

Supplementary Information

Long-life Aqueous Zinc-iodine Flow Batteries Enabled by Selectively Intercepting Hydrated Ions

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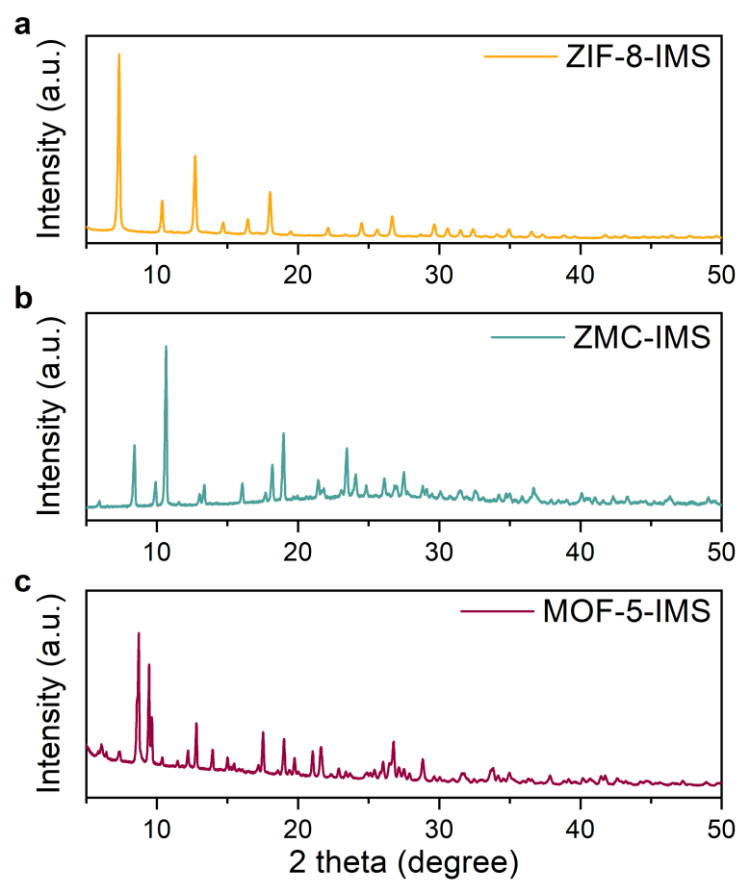
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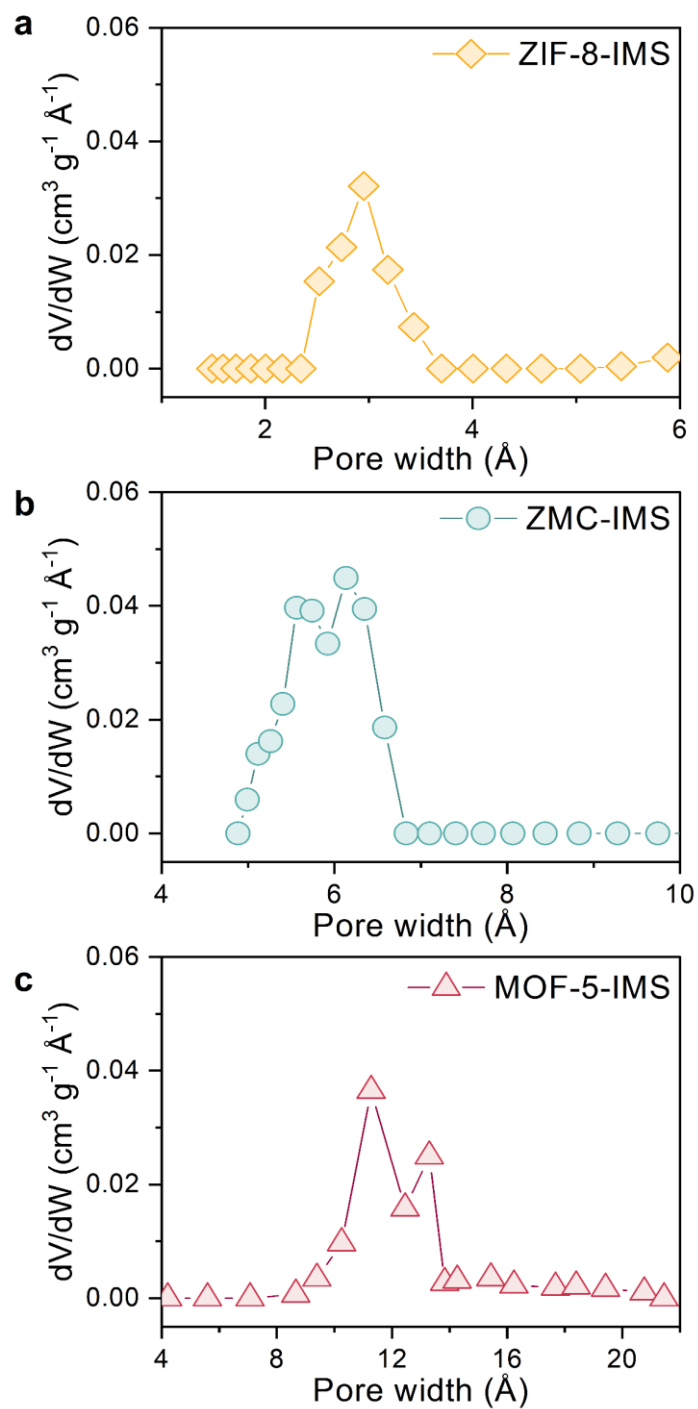
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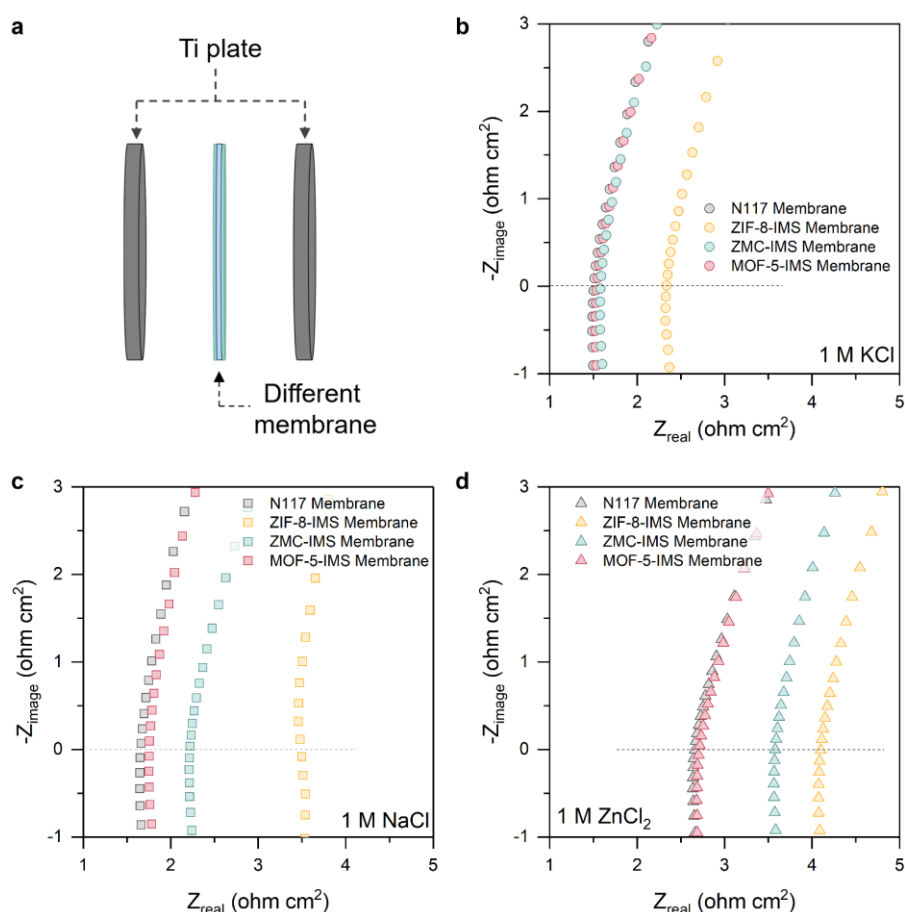
Supplementary Fig. 1 | Crystal structures of different IMS. XRD patterns of (a) ZIF-8-IMS, (b) ZMC-IMS, and (c) MOF-5-IMS.



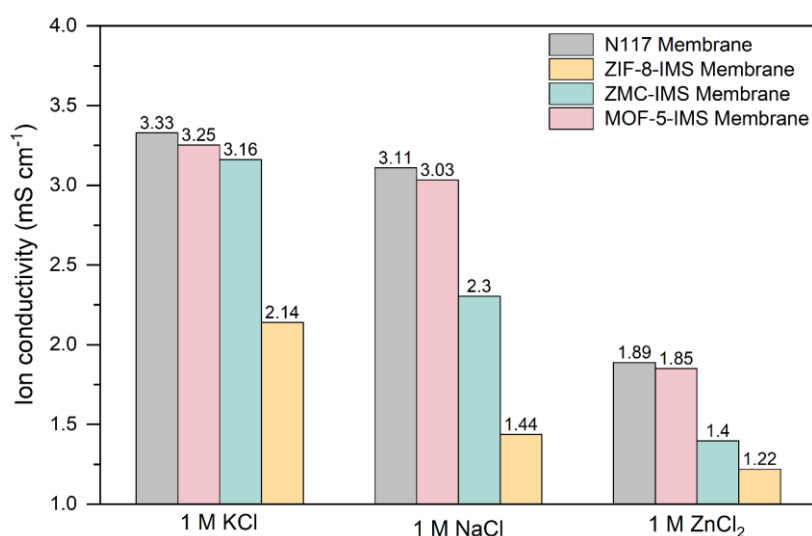
Supplementary Fig. 2 | Pore size analysis of different IMS. Pore size distributions of (a) ZIF-8-IMS, (b) ZMC-IMS, and (c) MOF-5-IMS.



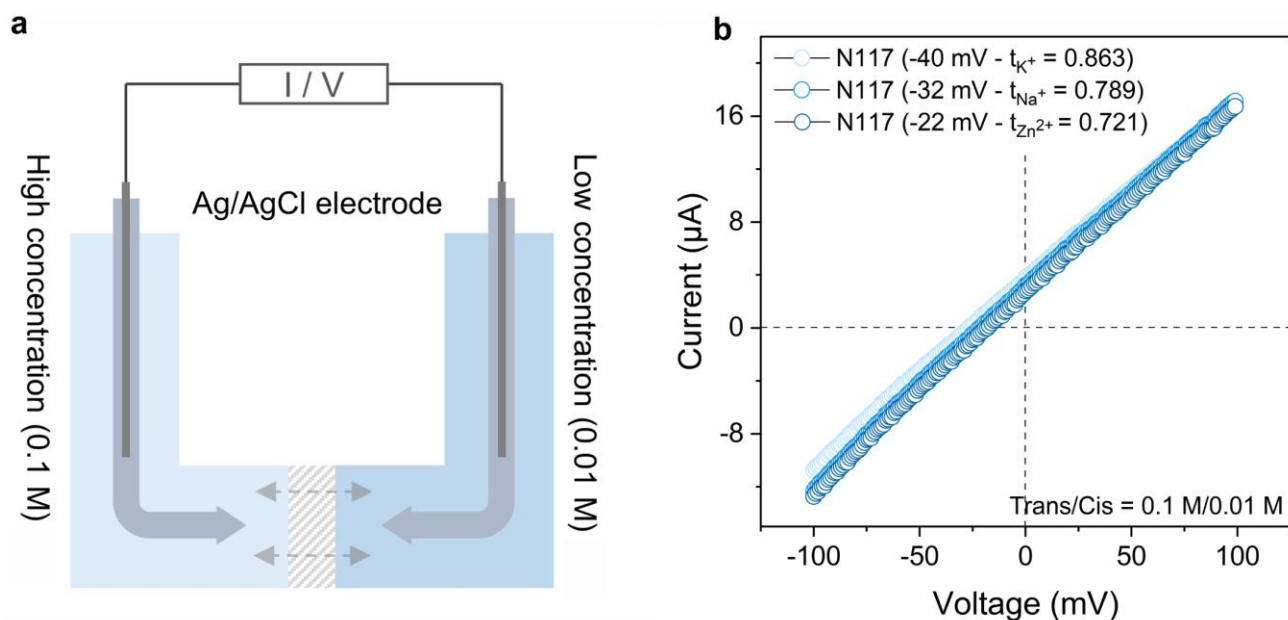
Supplementary Fig. 3 | Membrane preparation. Schematic illustration and digital images of the membranes prepared with the slurry.



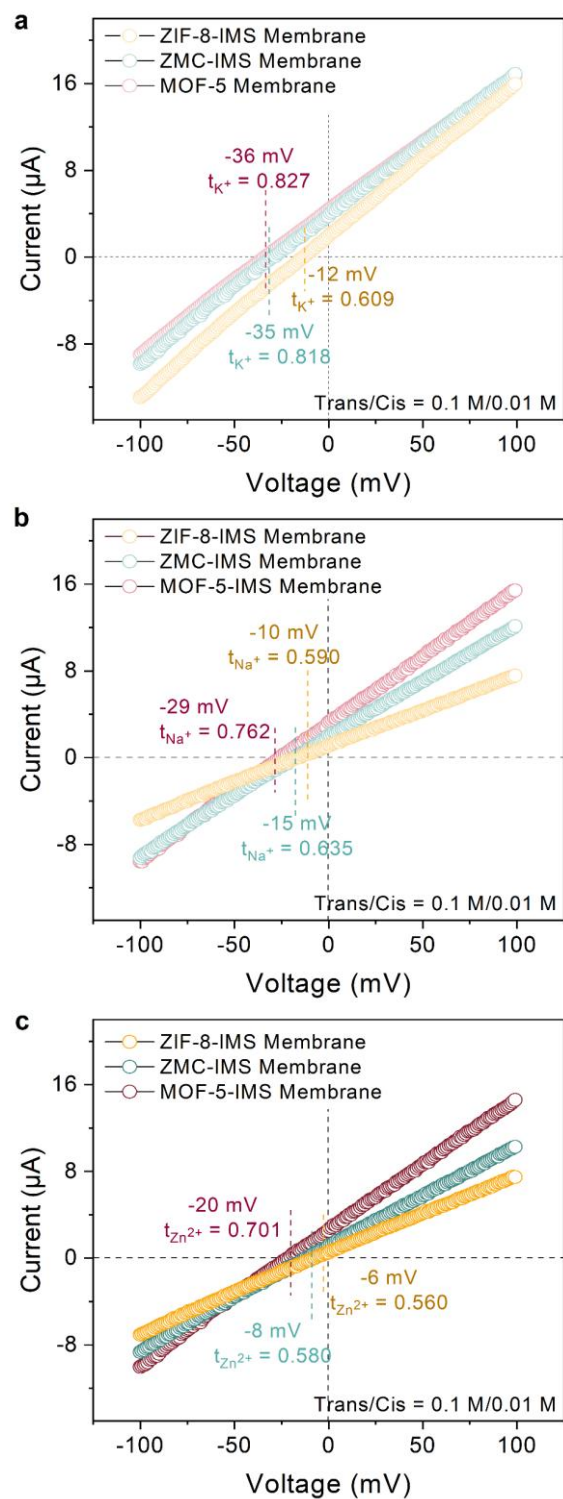
Supplementary Fig. 4 | Ion conductivity measurement of electrolyte with different salts. (a) Schematic diagram for membrane resistance measurement. (b), (c) & (d) Nyquist plots of different membranes in 1 M KCl, 1 M NaCl, and 1 M ZnCl_2 .



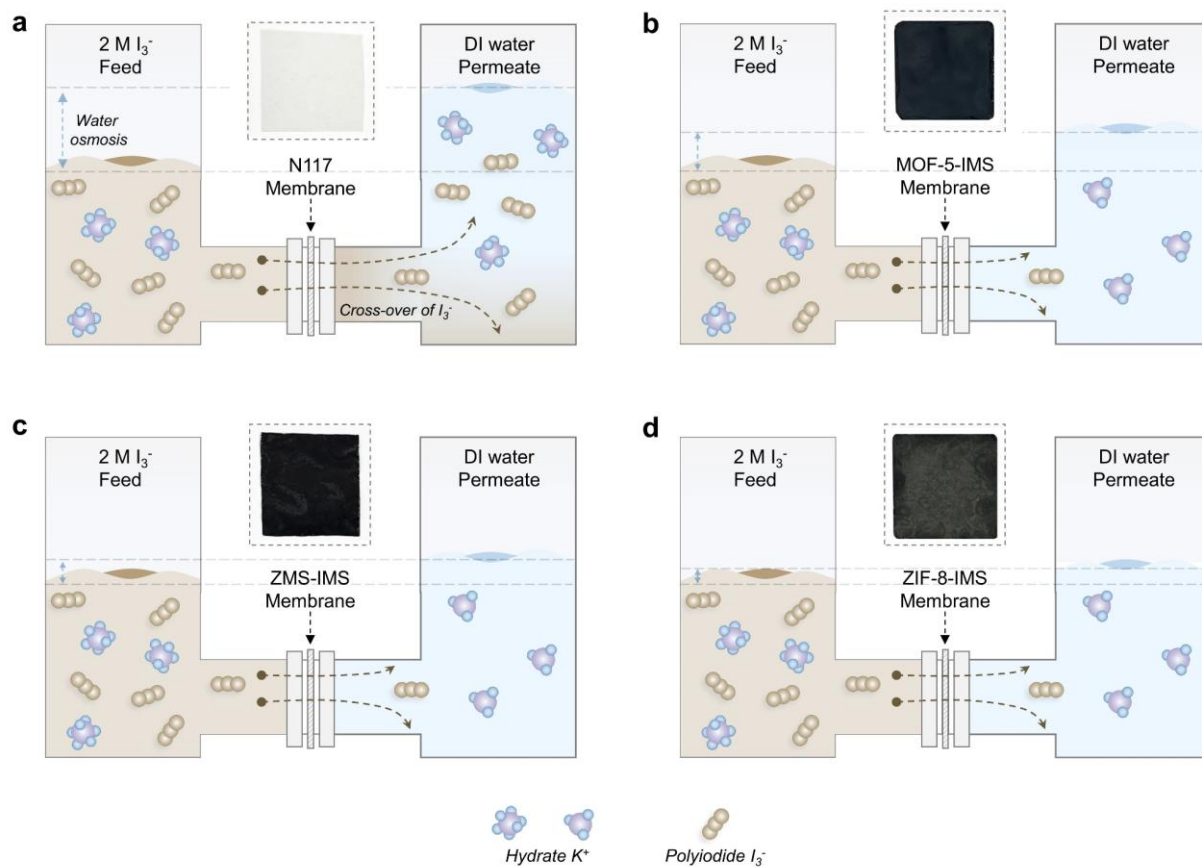
Supplementary Fig. 5 | Ion conductivity results of electrolyte with different salts. Ion conductivity of different membranes in 1 M KCl, 1 M NaCl, and 1 M ZnCl₂.



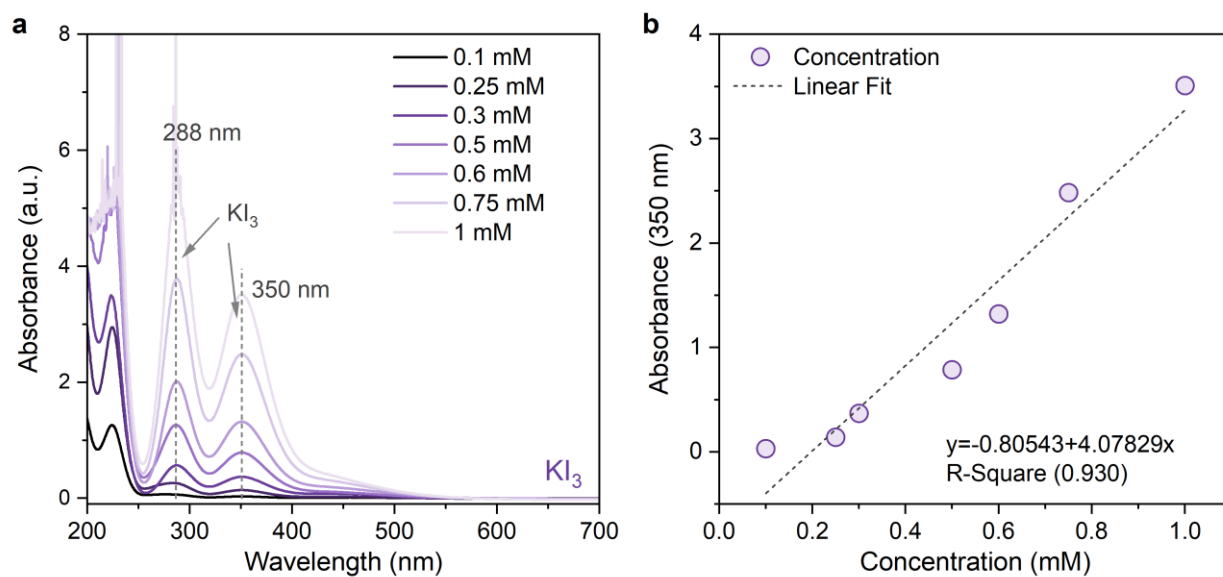
Supplementary Fig. 6 | Ion transference number measurement. (a) Schematic illustration of V-I testing device; (b) the cation transference numbers of K⁺, Na⁺, Zn²⁺. The transference numbers calculated from V-I profiles for 0.1 M/0.01 M concentration gradients in the two reservoirs connected by an N117 membrane.



