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THE TEMPERATURE DEPENDENCE OF THE DAMAGE MICROSTRUCTURES IN NEUTRON-IRRADIATED VANADIUM*

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L. L. HORTON AND K. FARRELL

Metals and Ceramics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831

Vanadium and vanadium with boron carbide additions (V-B_4C) were irradiated to ~1 dpa in the Oak Ridge Research Reactor at controlled temperatures ranging from 455 to 925 K. The V-B_4C alloy was enriched in ^{10}B, which produced ~3900 at. ppm helium. In the vanadium specimens, the dislocation microstructures varied from clusters of small (<50 nm diam) dislocation loops (455 to 625 K) to larger, homogeneously distributed loops at higher temperatures. Their Burgers vectors were a/2<111>. The V-B_4C specimens contained only tangled dislocation segments. Cavities were observed in all specimens. The cavity concentration decreased and the average diameter increased with increasing irradiation temperature. At 725 K, the maximum swelling was observed in both the vanadium (0.1%) and V-B_4C (1.4%). At comparable temperatures the cavities in the V-B_4C specimens were smaller and more numerous than those in the vanadium specimens. Helium bubbles were found on the grain boundaries in all of the V-B_4C specimens.

1. INTRODUCTION

A tokamak blanket design study by D. L. Smith et al.[1] selected vanadium-based alloys as the best material for structural applications with a lithium-cooled blanket. Previous studies have shown that vanadium-based alloys have low swelling after both neutron and ion irradiation. However, there is little information on the temperature dependence of neutron-irradiation damage or on the effects of the helium levels expected in a fusion reactor. (See the review of Gold and Harrod.)[2] For vanadium, the calculated ratio of helium concentration to displacement damage is ~5 at. ppm/dpa; in fission reactors, the ratio is <0.1 at. ppm/dpa.[3] One method of studying the effects of higher helium concentrations on the development of damage microstructures in fission reactors involves doping with the isotope ^{10}B which has a large cross section for the thermal neutron reaction $^{10}B + n \rightarrow {}^4He + {}^7Li$. A disadvantage of this technique is that the He/dpa ratio is not constant; most of the helium is produced early in the irradiation.

In this investigation, the irradiation temperature dependence of damage microstructures in neutron-irradiated pure vanadium and vanadium with boron carbide additions (V-B_4C) enriched in ^{10}B have been studied.

2. EXPERIMENTAL PROCEDURE

The vanadium specimens were fabricated from 2-pass zone refined stock (~100 wt ppm interstitial impurities). The V-B_4C specimens contained 760 wt ppm (~3900 at. ppm) ^{10}B and 745 wt ppm C; the concentration of the other interstitial impurities was <300 wt ppm. Disk specimens (3 mm diam × 0.5 mm thick) were heat-treated at ~1275 K in a vacuum at a pressure of ~1 mPa for 1 h for vanadium and 0.5 h for V-B_4C. The disks were irradiated in helium-filled, stainless steel capsules in the Oak Ridge Research Reactor (ORR). Flux monitors were located in 14 positions over the experiment to assure an accurate flux measurement for each capsule. The temperature of each capsule

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was monitored and controlled with individual heaters to within ±5° of the selected temperature. Additional details of the experiment can be found in Ref. 4. The irradiation parameters are listed in Table 1. The damage levels (dpa) were calculated from the measured fluences using the cross sections for the ORR spectrum provided by Gabriel et al.[3] The calculated helium levels were <0.1 at. ppm for vanadium and ~3900 at. ppm for $V\text{-}B_4C$. Microstructural examinations were done with JEM 100CX and Philips EM 400T/FEG analytical electron microscopes operated at 120 KeV.

3. RESULTS

3.1. Vanadium

The unirradiated vanadium specimens had a low dislocation density of $<10^{11} m^{-2}$. In the irradiated specimens the dislocations varied from clusters of small (<50 nm diam) dislocation loops for irradiations between 455 and 625 K to larger, homogeneously distributed loops and line segments at higher temperatures. The clusters of small loops were often associated with dislocation line segments. Analyses of the geometry of the loops formed at 625 K indicated Burgers vectors of a/2<111>. In general, all of the loops within individual clusters did not have the same specific Burgers vector. At 723 and 773 K the dislocation loops retained the a/2<111> Burgers vectors. More detailed analyses of the dislocation microstructures are in progress and will be the subject of a separate publication.

