

Theory of Fission Gas Release During Grain Growth

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ABSTRACT

The paper provides a theoretical framework for calculation of fission gas release process by diffusion, irradiation-induced re-solution, grain boundary saturation, and grain growth in UO₂ fuel under time varying temperature loads. We suppose that UO₂ grains are spherical of equal size. The diffusion equation for gas atoms in the grain, which allows for the time variations of gas diffusion coefficient, gas production rate, and grain size, is formulated. The equation is subjected to a boundary condition, which accounts for grain boundary gas re-solution (to the grain) and grain boundary saturation prior to release. A kinetic equation for grain growth is coupled to the above boundary value problem. Analytical solutions to the aforementioned equations are obtained. The obtained equations are used to calculate fission gas release and grain growth as functions of temperature and time. Correlation between grain growth and gas release has been evaluated and the results are discussed in light of experimental data.

INTRODUCTION

In nuclear fuel noble gases xenon and krypton are generated during fission of uranium and plutonium isotopes. A fraction of these gaseous fission products is released into the free volume of fuel rod increasing the internal fuel rod pressure. Also the gas released into the fuel pellet-cladding gap degrades the thermal conductance of the gas in the gap causing higher fuel temperature and further enhancement of fission gas release. Fission gas release is generally considered to be potentially a life-limiting (burnup-limiting) phenomenon because of its consequence on fuel cladding integrity due to excessive rod internal pressure.

Fission gas release process in UO₂ fuel is considered to consist of two mechanisms: an athermal release and thermal release. The athermal release accounts for the contribution of release caused by *direct recoil* of fission fragments within a layer equal to the range of the fission fragments in the fuel ($\approx 10 \mu\text{m}$); and by a *knockout* mechanism, which is an elastic collision between fission fragments and fission product gas atoms in the fuel [1]. Athermal release is primarily fission rate dependent and in light water reactors it is almost a linear function of fuel burnup up to a burnup of around 40 MWd/kgU.

More specifically, the rate of the number of gas atoms released by athermal process can be written as: $dN_r / dt \approx N_g \dot{\phi}$ where N_g is the number of gas atoms generated per unit volume of fuel, $N_g = \dot{\phi} t y$, y is the fission yield, $\dot{\phi}$ is the fission rate and t is the irradiation time. The release fraction is $F_{ath} = N_r / N_g \sim \dot{\phi} t = C u$ where u is the fuel burnup [MWd/kgU] and C is a constant of proportionality. For example, Lorenz [2] based on the evaluation of athermal fission product gas release data, obtained from rods irradiated up to an average burnup of around 40 MWd/kgU, found that $C=8.5 \times 10^{-8}$. However, more recent data from rods operating at low powers indicate an enhancement in athermal fission gas release beyond burnup of around 45 MWd/kgU almost in an exponential manner. An engineering model to capture this phenomenon was suggested in reference [3].

The thermal release is highly temperature dependent and is driven by thermal processes in irradiated fuel. These include gas diffusion to grain boundaries, grain growth, grain boundary saturation and release. More specifically, in-reactor experiments performed on nuclear fuel, by ramping fuel rods to different power levels, indicate that at above a certain linear heat generation rate (LHGR) fission product gas release is enhanced by grain boundary sweeping. The increase in fuel temperature due to the rise in LHGR enhances volume diffusion of fission product gases to the grain boundaries of UO₂ fuel, and also may cause grain growth, which sweeps the gases to the grain boundaries. Upon saturation of grain boundaries with gases, release to the free volume of the fuel rod will occur.

In the last decades there have been many efforts to develop physically based theoretical frameworks for calculations of fission product gas release in nuclear fuel. From our vantage point these comprise seminal works of Booth and Kennedy [4] on intragranular diffusional release and Speight [5] which included the effect of gas precipitation, re-solution and grain boundary saturation and release. The problem of time varying parameters (diffusion coefficient and gas production rate) was analyzed among others by Mathews and Wood [6], and later by Forsberg and Massih [7] analytically. Lassman and Benk [8]

and Lassmann [9] have recently evaluated some of these methods. Moreover, Forsberg and Massih [10, 11] extended the time-varying conditions to include the effect of re-resolution and the intergranular gas release. A general theoretical method for calculation of the release of volatile fission gas products has been provided by Paraschiv, Paraschiv, and Grecu [12] which include the phenomena of grain growth, grain boundary bubble growth and re-resolution based on their earlier studies on the release of stable fission product gases from nuclear fuel.

In the present paper we attempt to provide a general method for calculation of thermal fission product gas release process by gas atom diffusion, irradiation-induced re-resolution, grain boundary saturation, and grain growth in UO_2 fuel under time varying temperature loads. We suppose that UO_2 grains are spherical of equal size, i.e. the so called the equivalent sphere model of Booth [4]. Then the diffusion equation for gas atoms in the grain, which allows for the time variations of gas diffusion coefficient, gas production rate, and grain size, is formulated. The equation is subjected to a boundary condition, which accounts for grain boundary gas accumulation, re-resolution (to the grain), and grain boundary saturation prior to release. A kinetic equation for grain growth is coupled to the above boundary value problem. The governing equations for this problem and some limited calculations were briefly presented in our earlier overview paper in 1994 [3]. Here analytical solutions to the aforementioned equations are presented. The obtained equations are used to calculate fission gas release and grain growth as functions of temperature and time.

