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Photonically enhanced flow boiling in a channel coated with carbon nanotubes

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High heat dissipation rates are enabled by multi-phase cooling schemes owing to latent heat uptake. We demonstrate enhanced flow boiling from a carbon nanotube (CNT)-coated copper surface exposed to low-intensity ultraviolet (UV)-visible excitation. Compared to non-illuminated results, the average boiling incipience temperature decreased by 4.6 °C and heat transfer coefficients improved by 41.5% with light exposure. These improved results are attributed to augmented hydrophilicity upon exposure to UV light and possible nanoscale opto-thermal effects, and suggest opportunities for active temperature control of temperature-sensitive devices. © 2012 American Institute of Physics. [doi:10.1063/1.3681594]

The ongoing miniaturization of electronic devices has been accompanied by an increasing thermal load per unit surface area. This phenomenon has necessitated conceiving advanced cooling schemes for thermal management. The utilization of the latent heat of vaporization of a fluid makes boiling an attractive heat dissipation scheme. Flow boiling has been a topic of intense research during the past decades because of its potential to achieve high heat dissipation rates while maintaining relatively uniform temperatures across the boiling surface. Several approaches have been pursued in prior work to improve the performance of boiling surfaces. Surface modifications provide a means of enhancing boiling performance and include surface coatings,¹⁻⁴ augmentation of surface roughness,⁵ and use of nanostructured surfaces.⁶⁻¹⁰ High critical heat flux (CHF) has also been realized with wicking structures that provide continuous replenishment of liquid to the boiling surface by capillary action, thereby delaying the dry out of the heated region.^{11,12} However, these methods are passive and do not facilitate active control of surface temperatures and two-phase flow patterns in the cooling system. Active control is desirable in many of today's emerging technologies because of irregularities in the system environment, changing power loads, and the potential to reduce energy consumption. This study explores the possibility of using light to actively control the flow boiling performance. In particular, photonically excited carbon nanotube (CNT)coated copper surfaces are examined as a means to actively control boiling phenomena.

CNTs are extremely thin tubes of graphitic carbon with outer diameters ranging from 1 to 100 nm and typical lengths from 1 to $50 \,\mu$ m. They possess several unique advantages that could be exploited to improve boiling performance. CNT arrays can provide zero angle and "reservoir-type" cavities that are extremely effective at initiating and sustaining bubble nucleation.⁹ CNTs are also known to possess high

axial thermal conductivity.¹³ The heat transfer area is expected to increase substantially, with CNTs acting as high thermal conductivity fins. Metallized CNT arrays have been shown to be efficient wicking structures due to capillary size effects that scale inversely with pore size.¹²

One disadvantage of the multi-walled CNTs, however, is their inherent hydrophobicity. A recent report indicates that multi-walled CNTs exhibit a reversible hydrophobic to hydrophilic behavior upon exposure to low-intensity UV light.¹⁴ This property of CNTs could possibly be exploited to obtain an active tunable surface wetting behavior of a CNTcoated surface and an improved thermal performance. Another foreseeable enhancement effect associated with photo-stimulated CNTs is associated with the localized absorption of light. Nanostructured surfaces absorb light by trapping photons in small cavities and locally enhancing electromagnetic fields. Consequently, the local heat flux on a nanoscale feature such as a CNT can be considerably higher than the average, triggering nucleation of vapor embryos. For example, localized light absorption in CNTs has been reported to produce high local temperatures reaching 1500 °C with a low optical energy flux of 0.1 W/cm².¹⁵ The overall goal of this study is to characterize the changes in flow boiling behavior caused by photonic excitation of CNTcoated surfaces.

Figures 1 and 2 depict the construction of the flow boiling test module and the schematic of the flow loop designed to deliver the working fluid to the test module, respectively. Detailed description of the test module and the design of the flow loop are provided in supplementary information (SI).¹⁶ Deionized and degasified water was used as the working fluid for this study.^{17,18} Test surfaces were illuminated with a low intensity UV-visible (UV-VIS) light-emitting diode (LED) area light with an optical energy flux of 83 mW/cm². The specifications of the light source utilized for photonic excitation are outlined in SI.¹⁶ A low-intensity light source was used to minimize heating of the sample surface by the

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FIG. 1. (Color online) Schematic of the flow boiling test module depicting the four main components and the heating network. Inset shows a schematic of the CNT-coated copper substrate.

UV-VIS light, ensuring that any enhancements in flow boiling performance due to photo-excitation are not the result of broad area surface heating by the light source. Assuming 100% light absorption, the energy fluxes from the light source are at least two orders of magnitude less than the heat fluxes measured during flow boiling. We also note that only 21% of the area of the substrate was exposed to the light source.

