Influence of Competition between Intragranular Dislocation Nucleation and Intergranular Slip Transfer on Mechanical Properties of Ultrafine-Grained Metals

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Huge-scale atomistic simulations of shear deformation tests to the aluminum polycrystalline thin film containing the Frank–Read source are performed to elucidate the relationship between the inter- and intragranular plastic deformation processes and the mechanical properties of ultrafine-grained metals. Two-types of polycrystalline models, which consist of several grain boundaries reproducing easy and hard slip transfer, respectively, are prepared to investigate the effect of grain boundary on flow stress. While the first plastic deformation occurs by the dislocation bow-out motion within the grain region for both models, the subsequent plastic deformation is strongly influenced by the resistance of the slip transfer by dislocation transmission through grain boundaries. The influence of the competition between the intragranular dislocation nucleation and intergranular slip transfer on the material strength is considered. The nanostructured material’s strength depending on local defect structures associated with grain size and dislocation source length is assessed quantitatively.

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1. Introduction

In general the plastic deformation characteristics of coarse-grained metals (generally grain size \(d > 1 \mu m\)) is determined by the average quantity of collective motion of dislocations, because the initial stage of the plastic deformation is related to the activation of many pre-existing dislocation sources with various slip systems and then the interaction between dislocations in different slip planes contributes to the hardening process. In contrast to the conventional coarse-grained metals, nanocrystalline metals (\(d < 100 \text{ nm}\)) have a dramatic increase in the volume fraction located in the grain boundary region; hence mechanical properties of nanocrystalline metals cannot be simply predicted based on assumptions of the average quantity of collective motion of dislocations in grains. For example, grain size softening known as the inverse Hall–Petch effect is captured experimentally1,2) and grain rotation, grain boundary sliding and thermal activated local shear are proposed to be an underlying deformation mechanism.3–6) Moreover, dislocation emission from grain boundaries is also an important deformation mechanism for nanocrystalline metals because it is difficult to construct any intragranular dislocation source due to the very limited individual grain’s space. As characteristic lengths of these elementary processes like grain boundary sliding, grain boundary migration, dislocation emission from grain boundaries are about submicron-meter, these processes can be directly expressed by atomistic simulations7–13) and the mechanism is becoming clear.14)

On the other hand, recent metal processing techniques allow the effective control of grain size and microstructure in metallic systems, improving bulk material properties. It has been possible to produce ultrafine-grained (UGF) metals (100 nm < \(d < 1 \mu m\)) in bulk by severe plastic deformation process15–17) and these UFG metals achieve unique and excellent mechanical properties.18–20) For example, UFG copper with a large number of nano-twins shows the coexistence of high strength and high ductility21,22) and UFG iron obtains the improved fracture toughness at low temperatures as compared to coarse-grained iron.23,24) Therefore, UFG metals are strongly expected for new structural materials. However it is difficult to explain why these excellent mechanical properties breaking the trade-off relationship between strength and ductility (toughness) appear in UFG metals by using conventional theories or obtained elementary processes in atomistic simulations because the competition between the intragranular dislocation motion and grain-boundary-mediated plastic deformation processes could strongly influence the mechanical properties of UFG metals. More specifically, we should consider the relationship between the dislocation multiplication from the Frank–Read (FR) source, dislocation emission from grain boundaries and dislocation transmission through grain boundaries. We believe that understanding and systematizing the complex phenomena expressed by combining elementary processes appearing in the grain size of between 100 nm and 1 \(\mu m\) will develop up a new science for UFG materials.

A number of analytic models25–27) and computational simulations28–31) of the mechanical properties of UFG metals have been reported. In particular, atomistic simulation is a very useful tool for investigating the complex defect interactions including grain boundaries. Many atomistic simulations of the interaction between lattice dislocations generated from an intragranular dislocation source and grain boundaries have been reported32–38) and are making clear the detail interaction mechanism. However, most simulations have the two dimensional structures and the critical stress to activate the intragranular dislocation source does not show the size effect like grain size dependence. Because the critical stress for the dislocation multiplication from the FR source in
UFG metals becomes clearly close to the material strength observed in experiments, it is essential to consider the size dependence of the dislocation generation from an intragranular dislocation source in atomistic simulations to investigate the unique mechanical properties of UFG metals.

