

Supplementary Information

Title

Functionally Antagonistic Polyelectrolyte for Electro-Ionic Soft Actuator

Authors

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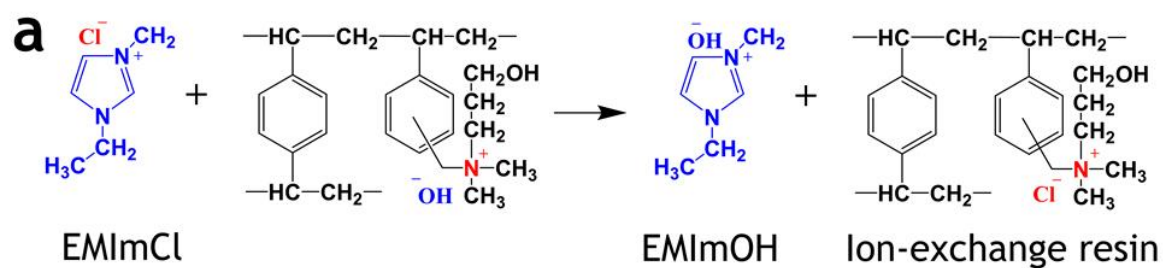
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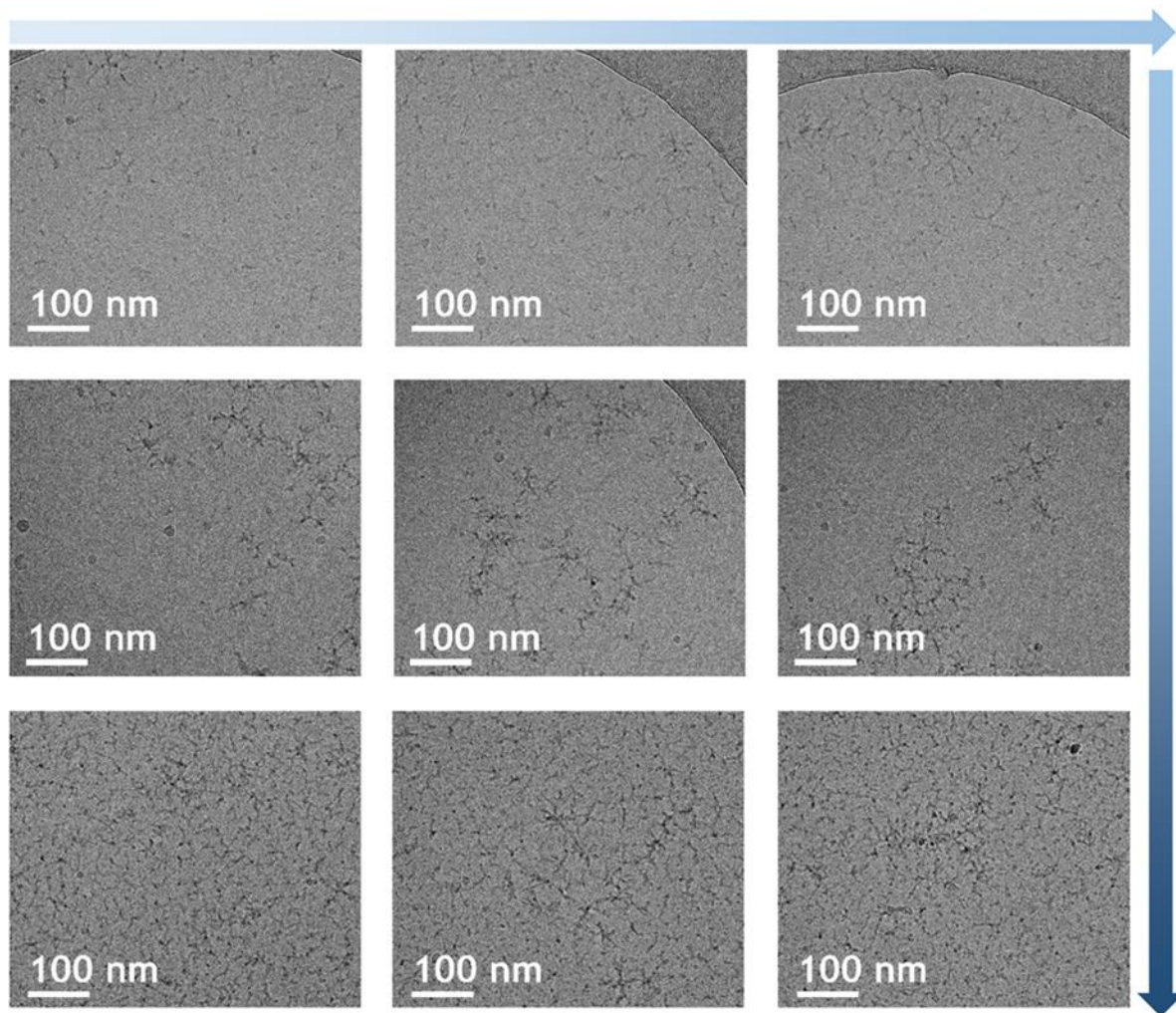
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Supplementary Figures and Tables

