Tracer Concentration Contours in Grain Lattice and Grain Boundary Diffusion

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Abstract

Grain boundary diffusion plays a significant role in fission gas release, which is one of the crucial processes dominating nuclear fuel performance. Gaseous fission products such as Xe and Kr generated during nuclear fission have to diffuse in the grain lattice and the boundary inside fuel pellets before they reach the open spaces in a fuel rod. These processes can be studied by 'tracer diffusion' techniques, by which grain boundary diffusivity can be estimated and directly used for low burn-up fission gas release analysis. However, only a few models accounting for the both processes are available and mostly handle them numerically due to mathematical complexity. Also the numerical solution has limitations in a practical use. In this paper, an approximate analytical solution in case of stationary grain boundary in a polycrystalline solid is developed for the tracer diffusion techniques. This closed-form solution is compared to available exact and numerical solutions and it turns out that it makes computation not only greatly easier but also more accurate than previous models. It can be applied to theoretical modelings for low burn-up fission gas release phenomena and experimental analyses as well, especially for PIE (post irradiation examination).

1. Introduction

Understanding the release mechanism of fission gases inside UO₂ fuel pellets during reactor operation is of great technical importance for the achievement of reliable high fuel performance. Build-up of the fission gases in a gap between fuel pellets and cladding not only impairs the heat transfer from fuel to coolant but also increases the internal gas pressure of the fuel rod. Corrosive fission products such as iodine may chemically attack the inner surface of the fuel cladding, thus, assist to breach it. Despite the great importance, in actual, complex and uncertain mechanism of the fission gas release imposes undesirably higher operational margin than needed, which has to be eliminated for higher performance operation. For these reasons the behavior of the fission gases in the irradiated UO₂ pellet has been extensively studied by

many researchers, either in-pile measurements or post-irradiation examinations.

The quantitative analysis for the fission gas release rate prediction involves the lattice diffusion inside grains and the grain boundary diffusion through grain boundaries. Thus the combined effect of the two diffusivities is an important factor in the analysis of the fission gas release although it is believed that the grain boundary diffusivity is orders of magnitude greater than the lattice diffusivity.

The diffusion processes in the fission gas release can be studied by 'tracer diffusion' techniques. Diffusion processes in the methods follow just the reverse way of the diffusion processes in the fission gas release : tracer atoms from a open surface diffuse through grain boundaries and then diffuse again from the grain boundary to the matrix. The grain boundary diffusivity can be estimated by a quantitative diffusion analysis.

The mathematical analysis of the grain boundary diffusion process began with Fisher [1]. He studied the effect of coupling lattice diffusion and grain boundary diffusion on the tracer penetration into a solid. He treated the grain boundary as an isolated, thin semi-infinite slab embedded in the lattice perpendicular to the free surface and obtained the approximation solution. Whipple [2] later presented the numerical solution using Laplace-Fourier transformation for the same problem. Suzuoka considered the case of finite amount of diffusant source, which means that the concentration of the surface source may vary with time [3]. Subsequently these stationary boundary modeling works have been carried out diversely and in detail in many references [4-7]. However, most of them handle the problem numerically, whose solution techniques are limited in a practical use.

In the mean time, grains in a fuel pellet grow and shrink according to grain growth kinetics, especially at elevated temperature at which nuclear reactors are operating. Thus the boundary movement ascribed to the grain growth greatly influences the fission gas release rate by lengthening or shortening the lattice dif-

fusion distance, which is the rate limiting step. Sweeping fission gases by the moving boundary contributes to the increment of the fission gas release as well. However, even for a tracer diffusion analysis, taking both the intragranular grain growth and the diffusion processes simultaneously into consideration is not easy. Glaeser and Evans studied the effect of grain boundary migration on this coupled diffusion problem with the assumption that the grain boundary movement is extremely faster than tracer atoms diffusion in the grain lattice [8].

Recently, Olander and El-Saied [9] proposed a comprehensive model which accounts for both stationary and moving grain boundary during the tracer diffusion. Their numerical solution improves the computational accuracy and the capability of handling moving grain boundary problem as well. However, it is also a numerical solution, thus, requires a computer and a compatible numerical solvers since it contains the unsolved partial differential equations in the model.

Current study on this coupled grain lattice and grain boundary diffusion problem consists of two parts : one is stationary and the other moving grain boundary case. Here in this paper an approximate analytical solution for the stationary grain boundary diffusion case is developed by extending previous Olander and El-Saied's work. Resultant closed-form solution, instead of numerical solution, is compared to available exact and numerical solutions. Second part for the moving grain boundary case will be presented later in this journal.

