# Slow Bubble Growth and Dissolution in a Viscoelastic Fluid

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**ABSTRACT:** A simple equation is derived for the time dependence of the bubble radius for the diffusion-induced slow growth or dissolution of a spherical gas bubble in a viscoelastic fluid of infinite extent. The constitutive equation for a first-order fluid and a surface–volume perturbation scheme are used to develop the solution, and the effect of viscosity level and elasticity on the bubble dynamics is considered. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci **67:** 2093–2103, 1998

**Key words:** bubble growth; bubble dissolution; viscoelasticity

## INTRODUCTION

A number of important problems in chemical and polymer processes involve the diffusion-induced growth or dissolution of spherical drops or bubbles. Consequently, there has been a relatively large number of investigations concerned with the growth or dissolution of spherical particles in inviscid, viscous Newtonian, and viscoelastic non-Newtonian fluids. For the case of inviscid fluids, a similarity transformation can be used to construct an exact analytical solution[1] for the growth of a sphere from a zero initial radius. However, in general, investigation of this nonlinear moving boundary problem necessarily involves the construction of asymptotic analytical solutions or the development of numerical solutions. For example, perturbation solutions have been derived for both the slow[2] and rapid[3] growth or dissolution of spherical particles. Many of the approximate analytical solutions to the transport equations describing the diffusion-induced growth or dissolution of an isolated sphere in an inviscid fluid are discussed and evaluated elsewhere.[4]

The analysis of course becomes more complicated when hydrodynamic resistance is incorporated into the transport problem. Barlow and Langlois[5] considered bubble growth in a Newtonian fluid using the coupled momentum and diffusion equations. Street,[6] Folger and Goddard,[7] and Papanastasiou et al.[8] analyzed the growth and/or collapse of bubbles in viscoelastic fluids in the absence of mass transfer effects. Zana and Leal,[9] Han and Yoo,[10] and Arefmanesh and Advani[11] studied the growth or dissolution of bubbles in viscoelastic fluids by incorporating both hydrodynamic and mass transfer effects in the analysis of the transport process. In these investigations, a variety of differential and integral constitutive equations were utilized. In all of the above studies, the momentum equations or the coupled diffusion and momentum equations were, for the most part, solved using numerical methods.

In some cases, the velocity of the bubble interface is small compared to the rate of growth of the concentration boundary layer, and it is possible to derive relatively simple results by limiting the analysis to slowly moving interfaces. Previously, it was shown[2] that it is possible to construct a perturbation solution for spherical moving boundary problems in inviscid fluids for the limiting case of slow growth or dissolution rates. It is important to emphasize here that it is the concentration driving force for bubble growth or dissolution, and not the diffusivity, which determines whether a bubble growth or dissolution process is slow or

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rapid. This will be evident when the appropriate dimensionless groups are defined below. A similar perturbation approach should be applicable to the analysis of the slow growth or dissolution of bubbles in viscous Newtonian and viscoelastic non-Newtonian fluids. In addition, there is an additional simplification in the analysis of spherical moving boundary problems for slowly moving interfaces in viscoelastic fluids. Since the diffusion-induced growth or collapse of a bubble in a fluid involves an unsteady flow field in a fluid initially at rest, it should be possible, in certain cases, to use the constitutive equation for a first-order fluid to describe the viscoelastic material.[12] The first-order fluid represents the first-order term of a retarded motion expansion for a linear viscoelastic fluid, valid for unsteady flows of fluids that were initially at rest. The utilization of the constitutive equation for a first-order fluid greatly simplifies the analysis of hydrodynamic effects in the growth or dissolution process.

The principal objective of this article is to derive a simple equation for the time dependence of the bubble radius for the diffusion-induced slow growth or dissolution of a spherical gas bubble in a viscoelastic fluid of infinite extent. The assumptions used in the problem formulation are listed in the second section of the article, and the equations describing the coupled mass transfer–hydrodynamic problem are presented in the third section. The perturbation solution for a slowly moving interface is developed in the fourth section of the article, and the basic predictions of the theory are discussed in the final section of the article.

## PROBLEM FORMULATION

The following assumptions are used to formulate equations for the growth or dissolution of a gas bubble in an infinite sea of a viscoelastic fluid.

1. The effect of heat released or absorbed during phase change is considered negligible so that the transport process is effectively isothermal.
2. The velocity field in the outer fluid is purely radial, and the concentration field is spherically symmetric.
3. The outer phase consists of a dissolved gas and liquid, and the amount of dissolved gas is small enough so that the density $\rho$ of the outer phase is essentially constant.
4. The bubble is a perfect sphere isolated in an infinite liquid phase, and the origin of the coordinate system is the bubble center, which is at rest.
5. There are no chemical reactions in the infinite outer phase.
6. All gravitational effects are negligible, and it is assumed that surface tension effects are small for the system of interest.
7. The bubble is a one-component system with uniform density $\hat{\rho}$, uniform pressure $p_g$, and a radial velocity field. The gas in the bubble is considered to be an inviscid fluid.
8. The diffusion process in the outer liquid phase is adequately described by a linear constitutive equation (Fickian diffusion), and the binary mutual diffusion coefficient, $D$, in the outer phase is effectively a constant.
9. The initial solute concentration in the outer liquid phase, $\rho_{10}$, and the initial pressure, $p_0$, are uniform. The initial bubble density corresponding to $p_0$ is $\hat{\rho}_0$, and the initial bubble radius is $R_0$.
10. There exists concentration equilibrium at the phase boundary, and this phase equilibrium is described by the following linear relationship:

$$\rho_1(R, t) = Kp_g \tag{1}$$

Here, $\rho_1(r, t)$ is the mass density of solute in the liquid phase, $R$ is the radius of the bubble, $t$ is time, $r$ is the radial position variable in spherical coordinates, and $K$ is a Henry's law constant. The solute concentration corresponding to the initial pressure $p_0$ is $\rho_{1E}$.

