

Lagrangian formalism for computing oscillations of spherically symmetric encapsulated acoustic antibubbles

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Antibubbles are gas bubbles containing a liquid droplet core and, typically, a stabilising outer shell. It has been hypothesised that acoustically driven antibubbles can be used for active leakage detection from subsea production facilities. This paper treats the dynamics of spherically symmetric microscopic antibubbles, building on existing models of bubble dynamics. A more complete understanding of microbubble dynamics demands that the effects of the translational dynamics is included into the Rayleigh-Plesset equation, which has been the primary aim of this paper. Moreover, it is a goal of this paper to derive a theory that is not based on ad-hoc parameters due to the presence of a shell, but rather on material properties. To achieve a coupled set of differential equations describing the radial and translational dynamics of an antibubble, in this paper Lagrangian formalism is used, where a Rayleigh-Plesset-like equation allows for the shell to be modelled from first principles. Two shell models are adopted; one for a Newtonian fluid shell, and the other for a Maxwell fluid shell. In addition, a zero-thickness approximation of the encapsulation is presented for both models. The Newtonian fluid shell can be considered as a special case of the Maxwell fluid shell. The equations have been linearised and the natural and damped resonance frequencies have been presented for both shell models.

Keywords: Microbubbles, spatio-temporal bubble dynamics, Rayleigh-Plesset equation.

1. Introduction

Recently, it has been proposed to locate offshore hydrocarbon production facilities below the sea instead of at the surface [1]. The construction of subsea production facilities reduces operation costs, and thereby allows for the production of hydrocarbons at greater depths. Several of

the new production fields in the Northern Hemisphere are placed in Arctic climates. Therefore, transportation processes at low temperatures are becoming of increasing importance. However, under the extreme conditions in such regions, leakages in transportation pipelines may be hard to detect [2].

Recently, a full overview of acoustic leakage detection methods was published [3]. Because of the similarity in acoustic response from bubbles and from other subsea phenomena, it has been hypothesised that acoustically driven antibubbles can be used for active leakage detection from subsea production facilities [4]. Antibubbles are gas bubbles containing a liquid droplet core. Typically, antibubbles are encapsulated by a stabilising outer shell.

This paper treats the dynamics of spherically symmetric microscopic antibubbles, building on existing models of bubble dynamics. In recent years it has been suggested that a more complete understanding of microbubble dynamics demands that the effects of the translational dynamics is included into the Rayleigh-Plesset equation [5]. Moreover, it is a goal to derive a theory that does not include any *ad-hoc* shell parameters, but is rather based on material properties, *e.g.*, the shear viscosity and the shear modulus. This is of interest as *ad-hoc* parameters describing the shell are not general, but depend on, *inter alia*, the bubble resting radius.

To achieve a coupled set of differential equations describing the radial and translational dynamics of an antibubble, Lagrangian formalism is used, where a Rayleigh-Plesset-like equation allows for the shell to be modelled from first principles. Two shell models are adopted; one for a Newtonian fluid shell and one for a Maxwell fluid shell. In addition, a zero-thickness approximation of the encapsulation is presented for both models.

2. Theory

In Lagrangian formalism, a Lagrangian function $L = T - U$ is defined, where L is the Lagrangian function, T is the kinetic energy, and U is the potential energy. The Lagrangian equation (1) is given by

$$\frac{d}{dt} \frac{\partial L}{\partial \dot{q}_i} - \frac{\partial L}{\partial q_i} = - \frac{\partial F}{\partial \dot{q}_i}, \quad (1)$$

where F is the dissipative function which is expressed as a sum of the dissipating mechanisms such as the shear viscosity of the water and the shear viscosity of the shell, q_i is the generalised coordinate system, and the overdot indicates the first time derivative. Let us consider an antibubble as presented in Figure 1, where R_1 and R_2 are the instantaneous radii from the centre of the bubble to the two interfaces of the shell, and R_d is the radius of the droplet core inside the bubble. As the liquid droplet core can be considered incompressible, R_d is constant when the bubble undergoes oscillations and translation. The shell and the surrounding liquid are assumed incompressible, too. From these assumptions, L and F are found, and subsequently substituted into (1).

