Weakly nonlinear stability of ultra-thin slipping films*

HU Guohui**

(Shanghai Institute of Applied Mathematics and Mechanics, Shanghai University, Shanghai 200072, China)

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Abstract A weakly nonlinear theory is presented to study the effects of slippage on the stability of the ultra-thin polymer films. The nonlinear mathematical model is constructed for perturbations of small finite amplitude based on hydrodynamic equations with the long wave approximation. Results reveal that the nonlinearity always accelerates the rupture of the films. The influences of the slip length, film thickness and initial amplitude of perturbations on the rupture of the films are investigated.

Keywords: ultra-thin film, weakly nonlinear theory, hydrodynamic stability, rupture of thin film.

Liquid thin film flows are commonly encountered in nature and in numerous practical applications, e.g. in chemical engineering, materials process, or microelectronic systems. A great deal of theoretical studies have been performed to understand the stability, dynamics and dewetting of the flows. For ultra-thin films, whose mean thicknesses are less than 100 nm, the flows can be described by Navier-Stokes equations including the intermolecular body force or disjoining pressure terms[1]. Long wave or lubrication approximation is usually applied to obtain a set of simplified system since the film thickness is quite smaller than the streamwise length scale. An outline of the theoretical work based on long wave approximation was proposed by Oron, Davis and Bankoff[2]. Previous researches showed that the free films are inherently unstable due to the intermolecular potential while the films on a no-slip substrate might be stable when the effective Hamaker constant in molecular interaction is negative[3—6].

Most of the current theoretical studies focused on the films satisfying no-slip velocity condition on substrates. However, recent experiments and computations based on molecular dynamics simulation indicated that slipping velocity can be evidently found in macromolecular polymer films[7—9]. Various ranges of slip length were reported in different experiments, varying from a few nanometers to a thousand micrometers. Therefore, it might be expected that the slippage has great influences on their hydrodynamic behavior. A linear stability analysis was conducted by Sharma et al. for Newtonian fluid taking into account the effect of slippage[10]. Results showed that slippage can encourage the development of the perturbations and the rupture or the breakup, in the sense that the local film thickness becomes zero in a finite time, and can reduce the number density of holes for slip strong enough. To understand the nonlinear evolution and morphology of the flow, numerical simulations were performed henceforth based on Navier-Stokes equations with long wave approximation for both weak and strong slips[11, 12]. Results showed that the ratio of rupture time between the nonlinear computations and linear analyses is always less than 1, which reveals that the nonlinear effects can accelerate the breakup of the films.

Compared with the linear theory and numerical simulation, weakly nonlinear theory (WNT) has advantages that it can not only obtain the analytical solution which is convenient to analyze, but also capture the essential physics of the problem, although it is not fully quantitatively accurate. The stabilities of no-slip on substrate and free thin films subject to finite amplitude disturbances have been examined by the weakly nonlinear theory[3, 4, 6, 13]. For free films, Erneux et al. gave an analytical estimate of the rupture time[3, 4]. Sharma et al. discussed the roles of different kinds of forces, such as viscous effect and surface tension, on the stability of the flows, and found that intermolecular force could promote the development of dominant perturbations in the nonlinear stage[4].

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** To whom correspondence should be addressed. E-mail: ghhu@staff.shu.edu.cn

This paper aims at applying the weakly nonlinear theory to the ultra-thin films with slippage. The hydrodynamic equations are described and the nonlinear mathematical model and related computational results are presented.

1 Hydrodynamic equations

A schematic diagram of the thin film with thickness h_0 is described in Fig. 1. The two-dimensional Navier-Stokes equations in (x, z) coordinates can be given as

$$\nabla \cdot v = 0,$$
$$\rho(v_t + v\nabla v) = \mu\nabla^2 v - \nabla p - \nabla\Phi, \quad (1)$$

where the subscript stands for partial differentiation, $v(u, w)$ is the velocity vector, p is the pressure, and Φ is the intermolecular body potential. μ, ρ, γ are dynamic viscosity, density, and surface tension, respectively. The van der Waals potential Φ is the most usually adopted long range force in the previous studies. In the non-retarded form it can be given as

$$\Phi = \frac{A}{6\pi h^3}, \quad (2)$$

where A denotes the Hamaker constant. To consider the retardation effect, the potential will be generalized as

$$\Phi = \frac{A}{6\pi h^3}\frac{1 + c_1 h/R}{(1 + c_2 h/R)^2}. \quad (3)$$

Here, R is a retardation parameter, the numerical constants c_1, c_2 equal 7.98 and 5.32, respectively, other physical parameters in the present study are $A = 10^{-20}$ J, $\gamma = 20$ mJ/m^2, $\mu = 1$ kg/(ms), $\rho = 1000$ kg/m^3, and $R = 100$ nm.

