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Modelling the breakup behaviour of threads in a two-phase system

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Modelling the Breakup Behaviour of Threads in a Two-Phase System

Final project

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Summary

In this study the breakup behaviour of viscoelastic threads is investigated using the finite element method. Newly developed MATLAB software is used for the initial calculations, after which the model is implemented in the SEPRAN finite element package. First, calculations are performed for viscous threads in a viscous matrix. The results from these calculations are in close agreement with analytical results (based on a linearized theory), even at finite deformations of the thread.

Further calculations focus on the behaviour of viscoelastic threads in air and in a viscous matrix. The calculations show a buildup of elongational stresses in the necks of the disturbed thread, resulting in a typical dumbbell shape of the thread and a significant retardation of the breakup process, which is in agreement with experimental results. Applying initial stresses in the thread model, to account for the stretching of the dispersed domains into threads, also slows down the breakup process considerably. The influence of disturbances of the interfacial tension along the interface is also studied and proves to be an important factor at the initial stage of the breakup process.
Chapter 1

Introduction

Many industrial processes involve dispersion of immiscible liquids into a blend. In polymer processing for example, melt blending of immiscible homopolymers is a common route to obtain materials with specific properties. The morphology achieved during the mixing process is an essential factor for the properties eventually obtained, and should therefore be carefully studied. There are two important stages in the mixing process which can be recognised. The first is the initial stage of mixing, when the dispersed domains are fairly large and the main driving force for deformation is the shear stress. The second stage starts once the dispersed domains have been stretched into long slender threads and the interfacial stress between the two phases becomes the main driving force for deformation, causing the dispersed phase to break up into droplets.

The first studies of such breakup processes were carried out by Rayleigh [1], who investigated the breakup of water jets in air taking the interfacial tension and inertia of the thread phase into account. Weber [2] added viscous stresses in the thread, while Tomotika [3] extended the analysis by taking the viscosity of the matrix into account. Most of the literature on viscoelastic threads is confined to polymer solutions in a gaseous matrix (e.g. Bousfield et. al. [4], Keunings [5]), while relatively little research incorporates a viscous matrix phase (Elmendorp [6]). This study focuses on the breakup process of viscoelastic threads in a viscous matrix, especially on the modelling of such systems. It is a continuation of previous research by Hermans [7] and Weijers [8], and has been partially published in Janssen [9].

First the more straightforward case of viscous threads, for which a linear theory is available, is studied. A model of the thread and the surrounding matrix material, as implemented in a finite element code, is described in Chapter 2. Here also some specific techniques concerning the mesh deformation and the implementation of interfacial stresses are discussed. In Chapter 3, the results of the calculations with viscous threads are given and compared to both analytical results according to Tomotika [3] and model experiments according to Janssen [9]. Chapter 4 deals with the breakup of viscoelastic threads, both in a viscous matrix and in air, in which case the
influence of the matrix can be neglected. For the simulations in a viscous matrix the results are also compared to model experiments. The influence of initial longitudinal stresses in the thread caused by the stretching of the thread is taken into consideration and its effects are studied. In Chapter 5 the calculations are extended to incorporate disturbances of the interfacial tension and some results are shown. Finally, in Chapter 6 some conclusions are drawn from the results of this study and recommendations for further research are given.
Chapter 2

The Breakup Model

2.1 Theory

During the mixing process, drops of the dispersed material are deformed into long, slender threads due to affine stretching. At a certain point, local radii are decreased such that the interfacial stress becomes the main deforming force. The interfacial stress tends to minimize the interfacial area between the two phases. As a consequence, small disturbances present at the interface of the thread will grow and finally result in the breakup of the thread into drops.

The thread can be modelled as a cylinder with a disturbance on the radius. These so-called ‘Rayleigh disturbances’ (named after Lord Rayleigh, who first studied the breakup of water jets in air, taking into account the interfacial tension and inertia of the thread phase [1]) are typically sinusoidal, as shown in Figure 2.1:

\[ h(z) = R + \alpha \sin \left( \frac{2\pi z}{\lambda} \right), \]

with \( h(z) \) the interface position, \( \alpha \) the disturbance amplitude, \( \lambda \) the disturbance wavelength and the average radius

\[ \tilde{R} = \sqrt{R_0^2 - \frac{\alpha^2}{2}} \]

with \( R_0 \) the initial radius. These disturbances will grow for wavelengths greater than the initial thread circumference \( 2\pi R_0 \), due to a monotonic decrease of the interfacial area. Disturbances with wavelengths smaller than the thread circumference will damp, for the growth of these disturbances would yield an (initial) increase of the interfacial area, as can be concluded from a theoretical analysis.

