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# An accurate numerical solution for mass diffusion-induced bubble growth in viscous liquids containing limited dissolved gas

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**Abstract**—A mathematical formulation based on potential theory is used to develop an accurate numerical technique to calculate the mass diffusion-induced growth of a spherical gas bubble surrounded by a viscous Newtonian liquid with a limited dissolved gas concentration under isothermal conditions. We solve for the complete concentration profile of the dissolved gas in the liquid enveloping the bubble to predict the bubble growth. These results are contrasted against the dynamics of bubble growth predicted by the widely used approach of employing polynomial profiles to describe the gas concentration. The influence of key dimensionless parameters on the differences in the bubble growth due to the choice of concentration profile is examined. The investigation reveals that the commonly used approximate solutions for the concentration profile not only underpredict the growth rate at the early stages of the growth but also overestimate the steady-state bubble radius due to the implicit assumption of an unlimited supply of dissolved gas in the liquid.

## 1. INTRODUCTION

THE BUBBLE growth and dissolution in liquids plays a key role in diverse fields such as boiling and vaporization, polymer processing and glass refining. The growth process is in general complicated, involving simultaneous mass, momentum and energy transfer between the expanding bubble and the fluid surrounding it. For example, in the case of mass diffusion-induced growth of a gas bubble, the movement of the gas–liquid interface depends on the gas pressure inside the bubble through the momentum transfer. On the other hand, the gas pressure and the bubble radius are related to each other and to the rate of gas diffusion through the conservation of mass. Also, the rate of gas diffusion itself depends on the movement of the interface through the diffusion equation. Hence the three equations, namely the momentum, the mass balance and the diffusion, governing the mass diffusion-induced growth of a gas bubble in a pool of liquid containing a dissolved gas are highly coupled and nonlinear. Due to these complexities, there is no known analytical solution to predict the bubble growth under general conditions.

In the past, analytical and numerical methods have been developed to predict bubble growth in liquids to model specific cases. The governing equations describing the spherically symmetric phase growth controlled by the transport of mass and heat were presented by Scriven [1]. He obtained an analytical solution for a bubble growing in an infinite pool of liquid using a similarity solution. The similarity solution is valid for a bubble growing from an initial zero radius. Due to the presence of a geometric length scale, namely the initial bubble radius, it is not possible to obtain similarity solutions to the diffusion equation for a bubble growing from a nonzero radius. Cable and Frade [2] extended the similarity solution to the cases of multicomponent bubbles, in which more than one gas diffuses into the bubble at the same time. They observed that the bubbles growing from a finite initial size always approached an asymptotic regime having constant bubble composition and that there was a linear relation between the radius and the square root of time which enabled them to define a similarity variable and obtain an analytical solution for the asymptotic regime.

Among the attempts made to obtain approximate solutions to the problem of the diffusion-induced growth of a gas bubble, Barlow and Langlois [3], in a pioneering paper, studied the diffusion-induced growth of a gas bubble in a viscous liquid. They considered both transfer of mass and momentum and simplified the diffusion equation by restricting the concentrating gradients to a thin boundary layer surrounding the bubble. Outside the boundary layer, the gas concentration was assumed to be undisturbed and equal to the initial concentration. A similar assumption was also used by Plesset and Zwick [4] in their study of thermal diffusion into a vapor bubble within a volume of superheated water. The above assumption allowed them to obtain an analytical solution for the early stages of the growth using the Laplace transform. Rosner and Epstein [5] used the moment integral method to predict the diffusion-induced growth of a bubble in highly supersaturated liquids. The use of

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### NOMENCLATURE

| | |
|---|---|
| $c$ | concentration of dissolved gas |
| $\acute{c}$ | gas concentration minus initial gas concentration, $c-c_0$ |
| $D$ | diffusion coefficient |
| $k_h$ | Henry's law constant |
| $P$ | fluid pressure |
| $P_a$ | atmospheric pressure |
| $P_f$ | applied pressure |
| $P_g$ | gas pressure |
| $r$ | radial coordinate |
| $R$ | bubble radius |
| $\dot{R}$ | derivative of $R$ with respect to time |
| $\ddot{R}$ | second derivative of $R$ with respect to time |
| $S$ | outer radius of shell |
| $t$ | time |
| $t_{ref}$ | reference time, $\mu/P_a$ |
| $v_r$ | fluid velocity in radial direction |
| $y$ | Lagrangian coordinate, $r^3-R^3$. |

Greek symbols

| | |
|---|---|
| $\beta$ | clustering parameter |
| $\lambda_1, \lambda_2$ | eigenvalues |
| $\mu$ | fluid viscosity |
| $\rho$ | fluid density |
| $\rho_g$ | gas density |
| $\sigma$ | surface tension |
| $\tau_{rr}$, $\tau_{\theta\theta}$, $\tau_{\phi\phi}$ | normal stresses |
| $\Phi$ | potential function |
| $\Psi$ | ratio of bubble volume to total volume of bubble and fluid. |

Subscripts

| | |
|---|---|
| e | equilibrium conditions |
| 0 | initial conditions. |

Superscript

| | |
|---|---|
| * | dimensionless quantities. |

the integral method to obtain approximate solutions to partial differential equations dates back to von Karman and Pohlhausen, who applied the method to the solution of boundary layer equations [6]. To solve the diffusion equation with the integral method, Rosner and Epstein assumed a polynomial to describe the concentration profile inside the boundary layer surrounding the bubble. Using different orders of polynomial profiles, several researchers have adopted this method to predict the diffusion-induced growth or collapse of a gas bubble in both viscous Newtonian and viscoelastic liquids [7–9]. Recently, Payvar [10] used a polynomial profile for the gas concentration to predict the mass transfer-controlled bubble growth during the rapid decompression of a liquid.

Using polynomial profiles for the gas concentration is tantamount to assuming that the bubble grows in an infinite pool of liquid, which happened in all the cases referred to above. In situations where a large number of bubbles nucleate and grow simultaneously in close proximity [11], the amount of liquid immediately surrounding a bubble is finite and so is the amount of the dissolved gas. Therefore, the hydrodynamics of the growth and the diffusion process will be different from the case of a bubble growing in an infinite medium. The underlying assumption in adopting a polynomial profile to describe the gas concentration is that at large distances from the bubble interface, the gas concentration remains unchanged and equal to the initial concentration. This assumption is not valid for a bubble surrounded by a finite amount of liquid with a limited concentration of dissolved gas and cannot predict the steady-state bubble radius correctly. Moreover, under such circumstances, it is not possible to obtain a similarity solution to the diffusion equation even for a bubble growing from zero initial radius. This is due to the presence of yet another geometric length scale, namely the dimension of the finite body of the medium surrounding the bubble. Therefore, it is necessary to solve the diffusion equation in its complete form numerically as opposed to using similarity solutions or approximate analytical methods.

