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Srivastava, A.K.

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STAINLESS STEEL

Arvind K. Srivastava
(Ph. D. thesis)

February 1977

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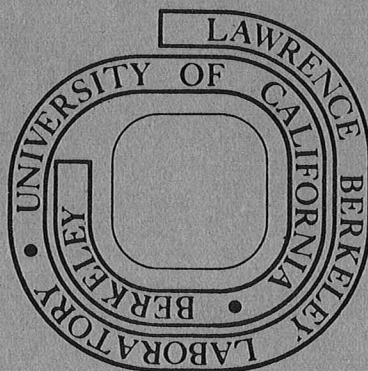
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PROTON INDUCED SWELLING IN TYPE 316 STAINLESS STEEL

Contents

Abstract	v
I. Introduction	1
II. Proton Damage Processes	8
A. Displacement of Atoms	8
B. Theory of Radiation Induced Swelling	13
C. Sputtering	16
D. Retention of Hydrogen	17
III. Experimental Procedure	19
A. Description of Irradiation Facility	19
B. Sample Holder Assembly	20
C. Sample Preparation and Swelling Measurement	21
IV. Results	23
V. Discussion	26
VI. Conclusions	31
ACKNOWLEDGEMENTS	32
References	33
Tables	38
Figure Captions	40
Figures	41

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PROTON INDUCED SWELLING IN TYPE 316 STAINLESS STEEL

Arvind K. Srivastava

Materials and Molecular Research Division, Lawrence Berkeley Laboratory
and Department of Nuclear Engineering,
University of California, Berkeley, California 94720

ABSTRACT

Annealed Type 316 stainless steel was irradiated by 140 keV protons at 625°C in vacuum ($\sim 1 \times 10^{-8}$ Torr) to study its swelling behavior. The maximum fluence used in the investigation, 2×10^{20} protons/cm², created a peak damage of 590 dpa (displacements per atom) based on 25 eV as the displacement threshold. The swellings integrated along the entire proton range were obtained from the measurements of the step-heights at the junction of the irradiated and unirradiated regions. The swelling-dpa relationship was ascertained from the integrated swellings measured at various doses and the corresponding depth-dpa profiles. There were three stages in the swelling regime. The rate of swelling in stage I (dpa < 200) increased continuously up to 0.8% per dpa. In stage II (425 > dpa > 200) the rate of swelling remained at its highest (0.8% per dpa). Rapid reductions in the rate began to occur beyond 425 dpa and the swelling saturated at about 260% at 500 dpa. The values of swelling at 200 dpa and 425 dpa were 46% and 226% respectively. The entire swelling regime could be explained by assuming reasonable changes in the dislocation density and void density as the void volume increases. To fit the experimental results it was necessary to assume that dislocations lying within a spherical shell of thickness 150Å around the voids did not contribute to void growth because of the effects of Pipe Diffusion, and that the dislocation's capture cross-section for interstitials was 4.2% higher than that for vacancies when the assumed displacement threshold (E_d) was 25 eV or alternatively 7.5% higher if E_d was taken as 40 eV. The reduction in the rate of swelling due to Pipe Diffusion amounted to 25% at a dislocation density of 4×10^{11} /cm² and 5% at 5×10^{10} /cm² when the inter-void spacing was larger than 2000Å.

I. INTRODUCTION

Numerous investigations have been carried out to examine neutron induced voids in metals and alloys¹⁻²⁵ since Cawthorne and Fulton¹ first reported void formation in Type 316 stainless steel which had been irradiated in the Dounreay Fast Reactor in U.K. A considerable effort in recent years has been made toward simulating fast neutron damage that in a typical reactor will occur in a period of a few years by using energetic charged particle irradiations such as electrons, protons, or ions of C, Ni, etc.^{3-9,26-35} Use of charged particles produces displacement of atoms at a rate thousands of times faster than in nuclear reactors. For example, fluxes of nickel ions and protons are now available that can produce about 2×10^{-2} displacements per atom (dpa) per sec and 2×10^{-3} dpa/sec respectively in comparison to $\sim 10^{-6}$ dpa/sec in a fast nuclear reactor. During irradiation lattice atoms are displaced creating interstitial-vacancy pairs. These point defects may diffuse in the lattice to form aggregates of their own kind. Three dimensional aggregates of vacancies i.e., voids have been found to grow during irradiation in the temperature range $0.3 T_M$ to $0.5 T_M$ causing swelling of the material. T_M is the absolute temperature of melting.

An interesting question which has yet to be answered is whether or not saturation of swelling would occur beyond a certain number of dpa in various structural materials. Saturation in this sense means no further swelling with further increase in dpa.

One of the materials most extensively studied has been Type 316 stainless steel (S.S. 316) because of its choice as a fuel cladding

material for the liquid-metal-cooled fast breeder reactors. Past accelerator simulation studies show a great deal of controversy over saturability of S.S. 316 swelling at high doses (greater than 100 dpa). The first report of swelling saturation was a result of work at Harwell using carbon ion bombardment. Mazey et al.³⁰ irradiated S.S. 316 containing 10 ppm He by 20 MeV carbon ions at 525°C. Their results show a saturation of swelling of about 12% at 400 dpa. Saturation of swelling in S.S. 316 was further suggested by 5 MeV nickel ion irradiation data of Hanford Engineering Development Laboratory.³⁶ A saturation value of 8% swelling at 450 dpa and 585°C was obtained.

In contrast to the above results the work of Keefer et al.²⁷ and of Johnston et al.³⁴ apparently demonstrated that saturation of swelling did not occur in S.S. 316. Irradiation experiments of Keefer et al.²⁷ using 1 MeV protons produced 20% volume increase at 50 dpa and 500°C with no sign of saturation.

Johnston et al.³⁴ have irradiated S.S. 316 containing no He by 5 MeV nickel ions to produce displacements as high as 230 dpa and found no indication of saturation of the swelling. Swelling at 230 dpa was about 20%. The corresponding swelling in S.S. 316 containing 15 ppm He was about 50%. All irradiations were done at 625°C which has been established as the temperature at which swelling is maximum in this material for equal doses.

There have been several mechanisms proposed which may limit the void growth and hence, may contribute toward saturation of swelling at high doses. One is the increasing dislocation density with fluence.³⁷ In the early stages of irradiation agglomeration of

interstitials produces dislocation loops. These loops have a larger interaction energy with interstitials than with vacancies because of larger lattice distortion around interstitials. Hence, interstitials are preferentially attracted toward these loops causing them to grow. On the other hand, any vacancy dislocation loops that may be nucleated would shrink and disappear because of the preferential absorption of interstitials. However, three dimensional aggregates of vacancies i.e., voids are usually also nucleated during irradiation with the help of insoluble gases such as helium (frequently on precipitate interfaces). Since irradiation produces vacancies and interstitials in equal numbers, and dislocations attract interstitials preferentially excess vacancies over interstitials are left to migrate to alternative sinks such as the voids, resulting in their growth. Growth of the interstitial dislocation loops causes increasing bias towards interstitials resulting in an increase in the rate of void growth. However, dislocations also absorb vacancies. Increasing dislocation density to the point where dislocation sinks predominate over all others leaves few vacancies to diffuse to voids. Thus, there should be two regions in the rate of void growth, an increasing rate followed by a decrease in the rate of growth. The optimum dislocation density at which the rate of swelling is highest is of the order of $10^{10}/\text{cm}^2$.

Experimental evidence of a low rate of swelling in cold worked S.S. 316 containing very high dislocation density ($>10^{11}/\text{cm}^2$) has been demonstrated in the neutron irradiation experiments of Straalsund et al.,³⁸

Stiegler and Bloom,²⁵ and Brager.²⁴ Increasing cold work resulted in a decrease in the rate of swelling.²⁴ Reduction in the rate of swelling by cold work has been further demonstrated in the proton irradiation results of Keefer and Pard,²⁹ nickel ion bombardment results of Johnston et al.,³⁴ and carbon ion irradiation experiments of Mazey et al.³⁰ In contrast, Norris³⁹ found a reduction in the swelling of nickel containing a low density of dislocations ($<10^9/\text{cm}^2$).

Another mechanism that may contribute to saturation of swelling is the interstitial injection into the voids by focused replacement collision sequences (FRCS).⁴⁰ If a lattice atom is struck by the irradiating particle or another atom such that the transferred energy is less than the displacement threshold the lattice atom may not be displaced to form a vacancy-interstitial pair in the bulk. However, if the struck lattice atom is within a few atomic distances (approximately 10) of the surface of a void and if the direction of the momentum transfer is nearly the same as that of a close-packed direction of atoms then the struck lattice atom could get displaced replacing a neighboring atom which in turn may collide with the next neighbor and thus, the sequence will propagate along the row. When such a sequence of replacement collisions intersects the surface of a void a surface atom will be ejected. The atom thus ejected will decrease the size of the void. Some of the vacancies created in the bulk by FRCS may diffuse back into the void. However, a fraction of the vacancies will diffuse into the lattice and may be lost at

dislocations or grain boundaries resulting in a net void shrinkage.

Clear evidence of such vacancy injection has been narrated by Nelson⁴⁰ wherein gold was irradiated by 1 MeV electrons. Electrons of this energy transfer only about 21 eV to gold atoms, which is below the displacement threshold of 35 eV. Transmission electron microscopy examination showed a large density of vacancy tetrahedra extending hundreds of Angstroms below the surface. It was suggested that such tetrahedra were formed from vacancies left in the sample as a consequence of FRCS intersecting the surface.

Hardness and Li⁴¹ have suggested Pipe Diffusion of point defects along dislocation core as a possible means by which rapid recombination of vacancies diffusing out of the void along an intersecting dislocation and interstitials arriving at the dislocation from the bulk could be achieved. Flow of vacancies out of the void will lead to shrinkage of the void. However, this recombination of vacancies and interstitials by Pipe Diffusion can take place in the vicinity of a void only within a spherical shell of thickness $\bar{y}$, the mean free path of vacancies along a dislocation core. $\bar{y}$ is the distance a vacancy can move away from the void along an intersecting dislocation before evaporating into the bulk. Thus, Pipe Diffusion has the effect of making dislocations intersecting the voids, within a spherical shell of thickness $\bar{y}$, incapable of providing the necessary bias for interstitials that causes void growth.

Experimental evidence of the effect of Pipe Diffusion on void growth was demonstrated by Norris.⁴² It was shown that under electron irradiation of nickel at 450°C a void connected to foil surfaces (sink for vacancies) by dislocations shrank as opposed to voids that were not so connected.

When the contribution to void growth due to preferential attraction of interstitials to dislocations (bias reduced due to the effects of pipe diffusion) is balanced by void shrinkage resulting from the injection of interstitials into voids due to FRCS saturation of swelling would occur.

The problems related to the study of saturation effects are long irradiation times and measurements of large amounts of swelling (>50%).

Longer times of irradiation may cause increased contamination of the sample. As shown in Fig. 6 the energy loss in nuclear collisions and hence, the rate of displacement of atoms increases with depth first, and then drops down quickly. Hence, to produce saturation along the entire range in the shortest length of time the penetration depth of protons should be as small as possible. However, since a free surface provides a sink for point defects the penetration depth should not be less than $2000\text{\AA}$ to keep surface effects on swelling at a minimum. Contamination of sample from hydrocarbons due to pump oil in the system must be avoided by using oil-free pumps on the irradiation chamber. Also, an oil trap must be placed between the beam tube and the chamber to prevent creeping of oil along the tube into the chamber.

Most measurements of swelling have been made by TEM. The method is very attractive since voids can be directly observed so that their shapes, densities could be determined. However, there are great limitations to this method when large swellings are being measured. Due to the irregular shapes of the voids significant errors are introduced in calculating their volumes. When the void sizes are comparable to the thickness of the foil the measurements could be quite unreliable. The inhomogeneous distribution of voids makes the problem worse because of the difficulty

in choosing typical regions in which to make measurements.

