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HEAT OR MASS TRANSFER-CONTROLLED DISSOLUTION OF AN ISOLATED SPHERE

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Abstract—A comprehensive set of finite-difference solutions describing the heat or mass transfer-controlled dissolution of isolated spheres is presented. The analysis is based on a generalized formulation which includes three specific classes of dissolution problems. A coordinate transformation which immobilizes the moving boundary and maps the infinite region of interest into a finite region is used to minimize the difficulties associated with a numerical analysis of this problem. Radius–time and particle lifetime results are reported for ranges of parameters which include the majority of physically important dissolution processes. In addition, the results of this investigation are used to determine the accuracy and range of applicability of various approximate analytical solutions.

NOMENCLATURE

$\hat{C}_{\hat{v}}$,	specific heat capacity at constant specific volume for outer phase;
D,	binary diffusion coefficient for outer phase;
$\Delta\hat{H}$,	specific enthalpy of phase change;
I_1,	integral defined by equation (14);
k,	thermal conductivity of outer phase;
R^*,	radius of bubble;
R_0,	initial radius of bubble;
r^*,	radial position variable;
T^*,	temperature;
T_E,	equilibrium temperature at particle surface;
T_O,	initial temperature in outer phase;
t^*,	time;
t_L,	dimensionless lifetime of particle;
$\hat{V}_I$,	partial specific volume of component I;
W,	quantity defined by equation (18).

The various definitions of N_a, N_b, R, r, t and Y are given in Table 1.

Greek letters

α,	thermal diffusivity of outer phase = $k/\rho\hat{C}_{\hat{v}}$;
β,	constant used in equation (7);
ρ_I^*,	mass density of component I;
ρ_{IE},	equilibrium mass density of component I at particle surface;
ρ_{IO},	initial mass density of component I in outer phase;
ρ,	density of outer phase;
$\hat{\rho}$,	density of particle;
τ,	correlating variable for particle lifetimes $= \dfrac{2N_a t_L \ln(1 + N_b)}{N_b}$;
ψ,	independent variable defined by equation (7).

INTRODUCTION

THE DISSOLUTION of a spherical particle controlled by the molecular diffusion of mass or conduction of heat is of considerable importance in a large number of technical problems. The lifetime of dissolving bubbles, droplets, and solid particles in an isothermal bulk phase is a prime consideration in the design of many types of processing equipment. Also, the mathematical analysis of this type of phenomenon is frequently applied in the interpretation of experiments designed to measure fundamental transport properties. Furthermore, the evaporation, condensation, sublimation, or melting of a spherical phase immersed in a second phase of the same component constitutes an important class of problems in the domain of heat transfer. In the commonly considered analysis of these related phenomena, the dissolving or shrinking of an isolated stationary spherical phase is regarded as being controlled by diffusion or conduction in an outer phase of infinite extent This is a nonlinear transport problem which involves the solution of the unsteady-state diffusion equation in the presence of a moving boundary.

Although an exact analytical solution based on a similarity transformation is available for the related process of growth from a zero initial radius [1–3], no such result has been derived for the case of spherical particle dissolution. Indeed, examination of the equations describing particle dissolution reveals that no similarity variable exists because the boundary conditions are not compatible with any one-parameter groups of transformations which render the differential equations conformally invariant since the initial particle radius is nonzero. Consequently, investigation of the nonlinear particle dissolution problem has, of necessity, proceeded by utilization of asymptotic analytical solutions [4–8], by application of weighted residual techniques to generate approximate analytical solutions [9], and by employment of finite-difference methods [7, 10, 11].

Although asymptotic and weighted residual solutions can be compact and easy to apply, their accuracy and useful range of validity are rarely available since error estimates for such solutions are usually difficult to obtain. Hence, it is clear that accurate finite-difference solutions can serve a twofold purpose. Not only do such solutions provide a quantitative description of an important physical situation, but, in addition, they serve as a basis for assessing the general applicability of the more desirable approximate analytical solutions. Accurate and comprehensive finite-difference solutions of the particle dissolution problem appear to be lacking. The results of Cable and Evans [10] have recently been shown [7] to be inaccurate and those of Readey and Cooper [11] are inaccurate and limited. The calculations of a third study [7] are also quite limited since the principal goal of that investigation was to present an improved asymptotic solution for the dissolution process.

In this paper, a relatively comprehensive set of finite-difference solutions describing the heat or mass transfer-controlled dissolution of spherical particles is presented. The ranges of conditions covered in this parametric study were determined by a survey of the characteristics of the various physical phenomena which can be classified as dissolution processes. Radius–time and particle lifetime results are reported, and the various approximate analytical solutions are examined in light of the present investigation.

FORMULATION OF EQUATIONS

In this section, we list the assumptions utilized in the derivation of the basic transport equations and present a single set of equations which, by proper definition of the variables, describes both heat and mass transfer-controlled particle dissolution. It proves convenient to divide the general class of spherical particle dissolution problems into three subclasses. Particle dissolution which is controlled by diffusion in an infinite binary bulk phase whose components have constant partial specific volumes is considered a Class I-A problem. Class I-B contains the diffusion-controlled dissolution problems with a constant density condition imposed on the outer binary phase. Finally, Class II consists of the problems for which one-component particle dissolution is controlled by heat conduction in the infinite bulk phase.

The following assumptions are utilized in the derivation of the transport equations for all three classes:

(1) The velocity field in the outer phase is purely radial.

(2) The concentration or temperature field is spherically symmetric.

(3) The origin of the coordinate system is the particle centre which is at rest.

(4) All gravitational effects are neglected.

(5) The particle is a perfect sphere isolated in an infinite bulk phase.

(6) Inertial, viscous, and surface effects are small so that the equations of motion for the system predict that the pressure is effectively uniform throughout the system for all times.

(7) The initial concentration or temperature profile in the outer phase is uniform.

(8) There exists concentration or temperature equilibrium at the phase boundary.

(9) The density of the spherical particle is constant.