2. Mathematical Formulation

Mathematical analysis of the moving grain boundary problem [1-4] in a tracer diffusion technique deals with the grain boundary as a single, semi-infinite thin slab of thickness δ intersecting the surface at a right angle. Diffusion species (tracer) on the surface is a radioactive isotope and completely soluble in the

lattice. Volume diffusion, grain boundary diffusion as well as grain boundary movement processes assist tracer deposition in a solid. Figure 1 shows the geometry and the processes considered in this formulation. Tracer atoms on the surface diffuse into a solid either by direct volume diffusion or by diffusion through the grain boundary followed by lateral volume diffusion from the boundary to the lattice. Since the grain boundary redeposits tracer behind when it moves, the tracer concentration ahead of the boundary is different from that behind it. Thus two lattice diffusion equations are needed to describe the lattice concentration distributions upstream and downstream of the moving boundary.

Taking the grain boundary as the origin of the coordinate x and the surface as the origin of y the tracer conservation equations are:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x} , \tag{1a}$$

$$\frac{\partial C'}{\partial t} = D \frac{\partial^2 C'}{\partial x'^2} + v \frac{\partial C'}{\partial x'} \tag{1b}$$

Tracer on the surface [$C_{gb}(0,t)=1$]

Fig. 1. **Geometry and Solute Diffusion Processes Involved in Tracer Deposition(slab model)**

where t is time and x and x′ are the lateral position ahead of and behind the moving grain boundary, respectively. C and C′ are the tracer concentrations also ahead of and behind the boundary, respectively. D is the lattice diffusion coefficient and v is the grain boundary velocity.

In equations (1a) and (1b), first terms represent normal Fickian diffusion, whereas second terms mean convective terms since the grain boundary taken as the frame of reference is moving with velocity v. Grain lattice diffusion is restricted to lateral direction parallel to the surface, thus, direct volume diffusion iny direction in the lattice is neglected. This assumption is valid at short annealing times and at depths at which solute supply to the lattice from the grain boundary is dominant.

Tracer concentration in the grain boundary, C_{gb}, is given by the balance equation:

$$\delta \frac{\delta C_{gb}}{\delta t} = \delta D_{gb} \frac{\partial^2 C_{gb}}{\partial y^2} + D \left[\left(\frac{\partial C}{\partial x} \right)_{x=0} + \left(\frac{\partial C'}{\partial x'} \right)_{x'=0} \right] \tag{2}$$

D_{gb} is the grain boundary diffusion coefficient

Equations (1) and (2) can be rewritten using following dimensionless parameters:

$$X = \frac{x}{E} , \qquad Y = \frac{y}{E} . \qquad \text{and } \tau = \frac{Dt}{E^2} .$$

For these parameters E is defined as $\frac{\delta D_{gb}}{2D}$. New equations are as follows:

$$\frac{\partial C}{\partial \tau} = \frac{\partial C^2}{\partial X^2} - A \frac{\partial C}{\partial X} \tag{3a}$$

$$\frac{\partial C'}{\partial \tau} = \frac{\partial^2 C'}{\partial X'^2} + A \frac{\partial C'}{\partial X'} \tag{3b}$$

for the diffusion in the grain lattice and

$$B \frac{\partial C_{gb}}{\partial \tau} = \frac{\partial^2 C_{gb}}{\partial Y^2} + \frac{1}{2} \left(\frac{\partial C}{\partial X} \right)_{X=0} + \frac{1}{2} \left(\frac{\partial C'}{\partial X'} \right)_{X'=0} \tag{4}$$
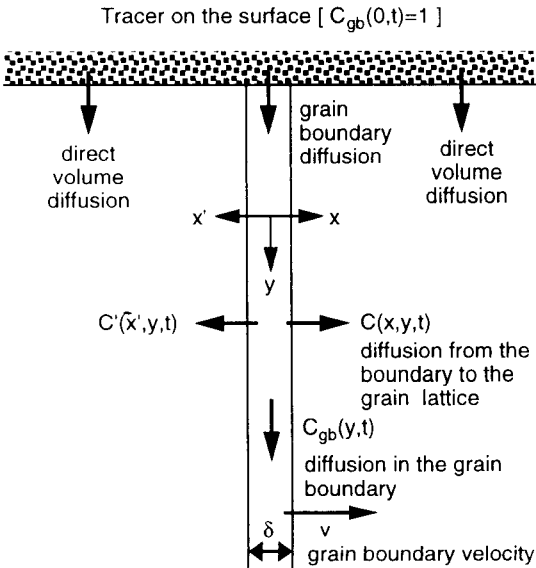
for the diffusion in the grain boundary. A is a dim-
ensionless grain boundary velocity and B is the ratio
of lattice and grain boundary diffusivities:

$$A = \frac{vE}{D} \, , \qquad\qquad B = \frac{D}{D_{gb}}$$

Because the grain boundary thickness δ is very
small compared to the distance of lateral penetration
of the tracer isotope, a very low ratio of D/D_{gb} is
needed to efficiently transport the tracer in the solid.
In general, the parameter B is in the order of 10^{-6}.
Thus the grain boundary storage capacity, $B\partial_{gb}/\partial\tau$, of
the tracer can be neglected. Thus in the mathemat-
ical formulation the grain boundary diffusion in
equation (4) can be dealt with as a quasi-stationary
form by setting $B\partial C_{gb}/\partial\tau = 0$.

As $\sqrt{Dt}$ is a characteristic lattice diffusion length,
D_{gb}/D should be comparable to $\sqrt{Dt}/\delta$. E represents
a characteristic length of this problem while A repres-
ents the relative importance of grain boundary move-
ment to the lattice diffusion in delivering tracer to the
interior of the polycrystal.

Fisher's, Whipple's, and Suzuoka's models corre-
spond to the limit for v equals zero, i. e., $A=0$ in
equations (3a) and (3b) while Glaeser-Evans' model
is the limit for D approaching zero.

Boundary conditions for these equations (3) and
(4) are:

$$C(0,Y,\tau) = C'(0,Y,\tau) = C_{gb}(Y,\tau) \text{ and}$$

$$C(\infty,Y,\tau) = C''(\infty,Y,\tau) = 0 \qquad (5)$$

for the lattice diffusion and

$$C_{gb}(0,\tau) = 1 \text{ and } C_{gb}(\infty,\tau) = 0 \qquad (6)$$

for the grain boundary diffusion, respectively. Initial
conditions are:

$$C(X,Y,0) = C'(X',Y,0) = 0 \qquad (7)$$

for the lattice diffusion and

$$C_{gb} = (Y,0) = 0 \qquad (8)$$

for the grain boundary diffusion, respectively.

In fact, the boundary conditions in equation (5)
require that separation of the grain boundaries, i. e.,
average grain size d be large compared to both the
characteristic lattice diffusion length, $\sqrt{Dt}$, and the
extent of motion of the grain boundary during an-
nealing process to prevent overlapping the lateral
concentration profiles from adjacent boundaries and
to insure that the tracer penetration through the
boundary does not reach the first parallel boundary
beneath the surface.

3. Approximate Analytical Solution for Stationary Grain Boundary Case

Equation (3) and (4) are to be solved with respect-
ive conditions (5) through (8) to obtain C_{gb} and C
and C'. In this section the development of an ap-
proximate analytical solution only for the stationary
grain boundary case, i. e., $A=0$, and zero tracer stor-
age capacity of grain boundary, i. e., $\partial C_{gb}/\partial\tau = 0$ is
introduced.

Reduced governing equations for this case are as
follows:

$$\frac{\partial C}{\partial\tau} = \frac{\partial^2 C}{\partial X^2} \qquad (9a)$$

$$\frac{\partial C'}{\partial\tau} = \frac{\partial^2 C'}{\partial X'^2} \qquad (9b)$$

With reduced tracer balance equation in the grain
boundary:

$$0 = \frac{\partial^2 C_{gb}}{\partial Y^2} + \frac{1}{2}\left(\frac{\partial C}{\partial X}\right)_{X=0} + \frac{1}{2}\left(\frac{\partial C'}{\partial X'}\right)_{X'=0} \qquad (10)$$

Initial and boundary conditions are identical to the
previous ones.

