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Journal of Nuclear Materials 246 (1997) 223–231

journal of nuclear materials

On the fission gas release from oxide fuels during normal grain growth

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Received 5 February 1996; accepted 27 March 1997

Abstract

A mathematical formalism for calculating the fission gas release from oxide fuels considering an arbitrary distribution of fuel grain size with only zero boundary condition for gas diffusion at the grain boundary is proposed. It has also been proved that it becomes unnecessary to consider the grain volume distribution function for fission products diffusion when the grain boundary gas resolution is considered, if thermodynamic forces on grain boundaries are only time dependent. In order to highlight the effect of the normal grain growth on fission gas release from oxide fuels Hillert's and Lifshitz and Slyozov's theories have been selected. The last one was used to give an adequate treatment of normal grain growth for the diffusion-controlled grain boundary movement in oxide fuels. It has been shown that during the fuel irradiation, the asymptotic form of the grain volume distribution functions given by Hillert and Lifshitz and Slyozov models can be maintained but the grain growth rate constant becomes time dependent itself. Experimental results have been used to correlate the two theoretical models of normal grain growth to the fission gas release from oxide fuels. © 1997 Elsevier Science B.V.

1. Introduction

As already pointed out by El-Saied and Olander [1], all published fission gas release models based on the Booth's sphere approximation, use, instead of the real distribution of the fuel grain sizes, a fixed number of equivalent average grain volume spheres from which the diffusion of fission gas at grain boundaries is calculated. Beyond their uninspired choosing of the coordinate system to describe the diffusion of the fission gas simultaneously with the grain growth, the Hillert's grain size distribution function [2], selected to exemplify the differences between the classical fission gas models and the new one, is only an idealized formulation of the time evolution of the grain size distribution that introduces the local curvature of the boundary as a measure of the global drift velocity of the grain boundary. Rather long range diffusion can be required for those cases of grain growth where impurities segregate to the moving boundary [4–6], as it is the case for the oxide fuels.

The structural changes affecting the fission gas release are influenced by both the as-fabricated fuel microstructure and the irradiation conditions. The out-of-pile tests on as-manufactured fuel reveals different grain growth kinetics for different manufacturers, mainly due to small fluctuations of the initial impurities content or the initial porosity [7].

Thus, in dissolved state,the impurities will retard grain growth through elastic attraction towards the open structure of the grain boundary [4]. The impurities,segregated at grain boundaries and the initial porosity, move along with the grain interfaces, hence reducing the rate of grain growth [8–10].

On the other hand, the degree of grain growth existing in a sample of irradiated UO_2 when annealed at high temperature is considerably smaller than that observed in an unirradiated UO_2 fuel sample subjected to the same annealing treatments [9].

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PII S0022-3115(97)00039-1

The abilities of grain boundaries to move during annealing of irradiated fuel is opposed by the increased concentration of impurities captured by the moving grain boundary, that lead to a progressive reduction of grain growth [9].

A similar effect is given by the intergranular gas bubbles [11]. Thus, they will exert a drag force on an advancing grain surface being swept along with the grain boundary [12].

Of particular interest, versus the grain morphology extensively studied in connection with various metal alloys and ceramics is the oxygen to metal ratio for the case of mixed oxide fuels [13].

This paper sets up the possibility of modelling the fission gas release from oxide fuels using an arbitrary grain volume distribution function when the resolution effect can be neglected. The Lifshitz and Slyozov's theory has been applied to the case of diffusion-controlled grain growth in oxide fuels versus the Hillert's model of grain growth.

2. Statement of the problem

It has been shown that in the diffusion regions bounded by moving boundaries where the moving law in time is known, the diffusion equation written for a time dependent coordinate system [15,16],

$$\frac{\mathrm{d}C_k}{\mathrm{d}t} = \nabla(D_k^a \nabla C_k) - C_k \nabla v^a + \sum_{j=1}^{r} \nu_{kj} b_j \qquad (1)$$

can be rewritten for an arbitrary fixed coordinate system using the Euler formula:

$$\nabla v^a = \frac{\mathrm{d}}{\mathrm{d}t} \ln J(x_0, t),$$

where $\nabla = \partial/\partial x =$ the divergence operator versus the coordinate system of the boundary velocity v^a, C_k is the concentration of the diffusing species k, D_k^a the diffusion coefficient in the coordinate system of the velocity v^a, ν_{kj} the stoichiometric number of species k in chemical reaction j, b_j the reaction rate of the chemical reaction j (per unit time) and $J(x_0, t)$ the Jacobian of the transformation from the time dependent coordinate system to a fixed one.

Assuming that the driving forces on the boundaries are only time dependent functions as it results from Ref. [16],

$$J(x_0, t) = J^a(t) = \prod_{j=1}^{3} a_j(t); \quad J^a(0) = 1,$$

where $a_1(t)$, $a_2(t)$, $a_3(t)$ are the components of the moving law in time of the moving boundary on the three directions.

