Condensate Droplet Size Distribution on Lubricant-Infused Surfaces

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Abstract

Condensation is a ubiquitous phenomenon in nature and industry. Heat transfer rates during dropwise condensation on non-wetting substrates can be 6-8X higher than heat transfer rates during traditional filmwise condensation on wetting substrates. Dropwise condensation on lubricant-infused surfaces (LIS, or SLIPS) is particularly interesting due to high droplet mobility on these surfaces. To accurately predict heat transfer rates during dropwise condensation, the distribution of droplet sizes must be known. Here we present condensation studies of water on aluminum-based lubricant-infused surfaces with a wide range of lubricant viscosities (12 – 2717 cSt) to determine droplet size distributions. Through optical imaging and microscopy, we show that the distribution of droplet sizes on LIS is independent of lubricant viscosity, and agrees well with the model developed by Rose for the distribution of droplet sizes on hydrophobic surfaces, especially in the range $10 < r < 100 \mu\text{m}$. Using artificial sweeping experiments and numerical modeling, we investigate the dependence of sweeping rates on the distribution of droplet sizes and on average heat transfer rates. The maximum size to which droplets grow before being swept decreases rapidly with only a modest decrease in sweeping period, from 750 to 62 μm . Yet, the distribution of droplet sizes and heat transfer rates are nearly unaffected by the change in sweeping period, due to a relative insensitivity of heat transfer to droplets with radii $r > 100 \mu\text{m}$ due to a high conduction resistance within these droplets. Our work provides an experimental and analytical framework to predict heat transfer and sweeping rates for water condensation on a vertical plate coated with a LIS or SLIPS surface.

Keywords: dropwise condensation; droplet size distribution; sweeping; heat transfer; lubricant infused surface (LIS); slippery liquid infused porous surface (SLIPS)

1. Introduction

Condensation is pervasive both in nature [1,2] and industrial applications, including power generation [3], closed-loop electronics cooling systems [4,5], air conditioning [6], desalination [6,7], and water harvesting [8]. On industrial condenser applications, which are typically covered with a high surface energy metal oxide (i.e. Al_2O_3 , CuO , FeO), condensation initiates in the filmwise mode, typified by the formation of a relatively thick liquid film ($\sim 100\text{ }\mu\text{m}$) on the cold surface which acts as a thermal resistance to heat transfer (Figure 1a). If the exterior condenser surface is coated with a low surface energy promoter, discrete droplets form, grow, and shed rapidly via gravitational force, allowing for the re-nucleation of small low-thermal-resistance droplets, in what has been termed dropwise condensation (Figure 1b) [9,10]. Heat transfer rates during dropwise condensation in a pure saturated vapor have been shown to be 6-8X higher than traditional filmwise condensation [9]. After 80 years of research [11], and limited progress due to durability concerns, dropwise condensation has recently gained renewed attention due to its potential in phase-change-based heat transfer applications and significant advances in microfabrication and functional coating techniques [9–16]. Specifically, researchers have been developing novel methods to further control the condensing surface wettability, which is known to strongly affect the mobility of condensate droplets. Mobile droplets, i.e. those with low contact angle hysteresis, clear nucleation sites more rapidly and thereby allow for high nucleation rates and thus high heat transfer [17].

Figure 1: Different stages of filmwise and dropwise condensation. (a) Filmwise condensation on a vertical plate. (b) Dropwise condensation on a vertical plate. (c) Schematic and (d) photograph of recently nucleated droplets shortly before reaching the effective transition radius r_e . These small droplets grow mainly via direct condensation. (e, f) For $r_e < r < r_{\max}$ droplets grow mainly through coalescence with neighboring droplets. (g, h) Once droplets are large enough, the gravitational body force overcomes pinning surface tension forces and droplets slide or sweep down the surface.

To promote dropwise condensation, the solid surface requires a low-energy promoter. Traditionally, low-energy coatings such as fluoropolymers (e.g. Teflon), or self-assembled monolayers (e.g. dioctadecyl disulphide) have been used [11]. However, these coatings experience low durability and short life times under vapor saturation conditions [3]. Chemical and topographical inhomogeneities create defects where droplets can pin and transition to filmwise condensation [18]. Similarly, microstructured surfaces are generally not suited for

continuous dropwise condensation, as condensate can nucleate within the microstructures and remain in the wetting Wenzel state even after coalescence and growth [19]. Only once the surface structures are small enough, on the order of 10 nm – 1 μ m, can droplets form in the favorable non-wetting Cassie-Baxter state [20–22]. However, stable dropwise and jumping-droplet condensation [23–26] on these nanostructured surfaces can only be achieved for low supersaturations [12,14] due to the presence of coalescence and flooding within surface structures once droplet nucleation densities approach the density of the structures [18,27]. Recently, a new class of surface suitable for dropwise condensation of both water and liquids with lower surface tensions has been introduced: Slippery Liquid-Infused Porous Surfaces (SLIPS), or Lubricant-Infused Surfaces (LIS) [14–16,28–33]. If designed correctly [28,34], the infused liquid, or lubricant, creates an atomically flat interface for water condensation. Due to the absence of defects on the surface, condensed water droplets have extremely low contact angle hysteresis ($\sim 2^\circ$) and very high mobility [28,33], leading to easy shedding and rapid clearing of nucleation sites with increased overall heat transfer [29]. Furthermore, the ultra-low surface energy of the infused liquid, coupled with its ultra-smooth interface, has enabled stable dropwise condensation of lower surface tension fluids ($> 20\text{mN/m}$), not possible with classical hydrophobic promoter surfaces [15].

Although the use of SLIPS, or LIS, for dropwise condensation has been proven to increase droplet mobility compared to traditional smooth solid surfaces [33], more fundamental studies of individual droplet heat transfer and droplet size distributions are needed in order to assess the heat transfer enhancement potential. Given the ultra-smooth liquid interface with the condensing water droplets, the dynamics of condensation may vary from those on smooth copper surfaces for which past droplet size correlations, most notable that of Rose and co-workers [35–37], have

been derived. Furthermore, SLIPS have been shown to have higher nucleation densities [29] and higher droplet mobility which could potentially shift the droplet number density towards smaller ranges [38]. In addition, the oil lubricant layer has the potential to cloak the condensing droplets, creating a vapor diffusion barrier and thus potentially reducing direct condensation and coalescence rates [39]. Hence, to be able to accurately predict dropwise condensation heat transfer rates on LIS or SLIPS, it is important to experimentally determine droplet size distributions for these types of surfaces.

In this work we present experimental measurements of droplet density functions on lubricant-infused surfaces infused with oils of various viscosities (12 – 2717 cSt) and for both cloaking and non-cloaking conditions. We compare our experimental data to the classical Rose correlation for droplet sizes [36]. Finally we integrate our droplet distribution model with the well validated individual droplet heat transfer theory to develop a numerical model of overall surface heat transfer performance. We also discuss the effect of sweeping and droplet mobility on the distribution of droplet sizes on SLIPS and LIS. The work here not only presents the first experimental characterization of condensate droplet distribution on SLIPS and LIS surfaces, it also provides a high fidelity modeling framework for the prediction of condensation heat transfer rates on SLIPS and LIS that can be extended to other phase change processes such as condensation frosting.