In this study, to investigate the influence of the competition among two plastic deformation phenomena of dislocation multiplication from the FR source and dislocation transmission through grain boundaries on the mechanical properties of UFG metals, we perform huge-scale atomistic simulations, in which about a hundred million atoms are used for the shear deformation to aluminum polycrystalline models. Before performing the complex defect interaction simulations, we investigate the two elementary processes. Firstly, the critical shear stresses needed to bow-out motion of models. Before performing the complex defect interaction simulations, in which about a hundred million atoms are simulated region with a regular periodicity and the periodic boundary conditions are applied to the $x$ and $y$ directions, the dimensions of the simulation cell $l_x$ and $l_y$ in the $x$ and $y$ directions can be determined as $2d$ and $\frac{\sqrt{3}d}{2}$, respectively. This model includes four grains numbered as shown in Fig. 1(b). At first, four circular grains whose radius is slightly longer than the circumradius of hexagon are arranged in the Voronoi centers, and the grain regions are determined by Voronoi division technique. When the atoms around the boundary between grains are located closed to the other atoms within less than 70% of the distance to nearest neighboring, one of the pair atoms is eliminated.

We consider two types of grain boundaries models to control the resistance of dislocation transmission though grain boundaries. We call the two types of models “GBI model” and “GBII model”. In both models, the crystal orientation of the center grain is exactly the same; [110], [112] and [111] in the $x$, $y$ and $z$ directions, respectively, which are the same orientations of the FR model shown in Fig. 1(a). In the case of GBI model, the three other grains have the same [111] orientation axis in the $z$ direction and other orientations in the $x$ and $y$ directions are determined by rotating different angles around the $z$ direction from the center grain for each grain: 32.2° for grain 2, 52.7° for grain 3 and 27.8° for grain 4; hence all grain boundaries correspond to {111} tilt boundaries and all grains have the common (111) slip plane in the $z$ direction. The coincidence site lattice relations are formed at the boundaries between the central grain and other grains if they constructs the symmetric grain boundary plane, and the actual asymmetric grain boundary energies $\gamma^2$, $\gamma^3$ and $\gamma^4$ calculated using the EAM potential are 472, 497 and 473 mJ/m², respectively. On the other hand, in the case of GBII model, the three other grains have the same [100] orientation in the $z$ direction; therefore the grain boundary structures between the center grain and other grains are quite different from those of GBI model and the center grains and others have no common (111) slip plane. Consequently, we can regard GBI and GBII models as easy and hard slip transfer from the center grain to the neighbor grains, respectively. In this study, $d$ changes from 10 to 100 nm and $l_d$ is fixed to 35 nm in all models to investigate the grain size dependence of $\gamma^G$.

Finally, we combine the FR model with the GBI or GBII model to investigate the mechanical properties of UFG metals as shown in Fig. 1(c). We call these models “GBI-FR model” and “GBII-FR model”, respectively. In this study, $d$ is
fixed to 100 nm and $L$ is changed between 5 nm and 40 nm. The dimensions of the simulation cell and the total number of atoms are almost the same as GBI and GBII models.

2.2 Simulation procedure

Stable configurations of all models are obtained by the two processes. Firstly, a thermal equilibrium state of defect regions for each model is obtained by molecular dynamics simulation at 300 K via the velocity-scaling scheme and the analysis temperature gradually decreases to 0 K. Subsequently, the stable atomic configuration under zero stress at 0 K condition is finally obtained by the conjugate gradient method.

