Supplementary information:

Structural Investigations of Molecular Solutes within Nanostructured Ionic Liquids

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SAXS Measurements

Peak II Discussion

Peak II reflects the distance between similarly charged ions within the polar network. These results are depicted in Figs S1 and S2. From the data for the undiluted ILs, it is apparent that the correlation distance for peak II is not greatly affected by the cation alkyl chain for $[NTf_2]^-$ ILs, whereas a small increase of 0.13 Å is observed for the $[OTf]^-$ IL with the increase in alkyl chain from butyl to decyl with a significant decrease of 0.39 Å observed for the same change in the cation of the $[Me_2PO_4]^-$ ILs. The free volume within ILs bearing these anions has been found to increase in the order $[C_4C_1im][Me_2PO_4]$ << $[C_4C_1im][OTf]$ < $[C_4C_1im][NTf_2]$.[1] ILs containing more basic anions such as $[Me_2PO_4]^-$ also have a higher propensity to form more compact π-stacked structures than anions such as $[NTf_2]^-$.[2, 3] Hence as the alkyl chain increases, it is more likely that it will be excluded from the polar regions of the more compact $[C_nC_1im][Me_2PO_4]$ ILs, whereas there will be more flexibility for its partial incorporation into the polar regions of the other two ILs. Given the weakly interacting nature of the $[C_nC_1im][NTf_2]$ ILs this does not lead to a perturbation of the distance between ions whereas the more compact $[C_nC_1im][OTf]$ ILs experience a small expansion.

The effect of solutes on the position of peak II is less clear and some apparent discrepancies occur. For example, both ACN and DMSO were classified at highly dipolar solutes that partition preferentially into the polar regions of the ILs. Despite their similar behaviour based on the peak I results, the correlation distance corresponding to peak II is observed to not change significantly for DMSO whereas it increases notably for the $[NTf_2]^-$ ILs and $[C_{10}C_1im][Me_2PO_4]$ for ACN. These differences in behaviour are likely to arise from a complex interplay of the preferred conformations of the solute relative to the IL ions as well as partitioning within the domains leading to more

difficult to rationalise trends. Nonetheless, for CyOAc which was identified as preferentially partitioning in the non-polar domains for all ILs, no significant change in the correlation distance for peak II could be observed outside of a subtle increase of $[C_4C_1im][NTf_2]$ which is consistent with its limited solvation within this region of these ILs.

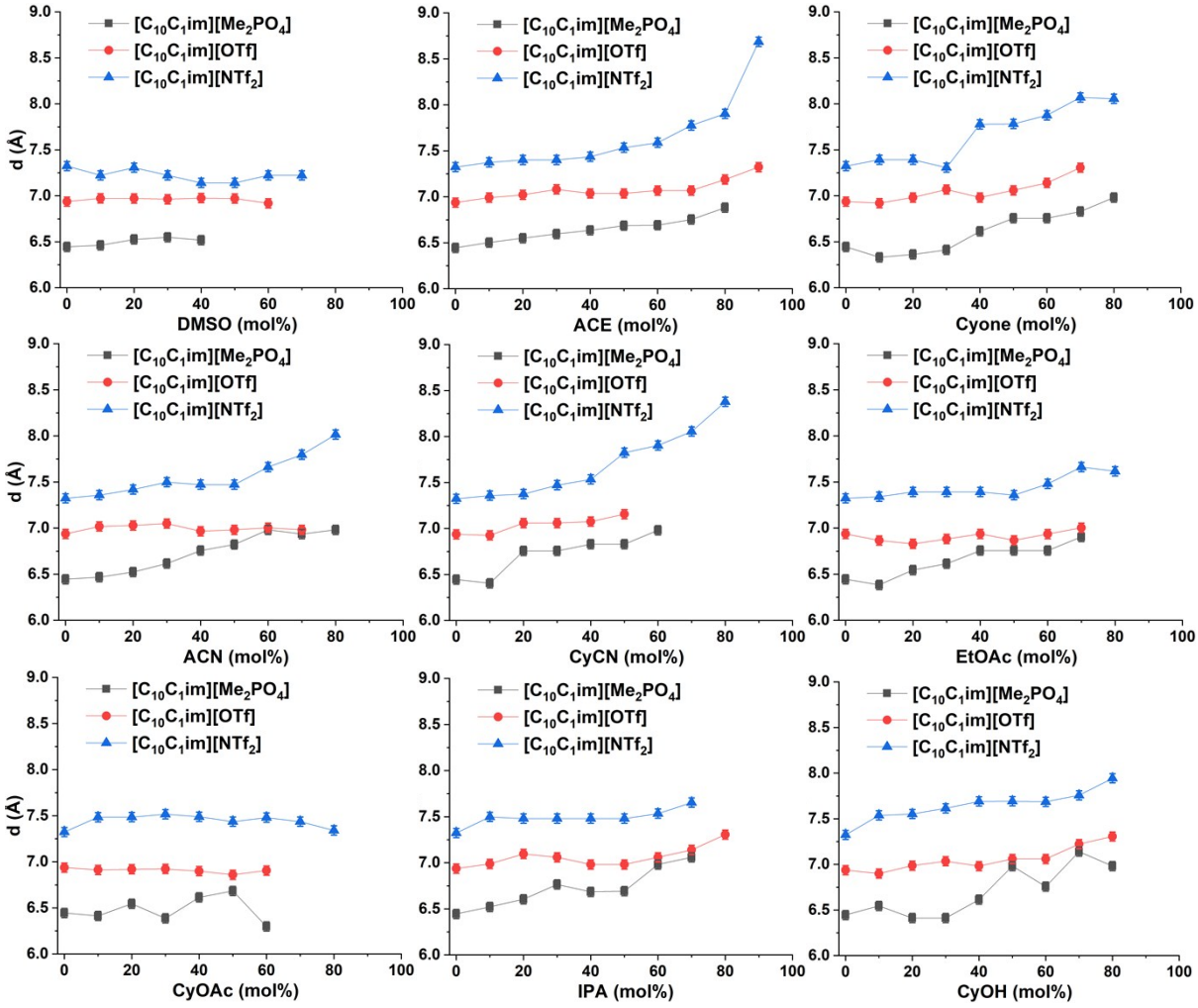

Figure S1 Correlation distance (d) of the peak maximum for peak II across all $[C_{10}C_1im]^+$ ILs for which peak II could be identified.

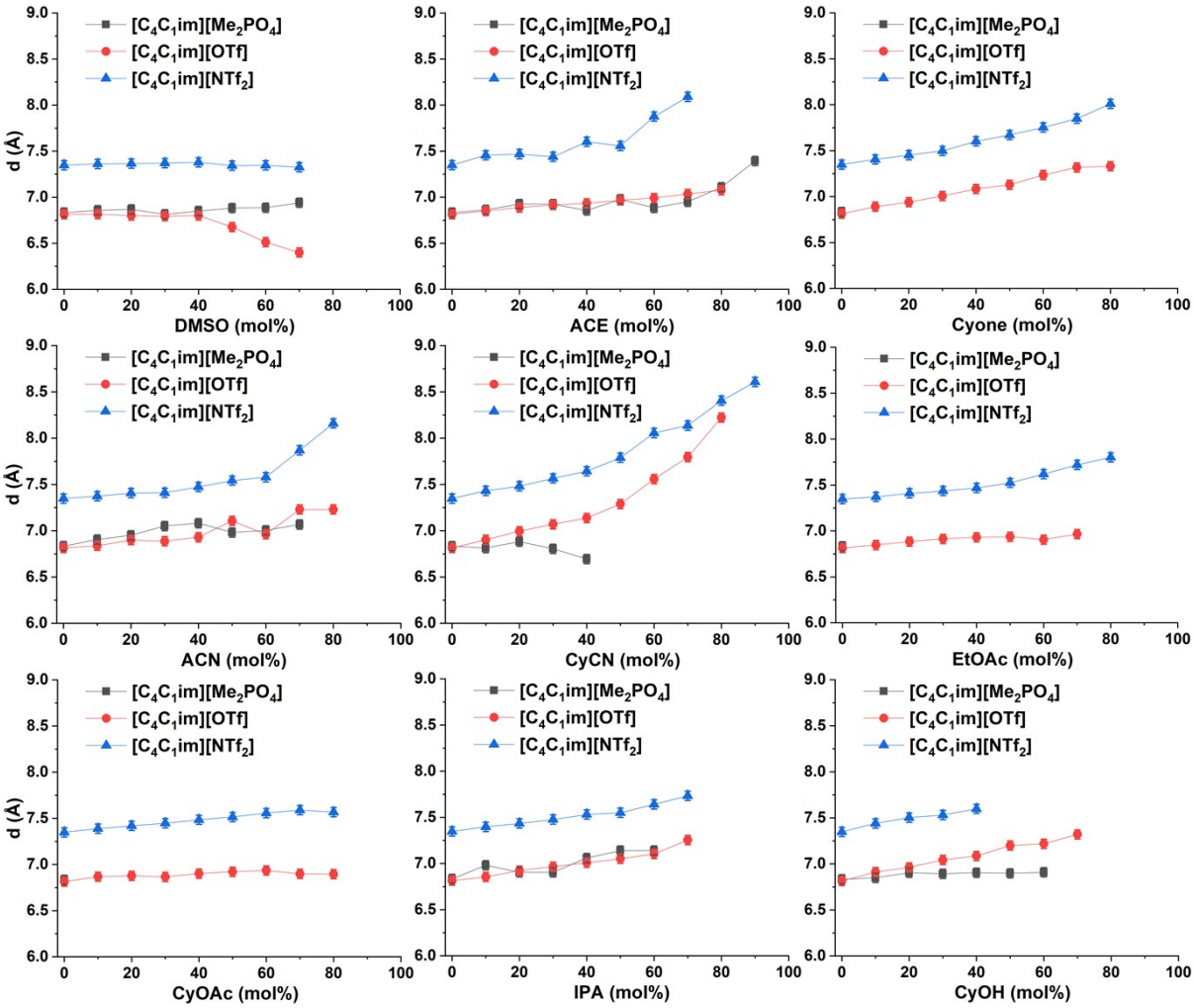

Figure S2 Correlation distance (d) of the peak maximum for peak II across all $[C_4C_1im]^+$ ILs for which peak II could be identified.

SAXS Fitting Data

Table S1. Fitted peak positions for all ILs, molecular solutes and mixtures from the SAXS patterns obtained. NM = not miscible. Blank spaces depict solutions where either peak 1 or peak 2 could not be determined.

Solute	Ionic liquid	Mol% solute	q peak 1 ($Å^{-1}$)	q peak 2 ($Å^{-1}$)	d peak 1 (Å)	d peak 2 (Å)
N/A	$[C_{10}C_1im][Me_2PO_4]$	0	0.272	0.975	23.07	6.44
N/A	$[C_{10}C_1im][NTf_2]$	0	0.283	0.858	22.20	7.32
N/A	$[C_{10}C_1im][OTf]$	0	0.256	0.906	24.59	6.94
N/A	$[C_4C_1im][Me_2PO_4]$	0	0.477	0.920	13.18	6.83
N/A	$[C_4C_1im][NTf_2]$	0	0.546	0.855	11.51	7.35
N/A	$[C_4C_1im][OTf]$	0	0.512	0.922	12.27	6.81
ACE	$[C_{10}C_1im][Me_2PO_4]$	10	0.277	0.966	22.66	6.50
		20	0.284	0.959	22.11	6.55
		30	0.287	0.953	21.92	6.60
		40	0.290	0.947	21.66	6.64
		50	0.292	0.940	21.50	6.69
		60	0.292	0.939	21.51	6.69
		70	0.287	0.930	21.92	6.75
		80	0.273	0.913	22.98	6.88
		90				
ACE	$[C_{10}C_1im][NTf_2]$	10	0.281	0.852	22.36	7.37
		20	0.290	0.849	21.67	7.40
		30	0.292	0.849	21.52	7.40
		40	0.296	0.845	21.23	7.44
		50	0.303	0.834	20.74	7.53
		60	0.310	0.828	20.27	7.59
		70	0.323	0.808	19.45	7.78
		80	0.323	0.795	19.44	7.90
		90	0.337	0.723	18.64	8.69
ACE	$[C_{10}C_1im][OTf]$	10	0.259	0.899	24.24	6.99
		20	0.265	0.895	23.67	7.02
		30	0.267	0.887	23.49	7.08
		40	0.270	0.893	23.27	7.04
		50	0.274	0.893	22.93	7.04

		60	0.277	0.889	22.68	7.07
		70	0.274	0.889	22.93	7.07
		80	0.296	0.874	21.23	7.19
		90	0.294	0.858	21.37	7.32
ACE	$[C_4C_1im][Me_2PO_4]$	10	0.472	0.916	13.32	6.86
		20	0.462	0.907	13.60	6.93
		30	0.456	0.907	13.77	6.92
		40	0.461	0.916	13.64	6.86
		50	0.410	0.901	15.31	6.98
		60	0.395	0.913	15.91	6.88
		70	0.334	0.904	18.81	6.95
		80		0.884		7.11
		90	0.544	0.850	11.55	7.39
ACE	$[C_4C_1im][NTf_2]$	10	0.553	0.843	11.36	7.46
		20	0.502	0.841	12.53	7.47
		30	0.517	0.845	12.16	7.44
		40	0.479	0.827	13.11	7.60
		50		0.831		7.56
		60		0.798		7.88
		70		0.777		8.09
		80		0.524		11.99
		90				
ACE	$[C_4C_1im][OTf]$	10	0.492	0.917	12.78	6.85
		20	0.478	0.912	13.15	6.89
		30	0.465	0.909	13.52	6.92
		40	0.464	0.906	13.54	6.93
		50	0.441	0.902	14.26	6.97
		60	0.399	0.899	15.75	6.99
		70	0.374	0.893	16.80	7.03
		80	0.325	0.888	19.33	7.08
		90				
ACN	$[C_{10}C_1im][Me_2PO_4]$	10	0.278	0.971	22.59	6.47
		20	0.282	0.963	22.31	6.52
		30	0.284	0.950	22.12	6.62
		40	0.288	0.930	21.82	6.76

		50	0.297	0.921	21.19	6.82
		60	0.302	0.900	20.83	6.98
		70	0.316	0.906	19.88	6.94
		80	0.306	0.900	20.53	6.98
		90	0.314		20.01	
ACN	$[C_{10}C_1im][NTf_2]$	10	0.294	0.854	21.37	7.36
		20	0.294	0.847	21.37	7.42
		30	0.292	0.838	21.52	7.50
		40	0.297	0.841	21.16	7.47
		50	0.301	0.841	20.87	7.47
		60	0.310	0.820	20.27	7.66
		70	0.328	0.806	19.16	7.80
		80	0.341	0.784	18.43	8.01
		90	0.367		17.12	
ACN	$[C_{10}C_1im][OTf]$	10	0.263	0.895	23.92	7.02
		20	0.262	0.894	23.98	7.03
		30	0.264	0.891	23.82	7.05
		40	0.269	0.902	23.38	6.97
		50	0.272	0.900	23.10	6.98
		60	0.281	0.897	22.36	7.00
		70	0.290	0.900	21.67	6.98
		80	0.309		20.33	
		90	0.325		19.33	
ACN	$[C_4C_1im][Me_2PO_4]$	10	0.477	0.910	13.17	6.90
		20	0.468	0.904	13.42	6.95
		30	0.475	0.891	13.24	7.05
		40	0.472	0.887	13.31	7.08
		50	0.468	0.900	13.44	6.98
		60	0.461	0.897	13.62	7.00
		70	0.445	0.889	14.11	7.07
		80	0.400		15.71	
		90				
ACN	$[C_4C_1im][NTf_2]$	10	0.554	0.852	11.34	7.37
		20	0.572	0.848	10.99	7.41
		30	0.587	0.848	10.71	7.41

		40	0.617	0.841	10.18	7.47
		50	0.549	0.833	11.45	7.54
		60	0.626	0.829	10.04	7.58
		70	0.287	0.799	21.87	7.87
		80		0.770		8.16
		90		0.643		9.77
ACN	$[C_4C_1im][OTf]$	10	0.476	0.919	13.20	6.84
		20	0.436	0.911	14.41	6.90
		30	0.496	0.912	12.67	6.89
		40	0.494	0.907	12.73	6.93
		50	0.456	0.884	13.78	7.11
		60	0.495	0.902	12.70	6.96
		70		0.869		7.23
		80	0.440	0.869	14.28	7.23
		90				
CyCN	$[C_{10}C_1im][Me_2PO_4]$	10	0.281	0.981	22.38	6.40
		20	0.286	0.930	21.96	6.76
		30	0.283	0.930	22.23	6.76
		40	0.292	0.920	21.50	6.83
		50	0.289	0.920	21.75	6.83
		60	0.290	0.900	21.68	6.98
		70	0.272		23.10	
		80	0.248		25.34	
		90	0.214		29.36	
CyCN	$[C_{10}C_1im][NTf_2]$	10	0.300	0.854	20.94	7.36
		20	0.299	0.852	21.01	7.37
		30	0.306	0.841	20.50	7.47
		40	0.311	0.834	20.21	7.53
		50	0.315	0.803	19.92	7.82
		60	0.330	0.795	19.04	7.90
		70	0.328	0.780	19.17	8.06
		80	0.330	0.750	19.04	8.38
		90	0.340		18.48	
CyCN	$[C_{10}C_1im][OTf]$	10	0.261	0.907	24.06	6.93
		20	0.267	0.890	23.54	7.06

		30	0.271	0.890	23.19	7.06
		40	0.276	0.888	22.74	7.08
		50	0.281	0.878	22.37	7.16
		60	0.283		22.24	
		70	0.283		22.21	
		80	0.274		22.91	
		90	0.274		22.93	
CyCN	$[C_4C_1im][Me_2PO_4]$	10	0.467	0.920	13.45	6.83
		20	0.428	0.922	14.69	6.81
		30	0.395	0.913	15.91	6.88
		40	0.363	0.923	17.31	6.80
		50	0.320	0.939	19.63	6.69
		60	NM	NM		
		70	NM	NM		
		80	NM	NM		
		90	NM	NM		
CyCN	$[C_4C_1im][NTf_2]$	10	0.508	0.846	12.38	7.43
		20	0.504	0.840	12.48	7.48
		30	0.487	0.830	12.91	7.57
		40	0.477	0.822	13.16	7.64
		50	0.370	0.807	16.98	7.79
		60	0.330	0.780	19.04	8.06
		70	0.340	0.772	18.48	8.14
		80	0.330	0.748	19.04	8.41
		90	0.320	0.730	19.63	8.61
CyCN	$[C_4C_1im][OTf]$	10	0.473	0.910	13.30	6.90
		20	0.443	0.898	14.18	6.99
		30	0.404	0.889	15.57	7.07
		40	0.375	0.880	16.75	7.14
		50	0.350	0.862	17.95	7.29
		60	0.320	0.831	19.63	7.56
		70	0.270	0.806	23.27	7.80
		80		0.764		8.22
		90				
CyOAc	$[C_{10}C_1im][Me_2PO_4]$	10	0.277	0.980	22.66	6.41

