Supporting Information

Synthesis of Freestanding Amorphous Giant Carbon Tubes with Outstanding Oil Sorption and Water Oxidation Properties

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Experimental Section

***Preparation of graphene foam (GF)*:** Ni foam was used as template for CVD grow of graphene foam after being washed in water and ethanol respectively by sonication to eliminate contaminants residing on its surface. It was placed in a quartz tube of outer diameter 25 mm and inner diameter 22 mm, and was heated to 1000 ^{o}C at a heating rate of 5 ^{o}C min^{-1} in a horizontal tube furnace under H_2 (120 s.c.c.m. H_2 5%, Ar 95%) and kept at this temperature for 1 h to clean its surface and remove Ni oxides. After that, the H_2 flow (120 s.c.c.m. H_2 5%, Ar 95%) was reduced to 60 s.c.c.m and a small amount of CH_4 was simultaneously introduced into the reaction tube at ambient pressure. The flow rate of CH_4 is 15 s.c.c.m, corresponding to concentration of 20 vol% in the total gas flow. After 1 h of reaction-gas mixture flow, the samples were cooled to room temperature under H_2 (60 s.c.c.m. H_2 5%, Ar 95%). Then the sample was put into a $FeCl_3$ (1 M) and HCl (1 M) solution at room temperature for an overnight to completely dissolve Ni foam to obtain graphene foam (GF).

***Pump oil sorption measurement of GF*:** GF was soaked in pump oil and maintained for 24 h to measure the saturation adsorption capacity. The oil-adsorption capacity of GF was calculated by the ratio between the maximum adsorbed oil quantity m_{oil} (g) and the GF mass m_{GF} (g) according to the following equation:

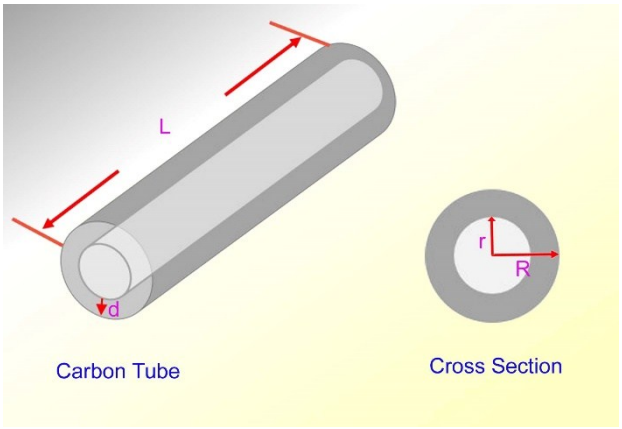
$$q = \frac{m_{oil}}{m_{GF}} = \frac{m_f - m_{GF}}{m_{GF}} \quad (1)$$

Where q is the sorption capacity (g/g), m_f is the weight (g) of the wet GF the moment when the liquid drops don't drip in succession during naturally draining.

Characterization

Contact angle was measured on a JC2000C1 Contact Angle Measurement (Shanghai Zhongchen Digital Technique Equipment Co., Ltd., China) at room temperature. Energy dispersive spectroscopy (EDS) and element mapping were performed by a field-emission scanning electron microscope (FE-SEM, Quanta 200). X-ray photoelectron spectra (XPS) of quartz wool, GCTs and N-GCTs were obtained using a PHI 5000 Versaprobe II X-ray Photoelectron spectrometer.

Calculation of wall thickness of freestanding GCTs



Scheme S1. Schematic demonstration of GCT structure.

According to GCT structure (Scheme S1), the following equations are given out.

$$V = (\pi R^2 - \pi r^2) \cdot L \quad (2)$$

$$S = 2\pi R \cdot L + 2\pi r \cdot L \quad (3)$$

$$R = r + d \quad (4)$$

$$m = \rho V \quad (5)$$

Where d is wall thickness of GCT, L is length of GCT, R is radius of GCT, r is radius of hollow structure of GCT, m is mass of carbon tube, V is volume of carbon in GCT, ρ is density of carbon (2 g cm^{-3}), and S is surface area of GCT. The value of d is obtained and equal to 21 nm

after the deduction of the above four equations under the premise that the value of *d* is much smaller than that of *r*.

