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***In-situ* observation of microstructural development in Fe-Cr-Ni alloys under dual-beam irradiation**

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Abstract. — To investigate the effect of gaseous elements on point defect cluster formation, such as dislocation loops and cavities, *in-situ* observation of dual-beam irradiation has been carried out for Ni and commercial stainless steels by using 1300 kV HVEM and 300 kV ion-accelerator facility. During dual-beam irradiation to Ni, small cavities nucleated at only compression side of faulted interstitial loops. From the loop growth experiment in both of Ni and stainless steels, it was confirmed that injected He can increase the migration energy of vacancies, which means He has a strong binding energy to vacancies. Therefore, He atoms and vacancies can move to the compression side of dislocations. In commercial 316 steels the cavity nucleation was strongly enhanced by the dual-beam irradiation, and the bi-modal distribution in cavity size was developed at higher He concentration. From the dual-beam irradiation with different He injection rate, it was clarified that the cavity formation with small size is needed to accumulate some amount of He in the matrix. These suggest that injected He atoms can be stored in the internal sinks.

1. Introduction.

In last decade, several groups have tried to connect an electron microscope with an ion-accelerator, for example, at Harwell [1], University of Virginia [2], Argonne National Laboratory [3], Hokkaido University [4] and Kyushu University [5]. A high voltage electron microscope (HVEM) connected with an ion-accelerator has many advantages in various research fields, such as *in-situ* observation of defect clusters, bubbles, surface modification process.

The incident electron beam in HVEM produces large amount of point defects; vacancies and interstitial atoms, which usually develop to observable dislocation loops and voids. However, we also have to consider the effect of gaseous elements, He and H, which come from some nuclear reaction or surrounding in neutron irradiation conditions. The dual-beam irradiation of electrons and He ions is useful to understand the radiation-damage process in materials of fusion and fission reactor devices.

The authors have installed 1300 kV HVEM and 300 kV accelerator for studying on radiation damage process in materials. The aim of this study is to clarify the effect of injected He on the development process of dislocation loops and voids which are produced by dual-beam irradiation.

2. Experimental procedure.

2.1 DUAL-BEAM IRRADIATION FACILITY — 1300 kV HVEM (H-1300) has been equipped with 300 kV ion-accelerator (WEC-300) which includes a hot PIG typed gaseous ion source. The incident angle of ion beam to electron beam is 30 degrees. Experimental details are found in another article [4].

By using this facility, irradiation with different modes was carried out on three areas in the same grain, that is, simple electron irradiation, dual-beam irradiation and electron irradiation after He ion-injection. This method enables to compare directly the results from these irradiation modes, because other experimental condition, such as irradiation temperature, specimen thickness, incident angle and other observation conditions can be unchanged.

2.2 IRRADIATION — Pure Ni of Jonson Matthey and a typical commercial 316 austenitic stainless steel were used for the experiment. Irradiation was carried out in the order of simple electron irradiation, dual-beam irradiation and electron irradiation after ion-injection. Dual-beam irradiation was performed with 1000 keV electrons and 70 keV He⁺ at 470 to 830 K. Maximum dose of electrons was up to 12 dpa, which was equivalent to the irradiation of 90 min. Incident direction of electrons was $\langle 001 \rangle$ and $\langle 011 \rangle$ for pure Ni and stainless steel, respectively.

The depth distributions of injected He and induced damages were calculated using TRIM3D program [6]. Maximum depth of them were about 400 nm, therefore the thickness of examined area was decided to 400 nm using thickness fringes of [002] reflections. He concentration was decided by assuming that injected He can spread to whole thickness of specimens. The nominal concentration of ion-injected damage was calculated to be less than two order of magnitudes of electron-produced one, therefore, the contribution of ion-produced damages can be negligible to consider the growth process of dislocations and voids in this experiment.

3. Result and discussion.

3.1 BEHAVIOR OF DISLOCATION LOOPS IN Ni — Microstructural development in Ni during irradiation shows general patterns: almost all of loops were faulted $\langle 111 \rangle$ type, and grew in size with increasing of dose. They changed sometime to unfaulted ones, and finally turn to dislocation lines.