2. Dropwise Condensation Heat Transfer Theory

Dropwise condensation is a hierarchical and highly transient process [40]. Droplets nucleate with a critical nucleation radius, $r_{\min} \sim 10 - 100$ nm for water [41], and a nucleation site density N_s . Droplets then grow *via* direct condensation until they come in contact with neighboring

droplets at an effective transition radius $r_e = 1/\sqrt{4N_s} \sim 0.5 - 10 \mu\text{m}$ for water, as shown in Figure 1c,d. After transition the droplets grow mainly *via* coalescence with other droplets (Figure 1e,f). On an inclined surface the droplets will reach a maximum diameter, $r_{\text{max}} \sim 1 \text{ mm}$ for water, at which gravitational forces overcome contact line pinning and the droplet slides down the surface, as shown in Figure 1g,h. Droplets can also be removed *via* sweeping or falling droplets prior to reaching r_{max} , in which case nucleation sites are cleared more rapidly and overall heat transfer rates increase.

Despite being a highly transient process, dropwise condensation heat transfer can be modeled using a steady-state formulation by combining individual droplet conduction analysis with time averaged droplet density functions [35–37,42,43]. For a single droplet, heat transfer can be thought of as a network of thermal resistances [35,42,44–46,30]. The network of serial and parallel resistances is illustrated in Figure 2a,b for droplets on a LIS. The heat transfer q_d through a single droplet is then

$$q_d = \frac{\Delta T_{\text{tot}}}{R_{\text{tot}}}, \quad (1)$$

where ΔT_{tot} is the overall temperature difference between the substrate temperature T_s and the vapor saturation temperature T_{sat} , and R_{tot} is the total thermal resistance through the coating and droplet. The overall thermal resistance R_{tot} through the surface structures and a droplet consists of the interfacial resistance R_i , conduction resistance R_{cond} and the structure resistance R_s , which includes resistances due to the lubricant (usually oil), the micro- or nanostructures (height h_B) and the low-energy surface coating (thickness δ_{coat}), as shown in Figure 2b. It has been recently shown that water droplets can nucleate within the lubricant layer [38]. However, for simplicity, we assume here that droplets nucleate on top of the lubricant layer and are thus not subject to an additional diffusion resistance.

Figure 2: Heat transfer model for dropwise condensation. (a) Total thermal path through a condensing droplet: structure resistance R_s , conduction resistance R_{cond} , and interfacial resistance R_i . Curvature effects cause a temperature jump ΔT_c at the liquid-vapor interface. (b) The structure resistance consists of composite thermal resistances through the micro- or nanostructures, the low surface energy coating, and the infused oil layer. (c) Individual droplet heat transfer (Eq. (10)) and droplet size distribution (Eq. (13)) as a function of droplet radius. The dashed line represents the transition radius between direct condensation and coalescence, r_e (sometimes called the coalescence length). (d) Individual droplet heat transfer normalized by the droplet base area (πr^2) and the cumulative fraction of overall surface heat transfer as a function of droplet radius. 95% of the total heat transfer happens for droplets smaller than 100 μm .

The temperature drop ΔT_i at the droplet-vapor interface arises due to an imbalance of evaporation and condensation of liquid and vapor molecules at the interface, respectively. The interface resistance can then be written as

$$R_i = \frac{1}{h_i 2\pi r^2 (1 - \cos \theta_A)}, \quad (2)$$

where h_i is the liquid-vapor interfacial heat transfer coefficient, and θ_A is the advancing contact angle. The interfacial heat transfer coefficient can be derived from kinetic theory and can be described as [45,47,48]

$$h_i = \frac{2\sigma}{2 - \sigma} \frac{1}{\sqrt{2\pi R_g T_s}} \frac{\rho_v \Delta h_{fg}^2}{T_s}, \quad (3)$$

where ρ_v is the vapor density, Δh_{fg} the latent heat of evaporation, R_g the specific gas constant, and σ the condensation accommodation coefficient. The accommodation coefficient represents the fraction of vapor molecules that will be captured by the liquid droplet. It is often assumed that the accommodation coefficient is equal to unity [42,45]. However, experimental data suggest that for droplets the evaporation and condensation coefficients at atmospheric pressure in the presence of non-condensables and possible hydrocarbon contaminants are closer to $\sigma = 0.04$, which will be used throughout this work [47]. It is also important to note, although more accurate interfacial heat transfer coefficient formulations derived from kinetic theory are available [49–52], they have not been used here due to difficulty of implementation into an analytical based heat transfer model.

Next, a temperature jump ΔT_c due to the droplet curvature, known as the Kelvin effect, has to be taken into account [53,54]. For a droplet with radius r the curvature temperature drop can be expressed as

$$\Delta T_c = \frac{2T_{sat}\gamma_l}{\Delta h_{fg}r\rho_l}. \quad (4)$$

Equation (4) can be re-arranged to

$$\Delta T_c = \frac{r_{min}}{r} \Delta T_{tot}, \quad (5)$$

where r_{min} is the supersaturation dependent critical (or minimum) nucleation radius [55]:

$$r_{min} = \frac{2T_{sat}\gamma_l}{\Delta h_{fg}\rho_l(T_{sat} - T_s)}. \quad (6)$$

For water at atmospheric pressure the critical droplet radius will be on the order of few nanometers ($r_{min} \approx 5$ nm for $\Delta T_{tot} = 5$ K and $T_{sat} = 100^\circ\text{C}$).

If the droplet liquid-vapor interfacial temperature, T_i , is assumed to be uniform, then an analytical temperature drop expression associated with conduction through the droplet, ΔT_{cond} , can be approximated by integration of the local thermal resistance from the droplet base to the liquid-vapor interface, taking into account the advancing contact angle between the droplet and the solid [30]. The conduction resistance then becomes [42]:

$$R_{cond} = \frac{\theta_A}{4\pi r k_l \sin \theta_A}, \quad (7)$$

where k_l is the liquid droplet thermal conductivity. Although the assumption of constant liquid-vapor interfacial temperature is only a crude approximation [56], it is maintained here due its ease of analytical implementation.

Assuming that the substrate temperature and droplet base temperature are uniform, the temperature gradient through the infusion layer will be uniform as well. The structure resistance beneath the droplet includes conduction resistances through the micro- or nanostructures, the low-energy coating, and the lubricant layer. Assuming that the oil infusion and the structures form a level interface with the liquid droplet, and the coating thickness is much smaller than the structure dimensions ($\delta_{coat} \ll h_B$), the structure resistance can be formulated as [30,45]:

$$R_s = \frac{1}{\pi r^2 k_{coat} \sin^2 \theta} \left(\frac{k_B \varphi}{\delta_{coat} k_B + h_B k_{coat}} + \frac{k_{oil}(1 - \varphi)}{\delta_{coat} k_{oil} + h_B k_{coat}} \right)^{-1}, \quad (8)$$

where k_B and k_{oil} are the thermal conductivities of the micro- or nanostructures (here: Boehmite) and the lubricant layer (here: oil), and φ is the solid fraction (here: $\varphi \approx 0.1$).