Simple shear deformation tests are performed by the conjugate gradient method. Two displacement control (DC) regions are set on the top and bottom region with about 1 nm width along the $z$ direction as shown in Fig. 1. Constant shear strain increment $\Delta \gamma_z$ of $7.5 \times 10^{-4}$ for the FR model or $5 \times 10^{-4}$ for other models is loaded step-by-step by controlling the $x$-directional displacement of atoms in the top-DC region. During the shear deformation tests, atomic displacements in all directions in the bottom-DC region are fixed and atomic displacements except for the $x$ direction in the top-DC region are not controlled. The fully relaxed configurations under each $\Delta \gamma_z$ are determined when the energy norms of all the atoms converged at 0.01 eV/Å or when the convergence step reaches a given value of 2000 step.

Atomic images shown in this paper are created using ATOMEYE visualization software, where only the atoms located on the defect region whose potential energies are higher than those of non-defect region are extracted. To perform huge-scale atomistic simulations, the original parallelized code based on message passing interface through 4096 cores is developed, and the maximum simulated region (0.24 x 0.21 x 0.035 µm³) is divided into 32 x 32 x 4.

3. Results and Discussions

3.1 Dislocation multiplication from Frank–Read source

Simple shear deformation tests are performed to FR models with the source length $L$ from 5 to 40 nm. Shear stress–strain curves are shown in Fig. 2(a). While stress–strain curves of different $L$ show the same slope of the elastic region, they show the different maximum shear stress. Each drop indicates the $t_{FR}^+$ for the bow-out motion with different $L$. The bow-out motion in case of $L = 5$ nm during shear deformation is visualized in Fig. 2(b). We can clearly observe that the bow-out motion from the upper FR source under shear deformation is expressed in the atomistic simulations and the FR source is recovered after enclosing the dislocation segment taking the shape into the dislocation loop. Consequently, dislocation multiplication from FR source can be reproduced with the atomic resolution. However, it should be noted that the pinning effect of the sessile dislocation on {112} plane on the bow-out motion is not perfect; namely the position of pinning point slightly migrates during deformation by changing the slip plane from {112} to {111} in localized regions of the sessile dislocations; hence the length between pinning points become slightly longer than the initial length $L$. Moreover, when the initial length of FR source is longer than 20 nm, the dislocation dipole tends to form a 60° dislocation dipole which is stable connection under deformation and is difficult to bow out. Therefore the actual dislocation length for bow-out motion becomes shorter due to the movement of pinning point. The relationship between $t_{FR}^+$ and the initial/actual lengths of FR source $L$ and the definition of the actual FR source length are shown in Fig. 2(c). The critical stress calculated by the simple Orowan model $t_{OR}^+$ is displayed in the black line for comparison. Because FR models are composed of two dislocation segments whose line-sense vector is opposite to one another, the effect of the other dislocation segment on $t_{OR}^+$ should be taken into account. Based on the assumption that the stress field containing dislocations can be expressed by the superposition of the analytical solution of the dislocation theory, we simply estimate the effect of the other segment on $t_{OR}^+$ by calculating the shear stress of the edge dislocation on the slip plane at the distance $h$ of the other dislocation of the double FR sources. The obtained maximum shear stress is about 200 MPa, so we suppose that this value is also necessary to move the dislocation line in addition to $t_{OR}^+$. The modified Orowan model including the contribution of the dipole effect is also shown in the red line in Fig. 2(c). While the bow-out stress $t_{FR}^+$ does not show the inverse relation to the initial dislocation length due to the change of pinning distance during the deformation, it decreases in definite inverse proportion to the actual dislocation length, that is $t_{FR}^+ \propto L^{-1}$. Moreover, the modified Orowan model can represent the results of atomistic simulations better than the simple Orowan model. Consequently, the size effect of the dislocation emission phenomenon from the intragranular dislocation source can be expressed by the double FR source model in atomistic simulations.

