# Electronic supplementary information

# A novel superhydrophobic bulk material

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## Experimental section

### Materials

Pristine multiwalled CNTs with a diameter of 30-50 nm and a length of about 30 μm were purchased from Chengdu Organic Chemicals Co., Ltd., China (synthesized by a CVD process; purity > 99.9 %). PTFE aqueous dispersion (60 wt%) was supplied by Chenguang Research Institute of Chemical Industry, Chengdu of China.

### Preparation of the superhydrophobic bulk material

0.8 g CNTs and 4 g PTFE aqueous dispersion were added in 30 mL of chloroform, and the dispersion was stirred at 50 °C until the chloroform has evaporated to dryness. Then the mixture of CNTs and PTFE was placed into a mold and pressed under a pressure of 256 KPa for 30 min at 390 °C. After cooling, the sample was demolded.

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## Characterization

Contact angle (CA), sliding angle (SA), and contact angle hysteresis (CAH) measurements were performed using a Krüss DSA 100 (Krüss Company, Ltd., Germany) apparatus at ambient temperature. CAH was observed by measuring the difference between advancing angle ($\Theta_\mathrm{A}$) and receding angle ($\Theta_R$) on room temperature on the KRÜSS DSA 100 apparatus. $\Theta_\mathrm{A}$ and $\Theta_R$ were collected with adding and withdrawing from the droplet, respectively. The volume of water droplet in each measurement was approximately 5 $\mu L$. Scanning electron microscopy measurements were carried out using a JSM-6701F field-emission scanning electron microscopy (FESEM, JEOL, Japan). X-ray photoelectron spectroscopy (XPS) characterization was performed on a PHI-5702 electron spectrometer using an Al Ka line excitation source with the reference of C 1s at 285.0 eV. Thermal measurement was carried out using a thermogravimetric analysis instrument (TGA, PerkinElmer PE7) under air atmosphere at a rate of 10/min.

## XPS and FESEM analysis

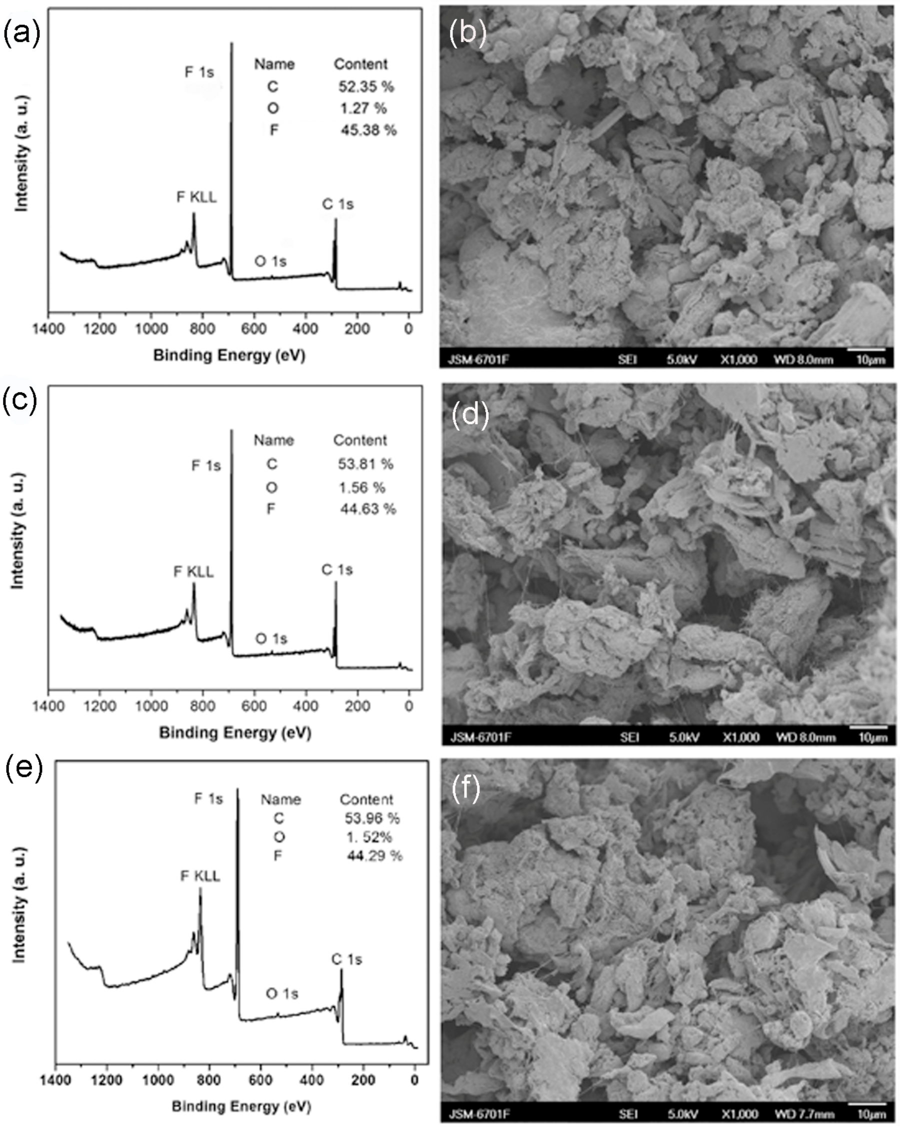

**Figure S1**. XPS survey spectrum and FESEM images for the top surface of the bulk material (a, b), the interior of the bulk material (c, d), and the top surface of the bulk material after the burning process (e, f).

