1. INTRODUCTION

The discovery that gas bubbles can be used as contrast agents in medical ultrasound examinations has significantly increased the interest in studying forced-oscillations of spherical gas bubbles.\(^1\)\(^-\)\(^3\) In this promising application, tiny gas bubbles are forced to undergo resonance when subjected to a properly-tuned acoustic field. The resonance phenomenon enables one to distinguish the echo produced by the contrast bubbles from that produced by the surrounding tissues.\(^1\)\(^-\)\(^3\) Unfortunately, free bubbles may oscillate chaotically under certain conditions thereby damaging body tissues.\(^4\) Another problem with free air bubbles is that they can easily dissolve and/or coalesce thereby reducing image quality.\(^5\)\(^,\)\(^6\) To avoid such undesirable side effects, in recent years encapsulated gas bubbles are increasingly replacing free bubbles as contrast agents. The elasticity of the shell can limit bubble’s growth to such an extent that it always remains within a tolerable range. Also, encapsulated bubbles usually contain nitrogen (or perfluorocarbons) which can significantly reduce the diffusion of gas to the surrounding liquid in order to give the bubble a longer lifetime. Not relying on their size, the working mechanism of encapsulated bubble is completely different from that of free bubbles. That is, encapsulated bubbles rely heavily on the backscattering of the ultrasound pulse for improving image quality. Alternatively, the generation of subharmonics, ultraharmonics, and second harmonics can also be used for increasing image contrast. The theoretical advantage of harmonics over the fundamental frequency is that only contrast agent microbubbles resonate with harmonic frequencies, while adjacent tissues do not resonate (or resonate insignificantly). This is particularly true for the second harmonics so much so that equipment are now commercially available to detect second harmonics in order to enhance image quality. As a matter of fact, second harmonic imaging has enabled imaging extremely small vessels (say, down to 40 microns) which would otherwise be missed with conventional methods.

Allen and Roy\(^7\)\(^,\)\(^8\) have theoretically shown that the viscoelastic behavior of physiological fluids can be responsible for the generation of second harmonics, at least when free bubbles are used as contrast agents. In a recent work dealing with free-bubble contrast agents, Ahmadpour et al\(^9\) have shown that second harmonics can also be generated for inelastic fluids provided that the fluid of interest is thixotropic. The present work can be regarded as an extension to their work in that it will be shown that even for encapsulated gas bubbles, the second harmonics can be generated provided that the medium surrounding the bubble is thixotropic. To show this, we are going to rely on one of the simplest thixotropic fluid models available, i.e., the Moore’s model.\(^10\) Our interest in this thixotropic fluid model stems mainly from its simplicity,

---

**Dynamics of Encapsulated Gas Bubbles Immersed in Thixotropic Fluids**

**M. JALALISENDI, M.A. RANJBAR, and K. SADEGHY**

*University of Tehran, College of Engineering, School of Mechanical Engineering. P.O. Box: 11155-4563, Tehran, Iran*

(Received: October 17, 2011)

In the present work, forced oscillations of a single, spherical, encapsulated gas bubble, immersed in an unbounded thixotropic liquid, is investigated numerically. Relying on the Moore’s model to represent the thixotropy of the liquid, and the Kelvin-Voigt model to represent the viscoelasticity of the enclosing shell, the modified integro-differential Rayleigh-Plesset equation is solved numerically using the Gauss-Laguerre Quadrature (GLQ) method. It is shown that the thixotropic behavior of the surrounding liquid plays a key role in the emergence of harmonics. It is also shown that the viscoelastic properties of the shell material and also its wall thickness can dramatically affect the rise of harmonics for encapsulated contrast bubbles used in medical sonography.

**Key Words:** Encapsulated Bubble / Thixotropic Fluid / Forced Oscillation / Contrast Agent
and also the fact that it encompasses all the basic facets of structural-based thixotropic fluid models. Another advantage of this thixotropic fluid model is that it represents inelastic thixotropic fluids only. Thus, it excludes complications which might arise from the fluid’s elasticity thereby making interpretation of numerical results much easier. To this should be added the fact that in recent years we have acquired some experience working with this simple but robust thixotropic fluid model.\textsuperscript{9,11,12} And, due to the qualitative nature of the present work, this thixotropic fluid model was deemed most useful for our simulations.

Having decided on the fluid model, one has to decide on the solid model to be used for representing the shell material. In the present work we are going to rely on one of the simplest viscoelastic solid models available, i.e., the Kelvin-Voigt model, to represent the shell material. The main advantage of this viscoelastic model is that it separates the contributions of the viscous and elastic behaviour of the shell material explicitly. To this should be added the fact that, this viscoelastic solid model has successfully been used by Church\textsuperscript{13} in his theoretical study of encapsulated gas bubbles immersed in Newtonian fluids [see Ref. 14 for a review on the performance of other viscoelastic models].

