SANDIA REPORT

SAND2002-3966
Unlimited Release
Printed March 2003

Analytical Modeling of Fission Product Releases by Diffusion from Multicoated Fuel Particles

Fred Gelbard

Prepared by
Sandia National Laboratories
Albuquerque, New Mexico 87185 and Livermore, California 94550

Sandia is a multiprogram laboratory operated by Sandia Corporation,

a Lockheed Martin Company, for the United States Department of Energy's
National Nuclear Security Administration under Contract DE-AC04-94AL85000.

Sandia National Laboratories

Issued by Sandia National Laboratories, operated for the United States Department of Energy by Sandia Corporation.

Available to DOE and DOE contractors from
U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831

Telephone: (865)576-8401
Facsimile: (865)576-5728
E-Mail: reports@adonis.osti.gov
Online ordering: http://www.doe.gov/bridge

Available to the public from
U.S. Department of Commerce
National Technical Information Service
5285 Port Royal Rd
Springfield, VA 22161

Telephone: (800)553-6847
Facsimile: (703)605-6900
E-Mail: orders@ntis.fedworld.gov
Online order: http://www.ntis.gov/help/ordermethods.asp?loc=7-4-0#online

SAND2002-3966
Unlimited Release
Printed March 2003

Analytical Modeling of Fission Product Releases by Diffusion from Multicoated Fuel Particles*

Fred Gelbard
Modeling and Analysis Department
Sandia National Laboratories
P. O. Box 5800
Albuquerque, NM 87185-0748

Abstract

Three levels of fission product diffusional release models are solved exactly. First, the Booth model for a homogenous uncoated spherical fuel particle is presented and an improved implementation is suggested. Second, the release from a fuel particle with a single barrier layer is derived as a simple alternative to account for a coating layer. Third, the general case of release from a multicoated fuel particle is derived and applied to a TRISO-coated fuel. Previous approaches required approximate numerical solutions for the case of an arbitrary number of coatings with arbitrary diffusivities and arbitrary coating interface conditions.

[*] This work was supported by the US Nuclear Regulatory Commission and was performed at Sandia National Laboratories, which is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the US Department of Energy under Contract DE-AC04-94AL85000.

Intentionally Left Blank

Table of Contents

I. Introduction 6

II. Fractional Release By Diffusion From A Homogeneous Spherical Fuel Particle 10

III. Lumped Fuel and Barrier Model.. 18

IV. Exact Multilayer Model... 26

V. TRISO-Coated Fuel Example... 35

VI. References.. 39

I. Introduction

Nuclear fuels encased in multicoated barriers are being considered for advanced High Temperature Gas Reactors (HTGRs). This fuel configuration has the potential to enhance safety by containing fission products within the fuel. However, even if the barriers remain structurally intact, experiments have shown that at high temperatures a significant fraction of the fission products may escape through the barriers [Kurata et al., 1981; Hayashi and Fukuda, 1989; Schenk et al., 1990].

Modeling all the details of fission product release from multicoated fuels is complicated by the variety of phenomena that occur. First, due to manufacturing limitations, the geometry is not exactly a system of perfectly uniform spherical coatings [Minato et al., 1994; Petti et al., 2002]. Furthermore, under a temperature gradient, the system deforms due to the so-called “amoeba effect” [Iwamoto et al., 1972; Stansfield et al., 1975]. Second, due to stresses, coatings may crack or delaminate [Miller et al., 2001]. Third, there are also reports of fission products reacting with barrier coatings, resulting in nonuniform variations in the barrier thickness [Smith, 1979a, 1979b; Tiegs, 1982; Lauf et al., 1984]. Fourth, at high temperatures, diffusion through the barriers can be significant [Kurata et al.1981; Hayashi and Fukuda, 1989]. Finally, due to temperature variations across the fuel particle, mass transport is not entirely uncoupled from heat transfer.

Clearly, models that are restricted to only diffusive mass transfer of fission products do not capture all the phenomena. However, surprisingly, such models, with assumed perfect spherical symmetry, have successfully modeled releases even from a fuel particle that has been pierced with a laser-drilled hole [Amian and Stover, 1982]. Therefore, there is utility in advancing such models for quantifying the effects of fission product diffusion.

Booth [1957] developed a popular diffusion model for uncoated spherical grains. To utilize this approach for fuel elements that are more than an order of magnitude larger, variations in the diffusivity are spatially averaged. An advantage of the Booth model is that the release fraction is a simple analytical expression, thus eliminating extensive computations. However, for multicoated fuels, only numerical solutions [Brown and Faircloth, 1976; Kurata, 1981; Amian and Stover, 1982], or semi-analytic approximations [Smith et al., 1977] have been documented in the literature. In all these cases, significant computational resources are required and the results are limited by discretization or approximation errors.

For reactor safety analyses, accurate but computationally efficient release models from multicoated fuels are needed. In this work, a new exact analytical solution for diffusion through a multicoated fuel is given. Thus, discretization and approximation errors are eliminated. Furthermore, the solution can be rapidly determined for an arbitrary number of layers, with arbitrary diffusivities for each fission product in each layer, with an arbitrary initial fission product concentration in each layer, and with arbitrary concentration discontinuities at interfaces between layers. The exact solution is based on work developed for heat conduction through composite media [Mulholland and Cobble, 1972; Ozisik, 1980]. However, for fission product release, the ability to specify discontinuous concentrations at coating interfaces has been added. An alternate approach for obtaining exact solutions to multilayer problems is based on Laplace Transforms [Carslaw and Jaeger, Chapter 12.8, 1976]. However, this approach is difficult to apply for more than two layers [Ozisik, p. 262, 1989]. Furthermore, the transform technique is complicated for nonzero initial conditions, and all the examples presented, even for two layers, are limited to zero initial conditions [Carslaw and Jaeger, Chapter 12.8, 1976].

We begin in Section II by deriving in detail the Booth model for the release fraction [Booth, 1957]. One reason for presenting the derivation is that the original work is not readily available. More importantly, we show that the current method for utilizing the Booth model [Ramamurthi and Kuhlman, 1990], suffers from a significant discontinuity that is readily corrected. Next we show in Section III that an exact expression for the release fraction can be obtained for a single coating used as a diffusive barrier. This lumped fuel and barrier solution assumes that all the resistance is in this barrier. An earlier analysis restricted the diffusivity in the barrier to be identical to that in the fuel [Rosenberg et al., 1965]. The steady state behavior of a single coating barrier has been analyzed [Dunlap and Gulden, 1968]. Fukuda and Iwamoto [1975] report that Baurmann [1970] developed a transient single coating barrier model. Unfortunately, we could not obtain a copy of Baurmann’s work. An exact solution is then derived in Section IV for the general case of an arbitrary number of layers. We show that in the limit of a single layer with a much lower diffusivity than the fuel, the exact multilayer model approaches the simpler lumped fuel and barrier model. To fully demonstrate the capabilities of the solution developed in this work, the simulation of a release from a TRISO-coated fuel geometry is given in Section V.

