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Journal of Nuclear Materials 271&272 (1999) 68–72

journal of nuclear materials

High energy cascades in gold as studied by high energy self-ion irradiation

N. Sekimura [a,*], Y. Kanzaki [a], N. Ohtake [a], J. Saeki [a], Y. Shirao [a], S. Ishino [b], T. Iwata [c], A. Iwase [c], R. Tanaka [d]

[a] *Department of Quantum Engineering and Systems Science, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113, Japan*
[b] *Department of Nuclear Engineering, Tokai University, 1117 Kitakaname, Hiratsuka, Kanagawa 259-12, Japan*
[c] *Tokai Establishment, Japan Atomic Energy Research Institute, 2-4 Shirakata-Shirane, Tokai-mura, Naka-gun, Ibaraki 319-11, Japan*
[d] *Takasaki Establishment, Japan Atomic Energy Research Institute, 1233 Watanuki-cho, Takasaki, Japan*

Abstract

Primary knock-on atom (PKA) energy spectrum extends up to several hundreds of keV in fusion reactor materials irradiated with 14 MeV neutrons. When we are going to evaluate materials behavior in an expected d-Li type intense neutron source, fundamental knowledge on effects of high energy PKA on formation of cascade damage, microstructural evolution and properties of irradiated materials is required. In this study, 170 MeV self-ion irradiation of thin foils of gold were performed to estimate effects of very high energy PKA on formation of defect clusters by cascade damage and its interactions. Defect clusters of vacancy type were observed in the thin foils of gold irradiated to 5×10^{13}–1×10^{15} ions/m^2. In the case of irradiation with 170 MeV self-ions of which the projected range exists at 6.3 μm from the ion incident surface, films of gold, 1.1, 3.0, 4.3, 6.3 and 7.2 μm in thickness, were placed in front of the 50 nm thick specimens to change PKA energy spectrum within the specimens. The number of vacancy clusters within a cluster group formed by a PKA varied with the thickness of gold film. High energy PKA was found to increase the number of defect clusters. However, size distributions of defect clusters were not strongly dependent on PKA energies. Interactions of high energy cascades result in the appearance of new defect clusters near the existing defect cluster groups in the higher dose range. Dose dependence of defect cluster density was similar to that observed in 14 MeV and fission neutron irradiated specimens. The contribution of PKA higher than 400 keV to the interactions of cascades is estimated from the calculated PKA energy spectrum and low energy self-ion irradiation data.

1. Introduction

Cascade collision sequences initiated from a high energy primary knock-on atom (PKA) produce a high density of point-defects, which can result in the formation of vacancy clusters in the core of the cascade. The PKA energy spectrum extends up to several hundreds of keV in fusion reactor materials irradiated with 14 MeV neutrons, and the cascade damage structure is strongly dependent on PKA energy.

We have been studying PKA energy dependence of cascade damage formation by using thin foils of gold. As self-ion irradiation up to several hundred keV can be regarded as a monoenergetic PKA, neutron irradiation effects can be understood by integrating cascade damage formation under the PKA energy spectrum. We have made a model to describe defect cluster formation in thin foils of gold under 14 MeV and fission neutron irradiation from self-ion irradiation data and calculated PKA energy spectrum [1]. This model can successfully estimate the distribution of the number of defect clusters under 14 MeV and fission neutron irradiation which are produced from a PKA in the low fluence region where defect clusters are not affected by other cascades.

* Corresponding author. Tel.: +81-3 38122111; fax: +81-3 3812 1794; e-mail: sekimura@q.t.u-tokyo.ac.jp.

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The interaction of cascades produces new vacancy clusters near the pre-existing groups of clusters, resulting in super-linear dose dependence of the density of vacancy clusters [2]. By comparing accumulation processes of vacancy clusters in thin foils of gold under three irradiation conditions with different PKA energy spectrum, we could find a minimum PKA energy to convert invisible vacancy agglomerates to visible clusters as viewed by a transmission electron microscope (TEM).

