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Letter to the Editor

Point-defects in irradiated UO_2

1. Introduction

An integral component of fission-gas behavior theories is the modeling of the point defects (vacancies and interstitials) in the fuel during irradiation. These are calculated by balancing the rate of production by fission to the rates of removal at a variety of extended defects (e.g., bubbles, dislocations, grain boundaries) and the rate of recombination. There exists a very extensive literature on this phenomenon in monatomic solids, mainly metals and alloys, in connection with the processes of void formation, irradiation creep and irradiation growth. The point-defect models for polyatomic solids such as UO_2 often adopt the metals theory en bloc, without accounting for either the two-components in the solid or the fact that they are ionized.

1.1. Post-irradiation

Analyses of post-irradiation point-defect concentrations in UO_2 take into account equilibrium between all four ions. The point-defects are sustained by equilibrium processes, namely Frenkel pairs and Schottky defects. Typical of the models that invoke point-defect equilibria are those proposed by Griesmeyer and Ghoniem [1] and by MacInnes and Winter [2]. The four point-defects are uranium interstitials and vacancies and oxygen interstitials and vacancies. Their concentrations are related by the equilibrium constants for the Frenkel equilibria:

$$x_{VO}x_{IO} = K_{FO} \quad \text{and} \quad x_{VU}x_{IU} = K_{FU}$$

and the Schottky equilibrium:

$$x_{VU}x_{VO}^2 = K_S$$

where $x_{i,j}$ represents the fractional occupation of element j on site-type i.

Prior irradiation is taken into account as a cation site fraction of the fission products that have replaced some of the fissioned uranium. Even though there are two fission products per vanished uranium, not enough of them are soluble in UO_2 to fill all of the vacated cation sites. As a result, the number of cation lattice sites decreases and the oxygen ions released from the disappearing anion lattice sites move to sites on the anion interstitial lattice. For each interstitial anion, electrical neutrality requires that two cations be oxidized from U^{4+} to U^{5+}. The higher-valence uranium constitutes another type of point-defect that must be considered in the analysis of the imperfect crystal.

1.2. During irradiation

In the in-pile case, the point-defects exist in a medium with very high production rates due to collisions of fission fragments with lattice ions. In treating UO_2 under irradiation, the equilibrium point-defect concentrations represented by the Frenkel and Schottky equilibria are many orders of magnitude smaller than those sustained by fission fragment collisions with lattice ions. Instead of characterizing the bulk population, these equilibria are relegated to providing fixed point-defect concentrations at the surface of the extended defects.

2. Damage cascade

Each collision of a fission fragment with a lattice atom creates a primary knock on atom (PKA) which loses energy by collisions with neighboring atoms, thereby creating a "cascade" of vacancies and interstitials of both O and U.[1] Most of these recombine during the subsequent stage when the damage explosion of the cascade contracts, which is called cascade cooldown. The small fraction of the point-defects that survive recombination are partitioned between clusters of V_s and I_s and single point-defects, which are free to migrate in the bulk solid. Similar post-cascade debris is produced in metals exposed to high-energy neutrons.

3. Point-defects in UO_2

All models of irradiated UO_2 in the literature treat only one of the two ionic species, invariably the uranium ion. The oxygen ion is simply ignored, and the analysis proceeds as if the solid were a monatomic metal. In most papers, no justification for neglecting oxygen point-defects is offered; occasionally the single component treated is not even identified. When the failure to consider anion point-defects is acknowledged, the justification advanced is that the mobilities of the cation defects are very much smaller than those of the anion. The underlying picture is that of two anions tagging along wherever a cation moves. Treatments of this type are presented in Refs. [1,3–6].

Irradiation in a ceramic such as UO_2 creates four types of structural point-defects: vacancies in the cation and anion sublattices and interstitials of the anion (O^{2-}) and the cations. The latter are a combination of U^{4+}, U^{5+} and fission products (fp) with an average valence denoted by V_{fp}.