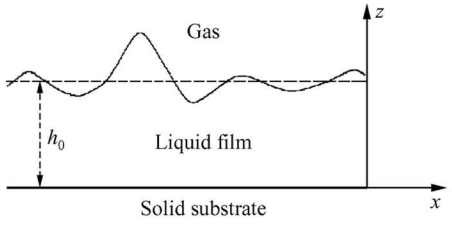

Fig. 1. Schematic diagram of thin film flow.

The boundary conditions on the substrate $z = 0$ are expressed as

$$w = 0, \quad u_z - b^{-1}u = 0, \quad (4)$$

where the slip length b is defined the same as that in the publications available, implying the shear stress balance at the solid-liquid interface. The case $b = 0$ corresponds to no-slip condition, i.e. $u = 0$.

The dynamic and kinematic conditions are written respectively on the free surface $z = h(x, t)$ as

$$(u_z + w_x)(1 - h_x^2) + 2h_x(w_z - u_x) = 0, \quad (5)$$
$$2\mu[(1 - h_x^2)w_z - h_x(u_z + w_x)](1 + h_x^2)^{-1} - p - \gamma h_{xx}(1 + h_x^2)^{-3/2} = 0, \quad (6)$$
$$h_t + uh_x = w. \quad (7)$$

The length and time are scaled with the thickness of film h_0 and kinematic viscosity coefficient υ. Then the dimensionless parameters are obtained as follows:

$$x^* = \frac{x}{h_0}, \quad z^* = \frac{z}{h_0}, \quad t^* = \frac{t\upsilon}{h_0^2}, \quad h^* = \frac{h}{h_0},$$
$$B = \frac{b}{h_0}, \quad u^* = \frac{uh_0}{\upsilon}, \quad w^* = \frac{wh_0}{\upsilon},$$
$$p^* = \frac{(p - p_g)h_0^2}{\rho\upsilon^2}, \quad A^* = \frac{A}{h_0\rho\upsilon^2}, \quad \Gamma = \frac{\gamma h_0}{\rho\upsilon^2}.$$

Applying the long wave approximation to Eq. (1) and boundary conditions Eqs. (4)—(7), the equations are transformed into a set of nonlinear differential equations for the horizontal component of velocity and film thickness, which reads

$$u_t + uu_x - \Gamma h_{xxx} + \Phi_x + B^{-1}uh^{-1} = 4u_{xx} + 4u_x h_x h^{-1}, \quad (8)$$
$$h_t + (uh)_x = 0. \quad (9)$$

Details of the mathematical operations can be found in Ref. [12]. It should be pointed out that the variables in Eqs. (8) and (9) have been rewritten in the long-scale form, and the superscripts, which denote the dimensionless parameters, are omitted for the sake of simplification.

2 Mathematical formulation of the weakly nonlinear theory

To investigate the nonlinear development of small amplitude perturbations, the film thickness is expressed as

$$h(t) = 1 + h' = 1 + \varepsilon y_1(t)\cos kx + \varepsilon^2 y_2(t)\cos 2kx + O(\varepsilon^3), \quad (10)$$

where y_1 and y_2 are the amplitudes of the first and second Fourier modes, k is the wave number of the perturbations. Substituting Eq. (10) into Eq. (9) yields the expression of u up to the order of $O(\varepsilon^3)$,

$$u = -\frac{1}{k}\left[\varepsilon y_1'\sin kx + \varepsilon^2\left(\frac{1}{2}y_2' - y_1'y_1\right)\sin 2kx\right] + O(\varepsilon^3). \quad (11)$$

The primes in Eqs. (10) and (11) mean the deriva-

tives respect to time. Substituting Eqs. (10) and (11) into Eq. (8), a series of equations of different orders of ε can be obtained by mathematical manipulations. The equations of the first two orders are

$$O(\varepsilon):\ y_1'' + (B^{-1} + 4k^2) y_1' + (k^4 + \Phi_{\varepsilon 0}) y_1 = 0, \tag{12}$$

$$O(\varepsilon^2):\ y_2'' + (B^{-1} + 16k^2) y_2' + (16k^4 + \Phi_{\varepsilon 1}) y_2 = 2y_1'' y_1 + 4y_1'^2 + (24k^2 + 4B^{-1}) y_1' y_1 - \Phi_{\varepsilon 2} k^2 y_1^2, \tag{13}$$

where the van der Waals body force

$$\Phi_x = -\frac{A}{6\pi} k [\varepsilon \Phi_{\varepsilon 0} y_1 \sin kx + \varepsilon^2 (\Phi_{\varepsilon 1} y_2 + \Phi_{\varepsilon 2} y_1^2) \sin 2kx].$$

