8.4 Dislocation vs. Production Bias Revisited with Accounted of Radiation-Induced Emission Bias: Void Swelling Under Electron and Light Ion Irradiation — V. I. Dubinko (NSC Kharkov Institute of Physics and Technology, Kharkov 61108, Ukraine), S. Y. Hu, Y. L. Li, C. H. Henager and R. J. Kurtz (Pacific Northwest National Laboratory)

This is the Extended Abstract of a manuscript submitted for publication in Philosophical Magazine & Philosophical Magazine Letters [1].

The preferential attraction to sinks of self-interstitial atoms (SIAs) over that of vacancies is the fundamental cause of the void swelling, and ultimately the evolution of microstructure in irradiated material. This preferential attraction is described mathematically in terms of bias factors that can be evaluated by solving diffusion equations for point defects (PDs) with drift terms included. The dislocation bias deduced from theory can be on the order of 0.5-1 (increasing with increasing sink strength) in a marked contrast with phenomenological values of 0.01-0.005 assumed in the early rate theory models in attempt to explain experimentally observed swelling rates [2]. Later this became one of the reasons that prompted the development of models based on production bias (PBM) as the main driver for swelling, which assumed that the dislocation bias was much lower than that predicted by theory. However, the PBM in its present form fails to account for important and common observations: the indefinite void growth often observed under cascade irradiation and the swelling saturation observed under high dose irradiation and in void lattices.

It is known that not all the energy of the primary knock-on atom (PKA) is spent for the production of stable defects. A considerable part of the PKA energy is spent for production of unstable Frenkel pairs (UFPs) [3, 4] and mobile lattice vibrations, such as focusons [5-8] and quodons [9-13], which can interact with extended defects and produce Schottky defects in their vicinity. A Schottky defect is a single vacancy or self-interstitial atom, or a small defect cluster, which can be emitted from extended defects (EDs) such as voids, dislocations and grain boundaries [14-20], and which does not require a counterpart of opposite sign in contrast to the bulk production of Frenkel pairs, in which the total numbers of vacancies and self-interstitial atoms *must be equal*. Another important difference between Frenkel and Schottky defects is the energy required for their formation. It is much lower for Schottky defects, especially for vacancies. It is well known that the Frenkel pair formation energy, $E_{FP}^{f,th}$, in metals under thermal equilibrium (when the crystal has enough time to adjust itself to the minimum free energy structure) is given by the sum of the SIA and vacancy formation energies, the former being is several times higher than the latter (e.g. in Cu, $E_{FP}^{f,th}$=3 eV, $E_{v}^{f,th}$=1.3 eV, $E_{FP}^{f,th} = E_{i}^{f,th} + E_{v}^{f,th}$=4.3 eV [21]). At the same time, the *ballistic* displacement energy required for stable Frenkel pair formation, $E_d \approx 25$ eV, is much higher than the thermal value, and so it can exceed the energy of vacancy production by an order of magnitude, which should result in the biased formation of vacancies as compared to SIAs near the ED surfaces under irradiation. This biased PD formation may be called *the radiation-induced emission* of vacancies from EDs, since the number of vacancies formed at the ED surface exactly equals the number of vacancies escaped from the ED. As a result, the number of atoms in the ED immediately decreases or increases depending on the ED type. For example, the number of vacancies in a void or a vacancy loop will be *decreased* while the number of atoms in a SIA loop will be *increased* in response to this ballistic vacancy emission.

