Mathematical Analysis of Bubble Dissolution

J. L. DUDA and J. S. VRENTAS
The Dow Chemical Company, Midland, Michigan

A perturbation series solution is derived for isothermal bubble dissolution and bubble growth from an initially finite size. The new solution is superior in several aspects to the quasi-stationary and quasi-steady state approximations derived previously. The accuracy and range of validity of the new results are investigated by comparison with finite-difference solutions of the equations governing bubble growth or dissolution. In addition, previous numerical solutions of the problem are compared to the finite-difference results of this study.

Numerous studies have been concerned with the analysis of bubble dissolution in undersaturated liquids either because the bubble behavior is of direct interest such as in the rate of removal of gas bubbles in glass making (*9*) or because the measurement of bubble dissolution serves as a useful technique for determining the diffusivities of dissolved gases in liquids (*11*). The commonly considered problem involves dissolution of an isolated stationary spherical bubble controlled by diffusion in a liquid phase of infinite extent. Mathematically, this problem reduces to the solution of the unsteady state diffusion equation subject to appropriate initial and boundary conditions and to the existence of a free boundary. The moving phase boundary removes this problem from the linear domain and special methods must be utilized to circumvent this difficulty. For the case of a bubble growing from a zero initial radius, exact solutions based on a similarity transformation have been obtained (*3, 10, 14*). However, because of the finite initial bubble size, the partial differential equation describing either bubble dissolution or growth from nonzero initial radius cannot be cast into an ordinary differential equation by a similarity transformation, and no exact solutions are available for these instances.

The rate of growth of the thickness of the concentration boundary layer around a dissolving bubble is usually fast compared to the rate of movement of the free surface, and this particular characteristic has been the basis for approximate solutions of the bubble dissolution problem. Epstein and Plesset (*7*) and others (*5, 8, 11*) have obtained approximate quasi-stationary solutions by neglecting the convective transport and by solving the resulting diffusion equation for the case where the bubble surface is considered stationary. The mass flux at the phase boundary determined from the solution of the simplified diffusion equation is then used to establish the motion of the bubble surface. Further simplification of the diffusion equation by neglecting the time derivative results in a more approximate solution which is usually referred to as the *quasi-steady state approximation* (*2*). The approximations incorporated in these solutions appear reasonable but the way they are imposed is not as rigorous from a mathematical point of view as a more formal approach might be.

Recently, numerical solutions for specific cases of bubble dissolution have appeared in the literature (*4, 13*) as attempts to gain insight into the bubble dissolution process. The results of Cable and Evans (*4*) show that both the quasi-stationary and the quasi-steady state approximations are in considerable variance with the finite-difference results. Since the latter results are considered more accurate because the complete equations are solved, the validity of the approximate analytical solutions appears to be in considerable doubt. However, the numerical methods applied are also suspect, so that the exact nature of the bubble dissolution process remains an unanswered question, as is also the possibility of formulating useful approximate analytical solutions to the problem.

Therefore, further examination of the gas bubble dissolution problem is undertaken in this investigation. A perturbation technique described elsewhere (*6*) is used to obtain a series solution for the complete diffusion equation and the associated boundary conditions. The zero-order term of this solution is equivalent to the quasi-stationary solutions derived previously but in a simpler form, and the formulation of the first-order term extends the range of applicability further than the idealized stationary solution. There are basically three reasons for utilizing a perturbation series technique (which essentially is a systematic scheme of successive approximations) to analyze the bubble dissolution problem. First, the accuracy and range of validity of the zero-order results can be extended by the addition of higher order terms. Thus, the gap between quasi-stationary solutions for spheres and the thin boundary-layer solutions of the type proposed by Plesset and Zwick (*12*) is partially filled, and matching of the two regimes can more easily be accomplished. Secondly, embedding the quasi-stationary approximation as a first step in an orderly scheme of successive approximations is mathematically more rigorous than intuitively eliminating certain terms. Finally, higher-order approximations often provide insight about the nature of the zero-order result, as is dramatically illustrated below.

The perturbation analysis is shown to be valid over a limited range for the description of the related problem of growth of an initially finite sphere in a supersaturated liquid. In addition, numerical solutions of the finite-difference forms of the exact equations describing the bubble dissolution and bubble growth problems are obtained in order to check the approximate perturbation solutions. These solutions indicate that in many cases the previously presented numerical solutions contain significant errors due presumably to inadequate finite-difference approximations for the concentration gradients near the surface of the bubble.

