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# ON AN INTEGRAL FORMULATION FOR MOVING BOUNDARY PROBLEMS*

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**Abstract.** The classical moving boundary problems arising in the freezing or chemical reaction of spheres, cylinders and slabs are considered. An integral method is employed to formally effect the integration of the motion of the moving boundary. This formal integration permits upper and lower bounds to be deduced for the motion and in particular simple upper and lower bounds are established for the time to complete freezing or reaction (that is, when the moving boundary reaches the centre of the sphere or cylinder). In addition an improved second upper bound on the motion is achieved by demonstrating that the dimensionless temperature or concentration is bounded above by the standard pseudo steady state approximation. The use of the integral formulation as an iterative scheme and the generalisations for a time dependent surface condition and a non nlinear diffusivity are also briefly considered.

**1. Introduction.** Moving boundary problems are frequently encountered in science and engineering and are important in many industrial processes such as casting thermoplastics or metal, freezing or thawing of foods and the production of ice. In chemical engineering the chemical reaction of a spherical particle is an important process. In this paper we consider the classical moving boundary problems arising in the freezing or chemical reaction of spheres, cylinders and slabs and we establish essentially two new results. Firstly we show that an integral formulation of the problem leads to a formal integration of the motion of the boundary and secondly we show that the temperature or concentration is bounded above by the pseudo steady state approximation. The first result means that upper and lower bounds can be deduced for the motion of the boundary and the time to complete freezing or reaction, whilst the second result gives rise to an improved upper bound for the motion of the boundary. For an extensive review of related literature we refer the reader to Davis and Hill [1].

Henceforth for convenience we detail results only for the spherical geometry. Corresponding results for cylinders and slabs can be deduced in an entirely analogous manner and the main results can be found in a subsequent section. The classical Stefan problem

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for a sphere with Newtonian cooling on the surface is as follows:

$$\frac{\partial c}{\partial t} = \frac{\partial^2 c}{\partial r^2} + \frac{2}{r}\frac{\partial c}{\partial r}, \qquad R(t) < r < 1, \tag{1.1}$$

$$c(1,t) + \beta\frac{\partial c}{\partial r}(1,t) = 1, \qquad c(R(t),t) = 0, \tag{1.2}$$

$$\frac{\partial c}{\partial r}(R(t),t) = -\alpha R'(t), \qquad R(0) = 1, \tag{1.3}$$

where $\alpha$ and $\beta$ are positive and non-negative constants, respectively, $c(r, t)$ is the dimensionless temperature or concentration and $R(t)$ represents the position of the moving boundary. It is important to note that in both the freezing and reaction problems the temperature or concentration is non-dimensionalised so that $c(r, t)$ satisfies,

$$0 \leqslant c(r,t) \leqslant 1. \tag{1.4}$$

The time to complete freezing, deoted by $t_c$, is defined by $R(t_c) = 0$. Since there is no known exact solution to this problem, various numerical and semi-analytical techniques have been devised.

Numerical solutions for such problems are given by Tao [12] while a number of authors have presented perturbation and boundary layer approaches to the problem (for example, Pedroso and Domoto [3], [4], Riley, Smith and Poots [5], Soward [10] and Stewartson and Waechter [11].) Integral approaches are given, for example by Goodman [2], Savino and Siegel [6], Shih and Chou [7], Shih and Tsay [8], and Theofanous and Lim [13]. Here we utilize essentially the integral formulations employed in [6, 7, 8, 13] which is not always apparent due to differences in notations and in transformations of basic variables. The thrust of this paper is to exploit this method to effect a formal integration of the motion of the boundary and consequently deduce a number of simple bounds. These simple, readily obtainable results should be contrasted with the elaborate calculations involved in many of the above papers.

The pseudo steady state approximation for this problem is formally obtained simply by neglecting the time partial derivative in (1.1). The resulting temperature and motion of the boundary are then given by

$$c_{\text{pss}}(r,t) = \frac{[r - R_{\text{pss}}(t)]}{r[1 + (\beta - 1)R_{\text{pss}}(t)]}, \tag{1.5}$$

$$t = \frac{\alpha}{6}\left[1 + 2\beta - 3R_{\text{pss}}^2 - 2(\beta - 1)R_{\text{pss}}^3\right], \tag{1.6}$$

and this solution can be shown to be asymptotically valid for large $\alpha$ (see Pedroso and Domoto [3]). In what follows we need to distinguish between the pseudo steady state motion of the boundary, (1.6), and the actual motion. We therefore introduce a more convenient notation

