



## Enhancing the onset of pool boiling by wettability modification on nanometrically smooth surfaces<sup>☆</sup>

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### ABSTRACT

Pool boiling experiments are performed with degassed water on highly smooth surfaces of two different wettabilities: hydrophilic and hydrophobic cases. Boiling curves and visual observations on the boiling have been performed. The onset of nucleate boiling (ONB) has been measured and the influence of the wettability has been quantified. As the inherent mean roughness of the glass substrates was lower than one nanometer it was possible to show the sole effect of the wettability. No hysteresis in the boiling curve was observed for both cases. The ONB was observed after 3.5 °C superheat on the hydrophobic case and the heat transfer coefficient (HTC) changed suddenly from the one of a convection regime (1.5 kW/m<sup>2</sup> K) to the one of a nucleate boiling regime (4 kW/m<sup>2</sup> K). On the contrary for the hydrophilic case, despite superheat above 37 °C and presence of boiling, the HTC was kept as the one of the convection regime.

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### 1. Introduction

Boiling is a natural phenomenon that occurs in daily life but also in many industrial processes. When the heating is supplied by a solid surface, both local liquid and solid temperatures are function of the local heat transfer, which, at the starting of boiling (the so-called “boiling onset”) depends on many parameters, among which the surface characteristics, such as the surface roughness and material properties. The temperature at which the boiling appears is always higher than the saturation temperature of the liquid: a “superheat” is needed. Among the surface-related key parameters two are of primary importance: the surface topographical features and its wettability [1–3]. Changing the solid surface to study the boiling has the consequence to modify these two parameters, that’s why, up to now there is some debate on how these two parameters are acting independently of each other

Looking at the surface topography, it has been recently shown that single cavities of the order of 10 microns large on silicon wafers can trigger the boiling [4–6]. The shape of the cavity is also of primary importance [7–9] for the nucleation. Several works have shown using highly smooth surfaces that nanometric features influence nucleation [10–13]. Bourdon and coworkers [14] showed that differences of roughness on highly polished bronze surfaces down to few nanometers in amplitude modify the superheat needed to get boiling.

On the other side, various authors have also shown the influence of wettability on the onset of boiling [2,7,15]. Many studies on boiling are performed with changes either in the roughness or the topography of the surface [16–20]. Only recently authors have used glass or silicon wafer surfaces with self assembling monolayer over the substrate in order to modify the wettability of a solid without affecting its topography (a monolayer is usually of the order of one or two nanometers height) in transient events [21,22] or stationary conditions [23,24,14].

In the present study we perform experiments of incipient pool boiling of degassed water on glass substrates presenting subnanometer roughness but different wettability. Surfaces with various wettability conditions are used in order to quantify its effect independently of topographical effects. The boiling curve is determined and the heat transfer coefficient (HTC) calculated. The wettability effect on this parameter is estimated.

### 2. Experimental procedure

#### 2.1. Materials

With the goal to measure the isolated effect of the wettability on the onset of nucleate boiling (ONB), possible roughness contribution has been minimized as much as possible. For that, we used glass surfaces (float glass; Oedenkoven Fr S.A.) because of its small inherent roughness without necessity to polish it. Moreover glass material is strong enough to prevent any damage on the surface during boiling in contrast to metallic surfaces [14]. Milli-Q water was used during the boiling experiments. OTS (Octadecyltrichlorosilane; Sigma-Aldrich, Germany)

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was used to add a hydrophobic monolayer to the glass substrate. All chemicals were used without further purification.

## 2.2. Experimental setup

We measure the superheat needed to have the onset of boiling on both hydrophilic and hydrophobic surfaces. A ceramic cartridge (Acim Jouanin H6.5X32X175) of 175 Watts is placed in a housing made in copper. This cartridge is used to heat the surface and reach boiling on it. A spring is placed between the bottom of the copper housing and the bottom of the box to press the heater with a constant strength on the surface. An internal heater (80 W) is placed in the boiling chamber to heat up the water. Two external heating tapes are placed on the walls of the chamber to compensate the thermal leakages. The boiling chamber is made in alumina and the heater is surrounded by Teflon. A heat flux meter with 3 embedded T-thermocouples (Captec, France) is placed between the copper and the tested surface. 2 additional K-thermocouples are placed in the water and connected to a proportional–integral–derivative (PID) controller to check the water temperature. A pressure gauge is connected to the chamber to measure its pressure. The boiling chamber is connected to bellows in order to modify the internal pressure. All the thermocouples and the pressure gauge are connected to a computer using a data acquisition system (Agilent A34970A data acquisition/switch unit, USA). Fig. 1 shows a scheme of the experimental setup.

