Supporting information

Superhydrophilic-Superoleophobic Coatings

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Experimental

Materials. Poly(diallydimethylammonium chloride) (PDDA, $Mw = 200\ 000-350\ 000$) and perfluorooctanoic acid (CF₃(CF₂)₆COOH) were all purchased from Sigma-Aldrich. Sodium perfluororooctanoate (0.10 M) was prepared by the reaction of perfluorooctanoic acid with NaOH in water. SiO₂ nanoparticles (average diameter ~ 20 nm) were purchased from Zhoushan Nanomaterials Co., China.

PDDA-PFO/SiO₂ synthesis: The SiO₂ nanoparticles were ultrasonically dispersed in PDDA ($Mw = 200\ 000-350\ 000$) aqueous solution (100 mL, 1.0 mg/mL), and a sodium perfluorooctanoate aqueous solution (20 mL, 0.1 M) was added dropwise under stirring at ambient temperature. A slurry was generated immediately due to PFO anions coordinated to quaternary ammonium groups of PDDA. The product was rinsed with deionized water and dried.

Spray-coating: PDDA-PFO/SiO₂ (0.2 g) was dispersed in ethanol (20 mL) under sonication, and the resulting suspension was sprayed onto substrates (copper, glass slide, and stainless steel mesh) with 0.2 MPa nitrogen gas using a spray gun

(Shanghai, Lotus brand, no. 1). The nanocomposite coatings were dried at room temperature for 2 h allowing the ethanol to evaporate completely.

Characterization: Contact angle measurements were performed using a Krüss DSA 100 (Krüss Company, Ltd., Germany) apparatus at ambient temperature. The volumes of various liquids in all measurements were approximately 5 μ L. Scanning electron microscopy measurements were carried out using a JSM-6701F field-emission scanning electron microscopy (FESEM, JEOL, Japan). X-ray photoelectron spectroscopy (XPS) analysis of the sample was performed on a PHI-5702 electron spectrometer using an Al K α line excitation source.



Figure S1. FESEM images of the nanocomposite coatings with various SiO_2 contents: a) 25, b) 40, c) 50, d) 67 wt%. An increase in the nanoparticle concentration causes the coating surface to generate less smooth areas and more particle aggregates.



Figure S2. The contact angles and sliding angles with rapeseed oil, diesel, and dodecane on the as-prepared PDDA-PFO/SiO₂ coating. These liquid droplets are all in spherical shapes with the contact angles greater than 150° and low sliding angles.



Figure S3. The contact angles with water and hexadecane on the nanocomposite coatings as a function of SiO_2 mass fraction. The coatings become superhydrophilic-superoleophobic for all SiO_2 concentrations above 50 wt%.



Figure S4. a) Survey spectrum and b) high-resolution C 1s spectrum of the PDDA-PFO/SiO₂ coating after plasma treatment. The content of oxygen element increases from 13.1 % to 30.8 %.



Figure S5. FESEM images of the stainless steel mesh before and after spray-coated with the PDDA-PFO/SiO₂ nanocomposite. After spray-coated with the PDDA-PFO/SiO₂, the smooth structures of stainless steel wires become roughness with large numbers of protrusions and pores.

Movie S1. Movie for the superhydrophilic-superoleophobic properties of the PDDA-PFO/SiO₂ coating after plasma treatment.



Movie S2. Movie for the oil/water separation experiment.

