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Size dependence of elastic mechanical properties of nanocrystalline aluminum

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ABSTRACT

The effect of grain size on the elastic mechanical properties of nanocrystalline pure metal Al is quantified by molecular dynamics simulation method. In this work, the largest nanocrystalline Al sample has a mean grain size of 29.6 nm and contains over 100 millions atoms in the modeling system. The simulation results show that the elastic properties including elastic modulus and ultimate tensile strength of nanocrystalline Al are relatively insensitive to the variation of mean grain size above 13 nm yet they become distinctly grain size dependent below 13 nm. Moreover, at a grain size < 13 nm, the elastic modulus decreases monotonically with decreasing grain size while the ultimate tensile strength of nanocrystalline Al initially decreases with the decrease of the grain size down to 9 nm and then increases with further reduction of grain size. The increase of ultimate tensile strength below 9 nm is believed to be a result of an extended elasticity in the ultrafine grain size nanocrystalline Al. This study can facilitate the prediction of varied mechanical properties for similar nanocrystalline materials and even guide testing and fabrication schemes of such materials.

1. Introduction

Nanocrystalline metals and alloys – with mean grain sizes < 100 nm – have strengths exceeding those of coarse-grained and even alloyed metals [1,2], and are thus expected to have many applications [3]. For example, pure nanocrystalline Al (with mean grain size of about 50 nm) has yield strength at the level of 500 – 600 MPa [4,5], which is over 10 times higher than that of the conventional coarse-grained polycrystalline Al (e.g. with mean grain size of about 40 μm) [5]. In general, it is observed that the yield strength increases with the decrease of the mean grain size larger than about 100 nm [6]. However, when the grain size is below about 20–10 nm, the yield strength instead decreases with decreasing the grain size. This is commonly known as the Hall–Petch breakdown [7,8]. Clearly, the mechanical properties and deformation of metals and alloys are strongly influenced by their grain sizes, especially at the nanometer scale [9,10].

In order to design nanocrystalline metals and alloys with optimum and/or tunable mechanical properties is it important to quantify their grain size dependence. The ability to completely characterize the grain size dependence of mechanical properties should also lead to the development of better materials-physics-based manufacturing and processing. However, it is not feasible in experiments to precisely control the grain size level in the synthesized nanocrystalline bulk samples. It is also challenging to fabricate ultrafine (< 5–10 nm) grain sized nanocrystalline bulk materials with densities close to theoretical levels using modern technologies. This is because of the existence of nanopores or nanovoids in the sintered nanocrystalline bulk materials resulted from various processing [11]. Therefore, a complete grain size dependence of mechanical properties cannot be easily obtained for nanocrystalline bulk materials using experimental techniques. However, computational materials modeling (especially atomistic simulations) is capable of providing robust and accurate measurements of the grain size dependence of mechanical properties for different nanocrystalline materials.

Aluminum, one of the most widely used lightweight metals, and its alloys are materials with promising continuing applications to meet the future challenge of pollution reduction and energy efficient transportations [12]. Thus, there is an urgent need to design and develop ultra-strong Al based alloys. Nanostructuring is considered as one of the most efficient ways to improve the mechanical properties of material systems [13]. In the literature, there are a number of scattered reports by both experiments and theoretical approaches to study the size effect of mechanical properties [4,5] and deformation mechanisms [14,15] in nanocrystalline pure metal Al, but no complete grain size dependence has been reported so far. In this work, we employ classical molecular dynamics (MD) simulations to quantify the grain size dependence of mechanical elastic properties of a range of samples of nanocrystalline pure metal Al. The complete knowledge of grain size dependence of

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mechanical properties of nanocrystalline pure metal Al will provide important fundamental basis for the design and development of Al based alloys with strong and/or tunable mechanical properties.

2. Simulation and analysis methods

2.1. Preparation of nanocrystalline samples

A set of 14 different 3-dimentional (3D) nanocrystalline atomic structures are generated using a Voronoi construction [16]. Each 3D nanocrystalline sample is cubic in shape and consists of 20 grains with a lognormal size distribution. As the side length of the cubic sample increases from 5 nm to 120 nm, the nanocrystalline sample increases in the number of atoms from 7.31×10^3 to 1.04×10^8. The mean grain size (d) is found to vary from 1.1 to 29.6 nm. In addition, a cubic sample of single crystal pure metal Al with side length as 40 nm (5.00×10^5 atoms) is also created and studied by MD for comparison purpose. In this study, MD simulations are performed using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) open-source code [17]. For the MD modeling, we use the embedded atom method (EAM) potential developed by Mishin et al. [18] established on the basis of experimental data and *ab initio* calculations.

