Supporting Information

Electrochemical Properties of Poly(vinylidene fluoride-*co*-hexafluoropropylene) Gel Electrolytes with High-Concentration Li Salt/Sulfolane for Lithium Batteries

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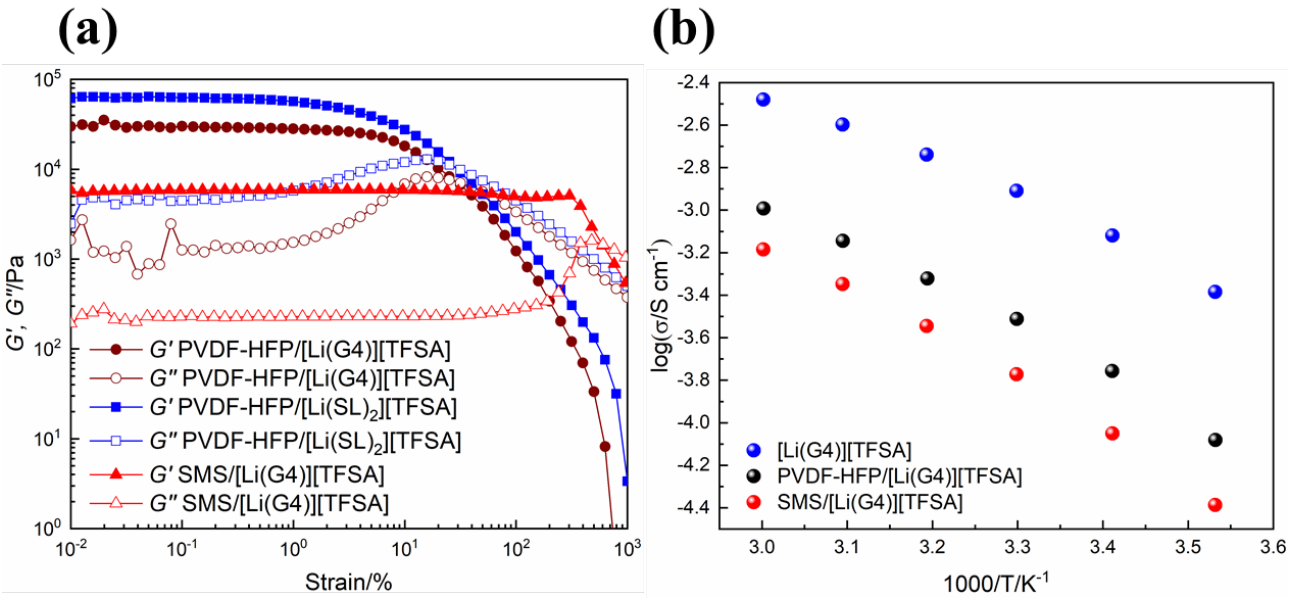

Figure S1. (a) Elastic modulus as a function of dynamic stress amplitude of gel based on 70 wt% highly concentrated electrolytes in polymer matrix at 30 °C. (b) Ionic conductivity of [Li(G4)][TFSA] gel electrolytes with the different polymer matrix at the weight ratio of [Li(G4)][TFSA] : polymer = 70 : 30. The number-average molecular weight, M_n, of SMS is 39.7 kDa. The weight fraction of the PSt block, f_{PSt}, is 46.6 wt%. Regarding the SMS, the details are described in a previous paper (Y. Kitazawa et al., *Macromolecules*, **47**, 6009 (2014)).