In this paper we modify the Frenkel pair 3-D diffusion model (FP3DM) with account of the radiation-induced vacancy emission from EDs and production bias. The swelling rate taking into account the absorption, emission and production biases can be expressed as [1]:

$$\frac{dS}{dt} = D_i \bar{c}_i \frac{k_{vv}^2 k_{dv}^2}{k_v^2} B_{net}\,,\ B_{net} \equiv B_{ab} - B_{em} + B_p\,, \tag{1}$$

$$B_{ab} \equiv \frac{Z_v^V Z_i^d - Z_i^V Z_v^d}{Z_v^d} + \frac{Z_v^V k_{Gi}^2 - Z_i^V k_{Gv}^2}{Z_v^d \rho_d}\,, \tag{2}$$

$$B_{em} \equiv \frac{k_v^2}{k_d^2} Z_v^V \frac{\left(D_v \bar{c}_v^{eq} - D_v c_v^{eq,V}\right)}{D_i \bar{c}_i}, \tag{3}$$

$$B_p \equiv \frac{\varepsilon_i k_{eff} K}{k_{dv}^2 D_i \bar{c}_i}\left(1 - \frac{k_v^2 \bar{R}_V}{8 l_g}\right), \tag{4}$$

where B_{net} is the net bias. B_{ab}, B_{em} and B_p are absorption, emission and production biases, respectively. B_p contains two new parameters, namely, k_{eff} and ε_i.

To demonstrate the modified model, we first evaluate the swelling rate observed in austenitic steel [2] and pure Fe-Ni-Cr alloy [22]. Figure 1 shows the comparison of the swelling rates measured and predicted by FP3DM and modified FP3DM. It can be seen that the swelling rate predicted by the modified FP3DM agrees with experimental data both qualitatively and qualitatively.

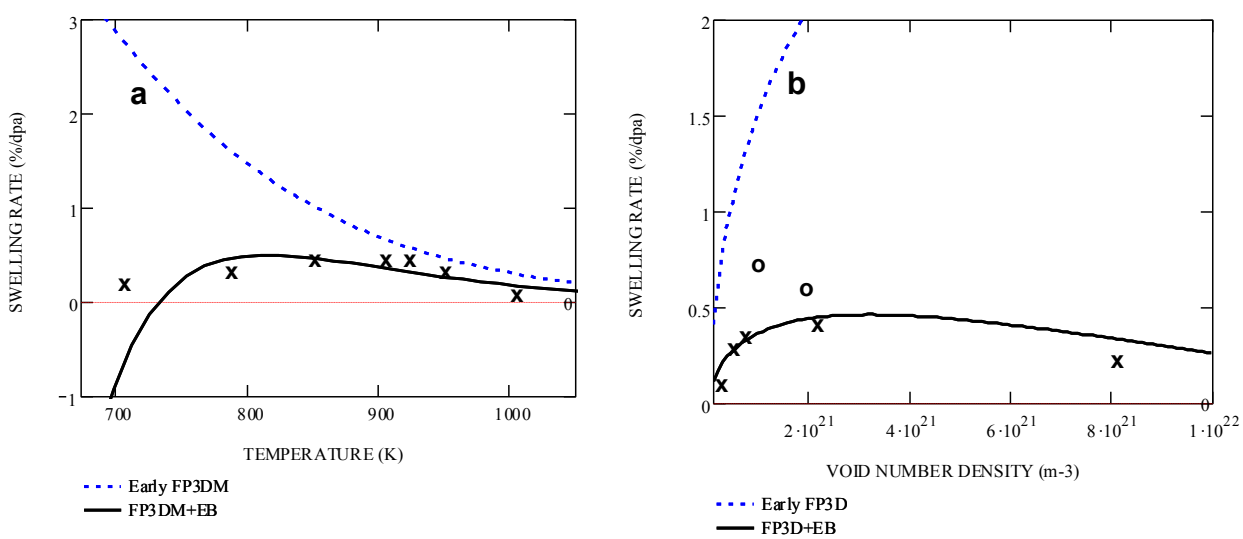

Figure 1. Dependence of the swelling rate predicted by FP3D and the modified (FP3D+EB) on temperature and the void number density vs. experimental data. Experimental data for austenitic steel [2] are shown in (a) and for a pure Fe-Ni-Cr alloy [22] in (b), where x and o represent residual gas and 10 appm pre-injected helium.

