Property-Relaxation Correlations in 3D-Siloxane/Polyether Hybrid Polymer Electrolytes

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Determination of the lithium transference numbers

The lithium transference numbers of the electrolytes were determined by potentiostatic polarization measurements, as elsewhere reported.[6] Each sample was sandwiched between lithium electrodes. After the assembly, each cell was stored at 50 °C for 24 h. After cooling to room temperature, an overpotential of 20 mV was applied to the working electrode. The resulting current flow was monitored and the polarization was maintained until a steady state was reached. The lithium transference numbers (t^+) results from the following equation:

$$t^+ = \frac{I_s}{I_0} \cdot \frac{(\Delta V - R_0 I_0)}{(\Delta V - R_S I_S)}$$

where I_s and I_0 are the steady state and initial current, respectively, ΔV is the applied polarization, R_0 and R_s are the interface resistances at the beginning and at the end of the experiments, respectively. These were determined by impedance spectroscopy, as the difference between the high-low frequency intercepts in the Nyquist plot. The temperature was controlled by placing the cell in a climate-chamber (Weiss, WK3), at a temperature of 25 ± 0.5 °C.

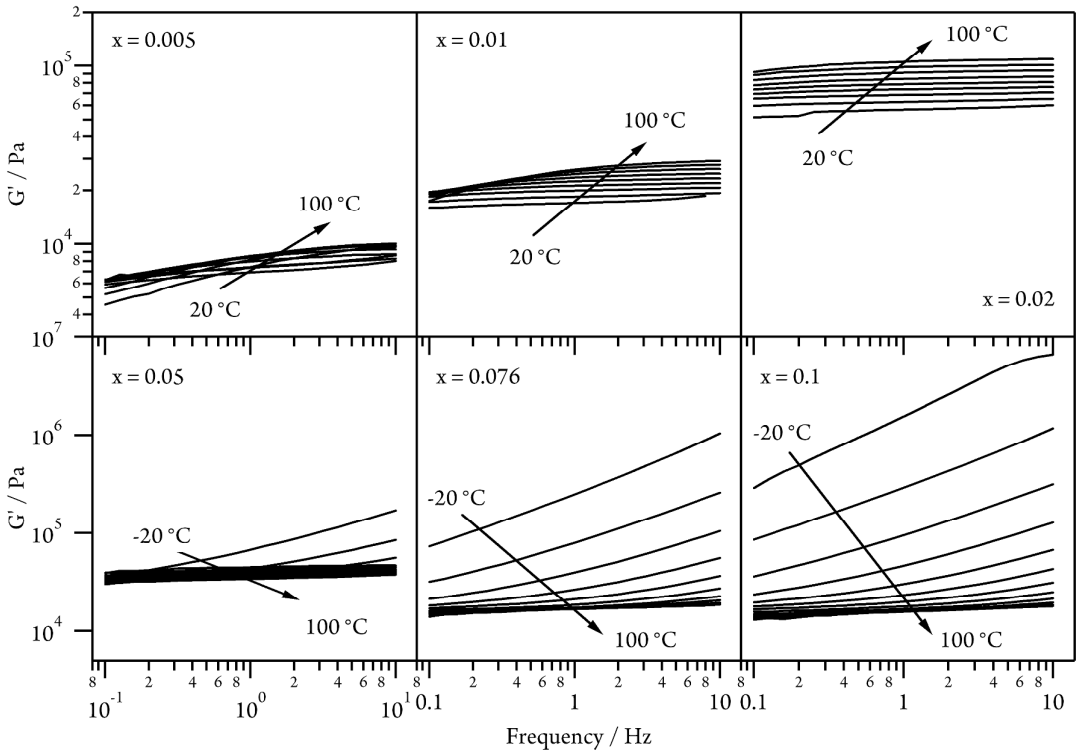

Figure S1: Storage shear modulus of the seven hybrid electrolytes [HP/($LiClO_4$)$_x$] in the high temperature regime.

