Journal of Nuclear Materials 377 (2008) 406–408

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Letter to the Editor

Calculation of grain boundary gaseous swelling in UO_2

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ARTICLE INFO

Article history:
Received 16 October 2007

ABSTRACT

Swelling of UO_2 fuel due to intergranular fission gas bubbles is calculated as a function of irradiation time (or burnup) using an effective semi-analytical method. The model can be used concurrently to evaluate fission gas release and gaseous swelling during irradiation.

During reactor service the volume of UO_2 fuel increases continuously with *burnup* (megawatt-days of thermal output per kg uranium). The swelling is defined as the increase in volume by the fission products located in the fuel. The solid fission products are theoretically predicted to contribute to fuel swelling on the average by 0.032% per MWd(kgU)^{-1} [1]. The contribution of gaseous fission products to fuel swelling includes rare gases such as krypton and xenon in solid solution and the volume change arising from the formation of fission gas filled bubbles. For the gases in solid solution and the small intragranular gas bubbles, it is estimated that they furnish about 0.056% per MWd(kgU)^{-1} to matrix swelling rate [2]. The intergranular gas bubbles can make the largest contribution to volume change depending on temperature and their amount. Early studies [3] indicated large bubbles of diameter around few microns on grain faces and also along grain edges. At high exposures and temperatures the bubbles interlink forming a tunnel network which concurrently leads to gaseous swelling and gas release [4,5]. Fission gas swelling in UO_2 fuel due to intergranular gas (grain face) bubbles can be expressed by

$$\left(\frac{\Delta V}{V}\right)_{\mathrm{gb}} = \frac{4\pi r_{\mathrm{b}}^3\omega}{3} n_{\mathrm{b}}, \tag{1}$$

where $\Delta V/V$ is the relative increase in volume, r_b the bubble radius, ω a geometric factor accounting for the ellipsoidal shape of a bubble, n_b the number of intergranular bubbles per unit volume of grain, and we also denote the volume of an intergranular bubble by $V_b = 4\pi r_b^3\omega/3$. Here $\omega = 1 - 1.5\cos\theta + 0.5\cos^3\theta$ and θ is the dihedral angle between the bubble surface and grain boundary [6]. In writing Eq. (1), we tacitly assumed that all intergranular bubbles have equal size. Note that n_b can be related to the number of gas bubbles per unit area of grain face N_b via $n_b = 3N_b/2R$, where R is the radius of (spherical) grain.

Now following observations by Hargreaves and Collins [7], we suppose that before the occurrence of grain boundary gas saturation, n_b (or N_b) is fairly constant, so that swelling can be evaluated by calculating the concentration of gas atoms on the grain boundaries and the size of each bubble. If the gas in the bubble obeys the van der Waals law, we write

$$N = \frac{1}{k_{\mathrm{B}}T}(V_{\mathrm{b}} - bN)\left(P + \frac{aN^2}{V_{\mathrm{b}}^2}\right), \tag{2}$$

where N is the number of gas atoms in the intergranular bubble, P the pressure inside the bubble, k_B the Boltzmann constant, T the absolute temperature, and a and b are phenomenological constants. The number of intergranular gas atoms per unit volume of grain C_b is expressed by $C_b = N\,n_b$. For gases such as Xe and Kr, which are of interest here, the second term in the second parenthesis of Eq. (2) is relatively small and hence is neglected in the ensuing calculations.

The bubble gas pressure is given by the Laplace equation [8], namely $P = 2\gamma/r_b + P_{ext}$, where γ is the surface tension of the bubble and P_{ext} is any externally applied hydrostatic stress due to fuel rod gas pressure and/or to fuel-cladding contact pressure. Supposing gas in the bubble obeys the van der Waals equation of state, then the intergranular gas density is expressed as

$$C_{\mathrm{b}} = \frac{4\pi}{3} r_{\mathrm{b}}^3\omega n_{\mathrm{b}} \frac{P_{\mathrm{ext}} + 2\gamma/r_{\mathrm{b}}}{b(P_{\mathrm{ext}} + 2\gamma/r_{\mathrm{b}}) + k_{\mathrm{B}}T}. \tag{3}$$

When the gas atoms saturate the grain boundaries $C_b = C_{bs}$, where

$$C_{\mathrm{bs}} = \frac{4\pi}{3} r_{\mathrm{bs}}^3\omega n_{\mathrm{b}} \frac{P_{\mathrm{ext}} + 2\gamma/r_{\mathrm{bs}}}{b(P_{\mathrm{ext}} + 2\gamma/r_{\mathrm{bs}}) + k_{\mathrm{B}}T}, \tag{4}$$

and r_{bs} is the bubble radius at the onset of interlinkage (saturation). Knowing the values of r_{bs} and C_b, the latter from fission gas release model calculation and the former from observations, n_b and r_b can be determined from Eqs. (3) and (4), respectively, from which we write