11. The incompressible, effectively pure liquid in the outer phase is described by the constitutive equation for a first-order fluid, valid for unsteady flows of fluids that are rest for $t < 0$. For a first-order fluid, the extra stress $\underset{\sim}{S}$ is given by the expression

$$\underset{\sim}{S} = \eta_0 f(t) \underset{\sim}{A}_1 \tag{2}$$

where $\eta_0$ is the viscosity of the fluid at zero shear rate, and $\underset{\sim}{A}_1$ is the first Rivlin–Ericksen tensor evaluated at time $t$. Also,

$$f(t) = \left[ 1 - \frac{\int_t^\infty G(s)ds}{\eta_0} \right] \tag{3}$$

where $G(s)$ is the shear stress relaxation

modulus of linear viscoelasticity. A simple, special expression for $G(s)$ is

$$G(s) = a\lambda e^{-s/\lambda} \tag{4}$$

where $a$ and $\lambda$ are constants for a given material. For this case,

$$f(t) = 1 - e^{-t/\lambda} \tag{5}$$

and $\lambda$ can be identified as a characteristic relaxation time for the fluid. Justification for using this constitutive equation for slow growth or dissolution processes is discussed below.

12. The density of the gas bubble is much smaller than the liquid density so that

$$\frac{\hat{\rho}_0}{\rho} \cong 0 \tag{6}$$

13. Pressure changes in the liquid phase caused by inertia effects are small. As will be evident below from the dimensionless forms of the transport equations, the magnitude of this effect is governed by the dimensionless group $N_I$, as follows.

$$N_I = \frac{\rho D^2}{p_0 R_0^2} \tag{7}$$

For most cases of interest,

$$N_I \cong 0 \tag{8}$$

14. The gas in the bubble is an ideal gas so that, at constant temperature,

$$\frac{\hat{\rho}}{\hat{\rho}_0} = \frac{p_g}{p_0} \tag{9}$$

## FORMULATION OF EQUATIONS

The above assumptions can now be used as the basis for the derivation of specific equations for the transport process from appropriate equations of change in the bulk fluids and from appropriate jump conditions at the phase boundary. The transport process in the bulk fluids is described by the overall continuity equation in the liquid,

$$\nabla \cdot \underset{\sim}{v} = 0 \tag{10}$$

by the continuity equation for the bubble phase,

$$\frac{\partial \hat{\rho}}{\partial t} + \nabla \cdot (\hat{\rho} \underset{\sim}{\hat{v}}) = 0 \tag{11}$$

by the equation of motion for an incompressible first-order fluid,

$$\rho\left(\frac{\partial \underset{\sim}{v}}{\partial t} + \underset{\sim}{v} \cdot \nabla \underset{\sim}{v}\right) = -\nabla p + \eta_0 f(t) \nabla^2 \underset{\sim}{v} \tag{12}$$

and by the species continuity equation for the dissolved gas in the outer liquid phase,

$$\frac{\partial \rho_1}{\partial t} + \underset{\sim}{v} \cdot \nabla \rho_1 = D \nabla^2 \rho_1 \tag{13}$$

In these equations, $\underset{\sim}{v}$ and $\underset{\sim}{\hat{v}}$ are the velocity vectors in the liquid and bubble phases, respectively, $p$ is the pressure in the liquid phase, and gravitational effects have been neglected. Similarly, the transport process at the phase interface is described by the overall jump mass balance,

$$\hat{\rho}(\underset{\sim}{\hat{v}} \cdot \underset{\sim}{n}^* - \underset{\sim}{U}^* \cdot \underset{\sim}{n}^*) = \rho(\underset{\sim}{v} \cdot \underset{\sim}{n}^* - \underset{\sim}{U}^* \cdot \underset{\sim}{n}^*) \tag{14}$$

by the jump species mass balance for the polymer,

$$\hat{\rho}_2(\underset{\sim}{\hat{v}}_2 \cdot \underset{\sim}{n}^* - \underset{\sim}{U}^* \cdot \underset{\sim}{n}^*) = \rho_2(\underset{\sim}{v}_2 \cdot \underset{\sim}{n}^* - \underset{\sim}{U}^* \cdot \underset{\sim}{n}^*) \tag{15}$$

and by the jump linear momentum equation,

$$\hat{\rho}\underset{\sim}{\hat{v}}(\underset{\sim}{\hat{v}} \cdot \underset{\sim}{n}^* - \underset{\sim}{U}^* \cdot \underset{\sim}{n}^*) - \underset{\sim}{\hat{T}} \cdot \underset{\sim}{n}^* = \rho \underset{\sim}{v}(\underset{\sim}{v} \cdot \underset{\sim}{n}^* - \underset{\sim}{U}^* \cdot \underset{\sim}{n}^*) - \underset{\sim}{T} \cdot \underset{\sim}{n}^* \tag{16}$$

In these equations, $\underset{\sim}{n}^*$ is the unit normal vector at the phase interface pointing into the liquid phase, $\underset{\sim}{U}^*$ is the velocity of the phase boundary, $\rho_2$ and $\hat{\rho}_2$ are the polymer mass densities in the liquid and bubble phases, $\underset{\sim}{v}_2$ and $\underset{\sim}{\hat{v}}_2$ are the velocities of the polymer in the liquid and bubble phases, and $\underset{\sim}{T}$ and $\underset{\sim}{\hat{T}}$ are the total stress tensors in the liquid and bubble phases, respectively. All of the quantities in eqs. (14)–(16) must, of course, be evaluated at the phase interface.