Now, with all the necessary thermal resistances identified, Eq. (1) can be written as:

$$q_d = \frac{\Delta T_i + \Delta T_c + \Delta T_{cond} + \Delta T_s}{R_{tot}} = \frac{\Delta T_{tot} - \Delta T_c}{R_i + R_{cond} + R_s}. \quad (9)$$

Combining equations (2), (5), (7), and (8) with (9), explicit formula for the single droplet heat transfer rate becomes:

$$q_d = \frac{\pi r^2 (T_{sat} - T_s) \left(1 - \frac{r_{min}}{r}\right)}{\frac{1}{2h_i(1 - \cos \theta_A)} + \frac{r \theta_A}{4k_l \sin \theta_A} + \frac{1}{k_{coat} \sin^2 \theta_A} \left(\frac{k_B \varphi}{\delta_{coat} k_B + h_B k_{coat}} + \frac{k_{oil}(1 - \varphi)}{\delta_{coat} k_{oil} + h_B k_{coat}} \right)^{-1}}. \quad (10)$$

For very small droplets ($r < 100$ nm), the curvature resistance is dominant while conduction through the droplet becomes more important as droplets grow ($r > 10$ μ m). The individual droplet heat transfer is smallest for small droplets and grows as the radius increases, as shown in Figure 2c. Due to a positive spreading coefficient with water, many lubricants (oils) are likely to form a thin oil layer around the individual water droplets, i.e. to cloak them [28,33]. The thickness of the cloaking layer (t) is small, typically on the order of few tens of nanometers [39]. For droplets with $r > 10$ μ m the associated thermal resistance ($\sim t/k_{oil} \approx 10^{-7}$ K·m²/W) for conduction through the oil layer is small compared to that through the droplet ($\sim r/k_{water} \approx 10^{-5}$ K·m²/W) and can be neglected.

To obtain an overall heat transfer rate, Eq. (10) has to be integrated with respect to all existing droplet sizes on the condensing surface. Since the condensation process is highly transient, we need to assume a steady state behavior to obtain an average heat flux. Dividing the droplet growth into the two aforementioned regimes: direct condensation and droplet coalescence with the effective transition radius r_e , and assuming a number density $n(r)$ for small droplets with $r_{min} \leq r \leq r_e$ and $N(r)$ for $r > r_e$, the total heat transfer rate is then [42,46]:

$$q''_{tot} = \int_{r_{min}}^{r_e} q_d(r) n(r) dr + \int_{r_e}^{\hat{r}} q_d(r) N(r) dr, \quad (11)$$

where q_d is given by Eq. (10). The upper bound for the integration for coalescence dominated growth droplets, $\hat{r}$, is related to the maximum droplet radius before sliding, $r_{\max}$. A force balance on the droplet just as it begins to de-pin and slide on a vertical surface due to the gravitational body force gives [45]:

$$r_{\max} = \left(\frac{6(\cos \theta_R - \cos \theta_A) \sin \theta_e \gamma_l}{\pi(2 - 3 \cos \theta_e + \cos^3 \theta_e) \rho_l g} \right)^{1/2}, \quad (12)$$

where θ_A , θ_R , and $\theta_e = \cos^{-1}(0.5 \cos(\theta_A) + 0.5 \cos(\theta_R))$ are the advancing, receding, and equilibrium contact angles, respectively, γ_l and ρ_l are the droplet surface tension and density, respectively, and g is the gravitational constant. Comparison to experimental data shows that $\hat{r} \approx r_{\max}/1.3$ [44]. For water droplets on LIS used in this work we obtain $r_{\max} \approx 460 \mu\text{m}$.

The most commonly used correlation for the number density of larger droplets ($r > r_e$) was developed by Rose and co-workers for dropwise condensation on flat copper plates treated with promoter layers [35,36,44]:

$$N(r) = \frac{1}{3\pi r_e^2 \hat{r}} \left(\frac{r}{\hat{r}} \right)^{-2/3}. \quad (13)$$

Tanaka *et al.* independently studied the distribution of drop sizes and found similar results [37]. The distribution for small droplet sizes ($r < r_e$) has been derived elsewhere, and can be expressed as [42]:

$$n(r) = \frac{1}{3\pi r_e^3 \hat{r}} \left(\frac{r_e}{\hat{r}} \right)^{-2/3} \frac{r(r_e - r_{\min})}{r - r_{\min}} \frac{A_2 r + A_3}{A_2 r_e + A_3} \exp(B_1 + B_2), \quad (14)$$

where A_i and B_i are parameters that can be solved for by enforcing a continuous boundary condition $n(r_e) = N(r_e)$ [45]:

$$A_1 = \frac{\Delta T_{\text{tot}}}{\rho_l \Delta h_{fg} (1 - \cos \theta)^2 (2 + \cos \theta)}, \quad (15)$$

$$A_2 = \frac{\theta}{4k_l \sin \theta}, \quad (16)$$

$$A_3 = \pi r^2 (R_i + R_s), \quad (17)$$

$$B_1 = \frac{A_2}{\tau A_1} \left[\frac{r_e^2 - r^2}{2} + r_{min}(r_e - r) - r_{min}^2 \ln \left(\frac{r - r_{min}}{r_e - r_{min}} \right) \right], \quad (18)$$

$$B_2 = \frac{A_3}{\tau A_1} \left[r_e - r - r_{min} \ln \left(\frac{r - r_{min}}{r_e - r_{min}} \right) \right], \quad (19)$$

$$\tau = \frac{3r_e^2 (A_2 r_e + a_3)^2}{A_1 (11A_2 r_e^2 - 14A_2 r_e r_{min} + 8A_3 r_e - 11A_3 r_{min})}. \quad (20)$$

With equations (13) and (14) through (20), the continuous droplet number density function can be solved, and is plotted in Figure 2c for $r_{\max} = 0.46$ mm.

Figure 2d shows the heat flux per droplet, *i.e.* $q_d'' = q_d/(\pi r^2)$, and the cumulative heat transfer fraction $f = \int q(r)/q_{\text{tot}}$ as a function of droplet radius. It reveals that larger droplets are mostly inactive, *i.e.* more than 80% of the total heat transfer occurs for droplets $1 \mu\text{m} < r < 100 \mu\text{m}$. Thus increasing the mobility of large droplets along with the small droplet number density is necessary to achieve an increase in the overall heat transfer rate. To test the validity of the theory, and to obtain droplet distributions valid for SLIPS and LIS surfaces, we used condensation experiments.

