Electronic Supplementary Information

Carbon Nanotube-Bonded Graphene Hybrid Aerogels and an Application to Water Purification

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Figure S1. TEM images of interface between graphene and CVD-grown CNT. (b), (c) and (d) are high magnification images of the red rectangular part in Figure (a)



Figure S2. SEM images of 3D graphene hybrid structures at low-magnification. (a) G-MS, (b) G-MN, (c) G-CNT_A and (d) G-CNT_B.



Figure S3. Characterization of 3D graphene-CNTs hybrid structures. (a) XRD patterns,(b) Raman spectra,



Figure S4. SEM images on CNTs bridging between graphenes. (a) and (b) SEM images are the magnified images of Figure 2c



Figure S5. Nitrogen sorption isotherms of 3D graphene-CNTs hybrid structures



Figure S6. Pore size distribution of 3D graphene hybrid structures measured by $N_{\rm 2}$ sorption .



Figure S7. Viscoelastic properties of 3D graphene-CNTs hybrid structures. (a) and (b) storage modulus and loss modulus as a function of compressive strain variation at room temperature. (c) and (d) storage modulus and loss modulus as a function of temperature.



Figure S8. UV-vis spectra of the dyes solution according to absorption time. (a) methyl orange, (b) congo red, (c) crystal violet and (d) methylene blue



Figure S9. EDS spectra of 3D graphene hybrid structures. (a) G-MS, (b) G-MN, (c) G-CNT_A and (d) G-CNT_B.

	Element	Weight%	Atomic%
G-MS	C(K)	28.68	53.17
	O(K)	12.21	17.00
	Ni(K)	29.35	18.69
	CI(K)	29.75	11.14
G-MN	C(K)	65.47	87.30
	O(K)	4.50	4.50
	Ni(K)	30.04	8.20
	C(K)	86.30	96.15
G-CNT_A	O(K)	1.20	1.01
	Ni(K)	12.49	2.85
	C(K)	90.03	97.04
G-CNT_B	O(K)	1.29	1.05
	Ni(K)	8.68	1.92

Table S1. Element composition of 3D graphene hybrid structures by EDS



Wavelength (nm)

Figure S10. (a) Color of the original solution turned blue due to desorbed dye (marked original) changes to white gradually after each successive rinse. (b) UV-vis adsorption spectra of the original solution containing dye, the solution made clear with fresh G-CNT_A and the solution made clear with regenerated G-CNT_A. The inset shows optical images of the solution before and after adsorption (1st_abs.: a C-CNT_A is first used for the adsorption, 2nd_abs.: the structure is reused for the adsorption after regeneration in CTAB solution)



Figure S11. Solvent/oil absorption from water surface by 3D graphene-CNTs hybrid structures. (a) The sequential photographs of G-CNT_A absorbing toluene (dyed with oil blue) on water surface. Movement of the structure on the water surface is controlled by external magnet. (b) Absorption capacity (Q) of 3D graphene-CNTs hybrid structures for toluene. (c) Absorption capacity measured for a range of oils and organic solvents.



Figure S12. (a) Regeneration capacity of the G_CNT_A for the adsorbing solvent. (d) Photographs showing the regeneration process of the G_CNT_A.

	Properties									
Sample	Density (mg/cm³)	Electrical conductivity (S/cm)	Young's modulus (kPa)	Specific surface area (m²/g)	Magnetic property			Viscoelastic (kPa)		
					Satura- tion (emu/g)	rema- nent (emu/g)	Coer- cive(Oe)	Stora -ge	loss	
G-MS	37.5	0.001	4	184.1	1.5	-	-	1.35	0.205	
G-MN	20.3	0.0185	17	188.24	13.34	4.95	247	4.58	0.401	
G-CNT_A	60.2	0.179	169	289.0	11.77	5.5	265	131.0	10.73	
G-CNT_B	62.4	0.189	466	243.7	7.44	3.53	272	450	45	

Table S2. A summary of properties of 3D graphene hybrid structures.