Experimental analysis of effect of speed and time on aluminium nanoparticles fabricated by high energy ball milling

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Abstract. Metal Matrix Composites are technologically advance material identified in recent era as a substitute for traditional engineering materials because of their excellent properties. Out of the available material, demand for Aluminium Matrix Composites are increasing in the sector of aerospace, automobile and defense applications. The Aluminium Matrix Composites are majorly accepted because of their excellent physical and mechanical properties. There are various manufacturing techniques developed for the fabrication of AMCs. High energy ball milling is a one of the most effective and versatile routes for fabrication of nano aluminium powder. The Al powder of 99.55% purity was taken for experimentation. The 60gm of Al powder was taken in stainless steel vial with BPR as 5:1. The stainless steel balls of diameter 4mm was used as a grinding media. The ball milling was performed at different speed and time in presence of 60ml toluene as process control agent. The characterization of powder particle was done with XRD and SEM. The analysis of peak were done on origin software. The optimum speed and milling time for fabrication of Al nanoparticles of size 36 nm were found to be at 250rpm and 3 to 6 hours.

Keywords: Aluminium, High Energy Ball Milling, X-Ray diffraction, Scanning Electron Microscope, Particle size

1. Introduction
The nano scale technology is very young and key technology and having potential application in the field of engineering, cosmetics, food product, textiles, biomedicine and domestic applications. Many research has adopted various methods of manufacturing out of that mechanical alloying is the best suitable method, which is also known as high energy ball milling. The main purpose of mechanical alloying is change the shape of particles, reduce particle size by continuous cold welding and fracturing. To achieve this it is require to use ductile material for MA process [2]. The aluminium (Al) and its alloys are frequently examined material because of its exceptional properties like corrosion resistance, low density and high strength to weight ratio. The ultrafine metallic powder have reported a wide applications in engineering because of large specific area and high reactivity and having particle size in nm [12]. A substantial improvement in mechanical properties are reported with particle size reaches to nanometer [4]. There are basically two terms associated with in high energy ball milling which are Mechanical Alloying and Mechanical Milling. Mechanical Alloying (MA) indicates mixture of one or two metal particles
(different metal) are milled together in ball milling machine to obtain the homogenous alloy. Mechanical milling is associated with milling of pure metals, intermetallics or prealloyed powders where homogenization is not an important factor. Mechanical milling is used to reduce size of metal particles [3]. The main difference between mechanical alloying and milling are that the time required for milling is half than the alloying also intention of milling is to reduce the particle size or transformation of particle by mechanical means [1].

The various types of high energy ball millings (HEBMs) are available for manufacturing of nano crystalline and amorphous powder having different capacity, efficiency and controlled conditions with cooling arrangement to maintain the temperature of process. The widely used HEBMs for production of Al alloys are attritor, planetary, and Spex shaker mills. Attritators are used for manufacturing of larger quantities upto 100 Kg. The planetary mills are used for milling volume with variety of options. In planetary mill input energy can be changed by changing speed of mill upto 1000 rpm. Spex shakers ball mills are used for small quantities in a very short time. In this experimental work, researcher was used planetary ball mill. [11].The high energy Planetary ball milling consist of a container (Vial), lid, Supporting disc, grinding balls, material to be grind. The container is kept on rotating supporting disc, both are rotating opposite to each other as these are installed on planet like drive mechanism. Hence it is called as planetary ball mill. The centrifugal force developed by vial and supporting disc acts on the vial content. Due to rotation, grinding balls collide with each other and container wall. The small amount of powder trapped between grinding balls, wall and grinding balls. Due to this action, morphology of powder particle could change [11].

In this work, high energy ball milling of Al powder performed at different speed and time in presence of process control agent (PCA). When liquid is used during grinding, process is known as wet grinding [3]. The XRD and SEM analysis of ball milled Al powder has been performed to know the present of carbide and oxide phase and to reveal the change in morphology of Al powder after the ball milling [7] [14].

The characterization of nano materials is very important from application point of view by understanding its properties. The performance of composites changes as the size of particle changes. Different methods are available to reveal the characteristics of materials.

Characterization Techniques:
1) Chemical Characterization
2) Structural Characterization

Chemical characterization is performed to internal chemical structural details and to know shape, size, crystallinity and lattice constant structural characterization is used. In this work, for characterization of ball milled aluminium powder, structural characterization method is adopted. In structural characterization, X-Ray diffraction and Scanning Electron Microscopy (SEM) are used.