		20	0.278	0.960	22.62	6.54
		30	0.276	0.984	22.76	6.39
		40	0.269	0.950	23.39	6.61
		50	0.266	0.940	23.64	6.68
		60	0.256	0.998	24.54	6.30
		70	0.227		27.67	
		80	0.221		28.43	
		90	0.271		23.19	
CyOAc	$[C_{10}C_1im][NTf_2]$	10	0.294	0.840	21.37	7.48
		20	0.297	0.839	21.16	7.49
		30	0.299	0.836	21.04	7.52
		40	0.300	0.839	20.94	7.49
		50	0.300	0.845	20.93	7.44
		60	0.300	0.840	20.95	7.48
		70	0.297	0.845	21.13	7.44
		80	0.292	0.856	21.52	7.34
		90	0.274		22.93	
CyOAc	$[C_{10}C_1im][OTf]$	10	0.258	0.909	24.32	6.91
		20	0.260	0.908	24.14	6.92
		30	0.261	0.908	24.10	6.92
		40	0.260	0.911	24.13	6.90
		50	0.259	0.916	24.26	6.86
		60	0.255	0.910	24.65	6.90
		70	0.244		25.72	
		80	0.230		27.32	
		90	0.190		33.07	
CyOAc	$[C_4C_1im][Me_2PO_4]$	10	NM	NM		
		20	NM	NM		
		30	NM	NM		
		40	NM	NM		
		50	NM	NM		
		60	NM	NM		
		70	NM	NM		
		80	NM	NM		
		90	NM	NM		

CyOAc	$[C_4C_1im][NTf_2]$	10	0.477	0.850	13.17	7.39
		20	0.431	0.847	14.57	7.42
		30	0.398	0.844	15.78	7.45
		40	0.340	0.840	18.46	7.48
		50	0.340	0.836	18.48	7.51
		60	0.305	0.831	20.60	7.56
		70	0.267	0.828	23.53	7.59
		80		0.830		7.57
		90		0.900		6.98
CyOAc	$[C_4C_1im][OTf]$	10	0.430	0.915	14.61	6.87
		20	0.398	0.914	15.78	6.88
		30	0.348	0.915	18.06	6.87
		40	0.320	0.910	19.63	6.90
		50	0.280	0.908	22.44	6.92
		60		0.906		6.94
		70		0.911		6.90
		80		0.911		6.90
		90				
CyOH	$[C_{10}C_1im][Me_2PO_4]$	10	0.283	0.960	22.22	6.54
		20	0.284	0.980	22.11	6.41
		30	0.284	0.980	22.11	6.41
		40	0.298	0.950	21.06	6.61
		50	0.330	0.900	19.04	6.98
		60	0.314	0.930	19.99	6.76
		70	0.346	0.880	18.17	7.14
		80	0.342	0.900	18.39	6.98
		90	0.467		13.44	
CyOH	$[C_{10}C_1im][NTf_2]$	10	0.295	0.834	21.33	7.54
		20	0.294	0.832	21.37	7.55
		30	0.298	0.825	21.06	7.61
		40	0.299	0.817	20.99	7.69
		50	0.300	0.817	20.94	7.69
		60	0.297	0.817	21.18	7.69
		70	0.290	0.810	21.67	7.76
		80	0.261	0.791	24.07	7.94

		90				
CyOH	$[C_{10}C_1im][OTf]$	10	0.262	0.911	24.00	6.90
		20	0.266	0.899	23.59	6.99
		30	0.270	0.893	23.23	7.04
		40	0.282	0.900	22.32	6.98
		50	0.292	0.890	21.53	7.06
		60	0.302	0.890	20.82	7.06
		70	0.314	0.870	20.03	7.22
		80	0.330	0.860	19.04	7.31
		90	0.340		18.48	
CyOH	$[C_4C_1im][Me_2PO_4]$	10	0.464	0.917	13.53	6.85
		20	0.470	0.910	13.37	6.90
		30	0.454	0.912	13.84	6.89
		40	0.440	0.910	14.29	6.90
		50	0.447	0.911	14.04	6.90
		60	0.451	0.909	13.92	6.91
		70	0.476	0.782	13.21	8.04
		80	0.452		13.90	
		90	0.481		13.05	
CyOH	$[C_4C_1im][NTf_2]$	10	0.509	0.845	12.35	7.44
		20	0.521	0.838	12.07	7.50
		30	0.456	0.834	13.79	7.53
		40		0.827		7.60
		50	NM	NM		
		60	NM	NM		
		70	NM	NM		
		80	NM	NM		
		90	NM	NM		
CyOH	$[C_4C_1im][OTf]$	10	0.474	0.909	13.25	6.91
		20	0.454	0.902	13.85	6.96
		30	0.426	0.892	14.74	7.04
		40	0.414	0.887	15.16	7.09
		50	0.410	0.873	15.32	7.20
		60	0.400	0.870	15.71	7.22
		70	0.400	0.858	15.71	7.32

		80		0.520		12.08
		90		0.580		10.83
Cyone	$[C_{10}C_1im][Me_2PO_4]$	10	0.275	0.992	22.87	6.33
		20	0.276	0.988	22.76	6.36
		30	0.277	0.980	22.71	6.41
		40	0.278	0.950	22.60	6.61
		50	0.285	0.930	22.07	6.76
		60	0.285	0.930	22.06	6.76
		70	0.285	0.920	22.04	6.83
		80	0.276	0.900	22.76	6.98
		90	0.221		28.41	
Cyone	$[C_{10}C_1im][NTf_2]$	10	0.301	0.850	20.88	7.39
		20	0.305	0.850	20.61	7.39
		30	0.310	0.860	20.27	7.31
		40	0.320	0.808	19.63	7.78
		50	0.322	0.807	19.50	7.78
		60	0.330	0.798	19.04	7.88
		70	0.345	0.779	18.21	8.07
		80	0.345	0.780	18.21	8.06
		90	0.350	0.831	17.95	7.56
Cyone	$[C_{10}C_1im][OTf]$	10	0.269	0.908	23.40	6.92
		20	0.268	0.900	23.41	6.98
		30	0.274	0.889	22.94	7.07
		40	0.279	0.900	22.50	6.98
		50	0.287	0.890	21.92	7.06
		60	0.293	0.880	21.44	7.14
		70	0.300	0.860	20.94	7.31
		80	0.304		20.65	
		90	0.310		20.27	
Cyone	$[C_4C_1im][Me_2PO_4]$	10	NM	NM		
		20	NM	NM		
		30	NM	NM		
		40	NM	NM		
		50	NM	NM		
		60	NM	NM		

		70	NM	NM		
		80	NM	NM		
		90	NM	NM		
Cyone	$[C_4C_1im][NTf_2]$	10	0.517	0.849	12.15	7.40
		20	0.508	0.843	12.37	7.45
		30	0.511	0.838	12.29	7.50
		40	0.485	0.826	12.96	7.60
		50	0.428	0.819	14.69	7.67
		60	0.400	0.810	15.71	7.75
		70	0.400	0.801	15.71	7.85
		80	0.370	0.784	16.98	8.01
		90				
Cyone	$[C_4C_1im][OTf]$	10	0.487	0.912	12.91	6.89
		20	0.462	0.905	13.61	6.94
		30	0.435	0.897	14.43	7.01
		40	0.413	0.887	15.21	7.08
		50	0.392	0.881	16.02	7.13
		60	0.380	0.868	16.53	7.24
		70	0.350	0.859	17.95	7.32
		80	0.320	0.857	19.63	7.33
		90	0.280		22.44	
DMSO	$[C_{10}C_1im][Me_2PO_4]$	10	0.281	0.972	22.36	6.46
		20	0.290	0.963	21.67	6.53
		30	0.297	0.959	21.18	6.55
		40	0.290	0.964	21.67	6.52
		50	0.310		20.27	
		60	0.330		19.04	
		70	0.340		18.48	
		80	0.320		19.63	
		90	0.360		17.45	
DMSO	$[C_{10}C_1im][NTf_2]$	10	0.292	0.870	21.52	7.22
		20	0.300	0.860	20.94	7.31
		30	0.300	0.870	20.94	7.22
		40	0.310	0.880	20.27	7.14
		50	0.300	0.880	20.94	7.14

		60	0.310	0.870	20.27	7.22
		70	0.330	0.870	19.04	7.22
		80	0.340		18.48	
		90	0.370		16.98	
DMSO	$[C_{10}C_1im][OTf]$	10	0.258	0.901	24.37	6.97
		20	0.258	0.901	24.36	6.97
		30	0.265	0.902	23.70	6.96
		40	0.265	0.901	23.68	6.98
		50	0.270	0.901	23.27	6.97
		60	0.279	0.908	22.54	6.92
		70	0.280		22.44	
		80	0.310		20.27	
		90	0.350		17.95	
DMSO	$[C_4C_1im][Me_2PO_4]$	10	0.480	0.916	13.08	6.86
		20	0.482	0.915	13.03	6.87
		30	0.479	0.922	13.11	6.81
		40	0.483	0.917	13.00	6.85
		50	0.484	0.913	12.99	6.88
		60	0.484	0.912	12.99	6.89
		70	0.497	0.905	12.64	6.94
		80	0.490		12.82	
		90	0.519		12.10	
DMSO	$[C_4C_1im][NTf_2]$	10	0.531	0.853	11.82	7.36
		20	0.536	0.853	11.73	7.36
		30	0.540	0.852	11.63	7.37
		40	0.546	0.852	11.52	7.38
		50	0.556	0.856	11.30	7.34
		60		0.855		7.35
		70		0.858		7.33
		80		0.895		7.02
		90				
DMSO	$[C_4C_1im][OTf]$	10	0.504	0.922	12.48	6.82
		20	0.514	0.924	12.23	6.80
		30	0.507	0.925	12.39	6.79
		40	0.507	0.924	12.40	6.80

		50	0.512	0.941	12.28	6.68
		60	0.553	0.965	11.36	6.51
		70	0.562	0.982	11.18	6.40
		80				
		90				
EtOAc	$[C_{10}C_1im][Me_2PO_4]$	10	0.275	0.984	22.81	6.38
		20	0.274	0.960	22.95	6.54
		30	0.278	0.950	22.57	6.61
		40	0.279	0.930	22.54	6.76
		50	0.275	0.930	22.82	6.76
		60	0.274	0.930	22.97	6.76
		70	0.260	0.910	24.15	6.90
		80	0.234		26.85	
		90	NM	NM		
EtOAc	$[C_{10}C_1im][NTf_2]$	10	0.300	0.856	20.94	7.34
		20	0.300	0.850	20.94	7.39
		30	0.300	0.850	20.94	7.39
		40	0.310	0.850	20.27	7.39
		50	0.316	0.854	19.88	7.36
		60	0.321	0.840	19.57	7.48
		70	0.334	0.820	18.81	7.66
		80	0.334	0.825	18.81	7.62
		90	0.314		20.01	
EtOAc	$[C_{10}C_1im][OTf]$	10	0.260	0.915	24.15	6.87
		20	0.262	0.920	24.02	6.83
		30	0.266	0.913	23.64	6.88
		40	0.269	0.906	23.36	6.94
		50	0.273	0.915	23.01	6.87
		60	0.271	0.906	23.21	6.94
		70	0.276	0.897	22.79	7.00
		80	0.268		23.48	
		90				
EtOAc	$[C_4C_1im][Me_2PO_4]$	10	NM	NM		
		20	NM	NM		
		30	NM	NM		

		40	NM	NM		
		50	NM	NM		
		60	NM	NM		
		70	NM	NM		
		80	NM	NM		
		90	NM	NM		
EtOAc	$[C_4C_1im][NTf_2]$	10	0.520	0.852	12.09	7.37
		20	0.499	0.848	12.59	7.41
		30	0.495	0.845	12.69	7.44
		40	0.481	0.841	13.07	7.47
		50	0.462	0.835	13.60	7.52
		60	0.431	0.825	14.58	7.62
		70	0.408	0.814	15.39	7.72
		80	0.365	0.805	17.21	7.80
		90				
EtOAc	$[C_4C_1im][OTf]$	10	0.484	0.918	12.99	6.85
		20	0.464	0.913	13.53	6.88
		30	0.458	0.909	13.73	6.91
		40	0.404	0.907	15.54	6.93
		50	0.433	0.906	14.51	6.94
		60	0.324	0.910	19.38	6.90
		70	0.298	0.902	21.10	6.97
		80				
		90				
IPA	$[C_{10}C_1im][Me_2PO_4]$	10	0.279	0.963	22.53	6.52
		20	0.285	0.951	22.08	6.60
		30	0.294	0.929	21.35	6.77
		40	0.282	0.940	22.31	6.68
		50	0.291	0.939	21.57	6.69
		60	0.308	0.900	20.41	6.98
		70	0.363	0.890	17.31	7.06
		80	0.410	0.850	15.32	7.39
		90	0.460	0.800	13.66	7.85
IPA	$[C_{10}C_1im][NTf_2]$	10	0.292	0.838	21.52	7.50
		20	0.299	0.840	21.01	7.48

		30	0.303	0.840	20.74	7.48
		40	0.303	0.840	20.74	7.48
		50	0.310	0.840	20.27	7.48
		60	0.305	0.834	20.60	7.53
		70	0.297	0.821	21.16	7.65
		80	0.277	0.806	22.68	7.79
		90		0.788		7.97
IPA	$[C_{10}C_1im][OTf]$	10	0.262	0.899	23.99	6.99
		20	0.268	0.885	23.45	7.10
		30	0.274	0.890	22.92	7.06
		40	0.281	0.900	22.36	6.98
		50	0.285	0.900	22.05	6.98
		60	0.292	0.890	21.49	7.06
		70	0.301	0.880	20.89	7.14
		80	0.302	0.860	20.79	7.31
		90	0.292	0.820	21.52	7.66
IPA	$[C_4C_1im][Me_2PO_4]$	10	0.474	0.900	13.26	6.98
		20	0.471	0.910	13.34	6.90
		30	0.476	0.910	13.20	6.90
		40	0.483	0.890	13.01	7.06
		50	0.490	0.880	12.82	7.14
		60	0.481	0.880	13.06	7.14
		70		0.804		7.82
		80	0.510	0.810	12.31	7.76
		90	0.568	0.816	11.06	7.70
IPA	$[C_4C_1im][NTf_2]$	10	0.554	0.849	11.34	7.40
		20	0.503	0.845	12.49	7.43
		30	0.529	0.840	11.88	7.48
		40	0.424	0.834	14.83	7.53
		50		0.832		7.55
		60		0.822		7.64
		70		0.813		7.73
		80		0.802		7.83
		90		0.775		8.10
IPA	$[C_4C_1im][OTf]$	10	0.500	0.917	12.57	6.85

20	0.480	0.908	13.10	6.92
30	0.467	0.902	13.45	6.97
40	0.457	0.897	13.75	7.01
50	0.436	0.891	14.41	7.05
60	0.432	0.885	14.54	7.10
70	0.410	0.866	15.32	7.25
80		0.850		7.39
90		0.790		7.95

SAXS Scattering Patterns

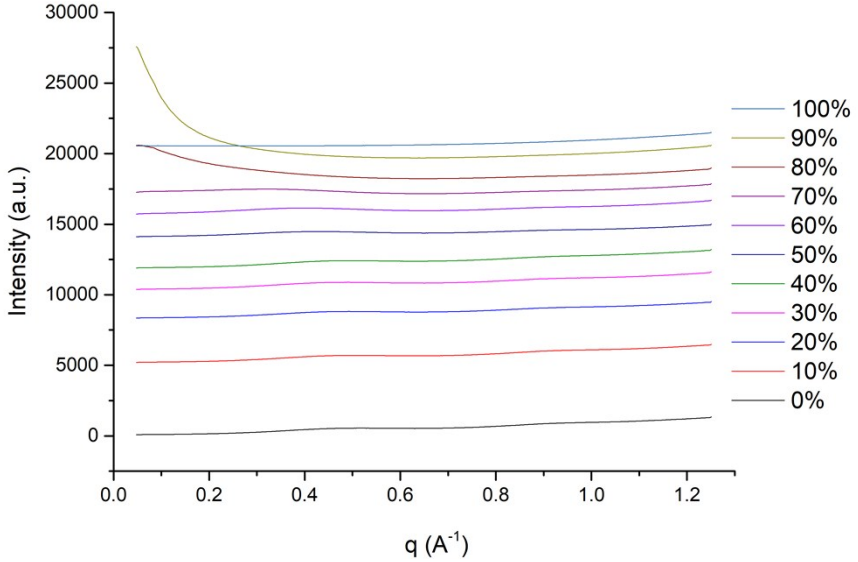

Figure S3. SAXS scattering patterns of ACE-[C_4C_1im][Me_2PO_4] mixtures.

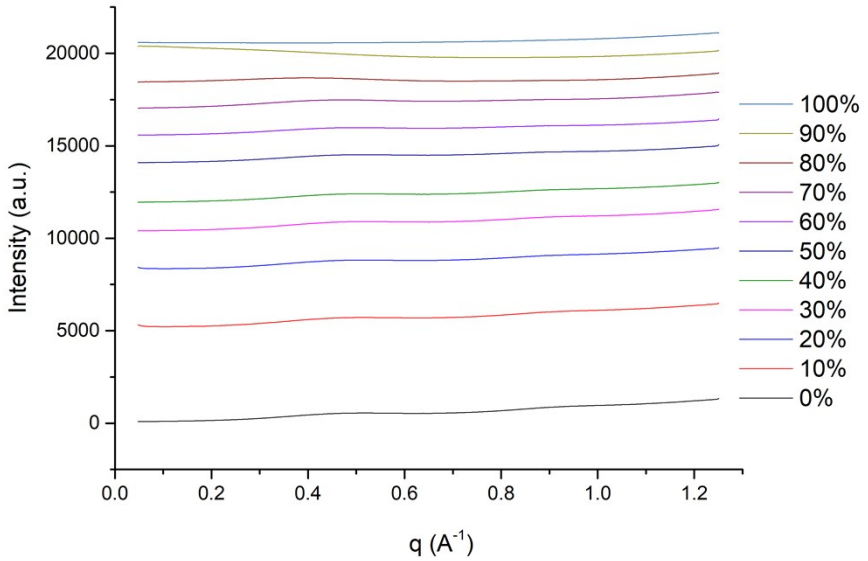

Figure S4. SAXS scattering patterns of ACN-[C_4C_1im][Me_2PO_4] mixtures.

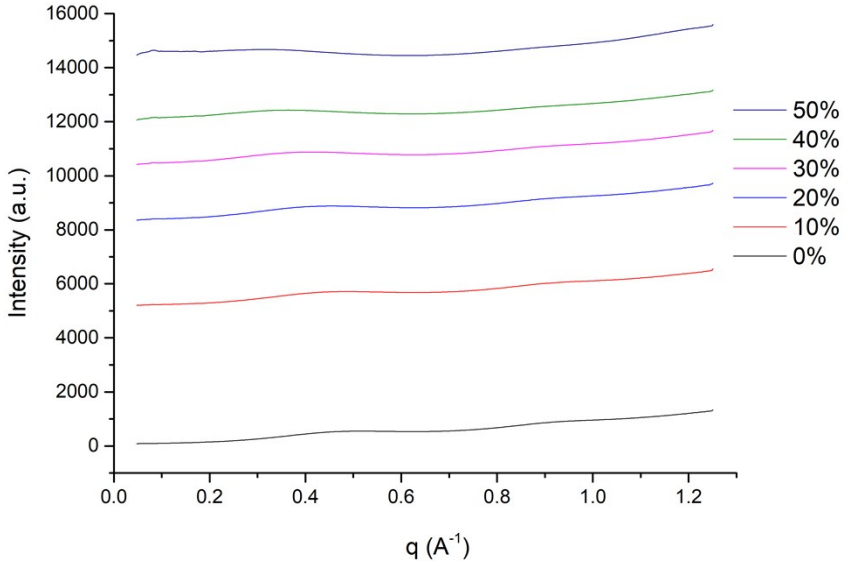

Figure S5. SAXS scattering patterns of CyCN-[C_4C_1im][Me_2PO_4] mixtures.

