Electronic Supplementary Material (ESI) for Materials Horizons
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Supporting Information for

Dendrimer-linked, Renewable and Magnetic Carbon Nanotube Aerogels

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Experimental

Synthesis of PAMAM crosslinked MWCNT aerogel: The pH value of the as-prepared MWCNT solution (10 mg·mL^{-1}) was adjusted to 8.0 by adding an appropriate amount of NaOH solution (2 moL·L^{-1}) dropwise. Then 3 mL above MWCNT solution was mixed with 200 μL PAMAM solution with the concentration of 30 mg·mL^{-1}. The mixture was stirred and sonicated for about 5 min to mix them well. After that, 30 mg GDL containing 200 μL aqueous solutions was added to the above mixture and sonicated for 30 s to initiate the gelation. Then the mixture was stayed put at ambient temperature until a uniform gel was obtained. The resulting hydrogel was washed with a large amount of deionized water for about one week to remove the hydrolysis product of GDL and then freeze-dried to obtain the MWCNT aerogel. Or the purified hydrogel was further turned into the alcogel by using alcohol to replace the water within the network of the hydrogel, and then dried with supercritical CO_2 to obtain MWCNT aerogel.

Synthesis of MWCNT/Fe_3O_4 composite aerogel: First, the Fe_3O_4 sol was prepared according to the procedure reported elsewhere [31]. The MWCNT/Fe_3O_4 composite aerogels were synthesized by the similar process to MWCNT aerogel, but the main difference between them

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is just by adding 150 μL 18 mg·mL^{-1} Fe_3O_4 sol to the MWCNT aerogel to obtain MWCNT/Fe_3O_4 composite aerogels.

Dye adsorption-desorption: In a typical experiment, 7 mg as-prepared MWCNT aerogel or the as-prepared hydrogel with 7 mg solid content were added into 50 mL of 20 mg·mL^{-1} MB aqueous solution followed by slowly stirring at room temperature. At a series of intervals, the mixture was sampled and the dye concentration remaining in the mixture was measured after centrifugation. The dye concentration was determined through a UV/vis spectrometer and calculated by the standard spectrophotometric method at the maximum absorbance of the dye. The amount of dye qt (mg·g^{-1}) absorbed with the MWCNT aerogel/hydrogel at time t, was calculated using the following equation:

$q_t = \frac{(C_0 - C_t) \times V}{M}$, where C_0 and C_t (mg·L^{-1}) are the initial and time t concentration of the dye, respectively, while V is the volume of dye solution (L) and M is the mass of the MWCNT aerogel/hydrogel used in this case (g). The pseudo-first-order kinetic equation is presented as: $\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t$, and pseudo-second-order kinetic equation is given as: $\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e}$, where q_e and q_t (mg·g^{-1}) are the dye amount adsorbed with the MWCNT aerogel/hydrogel at equilibrium and time t, respectively; k_1 and k_2 represent the pseudo-first-order (min^{-1}) and pseudo-second-order (g·mg^{-1}·min^{-1}) rate constant, respectively.

For selective adsorption, 7 mg as-obtained MWCNT aerogel were added into 50 mL RhB (4 mg·L^{-1}) and MB (4 mg·L^{-1}) mixed solution followed by slowly stirring at room temperature for 24 h to reach adsorption equilibrium. For desorption, PAMAM has been chosen as desorption agent to investigate the desorption behavior of the loaded dye within the MWCNT aerogel. After adsorption completely, an appropriate amount of PAMAM (with the mass ratio of PAMAM to aerogel 2:1, 10:1, 50:1, and 200:1, respectively) was added into above

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suspension to desorb the dye from the MWCNT aerogel matrix, and to promote desorption, 400 μL concentrated HCl was also added at the beginning of the desorption experiment.