One can see in Figure 1 (a) that the modified theory predicts swelling rate to become negative below some threshold temperature, which could not be expected from early FP3D. The physical reason for this prediction is based on the decrease of the mean PD fluxes ~$D_i\bar{c}_i$ with decreasing temperature due to more intense bulk recombination of PDs (or denser sink structure). As a result, the emission bias, B_{em}, which is inversely proportional to $D_i\bar{c}_i$, increases and prevails over the absorption bias, which should result in the void shrinkage. In order to test this prediction, one needs to produce voids at higher temperatures, and then continue irradiation at a lower temperature. Such experimental tests have been performed in several experimental setups [17-20, 23], one of which is described below. In experiments [17], nickel foils of 100 micron thickness have been irradiated with 1.2 MeV Cr ions at 873 K up to the total ion fluence of 10^{21} m^{-2}, which corresponded approximately to the irradiation dose of 25 displacements per atom (dpa) at the dose rate, $K = 7\times10^{-3}s^{-1}$. Examination of control samples in transmission electron microscope (TEM) has revealed formation of a high number density (~10^{21} m^3) of voids of 40-50 nm in diameter. The remaining foils have been irradiated subsequently up to the ion fluence of 10^{21} m^{-2} at two different temperatures, 798 K and 723 K, respectively. The resulting microstructure is shown in Figure 2, from which it is evident that the irradiation at lower temperatures has made the voids to <u>decrease in size</u>. The quantitative analysis of the void swelling confirms this conclusion: the void swelling has decreased by a factor of ~ 5 (Figure 3).

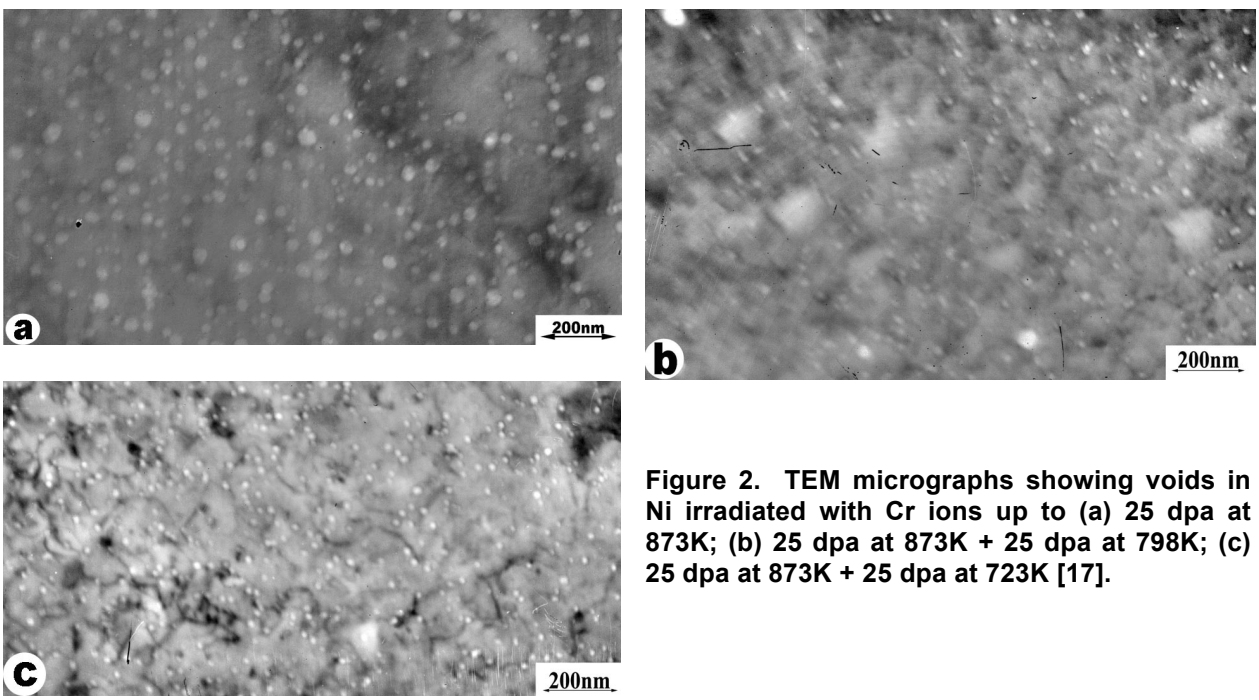

Figure 2. TEM micrographs showing voids in Ni irradiated with Cr ions up to (a) 25 dpa at 873K; (b) 25 dpa at 873K + 25 dpa at 798K; (c) 25 dpa at 873K + 25 dpa at 723K [17].

