

Supporting Information

Accelerating Ion Transport by Dynamic Asymmetry of Alternating Polymer electrolytes

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Polymer Characterization

Gel Permeation Chromatography with Multi-Angle Laser Light Scattering (GPC-MALLS)

An Agilent 1260 Infinity II GPC with dRI detector, isocratic pump, and autosampler together with three MZ Analysentechnik columns, and a Wyatt Dawn Helios II detector were used for all measurements. Tetrahydrofuran (THF, 1 mL/min) was used as solvent. The dn/dc was determined under the assumption of 100% mass recovery. The Astra software package was used for data collection and treatment.

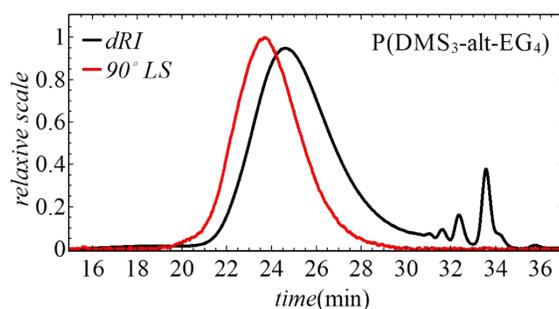


Figure S1: Differential Refractive Index (dRI) (black) and 90° Light Scattering (LS) (red) signal of the GPC-MALLS chromatogram on a relative scale for P(DMS₃-alt-EG₄).

¹H Nuclear Magnetic Resonance Spectroscopy

Spectra were collected on a Bruker AVIII-500 with a 5mm Prodigy TCI probe with deuterated benzene as solvent. The spectra are referenced to the residual solvent signal of benzene and chemical shifts reported in part per million.

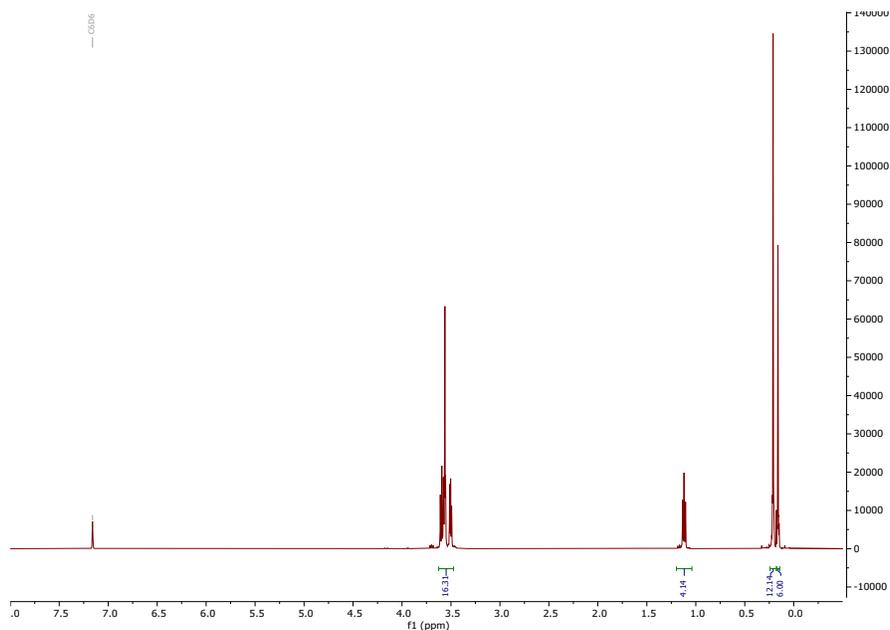


Figure S2: ¹H NMR of P(DMS₃-alt-EG₄) in deuterated benzene.

Differential Scanning Calorimetry (DSC)

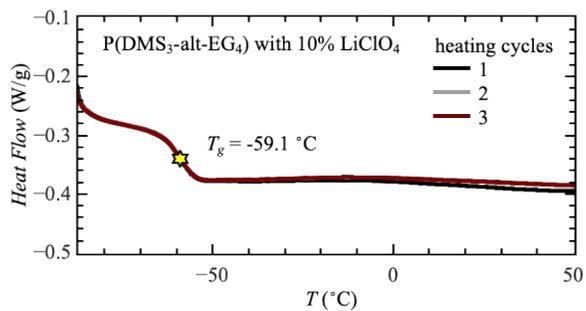


Figure S3: Heat flow as a function of temperature for P(DMS₃-alt-EG₄) with 10% LiClO₄ for all three heating cycles. Star symbol indicated the extracted glass transition temperature, T_g .