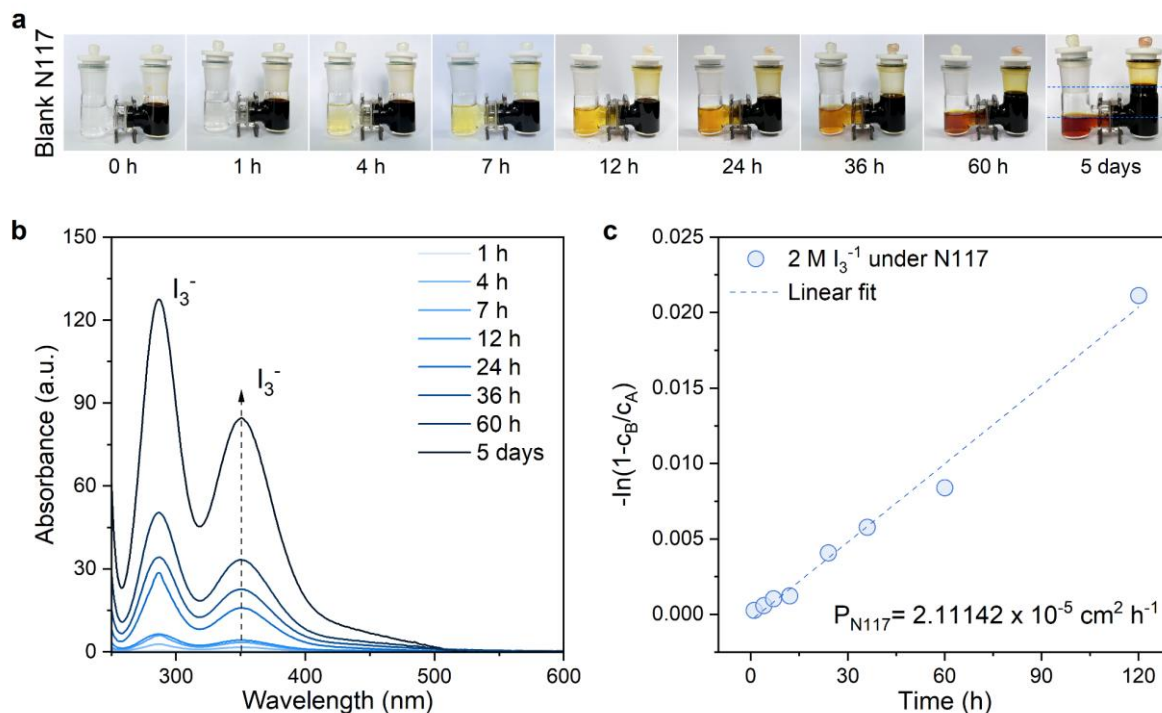
Supplementary Fig. 7 | Ion transference number of different IMS membranes. The cation transference numbers of (a) K^+ , (b) Na^+ , (c) Zn^{2+} under ZIF-8-IMS membranes, ZMC-IMS membranes and MOF-5-IMS membranes. The transference numbers calculated from V-I profiles for 0.1 M/0.01 M concentration gradients in the two reservoirs.



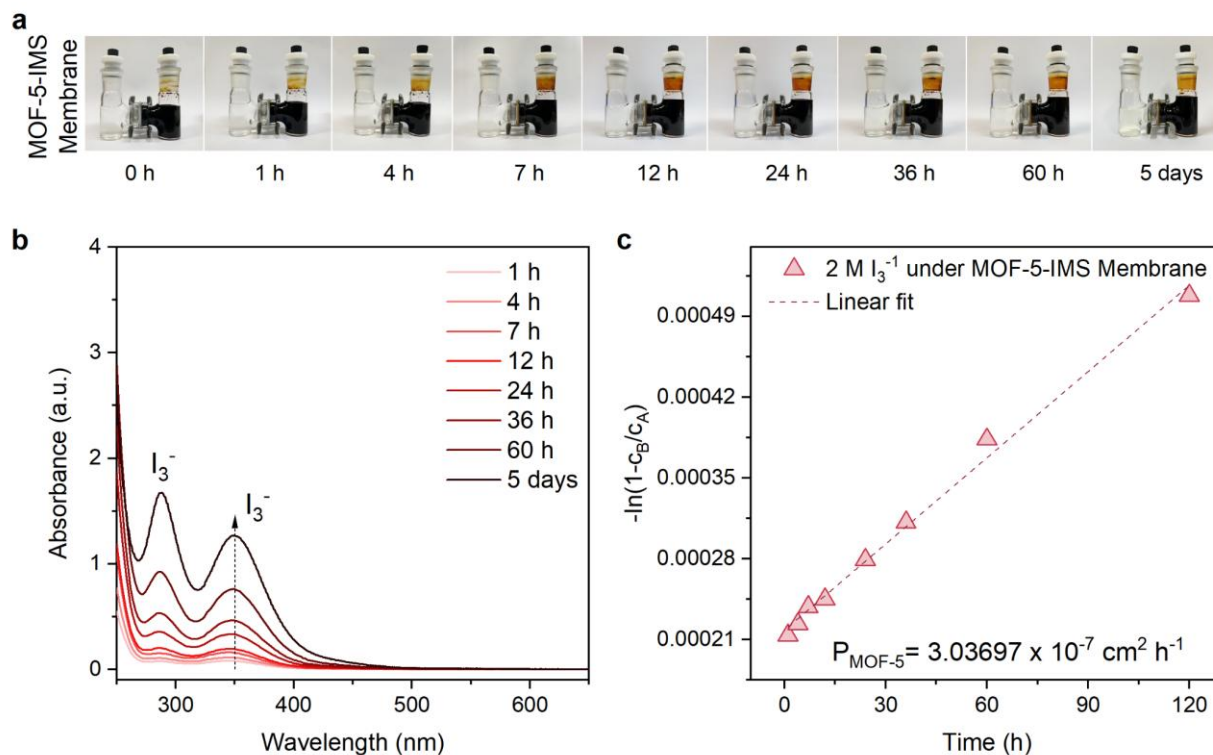
Supplementary Fig. 8 | Ionic permeation measurements. Two-compartment H-cell testing configuration of 2 M KI_x permeation using (a) N117 membranes, (b) MOF-5-IMS membranes, (c) ZMC-IMS membranes, and (d) ZIF-8-IMS membranes.



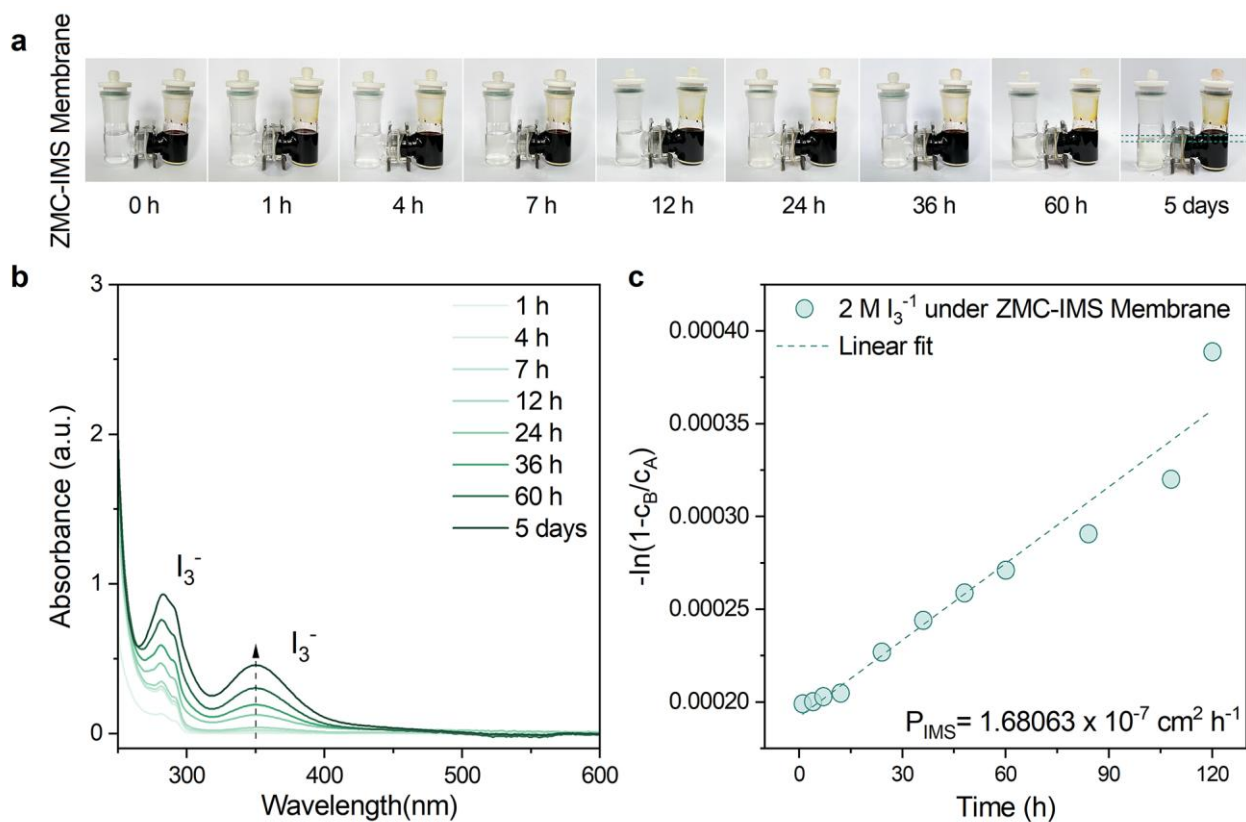
Supplementary Fig. 9 | Standard UV-visible spectra of aqueous KI_3 electrolytes. (a) UV-visible spectra of KI_3 under multiple concentrations between 0.1 mM to 1 mM; (b) Beer's law plot for I_3 multiple concentrations between 0.1 mM to 1 mM.



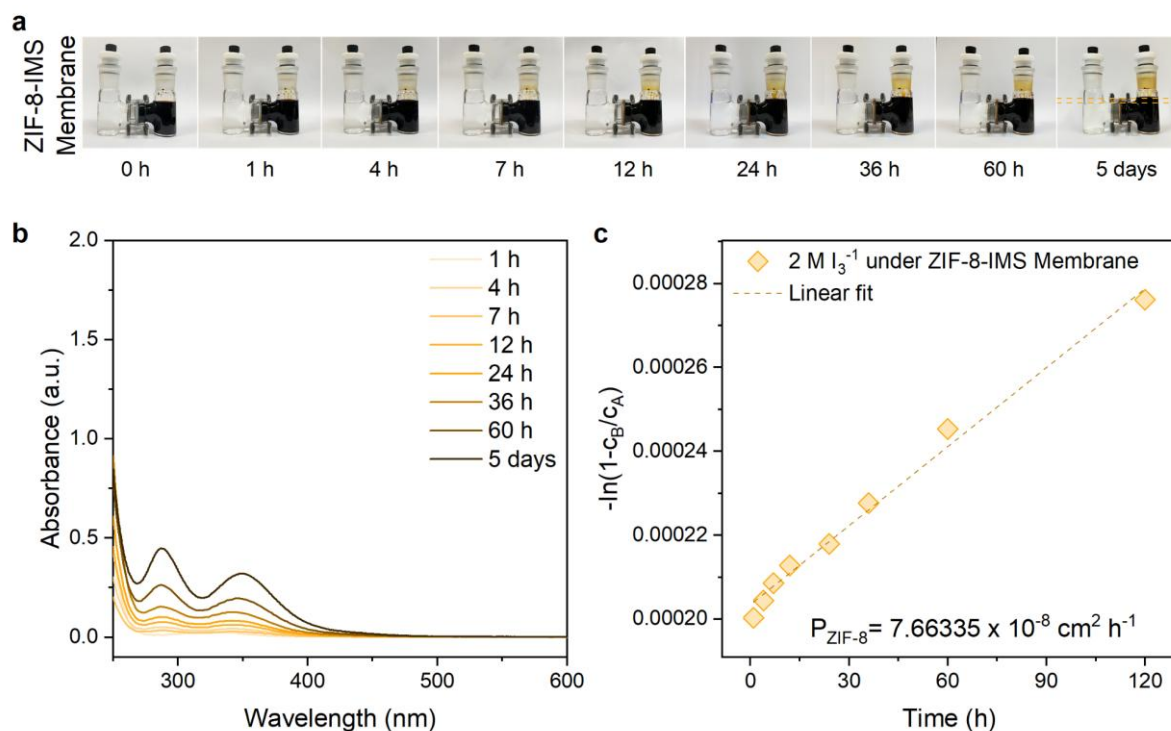
Supplementary Fig. 10 | KI_x permeation under N117 membranes. (a) photographs of the 2 M KI_x permeate solutions under the N117 membrane. (b) UV-vis of the KI_x permeate side. (c) $-\ln(1-c_B/c_A)$ vs. permeation time for the determination of permeability of 2 M KI_x under the N117 membrane. The fits in the $-\ln(1-c_B/c_A)$ vs. t (time) plots in (c) were obtained by linear fitting.



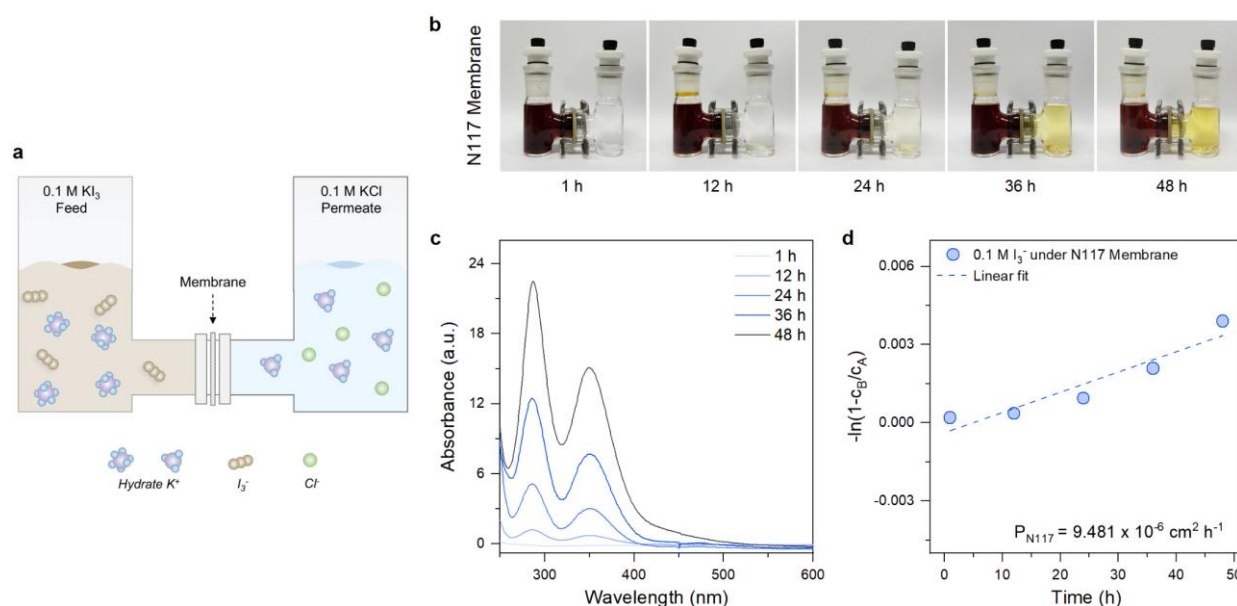
Supplementary Fig. 11 | KI_x permeation under MOF-5-IMS membranes. (a) photographs of the 2 M KI_x permeate solutions under the MOF-5-IMS membrane. (b) UV-vis of the KI_x permeate side. (c) $-\ln(1-c_B/c_A)$ vs. permeation time for the determination of permeability of 2 M KI_x under the MOF-5-IMS membrane. The fits in the $-\ln(1-c_B/c_A)$ vs. t (time) plots in (c) were obtained by linear fitting.



Supplementary Fig. 12 | KI_x permeation under ZMC-IMS membranes. (a) photographs of the 2 M KI_x permeate solutions under the ZMC-IMS membrane. (b) UV-vis of the KI_x permeate side. (c) $-\ln(1-c_B/c_A)$ vs. permeation time for the determination of permeability of 2 M KI_x under the ZMC-IMS membrane. The fits in the $-\ln(1-c_B/c_A)$ vs. t (time) plots in (c) were obtained by linear fitting.

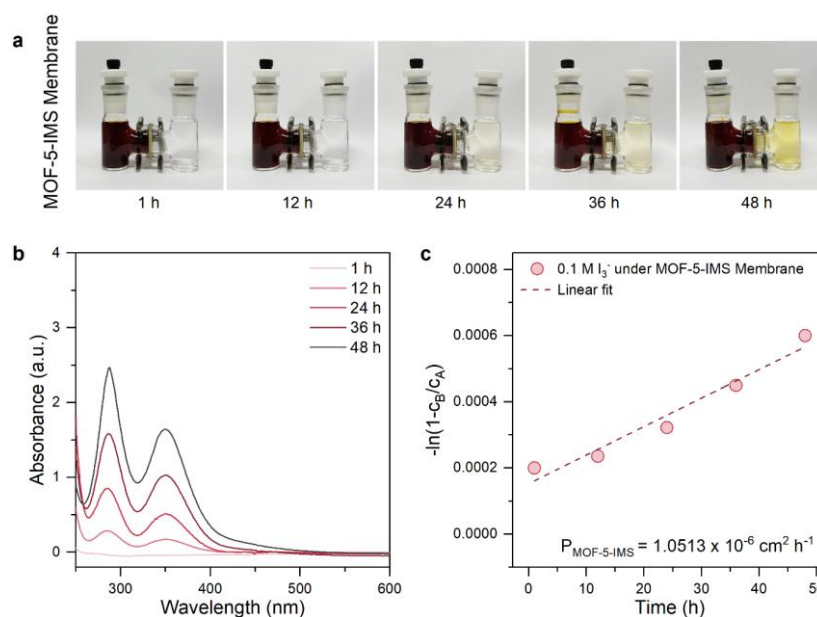


Supplementary Fig. 13 | KI_x permeation under ZIF-8-IMS membranes. (a) photographs of the 2 M KI_x permeate solutions under the ZIF-8-IMS membrane. (b) UV-vis of the KI_x permeate side. (c) $-\ln(1-c_B/c_A)$ vs. permeation time for the determination of permeability of 2 M KI_x under the ZIF-8-IMS membrane. The fits in the $-\ln(1-c_B/c_A)$ vs. t (time) plots in (c) were obtained by linear fitting.

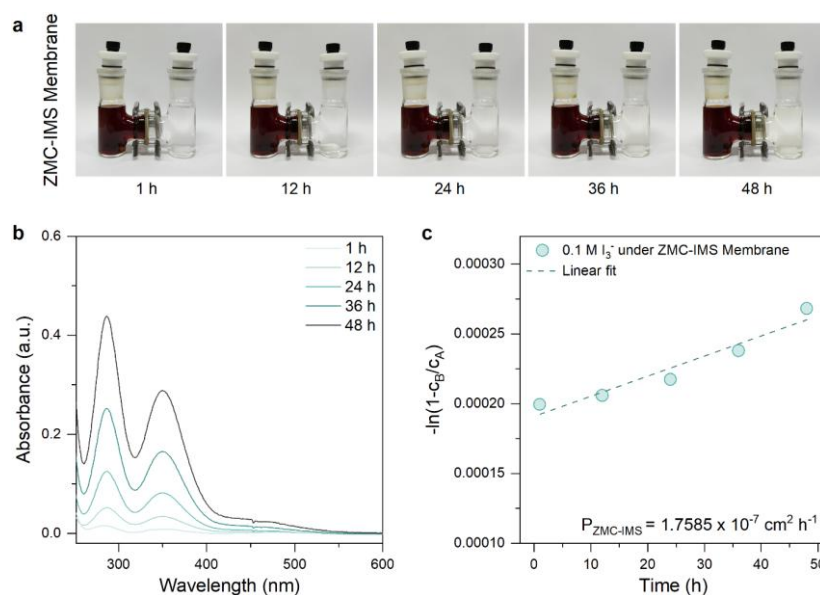


Supplementary Fig. 14 | KI_x permeation under N117 membranes based on low osmotic pressure.

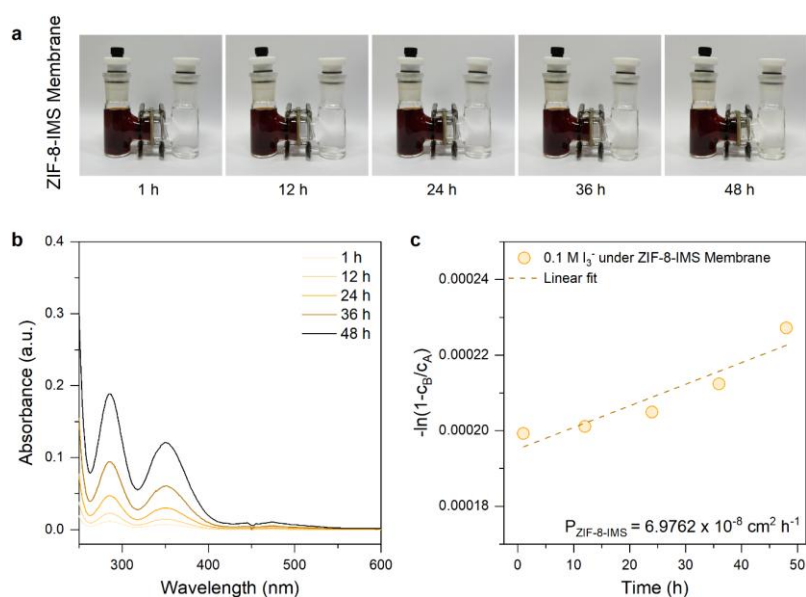
(a) Two-compartment H-cell testing configuration of 0.1 M KI_x feed side and 0.1 M KCl permeate side. (b) Photographs of the 0.1 M KI_x permeate solutions under the N117 membrane. (c) UV-vis of the KI_x permeate side. (d) $-\ln(1-c_B/c_A)$ vs. permeation time for determining permeability of 0.1 M KI_x under the N117 membrane. The fits in the $-\ln(1-c_B/c_A)$ vs. t (time) plots in (d) were obtained by linear fitting.



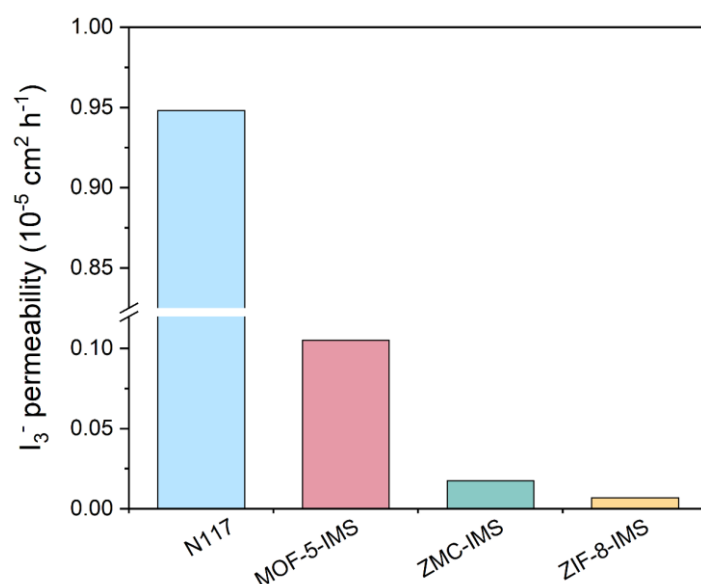
Supplementary Fig. 15 | KI_x permeation under MOF-5-IMS membranes based on low osmotic pressure. (a) Photographs of the 0.1 M KI_x permeate solutions under the MOF-5-IMS membrane. (b) UV-vis of the KI_x permeate side. (c) $-\ln(1-c_B/c_A)$ vs. permeation time for determining permeability of 0.1 M KI_x under the MOF-5-IMS membrane. The fits in the $-\ln(1-c_B/c_A)$ vs. t (time) plots in (c) were obtained by linear fitting.