Instrumentation: The structure of the obtained samples were characterized by XRD using monochromatic Cu Kα1 radiation (λ = 1.5406 Å) at 40 kV and 40 mA. The diffraction patterns were optimized with a step length of 0.001 ^{o} (2θ) over an angular range 10~70 ^{o} with a scanning speed of 0.01 ^{o}/s. The Raman spectra were measured on a Renishaw System 1000 with a 50 mW He-Ne laser operating at 514 nm with a CCD detector. SEM images were conducted at a Hitachi S-4800 field-emission-gun scanning electron microscope at 5-10 KV .The samples for SEM were prepared by putting samples on the conductive tape and then sprayed with Platinum. TEM was performed on a FEI Tecnai 20 at 20 KV. The BET specific area, pore size distribution and total pore volume data were obtained using ASAP 2010 (Micromeritics, USA) measurements at 77 K. Before measurement, all samples were degassed under vacuum at 100 ^{o}C for 10 h. UV spectroscopy was performed on UV-6100 double beam spectrophotometer (Shanghai Mapada).

Tables

Table SI1. Porous attribute of the resulting MWCNT and MWCNT/Fe_3O_4 aerogels

Aerogel		BET surface area (m^2/g)	Total pore volume (cm^3/g)	Average pore diameter (nm)
MWCNT	Sc CO_2 drying	154	0.30	7. 8
	Freeze drying	143	0.65	18.1
MWCNT/Fe_3O_4	Sc CO_2 drying	182	0.46	9.7
	Freeze drying	196	0.44	10.5

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Figures

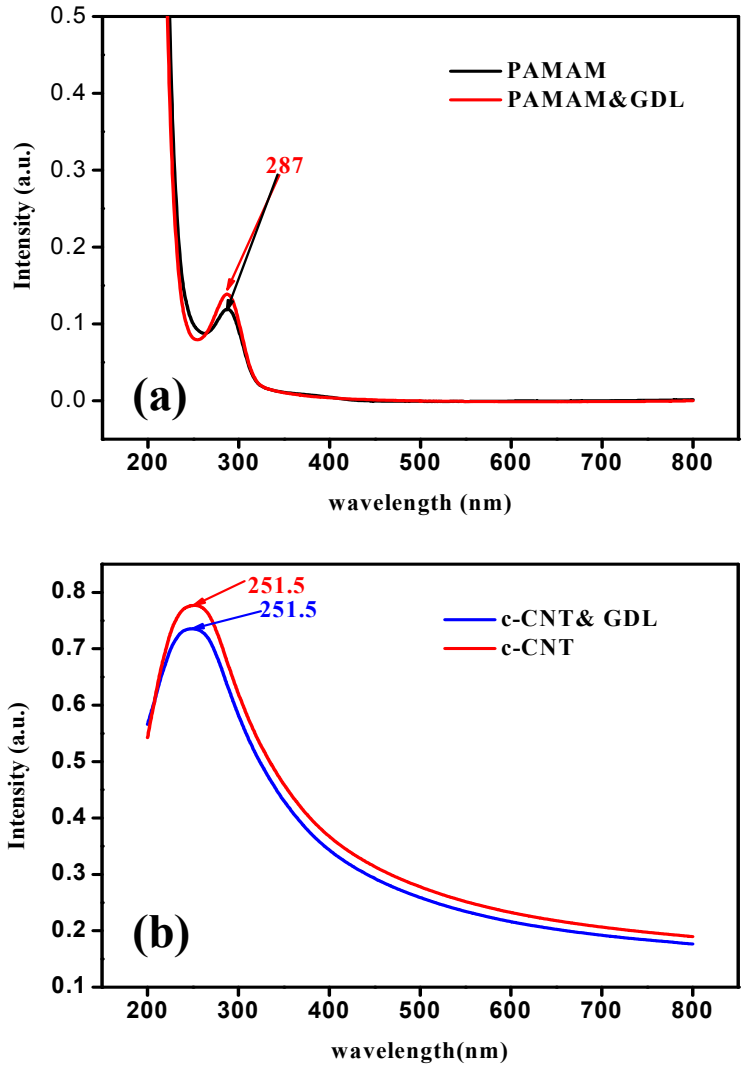

Figure SI1 UV-vis spectra of the mixtures between GDL and PAMAM (a) or between GDL and carboxylic carbon nanotubes (b). Both of the UV spectra shows no obvious peak shifts, revealing that GDL has no interaction with both PAMAM and MWCNTs, which guarantees that superfluous GDL and its hydrolyzed product can be removed completely from the resulting hydrogel after solvent-exchange process.

