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ATOM DISPLACEMENTS AND VOID FORMATION IN PROTON-IRRADIATED TYPE 316 STAINLESS STEEL

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ABSTRACT

The cross section for atomic displacement was calculated as a function of proton penetration into Type 316 stainless steel. The cross section at a given penetration is reduced from that used in previous work. This is the result of new information on proton range, modification of the cross-section calculations, and adoption of a higher displacement threshold energy. Swelling, void size, and void density data previously published are plotted against recalculated displacement damage. In general, the data are merely shifted to lower damage levels, and only one conclusion drawn in a previous publication appears to need modification. A short discussion on the effect of voids on proton energy and displacement cross section as a function of proton penetration is also included.

I. INTRODUCTION

In a recent publication,[1] the range of protons in Type 316 stainless steel, from both experimental and computational points of view, was discussed. The range of ~1 MeV protons was determined by the depth at which voids are observed after irradiation at 500°C. Calculated range-energy data which best agreed with the experimental results were then used to determine the dependence of proton energy on penetration. In this report, we proceed to a consideration of the atom displacement cross section and its dependence on proton energy, and hence on penetration. A procedure for calculation of the cross section at a particular energy is outlined; this procedure is more rigorous than one which was used previously.[2] The new cross-section values are used to revise the displacements per atom (dpa) calculated in previous work on stainless steel. Since the dpa obtained here differ substantially from those used in earlier publications,[3-5] revised swelling and other void data are presented.

The samples used in most of this work have been solution annealed for 1 hr at 980°C and aged for 8 hr at 760°C. This provided a stable microstructure in which void formation could be studied as it occurred during proton irradiation. Unfortunately, few samples with this microstructure have been irradiated with neutrons. Therefore, although we are in a position to calculate, with some assurance, the dpa generated during proton irradiation, we cannot yet make an attempt to correlate neutron and proton data vis-a-vis displacement damage.

II. DISPLACEMENT CALCULATIONS

The discussion in this section is based on reports by Keefer et al.[2] and Doran et al.[6] The latter report is the more complete, and the reader is referred to it for a detailed description of displacement calculations for various particles. Calculations for protons are sketched briefly in the paragraphs that follow. At a given proton energy, the displacement cross sections for iron and stainless steel have been found to be very similar.[6] Therefore, our calculations will be made for iron, as a reasonable substitute for stainless steel.

For protons in the energy range of interest (~ 1 MeV), the Rutherford scattering law holds; and, for protons of energy E incident on iron,

$$d\sigma(T) = 4\pi a_o^2 \, (\gamma Z_p)^2 Z_{Fe}^{\;2} \, \frac{M_p E_R^{\;2} dT}{M_{Fe} E\, T^2} \quad \cdot \qquad \ldots(1)$$

$d\sigma(T)$ is the differential cross section for transferring energy in the interval T to T+dT. In Equation 1, a_o is the radius of the first Bohr orbit 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. The effective proton charge is γZ, where $\gamma \leqq 1$ but differs significantly from 1 only at low proton energies ($\lesssim 0.2$ MeV). E_R is the Rydberg energy, 13.6 eV. With the appropriate numerical values, Equation 1 becomes, for the present case,

$$d\sigma(T) = 7.93 \times 10^{-13} \, \frac{\gamma^2}{E} \frac{dT}{T^2} \; , \qquad \ldots(2)$$

where E is in eV. The cross section for production of all displacements is given by

$$\sigma_d(E) = \int_{E_d}^{T_m} d\sigma(T) \cdot n(T) \ , \qquad \ldots(3)$$

where E_d is the threshold energy for creation of displacements, and T_m is the maximum energy that a particle of energy E can transfer to a lattice atom.

The term n(T) in Equation 3 has been introduced to account for secondary and higher-order displacements. At transfer energies below E_d, no displacements occur, and n(T) = 0. In the interval $E_d \leq T < 2E_d$, the primary knock-on atom is incapable of displacing a second atom, and n(T) = 1. In the interval $2E_d \leq T \leq T_m$,[6]

$$n(T) = \frac{\beta T_{dam}}{2E_d} \ . \qquad \ldots(4)$$

In Equation 4, β is a term introduced to account for anisotropic scattering. This is a modification of the old Kinchin-Pease[7] model, in which hard sphere (isotropic) scattering was assumed. Doran et al.[6] have recommended that a value of $\beta = 0.8$ be used. In Equation 4, T_{dam} is the total elastic energy available for the creation of displacements. That is, energy dissipated in electronic excitation, which does not contribute to displacements, has been deleted. From Robinson's[8] treatment of the Lindhard et al.[9] theory,

