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FLUID MECHANICS

MATHEMATICAL MODELING OF THE EFFECT OF VISCOELASTICITY ON THE FILM DRAINAGE BETWEEN INTERACTING DROPS

Dedicated to Professor Zapryan Zapryanov on the occasion of his 90th anniversary

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ABSTRACT: Mathematical model of the deformation and drainage of film between interacting drops is presented in the case when dispersed or continuous phases are viscoelastic fluids. The model is based on the assumptions of a gentle collision at small Reynolds numbers and small deformation at small capillary numbers. It consists of lubrication approximation in the film and creeping flow equation in the dispersed phase. The equations in the continuous and dispersed phases are coupled by continuity of the velocity and stress boundary conditions at the interface. Generalizations of the Maxwell-type rheological constitutive relation are used to model the viscoelastic effects: upper-convected Maxwell model in the drop and fractional Maxwell model in the film phase. Predictions of the effect of the extra elastic stresses on the film drainage are given.

KEY WORDS: Drop coalescence, Film drainage, Viscoelasticity, Upper-convected Maxwell model, Fractional Maxwell model.

1 INTRODUCTION

Viscoelastic behavior is exhibited by one or both phases in many industrial processes involving dispersions (blending of molten polymers, production of polymer foams, suspension polymerization, polymer coating, etc.), which can strongly affect subprocesses such as breakup and coalescence and, in turn, the properties of the final products. Thus, the investigations of coalescence in viscoelastic dispersions have not only theoretical but also practical significance. In recent decades a great progress has been made on the investigation of film drainage and rupture in Newtonian systems

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under different conditions: constant approach velocity or force [1, 2]; presence of insoluble [3–5] and soluble [6] surfactants. In recent years numerical solutions of generalized Newtonian film drainage have been presented. In [7] mobile drainage of film of power-law and Carreau fluids are considered. The power-law film drainage between Newtonian drops is studied in [8]. Drainage of Newtonian film between power-law drops is numerically investigated in [9]. However, the extension of such investigations to viscoelastic dispersions is still in its infancy.

In the present work the coalescence of axi-symmetrically interacting viscoelastic drops with a viscoelastic film between them is studied. Following the line of investigation used in the above-mentioned papers, the coalescence is split into three phases (see e.g. [10]):

(i) The external flow field, governing the frequency, strength and duration of collisions.

(ii) The process of film formation and drainage.

(iii) The destabilization of the film by the van der Waals and other intermolecular forces, leading to rupture.

Element (i) furnishes the initial and boundary conditions for (ii), which in turn provides those for (iii). Of the various elements involved, coalescence sub-process (ii) is especially sensitive to deviation from the Newtonian rheology: small changes in the stresses exerted on the film translate into large forces per unit volume.

In the present study sub-process (ii) is considered. The main goal is to investigate the influence of the viscoelasticity in both phases on the film drainage. In the dispersed phase an upper-convected Maxwell (UCM) model is used to describe the flow in the drops, which has the advantage to have only one extra parameter – relaxation time. UCM model seems to be satisfactory for small shear and elongational rates, which may be the case at the last, rate-determining, stage of the coalescence process – film drainage. Classical Maxwell model and its fractional generalizations are used to model the viscoelastic flow in the continuous (film) phase. The choice of fractional Maxwell model of the film flow is supported by recent experimental results (see [11]), which show that fractional Maxwell models are an excellent alternative for describing the dynamic rheological behavior of viscoelastic films.

The paper is organized as follows. In Section 2 the mathematical model is presented, the main parts of which are essentially the same as in the previous studies mentioned above. The new elements here are the viscoelastic models in the dispersed and continuous phases, respectively. In Section 3 numerical results are given for the case of drainage of Newtonian film between viscoelastic drops. The results are part of a previous work [12]. Predictions for the drainage rates of a viscoelastic film between Newtonian drops are also discussed. Conclusions of this study are presented in Section 4.

2 Mathematical formulation

Two drops with radii R_1 and R_2 are considered, which are of the same viscoelastic liquid and interact along the line connecting their centers in another immiscible viscoelastic fluid, see Fig. 1. Here μ is the viscosity and θ is the relaxation time of the drop phase. The continuous phase is characterized by viscosity μ/λ, λ being the viscosity ratio, and shear elasticity E.

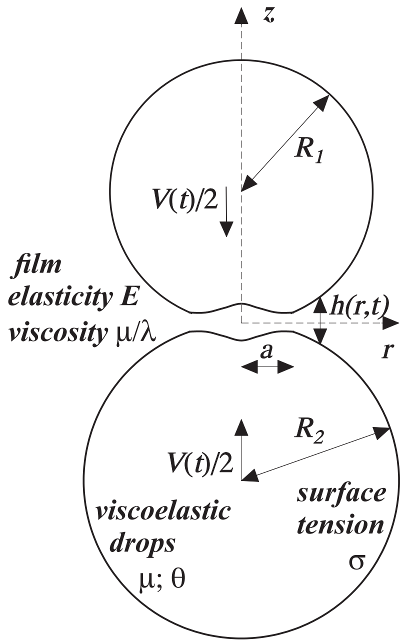

Fig. 1: Schematic sketch of the problem.