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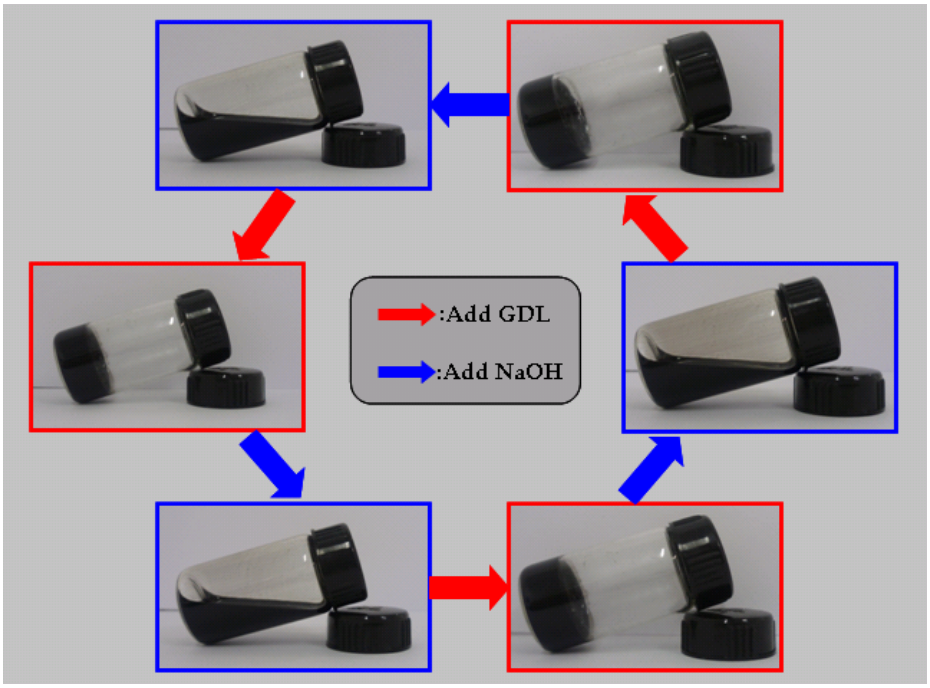

Figure SI2. Reversible sol-gel transition of the PAMAM-crosslinked MWCNT/Fe_3O_4 composite hydrogel

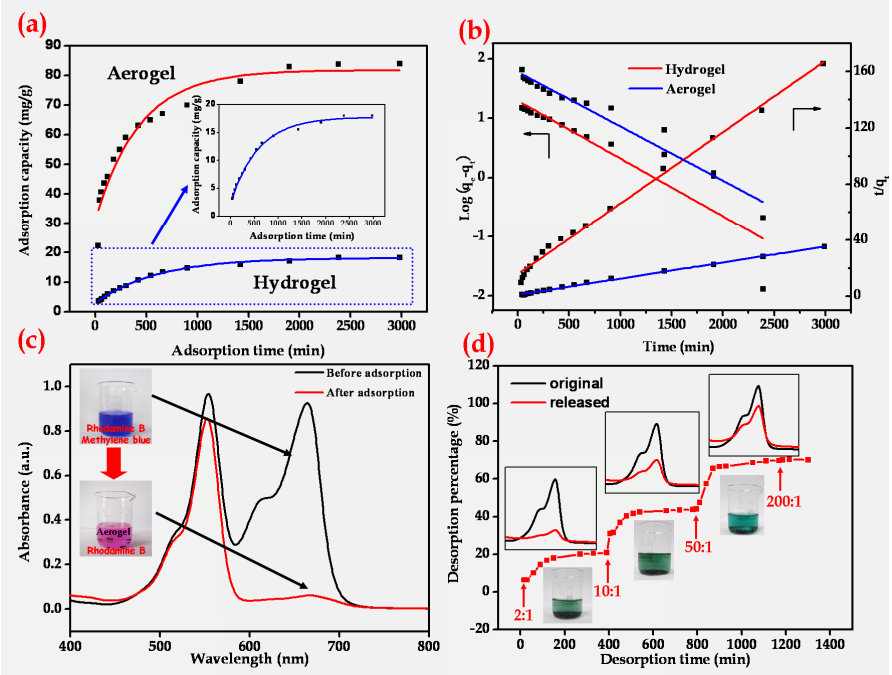

Figure SI3. Adsorption capacities at intervals (a) and corresponding kinetic model fits (b) of the dendrimer-crosslinked MWCNT hydrogel and aerogel as the adsorbent for MB removal, selective adsorption behavior (c) of the MWCNT aerogel toward the mixture of the MB and RhB, and controllable dosorption behavior (d) of the MWCNT aerogel through addition of the dendrimer PAMAM.