The organization of this paper is as follows. In section 2 the governing equations for the problem under consideration, i.e. diffusion of gas atoms in a growing grain, are presented. In section 3 approximate analytical solutions for the equations are derived. The relations for calculation of fission gas release under small time steps are derived in section 4. Section 5 provides the results of calculations carried out with the present theory. We close the report by discussing the relevance of time constants for diffusion and grain growth on fission gas release, and a brief discussion on UO_2 fuel microstructure studies to find the relation between grain growth and fission gas release.

EQUATIONS FOR GAS DIFFUSION IN A GROWING SPHERE

We consider the equivalent sphere model for UO_2 material, meaning that the grains of the material are spherical of equal size [4, 5]. The fission product gases are produced at a rate $\beta(t)$ in a grain with radius $R(t)$. The gases migrate to grain boundaries by diffusion with a diffusion coefficient designated as $D(t)$. The gas atoms reaching the boundary precipitate into intergranular bubbles where we denote the number of gas atoms in bubbles per unit area of grain boundary at time t by $N(t)$ with the grain boundary re-resolution rate, $B(t)=b\lambda/2$ (b is the grain boundary re-resolution probability and $\lambda/2$ the re-resolution depth from the grain face). All the considered variables are time dependent.

The differential equation for concentration of gas atoms at position r in the grain at time t , $C(r, t)$ is given by the law of conservation of mass, viz.

$$\frac{\partial C(r, t)}{\partial t} = D(t)\Delta_r C(r, t) + \beta(t) \quad \text{for } 0 < r < R(t) \quad (1)$$

with

$$\Delta_r = \frac{\partial^2}{\partial r^2} + \frac{2}{r} \frac{\partial}{\partial r} \quad (2)$$

The boundary conditions imposed on $C(r, t)$ are

$$\frac{\partial}{\partial r} C(0, t) = 0 \quad (3)$$

and

$$C(R, t) = \frac{B(t)N(t)}{D(t)} \quad (4)$$

The initial condition is $C(r, 0) = 1$. Since the gas diffusion and grain growth can occur simultaneously, Eq. (1) can be transformed to the equivalent equation of the form

$$\frac{\partial \mathcal{C}(\chi, \tau)}{\partial \tau} = \Delta'_\chi \mathcal{C}(\chi, \tau) + \mathcal{P}(\tau) \quad (5)$$

where

$$\chi \equiv \frac{r}{R(t)}, \quad \tau \equiv \int_0^t \frac{D(s)}{R^2(s)} ds \quad (6)$$

$$\Delta'_\chi = \Delta_\chi + \frac{\mathcal{R}_\tau}{\mathcal{R}} \frac{\partial}{\partial \chi} \quad (7)$$

$$\mathcal{R}_\tau \equiv \frac{\partial \mathcal{R}}{\partial \tau} \quad (8)$$

$$\mathcal{P}(\tau(t)) \equiv \frac{R^2(t)}{D(t)} \beta(t) \quad (9)$$

and the transformed re-solution rate:

$$\mathcal{B}(\tau(t)) \equiv \frac{R(t)}{D(t)} B(t) \quad (10)$$

The boundary condition given by Eq. (3) is expressed as

$$\frac{\partial \mathcal{C}(0, \tau)}{\partial \chi} = 0 \quad (11)$$

At the grain boundary $r = R(t)$, i.e., $\chi = 1$. We note that Eq. (5) includes explicitly the rate of change of grain radius $\mathcal{R}_\tau = d\mathcal{R}/d\tau$, which is governed by the grain growth kinetic law. The total amount of gas $G(t)$ per unit volume in a grain of radius R is

$$\begin{aligned} G(t) &= \frac{2\pi R^2 N + \int_0^R 4\pi r^2 C(r, t) dr}{4\pi R^3 / 3} \\ &= \frac{3D(t)C(R, t)}{2R(t)B(t)} + \frac{3 \int_0^R r^2 C(r, t) dr}{R^3(t)} \end{aligned} \quad (12)$$

Here the first term on the right hand side of Eq. (12) expresses the amount of gas residing on the grain boundary in equilibrium with the gas inside the grain, while the second term represents the amount of gas inside the grain whose distribution is governed by the diffusion equation.

We assume that the ratio $B(t)/\beta(t)$ is time independent, since both b and β are proportional to the fission rate. If no gas is released, we have

$$G(t) = \int_0^t \beta(s) ds \quad (13)$$

We now let $\mathcal{G}(\tau(t)) = G(t)$ and use the definitions in (6); hence Eq. (12) can be rewritten as

$$\frac{\mathcal{G}(\tau)}{3} = \frac{\mathcal{C}(1, \tau)}{2\mathcal{B}(\tau)} + \int_0^1 \chi^2 \mathcal{C}(\chi, \tau) d\chi \quad (14)$$

Here we note that $\mathcal{G}(\tau)$ (if no gas is released) can also be calculated by

$$\mathcal{G}(\tau) = \int_0^\tau \mathcal{P}(s) ds \quad (15)$$

When concentration of gas at the grain boundary reaches a certain saturation value, $C_s(t)$, given by

$$C_s(t) = \frac{B(t)N_s}{D(t)} \quad (16)$$

then gas release will occur, where N_s is the areal density of gas atoms at grain faces at saturation. In computations we have assumed first that $C_s(t)$ is linear in t , and then upon grain boundary saturation, only a fraction f of the intergranular gas is released. This means that upon release the gas concentration at grain boundary will be dropped to a value of $(1-f)C_s$ [10, 11].

