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STUDIES OF CRYSTALLITE SIZE AND LATTICE STRAIN IN AL-AL_2O_3 POWDERS PRODUCED BY HIGH-ENERGY MECHANICAL MILLING

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ABSTRACT

In the present study mechanical milling used to form nanocrystalline structure in Al-Al_2O_3 powders. The microstructure of the milled powders was investigated by X-r ay diffraction (XRD) analysis. It was found that the XRD peaks showed a significant broadening, which was related to grain refinement and lattice distortions. X-ray patterns were analyzed using Williamson–Hall treatment to determine the crystallite size and the lattice strain. nanocrystalline powders have been synthesized with microstructure showing a higher lattice strain and an evolution of the finest particles. Microhardness measurements and compression tests were performed to characterize the composite materials.

Keywords: XRD**;** Williamson–Hall**;** **Al**-Al_2O_3; Nanocomposites

1. Introduction

Nanocomposites, a high performance material exhibit unusual property combinations and unique design possibilities. The improvement in mechanical properties of nanocomposites makes them attractive materials for structural applications; their processing still presents significant challenges [1]. High-energy mechanical milling is a very effective process for synthesizing metal–ceramic composite powders. It allows incorporation of the metal and the ceramic phases into each powder particle. Since the nanostructure of each powder particle evolves through numerous deformations, fracturing and cold welding events, after a certain period of milling, powder microstructure homogeneity can be achieved at the same time as the nanostructure. [2].

Severe plastic deformation of the particles can lead to grain refining, accumulation of internal stress, change of the lattice parameter, and formation of cell structure [3]. The mechanisms of grain size refinement for the milled powders were identified on the basis of the microstructure and particle size studies given by Kuschke et al. [4]. They reported that

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plastic deformation, fracture and cold welding of powder particles were the three principle mechanisms which could operate in mechanical milling [4, 5]. The crystallite size and the lattice strain in nanocrystalline materials can be determined by X-ray line profile analysis.

The aim of the present work was to study the nanocrystalline structure formation in Al-Al_2O_3 powders deformed by high energy mechanical milling. The crystal structure and the phases present in the as-received and milled powders are identified using the X-ray diffraction technique. From diffraction patterns, using Williamson–Hall method, the grain size and the equivalent strain can be determined. TEM observations used in measuring the crystallite size and the results are compared with the results obtained from Williamson–Hall method.

2. Experimental work

2.1. Materials and milling

The materials used in the experiments were Commercial aluminum powder with a particle size of (-210+90 μm) and Commercial alumina powder with particle size smaller than 44 μm. Mechanical milling has been used to produce Al-20 wt. % Al_2O_3 composite powder. 50 gr mass of material were milled up to 45 h in a Planetary Monomill "Pulverisette 6". Grinding is carried out under Argon gas atmosphere at 300 rpm milling rotation speed and ball-to-powder ratio of 10:1. Stearic acid was used as a process control agent (PCA) to prevent the aggregation of powders during milling. The duration of milling process is fixed at 30 min in order to avoid temperature rise and the samples were taken at different times; 7, 12, 15, 21, 30, 38 and 45 h.

2.2. Structural analysis

X-ray diffraction (XRD) patterns were recorded for the as-received and milled powders using a Philips PW 1710 X-ray diffractometer using Cu Kα radiation (λ = 0.15406 nm) at 40 kV and 30mA settings. Scans were collected over a 2θ range of 20°- 90°. A smaller angular steps of 2θ = 0.06° was taken to measure the intensity of each Bragg reflection.