The transformation of these equations to specific results for the present problem is facilitated if the following dimensionless variables are introduced.

$$t^* = \frac{Dt}{R_0^2} \tag{17}$$

$$r^* = \frac{r}{R_0} \tag{18}$$

$$R^* = \frac{R}{R_0} \tag{19}$$

$$\hat{\rho}^* = \frac{\hat{\rho}}{\hat{\rho}_0} \tag{20}$$

$$v = \frac{R_0 v_r}{D} \tag{21}$$

$$\hat{v} = \frac{R_0 \hat{v}_r}{D} \tag{22}$$

$$C = \frac{\rho_1 - \rho_{10}}{\rho_{1E} - \rho_{10}} \tag{23}$$

$$C_S = \frac{\rho_1(R, t)}{\rho_{1E}} \tag{24}$$

In these equations, $v_r$ and $\hat{v}_r$ are the radial velocity components in the liquid and bubble phases, respectively. In the dimensionless forms of the transport equations, the asterisks will be dropped for convenience.

Integration of eqs. (10) and (11) produces the following results for the dimensionless velocity field:

$$v(r, t) = \frac{R^2}{r^2} v(R, t) \tag{25}$$

$$\hat{v}(R, t) = -\frac{R}{3\hat{\rho}} \frac{d\hat{\rho}}{dt} \tag{26}$$

Also, combination of an integrated form of eq. (12) with eqs. (1), (2), (14), (16), and (25) gives the following dimensionless expression:

$$C_S = 1 + \frac{4N_V f(t) v(R, t)}{R} + \mathrm{N}_I \left[ -(\hat{v} - v)\left( v - \frac{dR}{dt} \right) + 2v \frac{dR}{dt} + R \frac{dv}{dt} - \frac{v^2}{2} \right]_{r=R} \tag{27}$$

$$N_V = \frac{\eta_0 D}{p_0 R_0^2} \tag{28}$$

Evaluation of eq. (14) at the interface and substitution of eq. (26) produce the following result for the liquid phase velocity at the phase boundary:

$$v(R, t) = -\frac{R\hat{\rho}_0}{3\rho} \frac{d\hat{\rho}}{dt} + \frac{dR}{dt}\left( 1 - \frac{\hat{\rho}\hat{\rho}_0}{\rho} \right) \tag{29}$$

Another equation at the phase boundary can be derived by evaluating eq. (15) at the interface and by using eqs. (1), (9), and (29):

$$C_S \frac{dR}{dt} = -\frac{R}{3} \frac{dC_S}{dt} + \frac{N_a}{Q} \left( \frac{\partial C}{\partial r} \right)_{r=R} \tag{30}$$

$$Q = \frac{1 - \frac{C_s \rho_{1E}}{\rho}}{1 - \frac{\rho_{1E}}{\rho}} \tag{31}$$

$$N_a = \frac{\rho(\rho_{1E} - \rho_{10})}{\hat{\rho}_0(\rho - \rho_{1E})} \tag{32}$$

For most cases of interest, eqs. (6) and (8) are valid, and eqs. (27) and (29) can be reduced to the following simplified forms:

$$C_S = 1 + \frac{4N_V f(t)}{R} \frac{dR}{dt} \tag{33}$$

$$v(R, t) = \frac{dR}{dt} \tag{34}$$

Consequently, the species continuity equation for the dissolved gas, eq. (13), can be written as follows in dimensionless form:

$$\frac{\partial C}{\partial t} + \frac{R^2}{r^2} \frac{dR}{dt} \frac{\partial C}{\partial r} = \frac{\partial^2 C}{\partial r^2} + \frac{2}{r} \frac{\partial C}{\partial r} \tag{35}$$

Furthermore, the appropriate boundary conditions can be expressed as follows:

$$C(r, 0) = 0 \tag{36}$$

$$C(\infty, t) = 0 \tag{37}$$

$$C[R(t), t] = 1 + \frac{\rho_{1E}(C_S - 1)}{\rho_{1E} - \rho_{10}} \tag{38}$$

Also, the initial conditions for $R$ and $C_S$ are simply

$$R(0) = 1 \tag{39}$$

$$C_S(0) = 1 \tag{40}$$

The analysis of the bubble growth or dissolution problem thus involves solving eqs. (30), (33), and (35)–(40) for $C_S(t)$, $R(t)$, and $C(r, t)$. In general, a numerical method must be used to solve this nonlinear set of equations. However, for the case of slow growth or dissolution, it is possible to formulate a perturbation method, and this is considered in the next section.

## PERTURBATION SOLUTION

When the driving force for bubble growth or dissolution is small $[(\rho_{1E} - \rho_{10}) \to 0]$, it follows from eq. (32) that $N_a \to 0$. As $N_a \to 0$, it is evident from eqs. (30) and (33) that $R(t) = 1$ so that there is no change in the initial bubble radius for $N_a \to 0$. Hence, a slow growth or dissolution process is simply one for which the dimensionless group $N_a$ is small, and it is thus convenient to develop a parameter perturbation solution to the transport problem using $N_a$ as the small parameter. Consequently, we propose the following series expansions for the bubble growth or dissolution problem:

$$C(r, t) = C^0(r, t) + N_a C^1(r, t) + \cdots \quad (41)$$

$$C_S(t) = 1 + N_a C_S^1(t) + \cdots \quad (42)$$

$$R(t) = 1 + N_a R_1(t) + \cdots \quad (43)$$

It is evident that it is both the time dependence of the position of the bubble surface and the basic nonlinearity of the equation set that take the problem out of the exactly solvable class. Hence, it is convenient to utilize a surface–volume perturbation scheme to develop a solution to the transport problem. The following Taylor series expansions can be used to eliminate quantities evaluated at the phase interface:

$$C^i[R(t), t] = C^i(1, t) + \left(\frac{\partial C^i}{\partial r}\right)_{r=1} (R - 1) + \cdots \quad (44)$$

$$\left(\frac{\partial C^i}{\partial r}\right)_{r=R} = \left(\frac{\partial C^i}{\partial r}\right)_{r=1} + \left(\frac{\partial^2 C^i}{\partial r^2}\right)_{r=1} (R - 1) + \cdots \quad (45)$$