3.2 Influence of grain boundaries on activation of FR source

Before investigating the influence of the competition between intragranular dislocation motion and grain-boundary-mediated plastic deformation phenomena on the mechanical properties of UFG metals, the influence of grain boundaries on $t_{FR}^+$ is first investigated, because there is a possibility that the stress field of the dislocation bow-out is influenced by the grain boundary regions. A pair of FR sources as shown in Fig. 1(a) is inserted to each grain region of GBI models. All slip planes of FR sources in all grains are the same as (111) plane; hence the Peach-Koehler force acting on the mobile dislocations in FR sources located in the central grain becomes the maximum value under the applied shear deformation $\gamma_z$. As it is found from our preliminary study that the maximum stress for dislocation nucleation from grain boundaries is larger than $t_{FR}^+$, the first dislocation is activated from the FR source of the center grain.

Figure 3(a) shows the grain size dependence on $t_{FR}^+$ for GBI-FR models in which the initial source length $L$ is fixed to 20 nm. The values of $t_{FR}^+$ obtained by the FR models without grain boundaries are also presented by black dot line. Subsequently, we also investigate the influence of grain boundaries on the source length dependence on $t_{FR}^+$ for GBI-FR models that $d$ is fixed to 100 nm and the results are shown in Fig. 3(b), where the results of atomistic simulations for the
Fig. 1 Constructions of atomic models. (a) FR model. (b) GBI and GBII models composed of four regular hexagonal grains. (c) GBII-FR model ($d = 80$ nm and $L = 20$ nm).

Fig. 2 Bow-out behavior of Frank–Read source with various dislocation length $L$. (a) Shear stress–strain curves. (b) Dislocation configurations in case that $L$ is 5 nm. (c) Critical shear stress $\tau_{FR}$ in relation to the dislocation length $L$ calculated by atomistic simulations and the simple and modified Orowan models.

Fig. 3 Critical shear stress $\tau_{FR}$ in relation to (a) grain size $d$ and (b) actual FR source length $L$. 

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simple FR models and the analytical values based on the Orowan model are provided for comparison. In both cases as shown in Figs. 3(a) and 3(b), it cannot be clearly found the influence of grain boundaries on $\tau^{FR}$ in this study. However, our deformation simulations are performed at 0 K, so it is not easy for grain boundaries to change the stress field of the dislocation bow-out by the thermal activated local shear motion. Therefore, the dependence of grain boundaries on the $\tau^{FR}$ is still controversial issue.

3.3 Dislocation transmission through grain boundaries

To investigate the influence of the competition between intragranular dislocation motion and grain-boundary-mediated plastic deformation processes on the mechanical properties of UFG, shear deformation tests of GBI-FR and GBII-FR metals are performed. Grain size of all models is fixed to 100 nm and each model has different FR source length $L$ from 5 to 40 nm. As mentioned before, the former and latter models consist of grain boundaries having the ability of easy and hard slip transfer from the center grain to the neighbor grains, respectively, i.e., $\tau_{il}^{TM} > \tau_{il}^{FR}$.

Figures 4 and 5 represent the results of shear deformation tests of GBII-FR and GBI-FR models, respectively. Shear stress-strain behavior for various initial FR source lengths, defect configurations and stress distribution under shear deformation in case of GBII-FR model with $L = 5$ nm are shown in Figs. 4(a), 4(b) and 4(c), respectively. The same results for GBI-FR models are shown in Fig. 5. It should be noted that one FR source is introduced into only the center grain in GBII-FR model as shown in Fig. 4(b). However, in case of GBI-FR models with FR sources in all grains, other FR sources except for the center grain do not active during shear deformation as shown in Fig. 5(b); therefore we can ignore the influence of the number of FR sources on the mechanical properties when we compare the GBII-FR with GBI-FR models.

The first plastic deformation occurs by the dislocation bow-out motion in the center grain for all models; hence $\tau_{ij}^{GB}$ should also larger than any $\tau_{ij}^{FR}$. It is confirmed in Figs. 4(a) and 5(a) that the critical shear stress $\tau_{ij}^{GB}$ and $\tau_{ij}^{FR}$ for the incipient bow-out motion from the FR source in the center grain indicated red arrow is little different from $\tau_{ij}^{FR}$ of FR models as shown in Fig. 3(b). On the other hand, plastic deformation varies greatly after the first intragranular plastic deformation process. Namely, in all models, the first dislocation is activated from the FR source within the central grain region, and then the dislocation is absorbed into the grain boundary. The following plastic deformation is strongly influenced by the grain boundary structures.

