



Article Mathematical Modeling of the Wave Dynamics of an Encapsulated Perfluorocarbon Droplet in a Viscoelastic Liquid

Damir A. Gubaidullin 🔍, Dilya D. Gubaidullina and Yuri V. Fedorov *🔘

Department of Continuum Mechanics, Institute of Mechanics and Engineering, FRC Kazan Scientific Center, Russian Academy of Sciences, Kazan 420111, Russia

* Correspondence: kopperfildd@ya.ru

Abstract: A mathematical model has been developed and a numerical study of vapor bubble growth as a result of acoustic evaporation of an encapsulated perfluorocarbon droplet in a viscoelastic liquid is presented. The viscoelasticity of the droplet shell and the carrier liquid is taken into account according to the Kelvin–Voigt rheological model. The problem is reduced to solving a system of ordinary differential equations for the radius and temperature of the bubble, the radius of the droplet and the shell together with the thermal conductivity equation for the internal liquid. Spatial discretization of the thermal conductivity equation is carried out using an implicit finite difference scheme. ODEs are solved by the fifth order Runge–Kutta method with an adaptive computational step. To check the correctness of the numerical calculation in a particular case, the theory has been compared with known experimental data. The influence of the shear modulus of the shell and the carrier liquid, and the shell thickness on the radial dynamics of a vapor bubble inside an encapsulated droplet in an external viscoelastic liquid is demonstrated.

Keywords: mathematical model; acoustic field; perfluorocarbon drop; vapor bubble; viscoelastic shell and liquid; phase transition

MSC: 76T10; 80A20

1. Introduction

Recently, the study of emulsions with phase transitions, which have the following feature, has been of great interest. Under the action of ultrasound, vapor bubbles are formed inside the liquid droplets. This process is known as acoustic droplet vaporization [1]. The use of these emulsions has great potential in biomedicine [2,3], because, unlike conventional gas microbubbles, liquid droplets can be quite small and can easily flow through thin capillaries. For example, when using echographic imaging, liquid droplets flowing through the vascular system of tumors can evaporate and be used as contrast agents to image the internal structure of the tumor [4]. Perfluorocarbon droplets, such as decafluorobutane C_4F_{10} , octafluoropropane C_3F_8 and dodecafluoropentane C_5F_{12} are usually used as liquid droplets [5–7]. The choice of these substances is due to the fact that they remain metastable at physiological temperature and cannot evaporate spontaneously. Their evaporation requires acoustic energy.

The paper [8] presents experimental results on the evaporation of octafluoropropane and decafluorobutane droplets of different sizes in water. In the case of complete evaporation of the droplet, data were also obtained on the radial oscillations of a pure vapor bubble. It is shown that an increase in the initial droplet size leads to a stronger growth of the vapor bubble, as well as to an increase in the amplitude of its oscillations. Experimental data on the evaporation of a droplet of dodecafluoropentane in water are presented in [9], and a mathematical model of a vapor bubble is also considered, taking into account gas diffusion in an infinite perfluorocarbon liquid. It is noted that, without taking into account



Citation: Gubaidullin, D.A.; Gubaidullina, D.D.; Fedorov, Y.V. Mathematical Modeling of the Wave Dynamics of an Encapsulated Perfluorocarbon Droplet in a Viscoelastic Liquid. *Mathematics* 2023, *11*, 1083. https://doi.org/10.3390/ math11051083

Academic Editor: Amir Gubaidullin

Received: 9 February 2023 Revised: 18 February 2023 Accepted: 20 February 2023 Published: 21 February 2023



Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). gas diffusion, the vapor bubble collapses after the first acoustic cycle. The presence of gas diffusion allows the vapor bubble to survive the first collapse and continue to grow, which is well confirmed by experimental data on the observed time scales. A generalization of the mathematical model to the case of a finite size of a perfluorocarbon droplet was carried out in [10-12]. The effect of acoustic parameters, liquid properties and droplet size on the process of acoustic evaporation has been studied. To prevent rapid dissolution of droplets in the carrier liquid, they are usually covered with a polymer shell [13]. In [14], the propagation of acoustic waves in a viscoelastic liquid with coated perfluorocarbon droplets containing vapor bubbles was studied. The influence of the shell of inclusions on the dependences of the phase velocity and attenuation coefficient on the frequency of perturbations is illustrated. A good agreement between the presented theory and the known experimental data is found. If phase transitions are not taken into account, then the mathematical model [14] transforms into the model [15], which describes the acoustics of a viscoelastic liquid with encapsulated gas bubbles. The propagation of weakly nonlinear waves in a liquid with encapsulated gas bubbles was considered in [16,17]. The viscoelastic shell obeyed the Kelvin–Voigt rheological model. Both low-frequency long waves and highfrequency short waves were considered. Particular attention is paid to the compressibility of the shell and liquid, as well as heat transfer between the gas bubble and the carrier liquid. It is shown that the compressibility of the bubble shell and the process of heat transfer increase the scattering of the wave. In particular, it has been found that the scattering of the wave due to the compressibility of the shell is about ten times greater than due to the compressibility of the liquid. Wang et al. [18] obtained experimental data on the reflection of an acoustic wave from a water layer with air bubbles. A significant effect of the volume content of gas on the dependence of the reflection coefficient on the frequency of perturbations has been established. In [19], the problem of the propagation of a high-intensity acoustic wave in a liquid layer with spherical gas bubbles was considered. A system of differential equations for acoustic pressure and bubble radius is written out. Based on the Lie approach, the authors analyzed the resulting system of nonlinear equations, and for the first time, several exact analytical solutions were obtained. Wang et al. [20] derived an equation for radial oscillations of a gas bubble near the boundary of a solid body. The influence of the initial bubble radius, the distance to the solid wall and the period of acoustic pressure on the evolution of the bubble were studied numerically. It is shown that the maximum bubble radius increases with increasing pressure period and distance to the solid boundary. Other publications on the dynamics of vapor–gas bubbles can be found in the review [21]. The wave dynamics of dodecafluoropentane on a solid wall was experimentally studied using high-speed visualization [22]. Droplets of micron size were subjected to ultrasonic treatment with an amplitude in the range from 0.6 to 3 MPa and a frequency of 5 MHz. It was found that the expansion of the volume due to the phase change promotes the separation of droplets from the solid wall. A quantitative estimation of the influence of the size of a droplet or bubble, as well as the amplitude of ultrasound on the dynamic motion of the inclusion has been carried out. In [23], a model for the generation of a vapor bubble inside a liquid droplet using a two-frequency configuration of focused ultrasound, which consists of continuous low-frequency ultrasound and a short high-frequency pulse, is presented. This made it possible to shorten the onset time for the formation of vapor nucleation center and to change the ratio of the length and width of the region of formation of nucleation centers at a lower energy cost. In contrast to [11,12], where the van der Waals equation of state was used in modeling the acoustic evaporation of perfluorocarbon droplets, the Redlich–Kwong equation of state was used in [24]. This made it possible to predict a much higher vapor generation velocity, which is in better agreement with the experimental results. In addition, it is shown that the properties of droplets have a very strong effect on the nucleation center formation threshold compared to the acoustic parameters. The group of Sojahrood extensively investigated the dynamics of encapsulated bubbles (including the free bubble case) through various aspects such as bifurcation structure [25,26], sub- and super-harmonic behaviors [27,28] and the mechanics of nonlinear power dissipation [29]. In this paper, we present a mathematical model and numerically simulate the growth and radial oscillations of a vapor bubble inside an encapsulated perfluorocarbon droplet in an external viscoelastic liquid. The model proposed here can be used both to describe laboratory experiments, where water is usually used as the ambient liquid, and to describe various applications where a more complex liquid can act as a carrier phase.

2. Mathematical Model

Let us consider an encapsulated droplet of liquid with a vapor bubble at the center inside an array of viscoelastic liquid (Figure 1).



Figure 1. Scheme of a droplet: *v*—vapor, *e*—liquid phase, *s*—shell and *l*—carrier liquid.

The equations of continuity and momentum in a spherical coordinate system in the presence of central symmetry and assuming the incompressibility of liquid phases and shell are written as [14]

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 u_v \rho_v \right) = -\frac{\partial \rho_v}{\partial t}, \quad r < R_1, \tag{1}$$

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2u_k\right) = 0, \quad r > R_1, \quad k = e, \ s, \ l, \tag{2}$$

$$\rho_k \left(\frac{\partial u_k}{\partial t} + u_k \frac{\partial u_k}{\partial r} \right) = -\frac{\partial p_k}{\partial r} + \frac{\partial \tau_{rr,k}}{\partial r} + \frac{3\tau_{rr,k}}{r}, \quad r > R_1, \quad k = e, \ s, \ l. \tag{3}$$

Here ρ is the density, r is the radial coordinate, t is the time, u is the radial velocity, p is the pressure and τ_{rr} are the normal stresses. The solution of Equation (2) is written as follows

$$u_k = \frac{R_1^2}{r^2} U_k, \quad k = e, \ s, \ l, \tag{4}$$

where U_k is the velocity of liquid at the inclusion boundary. Boundary conditions are written at the boundary R_1 with phase transitions and R_2 without phase transitions

$$p_{e}(R_{1}) - p_{v}(R_{1}) + \frac{2\sigma_{1}}{R_{1}} = \tau_{rr,e}(R_{1}) - \tau_{rr,v}(R_{1}) + J(u_{e}(R_{1}) - u_{v}(R_{1})),$$

