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# Defect cluster formation in vanadium irradiated with heavy ions

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**Abstract**

Irradiation data of vanadium alloys have been accumulated by intensive irradiation experiments in fission reactors. In evaluating irradiation performance of the alloys in fusion environments, we should consider the effects of high energy cascade damage and transmutation reactions under 14 MeV neutron irradiation. Effects of high generation rate of helium on microstructural evolution and mechanical properties in vanadium alloys have been studied by several techniques including dynamic helium charging experiments (DHCE) and boron doping. However, fundamental understanding on defect cluster formation under cascade damage in vanadium has not yet been clarified in detail. In this study, the effect of cascade damage on vacancy cluster formation was investigated as a function of energy transfer by cascades using several kinds of heavy ion irradiations to thin foils specimens. No defect clusters were observed by transmission electron microscope (TEM) in thin foils of 99.8% pure vanadium irradiated with 200 and 400 keV self-ions ($V^+$) up to $1 \times 10^{16}$ ion/m$^2$ at room temperature. Thin foil specimens were also irradiated with $Au^+$ and $Xe^+$ ions to $1 \times 10^{16}$ ion/m$^2$. Energies of irradiating ions were 50, 100, 200, 300 and 400 keV. In the specimens irradiated with $Au^+$ ions, defect clusters of about 2–2.5 nm were detected by TEM. The areal density of the observed defect clusters increased with ion energy and was also found to be dependent on the thickness of the specimens. In the thin region of the specimens, density of the defect clusters per damage energy deposition increased with ion energy. These indicate that vacancy clusters are produced by high density of displacements in cascade damage. In the thicker region of the specimens, interstitials can easily annihilate vacancy clusters and form interstitial clusters. At the foil thickness of 20 nm, the minimum energy of gold ions to produce vacancy clusters was estimated to be 120 keV. This corresponds to the damage energy transfer density of 4.5 keV/nm/ion. 

## 1. Introduction

Vanadium alloys are one of the attractive candidate materials for fusion structural materials because of their low induced radioactivities after neutron irradiation in addition to good mechanical properties at elevated temperatures. Extensive efforts have been made to improve irradiation performance of vanadium alloys using fission reactors. Simultaneous effects of heavy irradiation damage and high helium generation rate under fusion irradiation environments have been studied using DHCE (dynamic helium charging experiments) [1,2] and boron-doping technique [3] in fission reactor. Solid transmutation effects on microstructural evolution have also been investigated [4,5].

However, very limited data are available on fundamental defect production from cascades in this metal. In BCC metals [6–8], cascade damage generally results in a very low density of defect clusters which are visible by transmission electron microscope (TEM) in irradiated specimens compared with FCC metals [9–11]. Recently molecular dynamics simulation of cascade damage from high energy primary knock-on atom (PKA) successfully demonstrated defect production behavior with an optimized interatomic potential for vanadium [12].

Irradiation experiments of FCC metals with self-ions have been successfully used to correlate single cascade effects on defect cluster formation with primary PKA energy spectrum under neutron irradiation [9,13]. As a first step of the experiments to investigate critical rules for vacancy cluster formation in vanadium, self-ion

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irradiation of thin foil specimens was performed under the wide range of energy and fluence. However, it was difficult to detect defect cluster formation in vanadium up to $1 \times 10^{16}$ $V^+$ ions/m$^2$ by TEM.

In this study, heavier $Au^+$ and $Xe^+$ ions were used to irradiate thin foil specimens to get a high density of displacement, and the formation of defect clusters is reported.

## 2. Experimental

3 mm disks of 99.8% pure vanadium were annealed at 1273 K for 1 h in a high vacuum with Zr foils followed by furnace cooling. This material contains 120 wt. ppm Oxygen in addition to metallic impurity atoms such as Al, Si, Fe, Cr and Cu. The specimens were electro-polished to get thin foils for TEM observation.