(10) Heat conduction and diffusion in the outer phase are adequately described by linear constitutive equations.

For Class I-A problems, the following additional assumptions must be introduced:

(11) The dissolution process proceeds isothermally. The effect of heat released or absorbed during phase change is considered negligible.

(12) The spherical particle is effectively a one-component phase.

(13) There is no chemical reaction in the infinite bulk phase.

(14) The binary mutual diffusion coefficient in the outer phase is constant.

(15) The partial specific volumes of the two components in the outer phase are constant.

For Class I-B problems, assumptions (11)–(14) must be utilized and assumption (15) is replaced by the following statement:

(16) The density of the outer phase is constant; this means that the partial specific volumes of the two components are not only constant but equal to each other.

The appropriate additional assumptions for Class II problems are as follows:

(17) The density of the outer phase is constant.

(18) The sensible heat of the particle phase relative to the interfacial temperature is small compared to the enthalpy of phase change.

(19) The thermal conductivity and the specific heat capacity of the infinite bulk phase are constant.

(20) Viscous dissipation in the system is negligible.

(21) The specific internal energy of the component of interest is a function only of the specific entropy and the density. Consequently, only the simplest caloric equation of state is considered in this study.

From the above assumptions and the phase rule, it is clear that the interfacial concentration for Class I-A and Class I-B problems and the interfacial temperature for Class II problems are constants set by external conditions. Hence, application of the appropriate conservation laws and jump conditions at the phase boundary in conjunction with the above assumptions yields the following dimensionless set of equations for heat or mass transfer-controlled particle dissolution [7]:

$$\frac{\partial Y}{\partial t} + \frac{R^2}{r^2}(N_a - N_b)\left(\frac{\partial Y}{\partial r}\right)\left(\frac{\partial Y}{\partial r}\right)_{r=R} = \frac{\partial^2 Y}{\partial r^2} + \frac{2}{r}\frac{\partial Y}{\partial r} \tag{1}$$

$$Y(r, 0) = 0 \qquad r > 1 \tag{2}$$

$$Y(\infty, t) = 0 \qquad t \geqq 0 \tag{3}$$

$$Y(R, t) = 1 \qquad t > 0 \tag{4}$$

$$\frac{\mathrm{d}R}{\mathrm{d}t} = N_a\left(\frac{\partial Y}{\partial r}\right)_{r=R} \tag{5}$$

$$R(0) = 1. \tag{6}$$

The definitions of the variables and parameters for these equations are given for all three classes of dissolution problems in Table 1. The remainder of this paper is concerned with solution of this set of equations, both by finite-difference

Table 1. Definitions of variables and parameters

Variable or parameter	Class I-A	Class I-B	Class II
Y	$\dfrac{\rho_I^* - \rho_{IO}}{\rho_{IE} - \rho_{IO}}$	$\dfrac{\rho_I^* - \rho_{IO}}{\rho_{IE} - \rho_{IO}}$	$\dfrac{T^* - T_O}{T_E - T_O}$
t	$\dfrac{Dt^*}{R_0^2}$	$\dfrac{Dt^*}{R_0^2}$	$\dfrac{\alpha t^*}{R_0^2}$
r	$\dfrac{r^*}{R_0}$	$\dfrac{r^*}{R_0}$	$\dfrac{r^*}{R_0}$
R	$\dfrac{R^*}{R_0}$	$\dfrac{R^*}{R_0}$	$\dfrac{R^*}{R_0}$
N_a	$\dfrac{\rho_{IE} - \rho_{IO}}{\hat{\rho}(1 - \hat{V}_1\rho_{1E})}$	$\dfrac{\rho(\rho_{IE} - \rho_{IO})}{\hat{\rho}(\rho - \rho_{IE})}$	$\dfrac{\rho(T_E - T_O)\hat{C}_p}{\hat{\rho}\Delta\hat{H}}$
N_h	$\dfrac{\rho_{IE} - \rho_{IO}}{(1/\hat{V}_I) - \rho_{IE}}$	$\dfrac{\rho_{IE} - \rho_{IO}}{\rho - \rho_{IE}}$	$\dfrac{(T_E - T_O)\hat{C}_f}{\Delta\hat{H}}$

methods and by approximate analytical techniques.

FINITE-DIFFERENCE METHOD AND RESULTS

Equations (1)–(6) present two complications which must be considered in any finite-difference solution of the dissolution process, the movement of the boundary between the two phases and the infinite extent of the bulk phase. Associated with this second complication is the fact that the gradients of concentration or temperature become very large near the moving boundary and, for an accurate finite-difference representation of the differential equations, closely spaced grid points are required in this region. On the other hand, a coarse grid is adequate for the major portion of the bulk phase. It has been suggested [7] that the inaccuracies of previous numerical studies of this problem [10, 11] are due to inadequate finite-difference approximations for the concentration or temperature gradients near the surface of the spherical particle. This previous investigation demonstrated that accurate numerical solutions can be obtained if a coordinate transformation is used to immobilize the moving boundary and map the infinite region into a region of finite extent. One appropriate transformation for the dissolution problem is

$$\psi = 1 - \exp\left[-\beta(r - R)\right] \tag{7}$$

and introduction of this new independent variable converts equations (1)–(5) to the following forms:

$$\frac{1}{\beta(1-\psi)}\frac{\partial Y}{\partial t} + \left[\frac{R^2\left(1 - \dfrac{N_h}{N_a}\right)}{\left[R - \dfrac{\ln(1-\psi)}{\beta}\right]^2} - 1\right] \times \frac{\mathrm{d}R}{\mathrm{d}t}\frac{\partial Y}{\partial \psi} = \beta(1-\psi)\frac{\partial^2 Y}{\partial \psi^2} - \beta\frac{\partial Y}{\partial \psi} + \frac{2}{R - \dfrac{\ln(1-\psi)}{\beta}}\left(\frac{\partial Y}{\partial \psi}\right) \tag{8}$$

