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# ANALYSIS OF THE INFLUENCE OF SURFACE ROUGHNESS AND NANOPARTICLE CONCENTRATION ON THE CONTACT ANGLE

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***Abstract.*** *Nanofluid pool boiling can modify the morphology of the heating surface and the physical properties of the base fluids, interfering directly on the vapor bubbles dynamics and on heat transfer. In this study, the effect of surface roughness and nanoparticle concentration on the contact angle was analyzed. The tests were carried out on copper surfaces with different roughness values, corresponding to a smooth surface (polished using an aluminum-oxide abrasive compound, corresponding to Ra = 0.03 µm) and a rough surface (polished using #600 emery paper, corresponding to Ra = 0.16 µm). The nanostructured surfaces were produced by maghemite nanoparticle deposition, for different mass concentration (0.29 and 2.9 g/l), via evaporation process of* $Fe_2O_3$*-deionized water nanofluid. After the deposition process, the surfaces were submitted to metallographic, roughness and wettability analysis. This paper mainly focuses on the variation of contact angle and wettability due to the microstructural change caused by the roughness and nanoparticle deposition process.*

***Keywords:*** *Nanofluids, nanostructured surfaces, contact angle, wettability, roughness.*

## 1. INTRODUCTION

In the past few years, the scientific community has been researching the effects of the promising interaction between high thermal conductivity nanoparticles and a base fluid, called "Nanofluids", first presented by Choi (1995). These nanoparticles of metals or metal oxides, diluted in a base fluid, are used to enhance the heat transfer properties of this base fluid and are currently being employed in different areas, in particular, microelectronics and micro cooling for producing compact and miniaturized equipment.

In the boiling process, nanoparticles can modify the characteristics of the heating surface and the physical properties of the base fluids, interfering directly on the mechanisms of formation and detachment of vapor bubbles. Thus, some issues must be better understood such as, the role of nanoparticle deposition on the surface wettability and how to control the deposition process, before applying this technique in micro scales devices.

Different authors (Liu et al., 2014; Heitch et al., 2014; Souza et al., 2014) applied nanostructured surfaces on nucleate boiling and all of them agree that the coating technique causes an enhancement in the critical heat flux (CHF). However, when independent studies are compared, contradicting trends are observed for the heat transfer coefficient (HTC) during pool boiling on nanostructured surfaces (Bang and Chang, 2005; Kim et al., 2004; Kim et al., 2006; Kim et al., 2007). Therefore, the effect of adding nanoparticles to a base fluid and, also, its interaction with the heating surface, on the HTC during pool boiling is still an open issue.

Both the HTC and the CHF behaviors can be attributed, mainly, to changes in the surface wettability caused by nanoparticle deposition during the nanofluid boiling process. The surface wettability is increased by the nanolayer which promotes the surface better rewetting leading to a higher CHF value. The contact angle reduction due to the nanostructure deposition partially explains the HTC behavior, since the bubble frequency and its departure diameter decreases with the nanostructure formation. Regarding HTC results, the divergences found in the open literature confirms that the boiling process is very sensitive to changes in the surface characteristics caused by nanofluids and nanostructured surfaces, especially the number and shape of micro-cavities.

The interaction between surface roughness and nanolayer formed by nanoparticles deposition also depends on nanofluids concentration (Ahamed and Hamed, 2012). According to them, nanoparticles deposition occurred at a slower rate for low nanofluids concentration (0.01 vol.%) resulting in an enhancement in the boiling HTC. This enhancement is due to the effect of thermal conductivity of nanofluids being more dominant than the effect of nanoparticle deposition on the surface.

For pool boiling on flat surfaces, many research efforts have been carried out to analyze the influence of nanostructured surfaces on boiling heat transfer. Lee et al. (2010) performed pool boiling experiments using water as working fluid and a coated surface with $Al_2O_3$, fabricated by anodizing technique. It was observed that the onset of nucleate boiling (ONB) and bubble departure diameter on coated surface was lower than that on non-coated surface and, consequently, the HTC was 20% higher for the surface with porous layer. Based on the visualization, the authors attributed the HTC improvement to an increase in the number of active nucleation sites and, also, to the combined effects of the nucleation sites interaction, the increase of vapor bubble frequency and the microlayer evaporation effect.