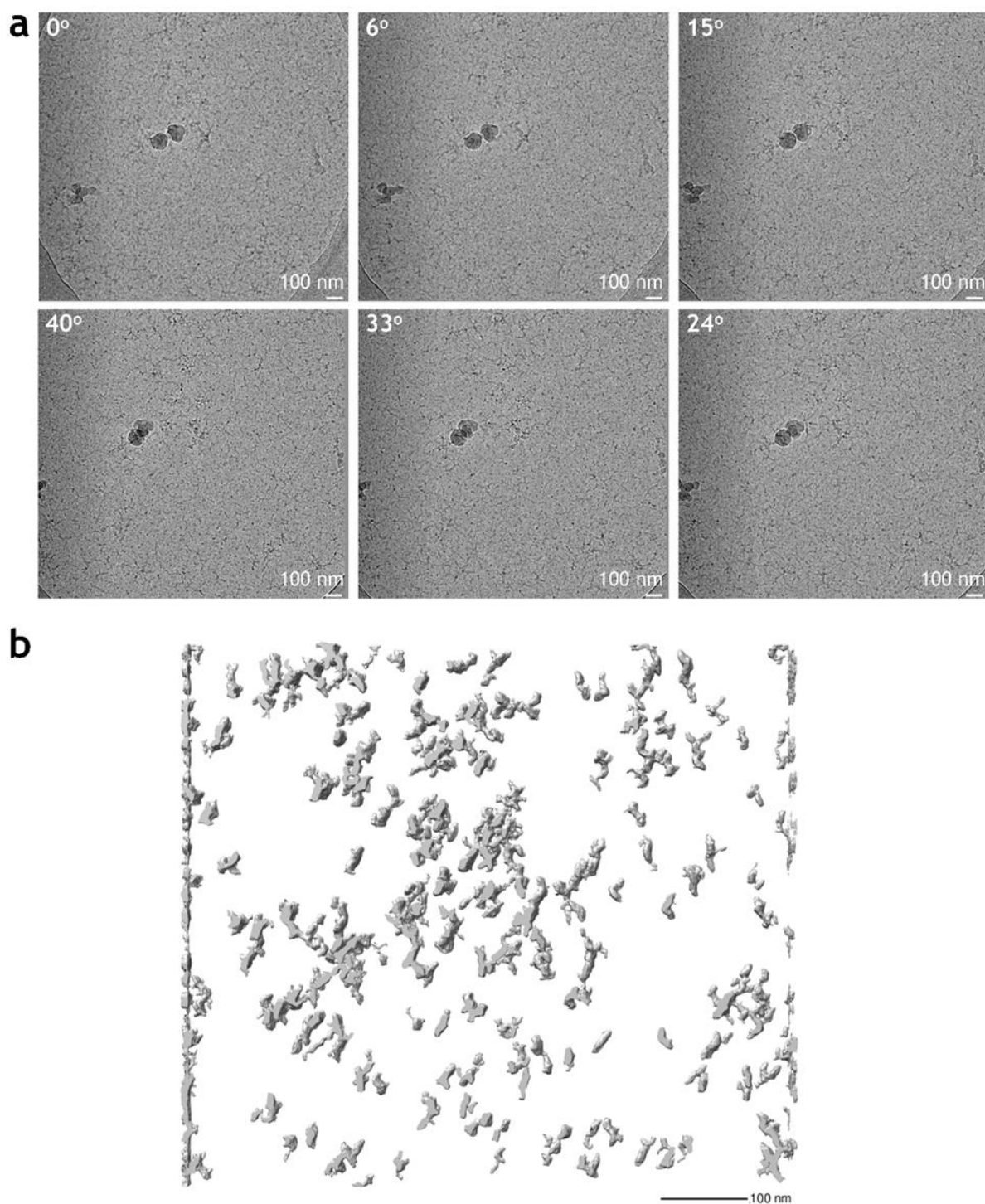


Supplementary Fig. 1. Preparing 1-ethyl-3-methylimidazolium hydroxide (EMImOH) by ion-exchange reaction. a. Scheme for ion-exchange reaction. **b.** Experimental setup for ion-exchange reaction. **c.** EMImOH product.

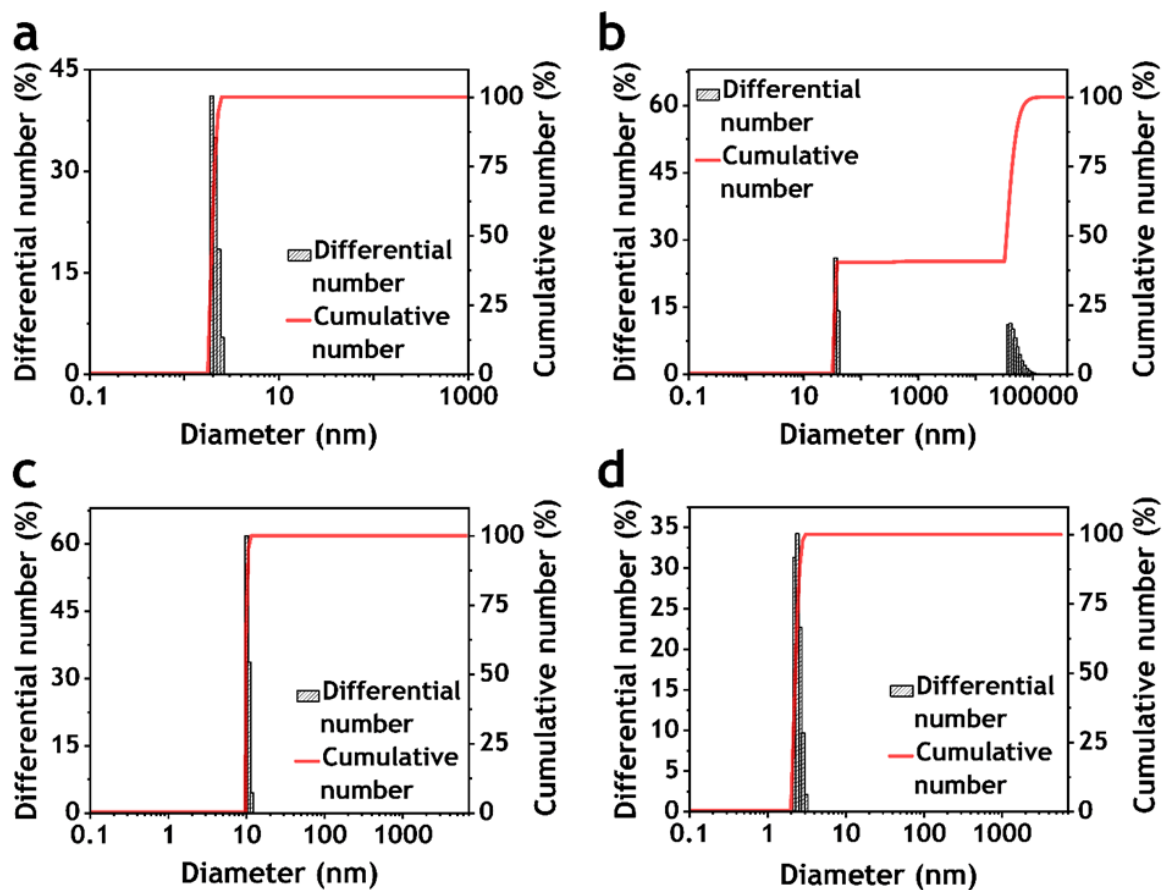


Supplementary Fig. 2. Cryo-TEM images of Nafion micelles at different concentrations.

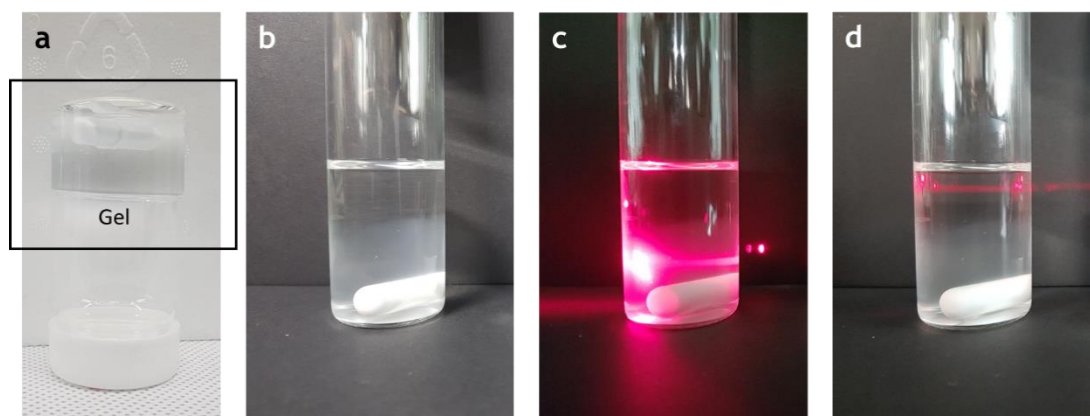
The arrow directions indicate the increases of concentration.