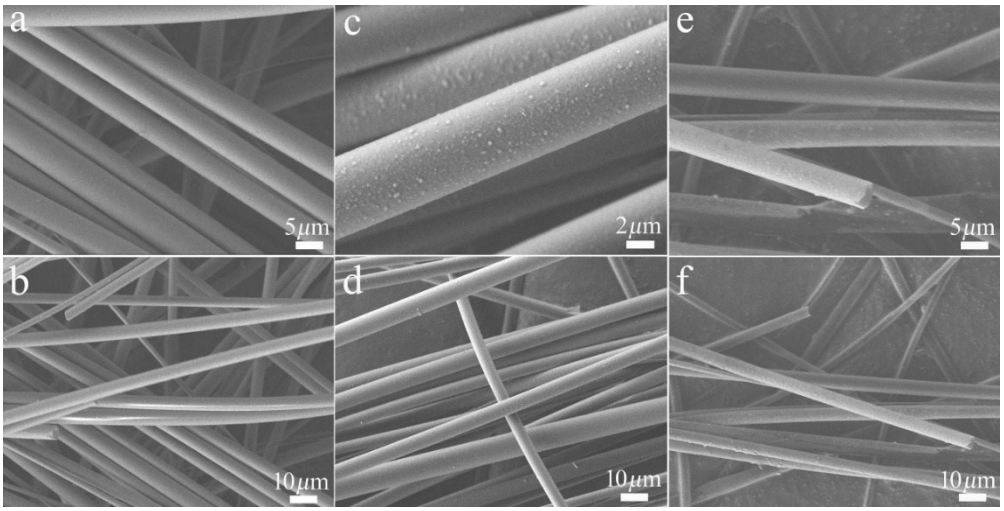


Fig. S1 FESEM images of (a, b) quartz wool, (c, d) quartz wire@carbon tubes composite, and (e, f) GCTs obtained by removing quartz wires.

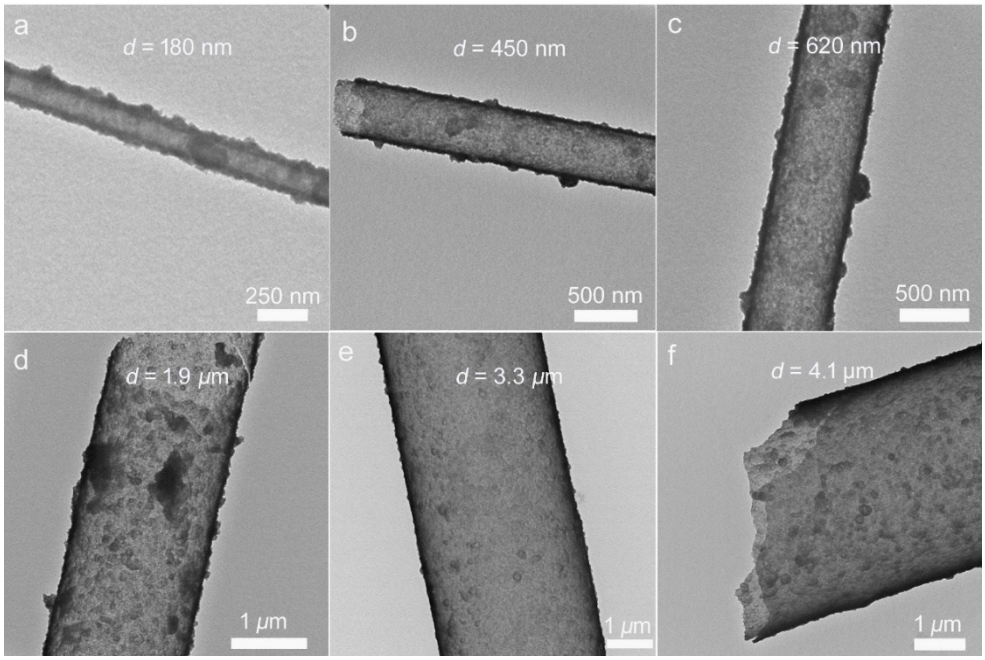


Fig. S2 TEM images of a few GCTs with different diameters.