The drops are considered to approach each other at specified velocity $V(t)$. In the numerical examples presented here the approach velocity is adjusted in time during the drainage process to maintain a constant interaction force. The same procedure can however be used for time dependent approach velocities or interaction forces that are more representative to actual drop collision. The interfacial tension σ at the drop interfaces is assumed to be constant.

2.1 Approximations and simplifications

The model is simplified by a number of approximations, which are valid in the limit of gentle collisions and which have been discussed in detail in [13]. The validity of the approximated model was confirmed recently by comparisons with numerical simulations at finite drop deformation [14]. Some of the simplifications are discussed below in regard to the restrictions on the applicability of the model as well as the advantages that they give for intensive numerical investigation of the film drainage in the framework of coalescence process.

A) Small deformation:

(1) $$a \ll R_i\,, \qquad i = 1,\ 2,$$

where a is the film radius defined by Eq. (15).

As noted in [10], assumption A) is not as restrictive as it looks, since only for gentle collisions drainage is typically rapid enough for coalescence to occur. The approximation A) enables a major simplification of the governing equations in the case of Newtonian film drainage, which become the same for drops of unequal radii ($R_1 \neq R_2$) and equal drops with an equivalent radius R_{eq}. The equivalent equal-drop radius is defined in [1, 10] as follows:

(2) $$R_{\text{eq}}^{-1} = 0.5\left(R_1^{-1} + R_2^{-1}\right).$$

Below the mathematical model is given in the case of drops of equal radius $R_1 = R_2 = R_{eq}$. A possible generalization to the case of drainage of viscoelastic film between drops of different radii is discussed in Section 2.3.

Assumption A) guarantees small interfacial gradient in the severely deformed region of the drops:

(3) $$\left|\frac{\partial h}{\partial r}\right| \ll 1\,, \qquad r \ll R_{\text{eq}},$$

where h and r denote local film thickness and radial coordinate, respectively (see Fig. 1). This allows another simplification - the interface is approximated as a plane as flow in the drop is concerned. This approximation leads to two important advantages. First, the solution in the dispersed phase is independent of the drop radius R_{eq}, which reduces the number of the parameters of the problem. Second, the direct boundary integral relation between the interfacial stress and velocity for the Stokes flow in half space, see [15], simplifies significantly the numerical procedure for simulations of film drainage between Newtonian drops. In the framework of previous study, [13], the approximation of flat drop interface has been verified via a comparison with finite element method (FEM) simulation of the flow in initially spherical drop of radius $R_{\text{eq}} \approx 10a$, taking into account the deformation of the drop as well. The comparison shows no difference between the results regarding film drainage, respectively film thickness h.

B) Inertial forces in the film and the adjacent dispersed-phase flow are negligible, which is in agreement with the assumption of gentle collisions.

C) The influence of the normal stresses in the film, is neglected. This allows the use of lubrication approximation in the film. Thus, the quasi-parallel flow in the film is approximated as the sum of a plug (uniform) part and a parabolic part.

Under the assumption B) the governing creeping flow equations of mass and momentum conservation are:

$$\nabla \cdot \mathbf{v}_b = 0\,, \tag{4}$$

$$\nabla p_b = \nabla \cdot \tau_b\,, \tag{5}$$

where p_b is the pressure, $\mathbf{v}_b$ – the velocity, and τ_b is the deviatoric stress tensor. Here b stands for d in the case of the model in the drop and f in the case of the film. For brevity the subscript f, denoting the variables in the film will be omitted further. To close the mathematical model (4)–(5) for the flow in the drop and film phase, a constitutive relation between the deviatoric stress tensor τ_b and the rate-of-deformation tensor $\dot{\epsilon}$ has to be given, where

$$\dot{\epsilon} = \nabla \mathbf{v}_b + (\nabla \mathbf{v}_b)^T\,. \tag{6}$$

Such rheological constitutive relations are described in following subsections, where the viscoelastic models of the dispersed phase and the film phase flows are discussed.

The continuity equation in the film (4) in cylindrical coordinates is

$$\frac{1}{r}\frac{\partial}{\partial r}(r v_r) + \frac{\partial v_z}{\partial z} = 0\,. \tag{7}$$

Here v_r is given by lubrication approximation of (5), which in the case of Newtonian film reads (see also [15]):

$$\begin{aligned} &\frac{\partial p}{\partial z} = 0\,,\\ &\frac{\partial p}{\partial r} = \frac{\mu}{\lambda}\,\frac{\partial^2 v_r}{\partial z^2}\,. \end{aligned} \tag{8}$$

After integration of (7) in z in the interval $[z_1,\, z_2]$ where $z = z_i(r,t)$, are the boundaries of the film, and taking into account that $z_2(r,t) = -z_1(r,t) = h(r,t)/2$ and $-v_z(z = z_1) = v_z(z = z_2) = \dfrac{1}{2}\dfrac{\partial h}{\partial t}$, the following equation for the evolution of the film thickness h is obtained:

$$\frac{\partial h}{\partial t} = -\frac{1}{r}\frac{\partial (rhu)}{\partial r}\,, \qquad u = u_u + u_p = \frac{1}{h}\int_{z_1}^{z_2} v_r(z)dz\,. \tag{9}$$

Here u is the mean velocity in the film, consisting of the sum of two parts: plug velocity u_u and parabolic velocity u_p. The plug part u_u is also the interface velocity and thus is directly related to the drop flow. The parabolic part u_p depends on the pressure gradient $\dfrac{\partial p}{\partial r}$ and the rheological constitutive relation.

