Linear oscillations of viscoelastic drops used for measuring the polymer retardation time

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Abstract

We study small-amplitude axisymmetric shape oscillations of a viscoelastic drop in a gas. The Oldroyd 8-constant model is used as the rheological constitutive equation of the liquid. The linearisation leads to a Jeffreys liquid with a frequency-dependent dynamic viscosity. The analysis of the time-dependent deformations of the drop surface yields the characteristic equation for the complex frequency. The damping rate and oscillation frequency are found to be determined by the viscous liquid behaviour and two time scales characterizing the elasticity of the liquid. It is shown that the polymer retardation time can be determined from the characteristic equation if the oscillation frequency and damping rate are measured. Preliminary data from experiments with acoustically levitated drops do not confirm the usual assumption applied in simulations that the ratio of the retardation and relaxation times ranged between 1/10 and 1/8.

Introduction

The deformations of a drop due to shape oscillations may influence transport processes across the drop surface, such as the evaporation and the aedrodynamic drag of the drop. Examples of technical systems where drop shape oscillations may be important are emulsions and fuel sprays. For their relevance for technical processes, drop oscillations have been under investigation since the times of Lord Rayleigh, who derived the angular frequency $\alpha_{m,0} \propto \sqrt{\sigma/\rho a^3}$ for linear oscillations of mode m of an inviscid drop with density ρ, radius a, and surface tension σ against the ambient vacuum [13]. Rayleigh's work was extended by Lamb, who included the influence of the drop viscosity and obtained expressions for the rate of decay of the oscillations in the limits of very high and very low drop viscosity [7]. Lamb also generalized Rayleigh's result by including the influence from a host medium with a non-negligible density [8]. He obtained a dependency of the angular oscillation frequency on a weighted mean of the two densities, $\alpha_m \propto \sqrt{\sigma/(m\rho_o + (m+1)\rho_i)a^3}$. The proportionality factors in the two relations above depend on the mode number m. The general (Newtonian) case of an oscillating viscous drop immersed in another liquid with non-negligible density and viscosity was analysed by Miller & Scriven [10]. From their work, a determinantal characteristic equation for the oscillating drop emerged.

The existing literature on drop oscillations is concentrated on Newtonian fluids. To date, the literature looking at oscillations of non-Newtonian, e.g., viscoelastic drops is still quite sparse. Bauer studied the oscillations of a viscoelastic drop under the influence of periodic temperature variations [1]. The author showed that, depending on the stress relaxation time of the liquid as compared to the oscillation period, the drop deformation is more or less influenced by the liquid elasticity. Khismatullin & Nadim presented an extensive theoretical analysis of linear shape oscillations of viscoelastic drops in a gas and found some interesting effects from the liquid elasticity. Those authors showed, e.g., the existence of a range of relaxation Deborah numbers where shape oscillations are due to the elasticity of the liquid, not due to surface tension [6].

In the present study we investigate theoretically the influence of viscoelasticity on small-amplitude shape oscillations of drops of polymer solutions. Our paper is organized as follows: in the following section we formulate the problem. Its solution in terms of velocity and pressure fields inside the oscillating drop, and the characteristic equation for the complex angular frequency following therefrom, is presented thereafter. Then we analyze and discuss the characteristic equation to describe various states of oscillation of viscoelastic drops caused by their relevant physical properties. Finally we propose a way to derive the retardation time of the polymers in the solutions from measurements of damped drop oscillations. In the last section we draw the conclusions from the results.

Formulation of the problem

The problem is governed by the equations of motion plus the viscoelastic rheological constitutive equation of the incompressible liquid. The equations are

$$\nabla \cdot \boldsymbol{v} = 0 \tag{1}$$

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for the continuity, and

$$\rho\left(\frac{\partial}{\partial t}+\boldsymbol{v}\cdot\boldsymbol{\nabla}\right)\boldsymbol{v}=-\boldsymbol{\nabla}\cdot\boldsymbol{\pi}+\rho\boldsymbol{f^B} \tag{2}$$

for the momentum balance, where the stress tensor $\boldsymbol{\pi}$ is defined as $\boldsymbol{\pi}=p\boldsymbol{\delta}-\boldsymbol{\tau}$. We specialise this equation for the case without body forces $\boldsymbol{f^B}$ and for small oscillation amplitudes as compared to the wavelength along a meridian of the drop. The resulting linearized equation of motion reads

$$\rho\frac{\partial\boldsymbol{v}}{\partial t}=-\boldsymbol{\nabla}\cdot(p\boldsymbol{\delta}-\boldsymbol{\tau}) \tag{3}$$

