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THE CAUSE OF HIGH-ANGLE BOUNDARIES FORMATION AT THE SEVERE PLASTIC DEFORMATION OF CRYSTALS: DEFORMATION FRAGMENTATION OR DYNAMIC RECRYSTALLIZATION

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Abstract. By transmission electron microscopy and back-scattered electron diffraction the structural parameters of deformation fragments and dynamically recrystallized grains in α-Fe and FeNi alloy have been analyzed upon severe plastic deformation in a Bridgman camera at a room temperature. It was detected that the initiation of high-angle grain boundaries in the deformed structure is associated with low-temperature dynamic recrystallization.

1. INTRODUCTION

Severe plastic deformations (SPD) is undoubtedly associated with extreme conditions [1]. This approach to material treatment has been of growing interest in recent years, since it allows considerable improvement of the physical and mechanical properties of metallic materials [2]. This is largely due to the creation of different fine-grained states, particularly ones that involve fragmentation processes. Four approaches to producing very high values of deformation are the ones most common today [3]: high pressure torsion (HPT) in a Bridgman camera, equal channel angular extrusion, accumulated rolling, and screw extrusion. In the first approach, a sample is placed between two anvils, one of which slowly turns with the simultaneous produc-tion of extremely high quasi-hydrostatic pressures (several GPa). In the second, a sample is extruded through two channels of equal diameter and inclined to each other at a certain angle (up to 90°). Plastic deformations in this case are so great that ordinary values of the relative degrees of deformation have no meaning, and we must switch to true logarithmic strains *e*. For HPT, they are defined as [4]:

$$e = \ln\left[1+\left(\varphi r / h\right)^2\right]^{0.5} + \ln\left(h_0 / h\right), \qquad (1)$$

where *r* and *h* are the radius and height of a sample disk treated in a Bridgman camera, and φ is the turning angle of the movable anvil. Number *N* of full turns of the movable anvil corresponds to the deformation at which $\varphi = 2nN$.

Structural states created at such severe deformations are very complicated and difficult ot predict. Unfortunately, most authors restrict the use of HPT to only mechanical treatment and studying the resulting structures and corresponding properties of materials, without analyzing the physical processes that occur immediately at extremely high degrees of plastic flow. In our opinion, classical dislocation and disclination approaches to the study of structural processes upon severe plastic deformation are insufficiently effective and require profound rethinking.

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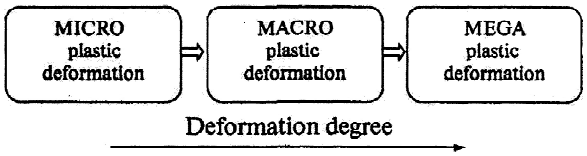

Fig. 1. Stage principle of the plastic deformation of solids.

Summarizing the many experimental studies of the structure of SPD-treated pure metals,, solid solutions and intermetallic compounds we can postulate the following feature of SPD process [5]:

- Fragmentation
- Existence of high-angle grain boundaries
- Lack of work hardening
- Low-temperature dynamical recrystallization
- High diffusibility
- Initiation or solving of non-equilibrium phases
- Amorphization.

The most refined concept of structures forming upon severe plastic deformation was proposed by V.V Rybin [6]. He succeeded in correctly describing phenomena that occur at significant degrees of defor-mation close to $e \sim 1$ based on ideas about the dominant role of disclination mode and related fragmentation processes. Analysis of numerous experimental data, especially for BCC crystals, led Rybin to the concept of a limit (critical) fragmented structure whose further evolution is impossible in the disclination mode [6]. Breakdown sites form on the boundaries of fragments that separate regions normally free of dislocations. The critical fragmented structure is an end product of plastic deformation that yields to the growing effect of external and internal stresses and should result in decay. This is true only for the early stages of HPT ($e < 2$) and thus does not describe structural changes that occur during later stages of deformation.

