Supporting Information for

The Importance of Transport Property Studies for Battery Electrolytes: Revisiting the Transport Properties of Lithium – *N*-Methyl-*N*-propylpyrrolidinium Bis(fluorosulfonyl)imide Mixtures.

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Table S1. Densities ρ of [Pyr$_{13}$][FSI] and {Li[FSI]+[Pyr$_{13}$][FSI]} Mixtures

	[Pyr$_{13}$][FSI] sample A, (CoorsTek)	[Pyr$_{13}$][FSI] sample B, (Kanto)	{Li[FSI] + [Pyr$_{13}$][FSI]} mixtures	
m_1 /mol·kg^{-1}	0	0	0.4811	0.9622
x_1	0	0	0.1292	0.2288
θ /^{o}C	ρ /g·cm^{-3}			
0.00	1.3597	1.3591	1.4066	1.4474
5.00	1.3556	1.3550	1.4023	1.4429
10.00	1.3515	1.3510	1.3981	1.4384
15.00	1.3474	1.3469	1.3939	1.4340
20.00	1.3433	1.3429	1.3896	1.4297
20.00	1.3434	-		-
25.00	1.3394	1.3388	1.3855	1.4254
30.00	1.3354	1.3348	1.3814	1.4212
40.00	1.3275	1.3268	1.3732	1.4127
50.00	1.3196	1.3190	1.3650	1.4043
60.00	1.3118	1.3113	1.3568	1.3960
70.00	1.3040	1.3036	1.3488	1.3878
80.00	1.2964	1.2961	1.3410	1.3797
90.00	1.2889	1.2886	1.3332	1.3719

[a] Standard uncertainties u are $u(T)$ = 0.01 K, $u(\rho)$ = 0.0003 g·cm^{-3} for the DMA5000 measurements,[25] but see Table 2 of the main text for u_r values based on purities.

Table S2. Viscosities η of [Pyr$_{13}$][FSI] and {Li[FSI]+[Pyr$_{13}$][FSI]} Mixtures

	[Pyr$_{13}$][FSI] sample A, (Coorstek)	[Pyr$_{13}$][FSI] sample B, (Kanto)	{Li[FSI] + [Pyr$_{13}$][FSI]} mixtures	
m_1 /mol·kg^{-1}	0	0	0.4811	0.9622
x_1	0	0	0.1292	0.2288
θ /^{o}C	η/mPa·s			
0.00	107.9	107.9	182.4	305.5
5.00	86.43	86.69	141.7	230.2
10.00	70.74	70.72	112.3	177.3
15.00	58.42	58.47	90.48	139.3
20.00	48.93	48.95	73.91	111.3
20.00	48.94	48.94	73.92	111.2
20.00	48.91	-	-	-
25.00	41.35	41.39	61.22	90.23
30.00	35.27	35.39	51.40	74.23
40.00	26.39	26.55	37.24	52.13
50.00	20.45	20.53	28.01	38.16
60.00	16.07	16.28	21.69	28.90
70.00	13.11	13.19	17.22	22.52
80.00	10.80	10.88	13.98	17.97
90.00	9.078	9.121	11.53	14.64

[a] Standard uncertainty u is $u(T) = 0.01$ K, relative expanded uncertainty U_r is $100 \cdot U_r(\eta)$ ($k = 2$) = 2.

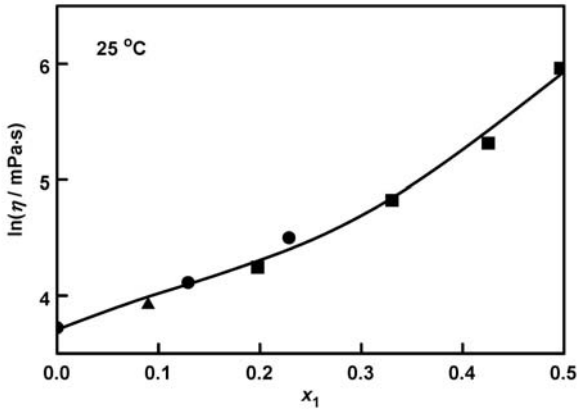

Figure S1. Viscosities of {Li[FSI]+[Pyr_{13}][FSI]} mixtures at 25 °C. Symbols: ●, this work; ▲, Hayamizu *et al.*[27]; ■, Yoon.[19] ln(η/mPa.s) = (3.7045 ± 0.068) + (3.43 ± 1.2) x_1 - (4.75 ± 6.0) x_1^2 +(13.89 ± 8.1) x_1^3, 100·u_r = 6.8.

