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Effects of temperature change on microstructural evolution in vanadium alloys under ion irradiation up to high damage levels

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Abstract

Recently, it has been pointed out that temperature change under irradiation strongly affects the microstructural evolution of materials. From an engineering point of view, it is very important to investigate the effects of temperature change under irradiation up to high damage levels. Ion irradiations were conducted in the High Fluence Irradiation Facility (HIT) up to 25 dpa. Both upward and downward temperature changes were performed with temperature combinations of 350/500 and 500/350 °C. Under irradiation with an upward temperature change, the microstructure in vanadium after the temperature change has been found to approach the microstructure of material irradiated at constant temperature with increasing damage level. On the other hand, significant effects of temperature change appeared in V–4Cr–4Ti–0.1Si at high damage level. Under irradiation with a downward temperature change, coarsening of defects was observed just after the downward temperature change. The microstructure approached the microstructure of the material irradiated at constant temperature with increasing damage level.

1. Introduction

In a fusion environment, structural materials are irradiated under non-steady state conditions, where several parameters such as temperature, thermal stress, and neutron flux are expected to change depending on operation mode or during startup and shutdown of the reactor.

In recent years, it has been pointed out that the effects of temperature change during irradiation on microstructural evolution are significant [1], and numerous experimental studies have been reported [2–4]. According to the results of both neutron and heavy ion irradiation experiments, shrinkage of interstitial loops occurs during an upward temperature change and growth of defects occurs during a downward temperature change [5].

When the materials are irradiated up to high damage levels, accumulation of extended defects causes swelling and/or degradation of mechanical properties. Accordingly, it may be difficult for a structural material to satisfy the proper performance that is demanded in a fusion reactor. In other words, the lifetime of the reactor is strongly dependent on resistance to irradiation. It is very important to obtain knowledge about the prediction of lifetime, for instance, whether the structural materials can be used safely up to 100 or 200 dpa. Thus, it is important to clarify the effect of temperature change on the prediction of the lifetime of the reactor. In regard to the effect of irradiation history, it has been reported that total dislocation densities of solution annealed and

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25% cold worked stainless steels approach a common value after 15 dpa irradiation at 400 °C [6]. In other words, microstructural evolution is determined by irradiation condition regardless of initial condition. It has also been reported that different sizes of precipitates in Ni–Al alloys approach a similar size after irradiation at 550 °C [7]. It is pointed out that whatever the initial starting configuration, the final state is one that is set solely by the irradiation condition. Not only from the viewpoint of fusion engineering but also a fundamental point of view, it is very interesting to gain knowledge of the 'history effect'; that is, whether the microstructure approaches the same state regardless of irradiation history. The objective of this study is to clarify the effect of 0.25 dpa pre-irradiation at different temperatures on the microstructural evolution of vanadium and vanadium alloys irradiated up to 25 dpa. Vanadium and vanadium alloys have been selected in this work because of their good performance as fusion structural materials [8–10].

2. Experimental procedures

Pure vanadium and V–4Cr–4Ti–0.1Si have been prepared. The alloy was arc-melted in an argon atmosphere. Specimens were cold rolled to 0.25 mm and were punched to 3 mm disks for transmission electron microscopy followed by mechanical polishing, electropolishing and annealing at 1100 °C for 2 h in a high vacuum (2×10^{-6} Torr), doubly wrapped with tantalum foils inside and zirconium foils outside.

Irradiations by 4 MeV Ni ions were conducted at the High Fluence Irradiation Facility, the University of Tokyo (HIT) using a Tandetron accelerator [11].

Damage levels were from 0.5 to 25 dpa and damage rates were about 5×10^{-4} dpa/s. Irradiations were performed either at constant temperatures of 350 and 500 °C or in a stepwise temperature sequence of 350/500 or 500/350 °C. To investigate the effect of short-term pre-irradiation temperature history on microstructure, materials were irradiated to 0.25 dpa followed by a stepwise temperature change, and then damage levels were set to 0.25–25 dpa during subsequent irradiation. There was an exception; namely, the irradiation of 500 °C (0.25 dpa)/350 °C (0.5 dpa) in pure vanadium was performed at the Research Institute for Applied Physics in Kyushu University with 2.4 MeV Cu ion. Irradiation conditions are illustrated in Fig. 1.

