Modelling of Intra-Granular Diffusion, Production and Removal in SOURCE 2.0

by:

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In this paper, the Release by Diffusion module of SOURCE IST 2.0 is discussed. The paper presents the physical processes modelled in this code module and discusses some of the numerical methods applied to the solution of the resulting equations. Application of Fick's Law requires solution of the concentration profile within the grain for isotopes of interest, which in turn requires modelling of isotope production through fission, decay of parent isotopes and neutron capture processes, isotope removal through radioactive decay and neutron capture, and transport via diffusion. The central calculational driver for the Release by Diffusion module, which evaluates the concentration profile, is the GrainDist *subroutine and its sub-programs: its overall structure and place in the Release by Diffusion module is briefly described. The differential equation for the combined diffusion/depletion/production equation is discretized using a Crank-Nicholson approach (*i.e. *second-order spatial discretization and first order temporal discretization). The central node is approached differently: the boundary condition that the spatial derivative of the concentration profile must be zero at the centre of the grain is used to derive the numerical expression solved for. Some discussion is provided on the handling of precursor isotopes and their effect on the evolution of the concentration profile of isotopes of interest. Some testing of the module has been performed and an example is given in which the calculated release rate for* ^{135}*I is compared to an analytical solution for a range of intra-granular spatial discretizations.*

1. Introduction

SOURCE IST 2.0 is a safety analysis code, currently in beta testing (Reference 1), which will model the mechanisms required to calculate fission product release from the fuel for a variety of accident scenarios, including large break loss of coolant accidents (LOCAs) with or without emergency core cooling and other design basis accidents. The goal of the model development is to generate models that are consistent with each other and phenomenologically based, insofar as that is possible given the state of theoretical understanding. SOURCE IST 2.0 is being developed by the Canadian nuclear industry as an Industry Standard Tool.

One of the main processes which drive the fission product release is diffusional release from the fuel grains. The release rate RR of a fission product from the grain surface is given by Fick's Law:

$$RR(t) = -S(t) \cdot D(t) \cdot \left.\frac{\partial C(r,t)}{\partial r}\right|_{r=R} \tag{1}$$

where:

r is the radial position within the grain (m)

t is time (s)
$RR(t)$ is the release rate (atoms/s)
$S(t)$ is the surface area of the grain (m^2)
$D(t)$ is the diffusion coefficient of the isotope in the fuel grains (m^2/s)
$C(r,t)$ is the concentration profile (atoms/(m^3))
R is the radius of the grain (m)

For a given isotope, the change in concentration with time is affected by the following:

1. The diffusion coefficient of the isotope in question, which is affected by the fuel temperature, temperature ramp rate, stoichiometric deviation and fission rate density
2. The removal rate of the isotope in question, which is affected by radioactive decay of the isotope and neutron capture of the isotope
3. The production rate of the isotope in question, which is affected by the direct fission yield of the isotope and the intra-granular distribution of the isotope's parents (both via radioactive decay and neutron capture)

The governing differential equation for the combined diffusion/depletion/production processes in spherical coordinates is:

$$\begin{aligned}\frac{\partial C(r,t)}{\partial t} &= D(t)\cdot\left(\frac{\partial^2 C(r,t)}{\partial r^2}+\frac{2}{r}\cdot\frac{\partial C(r,t)}{\partial r}\right)-\left[\lambda+\sigma\cdot\phi(t)\right]\cdot C(r,t)\\ &\quad+\sum_{FI} y_{FI}\cdot\dot{F}_{FI}+\sum_{DP} f_{DP}\cdot\lambda_{DP}\cdot C_{DP}(r,t)+\sum_{NP}\sigma_{NP}\cdot\phi(t)\cdot C_{NP}(r,t)\end{aligned} \tag{2}$$

where:

λ is the decay constant of the isotope (1/s)
σ is the neutron capture cross-section of the isotope (m^2)
$\phi(t)$ is the neutron flux (neutrons/($m^2\cdot s$))
y_{FI} is the effective direct yield of the isotope from fissionable isotope FI
$\dot{F}_{FI}$ is the fission rate of fissionable isotope FI
DP is an index for counting "decay parents"
f_{DP} is the branching fraction for decay of isotope DP to the isotope
λ_{DP} is the decay constant of isotope DP (1/s)
$C_{DP}(r,t)$ is the isotopic concentration of isotope DP (atoms/m^3)
NP is an index for counting "neutron capture parents"
σ_{NP} is the neutron capture cross-section isotope NP (m^2)
$C_{NP}(r,t)$ is the isotopic concentration of isotope NP (atoms/m^3)

The solution of the above equation for each isotope of interest along each chain of related isotopes is the main task of the Release by Diffusion module and the focus of this paper.

