Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supplementary Information

Sustainable Energy & Fuels

Micropores-in-Macroporous Gel Polymer Electrolytes for Alkali Metal Batteries

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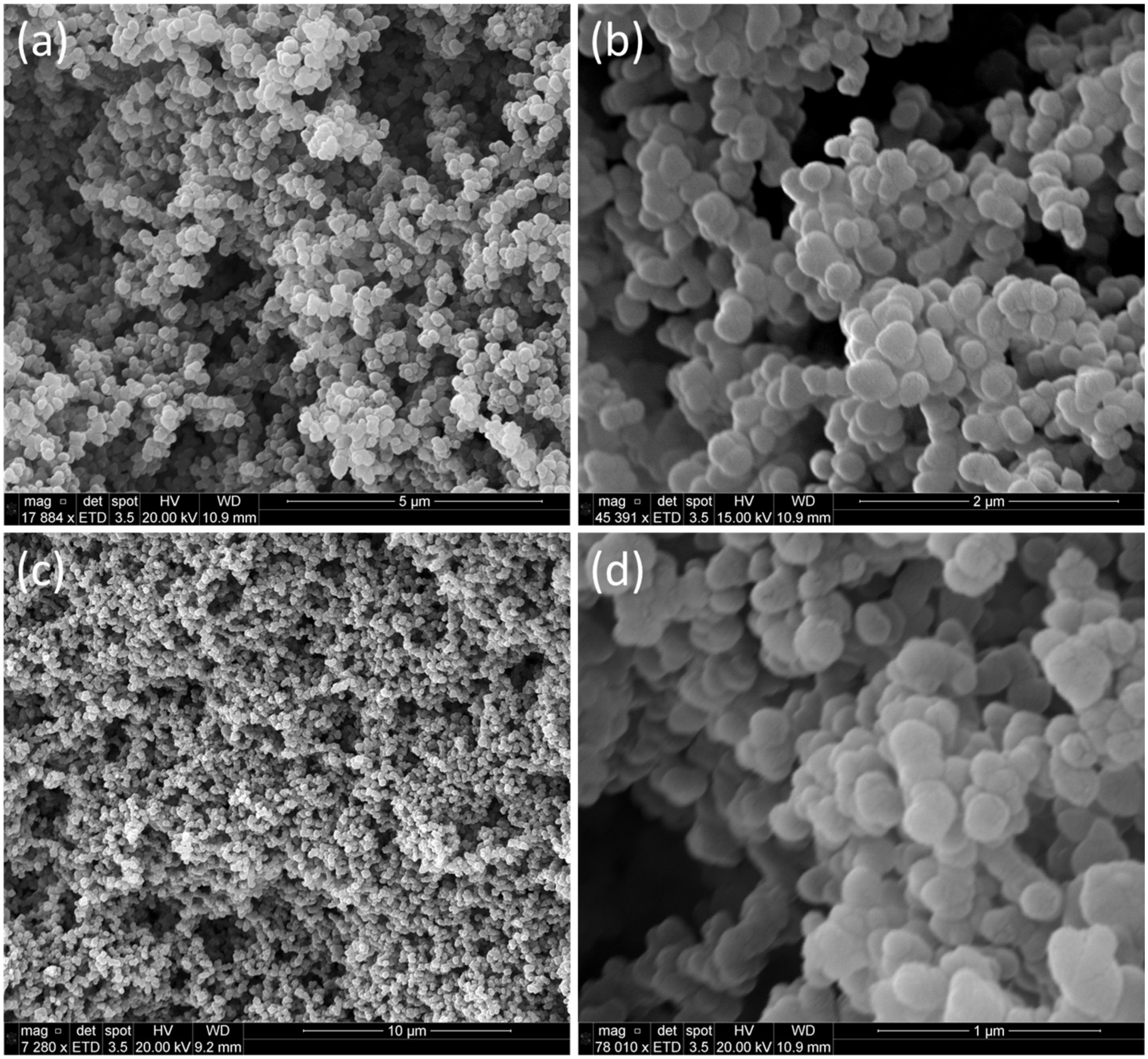

