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Effects of temperature change on the microstructural evolution of vanadium alloys under ion irradiation

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Abstract

The evolution of microstructure in vanadium alloys after upward and downward temperature changes has been closely examined. Vanadium alloys have been irradiated in the high fluence irradiation facility at the University of Tokyo (HIT). Irradiations have been performed either at constant temperature of 500°C or in a stepwise temperature sequence of either 350/500°C, 400/500°C, 450/500°C, 500/350°C or 350/500/350°C up to 0.5 or 0.75 dpa. After 350/500°C and 400/500°C temperature change irradiations, small dislocation loops have been observed. The density of these dislocation loops decreased with the pre-irradiation temperature. After 450/500°C irradiation, the microstructure was coarse, indicating that the initial temperature (450°C) was high enough to be in the regime, where the growth of defects mainly occurs. In the case of downward temperature change, microstructures coarser than those of higher temperature irradiations were observed. This apparent anomaly may be understood in terms of the rate theory.

1. Introduction

In the future fusion reactors, structural materials are expected to experience non-steady irradiation temperature, neutron flux and other parameters during reactor start-up/shut-down, plasma disruptions, etc. Numerous irradiation experiments have been performed in order to clarify the behavior of microstructural evolution under non-steady irradiation conditions [1–3]. According to these experiments, the irradiation temperature history strongly affects the microstructural evolution and resultant mechanical property changes of materials. It has been also pointed out that the effect of temperature variations on properties of materials can be very complicated after several cycles of temperature change. In previous studies, irradiation tests have been conducted under complicated temperature histories [4], so that microstructural processes are rather complicated. In order to extract and understand the process in each temperature step, single temperature step irradiations have been conducted in the present study. Vanadium alloys have been chosen in this study because of their importance in fusion reactor structure material R&D [5–7], and also because they have relatively simple solid solution microstructure. Since solute atomic size factor has been known to have strong effect on the microstructural evolution, a series of binary alloys covering a wide range of atomic size factors have been studied.

2. Experimental

Pure vanadium and two binary vanadium alloys, V–5 at.% Cr, V–5 at.% Nb, and a candidate alloy, V–4 at.% Cr–4 at.% Ti–0.1 at.% Si, have been prepared. Chromium and silicon are undersize elements in the vanadium matrix, while titanium and niobium are oversize elements. The alloys were prepared by arc-melting in an argon gas atmosphere, followed by cold rolling into sheets. Transmission electron microscopy (TEM) disks were punched from the alloy sheets and annealed at 1100°C in a high vacuum (2×10^{-6} Torr).

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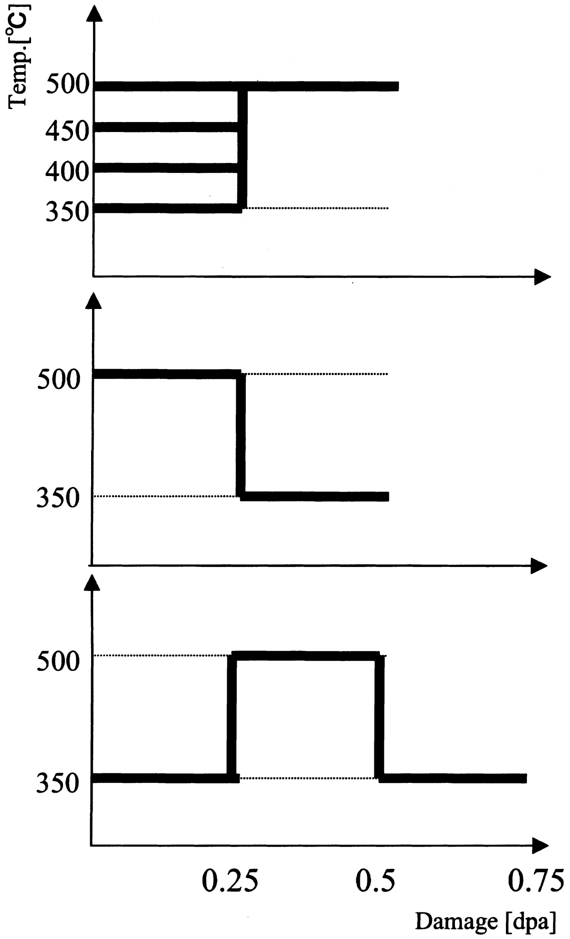

Fig. 1. Irradiation conditions.