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doi:10.1016/j.jnucmat.2008.03.011

$$\frac{C_b}{C_{bs}} = x^2\left(\frac{x+h}{1+h}\right)\left(\frac{1+(1+h)\vartheta}{1+(1+h/x)\vartheta}\right), \quad (5)$$

where we introduced parameters $x \equiv r_b/r_{bs}$, $h \equiv 2\gamma/r_{bs}P_{ext}$ and $\vartheta \equiv bP_{ext}/k_BT$. We re-express Eq. (5) in the form

$$\frac{x^3(x+h)}{x+(x+h)\vartheta} = \eta, \quad (6)$$

with $\eta \equiv (1+h)/[1+(1+h)\vartheta](C_b/C_{bs})$. Hence, knowing the value of η, then x and thereby r_b can be determined by solving Eq. (6) using a numerical scheme.

Now turning to the evaluation of gaseous swelling, we can substitute n_b in Eq. (1) from Eq. (3) to write

$$\left(\frac{\Delta V}{V}\right)_{gb} = \frac{3k_BT[1+\vartheta(1+h/x)]}{2P_{ext}(1+h/x)}\frac{\mathcal{N}}{R}, \quad (7)$$

where $\mathcal{N} = (2/3)RC_b$ is the number of gas atoms per unit area of the grain boundary. We note that when gas atoms saturate the grain boundaries, i.e., the intergranular bubbles interlink, $\mathcal{N} = \mathcal{N}_s \equiv (2/3)RC_{bs}$, where

$$\mathcal{N}_s = \frac{4r_{bs}\omega f_b P_{ext}(1+h)}{3k_BT\sin^2\theta[1+\vartheta(1+h)]}. \quad (8)$$

Hence, intergranular gaseous swelling saturates ($x = 1$) when

$$\left(\frac{\Delta V}{V}\right)_{gs} = \frac{2\omega f_b}{\sin^2\theta}\frac{r_{bs}}{R}, \quad (9)$$

where f_b is the fractional coverage of grain boundaries (by the bubbles) and ω was defined earlier. We may compare this result with that of Une [9] who simulated bubble density in UO_2 pellets by out-of-pile experiments. Une generated bubble swelling by sintering pellets in reducing atmosphere of H_2 and then annealing them in oxidizing atmosphere at temperatures of up to 2000 K. Thus, using the values $\theta \approx 56^\circ$, $r_{bs} = 1.0$ μm, $R = 7.5$ μm and $f_b = 1.0$ [9], we find $(\Delta V/V)_{gs} \approx 0.10$, which agrees with the observed and calculated value reported in ref. [9]. This result is also in agreement with the outcome of experiments conducted by Turnbull [10] on UO_2 pellets (grain size: 15 μm) irradiated at 2023 K up to about 4 $MWd(kgU)^{-1}$.

In Fig. 1 we have plotted $(\Delta V/V)_{gb}$ as a function of irradiation time at different temperatures up to the grain boundary saturation (for $T = 1700$ and $T = 1900$ K) using the aforementioned model with the intergranular gas density calculated according to the relations outlined in the Appendix. The input data to the model are presented in Tables 1 and 2, which have been used for calculations of fission gas release in earlier works [11,12]. We note that according to this input, Eq. (9) gives $(\Delta V/V)_{gs} = 0.0145$. The evolution of the intergranular gas bubble radius calculated through relation (6) is depicted in Fig. 2, while the evolution of the gas density on grain boundary normalized with the saturation value is displayed in Fig. 3. When $\mathcal{N}$ reaches $\mathcal{N}_s$ gas release would occur, which also would reduce the gaseous swelling and the mean gas bubble size according to the present theory.