Figure S1. SEM images of (a, b) HCFu and (c, d) HCPy particles after being removed from the PH polymer host (see Experimental Section)

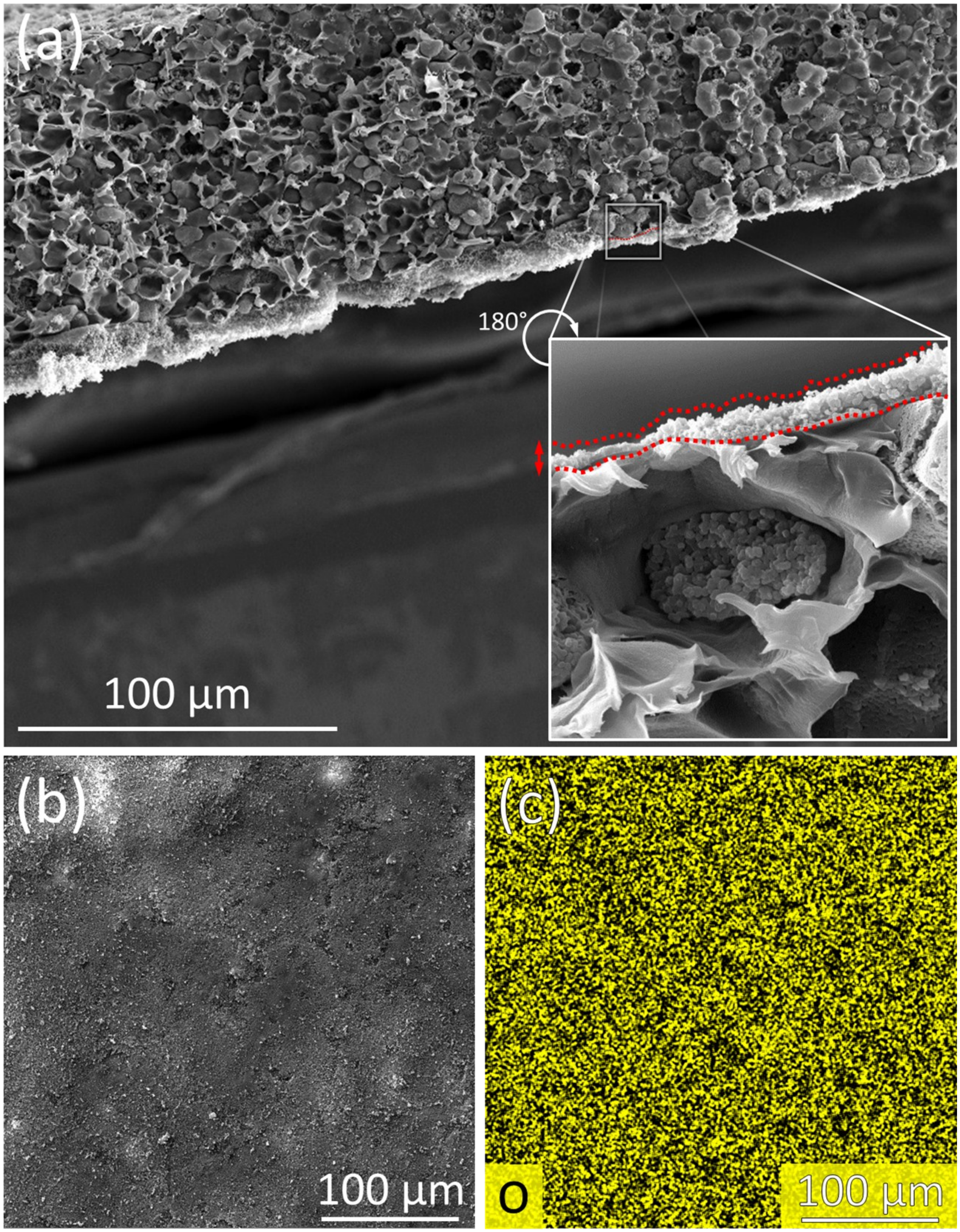

Figure S2. (a) Cross-sectional SEM image, (b) surface SEM image, and (c) oxygen-EDX image of HCFu-PH membrane

