Supporting Information

Highly Ionic Conductive and Mechanically Strong MXene/CNF Membranes for Osmotic Energy Conversion

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1. Interlayer spacing of composite membrane

In the case of MXene/CNF composite membrane, according to the equation (S1), the interlayer spacing between nanosheets can be calculated with the angle of (002) peak:[1]

$$2d\sin\theta = n\lambda \tag{S1}$$

Here, d is the layer spacing, θ is half the angle of (002) peak or between the incident wave and the scattering plane, λ is the wavelength of the incident wave (λ= 0.154 nm) and n=1 in equation (S1).

2. Surface Charge Density

Then we can estimate the surface charge density according to the equation (S2),[2]

$$\sigma = \frac{\varepsilon\varepsilon_0\zeta}{\lambda_d} \tag{S2}$$

in which σ is the surface charge, ε is the dielectric constant, ε_0 is the permittivity of vacuum, λ_d is the Debye length, and ζ is the zeta potential in equation (S2).

3. Energy Conveision Efficiency

The cation transference number under a specific concentration gradient can be calculated as:[3]

$$t_+ = \left(\frac{V_O}{(RT/zF)\ln\left(\frac{r_{c_H} c_H}{r_{c_L} c_L} \right)} + 1 \right) / 2 \tag{S3}$$

Herein, V_O, R, T, F, z, r, and c refer to the open-current potential, universal gas constant, absolute temperature, Faraday constant, charge number, activity coefficient of ions, and ion concentration, respectively.

Energy conversion efficiency is defined as the ratio of the electrical energy output to the gibbs free energy input, which can be calculated according to the following equation:

$$\eta_{max} = \frac{(2t_{+} - 1)^2}{2} \tag{S4}$$

4. Output Power Density

According to the following formula, we can calculate the theoretical maximum output power density under different concentration gradients:[4, 5]

$$P = V_0{}^2/4RA = V_O I_S/4A \tag{S5}$$

Here, V_O, I_S, R, A refer to the open-circuit potential, short-circuit current under corresponding concentration gradient, internal resistance, and cross-sectional area of membrane sample, respectively.

Figures and Tables

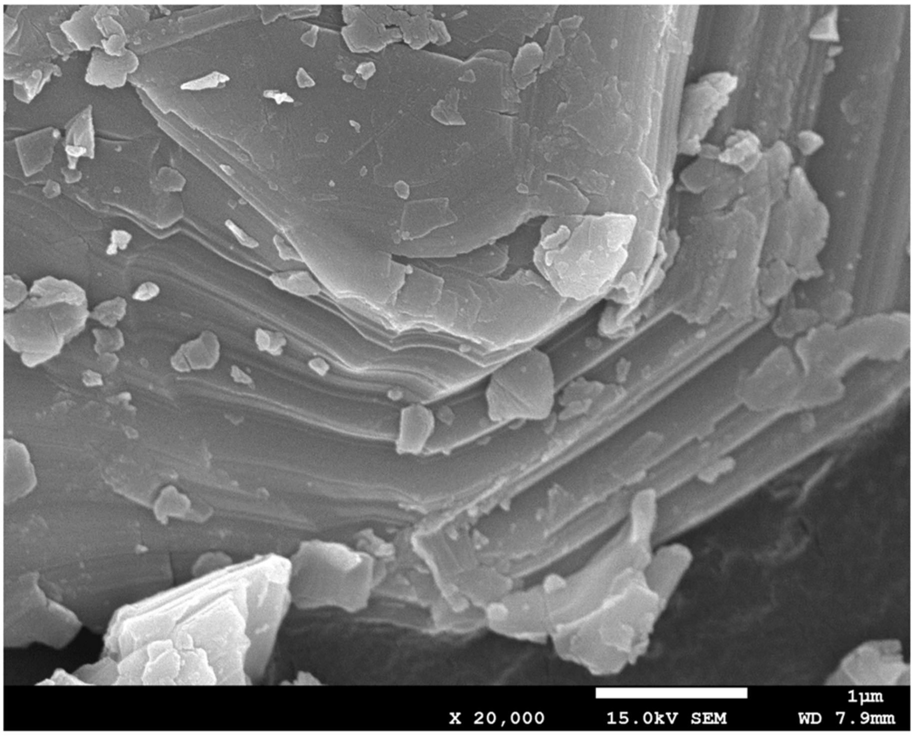

Fig. S1. The SEM image of MAX phase Ti_3AlC_2 powder.

