Simultaneous Breakup of Multiple Viscous Threads Surrounded by Viscous Liquid

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A scenario is proposed for the simultaneous breakup process of an arrangement of parallel viscous threads. At very close distances, threads will break up via an "in-phase" mode, that is, droplets will be formed at the same axial positions along the threads. In the case of threads that are further apart, breakup will occur via an "out-of-phase" mode, that is, on neighboring threads droplets will be formed at axial positions that are shifted by one-half of a wavelength. The smaller the viscosity ratio between the thread phase and the continuous phase, the closer the threads must be to break up via this in-phase mode. This scenario agrees with recent experimental results.

Introduction

The study of the breakup process of fluid filaments (liquid threads) is a classical problem in fluid mechanics literature which dates back more than a century. As early as 1873 Plateau (1873) studied the instability of a long cylinder of liquid and then, five years later, Rayleigh (1878) was the first to explain the instability of water jets in air as a surface tension driven process. Later on, Taylor (1934) performed experiments to study the formation of emulsions. One of his important observations was that a thread of viscous fluid surrounded by another viscous fluid could be stable in a shear flow. However, upon cessation of the flow, the filament became unstable and finally broke up into a line of equally spaced small droplets. Shortly thereafter Tomotika (1935, 1936) analyzed this phenomenon theoretically. Using a linear theory and taking into account the viscosity of both fluids, as well as interfacial tension, he was able to calculate the velocity field for the case of an infinitely long cylindrical thread in the presence of a small spatially periodic disturbance along the thread. His prediction for the droplet size, as a function of the viscosity ratio of the two fluids, was in good agreement with Taylor's experiments.

In more recent years the linear theory of Tomotika was extended in several ways (Goldin et al., 1969; Stone et al., 1986; Bousfield et al., 1986; Stone and Leal, 1989; Janssen, 1993) all for the case of a single breaking thread. In this article we begin to extend the theory of Tomotika to the situation of several viscous threads immersed in a viscous matrix. Tomotika's linear theory for the one-thread case will be summarized and then a scenario is proposed for the simultaneous breakup of an array of parallel threads. On the basis of a few qualitative/semi-quantitative arguments, both the influence of the distance between the threads and the viscosity ratio between the thread- and the matrix phase on this simultaneous breakup process will be discussed. Finally, the results obtained in a recent experiment (Elemans et al., 1997) are interpreted using this new scenario.

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Linear Theory for a Single Thread

Consider an infinitely long cylindrical thread of a fluid with viscosity μ_d surrounded by another fluid with viscosity μ_c under quiescent conditions. The subscripts d and c denote the disperse and the continuous phases respectively. The ratio of the two viscosities is defined as $p \equiv \mu_d/\mu_c$. (the symbol p is chosen to be in agreement with other publications, although there might be confusion with the pressure.) Because of thermal fluctuations, the thread is not precisely a cylinder. These fluctuations can be modeled as a sum of small periodic rotationally symmetric disturbances with wave number k ($k = 2\pi/\lambda$, where λ is the wave length.). Some of these periodic disturbances will grow driven by the interfacial tension σ, because the area between the two phases is decreased. Others will damp, because they will contribute to an increase in interfacial area. Tomotika determined the velocity field and the growth rate for an arbitrary wave number k, using a linear theory. He considered a disturbed thread with radius R given by

$$R(z,t) = a + \epsilon(t)\cos kz \tag{1}$$

where z is the coordinate along the axis of the thread, ϵ is the amplitude of the disturbance which is a function of time t, and a is the average radius.

Both phases are considered to be incompressible and inertia is neglected so that the system is described by the Stokes equations. The boundary conditions are given by

- No slip at the interface
- Continuity of the tangential stress at the interface
- Discontinuity of the normal stress at the interface due to the interfacial tension.