In the present work, we are going to rely on the Church’s formulations to simulate the behavior of encapsulated gas bubbles immersed in a thixotropic fluid obeying Moore’s model. The main objective of the present work is to see how thixotropic parameters representing structure breakdown and structure rebuild can affect the generation of harmonics in forced oscillation of encapsulated contrast bubbles. We are also interested in the role played by the viscoelastic properties of the shell material on the rise of such harmonics. To reach these objectives, we are going to start with presenting the Church’s formulation \textsuperscript{13} which is itself based on the general Rayleigh-Plesset equation. The Gauss-Laguerre Quadraure (GLQ) method will then be described briefly as the numerical method of solution to be used for solving the integro-differential equation governing bubble dynamics. We will then investigate the effect of fluid’s thixotropic parameters, and also the shell’s elasticity and thickness on the rise of second harmonics. The paper will be concluded with highlighting its major findings.

2. MATHEMATICAL FORMULATION

We consider a spherical cavity surrounded by a layer of a viscoelastic solid material and immersed in an unbounded liquid, as shown in Fig. 1.

The bubble, which is initially at rest, is set into motion by some external means (say, by exerting an acoustic pressure field). To come up with a set of tractable equations governing the bubble’s dynamics certain simplifying assumptions had to be made, chief among them are: i) there is no gravitational effect, ii) the motion always remains spherically symmetric, iii) the system is in isothermal state, iv) the liquid is incompressible, v) the shell material is incompressible, vi) the shell is very thin, vii) the trace of the stress tensor for both the liquid and the solid is zero, viii) the contribution of the perfect gas inside the bubble to the radial stress is negligible, ix) the interfacial tension at the gas side can be neglected, and x) the interfacial tension at the gas-shell side is negligible. Under these assumptions, the equation governing the dynamics of the shell and the liquid surrounding the encapsulated bubble (the so-called generalized Rayleigh-Plesset equation) can be obtained as [see Ref. 13 for the details of its derivation],

$$\rho (RR + \frac{3}{2}R^2) = p_o(t) - p_a(t) - \frac{2\Gamma}{R} + \int \frac{\tau_{rr,S}}{r} dr + \int \frac{\tau_{rr,L}}{r} dr$$

where \( \rho \) is the density, \( \Gamma \) is the shell-liquid interfacial tension, \( R(t) \) is the mean radius of the shell with \( R_0 \) being its initial value, \( R_i(t) \) is the radius of the inner wall, \( R_o(t) \) is the radius of the outer wall, \( p_o \) is the pressure of the gas inside the cavity, \( p_a(t) \) is the pressure at infinity, and \( \tau_{rr} \) is the radial stress. In this equation, the subscript “L” refers to the liquid with the subscript “S” referring to the solid. The encapsulated bubble, as shown in Fig. 1, can be set into motion when an acoustic pressure is exerted at infinity. The acoustic field is taken to be of the form: \( p_a(t) = p_b + p_A \sin \omega t \) where \( p_b \) is the ambient pressure, \( p_A \) is the amplitude of the acoustic field, and \( \omega \) is the angular frequency.

The two integrals appearing on the right-hand-side of Eq. 1 can be related to the deformation field provided that the constitutive equation (or, rheological model) of the

Fig. 1. Schematic showing an encapsulated gas bubble.
surrounding liquid and the solid material are known. As to the shell material, like Church\textsuperscript{33} we are going to assume that it is a viscoelastic solid which obeys the Kelvin-Voigt model; that is,

\[
\tau_{\alpha,s} = 2\mu_s \frac{\partial v_r}{\partial t} + 2G_s \frac{\partial u_s}{\partial t},
\]

(2)

where \(\mu_s\) is the shear viscosity of the shell material, and \(G_s\) is the shear modulus of the shell. In this equation \(v_r(r,t)\) is the radial velocity of particles comprising the shell with \(u(r,t)\) being their radial displacement. For shells made of incompressible materials, \(u(r,t)\) can be related to the bubble radius by,

\[
u(r,t) = \frac{R^2}{r}(R - R_0).
\]

(3)

Thus we have,

\[
3 \int_{r_e}^{r} \frac{\tau_{\alpha,s}}{r} \, dr = 3e \left( \tau_{\alpha,s} \right)_{r=r_e} = 12\epsilon \mu_s \frac{R^3}{n^2} + G_s \frac{R^3}{R} \left( \frac{R_0}{R} - \frac{1}{R} \right),
\]

(4)

where \(e\) is the shell thickness, and \(R_0\) is the initial radius. In this work, the shell thickness is assumed to remain virtually constant during bubble’s forced oscillations.