$$c_0(r,t) = c_{\text{pss}}(r,t), \qquad R_0(t) = R_{\text{pss}}(t). \tag{1.7}$$

In addition it is convenient to define here the function appearing in (1.6), namely

$$S_3(x) = 1 + 2\beta - 3x^2 - 2(\beta - 1)x^3. \tag{1.8}$$

We observe from (1.6) that the pseudo steady state estimate of the time to complete freezing is given by

$$t_0 = \frac{\alpha}{6}(1 + 2\beta). \tag{1.9}$$

A formal regular perturbation in powers of $\alpha^{-1}$, that is

$$c(r, t) = c_0(r, t) + \frac{C_1(r, t)}{\alpha} + o\left(\frac{1}{\alpha}\right), \tag{1.10}$$

gives rise to $C_1(r, t)$ which is singular as the boundary approaches the centre of the sphere. However the order one corrected approximation to the motion of the boundary is well defined for $R(t)$ tending to zero, and in fact we have

$$t = \frac{\alpha}{6}S_3(R) + \frac{(R-1)^2}{6}\left(\frac{(1+2\beta)+(\beta-1)R}{1+(\beta-1)R}\right) + o(1), \tag{1.11}$$

so that in particular the order one corrected estimate of the time to complete freezing is given by

$$t_1 = \frac{(\alpha + 1)}{6}(1 + 2\beta). \tag{1.12}$$

In the following section we utilize (1.4) to bound the motion of the boundary and show that the time to complete freezing satisfies the simple inequalities

$$\frac{\alpha}{6}(1 + 2\beta) \leqslant t_c \leqslant \frac{(\alpha + 1)}{6}(1 + 2\beta). \tag{1.13}$$

In the same section we also establish

$$c(r, t) \leqslant \frac{[r - R(t)]}{r[1 + (\beta - 1)R(t)]}, \tag{1.14}$$

which leads to an improved upper bound to the motion (which is in fact the first two terms on the right hand side of (1.11)).

In Sec. 3 we state the main results for cylinders and slabs while in Sec. 4 we consider the integral formulation as an iterative procedure. In Secs. 5 and 6 we respectively consider the appropriate generalisations of the results of Sec. 2 for problems with a time dependent surface condition and a non-linear diffusivity. In Sec. 7 some numerical results are presented. Finally in this section we remark that since we assume that a well behaved solution to this problem does in fact exist, we might refer the reader to the recent paperr of Solomon, Alexiades and Wilson [9] for consideration of such matters.

**2. Integral formulation and bounds.** On multiplying (1.1) by $r^2$ and integrating from $R(t)$ to $r$ we obtain, on using $(1.3)_1$,

$$\frac{\partial c}{\partial r}(r, t) = r^{-2}\frac{\partial}{\partial t}\int_{R(t)}^{r} \xi^2[\alpha + c(\xi, t)]\, d\xi, \tag{2.1}$$

and a further integration making use of $(1.2)_2$ gives,

$$c(r, t) = \frac{\partial}{\partial t}\int_{R(t)}^{r} \xi\left(1 - \frac{\xi}{r}\right)[\alpha + c(\xi, t)]\, d\xi, \tag{2.2}$$

and a change in the order of integration is justified by observing that the appropriate integrand is bounded on the region under consideration. From these two equations and $(1.2)_1$, we find

$$1 = \frac{\partial}{\partial t}\int_{R(t)}^{1} \xi[1 + (\beta - 1)\xi][\alpha + c(\xi, t)]\, d\xi, \tag{2.3}$$

which evidently integrates to give

$$t = \int_{R(t)}^{1} \xi[1 + (\beta - 1)\xi][\alpha + c(\xi, t)]\, d\xi, \tag{2.4}$$

or alternatively in terms of $S_3$ defined by (1.8) we have

$$t = \frac{\alpha}{6}S_3(R) + \int_{R(t)}^{1} \xi[1 + (\beta - 1)\xi]c(\xi, t)\, d\xi. \tag{2.5}$$

Since $c(r, t)$ is unknown (2.5) represents only a formal integration of the motion of the boundary. However from (1.4) and (2.5) it is apparent that the boundary motion satisfies the simple inequalities

$$\frac{\alpha}{6}S_3(R) \leqslant t \leqslant \frac{(\alpha + 1)}{6}S_3(R). \tag{2.6}$$

From (2.6) we obtain immediately the inequalities (1.13) for the time to complete freezing. These inequalities, although simple are clearly practically very useful, especially for large $\alpha$. It is apparent from (2.6) that the pseudo steady state boundary moves faster than the actual boundary, that is $R_0(t) \leqslant R(t)$. In the remainder of this section we establish (1.14) which leads to an improved upper bound for (2.6) but (1.13) unfortunately remains unchanged.