## 2.3. Preparation of the surfaces

All the experiments are carried on using glass surfaces. The treatment to add the hydrophobic layer on the glass and modify its wettability is chemical grafting using SAM [25,26,23] of OTS. The method to graft this monolayer is summarized hereafter. All the

glass surfaces were preliminary cleaned using  $\text{H}_2\text{O}_2/\text{H}_2\text{SO}_4$  (30/70) mixture then rinsed copiously with Milli-Q water and dried with  $\text{N}_2$ . A 30 minute exposure in a UV/ $\text{O}_3$  oven is then applied to the surface. After this cleaning method, the surface is completely hydrophilic ( $0^\circ$  contact angle).

## 2.4. Grafting method

After cleaning the surface as described above, the glass surfaces are immersed in 3 mM of OTS in  $\text{CCL}_4$ /hexadecane mixture (30/70 vol/vol) for 12 hours in a clean atmosphere of nitrogen where humidity is kept at 8 %RH and temperature at  $12^\circ\text{C}$  [27].

To remove possible deposited but ungrafted chemical, the surfaces are then immersed twice during 5 minutes in chloroform in an ultrasonic bath and dried with  $\text{N}_2$ .

## 2.5. Experimental procedure

The chamber is vacuumed down to 30 mbar before adding water in order to remove air and adsorbed gases inside the box. When the chamber is filled by degassed water, the internal heater is switched on in order to bring the water at its saturation temperature. A pressure gauge is connected to the tank to provide us the value of the pressure inside the chamber and thus of the correct saturation temperature of the water. When the saturation temperature is reached, the internal heater is switched off. External heaters are connected to a PID controller to compensate thermal leakage and to maintain the saturation temperature in the chamber. A heat flux meter with 3 T-thermocouples is placed between the copper housing of the heater and the glass surface (see Fig. 1). This sensor provides us a direct measure of the heat flux going to the tested surface.

Once the saturation temperature is reached, the cartridge heater is switched on to bring the copper housing at the saturated temperature

