Available online at www.sciencedirect.com
SCIENCE DIRECT®
Journal of Nuclear Materials 329–333 (2004) 1185–1189
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Cascade and subcascade structure in fission neutron irradiated fcc metals and their correlation to fusion neutron irradiation

Y. Satoh [a,*], M. Tsukada [a], H. Matsui [a], T. Yoshiie [b]

[a] *Institute for Materials Research, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980-8577, Japan*
[b] *Research Reactor Institute, Kyoto University, Noda, Kumatori-cho, Sennan-gun 590-0496, Japan*

Abstract

Thin foil specimens of Cu and Au were irradiated with fission neutrons at 285–295 K to neutron fluences of 7×10^{21} $\mathrm{n\,m^{-2}}$ (>0.1 MeV), using the Kyoto University Reactor. The structure of cascades and subcascades was examined by fitting the experimentally observed distribution of vacancy-type defect clusters to the calculated primary recoil energy spectrum, and was compared directly with that for fusion neutron irradiation. In Cu, 70% of cascades consist of only one defect cluster and 2% of more than four clusters. This is in contrast to larger cascades produced by 14 MeV neutrons that contain more than 10 clusters and extend 60 nm in diameter. The subcascade energy was 18 and 15 keV for Cu and Au, respectively. These values are about 30–60% higher than those obtained for fusion neutron irradiation.

1. Introduction

The defect structure development under high energy neutron irradiation starts with a large energy transfer from an incident neutron to a primary knock-on atom (PKA). The subsequent displacement collision cascade produces highly localized point defects. The PKA with a large recoil energy is known to divide into several damage zones called subcascades. After point defect reactions within cascades and subcascades, defect clusters and free point defects that have escaped from cascade damage zones remain, which leads to defect structural development.

In D–T fusion neutron irradiation experiments using the Rotating Target Neutron Source (RTNS-II), Kiritani and co-workers showed for several fcc metals that stacking fault tetrahedra (SFTs) are formed directly from subcascades [1,2]. The subcascade can be efficiently detected in thin foil specimens (that were thinned by electropolishing into the thickness suitable for direct observation with TEM) irradiated at relatively lower temperatures, such as room temperature. This is because high concentration of vacancies in subcascades form stable SFTs, and interstitial atoms escape promptly at specimen surfaces without seriously affecting vacancy clusters formed from subcascades. The analysis of cascade and subcascade structure based on the calculated primary recoil energy spectrum led to models of the PKA energy dependence of cascade structure ('recoil energy spectrum analysis') [2,3].

In the present study, we performed fission neutron irradiation of thin foil specimens at room temperature, in order to compare directly with the above fusion neutron irradiation data. The aim of this study is to examine a fission-fusion correlation of cascade and subcascade structure and the direct formation of point defect clusters from subcascades. We also deduced the PKA energy dependence of the subcascade structure, and compared with that obtained by fusion neutron irradiation. The present paper reports results on Cu and Au, which are typical materials of separated subcascades and closely spaced subcascades, respectively [2].

* Corresponding author. Tel.: +81-022 215 2067; fax: +81-022 215 2066.
E-mail address: ysatoh@imr.tohoku.ac.jp (Y. Satoh).

0022-3115/$ - see front matter
doi:10.1016/j.jnucmat.2004.04.265

2. Experimental procedure

Thin foil specimens of Au, Ag, Cu, and Ni were irradiated at 285–295 K, using the low temperature loop (LTL) and the Hydraulic Conveyor (Hyd) facilities at the Kyoto University Reactor (KUR), 5 MW light water reactor. In the LTL irradiation, the temperature of the specimen capsules was monitored and controlled at 290 K by supplying cold helium gas and using an electric heater. In the Hyd irradiation, the specimen capsule had no special apparatus for precise temperature control but was in the coolant water of the reactor. The coolant temperature increases gradually from the ambient temperature to about 320 K during one cycle operation (i.e., three days) at the normal power, 5 MW. In the present irradiation experiments at a reduced power of 0.1 MW, the coolant temperature stayed at 285–295 K. We made two irradiation runs with the LTL at three flux positions for each, and two irradiation runs with the Hyd.