We use the convention that when $R(t)$ is employed as the independent variable instead of $t$ we write

$$c(r, t) \equiv c^*(r, R). \tag{2.7}$$

From (2.2) and (2.3) we find on eliminating $R'(t)$

$$c^*(r, R) = \frac{\left[\alpha R(r - R) - \int_R^r \xi(r - \xi)(\partial c^*/\partial R)(\xi, R)\, d\xi\right]}{r\left(\alpha R[1 + (\beta - 1)R] - \int_R^1 \xi[1 + (\beta - 1)\xi](\partial c^*/\partial R)(\xi, R)\, d\xi\right)}. \tag{2.8}$$

We note that the validity of the pseudo steady state approximation for large $\alpha$ can be at least formally established from (2.8) by observing that the first terms in both the numerator and denominator will dominate if $\alpha$ is large. In order to establish (1.14) we have from (2.8)

$$c^*(r, R) - \frac{(r - R)}{r[1 + (\beta - 1)R]} = \frac{\left([1 + (\beta - 1)r]\int_R^r \xi(\xi - R)(\partial c^*/\partial R)(\xi, R)\, d\xi + (r - R)\int_r^1 \xi[1 + (\beta - 1)\xi](\partial c^*/\partial R)(\xi, R)\, d\xi\right)}{r[1 + (\beta - 1)R]\left(\alpha R[1 + (\beta - 1)R] - \int_R^1 \xi[1 + (\beta - 1)\xi](\partial c^*/\partial R)(\xi, R)\, d\xi\right)}, \tag{2.9}$$

and for $\beta \geqslant 0$ this expression is clearly negative provided that $\partial c^*/\partial R \leqslant 0$. This is certainly the case since

$$\frac{\partial c^*}{\partial R}(r, R) = \frac{1}{R'(t)} \frac{\partial c}{\partial t}(r, t), \tag{2.10}$$

and from physical considerations clearly $R'(t) \leqslant 0$ and $\partial c/\partial t \geqslant 0$. Thus (1.14) follows.

From (1.14) and (2.5) we obtain in place of (2.6) the following inequality,

$$\frac{\alpha}{6} S_3(R) \leqslant t \leqslant \frac{\alpha}{6} S_3(R) + \frac{(R-1)^2}{6}\left(\frac{(1+2\beta)+(\beta-1)R}{1+(\beta-1)R}\right), \tag{2.11}$$

which is an improved upper bound for the motion of the boundary but does not change (1.13). Again we remark that the upper bound in (2.11) is precisely the order one motion as given by (1.11). In the following section we state without derivation the main formulae for cylinders and slabs.

**3. Cylinders and slabs.** For the cylinder instead of (1.1) we have

$$\frac{\partial c}{\partial t} = \frac{\partial^2 c}{\partial r^2} + \frac{1}{r}\frac{\partial c}{\partial r}, \qquad R(t) < r < 1, \tag{3.1}$$

while (1.2) and (1.3) remain unchanged. In terms of $S_2$ defined by

$$S_2(x) = 2x^2 \log x + (1+2\beta)(1-x^2), \tag{3.2}$$

the pseudo steady state approximation is given by

$$c_0(r, t) = \frac{[\log r - \log R_0(t)]}{[\beta - \log R_0(t)]}, \qquad t = \frac{\alpha}{4} S_2(R_0). \tag{3.3}$$

Instead of (2.2) and (2.4) we have

$$c(r, t) = \frac{\partial}{\partial t}\int_{R(t)}^{r} \xi(\log r - \log \xi)[\alpha + c(\xi, t)]\, d\xi, \tag{3.4}$$

$$t = \int_{R(t)}^{1} \xi(\beta - \log \xi)[\alpha + c(\xi, t)]\, d\xi, \tag{3.5}$$

and from (1.4) and (3.5) we obtain the inequalities

$$\frac{\alpha}{4} S_2(R) \leqslant t \leqslant \frac{(\alpha+1)}{4} S_2(R), \tag{3.6}$$

$$\frac{\alpha}{4}(1+2\beta) \leqslant t_c \leqslant \frac{(\alpha+1)}{4}(1+2\beta). \tag{3.7}$$

Exactly as in the previous section we can establish that

$$c(r, t) \leqslant (\log r - \log R)/(\beta - \log R), \tag{3.8}$$

and from (3.5) and (3.8) we obtain

$$\frac{\alpha}{4} S_2(R) \leqslant t \leqslant \frac{\alpha}{4} S_2(R) + \frac{1}{4}\left((1+2\beta+R^2) - \frac{[2\beta(1+\beta)+1-R^2]}{(\beta - \log R)}\right), \tag{3.9}$$

which improves the upper bound in (3.6) but not in (3.7).