The dynamic behaviour of the liquid upon deformation is described by the rheological constitutive equation of the material. For the present viscoelastic liquid we use the Oldroyd 8-constant model given by the equation

$$\begin{aligned}\boldsymbol{\tau}+\lambda_1\frac{D\boldsymbol{\tau}}{Dt}+\frac{1}{2}\mu_0(tr\boldsymbol{\tau})\dot{\boldsymbol{\gamma}}-\frac{1}{2}\mu_1\{\boldsymbol{\tau}\cdot\dot{\boldsymbol{\gamma}}+\dot{\boldsymbol{\gamma}}\cdot\boldsymbol{\tau}\}+\frac{1}{2}\nu_1(\boldsymbol{\tau}:\dot{\boldsymbol{\gamma}})\boldsymbol{\delta}=\\ \eta_0\left[\dot{\boldsymbol{\gamma}}+\lambda_2\frac{D\dot{\boldsymbol{\gamma}}}{Dt}-\mu_2\{\dot{\boldsymbol{\gamma}}\cdot\dot{\boldsymbol{\gamma}}\}+\frac{1}{2}\nu_2(\dot{\boldsymbol{\gamma}}:\dot{\boldsymbol{\gamma}})\boldsymbol{\delta}\right]\end{aligned} \tag{4}$$

The parameters λ_1 and λ_2 are the stress relaxation and deformation retardation times of the liquid, respectively, and μ_0, μ_1, μ_2, ν_1, and ν_2 are further time constants of the model. Upon linearizaton, the Jeffreys material law is obtained. The equation is formulated with the symmetric and antimetric parts of the rate of deformation tensor

$$\dot{\boldsymbol{\gamma}}=\nabla\boldsymbol{v}+(\nabla\boldsymbol{v})^T \quad \text{and} \quad \boldsymbol{\omega}=\nabla\boldsymbol{v}-(\nabla\boldsymbol{v})^T \tag{5}$$

respectively. The irrotational derivatives of the stress tensor $\boldsymbol{\tau}$, and the rate of strain tensor $\dot{\boldsymbol{\gamma}}$, are defined as

$$\frac{D\boldsymbol{\tau}}{Dt}=\frac{\partial\boldsymbol{\tau}}{\partial t}+(\boldsymbol{v}\cdot\nabla)\boldsymbol{\tau}+\frac{1}{2}\{\boldsymbol{\omega}\cdot\boldsymbol{\tau}-\boldsymbol{\tau}\cdot\boldsymbol{\omega}\} \quad \text{and} \quad \frac{D\dot{\boldsymbol{\gamma}}}{Dt}=\frac{\partial\dot{\boldsymbol{\gamma}}}{\partial t}+(\boldsymbol{v}\cdot\nabla)\dot{\boldsymbol{\gamma}}+\frac{1}{2}\{\boldsymbol{\omega}\cdot\dot{\boldsymbol{\gamma}}-\dot{\boldsymbol{\gamma}}\cdot\boldsymbol{\omega}\} \tag{6}$$

The time dependency of the stress tensor $\boldsymbol{\tau}$ is given as

$$\boldsymbol{\tau}=\boldsymbol{T}(r,\theta)\cdot e^{-\alpha t} \tag{7}$$

where α is the complex angular frequency. The linearisation of this model leads to an equation for the deformation-induced stresses $\boldsymbol{\tau}$, which is characterised by a frequency-dependent viscosity as per

$$\boldsymbol{\tau}=\eta_0\frac{1-\alpha\lambda_2}{1-\alpha\lambda_1}\;\dot{\boldsymbol{\gamma}}:=\eta(\alpha)\;\dot{\boldsymbol{\gamma}} \tag{8}$$

The structure of the momentum equation is therefore formally identical to that of a Newtonian fluid with a frequency-dependent dynamic viscosity.

The continuity and momentum equations are formulated in spherical coordinates and solved subject to one kinematic and one dynamic boundary condition. A second dynamic boundary condition reveals the characteristic equation of the system. This will be presented in detail in the following.

Solution of the equations - the pressure and velocity fields

For analysing the flow in the oscillating drop we use the method of Levich and decompose the solenoidal flow field into an ideal-liquid and a real-liquid contribution (Helmholtz decomposition), assuming axial symmetry [9].