The irradiation conditions is illustrated in Fig. 1. Irradiation with 4 MeV nickel ion was conducted in high fluence irradiation facility located at the University of Tokyo (HIT) [8]. Irradiations were performed either at a constant temperature of 500°C or in a stepwise temperature sequence of 350/500°C, 400/500°C, 450/500°C, 500/350°C or 350/500/350°C. In the cases of upward temperature change, widely different temperature combinations, i.e. T_{HIGH} and T_{LOW}, are selected in order to understand the effects of pre-irradiation temperature. While keeping T_{HIGH} in the temperature regimes where mainly growth occurs, T_{LOW} has been chosen in such a way that it covers nucleation temperature regime [9]. Irradiation is interrupted during the temperature change in this study. The radiation damage is up to 0.5 or 0.75 dpa, and the damage rates are about 1.5×10^{-4} dpa.

After electro-polishing, the microstructure of alloys was examined by TEM (JEM 2010). In situ observation of the ion-irradiated specimens using an HVEM (JEM 1250) was performed at room temperature in order to determine if the extended defects formed by the ion-irradiation were of interstitial type or vacancy type. If the extended defect is of interstitial type, they will grow by electron irradiation at room temperature where the vacancy has small mobility, whereas the extended defect will shrink if it is of vacancy type.

3. Results

3.1. Upward temperature change

Fig. 2 shows the microstructure of V–5 Cr. Network dislocations were observed in the specimen irradiated at

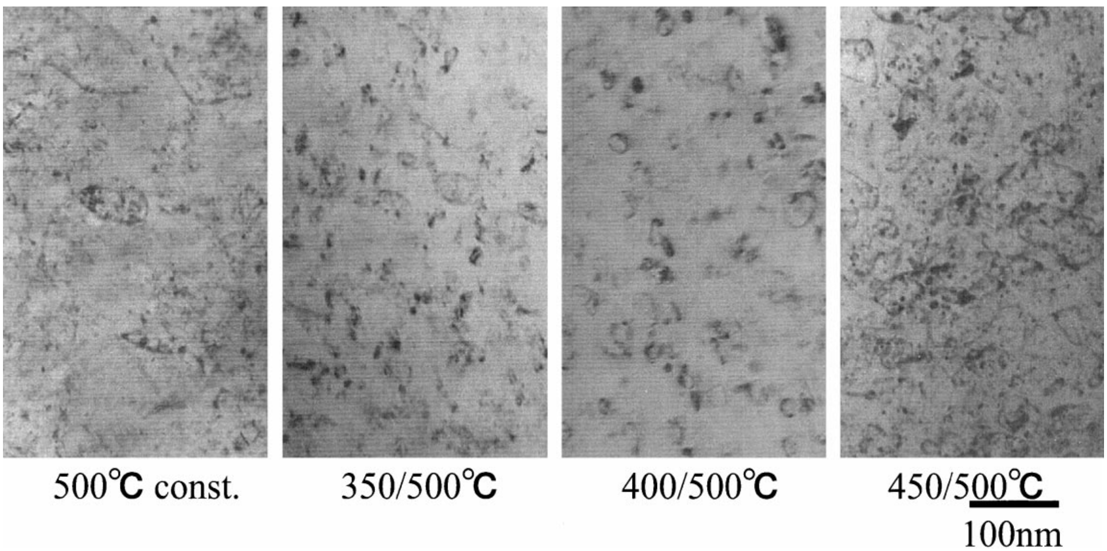

Fig. 2. Dislocation images of V–5 Cr after upward temperature change irradiation.