Table S3. Conductivities κ and MolarConductivities Λ of [Pyr_{13}][FSI] and {Li[FSI]+[Pyr_{13}][FSI]} Mixtures [a]

[Pyr_{13}][FSI] sample A, (Coorstek)			[Pyr_{13}][FSI] sample B, (Kanto)			{Li[FSI] + [Pyr_{13}][FSI]} mixtures (Coorstek)				
m_1/mol·kg^{-1}	0			0			0.4811		0.9622	
x_1	0			0			0.1292		0.2288	
θ/°C	κ/S·m^{-1}	Λ /μS·m^2·mol^{-1}	θ/°C	κ/S·m^{-1}	Λ /μS·m^2·mol^{-1}	θ/°C	κ/S·m^{-1}	Λ /μS·m^2·mol^{-1}	κ/S·m^{-1}	Λ /μS·m^2·mol^{-1}
0.01	0.3679	83.47	0.00	0.3737	84.79	0.00	0.2031	42.27	0.1249	24.21
4.99	0.4496	102.3	5.00	0.4564	103.9	5.00	0.2564	53.52	0.1627	31.64
10.00	0.5418	123.7	10.00	0.5489	125.3	10.00	0.3183	66.63	0.2074	40.47
14.99	0.6425	147.1	15.00	0.6493	148.7	15.00	0.3881	81.49	0.2596	50.79
20.00	0.7534	173.0	20.00	0.7628	175.2	20.00	0.4625	97.40	0.3196	62.73
25.00	0.8766	201.9	25.00	0.8850	203.9	25.00	0.5539	117.0	0.3867	76.13
25.01	0.8765	201.9	25.00	0.8891	204.8	25.00	0.5529	116.8	0.3871	76.21
30.00	1.012	233.8	30.00	1.010	233.4	30.00	0.6475	137.2	0.4624	91.31
40.00	1.308	303.9	40.00	1.318	306.3	40.00	0.8688	185.2	0.6370	126.5
50.00	1.642	383.8	50.00	1.650	385.8	50.00	1.123	240.7	0.8441	168.7
60.00	1.992	468.4	60.00	1.997	469.6	60.00	1.402	302.5	1.085	218.0
70.00	2.387	564.6	69.99	2.356	557.2	70.00	1.724	374.0	1.348	272.6
79.99	2.835	674.5	80.00	2.748	653.7	80.01	2.073	452.4	1.663	338.1

[a] Standard uncertainty u is $u(T) = 0.01$ K, relative expanded uncertainty U_r is $100 \cdot U_r(\kappa)$ ($k = 2$) = 2.

Table S4(a). PGSE NMR Self-diffusion Coefficients, D_{Si} of [Pyr_{13}][FSI] and {Li[FSI]+[Pyr_{13}][FSI]} Mixtures [a]

