Nucleate boiling heat transfer enhancement for water and FC-72 on titanium oxide and silicon oxide surfaces

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ABSTRACT

An experimental study was performed to investigate the nucleate boiling and critical heat flux (CHF) of water and FC-72 dielectric liquid on hydrophilic titanium oxide (TiO$_2$) nanoparticle modified surface. A 1 cm$^2$ copper heater with 1 µm thick TiO$_2$ coating was utilized in saturated pool boiling tests with water and highly-wetting FC-72, and its performance was compared to that of a smooth surface. Results showed that TiO$_2$ coated surface increased CHF by 50.4% and 38.2% for water and FC-72, respectively, and therefore indicated that boiling performance enhancement depends on the level of wettability improvement. A silicon oxide (SiO$_2$) coated surface, exhibiting similar surface topology, was tested to isolate the roughness related enhancement from the overall enhancement. Data confirmed that hydrophilicity of TiO$_2$ coated surface provides an additional mechanism for boiling enhancement.

1. Introduction

It is well-known that boiling heat transfer can be enhanced if the solid–liquid contact angle is lowered [1–2]. In their experimental work, Takata et al. [3–4] coated heater surfaces with TiO$_2$ layer and showed boiling enhancement for water. TiO$_2$ coating has a high affinity for water, and in turn significantly reduces water contact angle close to zero degrees and makes the surface superhydrophilic. Boiling heat transfer enhancement by using TiO$_2$ surfaces therefore has been of research interest recently. Additionally, fouling or scale on a surface can be inhibited by TiO$_2$ coating layer due to its photocatalysts effectiveness as reported by Wang et al. [5]. TiO$_2$ decomposes various organic substances and undesirable deposits in solid phase or soft mud by its strong oxidizing power. There are numerous potential applications of TiO$_2$ used in evaporators or evaporation concentrators found in the process industry to simultaneously improve heat transfer and antifouling. TiO$_2$ coating can also be adapted in other boiling heat transfer applications such as heat pipes and immersion cooling of micro-electronic components.

Literature on boiling enhancement of TiO$_2$ coated surfaces mostly included water as working fluid. Similar investigations with other commonly used fluids, such as FC-72, are still needed. Moreover, previous researchers [3–5] attributed the boiling enhancement of TiO$_2$ coating mainly to its high affinity for water. These studies neglected to address the effect of surface roughness which can easily change when a substrate is coated with TiO$_2$ layer. Many documented works, such as Kim et al. [6] and Li et al. [7] on the other hand, pointed out the boiling enhancement due to the micro and nano scale structured surfaces.

In this study we experimentally investigated the effect of TiO$_2$ coated surface on nucleate boiling and critical heat flux (CHF). Two types of fluids, water and FC-72, were chosen for saturated pool boiling tests. FC-72 is a dielectric fluid with very high wettability on most surfaces, and consequently, one might expect that TiO$_2$ coatings would make no improvement for boiling of FC-72. A smooth copper surface was considered as the reference surface for enhancement comparison. SiO$_2$ coated surface was also included as a non-hydrophilic surface which exhibits surface roughness similar to that of TiO$_2$ modified surface. With the use of SiO$_2$ coated surface in the tests, we aimed to rule out the corresponding boiling enhancement contribution attributable to the effect of surface roughness.

2. Experimental setup and procedure

The pool boiling test setup used in the present work is shown in Fig. 1. A cylindrical acrylic container with aluminum lids, 300 mm high and 200 mm in diameter, held the pool of 70–80 mm deep water or FC-72 working fluids at atmospheric pressure. The saturated pool condition inside the vessel was maintained by an auxiliary heater and monitored with two immersed type-T thermocouples and a pressure gage on the top lid. A reflux condenser mounted
above the pool condensed the vapor produced during the experiments and kept the liquid level constant. A 1 cm² copper heater was placed on a 25 mm thick bakelite substrate at the bottom of the pool and controlled by a programmable DC power supply. The bakelite layer also minimized heat loss from pool to ambient air.