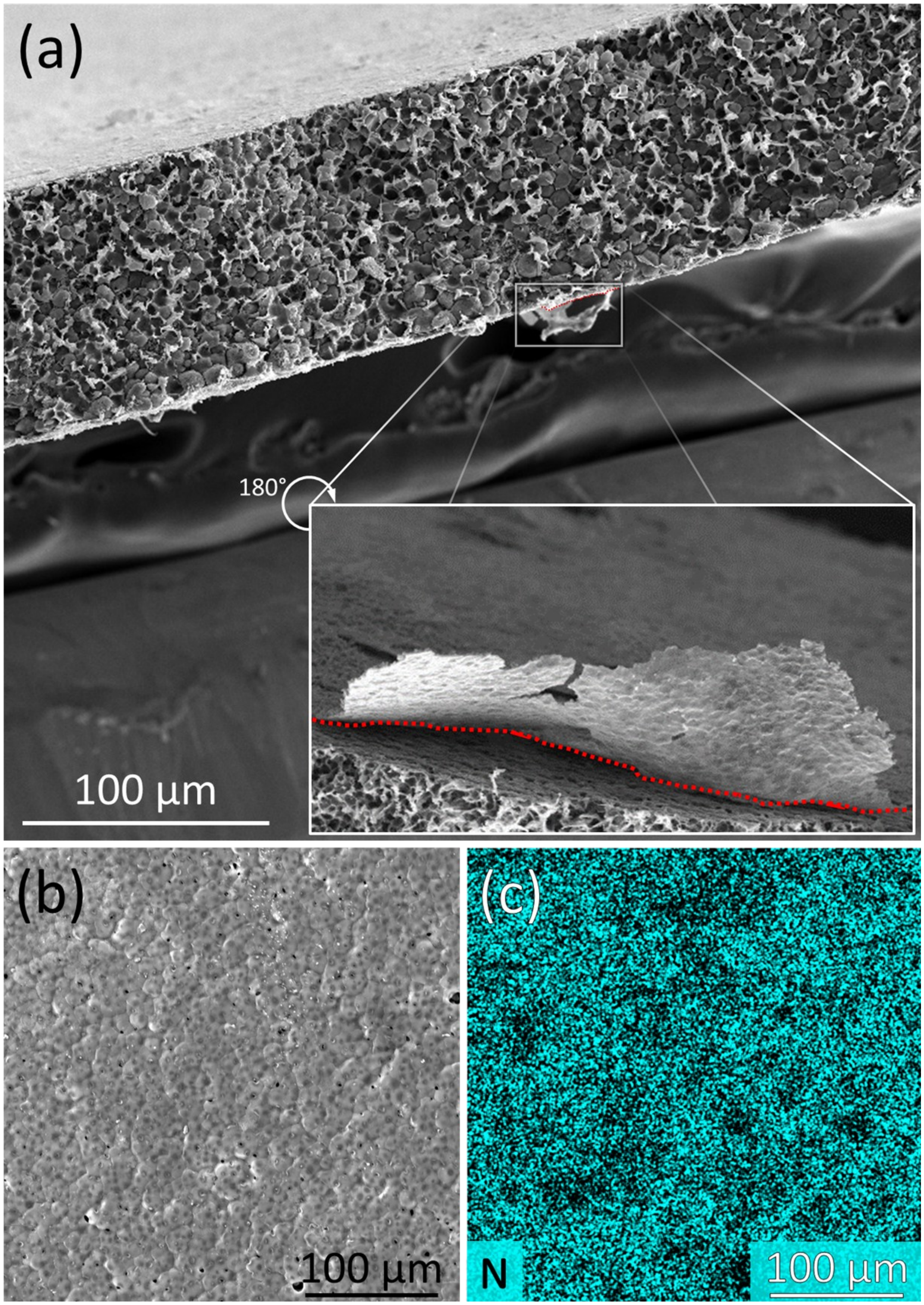

Figure S3. (a) Cross-sectional SEM image, (b) surface SEM image, and (c) nitrogen-EDX image of HCPy-PH membrane