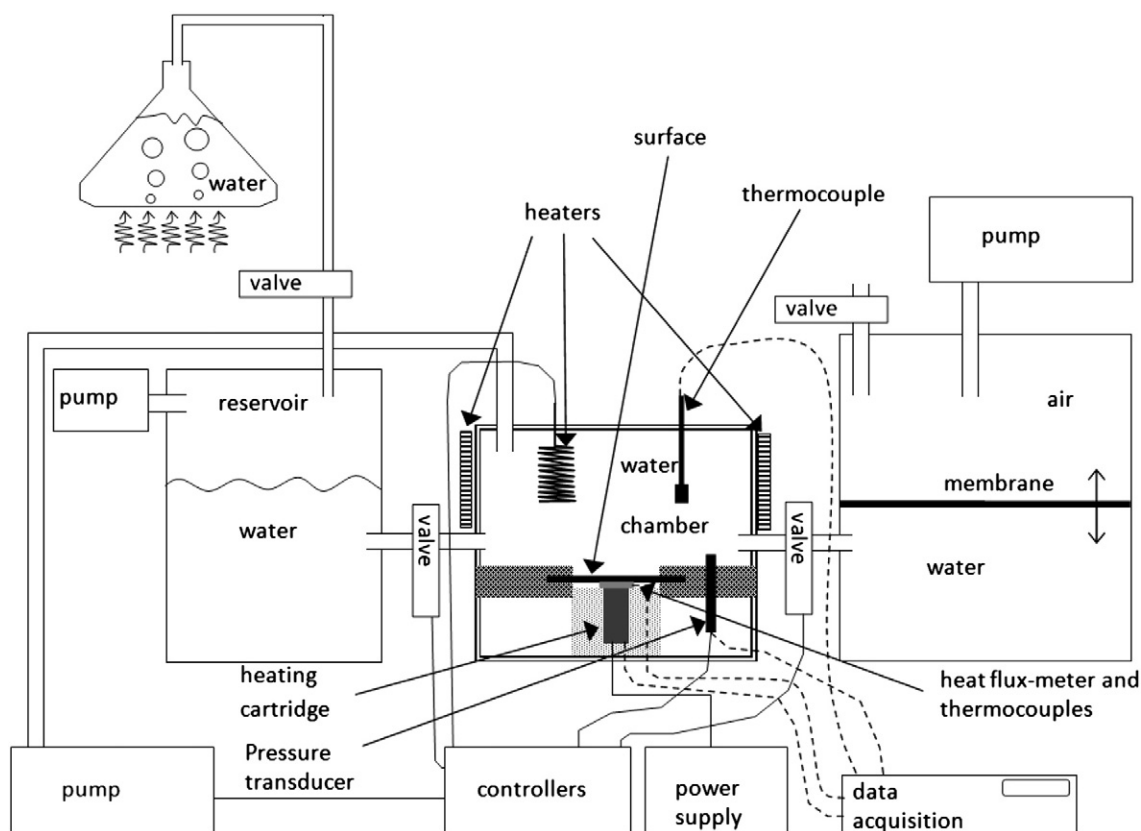


Fig. 1. Experimental setup.

of water. Then we increase by a tiny amount the power of the cartridge and we measure the temperatures and flux. Increasing step by step the power to the cartridge enables to build the boiling curve and the HTC.

### 3. Results and discussion

#### 3.1. Surface characterization

The roughness of the surfaces was measured using confocal microscopy and interferometry (Sensofar PLu Neox, Spain). The results are presented in Fig. 2 and Table 1. The mean roughness, here quantified by  $S_a$  and  $S_q$ , is one order of magnitude lower compared with highly polished metallic surfaces [14]. We have already shown [14] that the key parameter for the incipient boiling is not the mean roughness, but either the peak-to-valley depth either a parameter able to quantify the holes present on the surface (the parameter called “mean valley depth” is another good example). For this parameter also, the glass surface is an order of magnitude smoother than polished metallic surfaces. Nevertheless, while very smooth, the glass substrate presents defects and holes (see Fig. 2). On the bearing curve it is possible to see that 0.2% of the surface area presents holes as large as 16 nanometers depth. These holes are probably nucleation sites for the boiling.

The monolayer that is added to the glass substrate to modify its wettability doesn't affect the roughness parameters.

#### 3.2. Wettability effects

Pool boiling experiments are carried on using hydrophobic (grafted) and hydrophilic (ungrafted) surfaces. The contact angle measurements are shown in Table 2.

The grafted and ungrafted cases present a clear difference in the boiling curve (Fig. 3, top), since in the hydrophobic case, the first bubbles appear at a superheat of 3.5 °C and a significant change in the shape of the boiling curve occurs at a superheat of 4 °C. The slope of the boiling curve is highly increased because of the heat removal by the bubbles. In Fig. 3 (bottom) a visualization of the bubble generation on the surface is given for both cases. At the onset of the boiling only few bubbles appears at a very low frequency (in the range of the Hertz) for both cases. But as soon as the flux is increased the frequency increases drastically for the hydrophobic case: after less than one degree superheat above the onset of boiling fully developed boiling is visualized and bubble are covering the whole heated surface. On the contrary for the hydrophilic case the onset appears much later (at 22 °C of superheat). While increasing the flux the frequency increases until reach approximately 50 Hz on an area of 20 mm diameter for the maximum superheat we could reach (37 °C).

**Table 1**

Most common roughness parameters for our glass substrates.