In previous analyses of viscous threads in a viscous matrix by Tomotika [3], predictions have been made for the dominant growth rate (i.e. of the fastest growing disturbance which finally causes breakup), depending only on
the viscosity ratio between the two phases (thread/matrix). The disturbance amplitude is assumed to grow exponentially in time:

\[ \alpha = \alpha_0 e^{qt}, \]  

(2.3)

with growth rate

\[ q = \frac{\sigma \Omega(x, p)}{2\eta_c R_0}. \]  

(2.4)

\( \Omega \) is the dimensionless growth rate of the disturbance, which is a function of the wavenumber \( x \) and the viscosity ratio \( p \), see Figure 2.2:

\[ x = \frac{2\pi R_0}{\lambda}, \]  

(2.5)

\[ p = \frac{\eta_d}{\eta_c}, \]  

(2.6)

with \( \eta_d \) and \( \eta_c \) the viscosity of the dispersed (thread) and continuous (matrix) phase respectively. It is evident that the dominant wavenumber which belongs to the dominant growth rate is also only a function of the viscosity ratio \( p \). Dominant wavenumber \( x_m \) and corresponding growth rate \( \Omega_m \),
indicated by the solid line in Figure 2.2 are shown in Figure 2.3. It should be noted that the growth rate $\Omega$ according to Tomotika stems from a linear (small amplitude) analysis and its validity at greater amplitudes, especially when approaching breakup, needs verification.

### 2.2 Problem Definition

With the aforementioned theoretical considerations in mind, a finite element model was implemented, first in MATLAB [10] (which is a flexible environment during the development stage) and later (for computational economy) in the SEPRAN [11] finite element package. The flow is considered to be inertialess and the material an incompressible liquid, which reduces the balance equations of mass and momentum to:

$$ \nabla \cdot \vec{u} = 0, $$

(2.7)

with $\nabla$ the gradient operator and $\vec{u}$ the velocity vector and

$$ \nabla \cdot \sigma = 0, $$

(2.8)

with $\sigma$ the Cauchy stress tensor:

$$ \sigma = -pI + \tau, $$

(2.9)

split into the contribution from the pressure $p_a$ and the extra stress tensor $\tau$ that is given by a constitutive equation. When viscous flow is considered (for the matrix material and in the viscous calculations also for the thread material) the Newtonian constitutive equation holds:

$$ \tau = 2\eta D, $$

(2.10)
with $\eta$ the viscosity and $D$ the deformation rate tensor. For the viscoelastic calculations the Phan-Thien Tanner (PTT) model was used as constitutive equation, with the extra stress tensor:

$$\tau = \tau_s + \sum_{i=1}^{n} \tau_i,$$

(2.11)

consisting of a viscous 'solvent' mode $\tau_s$ and $n$ viscoelastic modes $\tau_i$. The solvent contribution is again given by Newtonian constitutive equation:

$$\tau = 2\eta_s D,$$

(2.12)

with $\eta_s$ the 'solvent viscosity'. The contributions of the viscoelastic modes to the extra stress tensor are given by:

$$\theta_i \nabla \tau_i \left( 1 + \frac{\varepsilon_i \theta_i}{\eta_i} \text{tr}(\tau_i) \right) \tau_i = 2\eta_i D,$$

(2.13)

with $\theta_i$ the relaxation time, $\eta_i$ the viscosity and $\varepsilon_i$ the non-linearity parameter of mode $i$. From Equation 2.13 it follows that the solvent mode can in fact be seen as a viscoelastic mode with $\theta = 0$, which is how it is implemented in the model. $\nabla \tau_i$ represents the Truesdell rate:

$$\nabla \tau = \dot{\tau} - L \cdot \tau - \tau \cdot L,$$

(2.14)

with $L$ the velocity gradient tensor and $\dot{\tau}$ the material time derivative related to the spatial time derivative by:

$$\dot{\tau} = \frac{\partial \tau}{\partial t} + \ddot{\tau} \cdot \nabla \tau.$$

(2.15)

Although multiple modes are needed to describe measured rheological data over a wide range of deformation rates, the number of modes used for most of the calculations presented in this report was only 1 plus the 'solvent mode', due to the excessive computation times which arise from multi-mode problems. (The computation time is proportional to the number of modes used). For a few simulations a 5 mode fit by Janssen [9] (see Appendix A) was used.

Since the breakup takes place at low Strouhal and Reynolds numbers, a quasi steady approach can be used, even though the flow problem is inherently transient. The Strouhal number indicates the ratio between local acceleration and acceleration due to non-uniformity of the flow, while the Reynolds number indicates the ratio between inertia and viscous forces.