2.1. Heaters and test surfaces

The heaters were built by soldering a 10 × 10 × 2 mm copper block onto a matching size, 1-mm thick, 5-ohm thick-film resistive heater. Heat flux was determined from the total power supplied into resistor per unit base area (\( q'' = V I / A \)). Heater temperature was monitored with two type-T, 36 AWG size thermocouples embedded halfway in the heater wall and located in the center and midway between the center and edge as indicated in Fig. 1. Temperature at the boiling surface was calculated by extrapolating average of two thermocouple readings through 1 mm distance to surface and assuming steady 1-D conduction through heater wall (\( T_w = T_{Cavg} - (q'' x) / k \)).

All copper test surfaces were initially polished using 600 mesh diamond abrasives and 0.1 μm lapping compound to have an almost mirror finish. The surfaces were cleaned using first acetone, and then 1% HCl. Finally the surfaces were rinsed with distilled water. After assigning one of these smooth copper surfaces as reference, other heaters were coated with TiO₂ and SiO₂ layers. Process involved depositing one drop (0.01 ml) of diluted 1% TiO₂ (or SiO₂)-ethanol solution onto the copper surface, spreading the solution across 1 cm² test surface, and heating the substrate to 200°C. When ethanol evaporated, 10 nm size particles bonded to surface forming an approximately 1 μm thick coating layer. Uniformity of the resulting coating was confirmed through visual inspection and consistent contact angle measurements at various locations of surface.

The smooth copper and TiO₂ or SiO₂ coated surfaces were characterized by scanning electron microscope (SEM) and optical profilometer. Fig. 2 includes SEM pictures of TiO₂ and SiO₂ coated surfaces at 1000 and 5000× magnifications and shows a similar topology in general. As shown in Fig. 3, detailed roughness measurements of these surfaces with optical profilometer indicated an average roughness, \( R_a \), of 1.55 and 1.41 μm for TiO₂ and SiO₂ coated surfaces, respectively. These measurements therefore, confirmed that TiO₂ and SiO₂ samples have comparable surface characteristics in terms of texture and roughness. In comparison, smooth copper surface had a much lower \( R_a \) at 0.3 μm.

2.2. Test conditions and procedure

All heat transfer tests were conducted in saturated pool at atmospheric pressure. Prior to each experiment, the test surface was soaked and cleaned with acetone, and rinsed with distilled water to remove contaminants. After installation of heater, the test vessel was evacuated and filled with the working fluid, water or FC-72. For FC-72 tests, an additional degassing process was carried out by boiling liquid pool for two hours to remove absorbed noncondensibles. Boundary condition in the tests was constant heat flux. Boiling curves were generated by increasing heat flux.
gradually in steps of 2 W/cm² up to CHF and recording corresponding heater temperatures every 3 s. Measurements at each step were continued until steady-state boiling condition was attained, typically in 2–3 minutes. When CHF was detected, the heat flux was set to 95% of CHF and then decreased gradually back to 0 W/cm².

2.3. Uncertainty analysis

Uncertainties were estimated mainly for heat flux and temperature measurements. The data acquisition system featured a digital voltmeter having a sensitivity of ±1 μV, a six-figure scale and an accuracy of 0.01% of the reading. The relative error of the heat flux was estimated to be within ±3.8% at the maximum heat flux rate. Embedded thermocouples in heater wall were carefully calibrated to an accuracy of ±0.1 °C.

3. Results and discussion

Static contact angles of water and FC-72 were measured on the three types of test surfaces using sessile drop method. Measurements were taken at elevated liquid and substrate temperatures to represent saturated pool boiling conditions better. Still images in Fig. 4 indicate water contact angles of 62°, 57° and 9° for smooth copper, SiO₂ coated and TiO₂ coated surfaces, respectively, all thermally activated after being exposed to 100 °C for 1 min. Results clearly demonstrated that TiO₂ has a high affinity for water. However, due to its highly wetting nature, FC-72 contact angle measurements on the same, thermally activated test surfaces resulted in nearly 0° with no distinguishable difference.