In this paper, we analyze the process of mass diffusion-induced growth of a gas bubble surrounded by a limited amount of liquid and dissolved gas and present a solution technique to accurately solve for the concentration profile of the dissolved gas. Bubble growth dynamics are predicted with this concentration profile. The results are compared with the predictions obtained by using polynomial profiles for the gas concentration. This enables one to establish the range of validity of the approximate solutions and investigate the errors which may result from the use of such solutions. This analysis is restricted to isothermal cases. The energy transfer, however, may affect the bubble growth in two ways. It may induce mass transfer due to the phase change (boiling) or it may affect the physical and transport properties of the gas–liquid system which are normally temperature dependent. The latter reason is of significance in viscous liquids such as polymer melts. Thus, the assumption of isothermal conditions will only affect the magnitude of the properties in the cases considered in this study and it is not expected to change the qualitative trend of the results and the conclusions of this analysis.

## 2. MATHEMATICAL FORMULATION

To analyze the growth of a bubble in a limited pool of liquid, we consider a spherical gas bubble surrounded concentrically by a hypothetical shell of liquid with finite thickness and constant mass. The assumption of the liquid shell around the bubble significantly simplifies the analysis due to the spherical symmetry. Figure 1 shows a schematic of the bubble and the liquid shell. In this figure, $R(t)$ is the bubble radius and $S(t)$ is the outer radius of the liquid shell. It is assumed that the bubble growth occurs under isothermal conditions, ensuring that the volume of the liquid in the shell remains constant. The continuity equation for the purely extensional flow of an incompressible fluid around the bubble yields the velocity distribution in the liquid:

$$v_r = \frac{\dot{R}R^2}{r^2} \tag{1}$$

where $v_r$ is the fluid velocity in the radial direction and $\dot{R}$ is the rate of bubble growth. The conservation of momentum for the fluid in the radial direction in terms of stresses is given as

$$\rho\left(\frac{\partial v_r}{\partial t}+v_r\frac{\partial v_r}{\partial r}\right) = -\frac{\partial P}{\partial r}+\frac{1}{r^2}\frac{\partial}{\partial r}(r^2\tau_{rr})-\frac{\tau_{\theta\theta}+\tau_{\phi\phi}}{r} \tag{2}$$

where $\rho$ is the density and $P$ is the pressure in the fluid. $\tau_{rr}$, $\tau_{\theta\theta}$ and $\tau_{\phi\phi}$ represent the stresses in the fluid and $\tau_{\theta\theta}$ is equal to $\tau_{\phi\phi}$ due to the spherical symmetry. To relate the stresses within the fluid to the gas pressure inside the bubble and to the applied pressure at the outer boundary of the shell, equation (2) is integrated along the radial direction from the bubble interface to the outer boundary of the shell. Use of the condition of stress continuity at the interface results in

$$\rho(\tfrac{1}{2}(\Psi^{4/3}-1)R\dot{R}^2-(\Psi^{1/3}-1)(R^2\ddot{R}+2R\dot{R}^2)) = (P_g-P_f)R-2\sigma+4\mu(\Psi-1)\dot{R} \tag{3}$$

where $\Psi$ is defined as $R^3/(V+R^3)$ and $V$, which is equal to $S^3-R^3$, is proportional to the volume of the liquid in the shell. $\ddot{R}$ is the second derivative of radius with respect to time, $P_g$ is the gas pressure inside the bubble, $P_f$ is the applied pressure outside the shell, $\sigma$ is the surface tension and $\mu$ is the fluid viscosity. For large values of $V$, $\Psi$ tends to zero and equation (3) reduces to the governing equation for bubble growth in an infinite medium. However, for small values of $V$, of the order of $R^3$ or less (finite shell thickness), the bubble growth rate will be higher due to a nonzero $\Psi$. The assumption of bubble growth in an infinite medium is not acceptable in such cases as it also implies that an unlimited amount of dissolved gas is available for diffusion into the bubble. This is clearly not the correct physics when the body of liquid is limited.

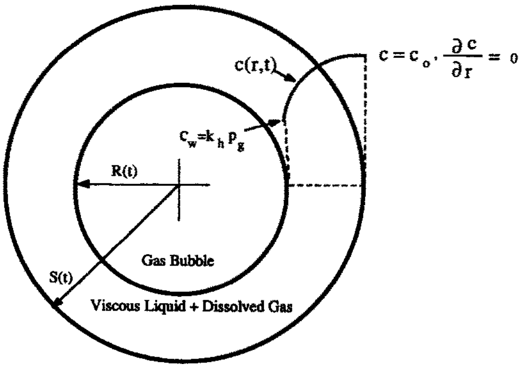

FIG. 1. Schematic diagram of a bubble surrounded by a liquid shell.

### 2.1. *Mass diffusion*

The mass balance for the gas inside the bubble can be expressed as

$$\frac{\mathrm{d}}{\mathrm{d}t}(\rho_g R^3) = 3\rho D R^2\left(\frac{\partial c}{\partial r}\right)_{r=R} \tag{4}$$

where $\rho_g$ is the density of the gas inside the bubble, $D$ is the diffusion coefficient and $(\partial c/\partial r)_{r=R}$ is the concentration gradient of the dissolved gas at the bubble interface. The left-hand side of equation (4) is the rate of accumulation of mass inside the bubble and the right-hand side is the rate of diffusion of gas from the liquid into the bubble. Due to the low pressures involved it is assumed that the gas inside the bubble is an ideal gas and its pressure is related to the gas concentration at the interface through Henry's law

$$c_w = k_h P_g \tag{5}$$

where $c_w$ is the gas concentration at the interface and $k_h$ is Henry's law constant. For low gas concentrations, Henry's constant is a function of the temperature only [12] and, hence, its magnitude remains constant under isothermal conditions. Under non-isothermal conditions, its dependence on the temperature is of the Arrhenius equation form. However, the change in the magnitude of $k_h$ under non-isothermal conditions will not affect the qualitative features of the results presented in this study.

Initially, the gas concentration is assumed to be uniformly distributed throughout the liquid with magnitude equal to $c_0$. The bubble expansion depletes the gas and induces a concentration gradient in the liquid shell which initiates the diffusion process governed by the following equation:

$$\frac{\partial c}{\partial t}+v_r\frac{\partial c}{\partial r} = \frac{D}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial c}{\partial r}\right) \quad R(t) \leqslant r \leqslant S(t). \tag{6}$$

The initial and boundary conditions for this equation are presented in the following section.

