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Hardening strain and recovery strain in nanocrystalline Ni investigated in tests with multiple stress changes

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ABSTRACT

Purpose: Dynamic recovery is interesting as it limits the maximal deformation strength of crystalline materials. Due to its small grain size, nanocrystalline Ni reaches its maximal strength after small strains < 0.1. It is shown that dynamic recovery contributes to strain and that its kinetics differs from that of hardening strain.

Design/methodology/approach: The kinetics of recovery was studied by performing a large stress reduction suppressing thermally activated glide of the hardening type. The transition to a new quasi-stationary state at reduced strain rate and stress was accelerated by incremental increases of stress.

Findings: During the transition the kinetics of deformation changes from that of recovery strain to the quasi-stationary one where hardening and recovery are coupled. The results are interpreted in terms of thermally activated hardening strain (in the grains) and thermally activated recovery strain (boundary mediated) linked by internal stresses. The activation volume of the hardening strain rate determined from the small stress increments is not inconsistent with the classical theory of thermally activated dislocation glide.

Research limitations/implications: It is proposed to better characterize dynamic recovery by performing small stress changes in the period of dominating recovery strain to quantify the kinetics parameters of recovery strain.

Practical implications: Disturbing deformation by sudden changes of stress is recommended as a suitable means to describe the kinetics of dynamic recovery. Recovery strains should enter the modeling of plastic deformation. This holds in particular for cases where dynamic recovery is prominent, e.g. at high stresses, high temperatures, and variable stresses (cyclic deformation, stress relaxation).

Originality/value: The stress change method described in this work is generally applicable in deformation testing independent of the type of testing machine, where inelastic strains are measured at the usual accuracy.

Keywords: Hardening; Recovery; Boundaries; Nanocrystalline; Stress changes

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MATERIALS

1. Introduction

Plastic deformation of crystalline materials is connected with storage of crystal defects (dislocations). Increasing dislocation content leads to strain hardening. The fact, that recovery of defects also is directly associated with inelastic strain, is less well known. The recovery strain, i.e., the inelastic strain being directly connected with dynamic recovery of defects, is usually not considered. However, it may rise to about 30% of the strain in monotonic deformation (see e.g. [1]) and even dominate when the stress is suddenly reduced. So it seems necessary to take the recovery strain into account to correctly describe inelastic deformation under service conditions on a microstructural basis. In the following the experimental evidence for recovery strain is highlighted.

Recovery strain was found in deformation at elevated temperatures under creep conditions [2,3] and confirmed in tests with full unloading [4]. To detect it, one must create conditions where dynamic recovery is dominating. This is possible by sudden stress reductions [5]. A convenient starting point is the state of quasi-stationary deformation. Here generation and recovery of dislocations are balancing each other so that the deformation strength at constant temperature and absence of fracture remains fairly constant except for slow changes caused by slow changes of the structure of the crystallites (subgrains) regarding spacing and boundary structure. So the intensity of recovery has reached its maximum value that can be reached in monotonic deformation. Inelastic strain of the "normal", work hardening kind, i.e., the strain leading to storage of dislocations, is driven by the applied stress σ . However, the dislocation obstacles exert back stresses on various spatial scales that reduce the thermal ("effective") stress component available for dislocation glide. Usually the effective stress is only a small fraction of the applied stress. This means that it is brought to zero even by moderate stress reductions. In this situation storage strain ceases for a while. Recovery seems to be necessary before deformation can continue. In the past, this simplistic expectation has led to the proposal that there would be an incubation period after a fast stress reduction where nothing happens except recovery. Many results have been obtained on the length of this recovery period with strain rate zero [6-8]. However, on a closer look it turned out that in general the strain rate is not exactly equal to zero. Rather straining goes on, first at negative rate (anelastic back flow, considered to be a reversible process), later at positive rates. The stress reduction tests in [2] were among the first to show that. At that time the observation of fast strain after a large stress reduction was termed "anomalous". But in fact it is a normal feature of deformation after large stress reduction. It is found virtually everywhere because recovery is always an important part of the microstructural processes in deformation.

Many works determine the activation volume *V* of deformation from change tests [9-13]. Usually the applied total (elastic plus plastic) strain rate $\dot{\epsilon}_{tot}$ is changed. The associated change of flow stress is measured and used to calculate *V* as $Mk_BT(dln \dot{\epsilon}_{tot}/d\sigma)$, where σ is the stress, *M* is the Taylor factor, k_B is the Boltzmann constant, *T* is the temperature and $\dot{\epsilon}$ is the strain rate. It is claimed that the rate change occurs at constant structure. The difference between applied total strain rate $\dot{\epsilon}_{tot}$ and inelastic strain rate $\dot{\epsilon}_{inel}$ is neglected. Often the change tests are performed under conditions where the flow stress has saturated implying that the density of free dislocations is in approximate dynamic equilibrium of generation and recovery. The parameter *V* is then interpreted in terms of thermally activated glide.