As to the liquid side, it is assumed that it is a thixotropic fluid which obeys obeys Moore’s model as its constitutive equation.\textsuperscript{30} In this particular rheological model, the stress tensor is related to the rate-of-deformation tensor by the Newtonian relationship \(\tau_{ij} = 2\mu_d \epsilon_{ij}\). But, unlike Newtonian fluids, for which the viscosity is constant at a given temperature, the viscosity of Moore fluids is assumed to depend on a structural parameter, \(\lambda\), which is itself time-dependent; that is,\textsuperscript{10}:

\[
\mu(t) = \mu_c + c\lambda(t),
\]

(5)

where \(\mu_c\) is the infinite-shear viscosity corresponding to complete structure build-up (i.e., \(\lambda = 0\)). In this equation the coefficient \(c\) is equal to \(\mu_0 - \mu_c\) where \(\mu_0\) is the zero-shear viscosity of the fluid corresponding to complete structure build-up (i.e., \(\lambda = 1\)). It needs to be mentioned that the difference between different structural models lies in the form of the kinetic equation adopted for the time evolution of the structural parameter. In the Moore’s model, the kinetic equation for the structural parameter is of the following form\textsuperscript{30},

\[
d\lambda/dt = a(1-\lambda) - b\sqrt{\Pi} \lambda,
\]

(6)

where “\(a\)” and “\(b\)” are positive material properties denoting structure build-up and structure break-down, respectively. In this equation, \(\Pi = d_t d_{th}\) is the second invariant of the deformation-rate tensor, \(d_t\). For purely-extensional deformation generated in the liquid surrounding the bubble during bubble’s forced oscillations, \(\sqrt{\Pi}\) can be interpreted as the rate of extension. Since, from continuity equation, the radial velocity is equal to \(v_r(r) = R\dot{R}/r^2\), the rate of extension can be obtained as \(\dot{R}/r = 2\sqrt{\Pi} [R^2 R_t/R]^{1/2}\).

As to the thixotropic parameters, \(a\) and \(b\), it should be noted that, “\(a\)” has the dimension of reciprocal time whereas “\(b\)” is dimensionless. Therefore, the ratio \(b/a\) can conveniently be taken as a characteristic time of the fluid. For thixotropic fluids, the characteristic time can conveniently be interpreted as the time needed by fluid elements for their viscosity change to occur. Therefore, it is anticipated that when this time constant is sufficiently large (say, larger than the characteristic time of the flow, i.e., \(1/\dot{\varepsilon}\)), thixotropic effects can have a significant influence on the flow characteristics. As will be shown shortly, this is indeed found to be true in our work (see Section 4).

From Eq. 5 one can conclude that under steady conditions, the structural parameter takes an equilibrium value equal to \(\lambda_c = a/(a+b)\). In addition, by rewriting Eq. 5 as \(d\lambda/dt = -(a+b)\lambda\), it can be seen that the rate equation is linear in \(\lambda\). As a result, \(\lambda\)-constant curves become straight lines passing through the origin revealing that for a given structure, the behavior is indeed Newtonian. It needs to be mentioned that in the Moore’s model, the greatest rate of build-up occurs when there is no structure left in the fluid. And, there is no build-up when there is complete structure. We also note that, for any value of the shear rate, there is no structural breakdown when there is no structure in the fluid and that the greatest rate of breakdown occurs when there is complete structure.

When a thixotropic fluid obeying Moore’s model is subjected to a shearing cycle in which the shear rate increases linearly with time to a maximum and then decreases at the same rate to zero (the so-called ramp test) the shear stress on the “up” curve is greater than the shear stress on the “down” curve.\textsuperscript{13} This can be attributed to a gradual breaking down of the fluid’s microstructures on the “up” curve which affects fluid’s response on the “down” curve. In practice, this behavior gives rise to the emergence of a hysteresis loop in the stress-shear rate relationship. On the other hand, if a fluid obeying Moore’s model is sheared at a constant shear rate, its viscosity decreases by the progress of time until it reaches an equilibrium value. To see this more clearly, one can integrate Eq. 5 at once to obtain,
\[ \lambda(t) = \lambda_0 - (\lambda_c - \lambda_0) \exp\left[\frac{-t}{\Lambda}\right] \quad (7) \]

where \( \lambda_0 \) is the initial value of the structural parameter, and \( \Lambda = 1/(\alpha + \beta \hat{e}) \) is the time constant (for a given \( \alpha, \beta \), and \( \hat{e} \)) of the fluid’s response. By inserting this equation into Eq. 5 one can easily see that, in steady shear, the viscosity of a thixotropic fluid obeying Moore’s model decreases exponentially in time, say from its initial value \( \mu_0 \) to an equilibrium value \( \mu_e (\geq \mu_0) \). Based on Eq. 6, this drop in viscosity is controlled by the time constant \( \Lambda \). Therefore, this time constant can be interpreted as a kind of relaxation time.