### 2.2. *Concentration profile*

To solve for the concentration profile, equation (6) is transferred to the Lagrangian coordinate, given as

$$y = r^3 - R^3(t). \tag{7}$$

This transformation eliminates the convective term. The diffusion equation in the new coordinate system is

$$\frac{\partial \acute{c}}{\partial t} = 9D\frac{\partial}{\partial y}\left((y+R^3)^{4/3}\frac{\partial \acute{c}}{\partial y}\right) \tag{8}$$

where $\acute{c}$ is defined as $c-c_0$. The amount of gas in the liquid and the bubble is assumed to be constant because there is no diffusion of gas to or from the liquid at its outer boundary. Mathematically, this implies that the concentration gradient at the outer boundary of the liquid is zero at all times. Hence, the boundary conditions for equation (8) are as follows:

$$\acute{c}|_{y=0} = k_h(P_g - P_{g0}) \tag{9a}$$

$$\left.\frac{\partial \acute{c}}{\partial y}\right|_{y=S^3-R^3} = 0 \tag{9b}$$

and as the initial concentration is equal to $c_0$

$$c'|_{t=0} = 0. \tag{10}$$

At the early stage of growth and near the interface, the concentration gradient in the liquid is large, which makes it difficult to obtain an accurate numerical solution of equation (8). To avoid this difficulty, we introduce a vector field $\mathbf{\Omega}$ with the components $\acute{c}$ and $9D(y+R^3)^{4/3}(\partial\acute{c}/\partial y)$ in the $(y-t)$ plane. Using equation (8), it can be shown that such a vector field is irrotational. Therefore, there exists a potential function $\Phi(y, t)$ such that

$$\mathbf{\Omega} = \nabla\Phi. \tag{11}$$

Hence, equation (8) can be written in terms of the potential field as

$$\frac{\partial \Phi}{\partial t} = 9D(y+R^3)^{4/3}\frac{\partial^2 \Phi}{\partial y^2} \tag{12}$$

where $\acute{c}$ is equal to $\partial\Phi/\partial y$. Equation (12) is easier to solve for the variable $\Phi(y, t)$, which is the integral of gas concentration with respect to the $y$ coordinate. The initial and boundary conditions given by equations (9a), (9b) and (10) written in terms of the new dependent variable are

$$\Phi(y, 0) = 0 \tag{13a}$$

$$\Phi_y(y=0, t) = k_h(P_g - P_{g0}) \tag{13b}$$

$$\Phi_{yy}(y = S^3-R^3, t) = 0. \tag{13c}$$

Solution of equation (12) allows one to calculate the exact concentration profile in the liquid. Equations (3), (4) and (12) constitute the system of governing equations describing the growth dynamics of a bubble.

It should be pointed out that the commonly used approach is to obtain an approximate solution to the diffusion equation by assuming a polynomial profile for the gas concentration with the moment integral method. The underlying assumption in this approach is that the gas concentration outside the boundary layer is uniform and equal to the initial concentration ($c_0$) [3, 4]. This implies that all derivatives of the concentration are equal to zero outside the boundary layer, allowing formulation of an *n*th-order profile for the concentration. Using this approach, equation (6) is multiplied by $r^2$ and then integrated with respect to the radial coordinate inside the boundary layer. If one chooses to describe the concentration with a second-order profile and substitute it into the integrated diffusion equation, one can calculate the bubble growth using mass balance without solving the diffusion equation explicitly [13]

$$\frac{\mathrm{d}}{\mathrm{d}t}(\rho_g R^3) = \frac{6\rho^2 D(c_0-c_w)^2 R^4}{\rho_g R^3 - \rho_{g0}R_0^3}. \tag{14}$$

The same method with a third-order profile for the gas concentration results in the following expression:

$$\frac{\mathrm{d}}{\mathrm{d}t}(\rho_g R^3) = \frac{27\rho^2 D(c_0-c_w)^2 R^4}{4(\rho_g R^3 - \rho_{g0}R_0^3)}. \tag{15}$$

It is obvious that the choice of a polynomial profile for the gas concentration influences considerably the solution of the diffusion equation. Given that the latter solution determines the growth rate of the bubble through equation (3), it appears that the choice of polynomial has a considerable influence on the predicted bubble radius $R(t)$. This does not happen with the method used in this paper, which solves explicitly and without any assumption for the gas concentration profile.

## 3. DIMENSIONLESS FORM OF THE GOVERNING EQUATIONS

The governing equations are transformed into a dimensionless form by scaling the variables with respect to characteristic values. The initial bubble radius ($R_0$) is used as the length scale and ($S_0^3 - R_0^3$), which is proportional to the initial liquid volume around the bubble, is used to scale the Lagrangian coordinate $y$. Atmospheric pressure, $P_a$, is used as characteristic value for pressure and $t_{\mathrm{ref}}$, which is unknown a priori, is used to scale the time. This leads to the following dimensionless variables (denoted with a superscript $*$):

$$P_g^* = \frac{P_g}{P_a} \tag{16a}$$

$$P_f^* = \frac{P_f}{P_a} \tag{16b}$$

$$R^* = \frac{R}{R_0} \tag{16c}$$

$$t^* = \frac{t}{t_{\mathrm{ref}}} \tag{16d}$$

$$y^* = \frac{y}{S_0^3 - R_0^3} \tag{16e}$$

$$\xi^* = \frac{P_g R^3 - P_{g0}R_0^3}{P_{g0}R_0^3} \tag{16f}$$

$$\Phi^* = \frac{\Phi}{S_0^3 - R_0^3}. \tag{16g}$$

Equations (3), (4) and (12) can now be expressed in dimensionless form as follows:

$$C_1(1/2(\Psi^{4/3}-1)R^*\dot{R}^{*2} -(\Psi^{1/3}-1)(R^{*2}\ddot{R}^*+2R^*\dot{R}^{*2})) = (P_g^*-P_f^*)R^* -4\frac{C_2}{C_2+R^{*3}}\dot{R}^*-C_3 \quad (17)$$

$$\frac{\mathrm{d}(P_g^*R^{*3})}{\mathrm{d}t^*} = 9C_4\left(\frac{C_5}{C_2}\right)R^{*4}\left.\frac{\partial^2\Phi^*}{\partial y^{*2}}\right|_{y^*=0} \quad (18)$$

$$\frac{\partial\Phi^*}{\partial t^*} = 9C_4(1/C_2)^{2/3}(y^*+1/C_2R^{*3})^{4/3}\frac{\partial^2\Phi^*}{\partial y^{*2}} \quad (19)$$

with the following boundary conditions:

$$\Phi^*(y^*,0) = 0 \quad (20a)$$

$$\Phi^*_{y^*}(y^*=0,t^*) = C_6(P_g^*-P_{g0}^*) \quad (20b)$$

$$\Phi^*_{y^*y^*}(y^*=1,t^*) = 0. \quad (20c)$$

Dimensional analysis shows that $t_{\mathrm{ref}}$, which is related to the momentum transfer, is equal to $\mu/P_a$. There are six dimensionless parameters in the above equations ($C_i$, $i = 1$–$6$). $C_1$ is a Reynolds number equal to $\rho R_0^2/\mu t_{\mathrm{ref}}$. $C_2$ is equal to $(S_0/R_0)^3-1$ and expresses the ratio of the volume of the liquid to the initial bubble volume. $C_2$ tending to infinity implies that there is an infinite amount of liquid surrounding the bubble. $C_3$ is the dimensionless surface tension, equal to $2\sigma/R_0P_a$. $C_4$ is equal to $D\mu/R_0^2P_a$ and represents the ratio of the time scale for the momentum transfer to the time scale of the gas diffusion. $C_5$ is equal to $\rho(R_gT)/P_a$ and is the ratio of liquid to the gas density when the latter is evaluated at atmospheric pressure and $C_6$ is equal to $k_hP_a$, which is a dimensionless Henry's constant.