$$J = \rho_{v}\left(\frac{dR_{1}}{dt} - U_{v}\right) = \rho_{e}\left(\frac{dR_{1}}{dt} - U_{e}\right),$$

$$p_{s}(R_{2}) - p_{e}(R_{2}) + \frac{2\sigma_{2}}{R_{2}} = \tau_{rr,s}(R_{2}) - \tau_{rr,e}(R_{2}), \quad u_{s}(R_{2}) = u_{e}(R_{2}) = \frac{dR_{2}}{dt},$$

$$p_{l}(R_{3}) - p_{s}(R_{3}) + \frac{2\sigma_{3}}{R_{3}} = \tau_{rr,l}(R_{3}) - \tau_{rr,s}(R_{3}), \quad u_{l}(R_{3}) = u_{s}(R_{3}) = \frac{dR_{3}}{dt}.$$
(5)

Here, σ is the surface tension coefficient, U_v is the velocity of vapor and J is the mass flow due to the phase change at the vapor/liquid interface. The normal stresses for a vapor and a liquid droplet are given in the form

$$\tau_{rr,v} = 2\mu_v \frac{\partial u_v}{\partial r} - \frac{2\mu_v}{3r^2} \frac{\partial r^2 u_v}{\partial r}, \quad \tau_{rr,e} = 2\mu_e \frac{\partial u_e}{\partial r}, \tag{6}$$

where μ is the dynamic viscosity. Since the viscosity of the vapor is less than the viscosity of the liquid, therefore $\tau_{rr,v} \ll \tau_{rr,e}$ and therefore, in the future, the value τ_v can be neglected.

For the carrier liquid and the shell of the droplet, we use the Kelvin–Voigt rheological model. Thus, the stress tensor component is related to the deformation tensor component by the following relation:

$$\tau_{rr} = 2G\varepsilon_{rr} + 2\mu\dot{\varepsilon}_{rr},\tag{7}$$

where *G* is the shear modulus and ε_{rr} is the deformation. A priori

$$\dot{\varepsilon}_{rr} = \frac{\partial u_s}{\partial r} \bigg|_{R_2} = \frac{\partial u_e}{\partial r} = -\frac{2R_2^2\dot{R}_2}{r^3} \Longrightarrow \varepsilon_{rr,s} = -\frac{2}{3r^3} \left(R_2^3 - R_{20}^3\right),$$

where R_{20} is the initial radius of the droplet. Then expression (7) takes the form

$$\tau_{rr,s} = -\frac{4R_2^2}{r^3} \left[\frac{G_s R_2}{3} \left(1 - \frac{R_{20}^3}{R_2^3} \right) + \mu_s \dot{R}_2 \right], \text{ and}$$

$$3 \int_{R_2}^{R_3} \frac{\tau_{rr,s}}{r} dr = -\frac{4G_s}{3} \left(1 - \frac{R_{20}^3}{R_2^3} \right) \left(1 - \frac{R_2^3}{R_3^3} \right) - \frac{4\mu_s R_2^2}{R_2^3} \left(1 - \frac{R_2^3}{R_3^3} \right) \dot{R}_2. \tag{8}$$

Similarly, we find the normal stress for the carrier liquid

$$\tau_{rr,l} = -\frac{4R_3^2}{r^3} \left[\frac{G_l R_3}{3} \left(1 - \frac{R_{30}^3}{R_3^3} \right) + \mu_l \dot{R}_3 \right], \text{ and}$$

$$3 \int_{R_3}^{\infty} \frac{\tau_{rr,l}}{r} dr = -\frac{4G_l}{3} \left(1 - \frac{R_{30}^3}{R_3^3} \right) - \frac{4\mu_l R_3^2}{R_3^3} \dot{R}_3.$$
(9)

Let us take into account the conditions of incompressibility of the shell and liquid, then we have

$$R_3^2 \dot{R}_3 = R_2^2 \dot{R}_2 = R_1^2 U_e. \tag{10}$$

The momentum Equation (3) is integrated from R_1 to infinity. In this case, the interval from R_1 to R_2 will correspond to the parameters of the liquid droplet, from R_2 to R_3 —to the parameters of the shell, from R_3 to infinity—to the parameters of the carrier liquid. Then, taking into account (4)–(10), after mathematical transformations, the equation of radial oscillations of the considered inclusion is derived