In the following, both anion and cation point-defects are included in an analysis of UO_2 undergoing irradiation. Restrictions related to electrical neutrality and the explicitly two-component solid result in some interesting conclusions.

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[1] The terms PKA and cascade are used interchangeably.

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4. Bulk characterization

To simplify this problem yet retain its essential features, the following assumptions are made:

1. The initial fuel is $UO_{2.00}$.
2. No Pu is produced during irradiation.
3. The soluble fission products exactly replace the uranium lost by fission in the cation sublattice. This implies that the sum of the fission yields of these fps is unity. In this picture, fission is equivalent to reduction of the U valence from 4+ to the yield-weighted average of the fission products. The following table shows the yields and the oxidation states of the fps.

Soluble fp	Yield	Valence
RE	0.53	3+
Zr	0.30	4+
Ba, Sr	0.15	2+
Mo	0.02	4+

In making the above yield assignments, formation of ternary oxides (molybdates, uranates, zirconates) has been neglected. Although Sr^{2+} is moderately soluble in UO_2, Ba^{2+} is much less so. However, this distinction is ignored and both of the these metal ions are assumed to dissolve completely in the fuel. Molybdenum is quite insoluble in UO_2, but it has been assigned a small solubility in order to bring the sum of the soluble-fp yields to unity in accord with Assumption No. 3 above.

From the above table, the average valence of the soluble fission products is:

$$V_{\mathrm{fp}} = \sum Y_i V_i = 3.2 \tag{1}$$

where Y_i and V_i are the nuclear yield and oxidation state, respectively, of fp i

The burnup β is defined as the number of fissions per unit volume, or:

$$\beta = \dot{F}t \tag{2}$$

where $\dot{F}$ is the fission rate in units of fissions per unit time per unit volume.

The inability of the soluble fission products to bind all of the oxygen released when uranium fissions (because $V_{\mathrm{fp}} < 4$) requires oxidation of some of the original U^{4+} to U^{5+}.

5. Point-defect behavior

In the following equations, the concentration C is defined as the number of a species per unit volume.

$C^o_{\mathrm{Scat}}, C^o_{\mathrm{San}}$ = cation and anion lattice sites.
C_{U4}, C_{U5} = U^{4+} and U^{5+} in the cation sublattice.
C_{fp} = soluble fps in the cation sublattice.
$C_{\mathrm{cat}} = C_{\mathrm{U4}} + C_{\mathrm{U5}} + C_{\mathrm{fp}}$ = all species in the cation sublattice.
C_{Vcat}, C_{Van} = vacant sites in the cation and anion sublattices.
$C_{\mathrm{Icat}} = C_{\mathrm{IU4}} + C_{\mathrm{IU5}} + C_{\mathrm{Ifp}}$ = total interstitial cations.
C_{O} = O^{2-} in the anion sublattice.
$C_{\mathrm{Ian}} = C_{\mathrm{IO}}$ = interstitial anion.

For the fluorite structure:

$$2C^o_{\mathrm{Scat}} = C^o_{\mathrm{San}} \tag{3}$$

where $C^o_{\mathrm{Scat}} = 1/\Omega$ and Ω is the volume of a UO_2 unit in uranium dioxide. All sites are completely filled prior to irradiation.

Element conservation during irradiation provides the following:

$$\text{uranium}: \quad C^o_{\mathrm{Scat}} = C_{\mathrm{U4}} + C_{\mathrm{U5}} + C_{\mathrm{fp}} + C_{\mathrm{IU4}} + C_{\mathrm{IU5}} + C_{\mathrm{Ifp}} \tag{4a}$$

$$\text{oxygen}: \quad C^o_{\mathrm{San}} = C_{\mathrm{O}} + C_{\mathrm{IO}} \tag{4b}$$