In Figure 2.4 an example of a mesh is shown with boundary conditions. When present, the unbounded matrix is approximated in radial direction by 10 times the undisturbed thread ratio (some simulations were done for viscoelastic threads in air, in which case the matrix viscosity can be neglected and therefore is not modelled). In axial direction the mesh is considered to be a repeating unit with symmetry at the ends. In this way mass is conserved.
in both phases and the interface is parallel to the thread axis at both ends of the thread. This approach implies that the length of the domain should be a multiple of $\frac{\lambda_m}{2}$, with $\lambda_m$ the dominant wavelength. Calculations have been done for domain lengths of $\frac{\lambda_m}{2}$, with an initially sinusoidal disturbance of the interface and $4 \cdot \frac{\lambda_m}{2}$, with an initially random disturbance of the interface, which is more in agreement with the real situation before breakup.

The interfacial tension $\sigma$, which is the driving force for the deformation in the problems considered, causes a normal stress jump at the interface between thread and matrix:

$$\Delta(\sigma : \bar{n}\bar{n}) = \sigma \left( \frac{1}{R_1} + \frac{1}{R_2} \right), \quad (2.16)$$

with $\Delta$ the difference between thread and matrix phase and $R_1$ and $R_2$ the main radii of curvature. $R_1$ is the radius in the r- z plane and $R_2$ in a perpendicular plane. These radii depend only on the shape of the interface $h(z)$ [12]:

$$R_1 = \left( 1 + \left( \frac{\partial h}{\partial z} \right)^2 \right)^{-\frac{1}{2}} \quad (2.17)$$

and

$$R_2 = -h \left( 1 + \left( \frac{\partial h}{\partial z} \right)^2 \right)^{-\frac{1}{2}}, \quad (2.18)$$

The evolution of the interface position can be determined through the kinematic condition:

$$\frac{\partial h}{\partial t} + u_z \frac{\partial h}{\partial z} = u_r. \quad (2.19)$$
2.3 The Finite Element Model

The obtained set of equations is solved using a mixed \((\tau, \bar{u}, p)\) formulation [13]. Due to axisymmetry the calculations can be performed in cylinder coordinates (quasi 2-D). The stress and velocity field are interpolated quadratically, while a bilinear interpolation is used for the pressure field. An operator splitting methodology according to Baaijens [13] is adopted to extract the advective parts of the constitutive equation, which are solved by application of a Time Discontinuous / Galerkin Least Squares (TD/GLS) method. Since the interpolations for stress and pressure are chosen discontinuous across the element boundaries, they can be eliminated on the element level, yielding an efficient solution algorithm.

The interface position \(h\) is solved in a separate problem after the velocity field is determined with the use of a Streamline Upwind Petrov-Galerkin (SUPG) algorithm [14]. It should be noted that, following from Equations 2.17 and 2.18, the discrete representation of the interface must be of class \(C^1\), i.e. twice differentiable, to preserve conformity. However, it is possible to integrate by parts the terms involving a second order derivative of the interface position, after which a formulation is obtained for which a representation of class \(C^0\) is admissible [5], thus giving the possibility to use linear elements to describe the interface position \(h\). For the calculations presented in this study quadratic elements were used to describe the interface.

The steady solution of the velocity field is used to displace the mesh for the next timestep, using an Arbitrary Lagrange Euler (ALE) algorithm. The mesh is rescaled in radial direction to keep the elements within the thread as well as in the matrix of the same relative size, while the mesh is not deformed in axial direction. This method has significant advantages over a purely Lagrangian approach (which leads to an unacceptable deformation of the elements) or Eulerian approach (which gives complications because the interface does not necessarily coincide with the nodal points and an interpolation procedure is needed).

The increment size is set at the beginning of the calculations, but decreases when the largest radial node displacement within one increment at the interface exceeds a user-defined fraction of the minimum thread radius, thus preventing the interface from reaching negative values due to discretisation in time:

\[
\Delta t = \min \left\{ \Delta t_0, \mu \frac{\min \{h(z)\forall z\}}{\max \{u_z, h(z), z\forall z\}} \right\}.
\]  
(2.20)

Here \(\Delta t_0\) indicates the initial increment size (typically 0.01 [s]) and \(\mu\) the user defined maximum decrease of \(h\) within one increment (typically \(2.5 \cdot 10^{-3}\) [-]).
Chapter 3

Viscous Threads

3.1 Experimental Results

The experiments by Janssen [9] referred to in this section, were carried out using viscous model liquids at room temperature (castor oil and silicon oil). The results are in close agreement with Tomotika's [3] linear analysis and will be discussed only briefly.