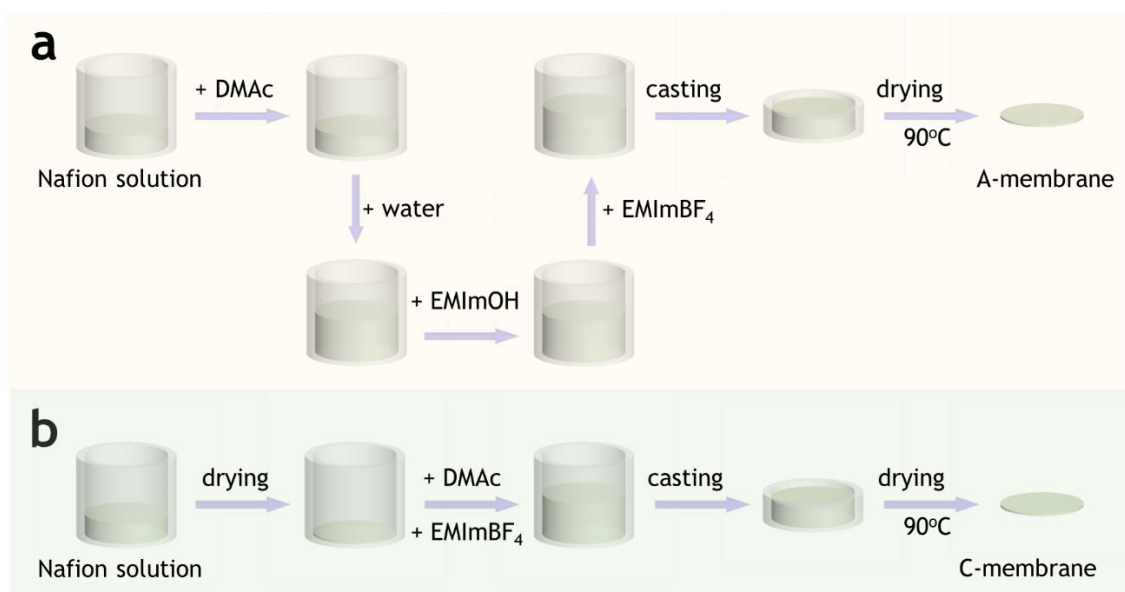
Supplementary Fig. 3. Cryo-TEM images of Nafion micelle. **a.** Images at the same position and different tilting angles. **b.** 3D image reconstructed from cryo-TEM tomography using software. The 3D image shows distorted micellar shapes in comparison to the real ones due to some noises while reconstructing from raw data with the help of software.



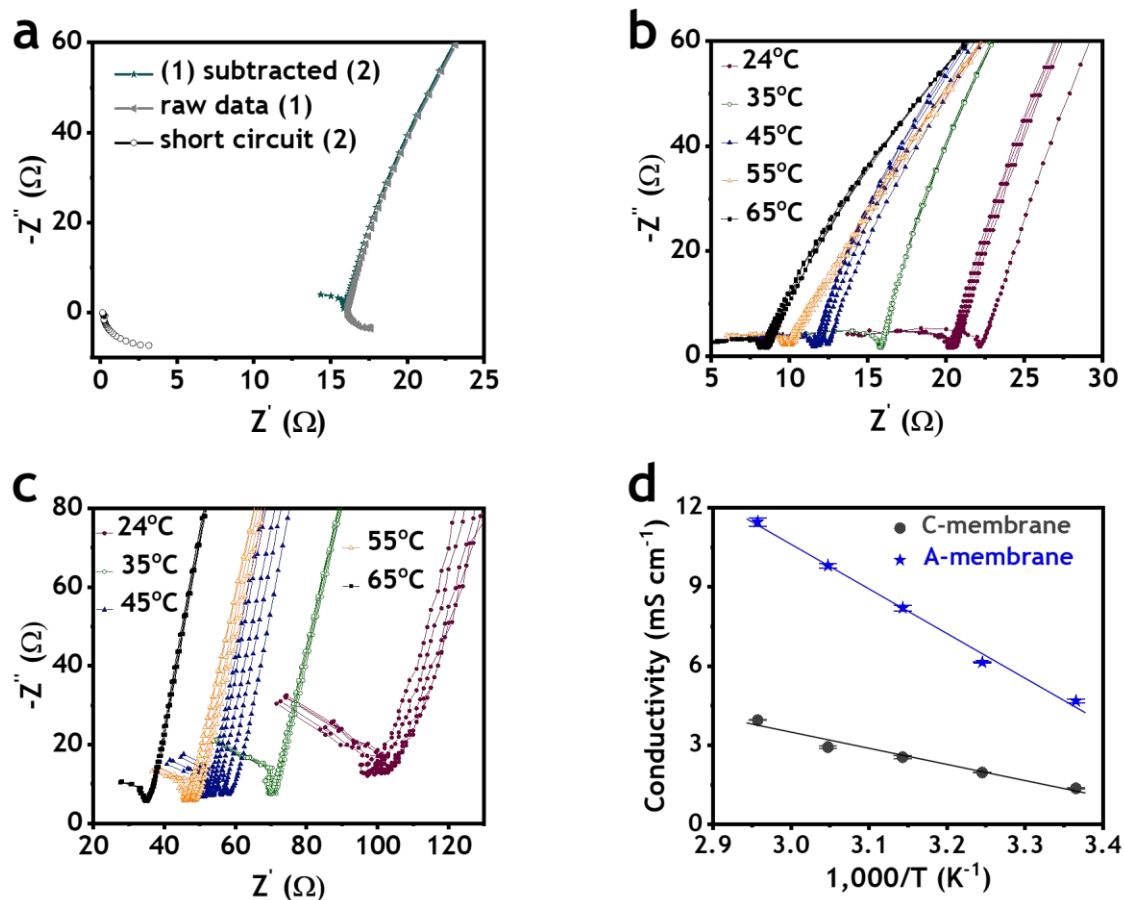
Supplementary Fig. 4. Particle size by dynamic light scattering (DLS). Nafion dispersions in water **a.** Before and **b.** After adding EMImBF₄ ionic liquid. Nafion dispersions in DMAc **c.** Before and **d.** After adding EMImBF₄ ionic liquid.