2. Overall Structure of the Release by Diffusion Module

The overall structure (*i.e.* calling hierarchy) of the Release by Diffusion module is shown in Figure 1. SUBROUTINE *Diffusion* is the overall controlling routine through which the Release by Diffusion module is accessed by the main program. Its primary function is to return to the calling routine the concentration profile of all of the isotopes in a series of decay chains at the end of a given interval, along with the diffusional release from the grain of all of these isotopes during this interval. It does this by calling *Redist*, *GrainDist* and then *Integrate* for each decay chain. If necessary, it may call *GrainDist* more than once in order to progress the solution over the entire time interval within the memory constraints of the array declarations.

One of the major overall design considerations for the *Diffusion* program unit is the large memory requirements of *GrainDist* subroutine and its sub-modules. In order to help to contain these requirements, the *GrainDist* routine is designed to allow for a maximum number of time steps. In the event that this maximum number of steps is insufficient to progress the solution across the entire interval, *GrainDist* changes the value of the time interval which was passed to it to the value which it was possible to progress the solution through. If this occurs, the *Diffusion* subroutine will subdivide the overall time interval as needed and call the *GrainDist* routine multiple times. Another, more significant contributor to memory efficiency is the use of sparse matrix techniques in handling the isotopic data for the isotopes in the chains.

3. Numerical Method Applied to *GrainDist* Subroutine

GrainDist and its subsidiary program modules (SUBROUTINEs *ABDBar*, *FindABCR*, *tridiag* and *StoreParent*) are intended to solve the combined diffusion/depletion/production equation using a Crank-Nicholson type discretization. This is a widely-used semi-implicit approach for solving the diffusion equation and so is a natural candidate for application to this more general problem.

For the balance of the discussion of the combined diffusion/depletion/production equation, we will define $A(t) = (\lambda + \sigma \cdot \phi(t))$ and $B(r,t) = \sum_{FI} y_{FI} \cdot \dot{F}_{FI} + \sum_{DP} f_{DP} \cdot \lambda_{DP} \cdot C_{DP}(r,t) + \sum_{NP} \sigma_{NP} \cdot \phi(t) \cdot C_{NP}(r,t)$.

In discretization of the Equation (2), $A(t)$, $B(r,t)$ and $D(t)$ must be handled carefully. Spatial discretization is handled in a simple manner, assuming a uniform spacing of nodes across the grain radius. This simplification was a design decision intended to reduce the complexity of the equations applied to the problem, but it is not the most computationally efficient way to provide sufficient detail near the grain surface. $B(r,t)$ is a function of location and *GrainDist* is structured such that the parents' concentration profiles are always defined before their children's. The nodal mesh is common to all of the calculations for the chain (indeed, the nodal mesh is a constant across all chains for each time interval).

Temporal discretization is more difficult. In principle, the discrete times for which data defining points for $A(t)$, $B(r,t)$ and $D(t)$ are available could be widely varying among themselves and from the time discretization which yields a converged solution for the isotope being considered. Therefore, for any given radial node r_j and time point t_n, we define $\overline{D}^n$, $\overline{B}_j^n$ and $\overline{A}^n$. These represent the average value of $D(t_n)$, $B(r_j,t_n)$ and $A(t_n)$ between t_{n-1} and t_n. For calculation of $\overline{D}^n$, this is assumed to be equivalent to the value of $D(t)$ for the average values of the underlying variables which drive the diffusion coefficient (such as temperature, temperature ramp rate, stoichiometric deviation, etc.).