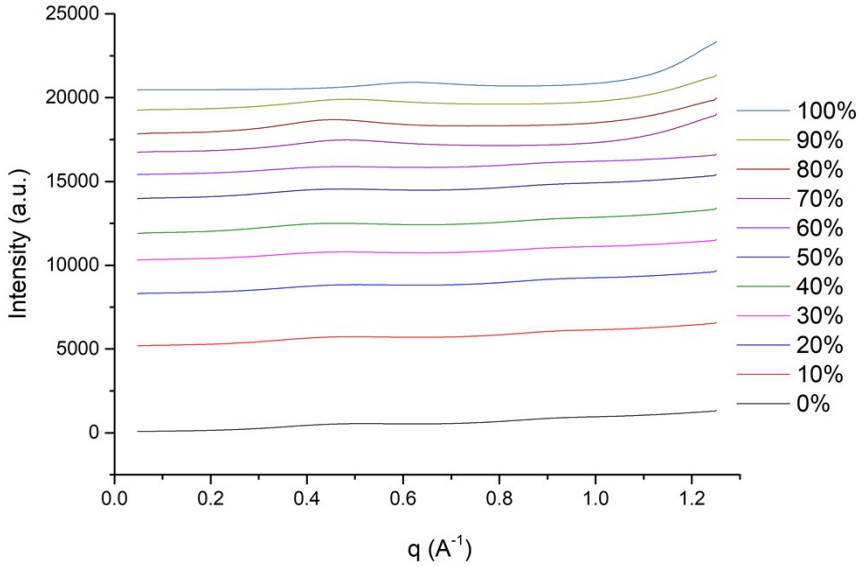

Figure S6. SAXS scattering patterns of CyOH-[C_4C_1im][Me_2PO_4] mixtures.

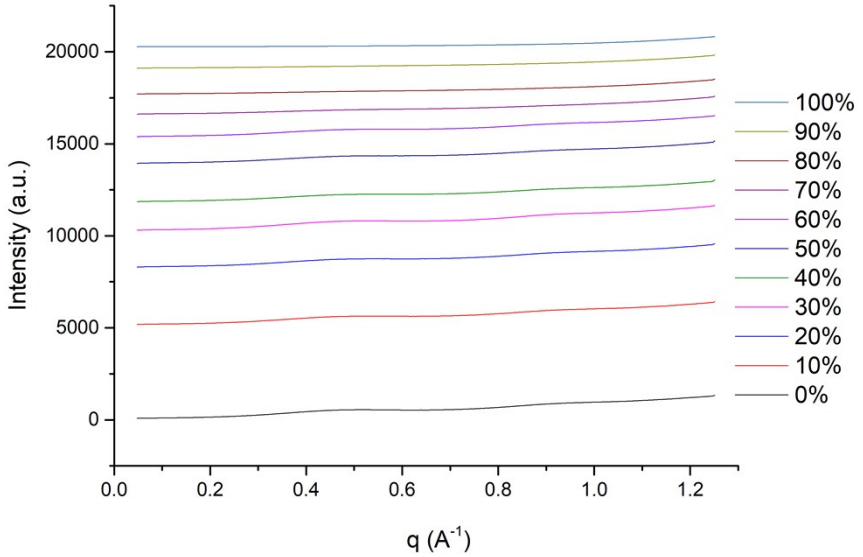

Figure S7. SAXS scattering patterns of DMSO-[C_4C_1im][Me_2PO_4] mixtures.

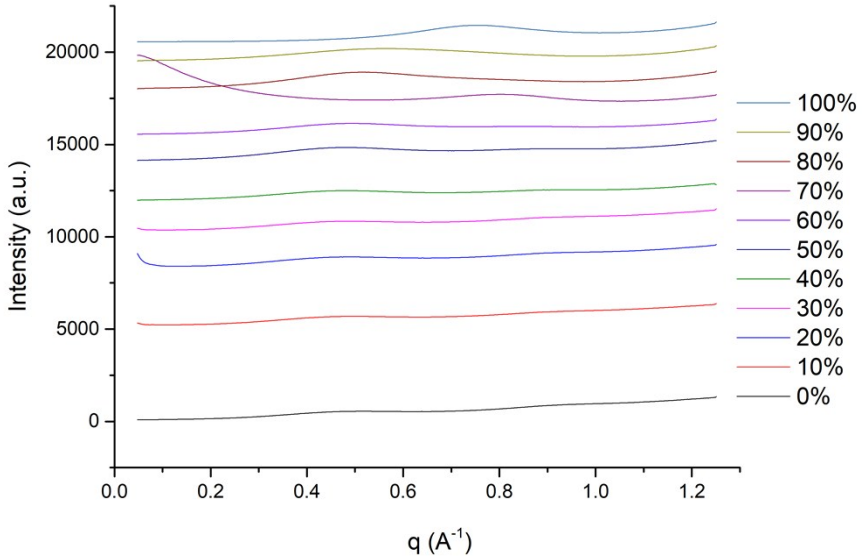

Figure S8. SAXS scattering patterns of IPA-[C_4C_1im][Me_2PO_4] mixtures.

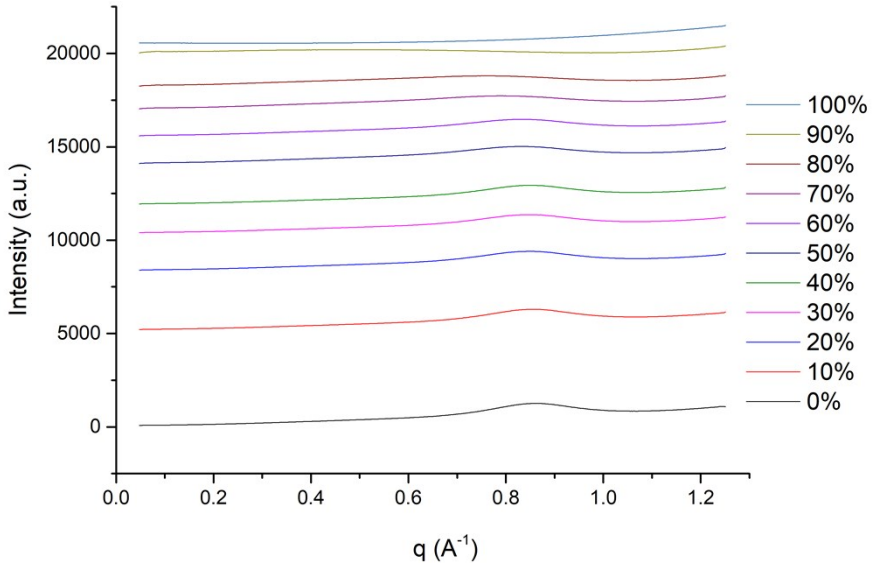

Figure S9. SAXS scattering patterns of ACE-[C_4C_1im][NTf_2] mixtures.

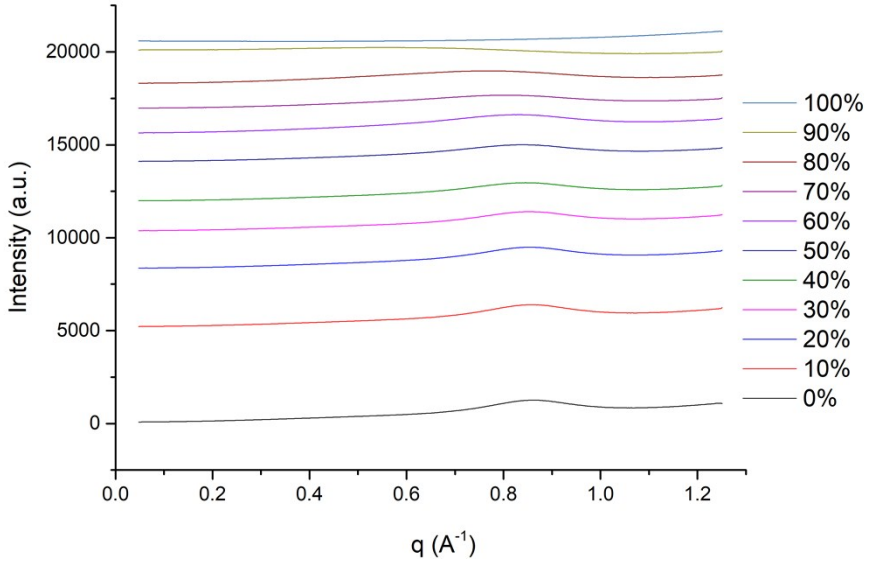

Figure S10. SAXS scattering patterns of ACN-[C_4C_1im][NTf_2] mixtures.

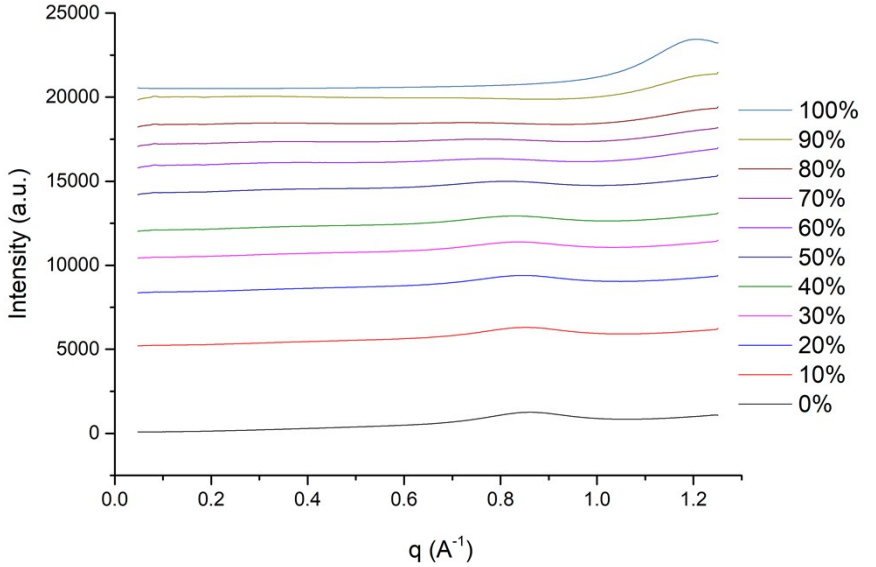

Figure S11. SAXS scattering patterns of CyCN-[C_4C_1im][NTf_2] mixtures.

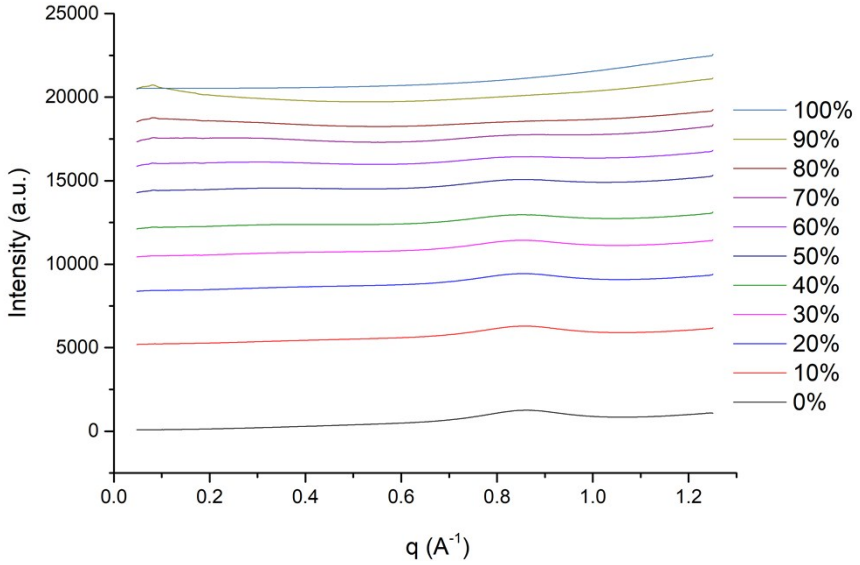

Figure S12. SAXS scattering patterns of CyOAc-[C_4C_1im][NTf_2] mixtures.

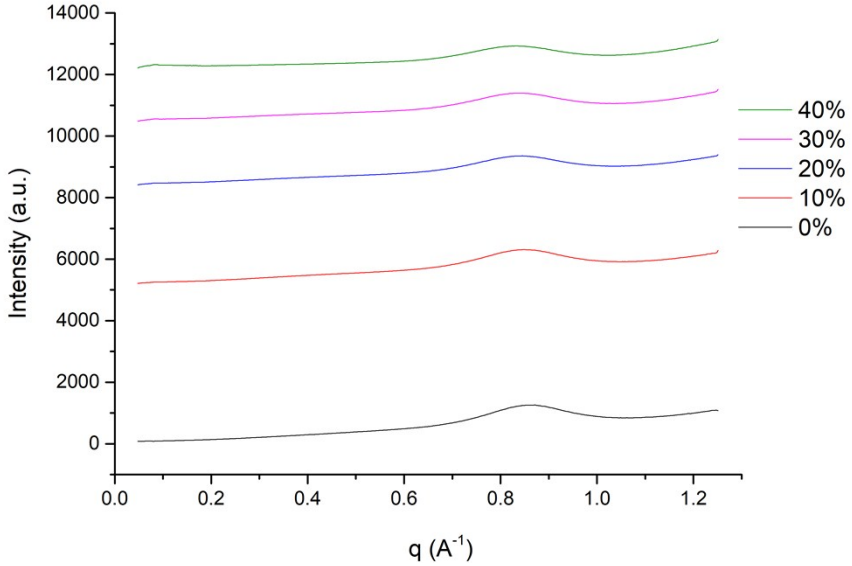

Figure S13. SAXS scattering patterns of CyOH-[C_4C_1im][NTf_2] mixtures.

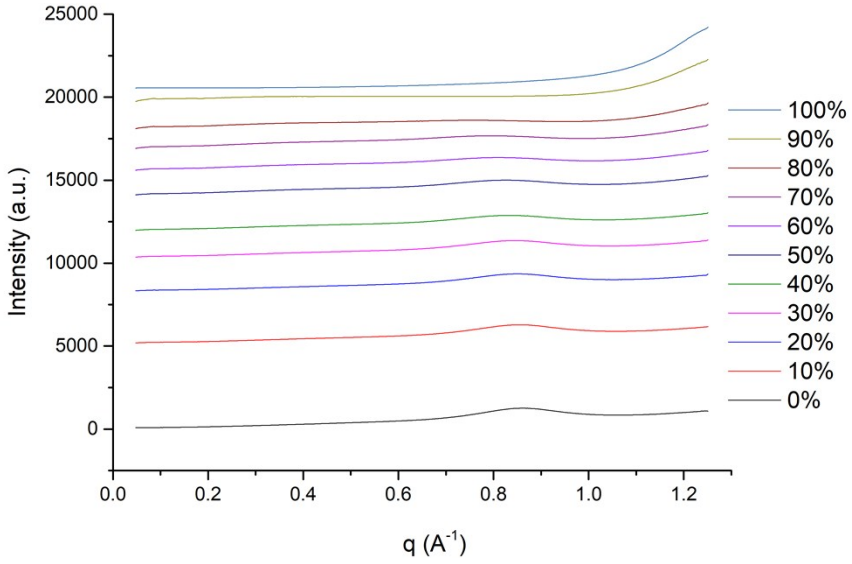

Figure S14. SAXS scattering patterns of Cyone-[C_4C_1im][NTf_2] mixtures.

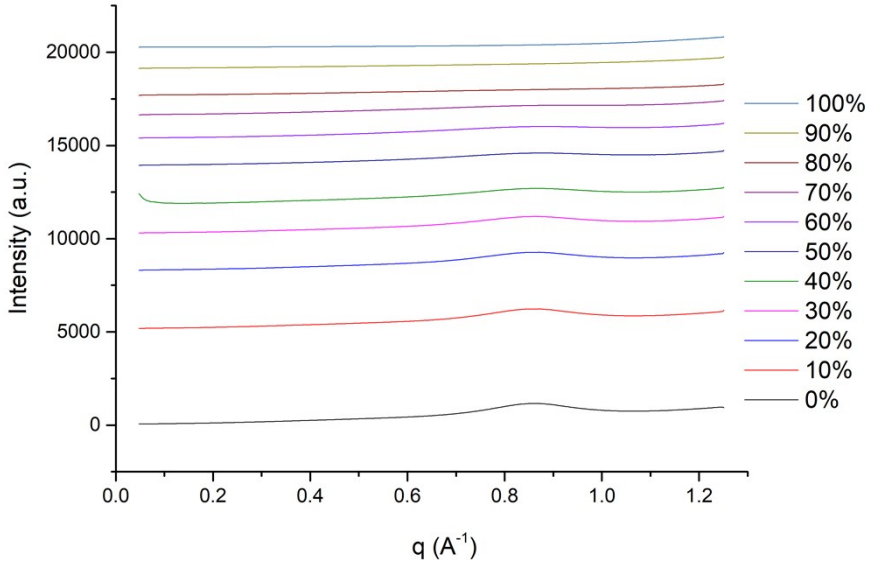

Figure S15. SAXS scattering patterns of DMSO-[C_4C_1im][NTf_2] mixtures.

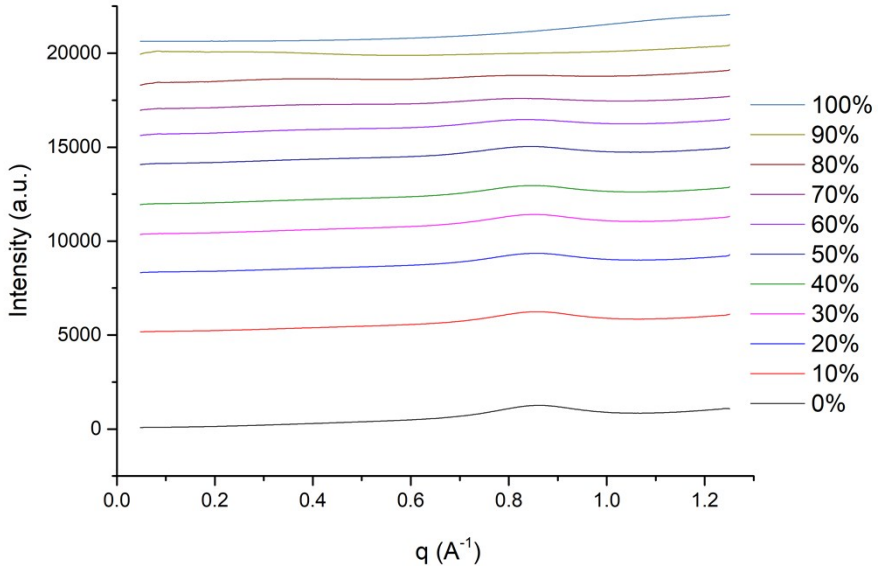

Figure S16. SAXS scattering patterns of EtOAc-[C_4C_1im][NTf_2] mixtures.

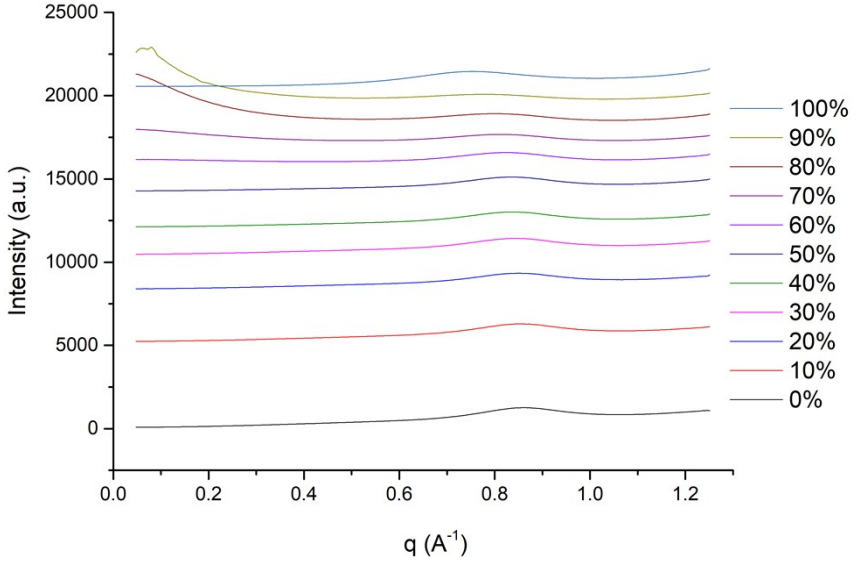

Figure S17. SAXS scattering patterns of IPA-[C_4C_1im][NTf_2] mixtures.

