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# Effects of Transmutation Products on the Formation of Interstitial Loops in High Purity Aluminum

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## Synopsis

Effects of the transmutation products during neutron irradiation in high purity aluminum on the formation of the interstitial loops were examined by the subsequent electron irradiation and electron microscopic observation. It is shown that Si atoms formed by $(n, \gamma)$ nuclear reaction enhance the nucleation of interstitial loops. It is pointed out that p or $\alpha$ formed by (n,p) or $(n, \alpha)$ nuclear reactions form its complex with vacancies at high temperature and these complexes also enhance the nucleation of the interstitial loops.

## I. Introduction

It is well known that many transmutation products are introduced into materials by nuclear reactions under fusion reactor conditions. The presence of gas bubbles formed in these materials lead to detrimental effects on the mechanical properties of plasma wall materials [1] , and consequently better understanding of their effects has been the subject of numerous experimental investigation. However,it is not well understood how transmutation products affect the nucleation of

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the interstitial type dislocation loops ( I-loops ) . This problem is also important to consider radiation effects of neutrons in metals.

Principal nuclear reactions in aluminum are : $^{27}Al(n,p)^{27}Mg$ and 100% of $^{27}Mg$ changes to $^{27}Al$ by $\beta$ decay, $^{27}Al(n,\gamma)^{24}Na$ and 79% of $^{24}Na$ changes to $^{24}Mg$ by $\beta$ decay and $^{27}Al(n,\gamma)^{28}Al$ and 12% of $^{28}Al$ changes to $^{28}Si$ by $\beta$ decay [2] .

Concerning to these elements, Ezawa et al. demonstrated that Si atoms in Al-Si alloy trap the self-interstitial atoms and cause a long incubation time necessary to grow up to visible size loops by election irradiation [3] . Shimomura and Kuwabara suggested that I-loops are nucleated heterogeneously at remained impurity sites even in zone-refined aluminum by 200keV election irradiation [4] . Kiritani remarked that there is a temperature range where I-loop nucleation is sensitive to the impurity [5] . Yoshida et al. suggested that vacancy -helium atom complexes act as nucleation sites of I-loop in Mo [6] . Ono et al. suggested that small size vacancy-hydrogen complexes also act as nucleation sites of I-loop in Al [7] . In these works, the trapping centers were introduced by ion irradiation or casting. Therefore, the purpose of the present work is to examine the effect of the transmutation products which were introduced into high purity aluminum actually by neutron irradiation. Further irradiations by electrons of these specimens must give valuable informations about the nucleation ofI-loop. I-loop formations by electron irradiation in very dilute Al-Si alloys and in high purity aluminum which was pre-irradiated with helium or hydrogen ions are also examined, and compared with the effects of transmutation products.

## II. Experimental Procedure

Materials used in the present work were zone-refined aluminum of 99.9999% purity [8] . The sheets or disks of thickness 0.01cm were pre-annealed at 800K for 3h in a vacuum of $1\times10^{-6}$ Torr. and supplied for neutron irradiation. Specimen A was irradiated in the fuel region of JMTR ( Japan Materials Testing Reactor ) to the fluence of $7.6\times10^{20}\cdot n\cdot cm^{-2}$ ( >1MeV ) at about 670K. Specimen B was irradiated in the region of the first Be reflector to the fluence of $1\times10^{19}\cdot n\cdot cm^{-2}$ ( >1MeV ) at about 470K and specimen C was irradiated in JOYO to the fluence of $9\times10^{21}\cdot n\cdot cm^{-2}$ ( >0.1MeV ) at about 670K. Specimen D was irradiated with 14MeV neutrons in RTNS-II at Lawrence Livermore National Laboratory to the fluence of $3\sim6\times10^{18}\cdot n\cdot cm^{-2}$ at 470K. These specimens were electrolitically polished and observed by an electron microscope JEM 200CX at Oarai branch of Tohoku University.