Now the solution for equation (9a) is considered
first since $X=X'$ and $C=C'$ by symmetry. As the left
boundary condition in equation (5) varies with time,

Duhamel's theorem [10] can be applied to find the analytical solution of C. First, C_{gb} as the left boundary condition for equation (9a) is assumed to be constant and then the equation is solved with the right side boundary and initial conditions. Let the solution be $g(X, \tau)$. Then, the ultimate solution of equation (9a) can be deduced from $g(X, \tau)$ for constant surface condition by the use of Duhamel theorem, leading to:

$$C(X, Y, \tau) = \int_0^\tau C_{gb}(Y, \lambda) \frac{\partial}{\partial \tau} g(X, \tau - \lambda) d\lambda \quad (11)$$

If C_{gb} is constant in equation (5), the solution of equation (9a) for constant surface condition is given by:

$$g(X, \tau) = erfc\left(\frac{X}{2\sqrt{\tau}}\right) \quad (12)$$

Since the behavior of C is to be evaluated only at X approaching zero, i. e., in $(\partial C / \partial X_{X \to 0})$, C_{gb} in the integral of equation (11) can, with acceptable accuracy, be expanded in a two-term Taylor series:

$$C_{gb}\left(Y, \tau - \frac{X^2}{4\xi^2}\right) = C_{gb}(Y, \tau) - \frac{X^2}{4\xi^2} \frac{\partial C_{gb}(Y, \tau)}{\partial \tau} \quad (13)$$

if ξ is defined as $X\sqrt{\tau - \lambda}$.

Substituting equation (13) into equation (11) and then integrating the equation in terms of ξ from $X/2\sqrt{\tau}$ to ∞ instead of λ from 0 to τ yields:

$$C = erfc(\theta)C_{gb} - \left(\sqrt{\frac{\tau}{\pi}} Xe^{-\theta} - \frac{1}{2} X^2 erfc(\theta)\right) \frac{\partial C_{gb}}{\partial \tau} \quad (14a)$$

where $\theta = \dfrac{X}{2\sqrt{\tau}}$.

Similarly the other pair solution of equation (14a) can be easily derived:

$$C' = erfc(\theta')C_{gb} - \left(\sqrt{\frac{\tau}{\pi}} X'e^{-\theta} - \frac{1}{2} X'^2 erfc(\theta')\right) \frac{\partial C_{gb}}{\partial \tau} \quad (14b)$$

where $\theta' = \dfrac{X'}{2\sqrt{\tau}}$.

θ and θ' are the dimensionless diffusional characteristic length in the direction of X and X', respectively. Derivatives of C and C' with respect to X and X' at $X = 0$ and $X' = 0$, respectively, are:

$$\left(\frac{\partial C}{\partial X}\right)_{X=0} = -\frac{C_{gb}}{\sqrt{\pi\tau}} - \sqrt{\frac{\tau}{\pi}} \frac{\partial C_{gb}}{\partial \tau} \quad (15a)$$

and

$$\left(\frac{\partial C'}{\partial X'}\right)_{X'=0} = -\frac{C_{gb}}{\sqrt{\pi\tau}} - \sqrt{\frac{\tau}{\pi}} \frac{\partial C_{gb}}{\partial \tau} \quad (15b)$$

Inserting these two equations into equation (10) produces a partial differential equation for the grain boundary concentration:

$$\frac{\sqrt{\tau}}{\pi} \frac{\partial C_{gb}}{\partial \tau} = \frac{\partial^2 C_{gb}}{\partial Y^2} - \frac{C_{gb}}{\sqrt{\pi\tau}} \quad (16)$$

This equation has to be solved in order to determine the ultimate analytical solutions C and C' in equations (14a) and (14b). Necessary boundary and initial conditions for this equation are the ones in equations (6) and (8) in the previous section. Hitherto derived expressions for C and C_{gb} are basically identical to Olander and El-saied's work. To solve them they resorted to numerical solutions.

Here another dimensionless variable, the diffusional characteristic length in the grain boundary, is introduced to solve the partial differential equation (16) analytically:

$$\phi = \frac{Y}{2(4\pi\tau)^{1/4}} \quad (17)$$

Using the chain rule of differentiation, equation (16) can be rewritten as:

$$\frac{\partial^2 C_{gb}}{\partial \phi^2} + 2\phi \frac{\partial C_{gb}}{\partial \phi} - 8C_{gb} = 0 \quad (18)$$

On applying the boundary and initial conditions in equations (6) and (8) to equation (18), the tracer concentration in the grain boundary is determined [11]:

$$C_{gb} = 2^4 \Gamma\left(\frac{4}{2} + 1\right) i^4 erfc(\phi) \qquad (19)$$

$i^4 erfc(\phi)$ in above equation is when $n = 4$ in the following definition:

$$i^n erfc(\phi) = \frac{2}{\sqrt{\pi}} \int_\phi^\infty \frac{(t-z)^n}{n!} e^{-t^2} dt \qquad (20)$$

Tracer concentration in the matrix can be obtained by substituting equation (19) into equation (14a) and (14b).