Souza et al. (2014) studied the effect of the deposition of $\gamma$-$Fe_2O_3$ nanoparticles (average size of 10 and 80 nm) on a horizontal copper disc during the saturated nucleate boiling of HFE7100. In their tests, it was considered two main conditions, unconfined and confined boiling. It was found a HTC increment of around 55% for the nanostructured surface with smaller nanoparticle compared to the surface without deposition. The results for the larger nanoparticles showed a decrease in the HTC of 29% compared to the case without deposition. The authors concluded that the microstructure of the nanostructured surfaces, corresponding to quantity and size of surface defects, influences the heat transfer process.

Three kinds of plain nickel-based chemical treatment surfaces and four kinds of nickel-based electrochemical treatment surfaces with nanostructure were used in the pool boiling experiment by Li et al. (2015). The effect of surface characteristic parameters, including nanoscale roughness ($R_a$), solid–liquid contact angle and effective heat transfer area ratio (defined by the authors as the ratio of the actual heat transfer area and its projected area, $r$) on the HTC were analyzed. Li et al. (2015) indicated that the HTC increases as $r$ increases and its enhancement range was basically the same with the increase range of contact angle.

Nolan et al. (2013) developed a study of HTC and CHF on nanostructure modified copper surfaces taking into account the nucleation site density and the contact angle. The nanostructured surfaces were created by an electrodeposition technique. For nanostructured surfaces, it was found an enhancement in the CHF of 142% and in the HTC of 33% compared to a finished plain copper surface. According to them, the contact angle reduction and the active nucleation site increase on the tested surfaces are responsible for CHF and HTC enhancement.

Heitch et al. (2014) analyzed the effect of nanostructured surfaces on the nucleate boiling of water, at saturation temperature and at atmospheric pressure. The nanostructured surface consisted of a Constantan tape nanostructured with molybdenum (by sputtering process) and maghemite (by nanofluid evaporation technique). The results were compared with experimental data for surfaces with different roughness and without nanoparticle deposition. It was observed an increase in the surface wettability for the nanostructured surface, mainly for maghemite deposition, and consequently an increase the CHF. An increase in the HTC occurred only for high heat fluxes values. The rough surfaces showed a highly hydrophobic behavior, by measurement of the apparent contact angle, and the HTC enhancement compared to the others surfaces tested was explained by the fact that the nucleation cavities are not flooded with the liquid, allowing the vapor trapping which increases the number of active boiling centers on the surface and, consequently, to an increase in heat transfer.

The main goal of this study is to analyze the effect of surface roughness and nanoparticle concentration on the contact angle of water. After the deposition process by nanofluid evaporation technique, the surfaces were submitted to metallographic, roughness and wettability analysis. The main focus is the variation of contact angle and wettability due to the microstructural change caused by the roughness and nanoparticle deposition process.

## 2. MATERIALS AND METHODS

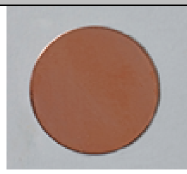
### 2.1 Test surface preparation

Table 1 shows the six types of copper discs, four of them being nanostructured, analyzed in this study. These surfaces consisted of one smooth surface, one rough surface, two smooth surfaces with nanoparticle deposition (at low and high concentration) and two rough surfaces with nanoparticle deposition (at low and high concentration). The nanostructures were produced by the deposition of maghemite nanoparticles via evaporation process of $Fe_2O_3$-deionized water nanofluid.

All the surfaces were initially polished using an aluminum-oxide abrasive compound (smooth surface) or #600 emery paper (rough surface), corresponding to a roughness, $R_a$, of 0.05 μm and 0.23 μm, respectively. Prior to the deposition processes, all surfaces were cleaned with 10% NaOH water solution and then cleaned with acetone.

