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**Transient Diffusion Through a Spherical Shell
into Its Finite Spherical Core**

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OAK RIDGE NATIONAL LABORATORY
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COMPUTER SCIENCES DIVISION

**TRANSIENT DIFFUSION THROUGH A SPHERICAL SHELL
INTO ITS FINITE SPHERICAL CORE**

**M. Reeves* and J. S. Tolliver
Computer Sciences Division**

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ABSTRACT

The problem of the diffusion of gas through a spherical shell into its spherical core is solved by using the Laplace transform technique. The transformed solution is obtained and then inverted to the corresponding solution in real space. This real space solution is slowly converging for small times. Therefore, an alternate expression is obtained that is appropriate and more rapidly convergent for small times. As an example, the first solution is used to calculate the theoretical gas content of the core of High-Temperature Gas-Cooled Reactor (HTGR) HISO-coated fuel particles from fuel-particle batch CR2261/HT after 850°C anneals in helium.

I. INTRODUCTION AND STATEMENT OF THE PROBLEM

High-Temperature Gas-Cooled Reactor (HTGR) bi-coated fuel particles consist of an inner core of ThO_2 fuel which is coated with a layer of porous carbon and a layer of dense carbon. The porous layer, called the buffer, serves as a reservoir for fission gasses while the dense outer layer serves as a pressure vessel. The high temperature inert gas permeability of the fuel particles is of interest and has been studied experimentally at Oak Ridge National Laboratory and the Oak Ridge Y-12 Plant by Morgan and Powell (1977) and by Morgan, Eatherly, and Powell (1977). This report considers the theoretical aspects of the problem by examining the diffusion of gas through a dense spherical shell into a low density spherical core. Nuclear Technology, Volume 35, is a special issue related to HTGR fuel particles and may be referred to for more detailed information about the fuel particles.

Figure 1 shows a composite sphere submerged in a gas of constant concentration c_0 . The geometry is characterized both by the radii a and b and by the porosities n and s . It is assumed that the pore dimensions of the inner region are sufficiently large compared to the mean free path of the gas that mass transport occurs essentially instantaneously. Such an assumption allows one to concentrate on the shell, which, with diffusivity D , is much more resistant to penetration by the gas.

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Fig. 1. Cross-sectional view of the spherical shell and its inner core together with geometrical and material parameters.

Diffusion within this shell is characterized by the equation

$$\frac{\partial c}{\partial t} = \frac{D}{r} \frac{\partial^2}{\partial r^2} (rc) \quad (1)$$

where c is the concentration, r is the radial distance, and t is the time. The initial condition is

$$c(r, t = 0) = 0 \quad (2a)$$

and the two boundary conditions are

$$c(r = b, t) = nc_0 \quad (2b)$$

and

$$\left[q \frac{\partial c}{\partial t} - D \frac{\partial c}{\partial r} \right]_{r=a} = 0 \quad (2c)$$

where

$$q = \frac{Sa}{3n} \quad (3)$$

Physically Eq. (2c) is based upon the characteristics of the inner core. Due to the assumption of instantaneous diffusion, the concentration at the inner core is independent of position and is proportional to the shell concentration evaluated at the interface. Because of the finiteness of the inner core, the rate of change of the concentration is then related to the interface flux, $-D_0 c / r$. The constant of proportionality q is related to the porosities and the radius of the inner core, as shown in Eq. (3).

Mathematically the internal boundary condition expressed in Eq. (2c) can be a source of considerable difficulty. The predicament here arises from the fact that eigenfunctions constructed to satisfy this boundary condition and the external boundary condition [Eq. (2b)] are not orthogonal. Thus it is quite difficult to incorporate the initial condition [Eq. (2a)] into this formulation. Carslaw and Jaeger [1959] mention this difficulty in the footnote on page 240. The solution, however, is easily obtained using the Laplace transformation

$$\bar{c}(\lambda) = L[c(t)] = \int_0^{\infty} e^{-\lambda t} c(t) dt \quad (4)$$

where λ is complex.