Equations (17)–(19) form the system of governing equations in dimensionless form for bubble growth that uses the exact concentration profile. The boundary conditions for these equations are

$$R^*(t^*=0) = 1 \quad (21a)$$

$$P_g^*(t^*=0) = P_{g0}^*. \quad (21b)$$

For the case of approximate solutions, where one assumes a polynomial profile for the gas concentration, equation (14) is written in dimensionless form as

$$\frac{\mathrm{d}\xi^*}{\mathrm{d}t^*} = \frac{C_4C_7}{\xi^*}(R^{*3}-\xi^*-1)^2\frac{1}{R^{*2}} \quad (22)$$

where $C_7$, which arises from nondimensionalization of equation (14), is equal to $6\rho^2k_h^2(R_gT)^2$ and expresses the gas concentration weighted with the ratio of the liquid density to the gas density at the atmospheric pressure. Equations (17) and (22) constitute the governing equations describing the bubble growth with a second-order profile for the gas concentration. If a third-order profile is used for the gas concentration, equation (22) will remain the same. However, parameter $C_7$ in this case will be equal to $27\rho^2k_h^2(R_gT)^2/4$.

## 4. NUMERICAL IMPLEMENTATION

The results presented in this study are confined to viscous fluids such as polymer melts. Polymeric melts may exhibit non-Newtonian and viscoelastic behavior under high deformation rates. However, the emphasis in this study in on the differences in the bubble growth predictions as a result of choosing different methods for solving the diffusion equation. Therefore, the effect of the non-Newtonian behavior of the fluid on the bubble growth was not considered in this paper. It has been shown elsewhere [14] that the fluid elasticity, in general, enhances the bubble growth at the early stages of the growth phenomenon. However, the bubble attains the same steady-state configuration eventually, suggesting that the fluid elasticity has a significant impact on the bubble growth only at the early stage of the process.

In the bubble growth in inviscid fluids, the viscous terms in the momentum equation are generally neglected compared to the inertia and surface tension terms. This is contrary to the bubble growth in highly viscous fluids where the viscous terms are dominant. To estimate the magnitude of the inertia terms in the momentum equation (equation (3)), we conducted an order of magnitude analysis. Using the equilibrium radius ($R_e$) and the initial gas pressure, each term in equation (3) was made dimensionless and of the order of one. The resulting Reynolds number ($\rho R_e^2P_{g0}/\mu^2$) was found to be much smaller than unity for viscous fluids, a fact which justified neglecting the inertia term.

The system of ordinary differential equations governing the bubble growth with the assumption of the polynomial profile for the gas concentration (equations (17) and (22)) are nonlinear and coupled, for which there is no known analytical solution. Therefore, a standard fourth-order Runge–Kutta method was used to solve them simultaneously. Equation (22) is singular at $t^* = 0$ and prompted the use of the following transformation:

$$\zeta^* = \xi^{*2}, \quad \zeta^*(0) = 0. \quad (23)$$

This transforms equation (22) to the following:

$$\frac{\mathrm{d}\zeta^*}{\mathrm{d}t^*} = 2\frac{C_4C_7(R^{*3}-\sqrt{\zeta^*}-1)^2}{R^{*2}}. \quad (24)$$

For the general case of the bubble growing in a limited body of liquid in which one needs to simultaneously evaluate the complete concentration profile, a combination of the Runge–Kutta method and an explicit finite difference scheme was employed to solve the coupled system of the governing equations (equations (17)–(19)). At the onset of the growth near the interface, concentration gradients are high; this poses numerical difficulties in solving the diffusion equation. Hence, we integrated the diffusion equation by using the new variable $\Phi$. Although $\Phi$ is not as steep as the concentration profile near the interface, it varies rapidly enough to warrant the use of a variable mesh with grid points clustered near the interface. We solve equation (19) on a uniformly spaced grid in the computational domain by employing the following transformation [15]:

$$\bar{y} = 1 - \frac{\ln[(\beta+1-y^*)/(\beta-1+y^*)]}{\ln[(\beta+1)/(\beta-1)]} \quad 1 < \beta < \infty \tag{25}$$

where $\bar{y}$ is the coordinate in the computational domain and $\beta$ is the clustering parameter. This transformation, which is used in boundary layer problems with large gradients near the wall, places more grid points near $y^* = 0$ as the clustering parameter approaches unity [15]. A variable clustering parameter is required in this case because with gas diffusion from the liquid into the bubble the concentration becomes less steep. Hence, to calculate the concentration gradient at the later stage of the growth process, a more uniform mesh is sufficient.

The calculation starts by making an initial guess for the concentration gradient at the interface. Using the initial guess, equations (17) and (18) are solved simultaneously to yield new values for the bubble radius and the gas pressure inside the bubble. Using the new radius and the gas pressure, equation (19) is solved and the concentration gradient at the interface is calculated. The calculated concentration gradient is then compared with the initial guess. If the error is less than a prespecified value, the variables are updated and the calculations are repeated for the next time step. Otherwise, the initial guess is revised and the iteration continues until convergence is achieved. Throughout this study, $10^{-5}$ was used as the relative convergence criterion.

## 5. STABILITY ANALYSIS

To investigate the stability of the coupled system of ordinary differential equations (equations (17) and (18)) about the singular points, we set the right-hand side of these equations equal to zero. The system has one equilibrium point at which $\partial^2\Phi^*/\partial y^{*2}$ is equal to zero and the bubble radius and pressure are equal to the equilibrium radius and pressure. The system of equations is nonlinear. Therefore, to investigate the stability, we linearized these equations about the equilibrium point. The eigenvalues of the resulting Jacobian matrix are given by

$$\lambda_1 = 0 \tag{26a}$$

$$\lambda_2 = -\left(\frac{3}{4C_2}R_e^{*3} + \frac{C_3}{2C_2}R_e^{*2} + \frac{C_3}{2R_e^*} + \frac{3}{4}\right) \tag{26b}$$

where $R_e^*$ is the dimensionless equilibrium radius. The eigenvalues are both real and less than or equal to zero, which indicates that the equilibrium point is a stable node [16].

