

## Supporting Information

# Aqueous biphasic systems composed of ionic liquids and acetate-based salts: phase diagrams, densities and viscosities

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**Table S1.** Experimental weight fraction data for the binodal curves of the systems composed of IL + KCH<sub>3</sub>CO<sub>2</sub> + H<sub>2</sub>O at 298 K and atmospheric pressure (0.1 MPa).<sup>a</sup>

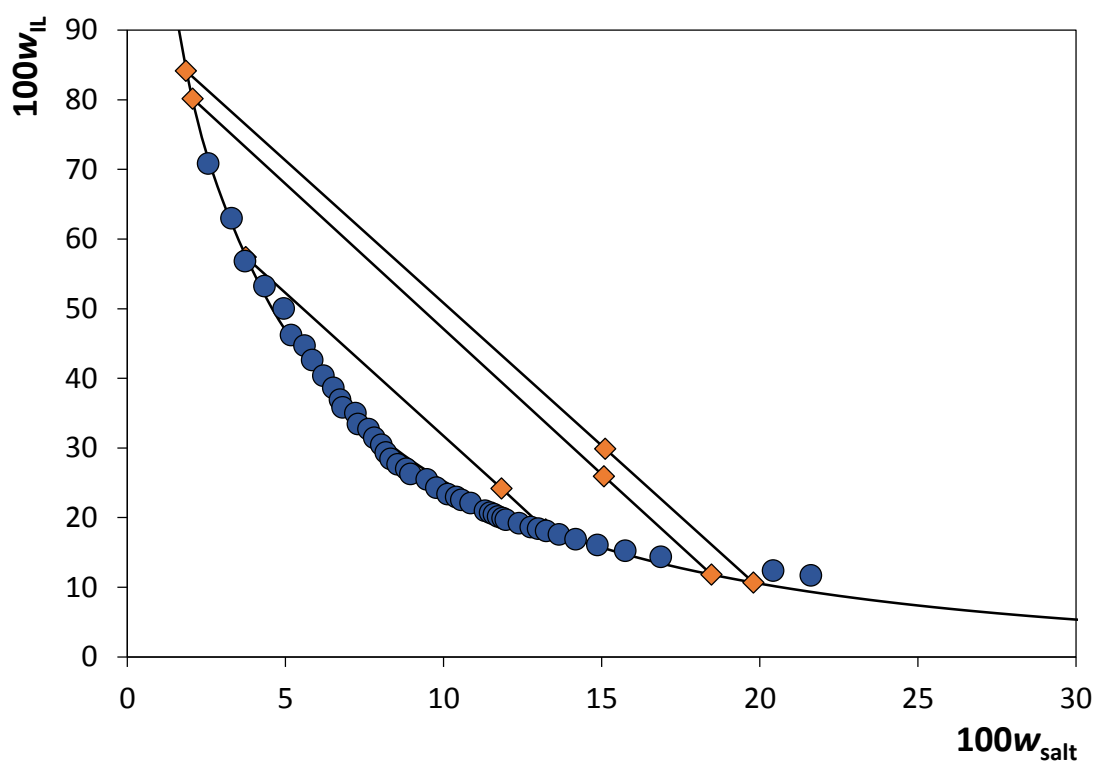
[C <sub>4</sub> mim][CF <sub>3</sub> SO <sub>3</sub> ] <i>M<sub>w</sub></i> = 252.23 g·mol <sup>-1</sup>		[C <sub>4</sub> mim][SCN] <i>M<sub>w</sub></i> = 197.19 g·mol <sup>-1</sup>		[C <sub>4</sub> mim][N(CN) <sub>2</sub> ] <i>M<sub>w</sub></i> = 205.15 g·mol <sup>-1</sup>			
100 <i>w</i> <sub>IL</sub>	100 <i>w</i> <sub>salt</sub>	100 <i>w</i> <sub>IL</sub>	100 <i>w</i> <sub>salt</sub>	100 <i>w</i> <sub>IL</sub>	100 <i>w</i> <sub>salt</sub>		
70.88	2.54	24.33	9.77	67.16	1.46	72.95	9.05
63.04	3.28	23.41	10.12	61.81	3.71	69.76	9.63
56.87	3.72	23.03	10.40	57.14	5.67	65.65	11.52
53.30	4.33	22.57	10.56	54.27	6.54	61.65	13.08
50.07	4.94	22.12	10.85	50.11	8.27	56.45	15.20
46.25	5.17	21.03	11.31	46.81	9.70	54.09	16.35
44.75	5.59	20.75	11.46	39.28	12.12	51.82	17.76
42.69	5.84	20.52	11.59	33.62	13.94	46.68	20.37
40.44	6.19	20.22	11.72	26.94	16.66	46.23	20.92
38.67	6.50	20.00	11.86	17.78	21.25	42.78	22.82
36.98	6.72	19.72	11.96	13.11	24.78	39.52	24.69
35.86	6.80	19.22	12.37			38.18	25.43
35.06	7.20	18.69	12.75			35.99	27.05
33.47	7.29	18.44	12.99			33.51	28.85
32.77	7.62	18.12	13.24			30.44	30.66
31.49	7.80	17.62	13.64			28.13	32.33
30.49	8.02	16.94	14.17				
29.41	8.17	16.12	14.86				
28.48	8.32	15.28	15.74				
27.68	8.55	14.42	16.86				
27.05	8.81	12.45	20.42				
26.30	8.95	11.75	21.61				
25.54	9.47						

<sup>a</sup> The standard uncertainty for the weight fraction  $u(100 w)$  is 0.01, the standard uncertainty for the temperature  $u(T)$  is 1 K, and the standard uncertainty for pressure  $u(P)$  is 10 kPa.