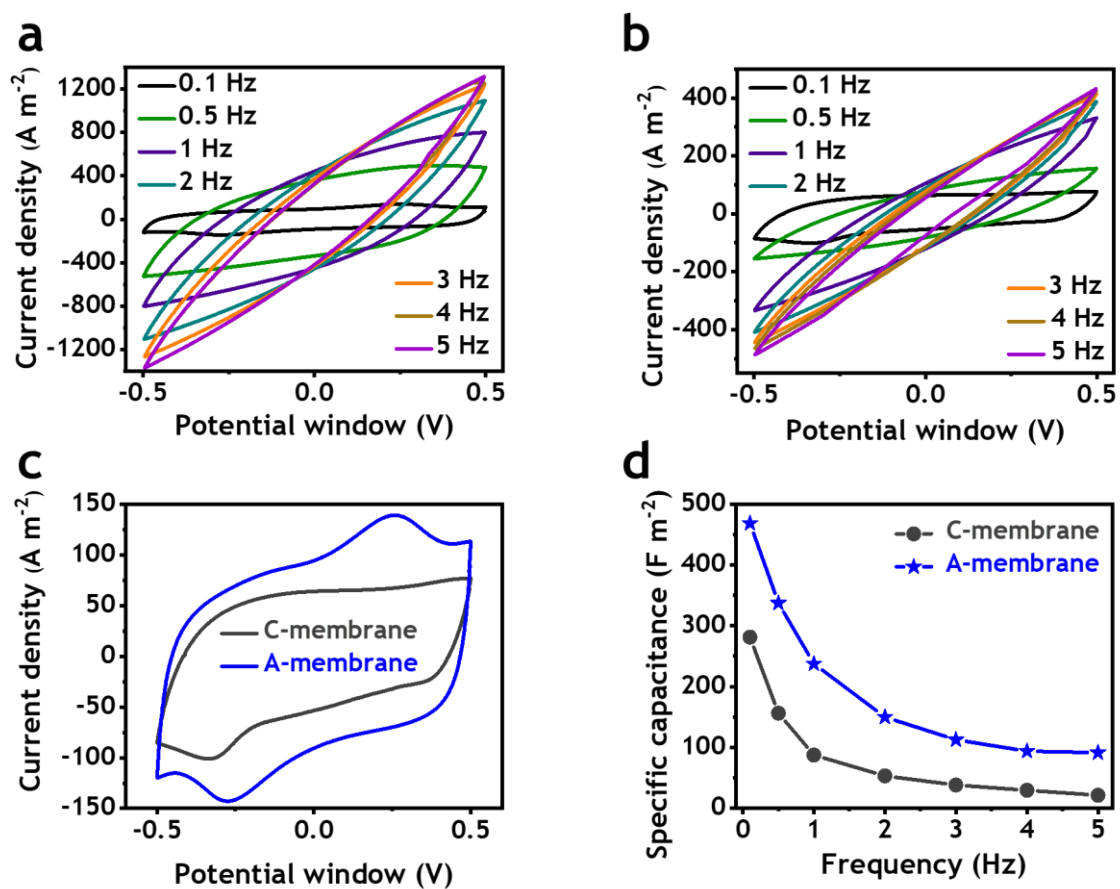
Supplementary Fig. 5. Effect of water content on micellar rheology. **a.** strong Nafion gel in low water content. **b.** precipitated Nafion micelles in high water content. **c.** Laser scattering of precipitated Nafion micelles in high water content. **d.** Laser propagation at high position.



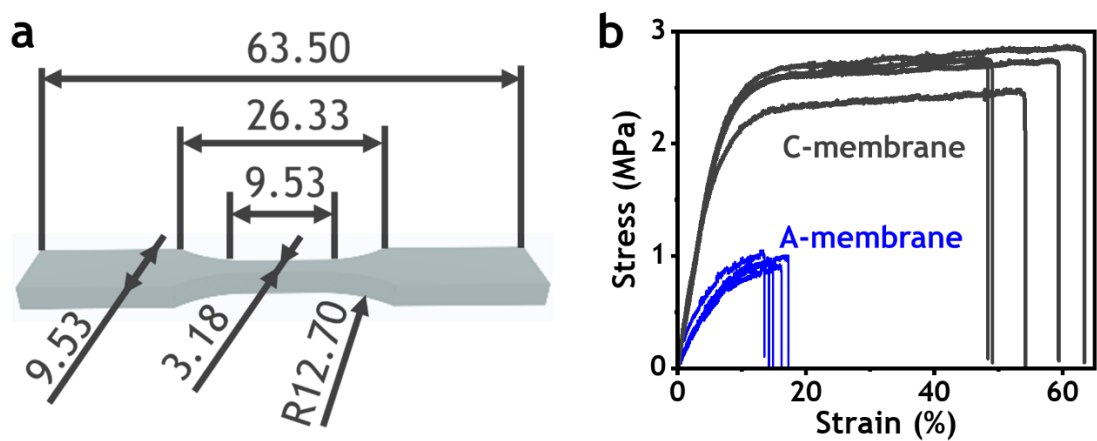
Supplementary Fig. 6. Procedures for fabricating polyelectrolyte membranes. a. Antagonistic membrane (A-membrane). **b.** Conventional membrane (C-membrane).



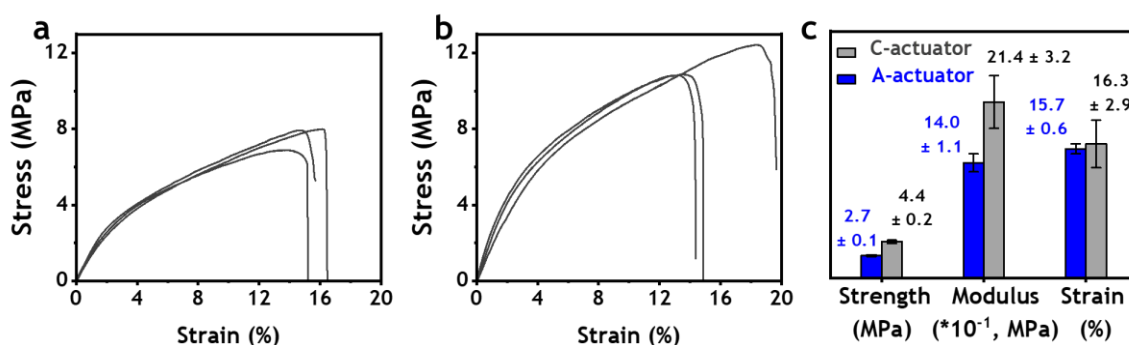
Supplementary Fig. 7. Ionic conductivity. **a.** Calibration of raw electrochemical impedance spectroscopy (EIS) data. **b.** EIS spectra of A-membrane. **c.** EIS spectra of C-membrane. **d.** Ionic conductivity calculated from EIS data.