Solutions of the equations of change - ideal liquid

For the ideal liquid (subscript i), viscous stresses disappear from the momentum balance. The continuity equation and the linearised momentum equations in the radial (r) and the angular (θ) directions read

$$\frac{1}{r^2}\frac{\partial}{\partial r}(r^2v_{r,i})+\frac{1}{r\sin\theta}\frac{\partial}{\partial\theta}(v_{\theta,i}\sin\theta)=0 \tag{9}$$

$$\rho\frac{\partial v_{r,i}}{\partial t}=-\frac{\partial p_i}{\partial r}\quad,\quad\text{and}\quad\rho\frac{\partial v_{\theta,i}}{\partial t}=-\frac{1}{r}\frac{\partial p_i}{\partial\theta} \tag{10}$$

Describing the velocity field as a gradient of the scalar velocity potential ϕ turns the continuity equation into the Laplace equation for the velocity potential, which is solved with a separation ansatz to yield

$$\phi(r,\theta) = C_\phi r^m P_m(\cos\theta)\ e^{-\alpha t} \tag{11}$$

where the degree m of the Legendre polynomial $P_m(\cos\theta)$ plays the role of a mode number of the oscillation, representing the number of lobes of the shape along a meridian of the drop. The velocity components of the ideal fluid are

$$v_{r,i}\left(=\frac{\partial\phi}{\partial r}\right) = C_\phi m r^{m-1}\ \ P_m(\cos\theta)\ e^{-\alpha t} \tag{12}$$

$$v_{\theta,i}\left(=\frac{1}{r}\frac{\partial\phi}{\partial\theta}\right) = -C_\phi r^{m-1}\ \ P'_m(\cos\theta)\ \ \sin\theta\ e^{-\alpha t} \tag{13}$$

The pressure is obtained by integration of one of the two momentum equations (10) as

$$p = -\rho\frac{\partial\phi}{\partial t} = C_\phi \rho\alpha r^m P_m(\cos\theta)\ e^{-\alpha t} \tag{14}$$

The integration constant C_ϕ will be determined later.

Solutions of the equations of change - real liquid

The real liquid dynamic behaviour (subscript n) is determined by the deformation-induced stresses. The related continuity and momentum equations read

$$\frac{1}{r^2}\frac{\partial}{\partial r}(r^2 v_{r,n}) + \frac{1}{r\sin\theta}\frac{\partial}{\partial\theta}(v_{\theta,n}\sin\theta) = 0 \tag{15}$$

$$\rho\frac{\partial v_{r,n}}{\partial t} = \eta(\alpha)\left[\frac{1}{r^2}\frac{\partial^2}{\partial r^2}(r v_{r,n}) + \frac{1}{r^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial v_{r,n}}{\partial\theta}\right)\right] \tag{16}$$

$$\rho\frac{\partial v_{\theta,n}}{\partial t} = \eta(\alpha)\left[\frac{1}{r^2}\frac{\partial^2}{\partial r^2}\left(r^2\frac{\partial v_{\theta,n}}{\partial r}\right) + \frac{1}{r^2}\frac{\partial}{\partial\theta}\left(\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}(v_{\theta,n}\sin\theta)\right) + \frac{2}{r^2}\frac{\partial v_{r,n}}{\partial\theta}\right] \tag{17}$$

We formulate the velocity field by the Stokesian stream function as per

$$v_{r,n} = -\frac{1}{r^2\sin\theta}\frac{\partial\psi}{\partial\theta}\ ,\quad v_{\theta,n} = \frac{1}{r\sin\theta}\frac{\partial\psi}{\partial r} \tag{18}$$

Introducing these definitions into one of the momentum equations (16) or (17) yields the differential equation for the stream function

$$\frac{\rho}{\eta}\frac{\partial\psi}{\partial t} = \frac{\partial^2\psi}{\partial r^2} + \frac{\sin\theta}{r^2}\frac{\partial}{\partial\theta}\left(\frac{1}{\sin\theta}\frac{\partial\psi}{\partial\theta}\right) \tag{19}$$

Solving this equation with a separation ansatz we obtain

$$\psi(r,\theta,t) = C_\psi q r j_m(qr)\ \sin^2\theta\ P'_m(\cos\theta)\ e^{-\alpha t} \tag{20}$$

where we have denoted $q = \sqrt{\alpha\rho/\eta(\alpha)} = \sqrt{\alpha/\nu(\alpha)}$ and j_m is a spherical Bessel function of the first kind and order m. The prime denotes the derivative of a function w.r.t. its argument. From the stream function we obtain the non-ideal velocity components

$$v_{r,n}(\theta,r,t) = -C_\psi q^2\frac{j_m(qr)}{qr}m(m+1)P_m(\cos\theta)\ e^{-\alpha t},\ \text{ and }\ v_{\theta,n} = C_\psi q^2\left(j'_m + \frac{j_m}{qr}\right)\sin\theta P'_m(\cos\theta)\ e^{-\alpha t} \tag{21}$$

