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UNDERSTANDING ENHANCED BOILING WITH TRITON X SURFACTANTS

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Heat transfer performance in pool boiling is largely dictated by bubble growth, departure, and number of nucleation sites. It is a well known phenomenon that adding surfactants can lower the liquid-vapor surface tension and increase the bubble departure frequency, thereby enhancing heat transfer. In addition to faster departure rates, surfactants are observed to dramatically increase the number of nucleation sites, which cannot be explained by simple surface tension arguments. Furthermore, it is not well understood which surfactant properties such as chemical composition and molecular structure affect boiling most significantly. From our experiments using Triton X-100 and Triton X-114 nonionic surfactants, we attribute boiling enhancement mainly to adsorption to the solid-liquid interface. Using the Mikic-Rohsenow model for boiling, a simple linear adsorption model, and the Cassie-Baxter description for contact angle, we developed a model that shows agreement with experimental results. This work offers some insights on how to predict boiling enhancement based on surfactant chemistry alone, which may aid in choosing optimal surfactants for boiling in the future.

NOMENCLATURE

a_s Projected molecular area of a single surfactant
 A Area of copper block
 c_p Liquid specific heat
 C_1 Surfactant monomer concentration
 $C_{1,bulk}$ Bulk monomer concentration
 $C_{1,s}$ Subsurface ($z = 0$) monomer concentration

D_1 Monomer diffusion coefficient
 D_b Diameter of bubble
 f_b Frequency of bubble departure
 f_s Area fraction of surface covered by surfactants
 g Gravitational acceleration
 h Heat transfer coefficient
 h_{fg} Enthalpy of vaporization
 k Thermal conductivity of copper block
 k_l Thermal conductivity of liquid
 L Length of copper block
 m Power-law exponent
 n Number of nucleation sites per unit area
 n_{EO} Number of EO groups
 N_A Avogadro constant
 P Perimeter of copper block
 q'' Heat flux
 q_{avg} Average heat flux per nucleation site
 R Molar gas constant
 R_s Constant in Mikic-Rohsenow nucleation model
 t Time variable
 T_{sat} Saturation temperature
 T_∞ Ambient temperature
 z Spatial dimension along the length of the copper block
 z Spatial dimension normal to liquid-vapor interface
 β Constant in power-law fit
 Γ_{lv} Surfactant concentration at liquid-vapor interface
 Γ_{sl} Surfactant concentration at solid-liquid interface
 γ_v Liquid-vapor surface tension
 $\gamma_{v,0}$ Liquid-vapor surface tension of pure water

- ΔT Difference between surface temperature and saturation temperature
 η Monomer adsorption tendency
 θ Contact angle with no surfactants at liquid-vapor interface
 θ_p Contact angle on flat boiling surface with no surfactants at liquid-vapor interface
 θ_s Contact angle on surfactant phase with no surfactants at liquid-vapor interface
 ρ_l Liquid density
 ρ_v Vapor density
 ϕ Defect cone angle
 Ω Geometric nucleation parameter

INTRODUCTION

Boiling is an important heat transfer mechanism used in a wide range of industrial applications such as power generation, chemical production, refrigeration, and electronics cooling [1]. Significant interest around boiling or any phase change heat transfer mechanism is due to the large amount of heat that can be transferred across a very small temperature difference. For boiling, when a hot surface in contact with the liquid, vapor bubbles nucleate on the solid-liquid interface at cavities. As the bubbles grow larger, buoyancy forces overcome surface tension forces and bubbles depart from the surface allowing a new bubble to nucleate at the same spot. Thus, increasing roughness to increase nucleation sites [2] and/or using hydrophilic surfaces to reduce surface tension forces are common methods to promote higher heat transfer rates [3]. Adding surfactants and other polymeric additives to the liquid is also known to enhance heat transfer [4]. Wasekar suggested that boiling enhancement is highly correlated to dynamic surface tension and showed that enhancement only occurs below the critical micelle concentration (CMC) [1], which is the concentration at which surfactant monomers aggregate into micelles. To try to model the enhancement, Sher et al. incorporated the Rohsenow correlation to describe complex enhancement behavior [5] and Wen et al. incorporated a wetting correlation with the Mikic-Rohsenow model [6]. However, a comprehensive description of the mechanism for boiling enhancement is lacking, nor is there a clear relationship between the enhancement and the properties of surfactants such as molecular weight, structure, and composition.