In a similar manner as above integration of the momentum equation in the film in z direction in the interval [0, z_i] results in the following relation for the tangential stress $\tau_f = \tau(z_i)$, exerted on the interface $z = z_i$ by the film:

$$\tau_f = -\frac{h}{2} \cdot \frac{\partial p}{\partial r}\,. \tag{10}$$

To obtain expression (10) of the tangential stress at the interfaces $z_2 = -z_1 = h/2$ no assumptions about the rheological relation between the stress and the rate-of-deformation tensors are made. Only the lubrication approximation in the film is used, where the film flow is symmetric with respect to the plane $z = 0$ and the pressure p is constant in z direction. Therefore, relation (10) is valid independently of the rheology in the film. In the case of power-law fluid in the film phase this was confirmed in the recent article [8].

The boundary conditions at the interface consist of continuity of the tangential velocity and tangential stress:

$$u_u = (v_d)_r|_{\text{interface}}\,, \tag{11}$$

$$\tau_f = (\tau_d)_{zr}|_{\text{interface}}\,. \tag{12}$$

The normal stress boundary condition, associated with a pressure jump due to the interfacial tension σ, is:

$$p = \frac{2\sigma}{R_{\text{eq}}} - \frac{\sigma}{2r}\frac{\partial}{\partial r}\left(r\frac{\partial h}{\partial r}\right). \tag{13}$$

The above equation is obtained under the assumptions discussed earlier and a simplification A), based on the small interfacial slope assumption (3). Thus, the terms $\left(\dfrac{\partial h}{\partial r}\right)^2$ in the original Young-Laplace equation can be neglected. In a recent study [16] the authors report a discontinuity in the tangential stress τ_f and associate this with the use of the linearized Young-Laplace equation (13). In fact, in [16] the condition of zero pressure far from the film region is not satisfied, see eq. (11) and Fig. 11 in [16]. Thus, small inaccuracy in the pressure gradient translates in a significant error in the tangential stress. Indeed, it follows from (10) that the tangential stress is proportional to the product of the pressure gradient and the film thickness h. This explains the reported discontinuity which appears at the end of the computational domain, where dimensionless thickness of the film is of order of 200.

In the present work, as in [9, 13], the boundary condition of zero pressure (14) is satisfied with accuracy of order 10^{-9} and using the linearized Young-Laplace equation (13) the solution is smooth without any discontinuity. Our investigation shows that the difference in the results in the film region using both versions of Young-Laplace equation is negligible, which in fact is supported by the results in [16]. The simplified version (13) has an advantage that the number of parameters of the problems is reduced. Indeed, using the original Young-Laplace equation in [16] capillary number Ca appears as an additional parameter in the dimensionless model.

The outer boundary conditions for pressure and approach velocity at sufficiently large distance from the film region ($r = r_{\text{large}}$) are

$$p(r_{\text{large}}) = 0 \quad \text{and} \quad \left(\frac{\partial h}{\partial t}\right)_{r_{\text{large}}} = -V(t)\,, \tag{14}$$

where $V(t)$ is adjusted so that

$$2\pi \int_0^{r_{\text{large}}} p(r) \cdot r\, dr = F = \frac{2\pi a^2 \sigma}{R_{\text{eq}}}\,. \tag{15}$$

Here F is the interaction force which is chosen as a constant; the constant a is a measure of the film radius. The initial condition for the interface is

$$h(t=0) = h_0 + \frac{r^2}{R_{\text{eq}}}\,, \tag{16}$$

which corresponds to initially undeformed drops.

As in previous studies, a transformation of the variables in the equations is applied which makes the governing equations dimensionless and reduces the number of parameters. The transformation is

$$\begin{aligned} r^* &= \frac{r}{R_{\text{eq}} a^*}\,, \quad z^* = \frac{z}{R_{\text{eq}} a^*}\,, \quad h^* = \frac{h}{R_{\text{eq}} a^{*2}}\,, \quad t^* = \frac{t\sigma a^*}{R_{\text{eq}}\mu}\,, \\ p^* &= \frac{p R_{\text{eq}}}{\sigma}\,, \quad p_d^* = \frac{p_d R_{\text{eq}}}{\sigma a^*}\,, \quad v^* = \frac{v\mu}{\sigma a^{*2}}\,, \quad \tau^* = \frac{\tau R_{\text{eq}}}{\sigma a^*}\,, \quad \lambda^* = \lambda a^*\,, \end{aligned} \tag{17}$$

where the dimensionless film radius a^* is defined as $a^* = a/R_{\text{eq}}$.

Applying (17), the dimensionless mathematical model in the case when both phases are Newtonian has only one hydrodynamical parameter - the dimensionless group λ^*, containing viscosity ratio λ. Three additional parameters, initial separation h_0^*, initial approach velocity $V^*(0)$ and r^*_{large} enter the model via the initial and boundary conditions, however they have no influence on the film drainage if chosen sufficiently large. Additional dimensionless parameters that appear in the model when one or both phases are viscoelastic are discussed below.

