Proceedings of the Korean Nuclear Society Autumn Meeting
Seoul, Korea, October, 1995

CONCENTRATION CONTOURS IN LATTICE AND GRAIN BOUNDARY DIFFUSION IN A POLYCRYSTALLINE SOLID

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Abstract

Grain boundary diffusion plays significant role in the fission gas release, which is one of the crucial processes dominating nuclear fuel performance. Gaseous fission products such as Xe and Kr generated inside fuel pellet have to diffuse in the lattice and in the grain boundary before they reach open space in the fuel rod. In the mean time, the grains in the 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 diffusion 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.

Lattice and grain boundary diffusion processes in the fission gas release can be studied by 'tracer diffusion' technique, by which grain boundary diffusivity can be estimated and used directly for low burn-up fission gas release analysis. However, even for tracer diffusion analysis, taking both the intragranular grain growth and the diffusion processes simultaneously into consideration is not easy. Only a few models accounting for the both processes are available and mostly handle them numerically. Numerical solutions are limited in the practical use.

Here in this paper, an approximate analytical solution of the lattice and stationary grain boundary diffusion in a polycrystalline solid is developed for the tracer diffusion techniques. This short closed-form solution is compared to available exact and numerical solutions and turns out to be acceptably accurate. It can be applied to the theoretical modeling and the experimental anaysis, especially PIE (post irradiation examination), of low burn-up fission gas release.

1. Introduction

Understanding the release mechanism of fission gases in the UO_2 fuel during reactor operation is of great technical importance for the achievement of reliable high fuel performance.

The build-up of the fission gases in the gap between fuel and cladding not only impairs the heat transfer from the fuel to the coolant but also increases the internal pressure of fuel rods.

Despite the great importance, in actual, the complex and uncertain mechanism of the fission gas release imposes undesirably higher marginal operation than needed, which has to be eliminated for higher performance operation.

The quantitative analysis for the fission gas release involves lattice diffusion in the grains and grain boundary diffusion through the grain boundaries. Thus the combined effect of the two diffusivities is an important factor in the analysis of the fission gas release.

Diffusion processes in the fission gas release can be studied by 'tracer diffusion' techniques. The method follows just reverse way of the diffusion processes.

2. Mathematical Formulation

Taking the grain boundary as the origin of the coordinate x and the surface as the origin of y the tracer conservation equations are[1-3]:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x}$$

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

with initial condition, $C(x,y,0) = C'(x',y,0) = 0$, and boundary conditions,
$C(0,y,t) = C'(0,y,t) = C_{gb}(y,t)$ and $C(\infty,y,t) = C'(\infty,y,t) = 0$.
D is the lattice diffusion coefficient and v is the grain boundary velocity.

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

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

with boundary conditions, $C_{gb}(0,t) = 1$ and $C_{gb}(\infty,t) = 0$, and initial condition, $C_{gb}(y,0) = 0$.
D_{gb} is the grain boundary diffusion coefficient.

By using following dimensionless parameters:

$$X = \frac{x}{E}, \quad Y = \frac{y}{E}, \quad \tau = \frac{Dt}{E^2}, \text{ and } E = \frac{\delta D_{gb}}{2D}$$

new governing equations can be rewritten for the diffusion in the lattice

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

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

with initial condition, $C(X,Y,0) = C'(X',Y,0) = 0$, and boundary conditions,
$C(0,Y,\tau) = C'(0,Y,\tau) = C_{gb}(Y,\tau)$ and $C(\infty,Y,\tau) = C'(\infty,Y,\tau) = 0$.
and for the diffusion in the grain boundary

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

with boundary conditions, $C_{gb}(0,\tau)=1$ and $C_{gb}(\infty,\tau)=0$, and initial condition, $C_{gb}(Y,0)=0$.

A is a dimensionless grain boundary velocity and B is the ratio of lattice and grain boundary diffusivities:

$$A=\frac{vE}{D} \quad \text{and} \quad B=\frac{D}{D_{gb}}.$$

3. Approximate Analytical Solution For Stationary Grain Boundary Case

Reduced governing equations for the case are as follows:

$$\frac{\partial C}{\partial \tau}=\frac{\partial^2 C}{\partial X^2} \tag{1a}$$

$$\frac{\partial C'}{\partial \tau}=\frac{\partial^2 C'}{\partial X'^2} \tag{1b}$$

with reduced tracer balance equation in the grain boundary:

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

Initial and boundary conditions are identical to the previous ones.

Since $X = X'$ and $C = C'$ by symmetry $C(X,Y,\tau)=0$ is considered first.