For the slab we can use the same Eqs. (1.2) and (1.3) while instead of (1.1) we have

$$\frac{\partial c}{\partial t} = \frac{\partial^2 c}{\partial r^2}, \qquad R(t) < r < 1, \tag{3.10}$$

and we interpret the time $t_c$ to complete freezing as the time taken for the moving boundary to reach the origin. With $S_1$ defined by

$$S_1(x) = 1 + 2\beta - 2(1+\beta)x + x^2, \tag{3.11}$$

we obtain the following formulae:

$$c_0(r,t) = \frac{[r - R_0(t)]}{[1 + \beta - R_0(t)]}, \qquad t = \frac{\alpha}{2} S_1(R_0), \tag{3.12}$$

$$c(r,t) = \frac{\partial}{\partial t} \int_{R(t)}^{r} (r - \xi)[\alpha + c(\xi, t)]\, d\xi, \tag{3.13}$$

$$t = \int_{R(t)}^{1} (1 + \beta - \xi)[\alpha + c(\xi, t)]\, d\xi. \tag{3.14}$$

Equations (1.4) and (3.14) give

$$\frac{\alpha}{2} S_1(R) \leqslant t \leqslant \frac{(\alpha+1)}{2} S_1(R), \tag{3.15}$$

$$\frac{\alpha}{2}(1 + 2\beta) \leqslant t_c \leqslant \frac{(\alpha+1)}{2}(1 + 2\beta), \tag{3.16}$$

while the inequality

$$c(r,t) \leqslant (r - R)/(1 + \beta - R), \tag{3.17}$$

and (3.14) gives

$$\frac{\alpha}{2} S_1(R) \leqslant t \leqslant \frac{\alpha}{2} S_1(R) + \frac{(R-1)^2}{6}\left(\frac{1 + 3\beta - R}{1 + \beta - R}\right). \tag{3.18}$$

Unlike the sphere and cylinder we obtain an improved upper bound for $t_c$, namely

$$\frac{\alpha}{2}(1 + 2\beta) \leqslant t_c \leqslant \frac{\alpha}{2}(1 + 2\beta) + \frac{1}{6}\frac{(1 + 3\beta)}{(1 + \beta)}. \tag{3.19}$$

We note that in the case $\beta$ zero the problem for the slab admits the well known exact solution

$$c(r,t) = 1 - \operatorname{erf}\left(\frac{1 - r}{2\sqrt{t}}\right) \Big/ \operatorname{erf}\left(\frac{\gamma}{2}\right)^{1/2}, \tag{3.20}$$

$$R(t) = 1 - (2\gamma t)^{1/2}, \tag{3.21}$$

where $\gamma$ denotes the positive real root of

$$\alpha e^{\gamma/2}\left(\frac{\pi\gamma}{2}\right)^{1/2} \operatorname{erf}\left(\frac{\gamma}{2}\right)^{1/2} = 1, \tag{3.22}$$

and it can be shown that these results satisfy the above equations (3.13)–(3.19) with $\beta = 0$. This exact solution is compared graphically with the pseudo steady state solution in the final section of the paper. We also observe that for $\beta = 0$, (3.18) and (3.19) together with the exact result (3.21) both reduce to the same inequalities, namely

$$\alpha \leqslant \gamma^{-1} \leqslant \alpha + \tfrac{1}{3}. \tag{3.23}$$

Numerical values given in Table 1 indicate the validity of these inequalities.

| $\alpha$ | $\gamma^{-1}$ | $\alpha + \frac{1}{3}$ |
|---|---|---|
| 0.20 | 0.4453 | 0.5333 |
| 0.25 | 0.5043 | 0.5833 |
| 0.50 | 0.7801 | 0.8333 |
| 1.00 | 1.3005 | 1.3333 |
| 2.00 | 2.3145 | 2.3333 |
| 5.00 | 5.3251 | 5.3333 |
| 10.00 | 10.3291 | 10.3333 |
| 50.00 | 50.3325 | 50.3333 |
| 100.00 | 100.3329 | 100.3333 |
| 500.00 | 500.3332 | 500.3333 |

TABLE 1. Values of $\gamma^{-1}$ and $\alpha + \frac{1}{3}$ for various values of $\alpha$.

