CONF-751006--6

75-114

RECOVERY IN STAGES I AND II OF THERMAL AND FISSION NEUTRON IRRADIATED MOLYBDENUM

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ABSTRACT

The influence of initial dose and irradiation doping upon the recovery of Mo was studied for the markedly different types of damage produced by thermal and fission neutrons. The features of the Stage I recovery commonly seen for several FCC metals can be identified, including a I_D peak at 40°K. The typical dose-dependent behavior of a I_E subpeak was observed at ∿ 47°K, and evidence for free interstitial migration is further supported by irradiation doping results. Stage II shows a first-order peak at 120°K in which the population percentage increases with increasing initial dose in opposite fashion to FCC impurity detrapping peaks.

INTRODUCTION AND EXPERIMENTAL

The production and recovery of thermal and fission neutron damage were studied in nearly identical Mo wires with resistance ratios of 1900 (thermal) and 1790 (fission). The specimens were mounted in a common irradiation capsule located in the sample chamber of the ORNL low temperature irradiation facility[1] which receives a highly thermalized neutron flux. The capsule contained liquid helium at 4.9°K during irradiation and 3 atm of exchange gas during annealing studies. The fission neutron sample in the lower half of the capsule was inserted into a small ^{235}U tubular neutron converter lined inside with Gd foil to complete the absorption of incoming thermal neutrons. This arrangement produced very pure fission neutron damage in the sample from a calculated fission neutron flux of 5.1×10^{10} n/cm^{2}sec. The thermal neutron sample in the upper half of the capsule where the average fission flux was negligible received a thermal flux of 1.4×10^{12} n/cm^{2}sec. The location of both samples in a common

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irradiation capsule provides the opportunity to compare closely production and recovery results for damage produced by primary recoils with greatly different energies. In Mo only 5% of the thermal neutron captures produce transmutations (^{99}Tc, ^{101}Ru during an experiment, whereas every capture is estimated (see Table 1) to produce 1.8 Frenkel pairs on the average. Since only about one Frenkel pair in 36 has a related transmutation, their influence is not expected to be significant.

In addition to production characteristics, dose-dependent recovery and irradiation doping experiments were done to determine the temperature at which free interstitials migrate. Although there is agreement on the temperatures at which recovery processes occur in the range below 55°K, there is not complete agreement as to which is related to free interstitial migration. The delineation of free interstitial migration from other low temperature recovery is important, since it determines the nature and amount of damage remaining at higher temperatures.

DAMAGE PRODUCTION

Damage resistivity, $\Delta\rho$, was measured after irradiation at 4.9°K for a low dose, Φ, where no significant irradiation annealing is expected.[2] $\Delta\rho$ and Φ along with the incremental damage rate $\Delta\rho/\Phi$ are listed in Table 1. The fission neutron rate of 3.6×10^{-25} ($\Omega cm^3/n$) is in close agreement with 3.2×10^{-25} measured by Horak and Blewitt[2] with a very slightly energy degraded fission neutron spectrum. The specific resistivity, $\delta\rho$, related to each type of neutron-atom recoil event is calculated by $\delta\rho = \Delta\rho/\sigma\Phi$ where σ is the thermal neutron capture or the average fission neutron scattering cross section, and the results are in Table 1. The damage ratio, $\delta\rho_F/\delta\rho_T$, depends only upon measured quantities and is the ratio of the amount of damage produced by each type of recoil event. Table 1 shows that one fission neutron scattering event produces 35 times as much damage as one thermal neutron capture event. The number of Frenkel pairs produced by each type of event can be estimated by $n_F = \delta\rho/\rho_F$ if the Frenkel pair resistivity ρ_F is known. We choose a value of $\rho_F = 8.5\times10^{-4}$ Ωcm/unit conc. of pairs from a commonly used empirical rule that $\rho_F = 1.5$ times the room-temperature resistivity. Values of n_F are listed in

Table 1. Characteristics of Neutron Damage Production

	Thermal	Fission
$\Delta\rho$ (nΩcm)	0.55	1.72
Φ (n/cm^2) (low dose)	1.4×10^{17}	4.8×10^{15}
$\Delta\rho/\Phi$ (Ωcm^3/n)	3.9×10^{-27}	3.6×10^{-25}
σ (barns)	2.5	5.6
$\delta\rho$ (Ωcm/unit conc. recoil event	15.6×10^{-4}	550×10^{-4}
Damage ratio (fission/thermal)	35	
n_F	1.8	65
Calculated damage energy (eV)	133	18,000
Damage energy ratio (fission/thermal)	135	

Table 1. Also in Table 1 are new damage energy values[3,4] calculated for each type of recoil event. Although significant disagreement between the measured (35) and the calculated (135) damage energy ratios remains, this comparison is improved over old results. The net result is that these two types of recoil events cannot be compared on the basis of a simple model which predicts damage production to be proportional to damage energy.