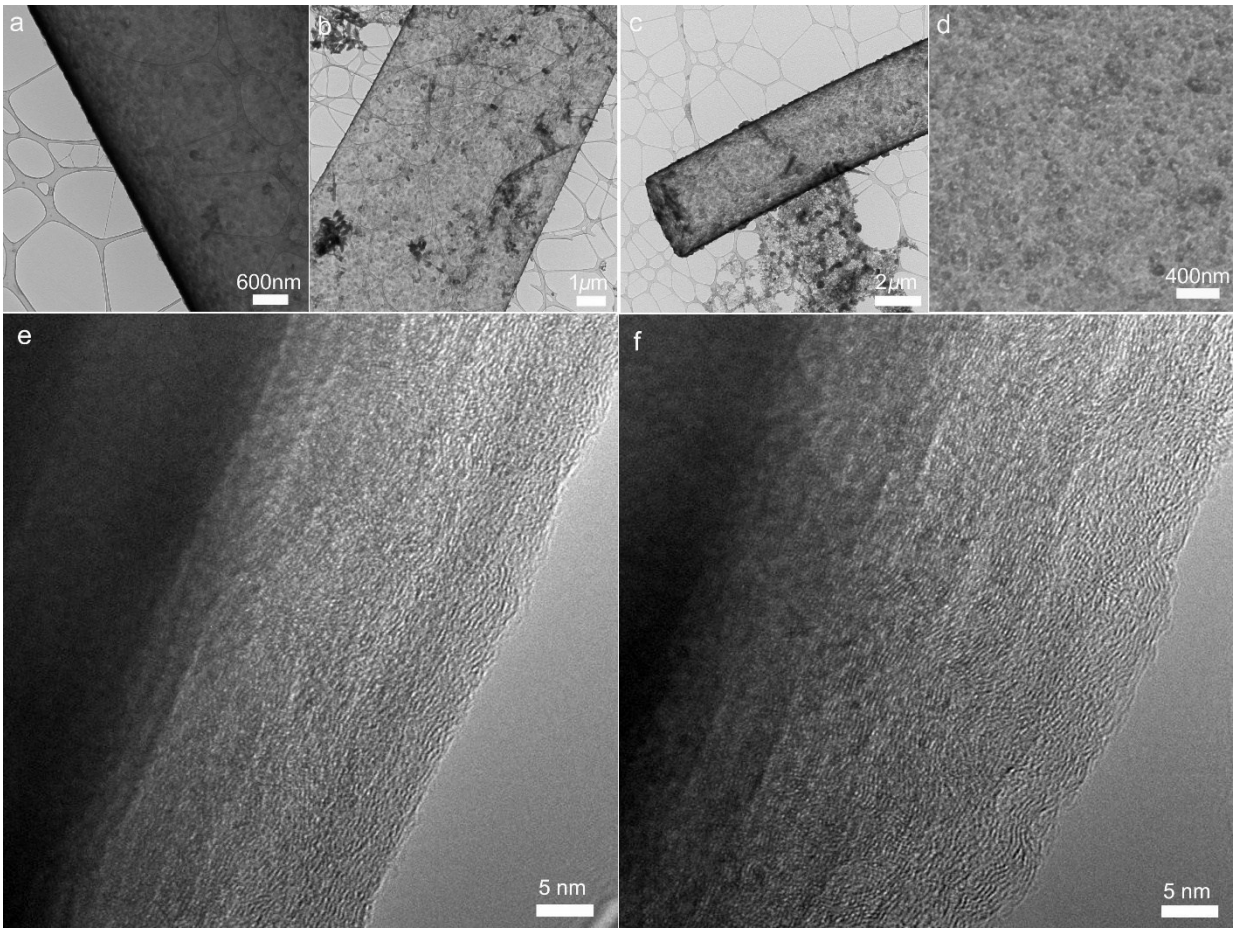


Fig. S3 TEM images of (a-c) GCTs and (d) single layer wall of a GCT (The upper surface is interior surface of GCT); (e, f) HRTEM images of GCTs.

