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Repeated load relaxation testing of pure polycrystalline nickel at room temperature using nanoindentation

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We present the results of repeated relaxation tests using nanoindentation to derive the activation volume of the dislocation velocity and the ratios of the dislocation density and dislocation velocity. An experimental technique, based on classical uniaxial relaxation experiments, was developed to establish a constant strain during repeated load relaxation transients and then to calculate the stiffness of unloading, and therefore the hardness, across the transients with acceptable results. We found that the activation volume of the dislocation velocity from our nanoindentation methodology was in good agreement when compared to the same reported for uniaxial experiments. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4862799]

A plastically deforming crystalline solid is subject to various competing atomistic processes that contribute to the flow stress and the ability for the material to work harden. In the case of metals, these processes are primarily governed by the mechanisms that influence the movement of dislocations and their direct dependence on stress and temperature. The use of single transient tests (either constant load or constant strain) to study the deformation mechanisms in plastically deforming crystalline materials has been examined extensively using techniques primarily developed for monotonic uniaxial test machines.[1,2] These tests have provided the capability for the measurement of two key quantities, the strain rate sensitivity of the stress (m) and the activation volume (V^*), which are used to characterize the kinetic aspects of plastic deformation.[3–7] These two quantities are important for determining the macroscopic deformation mechanisms that are operating but provide little information on the specific microscopic processes that can be contributing to the flow stress. To address this, repeated transient tests (either repeated stress relaxations or creep) were developed to study the contribution of specific dislocation mobility mechanisms on the flow stress and in particular the relative influence of the thermally activated and athermal components.[8–10]

In recent years, the use of instrumented indentation testing (nanoindentation) to conduct similar transient tests (constant load or constant strain) and develop meaningful relationships with those performed in a uniaxial machine has been well documented in literature.[11–13] Caijun *et al.*[11] explained that the precaution when using these methods is in the interpretation of the data given the complex stress state that exists in indentation that is not present in traditional uniaxial tests. However, they provide a detailed analysis of different transient tests in indentation and conclude that meaningful power law creep parameters can be measured and are comparable to those measured in a uniaxial test. In addition, there have been multiple researchers who have used single transient methods to determine m and V^* for a variety of materials and have reported general agreement with those measured in uniaxial tests.[14–17] While examining the relationship between the indentation size effect (ISE) and m and V^*, Stegall and Elmustafa[18] concluded that using activation volume analysis from nanoindentation based on single transient creep tests V^* decreased with increasing hardness (or stress) while the strain rate sensitivity of the hardness was relatively constant regardless of load for a variety of FCC metals and alloys. These results were similar to the ones reported in the literature in prior uniaxial tests.[19–21] In this work, we present a methodology developed to perform repeated load (stress) relaxation tests using nanoindentation to obtain information about the relative mobility and density of dislocations. The measurement of dislocation densities and their interaction at varying load levels are keys to better understand the mechanisms that control the so called ISE.

The material chosen for this research was 99.95% pure polycrystalline Nickel. Two samples were cut to sizes suitable for indentation testing and annealed at 750 °C and followed by air cooling resulting in an average grain size of $\sim$100 μm.

A Nanoindenter XP (Agilent Technologies, Inc., Santa Clara, CA) equipped with a three-sided diamond Berkovich probe was used to conduct the repeated load (stress) relaxation tests. The Berkovich indenter was specifically fabricated to have a useful shape to about 30 μm for higher load (deeper depth) experiments. The test protocol was designed to initiate repeated loading cycles, each of which was followed by relaxation hold period of 30 s. During the relaxation hold segment the load was adjusted continuously to maintain the indentation depth constant and equal to the maximum value attained at the end of the preceding load segment. The displacement tolerance and the small incremental step load size for the dynamic calculation of the load during relaxation segment were adopted to achieve a constant indentation depth during relaxation period. Great care was taken to capture the final displacement upon termination of the loading cycle; this final displacement was used as the set-point for control

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during the relaxation hold segment. The tolerance and increment step size were varied for the different repeated loading cycles of the tests to minimize the noise during the relaxation phase. The load limit was varied from 50 mN to 500 mN. In this paper we only report on the 50 mN and 500 mN. The time to load for the first loading cycle was set at 20 s and the same loading rate was followed for subsequent cyclic loadings. Only the final relaxation segment is followed by the unloading cycle and the unloading rate was set equivalent to the loading rate. Figure 1 shows a typical set of repeated relaxations for tests at two different maximum loads, 50 mN and 500 mN.