Thus, for an arbitrary grain of a polycrystalline material, the diffusion Eq. (1) can be rewritten in the following form:

$$\frac{\partial u_k^a}{\partial t} = \nabla_{x_0} D'_k \nabla_{x_0} u_k^a(x_0, t) + \sum_{j=1}^{r} \nu_{kj} b'_j, \qquad (2)$$

where

$$u_k^a(x_0, t) = C_k(x_0, t) J^a(t) \qquad b'_j = b_k J^a(t)$$
$$D'_k = a_i^{-1}(t) D_k^{ij} a_j^{-1}(t). \qquad (3)$$

The thermodynamic requirements of only-time dependent forces on the grain boundaries imply the same concentration of species k everywhere at the grain boundary. Thus, if we accept some resolution (thermal or irradiation induced), the boundary concentration of species 'k' is only a time dependent function, $C_k(a, t) = \phi(t)$, and, consequently, based on substitutions Eq. (3),

$$u_k^a(a_0, t) = \phi(t) J^a(t). \qquad (4)$$

If concentration gradients arise on the grain boundaries, the excess of gas concentration will migrate easily along the grain boundary or will be trapped by the intergranular bubbles. For this case the boundary condition can be defined as [17]

$$\phi(t) = b n_s(t);$$
$$n_s(t) = \frac{N_1^a(t)}{S^a(t)} = \frac{\int_0^\infty g(V^a) N_1^a \mathrm{d}V^a}{1/2 \int_0^\infty \sigma^a g(V^a) \mathrm{d}V^a}, \qquad (5)$$

where b is the resolution parameter (m^{-1}) defined by Speight [17], n_s the surface density of the gas atoms, $g(V^a)$ the grain volume distribution function normalized to the unit volume of the polycrystal, N_1^a the total number of gas atoms arrived at the boundary of the arbitrary grain of radius 'a' by both sweeping and diffusion processes up to the time t and σ^a the grain area. The factor (1/2) has been introduced in Eq. (5) to take into account that the separation surface is bounded in every point by two neighboring grains. The index 'a' has been introduced to link the solution of the diffusion Eq. (2) to the grain volume distribution function.

It follows from Eq. (5) that, for a continuous grain volume distribution function, if the solution of Eq. (2) along with Eq. (4) can be calculated for an arbitrary grain of the polycrystal then, at any time, the total amount of species k arrived at grain boundary by both diffusion and sweeping processes, can be obtained by multiplying $n_s(t)$ to the total surface between the fuel grains.

Hence, if the exact solution of Eq. (2) along with the boundary condition (Eq. (4)) is known for the average volume grains of a polycrystal, then the total amount of species k arrived at the boundary of the average volume grain divided by its surface and multiplied to the total surface between the fuel grains in the unit volume will give the amount of the diffusing species k arrived at the grain boundary inside of the unit volume of the polycrystal. Or, in other words, up to a constant, using the grain growth law of the average grain size of the polycrystalline material, the solution derived in Ref. [19] can be accepted to be representative for any arbitrary distribution of grain sizes.

The particular case of the Booth's sphere model (perfect sink boundary condition for gas diffusion) does not require the same surface concentration between the fuel grains and thus the grains of the polycrystal can be treated as isolated spheres which grow or shrink simultaneously with fission products generation and diffusion outside the grains. Then, the boundary condition for Eq. (2) becomes $C_k(a(t), t) = 0$ and at any time the total amount of species k which remain inside of an arbitrary grain is

$$N_k^a(t) = \int_{V^a(t)} C_k(x, t)\,\mathrm{d}x = \int_{V_0^a} u_k^a(x_0, t)\,\mathrm{d}x_0. \tag{6}$$

The total amount of species k inside of the fuel grains from the unit volume of the polycrystal is given by

$$N_k(t) = \int_0^\infty g(V^a) N_k^a(t)\,\mathrm{d}V^a \cong \sum_{i \geq 1} g(V^{a_i}) N_k^{a_i} \Delta V^i. \tag{7}$$

The total amount of species k generated inside of an arbitrary grain up to the time t is given by [16]

$$Q_k^a(t) = \int_0^t \int_{V^a(t)} q_k(s)\,\mathrm{d}x\,\mathrm{d}s; \quad q_k(s) = \sum_{j=1}^{r} \nu_{kj} b_j \tag{8}$$

and the total amount of species k generated inside the unit volume is

$$Q^T(t) = 1 \int_0^t q_k(s)\,\mathrm{d}s. \tag{9}$$

Now, the total amount of the diffusing species k arrived at grain boundary by only the diffusion mechanism is

$$\begin{aligned} N_D^1(t) &= \int_0^\infty g(V^a)(Q_k^a - N_k^a)\,\mathrm{d}V^a \\ &\cong \sum_{i \geq 1} g(V^{a_i})(Q_k^{a_i} - N_k^{a_i})\Delta V^i \end{aligned} \tag{10}$$

and by diffusion and sweeping

$$N_T^1(t) = Q^T(t) - N_k(t). \tag{11}$$

Formal solutions of Eq. (6) have already been given for both stable species [18–21] and radioactive products [16] and they can be extended for a polycrystal which is characterized by a grain volume distribution function using Eqs. (7), (9) and (11).

Before dealing with the application of the above described method, a brief description of the analytical grain volume distribution functions, their fundamental aspects and their limits is necessary.