We also investigated the stability of the finite difference scheme using the von Neumann method. In this approach, the error at any time $t = n\Delta t$ is assumed to be of the form

$$E_{j,n} = e^{\gamma(n\Delta t^*)}\, e^{i\beta(j\Delta y^*)} \tag{27}$$

where $e^{\gamma\Delta t^*}$ is the amplification factor. Using this error in equation (19) and simplifying the result, the following equation is obtained for the amplification factor:

$$e^{(\gamma\Delta t^*)} = 1 - 36C_4\left(\frac{1}{C_2}\right)^{2/3}\frac{\Delta t^*}{(\Delta y^*)^2} \times\left(y^* + \frac{R^{*3}}{C_2}\right)^{4/3}\sin^2(\beta\Delta y^*/2). \tag{28}$$

The error remains bounded if the absolute value of the amplification is less than unity. For the relation given by equation (28), this condition is satisfied if

$$18C_4\left(\frac{1}{C_2}\right)^{2/3}\frac{\Delta t^*}{(\Delta y^*)^2}\left(y^* + \frac{R^{*3}}{C_2}\right)^{4/3} \leqslant 1 \tag{29}$$

which establishes the stability criterion for obtaining the stable solution of the diffusion equation.

## 6. RESULTS AND DISCUSSION

Our objective is to obtain the bubble growth history by solving the full diffusion equation and explore the ranges under which the approximate solutions are valid. We would also like to estimate any errors caused by the approximate solutions as there are two main underlying assumptions made to derive them. It is assumed that the concentration can be described by a polynomial profile and the concentration at the outer shell boundary is equal to the initial concentration at all times, which translates into an unlimited amount of gas available for diffusion. These two assumptions in the derivation of the concentration profile contribute to the errors in the prediction of the bubble radius during the growth process, as shown in Figs. 2–9.

At the onset of the growth process and at small times, the assumption of a constant concentration at the outer boundary is reasonable as the percentage of depleted dissolved gas is small and hence the errors in the predictions are due to the selection of polynomials to approximate the concentration profiles. Later on, more and more dissolved gas which was initially in the liquid diffuses into the bubble and eventually one would expect the bubble to attain an equilibrium radius when all the gas has diffused into the bubble. The numerical solution of the diffusion equations does predict this behavior and the correct equilibrium radius, which can be verified by performing a simple mass balance. However, the approximate solutions use an imposed boundary condition of an initial concentration at the outer shell boundary and hence the bubble will continue to grow, supplied by an unlimited amount of gas at the outer boundary. Under these assumptions it may never arrive at an equilibrium radius.

To examine the conditions under which it may be acceptable to use approximate solutions, we conducted a parametric study. The parameters which have a significant effect on the concentration profile and hence the growth dynamics of the bubble are the amount of liquid in the shell around the bubble, the initial concentration of the dissolved gas or the initial gas pressure and the diffusion coefficient. The influence of these parameters on the bubble growth were analyzed and compared with approximate solutions by varying the dimensionless parameters $C_2$, $P^*_{g0}$ and $C_4$, respectively. In this analysis an initial bubble radius of one micron, which is greater than the critical bubble size [11], was used to ensure the growth. All the results in this study are presented in terms of the dimensionless time ($t^* = t/t_{ref}$). The average value of $\mu$ for highly viscous liquids is about $10^4$ Pa s and, hence, the value of $t_{ref}$ in all the cases was chosen to be 0.1 s. The predictions of the bubble growth obtained by using polynomial profiles for the concentration were verified by comparing them with the results of Barlow and Langlois [3].

Figures 2–4 show the effect of thickness of the liquid shell (parameter $C_2$) enveloping the bubble on the bubble growth. The three different values of $C_2$ correspond to thin, medium and thick shells. The results show that as the thickness of the liquid shell enveloping the bubble increases, the times to reach the equilibrium radius increases. Also, since the mass of the gas within the shell increases with its thickness, the value of the equilibrium radius will be larger. At the onset of the growth, the approximate solution follows the bubble growth history within 3–10%. We define the onset or inception of the growth process as the initial growth period from time equal to zero to a time of the order of the time scale for the momentum transfer ($t_{ref}$). Mathematically, $0 \leqslant t^* \leqslant 1$ represents the onset of the growth process. Also, it may be noticed that there is not an appreciable difference between the second- and third-order polynomial solutions; hence use of higher order polynomials to describe the concentration does not result in higher accuracy. At large times, as the boundary condition at the outer surface is no longer valid, the approximate solutions do not depict the correct physics of the growth dynamics. It must be pointed out that the polynomial solutions overpredict (by a factor of 3 or 4) the final equilibrium radius. The relative error in the final equilibrium radius decreases as the shell thickness increases. This suggests that as the thickness of the shell of liquid surrounding the bubble tends to infinity, one would expect a constant relative error between the two solutions. This error will be determined by the choice of polynomial concentration profile, since the percentage of the gas diffusing out of the liquid will be small enough for the simplified boundary condition of undisturbed concentration at infinity to be satisfactory.

Figures 3, 5 and 6 demonstrate the effect of the diffusion coefficient on the growth dynamics by changing the value of the parameter $C_4$ by two and four orders of magnitude. An increase in the diffusion coefficient accelerates the diffusion process. Therefore, the bubble attains its equilibrium radius faster. However, for the case of the polynomial profile, increasing the diffusion coefficient results in a higher growth rate throughout the entire growth period. Therefore, the approximate solutions significantly overestimate the growth rate as seen from these figures. The error decreases as the diffusion coefficient

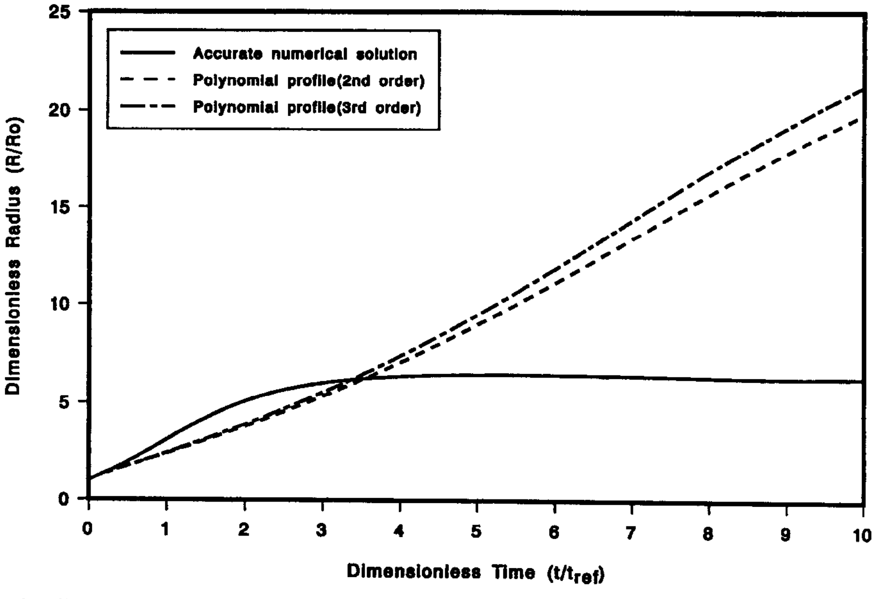

FIG. 2. Effect of shell thickness on bubble growth (thin shell, $C_2 = 10^3$, $C_3 = 0.4$, $C_4 = 100$, $P^*_{g0} = 10$).

