

## Supporting Information

### Covalently cross-linked TiO<sub>2</sub>-siloxane networks for robust superhydrophobic coating

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### Experimental section

#### Materials

Octadecyltrichlorosilane was obtained from Xilong Scientific Co., Ltd. 2-(3,4-Epoxy cyclohexyl)ethyltrimethoxysilane, bis(3-triethoxysilylpropyl)amine, titanium dioxide nanoparticles (20 nm), and phosphoric acid were purchased from Shanghai Macklin Biochemical Technology Co., Ltd. All chemicals were used directly without further treatment.

#### Superhydrophobic TiO<sub>2</sub>/silane nanocomposite coating

0.3 g of octadecyltrichlorosilane was dissolved in 10 mL of anhydrous ethanol and sonicated for 15 min. Subsequently, 1 g of TiO<sub>2</sub> nanoparticles was added, and the mixture was stirred at 45 °C for 2 h. The product was dried at 120 °C to obtain the alkylated TiO<sub>2</sub>.

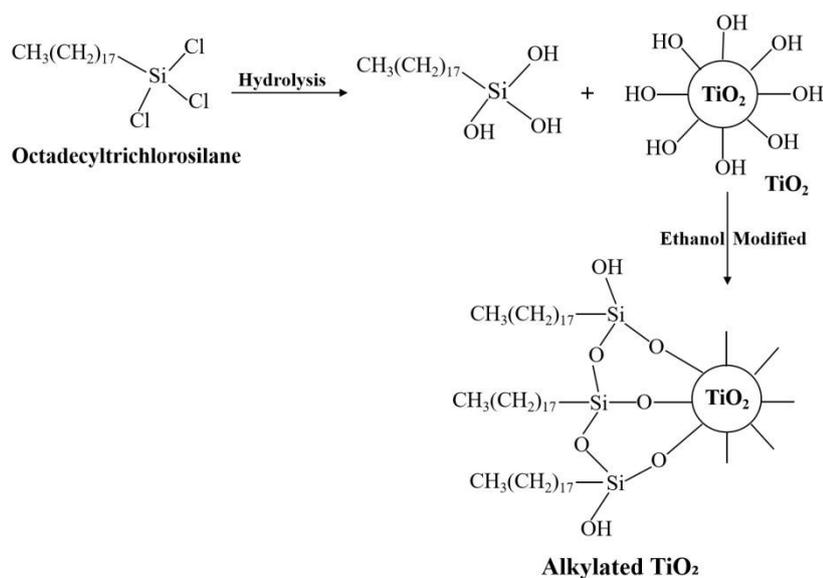
A mixture of 1.23 g of 2-(3,4-epoxy cyclohexyl)ethyltrimethoxysilane and 2.12 g of bis(3-triethoxysilylpropyl)amine was stirred in 17.8 g of anhydrous ethanol at 500 rpm. Then, 0.89 g of deionized water and 30 μL of phosphoric acid (to adjust pH to 2–2.5) were added. Stirring was continued at room temperature for 1.5 h to yield the epoxy-amino bifunctional siloxane oligomer.

1.1 g of the alkylated TiO<sub>2</sub> and 0.3 g of the siloxane oligomer were dispersed in 10 mL of ethanol under sonication. After stirring at 45 °C for 2 h, the resulting

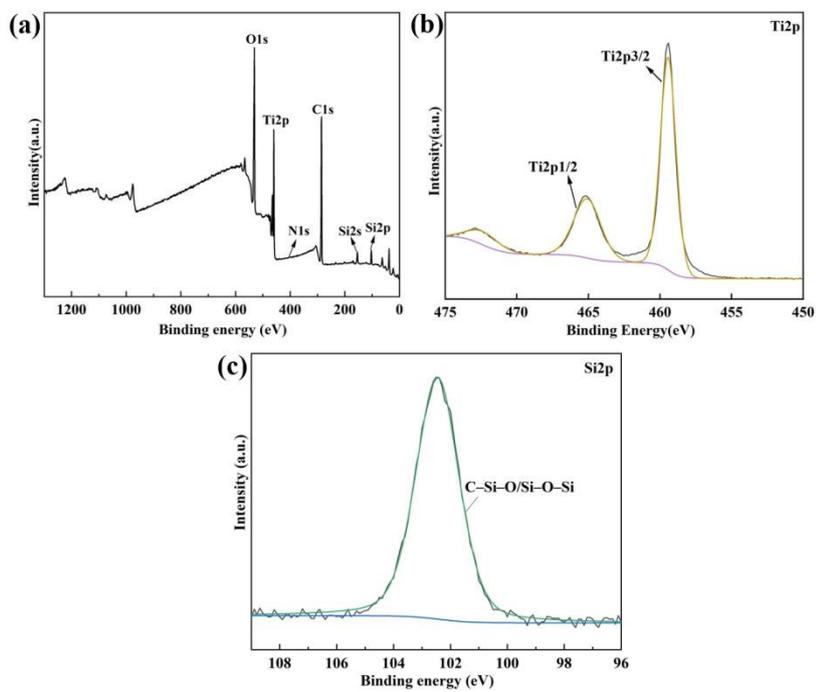
composite sol was applied onto a tinplate and cured at 120 °C to obtain the superhydrophobic TiO<sub>2</sub>/silane nanocomposite coating.