MATHEMATICAL ANALYSIS

Formulation of Equations

Consider the isothermal growth or dissolution of a one-component spherical bubble into an infinite liquid phase of binary constitution. The velocity field in the outer phase is purely radial, the concentration field is spherically symmetric, and the bubble is considered a perfect, isolated sphere. We further assume no chemical reactions, constant partial specific volumes, a constant diffusion coefficient, and a uniform initial concentration profile for the outer liquid phase. Finally, we consider situations for which the interior of the bubble can be assumed uniform and for which inertial, viscous, and surface tension effects are completely negligible. Thus, the equations of motion for the liquid phase predict that the internal bubble pressure can be set equal to the ambient pressure with negligible error, and, consequently, the growth or dissolution of the bubble is controlled strictly by the diffusion process in the liquid phase. It follows then that the bubble dynamics can be described by the following set of equations, where the origin of the coordinate system is the drop center which is assumed to be at rest:

$$\frac{\partial \rho_I}{\partial t} + \frac{f(t)}{r^2} \frac{\partial \rho_I}{\partial r} = D\left(\frac{\partial^2 \rho_I}{\partial r^2} + \frac{2}{r} \frac{\partial \rho_I}{\partial r} \right) \tag{1}$$

$$r^2 \overline{v} = R^2 \overline{v}(R, t) = f(t) \tag{2}$$

$$\overline{v}(R, t) = \frac{D(1 - \hat{V}_I \hat{\rho}) \left(\frac{\partial \rho_I}{\partial r} \right)_{r=R}}{\hat{\rho}(1 - \hat{V}_I \rho_{IE})} \tag{3}$$

$$\rho = \frac{1 - \hat{V}_I \rho_I + \hat{V}_J \rho_I}{\hat{V}_J} \tag{4}$$

$$\rho_I(r, 0) = \rho_{IO} \qquad r > R_o \tag{5}$$

$$\rho_I(\infty, t) = \rho_{IO} \qquad t \geqq 0 \tag{6}$$

$$\rho_I[R(t), t] = \rho_{IE} \qquad t > 0 \tag{7}$$

$$\frac{dR}{dt} = \frac{D \left(\frac{\partial \rho_I}{\partial r} \right)_{r=R}}{\hat{\rho}(1 - \hat{V}_I \rho_{IE})} \tag{8}$$

$$R(0) = R_o + 0 \tag{9}$$

Introduction of the dimensionless variables

$$\rho_I^* = \frac{\rho_I - \rho_{IO}}{\rho_{IE} - \rho_{IO}} \tag{10}$$

$$R^* = \frac{R}{R_o} \tag{11}$$

$$r^* = \frac{r}{R_o} \tag{12}$$

$$t^* = \frac{Dt}{R_o^2} \tag{13}$$

converts the pertinent equations above to

$$\frac{\partial \rho_I}{\partial t} + \frac{R^2}{r^2}(N_a - N_b)\left(\frac{\partial \rho_I}{\partial r} \right)\left(\frac{\partial \rho_I}{\partial r} \right)_{r=R} = \frac{\partial^2 \rho_I}{\partial r^2} + \frac{2}{r} \frac{\partial \rho_I}{\partial r} \tag{14}$$

$$\rho_I(r, 0) = 0 \qquad r > 1 \tag{15}$$

$$\rho_I(\infty, t) = 0 \qquad t \geqq 0 \tag{16}$$

$$\rho_I(R, t) = 1 \qquad t > 0 \tag{17}$$

$$\frac{dR}{dt} = N_a \left(\frac{\partial \rho_I}{\partial r} \right)_{r=R} \tag{18}$$

$$R(0) = 1 \tag{19}$$

$$N_a = \frac{\rho_{IE} - \rho_{IO}}{\hat{\rho}(1 - \hat{V}_I \rho_{IE})} \tag{20}$$

$$N_b = \frac{\rho_{IE} - \rho_{IO}}{\frac{1}{\hat{V}_I} - \rho_{IE}} \tag{21}$$

where the asterisks have been dropped for convenience. Close examination of the above set of equations reveals that no similarity variable exists since the boundary conditions are not compatible with any one-parameter groups of transformations which render the differential equations conformally invariant because the initial bubble radius is nonzero. It thus becomes necessary to solve a nonlinear partial differential equation rather than a linear ordinary differential equation as is the case for growth from zero size. Since the parameters N_a and N_b are usually small for the isothermal growth or collapse of gas bubbles at atmospheric pressure, we generate a series solution for this important case by formulating a parameter perturbation technique using the above two dimensionless groups as the perturbation parameters.

