$\underset{\sim}{Y}$ = V-phase composition matrix: y_{ij} is the mole fraction of component j in the V-phase in stage i

β = rj element of $(\underset{\sim}{B}^{-1})\underset{\sim}{A}$: j is the number of the L-phase variable being fixed, and r is the number of the V-phase variable which becomes a noniteration variable

$\underset{\sim}{\Gamma}$ = activity coefficient matrix for V-phase: γ_{ij} = activity coefficient for component j in V-phase in stage i

δ_{kl} = Dirac delta function; $\delta_{kl} = 1$ if $k = l$, otherwise it is zero

ν = iteration number

ξ_{jk} = interaction energy between the j-k pair of components

ρ_{ijk} = molar volume of component j divided by molar volume of component k in stage i

σ_1, σ_2 = Euclidean error norms, defined by Eqs. 15 and 16

$\underset{\sim}{\Lambda}$ = activity coefficient matrix for L-phase: λ_{ij} = activity coefficient for component j in L-phase in stage i

ω_{ijk} = modified Wilson parameter between the j-k pair of components in stage i

Superscripts

* = a modified vector or matrix obtained by eliminating one or more rows and/or columns

T = matrix transpose

APPENDIX

The modified Wilson Equation (Tsuboka and Katayama, 1975) was used for calculation of activity coefficients for the liquid phase. The derivatives of the activity coefficient with respect to composition and temperature are needed in the calculation of the Jacobian matrix. The partial derivatives for stage i are as follows.

$$\frac{d\lambda_{ij}}{dt_i} = -\left(\frac{\lambda_{ij}}{rt_i^2}\right)\left(\sum_k^m (x_{ik}\omega_{ikj}\xi_{kj}) \Big/ \sum_k^m (x_{ik}\omega_{ikj}) + \sum_k^m \left(x_{ik}\omega_{ijk}\sum_l^m x_{il}\omega_{ilk}(\xi_{jk}-\xi_{lk})\Big/ \left(\sum_l^m (x_{il}\omega_{ilk})\right)^2\right)\right) - \sum_k^m \left(x_{ik}(d\rho_{ikj}/dt_i\right)\Big/\sum_k^m (x_{ik}\rho_{ikj}) - \sum_k^m \left((x_{ik}(d\rho_{ijk}/dt_i)\left(\sum_l^m (x_{il}\rho_{ilk})\right) - x_{ik}\rho_{ijk}\sum_l^m (x_{il}(d\rho_{ilk}/dt_i))\right)\Big/\left(\left(\sum_l^m (x_{il}\rho_{ilk})\right)^2\right)\right) \quad \text{(A-1)}$$

$$d\lambda_{ij}/dx_{ik} = -\lambda_{ij}\left(\omega_{ikj}\Big/\sum_l^m (x_{il}\omega_{ilj}) + \sum_l^m\left(\left(\omega_{ijk}\delta_{lk}\left(\sum_p^m (x_{ip}\omega_{ipl})\right) - x_{il}\omega_{ijl}\omega_{ikl}\right)\Big/\left(\sum_p^m (x_{ip}\omega_{ipl})\right)^2\right) - \rho_{ikj}\Big/\sum_p^m (x_{ip}\rho_{ipj}) - \sum_l^m\left(\left(\omega_{ijk}\delta_{lk}\left(\sum_p^m (x_{ip}\rho_{ipl})\right) - x_{il}\omega_{ipl}\omega_{ikl}\right)\Big/\left(\sum_p^m (x_{ip}\rho_{ipl})\right)^2\right)\right) \quad \text{(A-2)}$$

where

ω_{ijk} = the modified Wilson parameter between the j-k pair of component in stage i and defined by:

$$\omega_{ijk} = \rho_{ijk}\exp(-\xi_{jk}/rt_i)$$

ρ_{ijk} = molar volume of component j divided by molar volume of component k in stage i

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Asymptotic Expansions for the Description of Gas Bubble Dissolution and Growth

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A new asymptotic expansion is presented to describe the dissolution or growth of an isolated, stationary gas bubble in a liquid. With the exception of long times, bubble radii calculated via this expansion are in excellent agreement with values obtained by numerical methods over a wide range of gas under(super)-saturations.

SCOPE

Even the simplest problem of mass transport between a gas bubble and the surrounding liquid, namely that of the dissolution (growth) of an isolated, stationary gas bubble in an isothermal, isobaric, liquid of infinite extent is complex. Although the governing equations are well known, exact solutions cannot be obtained in general because of the motion of the

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gas-liquid interface, and the resulting convective transport in the fluid. An exception is the special case of growth from zero initial size treated by Scriven (1959).

A number of approximate methods have been developed to deal with this problem. Some of these techniques permit simple analytical solutions to be obtained but do not yield accurate results. Other procedures, while more precise, involve some degree of numerical computation.

Thus, the objective of this study is to find a simple yet accurate procedure for the calculation of gas bubble radii during dissolution or growth for the system described above which also is generalizable to more complex bubble dissolution/growth problems.

CONCLUSIONS AND SIGNIFICANCE

A "short time" asymptotic expansion was found to describe gas bubble dissolution and growth. It has the virtue of being both simple and quite accurate. Use was made of the recognition that in the limit of zero time a similarity solution is possible for this problem. This solution was extended in time by an asymptotic expansion. The final result for the bubble radius given in Eq. 14 is seen to be compact, and convenient for evaluation by hand.

The importance of this procedure is recognized when one finds that this "small" time procedure appears to predict accurate values of bubble radii for almost the entire period of dissolution and over similarly long periods of growth. Furthermore, this procedure is applicable for a broad range of gas under(super)-saturations. Also, it appears that this method may be utilized to attack more complex gas bubble problems, such as those involving chemically reactive and/or multicomponent systems.

INTRODUCTION

Many articles have been written on the mathematical description of the problem of dissolution and growth of stationary bubbles in view of the practical applications of this subject. For instance, Pfeiffer and Krieger (1974) studied the dissolution of gas bubbles held stationary by attachment to a fiber. By fitting the observed radius-time data to theoretical predictions, they were able to estimate the diffusivities of various gaseous solutes in liquids. Another application for theoretical treatments of stationary bubble dissolution is the elimination of gas bubbles from glassmelts which are highly viscous. Bubble rise in such systems is negligibly slow. Our interest in this problem has been stimulated by potential applications to "Space Processing". In orbiting spacecraft, near free fall conditions prevail, and the dissolution and growth of stationary fluid droplets, of which bubbles form a limiting case, will be quite important.

One of the simpler systems which has been oft considered is that of a dissolving (growing) stationary, isolated spherical gas bubble in a liquid of large extent. However, even this problem is sufficiently complex (when proper accounting is made for the motion of the liquid caused by the movement of the interface) that, in general, approximations are required to obtain analytical solutions. A notable exception is the classical work of Scriven (1959) who developed an exact solution for growth from zero initial size using a similarity transformation. The problem of dissolution or growth from a non-zero initial size has been treated by many workers. Readey and Cooper (1966) derived the model equations, and solved them for certain ranges of the governing parameters using a numerical technique. A numerical scheme also was used by Cable and Evans (1967). Duda and Vrentas (1969, 1971) identified two smallness parameters N_a and N_b in the problem, and developed a perturbation expansion in these parameters. Duda and Vrentas obtained the concentration field to $0(N_a)$ (and $0(N_b)$ for fixed N_a) and the square of the bubble radius to $0(N_a^2)$. They also developed an accurate finite difference solution of this problem and demonstrated good agreement between their $0(N_a^2)$ result and the numerical solution for $|N_a| = 0.2$.

Rosner and Epstein (1972) investigated the effects of interface kinetics and surface tension in bubble growth using an integral method and the assumption of a thin concentration boundary layer to obtain closed form solutions. Viscous, inertial, surface tension, and interface kinetics effects were all included in a treatment by Szekely and Fang (1973) who used the method of finite differences for solving the governing equations. Ruckenstein and Davis (1970) analyzed the general problem of moving and stationary fluid spheres dissolving or growing in a surrounding fluid medium. They used the assumption of a thin concentration boundary layer, and employed a similarity transformation developed by Ruckenstein (1968) in obtaining an integro-differential equation for the radius. This equation was, in the general case, solved using numerical means.