RECOVERY

Figure 1 shows the isochronal recovery rates after two doses which differ by a factor of 4.3. The locations of the major recovery peaks agree with those found after electron[5,6] and other fast neutron[7,8] irradiations, although the percentage population differs according to particle type and initial dose. (Although the high and low doses are not equal for each type, the high thermal and low fission neutron doses are comparable.) An overall correspondence in the recovery patterns can be seen for the two types, but the peak resolution and population are reduced in the fission neutron damage. It is notable, however, that in the fission neutron damage the first-order processes indicated at 14, 23, and 27°K, which are probably close pair recovery, remain distinct for Mo, in contrast to Cu and Al which show only continuous recovery over the corresponding range.[9] This suggests that localized defect concentration in the displacement cascade is lower in Mo than in Cu and Al.

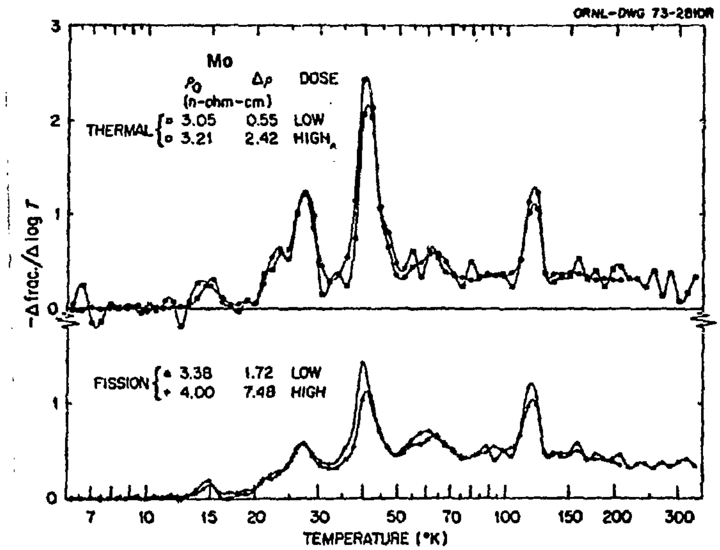

Fig. 1. Isochronal recovery rates of thermal and fission neutron damage in Mo. All annealing programs were identical with 5 min. pulses and $\Delta T = 0.055\ T$.

The relatively long span from 5 - 13°K with no recovery (the peak at 6°K and the negative peaks at 7 and 12°K are considered spurious since they occur only in the low dose thermal data, which are least accurate) strongly indicates the irradiation temperature of 4.9°K is low enough to freeze in all the damage produced in Mo. The overall resemblance of the recovery pattern to Stages I and II of the FCC metals is good if the peaks at 14, 23, and 27°K are identified as close pair substages, the peak at 40°K as I_D correlated pair recovery, and the peak at 120°K as the detrapping of defects from impurities. Certain features, however, differ from the FCC metals in the Stage II region between 50 - 300°K. The peak at 120°K increases in size with increasing initial dose which is opposite in behavior to that seen for Stage II detrapping peaks in FCC metals. Also, the portion of Stage II related to background rather than peak recovery is large compared to that in FCC metals for comparable defect concentrations.[10]

DOSE DEPENDENCE

We know of no dose-dependent studies which observed effects of free interstitial migration as seen for the FCC Stage I_E. The resolution of possible effects in previous fast neutron studies was obscured by high initial doses. Effects in previous electron studies may have been difficult to see because only small populations of free defects were produced due to low energy recoils. With variation of initial dose in the low dose range, as in the present experiment, two effects can be expected in the FCC model. First, a Stage I_E subpeak moves to lower temperatures with increasing dose. In the thermal neutron results in Fig. 1 the subpeak centered at 47°K follows this behavior and shows a relationship to the 40°K peak similar to that seen between the I_D and I_E peaks in FCC metals. Second, the recovery of a high dose should be greater than a low dose in Stage I_E because interstitial trapping reactions are reduced in favor of recombination with vacancies. The curves in Fig. 2 show this more clearly than Fig. 1. It can be noticed that enhanced recovery can also be detected in the 40°K peak. This is consistent with the idea[9] that if the concentration of defects ($\sim 6.5 \times 10^{-7}$ in the low thermal dose) is less than impurities, then the latter can trap correlated interstitials. Applying this argument to the 40°K peak of the fission neutron data suggests that some correlated pairs are located in sparsely populated regions where impurities can have a relatively strong influence. This result is not consistent with the usual fast neutron damage picture of a densely populated, vacancy-rich cascade fringed with relatively free interstitials.