Substitution of eqs. (41)–(43) into eqs. (30), (33), and (35)–(40) and utilization of eqs. (44) and (45) produce the following set of equations for the lowest order perturbation result:

$$\frac{\partial C^0}{\partial t} = \frac{\partial^2 C^0}{\partial r^2} + \frac{2}{r}\frac{\partial C^0}{\partial r} \quad (46)$$

$$C^0(r, 0) = 0 \quad (47)$$

$$C^0(\infty, t) = 0 \quad (48)$$

$$C^0(1, t) = 1 \quad (49)$$

$$C_S^1(t) = 4N_V f(t)\frac{dR_1}{dt} \quad (50)$$

$$C_S^1(0) = 0 \quad (51)$$

$$\frac{dR_1}{dt} = -\frac{1}{3}\frac{dC_S^1}{dt} + \left(\frac{\partial C^0}{\partial r}\right)_{r=1} \quad (52)$$

$$R_1(0) = 0 \quad (53)$$

Equations (46)–(53) can thus be used to determine the lowest-order perturbation series functions, $C^0(r, t)$, $C_S^1(t)$, and $R_1(t)$.

The solution to eqs. (46)–(49) is simply

$$C^0 = \frac{\operatorname{erfc}\left(\dfrac{r-1}{2t^{1/2}}\right)}{r} \quad (54)$$

and the following derivative can be used in eq. (52):

$$\left(\frac{\partial C^0}{\partial r}\right)_{r=1} = -1 - \frac{1}{\pi^{1/2} t^{1/2}} \quad (55)$$

Also, integration of eq. (52), utilization of eqs. (51) and (53), and substitution of eq. (50) produce the following ordinary differential equation for $R_1(t)$:

$$\frac{dR_1}{dt} + \frac{3R_1}{4N_V[1 - \exp(-tN_E)]} = \frac{3F(t)}{4N_V[1 - \exp(-tN_E)]} \quad (56)$$

Here,

$$F(t) = \int_0^t \left(\frac{\partial C^0}{\partial r}\right)_{r=1} dt' = -t - \frac{2t^{1/2}}{\pi^{1/2}} \quad (57)$$

and we have used the following dimensionless form of eq. (5):

$$f(t) = 1 - \exp(-tN_E) \tag{58}$$

$$N_E = \frac{R_0^2}{D\lambda} \tag{59}$$

The solution to eqs. (53) and (56) can be expressed as follows:

$$R_1(t) = \frac{3\exp\left(-\frac{3t}{4N_V}\right)}{4N_V[1-\exp(-tN_E)]^b} \times \int_0^t \frac{F(\tau)\exp\left(\frac{3\tau}{4N_V}\right)[1-\exp(-\tau N_E)]^b d\tau}{[1-\exp(-\tau N_E)]} \tag{60}$$

$$b = \frac{3}{4N_V N_E} \tag{61}$$

Hence, eqs. (43) and (60) give a relatively simple expression for the bubble radius, $R(t)$, valid for small $N_a$ and for any values of $N_V$ and $N_E$. The dimensionless group $N_V$ characterizes the importance of viscous effects in the bubble growth or bubble dissolution process; for an inviscid fluid, $N_V = 0$. The dimensionless group $N_E$ characterizes the importance of fluid elasticity; for an inelastic material, $\lambda \to 0$ and $N_E \to \infty$.

The above result for $R_1(t)$, of course, represents the lowest-order perturbation series result for the bubble radius, and higher-order results can be derived with a significant increase in the labor involved. In addition, it has been shown that the perturbation expansion for slow growth or dissolution in an inviscid outer liquid exhibits singular behavior at large times.[2] Although the zero-order solution is well behaved for all time, the first-order correction is unbounded at large times, and the perturbation expansion is not uniformly valid. In fact, there is singular behavior in both space and time, and a space–time matching scheme has to be developed to form a uniformly valid composite solution. Here, we present only the zero-order result, which will be useful for sufficiently small $N_a$.

Evaluation of $R(t)$ requires only a numerical integration of the integral in eq. (60). This is a relatively simple procedure, although it is necessary to handle an integrable singularity at the lower limit. Also, some simpler results can be derived from eq. (60) for several important limiting cases. For an inelastic material ($N_E \to \infty$), eq. (60) reduces to the following form:

$$R_1(t) = \frac{3\exp\left(-\frac{3t}{4N_V}\right)}{4N_V}\int_0^t F(\tau)\exp\left(\frac{3\tau}{4N_V}\right)d\tau \tag{62}$$

and integration yields the following result for cases in which elastic effects can be neglected:

$$R_1(t) = -\left(t - \frac{4N_V}{3}\right) - \frac{4N_V}{3}\exp\left[-\frac{3t}{4N_V}\right] - \frac{2t^{1/2}}{\pi^{1/2}} + \frac{2}{\pi^{1/2}}\exp\left[-\frac{3t}{4N_V}\right] \times \int_0^{t^{1/2}} \exp\left[\frac{3\eta^2}{4N_V}\right]d\eta \tag{63}$$

For $N_V = 0$, we recover the well-known result for growth or dissolution in an inviscid outer liquid:

$$R(t) = 1 + N_a\left[-t - \frac{2t^{1/2}}{\pi^{1/2}}\right] \tag{64}$$

For bubble growth or dissolution in very viscous materials (large $N_V$), the following result is valid for $(3t/4N_V) \ll 1$:

$$R(t) = 1 + \frac{3N_a}{4N_V}\left[-\frac{t^2}{2} - \frac{4t^{3/2}}{3\pi^{1/2}}\right] \tag{65}$$

