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Microstructural evolution and hardening of neutron irradiated vanadium alloys at low temperatures in Japan Material Testing Reactor

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Abstract

In order to clarify the mechanism of the large increment of DBTT due to low temperature neutron irradiation, the mechanical properties and the microstructural evolution were investigated in vanadium binary alloys irradiated in JMTR. From a series of microhardness tests conducted on these alloys after irradiation at 90–200°C, the hardness increased with increasing irradiation temperature, except for V–5Ti and V–5Nb. However, the hardening decreased with increasing irradiation temperature from 350°C to 400°C. The dislocation loops were observed by TEM after irradiation from 90°C to 200°C, while no void was observed at these irradiation temperatures. Positron annihilation spectroscopy (PAS) exhibited the nucleation of vacancy clusters in undersized binary alloys. The dislocation loop density increased with increasing irradiation temperature. It is considered that the irradiation hardening is mainly caused by dislocation loops. It has also been suggested that interstitial oxygen promote dislocation loop nucleation since interstitial oxygen and solute atoms interacted strongly and bound at around 200°C leading to enhanced nucleation of interstitial loops. © 1999 Published by Elsevier Science B.V. All rights reserved.

1. Introduction

Vanadium alloys have been shown to have very low ductile–brittle transition temperature (DBTT). Especially, the DBTT of blunt-notched V–4Cr–4Ti alloys, was below liquid nitrogen temperature (L-N-T) even after neutron irradiation with high damage level [1]. However, it has been reported recently that V–4Cr–4Ti irradiated at 100–200°C in HFBR has the DBTT shifted from L-N-T to 100°C [2]. In the present paper, the mechanical properties and the microstructural evolution has been investigated in neutron irradiated vanadium binary alloys up to 400°C in JMTR, in order to clarify the mechanism of the large increment of DBTT due to neutron irradiation.

2. Experimental

The alloys prepared are V–5 at.% Fe, V–1 at.% Si, V–5 at.% Cr, V–5 at.% Mo, V–5 at.% Ti and V–5 at.% Nb, and pure vanadium. These alloys have been selected to cover a wide range of atomic size factor of solutes in vanadium. The starting material was 99.9% pure dendritic vanadium and was electron beam melted for further purification. The purity levels of the materials for alloying ranged from 99.99% to 99.999%. The alloy ingots were obtained by arc melting in argon. These ingots were cold rolled to approximately 0.25 mm thick sheets. TEM disks were punched from sheet specimens and were doubly wrapped with tantalum and zirconium foils and sealed in evacuated quartz tubes. The recrystallization annealing was done at 1100°C for 2 h. The concentration of the interstitial impurities after the anneals was determined by chemical analysis; oxygen, carbon, and nitrogen concentration was about 0.1, 0.02, 0.001 at.%, respectively.

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Neutron irradiation was conducted in Japan Materials Testing Reactor (JMTR) at 90°C, 150°C with damage level of 0.03 dpa (neutron flux: 2.9×10^{19} n/cm^2, $E > 0.1$ MeV); and 200°C, 350°C, 400°C with 0.13 dpa (neutron flux: 1.2×10^{20} n/cm^2, $E > 0.1$ MeV) using temperature control with electric heaters. Specimens were encapsulated and sealed in a vacuum by electron beam welding. Transmission electron microscopy (TEM), Micro-Vickers hardness test, and positron annihilation spectroscopy (PAS) were conducted on disk specimens irradiated at 90°C, 150°C, and 200°C.

3. Results

3.1. Microhardness test

Fig. 1 shows the hardness changes of each alloy as a function of the atomic size factor of solute. From a series of microhardness tests conducted on these alloys after irradiation between 90°C and 200°C, the hardness decreased with increasing irradiation temperature for V–5Nb, a behavior typically observed in many metals. However, the hardness did not change for V–5Ti. Furthermore, the alloys with smaller relative atomic size than V–5Mo have shown an inverse irradiation temperature dependence; i.e. radiation induced hardening is greater for higher irradiation temperature. With increasing irradiation temperature from 200°C to 350°C, the data indicate a significant increase in hardness of all vanadium alloys. At a higher temperature (400°C), however, the hardness decreased for all vanadium alloys. It should be noted that irradiation fluence at temperature 200°C, 350°C, and 400°C was about four times larger than at 90°C and 150°C.