In summary, the components of the velocity field v_r and v_z, in terms of the stream function $\Psi(r,z,t)$ in and around the thread, are given by

$$v_{r,i} = \frac{1}{r}\frac{\partial \Psi_i}{\partial z};\ v_{z,i} = -\frac{1}{r}\frac{\partial \Psi_i}{\partial r} \quad (i=1,2) \tag{2}$$

where $i=1$ refers to the thread phase and $i=2$ refers to the matrix phase and where

$$\Psi_1(r,z,t) \equiv \left[A_1(t) r I_1(kr) + A_2(t) r^2 I_0(kr)\right]\sin kz \quad (r<a) \tag{3}$$

$$\Psi_2(r,z,t) \equiv \left[B_1(t) r K_1(kr) + B_2(t) r^2 K_0(kr)\right]\sin kz \quad (r>a) \tag{4}$$

where I_n and K_n are modified Bessel functions of order n, and the coefficients A_1, A_2, B_1 and B_2 are determined by the boundary conditions. Explicit expressions for these coefficients can be found in the Appendix. Full details can be found in Tomotika (1935). The disturbance amplitude is assumed to grow exponentially, that is, $\epsilon(t) = \epsilon_0 \exp qt$, with ϵ_0 the initial amplitude and q the growth rate. This growth rate can be written as (Tomotika, 1935)

$$q(k,p) = \frac{\sigma}{2\mu a}\Omega(x,p) \tag{5}$$

where $\Omega(x,p)$ is a dimensionless growth rate and x is a dimensionless wave number defined by $x \equiv ka$. For $q>0$, disturbances are growing, and for $q<0$, disturbances will damp out. The dimensionless wave number $x_{\max}$ of the fastest growing disturbance is the wave number for which $\Omega(x,p)$ is maximal. In Figure 1 the value of $x_{\max}$, as well as the corresponding dimensionless growth rate $\Omega_{\max} \equiv \Omega(x_{\max},p)$, can be found as a function of the viscosity ratio p. The size of the droplets which are formed is determined by this wave number. The volume of the new droplet is the same as the volume corresponding to one wave length of the initial disturbed thread, so $R_{\text{droplet}} = \sqrt[3]{3\pi/2x_{\max}a}$. Tomotika's theoretical results (Tomotika, 1935) for the droplet size are in agreement with the results of Taylor's (1934) experiments.

In Figure 2 the velocity fields v_r and v_z are plotted for that k value with maximum growth rate and for two different values of the viscosity ratio.

As both velocity components v_r and v_z are of the form $v = f(r).g(z)$, it is only the r-dependency at a certain z-coordinate which is shown in this figure. The particular z-coordinate chosen here is given by $2\pi z/\lambda = \pi/4$, that is, $z = \lambda/8$. The r-dependency of the radial and axial velocity components (v_r and v_z) varies with the viscosity ratio p, as can be seen in these two figures. The effect of the viscosity ratio is exaggerated by taking $p=4$. In our experiments, p varies only over one decade. In the next section, we will analyze the simultaneous breakup process of two parallel threads using the above solution for the velocity field in and around a single thread.

Approximation for Two Threads

Consider two identical parallel threads both with a radius equal to a. The distance between the centers of the two

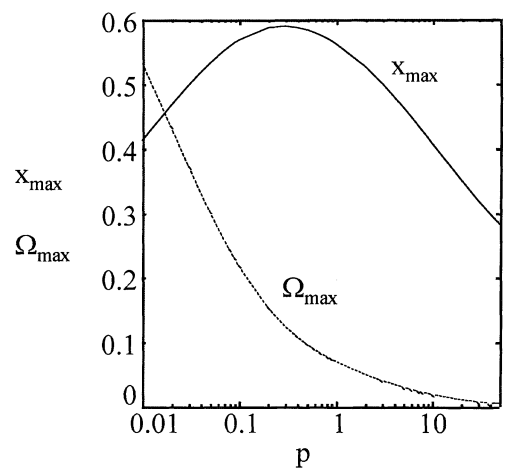

Figure 1. Dimensionless wave number $x_{\max}$ and its corresponding dimensionless growth rate $\Omega_{\max}$ as a function of the viscosity ratio p.