In case of GBII-FR model, the propagation of plastic deformation by the dislocation transmission through grain boundaries does not occur after the first dislocation is trapped at the grain boundary as shown in Fig. 4(b). Instead, the second bow-out motion from the FR source continuously occurs. At this time stress relaxation occurs only in the central grain and the stress in other grain regions increases elastically as shown in Fig. 4(c). Consequently, the applied shear stress continues to increase because of the partially yielding phenomenon only by the intragranular plastic deformation of the center grain. On the other hand, in the case of GBI-FR models, a part of absorbed dislocation easily passes through grain boundaries as shown in Fig. 5(b). There is a clear distinction of stress distributions before and after dislocation transmission through grain boundaries as shown in Fig. 5(c). Firstly, a uniform stress distribution is observed before the dislocation bow-out from the FR source shown in the left picture of Fig. 5(c). Afterward the elastic strain energy stored in the neighboring grain is relaxed by the slip transfer shown in the right picture of Fig. 5(c). Additionally, dislocation transfer beyond the grain boundaries leads to continuous yielding behavior shown in Fig. 5(a), that behavior is quite different from the GBII-FR model.

Figure 6 shows the influence of the FR source length $L$ on the critical shear stress to activate the FR source $\tau_{ij}^{FR}$ or $\tau_{ij}^{TM}$ and flow stress $\tau_0^{FM}$ of each model when shear strain is 0.07. As mentioned before, although $\tau_{ij}^{FR}$ and $\tau_{ij}^{TM}$ show the same dependence of $L$, $\tau_0^{FM}$ and $\tau_0^{FR}$ show the quite different dependences of $L$. The reason can be explained by the relationship between resistances of intragranular dislocation generation $\tau_{ij}^{TM}$ and intergranular slip transfer $\tau_{ij}^{TM}$. In case of GBII-FR models with little $L$ dependence of $\tau_0^{FM}$, the slip transfer does not occur when shear strain is 0.07 as shown in Figs. 4(b) and 4(c); therefore we can estimate that $\tau_{ij}^{TM}$ is much higher than $\tau_0^{FM}$. In such the situation that $\tau_{ij}^{TM} > \tau_0^{FM}$, the flow stress can be controlled not by the intragranular dislocation nucleation phenomenon but by the intergranular slip transfer phenomenon. On the other hand, in case of GBI-FR models where slip transfer has already occurred when $\gamma_{cr} = 0.07$, each flow stress takes a constant value from $\gamma_{cr} = 0.06$ to 0.075 as shown in Fig. 5(a) and the flow stress $\tau_0^{FM}$ shows the clear $L$ dependence as shown in Fig. 6. In the case of $L$ beyond 20 nm, the flow stress $\tau_0^{FM}$ takes almost a certain value as observed in GBII-FR models and the values of $\tau_0^{FR}$ are higher than $\tau_0^{FM}$; hence the resistance of plastic propagation phenomenon by the intergranular slip transfer could control the material strength. Although the critical slip transfer stress $\tau_0^{TM}$ actually obtained by superposing the applied stress and the stress generated by piled-up dislocations, no dislocation pile-up occurs when the first slip transfer phenomenon occurs in GBI-FR models; thus it can be supposed that $\tau_0^{FM}$ is closely related to the $\tau_0^{TM}$. When $L$ decreases below 20 nm, the flow stress $\tau_0^{FM}$ begins to increase and becomes close to $\tau_0^{FR}$; hence it can be estimated that $\tau_0^{FR}$ becomes larger than $\tau_0^{TM}$ with decrease in $L$ and the intragranular dislocation nucleation phenomena could control the material strength.