D_{Si}([Pyr_{13}][FSI]), sample B				D_{Si} {Li[FSI]+[Pyr_{13}][FSI]}									
m_1 /mol·kg^{-1}	0			0.4811					0.9622				
x_1	0			0.1292					0.2288				
θ /°C	$10^{12}\cdot D_{S+}$ /m^2·s^{-1}	$10^{12}\cdot D_{S-}$ /m^2·s^{-1}	D_{S+}/D_{S-}	$10^{12}\cdot D_{S[Li]+}$ /m^2·s^{-1}	$10^{12}\cdot D_{S[Pyr]+}$ /m^2·s^{-1}	$10^{12}\cdot D_{S-}$ /m^2·s^{-1}	$D_{S[Li]+}/D_{S-}$	$D_{S[Pyr]+}/D_{S-}$	$10^{12}\cdot D_{S[Li]+}$ /m^2·s^{-1}	$10^{12}\cdot D_{S[Pyr]+}$ /m^2·s^{-1}	$10^{12}\cdot D_{S-}$ /m^2·s^{-1}	$D_{S[Li]+}/D_{S-}$	$D_{S[Pyr]+}/D_{S-}$
5.00	14.0	17.3	0.81	7.50	8.98	10.3	0.73	0.87	4.69	5.31	6.00	0.78	0.89
15.00	21.3	26.4	0.81	12.0	14.2	16.2	0.74	0.88	8.14	9.25	10.2	0.79	0.90
25.00	31.0	37.7	0.82	17.9	22.0	24.0	0.75	0.92	13.0	14.8	16.0	0.81	0.93
40.00	51.0	61.6	0.83	30.4	37.1	41.1	0.74	0.90	23.0	26.9	28.5	0.81	0.95
60.00	83.9	99.9	0.84	55.0	67.8	73.8	0.75	0.92	42.5	50.8	52.7	0.81	0.96
80.00	129	152	0.85	86.0	108	117	0.73	0.92	68.2	84.3	84.7	0.81	1.00

[a] Standard uncertainty u is $u(T) = 0.01$ K, relative expanded uncertainty U_r is $100\cdot U_r(D_{Si})$ ($k = 2$) = 2.

Table S4(b). Steady Gradient Spin-echo NMR Self-diffusion Coefficients, D_{Si} of {Li[FSI]+[Pyr$_{13}$][FSI]} Mixtures [a]

D_{Si} (Li[FSI]+[Pyr$_{13}$][FSI])					
m_1 /mol·kg^{-1}	0.4811	0.9622			
x_1	0.1292	0.2288			
θ /°C	$10^{12}\cdot D_{S[Pyr]^+}$ /m^2·s^{-1}	θ /°C	$10^{12}\cdot D_{S[Pyr]^+}$ /m^2·s^{-1}	θ /°C	$10^{12}\cdot D_{S-}$/m^2·s^{-1}
20.09	17.3	25.12	14.5	25.22	15.9
39.99	36.5	30.09	17.6	29.77	18.8
49.77	48.9	30.09	17.7	30.14	18.8
59.82	66.2	49.87	36.9	30.15	18.9
59.83	66.0	70.64	66.1	39.86	27.4
79.82	107	79.83	82.8	40.12	28.5
79.84	106	79.83	80.6	40.13	27.1
		79.95	81.5	49.82	40.4
		80.02	81.8	49.86	37.7
				59.90	51.4
				70.51	71.6
				70.52	71.0
				80.12	85.5

[a] Standard uncertainty u is $u(T) = 0.01$ K, relative expanded uncertainty U_r is $100\cdot U_r(D_{Si})$ ($k = 2$) = 3.

[b] In the case of the cation, experiments were performed by varying only the gradient, through positive and negative ranges, at constant τ, the 90°-180° rf pulse spacing , due to the shortness of T_2, as is usual for ionic liquids. For the anion, the longer T_2 allowed variable τ runs, at constant field gradient, $\pm g$ as well. However the low signal to noise ratios produced anomalously low D_S values for the majority of the constant τ experiments, and so only variable τ results are reported. The problem was worse for the 9% mixture in that both types of experiment produced inconsistent values: hence no data are reported for the anion in this case.

Table S5. Velocity Correlation (f_{ij}), and Distinct Diffusion (D^d_{ij}) Coefficients, Laity Resistance Coefficients (r_{ij}), and Nernst-Einstein Δ for [Pyr$_{13}$][FSI]