The Crank-Nicholson approach is to use a second-order discretization in space and a first-order discretization in time. If such an approach is combined with the use of the variables $\overline{D}^n$, $\overline{B}_j^n$ and $\overline{A}^n$ using a nodal mesh of jMax elements, for all radial nodes except r_{jMax} (the grain surface) and r_1 (the grain centre), the resulting equation is as follows:

$$\begin{aligned}&-\frac{\overline{D}^n}{2\cdot\Delta r}\cdot\left(\frac{1}{\Delta r}-\frac{1}{r_j}\right)\cdot C_{j-1}^{n+1}+\left(\frac{\overline{A}^n}{2}+\frac{\overline{D}^n}{\Delta r^2}+\frac{1}{\Delta t^n}\right)\cdot C_j^{n+1}-\frac{\overline{D}^n}{2\cdot\Delta r}\cdot\left(\frac{1}{\Delta r}+\frac{1}{r_j}\right)\cdot C_{j+1}^{n+1}\\&=\frac{\overline{D}^n}{2\cdot\Delta r}\cdot\left(\frac{1}{\Delta r}-\frac{1}{r_j}\right)\cdot C_{j-1}^{n}-\left(\frac{\overline{A}^n}{2}+\frac{\overline{D}^n}{\Delta r^2}-\frac{1}{\Delta t^n}\right)\cdot C_j^{n}+\frac{\overline{D}^n}{2\cdot\Delta r}\cdot\left(\frac{1}{\Delta r}+\frac{1}{r_j}\right)\cdot C_{j+1}^{n}+\overline{B}_j^n\end{aligned}\qquad\text{spherical grains (3)}$$

$$\begin{aligned}&-\frac{\overline{D}^n}{2\cdot\Delta r}\cdot\left(\frac{1}{\Delta r}-\frac{1}{2r_j}\right)\cdot C_{j-1}^{n+1}+\left(\frac{\overline{A}^n}{2}+\frac{\overline{D}^n}{\Delta r^2}+\frac{1}{\Delta t^n}\right)\cdot C_j^{n+1}-\frac{\overline{D}^n}{2\cdot\Delta r}\cdot\left(\frac{1}{\Delta r}+\frac{1}{2r_j}\right)\cdot C_{j+1}^{n+1}\\&=\frac{\overline{D}^n}{2\cdot\Delta r}\cdot\left(\frac{1}{\Delta r}-\frac{1}{2r_j}\right)\cdot C_{j-1}^{n}-\left(\frac{\overline{A}^n}{2}+\frac{\overline{D}^n}{\Delta r^2}-\frac{1}{\Delta t^n}\right)\cdot C_j^{n}+\frac{\overline{D}^n}{2\cdot\Delta r}\cdot\left(\frac{1}{\Delta r}+\frac{1}{2r_j}\right)\cdot C_{j+1}^{n}+\overline{B}_j^n\end{aligned}\qquad\begin{array}{r}\text{"infinite"}\\\text{cylindrical grains}\\\text{(4)}\end{array}$$

This can be expressed as a matrix equation **M** · **u** = **r**, where the vector **u** is composed of the nodal concentrations, **r** is the vector of the right hand sides of the equations and **M** is a tri-diagonal matrix containing the equation coefficients. The matrix **M** can be expressed using three vectors **a**, **b** and **c** rather than a full array, where **a** represents the sub-diagonal of **M** (*i.e.*, $\mathbf{a}_j = \mathbf{M}_{j,\ j\text{-}1}$, j = 2…jMax-1), **b** the diagonal of **M** (*i.e.*, $\mathbf{b}_j = \mathbf{M}_{j,\ j}$, j = 2…jMax-1) and **c** the super-diagonal of **M** (*i.e.*, $\mathbf{c}_j = \mathbf{M}_{j,\ j\text{-}1}$, j = 1…jMax-2). As can be seen from the equations above, **M** is diagonally dominant, which means that a simple solution technique can be applied, which requires only matrix decomposition, forward substitution and back-substitution. No pivoting is required (see Reference 2).

In order to complete the matrix equation, expressions for j=jMax and j=1 need to be determined from the boundary conditions. The boundary condition which applies at the surface of the grain is simple. For j=jMax:

$$C_{jMax}^n = 0 \tag{5}$$

The boundary condition at the centre of the grain is that $\left.\frac{\partial C(r,t)}{\partial r}\right|_{r=0} = 0$. The partial derivative can be approximated with a second order discretization. If the boundary condition is applied, we are left with the following expression for j=1:

$$-3\cdot C_1^{n+1} + 4\cdot C_2^{n+1} - C_3^{n+1} = 0 \tag{6}$$

If this expression is used, then the resulting matrix equation for the entire nodal mesh is no longer tri-diagonal, which means that the simple tri-diagonal solver could no longer be applied. However, recall that, if Equations (3) and (4) are re-written using **a**, **b**, **c** and **r** for j=2, we obtain:

$$\mathbf{a}_2\cdot C_1^{n+1} + \mathbf{b}_2\cdot C_2^{n+1} + \mathbf{c}_2\cdot C_3^{n+1} = \mathbf{r}_2 \tag{7}$$

If we multiply Equation (6) by $\mathbf{c}_2$ and then add Equation (7) to it, we get the expression for j=1 which allows us to preserve a tri-diagonal matrix solution technique.