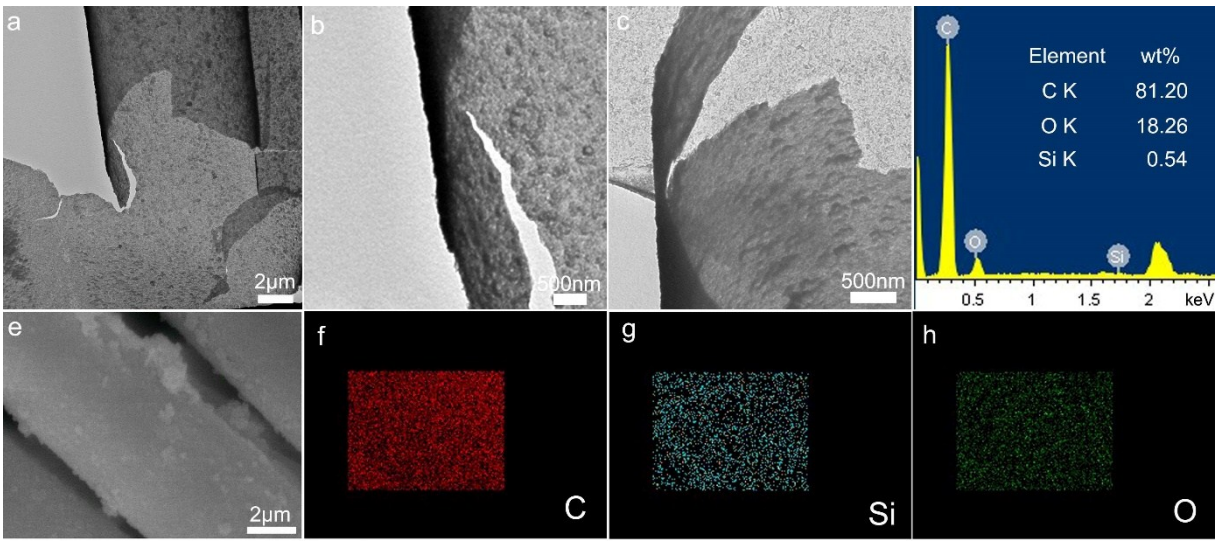


Fig. S4 (a-c) TEM images of cross-section, (d) EDS, (e) SEM image, and (f-h) element mapping of GCTs.