Supplementary Fig. 16 | KI_x permeation under ZMC-IMS membranes based on low osmotic pressure. (a) Photographs of the 0.1 M KI_x permeate solutions under the ZMC-IMS membrane. (b) UV-vis of the KI_x permeate side. (c) $-\ln(1-c_B/c_A)$ vs. permeation time for determining permeability of 0.1 M KI_x under the ZMC-IMS membrane. The fits in the $-\ln(1-c_B/c_A)$ vs. t (time) plots in (c) were obtained by linear fitting.

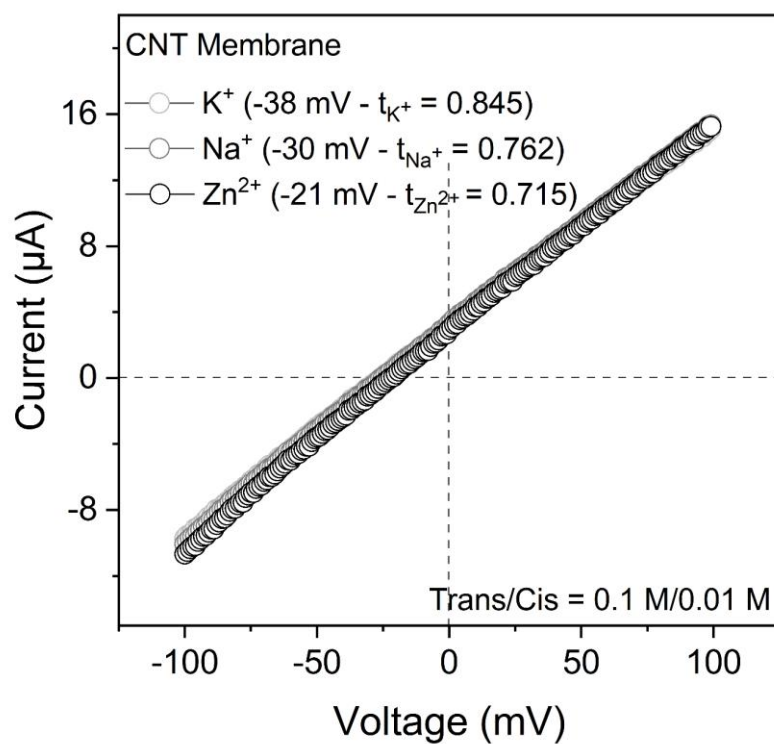


Supplementary Fig. 17 | KI_x permeation under ZIF-8-IMS membranes based on low osmotic pressure. (a) Photographs of the 0.1 M KI_x permeate solutions under the ZIF-8-IMS membrane. (b) UV-vis of the KI_x permeate side. (c) $-\ln(1-c_B/c_A)$ vs. permeation time for determining permeability of 0.1 M KI_x under the ZIF-8-IMS membrane. The fits in the $-\ln(1-c_B/c_A)$ vs. t (time) plots in (c) were obtained by linear fitting.

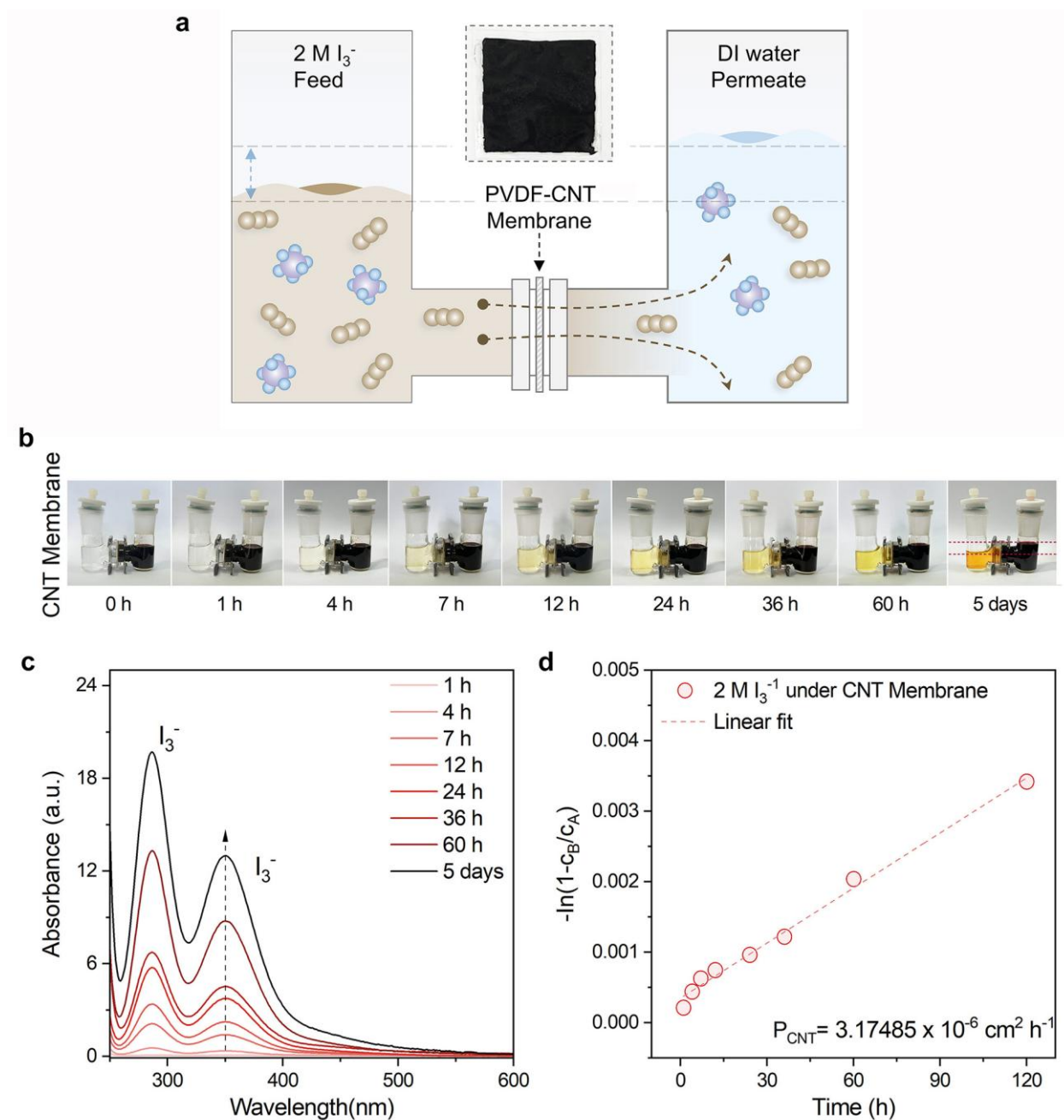


Supplementary Fig. 18 | Comparison of I_3^- permeation across different membranes. I_3^- permeability of N117, MOF-5-IMS, ZMC-IMS, and ZIF-8-IMS membranes.

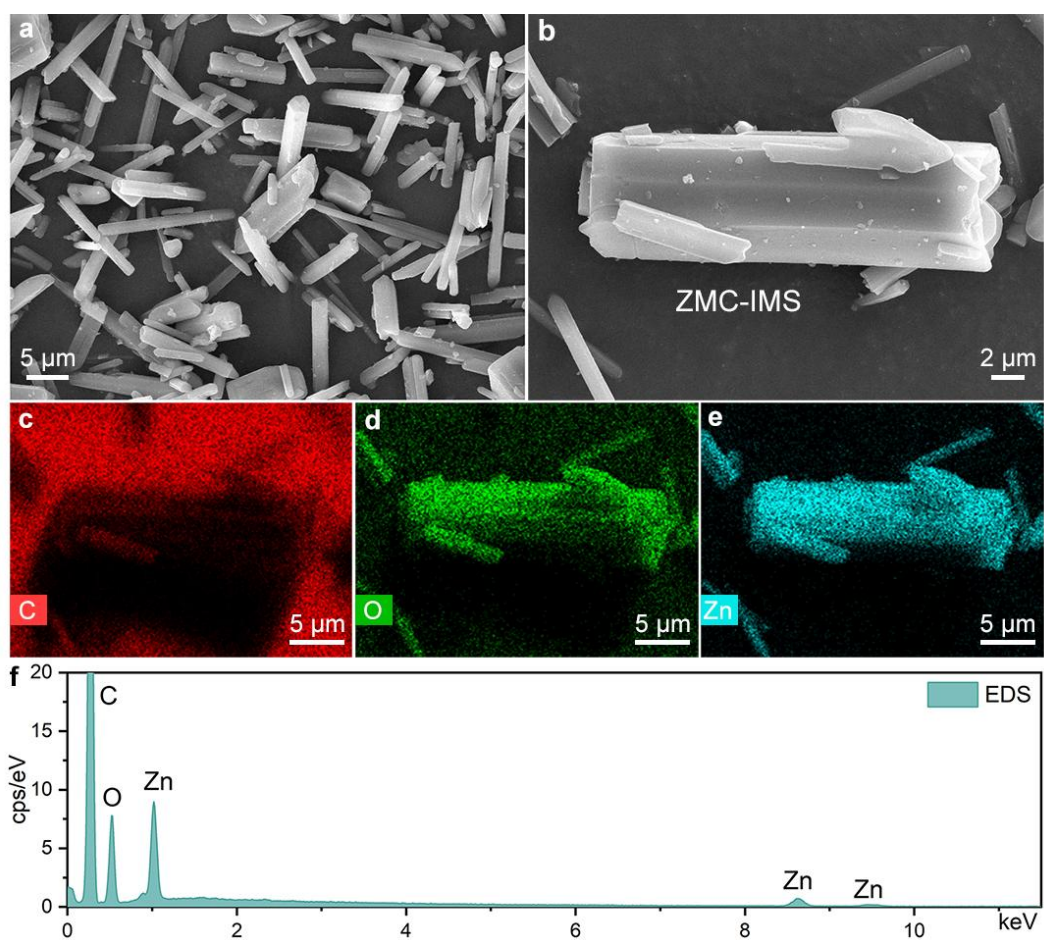
Supplementary Note: to mitigate the influence of osmotic pressure, we employed another H-type cell with a feed reservoir containing a lower concentration of KI_3 (0.1 M) and a permeate side filled with supporting salts (0.1 M KCl) to evaluate the permeability of all membranes (**Supplementary Fig. 14-17**). Notably, this testing approach is also widely utilized in other permeability assessments, which use equal concentrations of cations on the two sides to exclude the influence of cations.¹¹ As shown in **Supplementary Fig. 18**, the permeability of I_3^- through the N117 membrane exhibits a high value of $9.481 \times 10^{-6} \text{ cm}^2 \text{ h}^{-1}$, indicating poor blocking efficiency against polyiodides. When introducing ionic-molecular sieve (IMS)-based layers with different pore sizes, it is obviously observed that the MOF-5-IMS membrane ($1.0513 \times 10^{-6} \text{ cm}^2 \text{ h}^{-1}$) with large pore size ($\sim 12 \text{ \AA}$) shows only a slight improvement in blocking effectiveness for polyiodides, while both the ZMC-IMS ($1.7585 \times 10^{-7} \text{ cm}^2 \text{ h}^{-1}$) and ZIF-8-IMS ($6.9762 \times 10^{-8} \text{ cm}^2 \text{ h}^{-1}$) membranes demonstrate superior blocking performance. This suggests that regulating the ion channel size can effectively suppress crossover. Therefore, the permeation results obtained from the low osmotic pressure H-cell align with the trends observed in the previous measurements (**Fig. 2e**). This confirms that the ZMC-IMS membrane can precisely control the transport of hydrated ions to overcome both the challenges of polyiodide crossover and water migration.



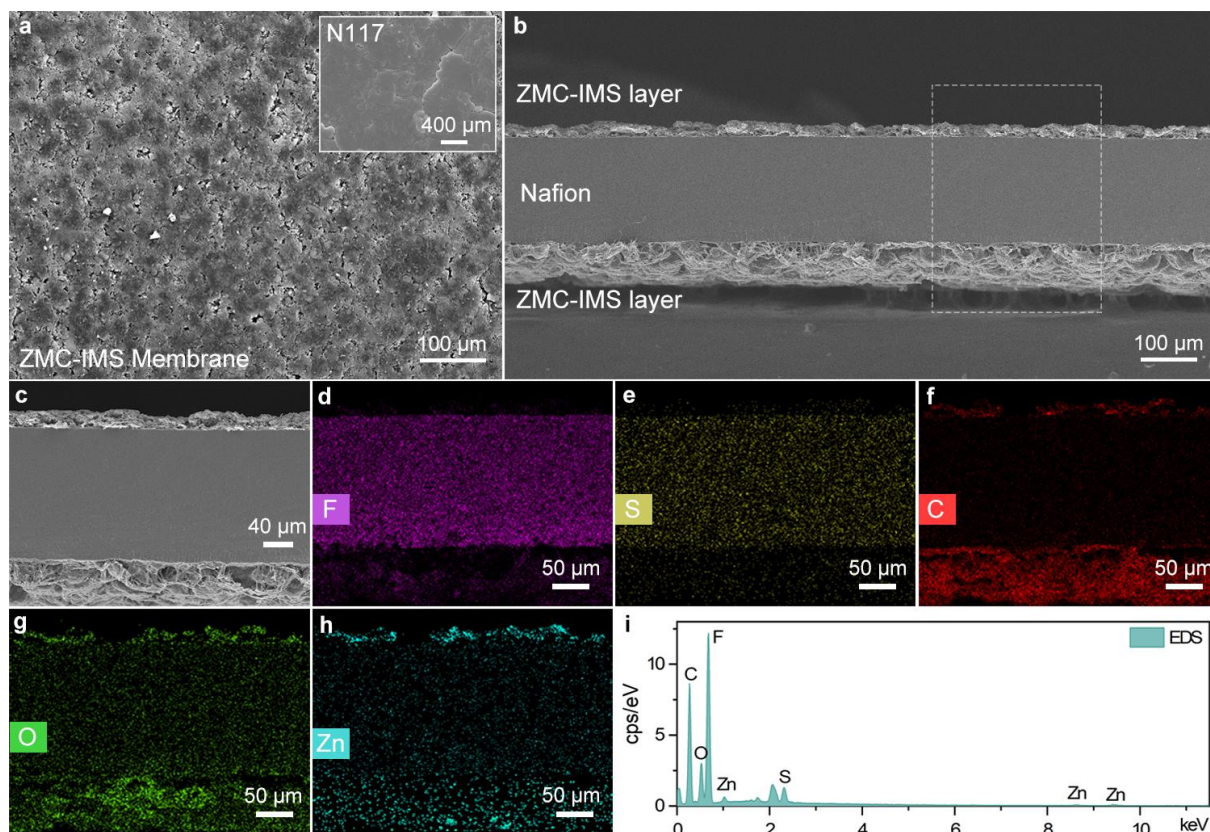
Supplementary Fig. 19 | Ion transference number of CNT membrane. The cation transference numbers of K^+ , Na^+ , Zn^{2+} . The transference numbers calculated from V-I profiles for 0.1 M/0.01 M concentration gradients in the two reservoirs connected by the CNT membrane.



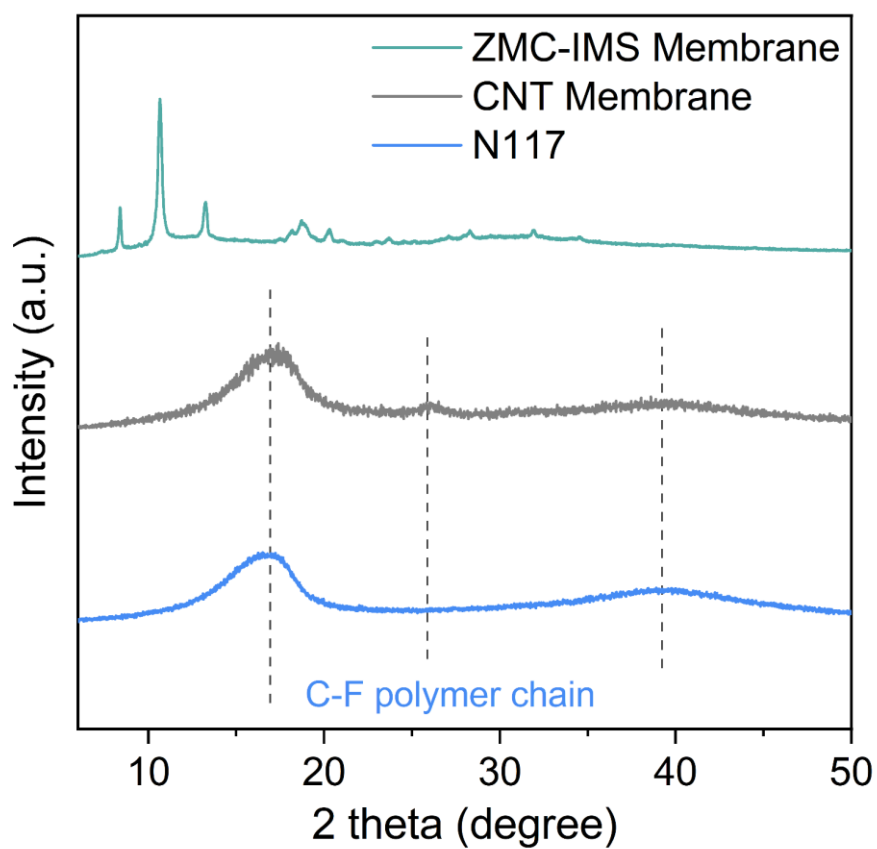
Supplementary Fig. 20 | KI_x permeation under CNT membranes. (a) Two-compartment H-cell testing configuration of 2 M KI_x permeation using (a) CNT membrane. (b) Photographs of the 2 M KI_x permeate solutions under the CNT membrane. (c) UV-vis of the KI_x permeate side. (d) $-\ln(1 - c_B/c_A)$ vs. permeation time for the determination of permeability of 2 M KI_x under the CNT membrane. The fits in the $-\ln(1 - c_B/c_A)$ vs. t (time) plots in (d) were obtained by linear fitting.



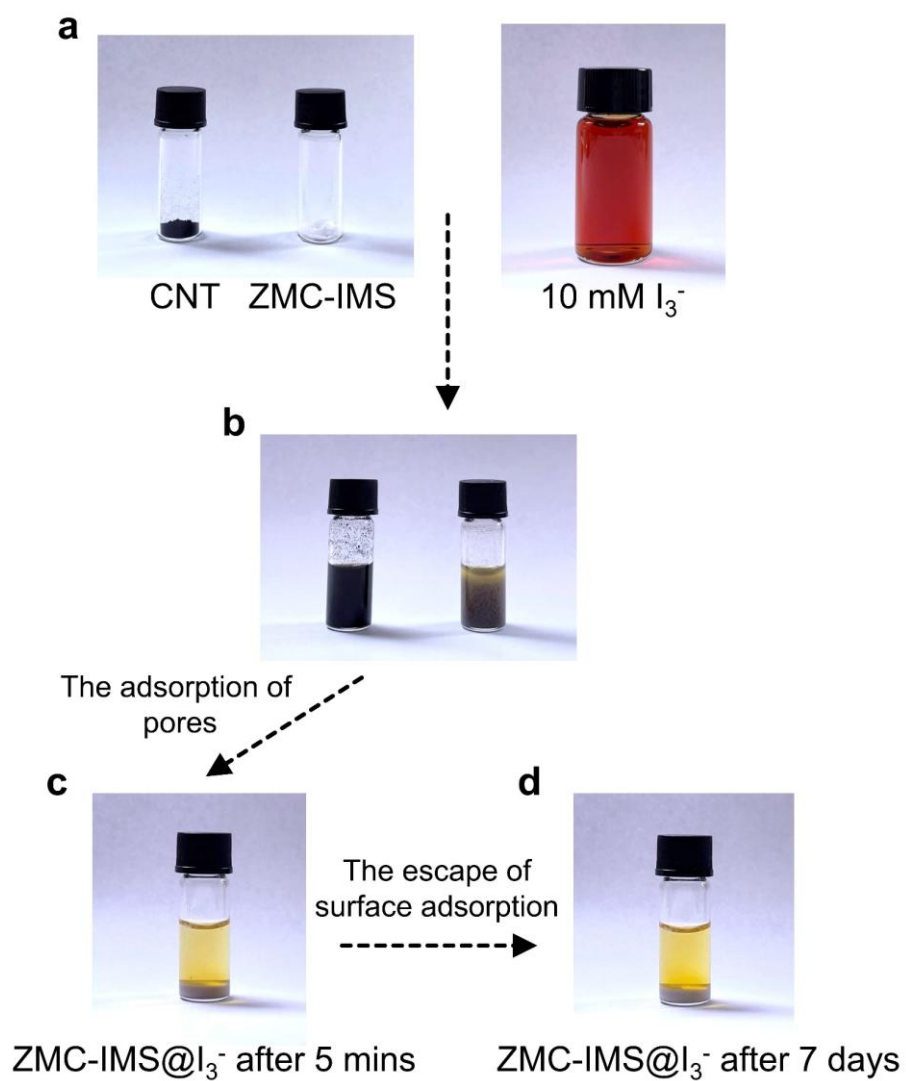
Supplementary Fig. 21 | Characterization of the ZMC-IMS powders. (a & b) SEM images of the ZMC-IMS. (c - f) The corresponding EDS-mapping of ZMC-IMS.