Techniques, such as the Williamson–Hall [6] and Halder–Wagner methods [7], are used to determine the grain size and the equivalent strain. The grain size refinement studies identified on the basis of the microstructure and particle size studies, reported that plastic deformation, fracture and cold welding of powder particles were the three principle mechanisms which operate during mechanical milling [4,8]. In this work, the analysis of the XRD peaks was performed via the Williamson–Hall method, in which the full width at half maximum (FWHM), β, due to sample imperfections is related to the crystallite size, D, and the strain, ε according to the following equations;

$$\beta^* = d^*\varepsilon + 1/D \quad (1)$$

Where, $$\beta^* = \beta \cos\theta / \lambda \quad (2)$$

and $$d^* = 2 \sin\theta/\lambda \quad (3)$$

θ is the Bragg angle and λ is the X-ray wavelength used. The experimental breadth of a given reflection was fitted for the peak breadth from the instrument by Voigt functions using the Originpro 8 software. FWHM is corrected, using silicon as standard reference material. Values of $\beta^*$and d^* can be calculated correctly from the previous equations. Then from plotting β^*against d^* crystallite size and strain can be obtained where, the intercept of the plot of β^* against d^* gives $1/D$ and the slope gives the strain [5].

The powders were characterized for their microstructure and distribution of the Al_2O_3reinforcement particles in the Al matrix using a JEOL-JSM 5400 F scanning electron microscope (SEM).

3. Results and Discussion

3.1. X- ray diffraction pattern and structural changes

X-ray diffraction patterns of the Al-Al_2O_3 powders milled with rotation speed 300 rpm at different periods of time are shown in Figures 1. From the figure it is clear that line broadening increases with milling time. It is attributed to high energy milling which involves repeated deformation, cold welding, and fragmentation. Structural changes such as decrease in crystallite size, accumulation of microstrain, and dislocations occur in deformed powder. So, the X-ray line broadening analysis has been used to characterize the microstructure in terms of crystallite size and lattice strain. Line profile analysis helps in ascertain the evolution of the apparent size, D, and the equivalent lattice strain, ε, with milling time [9]. The profiles of all Bragg reflections are broadened; this is related to the reduction of crystallite size and to the important lattice strain introduced by milling.

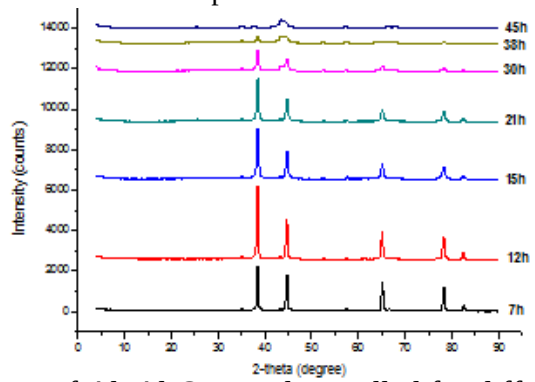


Fig. 1. XRD patterns of Al–Al_2O_3 powders milled for different times; 7, 12, 15, 21, 30, 38 and 45 h.

For appearing line broadening increase with milling time in XRD pattern, typical first peak lines of Al-Al_2O_3 powders milled for different times are plotted separately. Figure 2 shows profiles plotting for only the first peak (P1), of the milled powders for different times. It's noted that with milling time, peak broadening increases and the peak is shifted to lower θ values. The reason of this shift may be the change of the lattice parameter during milling. [10, 11].

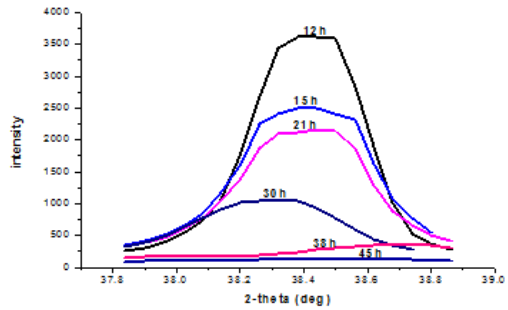


Fig. 2. Bragg reflections Profiles of the first peak- of milled Al–Al_2O_3 composites powders for different times (12, 15, 21, 30, 38, 45 hours).

The profiles of all Bragg reflections are broadened; this is related to the reduction of crystallite size and to the important lattice strain introduced by milling. To estimate the crystallite size and the lattice distortion, an inspection of the shape of the diffraction peaks was performed. The same calculations procedures are carried out for every peak of the two nanocomposites samples of different times for Al and Al_2O_3.

