Supporting Information For

Flexible, self-healing and highly stretchable polymer electrolyte via quadruple

hydrogen bonding for lithium-ion batteries

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Scheme S1. Synthesis of UPy-based monomer (UPyMA) from 6-methylisocytosine and 2-

isocyanatoethyl methacrylate



Figure S1. ¹H NMR spectrum of the synthesized (2-(3-(6-methyl-4-oxo-1,4-dihydropyrimidin-2-

yl)ureido)ethyl methacrylate) (UPyMA).



Figure S2. ¹H *NMR* spectrum of the synthesized 4-cyanopentanoic acid dithiobenzoate (CPADB).



Figure S3. ¹H *NMR* spectrum of the poly(ethylene glycol) methyl ether methacrylate (PEGMA)



Figure S4. ¹*H NMR* spectrum of PEG10-UPy.



Figure S5. FT-IR spectra of the P(PEGMA) and PEG10-UPy.



Figure S6. DSC traces showing the $T_{\rm g}$ of four types of copolymers and the $T_{\rm g}$ and $T_{\rm m}$ of

P(PEGMA).



Figure S7. The photo of polymer of P(PEGMA) without UPyMA.



Figure S8. Dynamic frequency sweep for PEG5-UPy at 60 °C.



Figure S9. The electrochemical stability window of the shPE.



Figure S10. Chronoamperometry of the Li/P(PEGMA)/Li cell at a potential step of 10 mV. The

inset shows the AC impedance spectra before and after polarization at 60 °C.



Figure S11. The stress-strain profile of PEG5-UPy.



Figure S12. Demonstration of the P(PEGMA) attached to a metal surface to support 10 g of mass.



Figure S13. Rate performance of the LFP/shPE/Li cell at 60 °C.