As the left boundary condition, $C(0,Y,\tau)$, varies with time, Duhamel's theorem[4] is applied to find the analytical solution of $C(X,Y,\tau)$.

That is,

$$C(X,Y,\tau)=\int_0^{\tau} C_{gb}(Y,\lambda)\frac{\partial}{\partial \tau}g(X,\tau-\lambda)d\lambda \tag{3}$$

where $g(X,\tau)$ is the solution of $C(X,Y,\tau)$ for constant surface condition.

If C_{gb} is constant, $g(X,\tau)=erfc\left(\frac{X}{2\sqrt{\tau}}\right)$.

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

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

if ξ is defined as $X/2\sqrt{\tau-\lambda}$.

Then the Duhamel's integration of equation (3) in terms of ξ from $X/2\sqrt{\tau}$ to ∞ instead of λ from 0 to τ yields:

$$C=erfc(\theta)C_{gb}-\left(\sqrt{\frac{\tau}{\pi}}Xe^{-\theta}-\frac{1}{2}X^2 erfc(\theta)\right)\frac{\partial C_{gb}}{\partial \tau} \tag{5a}$$

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

Similarly the other of the pair solution is

$$C'=erfc(\theta')C_{gb}-\left(\sqrt{\frac{\tau}{\pi}}X'e^{-\theta'}-\frac{1}{2}X'^2 erfc(\theta')\right)\frac{\partial C_{gb}}{\partial \tau} \tag{5b}$$

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

Insertion of the derivatives of C and C' in equations (5a-b) with respect to X and X' at X = 0 and X' = 0 produces a partial differential equation for the grain boundary concentration:

$$\frac{\sqrt{\tau}}{\pi}\frac{\partial C_{gb}}{\partial \tau} = \frac{\partial^2 C_{gb}}{\partial Y^2} - \frac{C_{gb}}{\sqrt{\pi\tau}}. \qquad (6)$$

This equation has to be solved in order to determine the ultimate analytical solutions C and C'. Necessary boundary and initial conditions for this equation are the ones in the previous section.

To solve the partial differential equation, another dimensionless variable is introduced:

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

Using the chain rule of differentiation, the partial differential equation (6) for the grain boundary concentration can be rewritten as:

$$\frac{\partial^2 C_{gb}}{\partial \phi^2} + 2\phi\frac{\partial C_{gb}}{\partial \phi} - 8C_{gb} = 0 \qquad (7)$$

On applying the boundary and initial conditions the tracer concentration in the grain boundary is determined[5]:

$$C_{gb} = 2^4\Gamma\left(\frac{4}{2}+1\right)i^4 erfc(\phi) \qquad (8)$$

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

$$i^n erfc(\phi) = \frac{2}{\sqrt{\pi}}\int_\phi^\infty \frac{(t-z)^n}{n!}e^{-t^2}dt \qquad (9)$$

Tracer concentration in the matrix can be obtained by substituting equation (8) into equations (5a) and (5b).

Then the tracer concentration in the lattice becomes:

$$C(\theta,\phi) = 2^4\Gamma(3)\left[erfc(\theta)i^4 erfc(\phi) - \frac{1}{4}\phi i^3 erfc(\phi)\left\{\frac{2}{\sqrt{\pi}}\theta e^{-\theta^2} - 2\theta^2 erfc(\theta)\right\}\right] \qquad (10)$$

If following recurrence relation is applied to above equation:

$$i^n erfc(\phi) = -\frac{\phi}{n}i^{n-1}erfc(\phi) + \frac{1}{2n}i^{n-2}erfc(\phi) \qquad (11)$$

equation (10) becomes:

$$C(\theta,\phi) = 2^4\Gamma(3)\left[i^4 erfc(\phi)\left\{erfc(\theta) + \frac{2}{\sqrt{\pi}}\theta e^{-\theta^2} - 2\theta^2 erfc(\theta)\right\} - \frac{1}{8}i^2 erfc(\phi)\left\{\frac{2}{\sqrt{\pi}}\theta e^{-\theta^2} - 2\theta^2 erfc(\theta)\right\}\right] \qquad (12)$$

As the second term in the right hand side is very small compared to the first term (≤1%), the lattice tracer concentration is reduced to:

$$C(\theta,\phi) = 2^4\Gamma(3)\left[i^4 erfc(\phi)\left\{erfc(\theta) + \frac{2}{\sqrt{\pi}}\theta e^{-\theta^2} - 2\theta^2 erfc(\theta)\right\}\right] \qquad (13)$$

