

Electronic Supplementary Information

Enhancement of the Donnan Effect through Capacitive Ion
Increase using an Electroconductive rGO-CNT Nanofiltration
Membrane

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Cross-flow filtration setup and salt rejection

Figure S1-S4

Cross-flow filtration setup and salt rejection

The cross-flow filtration setup consists of nitrogen cylinder, feed water tank, cross-flow filtration cell, DC power supply, peristaltic pump, concentrated water tank, and permeate water tank. A dispensing pressure vessel (5 L, Millipore, USA) was used as the feed water tank, and no pH adjust in the prepared salt aqueous solution. DI water was used to measure the pure water flux. A DC power (DH1765-1, Beijing Dahua, China) was used to supply the cell voltage between the GCMs and the counter electrode. A polycarbonate, custom-made, cross-flow filtration cell was used to evaluate the salt rejection of the GCMs. The filtration cell was designed with a steel ring capable of delivering an electrical current to the GCM (effective membrane surface area was 9.08 cm²) and a counter Ti electrode (effective electrode area was 7.06 cm²) located at the top of the cell, 10 mm above the GCM surface. Five flange bolts were screwed through the upper part and the under part of the filtration cell to fix the membrane and seal the cell. The feed water was circulated over the membrane under 1 bar pressure, and separated into permeate and retentate, which flowed into the concentrated water tank and permeate water tank, respectively. In each filtration experiment, a new GCM was installed into the filtration cell and stabilized after 1 h of compaction and pre-adsorption. The permeate for 12 h was collected to measure electrical conductivity (S230 Seven Compact, Mettler-Toledo, Switzerland) and flux mass (ME2002E, Mettler-Toledo, Switzerland). The retentate water was pumped into a concentrated water tank by a peristaltic pump (0.5 ml/min). The calculation methods of rejection ratio and water flux referenced previous researches.^{1,2}

By recording the mass of permeated water using a digital balance (ME2002E, Mettler Toledo, Switzerland) in certain period time, the permeate fluxes (J , L·m⁻²·h⁻¹·bar⁻¹) were calculated using the following equation:

$$J = \frac{V}{A \times t \times P} \quad (1)$$

Where V is the volume of permeated water, A is the effective area of membrane, t is

the permeation time of each tests, P is the transmembrane pressure.

