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Dynamics of thin liquid films on surfaces with a time-periodic wettability

Balram Suman, Satish Kumar *

Department of Chemical Engineering and Materials Science, University of Minnesota, 151 Amundson Hall, 421 Washington Avenue SE, Minneapolis, MN 55455, USA

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Abstract

The dynamics of thin liquid films on surfaces whose wettability changes in a time-periodic manner are examined in this work. A nonlinear evolution equation based on the lubrication approximation is used to describe the film height, and attractions due to van der Waals forces are incorporated. Film wettability is varied through an imposed sinusoidal modulation of the Hamaker constant. A linear stability analysis predicts that if the mean Hamaker constant is negative, disturbances at the film surface will eventually decay regardless of the amplitude and frequency of the oscillation. However, numerical solution of the evolution equation shows that the film can rupture at a given frequency if the amplitude is sufficiently large. The associated characteristic wavelength can be predicted from results for constant-wettability surfaces if an appropriate effective Hamaker constant is used. For positive mean Hamaker constants, film rupture can be accelerated, delayed, or prevented depending on how the Hamaker constant changes early in the oscillation cycle. The effects of spatial gradients in wettability are also considered, and it is found that oscillation can delay but not prevent rupture. Inclusion of short-range repulsive forces leads to the formation of droplet-like structures separated by ultra-thin films, but this can be prevented by sufficiently large and slow oscillations of the Hamaker constant. The results of this work may find use in applications that make use of surfaces whose wettability can be controlled by external stimuli.

Keywords: Thin liquid films; Wettability; Time-periodic; Stability; Hamaker constant

1. Introduction

Thin liquid films have been the topic of extensive fundamental study for several decades due to their relevance in numerous natural phenomena and industrial applications [1]. Theoretical investigations typically assume that the wettability of the surface on which the film rests is time-independent. Recently, however, surfaces whose wettability can be varied with respect to time have been developed, and these may find application in emerging technologies such as biochemical sensing, microfluidics, and self-cleaning coatings [2]. In the case where the wettability of a surface changes periodically with time, a thin liquid film may alternately find itself in conditions where wetting is favorable and unfavorable. It is then of interest to know what the final state of the film will be, and how that state depends on the amplitude and frequency of the oscillation. The goal of the present paper is to explore this issue theoretically through the framework of the lubrication approximation.

As reviewed by Liu et al., there are several methods by which the wettability of a surface can be dynamically varied [2]. One way is through surface modification with photo-, electro- and thermo-sensitive polymer molecules whose morphologies can be reversibly switched [3]. A second route is through self-assembled monolayers in which the self-organized structure changes in response to physical or chemical stimuli [4–6]. A third technique makes use of metallic oxide thin films that change wettability when exposed to light [7,8]. Rather than focusing on any one of these specific methods, we consider here a more general problem in which the Hamaker constant is time-varying. The Hamaker constant enters the equations of motion through an effective body force (or disjoining pressure), and this approach has been used in many prior studies to describe the phenomena of liquid film dewetting and rupture [1]. The equations of motion and boundary conditions are simplified using the lubrication approximation to yield a nonlinear partial

* Corresponding author. Fax: +1 612 626 7246.
E-mail address: kumar@cems.umn.edu (S. Kumar).

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differential equation describing film height. As the literature in this area has been discussed extensively elsewhere [1] and continues to evolve (e.g., Refs. [9–11]), we do not provide a review here. Whereas prior works, to the best of our knowledge, have assumed a time-independent Hamaker constant, the distinguishing feature of the present work is the consideration of the time-periodic case. In addition to examining surfaces with a spatially uniform wettability, we also briefly discuss the effects of wettability gradients and short-range repulsive forces. The problem is set up in Section 2, results and discussion are presented in Section 3, and conclusions follow in Section 4.