The boundary value problem presented by Eqs. (5)-(11) is amenable to numerical treatments. Since the numerical treatment of the problem gives rise to a set of stiff equations [13] the traditional Crank-Nicholson or explicit time integration technique is hardly suitable for this problem. Hence a finite difference method applying a Rosenbrock type time integration scheme [14] has been utilized to solve this boundary value problem, that is to calculate fission product gas release as a function of time. The details of this calculation will be presented elsewhere. In the ensuing sections we provide approximate analytical solutions for the foregoing differential-integral equations which agree with the aforesaid numerical solutions.

APPROXIMATE SOLUTIONS

The general solution of differential equation (5) can be expressed as the sum of the general solution of the corresponding homogeneous equation (complementary function) \mathcal{C}_c and any solution of the inhomogeneous equation (particular integral) \mathcal{C}_p . We write:

$$\mathcal{C}(\chi, \tau) = \mathcal{C}_c(\chi, \tau) + \mathcal{C}_p(\chi, \tau) \quad (17)$$

For the homogeneous part of the solution we have (cf. Eq. (14))

$$\frac{\mathcal{C}_c(1, \tau)}{2\mathcal{B}(\tau)} + \int_0^1 x^2 \mathcal{C}_c(x, \tau) dx = 0 \quad (18)$$

and the differential equation:

$$\frac{\partial \mathcal{C}_c}{\partial \tau} - \Delta'_\chi \mathcal{C}_c = 0 \quad (19)$$

The inhomogeneous part of the solution obeys:

$$\mathcal{P} = \frac{\partial \mathcal{C}_p}{\partial \tau} - \Delta'_\chi \mathcal{C}_p \quad (20)$$

and the integral equation (14):

$$\frac{\mathcal{G}(\tau)}{3} = \frac{\mathcal{C}_p(1, \tau)}{2\mathcal{B}(\tau)} + \int_0^1 x^2 \mathcal{C}_p(x, \tau) dx \quad (21)$$

The total amount of fission product release gas $\mathcal{F}(\tau)$ is calculated according to:

$$\mathcal{F}(\tau) = \int_0^\tau \mathcal{P}(s) ds - \mathcal{G}(\tau) \quad (22)$$

where the integral term represents the total amount of generated gas.

Expanding the complementary function $\mathcal{C}_c(\chi, \tau)$ in terms of a basis vector e_i satisfying relation (18)

$$\mathcal{C}_c = \sum_{i=1}^{\infty} \alpha_i e_i \approx \sum_{i=1}^n \alpha_i e_i \quad (23)$$

where α_i are constants and each $\mathcal{C}_c(\chi, \tau)$ satisfies relations (18) and (19). For the problem under consideration we factor out the main time dependence of e_i as:

$$e_i(\chi, \tau) = e^{-d_i \tau} \tilde{e}_i(\chi, \tau) \quad (24)$$

where $\tilde{e}_i(\tau)$ also satisfies relations (18) and (19). Introducing Eq. (24) into Eq. (19), we obtain:

$$\frac{\partial \tilde{e}_i}{\partial \tau} = \Delta'_x \tilde{e}_i + d_i \tilde{e}_i \quad (25)$$

Next we expand \tilde{e}_i as

$$\tilde{e}_i = \sum_{k=1}^{\infty} \lambda_i^k \hat{e}_i^k \approx \sum_{k=1}^2 \lambda_i^k \hat{e}_i^k \quad (26)$$

where \hat{e}_i^k satisfies Eq. (25) and the associating conditions. Also λ_i^k ($i \geq 1$) and d_i are selected such that the imposed integral and boundary conditions are satisfied for all τ . Moreover, we have $\lambda_i^0 = 1$. Hence using Eqs. (25) and (18); we have

$$\frac{\partial \hat{e}_i^k}{\partial \tau} = \Delta'_x \hat{e}_i^k + d_i \hat{e}_i^k \quad (27)$$

and

$$\frac{\mathcal{D}(\tau) \hat{e}_i^k(R, \tau)}{2\mathcal{B}(\tau)\mathcal{R}(\tau)} + \int_0^1 x^2 \hat{e}_i^k(xR, \tau) dx = 0 \quad (28)$$

where $\mathcal{D}(\tau) \Leftrightarrow D(t)$. Solution of the differential-integral equations (27) and (28) may be expressed as

$$\hat{e}_i^k = \sum_{m=0}^k \binom{k}{m} (2\tau)^{k-m} f_i^m \quad (29)$$

where f_i^m satisfy the following differential equations

$$\left. \begin{aligned} \Delta'_x f_i^0 + d_i f_i^0 &= 0 \\ \Delta'_x f_i^m + d_i f_i^m &= 2m f_i^{m-1} \quad (m \geq 1) \end{aligned} \right\} \quad (30)$$