$$\begin{split} \rho_{d1} R_1 \dot{U}_e &- 0.5 \rho_{d2} U_e^2 + 2 \rho_{d1} \dot{R}_1 U_e = p_v - p_l - \frac{2\sigma_d}{R_1} + \left(\frac{1}{\rho_v} - \frac{1}{\rho_e}\right) J^2 - \frac{4\mu_d U_e}{R_1} \\ &- \frac{4G_s}{3} \left(1 - \frac{R_{20}^3}{R_2^3}\right) \left(1 - \frac{R_2^3}{R_3^3}\right) - \frac{4G_l}{3} \left(1 - \frac{R_{30}^3}{R_3^3}\right), \quad p_l = p_\infty + p_a, \\ \rho_{d1} &= \rho_e + (\rho_s - \rho_e) \frac{R_1}{R_2} + (\rho_l - \rho_s) \frac{R_1}{R_3}, \quad \rho_{d2} = \rho_e + (\rho_s - \rho_e) \frac{R_1^4}{R_2^4} + (\rho_l - \rho_s) \frac{R_1^4}{R_3^4}, \end{split}$$

$$\sigma_d = \sigma_1 + \sigma_2 \frac{R_1}{R_2} + \sigma_3 \frac{R_1}{R_3}, \quad \mu_d = \mu_e \left(1 - \frac{R_1^3}{R_2^3} \right) + \mu_s \left(\frac{R_1^3}{R_2^3} - \frac{R_1^3}{R_3^3} \right) + \mu_l \frac{R_1^3}{R_3^3}, \tag{11}$$

$$\dot{\mathsf{R}}_1 = U_e + \frac{J}{\rho_e}.\tag{12}$$

In Equation (11), the term p_a determines the external acoustic pressure. If we take J = 0, then Equation (11) coincides with the equation obtained earlier in [30]. Estimates made in [31] show that for perfluorocarbons, the thermal diffusion length is about 10 centimeters for perturbation frequencies from 1 to 10 MHz. As a result, we can assume that the temperature inside the vapor bubble is equal to the temperature on its surface. Thus, the pressure and temperature inside the bubble are determined from the Clausius–Clapeyron equation

Ì

$$p_v = p_\infty \exp\left[\frac{l}{B_v} \left(\frac{1}{T_{s0}} - \frac{1}{T_v}\right)\right],\tag{13}$$

where *l* is the specific vaporization heat of liquid, T_{s0} is the temperature at the saturation line, T_v is the vapor temperature inside the bubble, and B_v is the reduced gas constant. Next, it is necessary to obtain an equation that describes the evolution of temperature T_v . To achieve this, the mass (1) and energy [32] conservation equations are written for the vapor phase in a spherical coordinate system under the following assumptions: the vapor in the bubble obeys the ideal gas law, the vapor density is not constant in time, the pressure, density and temperature inside the bubble are uniform $(\partial p_v / \partial r = \partial \rho_v / \partial r = \partial T_v / \partial r = 0)$, the adiabaticity of the bubble is preserved (the heat flow q_v of the vapor is zero)

$$\rho_v c_v \frac{\partial T_v}{\partial t} = \frac{\partial p_v}{\partial t},\tag{14}$$

$$o_v = p_v / (B_v T_v), \tag{15}$$

where *c* is the specific heat. We multiply Equation (1) by $c_v T_v$ and add it to Equation (14), we obtain

$$\frac{\partial(\rho_v c_v T_v)}{\partial t} + \frac{c_v \rho_v T_v}{r^2} \frac{\partial}{\partial r} \left(r^2 u_v \right) - \frac{\partial p_v}{\partial t} = 0.$$
(16)

Using the equation of state (15), the relationship between the heat of an ideal gas, the gas constant and the adiabatic exponent, and integrating Equation (16), we find

$$u_v = -\frac{r}{3\gamma_v}\frac{\dot{p}_v}{p_v}.$$
(17)

Combining the obtained expressions (17), (5) as well as the Clausius–Clapeyron Equation (13), we arrive at a differential equation for temperature T_v

$$\dot{T}_v = \frac{3\gamma_v B_v T_v^2}{lR_1} \left[\left(\frac{1}{\rho_v} - \frac{1}{\rho_e} \right) J - U_e \right].$$
(18)

To close the system of Equations (10)–(13), (15) and (18), it is necessary to set the mass flow *J*. Since the temperature inside the vapor bubble is uniform and does not depend on the radial coordinate, we write down the thermal conductivity equations for the droplet

$$\frac{\partial T_e}{\partial t} + u_e \frac{\partial T_e}{\partial r} = \frac{D_e}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial T_e}{\partial r} \right) + \frac{12\mu_e}{\rho_e c_e} \frac{u_e^2}{r^2}, \quad D_e = \frac{\lambda_e}{\rho_e c_e}.$$
(19)

Here λ is the thermal conductivity coefficient, *D* is the thermal diffusivity. Boundary conditions are set

$$r = R_1:$$
 $T_e = T_v,$ $J = \frac{\lambda_e}{l} \frac{\partial T_e}{\partial r};$ $r = R_2:$ $T_e = T_{\infty}.$ (20)

As in [8], it is assumed here that the temperature of the shell and liquid surrounding the droplet are constant. Thus, to determine the mass flow J, it is necessary to solve the thermal conductivity equation in partial derivatives (19) with boundary conditions (20). Thus, we obtained a closed system of nonlinear differential Equations (10)–(13), (15) and (18)–(20), which determines the growth and radial oscillations of a vapor bubble in an encapsulated perfluorocarbon droplet located inside an array of viscoelastic liquid.