3.2. Crystallite size and the lattice strain calculations by WH method

By using the previous equations; $\beta^* = \beta \cos\theta / \lambda$ and $d^* = 2 \sin\theta/\lambda$; values of $\beta^*$and d^* can be calculated. From Plotting β^* against d^*, the crystallite size *D*, and the lattice strain ε of composites at different times are obtained. Where, the curve intercept gives 1/*D* and the slope gives the strain. Collection of plotting β^* against d^* for all samples; 7, 12, 15, 21, 30, 38 and 45 hours of Al-Al_2O_3 is shown in Figure 3.

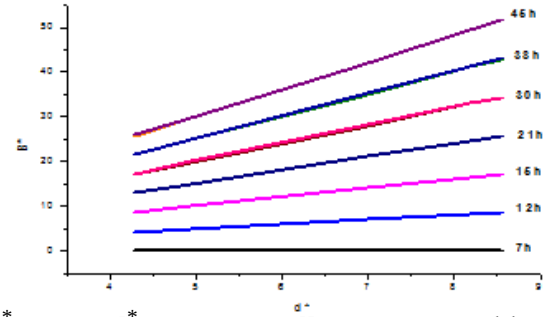


Fig. 3. Plotting β^*against d^* to get crystallite size *D*, and lattice strain ε of Al–Al_2O_3 – 300rpm

After getting D and ε values of aluminum and alumina for different samples of the Al-Al_2O_3 nanocomposite. Figure 4 shows the variation of the crystallite size and of the lattice strain of Al and Al_2O_3 against milling time obtained by WH method. It can be seen that the rate of Al grain refinement continuously decreases reaching, after 45h of milling, the value of 19.77nm. For alumina in Al-Al_2O_3 powders, the rate of the grain refinement continuously decreases reaching, after 45 h of milling, a value of 41.29 nm. On the other hand, the lattice strain shows a continuous increase to value of 1.16% for Al and a value of 0.663 % for Al_2O_3 . It's noted that the milling induces a higher lattice strain and an evolution of the finest particles.

The low crystallite size can be explained by the competition between the levels of stress produced by a milling device, and the degree of dynamic recovery in the milled material [5, 10]. Also, the local deformation of the matrix in the vicinity of the reinforcement particles is increased, accelerating the work hardening of the matrix, and thus, the extent of grain refinement [12]. It is also noticeable from crystallite size values, that crystallite size of alumina is bigger than that of Al. That is may be related to a degree of coverage of the reinforcement (Al_2O_3) particles by the aluminum i.e. encapsulation of reinforcement particle with Al. This explanation is in good agreement with others [13, 14]

Figure 5 shows TEM picture of the Al–Al_2O_3 composites powder milled at 300rpm for 45h. The crystallite size measurements were obtained using TEM observations in order to compare these results with results coming from XRD. XRD methods have been more commonly used to determine the grain size. Regarding the comparison between the results obtained from the Williamson–Hall method and direct TEM observations; Ungar [15] reviewed the meaning of size obtained from peak broadening in XRD patterns and

compared the size values obtained by XRD methods with those obtained by direct microstructural studies, e.g., TEM. He pointed out that the two measurements can be: (1) identical to each other, (2), totally different or (3) in good qualitative correlation.