Figure 1 shows a large faulted loops observed in Ni irradiated with two different modes at 670 K, which was contained inside of the specimen. In the case of dual-beam irradiation at 20 atppm He/dpa, a remarkable point is small contrasts existed only inside of the loops, as shown in figure 1a. These contrasts were confirmed to be small cavities by changing observation angles to observed condition. The cavities showed no obvious growth, but increase in the number density, which means that the cavities nucleated continuously on the inside of growing faulted loops during irradiation. In the case of irradiation after preinjection, as shown in figure 1b, there was no preferential cavity nucleation. It indicates that the concentration of He stored in the matrix is quite low, because the preinjected He atoms move easily to surfaces, grain boundaries and other sinks. Therefore, a dynamic supply of He to faulted loops is needed for the preferential cavity nucleation.

With increasing doses, the nucleation and growth of cavities were observed, except at simple electron-irradiation. In the case of electron irradiation, only dislocation segments were formed, but cavities were not confirmed during present experiment. This result suggests that point defects move easily in pure Ni, and cannot lead to a large amount of accumulation of point defects in such thin film of Ni. In the case of dual-beam irradiation, faulted loops were formed preferentially, and

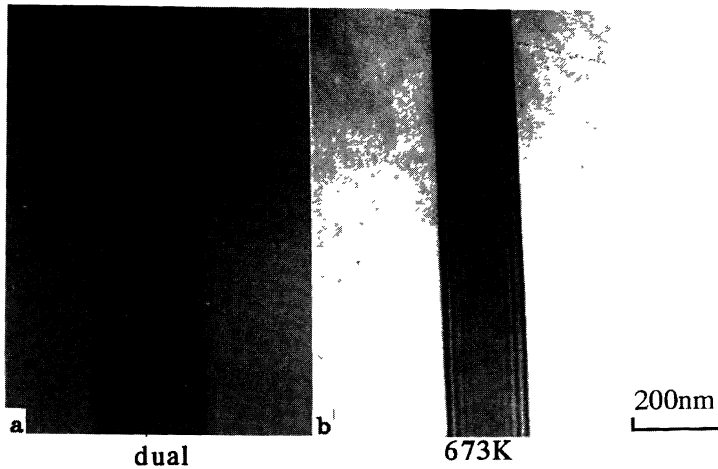


Fig. 1. — Stacking fault loops formed in Ni at early stage of irradiation at 670 K. Dual-beam irradiation at 20 ppm He/dpa (a), and electron-irradiation after hot preinjection (b).

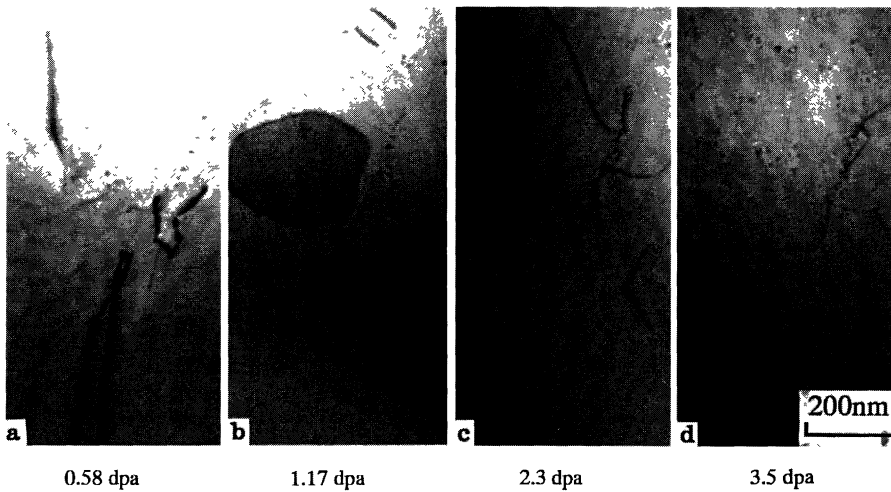
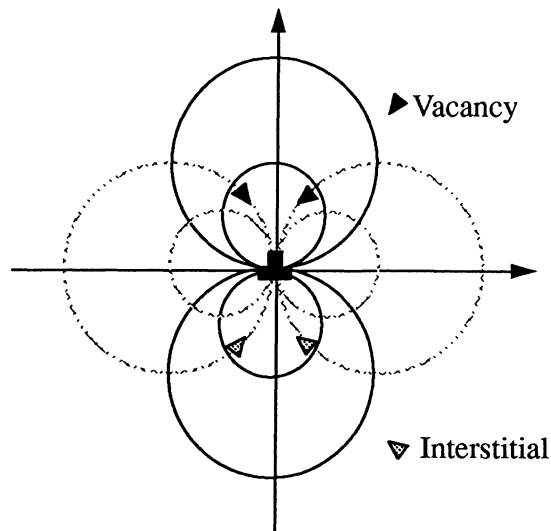