$$T_{dam} = T\ [1 + kg\ (\epsilon)]^{-1} \ , \qquad \ldots(5a)$$

where, for a pure metal of atomic weight A,

$$k = 0.1337\, Z^{2/3}/A^{1/2} \ , \qquad \ldots (5b)$$

$$g(\epsilon) = \epsilon + 0.4024\, \epsilon^{3/4} + 3.401\, \epsilon^{1/6} \ , \qquad \ldots (5c)$$

$$\epsilon = T/0.08693\, Z^{7/3} \ . \qquad \ldots (5d)$$

Using Equations 4 and 5a in Equation 3,

$$\sigma_d(E) = 7.93 \times 10^{-13} \frac{\gamma^2}{E} \left\{ \int_{E_d}^{2E_d} \frac{dT}{T^2} + \frac{\beta}{2E_d} \int_{2E_d}^{T_m} \frac{dT}{T\,[1 + kg(\epsilon)]} \right\} . \qquad \ldots (6)$$

Equation 6 has been integrated numerically by Doran et al. ; and σ_d for iron as a function of proton energy, E, has been presented in tabular form in Reference 6.

As can be seen by integration of Equation 6, σ_d for proton irradiation is quite sensitive to the choice of E_d, and it is important that the best possible value be assigned to E_d. Doran et al. used a value of 40 eV; this assignment was based on both theoretical and experimental results. Since 25 eV was used for E_d in our earlier work,[2] this accounts for most of a rather large difference in cross-section results obtained by Keefer and Doran. Since the report by Doran et al. is intended to serve as a basis from which experimenters in the field of void formation can compare their results, for the sake of consistency, 40 eV will be used for E_d in iron. It is believed, however, that this value may be revised, probably downward, in the face of future experimental and theoretical results. In fact, Jung et al.[10] have recently conducted experiments which bear on this problem. They report results on the anisotropy, with respect to direction of atom ejection in the metal lattice, of the threshold energy in copper and platinum. For copper, their mean threshold energy over all atom ejection directions is between 25 and 30 eV. The threshold energy for iron or stainless steel is probably not significantly different from that for copper, so the results of Jung _et al._ cast some doubt on the assignment of 40 eV to E_d.

In Reference 1, it was pointed out that the depth to which voids were observed exceeded by more than 10^{-4} cm the proton range calculated by Janni[11] at incident energies between 0.75 and 1.05 MeV. After considering, and abandoning, several mechanisms whereby the proton range might be extended by such a large amount, it was concluded that the proton range in iron is best represented by the calculations of Williamson _et al._[12] The dependence of proton energy on depth of penetration into iron (as a substitute for stainless steel) was then discussed. The new information on proton range lowers the proton energy, and therefore σ_d, at a particular depth. Further, as outlined previously, our displacement cross-section calculations have been revised, and inclusion of a higher displacement threshold energy decreases the cross section at a given depth. The dependence of proton energy, E, and displacement cross section, σ_d, on depth of penetration is shown in Figure 1. The solid curves for energy and cross section are taken from information given in Reference 6 and Equation 6, respectively. These curves have been drawn for the case of a proton with incident energy of 1.025 MeV.*

In Reference 1, the extension of proton range by the presence of voids was considered; this was based on the work of Schwartz and Ardell.[13] Recently, a more detailed account of the effect of voids, by Odette _et al._,[14] was received. This work prompted us to include, as an aside, a discussion of the effect of voids on proton energy, range, and displacement cross section in this report. Note that the solid curves in Figure 1 are plotted against "mass depth" (i.e., that depth through which protons pass in the absence of voids). The dependence

*To determine the energy and cross section at some depth for a proton at lower incident energy E', the ordinate can be shifted to the right until it intersects the energy curve at E'. The abscissa is then shifted a like amount.