3. Materials and Methods

Porous surfaces were fabricated using a simple fabrication technique where aluminum is immersed in hot water to create a nano-structured surface [57]. First, a polished sheet of aluminum (mirror-like finish, McMaster) of size 1 in x 2 in x 0.05 in was rinsed with acetone, IPA, and deionized (DI) water and blown dry with N_2 . Polished aluminum samples were necessary for these experiments due to the need for a highly reflective surface in order to observe

microscale droplets with a conventional DSLR camera (discussed later). Subsequently, the sheet of aluminum was immersed in hot DI-water near its boiling point ($\approx 95^{\circ}\text{C}$) and was kept there for approximately 15-20 min. The hot water causes a self-limited surface reaction at the aluminum surface to occur, and a thin (≈ 500 nm) boehmite layer is formed, as shown in the inset of Figure 3 [16]. Once the aluminum sheet cooled down to room temperature ($\approx 23^{\circ}\text{C}$), it was treated with air plasma for 1 min to clean the surface from any hydrocarbon contamination and to create free $-\text{OH}$ groups at the surface for effective functionalization. To functionalize the surface, Heptadecafluorodecyltrimethoxy-silane (HTMS) was deposited using vapor phase deposition. The cleaned porous surfaces were placed in a container with a vial of 1 mL HTMS toluene solution (5% v/v). A lid was placed on top to seal the container, followed by heating in an atmospheric pressure oven at 80°C for 3 hours. This process allows for the development of a highly conformal coating as the HTMS molecules evaporate from solution and re-deposit on the porous samples. Contact angle measurements (MCA-3, Kyowa Interface Science Ltd.) of ≈ 300 nL droplets on a HTMS coated smooth silicon wafer showed advancing/receding contact angles of $\theta_{\text{A}}/\theta_{\text{R}} \approx 127 \pm 5.5^{\circ} / 109 \pm 3.5^{\circ}$. Once the conformal coating had been applied, the samples were tested for their superhydrophobicity to ensure a high-quality coating was achieved. Contact angle measurements of ≈ 300 nL droplets on a HTMS coated porous aluminum surface showed advancing/receding contact angles of $\theta_{\text{A}}^{\text{app}}/\theta_{\text{R}}^{\text{app}} \approx 170 \pm 1.5^{\circ} / 165 \pm 4^{\circ}$. Finally, the porous boehmite nanostructures were infused with different oils using a spin coating technique at 500-2000 rpm, depending on the viscosity of the oil. Contact angle measurements of ≈ 300 nL droplets on all of the final infused surfaces showed advancing/receding contact angles on of $\theta_{\text{A}}/\theta_{\text{R}} = 122 \pm 2^{\circ}/118 \pm 2^{\circ}$.

Table 1 lists the lubricant oils with their respective kinematic viscosity, surface tension, and their tendency to cloak. On cloaking lubricants, such as Krytox oils [28,33], the thin lubricant layer surrounding the droplets must drain before the water droplets can coalesce. The kinetics of drainage are governed by the lubricant viscosity [58], and potentially affect the overall droplet size distribution. To test the hypothesis about lubricant viscosity-dependent size distributions, Krytox oils with a wide range of viscosities were used. To study the influence of cloaking on the number density of condensing water droplets, a non-cloaking Carnation Mineral Oil (Sonneborn LLC) was used.

Figure 3: Experimental setup. Steam carried by saturated N₂ and supplied to the sample at two vertically stacked locations (diameter tube ¼", distance between centers 0.3-0.5") condensed on the sample attached to a Peltier cold stage. A DSLR camera, connected to a monitor to assist focusing, was equipped with a regular 70-300 mm telephoto lens and an additional microscope or macro lens. A ring light supplied uniform lighting. A gravity bag and dual needle with gauge 25 simulated high sweeping rates. Inset: SEM image of the nanostructured polished aluminum surface prior to oil infusion showing the knife-like low solid fraction nanostructures with characteristic height and width length scales of ≈ 500 nm.

Table 1: Lubricant oil properties at room temperature.

Name	Viscosity ν [cSt]	Surface Tension γ_{lg} [mN/m]	Cloaking?
Krytox 16256	2717	19	Yes
Krytox GPL 106	822	18	Yes
Krytox 1514	142	18	Yes [28]
Krytox GPL 100	12	17	Yes
Carnation Mineral Oil	12	28 [16]	No [16]

Figure 3 shows the experimental setup for water condensation on LIS. The sample was attached to a Peltier based thermal plate (TP104SC, Instec), herein called the cold stage, using heat transfer tape (McMaster). The cold stage was connected to a temperature controller (mK2000, Instec) that held the cold plate at 0.05 ± 0.1 °C. A second water bath of DI-water was heated to 73 ± 3 °C using a hot plate. Compressed nitrogen was supplied at the bottom of the second bath water tank at a low flow rate. While rising through the hot water the nitrogen gas saturated with water vapor and was then guided through two thermally insulated 1/4" pipes to the sample surface. The outlets of the pipes were arranged vertically at a center-center distance of 0.3-0.5 in. The resulting condensation area spanned roughly 1 in horizontally and 0.7 in vertically and represented the top-most area of a vertical pate. All condensation experiments were done in ambient conditions ($\approx 23^\circ\text{C}$, 50% relative humidity). The presence of non-condensable gases (NCGs) was not a concern since steady-state droplet size distribution densities have been shown to be independent of the concentration of NCGs [43]. A Canon T3i camera captured images of the condensing water droplets at 5184 x 3456 pixels, and videos at 60 fps and 1280 x 720 pixels. A telephoto lens (70-300 mm, Sigma) was set to 300mm and infinity focus. To achieve various levels of magnification with a total range of droplet radii from 1 to 1500 μm , a macro lens (DCR-150, Raynox) or a 5x (TU Plan Fluor EPI, Nikon), 10x (TU Plan Fluor EPI,

Nikon), 20x (TU Plan Fluor EPI, Nikon), and 50x (TU Plan Fluor EPI ELWD, Nikon) magnification infinity focus microscope lenses with step-down adapters were attached to the regular telephoto lens. Table 2 lists the calibration for the different lens combinations for still photographs and videos. For calibration, the outer diameter of a gauge 33 needle or 10 μm Si pillars were measured and compared to the number of pixels within the feature. The sample was illuminated with a LED ring light (LED-144-YK, AmScope). The camera and lens setup were placed on a macro rail and fine linear translation stage (PT1, Thor labs) to allow image focusing. To make focusing easier, the camera was connected to an external monitor. Due to a very shallow depth of focus for high magnification shots, it was crucial to perfectly align the camera to the sample. Any tilt (vertically or horizontally) resulted in droplets that were out of focus and the automated droplet detection software (Matlab) failed.

Table 2: Calibration and resolution of lens setups.

lens	resolution photos [$\mu\text{m}/\text{pix}$]	resolution videos [$\mu\text{m}/\text{pix}$]
Raynox macro	3.15	13.07
5x microscope	0.5	2.11
10x microscope	0.26	1.06
20x microscope	0.14	0.616
50x microscope	0.0625	0.255

To capture droplet data during all stages of a sweeping period and to ensure statistical significance, we captured one image every 10-15 seconds. For the macro, 5x, 10x, and 20x lenses, a minimum of 25, 50, 100, and 100 images were analyzed, respectively. Figure 4 shows an exemplary set of randomly timed images for each magnification. Videos with a 50x lens were used to determine the nucleation site density of droplets. Droplets were counted in the direct condensation regime after a sweeping event, just before coalescence started, i.e. when droplets in

the swept area had a uniform size distribution and were closely packed. The reported values are an average of at least 15 swept areas. Overall, we estimate the error associated with the automated droplet detection to be less than 10%.

Figure 4: Three exemplary transient droplet size distributions captured with a DSLR camera coupled to a (a) macro lens, (b) 5x, and (c) 20x microscope lens.

