Microstructure and mechanical behavior of Al-xMg/5Al₂O₃ nanostructured composites

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Abstract

The effect of Mg content and milling time were investigated on the microstructure and microhardness values of Al-xMg/5Al₂O₃ (x = 0, 4, 8 and 12 wt %) nanostructured composite prepared via high energy milling technique. XRD results showed an acceleration of alloying process and formation of Al (Mg)ₓ by enhancing percentage of Mg element. Also, by increase in Mg percentage the grain size reduction was more considerable during milling treatment. Additionally, increment of the Mg content up to 12 wt% causes the increase in micro-strain of the samples (from 0.31 to 0.82%). Increase in Mg concentration accelerates the mechanical milling process. According to SEM results a coaxial and circular morphology with a uniform distribution of powder particles has been formed. Up to 12 wt% (for each milling time), significant increase in microhardness (215 HV) was carried out due to solid solution hardening and crystallite refinement. From 10 to 15 h, a slight increase in microhardness up to 218 HV can be observed.

1. Introduction

Al is the most extensively used substrate for the production of metal-based composite. This element has been noticed in plenty of industries due to its exclusive properties like low density to weight ratio, ability of hardening precipitation, great corrosion resistance and high electrical and thermal conductivity [1].

On the other hand, Al-based composites strengthened with ceramic particles prepared by the mechanical alloying process provides a combination of properties of both the metallic matrix and the ceramic reinforcement components resulting in improved physical and mechanical properties of the composite. Alumina particles were chosen as the reinforcement for several reasons, such as the homogeneous properties in various directions and the good formability of these materials [1–4], chemically inert with Al, applicability at high temperatures and benefits of resistance to creep [5]. Also the presence of Al₂O₃ particles in a metal matrix improves the hardness of this material at room and at higher temperatures [6]. According to research in the field of production of metal-based composites, the percentage of reinforcement phase varies according to the type of composite application. For example, it has been used to improve strength and hardness between 5 to 15% [6–9], to increase abrasion properties up to 30% and to improve electrical properties up to 70%. [10].

In the previous works [6, 11], the use of mechano-chemical treatments in the production of Al-X binary substrates (X = Cu, Zn, Si) instead of pure Al substrates has been done. Therefore the aim of present study is the choice of binary substrates prepared by mechanical milling method. Also the effect of high energy milling on morphology, microstructure and mechanical properties of the individual composite has been studied. Hence, the use of Al-Mg binary substrates has been considered in the production of Al substrate composites [12–17]. Research demonstrated that Mg addition to Al matrix improves strength and ductility, simultaneously [18]. Also, it has a remarkable effect on grain refinement and alloying acceleration. The former increases hardness, tensile strength, and yield strength [12, 14, 15 and 18]. The equilibrium solubility of Mg in Al at room temperature has been reported lower [19] than one atomic percentage while, this value can be increase up to 45 atomic percentages for Al-50at%Mg alloy [20]. Further research in this area has proved the formation of
intermetallic compounds with 20% Mg and above [12–20]. Therefore, the objective of this study is to investigate the effect of various compounds (0, 4, 8 and 12% Mg) which less has been investigated. Also, the effect of Mg content on morphology, microstructure and mirohardness of composites has been researched.

2. Experimental procedure

In this research, Al–xMg/5Al₂O₃ (x = 0, 4, 8 and 12 wt% of Mg) composite was milled in a high energy planetary mill under argon atmosphere for 2, 5, 10 and 15 h. The ball to powder mass ratio was constant and equal to 20:1 and the speed of movement of the mill container was chosen 250 rpm. In order to control the temperature of the process, the mill was stopped for 30 min after 30 min working. X-ray diffraction (XRD) model was used to characterize the formed phases and the grain size of the crystals, lattice strain and lattice parameter.

X-ray diffraction measurements were carried out in a Philips X’Pert High Score diffractometer using Cu Kα (λ = 1.5405Å) radiation over 30°–70° 2θ. To adjust the shift of zero angle, the silicon crystal was used as a standard sample. The crystallite size (grain size) and lattice strain (micro-strain) were estimated using the Williamson–Hall method [21]:

\[ B_i \cos \theta = K \lambda / d + 2 \varepsilon \sin \theta. \]  

(1)

Where \( B_i \) is the full-width at half-maximum of the diffraction peak, \( \theta \) is the diffraction angle, \( \lambda \) is the X-ray wavelength, \( d \) is the crystallite size, and \( \varepsilon \) is the lattice strain. \( B_i \) can be given as:

\[ B_i^2 = B_i^2 - B_i^2. \]  

(2)

Where \( B_i \) is the width at half-maximum of the Si powder peaks used for calibration and \( B_i \) is the evaluated width. Thus, it is clear that when we plot \( B_i \cos \theta \) against \( \sin \theta \) for each Al peaks, we get a straight line with slope \( (\varepsilon) \) and intercept \( K \lambda / d \). The crystallite size \( (d) \) can be calculated from \( K \lambda / d \) (K and \( \lambda \) are determinate) and lattice strain is slop line \( (\varepsilon) \) [21]. The crystallite size and micro-strain was determined using 3 angle peaks.

