TRANSPORT THEORY EVALUATION OF THE RELEASE OF SHORT-LIVED FISSION PRODUCTS FROM FUEL GRAINS

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ABSTRACT

The first barrier to the release of fission products from reactor fuel elements is the UO_2 matrix. An assessment of the fraction of a particular fission product being released from the fuel grains is necessary to assess subsequent release mechanisms and pathways. Although the movement of fission products within the fuel lattice is assumed to be by a concentration gradient driven diffusion process, the modelling of this process by diffusion theory requires some approximations to be made when applied to short-lived fission products. This paper applies the more rigorous conditions of transport theory to evaluate the probability of fission product release from a fuel grain as a function of the fission product mean free path and the size of the fuel grain.

1.0 INTRODUCTION

At low temperatures, the UO_2 pellets in CANDU fuel are an effective primary containment for the fission products produced during irradiation. At elevated temperatures (*i.e.*, >1000°C), various physical processes cause the movement of these fission products out of the ceramic fuel. The more important processes include diffusion of volatile fission products either as single gas atoms or as bubbles, growth of the fuel grains in which the moving grain boundary accumulates fission products from the fuel matrix through which it is growing, and direct release by recoil/knockout of fission products produced close to exposed UO_2 surfaces.

In diffusion and in grain growth, the time required to transport the fission product from the fuel matrix to a fuel grain surface is important as it delays the release of fission products from the fuel pellets. This is particularly significant in assessing the release of radioactive fission products, as a number of them will decay during the transport process and never reach the fuel pellet surface.

The most probable distance a fission product can travel in UO_2 between birth and decay is termed the fission product's diffusion length, L. Numerically L is equal to the square root of the ratio of the fission product's diffusion coefficient, D, to its decay constant, λ:

$$L = (D/\lambda)^{\frac{1}{2}} \quad \ldots (1)$$

The <u>average</u> distance a fission product travels in UO_2 between birth and decay, τ, is equal to twice its diffusion length, *i.e.*, $\tau = 2L$ (see Appendix A).

The average distance a fission product can travel before undergoing an event which can change its direction of travel is termed its transport mean free path (or jump distance), μ. The diffusion coefficient, D, is related to the transport mean free path by the classical Einstein equation,

$$D = (\mu^2/3)k \quad \ldots (2)$$

where k is the jump frequency (Reference 1).

This paper examines the modelling of the release of grain bound fission product inventory by diffusion during steady state operation. For radio-

active fission products, it can be shown that current modelling techniques provide an accurate approximation of fission product release by comparison to the more exact transport theory. However, the comparison also shows that release kinetics are independent of fission product concentration or concentration profile and depend only on the random migration of the fission products through the UO_2 lattice.

Section 2.0 defines the terms, cumulative and instantaneous fractional release. Section 3.0 describes the current diffusional release model proposed by Booth (Reference 1) and generalized by Kidson (Reference 2). Section 4.0 describes the transport theory approximation to the diffusion process. Section 5.0 presents a solution to the transport equation in terms of probabilities. Section 6.0 illustrates how the transport theory approximation can be applied in modelling grain bound release by diffusion.

2.0 RELEASE FRACTION

2.1 Volatile, Stable Fission Products

Stable fission products do not decay during their migration from the fission site where they are born to their escape at a fuel grain surface. In steady state, the number of stable fission products reaching the grain surface per unit time is exactly equal to the number of stable fission products born within the grain volume during the same time period. The probability that a volatile, stable fission product will be released from the fuel grain in which it was born is unity.

The instantaneous rate of release of a volatile, stable fission product is governed by the random migration of fission products within the fuel grain. In steady state, the release rate, R, exactly equals the birth rate, B, of the fission product in question (*i.e.*, the ratio, R/B, is unity).

If the fission rate is changing with time, the birth rate and the release rate will both change. Once another steady state is reached, R and B again become equal.

For stable fission products, it is customary to define the release fraction, f_R, as:

$$f_R = \int R\,dt \;/\; \int B\,dt \quad \ldots (3)$$

where the integration is from time zero. Note, f_R defined this way is the cumulative release fraction.