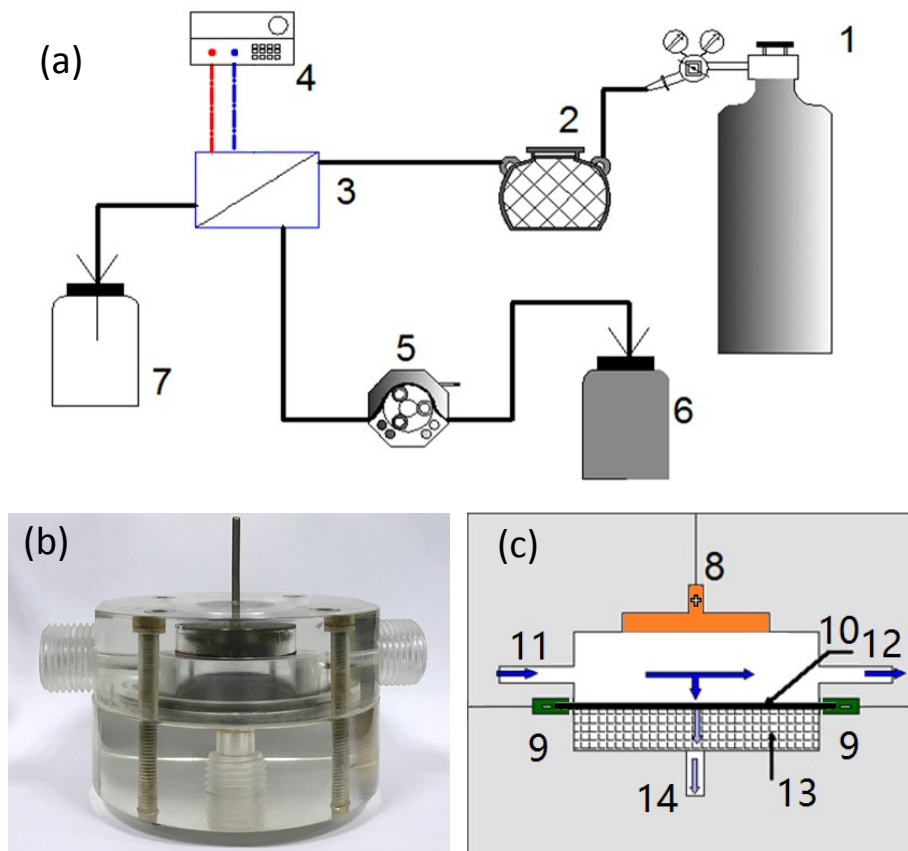


Fig. S1 (a) Filtration setup, (b) Photograph of the cross-flow filtration cell, and (c) Schematic illustration of the cross-flow filtration cell. (1) nitrogen cylinder; (2) feed water tank; (3) cross-flow filtration cell; (4) DC power supply; (5) peristaltic pump; (6) concentrated water tank; (7) permeate water tank; (8) Ti electrode; (9) steel ring; (10) GCM; (11) inlet; (12) outlet; (13) porous support; (14) permeate.

Surface morphology of GCMs with different CNT loading

The microstructure of the GCMs with various CNT content is shown in Fig. S2. The surface morphology of the rGO membrane was smoother than that of the GCM, and CNTs were more densely distributed on the GCMs as increasing CNT content. A wrinkled surface topography and a lamellar structure were observed on the image of the rGO membrane. The rGO-CNT composite loading on the PVDF substrate

membrane was with the thickness of about 185 nm for the GCM-33 (Fig. S2i).

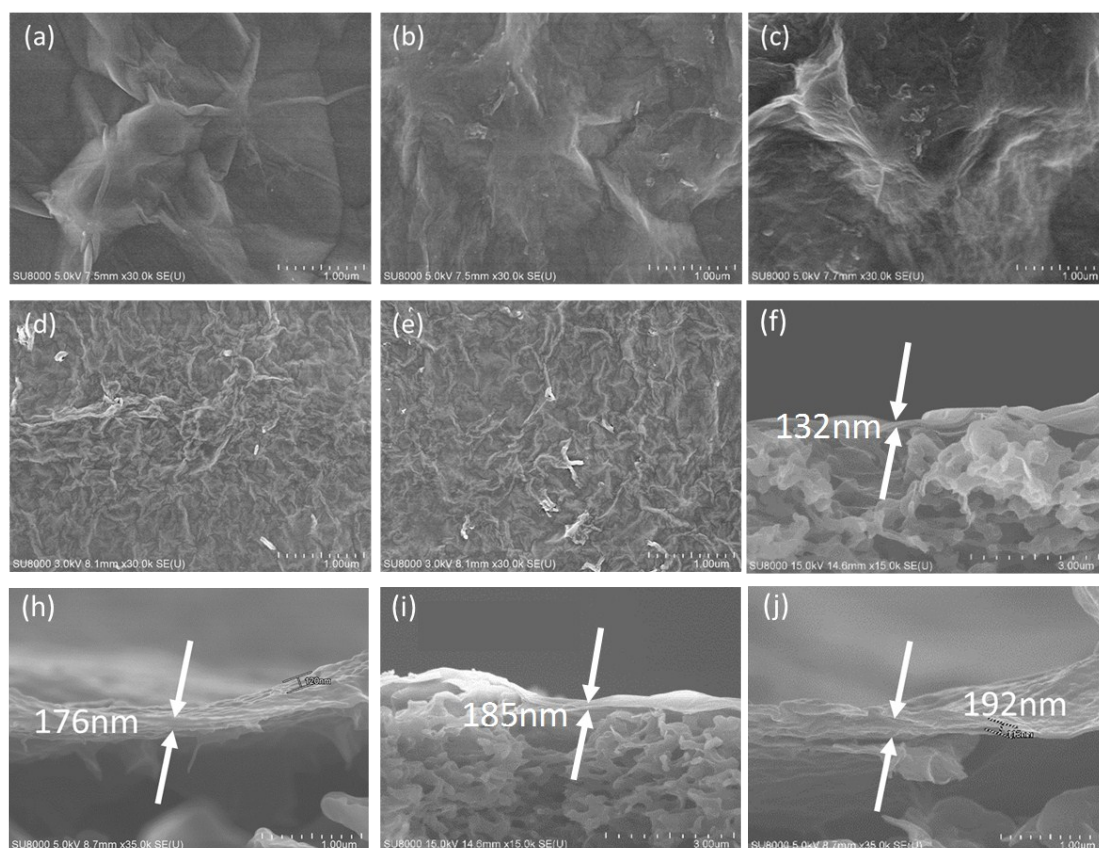


Fig. S2 SEM images of the GCMs with different CNT loading from 0 to 80%. (a) rGO membrane, (b) GCM-15, (c) GCM-33, (d) GCM-50, (e) GCM-80, and cross-sectional image of (f) rGO, (h) GCM-15, (i) GCM-33, (j) GCM-50.

