Boiling heat transfer on superhydrophilic, superhydrophobic, and superbiphilic surfaces

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A B S T R A C T

With recent advances in micro- and nanofabrication, superhydrophilic and superhydrophobic surfaces have been developed. The statics and dynamics of fluids on these surfaces have been well characterized. However, few investigations have been made into the potential of these surfaces to control and enhance other transport phenomena. In this article, we characterize pool boiling on surfaces with wettabilities varied from superhydrophobic to superhydrophilic, and provide nucleation measurements. The most interesting result of our measurements is that the largest heat transfer coefficients are reached not on surfaces with spatially uniform wettability, but on *biphilic* surfaces, which juxtapose hydrophilic and hydrophobic regions. We develop an analytical model that describes how biphilic surfaces effectively manage the vapor and liquid transport, delaying critical heat flux and maximizing the heat transfer coefficient. Finally, we manufacture and test the first *superbiphilic* surfaces (juxtaposing superhydrophobic and superhydrophilic regions), which show exceptional performance in pool boiling, combining high critical heat fluxes over 100 W/cm^2 with very high heat transfer coefficients, over 100 kW/m^{2}K.

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1. Introduction

A surface is called superhydrophilic (SHPi) if the apparent contact angle of water on the surface in air is close to zero, which induces spontaneous spreading. The high affinity of SHPi surfaces for water enhances capillary water transport [1,2], prevents dropwise condensation or fogging [3,4], and facilitates boiling [5]. A surface is called superhydrophobic (SHPo) if the apparent contact angle of water on the surface in air is larger than 150°. These surfaces are inspired by natural structures such as the lotus leaf and have a wealth of technical applications. SHPo surfaces self-clean [6], enhance condensation [7], mitigate frost buildup [8,9], and reduce hydrodynamic drag [1,10]. Typically, fabrication of SHPi or SHPo surfaces requires engineering a water-attracting or repelling surface to have a severe roughness on the sub-millimeter scale, which increases or decreases the true contact area with water, respectively. In recent reviews, enhanced liquid–vapor phase change was also described as a potential application of SHPi [11] and SHPo [12] surfaces. This potential enhancement is important for convective heat transfer technologies since convection associated with multiphase flow delivers the highest heat transfer coefficients,

typically one order of magnitude higher than single-phase forced convection, and two orders of magnitude higher than single-phase natural convection [13]. Few multiphase heat transfer measurements however have been made on SHPi or SHPo surfaces, with the exception of experiments involving single nucleation [14] or condensation [15].

The high heat transfer rates delivered by boiling are needed in industrial applications such as thermal generation of electricity, metallurgy, electronics cooling, and food processing. While flow boiling describes the boiling of liquids forced to move along hot solid surfaces, pool boiling, the mode studied here, describes a fluid heated on a hot surface and transported by buoyancy [16]. Two parameters measure the pool boiling performance. First, the heat transfer coefficient (HTC) is the ratio of the heat flux (q'') to the difference between the surface temperature and the boiling temperature of the fluid (ΔT), that is HTC = $q''/\Delta T$. The HTC describes the thermodynamic efficiency of the boiling exchange. Second, the critical heat flux (CHF) is the highest heat flux that a surface can exchange with a boiling fluid before the individual bubbles merge into a vapor layer that insulates the surface from the liquid. In the regime with $q'' <$ CHF, HTC typically increases with q'', as the solid surface interacts with an increased number of liquid and vapor pockets, maximizing the opportunity to transfer heat and mass across the liquid wedges of the multiple wetting lines. At CHF, the HTC is drastically reduced, which induces a significant and

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Nomenclature

A_{in}	area of influence (m^2)
c_p	specific heat (J/kg K)
CHF	critical heat flux (W/m^2)
d	diameter (m)
f	frequency (Hz)
g	acceleration due to gravity (m/s^2)
k	thermal conductivity (W/mK)
HTC	heat transfer coefficient (W/m^2K)
n'_a	active nucleation site density (sites/m^2)
p	pitch (m)
q''	heat flux (W/m^2)
SBPi	superbiphilic
SHPi	superhydrophilic
SHPo	superhydrophobic

T	temperature (°C or K)
ΔT	$T-T_{sat}$, superheat (K)

Greek symbols
γ	surface tension (N/m)
θ	wetting angle (°)
ρ	density (kg/m^3)

Subscripts
con	contact
d	departure
l	liquid
sat	saturation
v	vapor

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often destructive surface temperature increase [17–19] called dry-out.