2.2 Volatile, Radioactive Fission Products

Unlike a mobile, stable fission product which must eventually leave the fuel grain in which it is produced, a short-lived fission product can decay before it escapes. The shorter the fission product half-life, the more likely it is to decay before escape.

The average concentration, N, of a radioactive fission product within a fuel grain can be described by the simple differential equation:

$$dN/dt = B - \lambda N - R \quad \ldots (4)$$

At steady state, $dN/dt = 0$, the equilibrium average concen-tration is given by:

$$N_{eq} = (B - R)/\lambda \quad \ldots (5)$$

Due to radioactive decay, the ratio, R/B, must be less than unity under steady state conditions. The R/B ratio is the probability that a radioactive fission product, born within a fuel grain, will be released from the fuel grain before the fission product decays. The ratio, R/B, is termed the instantaneous release fraction.

Following reactor startup to steady power, the average fission product concentration in the fuel grain can be found by integrating equation (4) to yield:

$$N = N_{eq} (1 - e^{-\lambda t}) \ldots (6)$$
or
$$N = N_{eq} (1 - e^{-.693\ t/T½})$$

where T½ is the half life of the fission product in question. Steady operation for 3 to 4 half lives is sufficient to produce near equilibrium values of both N_{eq} and R/B.

3.0 DIFFUSION APPROXIMATION

In 1957, A.H. Booth proposed a model for the release of volatile fission products from fuel grains (Reference 2). Booth made three important assumptions:

1) fuel grains can be treated as spheres,
2) the fission product concentration at the surface of the fuel grain and the concentration gradient at the grain center are zero, and
3) the differential equation for radioactive fission product concentration, C,

$$D \nabla^2 C + B - \lambda C = dC/dt \ldots (7)$$

can be applied throughout the fuel grain volume.

The steady state solution of equation (7), for operation at constant birth rate, can be found analytically or evaluated using standard finite difference modelling approaches (see Appendix B). Algebraic solution of equation (7) yields the following expression for the instantaneous release fraction during steady operation:

$$R/B = 3(L/a)\{\coth(a/L) - (L/a)\} \ldots (8)$$

in terms of the dimensionless quantity, diffusion length to grain radius, (L/a).

4.0 TRANSPORT APPROXIMATION

It is common practice in neutronics calculations to augment physical dimensions by an extrapolation distance derived from transport theory analysis (Reference 4). This dimen-sion adjustment is required as the diffusion theory approx-imation (*i.e.*, equation (7)) is not accurate within three mean free paths of a physical boundary. The extrapolation distance is defined as $0.71\lambda_{tr}$, where λ_{tr} is the transport mean free path for neutrons.

Although neutron flux and fission product concentration are expressed in different units, and thus, so are the cor-responding diffusion coefficients, the transport mean free paths for both have the same significance. The transport mean free path for neutrons, λ_{tr}, and the transport mean free path for fission products, μ, both represent the average distance travelled before the neutron or fission product undergoes an event which could change their direction of travel.

By replacing the grain radius, a, in equation (8) with the augmented radius, $a' = a + 0.71\mu$, the diffusion approx-imation in Section 3.0 can be applied to a fuel grain greater than six mean free paths in diameter.

If the transport mean free path for fission products is small compared to typical fuel grain diameters, then the diffusion approximation and the transport approximation would yield essentially identical results for R/B.

5.0 MODELLING

5.1 Transport Approximation

The transport approximation is easily demonstrated using the finite difference solution shown in Appendix B. In the diffusion approximation the concentration is assumed zero at the physical grain surface. In the transport approximation, the fission product concentration is extrapolated to zero at a distance 0.71μ outside the physical boundary.

Figure 1 compares the diffusion approximation and the transport approximation for the instantaneous release fraction, R/B, as a function of the ratio of diffusion length to grain radius, L/a, for short-lived fission products. The analysis assumes a typical fuel grain radius of 5 microns and a transport mean free path of 1 micron. As can be seen, the diffusion approximation over-predicts the release fraction by an insignificant amount even for a transport mean free path of 1 micron.

5.2 Probability Solution to the Transport Model

An analytical expression for the instantaneous release fraction, R/B, is derived in Appendix C based on proba-bilities. The resulting expression is:

$$R/B = 3\Theta(1+e^{-1/\Theta}) \ldots (9)$$
$$+ 12\Theta^3(e^{-1/\Theta}(1 + 1/\Theta) - 1)$$

where Θ is the ratio of the diffusion length to the fuel grain radius, L/a.