3. Initial Conditions and Numerical Realization

If we assume that the vapor bubble at the initial moment of time exists in the form of a nucleation center $R_1(0) = R_{10}$ and is at rest, then the velocity of the liquid/vapor interface satisfies the condition $\dot{R}_1(0) = 0$. The initial values of the radius of a droplet and a droplet with a shell are also determined by $R_2(0) = R_{20}$, $R_3(0) = R_{30} = R_{20} + r_{s0}$, where r_{s0} is the initial thickness of the shell. Thus, the pressure in the bubble at the initial moment of time has the form

$$p_v(0) = p_{v0} = p_{\infty} + \frac{2\sigma_1}{R_{10}} + \frac{2\sigma_2}{R_{20}} + \frac{2\sigma_3}{R_{30}},$$

and its temperature is found from the Clausius-Clapeyron equation

$$T_v(0) = T_{v0} = \left(\frac{1}{T_{s0}} - \frac{B_v}{l}\ln\frac{p_{v0}}{p_{\infty}}\right)^{-1}.$$

For the thermal conductivity Equation (20), the initial condition is given as $T_e(r, 0) = T_{\infty}$. The numerical solution of ordinary differential Equations (10)–(12), (18) is carried out by the Runge–Kutta method of the 5th order with automatic step selection. To eliminate computational problems in solving the thermal conductivity (19) connected with the motion of the bubble and droplet walls, by analogy with [32], the boundaries are fixed using dimensionless variables. For the internal liquid phase, the variable $x = (R_2 - r)/(R_2 - R_1)$ is introduced, in this case, if $r \in [R_1, R_2]$, then $x \in [0, 1]$. Then the partial derivatives will be written as

$$\frac{\partial}{\partial r} = -\frac{1}{h}\frac{\partial}{\partial x}, \quad \frac{\partial}{\partial t} = \frac{\partial}{\partial t} + \frac{\dot{R}_2 - hx}{h}\frac{\partial}{\partial x}, \quad h = R_2 - R_1,$$

and the thermal conductivity Equation (19) is rewritten in the form

$$\frac{\partial T_e}{\partial t} = V_{e2} \frac{\partial^2 T_e}{\partial x^2} + V_{e1} \frac{\partial T_e}{\partial x} + V_{e0},$$

$$V_{e2} = \frac{D_e}{h^2}, \quad V_{e1} = -\left[\frac{1}{h} \left[\dot{R}_2 - \dot{h}x - \frac{R_1^2 U_e}{(R_2 - hx)^2}\right] + \frac{2D_e}{h(R_2 - hx)}\right],$$

$$V_{e0} = \frac{12\mu_e}{\rho_e c_e} \frac{R_1^4 U_e^2}{(R_2 - hx)^6}.$$
(21)

Equation (21) is approximated using an implicit scheme with errors of the first order in time and of the second order in space. The spatial coordinate *x* is discretized by *N* points with a step of Δx . Thus, the partial derivatives of temperatures can be estimated using the following second-order finite-difference schemes

$$\frac{\partial T_e}{\partial x} = \frac{T_e|_{i+1} - T_e|_{i-1}}{2\Delta x} + O(\Delta x^2),$$
$$\frac{\partial^2 T_e}{\partial x^2} = \frac{T_e|_{i+1} - 2T_e|_i + T_e|_{i-1}}{\Delta x^2} + O(\Delta x^2), \quad i = 2, ..., N - 1.$$

As a result, we arrive at a system of three-point equations, which can be solved, for example, by the runout method. When the temperature field at the time step is found, the mass flow *J* is found by the following formula

$$J = \frac{\lambda_e}{l} \frac{3T_v - 4T_e|_2 + T_e|_3}{2\Delta x} + O(\Delta x^2).$$

4. Results of Calculations

To check the constructed mathematical model and the chosen numerical implementation in the particular case in Figure 2 there is comparison of the vapor bubble radius versus time with the available experimental data [8] obtained by evaporating an octafluoropropane droplet of various sizes in water. Water temperature is $T_{\infty} = 310$ K, pressure is $p_{\infty} = 10^5$ Pa. The thermophysical parameters of octafluoropropane were taken from [8,14]. The external acoustic pressure was given in the form

$$p_{a} = \begin{cases} -P_{a}\sin(2\pi f_{a}t), & 0 \le t \le N_{a}/f_{a}, \\ 0, & t > N_{a}/f_{a}, \end{cases}$$

where $P_a = 0.5 \times 10^6$ Pa, $f_a = 8 \times 10^6$ Hz and $N_a = 2$. The initial radius of the vapor bubble is $R_{10} = 10^{-7}$ m.



