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Mechanical milling of aluminum powder using planetary ball milling process

M. Ramezani*, T. Neitzert
Centre of Advanced Manufacturing Technologies (CAMTEC), School of Engineering, Auckland University of Technology, Auckland, New Zealand
* Corresponding e-mail address: maziar.ramezani@aut.ac.nz

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ABSTRACT

Mechanical alloying has recently attracted considerable attention as researchers strive to enhance nanocomposite properties and extend their utility. The process can be performed at room temperature and homogeneous nanocomposite powders can be produced. In this paper, we investigated the effect of different ball milling parameters and operating conditions (milling time, ball size, processing control agent (PCA) and speed) in mechanical alloying of aluminum powder to achieve particle size reduction with less contamination. Two types of PCA, i.e. stearic acid and methanol have been used and microstructure evolutions at different operating conditions were studied. It was shown that the optimized milling parameters for aluminium composite are 100 stainless steel ball (10 mm), 200 rpm rotation speed with direction reversal and 1 min pause time after every 15 min running time, under argon gas for 30 hr of milling.
Keywords: Aluminum powder; Mechanical milling; Methanol; Microstructure; Milling parameters; Stearic acid

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1. Introduction

Mechanical alloying (MA) is a high energy ball milling process by which constituent powders are repeatedly deformed, fractured and welded by grinding media to form a homogeneous alloyed microstructure or uniformly dispersed particulates in a matrix (Suryanarayana, 2001). The main objectives of the milling process are to reduce the particle sizes (breaking down the material), mixing, blending and particle shaping. The process requires at least one fairly ductile metal (e.g. aluminium) to act as a host or binder. The major process in MA for producing quality powders of alloys and compounds with well-controlled microstructure and morphology is the repeated welding, fracture, and rewelding of the reactant mixed powders (Benjamin, 1992). It is critical to establish a balance between fracturing and cold welding in order to mechanically alloy successfully.

Two techniques are proposed to reduce cold welding and promote fracturing. The first technique is to modify the surface of the deforming particles by adding a suitable processing control agent (PCA) that impedes the clean metal to metal contact necessary for cold welding (wet milling). The second technique is to modify the deformation mode of the powder particles so that they are able to deform to the large compressive strains necessary for flattening and cold welding. (Zehetbauer and Zhu, 2009).

Whenever two grinding balls collide, a small amount of powder is trapped in between them. Typically, around 1000 particles with an aggregate weight of about 0.2 mg are trapped during each collision (White, 1979). During this process, the powder morphology could be modified. For the soft powders like aluminium, the flattened layers overlap and form cold welds. This leads to formation of layered composite powder particles consisting of various combinations of the starting ingredients (Schwarz and Koch, 1986). The work-hardened elements or composite powder particles may fracture at the same time. These competing events of cold welding (with plastic deformation and agglomeration) and fracturing (size reduction) continues

repeatedly throughout the milling period. Finally, a refined and homogenized microstructure can be obtained and the composition of the powder particles is the same as the proportion of the starting constituent powders (El-Eskandarany, 2001). Along with the cold welding event described above, some powders may also coat the grinding medium and/or the inner walls of the container. A thin layer of the coating is beneficial in preventing wear-and-tear of the grinding medium and also in preventing contamination of the milled powder with the debris.

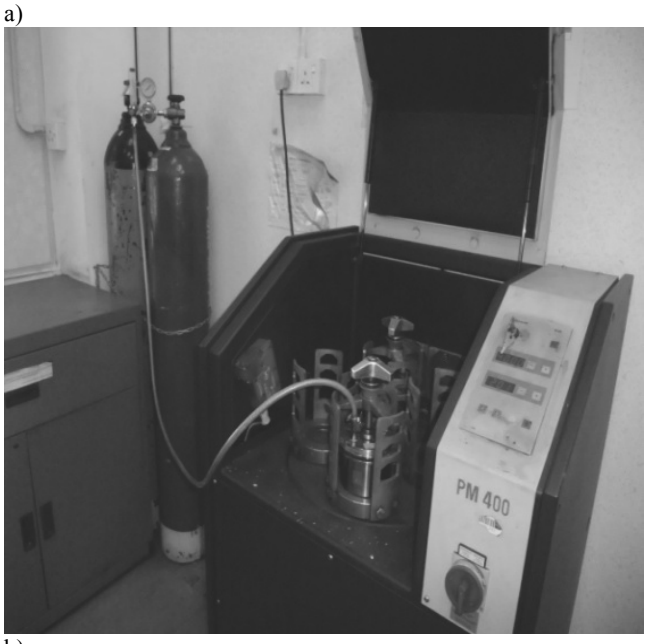
Several researches have been carried out previously to study the mechanical alloying. Calkaet et al. (2005) applied electric discharge assisted milling and conventional mechano-synthesis techniques to investigate the effects of milling conditions on the fracture and agglomeration of amorphous CoSiB ribbons produced by planar flow casting. Książek et al. (2007) investigated the kinetics of thermal decomposition of cadmium carbonate and basic zinc carbonate and the effect of intensive milling in a planetary ball mill on its parameters. Kakuk et al. (2009) modelled the milling process in a planetary ball mill and found a relationship between the angular velocity of the sundisk and the vials, and the geometrical parameters of the mill. Farahbakhsh et al. (2011) investigated the mechanical alloying of the copper powder and the formation of Ni-Cu solid solution coating on the Ni balls. Wang and Wang (2011) studied the microstructure evolution of high energy milled Al-50 wt% Si alloy during heat treatment at different temperatures. Md Rasib and Hussain (2011) carried out mechanical alloying of Fe-NbC composite at different milling speeds. Patel and Morsi (2012) studied the influence of mechanical alloying of Al-8Sn-5Mg elemental powders on the microstructure and properties of the sintered product.

