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SWELLING AS A FUNCTION OF DISPLACEMENT DAMAGE IN PROTON-IRRADIATED TYPE 316 STAINLESS STEEL

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SWELLING AS A FUNCTION OF DISPLACEMENT DAMAGE IN PROTON-IRRADIATED TYPE 316 STAINLESS STEEL

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ABSTRACT

Specimens of Type 316 stainless steel containing between 2 and 9 appm of preinjected helium, were proton irradiated at 500 and 600°C. Electron microscopy was used to determine volume swelling in specimens having undergone between 0.5 and 50 displacements per atom. Displacements were calculated from the Kinchin-Pease model with a threshold energy of 25 eV. Swelling at 500°C increased with damage up to about 25% for 50 displacements per atom. No evidence of a limit to swelling with increasing fluence was observed. The minimum void size increased with damage, and the void density increased with atom displacement rate. These results imply completion of nucleation early in the irradiation, followed by continued void growth. A specimen having undergone 100 displacements per atom at 500°C exhibited a void array which was so complex that a swelling determination could not be made. Ferrite, transformed from austenite during the irradiation, was detected in this specimen by electron diffraction as small crystallites within the austenite grains. Evidence of austenite-to-ferrite transformation was obtained for specimens with about 5 or more displacements per atom. A specimen irradiated to about 20 displacements per atom at 600°C exhibited frequent joining of voids to form a complex array. Preliminary examination indicates swelling at 600°C is greater than at 500°C. Voids are larger and less numerous at 600°C.

I. INTRODUCTION

It is important to determine the extent of radiation damage to core components of fast reactors at fluences of 10^{23} n/cm^2 (E > 0.1 MeV) and higher. However, several years of irradiation in test reactors and considerable expense are required to achieve these exposures.

Accelerator irradiations provide a rapid and inexpensive method of studying radiation effects. Although they are not a substitute for reactor irradiations, they can produce information that will allow an estimate to be made of high-fluence effects. The present report deals with the swelling of Type 316 stainless steel produced by irradiation with protons.

II. EXPERIMENTAL PROCEDURE

Specimens were irradiated at 500 and 600°C with protons having energies of either 0.75 or 1.0 MeV. The proton fluences were varied between 6×10^{17} and 1.3×10^{20} p/cm^2 at a flux of about 1×10^{15} p/cm^2-sec. Specimen regions having undergone between 0.5 and 100 displacements per atom were examined by electron microscopy.

The specimens used were 0.012-mm-thick foils having cross-sectional dimensions of 1.78 x 0.88 cm. Each specimen was solution-annealed at 980°C and then precipitation-annealed at 760°C. Formation of $M_{23}C_6$ carbides at austenite grain boundaries occurs during the latter anneal.(1) The average grain size was 20 to 30 μm. After heat treatment, each specimen was diffusion-bonded at 650°C to a copper support plate. During irradiation, this plate served as a thermal conductor between the specimen and a temperature control device.(2) Temperature control depended on proton beam stability and was generally within ±5°C with periodic fluctuations of ±10°C.

Before irradiation, specimens were injected with helium by means of a cyclotron.(3) These injections resulted in an even distribution of helium throughout the sample volume as has been verified by mass spectrometric analysis of front and back sections of injected foils. The temperature of samples undergoing injection does not rise significantly above ambience. The helium content of samples discussed here varied from 2 to 9 appm.

After irradiation, each specimen was removed from its copper plate by immersion in nitric acid and cut into several sections. Electron microscope foils were prepared from specific regions within the specimen thickness by electropolishing of a section. The thinning procedure was periodically interrupted so that x-ray absorption measurements could be made to determine the section thickness and hence the expected final foil position. Foil positions determined in this way are accurate within ±1000 Å.

The void number density and the distribution of void diameters in electron photomicrographs were determined by use of a particle-size analyzer. The void densities were calculated from estimated foil thickness. Contributions to the voidage for each size interval on the analyzer were determined and summed to give the total void volume.

III. DISPLACEMENT CALCULATIONS

The atom displacement calculations used in this work are based on the damage-production model of Kinchin and Pease.[4] Before proceeding with a description of the calculations, we must consider the assumptions made in the Kinchin-Pease formulation and test their applicability to low-energy proton irradiation.

In the formulation of their model, Kinchin and Pease assumed that the scattering events involving displaced atoms (knock-ons) and atoms of the lattice were hard sphere in character. That is, the atom-atom interactions were assumed to be screened, long-range, and of low energy. Since the number of secondary and higher-order displacements depends on the type of atom-atom interaction operating, it is necessary to know the nature of the interaction. A simple calculation in which the closest distance of approach is equated to the screening radius of the old Bohr theory, gives an approximate "border-line" energy for the transition from hard sphere to Rutherford scattering. For iron, this knock-on energy is approximately 5×10^4 eV. To see where we stand in our experiments, we must calculate the maximum energy, T_m, which can be transferred to an iron atom by an incoming proton of energy, E. From the usual expression for T_m, we find that for this case $T_m = 6.95 \times 10^{-2}$ E (eV). In our experiments, the incident proton energy has been 1.0 MeV or less, in which case $T_m \lesssim 7 \times 10^4$ eV. Since the <u>average</u> transfer energy is considerably less than this, and since our microscope foils are taken at proton penetrations corresponding to proton energies of about 0.6, 0.45, and 0.12 MeV, it is clear that the vast majority of primary knock-on atoms in our experiments possess energies well below 5×10^4 eV. We are, therefore, justified in accepting the Kinchin-Pease assumption of hard sphere scattering. Inclusion of a "hybrid" scattering law to take account of "border-line" energy transfer events would have almost no effect on our calculated number of displacements.