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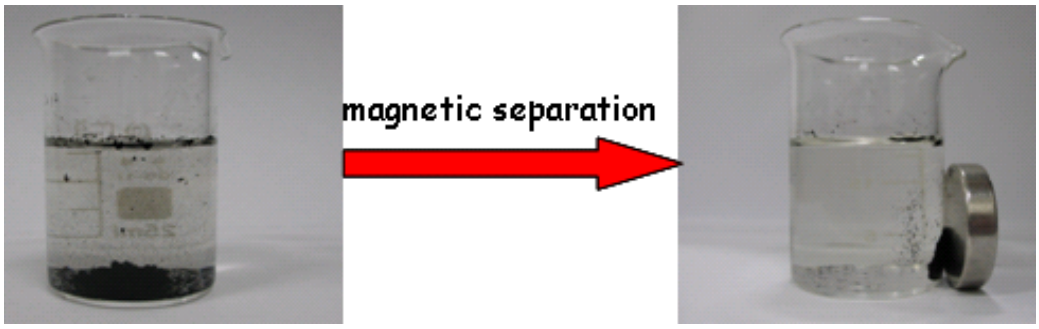

Figure SI4 Magnetic separation of the MWCNT/Fe_3O_4 composite aerogel from the aqueous solution after adsorption of the organic dye molecules.

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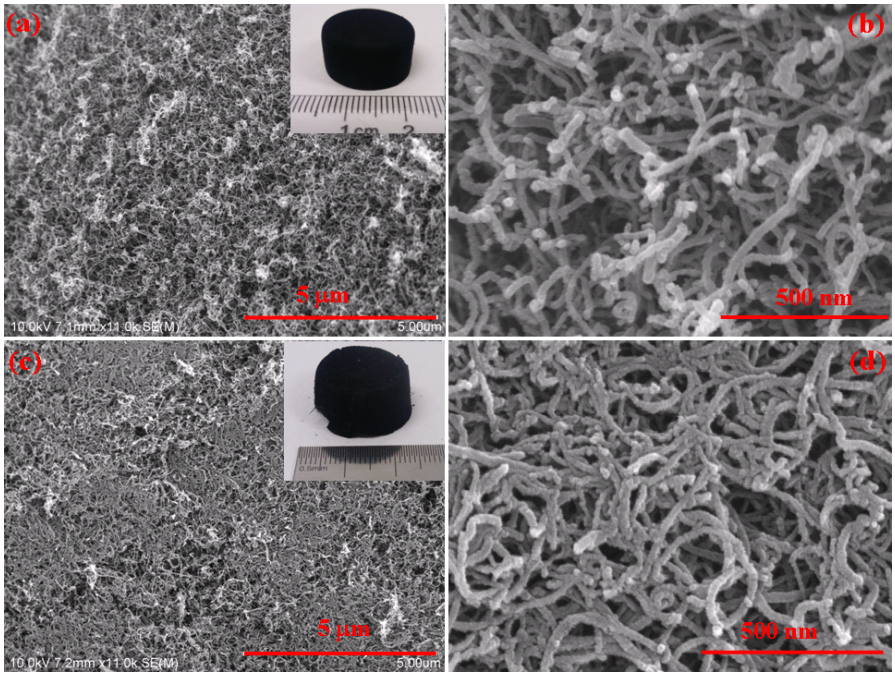

Figure SI5. Typical SEM images of the resulting carbon nanotube aerogels processed with supercritical CO_2 drying (a, b) or freeze drying (c, d). Insets in (a) and (c) are the digital photos of the monolithic carbon nanotube aerogels processed with supercritical CO_2 drying and freeze drying, respectively

