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Nanoparticle Deposition by Boiling on Aluminum Surfaces to Enhance Wettability

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ABSTRACT

Surface wettability of materials is important in a myriad of HVAC&R processes. This study is focused on how to manipulate wettability by nanofluid boiling nanoparticle deposition on aluminum surfaces. The focus is on aluminum, because it is commonly used as the material for heat transfer in air conditioning and refrigeration systems. The boiling deposition process occurs under atmospheric pressure, in a reservoir large compared to the sample size. The effect of nanoparticle concentration, boiling duration and surface initial roughness are studied by varying parameters one at a time while controlling the others. Al_2O_3 nanoparticles aredeposited on the aluminum surface, as confirmed by microscopy. The static contact angle of water is measured using a goniometer to characterize the wetting behavior. It is observed that the layer of Al_2O_3 nanoparticle deposited on aluminum surfaces enhances the wettability on the surface, and the higher the concentration of nanoparticle in the fluid during boiling deposition, the better the wetting of the surface. Surface roughness before and after the nanofluid boiling process is also investigated. The relationship between the surface topography and water wettability is discussed.

1. INTRODUCTION

It is known that surface wettability is important in boiling (Bourdon *et al.*, 2014), evaporation (Takata *et al.*, 2005), condensation (Wang and Chang, 1998), frosting/defrosting (Rahman and Jacobi, 2013), absorbtion refrigeration (Kim *et al.* 2003) and many other thermal processes important in air conditioning and refrigeration. Surfaces with enhanced wettability are preferred in certain conditions. It was found that when condensation occurs on the air-side of a heat exchanger, hydrophilic fins might save fan power compared to uncoated ones (Liu and Jacobi, 2009). The frost formed on the hydrophilic fins is more dense and thus exhibits higher thermal conductivity than that on hydrophobic fins (Hoke *et al.*, 2004). A hydrophilic surface enhances critical heat flux in pool boiling (Kandlikar, 2001) and reduces two-phase pressure drop in flow boiling (Phan *et al*,. 2011). Different surface wettability manipulation techniques have been developed to enhance the performance of heat exchangers. Takata *et al.* (2005) used TiO_2 with UV light to obtain a superhydrophilic surface for better boiling and evaporation heat transfer. Kim *et al.* (2003) used a plasma induced superhydrophilic fins to enhance thermal-hydraulic performance of a heat exchanger. These techniques involve a surface chemistry treatment, which often raises issues of cost during manufacturing and weathering when in use. The question then arises whether superhydrophilicity can be obtained by topography manipulation only in a way with potential for mass production.

There has been intensive study of heat transfer enhancement by nanofluids for both pool boiling (You *et al.*, 2003) and flow boiling (Ahn *et al.*, 2010). It has been found that critical heat flux (CHF) can be enhanced by nanofluids, due to the presence of a layer of nanoparticle deposition, which leads to surface wettability enhancement (Kim *et al.*, 2007). Microlayer evaporation at the base of the vapor bubble is considered to be the mechanism of nanoparticle deposition during boiling (Kwark *et al.*, 2010). Kim *et al.* (2007) claimed that surface wettability is changed by the nanoparticle layer because surface chemical composition and topography can be modified. It was reported that a change of surface roughness (Vafaei and Borca-Tasciuc, 2013) and capillarity (Kim and Kim, 2007) were observed. In this paper, the topography effect is the focus.

Surface roughness has an impact on both the boiling process and the surface wettablility itself. As a result, for the nanoparticle deposition by nanofluid boiling, it is important to study surface roughness before and after the boiling process in order to understand the mechanism of nanoparticle deposition and wettability change. It has been reported that the surface roughness after nanofluid boiling depends on the original surface roughness scale and the nanoparticle size (Kim *et al.*, 2007). However, little research has been done to investigate the surface roughness effect on wettability manipulation by nanofluid boiling. The growth rate of deposited layer was modeled by Kim *et al*. (2007). According to their model, the thickness of nanoparticle layer should be proportional to time, heat flux, nucleation site density and nanoparticle concentration in the nanofluid. The parameter space is large. In this paper, the research focus is on the influence of nanofluid concentration and surface roughness, which is also directly related to cavity and nucleation site density.

