Dose and Temperature Dependence of Void Swelling in Electron Irradiated Stainless Steel

Akimichi HISHINUMA, Yoshio KATANO and Kensuke SHIRAISHI

Japan Atomic Energy Research Institute*

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In a high voltage electron microscope, solution treated Type 316 stainless steel was electron-irradiated at temperatures in the range of 370-630°C to a dose of about 30 dpa. The swelling ($\Delta V/V$) induced by the irradiation beyond about 5 dpa is well described by an empirical equation, $\Delta V/V = A(dpa)^n$, under constant void and dislocation densities. With increasing irradiation temperature, the fluence exponent $n$ increases and the pre-exponent term $A$ decreases. At 550°C irradiation, the fluence exponent takes the value of 1.5 due to the diffusion-limited void growth. The value of $n$ larger than 1.5 at higher temperature (>550°C) is attributable to the surface reaction-limited void growth. The smaller value of $n$ for the low temperature (≤500°C) irradiation appears to arise from the dislocation-assisted vacancy diffusion. The peak swelling temperature of the specimen irradiated to 30 dpa is about 570°C, which shifts to a higher temperature with increase in electron dose.

KEYWORDS: stainless steels, electron beams, irradiation, swelling, voids, dislocations, high voltage electron microscope, mechanical radiation effects, dose rate, dose-response relationship, temperature dependence

I. INTRODUCTION

The void swelling is one of the major problems in austenitic stainless steels as core materials for use in fast breeder reactors or fusion reactors. A large amount of experimental data is available for the swelling in the stainless steels as a function of neutron fluence. However, there are wide disagreements concerning the functional relationship between swelling and fluence, and the same set of experimental points may be fitted by some different empirical equations. The Dounreay type of analysis (1) assumed a linear relationship between swelling ($\Delta V/V$) and fluence ($\phi t$) after an incubation period. The empirical equation used by American workers (2) is $\Delta V/V \propto \phi t^2$, where the fluence exponent $n$ is determined by the best fit.

The experimental results on neutron irradiated stainless steels (3)-(5) indicate that the fluence exponent $n$ is temperature dependent and takes the value $\sim 1$ near 400°C, increasing toward $\sim 2.5$ with irradiation temperature. For proton-irradiated Type 316 stainless steel, Keeper et al. (6) reported also the power law dependence of the void swelling; the fluence exponents are 0.68, 1.3 and 2.9 at irradiation temperatures of 400, 500 and 600°C, respectively. With a high voltage electron microscope (HVEM), the temperature dependence of the fluence exponent has not so far been clarified. Some experimental results (7)-(9) suggest that the fluence exponent is little dependent on the electron-irradiation temperature.

The void swelling theories based on diffusion-controlled void growth depict that the fluence exponent $n$ depends on metallurgical structure; the value lies in the range of 0.47-3 depending on the strength of dislocations, voids and precipitates as a sink for radiation-produced point defects (8)-(11). When the void growth is controlled by vacancy reaction at the void surface, the swelling is described by the fluence exponent of 3 (12). In HVEM, single point defects are produced uniformly through-

* Tokai-mura, Ibaraki-ken.
out the specimen without transmutation effects and collision cascades. Thus, the use of a HVEM is beneficial for studying the void swelling in comparison with the current theories.

The present paper describes the behavior of voids and dislocations produced in Type 316 stainless steel electron-irradiated in a HVEM. The dose and temperature dependence of the void swelling will be discussed together with the void growth under a constant density of sinks for radiation-produced point defects.

II. EXPERIMENTAL PROCEDURE

A sample used in this experiment was the Type 316 stainless steel solution treated for 10 min at 1,100°C. A chemical analysis of the materials is given in Table 1. Specimens for electron microscopy were prepared by electro-polishing in a solution of 95% acetic acid and 5% perchloric acid at a temperature below 15°C using a Tenupol jet electro-polishing unit.