X-ray photoelectron spectroscopy (XPS) analysis of the oxygen-related functional groups in the GCMs

From the C1s XPS spectra of the GCMs (Fig. S3), four peaks centered at 284.7, 286.2, 287.6, and 288.9 eV appeared, assigned to C–C, C–OH, C=O, and O–C=O groups, respectively.^{3, 4} The peak of C=O was not observed in the XPS spectra of the rGO membrane. The intensity of C–C group and C/O ratio increased when the CNT fraction increased from 0 to 80%. Based on the atomic ratios obtained from the XPS spectrum of the GCMs in Fig. S3e, the O/C ratio of the rGO membrane, GCM-15, GCM-33, GCM-50, and GCM-80 are calculated to be 0.18, 0.17, 0.15, 0.15, and 0.14,

respectively.

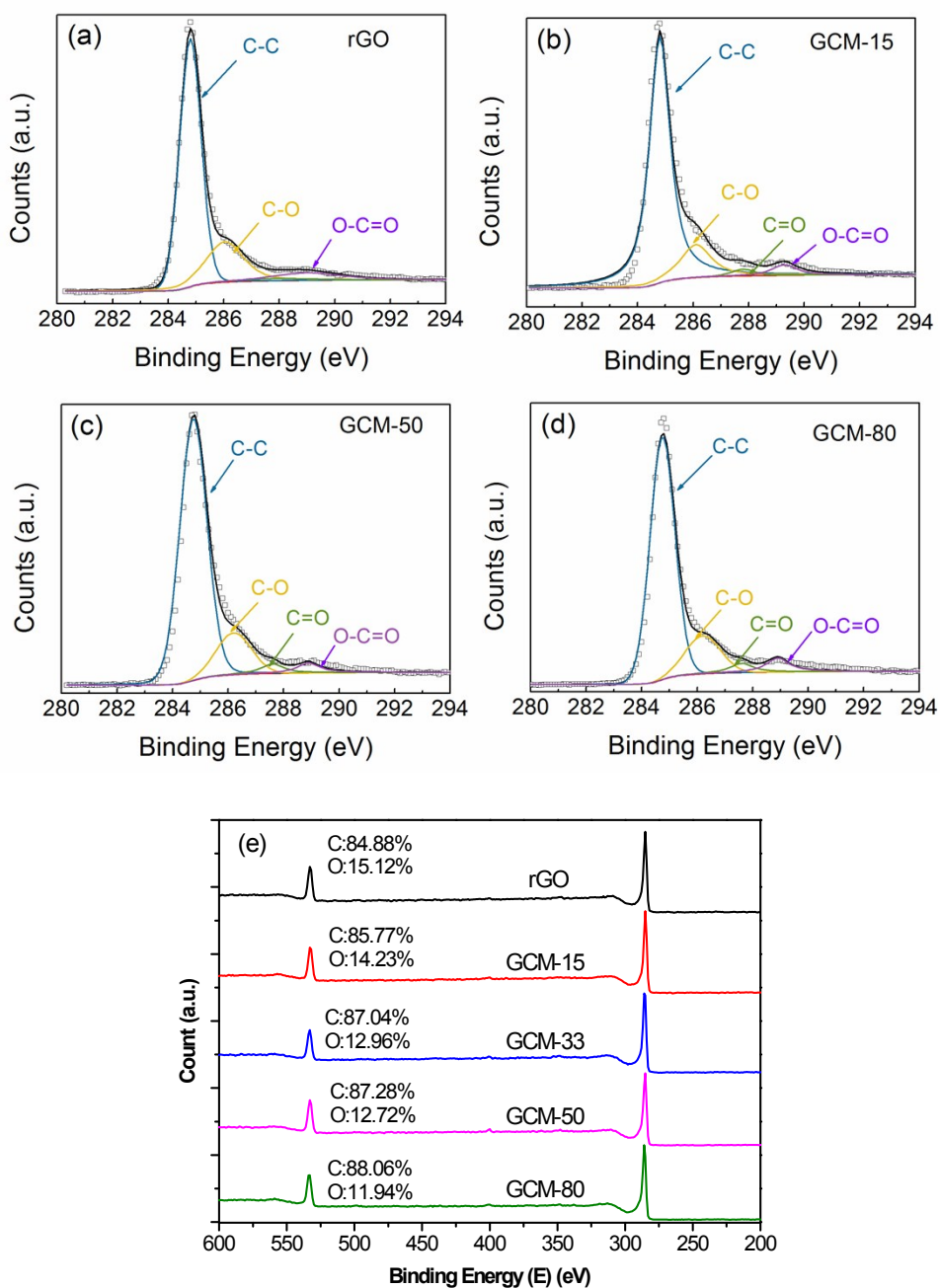


Fig. S3 XPS spectra of the GCMs with various CNT content

The Atomic Force Microscope (AFM) images of the GCMs and the corresponding roughness values

