

Supporting information

Emerging Conjugated Radical Polymer Cathode with Ultra-Long Cycle Life for an Entire Polymer Rechargeable Battery

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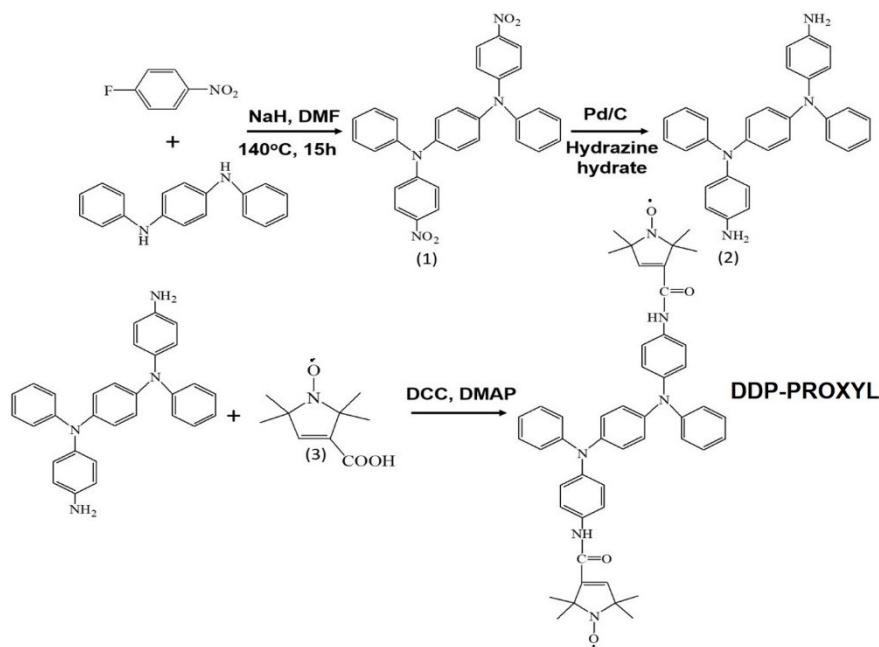


Figure S1 Synthetic route of the DDP-PROXYL

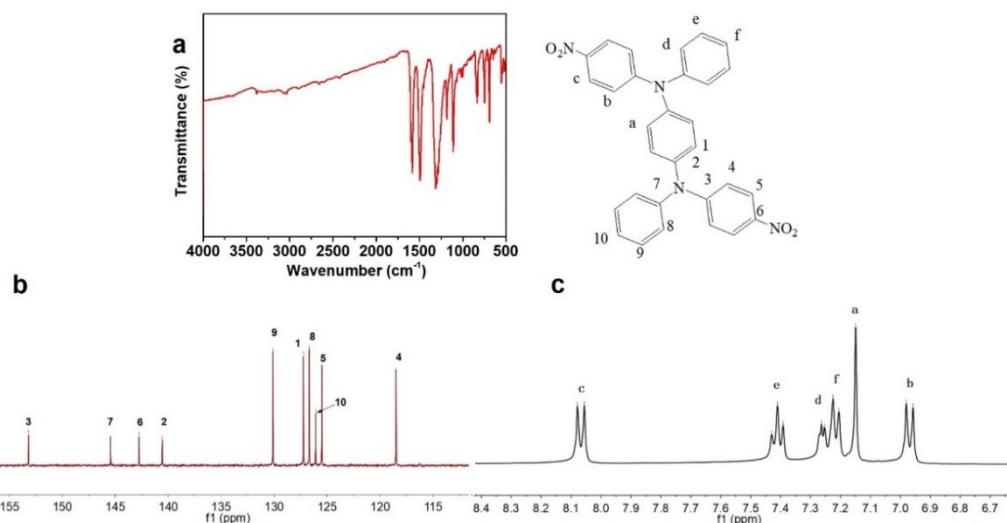


Figure S2 (a) FTIR, (b) ^{13}C NMR and (c) ^1H NMR spectra (400 MHz, $\text{DMSO}-d_6$) of *N,N'*-Bis(4-nitrophenyl)-*N,N'*-diphenyl-1,4-phenylenediamine (DDP- NO_2)

