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Ambient Pressure Dried Graphene Aerogels with Superelasticity and Multifunctionality

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Synthesis of Graphene Oxide (GO)

GO was synthesized from natural graphite (325 mesh) via a modified Hummers' method. Briefly, 1 g graphite powder, 3 g $KMnO_4$ and 60 mL concentrated H_2SO_4 solution without mix was respectively introduced in a refrigerator (-18 °C) for 30 min. Then the graphite powder, $KMnO_4$ and concentrated H_2SO_4 solution were successively introduced in a flask (250 mL). The mixture was stirred for 2 h and the temperature was controlled below 20 °C in ice bath. After these steps, the mixture was moved to oil bath at 50 °C and sequentially stirred for 6 hours. The result mixture was slowly poured into a 500 mL beaker with 120 mL of distilled water. The 30 % H_2O_2 solution was dropwise added into the mixture until no bubbling. Finally, the crude product was washed repeatedly with deionized water by centrifuge until the filtrate became neutral. The obtained brown-yellow graphene oxide was saved for use.

Fabrication of Ambient Dried Graphene Aerogel (ADGA)

In a typical procedure, graphene oxide (method, see Electronic Supplementary Information) dispersion (4 mg mL^{-1}, 10 mL) was first mixed uniformly with L-ascorbic acid (80 mg) by stirring and then sealed in a glass vial and heated for 1 h at 95 °C for synthesis of partially reduced graphene hydrogel (PRGH). Secondly, the obtained hydrogel was cooled at the room temperature and then accepted the

freeze-thaw process (FTP) in the freezer (-18 °C) and room temperature. Thirdly, the freeze-recast hydrogel was further reduced (95 °C) by the initial reductant to get completely reduced graphene hydrogel (RGH) by L-AA. Finally, the RGH was subjected to dialysis in water (to remove soluble species) and dried in the oven at 60 °C for 24 hours to obtain the ambient pressure dried graphene aerogel (ADGA).

Details of Characterization Methods

The density of ADGA was calculated by the weight and volume of cylindrical sample. The X-ray diffraction (XRD) patterns were conducted at room temperature using a Bruker D 8 X-ray diffractometer. Scanning electron microscopy (SEM) of monolith structures was performed using a JSM-7500F field emission SEM at an accelerating voltage of 5 kV. The different view sections for SEM observation were cut by a very thin blade. Transmission electron microscopy (TEM) analysis was conducted on a JEOL JEM-2200 FS instrument with an accelerating voltage of 200 kV. The graphene flakes were torn off from the ADGA and wrapped up by epoxy solution. Then the ultrathin sections from the cured epoxy include the ADGA flakes were observed by TEM. The XPS were obtained with an ESCA Lab 220i-XL electron spectrometer. Elemental analysis was conducted by Elemental Analyzer Vario EL III. The pr-GO solutions (reduction time is 15 min and 25 min) were dried by vacuum filtration and the pr-GO hydrogels (reduction time is respectively 30 min, 35 min, 45 min, 60 min and 120 min) were subjected to dialysis in water (to remove soluble species) before the XPS and elements analysis tests. The Raman analysis was recorded with a Renishawin Via Raman microscope (Britain) using an excitation wavelength of 532 nm. The compressive tests were performed in an Instron (Micro Tester, 5940, Instron) using a 10 N load cell and strain control mode with a strain rate of 3 % per second. The electrical conductivity was determined by a two-probe method using a digital multimeter (VC 890D. Shenzhen Victor Hi-tech Co. Ltd.).

Table S1. Elemental analysis results of graphene solid.

Reduction time of GO	C [wt%]	H [wt%]	O [wt%]	C/O (atomatic ratios)
0 min	46.57	2.53	50.90	1.22
15 min	48.38	2.66	48.96	1.32
25 min	49.88	2.57	47.55	1.40
30 min	57.46	2.28	40.26	1.90
35 min	64.29	2.07	33.64	2.55
45 min	65.75	2.05	32.20	2.72
60 min	68.07	1.78	30.15	3.01
2 h	68.67	1.85	29.48	3.11
6 h	86.31	1.02	12.67	9.08

Fig. S1 The influence of GO concentration on the volume of RGH (up) and ADGA (down). The concentration was from 1 to 5 mg mL^{-1}.