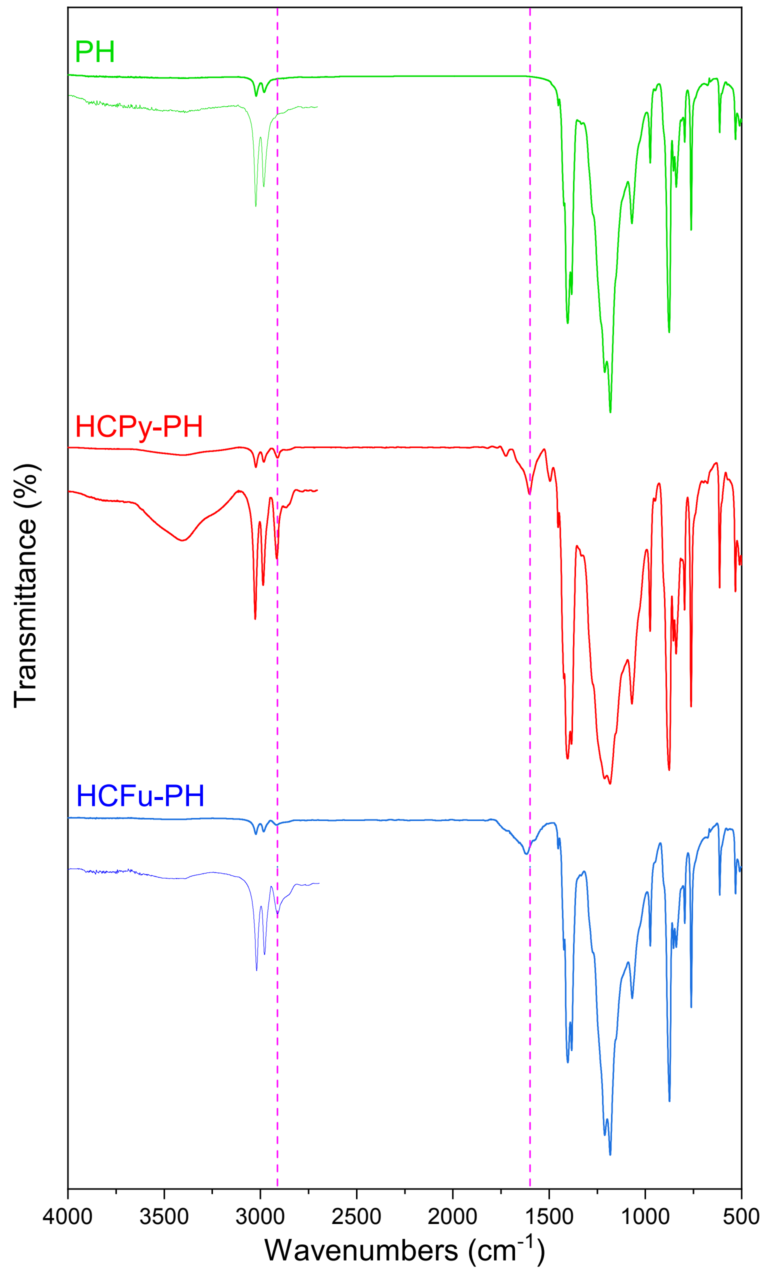

Figure S4. FTIR spectra of PH, HCPy-PH, and HCFu-PH membranes