Numerous approximate solutions of the problem of bubble dissolution and growth have appeared in the literature. Epstein and Plesset (1950) presented the quasi-stationary approximation wherein the mass flux at the bubble surface is evaluated from a model which neglects both convection in the liquid and interface motion. Analytical solutions of the quasi-stationary equations have been obtained by Frischat and Oel (1965). The utility of this approximation has been examined in recent articles by Weinberg et al. (1980) and Subramanian and Chi (1980). In another work (Subramanian and Weinberg, 1980), we have pointed out that accounting for the motion of the bubble boundary, while ignoring the resulting convective transport in the liquid (Tao, 1979, for example) results in a poorer approximation than the quasistationary result. The time derivative in the quasistationary equation is sometimes neglected to obtain the quasisteady model (Bankoff, 1966).

Most of the studies quoted above have either relied upon numerical solutions of partial differential equations or have utilized approximate techniques often leading to cumbersome expressions for the bubble radius. Here, we present a simple, yet powerful, method for the calculation of the radius of a shrinking (or growing) bubble. The procedure involves the use of an asymptotic expansion technique which provides compact analytical solutions for the time dependence of the bubble radius. For a simple illustration, we have chosen the case of the dissolution of an isolated, stationary single-component gas bubble in a liquid of infinite extent. However, the technique can be generalized in a straightforward fashion to handle more complex situations involving multicomponent and/or chemically reactive systems. As a matter of interest, it will be seen that the classical quasistationary concentration field is recovered as the zeroth order result in one version of the present method. It is shown that this concentration field may be improved in a systematic fashion.

A comparison of the present expansion with the parameter perturbation scheme employed by Duda and Vrentas (1969) is appropriate, and will be presented. Our scheme employs an orderly expansion in time instead of the parameters N_a and N_b used by the latter authors (while the present illustration deals with $N_b = 0$ for simplicity, the method can be employed when $N_b \neq 0$). The present expansion for the bubble radius does not contain an integral requiring numerical quadrature for evaluation such as the one present in the Duda-Vrentas result. Our scheme is applicable over a broad range of N_a values, and in fact, a truncated version of our expansion almost identically reproduces the Duda-Vrentas result where the latter is useful.

For large N_a where the Duda and Vrentas expansion diverges, as expected, the present expansion for the radius continues to be quite accurate. It is worth noting that the small time expansion developed here provides reasonably accurate predictions for the radius even at large times. However, in light of the asymptotic nature of the present expansion (limit $T \to 0$), it cannot be expected to represent the exact solution in the limit $T \to \infty$. This is particularly important in growth problems since all values of time, including "large" values, are meaningful for growth.

ANALYSIS

We consider the unsteady isothermal dissolution (or growth) of a bubble whose center of mass is stationary in a liquid of large extent. The bubble is assumed to be a perfect sphere. Consequently, the concentration field in the liquid is spherically symmetric, and the velocity field purely radial. Inertial, viscous, and surface tension effects are ignored. Therefore, the pressure inside the bubble is constant and equal to the pressure in the liquid. A pure component bubble is considered, and it is assumed that equilibrium conditions are maintained at the interface at all times so that the concentration of the gaseous species in the liquid at the interface is a constant. The partial specific volume of the gaseous species in the liquid is considered negligible.

Under the above assumptions, the scaled concentration field $C(T, r)$ will satisfy the following conservation equation and initial and boundary conditions.

$$\frac{\partial C}{\partial T} + \frac{dg}{dT}\frac{g^2}{r^2}\frac{\partial C}{\partial r} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial C}{\partial r}\right) \tag{1}$$

$$C(0, r) = 0 \tag{2a}$$

$$C(T, \infty) = 0 \tag{2b}$$

$$C(T, g(T)) = 1 \tag{2c}$$

Here, the reference length used in defining the scaled radial coordinate r and the scaled bubble radius g is the initial bubble radius a_o. The scaled time T is defined using the characteristic diffusion time a_o^2/D. Mass balance leads to:

$$\frac{dg}{dT} = N_a\frac{\partial C}{\partial r}(T, g(T)) \tag{3}$$

and the initial condition on $g(T)$ is:

$$g(0) = 1 \tag{4}$$

The driving force parameter N_a is positive for dissolution and negative for growth.

The problem statement is complete. Even though the scaled bubble radius $g(T)$ is the quantity of principal interest, due to the coupling between $C(T, r)$ and $g(T)$, both must be obtained simultaneously. The procedure used here for the solution of the coupled equations is based upon the fact that in the limit as $T \to 0$, a similarity solution may be found for the concentration field. We develop an asymptotic expansion in time which permits us to improve upon this lowest order similarity solution. Interestingly, this expansion, which is expected to describe the solution for "small" T, appears to be quite accurate in predicting the radius over almost the entire period of dissolution, and over similarly long periods in growth.

Solution

There are several possible routes one may use to obtain the solution for $g(T)$ via the asymptotic expansion technique. All such procedures, however, rely upon the immobilization of the interface using a suitable transformation. One way of achieving this is to define

$$Y = r - g(T) \tag{5}$$

At this point, it is convenient to introduce a transformed concentration field.

$$C_1(T, Y) = \frac{r}{g(T)}C(T, r) \tag{6}$$

As will be seen shortly, Eq. 6 has a special significance since it will permit us to recover the well-known quasi-stationary solution as the zeroth order concentration field in the expansion. Its function is to generate explicitly the steady curvature contribution to dg/dT in the radius equation. The use of Eqs. 5 and 6 in Eqs. 1 to 3 leads to the following set.

$$\frac{\partial C_1}{\partial T} + \frac{dg}{dT}\left[\left\{\frac{1}{g} - \frac{g^2}{(Y+g)^3}\right\}C_1 + \left\{\frac{g^2}{(Y+g)^2} - 1\right\}\frac{\partial C_1}{\partial Y}\right] = \frac{\partial^2 C_1}{\partial Y^2} \tag{7}$$

$$C_1(0, Y) = 0 \tag{8a}$$

$$C_1(T, \infty) = 0 \tag{8b}$$

$$C_1(T, 0) = 1 \tag{8c}$$

$$\frac{dg}{dT} = N_a\left(-\frac{1}{g} + \frac{\partial C_1}{\partial Y}(T, 0)\right) \tag{9}$$

We observe that the transformations have produced a simplification of the boundary condition at the interface by removing the time dependence. On the other hand, the conservation equation has become more complex (compare Eqs. 1 and 7). It may be noted that the new coordinate Y is measured from the moving interface. In this reference frame, fluid at the interface is *stationary*, while the remaining fluid moves away or toward the interface depending on whether the bubble is dissolving or growing. This is reflected in the modified convective transport term in Eq. 7 which is negligible near the interface. In the limit of small time, the coefficient of dg/dT becomes negligibly small since in this regime, the concentration "boundary layer" is quite thin, and thus $Y + g \approx g$. If the term involving dg/dT is dropped for all times, and the solution of Eqs. 7 and 8 is used in Eq. 9, the classical quasi-stationary approximation is recovered.