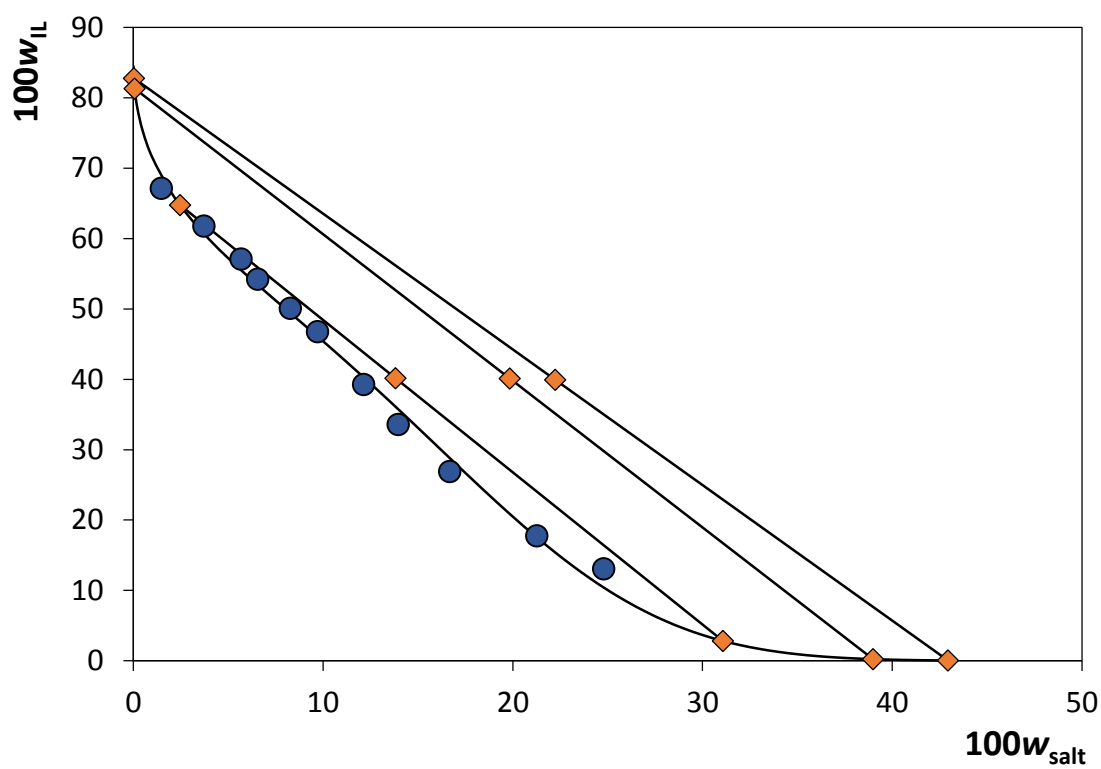
**Table S2.** Experimental weight fraction data for the binodal curves of the systems composed of IL + NaCH<sub>3</sub>CO<sub>2</sub> + H<sub>2</sub>O at 298 K and atmospheric pressure (0.1 MPa).<sup>a</sup>

[C <sub>4</sub> mim][CF <sub>3</sub> SO <sub>3</sub> ] <i>M<sub>w</sub></i> = 252.23 g·mol <sup>-1</sup>				[C <sub>4</sub> mim][SCN] <i>M<sub>w</sub></i> = 197.19 g·mol <sup>-1</sup>	
100 <i>w</i> <sub>IL</sub>	100 <i>w</i> <sub>salt</sub>	100 <i>w</i> <sub>IL</sub>	100 <i>w</i> <sub>salt</sub>	100 <i>w</i> <sub>IL</sub>	100 <i>w</i> <sub>salt</sub>
69.70	2.27	26.44	6.43	77.84	0.97
61.23	2.61	25.42	6.74	61.37	3.76
56.59	3.01	23.84	7.05	40.96	10.82
46.58	3.83	22.64	7.27	35.14	12.28
45.25	3.91	22.09	7.44	30.59	13.74
44.23	4.08	21.33	7.58	26.91	15.08
42.65	4.44	20.79	7.83	25.23	15.69
40.91	4.53	20.03	8.18	23.52	16.35
39.42	4.55	19.29	8.46	22.21	16.93
38.29	4.83	18.65	8.76	19.61	18.10
37.20	4.97	17.90	9.14	12.49	21.94
35.77	5.10	17.32	9.41		
34.79	5.29	16.56	9.88		
33.42	5.54	15.78	10.33		
31.77	5.57	14.64	11.16		
30.69	5.89	14.02	11.59		
29.28	6.04	13.02	12.39		
27.74	6.31	11.81	13.56		