As shown in Figure 2, the results of equation (9) are indistinguishable from the diffusion approximation given by equation (8). This agreement indicates that:

1)the transport mean free path for fission products is very small (*i.e.*, negligible extrapolation distance). (The value for μ derived in Reference 1 is less than 6 Å),

2)whereas the diffusion approximation uses concen-tration gradients and diffusion coefficients, the transport solution (equation (9)) uses only the diffusion coefficient. Therefore, the release of fission products is adequately described by a random walk analysis (Appendix C) of a fission product gas, and

3)the release fraction of a fission product (ignoring chemical effects) is not influenced by the presence of other fission product gases in the diffusing media.

At low temperature or for fission products with very short half-lives, the diffusion length is very small. Under such condition where Θ is much less than unity, equation (9) reduces to:

$$R/B = 3\Theta \ldots (10)$$

which is identical to the fractional release by recoil derived by Wise (Reference 5) and by Lewis (Reference 6).

6.0 MODEL APPLICATION

Figure 3 illustrates the steady state release fraction for I-131 (half-life equals 8 days) and Kr-87 (half-life equals 1.27 hours) from a 10 micron diameter spherical fuel grain using a typical diffusion coefficient for thermally activated diffusion given by the expression:

$$D = 7.6 \times 10^{-10}\, e^{-35350/T} \ldots \quad (11)$$

where D is in m^2/s and the temperature, T, is in Kelvin.

Although equations (8) and (9) were derived from steady state considerations, release during transients can be approximated by coupling the results of equation (4) with equation (9). For example, during an initial rise to power, the average fission product concentration is given by equation (6). The instantaneous release fraction, R/B, during the transient can be represented by the expression:

$$R/B = p(\Theta)\ N_{eq}(1 - e^{-\lambda t}) \ / \ N_{eq} \ldots \quad (12)$$
or
$$R/B = p(\Theta)\ (1 - e^{-\lambda t})$$

as $p(\Theta)$ is the release probability under isothermal conditions for the fission product average concentration based only on uniform birth rate throughout the fuel grain volume.

7.0 REFERENCES

(1) B.J. LEWIS and H.E. SILLS, "Fission-product transport and the diffusion approximation", J. Nuc. Mat., 184(1991)107-112.

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(3 G.V. KIDSON, "A Generalized Analysis of the Cumulative Diffusional Release of Fission Product Gases from an "Equivalent Sphere" of UO_2", J. Nuc. Mat., 88(1980)299-308.

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(5) C. WISE, "The Transport of Fission Products Close to the Fuel Surface", J. Nuc. Mat., 152(1988)102-113.

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APPENDIX A

Calculation of τ

Following Reference 1, considering an isotropic point source emitting S short-lived fission products per second in an infinite medium. Except at the origin, the steady-state diffusion equation is

$$\nabla^2 C - C/L^2 = 0 \qquad \ldots A.1$$

where spherical symmetry has been assumed. At any radius, r,

$$4\pi r^2(-D\ dC/dr) = S\ e^{-r/L} \qquad \ldots A.2$$

where the term $e^{-r/L}$ allows for the attenuation of the source fission products by decay. Imposing equation (A.2), with C remaining finite as r becomes infinite, yields the solution

$$C = (S/4\pi D)\ (e^{-r/L})/r \qquad \ldots A.3$$

The number of atoms, dC, which decay per second in a spherical shell of volume $dV = 4\pi r^2\ dr$ located between r and r+dr is therefore

$$dC = \lambda C dV = (S/L^2)\ re^{-r/L}\ dr \qquad \ldots A.4$$

The probability, p(r)dr, that a fission product emitted by the source decays in dr is just dC/S or

$$p(r)dr = (r/L^2)\ e^{-r/L}\ dr \qquad \ldots A.5$$

and the probability distribution function, p(r), is

$$p(r) = (r/L^2)\ e^{-r/L} \qquad \ldots A.6$$

Using equation A.6 as the weighting function and integrating from 0 to infinity, the average value of r is calculated as

