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Effects of temperature change on vanadium alloys irradiated in HFIR

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Abstract

To understand the effects of temperature change on pure vanadium and four vanadium model alloys (V–5Cr, V–5Ti, V–4Cr–4Ti, V–4Cr–4Ti–0.1Si), neutron irradiation was performed in high flux isotope reactor. The specimens were irradiated for a total of 8 irradiation cycles, which resulted in 4 dpa. Each irradiation cycle consisted 0.05 dpa at 498 K, followed by 0.45 dpa at 613 K. After the irradiation the TEM samples were electro-polished and examined by electron microscopy. The microstructures of the samples were compared with those of the same alloys, which were continuously irradiated at 613 K.

1. Introduction

It is well known that effects of varying temperature irradiation on microstructure are very significant in many pure metals and also in structural materials designed for fusion reactors [1–5]. Vanadium alloys exhibit important advantages as a candidate material for this reactor. Fission neutron irradiation using Japan Materials Testing Reactor (JMTR) [6] and our previous ion irradiation studies on vanadium alloys [7,8] showed that, microstructure at an elevated temperature was very sensitive to the pre-irradiation temperature and dose. Moreover, in V–4Cr–4Ti alloy, formation of Ti-oxide at 873 K is enhanced by pre-irradiation at 473 K, which resulted in hardening of the damage region [8]. The previous fission neutron irradiation using JMTR and the other ion irradiation studies were mainly focused on microstructural evolution to relatively low ($\simeq$1 dpa) dose irradiation. To understand effects of temperature change at higher dose levels, the varying temperature irradiation experiment using high flux isotope reactor (HFIR) was selected as one of the major tasks in the Japan–USA Fusion Cooperation Program (JUPITER Project) [9]. In the HFIR-13J varying temperature experiment, four irradiation zones were set, low-temperature (613 K) constant, high-temperature (793 K) constant, high-temperature variable (613/793 K) and low-temperature variable (498/613 K). In the variable zones, the temperatures were reduced to the low- temperature condition for the initial 10% period of each irradiation cycle, and elevated for the remaining 90% period. The present paper summarizes some key findings of pure vanadium and vanadium alloys under the lower temperature constant (613 K) and the low-temperature variable (498/613 K) conditions.

2. Experimental procedure

Pure vanadium and four model vanadium alloys (namely, V–5Cr, V–5Ti, V–4Cr–4Ti, V–4Cr–4Ti–0.1Si) were used in this study. In the vanadium alloys, the oxygen content varied over the range of 13–1889 appm. The detailed specimen composition is reported in Ref. [10]. Disk specimens for electron microscopy were wrapped with pure zirconium as a getter of oxygen and annealed 2 h at 1373 K prior to irradiation.

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The irradiation was conducted for a total of eight cycles in europium shielded RB* position of HFIR, which resulted in a fast fluence ($E > 0.1$ MeV) of 8×10^{21} n/m^2. This corresponds to 4 dpa in stainless steels. The experimental design, fabrication of the capsule and the irradiation temperature history of each cycle have been reported in Refs. [11,12].

3. Results

3.1. Pure vanadium

The microstructure of pure vanadium at constant temperature irradiation (613 K) and varying temperature irradiation (498/613 K) were compared. The comparison is shown in Figs. 1 and 2. In Fig. 1, two weak