The velocity and pressure fields

The solutions of the equations of motion are the velocity and pressure fields

$$v_r\,(r,\theta,t) = v_{r,i} + v_{r,n} = \left[C_\phi m r^{m-1} - C_\psi q^2 \frac{j_m(qr)}{qr} m(m+1)\right] P_m(\cos\theta)\;\; e^{-\alpha t} \tag{22}$$

$$v_\theta\,(r,\theta,t) = v_{\theta,i} + v_{\theta,n} = \left[-C_\phi r^{m-1} + C_\psi q^2 \left(\frac{m+1}{qr} j_m - j_{m+1}\right)\right] \sin\theta\;\; P_m'(\cos\theta)\;\; e^{-\alpha t} \tag{23}$$

$$p\,(r,\theta,t) = -\rho\frac{\partial\phi}{\partial t} = C_\phi \rho\alpha r^m P_m(\cos\theta)\;\; e^{-\alpha t} \tag{24}$$

The integration constants in these solutions are determined by the boundary conditions. The kinematic boundary condition states that the radial rate of displacement of the deformed surface of the oscillating drop

$$r_s\,(\theta,t) = a + \epsilon_0 P_m(\cos\theta)\;\; e^{-\alpha t} \tag{25}$$

equals the radial velocity at $r = a$, i.e.,

$$v_r|_{r=a} = \frac{\partial r_s}{\partial t} \tag{26}$$

Here, a is the equilibrium radius of the drop and ϵ_0 the (small) deformation amplitude. This condition yields for the integration constants

$$C_\phi m a^{m-1} - C_\psi q^2 \frac{j_m(qa)}{qa} m(m+1) = -\epsilon_0\alpha \tag{27}$$

One dynamic boundary condition states that the shear stress vanishes at the drop surface, i.e., that

$$\left.\left(\frac{1}{r}\frac{\partial v_r}{\partial\theta} + r\frac{\partial v_\theta/r}{\partial r}\right)\right|_{r=a} = 0 \tag{28}$$

The relation between the integration constants resulting from this condition reads

$$-C_\psi \frac{q}{a}(2qa j_{m+1}(qa) - q^2a^2 j_m(qa)) + 2(m^2-1)\left[-C_\psi q\frac{j_m(qa)}{a} + \frac{C_\phi}{m+1}a^{m-1}\right] = 0 \tag{29}$$

The two integration constants emerge as

$$C_\psi = -\frac{2(m-1)\epsilon_0\alpha a}{mq(2qa j_{m+1} - q^2a^2 j_m)} \quad \text{and} \quad C_\phi = -\frac{\epsilon_0\alpha}{m a^{m-1}}\left[\frac{2\left(m^2-1\right)}{2qa j_{m+1}/j_m - q^2a^2} + 1\right] \tag{30}$$

This solution is formally identical with the results of Chandrasekhar and Khismatullin & Nadim [3, 6].

The characteristic equation for the complex angular frequency

The characteristic equation for the complex frequency α is found from the dynamic boundary condition for the normal stress which must vanish at the drop surface. With the capillary pressure $p_0 = 2\sigma/a$ inside the drop in its undeformed state and the capillary pressure in the deformed droplet [8]

$$p_\sigma = \frac{\sigma}{a}\left[2 + (m-1)(m+2)\frac{\epsilon_0}{a}P_m(\cos\theta)\;\; e^{-\alpha t}\right] \tag{31}$$

the requirement of vanishing normal stress at the interface reads

$$p_\sigma + \tau_{rr} = p_0 + p, \;\text{ i.e., }\; \frac{\sigma}{a}\left[2 + (m-1)(m+2)\frac{\epsilon_0}{a}P_m(\cos\theta)\;\; e^{-\alpha t}\right] = \frac{2\sigma}{a} + C_\phi\rho\alpha r^m P_m(\cos\theta)\;\; e^{-\alpha t} - 2\eta(\alpha)\frac{\partial v_r}{\partial r} \tag{32}$$