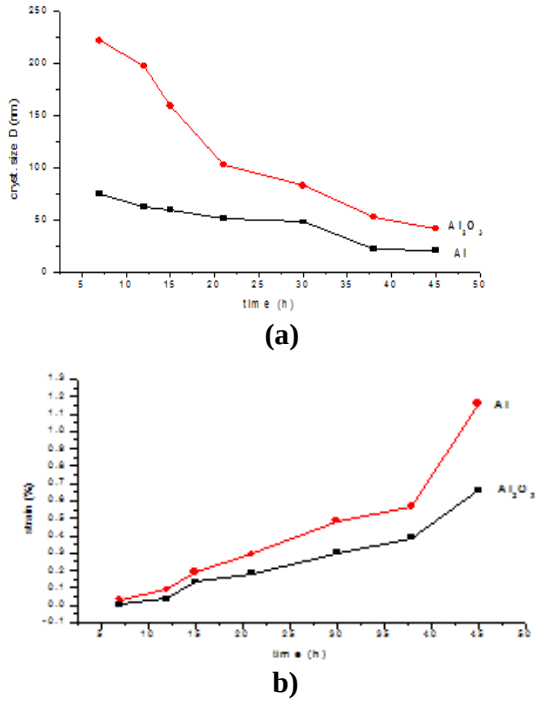


Fig. 4. Changes of Al and Al_2O_3 crystallite size and lattice strain of the milled Al-Al_2O_3 powders -300rpm (7, 12, 15, 21, 30, 38, 45 h), (a) Crystallite size. (b) Lattice strain

3.3. TEM analyses

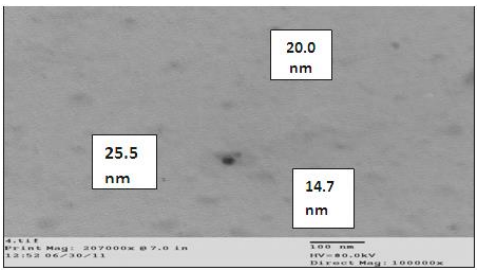
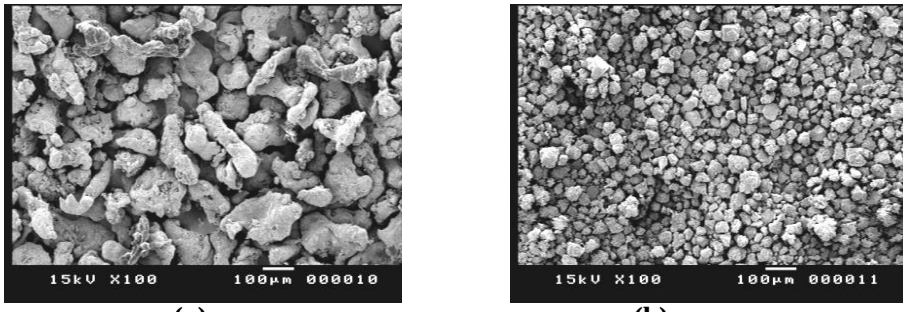


Fig. 5. TEM microstructure of Al–Al_2O_3 powders milled at 300rpm for 45 h

In this work the crystallite size value from WH method for Al–Al_2O_3 powders is 19.77 nm for Al and from direct TEM observation, range from 14.7 to 25.5nm. So, the grain sizes obtained by Williamson–Hall method and direct TEM observations are nearly the same i.e. they in agreement with Ungar first condition. Suryanarayana, C. [16] reported that the grain sizes obtained by the X-ray peak broadening studies, after incorporating the appropriate corrections, and the direct TEM techniques are expected to be the same, and this is achieved in this work and also in other cases [17,18].

3.4. Microstructural observations

Scanning Electron Microscope (SEM) analysis for the as-received powders aluminum and alumina is given in Figure 6. From which, Al particles are irregular in shape and alumina particles are almost spherical.



(a) **(b)**

Fig. 6. SEM images for the as-received powders: (a) Aluminum (b) Alumina

Figure 7 shows the SEM image of Al-Al_2O_3 powder particles after mixing the powders and before milling (0 hours milling time). The agglomerations of alumina particles are observed, which can be removed by the milling.