Fig. 2. — Microstructural change in Ni irradiated with dual-beam condition of 20 atppm He/dpa at 673 K. Cavities were nucleated at dislocation segments, which climbed away during irradiation.

cavities nucleated on the loops, as shown in figure 2, and they developed to relatively large voids. In the case of the irradiation after He injection at 670 K (hot preinjection), cavities aligned to dislocations where local concentration of vacancy and He could be higher. In the case of irradiation after He injection at room temperature (cold preinjection), cavities nucleated randomly with high number density, which means that preinjected He could stabilize defect clusters, especially vacancy typed ones.

Urban *et al.* [7] observed the formation of vacancy-type loops on the compression side of interstitial-type loops in electron-irradiated Ni, and concluded that the loops results from the trapping of vacancies by gaseous impurities. In general, from the elastic interaction between

Interaction between Dislocation and Point Defects



He Migrates with Vacancies

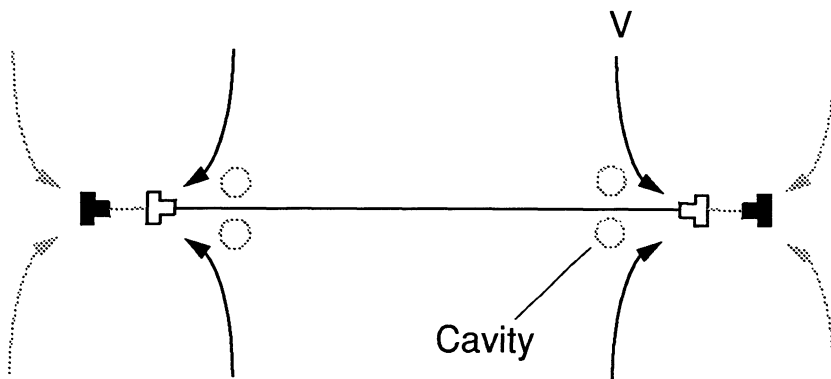


Fig. 3. — Schematic illustration of interaction between point defects and edge dislocation. Vacancies sink to compression side, and self-interstitials move to tension side.

point defects and dislocation, vacancies move to the compression side of the loops, and interstitials come to the tension side, as shown in figure 3. Therefore it is reasonable that vacancy-type defect clusters can form on the compression side of dislocations. As indicated in this experiment, He can assist cavity nucleation at the compression side where the vacancy concentration is higher.

Kiritani *et al.* [8] has proposed a useful method to measure the migration energy of vacancy from growth rate of interstitial typed loops. Figure 4 shows a relation between loop growth rate and temperature for Ni which were electron-irradiated under single and dual beams condition.

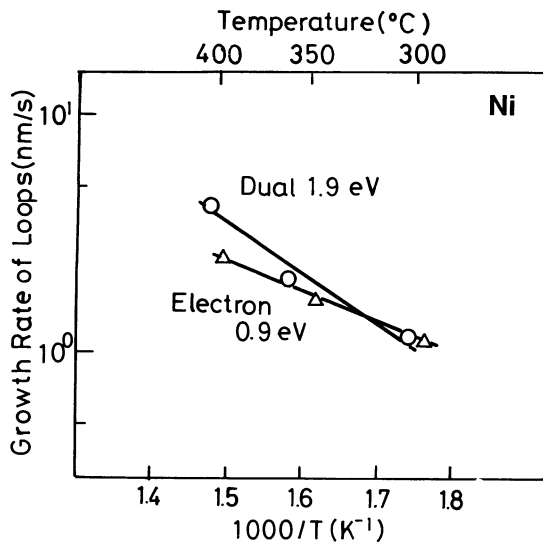


Fig. 4. — Relation between loop growth rate and temperature for Ni under single and dual beams irradiations at limited temperature.

