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Effects of nanoparticle deposition on surface wettability influencing boiling heat transfer in nanofluids

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Buildup of a porous layer of nanoparticles on the heated surface occurs upon boiling of nanofluids containing alumina, zirconia, or silica nanoparticles. This layer significantly improves the surface wettability, as shown by a reduction of the static contact angle on the nanofluid-boiled surfaces compared with the pure-water-boiled surfaces. The contact angle reduction is attributed to changes in surface energy and surface morphology brought about by the presence of the nanoparticle layer. The high surface wettability can plausibly explain the boiling critical heat flux enhancement in nanofluids.

Nanofluids are engineered colloidal suspensions of nanoparticles in a base fluid and exhibit a very significant enhancement (up to 200%) of the boiling critical heat flux (CHF) at modest nanoparticle concentrations. Since CHF is the upper limit of nucleate boiling, such enhancement offers the potential for major performance improvement in many practical applications that use nucleate boiling as their prevalent heat transfer mode. For example, the use of nanofluids with higher CHF could enable effective thermal management of ever smaller and more powerful electronic devices, enable power uprates in commercial nuclear plants, allow design of more compact heat exchangers for the chemical industry, etc. To study the two-phase heat transfer characteristics of nanofluids, we have conducted pool boiling experiments with water-based nanofluids containing Al2O3, ZrO2, and SiO2 nanoparticles at volume concentrations of 0.1%, 0.01%, and 0.001% (total of nine nanofluids). The nanofluids were prepared by dilution of concentrated (10 wt %) nanofluids purchased from various vendors. The size (effective diameter) of the nanoparticles in the dilute nanofluids was measured with dynamic light scattering and ranged from 110 to 210 nm for Al2O3 nanofluids, from 110 to 250 nm for ZrO2 nanofluids, and from 20 to 40 nm for SiO2 nanofluids. With these nanofluids we have measured a CHF enhancement of up to 50% in previous experiments. The heaters were flat plates, 5 mm wide, 45 mm long, and 0.05 mm thick and were made of grade 316 stainless steel. We observed that soon after nanofluid boiling is initiated, some nanoparticles precipitate on the heater surface and form irregular porous structures, which do not appear during boiling of pure water (Fig. 1). The energy dispersive spectrometer analysis of the surface confirmed that the porous layer is made of the same material of the nanoparticles used in the experiment. Similar nanoparticle precipitation was observed by other researchers experimenting with nanofluid boiling.

Various mechanisms of nanoparticle precipitation and adhesion to the heater surface have been hypothesized (e.g., liquid evaporation, surface chemical reactions, electric fields, and dip coating); however, regardless of the mechanism, the presence of a porous layer on the surface can undoubtedly have a significant impact on boiling heat transfer through changes in surface area, surface wettability, and bubble nucleation.

In this letter we intend to answer the following three questions. Does the nanoparticle layer change the wettability of the heated surface? What are the physical mechanisms that cause the wettability change? What are the consequences on nucleate boiling heat transfer and CHF?

In order to investigate surface wettability, the static contact angle \( \theta \) was measured for sessile droplets of pure water and nanofluids at 22 \(^\circ\)C in air on clean surfaces and nanoparticle-fouled surfaces. The sessile droplet method is a well established technique for assessing wetting of a solid by a liquid and has been used routinely in boiling heat transfer studies. Low values of the contact angle correspond to high surface wettability. Figure 2 shows that the contact angle decreases from about 70\(^\circ\) to about 20\(^\circ\) on the fouled surfaces. Such decrease occurs with pure water as well as nanofluid droplets, thus suggesting that wettability is enhanced by the porous layer on the surface, not the nanoparticles in the fluid. In another research, Wasan and Nikolov found that ordering of nanoparticles near the liquid/solid contact line can increase the spreading of nanofluids.