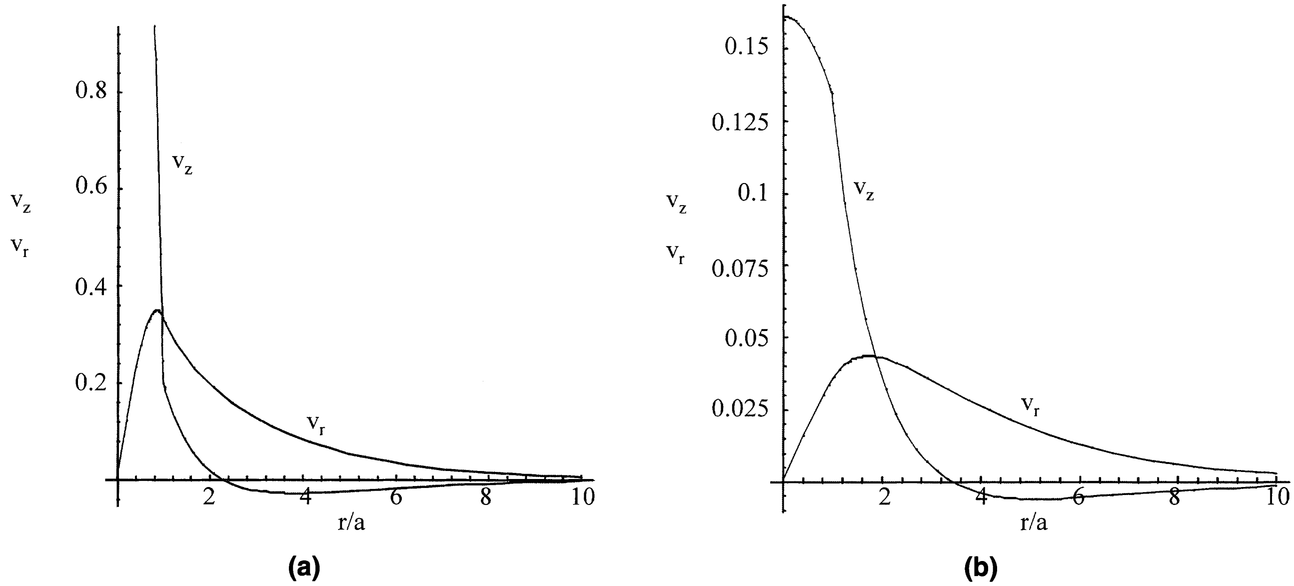

Figure 2. Velocities in the *r*- and *z*-direction for respectively $p = 0.04$ (Figure 2a) and $p = 4$ (Figure 2b).

threads is b. Both threads exhibit small thermal fluctuations. We want to study the phase relation between the disturbances of both threads, therefore, we assume that both threads have a periodic, rotationally symmetric disturbance with wave number k, while the phase difference between the two disturbances is α. Thus, the radii of the threads are given by

$$R_1(z,t) = a + \epsilon(t)\cos kz \tag{6}$$

$$R_2(z,t) = a + \epsilon(t)\cos(kz - \alpha) \tag{7}$$

Let us suppose that the threads are so far apart that the interaction between them can be neglected. Then, because of the linearity of the Stokes equations, the two solutions for the velocity fields around each thread can be superposed. So, the total velocity field is then given by $\boldsymbol{v} = \boldsymbol{v}_1 + \boldsymbol{v}_2$, where $\boldsymbol{v}_i$ is the velocity field generated by the disturbance at thread i if there were no other thread present.

The phase relation between the two threads can be determined by calculating the dissipation D in the surrounding fluid. The phase difference will be such that the dissipation will be minimal. The dissipation D is defined as

$$D = \int_V \mu \boldsymbol{D}:\boldsymbol{D} dV \tag{8}$$

where $\boldsymbol{D}$ is the rate of deformation tensor, given by $\boldsymbol{D} = \frac{1}{2}(\nabla \boldsymbol{v} + (\nabla \boldsymbol{v})^{\mathrm{T}})$. Because of the periodicity, we restrict the z-part of the volume integration to one wave length along the thread. After performing this integration, the dissipation can be written as