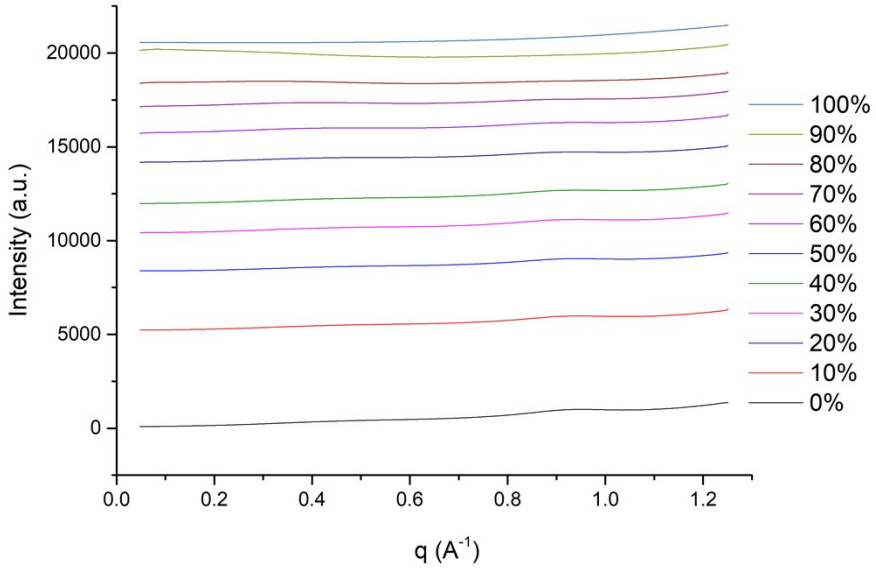

Figure S18. SAXS scattering patterns of ACE-[C_4C_1im][OTf] mixtures.

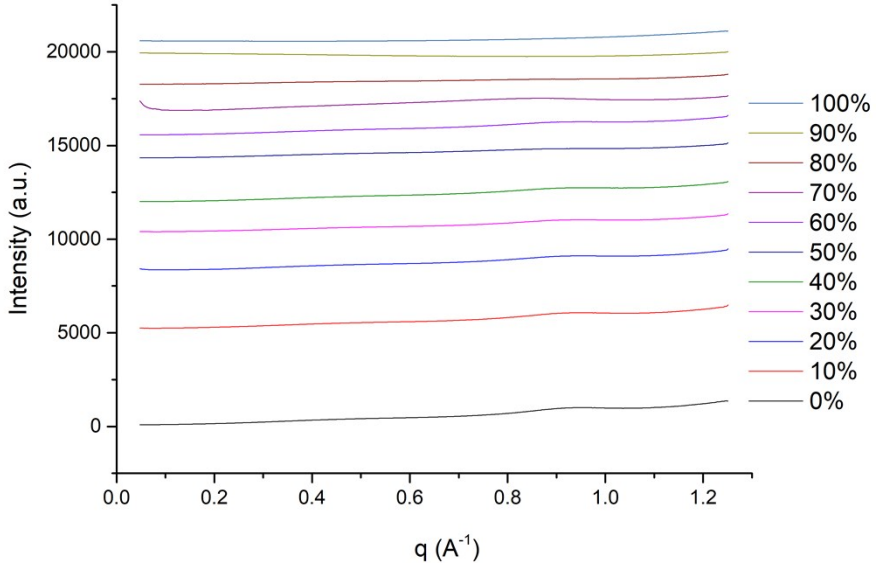

Figure S19. SAXS scattering patterns of ACN-[C_4C_1im][OTf] mixtures.

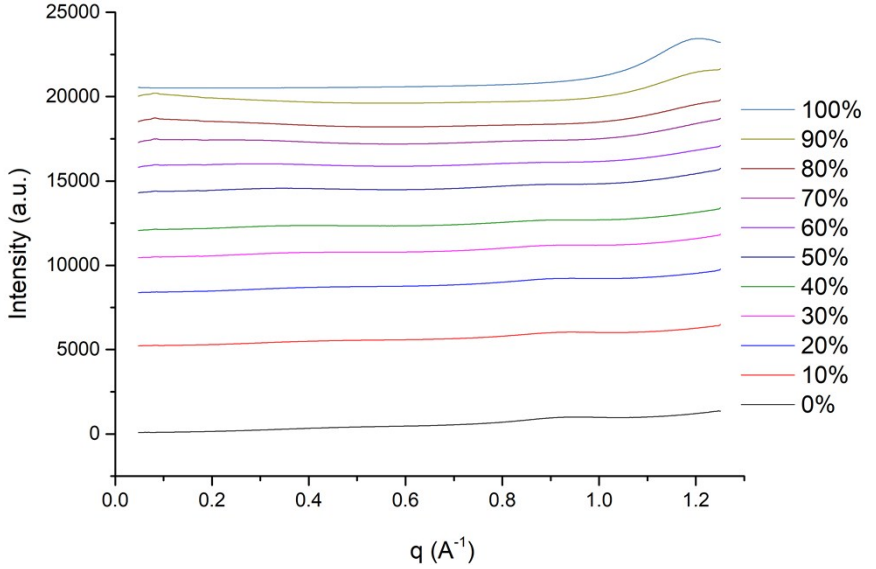

Figure S20. SAXS scattering patterns of CyCN-[C_4C_1im][OTf] mixtures.

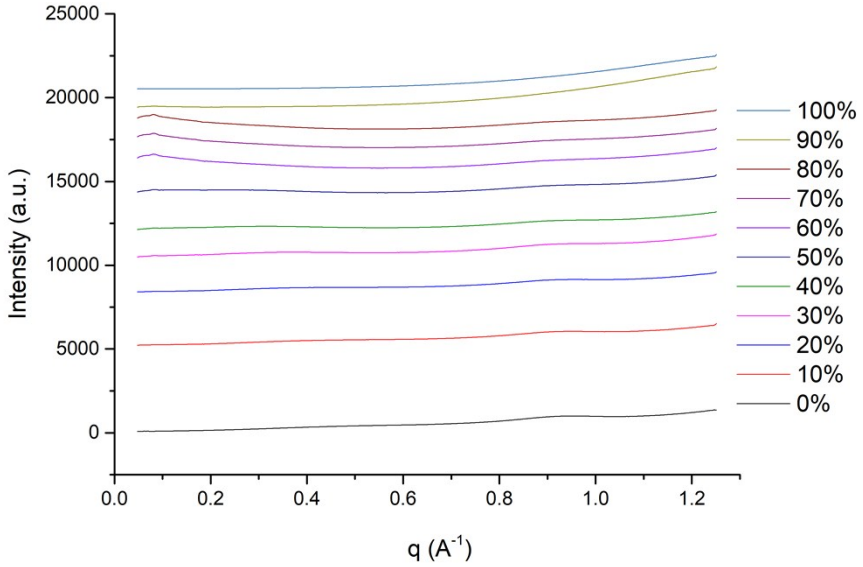

Figure S21. SAXS scattering patterns of CyOAc-[C_4C_1im][OTf] mixtures.

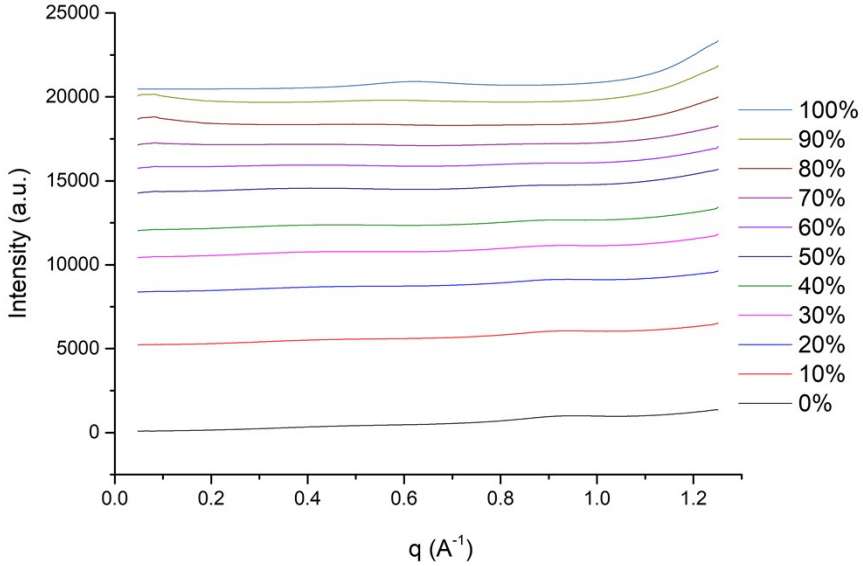

Figure S22. SAXS scattering patterns of CyOH-[C_4C_1im][OTf] mixtures.

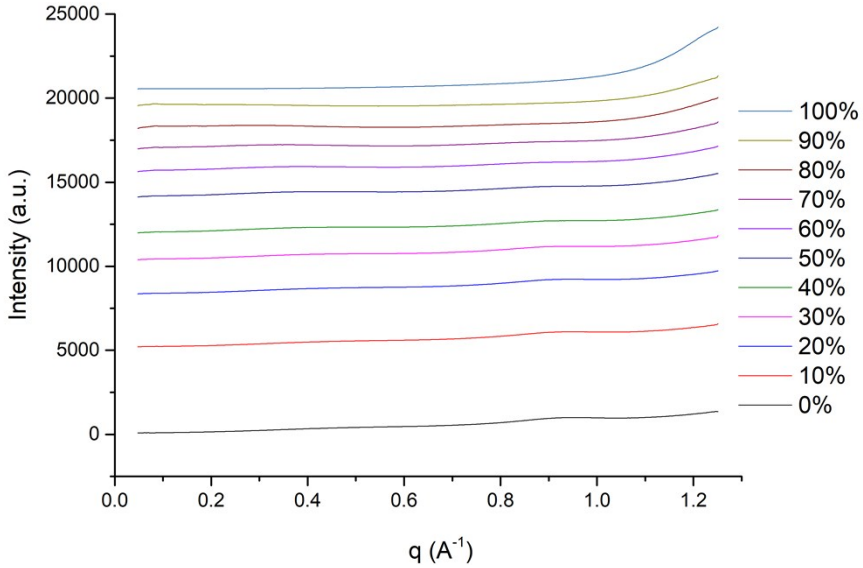

Figure S23. SAXS scattering patterns of Cyone-[C_4C_1im][OTf] mixtures.

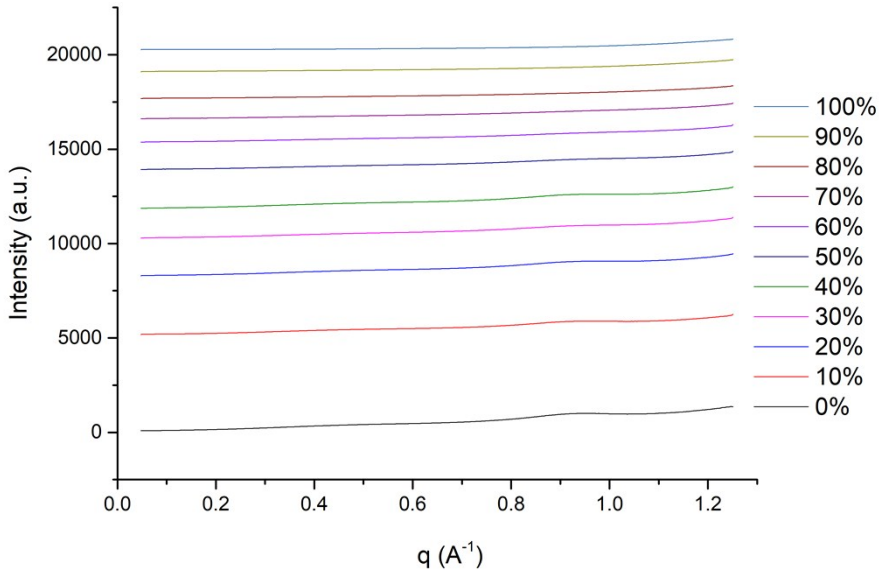

Figure S24. SAXS scattering patterns of DMSO-[C_4C_1im][OTf] mixtures.

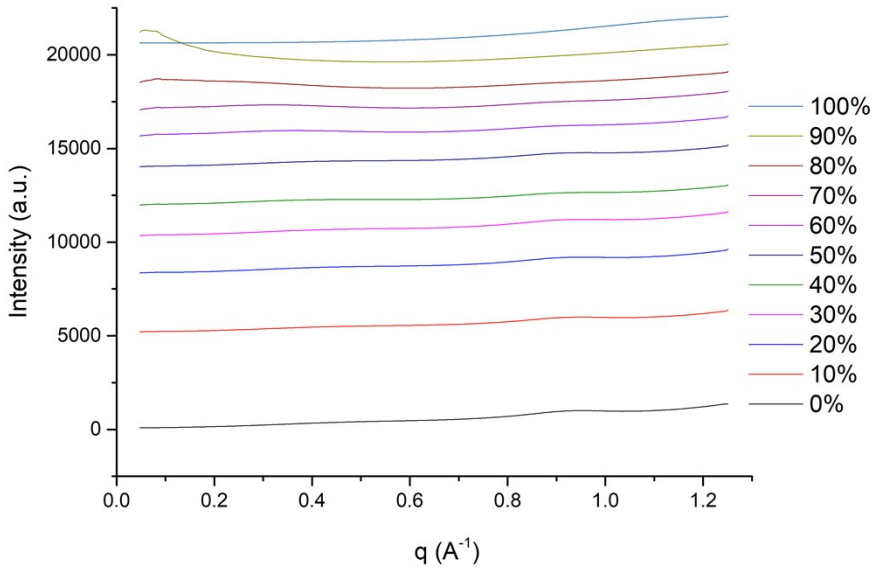

Figure S25. SAXS scattering patterns of EtOAc-[C_4C_1im][OTf] mixtures.

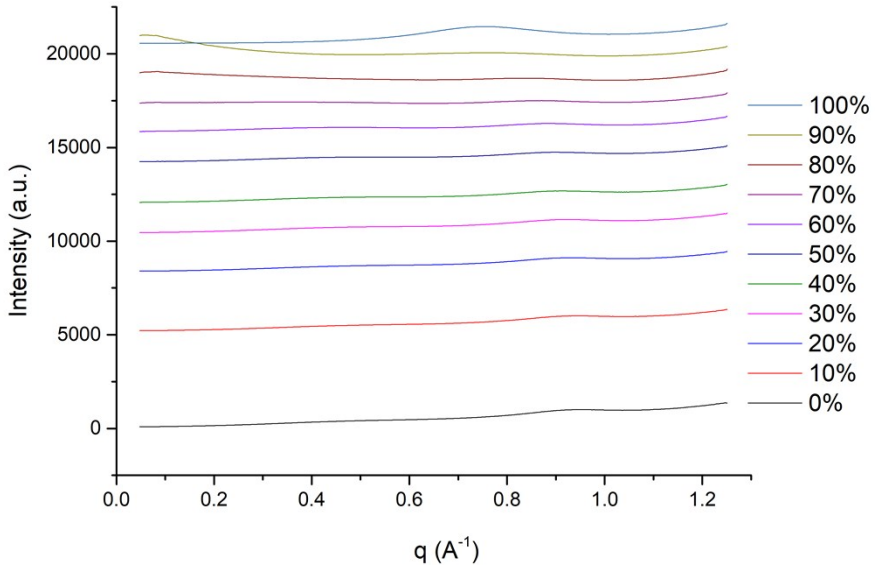

Figure S26. SAXS scattering patterns of IPA-[C_4C_1im][OTf] mixtures.

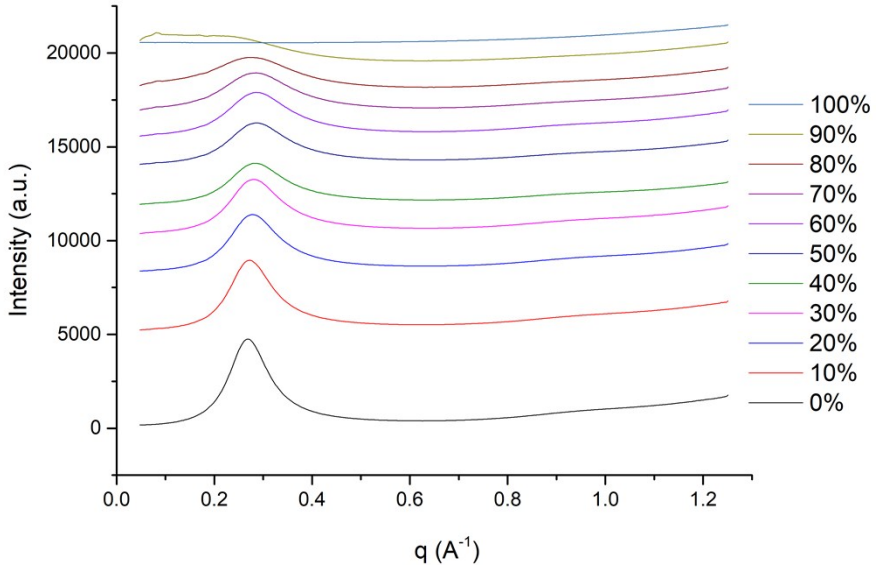

Figure S27. SAXS scattering patterns of ACE-[$C_{10}C_1$im][Me_2PO_4] mixtures.

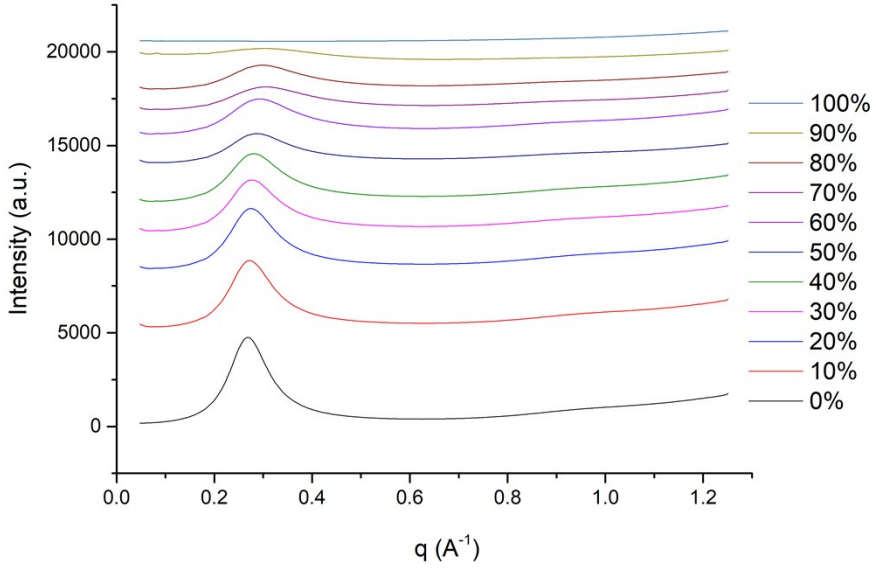

Figure S28. SAXS scattering patterns of ACN-[$C_{10}C_1$im][Me_2PO_4] mixtures.