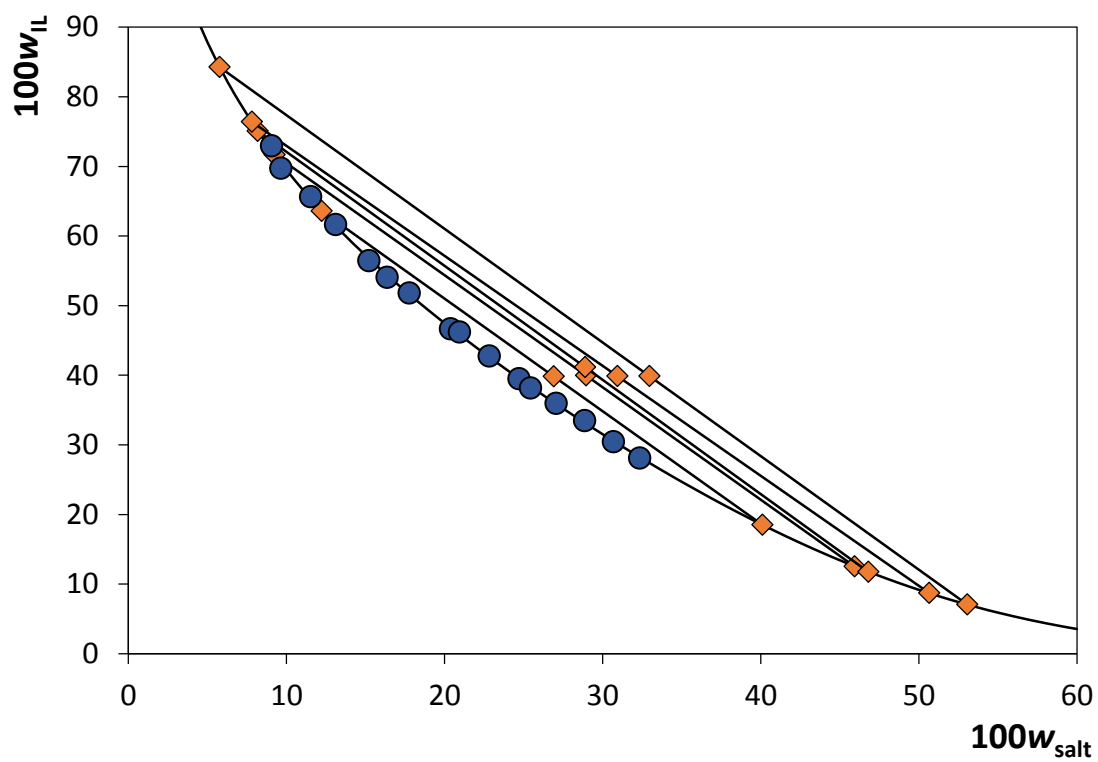
<sup>a</sup> The standard uncertainty for the weight fraction  $u(100 w)$  is 0.01, the standard uncertainty for the temperature  $u(T)$  is 1 K, and the standard uncertainty for pressure  $u(P)$  is 10 kPa.



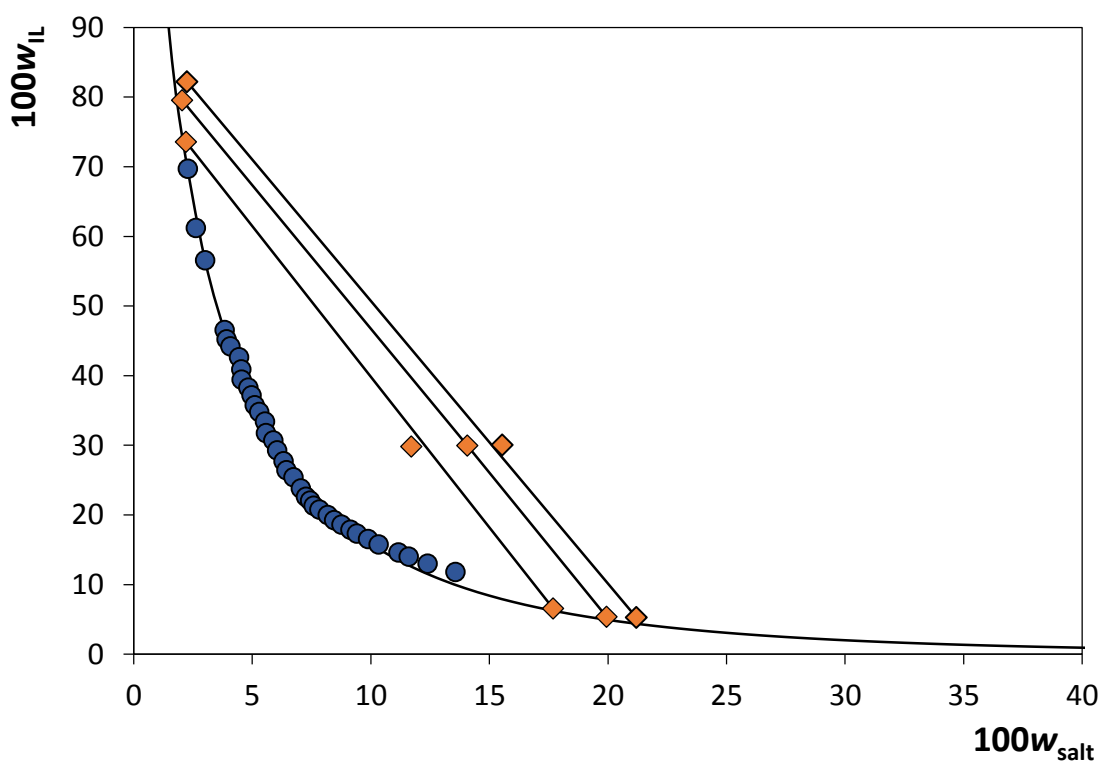
**Fig S1.** Phase diagram for the ternary system composed of [C<sub>4</sub>mim][CF<sub>3</sub>SO<sub>3</sub>] + KCH<sub>3</sub>CO<sub>2</sub> + H<sub>2</sub>O: binodal curve data (●); TL data (◆); adjusted binodal data through Equation 1 (—).