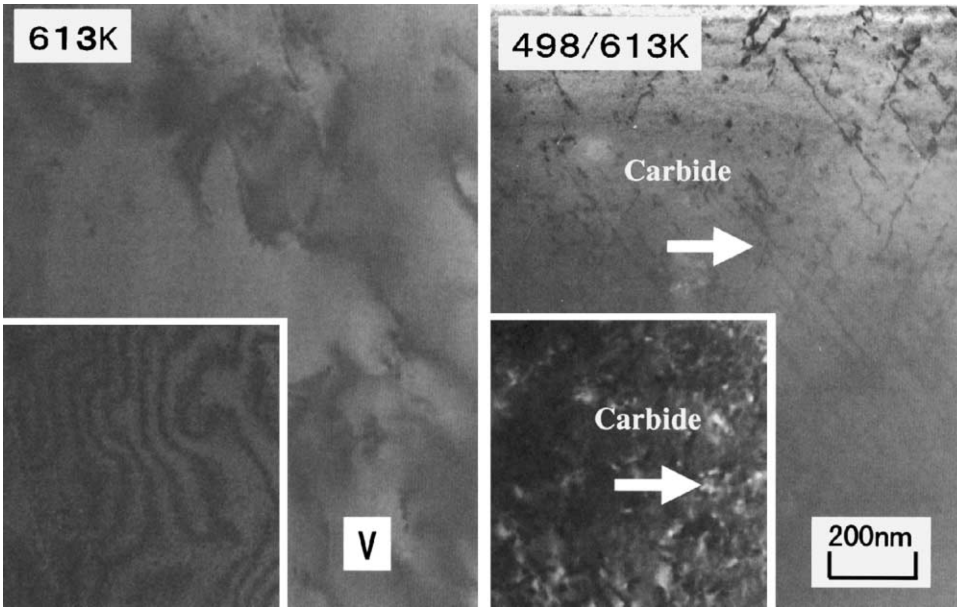

Fig. 1. Dislocation structures observed in pure vanadium at 613 and 498/613 K.

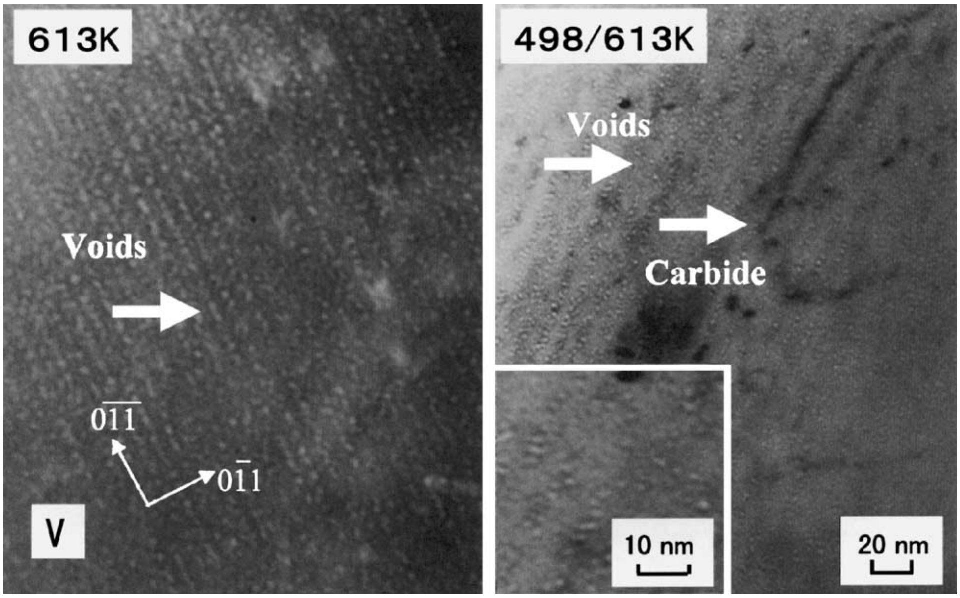

Fig. 2. Voids observed in pure vanadium at 613 and 498/613 K.