None of the papers mentioned above studied the effect of operating conditions on the MA process. The MA process is affected by several factors that are playing very important roles in the fabrication of homogeneous materials. It is well known that the properties of the milled powders of the final product, such as the particle size distribution, morphology and the degree of disorder, depend on the milling conditions and, as such, the more complete control and monitoring of the milling conditions, the better end product is obtained. The aim of this paper is to determine the effect of different ball milling parameters and operating conditions (milling time, ball size, PCA and speed) in Al matrix to achieve particle size reduction.

2. Experimental details

Planetary ball milling is carried out for fabrication of engineering materials via a mechanical alloying process. In planetary ball milling, the milling media contains considerably high energy, as the milling stock and balls come off the inner wall of the vial (milling bowl) and the effective centrifugal force can reach up to twenty times of gravitational acceleration. The centrifugal forces caused by the rotation of the supporting disc and autonomous turning of the vial act on the milling charge (balls and powders). Since the turning directions of the supporting disc and the vial are opposite, the centrifugal forces alternately are sync hronized and opposite. Therefore, the milling media and the charged powders alternatively roll on the inner wall of the vial, and are lifted and thrown off across the bowl at high speed. One advantage of this type of mill is the ease of handling the vials (45 ml to 500 ml in volume) inside a glove box.

a)



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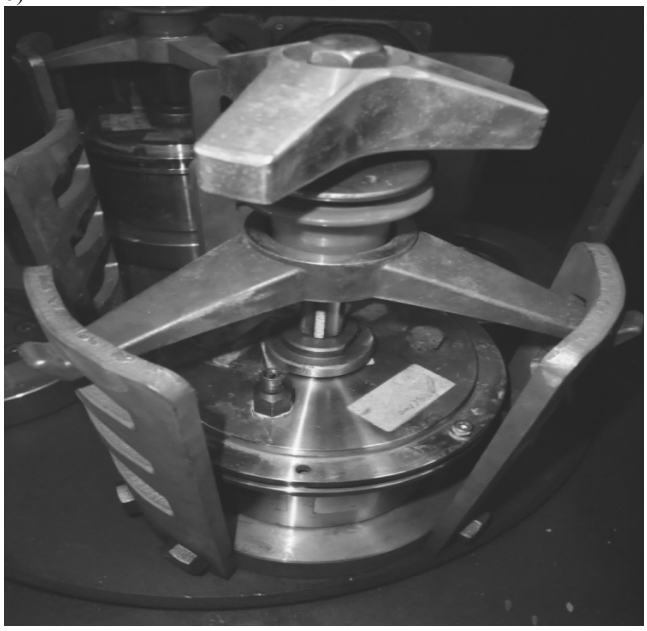


Fig. 1. Retsch PM400 (a), and 500 ml jar single station (b)

The aluminium powder used in this research has a purity of 99%. The particle size is 90%, 31.97 µm and is flaky in shape. In this study stearic acid ($C_{18}H_{36}O_2$ or CH_3 $(CH_2)_{16}COOH$) and methanol (CH_3OH) with purity of 99.99% are used as process control agent to avoid cold welding.

Table 1.
Summary of milling parameters

Time	Ball No.	Ball size (mm)	Stearic acid	Methanol	Speed (rpm)	Ratio (B/P)
0.5 hr	100	10	-	-	200	10
1 hr	100	10	-	-	200	10
3 hr	100	10	-	-	200	10
5 hr	100	10	2 wt%	-	200	10
7.5 hr	100	10	0-3 wt%	3.3 ml	200	10
10 hr	100	10	2 wt%		200	10
15 hr	100.31	10, 15, 20	2 wt%	3.3 ml	200.250	10
20 hr	100	10	2 wt%	-	200	10
25 hr	100.31	10, 15, 20	2 wt%	-	200	10
30 hr	100.31	10, 15, 20	2 wt%	3.3 ml	200.250	10
40 hr	100.31	10, 15, 20	2 wt%	3.3 ml	200.250	10

