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# Monitoring alloy formation during mechanical alloying process by x-ray diffraction techniques

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**ABSTRACT** Mechanical alloying (MA) is a novel processing technique that use high energy impact ball mill to produce alloys with enhanced properties and microscopically homogeneous materials starting from various powder mixtures. Mechanical alloying process was originally developed to produce oxide dispersion strengthened nickel superalloys. In principal, in high-energy ball milling process, alloy is formed by the result of repeated welding, fracturing and rewelding of powder particles in a high energy ball mill. In this process a powder mixture in a ball mill is subjected to high-energy collisions among balls. MA has been shown to be capable of synthesizing a variety of materials. It is known to be capable to prepare equilibrium and non-equilibrium phases starting from blended elemental or prealloyed powders. The process ability to produce highly metastable materials such as amorphous alloys and nano-structured materials has made this process attractive and it has been considered as a promising material processing technique that could be used to produce many advanced materials at low cost. The present study explores the conditions under which aluminum alloys formation occurs by ball milling of blended aluminum and its alloying elements powders. In this work, attempt was made in producing aluminum 2024 alloys by milling of blended elemental aluminum powder of 2024 composition in a stainless steel container under argon atmosphere for up to 210 minutes. X-ray diffraction together with thermal analysis techniques has been used to monitor phase changes in the milled powder. Results indicate that, using our predetermined milling parameters, alloys were formed after 120 minutes milling. The thermal analysis data was also presented in this report.

(Mechanical Alloying, Aluminum Alloys, Ball Milling)

## INTRODUCTION

Mechanical alloying is generally a dry, high energy milling process that produces alloys and composite powders by cold welding and fracturing of metal powders in a high energy impact mill. MA can be performed by using various types of ball mills such as vibratory mill, planetary mill and attritor type mill under various milling conditions. This process is considered as one of the novel processing technique that offers the potential of producing various useful materials by solid state reaction. Solid state reaction between pure elements during the mechanical alloying proceeds gradually under the permanent repeated action of powder deformation. This process is usually accompanied by strong deformation of crystalline lattices of milled powder [1]. Many of recent research efforts have reported application of MA process to synthesize various materials such as intermetallic powder engineered for specific applications, non equilibrium structure/microstructure materials including amorphous alloys, extended solid solutions, metastable crystalline phases, quasicrystals and nanocrystalline materials [2]. In the process, composite particles with a characteristically layered microstructure are formed in the initial stage of milling. Subsequently alloy phase is formed in refined lamellar structure and eventually amorphization of the alloys phase occurs. The formation of alloys phase and amorphization of powder particles during milling are strongly affected by milling conditions such as type of ball milling, milling intensity, milling

speed and milling time as well as starting powder composition [2-3]. Many research works reported have focused on the mechanism, the possible alloy systems, the composition ranges, morphological evolution, characteristics of the powders,etc. for the amorphization and nanocrystalline powder formation by MA [2].

Many mechanisms have been proposed for the MA process, but only two mechanisms have been accepted by many researchers [2]. The two mechanisms proposed for alloying process of MA involve; first, rapid solidification following local melting caused by collision between balls and the particles and second, solid state inter-diffusion reaction at new surfaces of elemental powders by impact friction and wear.

The use of elemental powders provided greater flexibility in the choice of the alloying and the process of mechanical alloying results in a uniform microstructure with better properties. Moreover, the powder metallurgy route of processing powders by mechanical alloying process overcomes some of the problems faced by liquid metallurgy route [4].

The aim of the present investigation was mechanical alloying of elemental powders of Al-4.4Cu-1.5Mg in a planetary ball milling and thereby preparing a reactive alloy powder with 2024 composition. In the present work, a Fritsch single stage planetary ball mill was used to carry out MA of elemental powder mixture with varying milling time and alloying development was monitored by x-ray diffraction technique.

