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Method Section

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How drops start sliding over solid surfaces

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Methods

Preparation of solid surfaces and liquid drops

The following chemicals were used to fabricate the solid substrates: trichloromethylsilane (TCMS, 99%, Sigma-Aldrich), 1*H*,1*H*,2*H*,2*H*-perfluorodecyltrichlorosilane (PFDTS, 96%, Alfa Aesar), 1*H*,1*H*,2*H*,2*H*-perfluorooctyltrichlorosilane (PFOTS, 97%, Sigma-Aldrich), Sylgard 184 (PDMS precursor, Dow Corning), Sylgard 184 curing agent (Dow Corning), trimethylsiloxy-terminated poly-(dimethlysiloxane) (PDMS) (200 cSt., ABCR), sulfuric acid (VWR, 95%), ammonia solution (VWR, 28%), hydrogen peroxide solution (Sigma-Aldrich, 34.5–36.5%), *n*-hexane (99.99%, Fisher Chemical), toluene (99.99%, Fisher Chemical), isopropanol (99.5%, Fisher Chemical), acetone (99.98%, Fisher Chemical), ethanol (absolute, 99.96%, VWR Chemicals), tetrahydrofuran (99.9%, Sigma-Aldrich), SU-8 3025 photoresist (Microchem), mr-Dev 600 developer (micro resist technology), 18NR-T (Dyesol) and sodium hydroxide (98%, Sigma-Aldrich). Reagents were used as received. Polished silicon wafers were obtained from Si-Mat. Thin glass slides of 24×60 mm^2 and 170±5 μm thickness were obtained from Carl-Roth. All syntheses were carried out at room temperature. The liquids drops used for force measurements included those of hexadecane, 1-butyl-2,3-dimethylimidazolium bis(trifluoromethanesulfonyl)- imide and water. Hexadecane (99%, Sigma-Aldrich) and 1-butyl-2,3-dimethylimidazolium bis(trifluoromethanesulfonyl)imide (99%, iolitec) were used as

1

29 received. Water with a typical resistivity of 18.2 MΩ·cm was obtained from a Sartorius Arium

30 661 VF Water Purification System.

31

Silicone Nanofilaments

33 Silicone nanofilaments were synthesized by modifying previously reported methods [1,2]. The

34 fabrication contained the following steps: First, 120 µL of TCMS was added into a reaction

35 chamber containing 50 mL of water-saturated *n*-hexane. The resulting solution was stirred for

36 60°s. Then, cover glass slides were immersed in the solution, before sealing the reaction

37 chamber. After 3 days the TCMS-coated glass slides were removed from the reaction chamber,

38 rinsed with *n*-hexane and dried under a nitrogen stream. The TCMS-coated glass slides were

39 activated in an oxygen plasma chamber (25 W, 2 min, 7 cm^3/min oxygen flow rate, Diener

40 Electronic Femto) before modification with the semi-fluorinated silane PFDTS. Therefore, the

41 activated substrates were immersed in a solution containing 25 µL of PFDTS dispersed in 50

42 mL of *n*-hexane for 20 min. Subsequently, the substrates were rinsed with *n*-hexane and dried

43 under a nitrogen stream. Representative scanning electron microscopic images of the

44 resulting silicone nanofilaments are shown in Fig. S1 (a & b).

45

Silicon Wafers

47 Pristine silicon wafers were cleaned by ultra-sonication in toluene, acetone and ethanol for 15

48 min, respectively. Subsequently, the pre-cleaned wafers were plasma-cleaned and activated

49 in an oxygen plasma chamber (300 W, 10 min, 6 cm^3/min oxygen flow rate). In order to

50 fluorinate the surfaces, 25 µL of PFDTS was mixed with 50 mL of *n*-hexane. The activated

51 silicon wafers were immersed in the solution for 5 to 15 min. Afterwards, the silicon wafers

52 were rinsed with *n*-hexane, followed by ultrasonication in isopropanol for 5 min. The cleaned

53 silicon wafers were dried under a nitrogen stream. It should be noted that the immersion time

54 influences the lateral adhesion force of the fluorinated silicon wafers. The scanning electron

55 microscopy analysis revealed featureless, smooth surfaces as shown in Fig. S1 (c & d).

