

Supplementary Information

Performance enhancement of aqueous ionic liquids with lower critical solution temperature (LCST) behavior through ternary mixtures

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Supplementary Note 1: NSal+PTFA Mixtures

Herein we show the properties of the eight NSal+PTFA mixtures, with comparisons to each mixture's precursor ILs at the same total IL composition as shown in Table S1 and S2. Table S3 shows the osmolality and phase separation temperatures of the NSal+PTFA mixtures at constant relative mixing ratios, $r = \frac{w_{NSal}(\%)}{w_{PTFA}(\%)}$, of 0.6, 1.0, 1.67, and 2.0.

Table S1: The performance of the NSal+PTFA mixtures relative to their precursor ILs. All osmolality measurements shown are based on five water activity measurements with a maximum standard deviation of 16% in osmolality. The maximum error in the cloud point measurements for T_c is 0.5 °C.

IL Mixture	Osmolality (mmol kg ⁻¹)	T_c (°C)	Osmolality Difference from NSal (mmol kg ⁻¹)	Osmolality Difference from PTFA (mmol kg ⁻¹)	T_c Difference from NSal (°C)	T_c Difference from PTFA (°C)
20 wt.% NSal + 10 wt.% PTFA	560.6 ± 46.4	40.0	170.3	-87.4	-6.5	9.0
10 wt.% NSal + 20 wt.% PTFA	604.4 ± 61.2	39.0	214.1	-43.6	-7.5	8.0
25 wt.% NSal + 15 wt.% PTFA	593.2 ± 48.5	41.5	134.6	-96.8	-3.5	10.5
15 wt.% NSal + 25 wt.% PTFA	831.8 ± 27.8	39	373.2	141.8	-6.0	8.0
25 wt.% NSal + 25 wt.% PTFA	774.3 ± 50.3	42.5	145.2	86.3	-6.5	10.5
30 wt.% NSal + 25 wt.% PTFA	875.8 ± 40.6	47.5	40.2	85.3	-9.5	11.5
25 wt.% NSal + 30 wt.% PTFA	878.0 ± 22.4	47.0	42.5	87.5	-10.0	11.0
30 wt.% NSal + 30 wt.% PTFA	1063.5 ± 44.9	51.0	21.5	170.5	-14.0	11.0

Table S2: Phase performance of the NSal+PTFA mixtures relative to their precursor ILs based on a phase separation at $T_{sep} = 70$ °C for 30 min in a water bath heater. All osmolality measurements shown are based on five water activity measurements with a maximum standard deviation of 16% in osmolality.

IL Mixture	WR Osmolality (mmol kg ⁻¹)	PMR	WR Osmolality Difference from NSal (mmol kg ⁻¹)	WR Osmolality Difference from PTFA (mmol kg ⁻¹)	PMR Difference from NSal	PMR Difference from PTFA
20 wt.% NSal + 10 wt.% PTFA	363.4 ± 24.8	1.61	121.8	-22.4	0.17	-0.78
10 wt.% NSal + 20 wt.% PTFA	405.9 ± 29.6	2.15	164.3	20.1	0.71	-0.24
25 wt.% NSal + 15 wt.% PTFA	431.7 ± 37.8	0.72	88.4	38.1	0.06	-0.18
15 wt.% NSal + 25 wt.% PTFA	444.0 ± 41.5	0.83	100.7	50.4	0.16	-0.08
25 wt.% NSal + 25 wt.% PTFA	553.9 ± 38.3	0.30	171.5	108.8	0.09	-0.11
30 wt.% NSal + 25 wt.% PTFA	597.7 ± 46.1	0.15	111.5	146.4	0.02	-0.11
25 wt.% NSal + 30 wt.% PTFA	458.6 ± 51.9	0.24	-27.6	7.3	0.12	-0.01
30 wt.% NSal + 30 wt.% PTFA	522.5 ± 38.1	0.13	-67.4	65.0	0.09	0.03

Table S3: NSal+PTFA mixtures at different constant relative IL weight composition ratios

($r = \frac{w_{NSal} (\%)}{w_{PTFA} (\%)} = const.$). All osmolality measurements shown are based on five water activity measurements with a maximum standard deviation of 16% in osmolality. The maximum error in the cloud point measurements for T_C is 0.5 °C.

Total IL Weight Percent (%)	Osmolality (mmol/kg)	Osmolality Std. Dev. (mmol kg ⁻¹)	T_C (°C)
$r = 0.6$			
10	540.4	23.6	40.0
20	766.4	26.1	35.0
30	741.6	16.7	38.0
40	942.4	31.6	40.0
50	1014.8	12.7	42.5
60	1242.7	16.8	51.0
70	2150.0	24.4	/
$r = 1$			
10	619.0	54.5	40.0
20	704.5	33.3	35.0
30	778.8	46.4	38.0
40	878.0	20.5	40.0
50	1115.6	25.5	42.5
60	1255.2	15.4	51.5
70	2152.3	16.8	/
$r = 1.67$			
10	533.7	20.1	40.0
20	657.2	37.4	36.0
30	749.5	25.5	38.5
40	965.0	30.3	40.5
50	1072.5	29.8	44.0
60	1420.2	17.4	54.5
70	2546.4	10.1	/
$r = 2$			
10	449.6	27.9	40.0
20	603.3	26.0	35.5
30	698.8	27.6	38.5
40	843.0	29.5	40.5
50	1006.9	33.1	44.0
60	1365.5	23.6	54.0
70	2389.6	17.9	/

Supplementary Note 2: NSal+PSal Mixtures

Herein we show the results for NSal+PSal mixtures not shown in the main text. Fig. S1 shows the data plane of the mixtures, where the ordinates 0 and 1 on the y-axis represent the precursors ILs, PSal, and NSal respectively. The osmolality as shown in Fig. S1(a) is higher than either precursor IL above 35 wt.% total IL composition and is peak at the 0.6 NSal IL mixture ratio ordinate. The critical phase separation temperature, T_C , is lower than NSal across the plane, but is higher than PSal as shown in Fig. S1(b). The WR phase purity is improved over either precursor IL below 50 wt.% total IL composition as shown in Fig. S1(c). However, as Fig. S1(d) shows, minimal improvement to the PMR is seen over the precursor ILs. Table S4 and S5 show the raw data for six NSal+PSal mixtures, with comparisons to each mixture's precursor ILs at the same total IL composition.