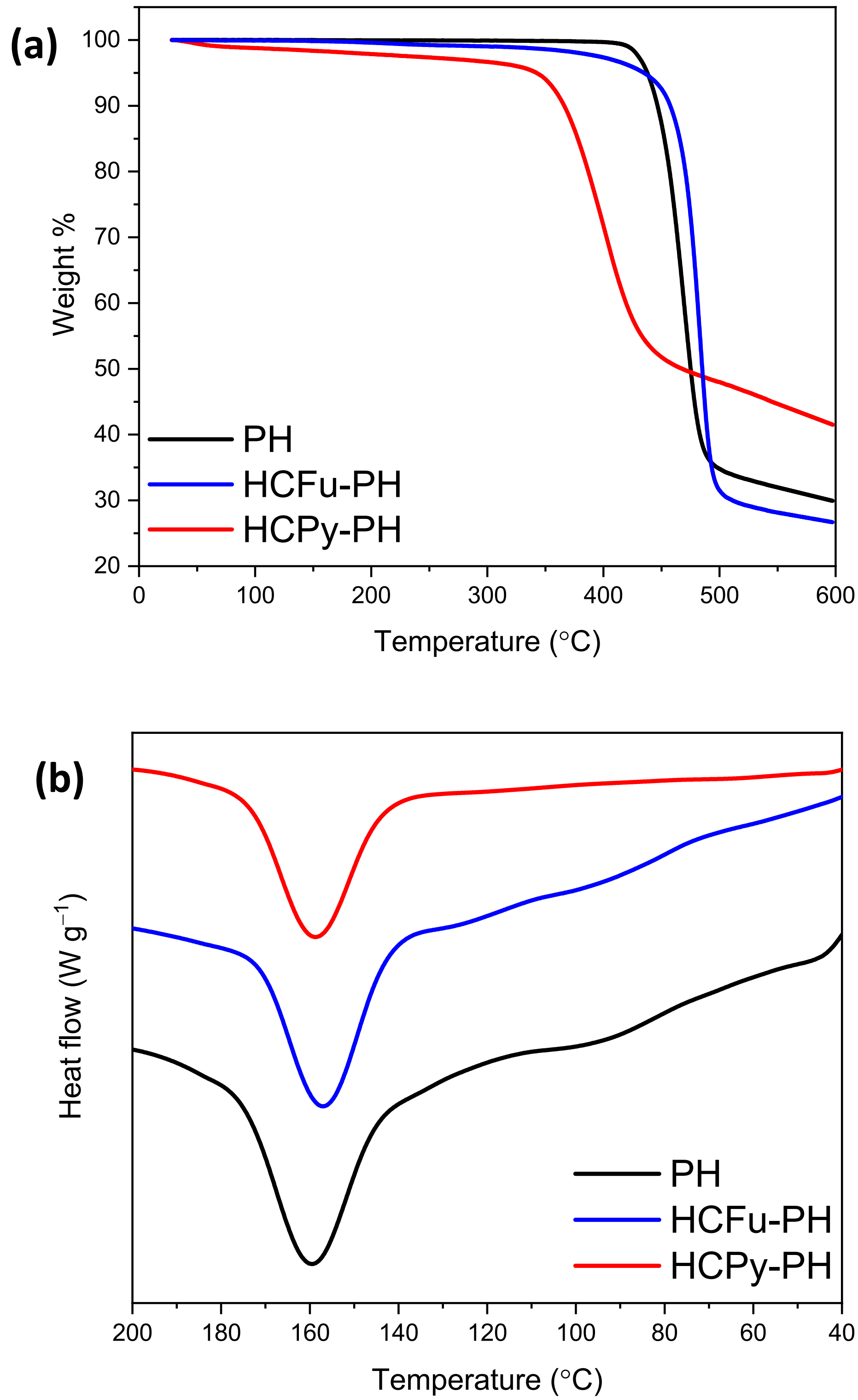

Figure S5. (a) TGA and (b) DSC curves of PH, HCPy-PH, and HCFu-PH membranes.