For the problem under consideration it is a good approximation to assume that in Eq. (7) $c \equiv \mathcal{R}_\tau / \mathcal{R}$ (appearing in the operator Δ'_x) is a weak function of time and moreover $c \ll 1$, meaning that the rate of grain growth is smaller than grain size growth. It can be shown that for this case $f_i^m(r)$ has the form

$$f_i^m(r) \approx \left(\frac{r\sqrt{d_i}}{d_i} \right)^m j_m(r\sqrt{d_i}) \quad (31)$$

where $j_m(z)$ are the Spherical Bessel functions of first order [15]. The values $\{\alpha_i\}_{i=1}^n$ in (23) are determined by the continuity requirement imposed on the solutions from one time-step to another using generalized moments:

$$W_j(C^-) = W_j(C^+) \quad (32)$$

where C^- is the preceding solution at the end of the previous time step, while C^+ is the current solution at the start of the current time step with

$$W_n(C) = \int_0^1 x^2 j_0(n\pi x) \mathcal{E}_c(xR, \tau) dx \quad (33)$$

The total amount of gas can be calculated by using Eq. (21) and Eq. (12), we obtain

$$G(t) = \frac{3D f_i^m(R)}{2BR} + 3 \int_0^1 x^2 f_i^m(Rx) dx \quad (34)$$

where $f_i^m(\bullet)$ is given by Eq. (31).

Now turning to the solution of the inhomogeneous equation (particular integral), we write the solution in terms of power series expansion as follows

$$\mathcal{C}_p(r, \tau) = \sum_{k=1}^{\infty} \Lambda_k(\tau) C_p^k(r, \tau) \approx \sum_{k=0}^2 \Lambda_k(\tau) C_p^k(r, \tau) \quad (35)$$

where $\Lambda_k(\tau) \equiv \lambda_k$ is assumed to be constant for $k \geq 1$ and

$$\Lambda_0(\tau) \equiv \lambda_0 + \int_0^\tau \mathcal{P}(s) ds \quad (36)$$

Also $C_p^k(r, \tau)$ satisfy the following conditions:

$$\frac{\partial C_p^k}{\partial \tau} = \Delta_r' C_p^k \quad (37)$$

$$C_p^k(r, 0) = r^{2k} \quad (38)$$

As for the case of complementary function we suppose that $c \equiv \mathcal{R}_\tau / \mathcal{R} \ll 1$ is a weak function of time. It can be shown that $C_p^k(r, \tau)$ may be expressed by:

$$C_p^k(r, \tau) = (2k+1)!! (2\tau)^{2k} M(-k, 3/2, -r^2/4\tau) \quad (39)$$

where $M(a, b, \bullet)$ is the confluent hypergeometric function, Kummer's function [15]; and hence with this choice, $C_p^k(r, \tau)$ satisfy Eqs. (37) and (38). For example, we have

$$C_p^0(r, \tau) = 1; \quad C_p^1(r, \tau) = r^2 + 6\tau; \quad C_p^2(r, \tau) = r^4 + 20r^2\tau + 60\tau^2 \quad (40)$$

The values for $\Lambda_k(\tau) \equiv \lambda_k$ ($k=1, 2$) in Eq. (35) are determined from the integral and boundary conditions, as for the homogeneous part, by employing Eq. (32). The total amount of gas in the inhomogeneous term is calculated according to:

$$\mathcal{G}_p = \frac{3D C_p^k(R, \tau)}{2BR} + 3 \int_0^R r^2 C_p^k(r, \tau) dr \quad (41)$$

where $C_p^k(r, \tau)$ for $\{k=0,1,2\}$ are given by relations (40). The weight integrals are evaluated as in (33) in the manner

$$W_n(C_p) = \int_0^1 x^2 j_0(n\pi x) \mathcal{C}_p(xR, \tau) dx \quad (42)$$

where the weight density function is taken as the zeroth Spherical Bessel function of the first kind.

GAS RELEASE UNDER SMALL TIME STEPS

The selection of the forms for the particular solution discussed in the foregoing section may give a poor adjustment to the release behavior in the case of short time power variations. This in turn, will cause the coefficients in the complementary solution to become large, thereby rendering the approximation of comprising only few terms in the power series as inadequate. Introducing a new term in the particular solution, which treats the small time step solution of the problem, can circumvent this trouble. A condition to be satisfied is that the entire gas inventory during the time step is placed in the particular solution according to

$$G(C^-) = G(C_p^+) \quad (43)$$

where C^- and C_p^+ are the old solution at the end of the old time step and the new solution at the start of the new time step, respectively. The aforementioned new term is included in the series given by Eq. (35) in the form:

$$C_p(r, \tau) \approx C_x + \sum_{k=0}^2 \Lambda_k(\tau) C_p^k(r, \tau) \quad (44)$$

where C_x satisfies the equation

$$\frac{\partial C_x(r, \tau)}{\partial \tau} = \Delta C_x(r, \tau) \quad (45)$$

with conditions

$$C_x(r, 0) = 0 \quad \text{and} \quad C_x(R, \tau) = \tau \quad (46)$$

Introduce an auxiliary function with $x = r/R$:

$$C_x(Rx, \tau) \equiv \tau - C_x(Rx, \tau) \quad (48)$$

This function satisfies

$$C_x(r, 0) = 0 \quad \text{and} \quad C_x(R, \tau) = 0 \quad (49)$$

The following choice for $C_y(r, \tau)$ is considered

$$C_x(Rx, \tau) = 2 \sum_{n=1}^{\infty} \frac{\sin(n\pi x)}{n\pi x} (-1)^{n+1} \frac{1 - e^{-n^2\pi^2\tau}}{n^2\pi^2} \quad (50)$$