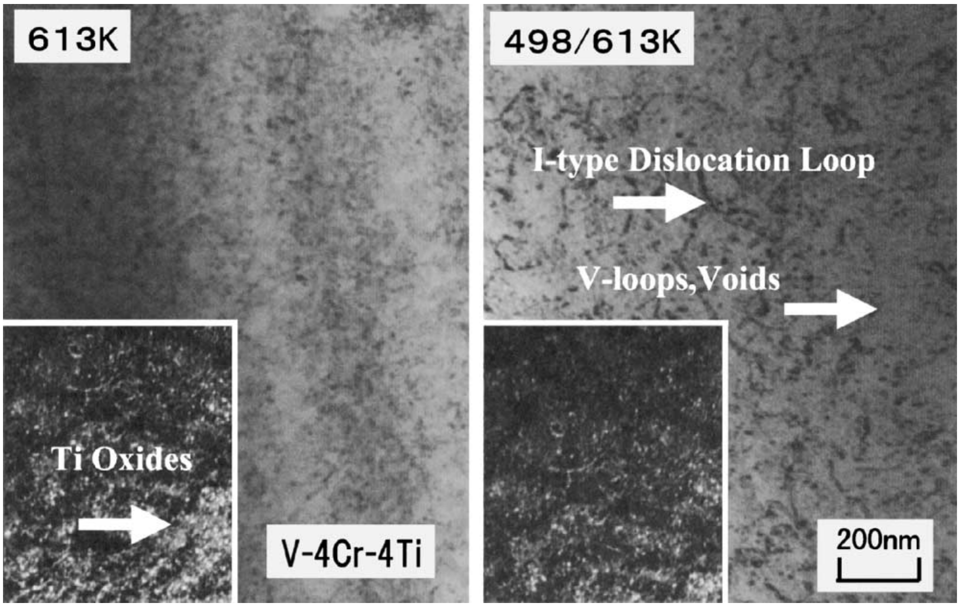

Fig. 3. Dislocation structures observed in V–4Cr–4Ti at 613 and 498/613 K.

beam images of the microstructure were inserted in the left corner of each photos. Fig. 2 shows the corresponding void contrast image at the same irradiation conditions. At the constant temperature irradiation, the dislocation density was very low of 3×10^{13} m^{-2}. As shown in Fig. 2, ordered array of voids have been observed at constant temperature irradiation. The ordered array of voids has a bcc lattice that is parallel to the host vanadium lattice. The average diameter and density of voids were 2.8 nm and 1.1×10^{24} m^{-3}, respectively. However, under varying temperature control, a high density of thin, long platelet shaped precipitates (shown by arrows in Fig. 1) was observed. They are oriented in $\langle 1\,0\,0 \rangle$ directions. Small voids of about 1.8 nm and a density of 1.4×10^{24} m^{-3} were randomly distributed in this sample.

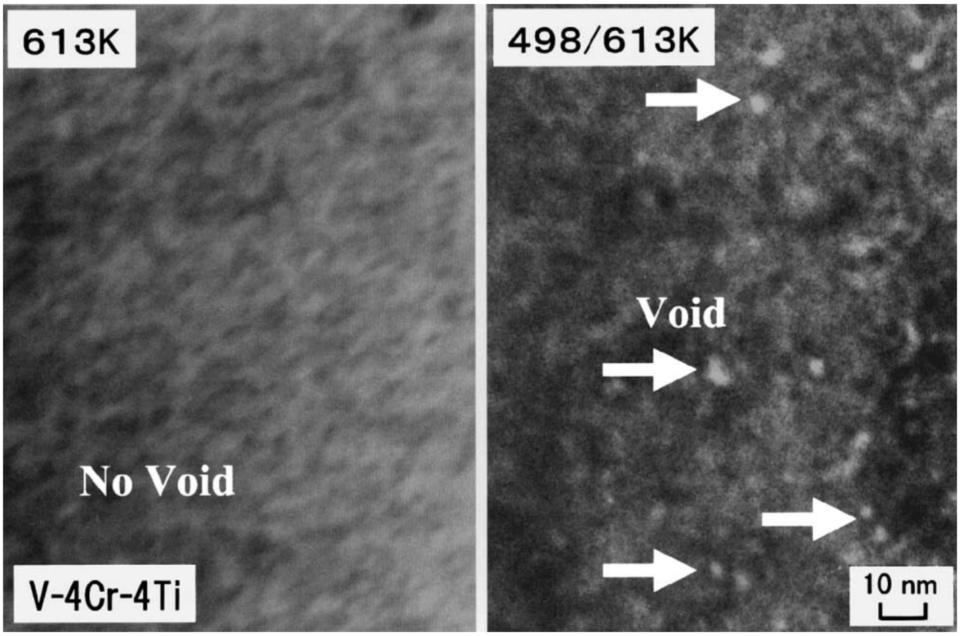

Fig. 4. Voids observed in V–4Cr–4Ti at 613 and 498/613 K.