In Figure 3.1 a few stages of the experiments are shown. The results clearly show the growth of a sinusoidal disturbance on the interface, at least during the initial stage of the breakup process. The measured amplitude $\alpha$ shows exponential growth, which is in agreement with the theory. Both the dominant growth rate $\Omega_m$, which can be determined from $\alpha(t)$ (with use of Equation 2.4) and the corresponding wave number $x_m$ are in close agreement with the theoretical values.

Figure 3.1: Experimental results of the breakup of a viscous thread (castor oil, $\eta_d = 0.74$ [Pa.s]) in a viscous matrix (silicon oil, $\eta_e = 0.94$ [Pa.s]), $\sigma = 4.1 \cdot 10^{-3}$ [N/m]; photographs every second.
3.2 Computational Results

The numerical simulations of the breakup process of viscous threads in a viscous matrix were done with a mesh which describes a part of $\frac{L_m}{2}$ of the thread length, with $\lambda_m$ corresponding to the dominant wave number $x_m$ of the matrix-thread configuration under consideration. As discussed before this is allowed due to symmetry considerations and simulations with various lengths yield a dominant wavenumber for this problem which is indeed in agreement with the theory. In Figure 3.2 a typical simulation is shown. The

![Figure 3.2: Calculated velocity field and interface of a viscous thread in a viscous matrix.](image)

The initial radius of the thread is $1$ [mm], with an initial disturbance amplitude of $10^{-2}$ [mm]. The viscosity of both the matrix and thread phase is $1$ [Pa.s] and the interface tension $10^{-2}$ [N/m]. These values are comparable to those of the model liquids used in the experiments described in the previous section.

Figure 3.3 shows the amplitude growth in time for this simulation (full line), compared to the prediction according to Tomotika’s theory (dashed line). The exponential growth rate agrees with the predicted value of the theory, up to very high deformations. The error (less than 10%, even at $\frac{\sigma}{\eta_0} = 0.6$) in Tomotika’s theory stems from the fact that non-linear effects start to play a role when breakup is approached. The formation of satellite drops is initiated (see Figure 3.1) and it is no longer valid to describe the disturbance as a perfect sine wave in this stage. Since the finite element implementation of the problem describes the interface location with a height function, it is not possible to simulate the breakup process until the actual breakup, although local mesh refinement in the neck region of the thread might give accurate results close before this moment.

Decreasing the amplitude of the initial disturbance yields results identical to those of the simulation described, except for a longer time until breakup. If a random noise is prescribed as the initial disturbance (which
Figure 3.3: Amplitude growth in time for the simulation and according to Tomotika's theory.

is in agreement with the actual situation during the breakup process) at a simulated length of $4 \cdot \Delta m$, the predicted dominant wave still emerges. A variation of the viscosity ratio $p = \frac{\nu_d}{\nu_c}$ in a range of $10^{-2}$ to $10^2 [-]$ also yields growth rates and dominant wave numbers which are in agreement with theoretically predicted values.
Chapter 4

Viscoelastic Threads

4.1 Experimental Results

This chapter focuses on the behaviour of viscoelastic threads. For the simulations of viscoelastic threads in a viscous matrix, comparisons are made to experimental results from Janssen [9]. Figure 4.1 shows a typical experiment with a viscoelastic model liquid in the thread phase (80 % corn syrup / 20 % water / 0.01 % polyacrylamide, see Appendix A). In contrast to the viscous experiments, in which a sinusoidal disturbance could be seen almost until the point of breakup, these experiments show the development of dumbbell shapes. The thin filaments which emerge inbetween the droplets are relatively stable and slow down the breakup process considerably. The liquid in the filaments slowly drains into the droplets, until the filament breaks and the loose ends retract toward the droplets. All viscoelastic liquids which were studied show similar results.

The typical breakup behaviour which is seen for these viscoelastic threads can be explained from the build up of orientational stresses. Due to the high strain rates, the elongational viscosity drastically increases and thus slows down the elongational flow in the filaments. The orientational stresses also arise from the deformation applied to the material prior to the breakup process, during the formation of the thread. In the next section the development of local stresses, as well as the influence of initial stresses on the breakup behaviour is studied with the use of finite element simulations.

4.2 Computational Results

First, simulations of viscoelastic threads in air are considered, in which case the influence of the matrix can be neglected and only the thread and the moving interface need to be modelled. The simulations were performed according to previous finite element calculations by Bousfield et. al. [4], using various meshes to investigate the influence of discretisation errors.

