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TITLE: KINETICS ANALYSIS OF DIFFUSION OF POINT DEFECTS TO DISLOCATIONS
FOLLOWING PULSED NEUTRON AND ELECTRON IRRADIATIONS

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Kinetics Analysis of Diffusion of Point Defects to Dislocations Following Pulsed
Neutron and Electron Irradiations

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Résumé - The kinetics of point defect-dislocation interactions following pulsed neutron and electron irradiations has been studied in high purity copper. In all experiments, data showed a very rapid increase in the modulus defect followed by a more gradual change. The magnitude of the rapid change in the modulus defect was proportional to the fluence. The initial rapid rise was too fast to allow for kinetics analysis; however, the slower continuous process was analyzed. Results are interpreted in terms of a model wherein interstitials are rapidly depleted and responsible for the initial rapid increase; the slower process is due to vacancy diffusion and arrival at dislocations.

Abstract - The kinetics of point defect-dislocation interactions following pulsed neutron and electron irradiations has been studied in high purity copper. In all experiments, data showed a very rapid increase in the modulus defect followed by a more gradual change. The magnitude of the rapid change in the modulus defect was proportional to the fluence. The initial rapid rise was too fast to allow for kinetics analysis; however, the slower continuous process was analyzed. Results are interpreted in terms of a model wherein interstitials are rapidly depleted and responsible for the initial rapid increase; the slower process is due to vacancy diffusion and arrival at dislocations.

I - INTRODUCTION

The identification of the defect or defects responsible for the observed dislocation pinning in copper around room temperature has been the objective of several investigations /1-6/. In the early work of Peró and Thompson /2/, the defect was preliminarily identified as the vacancy. In more recent work by Thompson and Buck /4/, however, the pinning defect has been interpreted as "an interstitial". Identification of the pinning defect in each case was based primarily on an interpretation of a derived value of the migration energy. An important feature of these results was the observation of "instantaneous" pinning followed by a "delayed" pinning process. Only this latter process was utilized in obtaining the migration energies. No comprehensive model describing both pinning mechanisms and the responsible defects has been presented.

Experimental conditions to study instantaneous and delayed pinning mechanisms can be obtained by pulsed irradiations of short enough duration to complete prior to the initial pinning process. In the experiments presented herein, we have used pulsed neutron (40 μ sec) and pulsed electron (400 μ sec) irradiations to study the arrival of the pinning defects in copper from 310 to 430 K. Using this method a controlled concentration of Frenkel pairs is produced instantaneously in the lattice and their diffusion to dislocations is observed as a function of time.

II - EXPERIMENTAL PROCEDURE AND ANALYSIS

Experimental details are similar to our previous work /7/. The pulsed neutron experiments were conducted using a fast burst assembly which produces a fission-like spectrum of about 40 nsec duration leading to a fluence of 10^{14} n/m². Electron irradiations were performed using 1.0 MeV electron in 0.4 second pulses generated by a mechanical shutter. Background modulus data was also taken and changes were subtracted for the experimental neutron data used in the kinetics analysis. The much larger changes in the electron results make this correction negligible.

The number of pinning points added to the dislocations can be related to the modulus defect following Simpson and Sosin /8/

$$n_d = \frac{(\Delta Y/Y)_0}{(\Delta Y/Y)} - 1 \approx N_{dy} = (\tau_0^2 - \tau^2) / (\tau^2 - \tau_c^2)$$

where the subscript 0 refers to the pre-irradiation values and the subscript c refers to the saturated value. The general features of the results for N_{dy} are shown in figure 1. This uncorrected neutron data starts with a smooth continuous increase in N_{dy} at 310K. As the temperature increases, a sharp increase in N_{dy} , particularly noticed first in the 350K data occurs followed by a smooth continuous increase. Note that the magnitude of the sharp rise increases by roughly a factor of two from 350 to 430K. The exact shape of this initial sharp increase is influenced by the uncertainty in the zero time inherent in the experimental technique. The data clearly suggest two pinning processes with different time/temperature behaviors.