(One should note Moore’s model represents inelastic fluids only, so that this term should not be confused with memory effects.) It is easy to see that for Newtonian fluids (\( \alpha = b = 0 \), \( \lambda = \lambda_0 = 1 \), \( \mu = \mu_0 \)) the relaxation time is equal to infinity. Obviously, based on our formulations, the lower the relaxation time, the larger is the deviation from Brownian behaviour. That is to say that, fluids with smaller relaxation times should exhibit thixotropic effects more significantly (i.e., they forget their initial configuration, \( \lambda_0 \), more dramatically upon reaching to the equilibrium configuration, \( \lambda_e \)). By rewriting the relaxation time in the form of \( \Lambda = 1/(a + b \hat{e}) \), one can also see that for a given \( \hat{e} \), and a given “a” (which is related to the Brownian motions only), the relaxation time decreases by an increase in \( b/a \), i.e., the characteristic time of the fluid. And, this corroborates our previous notion that by an increase in \( b/a \), fluid’s obeying Moore’s model should exhibit thixotropic effects more significantly. Base on Moore’s model, the second integral in Eq. 1 is obtained as,

\[ 3 \int_{r_i}^{r_{\infty}} \tau_{\infty} \mu(\lambda) \frac{R^2 R}{r^4} dr = -12 \int_{r_i}^{R_i} \mu(\lambda) \frac{R^2 R}{r^4} dr. \quad (8) \]

The governing equation then becomes,

\[ \rho \left( \ddot{R} + \frac{3}{2} \dot{R}^2 \right) = p_{\infty}(\frac{R}{R_i})^{\eta} - p_0(\frac{R}{R_i})^{2 - 2\tau} \int_{r_i}^{R} \mu(\lambda) \frac{R^2 R}{r^4} dr + 12c \left[ \mu_0 \frac{R}{R_i} + G_s \frac{R^2}{R_i} \right] \]

where \( p_0 \) has been replaced by the relationship \( p_{\infty}(R/R_i)^{\eta} \) using perfect gas assumption with \( p_{\infty} \) being the initial gas pressure and “k” as its polytropic exponent. It is to be noted that, this equation is virtually the same equation as that derived by Church, with the only difference being that now the fluid’s viscosity is structure-dependent, \( \mu(\lambda) \).

Now, before proceeding with solving this equation numerically, like Zana and Leaf we transform the problem to the Lagrangian frame of reference so that the difficulty with tracking a moving boundary can be avoided. To that end, we substitute: \( y = r_i - R(t) \). The r-momentum equation then transforms to:

\[ \rho \left( \ddot{R} + \frac{3}{2} \dot{R}^2 \right) = p_{\infty}(\frac{R}{R_i})^{\eta} - (p_0 + p_\Lambda \sin \omega t) - \frac{2\Gamma}{R} - 4R^{\frac{3}{2}} \int_0^{\frac{\pi}{2}} \frac{\mu}{(y + R^2)^2} dy + 12c \left[ \mu_0 \frac{R}{R_i} + G_s \frac{R^2}{R_i} \right] \]

\[ (9) \]

The above equation together with all other equations pertinent to our fluid mechanics problem are made dimensionless by substituting:

\[ \hat{R} = \frac{R}{R_0}, \quad \hat{v} = \frac{v}{R_0}, \quad \hat{t} = \omega t, \quad \hat{\mu} = \frac{\mu}{\mu_0}, \quad \hat{\xi} = \frac{\mu_0}{\mu_0}, \quad \hat{e} = \frac{e}{R_0}, \quad \hat{G}_s = \frac{G_s}{\mu_0 \omega} \quad (11) \]