3. Some observations about the normal grain growth

It is generally accepted that in a condensed matter the grains of a new phase will nucleate and grow until a quasistationary state, so-called normal grain growth, is reached [2–4,14,22–25]. After this first stage the grains will grow as a result of the reduction in grain boundary energy with only few changes of the polycrystal volume.

Experimental data have often been interpreted in terms of a log-normal distribution [22,23] of the grain sizes and faces. Accepting this distribution, Feltham was able to treat the normal grain growth as an uninvariant statistical problem in which the state of grain growth is obtained as a result of the surface tension-controlled rate of growth of the individual grains in the distribution [22].

In their theory on the kinetics of precipitation from supersaturated solid solutions, Lifshitz and Slyozov have shown that during the first stage the grains grow until the degree of supersaturation has so fallen that the critical radius (the equilibrium radius between the condensed matter and the new phase grains) caught up with the mean grain size [14]. The second stage is considered when the grains have reached an appreciable size and the supersaturation of the matrix can be neglected. They describe the second process as coalescence (the growth of the large grains at the expense of the smaller ones [2]).

With only few changes, their theory of grain growth can be applied for spatial systems where the long range diffusion along the grain boundaries cannot be neglected. In fact, the main assumption is already known as 'one grain model' [3,24–26], and it consists of considering the behavior of only one grain with the grain boundary energy of all the other grains smeared out throughout the system. The method has been applied by Saetre et al. [26] in order to improve the Hillert model [2] that will be discussed later.

The Lifshitz and Slyozov's theory rigorously proves that there is an asymptotic grain size distribution as a result of diffusion. The second order effects such as the grain shape, crystalline order and the elastic strain that result from the difference between the specific volume of the grains and the matrix do not alter the asymptotic grain distribution function nor influence its stability [14].

According to the latter observation, it can be proved that the diffusion approximation for grain growth can be maintained for the case of grain growth from oxide fuels during the fuel irradiation. Taking into account that in the second stage of grain growth the degree of supersaturation is small, then from the definition of the critical radius a_c, and from the condition of conservation of matter flows along the boundary of an arbitrary grain of radius 'a' it follows [14]

$$\frac{\mathrm{d}a}{\mathrm{d}t} = \frac{\alpha D_{\mathrm{eff}}}{a}\left(\frac{1}{a_c} - \frac{1}{a}\right) \tag{12}$$

$$\alpha = \frac{2\gamma_{\mathrm{gb}} \Omega' C_0}{kT},$$

where γ_{gb} is the grain boundary surface tension, Ω' the atomic volume associated to the grain boundary, $\Omega' - \Omega$, k the Boltzmann constant and C_0 the equilibrium concentration of atoms of the polycrystalline material. The effective diffusion coefficient, D_{eff} has been introduced in order to take into account the impurities segregated at grain boundaries.

In terms of reduced volume, $z = a^3/a_c^3$ and the dimensionless time $\tau = \ln(a_c/a_{c0})^3$, where a_{c0} is the initial critical radius, it follows from the conditions of the time-invariance of the grain volume distribution function in their theory, that the volume of an arbitrary grain varies in time as

$$\frac{dz}{d\tau} = -\left(z^{1/3} - \frac{3}{2}\right)^2 (z^{1/3} + 3). \tag{13}$$

Then, by integration

$$\frac{x}{x_0} = \frac{(3/2 - u_0)^{5/9}(u_0 + 3)^{4/9}}{(3/2 - u)^{5/9}(u + 3)^{4/9}} \times \exp\left(\frac{3}{3 - 2u_0} - \frac{3}{3 - 2u}\right), \tag{14}$$

with $u = z^{1/3}$ and $x = a_c/a_{c0}$.

The grain volume distribution function as a time-dependent function resulted from the Lifshitz and Slyozov's theory has the following form [14],

$$\varphi(z, \tau) = n(\tau)p(z) \tag{15}$$

with

$$n(\tau) = \frac{3A}{4\pi \bar{a}_0^3} e^{-\tau}; \quad \bar{a}_0 = \text{the initial mean grain radius} \tag{16}$$

where $n(\tau)$ is the number of the fuel grains per unit volume, A is the normalization constant (in our case $A = 0.9009$) and

$$p(z) = \begin{cases} \dfrac{27}{\left[32(z^{1/3} + 3)^7 (3/2 - z^{1/3})^{11}\right]^{1/3}} \exp\left(1 - \dfrac{3}{3 - 2z^{1/3}}\right) & z < \dfrac{27}{8} \\ 0 & z \geq \dfrac{27}{8} \end{cases} \tag{17}$$

$$\left(\int_0^\infty p(z)\,dz = \int_0^{z_0} p(z)\,dz = 1\right).$$

$p(z)$ is the probability that a grain shall have a reduced volume between z and $z + dz$. Also, the corresponding average grain growth law is [14]