Surface profilometry was used by Johnston et al.⁴³ to determine swelling caused by nickel ions by measuring heights of steps formed on the irradiated surface. Sample dimensions were chosen such that they were very large (approximately a factor of 100 or greater) compared to the penetration depth of the nickel ions. Hence, the irradiated volume was constrained from swelling in all directions except perpendicular to surface. Thus, swelling produced a step, between the unirradiated and irradiated surfaces, proportional to the integrated swelling along the entire ion range, minus sputtering of the surface by ions plus elevation of surface due to the deposited nickel. The greatest disadvantage of this method is that the swelling measured is not at one single dpa. Hence, several irradiations at various doses have to be done to determine the swelling-dpa relationship as explained in Chapter IV of this thesis.

Johnston et al.³⁴ compared swellings determined by step-height measurements and TEM measurements and found the former larger by a factor of about 1.5. However, they consider the former measurements more reliable since it is more direct.

The present investigation was undertaken to determine whether saturation of swelling would occur in S.S. 316 when it is irradiated by 140 keV protons to hundreds of displacements per atom at 625°C. Surface profilometry (method of step height measurements) was used to determine swelling.

II. PROTON DAMAGE PROCESSES

An energetic proton may cause sputtering of surface atoms or lattice atom displacements creating vacancy-interstitial pairs. Sputtering results in erosion of the material whereas formation and growth of voids due to clustering of vacancies may cause swelling. The implanted hydrogen may also contribute to the growth of voids, and therefore, to swelling.

A. Displacement of Atoms

As the proton enters the sample it begins to interact with atoms in the solid and loses energy in both inelastic and elastic collisions and thus slows down as it penetrates further into the solid. In elastic collisions with the lattice atoms the proton gets deflected from its original path. If the energy transferred to the lattice atom is greater than the displacement threshold the atom will be displaced leaving a vacant site and an interstitial atom. The primary knock-on atom (PKA) can in turn displace further lattice atoms. The inelastic collisions involve electronic excitation of atoms or ionization, and do not result in atomic displacements. The two types of collisions can be treated separately since, unlike the elastic collisions, in the inelastic collision there is little momentum transfer from the proton even though there can be a large energy transfer to the electrons. The separate treatment of inelastic (or electronic) and elastic (or nuclear) interactions results in a great simplification because nuclear scattering can be described by classical mechanics. Loss of energy by inelastic collisions predominates at energies much greater than M_1 keV (M_1 is Mass No. of projectile) whereas loss

by nuclear collisions is dominant at lower energies.

The nuclear energy loss curve for 140 keV protons (Fig. 6) used in the present investigation was obtained from Oen.⁴⁴ The method of calculation has been described by Oen et al.⁴⁵ To calculate the loss of energy due to nuclear collisions Oen⁴⁴ used the analytical approximation by Winterbon et al.⁴⁶ for the numerical form of the differential nuclear scattering cross-section developed by Lindhard et al.⁴⁷ Thomas-Fermi interaction potential was used in the development of the scattering cross-section. The analytical approximation⁴⁶ for the cross-section is

$$d\sigma = \frac{1}{2} \pi a^2 \lambda q^{-4/3} [1 + (2\lambda q^{2/3})^{2/3}]^{-3/2} dq \quad (1)$$

where $\lambda = 1.309$

$$q = \epsilon^2 \sin^2 \frac{\theta}{2} \quad (2)$$

where θ = center of mass scattering angle and

$$\epsilon = \frac{EM_2 a}{(M_1 + M_2) Z_1 Z_2 e^2} = \frac{E}{E_N} \quad (3)$$

Subscripts 1 and 2 refer to projectile ion and target atom respectively. M and Z are the mass and atomic number respectively, e is the electronic charge, E is the energy of the projectile ion, ϵ is the non-dimensional energy parameter and a is the screening length (Å).

$$a = 0.8853 a_o \left[Z_1^{2/3} + Z_2^{2/3} \right]^{-1/2} \quad (4)$$

where $a_o = 0.529\text{Å}$ is the Bohr radius.

The average loss of energy in nuclear collisions that results in displacement of atoms is

$$\frac{dE}{dL} = N \int_{E_d}^T T d\sigma \quad (5)$$

where T = transferred energy to the lattice atom when the energy of projectile is E , N is density of lattice atoms. E_d is the displacement threshold energy, T_m is the maximum energy transfer that can occur between the projectile and the lattice atom and L is the path length.

$$T = \frac{4M_1 M_2}{(M_1 + M_2)^2} \cdot E \cdot \sin^2 \frac{\theta}{2} = T_m \sin^2 \frac{\theta}{2} \quad (6)$$

In terms of the dimensionless path length parameter

$$\rho = \frac{LN^2 4M_1 M_2}{(M_1 + M_2)^2} = \frac{L}{L_N} \quad (7)$$

The dimensionless nuclear stopping power can be expressed as

$$\left(\frac{d\varepsilon}{d\rho} \right)_n = \left(\frac{L_N}{E_N} \right) \frac{dE}{dL} \quad (8)$$

Performing the integration in Eq. (5) one obtains

$$\begin{aligned} \left(\frac{d\varepsilon}{d\rho} \right)_n = \frac{9}{8\varepsilon} & \left\{ \ln \left[\frac{(2\lambda)^{1/3} \varepsilon^{4/9} + \sqrt{1 + (2\lambda)^{2/3} \varepsilon^{8/9}}}{(2\lambda)^{1/3} \gamma^{4/9} + \sqrt{1 + (2\lambda)^{2/3} \gamma^{8/9}}} \right] \right. \\ & \left. + \frac{(2\lambda)^{1/3} \gamma^{4/9}}{\sqrt{1 + (2\lambda)^{2/3} \gamma^{8/9}}} - \frac{(2\lambda)^{1/3} \varepsilon^{4/9}}{\sqrt{1 + (2\lambda)^{2/3} \varepsilon^{8/9}}} \right\} \end{aligned} \quad (9)$$

where

$$\gamma = \varepsilon \sqrt{\frac{E_d}{T_m}} \quad (10)$$

Equation (9) gives the nuclear energy loss per unit path length.

Whereas the loss of energy by the ion in nuclear collision can be described by Eq. (9), theories of electronic stopping are inadequate. White and Mueller⁴⁸ have experimentally determined the electronic energy loss in Fe by protons with energies 140 keV and below. These values have been used by Oen⁴⁴ in the calculation of depth--nuclear energy loss profile for 140 keV protons in Fe. Assuming little difference between the loss of energy by protons in collision with the atoms of Fe and of S.S. 316 the nuclear energy loss curve for Fe was used to calculate the rates of displacement of atoms in S.S. 316.

Total energy loss per unit path length is the sum of the nuclear and electronic losses.

$$\left(\frac{d\epsilon}{d\rho}\right)_{\text{total}} = \left(\frac{d\epsilon}{d\rho}\right)_n + \left(\frac{d\epsilon}{d\rho}\right)_e \quad (11)$$

and average total path is

$$\bar{\rho} = \int_0^{\epsilon_0} \left[\left(\frac{d\epsilon}{d\rho}\right)_{\text{total}} \right]^{-1} d\epsilon \quad (12)$$

where ϵ_0 is evaluated at the target surface.

In the case of protons slowing down in the stainless steel $\bar{\rho}$ can be significantly different from the penetration depth $\bar{\rho}_d$ measured in the direction of the proton beam at the surface. This is because of large deflection of the protons in elastic collisions with atoms having $M_2 \gg M_{\text{proton}}$.

$$\bar{\rho}_d = \int_0^{\bar{\rho}} \langle \cos\phi \rangle d\rho \quad (13)$$

where $\langle \cos\phi \rangle$ is the average value of the cosine of the scattering angle in the laboratory system of coordinates.

$$\langle \cos \phi \rangle = \exp \left[- \int_0^{\rho} N L_N d\rho \int_0^{\epsilon^2} (1 - \cos \phi) \frac{d\sigma}{dq} dq \right] \quad (14)$$

ϕ is related to q through Eq. (2) and

$$\tan \phi = \frac{\sin \theta}{\frac{M_1}{M_2} + \cos \theta} \quad (15)$$

The quantity of interest in dose-dpa calculations is the nuclear energy loss per unit penetration depth. This is equal to

$$\frac{d\epsilon}{d\rho_d} = \frac{1}{\langle \cos \phi \rangle} \frac{d\epsilon}{d\rho}_n \quad (16)$$

Slowing down of ions is a statistical process resulting in straggling of their penetration depth. Winterbon⁴⁹ has shown that the distribution of penetration depths is Gaussian. Using this result the average energy loss in nuclear collisions per unit penetration depth is

$$\overline{\frac{d\epsilon}{d\rho_d}}(\rho_d) = \int_0^{\infty} \frac{d\epsilon}{d\rho_d} (\rho_d' - \rho_d) \frac{1}{\sqrt{2\pi} \alpha} \times \exp \left\{ - \frac{(\rho_d' - \bar{\rho}_d)^2}{2\alpha^2} \right\} d\rho_d' \quad (17)$$

where

$$\alpha = \sqrt{\rho_d^2 - (\bar{\rho}_d)^2} \quad (18)$$

α is the straggling parameter. The number of interstitial-vacancy pairs per unit path length created by the proton was obtained by multiplying the nuclear energy loss (Eq. (9)) by $(\eta/2E_d)$ where E_d is the displacement threshold (25 eV) and $\eta = 0.8$ based on the work of Robinson.⁵⁰ The loss of PKA energy in secondary ionization is neglected because most recoils (~90%) by protons are less than 250 eV.

$$\frac{\overline{\Delta E}}{\Delta x} \text{ (nuclear collisions)} = \frac{E_N}{L_N} \cdot \frac{d\epsilon}{d\rho_d} \quad (19)$$

where x , ρ_d are the actual and non-dimensional lengths, respectively, along the direction of proton beam at the target surface.

Oen⁴⁴ has calculated $\frac{\overline{\Delta E}}{\Delta x}$ versus depth using Eq. (1) through Eq. (19) with $\alpha = 0.1 \mu\text{m}$. The curve is shown in Fig. 6. The lower cut off for transferred energy (E_d) for the curve is 20 eV. Choice of 25 eV for E_d introduces less than 5% error in the calculation of $\frac{\overline{\Delta E}}{\Delta x}$.

B. Theory of Radiation Induced Swelling

Bullough and Perrin⁵¹ developed expressions to predict the rate of growth of voids under irradiation. For mathematical simplicity, it was assumed that voids of concentration n_v per cm^3 were distributed uniformly throughout the matrix which was divided into n_v spheres of radius R , where

$$R = \left(\frac{3}{4\pi n_v} \right)^{1/3} \quad (20)$$

The dislocation density (n_d per cm^2) was considered, again for simplicity, as uniform throughout the matrix. The dislocation density is a sum of the dislocations before the start of irradiation and the dislocation loops that form during irradiation. To allow for the

preferential attraction toward interstitials the dislocations were assumed to have a larger capture cross-section for interstitials than for vacancies. Swelling could be considered as the sum of the volume increase of all idealized spherical voids of radius r_v during irradiation due to the presence of dislocations lying in the spherical shell of which the inner and outer radii are r_v and R respectively. The boundary at radius R represents the interface midway between neighboring voids.

The quasi-steady state fractional vacancy and interstitial concentrations $C_v(r)$ and $C_i(r)$ respectively in the spherical shell around each void are

$$D_v \left(\frac{d^2 C_v}{dr^2} + \frac{2}{r} \frac{dC_v}{dr} \right) + K - \alpha_R C_v C_i - D_v z_v n_d C_v = 0 \quad (21)$$

and

$$D_i \left(\frac{d^2 C_i}{dr^2} + \frac{2}{r} \frac{dC_i}{dr} \right) + K - \alpha_R C_v C_i - D_i z_i n_d C_i = 0 \quad (22)$$

where D_v and D_i are the vacancy and interstitial diffusion coefficients, respectively. α_R is the recombination coefficient. z_v and z_i are numbers, both of order unity, that characterize the capture cross-sections of dislocations for vacancies and interstitials, respectively ($z_i > z_v$). K is the rate of production of vacancies and interstitials.

$$K = \frac{\eta}{2E_d} \cdot \frac{\overline{\Delta E}}{\Delta x} \cdot \frac{I}{N} \quad (23)$$

where I is the proton current density expressed as protons per cm^2 per sec and N is atoms per cm^3 in the target S.S. 316.

$$\text{Percent swelling (S)} = \frac{V_v}{V_i} \times 100 = \frac{r_v^3}{R^3 - r_v^3} \times 100 \quad (24)$$

where V_v = void volume in the matrix

V_i = volume of the matrix prior to irradiation

The boundary conditions are obtained by putting zero-flow conditions across the spherical surface at $r = R$, i.e.,

$$\frac{dC_v}{dr} = 0$$

$$\frac{dC_i}{dr} = 0$$

And, at the surface of the void $r = r_v$

$$C_i = 0$$

$$C_v = \bar{C} = C_v^e \exp[\{(2\gamma_s/r_v) + P\} \Omega/kT_a]$$

where P is the hydrostatic compressive stress acting on the void, C_v^e is the equilibrium concentration of vacancies in the bulk, Ω is the atomic volume, k is the Boltzmann's constant, T_a is the absolute temperature, and γ_s is the surface energy. Pressure in the void due to any gas atoms present has been neglected in comparison to $\{(2\gamma_s/r_v) + P\}$, ($\gamma_s \approx 1000 \text{ ergs/cm}^2$).