Broadband Dielectric Spectroscopy

Havriliak Negami Fitting Parameters for P(DMS₃-alt-EG₄)

Table S1: Fitting parameters for the best description of ϵ'' vs f of P(DMS₃-alt-EG₄) (i.e., **Figure 2a** in

main text)

T ($^{\circ}\text{C}$)	τ_{HN} (s)	β	γ	$\Delta\varepsilon$
- 85	0.18 ± 0.01	0.80 ± 0.01	0.38 ± 0.01	2.21 ± 0.01
- 80	$(2.57 \pm 0.05) \cdot 10^{-3}$	0.76 ± 0.01	0.38 ± 0.01	2.21 ± 0.01
- 75	$(1.38 \pm 0.02) \cdot 10^{-4}$	0.74 ± 0.01	0.36 ± 0.01	2.27 ± 0.01
- 70	$(1.44 \pm 0.01) \cdot 10^{-5}$	0.72 ± 0.01	0.36 ± 0.01	2.31 ± 0.01
- 65	$(2.41 \pm 0.03) \cdot 10^{-6}$	0.71 ± 0.01	0.36 ± 0.01	2.33 ± 0.01
- 60	$(5.88 \pm 0.24) \cdot 10^{-7}$	0.72 ± 0.01	0.34 ± 0.01	2.41 ± 0.04

Vft Fit Parameter for σ_{DC} (guides for the eye)

Table S2: Fitting parameter for creating the guides for the eye for Figure 7a in main text using VFT equation. No physical meaning behind.

[Li+]/[EG]	σ_0 (S/cm)	T_0 (K)	A
0	$(4.43 \pm 0.36) \cdot 10^{-8}$	145.01 ± 0.96	$- 1148.43 \pm 19.91$
0.02	$(1.12 \pm 0.04) \cdot 10^{-4}$	166.05 ± 0.39	$- 644.88 \pm 6.82$
0.04	$(2.34 \pm 3.02) \cdot 10^{-3}$	163.12 ± 13.08	$- 833.23 \pm 268.87$
0.06	$(6.05 \pm 0.54) \cdot 10^{-3}$	166.30 ± 0.75	$- 869.93 \pm 16.26$
0.08	$(1.30 \pm 0.10) \cdot 10^{-2}$	171.60 ± 0.61	$- 902.18 \pm 13.74$
0.10	$(1.76 \pm 0.20) \cdot 10^{-2}$	176.84 ± 0.89	$- 902.01 \pm 20.13$
0.12	$(2.04 \pm 12.0) \cdot 10^{-2}$	183.77 ± 58.56	$- 890.26 \pm 1149.98$
0.14	$(1.38 \pm 6.27) \cdot 10^{-2}$	187.26 ± 25.11	$- 849.20 \pm 651.91$

Quasi Elastic Neutron Scattering

Fitting parameter of $P(\text{DMS}_3\text{-alt-EG}_4)$ at $T = 30^\circ\text{C}$

Table S3: Fitting parameter for the best description of Intermediate scattering function, $I(Q,t)$, as a function of time, t , for $\text{P}(\text{DMS}_3\text{-alt-EG}_4)$ at $T = 30^\circ\text{C}$ (**Figure 9 Main Text**).

Q (\AA^{-1})	$\tau(Q)$ (ns)	$\beta(Q)$
0.4	0.28 ± 0.02	0.44 ± 0.01
0.6	0.079 ± 0.004	0.42 ± 0.01
0.8	0.028 ± 0.001	0.43 ± 0.01
1.0	0.013 ± 0.001	0.42 ± 0.01
1.2	$0.007 \pm 6.63 \cdot 10^{-5}$	0.41 ± 0.01
1.4	$0.0036 \pm 3.13 \cdot 10^{-5}$	0.42 ± 0.01
1.6	$0.0025 \pm 1.95 \cdot 10^{-5}$	0.42 ± 0.01

Fitting parameter of $P(\text{DMS}_3\text{-alt-EG}_4)$ with $[\text{Li}^+]/[\text{EG}]=0.09$ at $T = 30^\circ\text{C}$

Table S4: Fitting parameter for the best description of Intermediate scattering function, $I(Q,t)$, as a function of time, t , for $\text{P}(\text{DMS}_3\text{-alt-EG}_4)$ with $[\text{Li}^+]/[\text{EG}]=0.09$ at $T = 30^\circ\text{C}$ (**Figure 10(a) Main Text**).