Then C in equation (14a) representing the tracer concentration in the lattice becomes:

$$C(\theta, \phi) = 2^4 \Gamma(3)\left[erfc(\theta) i^4 erfc(\phi) - \right.$$

$$\left. \frac{1}{4} \phi i^3 erfc(\phi)\left\{\frac{2}{\sqrt{\pi}} \theta e^{-\theta^2} - 2\theta^2 erfc(\theta)\right\}\right] \qquad (21)$$

If equation (21) is rewritten by using consecutive repeated integral forms of the error function and corrected based on the intuition from the many-terms Taylor series manipulation, the equation becomes:

$$C(\theta, \phi) = 2^4 \Gamma\left(\frac{4}{2} + 1\right) erfc(\theta) i^4 erfc(\phi)$$

$$- 2^3 \Gamma\left(\frac{3}{2} + 1\right) \phi i^3 erfc(\phi)\left\{\frac{2}{\sqrt{\pi}} \theta e^{-\theta^2} - 2\theta^2 erfc(\theta)\right\}$$

$$(22a)$$

Since $C = C'$ and $X = X'$ by symmetry, the tracer concentration in the left-side grain lattice of the grain boundary is as follows:

$$C(\theta', \phi) = 2^4 \Gamma\left(\frac{4}{2} + 1\right) erfc(\theta') i^4 erfc(\phi)$$

$$- 2^3 \Gamma\left(\frac{3}{2} + 1\right) \phi i^3 erfc(\phi)\left\{\frac{2}{\sqrt{\pi}} \theta' e^{-\theta'^2} - 2\theta'^2 erfc(\theta')\right\}$$

$$(22b)$$

4. Results and Discussion : Comparisons with Earlier Works

The approximate analytical solution in equation (22) was checked against the earlier works, Fisher's, Whipple's, and Olander-Elsaied's solutions. As mentioned, Fisher derived the following solution by assuming that the tracer concentration in the grain boundary is constant which corresponds to the left boundary condition for the tracer balance equation in the lattice:

$$C_F = C_{gb} erfc(\theta) \qquad (23a)$$

$$C_{gb,F} = e^{-2 \times 2\theta} \qquad (23b)$$

where $\theta = \frac{X}{2\sqrt{\tau}}$ and $\phi = \frac{Y}{2(4\pi\tau)^{1/4}}$ whereas Whipple used Fourier-Laplace transformation technique for the same problem. His technique is explained in detail in reference [2]. His exact solution is as follows:

$$C_W = erfc\left\{\left(\frac{4\pi}{\tau}\right)^{1/4} \phi\right\} + \left(\frac{4}{\pi\tau}\right)^{1/4} \phi \int_1^\Delta \frac{d\sigma}{\sigma^{3/2}}$$

$$exp\left\{-\left(\frac{4\pi}{\tau}\right)^{1/2} \frac{\phi^2}{\sigma}\right\} erfc\left\{\frac{(\sigma - 1)\sqrt{\tau}}{4} + \theta\right\}$$

$$(24a)$$

$$C_{gb,W} = erfc\left\{\left(\frac{4\pi}{\tau}\right)^{1/4} \phi\right\} + \left(\frac{4}{\pi\tau}\right)^{1/4} \phi \int_1^\Delta \frac{d\sigma}{\sigma^{3/2}}$$

$$exp\left\{-\left(\frac{4\pi}{\tau}\right)^{1/2} \frac{\phi^2}{\sigma}\right\} erfc\left\{\frac{(\sigma - 1)\sqrt{\tau}}{4}\right\} \qquad (24b)$$

where $\theta = \frac{X}{2\sqrt{\tau}}$, $\phi = \frac{Y}{2(4\pi\tau)^{1/4}}$ and $\Delta = \frac{D_{gb}}{D}$

Olander and El-saied's mathematical treatment was described in the previous section.