2.1. Kinetic energy

The kinetic energy T of the dynamic antibubble system is given by

$$T = \frac{1}{2} m_b \dot{x}^2 + T_L + T_S, \quad (2)$$

where T_L is the kinetic energy of the liquid surrounding the antibubble and T_S is the kinetic energy of the shell, and $m_b = \frac{4}{3}\pi R_d^3 \rho_L + \frac{4}{3}\pi (R_1^3 - R_d^3) \rho_g$ is the sum of the mass of the core

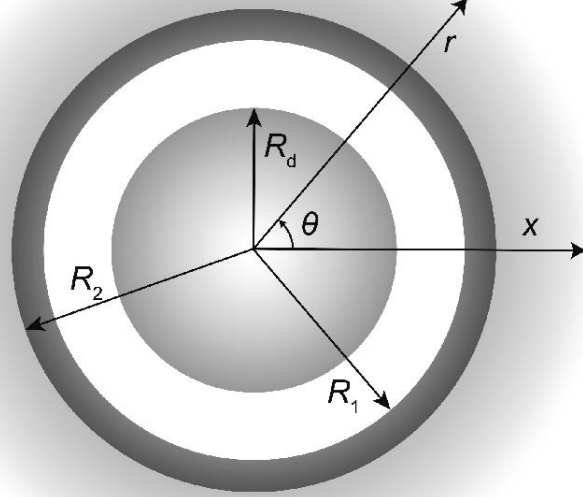


Fig. 1. Schematic of a fluid (opaque grey) containing an antibubble consisting of a droplet core (grey) of radius R_d , surrounded by a gas layer (white), and a thin shell (dark grey) of inner radius R_1 and outer radius R_2 . The antibubble is initially centred in the two respective coordinate systems used in this paper.

and the mass of the gas inside the bubble, in which ρ_L is the density of the liquid outside and inside the antibubble, R_{10} is the initial inner radius, and ρ_g is the density of the gas layer of the antibubble. The kinetic energy of an incompressible liquid is the following integral over volume V [6]:

$$T_L = \frac{\rho_L}{2} \int_V |\nabla\varphi|^2 dV, \quad (3)$$

where φ is the velocity potential of the liquid surrounding the bubble. We introduce a spherical coordinate system (r, θ, ϕ) that always has its origin in the centre of the antibubble. The centre of the antibubble is allowed to move exclusively in x -direction. The boundary condition at the surface $r = R_2$ is

$$\frac{\partial\varphi}{\partial r} = \dot{R}_2 + \dot{x} \cos\theta. \quad (4)$$

The velocity potential, which must satisfy Laplace's equation $\nabla^2\varphi = 0$, has the form

$$\varphi = \frac{a}{r} + \frac{b \cos\theta}{r^2}. \quad (5)$$

For the following functions a and b , (4) and (5) hold:

$$a(t) = -\dot{R}_1 R_1^2, \quad b(t) = -\frac{\dot{x} R_1^2}{2}. \quad (6)$$

Substituting (4) into (3), the kinetic energy of the incompressible surrounding fluid is given by

$$T_L = 2\pi\rho_L R_2^3 \left(\dot{R}_2^2 + \frac{\dot{x}^2}{6} \right). \quad (7)$$

When assuming an incompressible fluid, the damping due to acoustic radiation from the oscillating bubble cannot be accounted for. However, this problem can be overcome if we assume a weakly compressible fluid, as elegantly demonstrated in [7]. Considering the kinetic energy of the shell, the deformation of the shell is assumed negligible while the antibubble undergoes oscillations and translation. In this case, the volume of the shell V_S is constant, and the velocity inside the shell $v_S = R_1^2 \dot{R}_1 / r$. The assumption of an incompressible shell means that

$$R_2^3 - R_1^3 = R_{20}^3 - R_{10}^3, \quad (8)$$

and

$$R_1^2 \dot{R}_1 = R_2^2 \dot{R}_2, \quad (9)$$

where R_{20} is the initial outer resting radius. Thus, the kinetic energy of the incompressible shell can be written as

$$T_S = \frac{1}{2} \int_{V_S} \rho_S v_S^2 dV_S = 2\pi\rho_S R_1^3 \dot{R}_1^2 \left(1 - \frac{R_1}{R_2} \right), \quad (10)$$

where ρ_S is the density of the shell.