All of the nanocrystalline and single crystal samples are relaxed to equilibrium configurations at 300 K with a pressure of 0 bar by using a Nose/Hoover type equation of motion sampled from isothermal-isobaric (NPT) ensemble. Simulation times of 50–100 ps (i.e. 50,000 – 100,000 steps with a time step of 0.001 ps) are found to be adequate for such relaxations. Periodic boundary conditions (PBC) are also employed. Fig. 1(a) and (b) show two representative nanocrystalline samples after relaxation containing mean grain sizes (d) of 2.2 nm (5.94×10^4 atoms) and 13.4 nm (1.30×10^7 atoms), respectively. Royal-blue atoms with FCC crystal arrangement possess coordination number of 12, the rest of the atoms (e.g. light blue, yellow, red, etc.) have non-12 coordination numbers and usually are located at the grain boundary regions. It is noticed the equiaxed (spherical like) shape grains in Fig. 1(a) change to polyhedral grains in the nanocrystalline sample when the mean grain size is increased from 2.2 to 13.4 nm. This is in agreement with various experimental observations (e.g. TEM characterization) on the grain morphology of nanocrystalline materials with varied mean grain sizes [3].

2.2. Structural analysis of nanocrystalline samples

The statistical structure parameters of nanocrystalline samples as a function of mean grain size are computed, including mass density, atomic fraction of GBs, and the average coordination number, as shown in Fig. 2(a), (b), and (c), respectively. First of all, all of these structural parameters show distinct size dependence with respect to the mean grain size. Especially, when the mean grain size is reduced to below 15–10 nm, the size dependence of each structural parameter becomes more significant. The computed mass density in the single crystal Al is 2.67 g/cm^3, this is in close agreement with the experimental density of Al – 2.70 g/cm^3 [19]. The mass densities of the nanocrystalline samples are overall smaller than that of the single crystal sample (see Fig. 2a). The relative densities of nanocrystalline samples with mean grain sizes of 20 nm and 10 nm are found to be 99.6% and 99.2%, respectively. These relative densities are generally higher than that of the experimental nanocrystalline samples with similar grain size levels. For example, the relative densities of nanocrystalline Se [20] and Ni [3] are 98.2% and 94.0% with mean grain size of 20 nm and 10 nm, respectively. This is probably due to the existence of nanopores or sub-nanoscale porosities in the experiments resulted from various processing of nanocrystalline samples [11]. When the mean grain size is below a few nanometers, the relative density in the nanocrystalline sample decreases significantly, it is calculated that the relative densities of nanocrystalline samples with mean grain sizes of 5 nm and 1 nm are 98.7% and 96.3%, respectively.

The reduced density in nanocrystalline samples is a direct consequence of the increased atomic fraction of grain boundary (GB) atoms [21,22]. As shown in Fig. 2(b), the atomic fraction of GB atoms in the nanocrystalline sample increases with the decrease of mean grain size. When the mean grain size is reduced to lower than 15–1 nm, more than 5–50% of atoms locate at the GB regions in the nanocrystalline sample. The large atomic fraction of GB atoms (or volume fraction of GBs) in the nanocrystalline samples possesses non-12 coordination numbers (the single crystal Al is of FCC structure with coordination number as 12) therefore has disordered atomic arrangement. As a result, the atomic density at the GB regions in the nanocrystalline sample is lower than that of in a perfect crystal. As shown in Fig. 2(c), the average coordination number of the nanocrystalline sample is less than that of a single crystal and it deceases significantly with decreasing the mean grain size. This distinct grain size dependence of the