Formulation of Perturbation Method

We proceed by immobilizing the moving boundary and utilizing a volume perturbation scheme (6). Substitution of the new variables

$$\xi = \frac{r}{R} \tag{22}$$

$$C = \rho_I \xi \tag{23}$$

into Equations (14) to (19) yields the following nonlinear system of equations:

$$\begin{aligned} &\frac{\partial C}{\partial t} + \left[2N_a \int_0^t \left(\frac{\partial C}{\partial \xi} \right)_{\xi=1} dt' \right] \left(\frac{\partial C}{\partial t} \right) \\ &- 2N_a t \frac{\partial C}{\partial t} + (N_a - N_b) \left[\frac{1}{\xi^2} \left(\frac{\partial C}{\partial \xi} \right) \left(\frac{\partial C}{\partial \xi} \right)_{\xi=1} - \frac{C}{\xi^3} \left(\frac{\partial C}{\partial \xi} \right)_{\xi=1} - \frac{1}{\xi^2} \frac{\partial C}{\partial \xi} + \frac{C}{\xi^3} \right] \\ &- N_a \left[\xi \left(\frac{\partial C}{\partial \xi} \right) \left(\frac{\partial C}{\partial \xi} \right)_{\xi=1} - C \left(\frac{\partial C}{\partial \xi} \right)_{\xi=1} - \xi \frac{\partial C}{\partial \xi} + C \right] = \frac{\partial^2 C}{\partial \xi^2} \end{aligned} \tag{24}$$

$$C(\xi, 0) = 0 \tag{25}$$

$$C(\infty, t) = 0 \tag{26}$$

$$C(1, t) = 1 \tag{27}$$

$$R^2 = 1 + 2N_a \int_0^t \left(\frac{\partial C}{\partial \xi} \right)_{\xi=1} dt' - 2N_a t \tag{28}$$

We seek a solution in the form of a series in ascending powers of the perturbation parameters and thus assume the existence of the following double perturbation series:

$$C = C_0 + N_a C_1 + N_b C_2 + N_a N_b C_3 + N_a^2 C_4 + N_b^2 C_5 + \ldots \tag{29}$$

Introduction of Equation (29) into Equations (24) to (27) produces in the usual manner (1) the following differential equations and boundary conditions for the zero-order and first-order terms of the perturbation series:

$$\frac{\partial C_0}{\partial t} = \frac{\partial^2 C_0}{\partial \xi^2} \tag{30}$$

$$C_0(\xi, 0) = C_0(\infty, t) = 0 \tag{31}$$

$$C_0(1, t) = 1 \tag{32}$$

$$\frac{\partial C_1}{\partial t} + 2\left[\int_0^t \left(\frac{\partial C_0}{\partial \xi} \right)_{\xi=1} dt' \right] \left(\frac{\partial C_0}{\partial t} \right)$$

$$-2t\frac{\partial C_0}{\partial t}-\frac{1}{\xi^2}\frac{\partial C_0}{\partial \xi}+\frac{C_0}{\xi^3}+\xi\frac{\partial C_0}{\partial \xi}-C_0$$

$$+\left[\frac{1}{\xi^2}\frac{\partial C_0}{\partial \xi}-\frac{C_0}{\xi^3}-\xi\frac{\partial C_0}{\partial \xi}+C_0\right]\left(\frac{\partial C_0}{\partial \xi}\right)_{\xi=1}=\frac{\partial^2 C_1}{\partial \xi^2} \quad (33)$$

$$C_1(\xi,0)=C_1(\infty,t)=C_1(1,t)=0 \quad (34)$$

$$\frac{\partial C_2}{\partial t}-\frac{1}{\xi^2}\left(\frac{\partial C_0}{\partial \xi}\right)\left(\frac{\partial C_0}{\partial \xi}\right)_{\xi=1}+\frac{C_0}{\xi^3}\left(\frac{\partial C_0}{\partial \xi}\right)_{\xi=1}+\frac{1}{\xi^2}\frac{\partial C_0}{\partial \xi}-\frac{C_0}{\xi^3}=\frac{\partial^2 C_2}{\partial \xi^2} \quad (35)$$

$$C_2(\xi,0)=C_2(\infty,t)=C_2(1,t)=0 \quad (36)$$

From Equations (30) to (32) the zero-order solution is simply

$$C_0=\text{erfc}\left(\frac{\xi-1}{2\sqrt{t}}\right) \quad (37)$$

and, therefore, Equations (33) and (35) become

$$\frac{\partial C_1}{\partial t}+\left[\frac{1}{\pi t\,\xi^2}-\frac{2(\xi-1)}{\pi t}-\frac{(\xi-1)}{\sqrt{\pi t}}+\frac{1}{\xi^2\sqrt{\pi t}}-\frac{\xi}{\pi t}-\frac{\xi}{\sqrt{\pi t}}\right]e^{-\frac{(\xi-1)^2}{4t}}$$

