Supporting Information: Transport Properties of Ionic Liquid and Sodium Salt Mixtures for Sodium-Ion Battery Electrolytes from Molecular Dynamics Simulation with a Self-Consistent Atomic Charge Determination

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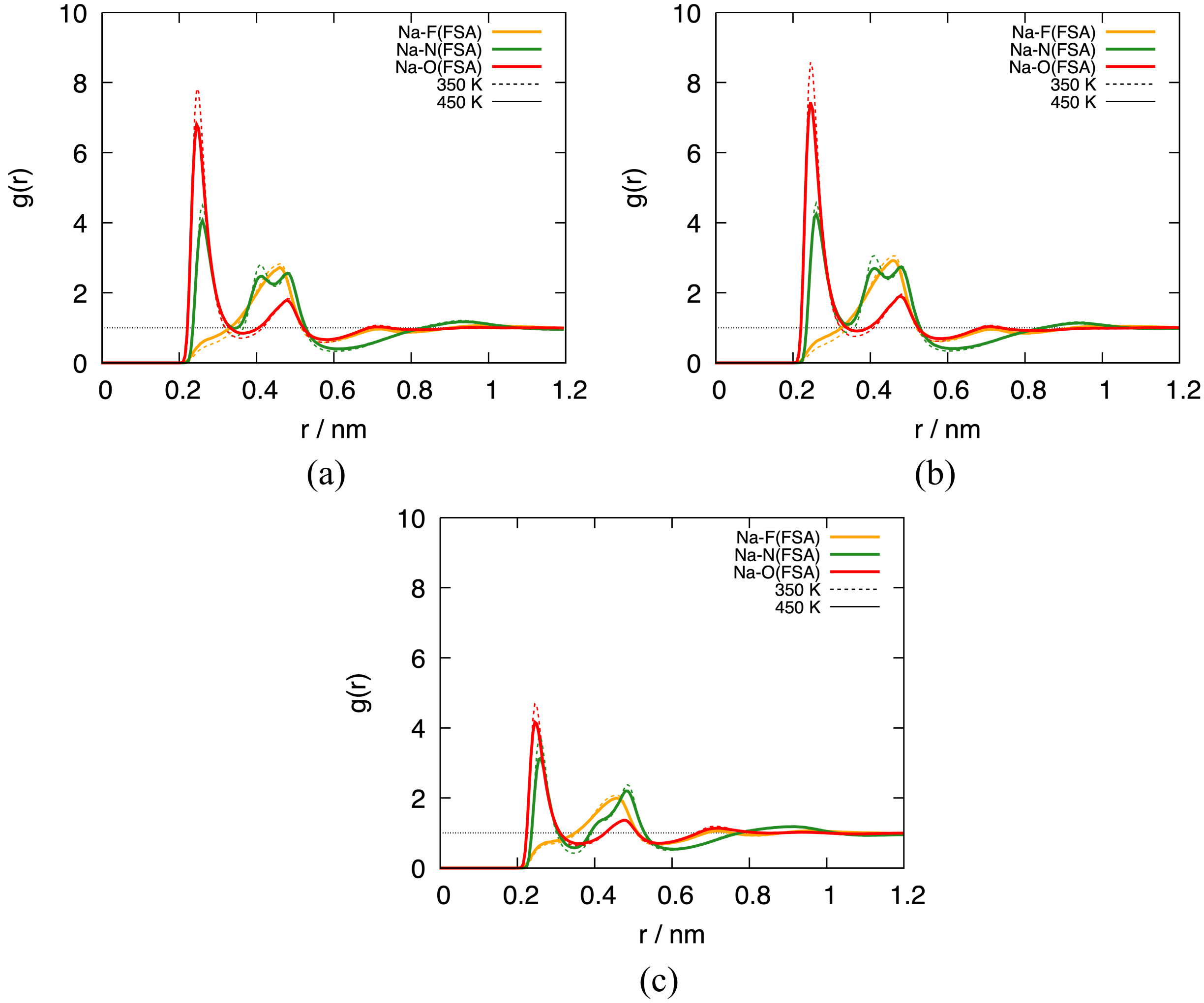

Figure S1: Pair correlation function centering at the sodium ion in (a) [C_2C_1im, Na][FSA], (b) [C_3C_1pyrr, Na][FSA], and (c) [K, Na][FSA], with x(Na[FSA]) = 0.5, at 350 and 450 K of the last iteration step.