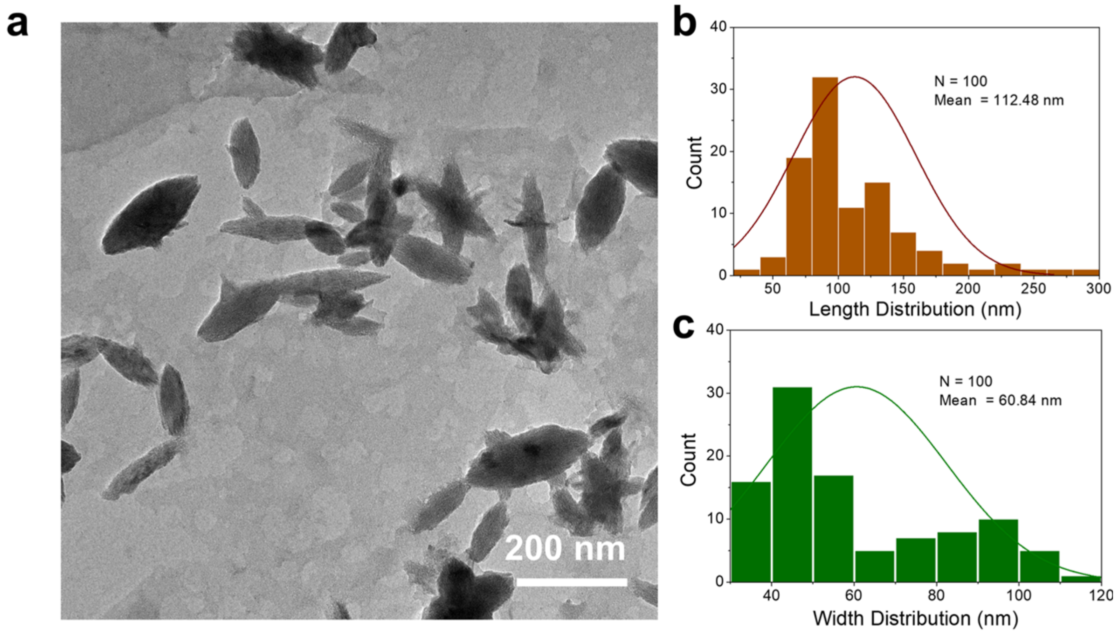

Fig. S2. (a)The Cryo-EM image of MXene nanosheets. The (b) length and (c) width of 100 MXene nanosheets were analyzed and counted according to Cryo-EM images.

Fig. S3. Photograph of a MXene suspension with a concentration of 0.5 $mg \cdot mL^{-1}$. MXene can be dispersed in water to form a stable suspension within at least one month.

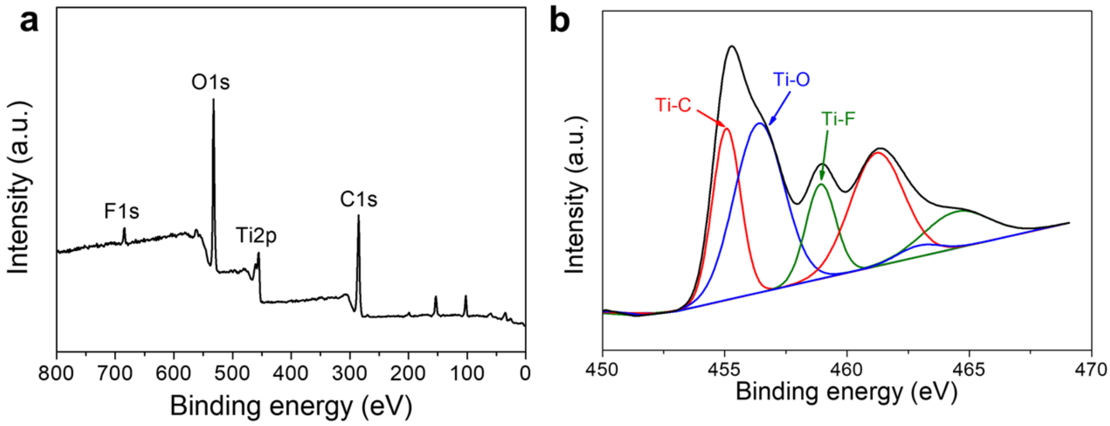

Fig. S4. X-ray photoelectron spectroscopy (XPS) characterization of Mxene membrane. a) XPS survey spectrum and b) high-resolution Ti2p spectrum of MXene,which clearly show the presence of oxygen and fluorine, indicating the existence of -OH and -F surface groups.