$$\tau = \int rp(r)pr\ /\ \int p(r)dr = 2L \qquad \ldots A.7$$

APPENDIX B

A Finite Difference Approximation for Concentration Driven Diffusion

If the spherical fuel grain is represented by a series of concentric, equal thickness shells, then the steady state diffusion equation:

$$D\ \nabla^2 C + B - \lambda C = 0 \qquad \ldots B.1$$

can be written as

$$D(C_- - C)S_-/\Delta r + D(C_+ - C)S_+/\Delta r + (B-\lambda C)V = 0 \qquad \ldots B.2$$

where D is the diffusion coefficient,
C is the fission product concentration on the current shell surface,
C_-, C_+ are the fission product concentrations on the surface of the previous and next shell respectively,
S_-, S_+ are the surface areas of spheres passing midway between C_- and C, and C and C_+ respectively,
Δr is the shell thickness,
B is the fission product birth rate,
λ is the fission product decay constant, and

V is the volume between S_- and S_+.

By rearranging and collecting like terms, equation B.2 can be re-written as:

$$aC_- + bC_+ - eC + f = 0 \quad \ldots \text{B.3}$$

where $a = DS_-/\Delta r$
$b = DS_+/\Delta r$
$e = a + b + \lambda V$, and
$f = BV$

Assuming the recursive relation:

$$C = uC_+ + p \quad \ldots \text{B.4}$$

and substituting it in equation A.4 for C_-, the further recursive relationships:

$$u = b/(e - au_-) \quad \ldots \text{B.5}$$
$$p = (f + p_-)/(e - au_-)$$

for the coupling coefficients can be developed ($u = 0$ and $p = (f/e)$ at the fuel grain center).

For the diffusion approximation, the concentration on the surface of the last shell is set to zero. The concentration on the surface of the other shells is found by applying equation B.4 recursively from the outer to inner shell.

For the transport approximation, the same procedure is used except that an additional shell of thickness 0.71μ, the extrapolation distance, and having zero birth rate, is added to the model of the fuel grain.

APPENDIX C

Escape Probability of Radioactive Fission Products from a Spherical Fuel Grain

A radioactive fission product has a diffusion length given by the expression:

$$L = (D/\lambda)^{\frac{1}{2}} \quad \ldots \text{C.1}$$

where D is the diffusion coefficient of the fission product, and
λ is the fission product's decay constant.

The average distance of travel is designated τ where $\tau = 2L$.

If the fuel grain (sphere A in Figure C.1) has an equivalent sphere radius **R**, then the probability density that it will arrive on the surface of a sphere B with radius **b**, with origin at radius **r**, before it decays, is given by the expressions:

$$P(\mathbf{r},\mathbf{b}) = 0, \text{ sphere B inside sphere A} \quad \ldots \text{C.2}$$
$$= (1+\mathbf{r}/(2\mathbf{b})+\mathbf{b}/(2\mathbf{r})-\mathbf{R}^2/(2\mathbf{br}))/2,$$
$$\text{sphere B intersecting sphere A}$$
$$= 1, \text{ otherwise}$$

The value of **r** is random within the fuel grain volume. Integrating over the grain volume yields the probability function:

$$P(\mathbf{b}) = 3(\mathbf{b}/\mathbf{R}-(\mathbf{b}/\mathbf{R})^3/12)/4 \text{ for } \mathbf{b}<2\mathbf{R} \quad \ldots \text{C.3}$$
$$= 1 \text{ for } \mathbf{b}\geq 2\mathbf{R}$$

The value of **b** obeys the probability density function given by:

$$P(\mathbf{b},\tau) = (\mathbf{b}/\tau^2)\ e^{-b/\tau} \qquad \ldots \text{ C.4}$$

Setting $(\tau/(2\mathbf{R}))$ to Θ and integrating **b** over the range 0 to infinity yields the escape probability as:

$$P = 3\Theta(1 + e^{-1/\Theta}) + 12\Theta^3(e^{-1/\Theta}(1 + 1/\Theta) - 1) \qquad \ldots \text{ B.5}$$

The escape probability is a unique function of the mean free path and the equivalent sphere diameter.