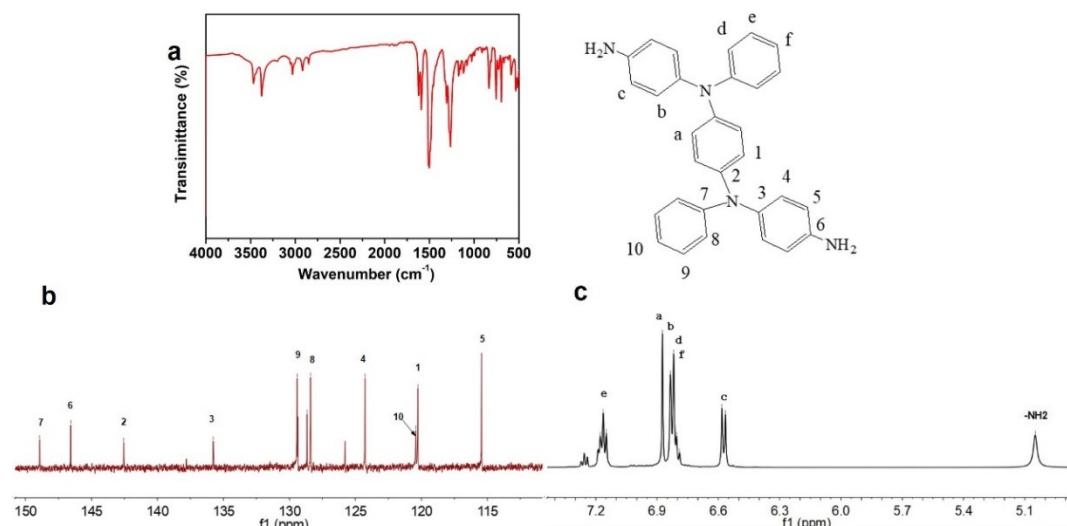


Figure S3 (a) FTIR, (b) ^{13}C NMR and (c) ^1H NMR spectra (400 MHz, $\text{DMSO}-d_6$) of *N,N'*-Bis(4-aminophenyl)-*N,N'*-diphenyl-1,4-phenylenediamine (DDP- NH_2)

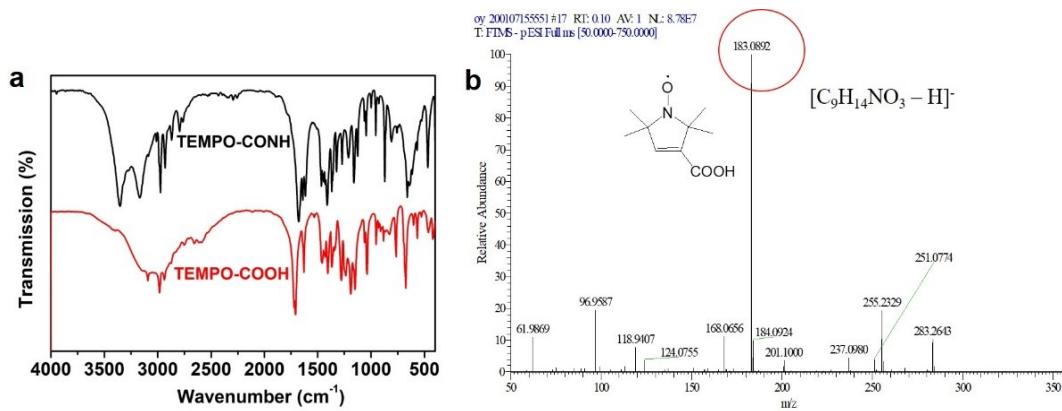


Figure S4 (a) FTIR, (b) MS (ESI) spectra of 3-carboxy-2,2,5,5-tetramethyl-3-pyrrolin-1-oxyl (PROXYL-COOH)