In Section 2.2 the case of drainage of Newtonian film between viscoelastic drops is considered. Mathematical model of drainage of viscoelastic film between Newtonian drops is given in Section 2.3. For the sake of brevity the asterisks denoting dimensionless variables and parameters are further omitted.

2.2 Drainage of Newtonian film between interacting viscoelastic drops

As discussed earlier, the creeping flow equations (4)–(5) in the drop phase

$$\nabla \cdot \mathbf{v}_d = 0\,, \qquad \nabla p_d = \nabla \cdot \tau_d\,, \tag{18}$$

have to be completed with a constitutive relation between the deviatoric stress τ_d and the rate-of-deformation tensor $\dot{\epsilon}$ given in (6). Here for modeling of the extra elastic stress in the case of viscoelastic disperse phase the upper-convected Maxwell model is used:

$$\tau_d + \theta \overset{\nabla}{\tau_d} = \mu \dot{\epsilon}, \tag{19}$$

where θ is the relaxation time and $\overset{\nabla}{\tau_d}$ is the upper-convected time derivative of the stress tensor τ_d, defined as:

$$\overset{\nabla}{\tau_d} = \frac{\partial \tau_d}{\partial t} + \mathbf{v}_d \cdot \nabla \tau_d - (\nabla \mathbf{v}_d)^T \cdot \tau_d - \tau_d \cdot (\nabla \mathbf{v}_d)\ . \tag{20}$$

Let us note that the one-mode UCM model, described above, is not the best choice from practical point of view. There exist multiple-mode viscoelastic models [17] (e.g. Phan-Thien-Thanner or Giesekus models) that can better describe viscoelastic flows. To get an idea about the influence of the extra elastic stresses on the film drainage compared to Newtonian drops, UCM model (19)–(20) is chosen here mainly because of the smaller number of parameters. For quantitative characterization of the effect of elasticity on the drop coalescence in industrially important viscoelastic dispersions more realistic models could be used.

In dimensionless variables the UCM model is (the asterisks are omitted):

$$De \frac{\partial \tau_d}{\partial t} + Wi \left[\mathbf{v}_d \cdot \nabla \tau_d - (\nabla \mathbf{v}_d)^T \cdot \tau_d - \tau_d \cdot \nabla \mathbf{v}_d \right] + \tau_d = \dot{\epsilon}\,, \tag{21}$$

where $De = \theta / T$ is the Deborah number and $Wi = \theta \sigma a^* / (\mu R_{\text{eq}})$ is the Weissenberg number. It is worth noting that the characteristic time T in the definition of the

Deborah number could be different from that in the drainage process $\mu R_{\text{eq}}/\sigma a^*$, used in the scaling (17). Thus, in general $De \neq Wi$. The numerical results presented in the next section are obtained in the limit of zero Deborah number ($De = 0$), corresponding to a quasi-steady UCM model.

To the end of this section the model will be completed with the governing equation in dimensionless form in the case of Newtonian film. This is in correspondence with the numerical results in Section 3.1 concerning Newtonian film drainage between viscoelastic drops. Mathematical model for viscoelastic film between viscoelastic drops can be written in a straightforward manner by combining the above viscoelastic model of the flow in the drop with the model for viscoelastic film drainage presented in the following subsection. This is possible because the boundary conditions at the interface are independent of the rheology of the flows in both phases (see eqs. (23)–(26)).

Equation (9) for the evolution of the film thickness h in the case of Newtonian film is

$$\frac{\partial h}{\partial t} = -\frac{1}{r} \cdot \frac{\partial (rhu_u)}{\partial r} + \frac{1}{r} \cdot \frac{\lambda}{12} \cdot \frac{\partial}{\partial r}\left(h^3 r \frac{\partial p}{\partial r}\right), \tag{22}$$

where the second term in the r.h.s. corresponds to the parabolic part of the film velocity, given by (8).

Some of the elements of the film drainage model in dimensionless form are the same as dimensional ones. Below they are given again for completeness

$$\tau_f = -\frac{h}{2} \cdot \frac{\partial p}{\partial r}; \tag{23}$$

the boundary conditions at the interface represent continuity of the tangential velocity and stress

$$u_u = (v_d)_r|_{\text{interface}}\,; \tag{24}$$

$$\tau_f = (\tau_d)_{zr}|_{\text{interface}}\,; \tag{25}$$

$$p = 2 - \frac{1}{2r}\frac{\partial}{\partial r}\left(r\frac{\partial h}{\partial r}\right); \tag{26}$$

the outer boundary conditions:

$$p(r_{\text{large}}) = 0 \quad \text{and} \quad \left(\frac{\partial h}{\partial t}\right)_{r_{\text{large}}} = -V(t)\,; \tag{27}$$

constant interaction force:

$$\int_0^{r_{\text{large}}} p(r) \cdot r\, dr = 1\,; \tag{28}$$

and the initial position of the interface $z = h/2$:

(29) $$h(t = 0) = h_0 + r^2 .$$

The mathematical model (21)–(29) is used in Section 3.1 for numerical simulation of film drainage of Newtonian film between viscoelastic drops.