3.2. Vanadium alloys (V–5Cr, V–5Ti, V–4Cr–4Ti, V–4Cr–4Ti–0.1Si alloys)

In vanadium alloys, the effects of low-temperature irradiation were relatively small, except for small void formation in the V–4Cr–4Ti alloy. In V–5Cr, the constant and varying temperature irradiation produced tangled dislocation structures, small vacancy type clusters (namely, loops and voids). Voids were randomly distributed in both cases. The measured size and density were 2.5 nm and 5.0×10^{23} m^{-3} at constant irradiation, respectively. At varying temperature irradiation, they were 3.0 nm and 5.8×10^{23} m^{-3}, respectively. Besides tangled dislocations, a very high density of precipitates was also formed in titanium containing alloys (V–5Ti, V–4Ti–4Cr, V–4Cr–4Ti–0.1Si). The microstructure of V–4Cr–4Ti at constant temperature irradiation (613 K) and varying temperature irradiation (498/613 K) were compared and are shown in Fig. 3. From our previous ion irradiation, fine precipitates (shown by arrows in an inserted weak beam image) were identified to be titanium oxides (TiO_2). Fig. 4 shows the corresponding void contrast image. Small voids were only observed in the samples irradiated in the varying temperature condition.

4. Discussions

In this study, carbide formation was observed in pure vanadium irradiated in the varying temperature condition (498/613 K). The enhanced formation of carbide in the varying temperature condition was also confirmed by copper ion irradiation. Fig. 5 provides comparison of the microstructure in the constant temperature irradiation (873 K) and varying temperature irradiation (873/473/873 K). Upper and lower photos show the dislocation contrast image and corresponding void contrast images, respectively. Irradiation details (irradiation temperature and doses) are given in the diagram below. In the copper ion irradiation experiments on pure vanadium, carbide formation was detected in the temperature range of 773–873 K but they were dissolved after very low-dose irradiation at lower temperatures [7]. As shown in the Fig. 5, the carbides were very sensitive to temperature variation and they dissolved at successive irradiation at 473 K (see (a) and (b) in the figure). But dissolution of carbide results in additional formation at the higher dose level. As shown in (d) and (e) in Fig. 5, the carbide density at constant irradiation at 873 K was about ten times lower than that of varying temperature