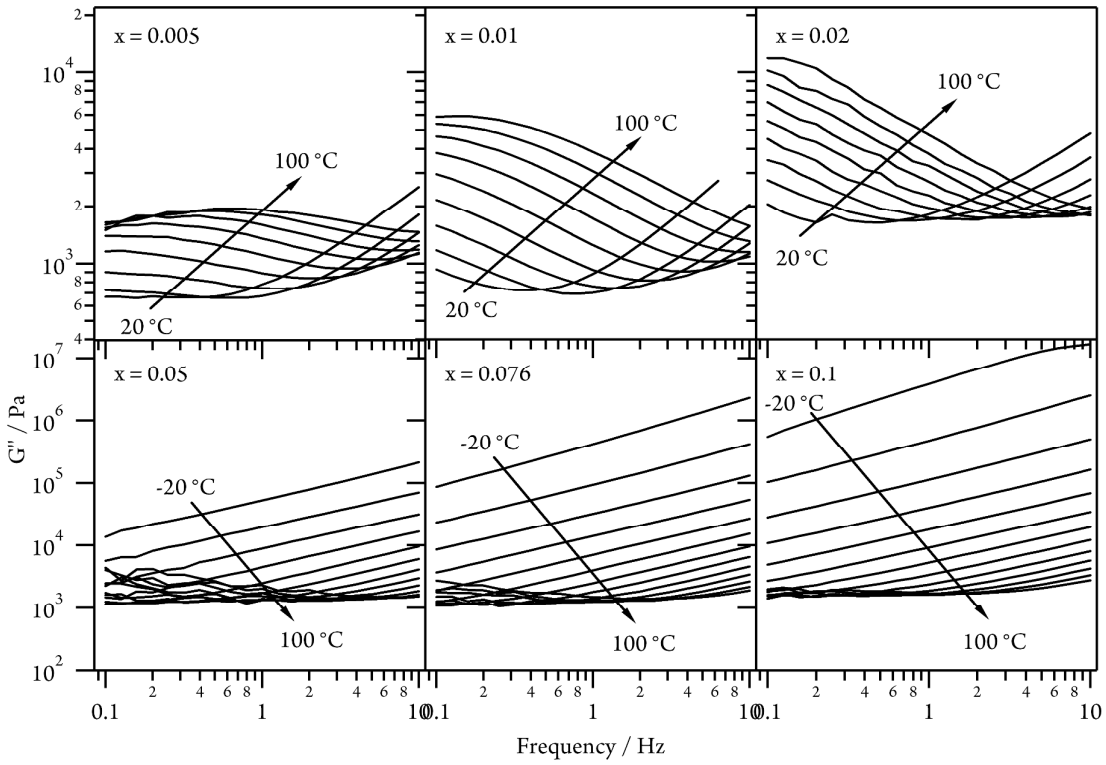

Figure S2: Loss shear modulus of the seven hybrid electrolytes [HP/($LiClO_4$)$_x$] in the high temperature regime.

