Supplementary Information (SI) for Journal of Materials Chemistry C.

Electronic supplementary information

Highly-conductive ion channels enable flexible actuators for robotic application

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Experimental section

Materials

Polyvinylidene difluoride (PVDF) was purchased from Xiya Chemical Technology Company. Ionic liquid 1-Ethyl-3-methylimidazolium tetrafluoroborate ([EMIM][BF_4]) was obtained from Shanghai Cheng Jie Chemical Company. PEDOT:PSS (1.3 wt.%±0.2%) was commercially available from Xin sheng New Energy Company. Hydroxylated multi-walled carbon nanotubes (MWCNTs) was purchased from Shanghai Macklin Biochemical Company. Poly(acrylic acid) (PAA, M. W~5000, 50 wt.% aqueous solution) was purchased from Shanghai Acmec Biochemical Company. sodium dodecyl benzene sulfonate was obtained from BKMAMLAB Company. N, N-dimethylacetamide (DMAC) was available from Energy Chemical Company. All the reagents were of analytical pure and used as received and deionized water was prepared for all experiments.

Preparation of PVDF coated PAA and MWCNTs (CNT@PVDF/PAA)

Firstly, 0.6 g PVDF and 25 mg MWCNTs were dissolved in 30 mL deionized water, and then 0.6 g sodium dodecyl benzene sulfonate was added and heated to form colloids (Figure S1a). Turned off the heat and continue stirring until the liquid cools to room temperature. Then the liquid is dripped on the glass plate (2.5 cm×7.5 cm), heated at 80°C for 4 h until the liquid was completely dried, and the powder sample CNT@PVDF/PAA is obtained by scraping it off with a knife (Figure S1b).

Preparation of the electrolyte and electrode layers

Firstly, 0.25 g CNT@PVDF/PAA powder was dispersed in 2 mL DMAC and stirred for 30 min, then 0.1 g [EMIM][BF_4] was added and stirred for 12 h to obtain a uniform solution (Figure S1c). After 1 h ultrasonic dispersion, it was injected onto the glass plate and heated at 80°C to become a soft black electrolyte film. Finally, the black PEDOT: PSS liquid was poured on an 8 cm×8 cm×1 cm polytetrafluoroethylene mold and heated at 80 ^{o}C for 4 h to obtain a soft black electrode film.

Assembly of the ionic actuators

Two PEDOT:PSS electrode films were laminated on the CNT@PVDF/PAA electrolyte film, hot pressed at 150 ^{o}C for 10 min, and cooled to room temperature.

After that, the film is carefully removed from the glass substrate to obtain an independent sandwich film. The actuator membrane was then cut with a knife into strips of standard size (5 mm×25 mm). The original PVDF electrolyte actuator was constructed using the same process.

The cross section and electrolyte surface of the actuator were observed by scanning electron microscope (SEM). The model of SEM was Hitachi SU8010. The actuation performance and electrochemical performance of the actuator were tested by CHI660 electrochemical workstation. A multifunctional digital four-probe tester (ST2258C) was used to measure the conductivity of the electrolyte. The mechanical properties of electrolyte film and actuator were tested by universal testing machine (ZwickRoell Z0.5TH). The Fourier transform infrared spectrum of the electrolyte was obtained by Bruker VERTEX 70 shock spectrometer. X-ray diffraction (XRD) was recorded at room temperature under 50 kV and 50 mA Cu K_a radiation using the X'Pert-Pro -Pro MPD X-ray diffractometer from the Netherlands.