Q (\AA^{-1})	$\tau(Q)$ (ns)	$\beta(Q)$ (constant)
0.4	2.84 ± 0.94	0.3
0.6	0.47 ± 0.06	0.3
0.8	0.13 ± 0.004	0.3
1.0	0.048 ± 0.001	0.3
1.2	0.019 ± 0.001	0.3
1.4	0.010 ± 0.0002	0.3
1.6	$0.0049 \pm 8.64 \cdot 10^{-5}$	0.3

Fitting parameter of $P(\text{DMS}_3\text{-alt-EG}_4)$ with $[\text{Li}^+]/[\text{EG}]=0.09$ and added toluene at $T = 30^\circ\text{C}$

Table S5: Fitting parameter for the best description of Intermediate scattering function, $I(Q,t)$, as a function of time, t , for $\text{P}(\text{DMS}_3\text{-alt-EG}_4)$ with $[\text{Li}^+]/[\text{EG}]=0.09$ and added toluene at $T = 30^\circ\text{C}$ (**Figure 14(a) Main Text**).

Q (\AA^{-1})	$\tau(Q)$ (ns)	$\beta(Q)$ (constant)
0.4	1.51 ± 0.035	0.38 ± 0.01
0.6	0.40 ± 0.006	0.35 ± 0.01
0.8	0.14 ± 0.002	0.33 ± 0.01
1.0	0.059 ± 0.001	0.32 ± 0.02
1.2	0.027 ± 0.0004	0.31 ± 0.01
1.4	0.0117 ± 0.0004	0.31 ± 0.01
1.6	0.0075 ± 0.0001	0.31 ± 0.01

Fitting parameter of $P(\text{DMS}_3\text{-alt-EG}_4)$ with $[\text{Li}^+]/[\text{EG}]=0.09$ and added acetonitrile at $T = 30^\circ\text{C}$

Table S6: Fitting parameter for the best description of Intermediate scattering function, $I(Q,t)$, as a function of time, t , for $\text{P}(\text{DMS}_3\text{-alt-EG}_4)$ with $[\text{Li}^+]/[\text{EG}]=0.09$ and added acetonitrile at $T = 30^\circ\text{C}$ (**Figure 14(b) Main Text**).

Q (\AA^{-1})	$\tau(Q)$ (ns)	$\beta(Q)$ (constant)
0.4	2.41 ± 0.07	0.34 ± 0.01
0.6	0.55 ± 0.01	0.33 ± 0.01
0.8	0.17 ± 0.002	0.33 ± 0.01
1.0	0.077 ± 0.0011	0.31 ± 0.02
1.2	0.034 ± 0.0005	0.30 ± 0.01
1.4	0.0174 ± 0.0007	0.29 ± 0.01
1.6	0.0088 ± 0.0001	0.28 ± 0.01

Fitting parameter of $P(\text{DMS}_3\text{-alt-EG}_4)$ with $[\text{Li}^+]/[\text{EG}]=0.09$ and more added acetonitrile at $T = 30^\circ\text{C}$

Table S7: Fitting parameter for the best description of Intermediate scattering function, $I(Q,t)$, as a function of time, t , for $\text{P}(\text{DMS}_3\text{-alt-EG}_4)$ with $[\text{Li}^+]/[\text{EG}]=0.09$ and more added acetonitrile at $T = 30^\circ\text{C}$ (**Figure S3**).

Q (\AA^{-1})	$\tau(Q)$ (ns)	$\beta(Q)$ (constant)
0.4	1.10 ± 0.03	0.40 ± 0.01

0.6	0.29 ± 0.01	0.38 ± 0.01
0.8	0.096 ± 0.002	0.38 ± 0.01
1.0	0.050 ± 0.001	0.34 ± 0.02
1.2	0.021 ± 0.0003	0.33 ± 0.01
1.4	0.011 ± 0.0005	0.32 ± 0.01
1.6	0.0073 ± 0.0002	0.30 ± 0.01

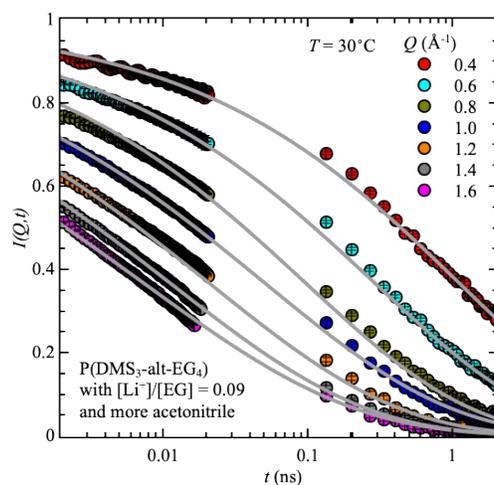


Figure S4: Intermediate scattering function, $I(Q,t)$, as a function of time, t , for P(DMS₃-alt-EG₄) with $[\text{Li}^+]/[\text{EG}]=0.09$ and more acetonitrile at $T = 30^\circ\text{C}$ for Q -values as indicated. Solid lines are the best description with equation (2).