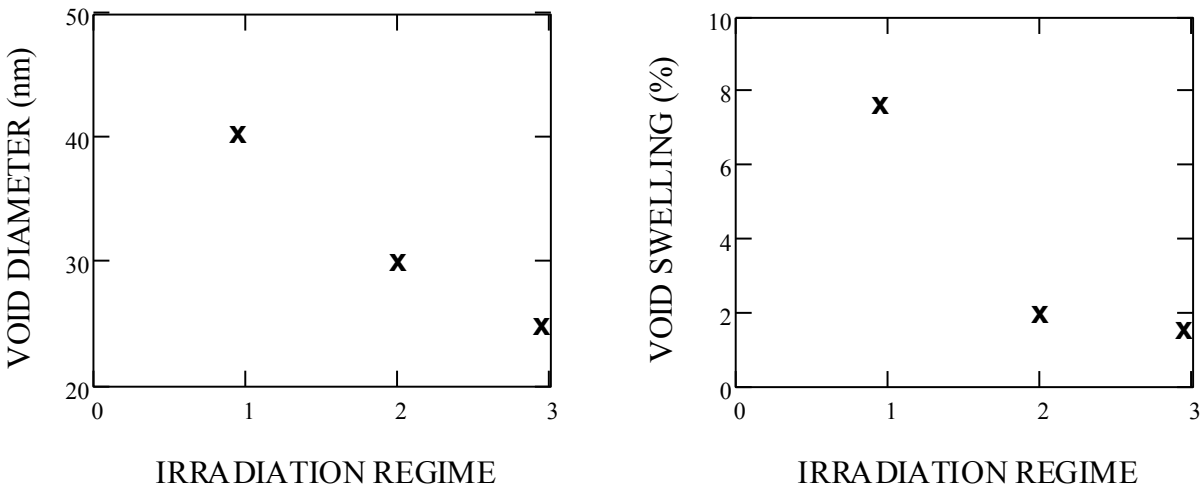

Figure 3. Mean void diameter and swelling in Ni irradiated with Cr ions up to 25 dpa at 873K (regime 1); 25 dpa at 873K + 25 dpa at 798K (regime 2) and 25 dpa at 873K + 25 dpa at 723K (regime 3). The size of symbols "x" corresponds to the mean error in void measurements [17].

Comparison of the PBM, FP3DM+EB and FP3DM+EB+PB models with experimental data [17] is shown in Figure 4. In the PBM their maximum values are estimated as follows [24, 25]: k_{eff} =0.1, ε_i = 0.2.