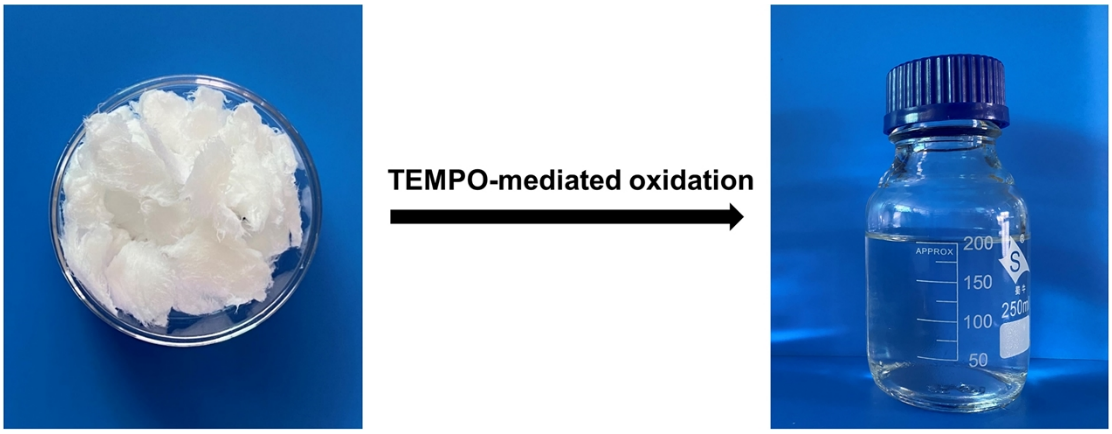

Fig. S5. Transparent CNF suspension deriving from bleached softwood pulp was prepared by TEMPO-mediated oxidation process.

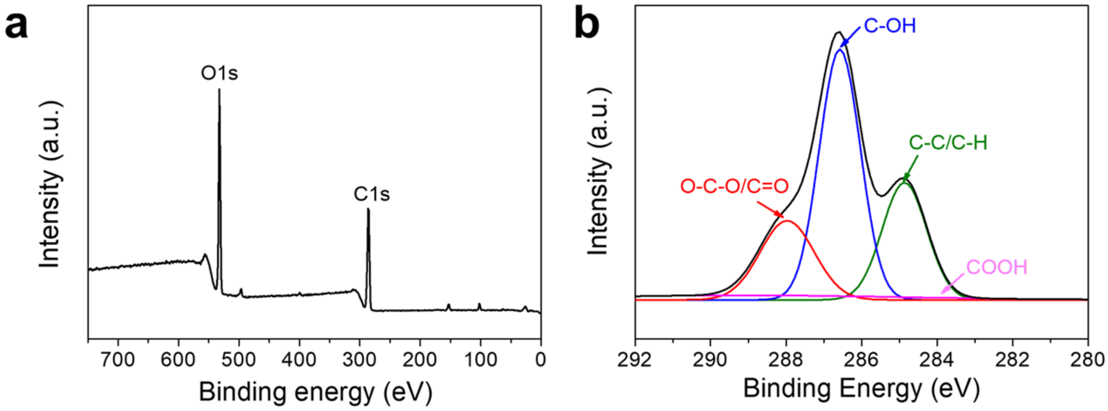

Fig. S6. (a) The XPS of CNF membrane. (b) C1s XPS spectra of the CNF, indicating the existence of surface functional carboxyl groups.

Table S1. Bonding-state peak locations and concentrations of the decomposed C1s energy state of the CNF.