$$D = D_{11} + D_{22} + D_{12}(b)\cos\alpha \tag{9}$$

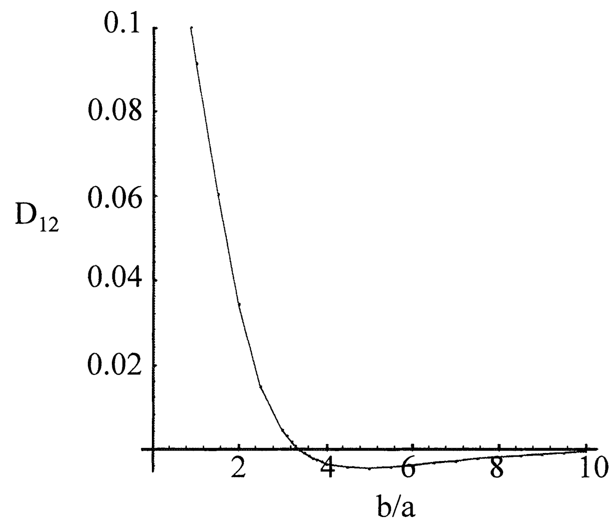

Figure 3. Interaction dissipation D_{12} for the case of $p = 0.91$.

where $D_{11} = D_{22}$ is the dissipation caused by $\boldsymbol{v}_1$ or $\boldsymbol{v}_2$, respectively. These terms are independent of b. $D_{12}(b)$ is the dissipation caused by the cross term in $\boldsymbol{D}:\boldsymbol{D}$ of $\boldsymbol{v}_1$ and $\boldsymbol{v}_2$ and depends on the distance between the two threads. $D_{12}(b)$ is plotted in Figure 3 for $p = 0.91$, a case also considered by Tomotika (1935).

For $b < 3.4$, D_{12} is positive so the dissipation D is then minimal for $\alpha = \pi$. Hence, the fastest growing disturbance occurs when the two threads break up via an out-of-phase mode. For $b > 3.4$, D_{12} is negative, so D is now minimal for $\alpha = 0$ and the two threads will break up via an in-phase mode. However, these phase relations will only be observed if the loss in dissipation is outweighed by the thermal noise. Otherwise, there is no phase relation between the breakup processes of both threads.

Scenario for the Breakup Process of Two Threads

The previous analysis is, strictly speaking, only valid if the two threads are far apart. If the threads are close together, the solution discussed for the total velocity field does not obey the boundary conditions at both cylinders. Nevertheless, based on the character of the solution for one thread, we can try to match the velocity field belonging to each of the threads such that the boundary conditions at the cylinders are satisfied as well as possible. This approach will lead to a scenario for the simultaneous breakup process of two parallel liquid threads.

Assume that the phase of the disturbance on thread 1 is fixed and consider the associated velocity field as if there were no other thread present. Next, we consider the disturbance on thread 2 due to this velocity field, assuming that only the phase of this disturbance can be different. For the moment, we ignore the fact that in reality the deformation of the threads is no longer axisymmetric. The wave number of the disturbance on thread 2 is the same as for thread 1. Of course, the part of the surface of thread 2, which is closest to thread 1, experiences the largest influence of thread 1.

Suppose that just outside thread 1, the z-component of the velocity field dominates (see Figure 2b). So, if thread 2 is close to thread 1, it experiences mainly the z-component of the velocity field of thread 1. Because of the no-slip condition, the velocity in the z-direction of the second thread will have the same direction as that of the first thread. Therefore, the disturbances on the second thread will follow the disturbances on the first one, and, on thread 2, maxima in amplitude will form at identical axial positions as on thread 1 in order to satisfy the conservation of mass. Hence, the threads will break up via an in-phase mode ($\alpha = 0$).

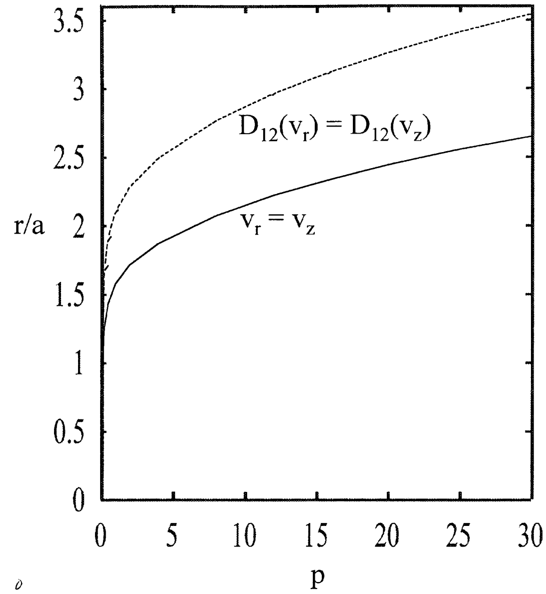

Figure 5. Boundaries between in-phase and out-of-phase regions based on the two criteria: $v_r = v_z$ and $D_{12}(v_r) = D_{12}(v_z)$.