II. Fractional Release By Diffusion From A Homogeneous Spherical Fuel Particle

The governing equation for Fickian diffusion and radioactive decay in spherical coordinates is given by

$$\frac{\partial c}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left[D r^2 \frac{\partial c}{\partial r} \right] - \lambda c \qquad (0 \leq r \leq R) \qquad \text{(II.1)}$$

where R is the radius of the sphere, D is the diffusivity, and is λ the radioactive decay rate. D is parameterized as

$$D = D_0 \exp\left[-\frac{E_0}{R_{gas} T} \right] \qquad \text{(II.2)}$$

where D_0 and E_0 are fitting parameters, T is the temperature, and R_{gas} is the ideal gas constant. Because the temperature may vary with time, we assume that D is a known function of time. We assume that there is no spatial variation of temperature, and thus the diffusivity is independent of r. The boundary and initial conditions are,

$$c = 0 \qquad (r = R,\ t > 0), \qquad \text{(II.3)}$$

$$c \text{ finite} \quad (r = 0), \text{ and} \tag{II.4}$$

$$c = c_0 \quad (0 \leq r \leq R,\ t = 0). \tag{II.5}$$

The condition given in Eq. (II.3) provides the maximum release. Alternatively, by introducing another parameter, a convective boundary condition can be imposed such that the flux is proportional to the concentration on the surface of the sphere [Fukuda and Iwanmoto, 1975].

Using the transformation $p = \exp(\lambda t)cr$, the governing equation can be given as

$$\frac{\partial p}{\partial t} = D\frac{\partial^2 p}{\partial r^2}. \tag{II.6}$$

The transformed boundary and initial conditions are,

$$p = 0 \qquad (r = R,\ t > 0), \tag{II.7}$$

$$p = 0 \qquad (r = 0), \text{ and} \tag{II.8}$$

$$p = rc_0 \quad (0 \leq r \leq R,\ t = 0). \tag{II.9}$$

The transformed system can be solved in terms of an eigenfunction expansion that satisfies the transformed boundary conditions. Let p(r,t) = Q(r)T(t), where Q is a function only of r, and T is a function of only t. Then the separated form of Eq. (II.6) is given by

$$\frac{T'}{DT} = \frac{Q''}{Q} = -\beta^2 \tag{II.10}$$

where β is a constant, and the primes represent differentiation with respect to the appropriate independent variable. To within a multiplicative constant, the solution to Eq. (II.10) for Q that satisfies Eq. (II.8) is

$$Q = \sin(\beta r). \tag{II.11}$$

From Eqs. (II.7) and (II.8) β can take on only discrete values of

$$\beta_n = n\pi/R \qquad (n = 1, 2, \ldots). \tag{II.12}$$

By using these values of β_n, the solution of Eq. (II.10) for T (within a multiplicative constant) can be given by

$$T = \exp\left[-\frac{n^2\pi^2\overline{D}t}{R^2}\right] \tag{II.13}$$

where the time-averaged diffusivity is given by

$$\overline{D} = \frac{\int_0^t D(x)\,dx}{t}. \tag{II.14}$$

Combining Eqs. (II.11) and (II.13) results in

$$p = \sum_{n=1}^{\infty} A_n \sin\left(\frac{n\pi r}{R}\right) \exp\left[-\frac{n^2\pi^2\overline{D}t}{R^2}\right] \tag{II.15}$$

where A_n are constants determined by the initial condition. Imposing the initial condition and transforming back to spherical coordinates results in

$$\frac{c}{c_0} = \frac{2e^{-\lambda t}R}{\pi r} \sum_{n=1}^{\infty} \frac{(-1)^{n+1}}{n} \sin\left(\frac{n\pi r}{R}\right) \exp\left[-\frac{n^2\pi^2\overline{D}t}{R^2}\right]. \tag{II.16}$$

The mass of fission product remaining in the sphere is given by

$$M(t) = \int_0^R 4\pi r^2 c\, dr = \frac{8c_0 R^3 e^{-\lambda t}}{\pi} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left[-\frac{n^2\pi^2\overline{D}t}{R^2}\right]. \qquad \text{(II.17)}$$

The fraction released or decayed is therefore given by

$$f = 1 - \frac{M(t)}{M(0)} = 1 - \frac{6e^{-\lambda t}}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left[-\frac{n^2\pi^2\overline{D}t}{R^2}\right]. \qquad \text{(II.18)}$$

An expression for the diffusivity variation with time is needed to determine $\overline{D}$, and then the fraction released or decayed as given by Eq. (II.18) can be determined. If Eq. (II.14) can be evaluated analytically, then an explicit expression for $\overline{D}$ can be substituted into Eq. (II.18). The case of constant D and no radioactive decay results in the classical Booth model [Ramamurthi and Kuhlman, p. 41, 1990]. However, the solution is actually not restricted to constant D, as long as the time-averaged diffusivity is used for D.

II.A. Booth Model for Constant Diffusivity

For constant diffusivity, $\overline{D} = D$, and $\lambda = 0$, the fraction released is given by

$$f_{Booth}\left(\frac{t}{\tau}\right) = 1 - \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}\exp\left[-\frac{n^2\pi^2 t}{\tau}\right] \qquad (D \text{ constant}, \lambda = 0). \qquad (II.19)$$

This function is plotted in Fig. II.1 as a function of the dimensionless time $t/\tau = Dt/R^2$. For t/τ much greater than approximately 0.1, only the first term in the series is needed, and thus

$$f_{Booth}\left(\frac{t}{\tau}\right) \to 1 - \frac{6}{\pi^2}\exp\left[-\frac{\pi^2 t}{\tau}\right] \qquad (t/\tau \gg 0.1, D \text{ constant, and } \lambda = 0) \quad (II.20)$$

where D is the effective diffusivity, and t is time. For $t/\tau < 1/\pi^2$ the fraction released is given by [Ramamurthi and Kuhlman, p. 41, 1990]

$$f_{Booth}\left(\frac{t}{\tau}\right) \to \sqrt{\frac{36t}{\pi\tau}} - 3\frac{t}{\tau} \qquad (t/\tau < 1/\pi^2, D \text{ constant, and } \lambda = 0) \qquad (II.21)$$

These two approximations are compared to the exact expression in Fig. II.1. Notice that these two approximations are adequate for representing the fraction released for all values of dimensionless time. The transition dimensionless time between the two approximations is given as $1/\pi^2 = 0.101$ [Ramamurthi and Kuhlman, p. 41, 1990]. However, the approximations do not intersect, and are not even closest at this value of dimensionless time. Therefore, if these two functional forms are retained we suggest instead using $t/\tau = 0.155$ as the transition point. At this value of dimensionless time the two approximations are closest.

