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# Article Unexpected frictional behavior of laser-textured hydrophobic surfaces

Hiba Jendoubi<sup>1</sup>, Olga Smerdova<sup>1</sup> and Noël Brunetière<sup>1,\*</sup>

- <sup>1</sup> Institut Pprime; CNRS, Université de Poitiers, ISAE-ENSMA, Poitiers, France
- \* Correspondence: noel.brunetiere@univ-poitiers; Tel.: +33-549-496-531
- 1 Abstract: Hydrophobic surfaces can allow a liquid to slip over the surface and can thus reduce
- <sup>2</sup> friction in lubricated contact working in a full film regime. Theory supports that the amount of slip
- can be increased if super-hydrophobic surfaces that are composed of a textured low surface energy
- <sup>4</sup> material are used. In this work, polytetrafluoroethylene (PTFE) polymer samples were textured
- 5 with a femto second laser to create super-hydrophobic surfaces by machining a hexagonal network
- of small circular holes with 10 and 20  $\mu$ m lattice sides. The frictional behavior of these surfaces
- 7 was compared to the smooth PTFE samples. Surprisingly, the textured surfaces revealed higher
- friction coefficients than the smooth surfaces. This higher friction can be explained by a change of
- wetting regime due to high pressure in fluid and a possible generation of vortices in the cavities.

10 Keywords: Surface texture; Hydrophobic surface; Friction; Lubrication

# 11 1. Introduction

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According to Holmberg and Erdemir[1], about 23% of total global energy consumption is used to fight friction and wear. It is thus essential to propose solutions to reduce friction in machine components. In the case of lubricated contacts, surface texturing has proven to be efficient in several configurations to reduce friction [2,3]. For instance, by creating a network of shallow grooves on the sliding surface, the friction and temperature in a mechanical seal can be reduced by 50 % [4,5].

Another solution to reduce friction was proposed by Spikes [6,7], which uses surfaces on which the lubricant fluid can slip, meaning that there is no adherence between the fluid and the surface. This slip is characterized by the slip length, which is defined as the distance between the surface and the point at which the relative fluid velocity would vanish. Salant and Fortier [8] showed theoretically that performance can be further improved by combining slip and and no-slip areas on the surface. The slip area lets more fluid enter the contact, as if the film was thicker. The heterogeneous slip/no slip bearing can thus be compared to a pocket or a step bearing. Several authors have since performed simulations to demonstrate the interest for this type of slip surface [9,10]. It is thus of importance to produce surfaces with slip that can be used in lubricated applications.

In their experimental work, Zhu and Granick [11] demonstrated that fluid can slip on a poorly wetted surface when its roughness height is lower than 6 nm. For lower roughness level, the level of slip, which is characterized by the slip length, is controlled by chemical interactions between the fluid and the surface; as early shown by Schnell [12]. In the case of confined flows, roughness can allow slip of the fluid [13]. Using molecular dynamics, Huang et al. [14] found that the slip length is strongly correlated to the contact angle formed between the solid surface and a sessile water drop. The contact angle characterizes by a simple measurement the intensity of chemical interactions of water with the surface. The slip length is higher when the contact angle is higher. In both studies [11,14], the value of the slip length is a few tens of nanometers. According to theoretical work [6–10], these surfaces could provide lower friction level when used

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Copyright: © 2021 by the authors. Submitted to *Lubricants* for possible open access publication under the terms and conditions of the Creative Commons Attribution (CC BY) licenses (https://creativecommons.org/licenses/<sup>38</sup> by/4.0/). materials with different contact angles and showed that higher contact angle values lead

- to a behavior corresponding to slip of the lubricant on the coated surface. In the case
- of limit lubrication regime, the combination of a hydrophic and a hydrobobic surfaces
- <sup>47</sup> in a water lubricated contact is also efficient to reduce friction thanks to an hydration
- 48 mechanism [18,19]. These very low film thicknesses lubrication corresponding to high
  - loading are however not considered in the present work.