To simulate higher sweeping rates and to study their effect on the distribution of droplet sizes, additional water droplets were manually supplied to the sample (see Figure 3). A dual twin tip needle with gauge 25 (TS25DSS-1/2, Adhesive Dispensing Ltd) was attached to a gravity bag (Enteral Feeding Gravity Bag, Dynarex) mounted ≈ 1 m above the sample and released individual water droplets ≈ 5 mm above the viewing area. The dispensed droplets would slide down the sample to sweep existing droplets at a rate that was controlled by the flow rate of the water feed. Droplet sweeping rates of 0.3 Hz, 0.6 Hz, and 1.4 Hz were studied.

4. Results and Discussion

4.1. Natural Droplet Size Distribution

We first measured the average, i.e. steady state, distribution of droplet sizes on all samples (Table 1) without artificial sweeping. Figure 5a shows the droplet number density $N(r)$ as a function of droplet radius on the five lubricant oils. All data points fall onto the same curve, irrespective of lubricant viscosity or tendency to cloak. It has been shown previously that steady state droplet size distributions are independent of the magnitude of heat transfer [35–37,43]. It can be expected that cloaking influences heat transfer rates, as the oil layer surrounding the water droplet acts as a diffusion barrier to water vapor. However, given the insensitivity of the steady state droplet size distribution on heat transfer rates, the present findings support previous observations of self-similar droplet number densities [36]. The viscosity of the lubricants studied here also does not influence the droplet size distribution of the condensing water droplets. The most predominant effect of lubricant viscosity is the dynamic friction between lubricant and droplet which influences droplet sweeping velocities. However, the droplet size distribution depends mostly on the maximum size that droplets can grow to just before sweeping, and does not depend on the droplet velocity after sweeping. The maximum radius to which a droplet can grow before gravity causes sweeping is independent of the lubricant viscosity. Thus during the direct condensation and coalescence regime, the distribution of droplet sizes is independent of the lubricant viscosity, as well as cloaking. However, we did notice a marked difference in durability when comparing the low and high viscosity lubricants. Low viscosity oils (12 cSt) experienced a relatively fast (~ 1 hour) depletion from the boehmite structures, irrespective of their tendency to cloak. After two hours of operation, the advancing contact angle of water droplets on the low viscosity oil surfaces increased from $\approx 120^\circ$ to $\approx 160^\circ$, that of non-infused

boehmite structures. While the LIS surfaces with lubricants of higher viscosity (> 140 cSt) could be used for many hours (>10 h) without loss of performance, boehmite structures started protruding from the Krytox GPL 100 and Carnation Oil shortly after condensation started. Similar observations of viscosity-dependent oil drainage have been reported previously [59]. Yet, there seems to be little to no effect on the average distribution of drop sizes even after partial depletion.

Figure 5: Steady state number density, i.e. droplet size distribution, and water droplet sliding on all LIS surfaces without artificial sweeping. (a) The dashed and dotted lines show the original and modified number density function by Rose [36] with $\hat{r} = 0.36$ mm, and the solid line shows the fit from Eq. (21). The left vertical line shows the theoretical maximum droplet radius ($r_{\text{max,th}} = 0.46$ mm), and the right dashed line the experimentally determined radius ($r_{\text{max,exp}} = 0.7 - 0.8$ mm). (b) Vertical center of mass droplet location along the sample as a function of time obtained with the macro lens. Initial droplet sliding sizes and resulting sliding velocities were determined to be $r_{0,\text{K-16256}} = 0.83$ mm/ $V_{\text{K-16256}} = 1.3$ mm/s, $r_{0,\text{GPL106}} = 0.97$ mm/ $V_{\text{GPL106}} = 1.90$ mm/s, $r_{0,\text{K-1514}} = 0.95$ mm/ $V_{\text{K-1514}} = 1.91$ mm/s, $r_{0,\text{GPL100}} = 0.77$ mm/ $V_{\text{GPL100}} = 55.3$ mm/s, and $r_{0,\text{Carnation}} = 1.08$ mm/ $V_{\text{Carnation}} = 22.5$ mm/s, respectively. Coalescence with subsequent droplets as sliding ensued resulted in increasing droplet size as it moved along the plate. However, the size difference was not appreciable for the droplet examined.

Figure 5a also includes the Rose model [36] (Eq. (13)) with $\hat{r} = 0.36$ mm. Our experimental data points agree well with the Rose model, especially in the range $10 < r < 200$ μm , irrespective of the lubricant oil. For $r < 10$ μm , the Rose model slightly underestimates the observed number density of condensing droplets. We propose two possible reasons for the underestimation. First,

Rose and co-workers were limited to observing droplets with $r > 10 \mu\text{m}$ [35], and thus any data below this droplet size is an extrapolation of their findings. Second, we attribute the underestimation to the discrepancy in the number of active nucleation sites, which are a function of supersaturation. While Rose and co-workers [35,44] used values up to $N_s = 2 \times 10^{12} \text{ m}^{-2}$ with $r_e \approx 400 \text{ nm}$ to validate their results, we observe nucleation site densities of approximately $N_s \approx 1-5 \times 10^{11} \text{ m}^{-2}$ with $r_e \approx 0.8-1.4 \mu\text{m}$ due to the presence of non-condensables which result in lower supersaturations and heat fluxes in our experiments. To better account for the higher number of small droplets, we can describe the number density function for droplets on LIS as:

$$N(r) = a r^{-b} . \quad (21)$$

The constants a and b can be determined from a fit to all data points, as shown in Figure 5a. For $[r] = \mu\text{m}$, we find that $a = 6 \times 10^7$, and $b = 3.05$, where the unit of a is such that $[N(r)] = 1/\text{mm}^3$. Note that b is slightly higher than most previously found exponents, which are generally between 2.6 and 2.7 [37,43,47]. Although the number of larger droplets is slightly overestimated by the Rose model, droplets in this size range ($r > 100 \mu\text{m}$) participate negligibly in the overall heat transfer process due to their increased thermal conduction resistance (Figure 2d) [35]. Thus, for heat transfer modeling purposes, the overestimation of large droplets is not critical and can be ignored. Figure 5a also shows the theoretical and experimental maximum droplet radius at the onset of sliding. While theoretical estimation of the maximum departure radius using Eq. (12) yields $r_{\text{max,th}} = 0.46 \text{ mm}$, we did not observe droplet sliding before droplets reached $r_{\text{max,exp}} = 0.7 - 0.8 \text{ mm}$. The discrepancy between model and experiment arises from 1) simplifications made in the development of the theoretical model governing Eq. (12) (ideal contact line, constant contact angle hysteresis, continuous shape of the contact line – oval, circle), and 2) edge effects

at the three-phase contact line of LIS surfaces where the lubricant oil forms a meniscus at the water interface and increases its adhesion force [39,58].

Although not a factor in the droplet distribution results and hence inconsequential to the overall surface heat transfer for the viscosities tested here, the velocity of droplet sliding down the surface was inversely proportional to the viscosity of the lubricant oil used, in agreement to previously reported studies [28,33]. Figure 5b shows the position of sliding droplets as a function of time for all lubricant oils tested. While the steady sliding velocity on Krytox 16256 ($\nu = 2712$ cSt) was only $V = 1.30$ mm/s, the droplet speed on Krytox 1514 ($\nu = 142$ cSt) was $V = 1.91$ mm/s and increased to $V = 55.3$ mm/s on Krytox GLP100 ($\nu = 12$ cSt). We also note that droplets on the non-cloaking carnation oil have higher sliding velocity than on the cloaking Krytox GLP100 oil of same viscosity. This observation suggests that in addition to lubricant viscosity, the lubricant meniscus near the macroscopic three-phase contact area influences droplet dynamics. Also note, however, although the infused oil viscosities tested here (12 – 2717 cSt) had no effect on the droplet distribution, this may not always be the case. One can imagine if the oil viscosity is so high that the sliding velocity of departing droplets is impeded, that flooding of the surface with large conduction limited droplets may occur.