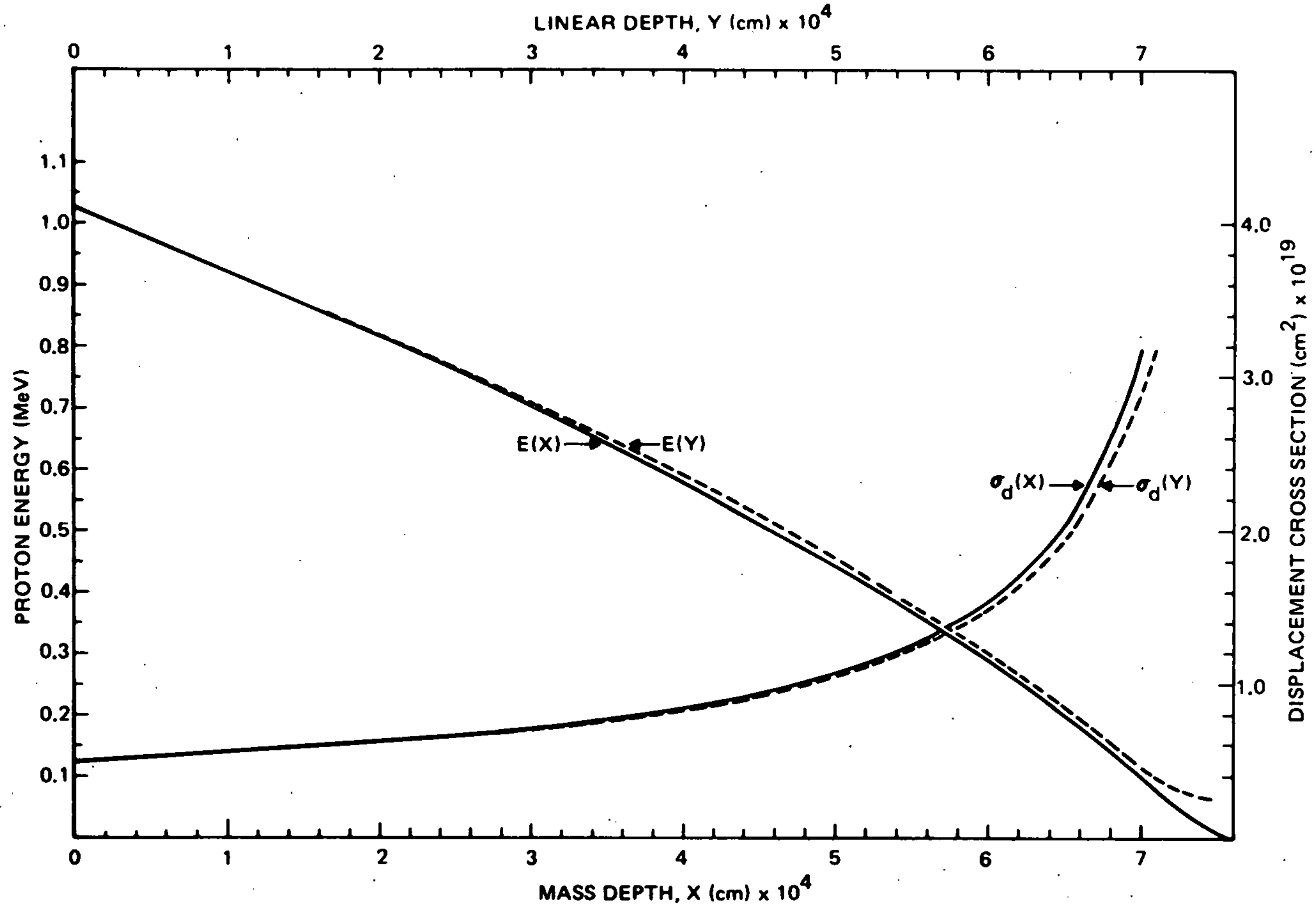

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Figure 1. Proton Energy and Displacement Cross Section vs Depth of Penetration in Type 316 Stainless Steel (Mass depth refers to samples without voids, while linear depth refers to samples containing voids. Although the two scales are the same, the linear and mass depths are plotted on different abscissas for emphasis.)

of proton energy and displacement cross section for a sample containing voids is shown by dashed curves. These curves were obtained by combining, according to the theory of Odette et al.,[14] swelling data obtained from a "cross-section" TEM foil with the calculated values of E and σ_d (solid curves). The foil from which the data were taken had a maximum swelling value of ~10%. Note that, except near the end of the proton range, the presence of voids has little effect on either E or σ_d. Of course, the greater the swelling, the greater will be the effect on energy and displacement cross section.

In most of our work, TEM foils are extracted from samples so that their planes lie normal to the proton beam. From Figure 1, it is obvious that the position of these foils is important, with respect to determining the number of defects created. In our extraction technique, the sample is back-thinned until the desired position is reached, typically 5×10^{-4} to 6×10^{-4} cm from the front surface. This position is monitored by measuring the thickness of the front portion of the sample. Since β-ray attenuation is used in these measurements, it is important to point out that we are measuring "mass depth," and that the solid curves in Figure 1 are appropriate for our use. In other thickness-measuring techniques not utilizing radiation attenuation, the presence of voids must be accounted for by the procedure used in getting the dashed curves in Figure 1.