Figure 2. Comparison of the dependence of the vapor bubble radius on time with the experimental data [8] (the cross shows the moment of complete evaporation of the droplet): $1-R_{20} = 10^{-6}$ m and $2-R_{20} = 1.85 \times 10^{-6}$ m; $r_{s0} = 0$, $\rho_s = \rho_l$, $G_s = 0$ and $G_l = 0$.

For data 1, the authors of the experiment [8] proposed to use the viscosity of the carrier phase $\mu_l = 0.006$ Pa·s, and for data 2— $\mu_l = 0.009$ Pa·s. In the case when the droplet completely turned into vapor (the current droplet radius R_2 coincided with the radius of the vapor bubble R_1 and $R_2 < R_1$), it was assumed that the mass transfer intensity *J* was equal to zero. In this case, the already pure vapor bubble then performed radial oscillations. It can be seen that the larger the radius of the liquid droplet, the greater growth of the vapor bubble inside it. It should be noted that an increase in the droplet size by a factor of 1.85 leads to an approximately twofold increase in the time for complete evaporation of the liquid. On the whole, there is good agreement between theory and experiment. Thus, the mathematical model and its numerical implementation are capable of correctly reproducing the key physical processes underlying the evaporation dynamics of volatile contrast agents with phase transitions.

Let us now turn to the consideration of the dynamics of an encapsulated octafluoropropane droplet in water. Butyl rubber with a density of $\rho_s = 1475 \text{ kg/m}^3$ and a viscosity of $\mu_s = 0.99 \text{ Pa} \cdot \text{s}$ is considered as a shell of a droplet. Shell thickness is $r_{s0} = 10^{-7} \text{ m}$. Figure 3 shows the dependencies of the vapor bubble radius on time for various values of the shear modulus of the droplet shell.



Figure 3. Vapor bubble radius at different shear moduli of the droplet shell: $1-G_s = 10^7$, $2-3 \times 10^7$, $3-5 \times 10^7$ Pa; $\mu_l = 9 \times 10^{-3}$ Pa·s and $R_{20} = 1.85 \times 10^{-6}$ m (the cross shows the moment of complete evaporation of the droplet).

As expected, an increase in the shear modulus of the droplet shell leads to a decrease in the growth of the vapor bubble, as well as to a faster attenuation of its radial oscillations. With an increase in the elasticity of the shell, the evaporation time of the droplet increases. For the shear modulus value $G_s = 10^7$ Pa the droplet evaporation time is approximately $t \approx 0.7 \,\mu$ s, while for the $G_s = 5 \times 10^7$ Pa value the evaporation time increases to $t \approx 4.6 \,\mu$ s. Thus, in this case, an increase in the shear modulus by a factor of 5 leads to an increase in the evaporation time of the encapsulated droplet by a factor of 6.5.

Figure 4 shows the time dependencies of the vapor bubble radius for various shell thicknesses.



Figure 4. Radius of a vapor bubble for different values of the initial shell thickness: $1-r_{s0} = 10^{-7}$, $2-1.2 \times 10^{-7}$, $3-1.3 \times 10^{-7}$ m; $G_s = 5 \times 10^7$ Pa and $R_{20} = 1.85 \times 10^{-6}$ m (the cross shows the moment of complete evaporation of the droplet).

It can be seen that the thicker the shell, the smaller the radius of the vapor bubble and the faster its radial oscillations attenuate. As in the previous case, the droplet evaporation time also increases. Calculations show that taking into account the elasticity of the external liquid will not change the qualitative picture of the curves. Thus, a sufficiently elastic and thick shell of the inclusion, as well as an elastic external liquid, will restrain the radial motions of the vapor bubble.

5. Conclusions

A system of differential equations that determines the radial oscillations of a vapor bubble inside an encapsulated perfluorocarbon droplet located in an external viscoelastic liquid under acoustic pressure action has been obtained. Numerical calculations have been carried out. In a particular case, a good agreement between theoretical calculations and known experimental data has been found. It is shown that an increase in the shear modulus of the shell of a droplet or an elastic carrier liquid, as well as an increase in the thickness of the shell, lead to a decrease in the growth of a vapor bubble, as well as to a decrease in the amplitude of its oscillations. With the current parameters of the problem, it was found that if the droplet radius is increased by a factor of 1.85, then the time for complete evaporation of the liquid will approximately double. However, if the droplet is covered with a thin elastic shell, then, depending on the properties and parameters of the shell, it is possible to increase the time of complete evaporation of the droplet by 6–7 times. This result is important from a practical point of view. Finally, as with any study, the limitations of the present model must be recognized and considered in future work: (1) shell viscoelasticity from the Maxwell, Zener and Oldroyd models; (2) shell anisotropy [33]; (3) small nonsphericity of the vapor bubble and droplet; (4) the Redlich–Kwong equation of state for the vapor phase [24].