In the case of electron irradiation the migration energy of vacancy was estimated to be 0.9 eV from the twice of the slope in figure 4. On the contrary, at the condition of dual-beams of 20 atppm He/dpa, the energy increased. It suggests that the difference in both of those energies could be explained to be a binding energy between vacancy and He atom.

Schilling [9] has calculated the binding energy of the complex of He atom and a vacancy in Ni to be 2.1 eV. This strong binding could suppress the flow of vacancies to defect sinks, such as dislocations. If some amount of vacancies are consumed by the formation of small cavities located at compression side of interstitial loops, as shown in figure 1a, the growth rate of loops could increase possibly. Also, after preinjection, He enhanced the nucleation of loops, which means that a strong binding can stabilize for cascade damages. It is possibly explained that large number of unfaulted loops can nucleate in preinjected areas even at low doses.

3.2 BEHAVIOR OF DISLOCATION LOOPS IN STAINLESS STEEL — Figure 5 shows typical sequences of dislocation loop formation under simple electron-irradiation. In this process, the growth of the loops were not so obvious compared to Ni, and the loop had irregular shapes. The reason for such low growth rate in this steel could be defect trapping and/or segregation of the high concentrated solute atoms [10].

In the case of the dual-beam irradiation, much difference cannot be observed in the size and number density of the dislocation loop compared to simple electron irradiation. The complicated loop structures were composed of small-sized loops with a high number density. It suggests, however, such small amount of He, which is estimated to be <50 atppm, cannot compete with the effect of solute atoms on the loop structure, as described above.

To estimate the vacancy migration energy in stainless steel, the same method of loop growth rate as in Ni has been applied. Figure 6 shows temperature dependence of the growth rates under simple and dual beam irradiations at the condition of 70 atppm He/dpa. In the case of simple electron irradiation, the energy was estimated as 1.4 eV, which is a little larger compared to other

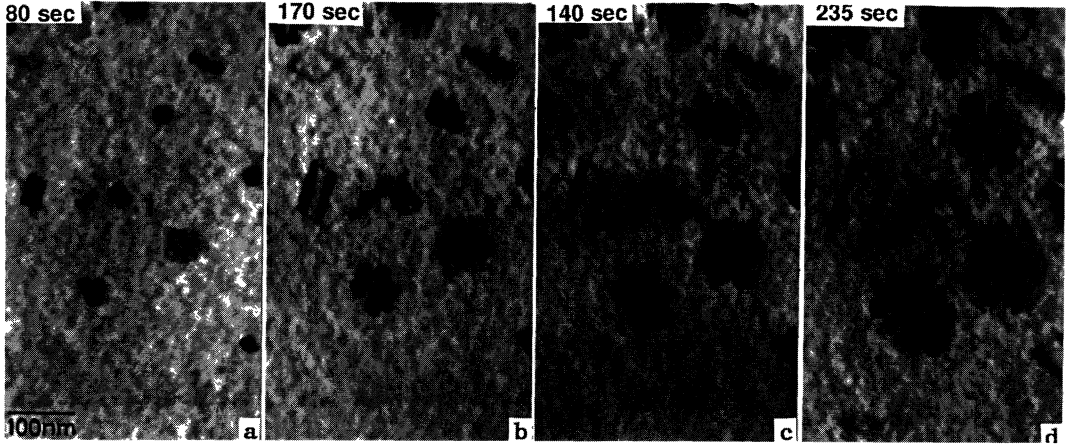


Fig. 5. — Typical sequences of dislocation loop formation under simple electron-irradiation at 770 K.

