## **Supporting Information for**

## Graphene Sponge for Efficient and Repeatable Adsorption and Desorption of Water Contaminations

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**Fig. S1** Digital photographs of rhodamine B solution after (a) just adding GO powders, and (b) adding GO powders for 5 min. The Zeta potential of GO is -29.5 mV, and the corresponding adsorption capacity of GO powders for rhodamine B is about 287.9 mg/g.

	GS1	GS2	GS3	GS4	GS5
Thiourea quantity (g)	0.1	0.3	0.5	0.5	0.5
Concentration of GO solution (mg/ml)	2	2	2	1	2
Size of GO	Small size (hundreds	Small size (hundreds	Small size (hundreds	Small size (hundreds	Large size
	nanometers)	nanometers)	nanometers)	nanometers)	(100 µ)
Specific surface area (m <sup>2</sup> /g)	120	149	150	399	79

Tab. S1 Different kinds of GSs used in the experiment.



Fig. S2 (a) SEM image and (b) the nitrogen adsorption-desorption isotherm of GSs prepared with a low concentration of GO solution (1 mg/ml, GO size: hundreds nanometers). The surface area obtained is  $399 \text{ m}^2/\text{g}$ .

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**Fig. S3** XPS spectra of GS1, GS2 and GS3. For the prepration of GS1, GS2 and GS3, 50 ml GO solution (GO size: hundreds of nanometers) with a concentration of 2 mg/ml was used, and the thiourea quantity was 0.1 g, 0.3 g and 0.5 g, repectively.



Fig. S4 (a) C1s, (b) O1s, (c) N1s, and (d) S2p XPS spectra of GS3.

Tab. S2 Element content of C, O, N, and S in GS1, GS2 and GS3.

	C (%)	O (%)	N (%)	S (%)
GS1	85.6	9.9	2.1	2.4

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GS2	83.2	8.8	3.9	4.1
GS3	82.6	7.3	5.2	4.9



Fig. S5 Raman spectra of GO, GS1, GS2, and GS3.



**Fig. S6** (a) Top view and (b) cross-section view of the GSs prepared without (left side) and with (right side) the use of thiourea. (c, d) SEM images of the GSs prepared (c) without and (d) with the use of

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thiourea.



Fig. S7 Molecular formula of (a) rhodamine B, (b) methylene blue, and (c) methyl orange.



**Fig. S8** The adsorption of GSs for different kinds of dyes. Left: rhodamine B ( $C_{28}H_{31}ClN_2O_3$ ; Relative molecular mass: 479); Middle: methylene blue ( $C_{16}H_{18}ClN_3S$ ; Relative molecular mass: 319.5); Right: methyl orange ( $C_{14}H_{14}N_3SO_3Na$ ; Relative molecular mass: 327).

Adsorbent	Adsorption amount (mg/g)	Equilibrium time (min)	Reference
GSs	72.5	180	Our work
Reduced GO	13.15	250	[1]
Magnetite/ Reduced GO	13.63	120	[2]
Multi-walled CNTs	36	180	[3]
Activated carbon fiber/CNT	2.5	120	[4]
Loguat seeds	3.97	300	[5]
Sago waste carbon	16.2	210	[6]
Surfactant-modified coconut coir pitch	13.3	90	[7]
hypercrosslinked			
polymeric	25-55	600	[8]
adsorbent (HJ-1)			
Activated carbon	20-400	30-300	[9-11]

Tab. S3 Rhodamine B adsorption performance comparison of GSs with other materials.

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Fig. S9 UV-Vis spectra of rhodamine B aqueous solution adsorbed by GSs for different time.



Fig. S10 Photograph of GSs prepared with 0.5 g (left one) and 0.7 g (right one) thiourea.



Fig. S11 SEM images and the nitrogen adsorption-desorption isotherm of GS1, GS2 and GS3.

Tab. S4 Structure and rhodamine B adsorption performance of GSs prepared with small and large GO

sheets.

CSs sample	Specific surface area	Zeta potential	Adsorption capacity
GSs sample	(m <sup>2</sup> /g)	(mV)	(mg/g)
Prepared with small GO sheets	150	1.4	72.5
(GS3)	150	-1.4	
Prepared with large GO sheets	70	1.2	40.2
(GS5)	19	-1.3	42.3

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Fig. S12 Cyclic performance of GSs for diesel oil and ethanol adsorption.

## Notes and references

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