

SUPPORTING INFORMATION

Ionic Liquid Mixtures – An Analysis of their Mutual Miscibility

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Table S1. Predicted LLE composition by COSMO-RS with different IL molecular models ([CA] and [C+A] models) at T=298 K for the 9 IL-IL mixtures reported in bibliography presenting two immiscible liquid phases.

IL 1 + IL 2	[CA] _{IL1} + [CA] _{IL2}		[C+A] _{IL1} + [C+A] _{IL2}	
	X ₂ ^{Phase 1}	X ₂ ^{Phase 2}	X ₂ ^{Phase 1}	X ₂ ^{Phase 2}
[P ₆₆₆₁₄][Cl] + [Pnmim][Cl]	0.204	0.795	Miscible	
[P ₆₆₆₁₄][Cl] + [Bmim][Cl]	0.129	0.870		
[P ₆₆₆₁₄][Cl] + [Prmim][Cl]	0.145	0.854		
[P ₆₆₆₁₄][Cl] + [Emim][Cl]	0.070	0.929		
[P ₆₆₆₁₄][Cl] + [Mmim][Cl]	0.052	0.947		
[P ₆₆₆₁₄][NTf ₂] + [Emim][NTf ₂]	0.290	0.979	0.033	0.966
[P ₆₆₆₁₄][NTf ₂] + [Prmpyr][NTF ₂]	0.223	0.986	Miscible	
[P ₆₆₆₁₄][NTf ₂] + [Epy][NTf ₂]	0.095	0.998	0.025	0.974
[P ₆₆₆₁₄][NTf ₂] + [Emim][MeSO ₃]	0.011	0.999	0.015	0.984

Table S2. Excess entropy (S^E) and activity coefficients of component 1 (γ_1) and 2 (γ_2) calculated by COSMO-RS, using [CA] model, at 298 K for binary IL-IL mixtures reported in bibliography.

