

## Supporting Information

### **3D nitrogen doped graphene gels as robust and sustainable adsorbents for dyes**

Jiyu Geng,<sup>a</sup> Leilei Si,<sup>a</sup> Haotian Guo,<sup>a</sup> Chenhui Lin,<sup>a</sup> Ye Xi,<sup>a</sup> Yang Li,<sup>abc</sup> Xilong Yan,<sup>abc</sup> Bawei Wang,<sup>ab\*</sup> Ligong Chen<sup>abc\*</sup>

<sup>a</sup> School of Chemical Engineering and Technology, Tianjin University, Tianjin 300350, P. R. China.

<sup>b</sup> Collaborative Innovation Center of Chemical Science and Engineering (Tianjin), Tianjin 300072, P. R. China.

<sup>c</sup> Tianjin Engineering Research Center of Functional Fine Chemicals, Tianjin, P. R. China.

\* Corresponding author. E-mail: [bwwang@tju.edu.cn](mailto:bwwang@tju.edu.cn) (Bawei Wang); [lgchen@tju.edu.cn](mailto:lgchen@tju.edu.cn) (Ligong Chen)

Reduced graphene oxide (rGO) was prepared by the similar method with glycol and hexanediol in the place of hexamethylenediamine and 1, 12-diaminododecane, denoted as rGO-2 and rGO-6, respectively.

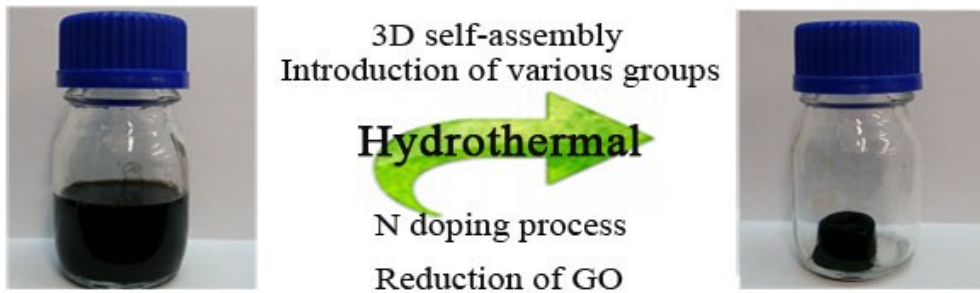


Fig. S1. Fabrication process pattern of NG gel.

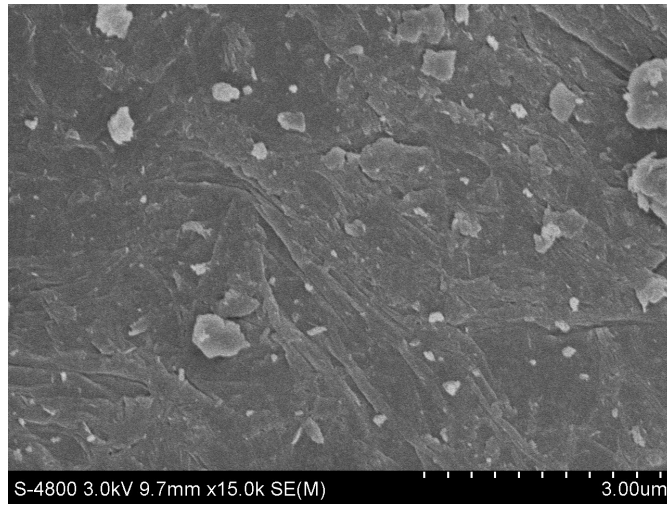


Fig. S2. SEM image of GO.

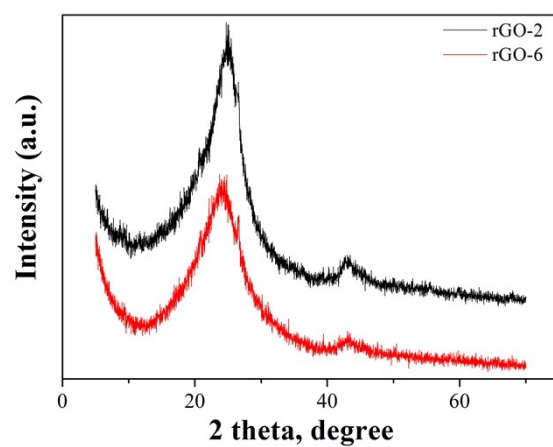


Fig. S3. XRD patterns of rGO-2 and rGO-6.