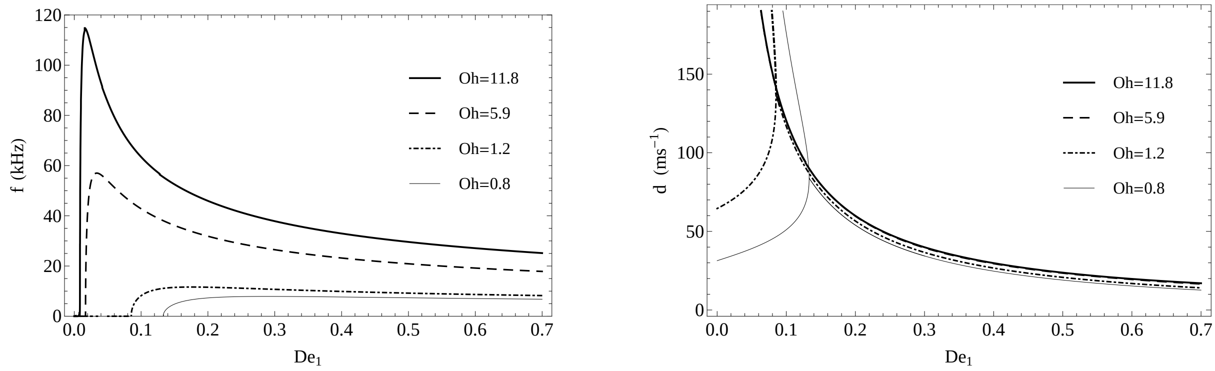

Figure 1. Left frequency and right damping rate as functions of the relaxation Deborah number De_1 for $\alpha_{m,0} = 24.17kHz$ with $De_2 = 0$.

where p is the pressure and τ_{rr} the viscoelastic normal stress in the radial direction due to the oscillations [3]. Denoting the angular frequency of small oscillations of an inviscid drop in the mode m by $\alpha_{m,0}$, which is given by the equation $\alpha_{m,0} = \sqrt{\sigma/\rho a^3}\sqrt{m(m-1)(m+2)}$ [13] and using the expressions (30) for the two integration constants, we may write the normal stress boundary condition (32) as

$$\frac{\alpha_{m,0}^2}{\alpha^2} = \frac{2(m^2-1)}{q^2a^2 - 2qaj_{m+1}(qa)/j_m(qa)} - 1 + \frac{2m(m-1)}{q^2a^2}\left[1 + \frac{2(m+1)j_{m+1}(qa)/j_m(qa)}{2j_{m+1}(qa)/j_m(qa) - qa}\right] \quad (33)$$

This is the characteristic equation for the complex angular oscillation frequency α of the drop. The equation is identical to the results of Lamb and Chandrasekhar [7, 3]. In the present case of a viscoelastic liquid, however, the kinematic viscosity ν, which appears in the argument qa of the spherical Bessel functions, is a function of the oscillation frequency α. We now analyse the behaviour of the oscillating drop by solving this equation.

Analysis of the characteristic equation for a viscoelastic drop

For solving the characteristic equation (33), we introduce $\nu(\alpha)$ resulting from the linearised material law (8). We use the definitions

$$y = \alpha/\alpha_{m,0}; \quad Oh = \eta_0/\sqrt{\sigma a \rho}; \quad De_1 = \alpha_{m,0}\lambda_1; \quad De_2 = \alpha_{m,0}\lambda_2 \quad (34)$$

which enter the argument qa of the spherical Bessel functions as per

$$q^2a^2 = \sqrt{m(m-1)(m+2)}\frac{y}{Oh}\frac{1-yDe_1}{1-yDe_2} \quad (35)$$

For the present paper we restrict our analysis to quadrupole oscillations, where $m = 2$. Since α occurs in the argument of the spherical Bessel functions, it is not possible to solve the characteristic equation analytically. For a numerical analysis of (33) we make use of the computer algebra software MATHEMATICA.

Before presenting our results, we validate our MATHEMATICA routine by reproducing some results of [6]. For a fluid characterised by $\rho = 10^3$ kg/m^3, $\sigma = 0.073$ N/m, and the drop radius $a = 0.1$ mm, we obtain the Rayleigh frequency $\alpha_{2,0} \approx 24.17$ kHz. The deformation retardation time λ_2 is set to zero, as in [6]. Varying the Ohnesorge number as a parameter, we reproduce Figure 4 of [6], which displays the dimensional frequency $f = [Im(y) \cdot \alpha_{m,0}]/(2\pi)$ and the dimensional damping rate $d = Re(y) \cdot \alpha_{m,0}$ as functions of the relaxation Deborah number De_1 with $De_2 = 0$. Figures 1 left and right display these results. As the parameter in this analysis, Khismatullin & Nadim used a Reynolds number defined as $Re = a^2\alpha_{m,0}\rho/\eta_0$, which we rather interpret as an Ohnesorge number, since it may be interpreted as a ratio of a viscous and a capillary time scale [6]. The identity $Re = \sqrt{m(m-1)(m+2)}/Oh$ holds. Therefore, for our case with $m = 2$, e.g., $Oh = 11.8$ is equivalent to $Re = 0.24$. Khismatullin & Nadim pointed out that there exists a critical relaxation Deborah number De_1^*, which marks the limit between the aperiodic mode of decay and the "real" oscillation mode. As shown in Figure 1 left, this critical Deborah number increases with decreasing Ohnesorge number. We can also see a change in the damping rate (Figure 1 right). While it increases with the Deborah number for the aperiodic cases, it decreases with further increasing elasticity. So the turn from the aperiodic mode to the oscillation mode is seen in the damping