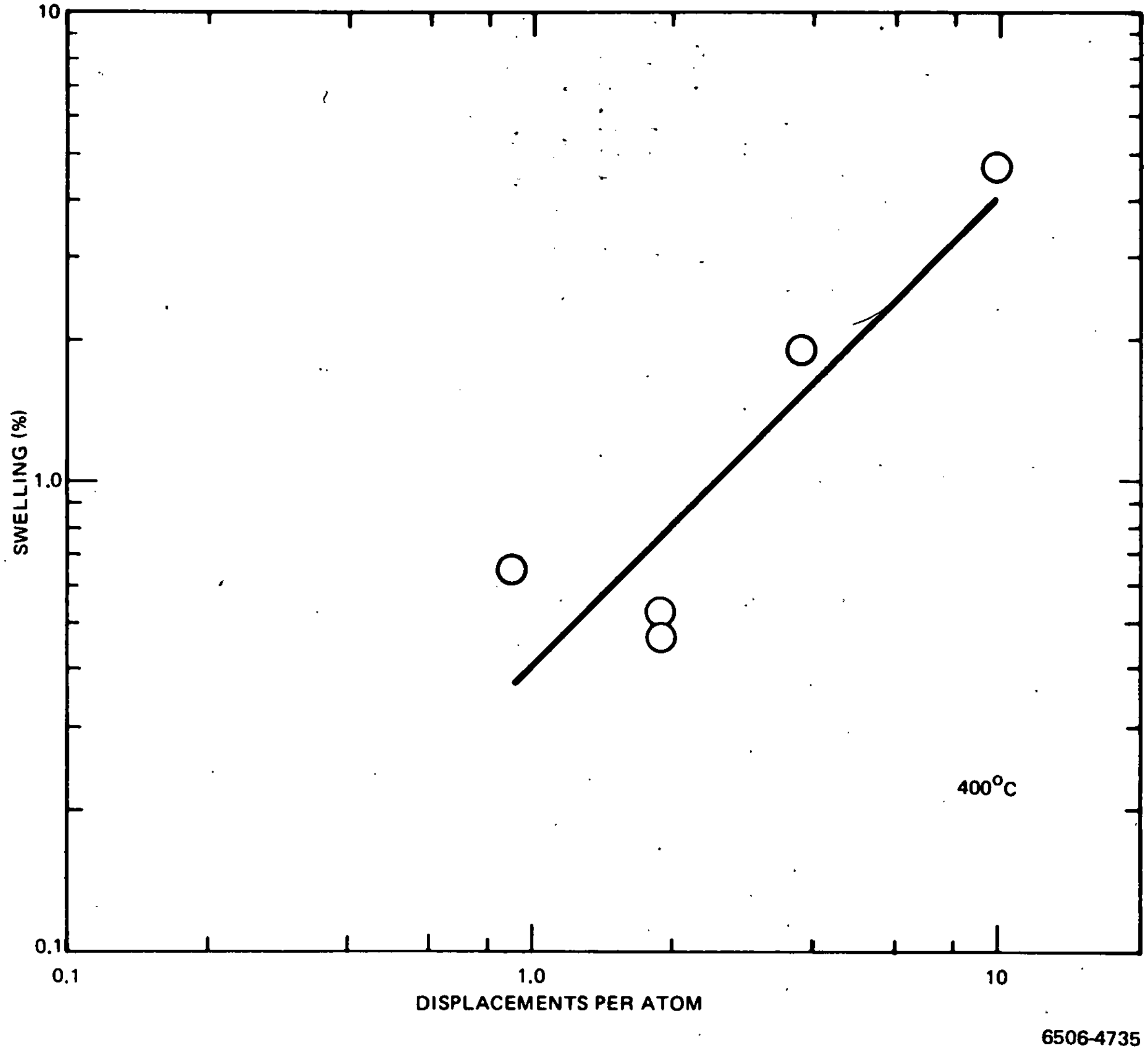

Figure 2. Swelling, $\Delta V/V_O$, in Percent vs Displacements Per Atom in Type 316 Stainless Steel Irradiated with 1 MeV Protons at 400°C (Samples were pre-injected with ~5 appm helium at room temperature.)

III. VOID DATA

Using the displacement cross-section curve in Figure 1 (solid line), the dpa for each TEM foil extracted from the solution-annealed and aged samples of Type 316 stainless steel have been recalculated. With one possible exception, to be discussed, the new dpa values do not materially affect conclusions drawn in previous publications.[3-5] In general, void data are simply shifted to the lower damage level, and remain approximately parallel to those published earlier. As examples, in Figures 2 through 6, are shown the dependence of swelling,* average void diameter, and void density on displacement damage. Lines through all data are drawn according to least squares fits. The swelling dependences at 400, 500, and 600°C are given by the following equations:

$$\frac{\Delta V}{V_o} (\%) = 0.41 \, (\text{dpa})^{0.99} \qquad 400°C \qquad \ldots (7a)$$

$$\frac{\Delta V}{V_o} (\%) = 0.57 \, (\text{dpa})^{1.4} \qquad 500°C \qquad \ldots (7b)$$

$$\frac{\Delta V}{V_o} (\%) = 0.10 \, (\text{dpa})^{2.6} \qquad 600°C \qquad \ldots (7c)$$