Bond	C-C/C-H	C-OH	O-C-O/C=O	COOH
Bind energy (eV)	284.9	286.6	287.9	289.2
Area (%)	24.51	48.31	20.16	7.02

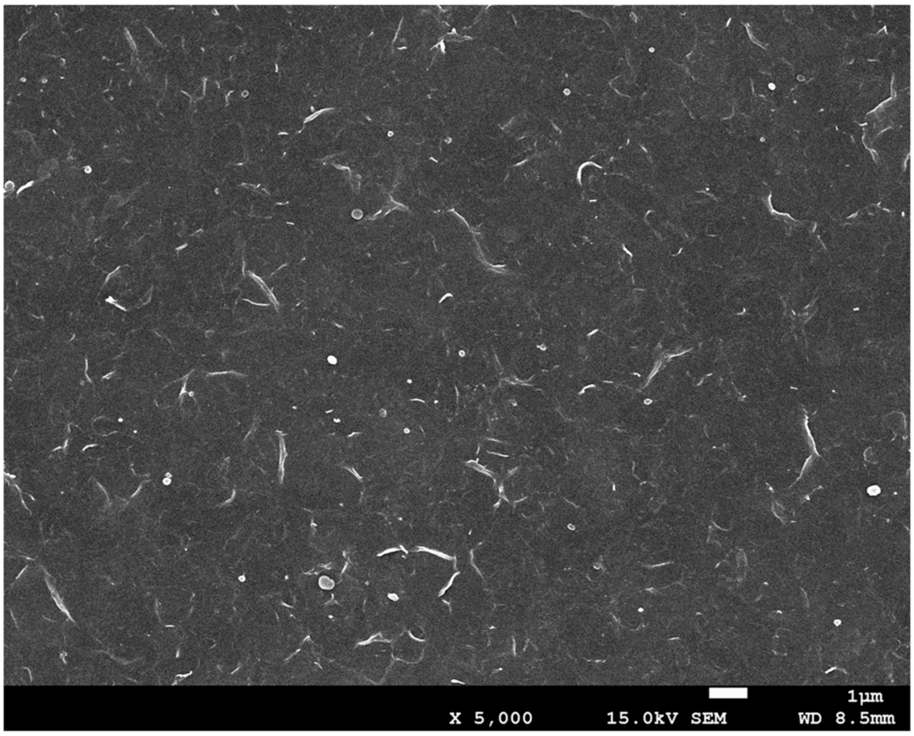

Fig. S7. The SEM image of MXene/CNF composite membrane surface.

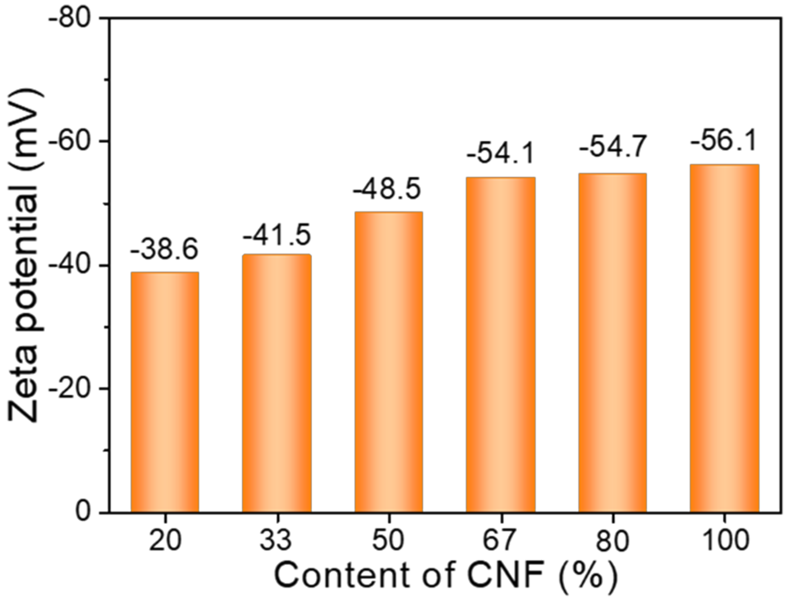

Fig. S8. The Zeta potential of the composite membrane with different CNF content.