They were irradiated with 200 and 400 keV self-ions ($V^+$) from $1.0 \times 10^{14}$ to $1.0 \times 10^{16}$ ions/m$^2$ at room temperature using a 400 kV Cockcroft–Walton type accelerator in the University of Tokyo, Tokai site. As very few defect clusters were detected by TEM observation of self-ion irradiated specimens up to the irradiation dose of $1.0 \times 10^{16}$ ions/m$^2$, thin foil samples were irradiated with heavier gold and xenon ions up to $1.0 \times 10^{16}$ ions/m$^2$ at room temperature to get a high density of displacement damage in cascades. Table 1 summarizes the irradiation conditions. Fig. 1 shows the calculated depth distributions of displacement damage in vanadium irradiated with 50–400 keV $Au^+$ ions. Damage production ends at 100 nm for 400 keV $Au^+$, so

Table 1
Irradiation conditions of heavy ion irradiation to thin foils of vanadium

| Target material | Irradiating ion | Ion energy (keV) | Ion fluence (ions/m$^2$) | Temperature |
|---|---|---|---|---|
| Thin foils of vanadium | $V^+$(self-ion) | 200 | $1.0 \times 10^{14}$–$1.0 \times 10^{16}$ | 300 K |
| | | 400 | $1.0 \times 10^{16}$ | |
| | $Xe^+$ | 200 | | |
| | | 250 | | |
| | | 300 | | |
| | | 400 | | |
| | $Au^+$ | 50 | | |
| | | 100 | | |
| | | 200 | | |
| | | 300 | | |
| | | 400 | | |

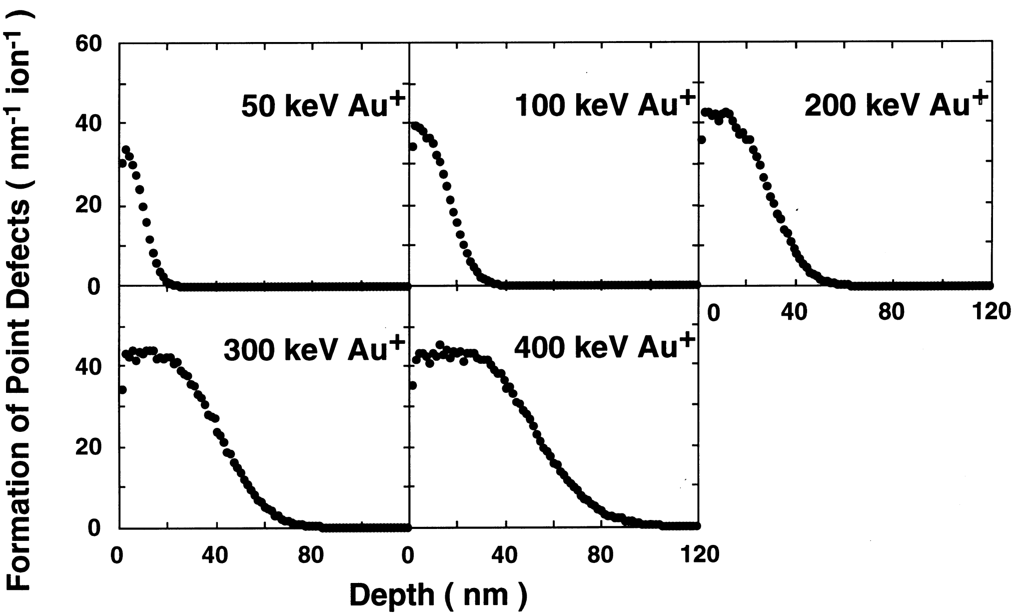


Fig. 1. Depth distribution of damage energy transfer in vanadium irradiated with $Au^+$ ions calculated by the TRIM 92 code.

we observed defect cluster formation in thin foils of vanadium less than 80 nm. These foil samples were also irradiated with 250–400 keV $Xe^+$ ions up to $1 \times 10^{16}$ ions/$m^2$ at room temperature. The irradiated specimens were observed by a TEM operated at 200 keV.

