## RAYLEIGH-TAYLOR INSTABILITY OF EXPANDED POLYMER FILMS

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Free layers of liquid - particularly liquid films accelerated by a gas pressure gradient - are unstable. Such instability (of the Rayleigh-Taylor type) was studied theoretically in [1] for plane films of an ideal fluid. The studies [2, 3] examined features of the development of perturbations in the case of plane films of Newtonian and rheologically complex fluids - especially viscoelastic polymers - undergoing acceleration by a gas pressure gradient. The investigations [4, 5] experimentally and theoretically studied the instability of cylindrical water films expanded by gas under high pressure in a cavity. The results obtained here showed that with relatively small gas pressure gradients, destruction of the cylindrical layer of liquid occurs as a result of the development of perturbations (Rayleigh-Taylor type). On the other hand, with an increase in the pressure gradient, destruction of the water films comes to be associated with cavitation occurring after reflection of an unloading wave from an external free surface (expansion of the cylindrical volume of liquid in -[4, 5] was initiated by the electrical explosion of an axial wire or explosive, creating a shock wave). This type of film failure is not typical of liquid polymer films, since these liquids are capable of supporting large tensile stresses without loss of continuity. The dynamics of expanded cylindrical films of polymer liquids in the absence of perturbations was examined in [6].

The goal of the present study is to theoretically investigate the instability of cylindrical films of polymer liquids expanded by gas at high pressure in a cavity. The findings may have application in the common practices of expansion and thermoforming of plastics [7]. If these processes are to be intensified and product quality is to be improved (such as by eliminating variations in wall thickness), then it will be necessary to have an understanding of the mechanism by which perturbations develop in polymer films undergoing expansion by a high-pressure gas in a cavity.

1. Formulation of the Problem of Film Instability. We will examine a cylindrical liquid film. Part of the cross section of this film is shown in Fig. 1a: the dashed lines show the free surfaces in the undisturbed case, the solid lines show the same surfaces in the presence of perturbations; the dot-dash lines represent the middle surfaces in both cases. We will study the plane problem, assuming that the parameters of the disturbed cylindrical film vary only in the azimuthal direction. We will use  $\varphi$  to denote the polar angle,  $R_0(t)$  to denote the radius of the undisturbed middle surface, and  $R(\varphi, t)$  to represent the radius of the middle surface in the presence of perturbations. Rayleigh-Taylor instability of a cylindrical film has the same cause as for a plane film [2, 3]. Flexural perturbations of the middle surface of the film and its thickness develop accordingly. The thinner sections of the film, having less inertia, are bent in the direction of motion. This in turn leads to the appearance of a force caused by a drop in gas pressure and corresponding movement of the liquid from the regions in which the film is contracted to regions where the film is thickneed.

However, the instability of a cylindrical film has several features which differ from the instability of a plane film. The characteristic time of growth of perturbations  $t_* \sim (h_0/a^0)^{1/2}$  [2, 3] (h<sub>o</sub> and a<sup>o</sup> are the thickness and the acceleration of the undisturbed film). In the case of an ideal fluid without surface tension

$$a^0 = \Delta P_0 / \rho h_0. \tag{1.1}$$

Here,  $\rho$  is the density of the liquid;  $\Delta P_0 = P_+^0 - P_-^0$  is the undisturbed gradient of the gas pressure on the film;  $P_+^0$  and  $P_-^0$  are the undisturbed gas pressures inside and outside and cavity.

For a plane film accelerated by a constant pressure gradient,  $t_*$  = const throughout the period of perturbation growth. On the other hand, as a result of the mass conservation

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condition, for an expanded cylindrical film  $h_0 \sim R_0^{-1}$ ,  $\Delta P_0 \sim P_+^0 \sim R_0^{-2\varkappa}$  in the present case of adiabatic expansion of gas in a cylindrical cavity ( $\varkappa$  is the adiabatic exponent). Thus, by virtue of (1.1),  $t_* \sim R_0^{\varkappa - 1}$  and, since  $\varkappa > 1$ , in the expansion of a cylindrical film of an ideal fluid free of surface tension, the rate of growth of perturbations is maximal in the initial stage of motion at small values of  $R_0$ . As the cylindrical film expands, gas pressure in the cavity decreases. This leads to slowing of perturbation growth.