Table 1
Parameters used for fission gas diffusion coefficient in uranium dioxide, $D = v_g D'/(v_g + g)$

Parameter	Unit	Definition
$D' = C_1 e^{-Q_1/k_BT} + C_2\dot{F}e^{-Q_2/k_BT} + C_3\dot{F}$	$m^2\,s^{-1}$	Diffusivity in trap free media
$v_g = 3.03\pi\ell\dot{F}(\bar{R}_b+\delta)^2$	s^{-1}	Intragranular gas bubble re-solution rate
$\bar{R}_b = 1.453\times10^{-10}\exp(1.023\times10^{-3}T)$	m	Intragranular bubble radius
$g = 4\pi\bar{R}_b C_b^t D'$	s^{-1}	Fission gas capture rate by intragranular gas bubble
$C_b^t = 1.52\times10^{27}/T - 3.3\times10^{23}$	m^{-3}	Total bubble density
$\ell = 6\times10^{-6}$	m	Fission fragment range
$\delta = 10^{-9}$	m	Damage radius of fission fragment
$\dot{F} = N_A\dot{F}_m$	$m^{-3}\,s^{-1}$	Fission density
$\dot{F}_m = 5.189\times10^{-14}q_v$	$mol\,m^{-3}\,s^{-1}$	Fission density
q_v	$W\,m^{-3}$	Power density
$\beta = 0.3\dot{F}_m$	$mol\,m^{-3}\,s^{-1}$	Fission gas production rate
$N_A = 6.022\times10^{23}$	mol^{-1}	Avogadro constant
$Q_1/k_B = 35247$	K	(Activation energy)/k_B
$Q_2/k_B = 13800$	K	(Activation energy)/k_B
$C_1 = 7.6\times10^{-10}$	–	–
$C_2 = 4.5\times10^{-35}$	–	–
$C_3 = 2.0\times10^{-40}$	–	–

Here, T is the absolute temperature, cf. [11,12] and references therein.

Table 2
Input data used in fission gas swelling computations for UO_2 fuel, cf. [11,12] and references therein

Parameter	Unit	Definition
$d_{pellet} = 8.480$	mm	Fuel pellet diameter
$\rho_T = 0.960$	–	Fraction of fuel density
$R = 7.5$	μm	Fuel grain radius
$P_{ext} = 1$	MPa	External pressure
$2\gamma/r_{bs} = 2.4$	MPa	Bubble surface tension to radius ratio
$\frac{4r_{bs}\omega f_b}{3\sin^2\theta} = 7.25\times10^{-8}$	m	Composite gas bubble parameter
$b = 5.16\times10^{-5}$	$m^3\,mol^{-1}$	van der Waals constant for Xe [14]
$v_b\lambda/\beta = 5.7\times10^{-8}$	$m^4\,mol^{-1}$	Ratio of re-solution rate to gas production rate
$q_l = 35$	$kW\,m^{-1}$	Linear power density

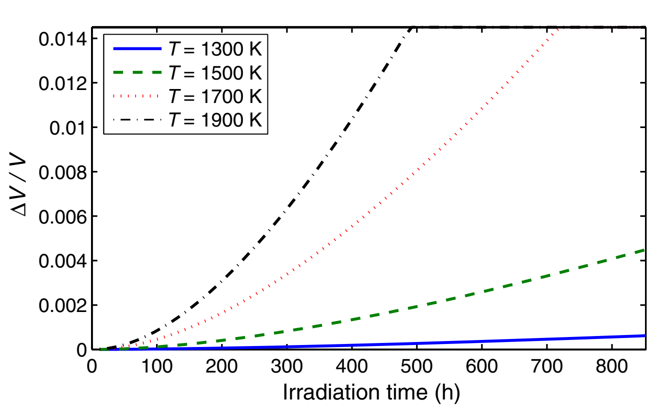

Fig. 1. Calculated fuel swelling due to intergranular gas bubbles (grain face bubbles) as a function of irradiation time.

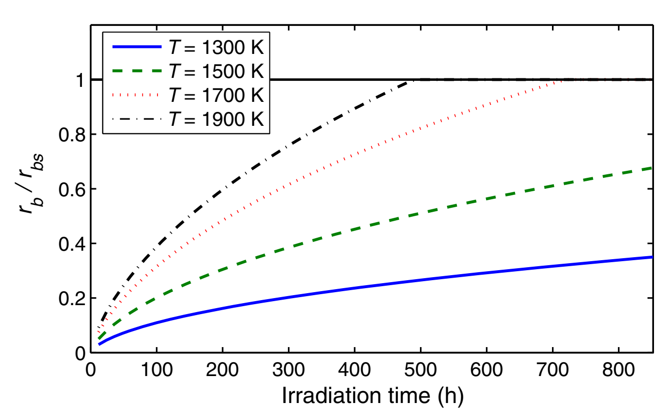

Fig. 2. Normalized intergranular bubble radius calculated as a function of irradiation time.