For obtaining analytical solution to the coupled diffusion Eqs. (21) and (22) the recombination term $\alpha_R C_v C_i$ may be neglected. Since recombination effects are not important at the temperature at which maximum void growth occurs (625 C for S.S. 316) the omission is justified. From the solution to Eqs. (21) and (22) the rate of growth of the void can be obtained as

$$\frac{dr_v}{dt} = \frac{(K - D_v z_v n_d \bar{C})}{r_v z_v n_d} L(\beta_v) - \frac{K}{r_v z_i n_d} L(\beta_i) \quad (25)$$

where

$$L(\beta) = \frac{\left[(R - r_v) \beta \cosh \beta(R - r_v) + (r_v R \beta^2 - 1) \sinh \beta(R - r_v) \right]}{[R \beta \cosh \beta(R - r_v) - \sinh \beta(R - r_v)]}$$

and $\beta = (z n_d)^{1/2}$, t is the time. For r_v greater than $100\text{\AA}$ the term $D_v z_v n_d \bar{C}$ is negligible compared to K ($\sim 10^{-3}/\text{sec}$) even when P reaches the yield point for S.S. 316. The diffusion coefficient and the equilibrium concentration of vacancies were calculated from the data of Ref. 51 using the following equations.

$$D_v = D_v^0 e^{-E_v^m/kT_a} \quad (26)$$

$$C_v^e = e^{-E_v^f/kT_a} \quad (27)$$

$$D_v^0 = 0.58 \text{ cm}^2/\text{sec}$$

$$E_v^m = 1.4 \text{ eV}$$

$$E_v^f = 1.6 \text{ eV}$$

$$\text{Rate of Swelling (percent per dpa)} = \frac{1}{K} \frac{4\pi r_v^2 (dr_v/dt)}{(4/3)\pi (R^3 - r_v^3)} \times 100 \quad (28)$$

C. Sputtering

Sputtering of atoms from the surface occurs when surface atoms receive energies in excess of the surface binding energy. This energy transfer may occur either in direct collision with the protons or in collision with energetic atoms displaced from lattice sites as a result of collision cascades. These cascades are created beneath the surface

by Primary Knock On atoms which receive their energies from the protons in nuclear collisions with them.

Extended theoretical treatments^{52,53} have been put forward to calculate sputtering yields, i.e., the average number of sputtered atoms per ion. Summary reports on sputtering are contained in several references.⁵⁴⁻⁵⁷ However, for light ions the agreement between theory and experiment is only within an order of magnitude.

Some experimental data exists on the sputtering yield of S.S. 304 by protons.⁵⁸ Assuming that there is little difference between the sputtering yields of S.S. 304 and S.S. 316 an average value of 5×10^{-4} atoms/proton was used in the present calculations.

D. Retention of Hydrogen

In the present investigation up to 2×10^{20} protons/cm² have been implanted at 625°C, average rate of implantation being 8×10^{14} protons/cm²-sec. If a fraction of the hydrogen implanted is retained in the matrix, it could help in the nucleation and growth of voids.

However, there is experimental evidence which shows that the implanted hydrogen is not retained in stainless steel at 625°C. Keefer and Pard⁵⁹ have examined proton irradiated samples for hydrogen retention using an Ion Microprobe Mass Analyzer. Transmission electron micrograph of the irradiated sample showed that the void volume increased by an order of magnitude in going from the surface to the end of the penetration depth. Also, most of the hydrogen is formed at the end-of-range. Because of largest void volume and of formation of hydrogen, concentration of

hydrogen should be greatest at the end-of-range if voids trap hydrogen. The results of Keefer and Pard⁵⁹ indicated that the hydrogen content was uniform throughout the sample. In view of this finding it was concluded that voids do not trap hydrogen.

III. EXPERIMENTAL PROCEDURE

A. Description of Irradiation Facility

A schematic diagram of the entire set-up is shown in Fig. 1. The proton beam used in the irradiation was obtained from a Van de Graaff accelerator. Species of ions were analyzed by the electromagnet at ⑥ and the desired species was sent into the beam tube ⑦→⑧. At positions ⑦ and ⑪ were two electromagnets with axes at right angles to each other and to the beam tube. These magnets were used to bend the beam in any desired direction. They helped to locate the beam on the target. The magnets were also used to sweep the beam both vertically and horizontally on the irradiated area. This was essential since the beam did not have a uniform intensity across its cross-section. Sweeping and locating the beam on the target was simultaneously accomplished by superimposing an alternating current on a direct current through the electromagnet coils. However, the alternating currents in the two magnets had different frequencies to avoid synchronization. One of the magnets had 60 cps input whereas the other had a variable frequency input (20 sec/cycle to 10 cycles/sec). Very slow sweep helped in determining the magnet current necessary for a desired maximum traverse of the beam during sweeping. While irradiating, the variable sweep speed was increased such that beam intensity on the target did not fluctuate widely.

After the beam left the sweeping magnets it entered the High Vacuum Chamber (HVC) as shown in Fig. 2. The HVC was pumped by a 6 in. DP creating a pressure of 1×10^{-8} Torr to 5×10^{-8} Torr at ③. The creep barrier ② maintained a 6 in. length of the beam tube at liquid nitrogen temperature all the time. This helped stop creeping

of pump oil along the beam tube into the irradiation chamber. Beyond HVC all seals were made up of either oxygen free copper or of gold except three viton O-rings, one in Inline valve at ④ and two in the Gate valve at ⑧. This kept the hydrocarbons at a minimum in the system.

The irradiation chamber (or sample chamber) at ⑥ shown in Fig. 2 was pumped by a 270 l/sec ion pump and a 6 in. titanium sublimation pump (TSP). At ⑤ there was a copper gasket with a 3/8 in. diameter hole through which beam entered the sample chamber. This small hole helped to keep the sample chamber pressure dependent only to a small extent on the pressure of HVC side. Before the start of the experiment the sample chamber was baked at 250°C for 24 hr and then pumped for another 24 hr to 5×10^{-9} Torr. The operating pressure of the sample chamber was about 1×10^{-8} Torr.

This set-up provided a uniform beam intensity on the target area and a clean environment such that samples irradiated for as long as 70 hr at 625°C came out uncontaminated.

B. Sample Holder Assembly

The sample holder assembly is shown in Fig. 3. The sample was held in an alumina block (85% purity). Ta heating wires made in the form of a helix sat in the groove at the bottom. The sample (1 in. dia, 3/8 in. thick) was held in the block in front of the heater wire with a 1/4 in. dia, 1/16 in. thick alumina spacer in between. A collimator made of S.S. 316 (1 in. dia, 1/4 in. thick) was mounted on the sample with the help of three screws made of Ta. The collimator was electrically insulated from the sample by alumina washers. Choice of Ta prevented diffusion bonding of screws to the

sample. The collimator had a 1/8 in. hole with a cross of 10 mil tungsten wires spot welded to it.

The thermal shields ⑥ are electrically connected to the sample. This coupling, when kept at +180 volts above the ground, acts as a faraday cup for beam monitoring.

C. Sample Preparation and Swelling Measurement

Samples (1 in. dia, 3/8 in. thick) were cut from S.S. 316 block prepared by Carpenter Technology, Inc. Sample of such a large size provided good temperature stability during occasional variations in the beam current and also reduced errors in the temperature measurement. Composition of the S. S. 316 is shown in Table I. Samples cut to size were then annealed at 1010°C for 1 hr in argon atmosphere and subsequently water quenched. Sample surface was mechanically polished. Final polishing was done in a vibratory polisher in a slurry of alumina powder having particle size of 500Å.

A 10 mil dia chromel-alumel thermocouple spot welded to the sample surface close to the irradiated area provided the temperature measurement. Temperature variations of $\pm 10^{\circ}\text{C}$ occurred during the experiment.

Figure 4 shows a circular irradiated area. The cross made of 10 mil tungsten wire on the collimator has masked an identical area on the irradiated surface as is evident in the figure. Swelling was measured across the masked area at eight different places. These measurements differed from each other by $\pm 10\%$. Figure 5 shows the profilometer trace of one of the measurements. The irradiated surface was very rough compared to the masked surface. The average

elevation of the irradiated surface above the masked surface was measured to determine swelling.

Clevite surfanalyzer 150 system profilometer was used for the measurements. Stylus had a 5 μ m dia diamond tip and bore on the specimen surface with a force of 50 mg.

IV. RESULTS

Five samples were irradiated at 625°C by 140 keV protons with fluences of $2.25 \times 10^{19}/\text{cm}^2$, $4.5 \times 10^{19}/\text{cm}^2$, $8.1 \times 10^{19}/\text{cm}^2$, $1.45 \times 10^{20}/\text{cm}^2$, and $2.0 \times 10^{20}/\text{cm}^2$. The average dose rate was about $8 \times 10^{14}/(\text{cm}^2\text{-sec})$ with variations of about $\pm 10\%$. Values of swelling, in terms of measured step-heights, of these samples are shown in Fig. 7. Each value is an average of eight measurements in different areas of the irradiated region. Values of step-heights were corrected for sputtering of the surface by protons. A sputtering yield of 5×10^{-4} atoms/proton was used in the calculations. The step-heights (L_{step}) are proportional to the swelling integrated along the entire proton range. The irradiated volume is constrained from swelling in all directions except perpendicular to the surface.

To obtain the swelling-dpa relationship the penetration depth of protons was divided into ten zones. Before irradiation each zone was $1000\text{\AA}$ wide perpendicular to the sample surface. After irradiation each zone width increased by $10 S(i)\text{\AA}$ ($= 1000 * S(i)/100$) where $S(i)$ is the average percent swelling for the i^{th} zone lying between $x(i-1)$ ($= (i-1) * 1000\text{\AA}$) and $x(i)$ ($= i * 1000\text{\AA}$), x being measured from the surface. Also,

$$L_{\text{step}}(\text{\AA}) = \sum_{i=1}^{i=10} (1000 * S(i)/100) \quad (29)$$

However, mass per unit area of the zones did not change during irradiation. The third column in Table II shows delineation of various zones in terms of mg/cm^2 . Table II also shows the average number of displacements per atom produced in each zone ($\text{dpa}(i)$) for all samples irradiated.

$$\text{dpa}(i) = t * K(i) \quad , \quad t = \text{time of irradiation}$$

$K(i)$ was calculated from Eq. (23) with $E_d = 25$ eV and $\eta = 0.8$. The average value of $\frac{\overline{\Delta E}}{\Delta x}$ for the i^{th} zone (i.e., $\frac{\overline{\Delta E}}{\Delta x}(i)$) to be used in Eq. (23) was obtained from Fig. 6. For any given sample the number of displacements rose from zone 1 to zone 7 and then dropped rapidly. The rise from zone 1 to zone 5 was rather slow while it was quite rapid from zone 5 to zone 7. Zone 7 had the highest number of displacements while zone 6 and zone 8 were the next highest. Thus most of the damage occurred in the three zones (6, 7 and 8). The highest number of dpa in zone 7 was 590 for sample 5.

