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Journal of Nuclear Materials 326 (2004) 97–105
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Theoretical analysis of two-stage fission gas release processes: grain lattice and grain boundary diffusion

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Received 27 October 2003; accepted 15 December 2003

Abstract

A theoretical analysis of two-stage fission gas release processes, grain lattice and grain boundary diffusion, coupled with bubble trapping and resolution, is carried out. Final results show that the conventional single-stage fractional release solution can be expanded for the two-stage release case with the introduction of a new dimensionless parameter $a/(\alpha + a)$ where α is the relative diffusivity ratio defined as D_{v}^{eff}/D_{gb}^{eff}. In fact, α is a temperature- and burn-up-dependent gas transport property. Recent evaluations of α demonstrate that grain boundaries can play significant roles in fission gas release, depending on the fuel power history, and the high burn-up behavior of fission gasses can be directly reflected in the two-stage parameter. Inter-granular bubble linkages effectively increase the two-stage parameter, which leads to apparent burn-up enhancement of the fractional fission gas release.

PACS: 28.41.Ak; 61.72.Mm; 66.30.Jt

1. Introduction

Fission gas release is of great concern in fuel performance evaluation, especially for high burn-up fuels. It affects gap conductance and fuel temperature, which in turn affect the release. Accurate prediction of the release is therefore essential not only for the rod pressure calculations that influence end-of-life rod pressures and LOCA (loss of coolant accident) analyses, but also for the radioactivity estimation in the gap that determines the radiological consequences. Currently in the nuclear fuel industry, end-of-life rod pressure limits the linear heat generation rate of the nuclear fuel rods when burn-up is greater than 30 GWd/MtU. In addition, Nuclear Regulatory Commission requires that bounding Anticipated Operational Occurrences (AOO) are included in the rod pressure analyses. The anticipated operational occurrences result in over-power transients of several minutes to hours in length, so that transient fission gas release analysis has also become important for licensing activities.

Since the LWR operation mode changed from annual to extended/high burn-up fuel cycles in the late 1980s, more attention has been paid to fission gas release phenomena [1–7]. There have been reports that fractional fission gas release is even accelerating with increasing burn-up. Therefore, for the development of high performance fuels, fission gas release is considered to be a potential design-limiting factor because of its crucial influence on the thermo-mechanical behavior of the current LWR fuel rods in heavy duty applications.

Uranium dioxide fuel pellets are a polycrystalline ceramic material consisting of many small grains. Fission gas atoms generated by fission reactions start to volumetrically diffuse onto grain boundaries and, on reaching there, continuously diffuse along the boundaries until they release to the open space in the fuel rod. The grain boundary is believed to have a significant role in the release, as do inter- and intra-fission gas bubbles, especially in the high burn-up fuels. Thus excellent

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doi:10.1016/j.jnucmat.2003.12.014

works on the grain-boundary-related fission gas release have been reported by several authors [8–13]. Particularly Olander's numerical model of solute transport in a polycrystalline body is one of the fundamental but pioneering works [11].

However, analytical approaches to the gas transport in the two regions of the operating fuels, grain lattice and grain boundary, have not been successful, and the current understanding of the grain boundary role and its related phenomena is still ambiguous. A diffusion model to describe the physical processes of the fission gas release in UO_2 fuel was first proposed by Booth [14]. In his model, fuel is treated as an assembly of uniform spheres with a single equivalent radius along with the perfect sink boundary condition. With the accumulation of in-pile experience, however, it has been revealed that gas–gas interactions lead to the formation of gas-filled intra-granular bubbles during the diffusion process when fuel burn-up increases. In addition, the perfect sink assumption does not conform to the micro-graphical examination results that gas atoms accumulate continuously in the grain boundaries, mostly causing the formation of inter-granular bubbles. It has also been found that fission fragments resolve the bubbles into the near-grain boundary region during reactor operation, and this augments the concentration of fission gas atoms in the grain boundary. The intra- and inter-granular bubbles, especially in high burn-up fuels, are schematically shown in Fig. 1.

In actuality, the precipitation of gas atoms in the bubbles and their resolution into the lattice complicate the analysis of fission gas release phenomena. Several mechanistic models have been proposed to take the non-zero grain boundary concentration into consideration [15–18]. Speight proposed a method to determine the grain boundary concentration [15] and Turnbull derived the analytic solution for the non-zero constant boundary concentration case [16]. Forsberg and Massih considered the time-dependent grain boundary condition and treated it numerically with the assumption that fission gas atoms release completely when the grain boundary concentration exceeds a certain saturated value and then accumulate again [18].

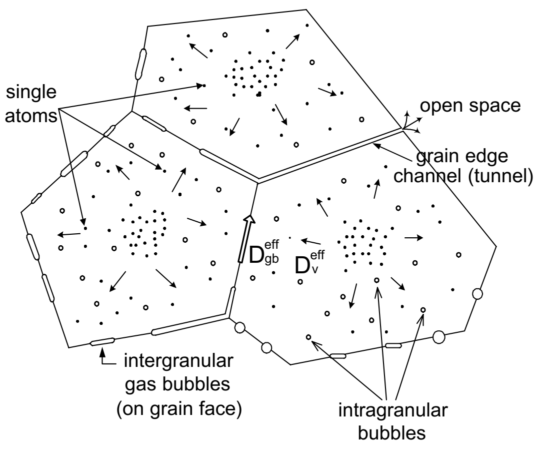

Fig. 1. A schematic of cross sectional view of a high burn-up fuel.