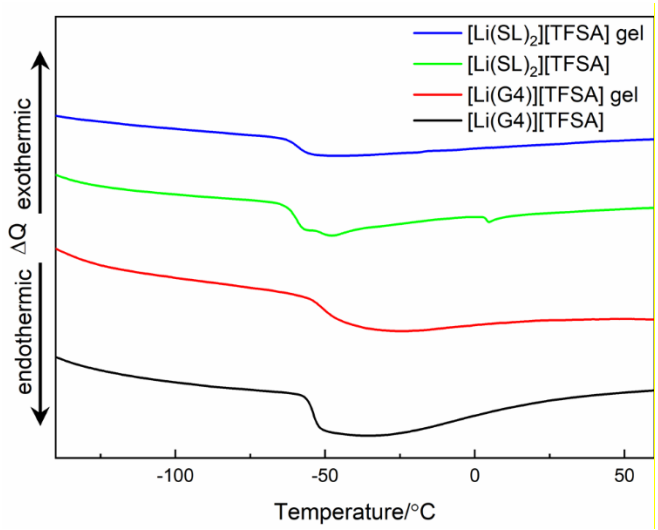

Figure S2. DSC thermograms of highly concentrated electrolytes (without PVDF–HFP) and gel electrolytes at a heating rate of 10 °C min^{-1} under nitrogen atmosphere.

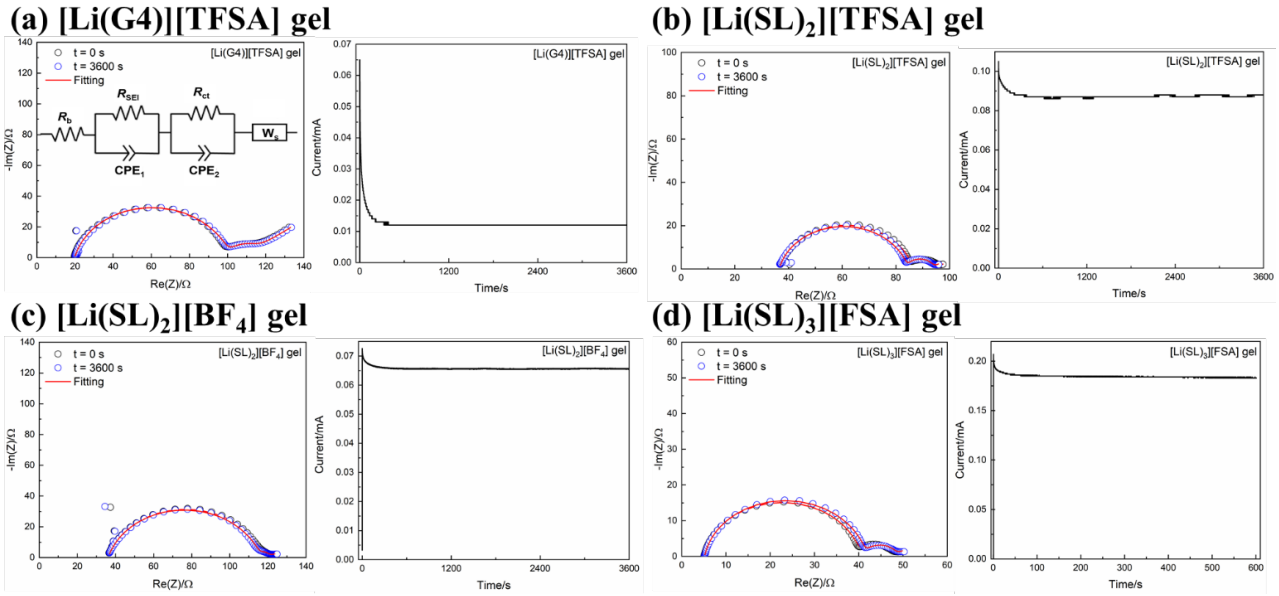

Figure S3. Nyquist plots before/after polarization and cheronoamperogramms of the [Li/Gel/Li] cells. The equivalent circuit shown in the inset was used to fit the data. The contact area of Li/gel electrolyte is 2 cm^2 and the thickness of each gel electrolyte is shown in **Table S1**.

Table S1. Thickness of gel electrolytes, I_Ω, I_{ss}, R'_{bulk}, R'_0, R'_{ss} and t_+ evaluated at 30 °C.