The morphology and the shape of powder particles were studied using a scanning electron microscope (SEM, Cam Scan MV2300).

Since the main goal of microhardness test was to understand the role of both work hardening and solid solution hardening on microhardness, thus the milled powders at different times were tested before sintering process to avoid omitting plastic deformation effect. The hardness of powders was determined by microhardness measurements using a Vickers indenter at a load of 250 milli-Newton and dwell time of 5 s. For measurement of microhardness, the powder mixtures were cold pressed under 300 MPa. The pressed specimens were in the form of cylinders with a diameter of 10 mm and a height of 5 mm. Prior to indentation, the surfaces of samples were polished using a sequence of increasing grit sandpaper followed by a series of diamond pastes. For each sample, at least 5 hardness tests were taken and their average was recorded.

3. Results and discussion

The XRD pattern of various samples after 2 h milling time is presented in figure 1. For \( x = 0 \), only the Al peaks are observed. For all samples the characteristic linear line of alumina are not detected due to their reduced particle size to sub-microns [21].

From \( x = 4 \) to 12, in addition to Al peaks, Mg lines are also observed. By increasing the Mg percentage, both intensity and number of visible peaks are increased. In all samples, with increasing Mg content the height of the peaks decrease and their width increased due to grain refinement and strain introduction.

Figure 2 illustrated the XRD patterns of various samples after 10 h milling. As can be seen the intensities of Mg peaks were smaller than those of the 2h milled samples. Nevertheless, for \( x = 4 \) and 8, Mg peaks are still visible and for \( x = 12 \) the Mg lines vanished completely.

The XRD patterns of 15 h milled samples as a function of Mg content are shown in figure 3. For samples with \( x = 4 \%), characteristic peaks of Mg phase are observed. The existence of Mg phase proves that the solid solution is not yet completed and it requires more milling time. For \( x = 8 \) and 12%, only related peaks of Al-Mg phase are observed (without any trace of Mg phase) due to the introduction of Mg atoms is in Al matrix which confirms the completion of alloying process [12–17].

The increase in Mg solubility in Al lattice during milling can be due to the formation of a nanocrystalline structure with a large volume fraction of the grain boundary [18]. Therefore, the faster reduction of the intensity of Mg peaks by enhancing the Mg concentration from 8 to 12% can be attributed to the finer grain size of substrate and further increase in grain boundaries by increment of Mg, which causes easier dissolution of Mg in Al matrix. In general, increasing the concentration of Mg accelerates the rate of solid solution formation. In
other words, by enhancing the Mg content, the supersaturated solid solution is formed in lower milling times and the alloying process is accelerated [1]. It is also shown in figures 1–3 that with the increase in Mg, the peaks become more broad caused by the grain refinement and strain accumulation in the matrix phase lattice [1].
The results of changes in grain size and lattice strain of the composite versus milling time are shown in figure 4. After 2 h milling, the crystallite size of all samples was reduced below the 100 nm. After 15 h milling, for $x = 0$, the grain size was reduced to about 65 nm. With increasing Mg, the grain size decreased more sharply, so that up to 12% Mg, the grain size decreased to about 30 nm.
Generally, during the milling process, because of the extreme plastic deformation, the density of point defects and dislocations increases. The lattice defects lead to increased internal energy and thermodynamic instability in powder particles.

In this case, the dislocations rearrange themselves to the lowest energy level and the low-angle boundaries are formed. Furthermore, dissolution of Mg in Al causes increase in the work hardening of powder particles and further increase in the density of dislocations \[22, 23\]. Increment of the dislocations density causes the conversion of low-angle grain boundaries to high-angle one at much shorter times, which accelerates the crystallite refinement process \[1, 12\].

Also, with milling time to 2 h lattice strain for all samples is same almost (about 0.30%). Higher times of milling (up to 15 h) lead to increase in point defect and the dislocations density due to mechanical work process and diffusion of Mg atoms with a larger atomic radius in the Al lattice. These factors result in accumulation of micro-strain of mixture powders (from 0.30 to 0.34% and 0.30 to 0.54% for sample whit 0 and 8% Mg, respectively). Additionally, increment of the Mg content up to 12%, has a significant effect on increasing the micro-strain of the composite (from 0.31 to 0.82%). It may be due to dissolution of Mg in the Al matrix completely and formation of the solid solution. The SEM images of 15 h milled samples versus Mg content are indicated in figure 5.