II. SOLUTION OF THE SUBSIDIARY EQUATION

By applying the Laplace transformation to Eqs. (1) and (2), one obtains the subsidiary equation

$$\frac{D}{r} \frac{\partial^2}{\partial r^2} (r\bar{c}) = \lambda \bar{c} \quad (5)$$

with

$$\bar{c}(r = b) = nc_0/\lambda \quad (6a)$$

and

$$\left[\lambda q \bar{c} - D \frac{\partial \bar{c}}{\partial r} \right]_{r=a} = 0 \quad (6b)$$

Integration of Eq. (5) yields an equation of the form

$$r\bar{c} = A \sinh(kr) + B \cosh(kr) \quad (7)$$

where

$$k = \sqrt{\lambda/D} \quad (8)$$

The boundary conditions of Eq. (6) permit determination of constants A and B so that the solution of the subsidiary equation may be written

$$r\bar{c} = bnc_0 \bar{Q} \quad (9a)$$

where

$$\bar{Q} = \frac{1}{2} \frac{f(k_{+1})}{f(k_{-1})} \quad (9b)$$

and

$$f(k_{\pm 1}) = (k_{\pm 1}^2 q_0 + 1) \sinh(k_{\pm 1}) + k_{\pm 1} \cosh(k_{\pm 1}) \quad (9c)$$

Quantities μ and λ are simple combinations of the radii:

$$\mu = r-a \text{ and } \lambda = b-a \quad (10)$$

III. INVERSION OF THE LAPLACE TRANSFORM SOLUTION

Here the inverse Laplace transform of Eq. (9) is desired. Isolating the dependence on λ and t in Eq. (9a), one obtains

$$rc(t) = \mathcal{L}^{-1}[rc(\cdot)] = \text{bnc}_e Q(t) \quad (11a)$$

and from the inversion theorem

$$Q(t) = \mathcal{L}^{-1}[\bar{Q}(\lambda)] = \frac{1}{2\pi i} \int_{\gamma-i\infty}^{\gamma+i\infty} e^{\lambda t} \bar{Q}(\lambda) d\lambda \quad (11b)$$

Parameter γ , a real quantity, is so chosen that all singularities lie to the left of the line $(\gamma-i\infty, \gamma+i\infty)$ in the complex plane (see Fig. 2).

The integral of Eq. (11b) is most easily evaluated by modifying the path of integration. Since $\bar{Q}$ may be shown to be a single-valued function of λ , the closed contour Γ (see Fig. 2) may be used. Further, since the infinite semicircular path within Γ contributes nothing, one may write Eq. (11b) as follows:

$$Q(t) = \frac{1}{2\pi i} \int_{\Gamma} e^{\lambda t} \bar{Q}(\lambda) d\lambda \quad (12)$$

The complex integral is therefore transformed into a form suitable for application of the Cauchy Residue Theorem, yielding

$$\int_{\Gamma} e^{\lambda t} \bar{Q}(\lambda) d\lambda = 2\pi i \sum_{j=0}^{\infty} K_j \quad (13)$$

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Fig. 2. Complex plane of the Laplace-transformation parameter λ . Locations of some of the poles in quantity Q are indicated.

where the K_j 's are the residues of the integrand evaluated at the poles enclosed within contour Γ .

The location of the poles may be determined by inspection of the denominator of $\bar{Q}$ in Eq. (9t). They are confined to the nonpositive real axis (see Fig. 2) as follows:

$$\lambda_0 = 0 \text{ and } \lambda_j = -D_{\alpha_j}^2, j=1,2,\dots \quad (14a)$$

where the α_j 's are the positive roots of the transcendental equation

$$\tan \alpha_j = \frac{\alpha_j}{1 - q_3 - 1} \quad (14b)$$

Residues of these poles are given by the expressions

$$K_0 = \frac{r}{b} \quad (15a)$$

and

$$K_j = \left[e^{\lambda_j t} f(k, \alpha_j) / \frac{d}{d\lambda} f(k, \lambda) \right]_{\lambda = -D_{\alpha_j}^2}, j=1,2,\dots \quad (15b)$$