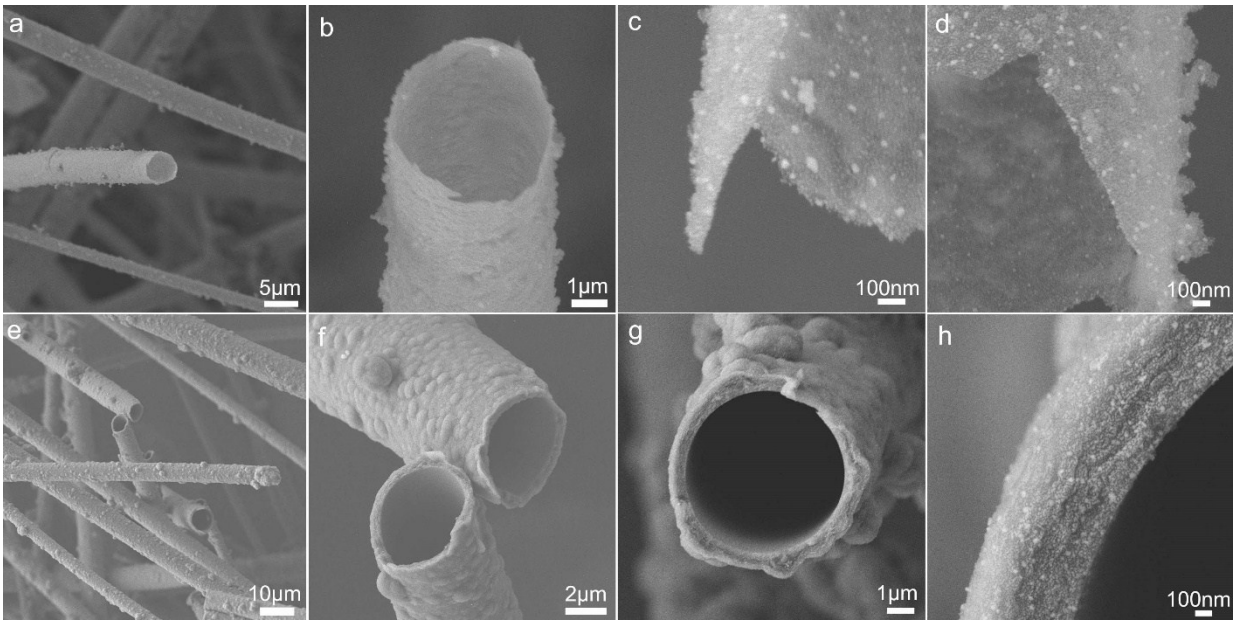


Fig. S5 FESEM images of GCTs prepared at (a-d) 950 °C and (e-f) 1050 °C.

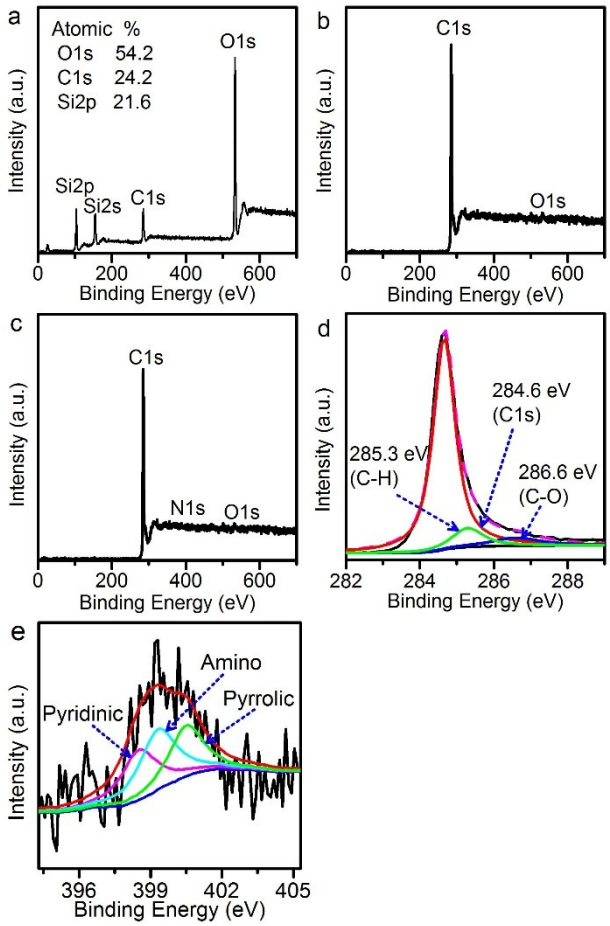


Fig. S6 XPS spectra of (a) quartz wool, (b) GCTs and (c) N-GCTs; High-resolution XPS spectra of (d) C1s and (e) N1s.

XPS spectra of quartz wool, GCTs and N-GCTs were shown in Fig. S6. The Si2p binding energy of about 103.5 eV corresponded to SiO_2 and the oxygen atoms were beyond stoichiometric ratio of O to Si for SiO_2 (Fig. S6a).[1] The XPS C1s core-level spectrum indicated the characteristic signals of crystalline carbon of GCTs (Fig. S6b and S6d). [1] After nitridation, the N1s peak was observed and could be best fit with three subsets of peaks at 398.5 eV, 399.3 eV and 400.5 eV (Fig. S6c and S6e), corresponding to the pyridine-like structure nitrogen, amino nitrogen and pyrrolic nitrogen, respectively.[2,3]