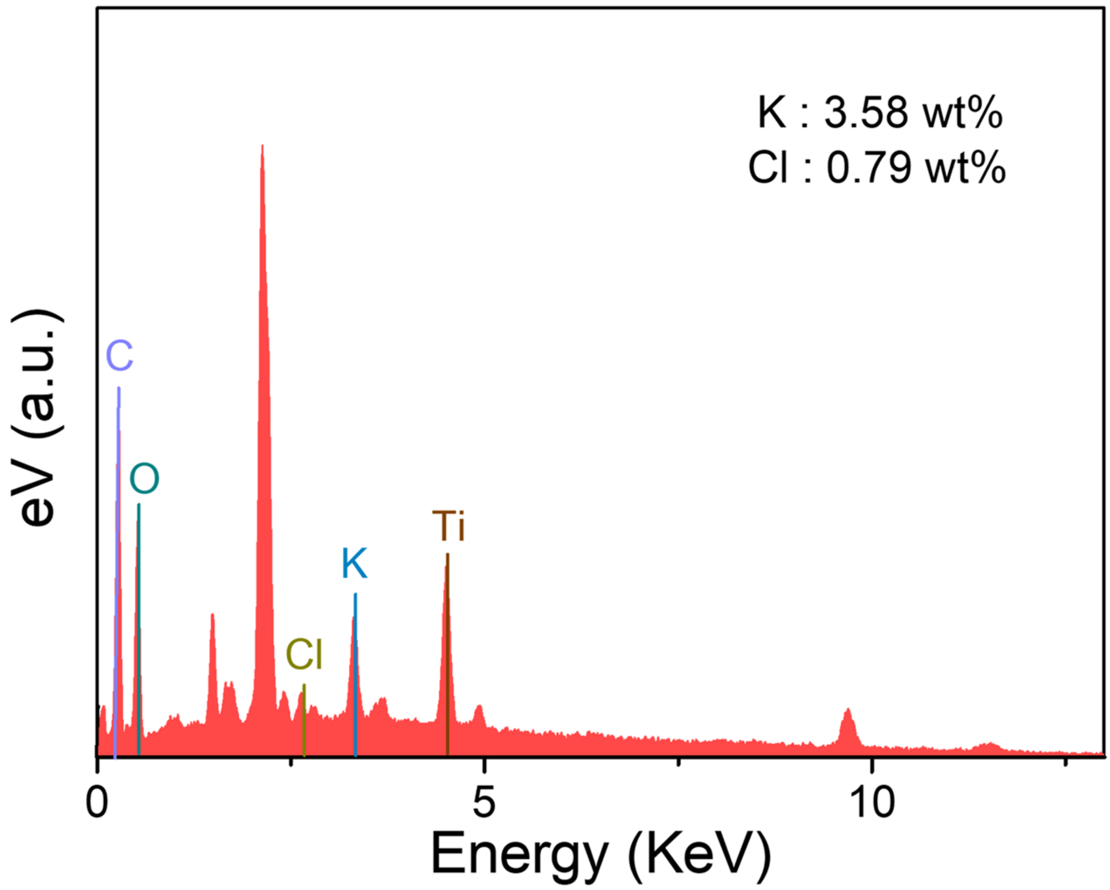

Fig. S9. The energy spectrum of the MXene/CNF composite membrane, in which the content of Cl^- is 0.79 wt% and the content of K^+ is 3.58 wt%, indicates that the composite membrane transports K^+ first and has good cation selectivity.