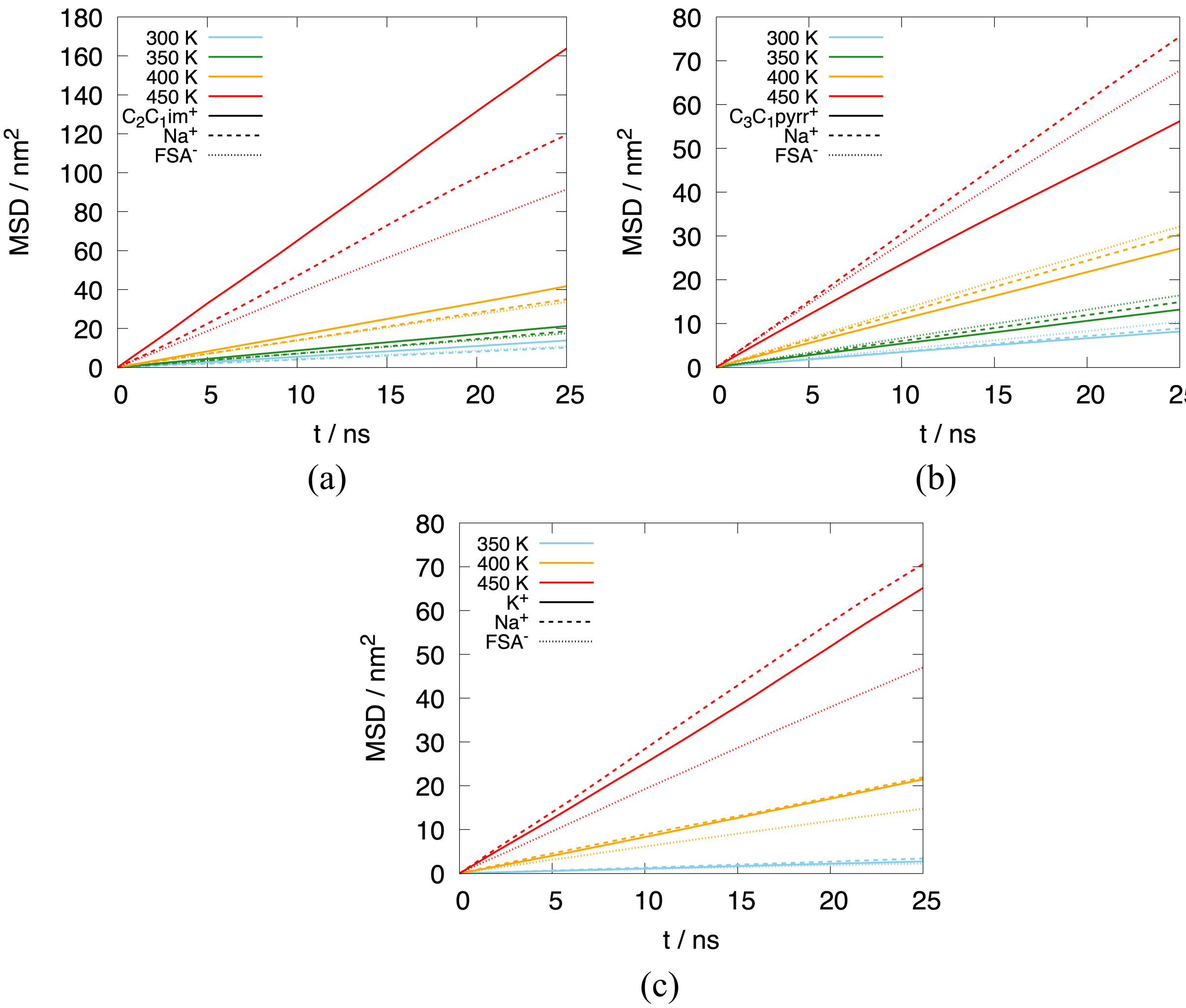

Figure S2: Mean square displacements (MSD) of ionic species in (a) [C_2C_1im, Na][FSA], (b) [C_3C_1pyrr, Na][FSA], and (c) [K, Na][FSA] at different temperatures. MSD for each ion species is sampled from 400 ns production run and the diffusion coefficient is calculated from the slope of MSD in the range of 0.5 to 15 ns.