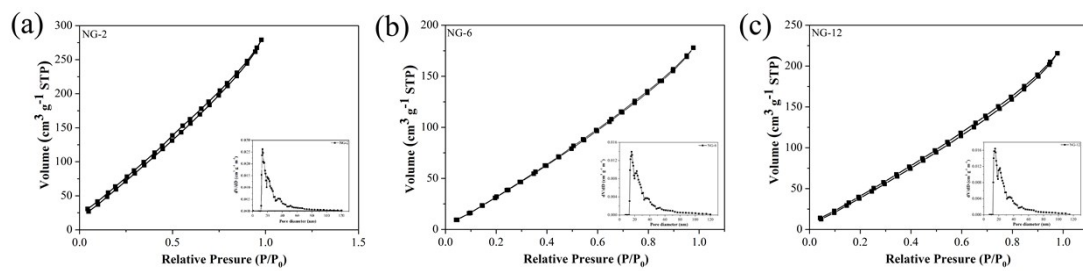


Fig. S4  $N_2$  adsorption-desorption isotherms and pore size distribution of NG samples: (a) NG-2; (b) NG-6; (c) NG-12.

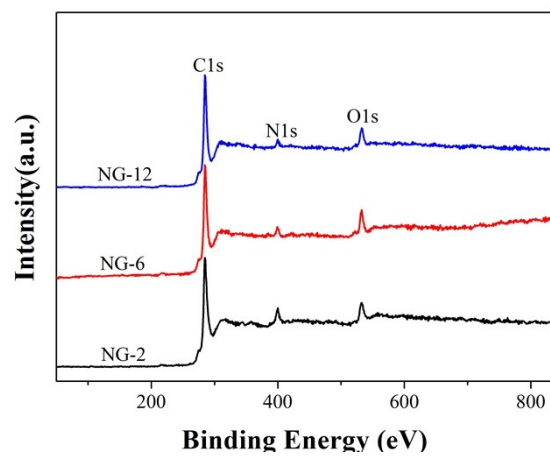


Fig. S5 XPS survey of NG samples

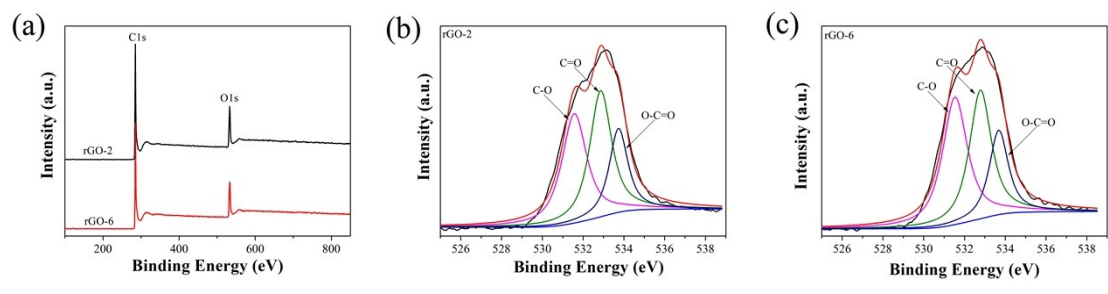


Fig. S6 High-resolution XPS spectra of (a) rGO samples and O 1s region for (b) rGO-2, (c) rGO-6.