Thus, we recognize that in the limit of $T \to 0$, a similarity solution is possible, and therefore extend this solution for finite but small T via an asymptotic expansion. We define

$$X = 2\sqrt{T} \tag{10a}$$

$$Z = \frac{Y}{2\sqrt{T}} \tag{10b}$$

$$F(X, Z) = C_1(T, Y) \tag{10c}$$

$$G(X) = g(T) \tag{10d}$$

The equations for $F(X, Z)$ and $G(X)$ are given below.

$$G(XZ + G)^3\left(2X\frac{\partial F}{\partial X} - 2Z\frac{\partial F}{\partial Z} - \frac{\partial^2 F}{\partial Z^2}\right)$$

$$+ 2\frac{dG}{dX}\left\{X^2(X^2Z^3 + 3X\,GZ^2 + 3G^2Z)F\right.$$

$$\left. - X(X^2GZ^3 + 3X\,G^2Z^2 + 2G^3\,Z)\frac{\partial F}{\partial Z}\right\} = 0 \quad (11)$$

$$G\frac{dG}{dX} = \frac{N_a}{2}\left\{-X + G\frac{\partial F}{\partial Z}(X, 0)\right\} \quad (12)$$

At this point, one may take the limit as $X \rightarrow 0$ and develop the zeroth order equations. These may then be subtracted, and suitable limiting processes employed to determine the appropriate asymptotic sequence of functions of X, and the higher order equations. However, inspection reveals that the following prescriptions may be used to generate the necessary asymptotic expansions.

$$F(X, Z) \sim \sum_{j=0}^{\infty} F_j(Z)\,X^j \quad (13a)$$

$$G(X) \sim \sum_{j=0}^{\infty} G_j\,X^j \quad (13b)$$

Substitution of Eqs. 13 into Eqs. 11 and 12 and subsequent matching of coefficients will lead to a set of ordinary differential equations for the coefficient functions $F_j(Z)$, and algebraic equations for the constants G_j. Similarly, substitution into the initial and boundary conditions given in Eqs. 8 will lead to the appropriate boundary conditions on $F_j(Z)$. The use of the initial condition on G reveals that $G_o = 1$. The relevant equations are reported in the Appendix. The equations for F_o may be solved immediately, and the result used to obtain G_1. The expressions for F_0 and G_1 then may be used in the governing equation for $F_1(Z)$. This equation, when solved, leads to a result for G_2, and the process may be continued to generate higher order results. As mentioned earlier, the zeroth order result for the transformed concentration field $F_0(Z)$ when used in Eq. 6 via Eq. 10c gives precisely the quasistationary concentration field. Thus, the present ordering procedure indeed produces the quasistationary field at the zeroth order. However, it should be recognized that the commonly employed quasistationary approximation involves the use of this zeroth order concentration field in the differential equation for the bubble radius (for instance, Eq. 12) without attempting to order this equation in X. Thus, the latter procedure is inconsistent in the sense that terms of all orders in X are permitted in the G equation while a concentration field valid only to $0(X^0)$ is used. The utility of the quasistationary result for the radius has been examined in other articles (Weinberg et al., 1980; Subramanian and Chi, 1980). It will be shown here that the present ordered approximations which are far simpler than the quasi-stationary result are more accurate than the latter.

The solution procedure for obtaining the functions F_j is tedious and details are given in the Appendix where solutions are reported for the first three functions F_0 to F_2. From these results, the following expressions for $G(X)$ and $H(X) = G^2$ may be obtained.

$$G(X) = 1 - \frac{N_a}{\sqrt{\pi}}X + \left(\frac{N_a}{3\pi} - \frac{1}{4}\right)N_a\,X^2$$

$$+ \left(\frac{5}{12} - \frac{8}{5\pi} + \frac{N_a}{18\pi}\right)\frac{N_a^2}{\sqrt{\pi}}X^3 + 0(X^4) \quad (14)$$

$$H(X) = 1 - \frac{2N_a}{\sqrt{\pi}}X + \left(\frac{5}{3\pi}N_a - \frac{1}{2}\right)N_a\,X^2$$

$$+ \left(\frac{4}{3} - \frac{16}{5\pi} - \frac{5}{9\pi}N_a\right)\frac{N_a^2}{\sqrt{\pi}}X^3 + 0(X^4) \quad (15)$$

The equation for F_3 is quite complex, and a formidable amount of bookkeeping and labor would be needed to obtain this function and hence the coefficient of X^4 in the G-expansion. This led us to seek alternative transformations which could result in simpler equations for the coefficient functions in the asymptotic expansion of the transformed concentration field. We found that transformations similar to those used by Duda and Vrentas (1969) in immobilizing the boundary, and transferring the curvature term did, indeed, lead to a simpler set of equations where the square of the scaled radius ($H = G^2$) appears as the primitive variable instead of the radius itself. Using these equations as a starting point, we were able to obtain an expansion for H to $0(X^4)$ which is given in the Appendix. However, it will be seen later that the above results good to $0(X^3)$ appear to be the best ones to use.

Relation to the Duda-Vrentas Expansion

In the case where the partial specific volume of the diffusing species in the liquid is negligible, the present N_a parameter is identical to that defined by Duda and Vrentas (1969) (with $N_b = 0$). Duda and Vrentas expanded their concentration field after suitable transformation in an asymptotic power series in N_a and obtained a result for $H(H = G^2)$ good to $0(N_a^2)$. It is interesting to examine the present expansion to determine if a direct relationship can be established between the two approaches. Specifically, it may be seen from Eq. 15 that as one goes to higher powers in X, the coefficients generally tend to contain higher powers in N_a. Unfortunately, we were unable to derive general results establishing lower bounds on the exponent of N_a in the coefficient of X^j for arbitrary j. However, it can be shown by expanding $L(X, W)$ and $H(X)$ in power series in N_a, matching coefficients, and solving the resulting equations via the present asymptotic expansion procedure that the *complete* $0(N_a)$ result is contained in Eq. 15 truncated at the X^2 term. This is, of course, in agreement with the result provided by Duda and Vrentas. The $0(N_a^2)$ equations are fairly complex, and although they most likely contain all higher powers of X, it is difficult to obtain results for terms beyond $0(X^4)$ without obtaining solutions of the higher order equations. In any event, one may employ Eq. 15 to obtain an *approximate expression* for H up to $0(N_a^2)$.

$$H = 1 - \left(\frac{2}{\sqrt{\pi}} + \frac{X}{2}\right)X\,N_a$$

$$+ \left(\frac{5}{3\pi} + \frac{X}{\sqrt{\pi}}\left\{\frac{4}{3} - \frac{16}{5\pi}\right\}\right)X^2N_a^2 + 0(N_a^3) \quad (16)$$

It should be re-emphasized that the coefficient of N_a^2 in Eq. 16 is incomplete since coefficients involving N_a^2 may appear in the higher order terms in the X-expansion of $H(X)$. However, we were encouraged to find that the coefficient of N_a^2 in Eq. 16 agrees quite closely with that given by Duda and Vrentas over a substantial range of values of time. More importantly, we observed that the results for H predicted from Eq. 16 for $|N_a| \lesssim 1.5$ were *practically indistinguishable* (in a plot) from the Duda-Vrentas result which we recomputed independently. For this purpose, the double integral appearing in the Duda and Vrentas result was evaluated numerically using Simpson's rule, and the accuracy of the calculations was verified by halving the interval sizes. We also checked our calculations (with $N_a = 0.2$) against the numerical results for H reported in this case by Duda and Vrentas. It will be seen later that $|N_a| \lesssim 1.5$ encompasses the range of N_a values where the $0(N_a^2)$ expansion is useful.

Relation to Scriven Solution

Scriven (1959) developed a similarity solution of the bubble growth problem for growth from zero initial size. If the gas density in the bubble is assumed negligible compared to the density of the liquid, his model equations would be similar to ours with the crucial difference that $g(0) = 0$. Of course, in this case, the reference length used in scaling our variables cannot be the initial bubble radius, but some arbitrarily chosen quantity. In identifying the relation to the Scriven solution, it is convenient to use Eq. 5, but not perform any transformations of the dependent variable $C_1(T, Y)$ such as the one shown in Eq. 6. Then, it may be seen that the new coordinate Z defined in Eq. 10b is related to the similarity coordinate η used by Scriven by an additive constant which is usually labelled the "Growth Constant" β.

$$Z = \eta - \beta \quad (17)$$

Thus, it is clear that in the special case $g(0) = 0$, for growth, we recover Scriven's equations and his solution at the zeroth order. Since his solution is an exact solution for this case, all the higher order corrections are identically zero, a fact which can be easily verified if the complete expansion is used.

For finite initial radius, in the case of growth, the present approach provides an ordered sequence of approximations to the bubble radius. When the bubble radius becomes much larger than the initial radius, it may be expected that the results will asymptotically approach the Scriven solution. However, for such large values of time, the present small-time expansion may not be suitable.