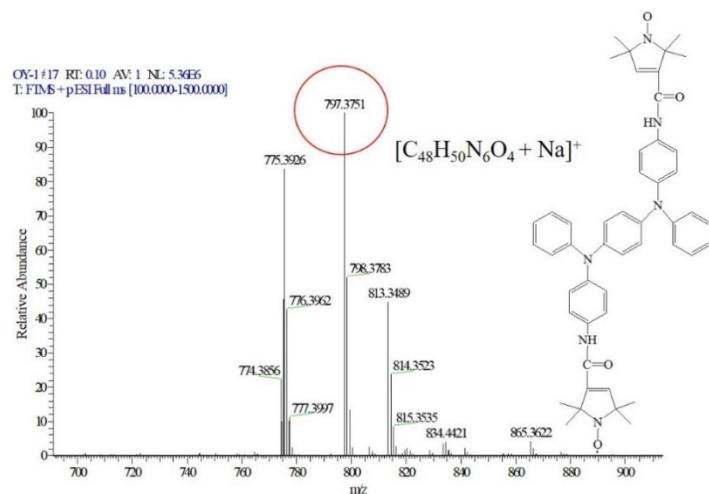


Figure S5 MS (ESI) spectra of DDP-PROXYL monomer:

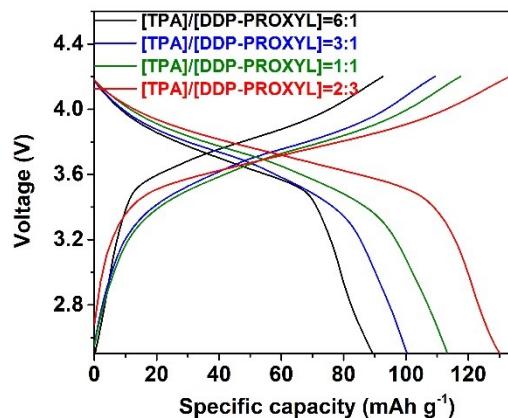


Figure S6 Galvanostatic charge/discharge curves of P(TPA-co-DDP-PROXYL) copolymer with different mole ratio of [TPA]/[DDP-PROXYL]