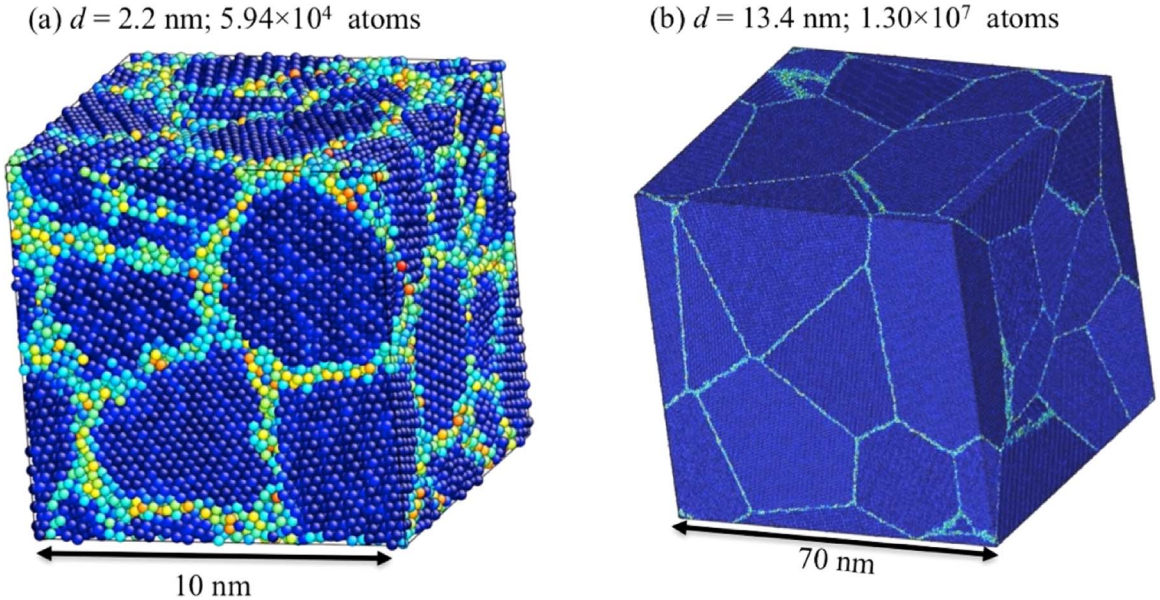

Fig. 1. Nanocrystalline Al bulk samples with representative different mean grain sizes d: (a) d =2.2 nm and (b) d =13.4 nm. Royal-blue atoms with FCC crystal arrangement possess coordination number of 12, the rest of the atoms (e.g. light blue, yellow, red, etc.) have non-12 coordination numbers and usually are found at grain boundary regions. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