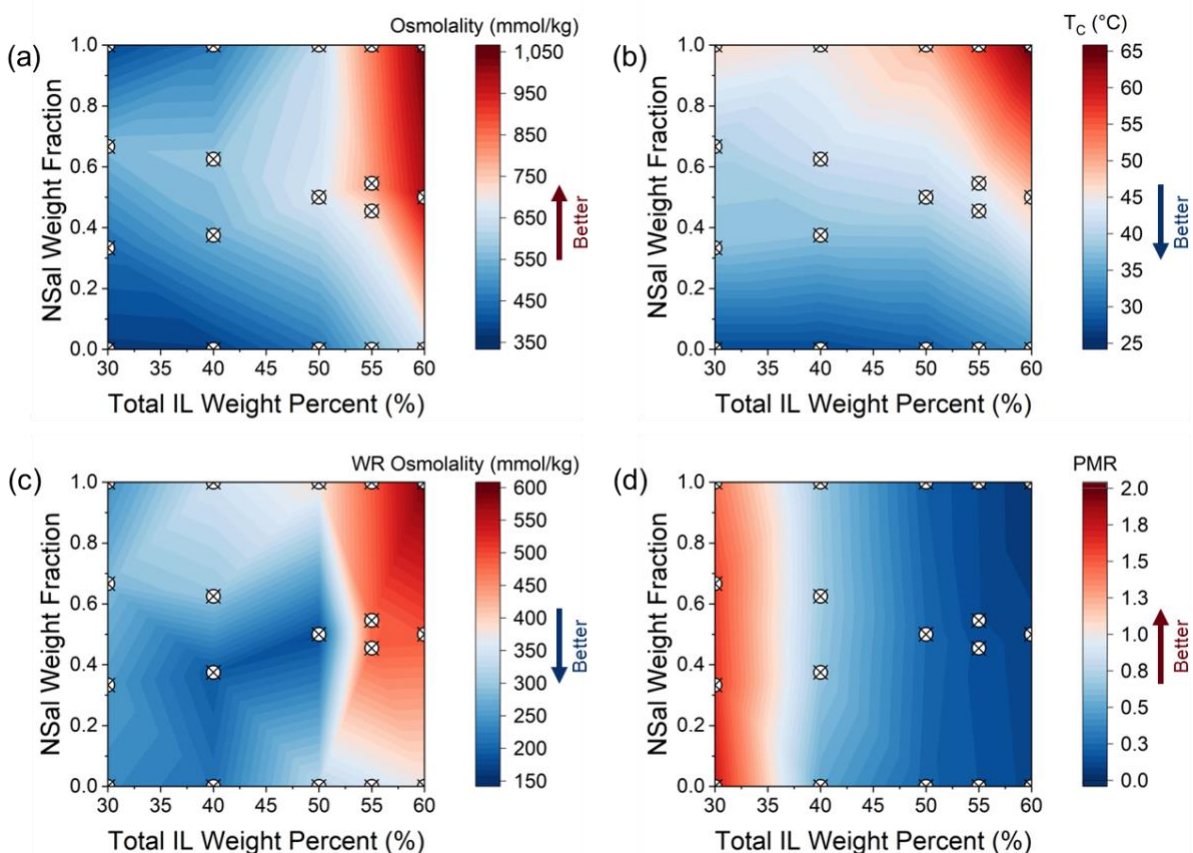


Fig. S1. Thermodynamic properties of the NSal+PSal ternary mixtures in terms of the weight fraction of NSal with respect to the total IL in the solution $\left(\frac{w_{NSal}(\%)}{w_{NSal}(\%) + w_{PSal}(\%)}\right)$ on the y-axis and the total IL weight percent in the solution on the x-axis (a) Osmolality. (b) Critical phase separation temperature, T_C . (c) WR phase osmolality. (d) WR phase to ILR phase mass ratio (PMR). All the data in (c) and (d) are based on a phase separation at $T_{sep} = 70^{\circ}\text{C}$ for 30 minutes in a water bath. All osmolality measurements shown in (a) and (c) are based on five water activity measurements with a maximum standard deviation of 16% in osmolality. The maximum error in the cloud point measurements for T_C shown in (b) is 0.5 $^{\circ}\text{C}$. The maximum error in the PMR measurements shown in (d) is 0.1%. The marked points correspond to measured data points as per the coordinates listed in Table 1 of the main manuscript.

Table S4: The performance of the NSal+PSal mixtures relative to their precursor ILs. All osmolality measurements shown are based on five water activity measurements with a maximum standard deviation of 16% in osmolality. The maximum error in the cloud point measurements for T_C is 0.5 °C.

