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THE INFLUENCE OF OXYGEN IMPURITY ATOMS ON DEFECT CLUSTERS AND RADIATION HARDENING IN NEUTRON-IRRADIATED VANADIUM*

R. Bajaj
Advanced Reactors Division
Westinghouse Electric Corporation
Madison, Pennsylvania 15663

and

M. S. Wechsler
Ames Laboratory ERDA and Department
of Materials Science and Engineering
Iowa State University
Ames, Iowa 50010

ABSTRACT

Single crystal TEM samples and polycrystalline tensile samples of vanadium containing 60-640 wt ppm oxygen were irradiated at about 100°C to about 1.3×10^{19} neutrons/cm^2 (E>1 MeV) and post-irradiation annealed up to 800°C. The defect cluster density increased and the average size decreased with increasing oxygen concentration. Higher oxygen concentrations caused the radiation hardening and radiation-anneal hardening to increase. The observations are consistent with the nucleation of defect clusters by small oxygen or oxygen-point defect complexes and the trapping of oxygen at defect clusters upon post-irradiation annealing.

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INTRODUCTION

Several investigations have revealed a post-irradiation annealing resistivity stage in BCC metals at about $0.2T_m$ (T_m = melting temperature in °K); for reviews see[1-4]. The resistivity annealing stage has been observed in neutron-irradiated vanadium at about 175°C ($\simeq 0.2T_m$) by Perepezko *et al.*[5], Stanley *et al.*[6], and McIlwain *et al.*[7]. The origin of the $0.2T_m$ annealing stage has been debated in the literature: Johnson[8,9]

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indicates an explanation in terms of the annealing of an intrinsic radiation-produced defect such as the vacancy, whereas Wechsler et al.[10] favor the trapping of interstitial impurity atoms at radiation-produced defect clusters. Internal friction measurements (Snoek damping) in neutron-irradiated vanadium have also been reported by Stanley et al.[6], Eto et al.[11,12], and McIlwain et al.[7]. The Snoek peak due to oxygen in solid solution is observed to decrease during annealing at temperatures between 100 and 200°C. The decrease in oxygen in solid solution during annealing deduced from resistivity measurements and from oxygen Snoek damping were observed to be in fairly good agreement[7].

Radiation-anneal hardening[13], an additional increase in yield stress upon post-irradiation annealing, is also observed at $0.2T_m$ in vanadium[14-20]. The present work indicates that radiation-anneal hardening may be due to the motion of interstitial impurities to radiation-produced defect clusters, which strengthens them as barriers to slip dislocations. The present work also shows that the increase in yield stress of vanadium as indicated by samples in the as-irradiated condition (irradiation temperature, about 100°C) also increases with increasing oxygen concentration, as was reported earlier by Hasson and Arsenault[21] and Arsenault and Pink[22]. These effects are correlated with the density and size distribution of defect clusters as revealed by transmission electron microscopy (TEM).

EXPERIMENTAL PROCEDURE AND RESULTS

Single crystal TEM samples and polycrystalline tensile samples were prepared with oxygen concentrations ranging from 60 to 640 wt ppm oxygen and irradiated at about 100°C to about 1.3×10^{19} neutrons/cm^2 (E>1 MeV). A portion of the samples were subjected to one-hour post-irradiation anneals at temperatures up to 800°C. The density and size distribution of defect clusters in the TEM samples were determined with a Zeiss particle size analyzer and the tensile samples were tested at room temperature at a strain rate of 1.67×10^{-4} sec^{-1}.

The defect cluster size distribution curves had a peaked shape. For samples in the as-irradiated condition, the average size was 138, 77, and

73 Å for samples with 95, 300, and 500 wt ppm oxygen, respectively, and upon post-irradiation annealing at 500°C, the average size increased to 310, 191, and 157 Å, respectively.

The total density of defect clusters of all sizes is shown in Fig. 1 as a function of the post-irradiation annealing temperature, with the values for as-irradiated samples plotted at the irradiation temperature. It appears that the density of defect clusters increases with increasing oxygen concentration. Furthermore, we note that the defect cluster density first starts to decrease at about 300°, 400°, and 500°C for 95, 300, and 500 wt ppm oxygen, respectively, which indicates that the defect clusters are more stable against annealing for vanadium with higher oxygen concentrations. For anneals above 600°C, the defect cluster density was too low to enable meaningful counts of defect clusters.