The maghemite ($\gamma$ –$Fe_2O_3$ with average particle size of 10 nm) nanoparticles were synthesized following Massart's method (Massart, 1982) through the precipitation of $Fe^{2+}$ and $Fe^{3+}$ salts in alkaline medium and dispersed in water. The $Fe_2O_3$-deionized water nanofluid was supplied by Prof. Maria de Fátima da Silva from NFA/ Instituto de Física/ Universidade de Brasília.

Table 1 – Description of heating copper discs tested in this study.

| Surfaces | Roughness, $R_a$ (μm) | Description | Surface Image |
|---|---|---|---|
| Smooth surface (SS) | 0.05 | Smooth surface without deposition. |  |
| Rough surface (RS) | 0.23 | Rough surface without deposition. |  |
| SS-Low concentration (SS-LC) | 0.09 | Smooth surface with maghemite deposition via evaporation process. 0.29 g/l nanofluid concentration. |  |
| SS-High concentration (SS-HC) | 0.21 | Smooth surface with maghemite deposition via evaporation process. 2.9 g/l nanofluid concentration. |  |
| RS-Low concentration (RS-LC) | 0.30 | Rough surface with maghemite deposition via evaporation process. 0.29 g/l nanofluid concentration. |  |
| RS-High concentration (RS-HC) | 0.41 | Smooth surface with maghemite deposition via evaporation process. 2.9 g/l nanofluid concentration. |  |

The nanostructured surfaces were prepared with two different volumetric concentrations of 0.29 g/l and 2.9 g/l, in order to verify the influence of nanolayer thickness on the wettability. In each case, a layer of suspension was firstly deposited onto the copper disc (20 mm diameter) using a syringe. The surface was connected to a power source, which allowed surface heating and consequent evaporation of the base fluid, leaving only the deposited nanoparticles. This procedure was similar to that used by Heitch et al. (2014) and Souza et al. (2014).

## 2.2 Characterization of test surfaces

The characterization of the samples was carried out using the following different techniques:

- Scanning electron microscopy (SEM) to obtain structural and chemical information. This procedure was performed using a EVO LS15 Zeiss® scanning electron microscope;

- Surftest SJ 301 model to obtain the average roughness ($R_a$) for each surface, with the same scanning area for all samples;
- Atomic force microscopy (AFM), model SPM-9600 with tapping mode, in order to obtain the topographic images of the surfaces.
- The contact angles were measured by analysis of pictures of a sessile droplet of water. The pictures were analyzed through two methods, implemented as a Java plug-in for the ImageJ software (freely available at Biomedical Imaging Group – EPFL): DropSnake based on B-spline snake to shape the drop and, LBADSA based on the fitting of the Young-Laplace equation to the image data. The contact angle measurements were performed using the experimental apparatus shown in Fig. 1, developed in the Heat Transfer and Micro-Fluidics Research Group at EESC-USP, Brazil.

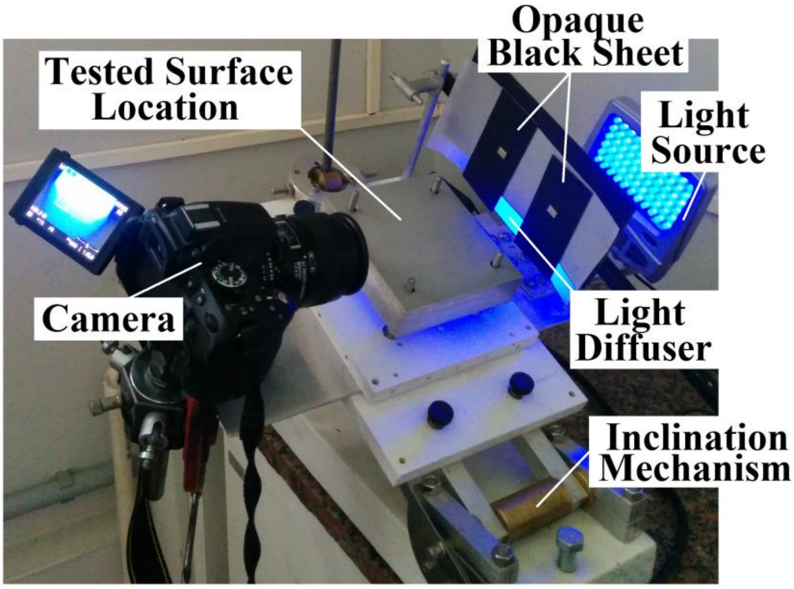
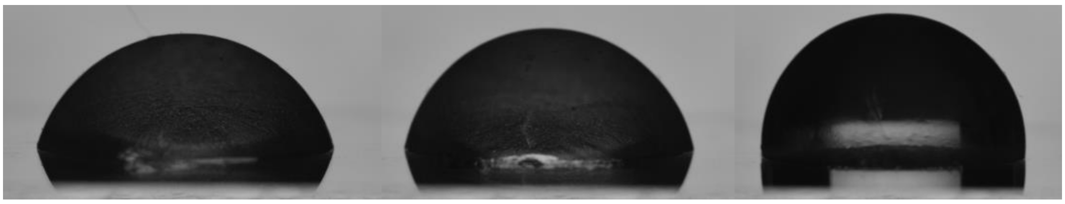