4.2. Heat Transfer and Sweeping Modeling

To quantify the influence of sweeping on average heat transfer rates, and to bring the present results into context, we developed a numerical model. The model determines sweeping rates f_{cycle} , maximum radius of sweeping droplets r_{sw} , maximum radius to which droplets grow before being swept r_s , and heat transfer rates q'' as a function of plate height y (maximum height $L = 1$ m). Figure 6 shows a schematic of the numerical model. Similar to Rose [44], we first assume that all falling, i.e. sweeping droplets, originate at the top of the plate. This assumption is

justified, as droplets further down the plate will not grow to $r = r_{\max}$ as they will be swept prior to reaching this critical size for onset of sliding, i.e. $r_s < r_{\max}$. We further assume that droplets with $r = r_{\max}$ are distributed evenly within this top region. The average area coverage of these larger droplets is $\eta \approx 82\%$, as derived from the data presented in section 4.1. We can then discretize the width of the plate into individual slots of width $B = 2r_{\max}/\eta$ (here: $B = 1.12$ mm), containing one droplet with $r = r_{\max}$ each. When droplets slide down the vertical plate they coalesce with smaller droplets ahead of them and grow in size. Assuming that the shape and contact angles of a sliding droplet are constant, the radius at location y becomes:

$$r_{sw}(y) = V(y)^{1/3} \cdot g(\theta_e), \quad (22)$$

where $g(\theta_e)$ is a geometric factor relating the contact angle to the volume of the droplet. For a spherical cap:

$$g(\theta_e) = \left(\frac{\pi(2 + \cos(\theta_e))(1 - \cos(\theta_e))^2}{3\sin^3(\theta_e)} \right)^{1/3}. \quad (23)$$

Figure 6: Schematic of numerical sweeping model. Droplets grow to $r_{\max}$ in the top-most region of the plate and are equally distributed within slots of width B . When droplets slide down the vertical plate they sweep areas in neighboring slots. Sweeping cones are marked with the blue dotted lines. At some point y down the plate (red dot)

where $r_{sw}(y) = B$ the condensing droplets get swept three times as often as at the top of the plate: once from droplet originating within the same slot, and twice from sliding droplets from the two neighboring slots. The same principle applies further down the plate, with droplets from other slots sweep the area. For every integration step, the droplets sweep an area $dA = dL \cdot 2r_{sw}(y)$ that contains droplets with a droplet size distribution following Eq. (13) with $r_{max}(y) = r_s(y)$.

We further assume that the droplet size distribution $N(r)$ follows the Rose model with $\hat{r}(y) = r_s(y)/1.3$ everywhere on the plate, and that it has a continuous boundary condition with $n(r)$ at r_e (here: $r_e = 1.58 \mu\text{m}$), and that no flooding occurs at the bottom of the plate due to accumulation of sweeping droplets.

To model sweeping rates as a function of plate height we first need to determine the steady state, i.e. average heat transfer and mass transfer rates at the top of the plate to define a characteristic time for dropwise condensation, t_{ch} . Using the latent heat of condensation, Δh_{fg} , we can write:

$$q''_{tot} = \frac{m''}{t_{ch}} \Delta h_{fg} , \quad (24)$$

where q''_{tot} is described by Eq. (11), and m'' is the area-normalized average mass of the condensed droplets. The mass of all condensed droplets can be described using the droplet size distribution:

$$m'' = \rho_l \left(\int_{r_{min}}^{r_e} V(r) n(r) dr + \int_{r_e}^{r_{max}} V(r) N(r) dr \right) , \quad (25)$$

where $V(r) = (r g(\theta_e))^3$. Note that the upper bound on the second integral is r_{max} as opposed to $\hat{r}$, which is typically used for the calculation of the heat flux. For condensation in a pure water vapor environment at $T_{sat} = 100^\circ\text{C}$ and $\Delta T = 5 \text{ K}$, the characteristic time is $t_{ch} = t_{cycle}(y = 0) = 2.05 \text{ s}$, which is in good agreement with previous experimental results on transient dropwise condensation [43,60].

To determine the sweeping rates and heat transfer along the plate for $y > 0$ we can discretize the plate into rows of height dL (here: $dL = 10\text{mm}$ for $L = 1 \text{ m}$). A sweeping droplet coalesces

with all smaller droplets in the area $dA = dL \ 2r_{sw}(y)$ ahead of it. Thus, the volume of the sweeping droplet increases as

$$V(y) = V(y - 1) + dV , \quad (26)$$

where

$$dV = \left(\int_{r_{min}}^{r_e} V(r)n(r)dr + \int_{r_e}^{r_s(y)} V(r)N(r)dr \right) dA . \quad (27)$$

The new radius of the sweeping droplet can be calculated using Eq. (22). Again, note the upper bound in the second integral. To calculate the maximum size to which droplets can grow before being swept, $r_s(y)$, we first need to determine the sweeping cycle time, or sweeping period, t_{cycle} . If $n_s(y)$ is the number of sweeping events within the characteristic time period t_{ch} , then we can write:

$$t_{cycle} = \frac{t_{ch}}{n_s} . \quad (28)$$

From geometric considerations, we can see that for $y > 0$:

$$n_s = 1 + 2 \frac{r_{sw}(y)}{B} . \quad (29)$$

The red dot in Figure 6 illustrates the relationship of the slot in which it is located and the interaction with droplets from neighboring slots. The first term on the right hand side of Eq. (29) stems from the sweeping droplet originating at the top of the same slot in which the red dot is located. For an imaginary position at the center of a slot (red dot), the droplets from the neighboring slots will each cause a sweeping event at the red dot when $r_{sw} = B$, and n_s becomes $n_s = 3$. Even further down the plate droplets from the slots next to the neighboring slots will also sweep the center slot, such that when $r_{sw} = 2B$, $n_s = 5$. Since we assume that r_{sw} is only a function of y (i.e. sweeping droplets in all slots behave the same), this progressive series of sweeping events gives rise to the second term on the right hand side of Eq. (29).

We can now calculate the radius to which droplets can grow in between sweeping cycles, assuming that sweeping originates at uniform time intervals at the top of the plate. Using a definition similar to Eq. (24), we can write:

$$t_{cycle}(y) = \frac{m''(y)}{q''(y)} \Delta h_{fg} = \frac{\rho_l \left(\int_{r_{min}}^{r_e} V(r)n(r)dr + \int_{r_e}^{r_s(y)} V(r)N(r)dr \right)}{\int_{r_{min}}^{r_e} q_d(r)n(r)dr + \int_{r_e}^{r_s(y)/1.3} q_d(r)N(r)dr}. \quad (30)$$

Numerically, through an iterative method, we can now solve for $r_s(y)$ and calculate the overall heat transfer as a function of plate height using the formula given in the denominator of Eq. (30).