Irradiation is assumed to create interstitials of the cationic species in the same ratio as their concentrations in the cation sublattice:

$$f_{\mathrm{Icat}} = \frac{C_{\mathrm{IU4}}}{C_{\mathrm{U4}}} = \frac{C_{\mathrm{IU5}}}{C_{\mathrm{U5}}} = \frac{C_{\mathrm{Ifp}}}{C_{\mathrm{fp}}} = \frac{C_{\mathrm{Icat}}}{C_{\mathrm{cat}}} \tag{5}$$

The burnup defined by Eq. (1) is related to the fission product concentrations by:

$$\beta = C_{\mathrm{fp}} + C_{\mathrm{Ifp}} = (1 + f_{\mathrm{Icat}})C_{\mathrm{fp}} \tag{6}$$

Electrical neutrality during irradiation is a feature unique to ceramics but is not a consideration during irradiation of metals. For the present system, it is:

$$4C_{\mathrm{U4}} + 5C_{\mathrm{U5}} + V_{\mathrm{fp}}C_{\mathrm{fp}} + 4C_{\mathrm{IU4}} + 5C_{\mathrm{IU5}} + V_{\mathrm{fp}}C_{\mathrm{Ifp}} = 2C_{\mathrm{O}} + 2C_{\mathrm{IO}}$$

This equation can be simplified by adding 4 × Eq. (4a), subtracting 2 × Eq. (4b) and making use of Eqs. (3), (5), and (6). The electrical-neutrality condition reduces to:

$$(1 + f_{\mathrm{Icat}})C_{\mathrm{U5}} = (4 - V_{\mathrm{fp}})\beta \tag{7}$$

In the original fuel, all C^o_{Scat} cation sites are filled with U. After burnup β, the concentration of remaining total uranium is $C^o_{\mathrm{Scat}} - \beta$. The fraction of total U as U^{5+}, irrespective of location, is:

$$g_5 = \frac{(1 + f_{\mathrm{Icat}})C_{\mathrm{U5}}}{C^o_{\mathrm{Scat}} - \beta} \tag{8}$$

Combining Eqs. (7) and (8):

$$g_5 = \left(\frac{4 - V_{\mathrm{fp}}}{C^o_{\mathrm{Scat}} - \beta}\right)\beta = (4 - V_{\mathrm{fp}})\frac{\mathrm{FIMA}}{1 - \mathrm{FIMA}} \tag{9}$$

In the second form of this equation, burnup is expressed as FIMA (fissions per initial metal atom):

$$\mathrm{FIMA} = \beta / C^o_{\mathrm{Scat}} \tag{10}$$

Eq. (9) shows that $g_5 = 1$ at a burnup FIMA* given by:

$$\mathrm{FIMA}^* = (5 - V_{\mathrm{fp}})^{-1} \tag{11}$$

For $V_{\mathrm{fp}} = 3.2$, FIMA* = 0.56. At FIMA > FIMA*, the U^{5+} is oxidized to U^{6+}.

6. Point-defect balances

The point-defect concentrations, C_{Icat} and C_{Ian} for interstitials and C_{Van} and C_{Vcat} for the vacancies, are determined from point-defect balances. To illustrate the important effects of irradiation, the relevant microstructure and processes are simplified as follows:

1. Irradiation produces only point-defects; clusters (V and I loops) are not included.
2. V–I recombination is neglected (except during cascade cooldown).
3. The only point-defect sinks are cavities (bubbles) and dislocations. Their concentrations are specified.
4. The equilibrium point-defect concentrations in the solid adjacent to these sinks (from the Frenkel and Schottky equilibria) are assumed to be small compared to the bulk concentrations, and are neglected.
5. Except for providing the cavities (bubbles), the behavior of the fission-gas is not considered

6. Quasi-steady state is assumed.
7. The diffusivities of cation vacancies and interstitials are the averages of U^{4+}, U^{5+} and fps.
8. Radiation enhancement of the diffusivities due to mixing in the fission spike is not included.
9. Re-solution of vacancies from the cavities is not included
10. Oxygen loss by reaction with the cladding is neglected.