The desired solution is obtained by combining Eqs. (9), (11), (12), (13), and (15). It is

$$rc = bnc_0 \left[\frac{r}{b} + \sum_{j=1}^{\infty} g(\alpha_j, \alpha_j) e^{-D_{\alpha_j}^2 t} \right] \quad (16a)$$

where

$$g(a, \rho) = \frac{2}{a} \frac{(1 - \rho^2 qa) \sin \rho a + qa \cos \rho a}{(2q + \rho) \sin \rho a + (\rho^2 qa - b) \cos \rho a} \quad (16b)$$

By letting a and b approach infinity in such a manner that their difference ρ remains finite, it may be easily shown that Eq. (16) reduces to the solution used by Jenkins, et al. [1969] for a region bounded by parallel planes. Absolute convergence, and hence convergence, may be established for Eq. (16a) by using a simple ratio test, except for the case $t = 0$. There the ratio test fails, and such convergence must be established by showing that the Cauchy integral exists. At small times, however, convergence may be quite slow, and a different expression is desirable.

IV. INVERSION OF THE LAPLACE TRANSFORM

FOR SMALL VALUES OF TIME

For small times one takes advantage of the sensitivity of Q to large s and k values, a fact which is apparent from the Inversion Theorem, Eq. (11), and the definition of k , Eq. (8). The idea is to expand $\bar{Q}$ in terms of negative exponentials and inverse powers of k . This procedure is the same as that used by Carslaw and Jaeger [1959] (see pp. 330-31, for example).

Using the definitions of the hyperbolic functions in terms of exponentials, quantity $\bar{Q}$ of Eq. (9b), takes the form

$$\bar{Q} = \frac{1}{2} \frac{e^{-k(z-a)} + e^{-k(z+a)}}{1 - e^{-2kz}} \quad (17)$$

Here

$$z = \frac{d-x+x^2}{d+x+x^2} \quad (18)$$

where

$$d = q/a = \frac{s}{3n} \quad (19)$$

from Eq. (3) and

$$x = 1/ka \quad (20)$$

Expanding the denominator of Eq. (17) as

$$(1 - e^{-2kz})^{-1} = \sum_{m=0}^{\infty} e^{-2mkz} \quad (21)$$

and multiplying by the numerator, one has

$$\bar{Q} = \frac{1}{\lambda} \sum_{m=0}^{\infty} \left\{ e^m e^{-k[(2m+1)\lambda - \mu]} - e^{m+1} e^{-k[(2m+1)\lambda + \mu]} \right\} \quad (22)$$

Quantity e^m may be analyzed further by using the binomial expansions

$$(\delta + y)^{-m} = \sum_{i=0}^{\infty} (-1)^i \binom{m+i-1}{i} \delta^{-(m+i)} y^i \quad (23a)$$

and

$$(\delta + y)^m = \sum_{i=0}^m \binom{m}{i} \delta^{m-i} y^i \quad (23b)$$

in which

$$\binom{m}{i} = \frac{m!}{i!(m-i)!} \quad (23c)$$

is the usual binomial coefficient. In the process of analyzing Eq. (18), variable y of Eq. (23) is set equal to $x + x^2$, $-x + x^2$, x , and $-x$, and parameter δ assumes the values d and unity.

After a considerable amount of algebra, one obtains the desired expression for z^m , namely

$$z^m = \sum_{w=0}^{\infty} B_{mw} x^w \quad (24)$$

where

$$B_{mw} = \sum_{i=0}^{I_u} \sum_{j=0}^{J_u} \sum_{p=p_c}^{P_u} (-1)^{w-j} \binom{m+w-i-j-1}{m-1} \quad (25a)$$

$$\cdot \binom{w-i-j}{j} \binom{m}{i-p} \binom{i-p}{p} d^{-(w-p-j)}$$

The summation limits are given by

$$I_u = \min(w, 2m) \quad , \quad (25b)$$

$$J_u = (w-i)/2 \quad , \quad (25c)$$

$$P_c = \max(0, i-m) \quad , \quad (25d)$$

$$P_u = i/2 \quad . \quad (25e)$$

For $m = 0$, Eq. (25a) yields $B_{0w} = \delta_{0w}$ where δ_{0w} is the Kronecker delta.