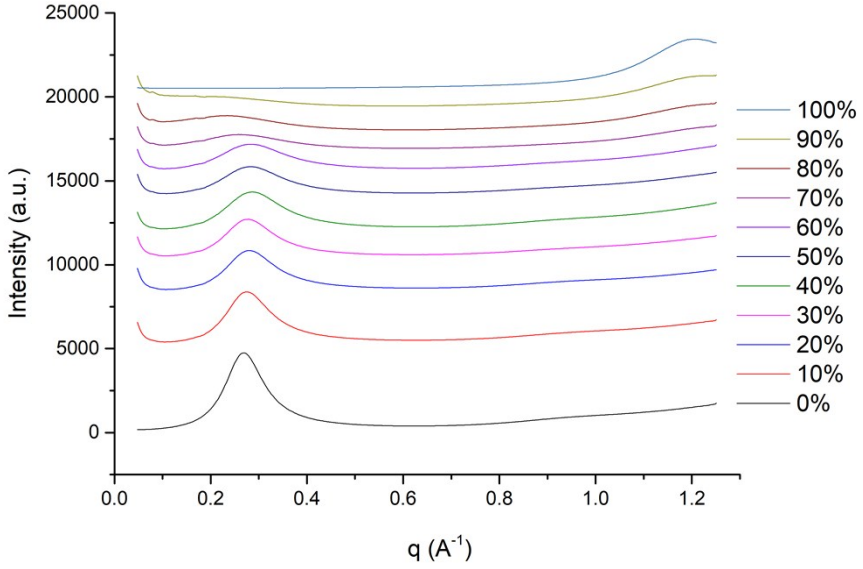

Figure S29. SAXS scattering patterns of CyCN-[$C_{10}C_1$im][Me_2PO_4] mixtures.

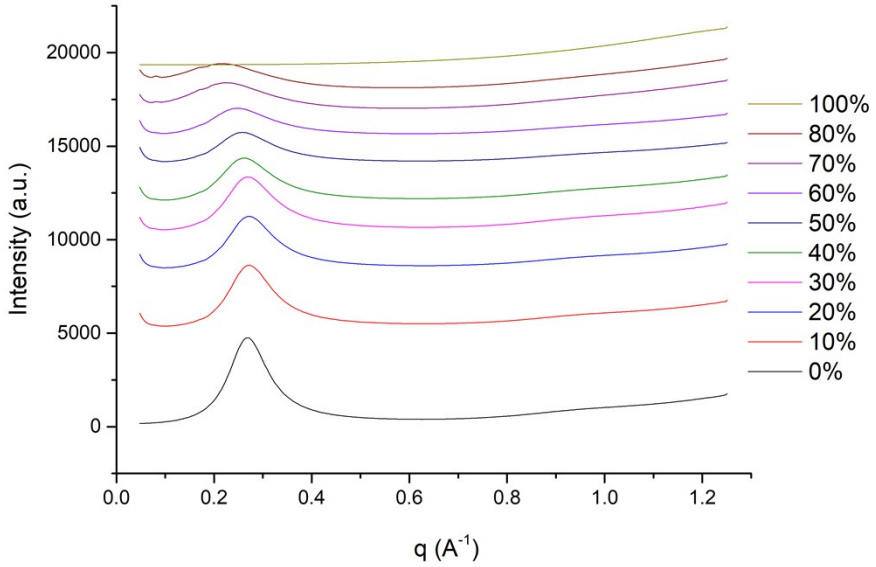

Figure S30. SAXS scattering patterns of CyOAc-[$C_{10}C_1$im][Me_2PO_4] mixtures.

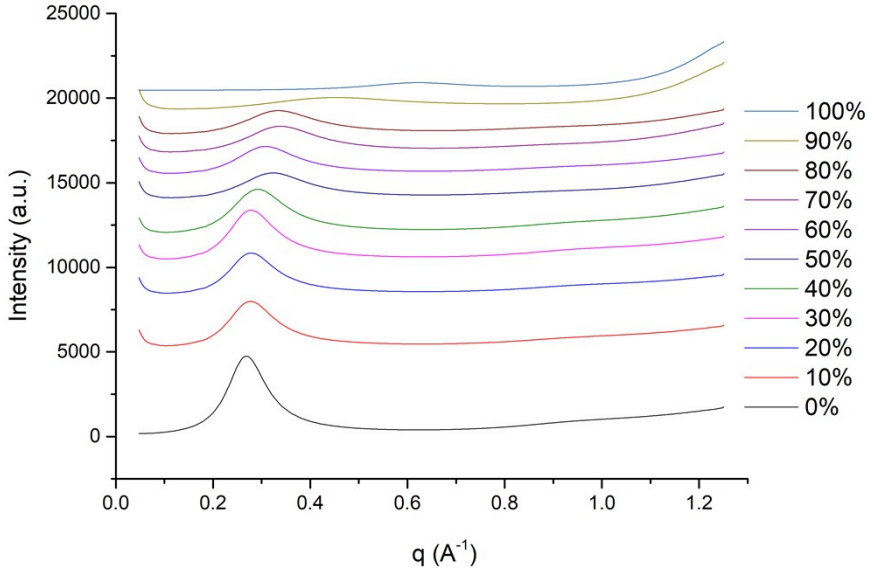

Figure S31. SAXS scattering patterns of CyOH-[$C_{10}C_1$im][Me_2PO_4] mixtures.

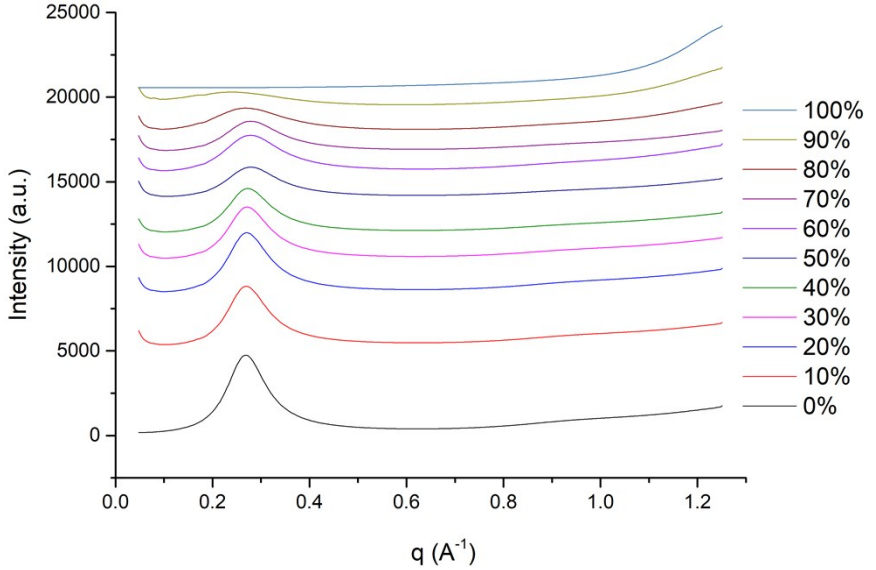

Figure S32. SAXS scattering patterns of Cyone-[$C_{10}C_1$im][Me_2PO_4] mixtures.

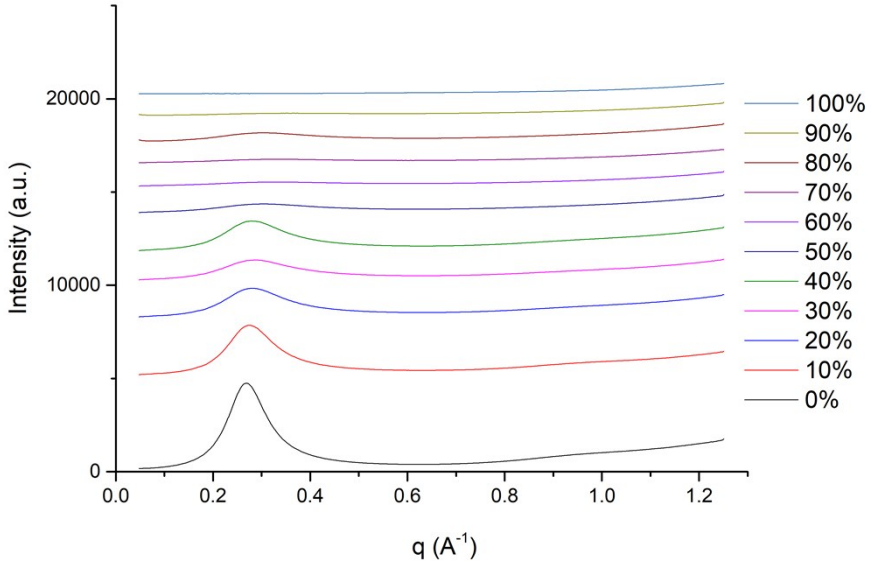

Figure S33. SAXS scattering patterns of DMSO-[$C_{10}C_1$im][Me_2PO_4] mixtures.

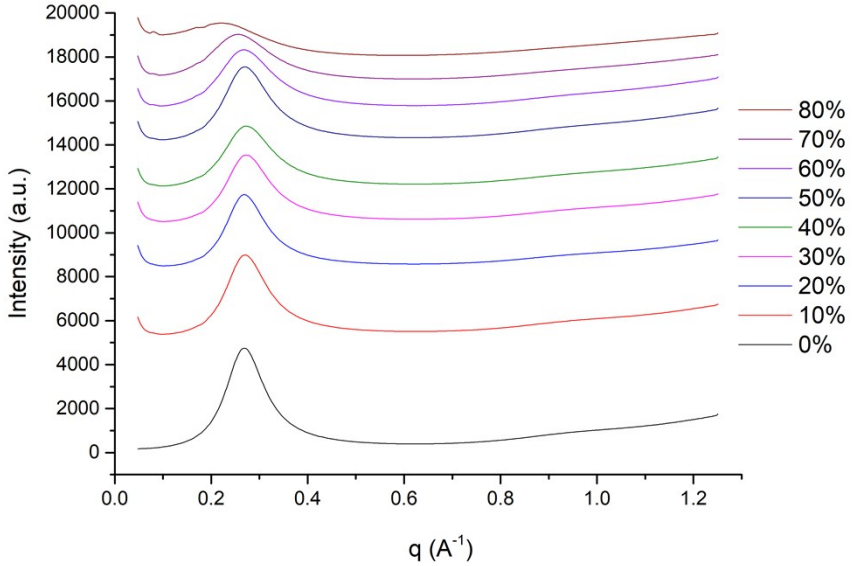

Figure S34. SAXS scattering patterns of EtOAc-[$C_{10}C_1$im][Me_2PO_4] mixtures.

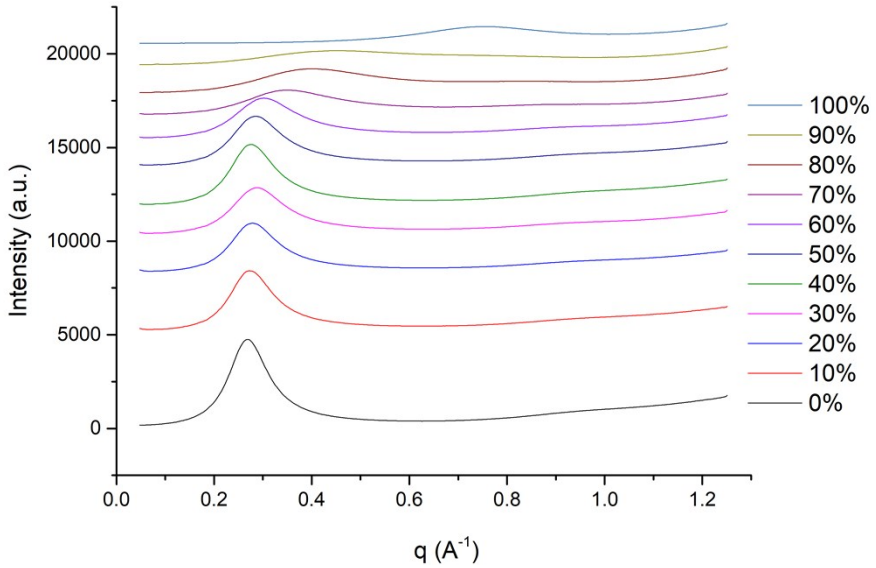

Figure S35. SAXS scattering patterns of IPA-[$C_{10}C_1$im][Me_2PO_4] mixtures.

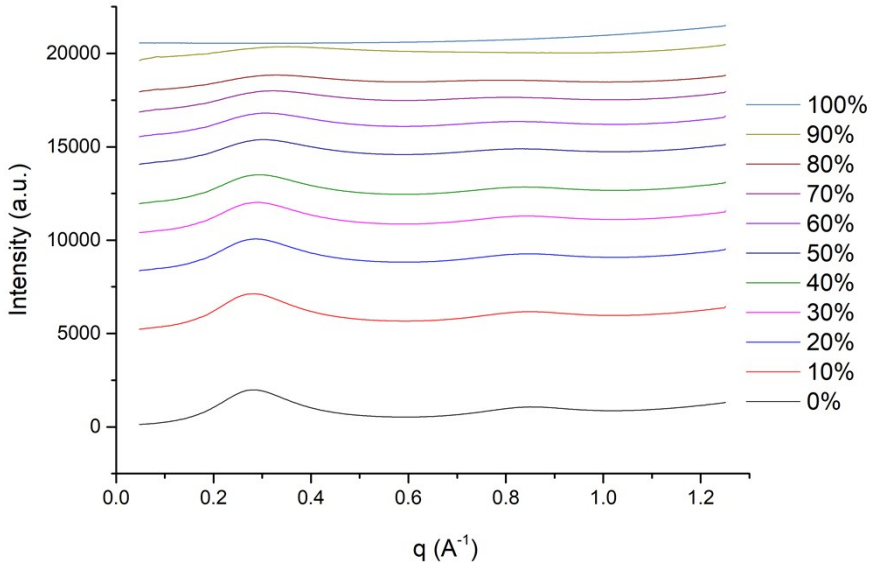

Figure S36. SAXS scattering patterns of ACE-[$C_{10}C_1$im][NTf_2] mixtures.

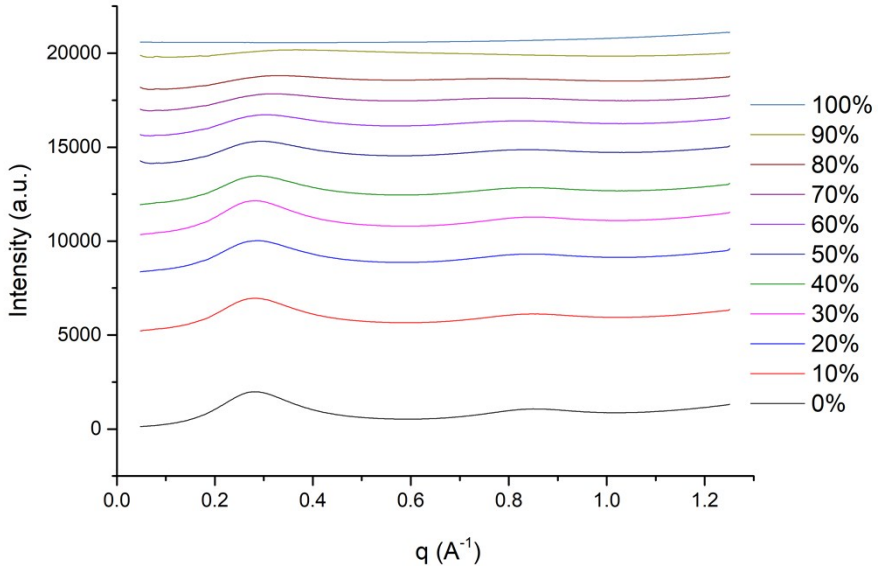

Figure S37. SAXS scattering patterns of ACN-[$C_{10}C_1$im][NTf_2] mixtures.

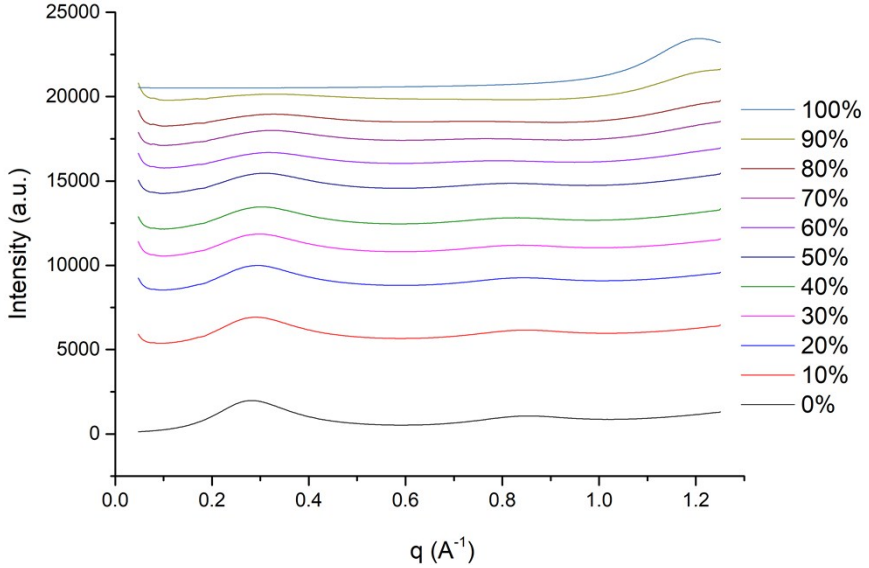

Figure S38. SAXS scattering patterns of CyCN-[$C_{10}C_1$im][NTf_2] mixtures.

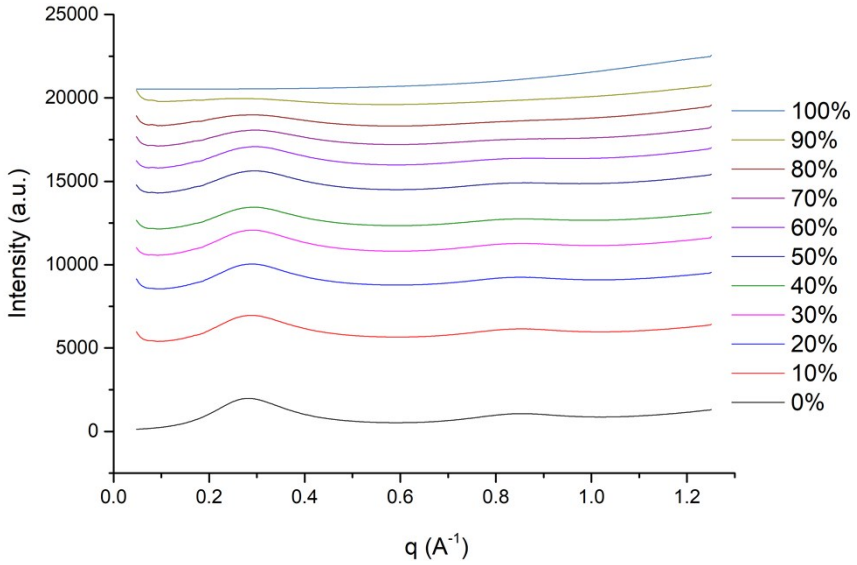

Figure S39. SAXS scattering patterns of CyOAc-[$C_{10}C_1$im][NTf_2] mixtures.