Because numerical solutions may have convergence problems with discontinuous functions, an even better approach would be to use approximations that result in a continuous release fraction with dimensionless time. This can be easily achieved by determining a scale factor s given by

$$s\left[\sqrt{\frac{36t}{\pi\tau}} - 3\frac{t}{\tau}\right] = 1 - \frac{6}{\pi^2}\exp\left[-\frac{\pi^2 t}{\tau}\right]. \qquad \text{(II.22)}$$

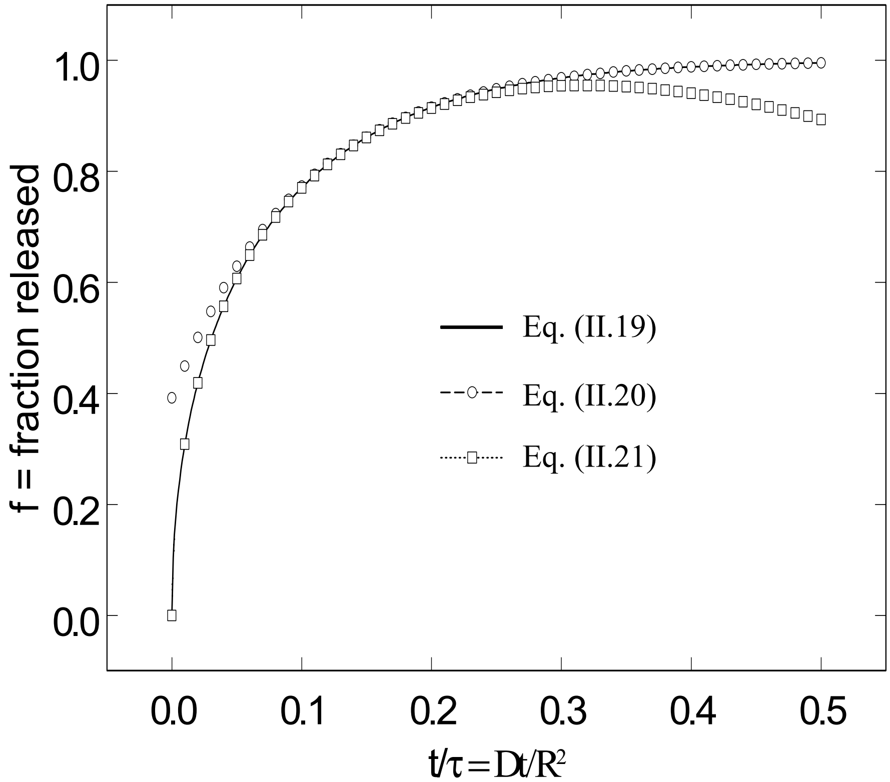
If the continuity condition is applied at $t/\tau = 0.155$, then

$$s = \frac{1 - \frac{6}{\pi^2}\exp\left[-0.155\pi^2\right]}{\left[\sqrt{\frac{(0.155)(36)}{\pi}} - 3(0.155)\right]} = 1.0006964 \qquad \text{(II.23)}$$

and for thus t/τ < 0.155 the approximation is

$$f_{\text{Booth}} = 1.0006964\left[\sqrt{\frac{36t}{\pi\tau}} - 3\frac{t}{\tau}\right] \quad (t/\tau < 0.155,\ \text{D constant},\ \lambda = 0) \qquad \text{(II.24)}$$

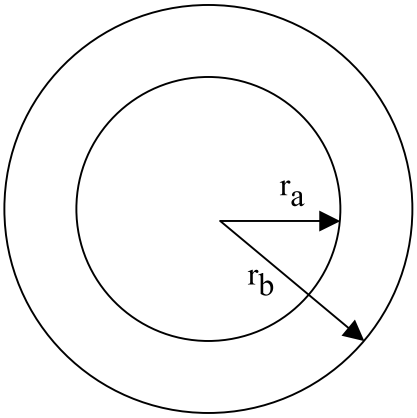
Figure II.1. Fraction released for Booth model



III. Lumped Fuel and Barrier Model

Consider the same fuel sphere of radius r_a, enclosed by diffusive barrier layer in the region $r_a \leq r \leq r_b$ as shown in Fig. III.1.

Figure III.1. Schematic of coupled lumped fuel and barrier model.



Because the dominant resistance is in the barrier layer, we will approximate the fission product concentration as uniform in the fuel. Thus the initial fission product mass can be given by

$$M(0) = \frac{4}{3}\pi r_a^3 K_1 c_a \tag{III.1}$$

where K_1 is a constant, and c_a is the concentrations in the regions $0 \le r \le r_a$. With this formulation there may be multiple interior layers, each with a different fission product concentration, but with an initial average fission product concentration of c_a. The concentration just inside the diffusive barrier at the $r = r_b$ will be approximated as

$$c\big|_{r=r_a} = K_2 c_a \qquad \text{(III.2)}$$

where K_2 is a constant. (The constants K_1 and K_2 are introduced to allow for discontinuous concentrations.) Equation (II.1) is still applicable in the diffusive barrier, however the boundary conditions are now

$$c = K_2 c_a \qquad (r = r_a,\ t > 0), \qquad \text{(III.3)}$$

$$c = 0 \qquad (r = r_b), \text{ and} \qquad \text{(III.4)}$$

$$c = 0 \qquad (r_a \le r \le r_b,\ t = 0). \qquad \text{(III.5)}$$

Eqs. (II.1), (III.3), (III.4), and (III.5) can be expressed in terms of dimensionless variables by defining a dimensionless distance, time, and scaled concentration respectively as

$$R = \frac{r - r_a}{r_b - r_a}, \qquad \text{(III.6)}$$

$$T = \frac{tD}{(r_b - r_a)^2}\text{, and} \qquad \text{(III.7)}$$

$$C = \frac{rc}{r_a K_2 c_a}\,. \qquad \text{(III.8)}$$

With these definitions, the dimensionless equation, and boundary and initial conditions are,

$$\frac{\partial C}{\partial T} = \frac{\partial^2 C}{\partial R^2} \qquad \text{(III.9)}$$

$$C = 1 \qquad (R = 0,\ T > 0), \qquad \text{(III.10)}$$

$$C = 0 \qquad (R = 1,\ T > 0),\ \text{and} \qquad \text{(III.11)}$$

$$C = 0 \qquad (0 \le R \le 1,\ T = 0). \qquad \text{(III.12)}$$

The solution to this system is given by

$$C = 1 - R - \frac{2}{\pi}\sum_{n=1}^{\infty}\frac{1}{n}\sin(n\pi R)\exp\left[-n^2\pi^2 T\right]. \qquad \text{(III.13)}$$

Thus in the region $r_a \le r \le r_b$

$$\frac{c}{c_a} = \frac{K_2 r_a}{r_a + R(r_b - r_a)} \left\{ 1 - R - \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{1}{n} \sin(n\pi R) \exp\left[-n^2 \pi^2 T \right] \right\} \tag{III.14}$$

From a mass balance, the rate of mass change in the region $r < r_a$ is given by the mass rate that diffuses into the barrier. Thus

$$\frac{d}{dt}\left[\frac{4}{3} \pi K_1 c_a r_a^3 \right] = 4\pi r^2 D \left.\frac{\partial c}{\partial r}\right|_{r=r_a} = \frac{4\pi r^2 D}{(r_b - r_a)} \left.\frac{\partial c}{\partial R}\right|_{R=0}, \tag{III.15}$$

which reduces to

$$\frac{dc_a}{dT} = -\frac{3K_2 c_a (r_b - r_a)}{K_1 r_a} \left\{ \frac{r_b}{r_a} + 2 \sum_{n=1}^{\infty} \exp\left[-n^2 \pi^2 T \right] \right\}. \tag{III.16}$$

