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Historical perspective

Stability and break-up of thin liquid films on patterned and structured surfaces

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ABSTRACT

Solid surfaces with chemical patterning or topographical structure have attracted attention due to many potential applications such as manufacture of flexible electronics, microfluidic devices, microscale cooling systems, as well as development of self-cleaning, antifogging, and antimicrobial surfaces. In many configurations involving patterned or structured surfaces, liquid films are in contact with such solid surfaces and the issue of film stability becomes important. Studies of stability in this context have been largely focused on specific applications and often not connected to each other. The purpose of the present review is to provide a unified view of the topic of stability and rupture of liquid films on patterned and structured surfaces, with particular focus on common mathematical methods, such as lubrication approximation for the liquid flow, bifurcation analysis, and Floquet theory, which can be used for a wide variety of problems. The physical mechanisms of the instability discussed include disjoining pressure, thermocapillarity, and classical hydrodynamic instability of gravity-driven flows. Motion of a contact line formed after the film rupture is also discussed, with emphasis on how the receding contact angle is expected to depend on the small-scale properties of the substrate.

Contents

1. Patterned and structured surfaces

Advances in manufacturing over the past several decades led to the ability to fabricate heterogeneous solid surfaces with well-controlled

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spatially periodic variation of physical and chemical properties. The manufacturing techniques are reliable, fast, and inexpensive, and the scales of spatial variation can be as small as fractions of a micron. While these surfaces have many potential applications, the focus of the present review is on their use in situations involving fluid flow. Two types of non-homogeneous surfaces will be discussed. The first one, referred to as patterned surface, is a flat surface with spatially periodic variation of chemical properties. Ultra-thin films of gold [1] or monolayers of octadecyltrichlorosilane (OTS) [2] have been widely used for chemical patterning of smooth substrates such as glass or silicon wafers; common patterns are alternating stripes and squares. The second type is a structured surface, which is characterized by periodic variations of topography (although some authors use the term "structured" for chemically patterned surfaces as well). Examples of structured surfaces include periodic arrays of parallel grooves and arrays of pillars of square or circular cross-section. When a structured surface appears to be in contact with liquid on macroscale, different configurations are possible at the much smaller scale of the structure [3]. The grooves or spaces between the pillars can be filled with liquid so that the liquid is in contact with the solid everywhere along the boundary. This configuration is referred to as the Wenzel state. However, there is also a possibility that the grooves or the spaces between the pillars are filled with the gas phase, a configuration referred to as the Cassie–Baxter state, although some authors refer to it as the Cassie state. Let us briefly discuss some of the applications which motivated the research discussed in the present review.

Using chemically patterned substrates has been suggested as an efficient approach to manufacturing patterns of soft materials by dewetting of liquid films on such substrates. This approach was demonstrated for polymer films in the pioneering experiments of Krausch et al. [4,5] using silicon substrate patterned by micron size metal lines. The substrate pattern resulted in a similar pattern in the polymer film after dewetting. Many other experimental studies of various liquid-substrate combinations show that microstructures of different shapes can be obtained using different types of substrate patterning [6,7]. However, the challenge in these types of applications is that the dewetted configurations can fail to replicate the substrate pattern. This is not surprising since dewetting is typically driven by an instability which may have an intrinsic scale different from the scale of the patterning. The instability length scale depends on film thickness, so rather poor results in terms of substrate pattern replication are observed for both large and small values of the thickness, i.e. when the instability length scale is far from the lateral scale of the patterning. Mathematical models of instability of liquid films on chemically patterned surfaces have been developed in a number of studies to define the optimal conditions for the pattern transfer and are discussed in detail in the present review. More recent applications of these models include novel methods for manufacture of flexible electronics and displays [8]. Other closely related applications of dewetting of patterned surfaces include manufacture of components of microfluidic devices [9].