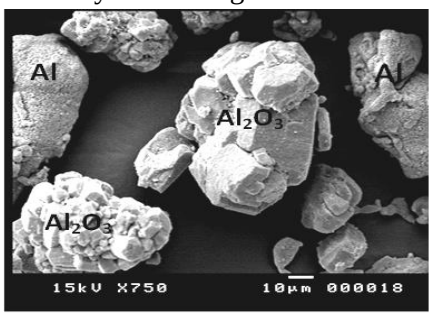


Fig. 7. SEM image of Al-Al_2O_3powder particles before milling (0 hours)

Figure 8 show the SEM images of Al-Al_2O_3 powder particles for 300 rpm milling speed at different milling times. A change in powder particle size happened with milling time, at 7 hours milling time, Figure 8 (a), the agglomerations of alumina are still observed, but they are smaller. At this milling time, flattened particles formed. This is because Al particles are soft and their sizes are increased by cold welding, which becomes prevalent and leading to an increase in particles size. After 30 hours milling time, particles morphology changed from flattened to flake-like. At this milling time, due to work hardening of the ductile Al powders, along with cold welding of the particles, fragmentation of particles happened with particle size reduction. Also, alumina particles have been moved in the cold welded Al and confined between them as shown in Figure 8 (b). At 45h milling time, Figure 8 (c) alumina clusters have been disappeared and distributed uniformly. Nanometer size particles of alumina are covered by Al nano size particles. The powder particles became more uniform in size compared to the early stages of milling.

3.4.1. Mechanical characterization

Mechanical testing is applied on the sintered consolidated specimens to make an evaluation of mechanical properties. Microhardness measurements and compression tests

were performed to characterize the composite materials. The values of microhardness, after compaction and sintering processes, for Al-20wt. %Al_2O_3 nanocomposite is 83 HV. Microhardness of aluminum is about 30 HV. So it is clear from the obtained result that Microhardness increased after the addition of reinforcement particles and milling. Increasing the high energy ball milling duration increases the deformation and work hardening of powders. Moreover, the presence of hard reinforcing particles might enhance the work hardening rate of the matrix resulting in increase in hardness. The general trend was observed by others [19, 20]

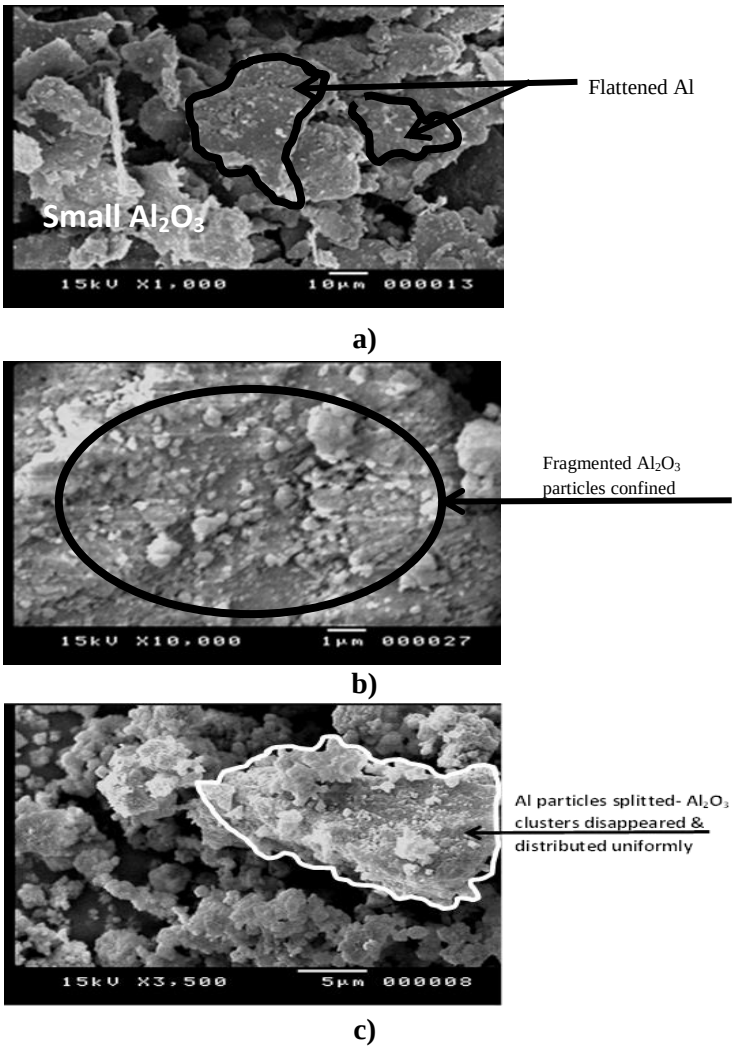


Fig. 8. SEM micrographs of Al–Al_2O_3 powders - 300rpm after (a) 7 h, (b) 30 h, (c) 45h.

