Macroscopic mechanism of rupture of free liquid films

Yulii D. Shikhmurzaev

School of Mathematics, University of Birmingham, Birmingham B15 2TT, UK

Abstract

A model describing a macroscopic mechanism of the rupture of a free liquid film is introduced and analysed in the framework of the thin-film approximation. The process is shown to be driven by the surface-tension gradient arising when the rate of variation of the free-surface area due to external disturbances becomes comparable with the inverse surface-tension-relaxation time. The proposed mathematical description of the rupture phenomenon does not require the introduction of intermolecular forces into the equations of macroscopic fluid mechanics. ***To cite this article: Y.D. Shikhmurzaev, C. R. Mecanique 333 (2005).***
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Résumé

Mécanisme macroscopique de rupture de couches minces libres. Dans le cadre de l'approximation de couches minces, on introduit et on analyse un modèle macroscopique de rupture d'une couche mince liquide. On démontre que le processus est alimenté par le gradient de la tension surfacique chaque fois quand le taux de variation de l'aire de surface libre provoqué par les perturbations extérieures devient comparable avec l'inverse du temps de relaxation de la tension surfacique. La description mathématique du phénomène de rupture proposée ici ne nécessite pas l'introduction explicite des forces intermoléculaires dans les équations de la mécanique des fluides macroscopique. ***Pour citer cet article : Y.D. Shikhmurzaev, C. R. Mecanique 333 (2005).***
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E-mail address: yulii@for.mat.bham.ac.uk (Y.D. Shikhmurzaev).

1. Introduction

The rupture of a free liquid film is an example from a wide class of fluid motion where the flow domain's topology changes in a finite time. The peculiarity of this phenomenon is that, unlike the breakup of a cylindrical jet, in the planar geometry of a film the capillary pressure is always a stabilizing factor tending to counter the effects of external disturbances and restore the initial shape of the film. In the literature, the rupture of films has been studied in the situation where the film is already so thin that the opposite interfaces feel each other's presence via long-range intermolecular forces, which then act as a breaking mechanism [1,2]. In the present Note, we examine what happens before that and whether or not the process of thinning a free film with a thickness on a macroscopic rather than molecular scale and its eventual rupture can be modelled in the framework of fluid mechanics without invoking intermolecular forces.

The key physical idea of the present study is as follows. The deformation of a free surface associated with variation of its area is a mechanism that drives the liquid-gas interface out of its equilibrium state. This influence is opposed by the relaxation mechanism rooted in thermodynamics that tends to restore the equilibrium state of the interface and bring its surface properties, such as the surface tension, back to their equilibrium values. In almost all flows, the characteristic time scale associated with the rate-of-change of the free-surface area is by many orders of magnitude larger than the surface-tension-relaxation time so that, if there are no other macroscopic factors influencing the surface tension (such as the temperature or surfactant concentration gradients, external electromagnetic fields, etc.), one can consider the surface tension of the liquid-gas interface to be a known constant.

The situation changes in the case of the rupture of a free film. In the process of thinning of a free film, to leading order in the aspect ratio, the velocity profile across the film is that of a plug flow so that for the relative rate at which the fresh free surface area S is created one has $(1/S)\,dS/dt \propto (1/h)\,dh/dt$ and it tends to infinity (and hence the corresponding time scale tends to zero) as the film's thickness h decreases. Thus, for sufficiently thin films external disturbances can cause significant deviation of the interfacial properties, first of all the surface tension, from their equilibrium values. Then, for a general case of a spatially nonuniform disturbance one will have a surface tension gradient which generates the Marangoni flow, and if the film is sufficiently thin, it can be ruptured due to the resulting motion before the thermodynamic equilibrium of the interface is restored. We examine this scenario using a simple model of fluid flow with forming/disappearing interfaces [3], which was developed for, and shown to be successful in, describing dynamic wetting [3–5]. In the present work, the model is used without any adhoc alterations.

