Supporting Information

Crosslinked Hyperbranched Polyglycerol-Based Polymer Electrolytes for Lithium Metal Batteries

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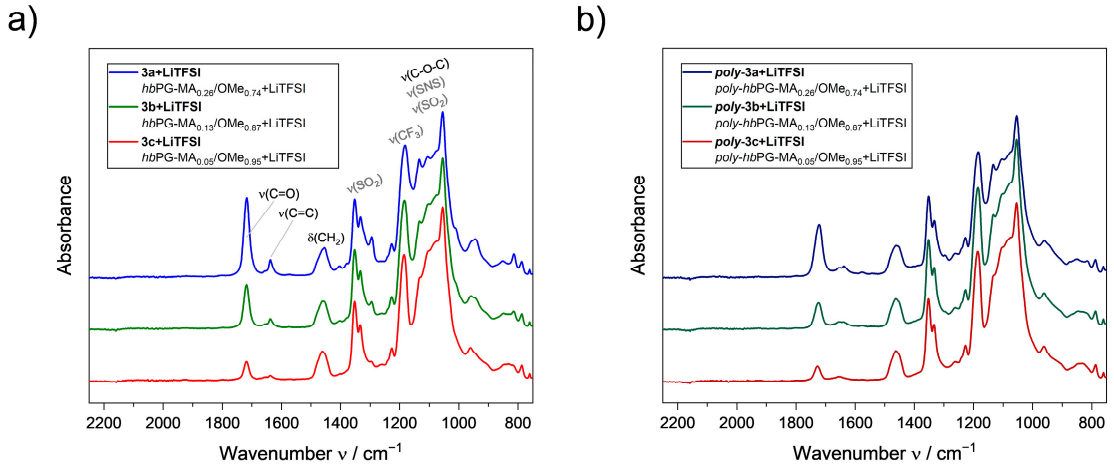
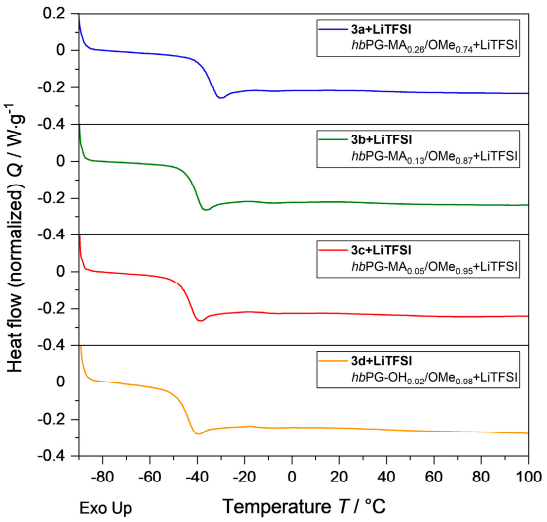
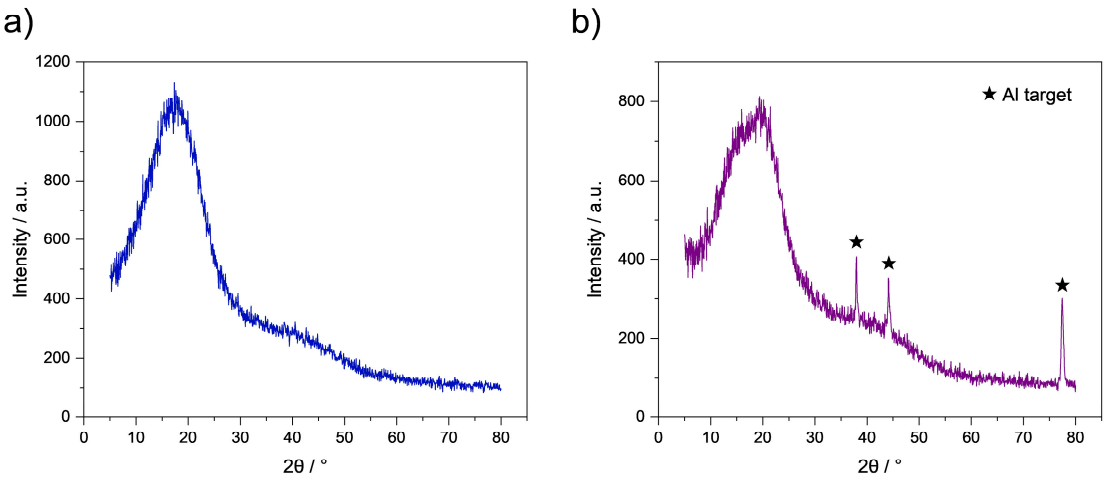