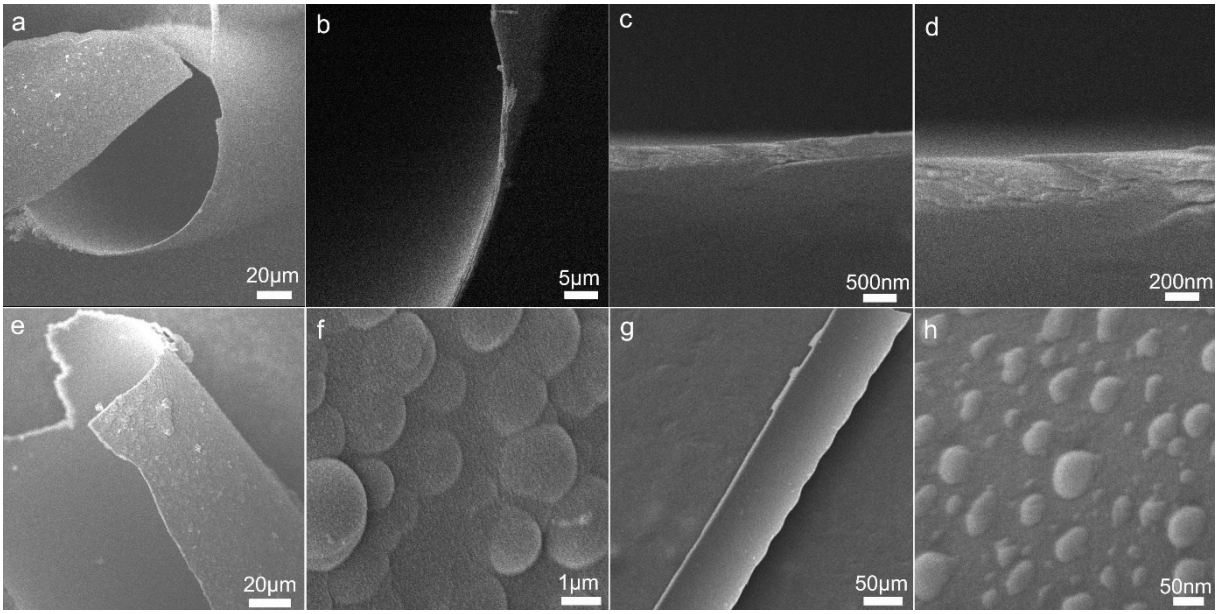


Fig. S7 FESEM images of (a-d) cross-section, (e, f) external surface and (g, h) internal surface of GCT-125 prepared at 1100 ^{o}C using quartz wires (125 μm).

FESEM images of the ultralong GCTs made from macroscopic quartz wires (125 μm) at 1100 ^{o}C were shown in Fig. S7. The characterizations indicated perfect tubular structure of the GCTs despite a large inner diameter of 125 μm (Fig. S7a), suggesting their structural robustness. It could be attributed to their thickness of about 300 nm (Fig. S7b-d). Formation of micrometer bulges and nanometer bumps was observed on the internal surface (Fig. S7e and S7f), which could be resulted from variation of chemical components or crystallinity in different

regions of quartz wire surface that caused different activity in catalyzing epitaxial growth of carbon.

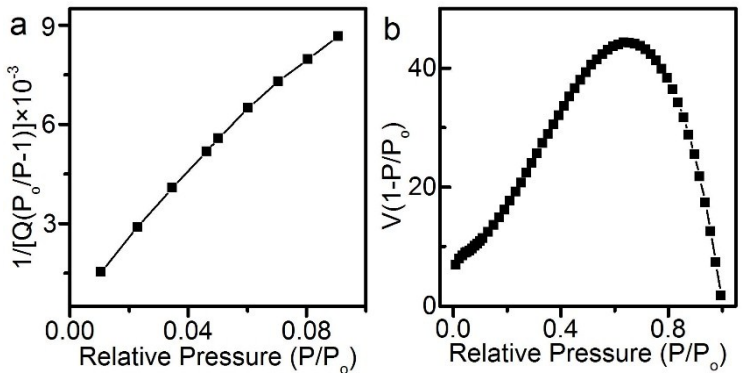


Fig. S8 (a) BET surface area plot and (b) calculated Rouquerol plot of GCTs.

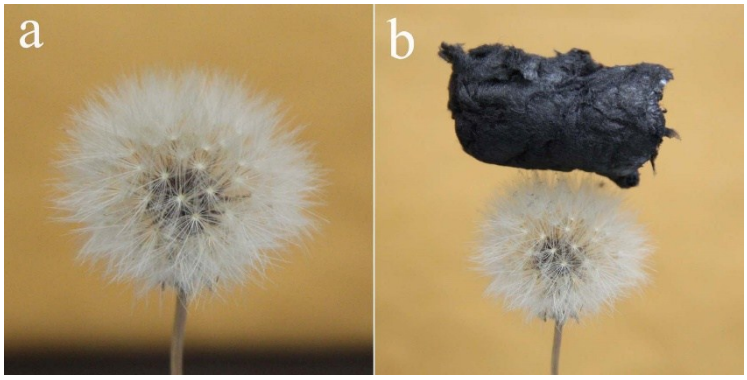


Fig. S9 Optical images of (a) mature dandelion, (b) freestanding GCT mat lying on the villus of mature dandelion floret with no shape change of villus, showing extremely low density.