The stiffness of contact was determined by the slope of the load and displacement curves during the unloading segment of the test and the data were used to determine the mechanical properties by the Oliver–Pharr analysis.[22] The nanoindenter continuously records displacement, load, and time throughout the plunge. The hardness is calculated based on contact depth at the point of unloading following the end of the hold segment for a standard load control test. To accomplish this, the contact depth was extrapolated through the hold segment using the as calculated values provided by the test protocol at the end of the hold time at the point of unloading. The contact depth was determined by establishing the stiffness through the hold segment according to

$$S = \sqrt{\frac{4P}{\pi * (\frac{H}{Er})}}, \tag{1}$$

where S is the stiffness, P is the load, H is the hardness, and E_r is the reduced modulus.

Based on Eq. (1), we were able to back out the stiffness values at different load levels. Next, the contact depth was calculated using Eq. (2) and subsequently the contact area by Eq. (3). The hardness is determined from the load and the contact area

$$h_c = h - \varepsilon\left(\frac{P}{S}\right), \tag{2}$$

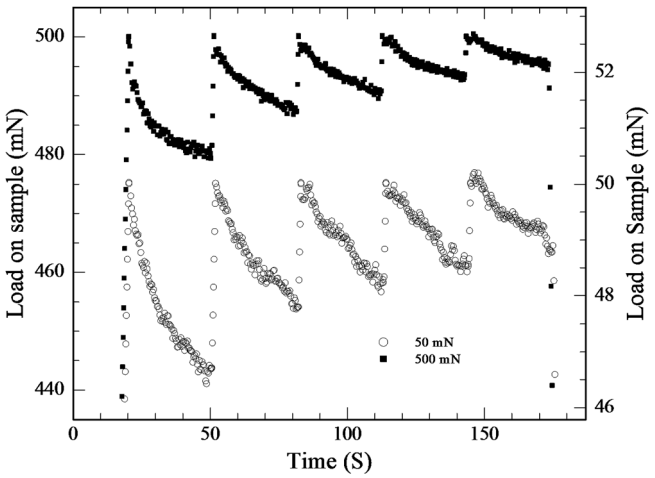

FIG. 1. Incremental load relaxations versus time for maximum loads of 500 mN and 50 mN.

where h_c is the contact depth, h is the displacement, ε is 0.75 for a Berkovich tip, P is the load, and S is the stiffness

$$A_c = f(h_c). \tag{3}$$

Figure 2(a) shows the indentation load versus the displacement for a 50 mN load control experiment with five incremental loading steps identified as A through E. The load displacement curve is represented by the solid line and the dotted line shows the extrapolated contact depth. To confirm that the extrapolated values in Figure 2(b) were derived correctly, the hardness values were calculated using the load corresponding to the intermediate increments A through D of Figure 2(a) since the hardness value at E is automatically retained by the machine. It was found that the hardness values correlate well with the load control method and the results are indicated in Table I.

The purpose of this work was to develop a technique capable of examining the coupled relationship between V^* and

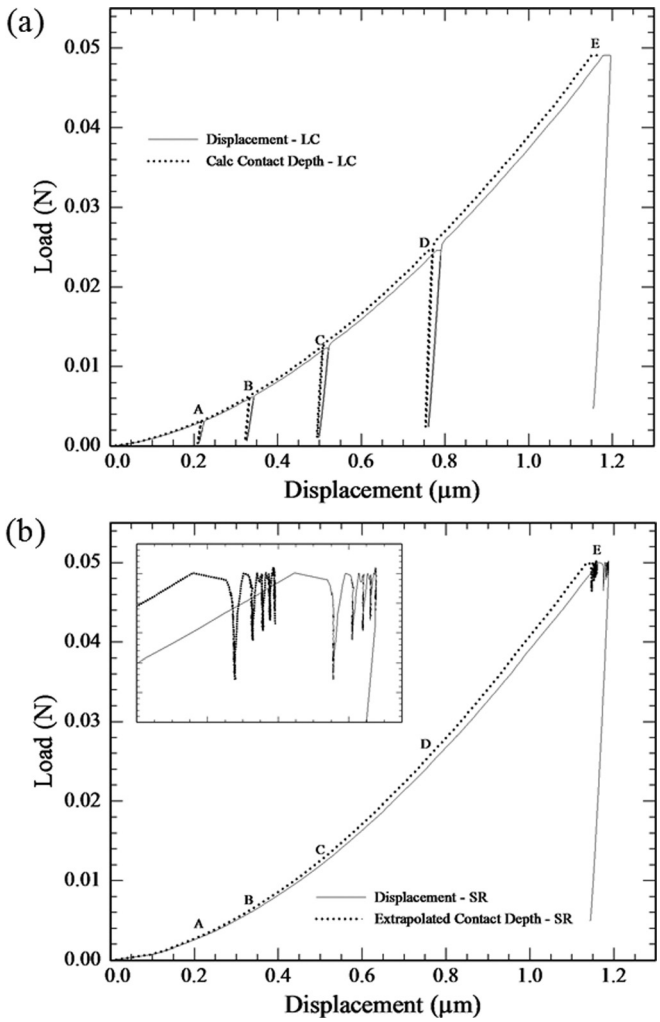

FIG. 2. (a) Load versus displacement curve using the load control test protocol with 5 incremental unloading segments. The dotted line shows the calculated contact depth while the hardness calculated at each unloading are represented by A through E and are given in Table I. (b) Load versus displacement curve using the load control test protocol modified for repeated load relaxations. The dotted line shows the calculated contact depth while the hardness calculated at each unloading are represented by A through E and are given in Table I.