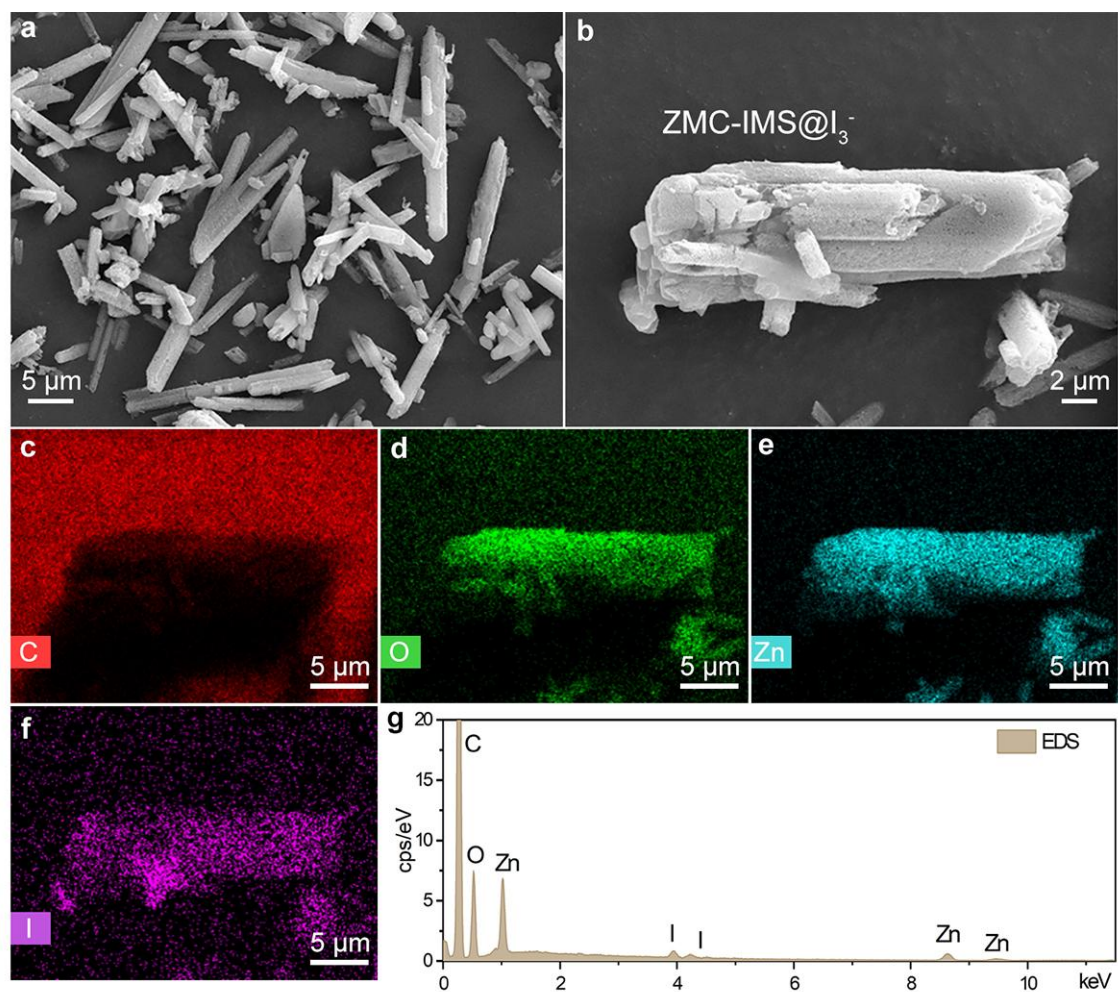
Supplementary Fig. 22 | Morphology of ZMC-IMS membrane. SEM images of (a) surface morphology and (b) & (c) cross-section of ZMC-IMS membranes. (c) Cross-sectional SEM image and (d - i) the corresponding EDS-mapping of ZMC-IMS membranes. (The inset: surface morphology of pristine N117 membranes)



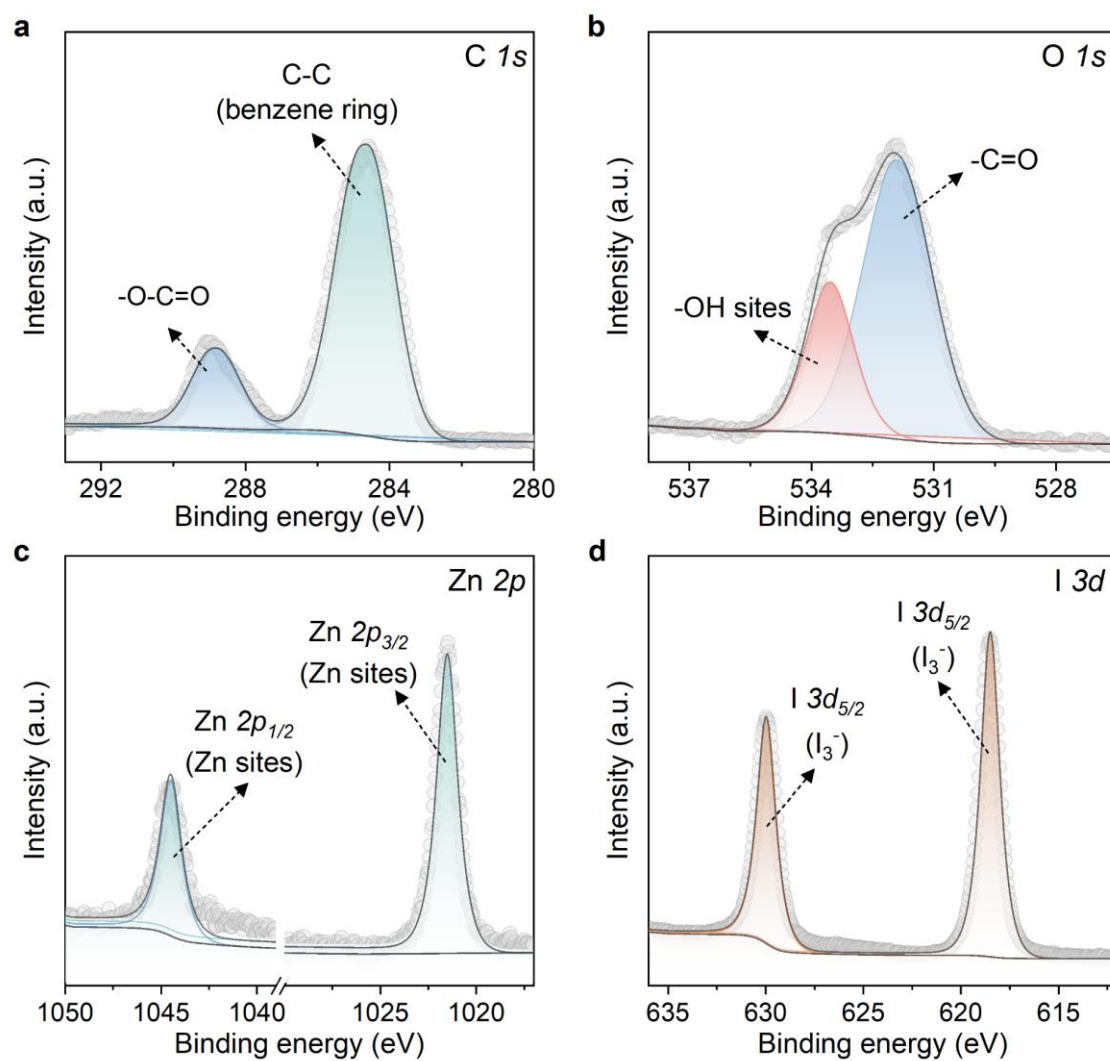
Supplementary Fig. 23 | Crystal structures of different membranes. XRD pattern of pristine N117, CNT and ZMC-IMS membranes.



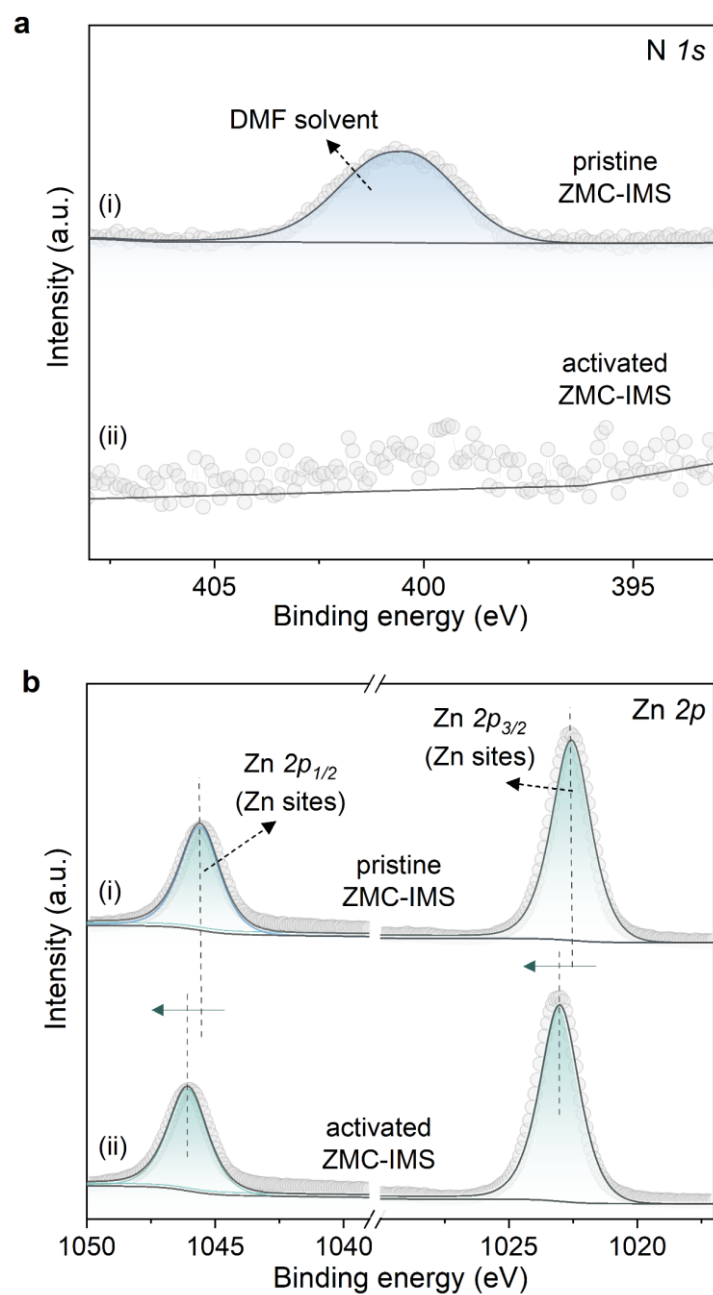
Supplementary Fig. 24 | Adsorption capacity of ZMC-IMS for I_3^- . Photographs of CNT and ZMC-IMS before and after adsorption the I_3^- .



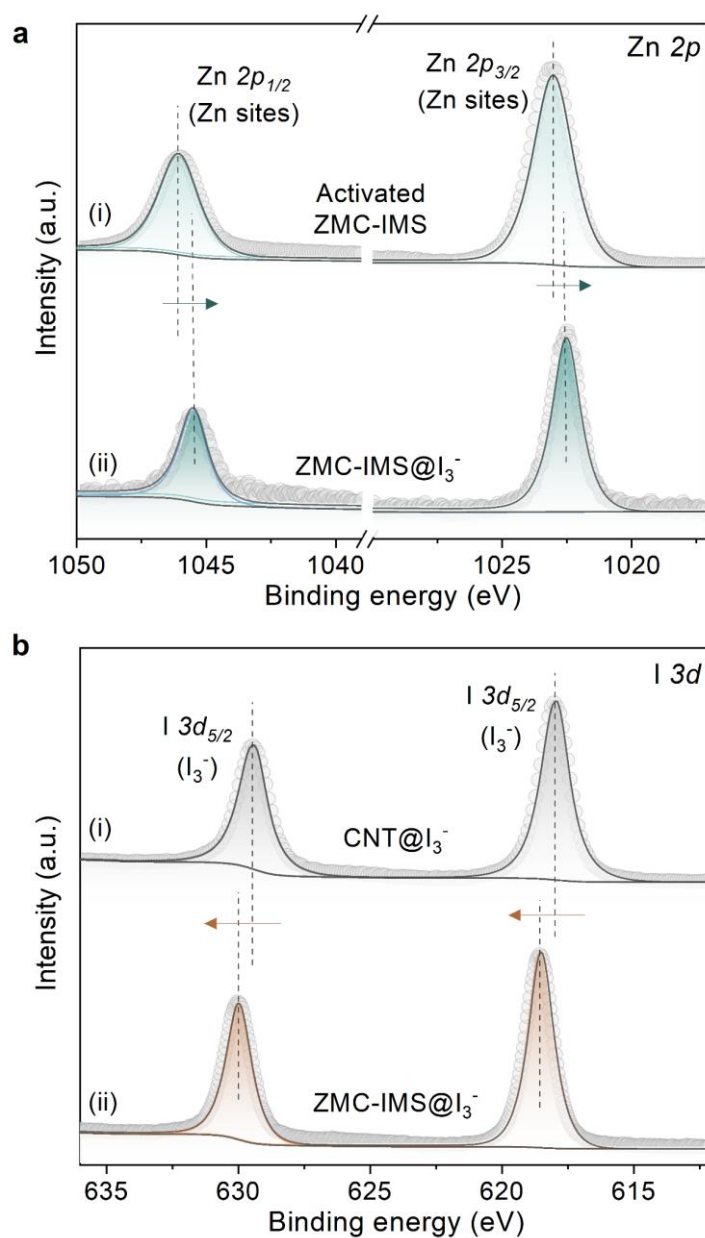
Supplementary Fig. 25 | Morphology of the ZMC-IMS@I₃⁻. (a & b) SEM images of the ZMC-IMS after dipping in 0.1 M I₃⁻. (c - g) The corresponding EDS-mapping of ZMC-IMS membranes.



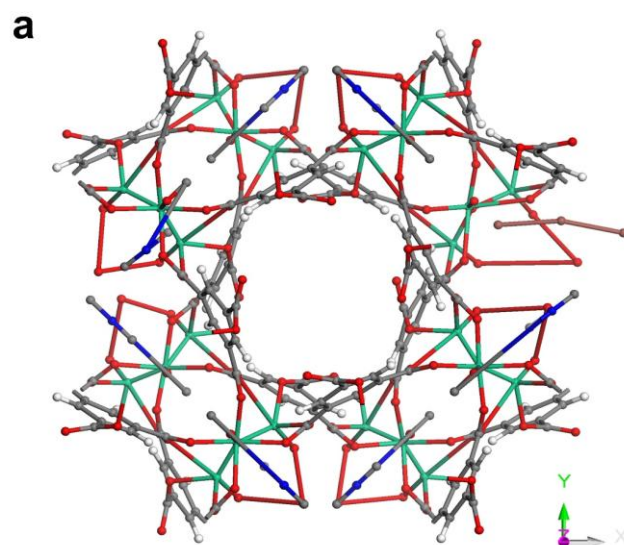
Supplementary Fig. 26 | Electronic structure of ZMC-IMS@I₃⁻. (a - d) C 1s, O 1s, Zn 2p, and I 3d XPS depth profiles of ZMC-IMS@I₃⁻.



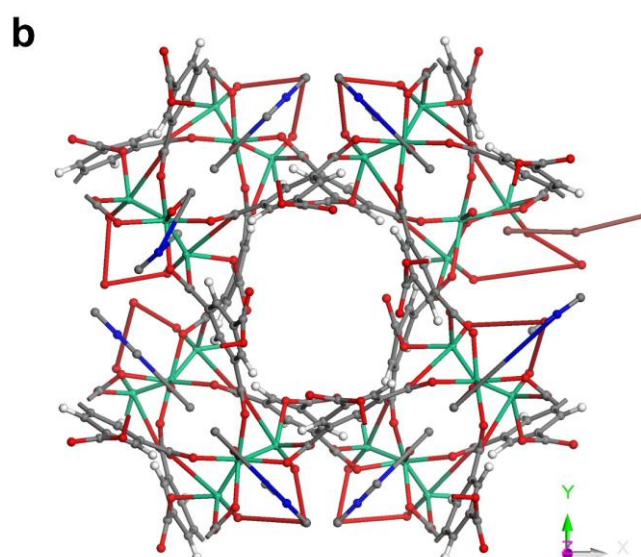
Supplementary Fig. 27 | Electronic structure of ZMC-IMS after activation. (a) N 1s and (b) Zn 2p XPS depth profiles of pristine ZMC-IMS and activated ZMC-IMS.



Supplementary Fig. 28 | Electronic structure of different materials after absorbing I₃⁻. (a) Zn 2p XPS depth profiles of activated ZMC-IMS and ZMC-IMS@I₃⁻. (b) I 3d XPS depth profiles of CNT@I₃⁻ and ZMC-IMS@I₃⁻.

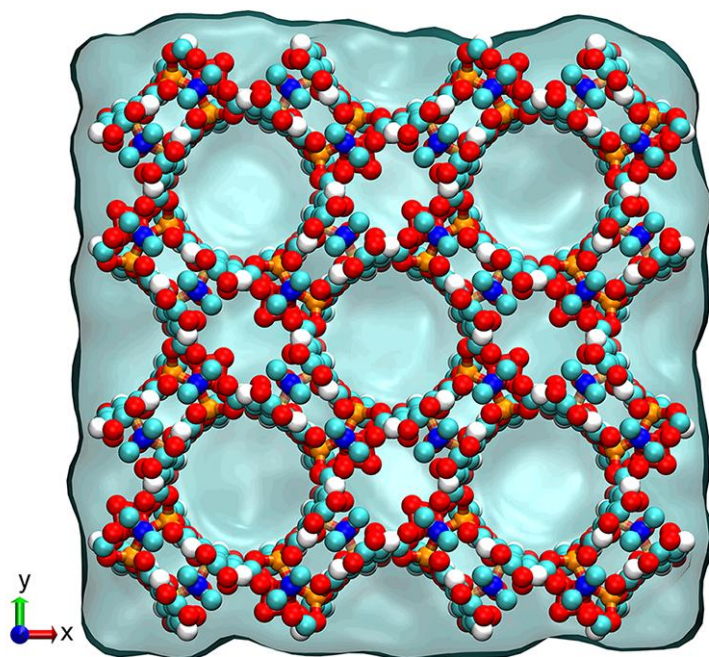


*ZMC-IMS@I₃⁻
on metal active sites*

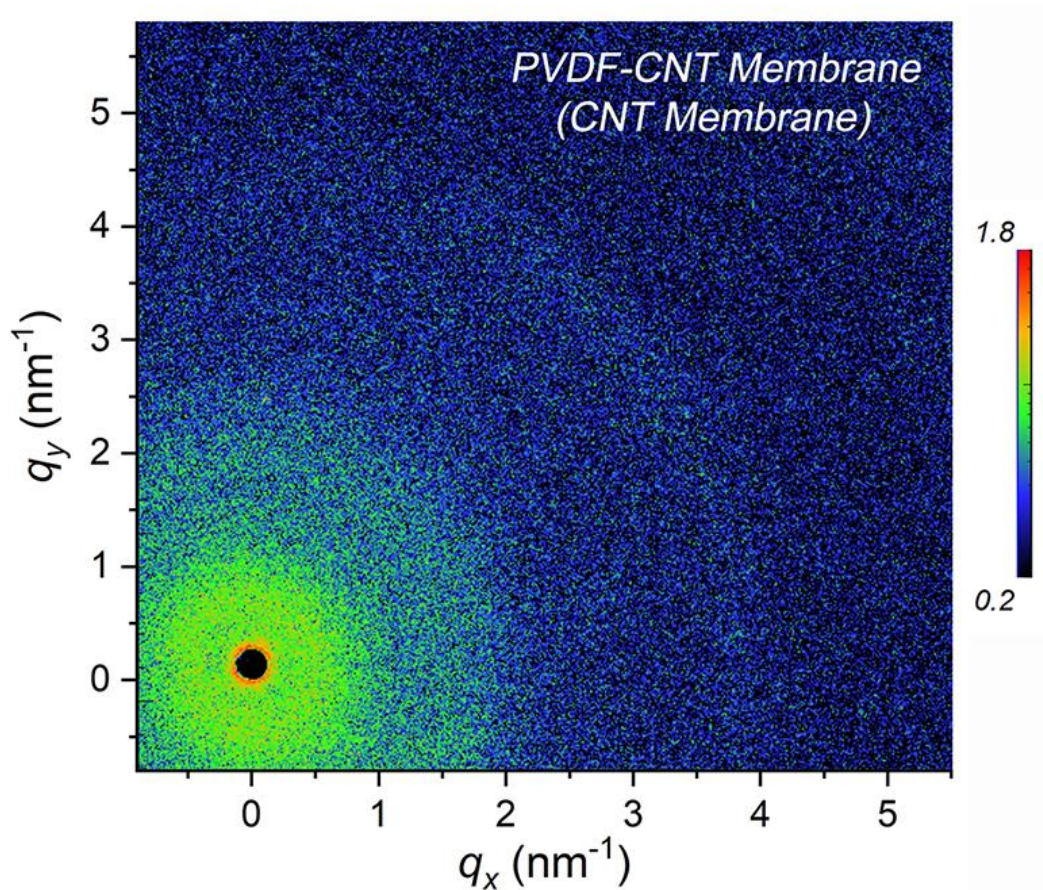


*ZMC-IMS@I₃⁻
on hydroxyl active sites*

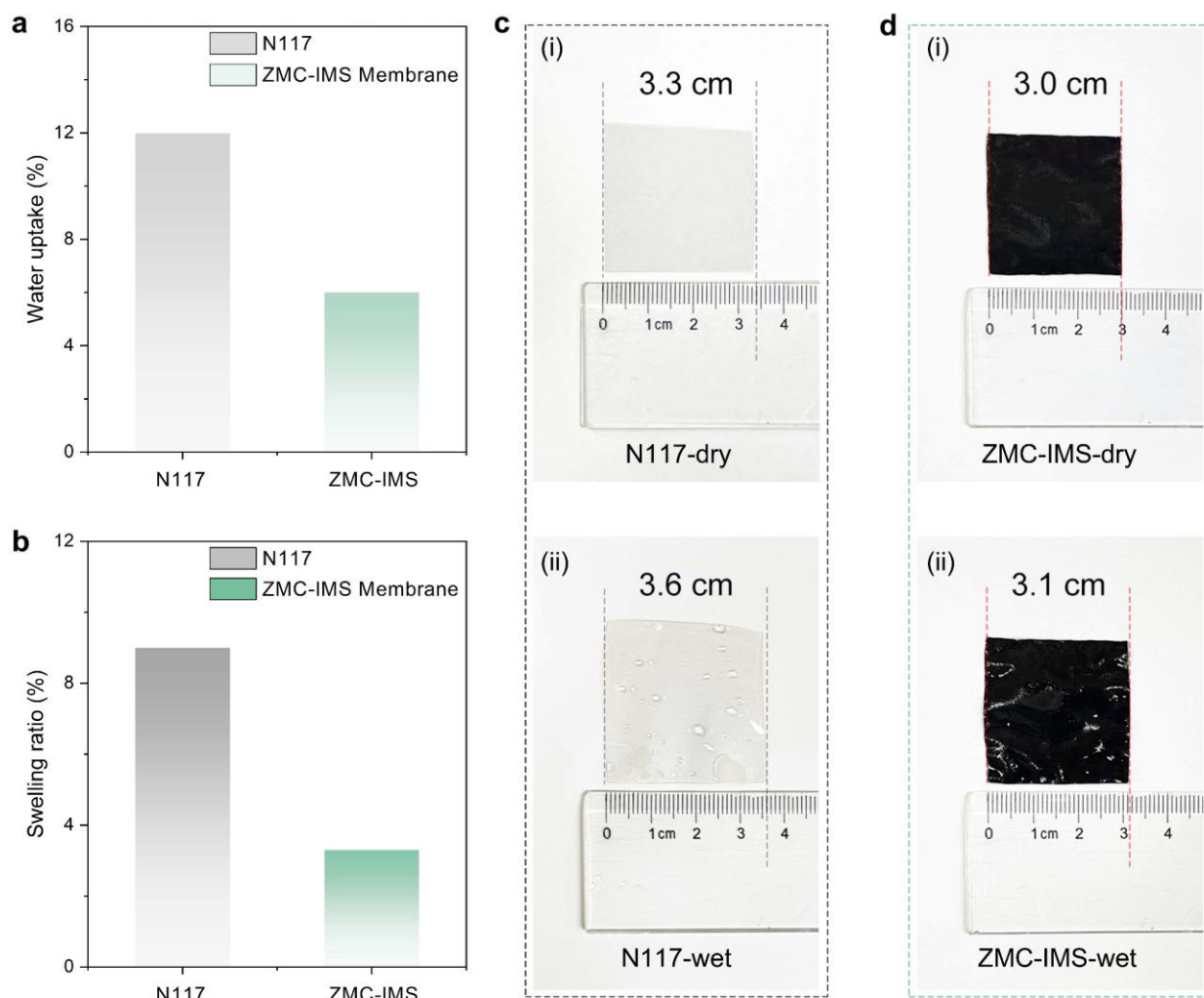
Supplementary Fig. 29 | Density functional theory (DFT) calculation structure. The interaction model between ZMC-IMS and I₃⁻ under (a) metal active sites and (b) oxygen active sites.