3.2. Positron lifetime measurements

The positron lifetime spectra were analysed with the programs 'Positronfit' and 'Resolution' in terms of one or two exponential components to obtain the best fit after subtracting background and the source contribution to the spectrum. Fig. 2 depicts the results of positron lifetime measurements on vanadium alloys neutron irradiated at temperature 90°C, 150°C and 200°C. The lifetime spectra obtained were decomposed into two components. The shorter lifetime component (τ_1) indicates matrix, single vacancy or dislocation component, and the longer lifetime component (τ_2) indicates vacancy clusters component. These clusters consist of a small number of vacancies, i.e. ten or few tens of vacancies [3], which are unobservable by TEM. The intensity of longer life time component (I_2) is also shown in this figure. The presence of vacancy clusters due to irradiation on the alloys with smaller solute atomic size than V–5Mo is clearly indicated in this figure. As seen here, an increase of τ_2 with irradiation temperature, especially from 90°C to 150°C means the increasing size of vacancy clusters. This tendency is usually observed in neutron irradiated specimens at higher temperature. The absence of vacancy clusters in V–5Ti and V–5Nb is also evident from these measurements. Since Ti and Nb have greater atomic size than vanadium, vacancies are trapped by these solute atoms so that migration of these vacancies are effectively suppressed [4].

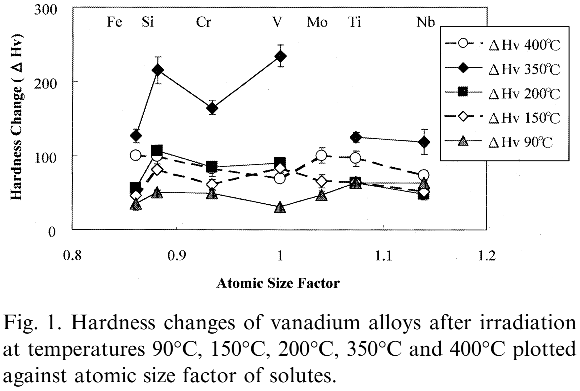

Fig. 1. Hardness changes of vanadium alloys after irradiation at temperatures 90°C, 150°C, 200°C, 350°C and 400°C plotted against atomic size factor of solutes.

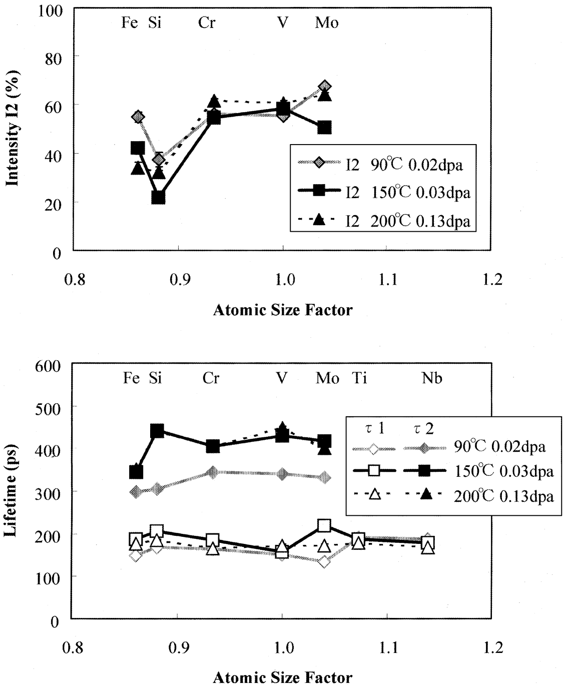

Fig. 2. Positron annihilation parameters of vanadium alloys after irradiation at 90°C, 150°C and 200°C versus atomic size factor of solutes. I_2 and τ_2 are the intensity and the lifetime of the second spectral component, and τ_1 is the shorter lifetime component.

3.3. Microstructural observations

Figs. 3 and 4 show the dislocation loops observed by TEM after irradiation at 90°C, 150°C and 200°C, respectively. Voids were not observed at these irradiation temperatures. The micrographs are arranged from left to right with increasing atomic size factor of the alloy. Dislocation loop density and average radius are determined from these figures and are plotted against the relative atomic size factor in Fig. 5. As seen in Fig. 5, the loop size in pure vanadium is bigger than in other alloys, and the loop size of all specimens somewhat