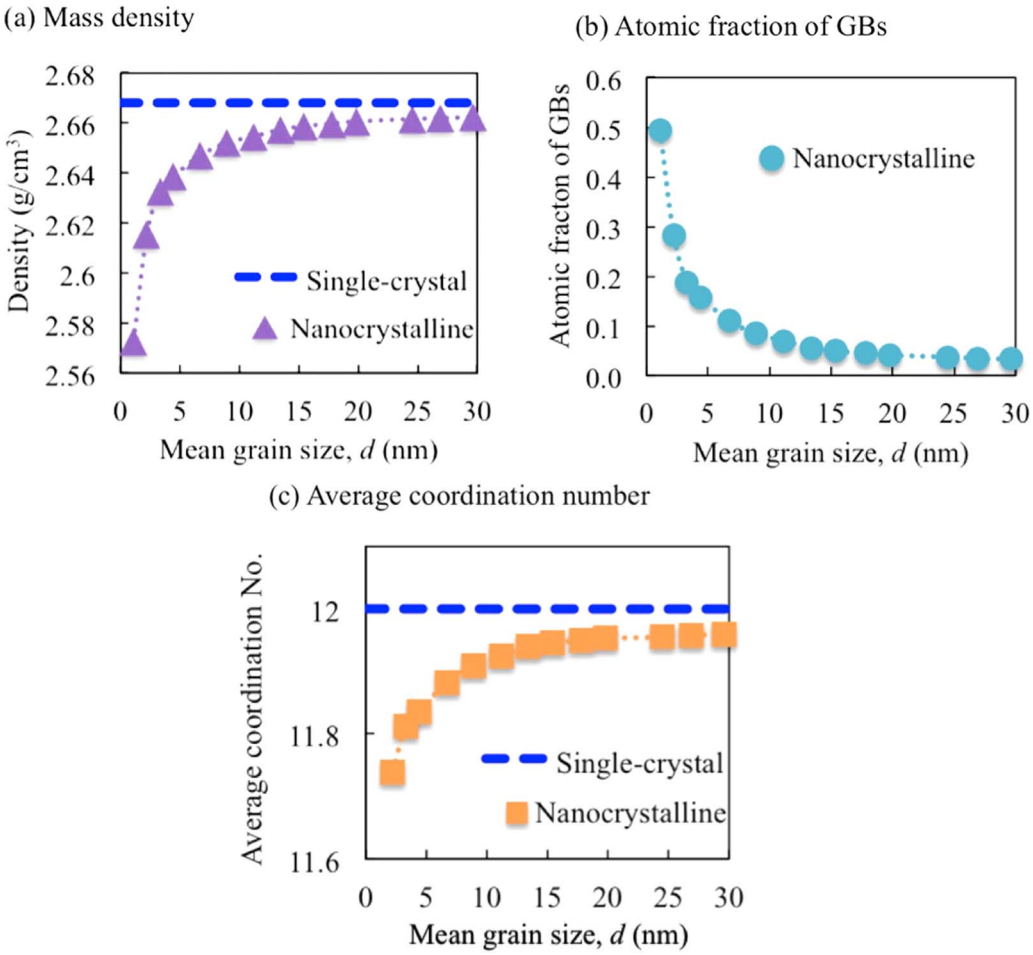

Fig. 2. Structural parameters of nanocrystalline Al bulk samples as a function of mean grain size *d*: (a) mass density; (b) atomic fraction of grain boundaries (GBs); (c) average coordination number. The blue dashed lines in (a) and (b) indicate the levels of corresponding structural parameter values of the single crystal Al bulk sample. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

nanocrystalline structures are the fundamental reasons that lead to size dependent physical, chemical and mechanical properties in nanocrystalline materials [23,24].

2.3. Uniaxial tensile simulation

As shown above, the EAM potential used in this MD modeling work can nicely represent various structural parameters of nanocrystalline and single crystal bulk Al. Consequently, we carried out MD simulations of uniaxial tension deformation along *x*- axis for the 14 nanocrystalline and an additional single crystal samples. Periodic boundary conditions are used. The uniaxial tension load is then simulated by changing the length of the simulation model along the *x*- axis. For each nanocrystalline sample, a uniform strain field along the required direction is accomplished by repeatedly scaling the corresponding unit cell and the atomic positions by a factor of 1.01 of the initial coordinates, and then relaxing the model for 50 ps in between rescaling steps. This step of tensile/relaxation is repeated until a strain of 0.5 was reached. The tensile strain here is the engineering strain, defined as $\varepsilon=\Delta L/L_0$, where ΔL is the change relative to the original length L_0. In this work, a strain rate of 10^{10} s^{-1} is applied to all nanocrystalline samples to investigate the nanosize effect on the elastic properties of nanocrytalline Al. During deformation, the lateral boundaries of each sample are kept constant at zero pressure. For comparison, the same MD simulation has been done for the single crystal Al. This tensile method is comparable to techniques reported previously in the study of silica nanowires [25] and metallic nanowires [26].

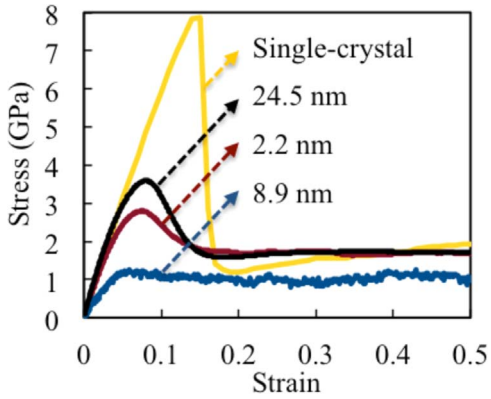

Fig. 3. Size dependence of stress–strain curves of the single crystal and nanocrystalline Al bulk samples with varied mean grain sizes at a strain rate of 10^{10} s^{-1}.

3. Results and discussion

Fig. 3 shows a number of representative stress–strain curves of nanocrystalline samples with comparison to the case of single crystal sample. The tensile behavior is similar in all cases before reaching the ultimate tensile strength (UTS) that corresponds to the peak value of the stress–strain curve. At the initial stage, the value of each stress–strain curve increases linearly but with different slopes up to different levels (or yield points) in different nanocrystalline samples. This behavior corresponds to the purely elastic region. The different slopes