$$\frac{\mathrm{d}R}{\mathrm{d}t} = \beta N_a\left(\frac{\partial Y}{\partial \psi}\right)_{\psi=0} \tag{9}$$

$$Y(\psi, 0) = 0 \tag{10}$$

$$Y(1, t) = 0 \tag{11}$$

$$Y(0, t) = 1. \tag{12}$$

In terms of the new variable, ψ, the transport which controls the dissolution process occurs in a fixed finite region. The parameter β is a constant which permits some flexibility in the coordinate transformation and is used in distributing grid points for the most accurate representation of the differential equations. If a central difference form for derivatives with respect to ψ and a backward form for derivatives with respect to time are employed, equations (8) and (9) and the associated boundary conditions can be solved by an implicit finite-difference scheme which utilizes a Gaussian elimination method to solve a tridiagonal matrix system. The implicit finite-difference technique for the solution of parabolic partial differential equations which is used in this analysis is well known [12] and the details are not included in this paper.

The mesh sizes for the radial and time variables were varied in order to establish the convergence of the finite-difference solution to the solution of the differential equations. To obtain accurate solutions with a reasonable number of time steps, it was found expedient to start with very small time steps and increase the size of these steps as the particle dissolution process proceeds. In many cases, particularly at small values of N_a, the rate of dissolution increases near the end of the process and it was necessary to decrease the size of the time steps in this region. The absolute size of the time steps was dictated by the parameters N_a and N_b and, for most cases, approximately 600 steps in time were necessary for an accurate description of the complete dissolution process. In the ψ direction, 160 evenly spaced points were found to be adequate. For the optimum use of these grid points, the range of β was varied from 0·5 for slowly dissolving particles to a value of 20 for the cases where the rate of movement of the free surface is fast compared to the growth of the boundary layer.

Table 2. Calculated dissolving particle lifetimes

N_a	t_L	τ
	$N_b = 10^{-5}$	
10^{-8}	5·00 × 10^{7}	1·00
10^{-7}	5.00 × 10^{6}	1·00
10^{-6}	5·00 × 10^{5}	1·00
10^{-5}	4·99 × 10^{4}	0·998
10^{-4}	4·95 × 10^{3}	0·990
10^{-3}	4·84 × 10^{2}	0·968
10^{-2}	4·51 × 10^{1}	0·902
	$N_b = 10^{-3}$	
10^{-6}	5·00 × 10^{5}	1·00
10^{-5}	4·99 × 10^{4}	0·998
10^{-4}	4·95 × 10^{3}	0·990
10^{-3}	4·84 × 10^{2}	0·968
10^{-2}	4·52 × 10^{1}	0·936
10^{-1}	4·00	0·800
1	2·84 × 10^{-1}	0·568
	$N_b = 10^{-2}$	
10^{-5}	5·01 × 10^{4}	0·997
10^{-4}	4·97 × 10^{3}	0·989
10^{-3}	4·86 × 10^{2}	0·967
10^{-2}	4·65 × 10^{1}	0·925
10^{-1}	4·02	0·800
1	2·85 × 10^{-1}	0·567
10^{1}	1·61 × 10^{-2}	0·320
	$N_b = 10^{-1}$	
10^{-4}	5·21 × 10^{3}	0·993
10^{-3}	5·11 × 10^{2}	0·974
10^{-2}	4·86 × 10^{1}	0·926
10^{-1}	4·22	0·804
1	3·02 × 10^{-1}	0·576
10^{1}	1·71 × 10^{-2}	0·326
10^{2}	8·76 × 10^{-4}	0·167
	$N_b = 1$	
10^{-3}	6·90 × 10^{2}	0·956
10^{-2}	6·75 × 10^{1}	0·936
10^{-1}	6·01	0·833
1	4·51 × 10^{-1}	0·625
10^{1}	2·71 × 10^{-2}	0·376
10^{2}	1·43 × 10^{-3}	0·198
10^{3}	7·26 × 10^{-5}	0·101

The range of conditions covered in this study and the calculated dimensionless particle lifetimes are presented in Table 2. The quantity τ is a convenient correlating variable for lifetimes suggested by one of the analytical solutions described below. The majority of dissolution problems which are of practical importance fall within the range of conditions considered in this study. A graphical representation of the lifetime results is presented in Fig. 1. Maximum computer time on a Burroughs B5500 computer was 500 s. In many applications, the radius of the particle as a function of time is of interest as well as the particle lifetime, and Figs. 2–6 show the rates of dissolution for the cases considered in this study. Each of these figures corresponds to a specific value of N_b, and it is evident that, as N_a is increased, a larger fraction of the dissolution process occurs during a given fraction of the lifetime of the sphere.

A comparison of the finite-difference solutions of this study and results calculated from an asymptotic perturbation solution discussed below is presented in Fig. 7. This graph shows that the particle lifetimes calculated from the perturbation solution differ by 2 per cent or

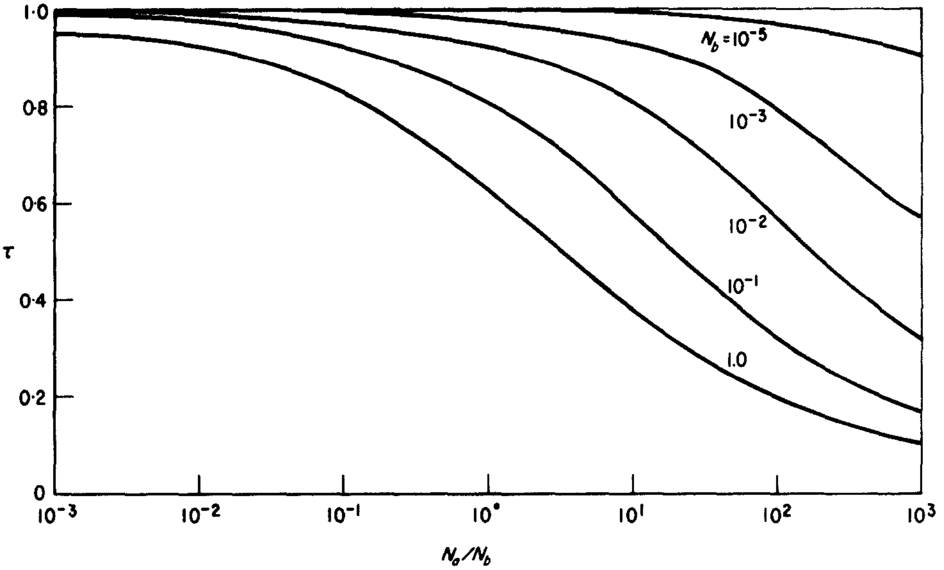

FIG. 1. Parametric correlation of particle lifetimes.