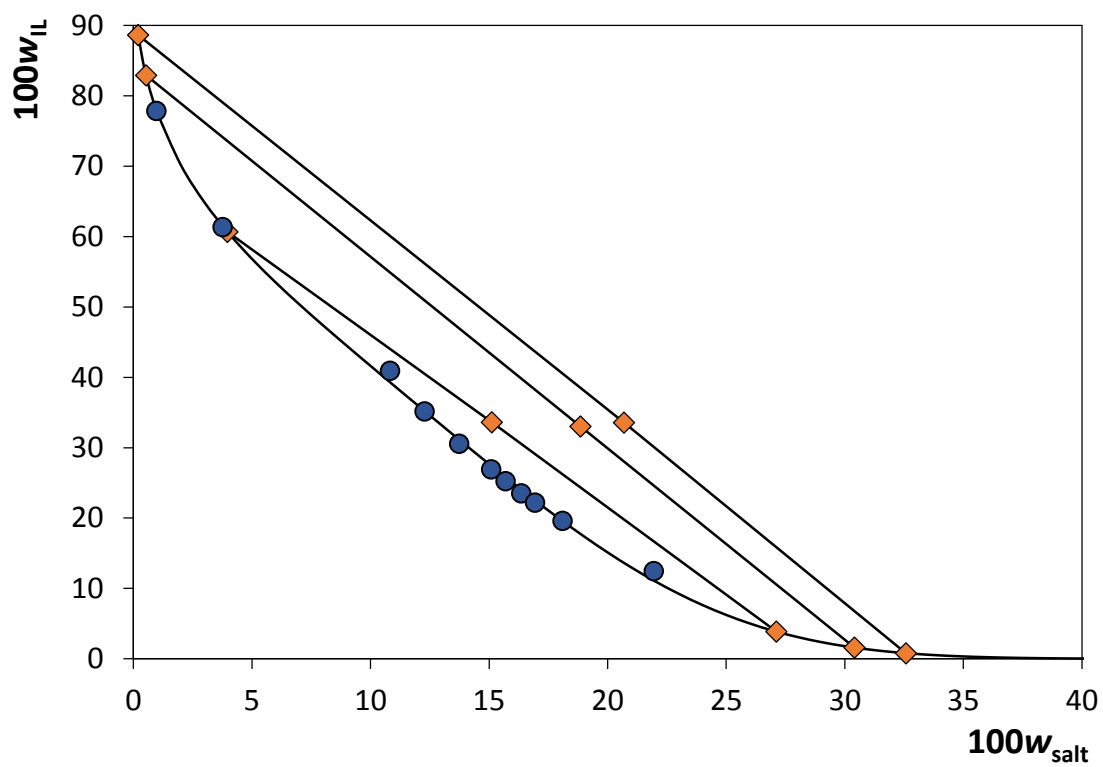
**Fig S2.** Phase diagram for the ternary system composed of  $[\text{C}_4\text{mim}][\text{SCN}] + \text{KCH}_3\text{CO}_2 + \text{H}_2\text{O}$ : binodal curve data (●); TL data (◆); adjusted binodal data through Equation 1 (—).



**Fig S3.** Phase diagram for the ternary system composed of  $[\text{C}_4\text{mim}][\text{N}(\text{CN})_2] + \text{KCH}_3\text{CO}_2 + \text{H}_2\text{O}$ : binodal curve data (●); TL data (◆); adjusted binodal data through Equation 1 (—).