	Thickness [μm]	I_Ω [mA]	I_{ss} [mA]	R'_{bulk} [Ω]	R'_0 [Ω]	R'_{ss} [Ω]	t_+ [-]
[Li(G4)][TFSA]	76	0.080	0.012	19.4	88.1	88.4	0.03
[Li(SL)$_2$][TFSA]	71	0.106	0.088	36.6	56.1	56.0	0.65
[Li(SL)$_2$][BF$_4$]	73	0.072	0.066	35.9	82.9	81.4	0.73
[Li(SL)$_3$][FSA]	69	0.203	0.183	5.17	41.9	42.5	0.53

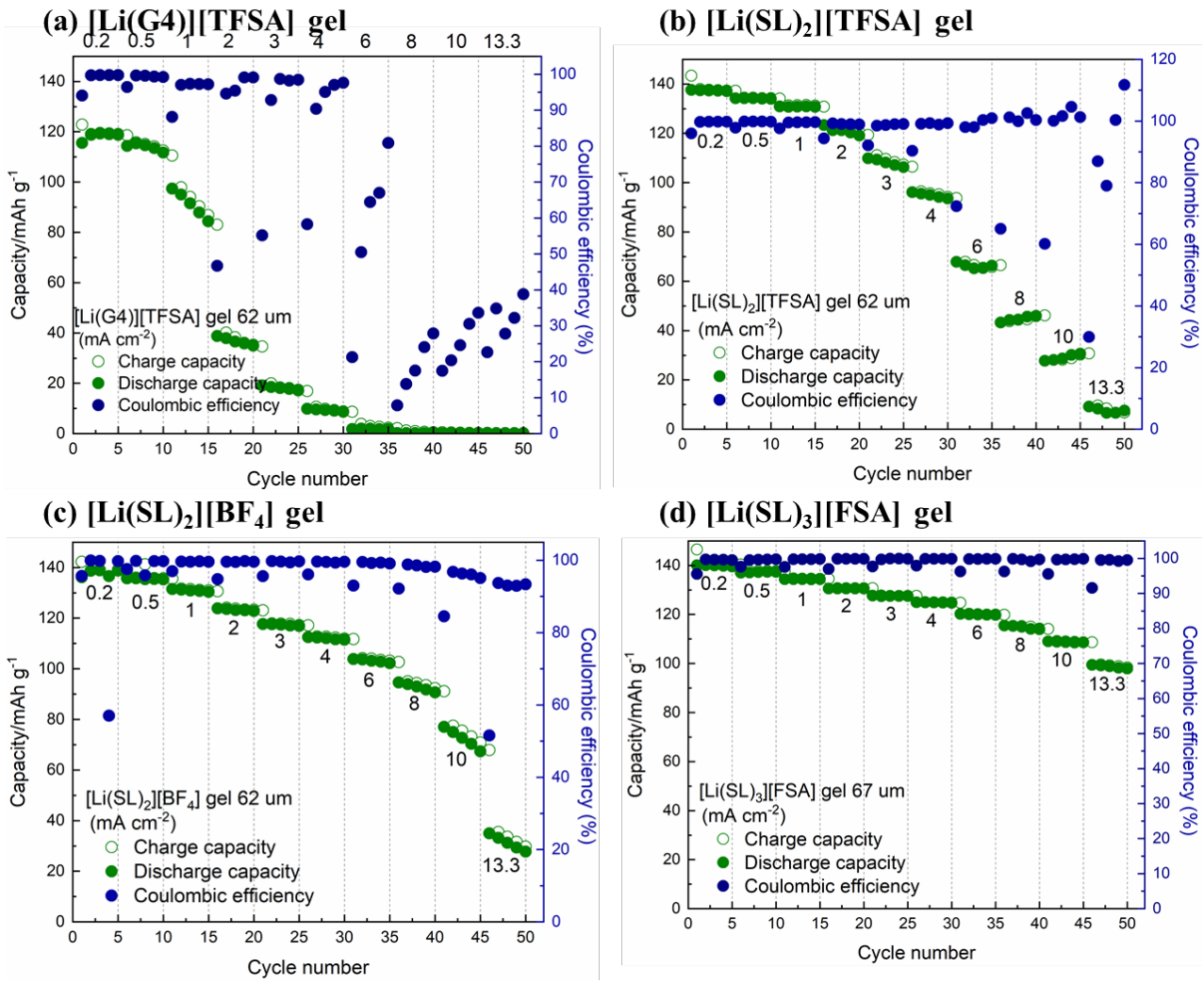