A second important assumption made in the Kinchin-Pease model is the following: if the energy of a knock-on is sufficiently low so that its velocity is less than that of an orbital electron, <u>all</u> energy loss by that knock-on is by elastic nuclear processes. Since, in fact, inelastic losses to electrons do continue to occur at low energies, the importance of the inelastic loss process must be determined. Obviously, the Kinchin-Pease assumption might tend to overestimate the number of additional displacements created by knock-ons. Lindhard and co-workers[5] have made detailed calculations of the stopping cross sections for both electronic and nuclear interactions. We have included their results in calculations made in a later paragraph. The correction term for electronic interactions is found to be small for our experiments.

In the Kinchin-Pease model, it is also assumed that energy is dissipated via random collisions. Therefore, the possibility of channeling and focusing processes is not taken into account. Because of the relatively low energies of primary knock-ons created in our experiments, channeling events can safely be ignored. Focusing probably does occur and, since such a process would lead to somewhat fewer than the expected number of displacements, we might artificially account for it by raising slightly the threshold energy, E_d, for displacement production. However, we do not have an accurate E_d value for the alloys we irradiate, and rather arbitrary adjustments from our estimated value of 25 eV seem fruitless. In any case, the effect of focusing is expected to be small.

Finally, the Kinchin-Pease model allows for no mutual recombination of interstitials and vacancies. Clearly, because of the mobility of both interstitials and vacancies at the temperatures at which we irradiate, recombination may be of considerable importance. We think, however, that solution to this problem must await the application of computer techniques. In the meantime, we can only ignore recombination and make calculations which are appropriate only for 0°K. (Spontaneous recombination of close pairs probably occurs even at this temperature, but its extent is not known.)

We can now proceed with a calculation of the number of displacements created by proton irradiation. The total number of displacements per atom, $N_d(E)$, produced by a fluence of φt particles of energy E is given by

$$N_d(E) = \varphi t \int_{E_d}^{T_m} d\sigma(T) \cdot n(T) \quad . \qquad \ldots (1)$$

In Equation 1, $d\sigma(T)$ is the differential cross section for transferring energy in the interval T to T + dT. For protons in the energy range of interest to us, the Rutherford scattering law holds, and we can write for protons incident on iron

$$d\sigma(T) = 4\pi a_o^2 Z_p^2 Z_{Fe}^2 \frac{M_p}{M_{Fe}} \frac{E_R^2}{E} \frac{dT}{T^2} \quad . \qquad \ldots (2)$$

In Equation 2, a_o is the Bohr radius of the hydrogen atom, and Z_p, M_p and Z_{Fe}, M_{Fe} are the charge numbers and masses of protons and iron atoms, respectively. E_R is the Rydberg energy, 13.6 eV. With the appropriate numerical values, Equation 2 becomes

$$d\sigma(T) = 7.88 \times 10^{-3} \frac{1}{E} \frac{dT}{T^2} \quad . \qquad \ldots (3)$$

In Equation 1, n(T) is the total number of displacements per primary recoil atom of energy T. In the interval $E_d \leqq T \leqq 2E_d$, the primary is incapable of displacing a second atom, so n(T) = 1. In the interval $2E_d \leqq T \leqq T_m$, we let

$$n(T) = f(T) \frac{T}{2E_d} \quad , \qquad \ldots (4)$$

where f(T) is a factor introduced to include the loss of recoil atom energy in inelastic collisions. From Lindhard et al.,[5] f(T) is the fraction of primary energy which is dissipated in elastic collisions.

$$f(T) = \frac{S_n}{S_n + S_e} \quad , \qquad \ldots (5)$$

where S_n is the cross section for elastic nuclear stopping and S_e is the cross section for inelastic electronic stopping. S_n is independent of particle energy and has a value of 3.55×10^{-25} cm^2/atom. $S_e = CT^{1/2}$, where $C = 4.15 \times 10^{-28}$ $cm^2 - eV^{-1/2}$.

From Equations 1, 3, 4, and 5,

$$N_d(E) = 7.88 \times 10^{-13} \frac{\varphi t}{E} \left[\int_{E_d}^{2E_d} \frac{dT}{T^2} + \frac{S_n}{2E_d} \int_{2E_d}^{T_m} \frac{dT}{T(S_n + CT^{1/2})} \right] \quad .$$

Integrating and putting in the limits,

$$N_d(E) = 3.94 \times 10^{-13} \frac{\varphi t}{E \cdot E_d} \left[1 + \ln \frac{T_m}{2E_d} - 2\ln \left\{ \frac{S_n + CT_m^{1/2}}{S_n + C(2E_d)^{1/2}} \right\} \right] , \qquad \ldots (6)$$

where E_d is assumed to be 25 eV.

Equation 6 is the expression we are now using in calculating the displacements in proton irradiated samples. The third term in the brackets in Equation 6 is the correction term for electronic stopping; it amounts to a correction of less than 10% at even our highest proton energy, so we are left with essentially the old Kinchin-Pease result.

The energy of a proton as a function of its initial energy and depth of penetration into stainless steel can be obtained from the tabulations of Janni.[6] The energy at selected penetration distances is then used in Equation 6 to obtain the number of displacements per unit proton fluence.

IV. DISCUSSION OF EXPERIMENTAL RESULTS

The results for all of the samples are given in Table 1, where they are arranged in order of increasing damage for each temperature. Since the damage produced by the protons increases with penetration into the sample, several electron microscope foils representing different degrees of damage can be extracted from the same irradiated sample.

The increase in sample volume per unit volume (swelling) as a function of calculated number of displacements is shown in Figure 1. The points represent averages of swelling data obtained from several photomicrographs of the same foil. The spread in the data used in forming these averages is sometimes large and is considered to be representative of the true swelling variation in a foil. Swelling deviations are thought to be the result of microstructural inhomogeneities within small regions of a sample. Uncertainties in foil position within the sample cannot account for such results. Further, it is unlikely that significant inhomogeneities in helium content or proton beam density could occur over the small distances involved here.

