Interplay of Chain Dynamics and Ion Transport on Mechanical

Behavior and Conductivity in Ionogels

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Figure S1 DSC traces of ionic liquid and P(DMAA-co-MAAc) matrix.



Figure S2 (a) Debonding force versus time curves on different substrates collected from probe-tack tests. (b) Adhesive energy W_{ade} of samples on different substrates, including glass, PDMS (Polydimethylsiloxane) and PMMA (Polymethyl methacrylate), and stainless steel.



Figure S3 Resistance changes rate of the flexible strain sensor base on ionogel-0.8 at (a) -10 °C, (b) 25 °C, and (c) 100 °C.



Figure S4 Rough comparison between this work and previously reported ionogel materials in terms of transparency,

conductivity, stretchability, mechanical strength, and toughness1-9.



Figure S5 Isothermal plots of real part, ε' , imaginary part, ε'' and dc conduction free dielectric loss, ε''_{der} of the complex permittivity (ε^*) as a function of applied frequency, respectively, for (a) ionogel-0.65, (b) ionogel-0.7, (c) ionogel-0.75, (d) ionogel-0.8, (e) ionogel-0.85 and (f) ionogel-0.9. The solid lines through the ε''_{der} plots show the fitting of experimental data to HN formalism.



Figure S6 Isothermal plots of real part, σ' and imaginary part, σ'' of the complex conductivity (σ^*) as a function of applied frequency, respectively, for (a) ionogel-0.65, (b) ionogel-0.7, (c) ionogel-0.75, (d) ionogel-0.8, (e) ionogel-0.85 and (f) ionogel-0.9. The solid lines through the σ' plots show the fitting of experimental data to RBM formalism.



Figure S7 Plots of the inverse of segment relaxation time $(1/\tau_s)$ versus hopping frequency (ω_H) of ionogel-m (m: 0.65-0.9).



Figure S8 Plots of dc conduction σ_{dc} vs hopping frequency ω_H of these ionogels. The dashed lines represent the linear fitting of ionogel-m (m: 0.65-0.9).

	Slope
ionogel-0.65	1.07
ionogel-0.7	1.14
ionogel-0.75	1.11
ionogel-0.8	1.26
ionogel-0.85	1.26
ionogel-0.9	1.23

Table S1 Slope of the plots of dc conduction σ_{dc} vs hopping frequency ω_H of ionogel-m (m: 0.65-0.9).



Figure S9 Normalized inverse of temperature dependent ionic conductivity ($^{\sigma_{dc}}$) using the Vogel temperature $T_0^{"}$ for all the ionogel-m (m: 0.65-0.9).

Table S2 WLF fitting constants C_1 and C_2 of temperature dependent the natural logarithmic plot of the horizontalshift factors a_T of ionogel-0.14-m (m: 0.65-0.9).

	C1	C2
ionogel-0.65	9.82	260.47
ionogel-0.7	10.56	160.83
ionogel-0.75	11.4	242.06
ionogel-0.8	10.93	231.17
ionogel-0.85	3.99	175.78
ionogel-0.9	1.33	131.44



Figure S10 Time-concentration horizontal shift factor a_m (black plots) and vertical shift factor b_m (red plots) as a function of ionic liquid contents m. Reference content m=0.65.



Figure S11 Loss factor $Tan \,\delta \,(= \frac{G''}{G'})$ following the principle of time-concentration superposition of all the ionogel-m (m: 0.65-0.9). **Table S3** Vertical shift-factor (a_s) of inverse of segmental relaxation time (1/ τ_m) of ionogel-0.14-m

(m:	0.65-0.9) in	Figure	7.
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	a _s
ionogel-0.65	4.23
ionogel-0.7	6.96
ionogel-0.75	9.76
ionogel-0.8	11.96
ionogel-0.85	11.41
ionogel-0.9	8.65



Figure S12 The relationship between the work of extension and conductivity and segment

relaxation time.

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