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Supplementary Information for

Binder-free δ-MnO_2@reduced graphene oxide composite film as a bi-functional electrode for aqueous rechargeable sodium-ion battery and hybrid capacitive deionization

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Calculations of deionization performance

$$SAC = \frac{(C_0 - C)V}{m} \qquad (1)$$

$$ASAR = \frac{SAC}{t} \qquad (2)$$

where V (mL) is the volume and m (mg) represent the total electrode mass. Meanwhile, C_0 and C (mg g^{-1}) refer to the initial and equilibrium concentrations of the NaCl solution, respectively. In addition, t (min) is the charging time for the desalination process.

$$\Lambda = \frac{SAC \times F}{1000 \times 58.44 \times \Sigma} \qquad (3)$$

where F is Faraday constant (96485 C mol^{-1}), 58.44 is the molar mass of NaCl and Σ is integral charge during the desalination process (charge, C g^{-1}).

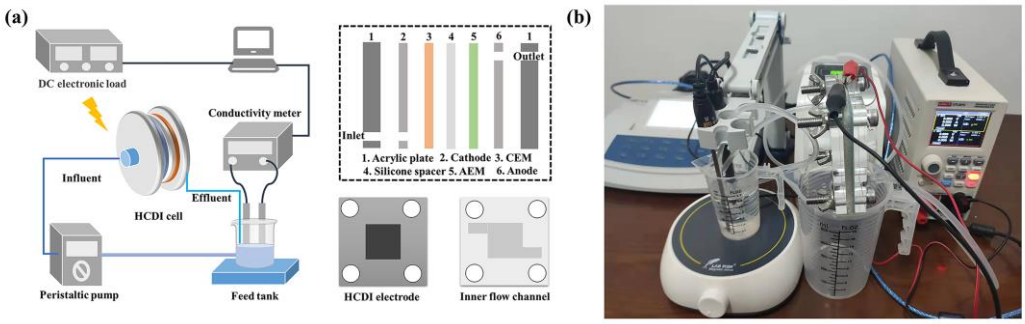

Fig. S1 (a) Schematic illustration of batch mode HCDI unit and internal structure and (b) Photographical image of HCDI experimental set up.

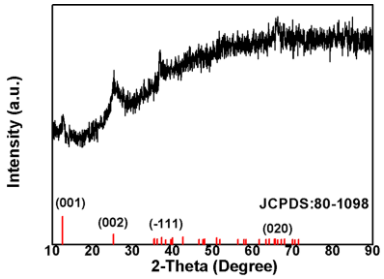

Fig. S2 XRD patterns of δ-MnO_2@rGO/CC sample.

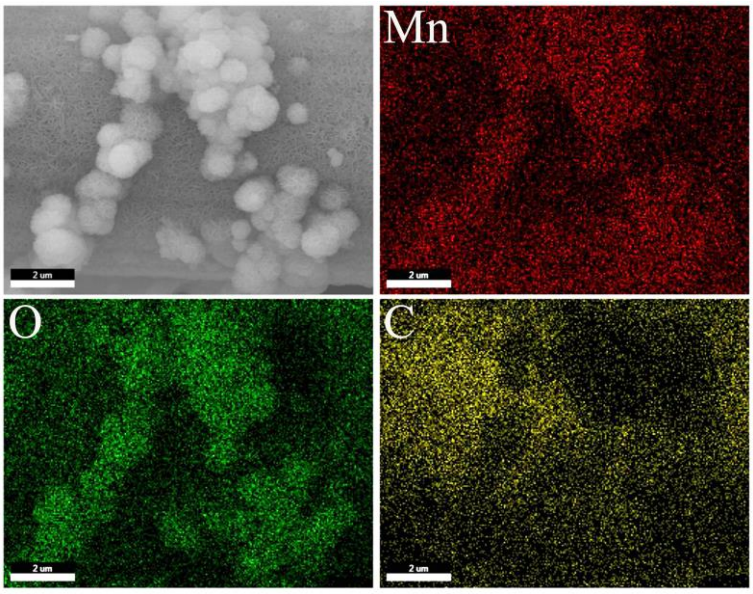

Fig. S3 EDS elemental mapping of δ-MnO_2@rGO/CC sample.

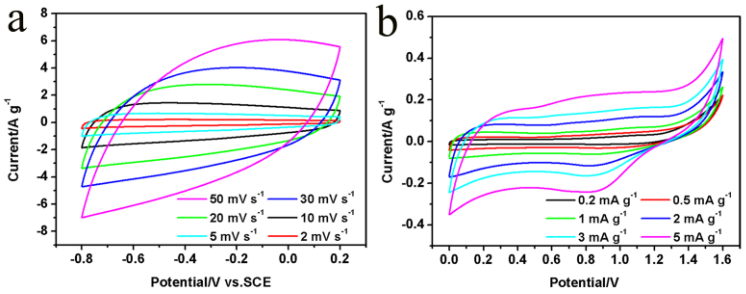

Fig. S4 CVs of AC (a) and δ-MnO_2@rGO/CC//AC in full cell system (b).