The effect of high energy cascades also needs clarifying in estimating microstructural evolution and property changes in fusion reactor materials. In evaluating irradiation performance of the candidate materials by an expected d-Li type intense neutron source which has high energy tails of neutrons, we should have fundamental knowledge on effects of high energy PKA on formation of cascade damage.

In this study, the effect of high energy cascades in thin foils of gold on defect cluster formation is investigated using 170 MeV self-ions. There are high energy components of PKA in the specimens irradiated with energetic heavy ions. It is also well known that the PKA energy spectrum changes with the depth from the incident ion surface. By placing films of gold in front of the thin foil specimens for TEM observation, we can change the PKA energy spectrum in the thin foil. The effect of high energy cascades on vacancy cluster formation is also investigated.

2. Experimental

Thin foils of 99.99% pure gold were annealed at 973 K for 3.6 ks and electro-polished for microstructural observation by TEM. These foil specimens were irradiated with 170 MeV gold ions at room temperature using a 20 MV tandem accelerator in the Tokai Establishment of Japan Atomic Energy Research Institute. Here we mainly discuss the results at 1.0×10^{14} ions/m^2, and only limited data are reported to discuss the effect of high energy PKA on defect clusters formation. Details of dose dependence study from 5×10^{13} to 1×10^{15} ions/m^2 will be published elsewhere [3].

Peak displacement damage depth in gold by 170 MeV self-ions without the degradation film exists at 6.3 μm from the ion incident surface. To change PKA energy spectrum within the thin foil specimens, films of gold, 1.1, 3.0, 4.3, 6.3 and 7.2 μm in thickness, were placed in front of the specimens. Fig. 1 shows the recoil energy spectra calculated by the TRIM-89 code for the five different film thicknesses for 170 MeV gold ions. It should be noted that when the gold film of 6.3 μm in thickness is placed, relative fraction of PKA higher than 200 keV is larger than other cases.

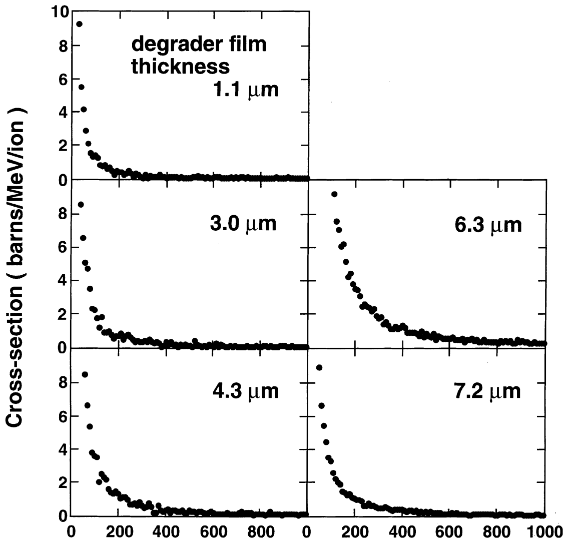

Fig. 1. Recoil energy spectra for five different film thicknesses for energy degradation of 170 MeV Au^+ ions calculated by the TRIM code.

Irradiated samples were observed by TEM operated at 200 kV. Typical foil thickness for TEM observation was less than 80 nm.

3. Results

3.1. Number of vacancy clusters in a cluster group

Fig. 2 shows the micrographs of defect clusters observed in thin foil specimens irradiated with high energy self-ions to 1.0×10^{14} ions/m^2 passing through the degrader films. All of these clusters are considered to be of vacancy type, because intersitials can easily escape to the specimen surface at room temperature. Groups containing many vacancy clusters are observed. It was relatively easy to separate groups of defect clusters even at the energy degrader thickness of 6.3 μm, and the observed defect clusters in a cluster group are considered to come from sub-division of cascades from a PKA.

Fig. 3 shows the distributions of the number of vacancy clusters in a group of clusters. The number of vacancy clusters within a cluster group varies with the thickness of gold degrader films. High energy PKAs are found to increase number of defect clusters. Groups containing higher numbers of clusters are observed especially at the gold film thickness of 6.3 μm. The maximum number of defect clusters in one group of clusters is found to be 14.