2. Thin-film approximation

To study a free-surface flow with a topological transition of the flow domain in the framework of the theory of fluid motion with forming interfaces [3] one has to consider solutions of the Navier–Stokes equations,

$$\nabla \cdot \mathbf{u} = 0, \qquad \rho(\partial \mathbf{u}/\partial t + \mathbf{u} \cdot \nabla \mathbf{u}) = -\nabla p + \mu \nabla^2 \mathbf{u} \tag{1}$$

satisfying at an a priori unknown free surface $f(\mathbf{r}, t) = 0$ with the inward normal $\mathbf{n} = \nabla f/|\nabla f|$ the boundary conditions

$$\partial f/\partial t + \mathbf{v}^s \cdot \nabla f = 0 \tag{2}$$

$$-p + \mu \mathbf{n} \cdot [\nabla \mathbf{u} + (\nabla \mathbf{u})^*] \cdot \mathbf{n} = \sigma \nabla \cdot \mathbf{n}, \qquad \mu \mathbf{n} \cdot [\nabla \mathbf{u} + (\nabla \mathbf{u})^*] \cdot (\mathbf{I} - \mathbf{nn}) + \nabla \sigma = 0 \tag{3}$$

$$\rho(\mathbf{u} - \mathbf{v}^s) \cdot \mathbf{n} = (\rho^s - \rho^s_e)\tau^{-1}, \qquad \partial \rho^s/\partial t + \nabla \cdot (\rho^s \mathbf{v}^s) = -(\rho^s - \rho^s_e)\tau^{-1} \tag{4}$$

$$(1 + 4\alpha\beta)\nabla \sigma = 4\beta(\mathbf{v}^s - \mathbf{u}) \cdot (\mathbf{I} - \mathbf{nn}), \qquad \sigma = a\rho^s - b(\rho^s)^2 \tag{5}$$

together with some conditions in the far field specifying a particular flow and initial conditions. In addition to the known kinematic condition (2) and conditions on the normal and tangential stress (3), where $\mathbf{I}$ is the metric tensor,

the model accounts for the mass exchange between the bulk and the surface phase (4) as the interface relaxes towards its equilibrium state with the surface density ρ^s tending to its equilibrium value ρ_e^s (τ is the relaxation time; $\mathbf{u}$ and $\mathbf{v}^s$ are the bulk and the surface-phase velocity, respectively). For spatially nonuniform flows, like the rupture of a film, the appearing gradient of the surface tension σ, first, influences the flow via the tangential-stress condition (the Marangoni effect; the second condition in (3)) and, secondly, forming a torque with the tangential stress, it makes the tangential to the interface components of the surface velocity $\mathbf{v}^s$ deviate from the corresponding component of the bulk velocity $\mathbf{u}$ evaluated at the interface (the first condition in (5)). The equation of state in the surface phase (the second equation in (5)) is taken in a simple barotropic form approximating the general equation of state for the surface phase, $\sigma = f(\rho^s, T)$, where T is the absolute temperature, for the process of interface formation. The concept of the 'surface density' as a parameter of state was first introduced by Gibbs [6] and since then used in different forms, together with the notion of surface velocity, by many authors [7]. Eq. (5) takes into account that the surface tension decreases from its equilibrium value $\sigma_e = \sigma(\rho_e^s)$ if the surface phase becomes compressed or extremely rarefied. In particular, a hypothetical instantly created free surface corresponds to $\rho^s = 0$ and, as one would expect, has zero surface tension; the latter is acquired as the molecular motion leads to the formation of a certain structure of the interfacial layer, which is macroscopically modelled as a two-dimensional 'surface phase'. Estimates for material constants α, β and τ for some fluids have been obtained by analysing experiments on dynamic wetting [5].