In this study, we conduct a theoretical analysis of fission gas release in the two regions, grain lattice and grain boundary. Two simultaneous time-dependent gas atom balance equations, coupled with bubble trapping and resolution, are formulated with the appropriately defined diffusion coefficients. Then, the two equations are reduced to a single partial differential equation by incorporating the relative diffusivity ratio into a time-dependent surface boundary condition of the third kind, a combined Dirichlet and Neumann condition. In the following section, most of the discussion is devoted to the simplified quasi-steady state solution, primarily the role of the grain boundary and the introduction of the two-stage parameter in the current two-stage fission gas release analysis.

2. Mathematical formulation

First, coupled with the bubble trap and the bubble resolution, fission gas transport is broken down into the two principal processes: the effective grain lattice diffusion, and the effective grain boundary diffusion. Fig. 2 schematically shows the fundamental processes of the current two-stage model, with the assumption that a grain has an ideal tetrakaidecahedron structure and thus the grain boundary can be treated as a flat surface between two neighboring grains. It is further postulated for the extension to high burn-up cases that fission gas bubbles at the grain edges are linked together to form grain edge tunnels that are finally connected to the open space inside the fuel rod. This means that, after the fission gases volumetrically diffuse through the grain lattice and reach the grain boundaries, they continuously surface-diffuse again along the grain boundaries and release on arrival at the edge tunnels of grain surfaces. This provides the basis of cylindrical geometric approach to solve the following solute transport formulation in the grain boundary.

Because each grain of UO_2 fuel is treated as a homogeneous matrix in this analysis, an effective diffusion coefficient D_{trap}^{eff} can be chosen for the description of the gas transport in the grain lattice. As defined by Speight, the coefficient includes the effects of fission gas atoms precipitating into the bubbles and their resolution into the lattice in the following way [15]:

$$D_{trap}^{eff} = \left(\frac{b}{b+g}\right) D_{trap}^{in\text{-}pile} \quad (1)$$

where g is the probability of the gas atom being captured by the bubbles and b is the probability of the bubble's resolution by fission fragments. Of course, $D_{trap}^{in\text{-}pile}$ is defined as a lattice diffusion coefficient in the presence of the fission reaction, which reflects much faster re-entry of the gas or release of the gas from crystalline traps under the in-pile environment in the intrinsic diffusion coefficient [15,19]. For simplicity, the effective coefficient D_{trap}^{eff} is represented by the notation D_v^{eff} throughout this paper.

The governing equation for the effective grain lattice diffusion is then

$$\frac{\partial C_v}{\partial t} = \beta + \frac{1}{R^2}\frac{\partial}{\partial R}\left(D_v^{eff} R^2 \frac{\partial C_v}{\partial R}\right) \tag{2}$$

with the initial condition $C_v(R,0) = 0$, and boundary conditions $C_v(0,t) =$ finite and $C_v(a,t) = \overline{C}_{gb}(t)$. In the equation, C_v is the volumetric fission gas concentration within the grain, β is the fission gas generation rate, and a is the equivalent radius of the grain. The lattice diffusion term in the RHS of Eq. (2) is expressed in spherical coordinates because the polyhedral grain is treated as an equivalent sphere in this analysis. Note that the surface boundary condition is the time-dependent average grain boundary concentration that is to be solved simultaneously.

Now, the fission gas atom concentration in the grain boundary is expressed as

$$\delta\frac{\partial C_{gb}}{\partial t} = \delta\frac{1}{r}\frac{\partial}{\partial r}\left(D_{gb}^{eff} r \frac{\partial C_{gb}}{\partial r}\right) - 2D_v^{eff}\left(\frac{\partial C_v}{\partial R}\right)_{R=a} \tag{3}$$

subject to the initial condition $C_{gb}(r,0) = 0$, and the boundary conditions $C_{gb}(0,t) =$ finite and $C_{gb}(s,t) = 0$. In Eq. (3), C_{gb} is the fission gas concentration in the grain boundary, δ is the grain boundary thickness, and s is the equivalent radius of the grain surface. The effective grain boundary diffusion coefficient D_{gb}^{eff} can be defined similarly to D_v^{eff} in Eq. (1), because qualitatively intergranular bubbles can also trap the inter-granularly diffusing gas atoms and the bubbles can be resolved into the boundary region by the fission fragments. The two principal fission gas transports, D_v^{eff} and D_{gb}^{eff}, are schematically shown in Figs. 1 and 2. Fundamentally Eq. (3) is identical to the Olander's formulation for the combined grain boundary and lattice diffusion in a fine-grained ceramic [11].

