Correlation of Microstructural Evolution in V-4Cr-4Ti by Heavy Ion and Neutron Irradiations*)

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The evolution of interstitial dislocation loops in V-4Cr-4Ti during heavy ion and neutron irradiation was investigated in comparison with that during electron irradiation at temperature ranging from 407 K to 673 K. Temperature and damage rate dependence of the loop number density by heavy ion and neutron irradiations agreed well with those by electron irradiation obeying Sink and Recombination Dominant kinetics at low and high temperature, respectively. Heavy ion and neutron irradiations induced higher loop number density than that of electron irradiation at 563 - 673 K. The difference may be attributed to cascade-enhanced vacancy loop formation.

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1. Introduction

Vanadium based alloys are promising candidate materials for structural components of blankets because of their low activation characteristics and high temperature strength. Especially V-4Cr-4Ti has been regarded as the leading candidate because of its balanced high temperature strength, low temperature ductility, workability and so on [1, 2]. However, this material has a severe low temperature operation limit caused by irradiation embrittlements. At present the limit is regarded as being not less than 673 K.

The radiation embrittlement is known to be originated from the formation of high density defect clusters during irradiation, which can cause hardening, flow localization during plastic deformation and reduction of fracture resistance [3].

Defect clusters formed in V-4Cr-4Ti by irradiation were identified to be dislocation loops, Ti-rich precipitates and their complexes, whose fraction is dependent on temperature. Extensive researches on dislocation loop evolution have been carried out using electron beams in HVEM (High Voltage Electron Microscope) for vanadium alloys [4–6]. However, electrons cannot cause displacement cascade which is the characteristic of fusion neutron irradiation effects.

The purpose of this study is to investigate dislocation loop evolution processes of V-4Cr-4Ti under heavy ion and neutron irradiations, which produce cascade damage, and correlate with those obtained by the past studies using HVEM. The understanding on the kinetics of microstructural evolution under irradiation with various species is expected to enhance predictability of radiation embrittlement of V-4Cr-4Ti in fusion conditions.

2. Experimental Procedure

The material used is a high purity V-4Cr-4Ti alloy designated as NIFS-HEAT-2 [7], heat treated at 1273 K for 2 hr.

Ion irradiation was carried out with 2.4 MeV Cu^{2+} ions using Tandem Accelerator at RIAM, Kyushu University, with the current of 8.0 nA/mm^2. Figure 1 shows SRIM code calculation of depth profile of displacement and Cu atom deposition rates. After irradiation, the surface of ~600 nm were removed by electro-polishing follower by back-thinning to perforation. The observed area

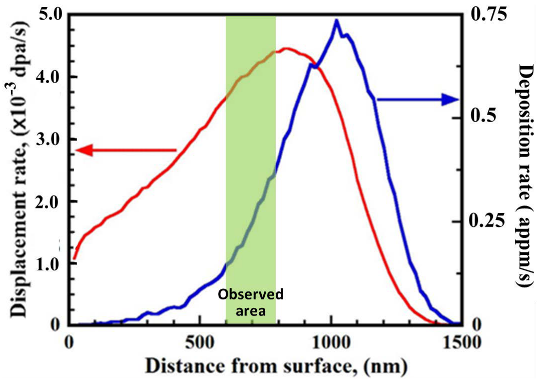

Fig. 1 Depth profile of displacement rate and ion deposition rate by 2.4 MeV Cu^{2+} on V-4Cr-4Ti in the present condition.

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Table 1 Summary of irradiation conditions.

Irradiation	Temperature	dpa	dpa/s
2.4MeV Cu^{2+} ions	403-413K	1.0	4.5×10^{-3}
	473K	1.0, 2.5, 10	4.5×10^{-3}
	573K	1.0	4.5×10^{-3}
	673K	0.15, 1.0	4.5×10^{-3}
Fission Neutrons (BR2)	363K	0.20	1.1×10^{-7}
Fission Neutrons (JMTR)	563K	0.08	4.4×10^{-8}

is schematically shown in Fig. 1. The result shows that ~30 appm Cu deposited at the observed area after irradiation to 1 dpa. Transmission Electron Microscope (TEM) observations of microstructures were carried out with JEM-2000FX at RIAM, Kyushu University.