Subject to the above restrictions, the four point-defects balances are:

$$\text{cation vacancies}: \quad G_{\text{cat}}\dot{F} = (Z_{\text{CVcat}}4\pi R_{\text{C}}N_{\text{C}} + Z_{\text{dVcat}}\rho)D_{\text{Vcat}}C_{\text{Vcat}} \tag{12}$$

$$\text{cation interstitials}: \quad G_{\text{cat}}\dot{F} = (Z_{\text{CIcat}}4\pi R_{\text{C}}N_{\text{C}} + Z_{\text{dIcat}}\rho)D_{\text{Icat}}C_{\text{Icat}} \tag{13}$$

$$\text{anion vacancies}: \quad G_{\text{an}}\dot{F} = (Z_{\text{CVan}}4\pi R_{\text{C}}N_{\text{C}} + Z_{\text{dVan}}\rho)D_{\text{Van}}C_{\text{Van}} \tag{14}$$

$$\text{anion interstitials}: \quad G_{\text{an}}\dot{F} = (Z_{\text{CIan}}4\pi R_{\text{C}}N_{\text{C}} + Z_{\text{dIan}}\rho)D_{\text{Ian}}C_{\text{Ian}} \tag{15}$$

where G_{cat} is the number of cation V–I pairs per fission (after recombination during cascade cooldown), G_{an} is number of anion V–I pairs per fission (after recombination during cascade cooldown), R_{C} is cavity radius, N_{C} is cavity (bubble) number density (m^{-3}), ρ is network dislocation density (m^{-2}) and $D_{j\text{cat}}$, $D_{j\text{an}}$ are the diffusivities of point-defect j (V or I) on the cation and anion sublattices, respectively (m^2/s)

Z_{kjm} is a bias factor. The subscripts kjm denote:

- sink (extended defect) of type k (C for cavity or d for dislocation);
- point-defect j (V or I);
- sublattice m (cat or an).

The diffusion coefficients of the point-defects in UO_2 are not very well known. Matzke [11–13] gives the following for uranium:

$$D_{\text{Vcat}} = 10^{-7}\exp(-28,000/T)$$
$$D_{\text{Icat}} = 7 \times 10^{-7}\exp(-22,000/T) \quad \text{m}^2/\text{s}$$

The corresponding oxygen point-defect diffusivities deduced by Breitung [14] are[2]:

$$D_{\text{Van}} = 1.4 \times 10^{-4}\exp(-32,000/T)$$
$$D_{\text{Ian}} = 1.3 \times 10^{-6}\exp(-12,000/T) \quad \text{m}^2/\text{s}$$

However, radiation-induced increases of the diffusion coefficients are not accounted for in the above formulas.

7. Production of point-defects by fission

In UO_2, the numbers of point-defects produced per fission are different for the anions and the cations. In an early study, Soullard and Alamo [7] give 2.7×10^4 cations and 7.3×10^4 anions. Matzke [8] gives 1.5×10^4 Frenkel pairs (i.e., a V and an I) per fission, but does not distinguish between O and U defects. These figures do not account for recombination within the cascade, which is a very important feature of metal irradiation. Very likely the above numbers should be reduced by about an order of magnitude to give an accurate estimate of the production of point-defects that are free to migrate to sinks in the solid (G_{an} and G_{cat}). However, the important point is that the two values given by Soullard and Alamo [7] differ by nearly a factor of three. In numerical simulations of 1–80 keV PKAs in UO_2, Van Brutzel and Crocombette [9] found that the ratio of O/U Frenkel pairs was nearly exactly two. However, the number of Frenkel pairs varied as the 0.85 power of the energy, not linearly as expected from the NRT theory [10].