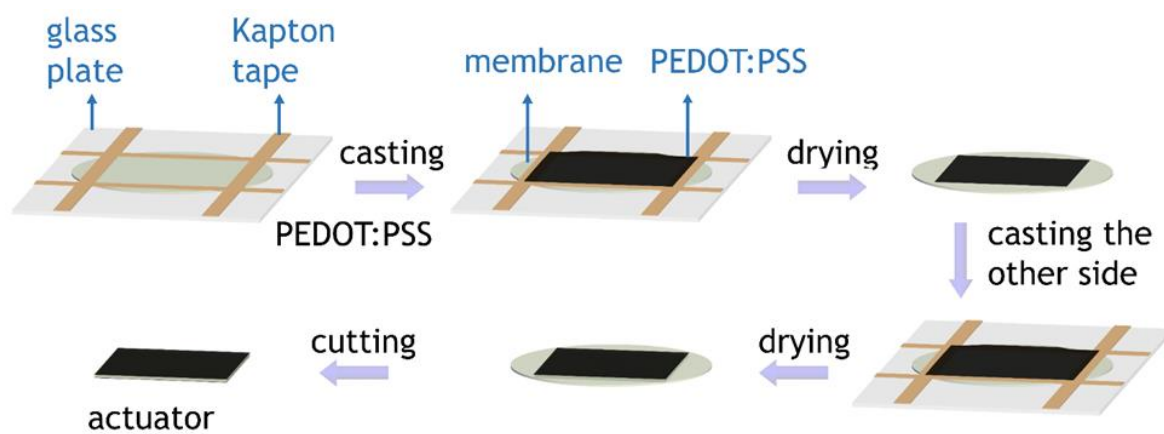
Supplementary Fig. 8. Specific capacitance of actuators. **a.** Cyclic voltammetry (CV) curves of A-membrane actuator. **b.** CV curves of C-membrane actuator. **c.** Typical comparison of CV curves of the two samples at 0.2 V s^{-1} (0.1 Hz). **d.** Specific capacitance calculated from CV data.



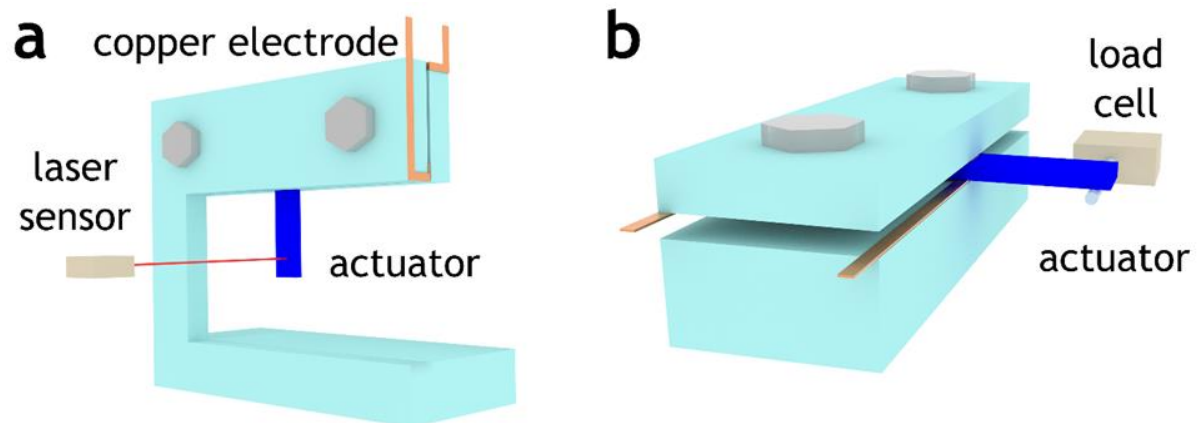
Supplementary Fig. 9. Mechanical properties. **a.** Dimensions of tensile test sample according to ASTM D638-V. **b.** Stress-strain curves.



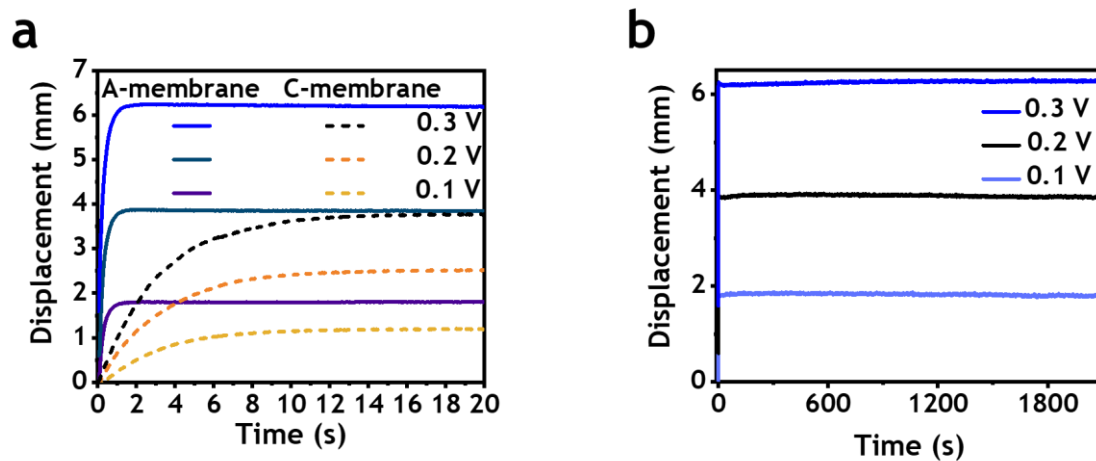
Supplementary Fig. 10. Mechanical properties of actuators. a. Stress-strain curves of A-actuator. **b.** Stress-strain curves of C-actuator. **c.** mechanical properties.



Supplementary Fig. 11. Procedure for fabricating actuator.