2. X-Ray Diffraction Technique

X-ray diffraction is a non-destructive technique which gives very important information about lattice structure of a crystalline structure like unit cell dimensions, bond angle, chemical compositions and crystallographic structure of natural and fabricated materials. The XRD machine works on well-known Bragg’s Law ($\lambda = 2dsin\theta$)  

$$\lambda = 2dsin\theta$$  \hspace{1cm} (1)

where $\lambda$ is the wavelength of incident X-ray, $d$= interplanar spacing and $\theta$ is the angle of incidence. In this case X-ray source is used for identification and characterization of crystalline solids. It is used to obtain the average size of the particles using Scherrer formula:

$$a = 0.9 \lambda/Bcos\theta$$  \hspace{1cm} (2)

where $a$= average particle size, $\lambda$ is the wave length of incident X-ray, $B$= full width of half maxima in radians and $\theta$ is the angle of incidence [10] [5].
The XRD experimentation was conducted at UGC-DAE Consortium for Scientific Research, Indore on the D8 advanced XRD machine.

3. Scanning Electron Microscopy
In case of scanning electron microscope, develops the image by scanning the sample surface with high-energy beam of electron. The electron beam scanned over the surface of sample to measure the electrons that are scattered back. The high energy electron interact with atoms on the surface which producing signal that contains the surface topography of the sample [13]. The SE M analysis were conducted at UGC-DAE Consortium for Scientific Research, Indore on JEOL Scanning Electron Microscope.

4. Experimental Detail
In this study Aluminium is selected as matrix material as it is emerged as important material for industrial and commercial applications. Aluminium possesses excellent properties, light in weight, strength is equal to construction steel, easy to form into rolled sheet. Here aluminium powder is mixture of spherical and elongated shape particle having average diameter of 23.99 micron and average length of 31.38 micron. The Al powder is received from PASHWAMANI METALS, Mumbai having purity of 99.55% and particle size of 20 micron. The selection of process parameters namely ball milling speed (rpm), ball milling time (hrs) and CNT diameter (nm) were made on the basis of literature survey [6]. Three process parameters are hence selected to conduct this experiment namely ball milling speed (rpm), ball milling duration (hours) and CNT diameter (nm). Table 1 indicates the process parameters and the values for the same.

| Factors                  | LEVELS |                 |             |             |
|-------------------------|--------|-----------------|-------------|-------------|
|                         | LOW    | MEDIUM          | HIGH        |             |
| Ball milling speed (rpm) | 150    | 200             | 250         |             |
| Ball milling time (hours)| 3      | 6               | 12          |             |
| CNT diameter (nm)       | 8      | 20              | 50          |             |

An L9 orthogonal array was chosen for 3 factors and 3 levels with the primary intention of optimizing the number of experiments. The Minitab 17 software was used to carry out the design of experiments. Table 2 illustrates the final design of experiments using the L9 orthogonal array.

| Expt. No. | Control Factors |
|-----------|-----------------|
|           | S (rpm) | T (hours) | D (nm) |
| 1         | 150      | 3         | 8      |
| 2         | 150      | 6         | 20     |
| 3         | 150      | 12        | 50     |
| 4         | 200      | 3         | 50     |
| 5         | 200      | 6         | 8      |
| 6         | 200      | 12        | 20     |
| 7         | 250      | 3         | 20     |
| 8         | 250      | 6         | 50     |
| 9         | 250      | 12        | 8      |

With above parameters, ball milling of Al powder was performed in FRITSCH Pulverisette 6 make planetary ball milling machine in 250ml stainless steel jar with stainless steel balls. The weight of each ball was 4g and ball to powder ration was taken as 5:1 during milling operation. The dwell time was
takes as 15 minutes after 1 hour and reversal of rotation after each hour so that powder is milled properly. After rotation of 1 hour, ball milling is given rest for 15 minutes in order to avoid rapid increase in temperature during milling operation. The toluene (C6H5CH3) was used as a process control agent to control the sudden rise in temperature inside the vial during milling process. The 60ml toluene quantity was taken for wet milling. The purpose of PCA are to control the cold welding of powder to ball and vial. The XRD diffraction pattern was recorded from 20° to 90° on D8 advance XRD system having 2.2Kw sealed tube of Cu as X-ray source.

5. Results and Discussions
5.1 Characterization of Powder:
The characterization of raw and ball milled Al powder were done by XRD and SEM to determine the changes in shape and size of Al particles. The aluminium powder were ball milled at 150, 200 and 250rpm for 3,6,12 hrs. The crystalline nature of milled aluminium powder was examined by using XRD technique. Fig. 3 shows XRD patterns of the raw and ball milled aluminium powder for different time and speed reveals that there was an appreciable change in particle size of aluminium powder. The average particle size for raw and ball milled aluminium powder were estimated by Scherrer formula equation(1).

