

Electronic Supplementary Information

Free-standing flexible film as a binder-free electrode for efficient hybrid deionization system†

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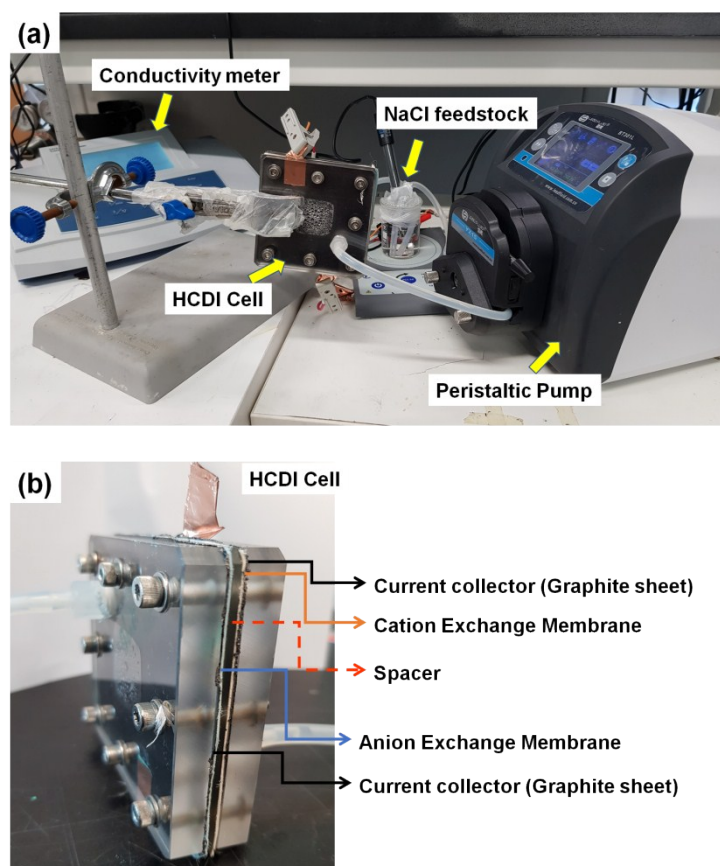


Fig. S1 Photographical image of hybrid capacitive deionization (HCDI) experimental set up (a) and the HCDI cell unit (b).

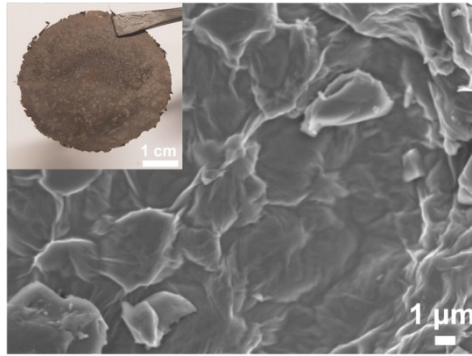


Fig. S2 SEM image of AC@rGO film.

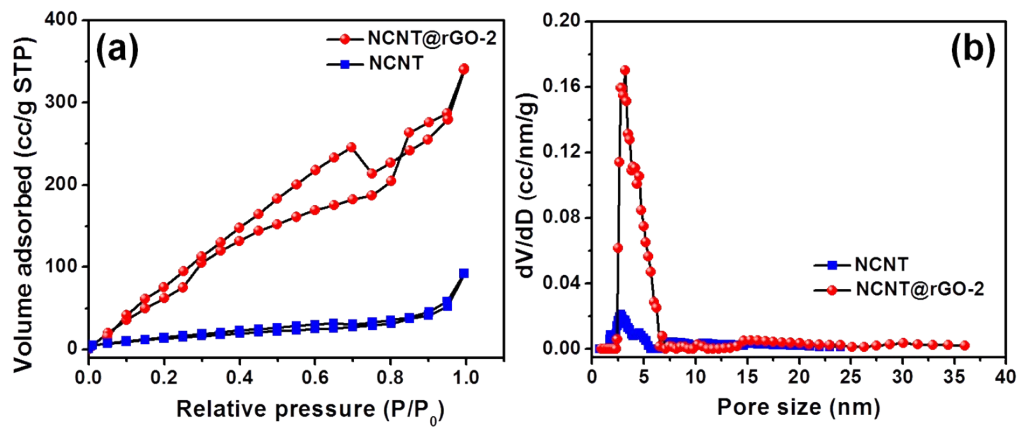


Fig .S3 a) Nitrogen adsorption-desorption isotherms, (b) pore size distribution of NCNT composite and NCNT@rGO-2 film.