Boiling curves of smooth copper, SiO₂ coated and TiO₂ coated surfaces for water and FC-72, in the form of steady state surface superheats as a function of heat fluxes, are presented in Fig. 5. In all tests, heat flux was increased in steps of approximately 2 W/cm² up to CHF and then decreased back in a similar manner. In water tests, all heaters gave nearly identical boiling curves for increasing and decreasing heat flux conditions. At heat fluxes <60 W/cm², all surfaces performed similarly. At higher heat fluxes, SiO₂ coated surface performed slightly better than smooth copper surface, and TiO₂ coated surface provided a significant enhancement over both surfaces. Smooth copper surface reached to CHF at 121 W/cm² which is very close to Zuber’s CHF limit of 120 W/
cm², calculated by

\[ q_{\text{CHF}} = C_k \rho_v h_{fg} \left( \rho_l - \rho_v \right) \sigma^{1/4} \]

where \( C_k \) is 0.131 and all fluid properties are at 100°C and one atmosphere. SiO₂ coated and TiO₂ coated surfaces gave higher CHF values of 134 and 182 W/cm², respectively, improving smooth copper surface's CHF by 10.7% and 50.4%. In FC-72 tests, surface temperature overshoot at boiling incipience occurred at 16–17°C superheat due to high wettability of FC-72 on all test surfaces. Beyond incipience, matching boiling curves were observed for increasing and decreasing heat flux conditions of each heater, indicating a consistent trend with earlier studies [8,9]. SiO₂ coated surface showed an obvious enhancement over smooth copper surface at heat fluxes >5 W/cm². TiO₂ coated surface outperformed other two surfaces throughout the entire nucleate boiling range. CHF attained by smooth copper surface was 16.5 W/cm², which is comparable to previously reported values at the range of 13.2–19 W/cm² [8,10], and higher than the Zuber's CHF estimation at 15.1 W/cm² when \( C_k \) is 0.131 and all fluid properties are at 56°C and one atmosphere. It is also known that below a certain heater size length-scale, CHF increases with decreasing heater size and Zuber's correlation underpredicts observed CHF [10]. SiO₂ and TiO₂ coated surfaces offered higher CHF values at 18.8 and 22.8 W/cm², respectively, and resulted in 13.9% and 38.2% CHF enhancement over smooth copper surface.

For an overall performance comparison of all test surfaces in water and FC-72, heat transfer coefficients, defined as

\[ h = q/(T_w - T_{sat}) \]

in high heat flux range of water tests, SiO₂ and TiO₂ coated surfaces provided up to 8.5% and 36.2% higher heat transfer rate, respectively, over smooth copper surface. In FC-72 tests, the improvement by SiO₂ and TiO₂ coated surfaces was more at up to 45.6% and 91.2%, respectively. Additionally, FC-72 data illustrate that heat transfer coefficients, or slope of the boiling curves, start to decrease as they approach to CHF. In this region, as more nucleation sites become active, vapor covered dry surface area also increases and leads to a less effective heat transfer.

Present data demonstrates that TiO₂ coated surface greatly enhances nucleate boiling and CHF performance of both water and FC-72 over our reference smooth copper surface. Previous studies [3–5] attributed their similar enhancement mainly to TiO₂'s high affinity for water. In case of any thermally-activated or photo-activated superhydrophilicity, through the application of heat or high energy light, TiO₂ crystal surface itself is reduced, and oxygen vacancy is created. The oxygen vacancy then bonds to water molecule in the air. Finally, hydroxyl radical (OH) is generated on the surface that acts as chemisorbed water layer. This mechanism explains the reason of high TiO₂ affinity for water.