It was noted in [7] that changes to HPT in technically pure iron were accompanied by jump-type changes in the material's structure that occurred at critical value $e_c \sim 1$. The mechanical behavior of the material also changed: the mechanical hardening obeyed a linear instead of a parabolic law upon severe deformation. A copper wire was HPT-treated in [8] ($e = 1.6$ and 3.7); cyclic variations in the structure were observed as deformation grew: fragmented structure ($d = 0.2$ μm) ⇒ recrystallized structure ⇒ fragmented structure ($d = 0.1$ μm, where d is was the mean size of fragments.

Energy aspects of a solid behavior under a large load were considered in [9]. It was suggested that SPD can be observed not only at special deformation procedures as was commonly supposed before [10], but at the any one deformation procedure (e.g., rolling) following after microplastic and macroplastic stages with extremely high value of deformation (Fig. 1). If a finite-dimension solid can be presented as a mechanical dissipative system [10], we suggest that some mechanical energy is introduced into the solid under deformation action. A plastic flow is an obvious dissipation (relaxation) channel for this energy. When the energy's dissipative capabilities are exhausted, another dissipation channel can be used, e.g., mechanical fracture. However, the high amount of accumulated elastic strain energy can result in additional channels of dissipation: low-temperature dynamic recrystallization, phase transformations, and the release of thermal energy. With SPD, the formation and growth of cracks is partially or completely suppressed when the component of hydrostatic compression stress is high, and the progress of the fracture is considerably hindered. In the other words, using HPT or similar loading procedures, we force the solid to deform without fracture.

If the limit structure concept is correct [6], then dislocation-disclination accommodation is effective up to a certain limit that corresponds to the formation of the critical defect structure, and other factors should act as additional channels of mechanical energy dissipation. Three possible structural scenarios of SPD development were suggested in [11] on the basis of the energy approach. In materials where dislocation (disclination) transformations made much easier (e.g., in pure metals), low-temperature dynamic recrystallization occurs after fragmentation. Local regions of the structure become free of defects and local stresses, and a plastic flow begins again in new recrystallized grains as a result of dislocation and disclination modes. Dynamic recrystallization then acts like a high-power additional channel of elastic strain energy dissipation. If the mobility of plastic deformation carriers is relatively low (e.g., in solid solutions or in intermetallic compounds), phase transitions serve as a high-power channel of dissipation, most often the crystal-to-amorphous state transition. As a result, the plastic flow localizes in an amorphous matrix without deformation hardening or the accumulation of high internal stresses. The choice of one scenario of structural transformation or another depends, among other things, on parameter T_{SPD}/T_m, where T_{SPD} is the SPD temperature with allowance for the probable effect of heat release and T_m is the material melting temperature.