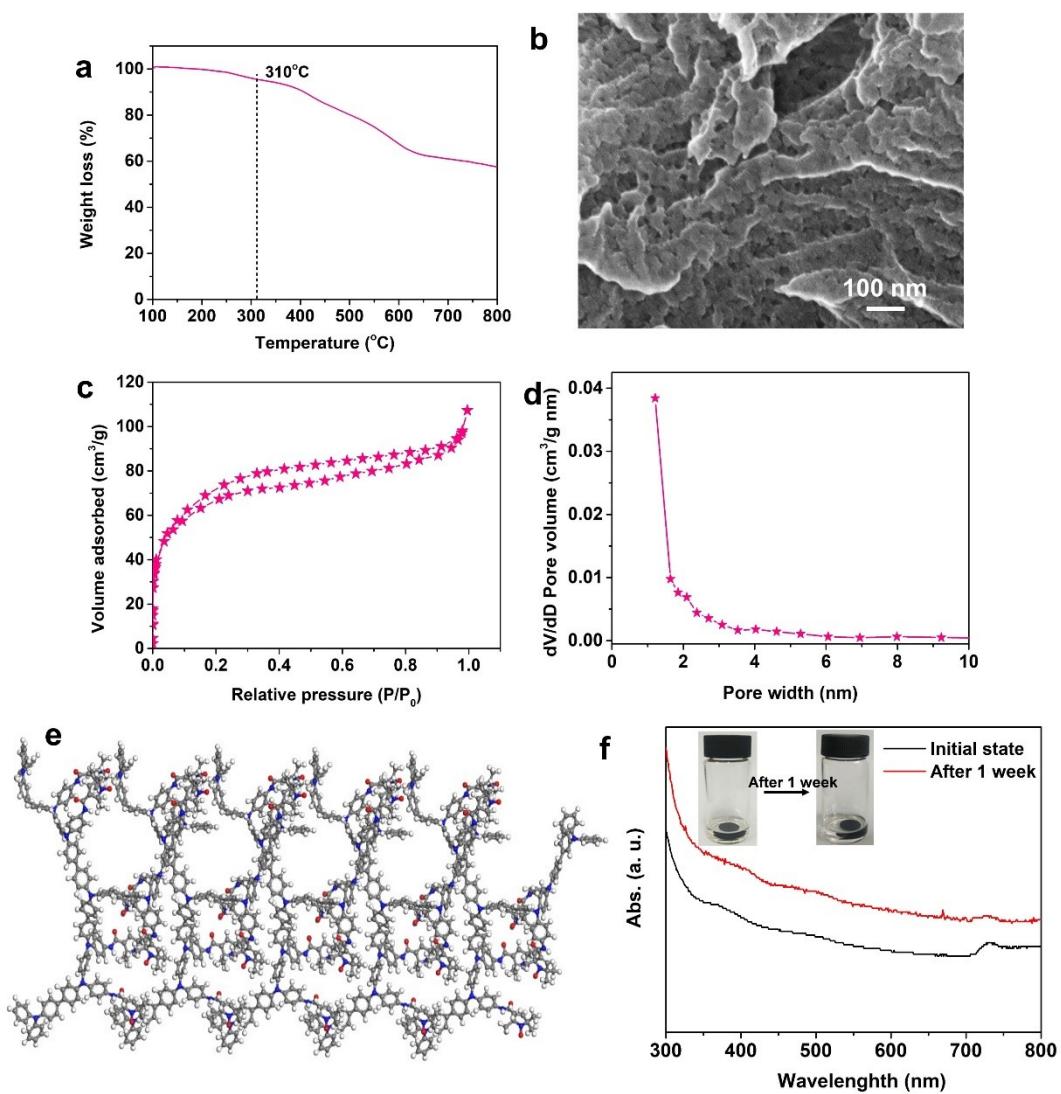


Figure S7 (a) TGA, (b) SEM image, (c) N_2 sorption isotherms at 77 K, (d) pore size distribution and (e) structure simulation of P(TPA-co-DDP-PROXYL) copolymer. (f) UV spectra of the electrolytes immersed by P(TPA-co-DDP-PROXYL) for 1 week.

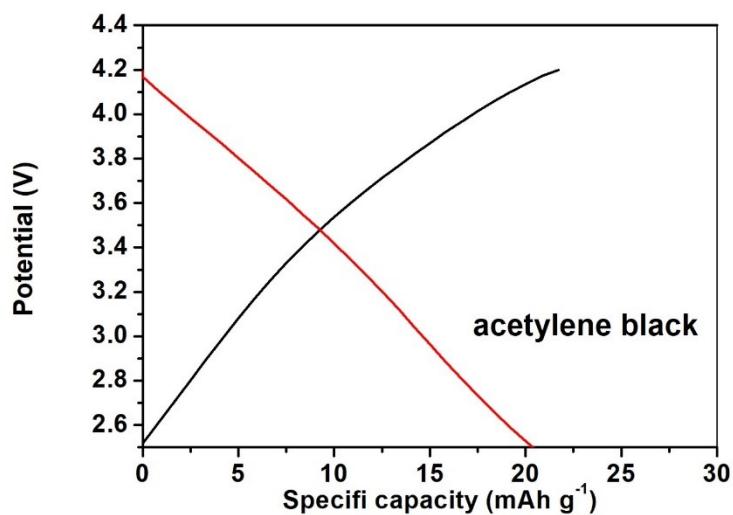


Figure S8 Specific capacity of acetylene black at 20 mA g^{-1} .

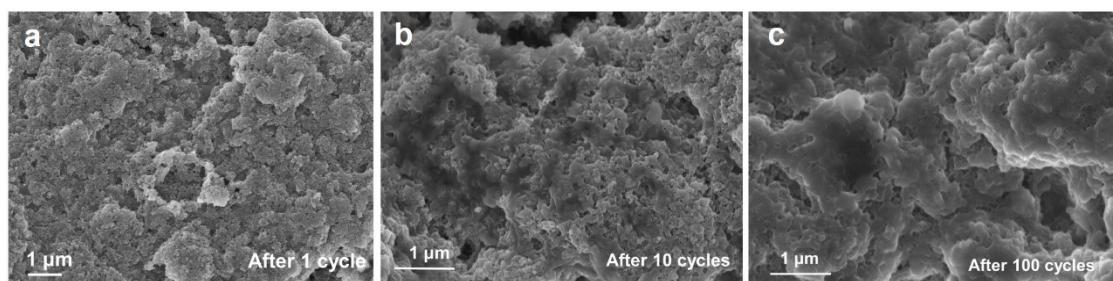


Figure S9 SEM images of P(TPA-co-DDP-PROXYL) electrode with different cycle times at 500 mA g^{-1}