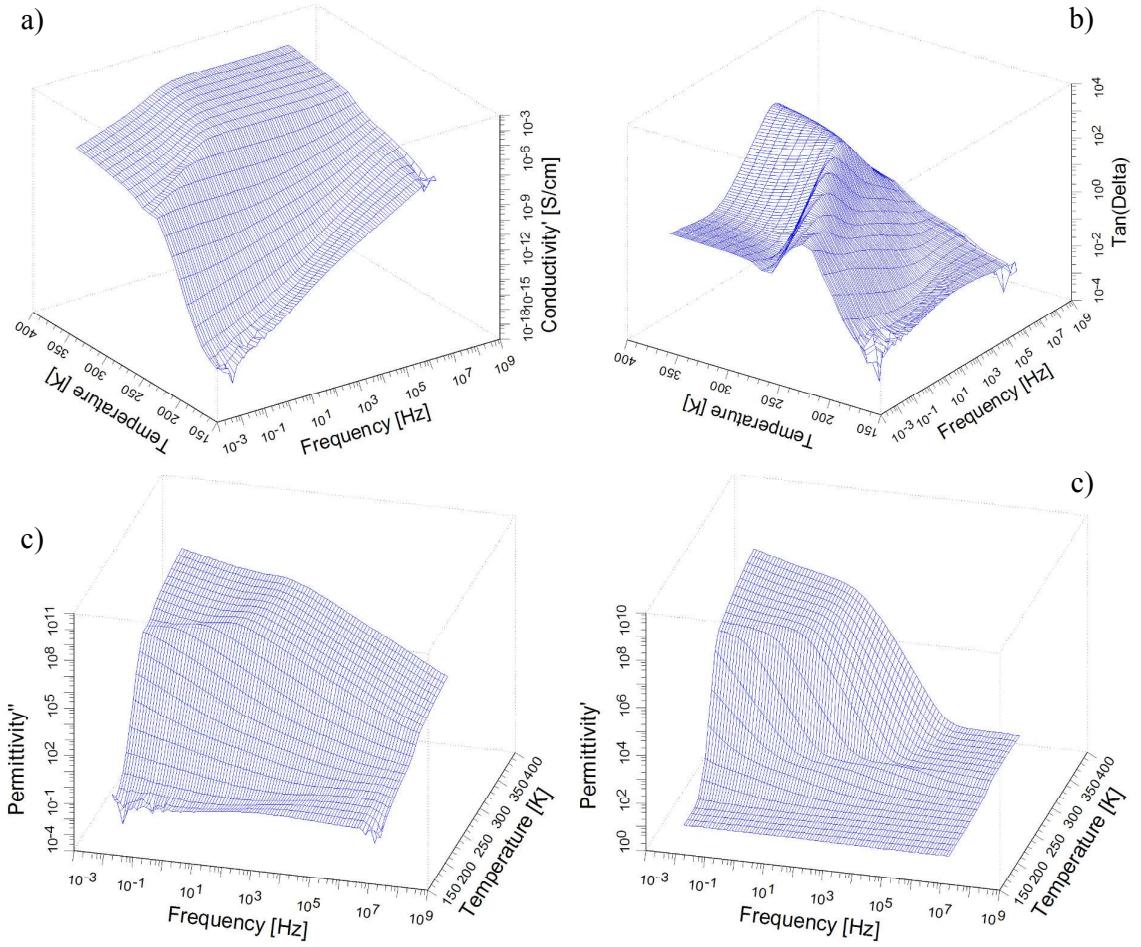

Figure S3: 3D electric spectra of [HP/$(LiClO_4)_{0.005}$] between -100 and 100 °C, 10^{-2} and 10^7 Hz: a) real conductivity (σ'); b) tanδ ($\varepsilon''/\varepsilon'$); c) imaginary permittivity (ε''); d) real permittivity (ε').