The total amount of gas in the grain is now calculated with the new term according to:

$$\mathcal{G}_p \left(C_x \left(\frac{r}{R}, \frac{\tau}{R^2} \right) \right) = \frac{\tau}{R^2} - \frac{1}{15} + \sum_{n=1}^{\infty} \frac{6e^{-\frac{n^2\pi^2\tau}{R^2}}}{n^4\pi^4} + 3 \frac{D}{2BR} \frac{\tau}{R^2} \quad (51)$$

For small time steps Eq. (51) is approximated by

$$\mathcal{G}_p \left(C_x \left(\frac{r}{R}, \frac{\tau}{R^2} \right) \right) \approx \frac{4\tau}{R^2} \sqrt{\frac{\tau}{\pi R^2}} - \frac{3}{2} \left(\frac{\tau}{R^2} \right)^2 + 3 \frac{D}{2BR} \frac{\tau}{R^2} + O \left(\frac{\tau}{R^2} \right)^\infty \quad (52)$$

Finally the weight integrals, for C_x , corresponding to Eq. (42) are in the form (for $j=1,2,3,\dots$):

$$W_j \left(C_x \left(\frac{r}{R}, \frac{\tau}{R^2} \right) \right) = \frac{\tau}{R^2} \frac{(-1)^{j+1}}{(j\pi)^2} - \frac{(-1)^{j+1}}{(j\pi)^4} \left(1 - \exp \left(-\frac{j^2 \pi^2 \tau}{R^2} \right) \right) \quad (53)$$

CALCULATION OF GAS RELEASE

The areal density of gas atoms at grain faces $N(t)$ is determined by Eqs. (12) and (13), i.e.:

$$\frac{N(t)}{R(t)} = \frac{2}{3} \int_0^t \beta(s) ds - \frac{2 \int_0^R r^2 C(r,t) dr}{R^3(t)} \quad (54)$$

When $N(t)$ reaches the saturation value N_s , release will occur. The quantity N_s can be calculated through an equation of state for gas atoms in the grain boundary. The density of gas at the grain boundary at saturation is $C_{gb}^s \equiv C_s = 3N_s / 2R$. We assume that once the grain boundary gas saturation occurs, gas release is controlled by a continuous process, meaning that the gas concentration within the intergranular bubbles is the average of the concentrations before and after the release, or $C_{av} = [C_s + (1-f)C_s] / 2$, here f is the fraction of gas released at saturation, which in our computations is taken to be $f=0.7$. Note that this formulation is equivalent to saying that gas release occurs by a continuous process above $0.65 C_s$. The amount of gas release during the time step is calculated by taking the gas content initially present in the intergranular bubbles, adding the gas generated during the time step, and subtracting the amount of gas present at the end of the time step. This technique reasonably approximates several discrete gas releases that would require many short time steps and long computer time.

COMPUTATIONS

In this section we present the results of our computations performed by applying the foregoing equations to calculate fission product gas release and the associating grain growth in UO_2 fuel under isothermal conditions. The computations not only provide insight to connection between release and grain growth but also offer sample cases for verification of release calculations in fuel modeling computer codes. The material properties used for the calculations consist of relations for UO_2 fuel grain growth and the diffusion coefficient for fission gas in UO_2 during irradiation. The grain growth kinetic model used is that given by Ainscough, Oldfield, and Ware [16] which is outlined in Table 1. This model is for equiaxed grain growth valid over the range 1300-1650°C for sintered stoichiometric UO_2 pellets with densities between 94 to 99% of theoretical density. The relations utilized for gas diffusion coefficient in for UO_2 fuel are based on the work of Speight [5], White and Tucker [17], Turnbull, White and Wise [18], and Matzke [19]. The relations are presented in Table 2 and the associating constants in Table 3.

The gas arriving at grain boundaries with a given rate will eventually saturate the boundaries through a network of interconnected bubbles. If the ideal gas equation of state is assumed, the density of the intergranular gas bubble at saturation is given by [17] $N_s = 8.72 \times 10^{-9} (2\gamma/r_f + P_{ext}) / T$ [atoms/m²], where r_f is the projected radius of the curvature of the capillary surface of the bubble and γ its surface tension, $2\gamma/r_f = 2.4 \times 10^6$ [Pa], T the temperature [K], and P_{ext} the external gas pressure.

The relations presented in this paper are programmed in a computer routine for calculation of fission gas release and grain growth as a function of irradiation time at different constant temperatures. The required input parameters for our computations are listed in Table 4. Fission gas production is taken as $\beta = 0.3\phi$, where the value of ϕ is given in Table 3.