## Shaping and cutting process

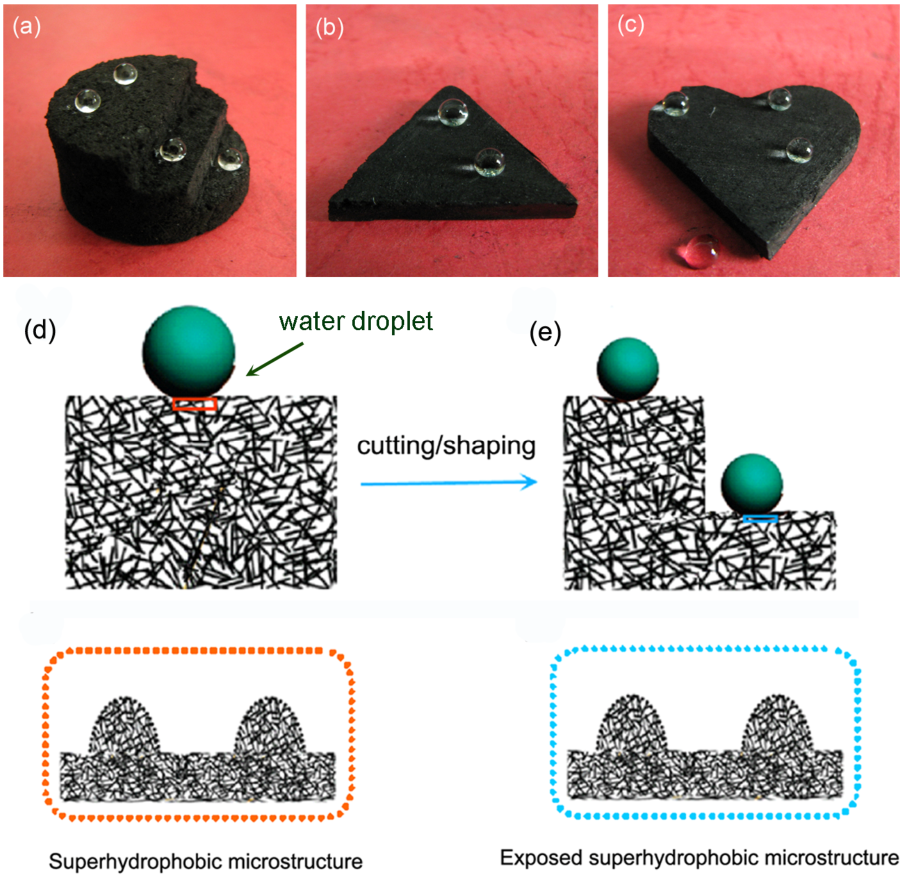

**Figure S2a-c** shows that the bulk material can be cut and machined into desired shapes without sacrifice its original superhydrophobicity. This is because the low-surface-energy microstructures extending throughout the whole volume of the bulk material, which made the freshly exposed surfaces created by cutting and shaping also exhibit superhydrophobic (see Figure S2d and e). The superhydrophobicity is therefore retained even after accidental damage.

More importantly, when the bulk material lost its superhydrophobicity after a long abrasion process, it can restore superhydrophobic once more just by cutting the outmost layer.

## Scratch test

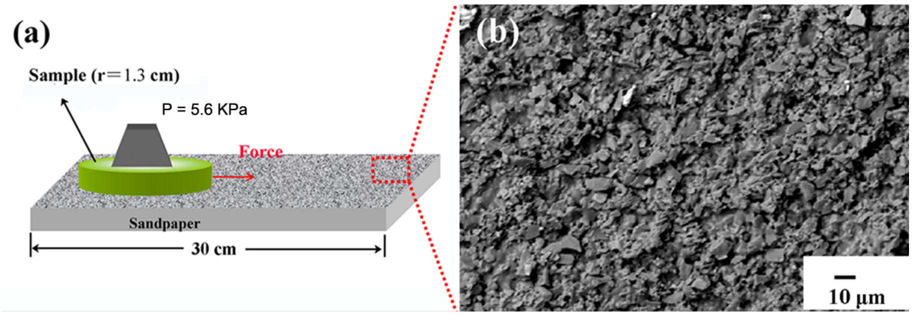

**Figure S3.** (a) Schematic illustration of the methodology of the scratch test. (b) SEM image of the sandpaper.

Figure S3 illustrates the methodology of the scratch test: 1500-mesh sandpaper served as an abrasion surface, with the bulk material surface to be tested facing this abrasion material. While a pressure (5.6 KPa) was applied to the bulk material, the bulk material was dragged in one direction with a speed and abrasion length of 3 cm/s and 30 cm, respectively.