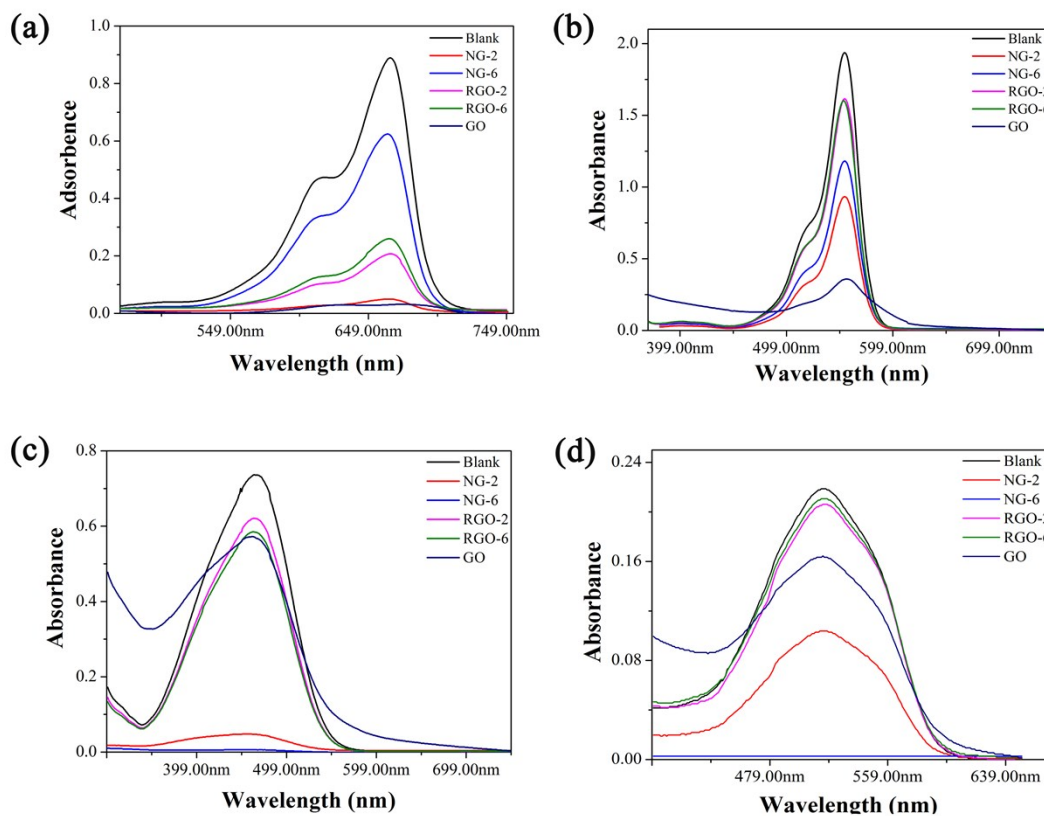


Fig. S7 UV-vis adsorption spectra of different dye solutions: (a) MB, (b) RhB, (c) MO and (d) AR after various adsorbents adsorption for 12 h.

Table S1 Comparison of adsorption values of GO, RGO and NG for adsorption of different dyes.

	MB(mg g <sup>-1</sup> )	RhB( mg g <sup>-1</sup> )	AR( mg g <sup>-1</sup> )	MO( mg g <sup>-1</sup> )
GO	124.49	98.58	24.90	19.17
RGO-2	79.19	27.43	5.77	12.81
RGO-6	73.02	28.98	3.72	16.80
NG-2	108.89	63.83	52.66	60.64
NG-6	52.92	36.84	174.86	161.31
NG-12	5.58	8.67	64.75	72.85



Table S2 Kinetics parameters of NG-2 and GO for the adsorption of MB and RhB.

Kinetic model	Parameters	MB		RhB	
		NG-2	GO	NG-2	GO
Pseudo-first-order	$q_e$ (mg g <sup>-1</sup> )	110.09	124.49	64.06	98.58
	$k_1$ (h <sup>-1</sup> )	0.12	0.10	0.14	0.13
	$R^2$	0.9999	0.9970	0.9993	0.9988
Pseudo-second-order	$q_e$ (mg g <sup>-1</sup> )	108.34	56.12	107.07	13.12
	$k_2$ (g (mg <sup>-1</sup> h <sup>-1</sup> )) × 10 <sup>-3</sup>	1.43	3.90	0.46	16.52
	$R^2$	0.9791	0.9641	0.9235	0.9662

