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Short communication

Correlation between the minimum grain size obtainable by ball milling and lattice strain

Farghalli A. Mohamed

Department of Materials Science and Engineering, University of California, Irvine, CA 92697-2575, USA

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ABSTRACT

Primary among the top-down processing techniques that can be used to synthesize nanocrystalline materials is ball milling. During milling, the gain size decreases with milling time, reaching a minimum grain size, d_{min}. Minimum grain size attainable is a characteristic of each material. Ball milling introduces considerable amount of lattice strain, ε. Such a strain increases rapidly with time, reaching a maximum value that coincides with d_{min}. By applying a recently developed dislocation model, which relates, d_{min}, to several physical parameters (such as the stacking fault energy and hardness), and by adopting several fundamental concepts, which are related to deformation behavior in materials, it is shown that d_{min}/b depends on $(1/\varepsilon)^{0.94}$, where$b$is the Burgers vector.The validity of this functional dependence is examined by analyzing experimental data on several nanocrystalline materials.

1. Introduction

An effective approach to strengthen metallic systems such as Al and its alloys is via grain refinement. In general, as the grain size, d, is refined, the strength increases $1/\sqrt{d}$ according to the Hall-Petch relationship [1,2] that can be represented by:

$$\tau = \tau_o + c/\sqrt{d} \quad (1)$$

where τ is the flow stress, τ_o is a friction stress and c is a constant. Eq. (1) is also valid when the yield strength is replaced by the hardness, H ($H = 6\ \tau$).

There are two types of processes that can be used for grain refinement: Top-down processes and bottom-up processes. The former processes refer to processes that have been used for grain refinement by structural decomposition. In contrast to bottom-up processes, which are normally associated with low mass buildup rate and small sample dimension, top-down processes are deemed to be cost-effective and capable of producing nanocrystalline or ultrafine-grained materials in large quantities.

In the last two decades, considerable efforts have been devoted to exploring novel top-down processing techniques and to understanding the underlying mechanisms for structural decomposition. Among these methods, ball milling [3] has received considerable attention.

Ball milling induces heavy cyclic deformation in powders, and is consists of repeated welding, fracturing, and rewelding of powder particles [4,5]. As a result of this cyclic deformation during milling, materials experience severe impact deformation, resulting in grain refinement. Over the past several years, the characteristics of crystal refinement and development of nanostructures during ball milling have been studied extensively. These studies have led to several important findings. First, Oleszak and Shingu [6] have observed that the crystallite size decreases with milling time and that continuous milling leads to a minimum grain size, which is a characteristic of each metal. Eckert et al. [7] have suggested that the minimum average grain size, d_{min}, is obtained as a result of a balance between the formation of dislocation structure and its recovery by thermal processes. Third, Fecht [8] has proposed a phenomenological approach for grain refinement during milling that involves the following three stages: (a) the localization of a high dislocation density in shear bands, (b) the annihilation and recombination of dislocations, forming cells and subgrains (recovery), and (c) the transformation of subboundaries into high grain boundaries.

By utilizing the aforementioned findings and suggestions, a theoretical dislocation model [9], which quantitatively describes the dependence of the minimum grain size, d_{min}, on several physical parameters in a nanocrystalline (nc) material, was developed. The model may be represented by [9]:

$$d_{min}/b = A_3 \exp(-\beta Q/4RT)(D_{po} Gb^2/\nu_o kT)^{0.25}(\gamma/Gb)^{0.5}(G/\tau)^{1.25} \quad (2)$$

where b is the value of the Burgers vector, A_3 is a dimensionless constant, $\beta$is a constant, Q is the self-diffusion activation energy, R is the gas constant, T is the absolute temperature, D_{po} is the frequency factor for pipe diffusion, G is the shear modulus, ν_o is the initial dislocation

E-mail address: famohame@uci.edu.

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velocity, k is Boltzmann's constant, $\gamma$is the stacking fault energy and $\tau$is the shear stress. The above equation was developed by combining elements of dislocation theory with well-established fundamental concepts and observations. As shown by Eq. (2), the model predicts that the minimum grain size, $d_{\min}$, depends on hardness, stacking fault energy, and an exponential function of the activation energy for recovery. These predictions were initially found to be in good agreement with experimental data reported for nc-FCC and nc-BCC metals [9]. In addition, the model was extended to nc-HCP metals [10]. Furthermore, it has been demonstrated [11,12] that the dislocation model for ball milling is quantitatively applicable to the description of various severe plastic deformation (SPD) processes such as Equal Channel Angular Pressing (ECAP) and high-pressure torsion (HPT), regardless of differences in the mechanics and kinetics of the process, and the range of the minimum grain size produced by each process