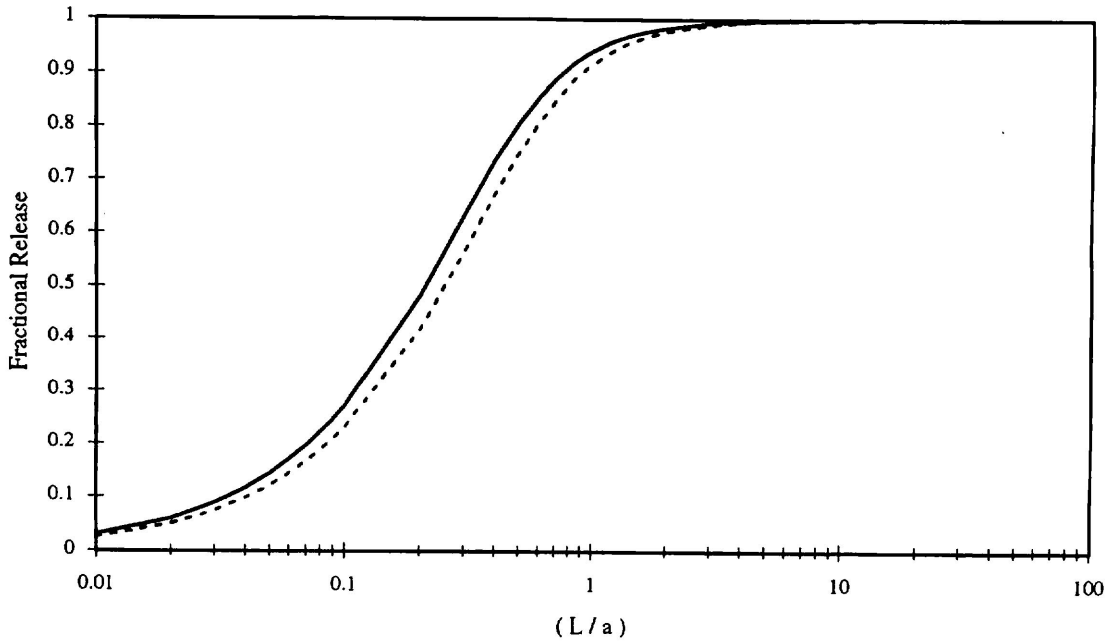

Figure 1
The probability of a short-lived fission product escaping from a spherical fuel grain. The solid line represents the diffusion theory evaluation of the escape probability. The dashed line represents the transport approximation. A grain radius of 5 microns and a transport mean free path of 1 micron are assumed.