We also define viscosity and amplitude ratios by \( \xi = \frac{\mu_0}{\mu_\Lambda} \) and \( \delta = \frac{e}{R_0} \omega \) respectively. In dimensionless form (having dropped the “hat” sign above dimensionless variables for convenience) the governing equations become,

\[ \ddot{R} + \frac{3}{2} \dot{R}^2 = [(C_p + We)(\frac{1}{R})^{\eta} - C_s(1 + \delta \sin t)] - \frac{We}{(Re)_s} \frac{1}{(R)^2} \int_0^{\pi/2} \frac{\mu}{(y + R^2)^2} dy - 12c \left[ \frac{R}{(Re)_s} \left[ \frac{G_s}{R^4} + \frac{1}{2} (R - 1) \right] \right] \]

\[ (12) \]

\[ \frac{\mu}{\xi} + (1 - \xi) \frac{\lambda}{\lambda} \]

\[ (13) \]

\[ \frac{d\hat{a}}{dt} = -2\sqrt{3} \frac{b\lambda}{R^3} \left[ \frac{R^2 \dot{R}}{r^4} \right] + a(1 - \lambda) \]

\[ (14) \]

The dimensionless numbers appearing in the above equations are the liquid Reynolds numbers, \( (Re)_l \), the shell Reynolds number \( (Re)_s \), and the pressure coefficient, \( C_p \), and the liquid Weber number. They are defined by,

\[ (Re)_l = \frac{\rho_0 R^2 a^2}{\mu_0}; \quad (Re)_s = \frac{\rho_0 R^2 a^2}{\mu_s}; \quad C_p = \frac{p_0}{\rho_0^2 R_0^4}; \quad We = \frac{2\Gamma}{\rho_0^2 R_0^3}. \]

\[ (15) \]

The governing equations, Eqs. 12-14, are seen to constitute an initial-value system of ordinary differential equations. To be amenable to a numerical solution, initial values should be assigned to the pertinent parameters. In dimensionless form, the initial values required to close the problem are: \( R(0) = 1, \dot{R}(0) = 0, \lambda(0) = 1 \) Having decided on the initial
conditions, one can then proceed with solving these equations numerically. Due to the presence of the integral term in Eq. 12, solving this equation is not an easy task, at least using conventional ODE solvers. Thus, we have decided to rely on a new method developed recently by Amini and Sadeghy\textsuperscript{16} to tackle this term. The method is based on the Gauss–Laguerre Quadrature (GLQ) formulation which can calculate this integral in the interval \([0, \infty]\). The GLQ method conveniently subdivides the integration domain into segments of unequal lengths such that more points can be placed adjacent to the bubble’s wall \(\text{[see Ref. 16, for more details]}\). As a result, steep variation in the structural parameter, \(\lambda\), can better be resolved in the vicinity of the bubble. To ensure grid-independent results, we have found it necessary to use at least 200 nodes in all of our simulations.

### 3. RESULTS AND DISCUSSIONS

After verifying the code developed in this work using numerical data available for free bubbles \((e = 0)\) immersed in thixotropic fluids of the Moore’s type \(\text{[see, Ref. 9]}\) the code was used for simulating the forced oscillations of encapsulated bubbles immersed in the Moore fluid. Our main objective is to investigate the effect of the fluid’s thixotropic parameters \((a \text{ and } b)\) on the bubble’s response. We are also interested in investigating the importance of the viscosity ratio, \(\xi = \mu_L/\mu_0\), on the harmonics generation. The role played by the shell’s viscoelasticity and its wall thickness on the rise of harmonics are other objectives of the present work.

For our simulations to closely resemble real situations, we have tried to rely on physical data reported in the open literature for the material properties and ultrasound parameters pertinent to the field of contrast agent microbubbles.\textsuperscript{1-3)} Typical physical data used in the simulations are \textsuperscript{5-7)}:

\[1 < R_0 < 5 \mu m, \ 0.5 < \omega < 5 \text{ MHz}, \ 0.001 < \mu_k < 0.005 \text{ Pa.s}, \ 0.05 < \mu_s < 0.5 \text{ Pa.s}, \ 0 < G_s < 20 \text{ MPa}, \ \Gamma = 0.05 \text{ N/m}, \ \text{and } \rho = 1000 \text{ kg/m}^3.\] The range of dimensionless numbers are:

\[0.5 \text{ Re}_L < 50; \ 0.01 < \text{Re}_S < 1; \ 0.5 < \text{Cp} < 10; \ 0.1 < \text{We} < 5.\]

Figure 2 shows the effect of the fluid’s thixotropic parameters, \(a\) and \(b\), on the bubble’s response to acoustic pressure fields. The results presented in this figure are for a typical values of \(\text{Cp} = 1, \text{Re}_L = 1, \text{Re}_S = 0.02, \text{We} = 0.5, G_s = 1, \text{and } e = 0.001, \) \(\xi = 0.1, \text{and } \delta = 2\). We have decided to present data for different \(b/a\) values only. This is because our preliminary numerical results have shown that it is the ratio \(b/a\) which controls the bubble’s response not their absolute separate values. That is to say that, for the same \(b/a\) ratio, the same behavior is predicted to prevail in the bubble’s response. This is not surprising realizing the fact that this ratio is the characteristic time of our thixotropic fluid. As can be seen in Fig. 2, this ratio has a significant effect on the rise of harmonics in the bubble’s response. That is, while there is virtually no harmonics present at small \(b/a\) ratios \(\text{i.e.}, \text{by approaching the Newtonian extreme})\) harmonics and second harmonics start to emerge when this ratio is sufficiently large.