Eq. (3) must be solved using cylindrical geometry because the gas transport process in the grain boundary is basically a surface diffusion from the center of the grain surface to the edge, as seen in Figs. 2 and 3. In this formulation, the unique fission gas supply to the grain boundary is the gas atoms arriving at the two adjacent boundaries that face each other. This is the reason the last term in the RHS is doubled.

Because $D_v^{eff}(\frac{\partial C_v}{\partial R})_{R=a}$ in Eq. (3) is a function only of time, it can be represented as $f(t)$. Hence, when $\xi = D_{gb}^{eff}$ and $k = -\frac{\delta\xi}{2}$ are introduced, the governing equation can be rewritten as

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial C_{gb}}{\partial r}\right) + \frac{1}{k}f(t) = \frac{1}{\xi}\frac{\partial C_{gb}}{\partial t}. \tag{4}$$

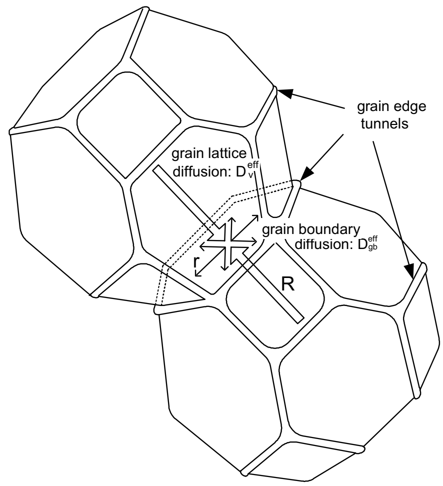

Fig. 2. A schematic of two principal diffusion processes in the tetrakaidecahedron grain: grain lattice diffusion D_v^{eff} and grain boundary diffusion D_{gb}^{eff}.

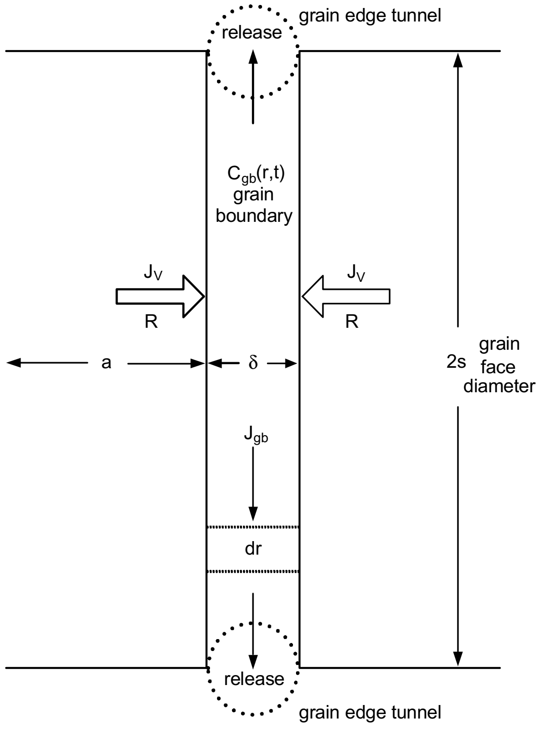

Fig. 3. Balance diagram of fission gas atom concentration within the grain boundary.

The general solution of this equation can be determined by using the Green's function method:

$$C_{gb}(r,t) = \frac{\xi}{k}\int_{\tau=0}^{t} d\tau \int_{r'=0}^{s} r' G(r,t|r',\tau) f(r',\tau)\, dr' \tag{5}$$

and the Green's function suitable for this case is known to be [20,21]:

$$G(r,t|r',\tau) = \frac{2}{s^2}\sum_{m=1}^{\infty} e^{-\xi\beta_m^2(t-\tau)} \frac{J_0(\beta_m r)}{J_1^2(\beta_m s)} J_0(\beta_m r'), \tag{6}$$

where J_0 is the 0th ordinary Bessel function of the first kind and β_m is the mth root coefficient.

Therefore, $C_{gb}(r,t)$ becomes

$$C_{gb}(r,t) = \frac{2\xi}{ks^2}\sum_{m=1}^{\infty} \frac{J_0(\beta_m r)}{J_1^2(\beta_m s)} \times \int_{\tau=0}^{t} e^{-\xi\beta_m^2(t-\tau)} f(\tau)\, d\tau \int_{r'=0}^{s} r' J_0(\beta_m r')\, dr'. \tag{7}$$

From the following integration property of Bessel functions:

$$\int_0^s r' J_0(\beta_m r')\, dr' = \frac{s}{\beta_m} J_1(\beta_m s), \tag{8}$$

$C_{gb}(r,t)$ reduces in this way

$$C_{gb}(r,t) = -\frac{4}{\delta s}\sum_{m=1}^{\infty} \frac{J_0(\beta_m r)}{\beta_m J_1(\beta_m s)} \int_0^t e^{-\xi\beta_m^2(t-\tau)} f(\tau)\, d\tau. \tag{9}$$

Now, the average grain boundary concentration of fission gas atoms can be obtained by the following definition:

$$\overline{C}_{gb}(t) = \frac{2}{s^2}\int_0^s C_{gb}(r,t) r\, dr. \tag{10}$$

Hence, we obtain

$$\overline{C}_{gb}(t) = -\frac{8}{\delta s^2}\sum_{m=1}^{\infty} \frac{1}{\beta_m^2}\int_0^t e^{-\xi\beta_m^2(t-\tau)} f(\tau)\, d\tau. \tag{11}$$

Eq. (9) shows that the solutions of Eqs. (2) and (3) are directly coupled. That is, to solve Eq. (2) completely, we must find $C_{gb}(r,t)$, which also requires the solution of $C_v(R,t)$ again.