Cavities were observed in all specimens. Example micrographs are shown in Fig. 1. The cavities were equiaxed but did not have a distinct crystallographic shape. The distribution of cavities was fairly homogeneous for all irradiation temperatures except 775 K. At the lower irradiation temperatures, however, the cavity concentration may be lower near the loop clusters than elsewhere in the specimens. Despite high cavity concentrations at the lower irradiation temperatures, no cavity lattices were observed. As shown in Fig. 2, the volume-averaged cavity diameter increased and the cavity concentration decreased with increasing irradiation temperature. The quantitative data for the 445 K specimen plotted in Fig. 2 are approximate values since the specimen quality was not sufficient for accurate measurements. Maximum swelling of 0.1% was measured for an irradiation temperature of 725 K.

TABLE 1
Irradiation parameters for vanadium alloys in ORR-228

Material	Irradiation Temperature		Fluence (>0.1 MeV) (neutrons-m^{-2})	Damage level (dpa)
	(K)	(T/T_m)		
V	455	0.21	1.1×10^{25}	1.0
V	550	0.25	1.3×10^{25}	1.2
V	625	0.29	1.2×10^{25}	1.1
V,$V\text{-}B_4C$	725	0.33	1.3×10^{25}	1.2
V,$V\text{-}B_4C$	775	0.36	1.0×10^{25}	1.0
$V\text{-}B_4C$	825	0.38	1.2×10^{25}	1.1
$V\text{-}B_4C$	925	0.43	0.7×10^{25}	0.6

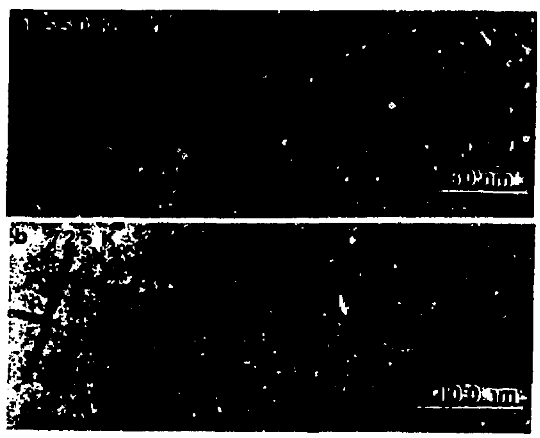

FIGURE 1
Examples of the cavity microstructures observed in vanadium. (a) 550 K, overfocused. (b) 725 K, underfocused. Note the platelet precipitates

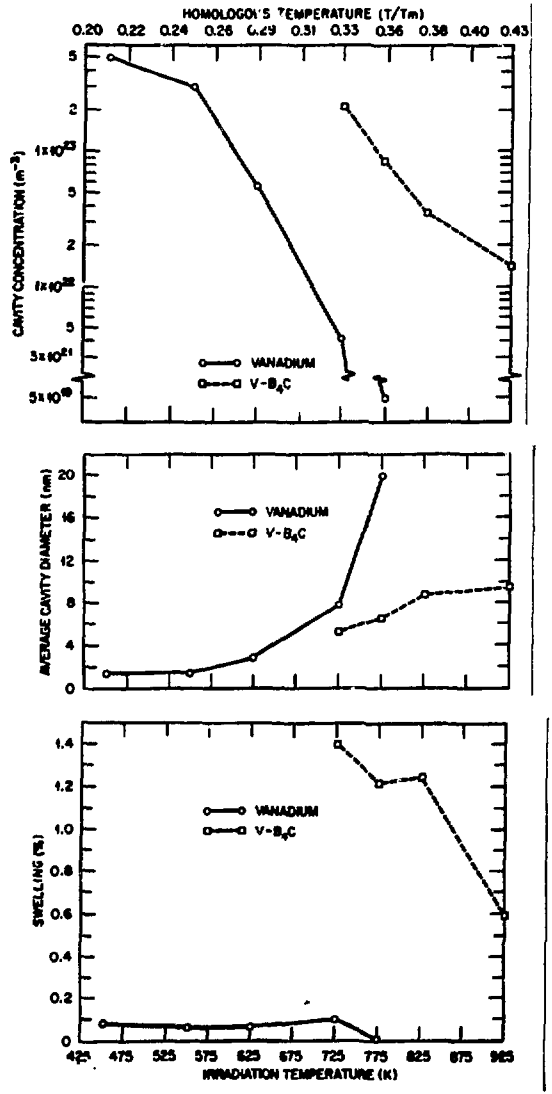

FIGURE 2
Quantitative cavity data for neutron-irradiated vanadium and $V\text{-}B_4C$

Irradiation-induced platelet precipitates were observed in the specimens irradiated at 725 and 775 K. These precipitates, seen as fringed areas in the micrograph of Fig. 1(b), lie on {012} with their long axis along <100>. Further analyses are planned to determine the composition of the precipitates.