THEORY

Surfactant solutions below the CMC are monomeric while above the CMC, surfactants aggregate into micelles. The CMC is typically a very small concentration on the order of a few mM; therefore, below the CMC many bulk properties such as viscosity, thermal conductivity, specific heat, and saturation temperature are virtually unaffected [4, 7–9]. For instance, boiling point elevation was calculated to be 2×10^{-4} K for Triton

X-100 at the CMC using the Clausius-Clapeyron relation and Raoult's Law. On the other hand, surface tension is significantly reduced due to the tendency of surfactants to adsorb at interfaces. Since the concentrations studied in this paper will be submicellar ($C_1 < C_{CMC}$), all fluid properties except for surface tension, γ_{lv} , are assumed to be invariant with surfactant concentration.

Dynamic Surface Tension

When a liquid-vapor interface is created during bubble formation in a submicellar solution, surfactants adsorb at the interface by diffusion. Assuming a diffusion controlled process, the following governing equation for monomer concentration can be used:

$$\frac{\partial C_1}{\partial t} = D_1 \frac{\partial^2 C_1}{\partial z^2} \quad (1)$$

Using conservation of mass, the following boundary condition is applied:

$$\frac{\partial \Gamma_{lv}}{\partial t} = D_1 \left(\frac{\partial C_1}{\partial z} \right)_{z=0} \quad (2)$$

where Γ_{lv} is the monomer surface concentration (adsorption). Solving (1) leads to the well-known Ward-Tordai solution:

$$\Gamma_{lv} = 2C_{1,bulk} \sqrt{\frac{D_1 t}{\pi}} - 2 \sqrt{\frac{D_1}{\pi}} \int_0^{\sqrt{t}} C_{1,s}(t - \tau) d\sqrt{\tau} \quad (3)$$

where $C_{1,bulk}$ is the bulk concentration ($x \rightarrow \infty$) and $C_{1,s}$ is the subsurface concentration ($x = 0$). Assuming that the subsurface concentration is zero, (3) can be simplified to:

$$\Gamma_{lv} = 2C_{1,bulk} \sqrt{\frac{D_1 t}{\pi}} \quad (4)$$

While the simplification is commonly reported in the literature, it should be noted that it is a nonphysical solution since it predicts an infinite flux at $t = 0$ [10]. However, Moorkanikkara and Blankschtein have shown that using a more accurate mixed energy barrier-diffusion control model also leads to an apparent $t^{1/2}$ dependence [10]. Thus, for simplicity, the asymptotic limit of the simple diffusion model (4) is used. Using an ideal gas type surface equation of state $\Pi = \gamma_{v,0} - \gamma_{lv} = \Gamma_{lv} RT$, the dynamic surface tension in the short-time limit is given by:

$$\gamma_{lv} = \gamma_{v,0} - 2RTC_{1,bulk} \sqrt{\frac{D_1 t}{\pi}} \quad (5)$$

The validity of the $t^{1/2}$ dependence of dynamic surface tension has been demonstrated with a variety of surfactants, including Triton X-100 [11] for timescales less than 50 ms. Using data from Liu et al. [11], we calculated that a value of $8 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$ is appropriate for the diffusion coefficient, D_1 . Data from Fainerman et al. shows that the diffusion coefficient should be similar for Triton X-100 and Triton X-114 [12]; therefore, the same value for the diffusion coefficient is used in this study. A key aspect of dynamic surface tension is that having a high diffusion coefficient is the most important parameter in lowering surface tension, and thereby enhancing boiling.