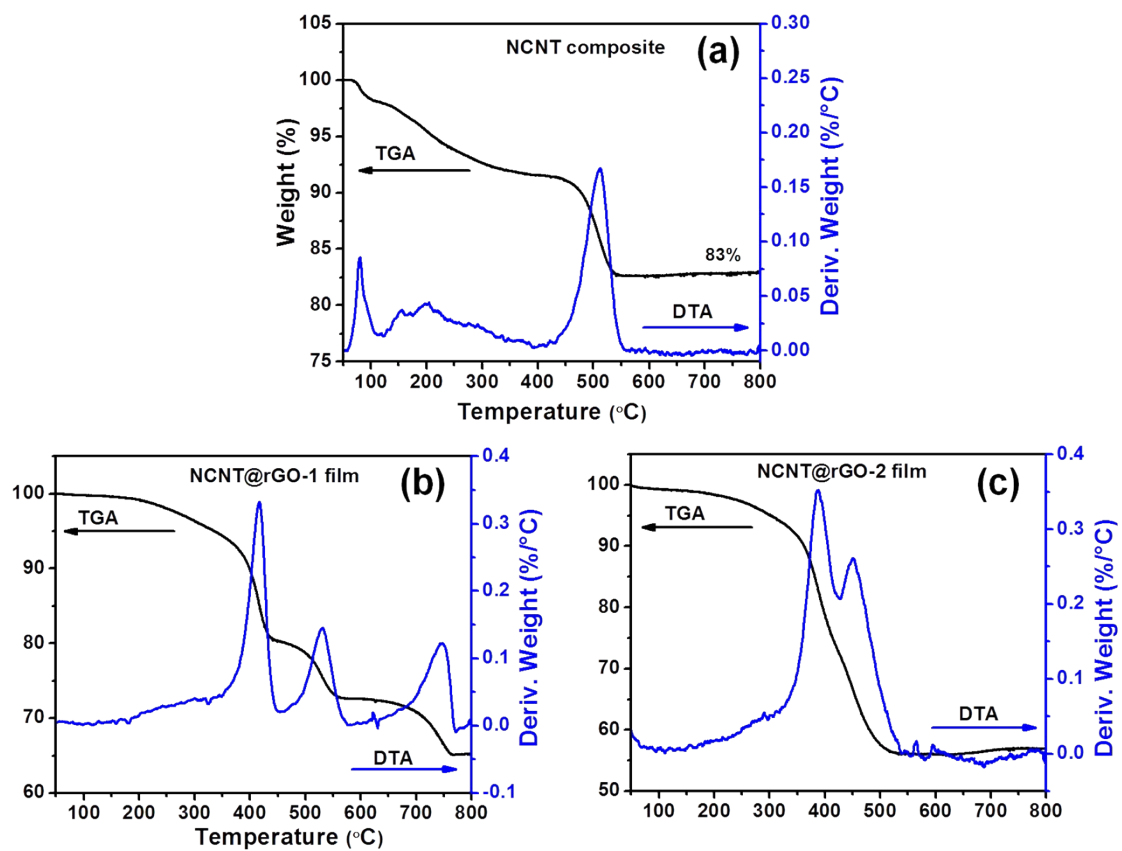


Fig. S4 DTA curves for (a) NCNT composite; (b) NCNT@rGO-1 film; (c) NCNT@rGO-2 film.

Table S1 FTIR analysis results and comparisons of GO, NCNT composite and NCNT@rGO film

| Electrode material | Band | Characteristic peak |
|--------------------|---|--|
| 1) GO | ~3000 - 3400 cm ⁻¹ 2922, 2850, 1717, 1633, 1398 and 1099 cm ⁻¹ | stretching vibrations of surface hydroxyl groups of -COOH and H ₂ O asymmetric C-H, symmetric C-H, carbonyl (C=O), aromatic (C=C), carboxyl (O=C-O) and alkoxy (C-O) stretching vibrations |
| 2) NCNT composite | 477 and 718 cm ⁻¹ 907 cm ⁻¹ 1637 and 3214 cm ⁻¹ 3400 cm ⁻¹ | Ti-O vibrations stretching peak of Ti-O-Na stretching and bending modes of hydroxyl (-OH) and water molecules coordinated to Ti ⁴⁺ ions stretching vibration of hydroxyl groups from O=C-OH and C-OH present in MWCNTs |
| 3) NCNT@rGO film | 477 and 718 cm ⁻¹ 907 cm ⁻¹ 3400 cm ⁻¹ 1570 cm ⁻¹ 1160 cm ⁻¹ | Ti-O vibration stretching peak of Ti-O-Na stretching vibration of hydroxyl groups from O=C-OH and C-OH present in MWCNT C=C stretching of rGO C-OH stretching of rGO |