Since aluminum is commonly used for heat exchangers, this metal is used as the specimen for the experiments. An aqueous Al_2O_3 nanofluid was used to deposit Al_2O_3 nanoparticles on aluminum surface of varies roughness by pool boiling, so as to investigate the enhancement of surface wettability due to topography change. The experimental apparatus and procedure are introduced in section 2 and 3. The surface wettability data and its dependence on boiling conditions and roughness are analyzed in section 4.

2. SURFACE PREPARATION

The surface preparation includes surface roughness control and preparation of nanofluids before boiling, followed by nanofluid pool boiling nanoparticle deposition process.

2.1 Surface Roughness Control

Since roughness affects both the boiling behavior and the nanoparticle coated surface morphology, surface roughness control is necessary for a prudent investigation. The surface of each specimen as received from machine shop was anisotropic, where machine marks and grooves of certain pattern can be observed by the naked eye. For this study, an isotropic surface with certain roughness is preferred. A 400 grit alumina sanding sheet was used to obtain a smooth surface, and 60 grit silicon carbide sanding sheet for rough surface. Aluminum oxide abrasive powder of average size 12.5 µm dispersed in a water based fluid used as the slurry on a lapping machine (Lapmaster 12). The specimens were lapped at 60 rpm for 4 hr. After lapping, the machine marks and grooves of the original machined surface were gone, and final finish was mirror like.

2.2 Nanofluid Preparation

The nanofluid was prepared by mixing dry nanoparticle powder and pure water. The Al_2O_3 nanoparticles have an average size of 40 nm. Alumina powder was weighed using a precision balance (Mettler AE200) that has an accuracy of ± 0.1 mg. An Erlenmeyer flask was used to mix the nanoparticle with pure water. The flask was then placed into an Ultrasonic Bath (Bransonic 1510R) for 30 min at a frequency of 42 kHz to achieve a near uniform dispersion of nanoparticles in water. Then, the nanofluid in the flask was poured into the pool for boiling experiment. In order to minimize the nanofluid residual inside the flask, pure water was added to wash the flask three times, and the liquid was poured into the pool. The final concentration of the nanofluids are 0.01wt%, 0.1 wt %, 1 wt %.

2.3 Boiling Deposition Experiment

2.3.1 Experimental apparatus: The vessel is 180 mm x180 mm with a wall thickness of 5 mm, as shown in Figure 1. The cylinder is made of Pyrex borosilicate glass which tolerates temperatures up to 230 ^{o}C. The top and bottom of the vessel are covered by high temperature UHMW plastic plates with sensor hang at the top and the specimen inserted at the bottom. The top plate has a vent, cooling coil, a K-type thermocouple and an absolute pressure sensor. The test specimen is located at the center and leveled with the top surface of a PTFE block. The gap between the block and the test specimen is filled by RTV silicone sealant.

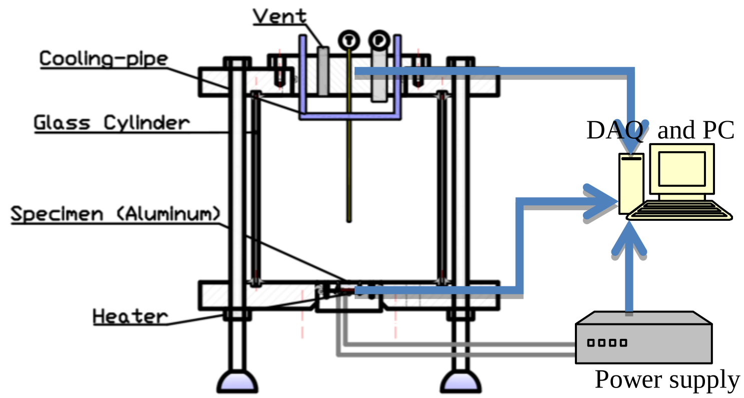


Figure 1: Sketch of boiling deposition experimental facility

The structure of heating section and the specimen is shown in Figure 2. The test specimen is a 20 × 20 mm aluminum block. A square WatLow ULTRAMIC ceramic heater located under the aluminum blocked is used to heat the specimen. A K-type thermocouple was used to measure the temperature of the heater to monitor the heating process. Another K-type thermocouple was inserted into a 0.5 × 5 mm hole in the aluminum specimen, 6.4 mm below the boiling surface, to monitor the temperature of the aluminum. Temperature and pressure readings were captured by an NI 9213. The heating power of the heater was controlled by AGN5771 DC power supply, which has a capacity of 1.5 kW. The heat flux at the boiling surface was found by calculating the DC electric power supplied to the heater divided by the boiling area of the specimen.