2.3 Drainage of viscoelastic film between interacting Newtonian drops

As mentioned earlier, the boundary and outer conditions (24)–(29), being independent of the rheology of the phases, are the same also in the case of viscoelastic film and are not given here.

For the modeling of the viscoelastic film flow a time-fractional generalization of the Maxwell viscoelastic constitutive equation is considered. The motivation of this choice is based on the following. First, the spatial part of the upper-convected operator in (20) has no contribution in the case of lubrication approximation (unidirectional velocity $v_r(z)$ in the film). Second, findings in recent experimental studies (see e.g. [11] and references therein) on the behaviors of viscoelastic films show that the fractional Maxwell model optimally adjusts the dynamic and creep rheological behaviors. In all examples of viscoelastic materials, considered in [11], best fit is obtained with a fractional Maxwell model with two modes (i.e. with two spring-pot elements).

Consider first the classical Maxwell model, which relates the deviatoric stress τ with the rate-of-deformation tensor $\dot{\epsilon}$ in dimensionless form as follows

(30) $$\tau + Wi\frac{\partial \tau}{\partial t} = \frac{1}{\lambda}\dot{\epsilon},$$

where the Weissenberg number is defined as $Wi = \lambda\sigma/(ER_{\text{eq}})$.

The two-mode time-fractional generalization of the classical Maxwell model (30) is given bellow in its most popular form, see e.g. [11]. It represents two spring-pots (called also Scott-Blair elements) with parameters (E_1, θ_1, α) and (E_2, θ_2, β), where E_j and θ_j are elasticity and relaxation time for j-th mode, and the fractional parameters α and β satisfy $1 \geq \alpha \geq \beta \geq 0$. The fractional Maxwell constitutive equation is:

(31) $$\tau + \theta^{\alpha-\beta}\frac{\partial^{\alpha-\beta}\tau}{\partial t^{\alpha-\beta}} = E\theta^{\alpha}\frac{\partial^{\alpha}\epsilon}{\partial t^{\alpha}} ,$$

where

$$E = E_1\left(\frac{\theta_1}{\theta}\right)^{\alpha}, \qquad \theta = \left(\frac{E_1\theta_1^{\alpha}}{E_2\theta_2^{\beta}}\right)^{\frac{1}{\alpha-\beta}} .$$

In (31) the Caputo fractional derivative of order γ, $0 < \gamma < 1$, is used

$$\frac{\partial^\gamma f(t,\cdot)}{\partial t^\gamma} = \frac{1}{\Gamma(1-\gamma)} \int_0^t (t-\xi)^{-\gamma} \frac{\partial f(\xi,\cdot)}{\partial \xi}\, d\xi\,, \tag{32}$$

where Γ is the Euler Gamma function.

Taking into account that the elasticity is the ratio between the viscosity and the relaxation time, $E_j = \mu/(\lambda\theta_j)$, and applying the transformation (17), we rewrite the fractional Maxwell constitutive equation (31) in a dimensionless form, which is more suitable in the context of the film drainage model

$$\mathcal{A}_t(\tau) := \left(\frac{\theta_1}{\hat{t}}\right)^{1-\alpha} \frac{\partial^{1-\alpha}\tau}{\partial t^{1-\alpha}} + Wi\left(\frac{\theta_2}{\hat{t}}\right)^{-\beta} \frac{\partial^{1-\beta}\tau}{\partial t^{1-\beta}} = \frac{1}{\lambda}\,\dot{\epsilon}\,, \tag{33}$$

where $\hat{t} = \mu R_{\text{eq}}/(\sigma a^*)$ is the characteristic time defined in (17) and the Weissenberg number $Wi = \lambda\sigma/(E_2 R_{\text{eq}})$. To obtain (33) fractional differentiation of order $1-\alpha$ is applied to both sides of eq. (31) and the property that the composition of fractional derivatives of order α and β is fractional derivative of order $\alpha+\beta$ is employed. This property holds since $\tau(t=0) = 0$. For convenience, the composite time-fractional operator $\mathcal{A}_t$ is introduced in (33).

In the case $\beta = 0$ the two-mode fractional model (33) reduces to a single mode fractional Maxwell model that represents combination of spring and spring-pot element, while at $\alpha = 1,\ \beta = 0$ the classical Maxwell model (30) is recovered.

Applying $\mathcal{A}_t$ to the momentum conservation equation $\nabla p = \nabla\cdot\tau$ and taking into account (33) an equation for the velocity in the film is obtained as follows:

$$\mathcal{A}_t(\nabla p) = \mathcal{A}_t(\nabla\cdot\tau) = \nabla\cdot\mathcal{A}_t(\tau) = \frac{1}{\lambda}\,\nabla\cdot\dot{\epsilon}\,. \tag{34}$$

Using the assumption of lubrication approximation, $(\partial p/\partial z = 0)$, equation (34) implies:

$$\mathcal{A}_t\left(\frac{\partial p}{\partial r}\right) = \frac{1}{\lambda}\frac{\partial^2 v_r}{\partial z^2}\,, \tag{35}$$

which can be considered as a generalization of the classical lubrication theory in the case of fractional Maxwell model. Let us note that if $\alpha = 1$ and $Wi = 0$ then $\mathcal{A}_t$ is the identity operator and (35) reduces to the classical one (8). It can be seen from (35) that the viscoelastic flow in the film has a parabolic profile, as in the case of Newtonian film. Let us note that parabolic profile flow has been obtained by other authors (see e.g. [18]) using different viscoelastic models under the assumption of thin-film approximation or unidirectional flow.