Figure 1 - Experimental apparatus to measure contact angle of droplets and instruments.

The apparatus consists basically of the test surface, a camera, an adjustable blue LED light source, pipettes, sheet of tracing (light diffuser) and opaque black papers (for less reflections of the droplet), and an aluminum block in which was fixed the test surface (Netto et al., 2013). It was possible to interchange the test surface and alter its inclination through a worm screw connected to a flywheel. The measurements were repeated at two different randomly selected locations on the surfaces.

The method developed by Netto et al. (2013) and used in the present study was validated by comparison with angles previously known. Semi-spherical bodies made of glass were used, with shapes similar to the 2D image of the droplet, positioned on the surface, Fig. 2.



(a) (b) (c)

Figure 2 - Semi-spherical glass proof bodies positioned on the experimental apparatus to validate contact angle measurements.

After taking pictures of each semi-spherical proof body positioned on the test surface, three different operators analyzed each image and measured its contact angle by using the plug-in for the ImageJ software. Table 2 compares the contact angle based on the measurements and the respective estimated value from geometrical relations. Also, it shows the mean absolute error (MAE) of the measurement that confirms that this method of picture analysis of a sessile droplet is accurate.

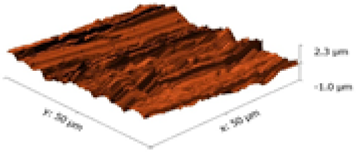
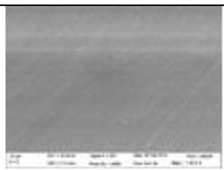
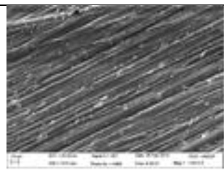
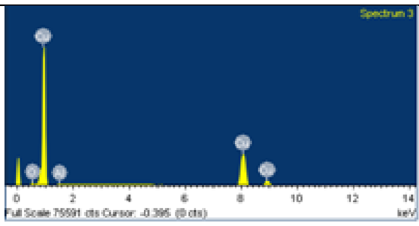
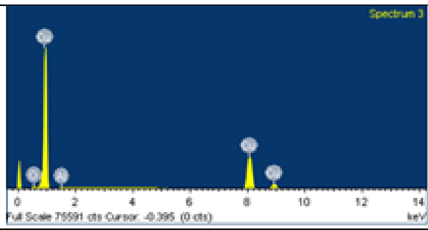
Table 2 - Results of the contact angle measured on three semi-spherical glass proof bodies simulating droplets.

| | **Semi-cylinder (a)** | **Semi-cylinder (b)** | **Semi-cylinder (c)** |
|---|---|---|---|
| Operator 1 | 74.074 | 76.800 | 93.837 |
| Operator 2 | 74.512 | 76.610 | 93.986 |
| Operator 3 | 73.509 | 76.435 | 93.879 |
| Estimated value | 73.827 | 77.916 | 94.885 |
| $MAE = \frac{1}{N}\sum_{i=1}^{N}\left\|\frac{\theta_{calc}-\theta_{exp}}{\theta_{calc}}\right\|$ | 0.3% | 1.7% | 1.0% |

## 3. RESULTS

Table 3 shows the characterization of the smooth (SS) and rough surfaces (RS). The AFM and SEM images show a few topographic irregularities for SS compared to the RS, as showed by distance peak-to-valley. The chemical analysis showed the predominance of Cu with aluminum oxide as residual material. As described previously, the surface roughness, $R_a$, for SS and RS was 0.05 µm and 0.23 µm, respectively.