The rate of integrated swelling, defined as the ratio of the changes in measured step-heights and in the proton fluences ($\Delta L_{\text{step}} / \Delta(It)$), appeared to increase up to a dose of about $8 \times 10^{19} / \text{cm}^2$, and then remained at its highest until about $1.4 \times 10^{20} / \text{cm}^2$. The rate then appeared to have decreased. Step-height measured at a fluence of $8.1 \times 10^{19} / \text{cm}^2$ for sample 3 was only $1625 \text{ \AA}$. When the fluence was increased to $1.45 \times 10^{20} / \text{cm}^2$ in sample 4, the measured step-height became $6520 \text{ \AA}$. Thus, for a rise of $6.4 \times 10^{19} / \text{cm}^2$ in the fluence the increase in the measured step-height was about $4900 \text{ \AA}$, the rate of integrated swelling being $7.6 \times 10^{-17} \text{ \AA} / (\text{proton}/\text{cm}^2)$. The magnitude of this ratio decreased to $4.4 \times 10^{-17} \text{ \AA} / (\text{proton}/\text{cm}^2)$ when the fluence was increased from $1.45 \times 10^{20} / \text{cm}^2$ in sample 4 to $2.0 \times 10^{20} / \text{cm}^2$ in sample 5.

Figure 8 shows a swelling curve that gives integrated swellings for every sample irradiated close to the experimentally measured values. The method of obtaining the swelling curve is explained in Chapter V. The integrated swelling for any given sample was calculated by adding

the swellings obtained from the swelling curve for each of the ten zones (Eq. (29)). The swelling curve in Fig. 8 has three stages. In stage I ($\text{dpa} < 200$) the rate of swelling (Eq. (28)), defined as the rate of change of swelling per unit dpa, rises with increasing dose. Stage II ($425 > \text{dpa} > 200$) is marked by a very high rate of swelling ($0.8\%/ \text{dpa}$). In stage III ($\text{dpa} > 425$) the rate of swelling decreases rapidly.

V. DISCUSSION

The swelling behavior of solution annealed Type 316 stainless steel in proton irradiation could be explained in view of the effects of pipe diffusion, the changing dislocation density and void density as void folume increases.

The effects of void density (n_v/cm^3) and dislocation density (n_d/cm^2) on the calculated rate of swelling (Eq. (28)) are plotted in Figs. 9 and 10 respectively. Figure 9 shows that the rate of swelling first increases with an increase in n_{vo} ($=n_v(1 + S/100)$) and then decreases. S is percent swelling (Eq. (24)). n_d was kept constant at $5 \times 10^{10}/\text{cm}^2$ for the calculations. The choice of n_{vo} as the variable has an advantage over n_v since no change in the value of n_{vo} indicates that neither nucleation nor coalescence of voids has taken place. In contrast, n_v decreases continuously by a mere increase in swelling. A decrease in the value of n_{vo} suggests coalescence of voids. Figure 9 also shows that at larger swelling the swelling-void density curve is shifted toward lower n_{vo} . Figure 10 shows the effect of dislocation density on the rate of swelling. The rate increases with n_d until n_d reaches a medium value ($\sim 10^{10}/\text{cm}^2$). The rate then decreases with increasing n_d . A constant value of n_{vo} ($5 \times 10^{13}/\text{cm}^3$) is assumed at 40% swelling ($z_i = 1.042$ and $z_v = 1$ for Figs. 9-11).

The pipe diffusion of vacancies results in a reduction in the rate of void growth. The reduction was calculated by subtracting the excess interstitials over vacancies absorbed by dislocations in a spherical shell of thickness $\bar{y}$ around the voids where $\bar{y}$ is the mean free path of vacancies along a dislocation core.

$$\left(4\pi r_v^2\right) \left(\frac{dr_v}{dt}\right)_{\bar{y}} = \left(4\pi r_v^2\right) \left(\frac{dr_v}{dt}\right)_{\bar{y}=0} - \int_{r_v}^{(r_v+\bar{y})} (4\pi r^2) (D_i C_i z_i n_d - D_v C_v z_v n_d) dr$$

The various parameters in the above equation are defined in Chapter II. $\bar{y} = 0$ means no effect of pipe diffusion. $\left(\frac{dr_v}{dt}\right)_{\bar{y}=0}$ was obtained from Eq. (25). The reduced rate of swelling was then computed from Eq. (28) using $\left(\frac{dr_v}{dt}\right)_{\bar{y}}$. The calculated reductions in the rate of swelling were about 25% and 5% for n_d equal to $4 \times 10^{11}/\text{cm}^2$ and $5 \times 10^{10}/\text{cm}^2$ respectively when $\bar{y} = 150\text{\AA}$. These calculations were made when the void separation $(2R - 2r_v)$ was greater than $2000\text{\AA}$ where r_v is the void radius and R is defined by Eq. (20).

The swelling curve (Fig. 8) was obtained by trial and error by developing several functions $S(\text{dpa}(i))$, i being the zone number in the irradiated sample, such that for each function the calculated values of L_{step} (Eq. (29)) for every sample irradiated were in agreement with the experimental values (Fig. 7). Amongst the several functions $S(\text{dpa})$ thus determined the function of which the slopes (i.e., rates of swelling) could be obtained from Eq. (28) by assuming continuous changes in n_{vo} and n_d was selected as the swelling curve. However, to satisfy the above conditions in the choice of the swelling curve it was necessary to assume that $\bar{y} = 150\text{\AA}$. Figure 11 shows the changing parameters n_{vo} and n_d required to obtain the swelling curve. The assumptions of n_{vo} and n_d appear to be reasonable based on the work of Johnston et al.³⁴ and McDonald and Taylor.³¹

In the nickel-ion irradiation of S.S. 316 at 625°C by Johnston et al.³⁴ void density first increased from about $2.5 \times 10^{13}/\text{cm}^3$ at 0.2% swelling to $6 \times 10^{13}/\text{cm}^3$ at 1% swelling. Void density then decreased slightly to about $5 \times 10^{13}/\text{cm}^3$ at 20% swelling indicating no further nucleation of voids in this range (1% to 20% swelling). In nickel ion irradiation of S.S. 304 at 600°C by McDonald and Taylor³¹ the dislocation density was measured to be of the order of $10^{11}/\text{cm}^2$ for values of swelling less than about 10%. Dislocation density then appeared to decrease with increasing swelling.

Figure 11 shows that the calculated value of n_{vo} remains unchanged at $5 \times 10^{13}/\text{cm}^3$ for $50 < \text{dpa} < 150$ ($4\% < S < 20\%$) i.e., no further nucleation takes place in this range. However, dislocation density decreases from $4 \times 10^{11}/\text{cm}^2$ at 4% swelling to $1.5 \times 10^{11}/\text{cm}^2$ at 20% swelling. The loss in the dislocation density occurs because of the image force attraction of dislocations towards void surfaces.⁶² The void diameters at 4% swelling and 20% swelling are about 1150Å and 2000Å respectively. The corresponding void separations ($2R - 2r_v$) are 2270Å and 1600Å respectively. Thus, void diameter has exceeded the void separation at 20% swelling and coalescence begins to occur. At 200 dpa (46% swelling) the void diameter and void separation are 2600Å and 1220Å respectively. At such proximities of voids coalescence is very significant, such that void number density (n_{vo}) decreases in stage II from $4 \times 10^{13}/\text{cm}^3$ at 200 dpa to $4 \times 10^{12}/\text{cm}^3$ at 425 dpa while swelling increases from 46% to 226%, i.e., the irradiated volume increases by a factor of 2.2 ($= (226+100)/(100+46)$). The dislocation density, however, decreases by only a factor of about 3 during stage II

from $7 \times 10^{10}/\text{cm}^2$ at 200 dpa to $2.5 \times 10^{10}/\text{cm}^2$ at 425 dpa. The rate loss of dislocations in stage II is lower than in stage I since most dislocations surrounding the voids were annihilated in stage I due to the image force attraction. At 425 dpa the void diameter and the void separation are about $1 \mu\text{m}$ and $1320 \text{\AA}$ respectively. At such proximities and large void diameters almost all climbing dislocations must terminate at neighboring void surfaces. This results in a rapid loss of the dislocation density in stage III (dpa > 425) and consequently, rapid reductions in the rate of swelling occur with increasing dpa in stage III. There is no change in the value of n_{vo} in stage III since the irradiated volume increases by only about 10% in this region.

The values of z_i and z_v , the preference factors of dislocations for interstitials and vacancies respectively, used to obtain the swelling curve were 1.042 and 1 respectively. However, the dpa were calculated based on 25 eV as the displacement threshold (E_d). If E_d were chosen as 40 eV, the generally accepted value at present,⁶⁰ then calculated dpa values are reduced by a factor of 1.8 and hence, to obtain the required rates of swelling (per dpa) z_i should be increased to 1.075. This value of z_i is closer to 1.08 as suggested by R. Bullough.⁶¹

Since swelling reaches as high as 260% in the present investigation and the entire swelling behavior including saturation effects appears to be explainable on the basis of reasonable estimates of changes in void number density and dislocation density the experiments do not provide any evidence for a significant contribution to saturation of focussed replacement collision sequences that inject the interstitials directly into the void.

Figure 12 shows a comparison of swelling in annealed S.S. 316 due to proton (present investigation) and nickel ion irradiations.³⁴ Johnston et al.³⁴ chose 33 eV for displacement threshold in their calculations of the number of displacements due to nickel ions in contrast to 25 eV in the present work. Hence, the number of displacements, as reported by Johnston et al.,³⁴ were multiplied by $\left(\frac{33}{25}\right)$ for the comparison in Fig. 12. Proton induced swelling was about a factor of 10 higher than the swelling due to the nickel ions. A similar relationship for S.S. 304 was observed by Taylor and McDonald.⁶³ At 16 dpa and 600C proton irradiation⁶³ produced 1.4% swelling in comparison to 0.1% swelling at 18 dpa and 600C due to nickel ion irradiation.⁶³ The large discrepancy between swellings due to proton and nickel ion bombardments could be explained in view of the manner in which the displacements are created by the two ions. Figure 6 shows that at a depth of about $6500\text{\AA}$ the nuclear energy loss is maximum. Around this peak the proton creates approximately an average of only 6 interstitial-vacancy pairs within a depth of $1000\text{\AA}$. The point defects thus created are free to migrate toward voids, dislocations, grain boundaries, etc and may annihilate themselves through recombination only by random walk. In contrast, the nickel ion creates about 1600 $\left(\sim 1200 \times \frac{33}{25}\right)$ vacancy-interstitial pairs,³² in the form of a cascade, in the peak swelling region within a depth of $1000\text{\AA}$. Many of these point defects should be expected to annihilate themselves by recombination within the collision cascade itself. Hence, only a fraction would be left to migrate to voids and other sinks. Thus, for the same calculated dpa for nickel ion and proton irradiations swelling would be less for the former.

VI. CONCLUSIONS

- (1) There were three stages in the swelling regime of Type 316 stainless steel due to proton irradiation at 625C.
 - (a) Stage I (dpa < 200). Rate of swelling increased continuously up to 0.8% per dpa with increasing dose (assumed displacement threshold was 25 eV).
 - (b) Stage II (425 > dpa > 200). Rate of swelling remained high (0.8% per dpa).
 - (c) Stage III (dpa > 425). Rate of swelling decreased continuously with dose. Saturation occurred at about 500 dpa and 260% swelling.
- (2) Values of swelling at 200 dpa and 425 dpa were 46% and 226% respectively.
- (3) The observed swelling behavior of Type 316 stainless steel could be explained by assuming reasonable changes in dislocation density and void density as irradiation progresses.
- (4) The contribution of pipe diffusion in reducing the rate of swelling was estimated to be significant when the dislocation density was of the order of $10^{11}/\text{cm}^2$, amounting to 25% at $4 \times 10^{11}/\text{cm}^2$.
- (5) A reasonably good fit between the experimental and calculated swelling curves required a 4.2% greater capture cross-section for interstitials than that for vacancies at the dislocations when the displacement threshold (E_d) was assumed to be 25 eV or alternatively 7.5% greater if E_d was taken as 40 eV.