TABLE I. Comparison of the hardness results from load control and the extrapolated load relaxation experiments.

	Hardness (GPa)	
	Measured	Extrapolated
Interval	LC	SR
A	2.60	2.58
B	2.22	2.19
C	1.93	1.93
D	1.67	1.71
E	1.46	1.46

the components controlling the dislocation mobility using nanoindentation. Some researchers have argued that attempting to perform repeated relaxation testing in indentation is problematic to obtain valuable results due to the complex state of strain ahead of the indenter.[23] We demonstrated that a repeated relaxation indention experiment can be implemented to equivocally generate results similar to those obtained using traditional uniaxial experiments.[8,10,23] Further, we have shown that the hardness (stress) can be determined with confidence at various load levels by extrapolation of the contact stiffness using the Oliver and Pharr analysis allowing for the precise monitoring during the relaxation hold segments. Therefore, we believe using repeated relaxation indentation experiments can be used to examine dislocation mechanisms that may be influencing the ISE. Based on this perspective, we are specifically interested in establishing relationships between dislocation density and velocity in the presence of an ISE.

In dislocation plasticity, the activation volume represents the volume in which dislocations are mobile during thermal activation. In terms of the measured hardness, the apparent activation volume is generally defined by Eq. (4) and linked to the strain rate sensitivity of the hardness by Eq. (5).[14] In these equations k_B is Boltzmann's constant, T is the absolute temperature (K), H is the hardness, $\dot{\varepsilon}$ is the strain rate, and 9 is a factor that represents the Taylor factor and the ratio of hardness to yield strength. It can be seen that the activation volume is a rate dependent quantity

$$V^* = 9k_bT\left(\frac{\partial \ln \dot{\varepsilon}}{\partial \ln H}\right), \tag{4}$$

$$m = \left(\frac{\partial \ln H}{\partial \ln \dot{\varepsilon}}\right). \tag{5}$$

The Orowan equation is represented by Eq. (6) and shows that the strain rate is a function of the density of a moving dislocation ρ_m with an average velocity v

$$\dot{\varepsilon} = b\rho_m v. \tag{6}$$

Caillard and Martin showed that when Eqs. (4) and (6) are combined, V^* is examined based on the relative contributions of the ρ_m and v, but it is not possible to discern which is dominating the stress.[8] We modified their relationship to take into account the ratio of the flow stress to hardness and the corresponding result is given by

$$V^* = 9k_bT\left(\frac{\partial \ln \rho_m}{\partial H} + \frac{\partial \ln v}{\partial H}\right). \tag{7}$$

The activation volume V^* is classically determined based on Eq. (4) from a single transient creep or load relaxation test.[13] However, in order to examine the contribution of the rate of the dislocation density to the hardness and the rate of the dislocation velocity to the hardness as given by Eq. (7), we need to define V_r, a phenomenological activation volume and V, activation volume of the dislocation velocity which are both needed for the stress relaxation analysis according to Caillard and Martin.[8] The first of these is V_r which is experimentally determined in conjunction with a time constant C_r to characterize the logarithmic nature of the relation. The second value V is a measure of the dislocation density moving between the beginning and end of successive relaxations. Therefore V_r, C_r, and V can be defined by the relations given in Caillard and Martin[8] and modified here to account for hardness as

$$\Delta H = \left(\frac{-9K_bT}{V_r}\right)\ln\left(\frac{1+t}{C_r}\right), \tag{8}$$

$$V = \frac{9K_bT}{\Delta H}\left(\ln\frac{\dot{\varepsilon}_{i2}}{\dot{\varepsilon}_{f1}}\right). \tag{9}$$

In order to investigate the ISE from an activation volume of the dislocation velocity perspective, V, a parameter Ω which represents the ratio V_r to V needs to be defined to draw the relationship between ρ_m and v

$$\Omega = (1+\beta)\left(1+\frac{K_r}{M}\right), \tag{10}$$

where the value of K_r is the work hardening coefficient and M is the combined modulus of the machine and the specimen.[8] The value of C_r is used to determine the ratio of ρ_m/ρ_{mo} that describes the change in the mobile dislocation density over the transient to the initial mobile dislocation density. This ratio is given by[8]

$$\left(\frac{\rho_m}{\rho_{mo}}\right) = \left(\frac{C_r}{C_r+t}\right)^{\frac{\beta}{1+\beta}}. \tag{11}$$