In fact, there are several ways to solve the equations simultaneously. One approach is a numerical method and another may be an iterative solution technique that begins with the assumption of a simple but plausible function for $C_v(R,t)$. However, these transient solution techniques do not seem to be straightforward. Nor may it be easy to grasp the real meaning of the two competing fission gas transport processes and their combined effects on the release, especially in high burn-up fuels. Therefore, even with this complete transient formulation, most of the following discussion is devoted to the simplified quasi-steady state solution, leaving the transient solution for a later paper.

3. Analytical solution using boundary conditions of the third kind

Based on the properties of Bessel functions, the average grain boundary concentration in Eq. (11) can be related to the rate at which fission gas atoms leak to the open space. Because $\partial\{J_0(\beta_m r)\}/\partial r = -\beta_m J_1(\beta_m r)$, the first derivative at the grain edge reduces to

$$-\delta \left.\frac{\partial C_{gb}}{\partial r}\right|_{r=s} = -\frac{4}{s}\sum_{m=1}^{\infty}\int_0^t e^{-\xi\beta_m^2(t-\tau)} f(\tau)\, d\tau. \tag{12}$$

This reduction reveals that the two Eqs. (11) and (12) are mathematically in an identical form except for the constants around the summation symbol.

Under typical reactor operating conditions, $\xi\beta_m^2(\tau - t)$ is generally far smaller than unity and thus terms with $m > 2$ attenuate very fast. Comparison of the two equations, therefore, results in the following relation between the average concentration and the leakage flux:

$$\frac{\delta s\beta_1^2}{2}\overline{C}_{gb}(t) \cong \delta \left.\frac{\partial C_{gb}}{\partial r}\right|_{r=s}. \tag{13}$$

Most nuclear power plants are operated in the steady state with the designed constant power, although the linear heat generation rate and the temperature profile inside a fuel rod differ from rod to rod and change slowly with fuel burn-up. In this sense, fuel performance analyses, even for the initially loaded reactor core, are carried out on the basis of summation of the steady states attained during each segmented time frame. In such states without rapid power ramp or drop, it can be postulated that there is no sudden accumulation or depletion of fission gas atoms in the grain boundary. That is, all the fission gas atoms reaching the grain boundaries diffuse towards the grain edges and eventually leak out to the open space in the fuel rod, maintaining a constant grain boundary concentration. This assumption seems to be reasonable, particularly for high burn-up, because it is believed that the grain boundary can only retain a certain amount of fission gas atoms, i.e., the saturated concentration. In this case, the leakage rate out of the grain surface becomes equal to the arrival rate at the grain surface.

That is,

$$\delta D_{gb}^{eff} \left.\frac{\partial C_{gb}}{\partial r}\right|_{r=s} \cong s D_v^{eff} \left.\frac{\partial C_v}{\partial R}\right|_{R=a}. \tag{14}$$

Then the combination of Eqs. (13) and (14) yields

$$\overline{C}_{\rm gb}(t) = \frac{2}{\delta\beta_1^2}\frac{D_{\rm v}^{\rm eff}}{D_{\rm gb}^{\rm eff}}\frac{\partial C_{\rm v}}{\partial R}\bigg|_{R=a} \equiv \alpha\frac{\partial C_{\rm v}}{\partial R}\bigg|_{R=a}, \tag{15}$$

where $\alpha = \frac{2}{\delta\beta_1^2}\frac{D_{\rm v}^{\rm eff}}{D_{\rm gb}^{\rm eff}}$.

Eq. (15) vividly demonstrates that the surface boundary condition for the balance Eq. (2) can be replaced with a boundary condition of the third kind, a combined Dirichlet and Neumann condition:

$$C_{\rm v}(a,t) = \overline{C}_{\rm gb}(t) = \alpha\frac{\partial C_{\rm v}}{\partial R}\bigg|_{R=a}. \tag{16}$$

That is,

$$\alpha\frac{\partial C_{\rm v}}{\partial R}\bigg|_{R=a} - C_{\rm v}(a,t) = 0. \tag{17}$$

This means that Eq. (3), representing the balance of the fission gas atoms in the grain boundary, is incorporated into a boundary condition of the third kind for Eq. (2) without any significant loss of analytical foundation. This incorporation simplifies both the computational treatment and the analysis of complicated fission gas release processes. Note that the equivalent radius of the grain surface s drops out in the course of derivation.