Supporting Figures

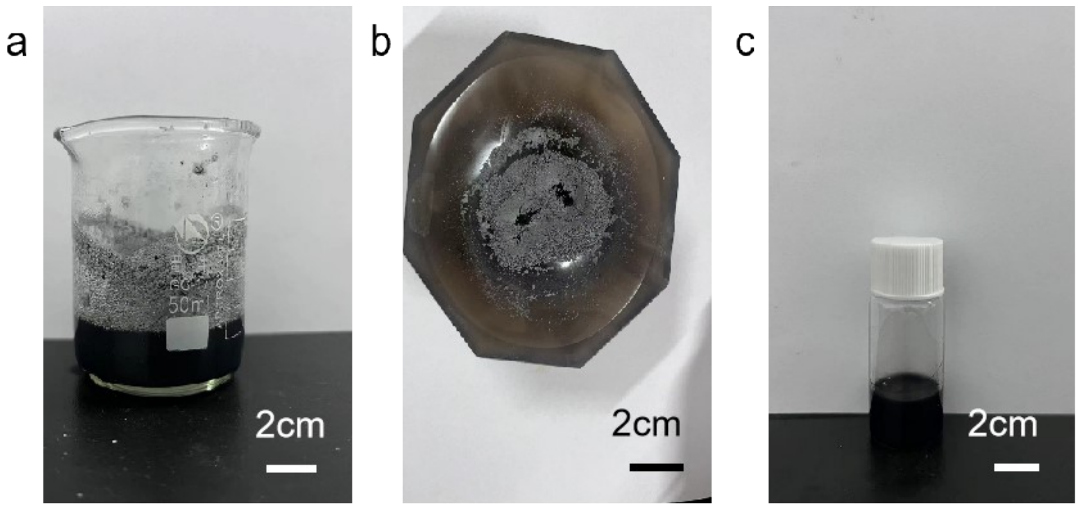

Figure S1. Photographs of electrolyte preparation. (a) CNT@PVDF/PAA dispersion. (b) CNT@PVDF/PAA powder. c) The homogeneous CNT@PVDF/PAA dispersion in organic solvent.

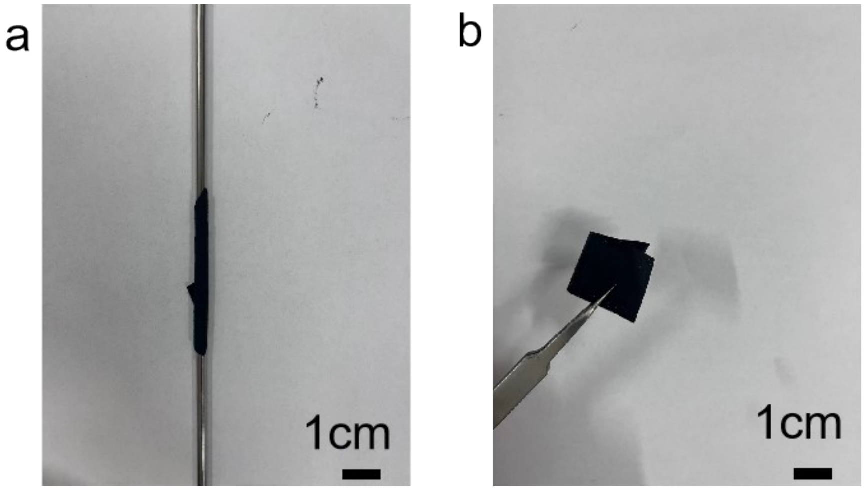

Figure S2. The electrolyte membrane is (a) twisted on a rod and (b) fold by a tweezer.

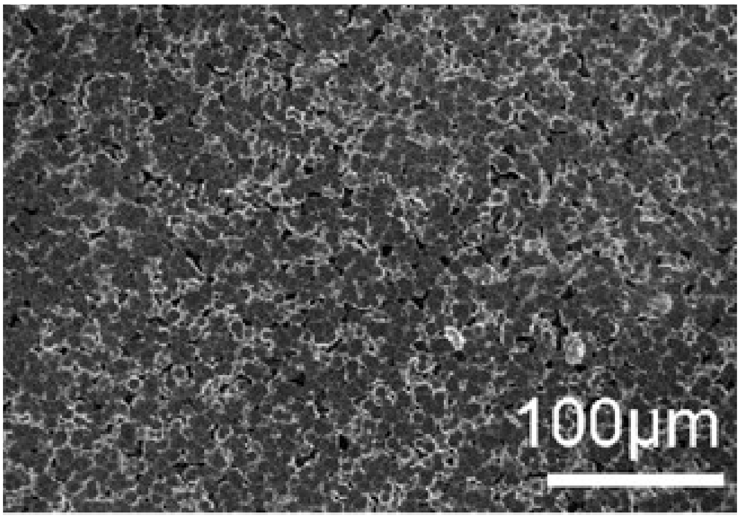

Figure S3. The surface SEM image of PVDF electrolyte membrane.

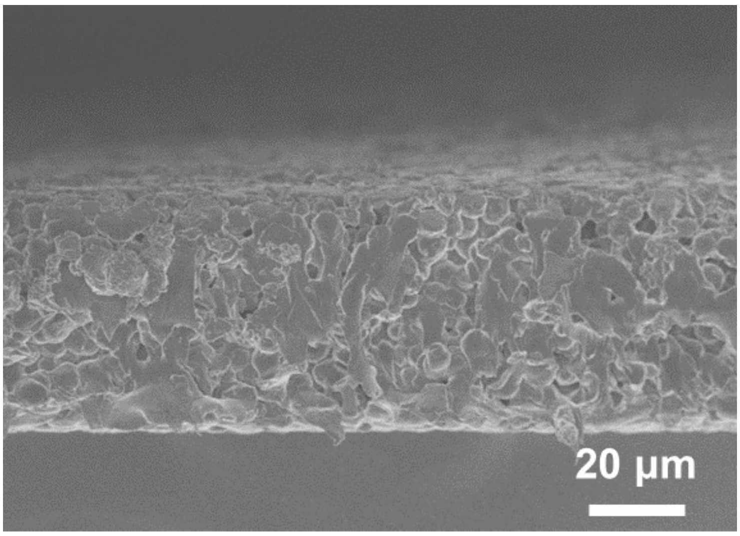

Figure S4. The cross-sectional SEM image of PVDF electrolyte membrane.