Finite Difference Solution

In order to establish the region of utility of the X-expansion to various orders, and the N_a-expansion, it is important to have available an accurate solution of Eqs. 1 to 4. For this purpose, we have solved these equations using the method of finite differences. The basic approach used is very similar to that suggested by Duda and Vrentas (1969). A transformation to a new coordinate θ defined by:

$$\theta = 1 - \exp[-\sigma\{r - g(T)\}] \quad (18)$$

is made so that the region $g \leq r < \infty$ is mapped into $0 \leq \theta < 1$ which is convenient for the finite difference formulation. Since the details of the procedure already have been presented elsewhere (Subramanian and Chi, 1980) they will be omitted here.

RESULTS AND DISCUSSION

Here we present the results of calculations of the time dependence of bubble radii (and h) employing Eqs. 14 and 15. It may be noted that the symbols h and H represent the same entity, namely, the square of the reduced radius. The different symbols were used in the text to identify the different functional dependences on their respective arguments T and X. However, in the discussion, the symbols h and H will be used interchangeably (the same applies to g and G). These results will be compared with values obtained via finite difference and N_a expansion techniques for a wide range of N_a values. Bubble growth will be considered as well as bubble dissolution. However, computations pertaining to the latter process will predominate since the prediction of bubble radii at times approaching dissolution provides the most demanding test of approximate techniques.

In Figure 1 the square of the reduced bubble radius (h) is plotted against reduced time for $N_a = 1.5$. The solid line shows the values of h found by use of the numerical solution of the finite difference equations. These results are accurate to at least three significant figures, and will be taken as the exact solution of Eqs. 1-4 for purposes of evaluation of the approximate methods. The remaining curves in this figure illustrate the time dependence of h found using Eq. 15 to different orders in X. The dotted curve was generated by employing the result from Eq. 15 plus the $0(X^4)$ contribution derived in the Appendix. One notes that at long times this calculation produces a divergence in h. This behavior clearly illustrates the asymptotic nature of the "small time" expansion. *As is evident from Figure 1, and as we observed to be generally true with exceptions noted below, the expansion taken through $0(X^3)$ proves to be the most accurate expression for h (or g).* In fact, the dashed curve in Figure 1

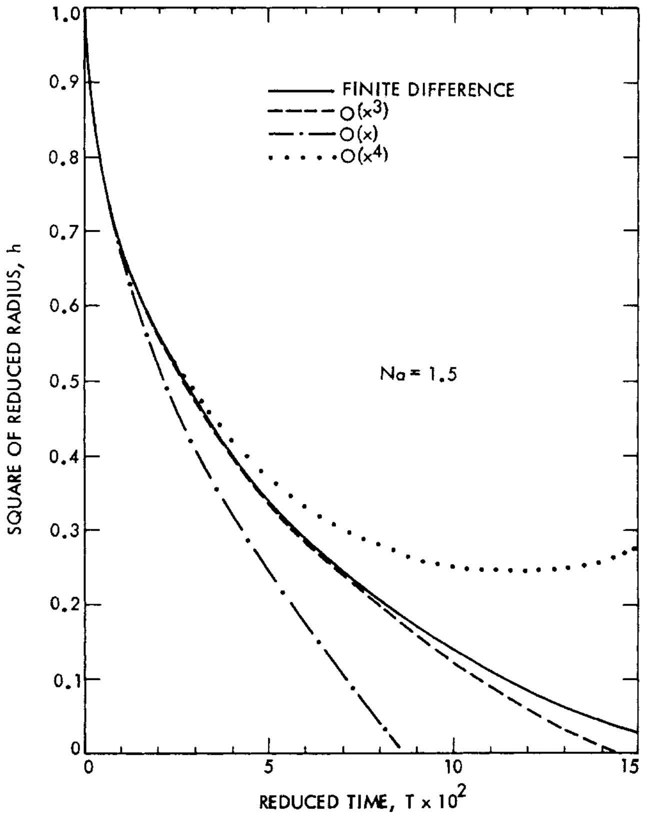

Figure 1. Comparison of the square of reduced radius (h) as a function of reduced time (T) calculated from the finite difference, $0(X)$, $0(X^3)$, and $0(X^4)$ solutions; N_a = 1.5.

($0(X^3)$) shows excellent agreement with the finite difference results, except at the largest times where there are moderate discrepancies. The expansion through $0(X^2)$ yields h values

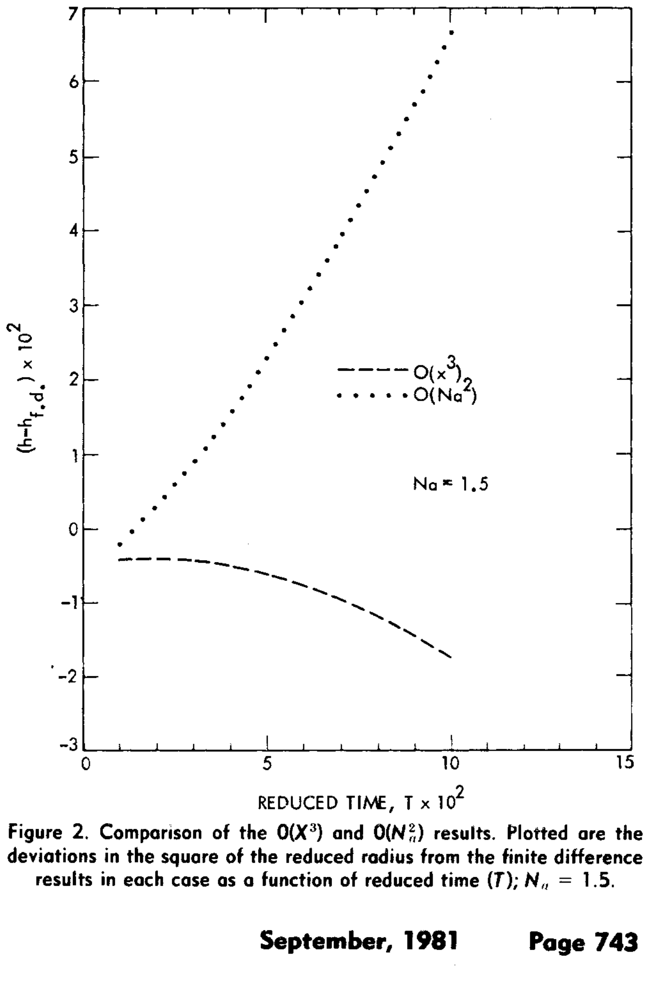

Figure 2. Comparison of the $0(X^3)$ and $0(N_a^2)$ results. Plotted are the deviations in the square of the reduced radius from the finite difference results in each case as a function of reduced time (T); N_a = 1.5.

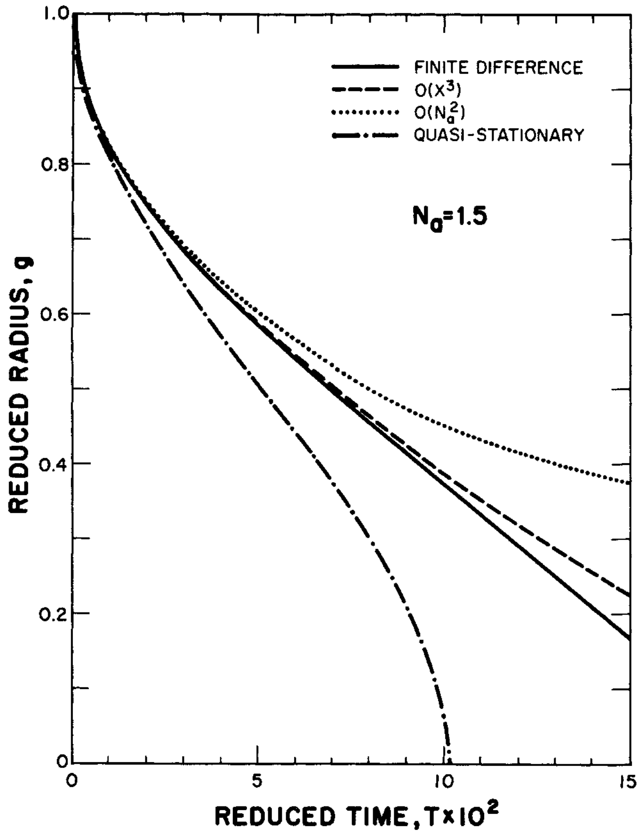

Figure 3. Comparison of the reduced radius (g) as a function of reduced time (T) calculated from the finite difference, $0(X^3)$, quasistationary and $0(N_a^2)$ solutions; N_a = 1.5.