$$\bar{a}^3 = a_c^3 = \frac{8}{9} \frac{\gamma_{gb} \Omega' C_0 D_{eff}}{kT} t$$

or, for an arbitrary time step Δt,

$$\bar{a}^3 - \bar{a}_0^3 = K \Delta t, \tag{18}$$

where the rate constant K is given by

$$K = \frac{8}{9} \frac{\gamma_{gb} \Omega' C_0 D_{eff}}{kT}. \tag{19}$$

El-Saied and Olander [1] use in their model of fission gas release during normal grain growth the Hillert's grain volume distribution function [2]. As mentioned above, this idealized theory of normal grain growth has been criticized over the years because of the unphysical statement of the problem when the local curvatures of the grains are considered as driving forces of the grain volume changes [3]. In order to improve the physical meaning of the Hillert's model, Fischmeister and Grimvall [27] and also Saetre, Hundery and Ryum [26] introduce the concept of 'one grain model' and prove the similarity between the Hillert approach and the Lifshitz–Slyozov theory. From definition of the drift velocity, the rate of change of an arbitrary grain size 'a' is determined by the pressure, Δp on the grain boundary due to its curvature to eliminate the smeared out grain boundary energy, that in the one grain model is defined as [26]

$$\frac{da}{dt} = M \Delta P = M\sigma\left(\frac{1}{a_c} - \frac{1}{a}\right), \tag{20}$$

where M is the grain boundary mobility and σ is the specific grain boundary energy. In terms of reduced grain radius $u = a/a_c$ and the dimensionless time $\tau = \ln(a/a_c)^2$ it follows from the conditions of the time-invariance of the grain volume distribution function, that the radius of an arbitrary grain varies in time as

$$\frac{du}{d\tau} = -\frac{(2 - u)^2}{2u}. \tag{21}$$

The evolution of the grain boundary derived from the Hillert's theory of grain growth is

$$2\left[\frac{1}{2 - u_0} - \frac{1}{2 - u}\right] - \ln\left(\frac{2 - u}{2 - u_0}\right) = \frac{1}{2} \ln \frac{a_c}{a_{c0}}. \tag{22}$$

The grain volume distribution function as a time-dependent function derived by Hillert is

$$\chi(u, \tau) = n(\tau)h(u),$$

with

$$n(\tau) = \frac{3B}{4\pi \bar{a}_0^3} e^{-3/2\tau}; \quad \bar{a}_0 = \text{the initial mean grain radius}$$

where $n(\tau)$ is the number of the fuel grains per unit volume, B the normalization constant (in our case $B = 1.0453$) and the probability function,

$$h(u) = \begin{cases} \dfrac{3u}{(2 - u)^5} (2e)^3 e^{-6/(2-u)} & u < 2 \\ 0 & u > 2 \end{cases}. \tag{23}$$

The moving law in time of the mean grain radius corresponding to Eq. (23) is

$$\bar{a}^2 - \bar{a}_0^2 = K\Delta t, \tag{24}$$

$$K = \tfrac{81}{64} M\sigma. \tag{25}$$

Besides the theories about the above mentioned normal grain growth, the problem of grain growth during oxide fuel irradiation is more complex due to the impurities or the fission gas bubbles which occur as a result of the production of the fission products. In fact, because the fraction of metal-oxide atoms at grain boundaries is extremely small when compared to that of bulk atoms, very small amounts of impurities may completely dominate grain growth phenomena because of solute segregation [6]. To take into account this aspect, an effective diffusion coefficient has been used to describe the rate of arbitrary grain radius changes in Eq. (12). It can fluctuate in time as a result of the retarding effect on grain boundaries induced by either intergranular gas bubble nucleation and growing or by the solid fission products, but the mathematical formalism associated to the theory of normal grain growth allows a distinct formulation of the problem. Because of the canonical form of the grain volume distribution function, a dimensionless time has been involved in both Lifshitz and Slyozov's and fellert's theories on the normal grain growth, and the retarding effect induced by impurities can be accounted for by a separate treatment of the grain growth constant K (Eq. (19)) as a time dependent function. According to the latter observation Eqs. (18) and (24) can be written in the more general form,

$$\bar{a}^m - \bar{a}_0^m = K'(t)\Delta t, \tag{26}$$

where $m = 2$ for Hillert's model and $m = 3$ for Lifshitz and Slyozov's model,

$$K'(t) = KF(t) \tag{27}$$

and the rate constant K that corresponds to Eqs. (19) and (25) can be defined as [4]

$$K = \frac{k_0}{T} \exp\left(-\frac{Q_s}{T}\right), \tag{28}$$

where Q_s is the activation energy for the coarsening process and k_0 is a kinetic constant.

The function $F(t)$ must be linked to the drag forces exerted by impurities and for the case when the grain growth is completely stopped, $F(t) = 0$. It results from the above treatment of the rate constant $K'(t)$ that even if the solid fission products migrate and precipitate or intergranular bubbles nucleate and grow on the grain boundaries, the grain volume distribution function remains invariant during the fuel irradiation. Similar functions have already been used [8,10–12,28,29], to consider the retarding effect induced by intergranular gas bubbles on the grain boundary.