To solve this problem, Laplace transform method can be used with the variable substitution technique using dimensionless variables. The analytical solution using the method is developed in Appendix A. Once the solution of Eq. (2) subject to the new boundary condition is obtained, J, the leakage rate of fission gas atoms per unit surface area per unit time, can be easily derived from Fick's first law.

Then, according to the following definition of fractional fission gas release:

$$F = \frac{4\pi a^2 \int_0^t J\,{\rm d}t'}{4/3\,\pi a^3 \beta t} = \frac{3}{a\beta t}\int_0^t J\,{\rm d}t' \tag{18}$$

the fractional release is finally obtained

$$F \cong \frac{4}{\sqrt{\pi}}\left(\frac{a}{\alpha+a}\right)^2\left(\frac{D_{\rm v}^{\rm eff}t}{a^2}\right)^{1/2} - \frac{3}{2}\left(\frac{a}{\alpha+a}\right)\left(\frac{D_{\rm v}^{\rm eff}t}{a^2}\right). \tag{19}$$

As seen in Eq. (19), multiples of the two-stage parameter, $a/(\alpha+a)$, appear in each term of the simple Booth solution, factorizing it with the new dimensionless property. This result clearly shows that current two-stage mathematical model reduces to the simple Booth single-stage model when $\alpha = 0$, i.e., when grain boundary diffusivity is infinite, which corresponds to the perfect surface sink condition.

In the post-irradiation examination (PIE) case, it is easily derived that the exactly same multiples of the two-stage parameter also show up in each term of the simple Booth solution, as in the in-pile case. The solution is as follows:

$$F \cong \frac{6}{\sqrt{\pi}}\left(\frac{a}{\alpha+a}\right)^2\left(\frac{D_{\rm v}^{\rm eff}t}{a^2}\right)^{1/2} - 3\left(\frac{a}{\alpha+a}\right)\left(\frac{D_{\rm v}^{\rm eff}t}{a^2}\right). \tag{20}$$

4. Discussion

Fission gas release phenomenon is strongly related to micro-structural changes in the fuel pellets. At low burn-up, these changes are confined to the central region of the pellet. However, pronounced changes take place even at the periphery in high burn-up rods. In-pile experiences have certainly demonstrated that intra- and inter-granular fission gas bubbles nucleate and grow inside the pellet as burn-up increases, and thus the fractional release becomes higher in the high burn-up fuel than in standard rods with low enrichment. It even tends to accelerate when burn-up exceeds around 45–50 GWd/MtU [1,5], although this is undoubtedly in conjunction with the thermal conductivity decrease of the fuel and the thermal conductance reduction of the gap.

Nevertheless, in his model, Booth assumed a fuel pellet to be an assembly of homogeneous uniform spheres and the fission gas storage capacity of the grain boundary to be zero [14]. Then he moved on to the reduction of the real two-stage fission gas transport processes to a simplified single-stage process by including the grain boundary migration in the presumably enlarged grain lattice diffusion. ANSI/ANS-5.4 adopted the model with the best-fitting parameters based on the empirically selected data because it is simple but useful for phenomenological analysis, although it does not properly account for high burn-up fuel behavior [22].

Later Speight successfully showed that simple adoption of the new diffusion coefficient $D_{\rm trap}^{\rm eff} = (b/b+g)D_{\rm trap}^{\rm in\text{-}pile}$ can make the Booth concept applicable even for high burn-up cases without solving two simultaneous balance equations, one for the fission gas concentration in the grain matrix and the other in the intra-granular bubbles [15]. He also raised the issue that the grain boundary must have some capacity for the storage of fission gases, i.e., the gas concentration in the boundary is no longer zero, and can probably be expressed in the following way: $C(a) = b\lambda N/2D$, where λ is the average distance of an atom ejected into the lattice by a fission fragment spike and N is the instantaneous number of gas atoms in the bubbles per unit area of grain boundary.

In dealing with this issue, Forsberg and Massih recently made outstanding progress and offered a numerical procedure to solve the non-zero grain boundary problem [18,23]. Their model is used now as one of the high burn-up models in the FRAPCON-3 code with some modifications [24]. Despite the strengths of the model, however, it was reported that their original model under-predicts much steady state in-pile data, especially at high burn-up above 45 GWd/MtU, and all of the ramped power data. In addition, some limitations and room for improvement in their treatment still remain. Because their solution is numerical, it is not easy to understand the role of the grain boundary and its effect on the resulting release fraction, even after the computational procedure is completed. Decisively, their assumption that all fission gas atoms in the grain boundaries release when the inter-granular concentration exceeds a certain saturation value and accumulate again from the beginning is disputable. As well, at least one or two vague parameters must be quantified before the model can be applied.