First, 40 g pure aluminium was ball milled using 500 ml stainless steel mixing planetary ball mill Retsch 400M. The device is shown in Fig. 1 and has 110 mm inner diameter and 110 mm height, effective sun wheel diameter of 300mm, speed ratio of 1: 2.5, interval operation with direction reversal and 1 min pause time after each 15 min running time. The jars were ran under argon gas and then were agitated with 3 different stainless steel (made of the same material as the grinding jar) milling balls size of 10, 15 and 20 mm at two different rotation speeds of 200 and 250 rpm. Two different ball numbers of 31 and 100 were used for varying milling times up to 40 hr. Stearic acid [$CH_3(CH_2)_{16}COOH$] of 0, 1, 2, and 3 wt% and 3.33 ml methanol [CH_3OH] were added as a process control agent separately and giving a ball to powder weight (B/P) ratio of 10:1 to reach a fine powder particle size regarding to powder morphology and to minimize cold welding of the aluminium particles. In addition, the PCA was used to prevent powders from sticking to the balls and the jar wall.

Mixing was carefully controlled under inert gas and afterward was transferred to the glove box to avoid impurities. Impurities in the sample influence the physical, interface reaction and mechanical properties of milled aluminium. The milling parameters are summarized in Table 1.

In this research, particle size and distributions of powder were determined using a CILAS 1190 instrument with a measurement range from 0.04 to 2,500 µm to determine the effect of milling parameters on feeding martial size. A field emission scanning electron microscope (FESEM) model LEO supra 55 with up to 40 nA probe current was used to observe the powder morphology and contamination of the powder. XRD characterization of the powders was performed with a Bruker type using CuKα radiation at 4.8 kW to define the essential quality and crystallography of the existing elements in aluminium.

Thermal and oxidation stability of materials and phase transformation during the mechanical milling process were studied with a thermo gravimetric analyser (TA instruments SDTQ600) to specify any change in melting point of aluminium powder after each time of setting new milling parameters.

3. Results and discussions

The raw material characterization involves the determination of size, particle distribution, and morphology of aluminium powder that was characterized by SEM, EDX, laser particle measurement, XRD and DSC.

It was found that aluminium powder has an average particle size of 31.97 µm as illustrated in Fig. 2. Due to the wide range of particle size of aluminium powder, milling process is necessary to get a homogenous particle size. Fig. 3 illustrates XRD analysis of milled aluminium powder for 7.5-40 hr which is used in this research. Five peaks can be observed at 2υ=38.5, 44.9, 65, 78 and 82.5°. The observed lattice peaks of Al are very close to the reference value of 0.40 nm. Compared to the pattern of the starting powders, the milled powders show a broadened intensity which means the powder size is effectively reduced after milling. The DSC machine proved the existance of pure aluminium powder (9.171 mg) with using Pt pan and 10°C/min temperature rate for 2 hr and 40 min. The melting point of unmilled and 7.5-40 hr milled aluminium are almost the same (~660°C) and milling did not have a major effect on the DSC analysis which could be useful for controlling milling temperature to avoid any phase changing or carbide formation during powder preparation.

In this project, the main milling parameters were changed to reach minimized aluminium matrix particles size and ameliorative aluminium particle morphology. Achieving smaller particle sizes with fine distribution is one of the challenges in milling of aluminium. On the other hand, trying to shift particle morphology from flake to spherical with keeping a minimized particle size is

another aim of this project. Having a spherical shape and as small as possible particle size would be advantageous for metal matrix composites and sintering processes.