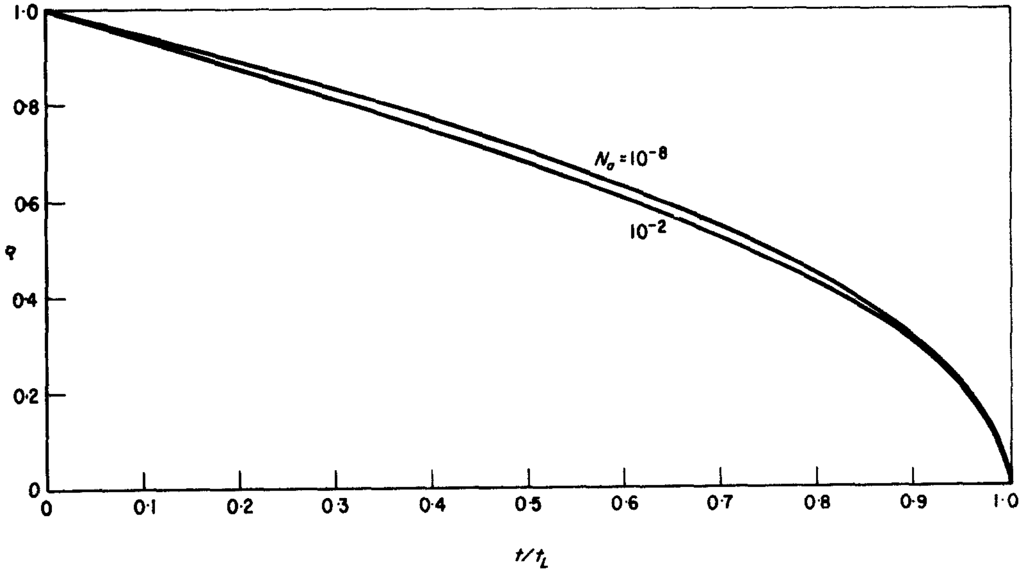

FIG. 2. Radius–time curves for $N_b = 10^{-5}$

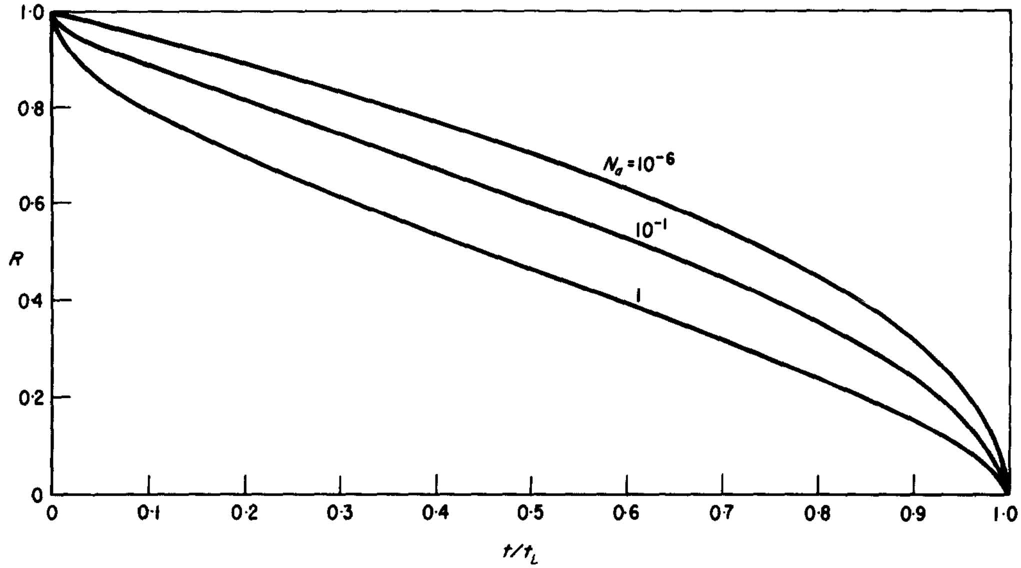

FIG. 3. Radius–time curves for $N_b = 10^{-3}$.

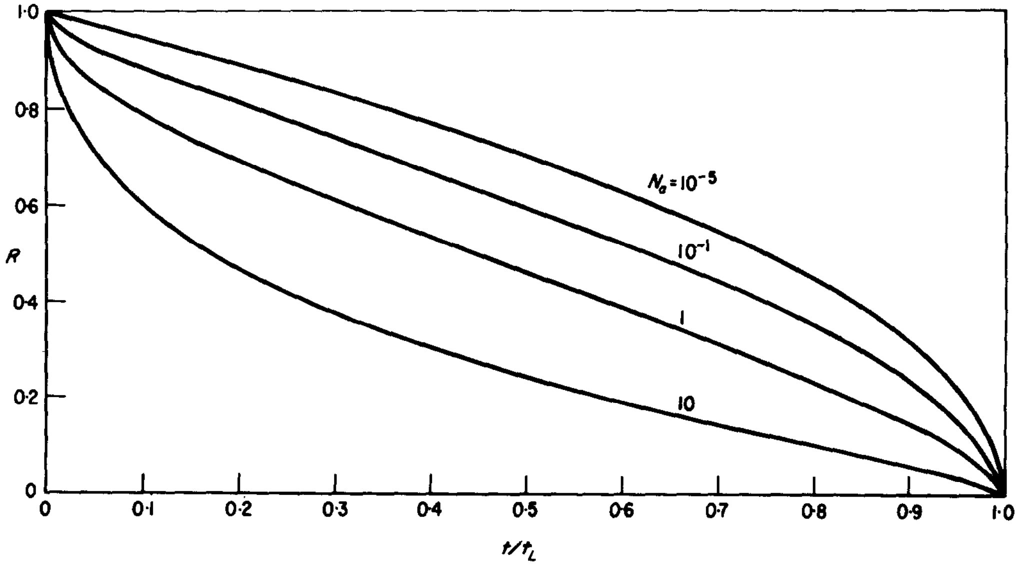

FIG. 4. Radius–time curves for $N_b = 10^{-2}$.