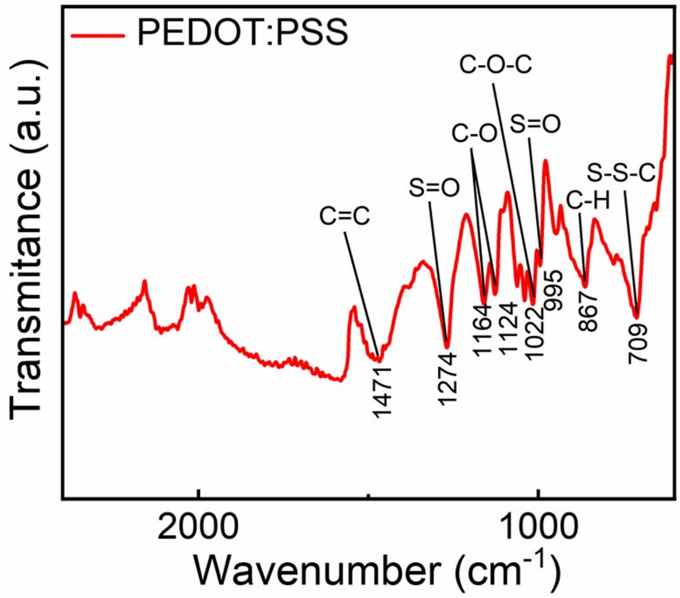

Figure S5. FTIR spectrum of PEDOT:PSS electrode.

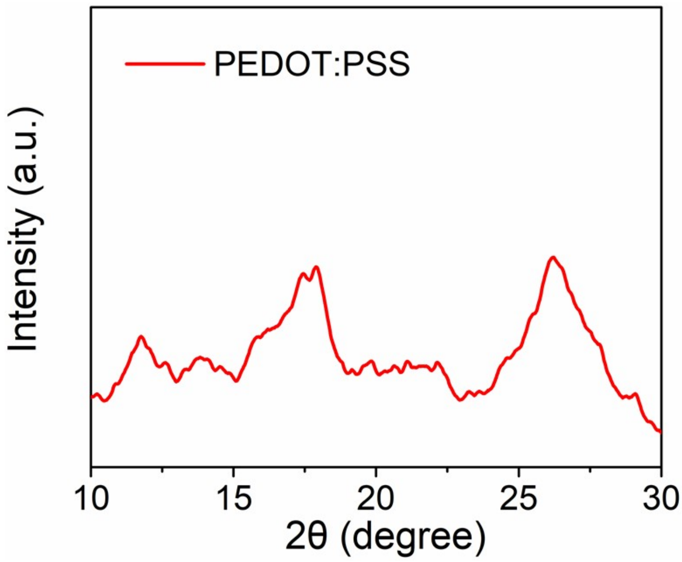

Figure S6. XRD pattern of PEDOT:PSS electrode.

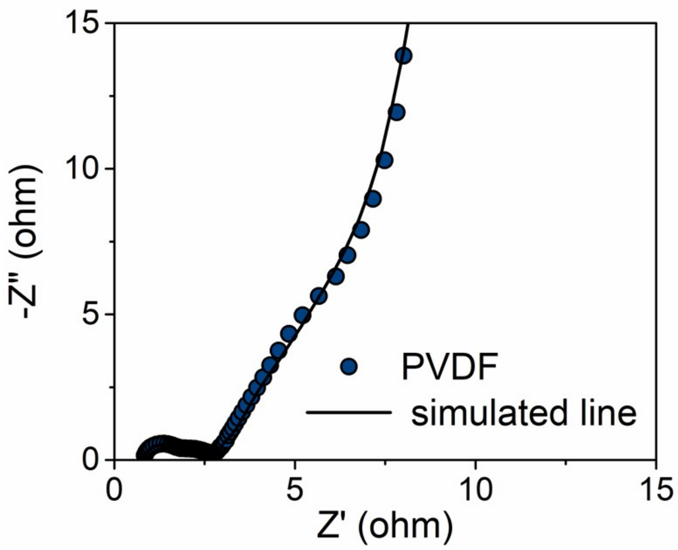

Figure S7. The EIS plot of PVDF based actuator.