### Characterization

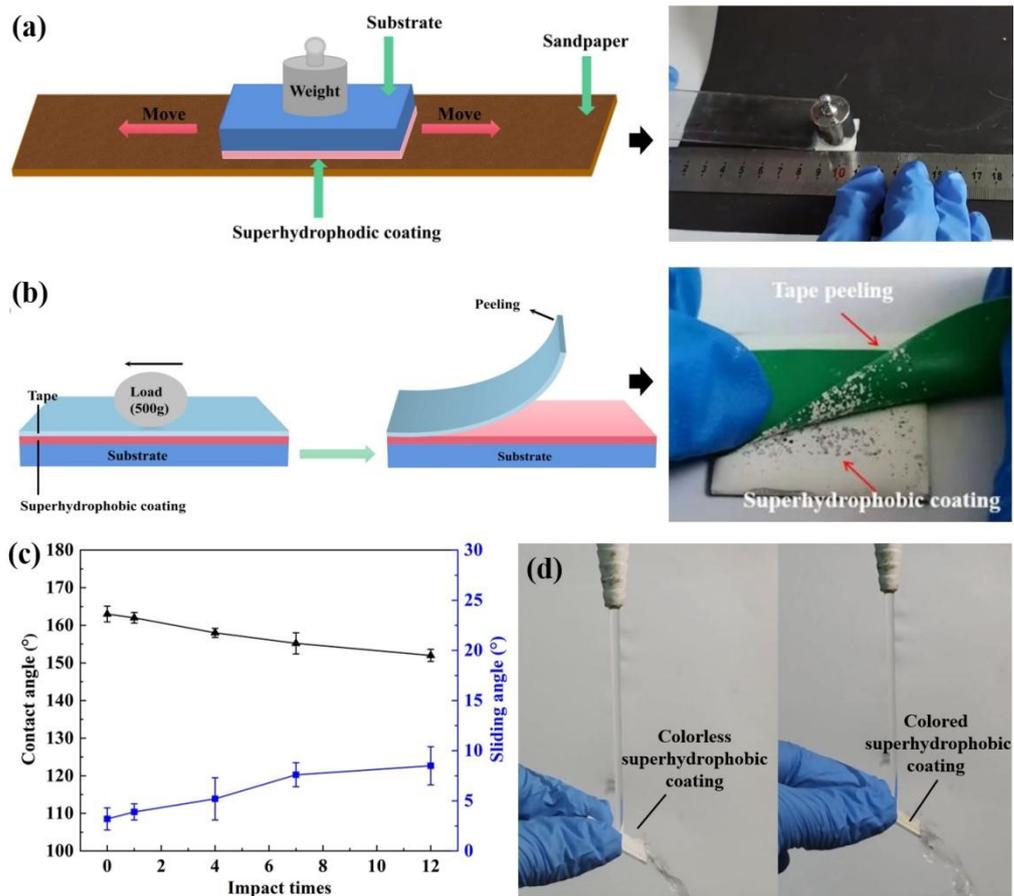
The surface morphology of the samples was examined using a scanning electron microscope (Apreo 2C, Thermo Fisher Scientific, USA) after gold sputtering, operating at 10–30 kV. The chemical composition of the samples were characterized by X-ray photoelectron spectroscopy (K-Alpha, Thermo Fisher Scientific, USA) using a monochromatic Al K $\alpha$  source ( $h\nu = 1486.6$  eV). Fourier-transform infrared spectroscopy (Nicolet iS10, Thermo Fisher Scientific, USA) was performed in ATR mode over the range 4000–400 cm<sup>-1</sup>. Surface topography and roughness were measured with an atomic force microscope (Dimension Edg, Bruker, Germany) in tapping mode using a NanoScope IIIA system, scanning an area of 1  $\mu\text{m} \times 1 \mu\text{m}$ . Water contact angles were determined using a goniometer (JCY-2, Shanghai Fangrui Instrument Co., Ltd., China) with 5  $\mu\text{L}$  deionized water droplets at 25 $\pm$ 2 °C, reported values are averages of at least five measurements.