Fig. S10 Water contact angle measurement of GCT mat. A contact angle of 121 ° for water on the GCT mat showed good hydrophobicity.

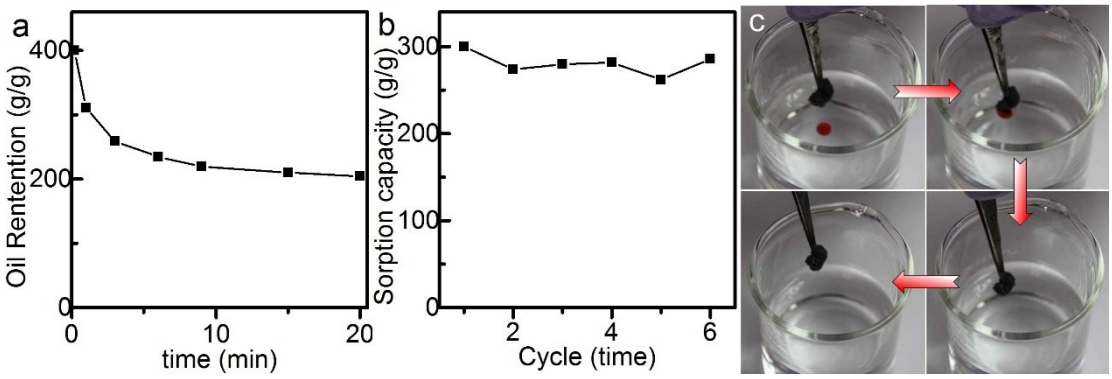


Fig. S11 (a) Retention capability of GCTs for pump oil. (b) Recyclability of GCTs. Sorption capacities of ethanol using GCTs and released its vapor under heat treatment (80 °C) for 6 cycles. (c) Demonstration of oleic acid (dyed with Sudan III) adsorption on the surface of water by using a piece of GCT mat.

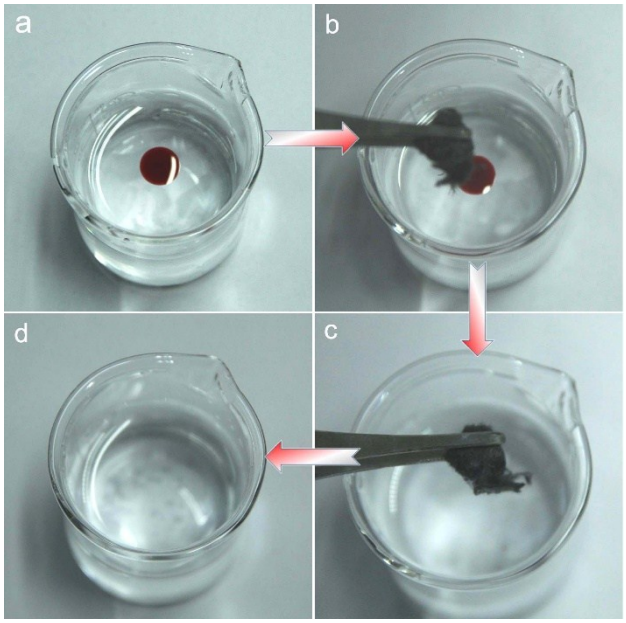


Fig. S12 Demonstration of diesel (dyed with Sudan III) adsorption on the surface of water by using a piece of GCT mat.

Movies: (S1) Side-view of dyed chloroform-water separation

(S2) Top-view of dyed oleic acid-water separation

References

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2 L. Zhou, X. Qu, D. Zheng, H. Tang, D. Liu, D. Qu, Z. Xie, J. Li and D. Qu, ACS Appl. Mater. Interfaces 2017, 9, 41332-41338.

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