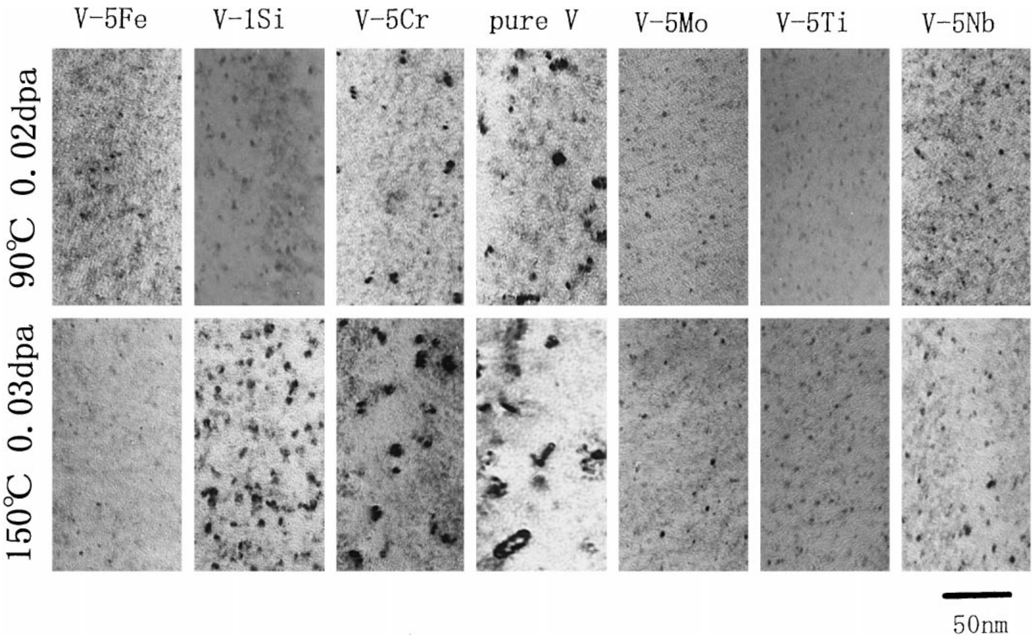

Fig. 3. Dislocation microstructures of vanadium alloys after irradiation in JMTR at 90°C and 150°C with 0.03 dpa.

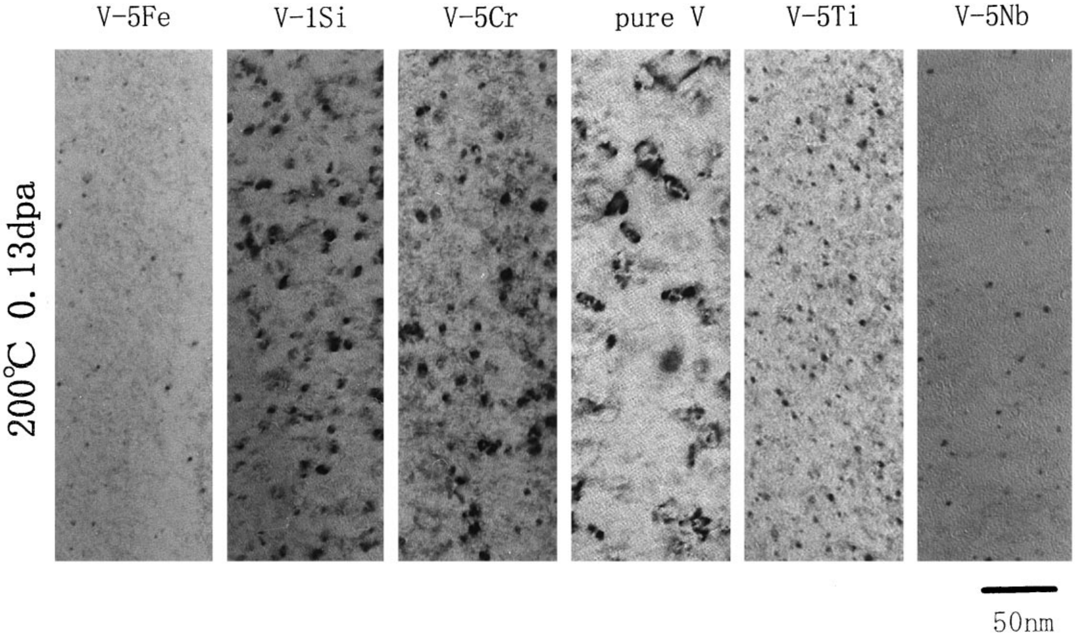

Fig. 4. Dislocation microstructures of vanadium alloys after irradiation in JMTR at 200°C with 0.13 dpa.