In order to attempt to correlate the density and size distributions with radiation hardening, we employ a dispersed barrier model, such as that applied to neutron-irradiated niobium by Tucker and Wechsler[23] and Loomis and Gerber[24]. The increase in yield stress due to the radiation-produced barriers (defect clusters) is given by

$$\Delta\sigma = \sigma_i - \sigma_u = 2\,\frac{K_1}{K_2}\,Gb\left(\sum_j n_j d_j\right)^{1/2} \tag{1}$$

where σ_i and σ_u are the yield stresses in the as-irradiated and post-irradiation-annealed material, respectively, G is the shear modulus, b is the Burgers vector of the slip dislocation, and n_j is density of defect clusters in the jth size interval centered at diameter d_j. The constant K_1 arises from setting the force F necessary to surmount the barriers equal to $F = K_1Gb^2$. Also, K_2 arises from the possibility that

$$\ell = K_2\bar{\ell} \tag{2}$$

where ℓ is the effective interbarrier distance, $\bar{\ell}$ is the average interbarrier distance

$$\ell = \left(\sum_j n_j d_j\right)^{-1/2} \tag{3}$$

and K_2 is a constant close to unity that incorporates geometrical considerations as discussed by Kocks[25]. We may calculate $\bar{l}$ from our measured size distributions and the results are shown in Fig. 2 as a function of oxygen concentration. We see that $\bar{l}$ decreases with increasing oxygen concentration.

Figure 3 consists of a plot of $\Delta\sigma = \sigma_i - \sigma_u$ for vanadium with 60, 205, and 640 wt ppm oxygen as a function of annealing temperature. We see that the increase in yield stress upon irradiation and the radiation-anneal hardening increase with increasing oxygen concentration. Furthermore, the general recovery toward the unirradiated yield stress starts at about 500°C and is completed at 700°C.

DISCUSSION

The correlation between the density and size distribution of the defect clusters as determined by transmission electron microscopy and the radiation hardening as determined by yield stress measurements was carried in accordance with Eqs. (1-3). Values of $\bar{l}$ and $\Delta\sigma$ were taken from Figs. 2 and 3, respectively, and G and b were taken equal to 6.77×10^3 kpsi and 2.63 Å, respectively. The resulting values of K_1/K_2 are plotted versus annealing temperature in Fig. 4. The increase in K_1/K_2 with increasing annealing temperature is interpreted as an indication that oxygen migrates to the defect clusters during post-irradiation annealing and strengthens the defect clusters as barriers to dislocation motion. Thus, we conclude that the radiation-anneal hardening is due chiefly to the influence of interstitial impurity trapping in strengthening the defect clusters and not to changes in the density and size distribution of the clusters. This conclusion agrees in general terms with those of Shiraishi et al.[17] and Smidt[20], although there are some differences in detail. The conclusion also agrees with the earlier work on neutron-irradiated niobium[23,26], but it differs somewhat with the interpretation of Loomis and Gerber[27] who correlated the radiation hardening with defect clusters of sizes only below 70 Å.

The increase in the defect cluster density with increasing oxygen concentration (Fig. 1) upon irradiation at about 100°C is believed to be due to the formation during irradiation of small aggregates of oxygen atoms or oxygen-point defect complexes which serve as nucleation centers for the formation of the defect clusters. This is consistent with the results of Eto et al.[12], who observed a decrease in Snoek damping due to oxygen in vanadium upon irradiation at about 60°C to 2×10^{17} and 5×10^{19} neutrons/cm^2 (E>1 MeV). The increased stability against annealing with increasing oxygen concentration (Fig. 1) may be due to the trapping of oxygen at the defect clusters. As indicated in the Introduction, oxygen trapping may also be responsible for the $0.2T_m$ resistivity annealing stage and for the decrease in Snoek damping upon post-irradiation annealing. It also provides an explanation for the retardation of strain aging that has been observed in irradiated vanadium containing oxygen[27].

CONCLUSIONS

1. Upon increasing the oxygen concentration, the defect cluster density in neutron-irradiated vanadium increases, the average size decreases, and the stability against annealing increases.

2. Higher oxygen concentrations in vanadium also have the effect of increasing the radiation hardening and radiation-anneal hardening.

3. Based on a dispersed barrier model embracing defect clusters of all sizes, the radiation-anneal hardening is due principally to a strengthening of the defect clusters as barriers to slip dislocation motion, not to changes in the density and size distribution of the defect clusters.

4. The observations are consistent with the nucleation of defect clusters by small oxygen or oxygen-point defect complexes upon irradiation and the trapping of oxygen at defect clusters upon post-irradiation annealing.

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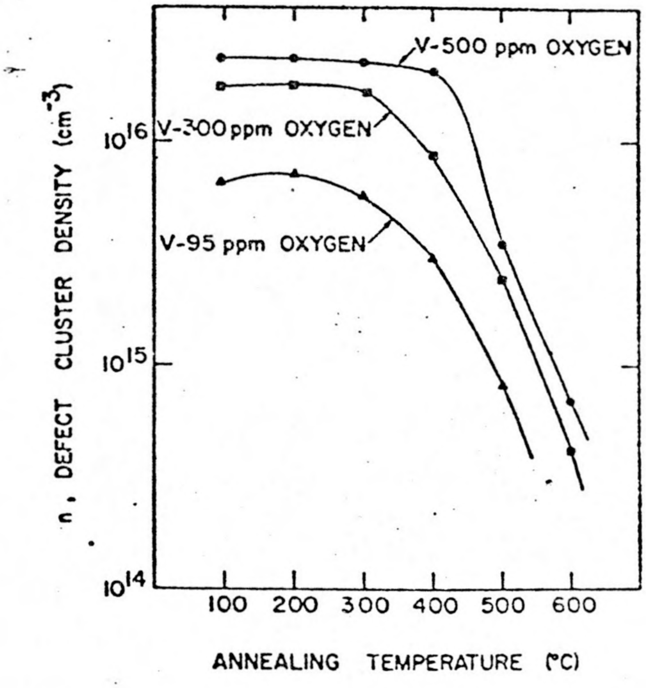