This is incorrect. A quasi-stationary (qs) state depends on applied stress σ . Changing σ means changing the microstructure. So the assumption of "constant structure" is violated and V does not have the anticipated meaning. To determine V care must be taken to measure the very first material response to the change where the transient to the new qs state is just beginning so that the change of the microstructure may still be negligible. This requires to determine the inelastic strain rate $\dot{\varepsilon}_{inel}$. It results from the difference of $\dot{\varepsilon}_{tot}$ and elastic strain strain rate $\dot{\varepsilon}_{el} = \dot{\sigma}/E_{eff}$ where $E_{\rm eff}$ is the effective elastic modulus covering the elastic strain contributions from the specimen and the deformation machine. But conversion of $\dot{\varepsilon}_{tot}$ into $\dot{\varepsilon}_{inel}$ is rarely done. Tests with sudden changes of applied stress σ from one level to another constant level are a better alternative to rate change tests because the elastic strain rate $\dot{\varepsilon}_{el}$ is vanishing in the periods of constant σ so that $\dot{\varepsilon}_{tot}$ equals $\dot{\varepsilon}_{inel}$.

We conclude that valid determinations of the activation volume of thermally activated glide are rare. This holds in particular for nanostructured materials. Due to the small grain size, strain hardening is so rapid that a qs state with saturated flow stress at constant strain rate is quickly reached. In this work we discuss change tests on nc Ni where the existence of recovery strain is proven and the activation volume for thermally activated glide is measured.

2. Results

Tests with multiple stress changes have been described in [1]. Figure 1 shows the time history of the test on nanocrystalline Ni with 35 nm grain size. It consists of an initial phase where the qs flow stress is established at high rate so that there is room for large reductions of stress without immediate stop of deformation. Then the stress was reduced within 1 s to $R_{\sigma} = 0.4$ of the initial qs flow stress $\sigma_0 = 2$ GPa. After the σ -reduction the progress of inelastic strain rate ($\dot{\varepsilon}_{el}$ is zero) was followed (see below). When $\dot{\varepsilon}_{inel}$ had become too low, σ was increased again by a small amount. This procedure was repeated until the new qs state at the reduced level of stresses and strain rates was reached. The test ended by fast deformation to fracture.



Fig. 1. Stress history of stress change test consisting of tensile deformation at constant $\dot{\epsilon}_{tot} = 10^{-3}$ /s to 2 GPa, fast unloading to 0.8 GPa, incremental stress increases, deformation at constant $\dot{\epsilon}_{tot} = 10^{-3}$ /s to ultimate tensile stress and beyond

Figure 2 (a) shows the deformation path after the large σ -reduction in the field of $\dot{\varepsilon}_{inel}$ versus stress, both normalized to the starting values σ_0 and $\dot{\varepsilon}_{inel,0}$. The test has been done *in situ* under synchrotron radiation allowing for the measurement of the peak broadening (in terms of the

full-width at half-maximum – FWHM) from different diffraction peaks. Such peak broadening can be influenced by the variations of strain in the material. As the applied stress increases, the magnitudes of the internal stresses increase and the peak width broaden.

Figure 2 (b) demonstrates the overall changes of the peak broadening Δ FWHM (changes within each stress step (Fig. 6 in [1]) are neglected for simplicity). Here the results of {311} and {200} diffraction peaks are displayed. Just after the large σ -reduction FWHM declines sharply in parallel to back flow. The decrease of peak width continues when $\dot{\epsilon}_{inel}$ has become positive again (Fig. 2 (a)). Only at stresses of 1.30 to 1.68 GPa FWHM starts to increase again till the end of the incremental stress increases.