Combining Eqs. (20), (22), and (24), one obtains

$$\bar{Q} = \sum_{m,w=0}^{\infty} \left[B_{mw} \bar{R}_{mw}(-\phi) - B_{m+1,w} \bar{R}_{mw}(\phi) \right] \quad (26)$$

in which

$$\bar{R}_{mw}(\rho) = \frac{1}{\lambda} \frac{1}{(ka)^5} e^{-k\omega_m(\rho)} \quad (27)$$

and

$$\omega_m(\rho) = (2m+1)\ell + \rho \quad (28)$$

Taking note of the definition of k in Eq. (8), one may write

$$\bar{R}_{mw}(\rho) = (\sqrt{D}/a)^w \frac{1}{\lambda^{1+s/2}} e^{-k\omega_m(\rho)} \quad (29)$$

The inverse Laplace transform of $\bar{R}$ may be obtained from tables. (See, for example, Appendix V in Carslaw and Jaeger [1959].) The result is

$$R_{mw}(\rho) = \left(\frac{2\sqrt{Dt}}{a} \right)^w i^w \text{erfc} \frac{\omega_m(\rho)}{2\sqrt{Dt}} \quad (30)$$

where $i^w \text{erfc}(x)$ represents the w -th integral of the complementary error function. (See, for example, Appendix II of Carslaw and Jaeger [1959].)

By analogy with Eqs. (26) and with the aid of Eq. (30), quantity Q may be written as

$$Q = \sum_{m,w=0}^{\infty} \left(\frac{2\sqrt{Dt}}{a} \right)^w \left[B_{mw} i^w \text{erfc} \frac{\omega_m(-\rho)}{2\sqrt{Dt}} - B_{m+1,w} i^w \text{erfc} \frac{\omega_m(\rho)}{2\sqrt{Dt}} \right] \quad (31a)$$

According to Eq. (11a), the desired solution is thus

$$rc = bnc_0 Q \quad (31b)$$

On the surface it appears that Eq. (31) recovers the initial condition, Eq. (2a), since at $t = 0$ the complementary error functions equal zero. It also seems reasonable that Eq. (31a) converges whenever $t \leq T$, where T is a radius of convergence yet to be determined. To establish these two points rigorously, however, is quite difficult and has not been done to date. The major problem comes from the coefficient B_{mw} defined by Eqs. (25). Whenever indices m and/or w approach infinity, quantity B also becomes an infinite series. Furthermore, the binomial coefficients of infinite indices become quotients of infinite products. Thus to investigate the behavior of B_{mw} as either or both of its indices go to infinity, one must investigate infinite sums of infinite products. A further complication is the possibility of negative terms in Eq. (25a). The solution expressed in Eq. (31) should therefore be investigated numerically prior to implementation.

The only useful, albeit non-rigorous, estimate of the radius of convergence which the authors have found is determined by setting all of the complementary error functions in Eq. (31a) to unity. (If the terms of the series were non-negative, this procedure would produce a reference series for use in a comparison test. One would then have a rigorous proof of convergence within the region of convergence of the reference series.) Such a series

may be summed exactly by simply reversing the binomial-expansion process used to develop Eqs. (24) and (25) from Eq. (18). The radius of convergence for Eq. (23a), with $y = 2\sqrt{Dt}/a$, provides the desired relation:

$$T \approx \frac{ad}{2D} = \frac{an}{6Ds} \quad (32)$$

where Eq. (19) is used to obtain the last expression.