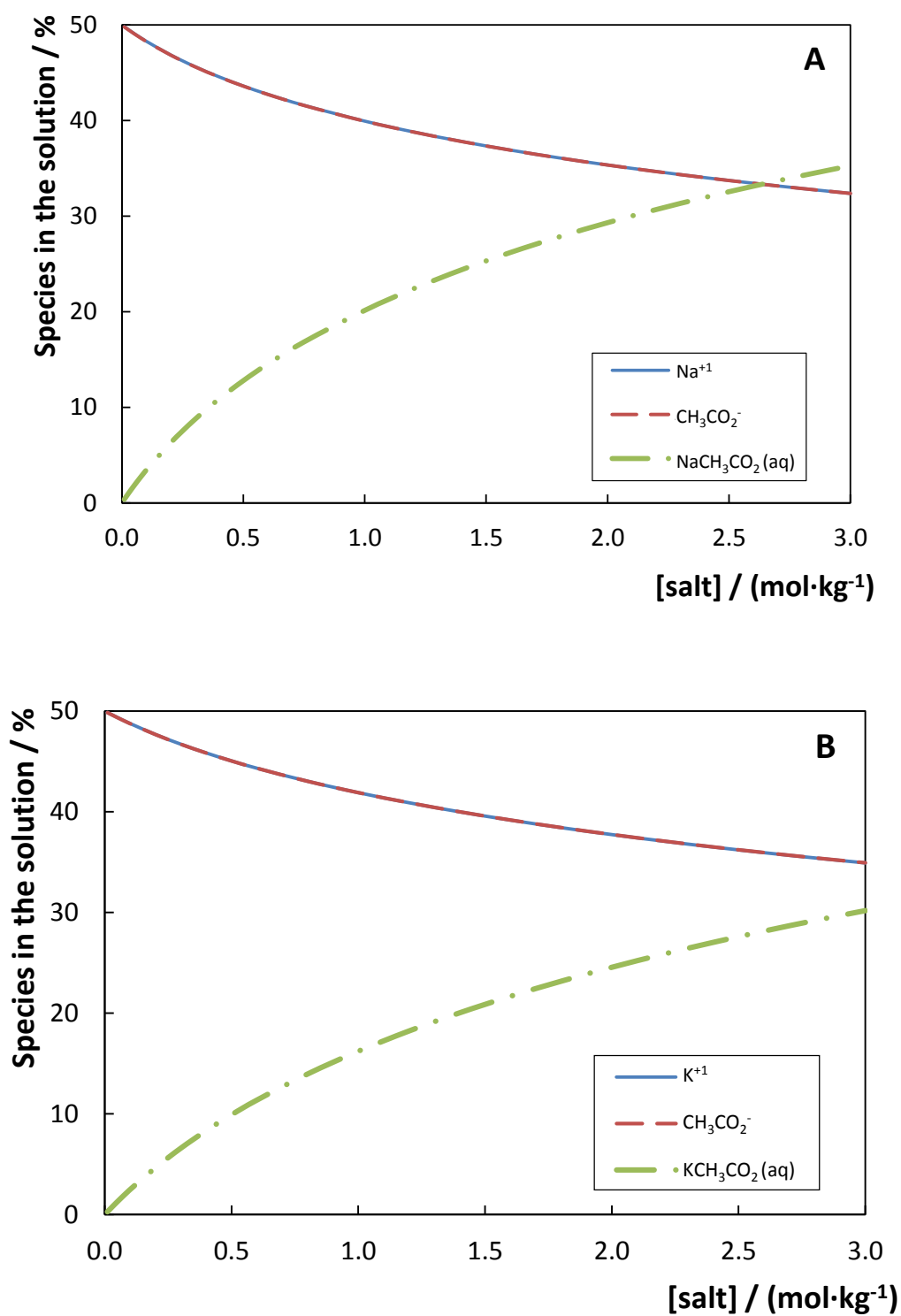


**Fig S4.** Phase diagram for the ternary system composed of [C<sub>4</sub>mim][CF<sub>3</sub>SO<sub>3</sub>] + NaCH<sub>3</sub>CO<sub>2</sub> + H<sub>2</sub>O: binodal curve data (●); TL data (◆); adjusted binodal data through Equation 1 (—).

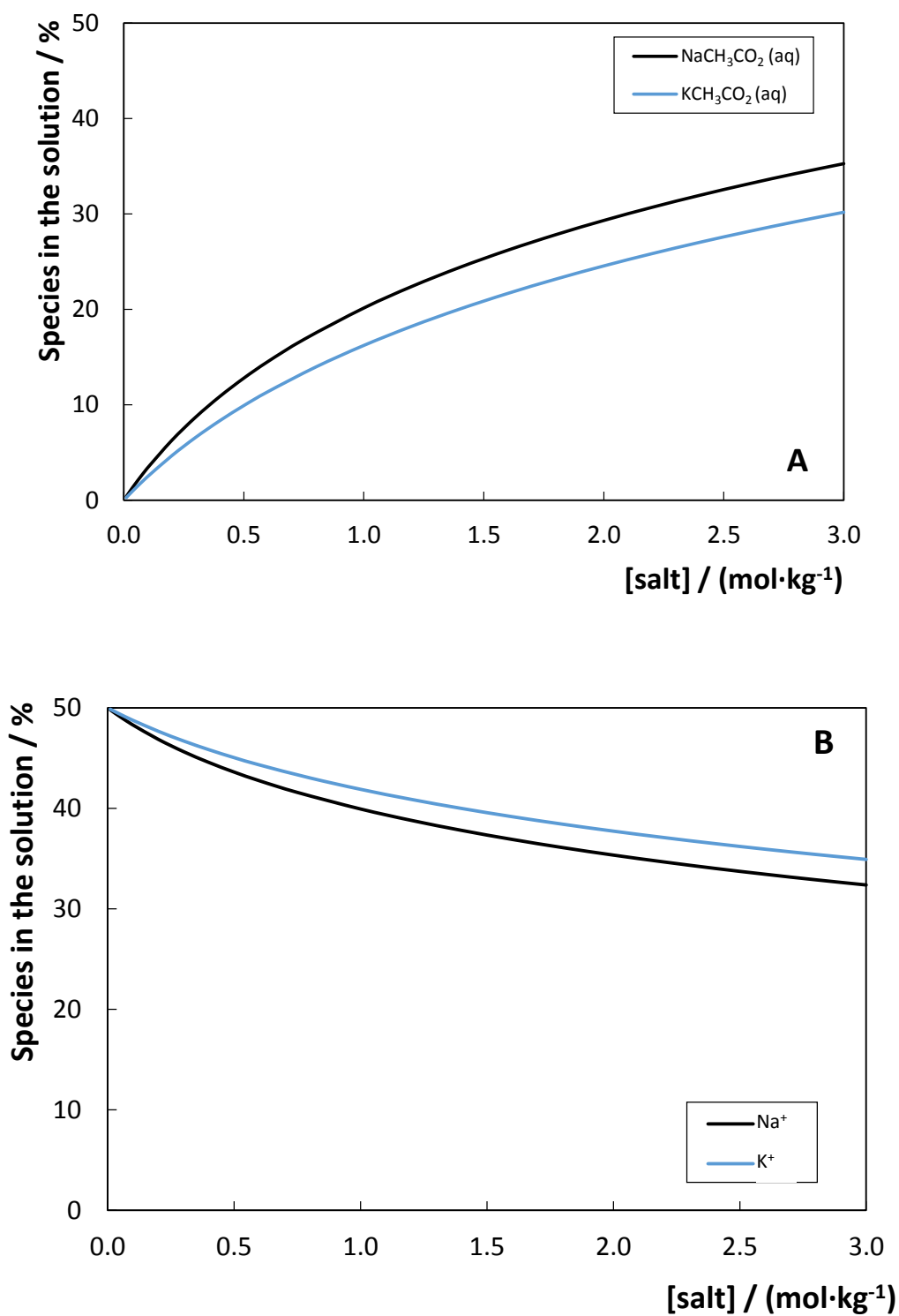


**Fig S5.** Phase diagram for the ternary system composed of [C<sub>4</sub>mim][SCN] + NaCH<sub>3</sub>CO<sub>2</sub> + H<sub>2</sub>O: binodal curve data (●); TL data (◆); adjusted binodal data through Equation 1 (–).





**Fig S6.** Ions speciation profiles of (A) NaCH<sub>3</sub>CO<sub>2</sub> and (B) KCH<sub>3</sub>CO<sub>2</sub> in aqueous solutions at 298 K (taking into account the experimentally measured pH values of the solutions) estimated by VisualMINTEQ.