4. Application

As mentioned by Andersen and Grong [4], the normal grain growth in metals and alloys is a diffusion controlled process driven by the reduction in grain boundary energy. Under heat treatment conditions the normal grain growth is well described by the following empirical equation:

$$\bar{a}^m - \bar{a}_0^m = k_0 \exp\left(-\frac{Q_{\text{app}}}{T}\right) t, \tag{29}$$

where 'a', 'a_0', are the current and initial mean grain radius, 'm' the time exponent, Q_{app} the apparent activation energy of grain growth and k_0 a kinetic constant. For most metals and alloys, the time exponent, m, in Eq. (29) varies typically between 2.5 and 10.0 due to the drag forces exerted by impurity elements in solid solution. Only in case of ultrapure metals annealed at very high temperatures, the time exponent may approach a constant value of 2, that corresponds to the limiting case where the migration rate is directly proportional to the driving pressure [4]. Taking $F(t) = 1$ in Eq. (27) the similarity between Eqs. (26)–(28) and Eq. (29) is obvious.

As already shown [18], the specific grain growth laws for every fuel manufacturer must be used when the effect of the grain growth on the fission gas release (FGR) from oxide fuels is studied. In order to highlight the effect of a theoretical grain size distribution function on FGR according to Eq. (29), both Hillert's and the Lifshitz and Slyozov's models have been calibrated with the experimental measurements of the average grain size from [7]. For simplicity, the least squared method has been used to compute the kinetic constants and the following results have been obtained:

$$m = 2; \quad k_0 = 0.41 \times 10^{-6}\ \text{m}^2/\text{s}; \quad Q = 350\ \text{kJ}, \tag{30}$$

$$m = 3; \quad k_0 = 0.14 \times 10^{-8}\ \text{m}^3/\text{s}; \quad Q = 411\ \text{kJ}. \tag{31}$$

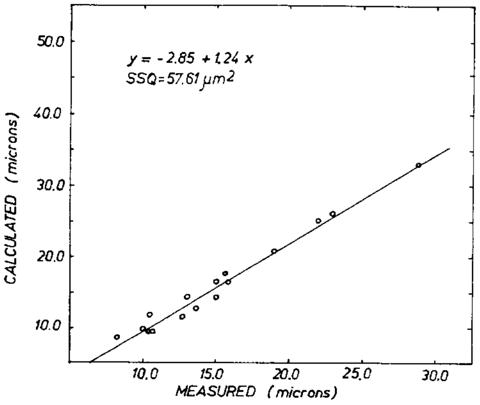

Fig. 1. The regression line (calculated versus measured values) for a power type grain growth law ($m = 3$) according to Lifshitz and Slyzov's model for normal grain growth.

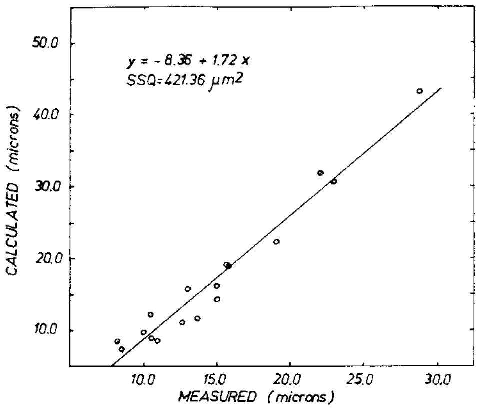

Fig. 2. The regression line (calculated versus measured values) for a power type grain growth law ($m = 2$) according to Hillert's model for normal grain growth.

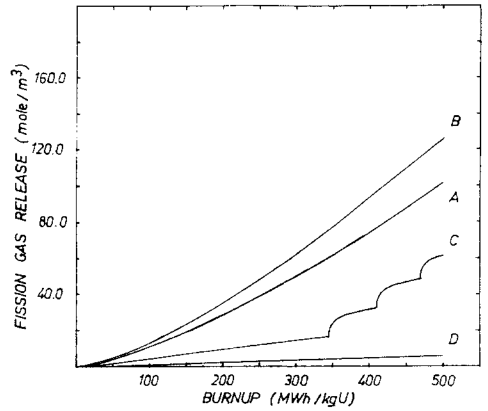

Fig. 4. Fission gas release per unit volume for a constant 1200°C temperature history, using Hillert's model for normal grain growth (A), and the approximation of diffusion from the average volume grain [19] of the same grain growth law ($m = 2$) with the correction parameters, $b = 0$ (B), $b = 10$ (C), and $b = 50$ (D).

The results obtained for $m = 3$ (Eq. (31)) are slightly different than the same results previously calculated because of the more accurate method used in [7], but our interest has been focused to only point out the effect of the two normal grain growth models on FGR when the real grain volume distribution function is modeled. Also the time dependence of the grain growth rate (Eq. (27)) has been neglected. Further analyses regarding the retarding effect induced by the intergranular fission gas bubbles and the metallic precipitates on grain boundary are necessary.

The regression lines given in Figs. 1 and 2 show very different slopes versus the first bisectrix ($y = x$) and with closer fit for $m = 3$. It results that a larger overestimation of the average grain size could be expected at higher values of the grain size (long times) for $m = 2$. Both the physical state of starting ammonium diuranate precipitate and the sintering parameters (levels of the fuel temperature and time) will profoundly influence growth of the resulting UO_2 grains [30]. This explains the closer result of the linear regression from Fig. 1.