The line through the 500°C data points in Figure 1 was drawn according to a least squares fit. The equation for this line is

$$\frac{\Delta V}{V}(\%) = 0.17\, N_d^{1.2} \quad , \qquad \ldots(7)$$

where ΔV is the total void volume in a sample of volume V, and N_d is the calculated number of displacements per atom. In the region between about 1 and 50 displacements per atom, there appears to be no change in the dependence of swelling on damage.

The data represented by the point at about 0.5 displacements per atom in Figure 1 were not used in the least squares fitting. At this low damage, void formation is at an early stage, and the resolvable voids are very inhomogeneously distributed. Therefore, accuracy in determining swelling is extraordinarily low, and the data were ignored in the fitting.

A photomicrograph of the void array in a specimen having undergone 50 displacements per atom at 500°C is shown in Figure 2. From this micrograph, the

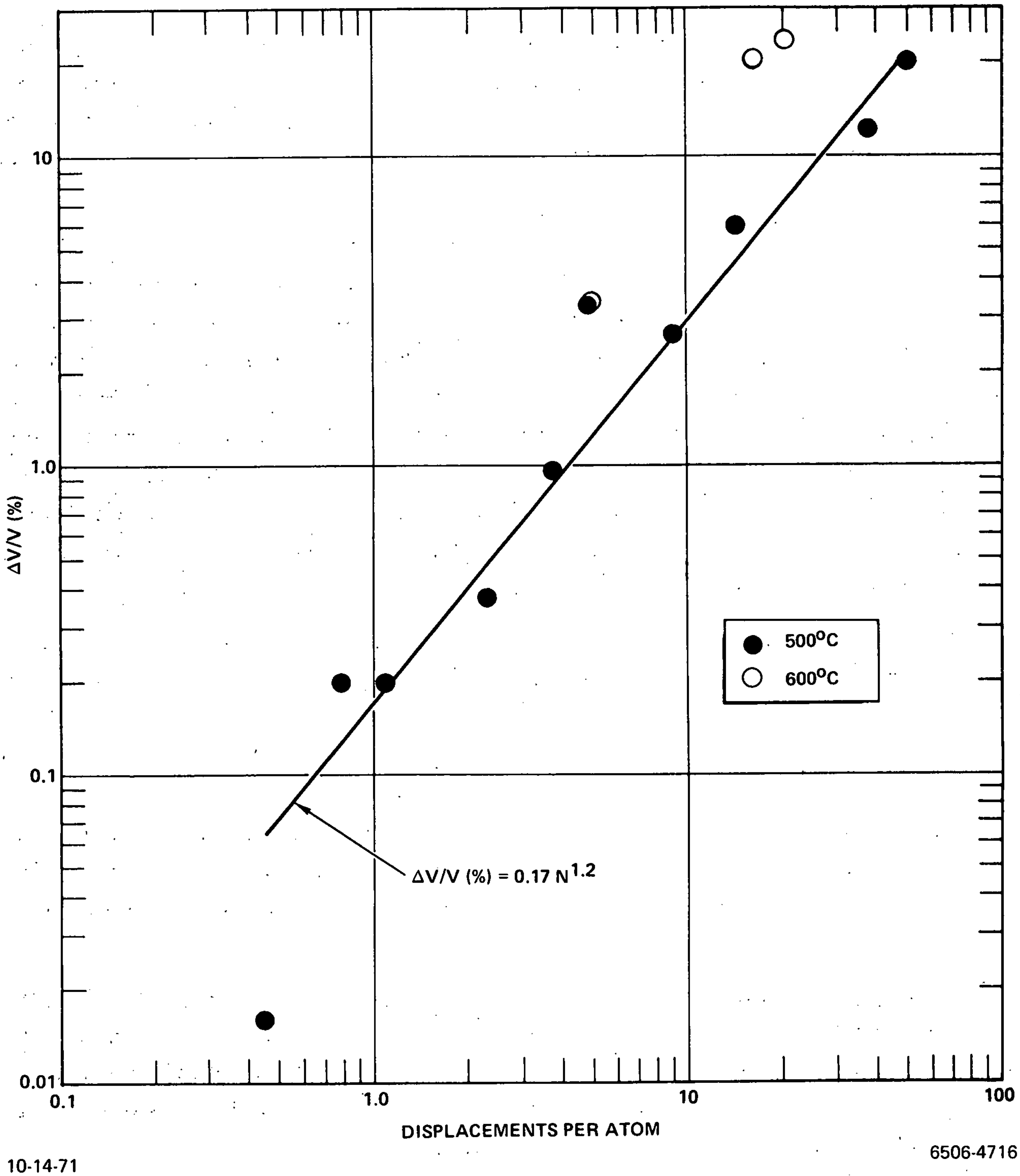


Figure 1. Swelling at 500 and 600°C as a Function of Displacement Damage for Proton-Irradiated Type 316 Stainless Steel Containing Helium