Integrating with the initial condition of $c_b = c_{bo}$ at $T = 0$, results in

$$c_a = c_{ao} \exp\left\{ -\frac{K_2 (r_b - r_a)}{K_1 r_a} \left[\frac{3T r_b}{r_a} + f_{Booth}(T) \right] \right\}. \tag{III.17}$$

The mass contained in the system is given by

$$M(T) = \frac{4}{3}\pi r_a^3 K_1 c_a + \int_{r_a}^{r_b} 4\pi r^2 c \, dr \, . \qquad \text{(III.18)}$$

The integral is zero at T = 0, and for T > 0 can be given by

$$\int_{r_b}^{r_c} 4\pi r^2 c \, dr = 4\pi(r_b - r_a)K_2 r_a^2 c_a \left\{ \frac{1}{2} - \frac{4}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{(2n-1)^2} \exp\left[-(2n-1)^2 \pi^2 T \right] \right\} \quad .$$

$$+ 4\pi(r_b - r_a)^2 K_2 r_a c_a \left\{ \frac{1}{6} - \frac{2}{\pi^2} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \exp\left[-n^2 \pi^2 T \right] \right\}$$

(III.19)

The fraction released by a diffusive barrier system is given by

$$f_{db}\left(T, K_1, K_2, \frac{r_b}{r_a} \right) = 1 - \frac{M(T)}{M(0)} \qquad \text{(III.20)}$$

where the subscript “db” stands for diffusive barrier. In dimensional form there are seven variables: t, D, c_{bo}, r_a, r_b, K_1 and K_2. However, in dimensionless form we see from Eq.

(III.20) that the fraction released can be expressed in terms of only four dimensionless variables. This is a significant simplification of the problem.

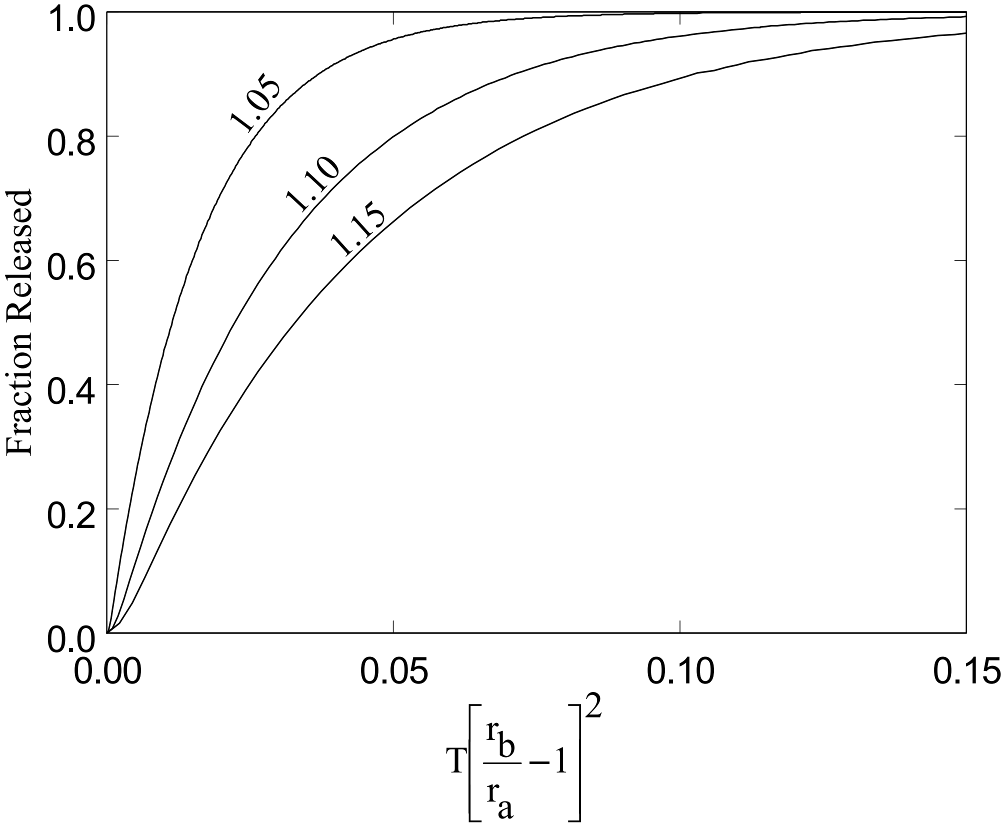
As an example, consider a typical layer thickness for SiC for various TRISO-coated fuels as given in Table III.1. Notice that the thickness of the SiC layer is on the order of 10% of the radius of the sphere interior to the SiC layer. Thus we will consider the range $1.05 < r_b/r_a < 1.15$ by holding the inner radius fixed and varying the outer radius. In addition, for convenience we specify that K_1 and K_2 are unity. With these parameters, we see from Fig. III.2 that for $T[r_b/r_a - 1]^2$ greater than approximately 0.1, the fraction released is more than 0.8. (The x-axis variable is chosen this way such that this variable is independent of the outer radius. Thus the x-axis variable is independent of the curve selected in the figure.) Also, the greater the thickness of the barrier compared to the size of the fuel, the slower the release.

Table III.1. Layer thickness for various fuels (expressed in micrometers, Petti et al., 2002).

Layer	NPR	MODUL	HTTR	HTR-10	HRB-4
Fuel*	200	500	600	501	367
Buffer	101.7	92	60	84.8	45
IPyC Inner Pyrolytic Carbon	52.9	39	30	43	31
SiC Silicon Carbide	35.3	35	25	40	34
OPyC Outer Pyrolytic Carbon	39.1	40	45	38.5	30

*Fuel thickness is for the fuel diameter.

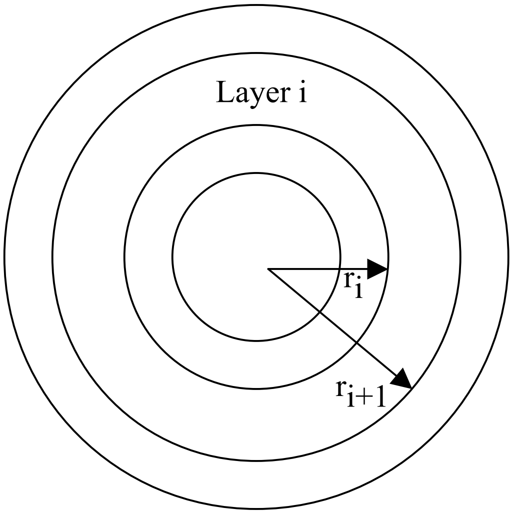
Figure III.2. Fraction released for lumped fuel and barrier model. The curves as labeled are for r_b/r_a = 1.05, 1.10, and 1.15.



IV. Exact Multilayer Model

TRISO-coated fuel consists of multiple layers, and a full analysis requires accounting for diffusional resistance in each layer. Such systems can be modeled as a layered sphere in which the layers may have different thicknesses and diffusivities for each fission product. A schematic of such a layered sphere is shown in Figure IV.1 with four layers (i.e. three coatings).

Figure IV.1. Schematic of coupled multilayer model.