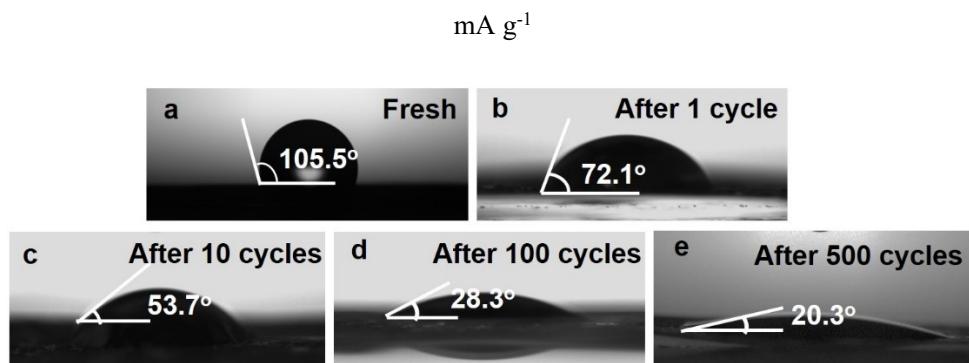


Figure S10 Water contact angle of electrode before and after cycling

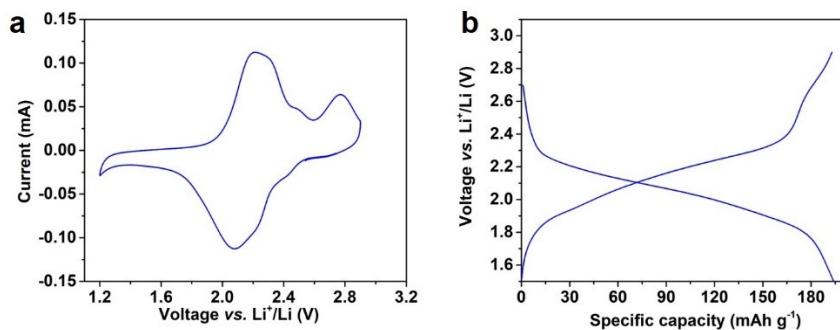


Figure S11. CV curve at the scan rate of 0.5 mV s^{-1} and (b) galvanostatic charge/discharge curves for the first three cycles at 20 mA g^{-1} of PAQS