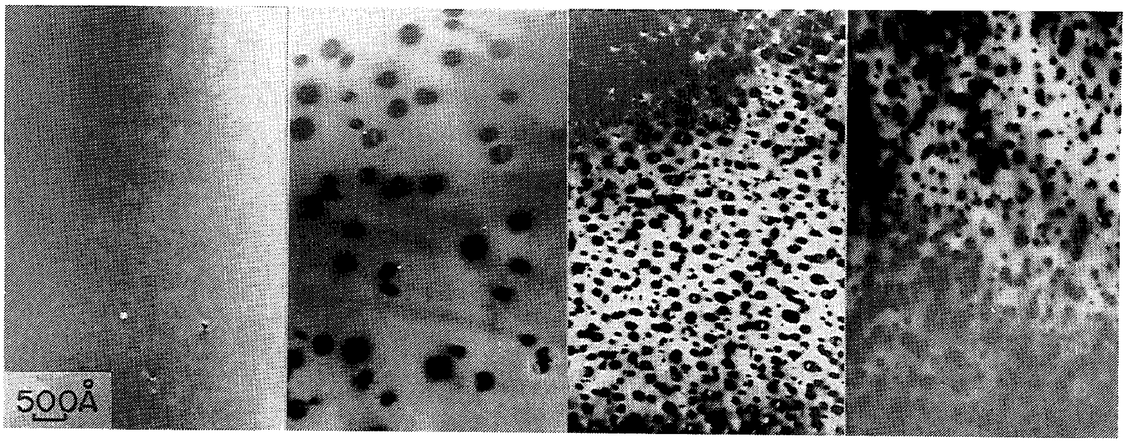
To compare with the effects of neutron irradiation, same quality specimens were electrolitically polished and irradiated with 17keV helium ions or 15keV hydrogen ions at a flux of $1\times10^{13}ions\cdot cm^{-2}\cdot s^{-1}$ at various temperatures.

To examine the effects of trapping of self-interstitial atoms by transmutation products and the formation of I-loops, pre-irradiated specimens with neutrons or ions were further irradiated with 200 keV electrons at a flux of $2\times10^{18}\sim2\times10^{19}\,e\cdot cm^{-2}\cdot s^{-1}$.

## III. Results and Discussion

### 1. Effects of silicon

In neutron irradiated specimens A, B and C, any radiation damages were not observed by the standard technique of electron-microscopy, except voids rarely observed in the speimen A, as shown in Fig.1-(a), and in the specimen B. This results mean that the temperatures of the neutron irradiation were too high to form any clusters of point defects in aluminum. When these specimens were further irradiated with 200keV electrons at a flux of $6\times10^{18}\,e\cdot cm^{-2}\cdot s^{-1}$ at room temperature, however, many I-loops were observed depending on the condition of the neutron irradiation as shown in Fig.1-(b), (c) and (d). The densities of I-loops in these figures are $1.3\times10^{10}$, $1.2\times10^{11}$ and $1.1\times10^{11}\,cm^{-2}$, respectively. In pure aluminum without the pre-irradiation with neutrons, the density of I-loops formed by the similar electron irradiation was at most $4\times10^{9}\,cm^{-2}$, and did not increase even for a long time irradiation, as seen in Fig.2. Even if the flux of electrons was changed between $2\times10^{18}$ and $2\times10^{19}\,e\cdot cm^{-2}\cdot s^{-1}$, the increase of the loop density was less than 20%, which is consistent with the result demonstrated by Shimomura and Kuwabara [4]. Therefore, these present results indicate that the transmutation products by the neutron irradiation caused the increse of I-loop density in the subsequent electron irradiation. The concentration of transmutation products introduced into the specimen by (n,p), (n,$\alpha$) and (n,$\gamma$) reactions was estimated, using the data of the cross section [9] and the



(a) (b) (c) (d)

Fig.1 Electron micrograhs in (a) specimen A irradiated in JMTR at 670K. (b) specimen B, (c) specimen A and (d) specimen C, which were further irradiated with electrons at room temperature.