IL Mixture	Osmolality (mmol kg ⁻¹)	T_C (°C)	Osmolality Difference from NSal (mmol kg ⁻¹)	Osmolality Difference from PSal (mmol kg ⁻¹)	T_C Difference from NSal (°C)	T_C Difference from PSal (°C)
20 wt.% NSal + 10 wt.% PSal	560.6 ± 34.3	39.5	170.3	184.9	-7.0	12.0
10 wt.% NSal + 20 wt.% PSal	444.0 ± 57.2	37.5	53.7	68.3	-9.0	6.5
25 wt.% NSal + 15 wt.% PSal	577.4 ± 29.9	41.5	118.9	196.1	-3.5	10.5
15 wt.% NSal + 25 wt.% PSal	543.8 ± 49.2	37.5	85.2	162.5	-7.5	6.5
25 wt.% NSal + 25 wt.% PSal	662.8 ± 30.4	40.5	33.7	207.6	-8.5	8.5
30 wt.% NSal + 25 wt.% PSal	803.6 ± 43.6	44.0	-31.9	247.9	-13.0	8.0
25 wt.% NSal + 30 wt.% PSal	728.1 ± 52.1	42.0	-107.4	172.5	-15.0	6.0
30 wt.% NSal + 30 wt.% PSal	985.4 ± 26.6	48.0	-56.6	329.3	-17.0	8.0

Table S5: Phase performance of the NSal+PSal mixtures relative to their precursor ILs based on a phase separation at $T_{sep} = 70$ °C for 30 min in a water bath heater. All osmolality measurements shown are based on five water activity measurements with a maximum standard deviation of 16% in osmolality.

IL Mixture	WR Osmolality (mmol kg ⁻¹)	PMR	WR Osmolality Difference from NSal (mmol kg ⁻¹)	WR Osmolality Difference from PSal (mmol kg ⁻¹)	PMR Difference from NSal	PMR Difference from PSal
20 wt.% NSal + 10 wt.% PSal	285.2 ± 14.5	1.57	43.53	49.11	0.14	-0.16
10 wt.% NSal + 20 wt.% PSal	251.7 ± 12.7	1.61	10.03	15.62	0.17	-0.12
25 wt.% NSal + 15 wt.% PSal	297.5 ± 33.2	0.68	-45.82	83.72	0.02	0.17
15 wt.% NSal + 25 wt.% PSal	191.5 ± 39.1	0.65	-151.84	-22.30	-0.01	0.15
25 wt.% NSal + 25 wt.% PSal	180.3 ± 49.5	0.25	-202.12	-143.98	0.04	0.03
30 wt.% NSal + 25 wt.% PSal	481.0 ± 51.6	0.13	-5.14	132.66	0.00	-0.04
25 wt.% NSal + 30 wt.% PSal	483.2 ± 37.5	0.15	-2.91	134.89	0.03	-0.01
30 wt.% NSal + 30 wt.% PSal	492.2 ± 30.5	0.09	-97.63	119.82	-0.03	-0.01

Supplementary Note 3: PTFA+PDMBS Mixtures

Herein we show the results for PTFA+PDMBS mixtures not shown in the main text. Fig. S2 shows the data plane of the mixtures, where the ordinates 0 and 1 on the y-axis represent the precursors ILs, PDMBS, and PTFA respectively. The osmolality as shown in Fig. S2(a) is higher than either precursor IL above 35 wt.% total IL composition and is peak at the 0.7 PTFA IL mixture ratio ordinate. The critical phase separation temperature, T_C , is lower than either precursor IL over the entire mixing plane as shown in Fig. S2(b). The WR phase purity is not improved over either precursor over the entire plane as shown in Fig. S2(c). However, as Fig. S2(d) shows, between 40-50 wt.% total IL composition, the PMR is slightly improved over either precursor IL at the 0.5 PTFA IL mixture ratio ordinate. Tables S6 and S7 show the raw data for six PTFA+PDMBS mixtures, with comparisons to each mixture's precursor ILs at the same total IL composition.