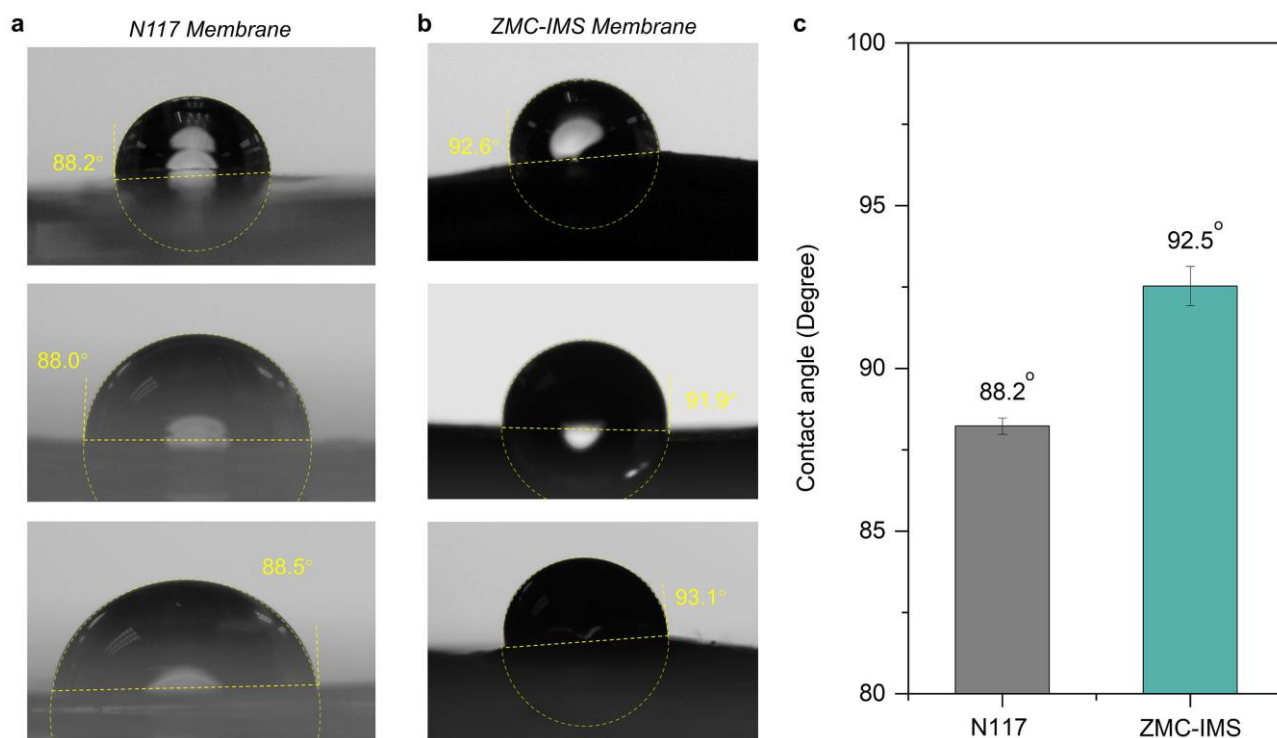
Supplementary Fig. 30 | Molecular dynamics simulation structure. Simulated ZMC-IMS membranes with a nanochannel, where zinc, carbon, nitrogen, oxygen, and hydrogen atoms are represented in orange, cyan, blue, red, and white, respectively.



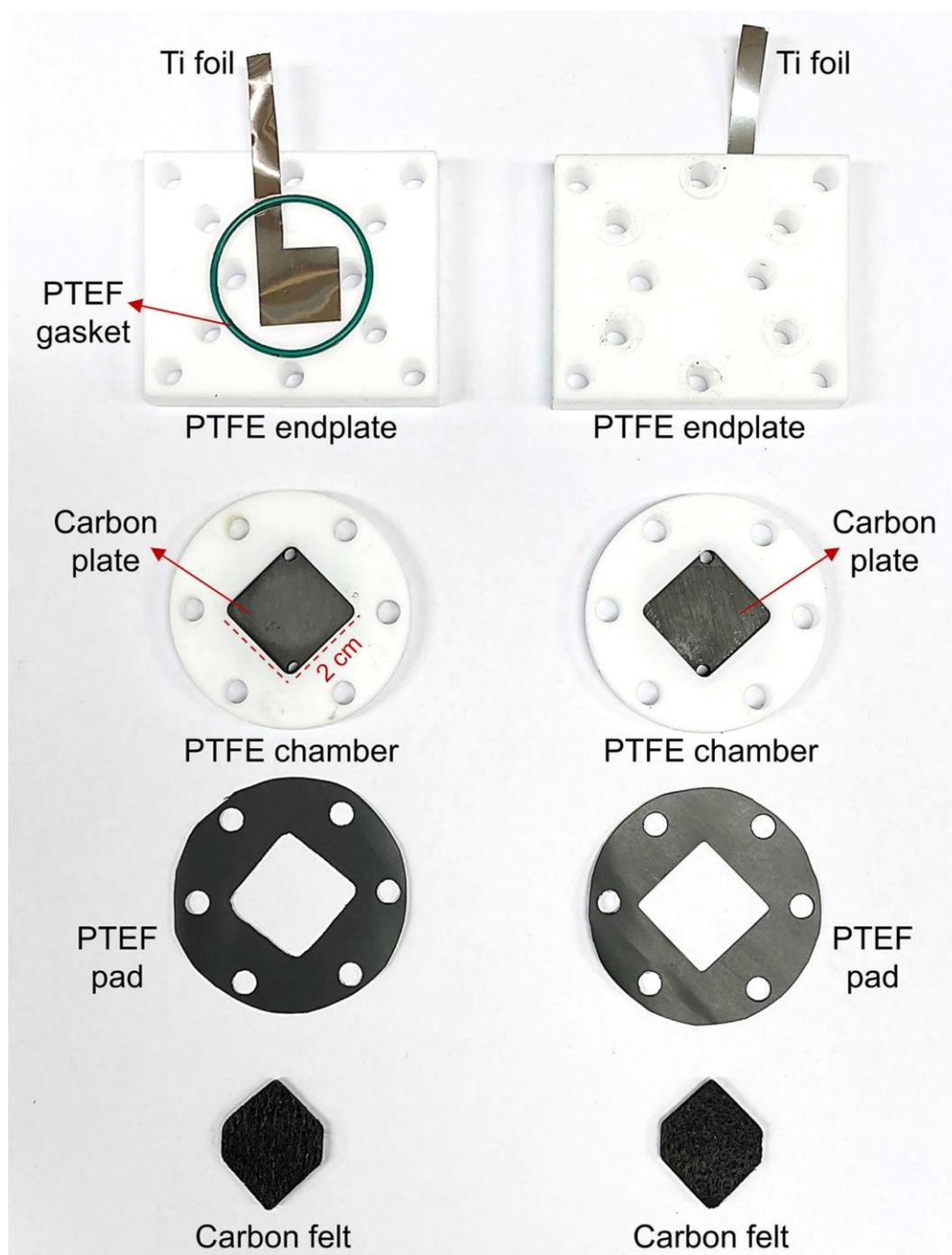
Supplementary Fig. 31 | Small-angle X-ray scattering measurement. Two-dimensional scattering pattern of CNT membranes.



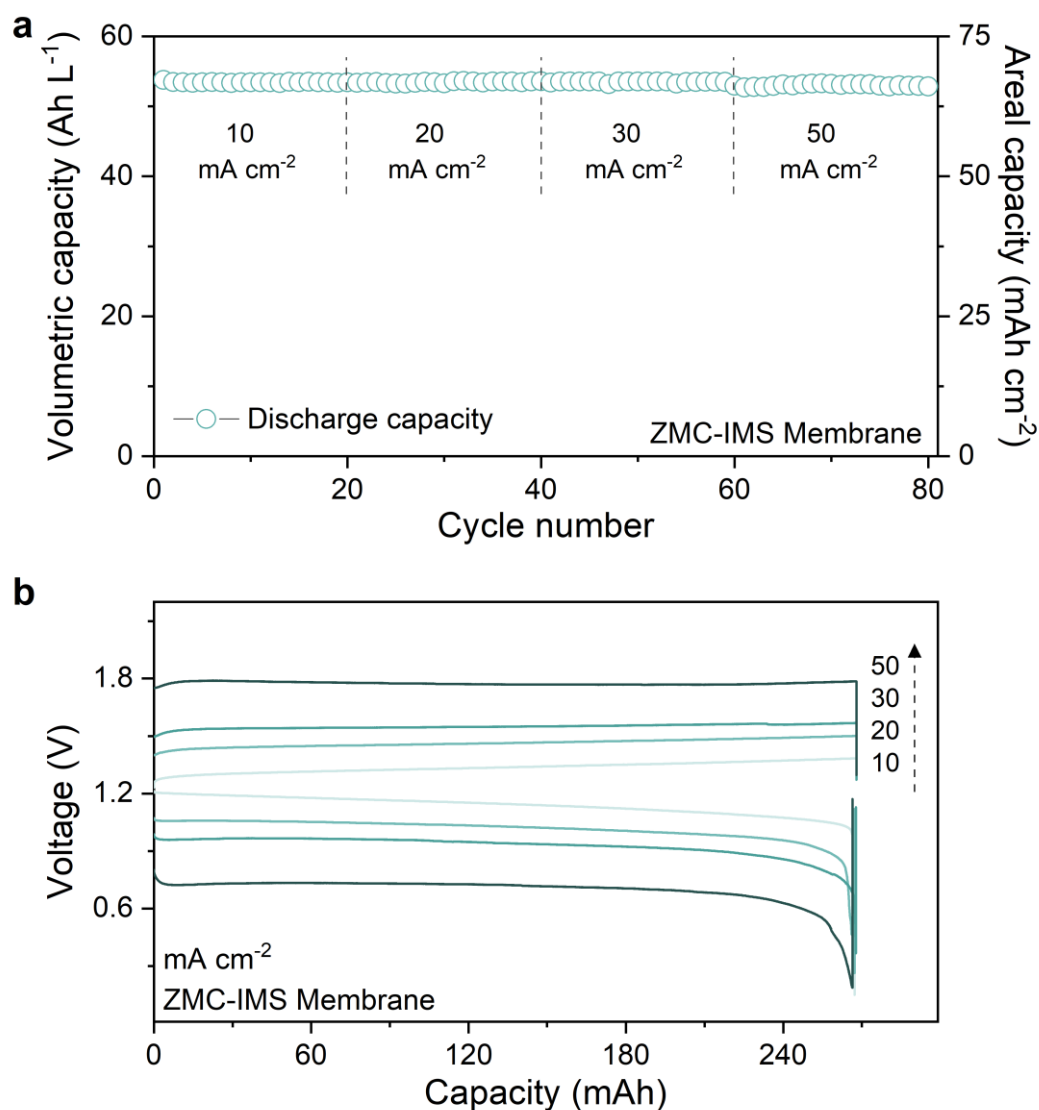
Supplementary Fig. 32 | Comparison of water uptake of different membranes. (a) Water uptake (b) swelling ratio of N117 and ZMC-IMS membranes. (c & d) photographs of the swelling behavior of N117 and ZMC-IMS membranes.



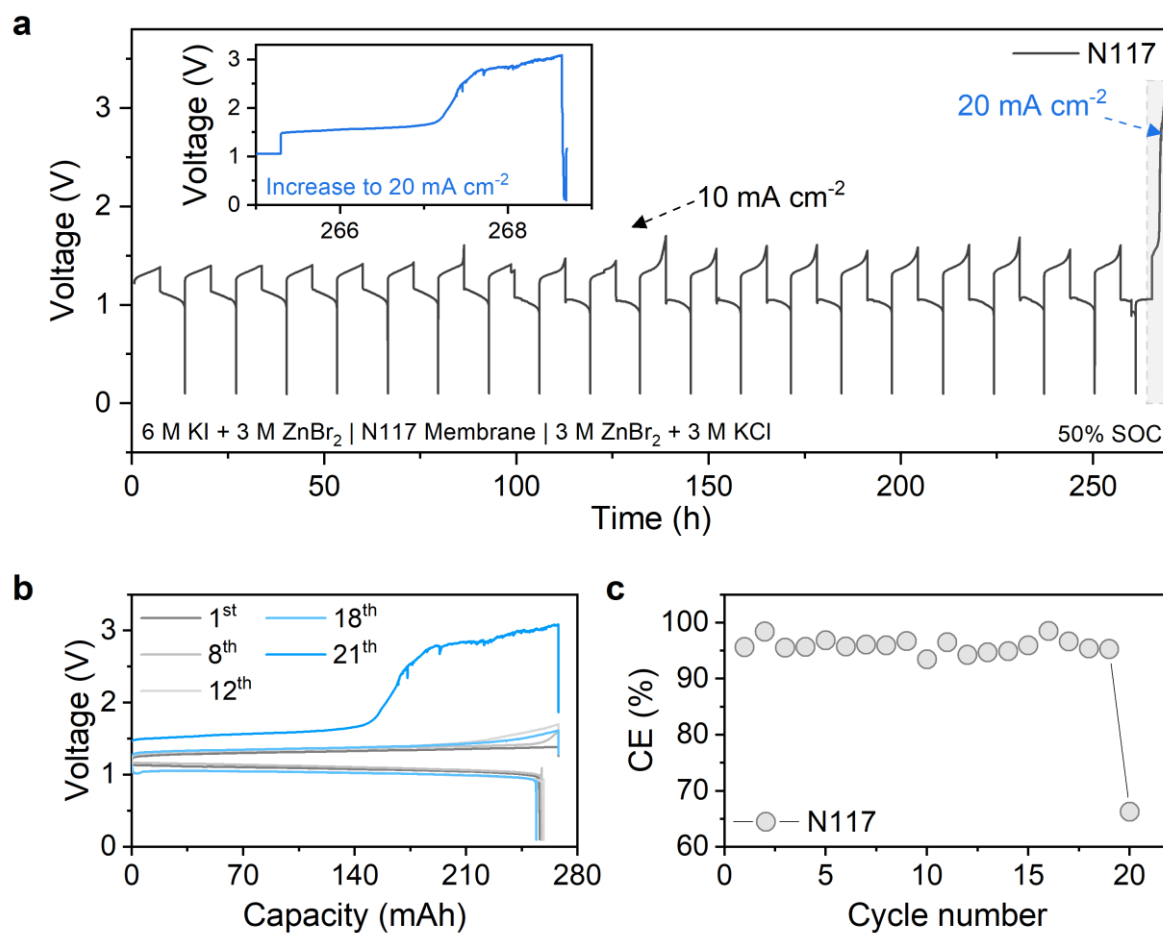
Supplementary Fig. 33 | Comparison of hydrophilicity and hydrophobicity of different membranes. The Digital image of the contact angle by water on (a) N117 and (b) ZMC-IMS membranes. (c) Comparison of contact angles with error bars derived from triplicate measurements. All error bars represent standard deviations from three independent tests.



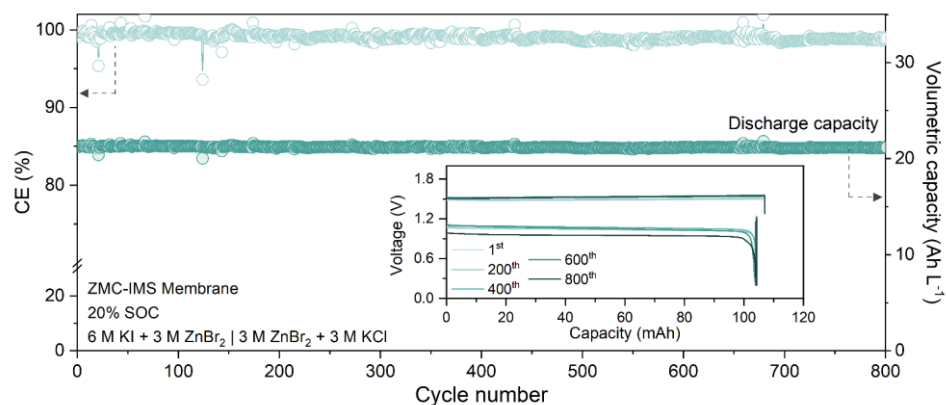
Supplementary Fig. 34 | Components of flow cell. Photographs of the cell components of $2 \times 2 \text{ cm}^2$ cell for flow cell tests.



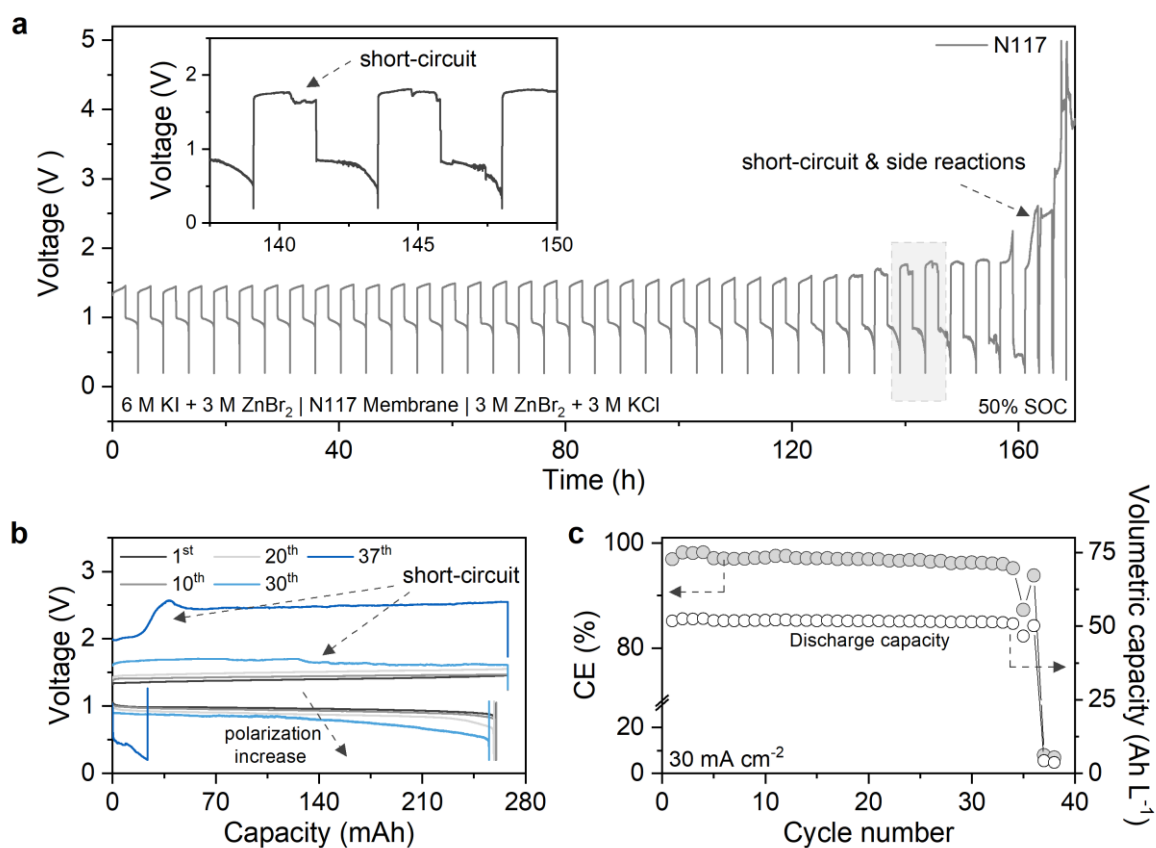
Supplementary Fig. 35 | Rate performance of Zn-I FBs with ZMC-IMS membranes. (a) Galvanostatic cycling, (b) voltage profiles, and (c) CE of Zn-I FBs with ZMC-IMS membranes (5 ml of 6 M KI + 3 M ZnBr₂ | 5 ml of 3 M ZnBr₂ + 3 M KCl, 4 cm² membrane area, 50% SOC) under 10 and 20 mA cm⁻².