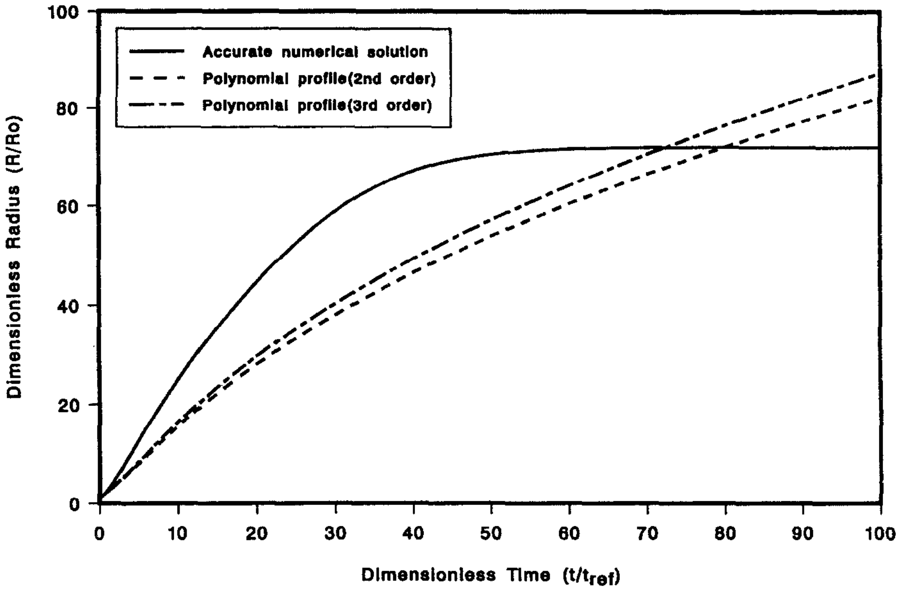

FIG. 3. Effect of shell thickness on bubble growth (medium thickness shell, $C_2 = 10^6$, $C_3 = 0.4$, $C_4 = 100$, $P^*_{g0} = 10$).

is decreased. For low values of the gas diffusivity and at the early stages of the growth process the error in the predictions is negligible.

The influence of the initial gas concentration on the growth is depicted in Figs. 3, 7 and 8. The results are for three different values of the dimensionless initial gas pressure of 5, 10 and 20, which are related to the initial gas concentration via Henry's law. Higher initial gas pressures were not chosen because they may violate the ideal gas law and because they are seldom observed in practical situations. These predictions show that the polynomial profiles underestimate the growth rate initially. However, the error decreases as the initial gas concentration is increased, indicating that polynomial profiles are reasonable approximations at higher initial gas concentrations.

Figure 9 shows a comparison between the exact solution and the third-order profiles at the inception of the growth process for three different values of the parameter $C_4$, which measures the time scale for diffusion as normalized with the time scale for the momentum transfer. The results indicate that the

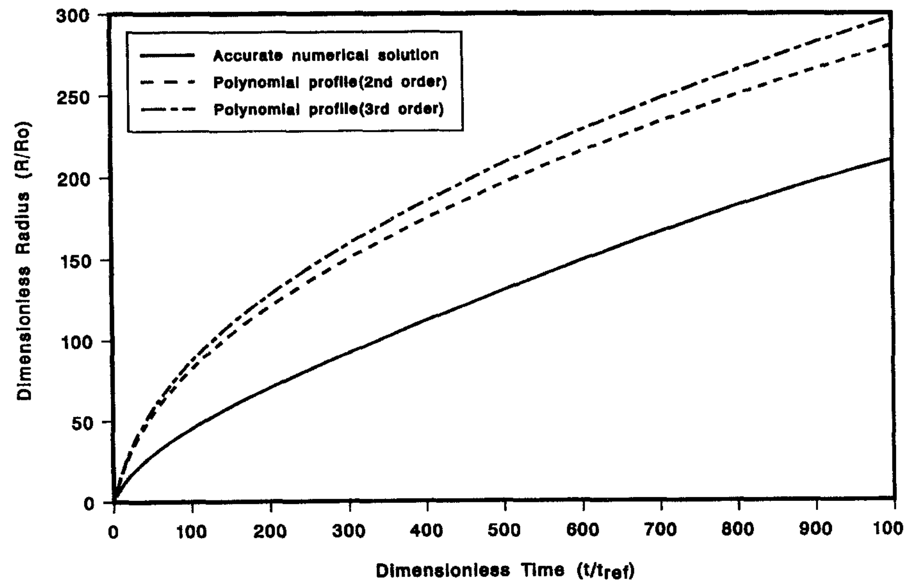

FIG. 4. Effect of shell thickness on bubble growth (thick shell, $C_2 = 10^8$, $C_3 = 0.4$, $C_4 = 100$, $P^*_{g0} = 10$).

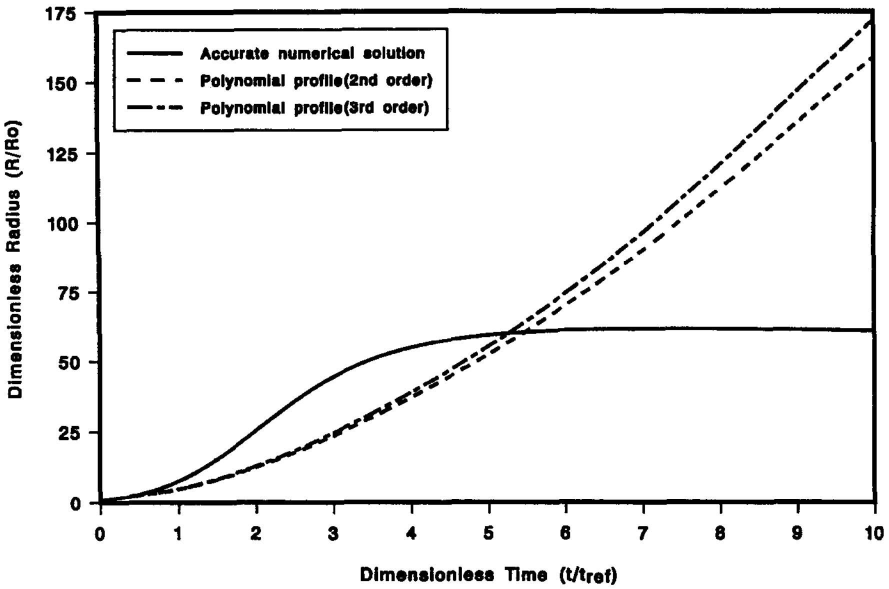

FIG. 5. Effect of gas diffusivity on bubble growth (high gas diffusivity, $C_2 = 10^6$, $C_3 = 0.4$, $C_4 = 10^4$, $P_{g0}^* = 10$).

polynomial profiles become more accurate for lower values of $C_4$. Actually, the relative difference between the two solutions is less than 6% when $C_4$ is less than or of the order of unity. Further investigation revealed that this is true irrespective of the magnitudes of the parameters $C_2$ and $P_{g0}^*$. This indicates that polynomial profiles are good approximations for describing the concentration to predict diffusion-induced growth of a gas bubble up to the time scale of the order of $t_{ref}$ and only if the diffusion time scale is smaller than $t_{ref}$.