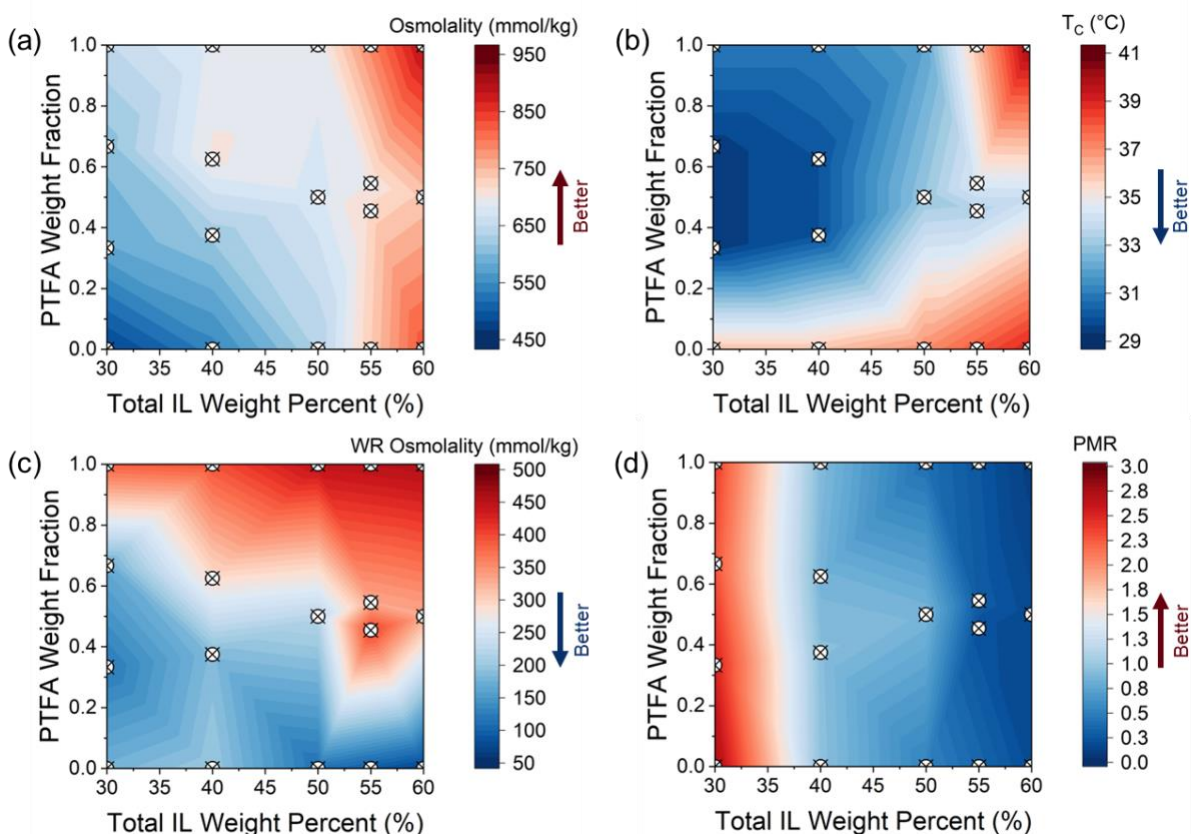


Fig. S2. Thermodynamic properties of the PTFA+PDMBS ternary mixtures in terms of the weight fraction of PTFA with respect to the total IL in the solution $\left(\frac{w_{PTFA}(\%)}{w_{PTFA}(\%) + w_{PDMBS}(\%)}\right)$ on the y-axis and the total IL weight percent in the solution on the x-axis (a) osmolality (b) critical phase separation temperature (c) WR phase osmolality (d) WR phase to ILR phase mass ratio (PMR). All the data in (c) and (d) are based on a phase separation at $T_{sep} = 70^{\circ}\text{C}$ for 30 minutes in a water bath. All osmolality measurements shown in (a) and (c) are based on five water activity measurements with a maximum standard deviation of 16% in osmolality. The maximum error in the cloud point measurements for T_C shown in (b) is 0.5°C . The maximum error in the PMR measurements shown in (d) is 0.1%. The marked points correspond to measured data points as per the coordinates listed in Table 1 of the main manuscript.

Table S6: The performance of the PTFA+PDMBS mixtures relative to their precursor ILs. All osmolality measurements shown are based on five water activity measurements with a maximum standard deviation of 16% in osmolality. The maximum error in the cloud point measurements for T_c is 0.5 °C.

IL Mixture	Osmolality (mmol kg ⁻¹)	T_c (°C)	Osmolality Difference from PTFA (mmol kg ⁻¹)	Osmolality Difference from PDMBS (mmol kg ⁻¹)	T_c Difference from PTFA (°C)	T_c Difference from PDMBS (°C)
20 wt.% PTFA + 10 wt.% PDMBS	607.8 ± 83.3	29.5	-40.2	130.8	-1.5	-6.5
10 wt.% PTFA + 20 wt.% PDMBS	580.9 ± 107.0	29.5	-67.1	103.9	-1.5	-6.5
25 wt.% PTFA + 15 wt.% PDMBS	703.3 ± 54.2	30.0	13.3	161.8	-1.0	-6.0
15 wt.% PTFA + 25 wt.% PDMBS	621.3 ± 93.6	30.0	-68.7	79.8	-1.0	-6.0
25 wt.% PTFA + 25 wt.% PDMBS	677.9 ± 26.8	33.0	-10.1	42.9	1.0	-4.0
30 wt.% PTFA + 25 wt.% PDMBS	701.9 ± 68.9	34.5	-88.6	-30.4	-1.5	-3.5
25 wt.% PTFA + 30 wt.% PDMBS	723.3 ± 99.4	33.5	-67.2	-8.9	-6.5	-5.5
30 wt.% PTFA + 30 wt.% PDMBS	738.3 ± 60.6	34.0	-154.7	-91.2	-6.0	-5.0

Table S7: Phase performance of the PTFA+PDMBS mixtures relative to their precursor ILs based on a phase separation at $T_{sep} = 70\text{ }^{\circ}\text{C}$ for 30 min in a water bath heater. All osmolality measurements shown are based on five water activity measurements with a maximum standard deviation of 16% in osmolality.

IL Mixture	WR Osmolality (mmol kg ⁻¹)	PMR	WR Osmolality Difference from PTFA (mmol kg ⁻¹)	WR Osmolality Difference from PDMBS (mmol kg ⁻¹)	PMR Difference from PTFA	PMR Difference from PDMBS
20 wt.% PTFA + 10 wt.% PDMBS	169.2 ± 24.4	2.29	-216.6	-8.9	-0.10	-0.48
10 wt.% PTFA + 20 wt.% PDMBS	121.2 ± 28.8	2.53	-264.5	-56.8	0.14	-0.24
25 wt.% PTFA + 15 wt.% PDMBS	290.8 ± 39.3	0.85	-102.9	90.4	-0.05	-0.09
15 wt.% PTFA + 25 wt.% PDMBS	185.9 ± 31.4	0.93	-207.7	-14.5	0.02	-0.02
25 wt.% PTFA + 25 wt.% PDMBS	221.6 ± 25.7	0.87	-223.6	107.0	0.46	0.34
30 wt.% PTFA + 25 wt.% PDMBS	332.1 ± 29.2	0.28	-119.2	238.7	0.03	-0.07
25 wt.% PTFA + 30 wt.% PDMBS	406.0 ± 35.0	0.22	-45.3	312.6	-0.03	-0.14
30 wt.% PTFA + 30 wt.% PDMBS	329.9 ± 42.1	0.18	-127.6	257.6	0.08	-0.01