TEM foils were prepared using a sectioning technique followed by back-thinning so that the damage region could be observed. Damage calculations were performed by the TRIM-code [12]. The amount removed from the irradiated surface by sectioning was determined using a laser microscope. The electrolyte used in this work was a mixture of 4 parts ethanol and 1 part hydrosulphuric acid.

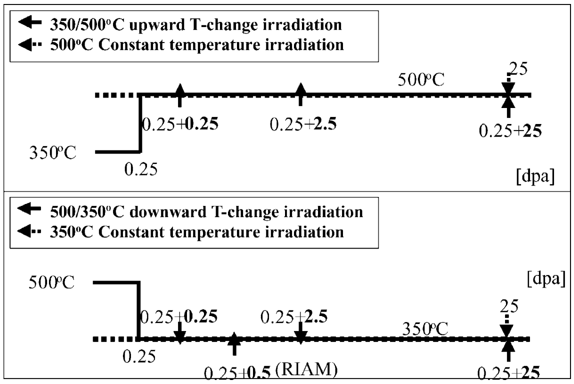

Fig. 1. Irradiation conditions. Heavy ion irradiations with upward or downward temperature change have been performed in HIT.

3. Results and discussion

3.1. 350/500 °C upward temperature change

Fig. 2(a) shows the microstructure of pure vanadium under irradiation with an upward temperature change. About 30 nm diameter dislocation loops were formed under 350/500 °C (0.25 dpa) irradiation. These dislocation loops grew to line dislocations after 2.5 dpa irradiation at the higher temperature of 500 °C. In this condition, needle-like precipitates and cavities were also observed in pure vanadium. The average cavity size was 4 nm and its number density was 4.97×10^{22} m^{-3}. This kind of precipitate has been reported in several papers, in which the precipitates were determined as vanadium carbide [13]. It has been reported often that cavities tend to coexist with precipitates in pure vanadium [13]. Tangled dislocation lines were observed with increasing damage level up to 25 dpa (350/500 °C (25 dpa)). In this condition, both precipitates and cavities disappear and the microstructure seems to approach the microstructure of the material irradiated at a constant temperature of 500 °C. Dissociation of precipitates seems to occur, perhaps because the precipitates become unstable in the presence of cascade events induced by the incident ion.

Fig. 2(b) shows the microstructure of V–4Cr–4Ti–0.1Si under irradiation with an upward temperature change. At 0.25 dpa irradiation after the upward temperature change, a high density of defect clusters was formed with dot-like contrast. These tiny defect clusters are expected to be a mixture of dislocation loops of interstitial type and precipitates of titanium oxide, although the determination of their nature was difficult because of their size. After 2.5 dpa irradiation at high temperature, dislocation lines and tiny defect clusters were observed. Although the average size of these tiny defect clusters did not change clearly with increasing damage level, the number density of defect clusters