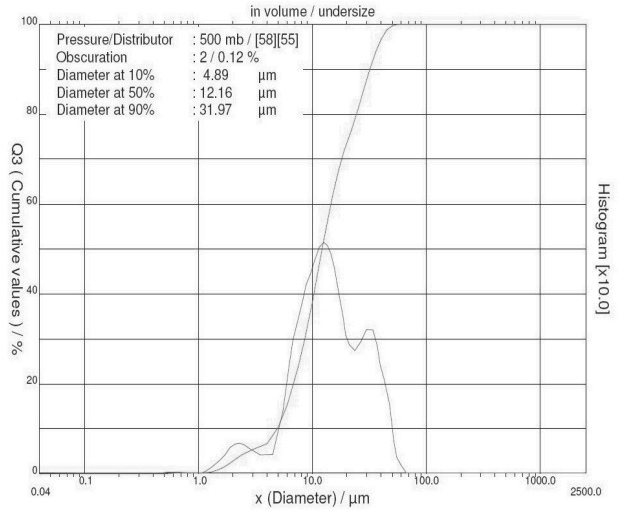


Fig. 2. Particle analyser graph (CILAS 11900) of pure aluminium

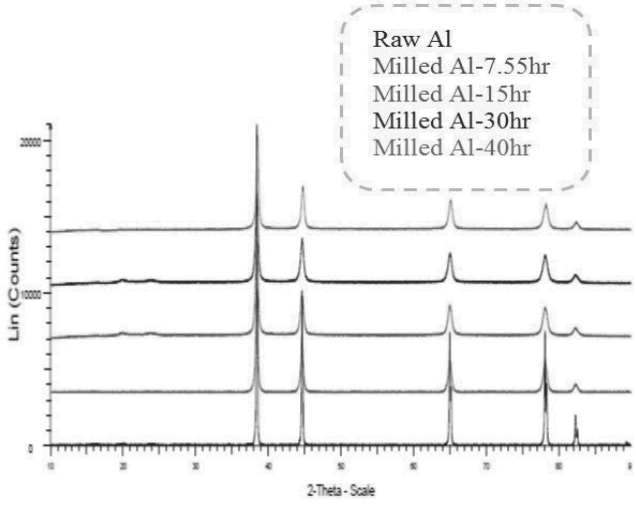


Fig. 3. X-Ray diffraction (XRD) pattern of unmilled and milled aluminium powder

3.1. Role of inert gas in MA

The aluminium particles were milled for 0.5-3 hr with 100 balls (10 mm) at 200 rpm rotation speed, without a processing control agent. Argon gas was applied to aluminium powder and the result was shifting the morphology of particles from flake to spherical shape and an increase in particle size, welding and agglomeration with a rough surface up to ~1.5 mm after 1 hr milling. The exception was observed after 3 hr of milling when particle size was reduced due to higher impact forces and temperature inside the jar that forced particle towards higher density, lower porosity and closer to a spherical shape.

The level of contamination was monitored by EDX analysis which did not show a high rate of contamination level for 0.5-3 hr milling. Fig. 4 shows the changes in aluminium particle size for 0.5-3 hr of milling time without using argon gas. It should be noted that for milling of aluminium power without argon gas, powder was melted and stuck to the jar wall even after 30 min of milling therefore for further optimization parameters, argon gas was applied in all subsequent milling processes.

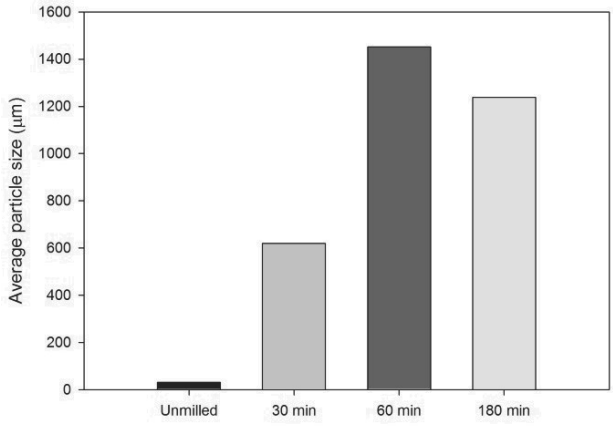


Fig. 4. Laser particle analyser for unmilled and 0.5-3 hr milled aluminium without argon and PCA

3.2. Role of milling time in MA

In this study, 2 wt% of stearic acid as a PCA, 200 rpm rotation speed and 100 balls (10 mm) were applied, subsequently milling times were increased from 5 up to 40 hr (5, 7.5, 10, 15, 20, 30 and 40 hr accordingly) to see the effect of milling time on aluminium powder shape, size and contamination level and the results are shown in Fig. 5.

The aluminium matrix looks differently when milling times were changed. Fig. 6 shows that the main particle size was increasing from ~31µm to ~90 µm after 5 hr of milling as the aluminium powder welding mechanism and agglomerate were higher than fracturing. It was decreasing until 15 hr of milling (~70 µm) due to the ball impact and friction and again increasing continuously up to 40 hr of milling (215 µm).

The reason for this behaviour is that during the milling process the temperature is going up very fast so after a certain temperature PCA was not able to control the cold welding and it seems that warm welding occurred (depending on the powder melting point). Even at this stage, the impact energy was not enough to break down the particle (sometimes the temperature could reach up to 500°C and pressure reach up to 500 kPa). Generally, in this project the particle size and shape were considered as the most important factors of an aluminium matrix although contamination (O, Fe and C) and phase changing were investigated by EDX as well.