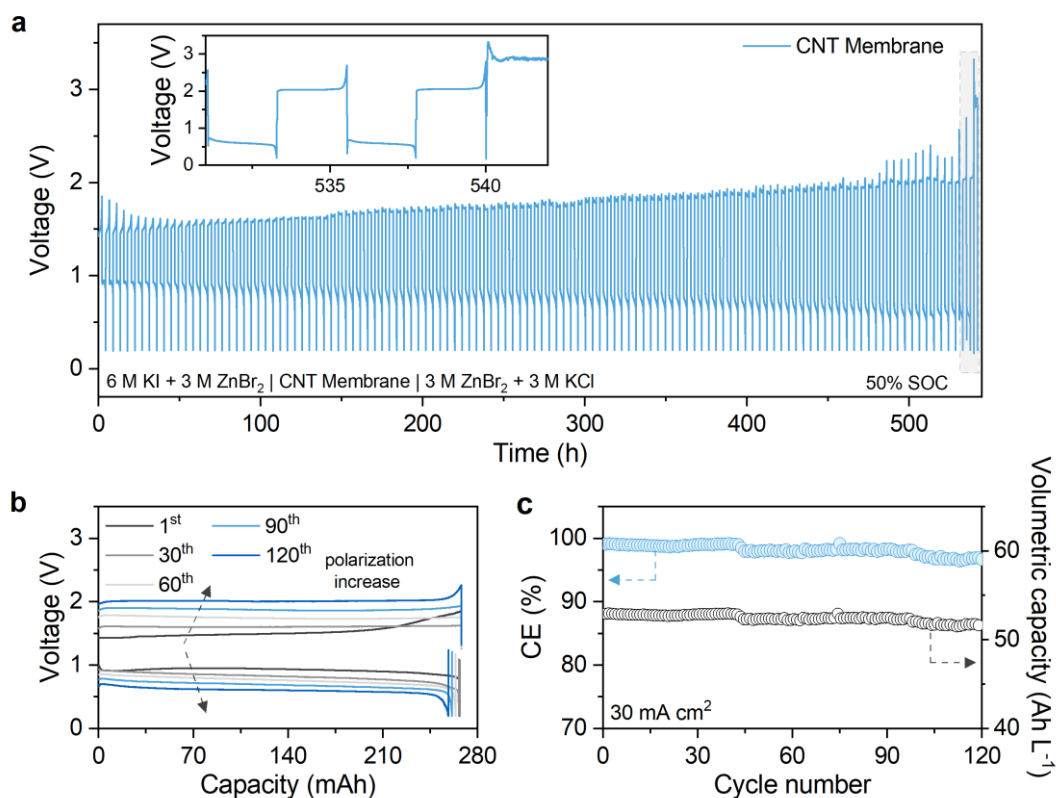
Supplementary Fig. 36 | Rate performance of Zn-I FBs with N117 membranes. (a) Galvanostatic cycling and (b) voltage profiles of Zn-I FBs with N117 membranes (5 ml of 6 M KI + 3 M ZnBr₂ | 5 ml of 3 M ZnBr₂ + 3 M KCl, 4 cm² membrane area, 50% SOC) under 10, 20, 30, and 50 mA cm².



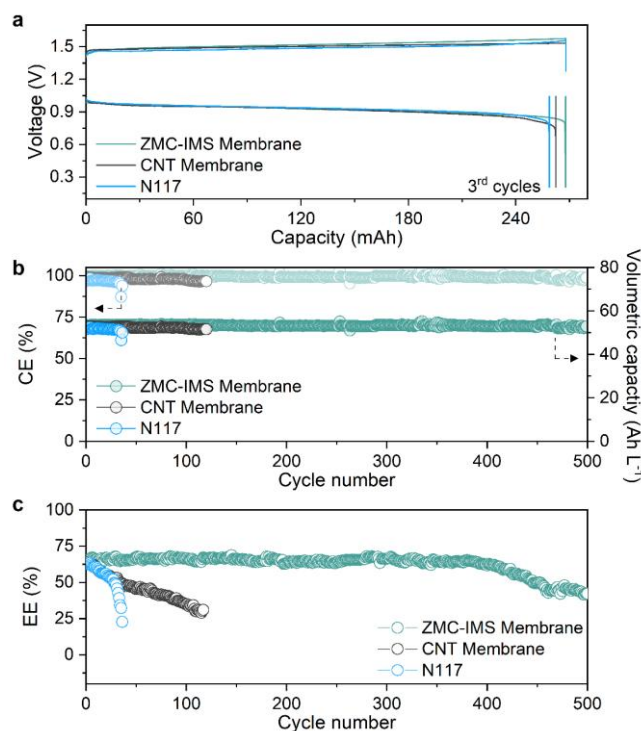
Supplementary Fig. 37 | Long cycling performance of Zn-I FBs with ZMC-IMS membranes at 20% SOC. CE of Zn-I FBs with ZMC-IMS membranes (5 ml of 6 M KI + 3 M ZnBr₂ | 5 ml of 3 M ZnBr₂ + 3 M KCl, 4 cm² membrane area, 20% SOC) under 30 mA cm⁻². (The inset: voltage profiles of Zn-I FBs with ZMC-IMS membrane)



Supplementary Fig. 38 | Long cycling performance of Zn-I FBs with N117 membranes at 50% SOC. (a) Galvanostatic cycling, (b) voltage profiles, and (c) CE of Zn-I FBs with N117 membranes (5 ml of 6 M KI + 3 M ZnBr₂ | 5 ml of 3 M ZnBr₂ + 3 M KCl, 4 cm² membrane area, 50% SOC) under 30 mA cm⁻².



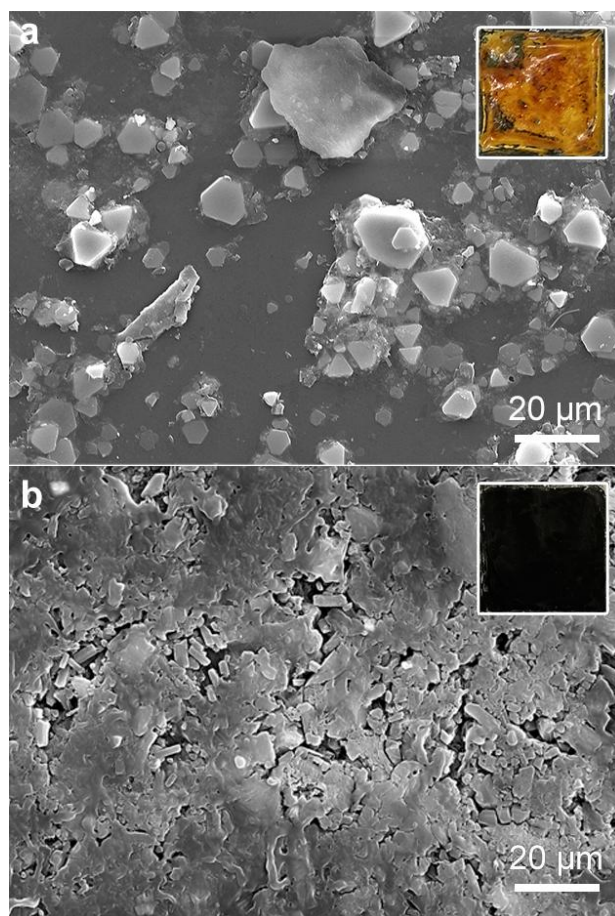
Supplementary Fig. 39 | Long cycling performance of Zn-I FBs with CNT membranes at 50% SOC. (a) Galvanostatic cycling, (b) voltage profiles, and (c) CE of Zn-I FBs with CNT membranes (5 ml of 6 M KI + 3 M ZnBr₂ | 5 ml of 3 M ZnBr₂ + 3 M KCl, 4 cm² membrane area, 50% SOC) under 30 mA cm⁻².



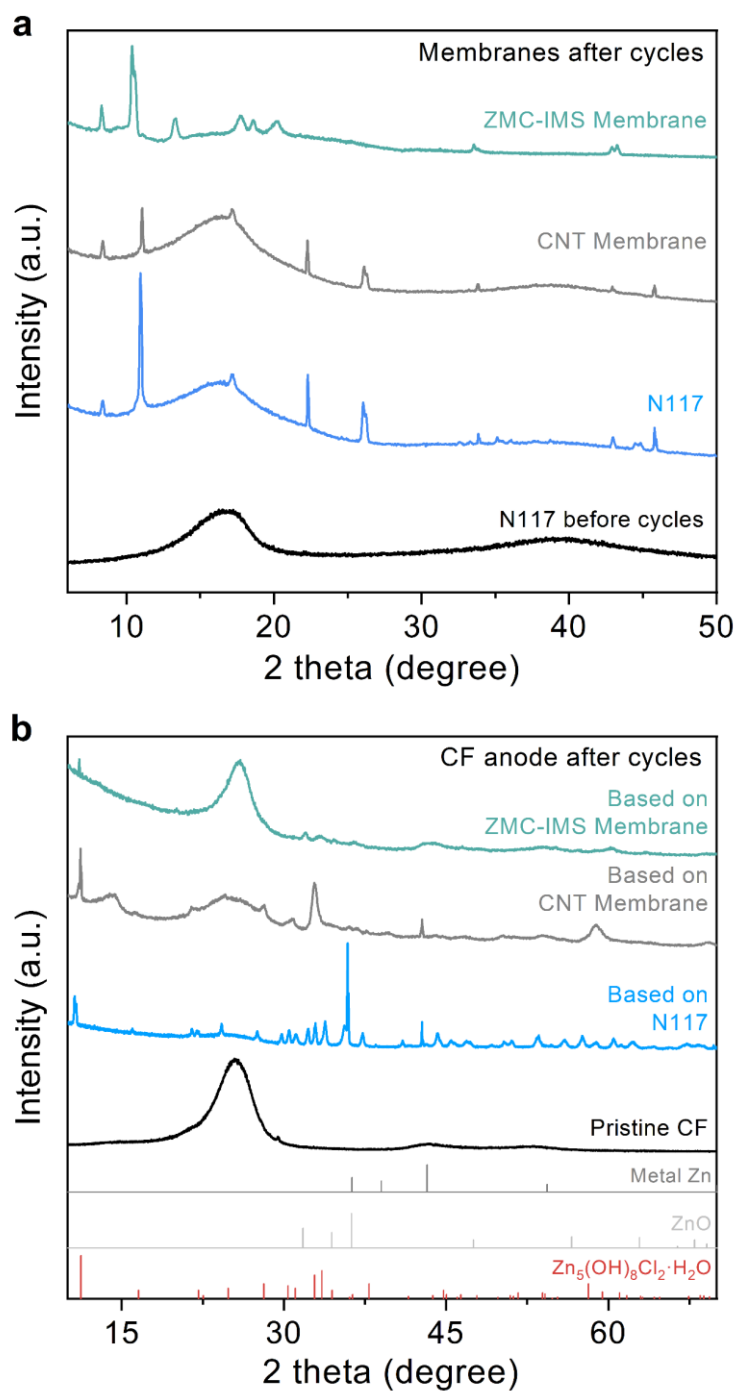
Supplementary Fig. 40 | Long cycling performance comparison of Zn-I FBs with different membranes. Comparison of (a) voltage profiles, (b) CE, and (c) EE of Zn-I FBs with N117, CNT and ZMC-IMS membranes (5 ml of 6 M KI + 3 M ZnBr₂ | 5 ml of 3 M ZnBr₂ + 3 M KCl, 4 cm² membrane area, 50% SOC) under 30 mA cm⁻².

Supplementary Note: It should be clarified that current densities of 20–50 mA cm⁻² are reasonable for conventional FBs using Nafion membrane. This current limitation is primarily due to the membrane with low ion conductivity and high ohmic resistance. Despite operating in soft conditions, reported cycle lifetimes of Zn-I FBs remain disappointingly short. Despite these soft conditions, reported cycle lifetimes of Zn-I FBs remain disappointingly short. Meanwhile, high SOC (>50%) risks I₂ solid precipitation in I-based posolyte, severely degrading performance, which means 50% SOC represents a practical upper limit for Zn-I FB systems in the normal electrolyte. However, many studies use excess electrolyte volumes at low SOC (<20%) to neglect water migration issues of Zn-I FBs, which sacrifices capacity utilization and increases costs. In stark contrast, to the best of our knowledge, our work achieves the highest I-based SOC condition with 2000 h of stable cycling at 30 mA cm⁻² and >99% high CE and 53.2 Ah L⁻¹_{posolyte} high volumetric energy density, demonstrating resilience under conditions that are indeed “harsh” for Zn-I FBs chemistry.

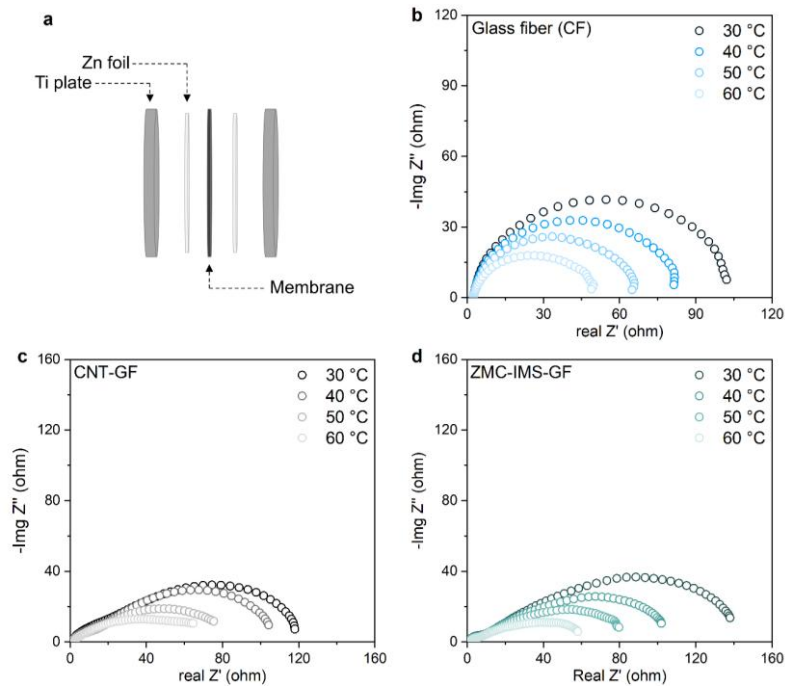
On the other hand, precise retention of hydrated ions by the ZMC-IMS membrane can allow smaller hydrated ions to rapidly transport through the membrane channels, thereby avoiding energy losses associated with dehydration and rehydration that can occur in ultra-small pore sizes.¹² As shown in **Supplementary Fig. 40**, the GCD curves of Zn-I FBs with the ZMC-IMS membrane exhibit a slight increase compared to those with the N117 membrane. Despite this minor effect, under high SOC and high areal capacity testing conditions, the ZMC-IMS membrane enables highly stable and reversible cycling of the Zn-I FBs at current densities ranging from 10 to 50 mA cm⁻² (**Fig. 4a & b**). Therefore, we believe that the introduction of the coating layer has a minimal impact on polarization at high rates, primarily serving to enhance the cycle life of the FBs.



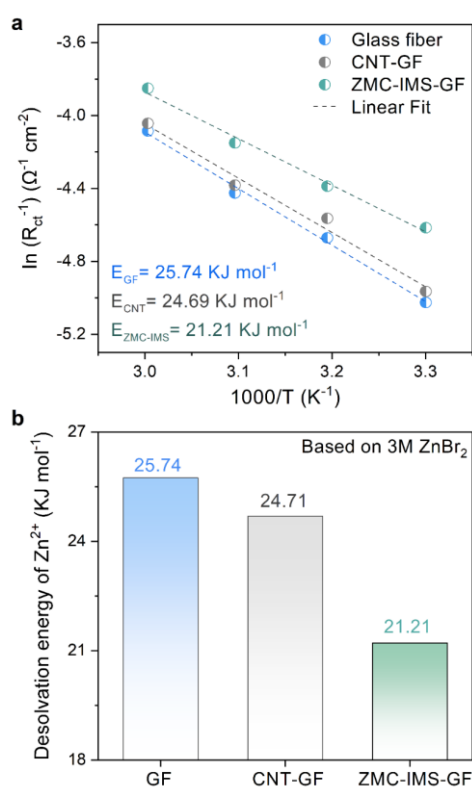
Supplementary Fig. 41 | Morphology of membranes after cycling. SEM images of (a) N117 membrane and (b) ZMC-IMS membrane on the anode side in the discharging state after cycles. (The inset: photographs of N117 and ZMC-IMS membranes after cycles)



Supplementary Fig. 42 | Structure of membranes after cycling. (a) XRD pattern of N117, CNT, and ZMC-IMS membranes on the anode side in the discharging state after cycles. (b) XRD pattern of CF anode using N117, CNT, and ZMC-IMS membranes in the discharging state after cycles, wherein PDF cards of $\text{Zn}_5(\text{OH})_8\text{Cl}_2 \cdot \text{H}_2\text{O}$ and ZnO by-products are #07-0155 and #36-1451.

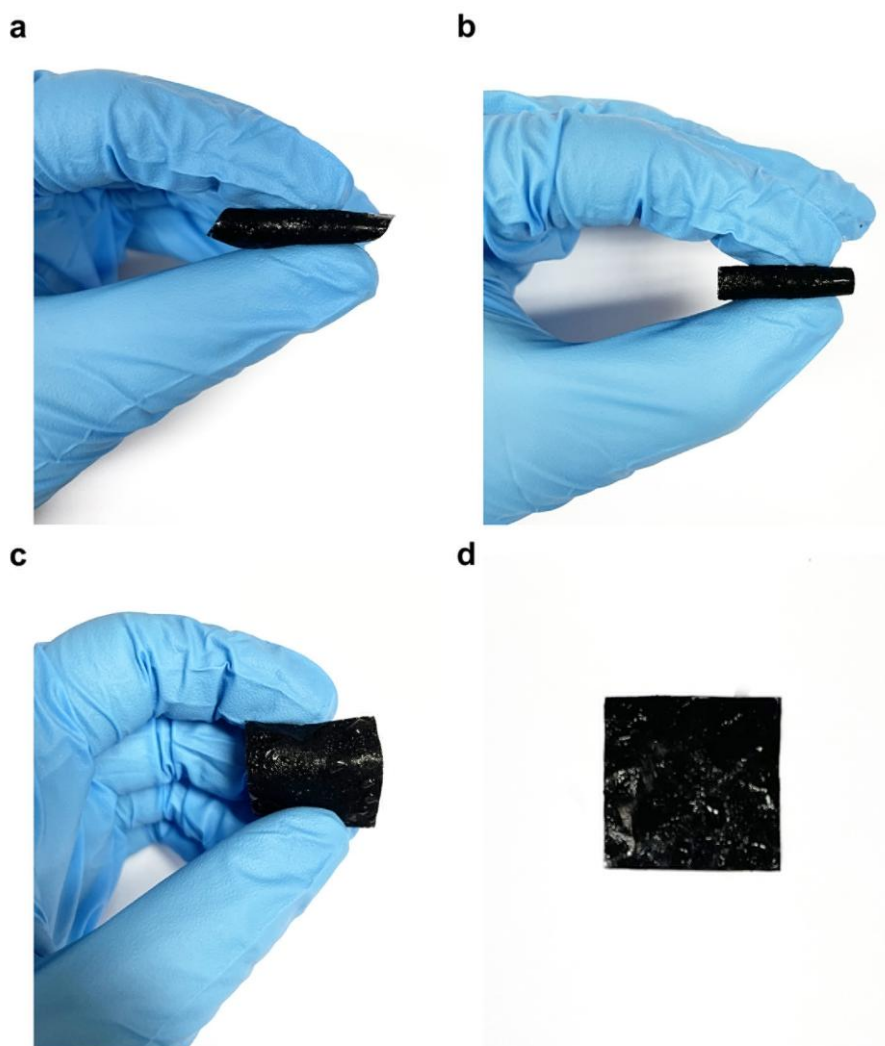


Supplementary Fig. 43 | Effects of different membranes on Zn anode reactions. Schematic illustration of Zn symmetric cells for testing Nyquist plots. Nyquist plots of Zn symmetric cells with (b) glass fiber, (c) CNT-GF, and (d) ZMC-IMS-GF membrane at different temperatures.



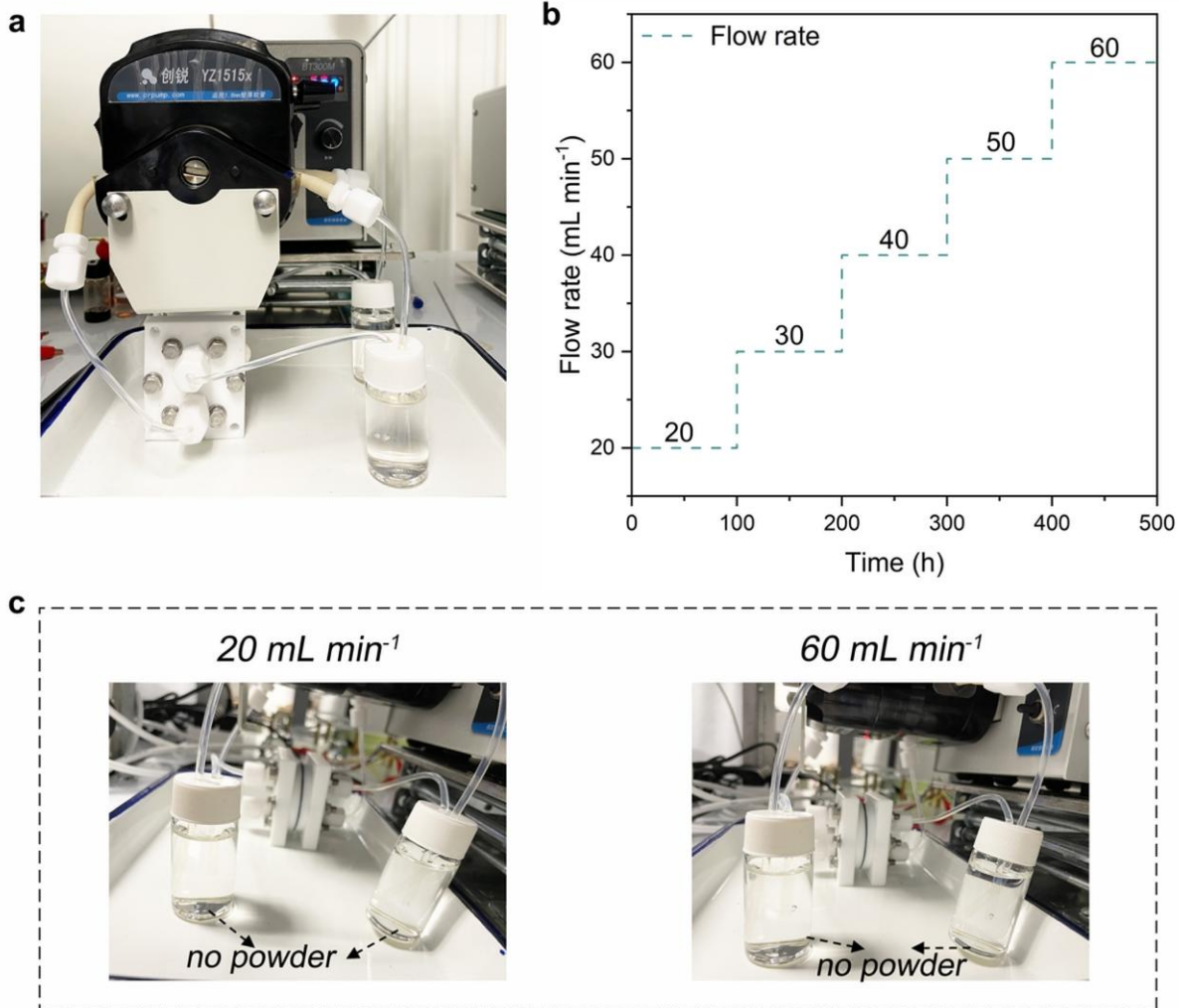
Supplementary Fig. 44 | Energy of different membranes on Zn anode reactions. The Arrhenius plot and (b) calculated desolvation energy based on the charge-transfer resistances with glass fiber, CNT-GF, and ZMC-IMS-GF membrane.