For layer i, the inner and outer radii are r_i and r_{i+1}, respectively, where $1 \leq i \leq L$, and $r_1 = 0$. The governing equation for the concentration in each layer is given by

$$\frac{\partial c_i}{\partial t} = \frac{D_i}{r^2}\frac{\partial}{\partial r}\left[r^2 \frac{\partial c_i}{\partial r}\right] \qquad (r_i < r < r_{i+1},\ i = 1, 2, \ldots, L) \qquad \text{(IV.1)}$$

where D_i is the diffusion coefficient in layer i. It is assumed that the diffusion coefficient is constant within each layer, but may vary from layer to layer. The boundary condition at $r = r_{L+1}$, $c = 0$, and at $r = 0$, c is finite. The interface conditions at $r = r_{i+1}$, $i = 1, 2, 3, \ldots, L-1$ are given by continuity of the flux

$$D_i \frac{\partial c_i}{\partial r} = D_{i+1} \frac{\partial c_{i+1}}{\partial r} \qquad (r = r_{i+1},\ i = 1, 2, \ldots, L-1) \qquad \text{(IV.2)}$$

and a general concentration relationship

$$\gamma_i c_i = \gamma_{i+1} c_{i+1} \qquad (r = r_{i+1},\ i = 1, 2, \ldots, L-1). \qquad \text{(IV.3)}$$

The partition factor γ_i, is introduced to allow for discontinuous concentrations at the interface [Dunlap and Gulden, 1968; Brown and Faircloth, 1976]. An analytical eigenfunction expansion approach for solving similar multilayer problems in heat conduction has been reported [Mulholland and Cobble, 1972; Ozisik, 1980]. We now extend this expansion approach to allow for the interface condition given in Eq. (IV.3).

We begin by separating Eq. (IV.1) into a product of a spatial function in layer i given by $\Psi_i(r)$, and a temporal function given by $\Gamma(t)$. In anticipation of an eigenvalue problem for the n-th eigenvalue, the separated governing equation becomes

$$\frac{D_i}{r^2 \Psi_{i,n}} \frac{d}{dr}\left(r^2 \frac{d\Psi_{i,n}}{dr} \right) = \frac{1}{\Gamma(t)} \frac{d\Gamma}{dt} = -\beta_n^2 \quad (r_i < r < r_{i+1},\ i = 1, 2, \ldots, L) \qquad \text{(IV.4)}$$

where β_n is a constant eigenvalue, and $n = 1, 2, \ldots, \infty$. The eigenfunctions satisfy the equation

$$\frac{1}{r^2} \frac{d}{dr}\left(r^2 \frac{d\Psi_{i,n}}{dr} \right) + \frac{\beta_n^2}{D_i} \Psi_{i,n} = 0 \quad (r_i < r < r_{i+1},\ i = 1, 2, \ldots, L) \qquad \text{(IV.5)}$$

and are given by

$$\Psi_{i,n}(r) = \frac{A_{i,n}}{r} \sin\left(\frac{r\beta_n}{\sqrt{D_i}} \right) + \frac{B_{i,n}}{r} \cos\left(\frac{r\beta_n}{\sqrt{D_i}} \right) \qquad \text{(IV.6)}$$

where $A_{i,n}$, $B_{i,n}$ are constants that are determined from the boundary and interface conditions. The eigenfunctions also satisfy the orthogonality relationship given by

$$\sum_{i=1}^{L} \gamma_i \int_{r_i}^{r_{i+1}} r^2 \Psi_{i,n} \Psi_{i,s} \, dr = 0 \qquad (n \neq s) \tag{IV.7}$$

where n and s are the indices of two different eigenfunctions. (A proof of this orthogonality relationship can be obtained by following the approach given in Ozisik [pp. 331-334, 1980].) The solution for Γ is readily obtained to within a multiplicative constant as

$$\Gamma(t) = \exp\left(-\beta_n^2 t\right). \tag{IV.8}$$

The exact solution for the concentration in layer i can now be expressed as,

$$\begin{aligned} c_i(r,t) &= \sum_{n=1}^{\infty} C_n \exp\left(-t\beta_n^2\right) \Psi_{i,n}(r) \\ &= \sum_{n=1}^{\infty} \frac{C_n \exp\left(-t\beta_n^2\right)}{r} \left[A_{i,n} \sin\left(\frac{r\beta_n}{\sqrt{D_i}}\right) + B_{i,n} \cos\left(\frac{r\beta_n}{\sqrt{D_i}}\right) \right] \end{aligned} \quad . \tag{IV.9}$$

where C_n is a constant. For L layers, there are L-1 interfaces, and thus 2(L-1) homogeneous interface equations for $A_{i,n}$ and $B_{i,n}$ given by Eqs. (IV.2) and (IV.3). In addition, the end conditions at $r = 0$ and $r = r_{L+1}$ provide two more homogeneous equations for a total of 2L equations. From the condition at $r = 0$, we can immediately assign $B_{1,n} = 0$, thus eliminating one coefficient and one equation. The matrix must be singular for this system of 2L-1 homogeneous equations to have a solution. This singularity requirement determines the eigenvalues β_n. For each eigenvalue, the coefficients $A_{i,n}$ and $B_{i,n}$ are the null vectors of the singular matrix. The singular matrix times the null vector is always a vector of zeroes, without all the elements of the null vector being zero. These null vectors are unique only to within a multiplicative constant, and may be normalized to one. (Ozisik [1980] suggests arbitrarily selecting an element of the null vector to be unity to resolve nonuniqueness. This approach is not recommended because the selected element may be zero, and by using such an arbitrary assignment the resulting solution may be wrong.)

The constants C_n are determined by satisfying the initial conditions and by using the orthogonality property of the eigenfunctions given in Eq. (IV.7). The result is

$$C_n = \frac{1}{N_n} \sum_{i=1}^{L} \gamma_i \int_{r_i}^{r_{i+1}} r c_i(r, t=0) \left[A_{i,n} \sin\left(\frac{\beta_n r}{\sqrt{D_i}} \right) + B_{i,n} \cos\left(\frac{\beta_n r}{\sqrt{D_i}} \right) \right] dr \,. \quad \text{(IV.10)}$$

For c initially constant in each layer i, $c_i(r, t = 0) = c_i(0)$. For this initial condition the integral for C_n can be determined analytically, and thus

$$C_n = \frac{1}{N_n \beta_n^2} \sum_{i=1}^{L} \gamma_i c_i(0) D_i \left\{ \begin{array}{l} A_{i,n}\left[\sin\left(\frac{\beta_n r}{\sqrt{D_i}}\right) - \left(\frac{\beta_n r}{\sqrt{D_i}}\right)\cos\left(\frac{\beta_n r}{\sqrt{D_i}}\right)\right] \\ + B_{i,n}\left[\cos\left(\frac{\beta_n r}{\sqrt{D_i}}\right) - \left(\frac{\beta_n r}{\sqrt{D_i}}\right)\sin\left(\frac{\beta_n r}{\sqrt{D_i}}\right)\right] \end{array} \right\}_{r=r_i}^{r=r_{i+1}} , \tag{IV.11}$$

where the norm is given by,

$$N_n = \sum_{j=1}^{L} \gamma_j \int_{r_j}^{r_{j+1}} \left[A_{j,n} \sin\left(\frac{r\beta_n}{\sqrt{D_j}}\right) + B_{j,n} \cos\left(\frac{r\beta_n}{\sqrt{D_j}}\right) \right]^2 dr . \tag{IV.12}$$