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REFERENCES

1. C. Cawthorne and E. J. Fulton, Nature 216, 575 (1967).
2. Proceedings of a Symposium on Radiation Damage in Reactor Materials, Vienna, IAEA (1969), Vol. II.
3. Irradiation Effects on Structural Alloys for Nuclear Reactor Applications, ASTM STP 484 (1970).
4. S. F. Pugh, M. H. Loretto and D. I. R. Norris, eds., Voids Formed by Irradiation of Reactor Materials (British Nuclear Energy Society, 1971).
5. J. W. Corbett and L. C. Ianniello, eds., Proceedings of the 1971 International Conference on Radiation-Induced Voids in Metals (USAEC, Oak Ridge, TN, 1972).
6. Effects of Radiation on Substructure and Mechanical Properties of Metals and Alloys, ASTM STP 529 (1973).
7. R. S. Nelson, ed., The Physics of Irradiation Produced Voids, AERE-R7934 (1975).
8. D. I. R. Norris, Rad. Eff. 14, 1 (1972), Part I.
9. D. I. R. Norris, Rad. Eff. 15, 1 (1972), Part II.
10. G. L. Kulcinski, B. Mastel and H. E. Kissinger, Acta Met. 19, 27 (1971).
11. S. D. Harkness and Che-Yu Li, Met. Trans. 2, 1457 (1971).
12. R. C. Ran, R. L. Ladd and J. Moteff, J. Nucl. Mater. 33, 324 (1969).
13. J. O. Stiegler and E. E. Bloom, Rad. Eff. 8, 33 (1971).
14. E. E. Bloom, J. O. Stiegler and C. J. McHargue, Rad. Eff. 14, 231 (1972).
15. L. M. Leitnaker, E. E. Bloom and J. O. Stiegler, J. Nucl. Mater 49, 57 (1973/74).

16. H. R. Brager and J. L. Straalsund, J. Nucl. Mater. 46, 134 (1973).
17. E. E. Bloom and J. O. Stiegler, J. Nucl. Mater. 36, 331 (1970).
18. K. Farrell, A. Wolfenden and R. T. King, Rad. Eff. 8, 107 (1971).
19. S. D. Harkness, B. J. Kestel and S. G. McDonald, J. Nucl. Mater. 46, 159 (1973).
20. H. R. Brager and J. L. Straalsund, J. Nucl. Mater. 46, 103 (1973).
21. E. E. Bloom, An Investigation of Fast Neutron Radiation Damage in an Austenitic Stainless Steel, USAEC Report ORNL-4580, Oak Ridge National Laboratory (1970).
22. N. H. Packan, Voids in Neutron Irradiated Aluminum, USAEC Report ORNL-TM-3109, Oak Ridge National Laboratory (1971).
23. K. Ehrlich and N. H. Packan, J. Nucl. Mater. 46, 77 (1973).
24. H. R. Brager, J. Nucl. Mater. 57, 103 (1975).
25. J. O. Stiegler and E. E. Bloom, J. Nucl. Mater. 41, 341 (1971).
26. D. I. R. Norris, J. Nucl. Mater. 40, 66 (1971).
27. D. W. Keefer, A. G. Pard and D. Kramer, Swelling as a Function of Displacement Damage in Proton-Irradiated 316 Stainless Steel, Ref. 5, p. 511.
28. D. W. Keefer and A. G. Pard, J. Nucl. Mater. 45, 55 (1972/73).
29. D. W. Keefer and A. G. Pard, Void Formation in Cold-Worked Type 316 Stainless Steel Irradiated with 1-MeV protons, Paper presented at the 7th ASTM Symposium on Effects of Radiation on Structural Materials, June 1974, Gatlinburg, TN. To be published in the proceedings.
30. D. J. Mazey, J. A. Hudson and R. S. Nelson, J. Nucl. Mater. 41, 257 (1971).

31. S. G. McDonald and Anthony Taylor, Void Swelling Behavior of Types 304 and 316 Stainless Steel Irradiated with 4-MeV Ni^+ Ions, Ref. 6, p. 228.
32. W. G. Johnston, T. Lauritzen, J. H. Rosolowski and A. M. Turkalo, The Effect of Metallurgical Variables on Void Swelling, General Electric Co. Report No. 76 CRD 019 (1976).
33. W. G. Johnston, J. H. Rosolowski, A. M. Turkalo and T. Lauritzen, J. Nucl. Mater. 47, 155 (1973).
34. W. G. Johnston, J. H. Rosolowski, A. M. Turkalo and T. Lauritzen, J. Nucl. Mater. 48, 330 (1973).
35. W. G. Johnston, J. H. Rosolowski, A. M. Turkalo and T. Lauritzen, J. Nucl. Mater. 54, 24 (1974).
36. J. J. Laidler, Discussion by Attendees on Void Formation in Metals During Ion Bombardment, Ref. 5, p. 447.
37. K. Urban, M. Wilkens, and A. Seeger, Application of High-Voltage Electron Microscopy to the Study of Voids and other Point-Defect Agglomerates, Ref. 5, p. 550.
38. J. L. Straalsund, H. R. Brager and J. J. Homes, Effects of Cold Work on Void Formation in Austenitic Stainless Steel, Ref. 5, p. 142.
39. D. I. R. Norris, Phil. Mag. 23, 135 (1971).
40. R. S. Nelson, J. Nucl. Mater. 57, 77 (1975).
41. S. D. Harkness and Che-Yu Li, Theoretical Study of the Swelling of Fast-Neutron-Irradiated Materials, Ref. 5, p. 798.
42. D. I. R. Norris, Discussion by Attendees on Swelling of Fast-Neutron-Irradiated Materials, Ref. 5, p. 821.

43. W. G. Johnston, J. H. Rosolowski, A. M. Turkalo and T. Lauritzen,
J. Nucl. Mater. 46, 273 (1973).
44. O. S. Oen, Oak Ridge National Laboratory, private communication (1975).
45. O. S. Oen, J. Narayan and T. S. Noggle, Ion Radiation Damage,
Application of Ion Beams to Metals, S. T. Picraux, E. P. EerNisse
and F. L. Vook, eds. (Plenum Publishing Corp., NY).
46. K. B. Winterbon, Peter Sigmund and J. B. Sanders, Mat. Fys. Medd.
Dan. Vid. Selsk. 37(14), (1970).
47. J. Lindhard, V. Nielsen and M. Scharff, Mat. Fys. Medd. Dan. Vid.
Selsk 36(10), (1968).
48. W. White and R. M. Mueller, Phys. Rev. 187, 499 (1969).
49. K. B. Winterbon, Rad. Eff. 15, 73 (1972).
50. M. T. Robinson, The Dependence of Radiation Effects on the Primary
Recoil Energy, Ref. 5, p. 397.
51. R. Bullough and R. C. Perrin, Theory of Void Formation and Growth
in Irradiated Materials, Ref. 5, p. 769.
52. Peter Sigmund, Phys. Rev. 184, 383 (1969).
53. W. Brandt and R. Laubert, Nucl. Instrum. Meth. 47, 201 (1967).
54. M. Kaminsky, Atomic and Ionic Impact Phenomena on Metal Surfaces
(Academic Press, Springer, 1965).
55. G. Carter and J. S. Colligon, Ion Bombardment of Solids (American
Elsevier, 1968).
56. M. W. Thompson, Defects and Radiation Damage in Metals
(Cambridge, 1969).
57. R. J. MacDonald, Adv. Phys. 21, 457 (1970).
58. F. Fairbrother, Jr., et al., Vacuum 4, 112 (1954).

59. D. W. Keefer and A. G. Pard, J. Nucl. Mater. 47, 97 (1973).
60. D. G. Doran, J. R. Beeler, N. D. Dudey and M. J. Fluss, Hanford Engineering Dev. Lab., HEDL-TME-73-76 (1973).
61. R. Bullough, Recent Advances in the Rate Theory of Void Swelling-Cascade Effects, Ref. 7, p. 275.
62. J. R. Willis and R. Bullough, Ref. 4, p. 133.
63. A. Taylor and S. G. McDonald, Void Formation in Proton- and Nickel-Irradiated 304 Stainless Steel, Ref. 5, p. 499.

Table 1. Composition of type 316 stainless steel (%).

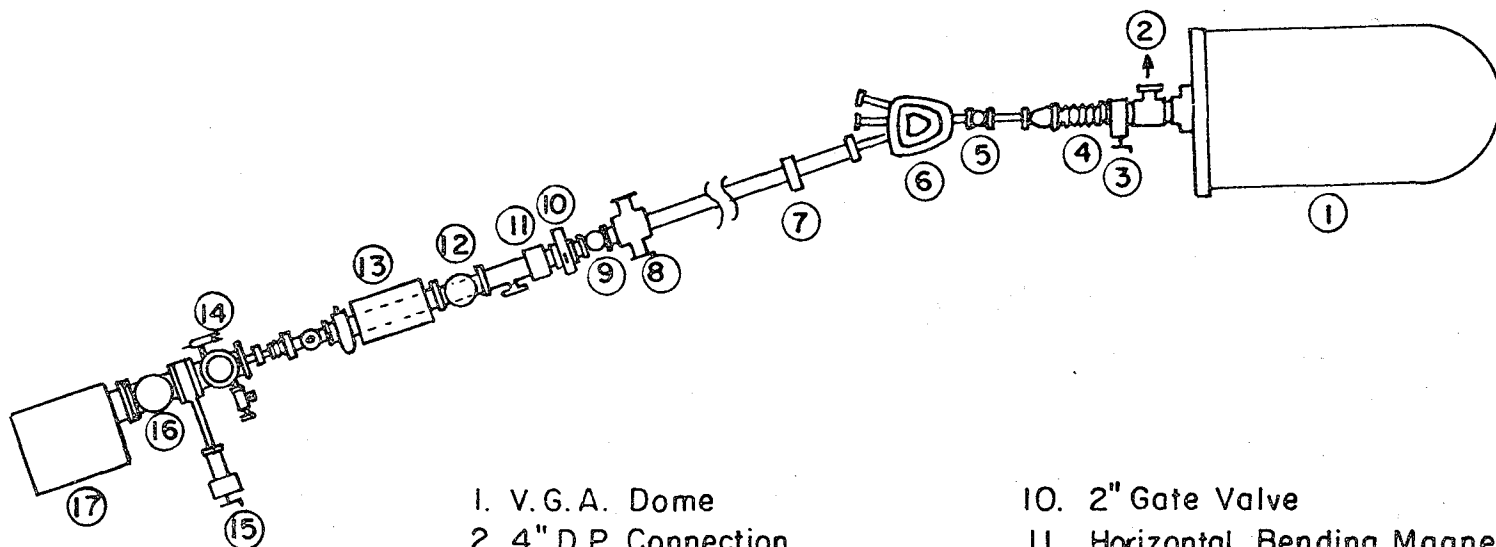
C	Mn	Si	P	S	Mo	Cr	Ni	Fe
0.05	1.73	0.52	0.004	0.008	2.52	17.59	13.64	63.93

Table II. Displacements per atom vs depth.

Zone No.	Penetration Depth		Sample 1 dpa	Sample 2 dpa	Sample 3 dpa	Sample 4 dpa	Sample 5 dpa
	Before Irradiation (Å)	mg/cm ²					
1	0-1000	0-0.08	12	24	43	76	105
2	1000-2000	0.08-0.16	14	27	49	88	120
3	2000-3000	0.16-0.24	15	31	56	100	136
4	3000-4000	0.24-0.32	20	40	72	129	177
5	4000-5000	0.32-0.40	31	62	112	200	275
6	5000-6000	0.40-0.48	51	102	184	328	450
7	6000-7000	0.48-0.56	67	134	241	430	590
8	7000-8000	0.56-0.64	49	98	177	314	430
9	8000-9000	0.64-0.72	17	34	62	110	151
10	9000-10,000	0.72-0.80	2	4	8	14	19
Total			278	556	1004	1789	2453

FIGURE CAPTIONS

- Fig. 1. Proton irradiation set-up.
- Fig. 2. Proton irradiation set-up (continued).
- Fig. 3. Sample holder assembly.
- Fig. 4. Proton irradiated surface.
- Fig. 5. Profilometer trace across the masked area.
- Fig. 6. Energy loss of 140 keV proton in nuclear collisions vs depth.
- Fig. 7. Measured step-height vs proton dose.
- Fig. 8. Swelling-dpa curve.
- Fig. 9. Effect of void density on the rate of swelling (calculated).
(Dislocation density = $5 \times 10^{10}/\text{cm}^2$.)
- Fig. 10. Effect of dislocation density on the rate of swelling (calculated).
(Void density, $n_{\text{vo}} = 5 \times 10^{13}/\text{cm}^3$, swelling = 40%.)
- Fig. 11. Calculated void and dislocation densities to obtain the swelling-dpa curve.
- Fig. 12. Comparison of swellings due to proton and nickel ion irradiations.