Supplementary Note 4: IL Kinetics

The temporal effects on the purity of the WR phase based on a phase separation at $T_{sep} = 70$ °C in a water bath heater of the ILs PTFA, PDMBS, NSal, and PSal at a 40 wt.% IL composition are shown in Fig. S3(a) to (d). The use of a stir bar to stir the IL solution delivers a statistically marginal decrease in the osmolality of the WR phase (i.e., a higher water purity) beyond the 80-minute mark for all the ILs. Overall, stirring cannot enhance the WR phase purity significantly, with a maximum improvement of 50-100 mmol kg⁻¹ in the case of 40 wt.% PTFA over the unstirred solution after 10 minutes of heating as shown in Fig. S3(a). On the other hand, stirring has a detrimental effect on the purity of the WR phase in the case of solutions with more than one IL species as shown in Fig. S3(e) to (g). In either the 25 wt.% NSal + 15 wt.% PSal or 15 wt.% PTFA + 25 wt.% PDMBS solutions, the WR phase purity is higher (lower osmolality) in the case of the unstirred solutions. For 40 wt.% PTFA two heating modes were investigated: water bath heating (uniform heating around the vial) and hotplate heating (surface heating at the bottom of the vial) for 10, 20, 40, 80, and 160 minutes as shown in Fig. S3(a). Heating the IL solution with a hotplate (blue line) results in the least pure WR phase (highest osmolality) across all configurations.

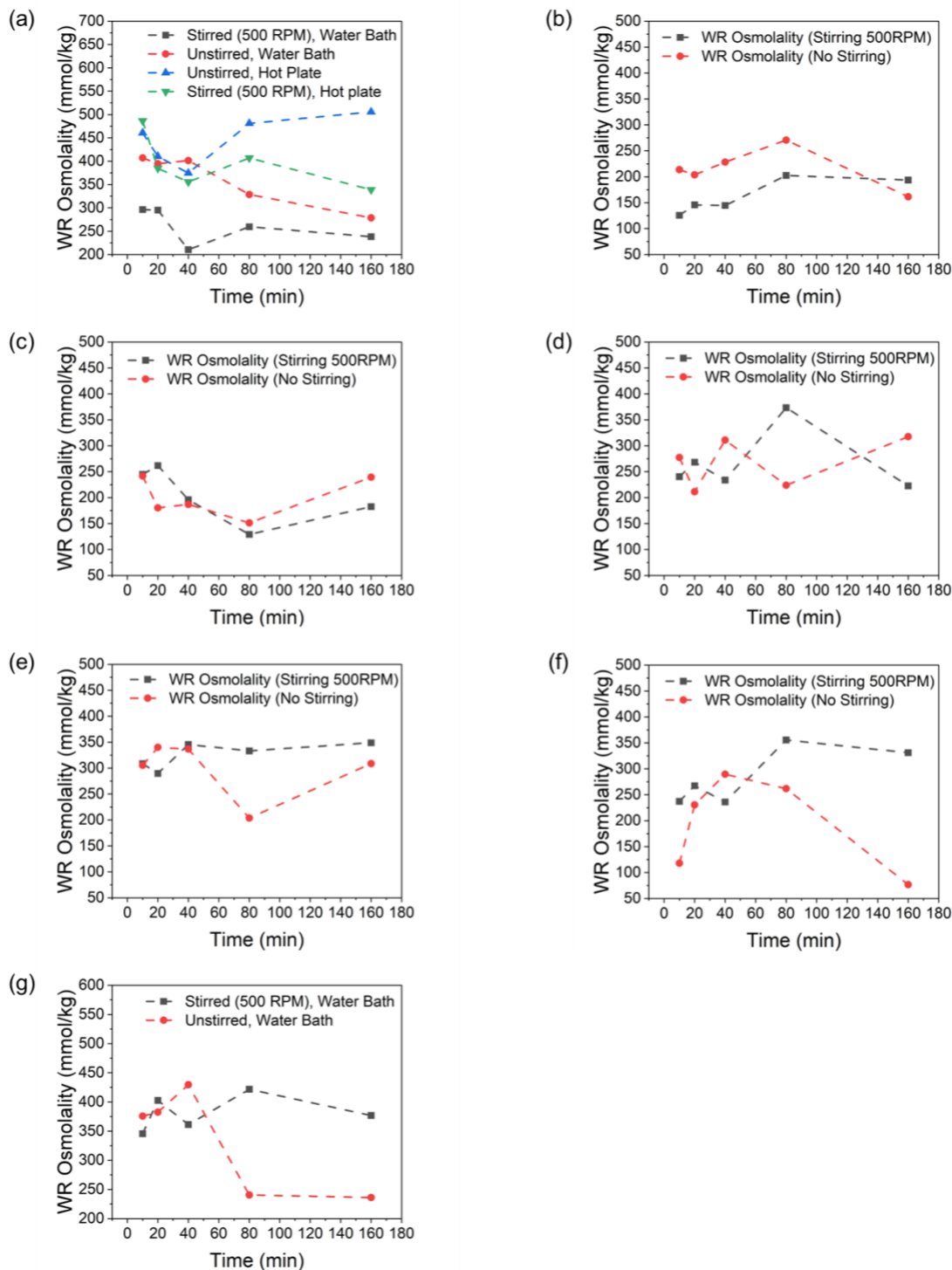


Fig. S3. Purity of the WR phase as a function of time and stirring in a water bath heater at $T_{sep} = 70\text{ }^{\circ}\text{C}$. (a) 40 wt.% PTFA. (b) 40 wt.% PDMBS. (c) 40 wt.% NSal. (d) 40 wt.% PSal. (e) 25 wt.% NSal + 15 wt.% PSal. (f) 15 wt.% PTFA + 25 wt.% PDMBS. (g) 15 wt.% NSal + 25 wt.% PTFA. Each data point represents an independent 10 mL sample. All osmolality measurements shown in (a) to (f) are based on five water activity measurements with a maximum standard deviation of 16% in osmolality.