2.2. Potential energy

The potential energy U of the antibubble system is given by

$$U = U_g + U_\sigma + U_X, \quad (11)$$

where U_g is the potential energy of the gas inside the antibubble, U_σ is the potential energy owing to surface tensions at the gas–shell and shell–liquid interface, and U_X is the work done by the external pressure on the outer surface of the shell. Let us consider a pressure change in the surrounding fluid under adiabatic conditions. For an ideal gas, the potential energy of the gas inside the antibubble is

$$U_g = \frac{p_g V}{\gamma - 1}, \quad (12)$$

where p_g is the instantaneous pressure inside the antibubble, $V = \frac{4}{3}\pi (R_1^3 - R_d^3)$ is the instantaneous gas volume inside the antibubble, and γ is the polytropic exponent of the gas. Using that $p_g/p_{g0} = (V_0/V)^\gamma$, in which p_{g0} is the initial gas pressure inside the antibubble at rest and V_0 is the initial gas volume inside the antibubble at rest, the potential energy of the gas inside the antibubble can be written as

$$U_g = \frac{p_{g0} V_{g0}}{\gamma - 1} \left(\frac{R_{10}^3 - R_d^3}{R_1^3 - R_d^3} \right)^{(\gamma-1)}, \quad (13)$$

where

$$p_{g0} = p_0 + \frac{2\sigma_1}{R_{10}}, \quad (14)$$

where p_0 is the ambient pressure and σ_1 is the surface tension of the gas–shell interface. The potential energy owing to surface tensions at the gas–shell and shell–liquid interface is given by

$$U_\sigma = 4\pi R_1^2 \sigma_1 + 4\pi R_2^2 \sigma_2, \quad (15)$$

where σ_2 is the surface tension of the shell–liquid interface. The work done by the external pressure on the outer surface of the shell is given by

$$U_X = \frac{4\pi}{3} R_2^3 (p_0 + P), \quad (16)$$

where $P(x, t)$ is the driving pressure function.

2.3. Dissipative function

Energy is dissipated as the antibubble oscillates and translates in the surrounding fluid. The total dissipation is given by

$$F = F_L + F_S = \int_{V_L} f_L dV + \int_{V_S} f_S dV, \quad (17)$$

where F_L and F_S are the dissipative functions of the viscous fluid and the shell, respectively, f_L and f_S are the respective density functions of the dissipative functions, and V_L is the volume of the fluid outside the bubble. For the surrounding fluid, f_L is given by [8]

$$f_L = \eta_L \left(v_{ij} - \frac{1}{3} \delta_{ij} v_{kk} \right)^2 + \frac{1}{2} \zeta_L v_{kk}^2, \quad (18)$$

where η_L is the shear viscosity of the fluid, ζ_L is the bulk viscosity of the liquid, δ_{ij} is the Kronecker delta, and v_{ij} is the rate-of-strain tensor of the surrounding fluid, which is given by [5]

$$v_{ij} = \frac{1}{2} \left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right), \quad (19)$$

where $v_{i,j,k}$ is the liquid velocity. Assuming an incompressible surrounding fluid, $v_{kk} = \vec{\nabla} \cdot \vec{v} = 0$. Hence, (18) is simplified to

$$f_L = \eta_L (v_{ij})^2, \quad (20)$$

where v_{ij} is given by [5]

$$\begin{aligned} (v_{ij})^2 &= \frac{6R_2^4 \dot{R}_2^2}{r^6} + \frac{9R_2^6 \dot{x}^2}{8r^8} + \frac{9R_2^2 \dot{R}_2 \dot{x}}{r^4} \left(\frac{R_2}{r} - \frac{R_2^3}{r^3} \right) \cos \theta \\ &+ \frac{\dot{x}^2}{8} \left(\frac{27R_2^2}{r^4} - \frac{54R_2^4}{r^6} + \frac{18R_2^6}{r^8} \right) \cos^2 \theta. \end{aligned} \quad (21)$$