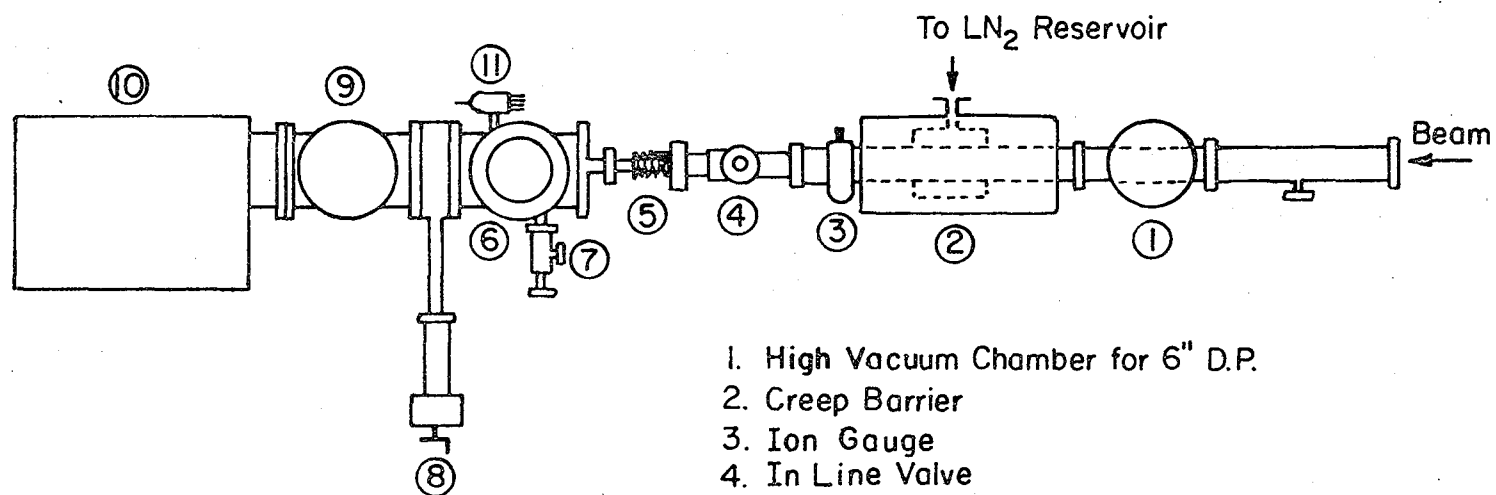


- | | |
|----------------------------|--|
| 1. V.G.A. Dome | 10. 2" Gate Valve |
| 2. 4" D.P. Connection | 11. Horizontal Bending Magnet |
| 3. 2" Gate Valve | 12. High Vacuum Chamber
for 6" D.P. |
| 4. Bellows | 13. Creep Barrier |
| 5. View Window | 14. Sample Chamber |
| 6. Main Bending Magnet | 15. 6" Gate Valve |
| 7. Vertical Bending Magnet | 16. Ti Sublimation Pump Chamber |
| 8. Horizontal Slits | 17. Ion Pump |
| 9. View Window | |

Fig. 1.

XBL 7511-9435

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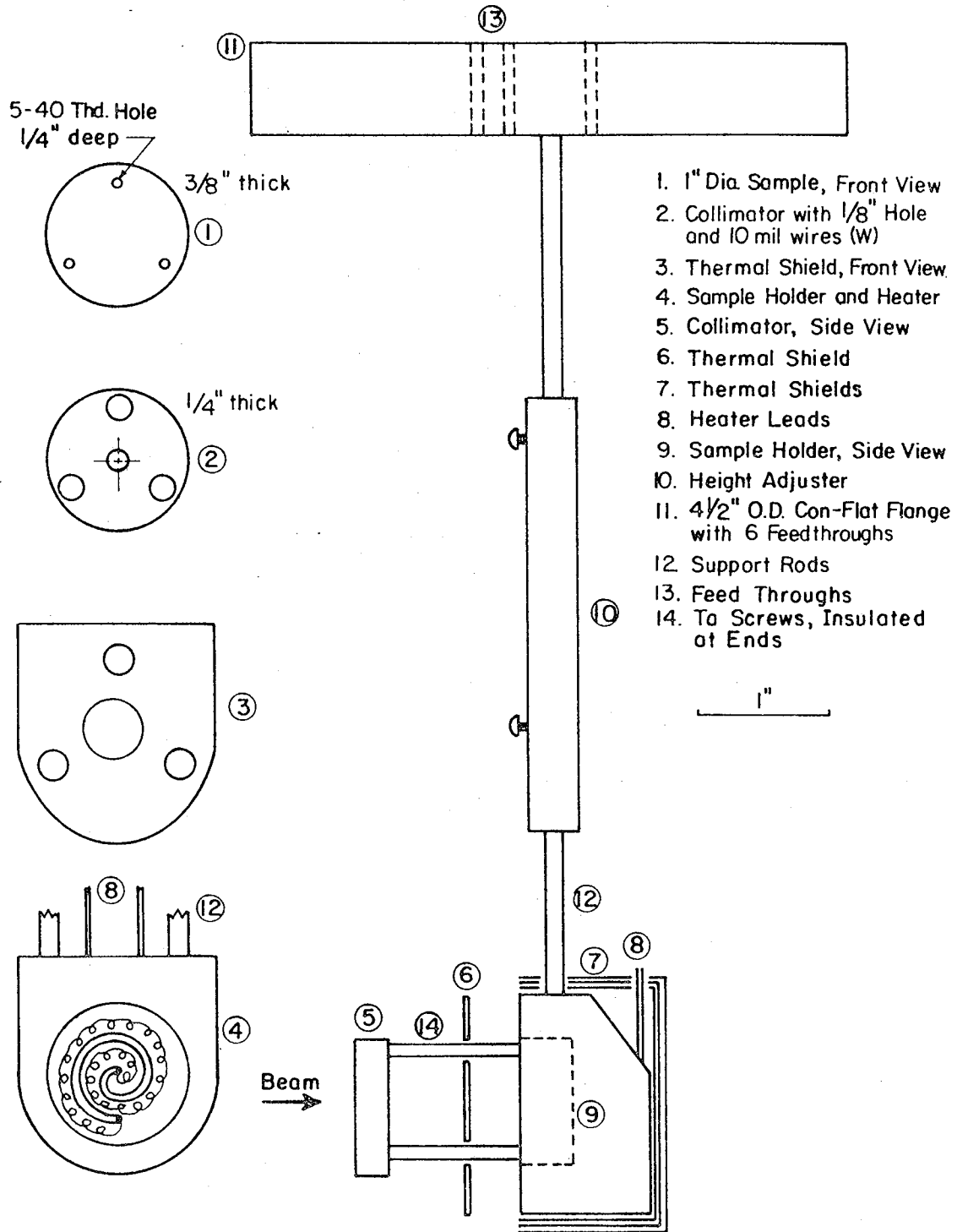


1. High Vacuum Chamber for 6" D.P.
2. Creep Barrier
3. Ion Gauge
4. In Line Valve
5. Bellows
6. Sample Chamber
7. 1" Gold Seal Valve
8. 6" Gate Valve
9. 6" Titanium Sublimation Pump
10. 270 ℓ /sec. Ion Pump
11. Sample Ion Gauge

XBL 7511-9436

Fig. 2.

-43-



XBL 7511-9437A

Fig. 3.

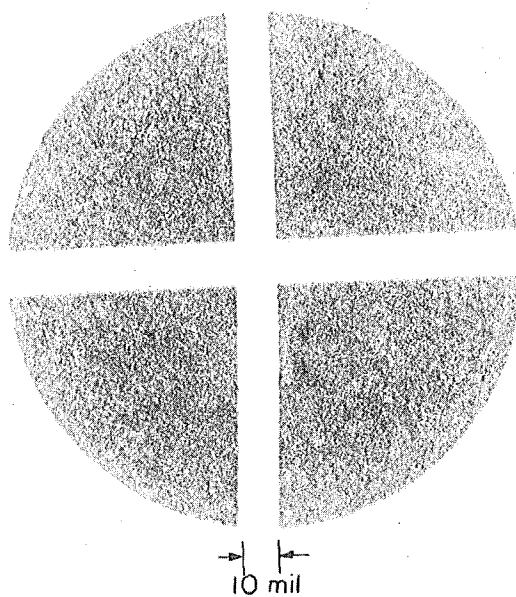
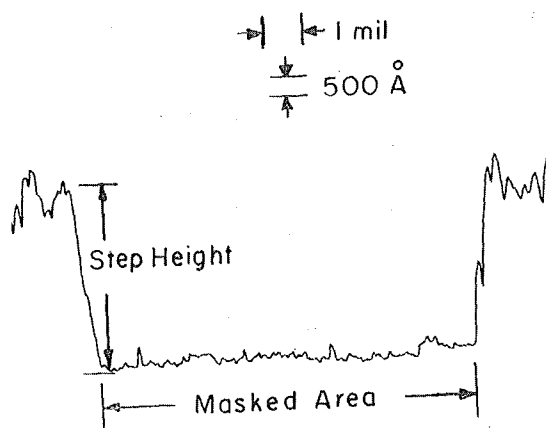
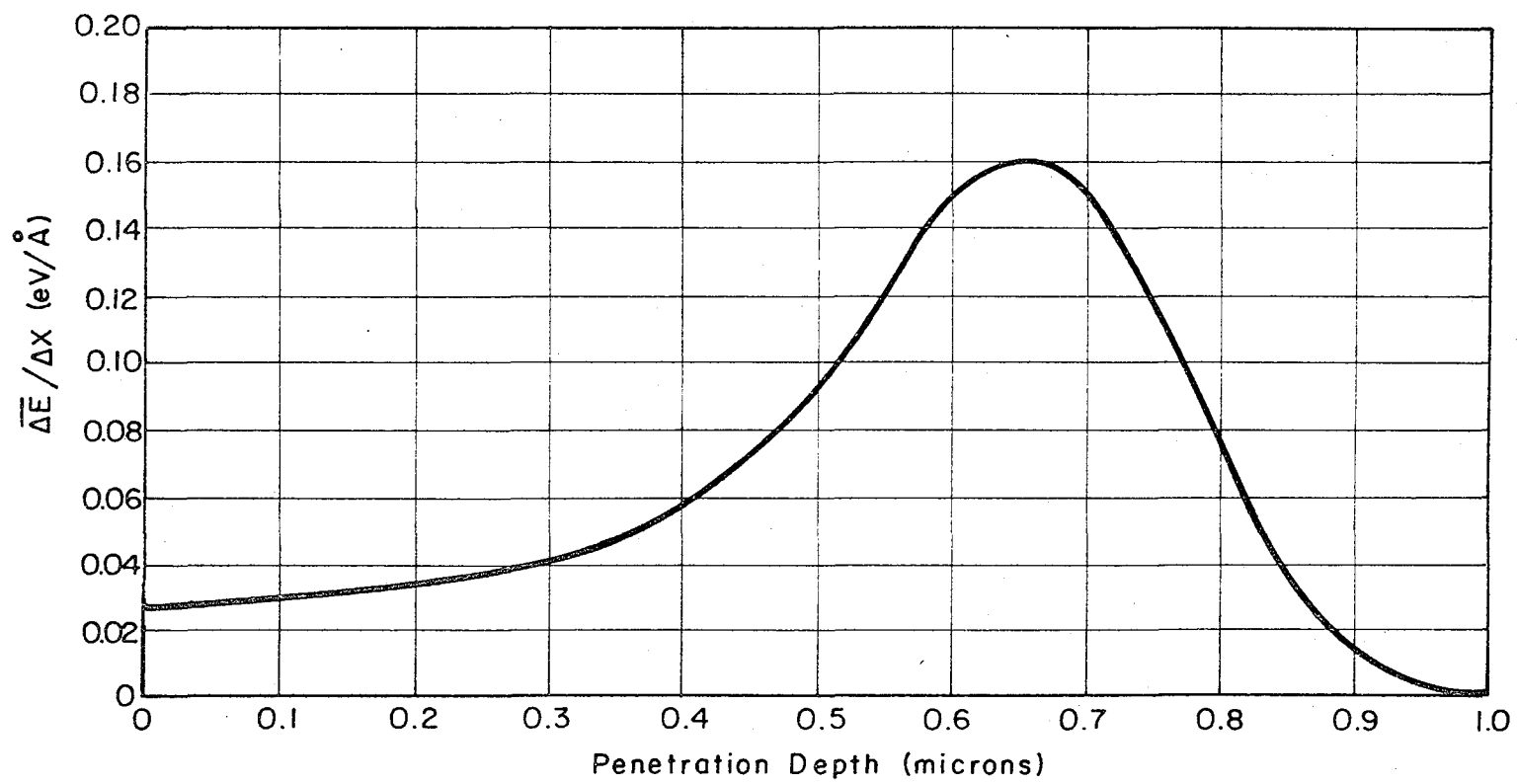


Fig. 4



XBB 765-4046

Fig. 5



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Fig. 6.