Considering highly wetting characteristic of FC-72 fluid, we would not necessarily expect an improvement in FC-72 boiling performance with the use of TiO₂ coated heater. Furthermore, despite the non-conclusive contact angle measurements, we can anticipate that TiO₂ affinity for FC-72 (C₆F₁₄) would not be as high due to the lack of OH radical. Nevertheless, present FC-72 results also showed an obvious enhancement with TiO₂ modified surface indicating that it possibly allows for more effective liquid-solid interaction, reduces the dry patches caused by growing bubbles and eventually enhances nucleate boiling and CHF.

Data from SiO₂ sample can be utilized to understand the effect of surface roughness on overall enhancement which was not spe-

![Fig. 5. Boiling curves for smooth copper and SiO₂/ TiO₂ coated surfaces in water and FC-72.](image)

![Fig. 6. Heat transfer coefficients as a function of heat flux for smooth copper and SiO₂/ TiO₂ coated in water and FC-72.](image)
cifically addressed in previous literature. With a very similar surface condition as shown in Figs. 2 and 3, SiO2 coated surface can simulate the roughness related enhancement that TiO2 coated surface provides. In water tests, SiO2 coated surface barely helped to increase performance over smooth copper surface, suggesting that enhancement is mainly due to high affinity of TiO2, as reported before. On the other hand, in FC-72 tests, SiO2 coated surface certainly improved performance, implying that surface roughness plays an important role in enhancement. This seemingly observational conclusion can be explained with a closer look at active cavity size range analysis [11] for water and FC-72 based on Eq. (1), where $\delta_i$ is estimated as $k_i/h$ with $h$ evaluated at the incipience of boiling.

\[
\left\{ \begin{array}{l}
R_{c,\text{max}} \\
R_{c,\text{min}}
\end{array} \right\} = \frac{\delta_i}{4} \left[ 1 - \frac{T_{\text{sat}} - T_i}{T_w - T_i} \left( \frac{T_w - T_i}{T_{\text{sat}} - T_i} \right)^2 \right] \frac{12 \pi \sigma T_{\text{sat}}}{p_i h \delta_i (T_w - T_i)}
\]

Fig. 7 shows active cavity size range for saturated boiling of water and FC-72 in terms of the surface superheat. By assuming that the average roughness of SiO2 and TiO2 coated surfaces (=1.5 μm) corresponds to an average cavity radius of 1 μm, we can estimate the contribution of this roughness level in boiling. As indicated in the plots, 1 μm average cavity radius cannot help boiling of water until surface superheat reaches 27°C (nevertheless, heaters still have numerous scratches after surface preparation that serve as relatively large sites and make boiling inception possible at low superheats as observed in Fig. 5). Same size cavity radius can help boiling of FC-72 starting only at 4°C surface superheat. Hence, it can be concluded that the roughness level on SiO2 coated surface determined its enhancement level over smooth copper in water and FC-72.

Repeating the tests several times with TiO2 and SiO2 coated heaters gave consistent results suggesting that the critical surface conditions of surface wettability and roughness remained nearly same.

4. Conclusion

Effect of a thin layer of hydrophilic titanium oxide (TiO2) coating on the nucleate boiling and the CHF was experimentally studied. In saturated water and FC-72 pool boiling tests, TiO2 coated surface increased CHF by 50.4% and 38.2%, respectively over a smooth copper surface, and indicated that TiO2 enhances boiling performance, of even a highly wetting fluid, FC-72, depending on the level of wettability improvement. It was therefore concluded that hydrophilic TiO2 surface possibly allows for more effective liquid-solid interaction, reduces the dry patches caused by growing bubbles and eventually enhances both nucleate boiling and CHF. A SiO2 coated sample, having surface roughness similar to TiO2 coated one, was also tested to isolate the roughness related enhancement from the overall enhancement. Data confirmed that hydrophilicity of TiO2 coating provides an additional mechanism for boiling enhancement, beyond increasing active nucleation site density.

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References