Supporting Information

The structure and hydrogen-bond properties of N-alkyl-N-methyl-pyrrolidinium bis(trifluoromethylsulfonyl)imide and DMSO mixtures

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Table S1. The peak positions (cm^{-1}) of positive and negative bands in Figure 2A2, B2, C2 and D2.

	[C3MPyr]$^+$–[Tf_2N]$^-$–DMSO−d_6		[C4MPyr]$^+$–[Tf_2N]$^-$–DMSO−d_6		[C6MPyr]$^+$–[Tf_2N]$^-$–DMSO−d_6		[C8MPyr]$^+$–[Tf_2N]$^-$–DMSO−d_6	
x(DMSO−d_6)	Positive peak	Negative peak	Positive peak	Negative peak	Positive peak	Negative peak	Positive peak	Negative peak
0.9	2129.1	2120.4	2129.3	2120.6	2129.1	2120.4	2129.1	2120.1
0.8	2129.8	2121.1	2129.8	2121.1	2129.8	2120.9	2129.5	2120.9
0.7	2130.3	2121.3	2130.5	2121.6	2130.3	2121.3	2130.0	2121.3
0.6	2130.8	2121.8	2130.8	2121.8	2130.8	2121.8	2130.5	2121.6
0.5	2131.2	2122.1	2131.2	2122.1	2131.0	2122.1	2130.8	2121.8
0.4	2131.5	2122.3	2131.5	2122.3	2131.2	2122.1	2131.0	2122.1
0.3	2131.7	2122.3	2131.7	2122.3	2131.5	2122.3	2131.2	2122.3
0.2	2132.0	2122.6	2131.7	2122.6	2131.5	2122.6	2131.2	2122.3
0.1	2132.2	2122.8	2132.0	2122.8	2131.7	2122.6	2131.5	2122.6

Table S2. Three topological properties (a.u) at the BCPs of the hydrogen-bonds between cation and anion at the most stable geometry of $[CnMPyr]^+–[Tf_2N]^–$DMSO$–d_6$ complexes obtained using the AIM theory by the m06-2x/6-311g(d,p) method.

Complex	Hydrogen-bond	Distance	ρ_{BCP}	$\nabla^2\rho_{BCP}$	$H_{BCP}(10^{-3})$
$[C3MPyr]^+–[Tf_2N]^–$DMSO$–d_6$	O3···H29	2.190	0.016	0.060	1.868
	O3···H36	2.251	0.014	0.051	1.712
	O4···H32	2.282	0.014	0.050	1.564
	O4···H39	2.437	0.010	0.043	1.696
	O4···H36	2.428	0.011	0.041	1.360
	O2···H26	2.496	0.010	0.037	1.193
	O4···H41	2.500	0.009	0.034	1.115
$[C4MPyr]^+–[Tf_2N]^–$DMSO$–d_6$	O3···H30	2.210	0.016	0.056	1.742
	O3···H37	2.233	0.015	0.053	1.784
	O4···H40	2.362	0.012	0.050	2.020
	O4···H33	2.292	0.014	0.049	1.519
	O4···H37	2.475	0.010	0.038	1.239
	O2···H27	2.500	0.010	0.038	1.219
	O4···H41	2.500	0.010	0.036	1.155
$[C6MPyr]^+–[Tf_2N]^–$DMSO$–d_6$	O3···H39	2.195	0.016	0.058	1.987
	O3···H32	2.220	0.016	0.055	1.699
	O4···H42	2.342	0.013	0.053	2.138
	O4···H35	2.277	0.014	0.051	1.607
	O4···H43	2.496	0.010	0.039	1.248
	O4···H39	2.500	0.010	0.036	1.163
	O2···H29	2.500	0.010	0.036	1.162
$[C8MPyr]^+–[Tf_2N]^–$DMSO$–d_6$	O3···H41	2.168	0.016	0.062	2.120
	O4···H44	2.322	0.013	0.056	2.242
	O4···H37	2.291	0.014	0.049	1.566
	O3···H34	2.275	0.014	0.048	1.435
	O4···H45	2.530	0.010	0.040	1.274

O4···H41	2.558	0.009	0.034	1.098
O2···H31	2.574	0.009	0.033	1.094

Table S3. Three topological properties (a.u) at the BCPs of the hydrogen-bonds between cation and anion at the most stable geometry of 2[CnMPyr]$^+$−2[Tf_2N]$^-$−DMSO−d_6 complexes obtained using the AIM theory by the m06-2x/6-311g(d,p) method.