The irradiation conditions are summarized in Table 1. The temperature varied between 403 and 413 K in the case denoted as 403 - 413 K for irradiation temperature in Table 1.

Neutron irradiations were carried out in JMTR and BR2 with disc specimens of 3 mm diameter and 0.15 - 0.25 mm thick. The specimen was electropolished from the both sides to perforation. TEM microstructural observations were carried out with JEM-2000FX at Oarai Center of IMR Tohoku University. Some of the results of JMTR irradiation was already reported elsewhere [8].

3. Results

Figure 2 shows microstructure after irradiation with 2.4 MeV Cu^{2+} ions to 1 dpa. At 403 - 413 K, 473 K, and 573 K, high density of dot images, which are known to represent small dislocation loops, were observed as well as loops. Weak beam dark field (WBDF) images, shown at the upper right corners, clearly show dot and loop images.

At 673 K and 1 dpa, loops and precipitates were mixed. In this case, an imaging in a diffraction condition far removed from Bragg conditions ($s \gg 0$: s is the deviation from Bragg condition) was also carried out and shown in lower area of Fig. 2 (d). Because the images in the conditions far removed from Bragg condition only exhibit the precipitates, comparison of the two images allows identification of dislocation loops. Also carried out was an irradiation to 0.15 dpa, when only dislocation loops were observed as shown in Fig. 2 (e). By these two ways, estimation of the loop number density was possible at 673 K. The loop number density derived by the two methods were close with each other. The loop number density data will be shown in the next section, comparing with those by neutron and electron irradiations.

Figure 3 shows irradiation dose dependence of microstructural evolution at 473 K. At 1 and 2.5 dpa, dislocation loops are observed predominantly. WBDF images, shown at the upper right corner, clearly shows dot and loop images. At 10 dpa tangled dislocation network are formed with lower density of loops. No evidence of precipitation

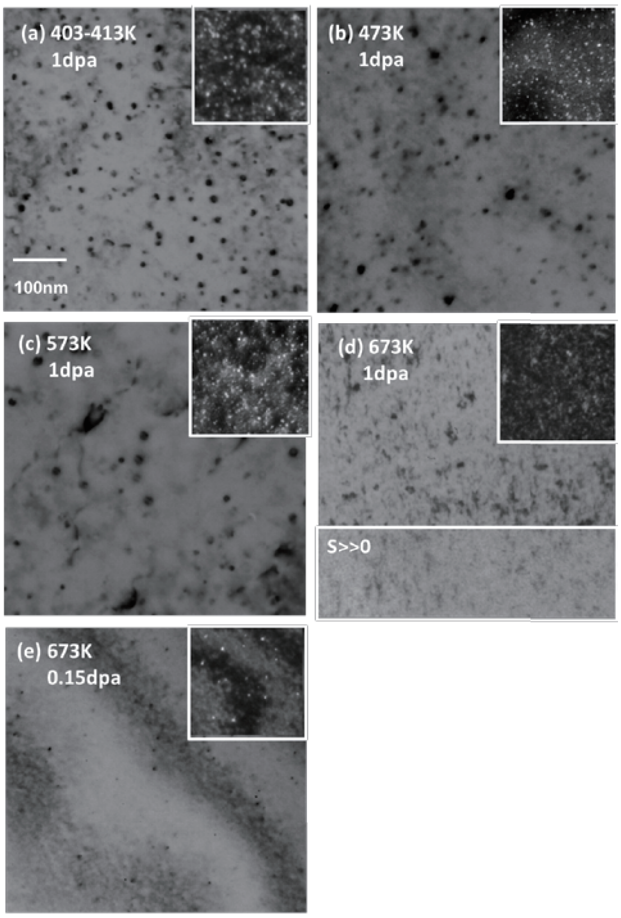

Fig. 2 Microstructural evolution by 2.4 MeV Cu^{2+} ion irradiation. Upper right corners show WBDF images.