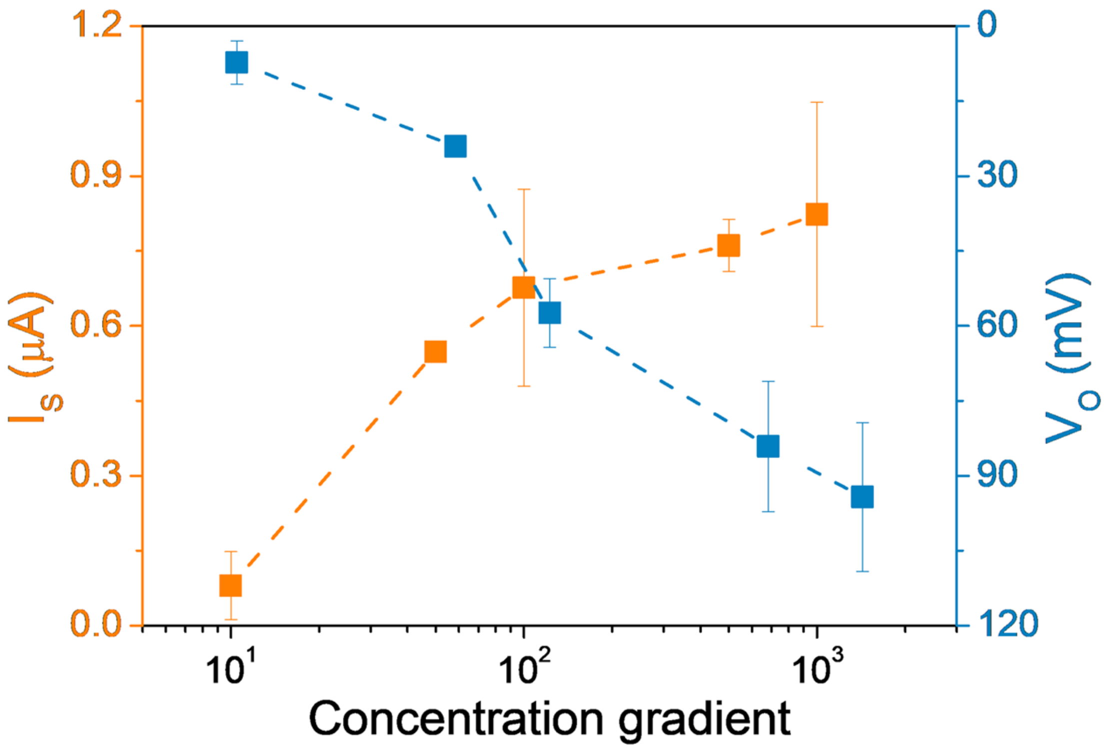

Fig. S10. The open circuit voltage (V_O) and short circuit current (I_S) of membrane (50% CNF) under a seriesof concentration gradients.

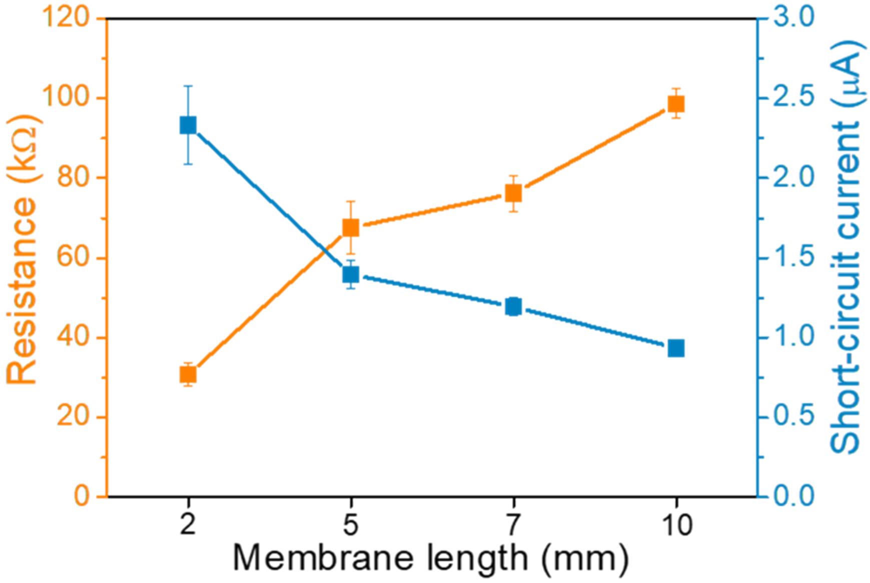

Fig. S11. Resistance and short-circuit current of membrane (50% CNF) with width of 5 mm and lengths of 2 mm, 5 mm, 7 mm and 10 mm under 1000-fold concentration gradient.

