Condensation Limits on Biphilic Surfaces

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1. Introduction

Water vapor condensation is routinely observed in nature and have a large influence on the performance of engineered systems such as power generation [1], and high-/heat-flux thermal management [2]. In particular, condensation on non-wetting surfaces, termed dropwise condensation, has been the topic of many investigation due to its enhanced heat transfer coefficient when compared to filmwise condensation [3]. With the advancement of micro/nanofabrication techniques in the past decade, superhydrophilic surfaces with ultra-high adhesion and biphilic surfaces which combine hydrophilic and hydrophobic regions in a single surface have been developed [4]. When water vapor condenses, droplet nucleation preferentially occurs on the regions which have relatively high surface energy [5]. For instance, in the case of condensation on hydrophobic or superhydrophilic surfaces, many droplets nucleate on region with defects which act to expose the high surface energy substrate. This phenomenon can be found on a range of surfaces ranging from hydrophilic to superhydrophilic. Therefore, rationally designed biphilic surfaces offer a controlled means of controlling heterogeneous nucleation by including high surface energy hydrophilic spots for preferential nucleation. A number of studies have been conducted in effort to fabricate the biphilic surfaces and find advantages of using them. Varanasi et al. fabricated biphilic posts and achieved successfully control of heterogeneous water nucleation [6]. Yao et al. studied the droplet contact angle behaviour on the biphilic surfaces [7], while Betz et al. manufactured and tested boiling heat transfer on super-biphilic surfaces [8]. However, past techniques to fabricate biphilic surfaces are either expensive, or labour intensive, involving many fabrication steps. Furthermore, the governing physics and limits of spatially controlled heterogeneous nucleation remain unexplored. Here, we develop a simpler method to fabricate silicon-based smooth and micro-structured biphilic surfaces. By studying the efficacy of our fabrication process using optical microscopy, we elucidate the breakdown of biphilicity for geometries having sparse hydrophilic areas exceeding pitch-to-spot size ratios of ≥50. Using three-dimensional (3D) numerical simulations, we develop an understanding of the transient temperature profiles during condensation in ambient conditions and identify a vapor diffusion mediated breakdown mechanism. Our work not only develops a simpler means to attain biphilicity, it sheds new light on the limits of achieving spatial control of heterogeneous nucleation of water vapor on surfaces.

2. Fabrication

To create biphilic surfaces, first, we fabricated micropillar arrays on silicon wafers via standard photolithography (Figure 1. (a)-(d)). Note that photore sist was not removed after etching due to a crucial lift-off process at the end. After etching, the surfaces were coated with a conformal layer (≥100 nm) of Octafluorocyclobutane (C₄F₈) by chemical vapor deposition to uniformly functionalize the surfaces to be hydrophobic and to ensure dropwise condensation (Figure 1. (e)). To render the surfaces biphilic, we utilized the lift-off method to remove the masking photore sist on pillar tops, thereby removing all hydrophobic coatings on top of the photore sist patches (Figure 1. (f)). After completing the fabrication process, the surfaces became biphilic with hydrophilic pillar tops (smooth silicon) and hydrophobic pillar sides and sample base (C₄F₈ coated). To study the breakdown mechanisms of biphilicity during condensation, smooth and structured biphilic surfaces having different geometric designs were fabricated, with pillar center-to-center spacings of P = 15 to 150 µm, lengths L = 10 to 15 µm, and pillar heights h = 0 to 15 µm. The novel lift-off technique allowed us to make the biphilic surfaces much faster as it can be easily applied to various heights and designs. For comparison, we also fabricated homogeneous and conformally coated hydrophobic surfaces having the identical designs as the biphilic surfaces. To fabricate the hydrophobic surfaces, all process steps were identical to the biphilic process, however, the last lift-off step was skipped. Therefore, top, bottom, and all sides of the pillars were uniformly covered with C₄F₈ with having the same intrinsic advancing and receding contact angles as the hydrophobic areas on the biphilic surfaces.