8. Bias factors

Significant differences between irradiated ceramics and irradiated metals are the bias factors. In metals where V and I absorption by the extended defects is diffusion-controlled, cavities are neutral sinks in the sense that $Z_{\text{CV}} = Z_{\text{CI}}$. In metals, the bias factor of dislocations for vacancies is $Z_{\text{dV}} = 1$, but for interstitials, $Z_{\text{dI}} > 1$. In this notation, Z_{CV} means the bias factor for vacancy absorption by cavities, Z_{dI} denotes the bias factor of interstitial absorption by dislocations. Z_{CI} and Z_{dV} are obvious variations.

In a ceramic such as UO_2, the bias factors are more complex than those for metals. For example, Likhanskii and Zborovskii [5] have performed a detailed stress analysis of the interaction of U vacancies with edge dislocations in UO_2 which indicated a departure of the concentration profile from that calculated with no interaction. In particular, they found that the cation V concentration near the core of the dislocation has a local maximum which reduces the bias factor and accelerates growth of intragranular bubbles. However, electrostatic effects were not considered.

Because the point-defects are electrically-charged ions, the combined mechanical and electrostatic interactions of the V_s and I_s with cavities are not the same and also different from V and I interactions with dislocations. In addition, the fluxes of point-defects to the sinks are restricted by an electrical-neutrality condition and a composition condition, as described below.

Based on these arguments, eight different bias factors are included in the analysis. If the eight bias factors are known, Eqs. (12)–(15) are readily solved for the four diffusivity-concentration products. However, the cation diffusivities are undoubtedly functions of burnup (FIMA) and U^{5+} fraction (g_5).

9. Special restrictions in ceramics

In ceramics, there are two conditions relating point-defect fluxes to composition and electrical neutrality that are not required in analysis of point-defect behavior in irradiated metals.

9.1. The ratio of the net fluxes of anions (O) to cations (U + fp) towards the sinks equals the bulk ratio

For cavities this condition is expressed by:

$$\frac{Z_{\text{CIan}}D_{\text{Ian}}C_{\text{Ian}} - Z_{\text{CVan}}D_{\text{Van}}C_{\text{Van}}}{Z_{\text{CIcat}}D_{\text{Icat}}C_{\text{Icat}} - Z_{\text{CVcat}}D_{\text{Vcat}}C_{\text{Vcat}}} = \left(\frac{\text{an}}{\text{cat}}\right)_{\text{bulk}} = 2 \tag{16a}$$

The vacancy fluxes are equivalent to atom fluxes in the opposite direction because atoms move away from the sink as vacancies move towards it. Eq. (16a) assures that the solid composition adjacent to the cavity is the same as that in the bulk solid. It can be shown that this condition is also equivalent to the requirement that the vacancies that constitute the cavities consist of one cation vacancy and two anion vacancies (i.e., a neutral trivacancy).

Eq. (16a) does not imply that the O/U ratio in the immediate vicinity of an extended defect cannot differ from the bulk value. Rather, it means that once a local O/U ratio is established near a sink, the continuing fluxes of point-defects must reflect the stoichiometry of the bulk. Otherwise, macroscopic deviations from stoichiometry would result in a continual buildup of the free-energy of the solid.

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[2] The activation energy of D_{Van} does not agree with the value given by Matzke [11–13].

The corresponding equation for dislocations is:

$$\frac{Z_{\mathrm{dIan}}D_{\mathrm{Ian}}C_{\mathrm{Ian}} - Z_{\mathrm{dVan}}D_{\mathrm{Van}}C_{\mathrm{Van}}}{Z_{\mathrm{dIcat}}D_{\mathrm{Icat}}C_{\mathrm{Icat}} - Z_{\mathrm{dVcat}}D_{\mathrm{Vcat}}C_{\mathrm{Vcat}}} = \left(\frac{\mathrm{an}}{\mathrm{cat}}\right)_{\mathrm{bulk}} = 2 \tag{16b}$$

9.2. *The net-charge flux towards the sinks is zero*

The flux of point-defect j in sublattice m towards sink k is denoted by:

$$J_{kjm} \propto Z_{kjm}D_{jm}C_{jm} \tag{17}$$

The net negative-charge flux to the sinks is the O interstitial flux less the O vacancy flux.

net $(-)$ flux to sink $k = 2(J_{k\mathrm{Ian}} - J_{k\mathrm{Van}})$

The factor of two accounts for the valence of the anion.