Substitution of (25) into (20) and subsequent integration over V_L , yields the dissipative function for the liquid

$$F_L = 8\pi \eta_L R_2 \dot{R}_2^2 + 3\pi \eta_L R_2 \dot{x}^2. \quad (22)$$

Finally, the dissipation function F_S for the viscous shell is the missing piece of the puzzle. For f_S we have [5, 8]

$$f_S = \eta_S (v_{ij})^2 = \eta_S \left[\left(\frac{\partial v_S}{\partial r} \right)^2 + \frac{2v_S^2}{r^2} \right], \quad (23)$$

where $v_S = R_1^2 \dot{R}_1 / r^2$ is the radial velocity in the shell. Now, integrating (23) over V_S results in

$$F_S = 8\pi \eta_S (R_{20}^3 - R_{10}^3) \frac{R_1 \dot{R}_1^2}{R_2^3}. \quad (24)$$

2.4. General Rayleigh-Plesset equation for a fluid shell

The expressions derived in the sections above for the respective kinetic energies, potential energies and the dissipative functions, or more specifically the translational kinetic energy of the antibubble (2), the kinetic energy of incompressible surrounding fluid (7), the kinetic energy of the shell (10), the potential energy of the gas (13), the potential energy of the surface-free energy (15), the potential energy from the work done on the antibubble by the external pressure on the outer surface of the bubble (16), the dissipative function for the liquid (22), and the dissipative function for the shell (24) are now combined to express L and F , respectively. Substituting the resulting expressions into (1), where R_1 and x are the generalised coordinates, whilst (8) and (9) are used to express R_2 in terms of R_1 , a set of coupled second order differential equations is obtained that describe the radial and translational dynamics of an antibubble with a fluid shell of finite shell thickness.

Studying the equations above, it can be recognized that modelling the shell as a fluid is not a complex operation to achieve. For a fluid shell, the dissipative function F_S for the shell is left undefined. It can now be shown that a set of equations, which are general, governing the radial and translation dynamics of an antibubble with a fluid shell can be obtained, albeit with some skill and cunning, eventually resulting in an equation set consisting of a Rayleigh-Plesset-like equation, and a translational equation:

$$\begin{aligned} R_1 \ddot{R}_1 \left[1 + \left(\frac{\rho_L - \rho_S}{\rho_S} \right) \frac{R_1}{R_2} \right] + \dot{R}_1^2 \left[\frac{3}{2} + \left(\frac{\rho_L - \rho_S}{\rho_S} \right) \left(\frac{4R_2^3 - R_1^3}{2R_2^3} \right) \frac{R_1}{R_2} \right] \\ = \frac{\rho_L \dot{x}^2}{\rho_S 4} + \frac{1}{\rho_S} \left[p_{g0} \left(\frac{R_{10}^3 - R_d^3}{R_1^3 - R_d^3} \right)^\gamma - \frac{2\sigma_1}{R_1} - \frac{2\sigma_2}{R_2} - p_0 - P(x, t) \right. \\ \left. - 4\eta_L \frac{R_1^2}{R_2^3} \dot{R}_1 + S \right], \end{aligned} \quad (25)$$

and

$$m_b \ddot{x} + \frac{2\pi}{3} \rho_L \frac{d}{dt} (R_2^3 \dot{x}) = -\frac{4\pi}{3} R_2^3 \frac{\partial}{\partial x} P(x, t) + F_d, \quad (26)$$

where F_d is the drag force, and S describes the radial stress in the shell, which can be represented by

$$S = 3 \int_{R_1}^{R_2} \frac{\tau_{rr}^{(S)}(r, t)}{r} dr, \quad (27)$$

where $\tau_{rr}^{(S)}$ is the radial tension function of the shell.