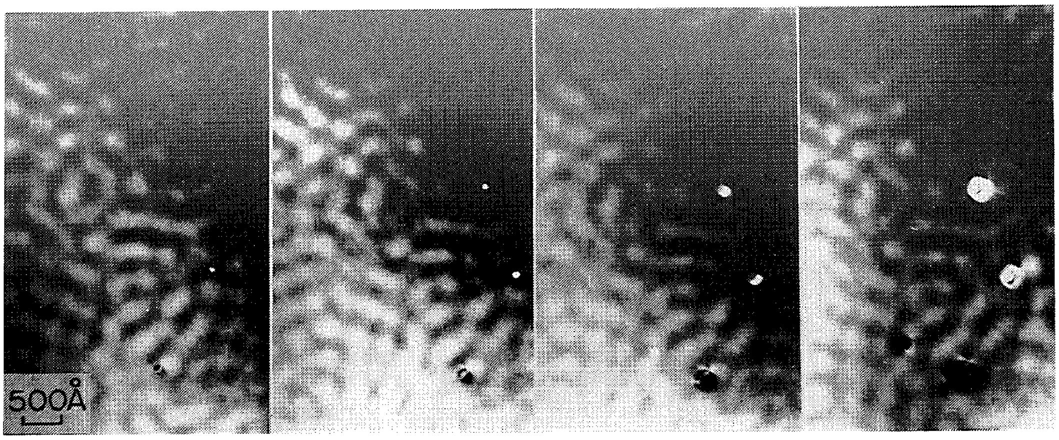


Fig.2 Dislocation loops in the same area of the specimen irradiated with electrons for 4, 6, 10 and 15 min, respectively.

Table I CONCENTRATION OF TRANSMUTATION PRODUCTS

| Reactor ($n/cm^2$) | Irr.Temp.(K) | p | $\alpha$ | Si | Mg |
|---|---|---|---|---|---|
| JOYO ($9x10^{21}$>0.1MeV) | 670 | $1x10^{-5}$ | $7x10^{-7}$ | $1x10^{-4}$ | $6x10^{-7}$ |
| JMTR ($7.6x10^{20}$>1MeV) | 670 | $4x10^{-6}$ | $8x10^{-7}$ | $8x10^{-5}$ | $6x10^{-7}$ |
| JMTR ($1x10^{19}$ >1MeV) | 470 | $5x10^{-8}$ | $3x10^{-8}$ | $3x10^{-6}$ | $2x10^{-8}$ |
| RTNS-II ($3\sim 6x10^{18}$) | 470 | $2\sim 5x10^{-7}$ | $4\sim 8x10^{-7}$ | | $3\sim 6x10^{-7}$ |

energy spectrum of the irradiated neutrons supplied by Japan Atomic Energy Research Institute. The calculated results are tabulated in Table I. It is clear that silicon atom produced by (n, $\gamma$) reaction in JMTR and JOYO is the predominant element. To compare the effects of Si introduced into the specimen by transmutation and casting, the dilute alloys, i.e. Al-30ppmSi and Al-150ppmSi, were similarly irradiated with electrons. The results are shown in Fig.3, where the loop densities are 6.5 x $10^{10}$ and 1.3 x $10^{11}cm^{-2}$, respectively. The solubility limit of Si in aluminum at room temperature may be estimated by the extrapolation of the solubility data at high temperature [10] to be about 40-60ppm. Therefore, it is noted that the loop density sharply depends on the concentration of Si introduced into the specimen by the both way, if the concentration is lower than the solubility limit. One of reasons why the I-loop density dully depends on Si concentration above the solubility limit must be a clustering of Si.

Silicon atom is an under size element in aluminum [11,12]. Ezawa et al. demonstrated that the under size element in aluminum interacts strongly with the self-interstitial atoms and causes a longer incubation time for the loop growth [3]. However, the concentration of Si in their specimen is too high to judge whether the trapping effects are due to isolated Si atoms or the clusters. Shimomura and Kuwabara indicated that I-loops are nucleated heterogeneously at remaining impurity even in zone-refined aluminum [4]. However, the kind of the impurity did not identified.

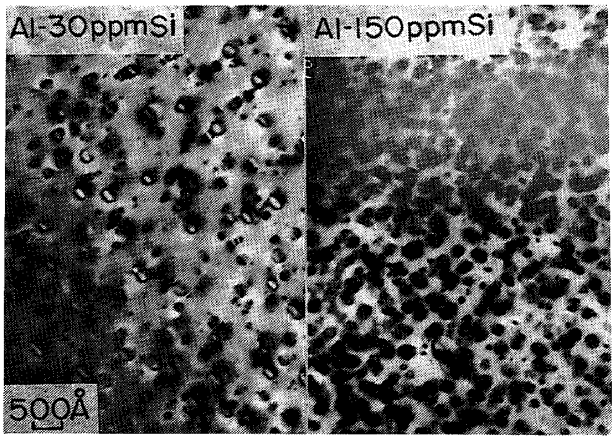


Fig.3 Dislocation loops in Al-30ppmSi and Al-150ppmSi.

