Influence of grain size on the nucleation and development of plasticity in nanocrystalline FeNi films

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Abstract. A molecular dynamics study of the features of the nucleation and evolution of plastic deformation in nanosized nanocrystalline FeNi films under uniform uniaxial tension is carried out. The dependences of the strength properties of the films on the grain size are obtained. It is found that the dependences of stresses at which defects begin to nucleate, maximum strength is achieved and plastic flow is realized, on the grain size have a pronounced maximum. The optimal grain structure for which the nanocrystalline film has the maximum strength at uniaxial stretching is determined. The calculated dependences allow determining the critical grain size at which the Hall-Petch relationship is reversed. It is shown that the nucleation and initial development of plasticity in nanosized films with grain size above the critical one has a dislocation nature. The twinning contribution to the film plasticity increases at strains corresponding to plastic flow.

1. Introduction
The grain size is an important material characteristic that determines the activation or suppression of various mechanisms of plastic deformation. As a rule, as the grain size decreases to hundreds or tens of nanometers, the strength of material increases in comparison with coarse-grained counterparts. It is known that the onset of plasticity in metallic coarse-grained materials has a dislocation nature. In nanocrystalline metals with a grain size less than 20-30 nm, the dislocation mobility is significantly suppressed, since the Peierls barrier, which prevents dislocation slip, is sufficiently high [1]. Therefore, in nanocrystalline materials, along with dislocation processes, various grain boundary deformation mechanisms [1-3] or twinning [4-6] are activated. One of the main grain boundary processes is grain boundary sliding, which, in particular, prevails during superplastic deformation of nanocrystalline metals [7]. The important role of grain boundary sliding in the plasticity of nanomaterials at high stresses and strain rates is also confirmed by computer simulations [8,9]. Grain boundary sliding is realized in the form of plastic shears localized in the region of grain boundaries. They lead to the formation of structural defects at triple junctions, which become sources of internal stresses [10]. In turn, these defects can lead to the nucleation of nanocracks and subsequent brittle fracture of the material. Simultaneously with grain boundary sliding, accommodation processes can be initiated in the material, which are caused by the transformation of defects formed in triple junctions due to stress relaxation localized there. This leads to an increase in the ductility of the material. Revealing the nature of accommodation of grain boundary sliding in nanocrystalline materials is of great scientific and practical interest. The mechanisms of accommodation of grain boundary sliding in
such materials can be the emission of lattice dislocations from triple junctions [7], twinning [11,12], migration and splitting of grain boundaries [13,14], rotational deformation [15]. This behavior during deformation is typical for bulk nanocrystalline materials. However, in nanoscale nanocrystalline films, the mechanisms of nucleation and development of plasticity can differ, which is caused by a large fraction of the material located near free surfaces.

The leading role of particular accommodation process is associated with the grain size in the nanocrystalline material. The strength characteristics of nanocrystalline metallic materials increase with a decrease in the grain size only up to a certain density of grain boundaries, which is well described by the Hall-Petch relationship [16]. Starting from a certain critical grain size, further refinement of the grain structure leads to a softening of the material. For such materials, strength and grain size are related by the inverse Hall-Petch relationship. The physical nature of the inverse Hall-Petch relationship is still a subject of discussions [16-18].

The transience and small spatial scales of the processes that determine the behavior of materials at the atomic level impose difficulty on experimental investigations. At the same time, computer simulation allows one to study in detail the features of the nucleation and development of structural rearrangements under mechanical loading including high-energy action on the material [19-24].

In this work, the features of the nucleation and development of plasticity in nanosized nanocrystalline FeNi films under uniaxial uniform tension were investigated. We studied the effect of grain size on the mechanical characteristics of the samples to determine the optimal grain structure, at which the simulated films have the maximum strength under uniaxial tension.

2. Methods
The simulation was carried out on the base of the molecular dynamics method using the LAMMPS software package [25]. The interatomic interaction in the Fe-Ni system was described by the many-body potential developed in the framework of the embedded atom method [26]. The chemical composition of the samples corresponded to the equiatomic composition: Fe50%-Ni50%. The loaded samples consisted of 8 grains with approximately the same size. The centers of the grains were located at the sites of the conditional hcp lattice. The Voronoi tessellation was used to construct the grains. The grain sizes varied from 5 to 20 nm. For each grain size, 10 samples were loaded with different misorientation angles between the grains. The sample sizes, depending on the grain size, varied in the range from 10×8×8 nm to 40×32×32 nm. The corresponding number of atoms in the samples ranged from 63 000 to 4 080 000.

The simulated sample with a grain size of 20 nm is shown in figure 1. Periodic boundary conditions were used along the X and Y directions, and free surfaces were specified in the Z direction. The simulated samples were nanosized films, with two layers of grains parallel to the free surfaces. Uniaxial tension of the film was specified in the form of uniform stretching along the X direction at a constant rate of 5 m/s by means of a corresponding increase of the X coordinate for all atoms.

![Figure 1](image_url)

Figure 1. The initial structure of the nanocrystalline FeNi film with 20 nm grain size. Each grain is highlighted in a different color in figure (a). Only atoms on free surfaces and at grain boundaries are shown in figure (b).
During relaxation and loading of the samples, the thermostat rescaled a temperature to 300 K, and barostat maintained zero stress in the direction of the Y axis. The identification of structural defects in loaded samples was carried out using the Common Neighbor Analysis (CNA) algorithm [27]. The structure of the sample was visualized in the OVITO software package [28].