To explain this behavior, one should note that an increase in the characteristic time of the fluid means a more severe deviation from Newtonian behavior. Therefore, thixotropic effects are expected to manifest themselves more profoundly the larger the \(b/a\) ratio. At high \(b/a\) ratios, harmonics might be generated \(\text{even when the amplitude ratio is small})\) simply because the competition between the characteristic time of the fluid and the characteristic time of the flow \(\text{i.e., the inverse of the extension rate which is itself related to the frequency of the acoustic field through the equation of motion})\) adds severely to the nonlinearity of the dynamical system. As a result, we

![Figure 2](image_url)

**Fig. 2.** Effect of the thixotropic ratio, \(b/a\), on the bubble response: a) \(b/a = 1\), b) \(b/a = 0.2\), c) \(b/a = 0.001\).
will be witnessing harmonics which are normally absent for Newtonian fluids under the same set of acoustic parameters. (We would like to stress that harmonics can be observed even for Newtonian fluids provided that the amplitude of the driving acoustic force is sufficiently strong. Under these conditions, the motion ceases to be periodic and exhibits a chaotic behavior. Interestingly, the route to this chaotic behavior goes through a successive series of bifurcations to subharmonic oscillations of increasingly longer period [see, for example, Ref. 18].)

Figure 3 shows the effect of the viscosity ratio, $\zeta$, on the bubble’s response obtained at typical values of $b/a = 5$, $C_p = 3$, $Re_L = 0.5$, $Re_S = 0.02$, $We = 0.5$, $G_S = 1$, $e = 0.001$, and $\delta = 3.5$. The viscosity ratio, which is less than one for thixotropic fluids, can be regarded as a good index for the severity of the thixotropic behavior. As can be seen in Fig. 3, when this ratio approaches one (which corresponds to Newtonian fluids) the second harmonics becomes weaker and weaker. But, by a decrease in the viscosity ratio, the harmonics become stronger and stronger. To explain this behavior, one should note that a small viscosity ratio means that there is going to be a broad range of viscosities present all at the same time in the fluid system. These viscosities correspond to a wide range of diffusion times which can affect momentum transfer to such an extent that harmonics start to emerge.

Figure 4 shows the effect of the shell’s elasticity, $G_s$, on the bubble response. The results reported in this figure are obtained at typical values of $b/a = 20$, $C_p = 3$, $Re_L = 2$, $Re_S = 0.02$, $We = 0.5$, $\zeta = 0.1$, and $\delta = 2$. As can be seen in this figure, at small elastic moduli the bubble behaves quite chaotically (see Fig. 4a). But, by an increase in $G_s$ the elasticity of the shell becomes more dominant such that the response of the bubble becomes quite periodic exhibiting multiple harmonics (see Fig. 4b). However, if the shell is too elastic, the harmonics are completely wiped out from the bubble’s response (see Fig. 4c). Obviously, elasticity of the encapsulated layer has a significant effect on the response of contrast bubbles in thixotropic fluids so that it can be used as the controlling parameter. That is, choosing a shell material with appropriate elasticity not only does stabilize the bubble, but it also enhances the likelihood of

![Figure 3](image1.png)
![Figure 4](image2.png)

Fig. 3. Effect of the viscosity ratio, $\zeta$, on the bubble’s response obtained at:
(a) $\zeta = 0.05$, (b) $\zeta = 0.2$, (c) $\zeta = 0.8$.

Fig. 4. Effect of the shell’s elasticity, $G_s$, on the bubble response:
(a) $G_s = 0$, (b) $G_s = 20$, (c) $G_s = 60$. 
the generation of useful harmonics.