![FIG. 1. Scanning electron microscope images of stainless steel surface boiled in (a) pure water, (b) 0.01 vol % Al2O3 nanofluid, (c) 0.01 vol % ZrO2 nanofluid, and (d) 0.01 vol % SiO2 nanofluid.](Image)
To understand the dramatic enhancement of wettability, we consider Young’s equation

\[ \cos \theta = \frac{\gamma_{SV} - \gamma_{SL}}{\gamma_{LV}} \]

which relates the static contact angle to the so-called adhesion tension \( \gamma_{SV} - \gamma_{SL} \) and the surface tension \( \gamma_{LV} \). The adhesion tension of water on clean steel is \( \sim 10 \) mN/m and its surface tension is \( \sim 72 \) mN/m, so Young’s equation yields a contact angle of about 82°, which is in reasonable agreement with the measured angle [Fig. 2(a)]. If the surface is not smooth, the effective solid-liquid contact area differs from the smooth contact area. Wenzel\(^{12}\) defines a roughness factor \( r \) as the ratio of the effective contact area to the smooth contact area. The free energy of the solid-liquid interface on a rough surface is then \( r \) times the free energy of a perfectly smooth surface with the same apparent contact area. Therefore, Young’s equation needs to be modified as follows:

\[ \cos \theta' = \frac{\gamma_{SV} - \gamma_{SL}}{\gamma_{LV}} r = r \cos \theta, \]

where \( \theta' \) is the apparent contact angle. Note that the measured contact angles shown in Figs. 2(c) and 2(d) are, in fact, apparent contact angles because of the presence of the porous layer. Equation (2) suggests that the contact angle depends on three parameters, (i) the surface tension (\( \gamma_{LV} \)), (ii) the adhesion tension (\( \gamma_{SV} - \gamma_{SL} \)) and (iii) the roughness factor. The surface tension of our nanofluids was measured with a Sigma 703 tensiometer and found very close (within ±3%) to that of pure water. On the other hand the adhesion tension of water increases significantly in going from a clean metal to an oxide, e.g., from \( \sim 10 \) mN/m (stainless steel) to \( \sim 60 \) mN/m (alumina). Such change in adhesion tension alone reduces the contact angle to \( \sim 34^\circ \), as calculated from Eq. (2) assuming \( r=1 \). This is consistent with other studies showing that surface oxidation decreases the contact angle.\(^9\) The porous layer also increases the effective contact area. Thus the roughness factor \( r \) is greater than unity, which also contributes to the contact angle reduction in our case. To evaluate \( r \), we used a Tencor P-10 surface profilometer, which gave the images shown in Fig. 3. The surface boiled in pure water is very smooth, while the surface boiled in nanofluid presents structures consistent with the images of Fig. 1. The estimated surface areas are of the order of 84 000 and 470 000 \( \mu \)m² for the surface boiled in pure water and the surface boiled in nanofluid, respectively, resulting in \( r \sim 5.6 \). This is to be considered an upper bound estimate for \( r \), because it assumes that the pores are completely filled with liquid. In reality the capillary pressure pulls the liquid into the pores until it is balanced by the pressure of the trapped gases. For \( r \sim 5.6 \) the apparent contact angle decreases to \( \sim 39^\circ \), as calculated from Eq. (2) with nominal adhesion tension (\( \sim 10 \) mN/m) and surface tension (\( \sim 72 \) mN/m). In summary, a simple analysis of the modified Young’s equation suggests that the enhancement in wettability (decrease in contact angle) is caused by a combination of two effects, i.e., an increase of adhesion tension and an increase of surface roughness. Both effects are at work and both effects are large enough to cause the observed reduction in contact angle.

We shall now discuss the impact of wettability enhancement on boiling heat transfer in nanofluids. Activation of microcavities on the surface is generally described by the criterion of \( \theta \geq \beta \), where \( \beta \) is the opening angle of an idealized conical cavity. According to Yang and Kim,\(^{13}\) the number of active cavities per unit surface area, \( N_a \), is

\[ N_a = N_i \int_{R_{\text{min}}}^{R_{\text{max}}} \varphi(R)dR \int_0^\theta \phi(\beta)d\beta, \]

where \( N_i \) is the cavity areal density, \( R_{\text{max}} \) and \( R_{\text{min}} \) are the maximum and minimum active cavity radii, respectively, \( \varphi(R) \) is the cavity radius distribution function, and \( \phi(\beta) \) is the distribution function for the cavity opening angle. Therefore, a decrease of the contact angle, as was observed on nanoparticle-fouled surfaces, will tend to decrease the number of active cavities. Plausibly this contributes to the decrease in bubble nucleation in nanofluids with respect to pure surfaces.