$$+\left[\frac{1}{\xi^3\sqrt{\pi t}}+\frac{1}{\xi^3}-\frac{1}{\sqrt{\pi t}}-1\right]\text{erfc}\left(\frac{\xi-1}{2\sqrt{t}}\right)=\frac{\partial^2 C_1}{\partial \xi^2} \quad (38)$$

$$\frac{\partial C_2}{\partial t}-\left[\frac{1}{\xi^2\sqrt{\pi t}}+\frac{1}{\xi^2\,\pi t}\right]e^{-\frac{(\xi-1)^2}{4t}}-\left[\frac{1}{\xi^3}+\frac{1}{\xi^3\sqrt{\pi t}}\right]\text{erfc}\left(\frac{\xi-1}{2\sqrt{t}}\right)=\frac{\partial^2 C_2}{\partial \xi^2} \quad (39)$$

Equations (34) and (38) can be solved by standard linear methods yielding the equation

$$C_1(\xi,t)=\left[2(\xi-2)\sqrt{\frac{t}{\pi}}+\frac{2\xi}{\pi}\right]e^{-\frac{(\xi-1)^2}{4t}}+\left[2\sqrt{\frac{t}{\pi}}-\frac{2}{\pi}+\frac{5}{2}(\xi-1)+\frac{1}{2\xi}-\frac{\xi^2}{2}\right]\text{erfc}\left(\frac{\xi-1}{2\sqrt{t}}\right)-I_0(\xi,t) \quad (40)$$

where

$$I_0(\xi,t)=\frac{1}{\pi}\int_0^\infty\int_0^t\frac{\exp\left[-\frac{(\xi-1)^2+n^2}{4(t-\lambda)}\right]\sinh\left[\frac{(\xi-1)n}{2(t-\lambda)}\right]}{(1+n)^2\,(t-\lambda)^{1/2}\,\lambda^{1/2}}\times\left[\frac{e^{-\frac{n^2}{4\lambda}}}{\sqrt{\pi\lambda}}+\frac{\text{erfc}\left(\frac{n}{2\sqrt{\lambda}}\right)}{(1+n)}\right]d\lambda\,dn \quad (41)$$

Similarly, the solution to Equations (36) and (39) can be shown to be

$$C_2(\xi,t)=I_0(\xi,t)+\frac{(\xi-1)}{2\xi}\,\text{erfc}\left(\frac{\xi-1}{2\sqrt{t}}\right) \quad (42)$$

and, consequently, Equations (29), (37), (40), and (42) constitute a first-order series approximation to the concentration field in the liquid surrounding the bubble. Finally, from Equations (28), (29), (37), (40), and (42) we derive the following result for the bubble radius:

$$R^2=1-2N_a\left(t+2\sqrt{\frac{t}{\pi}}\right)+N_a^2\left[\frac{8t^{3/2}}{3\pi^{1/2}}+2t+\frac{8t^{1/2}}{\pi^{3/2}}-2I_1(t)\right]+N_aN_b\,[t+2I_1(t)]+\ldots \quad (43)$$

$$I_1(t)=\int_0^t\int_0^\infty\frac{\text{erfc}\left[\frac{n}{2(t-\lambda)^{1/2}}\right]}{(1+n)^2\sqrt{\pi\lambda}}\times\left[\frac{e^{-\frac{n^2}{4\lambda}}}{\sqrt{\pi\lambda}}+\frac{\text{erfc}\left(\frac{n}{2\sqrt{\lambda}}\right)}{(1+n)}\right]dn\,d\lambda \quad (44)$$

Results entirely equivalent to those given above can also be derived by a surface-volume perturbation scheme (6) which involves writing a series expansion for the boundary motion. Equation (43) provides explicit results for a second-order approximation to the bubble size. Higher-order approximations can of course be derived by computing the higher-order functions of the perturbation series for the mass density, but the labor involved rapidly becomes excessive. Calculation of the boundary position from Equation (43) is very simple if the functional form of the integral, $I_1(t)$, is known. This integral has been evaluated numerically and its functional dependence is depicted in Figures 1 and 2. Numerical integration of

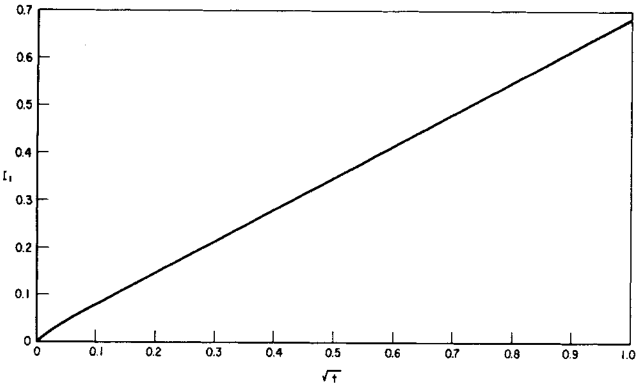

Fig. 1. Functional dependence of I_1 (t).