3. Experiments

Droplet nucleation during atmospheric water vapor condensation was studied using a custom-built top-view optical light microscopy setup. The biphilic surfaces were horizontally mounted to a cold stage using a thin layer of thermal grease and cooled to the target temperature of T_Air = 7°C with cooling rate of T_AIR = 5, 10, 20 °C/min in a laboratory environment having ambient temperature of T_Air = 23 ± 2°C and a relative humidity (RH) of φ = 42 ± 2%. Video recordings were performed at variable frame rates with a high-resolution camera (DS-Q2, Nikon) attached to an upright microscope (Eclipse LV100, Nikon). Top-view imaging was performed with a 20x (Tu Plan Fluor EPI, Nikon, Lens) lens. The working distance was measured to be 5 ± 0.5 mm.

4. Numerical Model

In order to understand the temperature distributions on biphilic pillar surfaces, and to conduct transient water vapor diffusion simulations, we developed a transient heat transfer simulation of the biphilic surfaces using the commercial code ANSYS FLUENT v 17.1. Due
to the high complexity of analysing the entire domain, only one-unit cell having both smooth silicon and CaF$_2$ coated areas was simulated with suitable symmetry boundary conditions. All operating conditions were set as same as the ambient condition during experiments.

5. Results and Discussion

In order to test the fidelity of the biphilic surfaces, we first performed condensation tests to characterize the efficacy of spatial control of heterogeneous nucleation. Figure 2 shows time lapse images of top-view condensation on the homogeneous, flat biphilic, and structured biphilic surfaces. Good selectivity was observed for low pitch to length ratios (Figure 2. (a)). However, breakdown of biphilicity occurred when the spacing between hydrophilic features increased (Figure 2. (b)). To quantify the spatial control of heterogeneous nucleation during condensation on biphilic surfaces, we defined the biphilic effectiveness as follows:

$$\text{Effectiveness} = \frac{\text{Number of droplets on a pillar top}}{\text{Total number of droplets}} \times 100(\%)$$

(1)

Figure 3 shows the effectiveness on nine different pillar designs as a function of ratio of pitch (center-to-center, $P$) to length (L) of the pillars ($P/L$) for hydrophobic samples. The first four ratios have $L = 10 \text{ µm}$ with different center-to-center distance, and the remaining five have $L = 15 \text{ µm}$ with different center-to-center distance. We also conducted nucleation on three differing pillar heights (5, 10, and 15 µm) in effort to determine the effect of cooling rate and pillar heights on effectiveness. For each sample, we collected data on three spatially random locations for every case and averaged the values and determined the standard deviation. As we can see in Figure 3, nucleation barely occurred on pillar tops for all cases because the hydrophobic surfaces did not have wettability differences. Most nucleation occurred on the bottom of the surfaces for all cases except when $L = 15 \text{ µm}$ and 5 K/min cooling rate.

Figure 4. Effectiveness of nucleation on the biphilic surfaces for nine different $P/L$ ratio with having (a) 0 µm, (b) 5 µm, (c) 10 µm, and (d) 15 µm pillar heights.

6. Conclusions

In this study, novel fabrication technique implemented in effort to fabricate the biphilic surfaces. By comparing the hydrophobic and biphilic nucleation behaviour, we showed that the biphilic surfaces which have different center-to-center distance, length, and height can provide preferable nucleation zones for spatial control of the nucleation as shown in previous studies. ANSYS FLUENT transient simulation was conducted in effort to predict heat transfer and temperature difference of the surfaces while the temperature cooled down. By comparing various ratio of center-to-center and length ($P/L$), we found that the breakdown occurred at high pillar spacing and this is mainly due to a decrease in the formation of isolated vapor depletion zones and temperature of a bottom of the surfaces. We also found that nucleation could be occurred even with very small temperature difference between a pillar top and the bottom of the surfaces. This study offers significant insights into the limits of biphilic condensation and provides avenues for further improvement of the biphilic condensing surfaces for a plethora of applications.

References