## 7. CONCLUSION

A numerical technique to solve for the exact concentration profile is introduced and used in the equations that govern the mass diffusion-induced bubble growth in a viscous liquid containing limited dissolved gas under isothermal conditions. Scaling analysis is performed before the resulting numerical predictions are compared with previous approximate numerical solutions of the diffusion-induced growth of a gas bubble in a viscous liquid. The approximate solutions

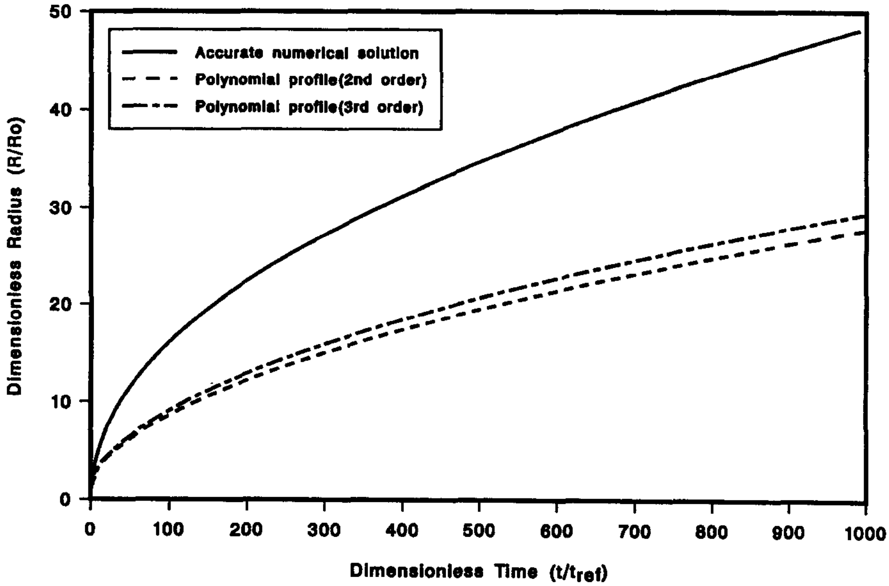

FIG. 6. Effect of gas diffusivity on bubble growth (low gas diffusivity, $C_2 = 10^6$, $C_3 = 0.4$, $C_4 = 1$, $P_{g0}^* = 10$).

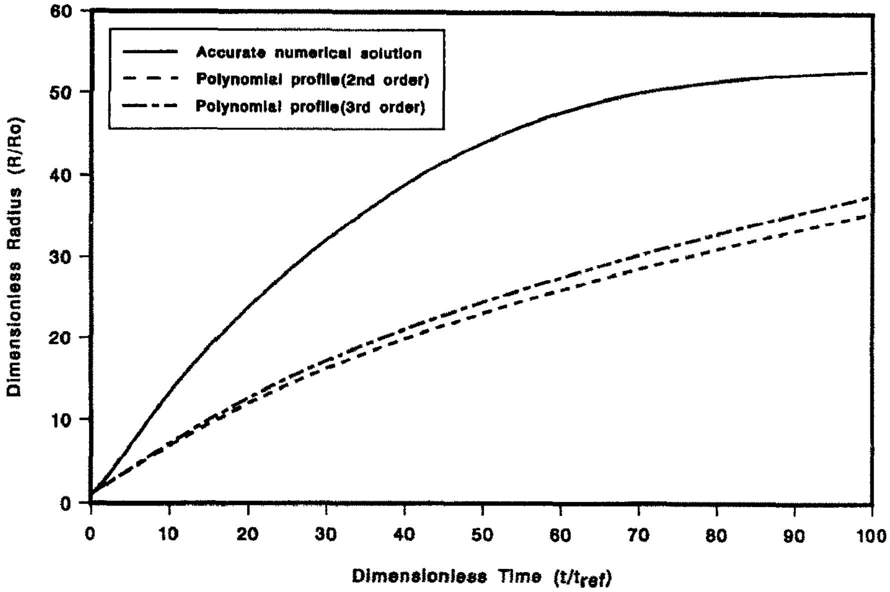

FIG. 7. Effect of initial concentration of dissolved gas on bubble growth (low gas concentration, $C_2 = 10^6$, $C_3 = 0.4$, $C_4 = 100$, $P^*_{g0} = 5$).

considered in this analysis were obtained using second- and third-order polynomial profiles for the gas concentration in the liquid. The influence of various dimensionless parameters on the dynamics of growth was investigated and the results were compared with the approximate solutions to establish the conditions under which the latter can be used without introducing significant errors. The thickness of the liquid shell around the bubble, the initial concentration of the dissolved gas and the gas diffusivity were among the parameters varied through appropriate dimensionless numbers. The parametric study revealed that the approximate solutions are satisfactory at the onset of the growth if the parameter $C_4$ is of the order of one. At the later stages of the process, the approximate solutions do not model the correct physics due to the boundary condition of an unlimited amount of dissolved gas in the liquid. We therefore recommend the use of approximate solutions only at the inception of growth and when the diffusion time

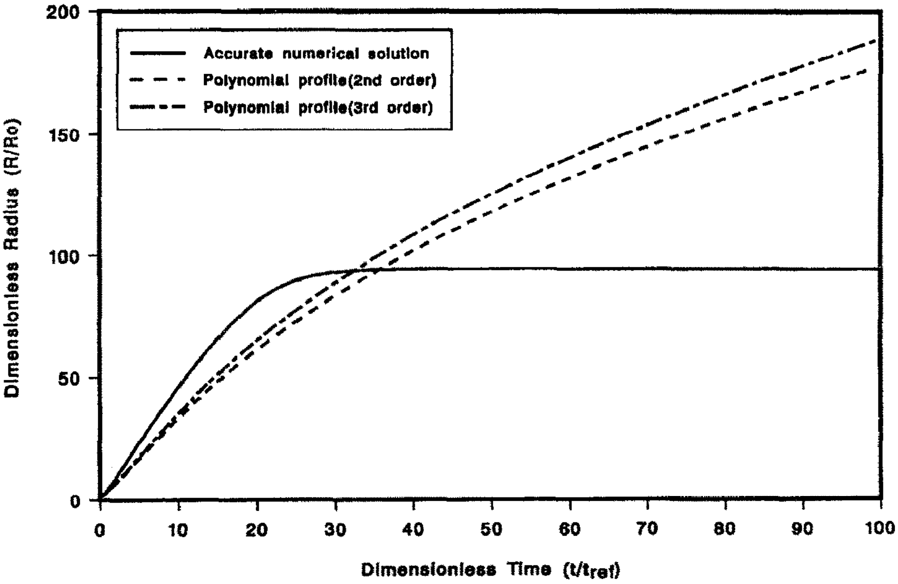

FIG. 8. Effect of initial concentration of dissolved gas on bubble growth (high gas concentration, $C_2 = 10^6$, $C_3 = 0.4$, $C_4 = 100$, $P^*_{g0} = 20$).

