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FISSION NEUTRON DAMAGE IN NIOBIUM

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ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION
Oak Ridge, Tennessee

February 1976

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THE CHARACTERISTICS OF 15 MeV AND
FISSION NEUTRON DAMAGE IN NIOBIUM*

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ABSTRACT

Displacement damage by 15 MeV (d-Be source) and fission neutrons at 30°C in high purity niobium single crystals has been studied by transmission electron microscopy. The fluence of the 15 MeV neutrons was $1.5 - 2.0 \times 10^{17} \text{ n/cm}^2$ and the fluence of the fission neutrons ($5 \times 10^{17} \text{ n/cm}^2$) was chosen so that samples from both types of irradiations had approximately the same damage energy. In both 15 MeV and fission neutron irradiated specimens, the loops were observed to be about 2/3 interstitial and 1/3 vacancy type. The analysis of Burgers vectors of the dislocation loops showed that more than 2/3 of the loops were perfect $a/2\langle 111 \rangle$ and that the rest were $a/2\langle 110 \rangle$ faulted. It is concluded that at equal damage energies, the detailed nature of the damage is the same for 15 MeV and fission neutron irradiated niobium.

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*Research sponsored by the U. S. Energy Research and Development Administration under contract with the Union Carbide Corporation.

1. INTRODUCTION

An understanding of the nature of displacement damage due to high energy neutrons is an important consideration for the evaluation of fusion neutron radiation effects and for the simulation of fusion neutron damage by fission neutrons. In this paper, we have investigated the characteristics of defect clusters resulting from high energy neutrons (d-Be source) and fission neutrons in niobium. The primary recoil spectrum in Nb from the d-Be source is similar to that from pure 15 MeV neutrons (D-T source) [1]. Transmission electron microscopy was used to determine the vacancy-interstitial nature of observed loops, their Burgers vectors and the incidence of multiple clusters of loops based on examination of large numbers of loops in both the high energy and fission neutron irradiated specimens.

2. EXPERIMENTAL DETAILS

The high energy neutrons were generated at the Oak Ridge Isochronous Cyclotron (ORIC) by stopping a 40 MeV deuteron beam in a thick Be target. The deuteron-breakup (d,n) reaction results in a neutron spectrum which is broadly distributed in energy about a peak at approximately 15 MeV with a small fraction extending above 30 MeV. The details of this source are described elsewhere [2]. The ORIC source produces a maximum flux $2 \times 10^{12} \text{ n/cm}^2 \text{ sec}$ at the specimen chamber with a spatial full-width-half maximum of approximately 7mm. This source has been well characterized in terms of spectrum and dosimetry and detailed calculations of damage energy and associated

quantities have been reported previously [1,2]. The niobium single crystals were grown from .05cm thick sheets of zone - refined starting material using the strain anneal technique. The impurity content in the annealed Nb is given in Table 1. The detailed irradiation technique has been described in ref. 2. Briefly, the irradiations were carried out at about 30°C on a continuous basis over a period of 30 hours with a total spectrum dose of $1.8 \times 10^{17} \text{n/cm}^2$ at the center of the sample. Dosimetry foils were placed before and after the sample capsule.

The low energy neutron irradiations were performed in the CP-15 facility of the Oak Ridge Bulk Shielding Reactor where the neutron spectrum is very similar to the U^{235} fission spectrum. The specimens were irradiated to a fluence of $5 \times 10^{17} \text{n/cm}^2$ ($E > 0.1 \text{ MeV}$) at a flux of $5 \times 10^{12} \text{n/cm}^2 \text{ sec}$ (temperature of irradiation $\sim 40^\circ\text{C}$).