The results of our computation of fission product gas release as a function of temperature, at constant linear power density of 43 kW/m (592 W/m³) for various irradiation times are shown in Fig. 1. The associating results for the evolution of grain size are plotted in Fig. 2. It can be seen from Fig. 1 that after a certain irradiation time fission gas release increases with

temperature reaching a maximum value then decreasing as temperature is increased. The reason for the decrease is the increase in grain size with temperature resulting in a longer diffusion path for gas atoms to reach the grain boundary, Fig. 2.

Table 1. UO₂ grain growth description according to Ainscough et al. [16]

$\frac{dR}{dt} = 0.25k_t \left(\frac{1}{R} - \frac{1}{R_m} \right)$ for $R \leq R_m$	[m/h]
$k_t = 5.24 \times 10^{-5} \exp\left(\frac{-2.67 \times 10^5}{8.314T}\right)$	[m ² /h]
$R_m = 1.115 \times 10^{-3} \exp(-7620/T)$	[m]
R : grain radius at time t	[m]
T : Temperature	[K]

Table 2. Relations used for gas diffusion coefficient in UO₂ fuel [17, 18]

$D_{eff} = \frac{b'D'}{b'+g}$	Effective diffusion coefficient used in governing equation for gas release
$D' = C_1 e^{-Q_1/RT} + 4C_2 \sqrt{\varphi_m} e^{-Q_2/T} + 4C_3 \varphi_m$	Gas diffusion coefficient in trap free media [m ² /s]
$b' = 3.03\pi l \varphi_m (\bar{R}_b + Z_0)^2$	Intragranular bubble gas re-resolution rate [1/s]
$\bar{R}_b = 1.453 \times 10^{-10} \exp(1.023 \times 10^{-3} T)$	Mean intragranular bubble radius [m]
$g = 4\pi \bar{R}_b C_b^{tot} D'$	The probability per second that a gas atom in solution is captured by a intragranular bubble
$C_b^{tot} = 1.52 \times 10^{27} / T - 3.3 \times 10^{23}$	Total gas bubble density [1/m ³]
$l = 6 \times 10^{-6}$ [m]; $Z_0 = 10^{-9}$ [m]; T : Temperature [K]; φ_m : fission density (see Table 3)	

Table 3. Coefficients and constants for the gas diffusivity relations presented in Table 2

$C_1 = 7.6 \times 10^{-10}$	$Q_1 = 7 \times 10^4$ [cal mol ⁻¹]
$C_2 = 1.41 \times 10^{-25}$	$Q_2 = 1.38 \times 10^4$ [K]
$C_3 = 2.0 \times 10^{-40}$	$R = 1.986$ [cal mol ⁻¹ K ⁻¹]
$\varphi_m = N_A \varphi \left[\frac{\text{fission}}{\text{m}^3 \text{s}} \right]$	$N_A = 6.022 \times 10^{23}$ [atoms/mole]
$\varphi = 5.189 \times 10^{-14} q_v \left[\frac{\text{mole fission}}{\text{m}^3 \text{s}} \right]$	
q_v : Volumetric heat generation rate	[W/m ³]

Table 4. Values selected for the input variables in the computations

$q_l = 43$	Linear power density [kW/m]
$q_v = 592$	Volumetric power density [MW/m ³]
$W = 63.9$	Fuel rating [W/kgU]
$R(0) = 5 \times 10^{-6}$	Initial UO ₂ grain radius [m]
$P_{ext} = 0.0$	External gas pressure acted on intergranular gas bubbles
$B(t)/\beta(t) = 5.7 \times 10^{-8}$	Ratio of gas resolution velocity to gas production rate [ms ⁻¹ /mol m ⁻³ s ⁻¹]
$\beta = 0.3\varphi = 1.557 \times 10^{-14} q_v$	mol m ⁻³ s ⁻¹ (see Table 3)

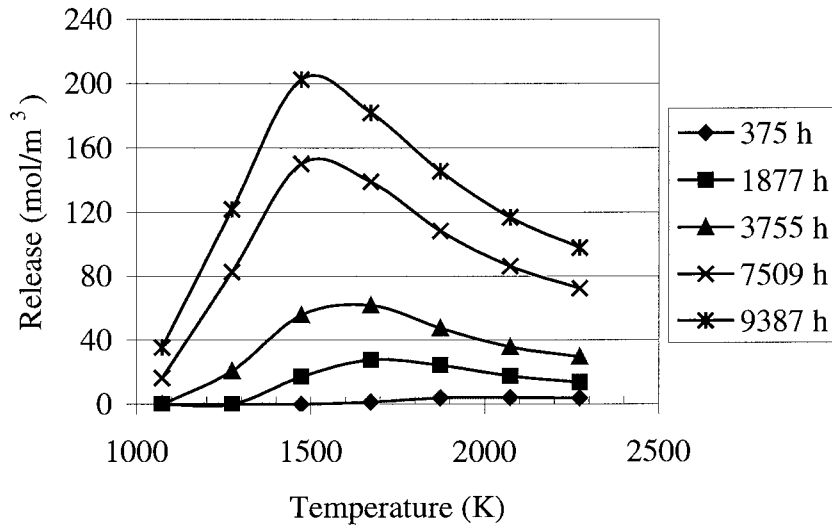


Fig. 1. Evolution of fission gas release in UO₂ fuel as a function of temperature at a constant power density.