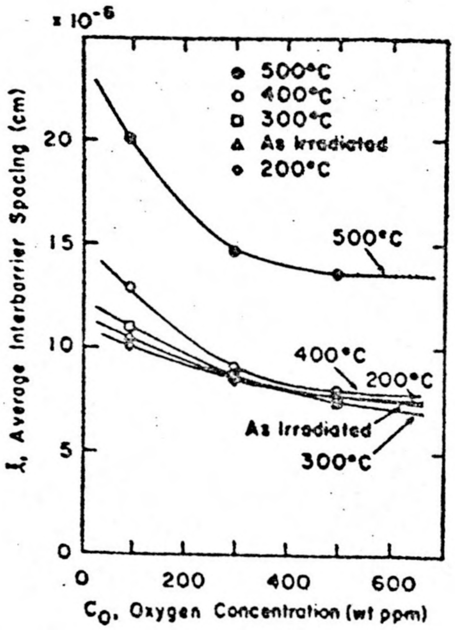

Fig. 1: Density of defect clusters versus post-irradiation annealing temperature. Irradiation dose and temperature, 1.4×10^{19} neutrons/cm^2 (E>1 MeV) at 95°C. One-hour anneals; as-irradiated plotted at 95°C.

Fig. 2: Average interbarrier distance versus oxygen concentration for as-irradiated and post-irradiation-annealed vanadium.

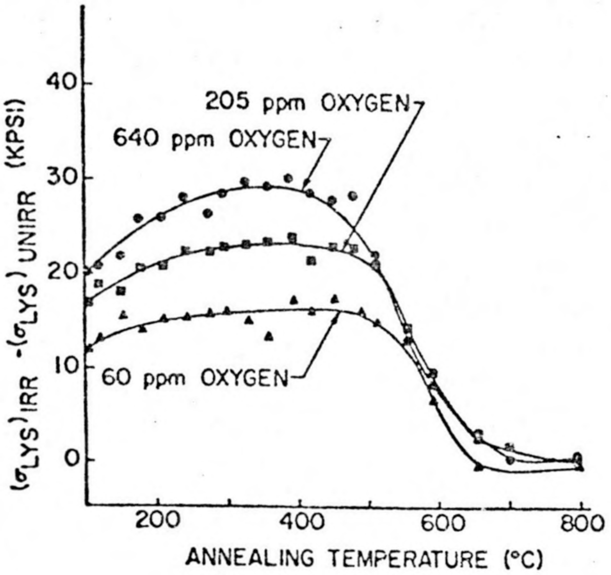

Fig. 3: Difference between lower yield stresses for irradiated and unirradiated vanadium containing 60, 205, and 640 wt ppm oxygen versus annealing temperature. Irradiation dose and temperature, 1.2×10^{19} neutrons/cm^2 (E>1 MeV) at 105°C. One-hour anneals. Room temperature tests.

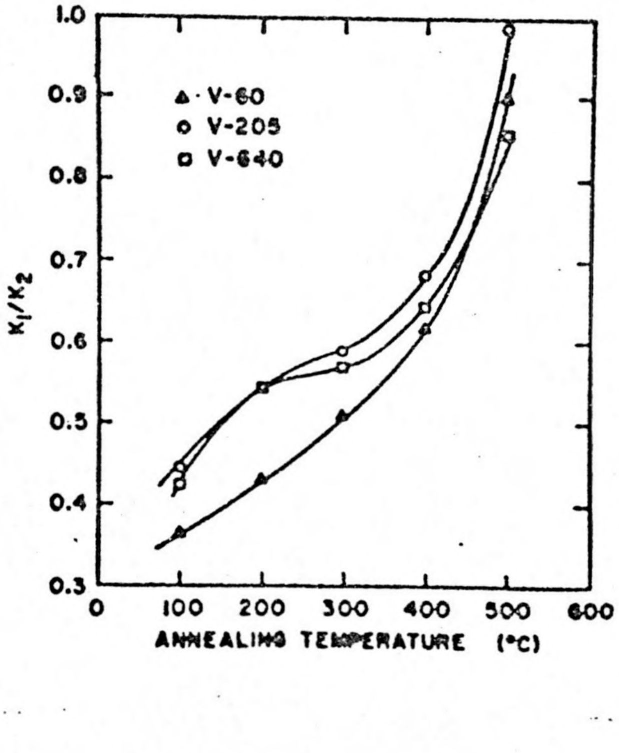

Fig. 4: K_1/K_2 versus post-irradiation annealing temperature for vanadium - 60, 205, and 640 wt ppm oxygen.