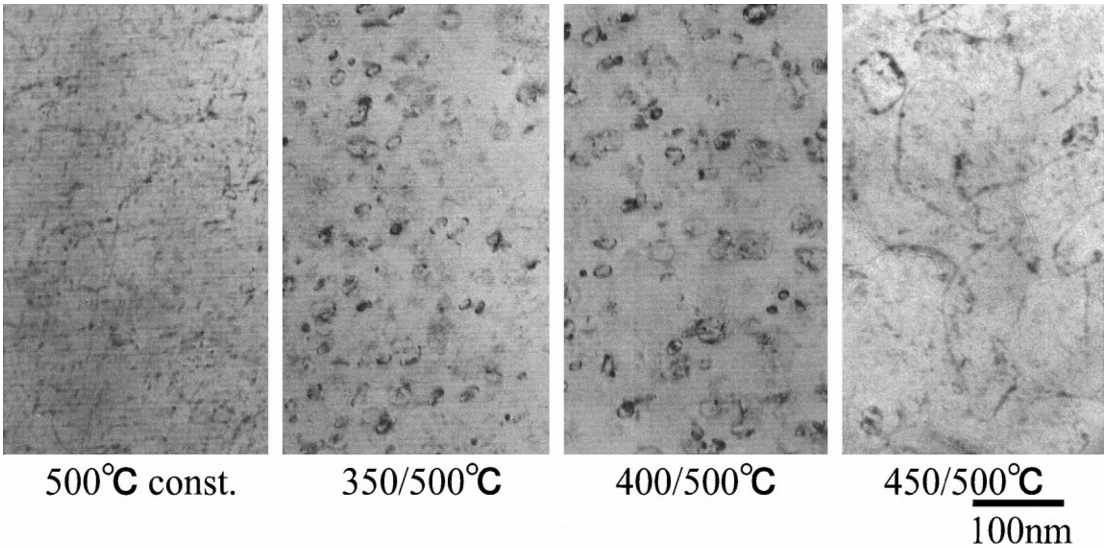

Fig. 3. Dislocation images of vanadium after upward temperature change irradiation.

a constant temperature of 500°C. Dislocation loops were observed after the irradiations, where pre-irradiations at T_{LOW} were conducted at a lower temperature than the subsequent, final irradiation temperature of T_{HIGH}. The size of these dislocation loops increased with the T_{LOW}.

Fig. 3 shows the microstructure of pure vanadium. Dense precipitates and network dislocations were observed in the specimen irradiated at 500°C. According to the previous study, the needle-like precipitates (vanadium carbide) were observed in ion-irradiated vanadium at 500°C [10]. Both of these precipitates seem to have the same structure. Dislocation loops were observed in specimens irradiated under stepwise temperature change. Although the size of the loops increased with the pre-irradiation temperature, the density of the loops decreases rapidly with the pre-irradiation temperature and coarse microstructure is observed after 450/500°C irradiation.

Fig. 4 shows the microstructure of V–5 Nb. A coarse microstructure is observed in the specimen irradiated under 500°C without pre-irradiation at a lower temperature. The microstructural evolution behaved in a similar manner as with V–5 Cr and pure vanadium when irradiated at 400/500°C and 450/500°C, temperature