Supplementary Note 5: IL Synthesis

The NMR spectra for both NSal and PSal are shown in Fig. S4(a) and (b) respectively. The Shimadzu IRAffinity-1 Fourier Transform Infrared (FTIR) spectrometer was utilized to perform attenuated total reflection (ATR) FTIR on NSal and PSal. The ATR-FTIR spectra for both ILs shown in Fig. S5 illustrate a strong presence of the -OH stretching mode near 3300 cm^{-1} , confirming the presence of the hydroxyl groups in the two ILs.

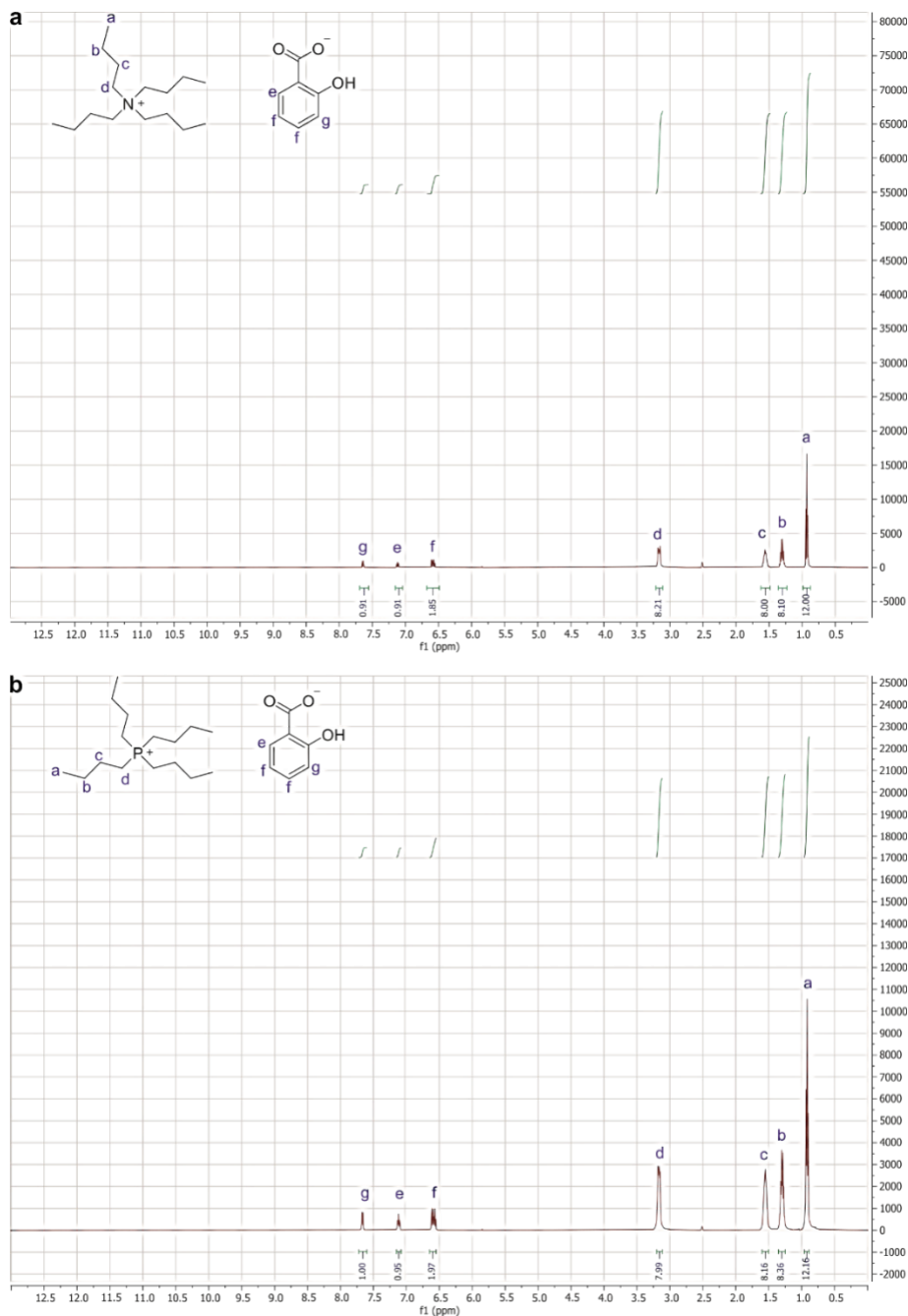


Fig. S4. (a) ^1H NMR of tetrabutylammonium salicylate (NSal) and (b) tetrabutylphosphonium salicylate (PSal)