The parabolic part of the velocity in the film with boundaries $[z_1,\ z_2]$ given by (35) is

$$(v_r(z))_p = \frac{\lambda}{2}\,\mathcal{A}_t\Big(\frac{\partial p}{\partial r}\Big)(z - z_1)(z - z_2)\,. \tag{36}$$

Thus, in the case of time-fractional Maxwell model (33) the evolution equation for the film thickness (9) becomes:

$$\frac{\partial h}{\partial t} = -\frac{1}{r}\cdot\frac{\partial(rhu_u)}{\partial r} + \frac{1}{r}\cdot\frac{\lambda}{12}\cdot\frac{\partial}{\partial r}\Big[h^3 r\mathcal{A}_t\Big(\frac{\partial p}{\partial r}\Big)\Big]. \tag{37}$$

Below we discuss a possible application of the results for equal drops to that of film drainage between drops of different radii (R_1 and R_2). The main concern is whether the tangential stresses, respectively velocities, will be equal at the bounding interfaces $z = z_1$ and $z = z_2$ in the case of an asymmetric film. To address this in the case of viscoelastic film the tangential stresses that the film flow (36) exerts on the drops are evaluated. The velocity gradients ($\dot{\epsilon}$) at the interfaces are related by

$$\frac{\partial v_r}{\partial z}(z = z_1) = -\frac{\partial v_r}{\partial z}(z = z_2) = -\frac{\lambda h}{2}\,\mathcal{A}_t\left(\frac{\partial p}{\partial r}\right) \tag{38}$$

under the assumption that the velocities at the bounding interfaces are equal, $v_r(z_1) = v_r(z_2)$. (Since they are equal at $t = 0$ due to the initial conditions, they will stay equal as far as the tangential stresses are equal, $\tau_f(z = z_1) = \tau_f(z = z_2)$.) Taking into account (33) it follows:

$$\mathcal{A}_t(\tau_f(z = z_1)) = \mathcal{A}_t(\tau_f(z = z_2)) = -\frac{h}{2}\,\mathcal{A}_t\Big(\frac{\partial p}{\partial r}\Big). \tag{39}$$

Due to the injectivity property of the operator $\mathcal{A}_t$ (which can be easily established by the use of Laplace transform), (39) implies

$$\tau_f(z = z_1) = \tau_f(z = z_2) = -\frac{h}{2}\,\frac{\partial p}{\partial r}\,, \tag{40}$$

which is consistent with (10).

The above discussion indicates that for the considered in the present section viscoelastic models the results for the film thickness in the case of equal size drops could be used also to describe the film drainage between drops of unequal radii.

The presence of time derivatives in $\mathcal{A}_t$, respectively in (37), requires an extra initial condition. A natural choice, consistent with the initial condition (16) of undeformed drops, is that the interfaces are at rest at the beginning

$$\frac{\partial h}{\partial t}(t = 0) = 0\,. \tag{41}$$

This also implies $\dfrac{\partial p}{\partial t}(t = 0) = 0$ and from (26), (29), and (36) $v_r(t = 0) = 0$.

3 Numerical Method, Results and Discussion

The general numerical scheme is the same as that used in earlier studies [9, 13] with the only difference with respect to the numerical methods for the non-linear drop equations (18)-(21) and the non-local time-fractional equations in the film (30)-(37). To better understand the effect of the extra elastic stresses on the film drainage, only in one of the phases at a time the model is considered viscoelastic. That is, two combinations are studied: viscoelastic drops in Newtonian fluid and viscoelastic film between Newtonian drops.

3.1 Viscoelastic drops

The equations (18)-(21) in the drop phase are solved by means of a finite element method, based on the work [17]. The finite elements used are of second order approximation for the velocity and linear for the pressure. For the spatial discretization non-uniform meshes of triangular elements are used. At the last stage of the process, film drainage, in the area adjacent to the film $(r < 1.2$ and small $z)$ the spatial step is of order 0.02 and constantly increases becoming two orders of magnitude larger far from the film (at large r and z). The mesh for discretization of the one dimensional film equations (22), (26) consists of the nodes of the finite elements mesh at $z = 0$.

The results presented bellow aim qualitative investigation of the influence of the viscoelasticity of the drop flow on the film drainage. Thus, they concern the partially mobile limit (small λ-values), where the influence of the flow in the drops on the film flow is strong. Therefore, the case $\lambda = 0.05$ is considered, for which the parabolic contribution to the film velocity, being proportional to λ in (22), is negligible and the film drainage depends mostly on the drop flow.