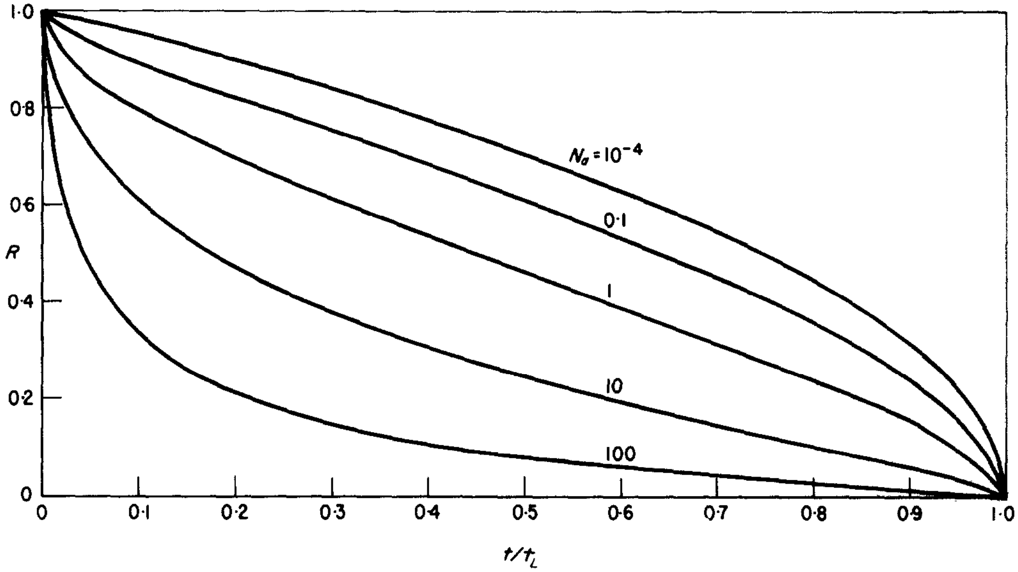

FIG. 5. Radius–time curves for $N_b = 10^{-1}$

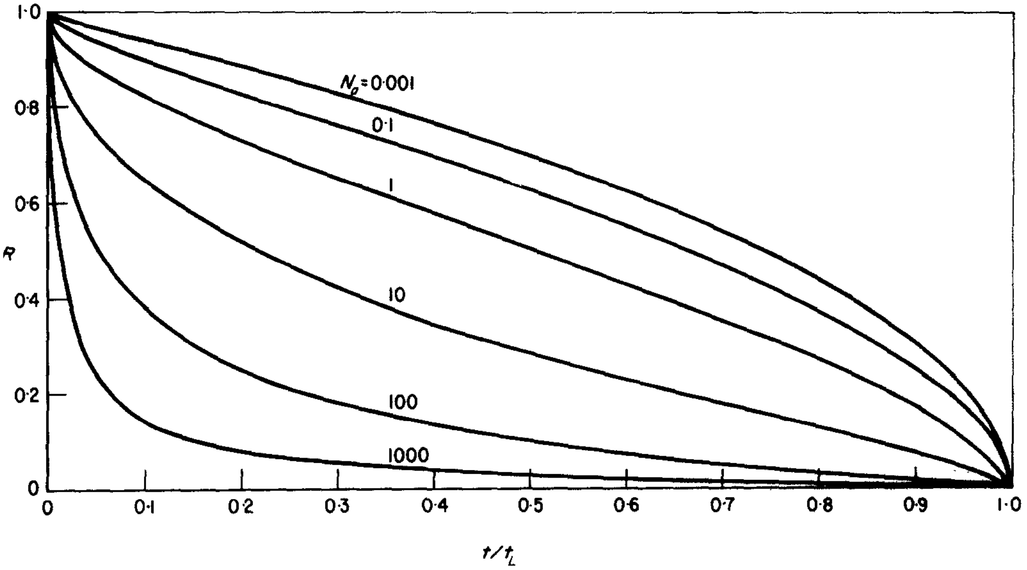

FIG. 6. Radius–time curves for $N_b = 1$.

less from those calculated from the finite-difference solution when $N_a \leqq 0{\cdot}01$ and $N_b \leqq 0{\cdot}01$. Good agreement is also found for the radius–time curves. Thus, for cases where N_a and N_b are less than 0·01, the results presented above were obtained from the perturbation solution.