As was shown in [2, 3], the presence of longitudinal elastic stresses associated with preliminary tension of the plane polymer film of liquid helps slow the development of perturbations. In a cylindrical film, the longitudinal elastic stresses (in the azimuthal direction) are accumulated directly during expansion of the film. Thus, this factor — absent for unstretched planar films — also helps slow the growth of disturbances in cylindrical films and, as shown below, may lead to complete suppression of perturbations.

As in [2, 3], we will conduct our theoretical study of Rayleigh-Taylor instability in the thin-film approximation, averaging all of the parameters over the film thickness. We introduce a coordinate system connected with the disturbed middle surface of the film. The longitudinal (azimuthal) direction in the film is indicated by the unit vector of a tangent  $e_1$  to a section of the middle surface of the film, while the transverse direction is indicated by the unit vector of a normal  $e_2$  (Fig. 1a). We use the general equations of the dynamics of thin films [8, 9] to obtain the equations of continuity and momentum in the form

$$\frac{\partial hR}{\partial t} + \frac{\partial}{\partial \varphi} \left[ h \left( V_e - U_e \right) \right] = 0, \qquad (1.2)$$

$$\frac{\partial \rho \mathbf{V} hR}{\partial t} + \frac{\partial}{\partial \varphi} \left[ \rho \mathbf{V} h \left( V_e - U_e \right) \right] = \frac{\partial}{\partial \varphi} \left( N_* \mathbf{e}_1 \right) + \mathbf{P}_* R,$$

where h is the thickness of the film;  $\mathbf{V} = d\mathbf{R}/dt$  is the absolute velocity of a liquid particle;  $\mathbf{U} = \partial \mathbf{R}/\partial t$  is the translational velocity of the coordinate system; N<sub>\*</sub> is the internal force in the film (in the azimuthal direction); P<sub>\*</sub> is the force per unit length of the middle surface, connected with the gradient of gas pressure on the film; the subscript e denotes projection on the tangent.

We will introduce small perturbations of the radius of the middle surface and the thickness of the film  $\delta$  and  $\chi$ 

$$R(\varphi, t) = R_0(t)[1 + \delta(\varphi, t)], \ h(\varphi, t) = h_0(t)[1 + \chi(\varphi, t)]$$
(1.3)

and the relative longitudinal velocity in the chosen coordinate system  $w = V_e - U_e$ , which is also a small perturbation.

We will subdivide the forces  $N_*$  and  $P_*$  into undisturbed and disturbed components,  $N_* = N + n$ ,  $P_* = P + p$  (n and P are small perturbations). It is interesting to examine the case of intensive expansion of the film, this expansion taking place over a period of time which is instantaneous compared to the relaxation time of the liquid. In such an instantaneous process, the points of contact of the macromolecules of the polymer behave in the same manner as the chemical cross-links in rubber macromolecules, while the rheological behavior of the fluid as a whole corresponds to non-Hookian behavior governed by the relation [10]

$$\mathbf{\tau}' = 2G(\mathbf{B} - \alpha \mathbf{B}^{-1}). \tag{1.4}$$

Here,  $\tau'$  is the deviator of the stress tensor; B is the Green tensor; G is the elastic modulus;  $\alpha$  is a dimensionless rheological parameter.

We will calculate N and n using (1.4). We will also calculate the forces P and p, which are connected with the gas pressure gradient in a manner similar to that for a plane

film [3]. We ignore perturbations of the pressure of the surrounding gas, which is permissible if the density of the gas in the cavity is more than one order less than the density of the liquid [2, 3]. Since we are studying concentrated polymer systems, the contribution of surface tension will also be negligible compared to the forces associated with entropic elasticity. As a result, in an approximation which is linear with respect to the perturbations, we use (1.2) to obtain the following equations of the problem of film instability

$$h_{0} = h_{00}r^{-1},$$

$$\frac{dV_{0}}{dt} = \frac{P_{+}^{0} - P_{-}^{0}}{\rho h_{0}} - \frac{2G\left(1 + \alpha\right)\left(r^{2} - r^{-2}\right)}{\rho R_{0}}, \quad r = \frac{R_{0}}{R_{00}},$$