Figure S4. Rate performances of Li/$LiCoO_2$ cells with (a) [Li(G4)][TFSA], (b) $[Li(SL)_2][TFSA]$, (c) $[Li(SL)_2][BF_4]$ and (d) $[Li(SL)_3][FSA]$ gel electrolytes at 30 °C.

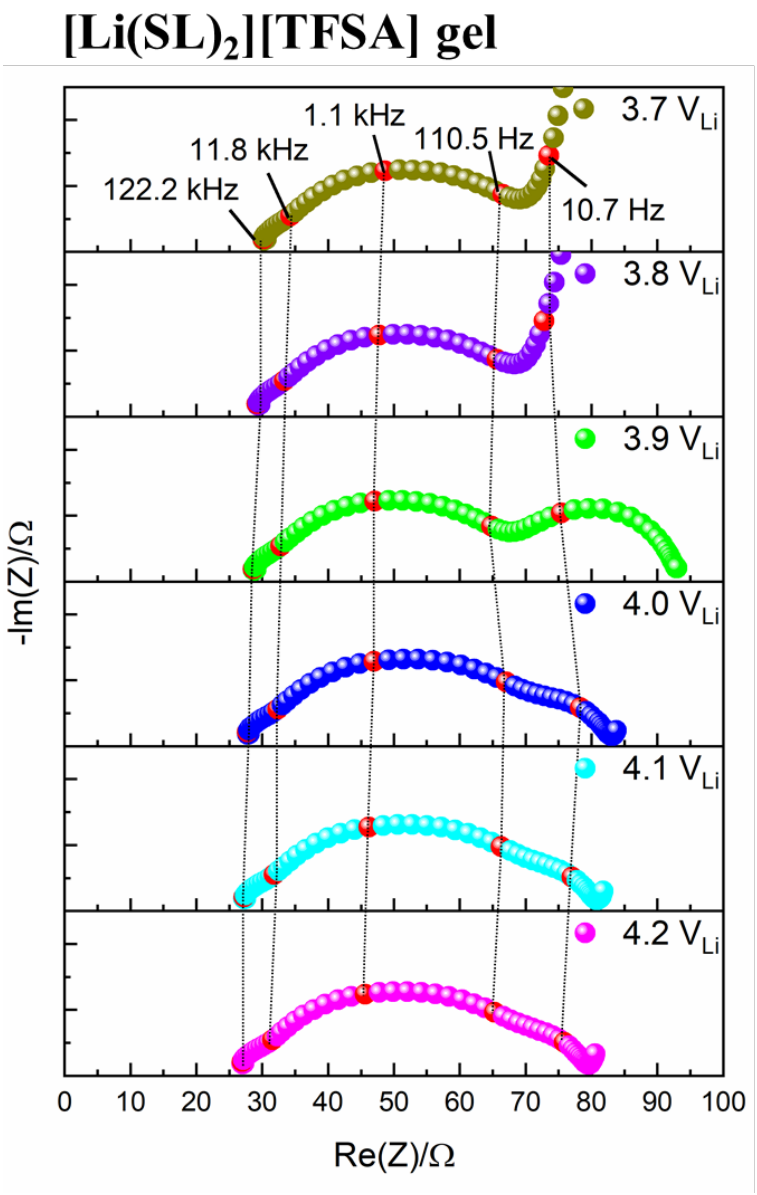

Figure S5. Nyquist plots of a [Li/[Li(SL)$_2$][TFSA] gel/$LiCoO_2$] cell measured at 30 ℃. The electrode areas of Li metal and $LiCoO_2$ electrodes are 2 cm^2 and 1.5 cm^2, respectively.