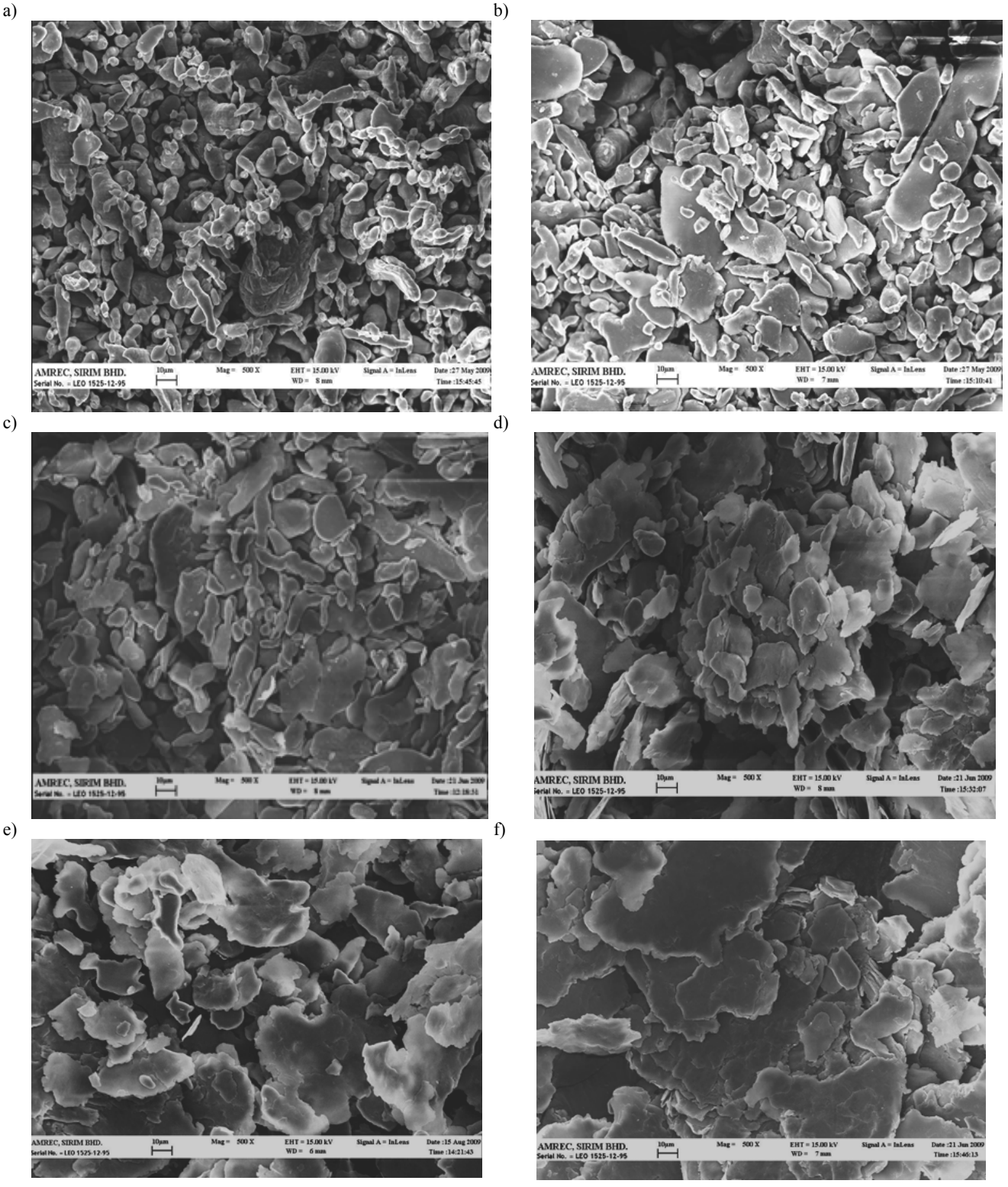


Fig. 5. SEM micrograph for: a) unmilled pure aluminium, b) 5 hr milled aluminium powder, c) 10 hr milled aluminium powder, d) 20 hr milled aluminium powder, e) 30 hr milled aluminium powder, f) 40 hr milled aluminium powder

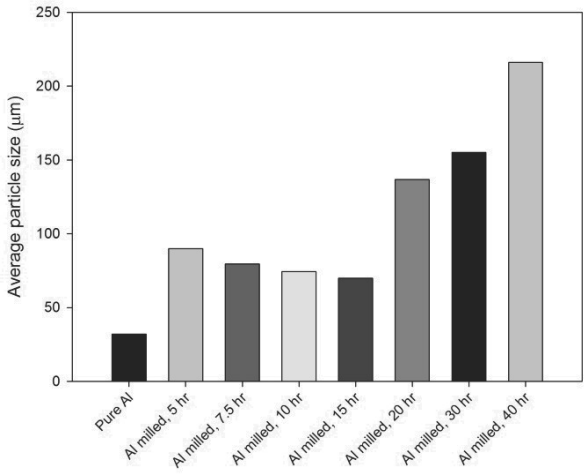


Fig. 6. Laser particle analyser results of pure and milled aluminium in different milling times

3.3. Role of PCA in MA

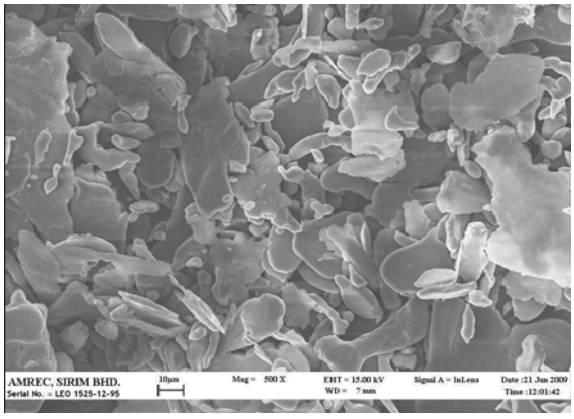
At 7.5 hr of milling, 1-3% of the stearic acid with 100 balls (10 mm) and 200 rpm speed were added. Fig. 7 and Fig. 8 show the results from the morphology analysis. It can be seen that in the case without PCA, the aluminium particles started welding together and forming large particles with a rough surface, where some particle size approached ~500 µm (where the impact energy of the balls formed a flakes shape). The particle size of ~90 µm could be achieved when milling was continued under 3 wt% PCA. In fact, without adding any amount of PCA one could get a more spherical shape or convert flake shape of original powder to spherical which has better features for sintering rather than flake shape, but achieving smaller particle size becomes a challenge. The option would be either to prepare the aluminium powder in both bigger particle size and closer to spherical shape or fine particle but flake in shape to be used as metal matrix composites for higher diffusion rates in powder consolidation applications.