T/K	f_{++} [a]	f_{--}	f_{+-}	D^d_{++} [b]	D^d_{--}	D^d_{+-}	Δ	r_{++} [c]	r_{--}	r_{+-}	$r_{+-}^2/(r_{++}r_{--})$
273.15	-0.879	-2.28	-1.13	-7.75	-20.1	-9.93	0.163	196	112	222	2.2
278.15	-1.17	-2.91	-1.41	-10.3	-25.6	-12.4	0.178	153	87.5	181	2.5
283.15	-1.51	-3.66	-1.74	-13.3	-32.1	-15.2	0.191	121	69.3	150	2.7
288.15	-1.93	-4.53	-2.12	-16.8	-39.6	-18.5	0.203	97.9	55.9	126	2.9
293.15	-2.41	-5.52	-2.54	-21.0	-48.1	-22.2	0.213	80.2	45.8	107	3.1
298.15	-2.96	-6.64	-3.02	-25.7	-57.7	-26.3	0.222	66.7	38.1	91.7	3.3
303.15	-3.58	-7.90	-3.56	-31.0	-68.4	-30.8	0.229	56.2	32.2	79.5	3.5
313.15	-5.04	-10.8	-4.80	-43.4	-93.0	-41.3	0.240	41.5	23.9	61.2	3.8
323.15	-6.75	-14.2	-6.29	-57.8	-122	-53.8	0.245	32.1	18.7	48.5	3.9
333.15	-8.70	-18.1	-8.02	-74.0	-154	-68.2	0.246	25.8	15.3	39.4	4.0
343.15	-10.8	-22.4	-10.0	-91.5	-189	-84.7	0.242	21.5	13.0	32.7	3.8
353.15	-13.1	-27.0	-12.3	-110	-227	-103	0.236	18.4	11.4	27.7	3.6
$100 \cdot u_r$	6	4	1	6	4	1	**0.03** [d]	6	3	1	

Units: [a] f_{ij}: $10^{-15} \cdot m^5/(mol \cdot s)$. [b] D^d_{ij}: $10^{-12} \cdot m^2/s$. [c] r_{ij}:$10^{12} \cdot J \cdot s \cdot m^{-2} \cdot mol^{-1}$ [d] Actual uncertainty, not a percentage.

Table S6. Apparent Molar Volumes of {Li[FSI]+[Pyr$_{13}$][FSI]} Mixtures

$V_{\phi 1}$\{Li[FSI] (1) - [Pyr$_{13}$][FSI] (0)\}					
m_1 /mol·kg^{-1}	0.3605	0.4811		0.9622	
x_1	0.1	0.1292		0.2288	
θ /°C	$V_{\phi 1}$/cm^2·mol^{-1} [a]	θ /°C	$V_{\phi 1}$/cm^2·mol^{-1}	θ /°C	$V_{\phi 1}$/cm^2·mol^{-1}
10.0	94.2	0.0	82.0	0.0	82.9
15.0	94.5	5.0	82.4	5.0	83.3
20.0	94.8	10.0	82.6	10.0	83.6
25.0	95.1	15.0	82.8	15.0	83.9
30.0	95.4	20.0	83.2	20.0	84.2
35.0	95.7	25.0	83.4	25.0	84.4
40.0	95.9	30.0	83.6	30.0	84.7
45.0	96.2	40.0	84.1	40.0	85.2
50.0	96.6	50.0	84.7	50.0	85.7
55.0	96.9	60.0	85.3	60.0	86.2
60.0	97.2	70.0	85.9	70.0	86.7
65.0	97.5	80.0	86.3	80.0	87.2
70.0	97.8	90.0	86.7	90.0	87.6
75.0	98.1				
80.0	98.4				
U /cm^3·mol^{-1} [b]	1.7 or 7 [b]		0.5		0.2

[a] Calculated from the data of Hayamizu *et al.*[27]

[b] U is the relative expanded uncertainty. This is calculated assuming an uncertainty of 0.05% in the molarities for both data sets (Hayamizu *et al.* did not give a value), and 0.0002 g·cm^{-3} in the densities, since both the Hayamizu group and ourselves used the same vibrating tube densimeter model. However Hayamizu et al conservatively listed values to only 3 decimal places, which gives U = 7 cm^3/mol. U increases with decreasing m_1.

Table S7. Nernst-Einstein Deviation Parameters Δ for {Li[FSI] +[Pyr_{13}][FSI]} Mixtures.