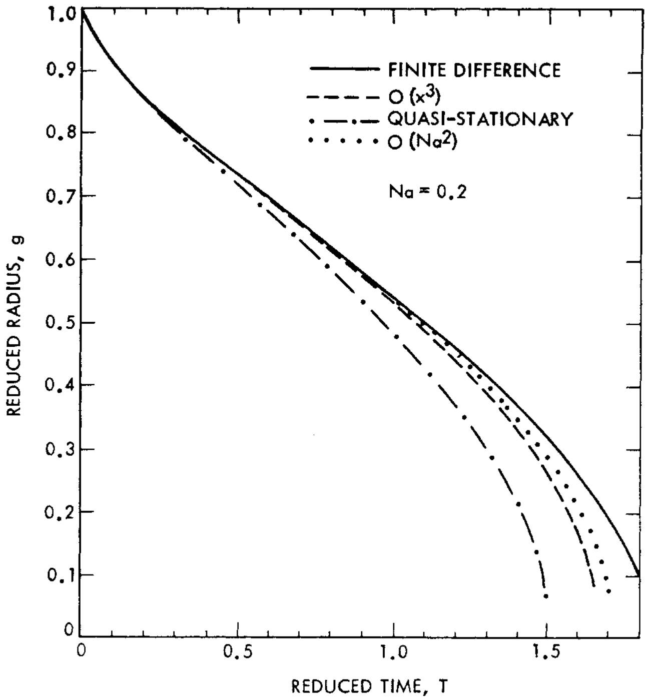

Figure 4. Comparison of the reduced radius (g) as a function of reduced time (T) calculated from the finite difference, $0(X^3)$, quasistationary, and $0(N_a^2)$ solutions; N_a = 0.2.

nearly identical to those given by the $0(X^3)$ expansion and hence is not shown in the figure. Also shown in the figure is a linear approximation for h in X which, as may be expected, performs rather poorly.

Duda and Vrentas demonstrated that their expansion to $0(N_a^2)$ is excellent at $N_a = 0.2$. We have, in fact, verified by independent calculations (not shown here) that their expansion does quite well even at a value of $N_a = 0.5$. Therefore, it is interesting to determine the bounds of the region where the $0(N_a^2)$ expansion is useful. With this aim in mind, we have tried to provide several comparisons of results from our $0(X^3)$ expansion, the Duda-Vrentas $0(N_a^2)$ expansion, and the finite difference results. For this purpose we evaluated the Duda and Vrentas results independently as indicated earlier. However, on the plots shown, our Eq. 16 yields results which are indistinguishable

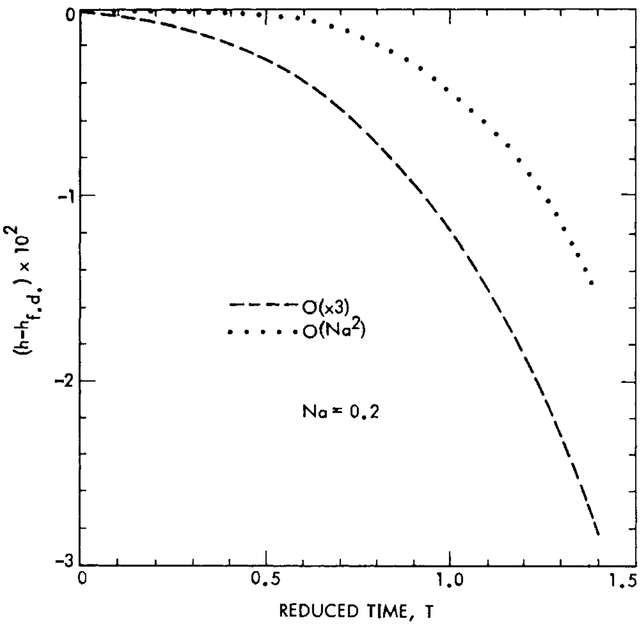

Figure 5. Comparison of the $0(X^3)$ and $0(N_a^2)$ results. Plotted are the deviations in the square of the reduced radius from the finite difference results in each case as a function of reduced time; N_a = 0.2.

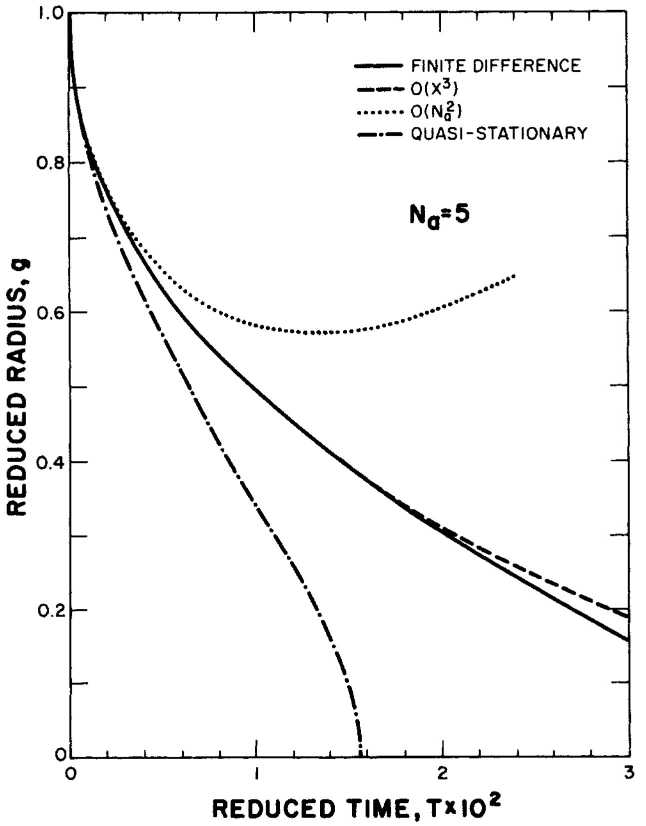

Figure 6. Comparison of the reduced radius (g) as a function of reduced time (T) calculated from the finite difference, $0(X^3)$, quasistationary, and $0(N_a^2)$ solutions; N_a = 5.

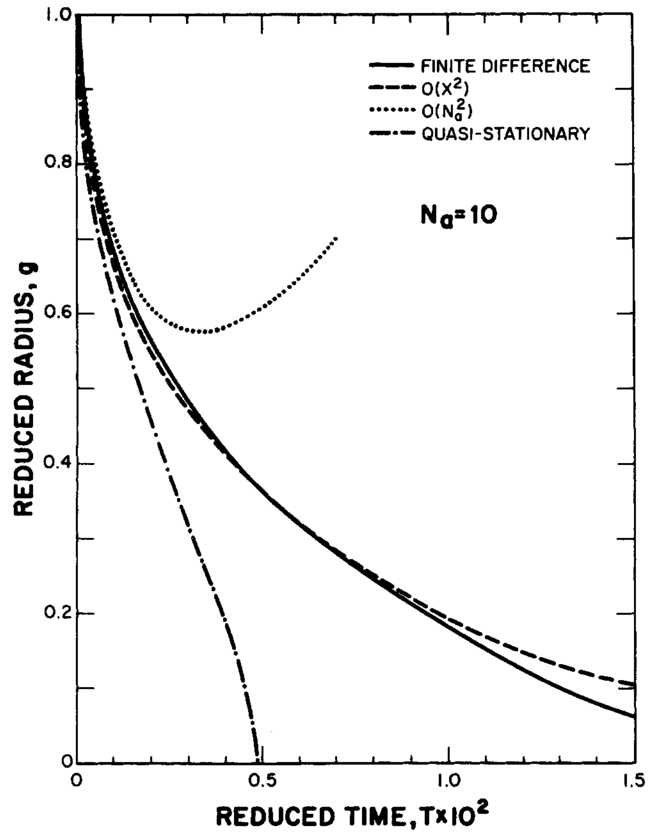

Figure 7. Comparison of the reduced radius (g) as a function of reduced time (T) calculated from the finite difference, $0(X^2)$, quasistationary, and $0(N_a^2)$ solutions; N_a = 10.

from those of Duda and Vrentas. In Figure 2, we illustrate the differences among the finite difference (exact) results and the two approximate calculations of h for the same N_a value of 1.5. The dashed line refers to the $0(X)^3$ calculation described above and the dotted line was drawn using the Duda and Vrentas expansion through $0(N_a^2)$. It is clear that the $0(X)^3$ expansion gives a more accurate result for h than does the $0(N_a^2)$ expansion of Duda and Vrentas for this value of N_a.