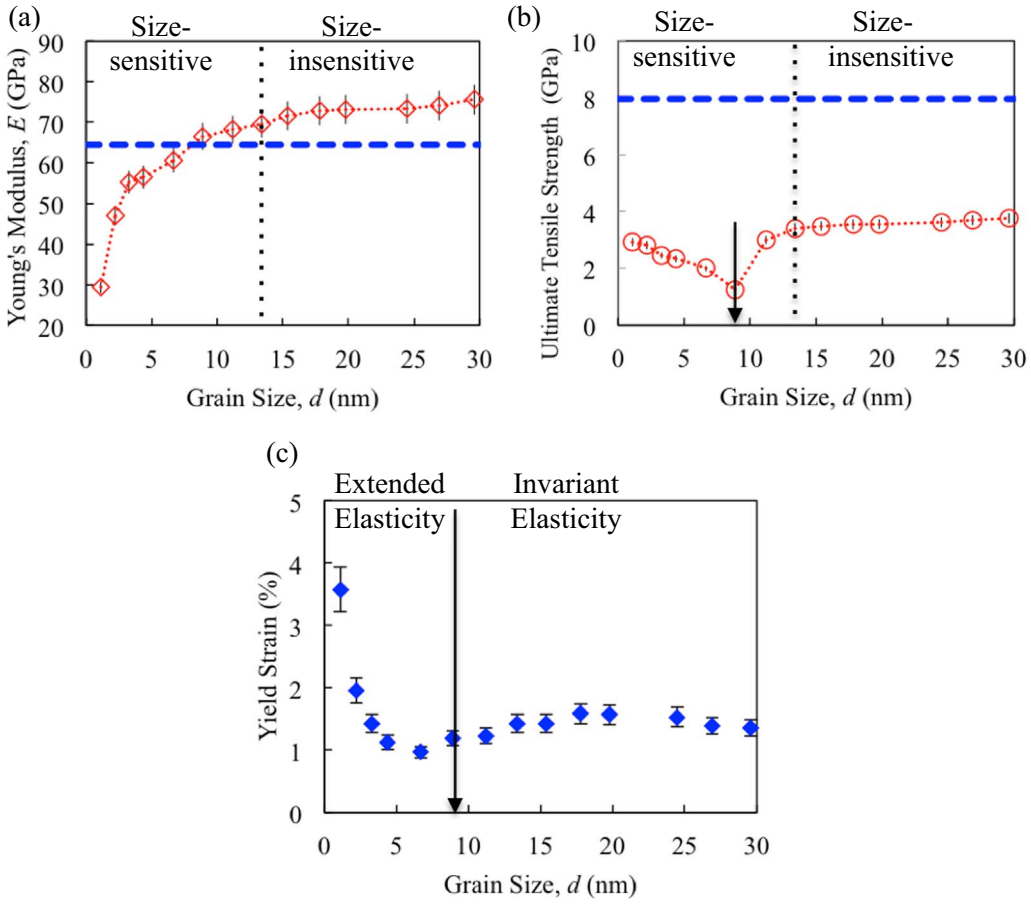

Fig. 4. Size dependence of elastic properties with error bars indexed for the nanocrystalline Al samples: (a) elastic modulus (E); (b) ultimate tensile strength; (c) yield strain. The blue dashed lines indicate the levels of corresponding property values of the single crystal sample. The strain rate is 10^{10} s^{-1} in the MD modeling. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

indicate different elastic moduli in different nanocrystalline samples. According to Hooke's Law, by numerical fitting with a linear function "Stress = Elastic Modulus×Strain" [27], the elastic moduli can be calculated for all the nanocrystalline and single crystal samples.

Fig. 4(a) shows the grain size dependence of elastic moduli of nanocrystalline samples, the value of elastic modulus for the single crystal sample is indicated by the blue dashed line. There are two regions in the grain size dependence of elastic moduli of nanocrystalline samples: one is grain size insensitive region at larger grain sizes (e.g. > 13 nm), the other is grain size sensitive region at small grain sizes (e.g. < 13 nm). In particular, with decreasing the grain size in the nanocrystalline sample, the elastic modulus remains almost invariant down to about 13 nm with a value about 75 GPa, which is 17.2% higher than that of the single crystal sample. This agrees very well with the experimental measurements (see Table 1) [28]. The MD computed elastic modulus of nanocrystalline sample with a mean grain size of 29.6 nm is 75.6 GPa, which is very close to the experimental value of 76.1 GPa measured for a nanocrystalline Al bulk with a mean grain size of 54.3 nm [28]. At grain size below 13 nm, the elastic modulus decreases remarkably with further reducing the grain size. This indicates a significant 'softening' of the nanocrystalline sample at ultra small grain size (< 13 nm) since the elastic modulus is a measure of stiffness of a solid material.

After the initial linear region in the stress–strain curves (see Fig. 3), all the nanocrystalline and single crystal samples undergo plastic deformation until the UTS is reached in each case. Fig. 4(b) shows the MD modeled UTS as a function of grain size of nanocrystalline samples at a strain rate of 10^{10} s^{-1}. By performing the same MD tensile test modeling as that for nanocrystalline samples, we have obtained the

Table 1
Elastic modulus Al bulk in the form of single crystal, nanocrystalline (mean grain size d, unit: nm), and conventional coarse-grained polycrystalline by MD calculations and/or experimental measurements.