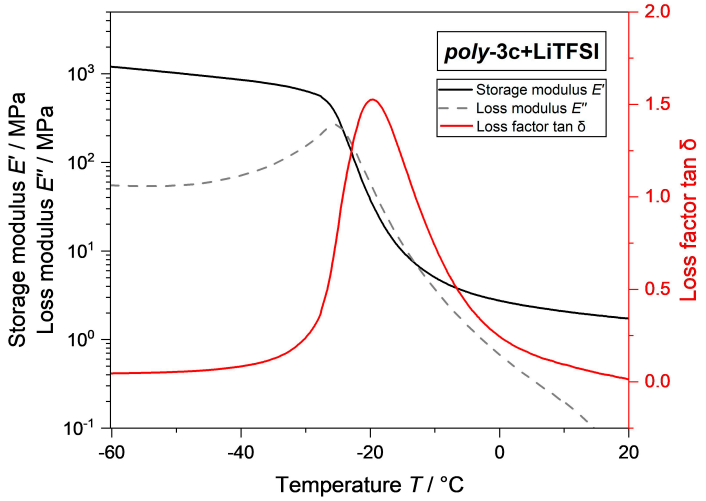
SI-Figure S1. (**a**) FTIR spectra of *hb*PG-MA$_x$/OMe$_y$+LiTFSI precursors. (**b**) FTIR spectra of UV cured *poly-hb*PG-MA$_x$/OMe$_y$ SPEs.



SI-Figure S2. DSC thermograms of *hb*PG-MA$_x$/OMe$_y$+LiTFSI precursors with various degree of methylation and methacrylation as well as the DSC thermogram of the *hb*PG-OH$_{0.02}$/OMe$_{0.98}$+LiTFSI electrolyte.



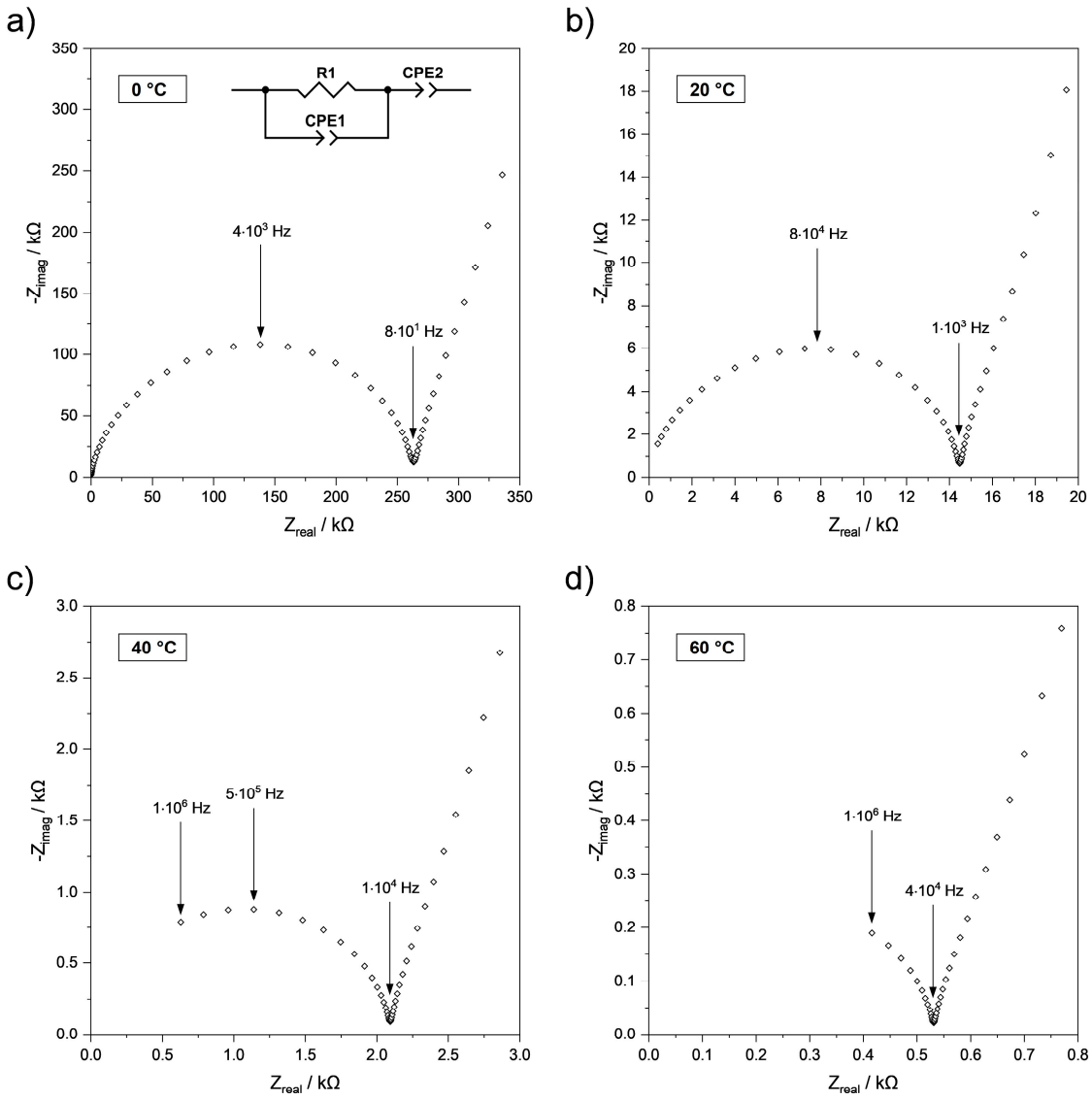
SI-Figure S3. XRD diffractograms of *poly-hb*PG-MA$_{0.05}$/OMe$_{0.95}$+LiTFSI measured (**a**) on a glass sample holder and (**b**) on an Al target in an inert sample chamber.