In Figure 3 additional comparisons are made among the Duda and Vrentas expansion to $0(N_a^2)$, our expansion to $0(X^3)$, the quasi-stationary approximation, and the finite difference results. The N_a value is again 1.5, but here the reduced radius, g, is shown as a function of time. Although the $0(X^3)$ expansion is superior to the $0(N_a^2)$ and quasi-stationary results over the time domain shown, all of these calculations exhibit moderately large errors at times close to bubble dissolution.

At the smaller value of N_a = 0.2, both the $0(X^3)$ and $0(N_a^2)$ results for g agree quite well with the finite difference results as shown in Figure 4. We generally observed that for small values of N_a, the H-expansion to $0(X^3)$ proved to be *slightly* more accurate than the G-expansion to the same order even though both expansions formally possess the same degree of accuracy. Thus, we have shown in Figure 4, the square root of the result for H from Eq. 15. In addition, the results from the quasistationary model (Weinberg et al., 1980) also are shown for comparison in Figure 4. Clearly, the quasi-stationary procedure is inferior to either of the two approximate techniques mentioned above. From an inspection of Figure 4, it may be noted that the $0(N_a^2)$ expansion does somewhat better than the $0(X^3)$ expansion over the entire time regime. This feature is illustrated more dramatically in Figure 5 where the differences among the finite difference and $0(N_a^2)$ and $0(X^3)$ results for h are shown for N_a = 0.2.

In general, one would anticipate that the Duda and Vrentas expansion would become more accurate as the value of N_a is decreased. On the other hand, the X-expansion yields quite accurate bubble radii at large N_a since large N_a implies small dissolution times. These points are well illustrated in Figures 6

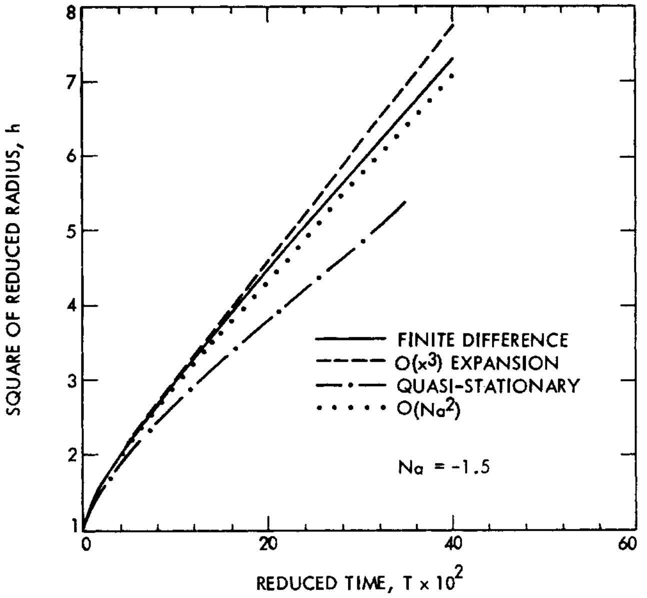

Figure 8. Comparison of the square of the reduced radius (h) as a function of reduced time (T) calculated from the finite difference, $0(X^3)$, quasi-stationary, and $0(N_a^2)$ solutions; N_a = −1.5 (growth).

and 7 where the reduced bubble radius is shown as a function of time for two large N_a values (5 and 10, respectively). One observes that while there is very good agreement between the finite difference values of g and the results from the X-expansion, the $0(N_a^2)$ calculation diverges for both N_a values. As mentioned earlier, there is no reason to expect the small -N_a expansion to do well at such large values of N_a, and the reason for its inclusion is its remarkable performance at N_a = 1.5. It might be noted that for N_a = 10, the $0(X^2)$ result for G does slightly better than the $0(X^3)$ values, and therefore, has been used. Also included for comparison in Figures 6 and 7 is the quasistationary solution which may be seen to do poorly in comparison to the X-expansions. Large N_a values correspond physically to large

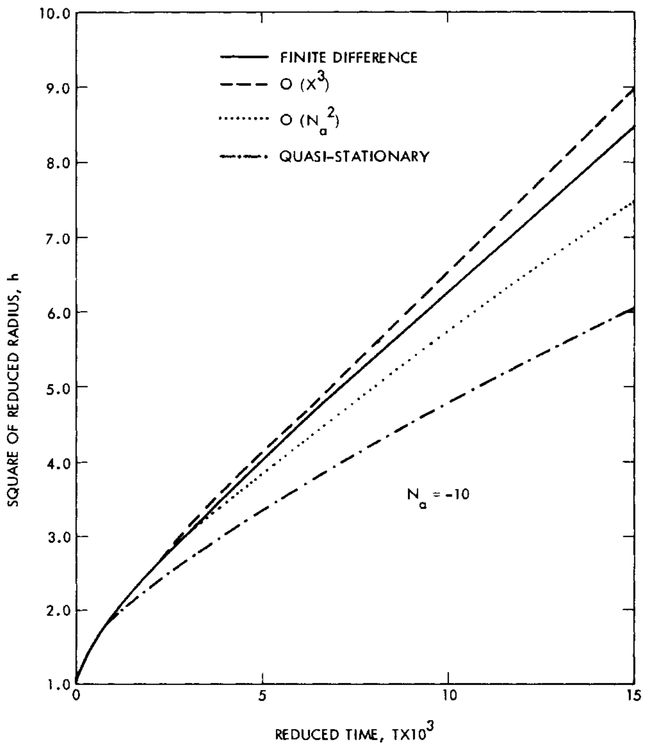

Figure 9. Comparison of the square of the reduced radius (h) as a function of reduced time (T) calculated from the finite difference, $0(X^3)$, quasi-stationary, and $0(N_a^2)$ solutions; N_a = −10 (growth).

gas solubility in the liquid and/or a very undersaturated liquid. Such situations occur commonly, and the values of N_a used here are physically realistic ones. Of course, for extremely large N_a, the partial specific volume of the dissolved species in the liquid would be non-negligible, and the assumption of isothermality would not be as valid.

Figures 8 and 9 are employed to demonstrate our calculations as applied to the problem of bubble growth from a non-zero initial bubble radius. In Figure 8 we have plotted $0(X^3)$, $0(N_a^2)$, and quasistationary calculations as well as the finite difference values for h for $N_a = -1.5$. While all the solutions are quite close, the $0(N_a^2)$ result here gives the best overall agreement with the finite difference results. However, for a larger $|N_a|$, namely $N_a = -10$, the $0(X^3)$ expansion does better as shown in Figure 9. It is interesting to note that the $0(N_a^2)$ approximation can adequately predict bubble radius for *growth* even at such a large value of $|N_a|$, while it fails quite badly in describing bubble *dissolution* for the same value of $|N_a|$. (Compare Figures 7 and 9.)