The solution of the current two-stage model developed here does not have those fundamental weaknesses because it mechanistically follows the two real fission gas transport processes with no sacrificial assumptions. As shown in Eqs. (19) and (20), this model successfully sorts out the role and the effect of the grain boundary with the two-stage parameter $a/(\alpha + a)$, where a is the equivalent grain radius and α is the ratio of diffusivities, defined as $\frac{2}{\delta\beta_1^2}\frac{D_v^{\mathrm{eff}}}{D_{\mathrm{gb}}^{\mathrm{eff}}}$ previously. It is easily seen that the ratio α has the unit of length and thus the new parameter turns out to be dimensionless because the unit of the root coefficient of Bessel function β_1 is the reciprocal of length. According to the two-stage parameter, two competing physical quantities, the equivalent grain radius a and the diffusivities ratio α, seem to determine the role and the effect of the grain boundary on the release of fission gas. In fact, a is not just a radius, but can be interpreted as a volume-to-surface ratio, whereas α can be regarded as a volume-to-surface diffusion ratio. Unless grain growth takes place seriously during reactor operation, the volume-to-surface ratio is a geometric constant, typically 5–10 μm, which is set during the manufacturing process through sintering. On the other hand, α, the ratio of grain lattice over grain boundary diffusion, is a variable transport property that is strongly dependent on both the fission gas species and the temperature and the burn-up according to reactor power history.

The new parameter reduces to unity only when α is much less than a, that is, when grain size is relatively large compared with a low grain lattice diffusivity or when grain boundary diffusivity is far much greater than grain lattice diffusivity. This corresponds to the case formulated in the simple Booth model. When α becomes similar to or greater than the grain radius a, Eqs. (19) and (20) show that the grain boundary must significantly affect the whole fission gas release process, because the square of the parameter appears in the dominant first term of the prediction expression.

In this model, the role of the grain boundary is, in actuality, incorporated in α, the ratio of two diffusivities. It is easily understood that α is basically $D_v^{\mathrm{eff}}/D_{\mathrm{gb}}^{\mathrm{eff}}$ with the unit of centimeter, because δ is on the order of 10^{-8} cm, β_1 is $2.405/s$ where s is the equivalent radius of a grain surface which is on the order of 10^{-4} cm, and thus $2/\delta\beta_1^2$ is close to unity. In fact, it is well known that the diffusion coefficient is one of the most critical properties for accurate estimation among those used in the fission gas release prediction. Nowadays, several data for D_v [25–31] and very few data or estimation for D_{gb} are available. In any case, however, it is very risky to take the absolute values because even the lattice diffusivity of Xe in UO_2 is not well established. For example, currently available data sets present a spread of a factor of 10^3 in D_v at 1400 °C (5×10^{-15}–10^{-12} cm^2/s). Fortunately, α requires only the ratio, not the absolute values.

Like the diffusion coefficient itself, the diffusivities ratio α is dependent on both temperature and burn-up. Although very different activation energies for grain lattice and grain boundary diffusions have been reported, it is generally accepted that the relative diffusivity of the grain boundary to that of the grain lattice increases with decreasing temperature. Because the LWR fuel temperature is in the middle range, from 1000 to 1700 °C during reactor operation, the diffusivities ratio α may not be very small, but be similar to the grain radius a. Thus, the role of the grain boundary may not be negligible under LWR operating conditions.

Recent evaluation of the currently available experimental data supports that in some reactor operation environments, the grain boundary can play a significant role in the fission gas transport process. Lately, Olander and Uffelen made a careful estimation of these diffusivities based on their combined grain boundary and lattice diffusion analysis [8]. They reported that $D_v/D_{\mathrm{gb}} \cong 10^{-2}$–$10^{-6}$ in the temperature range from 1000 to 1700 °C. Because α is a relative ratio, the 'effective' in the original definition can be dropped and thus their result can be directly applied to the current discussion. Kim experimentally investigated the lattice diffusion coefficients of xenon gas both in a sintered UO_2 and in a single crystal urania under the various oxygen potentials. He also estimated D_v/D_{gb} based on their model, which turns out to be about 10^{-3} [32].

In this case, the parameter ranges from 10^{-2} to 1 since the grain radius of commercial fuel pellets never exceeds 10^{-3} cm. This means that grain boundary trapping can suppress the fractional release down to $10^{-4} \sim 1$, unless grain boundary diffusivity is very high. Fig. 4 shows the suppression of the release fraction as a function of α when the practical grain size a is considered. As expected, the trapping of fission gas atoms in the grain boundary makes the diffusivity ratio α similar to a by retarding the inter-granular diffusion, thus increasing the ratio. In this case, the release must be less

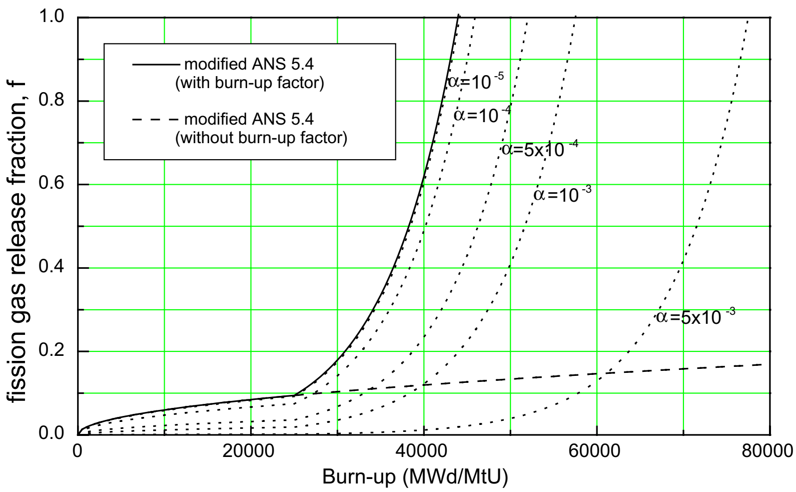

Fig. 4. Suppression of the fractional fission gas release as a function of α based on the two-stage parameter. Comparison is made against the modified ANS5.4 model with burn-up enhancement factor, $100^{\mathrm{Max}(0,\mathrm{Bu}-25\,000)/21\,000}$.