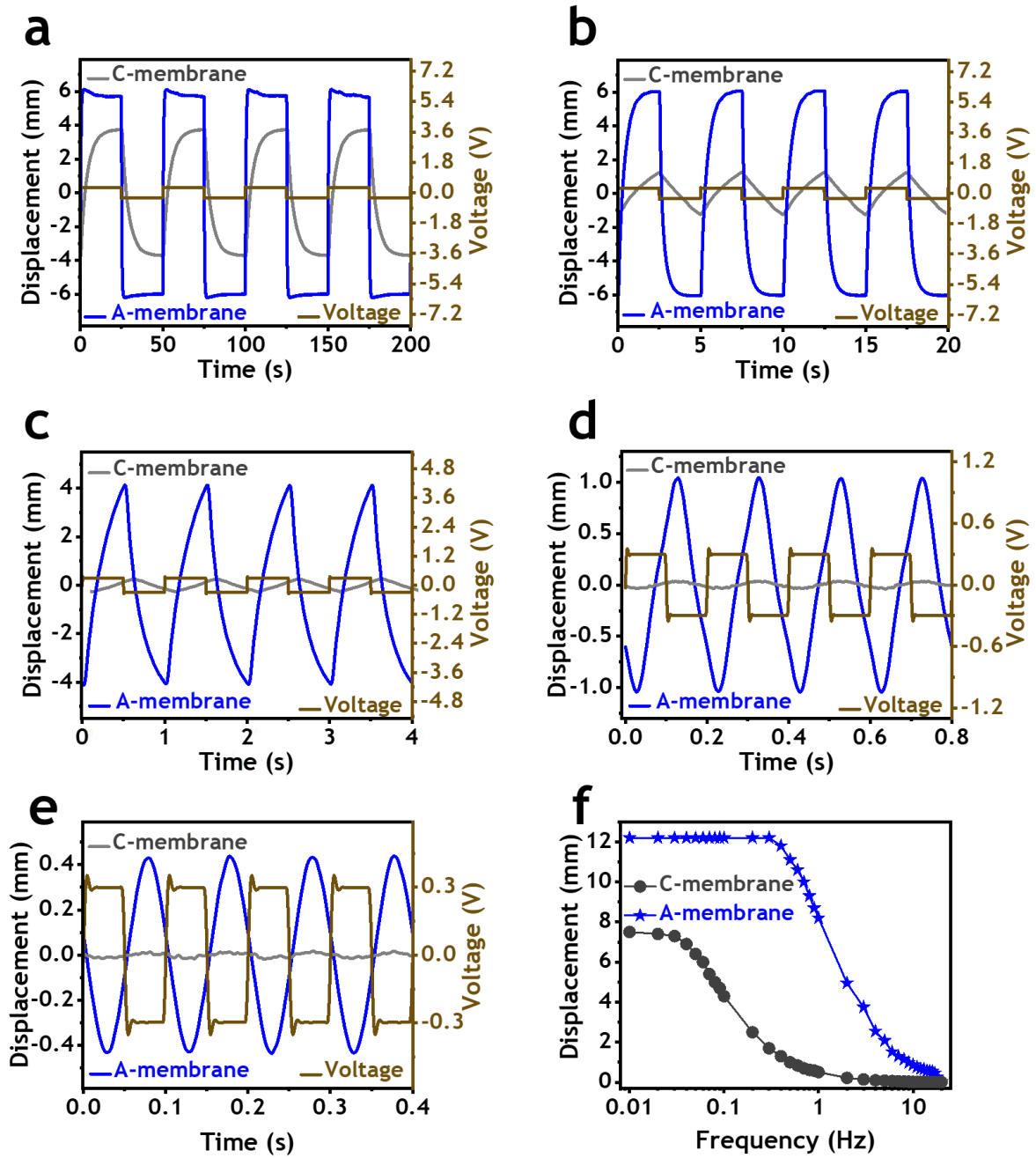


Supplementary Fig. 12. Actuator characterizations. Measurement setups for **a**. Bending displacement and **b**. Blocking force.

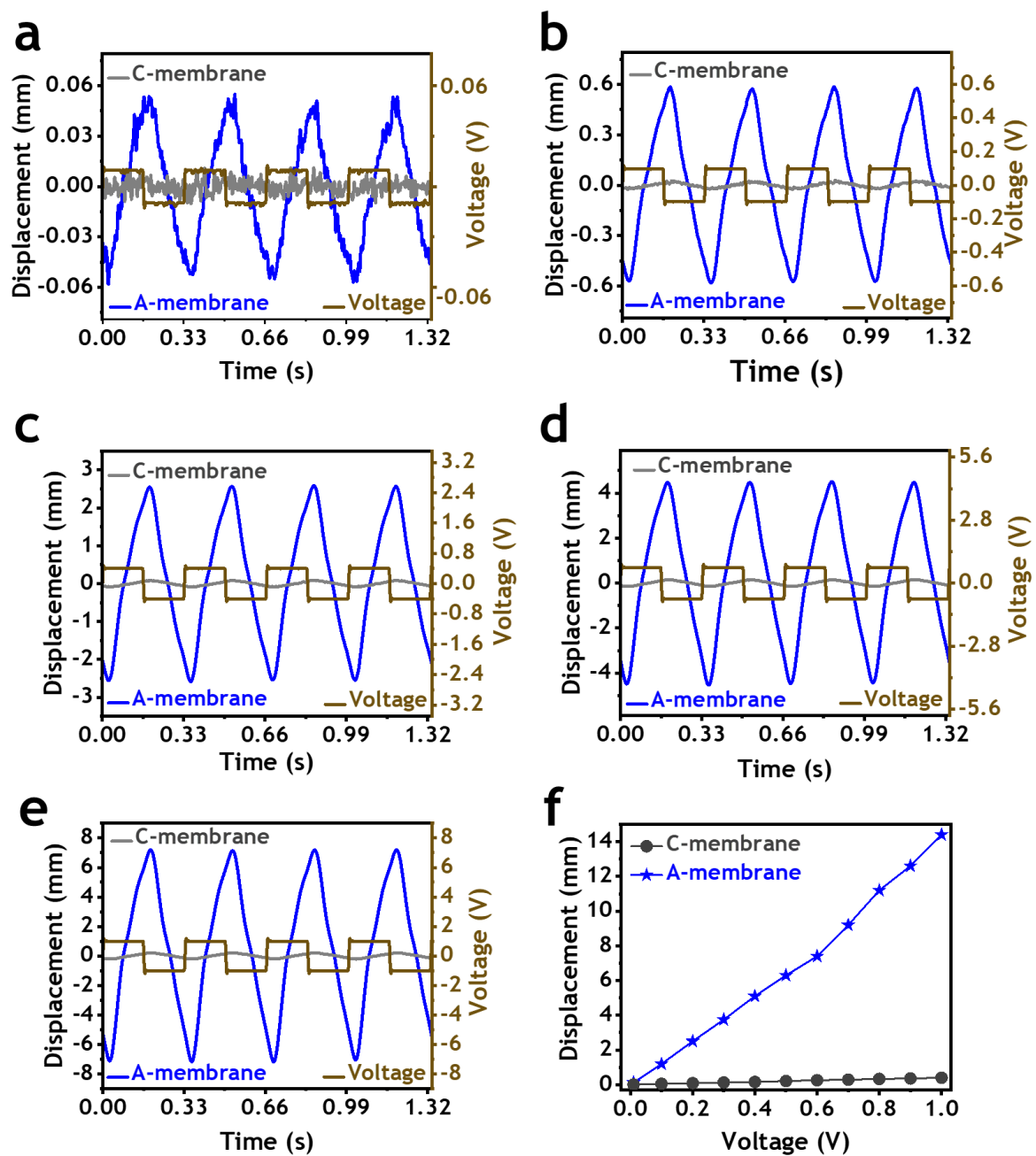


Supplementary Fig. 13. Direct current (DC) response. a. DC response at different voltages.

