Electronic Supplementary Information

Nitrogen-doped graphene composites as efficient electrodes with enhanced capacitive deionization performance

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Preparation of melamine-formaldehyde (MF) nanoparticles [1]: 2.8 g of melamine and 6 mL of 37.0 wt % formaldehyde aqueous solution were mixed in a 50 mL two-necked round bottom flask connected to a reflux condenser and equipped with stirring rod. Then, the pH value of the solution was adjusted to 9.0-9.5 by 2.5 mL of triethanolamine (TEA) under mechanical stirring. The mixture was heated to 55-60 ^{o}C. After 30 min, the MF pre-polymer solution was obtained and transferred to 100 mL of PVA solution (0.5%). After 5 min, turbidity of the liquid was occurred, which indicated the progressing formation and growth of the MF particles. The reaction was quenched with ice water. The dispersion of MF particles was centrifuged (10 min, 10000 rpm) and the supernatant was discarded. The obtained MF particles were repeated washing three times with deionized water, prior to freeze drying in vacuum for 24 h.

Table S1. The specific surface area, average pore diameter and total pore volume of RGO and RGO-MF composites.

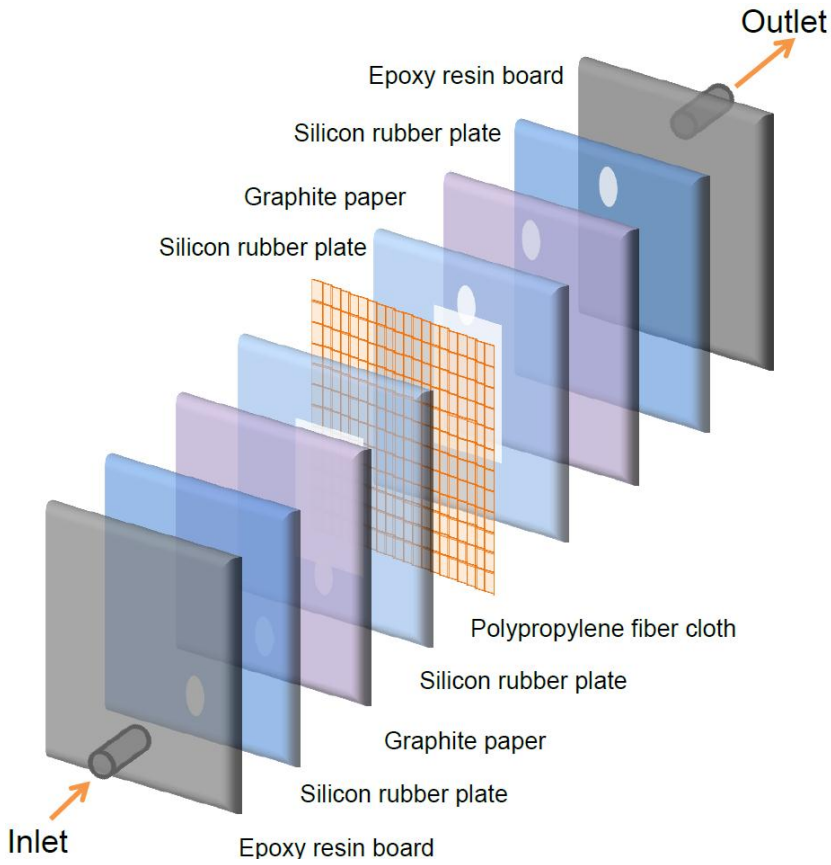
Samples	Specific surface area (m^2/g)	Average pore diameter (nm)	Total pore volume (cm^3/g)
RGO-MF-10	192	6.2	0.57
RGO-MF-20	352	7.2	1.1
RGO-MF-50	297	7.3	0.95
RGO	120	8.8	0.3

Table S2. Atomic percentage of carbon, nitrogen and oxygen in GO, RGO, MF, and RGO-MF composites.

Atomic percent%	GO	RGO	RGO-MF-10	RGO-MF-20	RGO-MF-50	MF
C	65.32	96.59	84.82	84.58	82.39	74.63
N	0	0	3.91	10.86	15.28	16.31
O	34.68	3.41	11.27	4.55	2.33	9.07

Table S3. Ratios of pyridinic and quaternary nitrogen of MF and RGO-MF composites, evaluated from the peak separation analyses of N1s core level peaks.

Samples	398.2	399.5	401.3
MF	0.56	0.32	0.12
RGO-MF-10	0.47	0.24	0.29
RGO-MF-20	0.47	0.23	0.30
RGO-MF-50	0.42	0.30	0.28



Scheme S1. Schematic illustration of CDI device.