Significant amount of work on structured surfaces in the Wenzel state has been motivated by heat transfer applications. In these applications, liquid films are typically flowing over heated substrates and the main quantity of interest is the heat transfer coefficient, i.e. the ratio of the heat flux to the temperature difference across the film. Structuring can lead to changes in the heat transfer coefficient compared to flat substrates under similar conditions [10,11]. The reasons for the change are two-fold. First, structuring can lead to changes in the flow patterns in the film. Second, it affects the conditions when the film breaks up and dry patches are formed. The local heat transfer coefficient over the dry areas is typically low, but it can be very high near the contact lines formed at the edges of dry patches, i.e. the lines of contact between the liquid film surface and the solid substrate. To understand this complex interplay of different effects, mathematical modeling is essential, especially when the optimal conditions for heat transfer enhancement are sought.

Structured surfaces in the Cassie–Baxter state are of interest for a number of applications. One is the development of the so-called self-cleaning surfaces. The idea is based on the ability of a large class of structured surfaces, referred to as superhydrophobic, to strongly repel water. A dramatic demonstration of that is an experiment in which a liquid droplet bounces off such surface as if it were a solid ball [12,13]. As a result, stain formation from liquid droplets which spread and then evaporate on typical everyday surfaces can be avoided as droplets bounce or roll off a superhydrophobic surface before they evaporate. Similar ideas motivated recent works on using the small-scale structuring to achieve anti-fogging [14] and antibacterial [15] substrate properties. Another application of structured surfaces is drag reduction. For example, using structured surfaces as walls of a microchannel leads to reduction of viscous resistance of pressure-driven flow in such channel [16]. To better understand the origin of this effect, consider viscous flow in the vicinity of a wall structured by an array of parallel grooves, oriented in the direction transverse to the flow, as shown in Fig. 1. When the grooves are in the Cassie–Baxter state, parts of the bottom boundary of the liquid film are the groove menisci at which the no-slip condition is not applicable. In fact, there is likely to be significant slippage there due to the fact that gas viscosity is much smaller than that of the liquid. This leads to the reduction of the overall viscous flow resistance in the channel. The same qualitative conclusions can be made for a variety of different types of structuring, such as arrays of pillars.

The possibility of drag reduction by using structured surfaces in the Cassie–Baxter state has been explored both experimentally and theoretically. Experimental work on channels with structured walls was pioneered by Ou et al. [16], who found decrease in viscous resistance of about 40% due to the use of structuring. Theoretical description of surfaces in the Cassie–Baxter state is usually based on the idea of treating the structured surface as a flat homogeneous surface at which the viscous flow equations satisfy the Navier slip condition,

$$u = \lambda_{eff} \frac{\partial u}{\partial y}, \tag{1}$$

where u is the flow velocity in the direction along the surface and the y-axis is in the direction normal to it. All the details of micro- or nanostructuring are thus incorporated into the effective slip length, λ_{eff}. Significant amount of theoretical work has been aimed at finding formulas for the effective slip length in terms of the parameters of the structuring. These studies have been reviewed in detail in Chapter 5 of [17], so we do not discuss them here. Instead, we briefly mention some of the recent developments in the field. The main focus of recent studies has been on using the numerical simulations of the gas flow in the grooves to obtain more accurate description of the effect of gas-filled structure on the global flow solutions. Of particular interest are the regimes when the groove menisci deformations are significant. Note that many derivations of the effective slip formulas were based on the assumption of flat menisci, while in reality the menisci can be curved, with possibilities

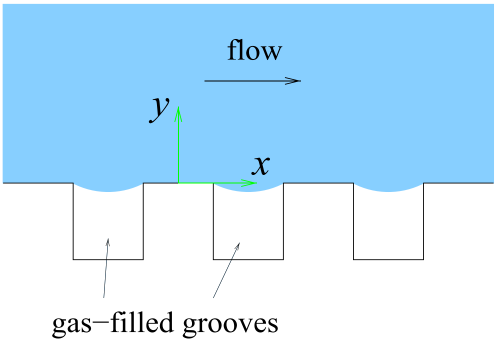

Fig. 1. A sketch illustrating flow past a structured surface in the Cassie–Baxter state. Liquid slippage over the menisci regions leads to an effective slip length.