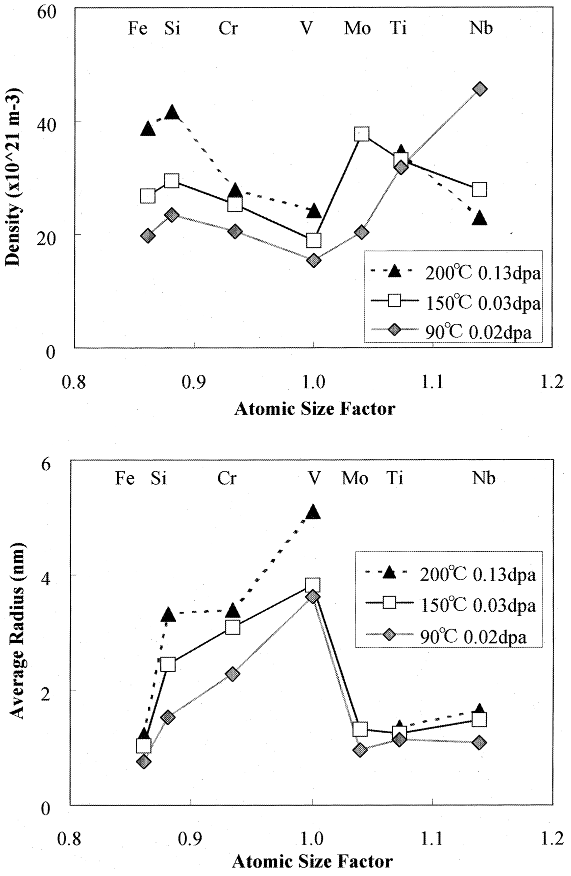

Fig. 5. Dislocation loop density and size determined from electron micrographs as shown in Figs. 3 and 4 plotted as a function of atomic size factor.

increased with increasing irradiation temperature. After irradiation between 90°C and 200°C, the loop density decreased with increasing irradiation temperature for V–5Nb. However, it did not change for V–5Ti. Moreover, the smaller atomic size alloys than V–5Mo have shown an inverse irradiation temperature dependence; i.e. the loop density is greater for higher irradiation temperatures.

A tendency for growth of dislocation loops to form dislocation networks was observed in the alloys after irradiation at temperatures 350°C and 400°C. Only dislocation loops, however, were observed in pure vanadium irradiated at 350°C. Furthermore, the dislocation density decreased with increasing irradiation temperature from 350°C to 400°C, except for V–5Ti.

It should be noted that the dislocation microstructures of V–5Ti are obviously decorated with some precipitates. Dislocation density has been measured from micrographs taken with $g = 110$. Thus, one half of the dislocations are out of contrast for Burgers vector $b = a/2\langle 1\ 1\ 1\rangle$, and one third are out of contrast for $b = a/2\langle 1\ 0\ 0\rangle$.

Fig. 6 shows the cavity images of all specimens after irradiation at 350°C and 400°C. No voids were observed in oversized vanadium alloys, i.e. V–5Ti and V–5Nb. This tendency agrees with the results of positron lifetime measurements described in the previous section. Furthermore, the void size becomes larger in the alloys with smaller atomic size factor. Thus, there is a clear tendency that alloys with strongly negative atomic size factors enhance cavity formation or its growth and vice versa [5–7]. Voids were also not observed in irradiated pure vanadium at 350°C. As seen in Fig. 6, the void size became larger while void density decreased with increasing irradiation temperature.

4. Discussion

Let us first consider the absence of vacancy clusters in V–5Ti and V–5Nb after irradiation at 90°C, 150°C and 200°C, as shown by the result of PAS. It is suggested that reduction of vacancy migration in neutron irradiated V–5Ti and V–5Nb at low temperature is due to trapping of vacancies by solute atom, since Ti and Nb have bigger atomic size than vanadium. From the result of microstructural observation by TEM, nucleation of voids was remarkable for irradiation between 90°C and 200°C. On the other hand, bigger voids observed after irradiation at 400°C with low density has provided the evidence that the void growth became dominant at this irradiation temperature.

In the present investigation, the dislocation loop density of V–5Fe, V–1Si, V–5Cr, pure V, and V–5Mo increased with increasing irradiation temperature from 90°C to 200°C. The result of internal friction measurement on neutron irradiated pure vanadium at temperature 90°C obtained from a parallel investigation [8] indicates that the oxygen impurities were trapped by irradiation induced defect clusters at low irradiation temperature such as 90°C. However, they were released and appeared in the matrix as free oxygen by increasing the annealing temperature above 200°C. It is considered that the dislocation loop nucleation was promoted by decreasing mobility of self-interstitial atoms, since the free oxygen in the matrix increased with increasing irradiation temperature. Thus, the dislocation loop density increases with increasing irradiation temperature.

