

Regular Article

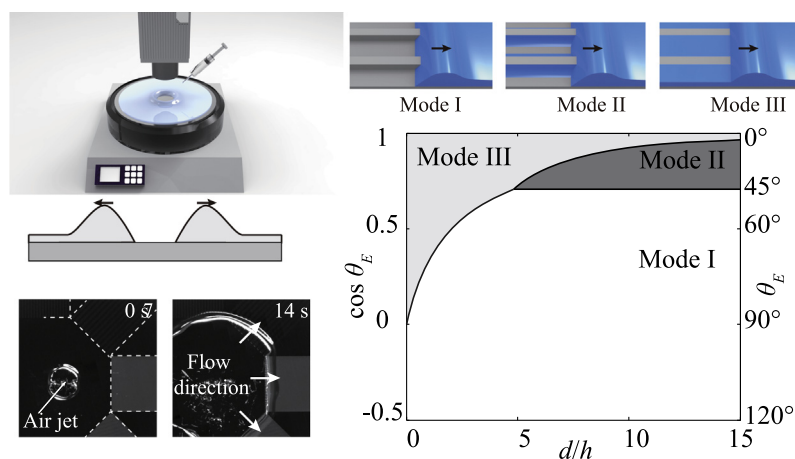
Viscous dewetting of metastable liquid films on substrates with microgrooves

Taehong Kim, Wonjung Kim*

Department of Mechanical Engineering, Sogang University, Seoul 04107, Republic of Korea



GRAPHICAL ABSTRACT



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ABSTRACT

We present a combined experimental and theoretical investigation of dewetting on substrates with parallel microgrooves. A thin, static liquid film has an equilibrium thickness so as to minimize the sum of the surface free energy and the gravitational potential energy. When the thickness of a liquid film is less than the equilibrium thickness, the film seeks the equilibrium through contraction of the wetted area, which is referred to as dewetting. We experimentally observed the dewetting of thin, metastable liquid films on substrates with parallel microgrooves. The experiments revealed that the films retract in the direction along the grooves and leaves liquid residues with various morphologies. We classify the residue morphologies into three modes and elucidate the dependence of the mode selection on the groove geometry and the equilibrium contact angle of the liquid. We also experimentally examined the dynamic motion of the receding contact lines of the dewetting films, and developed a mechanical model for the receding speed. Our results provide a basis for controlling liquid films using microstructures, which is useful for lubricant-impregnated surface production, painting, spray cooling, and surface cleaning.

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1. Introduction

Wetting refers to the spontaneous spreading of a liquid on a dry substrate. The term 'dewetting' is coined to describe the reverse

* Corresponding author.

E-mail address: wonjungkim@sogang.ac.kr (W. Kim).

phenomenon, the spontaneous contraction of a liquid film. When a liquid with a finite volume forms a film on a substrate, the contact angle is determined by Young's equation that dictates the balance of the interfacial energies between the three phases: solid, liquid, and gas. For liquid film with a given contact angle, the film thickness determines the interfacial area, thus the surface free energy, and the gravitational potential energy. The gravitational potential energy increases with the height, but the surface free energy decrease. Therefore, there is an equilibrium thickness at which the sum of two energies is minimized. If the film thickness is less than the equilibrium thickness, the film seeks the equilibrium by contraction of the interfacial area, thus resulting in dewetting.

In daily life, dewetting is observed in the rupture of a thin liquid film on a washstand after washing or in the rapid shrinkage of a water film after rain droplet impact on a car windshield [1]. When birds dive into water and come back out, their water-repellent feathers facilitate the rapid dewetting of water [2]. Dewetting is encountered in engineering problems such as inkjet printing [3], in which dewetting after ink droplet impact on paper should be suppressed for better-quality printing. Dewetting is also widely used to organize materials into micro- or nano-structures [4], including the ordered arrangement of polymer droplets [5,6] and films [7].

The surface free energies of the solid-gas and solid-liquid interfaces depend on the area, so that surface morphology provides the means to control wettability. This technique is exploited by various natural systems, including plant leaves [8], animal eidermis [9,10], and insect wings [11] and legs [12]. Inspired by these natural strategies, engineers have developed functional substrates with microstructures for liquid transport [13], self-cleaning [14], anti-corrosion [15], water evaporation [16], anti-icing [17], and water-condensation [18]. Recently, micropatterned surfaces with impregnated lubricants have attracted great interests thanks to their advantages in terms of condensation enhancement [19], drag reduction [20], and anti-scaling [21].