The material is described using the PTT constitutive model with $\varepsilon = 0$, effectively resulting in an Oldroyd-B model. The remaining parameters are
Figure 4.1: Experimental results of the breakup of a viscoelastic thread (80 % corn syrup / 20 % water / 0.01 % polyacrylamide) in a viscous matrix (silicon oil, \( \eta_e = 2.0 \text{[Pa.s]} \), \( \sigma = 1.8 \times 10^{-2} \text{[N/m]} \); photographs every 3 seconds.

\[ \eta_s = 0.25 \text{[Pa.s]}, \quad \eta_l = 0.75 \text{[Pa.s]} \text{ and } \theta_l = 30 \text{[s].} \]

The thread radius \( R_0 = 1 \text{[m]} \) at a domain length \( L = 10 \text{[m]} \), while the surface tension \( \sigma = 1 \text{[N/m]} \). Note that these values are chosen to comply with the dimensionless parameters introduced in Bousfield et. al. [4] (see Appendix B) and are not necessarily physically realistic.

A typical example of the development of the velocity field and interface during such a simulation is shown in Figure 4.2. This simulation very clearly shows the development of drops connected by stable filaments. After circa 15 seconds the breakup process is considerably retarded; the connecting filament shows no development of further disturbances and tends to stay mostly cylindrical instead of developing into a satellite drop. The flow field however shows that the drainage of liquid from the filament into the drop, which is observed in the experiments, does not take place. The liquid close to the drop tends to flow in the direction of the filament, which indicates that a satellite drop will eventually emerge.

If an insufficiently fine mesh is used (less than \( 40 \) elements to describe the interface), the connecting filament quickly becomes unstable and breaks. Examples of simulations with various mesh sizes are given in Appendix C.

The stability of the thread in these experiments may be caused by the use of the Oldroyd-B constitutive model, which gives relatively high elongational stresses under uniaxial strain. If a PTT model is introduced by
Figure 4.2: Calculated velocity field and interface of a viscoelastic thread in air.

setting the non-linearity parameter $\varepsilon$ to 0.01 [-], the simulations show that the filaments become significantly more unstable, which is in agreement with this assumption.

The following simulations investigate the behaviour of viscoelastic threads in a viscous matrix, with a mesh which describes a part of $4 \cdot \lambda_m^2$. It should be noted that the definition of $\lambda_m$ stems from the linear viscous analysis by Tomotika and should be used with care in the viscoelastic calculations, as it depends on the viscosity ratio $p = \frac{\eta_d}{\eta_c}$. For the viscoelastic case we define

$$\eta_d = \eta_s + \sum_{i=1}^{n} \eta_i,$$  \hspace{1cm} (4.1)

which is applicable in the initial stage of the breakup, when no orientational stresses have build up yet. This definition loses its validity if initial stresses, due to stretching of the thread before the actual interfacial tension driven breakup process, are taken into account. By choosing a domain length which is a significant factor longer than $\frac{\lambda_m}{2}$, it is hoped that the influence of incorrect boundary conditions due to a deviation of $\lambda_m$ is minimised, at least in the central part of the domain. Due to limited computational resources the mesh width is chosen only 60 elements, i.e. 15 elements per $\frac{\lambda_m}{2}$, which is obviously less than the advised 40 elements.

In Figure 4.3 a typical example of a simulation is shown. The initial
radius of the thread is 1 [mm] and a random disturbance (white noise, $\alpha$ is $10^{-4}$ [mm]) is initially applied. The viscosity of the viscous matrix equals 1 [Pa.s], while a 1 mode PTT fit is used for the thread phase, with $\eta_s = 0.6$ [Pa.s], $\eta_1 = 0.4$ [Pa.s], $\theta_1 = 15$ [s] and $\varepsilon_1 = 0.01$ [-], yielding a viscosity ratio $\rho = 1$ [-]. The dimensionless parameters introduced by Bousfield et al. can be calculated for this case and yield values comparable to the calculations in air, as shown in Appendix B. The simulation shows the development of drops with a filament in between, but unlike the experiments this filament is rather unstable and quickly starts to deform further into a smaller satellite drop.

For some simulations a uniform initial stress distribution was applied resulting from a uniaxial strain simulation at various strain rates ($\dot{\varepsilon} = 0.0, 0.1, 0.2, 0.3, 0.4$ [s$^{-1}$]) to account for the stretching of the thread before breakup. These simulations gave similar results as those shown in Fig. 4.3, only giving longer breakup times at higher initial stresses. The initial growth rate $\Omega$ as a function of time for these simulations is shown in Fig 4.4. An initial peak in $\Omega$ is seen when initial stresses are taken into account, which becomes higher when the initial stresses are higher. It is not clear if this phenomenon is present in the experiments, since the absolute growth rate $\dot{\alpha}$ is very small during the initial stage of the breakup process. The dimensionless growth rate then quickly (within $\frac{1}{10}$ of $\theta$) falls back to a value below Tomotika's prediction for the equivalent viscous experiment at viscosity ratio $\rho = 1$ [-]. If no initial stresses are taken into account, $\Omega$ is
approximately equal to Tomotika's prediction until very close before breakup. This is not in agreement with experimental results, where the breakup process is retarded due to the formation of stable filaments in a very early stage. It can be concluded that the initial stresses due to the stretching of the thread have a significant influence on the breakup behaviour of viscoelastic threads.