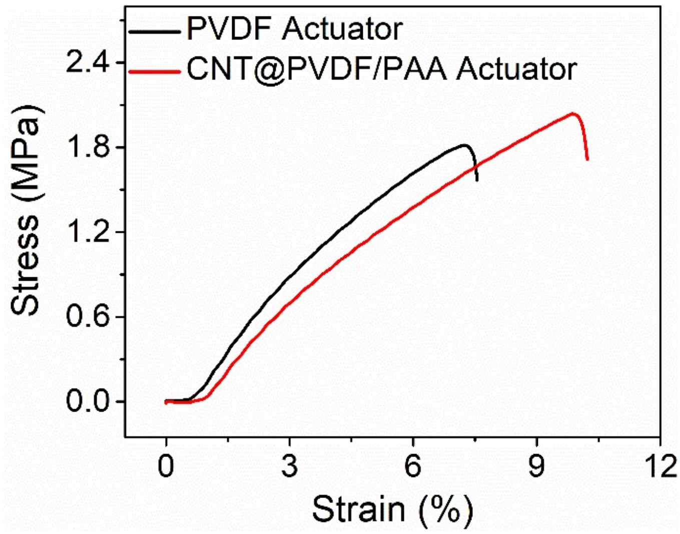

Figure S8. Strain-Stress curves for PVDF and CNT@PVDF/PAA based actuators.

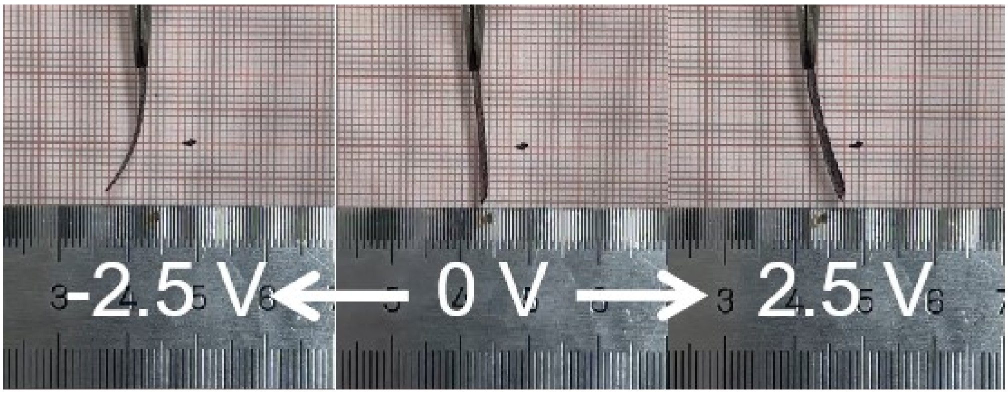

Figure S9. The actuation of PVDF electrolyte based actuator at voltage of 2.5V.

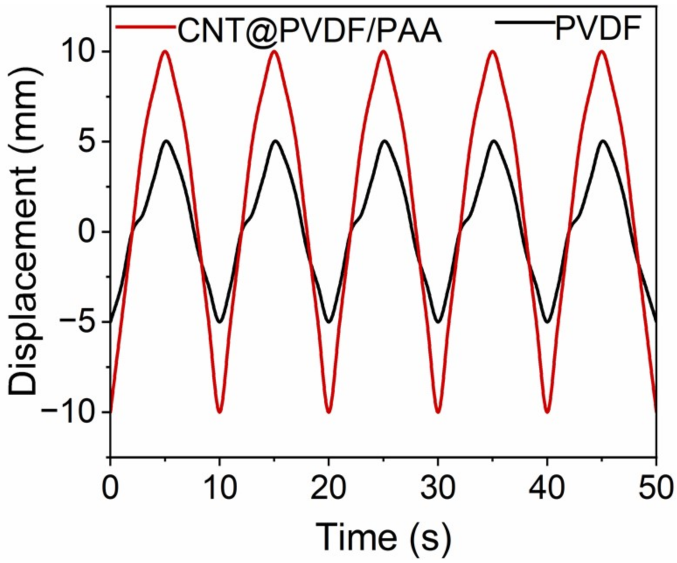

Figure S10. Displacement comparison of PVDF and CNT@PVDF/PAA actuators under the frequency of 0.1 Hz and voltage of 2.5 V.