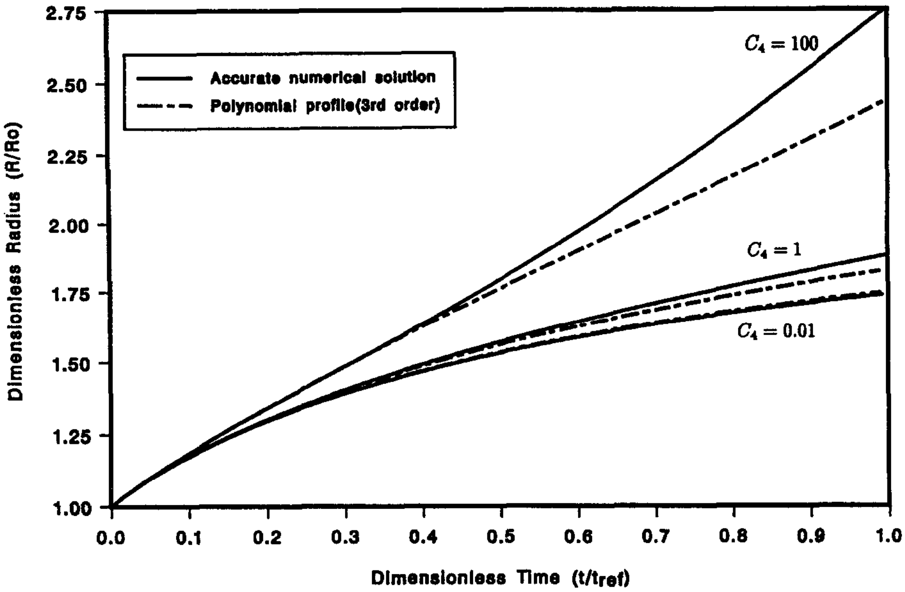

FIG. 9. Comparison betwen solutions at inception of growth ($C_2 = 10^6$, $C_3 = 0.4$, $P^*_{g0} = 10$).

scales are smaller or of the order of the time scale for momentum transfer.

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## SOLUTION NUMERIQUE PRECISE POUR LA CROISSANCE DE BULLE INDUITE PAR LA DIFFUSION MASSIQUE DANS DES LIQUIDES VISQUEUX CONTENANT DES GAZ DISSOUS

**Résumé**—Une formulation mathématique basée sur la théorie potentielle est utilisée pour développer une technique numérique précise afin de calculer la croissance induite par la diffusion massique d'une bulle de gaz sphérique entourée par un liquide newtonien visqueux avec une concentration limite de gaz dissous sous des conditions isothermes. Le profil complet de concentration de gaz dans le liquide autour de la bulle est déterminé pour prédire la croissance de la bulle. Ces résultats sont en contraste avec la croissance prédite par l'approche usuelle de profils polynomiaux pour décrire la concentration en gaz. On examine l'influence des paramètres dimensionnels actifs sur les différences dans la croissance de bulle dues au choix du profil de concentration. Il est révélé que les solutions approchées couramment utilisées pour le profil de concentration non seulement sousestiment la vitesse de croissance au début mais aussi surestiment le rayon de bulle à l'état permanent, à cause de l'hypothèse implicite de la fourniture illimitée de gaz dissous dans le liquide.

# EXAKTE NUMERISCHE LÖSUNG DES DURCH STOFFTRANSPORT GESTEUERTEN BLASENWACHSTUMS IN VISKOSEN FLÜSSIGKEITEN, DIE BEGRENZTE MENGEN GELÖSTES GAS ENTHALTEN

**Zusammenfassung**—Mit Hilfe der mathematischen Beschreibung im Rahmen der Potentialtheorie wird eine genaue numerische Technik zur Berechnung des durch Stofftransport gesteuerten Wachstums einer sphärischen Blase in einem viskosen Newton'schen Fluid mit begrenzter Konzentration gelösten Gases unter isothermen Bedingungen entwickelt. Um das Blasenwachstum zu bestimmen, wird für das gelöste Gas das vollständige Konzentrationsprofil in der Umgebung der Blase berechnet. Die Ergebnisse werden jener Dynamik des Blasenwachstums gegenübergestellt, die unter Verwendung der weit verbreiteten Näherung polynomischer Profile zur Beschreibung der Gaskonzentration ermittelt wurde. Der Einfluß der wesentlichen dimensionslosen Parameter auf das unterschiedliche Blasenwachstum—abhängig von der Wahl des Konzentrationsprofils—wird untersucht. Die Untersuchung zeigt, daß die üblicherweise verwendeten Näherungslösungen für das Konzentrationsprofil nicht nur die Wachstumsgeschwindigkeit im frühen Stadium unterschätzen, sondern auch durch die implizite Annahme einer unbeschränkten Versorgung mit gelöstem Gas in der Flüssigkeit den Blasendurchmesser im stationären Zustand überschätzen.

# ТОЧНЫЙ ЧИСЛЕННЫЙ РАСЧЕТ РОСТА ПУЗЫРЬКОВ ЗА СЧЕТ ДИФФУЗИИ В ВЯЗКИХ ЖИДКОСТЯХ, СОДЕРЖАЩИХ РАСТВОРЕННЫЙ ГАЗ ОГРАНИЧЕННОЙ КОНЦЕНТРАЦИИ

**Аннотация**—Для разработки метода численого расчета, обусловленного диффузией роста сферического газового пузырька, окруженного вязкой ньютоновской жидкостью с ограниченной концентрацией растворенного газа в изотермических условиях, используется математическая формулировка на основе теории потенциала. Для расчета роста пузырьков определяется профиль концентраций растворенного газа в жидкости, окружающей пузырек. Полученные результаты сравниваются с динамикой роста пузырьков, рассчитанной широко растпространенным методом с использованием полиномильных профилей. Исследуется влияние основных безразмерных параметров на рост пузырьков при различном выборе профиля концентраций. Показано, что общепринятые приближенные решения для профиля концентраций не только занижают интенсивность роста на ранних стадиях, но и завышают стационарный радиус пузырька в силу неявного предположения о неограниченной подаче растворенного газа в жидкость.