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Formation of Nanostructured Al-Mg-Si Alloys and Evaluation Its Properties

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In this study, nanostructured Al-Mg-Si (Al6061) alloy was prepared from elemental powders by mechanical alloying and heat treatment. 98.4% aluminum, 1% magnesium, 0.6% silicon powders were mixed and mechanically alloyed under argon atmosphere. The rotation speed of 500rpm and ball to powder ratio of 10:1 was employed. The mechanical alloyed powder was isothermally heat treated at 400 °C for 2 h under argon atmosphere.

The results showed that after 10h of milling, a solid solution of Al-Mg-Si with a grain size of ~ 40 nm was produced. The as milled and annealed powder was characterized by scanning electron microscopy (SEM) and X-ray diffraction (XRD). The microhardness values of alloy increase by increasing MA time. Mg_2Si particles precipitate from solid solution during subsequent annealing. The as milled powder appeared to have good thermal stability against grain growth so that the grain size after annealing remained constant (~ 40 nm).

Keywords: Elemental powder; Mechanical alloying; Nanostructure; Al6061.

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1. INTRODUCTION

6xxx Aluminum alloys are of particular interest to both the aerospace industry and automotive industry because of their attractive combinations of properties. The benefits of 6xxx alloys include medium strength, formability, weldability, corrosion resistance, and low cost. 6xxx alloys have superior corrosion resistance compared to 2xxx and 7xxx alloys. The low cost of 6xxx alloys is especially significant to the aerospace industry, that relies heavily on the more expensive 2xxx and 7xxx alloys [1].

Commercially available coarse grain aluminium alloys have exhibited poor hardness and strength. To improve the mechanical properties of Al alloys for structural applications, nanocrystalline (NC) or ultrafine grain (UFG) materials are being developed nowadays. According to the well-known Hall–Petch relationship, with decreasing grain sizes the strength is highly increased in nanocrystalline materials [2,3].

The nanostructure materials can be synthesized by several techniques that are capable of producing grain sizes of 20–400 nm2 One proven synthesis technique available to produce nanocrystalline materials is mechanical milling/mechanical alloying (MA) [4]. The MA process consists of repeated cold welding, fracturing and cold welding of powder particle mixture in a high-energy ball mill [5]. The repeated deformation, cold welding and fragmentation, during MA lead to the structural changes like decrease in the crystallite size and the accumulation of lattice strain occur in severely deformed powder [6].

In this regard, many researches have been focused on nanostructured aluminum alloys. For example, Tavoosi et al. [7] studied the effect of mechanical milling on solution behavior of Al–Zn system. Yazdian et al. synthesized nanostructured Al7075 alloy by mechanical alloying of mixed elemental powders [8] In another studies, Jaffari et al. [9] illustrated the influence of milling on phase transformation in nanostructured 2024 aluminum alloys.

In this study, we used elemental powder of Al, Mg and Si and prepared nanostructured Al-Mg-Si (Al6061) alloy by mechanical alloying. The effect of isothermal annealing was also studied.

2. EXPERIMENTAL

Powder of Al-Mg-Si (Al6061) alloy was produced by mixing of elements with appropriate percentage. The elemental powders were aluminum (99.99%), magnesium (99.98%) and silicon (99.98%). The powder mixture with composition of 1% Mg, 0.6% Si, bal. Al(wt.%) was milled in a planetary ball mill. The MA conditions were: 10 h, 500 rpm, 10/1 balls/load ratio (by wt.) with a 1% of stearic acid powder as process control agent (PCA). MA was performed in Argon atmosphere. The milling media consisted of five 20 mm diameter balls confined in a 120 ml volume vial. After mechanical milling powders were isothermally heat treated in argon atmosphere for 2 hours. Structural changes of powders during milling and subsequence annealing were investigated by X-ray difractometery(XRD). A Philips diffractometer (40 kV) with Cu K_ radiation ($\lambda = 0.15406$ nm) was used for XRD measurements. The XRD patterns were recorded in the 2θ range of 20–90◦ (step size 0.03◦, time per step 1 s). The Al grain size was estimated from broadening of XRD peaks using Williamson–Hall formula [10]. The Williamson–Hall relationship is expressed as follows:

$$\beta\cos\theta = 0.9\lambda/D + \varepsilon\sin\theta \qquad (1)$$

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In this equation θ is Bragg diffraction angle, λ, the wave radiation used (1.54 Å), ε, the effective strain associated with mechanical alloying and A(constant)=1. The ßcosθ for several peaks were calculated and the best line drawn through them. The slope of the line represents the average internal strain (ε) and intercept gives the grain size.