2. Problem formulation

We consider a thin liquid film bounded below by a rigid horizontal substrate and above by a passive gas. Assuming that the mean height of the film, h_0, is much smaller than the wavelength of a typical disturbance, λ, allows us to apply the lubrication approximation. Letting $\epsilon = 2\pi h_0/\lambda \ll 1$, we define the following dimensionless variables:

$$X = \frac{\epsilon x}{h_0}, \qquad Z = \frac{z}{h_0}, \qquad T = \frac{\epsilon U_0 t}{h_0}. \tag{1}$$

Here, x and z are the coordinates in the horizontal and vertical directions, respectively, and t denotes time. The characteristic velocity in the problem is denoted by U_0, which may be taken as $\mu/(\rho h_0)$, where the μ and ρ are the fluid viscosity and density, respectively. The horizontal and vertical velocity components are scaled by U_0 and ϵU_0, respectively. The scale for pressure is $\mu U_0/\epsilon h_0$, as is the scale for potentials associated with body forces. In this work, we consider only body forces due to van der Waals attraction and short-range repulsion:

$$\phi = \frac{A}{h^3} - \frac{B}{h^4}, \tag{2}$$

where h is the interfacial height, ϕ is the potential for the body force, A is a Hamaker constant, and B is a coefficient characterizing the strength of the short-range repulsive forces. Positive values of A drive film dewetting and rupture, and negative values of A (and positive values of B) impart stability. In our work, we will allow A to be a function of x in order to model the effects of a spatially varying wettability (Section 3.3).

When the above scalings are substituted into the Navier–Stokes equations and appropriate boundary conditions (balance of shear and normal forces at the liquid–gas interface; no-slip and no-penetration at the substrate), and only the leading order terms are retained after suitable rescalings, a nonlinear evolution equation for the dimensionless interfacial height, $H = h/h_0$, can be derived. As very similar derivations have been given elsewhere [1,11], we simply quote the final result:

$$\partial_T H + \frac{\partial_X H}{H}\partial_X A^* - \frac{A^*}{H^2}(\partial_X H)^2 - \frac{\partial_X^2 A^*}{3} + \frac{A^*}{H}\partial_X^2 H + \frac{8B^*}{3H^3}(\partial_X H)^2 - \frac{4B^*}{3H^2}\partial_X^2 H + \frac{1}{3C}\partial_X\left(H^3\partial_X^3 H\right) = 0, \tag{3}$$

where $A^* = \epsilon A/(\rho U_0^2 h_0^3)$ and $B^* = \epsilon B/(\rho U_0^2 h_0^4)$, with $C = U_0\mu/(\epsilon^3\sigma)$ being a capillary number. In this work, the Hamaker constant is taken to vary sinusoidally with time:

$$A^* = A_0 + A_t, \tag{4}$$

where A_0 is a mean Hamaker constant and A_t is taken to be either

$$A_t = \tilde{A}\cos(\omega T), \tag{5}$$

or

$$A_t = \tilde{A}\sin(\omega T), \tag{6}$$

with $\tilde{A}$ and ω being the amplitude and frequency, respectively, of the Hamaker-constant oscillation. Equation (3) assumes that the liquid is Newtonian and incompressible. However, these assumptions may fail if the film becomes sufficiently thin [12]. In addition, the no-slip boundary condition may fail if the substrate is sufficiently smooth and nonwettable [12]. A constant surface tension is also assumed, but this may be inappropriate if the wettability changes are driven by temperature variations.

Equation (3) is solved by discretizing the spatial derivatives using fourth-order centered finite differences, applying periodic boundary conditions, and time-stepping with Gear's algorithm [11]. For cases where the substrate has no wettability gradients and film rupture occurs (A^* independent of X; $B^* = 0$), using 300 grid points is sufficient for obtaining accurate results. For cases with wettability gradients, 600 grid points are employed, and for those with short-range repulsion, 1800 grid points are taken. A film height of less than 0.0025 anywhere in the domain is used as the criterion for determining the rupture time. The length of the simulation domain is taken as 2π; rupture times and interfacial profiles were found to be insensitive to longer domain lengths and to smaller values of the height used to define the rupture criterion.

A sinusoidal perturbation of the liquid–gas interface with amplitude 0.01, wavelength 2π, and phase angle $\pi/2$ is used as an initial condition for cases where wettability gradients are absent. Random initial conditions generated by averaging a sum of sine waves with random phases and amplitudes (no larger than 0.01) were employed for cases with wettability gradients. When wettability gradients are present, knowing where film rupture occurs is just as important as knowing the rupture time itself, which justifies the use of random initial conditions [11]. When they are absent, the place where rupture occurs is less important, so a simpler initial condition is used here; rupture times obtained with this initial condition were found to be the same as those obtained with random initial conditions.

3. Results and discussion

In Sections 3.1 and 3.2, we address the situation where there are no wettability gradients and film rupture can occur. In Sections 3.3 and 3.4, we address the effects of wettability gradients and short-range repulsion, respectively.