For j=1:

$$(\mathbf{a}_2 - 3\cdot\mathbf{c}_2)\cdot C_1^{n+1} + (\mathbf{b}_2 + 4\cdot\mathbf{c}_2)\cdot C_2^{n+1} = \mathbf{r}_2 \tag{8}$$

In order to solve the tri-diagonal matrix equation above, a value of Δt^n must be assumed. If the value of Δt^n is too high, a converged solution will not be achieved and calculational results will be meaningless. If Δt^n is too small, needless computational effort will be wasted without any significant improvement in the accuracy of the solution. Therefore, a convergence loop must be defined to set Δt^n as large as possible consistent with an accurate solution. The convergence loop needs to have a minimum concentration below which convergence is assumed. This is set in *GrainDist* to be that concentration at which there is 1 atom of the isotope in question within the nodal volume. For all nodes whose concentrations are above this minimum, the convergence criterion is that the nodal concentrations at $t+\Delta t^n$ using one step of size Δt^n and two steps of size $\Delta t^n/2$ all agree within the tolerance set by the variable Conv (currently set to $1 \cdot 10^{-6}$).

When a converged time step is achieved, a mechanism is required which will attempt to increase the time step as possible to increase calculational efficiency. The time step logic for *GrainDist* is shown in Figure 2. The approach is to reduce the time step until convergence is achieved and to attempt to increase it only when two successive trials of the same time step are both successful.

Note that evaluating the value of $\overline{B}_j^n$ for a given isotope requires evolution of the concentration profile of all of the parents of the isotope in question. This implies that, for some isotopes, the entire transient evolution of the concentration profile will need to be stored until it is used to evaluate the evolution of a child.

Associated with the diffusion modelled above will be a release of fission products from the grain. Equation (1) shows that the release rate RR(t) from the surface of the grain is equal to:

$$RR(t) = -4 \cdot \pi \cdot r_{Nr}^2 \cdot D(t) \cdot \left.\frac{\partial C(r,t)}{\partial r}\right|_{r=r_{Nr}} \qquad \text{spherical grains (9)}$$

$$RR(t) = -2 \cdot \pi \cdot r_{Nr} \cdot L \cdot D(t) \cdot \left.\frac{\partial C(r,t)}{\partial r}\right|_{r=r_{Nr}} \qquad \text{"infinite" cylindrical grains (10)}$$

where:

L is the grain length (m)

The value of $\left.\frac{\partial C(r,t)}{\partial r}\right|_{r=r_{Nr}}$ can be evaluated for each time step using the concentration profile which is determined for that time step by solving the tri-diagonal discretization of the combined diffusion/-depletion/production equation. The approach taken is to fit a second-order polynomial through the C_j^{Nr-2}, C_j^{Nr-1} and C_j^{Nr} (recall that $C_j^{Nr} = 0$) and solve for the slope of the polynomial at the grain surface. This approach is equivalent to using a second-order backward-difference approximation to the first derivative.

4. Testing of the Release Rate Calculation

The Release by Diffusion module has been tested by comparing its results against a known analytical solution for a range of different node mesh densities. Consider the steady-state solution for the concentration profile of the first member of a decay chain for the diffusion of a radioactive fission product from a spherical fuel grain (see Reference 3):

$$C(r) = \frac{B}{\lambda}\left(1 - \frac{r \cdot \sinh(\alpha R)}{R \cdot \sinh(\alpha r)}\right) \qquad (11)$$

where:

$C(r)$ is the steady state concentration (atoms/m^3),
r is radial position between 0 and R (m),
B is the production (birth) rate from fission (atoms/($m^3 \cdot s$)),
λ is the decay constant of the nuclide (1/s),
R is the radius of the spherical grain (m),
α is the "lumped" constant $(\lambda/D)^{1/2}$ (1/m) where D is the diffusion coefficient (m^2/s).