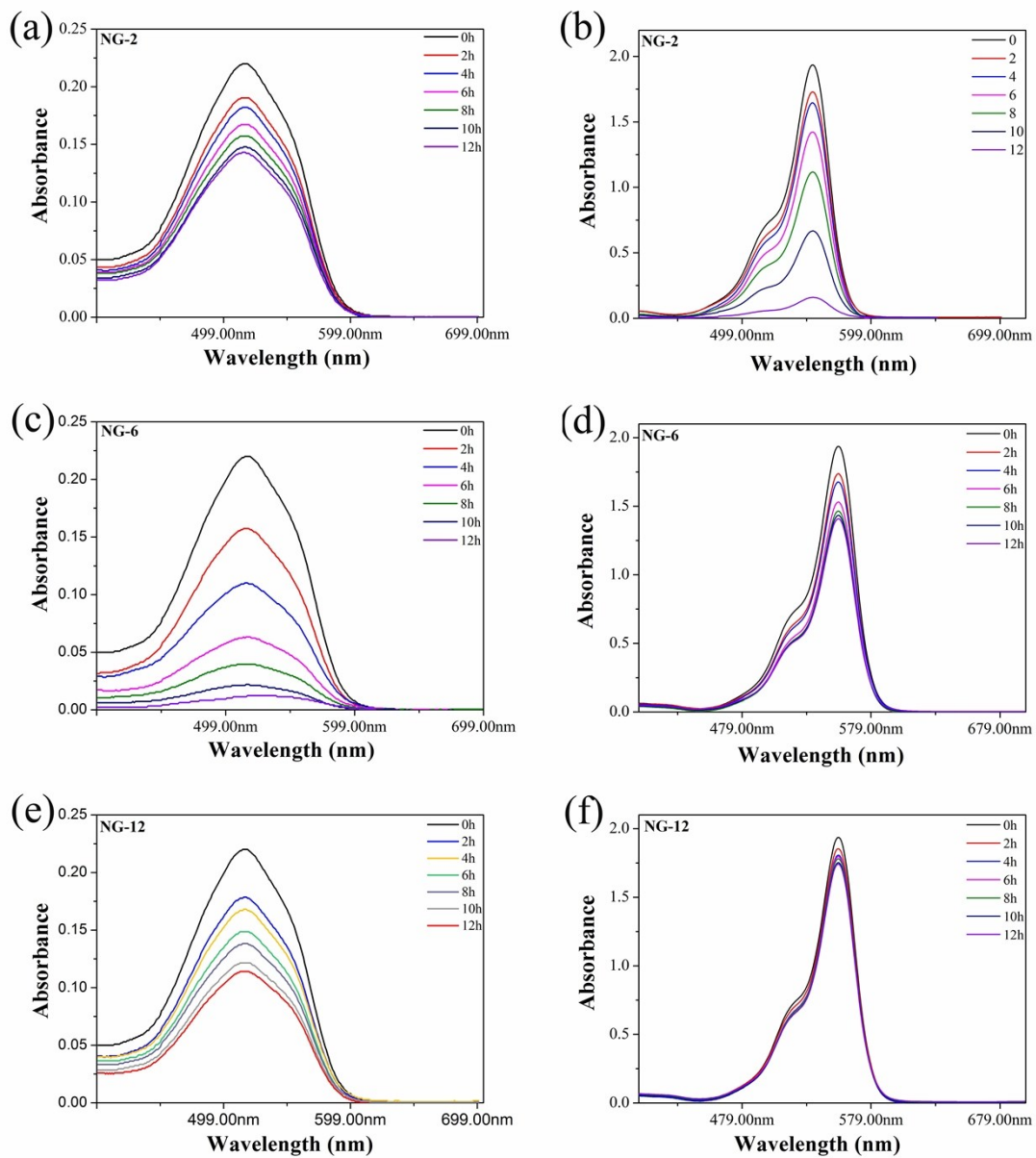


Fig. S8 UV-vis adsorption spectra of AR solutions after (a) NG-2, (c) NG-6 and (e) NG-12; RhB solutions after (b) NG-2, (d) NG-6 and (f) NG-12 adsorption for 12 h.