According to several reports [6,7,13], ball milling leads to a considerable enhancement of the lattice strain and such a strain increases rapidly, reaching a maximum value that coincides with the minimum grain size, $d_{\min}$. The broadening of Bragg peaks in the x-ray diffraction patterns of powder after milling is the result of both the small size of the diffracting grains and the lattice strain (Atomic level strain). The equation that describes the dependence of the measured peak width, $(\delta s)_o$ on the volume averaged grain size, d, the lattice strain, ε, and the reciprocal space variable, $s = 2\sin\theta/\lambda$, can be represented by [14]:

$$\frac{1}{(\delta s_o)} = \mathrm{d} - 4.\ \varepsilon^2 d\left[\frac{s}{\delta s_o}\right]^2 \tag{3}$$

$$Y = A - BX$$

On the basis of Eq. (3), the grain size, d, and the lattice strain, ε (root mean square (rms) strain) can be determined by (a) plotting Y against X for all of the measured peaks, and (b) performing a least-square fit.

It has been reported [7,15] that lattice strain as estimated from the x-ray broadening increases with decreasing grain size and that the maximum lattice strain increases with decreasing the minimum grain size. For example, maximum lattice strains in Al whose d_{min} is 25 nm and Ir whose d_{min} is 6 nm are 0.18% and 0.9%, respectively [7]. Despite this information, there is at present no quantitative analysis that can predict the relation between lattice strain, ε, and d_{min}. Accordingly, it is the purpose of this note to examine whether this relation can be established on the basis of Eq. (1) and fundamental concepts.

In order to explore the possibility of establishing a quantitative correlation between the minimum grain size, d_{min}, and lattice strain, ε, the following assumptions and approximations are made along with the following relationships:

(a) During milling, there are three sources that contribute to lattice strain: (i) dislocation density, (ii) grain boundaries, and (iii) Fe introduced during milling. Based on experimental evidence, it is suggested [7] that dislocation density, ρ, is the dominating contributor to lattice stain.

(b) The dislocation density is rated to stress by the following well-known expression [16]:

$$\tau/G = \alpha \mathrm{b}\sqrt{\rho} \tag{4}$$

Where α (~ 0.5) is a constant.

(c) As mentioned earlier, the grain size decreases with time during milling. This observation was reported initially by Oleszak and Shingu [6] and later was noted in other investigations [4–7]. Such a decrease in the grain size is related to the deformation energy provided by milling. This energy is a measure of deformation via dislocation multiplication and motion. The deformation energy per unit volume as a function of applied stress can, to a first approximation, be given by [9]

$$U = \int \tau d\varepsilon \tag{5a}$$

Table 1
Structural Properties of ball milled powders.

Metal or alloy	Strain (%)	$d_{\min}$ (nm)	b (nm)
Al (FCC)	0.18	25	0.286
Al 6061 (FCC)+	0.25	23	
Al 5083 (FCC)+	0.25	22.5	
Al-7.5Mg (FCC)+	0.2	22	
Ag (FCC)	0.16	25	0.288
Cu (FCC)	0.2	20.5	0.255
Ni (FCC)	0.4	11.7	0.249
Pd (FCC)	0.7	7	0.275
Rh (FCC)	0.3	7	0.267
Ir (FCC)	0.9	6	0.271
Fe (BCC)	0.35	8.1	0.248
W (BCC)	0.5	7	0.274

+The value of the Burgers vector, b, was taken equal to that of Al.

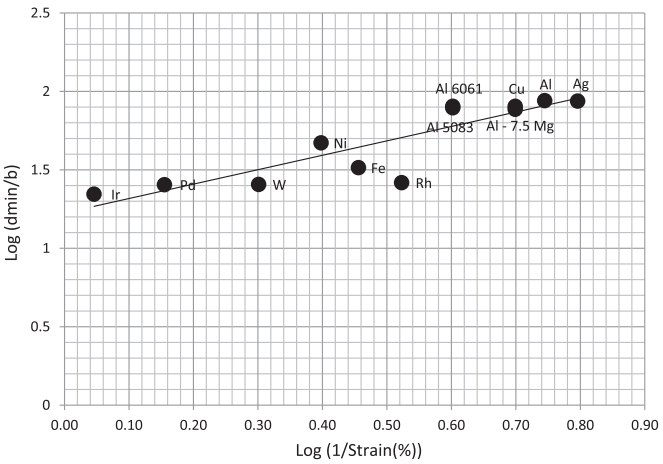

Fig. 1. Plot of the logarithm of the normalized minimum grain size, d_{min}/b, against the logarithm of the reciprocal of the lattice strain.