## 3. Results and discussion

### *3.1. Defect cluster formation by $Au^+$ ion irradiation*

$Au^+$ and $Xe^+$ ion irradiation produced visible defect clusters in thin foils of vanadium, whereas very few clusters were detected in the specimens irradiated with 200 and 400 keV self-ions up to $1 \times 10^{16}$ ions/$m^2$. Fig. 2 shows the density of observed defect images per unit area as a function of foil thickness under irradiation with $Au^+$ and $Xe^+$ ions. It was clearly observed that higher energy Au+ ion irradiation produced a higher density of clusters. Cluster density in the specimens irradiated with $Xe^+$ ions was one order of magnitude less than the $Au^+$ ion cases. In the specimens irradiated with 200 keV and higher energy of $Au^+$ ions, density of observed clusters was found to decrease with specimen thickness. In the case of 200 keV $Au^+$ ion irradiation, density of the defect clusters shows saturation at around 40 nm, whereas displacement damage depth extends up to 50 nm as shown in Fig. 1. The average size of observed defect clusters in vanadium thin foils irradiated with $Au^+$ and $Xe^+$ ions is found to be within a narrow band between 1.7–2.5 nm as shown in Fig. 3.

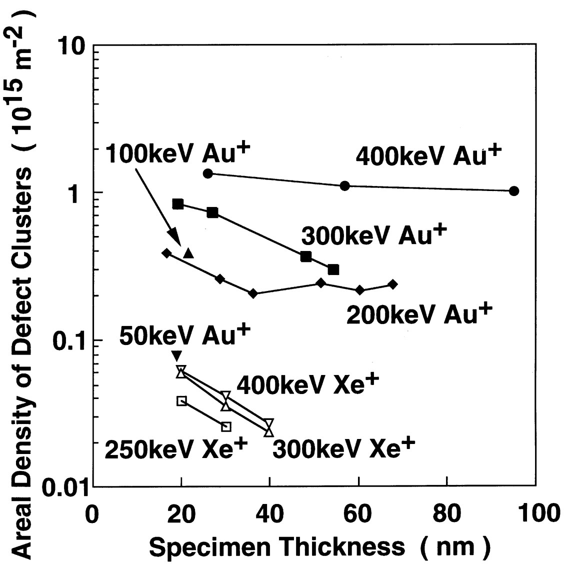


Fig. 2. Areal density of the defect clusters observed in vanadium thin foils irradiated with $Au^+$ and $Xe^+$ ions at room temperature as a function of specimen thickness.

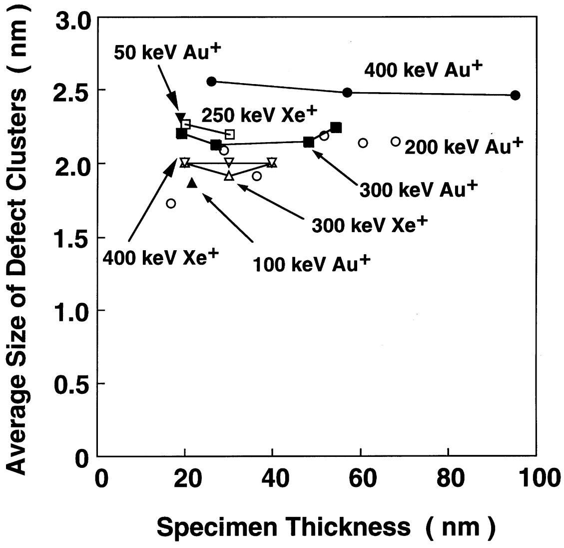


Fig. 3. Average size of the defect clusters observed in vanadium thin foils irradiated with $Au^+$ and $Xe^+$ ions at room temperature as a function of specimen thickness.