Figure 5 shows the effect of the shell Reynolds number, $R_{e_s}$, on the bubble’s response obtained at $b/a = 20$, $C_p = 1.5$, $R_{e_l} = 2$, $W_e = 0.5$, $G_s = 5$, $\xi = 0.1$, and $\delta = 2$. An increase in $R_{e_s}$ can be interpreted as a decrease in the shell’s viscosity, $\eta_s$, with the other parameters being fixed. As can be seen in this figure, at small Reynolds number (or large shell’s viscosity) the bubble behaves periodically exhibiting harmonics (see Fig. 5a). This behavior remains virtually intact by a further increase in the shell’s Reynolds number. However, at sufficiently high Reynolds numbers, the bubble behaves chaotically meaning that the shell’s viscosity cannot be too small.

Figure 6 shows the effect of the shell’s thickness, “$e$”, on the bubble response obtained at typical values of $b/a = 20$, $C_p = 4$, $R_{e_l} = 2$, $R_{e_s} = 0.02$, $W_e = 0.5$, $G_s = 10$, $\xi = 0.1$, and $\delta = 2$. Figure 6a, shows that a free bubble ($e = 0$) undergoes severe chaotic behavior for this set of parameters. As can be seen in Fig. 6b, adding a shell changes the bubble’s response completely from a chaotic behavior to a more periodic one so that harmonics are predicted to emerge. However, by increasing the shell thickness any further, the behavior of the bubble becomes periodic at the main frequency (see Fig. 6c). That is to say that, the harmonics disappear if the shell is too thick. This means that, much care needs to be exercised as to the selection of the shell’s thickness if the response is going to contain harmonics.

Figure 7 shows the effect of pressure amplitude ratio, $\delta$, on the bubble’s response obtained at typical values of $b/a = 5$, $C_p = 3$, $R_{e_l} = 0.5$, $R_{e_s} = 0.02$, $W_e = 0.5$, $e = 0.001$, $G_s = 1$, and $\zeta = 0.1$. As can be seen in this figure, this ratio has a dramatic effect on the bubble’s response and the harmonics picture. As a matter of fact, at sufficiently high amplitude ratios, the bubble behaves chaotically, exhibiting no harmonics whatsoever.

4. CONCLUDING REMARKS

Based on the results obtained in this work, one can conclude that the chaotic behavior observed for free bubbles in medical sonography can indeed be suppressed substantially through
the use of an encapsulating shell even in thixotropic fluids. But, for this to happen the shell should be of proper elasticity, viscosity and/or thickness. Also, the present work shows that for encapsulated bubbles surrounded by thixotropic fluids obeying Moore’s model, harmonics might be generated depending on the thixotropic parameters of the fluid, the properties of the shell, and the strength of the acoustic field. Interestingly, it is predicted that for thixotropic fluids it is the ratio of the breakup parameter to the breakdown parameter which is decisive in dictating the bubble response. The results presented in this work suggest that the for the generation of second harmonics in the bubble’s response, use can effectively be made of controlling the properties of the shell material. This is because normally the thixotropic parameters of the liquid surrounding the bubble cannot be tempered with—for physiological fluids, it is affected merely by illness and/or age of the patient.

The authors would like to acknowledge the financial support received from University of Tehran for conducting this research work under grant number 8106037/1/03. Special thanks are also due to the respectful reviewers for their constructive comments.

### Acknowledgement

The authors would like to acknowledge the financial support received from University of Tehran for conducting this research work under grant number 8106037/1/03. Special thanks are also due to the respectful reviewers for their constructive comments.

### Appendix:

For an encapsulated bubble growing or collapsing spherically in a liquid, the flow induced in the surrounding liquid and also the elongational deformation imposed to the shell material are purely radial so that, in spherical coordinate system, the equation of motion is the same for both the surrounding liquid and the encapsulating shell; that is [13],

\[
\rho \left( \frac{\partial^2 v_r}{\partial t^2} + v_r \frac{\partial v_r}{\partial t} \right) = -\frac{\partial \rho}{\partial t} + \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \tau_{rr} \right) - \frac{\tau_{00} + \tau_{02}}{r} \tag{A1}
\]

where \( \rho \) is the density, \( v_r \) is the radial velocity, \( p \) is the isotropic pressure, and \( \tau_{ij} \) is the stress tensor. This equation should be written once for the liquid (denoted by the subscript “L”) and the solid (denoted by the subscript “S”). But, before so doing, the above equation is simplified by assuming that the stress tensor is traceless for both the liquid and the shell material such that we have: \( \tau_{rr} = -\tau_{00} - \tau_{02} \). Further simplification arises from the notion that, thanks to spherical symmetry, we can write: \( \tau_{00} = \tau_{rr} \). The \( r \)-momentum equation then reduces to,

\[
\rho \left( \frac{\partial^2 v_r}{\partial t^2} + v_r \frac{\partial v_r}{\partial t} \right) = -\frac{\partial \rho}{\partial t} + \frac{\partial \tau_{rr}}{\partial t} + \frac{3 \tau_{rr}}{r} \tag{A2}
\]