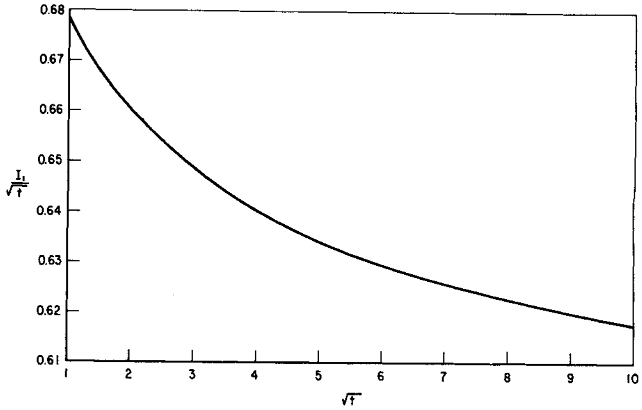

Fig. 2. Functional dependence of I_1 (t).

$I_1(t)$ is facilitated if Equation (44) is rewritten as

$$I_1(t) = 2\sqrt{\frac{t}{\pi}} - \sqrt{t}\int_0^1\int_0^\infty \frac{\text{erfc}\left(\frac{n}{2\sqrt{t\lambda}}\right)\text{erfc}\left[\frac{n}{2t^{1/2}(1-\lambda)^{1/2}}\right]}{\sqrt{\pi}\,(1+n)^3\,(1-\lambda)^{1/2}}\,dn\,d\lambda - \int_0^1\int_0^\infty \frac{e^{-\frac{n^2}{4\lambda t}}\,\text{erfc}\left[\frac{n}{2t^{1/2}(1-\lambda)^{1/2}}\right]}{(1+n)^2\,\pi\,\lambda^{1/2}(1-\lambda)^{1/2}}\,dn\,d\lambda \quad (45)$$

In addition, it can be shown from Equation (45) that

$$\lim_{t\to\infty}\left(\frac{I_1}{\sqrt{t}}\right) = \frac{1}{\sqrt{\pi}} \quad (46)$$

EVALUATION OF PERTURBATION SOLUTION

The perturbation series solution presented in the previous section is now evaluated as to its convergence properties and its accuracy in portraying the physical situation. In addition, we compare the present solution with previous results.

Comparison with Previous Approximate Solutions

As mentioned above, there exist two types of approximate analytical solutions to the bubble dissolution problem, the quasi-steady state approximation and the quasi-stationary approximation. Since there has been no attempt in this work to investigate the asymptotic nature of the former, it is not possible to comment precisely on its range of applicability and any advantages it possesses, if any, over the results of this investigation. However, since the bubble radius-time relationship derived in this study is as simple as the quasi-steady state result, it would appear that there is no advantage in using the more approximate equation from the quasi-steady state theory.

Equation (37) and the first-order terms of Equation (43) essentially constitute the quasi-stationary approximation of this investigation, and it is clearly evident that the form of the radius-time relationship is considerably simpler than previous quasi-stationary approximations (*7, 8, 11*). The complexity of the previous equations results essentially from not recognizing that the equation for the bubble radius is an infinite series and, hence, not disregarding incomplete higher-order terms. In summary, then, the complete bubble radius equation of this study is superior in several aspects when compared to the existing approximate solutions based on the quasi-stationary assumption. First of all, it is in a much simpler form, which makes it considerably easier to compute theoretical bubble dissolution rates or to extract diffusion coefficients from experimental dissolution data. Furthermore, the new equation provides more accurate results for a given set of conditions since it contains additional terms, and its range of applicability is hence greater. Finally, the new results are based on a mathematical formalism of successive approximations rather than a mathematically vague procedure of neglecting terms.