Author Contributions: Conceptualization, D.A.G. and Y.V.F.; methodology, Y.V.F.; software, D.A.G. and Y.V.F.; validation, Y.V.F.; formal analysis, Y.V.F.; investigation, D.A.G. and Y.V.F.; resources, D.A.G.; data curation, Y.V.F.; writing—original draft preparation, D.D.G. and Y.V.F.; writing—review and editing, D.D.G. and Y.V.F.; visualization, D.D.G. All authors have read and agreed to the published version of the manuscript.

Funding: This study was supported by the Russian Science Foundation, projects no. 21-71-00025 and 20-11-20070.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The data presented in this study are available upon request from the corresponding authors.

Conflicts of Interest: The authors declare no conflict of interest.

References

- Kripfgans, O.D.; Fowlkes, J.B.; Miller, D.L.; Eldevik, O.P.; Carson, P.L. Acoustic droplet vaporization for therapeutic and diagnostic applications. *Ultrasound Med. Biol.* 2000, 26, 1177–1189. [CrossRef] [PubMed]
- Sheeran, P.S.; Dayton, P.A. Phase-change contrast agents for imaging and therapy. *Curr. Pharm. Des.* 2012, 18, 2152–2165. [CrossRef] [PubMed]
- 3. Kee, A.L.J.; Teo, B.M. Biomedical applications of acoustically responsive phase shift nanodroplets: Current status and future directions. *Ultrason. Sonochem.* 2019, *56*, 37–45. [CrossRef]
- 4. Rapoport, N.; Gao, Z.; Kennedy, A. Multifunctional nanoparticles for combining ultrasonic tumor imaging and targeted chemotherapy. J. Natl Cancer Inst. 2007, 99, 1095–1106. [CrossRef] [PubMed]
- Sheeran, P.S.; Wong, V.P.; Luois, S.; McFarland, R.J.; Ross, W.D.; Feingold, S.; Matsunaga, T.O.; Dayton, P.A. Decafluorobutane as a phase-change contrast agent for low-energy extravascular ultrasonic imaging. *Ultrasound Med. Biol.* 2011, 37, 1518–1530. [CrossRef] [PubMed]
- Sheeran, P.S.; Luois, S.; Dayton, P.A.; Matsunaga, T.O. Formulation and acoustic studies of a new phase-shift agent for diagnostic and therapeutic ultrasound. *Langmuir* 2011, 27, 10412–10420. [CrossRef] [PubMed]
- Sheeran, P.S.; Luois, S.; Mullin, L.B.; Matsunaga, T.O.; Dayton, P.A. Design of ultrasonically-activatable nanoparticles using low boiling point perfluorocarbons. *Biomaterials* 2012, *33*, 3262–3269. [CrossRef]
- 8. Doinikov, A.A.; Sheeran, P.S.; Bouakaz, A.; Dayton, P.A. Vaporization dynamics of volatile perfluorocarbon droplets: A theoretical model and in vitro validation. *Med. Phys.* 2014, *41*, 102901. [CrossRef]
- 9. Shpak, O.; Stricker, L.; Versluis, M.; Lohse, D. The role of gas in ultrasonically driven vapor bubble growth. *Phys. Med. Biol.* 2013, 58, 2523–2535. [CrossRef]
- 10. Cho, S.; Son, G. A. level set method for bubble growth in acoustic droplet vaporization. *Int. Commun. Heat Mass Transf.* **2018**, *93*, 83–92. [CrossRef]