The only conclusion which may need to be altered from that drawn in previous work concerns the swelling which is obtained in the course of alternating helium injection at room temperature and proton irradiation at 500°C. In Reference 4, it was concluded (from two data points) that such a procedure apparently yielded a different dependence of swelling on displacement damage than is observed following a single helium injection and proton irradiation.

*Swelling is here defined as $\Delta V/V_o$, where ΔV is the increase in volume of a sample of original volume V_o. In the past, we have presented our data in terms of void volume fraction, $\Delta V/V$. There is little difference in the two quantities, except at values above roughly 10%.

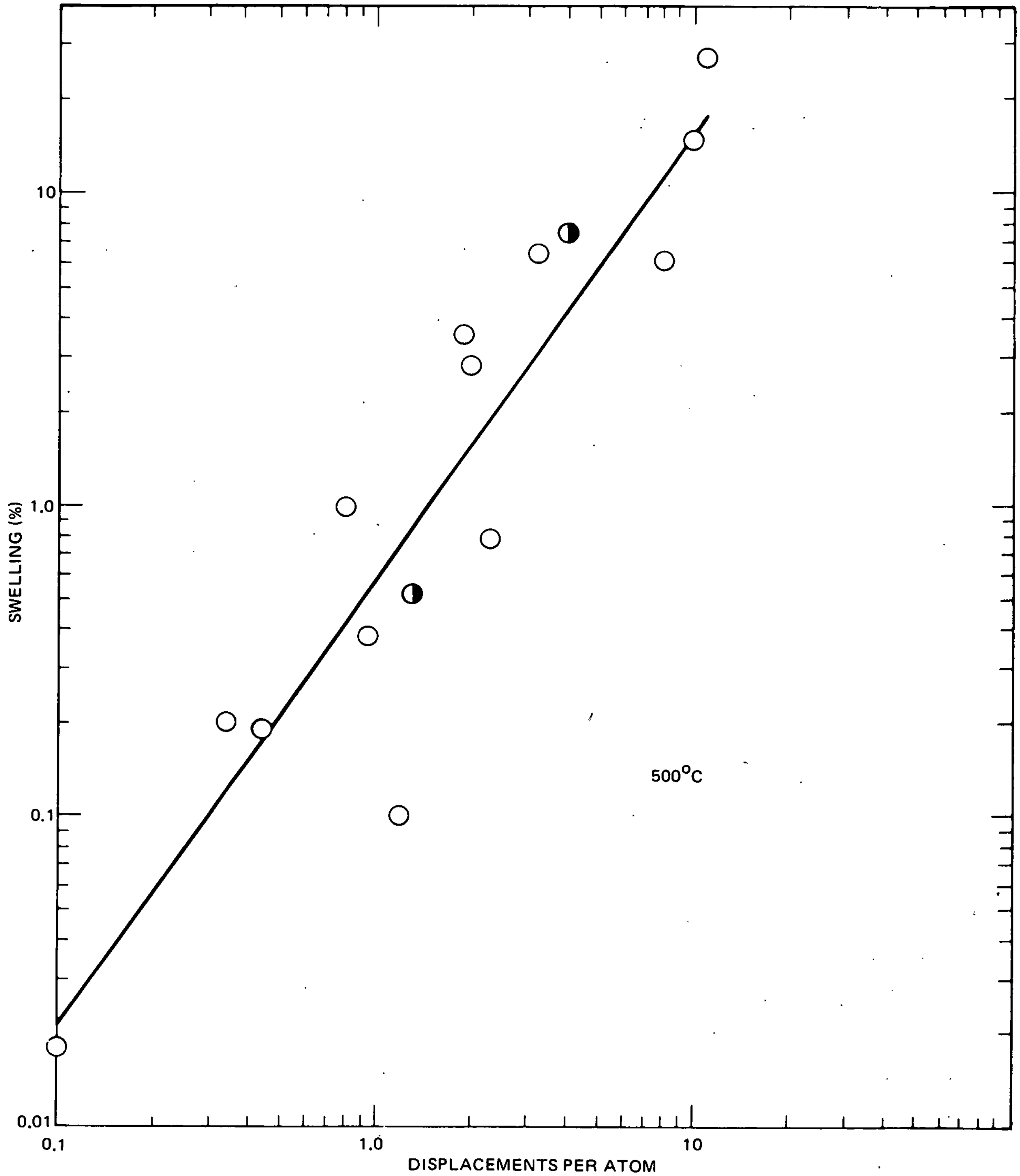

Figure 3. Swelling, $\Delta V/V_0$, in Percent vs Displacements Per Atom in Type 316 Stainless Steel Irradiated at 500°C with Protons Near 1 MeV. Open circles refer to samples which were pre-injected with ~5 appm helium at room temperature. Half-filled circles refer to a sample which was alternately injected with helium at room temperature and irradiated at 500°C with protons.