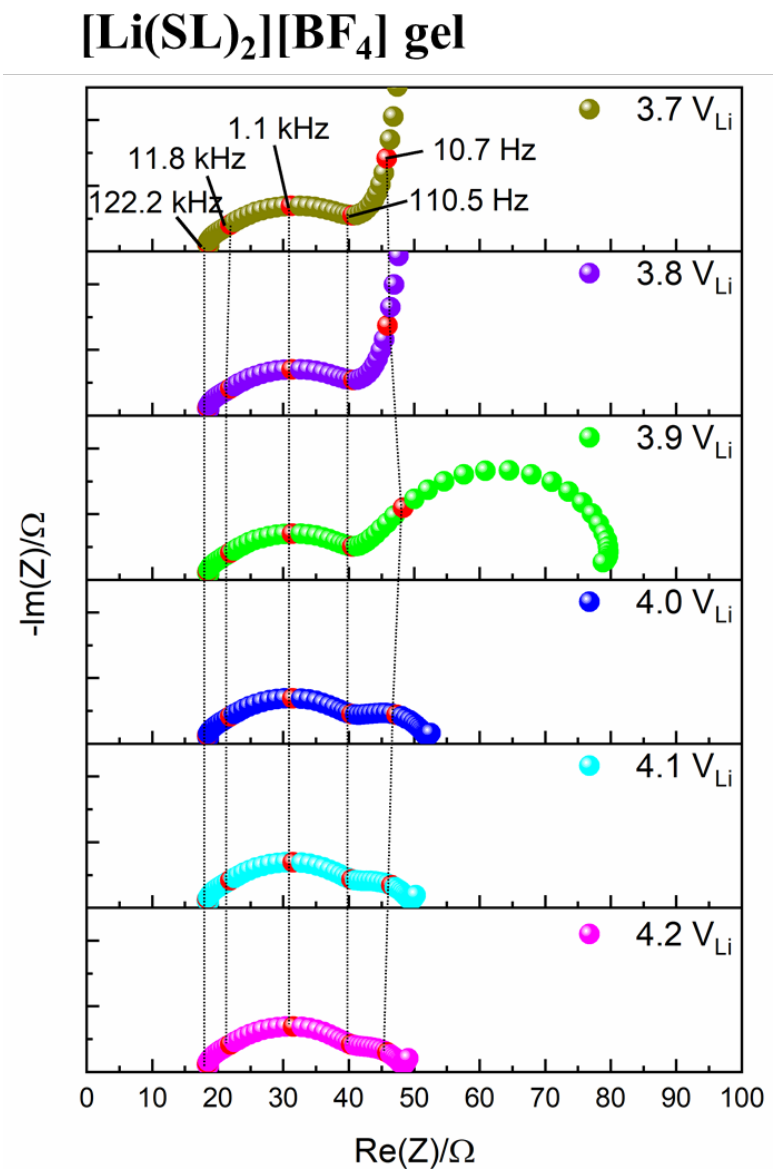

Figure S6. Nyquist plots of a [Li/[Li(SL)$_2$][BF$_4$] gel/LiCoO$_2$] cell measured at 30 ℃. The electrode areas of Li metal and LiCoO$_2$ electrodes are 2 cm^2 and 1.5 cm^2, respectively.