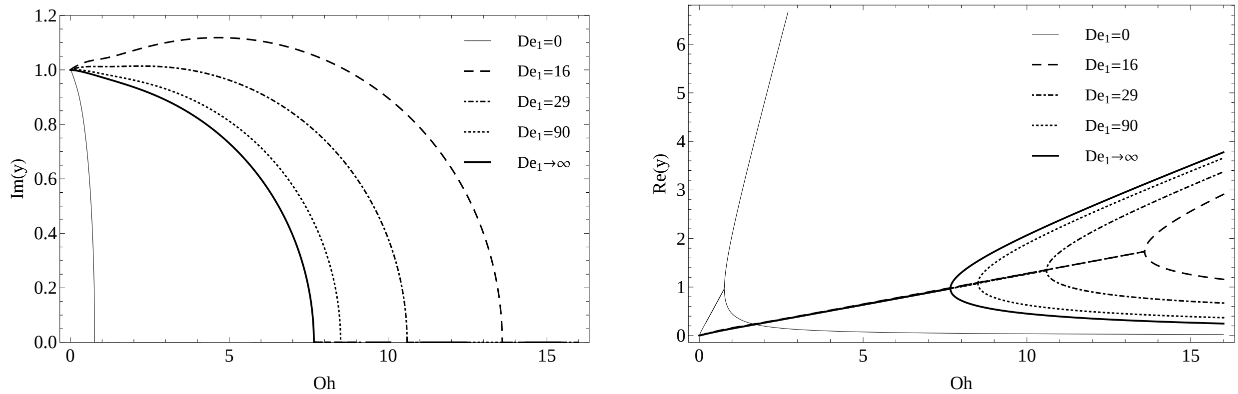

Figure 2. Non-dimensional (left) frequency and (right) damping rate as functions of the Ohnesorge number with $De_2 = De_1/10$.

rate as well, and the rate of decay of deformations assumes lower values for high supercritical Deborah numbers than in the aperiodic cases.

We now further analyse the characteristic equation (33). For this purpose, the retardation time λ_2 of the fluid must be given a value. This time scale is not readily determined [5]. What we know, however, is that the relation $\lambda_2 < \lambda_1$ must hold in order that the sign of the extra stress is correct [2]. In the literature, values for λ_2/λ_1 of $1/8$ and $1/10$ are commonly used, rather for historical than for physical reasons [5, 4, 12]. For our analysis we set $De_2/De_1 = 1/10$ for the moment. Our ultimate aim, however, is to propose a method to determine λ_2 from damped drop oscillation measurements. We are interested in the frequency and the damping rate as functions of the Ohnesorge number with the relaxation Deborah number De_1 as a parameter (Figures 2 left and right). For the inelastic case (De_1 = 0) we see that the non-dimensional frequency decreases with increasing Ohnesorge number. This corresponds to the result that, e.g., an increase of the viscosity reduces the frequency [11]. For supercritical Ohnesorge numbers greater than $Oh_0^* \approx 0.8$, where the subscript 0 denotes the (in this case vanishing) value of De_1, aperiodic modes of decay occur.

Moderate elasticity (De_1 > 0) makes the value of Oh^* increase and, therefore, widens the interval of Ohnesorge numbers where shape oscillations exist. It is important to mention, however, that increasing elasticity narrows this interval. E.g., while $Oh_{16}^* \approx 13.5$, we find that $Oh_{29}^* \approx 10.5$. It is, however, not possible to reach the critical Ohnesorge number of the purely viscous case as De_1 increases. Such a convergence behaviour can be expected, since the curves approach the purely viscous one with increasing relaxation Deborah number, but at a higher viscosity. Considering Oh^* as a function of De_1, we can say that

$$\lim_{De_1 \to \infty} Oh^* \approx 7.6$$

This is about 10 times the value observed for the Newtonian case, which is due to the influence of the retardation Deborah number De_2 on the Newtonian dynamic viscosity.

Figure 2 right shows the behaviour of the damping rate. For $Oh < Oh^*$, the dependency on Oh is linear, and for $Oh \gg Oh^*$ it is approximately linear, as found also by Prosperetti for the Newtonian case [11]. The state of the critical Ohnesorge number is seen here as well. The straight line seen for $Oh < Oh^*$ bends in a range of transition in a narrow range of Oh numbers and converges to another straight line. The asymptotics are parallel for all Deborah numbers investigated here. For zero Ohnesorge number, i.e. for the inviscid case, the damping rate is zero, as expected. The bifurcation of the damping rate at the state of onset of aperiodic modes was found by Chandrasekhar and Prosperetti also [3, 11]. The inelastic curve in Fig. 2 right is identical with the results by Prosperetti.