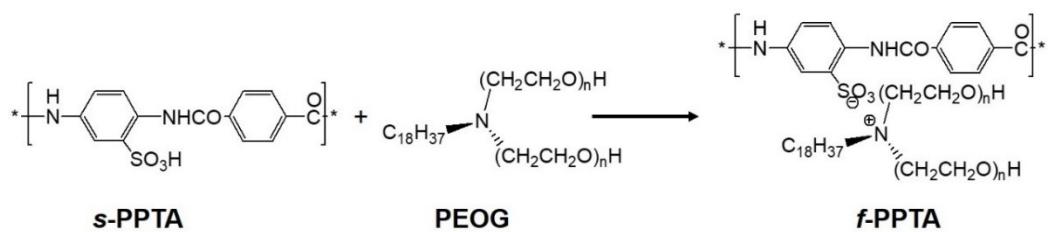


Figure S12. The synthesis route of the *f*-PPTA

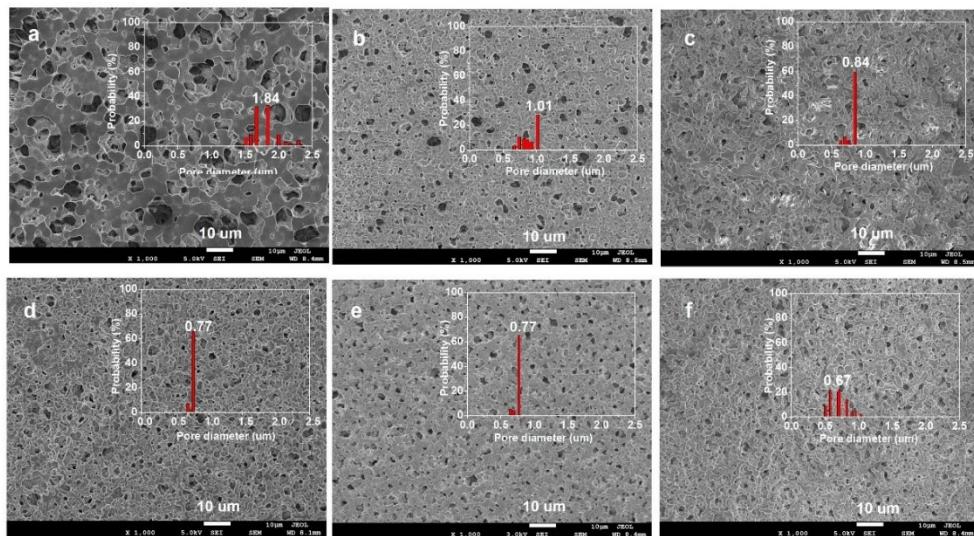


Figure S13 SEM images and pore size distributions of (a) PVDF-HFP, (b) PVDF-HFP/*f*-PPTA-1, (c) PVDF-HFP/*f*-PPTA-2, (d) PVDF-HFP/*f*-PPTA-3, (e) PVDF-HFP/*f*-PPTA-4 and (f) PVDF-HFP/*f*-PPTA-5

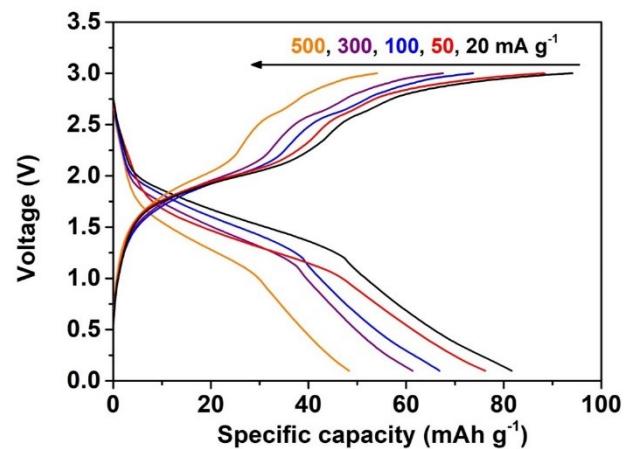
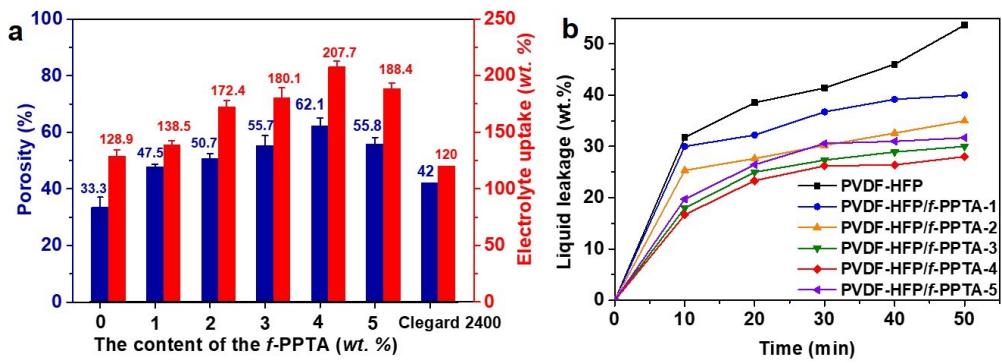


Figure S15 Charge/discharge profiles at different current density of the entire polymer rechargeable battery

Table S1. Electrochemical performance comparison of polymer cathode materials in LIBs