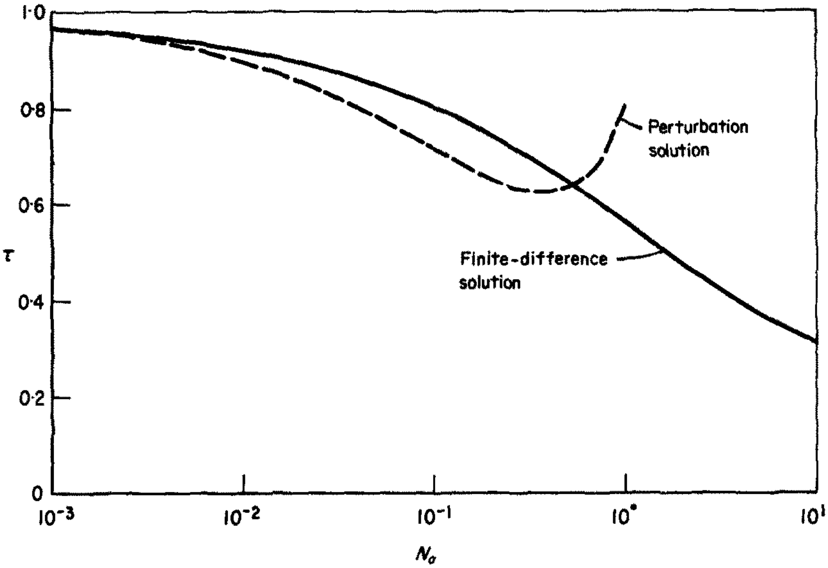

FIG. 7. Comparison of lifetimes calculated from finite-difference and perturbation solutions for $N_b = 0{\cdot}01$.

The radius–time curves and particle lifetime calculations presented here should provide accurate descriptions of the numerous physical situations which are adequately described by the basic set of equations presented above. In other important cases, some of the assumptions utilized above in deriving the dissolution equations are not valid (For example, the physical properties may be concentration or temperature dependent.), and the solutions of this paper can be regarded only as approximations to the true behavior. However, in many of these cases, the dominant mechanism of the dissolution process is described by the present calculations, and the results should be valuable for approximating physical phenomena which are not completely described by the dissolution model utilized here.

ANALYTICAL SOLUTIONS

As indicated above, analytical solutions to the particle dissolution problem have been obtained either by using asymptotic methods or by applying weighted residual techniques. Asymptotic methods yield solutions which are approximately valid when a parameter or variable of the problem is either very small or very large. Perturbation procedures are a typical and commonly used class of asymptotic methods. Such solutions, of course, by themselves describe only a part of the physically important range of dissolution processes. Weighted residual methods lead to approximate solutions which are obtained by demanding that the difference between these solutions and the exact solutions be minimized in some sense. Usually, weighted integrals of the residual are made to vanish, and the simpler weighted residual schemes are sometimes called integral methods [13]. These methods have been applied with success in obtaining accurate solutions to a variety of nonlinear conduction and diffusion problems [13], and a well formulated weighted residual solution can often provide accurate results for wide ranges of the variables and parameters of the problem. Preliminary work [9] has indicated the possibility that the integral method can satisfactorily describe the particle dissolution process. To our knowledge, however, no published comprehensive weighted residual results are available at the present time.

Basically, two types of asymptotic analytical solutions have been derived for description of particle dissolution phenomena, quasi-stationary solutions and quasi-steady-state solutions. The so-called quasi-stationary solutions have been obtained by neglecting the convective transport term of the unsteady-state diffusion or energy equation and then solving this equation with the particle surface considered stationary. The motion of the surface of the particle can then be determined by utilizing the heat or mass flux at the phase boundary as determined from the solution of the simplified transport equation. The quasi-stationary solution is apparently applicable to situations where convective transport is small compared to diffusive flow and where the rate of growth of the thickness of the concentration or temperature boundary layer surrounding a particle is fast compared to the rate of movement of the phase boundary. The quasi-stationary solution can thus be regarded as the asymptotic result for small N_a and N_b.

Epstein and Plesset [5] and Krieger *et al.* [6] both obtained quasi-stationary solutions essentially by the method described above. A more satisfactory approach is presented in a recent investigation [7] where a parameter perturbation solution of the dissolution process is derived by taking advantage of the smallness of N_a and N_b. The quasi-stationary solution is thus embedded as a first step in an orderly scheme of successive approximations. The first-order terms derived in the perturbation analysis extend the accuracy and range of validity provided by the zero-order or quasi-stationary solution and, in addition, furnish valuable insight about the structure of the solution. Furthermore, the zero-order perturbation solution, although computationally equivalent to available quasi-stationary results, is much simpler in form, apparently because incomplete higher-order terms have not been disregarded in previous investigations.

The radius–time relationship derived from the perturbation solution can be expressed as

$$R^2 = 1 - 2N_a\left(t + 2\sqrt{\frac{t}{\pi}}\right) + N_a^2\left[\frac{8t^{\frac{3}{2}}}{3\pi^{\frac{1}{2}}} + 2t + \frac{8t^{\frac{1}{2}}}{\pi^{\frac{3}{2}}} - 2I_1(t)\right] + N_aN_b[t + 2I_1(t)] \qquad (13)$$

where there is no N_b^2 term and where

$$I_1(t) = \int_0^t\int_0^\infty \frac{\operatorname{erfc}[\eta/2(t-\lambda)^{\frac{1}{2}}]}{(1+\eta)^2\sqrt{(\pi\lambda)}} \times \left[\frac{\exp(-\eta^2/4\lambda)}{\sqrt{(\pi\lambda)}} + \frac{\operatorname{erfc}(\eta/2\sqrt{\lambda})}{(1+\eta)}\right]\mathrm{d}\eta\,\mathrm{d}\lambda. \qquad (14)$$

Details of the perturbation solution, an expression for the concentration or temperature distribution around a particle, and graphs depicting the functional dependence of $I_1(t)$ are given elsewhere [7]. The zero-order perturbation result, which can be regarded as a modification of previous quasi-stationary solutions, is simply

$$R^2 = 1 - 2N_a\left(t + 2\sqrt{\frac{t}{\pi}}\right) \qquad (15)$$

It should be noted that, for given values of the two perturbation parameters, more terms will be needed to determine the particle radius with equivalent accuracy from the series solution at the longer values of time since a slower convergence rate can be expected towards the end of the interval of convergence, which can be inferred to be $0 < R < 2$. Hence, it is possible that the truncated perturbation series expansion for the radius, equation (13), will lead to significant errors near the end of the dissolution process even though the remainder of the radius–time curve is predicted quite accurately.