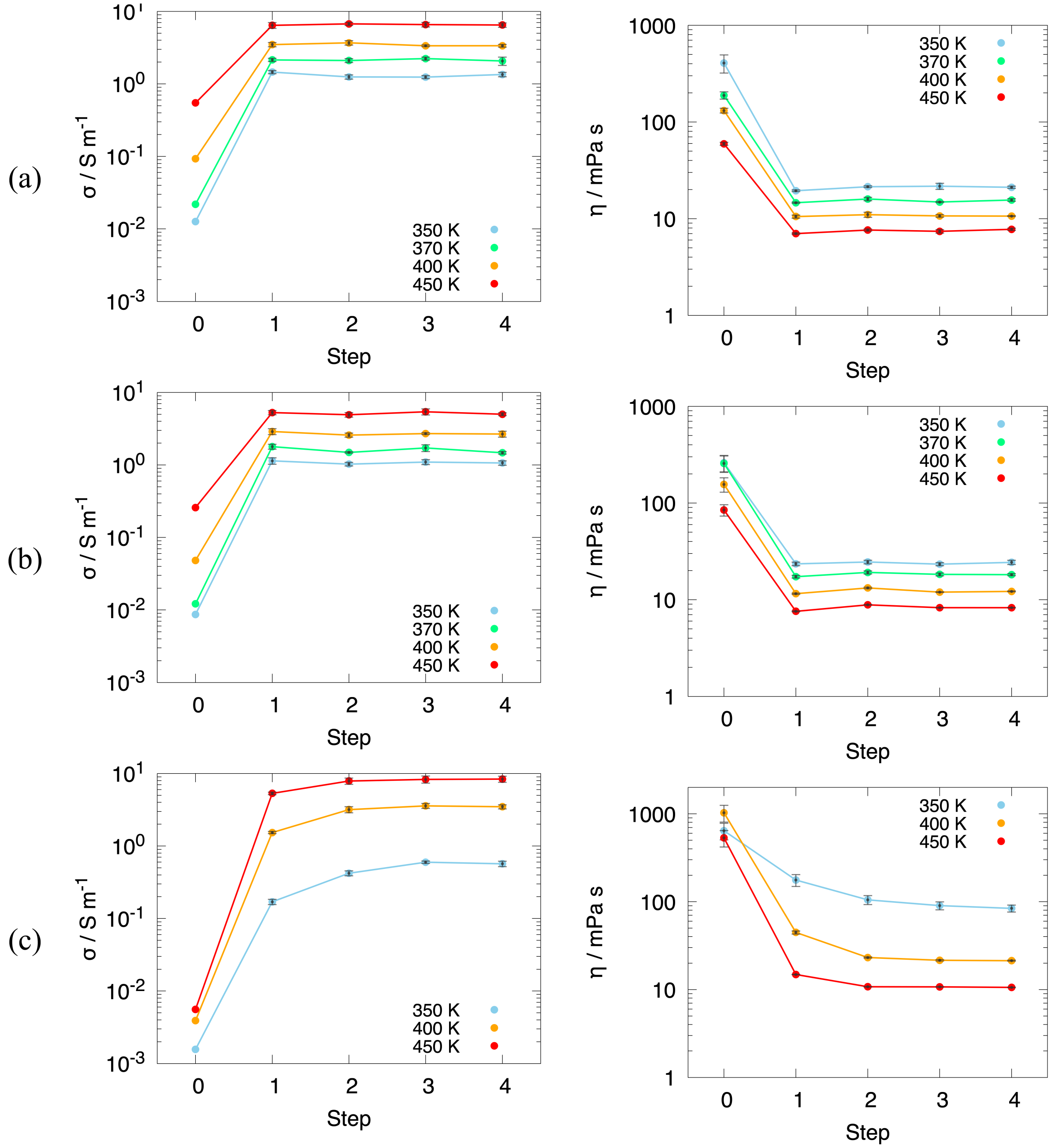

Figure S3: Convergence of the total electrical conductivity and shear viscosity of (a) [C_2C_1im, Na][FSA], (b) [C_3C_1pyrr, Na][FSA], and (c) [K, Na][FSA], with x(Na[FSA]) = 0.5, during MD/DFT iteration scheme. The vertical error bars correspond to the 95% confidence interval (twice the standard error), but they are mostly not appreciable on the scale of the plot.