Furthermore, for bubble growth or dissolution in very elastic materials ($N_E \to 0$), which are also very viscous (large $N_V$), the following result is valid for $(3t/4N_V) \ll 1$ and $tN_E \ll 1$:

$$R(t) = 1 - \frac{3}{4}N_a\left[\frac{t}{\left(N_E N_V + \frac{3}{4}\right)} + \frac{2t^{1/2}}{\pi^{1/2}\left(\frac{N_E N_V}{2} + \frac{3}{4}\right)}\right] \tag{66}$$

It is reasonable to expect that this equation should describe at least the early stages of slow bubble growth or dissolution in elastic, viscous molten polymers.

As noted previously, the basic perturbation result, eqs. (43) and (60), is valid for sufficiently small $N_a$ for all values of $N_V$ and $N_E$. Equations (63)–(66) are just simpler results, which can be obtained by introducing restrictions on $N_V$ and $N_E$. We conclude this section by stating restrictions on the applicability of the proposed result, eqs. (43) and (60). This result is restricted somewhat because it is based on two simplifications. First, the constitutive equation for a first-order fluid is used to describe the viscoelastic behavior of the liquid. As noted above, the first-order fluid can be regarded as a material described by the first-order term of a retarded motion expansion for a linear viscoelastic fluid, and it is restricted to unsteady flows generated from the rest state. Since the unsteady bubble growth or dissolution problem represents a flow field that is at rest for $t < 0$, the first-order fluid should describe the viscoelasticity of the liquid phase for an appropriate range of deformation conditions. It has been shown recently[13] that the first-order fluid will provide adequate predictions for the stress field for bubble growth or dissolution when

$$t|N_a| \leqq 1 \tag{67}$$

Hence, the first-order fluid can be used even when $|N_a|$ is large if $t$ is sufficiently small. The second simplification is the utilization of a perturbation method to derive a solution to the problem. The perturbation solution should be applicable when $|N_a|$ is sufficiently small. The parameter $N_a$ is 0.03 for the dissolution of oxygen in water and 0.8 for the dissolution of carbon dioxide in water. Typically, $|N_a|$ is of the order of unity or less for bubble–liquid systems of interest in polymer processing. For the range $0 < |N_a| \leqq 1$, the perturbation solution produces good results when

$$N_a t \leqq 0.25 \tag{68}$$

for bubble dissolution and when

$$|N_a| t \leqq 1.5 \tag{69}$$

for bubble growth. Consequently, from eqs. (67)–(69) and from the above discussion, it is evident that the proposed solution for a viscoelastic liquid, eqs. (43) and (60), is valid at least for $0 < |N_a| \leqq 1$ and for dimensionless times given by eq. (68) for bubble dissolution and by eq. (67) for bubble growth. For an inelastic liquid ($N_E \to \infty$), the restriction for bubble dissolution is still eq. (68), but, for bubble growth, eq. (67) is replaced by eq. (69).

Calculations carried out using an inviscid liquid phase[4] suggest that the proposed solution can be used to describe a significant fraction of the bubble dissolution process and a time period for the bubble growth process during which significant bubble growth has occurred. Unfortunately, however, the first-order fluid can be used to describe only a small part of a bubble growth process, which involves a 200-fold increase in the bubble radius.[10] Even though the first-order fluid can not be used to describe a bubble growth process with a large increase in the bubble radius, it is evident from the above perturbation solution that there is another possibility for determining the radius–time behavior for very elastic fluids ($N_E \to 0$). At this limit, it is clear that eq. (66) reduces to eq. (64), and, consequently, the radius–time behavior for bubble growth or dissolution in a very elastic first-order fluid can be estimated using an inviscid analysis. The various solutions that are available at the inviscid limit[4] can thus be utilized. It has been shown elsewhere[13] that this result is also valid for more general viscoelastic fluids, namely, integral viscoelastic materials. If we define the ratio $\theta$ by the expression

$$\theta = \frac{N_E N_V |N_a|}{N_E + |N_a|} \tag{70}$$

then it has been shown that the radius–time curve for an integral viscoelastic model will approach the inviscid limit when

$$\theta \ll 1 \tag{71}$$

It is thus evident from the above discussion that simple rheological models can be used to describe bubble growth or dissolution in elastic liquids for two cases. The first-order fluid model [eqs. (43) and (60)] can be used for $0 < |N_a| \leqq 1$ to provide adequate results when eqs. (67) and (68) are satisfied for bubble growth and bubble dissolution, respectively. In addition, the inviscid flow approximation can be used for all time when eq. (71) is satisfied. Finally, eq. (63) can be used for $0 < |N_a| \leqq 1$ and for all values of $N_V$ for a Newton-

ian fluid when eqs. (69) and (68) are satisfied for bubble growth and bubble dissolution, respectively.