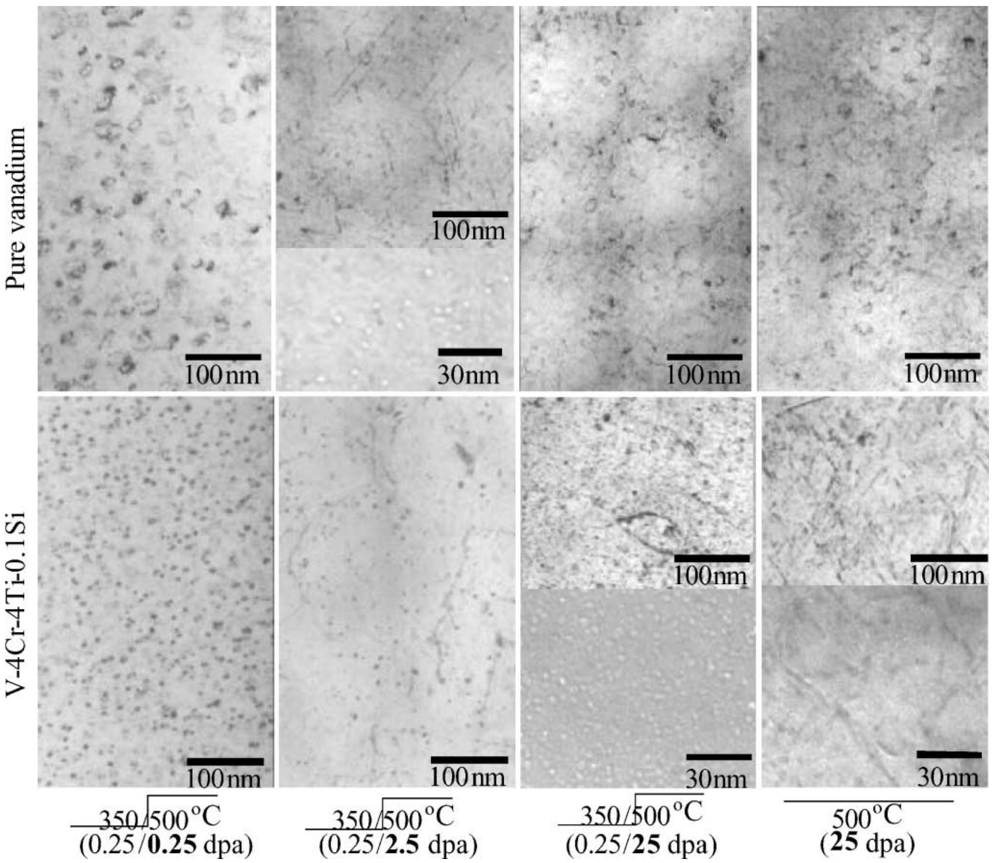

Fig. 2. (a) Microstructure of pure vanadium (above) and (b) V–4Cr–4Ti–0.1Si (below) under irradiation with upward temperature change.

decreased from 3.63×10^{22} to 4.82×10^{21} m^{-3}. On the assumption that the growth rates are different between dislocation loops and precipitates, the dotted tiny defect clusters, which remain constant in their size after 2.5 dpa irradiation at high temperature, were precipitates and the decrease in number density of tiny defects were dislocation loops. Under this irradiation condition (350/500 °C (2.5 dpa)), very tiny cavities were observed in V–4Cr–4Ti–0.1Si. This cavity formation should be discussed in terms of the effect of temperature change, because this alloy is well known as a good swelling resistant material, and there are few data on cavity observations at this damage level under constant temperature irradiation. With increasing damage level at high temperature to 25 dpa, the number density of dotted defect clusters increased to 2.63×10^{22} m^{-3}, but the size of these defect clusters did not show any significant change and remained about 5 nm. Under this irradiation condition (350/500 °C (25 dpa)), a high density of small cavities was clearly observed in V–4Cr–4Ti–0.1Si. It can be said that a quite different microstructure was observed compared to the 25 dpa constant temperature irradiation where swelling was restricted. In other words, large precipitates were formed with a high density without cavity formation under constant temperature irradiation. There is a suggestion that large precipitates and cavities do not coexist in V–4Cr–4Ti–0.1Si, which has been reported previous [14].

3.2. 500/350 °C downward temperature change

Fig. 3(a) shows the microstructure of pure vanadium under irradiation with a downward temperature change. Under 0.25 dpa irradiation at the lower the temperature after temperature change, dislocation lines and cavities were observed. This is in good agreement with previous work, which reported that coarsening of defects occurs after a downward temperature change [5]. With increasing damage to 0.5 dpa at the lower temperature, cavities could not be observed and dark field images show dense tiny dislocation loops superimposed on a microstructure of dislocation lines. From this damage level at the lower temperature, re-formation of defect clusters started and interstitials might shrink cavities because the mobility of vacancies is not so high at this temperature range. Under 500/350 °C (2.5 dpa) irradi-