56

57

SU-8 Pillars

59 SU-8 micropillar arrays were prepared on thin glass slides by photolithography [3]. The square

60 pillars were designed 25 μm high, with 50×50 μm^2 top areas. The pillar-pillar distance between

61 centers of two adjacent pillars in a row was 100 μm. The fabrication process consisted of the

62 following steps. First, glass slides (Carl-Roth) were cleaned by ultra-sonication in

63 tetrahydrofuran, acetone and ethanol for 15 min, respectively. Then, SU-8 photoresist was

64 spin-coated (500 rpm for 5 s followed by 3000 rpm for 30 s, SÜSS MicroTec) on the glass slides.

65 The coated glass slides were heated at 65 °C for 3 min, 95 °C for 10 min and then 65 °C for 30

66 min, respectively. Subsequently, the samples were slowly cooled down within two hours and

67 exposed to UV light (mercury lamp, 350 W) under a photolithography mask for 15 s (masker

68 aligner SÜSS MicroTec MJB3 UV400). To cross-link the photoresist, the samples were heated

69 up again at 65 °C for 3 min, 95° C for 10 min and 65 °C for 30 min, and then cooled down slowly.

70 Next, the samples were immersed in the SU-8 developer solution for 5 min, washed with

71 isopropanol and then dried in air, resulting in surfaces coated with SU-8 pillars. The dried

72 samples were immersed in 0.1 M NaOH solution overnight to hydrolyze the surfaces of the

73 SU-8 pillars. After rinsing the hydrolyzed surfaces with water and ethanol, the samples were

74 immersed in a solution containing 25 µL of PFDTS dispersed in 50 mL of n-hexane for 20 min.

75 Finally, the fluorinated SU-8 micropillar surfaces were rinsed with n-hexane and dried under a

76 nitrogen stream. Representative scanning electron microscopic images of an obtained SU-8

77 micropillar array are shown in Fig. S1 (e & f).

78

79 **PDMS**

80 Cross-linked PDMS surfaces: Sylgard 184 PDMS precursor was mixed with Sylgard 184 curing

81 agent in a ratio of 10:1 by weight and then degassed three times in a vacuum chamber for 5

82 min, respectively. Afterwards, the viscous mixture was deposited on glass slides and cured at

83 60 °C overnight. The scanning electron microscopy analysis revealed featureless, smooth

84 surfaces as shown in Fig. S1 (g & h).

85 Liquid-like PDMS surfaces: So termed "liquid-like" PDMS samples were prepared following the

86 method reported by Krumpfer and McCarthy.[4] The pre-cleaned cover slides were placed in a

87 2:1 mixture of sulfuric acid and hydrogen peroxide solution, placed in a water bath, and held

88 for 2 h at 70 °C. Then the samples were rinsed with pure water and ethanol. After being dried,

89 the cover slides were put into 500 mL ultraclean borosilicate glass vials (Duran Pure vials GL45

3

90 with a dust cover, Duran Group, Schott) and sufficiently wetted with PDMS. The vials were

91 resealed and placed in an oven for 24 h at 100 °C. Then the samples were properly rinsed with

92 hexane, acetone, and Milli-Q water and dried.