Eq. (IV.12) can be integrated analytically to give,

$$N_n = \sum_{j=1}^{L} \frac{\gamma_j \sqrt{D_j}}{\beta_n} \left[\begin{array}{l} \dfrac{\beta_n \left(r_{j+1} - r_j \right)}{2\sqrt{D_j}} \left[A_{i,n}^2 + B_{i,n}^2 \right] \\[2ex] + \left\{ \sin\left(\dfrac{2 r_j \beta_n}{\sqrt{D_j}} \right) - \sin\left(\dfrac{2 r_{j+1} \beta_n}{\sqrt{D_j}} \right) \right\} \dfrac{\left[A_{i,n}^2 - B_{i,n}^2 \right]}{4} \\[2ex] + A_{i,n} B_{i,n} \left[\sin^2\left(\dfrac{r_{j+1} \beta_n}{\sqrt{D_j}} \right) - \sin^2\left(\dfrac{r_j \beta_n}{\sqrt{D_j}} \right) \right] \end{array} \right] . \qquad \text{(IV.13)}$$

The mass contained in a multilayer system is given by

$$M(t) = \sum_{i=1}^{L} 4\pi \int_{r_i}^{r_{i+1}} r^2 c_i(r,t)\, dr$$

$$= \sum_{i=1}^{L} 4\pi \sum_{n=1}^{\infty} C_n \exp\left(- t\beta_n^2 \right) \left[\begin{array}{l} A_{i,n} \left\{ \dfrac{D_i}{\beta_n^2} \sin\left(\dfrac{r\beta_n}{\sqrt{D_i}} \right) - \dfrac{r\sqrt{D_i}}{\beta_n} \cos\left(\dfrac{r\beta_n}{\sqrt{D_i}} \right) \right\} \\[2ex] + B_{i,n} \left\{ \dfrac{D_i}{\beta_n^2} \cos\left(\dfrac{r\beta_n}{\sqrt{D_i}} \right) + \dfrac{r\sqrt{D_i}}{\beta_n} \sin\left(\dfrac{r\beta_n}{\sqrt{D_i}} \right) \right\} \end{array} \right]_{r_i}^{r_{i+1}} .$$

(IV.14)

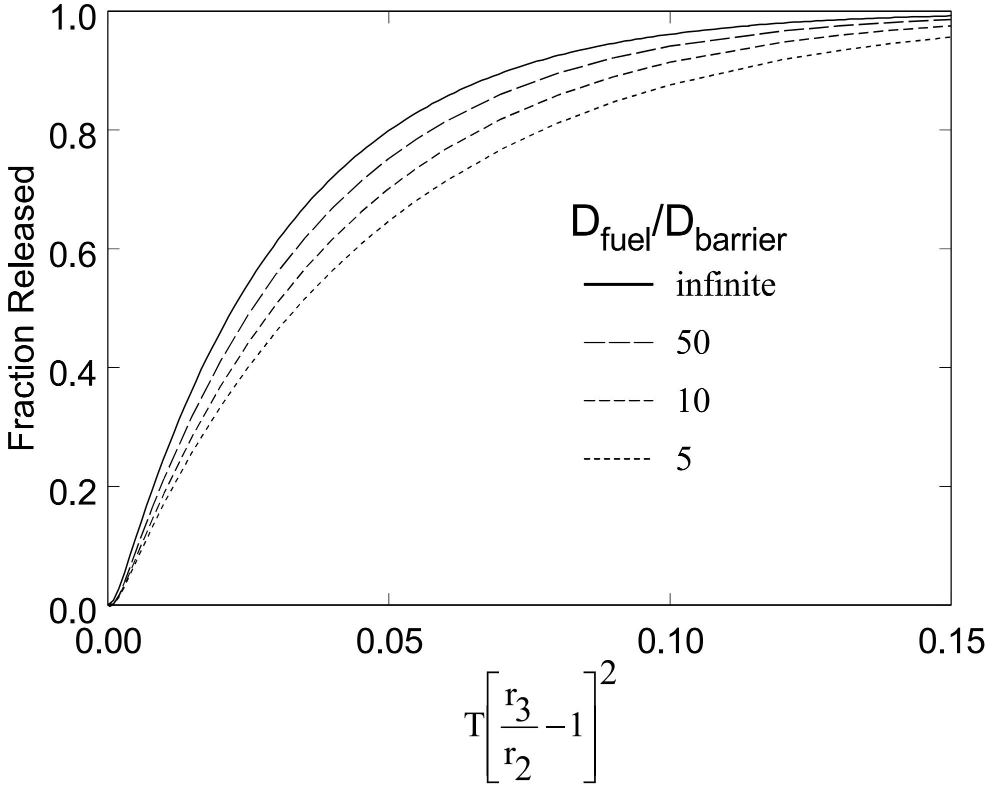
Thus the fraction released for a multilayer system is given by

$$f_{\text{multilayer}} = 1 - \frac{M(T)}{M(0)} . \qquad \text{(IV.15)}$$

The expression for $f_{\text{multilayer}}$ is exact, and thus eliminates discretization errors associated with numerical solutions for the concentration profile. However, determination of the eigenvalues β_n, does require numerical iteration. This iteration involves repeatedly evaluating the determinant of a small $(2L-1) \times (2L-1)$ matrix and finding the eigenvalue that makes the determinant zero. A simple secant technique is adequate for finding these eigenvalues. To reduce the possibility that an eigenvalue is skipped, the derivative for the determinant as a function of β at the eigenvalue is checked to see that it changes sign for successive eigenvalues. Typically, only 5 to 10 eigenvalues are needed for four significant figures of accuracy.

The general multilayer solution given by Eq. (IV.9) reduces to the two-layer coupled lumped fuel and barrier model given previously in the limit of $D_{\text{fuel}}/D_{\text{barrier}} = D_1/D_2 \to \infty$. This is shown in Figure IV.2 for $r_3/r_2 = 1.1$. The dimensionless time T is given by Eq. (III.7) with the diffusivity taken as that for the barrier layer. The solid curve was computed using Eq. (III.20), and the curves for finite D_1/D_2 were computed from Eq. (IV.15). For D_1/D_2 greater than about 50, the simpler lumped fuel and barrier model seems adequate for determining the release fraction.

Figure IV.2. Comparison of fraction released for lumped fuel and barrier model, and multilayer model for $r_3/r_2 = 1.10$, and $\gamma_1 = \gamma_2 = 1$. The limit of $D_{fuel}/D_{barrier} \rightarrow \infty$ is the solution given in Eq. (III.20). The three curves for finite $D_{fuel}/D_{barrier}$ correspond to the multilayer model.