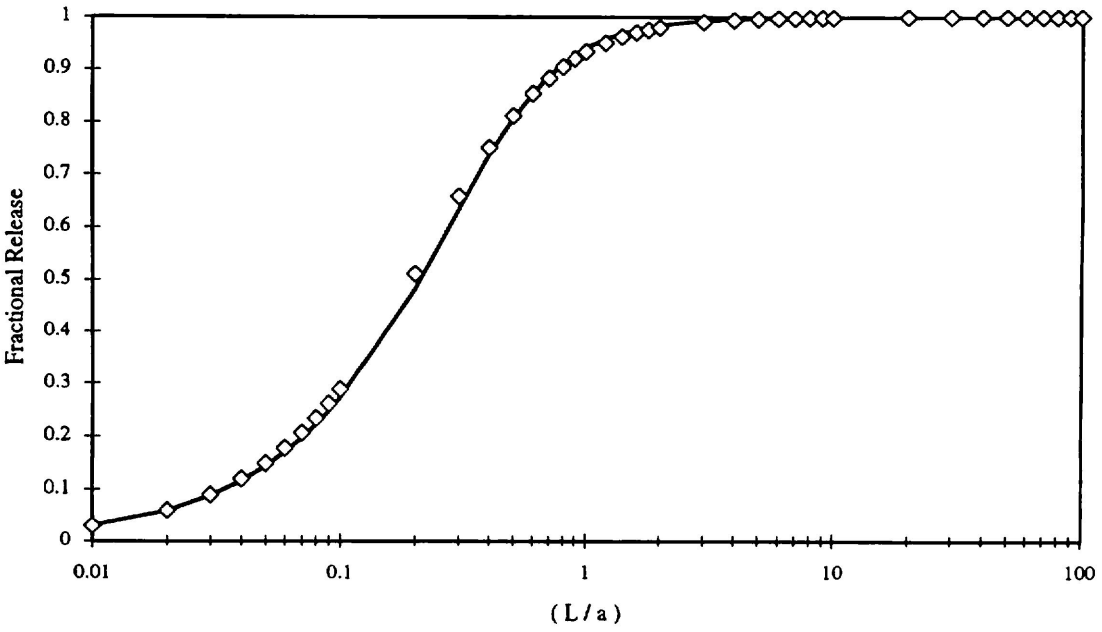

Figure 2
The probability of a short-lived fission product escaping from a spherical fuel grain. The solid line represents the diffusion theory model and the symbols are derived from probability analysis.

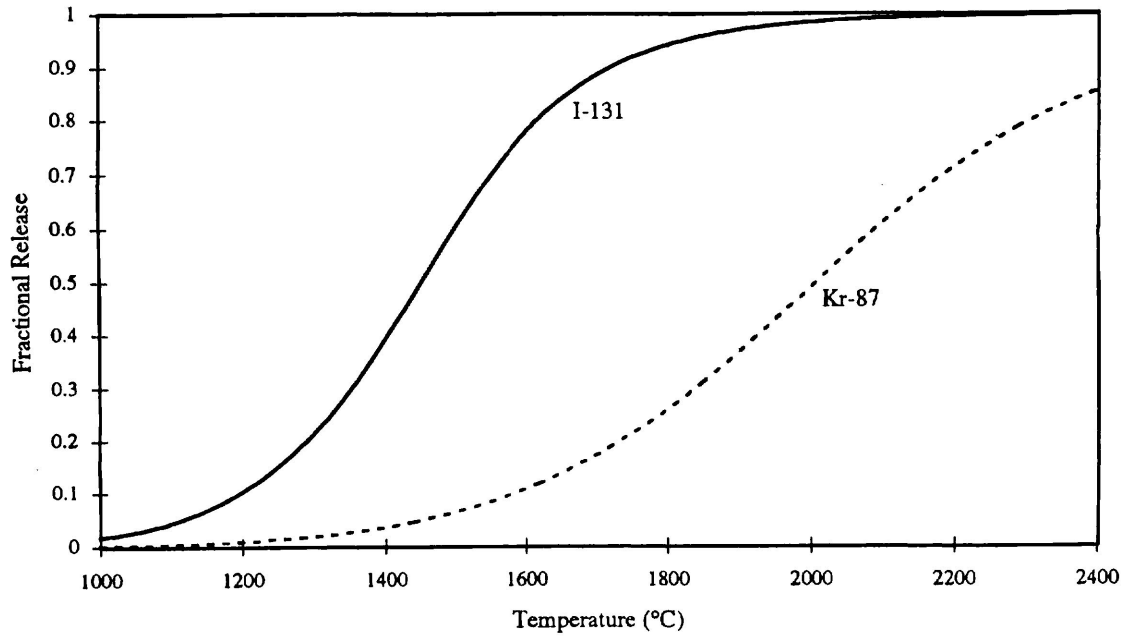

Figure 3
The release fraction (or escape probability) of I-131 and Kr-87 from a spherical fuel grain as a function of temperature calculated for thermally activated diffusion.

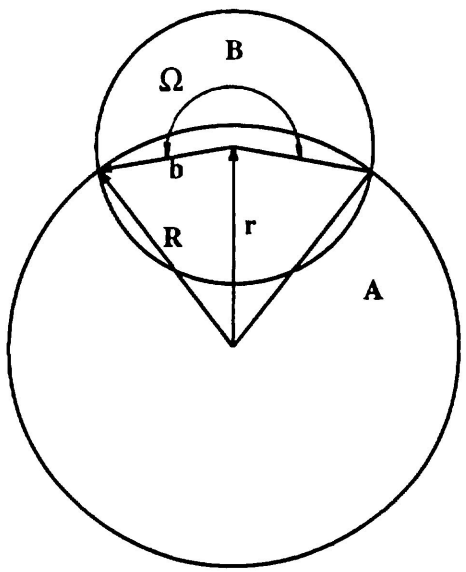

Figure C.1
Geometry for probability analysis.