The general three-dimensional problem can be considerably simplified in the thin-film approximation with the ratio of characteristic length scales in the directions normal and tangential to the film ϵ as a small parameter. We will consider this approximation for a plane two-dimensional flow in a film of incompressible Newtonian fluid of viscosity μ and density ρ surrounded by an inviscid dynamically-passive gas. Let the film's shape be described by $y = \pm h(x, t)$ in a suitably chosen Cartesian coordinate frame and u, u^s, v, v^s denote, respectively, the x and y components of the bulk and surface velocities. Then the thin-film approximation can be obtained by using the following asymptotic expansions:

$$x = L\epsilon\bar{x}, \quad y = L\epsilon^2\bar{y}, \quad t = t_\mu\epsilon^2\bar{t}, \quad hL^{-1} = \sum_{n=0}^{\infty}\epsilon^{2(n+1)}h_n \tag{6}$$

$$(u, u^s)U^{-1} = \sum_{n=0}^{\infty}\epsilon^{2n-1}(u_n, u_n^s), \qquad (v, v^s)U^{-1} = \sum_{n=0}^{\infty}\epsilon^{2n}(v_n, v_n^s)$$

$$pP^{-1} = \sum_{n=0}^{\infty}\epsilon^{2(n-1)}p_n, \quad (\sigma\sigma_e^{-1}, \rho^s ba^{-1}) = \sum_{n=0}^{\infty}\epsilon^{2n}(\sigma_n, \rho_n^s), \quad \text{as } \epsilon \to 0$$

where $L = \mu^2(\rho\sigma_e)^{-1}$, $t_\mu = \mu^3\rho^{-1}\sigma_e^{-2}$, $U = \sigma_e\mu^{-1}$, $P = \rho\sigma_e^2\mu^{-2}$. After the standard asymptotic analysis to leading order one arrives at the following set of equations:

$$\frac{\partial u_0}{\partial\bar{t}} + u_0\frac{\partial u_0}{\partial\bar{x}} = \frac{1}{h_0}\frac{\partial\sigma_0}{\partial\bar{x}} + \frac{4}{h_0}\frac{\partial}{\partial\bar{x}}\left(h_0\frac{\partial u_0}{\partial\bar{x}}\right), \qquad v_0 = -\bar{y}\frac{\partial u_0(\bar{x}, \bar{t})}{\partial\bar{x}} \tag{7}$$

$$\frac{\partial h_0}{\partial\bar{t}} + u_0^s\frac{\partial h_0}{\partial\bar{x}} = v_0^s, \qquad \frac{\partial\rho_0^s}{\partial\bar{t}} + \frac{\partial(\rho_0^s u_0^s)}{\partial\bar{x}} = 0 \tag{8}$$

$$\frac{\partial\sigma_0}{\partial\bar{x}} = 4\bar{\beta}(u_0^s - u_0), \qquad (u_0 - u_0^s)\frac{\partial h_0}{\partial\bar{x}} - (v_0 - v_0^s) = \bar{\chi}(\rho_0^s - \bar{\rho}_e^s) \tag{9}$$

$$\sigma_0 = a\rho_0^s - b(\rho_0^s)^2 \tag{10}$$

where $\bar{\beta} = \beta\mu\rho^{-1}\sigma^{-1}(1 + 4\alpha\beta)^{-1}$, $\bar{\chi} = a\mu(b\rho\sigma_e\tau)^{-1}$ and $\bar{\rho}_e^s = \rho_e^s ba^{-1}$. In obtaining (7)–(10) it was assumed that all nondimensional parameters appearing after the use of scaling (6) are of O(1) as $\epsilon \to 0$. In particular, this means that $\epsilon^2 t_\mu\tau^{-1} \to 0$ as $\epsilon \to 0$ so that, physically, the process described by (7)–(10) takes place on a time

scale small compared with τ and, according to the second equation (8), the relaxation mechanisms have no time to restore the equilibrium surface density (and hence the equilibrium surface tension).