Most of defect clusters observed in regions less than 30 nm thick are considered to be of vacancy type, where interstitials can easily escape to the specimen surface. In the thicker region of the specimens, interstitials annihilate vacancy clusters and also form interstitial clusters. It should be noted that most defect clusters were isolated, indicating no evidence of sub-cascade effects in vanadium. In thin foils of FCC metals such as gold, groups of vacancy clusters are observed after irradiation with heavy ions, and the number of vacancy clusters in a group is strongly dependent on ion energy and PKA energy under neutron irradiation [9].

Table 2 compares the defect yield obtained in 20 nm thick foils of vanadium under heavy ion irradiation with the reported yields in other BCC metal [6–8]. Here, defect yield is defined by the observed 'collapsed vacancy clusters' per incident ion observed with TEM. This value is dependent both on ion mass and energy. It should be noted that the measured defect yield in vanadium is lower than those in other BCC metals with larger atomic masses such as Fe, Mo and W.

### *3.2. Correlation between damage energy transfer and defect cluster formation*

To help understanding mechanisms of vacancy cluster formation, the density of the observed defect clusters per damage energy deposition averaged in 20 nm thick foils of vanadium is plotted in Fig. 4. Formation of defect clusters decreases with specimens thickness. However, in the very thin region of the specimens, defect cluster density increases almost linearly with ion energy. This supports the idea that clusters observed in the thin

Table 2
Defect yield in vanadium foils of 20 nm in thickness irradiated with heavy ions to $1.0 \times 10^{16}$ ions/m$^2$ at room temperature

| Ion (keV) | | Target | Defect yield | Reference |
|---|---|---|---|---|
| 200 | V | V | 0.00 | This work |
| 400 | V | V | 0.00 | |
| 200 | Xe | V | $<$ 0.001 | |
| 250 | Xe | V | 0.0039 | |
| 300 | Xe | V | 0.0059 | |
| 400 | Xe | V | 0.0063 | |
| 50 | Au | V | 0.0079 | |
| 100 | Au | V | 0.036 | |
| 200 | Au | V | 0.035 | |
| 300 | Au | V | 0.088 | |
| 400 | Au | V | 0.15 | |
| 80 | Fe | Fe | $\sim$ 0.001 | [7] |
| 80 | Xe | Fe | 0.0725 | |
| 80 | W | Fe | 0.195 | |
| 60 | Mo | (0 1 1)Mo | 0.12 | [8] |
| 60 | Sb | (0 1 1)Mo | 0.15 | |
| 60 | Xe | (0 1 1)Mo | 0.20 | |
| 120 | Sb | (0 1 1)Mo | 0.29 | |
| 180 | Sb | (0 1 1)Mo | 0.35 | |
| 60 | Au | W | 0.1 | [6] |

Other reported defect yield data in BCC metals are also shown. Defect yield is defined by the observed areal density of defect clusters per ion fluence.

specimen region are vacancy clusters formed by the high energy density in cascades. It appears from Fig. 4 that formation of the vacancy clusters under Au$^+$ ion irradiation seems to have a critical value of about 120 keV.

Fig. 5 shows calculated damage energy in a very thin region of the specimens for all the irradiation conditions in the present experiments. The minimum energy transfer to form vacancy clusters can be estimated from the damage energy deposition to be 4.5 keV/nm/ion. To prove this idea, Xe$^+$ ion irradiation experiments were performed between 200 and 400 keV. A low density of defect clusters was observed in the very thin region of

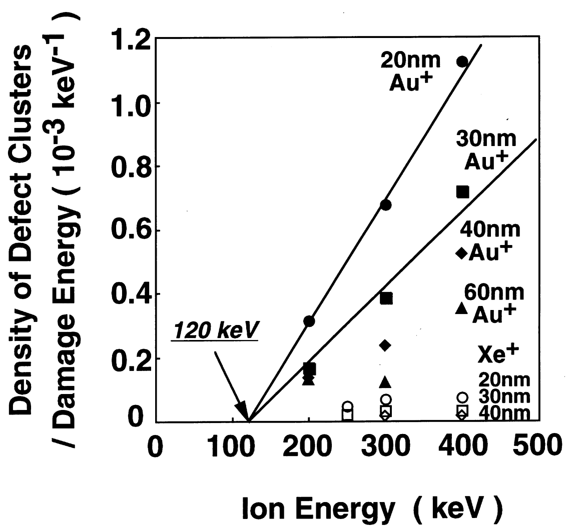


Fig. 4. Ion energy dependence of defect cluster density per damage energy in thin foils of vanadium irradiated with Au$^+$ and Xe$^+$ ions at room temperature.