Supplementary Note: It should be noted that uniform Zn deposition may arise from the modulation of Zn desolvation energy, as indicated by shifts in the Arrhenius dependence of charge transfer resistance observed in the EIS measurements of a symmetric cell (**Supplementary Fig. 43 and 44**).



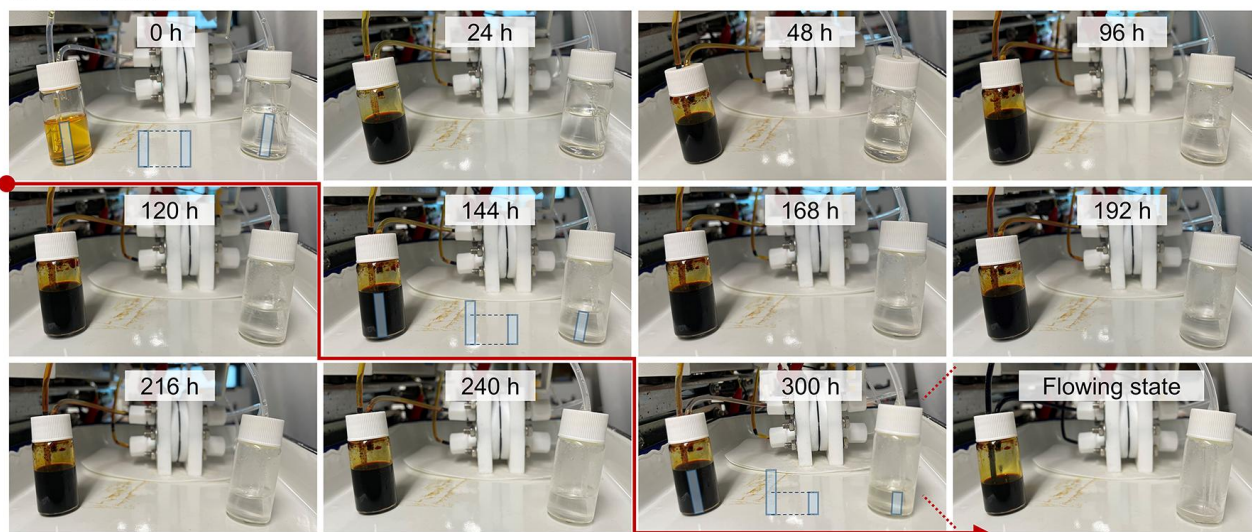
Supplementary Fig. 45 | Mechanical properties of ZMC-IMS membrane. (a - d) Photographs of ZMC-IMS membranes after 100 times of bending, and no coating fragments were found peeling off due to frequent bending from the photographs.

Supplementary Note: We incorporate CNTs as a membrane-forming agent in our ZMC-IMS slurry to enhance membrane formation. The addition of CNTs improves the uniformity and adhesion of the membrane, ensuring a more consistent and stable structure.¹³ Additionally, CNTs contribute to the electrical conductivity of the membrane to enhance charge transport, which can facilitate the reversible redox cycling of the iodide-based active materials, ensuring efficient and stable electrochemical reactions.^{14,15} Furthermore, incorporating CNTs can improve the mechanical properties of the membrane, providing greater resilience and stability during operation.¹⁶



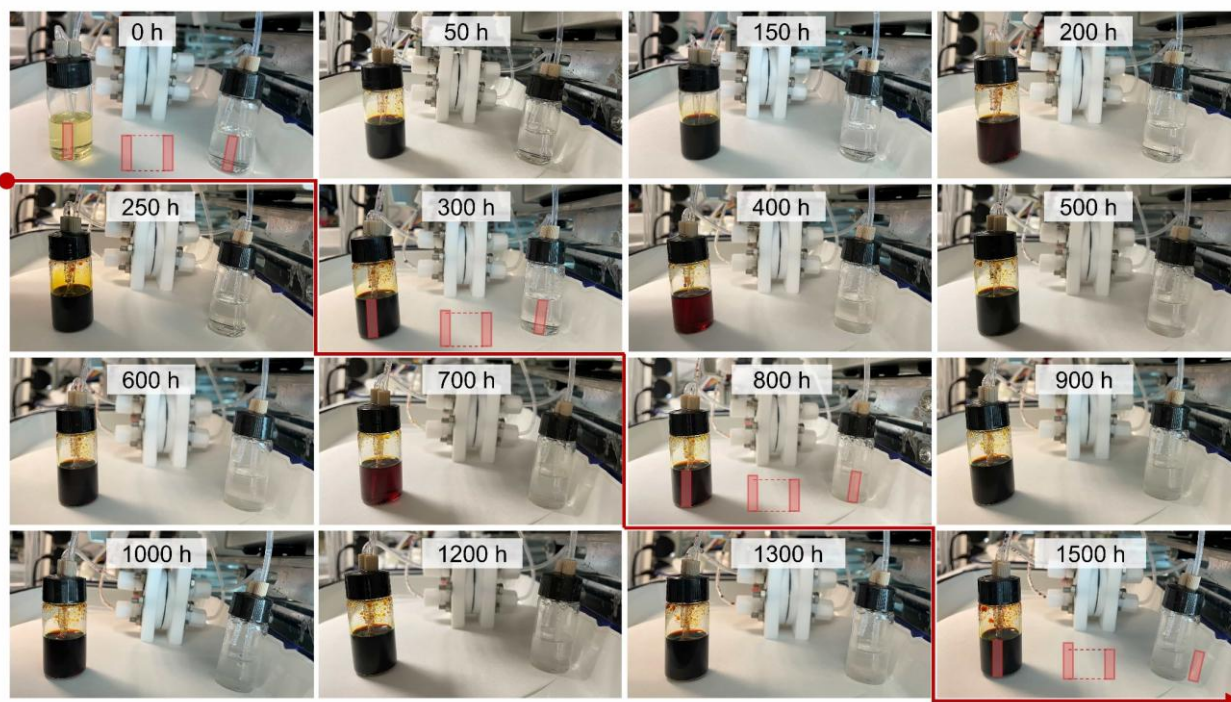
Supplementary Fig. 46 | Mechanical stability of ZMC-IMS membrane. (a) Photographs of the water flushing experiment on ZMC-IMS membranes. (b & c) digital images of residual in the water tank after 500 hours of water flushing under different flow rates.

N117 Membrane



Supplementary Fig. 47 | Photographs of water migration behavior under N117 membranes. The Zn-I FBs using N117 membranes showed a large volume change of posolyte and negolyte during flow cell operation of 6 M KI + 3 M ZnBr₂ | 3 M ZnBr₂ + 3 M KCl at 30 mA cm⁻² (4 cm²).

ZMC-IMS Membrane



Supplementary Fig. 48 | Photographs of water migration behavior under ZMC-IMS membranes. The Zn-I FBs using ZMC-IMS membranes showed a large volume change of posolyte and negolyte during flow cell operation of 6 M KI + 3 M ZnBr₂ | 3 M ZnBr₂ + 3 M KCl at 30 mA cm⁻² (4 cm²).

Supplementary Table 1 | Electrochemical comparison. Comparison of Zn-polyhalide FBs performance between this work and previously reported work.

Number	System	Working time (hour)	CE	Areal capacity (mAh cm ⁻²)	Volumetric capacity (Ah L ⁻¹ Posolyte)	Current density (mA cm ⁻²)	Per dis/charging time (hour)	Reference
1	Zn-I FBs	1500	88.5%	61	63.5	80	0.7625	<i>Angew. Chem.</i> 2018 , 130, 11341
2	Zn-I FBs	200	99.5%	35	40	40	0.875	<i>J. Power Sources</i> 2021 , 484, 229238
3	Zn-I FBs	60	99.2%	25	/	100	0.25	<i>Sci. Adv.</i> 2022 , 8, eabq4456
4	Zn-I FBs	159	99.2%	5.3	/	80	0.066	<i>Sci. Adv.</i> 2022 , 8, eabq4456
5	Zn-I FBs	2230	88.5%	160	57.6	40	4	<i>Sci. Bull.</i> 2021 , 66, 889
6	Zn-I FBs	140	99.5%	10	7	10	1	<i>Energy Environ. Sci.</i> , 2018 , 11, 2010
7	Zn-I FBs	136	95%	34	68	10	3.4	<i>Energy Environ. Sci.</i> , 2017 , 10, 735
8	Zn-I FBs	120	96%	35	35	100	0.35	<i>J. Mater. Chem. A</i> , 2022 , 10, 14090
9	Zn-I FBs	200	99%	3.35	/	20	0.1675	<i>J. Mater. Chem. A</i> , 2021 , 9, 16093
10	Zn-Br FBs	100	99%	40	/	80	0.5	<i>Small</i> 2019 , 15, 1901848
11	Zn-Br FBs	280	97.2%	40	7.2	40	1	<i>Adv. Mater.</i> 2020 , 32, 1906803
12	Zn-Br FBs	2000	97%	20	5.82	100	0.2	<i>Energy Environ. Sci.</i> , 2020 , 13, 2839
13	Zn-Br FBs	140	98.9%	40	6	80	0.5	<i>Adv. Funct. Mater.</i> 2021 , 31, 2102913
14	Zn-Br FBs	1100	99%	20	6	20	1	<i>J. Power Sources</i> , 2022 , 540, 231637
This work	Zn-I FBs	2050	99.5%	66.7	53.3	30	2.23	/

Supplementary Note: We grouped Zn-I, Zn-Br, and Zn-Cl as zinc-halogen systems due to their shared neutral/weakly acidic zinc anode chemistry and common challenges (e.g., water migration, crossover) for comparison purposes. Other systems are included for comparison and reference only, while Zn-I FBs are the primary focus.

Techno-economic analysis of redox FBs

Techno-economic modelling of emerging aqueous redox flow battery systems The LCOS of specific applications were calculated through the website tool (www.EnergyStorage.ninja, accessed: 18th September 2020). The webtool was developed by Schmidt's group involving the parameters in **Supplementary Table 2**. Full equations used to develop the webtool can be found in Schmidt's group.¹⁰ Considering the extended cycle life demonstrated by our Zn-I FBs, a 20-year project lifetime is adopted as a benchmark in the LCOS analysis, along with an 8% annual discount rate to account for discounted energy delivery over time, thereby underscoring the advantages of our developed Zn-I FB systems.

Supplementary Table 2 | Parameters for LCOS estimation of AFBs.

			<i>VRFBs</i>	<i>AQFBs</i>	<i>Zn-Fe FBs</i>	<i>Zn-Br FBs</i>	<i>Zn-I FBs (This work)</i>
<i>Investment cost-Power^a</i>	<i>USD/kW</i>	$C_p + C_{bop} + C_{add}$	472-627 <i>Ref¹⁷</i>	422.3	341.8	348.1	5502.7
<i>Investment cost-Energy^b</i>	<i>USD/kWh</i>	C_E	433-640 <i>Ref¹⁷</i>	72.8	50.84	105.32	48.9
<i>O&M cost-Power^c</i>	<i>USD/kW-yr</i>	C_{P-OM}	12	12	12	12	12
<i>O&M cost-Energy^c</i>	<i>USD/MWh</i>	C_{E-OM}	1	1	1	1	1
<i>Replacement cost^c</i>	<i>USD/kW</i>	C_{p-r}	0	0	0	0	0
<i>Replacement interval^d</i>	<i>cycles</i>	Cyc_r	8272	22314	22314	22314	40816
<i>End-of-life cost^c</i>	<i>%</i>	F_{EOL}	0%	0%	0%	0%	0%
<i>Discount rate^c</i>	<i>%</i>	DR	8%	8%	8%	8%	8%
<i>Round-trip efficiency^e</i>	<i>%</i>	η_{RT}	75%	79%	82.7%	85%	75%
<i>Self-discharge^c</i>	<i>%/day</i>	$\eta_{self\ idle}$	0%	0%	0%	3%	0%
<i>Cycle degradation^c</i>	<i>%/year</i>	Cyc_{deg}	0.000027 %	0.001%	0.001%	0.001%	0.0005%
<i>Cycle life^f</i>	<i>%</i>	$Cyclife$	8272	22314	22314	22314	40816
<i>Depth of discharge^e</i>	<i>%/day</i>	DOD	80%	85.44%	55.00%	13.40%	50.00%
<i>Daily time degradation^e</i>	<i>%/year</i>	$T_{deg, day}$	NA	0.05%	0.10%	3%	0.005%

<i>Annual time degradatine^e</i>	<i>Years</i>	<i>T_{deg}</i>	1.70%	16.70%	30.60%	36.50%	1.80%
<i>Shelf life^f</i>	<i>Seconds</i>	<i>T_{shelf}</i>	12.8	1.22	0.61	0.61	10.4
<i>Response time^c</i>	<i>years</i>		< 10	< 10	< 10	< 10	< 10
<i>Construction time^c</i>		<i>T_c</i>	0.5	0.5	0.5	0.5	0.5
<i>Reference Sources</i>			www.EnergyStorage.ninja	Joule, 2018, 2, 1894–1906	iScience, 2018, 3, 40–49	Energy Environ. Sci., 2020, 13, 2839–2848	This work

[a] Investment cost-Power includes power cost (C_p , USD kW⁻¹), balance-of-plant cost (C_{bop} , USD kW⁻¹), and additional cost (C_{add} , USD kW⁻¹). The power cost (C_p , USD kW⁻¹) was calculated based on Darling's group from:¹⁸

$$C_p = \frac{C_a}{\varepsilon_{sys,d} j_d V_d} = \frac{C_a R}{\varepsilon_{sys,d} U^2 \varepsilon_{V,d} (1 - \varepsilon_{V,d})} \quad (S1)$$

where c_a is the combined stack component cost per unit area (US\$ m⁻²). The component costs were included in **Supplementary Table 2**. j_d is the current density during discharge (kA m⁻²) and V_d is the discharge voltage (V). According to power conversion, pumping, heating and/or cooling, $\varepsilon_{sys,d}$ is the system efficiency during discharge and taken as a constant of 0.94. $\varepsilon_{V,d}$ is the discharge voltage efficiency, which means discharge voltage divided by open circuit voltage. R is the total area-specific resistance (ASR) of the cell (Ω m², measured in **Supplementary Fig. 49**); U is the open circuit voltage of the cell (V). The relationship between j_d , V_d , U and R is

$$V_d = U - j_d \times R \quad (S2)$$

The C_{bop} defined the costs of accessories for flow batteries, including heating/cooling equipment, state-of-charge, power managing electronics and pumps. The C_{add} term included other costs such as sales, administration, depreciation, warranty, research and development, profit margin, etc. for installation of the energy storage system. According to the Lu's groups, we here also took the corresponding averaged upper and lower bound values (at 95% confidence) for AFBs as the value for C_{bop} (USD 202.5 kW⁻¹) and C_{add} (USD 87.5 kW⁻¹).³ Investment cost-Power for all AFBs are listed in **Supplementary Table 5**.

[b] Investment cost-Energy (C_E , USD kWh⁻¹) was calculated based on Darling's group:¹⁷

$$C_E = \frac{c_{chemical} + c_{tank}}{\varepsilon_{sys,d} \varepsilon_{q,rt} \varepsilon_{V,d}} \quad (S3)$$

where $c_{chemical}$ is the total chemical cost (USD) normalized by the achieved energy density (USD kWh⁻¹), c_{tank} is the total tank cost (US\$ L⁻¹, taken as USD 0.15 L⁻¹) normalized by the achieved energy density (USD kWh⁻¹).¹⁷ The bulk prices of chemicals are listed in **Supplementary Table 4**. $\varepsilon_{q,rt}$ is round-trip coulombic efficiency. $c_{chemical}$ combines both the costs of active materials and supporting salts. Investment cost-Energy for all AFBs is listed in **Supplementary Table 5**.

[c] Power and energy specific operation and maintenance (O&M) cost (C_{P-OM} and C_{E-OM}), replacement cost (C_{P-r}), fraction of end-of-life cost to investment cost (F_{EOL}), discount rate (r), self-discharge rate ($\eta_{self-dis}$), response time and construction time (T_c) were assumed the same as all vanadium redox flow batteries.¹⁰

[d]

[e] Round-trip efficiency (η_{RT}), cycle degradation rate (Cyc_{deg}), daily time degradation rate ($T_{deg,day}$), annual time degradation rate (T_{deg}), and depth of discharge (DOD) were determined from results obtained from each RFB.

[f] The cycle life (Cyc_{life}) and shelf life (T_{shelf}) could be modeled as the degradation of energy storage capacity to an end-of-life value of 80% relative to initial capacity ($Cap_{nom,E}$):¹⁰

$$Cyc_{Deg} \left[\frac{\%capacity}{cycles} \right] = 1 - \exp \left(\frac{\ln(0.8)}{Cyc_{life}} \right) = 1 - 80\% \left(\frac{1}{Cyc_{life}} \right) \quad (S4)$$

$$T_{Deg} \left[\frac{\%capacity}{year} \right] = 1 - 80\% \left(\frac{1}{T_{shelf}} \right) \quad (S5)$$

Supplementary Note: Regarding discounts, our model adopted a discounted cash flow approach consistent with commercial VRFB analyses (e.g., 8% annual rate, as referenced in the model of www.EnergyStorage.ninja), which inherently prioritizes near-term performance.

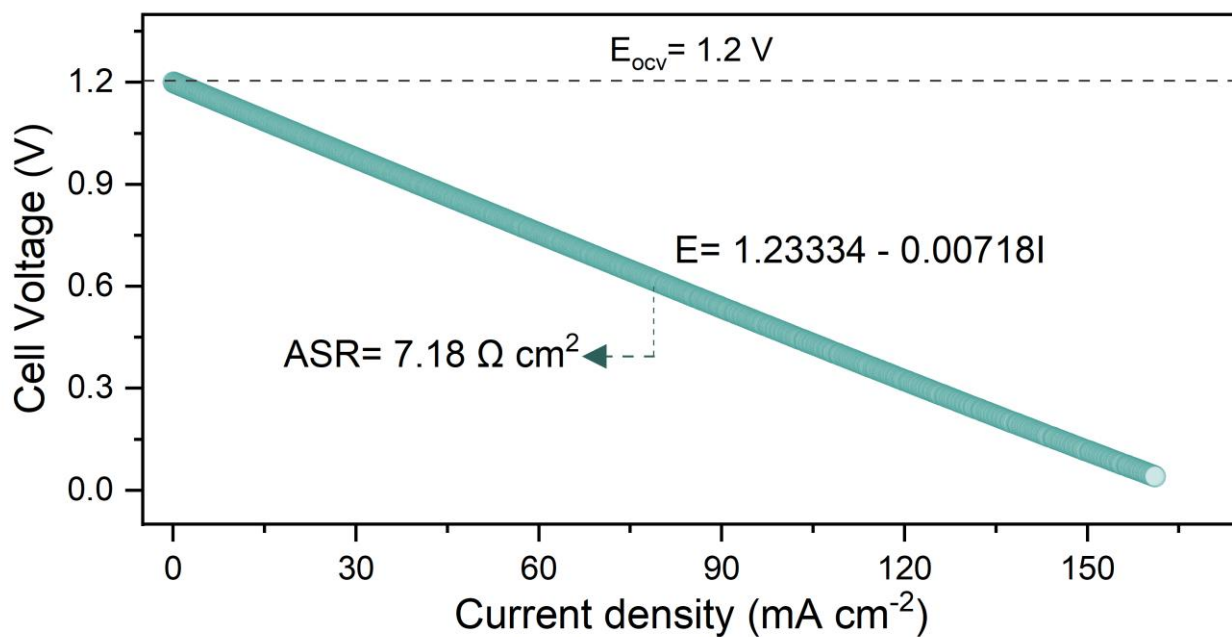
Application parameters: bulk energy storage

Discharge duration	Hours	T_d	1 - 23 h
Annual cycles	Cycles	Cyc_{pa}	180
Electricity cost	USD kWh ⁻¹	$Electricity-P$	0.02

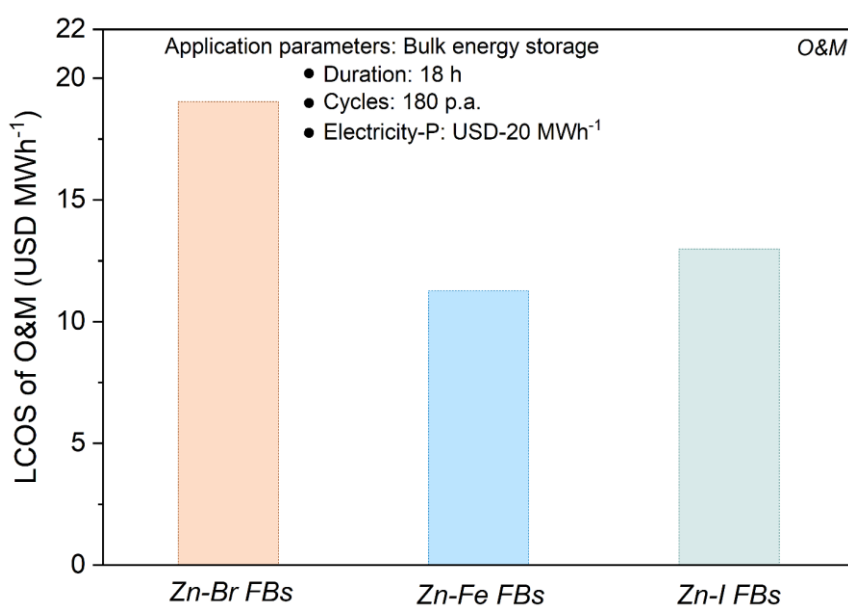
Supplementary Table 3 | Costs of reactor components for AFBs.