**Fig S7.** Ions speciation profiles comparison of (A) CH<sub>3</sub>CO<sub>2</sub>-based salts in aqueous solutions and (B) K<sup>+</sup> and Na<sup>+</sup> cations at 298 K (taking into account the experimentally measured pH values of the solutions ) estimated by VisualMINTEQ.

**Table S3.** Critical point of each system composed of IL + salt + H<sub>2</sub>O at 298 K and atmospheric pressure (0.1 MPa).<sup>a</sup>

IL	salt	$f$	$g$	$R^2$	Critical point		
					100 $w_{\text{IL}}$	100 $w_{\text{salt}}$	100 $w_{\text{H}_2\text{O}}$
[C <sub>4</sub> mim][CF <sub>3</sub> SO <sub>3</sub> ]		4.05 ± 0.18	4.48 ± 3.14	0.998	34.20	7.38	58.42
[C <sub>4</sub> mim][SCN]	KCH <sub>3</sub> CO <sub>2</sub>	1.60 ± 0.42	15.97 ± 16.04	0.935	37.32	13.33	49.35
[C <sub>4</sub> mim][N(CN) <sub>2</sub> ]		1.47 ± 0.21	4.43 ± 12.72	0.944	40.23	24.28	35.49
[C <sub>4</sub> mim][CF <sub>3</sub> SO <sub>3</sub> ]	NaCH <sub>3</sub> CO <sub>2</sub>	2.49 ± 0.14	29.61 ± 2.69	0.997	40.64	4.43	54.92
[C <sub>4</sub> mim][SCN]		5.25 ± 1.11	-80.17 ± 33.50	0.957	18.06	18.72	63.22

<sup>a</sup> The standard uncertainty for the weight fraction  $u(100 w)$  is 0.01, the standard uncertainty for the temperature  $u(T)$  is 1 K, and the standard uncertainty for pressure  $u(P)$  is 10 kPa.

**Table S4.** Viscosity of the top ( $T$ ) and bottom phases ( $B$ ) for the systems composed of IL +  $\text{KCH}_3\text{CO}_2 + \text{H}_2\text{O}$  (equilibrated at 298.15 K) and atmospheric pressure (0.1 MPa).<sup>a</sup>

<b><math>\text{KCH}_3\text{CO}_2</math></b>										
$T / \text{K}$	[C <sub>4</sub> mim][CF <sub>3</sub> SO <sub>3</sub> ] TL1		[C <sub>4</sub> mim][SCN] TL1		[C <sub>4</sub> mim][SCN] TL2		[C <sub>4</sub> mim][N(CN) <sub>2</sub> ] TL1		[C <sub>4</sub> mim][N(CN) <sub>2</sub> ] TL2	
	$\eta_T /$ (mPa·s)	$\eta_B /$ (mPa·s)	$\eta_T /$ (mPa·s)	$\eta_B /$ (mPa·s)	$\eta_T /$ (mPa·s)	$\eta_B /$ (mPa·s)	$\eta_T /$ (mPa·s)	$\eta_B /$ (mPa·s)	$\eta_T /$ (mPa·s)	$\eta_B /$ (mPa·s)
298.15	1.9625	5.9913	7.3317	2.9520	8.5128	3.1953	12.0800	8.2306	13.4050	9.9793
303.15	1.7417	5.0248	6.2160	2.6013	7.0631	2.8009	-	-	11.2220	8.3976
308.15	1.5147	4.3524	5.3500	2.3145	5.9957	2.4852	8.6297	6.0234	9.5467	7.1840
313.15	1.3147	3.7894	4.6254	2.0572	5.1567	2.2086	-	-	8.1801	6.1818
318.15	1.2531	3.3777	4.0930	1.8838	4.5476	2.0114	6.4882	4.6146	7.1684	5.4495
323.15	1.1515	3.0251	3.6235	1.7157	4.0163	1.8288	-	-	6.2998	4.8152
328.15	1.0595	2.7350	3.2269	1.5745	3.5793	1.6735	5.0570	3.6635	-	-

<sup>a</sup> The standard uncertainty in temperature  $u(T)$  is 0.02 K, the standard uncertainty for pressure  $u(P)$  is 10 kPa, and the combined expanded uncertainty in viscosity  $U(\eta)$  is 0.35 % with 95 % confidence level.

**Table S5.** Viscosity of the top ( $T$ ) and bottom phases ( $B$ ) for the systems composed of IL + NaCH<sub>3</sub>CO<sub>2</sub> + H<sub>2</sub>O (equilibrated at 298.15 K) and atmospheric pressure (0.1 MPa).<sup>a</sup>

$T / \text{K}$	<b>NaCH<sub>3</sub>CO<sub>2</sub></b>					
	[C <sub>4</sub> mim][CF <sub>3</sub> SO <sub>3</sub> ] TL1		[C <sub>4</sub> mim][SCN] TL1		[C <sub>4</sub> mim][SCN] TL2	
	$\eta_T /$ (mPa·s)	$\eta_B /$ (mPa·s)	$\eta_T /$ (mPa·s)	$\eta_B /$ (mPa·s)	$\eta_T /$ (mPa·s)	$\eta_B /$ (mPa·s)
298.15	3.0276	11.1220	7.9117	5.2143	8.54540	5.23450
303.15	2.6053	9.4382	6.6993	4.3763	7.09410	4.39850
308.15	2.2787	8.1056	5.7411	3.7323	5.96920	3.75870
313.15	1.9896	6.9824	4.9388	3.2064	5.07600	3.23250
318.15	1.7982	6.1502	4.3491	2.8205	4.45730	2.84610
323.15	1.6172	5.4253	3.838	2.4918	3.92400	2.51550
328.15	1.4650	4.7748	3.4133	2.2201	3.48330	2.24370

<sup>a</sup>The standard uncertainty in temperature  $u(T)$  is 0.02 K, the standard uncertainty for pressure  $u(P)$  is 10 kPa, and the combined expanded uncertainty in viscosity  $U(\eta)$  is 0.35 % with 95 % confidence level.