	Component 1	Component 2	Ref.	$-T \cdot S^E$ (J/mol)	γ_1	γ_2
1	[1,3Bmpy][BF ₄]	[1,3Bmpy][DCN]	35	657	-0.19	-0.17
2	[Pnmim][NTf ₂]	[Pnmim][Br]	34	1341	0.08	0.14
3	[Emim][CF ₃ CO ₂]	[Emim][CH ₃ CO ₂]	7	619	-0.11	-0.10
4	[Bmim][EtSO ₄]	[Emim][NTf ₂]	9	693	0.03	-0.01
5	[Emim][BF ₄]	[Emim][DCN]	21	599	-0.03	-0.02
6	[Emim][EtSO ₄]	[Emim][NTf ₂]	9	749	0.10	0.06
7	[Prmpyr][NTf ₂]	[Prmpyr][DCN]	33	454	0.07	0.09
8	[Bmim][BF ₆]	Bmim][BF ₄]	20	188	-0.03	-0.02
9	[Emim][BF ₄]	[Emim][PF ₆]	6	182	-0.02	-0.02
10	[Emim][EtSO ₄]	[Emim][MeSO ₃]	6	176	-0.03	-0.02
11	[Bpy][BF ₄]	[1,4Bmpy][BF ₄]	5	97	-0.03	-0.02
12	[Bmim][BF ₄]	[Bmim][MeSO ₄]	20	55	-0.02	-0.01
13	[Pnmim][PF ₆]	[Pnmim][CF ₃ CO ₂]	34	81	0.01	0.01
14	[P ₄₄₄₄][NTf ₂]	[Bmim][NTf ₂]	17	164	0.04	0.08
15	[Emim][EtSO ₄]	[Emim][MeSO ₄]	6	8	0.01	0.01
16	[Prmpyr][NTf ₂]	[Empyr][NTf ₂]	33	0	0.00	0.00
17	[Pnmim][NTf ₂]	[Bmim][NTf ₂]	25	-1	0.01	0.00
18	[Prmpyr][NTf ₂]	[Bmpyr][NTf ₂]	33	-2	0.01	0.01
19	[Epy][EtSO ₄]	[Epy][MeSO ₄]	6	8	0.01	0.01
20	[Hxmim][NTf ₂]	[Bmim][NTf ₂]	25	-9	0.01	0.01
21	[Bpyr][NTf ₂]	[Epyr][NTf ₂]	33	-13	0.01	0.01
22	[N ₈₈₈₄][NTf ₂]	[N ₈₈₈₄][NO ₃]	4	130	0.05	0.09
23	[Emim][NTf ₂]	[Bmim][NTf ₂]	40	-13	0.02	0.01
24	[Bmpyr][NTf ₂]	[Mmpyr][NTf ₂]	33	-16	0.01	0.02
25	[Hxmim][BF ₄]	[Bmim][BF ₄]	20	-4	0.02	0.02
26	[1,3Bmpy][BF ₄]	[1,3Ompy][BF ₄]	35	-29	0.03	0.02
27	[Prmpyr][NTf ₂]	[Hxmpyr][NTf ₂]	33	-32	0.03	0.02
28	[Pnmpyr][NTf ₂]	[Emim][NTf ₂]	33	31	0.03	0.03
29	[Omim][NTf ₂]	[Bmim][NTf ₂]	25	-46	0.03	0.04
30	[Mmim][NTf ₂]	[Bmim][NTf ₂]	25	-43	0.04	0.03
31	[Pnmpyr][NTf ₂]	[Mmpyr][NTf ₂]	33	-56	0.03	0.04
32	[Emim][NTf ₂]	[Hxmim][NTf ₂]	39	-52	0.06	0.04
33	[Bpy][BF ₄]	[Bpy][NTf ₂]	23	3	0.09	0.05
34	[Bmim][BF ₄]	[Omim][BF ₄]	6	-24	0.08	0.07
35	[Dcmim][NTf ₂]	[Bmim][NTf ₂]	25	-82	0.05	0.08
36	[Pnmpyr][NTf ₂]	[Emim][NTf ₂]	33	-78	0.06	0.09
37	[Hxmim][BF ₄]	[Emim][BF ₄]	20	-56	0.08	0.11
38	[Bpy][BF ₄]	[1,4Bmpy][NTf ₂]	5	-57	0.15	0.10
39	[Pnmim][Cl]	[P ₆₆₆₁₄][Cl]	39	1101	0.91	0.42
40	[Bmim][Cl]	[P ₆₆₆₁₄][Cl]	39	1298	1.09	0.49
41	[Prmim][Cl]	[P ₆₆₆₁₄][Cl]	39	1475	1.24	0.51
42	[Emim][Cl]	[P ₆₆₆₁₄][Cl]	39	1756	1.44	0.56
43	[Mmim][Cl]	[P ₆₆₆₁₄][Cl]	39	2070	1.64	0.59
44	[Emim][NTf ₂]	[P ₆₆₆₁₄][NTf ₂]	39	-716	0.84	0.32
45	[Prmpyr][NTf ₂]	[P ₆₆₆₁₄][NTf ₂]	33	-829	0.92	0.37
46	[Epy][NTf ₂]	[P ₆₆₆₁₄][NTf ₂]	40	-954	1.29	0.50
47	[Emim][MeSO ₃]	[P ₆₆₆₁₄][NTf ₂]	39	-298	1.91	0.95