## DISCUSSION

In the absence of surface tension effects, the bubble growth or bubble dissolution process depends on the following three dimensionless groups: $N_E$ (elasticity level), $N_V$ (viscosity level), and $N_a$ (speed of growth or dissolution process). Although there has been a considerable amount of work on this problem,[5–11] important parts of the parameter space for $N_E$, $N_V$, and $N_a$ have not been considered. For example, Han and Yoo[10] obtained experimental data for bubble growth involving carbon dioxide in molten polystyrene. For this system, $N_V$ was large (22), $N_E$ was very small ($2 \times 10^{-3}$), and $|N_a|$ was of the order of unity ($N_a = -1.22$). None of the seven studies cited above considers results for this range of parameters, which can be considered to be typical for gas bubbles in molten polymers. Street,[6] Folger and Goddard,[7] and Papanastasiou et al.[8] did not include mass transfer effects, and the article of Barlow and Langlois[5] is limited to Newtonian fluids. In addition, the solutions of Barlow and Langlois, Han and Yoo,[10] and Arefmanesh and Advani[11] are, in the strictest sense, valid only for values of $|N_a|$ significantly greater than unity since they all incorporate the thin boundary layer assumption in their developments. Consequently, six of the seven studies are not applicable to typical cases where $|N_a|$ is of the order of unity or less. The article by Zana and Leal[9] does consider this range for $N_a$ for the case of bubble dissolution, but the highest value used for $N_V$ was 0.1. In addition, most of the results were obtained using $N_E = 10$, and the lowest value of $N_E$ was unity. Hence, most of the results presented by Zana and Leal are valid only when elastic effects are rather weak.

The present perturbation solution is valid for low values of $|N_a|(0 < |N_a| \leqq 1)$ rather than large values, and there is no restriction on either $N_V$ or $N_E$. Consequently, one contribution of this article is the consideration of an important part of the parameter space for $N_a$, $N_E$, and $N_V$, which has not been considered before. A second contribution is the development of an analytical solution (only a numerical integration is involved) so that numerical solutions of differential equations are not required. Results are presented first for Newtonian fluids and then for very viscous, elastic materials.

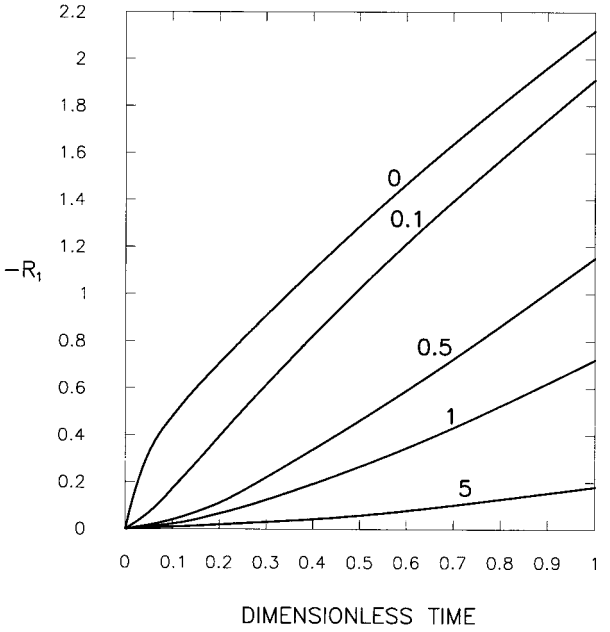


**Figure 1** Dependence of $R_1$ on dimensionless time for a Newtonian fluid. Numbers on curves are values of $N_V$.

The time dependence of $R_1$ for Newtonian fluids [defined by eq. (63)] is presented in Figure 1 for five values of $N_V$. The results for $R_1$ from this figure can be combined with eq. (43) to determine the effect of viscosity level on bubble growth and bubble dissolution for any value of $N_a$ in the range $0 < |N_a| \leqq 1$. Results for bubble dissolution for $N_a = 1$ are presented in Figure 2 for three values of $N_V$, including the inviscid limit ($N_V = 0$). It is evident from Figures 1 and 2 that the viscosity level can have a very pronounced effect on the bubble radius–time curve, and there will be significantly slower growth and dissolution in viscous materials than in an inviscid outer fluid. There appear to be no previous results for bubble growth in Newtonian liquids for small values of $|N_a|$. Zana and Leal[9] presented results for bubble dissolution in Newtonian fluids for $N_V \leqq 0.1$. Their results are similar to those presented in Figure 2, but direct comparison is not possible because their time coordinate is normalized using an unspecified reference time. The present results are easy to use since either an analytical result [eq. (63)] or a curve of $R_1$ values (Fig. 1) can be utilized. Evaluation of eq. (63) is straightforward since the integral in this equation has been tabulated (Dawson's integral).