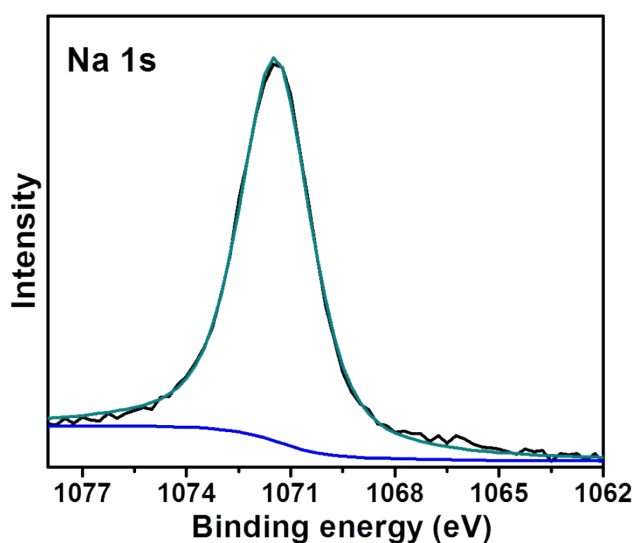


Fig. S5 High resolution spectrum of Na 1s corresponding to NCNT@rGO film.

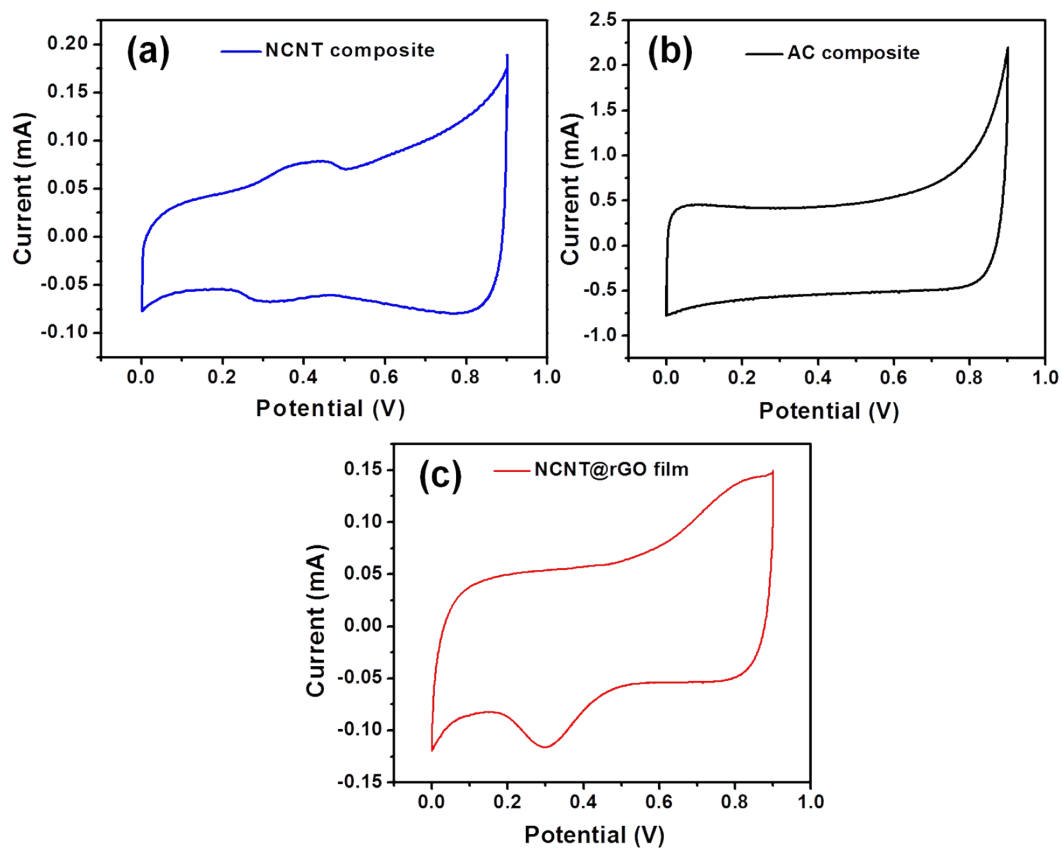


Fig. S6 Cyclic voltammograms curves in a voltage range of 0.0-0.9 V for (a) NCNT composite; (b) Activated carbon composite; and (c) NCNT@rGO film electrode at a scan rate of 1 mV/s in 3M NaCl solution.