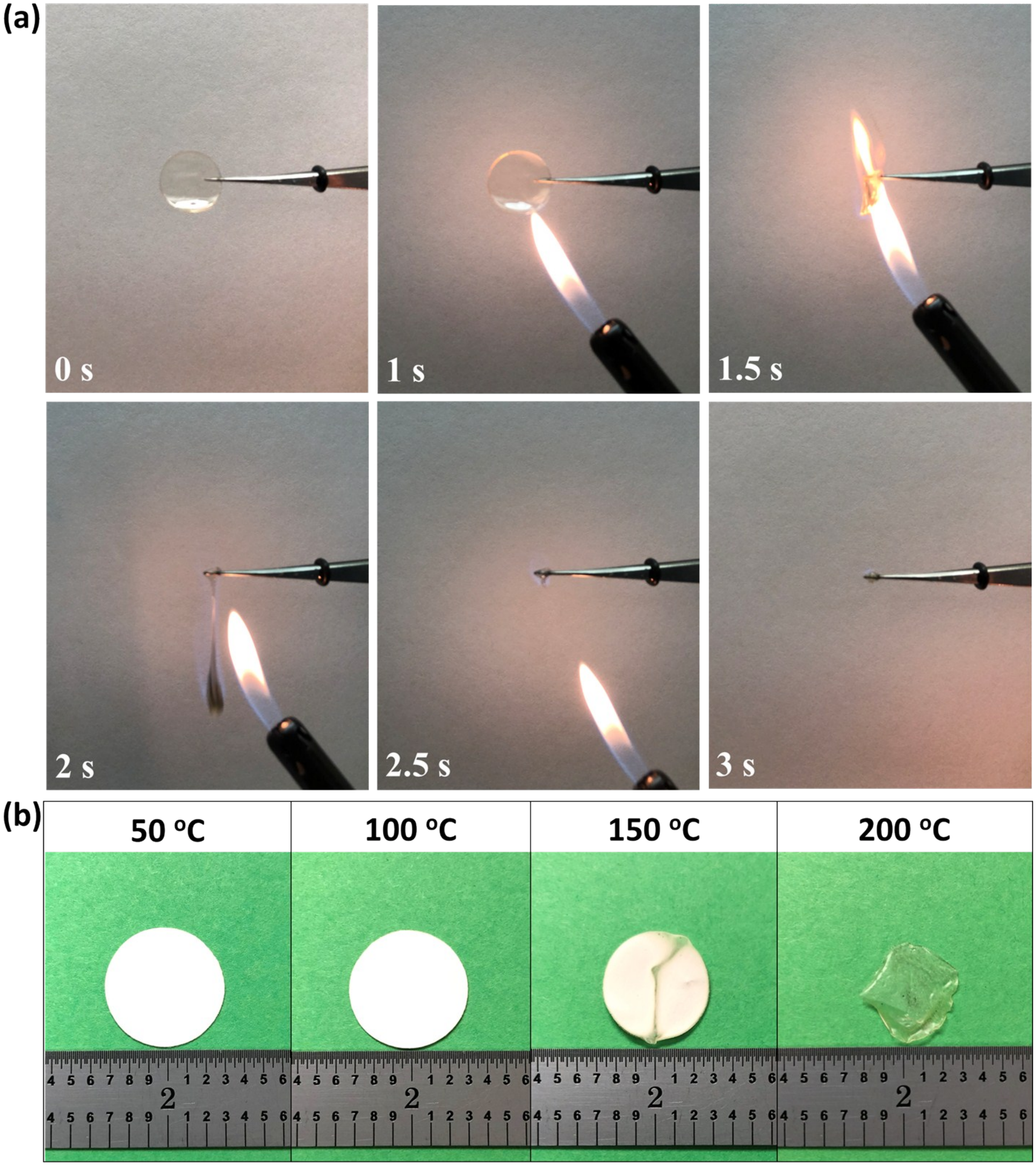

Figure S6. (a) Flame and (b) shrinkage tests for PH membrane.