In Figure 4.5 the development in time of the axial stress $\tau_{zz}$ at the symmetry axis of the thread is shown in the neck and bulb of a simulation with prescribed initial stresses ($\dot{i} = 0.4 \text{ [s}^{-1}\text{]}$) and compared to the stress relaxation when no deformation takes place ('fixed point'). It clearly shows the build up of elongational stresses in the connecting filaments between the drop, which quickly increase as the filament becomes thinner. Although this phenomenon was expected from the model experiments, it is not sufficiently present in these calculations to prevent the breakup of the filament in an early stage.

Part of the problem is the use of an insufficient constitutive description of the viscoelastic material. Calculations using a 5 mode PTT fit (see Appendix A) give better results as shown in Figure 4.6. Although the development of a satellite drop is still initiated, the simulation using a 5 mode fit shows a significant retardation of the breakup process after circa 8 [s]. For computational economy only a domain of $\frac{\lambda}{2}$ is used for these calculations. A simulation with a 1 mode fit under identical conditions is given to show the improvement using a more accurate material description.
Figure 4.5: Calculated stresses $\tau_{xx}$ at the center of the thread.

5 mode fit
$t=6.0$  6.9  8.0  8.9  9.4 [s]

Figure 4.6: Comparison between a 1 and 5 mode PTT fit.

1 mode fit
$t=6.0$  6.9  7.8  8.9  9.1 [s]
Chapter 5

The Influence of Disturbances of the Interfacial Tension

5.1 Some Considerations

The simulations previously described have all assumed the interfacial tension to be a constant over the interface. However, this may not always be the case, especially when the viscoelastic thread is a solution of polymer chains in a viscous solvent like the model liquids used in Janssen’s experiments, which is not necessarily homogeneous. The concentration of polymer chains at the interface, acting as a surfactant may vary due to diffusional and convectional processes within the thread and thus cause a disturbance of the interfacial tension along the interface. In the next section some simulations with a non-constant interfacial tension are described. It should be noted that the surfactant itself is not (yet) modelled, which implies that other possible effects of its presence, such as so-called Marangoni flows, following from tangential stresses due to surfactant gradients along the interface, are not taken into account. However, Stone [15] indicates that the assumption that the only effect of the surfactant is modifying the surface tension is quite reasonable at low surfactant concentrations.

5.2 Computational Results

For these simulations a sinusoidal disturbance of the interfacial tension \( \sigma \) is introduced, defined (in accordance with Equation 2.1) as

\[
\sigma(z) = \sigma_0 + \beta \sin \left( \frac{2\pi z}{\lambda_\sigma} \right),
\]

(5.1)

with \( \sigma_0 \) the average surface tension, \( \beta \) the disturbance amplitude and \( \lambda_\sigma \) the wavelength of the disturbance. To be able to compare the influence of the
the disturbances of the surface tension to the ‘Rayleigh disturbances’ on the interface, we define

\[ \chi = \frac{\beta \hat{R}}{\alpha \sigma} \]  

(5.2)

as the disturbance ratio between the interfacial tension and the interface itself. During the initial stage of breakup \( \hat{R} \) can be replaced by \( R_0 \), since \( \alpha \ll R_0 \) (see Equation 2.2). Note that negative values of \( \chi \) indicate a disturbance of the interfacial tension which is in counterphase with the disturbance of the interface.

We consider a domain of \( \lambda_m = \lambda_m \) and look at the initial behaviour of the thread for various \( \chi \). Calculations were performed for \(-10 \leq \chi \leq 10 [-]\) and the growth rate of the interface was studied. Figure 5.1 shows the relation between the dimensionless growth rate \( \Omega \) and the disturbance ratio \( \chi \). The results show a significant influence of a disturbance of

![Figure 5.1: Calculated dimensionless growth rate \( \Omega \) for various disturbance ratios \( \chi \).](image)

the interfacial tension on the initial growth rate of interfacial disturbances, even yielding negative values (damping) when \( \chi \) becomes smaller than a critical value \( \chi_c \approx -3.5 [-]\). Various calculations to verify the consistency of the results with different \( \sigma, \alpha \) and \( \beta \) show that the parameter \( \chi \) is indeed determinative for the initial breakup behaviour under the prescribed conditions.