Therefore, it is suggested by the present simulation of polycrystalline model that the material strength can be determined according to the relationship between $\tau_{ij}^{FM}$ and $\tau_{ij}^{FR}$, that is, the material strength is dominated by the intragranular dislocation nucleation stress when $\tau_{ij}^{FR} > \tau_{ij}^{TM}$, and it is determined by the intergranular slip transfer stress when $\tau_{ij}^{TM} > \tau_{ij}^{FR}$. In the former case, because the intragranular dislocation nucleation is harder to occur than the intergranular slip transfer, the dislocation pile-up phenomenon is not required to plastic deformation propagation by the slip transfer; hence the flow stress could show the $L^{-1}$ dependence. When the FR source length $L$ is related to the grain size $d$, the flow stress also shows the $d^{-1}$ dependence. In the latter case, because the intragranular dislocation nucleation
is easier to occur than the intergranular slip transfer, the dislocation pile-up phenomenon is required to generate stress concentration at the grain boundary. Based on the conventional pile-up model, the distance $D$ between the intragranular dislocation source and grain boundary where slip transfer will occur controls the magnitude of the stress concentration by the dislocation piled-up; hence the yield or flow stress probably show the $d^{-1}$ or $d^{-1/2}$ dependence when the distance $D$ is a function of $d$. Consequently, mechanical properties of UFG metals containing FR source in grains are strongly controlled by the competition between the intragranular dislocation nucleation and intergranular slip transfer phenomena. Further investigations of the competition between the intra- and intergranular plastic deformation phenomena by the atomistic simulation will give a clue to solve an interesting problem that why the strength of UFG metals shows the unique size dependence like $d^{-1}$ or $d^{-1/2}$.

Fig. 4 Competition between the intragranular dislocation nucleation and intergranular slip transfer under shear deformation of GBII-FR models. (a) Shear stress–strain curves. (b) Defect configuration during bow-out motion in case of $L = 5$ nm. (c) Shear stress distributions before ($\gamma_{fr} = 0.053$) and after ($\gamma_{fr} = 0.068$) bow-out motion in case of $L = 5$ nm.

Fig. 5 Competition between the intragranular dislocation nucleation and intergranular slip transfer under shear deformation of GBII-FR models. (a) Shear stress–strain curves. (b) Defect configuration during bow-out motion in case of $L = 5$ nm. (c) Shear stress distributions before ($\gamma_{fr} = 0.053$) and after ($\gamma_{fr} = 0.068$) bow-out motion in case of $L = 5$ nm.

Fig. 6 FR source length $L$ dependence of the initial bow-out stress $\tau_{fr}^{I}$ and $\tau_{fr}^{II}$ and the flow stress $\tau_{fr}^{II}$ of $\gamma_{fr} = 0.07$ in GBI-FR and GBII-FR models.
4. Conclusion

The influence of the competition between the intragranular dislocation motion and grain-boundary-mediated plastic deformation processes on the mechanical properties of UFG metals is investigated by huge-scale atomistic simulations of about 100 million atoms. We construct two-type of aluminum polycrystalline models containing the Frank–Read source with different grain boundary characteristics. The first plastic deformation occurs by the dislocation bow-out motion in the center grain for both models and the critical shear stress for plastic deformation occurs by the dislocation bow-out motion. The clear source length dependence that is almost the same as the simple theoretical model estimated by the dislocation theory; therefore, the existence of grain boundary does not influence the critical shear stress to dislocation bow-out motion in our simulations. The subsequent plastic deformation is strongly influenced by the resistance of the slip transfer by dislocation transmission through grain boundaries; namely grain boundary structures control the deformation mechanism. When \( r^{FR} > r^{TM} \), the material strength is dominated by the intragranular dislocation nucleation stress, on the other hand when \( r^{TM} > r^{FR} \), the material strength is determined by the intergranular slip transfer stress. Consequently, our simulations show that this competition between intragranular dislocation nucleation \( r^{FR} \) and the intergranular slip transfer \( r^{TM} \) is closely related to the unique size dependence of mechanical properties of UFG metals and also give the motivation to control grain boundary characteristic to obtain the excellent mechanical properties as reported the UFG copper containing many nano-twins.

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