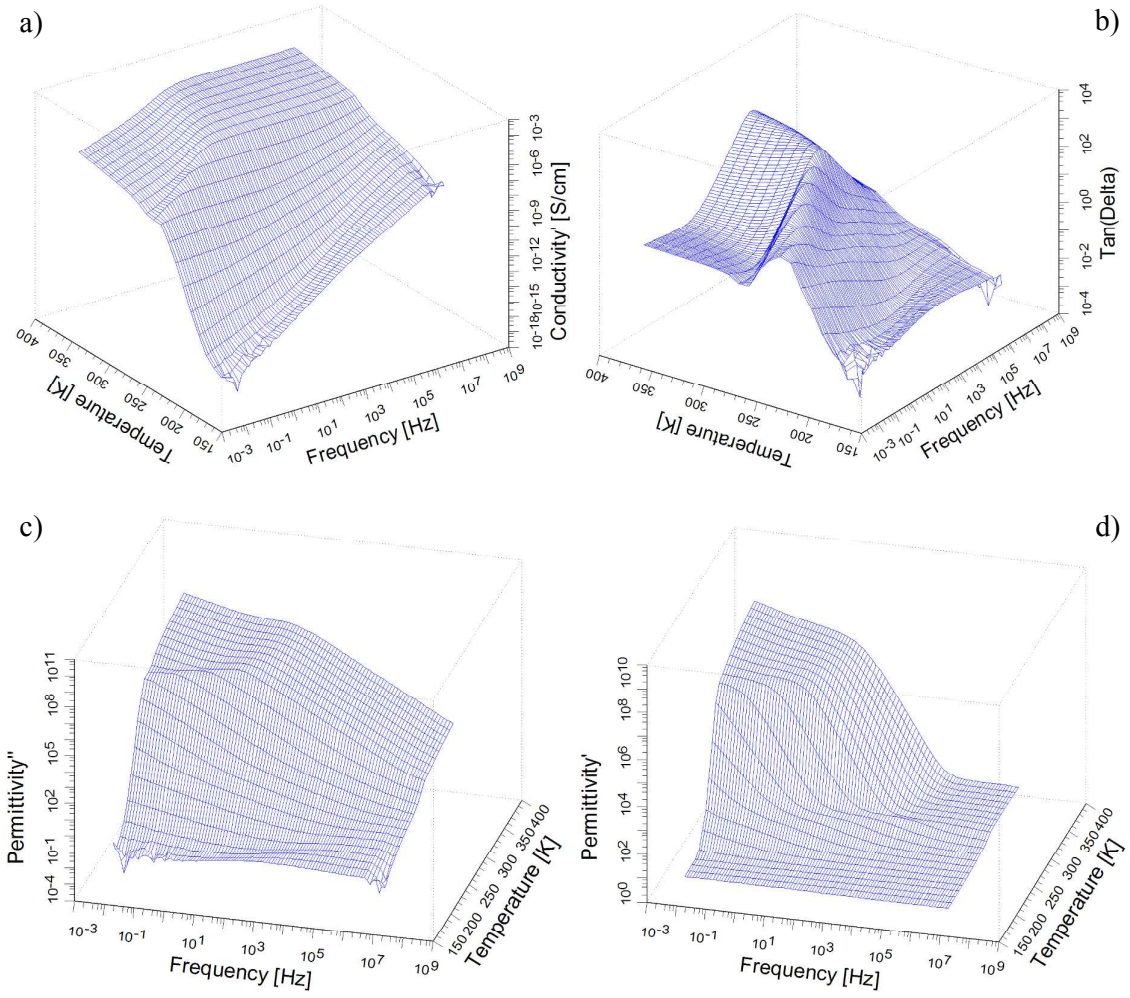

Figure S4: 3D electric spectra of [HP/$(LiClO_4)_{0.01}$] between -100 and 100 °C, 10^{-2} and 10^{7} Hz: a) real conductivity (σ'); b) tanδ ($\varepsilon''/\varepsilon'$); c) imaginary permittivity (ε''); d) real permittivity (ε').