Since the summation of the three terms in { } in above equation is close to $e^{-\theta^2}$, the tracer concentration in the lattice in equation (10) can simplify to the following form:

$$C(\theta,\phi) = 2^4\Gamma(3) i^4 erfc(\phi) e^{-\theta^2} \tag{14a}$$

Since $C = C'$ and $X = X'$ by symmetry,

$$C'(\theta',\phi) = 2^4\Gamma(3) i^4 erfc(\phi) e^{-\theta'^2} \tag{14b}$$

4. Results And Discussion: Comparisons With Earlier Works

Fisher's solution[1]:

$$C_{gb,F} = e^{-2\sqrt{2}\phi}, \qquad C_F = C_{gb} erfc(\theta)$$

$$\text{where } \theta = \frac{X}{2\sqrt{\tau}} \text{ and } \phi = \frac{Y}{2(4\pi\tau)^{1/4}}.$$

Whipple's solution[2]:

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

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

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

El-Saied and Olander's solution[6]:

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

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

$$\text{where } X = \frac{x}{E},\ \tau = \frac{Dt}{E^2},\ \text{and } \theta = \frac{X}{2\sqrt{\tau}}.$$

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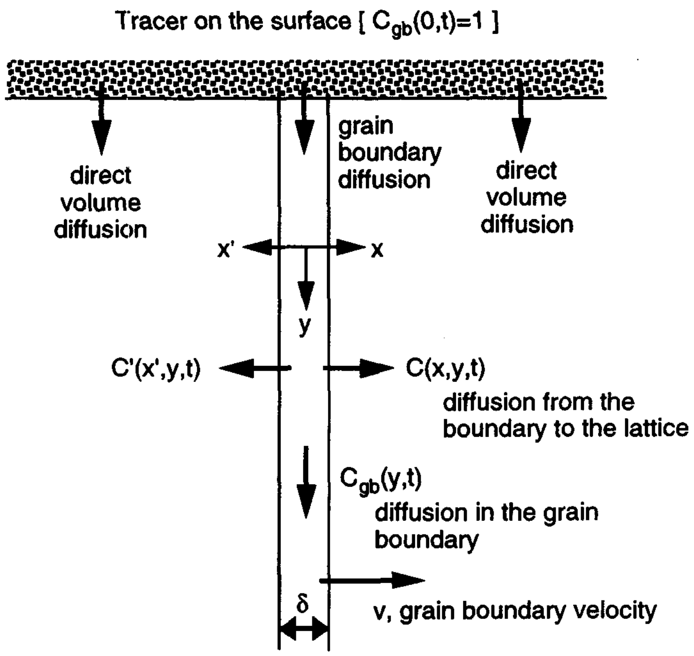

Figure 1 Geometry and solute diffusion processes involved in tracer deposition (slab model)

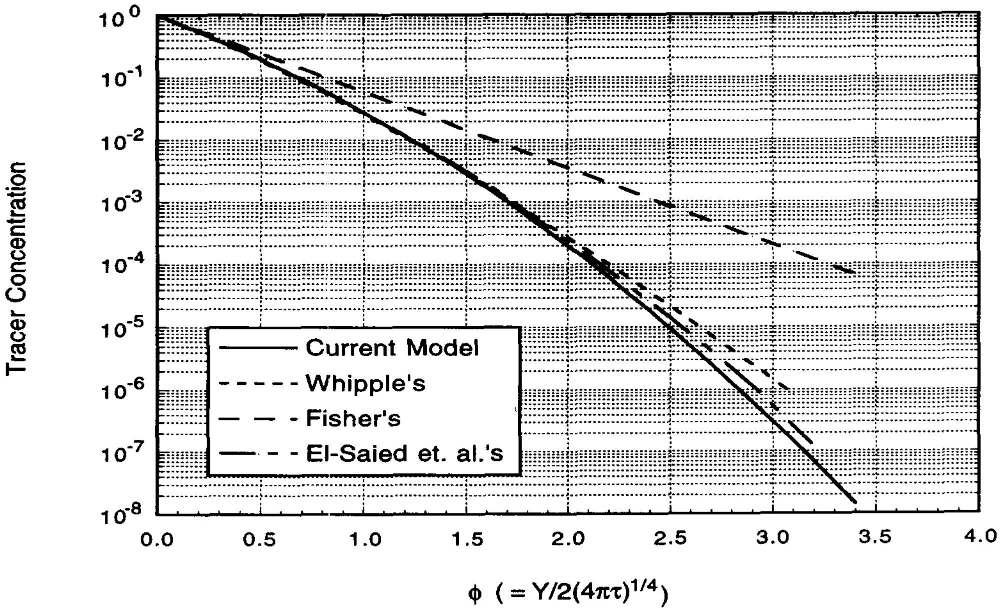

Figure 2 Tracer Concentration in the Grain Boundary