Table 3 - Results for the smooth and rough surface characterization.

| **Techniques** | **SS** | **RS** |
|---|---|---|
| AFM |  |  |
| SEM |  |  |
| $R_a$ | 0.05 µm | 0.23 µm |
| EDX |  |  |

The results for the characterization of the maghemite nanostructured surfaces, obtained by evaporation process of $\gamma$-$Fe_2O_3$-water nanofluid with different volumetric concentrations, show microstructural aspects with different grain size and porosity (see Tabs. 4 and 5). For all nanostructured surfaces, the chemical analysis shows the presence of Fe and O.

The surface roughness increases for all nanostructured samples compared to those without nanoparticle deposition, both for smooth and rough surfaces.

According to Ahmed and Hamed (2012), the interaction between surface roughness and nanolayer formed by nanoparticles deposition depends on nanofluid concentration. The SEM images show that as the volumetric concentration increases, surface deposition of the nanoparticles thickened and more micro-sized structures were formed on the surface. The SS-LC has only few nanoparticles adhered on the copper disc after the nanofluid evaporation process. However, the SS-HC presents a homogeneous coating layer with micro-scale grooves. This behavior is also observed for RS-LC compared to the RS-HC, where the $R_a$ is 0.41 µm.

The effect of nanoparticle deposition on the contact angle and, consequently, on the wettability are shown in Tab. 6. The wettability test provides the apparent static ($\theta$*), receding ($\theta_r$) and advancing ($\theta_a$) contact angles. To perform advancing and receding angle measurements, the authors tilted the sample until the drop of water begins to roll downhill.

It is worth mentioning that the size of the droplet covered a large surface area but it was still small enough to neglect the effect of gravity.

Table 4 – Results for the smooth and rough surface with nanoparticle deposition (via evaporation process of 0.29 g/l nanofluid concentration).

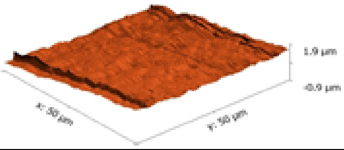
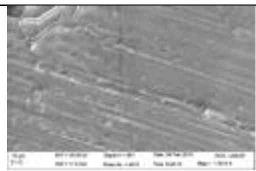
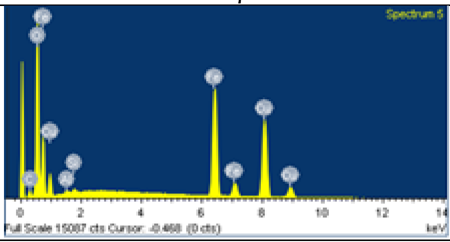
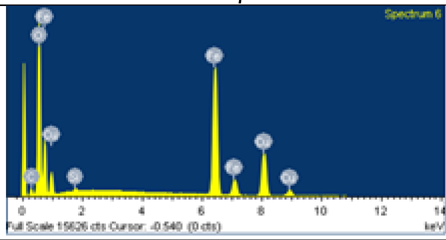
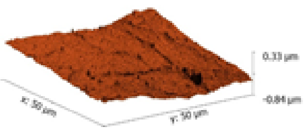
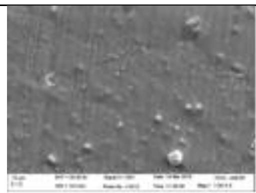
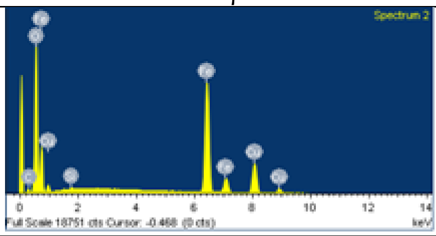
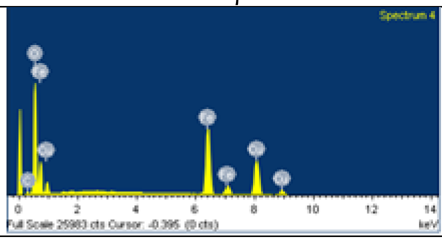
| **Techniques** | **SS-LC** | **RS-LC** |
|---|---|---|
| AFM |  |  |
| SEM |  |  |
| $R_a$ | 0.09 µm | 0.30 µm |
| EDX |  |  |