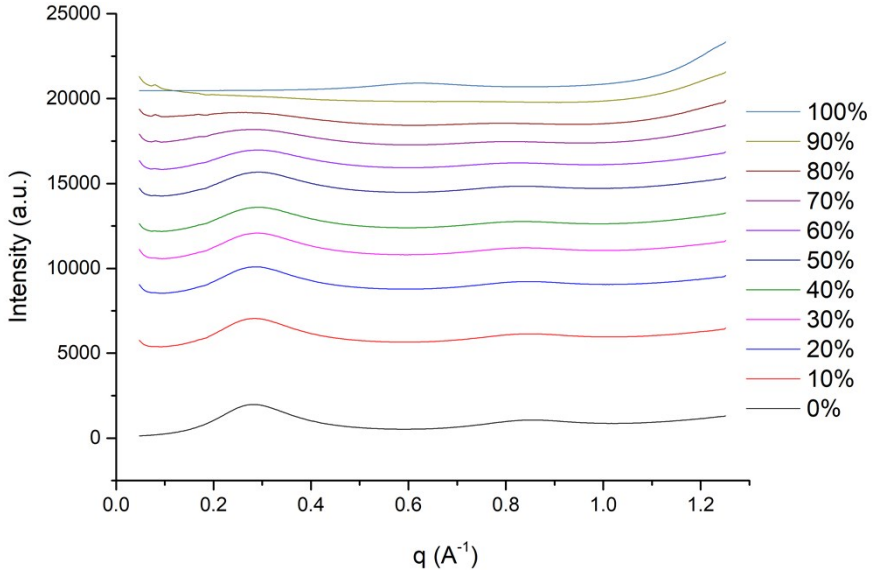

Figure S40. SAXS scattering patterns of CyOH-[$C_{10}C_1$im][NTf_2] mixtures.

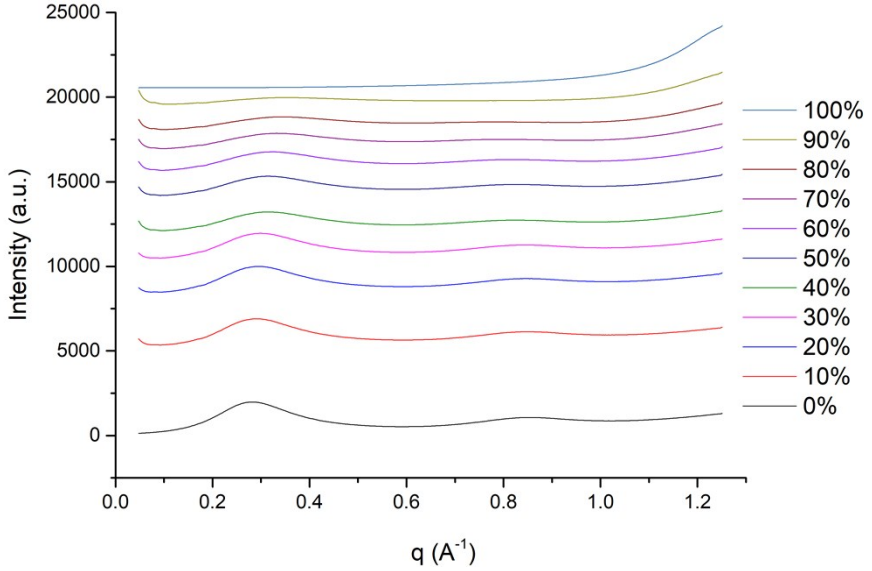

Figure S41. SAXS scattering patterns of Cyone-[$C_{10}C_1$im][NTf_2] mixtures.

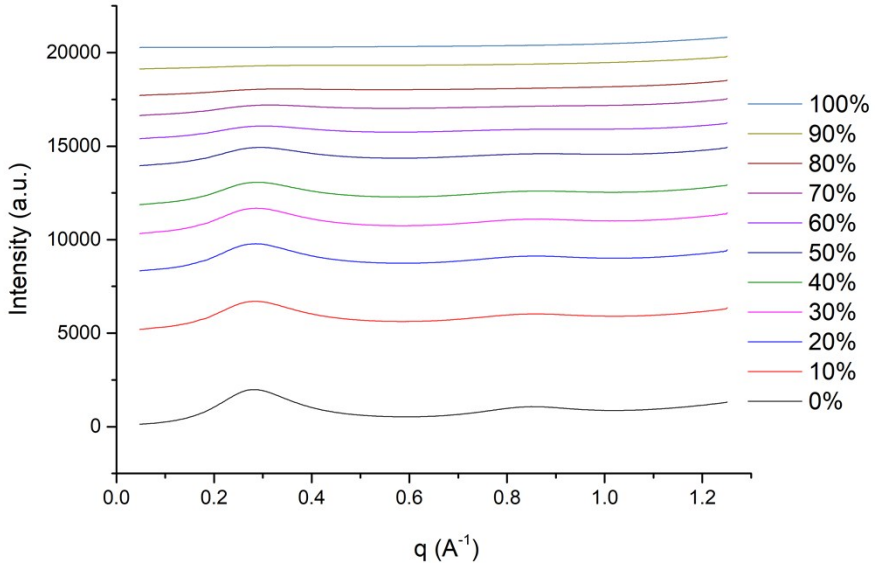

Figure S42. SAXS scattering patterns of DMSO-[$C_{10}C_1$im][NTf_2] mixtures.

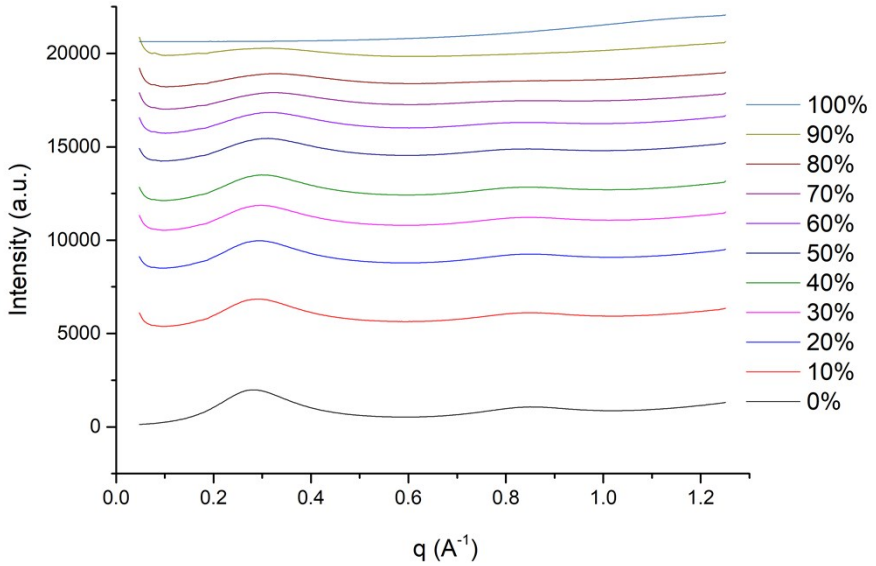

Figure S43. SAXS scattering patterns of EtOAc-[$C_{10}C_1$im][NTf_2] mixtures.

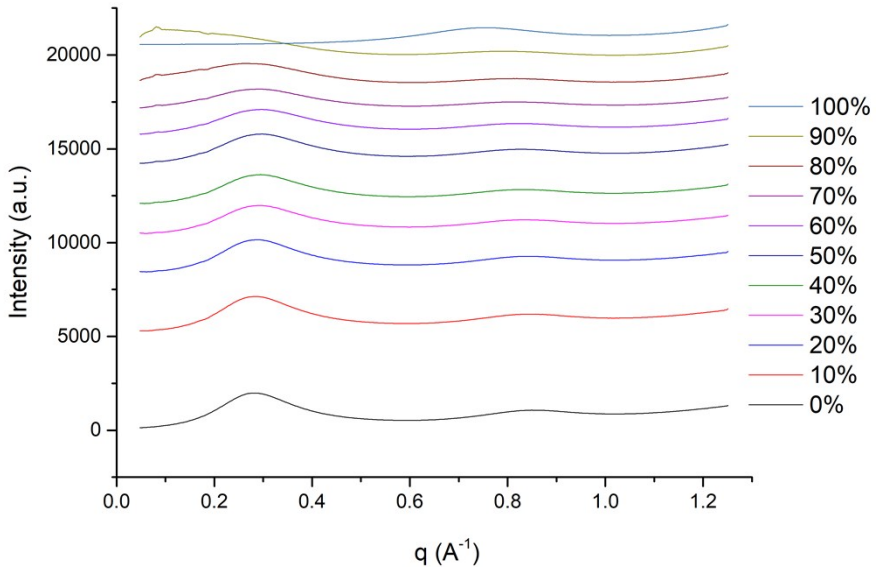

Figure S44. SAXS scattering patterns of IPA-[$C_{10}C_1$im][NTf_2] mixtures.

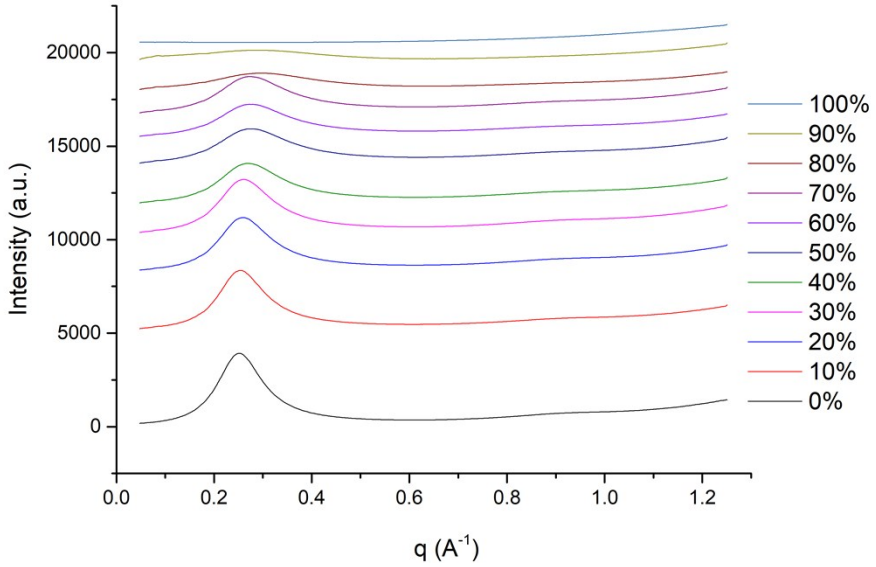

Figure S45. SAXS scattering patterns of ACE-[$C_{10}C_1$im][OTf] mixtures.

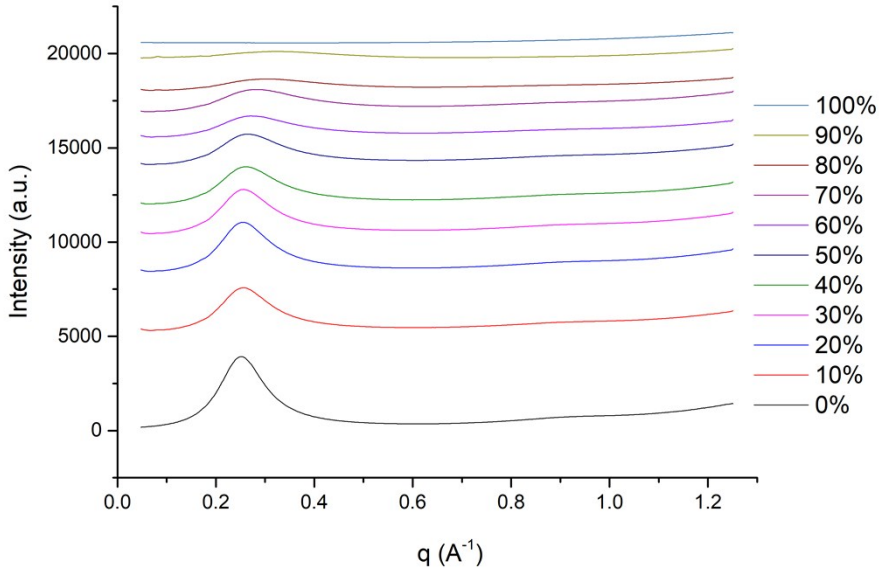

Figure S46. SAXS scattering patterns of ACN-[$C_{10}C_1$im][OTf] mixtures.

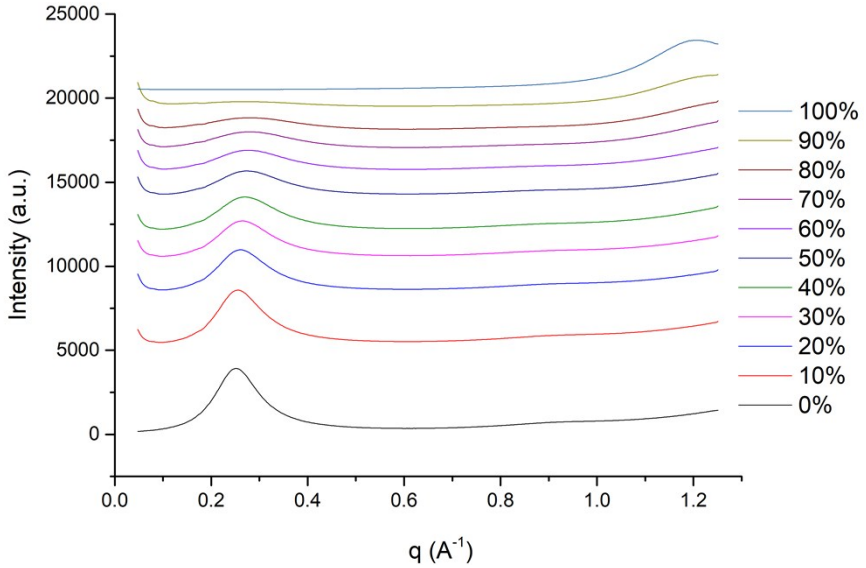

Figure S47. SAXS scattering patterns of CyCN-[$C_{10}C_1$im][OTf] mixtures.

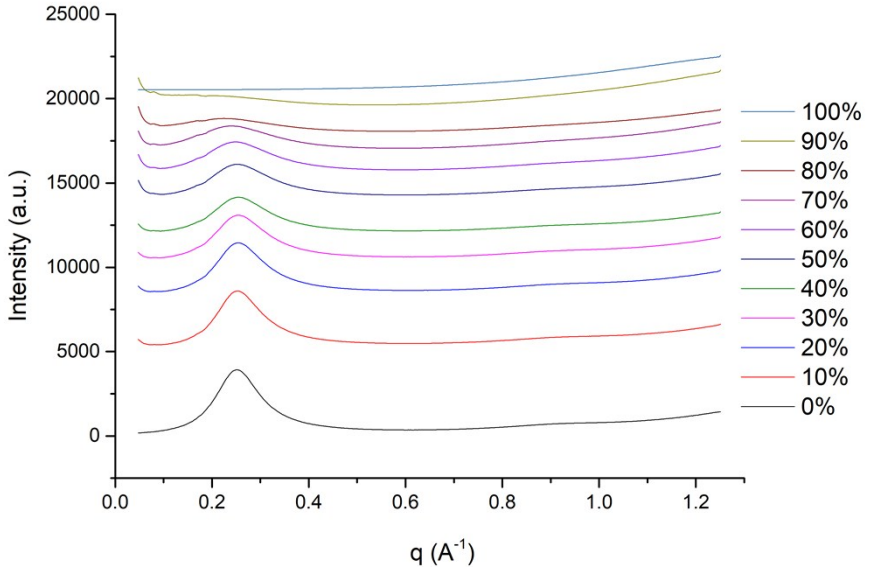

Figure S48. SAXS scattering patterns of CyOAc-[$C_{10}C_1$im][OTf] mixtures.

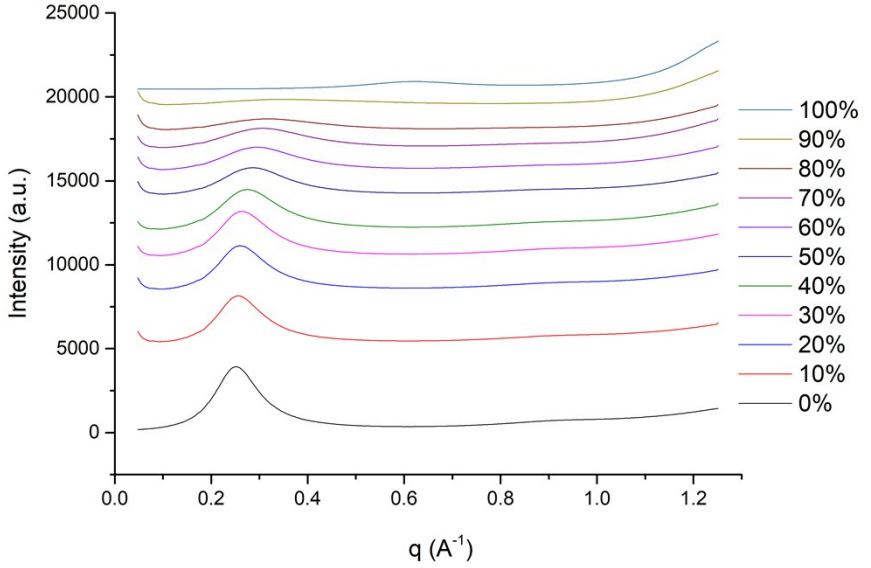

Figure S49. SAXS scattering patterns of CyOH-[$C_{10}C_1$im][OTf] mixtures.

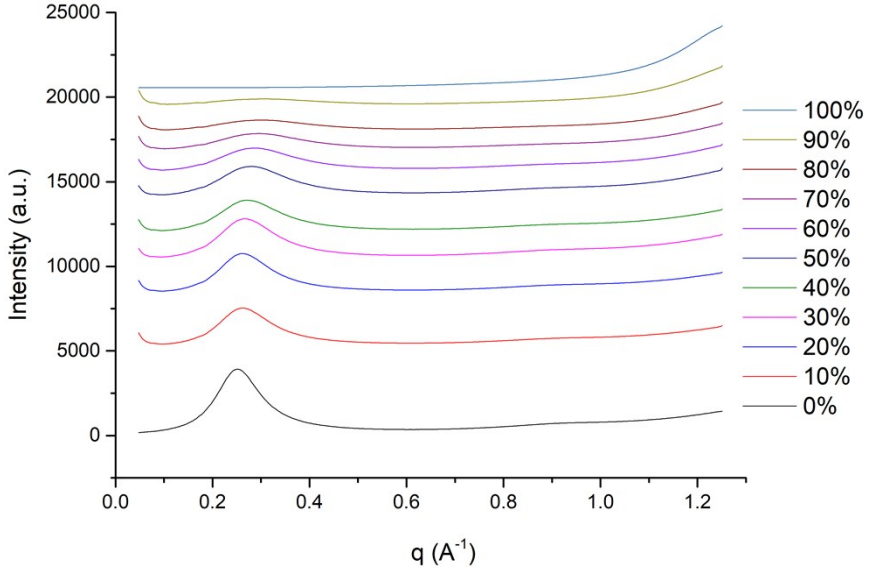

Figure S50. SAXS scattering patterns of Cyone-[$C_{10}C_1$im][OTf] mixtures.