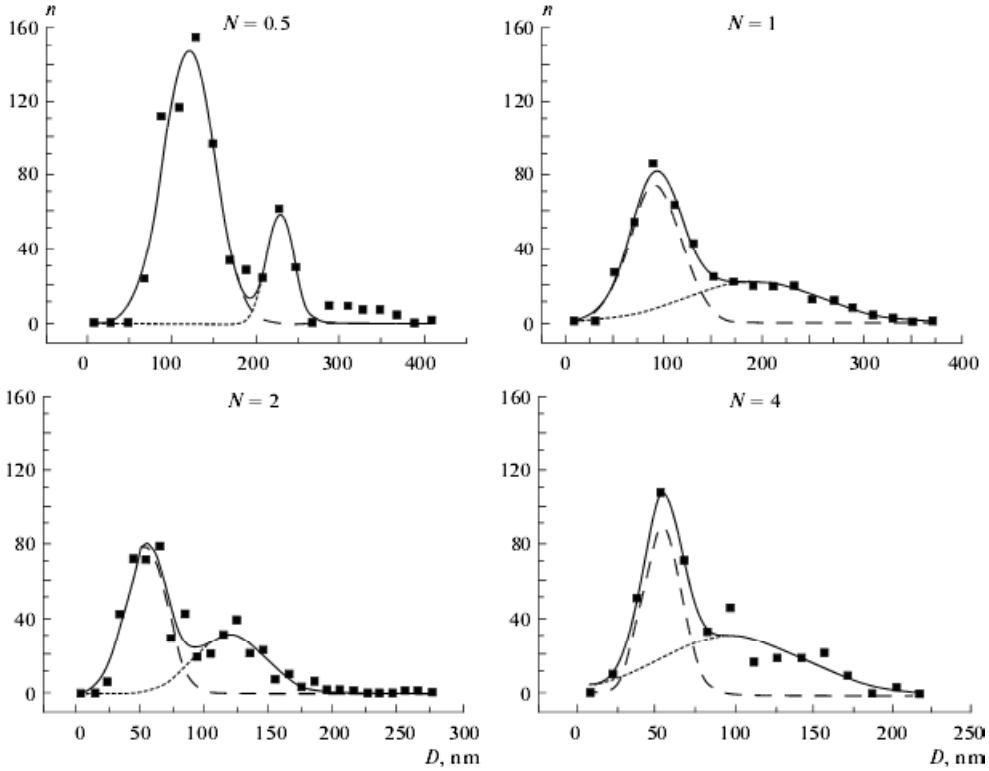

Fig. 2. Distribution histograms of grain and fragment sizes and their division into two Gaussian distributions after HPT with different values of full turns *N* for FeNi alloy: general histogram (solid), deformation fragments (dashed) and recrystallized grains (dotted).

Assuming the first scenario of structural transformations during SPD, we may *a priori* state that recrystallization during SPD is possible even at room temperature. Recrystallization processes (including dynamic ones) are purely diffusional [12]; we should therefore say that the diffusion and self-diffusion processes of substitution atoms required for the formation of recrystallization nuclei and their further growth can occur in iron, nickel, and other metals, along with their alloys at temperatures much lower than 0.3 T_m, where SPD experiments are usually carried out. This statement might appear incorrect at first glance; however, there is a great deal of experimental evidence that dynamic recrystallization during SPD actually occurs at room and even lower temperatures than those given in the literature [13,14]. Dynamic recrystallization can occur both with the growth of new recrystallized grains (discontinuous dynamic recrystallization) and without the formation of nuclei of new grains (continuous dynamic recrystallization) [14].

One feature of the structure of a metal material after SPD is existence of large local misorientations that correspond to the high-angle boundaries of grains [15]. It is believed that these boundaries largely determine unique properties of a material after SPD. But what is the physical nature of high-angle boundaries formation upon severe deformation? Unfortunately, this question has no correct answer within the current plastic deformation model, which uses a structural kinetics approach and the concept of mesodefects induced during deformation at relatively low true values of deformation (e ~ 1-2) [6,15]. This question remained essentially answered in [8], a detailed survey of this problem published relatively recently. The aim of this work is therefore to suggest a realistic mechanism of high-angle boundary formation in pure metals and diluted solid solutions during SPD.

2. EXPERIMENTAL

A Bridgman camera was used to produce HPT [4]. Polycrystalline α-Fe and single-phase polycrystalline FeNi alloy with FCC crystal structure were used as the material for our study. The mean size of grains was ~50 after slow cooling from 850 °C, and there was virtually no crystallographic texture. Thin (0.2 mm) plates were subjected to torsion deformation in the Bridgman camera at room temperature, constant quasi-hydrostatic pressure (P = 4 GPa), at 1 r/m rate of the movable anvil, and number N = 1/4, 1/2, 1, 2, 3, and 4 of continuous full turns of the movable anvil.

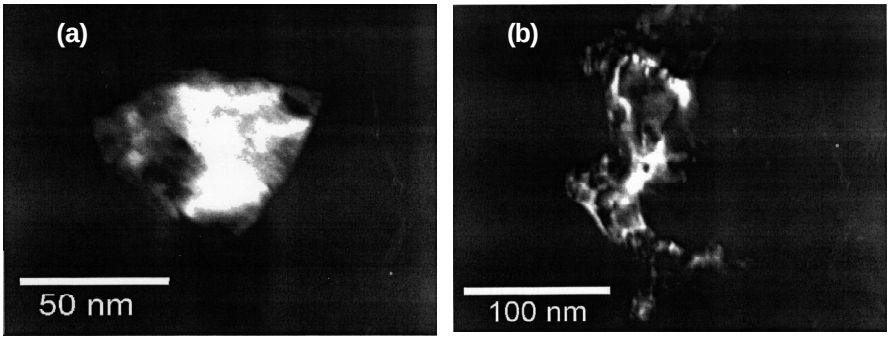

Fig. 3. Electron-microscopic images of the structure for the FeNi alloy after HPT (N = 2); (200) dark field images of recrystallized grain (a), and dislocation fragment (b).