The crystallite size for ball milled sample considered for study is given in table 3. The fig.1 shows the X-ray diffraction pattern of raw aluminium and ball milled aluminium powder prepared by high energy ball milling in planetary ball milling at different time and rpm. It was observed that raw aluminium powder has an average particle size of 44.35 nm estimated by Scherrer formula. The fig. 1 XRD pattern of aluminium powder before and after milling of aluminium powder at 150 rpm, 200rpm and 250rpm corresponding to 3,6 and12 hrs and enlarged section of first (Largest) peak of XRD. There are four sharp peaks detected corresponds to FCC structure of aluminium. The higher intensity of peak in Fig. 1(a) shows aluminium powder is more crystalline in nature and after ball milling, reduction in peak intensity followed by broadening of peak might be due to fine size of aluminium powder [9]. Compared to the pattern of raw aluminium powder, the milled powder shows broaden intensity peaks, indicates that powder size is effectively reduced [8]. The small shift observed in enlarged section of stacked largest peak is due to macrostrain in lattice. The change in crystallite size at particular rpm and hours is due to lattice strain and ductile nature of aluminium responsible for cold welding and deformation of aluminium particles. From fig. 2 it is found that with increase in RPM and milling time, there is a reduction in particle size. But at certain stages, there is increase in particle size due to the agglomeration, subsequent cold welding and fracture.

Table 3. Particle size of Aluminum nano-powder by Debye-Scherrer formula and particle size by Image J software from SEM images.

| Sample               | RPM | Hrs | Crystallite Size [nm] | Particle Size [μm] by Image J |
|----------------------|-----|-----|-----------------------|------------------------------|
| Raw Aluminium        | 150 | 3   | 52.64                 | 78.24                        |
|                      |     | 6   | 54.61                 | 63.87                        |
|                      |     | 12  | 51.01                 | 71.20                        |
| Ball Milled Aluminium| 200 | 3   | 61.69                 | 70.42                        |
| Powder               | 12  | 6   | 43.82                 | 165.06                       |
|                      |     | 3   | 42.13                 | 227.20                       |
|                      |     | 12  | 36.84                 | 128.32                       |
|                      | 250 | 6   | 36.03                 | 446.36                       |
|                      |     | 12  | 43.45                 | 333.90                       |
Figure (a)

Figure (b)

Figure (c)
Figure 1. X-ray Diffraction pattern of Raw and Ball Milled Aluminium Powder by High Energy Ball Milling. (a) Raw Aluminium Powder (b) 150 rpm at 3,6,12 hours (c) 200 rpm at 3,6,12 hours (c) 250 rpm at 3,6,12 hours and enlarged section of stacked largest peak.

Figure 2. Variation of Crystallite size versus milling time (hours) for Ball Milled Aluminium Powder.
(a) SEM images of raw aluminium power

(b) At 150 RPM 3 Hours

(c) At 150 RPM 6 Hours

(d) At 150 RPM 12 Hours
(e) At 200 RPM 3 Hours

(f) At 200 RPM 6 Hours

(g) At 200 RPM 12 Hours

(h) At 250 RPM 3 Hours
At 250 RPM 6 Hours

At 250 RPM 12 Hours

**Figure 3.** SEM images of Aluminium powder (a) Raw Aluminium Powder. At 150 RPM (b) 3 Hours (c) 6 Hours (d) 12 Hours. At 200 RPM (e) 3 Hours (f) 6 Hours (g) 12 Hours. At 250 RPM (h) 3 Hours (i) 6 Hours (j) 12 Hours.

Fig. 3 shows SEM images of raw aluminium powder and ball milled aluminium powder at 150, 200 & 250 RPM corresponding to 3, 6, 12 hours of milling time. The SEM analysis of raw aluminium powder has been done to check the surface morphology and shape of particles. From Figure (a) it is observed that aluminium particles are spherical and elongated in shape has an average grain size is 20 micron. The SEM analysis of ball milled aluminium powder has been done to check the change in morphology and flatness of the powder.

The initial powder size was 20 micron and after the ball milling particle size increases to 52 micron at 150 rpm 3 hours. Again slightly increases at 6 hours and further decreases to 51 micron. The changes were occurred due to due to welding of aluminium powder and fracture mechanism were less than agglomeration. Because of this, at 200 rpm 6 hours and 250 rpm 6 hours, there were sudden increase in particle size (measured on Image J software) and same time aluminium powder particle surface became flaky and fracture mechanism initiated as shown in fig 3 (i) and (j).

**6. Conclusion**

Aluminium matrix composites have wide applications in engineering and high energy ball milling is a most popular method of production of aluminium nanoparticles of low crystallite size 36nm. The morphology of ball milled aluminium powder is studied by XRD technique and Scanning Electron Microscope (SEM). The particle size is calculated by X-ray diffraction technique and the change in morphology is analyzed through SEM. At 3hrs and 6hrs, with further increase in rpm, crystallite size decreases. At 12 hrs, with increase in rpm, crystallite size decreases at 200 rpm and further increases at 250 rpm. This is due to cold welding phenomenon. Also from SEM analysis it is observed that at 200 rpm 12 hrs and 250 rpm 6 hrs, particle became flaky as increase in surface area of particles.
12hrs facture mechanism is initiated and with further increase in time there may be decrease in particle size.

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