It was shown above that the perturbation solution predicts the particle lifetime with less than 2 per cent error for N_a and N_b both equal to or less than 0·01, and accurate radius–time curves can also be calculated from the analytical solution for this range of perturbation parameters. Furthermore, from previous results [7], it follows that the major part of the radius–time curve can be calculated very accurately for values of N_a and N_b of 0·2. Therefore, the asymptotic analytical solution will yield reasonably accurate results for values of the perturbation parameters significantly greater than 0·01, although its use for larger values of N_a and N_b cannot be recommended without some reservation.

The quasi-steady-state solution to the dissolution problem is discussed by Bankoff [4] and by Kirkaldy [2], and the more general so-called extended quasi-steady-state solution is utilized by Rosner [14] and by Spalding [8]. These solutions appear to describe spherical dissolution phenomena under conditions where the interface velocity is small so that the temperature or concentration field eventually reaches what is effectively a steady-state distribution. Thus, a quasi-steady-state result can be regarded as the asymptotic solution for small N_a and large t. Unfortunately, there appears to be no detailed investigation available of the asymptotic nature of the quasi-steady-state solution, which basically should be derivable from a perturbation analysis similar to the one utilized to elucidate the basic structure of the quasi-stationary result. Indeed, a perturbation solution might be implemented by combining a parameter perturbation technique of the type utilized above with an inverse coordinate perturbation scheme. However, since the system would presumably have to be perturbed around a steady-state solution at infinite time, a perturbation approach of this type may well be susceptible to the problems inherent to forming an asymptotic expansion which runs opposite to the natural direction of time [15]. Hence, we shall here merely present the basic form of the quasi-steady-state solution and consider the possible range of validity by comparing it with the finite-difference results.

The extended quasi-steady-state result is the solution of

$$\frac{\mathrm{d}^2Y}{\mathrm{d}r^2} + \frac{2}{r}\frac{\mathrm{d}Y}{\mathrm{d}r} + \frac{R^2N_b}{r^2}\left(\frac{\partial Y}{\partial r}\right)_{r=R}\frac{\mathrm{d}Y}{\mathrm{d}r} = 0 \qquad (16)$$

subject to equations (3) and (4) with R^2 and $(\partial Y/\partial r)_{r=R}$ regarded as constants. The solution can be expressed as

$$Y = \frac{1 - \mathrm{e}^{W/r}}{1 - \mathrm{e}^{W/R}} \qquad (17)$$

with

$$W = R^2N_b\left(\frac{\partial Y}{\partial r}\right)_{r=R} \qquad (18)$$

Thus, from equations (5), (6), (17) and (18), it follows that

$$R^2 = 1 - \frac{2N_at\ln(1 + N_b)}{N_b} \qquad (19)$$

and the particle lifetime can be expressed as

$$t_L = \frac{N_b}{2N_a\ln(1 + N_b)}. \qquad (20)$$

A less general but more widely used quasi-steady-state solution is the result which is valid for small values of N_b as well as small values of N_a and large values of t. The equations corresponding to equations (19) and (20) for this case are simply

$$R^2 = 1 - 2N_at \qquad (21)$$

$$t_L = \frac{1}{2N_a}. \qquad (22)$$

In Table 3, the first-order perturbation solution, the zero-order or quasi-stationary solution, and the extended quasi-steady-state solution are compared with the finite-difference calculation for $N_a = N_b = 0{\cdot}01$. As would be expected, the two-term perturbation result is clearly superior. The quasi-stationary solution becomes significantly inaccurate near the end of the dissolution process and the extended quasi-steady-state result systematically and substantially underestimates the rate of particle dissolution. A similar conclusion about the extended quasi-steady-state solution can be drawn from Table 4 where it is compared to a finite-difference solution for $N_a = 0{\cdot}01$ and $N_b = 1$. Hence, results calculated from equation (19) are significantly in error even for values of N_a as low as 0·01. The possible ranges of N_a and N_b where equation (19) might be expected to yield good results can be obtained from Fig. 1. The extended quasi-steady-state result predicts $\tau = 1$; from Fig. 1 it is possible to determine, for each N_b, how accurately this solution predicts the particle lifetime for various values of N_a.

Table 3. Evaluation of analytical solutions for $N_a = N_b = 0{\cdot}01$

t	R^2			
	Finite-difference	Equation (13)	Equation (15)	Equation (19)
0·1	0·9910	0·9909	0·9909	0·9980
1·0	0·9581	0·9580	0·9574	0·9801
5·0	0·8531	0·8530	0·8495	0·9005
10·0	0·7370	0·7368	0·7286	0·8010
15·0	0·6269	0·6264	0·6126	0·7015
20·0	0·5203	0·5192	0·4991	0·6020
25·0	0·4164	0·4142	0·3872	0·5025
30·0	0·3149	0·3109	0·2764	0·4030
35·0	0·2156	0·2090	0·1665	0·3035
40·0	0·1188	0·1082	0·0573	0·2040
45·0	0·0257	0·0085	−0·0514	0·1045

Since the form of the perturbation solution is relatively simple, there appears to be no compelling reason to utilize the more approximate quasi-steady-state result for cases when both N_a and N_b are small. The extended solution given by equation (19) does yield results for all values of N_b and small N_a, a region not covered by the perturbation solution. However, such calculations should be treated with some caution since the asymptotic nature of the quasi-steady-state result is not clearly understood.

Table 4. Comparison of finite-difference and extended quasi-steady-state solutions for $N_a = 0{\cdot}01$ *and* $N_b = 1$

t	R^2	
	Finite-difference	Equation (19)
0·1	0·9941	0·9986
1·0	0·9718	0·9861
5·0	0·8988	0·9307
10·0	0·8173	0·8614
15·0	0·7395	0·7921
20·0	0·6637	0·7227
25·0	0·5895	0·6534
30·0	0·5164	0·5841
35·0	0·4444	0·5148
40·0	0·3731	0·4455
45·0	0·3028	0·3762
50·0	0·2332	0·3069
60·0	0·0967	0·1682
65·0	0·0305	0·0989
67·5	0	0·0643

A third asymptotic solution utilized with success in the treatment of bubble growth phenomena [16] is the thin boundary-layer solution. This solution applies to physical situations where the velocity of the particle surface is large compared to the rate of growth of the thickness of the concentration or temperature boundary layer; it can thus be regarded as the asymptotic perturbation result for large N_a. Although thin boundary-layer solutions have proved very valuable in the analysis of spherical growth phenomena, it is clear that such results would be of limited utility in the analysis of dissolution processes.