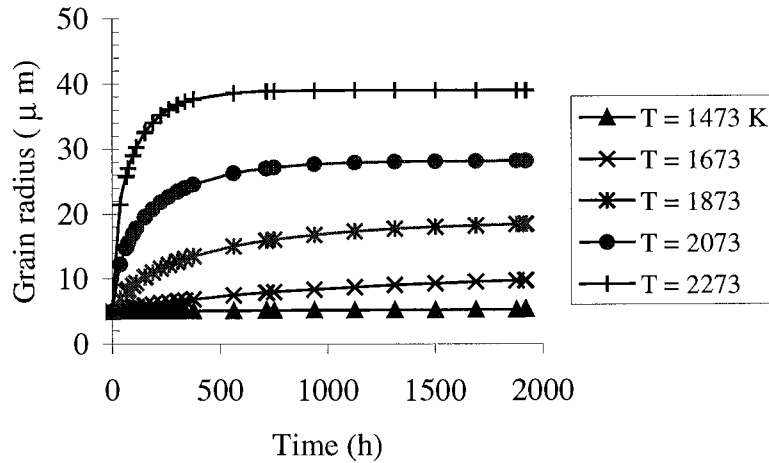


Fig. 2. Evolution of grain size in UO₂ fuel as a function of time and temperature.

DISCUSSION AND CLOSURE

The remarkable effect of the temperature dependence of fission gas release during grain growth (see Fig. 1) as predicted by the theory presented in the foregoing sections is briefly discussed in this section. This effect may be elucidated if we compare the time constants (or "frequencies") for gas diffusion and grain growth. The frequency for gas diffusion can be expressed by $\nu_D \equiv D/R^2$ whereas the corresponding quantity for grain growth is given by $\nu_G \equiv \dot{R}/R$, where $\dot{R} \equiv dR/dt$. Using Ainscough et al. [16] UO_2 grain growth description presented in Table 1 and the diffusion coefficient relations given in Table 2, we have calculated ν_D and ν_G as a function of temperature for a constant grain radius of $5 \mu\text{m}$, see Fig. 3. We observe that at a certain temperature the grain growth becomes much more rapid than the fission product gas diffusion in UO_2 . Since fission product gas diffusion time in UO_2 grain is relatively long, as can easily be estimated from $\nu_D^{-1} \equiv R^2/D$, this will result in lower release at higher temperatures, for which the grain size has reached its maximum saturation values (see Fig. 2), and grain boundary sweeping is no longer effective. This is an explanation of the effect seen in Fig. 1, which is obtained by our calculations using the theory presented in the foregoing sections. We should remark, nevertheless, that the results presented here depend on the precision and the range of validity of the material models (diffusivity and grain growth) utilized. Only direct comparison with accurate and detailed experiments can validate the prediction of the theory. We have consciously carried out our calculations beyond 2000 K, although we are aware that other mechanisms, which have not been taken into account in the present analysis, may contribute to fission product release beyond this temperature.

Grain growth concomitant with thermal release has been observed in boiling water reactor fuel both during normal operation [20] and in power ramp experiments [21]. Schrire and Lysell [21] studied UO_2 fuel microstructure and fission product distribution by power ramping a rod, pre-irradiated in a commercial BWR to a fuel burnup of 35 MWd/kgU , in Studsvik's R2 test reactor to a maximum linear power density of 43 kW/m and compared the results with a similar reference fuel rod which had experienced a maximum linear power density of 26 kW/m during its final cycle in the BWR. The power bump started with a 72 h irradiation at the peak LHGR of 25 kW/m in order to build sufficient amount of short lived fission products. Then ramping was performed at a rate of 0.69 kW/m.h to 43 kW/m , and held at this power for 3 hours to redistribute and release fission product gases. Schrire and Lysell's [21] scanning electron microscopy of fuel pellet revealed a strong coupling between fission product gas release and grain growth across pellet radius for the ramped rod. The average grain size at the center of the pellet (with radius of 5 mm) had increased by a factor of two (from 5.5 to 11 μm) and the release fraction from less than 0.05 to 0.74 from the center to the periphery of the pellet, respectively. The experiment clearly shows the importance of a physically based model to describe the phenomena of grain growth and fission product gas release in reactor fuel concurrently. Quantitative analyses of in-reactor experiments, such as that made by Schrire and Lysell requires implementation of the mathematical method in a nuclear fuel modeling computer program and simulation of the experiment. Our aim in the present paper has been to provide a consistent theoretical framework for that purpose.