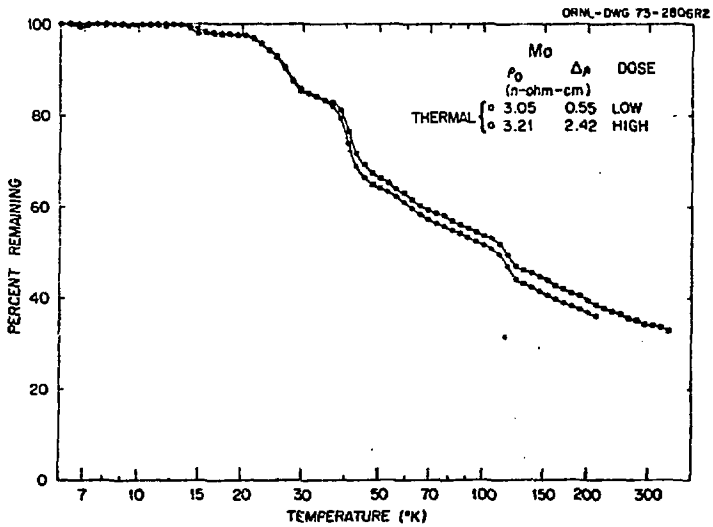

Fig. 2. Isochronal recovery of thermal neutron damage in Mo.

RADIATION DOPING

After anneal of the second (high dose) irradiation to 240°K, another nearly identical low dose isochronal study was made. The annealed damage remaining from the high dose (36% thermal, 42% fission) served to dope the samples. For several FCC metals it is well known that radiation doping similar to this enhances Stage I_E recovery. The enhancement occurs because during annealing of the doping damage the interaction cross section of the remaining interstitials is reduced due to clustering, while the surviving vacancies remain dispersed to give the doping damage an effectively vacancy-rich character. Figure 3 shows for both types of damage that enhanced recovery of the doped specimens begins near 42°K and proceeds in a manner consistent with that for FCC metals. The behavior of the Stage I_E subpeak near 47°K is not a shift of the undoped recovery to lower temperatures, but rather indicates the greater Stage I_E recovery in the doped case. Enhanced recovery in the 120°K peak for both types of damage is consistent with the idea that that this peak represents the release of trapped interstitials which then have a better opportunity in the doped case for recombination with a vacancy. Enhanced recovery in the fission neutron data between 50 - 100°K indicates that free interstitials may also be moving in this range, but the recovery spectrum does not permit interpretation of mechanisms.

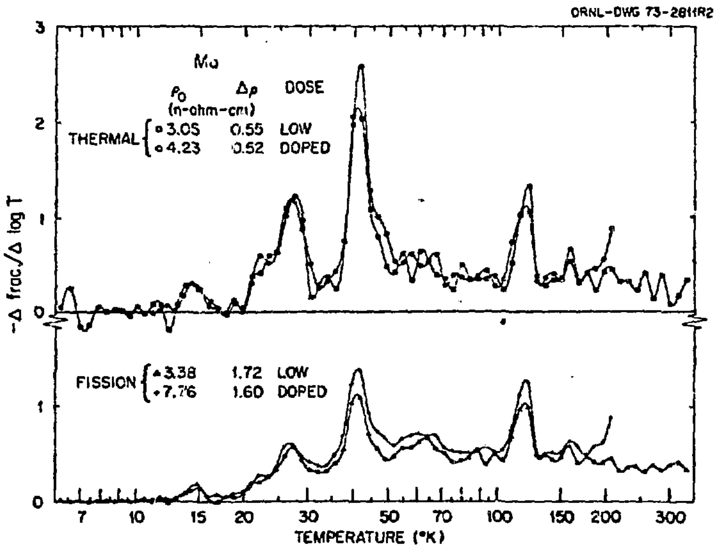

Fig. 3. Isochronal recovery rates of doped and undoped thermal and fission neutron damage in Mo.

CONCLUSIONS

1. The recovery of Mo up to 240°K can be identified with Stages I and II of the FCC metals, although some details differ.

2. Identification with the I_D substage is made for the 40°K peak, and the I_E peak is indicated but requires a smaller defect concentration than in the FCC metals to obtain comparable resolution. No indications of a free interstitial proposed by Hanada et al.[11] to move at 22°K are observed.

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