The smooth data following the sharp rise has been analyzed for its kinetic behavior. For the pulsed irradiation conditions, the arrival of defects at the dislocation can be viewed as a precipitation process with a relaxation time /9/. Starting from the general case of continuous relaxation spectra /10/ and following Alfrey and Doty /11/, the relaxation time τ can be found from the slope of ΔN_{dy} versus $\ln(t)$. Standard least-square methods were applied to fit fifth order polynomials to the data, avoiding the very short time, sharp rise. The migration energy obtained from

$$E^M = k \Delta \ln(\tau) [1/T_2 - 1/T_1]$$

was found to be 0.89 ± 0.27 eV, in good agreement with the single vacancy migration energy in copper /12/. To further test the hypothesis of a single migration energy, the value of E^M has been used to renormalize the data to be displayed as a single continuous curve using the relation

$$t = t \exp(E^M / k[1/T_c - 1/T])$$

where the reference temperature T_c is 370K. The results for the neutron irradiations are shown in figure 2. The temperature dependence of N_{dy} /1,7,9/ was accounted for by normalizing the data at the 370K results using an arbitrary point in the smooth part of each curve. The approximation to a continuous curve is remarkable.

III - DISCUSSION

The dose dependent neutron and electron data shown in figure 3 agree within roughly $\pm 5\%$, with the exception of the low dose 10^{-2} data. Examination of the data shows that both the sharp rise in N_{dy} and the long time behavior are proportional to dose. Additional information about the nature of the pinning defects can be obtained by examination of the normalized decrement δ/δ_0 . Let's assume that the sharp rise in N_{dy} is due to one type of pinning defect (interstitial) and the longer time smooth increase is due to a second defect (vacancy). The normalized decrement data in figure 4 show an initial increase in δ/δ_0 and at high temperature/dose δ/δ_0

starts to decrease after the initial rise forming an anomalous peak /14/. The values of δ/δ_0 for the 310 and 350K neutron data increase to a constant value. During the first 400 seconds only interstitials are pinning the dislocation at these temperatures. This interpretation is required to explain the increase in δ/δ_0 since it must be the defect responsible for the peaking effect. In terms of the nature of the peaking effect the effective value of u is less than u_0 /14/. As the temperature increases, a short time increase in δ/δ_0 remains but at longer time, δ/δ_0 starts to decrease. Further, the time at which the decrease clearly starts shifts to shorter times as the temperature increase. This is similar in nature to the temperature dependence of the N_V data. The arrival of vacancies which is strongly temperature dependent for the experimental conditions is responsible for the decrease in δ/δ_0 . Simpson and Kerkhoff /15/ have shown that while electron irradiation can produce the peaking effect in copper, quenched-in vacancies only produce decreases in δ/δ_0 . The dose dependence results of δ/δ_0 can be explained in a similar manner to the temperature data. The absolute number of interstitials and vacancies increases with the dose increase. Initially the primary effect is to increase δ/δ_0 due to the arrival of interstitials. The decrease in δ/δ_0 occurs because the absolute number of vacancies increases. If only interstitials were pinning δ/δ_0 would increase to a constant value for an effective value of $u < u_0$. Above u_0 , δ/δ_0 would go through a peak and then decrease and remain constant at a value dependent on dose.

VI - CONCLUSIONS

The combined analysis of pulsed neutron and electron irradiations of high purity copper including both modulus and decrement data reveals the existence of two distinct irradiation produced pinning processes. The first process occurs too rapidly in the temperature range 310 - 430 K to allow quantitative kinetic analysis. It produces an increase in the modulus defect and an increase in the decrement. This process is due to interstitial pinning. The second process produces an increase in the modulus and a decrease in the decrement. A kinetic analysis using the approximation method of Alfrey and Doty yields a migration energy of $E^m = 0.89 \pm 0.27$ eV which is consistent with the value of $E^m = 0.71$ eV for the single vacancy within the experimental and analytical uncertainties. This process is due to vacancy pinning. Work supported by the U. S. Department of Energy.

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Figure 1: Raw pulsed neutron data for N_{dy} vs $\log t$ at irradiation temperatures from 310 to 430 K. Doses in 10^{14} n/m²: 310K--10.2; 330K--11.1; 350K--7.58; 370K--9.58; 380K--10.5; 390K--10.4; 410K--10.1; 430K--9.75.

Figure 3: 370K pulsed neutron and electron data for N_{dy} vs $\log t$ at several doses /13/. The neutron (1n,...) results are normalized to the 4n data. The electron (1e,...) results are normalized to the 3e data. Note that $N_{dy}(e) = 10 N_{dy}(n)$.

Figure 2: Pulsed neutron data for N_{dy} plotted as a function of $\log t$ using $E = 0.8$ eV.

Figure 4: Decrement data plotted as a function of time for the pulsed neutron irradiations for several temperatures. See figure 1 for doses.