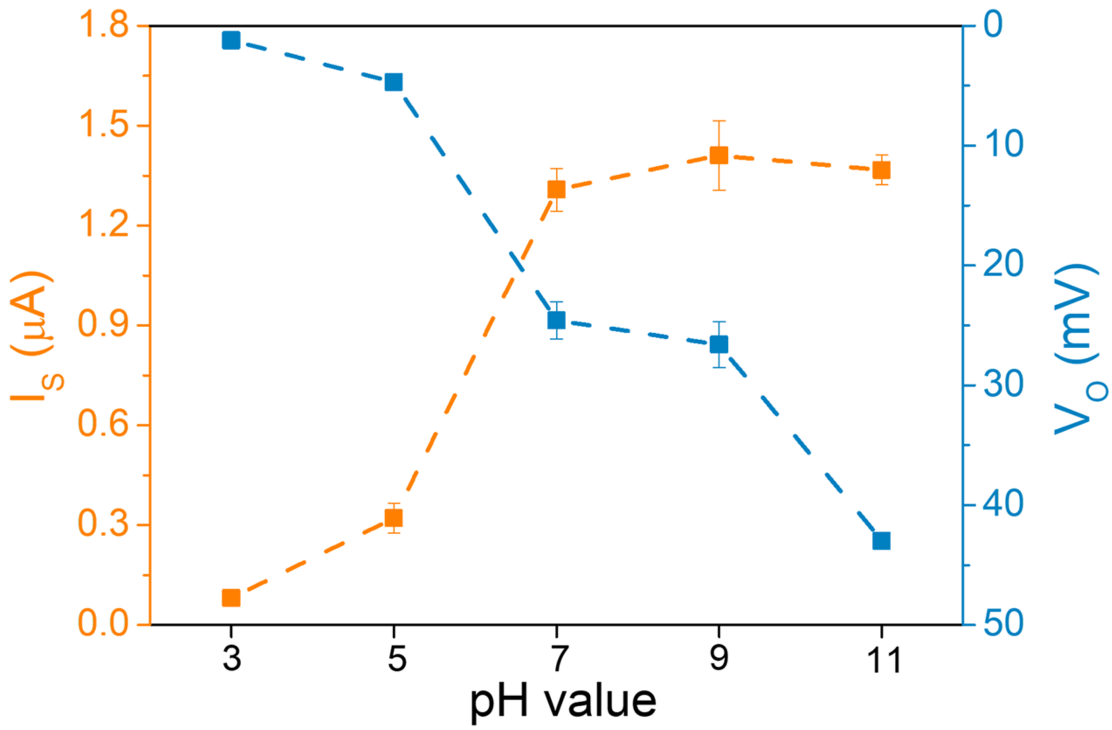

Fig. S12. The osmotic energy conversion performance of the composite membrane with CNF content of 50% over the pH range of 3-11. (C_H = 0.5 M NaCl, C_L = 0.01 M NaCl)

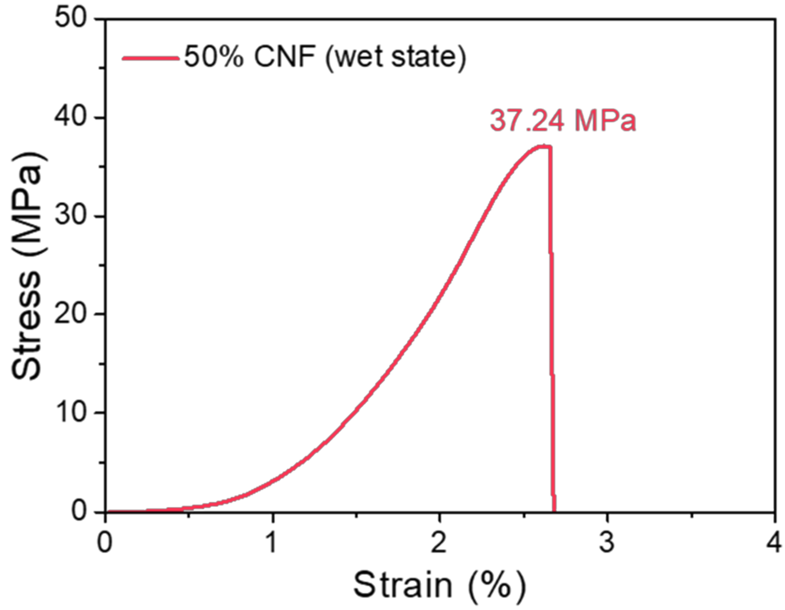

Fig. S13. Good mechanical strength of composite membrane (50% CNF) maintained from wet state.