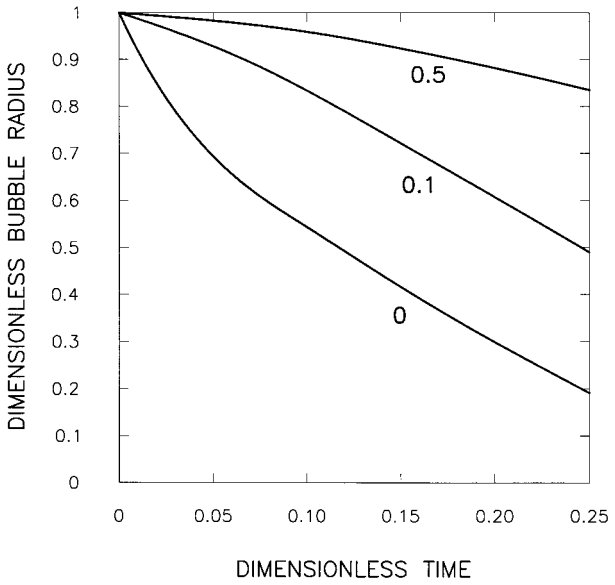


**Figure 2** Bubble dissolution for Newtonian fluid with $N_a = 1$. Numbers on curves are values of $N_V$.

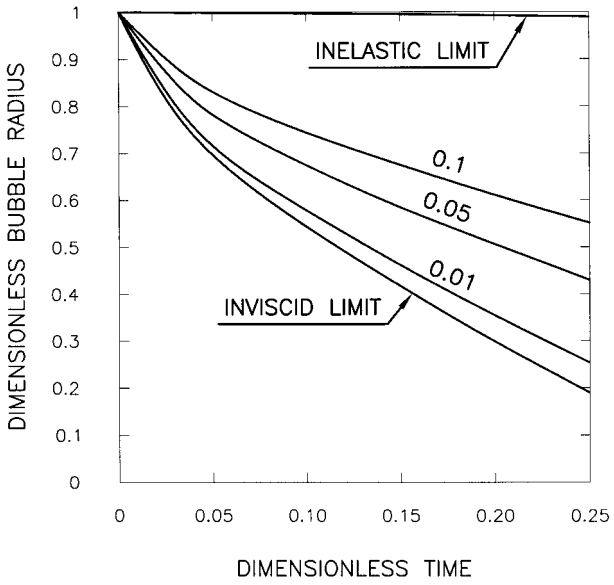


**Figure 4** Bubble dissolution for elastic fluid with $N_a = 1$ and $N_V = 10$. Numbers on curves are values of $N_E$.

The effect of fluid elasticity on bubble growth ($N_a = -1$) and bubble dissolution ($N_a = 1$) is illustrated in Figures 3 and 4, respectively. There appear to be no previous results for elastic bubble growth for small values of $|N_a|$ since previous studies[10,11] appear to have incorporated the thin boundary layer assumption (large $|N_a|$). Zana and Leal[9] presented dissolution results for small $N_a$, but, as noted above, they considered rather modest levels of viscosity (maximum $N_V = 0.1$) and elasticity (minimum $N_E = 1$). In Figures 3

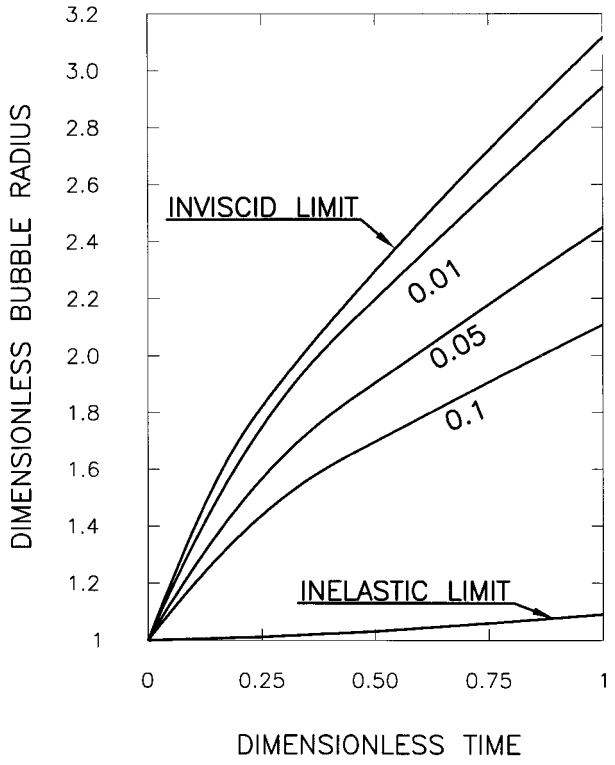


**Figure 3** Bubble growth for elastic fluid with $N_a = -1$ and $N_V = 10$. Numbers on curves are values of $N_E$.

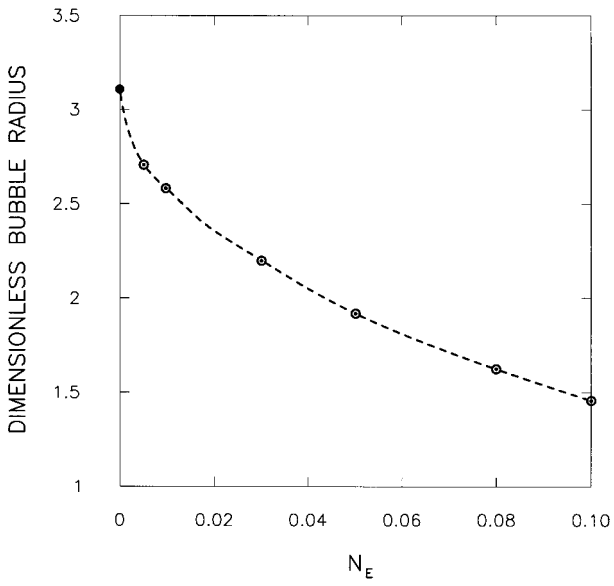


**Figure 5** Dependence of dimensionless bubble radius on $N_E$ for bubble growth with $t = 1$, $N_a = -1$, and $N_V = 10$. The solid circle represents the inviscid result and the open circles represent first-order fluid results.

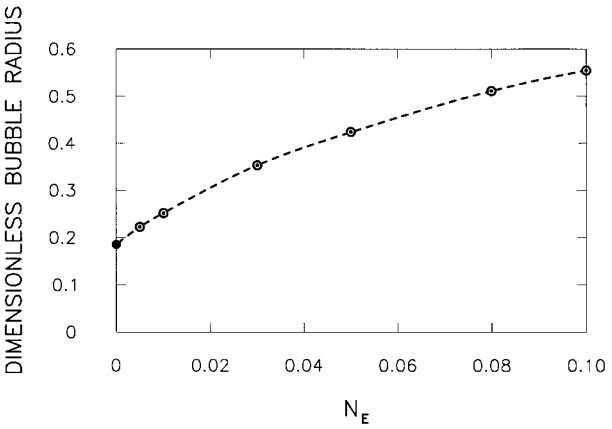


**Figure 6** Dependence of dimensionless bubble radius on $N_E$ for bubble dissolution with $t = 0.25$, $N_a = 1$, and $N_V = 10$. The solid circle represents the inviscid result, and the open circles represent first-order fluid results.