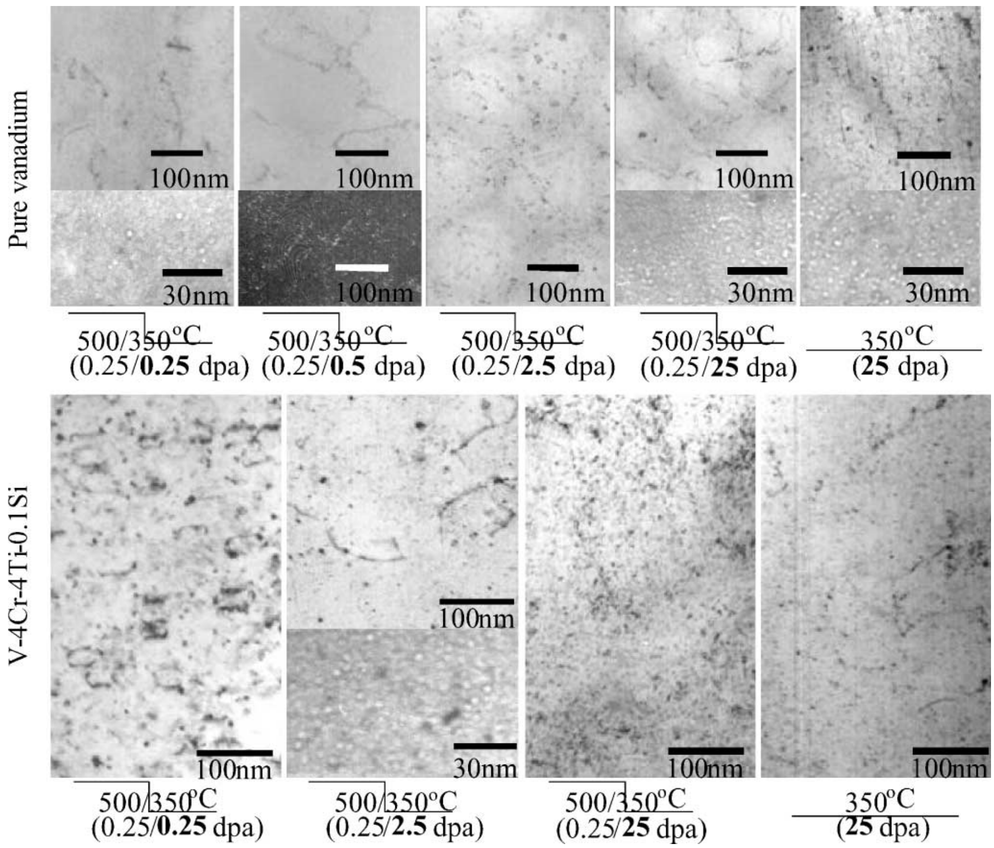

Fig. 3. (a) Microstructure of pure vanadium (above) and (b) V–4Cr–4Ti–0.1Si (below) under irradiation with downward temperature change.

ation, re-formed small size dislocation loops were clearly observed even in bright field conditions. Cavities were not observed after 2.5 dpa irradiation at the lower temperature after a temperature change, but they were formed again after 25 dpa irradiation at the lower temperature with a mean size of 3.44 nm and a number density of 3.20×10^{22} m^{-3}. By comparison, the number density of cavities was 1.22×10^{23} m^{-3} and their mean size was 4.08 nm under constant temperature irradiation of 350 °C. Hence, it can be said that the microstructure of the material subject to 500/350 °C (25 dpa) irradiation approached the microstructure of the material exposed to constant temperature irradiation. Precipitates (vanadium carbide) and cavities existed in pure vanadium under 25 dpa irradiation with temperature change, while there were no cavities under constant temperature irradiation at 500 °C. According to theories of point defect clustering, in this temperature regime, growth of vacancy clusters should be enhanced by raising the irradiation temperature. However, the inverse phenomenon was obtained in this study because of a strong correlation between precipitates and cavities in pure vanadium.