TABLE 1

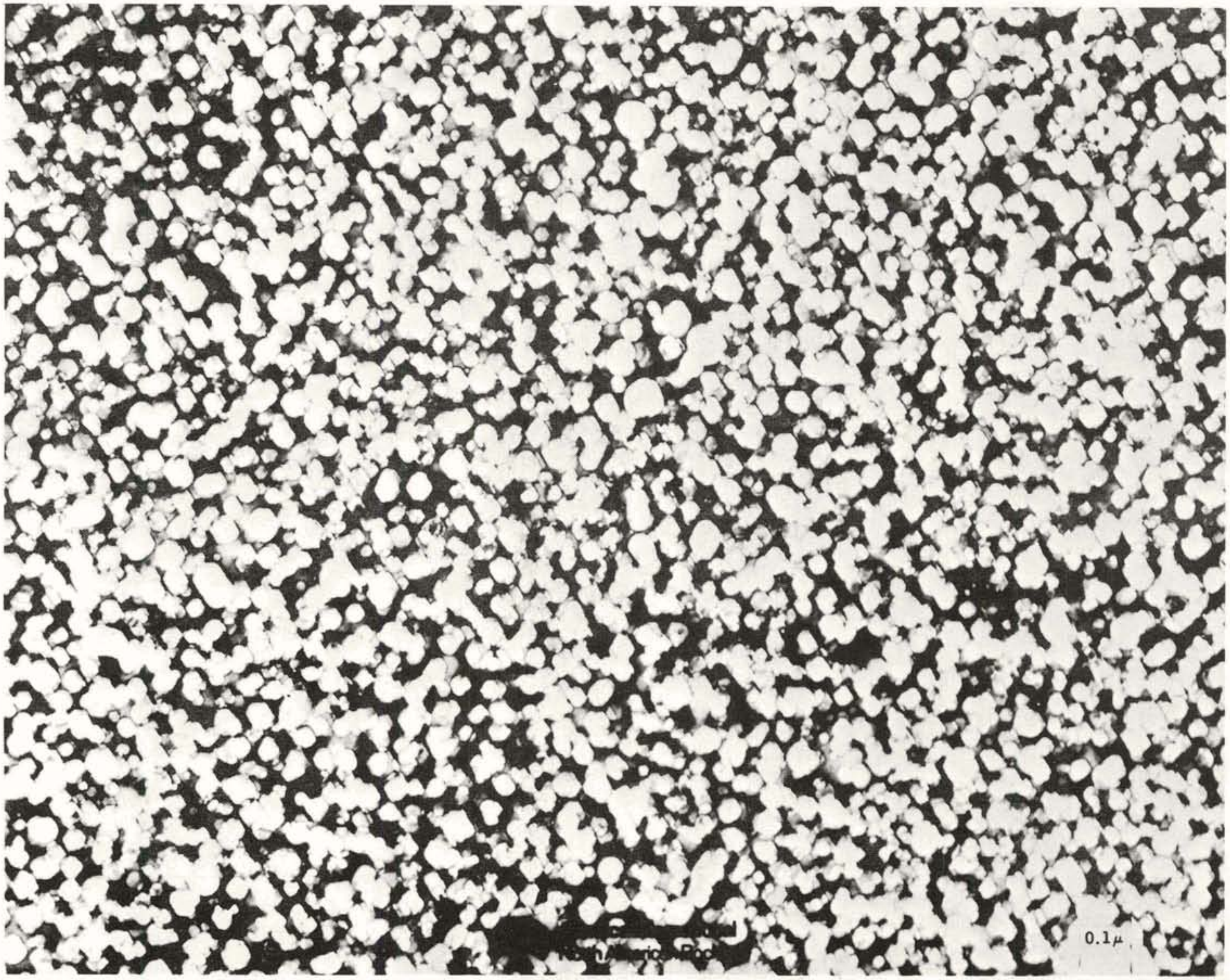
VOID FORMATION IN PROTON-IRRADIATED TYPE 316 STAINLESS STEEL

Sample Number	Temperature (°C)	Helium (appm)	Damage (Displacements/atom)	Damage Rate (Displacements/atom-sec)	Void Density ($/cm^3$)	Average Void Diameter (Å)	Volume Increase (%)
28	500	9	0.46	7.0×10^{-4}	1.8×10^{15}	52	0.018
6*	500	2	0.76	1.0×10^{-4}	3.0×10^{15}	110	0.20
6*	500	2	1.1	1.5×10^{-4}	9.6×10^{15}	70	0.19
36	500	4	2.3	1.7×10^{-4}	0.85×10^{15}	190	0.38
6*	500	2	3.8	5.2×10^{-4}	4.0×10^{15}	150	0.98
56A	500	6	4.8	3.9×10^{-4}	4.3×10^{15}	230	3.4
36	500	4	9.2	7.0×10^{-4}	3.0×10^{15}	240	2.7
56A	500	6	15.0	12.0×10^{-4}	7.2×10^{15}	260	6.0
59	500	6	25.0	3.6×10^{-4}	1.1×10^{15}	†	†
56B	500	6	38.0	8.8×10^{-4}	4.0×10^{15}	360	13.0
56B	500	6	51.0	12.0×10^{-4}	6.1×10^{15}	370	21.0
59	500	6	100.0	12.0×10^{-4}	§	§	§
38	600	6	16.0	6.8×10^{-4}	1.6×10^{15}	590	20.0
38	600	6	21.0	8.6×10^{-4}	2.0×10^{15}	590	24.0
80	600	5	5.0	10.0×10^{-4}	0.77×10^{15}	375	3.5

*Data from Sample 6 not included in Figure 6 because of large uncertainties in proton flux.

†Results considered invalid due to sample distortion.

§Gross swelling could not be measured on photomicrographs.



6-164

Figure 2. Voids in Type 316 Stainless Steel Proton-Irradiated at 500°C to Produce 50 Displacements per Atom (Sample 56B)

minimum, average, and maximum void diameters are 95, 405, and 880 Å, respectively, the void density is 5×10^{15} per cm^3, and the swelling is 24%. The morphology of these voids is typical of that obtained in other specimens irradiated to lower fluences at 500°C. As we have discussed previously,[2] the voids are bounded by {111} planes and frequently show truncation by {100} planes. Note that, although considerable void overlap occurs in Figure 2, there is little evidence of voids having merged to form irregularly shaped agglomerates.