As mentioned above, the perfect sink case (zero boundary condition) for Eq. (2) requires FGR analyses from a distribution of fuel grains, so that Eqs. (7), (14) and (22) for fission gas diffusion simultaneously with an arbitrary grain volume change have been applied for a finite number of grain size classes following both Hillert's and the Lifshitz and Slyozov's grain volume distribution functions (Eqs. (23) and (17)). Ten to twenty grain size classes are

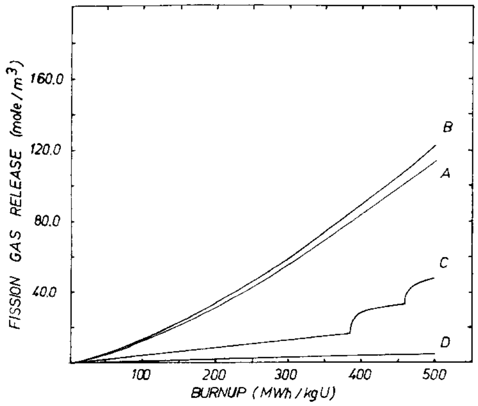

Fig. 3. Fission gas release per unit volume for a constant 1200°C temperature history, using Lifshitz and Slyzov's model for normal grain growth (A), and the approximation of diffusion from the average volume grain [19] of the same grain growth law ($m = 3$) with the correction parameters, $b = 0$ (B), $b = 10$ (C), and $b = 50$ (D).

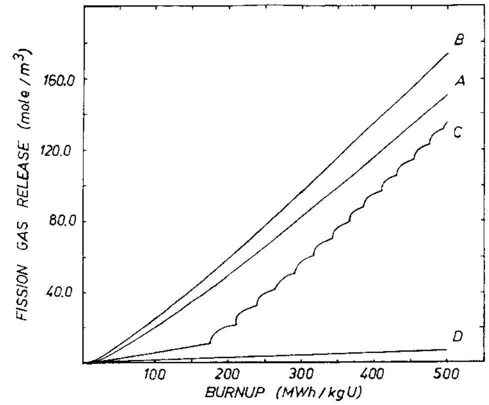

Fig. 5. Fission gas release per unit volume for a constant 1400°C temperature history, using Lifshitz and Slyzov's model for normal grain growth (A), and the approximation of diffusion from the average volume grain [19] of the same grain growth law ($m = 3$) with the correction parameters, $b = 0$ (B), $b = 10$ (C), and $b = 50$ (D).

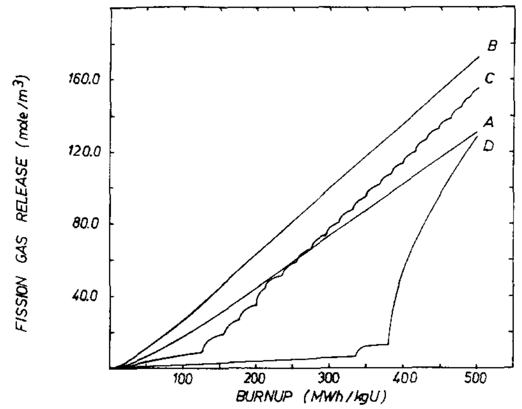

Fig. 6. Fission gas release per unit volume for a constant 1400°C temperature history, using Hillert's model for normal grain growth (A), and the approximation of diffusion from the average volume grain [19] of the same grain growth law ($m = 2$) with the correction parameters, $b = 0$ (B), $b = 10$ (C), and $b = 50$ (D).

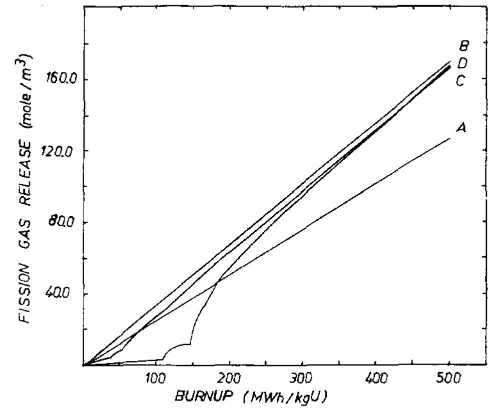

Fig. 8. Fission gas release per unit volume for a constant 1600°C temperature history, using Hillert's model for normal grain growth (A), and the approximation of diffusion from the average volume grain [19] of the same grain growth law ($m = 2$) with the correction parameters, $b = 0$ (B), $b = 10$ (C), and $b = 50$ (D).

enough to accurately describe the gas migration (diffusion and sweeping) for the two theoretical distribution functions during all the power histories analyzed (Figs. 3–12). The solutions of the diffusion Eq. 2 along with the boundary condition (Eq. 4) calculated in [20] and [19] have been used. In order to illustrate only the normal grain growth effect on fission gas release the linear power density has been fixed at 60 kW/m. Using a number of constant temperature histories (Figs. 3–8) corresponding to the equiaxed grain growth, comparative calculations of FGR have been performed both for the above mentioned distribution functions and for the generalized solution of diffusion equation simultaneously with the grain growth and considering the resolution effect from [19]. The 'correction parameter b' for gas resolution has been changed between 10 and 50 in order to illustrate the weight of gas resolution versus the normal grain growth laws used. All the other calculating methods and parameters for intergranular bubble growth and interlinkage are the same as in [19].