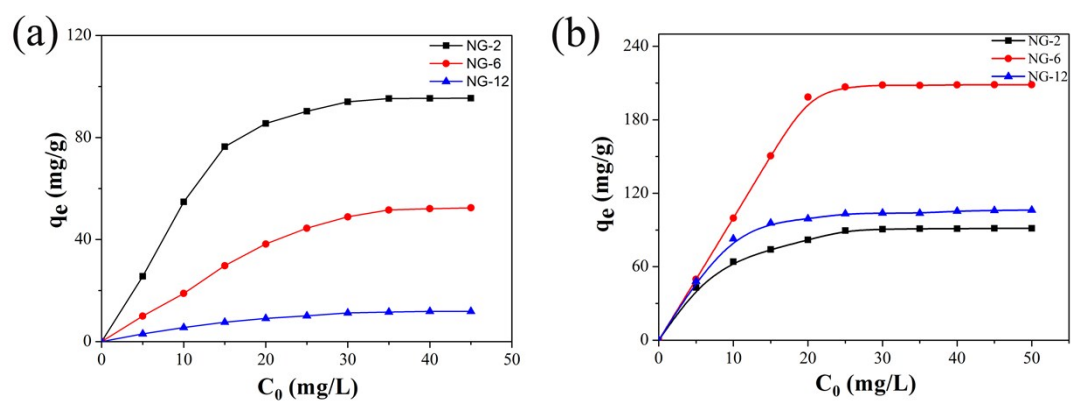


Fig. S9 Adsorption curves of (a) RhB and (b) AR on NG-2, NG-6 and NG-12.

Table S3 Langmuir and Freundlich isotherm parameters of NG-2, NG-6 and NG-12.

Isotherm model	Parameters	RhB			AR		
		NG-2	NG-6	NG-12	NG-2	NG-6	NG-12
Langmuir	$q_m$ ( $\text{mg g}^{-1}$ )	72.45	54.49	9.65	92.17	208.80	106.05
	$K_L$ ( $\text{L mg}^{-1}$ )	0.08	0.10	0.15	0.90	9.70	1.30
	$R_L$	0.3846	0.3333	0.2500	0.0528	0.0005	0.0372
	$R^2$	0.9985	0.9992	0.9987	0.9985	0.9940	0.9966
Freundlich	$K_F$ ( $\text{mg g}^{-1}$ )	1.52	2.87	15.82	31.67	262.42	40.64
	$(\text{L mg}^{-1})^{1/n}$						
	$n$	4.75	3.45	0.97	2.56	7.50	3.07
	$R^2$	0.9634	0.9816	0.9978	0.8892	0.8141	0.8164