Attempts have been made to understand the underlying physics to control wetting of a liquid film using microstructures. Tanner [22] pioneered the dynamics of the contact line of a spreading film driven by surface free energy, and he deduced that the radius of a spreading oil film on a bare surface is proportional to $t^{1/10}$ with t being time. Seemann et al. [23] experimentally studied the spreading of a water film on substrates with straight parallel grooves and elucidated the dependence of the spreading modes on the aspect ratio of the grooves and the contact angle. A few research groups investigated liquid spreading on a substrate with micropillars and suggested scaling laws to predict the spreading speed [24–26]. Recently, Kim et al. [27] have constructed scaling laws to estimate the velocity of hemiwicking on rough substrates by combining both the macroscopic and microscopic flow behaviours.

Although wetting and dewetting are phenomena governed by the same physical principles [28], the dynamics is different. It is because the viscous dissipation near the contact line is irreversible and the contact angle hysteresis results in the different interface geometries in wetting and dewetting. Redon et al. [1] studied the dynamics of viscous dewetting on a bare surface with the simplified description of moving contact lines. Snoeijer and Eggers [29] calculated an exact interface shape near the contact line and provided an improved model for the receding speed of the meniscus on a flat surface. Barasjen et al. [30] investigated the dewetting of a thin film on chemically patterned substrates using numerical analysis.

We here present a combined experimental and theoretical investigation of the dewetting of viscous liquids on substrates with parallel microgrooves. Our experiments show that dewetting films can leave residues in various forms that can not be observed from smooth substrates as the microstructures modulate the surface free energies and viscous dissipation near the contact line. We classify the observed residue morphologies into three modes. By examining the changes in surface free energy associated with the contact line motion on microstructured substrates, we elucidate the dependence of the residue morphology. We also estimate the viscous dissipation near the contact line and predict the receding velocity from the balance between free surface energy and viscous dissipation at each residue mode. The experimental details are described in Section 2, and the classification of the residue mode is given in Section 3. A mechanical model for the receding speed is developed in Section 4. As we limit our interest to dewetting of liquid films with micrometer thickness, the intermolecular interactions will be ignored in this study. There are a number of previous papers on the physics [31–35] and applications [36–40] of ultrathin (less than ~ 100 nm) film dewetting involving intermolecular interactions.

2. Experiments

We produced parallel-placed microgrooves made of an epoxy-based photoresist (SU-8) on 4-inch silicon wafers using photolithography. In a single wafer, we set sixteen rectangular regions, and created grooves with a specific dimension in each region. As shown in Fig. 1a, the rectangular patterned regions are visible when reflected by light. The height of the microstructures was $10\ \mu\text{m}$, and the structure width and distance between adjacent structures ranged from $10\ \mu\text{m}$ to $500\ \mu\text{m}$. To control the surface free energy of the substrate, we coated the wafer with Teflon, Octadecyltrichlorosilane (OTS), 1H,1H,2H,2H-perfluorooctyltrichlorosilane (PFOTS), or (3-Glycidyloxypropyl)trimethoxysilane (GPTMS). For Teflon coating, the substrate was covered with 1 wt

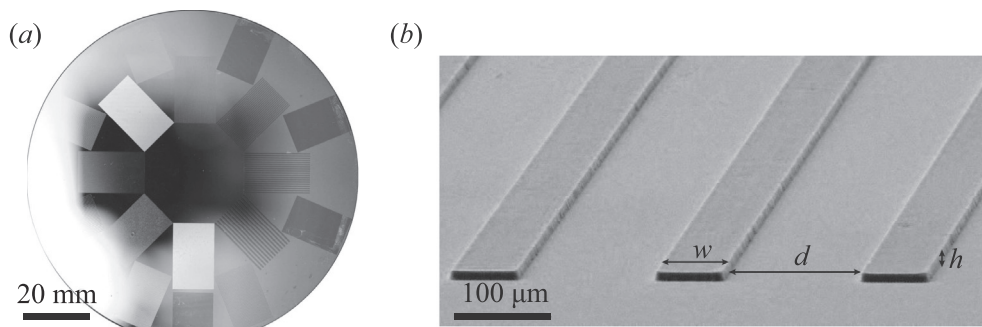


Fig. 1. Microstructures. (a) A single silicon wafer with sixteen rectangular regions, each of which has the grooves with specific dimensions. (b) Scanning electron microscope image of the microstructures.

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