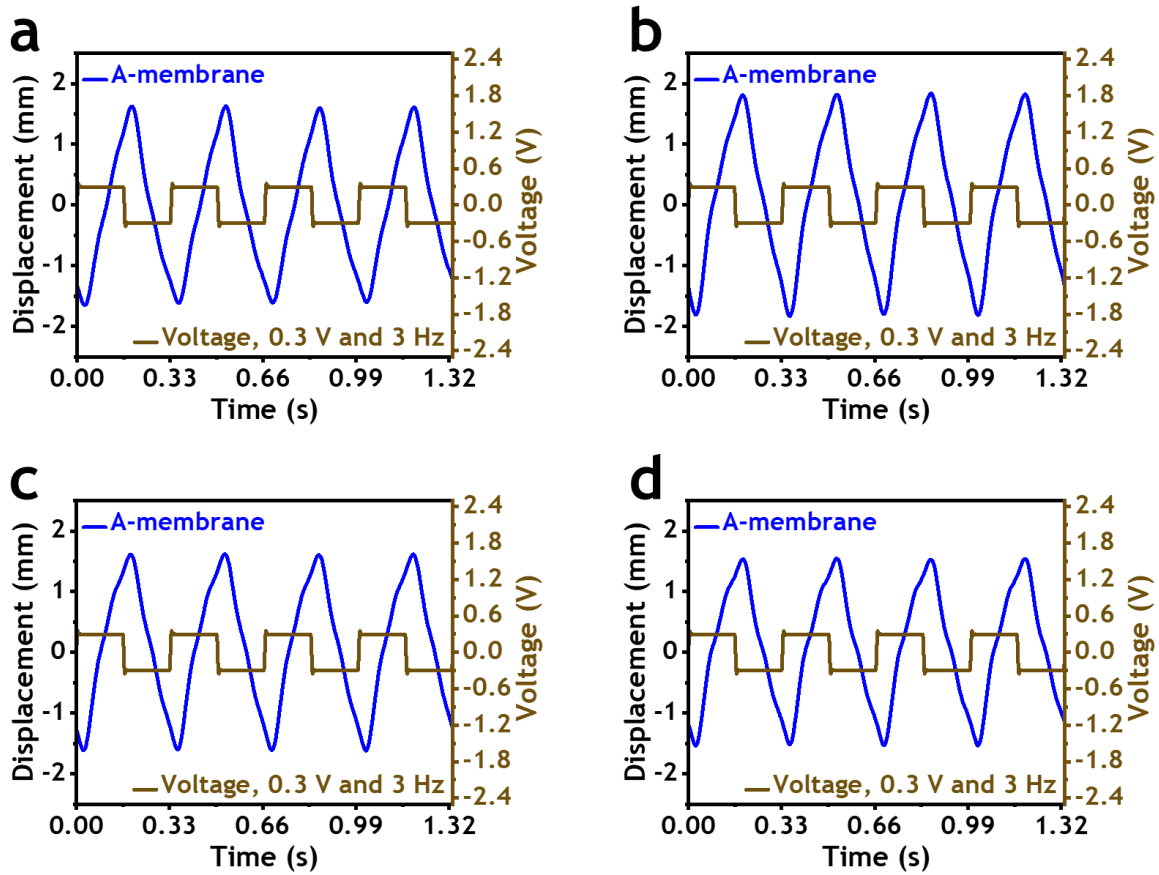
b. DC response over 35 minutes.



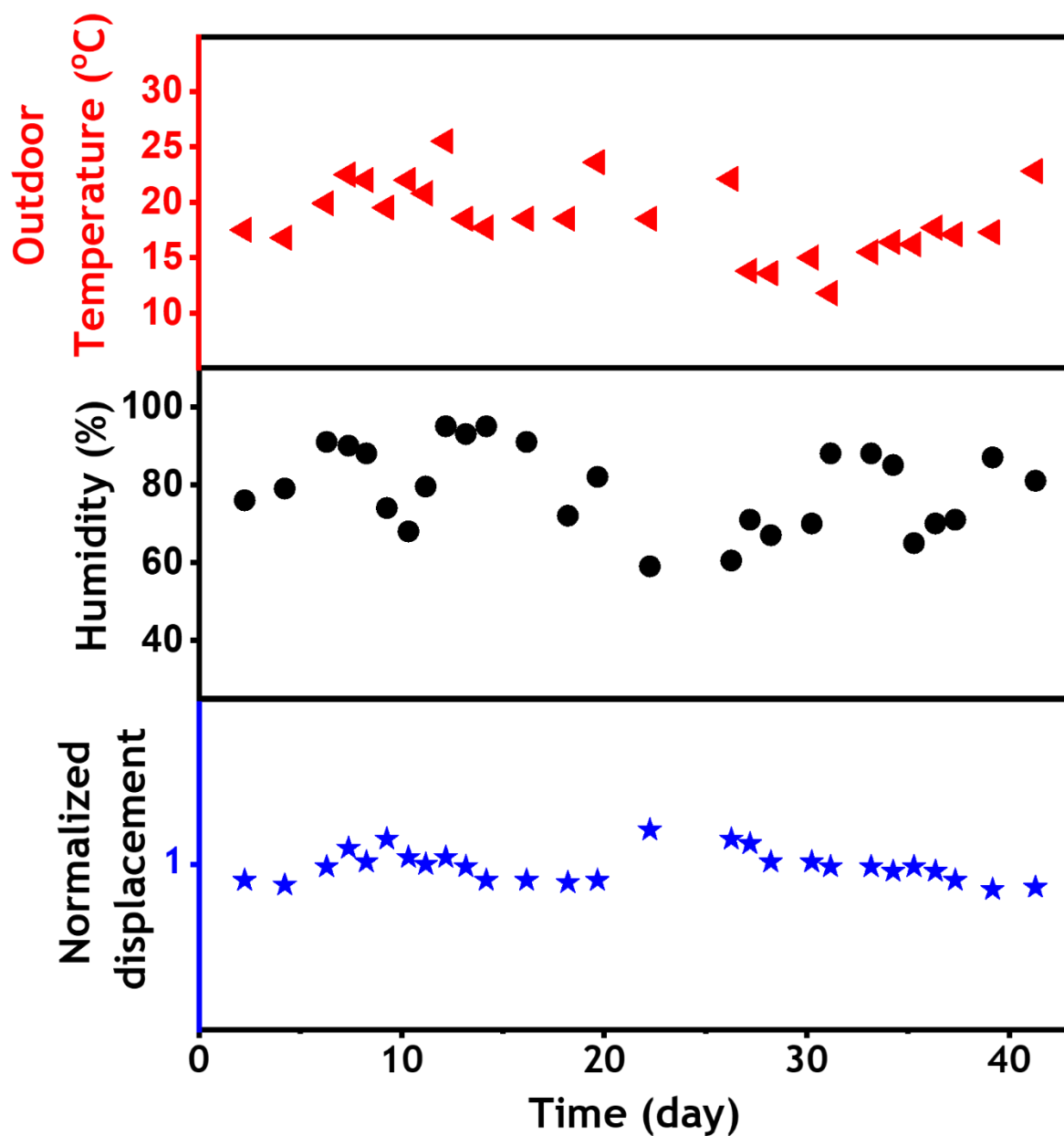
Supplementary Fig. 14. Bending displacement according to frequency. a. 0.02 Hz. b. 0.20 Hz. c. 1.00 Hz. d. 5.00 Hz. e. 10.00 Hz. f. From 0.01 to 20.00 Hz.