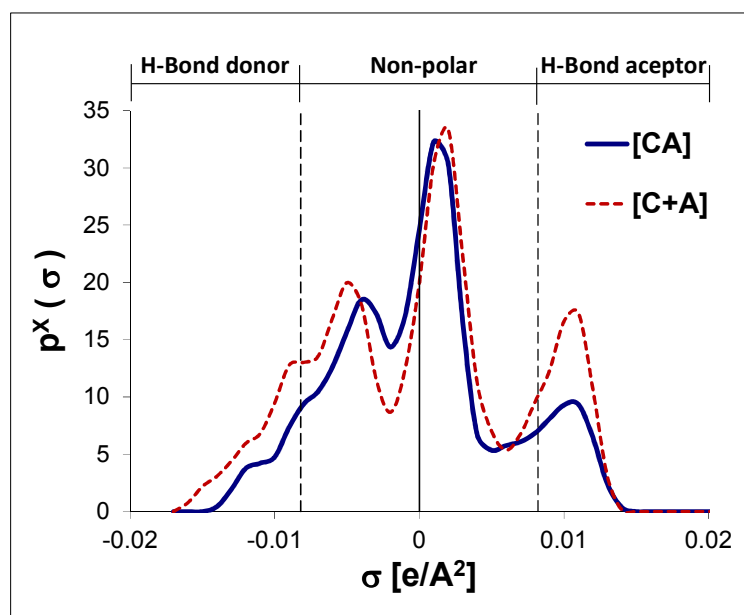


Figure S1. σ -Profile of [Emim][NTf₂] obtained using [CA] and [C+A] models in COSMO-RS calculations

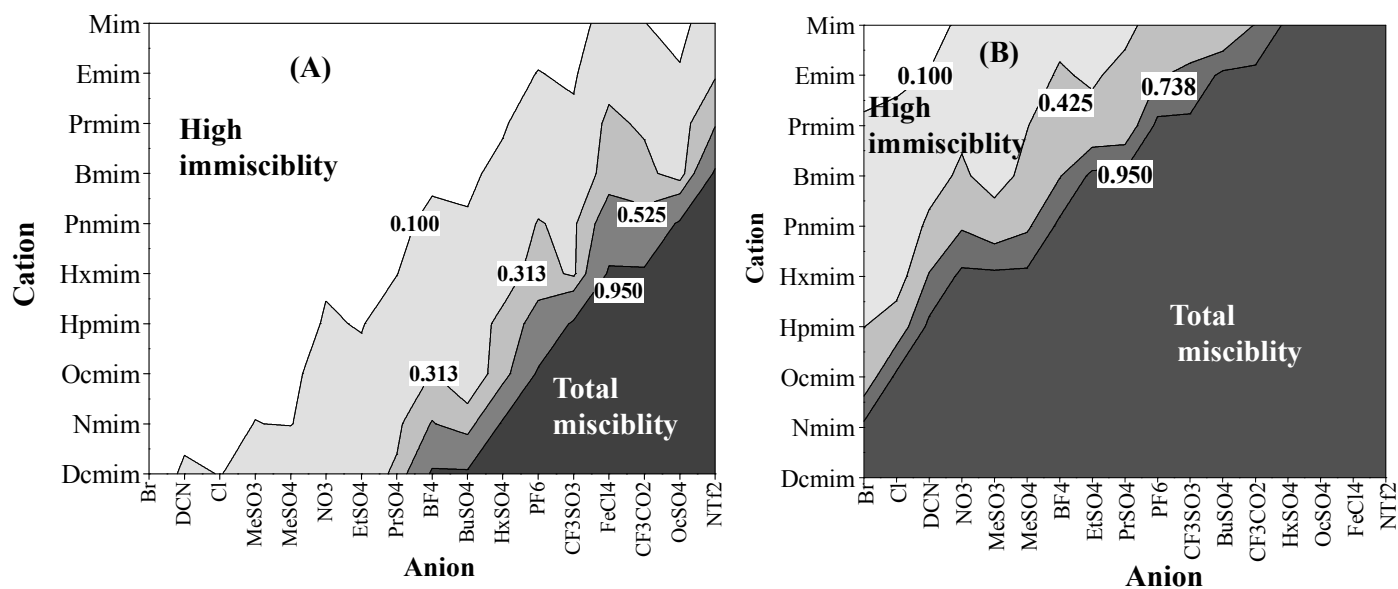


Figure S2. Screening of COSMO-RS predicted LLE composition ($X_{\text{Imidazolium-based IL}}$) of imidazolium-based IL in (A) [P₆₆₆₁₄][NTf₂] and (B) [P₆₆₆₁₄][Cl] rich phase for 170 binary equimolar mixtures at T= 298 K.