V. TRISO-Coated Fuel Example

TRISO-coated fuel consists of a micro spherical core of UO_2, and four coating layers. These layers in order away from the fuel are (1) a buffer layer of porous pyrolytic carbon, (2) an inner dense pyrolytic carbon (IPyC) layer, (3) a silicon carbide (SiC) layer, and (4) an outer dense pyrolytic carbon (OPyC) layer [Minato et al., 1994b; Moormann, et al., 2001]. For the purposes of the calculations in this section, we consider a specific TRISO-coated fuel in which the layer outer radii are 300, 360, 390, 415, and 460 μm, respectively. We also use the suggested diffusivities of Ruthenium at 1940 C for these layers. These diffusivities are 8×10^{-13}, 2×10^{-11}, 2×10^{-12}, 1.2×10^{-13}, and 2×10^{-12} m^2/s, respectively [Kurata et al., 1981]. To demonstrate the effects of a concentration jump at the interfaces, assume that the partition coefficients are given by $2\gamma_{SiC} = \gamma_{IPyC} = \gamma_{OpyC} = \gamma_{Buffer} = \gamma_{UO_2} = 1$. Thus we expect that the Ru concentration will drop by a factor of two from the PyC layers to the SiC layer.

Consider the case in which Ru is initially uniformly distributed only in the UO_2 layer and no Ru is initially in the other layers. The evolution of the concentration profile in all the layers for this case is shown in Fig. V.1. The Ru concentration shown in the figure is normalized to the initial concentration in the fuel. After one hour of simulated time the Ru concentration in the first half of the radial distance is still mostly at the initial concentration. Due to the low diffusivity in SiC, there is a sharp concentration drop in this layer. Furthermore, due to the partition coefficient for this layer, there is also a factor of two drop in concentration from the pyrolytic carbon layers to the silicon carbide layer. This interface drop is most prominent at the IPyC-SiC interface. Because of the small concentrations at the SiC-OpyC interface, it is difficult to discern a concentration discontinuity at this interface. After four hours of simulated time the Ru concentration in the buffer and IPyC layers increases due to diffusion of Ru from the UO_2 layer. After 7 hours of simulated time, much of the Ru has diffused out of the UO_2 layer. However, it is the amount of Ru out of the layered system that is of concern, and not just the amount left in the UO_2 layer. By using Eq. (IV.15) the released fraction is readily calculated and is shown in Fig. V.2. For this simulation, about 60 hours are required for more than 95% of the Ru to be released from the system.

Figure V.1. Fission product concentration profile evolution for TRISO-coated fuel using the diffusivities for Ru at 1940 C [Kurata et al., 1981]. The vertical dashed lines indicate the location of the interface between layers.

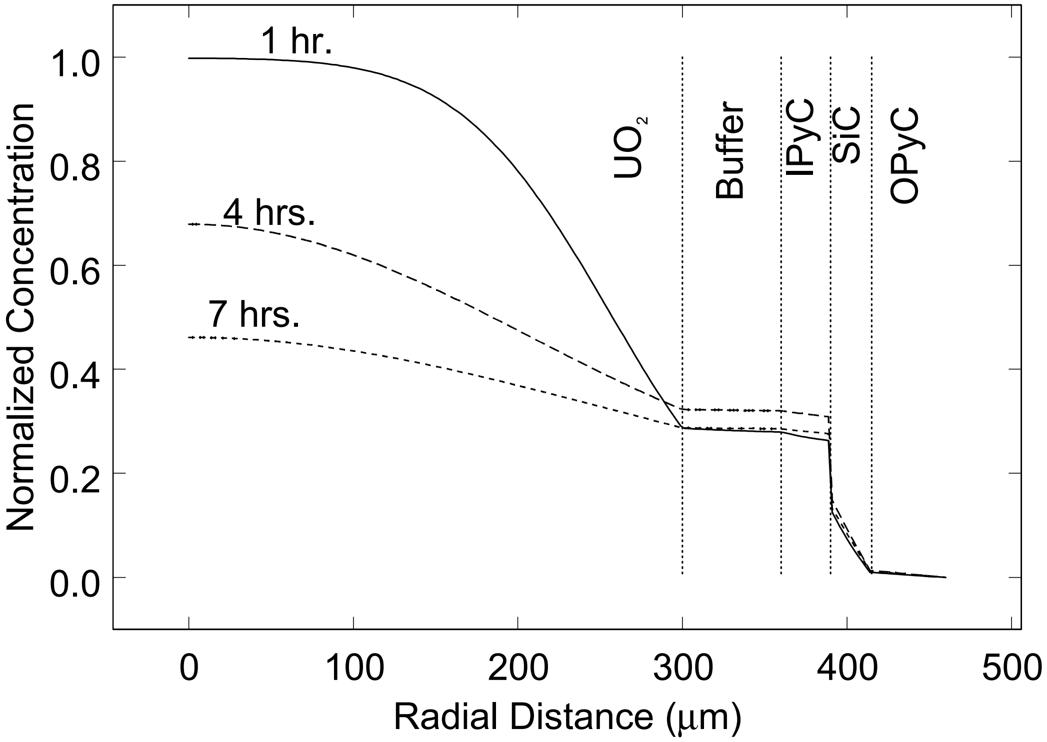
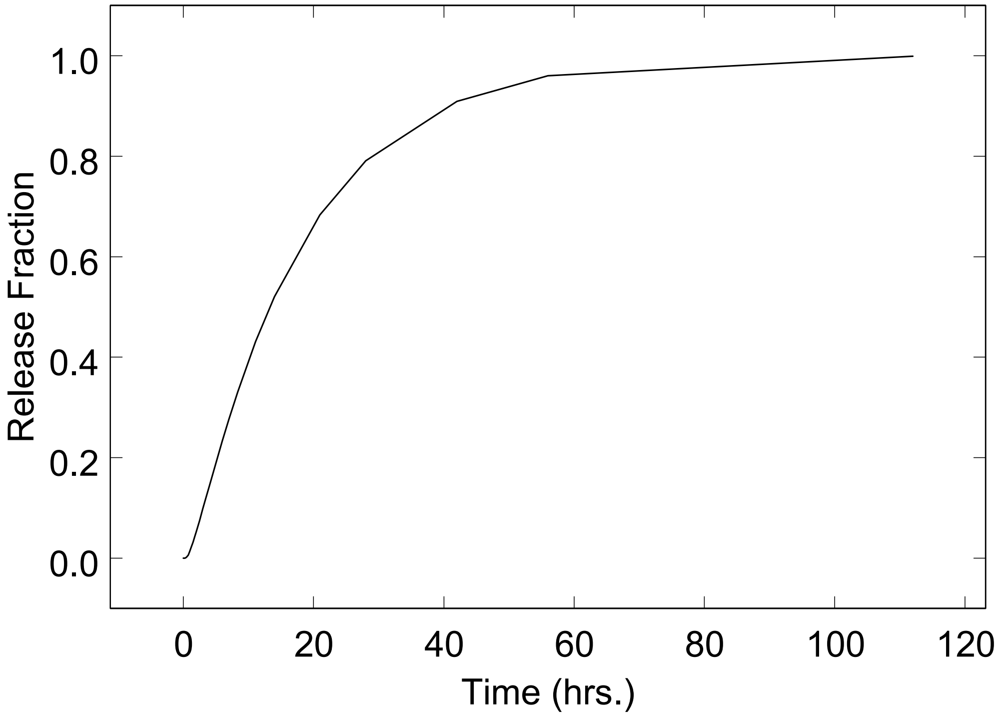


Figure V.2 Fission product release fraction for TRISO-coated fuel using the diffusivities for Ru at 1940 C [Kurata et al., 1981].