A much higher contact angle can be found on natural materials that are qualified as 50 super-hydrophobic, such as the lotus leaf [20]. A texture is combined with a low surface 51 energy material to give a Cassie Baxter state where the water drop lies on the top of 52 the texture asperities [21]. Ou et al. [22] were able to significantly reduce water flow friction in micro-channels by using textured surface with a surface treatment to reduce surface energy. The slip length of these surfaces was about 20  $\mu$ m. In 2006, Choi and Kim 55 [23] created an artificial superhydrophobic surface composed of sharp silicon asperities covered with a layer of PTFE (PolyTetraFluoroEthylene). They tested the surfaces on 67 a cone-and-plate rheometer and found slip length up to 20  $\mu$ m with water. If 30 % of glycerin is added to water, the slip length could reach about 50  $\mu$ m. This huge slip length 59 value can be explained by the air trapped between the asperities, which reduces the 60 apparent friction of the fluid on the surface. Different surface texture patterns were 61 tested by Srinivasan et al. [24] on a plate-on-plate rheometer. A slip length of about 40 62  $\mu$ m was reached in the best case. A summary of the recent research on this topic and the 63 slip length values obtained can be found in the paper of Solomon et al. [25].

Super-hydrophobic surfaces appear to be a very attractive solution to reduce friction 65 in lubricated contacts thanks to slip length values that can be higher than the thickness of the lubricating film. However, in the literature, these surfaces were tested in micro-67 channel flows or in a rheometer where the operating conditions are barely less severe 68 than in lubricated contacts because there is no hydrodynamic pressure generation. In the 69 present work, we designed super-hydrophobic surfaces by texturing flat PTFE samples 70 with a femto second laser [26]. The surfaces were tested on a tribometer with different 71 speeds and load conditions. We then compared the results to untextured surfaces to 72 study the possible friction reduction provided by superhydrophobic textured surface in 73 hydrodynamic lubrication regime. 74

# 75 2. Materials and methods

76 2.1. Samples

A plate of 2 mm thickness of pure polytetrafluoroethylene PTFE (Approflon, France) 77 was used to cut out circular samples with a manual press equipped with a cookie cutter 78 of 10 mm in diameter. The samples were polished with an automatic polishing machine 79 until 0.055  $\mu$ m arithmetic roughness (Sa) was obtained. After the polishing step, the 80 samples were washed with ethanol in an ultrasonic bath for 15 min and then dried 81 for 1 h in an oven at a temperature of 95  $^{o}$ C. Contact angle measurements are then 82 realized with a semi-automatic goniometer (Krüss DSA25) by posing a drop of 5  $\mu$ l 83 on the sample surface. For each sample, contact angle measurements are done in five 84 different areas. The values of contact angle were between  $107^{\circ}$  and  $112^{\circ}$  (see figure 1), 85 which is in agreement with the literature [27].





## 87 2.2. Surface texturing

To increase the hydrophobicity and reach a superhydrophobic state, the surfaces of 88 the PTFE samples were textured. To allow a better mobility of the water on the surface, 89 the objective was to obtain a Cassie Baxter wetting state, when the water drop lies on 90 the top of the asperities, and avoid the Wenzel wetting state, when the water drop fully 91 wets the asperities. Restrictions need to be imposed on the geometry of the pattern to 92 ensure that the Cassie Baxter will be the most favorable energetic state [28]. Moreover, 93 the texture must withstand mechanical shear loading during the friction tests. Thus, it 94 was decided to create a network of cavities rather than the slender pillars or ridges that are used in the literature [25]. In this case, the contact angle  $\theta_{textured}$  for the Cassie Baxter 96 state satisfies the relation: 97

$$\cos\theta_{textured} = f\cos\theta + f - 1,\tag{1}$$

where f is the ratio of the area of the top surface (out of the cavities) to the total area and 98  $\theta$  the contact angle for the smooth surface. To maintain a minimum material thickness 99 between the cavities, the surface fraction f cannot be lower than about 0.2. If  $\theta \simeq 110^{\circ}$ , 1 00 then a contact angle of 150 ° is expected theoretically for a textured surface. The surface 1 01 texture was manufactured with a femto-second laser (Manutech, France). The minimum 1 0 2 texture lateral size that can be machined with the laser is 10  $\mu$ m. A second set of samples 1 0 3 with a lateral size of 20  $\mu$ m was also machined to obtain a better control of f and of 1 04 the shape of the cavities. Figure 2 shows the topography measured with a white light 1 0 5 interferometer (Taylor Hobson Talysurf CCI 6000) on a sample with a 10  $\mu$ m pattern size 106 and on a sample with a 20  $\mu$ m pattern size. The average depth of the pattern is about 5 107  $\mu$ m for the small texture and about 10  $\mu$ m for the large texture. 108



**Figure 2.** Surface topography of the samples measured by white light interferometry: (**a**) Sample with 10  $\mu$ m lateral texture size. (**b**) Sample with 20  $\mu$ m lateral texture size.