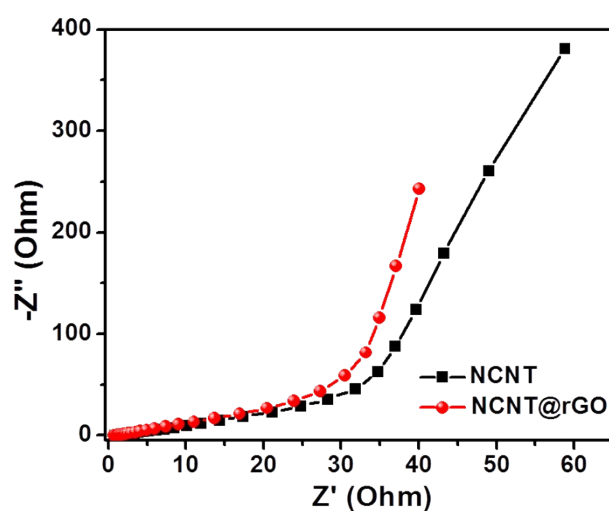


Fig. S7 Electrochemical impedance spectra of the as prepared electrodes NCNT composite and NCNT@rGO in 3M NaCl solution.

Table S2 Summary of the desalination performance, charge efficiency of electrodes in the HCDI system operated under an applied current of 1 mA

| HCDI system | Total mass of electrode (mg) | ($C_e - C_o$)* mg/L | Salt removal capacity* (mg/g) | Time consumed for desalination at applied current of 1 mA (min)* | Desalination rate performance* (mg/g/s) | Current density (mA/g) | Charge efficiency, Λ (%) |
|----------------------|---|-----------------------|-------------------------------|--|---|------------------------|----------------------------------|
| NCNT//AC composite | 17.3 (includes mass of active materials and the binders) | ~38.3 | ~109 | ~68 | ~0.026 | 116 | ~77 |
| NCNT@rGO-1//AC@rGO-1 | 14.9 | ~30.66 | ~103 | ~45 | ~0.038 | 147 | ~93.6 |
| NCNT@rGO-2//AC@rGO-2 | 14.7 | ~38.16 | ~129 | ~55 | ~0.039 | 145 | ~95.3 |

*where C_o (mg/L) is the initial NaCl concentration, C_e (mg/L) is the final equilibrated NaCl concentration. An average of 6 cycles are used for calculation.

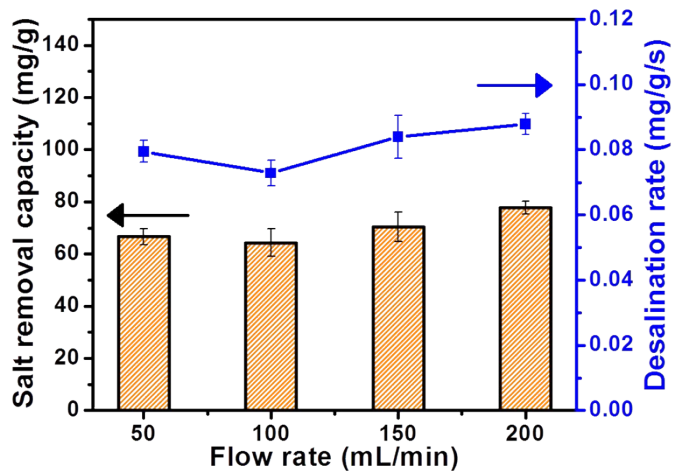


Fig. S8 Salt removal capacity and desalination rate of NCNT@rGO-1 electrode at different flow rates in ~3000 mg/L NaCl solution at an applied current of 2mA.

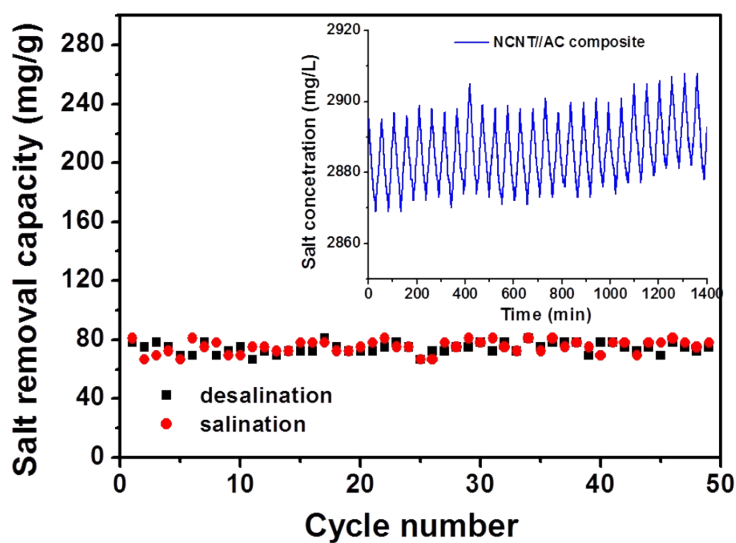


Fig. S9 (a) The cycling performance of the NCNT//AC system at an applied current of 1.5 mA; inset is the corresponding change in the conductivity pattern.