Al bulk	Elastic Modulus (GPa)	
	Current MD	Previous Exp.
Single crystal	64.4	67.2–69.5 [29]
Coarse-grained polycrystalline	/	70.0 [30] 70.6 [31]
Nanocrystalline (d)	75.6 (29.6 nm) 67.0 (11.1 nm)	76.1 [28] (54.3 nm) 60.2 [31] (11.1 nm)

UTS for the single crystal sample, as indicated by the blue dashed line in Fig. 4(b). The MD computed UTS of single crystal Al agrees well with the reported value (8.3 GPa [32]) in the literature by MD modeling at the same strain rate of 10^{10} s^{-1}. In addition, similar in the case of elastic modulus, the UTS is independent (or insensitive) on the grain size when the mean grain size is larger than 13 nm. However, when the mean grain size in nanocrystalline sample is lower than 13 nm, the UTS becomes significantly size dependent with further reducing the grain size. It is interesting to notice that, at grain size < 13 nm, the UTS of nanocrystalline sample initially decreases then increases again with a transition point corresponding to a grain size of 9 nm, as indicated by the solid arrow in Fig. 4(b).

At the region of grain size between 13–9 nm, we think that the decrease of UTS of nanocrystalline sample is probably due to some

dislocations sink or disappear at GBs during the process of tensile deformation [9,33]. In other words, not all of the dislocations tend to piling-up against the GBs to strengthen the material at the grain size between 13 nm and 9 nm. As shown via Supplementary animation (I), it is observed that, in the case of tensile deformation of a nanocrystalline Al bulk sample with mean grain size of about 13.4 nm, some dislocations sink at the grain boundary regions while the rest of the dislocations stabilize within the grain interior regions. This nanoscale deformation and strengthening mechanisms have been observed and reported in a large number of nanocrystalline samples in the literature [9,33].

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However, when the grain size is smaller than 9 nm, the UTS increases with further reducing the grain size in the nanocrystalline sample. This is found to relate with an extended elasticity in nanocrystalline samples with ultra small grain sizes. As shown in Fig. 4(c), the elasticity of nanocrystalline sample remains almost invariant at mean grain sizes larger than 9 nm (as indicated by the solid arrow in Fig. 4c), whereas, it is increased significantly with further reducing the grain size. The extended elasticity (region between 9 and 1 nm in Fig. 4c) in the nanocrystalline sample with ultra small grain size results in the increased UTS under tensile load. As shown in Supplementary animation (II), in the case of tensile deformation of a nanocrystalline Al bulk sample with mean grain size of about 2.2 nm, one observes that all the dislocations eventually disappear at the grain boundary regions. This is a direct consequence of the fact that the equilibrium distance between dislocations is larger than the grain size in this nanocrystalline sample. As a result, the elasticity region of nanocrystalline Al bulk sample is largely enhanced during tensile deformation. The extended elasticity has also been observed in various nanostructured pure metals using the widely available nano-indentation facilities [34]. Furthermore, Kuan et al. [34] reported that the elastic elongations (or strains) of nanoscale thin films Mg, Zr, Al, and Cu are measured to be about 3%, 6%, 5%, and 4%, respectively. This is in reasonable agreement with the present MD modeling results that the elastic strain is between 1–4% in different nanocrystalline Al samples with grain size ranging from 30 nm to 1 nm.

Supplementary material related to this article can be found online at http://dx.doi.org/10.1016/j.msea.2017.03.065.

4. Conclusions

In the present work, the effect of grain size on the mechanical elastic properties of the nanocrystalline pure metal Al has been quantified by employing MD modeling. The grain size dependence of elastic modulus and ultimate tensile strength of nanocrystalline Al only becomes distinct at grain size small than about 13 nm, above which these elastic properties are almost invariant with the change of grain size. In addition, at grain size < 13 nm, the elastic modulus decrease significantly with decreasing the grain size, while the ultimate tensile strength of nanocrystalline Al decreases with the decrease of grain size down to 9 nm but increases with further reducing the grain size. The increase of ultimate tensile strength below 9 nm is due to an extended elasticity in the ultrafine grain size nanocrystalline Al. We expect that the quantifications of grain size dependence of mechanical properties will have implications in the development of Al based nanostructured alloys for high performance structural applications with relevance to transportation systems.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.msea.2017.03.065.

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