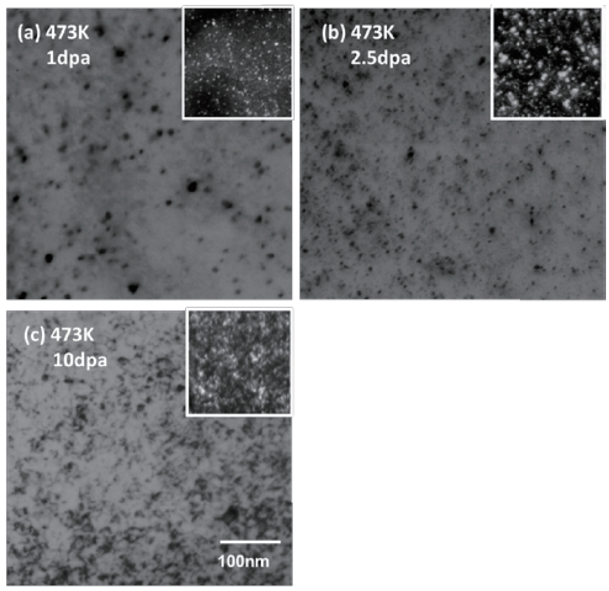

Fig. 3 Microstructural evolution versus dose at 473 K by 2.4 MeV Cu^{2+} ion irradiation. Upper right corners show WBDF images.

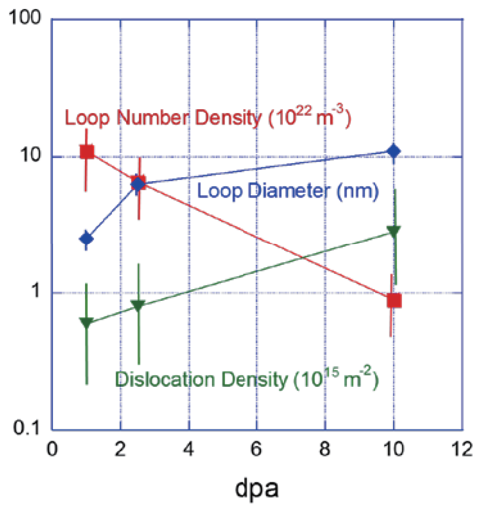

Fig. 4 Microstructural parameters as a function of dose (dpa) by 2.4 MeV Cu^{2+} ion irradiation at 473 K.

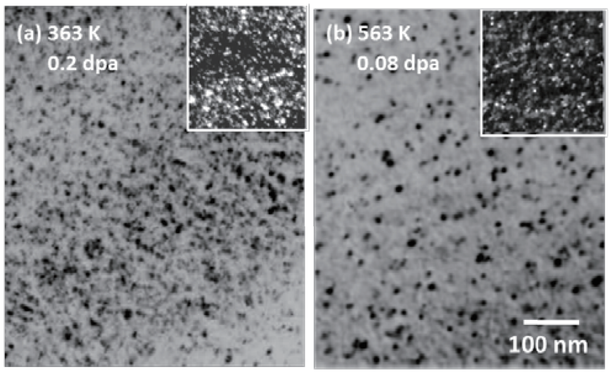

Fig. 5 Microstructural evolution by neutron irradiation. (a) Irradiation in BR2. (b) Irradiation in JMTR. Upper right corners show WBDF images.

was observed.