In Fig. 2 the film thickness profiles in the case of Newtonian drops, $Wi = 0$ (left), is compared to that for viscoelastic drops, $Wi = 2.4$ (right). The profiles in both parts of the figure are chosen to correspond to same minimal film thickness h_{min}. It is seen that the main difference between the film thickness profiles with respect to the gap shape is the separation between the drops at which the film and respectively dimple is formed. For Newtonian drops, $Wi = 0$, the film is formed at $h_{\text{min}} \approx 0.2$, while for viscoelastic drops, $Wi = 2.4$, the corresponding value is $h_{min} \approx 0.05$. Thus, due to the extra elasticity of the drop phase the film/dimple is formed at a smaller separation between the drops, which increases the drainage rate. This is seen in Fig. 3, where the evolution of the minimal film thicknesses is shown for different Weissenberg numbers.

To understand how the elasticity of the drops effects the film and dimple formation the following test is performed. The flow in the drop is calculated for increasing Wi at given tangential stress τ_f on the interface as a boundary condition. The tangential

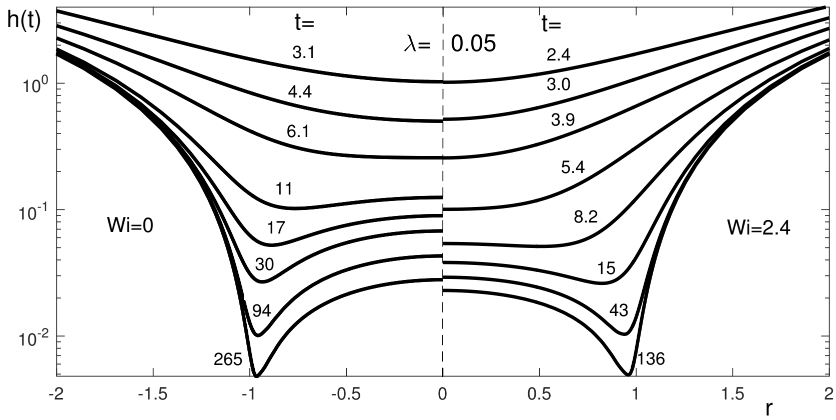

Fig. 2: Film thickness profiles at different times and $\lambda = 0.05$: Newtonian drops (left) and viscoelastic drops at $Wi = 2.4$ (right).

stress (see Fig. 4(a)) corresponding via (23), (26) to the film profile in the left part of Fig. 2 at $t = 6.1$ is chosen. The resulting interfacial velocity is shown in Fig. 4(b) for different Weissenberg numbers. It is seen that increasing Wi the maximal value of the interfacial velocity increases and moves towards the center of the film. Such dependence on the value of Wi indicates that the extra elasticity of the drop flow tries to pump out the fluid from the film and in this way preventing film as well as dimple formation.

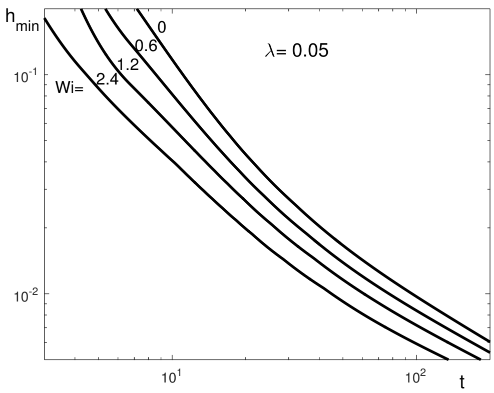

Fig. 3: The evolution of the minimal film thickness at different Weissenberg numbers.

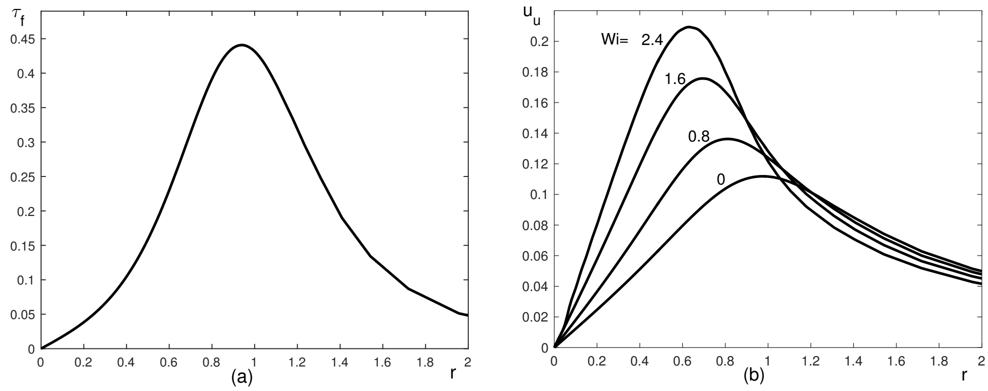

Fig. 4: Tangential stress τ_f corresponding to film shape at $t = 6.1$ from Fig. 2 at $Wi = 0$ (a). Interfacial velocity u_u corresponding to the tangential stress from the left part for different values of Wi (b).