Those four solutions were evaluated numerically by a mainframe IBM computer and the appropriate subroutines in NAG13 mathematical library [12]. In
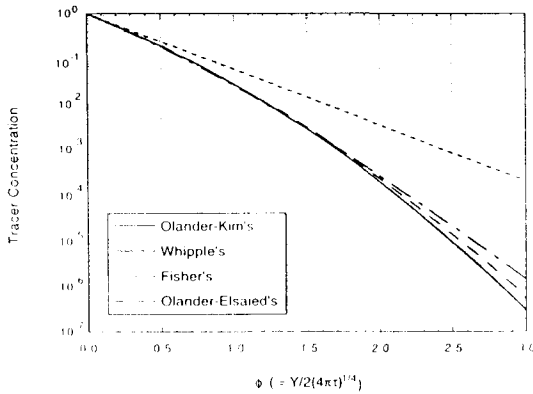
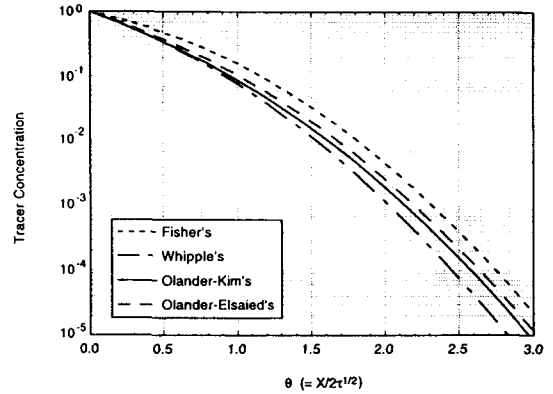Fig. 2. Tracer Concentration in the Grain Boundary

Fig. 3. Tracer Concentration in the Grain Lattice at ϕ =0.57

the computation the built-in error function in the library was used for the error function evaluations while the numerical values for the repeated integrals of the error function were taken from reference [11].

Figure 2 shows the tracer concentration in the grain boundary as a function of the dimensionless variable, ϕ =Y/2$(4\pi\tau)^{1/4}$. The figure demonstrates that all solutions except Fisher's agree well until ϕ is less than 2.0. As ϕ approaches 3.0, the concentration goes down below 10^{-6} and Olander-Kim's solution is about the factor of two or three smaller than Whipple's exact solution. Practically in most of tracer diffusion experiments the detection sensitivity is not higher than 10 ^{6}. Thus the applicable range of ϕ of the current solution lies in the practically important range encountered in the analysis of experimental results. On the other hand, Fisher' solution begins to deviate early even at around ϕ =0.5. In actual, when ϕ is small (less than 0.2) the effect of direct volume diffusion from the surface source remains significant. Thus Fisher's solution is very limited for the practical use though his is simple and easy to use.

In Figure 3 the tracer concentration profiles in the grain lattice are plotted for the four solutions in terms of $\theta = \dfrac{X}{2\sqrt{\tau}}$ at θ =0.57. The figure shows that Olander-Kim's solution is the closest to Whipple's exact solution though the profiles are all similar. It is easily seen that the tracer concentrations in the fig-

ure are actually falling very rapidly (below 0.01 after θ =1.5) like the error function if y ordinate is converted to linear scale. Though in the experimental analysis average tracer concentration is of much more practical use than the profile itself, this comparison is made in the figure since averaging a profile with the known profile is straightforward and, moreover, elucidation of the concentration profile in the grain lattice is important for the future moving grain boundary diffusion study.

Motivation for developing this model is to permit determination of grain boundary diffusivity easily and accurately through the analysis of grain boundary diffusion measurement. Grain boundary diffusion (self or foreign atom) in metals have been studied theoretically and/or experimentally by many authors [1,3, 5,9,13]. Olander interpreted the tracer surface diffusion experiments on UO_2 in terms of gas transport processes [14]. In order to enhance the accuracy of fission gas release rate estimation concerns of developing coupled lattice and grain boundary diffusion analysis technique have arisen instead of 'an equivalent grain diameter' or empirical approaches because they suffer many drawbacks. In that case a good knowledge of grain boundary diffusivity is essential. This Olander-Kim's model, the analytical model of Olander-Elsaied model, was indirectly compared with grain boundary diffusivity measurement data [9],

however, thorough review and estimation of this model with experimental data will be discussed in the following paper in which, on the basis of this solution, general solution for moving grain boundary diffusion and experimental data analysis will be dealt with.

5. Conclusions

An approximate analytical solution for the coupled lattice and stationary grain boundary diffusion case in a solid was developed. It turned out that the accuracy of the solution is comparable to Whipple's exact solution and easy to use since it has a closed-form and every term in the solution is analytically differentiable and integrable.

This work provides a platform for future moving grain boundary diffusion study which is practically important in the fission gas release analysis in reactor operating conditions.

Acknowledgments

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