The derivative of this expression, when evaluated at r=R, is:

$$\left.\frac{\partial C(r)}{\partial r}\right|_{r=R} = \frac{B \cdot R}{\lambda}\left(\frac{1}{R^2} - \frac{\alpha}{R \cdot \tanh(\alpha R)}\right) \tag{12}$$

and the resulting release rate can be calculated from Equation (1)

Using a fission rate of $1.55 \cdot 10^{19}$ 1/s, a yield of 6.28%, $\lambda = 2.93 \cdot 10^{-5}$ 1/s, R=20 μm and $D = 2.4 \cdot 10^{-18}$ 1/($m^2 \cdot s$), the release rate of I-135 can be calculated to be $1.4 \cdot 10^5$ atoms per second. In Figure 3, the fractional difference between this value and the prediction given by running *TestDiff* as a function of number of nodes in the grain is given. Convergence to within 1% is achieved by 400 nodes.

5. Conclusion

As part of the SOURCE IST 2.0 development effort, the Release by Diffusion module has been developed. This module applies a Crank-Nicholson type discretization to the combined diffusion/-depletion/production equation. Testing of the module indicates that it provides excellent agreement with the analytical solution of the steady-state release rate of the first element in a chain.

The Release by Diffusion module provides SOURCE IST 2.0 with the capability to model one of the main engines of fission product release. No previous CANDU fission product release code has represented the combined processes of isotope generation via fission of multiple actinide isotopes, decay and neutron capture; isotope depletion via decay and neutron capture; and diffusion of the isotope together in as physically representative a manner as this module does. The incorporation of this capability into SOURCE IST 2.0 contributes significantly to the fact that this code, when released, will be capable of far more detailed and representative assessments of fission product release than any code which has been used in the past in the Canadian nuclear industry.

6. References

1 D. H. Barber, F. C. Iglesias, L. W. Dickson, R. S. Dickson, M. J. Richards and R. A. Gibb, "SOURCE IST 2.0: Development and Beta Testing", presented at the 6th International Conference on CANDU Fuel, September 27-29, 1999, Niagara Falls, Ontario.

2 "Numerical Recipes in FORTRAN 77: The Art of Scientific Computing", ISBN 0-521-43064-X, Cambridge University Press, Second Edition, 1996

3 A.H. Booth, "A Suggested Method for Calculating the Diffusion of Radioactive Rare Gas Fission Products from UO2 Fuel Elements and a Discussion of Proposed In-Reactor Experiments That May Be Used to Test its Validity", Atomic Energy of Canada Report, AECL-700, 1957.