**FIG. 2.** (Color online) Contact angles of 5 \( \mu \)l sessile droplets on stainless steel surfaces, measured with a Krüss goniometer (uncertainty ±0.1°) equipped with a camera monitor. (a) Pure water droplet on surface boiled in pure water, (b) 0.01 vol % \( \text{Al}_2\text{O}_3 \) nanofluid droplet on surface boiled in pure water, (c) pure water droplet on surface boiled in 0.01 vol % \( \text{Al}_2\text{O}_3 \) nanofluid, and (d) 0.01 vol % \( \text{Al}_2\text{O}_3 \) nanofluid droplet on surface boiled in 0.01 vol % \( \text{Al}_2\text{O}_3 \) nanofluid. Similar results were obtained with the \( \text{SiO}_2 \) and \( \text{ZrO}_2 \) nanofluids.

**FIG. 3.** (Color online) Profilometer images of the stainless steel surface after boiling (a) pure water and (b) 0.1 vol % alumina nanofluid. The rms roughness values are \( \sim 0.1 \) and \( \sim 2 \) \( \mu \)m, respectively. Similar results were obtained with the other nanofluids.
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To estimate the impact of the observed contact angle on heating over CHF. Clearly, an increase in surface wettability also profoundly affects CHF. While a generally valid theory of CHF does not exist, the most credible hypotheses postulate that dry patches (or hot spots) develop on the heated surface at high values of the heat flux. These dry patches can be rewet or can irreversibly overheat, which causes CHF. Clearly, an increase in surface wettability promotes dry patch rewetting, thus delaying CHF. To estimate the impact of the observed contact angle change on CHF, we can use the simple model of Sadashivan et al. This model postulates that bubbles growing at neighboring active cavities can coalesce laterally at a certain stage during the growth phase before they depart from the surface. A certain volume of liquid is trapped between the vapor bubbles below the plane of coalescence [Fig. 5(a)]. Above the plane of coalescence, the vapor may form a large mushroom-shaped bubble that hovers over the surface for a time \( \tau_b \). The bubbles are assumed to have uniform size with radius \( r_b \), and be uniformly distributed over the heater surface. Therefore, for a \( 2r_b \times 2r_b \) unit cell the volume of liquid trapped below the plane of coalescence is

\[
\Delta \text{Vol} = 4 \pi r_b^2 \cos \theta^* - 3 \pi r_b^2 (3 \cos \theta^* - \cos^3 \theta^*). 
\]

The equivalent thickness of the liquid film is then calculated assuming uniform spreading of the trapped liquid over the surface as

\[
\delta_l = r_b \left[ \cos \theta^* - \frac{\pi}{12} (3 \cos \theta^* - \cos^3 \theta^*) \right].
\]

For given \( r_b \), the liquid film thickness increases with decreasing contact angle [Fig. 5(b)]. The time to dry out the liquid film, \( \tau_b \), can be estimated from the energy balance at the surface as

\[ \tau_b = \delta_l \rho L / q^* \]

where \( q^*, \rho \) and \( h_f \) are the heat flux, liquid density, and evaporation heat, respectively. Sadashivan et al. hypothesize that CHF occurs if \( \tau_d < \tau_b \). Since \( \tau_b \) depends mostly on hydrodynamics and only weakly on the heat flux, the increase in \( \tau_d \) has a direct effect on CHF; i.e., the value of the heat flux at which CHF occurs is proportional to \( \tau_d \). Equation (4) suggests that a contact angle reduction from 70° to 20° would increase the liquid layer thickness about fourfold, which results in a fourfold increase of the dryout time and thus in roughly a fourfold increase of the CHF. Recognizing that the CHF enhancement reported in the literature is of the same order of magnitude (up to 200%) but not quite so large, we can, however, conclude that the liquid layer model points to a strong nexus between CHF enhancement and surface wettability improvement caused by nanoparticle deposition.

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