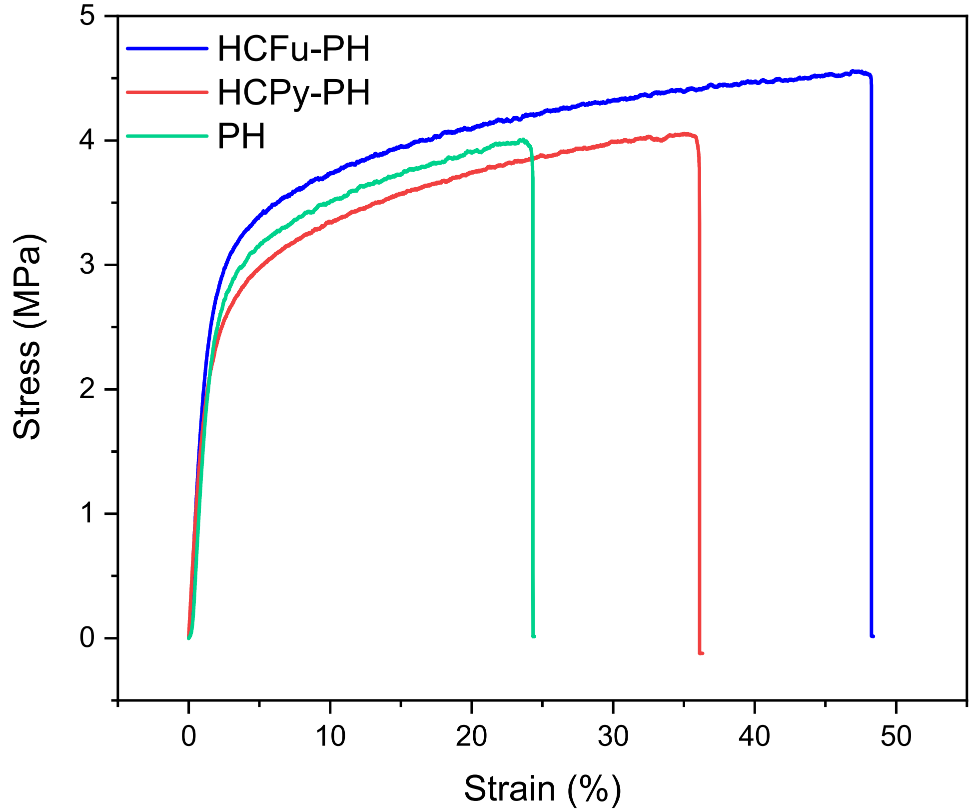

Figure S7. Stress-strain curves of PH, HCPy-PH, and HCFu-PH membranes.

Table S1. The performance comparison of recently reported GPEs for sodium and lithium batteries.

