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Erratum for: "Master equation and Fokker-Planck methods for void nucleation and growth in irradiation swelling", "Vacancy cluster evolution and swelling in irradiated 316 stainless steel" and "Radiation swelling behavior and its dependence on temperature, dose rate, and dislocation structure evolution"

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We have recently discovered an error in our void nucleation code used in three prior publications [1–3]. A term was omitted in the model for vacancy re-emission that (especially at high temperature) affects void nucleation and growth during irradiation as well as void annealing and Ostwald ripening of the size distribution after irradiation. The omission was not immediately detected because the calculations predict reasonable void densities and swelling behaviors when compared to experiment at low irradiation temperatures, where void swelling is prominent. (Comparable neutron irradiation experiments are less prevalent at higher temperatures, e.g., > 500 C.) Neglecting long-range interactions for simplicity, vacancies are thermally emitted from a sessile void of x vacancies and m inert gas atoms at a rate:

$$4\pi r(x-1)D_v(T)C_v^{eq}(T)e^{(E(x,m)-E(x-1,m))/kT} \quad (1)$$

in terms of the vacancy diffusivity, D_v and thermal equilibrium vacancy concentration, C_v^{eq} . Here, $r(x-1)$ is the radius of the (spherical) void or bubble minus one vacancy and $E(x, m)$ its energy. Given that:

$$\frac{4\pi}{3}r^3(x) = x\Omega \quad (2)$$

for atomic volume, Ω , and assuming:

$$E(x, m) = 4\pi r^2(x)\gamma(x) + F(x, m, T) \quad (3)$$

the difference for $x \gg 1$ is:

$$E(x, m) - E(x-1, m) \simeq \frac{2\gamma(x)}{r(x)}\Omega + 4\pi r^2(x)\left[\frac{d\gamma}{dr}\right]\Omega - P(x, m, T)\Omega \quad (4)$$

where γ is the (size-dependent) void surface energy and $P\Omega$ is the linearized difference of the Helmholtz free energy of the confined gas. The first term in Eq. 4 was inadvertently omitted from the calculation, and from an earlier version developed by Wehner and Wolfer [4]. After correcting and repeating our previous calculations, we show results for the incubation dose to reach 1% volumetric swelling in Fig 1. The temperature cutoff for void swelling is shifted downwards (cf. solid (dotted) curve for the old (new) results) because the larger energy difference promotes vacancy emission from the voids at high temperatures. The location of this cutoff temperature varies with dose rate as before, but it is also sensitive to the assumed

surface energy and internal gas pressure.

The results in Fig. 1 suggest that void formation energies must be reduced from the values predicted by the usual capillary approximation in order to agree with swelling experiments. It is well-known that the surface energy of a metal is affected by chemisorbed elements, like oxygen, sulfur, and carbon; and models commonly reduce ideal surface energies by about a factor of two to account for impurities. Similarly, helium content is expected to reduce the thermal emission of vacancies and stabilize small, sub-critical vacancy clusters. By setting the surface energy to a constant 0.8 J/m^2 (approximately 0.3 times its correct, clean value), and by generating helium at 0.3 appm/dpa and incorporating up to 0.8He/vacant site in small voids, we nearly reproduce the incubation times predicted by the original calculations (long-dashed curve). Evidently, a reduction in surface energy is sufficient to explain incubation at low temperatures, while helium accumulation is required for swelling at high temperatures. However, the constant, reduced surface energy is an incomplete and non-rigorous correction to the traditional capillary approximation in void nucleation. The proper influence of surface-active species and the roles of surface energy and surface stress (which quantities differ for solids) in void nucleation must be examined further. We will address these issues in a forthcoming publication and show that a correct treatment significantly reduces the void nucleation barrier versus expectations from the capillary approximation, moving back towards our earlier results.

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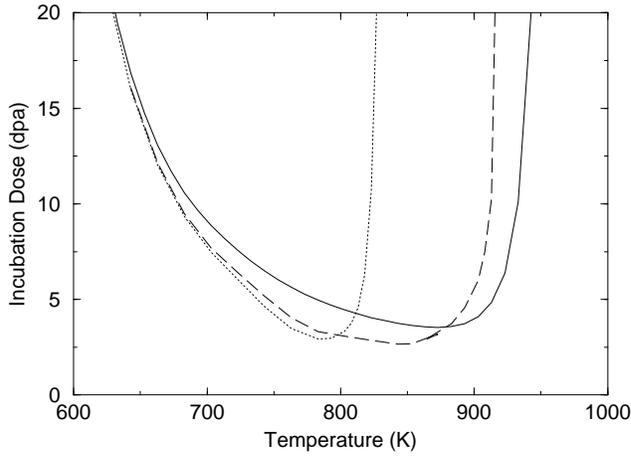


FIG. 1: Cumulative incubation dose in dpa required to reach $\Delta V/V = 1\%$ swelling as a function of temperature, at a dose rate of 10^{-6} dpa/s. The vacancy migration energy is $E_m = 1.35\text{eV}$; other parameters are as before, including the (true) bare surface energy. The solid curve shows the results of the original, erroneous calculation. In contrast, a corrected calculation predicts that swelling does not occur for clean void surfaces with no internal pressure (i.e., without oxygen or helium in the voids), the situation which the original work was intended to represent. The dashed line uses a surface energy that is reduced to 0.8 J/m^2 (approximately $0.3\times$ the bare value), with no internal gas pressure. The long-dashed curve shows the results for 0.8 J/m^2 , while introducing helium at 0.3 appm/dpa up to a maximum density of $0.8\text{ He/vacant site}$ in small voids.