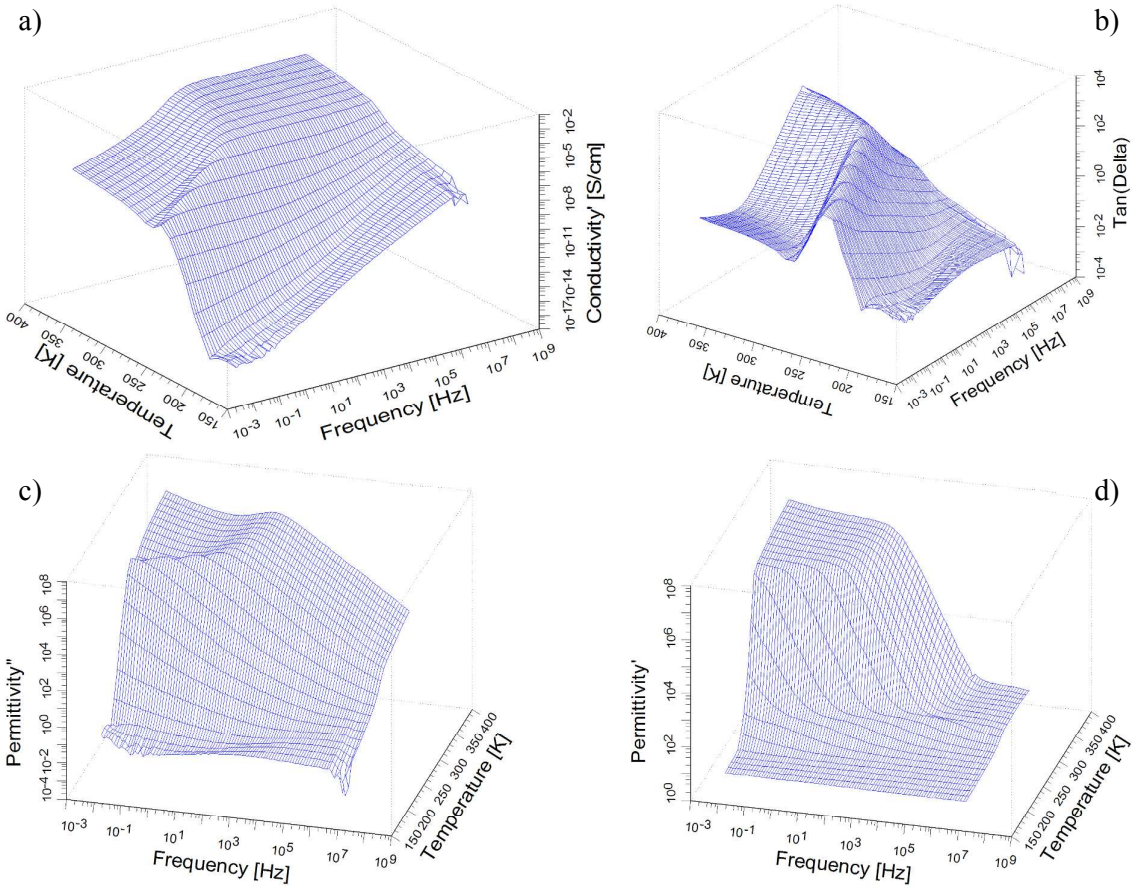

Figure S5: 3D electric spectra of [HP/$(LiClO_4)_{0.02}$] between -100 and 100 °C, 10^{-2} and 10^{7} Hz: a) real conductivity (σ'); b) tanδ ($\varepsilon''/\varepsilon'$); c) imaginary permittivity (ε''); d) real permittivity (ε').

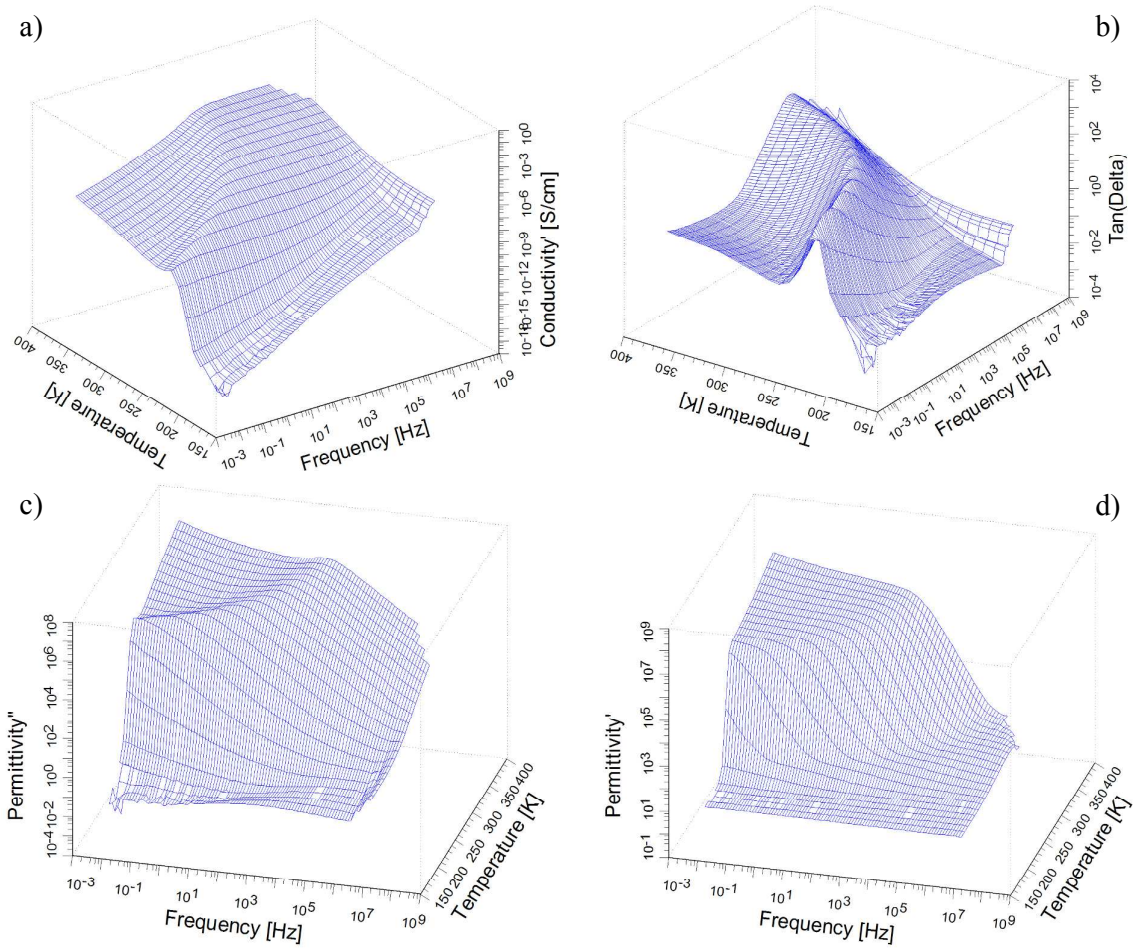

Figure S6: 3D electric spectra of [HP/($LiClO_4$)$_{0.05}$] between -100 and 100 °C, 10^{-2} and 10^7 Hz: a) real conductivity (σ'); b) tanδ ($\varepsilon''/\varepsilon'$); c) imaginary permittivity (ε''); d) real permittivity (ε').