As proposed by Salant and Fortier [8], the samples were only partially textured to 109 give a heterogeneous slip/no slip surface that will generate hydrodynamic lift between 110 the sample and the counterface. Indeed, in hydrodynamic lubrication, the lubricant 111 is drawn into the contact by the sliding movement of the surfaces. To maintain this 112 lubrication regime, part of the surfaces must form a converging space so that the outlet 113 section of the entrained fluid is smaller than the inlet. This design keeps the fluid 114 film under pressure and creates a lift force that pushes the two surfaces apart. We 115 were inspired by this principle by texturing only half of our samples. In this way, the 116 lubricating film will pass through the textured part, in which the flow of the fluid will be 117 facilitated by the slip, unlike the exit (the second half of the sample was kept smooth). 118 This will create a lift force that will prevent contact of the two surfaces and will maintain 119 a hydrodynamic lubrication regime throughout the friction test. The configuration of 120 the half textured sample is presented in figure 1. Three samples of each texture were 121 manufactured and tested. 1 2 2

## 123 2.3. Test rig and experimental conditions

The test rig that we used for these experiments is presented in figure 3 a) and 1 24 schematised in figure 3 b). The tribometer is composed of an horizontal arm mounted 125 on a frictionless air bearing allowing free vertical motion of the sample support. The 126 polymer sample was glued to the arm of the tribometer. Before the glue dries, the PTFE 127 disk was put into the contact with the counter-face, that is a smooth glass disk, to ensure 128 perfect alignment of the sample and the glass. After drying, a controlled load was 129 applied by means of a calibrated mass. Some water was poured on the glass disk to 1 30 lubricate the contact. The textured part of the sample was placed upstream with respect 1 31 to the direction of rotation of the disk, which allows the lubricant to pass through the 1 32 textured part first. Once the contact is established, the glass disk is rotated by a motor 133 mounted under the tribometer. A displacement sensor was mounted in front of a thin 1 34 elastic beam that maintains the sample support. When the sliding starts, the elastic 1 35 beam bends due to the friction resistance of the sample/disk contact. This bending 1 36 displacement was measured by the displacement sensor and related to the frictional 137 force of the contact by the stiffness of the setup measured prior to the experiments. 1 38



Figure 3. Presentation of the test rig : (a) Picture of the rig. (b) Principle of operation.

Several sliding conditions were tested by changing the sliding speed and normal load. The rotational speed  $\omega$  of 100 rpm, 150 rpm and 200 rpm and the calibrated masses from 10g to 70g were used. The radial position of the sample on the disk was  $R_d$  = 0.035 m. The test conditions are summarized in table 1. The averaged loading pressured calculated from the applied mass and the sample area is also indicated.

Each sample was used only once for each normal load and sliding speed combination to avoid any wear of the surface texture. Sliding duration was set at 30 seconds. The

friction coefficient presented later on was averaged on the last 25 seconds.

Table 1. Test conditions.

Parameter	Value
Sample material	PTFE
Rotating disk material	Glass
Lubricant	Water at 20°C
Rotational speed $\omega$ (rpm)	100, 150, 200
Applied mass (g)	10, 30, 50, 70
Corresponding averaged pressure (kPa)	1.3, 3.8, 6.4, 9.0

# 147 2.4. Simulation tool

A simulation tool was used to analyze the results of the test. The configuration of the studied problem is presented in figure 4. The sample of radius R is placed at distance  $h_0$  from a flat surface moving along the x direction. A possibility of integrating a flatness defect of the sample was introduced into the model by introducing two radii of curvature  $R_1$  and  $R_2$ .



Figure 4. Configuration of the simulated problem.