3. Results and discussion
Let us consider the most typical processes of nucleation and development of plasticity by the example of a nanocrystalline film with 20 nm grain size. Analysis of the simulation results showed that the features of the dependences of stress and the fraction of atoms with HCP symmetry of the nearest environment on the strain, shown in figure 2, correlate quite well with the features of the change in the defect structure of the loaded film (figure 3). Note that Shockley partial dislocations moving along slip planes leave intrinsic stacking faults (ISFs) behind. They are identified by the CNA algorithm as two layers of atoms with HCP symmetry of the nearest environment (hereinafter – HCP atoms) [29].

According to this algorithm, twin boundaries represent one layer of HCP atoms. Therefore, the fraction of HCP atoms in figure 2b is a rather informative characteristic that reflects the total area of nucleated defects.

\[ \text{Figure 2. Dependences of the stress along the } X \text{ axis (a) and the fraction of HCP atoms (b) on the strain of a nanocrystalline FeNi film with 20 nm grain size.}\]

The first maximum on the $\sigma(\varepsilon)$ curve in figure 2a will be called first yield stress. When the deformation corresponding to this maximum is reached, the first partial dislocation is generated and propagated in one of the grains of the film, leaving ISF behind which is highlighted in red in figure 3a. This leads to the gradual increase in the fraction of HCP atoms. The nucleated dislocation propagates through the grain body and stops at a neighboring interface, which can be either a grain boundary or a free surface. At further increase in strain to 3.30%, an absolute maximum (maximal stress) appears on the $\sigma(\varepsilon)$ curve. Starting from this strain, the nucleation and propagation of numerous structural defects takes place in the simulated film. This corresponds to an avalanche-like increase in the fraction of HCP atoms up to several percent (figure 2b). Such significant changes in the defect structure of the film lead to a remarkable decrease in stress (figure 2a). The defect structure corresponding to 4.25% strain is shown in figure 3b. After reaching this strain, partial dislocations had glided through almost all grains, leaving ISFs behind. Some ISFs, as a result of gliding of partial dislocations in a plane adjacent to them, are transformed into twins with a thickness of two atomic planes, which are highlighted in yellow in figure 3. When the simulated film is stretched in the interval of 4.50-6.25%, some ISFs “disappear” (figure 3c). This is due to trailing partial dislocations $1/6<112>{111}$ glide in the ISF plane, which lead to a shear corresponding to the full dislocation $1/2<110>{111}$, and restore an ideal FCC lattice. Approximately a half of all ISFs are transformed into thin twins simultaneously with the above process. When the sample is stretched in the interval of 6.25-8.00%, new ISFs are nucleated (figure 3d).
Figure 3. Defect structure of a nanocrystalline FeNi film with 20 nm grain size for different strain values: 3.10% (a); 4.25% (b); 6.25% (c); 8.00% (d); 10.50% (e); 15.00% (f). FCC atoms and atoms on the upper free surface are not shown. Atoms that belong to grain boundaries or free surfaces, intrinsic stacking faults and twin boundaries are colored in gray, red and yellow, respectively.
Then, up to 10.50% strain, the trailing dislocations glide along the ISFs and decrease the ISF area (figure 3e). As a result, some grains become almost defect-free. At the same time the area of twin boundaries remains virtually unchanged. Despite the fact that the fraction of HCP atoms for 10.50% strain is virtually the same as for 6.25%, in the interval between these strains a large number of split full dislocations glided in the grains, providing a significant decrease in stress. The stage of plastic flow begins at further deformation, at which the stresses and the fraction of HCP atoms change only slightly. This is due to the fact that twins begin to increase their thickness through the sliding of partial dislocations in adjacent planes at this stage of plastic deformation (figure 3f). Since the twin boundaries simply migrate to the adjacent plane in this case, the number of HCP atoms does not change. Almost all ISFs are transformed into twins. At the same time, grains without twins undergo multiple shears due to successive glide of Shockley leading and trailing dislocations.

As noted earlier, the grain size is the most important parameter of the internal structure that determines the physical and mechanical properties of the material. Therefore, it was of interest to reveal the effect of the grain size on the stresses at which the first yield stress occurs, the maximal stress is reached, and the plastic flow of the material is realized. In this case, the flow stress was calculated by averaging the stress in the interval of strains from 10 to 15%. The calculation results showed that the dependences of the first yield stress, maximal stress and flow stress on the grain size are significantly nonlinear and have a pronounced maximum. Note that the curves of dependences of first yield stress and maximal stress on average grain size in figure 4a correlate well with each other. FeNi nanocrystalline films with an average grain size of 12 nm are characterized by the highest strength with respect to the nucleation of structural defects. The critical grain size for the flow stress of nanocrystalline films is in the interval from 8 to 10 nm (figure 4b). This means that the flow stresses in nanocrystalline films with average grain sizes less than 8 and more than 10 nm are described by the inverse and classic Hall-Petch relationships, respectively.

**Figure 4.** Dependences of the average values of the maximum stress (black curve), first yield stress (red curve) (a) and flow stress (b), on the grain size in nanocrystalline FeNi films.

### 4. Conclusions
Based on the simulations performed, it can be concluded that the nucleation and development of plasticity in nanoscale films with a nanocrystalline structure under uniaxial tension has a dislocation nature. As the strain increases, twinning processes begin to play more important role in the deformation behavior of the films. It is shown that a decrease in the average grain size down to 8-10 nm increases the tensile strength of the films. This critical size defines the boundary at which the Hall-Petch relationship is reversed. At further decrease in the grain size, the strength of the films...
It is possible that this softening of the films is associated with the activation of intergranular processes, such as grain boundary sliding, grain boundary migration, grain rotations, etc.

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