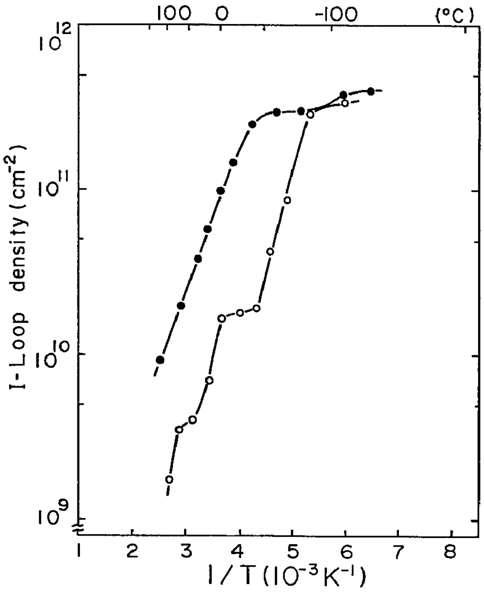


Fig.4 Loop densities in Al specimens, (●) pre-irradiated in JMTR to 1 x $10^{20}$n · $cm^{-2}$ at 630K, (○) without the pre-irradiation.

We can obtain very dilute Al-Si alloy which is suitable for the study of the interaction between Si and the self-interstitial atom by neutron irradiation of high purity aluminum at high temperature. The following electron irradiation at various temperatures must give worthwhile information about the binding strength between Si and the self-interstitial atom. An example of the dependence of the loop density in pre-neutron irradiated specimen on the temperature of the electron irradiation is shown in Fig.4. The comparison with the result in high purity aluminum without pre-neutron irradiation suggests that the difference in the loop density above 200K is due to Si atoms in solid solution. The trapping kinetics of the self-interstitial atoms by Si in Al will be developed in near future.

## 2. Effects of helium and hydrogen

In the case of specimen D, which was irradiated with 14MeV neutrons in RTNS-II, any radiation damages were not observed. However, as seen in Fig.5, many I-loops were developed by the subsequent electron irradiation. Figure 5 and Table I. suggest a possibility that helium or hydrogen atoms which were introduced into the specimen by (n, α) or (n, p) reactions also enhance the formation of I-loops in the subsequent electron irradiation. To confirm this point, thin foils ofaluminum were pre-irradiated with 17keV helium or

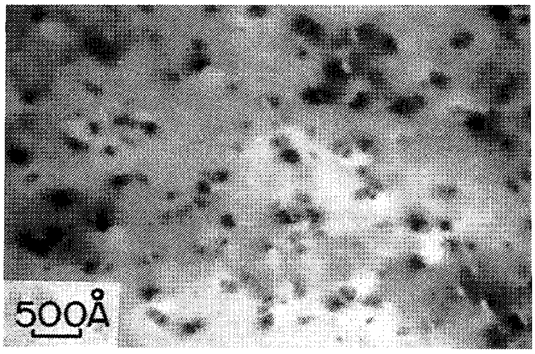
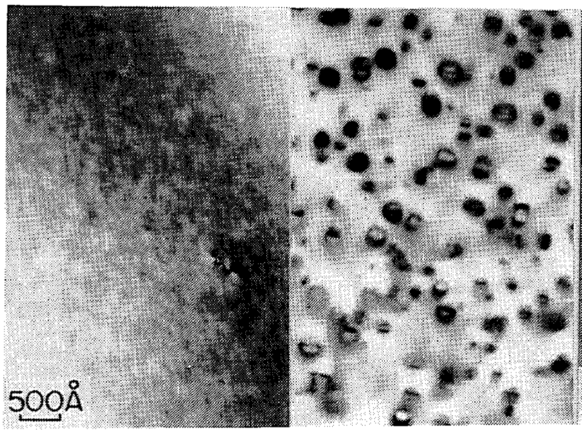
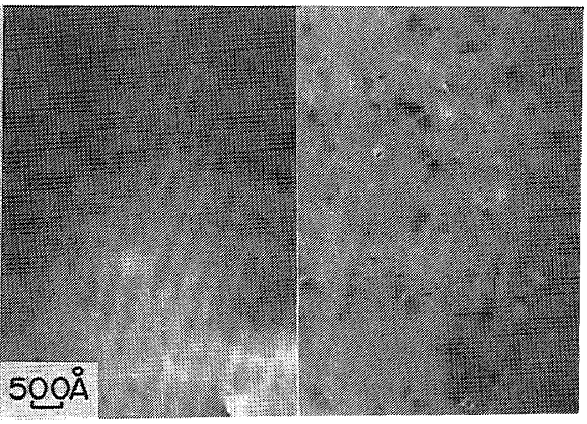


Fig.5 Dislocation loops in specimen D which was further irradiated with electrons.