Figure 7 shows calculated sweeping radius r_{sw} , maximum droplet radius before sweeping r_s , cycle period t_{cycle} , and normalized overall steady-state heat transfer rate as a function of plate height. As the sweeping droplets slide down the surface, they collect and absorb smaller droplets and grow in size. Simultaneously, the cycle period, and thus size to which droplets grow through direct condensation and coalescence before being swept, decreases. It is important to note that this decrease happens quickly over the first 10-20 cm of the plate and then approaches a constant of approximately $t_{cycle} \approx 0.5$ s and $r_s \approx 0.1$ mm, respectively. Similar trends have been reported before, where the sweeping period was nearly constant for large distances ($y/(2r_{max,0}) \geq 40$) away from the top of the plate [61]. Despite the strong decrease in maximum droplet size from top to bottom of the plate, the overall heat transfer rates decrease only minimally ($< 7.5\%$), due to the relative insensitivity of droplets with $r > 100$ μm on overall heat transfer rates [35,36]. At $L = 1$ m the largest droplets in the condensation distribution, r_s , are only slightly smaller than $r = 100$ μm , resulting in minimal heat transfer decrease. Indeed, it has been observed before that heat transfer rates are nearly constant over the length of a vertical plate [36,44]. While nucleation sites get cleared more frequently, sweeping droplets have a blanketing effect on significant parts of the surface such that the two contributions cancel each other out. Similar to the heat transfer

rates, the distribution of droplets sizes is hardly influenced by increased sweeping rates at the bottom of the plate. All of the above equations are independent of droplet sliding speed, and we do not expect a variation in average droplet sweeping and heat transfer rates for varying lubricant viscosities. The only difference between the various lubricant oils would be the time a sweeping droplet needs to reach the bottom of the plate, which is irrelevant for a steady-state solution and a discretization in space, not time.

Figure 7: Sweeping and heat transfer predictions for a tall vertical plate. (a) Calculated sweeping radius r_{sw} and radius to which droplets grow before being swept r_s as a function of plate height for a 100% water vapor environment at $T_{sat} = 100^\circ\text{C}$ and $\Delta T = 5\text{K}$. (b) Sweeping cycle time, or sweeping period, t_{cycle} and heat transfer rate normalized by the average heat transfer rate at the top of the plate as a function of plate height for the same conditions. For all parameters, results from numerical simulations are depicted as points and analytical fits as lines.

To facilitate estimations of droplet sizes, sweeping periods, and heat transfer rates on vertical plates, Figure 7 also includes analytical fits. Fitting to the numerical output, we can write for the radius of the sweeping droplets as a function of plate height:

$$r_{sw} = r_{max,0} + 2.85 \cdot 10^{-3} y^{0.43} , \quad (31)$$

where $y = 0$ m is the top of the vertical plate and $r_{\max,0}$ is the maximum droplet radius at the top of the plate ($[y] = \text{m}$, $[r] = \text{mm}$). The maximum droplet radius to which droplets grow before being swept can be written as:

$$r_s = 6.6 \cdot 10^{-5} y^{-0.28} . \quad (32)$$

The sweeping period can well be estimated using Eqs. (28), (29), and (31). The resulting average heat transfer rate as a function of plate height can be described by:

$$q''(y) = q_0''(1 - 0.075 y^{0.29}) , \quad (33)$$

where $q_0'' = 2.4 \cdot 10^5 \text{ W/m}^2$ is the heat transfer rate at the top of the plate for pure water vapor conditions, assuming the Rose model for the distribution of drop sizes.

4.3. Experimental Droplet Size Distribution with Artificial Sweeping

To experimentally verify the influence of sweeping on the distribution of droplet sizes we performed additional experiments with artificial sweeping where droplets were injected with a needle above the condensation area (Figure 3). Natural average sweeping periods through condensation and coalesces ranged from 9 to 24 s, as defined by the number of droplets sweeping through a specific spot in a 2 minute period, depending on the flow rate of the humid N_2 gas. Slow, medium, and fast artificial periods were 2.9, 1.8, and 0.7 s, respectively. After each sweeping event we observe re-nucleation at the same substrate locations. Figure 8 shows the droplet number densities $N(r)$ on the Krytox 1514 oil for the natural, slow, medium, and fast sweeping rates. As expected from numerical modeling presented in section 4.2, the time-averaged distribution of droplet sizes is independent of sweeping frequency. Simultaneously, we observe a strong decrease in the maximum diameter to which droplets grow before being swept. While droplets during natural sweeping at the top of a vertical plate grow to $r_s = r_{\max} = 0.7\text{-}0.8$

mm, this value decreases to $r_s = 0.5\text{-}0.6$ mm for the slow and medium sweeping rates, and to $r_s \approx 0.25$ mm in the case of fast artificial sweeping, as illustrated in Figure 8. Our data suggests that the distribution of droplet sizes on LIS is not a function of the maximum droplet size. Indeed, when fitting the Rose model for the distribution of droplet sizes (Eq. (13)) to our data (Figure 5a) by adjusting the exponent, we find the best agreement between data and model for an exponent of negative unity, i.e.

$$N(r) = \frac{1}{3\pi r^2 \hat{r}} \left(\frac{r}{\hat{r}}\right)^{-1}. \quad (34)$$

We see that the dependence of the maximum droplet size, $\hat{r} \approx r_s / 1.3$, cancels out, and we are left with

$$N(r) = \frac{1}{3\pi r^3}. \quad (35)$$

Figure 8: Steady state number density, i.e. droplet size distribution, for droplets on the Krytox 1514 oil with artificial sweeping. The time-averaged distribution of droplet sizes is independent of sweeping frequency for small droplets. The vertical dashed lines represent the size to which droplets grow before being swept.

Equation (35) is independent of the maximum droplet radius. Comparison with the power-law fit from this work reveals strong similarities between the exponent, $b = -3.05$, from Eq. (21) and the droplet size dependence in the modified Rose model (Eq. (35)). In fact, Eq. (13) was first derived as $N(r) \sim r^{m-3}$, where m was determined experimentally to $m = 1/3$ [36,37,62,63]. In the present study on LIS surfaces, we find that $m \approx 0$. The independence of the droplet distribution on the maximum droplet size before sweeping can also be explained with the self-similarity of droplet sizes at various magnification levels [36]. When small droplets nucleate, condense and coalesce, they are not yet influenced by the size to which they will eventually grow before sweeping. Thus, conceptually, we do not expect the size distribution of small droplets to differ for various sweeping periods and maximum droplet sizes.

4.4. Overall Surface Condensation Heat Transfer

Average heat transfer rates for the present condensation experiments with non-condensable gases can be estimated from a time-evolution of the distribution of droplet sizes. Figure 9a shows the evolution of condensing and coalescing droplets on a Krytox 1514 oil within one sweeping cycle (here: $t_{\text{cycle}} = 20$ s). Discretizing the cycle period into 63 time steps, we estimate the heat transfer rate averaged over the sweeping period to be $q''_{\text{exp}} = 5.22 \cdot 10^4$ W/m². The estimate was determined by balancing the mass of added condensate from image analysis with the energy released due to vapor-liquid phase change. Theoretically, using Eqs. (24) and (25) along with Eq. (21) and the experimentally observed cycle time, the average heat transfer would be $q'' = 4.3 \cdot 10^4$ W/m². Despite the fact that the subcooling temperature in the present experiments is unknown, the experimentally determined value is in excellent agreement with the theoretically predicted heat transfer rate, giving us confidence in the validity of the theoretical model presented in section 4.2.