Fig. 2. a) Normalized inelastic strain rate $\dot{\epsilon}_{inel,0}$, b) the overall changes of the peak broadening Δ FWHM versus normalized stress $R_{\sigma} = \sigma/\sigma_0$ in test with σ -reduction from 2 GPa to 0.8 GPa followed by incremental stress increase; qs: quasi-stationary strength

Figure 3 gives examples of the variation of $\dot{\varepsilon}_{inel}$ with time in selected parts of the test. In Figure 3 (a), deformation begins with back flow in the first stress step after the large σ -reduction. The rate of black flow declines rapidly and $\dot{\epsilon}_{inel}$ becomes zero after about 220 s. Then flow continues in positive direction again. The strain rate $\dot{\varepsilon}_{inel} > 0$ increases towards a maximum. Afterwards the deformation slows down again to approach the qs strain rate at $\sigma = 0.8$ GPa. This rate can be estimated from extrapolation to be less than 10⁻¹⁰/s [1]. The incremental increase of stress counteracts the decrease of rate, but does not stop it. This is demonstrated in more detail by the time history of inelastic strain in the first five stress levels after the big stress reduction shown in Figure 3a. In spite of the increase of stress from step to step, the average strain rate continues to decline. Without the stress increments the decline would be much stronger.

Figure 3 (b) shows ε_{inel} as function of time *t* in stress steps 21 to 24. Here the strain rate $\dot{\varepsilon}_{inel}$ increases from step to step and each stress increment induces so-called normal transients where the stress rate $\dot{\varepsilon}_{inel}$ is enhanced compared to the final ones. In the last step $\dot{\varepsilon}_{inel}$ remains relatively constant after the secondary transient indicating that a new qs state is approached.

3. Discussion

Our change tests show that the mechanism of deformation short after the large sudden stress reduction differs qualitatively from that in the final stress steps near the new qs state. In the first phase dynamic recovery is dominating as seen from the decrease of diffraction peak width. In this phase the strain rate decreases strongly with increasing (time and) strain even though the stress is stepwise increasing. Secondary transients cannot be detected. This may simply be due to the fact that the increase of strain per step is too small (in the order of 10^{-4}). However, there is an indication of slight decreases of $\dot{\varepsilon}_{inel}$ within each stress step.

In the last stress steps (Fig. 3 (b)) the material is much closer to the new qs state. Each stress increment causes a normal transient and the decrease of $\dot{\varepsilon}_{inel}$ becomes smaller despite the much larger strain interval per step compared to the first steps (Fig. 3 (a)).

In the following we interpret the two mechanisms as recovery strain (related with boundary mediated mechanisms of flow) and work hardening strain (by thermally activated dislocation glide related with storage of defects).



Fig. 3. a) Stress and inelastic strain during incremental stress steps 1 to 5 after the stress reduction as function of time, b) stress and inelastic strain during incremental stress steps 21 to 24 as function of time

3.1. Recovery strain

In the qs state at σ_0 the density of dislocations stored at boundaries and the related internal stresses are maximal. The large relative reduction of σ to $R_{\sigma} = 0.4$ stops dislocation glide in forward direction and causes back flow because the effective stress (applied stress diminished by athermal stress components including long-range back stress) becomes negative. This causes reduction of the amplitude of the internal stresses because the ε_{inel} difference between soft and hard regions is diminished. At the same time the dislocation density recovers because dislocation loops shrink (compare [14]; note that each dislocation by definition is part of a closed loop surrounding a slipped area).

More recovery occurs by continuing forward deformation in the hard regions where local stresses are still positive. The forward deformation here is carried by dislocations and by crystallite boundaries (including highangle ones) responding to stress by migration coupled with lattice shear in the course of atomic jumps across the boundaries. The forward flow in the hard regions also diminishes the internal stress amplitude for the same reason as the back flow. The effect of the recovery of dislocations and internal stresses resulting from the polarization of the dislocation structure is seen in the FWHM decrease. This decrease extends over a large part of the stress interval up to about $R_{\sigma} = 0.5$ corresponding to 1 GPa. In all of this range the strength is distinctly lower than it was in the initial state (σ_0 , 10⁻³/s) and also lower than in the new qs state that would be attained at the given stresses in the stress increment period. This is obvious from the fact that measured $\dot{\epsilon}_{inel}$ are always much higher than the qs rates would be (according to extrapolation of the dotted qs line in Figure 2. This means that the recovery of the dislocation density is associated with forward strains resulting from dislocations and boundaries. This strain was termed recovery strain. Extrapolation of the recovery strain rate to σ_0 shows that the fraction of recovery strain at qs deformation is in the order of 20%. After large reduction of stress the recovery strain makes the dominating contribution. As the recovery slows down, the recovery strain rate also slows down. This looks like strengthening, but is in fact only deceleration of recovery. When recovery has sufficiently proceeded, forward deformation of the normal type sets in again and accompanies the strain hardening to the new qs state.