### Table 1 Chemical composition of specimens

<table>
<thead>
<tr>
<th>Element</th>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>P</th>
<th>S</th>
<th>Cu</th>
</tr>
</thead>
<tbody>
<tr>
<td>%</td>
<td>0.06</td>
<td>0.46</td>
<td>1.55</td>
<td>0.014</td>
<td>0.008</td>
<td>0.27</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Element</th>
<th>Ni</th>
<th>Cr</th>
<th>Mo</th>
<th>B</th>
<th>Fe</th>
</tr>
</thead>
<tbody>
<tr>
<td>%</td>
<td>13.90</td>
<td>16.90</td>
<td>2.22</td>
<td>0.0014</td>
<td>Balance</td>
</tr>
</tbody>
</table>

The specimens were irradiated with 1 MeV electrons in a JEM-1000D, at temperatures in the range of 370~630°C to a total dose of about 30 dpa. The temperatures were deduced from the prior calibration of the temperature-power characteristic of the specimen holder. The electron flux was measured by using a Faraday cup installed above the screen. The average electron flux at the central area of irradiation was $3.4 \times 10^{15}$ to $4.0 \times 10^{15}$ e/cm²·sec. The electron flux corresponds to the damage rate of 5.7 to 6.7 dpa/hr taking the threshold energy of 25 eV. The thickness of the specimen was estimated to be about 0.5 to 1.0 μm by counting the number of equal thickness fringes from the specimen edge.

Electron images recorded at a magnification of ×20,000 were printed at a magnification of ×73,000. Measurements of void size were made within an area of 150 mm × 150 mm about the center of the irradiated area on each micrograph. The swelling was calculated from the summation of volume of voids within the area. In the calculation, the voids were assumed to be spherical in shape and classified by units of about 100 Å dia.

### III. EXPERIMENTAL RESULTS

Effect of irradiation temperature on the void component of the damage structure is shown in Photo. 1. The specimens were irradiated to a dose of about 30 dpa in the temperature range of 370~600°C. The voids observed in the specimen irradiated at lower temperatures are larger in void number density and smaller in diameter. A sharp decrease in the void number density with increasing the irradiation temperature was observed above 600°C, and no voids were formed in the specimen irradiated at 630°C.

The void number density, void diameter and void swelling are given in Figs. 1~3 respectively as a function of dpa. The void number density increases steeply at an initial stage of irradiation to be saturated at about 5 dpa (Fig. 1). More precisely, the density rather
The voids nucleated in the early stage of irradiation grow with electron dose (Fig. 2), and the average void diameter $D$ in Å units can be expressed by $D = B \times (dpa)^m$, where $B$ and $m$ are constant. The fluence exponent $m$ depends on the irradiation temperature and takes the value of 0.4 at 500°C. The value increases to 0.5 at 550°C and 0.6 at 570°C. At a total dose of about 30 dpa, the average void diameters are about 700, 1,000 and 1,600 Å for the irradiations at 500, 550 and 570°C, respectively.

Photo 1 Effect of irradiation temperature on void component of damage structure in Type 316 stainless steel electron-irradiated to dose of about 30 dpa

Fig. 2 Dose dependence of void diameter in Type 316 stainless steel electron-irradiated at 500, 550 and 570°C
The swelling (\(DV/V\)) can be expressed by a relation of the form \(DV/V = A(dpa)^n\) as indicated by the solid lines in Fig. 3. The lines through the data are drawn according to the best fit:

\[
\begin{align*}
500^\circ C: & \quad DV/V(\%) = 0.08 (dpa)^{1.1} \\
550^\circ C: & \quad DV/V(\%) = 0.02 (dpa)^{1.6} \\
570^\circ C: & \quad DV/V(\%) = 0.01 (dpa)^{1.8} \\
600^\circ C: & \quad DV/V(\%) = 0.002 (dpa)^{2.2}
\end{align*}
\] (1)

The fluence exponent \(n\) increases with increasing irradiation temperature. The pre-exponent term \(A\), on the contrary, decreases with increasing irradiation temperature.

Irradiation temperature dependence of void swelling in the specimens irradiated to doses of 20 to 100 dpa is shown in Fig. 4. The solid lines represent the experimental results and the broken lines the values extrapolated from the data assuming the Eq. (1). The peak swelling temperature is about 570°C in the specimens electron-irradiated to 30 dpa. The peak temperature seems to shift to a higher temperature with increase in electron dose.