Fig. 8 shows that after 7.5 hr of milling time the particle size became ~500 µm increasing from ~31 µm. Then, 1-3% of PCA (stearic acid) were added to change the fracture mechanism to cold welding. After adding 3 wt% of PCA, a particle size of ~72 µm was achieved and morphology was changed from flake to a more equiaxed shape with lower yield.

3.4. Role of ball size, ball number and rotation speed in MA

For a further investigation of mechanical milling parameters, the stainless steel ball size was changed from 10 mm (100 balls) to 10, 15 and 20 mm (altogether 31 balls), (different ball-DB) with B/P ratio (10:1) ; subsequently the rotation speed was also changed from 200 to 250 rpm (different speed-DS) and finally, both ball size and rotation speed were changed (DB-DS) with inclusion of 2 wt% stearic acid as PCA. Using a total number of 31 balls with 10, 15 and 20 mm ball size and 250 rpm rotation speed causes an increase in ball impact energy, however, it is accrued as the temperature inside the jar increases as well. As a result, after 15 hr of milling, the particle size reached up to ~220 µm and after 30 hr of milling, the particle size was increased to 260 µm (see Fig. 9 and Fig. 10) and the morphology changed to agglomerated particles.

a)



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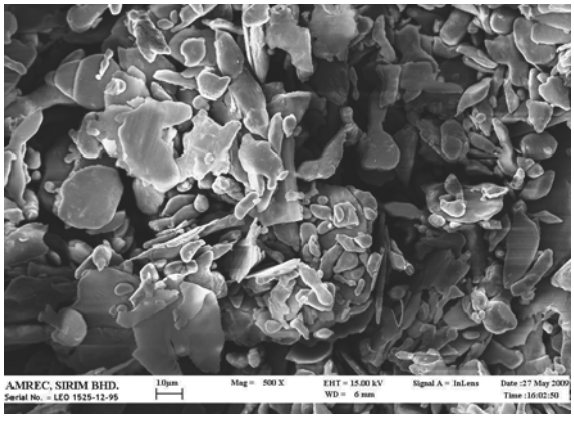

c)

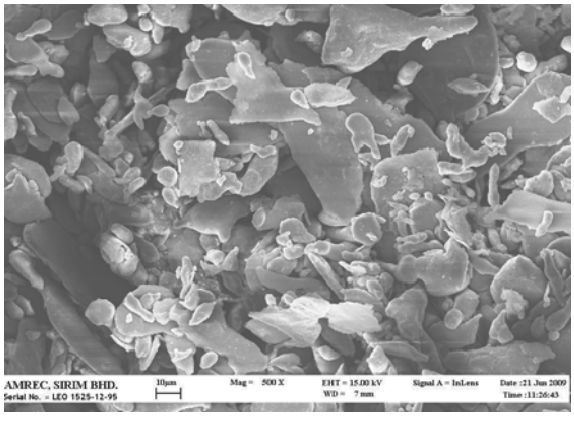

Fig. 7. SEM micrograph of 7.5 hr milled aluminium with: a) 1 wt% of stearic acid, b) 2 wt% of stearic acid, c) 3 wt% of stearic acid

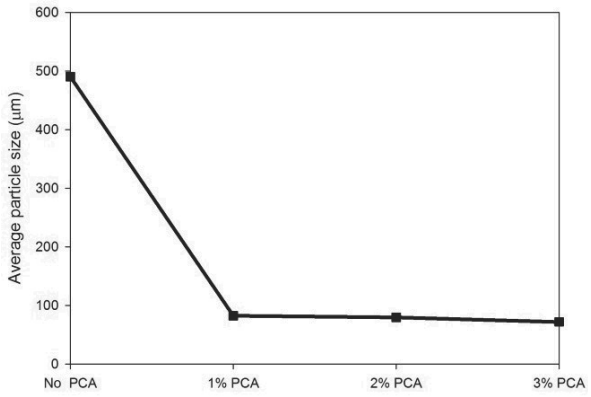

Fig. 8. Laser particle analyser results of 7.5 hr milled aluminium with 0 to 3 wt% of stearic acid