We have also irradiated a specimen at 500°C to 100 displacements per atom. As can be seen in Figure 3, the void array has become more complex with increasing damage. Because of this complexity, we are unable to determine an accurate swelling value for this sample. It is interesting that during this irradiation a portion of the sample was transformed from austenite to ferrite. Electron diffraction studies have shown that the ferrite occurs as small crystallites within the austenite grains. Because of the variation in displacement damage through the irradiated portion of the sample, it is difficult to determine the amount of transformation which is typical of the region which underwent 100 displacements per atom. In this region we estimate, from x-ray diffraction measurements, that more than 10% of the austenite was transformed to ferrite.

Ferrite has not yet been positively identified by electron diffraction studies of samples irradiated to lower fluences. However, those samples containing regions in which roughly 5 or more displacements per atom were created are magnetic. The degree to which the samples exhibit magnetism increases with increasing displacements. We have previously observed irradiation-induced austenite-to-ferrite transformation in Type 321 stainless steel, where the extent of transformation was greater than in Type 316.[7]

A specimen containing no helium was irradiated at 500°C to approximately 5 displacements per atom. Voids were produced, but the density was several orders of magnitude lower than that observed in samples containing helium. Void nucleation appears to be significantly enhanced by helium. Further, the hydrogen introduced during proton irradiation apparently plays no significant role in the nucleation of voids.

The voids produced by an irradiation at 600°C to 21 displacements per atom are shown in Figure 4a. Note that void faceting is somewhat less distinct than at

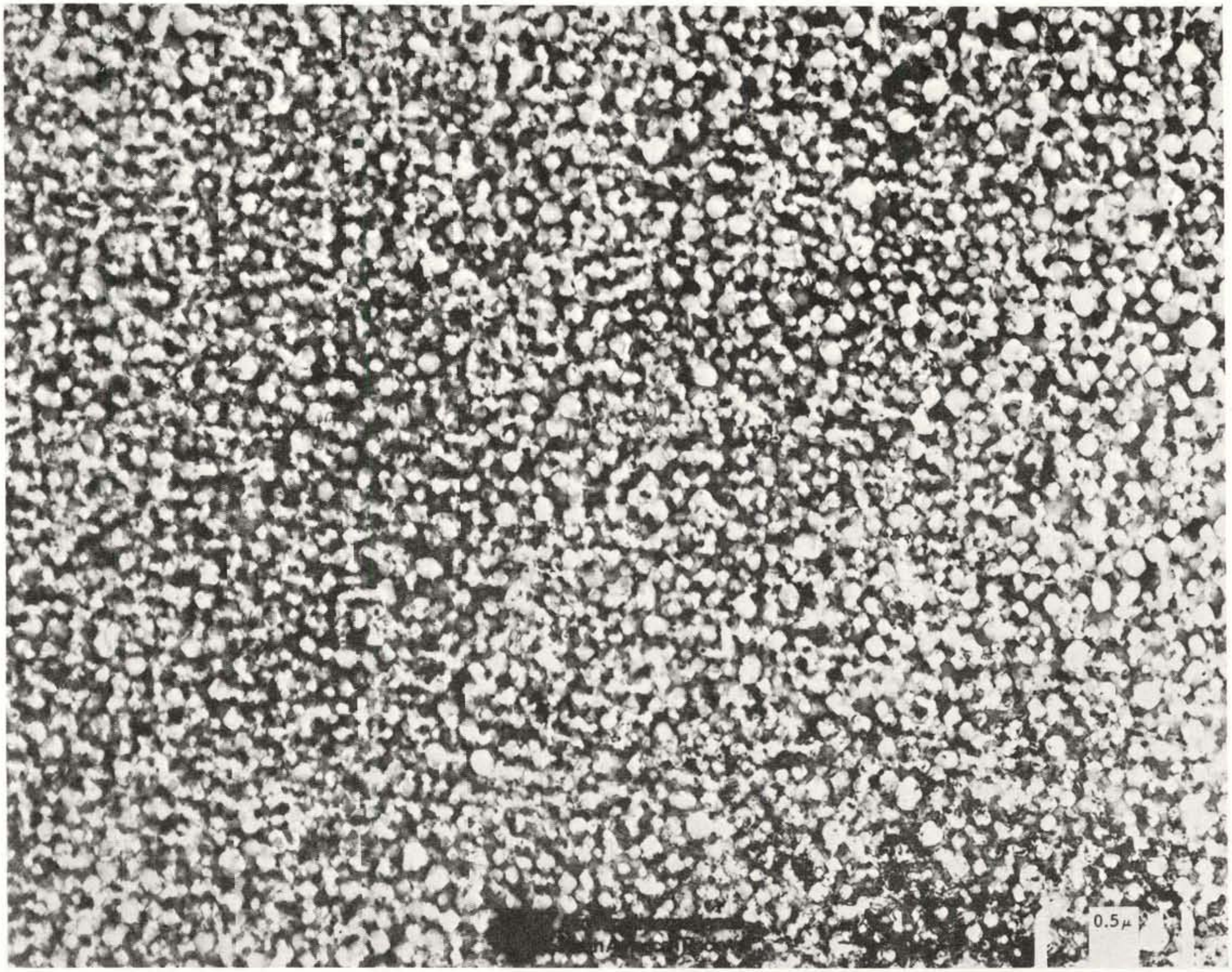


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Figure 3. Voids in Type 316 Stainless Steel Proton-Irradiated at 500°C to Produce 100 Displacements per Atom (Sample 59)