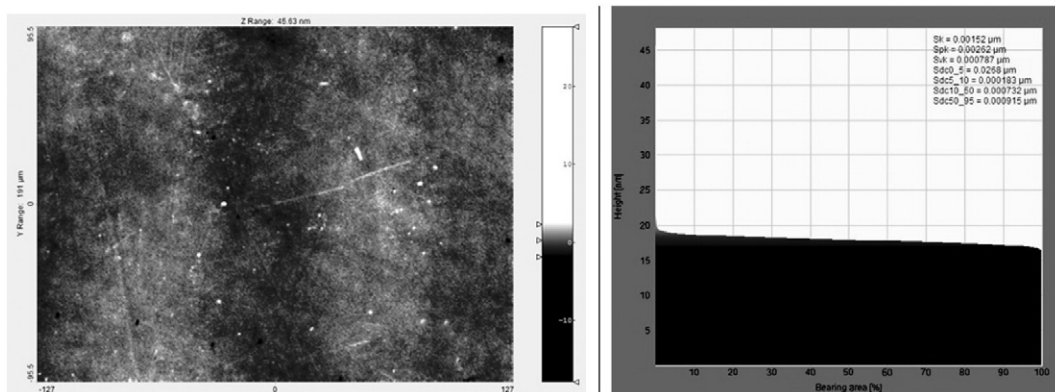
Roughness parameters	Values (nm)
$S_a$	0.46
$S_q$	0.61
$S_z$	45.6
$S_v$	17.9

In the hydrophobic case bubbles coalesce to form bigger bubbles that afterwards detach due to buoyancy. On the other hand, for the hydrophilic case bubbles are smaller and their density on the surface is lower. In the boiling curve of the hydrophilic case it was not possible to visualize any change of slope. That is probably due to the fact that it was not possible to reach the fully developed boiling state as seen on the hydrophobic case. Due to the limits of our experimental apparatus (i.e. for a superheat of 35 °C over the surface, the temperature of the heater cartridge was at 195 °C overshooting dangerously the maximum working temperature of the heat flux meter), we had to limit the imposed flux. This explains why fully developed boiling was not reached. Data uncertainties on the boiling curve increase at high power (Fig. 3).

Furthermore, on the hydrophilic surface, the bubbles are highly spherical and detach rapidly from the surface. In the hydrophobic case, bubbles are elongated and the departure time is longer. Each experiment was repeated 4 times to estimate the uncertainties of the temperature and heat flux measurements.

The hysteresis of the boiling curves was also measured and no significant hysteresis occurs in both cases (Fig. 4). The most logical reason of the lack of hysteresis is probably the fact that there is almost no nucleation sites related to cavities or specific topographical features on the surface as shown in Fig. 2.

The HTC are approximated by deriving the fit (hydrophilic:  $R^2 = 0.992$ ; hydrophobic, convective part:  $R^2 = 0.96$ ; hydrophobic, boiling part:  $R^2 = 0.999$ ) of the flux on the boiling curves. The HTC behavior (Fig. 5) shows that for ungrafted surfaces an effective ONB is not reached even with a superheat of almost 40 °C. In Fig. 5 a graphical scheme of the experimental results is given in order to represent in a clearer and evident way the physics related to the change of heat transfer regime. The grey part represents the standard deviation of the data. It is logical to have a large error bar for low heat flux as both heat flux and superheat are low. The presence of bubbles on the surface, visually detected by the camera (see supplementary material A) is usually linked to the ONB. The present data shows that it is not leading to a typical variation of the HTCs, which remain as for the single phase convection. This is particularly evident on the ungrafted or hydrophilic surface where the bubbles appear despite a HTC typical for convection (around 1.5 kW/m<sup>2</sup>K). While for the



**Fig. 2.** Topography of the glass surface at a scale of 254 μm × 191 μm (left) and the bearing curve relative this surface (right).