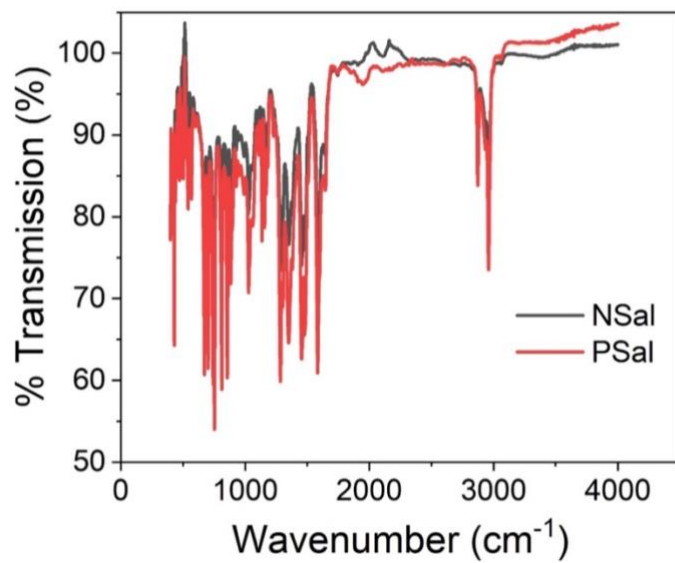


Fig. S5. ATR-FTIR results for NSal and PSal. The -OH stretching mode is present at 3300 cm⁻¹.

Supplementary Note 6: Ideal Entropy of Mixing and Chemical Potential

It should be noted that the increase in the ideal entropy of mixing upon going from two binary systems to a ternary system cannot account for the fact that the osmotic pressure of the ternary mixture is higher than either of the two binary mixtures at the same mole fraction of water. The ideal entropy of mixing in a binary mixture, which consists of water (denoted by subscript w) and some ionic liquid “a” (denoted by subscript a), can be written in the form given in Eq. S1, where n_i is the moles of species i , and x_i is the mole fraction of species i .

$$S_{mix} = -R[n_w \ln(x_w) + n_a \ln(x_a)] \quad (\text{S1})$$

Meanwhile, the ideal entropy of mixing in a ternary mixture can be written in the form given in Eq. S2.

$$S_{mix} = -R[n_w \ln(x_w) + n_a \ln(x_a) + n_b \ln(x_b)] \quad (\text{S2})$$

The chemical potential of water in a mixture can be expressed as the contribution from the ideal mixing entropy (μ_{ideal}), plus all other contributions, which includes non-ideal mixing entropy and the enthalpy of mixing (μ_{other}), as shown in Eq. S3.

$$\mu_w = \mu_{w,ideal} + \mu_{w,other} \quad (\text{S3})$$

In a binary mixture, the contribution of the ideal entropy of mixing to the chemical potential of water is given in Eq. S4.

$$\mu_{w,ideal} = -T \frac{\partial S_{mix}}{\partial n_w} = RT \left[\ln(x_w) + \frac{n_w}{x_w} \frac{\partial x_w}{\partial n_w} + \frac{n_a}{x_a} \frac{\partial x_a}{\partial n_w} \right] = RT \ln(x_w) \quad (\text{S4})$$

Meanwhile, Eq. 5 gives the ideal entropy of mixing’s contribution to the chemical potential of water in the ternary mixture, which simplifies to the same expression as in the binary mixture.

$$\mu_{w,ideal} = -T \frac{\partial S_{mix}}{\partial n_w} = RT \left[\ln(x_w) + \frac{n_w}{x_w} \frac{\partial x_w}{\partial n_w} + \frac{n_a}{x_a} \frac{\partial x_a}{\partial n_w} + \frac{n_b}{x_b} \frac{\partial x_b}{\partial n_w} \right] = RT \ln(x_w) \quad (\text{S5})$$

Thus, even though the entropy of mixing increases when two binary mixtures (IL “a” + water and IL “b” + water) are combined, the contribution of the ideal mixing entropy to the chemical potential of water remains unchanged (assuming the mole fraction of water is the same in a binary mixture “a”, binary mixture “b”, and the ternary mixture). Because the osmotic pressure is directly related to the chemical potential, the fact that the ternary entropy of mixing is greater than either of the constituent binary entropies of mixing cannot explain the increase in osmotic pressure of the ternary mixture over that of either of the binary constituents.

Supplementary Note 7: Ternary IL Mixtures and Weighted Average Models

The experimental ternary IL mixtures cannot be consistently modeled through a simple weighted averages model, either on a mass or a molar basis. For a given ternary mixture one approach is to model the properties of the ternary mixture based on a weighted of the partial properties of the two constituent IL species in the mixture. In a ternary mixture, the osmolality can be expressed in the form given in Eq. S6 and S7 on both a mass and a molar basis, where w_i is the weight fraction and x_i is the molar fraction of the IL species i .

$$Osm_{a+b} = \frac{w_a Osm_a + w_b Osm_b}{w_a + w_b} \quad (S6)$$

$$Osm_{a+b} = \frac{x_a Osm_a + x_b Osm_b}{x_a + x_b} \quad (S7)$$

Likewise, the critical phase separation temperature, T_c , can be expressed as given in Eq. S8 and S9, both on mass and a molar basis.

$$T_{c,a+b} = \frac{w_a T_{c,a} + w_b T_{c,b}}{w_a + w_b} \quad (S8)$$

$$T_{c,a+b} = \frac{x_a T_{c,a} + x_b T_{c,b}}{x_a + x_b} \quad (S9)$$

This simple model leads to significant errors in predicting a ternary mixture's osmolality or T_c , as shown in Table S8 and S9. Both methods severely underestimate the osmolality of the ternary mixtures contrary to the experimentally observed behavior illustrated in Figure 2 in the main text.