The resulting equation (12) corresponds to the linearized equations (8) and (9). The dispersion relation can be obtained by adopting the normal mode with the time growth rate ω, $y_1 = y_0 \exp(\omega t)$, then we get

$$\omega^2 + (B^{-1} + 4k^2)\omega + (k^4 + \Phi_{\varepsilon 0}) = 0. \tag{14}$$

Thus the two roots ω_1 and ω_2 of the equation can be calculated, and we suppose $\omega_1 > \omega_2$. The results obtained from the linear stability theory, which have been presented in Refs. [10] and [12], showed that the slip velocity will increase the growth rate of the perturbations and the wavelength of the most unstable mode, and accordingly decrease the number density of the holes.

To solve Eqs. (12) and (13), we should introduce initial conditions. Without loss of generality, the initial conditions for Eq. (12) are given such that the solution can be obtained as $y_1 = \exp(\omega_1 t)$. For Eq. (13) we simply set $y_2(0) = 0$, and $y_2'(0) = 0$. Therefore, these problems can be solved sequentially. Note that only the dominant terms will be reserved in the solutions since we only concentrate on the long time behavior.

3 Results

It can be easily found from Eq. (10) that at the most dangerous x-position for a given wave number k, the evolution of perturbed thickness can be always written as

$$h'(t) = h_l + h_{nl} = \varepsilon y_1(t) + \varepsilon^2 y_2(t). \tag{15}$$

Thus the rupture or blowup time T_b can be defined as the time when the thickness of the films vanishes. In addition, the rupture time T_l based on the linear stability theory is estimated to be

$$T_l = \ln(1/\varepsilon)/\omega(k_{\max}), \tag{16}$$

where $k_{\max}$ denotes the wave number of the most unstable mode.

The variation of thickness with time for the most unstable perturbation is shown in Fig. 2 for $h_0 = 50$ nm, $b = 10\mu$m, and $\varepsilon = 0.001$, together with the linear and second order terms. It can be seen that the nonlinear effect promotes the breakup of the film with slippage, which confirms the previous experimental and theoretical studies. The growth rate of the second order term is much larger than that of the first order one. It should be noted that for convenience of comparison to the previous work, the physical variables in the plots of this work are in dimensional form.

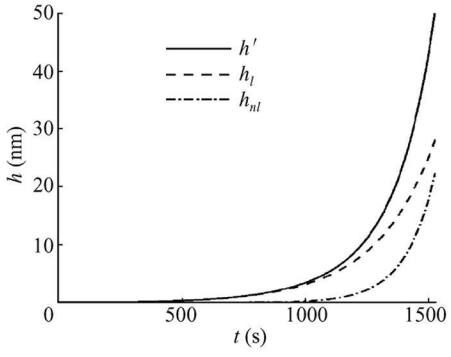

Fig. 2. Variation of the film thickness for the most unstable linear mode ($h_0 = 50$ nm, $b = 10\mu$m, and $\varepsilon = 0.001$).

The rupture time of the weakly nonlinear and linear theories varying with wave number k is given in Fig. 3 for different slipping lengths when $h_0 = 10$ nm. The result demonstrates that the slipping velocity enhances the growth of both linear and nonlinear disturbances, and thus reduces the breakup time of the liquid films. Although the rupture time based on the linear theory is always longer than the blowup time T_b, the nonlinearity does not change the wave number of the most unstable perturbation. Another feature of the plot is that the decreases of the breakup time are more apparent for perturbations with lower wave number, whereas the two curves of T_b and T_l are almost overlapped for higher wave number modes. The ratio between T_b and T_l is 0.471 for $k = 10^{-5}$ when $b = 10\mu$m, while the ratio equals 0.944 for the cutoff wave number $k = 0.256$.