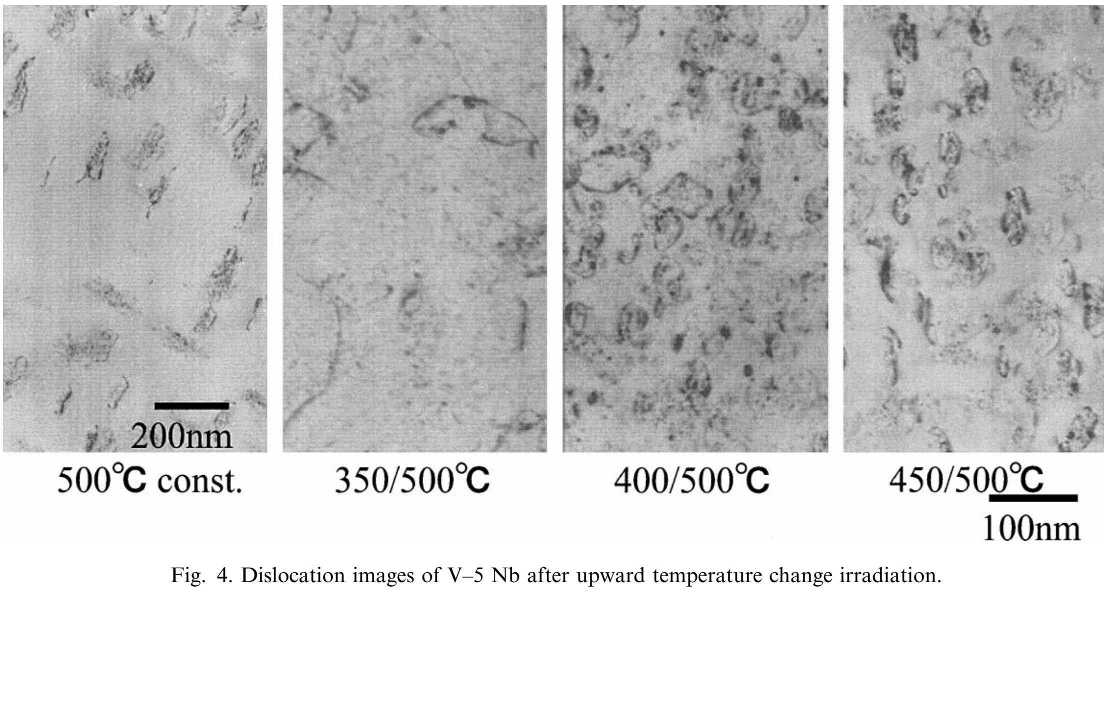

Fig. 4. Dislocation images of V–5 Nb after upward temperature change irradiation.

combination, but dense dislocation loops are not observed for 350/500°C irradiation in V–5 Nb. For the low temperature pre-irradiations, the stability of the extended defects against upward temperature changes was so poor that the extended defects shrank and disappeared.

3.2. Downward temperature change

Fig. 5 shows the cavity images of pure vanadium. After downward temperature change, i.e. 500/350°C irradiation, a coarse cavity microstructure was observed. After the 350/500/350°C three-step irradiation, cavities

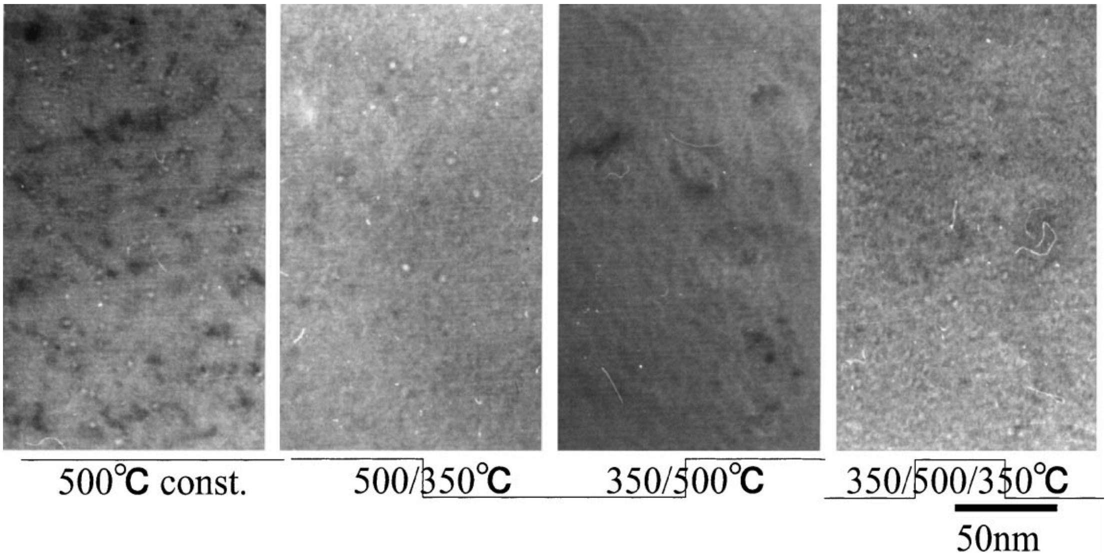

Fig. 5. Cavity images of vanadium after downward temperature change irradiation.