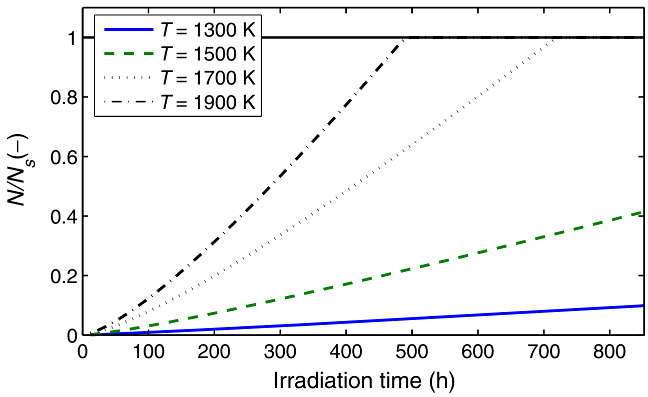

Fig. 3. Ratio of fission gas density within grain boundary to gas density at saturation calculated as a function irradiation time.

The evolution of grain boundary gas density, thereby the intergranular gas swelling, is a strong function of temperature. Our calculations, with the present theory and input parameter data, indicate that at 1900 K the grain boundary gets saturated after 500 h of irradiation ($\approx$1.4 MWd(kgU)$^{-1}$), whereas at 1300 K, it is saturated after 7716 h ($\approx$21.5 MWd(kgU)$^{-1}$).

Comparing our results with those obtained by using the intergranular swelling model of Spino et al. [2], who use a different approach and other parameters, we find that at 1300 K, $P_{\mathrm{ext}} \approx 0$ and burnup of about 6 MWd(kgU)$^{-1}$, our model gives $(\Delta V/V)_{\mathrm{gb}} \approx 0.3\%$, while theirs yields $(\Delta V/V)_{\mathrm{gb}} \approx 0.07\%$ for a grain size of 15 μm. Experiments to quantify the contribution of intergranular gaseous swelling at different temperatures are rare, therefore such experiments would be valuable for model evaluation. Composite parameters $\frac{2\gamma}{r_{\mathrm{bs}}}$ and $\frac{4 r_{\mathrm{bs}} \omega f_{\mathrm{b}}}{3 \sin^2 \theta}$ (Table 2), which are fuel microstructure entities, may need to be re-adjusted to describe intergranular gaseous swelling experiments under isothermal conditions. Our model presented here can readily be implemented in a fuel behaviour code to calculate fission gas release and gaseous swelling concurrently, where non-isothermal gaseous swelling can be evaluated.

Appendix A. Intergranular gas density

The number of gas atoms in intergranular bubbles per unit area of grain boundary at time t, $\mathcal{N}(t)$, which accounts for the effect of the irradiation-induced re-solution, is given in [13]. With a slight change of notation from [13], in terms of dimensionless parameters, we write

$$\mathcal{N}(\tau) = \frac{2\beta_{\mathrm{e}}}{k_1}\left[\tau + \frac{1}{k_2 k_3} - \frac{k_2 \mathrm{e}^{k_3^2\tau}\mathrm{erfc}(k_3\tau^{1/2}) + k_3 \mathrm{e}^{k_2^2\tau}\mathrm{erfc}(-k_2\tau^{1/2})}{k_2 k_3 (k_2 + k_3)}\right] + \mathcal{O}(\tau^{\infty}), \quad \text{for} \quad \tau < 1/\pi^2, \tag{A.1}$$

$$\mathcal{N}(\tau) = \frac{2\beta_{\mathrm{e}}}{3 + k_1}\left(\tau - \frac{1}{5(3 + k_1)}\right) + \sum_{m=1}^{\infty} \frac{4\beta_e \mathrm{e}^{-u_m^2\tau}}{u_m^2[u_m^2 + k_1(3 + k_1)]}, \quad \text{for} \quad \tau > 1/\pi^2, \tag{A.2}$$

where

$$k_2 = -\frac{k_1}{2} + \sqrt{\frac{k_1^2}{4} + k_1}, \tag{A.3}$$

$$k_3 = \frac{k_1}{2} + \sqrt{\frac{k_1^2}{4} + k_1}, \tag{A.4}$$

$$u_m = \arctan\left(\frac{k_1 u_m}{u_m^2 + k_1}\right) + m\pi, \tag{A.5}$$

and $k_1 = \nu_{\mathrm{b}}\lambda R/D$, $\beta_{\mathrm{e}} = \beta R^3/D$, $\tau = \int_0^t D(s)\mathrm{d}s/R^2 = Dt/R^2$. Here ν_{b} is the re-solution rate, λ the re-solution distance, D the gas diffusion coefficient, R the grain radius, and β the fission gas production rate per unit volume.

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