V. APPLICATIONS

The gas content of the inner core is of interest. As stated in Section I, it is assumed that diffusion in the core is instantaneous compared to diffusion through the shell so the concentration can be taken to be uniform throughout the core. The boundary conditions require that the pore concentrations on either side of the interface between the core and shell be equal. The gas uptake then is this uniform pore concentration times the volume of the inner core times the porosity of the inner core. Thus, from Eq. (16a) with $r=a$ one obtains the uptake U given by

$$U = \sqrt{U} \quad (33a)$$

where

$$\gamma = \frac{4}{3} a^2 b c_0 s \quad (33b)$$

$$\bar{U} = \frac{a}{b} - \sum_{j=0}^{\infty} g(\alpha_j, 0) e^{-D \alpha_j^2 t} \quad (33c)$$

and $g(\alpha, 0)$ is determined from Eq. (16b) with $\rho=0$, and the α_j 's are given by Eq. (14b). A computer program has been developed to calculate the gas uptake given by Eq. (33). The input parameters required are the dimensions a and b , the diffusivity D , the outside gas concentration c_0 , and the porosities n and s . Since c_0 appears only as a multiplicative factor in γ , these may be combined, for the purpose of calculating uptake, into the dimensions a and b and the three parameters D , q , and γ where q is the ratio $sa/3n$ defined by Eq. (3). Figure 3 illustrates two curves representing the uptake as functions of time for two different sets of parameters. The units are arbitrary; the

Fig. 3. Two sample curves showing the behavior of the theoretical gas uptake as a function of time for two different sets of parameters.

dimensions a and b and parameter γ are the same for each curve. Curve No. 1 has a much higher diffusivity D and also a larger value for the ratio q and would be fit well by a first-order approximation $U = A(1 - e^{-K(t-t_0)})$. Curve No. 2 demonstrates the departure from first order and indicates that the complete analysis as described in this document is necessary for proper treatment of the problem at small times. For exact treatment at even smaller times, it is necessary to calculate the uptake from Eqs. (31) of Section IV, but this has not proved necessary to date.

The uptake calculation has been combined with a data-fitting computer code to obtain the best fit of the theoretical uptake given by Eqs. (33) to experimental measurements of the gas uptake by HTGR biso-coated fuel particles. The experimental uptakes are input and the best theoretical fit is made returning the values of the diffusivity D and the porosities n and s . Figure 4 illustrates the experimental data and the theoretical fit for the helium uptake by fuel particle batch OR2261/HT at 850°C. A forthcoming report will detail the results of application of this data-fitting procedure to a series of HTGR fuel-particle gas uptake measurements.

Fig. 4. Experimental data and the theoretical fit for the helium uptake by HTGR fuel-particle batch OR2261/HT at 850°C. The theoretical fit yields $D = 4.055 \times 10^{-12} \text{ m}^2/\text{s}$, $n = 0.0471$, and $s = 0.1948$.

VI. CONCLUSION

The problem of diffusion through a spherical shell has been solved using the Laplace transformation technique. Two expressions for the solution have been obtained having the form of infinite series of special functions. The first expression is appropriate for all values of time while the second expression provides more rapid convergence at small times. Rigorous convergence has been established for all values of time in the former case, and a nonrigorous radius of convergence has been developed in the latter case. Application of the first solution has been made for the purpose of calculating the gas uptake as a function of time and for the purpose of obtaining the material parameters D , n , and s by a data-fitting process.

T	Radius of convergence of Eq. (31) ... T
t	Time ... t
U	Gas uptake of the inner core, Eq. (33a) ... U
$\bar{U}$	Dimensionless uptake, Eq. (33c)
x	Variable defined by Eq. (20)
y	Arbitrary variable, Eq. (23)
α_j	Parameters defined by Eq. (14)
β	Function defined by Eq. (18)
γ	Path parameter for inverse Laplace transform of Section III ... T^{-1}
	Uptake parameter of Section V defined by Eq. (33b)
δ	Arbitrary parameter, Eq. (23)
Γ	Closed integration contour
λ	Laplace transformation variable ... T^{-1}
λ_j	Location of j -th pole in complex plane
ρ	Radial distance from inner surface of shell ... L
ω	Parameter defined by Eq. (28) ... L

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