**Table 2**  
Wettability properties of the glass substrates used.

	Hydrophobic case		Hydrophilic case	
	$\Theta_{ad.} (^{\circ})$	$\Theta_{rec.} (^{\circ})$	$\Theta_{ad.} (^{\circ})$	$\Theta_{rec.} (^{\circ})$
Before boiling	113.9	94.7	0	0
After boiling	107.8	90.2	35.9	9.9

hydrophobic case the change of HTC is almost immediate in terms of temperature (within one degree), on the hydrophilic case the HTC remains the one of convection even with bubbles having appeared 15 °C before (that is to say between 22 °C and 37 °C of superheat). For the grafted surfaces the ONB is evident and it is already present for a superheat of 3.5 °C, confirming the very high effect of wettability on the boiling nucleation, without microscale cavities. The value of the HTCs for the grafted surfaces appears within the standard value range, reaching with water a value of about 4 kW/m<sup>2</sup>K.

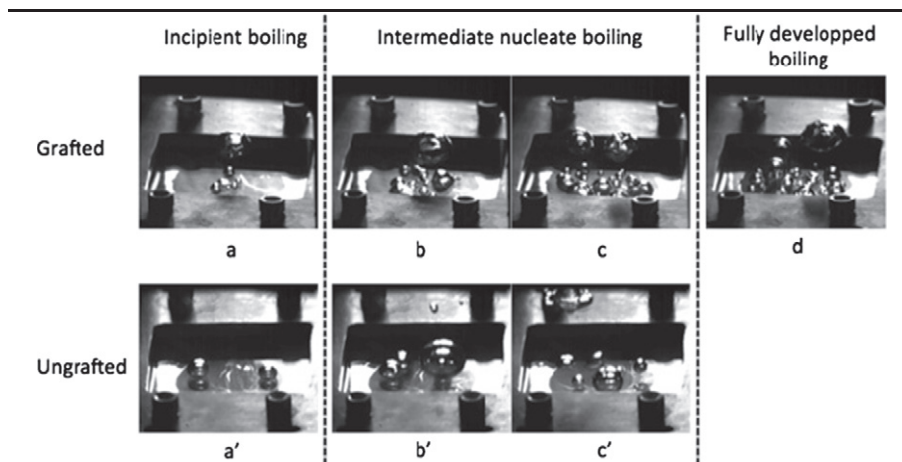
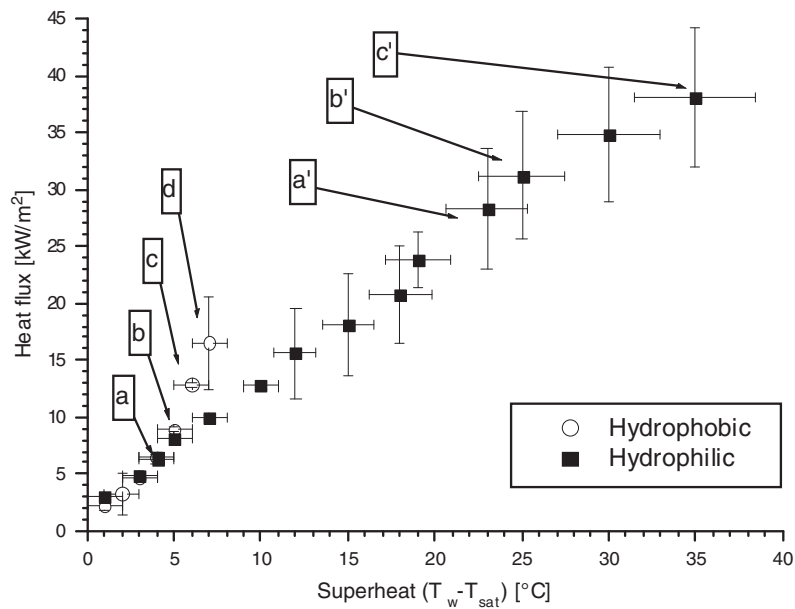
**4. Conclusions**

Pool boiling experiments of water were performed on highly smooth surfaces which are glass substrates. The wettability has been changed grafting a hydrophobic monolayer onto it, this means

without changing significantly the topography, i.e. the smoothness of the surface. Topography measurements show that almost no cavities are present on the surface. In this way the sole effect of the wettability is shown by visualization of the boiling and measurement of the boiling curves. On these highly smooth surface no hysteresis were found. The reduction of the wettability induced a reduction of the onset of boiling: bubbles appear on the surface with lower superheat (3.5 °C) on hydrophobic surface than on hydrophilic one (22 °C). The heat transfer regime was analyzed by calculation of the HTC. On the hydrophobic case when boiling appears the HTC passes from the value of 1.5 kW/m<sup>2</sup> K which is typical of convection to 4 kW/m<sup>2</sup> K which is typical for boiling. On the contrary for the hydrophilic case even with a superheat of 37 °C and presence of low boiling (50 Hz on a zone 20 mm diameter) the HTC is the one of convection and no change in its value was detected.