Two different sample surfaces were tested in this work. The first sample consisted of a bare oxygen free copper substrate and the second sample consisted of CNTs grown on an identical copper substrate. Both these substrates were roughened to a 400 grit sandpaper finish ensuring that equivalent copper surface morphologies were studied. Well-anchored CNTs were grown on the roughened copper substrates using a microwave plasma chemical vapor deposition (MPCVD) technique.^{19,20} CNT growth occurred under a hydrogen plasma with the addition of methane at a substrate temperature of 900 °C, as described in prior work.²⁰ The diameter of the CNTs varied roughly between 30 and 80 nm. Field emission scanning electron microscopy (FESEM) images (Figures 3(a) and 3(b)) revealed that the CNTs formed a moderately dense and randomly oriented mesh on the surface of the copper substrate.

The procedure adopted for obtaining the boiling curves and estimating the heat flux reaching the boiling surface is elaborated in SI.¹⁶ The flow boiling tests were performed with inlet temperature of 59.5 ± 0.5 °C and a volumetric



FIG. 2. (Color online) Schematic of the flow boiling experimental test loop.



FIG. 3. (Color online) FESEM image of CNTs grown on copper substrate: (a) low magnification and (b) high magnification—prior to testing. (c) High magnification—post testing and (d) flower like CuO nanostructures.

flow rate of 222 ml/min, yielding a Reynolds number Re \cong 675 and a mass velocity G \cong 38 kg/m²s. Since the goal of this study was to assess the effect of low intensity UV-VIS photonic excitation of CNT-coated copper surfaces on boiling incipience and bubble nucleation, and to avoid surface damage, all experiments were terminated well before CHF.

A total of nine flow-boiling experiments were conducted (T1-T9), two experiments with the bare copper substrate and seven experiments with the CNT-coated copper substrate. The first two flow-boiling experiments (T1, T2) with the bare copper substrate assessed the effects of UV-VIS photonic excitation on roughened copper surfaces (i.e., one experiment with UV-VIS light and one experiment without UV-VIS light). The next seven successive experiments (T3-T9) tested the flow boiling performance with the CNT-coated copper surface. Experiment T3 was conducted before light exposure, and experiments T4-T9 were conducted after alternating exposures to UV-VIS light (56 h for T4, 42 h for T6, and 58 h for T8) and complete darkness (24 h for T5, T7, and T9). The same CNT-coated substrate was used in all seven of these CNT boiling experiments (T3-T9).

We stress that the single-phase heat flux dissipation results of these seven experiments were consistent and no significant differences in heat transfer coefficient (HTC) were observed. Also, the mass velocity ($G \cong 38 \text{ kg/m}^2\text{s}$) employed in this work is expected to be too low to cause any substantial surface modifications.⁸ High magnification SEM images prior to and post testing (Figures 3(b) and 3(c)) reveal minimal erosion of CNTs even after approximately 70 h of thermal testing. Any possibility of erosion of these CNTs from the substrate leading to formation of nanofluids during the experiments is nullified as the coolant is drained out without any recycling (Figure 2).

Flow boiling data obtained in this study are shown in Figure 4, plotting the heat flux (q'') as a function of the difference between the mean surface temperature (T_s) and the water inlet temperature (T_{in}) . SI furnishes data used as input for uncertainty quantification.¹⁶ Figure 4(a) compares the flow boiling performance with and without light for the bare copper substrate. The data are within experimental error,

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FIG. 4. (Color online) Flow boiling curves for (a) bare copper surface and (b) CNT-coated copper surface with $G \cong 38 \text{ kg/m}^2 \text{s}$ and $T_{in} = 59.5 \pm 0.5 \text{ }^{\circ} \text{C}$.

indicating no significant enhancement due to UV-VIS illumination. Figure 4(b) depicts the results obtained for the seven boiling experiments with the CNT-coated substrate. The thermal characteristics of the CNT-coated substrate with light exposure (T4, T6, and T8) are consistent and within experimental uncertainties. As shown, there is a pronounced effect due to photonic excitation of the CNT-coated surface compared to the behavior prior to the first illumination sequence (T3). It was calculated that the average boiling incipience superheat reduced by 4.6 °C and the effective HTC increased by 41.5%. Interestingly, for the experiments conducted after darkness conditioning, the boiling curves do not revert to that of the original non-illuminated experiment. If the darkness-conditioned boiling curves (T5, T7, and T9) are considered separately, a leftward shifting trend is observed, suggesting an increased retention of the surface change that produced the original enhancement with increase in total time duration of photonic excitation.

Prior work on multi-walled CNTs exposed to UV light of intensity as low as $2 \mu W/cm^2$ revealed a reversible surface wetting behavior, switching from superhydrophobic to superhydrophilic, similar to many transition metal oxides.¹⁴ The prevailing explanation is that exposure to UV light decreases the tendency of oxygen molecules to adsorb such that hydrophilic hydroxyl groups are favored, making the CNTs hydrophilic. Hence, CNT-coated boiling surface area exposed to the light source is expected to exhibit hydrophilicity. Recently, mixed hydrophilic and hydrophobic areas named "hydrophobic networks" were reported to enhance boiling performance.² Likewise, the entire CNT-coated surface used in this study can be envisioned as a hydrophobic network with a central hydrophilic island (because of light exposure of 21% of the boiling surface area), acting to enhance heat removal rates as witnessed in experiments T4,T6, and T8.