It is assumed that the fluid flow between the sample and the moving surface is governed by the Reynolds equation. In addition, a mass conserving cavitation model is used [29], leading to:

$$F\frac{\partial}{\partial x}\left(\frac{h^3}{\mu}\frac{\partial D}{\partial x}\right) + F\frac{\partial}{\partial y}\left(\frac{h^3}{\mu}\frac{\partial D}{\partial y}\right) = 6V\left[\frac{\partial h}{\partial x} + (1-F)\frac{\partial hD}{\partial x}\right]$$
(2)

where *h* is the local film thickness and  $\mu$  the fluid viscosity. *F* is switch function and *D* a universal variable that is related to the pressure *p* of the fluid or its density  $\rho$ , depending if the fluid film is cavitated. In full film zones:

$$F = 1, \quad D = p, \quad \text{and} \quad \rho = \rho_0$$
(3)

and in zones of cavitation:

$$F = 0, \quad D = \frac{\rho}{\rho_0} - 1, \quad \text{and} \quad p = p_{cav}.$$
 (4)

In these equations,  $\rho_0$  is the liquid density and  $p_{cav}$  is the cavitation pressure. The Reynolds equation was discretized by the finite difference method and the system of equations was solved by a direct LU decomposition method for sparse matrices. Once the pressure in the fluid domain was calculated, the normal force *N* and the friction force *T* on the sample were calculated.

# 158 3. Results

## 159 3.1. Sample characterization

The contact angle results for the textured surfaces are given in table 2. A representa-160 tive image of a water drop on a textured surface can be seen in figure 1. As expected, the 161 texture significantly increased the contact angle that was initially close to 110° for the 162 flat smooth PTFE surface. The contact angle of textured surfaces reached values close 163 to, or higher than  $150^{\circ}$ . This value is considered to be the threshold, above which the 1 64 behavior is superhydrophobic. Furthermore, the texture with a 20  $\mu$ m lateral size gave 165 better results because of better precision of the laser machining and better control of the 166 surface fraction *f*. 167

Sample	Spacing	Static angle
T10-1	10 µm	133±1°
T10-2	$10 \ \mu m$	$143\pm2~^o$
T10-3	10 µm	$145\pm2~^o$
T20-1	20 µm	$147\pm2~^o$
T20-2	20 µm	$142\pm2~^o$
T20-3	20 µm	$153\pm3~^o$

Table 2. Contact angle of water drops on texture surfaces.

In addition to the contact angle characterization, the global shape of the samples 168 was measured with a 3D optical microscope (Alicona InfiniteFocus). It was then possible 169 to calculate the principal radii of curvature  $R_1$  and  $R_2$  of the surfaces of all of the samples 170 that we used. The results are presented in figure 5. These results show that the samples 1 71 were not perfectly flat, with a curvature radius of about 0.5 m. This curvature induced a 172 film thickness variation in the contact between the sample and the glass disk of several 173 tens of microns. It is noteworthy that this value is significantly larger than the texture 1 74 depth. 175



Figure 5. Principal curvature radii of the PTFE samples

#### 176 3.2. Friction tests

Figure 6 a) presents typical tangential force signals as a function of the sliding 177 distance for one sample of each type (smooth, textured with a lateral spacing of 10 178  $\mu$ m and textured with a lateral spacing of 20  $\mu$ m). Even if the average value of the 179 tangential force is stabilized, all the three signals exhibits some oscillations at three 1 80 different frequency levels (see figure 6 b). The high frequency corresponds to noise on 1 81 the signal. The oscillation with a period of about 0.07 s is the flexural natural frequency 182 of the tribometer. The damping of the fluid film is not sufficient to vanish the vibration 183 during the test length (30s). Finally an oscillation with a longer period probably induced 1 84 by the revolution period of the disk (about 0.4 s) is also viewable. The signals are 185 averaged over 25 s to eliminate the effect of these time oscillations. 186



**Figure 6.** Measured tangential force as a function of the distance or time for three type of samples. The loading mass is 30 g and the rotational speed is 150 rpm. a) Tangential force versus sliding distance for full test recording b) Tangential force versus time during the last half second

The results of the tests presented in figure 7 are averaged over three samples of each category (smooth, textured with a lateral spacing of 10  $\mu$ m and textured with a lateral spacing of 20  $\mu$ m). An error bar represents the variation among the three samples. The friction coefficient is the ratio of the friction force *T* over the normal load *N*. It is presented as a function of the duty parameter *G*, which is defined in the following way:

$$G = \frac{2\mu VR}{N} \tag{5}$$

where  $V = R_d \omega$  is the linear sliding speed of the sample/disk contact.