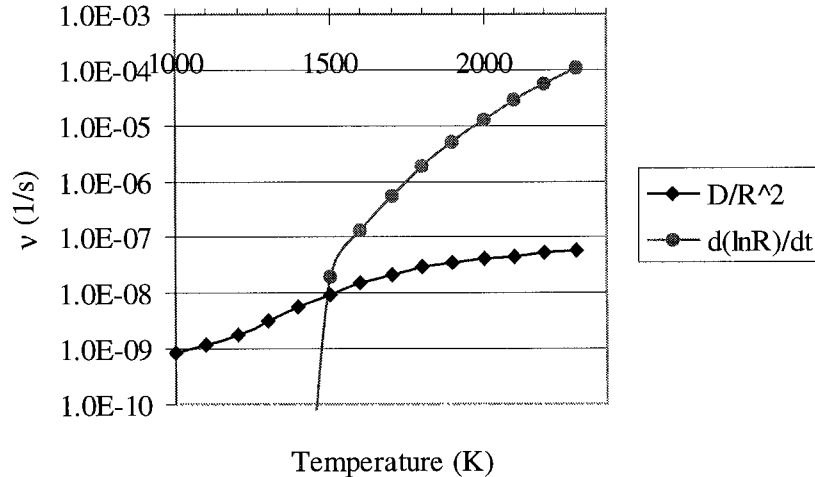


Fig. 3. Inverse time constants of fission product gas diffusion $\nu_D \equiv D/R^2$ in UO_2 and grain growth $\nu_G \equiv \dot{R}/R$ as a function of temperature for a UO_2 grain size of radius $R=5 \mu\text{m}$.

REFERENCES

1. Olander, D.R., *Fundamental Aspects of Nuclear Reactor Fuel Elements*, TID-26711-P1, National Technical Service, U. S. Department of Commerce, Springfield, Virginia, 1976.
2. Lorenz, R.A., "ANS-5.4 fission gas release model III, low temperature release," Proc. of the ANS Topical Meeting on LWR Fuel Performance, Portland, Oregon, April 1979.
3. Forsberg, K., Lindström, F. and Massih, A.R., "Modelling of some high burnup phenomena in nuclear fuel," Proc. of the IAEA Technical Meeting on Water Reactor Fuel Element Modelling at High Burnup and Experimental Support, Windermere, UK, September 1994.
4. Booth, A.H. "A method of calculating fission gas diffusion from UO_2 fuel and its application to the X-2-f test," AECL Report 496, 1957.
5. Speight, M.V., "Calculation on the migration of fission gas in material exhibiting precipitation and re-solution of gas atoms under irradiation," *Nuclear Science & Engineering*, Vol. 37, 1969, pp. 180-185.
6. Matthews, J.R. and Wood, M.H., "An efficient method for calculating diffusive flow to a spherical boundary," *Nuclear Engineering & Design*, Vol. 56, 1980, pp. 439-443.
7. Forsberg, K. and Massih, A.R., "Fission gas release under time varying conditions," *Journal of Nuclear Materials*, Vol. 127, 1985, pp. 141-145.
8. Lassmann, K. and Benk, H., "Numerical algorithms for intragranular fission gas release," *Journal of Nuclear Materials*, Vol. 280, 2000, pp. 127-135.
9. Lassmann, K., "Numerical algorithms for intragranular diffusional release incorporated in the TRANSURANUS code," International Seminar on Fission Gas Behavior in Water Reactor Fuels, Cadarache, France, September 2000.
10. Forsberg, K. and Massih, A.R., "Diffusion theory of fission gas migration in irradiated nuclear fuel UO_2 ," *Journal of Nuclear Materials*, Vol. 135, 1985, pp. 140-148.
11. Forsberg, K., Massih, A.R. and Andersson, K., "Calculation of fission gas migration in nuclear fuel with re-solution effect," Proc. of the Enlarged Halden Programme Group Meeting on Fuel Performance Experiments and Analysis, Sanderstølen, Norway, March 1986.
12. Paraschiv, M.C., Paraschiv, A. and Grecu, V.V., "A theoretical study of volatile fission products release from oxide fuels," *Journal of Nuclear Materials*, Vol. 275, 1999, pp. 164-185.
13. Press, W.H., Teukolsky, S.A., Vetterling, W.T. and Flannery, B.P., *Numerical Recipes in FORTRAN*, Second Edition, Cambridge University Press, Cambridge, 1992.
14. Hede, G., "Numerical Solution of Kinetic Equations for Fission Gas Release with Grain Growth," M.Sc. Dissertation, Uppsala University, UPTEC 94 034E, Uppsala, Sweden, March 1994.
15. Abramowitz, M. and Stegun, I.A., *Handbook of Mathematical Functions*, Dover, New York, 1965.
16. Ainscough, J.B., Oldfield, B.W. and Ware, J.O. "Isothermal grain growth kinetics in sintered UO_2 pellets," *Journal of Nuclear Materials*, Vol. 49, 1973/74, pp. 117-128.
17. White, R.J. and Tucker, M.O., "A new fission-gas release model," *J. Nuclear Materials*, Vol. 118, 1983, pp 1-38.
18. Turnbull, J., White R.J. and Wise, C. "The diffusion coefficient for fission gas atoms in uranium dioxide," Proc. of the IAEA Technical Committee Meeting on Water reactor Fuel Element Computer Modeling, Preston, UK, September 1988.
19. Matzke, H., "Gas release mechanisms in UO_2 - a critical review, *Radiation Effects*, 53, 1983, pp. 219-242.
20. Grapengiesser, G., Massih, A.R. and Nylund, O., "High burnup evaluation activities in Sweden and Finland," Proc. of the ANS International Topical Meeting on LWR Fuel Performance, Williamsburg, April 1988.
21. Schrire, D. and Lysell, G. "Fuel microstructure and fission product distribution in BWR fuel at different power levels," Proc. of the ANS-ENS International topical meeting on LWR fuel performance, April 1991, Avignon, France.