One also notes that the quasistationary approximation performs much better for the case of bubble growth. For example, from an inspection of Figure 8 it is seen that at $T = 5 \times 10^{-2}$, the reduced time required for the bubble radius to roughly double in size, the quasistationary prediction for h is excellent. However, in the case of bubble dissolution when the initial bubble radius has been reduced by about 75% of its initial values, serious errors are apparent in the use of the quasi-stationary result, Figure 4. Furthermore, it is clear from this figure that at long times the approximate calculations as well as the exact finite difference calculation predict a very rapid bubble shrinkage. These findings may be explained in a qualitative fashion by recourse to an inspection of Eqs. 7 and 9. One observes that one term in the expression for the rate of change of reduced bubble radius with time is inversely proportional to the reduced bubble radius (Eq. 9). Thus, for bubble dissolution problems when the reduced bubble radius is small the magnitude of g^{-1} will be large and the rate of reduction of the bubble radius will be sizeable. In the case of bubble growth, however, g^{-1} will decrease with time. As mentioned previously, the quasi-stationary approximation consists of neglecting the convective term in Eq. 7 which is proportional to dg/dT. In the N_a expansion procedure, the convective term is neglected in the first order result, and although it is included in higher orders it is treated as a small quantity. However, as illustrated above dg/dT attains a sizeable magnitude in the time regime close to bubble dissolution. This fact causes the observed asymmetry in the performance of the various approximations when applied to dissolution and growth problems and is responsible for their breakdown close to the dissolution point.

Another point worth noting from the finite difference results for growth is that the square of the reduced radius is practically linear in reduced time by the time the bubble has grown to about three times its initial radius. This is in agreement with the expectation that the Scriven solution is approached as an asymptote at large values of time. While the actual radius values are not sufficiently large for good agreement with the Scriven solution to be expected, the slopes dh/dT are remarkably close to the values predicted by Scriven ($4\beta^2$) for growth from zero initial size. For instance, for $N_a = -1.5$, when the radius is approximately 2.95 times the initial value, the value of dh/dT, from the finite difference result, is only a fraction of a percent away from the Scriven result. For $N_a = -10$, the deviation is approximately 7% when the radius is 3.25 times the initial value.

In summary, we may reiterate the virtues of the asymptotic expansion presented in this work. For large values of N_a, it predicts bubble shrinkage or growth more accurately than any approximate technique given to date. For small N_a values, a truncated version of the present expansion (Eq. 16) yields results for h which are virtually identical with those given by Duda and Vrentas to $0(N_a^2)$. In either case, the analytical results for h (and g) are extremely simple, and may be evaluated without resort to machine computation.

ACKNOWLEDGMENT

The authors are grateful for the financial support of this work provided by a contract from the National Aeronautics and Space Administration. Also, R. S. Subramanian wishes to express his gratitude for the hospitality extended to him by the Jet Propulsion Laboratory and the Chemical Engineering Department of the California Institute of Technology during his sabbatical year.

APPENDIX

The procedure employed for the solution of equations (11) and (12) along with the associated initial and boundary conditions by the use of the expansions presented in equations (13) is given below.

When the solution form given in equation (13a) is introduced into equation (11), and the coefficients of X^j are matched, the following set of ordinary differential equations for $F_j(Z)$ may be obtained upon some rearrangement and simplification.

$$\begin{aligned} F_j'' + 2Z\,F_j' - 2j\,F_j = \sum_{l=0}^{j-1} \Big\{ & -[Z^3\,G_{j-l-3} + 3\,Z^2\,H_{j-l-2} + 3\,Z\,I_{j-l-1} + J_{j-l}]F_l'' \\ & - \Big[2Z^4\,G_{j-l-3} + (j-l+4)\,Z^3\,H_{j-l-2} + 2Z^2(j-l+2)I_{j-l-1} + (j-l+2)\,Z\,J_{j-1}\Big]F_l' \\ & + \Big[l(2Z^3\,G_{j-l-3} + 6Z^2\,H_{j-l-2} + 6Z\,I_{j-l-1} + 2\,J_{j-l}) \\ & + \Big(2[j-l-3]Z^3\,G_{j-l-3} + 3[j-l-2]Z^2\,H_{j-l-2} + 2[j-l-1]Z\,I_{j-l-1}\Big)\Big]F_l\Big\} \end{aligned} \quad \text{(A-1)}$$

Here, H_j, I_j and J_j are the coefficients appearing in the asymptotic expansions of G^2, G^3 and G^4 respectively as shown below.

$$H = G^2 \sim \sum_{j=0}^{\infty} H_j\,X^j \quad \text{(A-2a)}$$

$$I = G^3 \sim \sum_{j=0}^{\infty} I_j\,X^j \quad \text{(A-2b)}$$

$$J = G^4 \sim \sum_{j=0}^{\infty} J_j\,X^j \quad \text{(A-2c)}$$

The coefficients H_j, I_j and J_j are related to ones of lower orders by the following relations.

$$H_j = \sum_{l=0}^{j} G_l\,G_{j-l} \quad \text{(A-3a)}$$

$$I_j = \sum_{l=0}^{j} G_l\,H_{j-l} \quad \text{(A-3b)}$$

$$J_j = \sum_{l=0}^{j} G_l\,I_{j-l} \quad \text{(A-3c)}$$

In equation (A-1), the convention that all functions and coefficients with negative subscripts are zero has been employed. The following boundary conditions on the functions $F_j(Z)$ may be derived by the use of equations (13a), (10) and (8).

$$F_j(0) = \delta_{j0} \quad \text{(A-4)}$$

$$F_j(\infty) = 0 \quad \text{(A-5)}$$

Furthermore, use of equation (13b) along with (10) and (4) leads to

$$G_0 = 1 \quad \text{(A-6)}$$

The introduction of equations (13) in equation (12), followed by the matching of the coefficients of like powers of X, the use of equation (A-6), and some simplification leads to the final result for G_{j+1} given below.

$$G_{j+1} = \frac{1}{j+1}\left[\frac{N_a}{2}\,(F_j'(0) - \delta_{j1})\right.$$

$$+ \sum_{n=1}^{j} G_n\left(\frac{N_a}{2} F'_{j-n}(0) - n\, G_{j+1-n}\right)\Bigg], \quad j \geq 0 \quad \text{(A-7)}$$

It may be observed that when $F'_j(0)$ is known for a given j, the next order coefficient G_{j+1} may be computed from equation (A-7).

The Solution for F_0 and G_1

The equations for F_0 obtained from equations (A-1), (A-4) and (A-5) by setting $j = 0$ are given below.

$$F''_0 + 2Z\, F'_0 = 0 \quad \text{(A-8)}$$

$$F_0(0) = 1 \quad \text{(A-9a)}$$

$$F_0(\infty) = 0 \quad \text{(A-9b)}$$

The solution is

$$F_0(Z) = erfc(Z) \quad \text{(A-10)}$$

The use of equation (A-10) along with equation (A-7) for $j = 0$ yields

$$G_1 = \frac{N_a}{2} F'_0(0) = \frac{-N_a}{\sqrt{\pi}} \quad \text{(A-11)}$$

The Solutions for Higher Order Coefficients

The use of the results for F_0 and G_1, along with the relations (A-3) in equation (A-1) for $j = 1$ leads to the following differential equation.

$$F''_1 + 2Z\, F'_1 - 2F_1 = \frac{-8}{\pi} N_a\, Z \exp(-Z^2) \quad \text{(A-12)}$$

Along with the homogeneous boundary conditions given in equations (A-4) and (A-5) for $j = 1$, equation (A-12) may be solved to yield

$$F_1(Z) = \frac{4N_a}{3\pi} Z \exp(-Z^2) \quad \text{(A-13)}$$

From equation (A-7) for $j = 1$, and equation (A-13) we get

$$G_2 = \frac{N_a}{4}\left[F'_1(0) - 1\right] = N_a\left(\frac{N_a}{3\pi} - \frac{1}{4}\right) \quad \text{(A-14)}$$

Similarly, the next higher function F_2 can be shown to satisfy

$$F''_2 + 2Z\, F'_2 - 4F_2$$

$$= \frac{4N_a}{\sqrt{\pi}} Z \exp(-Z^2)\left[\frac{2N_a}{3\pi} - 1 + \frac{3}{\sqrt{\pi}} Z - \frac{8N_a}{3\pi} Z^2\right] - \frac{6N_a}{\sqrt{\pi}} Z\, erfc(Z) \quad \text{(A-15)}$$

The solution for $F_2(Z)$ which satisfies equation (A-15) and the homogeneous boundary conditions from equations (A-4) and (A-5) for $j = 2$ is given below.

$$F_2(Z) = \frac{48N_a}{5\pi} i^2\, erfc(Z) + \frac{3N_a}{\sqrt{\pi}} Z\, erfc(Z)$$

$$+ \frac{N_a}{\sqrt{\pi}} \exp(-Z^2)\left[\frac{8N_a}{9\pi} Z^3 - \frac{6}{5\sqrt{\pi}} Z^2 + \left(\frac{N_a}{3\pi} + \frac{1}{2}\right)Z - \frac{12}{5\sqrt{\pi}}\right] \quad \text{(A-16)}$$

Here, $i^2\, erfc(Z)$ is a repeated integral of the error function defined in Abramowitz and Stegun (1968).