## MATERIALS AND METHODS

Pure elemental aluminum (Al), copper (Cu) and magnesium (Mg) powder with the mean particle size in the range of 75 micron and 63 micron respectively were selected. The elemental metal powders were mixed according to aluminum alloys 2024 composition ratio, i.e. Al-4.4Cu-1.5Mg and were blended and subsequently mechanically alloyed using Fritsch Planetary monomill. Mechanical alloying was carried out at room temperature for up to 210 minutes in a planetary ball mill using speed of 350 RPM. Due to short milling time, the powder produced was only partially alloyed. A ratio of 10:1 ball to powder was used. For this study, the mechanical alloying was carried out under an argon atmosphere and 0.6 wt% stearic acid ($C_{13}H_{36}O_2$) was used as a process control agent to avoid excessive cold welding of powder onto the ball and container surface during mechanical alloying. The mechanical alloying effect was evaluated by use of x-ray diffraction (XRD) analysis using Cu K$\alpha$ radiation with a wavelength of 1.54 angstrom. The mechanical alloyed powders were also investigated under SEM on a Philip XL40. Simultaneous thermal analysis (STA) comprises of Differential Thermal Analysis (DTA) and Thermogravimetric Analysis (TGA) of the MA powders were done using heating rates of 10 K/min.

## RESULTS AND DISCUSSION

During MA process, the morphology of Al and its alloying elements particles changed gradually. Figure 1 is SEM micrograph shows the typical change in the morphology of Al-4.4Cu-1.5Mg (2024 elemental alloy mixture) when subjected to MA process. It is obvious that the particles of Cu become finer and thinner during initial period of milling and finely dispersed in a lamellar microstructure of composite particles. A finer and more homogeneous distribution of Cu could have been produced if the milling had been continued sufficiently long. However, for the purpose of this study, milling has been discontinued after 210 minutes of MA.

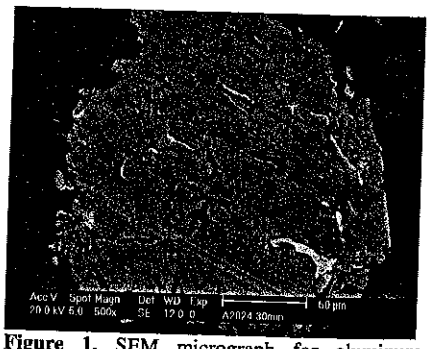


**Figure 1.** SEM micrograph for aluminum particle after 30 minutes of MA showing distribution of Cu layer.

All powder samples were then subjected to x-ray diffraction analysis and the x-ray diffraction spectrum patterns of the blended elemental aluminum with 2024 composition after MA were obtained and analyzed. The influence of milling time on the phase structure of the powder as presented in the x-ray diffraction spectrum is

clearly visible as peaks tend to broaden slightly with shifting in lattice spacing value, '**d**'. When the mechanical deformation was introduced into the powder during MA, particle and crystallite refinement occurs and lattice strain increases. This effects lead to broadening of the x-ray diffraction peaks and consequent decrease in the peak heights as observed in the x-ray diffraction spectrum. Analyses on the x-ray diffraction spectrum indicate that the '**d**' values representing lattice spacing of Al changes with increasing milling time.

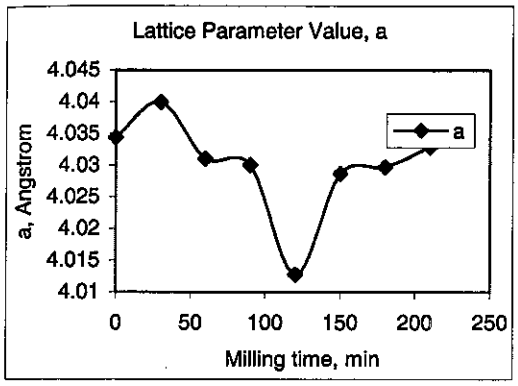


**Figure 2.** Change of lattice parameter value of aluminum as a function of milling time.

From this x-ray diffraction spectrum data obtained, the lattice parameter for MA aluminum powder was calculated from Al (111) plane. The changes in lattice parameter value, **a**, for aluminum with increasing milling time were plotted as a function of milling time and is shown in Figure 2. Result indicates that the '**a**' values calculated from Al (111) plane reflection increased slightly after 30 minutes milling and the value decreased progressively as milling continues and reached a minimum values for powder milled for 120 minutes. As milling continued, the lattice constant values increases progressively.

The reduction in lattice constant values for powder milled from 30 to 120 minutes could be due to effect of deformation in Al particles during milling. After 120 minutes it was believed that alloying has started with Cu and Mg dissolving in Al by diffusion and hence the lattice parameter values increases.