To highlight the evolution of the friction, a power law was fitted to each set of results. 188 For each sample family, the opacity of the markers is proportional to the applied load 189 making it possible to identify the effect of speed and load independently. Increasing the 1 90 load at a given speed makes the friction lower while increasing the speed leads at a given 191 load to a higher friction. For each family of samples, the friction increased with G, which 192 means that the lubrication regime is mainly hydrodynamic. The variations in friction 193 coefficient obtained for each category can be related to the difference in shape within 1 94 the samples (see figure 5). Unlike the expectations, the lowest friction was obtained 195 with the smooth surfaces, while the 10  $\mu$ m spacing textured surfaces gave the highest 196 friction value. Although the surface texturing was expected to allow slip of the fluid 197 on the surface by increasing the contact angle, the results revealed that this was not the 198 case. One possible explanation of this surprising finding is that the flatness defects of the 199 sample had an influence on the results. This hypothesis will be discussed and verified in 200 the following section. 201



**Figure 7.** Measured friction coefficient as a function of the duty parameter *G* for three type of samples. The values are averaged from three samples of each set and the error bar is the standard deviation. The opacity of the symbols is proportional to the load level. A power law fit curve is presented for each category.

## 202 4. Discussion

## 203 4.1. Simulation of the effect of surface curvature

According to the lubrication theory, no hydrodynamic pressure can be generated 204 between parallel flat surfaces. However, our results showed that a hydrodynamic lubri-2.05 cation regime can be reached with smooth samples. The hydrodynamic force is in fact 206 generated by the surface residual curvatures due to the polishing process. In addition, 207 this force depends on the surface curvature radii. This makes the comparison of the 208 performance of different samples difficult because their surfaces are not identical. To 209 highlight the effect of surface curvature, simulations were carried out on the configu-210 ration presented in figure 4. The simulations were performed for different values of 211 the curvature radii  $R_x$  and  $R_y$  of the sample and for different values of the central film 212 thickness  $h_0$ . The parameters that were used in the simulations are given in table 3. 213

Parameter	Value
Viscosity $\mu$ (Pa.s)	0.001
Averaged sliding speed $V$ (m.s <sup>-1</sup> )	1
Sample radius $R$ (m)	0.005
Curvature radius $R_x$ (m)	0.1 - 1
Curvature radius $R_y$ (m)	0.1 - 1
Domain size (m)	0.012
Number of nodes	256  imes 256
Cavitation pressure $p_{cav}$ (Pa)	0

Table 3. Parameters used for the simulation of figure 8.

The calculated friction coefficient  $f = \frac{T}{N}$  was presented as a function of the duty parameter *G* scaled by a shape factor:

$$G^* = G \times \left(\frac{\sqrt{R_x R_y}}{R}\right)^a \tag{6}$$

All of the simulation results with different surface curvatures were laid on the same line with the power law coefficient a = 0.355, as can be seen in figure 8. When presented as a function of this modified duty parameter  $G^*$ , the friction coefficient is independent on

the surface curvature. It is thus possible to fit the simulation results with a power law function:  $f = (1 + c_{1}) + c_{2} + c_{3} + c_{4} + c_{4}$ 

$$f = 6.16 \times (G^*)^{0.62} \tag{7}$$



**Figure 8.** Simulated friction coefficient as a function of the modified duty parameter  $G^*$  for different values of the surface curvature radii. A power law curve has been fitted on the results.

# 4.2. Discussion of the experimental friction results

To remove the effect of the residual curvature of the samples, the friction coefficient 215 is presented as a function of  $G^*$  in figure 9. The power law (7) obtained from simulations 216 has also been added to the figure. It is interesting to see that the friction coefficient 217 obtained with the smooth samples S2 and S3 is in good correlation with simulation law. 218 Meanwhile, sample S1 gives a slightly lower friction. When the surfaces are textured, a 219 significantly higher friction is obtained, except for a few conditions. This observation 220 stands for the two configurations of texture. Although an increase of the hydrophobicity 221 of the surfaces due to texturing was expected to provide a better sliding of the water and 222 to reduce friction, the opposite is obtained here. This means that the slip of water does 223 not take place. 224



**Figure 9.** Friction coefficient as a function of the modified duty parameter  $G^*$ , Eq. (6) - Experimental results and comparison to the numerically fitted curve

An analysis of the results of the literature ([13,22,23,25]) where a friction decrease was obtained thanks to surface texture enhancing hydrophobicity shows that in all cases the texture was composed of pillars or ridges parallel to the flow direction. Even if an amount of air is maintained between the texture, the fluid flow was facilitated between the ridges and pillars. In our case, the texture is composed of cylindrical cavities. Even if these cavities are efficient in increasing hydrophobicity and can maintain the air trapped in the cavities, there is no easy path for the water flow.