Figure 4 summarizes microstructural parameters. The loop number density is highest at 1 dpa, showing that the loop nucleation was completed before 1 dpa. Loops grow and coalesce and tangles with dose, decreasing their number density. The dislocation density, which is defined as a total dislocation length per volume, continues to increase because of formation and evolution of tangled dislocation network. The change shown in Fig. 4 is a typical initial microstructural evolution in irradiated metallic materials.

Figure 5 are microstructures of neutron-irradiated V-4Cr-4Ti (a) at 363 K and 0.2 dpa by BR2 and (b) at 563 K and 0.08 dpa by JMTR. The microstructure of the two cases are composed of dislocation loops. No evidence of precipitate formation was seen. The quantitative date will be shown in the following section with ion and electron irradiation data.

4. Discussion

Figures 6 and 7 summarize loop number density as a function of temperature and damage rate, respectively. The data by electron irradiations [4, 6] were also indicated for comparison. Figure 6 shows that the temperature dependence of the loop number density is very weak to ~473 K for electron irradiation and to ~573 K for ion irradiation, both followed by significant negative temperature dependence. A datum for neutron irradiation at 363 K, is close to those for electron and ion irradiations in the similar temperature range.

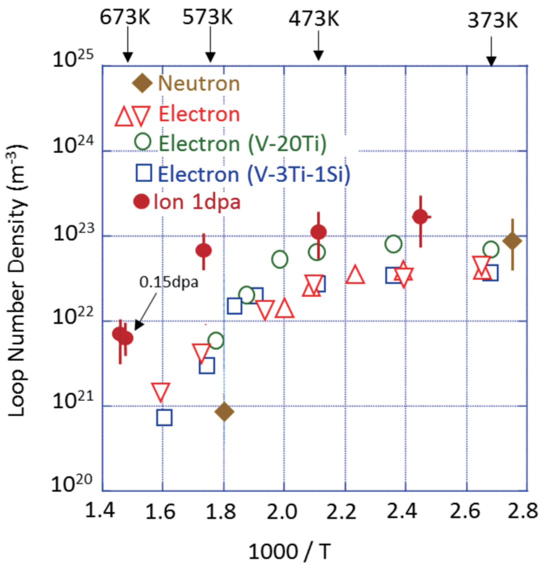

Fig. 6 Temperature dependence of dislocation loop number density. Data by electron irradiation for V-4Cr-4Ti ($0.8{\sim}1.3 \times 10^{-2}$ dpa/s) [6] and V-20Ti and V-3Ti-1Si (1.6×10^{-4} dpa/s) [4] are also shown for comparison.

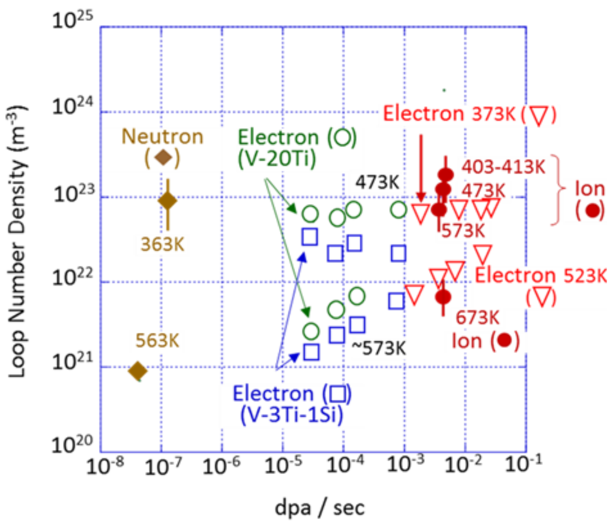

Fig. 7 Damage rate dependence of dislocation loop number density. Data by electron irradiation for V-4Cr-4Ti at 373 K and 523 K [6] and V-20Ti and V-3Ti-1Si at 473 K and ~573 K [4] are also shown for comparison.