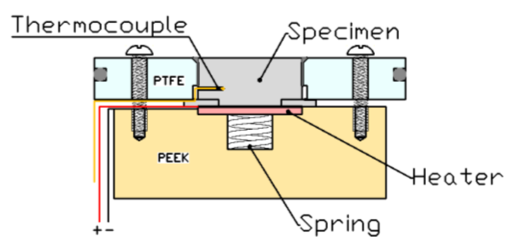


Figure 2: Structure of the heating section

2.3.2 Boiling Deposition Procedure: A circular auxiliary heater of 400 W located at the bottom of the vessel was used to heat up the liquid to saturation temperature, and to degas the liquid. When the change of thermocouple reading for the pool temperature was within the uncertainty of the bulk-fluid thermocouple (± 0.3 ^{o}C) in 10 minutes, it was considered that the liquid temperature profile was at steady state, so the boiling deposition process at the specimen surface began. The DC voltage on the heater was set to be 76 V for all boiling deposition processes, which gave about 140 ±5 kW/m^2 of heat flux at the boiling surface. The pool boiling deposition process was kept at the constant heat flux for 10 minutes, and then the specimen was carefully removed at the bottom without touching the surface and was protected in a case, so that the specimens were ready for surface characterization.

3. SURFACE CHARACTERIZATION

Before characterization, all original specimens of varies roughness were cleaned by washing in acetone, and then isopropanol, followed by deionized water, and again wash with isopropanol. Nitrogen gun was used to dry the surface thoroughly. Wettability and roughness investigations were undertaken on all clean surfaces. After surface characterization, these cleaned original specimens went through the boiling deposition process one by one, but each in a different nanofluid concentration. The nanofluid boiling treated surfaces again were investigated for their wettability and roughness through the same measuring procedure.

3.1 Contact angle Experiment

Contact angle measurements in this study were conducted after the coated surfaces were exposed in air for about a week after the boiling depostion process. The static contact angles of pure water on the surfaces before and after boiling were measured on a KSV CAM200 goniometer to characterize the surface wettability. The goniometer was calibrated using a 4mm diameter steel ball before the experiment. All contact angle experiments were conducted using a 5 μL pure water droplet. The size of the droplet was large enough to cover a surface area large compared to the topographical features but still small enough to neglect the effect of gravity. Dispenser control was used to locate each droplet onto the test surface softly with a consistent movement every time. Photographs of each droplet were obtained at a rate of 3 Hz, starting at the moment when droplet touched the surface. By analyzing the photographs using CAM2008, the contact angles were obtained. For each sample, the contact angles were measured at three different locations on the surface, namely center, edge and corner. The average contact angle of these three locations was used as the characteristic water contact angle of the surface. Between each measurement, the sample was rotated to allow the camera to capture surface static contact angle from different directions. At the end of the experiment for one sample, the contact angle at the center of the surface was measured again to check whether there was an influence from the environment or test procedure that affected the contact angle.

3.2 Roughness Measurement

The surface roughness was measured by a Tencor Alphastep IQ profilometer set on a vibration isolated table. The profilometer has a 5 μm radius 60^o cone stylus tip which has a maximum vertical resolution of 0.0012 nm. The surface was scanned over a 1000 μm length at a speed of 5 μm/s, and the sampling rate was 100 Hz, which results in a resolution of 0.05 μm. Very smooth uniform surface were scanned by a recipe with higher resolution: the scan length was 200 μm, the speed was 2 μm/s and the sampling rate was set to 200 Hz, so that the resolution was 0.01 μm. Each sample was scanned three times at different locations and towards varies directions on the surface. The average roughness factor was used to characterize the roughness of the surface.