Cathode	Liquid Electrolyte	Anode	Polymer Electrolyte/Separator	Conductivity (S/cm)	ΔV	Reported full-cell performance	Ref.
$LiFePO_4$	$LiPF_6$ in EC/DMC/EMC	Li	PVDF-LiPVAOB[a]	2.60×10^{-4}	4.8	25 cycles at 0.2 C	1
$LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$	LiTFSI in Dimethyl Sulfoxide	Li	Cellulose Membrane	6.34×10^{-3}	4.6	50 cycles at 0.2 C (90% C.R.)[h]	2
$LiFePO_4$	$LiPF_6$ in EC/DMC	Li	P(EGDA-co-VC)/PVDF-HFP [b]	1.49×10^{-3}	4.2	100 cycles at 0.3 C	3
$Li_{1.18}Co_{0.15}Ni_{0.15}Mn_{0.52}O_2$	$LiPF_6$ in EC/DMC/EMC	Li	BN[c]-(PVDF-HFP)	4.10×10^{-4}	–	300 cycles at 0.5 C (79% C.R.)	4
$LiFePO_4$	$LiPF_6$ in EC/DMC	Li	(PEGDE+DEBA+DPPO)[d] @PVDF-HFP	2.36×10^{-3}	4.2	200 cycles at 0.3 C (99.3% C.R.)	5
$LiFePO_4$	$LiPF_6$ in EC/DMC/ EMC/DEC/2 wt% VC	Li	Al-doped $Li_{6.75}La_3Zr_{1.75}Ta_{0.25}O_{12}$ @PVDF-HFP	0.74×10^{-3}	–	500 cycles at 2 C	6
$LiFePO_4$	$LiPF_6$ in EC/DEC	Li	PVDF-HFP- $Li_7La_3Zr_2O_{12}$	3.71×10^{-4}	4.65	200 cycles at 0.2 C (83.8% C.R.)	7
$LiCoO_2$	$LiPF_6$ in EC/DMC	Li	PEO/PMMA/P(VDF-HFP)/SiO_2	2.02×10^{-3}	5.3	100 cycles at 0.1 mA/cm^2 (99% C.R.)	8
$Na_3V_2(PO_4)_2F_3$	$NaPF_6$ in PC	Na	PEO/PMMA/P(VDF-HFP)/SiO_2	8.80×10^{-4}	4.9	100 cycles at 0.1 mA/cm^2 (93% C.R.)	8
$Na_3V_2(PO_4)_3$	$NaClO_4$ in EC/PC/ 5 wt% FEC	Na	Cross-linked MATEMP[e]	5.13×10^{-3}	5	1000 cycles at 1 C (81.6% C.R.)	9
$Na_{0.44}MnO_2$	$NaClO_4$ in PC/ 2 wt% FEC	Na	PVDF-HFP	1.91×10^{-3}	4	20 cycles at 0.1 C	10
$Na_3V_2(PO_4)_3$	$NaPF_6$ in EC/DMC/ 2 wt% FEC	Na	(cross-linked PEGDE-DPPO) @GF[f]	2.18×10^{-3}	4.8	2000 cycles at 1 C (95% C.R.)	11
$Na_3V_2(PO_4)_3$	$NaClO_4$ in EC/PC/ 5 wt% FEC	Na	Cross-linked MATEPP[g]	6.29×10^{-3}	4.9	10000 cycles at 5 C (69.2% C.R.)	12
$LiFePO_4$	$LiPF_6$ in EC/DEC	Li	HCFu-PH	6.4×10^{-3}	4.7	1000 cycles at 1 C (78% C.R.)	This work
$Na_3V_2(PO_4)_3$	$NaClO_4$ in PC/ 5 wt% FEC	Na	HCPy-PH	4.3×10^{-3}	4.5	1000 cycles at 1 C (94% C.R.)	This work

a. Lithium polyvinyl alcohol oxalate borate (LiPVAOB).
b. Poly (ethylene glycol) diacrylate-co-poly (vinylene carbonate) and PVDF-HFP.
c. Boron nitride (BN).
d. Cross-linked GPE based on poly (ethylene glycol) diglycidyl ether (PEGDE), diglycidyl ether of bisphenol-A (DEBA), and diamino-poly (propylene oxide) (DPPO).
e. Di(2-methylacryloyltrioxyethyl) methyl phosphonate (MATEMP)
f. Cross-linked poly(ethylene glycol) diglycidyl ether (PEGDE) and diamino-poly (propylene oxide) (DPPO) in the network of glass fiber (GF) membrane
g. Di(2-methacryloyltrioxyethyl)phenylphosphonate (MATEPP)
h. C.R.: Capacity retention

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