(a) (b)

Fig.6 Electron micrographs in Al (a) pre-irradiated with helium ion at 473K and (b) further irradiated with electrons at room temperature.



(a) (b)

Fig.7 Electron micrographs in Al (a) pre-irradiated with hydrogen ions at 473K and (b) further irradiated with electrons at room temperature.

15keV hydrogen ions to a fluence of 2 x $10^{13}$ ions · $cm^{-2}$ at 470K. In these specimens, any radiation damages were not observed as shown in Figs.6-(a) and 7-(a). However, many I-loops were developed by the subsequent electron irradiation as seen in Figs.6-(b) and 7-(b), respectively. These results strongly suggest that the trapping sites of the self-interstitial atoms are formed by the pre-irradiation of ions. At 473K, any clusters of vacancy or of the self-interstitial atoms can not be nucleated. Besides, the interaction between the self-interstitial atoms and thesegas atoms does not seem to be so strong [13] . Therefore, the possible trapping sites must be a helium-vacancy complexes or a hydro-gen-vacancy complexes.

We have pointed out that hydrogen-vacancy complexes which were formed by the pre-ion irradiation at high temperature act as nucleation sites of I-loops in the following electron irradiation. A depth distribution of these loops coincides with the projected ion range calculated by TRIM code [14] . The density of I-loops satisfied the Arrhenius relation to the inverse of the pre-ion irradiation temperature as shown in Fig.8. This relation yields an apparent binding energy Bapp=0.3eV. From a brief consideration on the kinetics under the ion irradiation, it was pointed out that Bapp may be related to a summation of binding energies to form a nucleus of hydrogen-vacancy complex and that the size of the nucleus is small [7] .

The specimen which was pre-ion irradiated at 473K to a fluence of 5 x $10^{14}$ ions · $cm^{-2}$ was further irradiated with electrons at various temperatures. A temperature dependence of thus formed I-loop density is shown in Fig.9. Comparing the results with results without the pre-ion irradiation, the flat stage at temperatures between 200K and 260K in the figure may be related to the trapping of the self-interstitial atoms at hydrogen-vacancy complexes. The study of the trapping kinetics will be continued.

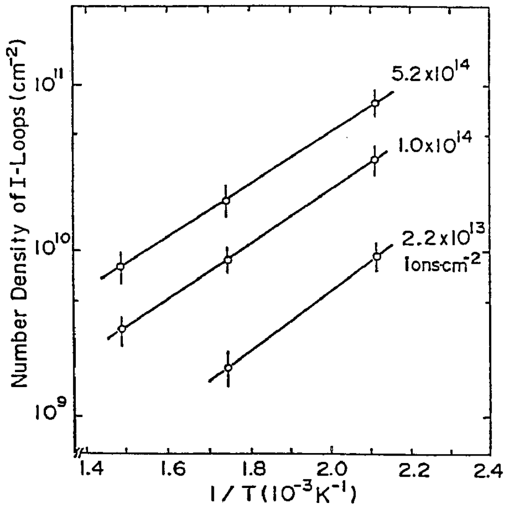


Fig.8 Arrhenius plot of the loop density as a function of the inverse of the pre-ion irradiation temperature.

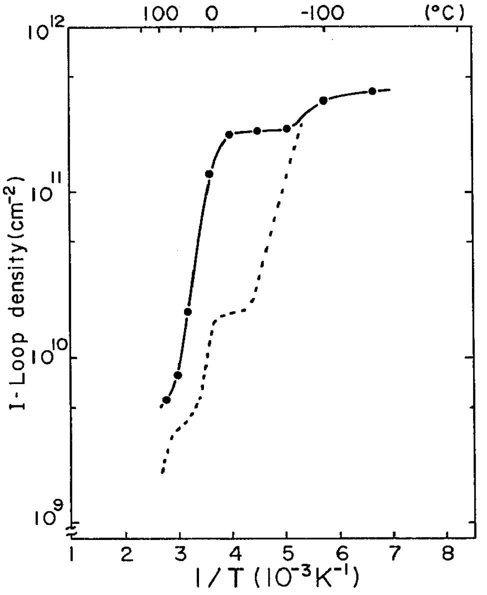


Fig.9 Loop densities in Al specimens as a function of the inverse of the electron irradiation temperature, (●) pre-ion irradiated at 473K, (…) without the pre-irradiation.

Acknowledgements

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