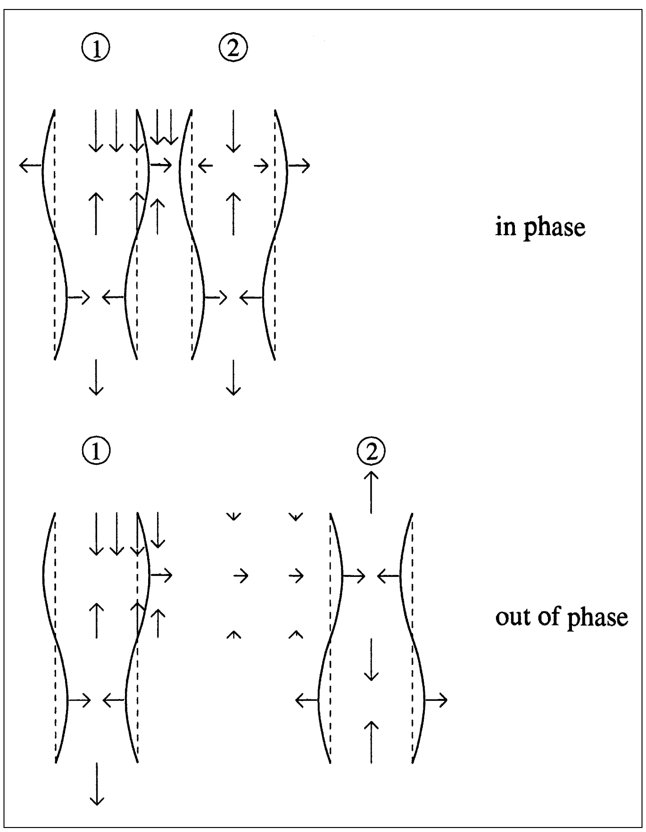

Figure 4. Phase relation between two disintegrating threads on the basis of the velocity field of one thread.

The arrows indicate the magnitude of the velocities v_r and v_z (not to scale).

At larger distances from thread 1, the r-component of its velocity field is dominant. If the second thread is located in that region, its disturbance will adapt to this velocity component. Maxima will be formed at those axial positions where the first thread is constricting, and, thus, the threads will break up via an "out-of-phase" mode ($\alpha = \pi$). These two cases are illustrated in Figure 4.

There are several possible criteria for the estimation of the critical distance between two threads where the mode of breakup changes from "in-phase mode" to "out-of-phase," that is where the r-component of the velocity field starts to dominate over the z-component. For example, the distance at a given point along the threads where these two velocity components are equal $v_r(z) = v_z(z)$, or where the contribution of v_r to the dissipation D equals that of v_z. The boundaries corresponding to these two criteria are plotted in Figure 5 as a function of the viscosity ratio. Although we realize that the viscosity ratios shown in this figure are much larger than those of the model systems that were used in the experiments, we deliberately exaggerated them in this figure to clarify the difference between the dividing lines based on the two criteria for change in the breakup mode.

It is to be expected that a more accurate estimate of this boundary will confirm the trend, as shown in Figure 5, that the critical distance increases with increasing p.

In conclusion, we want to propose the following scenario for the breakup of two parallel threads. If the distance b between the threads is small, breakup will occur via an "in-phase mode. When this b exceeds a certain critical value b_c, the threads will break up via an "out-of-phase" mode. Beyond an even larger distance b_T, breakup will not favor any specific phase relation between the breakup processes of the threads, because the thermal noise contribution to the total velocity field will dominate. The smaller the viscosity ratio p between the thread phase and the matrix phase, the smaller b_c will be.