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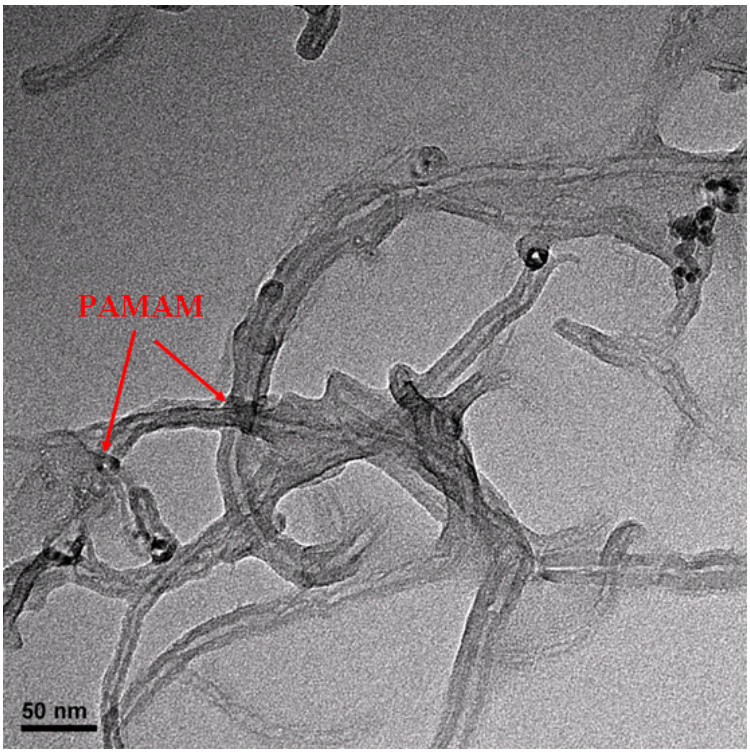

Figure SI6 TEM image of the resulting PAMAM-crosslinked MWCNT hydrogel.

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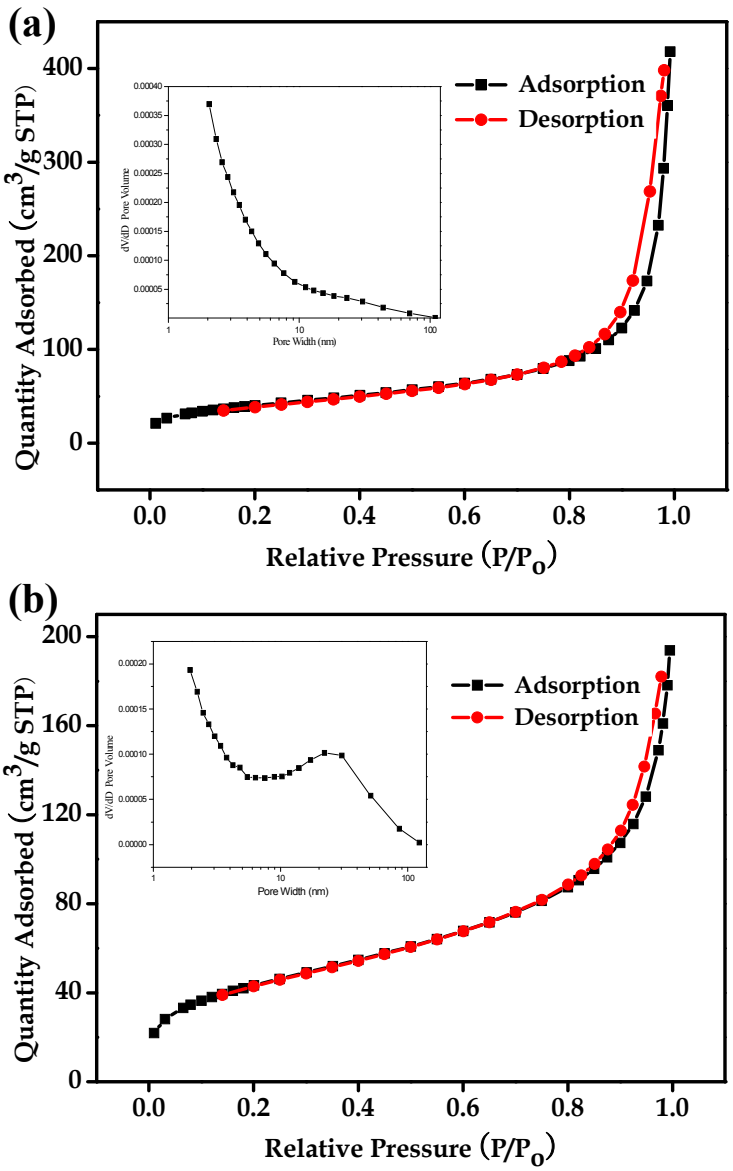