Photograph 2 shows the development of the dislocation component of the damage structure in the specimen irradiated at 550°C. Both diamond and irregular-shaped dislocation loops appeared in the initial stage of irradiation develop to dislocation lines and tangle to form dislocation networks. It should also be noted that the small double arc images arisen from strain field contrast were observed among the dislocation loops and dislocation lines (Photo. 2(a)). The dislocation density appears to be saturated dynamically at about 5 dpa. The development of the dislocation structure with the electron dose was not greatly different among the specimens irradiated at different temperatures.

**IV. DISCUSSION**

In this experiment, both void and dislocation densities are nearly constant beyond a dose of about 5 dpa. Under the conditions, a spherical diffusion field is set up around the void and the quasi-steady state current of vacancies into the void is given by:

\[
\frac{dn_v}{dt} = 4\pi Dr_v(\overline{C} - C^0) \frac{1}{1 + D/r_vk},
\] (2)

where \(n_v\) is the number of vacancies flowing into the void, \(D\) the diffusivity of a vacancy, \(r_v\) the void radius, \(\overline{C}\) the steady state vacancy concentration at a midpoint between adjacent voids, \(C^0\) the equilibrium vacancy concentration and \(K\) the rate constant at the void surface. When \(r_vk \gg D\), the flow rate of vacancies can be expressed by

\[
\left( \frac{dn_v}{dt} \right) = 4\pi Dr_v(\overline{C} - C^0).
\] (3)
In this case the vacancy current into the void becomes diffusion-limited. In the other extreme case, \( r_v K \ll D \), the rate becomes surface-limited as

\[
\left( \frac{dn_v}{dt} \right)_s = K 4\pi r_v^2 (\bar{C} - C^o).
\] (4)

Then, the current of vacancies into the void may be controlled by the surface-limited mechanism at higher temperatures.

In the diffusion-limited case, the rate of vacancy flux is proportional to the void radius as shown in Eq. (3) and the growth rate of the void is inversely proportional to the void radius:

\[
\frac{dr_v}{dt} = \frac{\Omega}{4\pi r_v^2} \frac{dn_v}{dt} = \frac{DD}{r_v} (\bar{C} - C^o),
\] (5)

where \( \Omega \) is the volume of a vacancy. It is clear from Eq. (5) that the void swelling increases proportionally with (dpa)\(^{1.5} \) under a constant void number density during irradiation. In the surface reaction-limited case, the vacancy flux into the void is proportional to the square of void radius (Eq. (4)) and then, the swelling increases proportionally with (dpa)\(^3 \). Although Bullough & Perrin\(^{(9,10)} \) derived the (dpa)\(^3 \) dependence of the swelling from the diffusion controlled void growth under the condition \( \rho_d r_v^2 > 1 \), where \( \rho_d \) is the dislocation density, the condition is not fulfilled in the present experiment; taking the dislocation density of \( \sim 10^{19} \text{cm}^{-2} \)\(^{(21)} \), the void diameter must be larger than 2,000 Å to satisfy the condition. From the above discussion, the fact that the fluence exponent \( n \) increases from 1.5 to 3 with increasing irradiation temperature suggests that the dominant mechanism for the void growth changes from the diffusion-limited to the surface reaction-limited with increasing irradiation temperature.

Using a Fe-18Cr-8Ni-Si alloy irradiated with 1 MeV electrons at 500°C, Okamoto et al.\(^{(14)} \) observed the voids with the strain field contrast arising from segregation of solute atoms at the void surface, and they discussed the possibility of the surface reaction-limited void growth in the alloy. An example of the voids with strain field contrast observed in the present experiment is shown in Photo. 3. A
number of voids with strain contrast is seen in the specimen irradiated at 500°C to a dose of 1 dpa (Photo. 3(a)). The void embryos grow to be resolvable after about 2 dpa irradiation (Photo. 3(b)). Relatively larger voids of about 300 Å dia. have also strain contrast as demonstrated in Photo. 4. In the photograph the two grains marked A and B were irradiated at the same time at 550°C to a dose of about 5 dpa. While the voids in the grain A have apparently no strain contrast, the voids must have the strain contrast as the void seen in the grain B when the image is taken in the Bragg condition to observe dislocation structure. The observations of the strain contrast around voids have been reported in electron-irradiated Type 304 and 316 stainless steels (16)~(18). Based on theoretical considerations, Wolfer (19) and Wolfer & Yoo (20) deduced that the segregation of impurity atoms onto the void surface results in an elimination of the void bias of radiation produced interstitials and leads to the surface-controlled void growth kinetics.