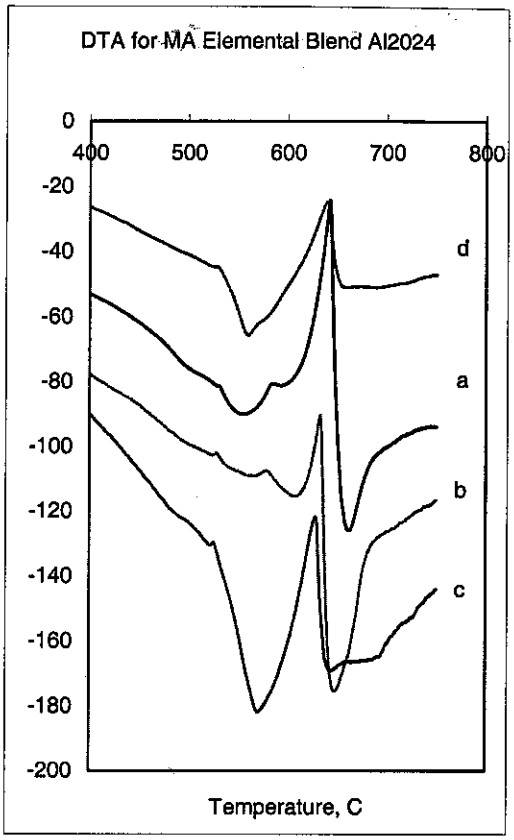


**Figure 3.** The DTA spectrum for elemental aluminum MA for
(a) 210 min, (b) 180 min (c) 150 min and (d) 120 min.

A differential thermal analysis (DTA) scan for non-milled powder mixture and for MA'ed powder mixture were taken and shown in Figure 3. This scan was taken to verify the effect of alloying time on the recrystallization and melting temperature of MA Al as a function of milling time. Characteristics of the DTA curve obtained for different MA times is the presence of an initial large exothermic peak due to recrystallization of MA Al powder and subsequently followed by a large endothermic peak due to melting of the matrix aluminum.

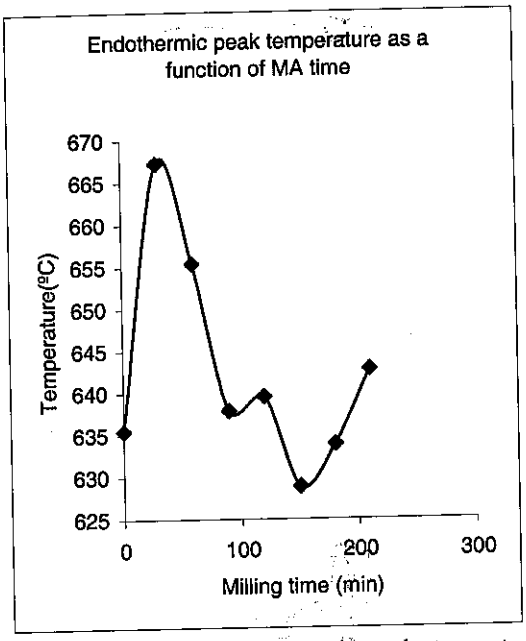


**Figure 4.** Changes in endothermic peak temperature represent melting of aluminum as a function of milling time.

Detailed analysis on this endothermic peak shows that the peak temperature change with milling time and this changes were plotted and shown in Figure 4. Result indicates that, this endothermic peak temperature that represents melting temperature of matrix aluminum alloy increases to highest value of 667 °C for powder milled for 30 minutes. However as milling continued, this endothermic peak temperature decreased with increase in milling time to the lowest value of 629 °C for 150 minute MA time. As milling continued, this endothermic peak temperature increased again. The trend of endothermic peak temperature changes with milling time can be seen to be following the same trend as changes in lattice parameter value with milling time calculated from x-ray diffraction spectrum as shown in Figure 2.

## CONCLUSION

Mechanical alloying process is a new novel process to produce alloys and it has been successfully used to produce several types of aluminum alloys with nano-structured morphology. On mechanically alloying blended elemental powder mixtures, interdiffusion between the components occurs and solid solution forms. The solid solubility limit is expected to increase with milling time as diffusion progresses. The alloying process can be monitored by x-ray diffraction technique, as the solid solubility levels have been generally determined from changes in the lattice parameter values that can be calculated from shifts in peak positions in the x-ray diffraction patterns.

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