- 11. Cho, S.; Son, G. A. Numerical study of droplet vaporization under acoustic pulsing conditions. *J. Mech. Sci. Technol.* **2019**, *33*, 1673–1680. [CrossRef]
- 12. Park, S.; Son, G. Numerical investigation of acoustic vaporization threshold of microdroplets. *Ultrason. Sonochem.* **2021**, *71*, 105361. [CrossRef]
- 13. Rapoport, N. Phase-shift, stimuli-responsive perfluorocarbon nanodroplets for drug delivery to cancer. *Wiley Interdiscip. Rev. Nanomed. Nanobiotecnol.* **2012**, *4*, 492–510. [CrossRef] [PubMed]
- 14. Gubaidullin, D.A.; Panin, K.A.; Fedorov, Yu.V. Acoustics of a liquid with droplets covered by a shell in the presence of phase transitions. *Fluid Dyn.* **2022**, *57*, 459–468. [CrossRef]
- Gubaidullin, D.A.; Fedorov, Yu.V. Acoustics of a viscoelastic medium with encapsulated bubbles. J. Hydrodyn. 2021, 33, 55–62. [CrossRef]
- Kikuchi, Y.; Kanagawa, T.; Ayukai, T. Physico-mathematical model for multiple ultrasound-contrast-agent microbubbles encapsulated by a visco-elastic shell: Effect of shell compressibility on ultrasound attenuation. *Chem. Eng. Sci.* 2023, 269, 117541. [CrossRef]
- 17. Kanagawa, T.; Honda, M.; Kikuchi, Y. Nonlinear acoustic theory on flowing liquid containing multiple microbubbles coated by a compressible visco-elastic shell: Low and high frequency cases. *Phys. Fluids* **2023**, *35*, 023303. [CrossRef]
- 18. Wang, Y; Chen, D.; Cao, X.; He, X. Theoretical and experimental studies of acoustic reflection of bubbly liquid in multilayer media. *Appl. Sci.* **2022**, *12*, 12264. [CrossRef]
- 19. Gordoa, P.R.; Pickering, A. Ultrasonic waves in bubbly liquids: An analytic approach. Mathematics 2021, 9, 1309. [CrossRef]
- Wang, X.; Zhou, S.; Shan, Z.; Yin, M. Investigation of cavitation bubble dynamics considering pressure fluctuation induced by slap forces. *Mathematics* 2021, 9, 2064. [CrossRef]
- 21. Prosperetti, A. Vapor bubbles. Annu. Rev. Fluid Mech. 2017, 49, 221-248. [CrossRef]
- 22. Kwon, S.; Son, G. Experimental study of ultrasound-triggered vaporization of microdroplets on a wall. J. Mech. Sci. Technol. 2022, 36, 1329–1335. [CrossRef]
- Zhao, Y.; Qin, D.; Chen, J.; Hou, J.; Ilovitsh, T.; Wan, M.; Wu, L.; Feng, Y. On-demand regulation and enhancement of the nucleation in acoustic droplet vaporization using dual-frequency focused ultrasound. *Ultrason. Sonochem.* 2022, 90, 106224. [CrossRef]
- Qin, D.; Zou, Q.; Lei, S.; Wang, W.; Li, Z. Predicting initial nucleation events occurred in a metastable nanodroplet during acoustic droplet vaporization. *Ultrason. Sonochem.* 2021, 75, 105608. [CrossRef]
- 25. Sojahrood, A.J.; Haghi, H.; Kolios, M.C.; Karshafian, R. Nonlinear dynamics and bifurcation structure of ultrasonically excited lipid coated microbubbles. *Ultrason. Sonochem.* **2021**, *72*, 105405. [CrossRef]
- 26. Sojahrood, A.J.; Haghi, H.; Porter, T.M.; Karshafian, R.; Kolios, M.C. Experimental and numerical evidence of intensified non-linearity at the microscale: The lipid coated acoustic bubble. *Phys. Fluids* **2021**, *33*, 072006. [CrossRef]
- 27. Sojahrood, A.J.; Haghi, H.; Karshafian, R.; Kolios, M.C. Nonlinear dynamics of acoustic bubbles excited by their pressure dependent subharmonic resonance frequency: Influence of the pressure amplitude, frequency, encapsulation and multiple bubble interactions on oversaturation and enhancement of the subharmonic signal. *Nonlinear Dyn.* **2021**, *103*, 429.
- Sojahrood, A.J.; Haghi, H.; Rostam, S.N.; Kolios, M.C.; Karshafian, R. On the threshold of 1/2 order subharmonic emissions in the oscillations of ultrasonically excited bubbles. *Ultrasonics* 2021, 112, 106363. [CrossRef] [PubMed]
- Sojahrood, A.J.; Haghi, H.; Karshafian, R.; Kolios, M.C. Classification of the major nonlinear regimes of oscillations, oscillation properties, and mechanisms of wave energy dissipation in the nonlinear oscillations of coated and uncoated bubbles. *Phys. Fluids* 2021, 33, 016105. [CrossRef]
- 30. Gubaidullin, D.A.; Fedorov, Yu.V. Acoustic waves in a liquid with gas inclusions containing a liquid layer and a viscoelastic shell. *High Temp.* 2022, *in press.* [CrossRef]
- Lacour, T.; Brasier, T.; Coulouvrat, F. Ultimate fate of a dynamical bubble/droplet system following acoustic vaporization. *Phys. Fluids* 2020, 32, 051702. [CrossRef]
- 32. Hao, Y.; Prosperetti, A. The dynamics of vapor bubbles in acoustic pressure fields. Phys. Fluids 1999, 11, 2008. [CrossRef]
- Chabouh, G.; Dollet, B.; Quilliet, C.; Coupier, G. Spherical oscillations of encapsulated microbubbles: Effect of shell compressibility and anisotropy. J. Acoust. Soc. Am. 2021, 149, 1240–1257. [CrossRef] [PubMed]

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.