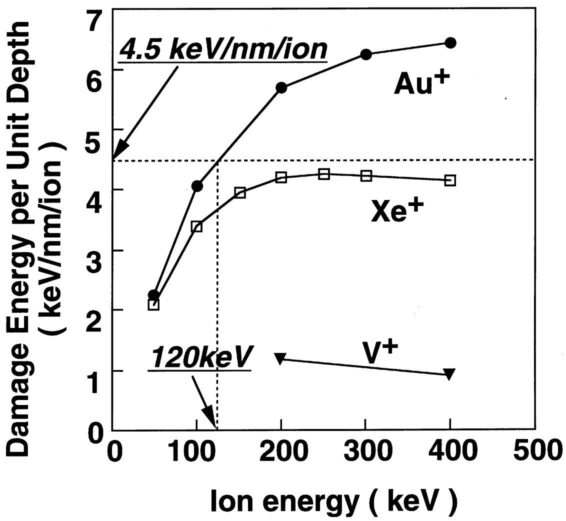


Fig. 5. Calculated damage energy in vanadium of 20 nm thickness irradiated with V$^+$, Xe$^+$ and Au$^+$ ions by the TRIM 92 code. 120 keV Au$^+$ ions introduces damage energy transfer of 4.5 keV/nm/ion in a vanadium foil of 20 nm thickness.

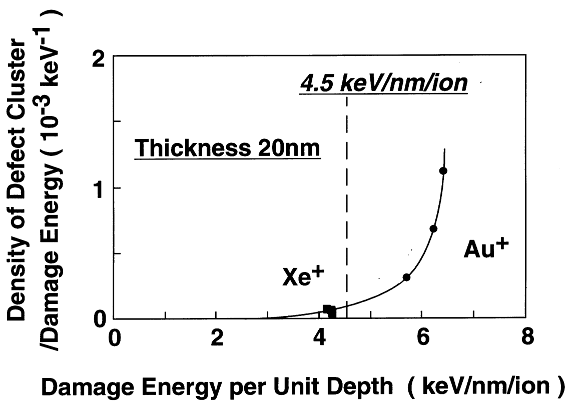


Fig. 6. Density of defect clusters per damage energy as a function of damage energy density in vanadium foil of 20 nm thickness.

the specimens irradiated with $Xe^+$ ions between 200 and 400 keV as shown in Fig. 2.

Fig. 6 shows again the density of defect clusters per damage energy at a foil thickness of 20 nm. From these results, 4.5 keV/nm/ion appears to be a threshold value for formation of vacancy clusters which can be observed by TEM in thin foils of vanadium, but the probability of vacancy cluster formation is dependent on damage energy density.

Irradiation experiments need to be conducted at temperatures below which vacancies are not mobile in vanadium in order to further develop our understanding of the dynamics of vacancy cluster formation.

## 4. Summary

The effect of cascade damage on defect cluster formation in vanadium was investigated using heavy ion irradiation. In the very thin region of the specimens, vacancy clusters were formed under gold ion irradiation, while very few clusters were detected in the specimens irradiated with 200 and 400 keV self-ions up to $1 \times 10^{16}$ ions/m$^2$. The density of vacancy clusters was found to be strongly dependent on ion energy. Only above a critical kinetic energy transfer density value of 4.5 keV/nm/ion in vanadium, visible vacancy clusters were formed from the cascade damage. In such a thin region, interstitials can escape to the specimen surface without annihilating the vacancy clusters.

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