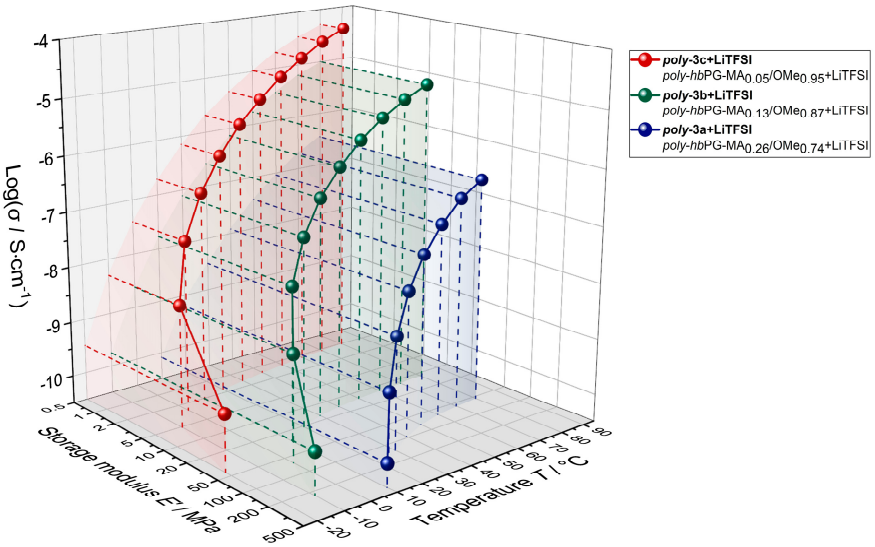
SI-Figure S4. Section of a DMA measurement of ***poly*-3c+LiTFSI** with displayed loss factor (tan δ).

SI-Table S1. Estimation of the shear modulus (G) based on the experimentally determined storage modulus (E') at 25 °C assuming a Poisson's ratio (μ) of 0.5 for each sample.