**4. Integral formulation as an iterative scheme.** The procedure of [6, 7, 8, 13] is to utilize (2.8) as an iterative scheme taking the pseudo steady state result as the initial approximation. Although we make no use of this procedure, we make one or two observations about this method for the sake of completeness. From (2.5) and (2.8) we see that we may generate successive approximations for $t(R)$ and $c^*(r, R)$ by

$$t_{n+1}(R) = \frac{\alpha}{6} S_3(R) + \int_R^1 \xi[1 + (\beta - 1)\xi] c_n^*(\xi, R)\, d\xi, \tag{4.1}$$

$$c_{n+1}^*(r, R) = \frac{\left(\alpha R(r - R) - (\partial/\partial R)\int_R^r \xi(r - \xi) c_n^*(\xi, R)\, d\xi\right)}{r\left(\alpha R[1 + (\beta - 1)R] - (\partial/\partial R)\int_R^1 \xi[1 + (\beta - 1)\xi] c_n^*(\xi, R)\, d\xi\right)}. \tag{4.2}$$

Due to the unusual nature of the integral Eq. (2.8) the problem of convergence of the scheme appears to be a non-trivial matter. We merely make the following comments.

Firstly we observe that the pseudo steady state approximation (1.5) and (1.6) arises out of (4.2) and (4.1) respectively by taking $c_{-1}(r, t) \equiv 0$ as the initial approximation. Thus the pseudo steady state solution may be considered as a first iteration of the scheme and not simply an arbitrary initial approximation as is commonly supposed. For the second iteration we find

$$t_1(R) = \frac{\alpha}{6} S_3(R) + \frac{(R - 1)^2}{6}\left(\frac{(1 + 2\beta) + (\beta - 1)R}{1 + (\beta - 1)R}\right), \tag{4.3}$$

$$c_1^*(r, R) = \frac{(r - R)\left(6\alpha R[1 + (\beta - 1)R]^2 + (r - R)[3 + (\beta - 1)(r + 2R)]\right)}{2r\left(3\alpha R[1 + (\beta - 1)R]^3 + 3\beta(1 - R)[1 + (\beta - 1)R] + (\beta - 1)^2(1 - R)^3\right)}, \tag{4.4}$$

and in principle it is possible to proceed further. However the calculations become rather tedious and the results are of doubtful value. For example, for $\beta = 0$ the scheme produces results which have undesirable properties when $\alpha > 1$. Thus

$$t_2(R) = \frac{(R-1)^2}{2}\left(\frac{\alpha}{3}(1+2R) + \frac{1}{5}\left(\frac{1+5\alpha R}{1+3\alpha R}\right)\right), \tag{4.5}$$

is not uniquely invertable for $\alpha > 1$. Moreover for $\beta = 0$ and $\alpha > 1$ we find that $c_2^*(r, R)$ has a denominator which vanishes for some $R$ in (0, 1) so that both $c_2^*(r, R)$ and $t_3(R)$ are unbounded. Clearly a detailed examination of the convergence of these schemes is necessary.

**5. Time dependent surface condition.** In this section we consider the problem (1.1), (1.2) and (1.3) with $(1.2)_1$ replaced by

$$c(1, t) + \beta \partial c(1, t)/\partial r = f(t), \tag{5.1}$$

where $f(t)$ is assumed to be a positive monotonically increasing function of time. We emphasize that generally the results of this section will not be applicable if $f(t)$ is not positive nor monotonically increasing. We employ the notation

$$g(t) = \int_0^t f(\tau)\, d\tau. \tag{5.2}$$

The pseudo steady state solution is given by

$$c_0(r, t) = \frac{f(t)[r - R_0(t)]}{r[1 + (\beta - 1)R_0(t)]}, \qquad g(t) = \frac{\alpha}{6} S_3(R_0), \tag{5.3}$$

where $S_3(x)$ is defined by (1.8). For the integral formulation (2.1) and (2.2) are still valid but in place of (2.4) we have

$$g(t) = \int_{R(t)}^1 \xi[1 + (\beta - 1)\xi][\alpha + c(\xi, t)]\, d\xi. \tag{5.4}$$