Table S8: Weighted average modeling approach for ternary IL mixtures on a mass basis.

Weighted Average (Mass Fraction Basis)				
IL Mixture	T_c (°C)	Osmolality (mmol/kg)	Difference from Experimental T_c (%)	Difference from Experimental Osmolality (%)
NSal+PTFA Mixtures				
20% NSal + 10% PTFA	50.3	427.1	25.8	-23.8
10% NSal + 20% PTFA	42.0	561.4	7.7	-7.1
25% NSal + 15% PTFA	46.0	454.2	10.9	-23.4
15% NSal + 25% PTFA	40.3	556.4	3.2	-33.1

25% NSal + 25% PTFA	39.6	519.7	-6.8	-32.9
30% NSal + 25% PTFA	39.9	502.9	-16.0	-42.6
25% NSal + 30% PTFA	38.4	535.9	-18.3	-39.0
30% NSal + 30% PTFA	38.8	519.1	-24.0	-51.2
PTFA+PDMBS Mixtures				
	T_c (°C)	Osmolality (mmol/kg)	Difference from Experimental T_c (%)	Difference from Experimental Osmolality (%)
20 wt.% PTFA + 10 wt.% PDMBS	36.3	522.7	23.2	-14.0
10 wt.% PTFA + 20 wt.% PDMBS	44.3	438.3	50.3	-24.5
25 wt.% PTFA + 15 wt.% PDMBS	35.4	537.8	17.9	-23.5
15 wt.% PTFA + 25 wt.% PDMBS	39.9	486.5	33.1	-21.7
25 wt.% PTFA + 25 wt.% PDMBS	34.8	545.6	5.3	-19.5
30 wt.% PTFA + 25 wt.% PDMBS	34.0	559.5	-1.6	-20.3
25 wt.% PTFA + 30 wt.% PDMBS	34.2	550.2	2.0	-23.9
30 wt.% PTFA + 30 wt.% PDMBS	33.5	562.5	-1.5	-23.8
NSal+PSal Mixtures				
	T_c (°C)	Osmolality (mmol/kg)	Difference from Experimental T_c (%)	Difference from Experimental Osmolality (%)
20% NSal + 10% PSal	41.0	386.2	3.8	-31.1
10% NSal + 20% PSal	38.0	382.1	1.3	-14.0
25% NSal + 15% PSal	39.7	380.7	-4.4	-34.1
15% NSal + 25% PSal	37.3	387.3	-0.6	-28.8
25% NSal + 25% PSal	37.3	384.4	-8.0	-42.0
30% NSal + 25% PSal	37.8	379.8	-14.2	-52.7
25% NSal + 30% PSal	36.5	387.4	-13.1	-46.8
30% NSal + 30% PSal	37.0	383.0	-22.9	-61.1

Table S9: Weighted average modelling approach for ternary IL mixtures on a molar basis.

Weighted Average (Molar Fraction Basis)				
IL Mixture	T_c (°C)	Osmolality (mmol/kg)	Difference from Experimental T_c (%)	Difference from Experimental Osmolality (%)
NSal+PTFA Mixtures				
20% NSal + 10% PTFA	29.6	251.4	-26.0	-123.0
10% NSal + 20% PTFA	31.6	429.7	-19.1	-40.7
25% NSal + 15% PTFA	27.9	271.8	-32.8	-118.2
15% NSal + 25% PTFA	29.4	414.2	-24.7	-100.8
25% NSal + 25% PTFA	26.4	344.5	-37.9	-124.8
30% NSal + 25% PTFA	25.7	318.1	-45.9	-175.3
25% NSal + 30% PTFA	26.5	372.6	-43.7	-135.6
30% NSal + 30% PTFA	25.8	344.0	-49.4	-209.1
PTFA+PDMBS Mixtures				
	T_c (°C)	Osmolality (mmol/kg)	Difference from Experimental T_c (%)	Difference from Experimental Osmolality (%)
20 wt.% PTFA + 10 wt.% PDMBS	20.6	313.7	-30.2	-93.8
10 wt.% PTFA + 20 wt.% PDMBS	33.0	323.7	11.7	-79.4
25 wt.% PTFA + 15 wt.% PDMBS	20.5	328.3	-31.7	-114.2
15 wt.% PTFA + 25 wt.% PDMBS	28.7	349.7	-4.4	-77.7
25 wt.% PTFA + 25 wt.% PDMBS	22.2	357.9	-32.6	-89.4
30 wt.% PTFA + 25 wt.% PDMBS	20.8	355.4	-39.6	-97.5
25 wt.% PTFA + 30 wt.% PDMBS	22.9	373.1	-31.8	-93.8
30 wt.% PTFA + 30 wt.% PDMBS	21.5	368.4	-36.9	-100.4
NSal+PSal Mixtures				
	T_c (°C)	Osmolality (mmol/kg)	Difference from Experimental T_c (%)	Difference from Experimental Osmolality (%)

20% NSal + 10% PSal	24.5	228.1	-37.9	-145.8
10% NSal + 20% PSal	28.5	287.3	-24.0	-54.5
25% NSal + 15% PSal	24.3	229.5	-41.5	-151.6
15% NSal + 25% PSal	27.1	282.7	-27.7	-92.3
25% NSal + 25% PSal	24.8	253.2	-38.8	-161.7
30% NSal + 25% PSal	24.3	240.8	-44.7	-233.7
25% NSal + 30% PSal	25.1	265.4	-40.3	-174.3
30% NSal + 30% PSal	24.6	252.1	-48.8	-290.9