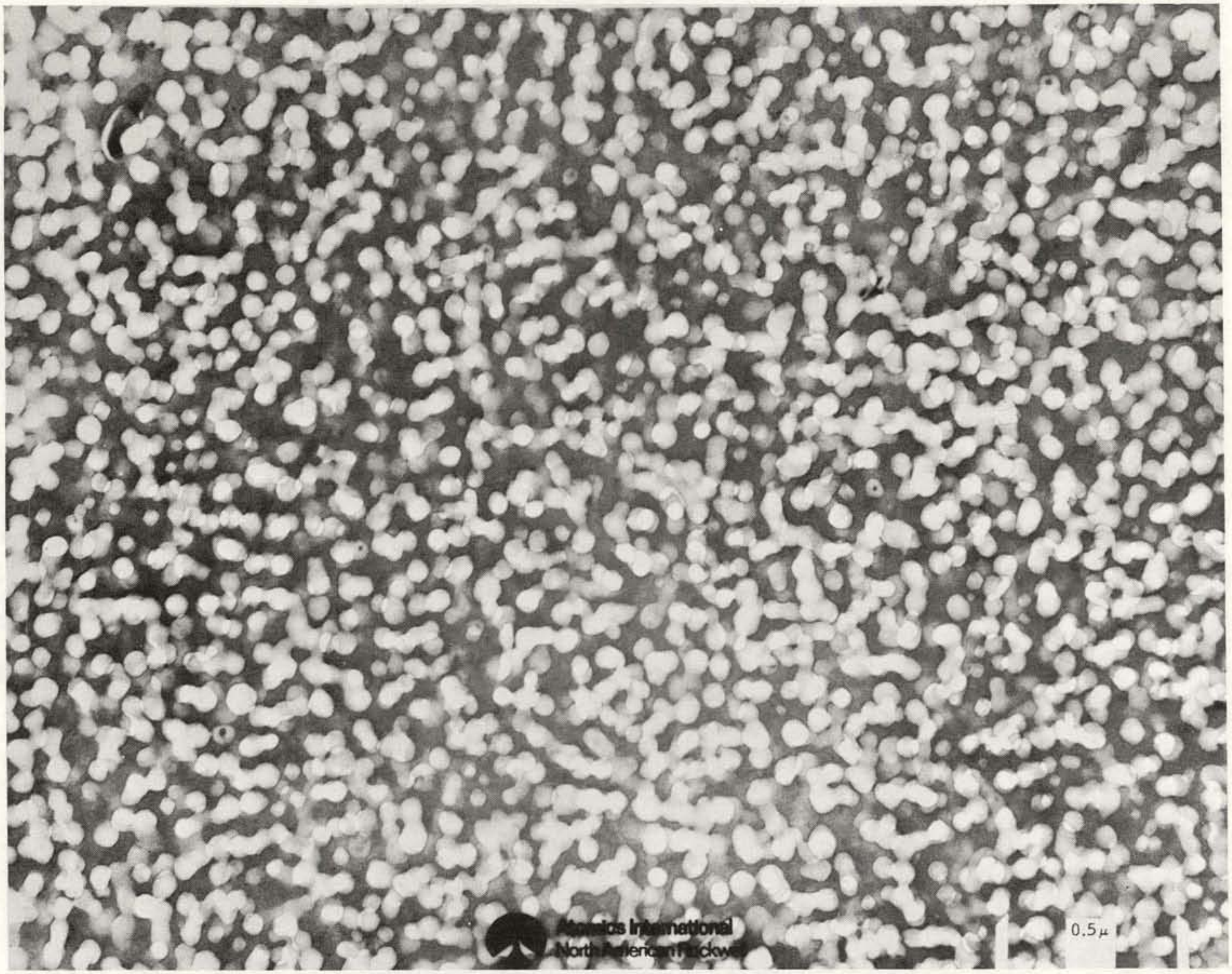

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Figure 4a. Voids in Type 316 Stainless Steel Proton-Irradiated at 600°C to Produce 21 Displacements per Atom (Sample 38)