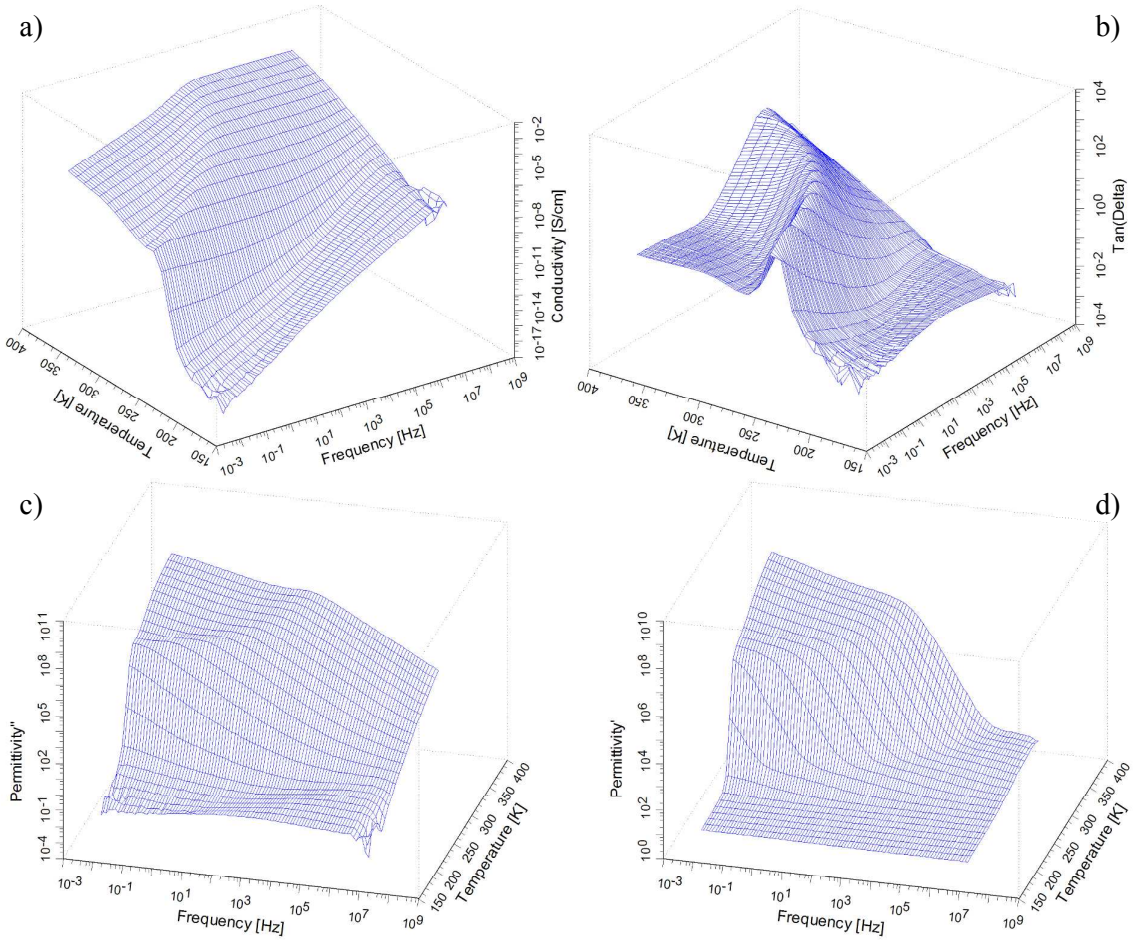

Figure S7: 3D electric spectra of [HP/$(LiClO_4)_{0.076}$] between -100 and 100 °C, 10^{-2} and 10^7 Hz: a) real conductivity (σ'); b) tanδ ($\varepsilon''/\varepsilon'$); c) imaginary permittivity (ε''); d) real permittivity (ε').