3.2. Boron-doped vanadium

The microstructure of the unirradiated $V\text{-}B_4C$ specimens contained dislocation line segments (density $\sim 10^{12} m^{-2}$) and blocky precipitates presumed to be carbides. The precipitates were located both in the grains and along the grain boundaries. Irradiation produced tangled dislocation lines. The dislocation density decreased from $8 \times 10^{13} m^{-2}$ for irradiation at 725 K to $6 \times 10^{12} m^{-2}$ at 925 K.

Homogeneous distributions of cavities were produced at all irradiation temperatures. This distribution suggests that the ^{10}B was in solution rather than in the precipitates prior to irradiation. The cavities were faceted; the cavity morphology was a truncated dodecahedron with {110} faces and {100} truncations. The cavity parameters are plotted with the vanadium data in Fig. 2. The cavity concentration decreased and the cavity diameter increased with increasing irradiation temperature. At 725 K (Fig. 3) and 775 K, the cavities in the $V\text{-}B_4C$ are smaller and have over an order of magnitude higher concentration than the cavities in vanadium. For $V\text{-}B_4C$, the highest

FIGURE 3
Cavity microstructures observed in neutron-irradiated $V\text{-}B_4C$ at 725 K, overfocused

swelling of 1.4% occurred at the lowest irradiation temperature of 725 K. At all irradiation temperatures, higher concentrations of cavities were observed on grain boundaries than in the grains (Fig. 4). For example, at 925 K the areal density of cavities on the grain boundaries was about an order of magnitude higher than in the matrix. Denuded zones were observed near the boundaries. Enhanced cavity concentration was also observed within the precipitates. In general, these cavities were smaller than the cavities observed elsewhere in the specimens.

4. DISCUSSION AND CONCLUSIONS

The types of features observed in these damage microstructures agree with those reported by other investigators. The dislocation microstructures for the vanadium specimens are similar to those reported by Elen.[5] The dislocation Burgers vectors were found to be a/2<111> in both studies. Elen also reported planar precipitates similar to those observed here. These precipitates, believed to result from the interaction of vanadium self-interstitials and interstitial impurities, were first observed by Wiffen and Stiegler.[6] Subsequently, many investigations have reported such precipitation in neutron-irradiated vanadium, but the composition remains unknown.[7]

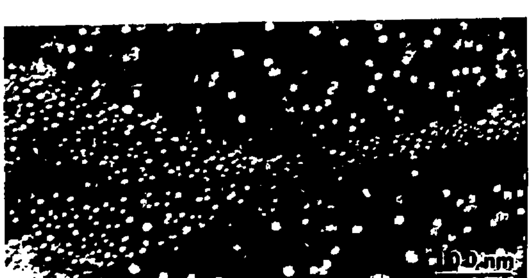

FIGURE 4
Cavities found on grain boundaries in $V\text{-}B_4C$, 925 K

Similarly, cavities are generally small and of high concentrations, and swelling is small.

However, the temperature-dependencies of the sizes and concentrations of cavities reported by various experimenters do not agree, even when allowances are made for different fluences. Table 2 compares the cavity parameters from the current study with earlier observations of neutron-irradiated vanadium. (The homologous temperatures are based on a melting temperature of 2173 K.) In all cases, the cavity diameter increases and the cavity concentration decreases with increasing irradiation temperature. In addition, with the exception of Adda,[7] all of the investigations report high concentrations ($>10^{21} m^{-3}$) of small (<10 nm diam) cavities for irradiation temperatures less than 0.36 T_m. There is insufficient data available to evaluate the reasons for the larger cavity diameters and lower cavity concentrations reported by Adda. Certainly, the lower purity of the material used by Adda would influence the swelling, but no information on the preirradiation microstructure was reported. The swelling values for all of the studies are quite low; the maximum swelling, 1.77%, was reported by Wiffen[8] for a fluence of 1.4×10^{26} neutron-m^{-2}. The low swelling in vanadium (and other refractory bcc metals) may result in part from the high concentrations of cavities promoting overall recombination of point defects by serving as the main point defect sink.