3. RESULTS AND DISCUSSION

3.1 Nature of Defects

The nature of the dislocation loops (vacancy or interstitial) was determined from their contrast behavior under two beam dynamical diffraction conditions as a function of loop position relative to the surface. Loop positions were determined with an uncertainty of $10\text{\AA}$ by stereomicroscopy. Fig. 1 a and b show micrographs under $[1\bar{1}0]$ dynamical diffraction conditions of 15 MeV neutron and fission neutron damage respectively. Small black dots in the micrographs are gold islands deposited on the surface as a reference for the stereomeasurements. The contrast behavior, i.e., the direction and shape of the

black-white contrast, was compared with recent contrast calculations which take the shear component of the Burgers vector into account and can treat loops of general orientations [3,4]. The direction of the contrast combined with the depth position in the foil identifies the vacancy or interstitial nature of a loop, and the results obtained for the 15 MeV irradiations are shown in Fig. 2, and the results for fission neutron irradiations shown in Fig. 3. In both cases approximately 2/3 of the loops are of interstitial type and 1/3 are of vacancy type.

3.2 Burgers Vectors of Dislocation Loops

In the past, it was assumed that the direction $\bar{L}$ of black-white contrast (the direction from the middle of the black lobe to the middle of the white lobe) under two-beam dynamical diffraction conditions would be parallel to the projection of the Burgers vector ($\bar{b}$) onto the image plane. More recent calculations have shown that $\bar{L}$ is not necessarily parallel to the projected direction of the Burgers vector and the following rule was found to hold true [4]:

$$\alpha = \frac{\beta + \gamma}{3} \quad (1)$$

where α is the angle between diffraction vector $\bar{g}$ and black-white direction $\bar{L}$, β is the angle between the diffraction vector ($\bar{g}$) and the Burgers vector $\bar{b}$ and γ is the angle between the diffraction vector $\bar{g}$ and the loop normal $\bar{n}$; $\bar{b}$ and $\bar{n}$ are projected onto the image plane. By applying the results of these calculations, the black-white

contrast of about 120 loops in each of the 15 MeV and fission neutron irradiated samples was analysed. It was found that in both irradiations more than 2/3 of the observed loops were perfect $a/2\langle 111 \rangle$ and the rest being $a/2\langle 110 \rangle$ faulted loops. The foil orientations used in the present experiments were such that it was not possible to unambiguously distinguish $a\langle 100 \rangle$ loops from both $a/2\langle 111 \rangle$ and $a/2\langle 110 \rangle$ loops. However, $a\langle 100 \rangle$ loops are believed to have high strain energy and the dislocation reaction:

which could result in the formation of $a\langle 100 \rangle$ loops is not expected to occur frequently.

3.3 Multiple Clusters

It has been reported that there are 'many' multiple clusters present in niobium irradiated with 15 MeV neutrons [5]. It was suggested that these multiple clusters were due to splitting of displacement cascades associated with high energy > 150 Kev primary recoil events and presumably not present in fission neutron irradiated material. If multiple clusters are formed in this way, one would expect to identify them by the presence of two or more vacancy clusters separated by a distance less than about $100\text{\AA}$. In the present work, this aspect of the damage has been studied using weak beam and high order stereo microscopy techniques to obtain high resolution three-dimensional information from three fields which contained in aggregate ~ 700 defect clusters. Examples of the structures observed in weak

beam micrographs are shown in Fig. 4. In these dark field micrographs, the bright defect cluster images arise only from electrons diffracted from the regions very close to the dislocation loops [6] and thus provide better information on the size and position of a cluster than can be obtained in conventional bright field micrographs. Only cluster images $< 100\text{\AA}$ apart represent possible multiple clusters and constitute $\sim 10\%$ of the total defect clusters observed. Stereo analysis has shown that in approximately one-half of these possible cases, the true separation distance is greater than $100\text{\AA}$. Analysis of the dislocation loop nature of the remaining closely spaced clusters showed that for about $3/5$ of these one or more of the clusters were interstitial in nature. The remaining cases where two or more vacancy clusters are less than $100\text{\AA}$ apart amount to about 2% of all the defect clusters in the sample volume studied. A similar study of the fission neutron irradiated specimens indicated a smaller percentage ($\sim 0.25\%$) of multiple clusters.