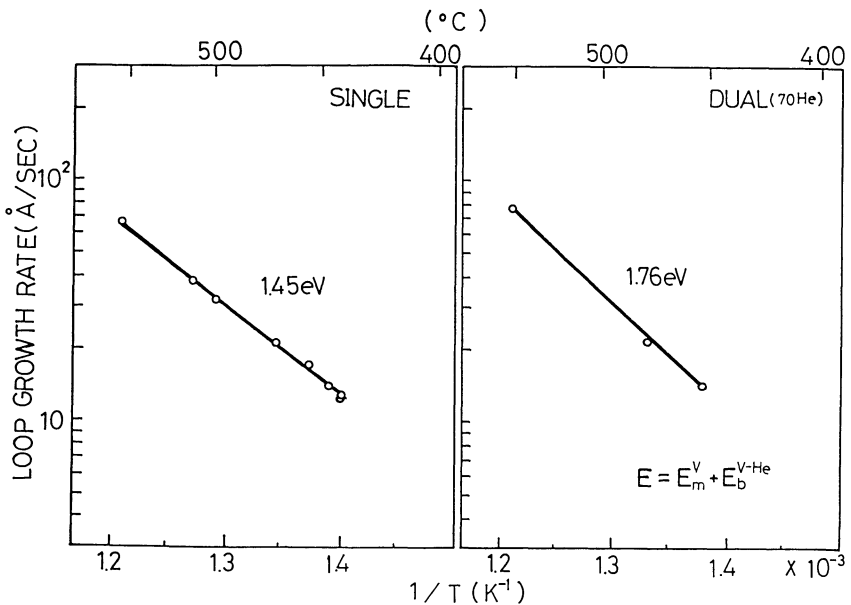


Fig. 6. — Temperature dependence of the growth rates under simple and dual-beam irradiations at the condition of 70 ppm He/dpa.

reported values, and could be caused by the difference in impurity level. In the case of dual-beam irradiation, the energy was estimated as 1.7 eV. The difference of about 0.3 eV should become from the effect of injected He, and can be considered as a binding energy in this stainless steel. The reason for the different binding energy as that of Ni could be explained by the effect of alloying elements and impurities contained in this stainless steel.

3.3 EFFECT OF He ON CAVITY FORMATION IN STAINLESS STEEL — With increasing dose cavity structures developed at both of simple and dual-beam irradiation. At simple electron irradiation, relatively large cavities were formed with low number density, and no cavity formation was observed at high temperature. It means that the nucleation of cavities finish at initial stage of the irradiation, and the growth process is predominate with increasing of doses.

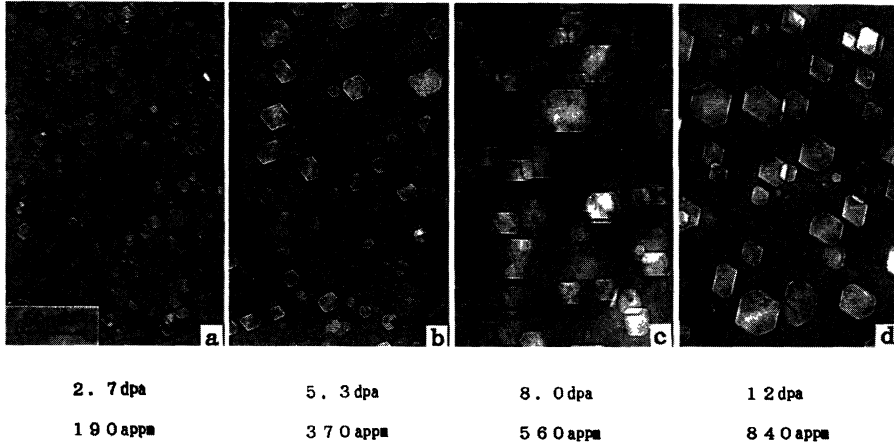


Fig. 7. — Microstructural change of cavities in standard 316 stainless steel during dual-beam irradiation of 70 atppm He/dpa at 773 K.

On the contrary, dual-beam irradiation influenced strongly the cavity formation; even at high temperature, relatively large numbers of cavities were observed. Figure 7 shows the development of cavity structures in stainless steel during dual-beam irradiation of 70 atppm He/dpa at 773 K. Those results show that He can stabilize the nucleus of cavities. Other point was that the number density of small cavities increased without suppressing the formation of large cavities. This means that the cavity nucleation is continuous with increasing concentration of He under dual-beam irradiation. It can be noted that the cavity structures was categorized into two types; large cavities with a small number density and fine cavities with a quite high number density, as shown typically in figure 7d. Such cavity structures are named “bi-modal distribution” [11], which means two typed cavities has different nature; voids which have relatively large size and low gas-pressure, and bubbles which have small size and high gas-pressure.