Eqs. (7)–(10) can be simplified further for medium to high-viscosity fluids by using the estimates for material constants of the model obtained from experiments on dynamic wetting [5]. These estimates show that for such fluids $\bar{\chi} \ll 1$ and $\bar{\beta} \gg 1$ and hence, to leading order in these parameters, it follows from (9) that one can neglect the difference between (u_0^s, v_0^s) and (u_0, v_0). Then, after simple algebra we arrive at an initial-value problem for the following set of nonlinear equations:

$$\frac{\partial u}{\partial t} + u \frac{\partial u}{\partial x} = \frac{1}{h} \frac{\partial \sigma}{\partial x} + \frac{4}{h} \frac{\partial}{\partial x} \left(h \frac{\partial u}{\partial x} \right) \tag{11}$$

$$\frac{\partial F}{\partial t} + \frac{\partial (Fu)}{\partial x} = 0, \quad F = (h, \rho^s); \qquad \sigma = a\rho^s - b(\rho^s)^2 \tag{12}$$

(Hereafter for brevity we drop the overbar and the subscript 0.) In (11), the surface tension gradient on the right-hand side appears as the only driving force, unlike the case of the breakup of a cylindrical jet [8], where the Marangoni effect and the capillary pressure due to the cross-sectional curvature play comparable roles.

3. The process of rupture

As already mentioned, the scaling (6) implies, in particular, that Eqs. (11), (12) operate on a time scale small compared with the surface-tension-relaxation time, and, with no relaxation in (12), $u \equiv \hat{u} = \text{const}$, $h \equiv \hat{h} = \text{const}$, $\rho^s \equiv \hat{\rho}^s = \text{const}$ is a solution of (11), (12) for arbitrary values of $\hat{u}$, $\hat{h}$ and $\hat{\rho}^s$. A linear stability analysis of this solution leads to a dispersion relationship of the form

$$\omega = k\hat{u} - 2ik^2 \left[1 \pm \left(1 + \frac{\hat{\rho}^s \lambda}{4k^2 \hat{h}} \right)^{1/2} \right], \quad \text{where } \lambda = \frac{d\sigma}{d\rho^s}(\hat{\rho}^s) \tag{13}$$

and ω and k are the angular frequency and wavenumber, respectively. Hence the solution is stable for $\lambda < 0$, which for the surface equation of state given by the second equation (12) corresponds to $\hat{\rho}^s > a/(2b)$ and unstable otherwise. In other words, it is stable if the rarefaction of the surface phase increases the surface tension, which then contracts the surface and replenishes ρ^s. On the other hand, if the surface tension decreases with decrease in ρ^s, then a local rarefaction of the surface phase due to an external disturbance leads to a local reduction in the surface tension whose gradient then acts to pull the film apart and reduce ρ^s even further. The dispersion relationship (13) also indicates that, unlike Rayleigh's instability of a cylindrical jet, the most destabilizing are short-wave disturbances.

In order to illustrate how the Marangoni effect incorporated in (11) and (12) leads to the rupture of a free film in the nonlinear regime consider the evolution of the film that after an external finite-amplitude disturbance makes $\rho^s \equiv \hat{\rho}^s$, where $\hat{\rho}^s$ is in the unstable zone ($\lambda \geqslant 0$). It is instructive to look at the borderline case where $\lambda = 0$, i.e. $\hat{\rho}^s = a/(2b)$. Thus, let us consider a small disturbance $\rho^s = a/(2b)[1 - A\exp(-x^2/l^2)]$, where A is the relative amplitude and l is the width of the disturbance.

Fig. 1 shows the film's profile at various times obtained via numerical integration of (11) and (12) for $A = 0.1$ and $l = 0.5$. As one can see, the initial disturbance of the surface tension indeed leads to the film's thinning due to the Marangoni effect. In the thin-film approximation $p = -2\partial u/\partial x$ and, as t increases, the pressure in the minimal cross-section goes through a global minimum (Fig. 2) suggesting a transition to a new dynamic regime. The essence of this regime follows from Fig. 1, which shows a gradual formation of a distinct structure consisting of a departing main body of fluid (macrofilm) and a vanishing residual film of an increasingly uniform thickness. Given that the (almost constant) thickness of the macrofilm and the (infinitesimal) thickness of the residual film are separated in
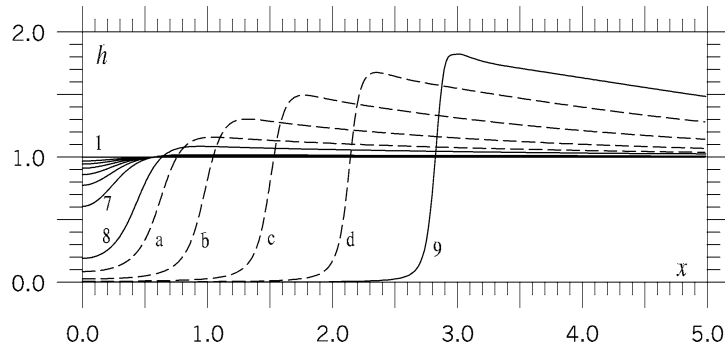
Fig. 1. Profiles of the free surface at different moments in time. Solid lines 1–9 are obtained for $t = 0, 10, 15, 20, 25, 30, 35, 40$ and 45, respectively. Dashed lines a–d correspond to $t = 41, 42, 43$ and 44.