From the equation corresponding to (2.8) and recalling that $\beta \geqslant 0$ we can show that instead of (1.4) and (1.14) we have together the inequalities

$$0 \leqslant c(r, t) \leqslant \frac{f(t)[r - R(t)]}{r[1 + (\beta - 1)R(t)]} \leqslant f(t). \tag{5.5}$$

From this and (5.4) we have, in the usual way, the following inequalities for the motion of the boundary, which are in fact generalisations of (2.6) and (2.11) respectively,

$$\frac{\alpha}{6} S_3(R) \leqslant g(t) \leqslant \frac{[\alpha + f(t)]}{6} S_3(R), \tag{5.6}$$

$$\frac{\alpha}{6} S_3(R) \leqslant g(t) \leqslant \frac{\alpha}{6} S_3(R) + f(t)\frac{(R-1)^2}{6}\left(\frac{(1+2\beta) + (\beta - 1)R}{1 + (\beta - 1)R}\right), \tag{5.7}$$

both of which yield

$$\frac{\alpha}{6}(1 + 2\beta) \leqslant g(t_c) \leqslant \frac{[\alpha + f(t_c)]}{6}(1 + 2\beta), \tag{5.8}$$

for the time to complete freezing $t_c$. For example if we take $f(t) = t$, then (5.8) gives

$$\left[\frac{\alpha}{3}(1+2\beta)\right]^{1/2} \leqslant t_c \leqslant \left[\frac{1}{3}(1+2\beta)\right]^{1/2}\left[\alpha + \frac{1}{12}(1+2\beta)\right]^{1/2} + \frac{(1+2\beta)}{6}. \quad (5.9)$$

**6. Non-linear diffusivity with no radiation.** In this section we consider the problem

$$\frac{\partial c}{\partial t} = r^{-2}\frac{\partial}{\partial r}\left(r^2 D(c)\frac{\partial c}{\partial r}\right), \qquad R(t) < r < 1, \quad (6.1)$$

$$c(1,t) = 1, \qquad c(R(t),t) = 0, \quad (6.2)$$

$$D(0)\frac{\partial c}{\partial r}(R(t),t) = -\alpha R'(t), \qquad R(0) = 1, \quad (6.3)$$

where $D(c)$ is a positive monotonically increasing function of $c$ such that $D(0)$ is non-zero. We introduce $F(c)$ such that

$$F(c) = \int_0^c D(\rho)\,d\rho. \quad (6.4)$$

The pseudo steady state approximation is given by

$$F(c_0(r,t)) = \frac{F(1)[r - R_0(t)]}{r[1 - R_0(t)]}, \qquad F(1)t = \frac{\alpha}{6}(R_0 - 1)^2(1 + 2R_0). \quad (6.5)$$

We find that in place of (2.2) and (2.5) we have

$$F(c(r,t)) = \frac{\partial}{\partial t}\int_{R(t)}^{r} \xi\left(1 - \frac{\xi}{r}\right)[\alpha + c(\xi,t)]\,d\xi, \quad (6.6)$$

$$F(1)t = \int_{R(t)}^{1} \xi(1-\xi)[\alpha + c(\xi,t)]\,d\xi. \quad (6.7)$$

The integral equation for $c^*(r,R) \equiv c(r,t)$ becomes

$$F(c^*(r,R)) = \frac{F(1)\left(\alpha R(r-R) - \int_R^r \xi(r-\xi)(\partial c^*/\partial R)(\xi,R)\,d\xi\right)}{r\left(\alpha R(1-R) - \int_R^1 \xi(1-\xi)(\partial c^*/\partial R)(\xi,R)\,d\xi\right)}, \quad (6.8)$$

and again assuming that $R'(t) \leqslant 0$ and that $\partial c/\partial t \geqslant 0$ we can establish from (6.8)

$$F(c(r,t)) \leqslant F(1)\frac{[r - R(t)]}{r[1 - R(t)]}. \quad (6.9)$$

Thus we have

$$0 \leqslant F(c^*(r,R)) \leqslant F(c_0^*(r,R)) \leqslant F(1), \quad (6.10)$$

and therefore provided that $D(c)$ is monotonically increasing we obtain

$$0 \leqslant c(r,t) \leqslant F^{-1}\left(\frac{F(1)[r - R(t)]}{r[1 - R(t)]}\right) \leqslant 1. \quad (6.11)$$