3.2 VISCOELASTIC FILM

In Section 2.3 film drainage model was presented where the extra viscoelastic stress in the film was introduced by time-fractional Maxwell rheological constitutive equation (33). This approach leads to an extra fractional integro-differential operator $\mathcal{A}_t$ in the equation for the film thickness (37). Thus, in the case of viscoelastic film the film thickness $h(r,t)$ is described by integro-differential equation in time t, which is of fourth order with respect to r. This together with the singularity of the kernels in the fractional derivatives poses significant challenges in the numerical simulation of the film drainage model. Finite-difference numerical approximation of the spatial derivatives in combination with Grünwald-Letnikov-type approximation of the time-fractional derivative will be subject of a future study, aiming numerical investigation of the drainage of viscoelastic film.

To get an idea how the extra elastic stresses in the film can affect the film drainage, a similar test as in the case of viscoelastic drops is performed. For the test the classical Maxwell model (30) is considered, where the viscous and the elastic parts of the stress are well separated. Thus, for a given film thickness profile, the parabolic film velocity that corresponds to the elastic part of the stress, u_{els}, is calculated and compared with the pure viscous part, u_{vis} ($u_p = u_{vis} + u_{els}$). Two film thickness profiles, see Fig. 5, that correspond to Newtonian film drainage at $\lambda = 10$ are considered. The film thickness profile at $t = 0.34$ is from the beginning of the drainage process - the film formation, while that at $t = 4.8$ is typical for the last stage when the film and the dimple are well established.

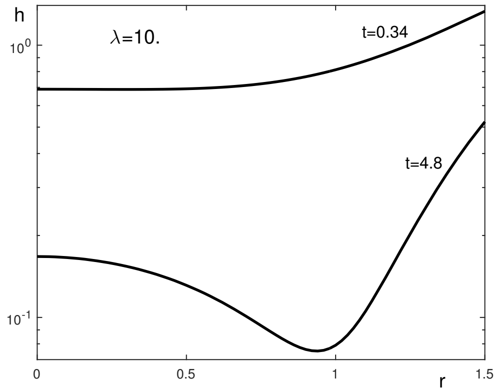

Fig. 5: Film profiles: at the film formation ($t = 0.34$); after the dimple is formed ($t = 4.8$).

The results of the test described above are presented in Fig. 6. It is seen that at the beginning of the film drainage process $t = 0.34$ the extra elastic part of the velocity u_{els} (dashed line) is significant, and at the periphery of the film $r \approx 1$ is even higher than u_{vis}. This indicates that at this stage (film formation) of the process the extra elastic stress in the film can contribute to a faster drainage. The profiles of the mean velocity shown in Fig. 6(a) are qualitatively similar to these presented in Fig. 4(b). Therefore, as in the case of viscoelastic drops, it could be expected that the viscoelastic film and the dimple will be formed at smaller distance between the

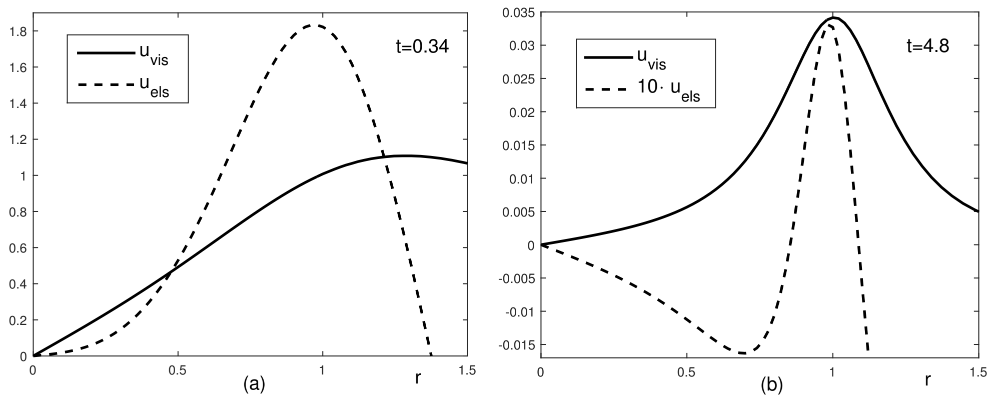

Fig. 6: The viscous, u_{vis}, and the elastic, u_{els}, parts at $Wi = 1$ of the mean parabolic film velocity corresponding to the film profiles from Fig. 5 at $t = 0.34$ (a) and $t = 4.8$ (b).

drops, compared to that of Newtonian film. At the last stage of the film drainage, however, the extra elastic part of the film mean velocity is negligible with respect the viscous one, see Fig. 6(b).

4 Conclusions

In this study a mathematical model for viscoelastic film drainage is presented based on UCM model in the drop phase and time-fractional Maxwell model in the film. It is shown that for the considered viscoelastic model in the film the profile of the film velocity is parabolic and depends on time fractional operator of the pressure gradient in the film, $\mathcal{A}_t\left(\frac{\partial p}{\partial r}\right)$. Regarding the effect of viscoelasticity on the film drainage, the main conclusion is that during their collision the drops are less deformed when the dispersed or film phase is viscoelastic than those of comparable Newtonian system at same minimal film thickness. Therefore, the film and the dimple are formed at thinner film thickness, which leads to a faster film drainage and in general can enhance the coalescence rate. This is in agreement with the predictions in [19] based on diffuse-interface method simulations of viscoelastic drop interaction.

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