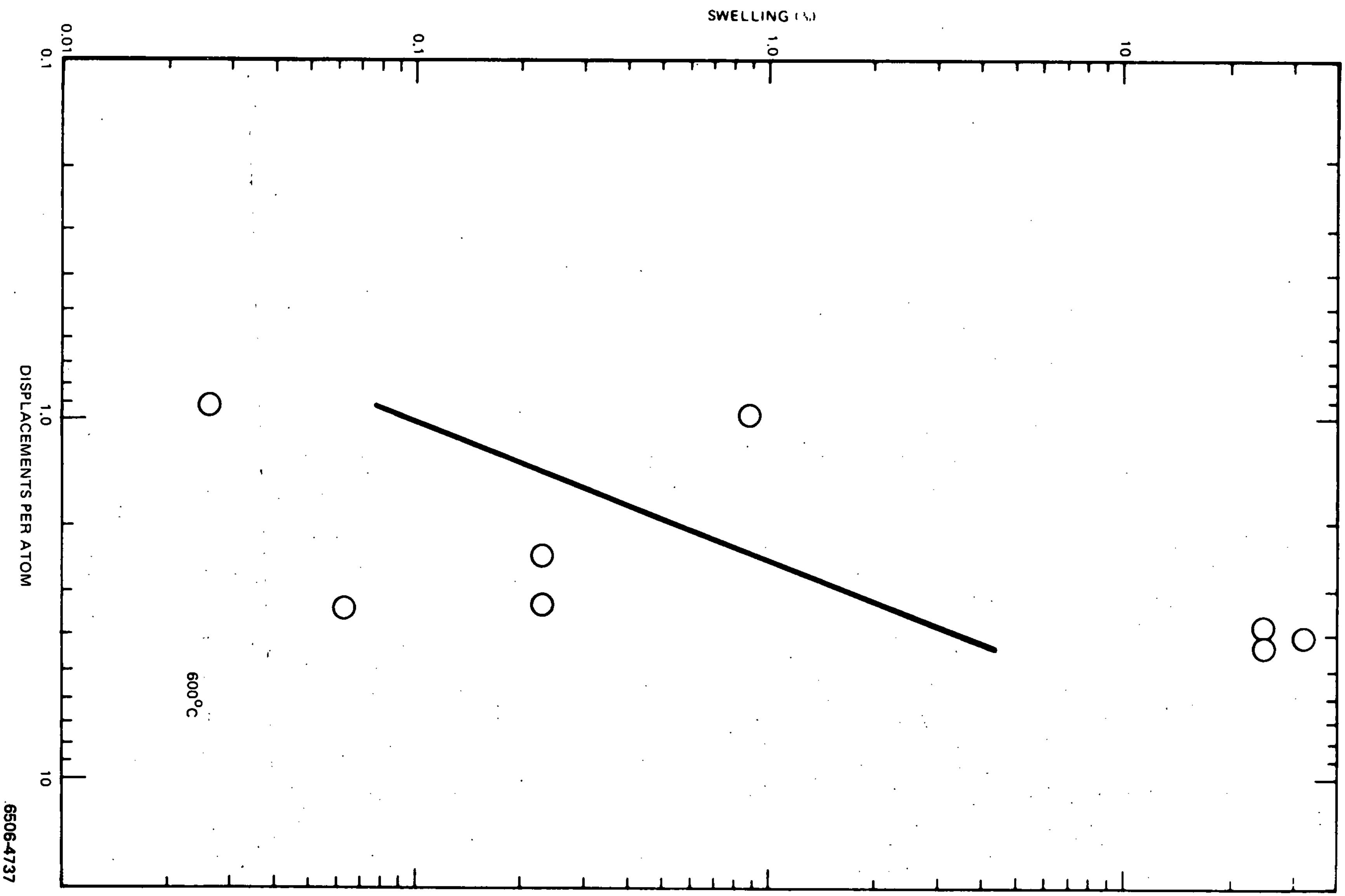

Figure 4. Swelling, $\Delta V/V_o$, in Percent vs Displacements Per Atom in Type 316 Stainless Steel Irradiated at 600°C with Protons Near 1 MeV (Samples were pre-injected with ~5 appm helium at room temperature.)