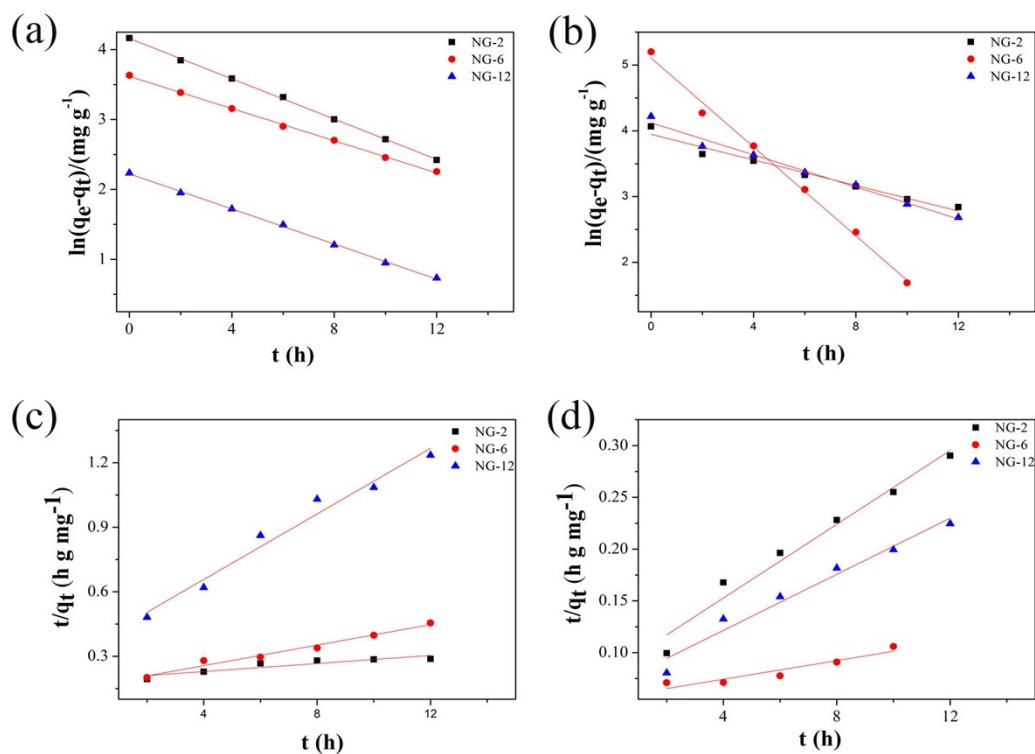


Fig. S10 Pseudo-first-order kinetic plots with (a) RhB, (b) AR on NG-2, NG-6 and NG-12; pseudo-second-order kinetic plots: (c) RhB, (d) AR on NG-2, NG-6 and NG-12.

Table S4 Kinetics parameters of NG-2, NG-6 and NG-12 for the adsorption of RhB and AR.

Kinetic model	Parameters	RhB			AR		
		NG-2	NG-6	NG-12	NG-2	NG-6	NG-12
Pseudo-first-order	$q_e$ ( $\text{mg g}^{-1}$ )	64.06	37.26	9.24	53.96	178.34	64.94
	$k_1$ ( $\text{h}^{-1}$ )	0.14	0.12	0.13	0.10	0.35	0.13
	$R^2$	0.9993	0.9989	0.9988	0.9970	0.9993	0.9982
Pseudo-second-order	$q_e$ ( $\text{mg g}^{-1}$ )	107.07	41.95	13.12	56.12	222.72	73.75
	$k_2$ ( $\text{g}(\text{mg}^{-1}\text{h}^{-1}))$ $\times 10^{-3}$	0.46	3.53	16.52	3.90	0.36	2.73
	$h_0$	5.27	6.21	2.84	12.28	17.86	14.85
	$R^2$	0.9235	0.9887	0.9662	0.9641	0.8545	0.9600

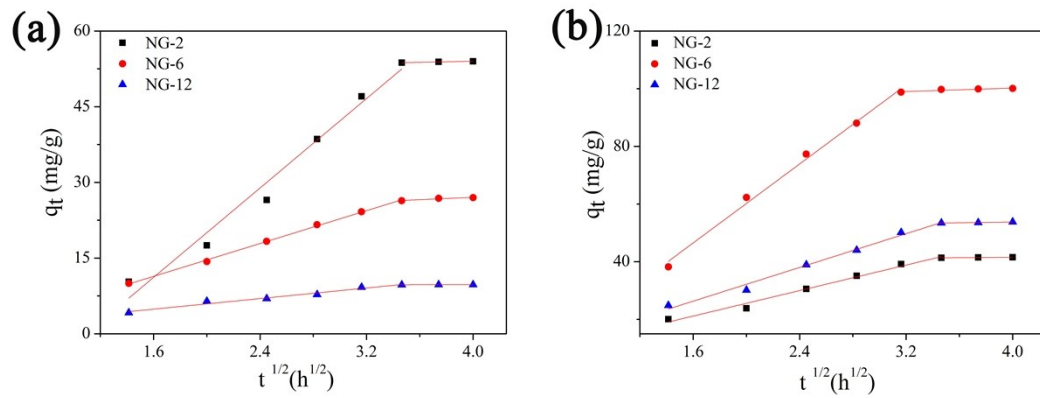


Fig. S11 Intra-particle diffusion kinetics plots with (a) RhB and (b) AR on NG-2, NG-6 and NG-12.

Table S5 Intra-particle diffusion model parameters for adsorption of RhB and AR .

Parameters	RhB			AR		
	NG-2	NG-6	NG-12	NG-2	NG-6	NG-12
$k_i$ (mg (g h <sup>1/2</sup> ) <sup>-1</sup> )	21.73	9.47	2.62	11.06	35.06	14.63
C (mg g <sup>-1</sup> )	23.52	1.66	0.69	3.44	15.53	2.90
R <sup>2</sup>	0.9734	0.9656	0.9677	0.9835	0.9515	0.9872