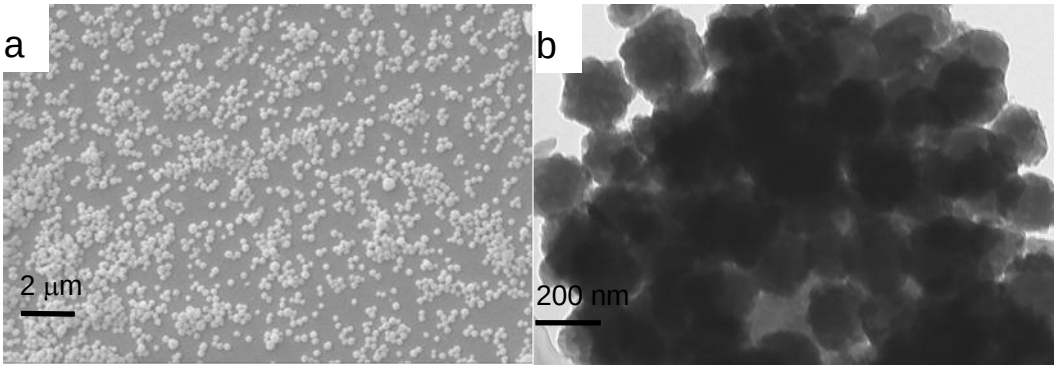


Fig. S1 (a) SEM and (b) TEM images of MF nanoparticles.

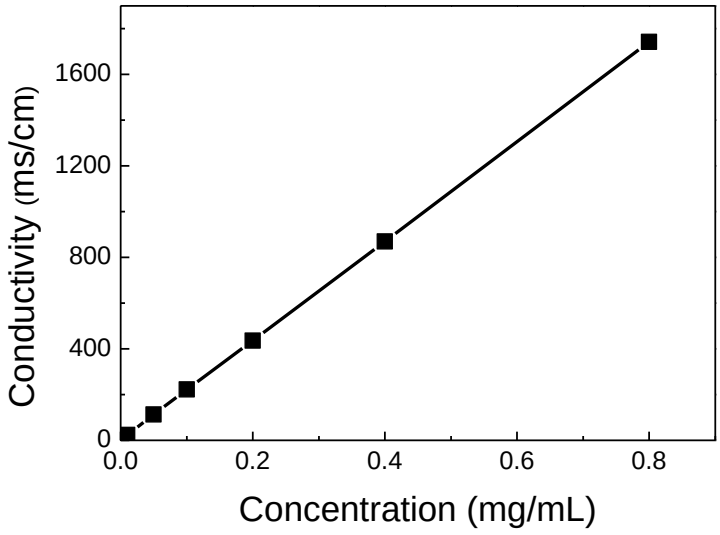


Fig. S2 The relationship between conductivity and concentration of NaCl solutions.

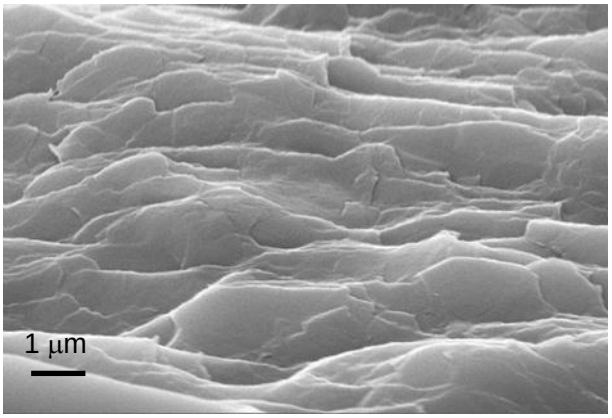


Fig. S3 SEM image of GO sheets.