Fig. 3(b) shows the microstructure of V–4Cr–4Ti–0.1Si under irradiation with a downward temperature change. In this alloy, 50 nm dislocation loops and dot-contrasted tiny defects were formed under 500/350 °C (0.25 dpa) irradiation. In this irradiation condition, the growth of defect clusters induced by a downward temperature change was not clearly observed. Comparable large dislocation loops might be formed under irradiation at 500 °C, and they might not grow rapidly after a downward temperature change. If the difference in growth rate between precipitates and dislocation loops is taken into account, dotted tiny defect clusters seem to be precipitates. With increasing damage level at lower temperature to 2.5 dpa, dislocation loops grew to dislocation lines, and cavities were also observed in V–4Cr–4Ti–0.1Si. Although the growth of defect clusters occurs just after the downward temperature change in pure vanadium, it was at 2.5 dpa that the defect clusters start to grow in V–4Cr–4Ti–0.1Si. It could be said that low mobility of point defects in V–4Cr–4Ti–0.1Si delays the growth of defects induced by a downward temperature change. Under 500/350 °C (25 dpa) irradiation, dislocation lines and a high density of fine defects were formed and there were no cavities. By comparison with the microstructure under 350 °C (25 dpa) constant temperature irradiation, there was no significant difference with or without temperature change.

3.3. Effect of temperature change on precipitates

A significant effect of temperature change appears in V–4Cr–4Ti–0.1Si after upward temperature change (Fig. 2(b)). In this alloy, it is well known that titanium oxides have a great influence on cavity formation. It is important to understand the mechanisms of microstructural evolution of precipitates. In this study, cavity formation did not occur in the presence of titanium oxide. It has been also reported that precipitates and cavities do not coexist under irradiation with temperature change in Fe–16Cr–17Ni–0.1P [15], because the precipitates shrunk during the upward temperature change. Moreover, in previous studies, it is reported that titanium oxides disappear after upward temperature change [16,17]. In the present work, it seems that precipitates had shrunk by upward temperature change and nucleation of cavities was enhanced during subsequent irradiation at high temperature. More work on the details is needed to understand the microstructural evolution of precipitates under irradiation with temperature change.

The fact that a high density of cavities was formed after irradiation with upward temperature change in V–4Cr–4Ti–0.1Si is very important from an engineering point of view, because it is important to investigate whether the material properties are affected by temperature change or not. In other words, the lifetime of a reactor may be affected by irradiation history or not. In the present work, it can be said that temperature change enhances swelling compared to constant temperature irradiation. Although swelling was only 0.6% after 25 dpa irradiation with temperature change in this study, there is a possibility that significant swelling will be induced with increasing damage levels. In the case of downward temperature change, it can be also pointed out that swelling is enhanced just after the temperature change because of the transitional growth of defect clusters.

4. Conclusion

Heavy ion irradiations with temperature change were conducted up to 25 dpa to understand microstructural evolution in vanadium and a vanadium alloy.

The microstructure of pure vanadium after upward temperature change approached the microstructure of material subject to constant temperature irradiation, and there was no significant effect of temperature history. On the other hand, a high density of cavities was formed after 25 dpa irradiation with upward temperature change in V–4Cr–4Ti–0.1Si, while there were no cavities but large precipitates under constant temperature irradiation. It seems that precipitates are very sensitive to the upward temperature change. It can be concluded that a short pre-irradiation at lower temperature caused significant 'history effects' on microstructural evolution in V–4Cr–4Ti–0.1Si.

Defect clusters that were formed by pre-irradiation at high temperature grew just after the downward temperature change followed by re-formation of new defect clusters under subsequent irradiation at lower temperature. With increasing damage levels at lower temperature, defect clusters grow and the microstructure approaches that of material subject to constant temperature irradiation. It is concluded that microstructural evolution is delayed by a downward temperature change under the irradiation conditions of this study. This delay was larger in V–4Cr–4Ti–0.1Si because of the lower mobility of point defects.

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