The neutron fluence was obtained by measuring ^{58}Co production in Ni specimens; the cross-section of ^{58}Ni(n,p)^{58}Co reaction in the LTL and the Hyd was obtained from the JENDL-3.2 nuclear data library [4] and the neutron spectrum of the respective facilities in KUR. The neutron spectrum is soft at the LTL facility because it is located farther from the reactor core: the fraction of fast neutrons (>0.1 MeV) to all neutrons is 10.7% and 15.0% at the LTL and the Hyd, respectively. In this paper the cross-section is defined with respect to all neutrons unless specifically noted. In Cu for example, the maximum damage rate in the LTL facility was 2×10^{-9} dpa/s, and that is almost equal to that in the Hyd facility at 0.1 MW operation. This is about an order of magnitude larger than the damage rate at RTNS-II.

The microstructure was observed with 200 kV TEM utilizing weak-beam dark-field images ($\mathbf{g} = 002$). The observation condition was the same as that for specimens irradiated with RTNS-II.

3. Results and discussion

3.1. Typical microstructures and defect accumulation

Fig. 1 shows typical defect structures observed in Cu and Au thin foil specimens after fission neutron irradiation at the LTL facility. TEM image contrast suggests that the majority of the defect clusters are SFTs of vacancy-type, which is a similar structure as in fusion neutron irradiation. They are thought to form directly from the high concentration of vacancies in cascades or subcascades.

Fig. 2 shows variation of the number density of defect clusters with the fluence of fast neutrons (>0.1 MeV). The number density of subcascade defects increases in proportion to the neutron fluence. The factor corresponds to the apparent cross-section for producing subcascade defect clusters; 0.15 and 0.08 barns for Cu and Au, respectively. The results on the fusion neutron irradiation [2] are also shown in the figure. They indicate that the number of defect clusters observed for a very low fluence is much less than that extrapolated from their proportional increase at higher fluence. This observation was interpreted as follows: some of defect clusters directly formed from subcascades stay invisible to TEM (latent defects), and they become visible by impact effects of large cascades formed in the vicinity of the latent defects [2]. In fission neutron irradiated Cu, the number density of defect clusters at the lowest fluence deviates from the proportional increase. This may suggest the presence of latent defect clusters in the fission neutron irradiation, but this needs further examination.

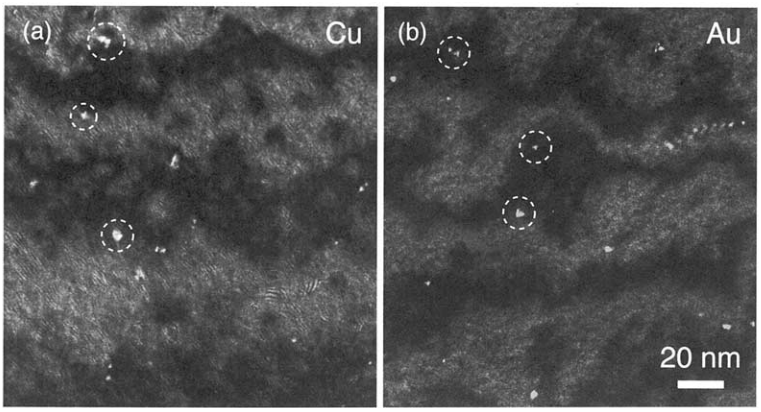

Fig. 1. Typical defect structure observed in thin foil specimens of (a) Cu and (b) Au, irradiated with fission neutrons at the LTL of the KUR. The irradiation was at 295 K to the fluence of 3.5×10^{21} n m^{-2} (>0.1 MeV). Each circle shows a group of defect clusters that is formed from a single PKA.