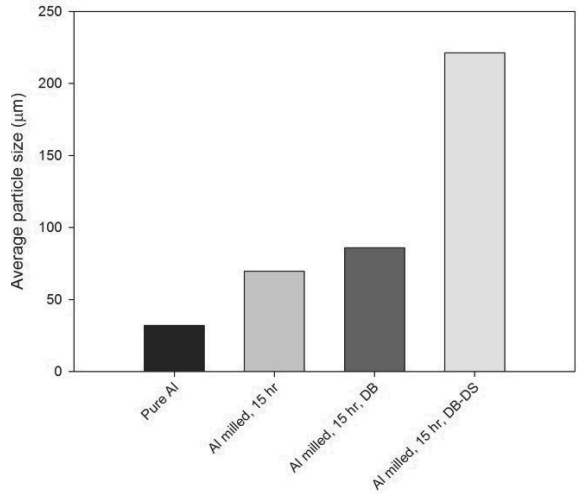

Fig. 9. Laser particle analyser results of 15 hr milled Al with different ball size (10, 15, 20 mm), ball no. (31) and rotation speed (250 rpm)

Fig. 11 shows that only after 40 hr of milling, the particle size has decreased due to a double break and fracture of half melted particles caused by very high impact energy. At this stage, the contamination level was high (Fe and O) with EDX scans of the specimens and powder morphology was not homogenous (mixing of flake and agglomerated particles). The powder sizes obtained for the aluminium (above 250 µm) can be so large to exclude any possibility of sintering. One way to control this effect is t hrough the changing of a process control agent (PCA) and decreasing the ball energy impact.

3.5. Role of changing PCA in MA

The milling process was carried out up to 40 hr (7.5, 15, 30 and 40 hr) with 200 rpm speed, 100 balls (10 mm) and 3.3 ml methanol as a PCA in order to examine the morphology, particle size and contamination of the aluminium matrix (see Fig. 12 and Fig. 13). For aluminium powder, methanol was used as a new process control agent and a more effective technique to create a breaking mechanism to avoid cold welding.

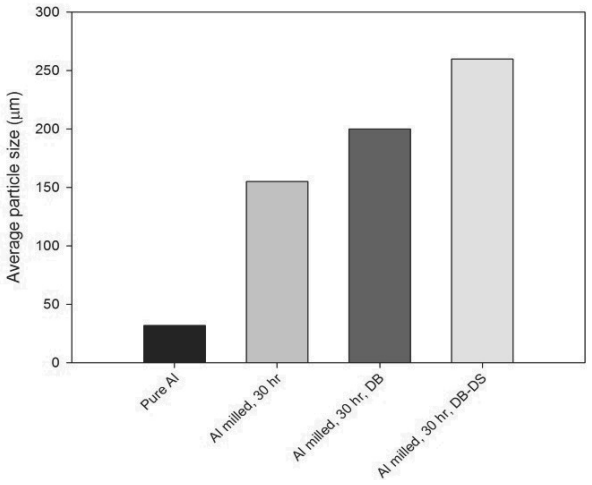

Fig. 10. Laser particle analyser results of 30 hr milled aluminium with different ball size (10, 15 and 20 mm), ball no. (31) and rotation speed (250 rpm)

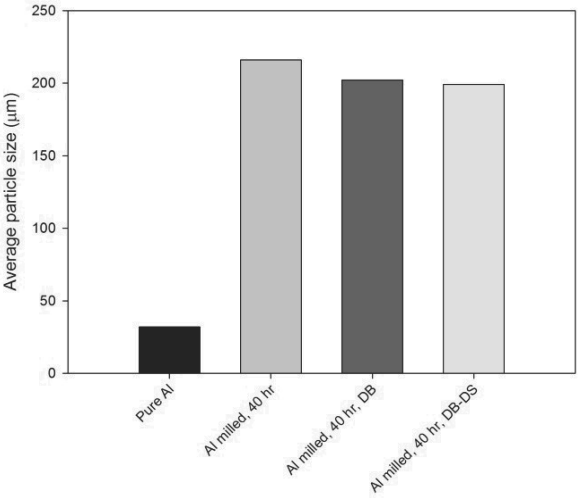

Fig. 11. Laser particle analyser results for pure aluminium and 40 hr milled Al with different ball sizes (10, 15 and 20 mm), ball no. (31) and rotation speed of 250 rpm.

Fig. 13 shows that aluminium powder size decreased to 8.9 µm after 30 hr of milling from ~31 µm for pure aluminium, and then the aluminium particles started to agglomerate when the milling time was increased up to 40 hr and the particle size was changed from ~9 µm to 12 µm in 40 hr of milling. However, for aluminium, the particle size started to increase after 30 hr of milling. This could be due to the low melting temperature of aluminium, because when the milling time is increasing, the heat inside the milling jar increases and agglomeration occurred causing the particle size to increase accordingly.