A relationship between the wavelength of the interfacial tension disturbance \( \lambda_\sigma \) and \( \chi_c \) can not be established, since a non-constant initial growth rate is found if the wavelengths of the interfacial tension disturbance and the interfacial disturbance differ. This is caused by the fact that the definition of \( \chi \) loses its validity under these conditions, since the local ratio between interface and interfacial tension gradient is no longer a constant, while the effect of the interfacial tension disturbance depends on the gradi-
ent of the interfacial tension along the interface. However, simulations for various wavelengths of the interfacial tension disturbance do indicate that the influence of these disturbances becomes smaller at greater wavelengths.

In figure 5.2 a simulation is shown over a domain of $4 \cdot \frac{\lambda_m}{2}$, with $\lambda_0 = 2 \cdot \lambda_m$. The interfacial tension has its maximum value at the domain boundaries and its minimum value at the center of the domain, with an amplitude of $20 \times$ the interfacial disturbance amplitude. The results clearly show that the interfacial disturbance in the region with a maximum interfacial tension grows at a higher rate than in the corresponding simulations with a constant interfacial tension, while the interfacial disturbance in the region with a minimum interfacial tension does not show any significant growth at all.

![Figure 5.2: Simulation with surface tension disturbance.](image)

and its minimum value at the center of the domain, with an amplitude of $20 \times$ the interfacial disturbance amplitude. The results clearly show that the interfacial disturbance in the region with a maximum interfacial tension grows at a higher rate than in the corresponding simulations with a constant interfacial tension, while the interfacial disturbance in the region with a minimum interfacial tension does not show any significant growth at all.
Chapter 6

Conclusions and Recommendations

• The simulations for viscous threads prove Tomotika’s linear analysis to be valid up to very large deformations. Only in the last stage of the breakup process deviations occur due to geometrical non-linearities.

• The present finite element model is capable of qualitatively describing most of the phenomena occurring during the breakup of viscoelastic threads. Quantitative differences between experiments and simulations still are relatively high.

• The drainage of liquid from the stable connecting filaments into the drops, which is one of the most typical phenomena found in the experiments with viscoelastic threads, can not be reproduced with the present model.

• The presence of initial stresses proves to be an important factor in the breakup process of viscoelastic threads, and at the same time one of the most uncertain. Further research should be done to gain insight in the initial stress distribution, as well as in the mechanisms resulting from these initial stresses and influencing the breakup process.

• Both an accurate description of the viscoelastic material (using a multi mode model) and a sufficiently refined mesh are necessary to describe the experimentally observed retardation of the breakup process due to the formation of stable connecting filaments between the drops.

• Simulations show a significant influence of disturbances of the surface tension on the initial breakup behaviour of the thread, if they are of the same order of magnitude as the interfacial disturbances. Some experimental research is needed to verify if such disturbances do actually occur in the systems considered in this study.
Appendix A

A 5 Mode PTT Fit for the Viscoelastic Model Liquid

The rheological characterization of the viscoelastic model liquid (80% corn syrup / 20% water / 0.01% polyacrylamide), which is modelled in the simulations, was performed by Janssen under simple shear on a Rheometrics RFS-II equipped with a cone and plate geometry. The data were fit to the PTT model described in Chapter 2, using 5 viscoelastic modes. Small amplitude oscillatory shear measurements were used to determine the linear viscoelastic parameters ($\eta$, $\theta$, and $\eta_b$), while the non-linearity parameter $\varepsilon$, which was assumed to be the same for all modes, was estimated from steady shear experiments.

The resulting parameters for the solvent mode and the viscoelastic modes of the best fit are given in the following table:

<table>
<thead>
<tr>
<th>$i$</th>
<th>$\eta$ [Pa.s]</th>
<th>$\theta$ [s]</th>
<th>$\varepsilon$ [-]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.43</td>
<td>0.01</td>
<td>0.02</td>
</tr>
<tr>
<td>1</td>
<td>0.0041</td>
<td>0.01</td>
<td>0.02</td>
</tr>
<tr>
<td>2</td>
<td>0.0466</td>
<td>0.0891</td>
<td>0.02</td>
</tr>
<tr>
<td>3</td>
<td>0.102</td>
<td>0.794</td>
<td>0.02</td>
</tr>
<tr>
<td>4</td>
<td>0.254</td>
<td>7.08</td>
<td>0.02</td>
</tr>
<tr>
<td>5</td>
<td>0.324</td>
<td>63.1</td>
<td>0.02</td>
</tr>
</tbody>
</table>