From (6.7) and (6.11) the inequalities corresponding to (2.6) and (2.11) are

$$\frac{\alpha}{6}(R-1)^2(1+2R) \leqslant F(1)t \leqslant \frac{(\alpha+1)}{6}(R-1)^2(1+2R), \quad (6.12)$$

$$\frac{\alpha}{6}(R-1)^2(1+2R) \leqslant F(1)t$$
$$\leqslant \frac{\alpha}{6}(R-1)^2(1+2R) + \int_R^1 \xi(1-\xi)F^{-1}\left(\frac{F(1)(\xi-R)}{\xi(1-R)}\right) d\xi, \tag{6.13}$$

and for $t_c$ both (6.12) and (6.13) yield simply

$$\frac{\alpha}{6} \leqslant F(1)t_c \leqslant \frac{(\alpha+1)}{6}. \tag{6.14}$$

Finally in this section we note that it is apparent from (6.6) that the case $\beta$ non-zero does not permit a similar analysis.

**7. Numerical results.** For the slab, with $\beta = 0$, Fig. 1 shows the variation of the exact solution $c^*(r, R)$, as derived from (3.20) and (3.21) and the pseudo steady state solution $c_0^*(r, R)$, derived from (3.12), at equal time intervals between $(6\gamma)^{-1}$ and $(2\gamma)^{-1}$. In Fig. 2 the variation, for the sphere, of $c_1^*(r, R)$, as given by (4.4) and $c_0^*(r, R)$, obtained from (1.5) is illustrated for $\alpha = 2$ and $\beta = .5$, at equally spaced positions of the moving boundary $R$. We observe that $c_1^*(r, R) \leqslant c_0^*(r, R)$. In Figures 3 and 4 we show, for the sphere and cylinder respectively the upper and lower bounds for the motion of the moving

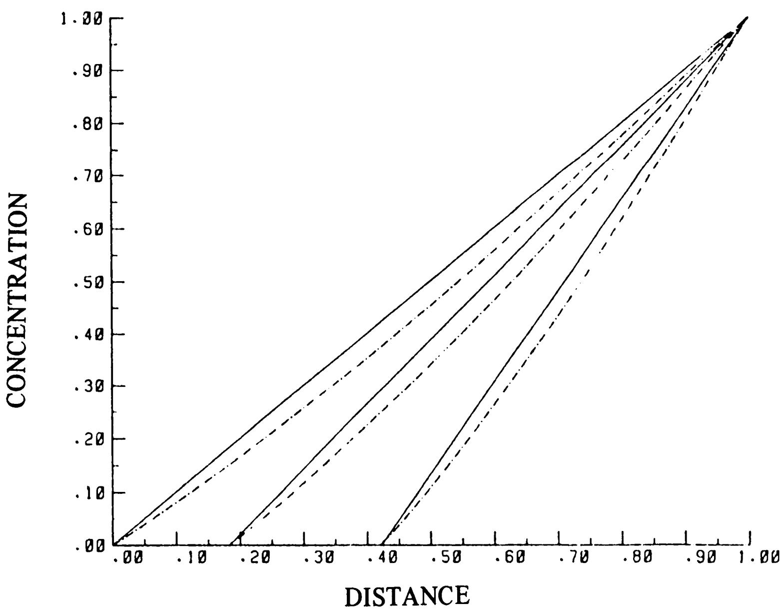


FIG. 1. Concentration verses distance for the slab with $\alpha = 1$ and $\beta = 0$ for exact $c^*(r, R)(-\cdot-)$ and $c_0^*(r, R)(—)$.