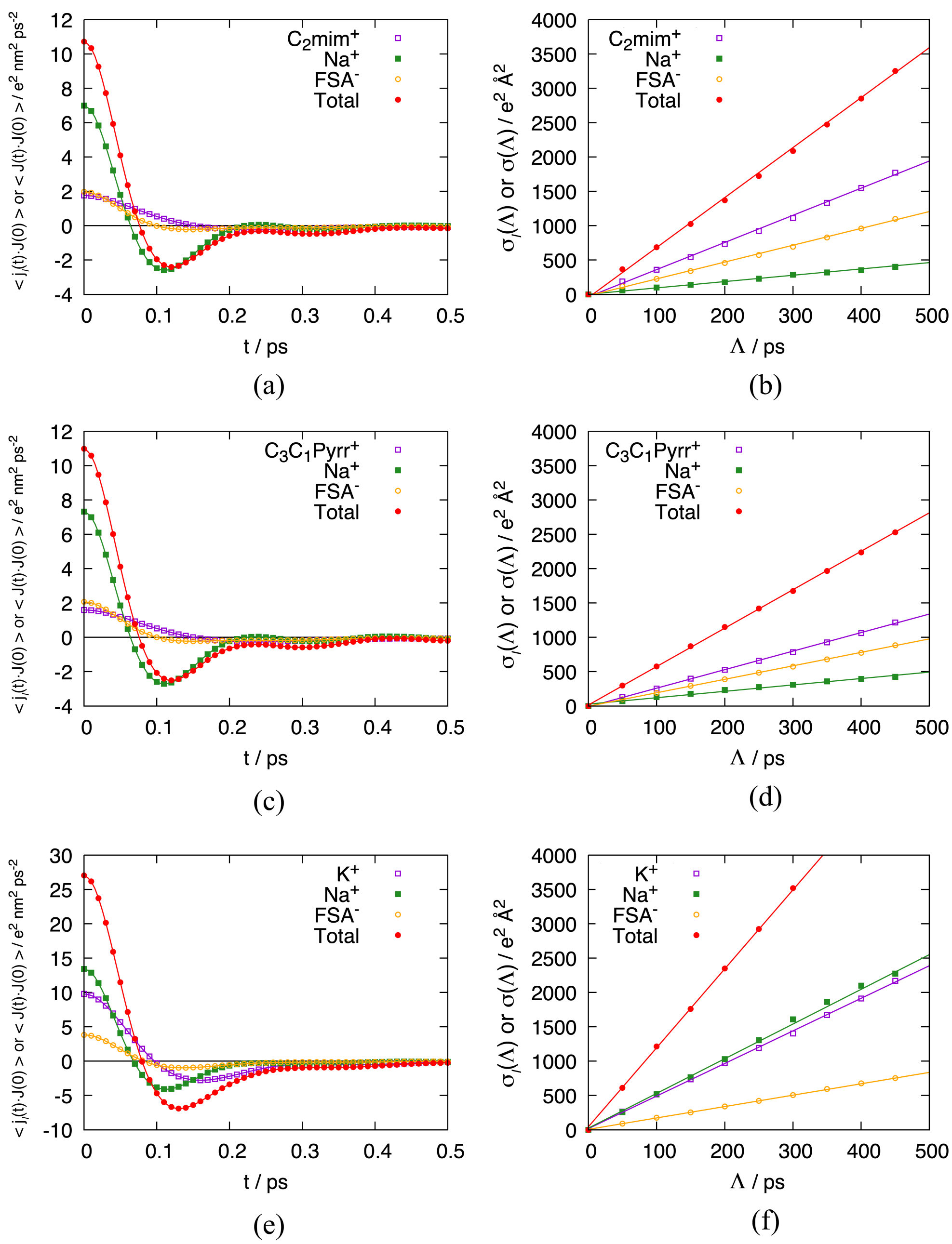

Figure S4: Correlation function of the charge current (the integrand of eq 3 or 6 in the main text) that was used to calculate the electrical conductivity (a, c, e). The numerical stability of the integration was improved using the Cesàro summation method (b, d, f). In (b), (d), and (f), the conductivity was obtained by the linear fit to $\sigma_I(\Lambda)$ or $\sigma(\Lambda)$ in the range of 10 ps $< \Lambda <$ 200 ps.

Table S1: Electrical conductivities of IL mixtures obtained from the Green-Kubo integration supplemented with the Cesàro summation method (eq 3 in the main text), σ; and Nernst-Einstein equation (the sum of eq 11 in the main text), $\sigma_{\mathbf{NE}}$; compared to those from experimental reports, $\sigma_{\mathbf{exp}}$.

T / K	${\sigma_{\mathrm{exp}}}^{\mathrm{S1–S3}}$ / S m^{-1}	σ / S m^{-1}	σ_{NE} / S m^{-1}
[C_2C_1im, Na][FSA][a]			
350	1.20	1.37 ± 0.19	1.81 ± 0.03
370	1.98	2.10 ± 0.14	2.78 ± 0.09
400	3.49	3.89 ± 0.23	4.98 ± 0.13
450	6.59	7.16 ± 0.53	9.20 ± 0.31
[C_3C_1pyrr, Na][FSA][a]			
350	0.57	1.07 ± 0.09	1.36 ± 0.04
370	1.03	1.63 ± 0.16	2.12 ± 0.05
400	1.99	2.87 ± 0.22	3.81 ± 0.07
450	4.26	5.26 ± 0.52	7.28 ± 0.22
[K, Na][FSA][b]			
350	0.13	0.80 ± 0.09	0.66 ± 0.02
400	1.79	3.56 ± 0.38	3.70 ± 0.08