Isothermal annealing was carried out at 300 °C, 400 °C for 2 h in order to form Mg_2Si precipitate.

3. RESULT AND DISCUSSION

3.1 Formation mechanism of Al-Mg-Si (Al6061) alloy

Powder phase changes were investigated during mechanical milling by XRD. The XRD patterns of as mixed and milled powders after various milling times are shown in Fig. 1. After 10 h of MA broadening of the Al peaks accompanied by remarkable decrease in their intensities occurred as a result of refinement of crystalline size and enhancement of lattice strain [11]. Small peaks of remaining Si are also seen on XRD pattern after 10 h of milling.

Repeated collision of ball to powders during ball milling can increase density of crystal defects (Dislocations , vacancies, grain boundaries). These defects have more open structures to solve atoms. On the other hand internal energy of the Al–Mg-Si mixture increase during ball milling. Therefore, system tends to reduce its internal energy by formation of a supersaturated solid solution and hence decrease phase interface [7] So MA process can increase the solubility of element. Hence all Mg atoms dissolved in Al, but some Si atom. The existence of initial Si in MA powder is due to the very low equilibrium solid solubility of Si in Al according to the Al-Si phase diagram.

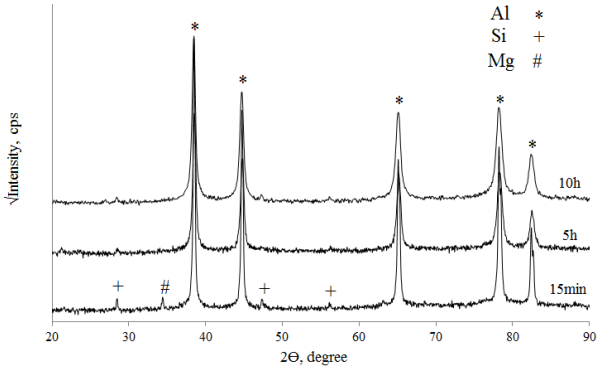

Fig. 1 – The XRD patterns of the Al- Mg-Si mixture milled for different times

Displacement of Al (111) XRD peak is presented in Fig. 2. By increasing milling time Al (111) peak shifted to lower angles. Two factors can cause the peaks shift in a diffraction pattern: Remaining uniaxial stresses and dissolution of atoms with different radius. In mechanical alloying there is high density of crystalline defects and dislocation because of frequent collision of ball to powders. Residual stress cannot shift XRD peaks of mechanically alloyed materials, because there are both compression and tension stress around each dislocation. It can be concluded that the peaks shift in mechanically alloyed material is due to the partial dissolution of Mg and Si in Al lattice.

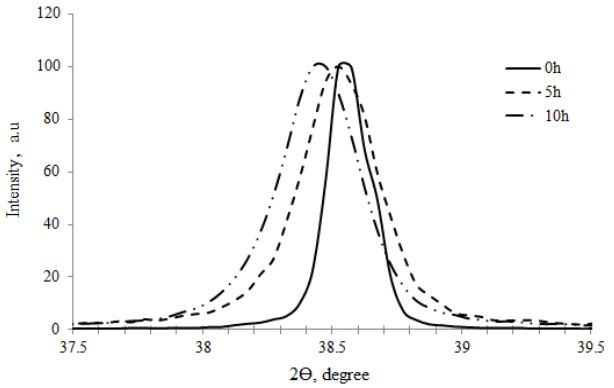

Fig. 2 – The displacement of Al (1 1 1) XRD peak during MA.