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**Fig. 3.** Boiling curve for hydrophilic (ungrafted) and hydrophobic (grafted) glass surfaces (top) and pictures of the boiling for both cases at different stages of the boiling.



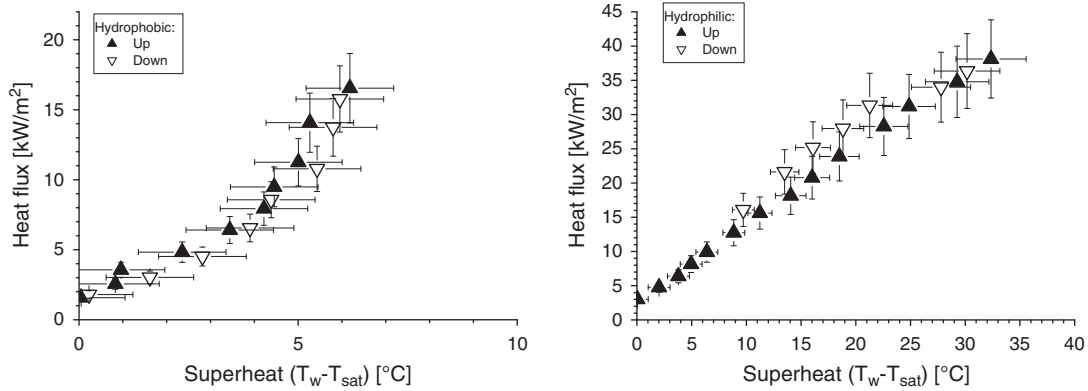


Fig. 4. Hysteresis in the boiling curve for both cases: hydrophobic (left) and hydrophilic (right).

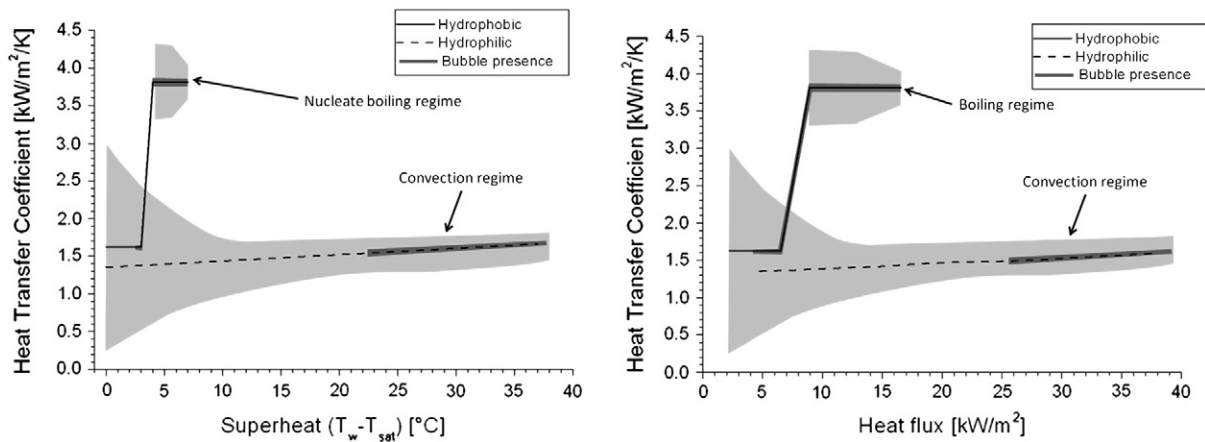


Fig. 5. The heat transfer coefficient (HTC) extrapolated from the boiling curves versus the superheat (left) and the heat flux (right) for both cases. The grey zone is providing an estimation of the error bar.

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