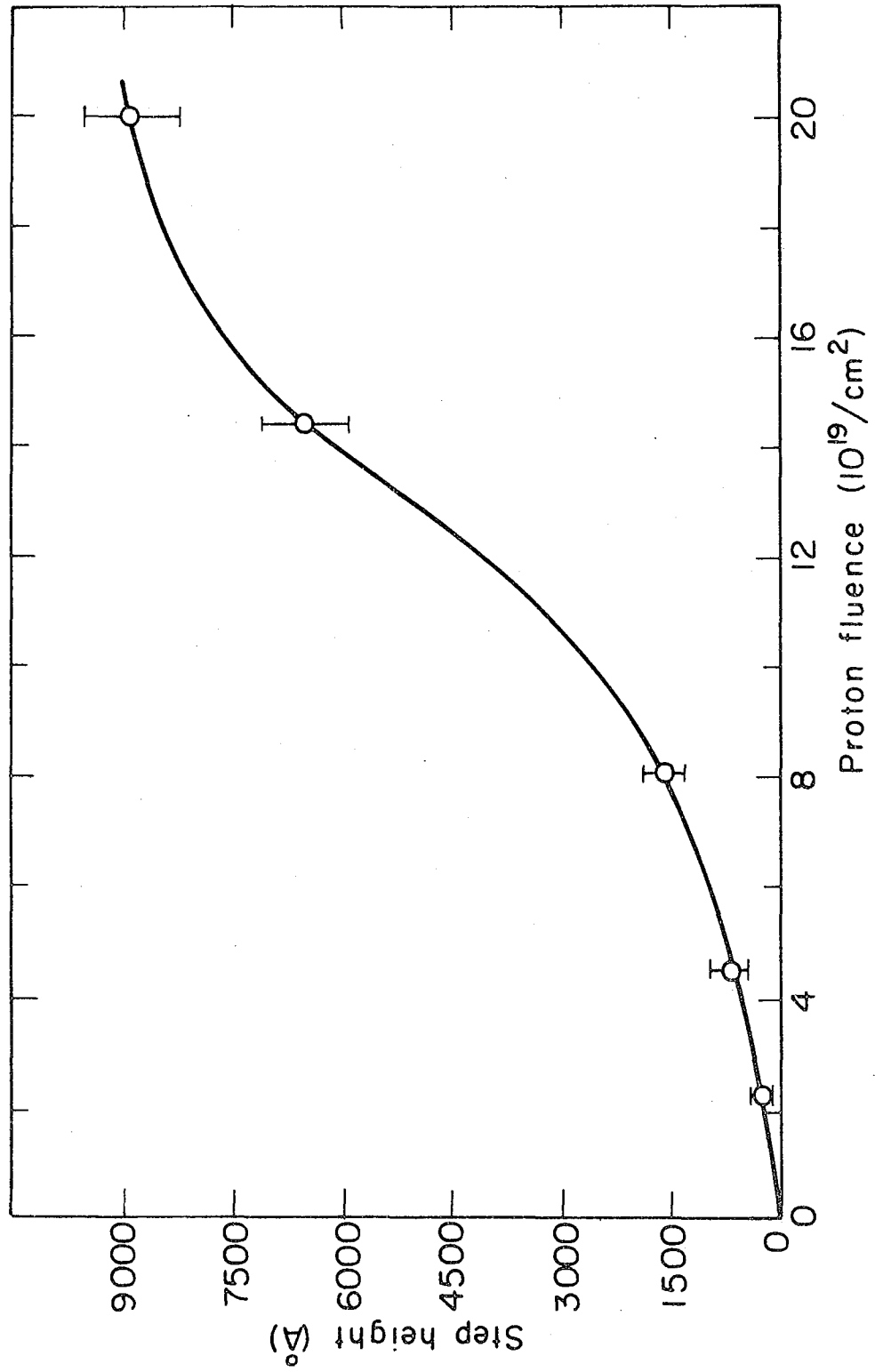


Fig. 7.

XBL762-6452

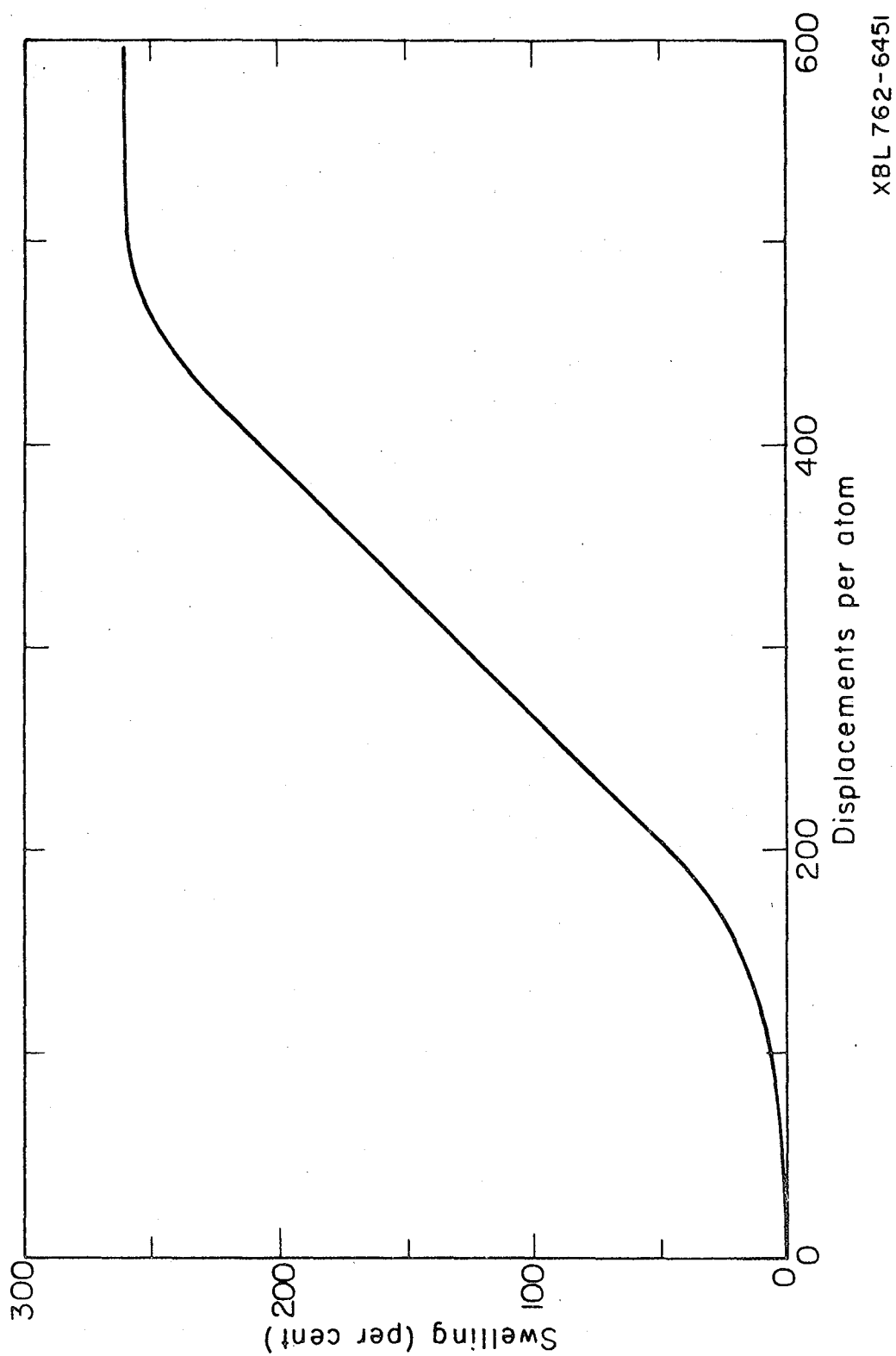


Fig. 8.