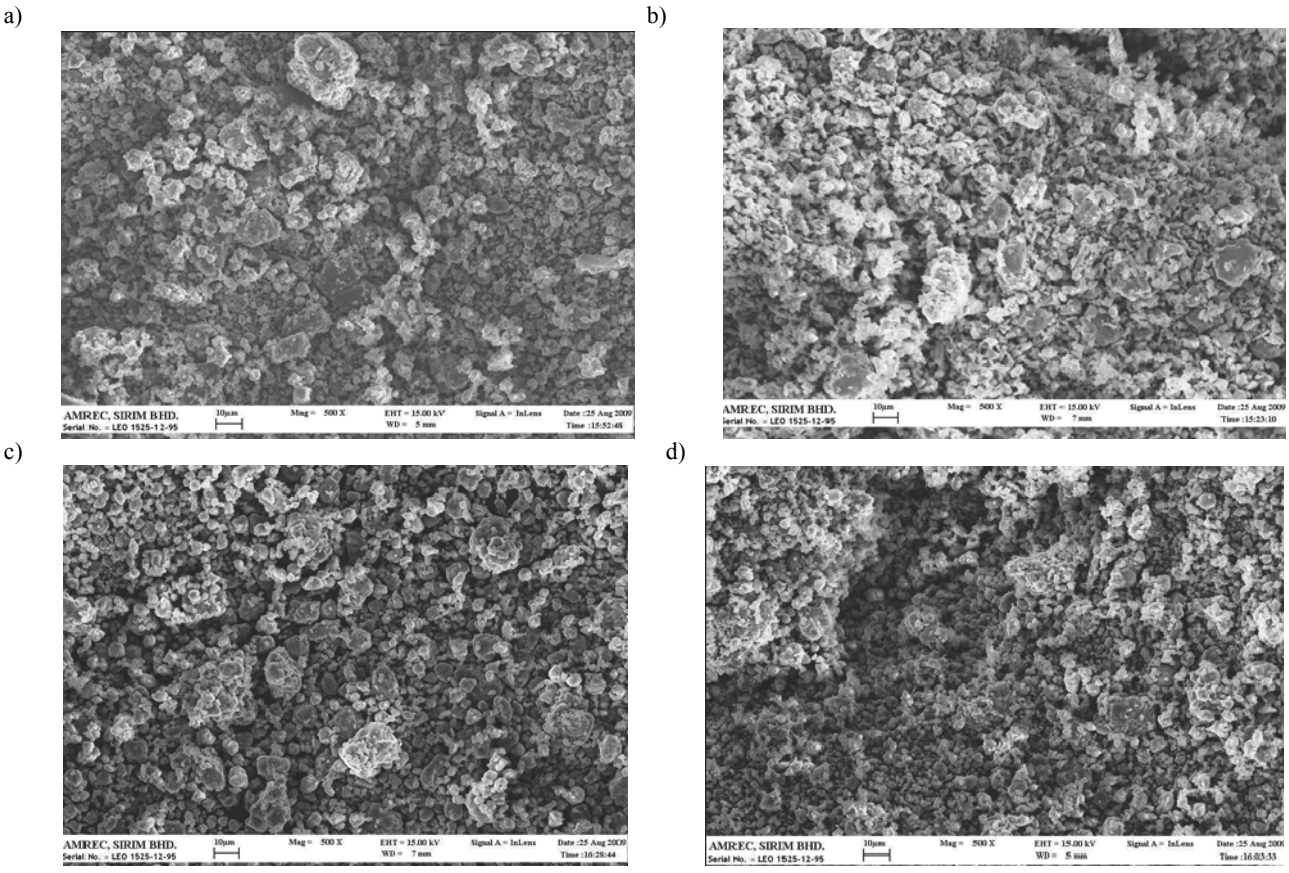

Fig. 12. SEM micrograph of: 7 hr (a), 15 hr (b), 30 hr (c) and 40 hr (d) milled aluminium with methanol

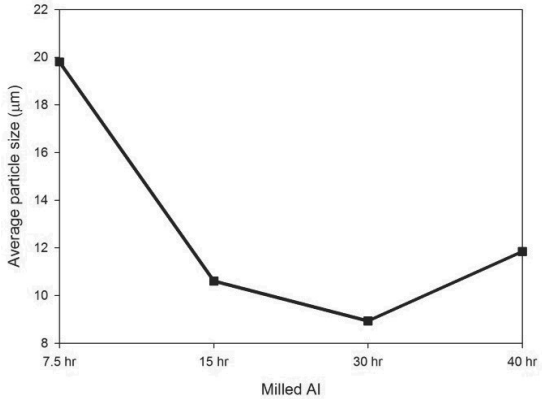

Fig. 13. Laser particle analyser results for 7.5-40 hr milled aluminium with methanol

4. Conclusions

Milling parameters for aluminium composite were successfully optimized with 100 stainless steel balls (10 mm), 200 rpm rotation speed with direction reversal and 1 min pause time after every 15 min running time, under argon gas for 30 hr of milling. However, as the PCA changed from stearic acid to methanol the particle size decreased to 8.93 µm and underwent some changes in shape from flake-like structures to more or less granular (disc) structures.

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