VI. References

Abramowitz, M. and I. A. Stegun (Eds.), *Handbook of Mathematical Functions*, Dover Publications, New York, 1972.

Amian, W. and D. Stover, "Cesium Release from Coated Fuel Particles with Failed Coatings," *Nuclear Technology*, **59**, 279-290, 1982.

Baurmann, K. W., JUL-685-PA, 1970. (As referenced by Fukuda and Iwamoto, 1975.)

Booth, A. H., "A Method of Calculating Fission Gas Release from UO_2 Fuel and its Application to the X-2-f Loop Test," Atomic Energy of Canada Limited - 496, 1957. (As referenced by Brown and Faircloth, 1976, and M. Maldovan, A. Denis, and R. Piotrkowski, *Nuclear Engineering and Design,* **187**, 327-337,1999.)

Brown, P. E. and R. L. Faircloth, "Metal Fission Product Behaviour in High Temperature Reactors – UO_2 Coated Fuel Particles," *Journal of Nuclear Materials*, **59**, 29-41, 1976.

Carslaw, H. S. and J. C. Jaeger, *Conduction of Heat in Solids*, Second Edition, Oxford University Press, 1976.

Dunlap, R. W. and T. D. Gulden, “Diffusion Model for Release of Fission Products from Coated Particle Fuels,” *Nuclear Science and Engineering*, **32**, 407-416, 1968.

Hayashi, K. and K. Fukuda, “Release Behavior of Fission Products from Coated Fuel Particles during Post-irradiation Heating at Abnormally High Temperatures,” JAERI-M, 89-003, 1989.

Iwamoto, K., S. Kashimura, and A. Kikuchi, “Observations of Kernel Migration in the Coated Fuel Particles under Temperature-Gradient Heating,” *Journal of Nuclear Science and Technology*, **9** (8), 465-472, 1972.

Kurata, Y. and K. Iwamoto, “Diffusion and Evaporation of Fission Products in Coated Fuel Particles,” *Journal of Nuclear Science and Technology*, **12** (3) 181-189, 1975.

Kurata, Y, K. Ikawa, and K. Iwamoto, “Fission Product Release from TRISO-Coated UO_2 Particles at 1940 to 2320 C,” *Journal of Nuclear Materials*, **98**, 107-115, 1981.

Lauf, R. J., T. B. Lindemer, and R. L. Pearson, “Out-Of-Reactor Studies of Fission Product-Silicon Carbide Interactions in HTGR Fuel Particles,” *Journal of Nuclear Materials*, **120**, 6-30, 1984.

Miller, G. K., D. A. Petti, D. J. Varacalle, and J. T. Maki, “Consideration of the Effects on Fuel Particle Behavior from Shrinkage Cracks in the Inner Pyrocarbon Layer,” *Journal of Nuclear Materials*, **295**, 205-212, 2001.

Minato, K., H. Kikuchi, K. Fukuda, N. Suzuki, H, Tomimoto, N. Kitamura, and M. Kaneko, “Internal Flaws in the Silicon Carbide Coating of Fuel Particles for High-Temperature Gas-Cooled Reactors,” *Nuclear Technology*, **106**, 342-349, 1994a.

Minato, K., T. Oagawa, K. Fukuda, M. Shimizu, Y. Tayama, and I. Takahashi, “Fission Product Behavior in Triso-Coated UO_2 Fuel Particles,” *Journal of Nuclear Materials*, **208**, 266-281, 1994b.

Moormann, R., W. Schenk, and K. Verfondern, “Source Term Estimation for Small-Sized HTRs: Status and Further Needs, Extracted from German Safety Studies,” *Nuclear Technology*, **135**, 183-192, 2001.

Mulholland, G. P. and M. H. Cobble, “Diffusion Through Composite Media,” *International Journal of Heat and Mass Transfer*, **15**, 147-160, 1972.

Ozisik, M. N., *Heat Conduction*, Chapter 8, Wiley, New York, 1980.

Ozisik, M. N., *Boundary Value Problems of Heat Conduction*, Dover Publications, p. 262, 1989.

Petti, D. A., J. T. Maki, J. Buongiorno, R. R. Hobbins, and G. K. Miller, "Key Differences in the Fabrication, Irradiation and Safety Testing of U. S. and German TRISO-coated Particle Fuel and Their Implications on Fuel Performance," Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho, INEEL/EXT-02-00300, June 2002.

Ramamurthi, M. and M. R. Kuhlman, "Final Report on Refinement of CORSOR – An Empirical In-Vessel Fission Product Release Model to U. S. Nuclear Regulatory Commission," p. 41, Battelle, Columbus, Ohio, 31 October 1990.

Rosenberg, H. S., D. L. Morrison, C. W. Townley, and D. N. Sunderman, "Postirradiation Fission-Product Release from Coated Fuel Particles," Battelle Memorial Institute, Columbus, Ohio, BMI-1734, July 7, 1965.

Schenk, W., G. Pott, and H. Nabielek, "Fuel Accident Performance Testing for Small HTRs," *Journal of Nuclear Materials*, **171**, 19-30, 1990.

Smith, C. L., "SiC-Fission Product Reactions in HTGR TRISO UC_2 and UC_xO_y Fissile Fuel I: Kinetics of Reactions in a Thermal Gradient," *Journal of the American Ceramic Society*, **62**, (11-12) 600-606, 1979a.

Smith, C. L., “SiC-Fission Product Reactions in HTGR TRISO UC_2 and UC_xO_y Fissile Fuel II: Reactions Under Isothermal Conditions,” *Journal of the American Ceramic Society*, **62**, (11-12) 607-613, 1979b.

Smith, P. D., R. G. Steinke, and D. D. Jensen, “Release of Metallic Fission Products from Multilayered Coated Particles,” *Nuclear Technology*, **34**, 475-482, 1977.

Stansfield, O. M., C. B. Scott, and J. Chin, “Kernel Migration in Coated Carbide Fuel Particles,” *Nuclear Technology*, **25**, 517-530, 1975.

Tiegs, T. N., “Fission Product Pd-SiC Interaction in Irradiated Coated-Particle Fuels,” *Nuclear Technology*, **57**, 389-398, 1982.

Distribution

USNRC	Farouk Eltawila, MS T10-E32 (Attn: Management Analyst)
USNRC	Charles Tinkler
USNRC	Richard Lee
MS0736	T. E. Blejwas, 6400
MS0744	D. A. Powers, 6400
MS0742	J. R. Guth, 6401
MS0748	G. E. Rochau, 6415
MS0748	J. E. Cash, 6415
MS0748	N. E. Bixler, 6415
MS0748	F. Gelbard, 6415
MS0748	P. Longmire, 6415
MS0748	R. O. Gauntt, 6515
MS9018	Central Technical Files, 8945-1 (1)
MS0899	Technical Library, 9616 (2)
MS0612	Review & Approval Desk, 9612 (1) For DOE/OSTI