and 4, very viscous ($N_V = 10$) and very elastic ($N_E = 0.01$) materials are considered. Both these figures illustrate that the high level of viscosity produces a dramatic decrease in the growth and dissolution rates in the absence of any fluid elasticity. As the elasticity level is increased away from the inelastic limit, there is a significant increase in the growth and dissolution rates, and, at high elastic levels ($N_E \to 0$), the radius–time curves approach the inviscid limit. The addition of elasticity counteracts the effects of high viscosity. More detailed graphs of the approach to the inviscid limit for $|N_a| = 1$ and $N_V = 10$ are presented in Figures 5 and 6 for bubble growth and bubble dissolution, respectively. The solid circles in these graphs are the calculated inviscid values of the bubble radius at $t = 1$ for bubble growth and at $t = 0.25$ for bubble dissolution. The open circles represent the first-order fluid results for various values of $N_E$ as this parameter is decreased towards zero. These figures illustrate the approach of the radius–time curves for first-order fluids to the inviscid limit as $N_E \to 0$. It should be noted that the approach to the inviscid limit as $N_E$ becomes small is valid for integral viscoelastic materials, and, therefore, radius–time curves for more general viscoelastic materials can be estimated in the limit of high elasticity by simply using the inviscid flow approximation. Finally, we present curves that illustrate what level of elasticity is needed to have a significant influence on a bubble dissolution process for different viscosity levels. The dimensionless bubble radius is evaluated for a dissolving bubble at $t = 0.05$ with $N_a = 1$ as a function of $N_E$ for two values of $N_V$, 1 and 10. The results are presented in Figure 7. For the lower level of viscosity ($N_V = 1$), elasticity will begin to have a significant influence for values of $N_E$ less than 10. For the higher level of viscosity ($N_V = 10$), elasticity will become important only when $N_E$ is less than 1. Not surprisingly, greater elastic effects are needed to counteract the influence of higher viscosity levels. This conclusion is evident from examination of the early time solution, eq. (66). The radius prediction for different values of

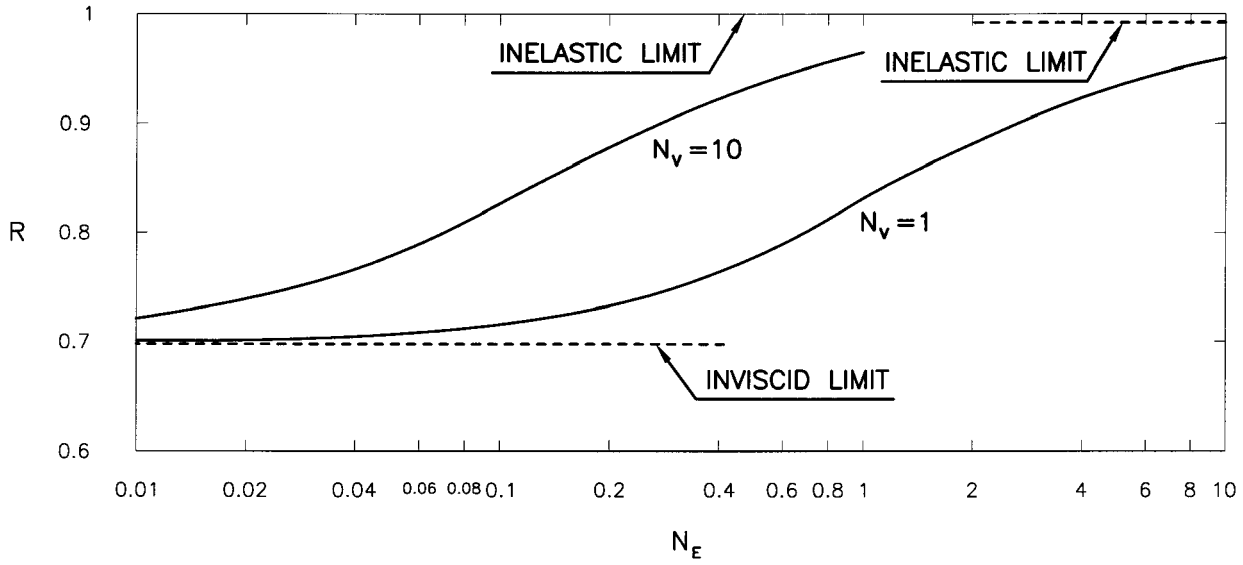


**Figure 7** Dependence of dimensionless bubble radius on $N_E$ for bubble dissolution with $t = 0.05$, $N_a = 1$, and two values of $N_V$.

$N_V$ is the same when $N_E$ is chosen so that the product $N_V N_E$ is kept constant.

It is fair to conclude that there are three contributions of the present study. First, an analytical expression is produced for bubble growth and dissolution in Newtonian and viscoelastic materials, and utilization of this result is clearly easier than numerically solving differential equations. Second, this article considers an important part of the parameter space for $N_a$, $N_E$, and $N_V$ that has not been considered before. Finally, this article provides some insight into the roles of viscosity and elasticity by illustrating how bubble growth and dissolution proceed from the inelastic limit to the inviscid limit as the elasticity of the system increases.

Finally, we note that it would be useful to compare theoretical predictions with experimental data, but it does not appear that a controlled study of the dissolution or growth of an isolated bubble in a polymeric liquid has been published. The study of Han and Yoo[10] provides bubble radius–time data for bubble growth during mold filling, but these experiments involve a somewhat uncertain pressure history and the possibility of significant bubble interactions. The lack of appropriate experimental data increases the importance of a theoretical analysis of the bubble growth or dissolution problem. The present theoretical predictions do appear reasonable from physical considerations. The presence of increased viscosity retards bubble growth or dissolution, whereas the introduction of elasticity appears to promote instantaneous expansion or contraction of a bubble, which overcomes the viscous retardation.

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