The change in Al lattice parameter is presented in Fig. 3. The Al lattice parameter was calculated using all Al XRD peaks and the mean value was reported. The lattice parameter of Al increases with increasing milling time. Increasing lattice parameter can be due to the dissolution of Mg and Si in Al lattice. In fact, Mg atoms have greater influence on Al lattice parameter than Si. Amount of Mg is higher than Si and all of Mg atoms dissolved in Al lattice but only partial dissolution of Si atoms are possible. Displacement of Al (111) XRD peak towards lower angles and increasing the Al lattice parameter by increasing milling time suggest the gradual dissolution of Mg and Si atoms in Al lattice and consequently formation of an Al-based supersaturated solid solution after 10 h milling.

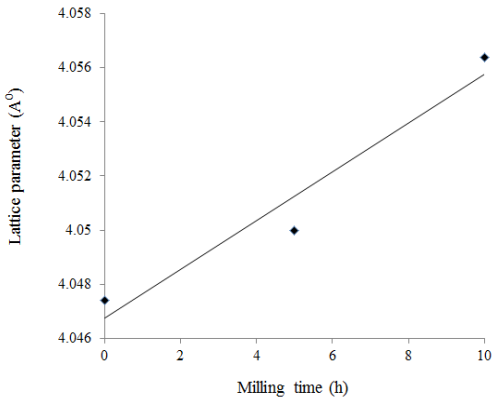

Fig. 3 – Al lattice parameter of Al-1% Mg-0.6% Si powder as-received and after different milling times

3.2 Isothermal annealing

The effect of isothermal annealing was studied. The as milled powder were isothermally annealed at 300°C and 400°C for 2 h. According to Fig. 4 Mg_2Si peaks (as the main precipitate in Al-Mg-Si alloys) are negligible after annealing at 300°C (Fig. 4b), but significant peaks of Mg_2Si were observed for powder after annealing at 400°C (Fig. 4c). Previous researches, reported formation of Mg_2Si below 250°C is negligible. It is also reported that if annealing temperature was above 370°C, significant amount of Mg_2Si could be formed. These accord well with the results obtained here [12,13].

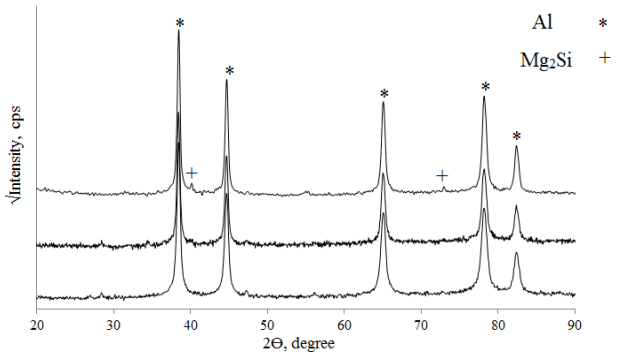

Fig. 4 – The XRD patterns of the mechanically alloyed Al- Mg-Si before and after isothermal annealing. a) mill for 10 h. b) milled for 10 h and annealed at 300°C for 2 h. c) milled for 10 h and annealed at 400°C for 2 h.

3.3 Grain size studies

Al grain size and lattice strain were calculated after different milling times. The broadening of the Al XRD peaks with increasing milling time is caused by the refinement of the crystalline size and strain induced during MA process. The Al grain size was estimated from broadening of XRD peaks using Williamson–Hall formula.[10] Because of mechanical deformation induced into the powders, Al grains refine and their internal strain increases. Mechanical deformation of powder increases dislocation density. Dislocation slips and then dislocation arranging formes a cellular structure, which change to nanometric grains by further milling. Fig. 5 shows changes of Al grain size and lattice strain as a function of milling time, in early stage of milling time Al grain size decrease sharply and then approaches a constant value at longer milling times. When the rate of creation and annihilation of dislocations become identical Al grain size becomes constant. After 5 hours of milling, the Al grain size was about 40 nm [14,15]. Al grain size and internal strain after 10 h milling were about 40 nm and 0.43, respectively.