It has been shown that the relation between stress, τ, and strain, ε, for many polycrystalline materials is parabolic [16] and may be expressed [17] as: $\tau = c_1 G\varepsilon^{0.5}$, where c_1 is a constant. By substituting for τin Eq. (5a), integrating from 0 to $\varepsilon$and writing the result of the integration in terms of ε, one obtains the following equation:

$$U = c_1 G\varepsilon^{1.5} \tag{5b}$$

(d) The Energy of dislocations per unit length can be expressed as [17]

$$E = c_2 G b^2 \tag{6}$$

where c_2 is a constant related to dislocation distribution and its characteristic (edge and screw).

(e) The total length of dislocations per unit volume is equal to twice the dislocation density, ρ (the number of dislocations per unit area) [18]

(f) The energy of dislocations per unit volume based on (d) and (e) can be written as [19]:

$$U = c_2 G b^2 \rho \tag{7}$$

Having provided the above information, attention is now placed on the correlation between the minimum grain size obtainable from ball milling, d_{min}, and lattice strain, ε. This correlation can be made via three steps:

Step 1: By comparing Eqs. (5b) and (7), dislocation density, ρ, can be expressed as:

$$\boldsymbol{\rho = c_3(\varepsilon^{1.5}/b^2)} \tag{8a}$$

where c_3 is a constant.

Then, by taking the root square of Eq. (8a), Eq. (8a) becomes:

$$\sqrt{\rho} = c_4(\varepsilon^{0.75}/b) \tag{8b}$$

where c_4 is a constant.

Step 2: Combining Eqs. (4) and (8b) to remove ρ leads to the following result:

$$\tau/G = c_5\varepsilon^{0.75} \tag{9}$$

where c_5 is a constant.

Step 3: By substituting for τ/G from Eq. (9) into Eq. (2), the following relation is obtained:

$$d_{min}/b = \mathrm{A}\exp\ (-\beta Q/4RT)(D_{po}Gb^2/\nu_o kT)^{0.25}(\gamma/Gb)^{0.5}(1/\varepsilon^{0.75})^{1.25} \tag{10a}$$

Eq. (10a) can be written in the following final form:

$$d_{min}/b = \mathrm{A}\exp(-\beta Q/4RT)(D_{po}Gb^2/\nu_o kT)^{0.25}(\gamma/Gb)^{0.5}(1/\varepsilon)^{0.94} \tag{10b}$$

Eq. (10b) predicts an almost linear relation between the minimum grain size obtainable from ball milling, d_{min}, and lattice strain, ε.

In order to examine quantitatively the validity of the above prediction, data on d_{min} and ε that are given in Table 1 were used. The data are available for seven FCC metals (Ag, Al, Cu, Ir, Ni, Pd, and Rh), 3 FCC alloys (Al-6061, Al-5083, and Al-7.5 Mg), and two BCC Metals (Fe and W). The values of d_{min} and ε were taken from references [6, 7,9]. Also included in the table are the values of the Burgers vector (9).

According to Eq. (10b), when the logarithm of the normalized minimum grain size, d_{min}/b, is plotted against the logarithm of the reciprocal of the lattice strain, ε, straight line having a slope of 0.94 results. The data in Table 1 are plotted using this presentation in Fig. 1. An examination of the figure reveals two characteristics: (i) the data of all metals scatter about a straight line, which represents the best fit (using the least-square method), and (ii) the slope of this line is 0.91. These two characteristics agree well with the prediction of Eq. (10b).

In summary, studies on ball milling have reported two primary findings: (a) the crystallite size, d, decreases with milling time and continuous milling leads to a minimum grain size, $d_{min,}$ which is a characteristic of each metal; and (b) the lattice strain increases rapidly, reaching a maximum value that coincides with the minimum grain size, d_{min}. In the present investigation, it is demonstrated that the relation between the minimum grain size obtainable from ball milling, d_{min}, and lattice strain, ε, can be described by the following equation:

$$d_{min}/b = B(1/\varepsilon)^{0.94}$$

where B is a parameter that depends on factors such the stacking fault energy and the activation energy for recovery. An analysis of experimental data reported for ball milling of several metals and alloys show that when the logarithm of the normalized minimum grain size, d_{min}/b, is plotted against the logarithm of the reciprocal of the lattice strain, ε, the plot results in a straight line having a slope of 0.91, in agreement with the prediction of the above developed equation.

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Data availability statement

The raw data required to reproduce these findings cannot be shared at this time as the data also forms part of an ongoing study

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