93 The flexibility of the O-Si-O bonds confers high mobility to PDMS chains. As only one end of

94 the PDMS is covalently grafted on the substrate, the remaining part of the PDMS keeps its

95 high mobility with rotational and/or bending motion. Thus, these grafted layers are

96 rotationally dynamic and behave "liquid-like". Water drops slide off these "liquid-like"

97 surfaces when tilting the surface by 4° (drop volume: 20 ml).[4]

98

99 **TiO$_2$ Nanoparticles**

100 TiO$_2$ surfaces consisting of approximately 20 nm TiO$_2$ nanoparticles (NPs) were fabricated from

101 TiO$_2$ nanoparticle paste (18NR-T, Dyesol) [5]. First, TiO$_2$ nanoparticle paste was coated on a

102 silicon wafer by doctor blade coating with 50 μm spacers. Then, the coated wafer was sintered

103 at 500 °C for 30 min, realizing nano-roughened TiO$_2$ structures. After cooling down, the TiO$_2$

104 structures were modified with PFOTS by chemical vapor deposition (CVD) in a desiccator for 2

105 h in vacuum. Scanning electron microscopic images of a resulting TiO$_2$ surface are shown in

106 Fig. S1 (i & j).

107

108 **Force Measurements**

109 **Sensitivity of the Laser Deflection System**

110 For the measurement of the lateral adhesion force by means of our laser deflection system

111 illustrated in Fig. 1b, a hollow rectangle capillary was used to move a liquid drop against a

112 substrate underneath [6,7]. A laser beam was incident on the capillary, which reflected the beam

113 to the center of a position sensitive detector (PSD). The capillary was manufactured in

114 borosilicate glass (VitroCom's Vitrotubes, CM Scientific). It was 35 mm long. The internal

115 dimensions of the capillary were 0.04 mm × 0.40 mm, with a wall thickness of 0.028 mm. In

116 order to increase the reflectivity of the capillary, its wide side was sputter-coated with a 5-

117 nm-thick layer of chromium and a 50-nm-thick layer of gold. The chromium was used as an

118 adhesion layer for the gold. When the laser hit the PSD, four photocurrents were generated

119 by the PSD at two opposite sides along the horizontal axis and two opposite sides along the

4

120 vertical axis. These four photocurrents were collected and converted into four voltages (U_{x1},

121 U_{x2}, U_{y1}, and U_{y2}) using an amplifier with a current-to-voltage converter. These four voltages

122 were read out in two values by the amplifier corresponding to the horizontal (x) and vertical

123 (y) coordinates of the laser spot on the PSD:

124
$$U_x = 5\text{V} \cdot \frac{U_{x1} - U_{x2}}{U_{x1} + U_{x2}}$$
Eq. 4

125
$$U_y = 5\text{V} \cdot \frac{U_{y1} - U_{y2}}{U_{y1} + U_{y2}}$$
Eq. 5

126 The divisions were performed by an analog divider (Burr-Brown DIV100). $U_y = U_{PSD}$ and

127 corresponded to the deflection of the capillary in the measurement of the lateral adhesion

128 force. These voltage values were digitalized in an AD converter (National Instruments (NI)).

129 Furthermore, the data acquisition software generated triggers for the cameras to ensure

130 synchronization between the cameras and the PSD signals.

131 The relationship between the deflection of the capillary, D, and the generated U_{PSD} represents

132 the sensitivity S of the laser deflection system:

133
$$S = \frac{U_{PSD}}{D}$$
Eq. 6

134 For calibration a defined deflection of the capillary was applied close to its free end by a

135 micromanipulator. A linear correlation was found between U_{PSD} and D for deflections up to

136 800 µm. The corresponding sensitivities were in the range of $4 - 5 \times 10^{-3}$ V/µm. We performed

137 a calibration for each new capillary and after each readjustment of the setup. Fig. S2 shows a

138 representative plot, with S = -4.16×10^{-3} V/µm and Pearson r = 0.9999. The sign of the voltage

139 corresponds to the direction of the laser displacement on the PSD along the y-axis.

