle. Same scenario was observed for Al-15wt%SiC. Five hours of milling was sufficient if only homogeneous mixing and alloying was the ultimate aim. Therefore, the MA technique was highly advantageous in achieving a homogeneous distribution of the reinforcement particles in the metal matrix composite as also reported elsewhere [2, 8, 11, 14].

3.3. Microstructural Evolution

Crystal size of the powders at different milling times is calculated by Scherrer's equation [14] as given in equation (2) from broadening of highest intensity XRD peaks.

$$D = \frac{k\lambda}{\beta \cos\theta} \quad \text{.....(2)}$$

where *k(=0.95)* is the proportionality constant; λ (=1.54178 Å) is the X-ray wavelength of Cu-Kα,; β is the full width at half maxima (FWHM) of the XRD peak in radians and θ is the Braggs' angle in degrees.

The minimum crystallite size reached values close to 29 nm when calculated by considering first XRD peak corresponding to Aluminium and 35.59 nm when considering first XRD peak corresponding to SiC in the Al-15wt.%SiC composite. SiC peaks were not detected by XRD in Al-5wt%SiC powders due to the small amount of SiC. Finer crystals would lead to finer grained powder metallurgy products and hence greater strength. Overall reduction in crystal size was much higher when weight fraction of SiC was increased from 5% to 10% by weight (Fig. 6). This demonstrates the role of SiC concentration in refining the microstructure and in achieving small crystal sizes over the same milling period. Another interesting finding was that in the initial stages of milling process, crystal size reduction was observed to be more when SiC content was less. However, the time at which steady state reached increased with increase in SiC concentration.

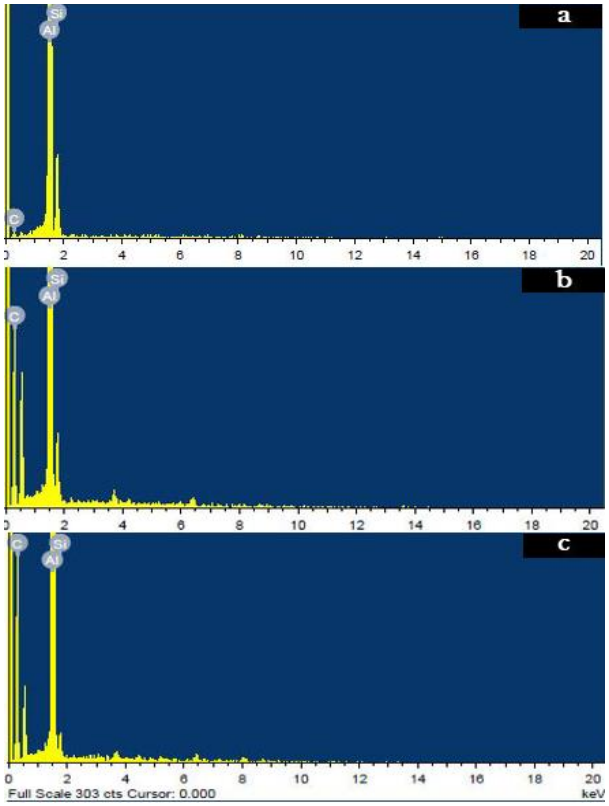

Fig. 5 EDS images of (a) unmilled Al-15wt%SiC (b) Al-15wt% SiC milled for 30 h (c) Al-5wt%SiC milled for 30 h

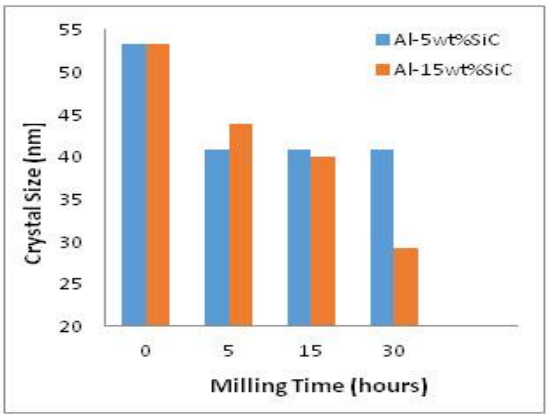

Fig. 6 Crystal size variation with time for different compositions of powders

4 CONCLUSIONS

- A simple horizontal conventional ball mill with some design modifications, developed by the authors can be used for successful mechanical alloying and high cost commercial ball mills may be avoided.
- High content of brittle reinforcement material (15 wt. % SiC) leads to more effective mechanical alloying as compared to 5 wt% SiC.
- Milling time and concentration of reinforcement particles are important parameters for structural evolution.
- Brittle reinforcement particles concentration reduces roundness and increases flatness of the composite particles.

- Inert milling environment, low milling speed and long milling time lead to contamination-free and efficient MA.
- Only a short period of milling (< 5 h) is sufficient to get homogenous composition of both of the phases.
- 30 h of ball milling gives better internal structure, smaller and rounded shaped particles.

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