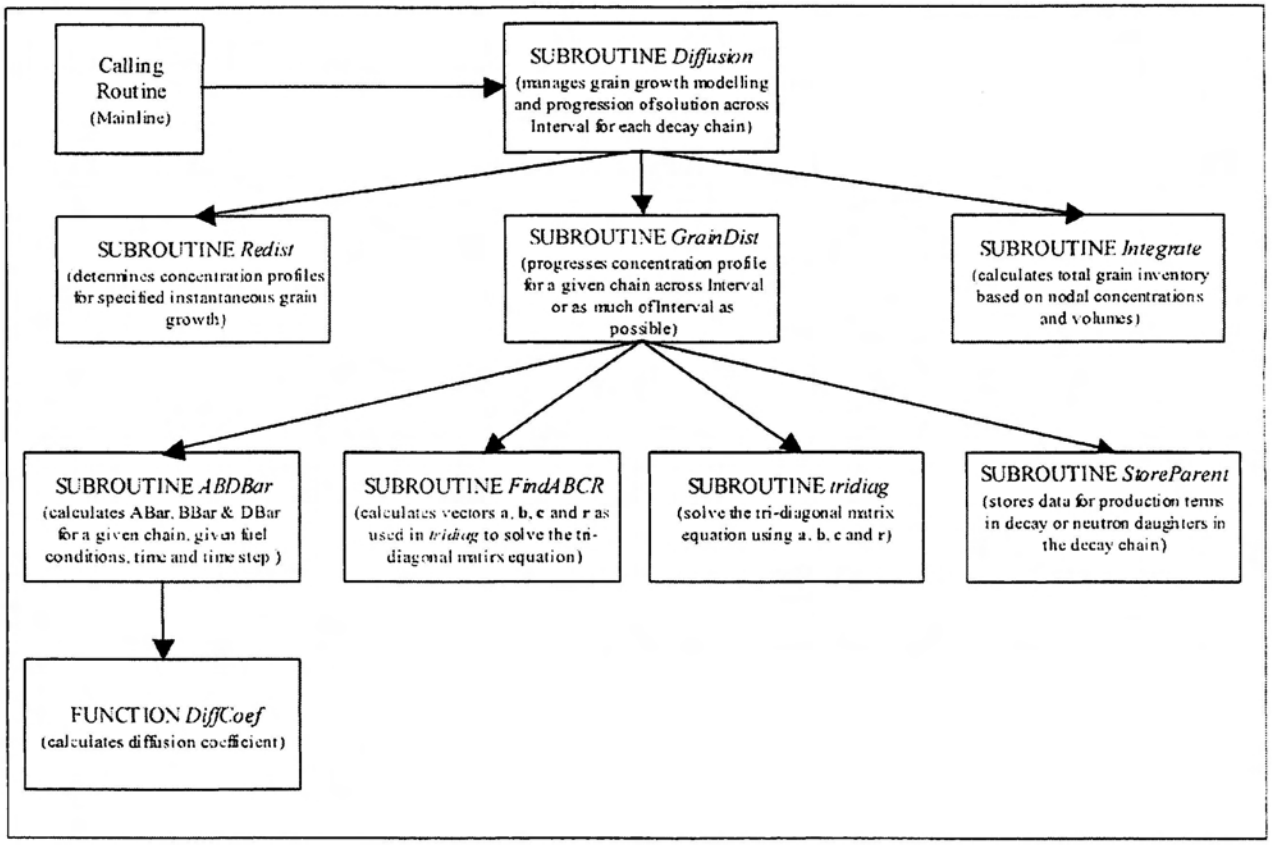

Figure 1: Calling Hierarchy of Program Modules in the Release by Diffusion Module