## The reusability of the bulk material

### i) The recycling of surface wettability

The bulk material lost its superhydrophobic properties after contamination with oil. However, the oil-fouled bulk material can regenerate the superhydrophobicity once more just by burning the oil in air. As shown in Table S1, the superhydrophobic properties of the bulk material differed slightly even with repeated contamination/regeneration cycles.

**Table S1.** The value of contact angle, sliding angle, and contact angle hysteresis as a function of the number of contamination/regeneration cycles.

| Contamination/regeneration cycles | Contact angle | Sliding angle | Contact angle hysteresis |
|---|---|---|---|
| 0 | 159 ° | 3 ° | 2.3 ° |
| × 1 | 159 ° | 3 ° | 2.2 ° |
| × 3 | 159 ° | 2.8 ° | 2.0 ° |
| × 5 | 160 ° | 2.8 ° | 1.8 ° |
| × 10 | 159 ° | 2.8 ° | 1.9 ° |

### ii) The recycling of oil-absorption capacity

The absorption capacity values, $W$ (wt/wt), were obtained by measuring the mass of the dry as-produced bulk material, and then measuring the mass of the absorbed oil. The ratio of the absorbed oil mass to the initial bulk material mass was taken as the $W$

(wt/wt) value, averaging out three samples. To ensure full saturation was obtained before weighing, the samples were left submerged in the oil (without water) overnight. The samples were then removed with sharp needle tweezers and immediately placed onto a weigh paper to be measured on the mass balance. Then the oils absorbed in the bulk material were removed by burning in air, and the burned material was used to repeat the above cycles. The oil-absorption capacity of the bulk material for gasoline, hexadecane, coal oil, and diesel was 3.69, 3.74, 3.78 and 3.95, respectively. The variation in absorption predominantly depends on the density of oil solvents, and increasing solvent density results in higher absorption capacity.

Importantly, as shown in Figure S4, the oil absorption capacity of the bulk material differs slightly even after 10 cycles of oil-removal cycles, demonstrating the stability of this oil sorbent material. Thus, our created bulk material is a good candidate in industrial oil-polluted water treatment and oil spill cleanup.

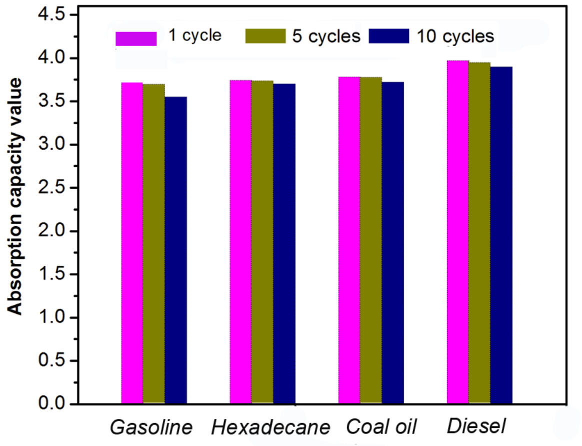

**Figure S4.** Oil-absorption capacities of the bulk material for four kinds of oils after different absorption/regeneration cycles.

## Contrast test

We also prepared CNTs and PTFE bulk materials, following the experimental procedure of the CNTs-PTFE bulk material. As shown in Figure S5a, the obtained CNTs bulk material was incompact and was fragile to mechanical contact. Moreover, the obtained PTFE bulk materials can only reach to hydrophobic state, with the CA value of 120$^{o}$ (see Figure S5b). The PTFE bulk material is superoleophilic to hexadecane, and when fouled by hexadecane, the PTFE bulk material can restore to its original hydrophobic properties after burning out the oil in air. Thus, it is the synergistic action of CNTs and PTFE that leads to a bulk material with superhydrophobicity, mechanical stability and easy reusability.

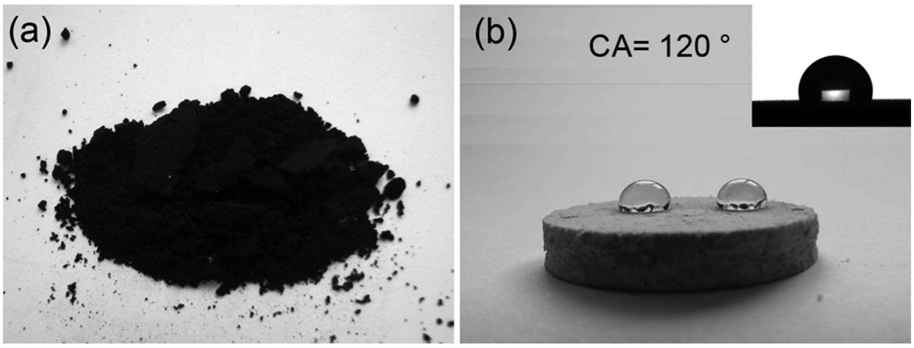

**Figure S5.** Optical image of the as-prepared CNTs (a) and PTFE (b) bulk materials. The obtained CNTs bulk material was fragile to mechanical contact, indicating its limited mechanical property. The obtained PTFE bulk material can only reach to hydrophobic sate.

**Video S1.** Video for the water-repelling and oil-attracting properties of the bulk material.