$$\frac{\partial\chi}{\partial t} + \frac{\partial\delta}{\partial t} + \frac{1}{R_{0}}\frac{\partial\omega}{\partial \phi} = 0,$$

$$\frac{\partialw}{\partial t} + w\frac{V_{0}}{R} + \frac{dV_{0}}{dt}\frac{\partial\delta}{\partial \phi} = -\frac{2G\left(1 + \alpha\right)}{\rho R_{0}}\left(r^{2} + 3r^{-2}\right)\frac{\partial\chi}{\partial 0},$$
(1.5)

$$\frac{\partial^2 \delta}{\partial t^2} + \frac{2V_0}{R_0} \frac{\partial \delta}{\partial t} + \frac{2\delta}{R_0} \frac{dV_0}{dt} + \frac{\chi}{R_0} \frac{dV_0}{dt} = \frac{P_+^0 - P_-^0}{\rho h_0 R_0} \delta + \frac{2G\left(1+\alpha\right)\left(r^2 - r^{-2}\right)}{\rho R_0^2} \frac{\partial^2 \delta}{\partial \varphi^2} + \frac{2G\left(1+\alpha\right)\left(r^2 + 3r^{-2}\right)}{\rho R_0^2} \chi_{ABB} + \frac{2G\left(1+\alpha\right)\left(r^2 + 3r^{-2}\right)}{\rho R_0^2} \chi_{ABB} + \frac{2G\left(1+\alpha\right)\left(r^2 - r^{-2}\right)}{\rho R_0^2} \frac{\partial^2 \delta}{\partial \varphi^2} + \frac{2G\left(1+\alpha\right)\left(r^2 + 3r^{-2}\right)}{\rho R_0^2} \chi_{ABB} + \frac{2G\left(1+\alpha\right)\left(r^2 - r^{-2}\right)}{\rho R_0^2} \frac{\partial^2 \delta}{\partial \varphi^2} + \frac{2G\left(1+\alpha\right)\left(r^2 - r^{-2}\right)}{\rho R_0^2} \frac{\partial^2 \delta}{\partial \varphi^2} + \frac{2G\left(1+\alpha\right)\left(r^2 + 3r^{-2}\right)}{\rho R_0^2} \chi_{ABB} + \frac{2G\left(1+\alpha\right)\left(r^2 - r^{-2}\right)}{\rho R_0^2} \frac{\partial^2 \delta}{\partial \varphi^2} + \frac{2G\left(1+\alpha\right)\left(r^2 - r^{-2}\right)}{\rho R_0^2} \frac{\partial^$$

where  $h_{00}$  is the initial undisturbed thickness of the film;  $R_{00}$  is the undisturbed initial value of the radius of the cylindrical cavity of the film;  $V_0(t)$  is the undisturbed rate of dispersion of the film. The first two equations express the continuity condition and the momentum balance for the undisturbed film, while the remaining three pertain to the disturbed motion. They represent the equation of continuity and projections of the momentum equation on a tangent and a normal to the middle surface of the film.

The gas pressure in the cavity of the cylindrical film is determined by the relation

$$P^{0}_{+} = P^{00}_{+} r^{-2\varkappa} \tag{1.6}$$

 $(P^{00}_+$  is the initial gas pressure in the cavity).

As in [6], we introduce the characteristic scale of velocity

$$v_{01} = \sqrt{\frac{P_{+}^{00}R_{00}}{(\varkappa - 1)\,\rho h_{00}}}.$$
(1.7)

Referring  $R_0$  to  $R_{00}$ ,  $V_0$  and w to  $v_{01}$ , and t to  $R_{00}/v_{01}$ , with allowance for (1.6) we convert Eq. (1.5) to dimensionless form

$$\frac{dr}{d\tau} = u, \quad a = \frac{du}{d\tau} = \frac{\varkappa - 1}{r^{2\varkappa - 1}} - 2Tr\left(1 - r^{-4}\right) - \frac{2P_2r}{Q},$$

$$\frac{\partial\chi}{\partial\tau} + \frac{\partial\delta}{\partial\tau} + \frac{\partialw_1}{\partial\varphi} = 0,$$