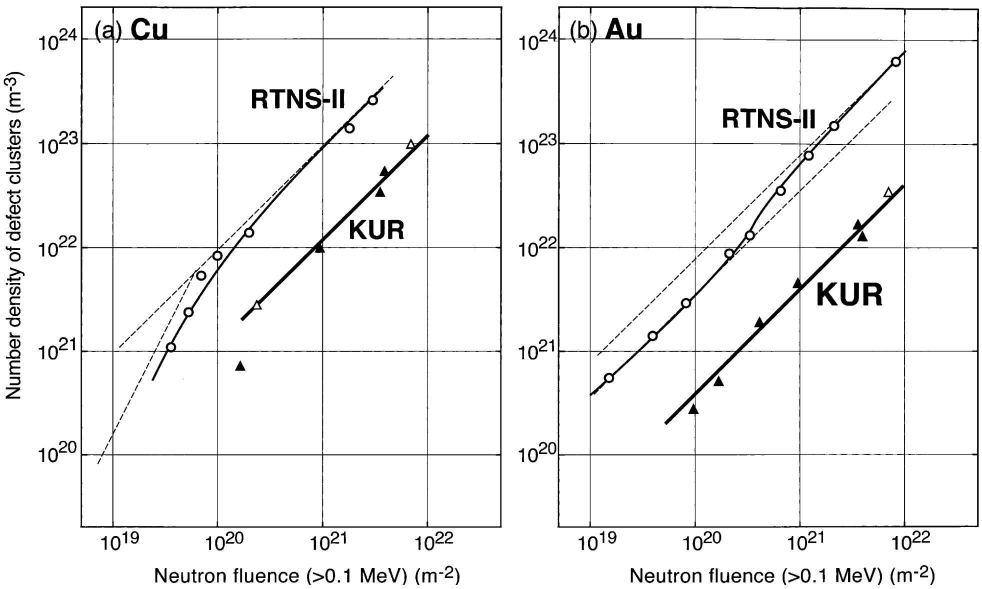

Fig. 2. Variation of the number density of defect clusters in (a) Cu and (b) Au irradiated as thin foils at room temperature by fission neutrons (KUR) and fusion neutrons (RTNS-II) [2,3]. Full and open triangles correspond to the results of the LTL and the Hyd of the KUR, respectively.

3.2. Average subcascade energy

An average damage energy assigned per subcascade defect is estimated in a similar method as in the analysis of fusion neutron irradiation experiments [2]. First we estimated the damage energy $E_D(E_{PKA})$ which is the part of the primary recoil energy E_{PKA} to produce structural damage on lattice atoms involved in cascade collision, which is estimated according to the theory by Lindhard et al. [5]. Then we consider division of the damage energy E_D into subcascades, assuming a threshold energy for subcascade defect formation E_{SC}, in an analogous procedure to the Kinchin–Pease model for Frenkel pair production. Namely, the number of subcascades N_{SC} produced from the PKA with the energy E_{PKA} is

$$N_{SC} = \begin{cases} 0 & (E_D < E_{SC}), \\ 1 & (E_{SC} \leqslant E_D \leqslant 2E_{SC}), \\ E_D/2E_{SC} & (2E_{SC} < E_D). \end{cases} \quad (1)$$

Then the cross-section for subcascade defect formation is expressed as

$$\sigma_{SC} = \int_0^{E_{max}} N_{SC}(E_{PKA}) \frac{d\sigma}{dE_{PKA}} dE_{PKA}, \quad (2)$$

where $d\sigma/dE_{PKA}$ is the differential cross-section for incident fission neutrons to produce collision to transfer an energy E_{PKA}: we adopted the cross-section calculated with SPECTER code [6].