The leftward shift of the darkness-conditioned boiling experiments (T5, T7, and T9) in Fig. 4(b) may correlate to the behavior of some transition metal oxides that when stored in the dark after exposure to low intensity light regain their hydrophobicity over extended times (1000 s of sec).²¹ Similar observations have been reported in case of multiwalled CNTs as well.¹⁴ The light intensity used in this work is four orders of magnitude greater than that reported in Ref. 14 and hence the relaxation to the hydrophobic state is expected to be much slower than 24 h reported therein. The total time duration of exposure (including previous tests) to photonic excitation prior to each of the experiments T5, T7, and T9 was 56, 98, and 156 h, respectively, and the substrate was allowed 24 h to partly relax back to the hydrophobic state in each case. We postulate that with an increase in the total time duration of photonic exposure with the darknessconditioned boiling experiments, the boiling surface turns more hydrophilic and requires longer time to recover its inherent hydrophobicity, thereby explaining the observed leftward shift of these boiling curves. It is interesting to note that the boiling curve of experiment T9 exhibits similar thermal performance characteristics to the photonically excited CNT-coated experiments (T4, T6, T8). This observation opens possibilities of prolonged photonic treatment of the CNT-coated surface for extending enhanced thermal performance.

A second hypothesis for enhanced heat dissipation in the case of photonically enhanced CNTs is based on localized light absorption in nanostructures. Localized thermal energy concentration due to absorption of light has been reported to result in increase in local temperature to the orders of several hundred degrees.^{15,22} The distribution of heat generated by the opto-thermal effect is highly dependent on the density of the nanotubes. When CNT densities are high, localized thermal densities would be less due to loss of heat into the bulk of the sample. SEM images (Figs. 3(a) and 3(b)) clearly show the copper surface beneath the CNT layer signifying the low number densities of CNTs which may aid in localized increase in temperature, enabling the formation and growth of vapor embryos beyond that of a non-illuminated surface. Thus photonic enhancement might aid in increasing the bubble nucleation density.

FESEM images obtained after the last experiment (T9) revealed the sporadic growth of flower-like CuO nanostructures.^{23,24} The rationale behind improved boiling performance could also be attributed to these CuO nanostructures. However, the growth of these nanostructures was not observed after preliminary boiling experiments with similar CNT coated substrates exposed to UV-VIS illumination for shorter duration (approximately 20 h), and consequently they are expected to have formed upon extended photonic excitation. Because the enhancement of parameters associated with boiling heat transfer such as onset of boiling and HTC was evident without the formation of these flower-like nanostructures, and the area fraction of the boiling surface occupied by the CuO nanostructures after the last experiment (T9) was 1-2 orders of magnitudes less than that of the CNTs, the formation of CuO nanostructures is not attributed as the primary mechanism for increasing the nucleation site density and bubble ebullience frequency.

Comparison of results for all the nine boiling tests reveals several additional features. The CNT-coated surface before exposure to light (T3) did not perform as well as the plain copper surface as indicated by the onset of boiling and slope of the boiling curve in Figures 4(a) and 4(b). This behavior is not totally unexpected, as heat dissipation from CNT-coated surfaces has shown contrasting trends in the past under different experimental conditions in case of flow in micro-channels.^{7–10} Mass velocity (G) constitutes a critical parameter in the enhancement/degradation of boiling performance with CNT-coated surfaces in flow boiling configurations reported in prior work.^{8,9} Both performance enhancement⁸ and degradation⁷ trends have been observed with low mass velocities (G \cong 86 kg/m²s), supporting the poorer heat dissipation characteristics of non-illuminated CNT-coated surface observed in this work ($G \cong 38 \text{ kg/m}^2 \text{s}$). Slightly different CNT growth protocols used in this work can influence the CNT quality, diameter, number density, array height, and anchoring to the copper surface. All the foregoing factors can affect flow boiling characteristics. However, optimizing the CNT parameters for enhanced heat dissipation and illuminating the CNT-coated surface with low-intensity illumination for additional thermal gains presents opportunities for active "programmable" temperature control.

In summary, we have demonstrated that CNT-coated surfaces can be photonically excited to achieve significant reductions in incipience superheat and increases in HTC during flow boiling. Two hypotheses are proposed for the observed boiling performance enhancement with lowintensity light illumination. Additional studies are warranted to study further the ability to control boiling by varying the intensity and pattern of the illumination. Funding from the U.S. Air Force Office of Scientific Research, Thermal Sciences (Program Manager: Dr. Kumar Jata) is gratefully acknowledged.

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