<i>Component</i>	<i>Cost (USD m⁻²)</i>	<i>Source</i>
<i>Graphite flow field</i>	<i>10^a</i>	<i>Ref^{18,19}</i>
<i>PTFE frame</i>	<i>15^a</i>	<i>Ref^{18,19}</i>
<i>Carbon felt</i>	<i>2^a</i>	<i>Ref^{18,19}</i>
<i>Fluorinated ion-exchange membrane (Nafion)</i>	<i>500</i>	<i>Ref^{18,19}</i>
<i>Fluorinated free membrane</i>	<i>25</i>	<i>Ref^{18,19}</i>
<i>PBI membrane</i>	<i>40</i>	<i>Ref^{19,20}</i>
<i>Gaskets, seals and manifolds total</i>	<i>1^a</i>	<i>Ref^{18,19}</i>

Supplementary Note: [a] Determined through averaging future state estimations at the year 2014.¹⁷



Supplementary Fig. 49 | Area-specific resistance (ASR) of the Zn-I FBs using ZMC-IMS membranes. The polarization curve of discharging Zn-I FBs (6 M KI + 3 M ZnBr₂ | 3 M ZnBr₂ + 3 M KCl) at 50% SOC.



Supplementary Fig. 50 | LCOS of O&M under different FB systems. Breakdown of the LCOS of O&M cost with a discharge duration of 18 h for Zn-Br FBs, Zn-Fe FBs, and Zn-I FBs.

Supplementary Note: As shown in **Supplementary Table 2**, the operational and maintenance (O&M) unit costs of energy and power are the same of 12 USD/kW-year and 1 USD/MWh across different FB systems, including AQ FBs, Zn-Fe FBs, Zn-Br FBs, and Zn-I FBs. Since the vanadium redox FB system is the most mature and commercialized, it serves as a key reference point for LCOS analysis. This similarity in O&M unit costs arises from common factors such as system complexity, maintenance requirements, and the necessity for regular monitoring and servicing, which do not significantly differ between various FB types. Therefore, different FB systems exhibit similar O&M unit costs. Finally, calculated by monetizing the energy storage systems, we obtained the total O&M costs for different Zn-based FB systems (Zn-Br, Zn-Fe, Zn-I FBs) under the simulation conditions of 8-hour duration, 180 annual cycles, and an electricity cost of 20 USD MWh⁻¹. As shown in **Supplementary Fig. 50**, The total O&M costs were 19.03, 11.26, and 12.99 USD MWh⁻¹, respectively. The Zn-Fe system has the lowest O&M cost due to its relatively inexpensive electrolyte design. However, the Zn-I system designed in this work offers a longer lifespan, contributing to a comparably low O&M cost as well. In contrast, the Zn-Br system has the highest O&M cost, primarily due to the expensive electrode and electrolyte prices, coupled with its lower cycling lifespan.

Supplementary Table 4 | The bulk price of chemicals.

<i>Chemical</i>	<i>Molecular weight (g mol⁻¹)</i>	<i>Price (USD kg⁻¹)</i>	<i>Source</i>
<i>KI</i>	<i>166</i>	<i>2.2</i>	<i>Ref^{17,19}</i>
<i>ZnBr₂</i>	<i>225.2</i>	<i>0.75</i>	
<i>KCl</i>	<i>74.55</i>	<i>0.71</i>	
<i>ZnCl₂</i>	<i>136.3</i>	<i>11</i>	
<i>KOH</i>	<i>56</i>	<i>0.82</i>	
<i>NaOH</i>	<i>40</i>	<i>0.34</i>	
<i>ZnO</i>	<i>81.39</i>	<i>1.3</i>	
<i>VO₂SO₄·4H₂O</i>	<i>235</i>	<i>10.2</i>	
<i>K₄Fe(CN)₆·3H₂O</i>	<i>422</i>	<i>2.15</i>	
<i>Na₄Fe(CN)₆·10H₂O</i>	<i>484</i>	<i>1.07</i>	
<i>anthraquinones</i>	<i>412</i>	<i>2.4</i>	
<i>1-Ethyl-1-methylpyrrolidinium bromide</i>	<i>194.11</i>	<i>200</i>	

Supplementary Table 5 | Parameters for estimating installed cost of AFBs.

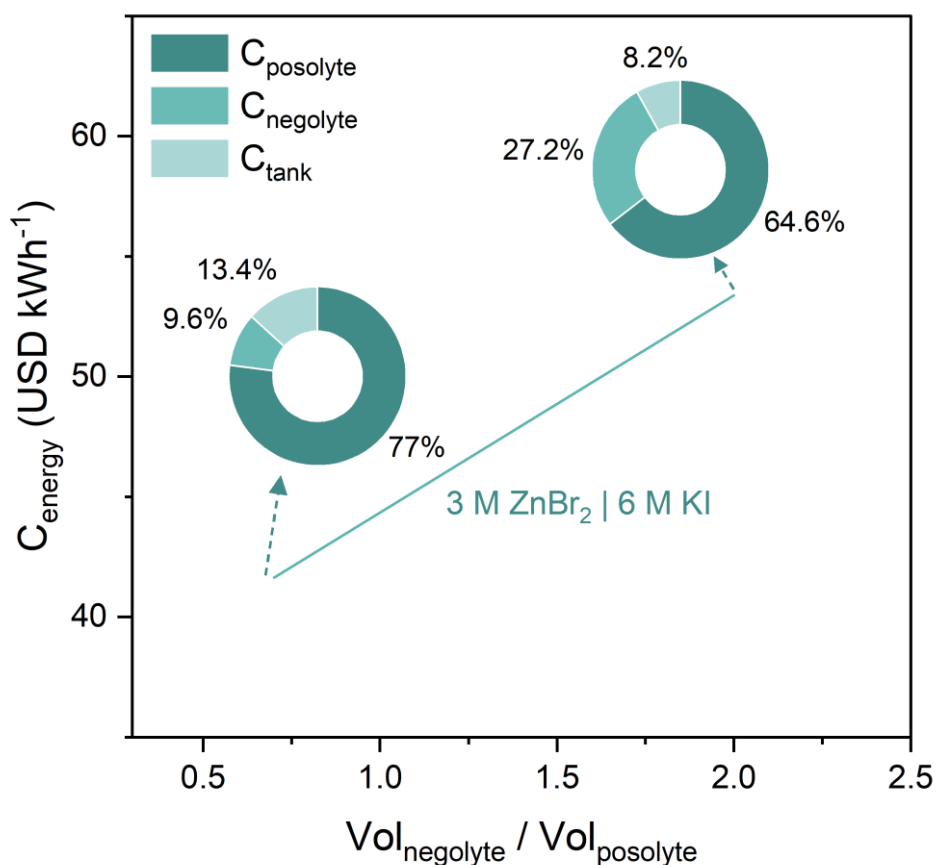
		<i>VRFBs</i>	<i>AQFBs</i>	<i>Zn-Fe FBs</i>	<i>Zn-Br FBs</i>	<i>Zn-I FBs (This work)</i>	<i>Zn-I FBs-future</i>
<i>Energy density</i>	<i>Wh L_{System}⁻¹</i>	<i>N.A</i>	17.2	12.72	6	27.66	33.32
<i>Posolyte cost</i>	<i>USD kWh⁻¹</i>		32.3	25.78	91.66	28.35	28.35
<i>Negolyte cost</i>	<i>USD kWh⁻¹</i>		17.6	5.56	91.66	7.14	1.82
<i>Tank cost</i>	<i>USD kWh⁻¹</i>		8.72	11.79	6.08	5.01	3.02
<i>Energy cost</i>	<i>USD kWh⁻¹</i>	<i>433–640 (ref.9)</i>	72.8	50.84	105.32	48.9	41.67
<i>ε_{v,d}</i>	%	<i>N.A</i>	99.94%	99.95%	97.2%	99.95	100%
<i>c_a</i>	<i>US\$ m⁻²</i>		112	112	127	542.1	67.1
<i>ASR</i>	<i>Ω cm⁻²</i>		1.5	1	1	11.8	5.1
<i>ε_{v,d}</i>	%		85.7%	87.6%	97.2%	88.13%	84.74%
<i>U</i>	<i>V</i>		1.05	1.05	1.8	1.18	1.18
<i>V_d</i>	<i>V</i>		0.9	1.63	1.67	1.04	1
<i>j_d</i>	<i>mA cm⁻²</i>		100	160	100	30	50
<i>Power cost (BOP cost and additional cost)</i>	<i>USD kWh⁻¹</i>	<i>472-527 (ref.9)</i>	422.3	341.8	348.1	622.5	492.1
<i>Prototype cell condition</i>	<i>Posolyte</i>	<i>N.A</i>	1.25 M Potassim ferrocyanide + 0.01 M KOH	60 mL 1 M Na ₄ Fe(CN) ₆ + 3 M KOH	60 mL 2 M ZnBr ₂ + 3 M KCl + 0.4 M MEP	6 M KI + 3 M ZnBr ₂	6 M KI + 3 M ZnBr ₂
	<i>Negolyte</i>	<i>N.A</i>	0.6 M 2,6-DBEAQ + 0.01 M KOH	60 mL 0.5 M Zn(OH) ₄ ²⁻ + 4 M NaOH	60 mL 2 M ZnBr ₂ + 3 M KCl + 0.4 M MEP	3 M ZnBr ₂ + 3 M KCl	3 M ZnBr ₂ + 3 M KCl
	<i>Membrane</i>	<i>Nafion</i>	<i>Fumasep E-620 (K)</i>	<i>PBI membrane</i>	<i>Porous membrane</i>	<i>IMS membrane</i>	<i>IMS with non-Nafion</i>
	<i>Volume ratio (V⁺:V⁻)</i>	<i>1:1</i>	<i>1.25:1.2</i>	<i>1:1</i>	<i>1:1</i>	<i>1:1</i>	<i>1:0.66</i>
	<i>Source</i>	www.EnergyStorage.ninja	<i>Joule</i> , 2018, 2, 1894–1906	<i>iScience</i> , 2018, 3, 40–49	<i>Energy Environ. Sci.</i> , 2020, 13, 2839–2848	<i>This work</i>	<i>Projected based on this work</i>

- [a] In practical implementation, the energy cost for AQFBs is computed based on the most optimal reported charge-balanced scenario rather than relying on demonstrated charge-excess outcomes.
- [b] Derived from **Supplementary Fig. 49**.
- [c] Optimization of the concentration ratio and volume ratio between the positive and negative electrolytes (**Supplementary Fig. 51**).
- [d] ASR is estimated as a summation of the resistance of KI + ZnBr₂, KCl + ZnBr₂ and membrane. The membrane resistance is taken from reference²¹, and the other two contributions were obtained from **Supplementary Fig. 52**. Assuming non fluorinated membrane with ASR $\approx 1.0 \Omega \text{ cm}^2$, future $\text{ASR} = R_{[\text{KI} + \text{ZnBr}_2]} + R_{[\text{KCl} + \text{ZnBr}_2]} + R_{\text{membrane}} = 1.64 + 2.459 + 1.0 \Omega \text{ cm}^2 = 5.1 \Omega \text{ cm}^2$.
- [e] Power cost is projected based on the optimized membrane cost, ASR and $\varepsilon_{V,d} = 0.8$.
- [N.A] Not applicable. The energy cost and power cost of VRFBs is reliably determined through empirical data obtained from literature.¹⁷ Therefore, the costs of these components are unnecessary.

Supplementary Note: The energy normalized installed cost ($C_{\text{installed}}$, USD kWh⁻¹) for Zn-I FBs was calculated based on Darling' groups:¹⁸

$$C_{\text{installed}} = C_E + \frac{(C_p + C_{\text{bop}} + C_{\text{add}})}{t_d} \quad (\text{S6})$$

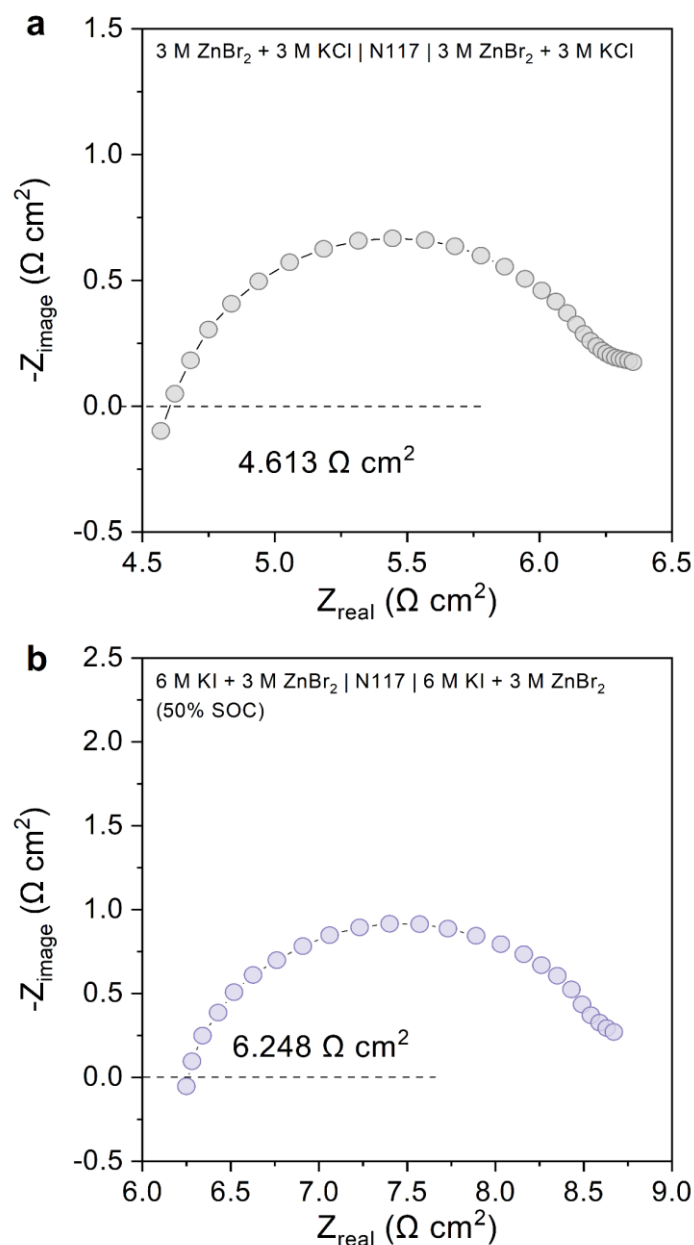
which is a combination of energy cost (C_E , USD kWh⁻¹), power cost (C_P , USD kW⁻¹), balance-of-plant cost (C_{bop} , USD kW⁻¹), additional cost (C_{add} , USD kW⁻¹) and the storage duration ($t_{d,h}$), which can be ascertained by calculating the ratio between the energy and power outputs of the battery system (E/P). Detailed parameters are comprehensively documented in **Supplementary Table 5-7**.



Supplementary Fig. 51 | Sensitivity analysis of the energy cost as a function of the volume ratio of negolyte and posolyte, assuming posolyte is the capacity limitation. The breakdown of the total energy cost of a Zn-I FB system (3 M ZnBr₂ | 6 M KI) with a volume ratio ($\text{Vol}_{\text{negolyte}}/\text{Vol}_{\text{posolyte}}$) of 0.66 (volume ratio to achieve charge balance, no excess of Zn²⁺) and 2.0 (with excess Zn²⁺) are shown in the donut charts.

Supplementary Note: Projection of future Zn-I FBs cost:

An approach to further reduce the energy cost of the Zn-I FBs is to minimize the amount of excess Zn-based negolyte in the posolyte-limited Zn-I FBs. The energy cost of the posolyte-limited Zn-I FBs (**Supplementary Fig. 51**) reduces as the volume ratio of negolyte to posolyte ($\text{Vol}_{\text{negolyte}}/\text{Vol}_{\text{posolyte}}$) decreases, *i.e.*, decreasing the excess amount of Zn²⁺. As shown in **Fig. 6c**, we forecasted the future total installed cost using the Zn-I FBs (6 M KI | 3 M ZnBr₂) performance by assuming no excess amount of negolyte (charge balance) for optimizing energy cost.



Supplementary Fig. 52 | EIS results of the symmetric cell. (a) EIS plot of 6 M KI + 3 M ZnBr₂ (50% SOC) symmetric cell separated by N117 membranes. (b) EIS plot of 3 M ZnBr₂ + 3 M KCl symmetric cell separated by N117 membrane.

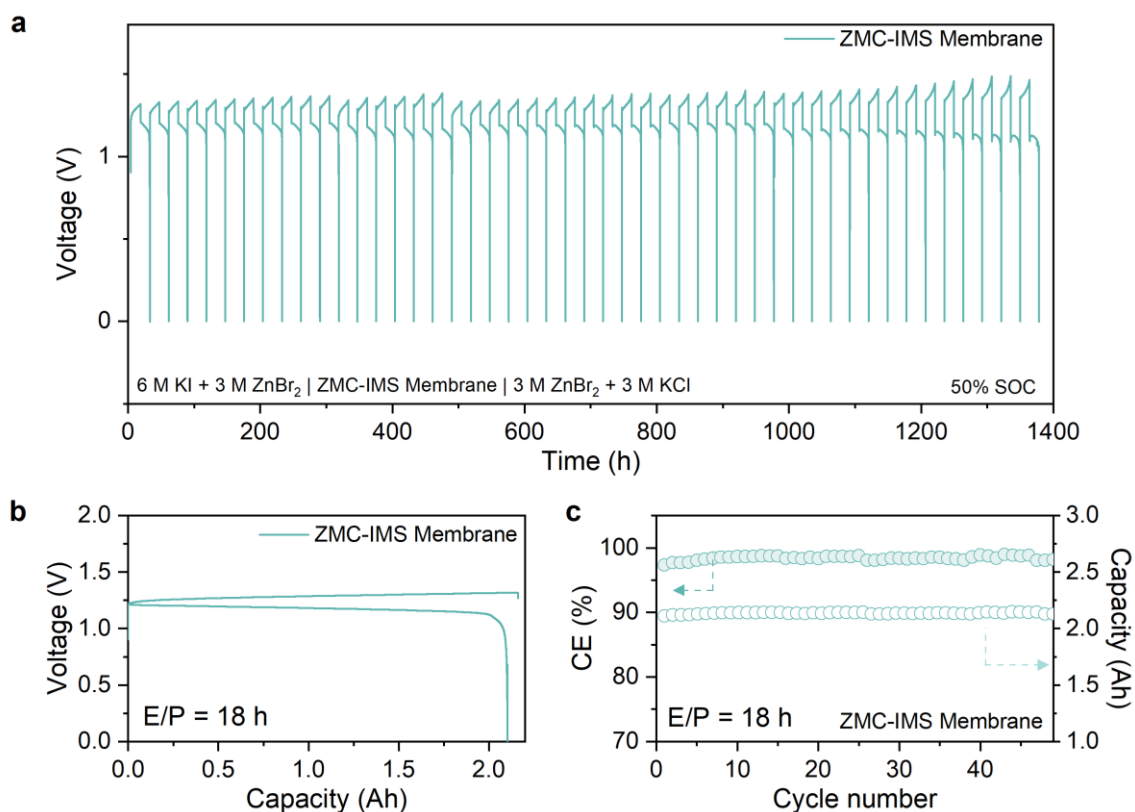
Supplementary Note: The ASR of N117 in K⁺ conductive electrolyte is tested and determined to be 1.33 $\Omega \text{ cm}^2$.²² Thus, the resistance of electrolytes (6 M KI + 3 M ZnBr₂) could be assessed to be 1.64 $\Omega \text{ cm}^2$ and the resistance of electrolytes (6 M KI + 3 M ZnBr₂) could be estimated to be 2.46 $\Omega \text{ cm}^2$ from the high frequency intercept of EIS plots, respectively.

Supplementary Table 6 | Present and future values of C_E , C_P , C_{bop} , and C_{add} for aqueous FBs

	<i>Zn-I FBs</i>	
	<i>Present</i>	<i>Future</i>
C_E (USD kWh ⁻¹)	48.9	41.64
C_P (USD kW ⁻¹)	5502.7	202.1
C_{bop} (USD kW ⁻¹)	202.5	202.5
C_{add} (USD kW ⁻¹)	87.5	87.5

Supplementary Table 7 | Installed cost breakdown of Zn-I FB systems at 270 kWh and 3600 kWh with fixed 15 kW power

	<i>System energy (kWh)</i>	<i>C_E ($\times 10^3$ USD)</i>	<i>C_P ($\times 10^3$ USD)</i>	<i>C_{bop} ($\times 10^3$ USD)</i>	<i>C_{add} ($\times 10^3$ USD)</i>	<i>C_{total} ($\times 10^3$ USD)</i>
<i>Zn-I FBs</i>	270	13.2	82.54	3.04	1.31	100.09
	3600	176.04	82.54	3.04	1.31	262.93
<i>Zn-I FBs-future</i>	270	11.24	3.04	3.04	1.31	18.63
	3600	149.9	3.04	3.04	1.31	157.29



Supplementary Fig. 53 | Long discharge duration cycling performance of Zn-I FBs flow cell with high E/P. 41 mL 6 M KI + 3 M ZnBr₂ | ZMC-IMS membrane | 41 mL 3 M ZnBr₂ + 3 M KCl Zn-I FBs flow cell at 120 mA with E/P=18 h (50% SOC) (The inset: representative voltage profile; CE and capacity retention over 1000 hours cycling).

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