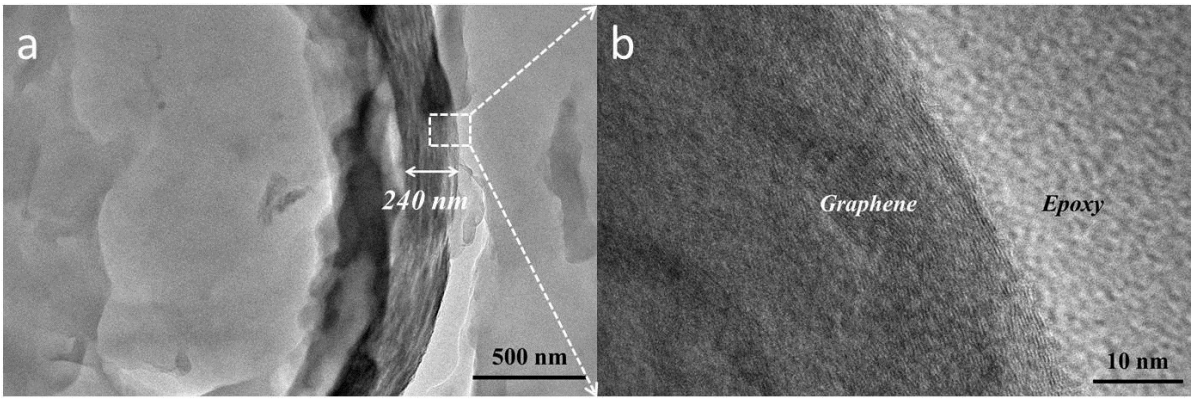

Fig. S2 TEM images of a typical cell wall of the ADGA. The density of this sample is 8.1 mg cm^{-3}.

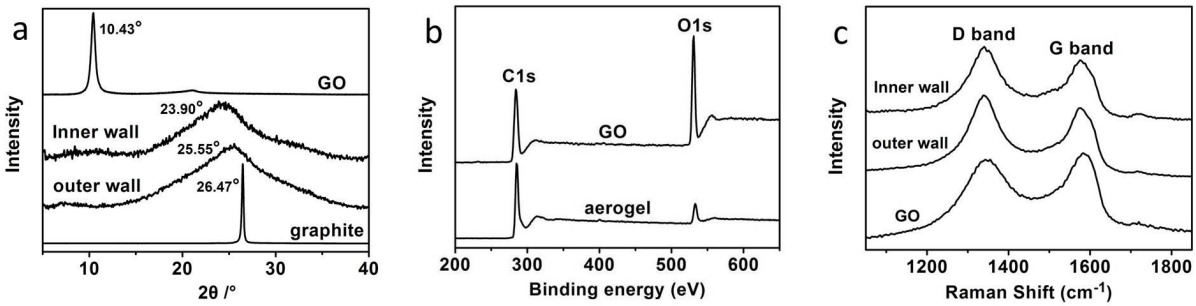

Fig. S3 (a) XRD patterns of GO, ADGA (inner wall and outer wall) and graphite. (b) XPS wide scan spectra of GO and the ADGA. (c) Raman spectra of GO and ADGA (inner wall and outer wall).

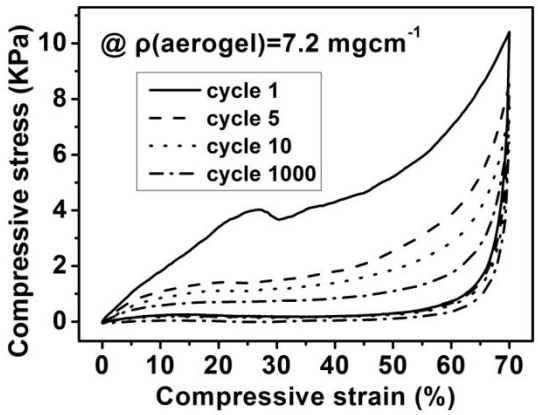

Fig. S4 The ADGA (7.2 mg mL^{-1}) was compressed at strain of 70% for 1000 cycles. It shows the stress–strain curves of cycle 1, cycle 5, cycle 10 and cycle 1000.