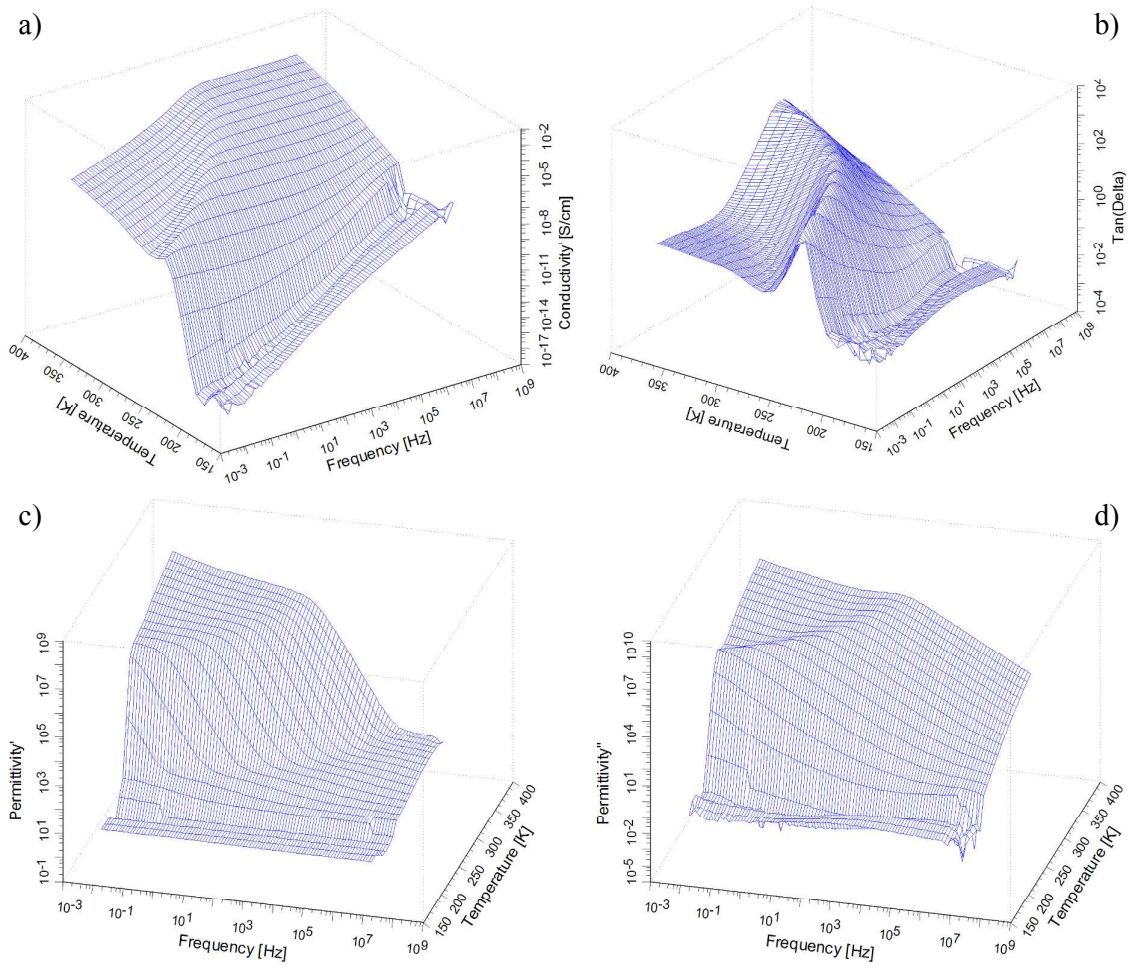

Figure S8: 3D electric spectra of [HP/$(LiClO_4)_{0.1}$] between -100 and 100 °C, 10^{-2} and 10^7 Hz: a) real conductivity (σ'); b) tanδ ($\varepsilon''/\varepsilon'$); c) imaginary permittivity (ε''); d) real permittivity (ε').