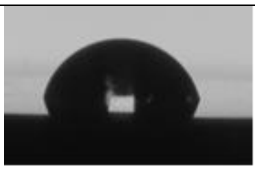
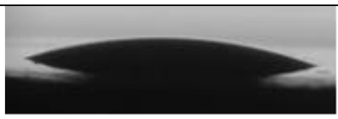
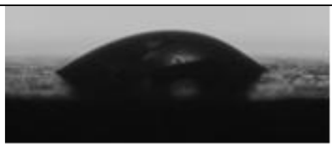
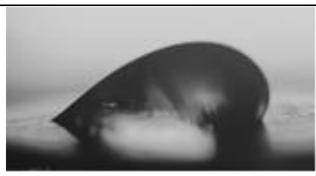
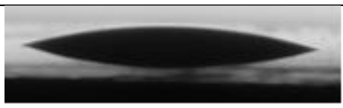
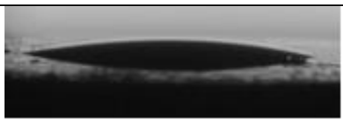
Table 5 – Results for the smooth and rough surface with nanoparticle deposition (via evaporation process of 2.9 g/l nanofluid concentration).

| **Techniques** | **SS-HC** | **RS-HC** |
|---|---|---|
| AFM |  |  |
| SEM |  |  |
| $R_a$ | 0.21 µm | 0.41 µm |
| EDX |  |  |

According to static contact angle measurements, the contact angle decreases with increasing the nanofluid volumetric concentration, for both smooth and rough surfaces, indicating an increase in the wettability.

The SS sample show a hydrophobic behavior, i.e., with a static contact angle higher than 90º. All surfaces with nanoparticle deposition show a wetting behavior and, at high nanofluid concentration, water wets the surface with an angle lower than 10° indicating that superhydrophilicity is achieved. For theses cases, it was not possible to observe the contact angle hysteresis.

Table 6 – Wettability results for all surfaces tested in this study.

| Surface | Static | Advancing / Receding |
|---|---|---|
| SS | 95°  |  93° / 23° |
| RS |  75° |  80° / 4° |
| SS-LC |  15° | <10° Completely Wetting |
| RS-LC |  39° |  114° / 32° |
| SS-HC |  < 10° | <10° Completely Wetting |
| RS-HC |  < 10° | <10° Completely Wetting |

## 4. CONCLUSIONS

An analysis on the effect of surface roughness and nanoparticle concentration on the contact angle of water was carried out. A metallographic, roughness and wettability characterization, after the deposition process by nanofluid evaporation technique, was presented. The main results are the following:

- ✓ The roughness of coated surface is a function of the nanofluid concentration and original surface condition. As the volumetric concentration increases the surface roughness ($R_a$) also increases;
- ✓ Nanofluid evaporation technique on copper surfaces with different roughness causes a noticeable increase in the wettability, observed by the decrease in the static contact angle. In the present study, the higher nanofluid concentration, the lower the contact angle of water on the coated surface. This behavior could be due to the topography modification;
- ✓ Nanolayer formed by nanoparticles deposition affects both surface roughness and surface wettability. It was observed that the nanolayer has a higher influence on the surface wettability than the original surface roughness.

Based on the metallographic, roughness and wettability results, as well as the experimental results for pool boiling using the surfaces tested in this study, it will be possible to analyze which mechanism has more impact on the heat transfer coefficient.

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