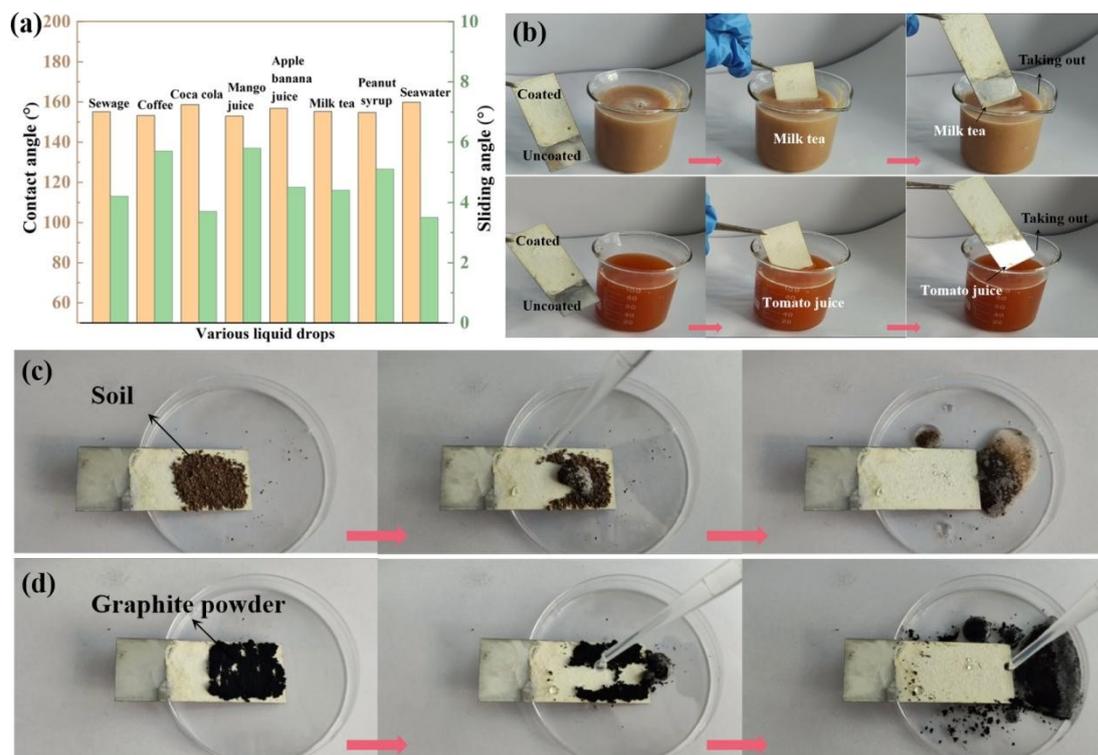
**Fig. S1** Hydrolysis of octadecyltrichlorosilane and ethanol-mediated modification of hydroxylated TiO<sub>2</sub> to alkylated TiO<sub>2</sub>.



**Fig. S2** (a) XPS survey spectra, (b) Ti2p, and (c) Si2p high-resolution XPS spectra of the superhydrophobic coating.



**Fig. S3** (a) Schematic of the sandpaper abrasion test (top) and its experimental setup (bottom): The superhydrophobic coating (on the substrate) undergoes reciprocating motion against sandpaper under a specified load. (b) Schematic of the tape-peeling test (left) and its experimental demonstration (right): A high-bond-strength tape is uniformly pressed onto the coating surface using a 500 g steel roller (two passes), followed by a 90 s dwell period before peeling. (c) Water contact angle and sliding angle of the superhydrophobic coating as a function of impact times from a simulated rainwater jet traveling at ~10 m/s. (d) Experimental demonstration of a simulated rainwater jet (traveling at ~10 m/s) impacting a colorless superhydrophobic coating (left image) and a colored superhydrophobic coating (right image).



**Fig. S4** (a) The contact angle and sliding angle of different droplets on the coating surface. (b) Uncoated chromium free passivated tinplate (lower image) and superhydrophobic coated chromium free passivated tinplate (upper image) immersed into milk tea and tomato juice, respectively. Self-cleaning process for (c) soil and (d) graphite powder on the superhydrophobic coating: soil and graphite powder is deposited on the coating, and impinging water droplets wash away the soil and graphite powder to restore a clean surface.