Boiling

The behavior of boiling is difficult to analyze from a very fundamental point of view; thus, previous studies in the past have focused on developing correlations. The Jakob & Fritz model [13] is a simple correlation for bubble departure frequency, f_b , which is appropriate for pressures close to atmospheric pressure:

$$f_b = (0.078 \text{ m s}^{-1}) \frac{1}{D_b} \quad (6)$$

To determine the bubble departure diameter, D_b , as a function of surface tension, Cole & Rohsenow's correlation [14] can be used:

$$D_b = 1.5 \times 10^{-4} \sqrt{\frac{\gamma_v}{g(\rho_l - \rho_v)}} \left(\frac{\rho_l c_p T_{\text{sat}}}{\rho_v h_{\text{fg}}} \right)^{5/4} \quad (7)$$

Using properties of water at 100°C , the average bubble lifetime ($1/f_b$) is approximately 30 ms.

According to the Mikic-Rohsenow description of boiling [15, 16], the heat flux due to boiling (neglecting convective heat transfer) is given by:

$$q'' = \pi D_b^2 n q_{\text{avg}} \quad (8)$$

$$n = \left(\frac{R_s \Omega(\theta, \phi) h_{\text{fg}} \rho_v}{2 T_{\text{sat}} \gamma_v} \right)^m \Delta T^m \quad (9)$$

$$q_{\text{avg}} = \frac{2k_1 \Delta T}{\sqrt{\pi \frac{k_1}{\rho c_{p,l}}}} \sqrt{f_b} \quad (10)$$

where n is the number of active nucleation sites, and q_{avg} is the time-averaged heat transfer during the formation of one vapor

bubble. The constant m is empirically determined to be 6 for most surface/liquid combinations and R_s is used as a fitting parameter that is related to roughness and cavity size. R_s should vary from surface to surface ($R_s = 3.11 \times 10^{-5} \text{ m}$ for Triton X-114 experiments and $R_s = 2.92 \times 10^{-5} \text{ m}$ for Triton X-100 experiments). The liquid-vapor surface tension, γ_v , is the dynamic surface tension, which is a function of concentration and bubble lifetime as described in the previous section. However, the calculated reduction in surface tension for typical bubble lifetimes is relatively small (0-3 mN/m) and cannot account for the observed enhanced in heat transfer. Instead, the number of nucleation sites must reflect the presence of surfactants. In particular, the Ω term in (9) is a function of the initial contact angle, θ , and cone angle, ϕ . The cone angle is taken to be 29° , which is a reasonable value considering measurements taken by Yang and Kim [17]. Ω arises out of pure geometric arguments on how a vapor bubble grows out of a conic cavity. It is a complicated expression and can be found in Lorenz' thesis [18]. Therefore, the only term in (9) that would be modified by the presence of surfactants is the initial contact angle, θ . Note that this is the contact angle of pure water on a completely flat surface since the initial angle assumes no surfactants have yet adsorbed at the liquid-vapor interface.

Adsorption

Assuming the surfactant adsorption forms a heterogeneous surface, the initial contact angle, θ , on a purely flat surface can be described by the Cassie-Baxter equation [19]

$$\cos(\theta) = (1 - f_s) \cos(\theta_p) + f_s \cos(\theta_s) \quad (11)$$

where f_s is the area fraction of the surface covered by surfactants, θ_p is the contact angle with no surfactants adsorbed, and θ_s is the contact angle over pure surfactant. From measurements taken from experimental imaging of boiling on a heated surface, θ_p was determined to be 40° . Due to the difficulty in measuring θ_s , we use this as a fitting parameter. The area fraction, f_s , is related to the surface concentration, Γ_{sl} , and projected monomer area, a_s , as follows:

$$f_s = \Gamma_{\text{sl}} N_A a_s \quad (12)$$

where N_A is the Avogadro constant. According to data from Partyka et al. [20], a_s is approximately 33 \AA^2 and 38 \AA^2 for Triton X-114 and Triton X-100, respectively. Typically, the Langmuir isotherm is used to model surfactant adsorption; however, data from Partyka et al. and Soboleva et al. show that the adsorption behavior is approximately linear with concentration up to the CMC [20, 21]. Accordingly, Henry's law

$$\Gamma_{\text{sl}} = \eta C_1 \quad (13)$$

can be used to determine the surface concentration, Γ_{sl} , where η represents the tendency of a surfactant to adsorb at the solid surface. From data by Partyka et al., η values of 1.7×10^{-5} m and 1.4×10^{-5} m were calculated for Triton X-114 and Triton X-100, respectively. For a surfactant with a lower CMC, such as Triton X-114, we expect that the slope should increase since surface saturation should occur earlier. In addition, Triton X-114, due to having a smaller footprint of 33 \AA^2 , reaches a higher saturation concentration which further increases the value of η . Combining (13), (12), and (11) into the Ω term of (9) then gives the dependency of the surfactant adsorption on nucleation behavior.

EXPERIMENTAL

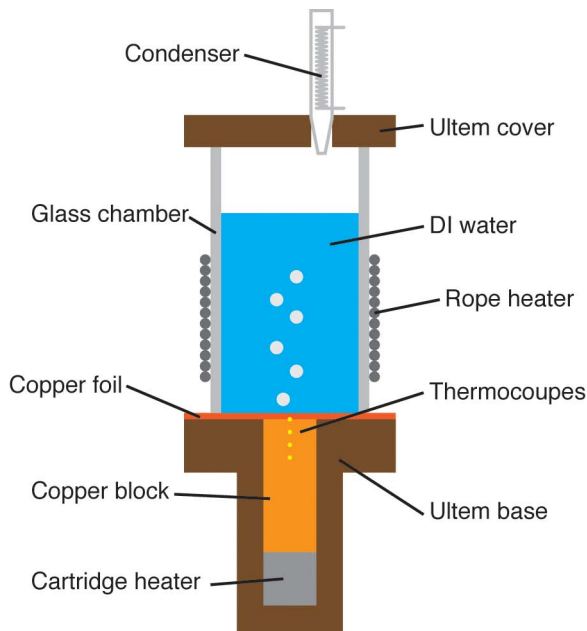


FIGURE 1. SCHEMATIC OF THE POOL BOILING TEST RIG. HEAT IS PROVIDED BY A CARTRIDGE HEATER LOCATED BELOW A COPPER BLOCK, WHILE A ROPE HEATER MINIMIZES HEAT LOSS AND MAINTAINS SATURATION CONDITIONS. FOUR EQUALLY-SPACED THERMOCOUPLES ARE USED TO DETERMINE THE HEAT FLUX.

We conducted pool boiling experiments with DI water where we measured surface temperature and heat flux. By adding surfactants to DI water, we were able to quantify the enhancement in heat transfer. Two nonionic surfactants, Triton X-114 and Triton X-100 (Sigma-Aldrich), were used in this study. These surfactants both share the same hydrophobic octylphenyl group with differing number of ethoxylate (EO) groups. They are both

described by the chemical formula $C_{14}H_{22}O(C_2H_4O)_{n_{EO}}$, where n_{EO} is the number of EO groups (see Table 1). Longer EO chains cause the surfactant to be more hydrophilic, resulting in reduced surfactant affinity and a higher CMC. As a result, the CMC for the longer EO chained Triton X-100 is higher.

Solutions of Triton X-114 and Triton X-100 were prepared in concentrations of 1.38 % and 2.17 % v/v, respectively, in deionized (DI) water. A custom boiling rig was filled with 400 mL of DI water (see Fig. 1). 100 μ L and 250 μ L syringes were used to apply a small amount of these prepared solutions into the chamber of the boiling rig. A programmable DC power supply (KLP, Kepco) was used to power a cartridge heater in the boiling rig. The power supply was controlled with LabVIEW (National Instruments) and power was ramped up linearly from 30 W to 100 W at a rate of 2.33 W min^{-1} . A rope heater wrapped around the glass chamber helped maintain bulk water temperature at T_{sat} . A condenser fitted on top of the rig ensured that water did not evaporated away.