Figure 6 shows that all the loop number density data within the temperature range of 363 K to 473 K lie between 0.3 to 3 × 10^{23} m^{-3}, showing weak temperature and irradiation species dependence. On the other hand, data at the temperature higher than 523 K have clear negative temperature dependence. However the loop number density by ion irradiation is significantly higher than that of electrons in this temperature range in spite of similar damage rate. The low loop number density by neutrons at 563 K can be explained by its very low dose rate and is correlated in Fig. 7 using damage rate as a parameter.

A comparison of loop number density taking the damage rate effects into account is possible by Fig. 7. The data at temperature ranging from 363 K to 473 K lie between 0.3 to 3 × 10^{23} m^{-3}, showing weak damage rate dependence. Data at 523 K to 673 K show clear damage rate dependence, but data by ions at 573 K and 673 K and neutron at 563 K show significantly higher loop number density than those by electrons in the same temperature range.

Therefore the dislocation data for ion, neutron and electron irradiations can be summarized as follows.

(1) At 473 K and below, the loop number density by ion, neutron and electron irradiations are close with each other and have weak dependence on temperature and damage rate.
(2) Significant temperature and damage rate dependence appear at 563 K and above.
(3) At 563 K and above the loop number density by ion and neutron irradiation is significantly higher than those of electron irradiation.

The electron irradiation data have been analyzed using chemical rate equations and the following two analytical expressions were derived which can be applied to many metals and alloys [9, 10].

(a) Loops are preferentially formed at a limited number of interstitial trapping sites (heterogeneous nucleation), referred as Sink Dominant regime. In this case the loop number density is equal to the effective interstitial trap number density, without having temperature and damage rate dependence.
(b) Loops are formed by mutual clustering of interstitials (homogeneous nucleation), referred as Recombination Dominant regime. In this case the loop number density is a negative function of temperature and positive function of damage rate. The meaning of the activation energy derived by the slope of Arrhenius plot of the loop number density is shown in Fig. 8 (b).

Comparing these pictures with the present results shown in Figs. 6 and 7 and summarized in this section, the defect processes at 473 K and below and at 563 K and above corresponds to the Sink Dominant and Recombination Dominant regimes, respectively. However, these pictures cannot explain high loop number density by ion and neutron irradiations only at 563 K and above.