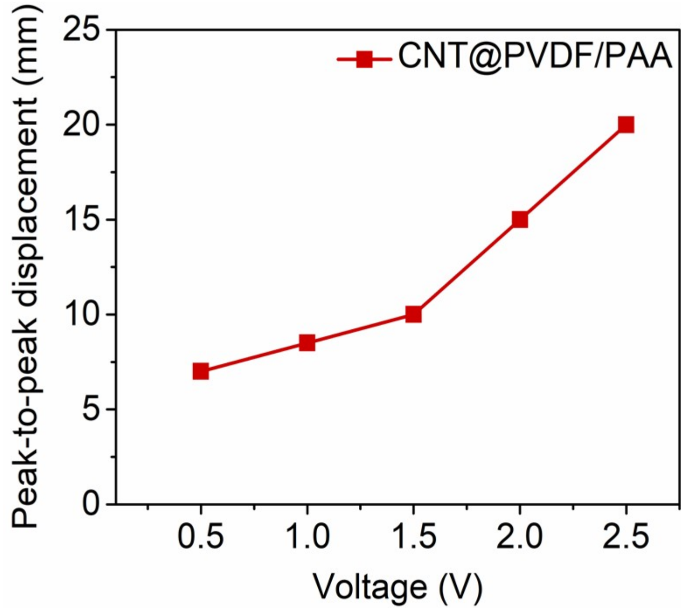

Figure S11. Displacement variation of CNT@PVDF/PAA actuator at different voltages.

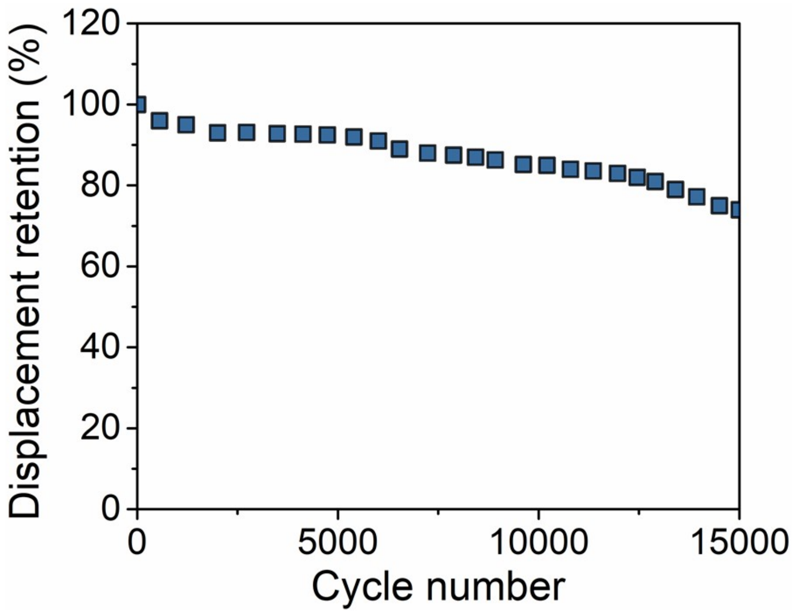

Figure S12. Cycling performance of PVDF based actuator.

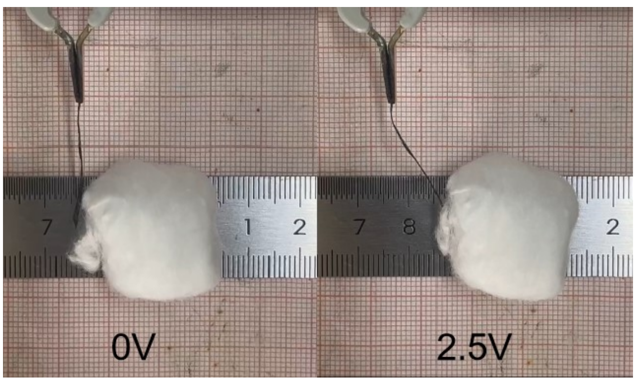

Figure S13. The blocking force test of the actuator.

Table S1. Simulated results of EIS plots of PVDF and CNT@PVDF/PAA actuators.

Element	PVDF	CNT@PVDF/PAA
R1	0.8118	0.8123
R2	0.7001	0.5305
R3	1.246	0.7681
R4	**157.8**	**1.501**
CPE1	1.104	1.095
CPE2	0.6476	0.5728
CPE3	0.7582	0.7808
W	**1.375**	**0.0391**

Table S2. Mechanical properties of electrolytes and actuators.

Material/Device	Tensile strength (MPa)	Elongation (%)
PVDF	0.6	7.6
CNT@PVDF/PAA	1.5	41.2
PVDF Actuators	1.5	7.5
CNT@PVDF/PAA Actuators	2.1	9.9