The lower the irradiation temperature, the smaller the fluence exponent in the power law expression for void swelling becomes. The exponent is expected to be 1.0 for the irradiation below 500°C. In a low temperature region, both the dislocation and void densities are very high and the vacancy flow into the void is affected by near-by dislocations; the voids in the vicinity of dislocation grow more rapidly than the other voids (22). In these circumstances, the flow of vacancies into a void is independent of the void radius, which results in the (dpa)1.0 dependence of void swelling. The fluence exponent smaller than 1.5 can be also deduced from the theory for the diffusion-limited void growth under the condition that the dislocation density is low and the voids are dominant sinks for point defects (8)~(10). In the low temperature irradiation, however, the dislocation density is so high that the condition can never be satisfied. Therefore, in a low temperature region (≤500°C) in this experiment, the controlling mechanism for void swelling...
growth is not likely to be simple diffusion-limited.

The pre-exponent term $A$ is proportional to the void number density. Both the density and the term $A$ decrease with increasing irradiation temperature. The values of $A$ is affected by the void-free zones at the specimen surfaces, and wider the void-free zone is produced in the specimen irradiated at higher temperatures\(^{(23)}\). It should, however, be noted that the void free zone does not affect the fluence exponent $n$ under the condition of constant void number density.

The void nucleation rate can be calculated by the model developed by Russell\(^{(24)}\) and Katz & Wiedersich\(^{(25)}\) as discussed in a previous paper\(^{(21)}\). The calculated void nucleation rates with no internal sinks are compared with the measured void number densities as shown with a broken line in Fig. 5. The curve in arbitrary units was fitted at 550°C with the measured void number density. In the nucleation rate calculation, the ratio of the arrival rate of interstitials to that of vacancies, $\beta_i/\beta_v$, was assumed to be 0.999 and independent of irradiation temperature. The change in the calculated void nucleation rate with irradiation temperature is fairly good agreement with the change in the experimentally observed void number density. The discrepancy between the curves in Fig. 5 at higher temperatures may be caused following the assumption that both the bias factor $\beta_i/\beta_v$ and the incubation period for void nucleation were independent of irradiation temperature. It is, in fact, reported that the bias factor decreases with irradiation temperature\(^{(19)(26)}\), and that the incubation period is shorter at higher temperature\(^{(27)}\).

The shift of the peak swelling temperature to a higher temperature as proceeding the irradiation is arised from the temperature dependence of fluence exponent and void density; the increase of the irradiation temperature increases the fluence exponent and decreases the void number density. In a low dose region, void size is very small and the void number density is dominant to the swelling, thereby causing relatively large swelling in the specimens irradiated at lower temperatures. The void size effect on the swelling becomes greater than that of the void density as the electron irradiation proceeds. Therefore, the peak swelling temperature shifts to a higher temperature with increase in electron dose.

### V. CONCLUSIONS

The dose and temperature dependence of swelling in Type 316 stainless steel electron-irradiated to a dose of about 30 dpa in the temperature range of 370~630°C were studied using a HVEM, and the conclusions obtained are as follows:

1. Both the void and dislocation densities increase steeply with dose in the early stage of the irradiation and become nearly constant beyond a dose of about 5 dpa.
2. The swelling is well described with $\frac{J}{V}/V(%) = A(\text{dpa})^n$ as a function of electron dose. The increase of the irradiation temperature increases the fluence exponent $n$, and decreases the pre-exponential term $A$ which is proportional to the void number density.
3. At 550°C irradiation, the fluence expo-
nent $n$ takes the value of 1.5 due to the diffusion-limited void growth.

(4) At high temperature ($>550\degree C$), the surface reaction-limited void growth contributes to the swelling.

(5) The smaller value of $n$ for the low temperature ($\leq 500\degree C$) irradiation appears to result from the dislocation-assisted diffusion.

(6) The peak swelling temperature of the specimen irradiated to 30 dpa is about 570$\degree C$, which shifts to a higher temperature with increase in electron dose.

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