Even though the irradiation fluence at temperature 200°C was about four times larger than at 90°C and 150°C, the increase of radiation induced hardening shows a good correlation with the increase of dislocation loop density. Thus the difference of irradiation fluence in this case does not affect the conclusion.

From the argument presented above we suggest that the increase of irradiation hardening is caused by the increase of dislocation loop density. In order to con-

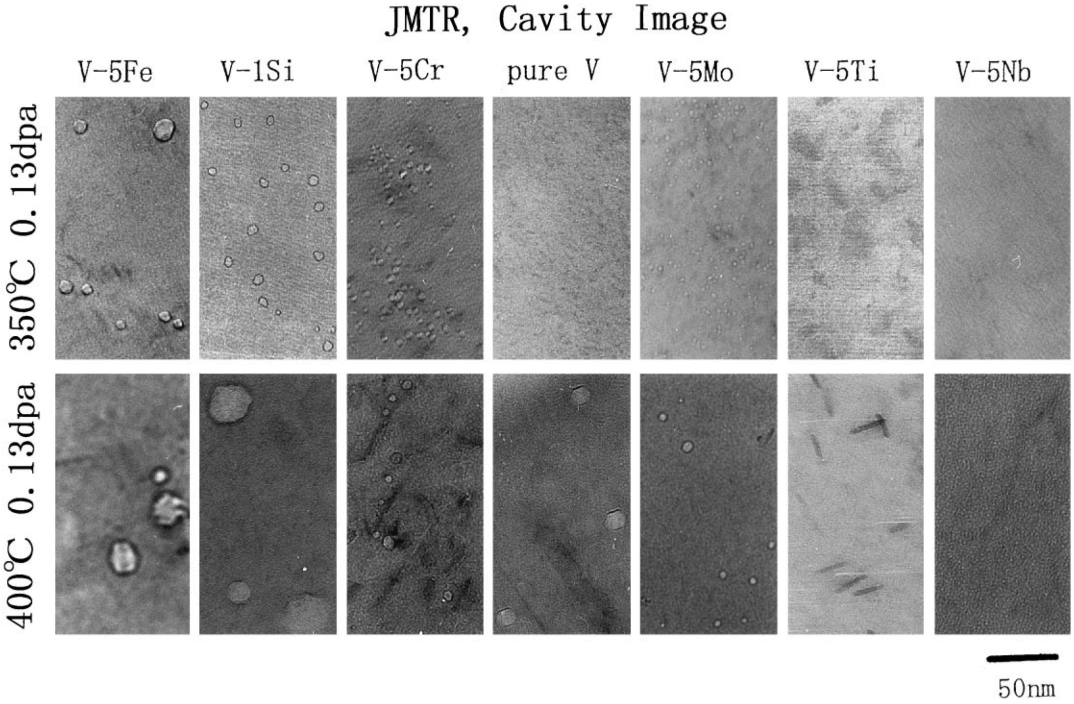

Fig. 6. Cavity microstructures of vanadium alloys after irradiation in JMTR at 350°C and 400°C with 0.13 dpa.

firm this suggestion, the yield stress increase has been calculated from the dislocation loop density and size based on the dispersed barrier hardening model. The calculated stress has been compared to the experimental values from microhardness tests [9]. A good agreement is obtained between the yield stress increase from hardness test and calculated values from dislocation loops. After irradiation at around 400°C, the coarsening of dislocation loops and growth of cavities in all vanadium alloys occur and are considered to cause the decrease in hardening as compared with 350°C irradiation.

5. Conclusions

After neutron irradiation at 90°C to 200°C, pure vanadium and all vanadium alloys except for V–5Ti and V–5Nb, the dislocation loop density and radiation induced hardening both increase with increasing irradiation temperature. It is considered that the irradiation hardening is mainly caused by dislocation loops. It has been also suggested that the increasing mobile oxygen in the matrix at intermediate irradiation temperatures (150–350°C) promoted dislocation loop nucleation.

Consequently, it is deduced that a low oxygen concentration would produce a reduction of radiation hardening and embrittlement of vanadium alloys after neutron irradiation at low temperatures. The large radiation-induced increase in DBTT is diminished in vanadium alloys at irradiation temperature above 400°C due to coarsening of dislocation loops and cavities which reduces the amount of radiation hardening.

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