T/ °C	x_1 = 0 (sample A)	x_1 = 0 (sample B)	x_1 =0.1 [2]	x_1 = 0.1292	x_1 = 0.2288
0	0.163	0.156	0.253	0.277	0.295
5	0.178	0.170	0.237	0.289	0.302
10	0.191	0.183	0.227	0.300	0.309
15	0.203	0.196	0.222	0.310	0.316
20	0.213	0.207	0.221	0.319	0.324
25	0.222	0.217	0.223	0.327	0.331
30	0.229	0.225	0.228	0.333	0.337
40	0.240	0.239	0.244	0.344	0.348
50	0.245	0.249	0.266	0.350	0.356
60	0.246	0.254	0.289	0.352	0.361
70	0.242	0.255	0.314	0.351	0.362
80	0.236	0.253	0.339	0.347	0.361

[a] Calculated from the data of Hayamizu *et al.*[27]

Table S8. Coefficients of Eqn (14), $D_{si} = \exp(a + b\, m_1)$. (Fig 18).

Property	T/°C	a	b/(kg·mol^{-1})	average devn/%	a	b/(kg·mol^{-1})	
		m_1 = 0 to 0.9622 (this work)			m_1 < 3.243 (data of Yoon *et al.*[19])		
$D_s[Pyr]^+$ /10^{-12}·m^2·s^{-1}	5	2.635 ± 0.018	-0.9871 ± 0.029	1.1			
	25	3.452 ± 0.018	-0.7819 ± 0.029	1.1	3.554 ± 0.14	-0.6174 ± 0.069	12
	40	3.934 ± 0.018	-0.6600 ± 0.029	1.1	3.998 ± 0.12	-0.5521 ± 0.062	11
	60	4.450 ± 0.018	-0.5300 ± 0.029	1.1	4.528 ± 0.11	-0.4748 ± 0.054	10
	80	4.856 ± 0.018	-0.4280 ± 0.030	1.1			
$D_s[Li]^+$ /10^{-12}·m^2·s^{-1}	5	2.491 ± 0.024	-0.9738 ± 0.039	1.5			
	25	3.273 ± 0.009	-0.7587 ± 0.015	0.6	3.170 ± 0.25	-0.540 ± 0.11	12
	40	3.751 ± 0.007	-0.6512 ± 0.011	0.4	3.623 ± 0.22	-0.473 ± 0.10	11
	60	4.279 ± 0.013	-0.5580 ± 0.021	0.8	4.165 ± 0.19	-0.3927 ± 0.086	9
	80	4.716 ± 0.027	-0.5109 ± 0.043	1.6			
$D_s[FSI]^-$ /10^{-12}·m^2·s^{-1}	5	2.841 ± 0.010	-1.087 ± 0.016	0.6			
	25	3.642 ± 0.002	-0.9052 ± 0.004	0.2	3.655 ± 0.080	-0.6780 ± 0.041	7
	40	4.115 ± 0.002	-0.7976 ± 0.004	0.2	4.022 ± 0.075	-0.6204 ± 0.038	6
	60	4.621 ± 0.007	-0.6824 ± 0.011	0.4	4.459 ± 0.070	-0.5515 ± 0.036	6
	80	5.018 ± 0.010	-0.5920 ± 0.016	0.6			

Table S9. Coefficients of Eqn (14), $\eta = \exp(a + b\, m_1)$.

Property	T/°C	a	b/(kg·mol^{-1})	average devn/%
m_1 = 0 to 0.9622 (this work and Hayamizu *et al.*[27])				
η/mPa·s	10	4.239 ± 0.028	0.9695 ± 0.050	2.1
	25	3.706 ± 0.025	0.8214 ± 0.044	1.8
	40	3.262 ± 0.021	0.7136 ± 0.038	1.6
	60	2.775 ± 0.017	0.6076 ± 0.031	1.3
	80	2.375 ± 0.018	0.5298 ± 0.031	1.3

Table S10. Coefficients of Eqn (14), $\Lambda = \exp(a + b\, m_1)$.

Property	T/°C	a	b/(kg·mol^{-1})	average devn/%
m_1 = 0 to 0.9622 (this work)				
Λ /μS·m^2·mol^{-1}	0	4.417 ± 0.052	-1.302 ± 0.083	3.1
	10	4.809 ± 0.050	-1.175 ± 0.080	3.0
	25	5.298 ± 0.050	-1.025 ± 0.080	3.0
	40	5.704 ± 0.046	-0.9177 ± 0.074	2.7
	60	6.133 ± 0.042	-0.7971 ± 0.068	2.5
	80	6.740 ± 0.028	0.6859 ± 0.046	1.7