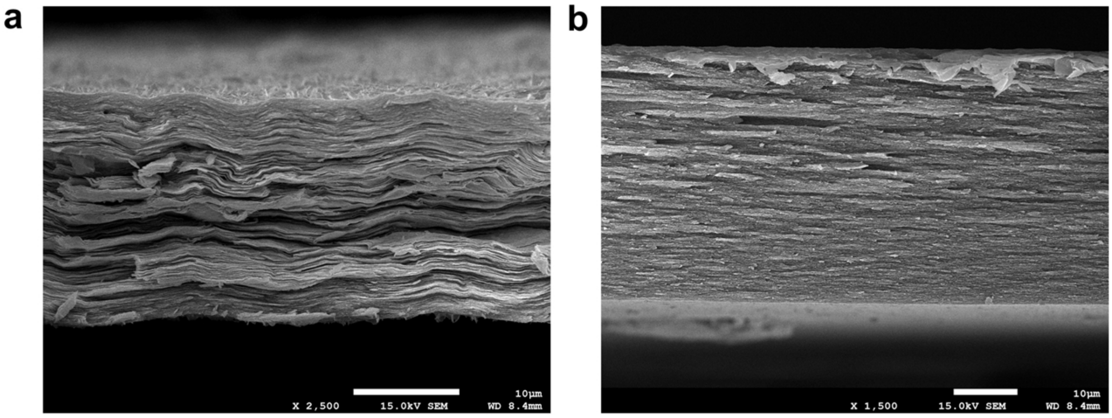

Fig. S14. Cross-sectional SEM images of (a) MXene and (b) CNF membrane after fracture.

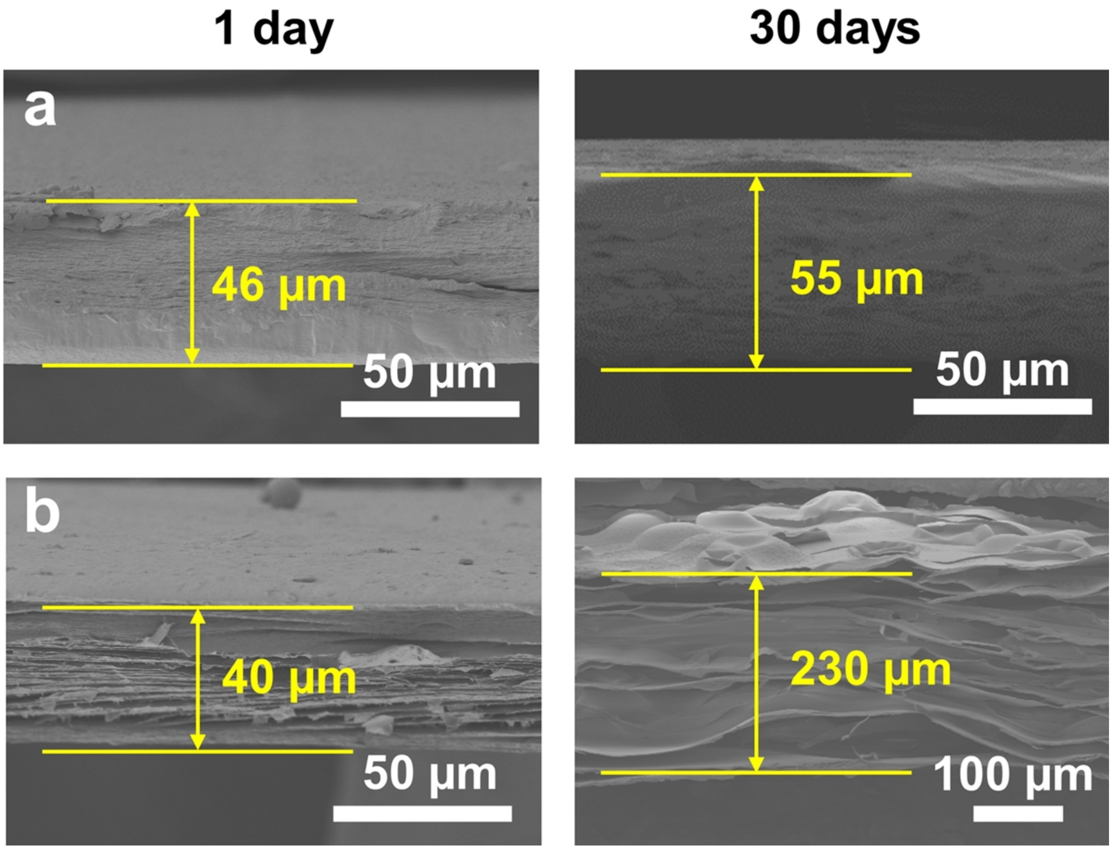

Fig. S15. Cross-sectional SEM images of the (a) composite membrane with 50% CNF and (b) pure CNF membrane before and after soaking in 1M KCl solution for 90 days.

Reference

1 C. Zhu, P. Liu, B. Niu, Y. Liu, W. Xin, W. Chen, X. Y. Kong, Z. Zhang, L. Jiang and L. Wen, *J Am. Chem. Soc.*, 2021, DOI: 10.1021/jacs.0c11251.

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