The structure was studied after THP using a JEM-200CX transmission electron microscope with an accelerating voltage of 160 kV in the dark field mode. A JSM-7500F scanning electron microscope with a resolution of 0.05 μm and an attachment for EBSD analysis were also used. All studies were conducted in regions corresponding to approximately one-half the radius of the sample disks.

3. RESULTS AND DISCUSSION

TEM histograms of the distribution of grain sizes and fragments after deformation were obtained for different N (Fig. 2). Parameter $D = 4S$ was selected as the characteristic size of a structural element, where S is the image area of the grain or fragment. Analysis of all the distributions revealed two maxima (a bimodal distribution) whose parameters changed as functions of the degree of deformation, while the bimodal character of the distributions was maintained.

We showed in [9] and in a number of other experiments [14] that within the proposed concept of the existence of additional channels for elastic strain energy dissipation upon HPT, the SPD processes in metals and solid solutions at room temperature are accompanied by dynamic recrystallization. Electron microscope analysis of the structure of our pure metal and alloy sample after experiencing different modes of deformation in the Bridgman camera also showed clear signs of similar processes (Fig. 3). Dark-field electron microscope images of the structure clearly showed individual grains that were almost regular hexahedrons and a low concentration of defects (Fig. 3a), testifying to the formation of such grains during dynamic recrystallization via the migration of high- angle boundaries formed according to one of the possible relaxation mechanisms of dislocation structure transformation under the conditions of a high concentration of point defects and high gradients of inner stresses [15]. At the same time, some grains (fragments) are of irregular shape and include high concentrations of defects and noticeable local distortions (Fig. 3b), indicating their formation through deformation fragmentation [6]. Each of the two grain types revealed in the structure after HPT treatment is clearly characterized by its own size distribution. In other words, the histograms in Fig. 2 are in fact combinations of two histograms of distributions of grains and fragments that differ in origin.

Fig. 2 also shows examples of the division of the experimentally obtained bimodal histograms for different N into two Gaussian distributions. Analysis of electron microscope images shows that the distributions shown by dashed curves correspond to fragments of deformation origin, while those shown by dotted curves correspond to recrystallized grains. The fractions of the volume occupied by deformation fragments C_f and recrystallized grains C_{rg} were then calculated for each mode of deformation by calculating the relative areas under the Gaussian distributions (Fig. 4a) and the average size of regions corresponding to fragments D_f and recrystallized grains D_{rg} (Fig. 4b). It can be seen that at first, C_f sharply increases with N; it then gradually decreases until it reaches a constant value of 0.6. Parameter C_{rg} obviously includes not only recrystallized grains but the initial grains as well; it first sharply falls to 0.2 and then rises to 0.4, where it remains almost constant. Parameter $k = C_{rg}/C_f$ at $N > 1$ is in our opinion an important structural constant and depends on the nature of the material and the HPT parameters, which largely determine its physical and mechanical properties (particularly the ratio between strength and plasticity). To confirm this, we must emphasize that the measured microhardness grew sharply when $N < 1$ and remained constant when $N > 1$. Dynamic equilibrium