Samples	Reversibl e capacity (mAh g ⁻¹)	Rate capability (capacity rentantion (mAh g ⁻¹)@current density (mA g ⁻¹)	Cyclic life			Ref
			capacity rentantion (mAh g ⁻¹)@cycle number@current density (mA g ⁻¹)	Discharge voltage (V)		
PTDATA	133.1	90.9@500	98.2@100@20	3.6	1	
YPTPA	105.7	97.6@2000	92.9@1170@differe nt current densities	3.6	2	
PTTPAB	87	84@500	77@150@100	3.7	3	
DANI-PYR	113	58@1500	85@180@20 60@600@500	3.2	4	
PTPA-PO	134	90@500	121@100@20	3.8	5	
PGVS	104	40@510	40@500@510	3.15	6	
PPy-C-TEMPO	115	-	86@50@20	3.5	7	
PTMA-co-GMA	104	82@550	103@50@55	3.6	8	
Perylene	90	-	50@1800@-	3.8/3.3	9	
PTPAFc	100.2	90@500	-	3.6	10	
P1a	33	30@700	30@30000@700	3.6	11	
p-DPPZ	170	125@200	125@500@200	4.1/3.3	12	
PTMA	104.2	40@500	81@100@20	3.6	13	
P(TPA-co- DDP- PROXYL)	127.3	100.9@2000	93.8@3000@500 71.2@3000@2000	3.7	This work	

Table S2 Porosity, electrolyte uptake and ionic conductivity of P(VDF-HFP)/*f*-PPTA membrane and Clegard 2400

Sample	Ionic conductivity σ (mS cm ⁻¹) 20°C
P(VDF-HFP)	0.67 (+0.024)
P(VDF-HFP)/ <i>f</i> -PPTA-1	1.02 (+0.057)
P(VDF-HFP)/ <i>f</i> -PPTA-2	1.13 (+0.006)
P(VDF-HFP)/ <i>f</i> -PPTA-3	1.43 (+0.132)
P(VDF-HFP)/ <i>f</i> -PPTA-4	1.65 (+0.050)
P(VDF-HFP)/ <i>f</i> -PPTA-5	1.56 (+0.076)
Clegard 2400	0.60

References

1. C. Su, H. He, L. Xu, K. Zhao, C. Zheng and C. Zhang, *J. Mater. Chem. A*, 2017, **5**, 2701-2709.
2. C. Zhang, X. Yang, W. Ren, Y. Wang, F. Su and J.-X. Jiang, *J. Power Sources*, 2016, **317**, 49-56.
3. Z. Chen, W. Li, Y. Dai, N. Xu, C. Su, J. Liu and C. Zhang, *Electrochim. Acta*, 2018, **286**, 187-194.
4. C. J. Yao, J. Xie, Z. Wu, Z. J. Xu, S. Zhang and Q. Zhang, *Chem-Asian J*, 2019, **14**, 2210-2214.
5. J. Xiong, Z. Wei, T. Xu, Y. Zhang, C. Xiong and L. Dong, *Polymer*, 2017, **130**, 135-142.
6. T. Suga, H. Ohshiro, S. Sugita, K. Oyaizu and H. Nishide, *Adv. Mater.*, 2009, **21**, 1627-1630.
7. L. Xu, F. Yang, C. Su, L. Ji and C. Zhang, *Electrochim. Acta*, 2014, **130**, 148-155.
8. Shaoyang Wang, *ChemSusChem*, 2020, **13**, 2371–2378.
9. I. A. Rodriguez-Perez, C. Bommier, D. D. Fuller, D. P. Leonard, A. G. Williams and X. Ji, *ACS Appl. Mater. Inter.*, 2018, **10**, 43311-43315.
10. C. Su, Y. Ye, L. Xu and C. Zhang, *J. Mater. Chem.*, 2012, **22**, 22658.
11. P. Acker, L. Rzesny, C. F. N. Marchiori, C. M. Araujo and B. Esser, *Adv. Funct. Mater.*, 2019, **29**, 1906436.
12. G. Dai, X. Wang, Y. Qian, Z. Niu, X. Zhu, J. Ye, Y. Zhao and X. Zhang, *Energy Storage Mater.*, 2019, **16**, 236-242.
13. Y. Ou, Y. Zhang, Y. Xiong, Z. Hu and L. Dong, *Eur. Polym. J.*, 2021, **143**, 110191.