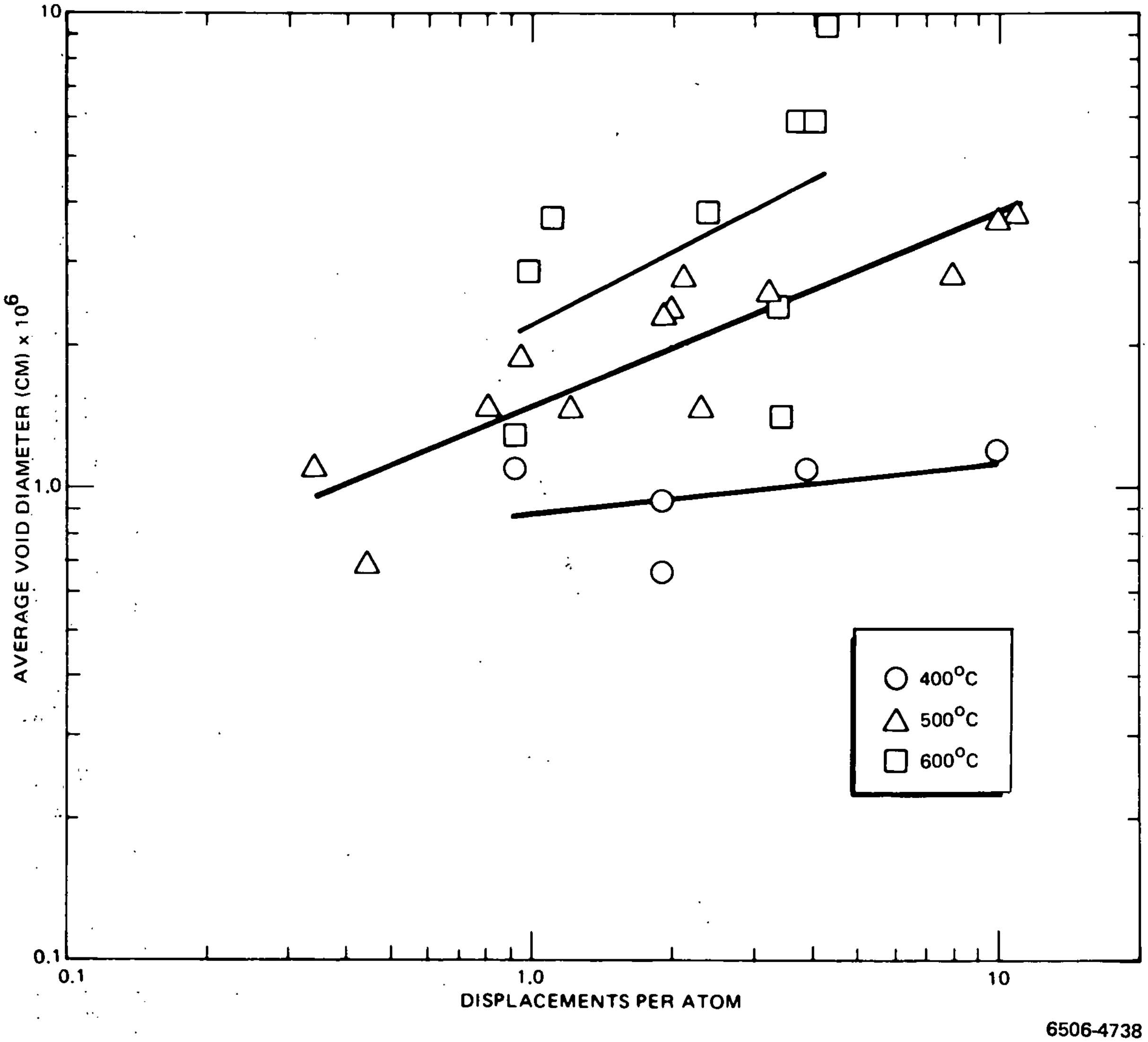

Figure 5. Average Void Diameter vs Displacements Per Atom in Type 316 Stainless Steel Irradiated at 400, 500, and 600°C with Protons Near 1 MeV (Samples were pre-injected with ~5 appm helium at room temperature.)