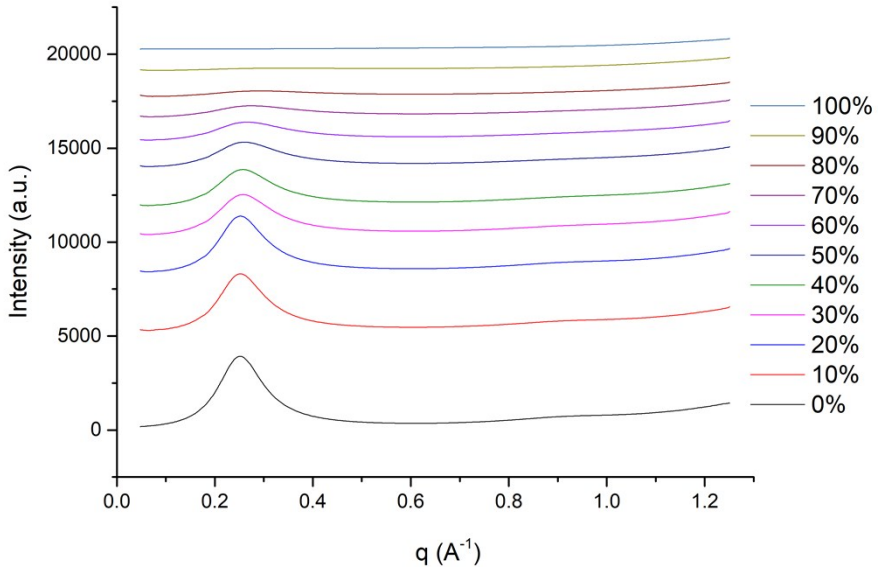

Figure S51. SAXS scattering patterns of DMSO-[$C_{10}C_1$im][OTf] mixtures.

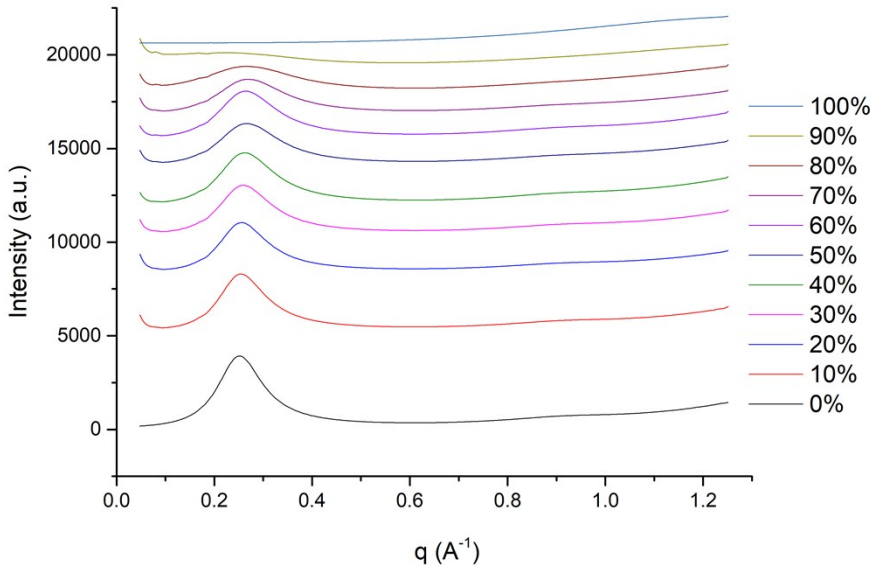

Figure S52. SAXS scattering patterns of EtOAc-[$C_{10}C_1$im][OTf] mixtures.

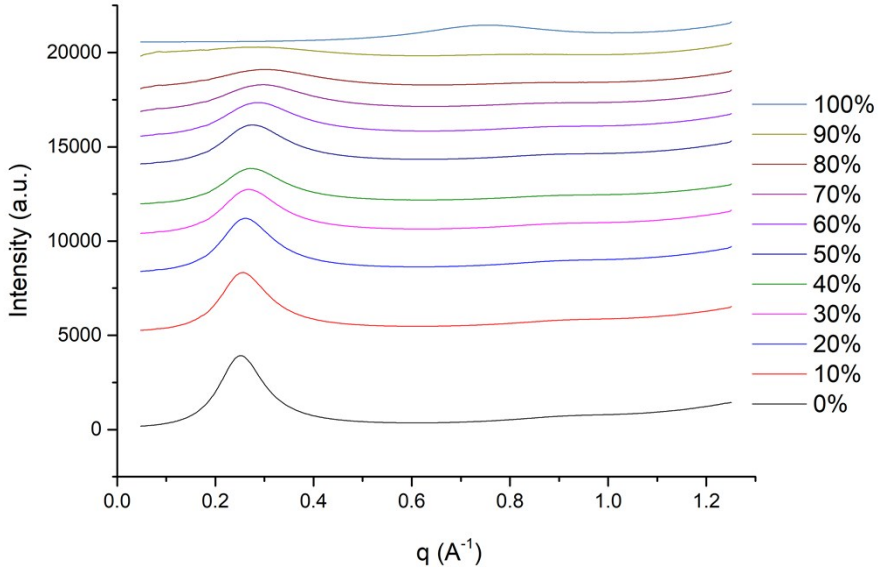

Figure S53. SAXS scattering patterns of IPA-[$C_{10}C_1$im][OTf] mixtures.

Exemplar Fits of SAXS Scattering Patterns for DMSO-[C_4C_1im][NTf_2]

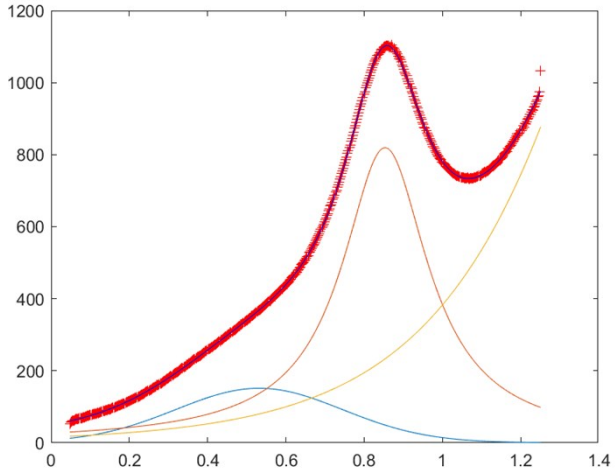

Figure S54. SAXS scattering pattern and associated fit of 10 mol% DMSO-[C_4C_1im][NTf_2].

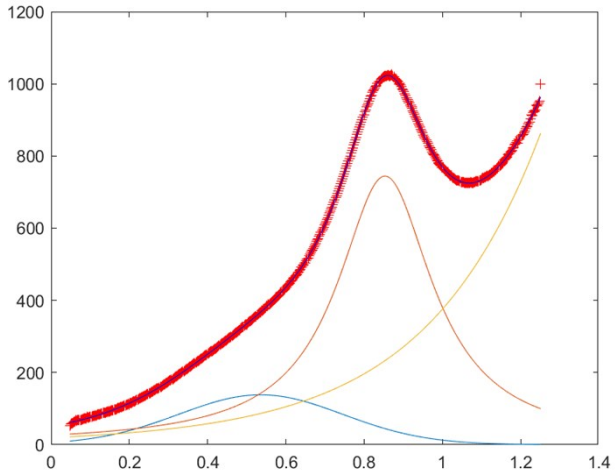

Figure S55. SAXS scattering pattern and associated fit of 20 mol% DMSO-[C_4C_1im][NTf_2].

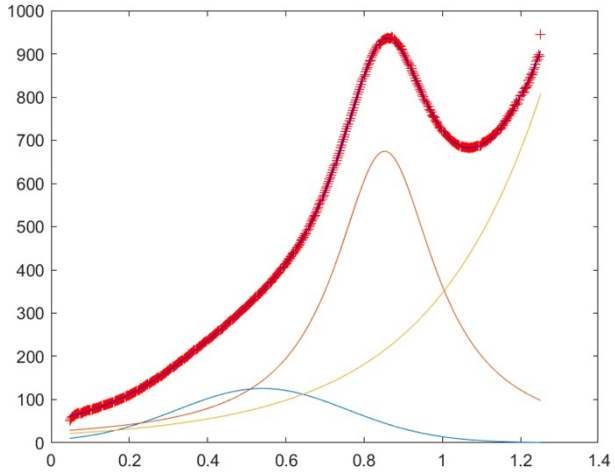

Figure S56. SAXS scattering pattern and associated fit of 30 mol% DMSO-[C_4C_1im][NTf_2].

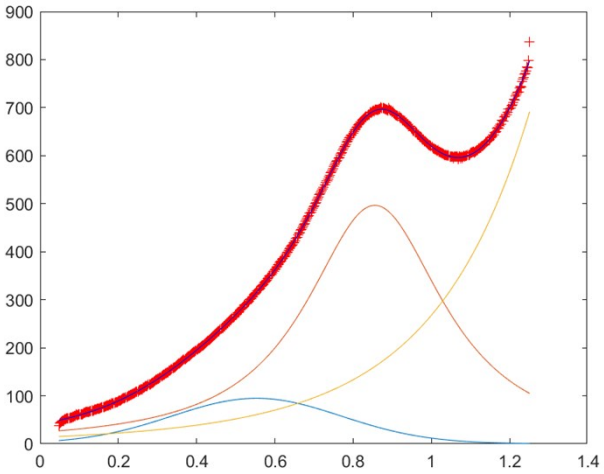

Figure S57. SAXS scattering pattern and associated fit of 40 mol% DMSO-[C_4C_1im][NTf_2].

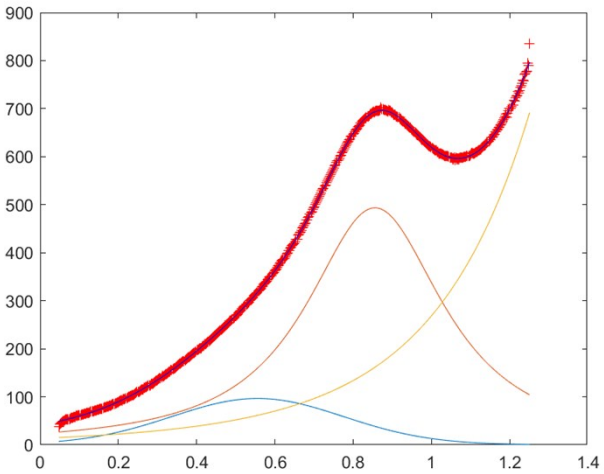

Figure S58. SAXS scattering pattern and associated fit of 50 mol% DMSO-[C_4C_1im][NTf_2].

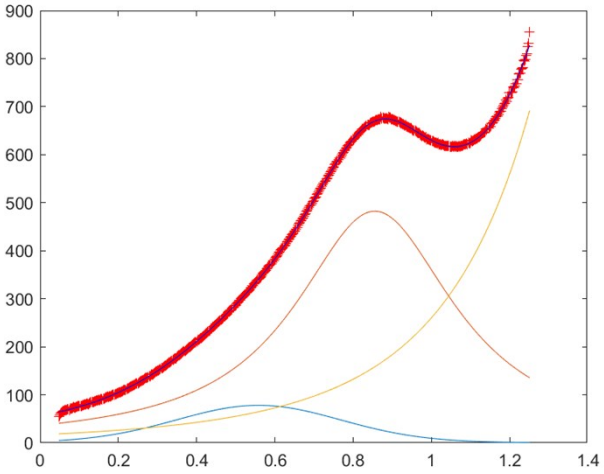

Figure S59. SAXS scattering pattern and associated fit of 60 mol% DMSO-[C_4C_1im][NTf_2].

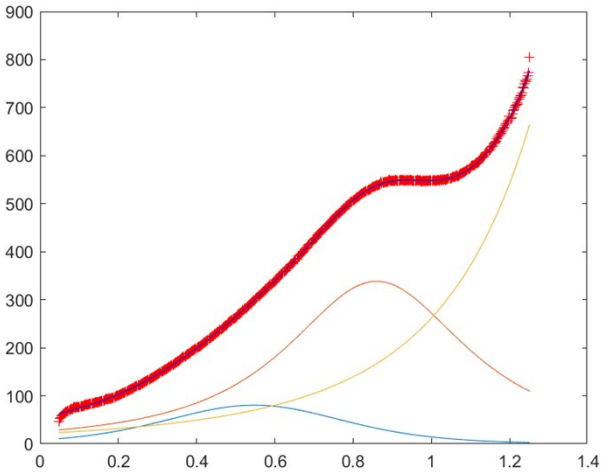

Figure S60. SAXS scattering pattern and associated fit of 70 mol% DMSO-[C_4C_1im][NTf_2].

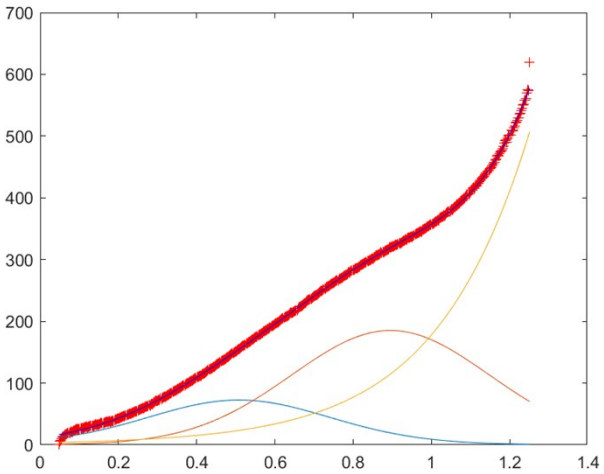

Figure S61. SAXS scattering pattern and associated fit of 80 mol% DMSO-[C_4C_1im][NTf_2].

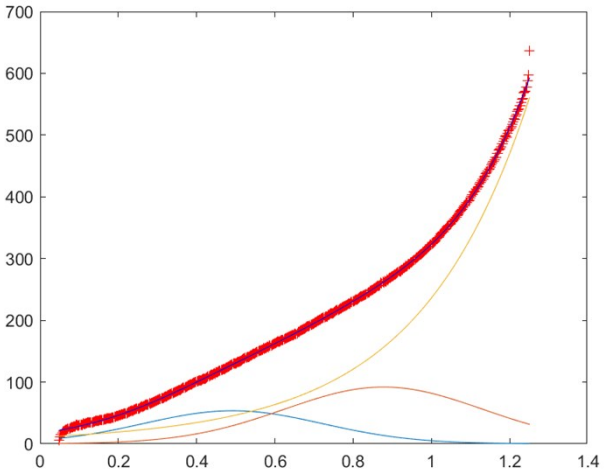

Figure S62. SAXS scattering pattern and associated fit of 80 mol% DMSO-[C_4C_1im][NTf_2].

IR Spectroscopy Measurements

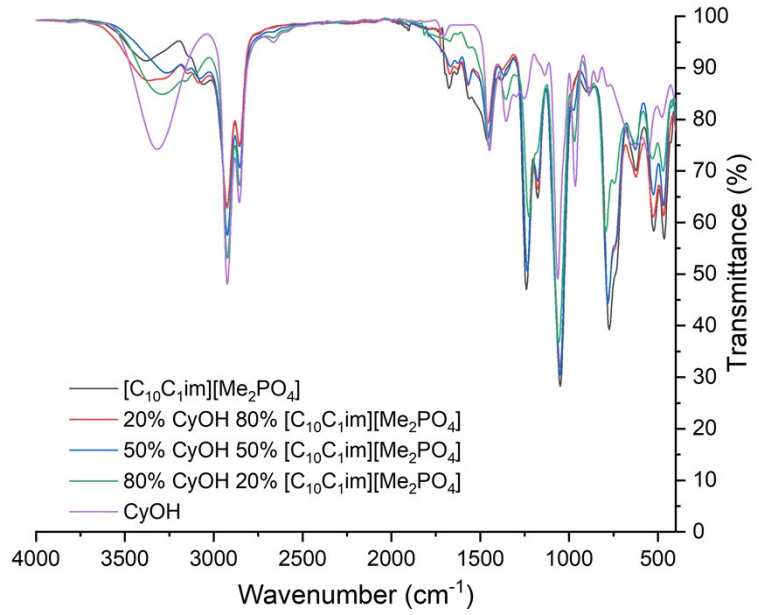

Figure S63. Complete IR spectrum for CyOH and $[C_{10}C_1im][Me_2PO_4]$ mixtures.

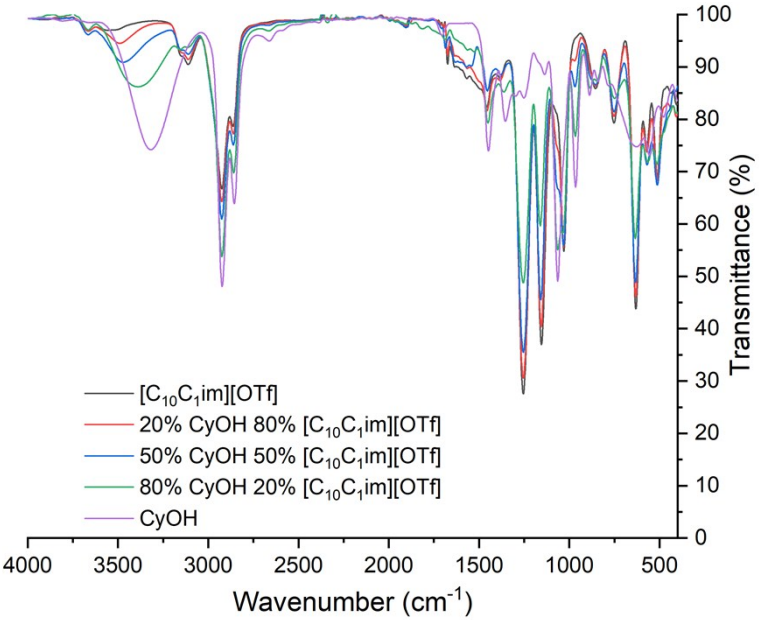

Figure S64. Complete IR spectrum for CyOH and $[C_{10}C_1im][OTf]$ mixtures.

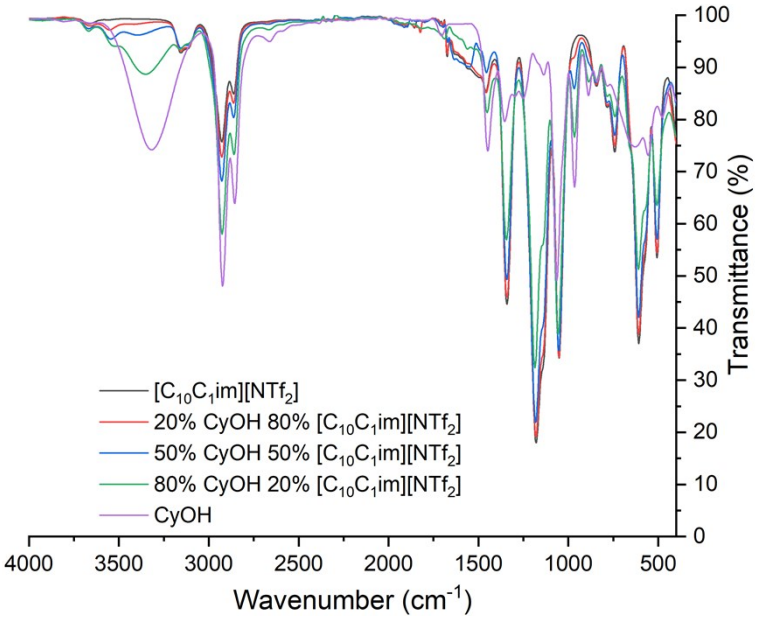

Figure S65. Complete IR spectrum for CyOH and $[C_{10}C_1im][NTf_2]$ mixtures.