Convergence Properties

It is of course desirable from both purely mathematical and practical points of view to investigate the convergence properties of the series expansions derived above. Ideally, it would be of value to delineate the range of the perturbation parameters for which the above infinite asymptotic perturbation series converge and to be able to determine how many terms of the series are necessary to obtain a desired degree of accuracy for given values of the perturbation parameters. However, mathematical convergence depends on possessing a knowledge of the behavior of terms of indefinitely high order, which of course are not available in the present instance. Hence, of necessity, we concentrate on determining over what range of the perturbation parameters the above truncated series describe the true solution to within a given degree of accuracy. The accuracy aspect of the perturbation results is discussed in the following section.

In any physical problem, the coefficients of the perturbation expansion are functions of the space and time variables, and a perturbation series is said to be uniformly valid in these variables only if the error is uniformly small (*15*). Nonuniformities in the perturbation solution arise in singular perturbation problems, in direct coordinate perturbation schemes, and, in general, any time when one of the operations performed on the perturbation expansion is not justified. Detailed examination of Equation (43) indicates that the series expansion for the bubble radius apparently diverges for large but finite values of time. Although the reason for such a nonuniformity is not clear from an analysis of the volume perturbation scheme, inspection of the formalism of the surface-volume technique mentioned above strongly suggests that this series divergence is injected into the development by the operation of dividing by the perturbation series for R. If this conjecture is indeed true, then the results of this investigation are valid only for any combinations of the values of time and of the perturbation parameters for which $0 < R < 2$.

Such a limitation basically presents no difficulty for bubble dissolution because the entire dissolution process is of course within this domain. However, one can expect that, for set values of the perturbation parameters, more terms will be needed for determining the bubble radius with equivalent accuracy from the series solution at the longer values of time since slower convergence can be expected towards the end of the interval of convergence. In addition, utilization of Equation (43) for bubble growth is limited to the early stages, when $R < 2$. However, since there exists an asymptotic solution for the later stages, it is possible to describe completely bubble growth from finite initial size by matching the two solutions.

Utilization of a similarity transformation gives as the solution to Equations (14) to (18) for growth from zero size

$$\rho_I = \frac{I_2(\eta)}{I_2(\beta)} \quad (47)$$

$$I_2(\eta) = \int_\eta^\infty \frac{\exp\left[-\lambda^2 - \frac{2\beta^3}{\lambda}\left(1-\frac{N_b}{N_a}\right)\right]}{\lambda^2}\,d\lambda \quad (48)$$

where the growth constant, β, can be evaluated from

$$N_a = -2\beta^3 \exp\left[\beta^2 + 2\beta^2\left(1-\frac{N_b}{N_a}\right)\right] I_2(\beta) \quad (49)$$

and where

$$\eta = \frac{r}{2\sqrt{t}} \quad (50)$$

$$R = 2\beta\sqrt{t} \quad (51)$$

Accuracy of Derived Solution

It appears that the best method, at the present time, for evaluating the accuracy of perturbation solutions is to compare them with finite-difference solutions of the original equations at several well chosen values of the perturbation parameters. Finite-difference solutions of nonlinear partial

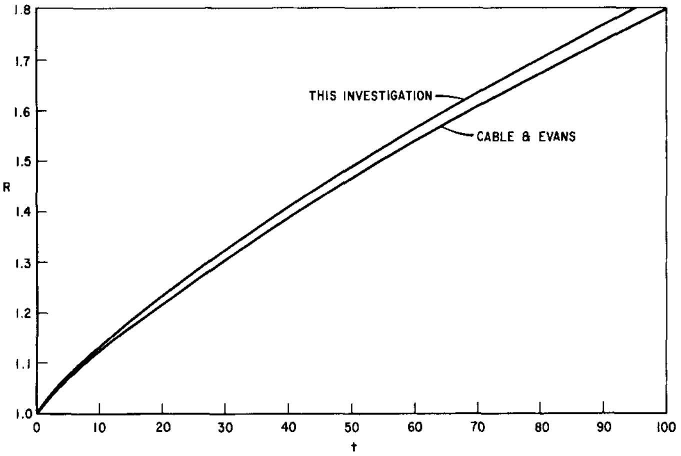

Fig. 3. Comparison of finite-difference solutions for $N_a = -.01$, $N_b = 0$.

differential equations can provide a good basis for assessing the applicability of truncated perturbation series since numerical solutions can be very accurate if proper precautions are taken. However, finite-difference solutions are really not convenient replacements for perturbation series solutions because they are not as compact and because they must be repeated for each new set of conditions whereas series solutions are immediately applicable for any values of the perturbation parameters within the range of validity. In this investigation, we do not conduct an extensive investigation to map the region of the perturbation parameters, N_a and N_b, in which the truncated series expansion, Equation (43), will predict the bubble radius with an error less than some desired error level. We prefer to concentrate on providing a valid finite-difference method for carrying out such a program and presenting a few examples of the type of accuracy that can be expected from the perturbation solution.