than that predicted without the grain boundary. This is in good agreement with the in-pile experiences that fission gases are far more mobile in single crystal than in polycrystalline UO_2 [33,34].

Most fission gas release measurements on irradiated PWR fuel rods demonstrate a threshold burn-up of about 20 GWd/MtU, depending on the power history. This observation is consensually explained in the following way: during the incubation period all gas atoms arriving at the grain boundaries are trapped, inter-granular bubbles nucleate and grow in the boundaries, they interlink themselves on the grain surfaces and at the edges, and eventually the inter-linkage triggers the fission gas release. Despite these findings, none of the prediction models, mechanistic or empirical, include the grain boundary diffusion process as a release mechanism. Instead, most of them impose a burn-up enhancement factor in their lattice diffusion coefficients to make up for the discrepancy.

In the current two-stage model, the inter-granular high burn-up behavior of the fission gasses can be directly reflected in the two-stage parameter, more specifically in the diffusivities ratio α in terms of burn-up dependence. During the incubation period, grain boundary diffusivity is relatively low, so α is similar to the grain radius a, which suppresses the fission gas release. However, once inter-granular bubbles begin to interlink the fission gas atoms trapped in the grain boundaries become more mobile, which increases the grain boundary diffusivity. Then the diffusivity ratio α decreases and the two-stage parameter increases from far less than unity to unity, leading to apparent burn-up enhancement of the fractional fission gas release.

5. Conclusions

A theoretical analysis of fission gas release in the two regions, grain lattice and grain boundary, is carried out in this study. Two simultaneous time-dependent balance equations for fission gas atoms are formulated, coupled with bubble trapping and resolution. The two equations are then reduced to a single partial differential equation with the incorporation of the relative diffusivity ratio α into a time-dependent surface boundary condition of the third kind, a combined Dirichlet and Neumann condition.

Laplace transform method with variable substitution technique produces the following solutions:

$$F \cong \frac{4}{\sqrt{\pi}}\left(\frac{a}{\alpha+a}\right)^2\left(\frac{D_v^{\mathrm{eff}}t}{a^2}\right)^{1/2} - \frac{3}{2}\left(\frac{a}{\alpha+a}\right)\left(\frac{D_v^{\mathrm{eff}}t}{a^2}\right)$$

for the in-pile case and

$$F \cong \frac{6}{\sqrt{\pi}}\left(\frac{a}{\alpha+a}\right)^2\left(\frac{D_v^{\mathrm{eff}}t}{a^2}\right)^{1/2} - 3\left(\frac{a}{\alpha+a}\right)\left(\frac{D_v^{\mathrm{eff}}t}{a^2}\right)$$

for the PIE case, respectively, where α is the relative diffusivities ratio, defined as $\frac{2}{\delta\beta_1^2}\frac{D_v^{\mathrm{eff}}}{D_{gb}^{\mathrm{eff}}}$. As seen in the equations, the solutions successfully explain the role and the effect of the grain boundary in terms of a new dimensionless parameter $a/(\alpha+a)$, a so-called two-stage parameter.

The new parameter reduces to unity only when α is much less than a, that is, when grain size is relatively large compared with the low grain lattice diffusivity or when grain boundary diffusivity is very much greater than grain lattice diffusivity. The latter corresponds to the case formulated in the simple Booth model. When α becomes similar to or greater than grain radius a, the solutions show that the grain boundary must significantly affect the whole fission gas release process, because the square of the parameter appears in the dominant first term of the prediction expression.

In fact, grain radius a is a geometric constant, typically 5–10 μm. However, α the ratio of grain lattice over grain boundary diffusivity, α is a variable transport property that is strongly dependent on both the fission gas species and the temperature and the burn-up. This means that the role of the grain boundary, in practice, depends on the ratio α, which is basically D_v^{eff}/D_{gb}^{eff} with the unit of centimeter. A recent report has shown that $D_v/D_{gb} \cong 10^{-2}$–10^{-6} in the temperature range from 1000 to 1700 °C. Therefore, the two-stage parameter reveals that the grain boundary can play significant roles in the fission gas release, depending on the fuel power history, because LWR fuel temperature is in the above temperature range during reactor operation.

