**Electronic Supplementary Information (ESI):** 

## Grafting Sulfonic and Amine Functional Groups on 3D Graphene for

## **Improved Capacitive Deionization**

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Fig. S1 CV curves of 3DGR, 3DNGR and 3DSGR at different scan rates.



Fig. S2 (a) SAC and CDI Ragone plots for the capacitor 3DGR-3DGR in a NaCl solution with different initial concentrations at a flow rate of 40 mL min<sup>-1</sup> at 1.4 V



Fig. S3 (a) Plots of NaCl solution conductivity *vs*. time and (b) the CDI Ragone plots for the capacitor 3DGR-3DGR with different voltage at a flow rate of 40 mL min<sup>-1</sup> in a 100 mg L<sup>-1</sup> NaCl solution.



Fig. S4 (a) Plots of NaCl solution conductivity *vs.* time and (b) the CDI Ragone plots for the capacitor 3DGR-3DGR with different flow rate in a 100 mg  $L^{-1}$  NaCl solution at 1.4 V.

Electrode material	Applied voltage (V)	Initial NaCl	Salt adsorption	
		concentration	capacity	Ref.
		(mg L <sup>-1</sup> )	(mg g <sup>-1</sup> )	
Graphene	1.2	50	1.85	1
Graphene/mesoporous carbon	2	40	0.73	2
Graphene nanosheets	2.0	250	8.6	3
GAC <sup>a</sup>	1.2	500	2.92	4
Carbon nanofibers	1.2	500	2.21	5
Hollow carbon	1.2	40	1.91	6
RGO-RF <sup>b</sup>	2.0	65	3.229	7
CNTs-RGO <sup>c</sup>	1.2	500	1.4	8
Graphene/carbon Nanotube	2	30	1.41	9
3DMGA <sup>d</sup>	1.6	50	3.9	10
3DNGR-3DSGR	1.2	100	6.4	This work
3DNGR-3DSGR	1.4	100	9.72	This work
3DNGR-3DSGR	1.4	500	13.72	This work

Table S1 Comparison of the salt adsorption capacity among various carbon electrode materials from the literatures

a: reduced graphene oxide (RGO) and activated carbon (AC) composites; b: reduced graphite oxidate-resol like material; c: carbon nanotubes (CNTs) and reduced graphene oxide (RGO); d: three-dimensional macroporous graphene architectures.

Electrode material	Applied voltage (V)	Initial NaCl concentration (mg L <sup>-1</sup> )	Charge efficiency	Ref.
Activated carbon	1.2	500	0.19	4
GAC <sup>a</sup>	1.2	500	0.24	4
CNTs	1.6	500	0.32	8
CNTs-RGO <sup>b</sup>	1.6	500	0.40	0
CFC <sup>c</sup>	1.4	250	0.41	11
CFC-SRGO <sup>d</sup>	1.4	250	0.46	
Carbon nanorods	1.2	500	0.56	12
Carbon	1.2	500	0.72	13
nanofibers				
3DNGR-3DSGR	1.4	500	0.85	This work

Table S2 Comparison of the charge efficiency among various carbon electrode materials from the literatures

a: reduced graphene oxide (RGO) and activated carbon (AC) composites; b: carbon nanotubes (CNTs) and reduced graphene oxide (RGO); c: carbon fibre cloth (CFC); d: carbon fibre cloth (CFC) and sulphonated reduced graphene oxide (SRGO) composites.

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