Free electrons have been neglected because UO_2 is an electrical insulator. A net charge around a sink may exist, but it cannot continue to grow as charges move towards it.

In an analogous fashion, cation fluxes deliver positive charges to the sinks:

net $(+)$ flux to sink

$$k = (J_{k\mathrm{Icat}} - J_{k\mathrm{Vcat}})\left[5\left(\frac{C_{\mathrm{U}}}{C_{\mathrm{cat}}}\right)g_5 + 4\left(\frac{C_{\mathrm{U}}}{C_{\mathrm{cat}}}\right)(1-g_5) + V_{\mathrm{fp}}\left(\frac{C_{\mathrm{fp}}}{C_{\mathrm{cat}}}\right)\right]$$

where the three terms in the brackets represent the contributions of U^{5+}, U^{4+} and fps. The first two parenthetical terms are the fractions of uranium ions on the cation sublattice. The last term is the component due to the fission product ions. Holes are not treated because they are attached to cations as U^{5+}.

Since

$(C_{\mathrm{U}}/C_{\mathrm{cat}}) = 1 - \mathrm{FIMA}$ and $(C_{\mathrm{fp}}/C_{\mathrm{cat}}) = \mathrm{FIMA}$,

net $(+)$ flux to sink $k = (j_{k\mathrm{Icat}} - j_{k\mathrm{Vcat}})[(4+g_5)(1-\mathrm{FIMA}) + V_{\mathrm{fp}} \times \mathrm{FIMA}]$

Using the flux notation of Eq. (17), equating the (+) and (−) fluxes and taking g_5 from Eq. (9), the charge-flux balance towards cavities becomes:

$$\frac{Z_{\mathrm{CIan}}D_{\mathrm{Ian}}C_{\mathrm{Ian}} - Z_{\mathrm{CVan}}D_{\mathrm{Van}}C_{\mathrm{Van}}}{Z_{\mathrm{CIcat}}D_{\mathrm{Icat}}C_{\mathrm{Icat}} - Z_{\mathrm{CVcat}}D_{\mathrm{Vcat}}C_{\mathrm{Vcat}}} = 2$$

which is identical to Eq. (16a). The analogous equation for the charge-flux balance towards dislocations is the same as Eq. (16b). Eqs. (16a) and (16b) do not preclude localized gradients of the O/U ratio around the extended sinks, nor does Eq. (16b) prohibit establishment of a localized electric field gradient around the extended defects. They simply require that once established, the fluxes of species and the fluxes of charges do not change with time.

10. Conclusions

The four bulk point-defect concentrations are determined by six equations. The apparent over-determination of the mathematical system means that only six of the eight bias factors can be independently fixed. That is, there exist two relations between bias factors that involve the bulk point-defect concentrations.

The bias factors in ionic solids are not the simple types applicable to metals [15]. Rather, in ceramics, the bias factors depend on the electric field that is set up wherever concentration gradients exist. The electric field is characterized by a shape and strength, and these two features adjust so that Eqs. (12)–(15), (16a) and (16b) are simultaneously satisfied. The creation of electric fields in diffusion in ionic solids is found in oxidation of metals [16–18] and is believed to be responsible for non-parabolic oxidation kinetics during Zircaloy oxidation in air [19]. Electric-field effects in the oxide film are probably the origin of the initial "cubic" rate law during Zircaloy oxidation in PWR water.

The analysis here is grossly simplified in order to more clearly illustrate how the two-component, ionic ceramic UO_2 differs from monatomic metals. No attempt has been made to calculate the sink strengths, but there is evidence from metal oxidation studies that the electric-field effect is important. There is much work to be done.

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