Size distributions of defect clusters are not strongly dependent on PKA energy spectrum as shown in Fig. 4. The average size of vacancy clusters is around 2.5 nm in

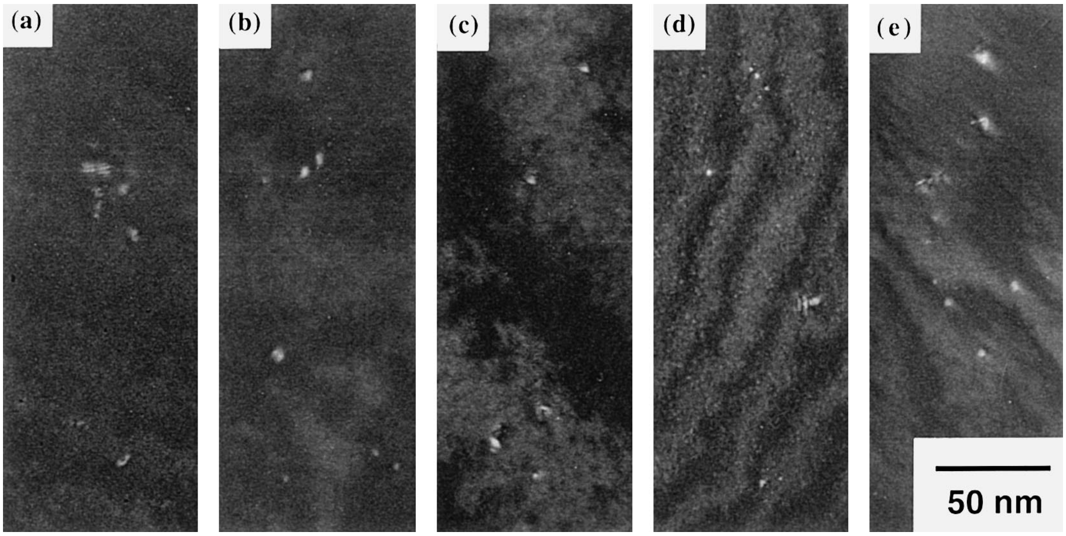

Fig. 2. Micrographs of defect clusters observed in thin foil specimens of gold irradiated with 170 MeV Au^+ ions to 1.0 × 1014 ions/m^2 passing through the degrader films of (a) 1.1, (b) 3.0, (c) 4.3, (d) 6.3 and (e)7.2 μm in thickness.

all the cases. It should be noted that the fraction of smaller clusters at 6.3 μm is larger than for other case.

3.2. Dose dependence of vacancy cluster formation

Fig. 5 shows the dose dependence of total density of vacancy clusters in thin foils of gold irradiated with degraded 170 MeV self-ions by 1.1, 3.0 and 6.3 μm gold films. At 1.1 μm, defect cluster density increases linearly with irradiation dose up to 1.0×10^{15} ions/m^2. However, at 3.0 μm, the defect cluster density shows deviation from the linear dose dependence. This is most clearly observed at 6.3 μm, where the deviation from the linear dose dependence starts from a lower dose.

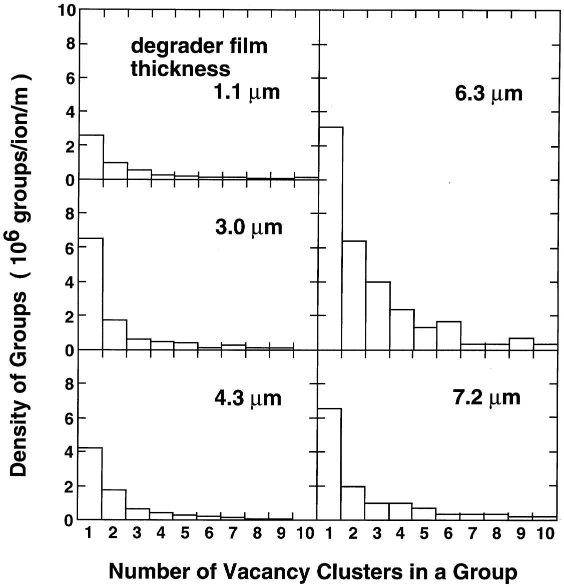

Fig. 3. Distributions of the number of vacancy clusters in a group of clusters in thin foils of gold irradiated with 170 MeV Au^+ ions to 1.0×10^{14} ions/m^2 passing through the degrader films of 1.1, 3.0, 4.3, 6.3 and 7.2 μm thickness.