Figure SI7 Sorption isotherms and corresponding pore size distribution curves (insets in each picture) of the supercirtical CO_2 dried (a) and freeze dried (b) MWCNT aerogels

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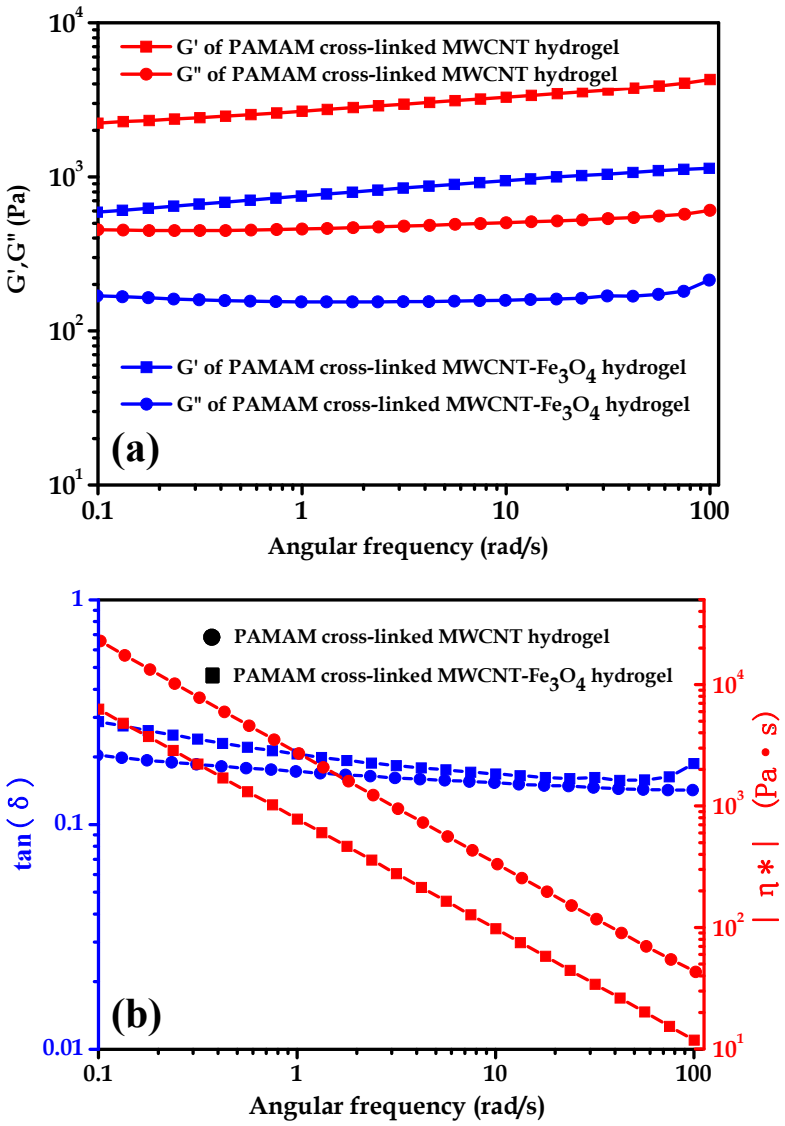

Figure SI8 Rheological investigation on the as-made MWCNT hydrogel and MWCNT/Fe_3O_4 composite hydrogel.