This latter concentration of vacancy cluster pairs is approximately that expected for a random distribution. The results for the 15 MeV neutron case appear to be much lower than the "many" multiple clusters [5] previously reported but greater than indicated by a previous study which failed to find multiple clusters [2]. This variability of results possibly reflects fluctuations in the damage distribution on a microscopic scale. We have not been able to establish a correlation of the apparent frequency of occurrence of the multiple clusters with the primary recoil spectra which is consistent with the density of single vacancy clusters. In view of the

low frequency of occurrence of these multiple clusters, we do not believe they significantly modify the conclusions arrived at on other grounds as to the similarity of 15 MeV and fission neutron damage.

3.4 Size Distributions

Fig. 5 shows transmission electron micrographs of 15 MeV and fission neutron damage under a high order reflection ($3\bar{3}0$). The high order diffraction condition was used in order to obtain better contrast and resolution of the defect images [7]. These results, shown in Fig. 6, are essentially the same as reported previously [2] but represent better statistics. There are general similarities in the size distributions of the two irradiations. The total point defect densities were obtained by integrating these size distributions. A ratio of 2.7 was found for the retained damage resulting from 15 MeV neutrons compared to that for fission neutrons. This result is in good agreement with the theoretical calculations [1].

4. CONCLUSIONS

1. From contrast behavior under dynamical diffraction conditions and stereomicroscopy it was found that in both irradiations, about 2/3 of the observed loops were interstitial and 1/3 vacancy type.
2. More than 2/3 of the observed loops were perfect $a/2\langle 111 \rangle$ loops and the remaining were $a/2\langle 110 \rangle$ faulted loops.
3. The incidence of multiple clusters was about 2.0% in the high energy neutron irradiations.

The main conclusion which can be drawn from the present study

is that the nature of the 15 MeV neutron damage is essentially the same as fission neutron damage for room temperature irradiations. One can therefore infer from this observation that the clustering behavior is nearly independent of the energy of incident neutrons and the damage can be scaled by the deposited damage energy.

ACKNOWLEDGMENTS

The authors acknowledge helpful discussions with J. B. Roberto, T. S. Noggle, M. J. Saltmarsh and L. H. Jenkins.

TABLE I

Impurity Analysis in Nb						
	H	C	N	O	Ta	W
wt. ppm -	<5	50	10	50	50	15

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FIGURE CAPTIONS

- Figure 1. Electron micrographs of 15 MeV (a) and fission (b) neutron irradiated niobium. The two beam dynamical diffraction conditions employed for these micrographs gives rise to the black-white contrast at the damage clusters which provides information for determining Burger's vectors and the vacancy-interstitial nature of the loops. The arrows give the directions of the $[1\bar{1}0]$ diffraction vector employed and their length corresponds to 0.2μ
- Figure 2. The depth position of loops versus sense of the black-white contrast $\bar{g} \cdot \vec{r}$ for 15 MeV neutron irradiated niobium. Identification of vacancy loops indicated by $\square$, identification of interstitial loops by $\boxtimes$.
- Figure 3. The depth position of loops versus sense of the black-white contrast $\bar{g} \cdot \vec{r}$ for fission neutron irradiated niobium. Identification of vacancy loops indicated by $\square$, identification of interstitial loops by $\boxtimes$.
- Figure 4. Weak beam electron micrographs of 15 MeV (a) and fission neutron (b) irradiated Nb. Diffraction conditions: $+g[1\bar{1}0]$ dark field with an excitation error of $2 \times 10^{-2} \text{ \AA}^{-1}$.
- Figure 5. Bright field micrographs with higher order diffraction conditions; (a) 15 MeV neutron dose = $1.8 \times 10^{17} \text{ n/cm}^2$, (b) Fission neutron dose = $5.0 \times 10^{17} \text{ n/cm}^2$. The arrows in the micrographs are along the $[3\bar{3}0]$ diffraction vector and they are 0.2μ long.
- Figure 6. Loop size distributions in the 15 MeV and fission neutron irradiated Nb.

ORNL-DWG 75-17944R

15 MeV Neutron Damage in Nb.

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Fission Neutron Damage in Nb.

0.2 μ