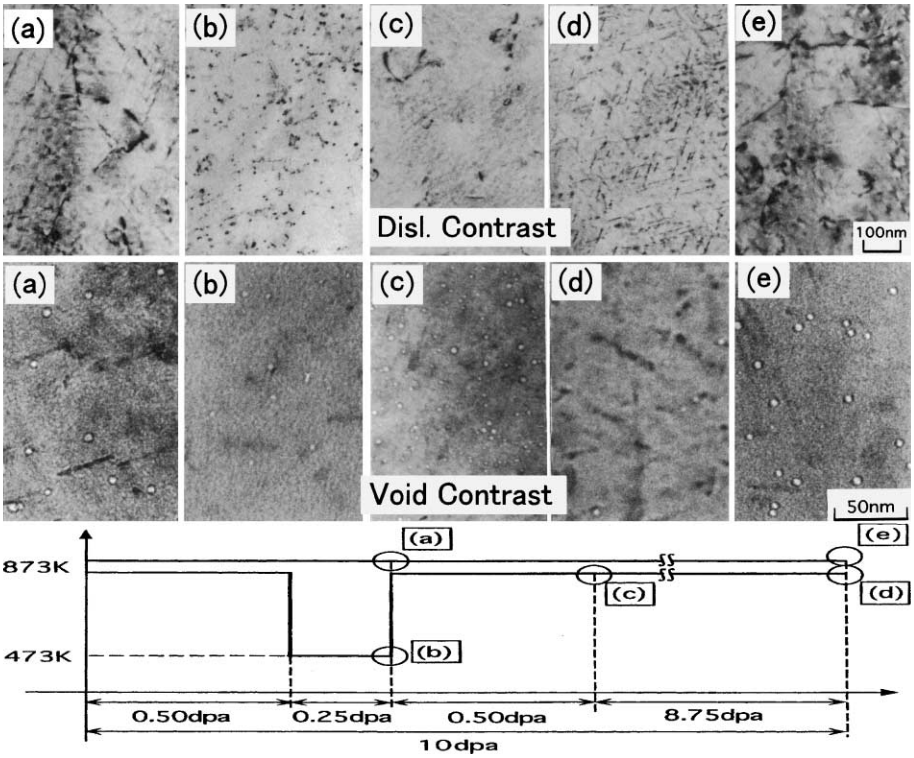

Fig. 5. Microstructure of copper ion irradiated pure vanadium at constant temperature (873 K) and varying temperature (473/873 K) up to a dose of 10 dpa. Irradiation details are given in the diagram below.

conditions to a dose of 10 dpa. And growth of voids was not prominent in the conditions where these carbides were observed. This means that void swelling resistance of pure vanadium will be affected by the dissolution and nucleation of carbide due to temperature variation.

Ordered void arrays were formed in pure vanadium at constant irradiation. The void arrangement in an ordered array is commonly observed in oxygen doped refractory metals (i.e. Nb [13]). In niobium, for example, the ordered void lattices are formed, if the oxygen–impurity concentration exceeds a threshold amount. The threshold concentration is between 60 and 400 appm for pure niobium. But the oxygen concentration of pure vanadium used in this study is only 13 appm. The concentration must therefore be below the threshold amount. The formation of void lattices may be due to unexpected oxygen diffusion from the irradiation environment. The detailed mechanism of enhanced carbide formation due to temperature variation was not understood. But it is well established that radiation induced precipitation is controlled by the migration of solute-point defect (vacancy and/or interstitial) complexes under irradiation. Therefore, enhanced carbide formation in pure vanadium may also be related to very high concentrations of vacancies and/or interstitials caused by the temperature variation.

In the present study, the effects of low-temperature irradiation on vanadium alloys were relatively small, except for the carbide formation in pure vanadium and small void formation in V–4Cr–4Ti alloy. Our previous studies reported that, when variations in temperature crossed a characteristic borderline temperature of microstructure evolution, the pre-irradiation at lower temperature is very efficient for suppression of interstitial loop formation. The results were explained by a vacancy rich condition, which appears temporarily at the beginning of the high-temperature irradiation [2–5]. Our previous study also showed that this temperature is about 673 K for pure vanadium [7]. In the case of V–4Cr–4Ti, vacancy clusters were very stable up to 773 K. In the present study, temperature variation did not cross these borderline temperatures (673 K for pure vanadium and 773 K for V–4Cr–4Ti alloy). But varying temperature irradiation produced random voids distribution in pure vanadium and fine voids formation in V–4Cr–4Ti. We believe that the void formation was close related with radiation induced precipitation (carbides and titanium oxides). Therefore, further analysis is needed to understand the void formation related with precipitation under varying temperature conditions.

5. Conclusions

Varying temperature (498/613 K) introduced carbide in pure vanadium. However, the effects of low-temperature irradiation on vanadium alloys were relatively small, except for the alloy V–4Cr–4Ti. In pure vanadium and V–5Cr alloy, small voids of about 3 nm diameter and tangled dislocation structures were observed in both irradiation conditions. In V–4Cr–4Ti alloy, on the other hand, a high density of small voids was only detected in the varying temperature condition 498/613 K.

Acknowledgements

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