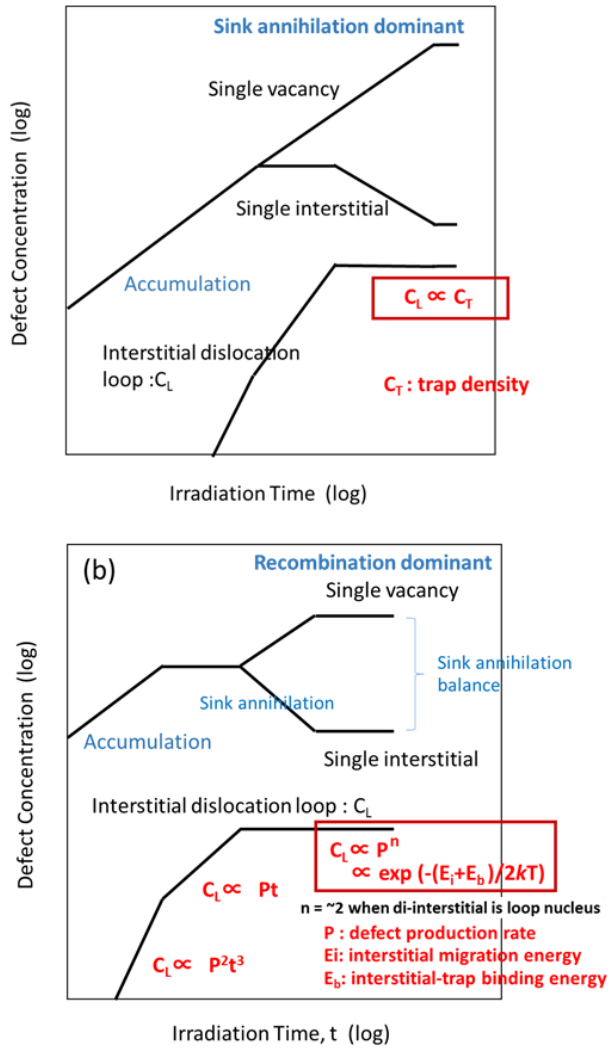

Fig. 8 Schematic representations of time dependence of defect cluster evolution derived by chemical rate equation models [9]. Also shown is the evolution of interstitial clusters with time derived by analytical and numerical analysis [10]. (a) Sink Dominant regime, and (b) Recombination Dominant regime.

A comparison of loop number density by electron and neutron irradiation was carried out for Fe-Cr-Ni austenitic ternaries [11], which also showed higher loop number density by neutrons relative to that by electrons considering the temperature and damage rate dependence of the density. Among the possible reasons pointed out in the paper is an enhancement of interstitial clustering and thus loop nucleation by cascade damage. This may explain the results shown in Fig. 6 that the loop number density by ion and neutron irradiations was slightly higher than that by electron irradiation in the temperature range of 363 - 473 K, but cannot explain significantly larger loop number density by ion irradiation than that of electron at 573 and 673 K because the cascade induced enhancement of interstitial loop nucleation should have weak temperature dependence.

One of the possible additional mechanisms would be that vacancy loops, induced by depleted zones of cascade damage, were also formed because vacancies are mobile in this temperature range. Unfortunately it is difficult to identify the type of small dislocation loops (vacancy type or interstitial type) by TEM observations. However, this idea is supported by a positron annihilation lifetime measurements [12] in which vacancy-Ti complexes were detected in V-4Cr-4Ti as a result of vacancy migration in the temperature range of 493 K to 623 K. The migration energy of vacancies can be obtained also by the temperature dependence of loop growth rate by HVEM [13]. The estimated vacancy migration energy for V-20Ti and V-3Ti-1Si were 1.36 eV and 1.22 eV, respectively [4]. Assuming 1 to 10 jumps per second is necessary for the clustering, the temperature where the vacancy clustering starts to occur, i.e. minimum temperature of vacancy clustering, is expected to be between 473 K to 573 K [4].

By heavy ion irradiation at 673 K and 1 dpa, a mixture of loops and precipitates was observed as shown in Fig. 2 (d). This means that the loop formation and precipitation progress simultaneously at this relatively high temperature range, which was more systematically shown in [14]. The two processes will influence with each other but the interaction will be dependent on temperature and damage rate. Modeling of microstructural evolution including point defects-solute interaction is essential for the prediction of microstructural changes under irradiation in the temperature range.

Microstructure-mechanical property correlation is a crucial study to apply the present data to the estimate of radiation hardening and embrittlement. An effort to evaluate mechanical properties using the irradiated specimens in common with the present study is in progress using nano-indentation techniques [15].

The mechanistic understanding on the difference of the irradiation effects by fission-neutron, ion and electron irradiations will enhance accuracy of the prediction of D-T fusion neutron irradiation effects to be established based on the surrogate irradiation means. In this case, the validation of the prediction is foreseen by the irradiation facilities of neutrons whose spectrum closely resembles that of D-T fusion neutrons, such as IFMIF (International Fusion Materials Irradiation Facility) [16].

5. Conclusion

The temperature and damage rate dependence of the interstitial dislocation loops number density in V-4Cr-4Ti produced by heavy ions, fission neutrons and electrons were well correlated with each other based on the conventional rate equation-based kinetic models in the temperature range of 403 - 493 K. However, effects of cascade damage, enhancing vacancy clustering, need to be taken into account at 573 K and 673 K where vacancies are highly mobile for the case of heavy ion and neutron irradiations. In this temperature range, where loop nucleation and precipitation proceeds simultaneously, defect-solute interaction will also be the key process and must be included in the modeling.

Acknowledgements

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