The rheological law suitable for the shell of a particular antibubble can now be applied. For a Newtonian fluid shell, the radial stress in the shell is given by

$$S = -4\eta_S \frac{\dot{R}_1 (R_{20}^3 - R_{10}^3)}{R_1 R_2^3}. \quad (28)$$

For a Maxwell fluid shell, the radial stress in the shell is given by

$$S = -4\eta_S \frac{D (R_{20}^3 - R_{10}^3)}{R_1^3 R_2^3}, \quad (29)$$

where $D(t) = -\lambda \dot{D}(t) + R_1^2 \dot{R}_1$, in which λ is the relaxation time.

2.5. Zero-thickness approximation

For antibubbles with a thin shell, *i.e.*, $R_S \equiv (R_2 - R_1) \ll R_1$, we can model the dynamics in first order without considering the correction for a finite shell. This can be done without a tangible loss of numerical accuracy. We may take the unrestrained radius equal to R_0 , $\rho_S \Rightarrow \rho_L$, $R_2 \Rightarrow R_1$, and $\sigma = \sigma_1 + \sigma_2$. Both the Newtonian and Maxwell shell models can now be represented with their respective zero-thickness approximation.

For a Newtonian fluid shell, the zero-thickness approximation is

$$R\ddot{R} + \frac{3}{2}\dot{R}^2 = \frac{\dot{x}}{4} + \frac{1}{\rho_L} \left[p_{g0} \left(\frac{R_0^3 - R_d^3}{R^3 - R_d^3} \right)^\gamma - \frac{2\sigma}{R} - p_0 - P(x, t) - 4\eta_L \frac{\dot{R}}{R} - 12\eta_S R_S \frac{\dot{R}}{R^2} \right], \quad (30)$$

whereas, for a Maxwell fluid shell, the zero-thickness approximation is

$$R\ddot{R} + \frac{3}{2}\dot{R}^2 = \frac{\dot{x}}{4} + \frac{1}{\rho_L} \left[p_{g0} \left(\frac{R_0^3 - R_d^3}{R^3 - R_d^3} \right)^\gamma - \frac{2\sigma}{R} - p_0 - P(x, t) - 4\eta_L \frac{\dot{R}}{R} - 12\eta_S R_S D \frac{\dot{R}}{R^4} \right]. \quad (31)$$

2.6. Linear analysis for an encapsulated antibubble

By linearisation of the respective models derived in the previous sections, we seek to understand underlying the mechanisms of oscillation. In this section, we illustrate the linearised versions' explicit expressions for the individual damping mechanisms, the linear natural resonance frequency, the second-order natural resonance frequency, and the damped linear resonance frequency for a finite thickness Maxwell shell. The damped linear resonance frequency is presented both with and without damping from reradiation.

For a small excursion $|x|$ of an antibubble where $|x| \ll R_{10}$, an analytical solution exists of the Rayleigh-Plesset-like equation (25), incorporating the radial stress of a Maxwell fluid shell (29). The small amplitude solution must satisfy

$$R_1 = R_{10} + x(t) \quad (32)$$

and

$$R_2 = R_{20} + \frac{R_{10}^2}{R_{20}^2} x(t) . \quad (33)$$

The coupling to the translational dynamics is also disregarded.