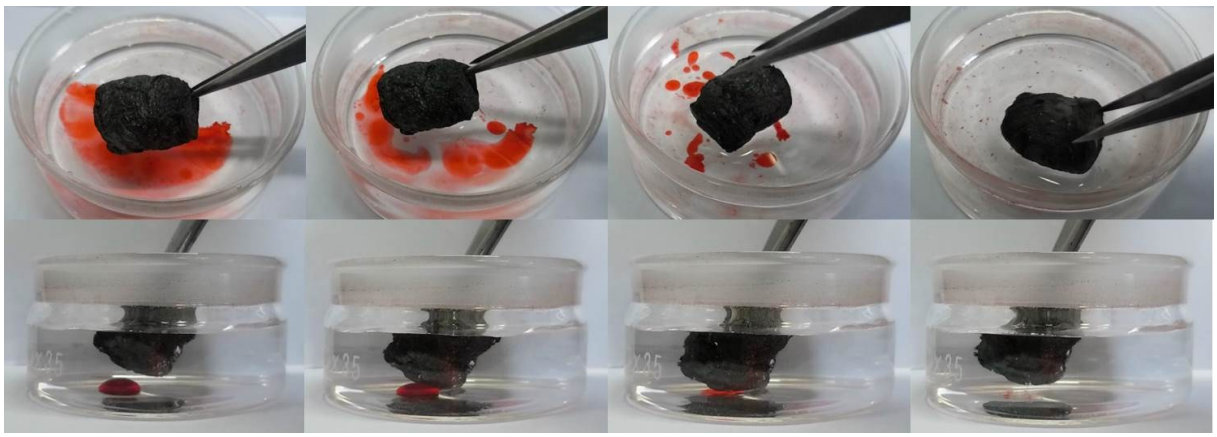

Fig. S5 The photos reveal the removal process of n-decane (dyed with Oil Red O) floating on water (up) and dichloromethane (dyed with Oil Red O) sinking below the water (down) using the ADGA.

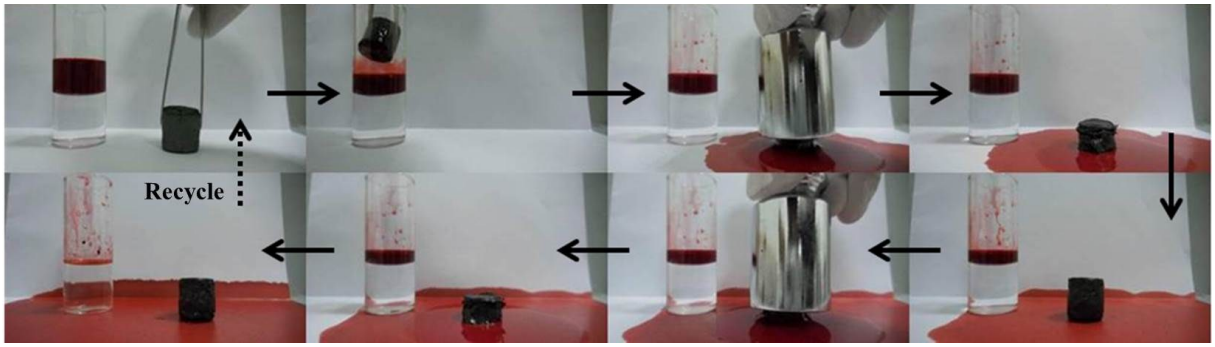

Fig. S6 Snapshots of the process of the absorption–squeezing cycles for oil (n-decane, dyed with Oil Red O) clear and collection using the ADGA. It indicates the ADGA is recycled and high efficient for oil clear and collection.