The conservation of mass imposes a constraint on the radial velocity \( v_r \) for both the liquid elements and the solid particles; that is,

\[
v_r(r, t) = \frac{\dot{R}_i(t)}{r} \left( \frac{R_i(t)}{r} \right)^2, \tag{A3}
\]

where \( R_i(t) \) is the radius of the inner wall, \( \dot{R}_i(t) \) is the velocity of the inner interface, and \( r \) is the radial distance from the center of the bubble to a point within the shell layer or the liquid. Equation A3 can be integrated from \( r \) to infinity (first from \( R_i \) to \( R_2 \) using parameters appropriate for the solid, and then from \( R_2 \) to infinity using parameters appropriate for the liquid). Neglecting the contribution of the gas to the motion, and also making use of the boundary conditions which can be imposed on the normal stress at the inner and outer interfaces, we will obtain [see Ref. 13 for the details],

Acknowledgement

The authors would like to acknowledge the financial support received from University of Tehran for conducting this research work under grant number 8106037/1/03. Special thanks are also due to the respectful reviewers for their constructive comments.
where $\rho_0$ is the density of liquid, $\rho_s$ is the density of the solid, $p_0$ is the initial gas pressure, $R_{i0}$ is the initial radius of the inner interface, $k$ is the polytropic exponent $\sigma_i$ is the gas-shell interfacial tension, $\sigma_s$ (to be denoted by the symbol $\Gamma$ in the main text) is the shell-liquid interfacial tension, and $p_s$ is the pressure at infinity. This equation, first derived by Church$^{13}$, is often referred to as the generalized Rayleigh-Plesset equation (A4).

The second integral then becomes,

$$\int_0^\infty \frac{\tau_{nL}}{R} \, r \, dr = -4\mu_L \frac{R_2^3}{R_1^3}$$

where $\mu_L$ is the viscosity of the liquid. For thixotropic fluids, since viscosity depends on the structural parameter and the structural parameter is nonuniform in the flow domain, $\lambda(t)$, this integral just becomes,

$$\int_0^\infty \frac{\tau_{nL}}{R} \, r \, dr = -12\int_{R_1}^{R_2} \mu(\lambda) \frac{R_1^3}{r^4} \, dr$$

As to the second integral in Eq. A4, Church$^{13}$ assumed that the shell is a viscoelastic solid obeying the Kelvin-Voigt model, for which we have,

$$\tau_{n,s} = 2\mu_s \frac{\partial v_s}{\partial r} + 2G_s \frac{\partial u}{\partial r}$$

where $\mu_s$ represents the viscous behavior of the shell, $G_s$ is its shear modulus, $v_s(r,t)$ is the radial velocity of particles comprising the shell, and $u(r,t)$ is the radial displacement of the shell particles. The second integral then becomes,

$$\int_{R_1}^{R_2} \frac{\tau_{n,s}}{r} \, dr = -4 \left[ \frac{R_2^3 - R_1^3}{R_1 R_2^2} \right] \left[ G_s (R_1 - R_{i0}) + \mu_s \frac{R_1^3}{R_2^2} \right]$$

This equation is valid also for thick shells. Assuming that the shell is very thin, Doinikov et al$^5$ have shown that this equation reduces to,

$$\int_{R_1}^{R_2} \frac{\tau_{n,s}}{R} \, dr = \frac{3e}{R} \left[ \tau_{n,s} \right]_{R_1}^{R_2} = 12e \left[ \frac{\mu_s}{R^2} + \frac{G_s}{R} \frac{R_{i0}^3 - 1}{R_{i0}^2 - 1} \right]$$

where $e = R_{20} - R_{i0}$ is the shell thickness, with $R_{20}$ being the shell’s outer radius under equilibrium conditions. Assuming that the shell is thin-walled, the equation governing the dynamics of encapsulated bubbles immersed in a thixotropic liquid of the Moore’s type can be obtained as$^{17}$,

$$\rho \left( \frac{R^2 + \frac{3}{2} R^2}{R} \right) = \rho_0 \left( \frac{R_{20}^2}{R} \right)^{\frac{1}{k}} - p_s(t) \left( \frac{2R^2}{R} \right)$$

$$12 \int_{R_1}^{R_2} \mu(\lambda) \left[ \frac{R^2}{R} \right] \, dr + 12e \left[ \frac{\mu_s}{R^2} + \frac{G_s}{R} \frac{R_{i0}^3 - 1}{R_{i0}^2 - 1} \right]$$

REFERENCES