Complex	Hydrogen-bond	Distance	ρ_{BCP}	$\nabla^2\rho_{BCP}$	$H_{BCP}(10^{-3})$
2[C3MPyr]$^+$–2[Tf_2N]$^-$–DMSO−d_6	O2···H29	2.272	0.013	0.048	1.732
	O5···H72	2.295	0.014	0.048	1.518
	O46···H68	2.367	0.013	0.048	1.580
	O4···H36	2.334	0.013	0.047	1.520
	O45···H26	2.378	0.012	0.043	1.502
	O43···H73	2.368	0.012	0.039	1.103
	O2···H69	2.470	0.011	0.037	1.013
	O43···H77	2.474	0.011	0.037	0.956
	N1···H76	2.443	0.012	0.036	1.222
	O2···H36	2.455	0.011	0.034	0.822
	N42···H73	2.528	0.011	0.033	0.803
	N42···H70	2.519	0.011	0.032	0.884
	O4···H32	2.507	0.008	0.028	0.912
	N42···H77	2.580	0.009	0.028	0.732
2[C4MPyr]$^+$–2[Tf_2N]$^-$–DMSO−d_6	O49···H72	2.361	0.013	0.048	1.615
	O4···H37	2.350	0.013	0.047	1.517
	O2···H30	2.285	0.013	0.046	1.666
	O48···H27	2.347	0.013	0.046	1.605
	O4···H40	2.435	0.011	0.043	1.607
	O5···H76	2.336	0.013	0.043	1.300
	O46···H84	2.479	0.010	0.040	1.400
	O46···H77	2.382	0.012	0.038	1.059
	O2···H73	2.476	0.011	0.037	1.044
	O46···H81	2.489	0.011	0.036	0.929
	N1···H80	2.471	0.011	0.034	1.126
	O2···H37	2.460	0.011	0.033	0.789

	N45···H77	2.533	0.011	0.033	0.796
	O3···H41	2.443	0.010	0.032	0.886
	N45···H74	2.507	0.011	0.032	0.916
	O4···H33	2.491	0.009	0.029	0.958
	N45···H81	2.588	0.009	0.027	0.714
2[C6MPyr]$^+$–2[Tf_2N]$^-$–DMSO−d_6	O55···H80	2.368	0.013	0.047	1.591
	O54···H29	2.346	0.012	0.046	1.608
	O4···H39	2.370	0.012	0.045	1.447
	O4···H42	2.413	0.011	0.045	1.686
	O5···H84	2.327	0.013	0.044	1.352
	O2···H32	2.318	0.012	0.043	1.530
	O52···H92	2.462	0.011	0.042	1.458
	O2···H39	2.402	0.012	0.037	0.919
	O52···H85	2.402	0.011	0.037	1.005
	N1···H88	2.439	0.012	0.036	1.232
	O52···H89	2.494	0.011	0.036	0.926
	O2···H81	2.491	0.011	0.035	0.968
	N51···H85	2.523	0.011	0.034	0.804
	O3···H43	2.430	0.010	0.032	0.901
	N51···H82	2.527	0.010	0.031	0.868
	N51···H89	2.572	0.010	0.028	0.740
2[C8MPyr]$^+$–2[Tf_2N]$^-$–DMSO−d_6	O5···H93	2.367	0.013	0.048	1.600
	O61···H31	2.339	0.013	0.047	1.631
	O62···H89	2.308	0.013	0.046	1.435
	O4···H44	2.411	0.011	0.045	1.710
	O2···H34	2.318	0.012	0.043	1.532
	O59···H101	2.455	0.011	0.042	1.485
	O59···H94	2.396	0.011	0.037	1.022
	O2···H41	2.400	0.012	0.037	0.922
	N1···H97	2.441	0.012	0.036	1.222
	O59···H98	2.497	0.011	0.036	0.923
	O62···H91	2.500	0.010	0.035	0.999
	N58···H94	2.518	0.012	0.034	0.815

O3···H45	2.451	0.010	0.031	0.840
N58···H91	2.534	0.010	0.031	0.851
N58···H98	2.576	0.010	0.028	0.732