Now, the linearised version of $D(t)$ can be expressed as

$$\dot{D} + \frac{1}{\lambda} D = \frac{R_{10}^2}{\lambda} \dot{x} \quad (34)$$

Taking $P(t) = P_a \exp(i\omega t)$, in which P_a is the pressure amplitude, a solution of (34) has the form

$$D(t) = ax + b\dot{x} . \quad (35)$$

When substituting (35) into (34), the constants a and b are found:

$$a = \frac{\lambda \omega^2 R_{10}^2}{1 + (\lambda \omega)^2} , \quad b = \frac{R_{10}^2}{1 + (\lambda \omega)^2} . \quad (36)$$

After substitution of (32) and (34) into (25), the resulting linearised equation can be written as:

$$\ddot{x} + \delta_M \dot{x} + (\omega_0^M)^2 x = -\frac{P(t)}{\alpha \rho_S R_{10}} , \quad (37)$$

where δ_M is the sum of the respective damping terms $\delta_M = \delta_L + \delta_S^M$, in which δ_L represents the damping from the viscous surrounding fluid and δ_S^M represents the damping from the Maxwell fluid shell, ω_0^M is the linear resonance frequency, and α is a coefficient related to the density difference between the shell and the surrounding liquid

$$\alpha = 1 - \left(1 - \frac{\rho_L}{\rho_S}\right) \frac{R_{10}}{R_{20}} . \quad (38)$$

The damping from the surrounding fluid can be expressed as

$$\delta_L = \frac{4 \eta_L R_{10}}{\alpha \rho_S R_{20}^3} , \quad (39)$$

whereas the damping from the Maxwell fluid shell can be expressed as

$$\delta_S^M = \frac{4 \eta_S (R_{20}^3 - R_{10}^3)}{\alpha \rho_S R_{10}^2 R_{20}^3 [1 + (\lambda \omega)^2]} . \quad (40)$$

The linear resonance frequency is given by

$$(\omega_0^M)^2 = \omega_0^2 + \lambda \omega^2 \delta_S^M , \quad (41)$$

where ω_0 is the linear natural resonance frequency of an antibubble with a fluid shell:

$$\omega_0^2 = \frac{1}{\alpha \rho_S R_{10}^2} \left[p_{g0} \frac{3\gamma}{1 - \left(\frac{R_d}{R_0}\right)^3} - \frac{2\sigma_1}{R_{10}} - \frac{2\sigma_2 R_{10}^3}{R_{20}^4} \right] . \quad (42)$$

From (41), it can be observed that the linear resonance frequency is dependent on ω , which is slightly unconventional [9]. However, it is not of major concern, because a damped system resonates with the linear damped resonance frequency when excited. To assess the linear damped, *i.e.*, the “real”, resonance frequency of (37), one studies a solution of the equation given by

$$x(t) = A e^{i(\omega t + \psi)}, \quad (43)$$

in which the phase ψ between the radial excursion and the acoustic excitation is

$$\psi = \arctan \left[\frac{\omega \delta_M}{\omega^2 - (\omega_0^M)^2} \right], \quad (44)$$

and the amplitude A is

$$A = \frac{P_a Q}{\alpha \rho_S R_{10} \omega_0^2}, \quad (45)$$

in which the Q-value of the antibubble $Q(\omega)$ is

$$Q(\omega) = \frac{\omega_0^2}{\sqrt{[\omega^2 - (\omega_0^M)^2]^2 + \omega^2 \delta_M^2}}. \quad (46)$$

The maximum value of (46) is the linear damped resonance frequency, which can be computed numerically.

From the linear resonance frequency for a Maxwell fluid shell, we can find the resonance frequency for a Newtonian fluid shell ω_0^N . Let us take $\lambda = 0$, $\delta_N = \delta_M$, in which δ_N is the total damping for an antibubble with a Newtonian fluid shell. Knowing that the linear damped resonance frequency for a Newtonian fluid shell must satisfy $(\omega_0^N)^2 = \omega_0^2 - \delta_N^2/4$, it can be rewritten as

$$(\omega_0^N)^2 = \frac{1}{\alpha \rho_S R_{10}^2} \left[\frac{3\gamma p_{g0}}{1 - \left(\frac{R_d}{R_0}\right)^3} - \frac{2\sigma_1}{R_{10}} - \frac{2\sigma_2 R_{10}^3}{R_{20}^4} - \frac{4\eta_L^2 R_{10}^4}{\alpha \rho_S R_{20}^6} - \frac{4\eta_S^2 (R_{20}^3 - R_{10}^3)^2}{\alpha \rho_S R_{10}^2 R_{20}^6} \right]. \quad (47)$$

3. Example

An example of two radius–time curves of an oscillating antibubble is shown in Figure 2. The curves are numerical solutions of (30), computed with the ode45 algorithm of MATLAB® (The MathWorks, Inc., Natick, MA, USA). Droplet core sizes were chosen 40% and 80% of the resting radius, respectively.