Cable and Evans (*4*) have conducted the most extensive investigation of formulating finite-difference solutions to the equations governing bubble growth and bubble dissolution. They utilized explicit finite-difference techniques, did not immobilize the moving boundary, and set up a finite-difference grid for an infinite region. Attempts to generate finite-difference solutions using this type of approach gave low growth and dissolution rates since it was difficult to represent the concentration gradient at the surface of the sphere very accurately. Cable and Evans also experienced this difficulty, especially at the higher growth and dissolution rates. To provide a more accurate representation of the surface concentration gradient, it was necessary to map the infinite region using a suitable transformation into a region of finite extent. Also, the method of solution is somewhat simpler if the moving boundary is immobilized and implicit finite-difference methods are used.

For the case of bubble growth from finite initial size, utilization of the transformation

$$\theta = 1 - \exp\left[-\alpha\left(\frac{r}{R} - 1\right)\right] \tag{52}$$

converts Equations (14) to (18) to

$$\frac{R^2}{\alpha(1-\theta)}\frac{\partial \rho_I}{\partial t} - \frac{\partial \rho_I}{\partial \theta}\frac{dR^2}{dt}\left[\frac{1-\frac{\ln(1-\theta)}{\alpha}}{2} - \frac{1-\frac{N_b}{N_a}}{2\left[1-\frac{\ln(1-\theta)}{\alpha}\right]^2}\right]$$

$$= \alpha(1-\theta)\frac{\partial^2 \rho_I}{\partial \theta^2} - \alpha\frac{\partial \rho_I}{\partial \theta} + \frac{2}{\left[1-\frac{\ln(1-\theta)}{\alpha}\right]}\left(\frac{\partial \rho_I}{\partial \theta}\right) \tag{53}$$

$$\frac{dR^2}{dt} = 2N_a\,\alpha\left(\frac{\partial \rho_I}{\partial \theta}\right)_{\theta=0} \tag{54}$$

$$\rho_I(\theta, 0) = 0 \tag{55}$$

$$\rho_I(1, t) = 0 \tag{56}$$

$$\rho_I(0, t) = 1 \tag{57}$$

The parameter, α, is a constant which allows some flexibility in the coordinate transformation. For bubble dissolution, a more suitable mapping is

$$\psi = 1 - \exp\left[-\alpha(r - R)\right] \tag{58}$$

and under this change of variable Equations (14) to (18) become

$$\frac{1}{\alpha(1-\psi)}\frac{\partial \rho_I}{\partial t} + \left[\frac{R^2\left(1-\frac{N_b}{N_a}\right)}{\left[R-\frac{\ln(1-\psi)}{\alpha}\right]^2} - 1\right]\frac{dR}{dt}\frac{\partial \rho_I}{\partial \psi}$$

$$= \alpha(1-\psi)\frac{\partial^2 \rho_I}{\partial \psi^2} - \alpha\frac{\partial \rho_I}{\partial \psi} + \frac{2}{R-\frac{\ln(1-\psi)}{\alpha}}\left(\frac{\partial \rho_I}{\partial \psi}\right) \tag{59}$$

$$\frac{dR}{dt} = \alpha\,N_a\left(\frac{\partial \rho_I}{\partial \psi}\right)_{\psi=0} \tag{60}$$

$$\rho_I(\psi, 0) = 0 \tag{61}$$

$$\rho_I(1, t) = 0 \tag{62}$$

$$\rho_I(0, t) = 1 \tag{63}$$

The application of implicit finite-difference techniques (*1*) to these equations is straightforward and details are omitted. Convergence of the finite-difference solutions to the solutions of the partial differential equations was established in the usual manner by varying the mesh sizes for the radial and time variables.

A comparison of the finite-difference methods of this investigation with those of Cable and Evans is presented in Figures 3 and 4 for typical cases of bubble growth and dissolution. It is clear that the results of Cable and Evans predict slower growth and dissolution rates, particularly in

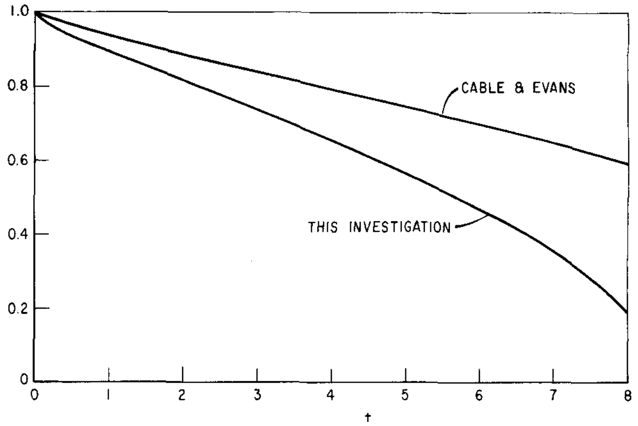

Fig. 4. Comparison of finite-difference solutions for $N_a = .05$, $N_b = 0$.