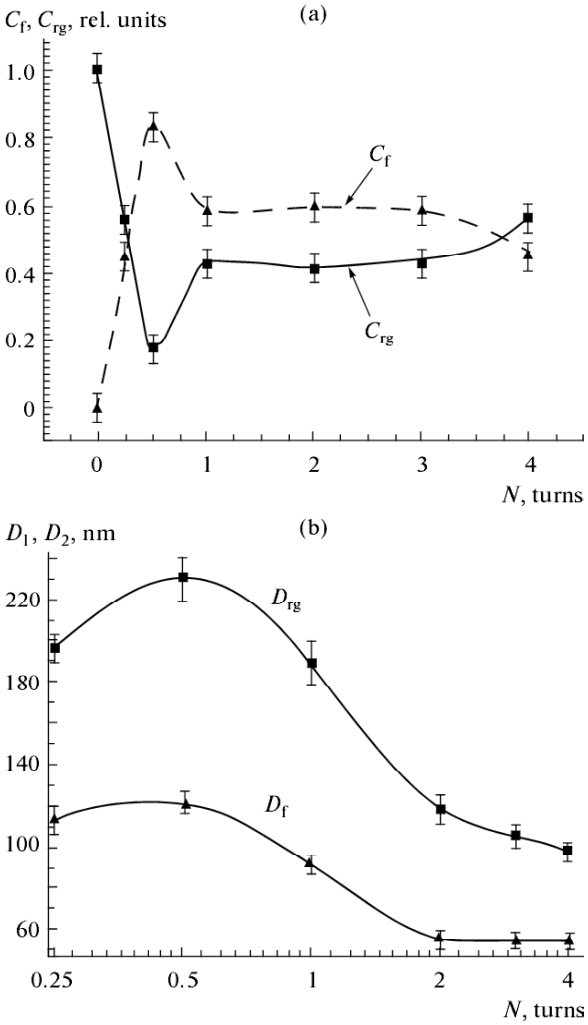

Fig. 4. Variations in (a) volume fractions C_{rg} and C_f and (b) mean sizes D_{rg} and D_f for recrystallized grains and deformation fragments, respectively, as functions of the number of full turns N in FeNi alloy.

between structural transformations F ⇔ RG is then observed during HPT. Dependences *D(N)* are simbate in Fig. 4; i.e., D_f and D_{rg} gradually fall to 50 and 100 nm, respectively, and then remain virtually constant when $N > 2$.

EBSD analysis allowed us to determine the characters of the distributions of grain-boundary misorientation angles φ for fragments and for grains formed at different N (Fig. 5). The fraction of grains with φ > 20° grew notably with N, while the grain-boundary angle distribution was invariable for all N: the misorientation maximum corresponded to φ = 40° - 50°. This contradicts the severe plastic deformation model [6], in which the φ maximum grows monotonally with N.

Let us now consider what we believe to be the most important result that follows from the above. We shall try to find a correlation between the fraction of dynamically recrystallized grains C_{rg} and the fraction of high-angle grain boundaries *a* under the same structural conditions of the alloy at different stages of HPT (with different N values). The values of α were calculated using the data in Fig. 5, with limitations on minimum φ (α_{20} if φ > 20°; α_{30} if φ > 30°; and so on). Parametric dependence $\alpha(C_{rg})$ were plotted for N = 1/2 - 4 and $\alpha = \alpha_{20} - \alpha_{50}$. Fig. 6 shows the experimental results $\alpha_{40}(C_{rg})$, where each point corresponds to a certain value of N. Linear correlation coefficient r was calculated using the standard technique and was in our case 0.912. In other words, a strict linear correlation was observed