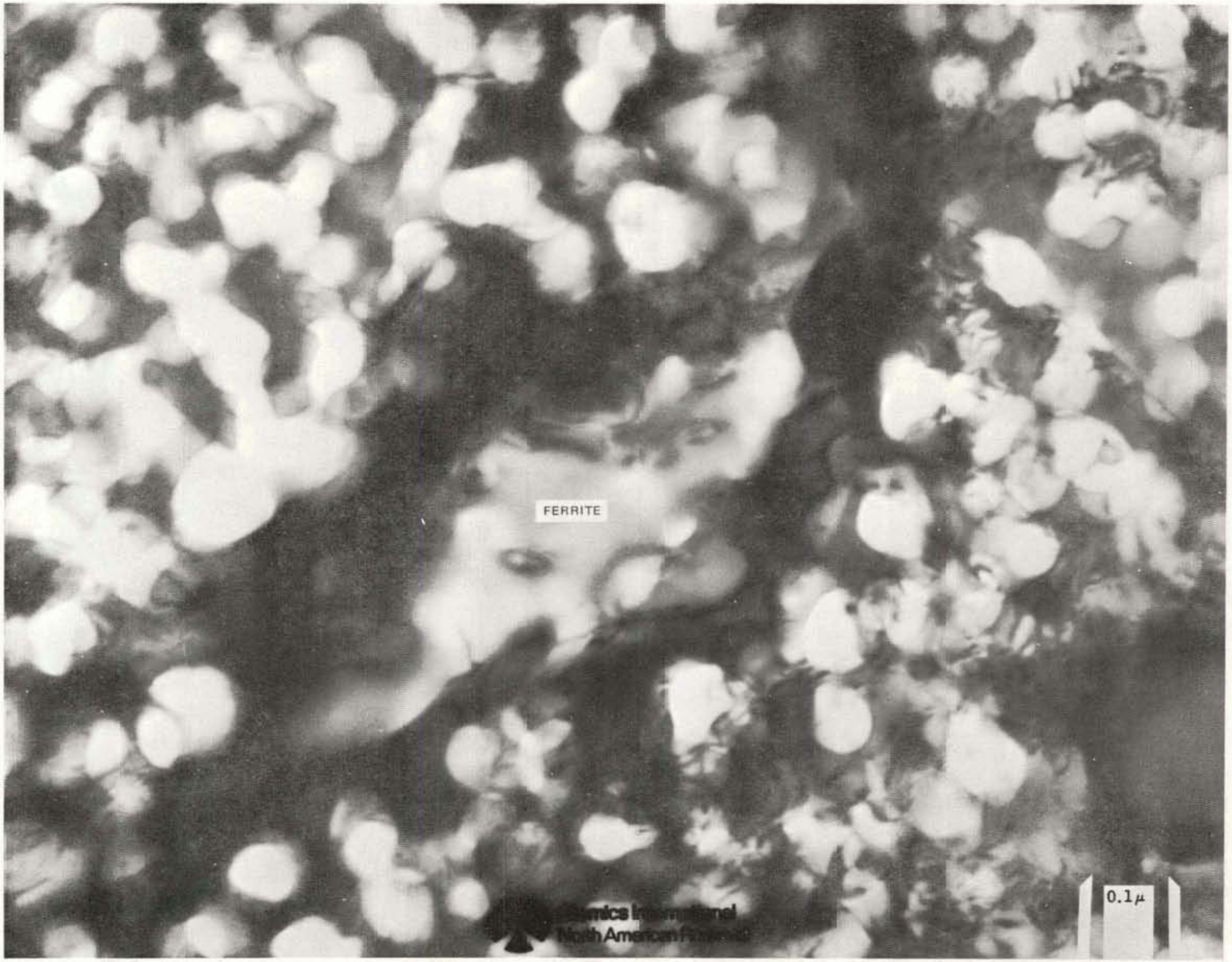

Figure 4b. Micrograph Showing a Region of Ferrite Which Transformed During Irradiation

500°C and that there is considerable evidence of voids having joined to form elongated complexes. As can be seen from Figure 1 and Table 1, the average swelling at this displacement level is about 24%, whereas at 500°C, the same damage produces only about 7% swelling. Also, the voids produced at 600°C are less numerous and larger than at 500°C.

Ferrite was detected by electron diffraction of the sample irradiated at 600°C. A small region of ferrite growing from a grain boundary is indicated in Figure 4b. Since no ferrite was observed by microscopy of samples irradiated to about 20 displacements per atom at 500°C, this preliminary results suggests the austenite-to-ferrite transformation occurs more readily as the irradiation temperature is increased to 600°C.

In Figure 5 we show a plot of the maximum, average, and minimum void diameters plotted as functions of the number of displacements created by irradiation at 500 and 600°C. The lines through the 500°C data are drawn according to least squares fits. The increase in the minimum void size with increasing damage is particularly interesting. This means that void nucleation occurs relatively early in an irradiation and that further defect production merely leads to growth of already-existing voids. Such an effect is not unexpected in view of the fact that in these experiments the entire helium content is injected prior to irradiation.

In irradiation at 500°C, no dependence of void density on number of atom displacements has been observed. However, as is shown in Figure 6, there is a distinct dependence of void density on atom displacement rate. From Figure 6 it is apparent that a higher displacement rate can result in an enhancement of void nucleation rate. Note also that, from Table 1, an increase in displacement rate at 600°C leads to an increase in void density. These results are in agreement with the observation made above that nucleation of voids occurs early in an irradiation.