**Table S6.** Density of the top ( $T$ ) and bottom phases ( $B$ ) for the systems composed of IL +  $\text{KCH}_3\text{CO}_2 + \text{H}_2\text{O}$  (equilibrated at 298.15 K) and atmospheric pressure (0.1 MPa).<sup>a</sup>

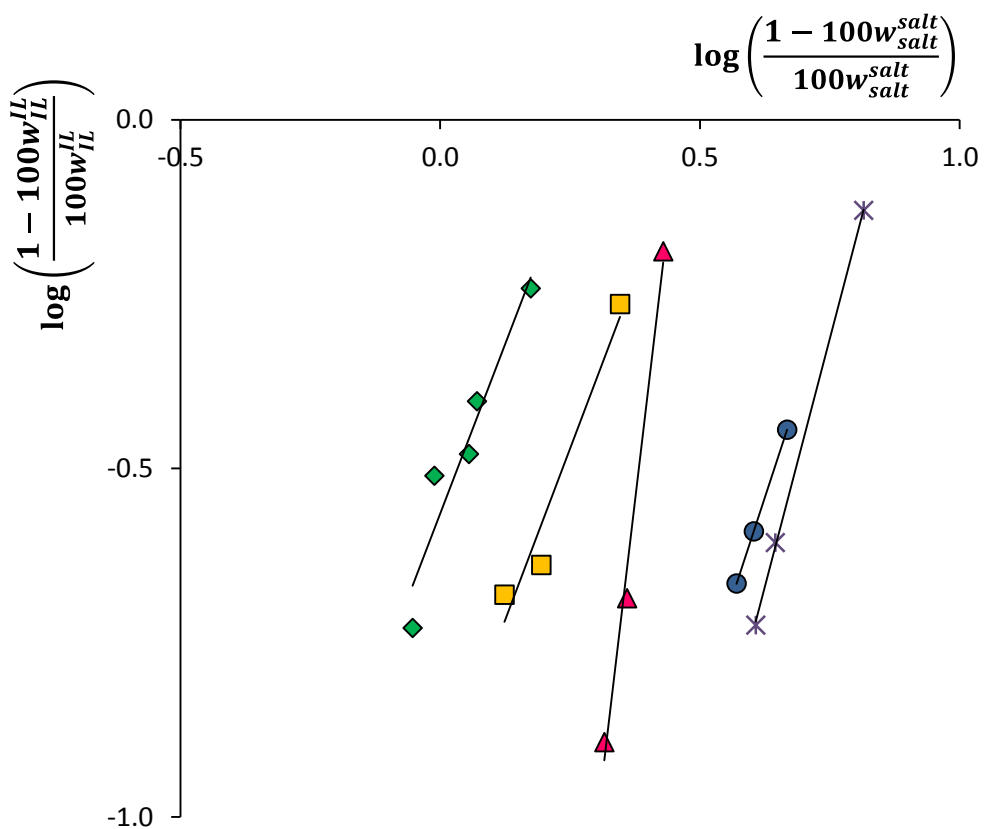
<b>KCH<sub>3</sub>CO<sub>2</sub></b>										
$T / \text{K}$	[C <sub>4</sub> mim][CF <sub>3</sub> SO <sub>3</sub> ] TL1		[C <sub>4</sub> mim][SCN] TL1		[C <sub>4</sub> mim][SCN] TL2		[C <sub>4</sub> mim][N(CN) <sub>2</sub> ] TL1		[C <sub>4</sub> mim][N(CN) <sub>2</sub> ] TL2	
	$\rho_T /$ (g·cm <sup>-3</sup> )	$\rho_B /$ (g·cm <sup>-3</sup> )	$\rho_T /$ (g·cm <sup>-3</sup> )	$\rho_B /$ (g·cm <sup>-3</sup> )	$\rho_T /$ (g·cm <sup>-3</sup> )	$\rho_B /$ (g·cm <sup>-3</sup> )	$\rho_T /$ (g·cm <sup>-3</sup> )	$\rho_B /$ (g·cm <sup>-3</sup> )	$\rho_T /$ (g·cm <sup>-3</sup> )	$\rho_B /$ (g·cm <sup>-3</sup> )
298.15	1.1079	1.2058	1.0707	1.1746	1.0706	1.1918	1.0822	1.272	1.0817	1.2878
303.15	1.1053	1.202	1.0673	1.1716	1.0673	1.1889	-	-	1.0783	1.2843
308.15	1.1026	1.1982	1.0640	1.1688	1.0640	1.1860	1.0755	1.2654	1.0749	1.2809
313.15	1.0998	1.1943	1.0607	1.1659	1.0607	1.1831	-	-	1.0715	1.2775
318.15	1.0969	1.1903	1.0573	1.1629	1.0574	1.1801	1.0691	1.2589	1.0682	1.2741
323.15	1.0939	1.1862	1.0540	1.1599	1.0541	1.1771	-	-	1.0648	1.2707
328.15	1.0909	1.1820	1.0507	1.1569	1.0507	1.1741	1.0638	1.2522	-	-

<sup>a</sup> The standard uncertainty in temperature  $u(T)$  is 0.02 K, the standard uncertainty for pressure  $u(P)$  is 10 kPa, and the combined expanded uncertainty in density  $U(\rho)$  is  $5 \times 10^{-4} \text{ g}\cdot\text{cm}^{-3}$  with 95 % confidence level.