The irradiation temperature at which the maximum swelling occurred is different for each of the investigations listed in Table 2. These differences may result, in part, from the flux differences in the various reactors and the different purities of the vanadium. The peak swelling temperature may also vary with fluence. However, in most neutron irradiation experiments, the temperature of individual specimens of capsules are not directly

TABLE 2
Vanadium swelling data

Investigator/ Material Purity	Fluence (>0.1 MeV) (10^{24} neutrons-m^{-2})	Irradiation Temperature			Cavity Parameters		
		(°C)	(K)	(Homologous)	Concentration (m^{-3})	Diam (nm)	Swelling (%)
Elen[5]	2.2	420	693	0.32	6×10^{21}	3.5	0.01
170 wt ppm[a]	3.7	630	903	0.42	1×10^{21}	7.0	0.02
	2.7	750	1023	0.47	1×10^{20}	11.0	0.007
Adda[7]	5.0	335	608	0.28	3.4×10^{20}	14.0	0.05
99.5% V	5.0	435	708	0.33	2.0×10^{20}	28.0	0.22
	5.0	600	873	0.40	1.5×10^{19}	78.0	0.38
	5.0	800	1073	0.49	—	—	—
Horton & Farrell	11.0	182	455	0.21	$\sim 5 \times 10^{23}$	<1.5	—
<100 wt ppm[a]	13.0	275	550	0.25	3.0×10^{23}	1.5	0.06
	12.0	350	625	0.29	5.6×10^{22}	2.8	0.07
	13.0	450	725	0.33	4.1×10^{21}	7.8	0.1
	10.0	500	775	0.36	$< 5 \times 10^{19}$	20.0	<0.02
Wiffen[8]							
220 wt ppm[b]	49.0	385	658	0.30	1.2×10^{22}	5.0	0.08
1250 wt ppm[b]	57.0	385	658	0.30	1.2×10^{23}	4.5	0.56
1000 wt ppm[a]	{ 97.0	475	748	0.34	1.1×10^{22}	8.2	0.34
	140.0	550	823	0.38	5.9×10^{21}	17.0	1.77
	140.0 }	600	875	0.40	2.0×10^{21}	23.0	1.47
1800 wt ppm[a]	25.0	710	933	0.43	—	—	—
Carlander et al.[9]	110.0	465	738	0.34	1.6×10^{22}	10.0	1.04
"Commercial	110.0	525	798	0.37	$1.2\text{–}8 \times 10^{21}$	10–17	0.3–1.1
Purity"	250.0	625	898	0.41	9.0×10^{21}	14.0	1.54
Bartlett et al.[10]	{360.0	450	723	0.33	1.35×10^{22}	6.0	0.2
6-pass zone	360.0	550	823	0.38	1.25×10^{21}	21.3	0.6
refined	360.0}	600	873	0.40	9.40×10^{20}	25.8	0.8
"Commercial	{360.0	450	723	0.33	1.06×10^{22}	11.0	0.7
Purity"	360.0	550	823	0.38	1.60×10^{21}	22.4	0.9
	360.0}	600	873	0.40	3.20×10^{20}	28.1	0.4

[a] Interstitial Impurities.

[b] C + N + O Impurities.

measured; they are estimated or extrapolated. Also, as indicated in Table 2, there is usually a large variation in fluence with irradiation temperature within a given experiment. This fluence variation combined with the uncertainty in irradiation temperatures makes determination of the actual peak swelling temperature difficult. In the present work the irradiation temperatures were measured and controlled continuously, and the fluence varied by only 20% for the total specimen set. Thus, the swelling values can be compared directly to determine that the peak swelling temperature is near 725 K.

Helium is known to enhance the nucleation of cavities and to extend bias-driven swelling to higher temperatures.[11] It is not surprising, then, that the V-B_4C specimens contained considerably higher concentrations of cavities than the vanadium at comparable temperatures and that the cavities in V-B_4C persisted to temperatures of 925 K (0.43 T_m). The progressive decline of swelling with increasing irradiation temperature (Fig. 2) indicates a bias-driven mode of swelling rather than one driven wholly by gas pressure. However, there is almost enough helium for the cavities to be equilibrium bubbles, especially at the higher irradiation temperatures. The level of swelling in $V-B_4C$ is higher than in vanadium because of the higher cavity density, and because the internal helium pressure reduces thermal vacancy emission. At grain boundaries, where bias-driven cavities cannot develop, the observed cavities must be equilibrium bubbles.

This comparison of damage microstructures and swelling levels in vanadium and $V-B_4C$ does not allow a proper assessment of the effects of intermediate He/dpa ratios such as those expected for fusion reactors. It simply confirms that helium stimulates cavity formation and forms bubbles at grain boundaries. Studies with lower and controlled rates of helium generation are needed for definitive conclusions.

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