The result for G_3 from equation (A-7) for $j = 2$ and equation (A-16) is

$$G_3 = \frac{N_a}{6}[F'_2(0) + G_1\, F'_1(0) + G_2\, F'_0(0)] - G_1\, G_2$$

$$= \left(\frac{5}{12} - \frac{8}{5\pi} + \frac{N_a}{18\pi}\right)\frac{N_a^2}{\sqrt{\pi}} \quad \text{(A-17)}$$

The Alternative Formulation

As indicated in the text, the calculation of results to the next higher order in the above equations, while straightforward in principle, entails a formidable task in bookkeeping. This led us to seek alternative transformations to obtain simpler starting equations. We found that the transformations employed by Duda and Vrentas (1969) for immobilizing the interface and transferring the curvature effect were quite suited for our purposes. Thus, in solving equations (1) to (4), we let

$$y = \frac{r}{g(T)} - 1 \quad \text{(A-18a)}$$

$$C_1(T, y) = (1 + y)\, C(T, r) \quad \text{(A-18b)}$$

$$h(T) = g^2(T) \quad \text{(A-18c)}$$

so that $C_1(T, y)$ and $h(T)$ satisfy the following set of equations.

$$h\frac{\partial C_1}{\partial T} + \frac{1}{2}\frac{dh}{dT}\left\{\frac{1}{(1 + y)^2} - (1 + y)\right\}\left\{\frac{\partial C_1}{\partial y} - \frac{C_1}{(1 + y)}\right\} = \frac{\partial^2 C_1}{\partial y^2} \quad \text{(A-19)}$$

$$C_1(0, y) = 0 \quad \text{(A-20a)}$$

$$C_1(T, \infty) = 0 \quad \text{(A-20b)}$$

$$C_1(T, 0) = 1 \quad \text{(A-20c)}$$

$$\frac{dh}{dT} = 2N_a\left\{-1 + \frac{\partial C_1}{\partial y}(T, 0)\right\} \quad \text{(A-21)}$$

$$h(0) = 1 \quad \text{(A-22)}$$

Now, define

$$X = 2\sqrt{T} \quad \text{(A-23a)}$$

$$W = \frac{y}{2\sqrt{T}} \quad \text{(A-23b)}$$

$$L(X, W) = C_1(T, y) \quad \text{(A-23c)}$$

$$H(X) = h(T) \quad \text{(A-23d)}$$

and expand $L(X, W)$ and $H(X)$ in asymptotic power series as before.

$$L(X, W) = \sum_{j=0}^{\infty} L_j(W) X^j \quad \text{(A-24a)}$$

$$H(X) = \sum_{j=0}^{\infty} H_j X^j \quad \text{(A-24b)}$$

The manipulations are similar to the earlier set so that the details will be omitted. Only the results for the coefficients H_j will be given below. In general,

$$H_0 = 1 \quad \text{(A-25a)}$$

$$H_{j+1} = \frac{N_a}{(j + 1)}\{L'_j(0) - \delta_{1j}\}, \quad j \geq 0 \quad \text{(A-25b)}$$

We get

$$H_1 = -\frac{2}{\sqrt{\pi}} N_a \quad \text{(A-26a)}$$

$$H_2 = N_a\left(\frac{5N_a}{3\pi} - \frac{1}{2}\right) \quad \text{(A-26b)}$$

$$H_3 = \frac{N_a^2}{\sqrt{\pi}}\left(\frac{4}{3} - \frac{16}{5\pi} - \frac{5N_a}{9\pi}\right) \quad \text{(A-26c)}$$

and

$$H_4 = N_a^2\left[\frac{83}{35\pi} - \frac{3}{4} + \frac{N_a}{\pi}\left\{\frac{63,706}{4,725\pi} - \frac{15,157}{3,360}\right\} + \frac{128}{63\pi^2}N_a^2\right] \quad \text{(A-26d)}$$

It should be noted that the expansions for the square of the radius given in equations (A-2a) and (A-24b) are identical since the expansion variable is precisely the same quantity ($X = 2\sqrt{T}$). Thus, the first four coefficients H_0 to H_3 calculated from equations (A-3a) and the results for G_0 to G_3 from equations (A-6), (A-11), (A-14), and (A-17) exactly match the results in equations (A-25a) and (A-26). However, in view of the different transformations used in arriving at the transformed concentration fields $F(X, Z)$ and $L(X, W)$, the coefficient functions $F_j(Z)$ and $L_j(W)$ are not equivalent functions of their arguments.

NOTATION

a = bubble radius
a_o = initial value of bubble radius
c = concentration of solute in the liquid
c_o = initial concentration of solute
c_i = concentration of solute at the interface
$C(T, r)$ = scaled concentration field of the solute in the liquid; $C = (c - c_o)/(c_i - c_o)$
$C_1(T, Y)$ = transformed concentration fields; defined in Eq. 6, for example
D = diffusion coefficient of the dissolved gas in the liquid
$F(X, Z)$ = transformed concentration field; $F(X, Z) = C_1(T, Y)$
$F_j(Z)$ = expansion coefficients defined in Equation (13a)
g = scaled bubble radius; $g = a/a_o$
$G(X)$ = scaled bubble radius; $G(X) = g(T)$
G_j = expansion coefficients defined in Eq. 13b
$h(T)$ = square of the scaled bubble radius; $h(T) = g^2(T)$
$H(X)$ = square of the scaled bubble radius; $H(X) = h(T)$
H_j = expansion coefficients defined in Eq. A-2a
$i^n erfc$ = repeated integral of the error function defined in Abramowitz and Stegun (1968)
I_j = expansion coefficients defined in Eq. A-2b
J_j = expansion coefficients defined in Eq. A-2c
$L(X, W)$ = transformed concentration field defined in Eq. A-23c
N_a = driving force parameter; $N_a = (c_i - c_\infty)/\rho$
r = radial distance (from bubble center) scaled by the initial value of the bubble radius
t = time
T = scaled time; $T = Dt/a_o^2$
W = new independent variable defined in Eq. A-23b
X = new independent variable defined in Eq. 10a
y = new independent variable defined in Eq. A-18a
Y = new independent variable defined in Eq. 5
Z = new independent variable defined in Eq. 10b

Greek Letters

β = "growth" constant appearing in Eq. 17
δ_{ij} = Kronecker delta function
η = Scriven's similarity coordinate; related to Z by Eq. 17
θ = scaled distance coordinate defined in Eq. 18
ρ = gas density in the bubble
σ = "stretching parameter" (Eq. 18)

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Long-Range Predictive Control

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Two process computer control algorithms which are based on long-range prediction are outlined. The methods are compared, similarities pointed out, and each is shown to become similar to dead-beat control under certain circumstances.

SCOPE

Two new process computer control techniques have been presented in the literature that have proven useful in industrial applications. Their success and the fact that both methods differ from the conventional state space or transfer function approaches have caused much interest in the petrochemical control community. Each of the new techniques is based on a non-minimal representation of the process, that is, the usual

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