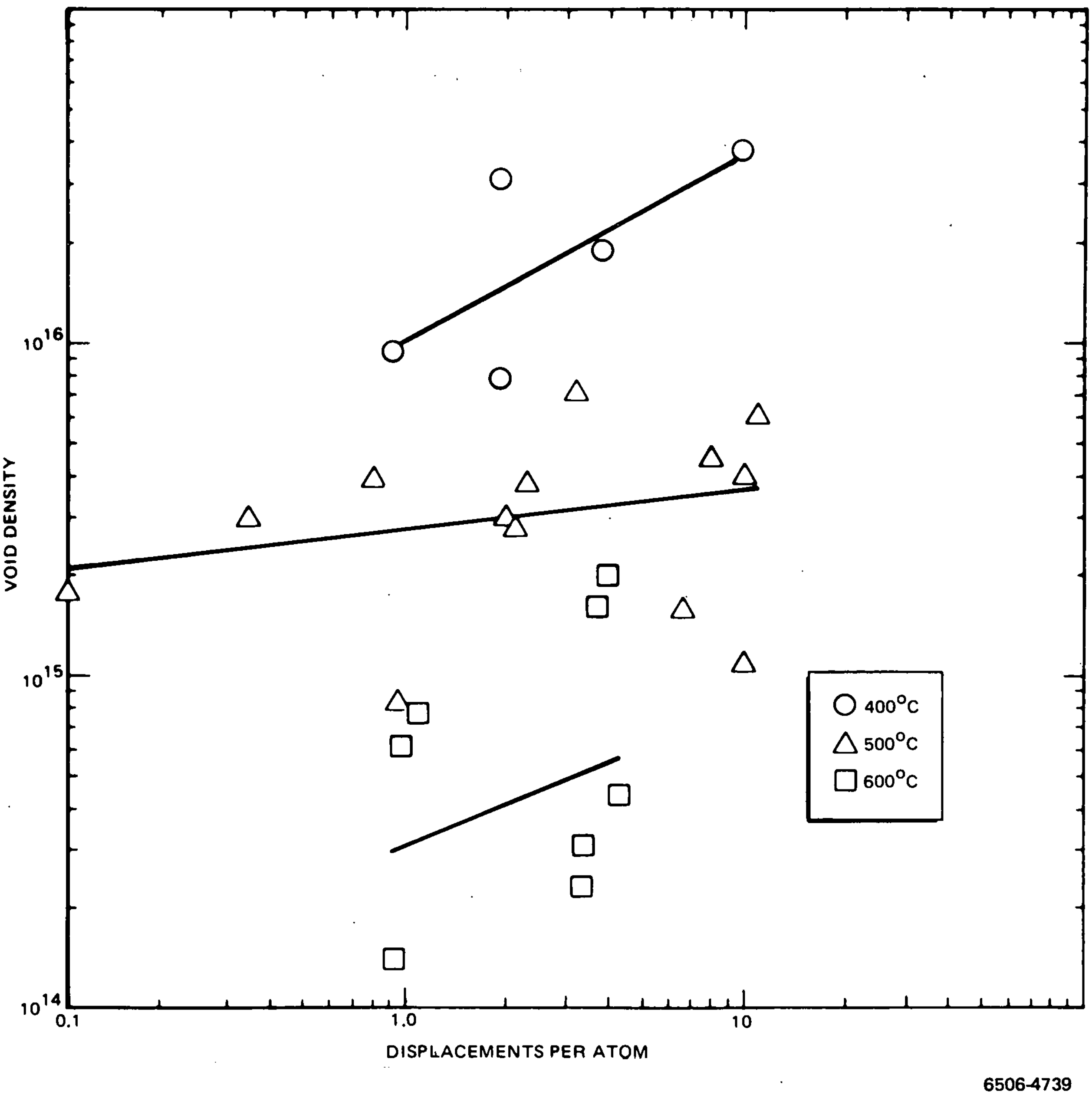

Figure 6. Void Density vs Displacements Per Atom in Type 316 Stainless Steel Irradiated at 400, 500, and 600°C with Protons near 1 MeV (Samples were pre-injected with ~5 appm helium at room temperature.)

The new dpa calculations have resulted in a shift in the data points for the alternate injection-irradiation experiment, such that they now lie much closer to the data for the single injection and irradiation procedure. The data for the alternate experiment are shown as half-filled circles in Figure 3. Other conclusions drawn in Reference 4 concerning void size, density, and nucleation remain the same.

IV. CONCLUSIONS

1) At a given sample depth and proton fluence, the calculated dpa have been revised downward by: (a) observation that the proton range exceeds that originally supposed, (b) revised calculations of the displacement cross section, and (c) use of 40 eV in place of 25 eV as the displacement threshold energy.

2) In general, data for swelling, void diameter, and void density are simply shifted to lower damage levels, and remain approximately parallel to the data which have been published earlier.

3) The dependence of swelling on dpa for a sample alternately helium injected and proton irradiated now is in better agreement with results obtained from samples given a single injection and irradiation.

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