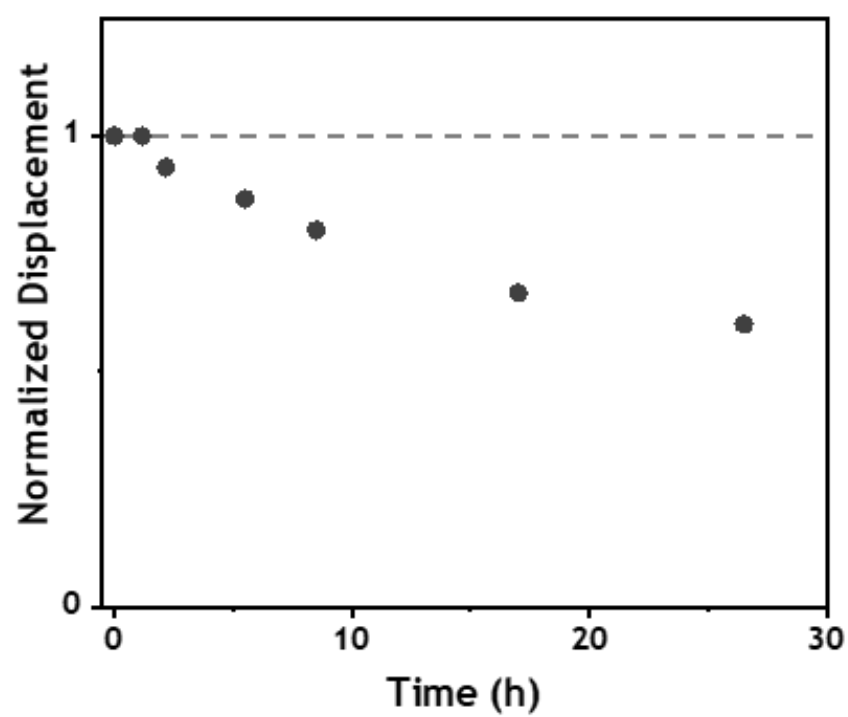
Supplementary Fig. 15. Bending displacement according to voltage. a. 0.01 V. b. 0.10 V. c. 0.40 V. d. 0.70 V. e. 1.00 V. f. From 0.01 to 1.00 V.



Supplementary Fig. 16. Durability A-membrane actuator at 0.3 V and 3.0 Hz. After a. 2 days. b. 10 days. c. 20 days. d. 40 days.



Supplementary Fig. 17. Durability of A-membrane actuator, humidity, and outdoor temperature. The temperature in the testing room was maintained between 18 and 20 °C. The humidity was between 60 and 90 %.



Supplementary Fig. 18. Durability of C-membrane actuator at 1.5 V and 3.0 Hz, normalized to the initial bending displacement.

Supplementary Table 1. Solvent systems for preparing Nafion dispersions.

Solvent systems	Solvents [g]				Nafion [g]
	water	1-propanol	DMAc	EMImBF ₄	
A-membrane	18.00	0.32	1.50	0.22	0.14
C- membrane	0.00	0.00	20.00	0.12	0.20

Supplementary Table 2. Hansen solubility parameters and distance parameters of Nafion and solvents.

Materials	Hansen solubility parameters [MPa ^{1/2}]				Distance parameter, R_a [MPa ^{1/2}]
	δ_t	δ_d	δ_p	δ_h	
Nafion ¹⁻³	23.5	17.4	12.5	9.6	$2.69 < R_0 < 20.09$
A-membrane ^{a)}	45.02	15.58	15.39	38.95	29.72
C-membrane	22.82	16.81	11.52	10.21	1.66
Water ²	47.8	15.6	16.0	42.3	33.08
1-propanol ²	24.5	16.0	6.8	17.4	10.06
2-propanol ²	23.5	15.8	6.1	16.4	9.87
EMImBF ₄ ⁴	26.2	17.9	14.8	12.2	3.61
DMAc ²	22.8	16.8	11.5	10.2	1.67
NMP ^{2,3}	22.9	18.0	12.3	7.2	2.69
Water/2-propanol ^{2,3}	35.7	15.7	11.1	29.6	20.09

^{a)}this study

Supplementary Table 3. Rise time and durability of the developed actuator and in the literature.

Electro-active ionic actuators			Durability (cycle, frequency) [day]	Rise time (voltage) [s]
Electrolytes	Electrodes	Thickness [μm]		
Nafion/EMIImBF ₄ ^{a)}	PEDOT:PSS	100	42 (>10 ⁷ , 3 Hz)	0.9 (0.3 V)
0.5M H ₂ SO ₄ solution ⁵	MoS ₂ /Kapton	26	3.0 (8000, 0.03 Hz)	200 (0.6 V)
COF-DT-SO ₃ Na ⁶	PEDOT:PSS	60	2.72 (23490, 0.1 Hz)	1 (0.5 V)
g-CN/PVDF/EMIImBF ₄ ⁷	g-CN	85	1.15 (100000, 1 Hz)	200 (3.0 V)
PMMA gel/LiClO ₄ ⁸	polypyrrole	220	0.46 (120000, 3 Hz)	>10 (1.0 V)
Nafion/EMIImBF ₄ ⁹	Ni-MOF/PEDOT:PSS	130	0.35 (30000, 1 Hz)	1.5 (1.0 V)
PSS-b-PMB/HMIImPF ₆ ¹⁰	CNT	70	0.31 (13490, 0.5 Hz)	80 (3.0 V)
PVDF-co-HFP porous/EMIImBF ₄ ¹¹	PEDOT:PSS	140	0.23 (20000, 1 Hz)	2.5 (0.5 V)
Nafion/EMIImBF ₄ ¹²	NSCOF/PEDOT:PSS	110	0.23 (20000, 1 Hz)	2.3 (0.5 V)
Nafion/EMIImBF ₄ ¹³	Triazine-PIM-1 CTF/ PEDOT:PSS	80-115	0.17 (15000, 1 Hz)	10 (0.5 V)
PS-b-PSS-EMIIm/EMIImBF ₄ ¹⁴	N,S co-doped graphene/PEDOT:PSS	100	0.16 (14000, 1 Hz)	5 (0.5 V)
Nafion/EMIImBF ₄ ¹⁵	MXene/ PEDOT:PSS	110	0.21 (18000, 1 Hz)	1 (0.5 V)
	PEDOT:PSS	110	0.15 (12600, 1 Hz)	14 (0.5 V)
1M LiCl or NaCl solution ¹⁶	V ₂ O ₅ nanofiber	11	-	>100 (1.0 V)
1M NaCl solution ¹⁷	CNT/PVC/CNT	245	-	>15 (1.0 V)
Other actuators				
Names		Materials		
Fuel-power artificial muscles ¹⁸		NiTi SMA/Pt	-	45
		CNT/Pt/Nafion	-	1800
Polymer composite humid actuator ¹⁹		Polypyrrole	-	300
Photomechanical actuator ²⁰		CNT/PDMS	-	10
Liquid crystal photoactuator ²¹		Liquid crystal polymer	-	2400
CNT/thermalplastic actuator ²²		CNT/Morthane	-	5
Liquid-crystal elastomer (LCE) thermal actuator ²³		LCE	-	360
Liquid-crystal elastomer (LCE) photo actuator ²⁴		LCE	-	1.3
Dielectric elastomer, applied voltage 4 - 6 kV ²⁵		Acrylic, silicone	-	0.001
Shape memory alloys (SMA) actuator ²⁶		SMA	-	5
CNT yarn muscles ²⁷		CNT	1.54 (>2x10 ⁶ , 15 Hz)	-
Fishing line and sewing thread muscles ²⁸		CNT/nylon	13.89 (>1.2x10 ⁶ , 1 Hz)	-

^{a)}this study

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