450	6.62	8.50 ± 0.67	9.58 ± 0.19

[a] x(Na[FSA]) = 0.50.

[b] x(Na[FSA]) = 0.56. Note that the values are at x(Na[FSA]) = 0.50 for Table 4 in the main text.

Table S2: Contribution of each ionic species to the electrical conductivity in IL mixtures [X, Na][FSA] at $T = 450\,\mathrm{K}$ with various compositions x(Na[FSA]).[a] These data are also plotted as Figure 10 in the main text.

x(Na[FSA])	$\sigma_{\mathrm{X^+}}$ / S m^{-1}	$\sigma_{\mathrm{Na^+}}$ / S m^{-1}	$\sigma_{\mathrm{FSA^-}}$ / S m^{-1}	σ / S m^{-1}
X = C_2C_1im				
0.1	5.32 ± 0.51	0.04 ± 0.19	3.27 ± 0.30	8.63 ± 0.72
0.3	4.20 ± 0.37	0.72 ± 0.25	2.63 ± 0.20	7.56 ± 0.43
0.5	3.38 ± 0.33	1.58 ± 0.31	2.20 ± 0.19	7.16 ± 0.53
0.7	1.98 ± 0.26	2.64 ± 0.33	1.49 ± 0.16	6.11 ± 0.54
X = C_3C_1pyrr				
0.1	4.59 ± 0.46	0.18 ± 0.14	3.27 ± 0.32	8.07 ± 0.72
0.3	3.54 ± 0.32	0.22 ± 0.26	2.48 ± 0.22	6.25 ± 0.57
0.5	2.32 ± 0.33	1.18 ± 0.20	1.74 ± 0.21	5.26 ± 0.52
0.7	1.44 ± 0.20	2.51 ± 0.25	1.29 ± 0.14	5.24 ± 0.42

[a] The conductivity is determined from the linear fit of Cesàro summation, and the deviation of total electrical conductivity, σ, from the sum of $\sigma_{\mathrm{X^+}}$, $\sigma_{\mathrm{Na^+}}$, and $\sigma_{\mathrm{FSA^-}}$ is a measure of (numerical) error in the fit.

References

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