140

141 **Spring Constant of the Capillary**

142 The spring constant κ of the glass capillary was calibrated by measuring the force acting on

143 the capillary with respect to the lateral deflection. First, the capillary was fixed at one end into

144 a copper holder as a fixture (Fig. S3). Then the entire fixture was mounted to a

145 micromanipulator (MMO-203, Narishige), which allowed us to move the fixture precisely (with

146 1 µm precision). A pin with supporting pedestals at the bottom and a pointy tip facing up was

147 placed on a microgram balance (Fig. S3). Using the micromanipulator, the free end of the

148 capillary was located right above the tip of the pin. Afterwards, the capillary was lowered

149 down slowly by means of the micromanipulator to engage with the tip until a slight deflection

150 of the capillary was generated. The place where the free end of the capillary was contacted

151 by the pin was within 1 ± 0.2 mm from the edge of the free end.

152 The microgram balance read out the weight that reflected the force, F, between the pin and

153 the deflected capillary, as F = mg, where g = 9.81m/s^2. Fig. S4 is a representative plot of the

154 calibration of the spring constant of the glass capillary. The relation between the force, F, and

155 the deflection, D, followed Hooke's law[8]

156
$$F = \kappa \cdot D$$
Eq. 7

157 with κ = 0.202±0.002 N/m.

158

159 **Force Conversion**

160 In order to obtain the lateral adhesion force, the voltage values directly collected from the

161 PSD need to be converted into forces. Combining Eq. 6 and Eq. 7 yielded the relationship

162 between the lateral adhesion forces, F_{LA}, and the generated voltage:

163
$$F_{LA} \equiv F = \kappa \cdot D = \kappa \cdot \frac{U_{PSD}}{S}$$
Eq. 8

164

165 **Substrate movement system**

166 Drops were deposited on solid substrates using a pipette (Thermo Scientific, Finnpipette F2).

167 In order to probe a velocity regime that spans 5 orders of magnitudes we implemented 3

168 different motorized stages to drive the relative motion between the drop and the substrate.

169 The low velocity regime of 2 µm/s to 200 µm/s was realized by a slow linear stage consisting

170 of a custom-made step motor whose single steps are approximately 34.2 nm. The medium

171 velocity regime of 200 µm/s to 1.6 mm/s was achieved by using a circular disk driven by a

172 linear motor (Faulhaber 2232 A 012 SR with transmission gear 22 EK 23014:1). The circular

173 disk rotated at a constant angular velocity in the regime of 0.02 rpm to 0.15 rpm. The liquid

174 drops were placed at a radius of 100 mm to the rotation center. The high velocity regime of

175 6.8 mm/s to 33.9 mm/s was realized again by a linear stage consisting of a custom-made step

176 motor which single steps are approximately 5 µm.

177

Drop imaging

A high speed camera (FASTCAM Mini UX100 type 800K-M-8G, Photron) equipped with a manual zoom lens was placed orthogonal to the drop motion. Its lens was focused on the drop with the narrow side of the capillary facing this orthogonal camera to monitor the change in front and rear contact angles as well as contact length. A second camera (Marlin F131B, Allied Vision) equipped with a manual zoom lens was placed parallel to the drop motion. Its lens was focused on the drop with the wide side of the capillary facing the parallel camera to monitor the change in contact width.

Before the sample was moved, the glass capillary was placed in the center of the drop. Both cameras were synchronized in time with the data acquisition system. Throughout an experiment the synchronization between force and optical measurements via the cameras was <60 ms. The drop was illuminated with two cold-light sources (KL 2500 LCD, Zeiss; L2, Leica), each of them located opposite to a camera, respectively. The contact angles, as well as the contact widths and lengths, were measured by means of ImageJ. The contact widths and lengths were measured using the known dimensions of the capillary as scale. Figs. S5-S10 show representative lateral adhesion forces with measured contact widths, contact lengths, and contact angles corresponding to the cases presented in Fig. 3a. Movies 1-7 show the motions of drops on different surfaces during the lateral adhesion force measurements corresponding to the cases presented in Fig. 2 and Fig. 3.

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