**Table S7.** Density of the top (*T*) and bottom phases (*B*) for systems composed of IL + NaCH<sub>3</sub>CO<sub>2</sub> + H<sub>2</sub>O (equilibrated at 298.15 K) and atmospheric pressure (0.1 MPa).<sup>a</sup>

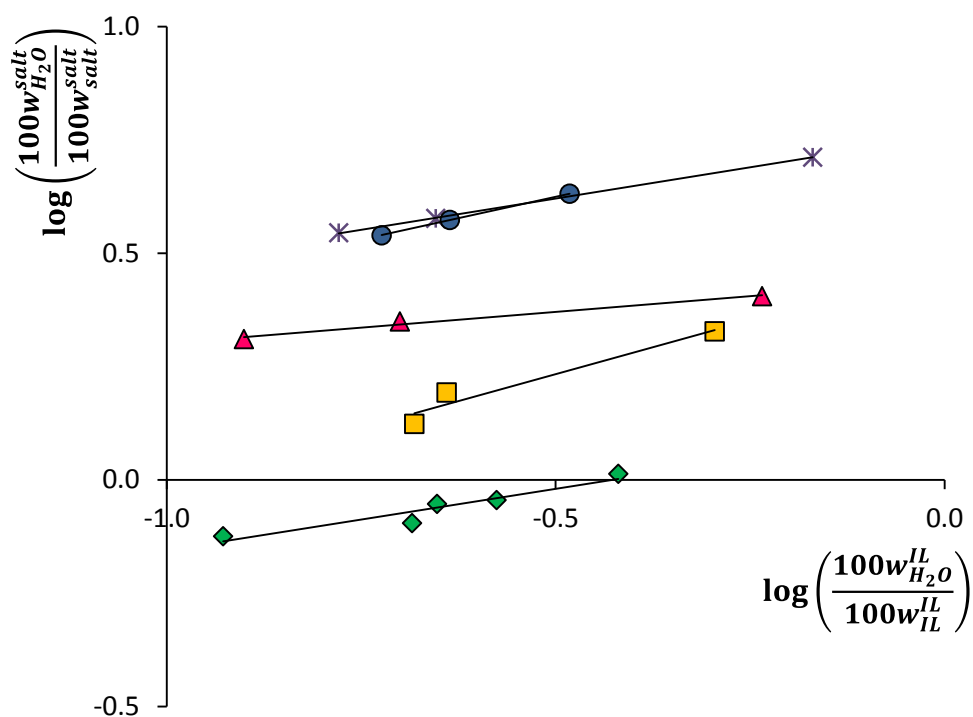
<i>T</i> / K	NaCH <sub>3</sub> CO <sub>2</sub>					
	[C <sub>4</sub> mim][CF <sub>3</sub> SO <sub>3</sub> ] TL1		[C <sub>4</sub> mim][SCN] TL1		[C <sub>4</sub> mim][SCN] TL2	
	$\rho_T /$ (g·cm <sup>-3</sup> )	$\rho_B /$ (g·cm <sup>-3</sup> )	$\rho_T /$ (g·cm <sup>-3</sup> )	$\rho_B /$ (g·cm <sup>-3</sup> )	$\rho_T /$ (g·cm <sup>-3</sup> )	$\rho_B /$ (g·cm <sup>-3</sup> )
298.15	1.1318	1.2509	1.0667	1.1594	-	1.1590
303.15	1.1290	1.2466	1.0633	1.1563	1.0460	1.1559
308.15	1.1262	1.2424	1.0599	1.1532	1.0426	1.1528
313.15	1.1233	1.2382	1.0566	1.1501	1.0391	1.1497
318.15	1.1204	1.2341	1.0532	1.1469	1.0356	1.1466
323.15	1.1173	1.2298	1.0498	1.1437	1.0321	1.1434
328.15	1.1142	1.2251	1.0465	1.1404	1.0285	1.1401

<sup>a</sup>The standard uncertainty in temperature  $u(T)$  is 0.02 K, the standard uncertainty for pressure  $u(P)$  is 10 kPa, and the combined expanded uncertainty in density  $U(\rho)$  is  $5 \times 10^{-4}$  g·cm<sup>-3</sup> with 95 % confidence level.



**Figure S8.** Othmer-Tobias correlation for each ternary system composed of salt + IL + H<sub>2</sub>O (cf. Table 3): [C<sub>4</sub>mim][CF<sub>3</sub>SO<sub>3</sub>] + NaCH<sub>3</sub>CO<sub>2</sub> (●); [C<sub>4</sub>mim][N(CN)<sub>2</sub>] + KCH<sub>3</sub>CO<sub>2</sub> (◆); [C<sub>4</sub>mim][SCN] + KCH<sub>3</sub>CO<sub>2</sub> (■); [C<sub>4</sub>mim][SCN] + NaCH<sub>3</sub>CO<sub>2</sub> (▲); [C<sub>4</sub>mim][CF<sub>3</sub>SO<sub>3</sub>] + KCH<sub>3</sub>CO<sub>2</sub> (\*).





**Figure S9.** Bancroft correlation for each ternary system composed of salt + IL + H<sub>2</sub>O (*cf.*

Table 3): [C<sub>4</sub>mim][CF<sub>3</sub>SO<sub>3</sub>] + NaCH<sub>3</sub>CO<sub>2</sub> (●); [C<sub>4</sub>mim][N(CN)<sub>2</sub>] + KCH<sub>3</sub>CO<sub>2</sub> (◆);

[C<sub>4</sub>mim][SCN] + KCH<sub>3</sub>CO<sub>2</sub> (■); [C<sub>4</sub>mim][SCN] + NaCH<sub>3</sub>CO<sub>2</sub> (▲); [C<sub>4</sub>mim][CF<sub>3</sub>SO<sub>3</sub>] +

KCH<sub>3</sub>CO<sub>2</sub> (\*).