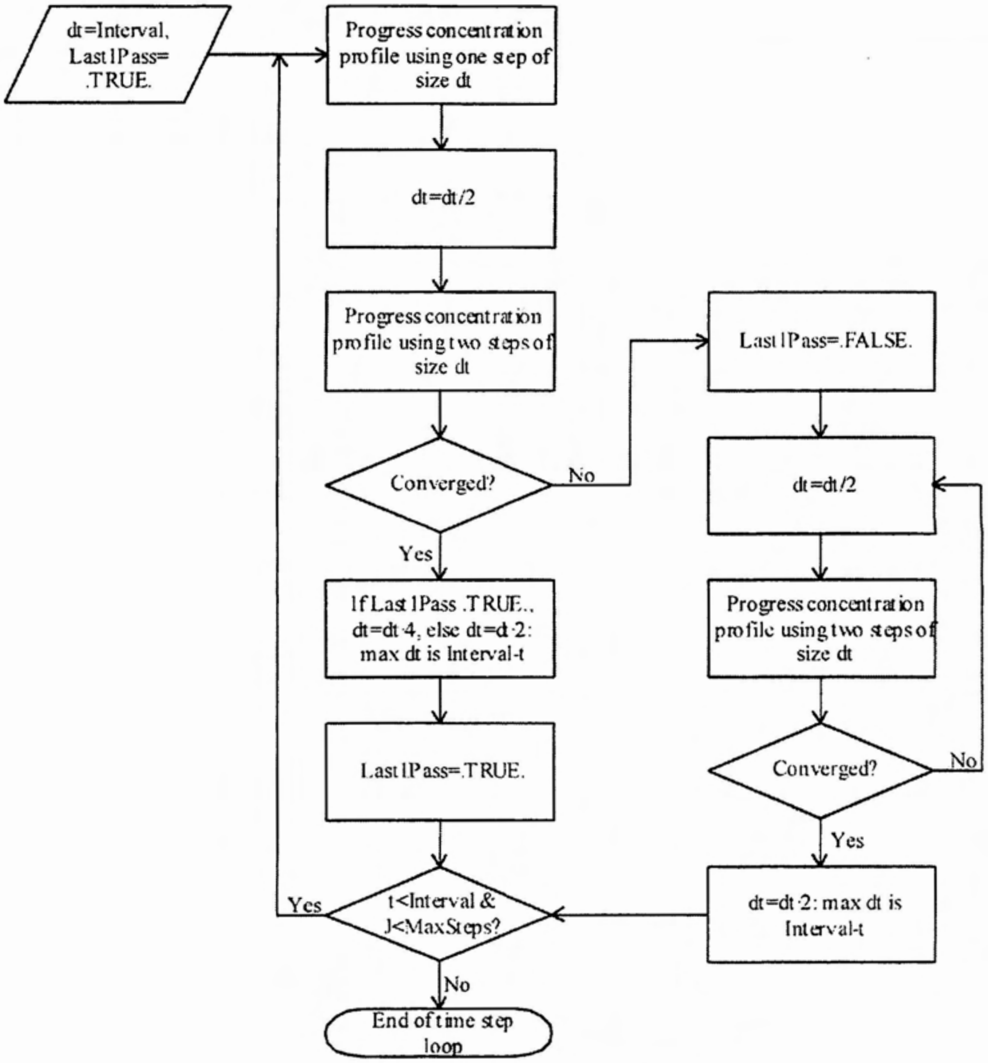

Figure 2: Time Step Control Logic for *GrainDist*

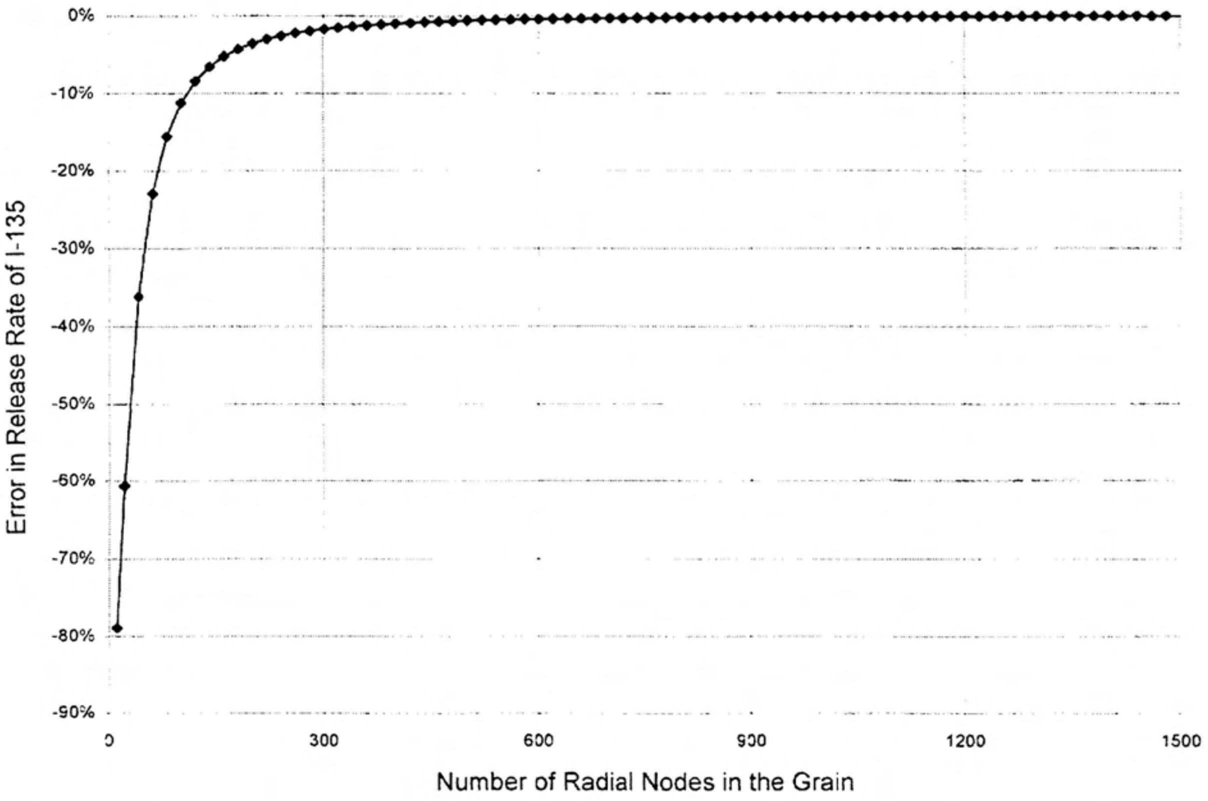

Figure 3: Error in Release Rate of I-135 as a Function of the Number of Radial Nodes for *GrainDist*