REFERENCES

1. G. Birkhoff, R. S. Margulies and W. A. Horning, Spherical bubble growth, *Physics Fluids* **1**, 201–204 (1958).
2. J. S. Kirkaldy, The time-dependent diffusion theory for condensation on spherical and plane surfaces, *Can. J. Phys.* **36**, 446–455 (1958).

3. L. E. Scriven, On the dynamics of phase growth, *Chem. Engng Sci.* **10**, 1–13 (1959).
4. S. G. Bankoff, Heat conduction or diffusion with change of phase, *Advances in Chemical Engineering*, Edited by T. B. Drew *et al.*, Vol. 5. Academic Press, New York (1964).
5. P. S. Epstein and M. S. Plesset, On the stability of gas bubbles in liquid–gas solutions, *J. Chem. Phys.* **18**, 1505–1509 (1950).
6. I. M. Krieger, G. W. Mulholland and C. S. Dickey, Diffusion coefficients for gases in liquids from the rates of solution of small gas bubbles, *J. Phys. Chem.* **71**, 1123–1129 (1967).
7. J. L. Duda and J. S. Vrentas, Mathematical analysis of bubble dissolution, *A.I.Ch.E. Jl* **15**, 351–356 (1969).
8. D. B. Spalding, *Some Fundamentals of Combustion*. Butterworth, London (1955).
9. D. E. Rosner, private communication.
10. M. Cable and D. J. Evans, Spherically symmetrical diffusion-controlled growth or dissolution of a sphere, *J. Appl. Phys.* **38**, 2899–2906 (1967).
11. D. W. Readey and A. R. Cooper, Molecular diffusion with a moving boundary and spherical symmetry, *Chem. Engng Sci.* **21**, 917–922 (1966).
12. G. E. Forsythe and W. R. Wasow, *Finite-Difference Methods for Partial Differential Equations*. John Wiley, New York (1960).
13. T. R. Goodman, Application of integral methods to transient nonlinear heat transfer, *Advances in Heat Transfer*, Edited by T. F. Irvine and J. P. Hartnett, Vol. 1. Academic Press, New York (1964).
14. D. E. Rosner, Lifetime of a highly soluble dense spherical particle, *J. Phys. Chem.* **73**, 382–387 (1969).
15. M. Van Dyke, *Perturbation Methods in Fluids Mechanics*. Academic Press, New York (1964).
16. S. G. Bankoff, Diffusion-controlled bubble growth, *Advances in Chemical Engineering*, Edited by T. B. Drew *et al.*, Vol. 6. Academic Press, New York (1966).

DISSOLUTION CONTRÔLÉE PAR UN TRANSFERT DE MASSE ET DE CHALEUR POUR UNE SPHÈRE ISOLÉE

Résumé—On présente un ensemble de solutions aux différences finies décrivant la dissolution contrôlée par le transfert de chaleur et de masse d'une sphère isolée. L'analyse est basée sur une formulation généralisée qui inclut trois classes spécifiques de problème de dissolution. Une transformation qui immobilise la frontière mouvante et transforme la région infinie intéressante en région finie est utilisée afin de minimiser les difficultés associées à une analyse numérique de ce problème.

Les résultats relatifs à la variation du rayon en fonction du temps et au temps de vie de la particule sont établis pour un ensemble de paramètres qui inclut la majorité des processus de dissolution physiquement importants. De plus, les résultats de cette recherche sont utilisés pour déterminer la précision et le domaine d'application des diverses solutions analytiques approchées.

WÄRME- UND STOFFÜBERGANGSABHÄNGIGE AUFLÖSUNG EINER EINZELNEN KUGEL

Zusammenfassung—Es wird ein umfassender Satz von Lösungen endlicher Differenzengleichungen angegeben, der das durch Wärme- oder Stoffübertragung bestimmte Schmelzen einer einzelnen Kugel beschreibt. Die Analyse beruht auf einer verallgemeinerten Lösung, die 3 besondere Arten von Schmelzproblemen einschliesst. Eine Koordinatentransformation, welche die frei beweglichen Grenzen festlegt und den interessierenden, unendlichen Bereich in einen endlichen abbildet, wird angewandt, um die Schwierigkeiten möglichst gering zu halten, die mit der numerischen Lösung des Problems zusammenhängen. Für Parameterbereiche, welch die meisten physikalisch bedeutsamen Schmelzprozesse einschliessen, werden als Ergebnisse die Abhängigkeit der Radien von der Zeit und die Lebensdauer der Kugeln angegeben. Zusätzlich werden die Ergebnisse dieser Untersuchung dazu benutzt, die Genauigkeit und den Anwendungsbereich verschiedener analytischer Näherungslösungen zu bestimmen.

РАСТВОРЕНИЕ ИЗОЛИРОВАННОЙ СФЕРЫ ПРИ УПРАВЛЯЕМОМ ТЕПЛОИЛИ МАССОПЕРЕНОСЕ

Аннотация—Описывается в доступном виде ряд решений в конечных разностях уравнений для случая растворения изолированных сфер при управляемом тепло- или массопереносе. Даётся анализ на основе учёта трёх типов задач по растворению. В целях упрощения задачи вводится преобразование координат, в которых игнорируется движение границы, и бесконечная область отображается в конечную. Для областей параметров, которые характеризуют большинство физически важных процессов растворения, приводятся значения характеристического времени и времени жизни частиц. Кроме того, результаты этого исследования используются для определения точности и границ применимости различных приближенных аналитических решений.