Fig. 1. Profiles de la surface libre aux moments différents. Les lignes continues 1–9 sont obtenus pour $t = 0, 10, 15, 20, 25, 30, 35, 40$, respectivement. Les lignes en pointillé a–d correspondent aux temps $t = 41, 42, 43$ et 44.
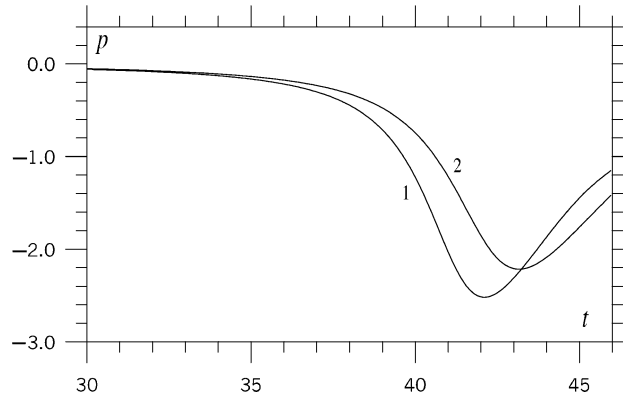

Fig. 2. The pressure in the minimal cross-section vs time for $l = 0.5$ (curve 1) and $l = 0.7$ (curve 2).

Fig. 2. La pression dans la section transverse minimale en fonction du temps pour $l = 0,5$ (courbe 1), et pour $l = 0,7$ (courbe 2).

scale, the late stage of the evolution of the residual film must take place in a self-similar regime. This regime is given by

$$u = x(t - t_0)^{-1}, \qquad (h, \rho^s, \sigma) = (H, R, aR)(t - t_0)^{-1}$$

where t_0, H and R are constants determined by the preceding evolution of the film. In this solution, both terms on the right-hand side of (11) become separately zero thus indicating that the process is driven by the departing ends of the macroscopic film whereas the residual film simply follows and gets thinner accordingly. Thus, there is no effect of the residual film on the departing ends of the macrofilm and hence of these ends on each other. In effect, this is what 'rupture' means in dynamic terms.

Since the thinning of the residual film does not lead to singularities in the solution, in practice, as one computes the global flow numerically, the residual film can be neglected once its thickness becomes smaller than the spatial resolution of the code.

The main feature of the mechanism of rupture considered above is that it does not require the free surfaces being brought together by the (singular) forces of attraction of the molecular origin, whose inclusion in macroscopic fluid mechanics is methodologically questionable. It is also noteworthy that, physically, the microscopic evolution of the film that follows after the opposite interfacial layers 'touch' is a natural continuation of the mechanism

described above. Indeed, 'touching' of the opposite interfaces means that each of them is no longer separating two bulk phases, and it is this position between the bulk phases (associated with asymmetry of intermolecular forces acting on the interfacial layer) that gives rise to the surface tension. Hence, 'touching' interfaces lose their surface tensions at the point of contact and, given that in the remaining parts of the film the surface tension is still nonzero, the film is pulled apart by the surface-tension gradient. This final 'microscopic rupture' is physically the same as the macroscopic process described in the present paper.

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