Figure 9: Time evolution of droplet condensation and comparison of heat transfer rates. (a) Heat transfer rates can be estimated from the time evolution of droplet condensation. Each image was obtained with a time step of 2.5 s. The sample surface was a Krytox 1514 lubricant oil, and images were obtained with a 5x microscope lens. (b,c) Comparison of heat transfer rates for filmwise and dropwise condensation with droplet size distributions following the Rose model (Eq. (13)) and the distribution for LIS, as presented here (Eq. (21)). (b) Influence of the coating thickness h_B on the relative heat transfer rates for a solid fraction $\phi = 0.1$. (c) 3-D color plot showing the relationship of coating thickness, microstructure solid fraction and heat transfer rate. The color represents the ratio of dropwise condensation on LIS (Eq. (21)) to filmwise condensation on bare substrates with a plate height of $y = 10$ cm and $\Delta T_{\text{tot}} = 5\text{K}$ (Eq. (36)). The white star marks the parameters of the present study.

To elucidate the effects of the newly identified droplet distribution results on LIS, or SLIPS, to overall surface heat transfer, we combined the experimental droplet distribution results with individual droplet heat transfer to predict an overall LIS heat transfer performance. Using Eq. (11) we can now compare the heat transfer rates following the distribution of droplet sizes for LIS, as derived in the present work (Eq. (21)), and with those following the Rose model (Eq. (13)). The black curve in Figure 9b shows the comparison as a function of coating thickness. Due to the underestimation of small droplet sizes in the Rose model, with the new distribution of droplet sizes on LIS we expect heat transfer rates to almost double compared to previous predictions.

In relation to filmwise condensation on a bare substrate, the promotion of dropwise condensation on LIS adds a thermal resistance between the substrate and vapor. Following Nusselt's theory for laminar filmwise condensation on a bare vertical substrate we can write [64]:

$$q''_{film} = \left(\frac{\Delta h_{fg} g (\rho_l - \rho_v) k_l^3}{4y \Delta T_{tot} \nu_l} \right)^{1/4} \Delta T_{tot}, \quad (36)$$

where ρ_v is the vapor density and ν_l is the liquid kinematic viscosity. To determine the effectiveness of lubricant-infused surfaces on overall heat transfer rates, we compare the heat transfer rates during dropwise condensation with those for filmwise condensation as a function of coating thickness h_B and solid fraction ϕ (Figure 9c). For a solid fraction of $\phi \approx 0.1$, LIS enhance heat transfer rates for coating thicknesses up to $h_B < 80 \mu\text{m}$. Especially for very thin coatings, such as boehmite ($h_B \approx 0.2\text{-}2 \mu\text{m}$), [65] we expect heat transfer enhancements on the order of 10-15 compared to traditional filmwise condensation. Within the realm of possible solid fractions ($\phi \approx 0.1\text{-}0.7$) their influence on heat transfer enhancement is minimal. The most important design parameter for the microstructured surface is the coating (oil) thickness, as it

greatly affects the surface thermal resistance. From a thermal perspective, the thickness of the porous surface structures should be minimized. Although structure length scale minimization might reduce possible self-healing effects of LIS surfaces [34], it has been shown to be the most beneficial in terms of retaining the lubricant due to larger capillary pressures generated by nanoscale pores when compared to microscale ones [66]. The work presented here provides an analytical and experimental framework to guide the design of SLIPS and LIS surfaces for dropwise condensation heat transfer applications.

5. Conclusions

We presented experimental and numerical results on the distribution of droplet sizes and the influence of sweeping on heat transfer rates for water dropwise condensation on SLIPS and LIS surfaces. Using statistical averaging of droplet densities to determine steady-state droplet size distributions and analytical-numerical modeling, we found that:

- 1) The average distribution of droplet sizes is independent of lubricant viscosity and follows the Rose model well, but underestimates the number of small droplets ($r < 10 \mu\text{m}$). With Eq. (21) we propose an alternative fit for $N(r)$ that accounts for a larger number of observed droplets at small sizes ($r < 10 \mu\text{m}$): $N(r) = 6 \times 10^7 r^{-3.05} \text{ mm}^{-3}$. Overall heat transfer rates can then be estimated using Eqs. (10), (11), and (14).
- 2) Surfaces with the low-viscosity oils ($\nu = 12 \text{ cSt}$) degraded quickly *via* lubricant drainage, whereas those surfaces with higher viscosity ($\nu > 140 \text{ cSt}$) remained stable for > 10 hours of operation. We thus recommend using oils with $\nu > 100\text{-}200 \text{ cSt}$ for water condensation on LIS.

- 3) The experimentally determined droplet sizes for the onset of sliding are almost twice as large as theoretically predicted ($r_{\max,ex} = 750 \mu\text{m}$ vs. $r_{\max,th} = 460 \mu\text{m}$). We propose that edge effects at the meniscus of the three-phase contact line between the oil, water, and air increase the droplet sliding resistance.
- 4) Artificial sweeping through droplet injection can represent increased sweeping rates on large vertical plates. The maximum size to which droplets grow before being swept decreases rapidly with only a modest decrease in sweeping period. While $r_s = r_{\max} \approx 750 \mu\text{m}$ with natural sweeping ($t_{\text{cycle}} = 9\text{-}24 \text{ s}$), $r_s \approx 62 \mu\text{m}$ for slow and medium artificial sweeping rates ($t_{\text{cycle}} \approx 1\text{-}2 \text{ s}$).
- 5) While the maximum size to which droplets grow *via* condensation and coalescence decreases with increasing sweeping rates and distance from the top of a vertical plate, the distribution of droplet sizes and heat transfer rates remain fairly constant throughout the height of a vertical plate. Due to a large conduction resistance through the liquid, only droplets with $r < 100 \mu\text{m}$ actively participate and influence the heat transfer process.
- 6) The most important design parameter for LIS is the coating thickness. Thicknesses of $\approx 0.5\text{-}5 \mu\text{m}$ can increase heat transfer rates up to 10-15 times compared to traditional filmwise condensation.
- 7) For the evaluation of heat transfer rates we assumed in this work that droplets nucleate and grow on top of the lubricant layer. However, recent studies have shown that droplets can nucleate within the lubricant layer and move upwards towards the lubricant-vapor interface as the droplets grow [38]. In this case we would expect an additional thermal resistance due to conduction and diffusion through the lubricant layer for small droplets ($r \sim h_B$). Since in the dropwise condensation model discussed in this work small droplets

contribute most to the overall heat transfer, actual heat transfer rates might be significantly lower than predicated due to this added resistance. Further experiments would need to verify and quantify the extent to which the location of droplet nucleation influences heat transfer rates.

Overall, this work presents new insights into the characteristics of dropwise condensation on lubricant infused surfaces and provides a framework for analytical modeling and estimation of heat transfer on large vertical plates.

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