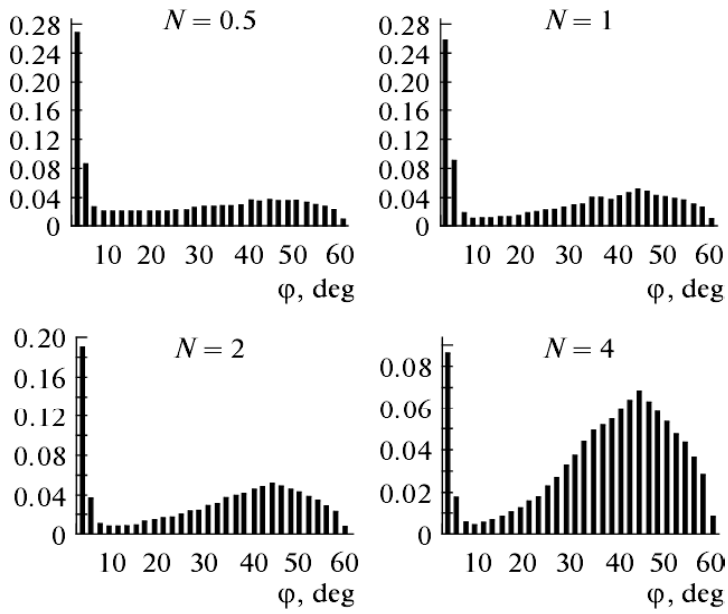

Fig. 5. Histograms of grain-boundary angle distribution φ in FeNi alloy for different numbers of full turns N (EBSD analysis).

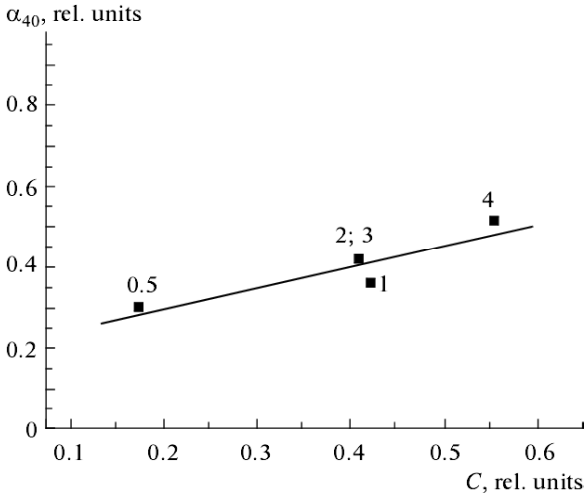

Fig. 6. Linear correlation of parametric dependence α_{40} (C_{rg}) for FeNi alloy; C_{rg} - the volume fraction of recrystallized grains and α_{40} - the fraction of grain boundaries with misorientation angle $\varphi \geq 40°$. The figures show the number of full turns *N*; correlation coefficient *r* = 0.912.

between the increase in the fraction of grains formed during dynamic recrystallization and that of the high-angle grain boundaries in the structure ($\varphi > 40°$). In our opinion, this clearly shows it is dynamic recrystallization, and not deformation fragmentation, that is responsible for many of the high-angle grain boundaries in the structure of the HPT-treated material. Let us note two important points:

(1) The maximum of high-angle grain boundary distribution is reached at angles φ = 40-50°, which correspond to the highest mobility of high-angle boundaries, and thus to the most rapid growth of recrystallization nuclei [14].

(2) The linear correlation coefficient *k* for parametric dependences $\alpha(C_{rg})$ was decreased when decreased the minimal φ value($\alpha_{50} \Rightarrow \alpha_{20}$), testifying to the growing contribution from dynamic fragmentation to the formation of middle- and low-angle grain boundaries.

We may assume that the structural sensitivity and magnetic properties of single-phase HPT-treated materials are determined by the ratio between the volume fractions of deformation fragments and recrystallized grains $k = C_f / C_{rg}$. The higher *k*, the greater the strength and magnetic hardness of a ferromagnetic. In our investigation for pure α-Fe and for FeNi alloy, *k* was close to unity. We may also assume that an increase in the volume fraction of recrystallized grains (a lower *k*) would correspond to an increase in the plasticity of the HPT- treated material; however, this hypothesis must be verified experimentally.

4. CONCLUSIONS

(1) The formation of structural regions corresponding to deformation fragments and dynamically recrystallized grains in the structure of α-Fe (BCC) and FeNi alloy (FCC) upon severe deformation in a Bridgman camera at room temperature was experimentally ascertained using the energy concept of additional channels for elastic strain energy dissipation (relaxation) during HPT.

(2) We showed via TEM and EBSD that the high concentration of high-angle grain boundaries in the structure of HPT-treated material is due mainly to dynamic recrystallization process but not only deformation fragmentation process, as was believed earlier.

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