Compression test is applied to determine the compressive strength of the material. Compression tests were performed and from the diameter and the applied loading conditions, stress-strain response of the material is calculated. The point of yielding is determined by drawing a line parallel to the initial linear region of the curve at a strain offset of 0.2 % strain. For Al-20wt.% Al_2O_3 nanocomposite, the compressive strength (CS

= 530 MPa) and the yield strength is calculated as (σy= 405 MPa). Figure 9 shows that a higher gain strength is obtained compared to the initial Al.

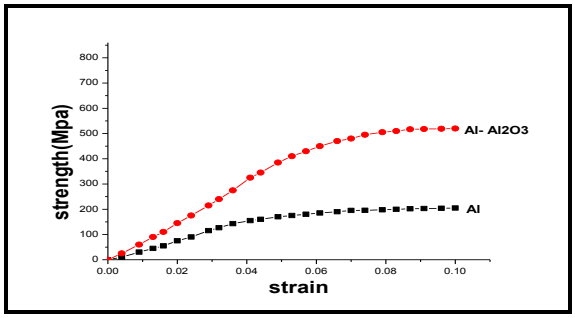


Fig. 9. Engineering stress-strain response of Al-Al_2O_3 nanocomposite and initial Al

Microhardness of aluminum is about 30 HV, while CS =300MPa and σy =200MPa. The increase in Microhardness and compressive strength in Al-20wt. % Al2O3 nanocomposite is attributed to the presence of hard reinforcing particles (Al2O3) and milling.

4. Conclusions

Study the evolution of the crystallite size and the lattice strain for milled aluminum composite powders (Al–Al2O3) is important in nanocomposites synthesis. The following conclusions can be made:

1- It's noted that the milling induces a higher lattice strain and an evolution of the finest particles, mainly in the extended times of milling.
2- From WH method, it can be seen that the rate of the grain refinement continuously decreases reaching, after 45hrs. of milling, the value of 19.77 nm.
3- The lattice strain shows a continuous increase to value of 1.16%.
4- Measurements of grain sizes obtained by the peak broadening in XRD patterns and the direct TEM techniques are nearly the same.
5- The presence of Al_2O_3 reinforcing particles enhances the hardness (83 HV) and compressive strength.

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دراسات لحجم الحبيبات وإجهاد الشبكة البلورية في متراكب نانوني من الألومنيوم – ألومنيا المنتج بالطحن الميكانيكي عالي الطاقة

الملخص العربي:

إهتم هذا البحث بدراسة حساب حجم الحبيبات بعد وصولها إلى حجم النانو في سبيكة متراكبة نانوية (ألومنيوم-ألومينا) والتي تم تصنيعها باستخدام طريقة التكوين الميكانيكي للسبائك. وقد أسفرت النتائج عن أنه ونتيجة لعملية الطحن فإن حجم الحبيبات يقل بينما يزيد الانفعال في الشبكة البلورية. قياسات الحجم للحبيبات تمت بتطبيق إحدى الطرق الحسابية وهى طريقة "وليمسون هال" وذلك عن طريق تحليل الأنماط الناتجة من الاشعة السينية وعمل معالجة لها, وأيضا تمت بالقياس المباشر باستخدام الميكروسكوب الإلكتروني النافذ وكانت النتائج تقريبا متشابهة. وللوقوف على بعض الخواص الميكانيكية لهذه السبيكة المتراكبة النانوية أجريت بعض الاختبارات (لصلابة, تحمل الانضغاط على العينات المكبوسة الملبدة, والنتيجة أن استخدام الالومينا في تقوية قالب الالومنيوم, والتوزيع المنتظم لها أدى الى تحسن ملحوظ في الخواص الميكانيكية للسبيكة النانوية المتراكبة الناتجة.