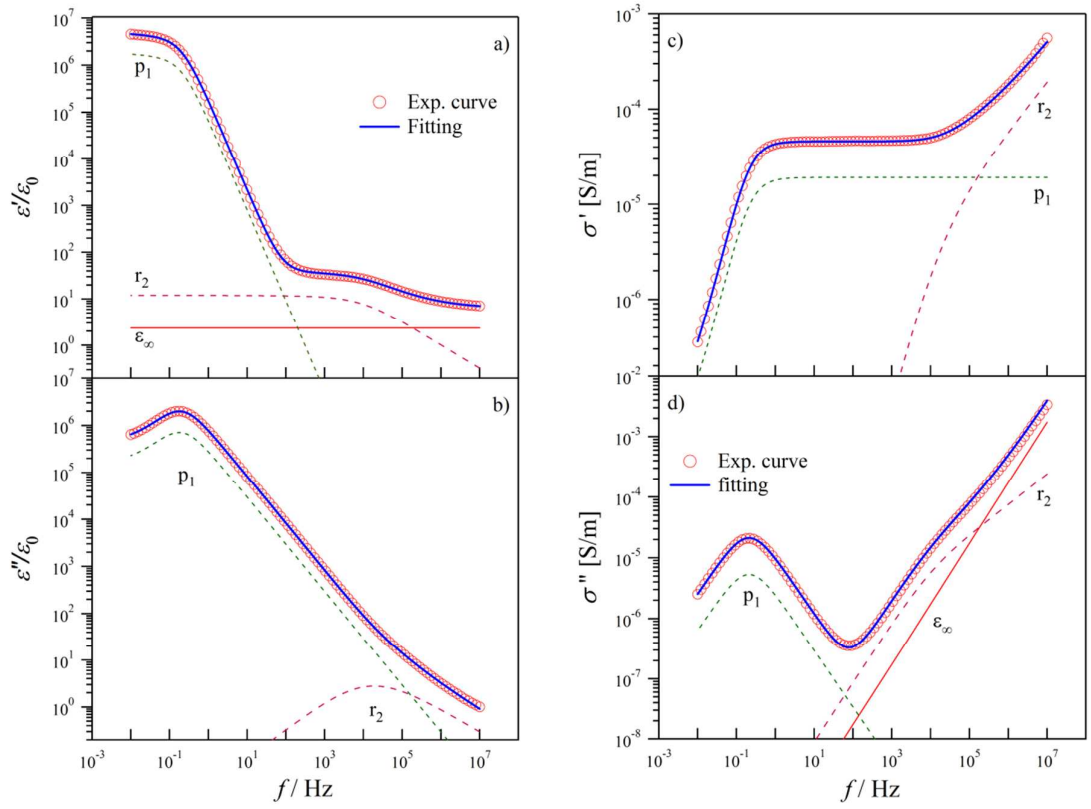

Figure S9: Fitting example of a broadband electric spectrum ([HP/($LiClO_4$)$_{0.02}$], 20 °C): a) real permittivity (ε'); b) imaginary permittivity (ε''); c) real conductivity (σ'); d) imaginary conductivity (σ'').

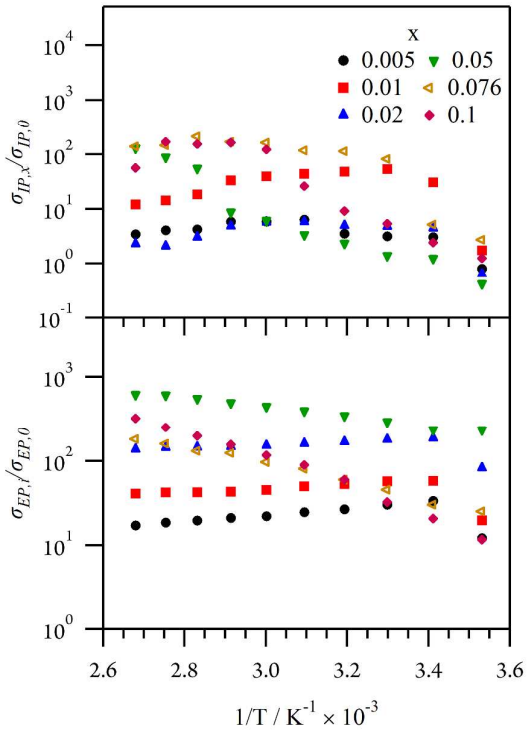

Figure S10: Relative conductivities between room temperature and 100 °C, with x = n($LiClO_4$)/n(EO).

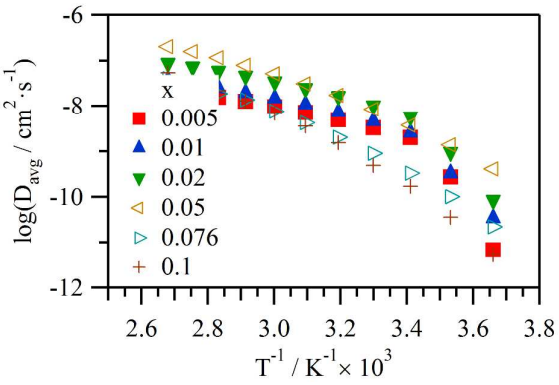

Figure S11: Nernst-Einstein average salt diffusion coefficients, plotted vs. 1/T.