In addition, during the simulations performed to obtain the results of figure 8, the maximum fluid pressure calculated in the fluid film varied between about 6.5 kPa and 270 kPa, depending on the value of  $G^*$ . This pressure would impact the interface between water and the air trapped in the cavities, as shown in figure 10. The curvature radius of the spherical interface is given by the Laplace equation:

$$R_c = \gamma \times \left(\frac{2}{p_\ell - p_g}\right) \tag{8}$$

where  $\gamma$  (= 0.071 N.m<sup>-1</sup>) is the water surface tension,  $p_{\ell}$  is the liquid pressure and  $p_g$  is the gas pressure. The maximum pressure differential that can withstand the interface depends on the diameter of the hole and the value of the contact angle  $\theta$  (see figure 10). It is thus possible to calculate a critical diameter  $d_c$  of a hole from the fluid pressure resulted from the simulations:

$$d_c = -\gamma \times \left(\frac{4\cos\theta}{p_\ell - p_g}\right). \tag{9}$$

Using the pressure calculated in the simulations, it is found that  $d_c$  is in the range 0.36  $\mu$ m for the highest pressure value to 15  $\mu$ m for the lowest pressure. The size of the texture used in the experiments is thus not small enough to ensure the stability of the water-air interface for all of the experimental conditions. It is thus possible for the water to fill the cavities, either partially or completely.



Figure 10. Effect of the liquid pressure on the air water interface shape.

Several papers [30,31] dedicated to the simulation of fluid flow over textured 237 surfaces with a shape ratio (texture depth to texture width) similar to our case have 238 demonstrated that flow vortices can be observed in the cavities, even at a very low 239 Reynolds number. More particularly, Mateescu et al. [30] showed that the viscous 240 dissipation in these vortices leads to an increase in drag or friction even for Reynolds 241 number values tending to zero. The most important parameter for drag increase was 242 shown to be the ratio of cavities depth to film thickness. Given that it is possible for 243 water to partially or completely fill the cavities due to the hydrodynamic pressure in 244 the liquid, the air-water interface can be destabilized thus creating water cavities (figure 245 11). The results of Mateescu et al. [30] suggest that some vortices can appear in our 246 samples and increase the friction. In this case, the texture would work as a labyrinth 247 seal, contrary to what was expected. 248



Figure 11. Vortices in the cavities increasing the sliding friction.

## 249 5. Conclusion

In this paper, the possibility to reduce friction in a hydrodynamic lubrication regime 250 with superhydrophobic textured surfaces was experimentally investigated. PTFE disks 251 were mechanically polished and textured with a femto second laser to create a hexagonal 252 network of cylindrical cavities on a half of the sample's surface. The results showed that 253 with cavities spacing of 10 and 20  $\mu$ m, the hydrophobic character of the samples was 254 increased with a contact angle up to 153°. Friction tests were conducted on these samples 255 and compared to the results of smooth samples of the same material. In contrast to what 256 was expected, these tests showed that the texture increased the friction coefficient. The 257 numerical simulations with Reynolds flow showed that the observed effect could not 258 be explained by the imperfect curvature of the sample surfaces. However, comparing 259 our results with the literature provided a plausible explanation in that the fluid partially 260 or completely filled the cavities due to high pressure in the contact. In addition, the 261 literature supports the hypothesis that vortices could appear in the cavities, leading to 262 an increase in friction due to the viscous dissipation. Indeed, the texture in our samples 263 was applied by creating holes instead of the pillars used by other researchers. Our 264 texture pattern was designed for better mechanical resistance of the surfaces to shear 265 due to friction. However, it seems that this type of texture modifies the flow compared 266 to the pillar texture, thus increasing the friction. This phenomenon can be avoided by 267 using smaller cavities (sub-micron scale), which would imply the need to change the 268 machining process. 269

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## 280 Abbreviations

- <sup>281</sup> The following abbreviations are used in this manuscript:
- 282
- 283 PTFE PolyTetreFluoroEthylene

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