Comparison of the Scenario with Experimental Results

The present scenario is compared with the results of model experiments that were performed to study the breakup of a planar array of parallel molten nylon-6 threads embedded in a molten poly(styrene) matrix. The experimental procedures are described in detail in Elemans et al. (1997). In the experiments, two types of poly(styrene) are used (SHELL PS N1000 and PS N7000, respectively), which have a different zero shear-rate viscosity. In combination with nylon-6 (Ultramid B3, BASF) as the thread phase, the viscosity ratio at 230°C (the temperature at which the experiments were performed) is 0.6 in the case of PS N1000 and 0.04 in the case of PS N7000.

Upon viewing the videotapes of the experiments involving PS N1000 as the matrix phase, it appeared that the "in-phase" behavior was observed for threads which were at small intermediate distances (Figure 6). In experiments where the threads were further apart, no distinct mode of breakup could be discerned. When the higher molecular PS N7000 was used as matrix polymer, the threads clearly broke up via an "out-of-phase" mode (Figure 7). This is in fair agreement with the presently proposed scenario. Changing the matrix polymer from PS N1000 into PS N7000 means a 15-fold decrease in viscosity ratio. The "in-phase" region, as mentioned in the previous section, is smaller, and the out-of-phase mode of breakup is more likely to occur.

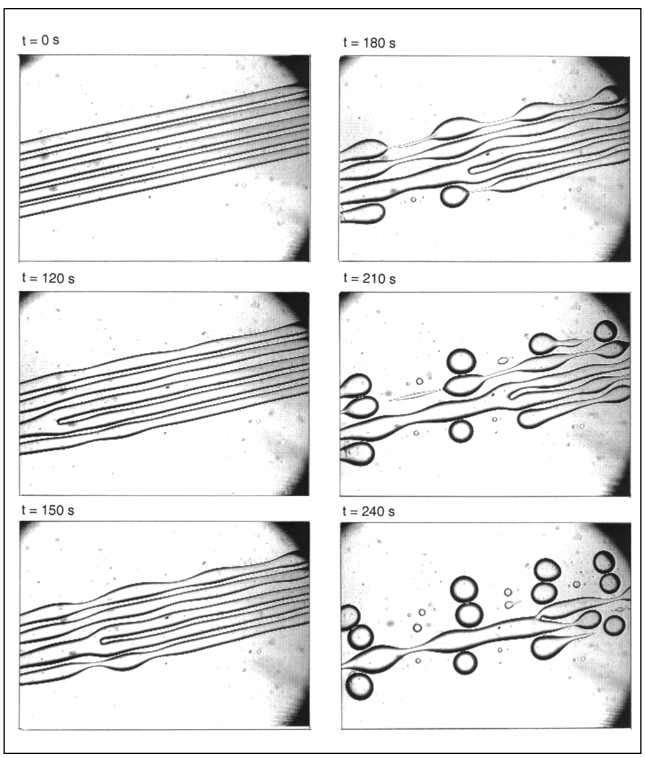

Figure 6. Process of the in-phase breakup of PA-6 threads in a matrix of PS N1000.

The photos were taken from a video screen at the following times: $t = 0$, 120, 150, 180, 210 and 240 s. The initial thread diameter was 70 μm. The viscosity ratio $p = 0.6$. The measurement was performed at 230°C.

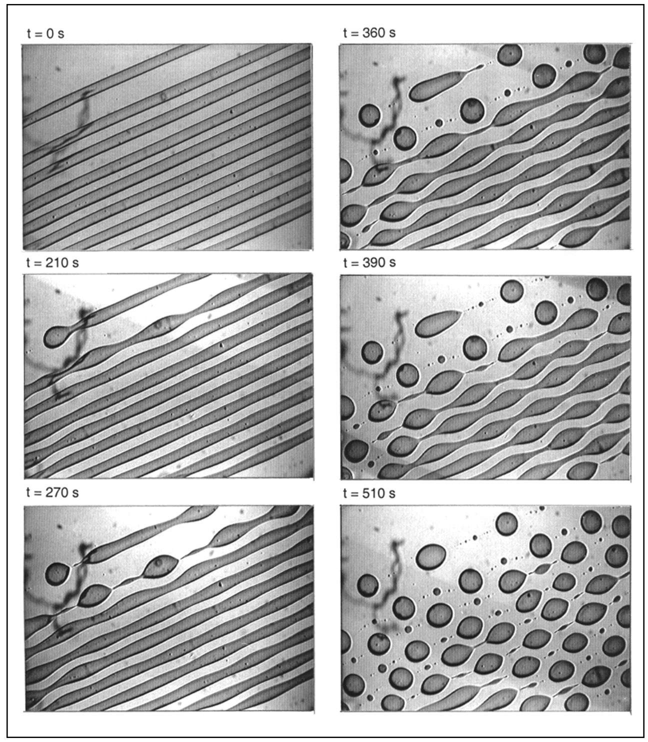

Figure 7. Process of the out-of-phase breakup of PA-6 threads in a matrix of PS N7000.