The surface as received from the machine shop was anisotropic, because of the machine marks and grooves of certain pattern left by the cutting tools. Because of its anisotropic nature, both the contact angle and roughness factor are different when measured parallel and perpendicular to the surface pattern. In contrast, the polished surfaces show no anisotropic behavior under wettability and roughness experiments when observed in different directions, as given in Table 1. The difference of surface roughness is within the scan resolution and the contact angle is within the measurement uncertainty. Such isotropic surface characteristics are required for the boiling deposition process in this study.

Table 1: Roughness and contact angle of as received original sample and polished samples.

Type of surface	Roughness factor Ra (μm)			Contact angle (degree)		
	Minimum	Maximum	Difference	Minimum $\pm 2^o$	Maximum $\pm 2^o$	Difference $\pm 4^o$
Original machined surface	0.2 ±0.05	0.9 ±0.05	0.7 ±0.1	94	109	15
Rough surface	0.9 ±0.10	1.3 ±0.10	0.4 ±0.2	43	48	5
Smooth Surface	0.2 ±0.05	0.3 ±0.05	0.1 ±0.1	37	33	4
Mirror like finish	0.01 ±0.01	0.01 ±0.01	0.0 ±0.02	87	96	3

4. RESULTS AND DISCUSSIONS

4.1 Surface wettability

Pure water static contact angles for each sample were measured to characterize wettability. Data are plotted against the concentration of nanofluids in Figure 3, where the red squares are for coated surface on an ultra-smooth mirror-like substrate and the blue diamonds are for a coated surface on a smooth substrate. The contact angle for a mirror-like surface drops from 93^o (red solid line) to 31^o after the nanoparticle deposition process by dilute nanofluid of 0.01 wt % for 10 min. This indicates a dramatic enhancement of surface wettability. For a smooth surface deposited with nanoparticle coating in 0.01 wt% nanofluid, about 20 degrees of decrease in water contact angle was observed compared to untreated smooth surface (blue dash line). As the nanofluid concentration increases, the wettability of coated surfaces increases. In the best case, water wetted the surface with an angle of about 6^o within 0.3 seconds, indicating that superhydrophilicity was achieved. That is a smooth substrate boiled in 1 wt% nanofluid.

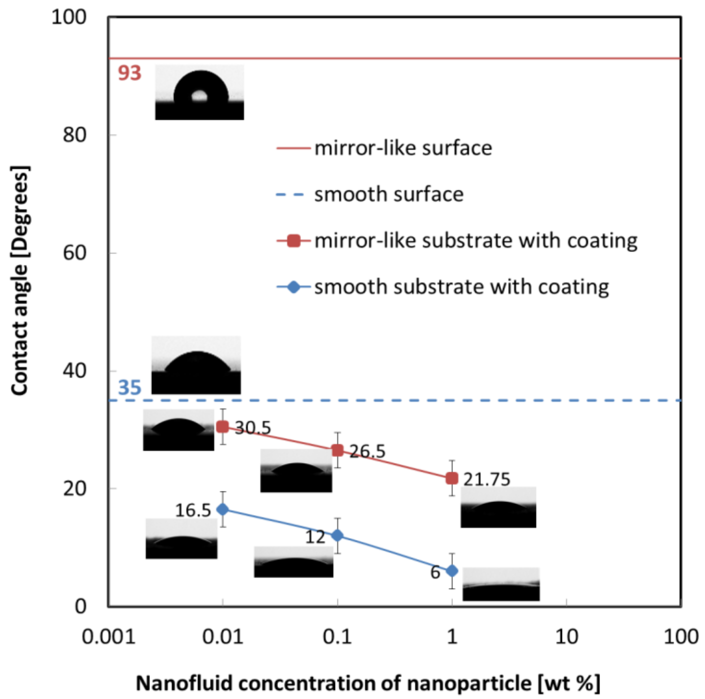


Figure 3: Contact angle before and after nanoparticle deposition process at varies nanofluid concentration.

Since aluminum oxidizes as soon as it contacts air, the surface is always covered by a thin but dense layer of aluminum oxide. When such surface is treated by depositing Al_2O_3 nanoparticles, the surface chemical composition does not change. Consequently, one can conclude that the wettability enhancement by nanofluid boiling nanoparticle deposition in this study is caused by topography modification.