The laws of normal grain growth for $m = 2$ and $m = 3$ (Eqs. (22) and (14)) along with the diffusion coefficient given in Ref. [31] have been used for the generalized solution [19] and Eq. (7). For a better understanding of the comparative studies presented in Figs. 3–12, the curves corresponding to the generalized solution [19] cumulate both the fission gas vented out in the void volume of the fuel element and the gas present inside the intergranular bubbles at any time. This amount is equivalent to the

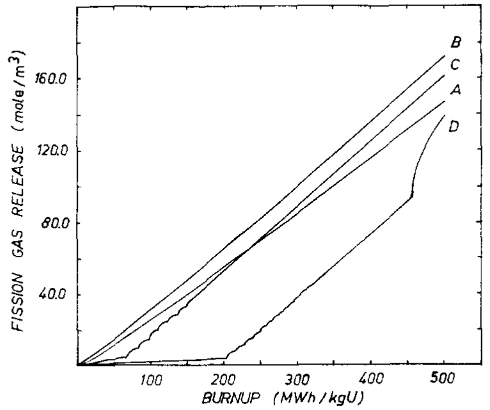

Fig. 7. Fission gas release per unit volume for a constant 1600°C temperature history, using Lifshitz and Slyzov's model for normal grain growth (A), and the approximation of diffusion from the average volume grain [19] of the same grain growth law ($m = 3$) with the correction parameters, $b = 0$ (B), $b = 10$ (C), and $b = 50$ (D).

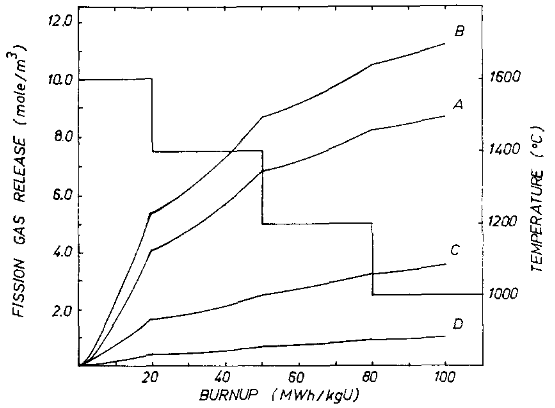

Fig. 9. Fission gas release per unit volume for a decreasing temperature history, using Lifshitz and Slyzov's model for normal grain growth (A), and the approximation of diffusion from the average volume grain [19] of the same grain growth law ($m = 3$) with the correction parameters, $b = 0$ (B), $b = 10$ (C), and $b = 50$ (D).

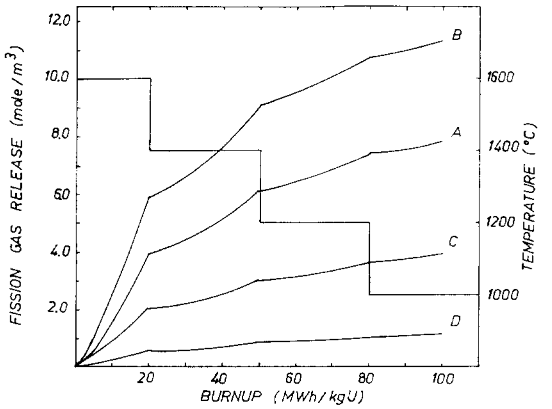

Fig. 10. Fission gas release per unit volume for a decreasing temperature history, using Hillert's model for normal grain growth (A), and the approximation of diffusion from the average volume grain [19] of the same grain growth law ($m = 2$) with the correction parameters, $b = 0$ (B), $b = 10$ (C), and $b = 50$ (D).

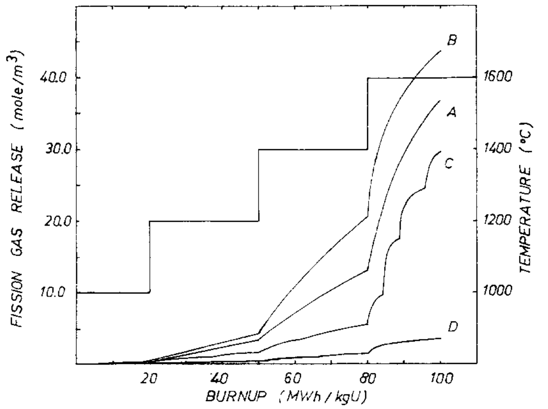

Fig. 12. Fission gas release per unit volume for an increasing temperature history, using Hillert's model for normal grain growth (A), and the approximation of diffusion from the average volume grain [19] of the same grain growth law ($m = 2$) with the correction parameters, $b = 0$ (B), $b = 10$ (C) and $b = 50$ (D).

results depicted using the two grain size distribution functions for the case of zero grain-boundary condition associated to Eq. (2). As it can be seen from Figs. 3–8 the amount of the fission gas release predicted by using the two distribution functions for the grain volume changes in the unit volume of the oxide fuel is always smaller than for the 'classical' average grain size model and zero boundary condition (curves A and B). When the resolution effect is accounted for (curves C and D) the slopes will drastically change with the value of the 'correction parameter' and this is more evident for the case of the Lifshitz and Slyozov model. Because of the approximation of the average grain size used for the curves B, the differences between the two models and between the curves A and B increase with the fuel temperature. When the resolution effect is accounted for, all the curves B, C and D arrive approximately to the same point in the case of $m = 2$ and 1600°C fuel temperature. As supposed in [19], when the time of the intergranular bubble interlinkage becomes smaller as the smallest time step chosen for calculation (in our case 1 MWh/kg U), a continuous network of interconnected tunnels can arise at the grain boundaries and the grain boundary condition would change correspondingly to the perfect sink one. This case can also be followed in Figs. 6–8 (curve D).