The photos were taken from a video screen at the following times: $t = 0$, 210, 270, 360, 390 and 510 s. The initial thread diameter was 70 micron. The viscosity ratio $p = 0.04$. The measurement was performed at 230°C (Elemans et al., 1997).

Conclusions

We have proposed the following scenario for the breakup process of two parallel viscous threads surrounded by another viscous liquid. For threads that are very close together, breakup occurs via an "in-phase" mode, while for threads that are somewhat further apart, this happens via an "out-of-phase" mode. If the distance between the threads is very large, there is no phase relation between the breaking threads.

The critical distance b_c is defined as the distance for which the in-phase breakup behavior changes into an out-of-phase breakup behavior. The value of b_c is a monotonically increasing function of the viscosity ratio p. This scenario is supported by the results of recent thread breakup experiments. We are aware of the fact that the real velocity field in and around a breaking thread is more complex than what is considered here. Figure 4, consequently, only hints at a possible explanation why both in and out-of-phase simultaneous breakup behavior occurs. A next step, therefore, would be to consider a stability analysis on the basis of the velocity field of two neighboring threads immersed in an unbounded fluid. Such a study is currently underway.

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Appendix

The boundary conditions for one single thread can be summarized as

$$\Delta \begin{pmatrix} A_1 \\ A_2 \\ B_1 \\ B_2 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ \sigma\epsilon(1-x^2)/2\mu xa \end{pmatrix}$$

where the matrix $\mathbf{\Delta}$ is given by

$$\Delta = \begin{pmatrix} kI_1(x) & xI_0(x) & -kK_1(x) & -xK_0(x) \\ kI_o(x) & 2I_0(x)+xI_1(x) & kK_0(x) & xK_1(x)-2K_0(x) \\ pkI_1(x) & p(xI_0(x)+I_1(x)) & -kK_1(x) & -xK_0(x)+K_1(x) \\ pkI_1(x)' & pxI_1(x) & -kK_1(x)' & xK_1(x) \end{pmatrix}$$

and $I_1(x)' = I_0(x) - I_1(x)/x$ and $K_1(x)' = -K_0(x) - K_1(x)/x$ are the derivatives with respect to x of the Bessel functions, where $x = ka$ is the dimensionless wave number. The coefficients A_1, A_2, B_1 and B_2 follow by Cramer's rule and are equal to

$$A_1(t) = -\frac{\sigma\epsilon(t)[1-x^2]}{2\mu xa}\frac{\det\mathbf{\Delta}_1}{\det\mathbf{\Delta}}$$

$$A_2(t) = \frac{\sigma\epsilon(t)[1-x^2]}{2\mu xa}\frac{\det\mathbf{\Delta}_2}{\det\mathbf{\Delta}}$$

$$B_1(t) = -\frac{\sigma\epsilon(t)[1-x^2]}{2\mu xa}\frac{\det\mathbf{\Delta}_3}{\det\mathbf{\Delta}}$$

$$B_2(t) = \frac{\sigma\epsilon(t)[1-x^2]}{2\mu xa}\frac{\det\mathbf{\Delta}_4}{\det\mathbf{\Delta}}$$

where $\mathbf{\Delta}_i$ is a 3×3 submatrix of $\mathbf{\Delta}$ and can be found by omitting the ith column and the fourth row of $\mathbf{\Delta}$.

The explicit expression for the dimensionless growth rate $\Omega(x,p)$ (see Eq. 5) in terms of these matrices is given by

$$\Omega(x,p) = \frac{1-x^2}{\det\mathbf{\Delta}}\left[a\det\mathbf{\Delta_2}I_0(x) - \det\mathbf{\Delta_1}I_i(x)\right].$$

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