the latter case. These slower rates are due presumably to an underestimation of the concentration gradients at the bubble surface. Consequently, the conclusions of these authors as to the applicability of the approximate analytical solutions are in doubt. The radius-time curves numerically generated in this investigation for bubble growth and those computed from the exact solution for growth from zero size are essentially indistinguishable at long times.

A comparison of perturbation and finite-difference solutions is presented in Table 1 for bubble dissolution with $N_a = 0.2$ and $N_b = 0$ and for bubble growth with $N_a = -0.2$ and $N_b = 0$. The finite-difference solutions are considered very accurate and any deviations between the numerical results and the perturbation solutions are presumably due to inadequacies in the approximate asymptotic perturbation series. Comparable or lesser errors were observed for the bubble radius for perturbation parameters anywhere in the interval —0.2 to 0.2. The second-order perturbation series is of course more accurate than the first-order result, and both approximations yield less accurate values for the bubble radius at the longer times because convergence of a series is slower towards the ends of the interval of convergence. Previous quasi-stationary approximations, such as the equation of Krieger, Mulholland, and Dickey (*11*), agree quite closely numerically with the first-order radius-time approximation of this study. Quasi-steady state results are in considerable disagreement with the numerical results of the present investigation.

The new results of this investigation obviously extend the accuracy and range of validity of previous work so that more confidence can be placed in results at a given set of conditions and errors less than a desired level can be obtained for a greater range of the perturbation parameters. However, even at very small values of the perturbation parameters, the truncated series expansion for the radius, Equation (43), will yield significant errors at long times as, of course, will any of the previous quasi-stationary approximations. Unless additional terms are added to the perturbation series, one cannot expect to obtain the same accuracy at longer times that is readily obtainable at the shorter times for the same values of the perturbation parameters. Hence, when dealing with series solutions for bubble growth and dissolution, it is necessary to consider the time interval as well as the values of the perturbation parameters when trying to determine under what conditions a truncated series will give a desired degree of accuracy.

TABLE 1. COMPARISON OF PERTURBATION AND FINITE-DIFFERENCE SOLUTIONS

Bubble Dissolution: $N_a = 0.2$, $N_b = 0$

		R^2	
t	Numerical solution	Second-order approximation	First-order approximation
0.011	0.9497	0.9493	0.9483
0.038	0.9008	0.8997	0.8968
0.125	0.8038	0.8036	0.7904
0.250	0.7028	0.7029	0.6743
0.400	0.6029	0.6032	0.5545
0.575	0.5018	0.5020	0.4277
0.770	0.4015	0.4008	0.2959
0.985	0.3018	0.2987	0.1580
1.225	0.2016	0.1937	0.0104
1.500	0.1002	0.0822	0.1528

Bubble Growth: $N_a = -0.2$, $N_b = 0$

		R^2	
t	Numerical solution	Second-order approximation	First-order approximation
0.15	1.250	1.254	1.235
0.41	1.503	1.503	1.453
1.01	2.001	2.003	1.858
1.66	2.503	2.512	2.246
2.33	3.005	3.028	2.621
3.01	3.506	3.550	2.987

NOTATION

C = dependent variable defined by Equation (23)
C_i = ith perturbation function for C
D = binary diffusion coefficient
I_0 = integral defined by Equation (41)
I_1 = integral defined by Equation (44)
I_2 = integral defined by Equation (48)
N_a = dimensionless parameter defined by Equation (20)
N_b = dimensionless parameter defined by Equation (21)
R = radius of bubble
R_o = initial radius of bubble
r = radial position variable
t = time
$\hat{V}_I$ = partial specific volume of component I
$\bar{v}$ = volume average velocity
α = constant used in Equations (52) and (58)
β = growth constant defined by Equation (49)
η = independent variable defined by Equation (50)
θ = independent variable defined by Equation (52)
ξ = independent variable defined by Equation (22)
ρ_I = mass density of component I
ρ_{IE} = equilibrium concentration of component I at phase interface
ρ_{IO} = initial concentration of component I in liquid phase
ρ = density of liquid phase
$\hat{\rho}$ = density of fluid phase
ψ = independent variable defined by Equation (58)

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