$$\frac{\partialw_1}{\partial\tau} + 2w_1 \frac{u}{r} + \frac{a}{r} \frac{\partial\delta}{\partial\varphi} = -2T\left(1 + 3r^{-4}\right) \frac{\partial\chi}{\partial\varphi},$$

$$\frac{\partial^2\delta}{\partial\tau^2} + \frac{2u}{r} \frac{\partial\delta}{\partial\tau} + \frac{a\delta}{r} + \frac{a\chi}{r} = 2T\left(1 - r^{-4}\right) \left(\delta + \frac{\partial^2\delta}{\partial\varphi^2}\right) + 2T\left(1 + 3r^{-4}\right)\chi.$$

$$Q = \frac{2h_{00}}{2}; \quad T = \frac{G\left(1 + \alpha\right)}{2}; \quad w_1 = \frac{w/v_{01}}{2}; \quad \tau = \frac{tv_{01}}{2}; \quad u = \frac{V_0}{2}.$$
(1.8)

Here  $P_2 = \frac{P_0^-}{\rho v_{01}^2}; \ Q = \frac{2h_{00}}{R_{00}}; \ T = \frac{G(1+\alpha)}{\rho v_{01}^2}; \ w_1 = \frac{w/v_{01}}{r}; \ \tau = \frac{tv_{01}}{R_{00}}; \ u = \frac{V_0}{v_{01}};$ 

The initial conditions for the undisturbed motion will be

$$\tau = 0, \ r = 1, \ u = 0. \tag{1.9}$$

2. Initial Stage of Film Expansion. As was shown in Part 1, the rate of growth of perturbations of a cylindrical film should be maximal at the beginning of its expansion. Thus, if the film is stable at the initial moments of expansion, then it will remain stable during all subsequent motion. We therefore focus our attention on the "rapid" instability at the initial moment of expansion. Assuming that the perturbations develop so rapidly that the undisturbed motion can be assumed "frozen," we find from (1.8) and (1.9) that

$$\frac{\partial \chi}{\partial \tau} + \frac{\partial \delta}{\partial \tau} + \frac{\partial w_1}{\partial \phi} = 0, \quad \frac{\partial w_1}{\partial \tau} + a_0 \frac{\partial \delta}{\partial \phi} = -8T \frac{\partial \chi}{\partial \phi}, \quad (2.1)$$

$$\frac{\partial^2 \delta}{\partial \tau^2} + a_0 \left(\delta + \chi\right) = 8T\chi, \quad a_0 = \varkappa - 1 - \frac{2P_2}{Q}.$$



Representing the perturbations in the form  $(\chi, w_1, \delta) = (\chi_0, w_{10}, \delta_0) \exp(\gamma \tau + is\phi)$  (the subscript 0 denotes amplitudes,  $\gamma$  denotes an increment,  $s = 1, 2, \ldots$  represents the wave number), we use (2.1) to obtain the dispersion equation

$$\gamma^{4} + 8T(1 + s^{2})\gamma^{2} + a_{0}s^{2}(16T - a_{0}) = 0.$$
(2.2)

The only solution of this equation which can lead to instability is

$$\gamma = \{-4T(1+s^2) + [16T^2(1+s^2)^2 - a_0s^2(16T-a_0)]^{1/2}\}^{1/2}.$$
(2.3)

At  $R_{00} \rightarrow \infty$ , Eqs. (2.2) and (2.3) will naturally change into the corresponding dispersion equations obtained in [2, 3] for a plane film.

Equation (2.3) will give a positive real value of  $\gamma$  under the condition that  $16T/a_0 < 1$ . Thus, with satisfaction of this condition at the beginning of dispersion of the film, the perturbations will increase (the initial stage of expansion of the film will be unstable). On the other hand, with allowance for (2.2) and (2.3), the case

$$\frac{16T}{a_0} \ge 1 \left( \frac{16G(1+\alpha)h_{00}}{\Delta P_{00}R_{00}} \ge 1 \right), \quad \Delta P_{00} = P_+^{00} - P_-^0$$
(2.4)

corresponds to neutral instability of the film (zero or purely imaginary values of  $\gamma$ ). Thus, in a linear approximation, the initial stage of film expansion is stable if condition (2.4) is satisfied. In fact, a small perturbation  $\delta$  of the middle surface of the film corresponds to its tension by the same order of magnitude. Given a sufficiently large value of the elastic modulus (G and, accordingly, T) along the film (in the azimuthal direction), a fairly large elastic force develops. As is known [2, 3], this force is a stabilizing factor.