4.2 Surface Topography

The roughness of each surface before boiling and after boiling deposition was measured. Figure 4, shows the surface roughness Ra (in micrometers) of surface before and after boiling deposition process against nanoparticle concentration. The roughness of clean surfaces before boiling are straight lines: blue dash line for the smooth surface that was polished by 400 grit paper, and red solid line for the mirror-like surface obtained by lapping in 12μm alumina water-based slurry. Nanoparticle deposition coated surfaces that have smooth substrate are plotted against the nanoparticle concentration in blue squares. The roughness of coated surfaces with a mirror-like finish before the boiling deposition process is in red triangles. It is observed that nanofluid boiling nanoparticle deposition treatment leads to a roughness higher than the uncoated surface. As the nanofluid nanoparticle concentration increases, roughness of the treated surface increases. This result is consistent for both the smooth substrate and mirror-like substrate. Figure 3 indicates that the roughness of the substrate has an impact on the topography of the coated surface. The smooth substrates (color blue in Figure 4) when deposited with nanoparticles, are consistently rougher than the coated surfaces with a mirror-like substrate (color red in Figure 4). As stated in section 2.3.2, all of the surfaces were boiled for 10 min at a heat flux of about 140 kW/m^2. For a particular nanofluid concentration, the only difference between each coated sample is the substrate. If the substrate plays a role, the reason is either the coating is too thin to cover the surface or the substrate itself has an impact on the coating process. Note that for boiling deposition at very low nanoparticle concentration, for instance 0.01 wt %, the mirror-like surface with coating is smoother (Ra=0.01 μm) than a 400 grit polished clean surface (Ra=0.27 μm). This implies that the deposited layer is too thin, that the surface topology determines the roughness of the coated surface directly. For the case of the rough surface (Ra=1.1 ±0.1 μm), after boiled in 0.1 wt% nanofluid, Ra increased to 1.4 ±0.1 μm, because the substrate is too rough to be covered by nanoparticle deposition in this experimental condition. It is reported that difference in surface roughness or cavity, would significantly influence the boiling process (Jones *et al.*, 2009). This nanofluid boiling nanoparticle deposition process is caused by micro-layer evaporation at the bottom of the bubble (Kwark *et al.*, 2004). It is anticipated that a large population of cavities in micro-scale would enhance the deposition process, while an ultra-smooth surface of nano-scale roughness would be inefficient in depositing nanoparticles by nucleate boiling. As a result, roughness of coated surface can be influenced by that of the substrate because the coating process is a function of the substrate surface condition.

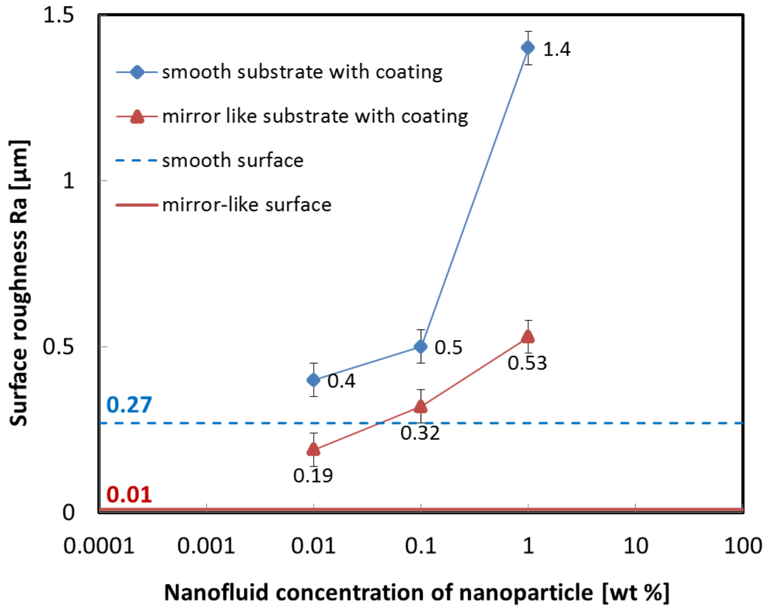
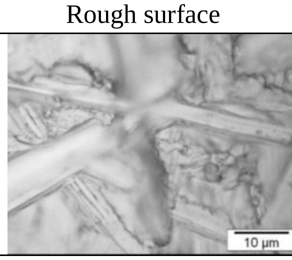
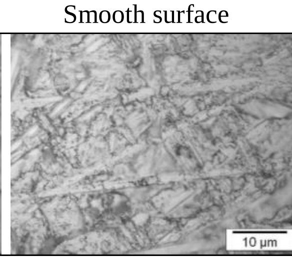
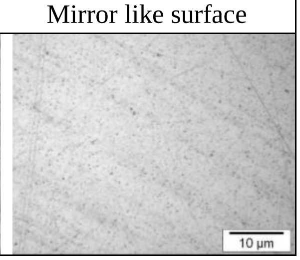
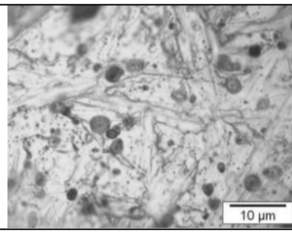
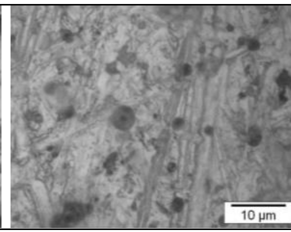
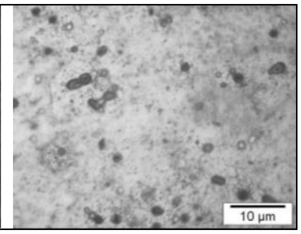