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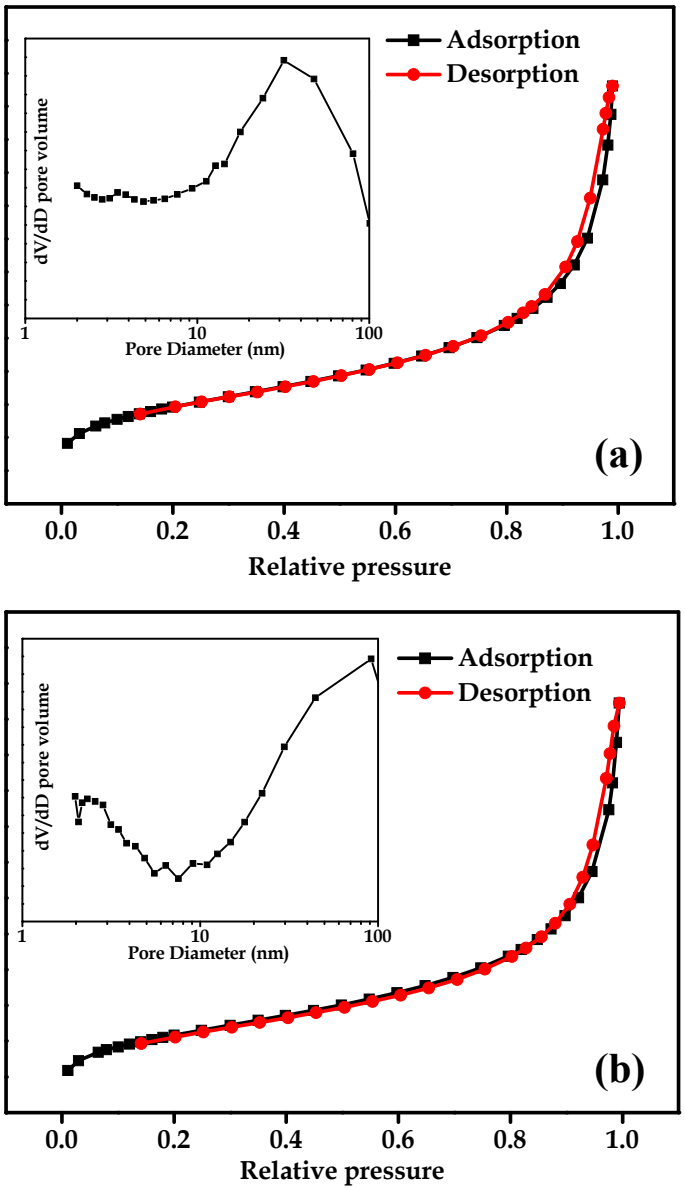

Figure SI9 Sorption isotherms and corresponding pore size distribution curves (insets in each picture) of the supercirtical CO_2 dried (a) and freeze dried (b) MWCNT/Fe_3O_4 composite aerogels

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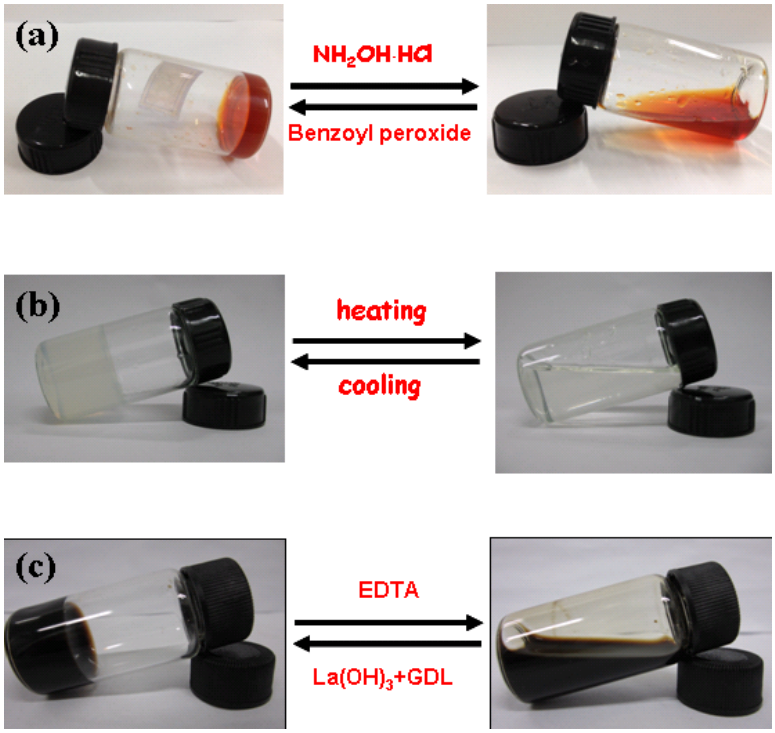

Figure SI10 Sol-gel transitions of the Fe-H_3BTC gel (a), agarose gel (b) and graphene oxide gel (c), respectively.