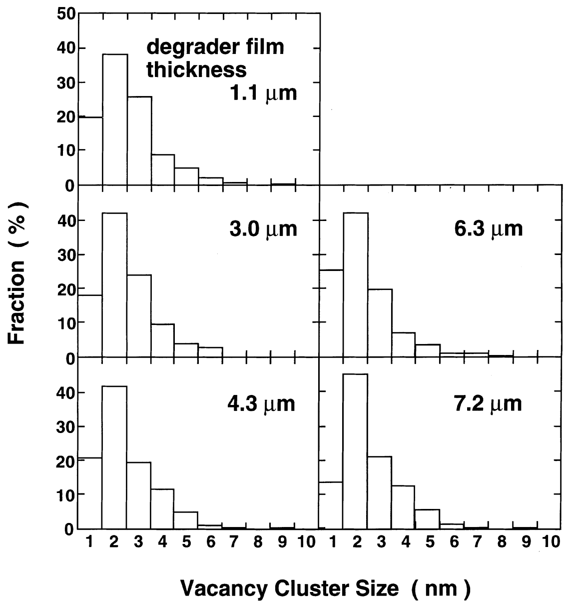

Fig. 4. Size distributions of vacancy clusters in a group of clusters in thin foils of gold irradiated with 170 MeV Au^+ ions to 1.0×10^{14} ions/m^2 passing through the degrader films of 1.1, 3.0, 4.3, 6.3 and 7.2 μm thickness.

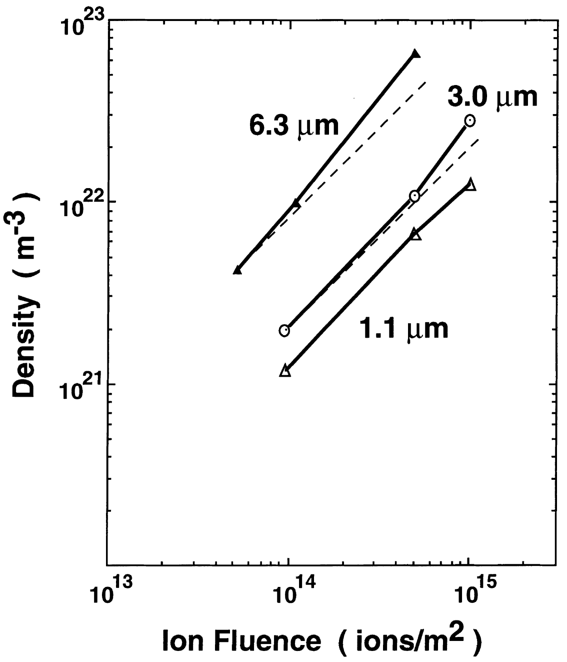

Fig. 5. Dose dependence of vacancy cluster density in thin foils of gold irradiated with degraded 170 MeV self-ions by 1.1, 3.0 and 6.3 mm degrader films.

We consider that the deviation arises from cascade interactions at higher doses. The deviation from the linear dose dependence in irradiated thin foils of gold is generally observed under 14 MeV neutron irradiation [4–6], fast neutron irradiation in the YAYOI reactor and also 21 MeV self-ion irradiation without degrader foils [2].

In the previous paper [2], we proposed that cascade damage in thin foils of gold introduces invisible agglomeration of vacancies in addition to the visible vacancy clusters. At higher dose, they are converted to visible but small clusters by interaction with nearby cascades.