In this experiment, small bubbles were detectable over 8 dpa. Figure 8 shows dose dependence of cavity number density in stainless steel irradiated with dual-beam of 70 atppm He/dpa at 773 K. A solid line shows the number density of large voids, and a dotted line shows the total number density of voids and small sized bubbles. This result indicates that the bubble formation needs to accumulate appropriate He concentratin in specimen matrix, where the critical concentration can assumed to be 500 atppm He in this irradiation condition. To confirm the critical He concentration, another dual-beam irradiation was carried out at more higher rate of He injection, 140 atppm He/dpa. The data is shown in figure 8 as a dotted line with closed circle. This result shows that the bubble formation can not be decided by defect concentration or dose, but by the accumulation of He. In general, injected He can move to several sinks, such as dislocation, cavities,

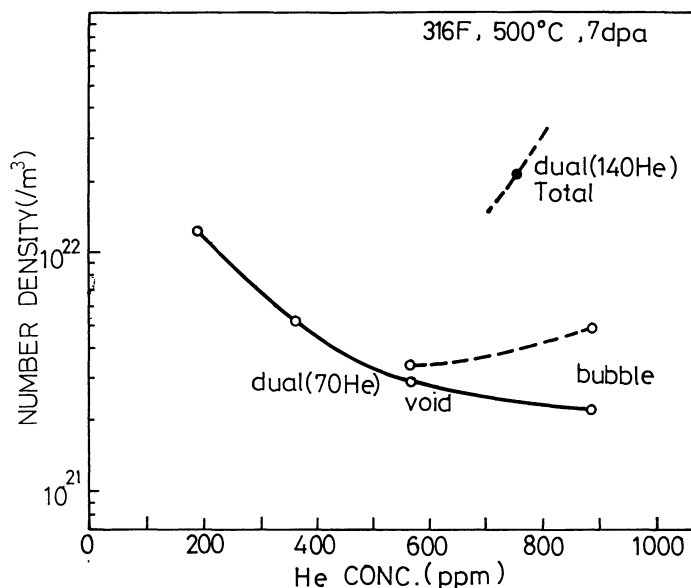


Fig. 8. — Dose dependence of cavity number density in standard 316 stainless steel. Small bubbles developed over 8 dpa, which is equivalent to about 500 atppm He, and the number increases slightly with increasing of irradiation dose.

grain boundaries and surfaces, however, the efficiency of He trapping for each sinks has not been decided, because there are no reliable data on the interaction among He atoms and sinks. In this experiment, the reason for the presence of critical He concentration to form bubbles indicates that the amount of He trapped at dislocation could be same, even if different irradiation condition.

Figure 9a shows the size distribution of cavities in the stainless steel after dual irradiation to several doses on 70 atppm He/dpa at 773 K. It can be easily recognized the size distribution showed “critical size”, which was about 5 - 10 nm. Mansur [10] has considered that such bi-modal size distribution is originated from the increase of surface energy by the gas pressure in cavities which can affect on the sink efficiency for point defects. The theory could explain that large voids can be grown by the help of gas pressure, but small ones, named as bubbles, cannot grow because of thermal emission of vacancies.

At higher concentration of He, as shown in figure 9b, almost all of cavities cannot grow. It means much amount of He enhances the cavity nucleation, but decreases the growth rate, which suggests that the amount of small sized cavities, which act as neutral sinks, should be increased with increasing of He concentration. In general it has been recognized that He has strong effect on cavity formation, because of increasing of the nucleation. However, these results indicate that He of high concentration can enhance the nucleation, but suppress the void swelling during dual-beam irradiation.

4. Summary.

Dual-beam irradiation of He⁺ ion and electron has been carried out for Ni and commercial 316 stainless steel by using HVEM/ion-accelerator facility. The result can be summarized as follows:

1) In dual-beam irradiated Ni, small cavities were nucleated on compression side of interstitial