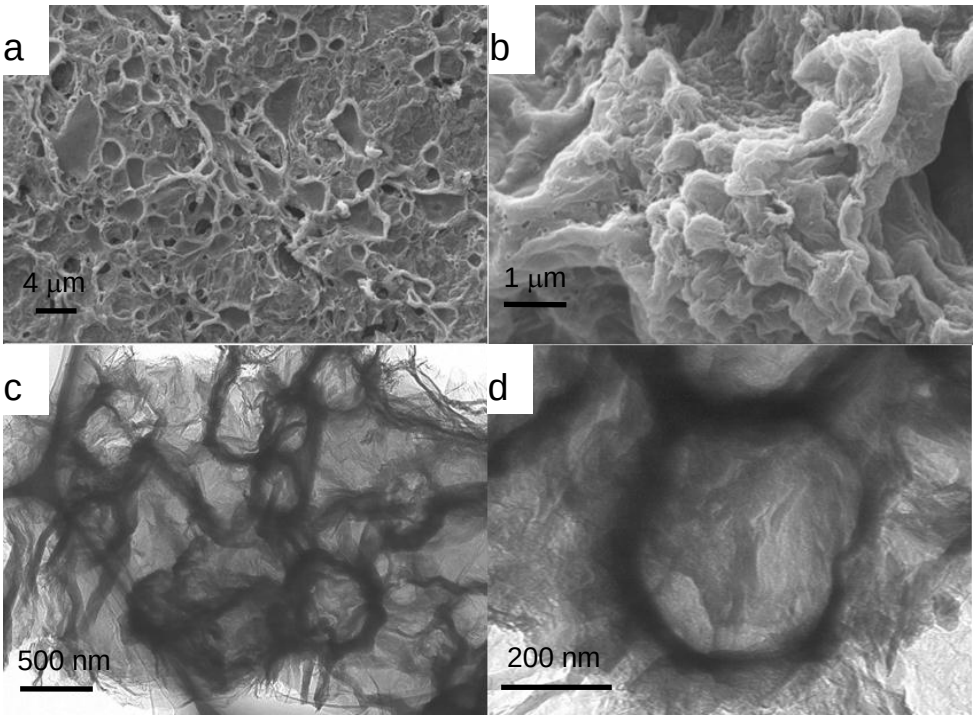


Fig. S4 (a,b) SEM and (c,d) TEM images of 3D RGO-MF-20 composite.

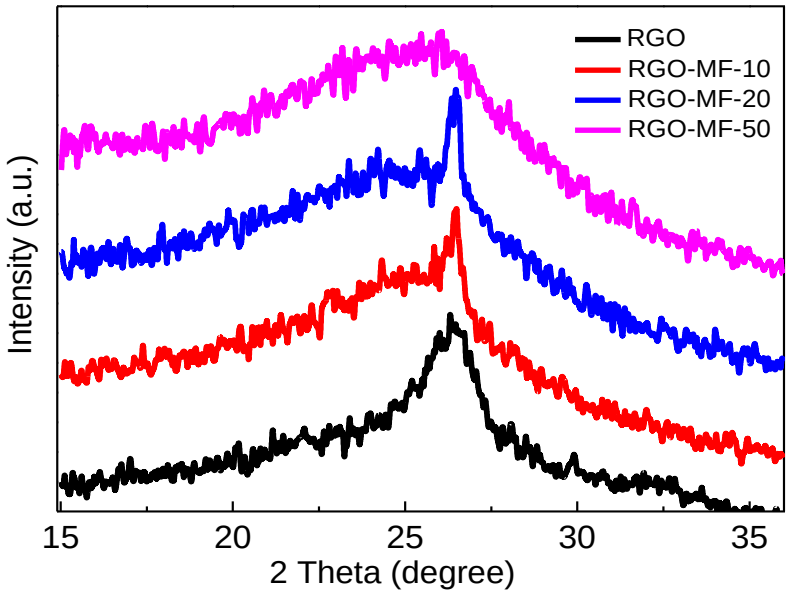


Fig. S5 XRD patterns of RGO and RGO-MF composites.

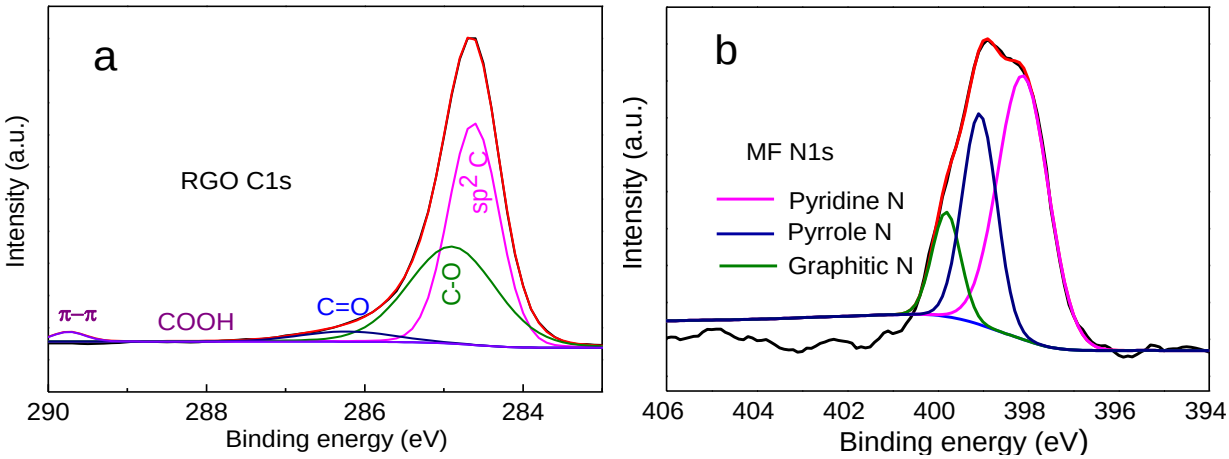


Fig. S6 (a) XPS C1s core level spectra of RGO and (b) XPS N1s core level spectra of MF.