Figure 4: Surface roughness before and after nanofluid boiling nanoparticle deposition process in varies concentrations.

Table 2 shows micrographs of the substrates taken by an optical microscope before and after boiling deposition treatment in a 0.1wt% nanofluid. It can be found from Table 2 that the ultra-smooth mirror-like surface (Ra=10 nm) presents a heterogeneous layer of coating after the nanofluid boiling nanoparticle deposition process. The dark circle with a dot at the center are the results of bubble nucleation and departure induced shear force acting on the nanoporous layer, according to Huitink *et al*. (2011). For smooth substrate (Ra=0.27 µm), the micro-scale scratches tends to be covered by a layer of coating after nanoparticle deposition process, but micro-scale grooves and scratches still exist. Similar behavior is observed for a rough surface (Ra=1.1 µm). In contrast, such structures are not observed on coated surface on an ultra-smooth substrate.

Table 2: Image comparison between treated and untreated surfaces.

	Rough surface	Smooth surface	Mirror like surface
Before boiling			

After boiling deposition process in 0.1wt% nanofluid			

4.3 Nanoparticle-Layer and Roughness Impact on Wettability

According to Wenzel's model, for hydrophilic surfaces, the apparent contact angle θ* decreases when surface areal roughness factor *r* increase (indicated in by the equation). This is inconsistent with the trend shown in Figure 5.

$$\cos\theta^* = \frac{\gamma_{SV}-\gamma_{SL}}{\gamma_{LV}} r = r\cos\theta$$

However, this could not explain why the contact angle of the coated smooth substrate exhibits about 10 degrees of lower static water contact angle than the coated mirror-like substrate at similar roughness, Ra≈0.5 µm. According to Kim and Kim (2007), capillary force may become important for wetting when the surface is deposited with nanoparticles by boiling in nanofluids. From the micrographs, micro-scale scratches of arbitrary pattern are observed on the coated "smooth" substrate, but not on the coated mirror-like substrate. When a water droplet wets the coated smooth substrate, the contact line spread out like wicking and lasts a few seconds, while on the coated mirror like surface the contact line is clean and stops spreading within one second.