The AFM images (Fig. S4) show that the GCMs had a rough surface. The corresponding Ra value of the rGO membrane, GCM-15, GCM-33, GCM-50, and

GCM-80 were 230, 270, 255, 244, and 235 nm, respectively.

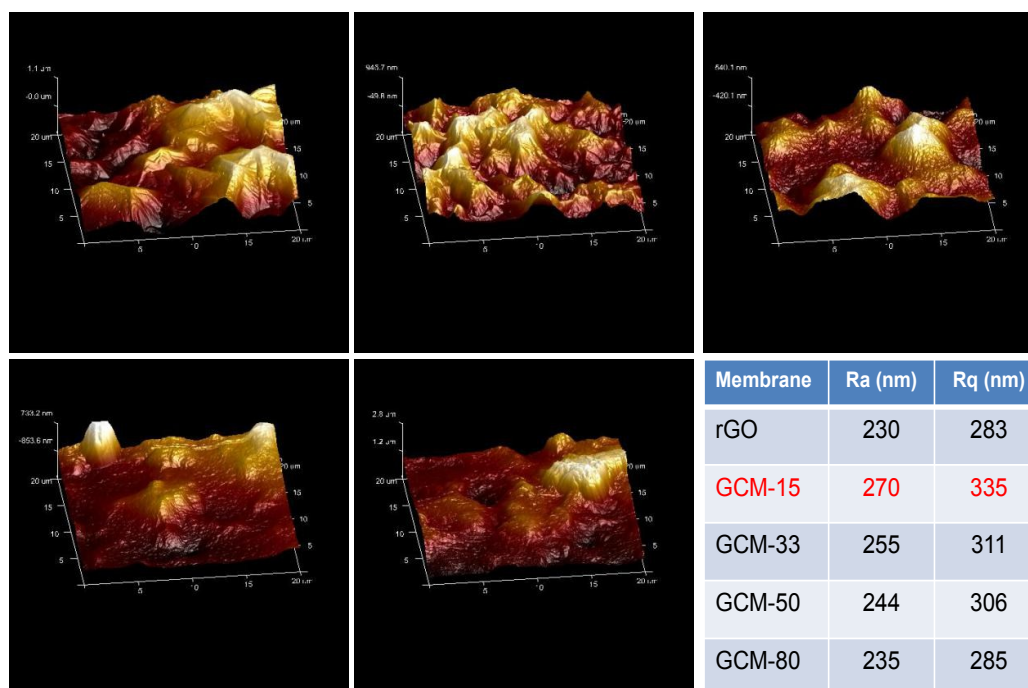


Fig. S4 AFM images of GCMs with various CNT content and the corresponding roughness values

Conductivities of applied salt solutions

In order to illuminate the ion permselectivity of the GCM under bias, we evaluated the NaCl, Na₂SO₄, and CaCl₂ rejection in the same concentration (0.05 mol/L) and in the same ionic strength (0.05 mol/L). The corresponding conductivities for various salt solutions were listed in Table S1.

Table S1. The electric conductivities of salt solutions with the same concentration and ionic strength.

	Conductivity (mS/cm)		
	NaCl	Na ₂ SO ₄	CaCl ₂
Concentration (0.05 mol/L)	4.55	7.34	8.49
Ionic strength (0.05 mol/L)	4.55	2.85	3.12

References

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