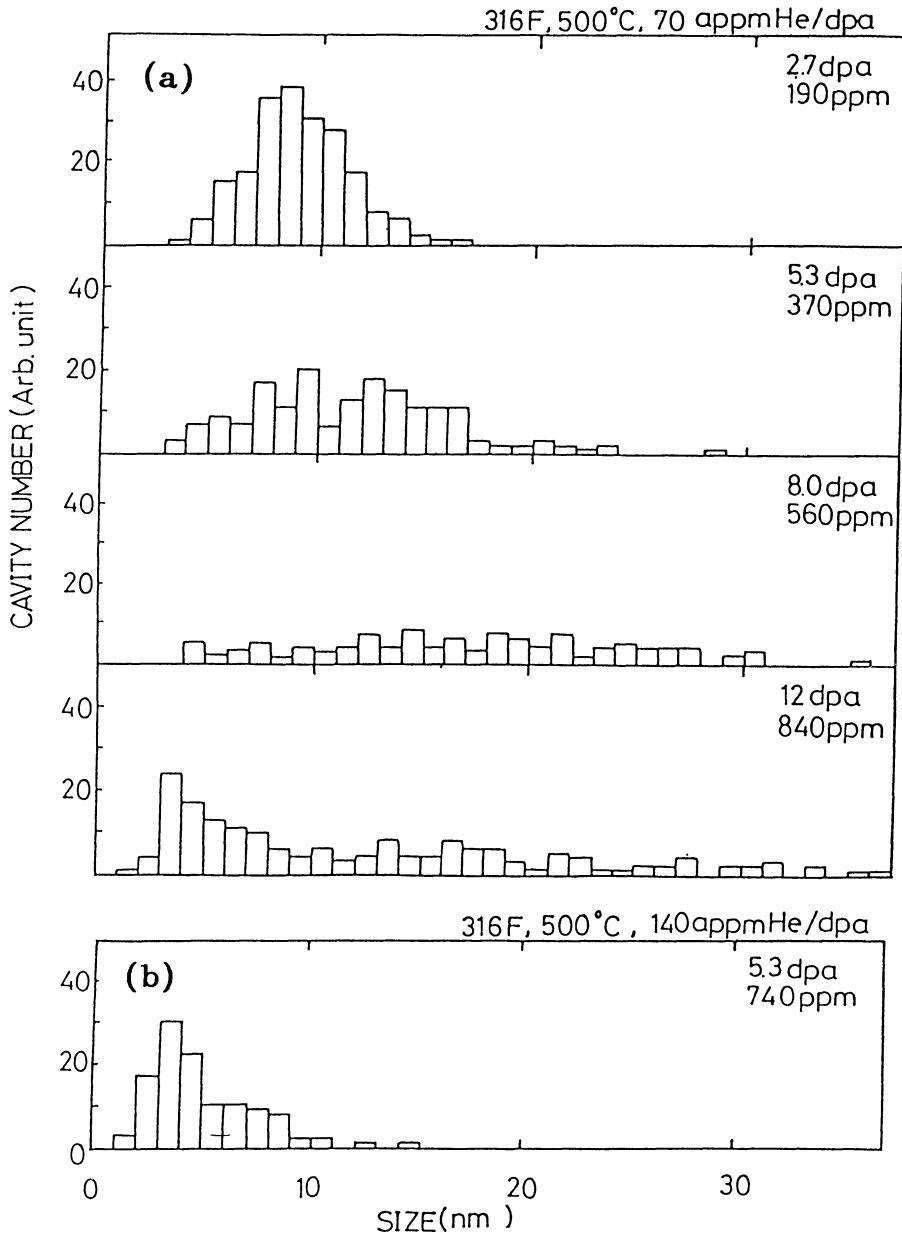


Fig. 9. — Cavity size distribution in standard 316 stainless steel during dual-beam irradiation at 773 K on different He condition: a) 70 atppm He/dpa, and b) 140 atppm He/dpa.

type loops with stacking fault. It means that injected He atoms move to dislocation loops with interacting with vacancies, and stabilize the cavity nucleus.

- 2) From the measurement of loop growth rate in both of Ni and stainless steels, it was suggested that injected He can increase the migration energy of vacancies.
- 3) In stainless steels the void number density was strongly increased by the dual-beam irradiation.

tion up to high doses. This fact indicates that He can enhance the nucleation of void even in commercial alloys which has a resistance for defect cluster formation.

- 4) From the dual-beam irradiation with different rate of He injection, it was confirmed that He accumulation is necessary to form small cavities. The fact means that injected He atoms can be stored to sinks, such as dislocations and grain boundaries, until small cavity formation.
- 5) The bimodal distribution of cavity size was developed at higher He concentration. The critical size of the cavities was 5-10 nm, and decreased slightly at high temperatures. However, the total volume of cavities decreased at higher He concentration, which means that small size cavities can act as a neutral sink for point defects.

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