Determination of the polymer retardation time from damped drop oscillations

In the above discussions, the polymer deformation retardation time λ_2 was set to 1/10 of the relaxation time λ_1. While the latter may be determined experimentally [14], the former is far more difficult to get. The characteristic equation of the oscillating drop (33) involves both time scales, and it is worth an investigation whether the equation could serve for deducing the value of λ_2 and the viscosity η_0 if all the other quantities are given. One prerequisite for this is that both the frequency and the damping rate of oscillations of a drop of the polymeric liquid must

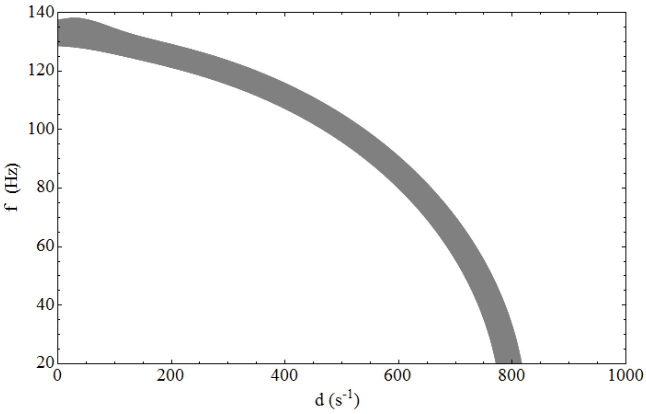

Figure 3. Ranges of frequencies and damping rates for periodic oscillations of a $1.96mm$ drop of an 0.05 % aqueous solution of the polyacrylamide P2540.

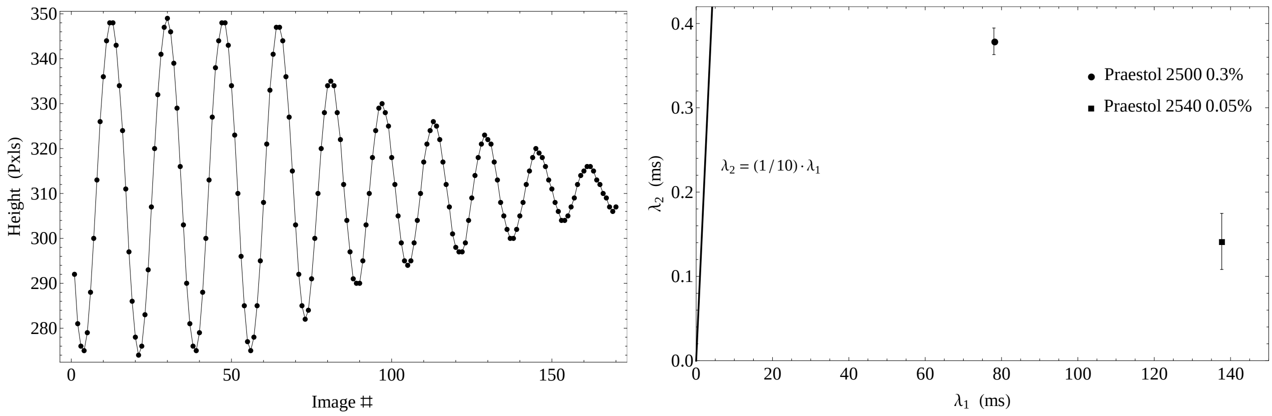

Figure 4. Left motion of the north pole of the levitated Praestol 2540 solution drop with time during a damped oscillation. Right pairs of time scales λ_1 and λ_2 for various polymer solutions.

be measured. This can be done experimentally with acoustically levitated drops, as shown by Trinh et al. for a Newtonian drop embedded in an immiscible Newtonian host liquid [15].

Since our interest is to determine the deformation retardation time of the polymeric substance in the solution, we look at solutions of the characteristic equation (33) for varying λ_2. The other quantity that is also unknown a priori is the viscosity η_0 in equation (8). Varying the retardation time in the range $0 \leq \lambda_2 \leq \lambda_1$ and the viscosity in the range $10^{-7} Pa \cdot s \leq \eta_0 \leq 1.5 Pa \cdot s$ for a drop of an aqueous solution of 0.05% mass polyacrylamide Praestol 2540 (Stockhausen Inc., Germany) ($\eta_0 = 1.521 Pa \cdot s$, $\rho = 998.8 kg/m^3$, $\sigma = 0.0765 N/m$, $\lambda_1 = 137.7 ms$) with the diameter $1.96mm$ we obtain the results in Fig. 3. We see that the range of dimensional frequencies and damping rates is a narrow band in the Gaussian plane of the real and imaginary parts of the complex angular frequency. These data represent that, with the values of oscillation frequency and damping rate known, the state of the drop defined by the pair of values of the unknown parameters is defined, so that the values may be determined by finding the point in this Gaussian plane.