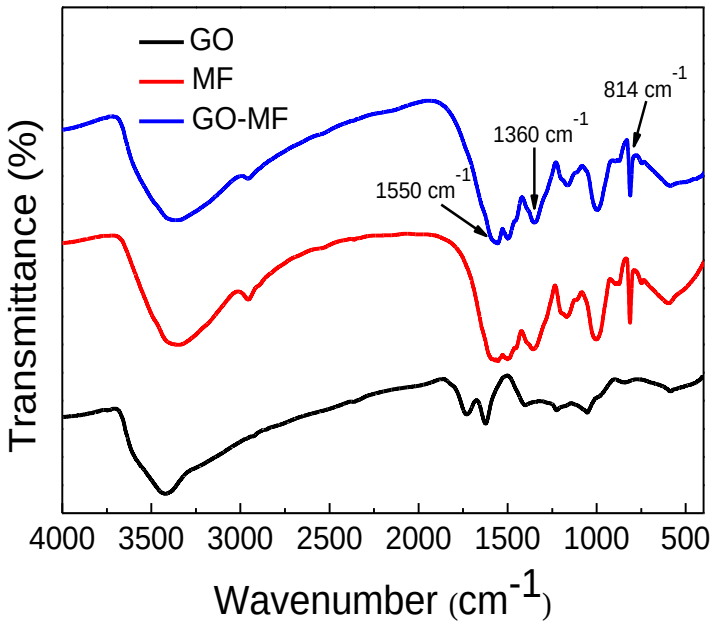


Fig. S7 FT-IR spectrum of GO, MF and RGO-MF-20 composite.

The peak at 1760 cm^{-1} for the C=O group disappears in RGO-MF-20 composite. The peaks of 1550, 1360, 814 cm^{-1} appear after the MF incorporation and calcination.

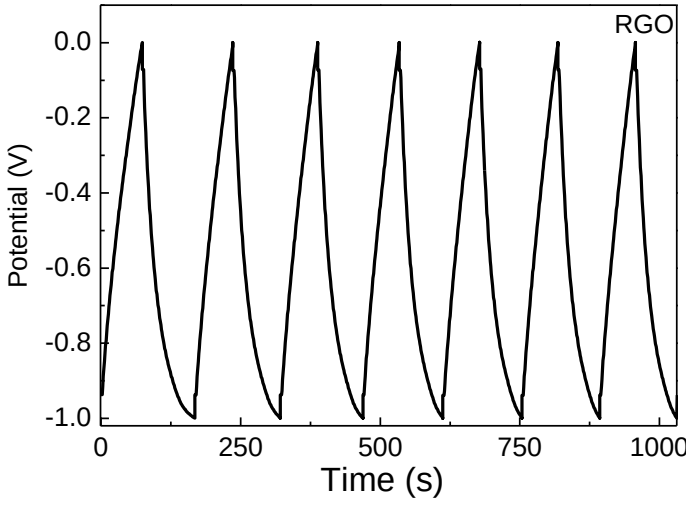


Fig. S8 Continuous GC curves of the RGO electrode with a current density of 0.3 A/g.

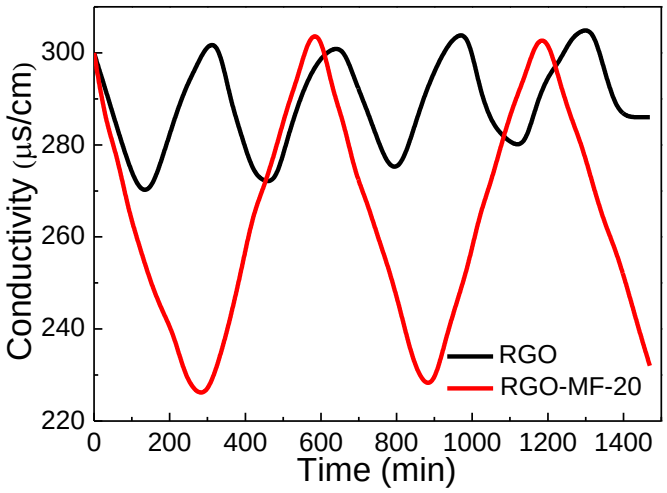


Fig. S9 The regeneration cycles of the RGO and RGO-MF-20 electrodes with the absence of an external potential.

Reference

(1) Ye, S. Q.; Wang, C. Y.; Liu, X. X.; Tong, Z.; Ren, B. Y.; Zeng, F. New Loading Process and Release Properties of Insulin from Polysaccharide Microcapsules Fabricated through Layer-by-Layer Assembly. *J. Control. Release* **2006**, *112*, 79-87.