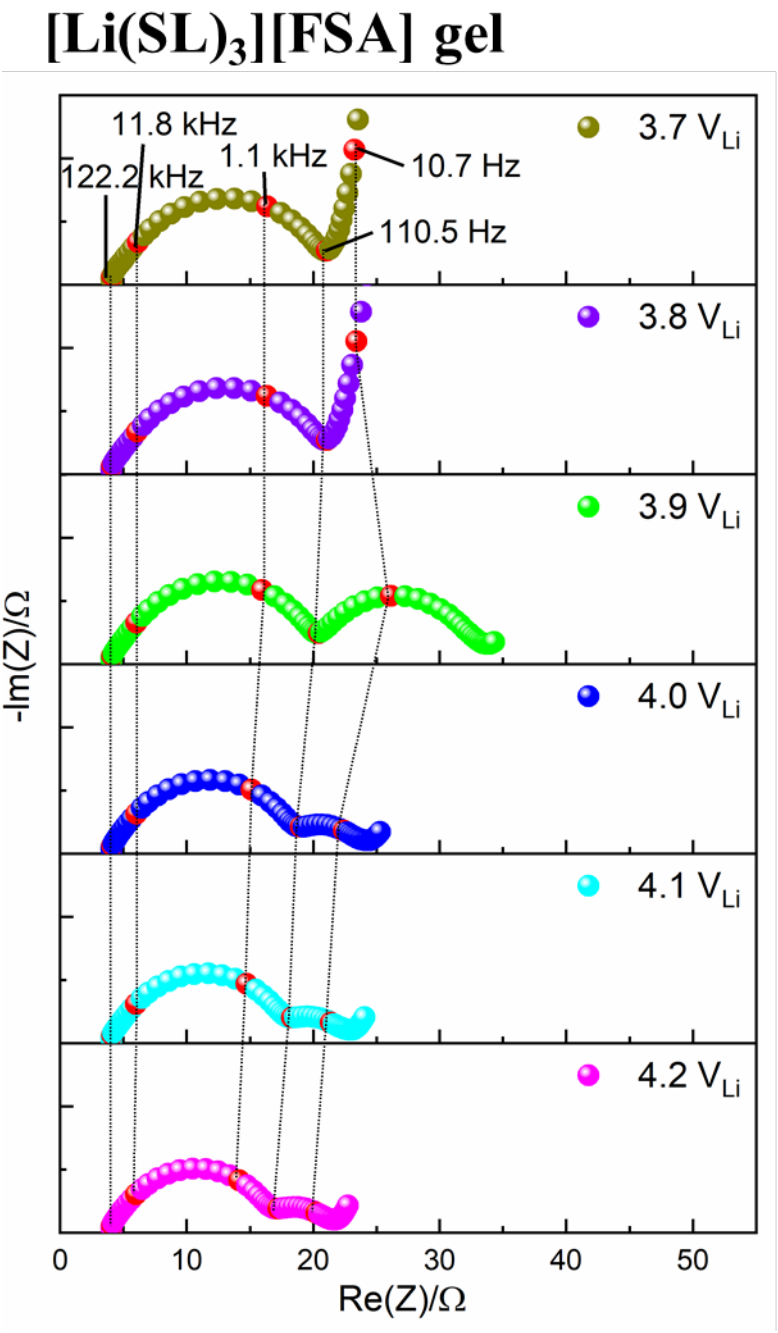

Figure S7. Nyquist plots of a [Li/[Li(SL)$_3$][FSA] gel/$LiCoO_2$] cell measured at 30 °C. The electrode areas of Li metal and $LiCoO_2$ electrodes are 2 cm^2 and 1.5 cm^2, respectively.

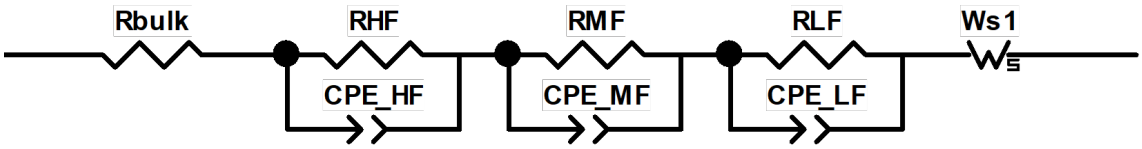

Figure S8. Equivalent circuit model for the [Li/Gel/$LiCoO_2$] cell. Constant phase elements (CPEs) are used instead of capacitances to fit the Nyquist plots. Warburg impedance (Ws1) represents the Li^+ ion diffusion impedance in the electrolyte and $LiCoO_2$ in the frequency range <1 Hz.