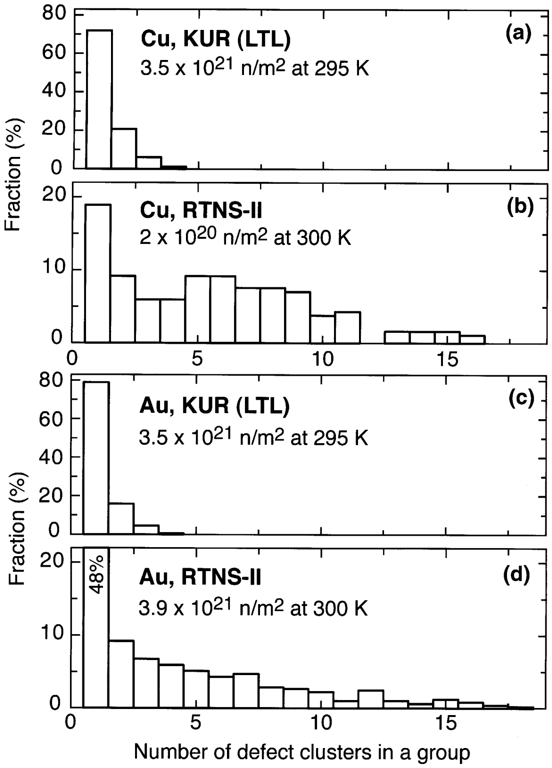

Fig. 3. Population of vacancy cluster groups plotted against the number of clusters contained in a group. The distributions in Cu and Au are compared between fission and fusion neutron irradiation. The results on the RTNS-II are taken from Refs. [2,3].

The average subcascade energy was estimated by varying the threshold energy E_{SC} in Eqs. (1) and (2) so that σ_{SC} corresponds to the experimental cross-section obtained in Section 3.1. The subcascade energy was 18 keV for Cu and 15 keV for Au. These values are higher than obtained for fusion neutron irradiation: 14 keV for Cu and 9 keV for Au. These results suggest that the efficiency of direct formation of vacancy clusters from subcascades is lower for fission neutrons than for fusion neutrons, even though the two irradiation environments are normalized to the primary recoil energy spectrum and the damage energy. A possible reason for the difference is that some fraction of PKAs with energy larger than E_{SC} will fail to form a defect cluster. This is expected in the relatively low recoil energy range, and will be more critical for fission neutron irradiation than for 14 MeV fusion neutrons.

3.3. Recoil energy spectrum analysis

In this section, we examine groups of defect clusters formed from a single PKA. As already shown by circles in Fig. 1, groups of defect clusters can be clearly recognized because the separation between subcascades is small even in Cu. This is in contrast to widely separated subcascades observed in fusion neutron irradiated Cu, in which grouping of defect clusters was made with help of the PKA energy spectrum [3].

Fig. 3 is the population histograms of observed defect cluster groups plotted against the number of clusters contained in a group. Both in Cu and Au, a large fraction of the defect clusters is isolated, and formation of subcascades is observed with only a low probability. For example in Cu, 70% of cascades consist of only one defect cluster and 2% more than four clusters. The size of defect group is smaller than 10 nm, even though it contains three defect clusters. This is in contrast to larger cascades produced by 14 MeV neutrons that contain more than 10 clusters and extend 60 nm in diameter [3].

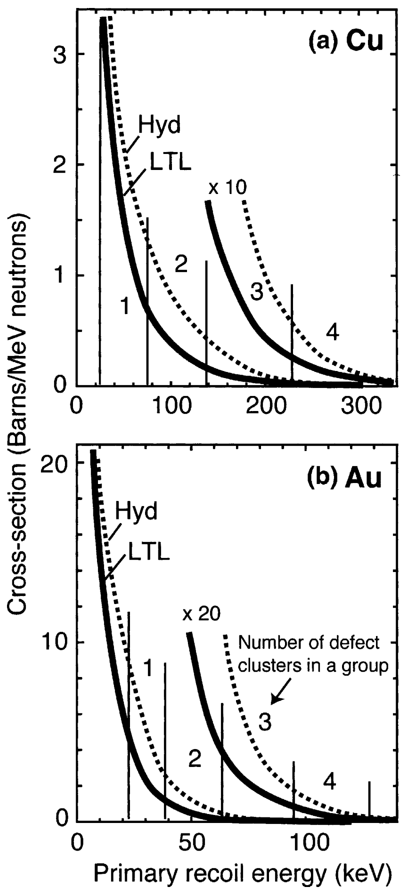

Fig. 4. Primary recoil energy spectrum in (a) Cu and (b) Au for fission neutron irradiation at the LTL and the Hyd of the KUR, and fitting of observed numbers of defect clusters in a group shown in Fig. 3(a) and (c).