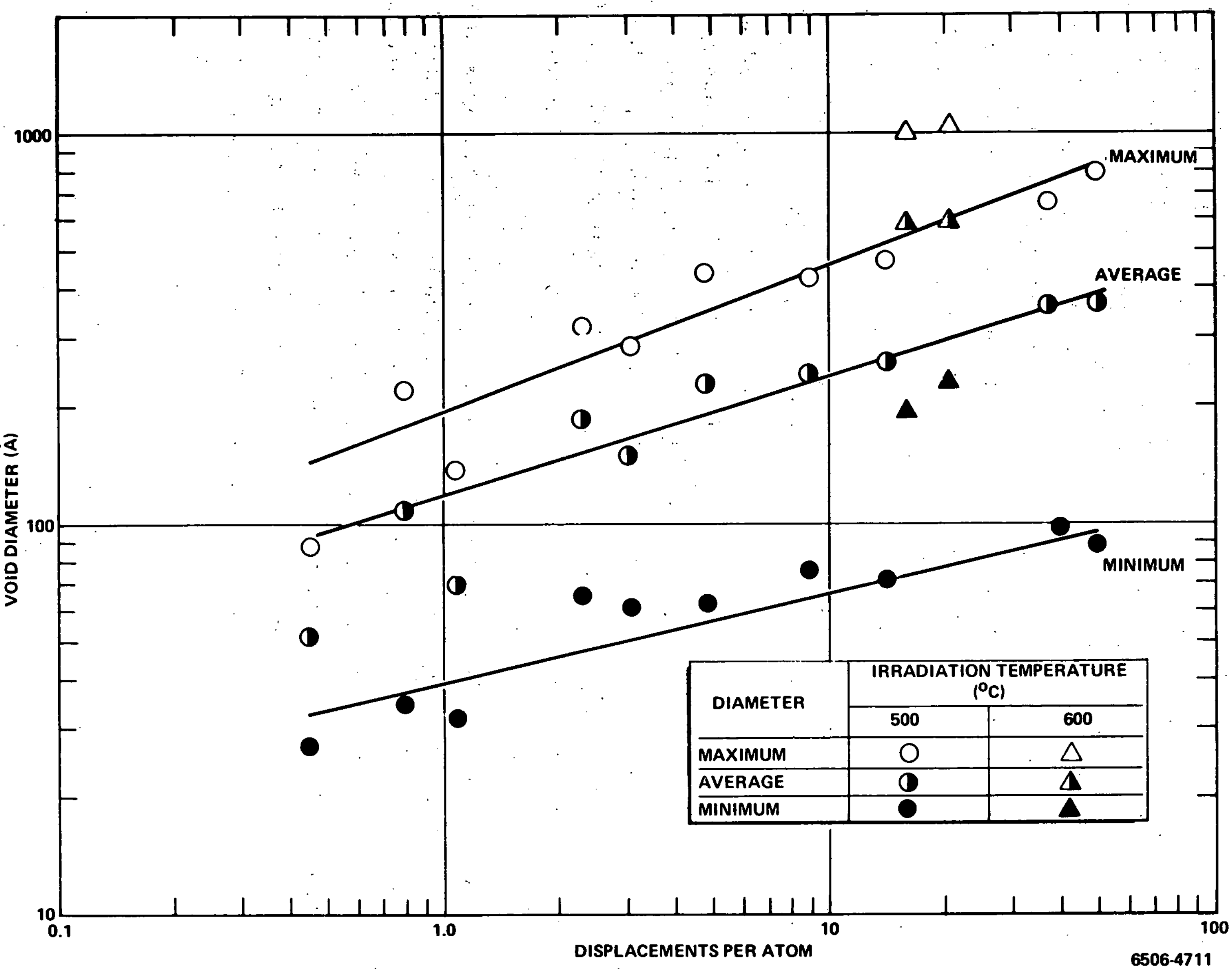

Figure 5. Maximum, Average, and Minimum Void Diameters as a Function of Displacement Damage for Proton-Irradiated Type 316 Stainless Steel

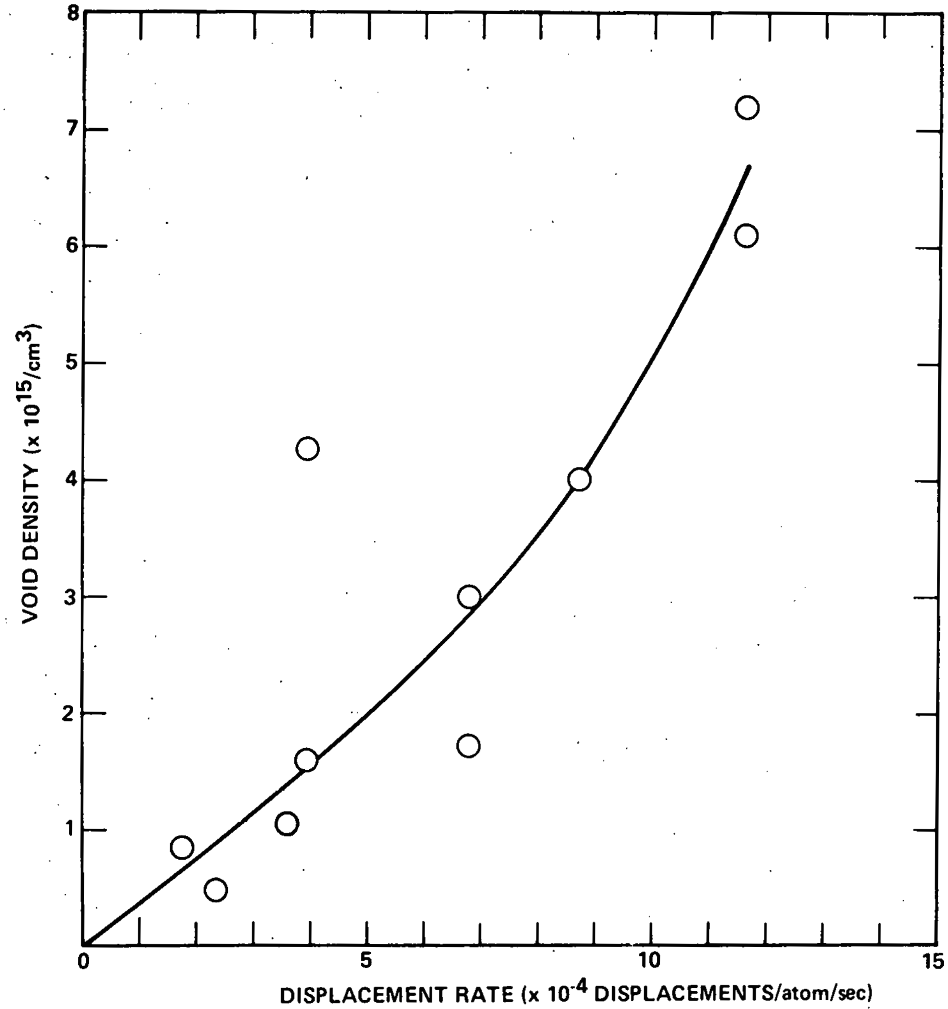

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Figure 6. Void Density as a Function of Atom Displacement Rate in Proton-Irradiated Type 316 Stainless Steel

V. SUMMARY

In proton irradiations performed at 500°C on samples pre-injected with helium, the volume swelling increases nearly linearly with number of atom displacements. There is no evidence of a limit to swelling up to a damage of 50 displacements per atom. Even at very large values of swelling, the voids show little evidence of agglomeration. The irradiations cause a progressive transformation of austenite to ferrite with increasing number of atom displacements. An increase in the minimum void size with number of atom displacements and an increase in void density with atom displacement rate both indicate that void nucleation occurs relatively early during the irradiation.

The swelling observed after 600°C irradiation to about 20 displacements per atom is greater than that at 500°C. The void number density is lower, the average void size is larger, and agglomeration of voids is frequent at 600°C.

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