It should be noted that, for a plane film, such stabilization may be realized just as a result of preliminary tension, since small flexural perturbations do not lead to elongation of its middle surface in a linear approximation. Inequality (2.4), as the stability condition, is further supported by the results of numerical solution of system (1.8). Here, the probability of violation of the stability condition due to nonlinear effects is low due to the fact that such effects are manifest slowly.

With an increase in s, the increment  $\gamma$  monotonically increases in accordance with (2.3). At  $s \rightarrow \infty$ ,  $\gamma$  approaches the asymptotic value

$$\gamma_* = a_0 \left(\frac{1 - 16T/a_0}{8T}\right)^{1/2}.$$
(2.5)

The assumption of "rapid" instability at the beginning of expansion is valid only in the case  $\gamma_{\star}/\sqrt{a_0} \gg 1$  (the characteristic time of expansion of the film at the initial stage  $\sqrt{R_{00}/(dV_0/dt)}$  is much greater than the characteristic time of increase in the perturbations). Accordingly, the final analytical condition of "rapid" instability will be  $16T/a_0 \ll 2/3$ .

<u>3. Results of Numerical Study of Instability.</u> Beginning with a certain value of the initial gas pressure gradient  $\Delta P_{00}$  and with fixed values for the remaining parameters of the liquid film, condition (2.4) is violated, and we can expect the manifestation of growing perturbations. The rate of increase in these perturbations changes during the expansion of the film cavity as a result of a reduction in gas pressure  $P_{+}^{0}$  and the development of azimuthal elastic stresses in the liquid. For small perturbations, these effects are studied by numerically solving system (1.8). Representing the perturbations in the form  $(\chi, w_1, \delta) = [X_0(\tau), -iW_0(\tau), D_0(\tau)] \exp(is\varphi)$ , we find from (1.8) that



Equations (3.1) were integrated numerically by the Kutta-Merson method together with the first two equations of (1.8). We assigned the following values to the parameters:  $\varkappa =$ 1.4, P<sub>2</sub> = 10<sup>-9</sup>, Q = 10<sup>-2</sup>. The values of s (natural numbers) were chosen from the interval [1, 2/Q]; s<sub>max</sub> = 2/Q = R<sub>00</sub>/h<sub>00</sub> corresponds to the shortest wavelength of perturbation, equal to  $2\pi h_{00}$  at  $\tau = 0$ . It makes no sense to perform calculations for perturbation wavelengths shorter than  $2\pi h_{00}$ , due to the use of equations of the longwave approximation (wavelength  $\ell \gg h_{00}$ ).

Without sacrifice of generality, along with the initial conditions for the undisturbed motion (1.9) we write the following compatibility conditions for the perturbations:

$$\begin{split} \tau &= 0, \quad D_0 = 1, \quad X_0 = -\frac{\gamma^2 + as^2}{\gamma^2 + 8Ts^2} \, D_0, \\ W_0 &= -\frac{\gamma}{s} \, (X_0 + D_0), \quad \frac{dD_0}{d\tau} = \gamma D_0, \end{split}$$

where  $\gamma$  is determined from (2.3) with a prescribed wave number s.

Figure 1b shows the dependence of the amplitudes of flexural perturbations of the middle surface  $D_0$  on time at T = 0.002; curve 1 corresponds to s = 5; 2) 10; 3) 20; 4) 40; 5) 80. In the given case,  $a_0 \simeq \varkappa - 1$ . Thus, in accordance with the result obtained in the previous section, we can expect an increase in perturbations at  $T = 0.002 < a_0/16 \simeq (\varkappa - 1)/16 = 0.025$ . In fact, at the initial moments of time in Fig. 1b, the amplitude of the perturbations increases exponentially, and the rate of increase is greater, the greater the wave number s. However, radial expansion of the film is accompanied by an increase in the azimuthal stress in the film. This slows the increase in perturbations, and the amplitude of the latter begin to decrease. The oscillations in the amplitude of the perturbations in Fig. 1b are connected with the competition between elastic and inertial forces typical of polymer liquids [11]. The "damping" of these oscillations is connected not with dissipation (which is absent) but with an increase in the azimuthal stress over time. The increment of the perturbations in the perturbations in the analytic of the perturbation (which is absent) but with initial stage coincides with the quantity calculated by means of (2.3).