Grain size after annealing at 400°C for 2 h was did not change and remained at about 40 nm. The as milled powder appeared to have good thermal stability against grain growth. Good thermal stability of as milled powder refers to 1) formation of Mg_2Si precipitate that prevents grain growth. 2) excess volume of nanomaterials. nanocrystals are structurally characterized by the ultrafine crystalline grains, and a large fraction of atoms located in the metastable grain boundaries in which the nearest neighbor configurations are much different from those in the crystallites. In other words, the grain boundary possesses an excess volume with respect to the perfect crystal lattice [16].

3.4 Microhardness studies

The microhardness values as a function of milling time are shown in Fig. 6. By increasing MA time the hardness of powder particle increased due to the refinement of grain size, introduction of lattice strain (Fig.5) as well as Mg and Si solid solution hardening and dispersed Si.

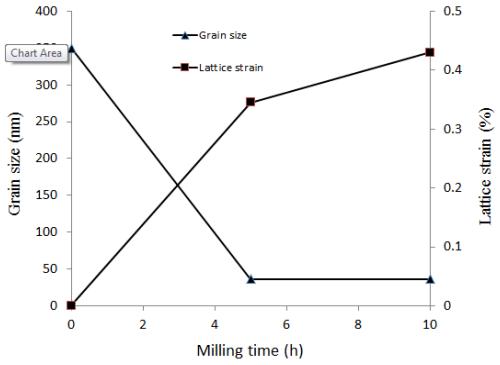

Fig. 5 – The grain size and lattice strain of Al matrix as a function of MA time

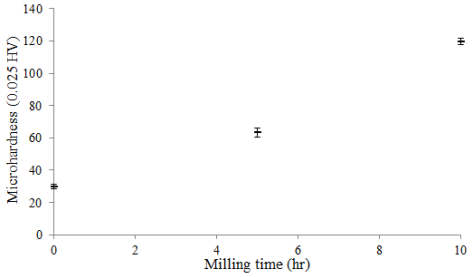

Fig. 6 – The microhardness value of Al–Mg-Si powder particles versus MA time

3.5 Morphological studies

Fig. 7 shows the morphology of powder particles at various milling times. In initial stage of MA the rate of powder agglomeration is higher than fragmentation which leads to an increase in powder particle size (Fig. 7b). As milling time increases the fracture of the powder particles predominates (Fig. 7c, d and e) reducing the powder particle size. For milling times longer than 30 h, the change of particle size was not significant indicating that an equal rate of agglomeration and fragmentation of powder particles is achieved.

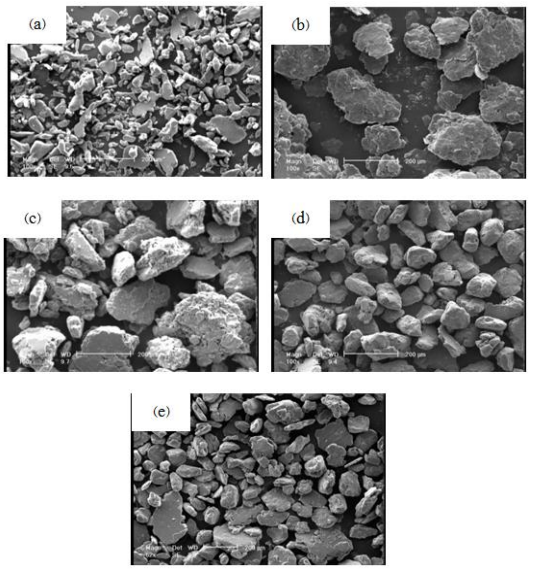

Fig. 7 – Changes in morphology of Al–Mg-Si powder particles during MA: (a) as-mixed powder and after (b) 2 h; (c) 5 h and (d) 7 h, (e) 10 h MA

4. CONCLUSIONS

Nanostructured Al6061(Al-Mg-Si) alloy can formed after mechanical alloying and isothermal annealing. MA of Al6061 by gradual dissolution of Mg and Si. After milling a nanostructured solid solution of Al with Si particle formed. By subsequent annealing Mg_2Si precipitate formed. MAed nanostructured Al6061 had good thermal stability and the grain size after isothermal annealing remained unchanged (~ 40 nm).

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