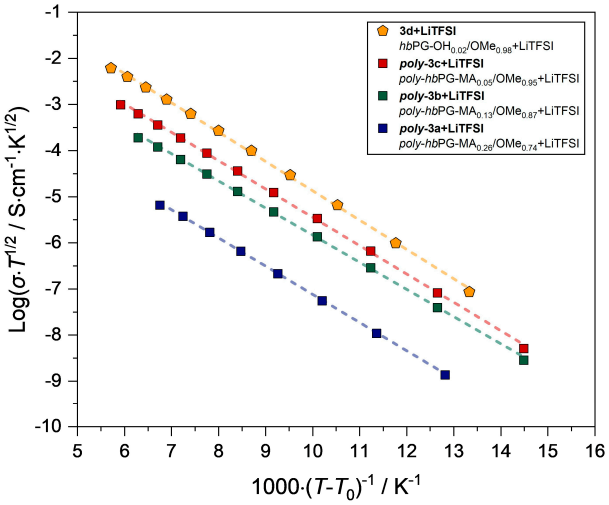
No.	E' / MPa	G / MPa
***poly*-3a+LiTFSI**	152 ± 28	≈ 51
***poly*-3b+LiTFSI**	22 ± 4	≈ 7
***poly*-3c+LiTFSI**	1.6 ± 0.4	≈ 0.5



SI-Figure S5. Nyquist plots of ***poly*-3c+LiTFSI** at different temperatures.



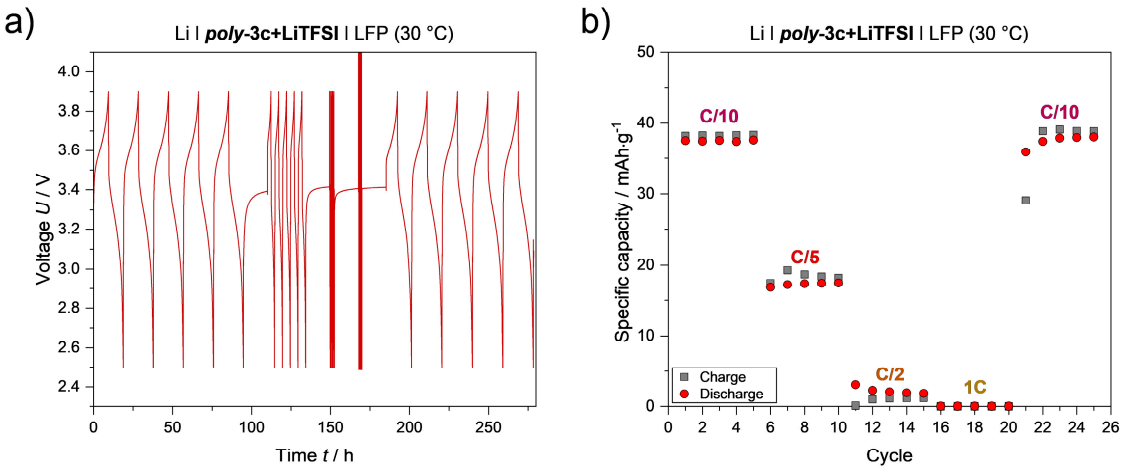
SI-Figure S6. Temperature dependence of ionic conductivity as well as mechanical strength of *hb*PG based polymer electrolytes (Li:O = 1:20) with different degrees of methylation and methacrylation.



SI-Figure S7. Temperature dependence of ionic conductivity of *hb*PG based polymer electrolytes (Li:O = 1:20) with different degrees of methylation and methacrylation using Vogel-Tammann-Fulcher (VTF) fitting curves.

SI-Table S2. Parameters determined through experimental data fitting using Vogel-Tammann-Fulcher (VTF) model.

No.	T_0 / K	Linear equation	E_p / kJ mol^{-1}	E_p / eV	R^2
***poly*-3a+LiTFSI**	205.15	–0.61x – 1.01	5.1	0.053	0.9998
***poly*-3b+LiTFSI**	194.15	–0.59x + 0.04	4.9	0.051	0.9995
***poly*-3c+LiTFSI**	184.15	–0.62x + 0.69	5.1	0.053	0.9994
3d+LiTFSI	178.15	–0.64x + 1.49	5.3	0.055	0.9994



SI-Figure S8. (**a**) Voltage profiles of a Li|***poly*-3c+LiTFSI**|LFP cell at different C-rates at 30 °C. (**b**) Specific charge/discharge capacities of a Li|***poly*-3c+LiTFSI**|LFP cell at different C-rates at 30 °C.