For each wave number, the perturbations increase until attainment of a certain value  $D_{0m}$  at  $\tau = \tau_m$ . Figure 2 shows the dependence of  $D_{0m}$  and  $\tau_m$  on s (curves 1-4 - T = 0.004; 0.006; 0.008; 0.01). The data for  $D_{0m}$  is shown by solid lines, while the data for  $\tau_m$  is shown by dashed lines. It should be noted that the curves in Fig. 2 were drawn through a discrete set of theoretical points corresponding to integral values of s. It is evident that with a certain value s =  $s_*$ , the maximum perturbation amplitude takes the greatest value  $D_*$  with fixed T. At s <  $s_*$ ,  $D_{0m}$  increases with an increase in s because - in accordance with (2.3) - the increment  $\gamma$  increases. On the other hand, at s >  $s_*$ , an increase in s corresponds to an increase in the stabilizing effect of the azimuthal elastic stress. Here, both the time required to reach the maximum and the value of  $D_{0m}$  decrease. The dependence of  $D_*$  on T is shown by curves in different scales in Fig. 3. With an increase in T from 0 to 0.025, the maximum perturbation amplitude rapidly decreases to  $D_* = D_0(0) = 1$ . At T >  $a_0/16 = 0.025$ , perturbations do not increase on a cylindrical film. This confirms that there is a region of stability (Part 2) and that inequality (2.4) is the stability condition.

The results of the calculations show that  $s_*$  increases with a decrease in T. At  $T \rightarrow 0$ , the theoretical points lie on a smooth curve  $s_*(T)$ ; on the other hand, as T approaches the value  $a_0/16 = 0.025$ , the relation  $s_*(T)$  becomes a step function due to the discreteness of the values of the wave numbers.

Figure 4 shows graphs of the dependence, on T, of the time  $\tau_*$  and the degree of tension  $r_*$  corresponding to the moment of attainment of the greatest amplitude  $D_*$ . As  $T \rightarrow 0.025$ ,  $r_* \rightarrow 1.37$ , and  $\tau_* \rightarrow 1.46$ . The circles show theoretical points. Meanwhile, the relations  $\tau_*(T)$  and  $r_*(T)$  are step functions due to the discreteness of s; smoothing curves 1 and 2 were drawn for the sake of clarity.

Analysis of the results permits the conclusion that as  $T \rightarrow 0$ , the characteristic scale of the perturbations  $\ell_{*} = 2\pi R_{0}(\tau_{*})/s_{*}$  decreases from the greatest amplitude.

The results obtained show that at each moment of time we can distinguish a perturbation with a wave number  $s_{\tau}$  (different at each moment) and an amplitude having the maximum value  $D_{\tau}$ . The wavelength of this perturbation  $\ell_{\tau} = 2\pi R_0/s_{\tau}$  is the characteristic scale of the pattern of perturbations on the film at the given moment of time. The solid lines in Fig. 5 pass through theoretical values of  $s_{\tau}$  at different moments of time, while the dashed lines show the dependence of  $s_{\tau}$  on r. Curves 1-4 correspond to  $T = 0.25 \cdot 10^{-3}$ ;  $0.5 \cdot 10^{-3}$ ;  $10^{-3}$ ;  $2 \cdot 10^{-3}$ . It is evident that  $\ell_{\tau}$  increases with an increase in time. Figure 6 shows the relation  $D_{\tau}(\tau)$  (with the same values of T as in Fig. 5). It is apparent that the maximum amplitude of the flexural perturbations at the beginning of motion increases almost exponentially. The amplitude then begins to decrease relatively slowly.

As a result of the increase in perturbations, failure of the film takes place at the moment of time  $\tau_1$ , when  $\chi(\tau_1) = -1$ . The characteristic size of the segments over which the film breaks down is  $\ell_1 = 2\pi R_0(\tau_1)/s_{\tau}(\tau_1)$ .

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