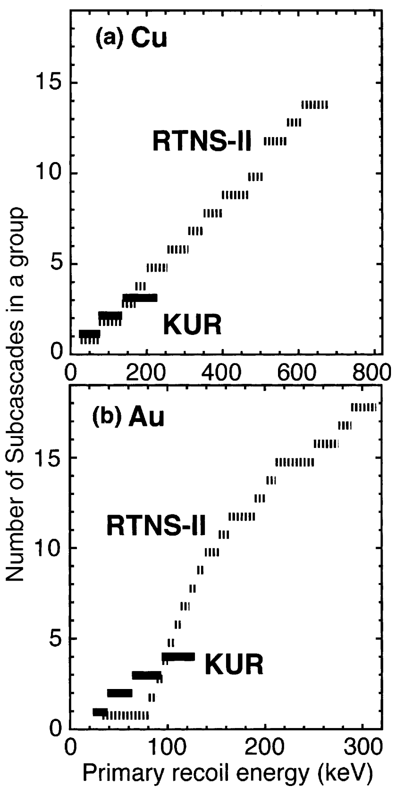

Fig. 5. Variation of the number of defect clusters in a group with the primary recoil energy: (a) Cu and (b) Au. The solid lines and broken lines are obtained from fission neutron irradiation at the LTL and fusion neutron irradiation at the RTNS-II, respectively.

All these features of spatial distribution of subcascades reflect the lower PKA energy in fission neutron irradiation.

We simply assumed in Fig. 4 that the groups with higher number of subcascades have been formed from PKA with higher energy. Then we obtain the relation between the PKA energy and the number of subcascade defects produced, as shown in Fig. 5. The dotted lines show the relation obtained from fusion neutron irradiation [2], which covers a wider primary recoil energy range; and the solid lines obtained from fission neutron irradiation, which gives lower energies. Cu shows a good correspondence in the lower energy range between fission and fusion neutron irradiation, while a discrepancy is observed in Au. Namely, the energy range from which a single isolated cascade results is much wider, 25–75 keV in fusion neutron irradiated Au. The results for fission neutrons are more reliable in the lower energy range, because they are not affected by higher energy recoils. A possible explanation for the discrepancy is an overestimate of isolated defect clusters in the grouping procedure of defect clusters formed by fusion neutron irradiation.

Acknowledgements

The authors are grateful to Professor M. Okada, Dr Q. Xu, and Mr K. Miyata at the Kyoto University Reactor for technical support in neutron irradiation experiments.

References

[1] M. Kiritani, T. Yoshiie, S. Kojima, J. Nucl. Mater. 141–143 (1986) 625.
[2] M. Kiritani, T. Yoshiie, S. Kojima, Y. Satoh, Radiat. Eff. Def. Solids 113 (1990) 75.
[3] Y. Satoh, I. Ishida, T. Yoshiie, M. Kiritani, J. Nucl. Mater. 155–157 (1988) 443.
[4] T. Nakagawa, T. Shibata, S. Chiba, T. Fukahori, T. Nakajima, Y. Kikuchi, T. Kawano, Y. Kanda, T. Ohsawa, H. Matsunobu, M. Kawai, A. Zukeran, T. Watanabe, S. Igarasi, K. Kosako, T. Asami, J. Nucl. Sci. Technol. 32 (1995) 1259.
[5] J. Lindhard, V. Nielsen, M. Scharff, P.V. Thomsen, Mat. Phys. Medd. Dar. V.D. Selk. 22 (1963) 10.
[6] L.R. Greenwood, R.K. Smither, ANL/FPP/TM-197, Argonne National Laboratory, 1985.