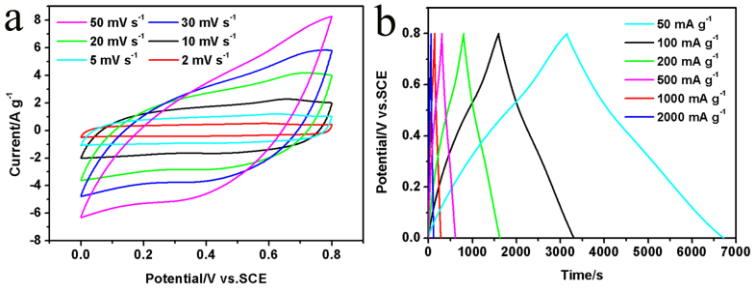

Fig. S5 (a) CVs of δ-MnO_2@rGO/CC electrodes at 2-50 mV s^{-1} in 1 M NaCl solution, (b) GCD curves of δ-MnO_2@rGO/CC electrodes at 50-2000 mA g^{-1} in 1 M NaCl solution.

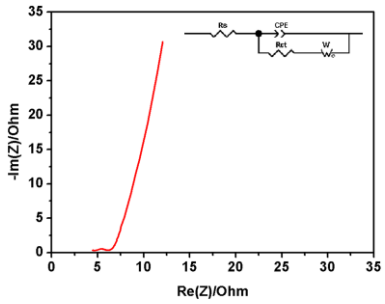

Fig. S6 Electrochemical impedance spectra of the as prepared δ-MnO_2@rGO/CC electrode in 1M NaCl solution.

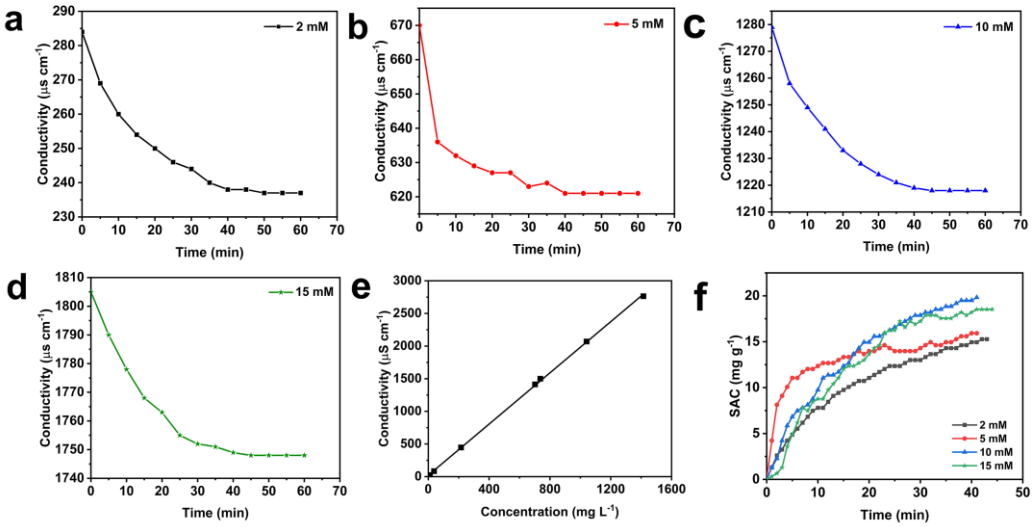

Fig. S7 (a-d) Electro-adsorption equilibrium curves of different feed concentrations, (e) Correlative curves of NaCl concentration (mg L^{-1}) corresponding to conductivity (μS cm^{-1}), (f) The curves of desalination amount vs. charging time of the δ-MnO_2@rGO/CC electrode.

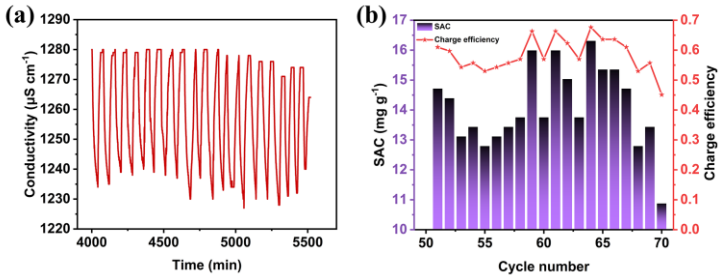

Fig. S8 (a) Cycling performance of 50-70 cycles, (b) Corresponding deionization capacity and charge efficiency response.

Table S1. Obtained impedance parameters and D_{Na} values of the δ-MnO_2@rGO/CC by EIS.

Electrode	R_s (Ω)	CPE-T	CPE-P	R_{ct} (Ω)	σ	D_{Na} ($cm^2\ s^{-1}$)
δ-MnO_2@rGO/CC	4.89	8.77×10^{-4}	0.99	0.95	2.30	2.52×10^{-4}

Table S2. Comparison of deionization performance of various MnO_2 electrodes reported in the literature.

Electrode	Applied voltage (V)	SAC ($mg\ g^{-1}$)	Cycle number	Ref.
δ-MnO_2@rGO/CC	1.2	19.8	50	this work
MnO_2/NPC	1.2	0.99	10	[1]
MnO_2/PSS/CNT	1.2	4.7	10	[2]
GNS@MnO_2	1.2	5.01	50	[3]
ep-AC@MnO_2	1.2	25.7	30	[4]
GO/PPy/MnO_2	1.2	38.4	30	[5]
Mn-Fe-PBAs/CC	1.0	14.47	30	[6]
MnO_2/AC	1.0	9.3	30	[7]
MnO_2/MWCNTs	1.2	6.65	-	[8]

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