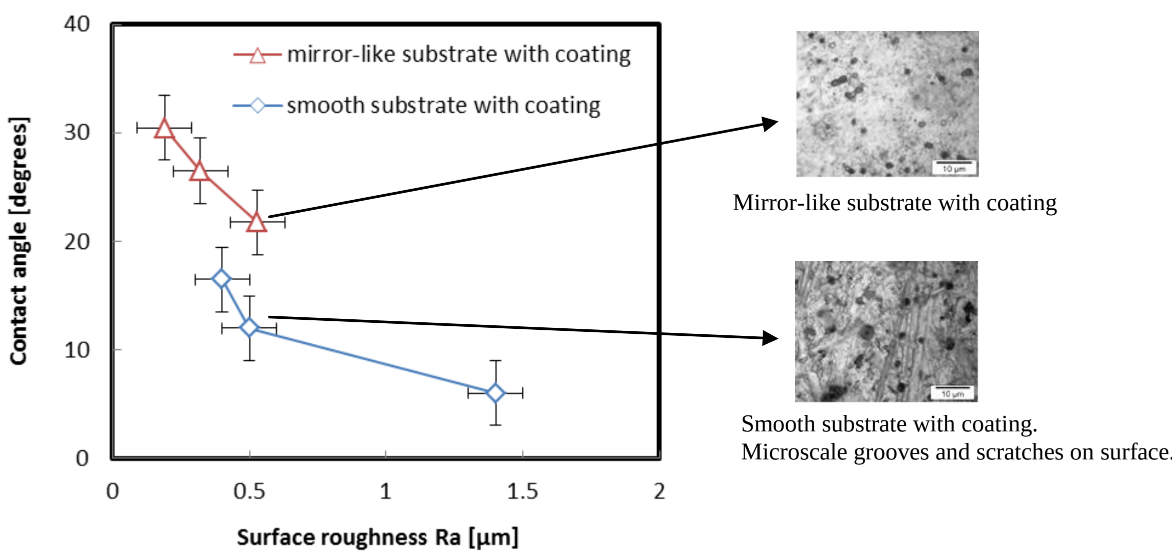


Figure 5: Contact angle vs. surface roughness of coated surface.

4.4 Potential application on heat exchangers and future work

It is important to study the potential of applying this nanofluid boiling nanoparticle deposition surface treatment in real HVAC&R applications. The first step is to investigate the topography of the fin on a heat exchanger. To start with, an aluminum flat fin and tube heat exchanger was used. A sample was cut from the flat fin and investigated by the profilometer and goniometer. The surface was observed to have lines and grooves in one direction, which is a result of the manufacturing process. When the stylus moves perpendicular to the lines, Ra= 0.13 ±0.02 µm, which is in the same scale of a 400 grit abrasive paper polished smooth surface. When scanned in parallel with the lines, Ra=0.03 ±0.02 µm, which is in the scale of a lapped ultra-smooth mirror-like substrate. The contact angle varied from $99^o \pm 2^o$ to $96^o \pm 2^o$. Sintering and other traditional metal manufacturing techniques can potentially be applied to enhance the attachment of the deposited layer to the substrate. Future work would involve nanoparticle deposition and wettability study on such surfaces.

5. CONCLUSIONS

Using the nanofluid boiling nanoparticle deposition treatment on aluminum surface of various roughnesses, a dramatic enhancement of wettability has been observed and considered to be a result of topography modification. The static contact angle of water on the coated surface is affected by the boiling condition of the nanoparticle deposition process. In the range of this study, the higher the nanofluid concentration, the lower the water contact angle on the treated surface.

It was observed that nanofluid boiling nanoparticle deposition treatment leads to a surface roughness higher than the uncoated ones. As the nanofluid concentration increases, the treated surface roughness increases. The roughness of coated surface is a function of the nanofluid concentration and substrate condition, because these factors influence the boiling process itself.

Capillary force may become important for a certain type surface, which explains why the contact angle of the coated smooth substrate exhibits about 10 degrees of lower static water contact angle than the coated mirror-like substrate at similar roughness, Ra≈0.5 μm. For a smooth substrate (Ra=0.27 μm), micro-scale grooves and scratches still exists after the nanoparticle deposition, which is not the case for those surfaces that has an ultra-smooth substrate.

An aluminum flat fin and tube heat exchanger was observed to have surface roughness within the range of this study. Future work would involve nanoparticle deposition and wettability study on such surface.

NOMENCLATURE

γ	surface tension	(N/m)
θ	Young's contact angle	(degree)
θ^*	apparent contact angle	(degree)
Ra	arithmetic average roughness	(μm)
r	areal roughness factor	(-)

Subscript

LV	liquid vapor interface
SL	solid liquid interface
SV	solid vapor interface

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