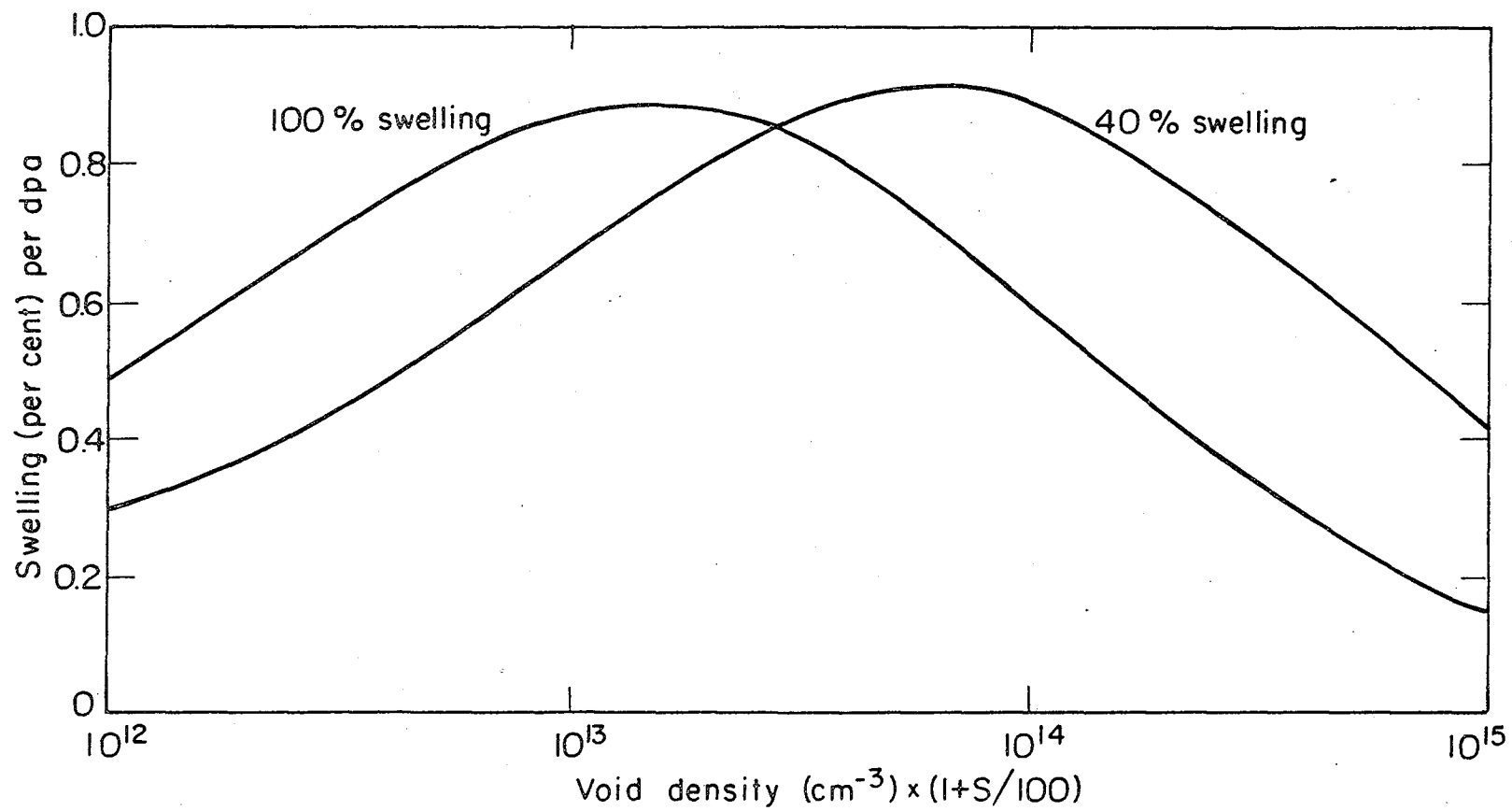
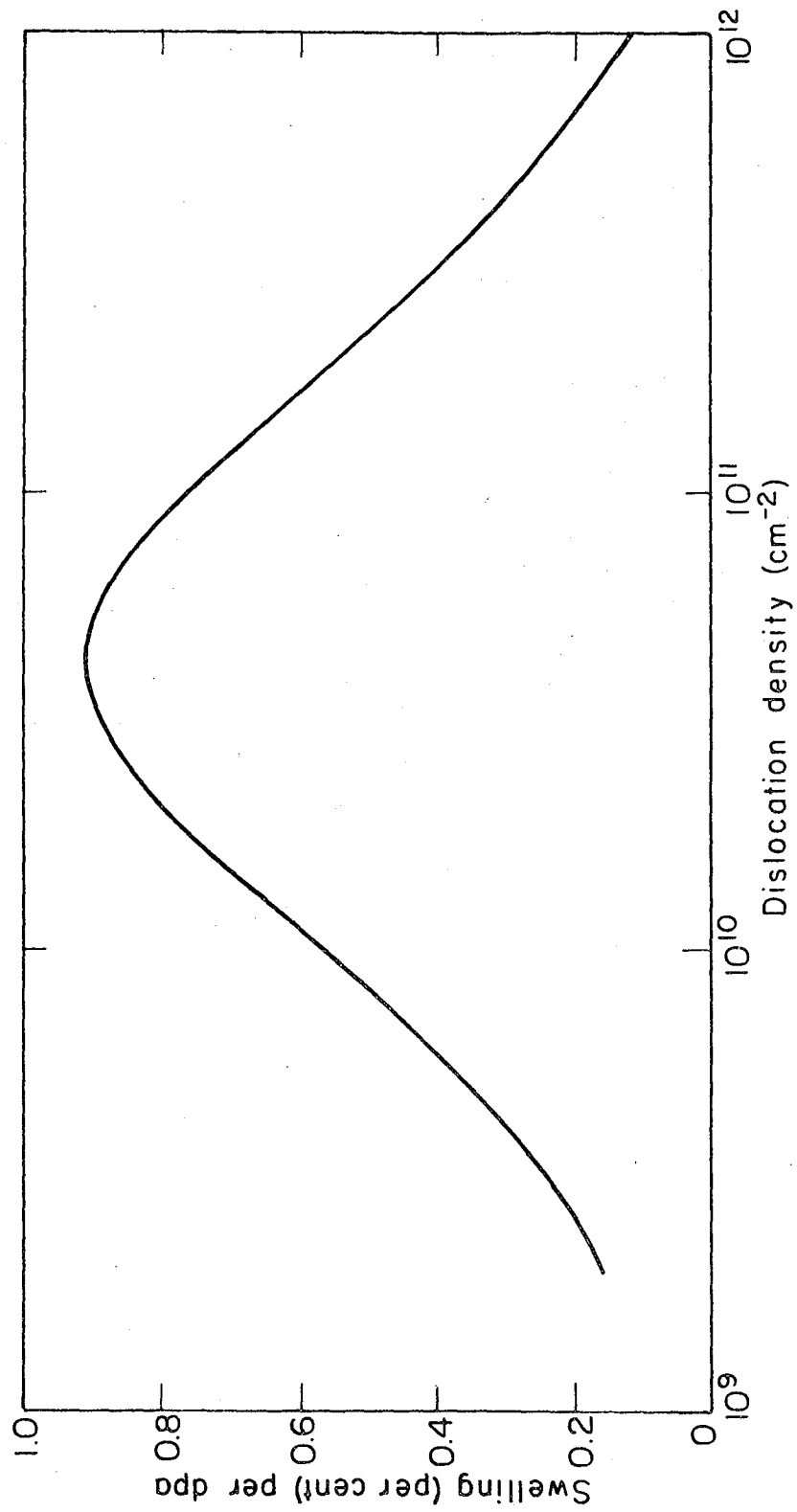


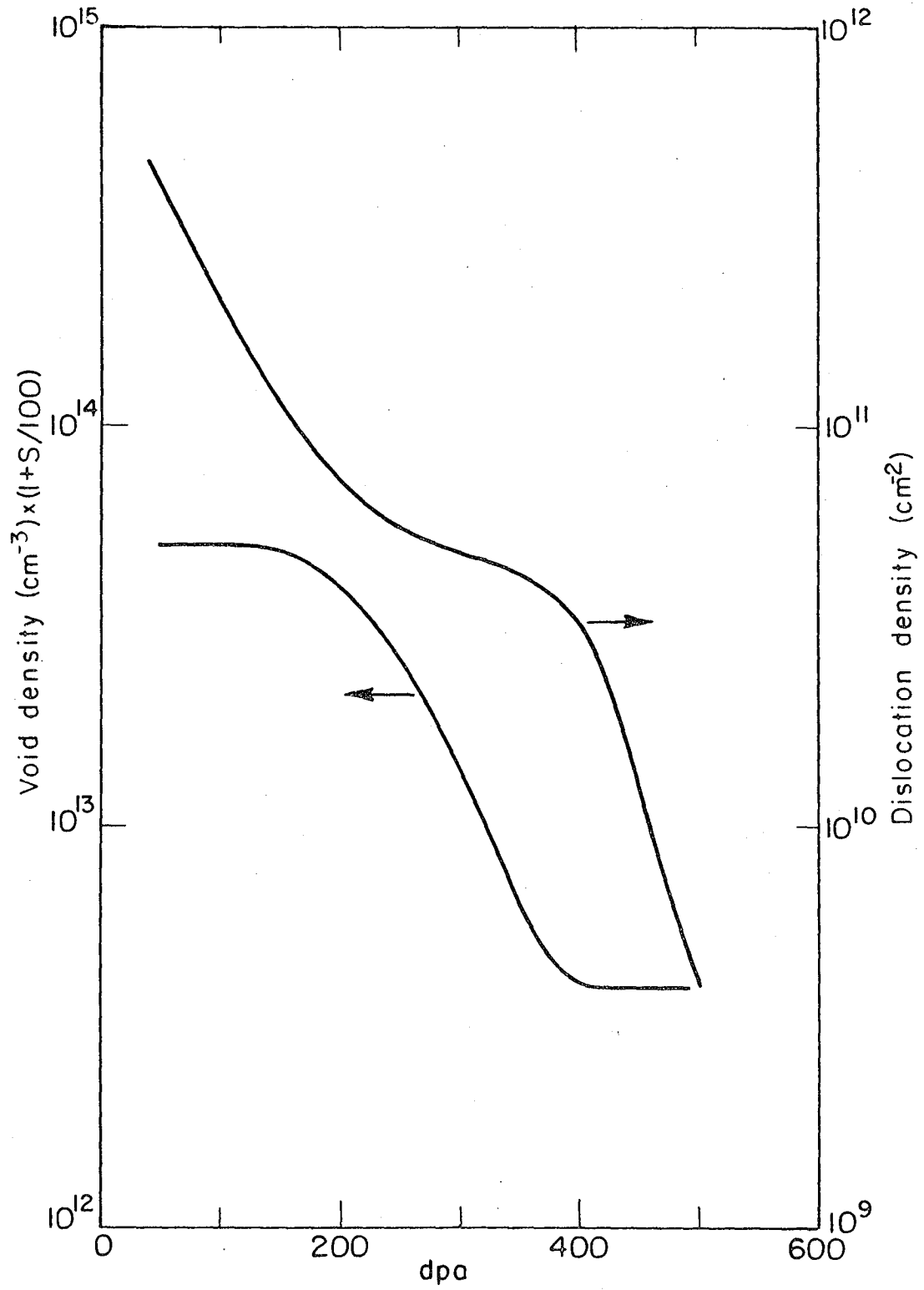
Fig. 9.

XBL 762 -6454



XBL 762-6453

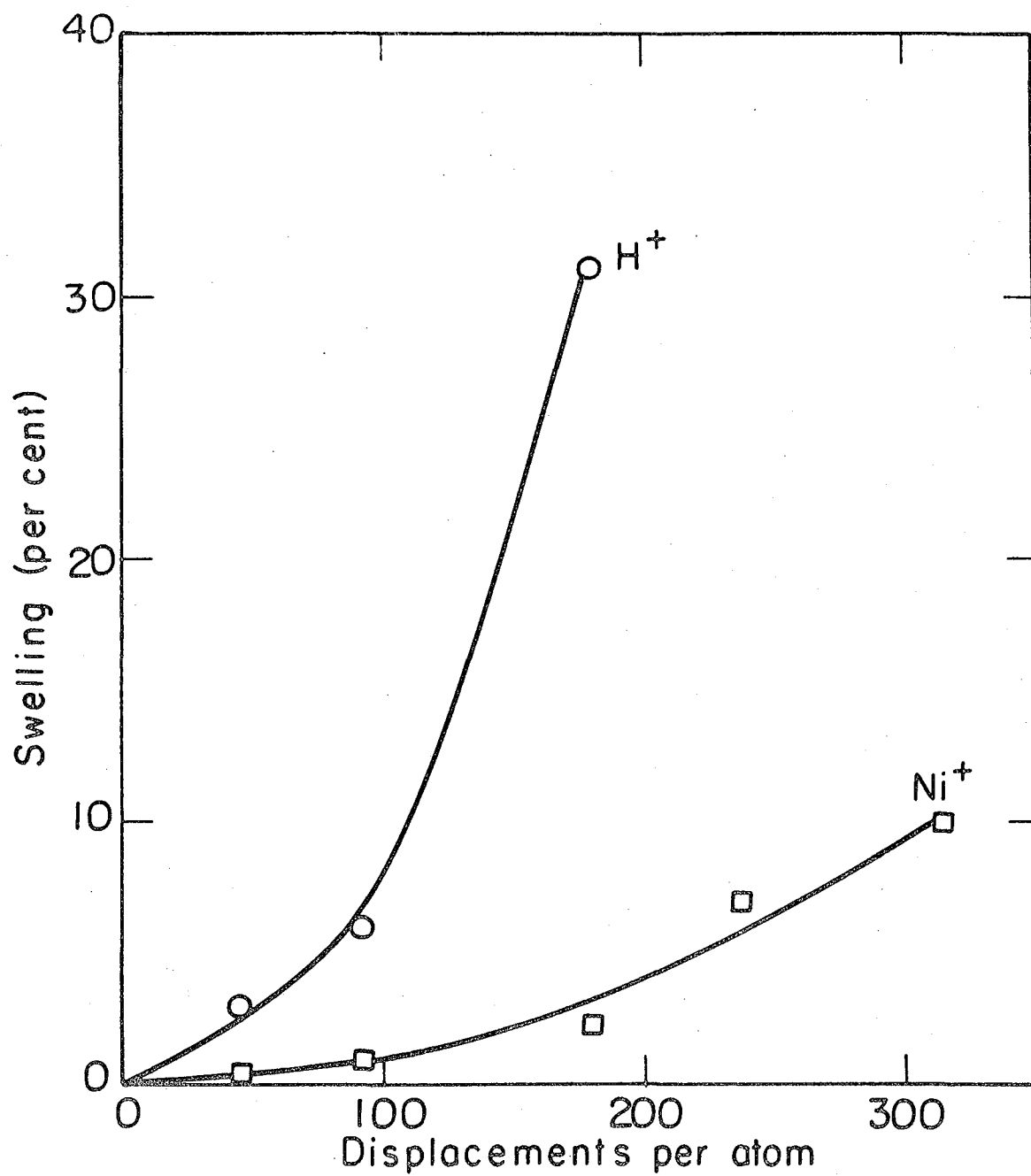
Fig. 10.



XBL 764-6772

Fig. 11.

-51-



XBL 764-6771

Fig. 12.

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