The effect of Mg presence on morphology in this figure is quite clear. For $x = 0$, after 15 h of milling, a flat and flaky morphology with a non-uniform distribution of powder particles has been obtained. With the increase in Mg content, the distribution of particles becomes more uniform and the particles size become smaller, so for $x = 4$ and 8%, a completely stable state has been created in the powder particles and a coaxial and circular morphology with a uniform distribution of powder particles has been formed. With the addition of magnesium, after 15 h, the flat and flaky morphology is converting to a coaxial and circular morphology that can be due to the ductility of both Al and Mg (ductile-ductile system). During milling, Mg atoms penetrate the Al lattice due to the local temperature rise at the point where the bullets collide with the powder particles. Also, because the atomic radius of Mg is larger than Al, the dissolution of Mg in Al causes distortion in the Al lattice and increases the work

![Figure 5. SEM images of samples after 15 h of milling (a) without magnesium (b) 4% Mg (c) 8% Mg (d) 12% Mg.](image-url)
hardening of the substrate powder particles and consequently increasing the density of the dislocations [22, 24 and 25].

Also, the presence of reinforcing particles (Al2O3) has led to increased hardness in composite powder particles, which leads to a faster increase in the density of dislocations, resulting in greater hardness in the powder particles and more brittle and prone particles to failure. Ultimately, these factors accelerate the mechanical milling process and create a stable state in the powder particles after 15 h of milling.

The microhardness changes of composite powders over milling time as shown in figure 6. It is observed that for Al/5Al2O3 composite the increase in hardness (to 112 HV) in terms of milling time (to 15 h) has a relatively constant slope. More mechanical milling results to enhance the work hardening in the powder particles of substrate and also causes the brittle and large particles of Al2O3 to be broken and distribution of the particles in the Al matrix, which this leads to further increase of the work hardening in the powder particles and consequently increase the dislocations density. By adding 4% Mg to Al/5Al2O3 composite, its hardness increases with a constant trend but with larger amounts (130 HV after 15 h).

By increasing the percentage of Mg to 8% from 2 to 10 h, the hardness values are raised, but the process of increasing the hardness is similar to the previous samples. From 10 to 15 h the hardness increases more rapidly (187 HV). With raising the Mg content to 12%, from 2 to 10 h the hardness increases rapidly up to 215 HV, and from 10 to 15 h the rate of hardness decreases and the hardness becomes almost constant (218 HV after 15 h).

The increasing hardness trend with rising milling time is attributed to the grain refinement, accumulation of strain and creation work hardening in the powder particles [5, 26, and 27].

Magnesium increases the hardness by two mechanisms, which are the Al (Mg) solid solution formation and the intensification of hardness due to the presence of solution Mg in the Al lattice [28].

The hardness increase for x = 4% sample (from 2 to 15 h) and for x = 8% sample (from 2 to 10 h) is due to the soluble Mg in the Al lattice, which results to the increase in hardness and dislocation density. The fast increase in microhardness (From 10 to 15 h and 2 to 10 h for x = 8 and 12% Mg, respectively) is related to the hardness of the solid solution [27, 29]. Furthermore, Al-Mg binary matrix due to the higher hardness capability than the single Al matrix result to further decrease in grain size during milling and further increase in microhardness compared to non-alloy substrate.

On the other hand, with increasing Mg percentage and increasing milling time more than 10 h (for x = 12% Mg) due to more increase in work hardening and density of dislocations, dynamic recovery occurs and work hardening speed decreases. Also, it can be related to the completion of solid solution formation process and static recovery of the matrix phase due to a local increase in temperature when the bullets hit to the powder particles [12].

Figure 6. Variation of microhardness of Al–xMg/5Al2O3 powder mixtures according to milling time (2, 5, 10 and 15 h) and different percentages of magnesium (x = 0, 4, 8 and 12% Mg).
4. Conclusion

The results of studying the microstructure and microhardness changes of Al–Mg/5Al₂O₃ (Mg = 0, 4, 8 and 12%) composites prepared by mechanical alloying are as follows:

1. Milling of Al–xMg/Al₂O₃ powder with different percentages of magnesium up to 15 h results in the formation of a nanostructured composite with a grain size between 65 to 30 nm.

2. Formation of Al(Mg)ss and acceleration of the mechanical alloying process were observed in samples with 8 and 12% Mg, due to complete dissolution of magnesium in the aluminum lattice, while the situation was not observed for another samples.

3. For sample without Mg, after 15 h of milling, a flat and flaky morphology has been obtained. While, increase of 8 and 12% of Mg accelerates the mechanical milling process and a coaxial and circular morphology with a uniform distribution of powder particles has been formed.

4. Increment of work-hardening in powder particles with increasing mechanical milling process time and the presence of reinforcing particles in the matrix, results to an increase in the density of dislocations and thus reduces the grain size and accelerates the process of Mg penetration into the Al lattice. The combination of these factors eventually leads to an increase in the microhardness of the composites.

5. With increasing Mg content up to 4 wt.% (for 15 h milling) microhardness enhances with a slight slope to 130 HV because of the presence of solution Mg in the Al lattice, and with increasing up to 12 wt.%, Significant increase in microhardness (215 HV) was carried out because of solid solution hardening and crystallite refinement. A slight increase in microhardness up to 218 HV (From 10 to 15 h milling) can be the result of the end of the solid solution formation process as well as the creation of a dynamic recovery (due to high work-hardening created in the matrix powder particles) and static recovery (due to a local increase in temperature when the bullets hit to the powder particles).

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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