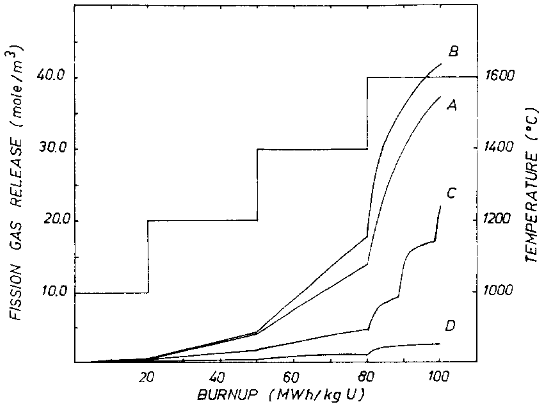

Fig. 11. Fission gas release per unit volume for an increasing temperature history, using Lifshitz and Slyzov's model for normal grain growth (A), and the approximation of diffusion from the average volume grain [19] of the same grain growth law ($m = 3$) with the correction parameters, $b = 0$ (B), $b = 10$ (C), and $b = 50$ (D).

The effect of the temperature changes during irradiation both for fission gas release models described in Ref. [19] and for the case when normal grain growth is considered are presented in Figs. 9–12. As it can be seen from these figures, the differences in FGR predicted between the zero grain boundary condition using the model described in Ref. [19] and the theoretical distribution function are maintained, with larger differences for Hillert's treatment of the normal grain growth. The result is expected from the above discussion about the growth parameters of the average grain size: larger grains are expected at longer times and higher temperatures so that these differences can be explained by the increased effect of the average grain size approximation versus a theoretical distribution of the fuel grains. However, when the resolution effect is considered, the grain size distribution effect on FGR can be neglected.

It must be mentioned that the fission gas release predictions are closely related to the average grain size predicted during grain growth. Thus, for both constant temperature histories (Figs. 3–8) and for time-varying temperature histories (Figs. 9–12) closer results for FGR are predicted for the smaller increased grains (the Lifshitz and Slyozov's model for normal grain growth versus Ref. [19]– curves A and B). Also, at lower temperatures, when both fission gas diffusion and gas sweeping due to grain growth are smaller, closer predictions of fission gas release are given in the perfect sink case for normal grain growth (curve A), versus the generalized solution for FGR from Ref. [19] (curve B).

According to the results presented above, when the fission gas resolution at the grain boundary can be neglected (as is the case of Xe and Kr species during the out-of pile annealing experiments) a careful selection of the normal grain growth models is required in order to satisfactorily describe fission gas migration at grain boundaries simultaneously with the grain growth. Otherwise, when the irradiation induced resolution is accounted for, the FGR models based only on the specific grain growth laws for every fuel manufacturer describe more accurately the volatile fission products evolution during irradiation [19].

5. Conclusions

Both as technological process of fabrication (sintering from UO_2 powders) and as a result of irradiation, the normal grain growth in oxide fuels is closer to the zero approximation of the grain size distribution function given by the Lifshitz and Slyozov theory (long range diffusion approximation) than the Hillert theory. Because of the solid fission products and intergranular fission gas bubbles, the as-supposed constant of grain growth would change in time and it has been so written that the normal grain growth can be correlated to the previously developed grain growth models which treat the irradiation effect.

As a result of fission products resolution, the thermodynamic requirement of only-time dependent forces on the grain boundaries in normal grain growth, involve the same concentration of fission gas on the boundaries. It has been proved that this condition permits to evaluate fission gas release from only a single class of the fuel grains and up to a constant (given by the surface to volume grain ratio) the diffusion from the equivalent sphere associated to the average volume grain is a good approximation of the real processes involved.

Comparative analyses of fission gas release for Hillert's and Lifshitz and Slyozov's models show large differences of FGR predictions between the FGR model based on the two theories of normal grain growth and the approximation of diffusion from a fixed numbers of equivalent average grain volume spheres. The FGR predictions based on Lifshitz and Slyozov's theory for normal grain growth and the average volume grain approximation of the same grain growth law (time exponent $m = 3$) are smaller, mainly because of the lower grain sizes predicted at the end of irradiation.

When the FGR results of the out-of-pile annealing tests are simulated the mathematical formalism presented can be used for better descriptions of FGR at the grain boundaries of the fuel samples.

Acknowledgements

This paper has been carried out under an International Atomic Energy Agency research contract. The authors would like to acknowledge to the IAEA Division of Nuclear Fuel Cycle and Waste Management for helpful technical advises and the IAEA Department of Research and Isotopes for prompt administrative assistance of the research. We would like also to thank to Dr O. Budan from RPA-NPG (Romanian Power Authority-Nuclear Power Group) for kindly assistance and to Dr I. Furtuna from INR for helpful comments of this work.

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