



# Wetting behavior on hybrid surfaces with hydrophobic and hydrophilic properties



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## ABSTRACT

Hybrid surfaces consisting of a micropillar array of hydrophobic and hydrophilic sites were designed and fabricated to understand the effects of their unique surface morphology and chemistry on droplet condensation. Droplet impingement experiments have revealed that hybrid surfaces exhibit high contact angles, which is characteristic of purely hydrophobic surfaces. However, little is known about the wetting behavior of droplets that nucleate and grow on hybrid surfaces during condensation. In fact, condensed droplets display a distinct wetting behavior during the droplet growth phase which cannot be reproduced by simply impinging droplets on hybrid surfaces. In this study, hybrid surfaces with three different spacing ratios were subjected to condensation tests using an environmental scanning electron microscopy (ESEM) and a condensation cell under ambient conditions. For hybrid surfaces with spacing ratio below 2, droplets were observed to form on top and sides of the micropillars, where they grew, coalesced with adjacent droplets, and shed after reaching a given size. After shedding, the top surface remained partially dry, which allowed for immediate droplet growth. For hybrid surfaces with spacing ratio equal to 2, a different wetting behavior was observed, where droplets basically coalesced and formed a thin liquid film which was ultimately driven into the valleys of the microstructure. The liquid shedding process led to the renucleation of droplets primarily on top of the dry hydrophilic sites. To better understand the nature of droplet wetting on hybrid surfaces, a surface energy-based model was developed to predict the transition between the two observed wetting behaviors at different spacing ratios. The experimental and analytical results indicate that micropillar spacing ratio is the key factor for promoting different wetting behavior of condensed droplets on hybrid surfaces.

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## 1. Introduction

Dropletwise condensation is characterized by significantly higher heat transfer, even by an order of magnitude greater when compared to filmwise condensation [1,2]. In the past few years, engineered surfaces have been designed and characterized to promote dropletwise condensation by making use of nano- or micro-scale features [3–13]. Most of the recent studies about dropletwise condensation have focused on superhydrophobic surfaces with micro-scale features. In those cases, it has been found that condensate droplets may partially penetrate the texture of the surfaces [3,12,13]. Under those conditions, droplets do not fully exhibit a

hydrophobic behavior, which could inhibit proper droplet shedding needed for enhanced dropletwise condensation. Condensate droplets under those conditions tend to grow to a large size (~2 mm) until they roll off the surface by gravity [14–16]. However, such a scenario is detrimental to the overall condensation process since large droplets have higher thermal resistance than smaller droplets.

Droplet size should also be reduced while increasing the droplet departure rate so more surface can become available for droplet renucleation. This in turn enables greater heat transfer per contact area [17]. Recent studies have focused on nanostructured surfaces from which suspended condensate droplets can spontaneously eject or jump when they reach a relatively small droplet diameter (less than 100 μm) [5,6,10,11,18]. However, such a movement could be lost when droplets grow very fast and coalesce with neighboring droplets [19,20]. Also, the air entrained in the nanocavities found in those surfaces could contribute to a decrease in heat transfer rate [11].

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Hybrid surfaces consisting of a micropillar array of hydrophobic and hydrophilic sites were designed to promote dropwise condensation. Although studies have reported on heterogeneous (discretized) nucleation behavior [7] and distinct wetting behavior [21] of droplets on hybrid surfaces, they provide little insight into the role of surface morphology on condensation. In this paper, we report on the effects of surface morphology and chemistry on the wetting behavior of condensate droplets on hybrid surfaces. Experimental observations reveal that condensate droplets form on top of each micropillar until they coalesce with neighboring droplets. Once they reach a given size, the coalesced droplets shed from the surfaces when the micropillar spacing is less than 50  $\mu\text{m}$ . However, coalesced droplets fill the valleys of the hybrid surfaces when the micropillar spacing is about 50  $\mu\text{m}$ , forming a thin liquid film which leads to a combination of dropwise and filmwise condensation mechanisms. Once the droplets have shed, the tops of the micropillars experience dropwise condensation again. In order to understand condensation phenomena on hybrid surfaces, the effects of surface morphology and surface chemistry on droplet growth dynamics were investigated analytically and experimentally.

## 2. Surface design and methods

### 2.1. Surface design criterion

It is known that with an increase in surface roughness, droplets prefer to sit on top of heterogeneous interfaces consisting of peaks and air gaps [22]. When droplets sit on a surface, the liquid could be in the Cassie–Baxter wetting state, where droplets sit on top of air-filled cavities, or in the Wenzel wetting state, where droplets wet the space or cavities below them. In order to design and fabricate hybrid surfaces that can ensure the Cassie–Baxter condition, a simple surface energy based analysis [4,8,13,23–25] has been undertaken to prescribe the desired wetting state for droplets. Surface energy based analysis of a predetermined hybrid surface including its boundary conditions was undertaken by accounting for the surface energy of a characteristic unit cell. Fig. 1a shows a droplet sitting on a hybrid surface in the Cassie–Baxter state [19]. For such a condition, the surface energy of the interfacial system 1 (IS1) within a unit cell is given by:

$$E = \gamma_{SL1} \times \left( \frac{a^2}{(a+b)^2} (a+b)^2 \right) + \gamma_{SV2} \times \left( 4ah + \left( 1 - \frac{a^2}{(a+b)^2} \right) (a+b)^2 \right) + \gamma_{LV} \times \left( \left( 1 - \frac{a^2}{(a+b)^2} \right) (a+b)^2 \right) + \gamma_{LV} \times \cos \theta_h (a+b)^2 \quad (1)$$

where  $\gamma_{SL1}$ ,  $\gamma_{SV2}$ , and  $\gamma_{LV}$  is the surface tension at the solid–liquid interface of the hydrophilic surface, surface tension at the solid–vapor interfaces of the hydrophobic surface, and the surface tension at the liquid–vapor interface, respectively. The micropillar width and gap size are denoted as  $a$  and  $b$ , respectively, and  $h$  denotes the micropillar height.  $\theta_h$  is the droplet contact angle on the hybrid surface.

Fig. 1b shows a droplet sitting on a hybrid surface in the Wenzel state [14]. For such a condition, the surface energy of the interfacial system 2 (IS2) within a unit cell is given by:

$$E = \gamma_{SL1} \times \left( \frac{a^2}{(a+b)^2} (a+b)^2 \right) + \gamma_{SL2} \times (4ah) + \gamma_{SL2} \times \left( \left( 1 - \frac{a^2}{(a+b)^2} \right) (a+b)^2 \right) + \gamma_{LV} \times \cos \theta_h (a+b)^2 \quad (2)$$

where  $\gamma_{SL2}$  is the surface tension at the solid–liquid interface of the hydrophobic surface.

To determine the critical condition corresponding to the transition between interfacial system 1 and interfacial system 2, the surface energies of both interfacial systems must be the same. Namely, Eqs. (1) and (2) are set equal to each other as follows:

$$\begin{aligned} & \gamma_{SV2} \times \left( 4ah + \left( 1 - \frac{a^2}{(a+b)^2} \right) (a+b)^2 \right) + \gamma_{LV} \\ & \times \left( \left( 1 - \frac{a^2}{(a+b)^2} \right) (a+b)^2 \right) \\ & = \gamma_{SL2} \times \left( 4ah + \left( 1 - \frac{a^2}{(a+b)^2} \right) (a+b)^2 \right) \end{aligned} \quad (3)$$

For a liquid droplet on a flat hydrophobic surface, the well-known Young's equation [21] can be expressed as follows:

$$\cos(\theta_{\text{equ}}) = \frac{\gamma_{SV2} - \gamma_{SL2}}{\gamma_{LV}} \quad (4)$$

where  $\theta_{\text{equ}}$  is the equilibrium contact angle of a liquid droplet on a flat hydrophobic surface

Eq. (3) can then be simplified by using Eq. (4) as follows:

$$\theta_{\text{critical}} = \cos^{-1} \left( \frac{A-1}{R-A} \right) \quad (5)$$

$$A = \frac{a^2}{(a+b)^2}$$

$$R = 1 + \frac{4h}{a(1+b/a)^2}$$

where  $\theta_{\text{critical}}$  is the critical contact angle that denotes the transition between interfacial systems 1 and 2.

The critical angle ( $\theta_{\text{critical}}$ ) obtained by using Eq. (5) was used to compare with the experimental contact angle data. Since it is difficult to find the equilibrium contact angle under ambient conditions [4,12,26], the average contact angle ( $\theta_{\text{ave}}$ ) was defined as follows:

$$\theta_{\text{ave}} = \frac{(\theta_{\text{adv}} + \theta_{\text{rec}})}{2} \quad (6)$$

where  $\theta_{\text{adv}}$  and  $\theta_{\text{rec}}$  are the advancing and receding angles of a liquid droplet on a flat hydrophobic surface, respectively.

The desired interfacial system 1 or 2 can be predicted using Eq. (5) by specifying the appropriate micropillar width ( $a$ ), gap size ( $b$ ), and micropillar height ( $h$ ). From Eq. (5), the calculated critical angle ( $\theta_{\text{critical}}$ ) can be calculated, which can then be compared with the average contact angle ( $\theta_{\text{ave}}$ ) obtained using Eq. (6). When the measured  $\theta_{\text{ave}}$  of a droplet on a flat hydrophobic surface is less than calculated  $\theta_{\text{critical}}$ , interfacial system 2 (as shown in Fig. 1b) becomes more favorable because of its lower surface energy. This explains why the hydrophobicity of the hydrophobic materials is not enough to promote and support interfacial system 1, since droplets would tend to fill the surface roughness (i.e. air cavities) to reach a lower surface energy state. On the other hand, when the measured  $\theta_{\text{ave}}$  is greater than the calculated  $\theta_{\text{critical}}$ , the interfacial system 1 becomes more favorable in terms of surface energy resulting in the settling of droplets on top of the micropillars. Therefore, Eq. (5) can be used to predict which of the two distinct wetting states may prevail on hybrid surfaces consisting of hydrophilic tops or spots and hydrophobic valleys exclusively.

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