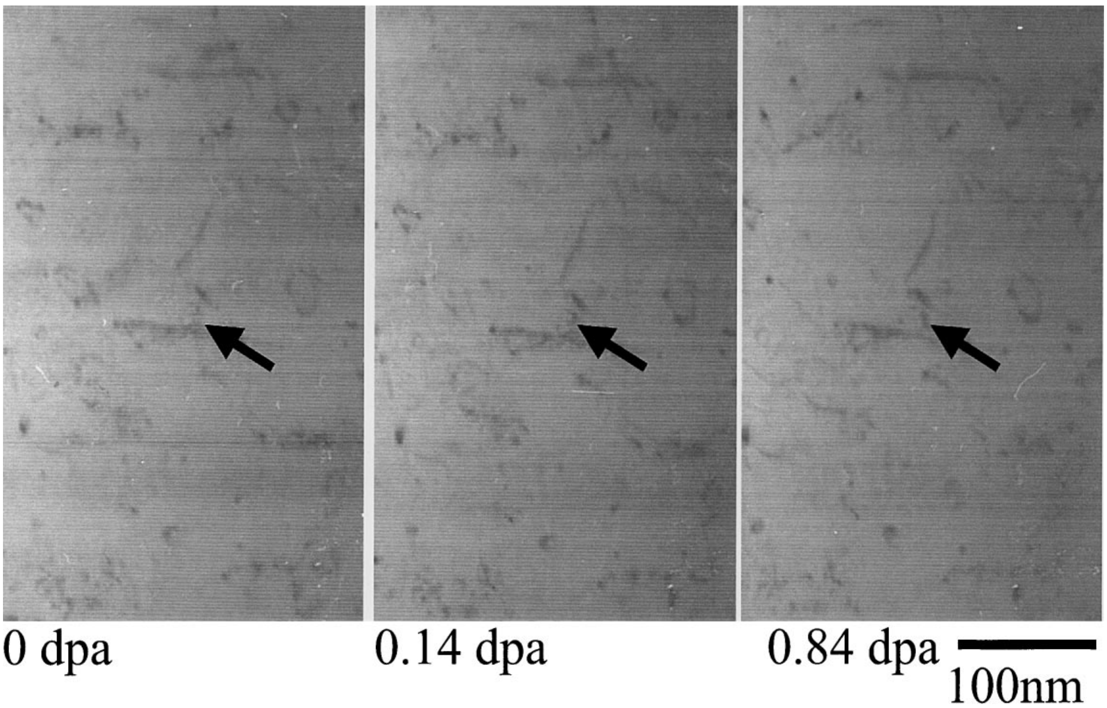

Fig. 6. Micrographs of HVEM in V–5 Cr after 500/350°C irradiation.

were observed despite the absence of cavities just after the 350/500°C irradiation sequence. Dislocation loops also grew in average size as a result of the downward temperature change from 500°C to 350°C.

3.3. *HVEM in situ observations*

A series of electron irradiations have been performed after ion irradiations in order to determine the nature of defects formed under ion irradiations. HVEM experiments were performed at room temperature and the damage rate were 4.5×10^{-3} dpa/s. As one example, a specimen of V–5 Cr after 500/350°C irradiation is selected here (Fig. 6). Since most of the loops grew during electron irradiation at room temperature, most of the dislocations formed under ion irradiations are judged to be of interstitial type. Tiny dislocation loops of interstitial type are formed during electron irradiation up to 0.84 dpa.

4. Discussion

4.1. *Processes during stepwise temperature increase during irradiation*

During upward temperature change, some extra vacancy flow is expected to occur during the upward temperature change because of a few mechanisms as described below. One of the mechanisms is the dissolution of tiny vacancy clusters caused by an increase in the irradiation temperature. During the pre-irradiation at a lower temperature, dense and tiny vacancy clusters are formed. Self interstitial atoms will annihilate with these tiny vacancy clusters which are invisible by TEM. As a result, the fraction of interstitials for the mutual recombination is small and unbalanced with the vacancy supply, resulting in vacancy surplus which shrink interstitial clusters. In this case, the dissolved and survived vacancies also have a chance to migrate and shrink interstitial cluster at higher temperatures. In addition to these processes, thermal dissociation of vacancy clusters might also enhance this process, although tiny vacancy clusters or microvoid is expected to be stable in this temperature range in vanadium [4,9]. In a previous study, recovery of defects was reported [10].