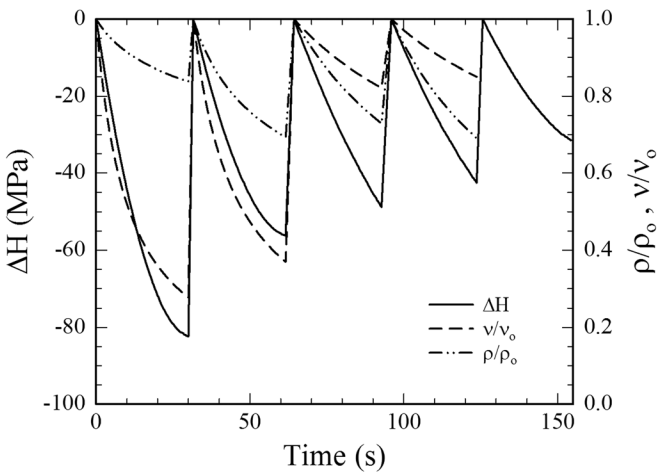

FIG. 3. Hardness (stress) relaxations versus time compared with the ratio of v/v_o and ρ/ρ_o for the 50 mN maximum load.

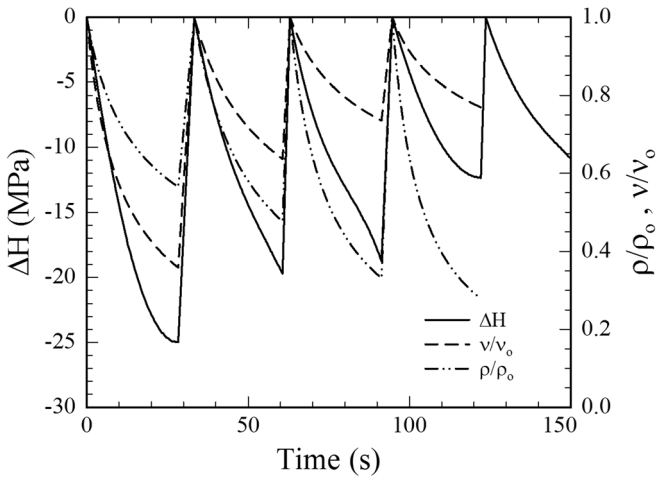

FIG. 4. Hardness (stress) relaxations versus time compared with the ratio of v/v_o and ρ/ρ_o for the 500 mN maximum load.

Finally, according to Caillard and Martin[8] the ratio of the dislocation velocity v can be determined using the relationship given in

$$\left(\frac{\rho_m}{\rho_{mo}}\right) = \left(\frac{v}{v_o}\right)^{\beta}. \tag{12}$$

Figures 3 and 4 show the change in hardness with the ratios for the mobile dislocation density and velocity versus time for the repeated transients of a 50 mN and 500 mN maximum loads, respectively. The data in these figures show that the dislocation velocity increases as the dislocation density decreases over each successive transient. The magnitude of the activation volume of the dislocation velocity, V, was found to be on the order of $\sim 30b^3$ at 50 mN and $\sim 70b^3$ at 500 mN for the first relaxation segments. The value of V at 500 mN of $\sim 70b^3$ is comparable to the value of $\sim 100b^3$ reported by Wang *et al*. for bulk polycrystalline Ni obtained in a uniaxial relaxation test .[23] Further, they reported that in general the value of V tended to be a factor of two smaller than V_r for their experiments.[23] We found a similar trend in our data where the ratio of V_r/V (Ω) according to Eq. (10), was on the order of 2 for the first relaxation, but in our case the ratio tends to increase with successive relaxations. We associate this increase in Ω to be influenced by the ISE and will explore this issue in depth in forthcoming research.

The purpose of this work is to develop an experimental technique for indentation, beyond that of single transient creep, capable of generating data to examine the coupled relationship between V, ρ, and v for metals that exhibit an ISE. We have presented a method for performing repeated hardness (stress) relaxation tests using nanoindentation and have demonstrated that this experimental technique can generate results similar to those obtained using traditional uniaxial experiments. Specifically, we derived the activation volume of the dislocation density V, the phenomenological activation volume V_r, which reasonably compare to previously published values for polycrystalline nickel in tension utilizing established closed form solutions. We found that the ratio of the initial to the final dislocation density decreases with increasing depth of indentation while the dislocation velocity increases. This finding would seem to indicate that as the depth of indentation decreases; the velocity of mobile dislocations is directly inhibited by the increase in the dislocation density and is at a minimum consistent with the accepted theory that the ISE is governed by a dislocation mechanism. A complete examination of the possible relationship between these results and the ISE along with additional pure metals will be covered in forthcoming research.

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