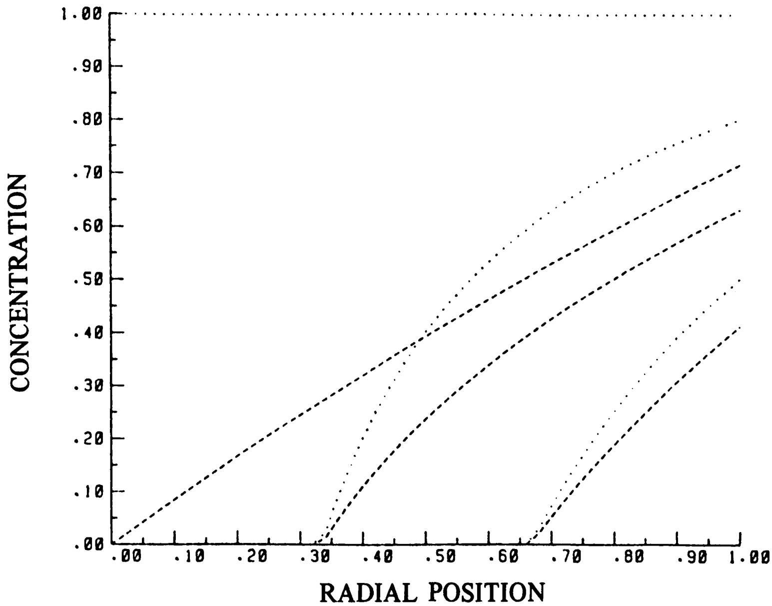


FIG. 2. Concentration verses distance for the sphere with $\alpha = 2$ and $\beta = 0.5$ for $c_0^*(r, R)(\cdots\cdots)$ and $c_1^*(r, R)(---)$.

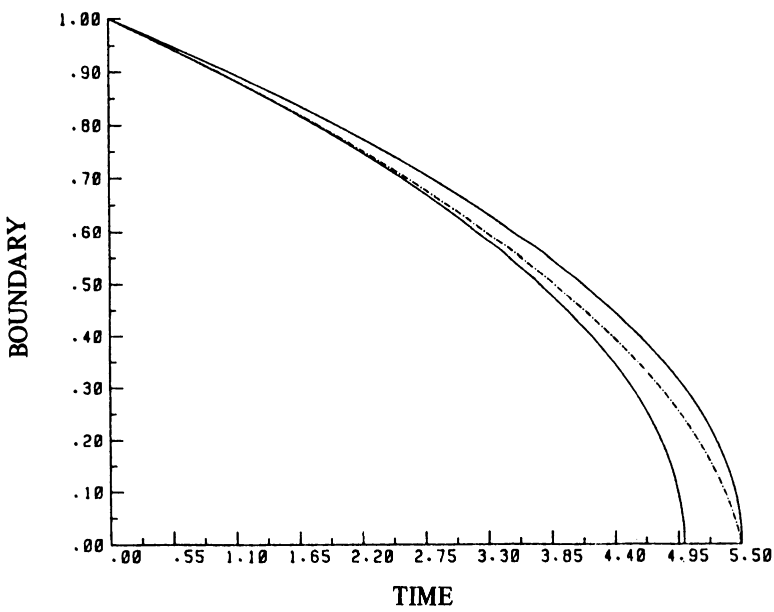


FIG. 3. Upper and lower bounds for the motion of the moving boundary for the sphere with $\alpha = 10$ and $\beta = 1$ (improved upper bound $-\cdot-\cdot-$).

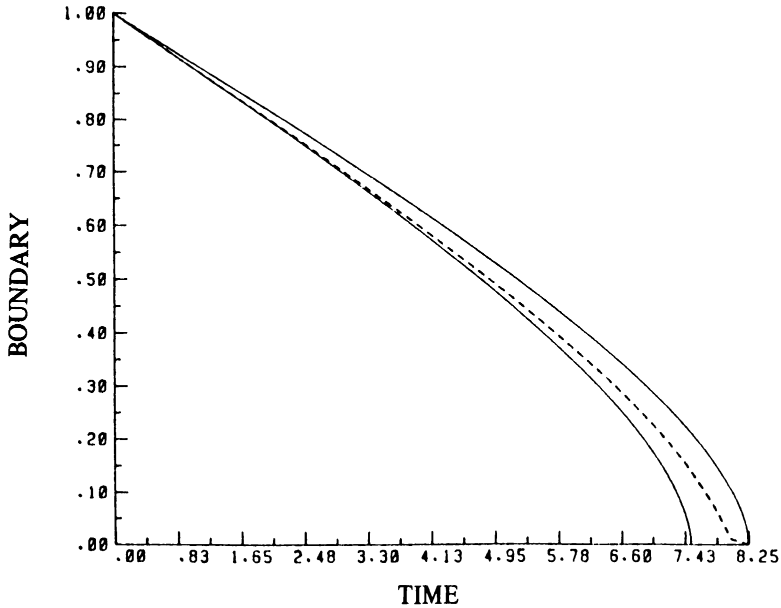


FIG. 4. Upper and lower bounds for the motion of the moving boundary for the cylinder with $\alpha = 10$ and $\beta = 1$ (improved upper bound $---$).

boundary, as determined from the inequalities (2.10), (2.11) and (3.6), (3.9), for $\alpha = 10$ and $\beta = 1$. It should be noted that as $\beta$ becomes larger, the improved upper bounds, given by (2.11) and (3.9), tend toward the lower bounds, except for when the boundary is very near the centre.

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