These values, when combined with a matrix with viscosity 1 [Pa.s], yield an initial viscosity ratio $p = 1.16 [-]$, which is close to value found for the 1 mode fit ($p = 1 [-]$), so changes in dimensionless growth rate $\Omega_m$ or wavenumber $x_m$ are negligible. The measured data and the corresponding PTT fits are shown in figure A.1. The top graph shows the dynamic viscosity $\eta'$, rigidity $\eta''$ and phase angle $\delta$ versus the angular velocity $\omega$. The bottom graph shows the shear viscosity $\eta$, and first normal stress difference $N_1$ versus the shear rate $\dot{\gamma}$ and the predicted elongational viscosity $\eta_e$ versus elongation rate $\dot{\varepsilon}$. For the elongational viscosity there are no measured data available, so only the prediction of the PTT model is given.
Figure A.1: Measured data and best fit for the viscoelastic model liquid
Appendix B

Dimensionless Parameters
According to Keunings

The results of the simulations by Bousfield et. al. [4] are expressed in terms of a set of dimensionless variables as follows:

\[ r = \frac{R}{R_0}, \quad (B.1) \]
\[ \zeta = \frac{z}{L}, \quad (B.2) \]
\[ \theta = \frac{\sigma t}{\eta_d R_0}. \quad (B.3) \]

The system is characterized by means of four dimensionless parameters:

\[ \alpha = \frac{R_0}{L}, \quad (B.4) \]
\[ \beta = \left( \frac{\rho \sigma R_0}{\eta_d^2} \right) \left( \frac{L}{R_0} \right)^2, \quad (B.5) \]
\[ \phi = \frac{\lambda_p \sigma}{\eta_d R_0}, \quad (B.6) \]
\[ \Lambda = \frac{\lambda_r}{\lambda_p}, \quad (B.7) \]

where \( \lambda_p \) denotes the polymer relaxation time and \( \lambda_r \) the retardation time, related by

\[ \lambda_r = \frac{\lambda_p \eta_s}{\eta_d}. \quad (B.8) \]

For the viscoelastic simulations in air performed for this research the parameters were chosen as follows:
The dimensionless parameters following from these values are \( \eta_1 = 0.75 \) and Eq. 4.1) (the parameter \( \eta_1 \) in the PTT model)

| \( \eta_s \) | 0.25          |
| \( \eta_d \) | 1.0           |
| \( \lambda_p \) | 30            |
| \( \epsilon \) | 0             |
| \( R_0 \) | 1             |
| \( L \) | 10            |
| \( \alpha \) | 10^{-2}       |
| \( \sigma \) | 1             |
| \( \rho \) | 0             |

(no inertia taken into account)

The dimensionless parameters following from these values are \( \alpha = 0.1 \), \( \beta = 0 \), \( \phi = 30 \) and \( \Lambda = 0.25 \), the same as the ones used for the simulations published by Bousfield.

For the simulations of a viscoelastic thread in a viscous matrix the parameters were chosen as follows:

| \( \eta_s \) | 0.6          |
| \( \eta_d \) | 1.0          |
| \( \lambda_p \) | 15           |
| \( \epsilon \) | 0.01         |
| \( R_0 \) | 10^{-3}      |
| \( L \) | 2.79 \cdot 10^{-2} |
| \( \alpha \) | 10^{-2}      |
| \( \sigma \) | 10^{-2}      |
| \( \rho \) | 0            |

The dimensionless parameters following from these values are \( \alpha = 0.04 \), \( \beta = 0 \), \( \phi = 150 \) and \( \Lambda = 0.6 \). These values are of the same order of magnitude as those found for the calculations by Bousfield.
Appendix C

The Influence of Mesh Refinement

The simulations described in this report are performed with a whole range of meshes, varying from $5 \times 5$ (quadratic) elements for the viscous calculations up to $60 \times 5$ elements for the viscoelastic calculations with a viscous matrix. By using different meshes for simulations under otherwise identical conditions, the influence of the discretisation errors induced by the mesh is examined. In Figure C.1 the results are given for the simulations according to Bousfield with a mesh of $20 \times 4$, $40 \times 4$ and $60 \times 4$ elements. The results clearly indicate a significant influence of the mesh size, especially when breakup is approached. Figure C.2 shows the simulations performed with the 5 mode PTT fit for the thread in a viscous matrix at different meshes ($20 \times 5$ and $60 \times 5$ elements). It is clear that the computation times involved with using a multi mode fit are only worthwhile when a sufficiently refined mesh is used. Increasing the accuracy of the material model at the cost of a less refined mesh does not improve the results of the simulations. The calculations generally indicate that 40 elements or more are needed to describe the interface over a domain of $\frac{1}{2}$ if one wants to be able to describe the typical viscoelastic phenomena of the breakup process.
Figure C.1: Simulations according to Bousfield with various meshes.

Figure C.2: Simulations for 5 mode fit with various meshes.
Bibliography