TABLE 1. SURFACTANT PROPERTIES

Surfactant	Avg. No. of EO Groups	CMC at 25 °C
TX-114	7.5	0.22 mM
TX-100	9.5	0.30 mM

A copper block with constant cross-sectional area of 4 cm^2 and four thermocouples spaced 8 mm apart were used to determine the heat flux. Since there are heat losses, the following fin equation was solved and used to fit the data to determine the heat flux at the surface. The fin equation is valid in this case since the Biot number is calculated to be 0.003:

$$0 = \frac{\partial^2 T(x)}{\partial x^2} - \frac{hP}{kA} (T(x) - T_{\infty}) \quad (14)$$

The boundary conditions used were $\left(\frac{\partial T}{\partial x}\right)_{x=L} = \frac{q_L''}{k}$ and $T(x=0) = T_{surface}$. An arbitrary value of L was applied while q_L'' and h were used as fitting parameters.

A 50 μ m copper foil serving as the boiling surface was soldered to the top of the copper block. Experiments were run within 12 hours after soldering so as to minimize the effects of oxidation. A CCD camera (Pixelink) aided by fiberoptic light was used to capture video of boiling at 120 fps.

RESULTS & DISCUSSION

Video snapshots of nucleate pool boiling of Triton X-114 solutions show that nucleation sites increased with increasing surfactant concentration below the CMC, which is approximately 0.2 mM (Fig. 2). The surface temperature also decreased with increasing surfactant concentration. Above the CMC, the solution reached the cloud point and surface temperature increased to approximately 112 °C which is higher than that of pure water (110 °C).

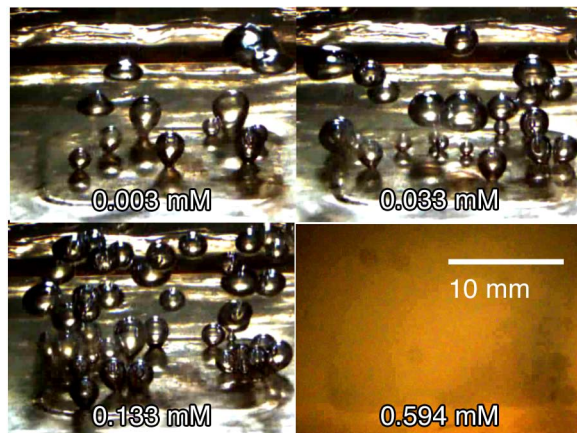


FIGURE 2. VIDEO SNAPSHOTS OF NUCLEATE POOL BOILING AT VARIOUS CONCENTRATIONS OF TRITON X-114 ($C_{CMC} \approx 0.2$ mM). IN ALL SNAPSHOTS, $q'' \approx 2.5$ W cm⁻².

From these images, it is apparent that boiling enhancement is mainly due to activation of new nucleation sites. Increased nucleation, however, cannot be explained by liquid-vapor surface tension arguments. At the start of a nucleation event, the lifetime of the liquid-vapor of surface tension is infinitesimally small, and according to the surfactant diffusion description in (4), no surfactants should be adsorbed at the interface. Accordingly, the frequency of bubble departure and bubble size should not be a significant mode of enhancement. The Mikic-Rohsenow description of nucleation sites in (9) has a surface tension dependency, but the small change in surface tension at such short timescales predicts relatively little change in the number of nucleation sites. To illustrate this discrepancy, we used the Mikic-Rohsenow model where the initial contact angle is equal to the plain surface contact angle ($q''(\theta = \theta_p)$ and Ω becomes invariant with concentration), and determined heat flux by changing γ_v alone using (5). We then fitted a general power-law to the boiling curves:

$$q''_{PL} = \beta \Delta T^{m+1} \quad (15)$$

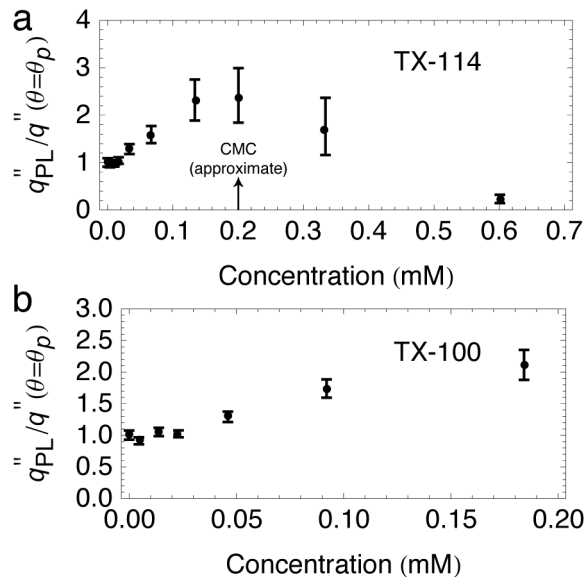


FIGURE 3. NUCLEATION SITE ENHANCEMENT FOR (a) TRITON X-114 AND (b) TRITON X-100, CALCULATED BY THE RATIO OF A POWER-LAW FIT OVER THE CONVENTIONAL MIKIC-ROHSENOW MODEL. THE NUMBER OF NUCLEATION SITES INCREASED LINEARLY WITH CONCENTRATION UP TO THE CMC.

By plotting the ratio $q''_{PL}/q''(\theta = \theta_p)$ as shown in Fig. 3, we show the discrepancy between the actual enhancement and that obtained using the Mikic-Rohsenow ignoring adsorption. We hypothesize that this discrepancy is due to adsorption of surfactant on the solid surface which would affect the initial contact angle, which in turn would affect the nucleation behavior. The shape of $q''_{PL}/q''(\theta = \theta_p)$ should be directly related to the adsorption behavior. The enhancement is approximately linear up to the CMC (0.2 mM for Triton X-114 and 0.3 mM for Triton X-100). As shown in Fig. 3a, the enhancement peaks near the CMC and boiling degrades beyond the CMC. The linear behavior is notable, since combining the adsorption behavior (11), (12), and (13) into the Ω term in (9) also results in approximately linear enhancement up to the CMC.

By taking into account adsorption and calculating the change in contact angle and nucleation using (11), (12), (13), and (9), we plotted the Mikic-Rohsenow model (8) and compared them to the boiling data. Obtaining the contact angle of the pure surfactant phase, θ_s , experimentally would be difficult as it requires measurement of the contact angle at the instantaneous moment of bubble nucleation. Therefore, θ_s is used as a fitting parameter. θ_s values of 47° and 45° were found suitable for both Triton X-114 and Triton X-100, respectively. The fact that $\theta_s > \theta_p$ is likely due to the existence of a hydrophobic (octylphenyl) component in the surfactant. The difference in

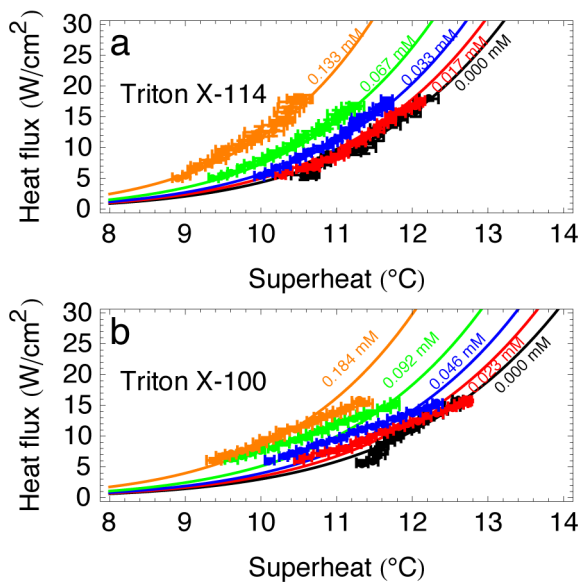


FIGURE 4. BOILING CURVES FOR (a) TRITON X-114 AND (b) TRITON X-100 WITH MIKIC-ROHSENOW MODEL TAKING INTO ACCOUNT SURFACTANT ADSORPTION.