These processes are considered to be dependent on PKA energy. Primary recoil atoms with energy >165 keV are assumed to be responsible for the deviation from the linear dose dependence. The minimum interaction energy of primary recoil atoms is estimated by considering interaction distance between cascades in three different irradiations [2]. Only a small fraction of high energy recoil atoms larger than this critical energy is effective in producing the non-linear dose dependence.

4. Discussion

A high energy PKA can produce larger numbers of clusters in a cluster group, since sub-cascade formation in gold can effectively produce vacancy clusters. However, it is difficult to separate the effect of cascade overlap from the direct effect of PKA to form larger numbers of clusters.

In the case of 6.3 μm of the gold degrader film, large numbers of vacancy clusters in a group were observed. However, dose dependence of vacancy cluster density starts to deviate from the linear dose dependence even at an ion fluence of 1.0×10^{14} ions/m^2 as shown in Fig. 5, and the fraction of smaller size vacancy clusters seems to be larger than other cases as shown in Fig. 4. In the previous paper, it was shown that smaller clusters were formed in thin foils of gold near the existing cluster group after dose dependence of the cluster density started to deviate from the linear dose dependence [2]. Therefore the observed distribution of the number of clusters in a group and cluster size in the case of 6.3 μm of gold degrader film are considered to be affected by other high energy cascades.

In the case of degrader film thickness of 1.1 μm, the density of vacancy clusters does not show clear deviation from the linear dose dependence. There is only small fraction of PKAs with higher than 400 keV, for example, compared with 6.3 μm case.

Fig. 6 compares estimated distributions of the number of vacancy clusters in a cluster group based on the self-ion irradiation data up to 400 keV [1] with the present data from the thin foil samples irradiated with degraded 170 MeV self-ions by 1.1, 3.0 and 6.3 μm gold films. This indicates that higher numbers of defect clusters cannot be estimated by cascade damage formation observed at 400 keV and the lower energy region. In the case of low dose irradiation of gold with

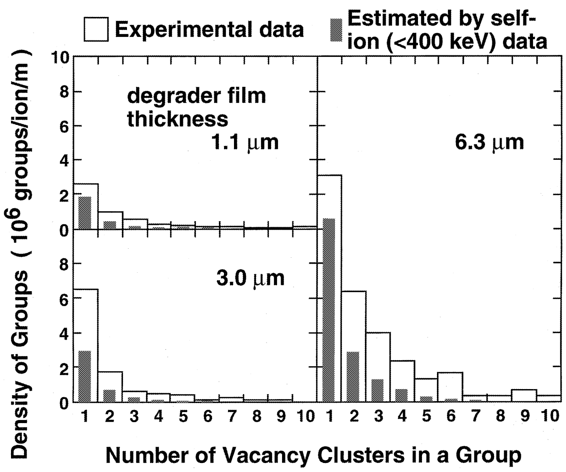

Fig. 6. Comparison of estimated and measured distribution of the number of defect clusters in a group in a thin TEM foil of gold. In front of the TEM specimens, gold films of 1.1, 3.0 and 6.3 μm in thickness are placed to degrade 170 MeV self-ions. Irradiation fluence is 1.0×10^{14} ions/m^2 in all cases.

14 MeV neutrons and fission neutrons, vacancy cluster density can be successfully estimated by self-ion data up to 400 keV. This covers the whole range of the PKA energy spectrum in these neutron irradiation conditions [1]. Again, larger numbers of vacancy clusters at 6.3 μm comes from both direct effect of high energy PKA and cascade overlap effects. However, single high energy PKA effects on vacancy cluster formation are detected in the case of thinner degrader film thicknesses.

5. Summary

Thin foils of gold are irradiated with degraded high energy self-ions. Effects of the recoil energy spectrum on the formation of cascade damage and vacancy clusters are investigated by changing the thickness of degrader films. Cascade damage by high energy recoil atoms produces groups of clusters with higher numbers of vacancy clusters. Interaction of cascades from high energy recoils causes the shift to non-linear dose dependence of vacancy cluster density.

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