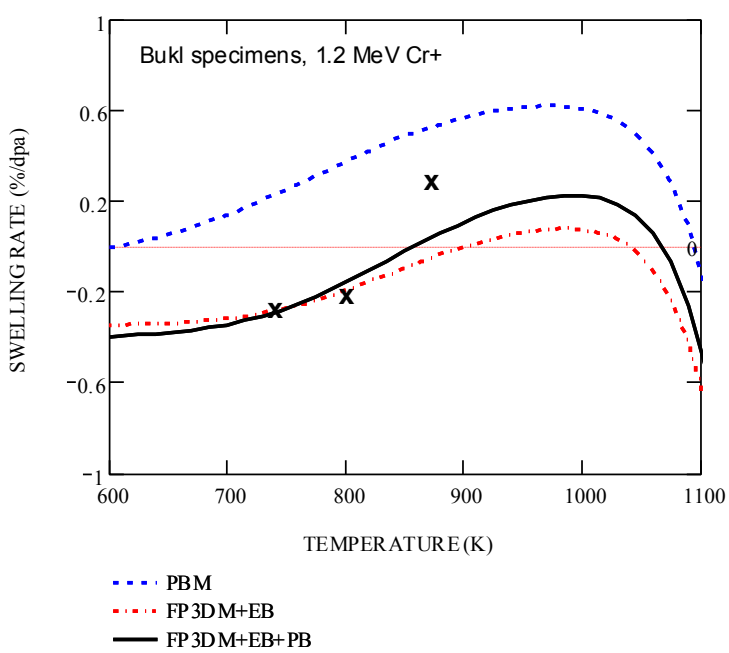

Figure 4. Temperature dependence of swelling rate in nickel bulk samples irradiated with 1.2 MeV Cr ions vs. experimental data [9]:

$K = 7\times10^{-3}$ dpa/s,
$\bar{R}_V = 20$ nm,
$N_V = 10^{21}\text{m}^{-3}$,
$\rho_d = 2\times10^{14}\text{m}^{-2}$.
$I_Q^0 = 5\times10^3 b$,
$\Delta E_d = 0.5$ eV,
$k_{eff} = 0.1$, $\varepsilon_i = 0.2$.

Other material parameters presented in Table 1.

One can see that the PBM cannot describe the observed low temperature reduction in swelling even qualitatively. On the other hand, the FP3DM modified with emission bias along (FP3DM+EB) can explain the swelling reduction but predicts a somewhat lower swelling rate at elevated temperatures and a lower threshold temperature than observed. The best agreement with experiment shows the model FP3DM+EB+PB that takes into account all three constituents of swelling, based on the absorption, emission and production biases.

In [1] we present new experimental results on sub-threshold electron irradiation, where the emission bias appears to be the only driver for microstructure evolution.

OUTLOOK

In order to forecast the behavior of nuclear materials in real radiation environment, the generation rate and propagation range of quodons should be modeled in different crystal structures as functions of defect concentrations and types. These factors seem to be of a primary technological importance since they offer a new insight on design of radiation-resistant materials.

Table 1. Material parameters used in calculations.

Parameter	Value
Atomic spacing, b, m	$3.23\times10P^{-10}$
Atomic volume of the host lattice, ω, m^{-3}	$2.36\times10P^{-29}$
Matrix shear modulus, μ, GPa	95 (Ni), 55 (Cu)
Interstitial dilatation volume, Ω_i	1.2ω, 0.6ω
Vacancy dilatation volume, Ω_v	-0.6ω
Bulk recombination rate constant, β_r, m^{-2}	10^{21}
Displacement energy, E_d, eV	30
Cascade efficiency for the stable PF production, k_{eff}	0.1
Fraction of point defects in the in-cascade clusters, $\varepsilon_{i,v}$	0.2
Maximum focuson energy, E_F, eV	30
Migration energy of vacancies, E_{vm}, eV	1.1 (Ni), 0.98 (Cu)
Pre-exponent factor, D_v^0	10^{-5}
Thermal vacancy formation energy at a free surface, E_{vf}^{th}, eV	1.8 (Ni), 1.3 (Cu)
Thermal vacancy formation energy at a dislocation, E_{vf}^{th}, eV	$E_v^{d,th} = E_v^{S,th} + \gamma_{SF} b^2$.
Stocking fault energy, γ_{SF}, J/m^2	0.1
Ballistic vacancy formation energy at a free surface	$E_v^0 = E_{vf}^{th} + E_{vm}$
Ballistic vacancy formation energy at the surface of a cavity with radius R_C and gas pressure P_g	$E_v^0 + \left(P_g - 2\gamma/R_C\right)\omega$
Surface energy, γ, J/m^2	2
Ballistic–thermal vacancy formation energy difference at dislocations, E_v^d, eV	0.5
Focuson propagation range, l_F, b	10
Quodon propagation range, l_Q, b	5×10^3

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