The dependences of the blowup time and the wave number $k_{\max}$ of the most dangerous modes on the film thickness are presented in Fig. 4 for different slip lengths. It can be found that the growth rate will decrease rapidly with the increasing mean thickness of

the films. Hence, the blowup time increases by several orders of magnitude. The meantime k_{max} declines in Fig. 4(b), which implies that the number density of the broken holes will reduce sharply, the same as that already reported in experimental work available. For thinner films, the increase of the slip length will drop down the wave number of the most unstable modes, as shown in Fig. 4(b).

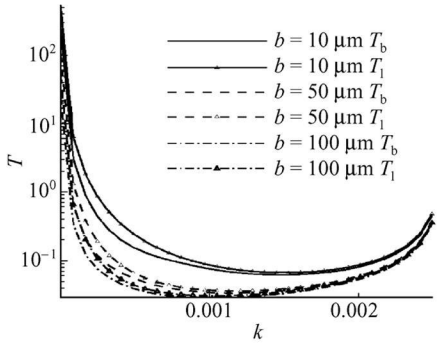

Fig. 3. Comparison of the breakup time based on linear and weakly nonlinear theories for different slipping lengths when h_0= 10nm and ε= 0.001.

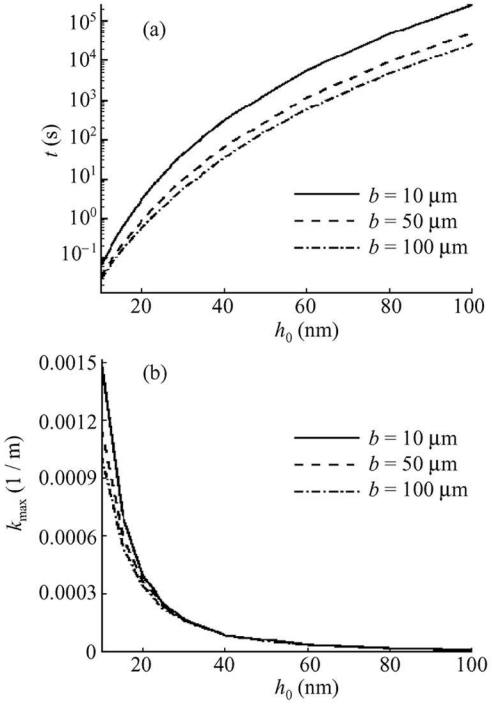

Fig. 4. Effects of film thickness and slip length on (a) the blowup time and (b) wave numbers of the most unstable perturbation when ε= 0.001.

Figure 5 considers the effect of the initial amplitudes of the perturbations on the ratio between T_b and T_l for different slip lengths when h_0= 40 nm. Generally, nonlinearities accelerate the film rupture since the ratios are always less than 1. The decrease of the ratio with the increasing initial amplitude indicates that the nonlinear term plays a more important role for larger ε. The dependence of the ratio is much weaker on the magnitude of the slip length, as shown in Fig. 5. It changes slightly from 0.75 to 0.82 for ε= 0.1, from 0.93 to 0.95 for ε= 0.0001 when b jumps from 10μm to 10cm. Anyway, as predicted by the weakly nonlinear theory, the destabilizing effects are stronger for smaller slip length.

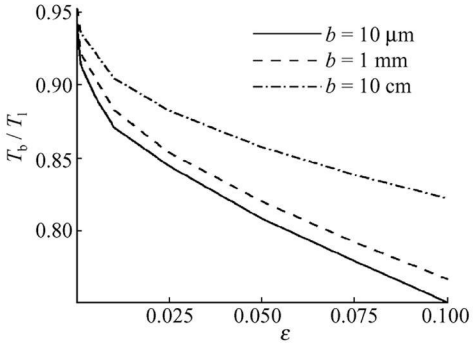

Fig. 5. Variation of the ratio between T_b and T_l with the initial amplitudes for different slip lengths when h_0= 40 nm.

4 Conclusion

To study the effects of the slippage on the stability of ultra-thin films, a mathematical model has been established based on the weakly nonlinear theory. The present analysis reveals that the nonlinearity always accelerates the breakup of the films. As already found in experiments and confirmed by linear analyses, the slipping velocity on the substrate enhances the development of perturbations and decreases the wave number of the most unstable mode, which leads to the decrease of the number density of the holes. The nonlinear effects are more apparent for perturbations with lower wave number. The results suggest that the ratio of nonlinear and linear breakup time will decrease as the magnitude of the initial perturbations increases, which implies the nonlinear effects become stronger. However, the ratio is almost independent of the strength of the slip length for a given initial perturbation.

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