Experimental determination of oscillation frequency and damping rate

For investigating the damped oscillation behaviour of single drops of viscoelastic liquids experimentally, we make use of the technique of acoustic levitation. This technique allows for the positioning of single liquid drops in the quasi-steady pressure field of a standing ultrasonic wave produced between two plates. One of the plates is a vibrating horn, which produces the waves. The other one acts as the reflector and has a concave curved surface in the present apparatus [16]. Oscillations of the levitated object may be excited by amplitude-modulating the ultrasound. Modulation frequencies up to $2kHz$ are achievable. For further details the reader is referred to [16].

For investigating an oscillating drop, the drop liquid is filled into an insulin syringe, which stands out for its very thin needle. With the needle tip close to a pressure node of the acoustic resonator, a portion of liquid is pushed out from the syringe to form the droplet. The achievable drop diameter ranges between $1.5mm$ and $2.5mm$. The drop resonance frequency is then determined by a modulation frequency sweep, monitoring the maximum occurring amplitudes of the oscillations. The drop is then steadily driven at that resonance frequency, and the modulation is switched off at a time $t = 0$, so that the drop carries out damped oscillations which eventually die out. This motion is recorded by a high-speed camera at a framing rate of $2kHz$, as shown in Fig. 4 left. From these data, the frequency and damping rate in the last part of the motion may be extracted, so that the real and imaginary parts of the complex angular frequency of this drop are known. Using these data for solving the characteristic equation numerically, we obtain the pair (λ_2, η_0) we want to determine for the liquid at hand. A summary of relaxation and retardation times of two different aqueous polyacrylamide solutions is shown in Fig. 4 right. Although these data should be regarded as preliminary, they show clearly that the usual practice in simulations of viscoelastic liquids to assume values between $1/10$ and $1/8$ for ratio λ_2/λ_1 may miss the correct value considerably. We are on the way to look more deeply into this method in order to establish it as a standard for determining deformation retardation times of polymeric liquids.

Conclusions

In the theoretical part of this study we analysed linear oscillations of viscoelastic drops in a gaseous environment. The solution of the equations of change for the problem yielded the characteristic equation for the complex oscillation frequency. Both the damping rate and the frequency of the oscillations are found to depend on the quantities for the viscous liquid behaviour and on the stress relaxation and deformation retardation time scales involved in the constitutive rheological equation. Preliminary experiments showed that the polymer deformation retardation time may be determined from measurements of frequency and damping rate of drops oscillating in an acoustic field. In further work on this subject, a robust algorithm for determining the retardation time from the characteristic equation and an accurate experimental technique will be developed.

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References

[1] Bauer, H.F., *Forschung im Ingenieurwesen* 52:81-88 (1986)
[2] Bird, R.B., Armstrong, R.C., Hassager, O., *Dynamics of polymeric liquids*, John Wiley & Sons, New York (1987)
[3] Chandrasekhar, S., *Proceedings of the London Mathematical Society* 9:141-149 (1959)
[4] Ho-Minh, D., Mai-Duy, N., Tran-Cong, T., *IOP Conference Series: Materials Science and Engineering* 10:012210 (2010)
[5] Huang, P.Y., Hu, H.H., Joseph, D.D., *Journal of Fluid Mechanics* 362:297-325 (1998)
[6] Khismatullin, D. B., Nadim, A., *Physical Review E* 63:061508 (2001).
[7] Lamb, H., Proceedings of the London Mathematical Society 13:51-66 (1881)
[8] Lamb, H., *Hydrodynamics*, Cambridge University Press (1932)
[9] Levich, V. G., *Physicochemical Hydrodynamics*, Prentice-Hall (1962)
[10] Miller, C.A., Scriven, L.E., *Journal of Fluid Mechanics* 32:417-435 (1968)
[11] Prosperetti, A., *Journal of Fluid Mechanics* 100:333-347 (1980)
[12] Phillips, T.N., Williams, A.J., *Journal of Non-Newtonian Fluid Mechanics* 87:215-246 (1999)
[13] Rayleigh, Lord, J.W.S., *Proceedings of the Royal Society London A* 29:71-97 (1879)
[14] Stelter, M., Brenn, G., Yarin, A.L., Singh, R.P., Durst, F., *Journal of Rheology* 44:595-616 (2000)
[15] Trinh, E., Zwern, A., Wang, T.G., *Journal of Fluid Mechanics* 115:453-474 (1982)
[16] Yarin, A.L., Brenn, G., Kastner, O., Rensink, D., Tropea, C., *Journal of Fluid Mechanics* 399:151-204 (1999)