The other mechanism is the difference in the motion efficiency of vacancies and interstitials. According to an analysis based on reaction kinetics, the steady-state concentration of vacancies is greater at lower temperatures, and the mobility of vacancies is lower than the mobility of interstitials [3]. As a result, a short period of vacancy rich condition should occur just after an upward temperature change during irradiation. In the present study, this process is unlikely to occur, because the concentrations of mobile point defects, i.e. single vacancies and single interstitials should diminish when the irradiation is interrupted during the temperature change. This process leads to the nucleation of loops during the subsequent irradiation.

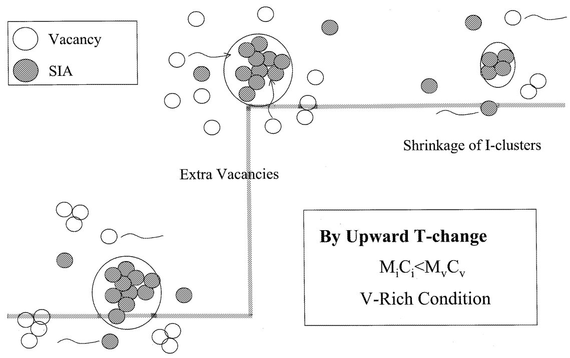

Fig. 7. The model during the upward temperature change irradiation.

Fig. 7 shows the model for the case of upward temperature change during irradiation. Small interstitial clusters nucleated during lower temperature irradiation, shrink by absorbing extra vacancies supplied from temperature change. After most of the unstable small interstitial clusters decrease in size, a coarse microstructure forms by the growth of surviving interstitial clusters. It appears that the stability of small clusters during the pre-irradiation at a lower temperature determines the final microstructure. For example, in the 350/500°C irradiation of V–5 Nb, a coarse microstructure is observed. This is in remarkable contrast with pure vanadium and V–5 Cr alloy, where rather fine dislocation loops dominate the microstructure. It is likely that the stability of small interstitial loops nucleated below 350°C is so low that the defects cannot survive to grow. It is clear from this observation that the stability depends on the added solute. The temperature regimes of nucleation and growth of defect seem to be shifted upward in V–5 Nb. In this alloy, the interaction between the oversized element niobium with vacancy is so strong that the mobility of vacancy is small due to the trapping effect by niobium atoms. As a result, the fraction of recombination site for Frenkel pairs increases, and consequently, the temperature regime where the interstitial migrate to sink is shifted to higher temperatures.

4.2. *Effects of downward temperature change*

In the case of the downward temperature change experiments, growth of both vacancy and interstitial clusters are observed despite vacancy-lean condition is expected from the rate theory during this temperature change [3]. The supply of extra vacancy seems to be needed for the growth of defect of vacancy type. Because the dissociation of vacancy cluster is regarded as thermal activation process, the stability is better in lower temperature. It is unlikely to occur that the tiny vacancy clusters become unstable during the downward temperature change. One possible reason for the growth of defects is as follows. Since the sink strength of point defect is large due to the dense extended defects formed during pre-irradiation at higher temperatures, the process of point defects migration to sinks is dominant compared with the mutual recombination of point defects during subsequent irradiation. The extended defects formed during pre-irradiation grow due to these migrating point defects. Consequently, coarse microstructures of both interstitial and vacancy types are observed.

5. Conclusions

1. Several upward and downward temperature change ion irradiations have been performed on several different vanadium alloys.

2. The character of almost all the dislocations was determined to be of interstitial type by in situ observations by HVEM.

3. It appears that the stability of small clusters during the pre-irradiation at a lower temperature determines the final microstructure.

4. After upward temperature changes, shrinkage of interstitial type extended defects occurred because of extra vacancy flow. Stability of small interstitial loops increases with the pre-irradiation temperature and depends on the component of alloys. The temperature regimes of nucleation and growth of defects are shifted upwards in V–5 Nb, because the interactions between point defects and solute atoms are strong.

5. After downward temperature change, growth of both vacancy and interstitial clusters was observed despite a vacancy-lean condition expected to occur as a result of a downward temperature change.

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