Short after the first large stress reduction to $R_{\sigma} = 0.4$ the recovery strain rate appears to play the decisive role. So the first little stress increments should give information on the kinetics of the recovery strain. These transients are

weakly pronounced and different from the later ones. It seems that the change of applied stress is insignificant compare to the local stress inherited from the qs deformation at $\sigma_0 = 2$ GPa.

3.2. Activation volume

The response to stress increments can be used to determine the very first inelastic strain rate measured in an $\varepsilon_{\text{inel}}$ -interval of a few 10⁻⁵ after a sudden incremental change of σ . This is an optimal approximation of a change test at constant structure. As mentioned before, V is immeasurably small short after the large stress reduction where recovery is dominating. After later stress increments the response is turning over to that of thermally activated glide of the storage type. So it presumably describes the relatively fast dislocation flow through the soft regions. The values of $V_{\text{glide}} = Mk_{\text{B}}T(\text{dln}\,\dot{\epsilon}/\,\text{d}\sigma)$ are shown in Figure 4. The term dln $\dot{\varepsilon}$ was determined from the rates measured before and after each stress increment. The scatter is large due to the experimental scatter in ε_{inel} and the necessity to keep the strain interval used for determination of $\dot{\varepsilon}_{inel}$ as small as possible. Two independent evaluations were performed where $\dot{\varepsilon}_{inel}$ was determined by fitting a straight line to the $\varepsilon_{\text{inel}}(t)$ curves of the type shown in Figure 3. Gray and black dots differ in the strain interval $< 10^{-4}$ used for fitting after the stress increments being smaller for the gray dots. The average values for V_{glide} in the last steps shown in Fig. 3b are nearly the same for both cases and give $V_{glide} = 22 b^3$.

These experimental values may be compared with the expectation of the classical theory of thermally activated glide of dislocations. It says that V is given by the width Δx and the spacing l_{th} of the thermal obstacles. In pure coarsegrained materials Δx is in the order of one atomic spacing, i.e. the magnitude b of the Burgers-vector and l_{th} is in the order of the dislocation spacing. A good estimate of the latter is bG/σ where G is the shear modulus. This yields $V = b^3 G / \sigma$. In the present case of nc Ni the stress is close to the mechanical threshold where the obstacle width has already become relatively small due to the large amount of mechanical activation of overcoming the obstacle. So Δx may be expected to be smaller than b so that $V = x_x b^3 G / \sigma$ with $0 < x_x < 1$. Here x_x is the numerical factor of $\Delta x/b$. It is seen from Fig. 4 that V/b^3 is approaching the line expected for coarse grained pure material with $x_x = 0.5$ at the end of the period of reduced stress close to the new qs state. So we get a value for the activation volume in the qs state at 1600 MPa of nc Ni that is not inconsistent with classical theory. This may indicate that the type of thermally activated flow carrying the deformation in nc Ni is virtually same as in coarse-grained material.

It has been proposed that the obstacle spacing is determined by the grains size *d*. However, this would give $V = 70 b^3$ for the same obstacle width 0.5 *b*. This value is distinctly higher than the measured one and therefore appears less probable.



Fig. 4. Normalized activation volume for thermally activated glide from stress increments versus normalized stress (G = 78.9 GPa: shear modulus of Ni); dots: individual data from each evaluated step (see text); open circles: average from steps 21 to 24 (Fig. 3 (b))

4. Conclusions

Multistep stress change tests confirm that strain is not only associated with storage, but also with recovery of defects.

After large relative stress reduction the rate of recovery strain is first negative. This type of recovery during back flow is simply the reversal of defect storage during forward flow.

As back flow ceases, the positive recovery strain becomes the dominating strain contribution. Its rate passes a relative maximum on the way toward the new quasistationary state at reduced stress where the recovery strain rate is lower by many orders of magnitude. The positive recovery strain, presumably located at the boundaries where dislocations had been stored, lets the material for a while appear soft in spite of the high defect content inherited from pre-deformation. When bulk deformation in forward direction sets in again, the strain associated with storage of defects begins to dominate again.

The transient responses of positive recovery strain and storage strain to incremental stress increases are different. The storage strain by dislocation glide exhibits a normal transient behavior with transient strain hardening. The activation volume of thermally activated glide is in the same order of b^3G/σ usually observed in pure materials. To determine reliable values of the activation volume of positive recovery strain, stress increments are not enough because the rate of recovery strain is declining too fast. To eliminate the effect of this decline, it is necessary to perform small stress cycles in the range of dominating positive recovery strain as done in [15,16].

For better understanding of crystal plasticity in application of materials under variable deformation conditions (see e.g. [14]) it seems necessary to explicitly introduce positive recovery strain in to the models.

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