Many measurements of in-pile fission gas release show a threshold burn-up of about 20 GWd/MtU. It is also well known that the growth of inter-granular bubbles and their linkage begins to trigger the release of trapped fission gases in the grain boundary when fuel burn-up exceeds the threshold burn-up. The introduction of the two-stage parameter, more specifically the burn-up dependent diffusivities ratio α, can successfully explain the inter-granular bubble behavior in high burn-up fuels. During the incubation period, grain boundary diffusivity is relatively low, so α is similar to grain radius a. This suppresses the fission gas release. However, once inter-granular bubbles begin to interlink, the fission gas atoms trapped in the grain boundaries become much more mobile, thus increasing the grain boundary diffusivity. Then the diffusivity ratio α decreases and thus the two-stage parameter increases from far less than unity to unity. This induces the high burn-up enhancement of the fission gas release.

Acknowledgements

This work is supported by Hanyang University, Seoul, Korea.

Appendix A

In the in-pile case, the fission gas generation term is not zero, as seen in Eq. (2), so must be included in the dimensionless transformation. When the dimensionless variable $u = RC + \frac{\beta R^3}{6D_v^{eff}}$ is introduced, Eq. (2) can be rewritten as

$$\frac{\partial u}{\partial t} = D_v^{eff}\frac{\partial^2 u}{\partial R^2}. \tag{A.1}$$

The initial and the boundary conditions are accordingly rewritten as $u(R,0) = \frac{\beta R^3}{6D_v^{eff}}$, $u(0,t) = 0$, and

$$\frac{\alpha}{a}\frac{\partial u}{\partial R}\bigg|_{R=a} - \frac{\alpha + a}{a^2}u\bigg|_{R=a} + (a - 2\alpha)\frac{\beta a}{6D_v^{eff}} = 0,$$

respectively.

Now the Laplace transform method is preferred for the solution of Eq. (A.1) with these initial and boundary conditions, because another crucial dimensionless parameter, $D_v^{eff}t/a^2$, is far less than unity under typical reactor operating conditions.

On taking the transformation by means of $\tilde{u} = \int_0^\infty e^{-\omega t}u(r,t)\,dt$, Eq. (A.1) becomes

$$\omega\tilde{u} - \frac{\beta R^3}{6D_v^{eff}} = D_v^{eff}\frac{\partial^2\tilde{u}}{\partial R^2} \tag{A.2}$$

and the accompanying conditions are transformed into $\tilde{u}(0) = 0$ and

$$\frac{\alpha}{a}\left(\frac{\partial\tilde{u}}{\partial R}\right)_{R=a} - \frac{\alpha + a}{a^2}\tilde{u}\bigg|_{R=a} + (a - 2\alpha)\frac{\beta a}{6D_v^{eff}}\frac{1}{\omega} = 0.$$

Then, the solution of Eq. (A.2) is obtained

$$\tilde{u}(R) = A\exp\left(\sqrt{\frac{\omega}{D_v^{eff}}}R\right) + B\exp\left(-\sqrt{\frac{\omega}{D_v^{eff}}}R\right) + \frac{\beta}{6D_v^{eff}}\frac{R^3}{\omega} + \frac{\beta}{\omega^2}R \tag{A.3}$$

with the coefficients A and B:

$$A = -B = \frac{\beta}{\omega^2}\left[\frac{\alpha}{a}\sqrt{\frac{\omega}{D_v^{eff}}}\left(e^{\sqrt{\frac{\omega}{D_v^{eff}}}a} + e^{-\sqrt{\frac{\omega}{D_v^{eff}}}a}\right) - \frac{\alpha + a}{a^2}\left(e^{\sqrt{\frac{\omega}{D_v^{eff}}}a} - e^{-\sqrt{\frac{\omega}{D_v^{eff}}}a}\right)\right]^{-1}.$$

The flux of fission gas release can also be transformed, in the following way:

$$\widetilde{J} = -\frac{D_v^{eff}}{a}\left[\left(\frac{\partial\tilde{u}}{\partial R}\right)_{R=a} - \frac{\tilde{u}}{a}\bigg|_{R=a} - \frac{\beta a^2}{3D_v^{eff}}\frac{1}{\omega}\right]. \tag{A.4}$$

Substitution of Eq. (A.3) into Eq. (A.4) results in a fractional form of solution in which $\tanh\sqrt{\omega a/D_v^{eff}}$ appears in both denominator and numerator. As mentioned, because $\tau = D_v^{eff}t/a^2 \ll 0.1$, the Laplace coefficient ω becomes very large, thus $\tanh\sqrt{\omega a/D_v^{eff}}$ becomes close to unity.

Hence, taking the inverse Laplace transformation of Eq. (A.4) and using the following definition of fractional fission gas release:

$$F = \frac{4\pi a^2\int_0^t J\,dt'}{4/3\,\pi a^3\beta t} = \frac{3}{a\beta t}\int_0^t J\,dt' \tag{18}$$

the fractional fission gas release is finally obtained

$$F \cong \frac{4}{\sqrt{\pi}}\left(\frac{a}{\alpha+a}\right)^2\left(\frac{D_v^{\mathrm{eff}}t}{a^2}\right)^{1/2} - \frac{3}{2}\left(\frac{a}{\alpha+a}\right)\left(\frac{D_v^{\mathrm{eff}}t}{a^2}\right). \quad (19)$$

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