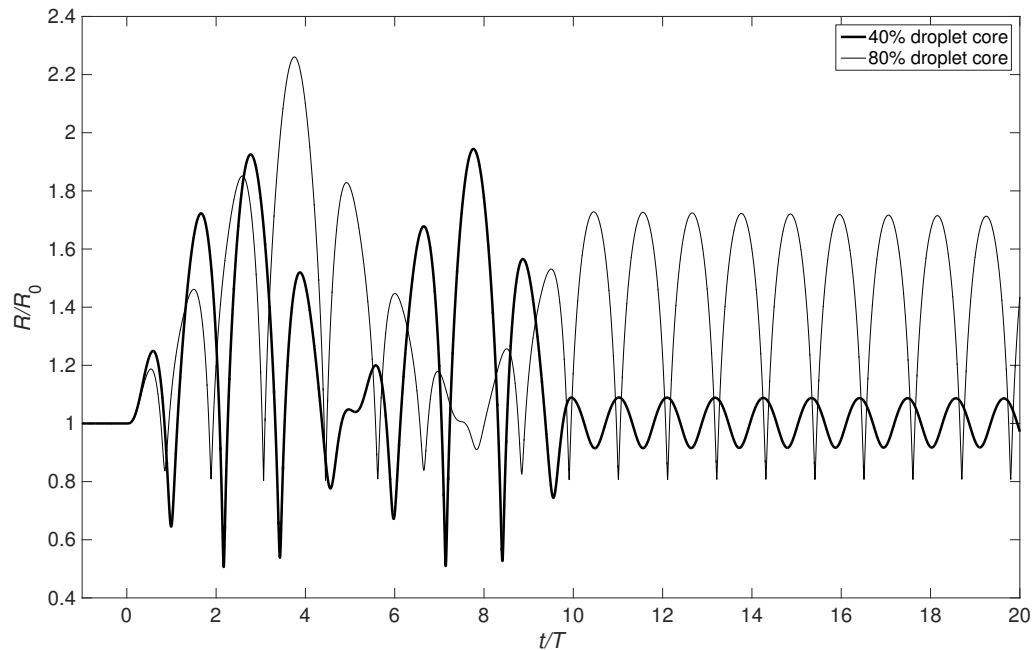


Fig. 2. Radius–time curves of an antibubble with a Newtonian shell.

The instantaneous radius has been normalised by the initial resting radius, and the time has been normalised by the period of the excitation pulse. An ambient pressure of $p_0 = 30$ atm, representing subsea conditions, was chosen. Other relevant parameters used were $P_a = 2$ MPa, $R_0 = 100 \mu\text{m}$, $R_S = 2$ nm, $\gamma = 1.4$, $\eta_L = 0.001$ Pa s, $\eta_S = 50$ Pa s, $\rho_L = 1054$ kg m $^{-3}$, $\sigma = 0.072$ N m $^{-1}$, and $\omega = 2\pi$ rad \times 200 kHz. Also, the coupling with (26) has been neglected here.

At the given excitation frequency, the antibubble excursions are clearly higher for the antibubble with the larger core. Especially, the asymmetric oscillation is worth noticing.

4. Conclusion

Using Lagrangian formalism, equations describing the spatio–temporal dynamics of antibubbles with a fluid shell have been derived, specifically for a Newtonian fluid shell and a Maxwell fluid shell. For both shell models, finite thickness shells and their zero-thickness approximations have been presented. The Newtonian fluid shell can be considered a special case of the Maxwell fluid shell. The equations have been linearised and the natural and damped resonance frequencies have been presented for both shell models.

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