these values of θ_s is a reflection of the more hydrophilic character (hence lower contact angle) of Triton X-100 compared to Triton X-114. In terms of the effect that modifying the θ_s parameter has on boiling, θ_s determines the degree of enhancement, where larger values lead to more enhancement due to easier nucleation on a hydrophobic surface. While fitting has been used, the relative differences in enhancements for a given surfactant at different concentrations is strictly dictated by the adsorption and nucleation theory described in this paper (fitting is performed on a surfactant-by-surfactant basis and not on an experiment-by-experiment basis). Since the trend in enhancement seems to be consistent with our model, we believe that this is evidence of the important role that adsorption plays in nucleation behavior.

As further validation, the contact angle close to bubble departure, θ , only changed a few degrees from low to high concentration, and this is consistent with what was observed in the boiling videos. This suggests that the liquid-vapor surface tension is not significantly reduced as predicted by dynamic surface tension theory. The model fits Triton X-114 better than Triton X-100 due to Triton X-100 exhibiting non-power-law behavior. While the exact reason for the non-power-law behavior is unknown, we speculate that differences in oxidation during soldering or differences in surface topology may have resulted in different nucleation cavity distributions. Since the cavity distribution is a key aspect of the Mikic-Rohsenow model, the power-law behavior may have been altered. Whether the fact that the non-power-law behavior during Triton X-100 experiments is due to an issue with the particular boiling surface or whether there

are other complicated dynamics would not invalidate our adsorption based model. Our model only predicts a multiplicative enhancement in the boiling curve, not a change in the power-law behavior. Thus, the inability to fit a power-law perfectly is a shortcoming of the empirical boiling correlations used and not of the adsorption based theory. However, further experiments with more surfactants, as well as experiments to better describe adsorption, are needed to completely validate our description of nucleation enhancement.

CONCLUSION

There are two modes of enhancement in surfactant nucleate boiling: faster bubble departure due to lower surface tension and increased nucleation. The latter significantly accounts for most of the observed enhancement. In regards to lowering of liquid-vapor surface tension, existing theory on dynamic surface tension suggests that having a high coefficient of diffusion is key in lowering surface tension and thereby reducing the force that must be overcome during bubble departure. Therefore, low molecular weight surfactants should be favored. With regard to nucleation, we believe the activation of new sites is mainly dictated by surfactant adsorption at the solid surface. At the start of a nucleation event, we do not consider surfactants at the liquid-vapor interface since this occurs in the extremely short-time limit. According to our description, adsorbed surfactants have an intrinsic contact angle higher than that of the plain boiling surface, thereby raising the contact angle according to the Cassie-Baxter equation. Using existing nucleation theory, we have shown that a higher initial contact angle should enhance the nucleation behavior significantly and the results from our model have shown agreement with boiling results. A key parameter in our model that strongly affects enhancement is the intrinsic contact angle of the surfactant phase, θ_s , where a higher value predicts greater enhancement. This parameter reflects the overall hydrophobicity of the surfactant. Surfactants with a more hydrophobic structure may be preferable for boiling. In the future, it is desirable to have a description of the relationship between concentration and adsorption based on fundamental molecular interactions and thermodynamics. Accordingly, nucleation behavior, and ultimately boiling enhancement, can be quantified in terms of molecular characteristics. The results of this work may allow for better understanding on how to use surfactants to improve performance in industrial boiling applications.

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