To date, two main strategies have been used to enhance the performance of surfaces for pool boiling. The first strategy enhances the performance at low heat fluxes, in the isolated bubble regime, by promoting nucleation and enhancing HTC [16]. This is made by either reducing the surface wettability [14,20–26] or by modifying the surface topology, via e.g. surface roughening, etching of cavities [20,27,28], or microporous coatings [29,30]. The second strategy enhances the performance at high heat fluxes, in the regime of slugs and columns [16], which results in an enhanced CHF. This is made by improving liquid transport, typically by increasing surface wettability [31,32], which also sharpens wetting angles and steepens thermal gradients [31]. Wettability can be enhanced by increasing the roughness of a hydrophilic surface [33] at the sub-millimeter scale. Note that some micro- and nanostructuring processes used to increase wettability come with the benefit of randomly distributed microcavities and defects [5,17,30,34], which also facilitate nucleation.

In this study, we fabricate surfaces with engineered wettability as shown in Fig. 1. We measure for the first time the density of active nucleation sites on SHPi and SHPo surfaces, an important input parameter needed for numerical simulations of boiling on such surfaces [35]. Our modeling and pool boiling measurements also show how the wettability of a surface, as well as the juxtaposition of regions of different wettabilities, control and enhance boiling heat transfer.

2. Materials and methods

2.1. Design and manufacturing of enhanced surfaces for pool boiling

2.1.1. Surface design and fabrication

Six types of surfaces are fabricated for this study, as shown in Fig. 1. The four types in the top row have spatially uniform wettabilities (hydrophilic, hydrophobic, SHPi, SHPo). The two types shown in the bottom row of Fig. 1 juxtapose hydrophilic and hydrophobic regions: this design induces a concurrent affinity for water and for water vapor, a *quality that we name biphilic*. Nature has examples of biphilic surfaces that enhance multiphase heat transfer. The biphilic wings of the Namib desert beetle optimize its water intake [7]; while hydrophilic regions of the wing help condensation, the hydrophobic regions guide the liquid to its mouth. Few biphilic surfaces have been fabricated [25,28,36], but they all have been shown to significantly enhance boiling heat transfer. In 1965, the first biphilic surface by Hummel [25], who

sprayed hydrophobic polymer drops onto a steel surface, showed a HTC 2–7 times higher than the bare steel surface. Biphilic surfaces were recently fabricated using microlithographic techniques [21,28,36]. The microfabricated biphilic surfaces by Betz et al. [28], shown on the bottom left of Fig. 1, exhibited not only HTCs 100% larger but also a CHF 65% larger than hydrophilic surfaces.

Pushing the *biphilic* concept to more extreme values of wettability, we have also manufactured superbiphilic (SBPi) surfaces. SBPi surfaces juxtapose SHPo and SHPi areas, as shown in the bottom right of Fig. 1. These surfaces were manufactured on silicon wafers using a combination of random nanostructuring processes, microlithography, and thin hydrophobic polymer coating, as follows. The random nanostructures are made by a DRIE using the black silicon method [37]. Next, the entire surface is exposed to oxygen plasma in a RIE machine for 30 min to create a 30 nm silicon dioxide layer, rendering the surface SHPi. To obtain a microscale pattern of SHPo areas on the SHPi field, a photolithographic process was employed on the SHPo surfaces. Teflon® fluoropolymer is spun onto the entire surface and baked; this additional coating is less than 100 nm thick and less than 5 nm rough. It preserves the original random structures of the etched silicon, as can be seen in the inset of Fig. 1. Photoresist with added surfactant (to aid wetting on the fluoropolymer surface) is spun onto the surface. The photoresist is patterned using photolithography. The exposed fluoropolymer is removed by oxygen plasma in an RIE machine for 3 min. Where the coated fluoropolymer is etched away, the underlying oxidized nanostructures, i.e., the SHPi surface, are exposed. Fig. 1 also characterizes the wettability of each surface, using visualization of 100 μL drops at ambient temperature.

2.1.2. Heater fabrication

Thin film heaters made of indium tin oxide (ITO) are directly deposited on the reverse side of the silicon wafer used to create the SBPi surfaces. The surface has a thermally grown oxide layer for electrical passivation. First, ITO is sputtered onto the silicon wafer in a custom Angstrom deposition chamber. The heater geometry of 1 cm × 3 cm is obtained by using a polycarbonate shadow mask. A target resistance of 50 Ohms was used to determine the ITO thickness, which was typically 300 nm. Copper electrodes of 1 cm × 1 cm were thermally deposited onto each end of the ITO heater, also using a polycarbonate shadow mask, leaving a 1 cm × 1 cm square of ITO exposed. The heater was electrically passivated by depositing a 50–100 nm layer of SiO_2 using a Semicore e-beam evaporator.