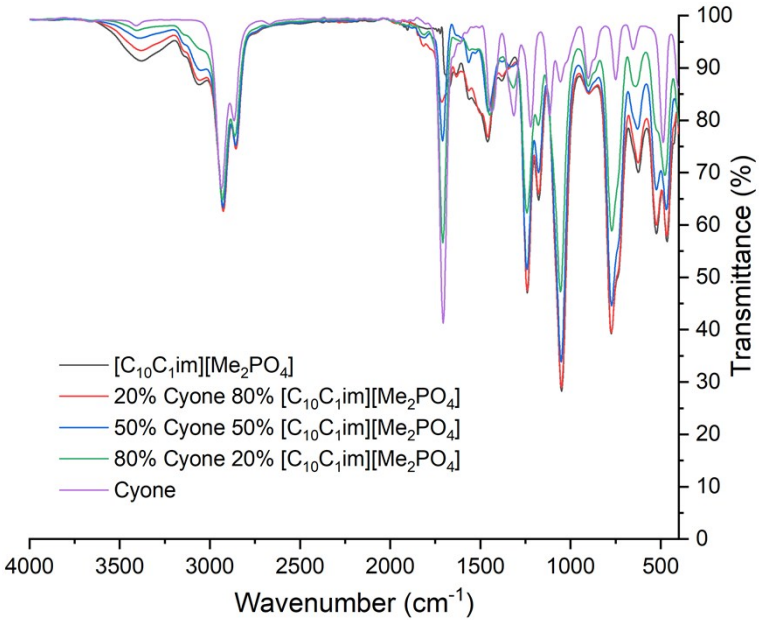

Figure S66. Complete IR spectrum for Cyone and $[C_{10}C_1im][Me_2PO_4]$ mixtures.

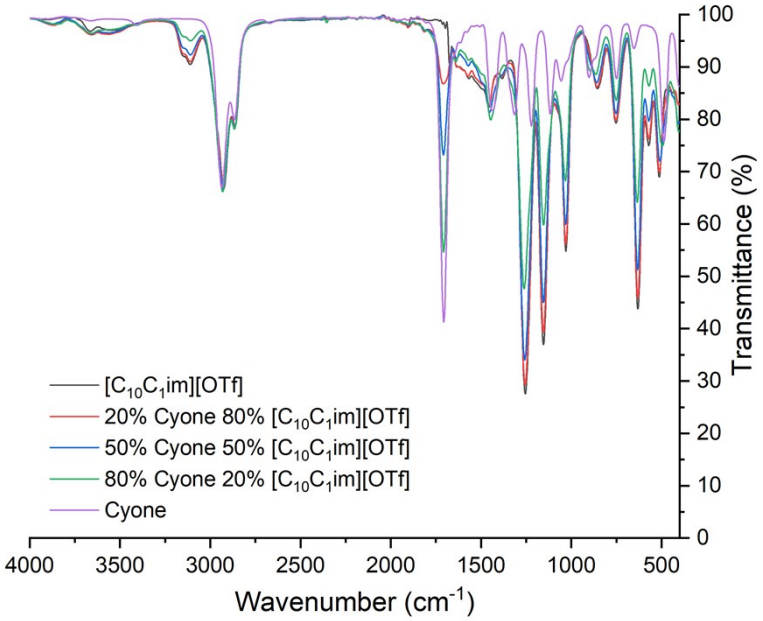

Figure S67. Complete IR spectrum for Cyone and $[C_{10}C_1im][OTf]$ mixtures.

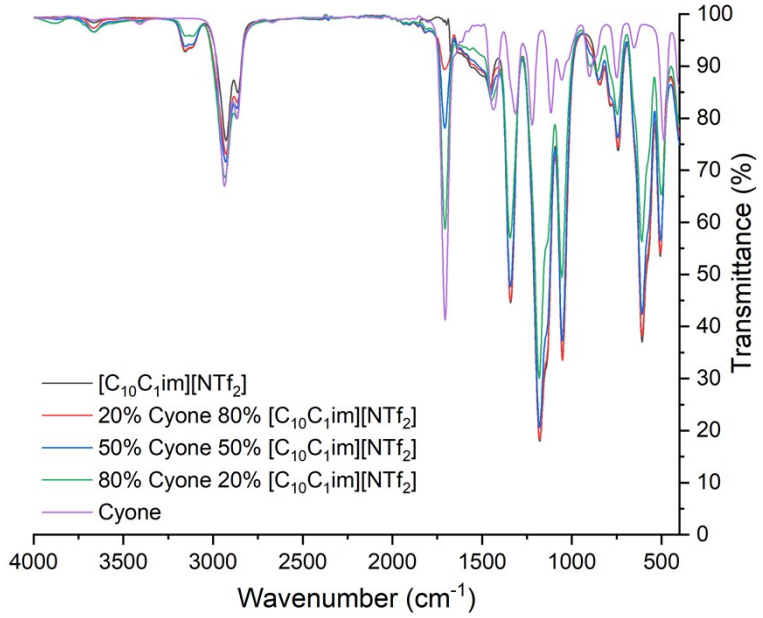

Figure S68. Complete IR spectrum for Cyone and $[C_{10}C_1im][NTf_2]$ mixtures.

Molecular Dynamics

Density Data

Table S2. Comparison of densities (in kg m^{-3}) obtained from molecular dynamics simulations with experimental values

Liquid	Simulated Density	Experimental Density	Deviation (%)
DMSO	1103.96 ± 0.04	1095.5	0.72
CyOAc	979.17 ± 0.05	966	1.36
CyOH	942.1 ± 0.3	948	-0.63
Cyone	943.64 ± 0.08	942.1	0.16
$[C_{10}C_1im][Me_2PO_4]$	1080 ± 1	1057.6	2.11
$[C_{10}C_1im][OTf]$	1169 ± 2	1150	1.60
$[C_{10}C_1im][NTf_2]$	1282.9 ± 0.9	1278.7	0.33

Radial Distribution Functions

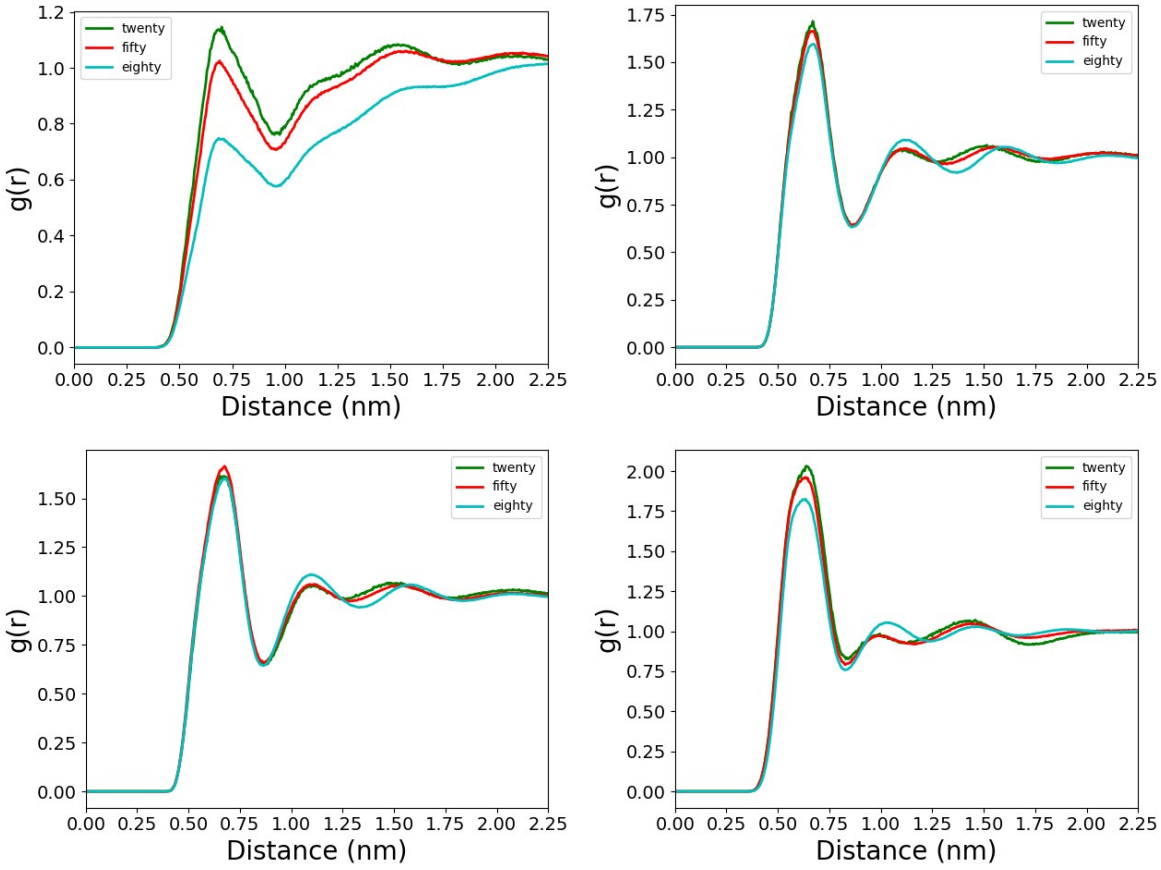

Figure S69. RDF of centre of mass of anion in $[C_{10}C_1im][NTf_2]$ to centre of mass of molecular solute for CyOAc, CyOH, Cyone and DMSO respectively (left to right from top) at 20 mol%, 50 mol% and 80 mol% of molecular solute.

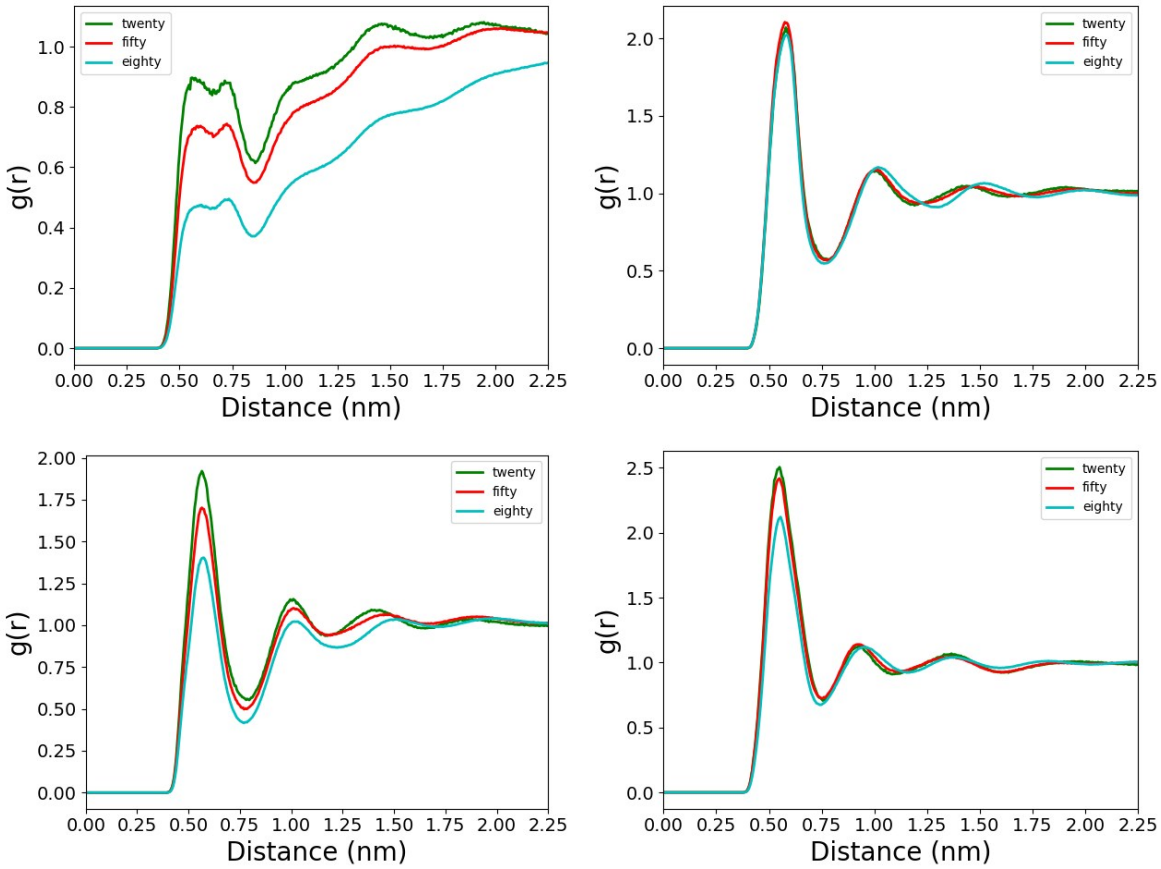

Figure S70. RDF of centre of mass of anion in [$C_{10}C_1$im][OTf] to centre of mass of solute for (left to right from top) CyOAc, CyOH, Cyone and DMSO respectively at 20 mol%, 50 mol% and 80 mol% of molecular solute.

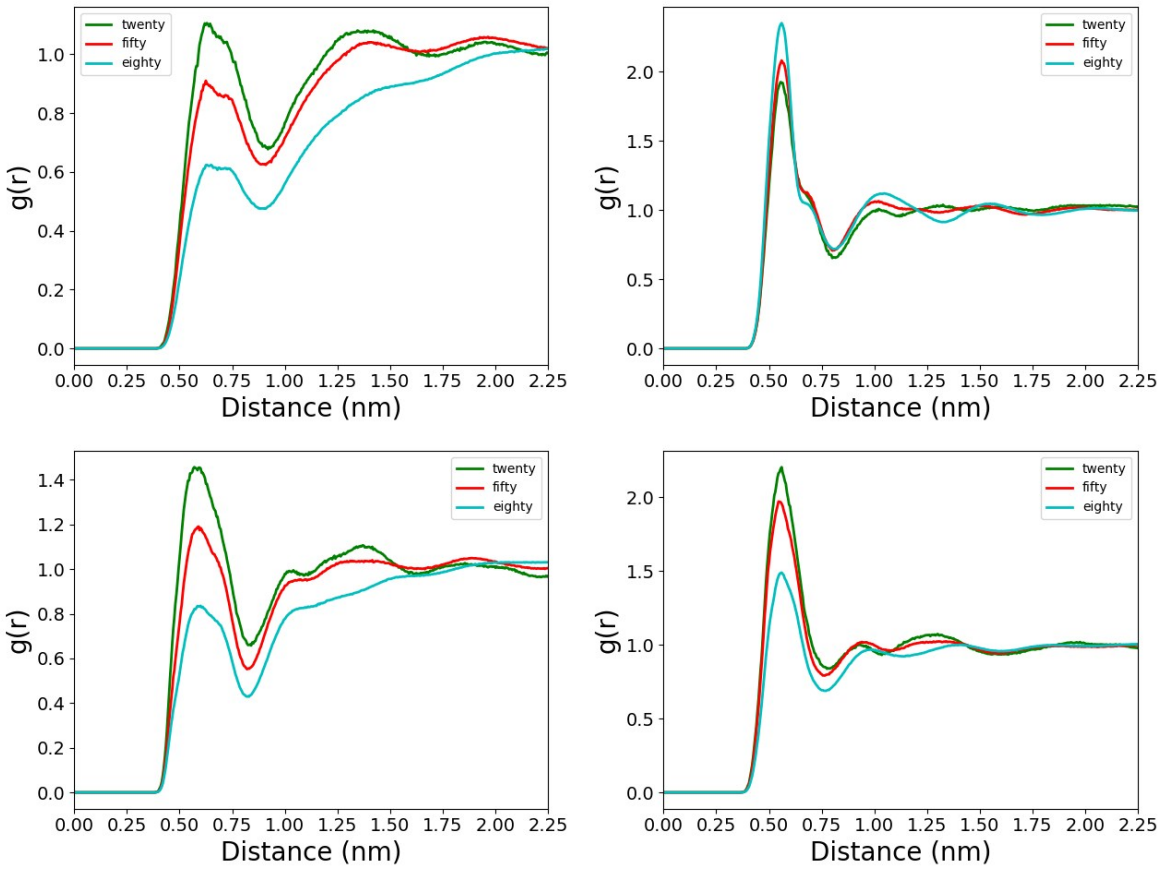

Figure S71. RDF of centre of mass of anion in $[C_{10}C_1im][Me_2PO_4]$ to centre of mass of solute for (left to right from top) CyOAc, CyOH, Cyone and DMSO respectively at 20 mol%, 50 mol% and 80 mol% of molecular solute.

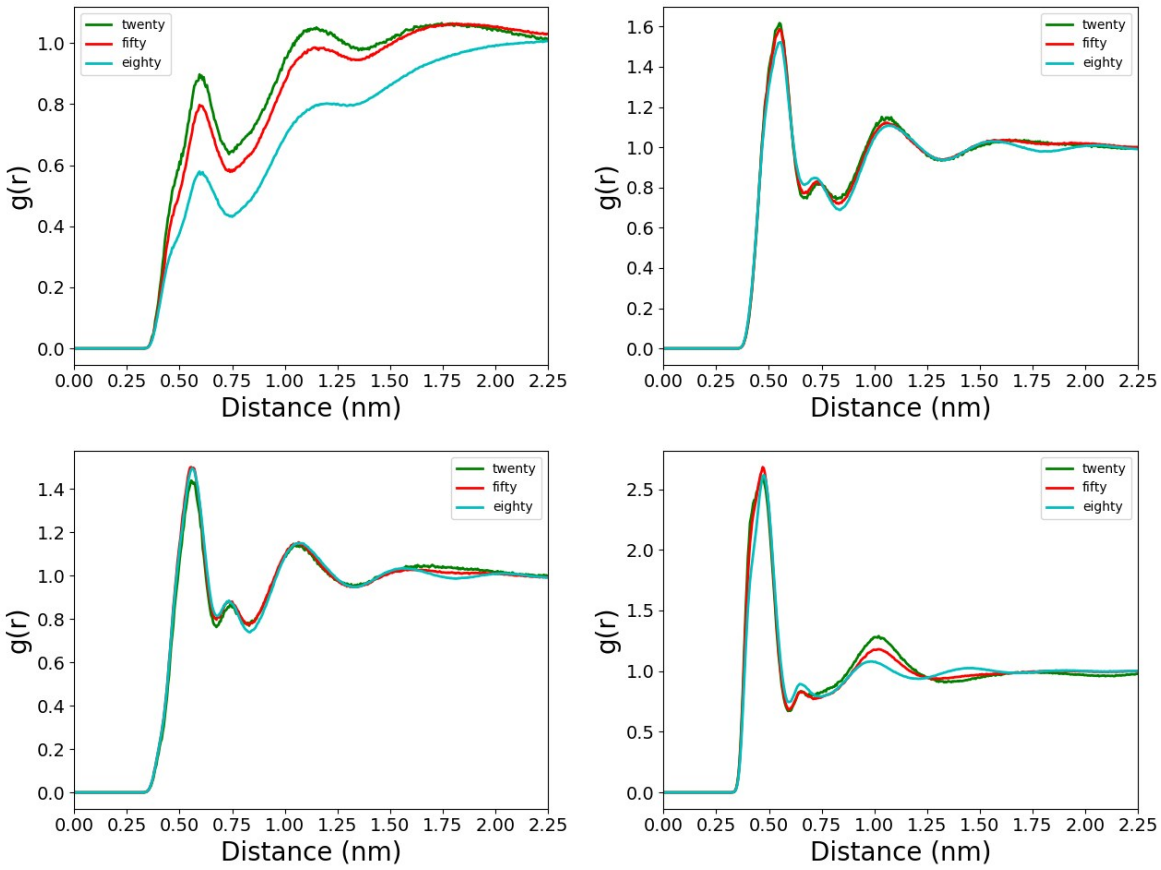

Figure S72. RDF of centre of mass of imidazolium ring in $[C_{10}C_1im][NTf_2]$ to centre of mass of molecular solute for (left to right from top) CyOAc, CyOH, Cyone and DMSO respectively at 20 mol%, 50 mol% and 80 mol% of molecular solute.

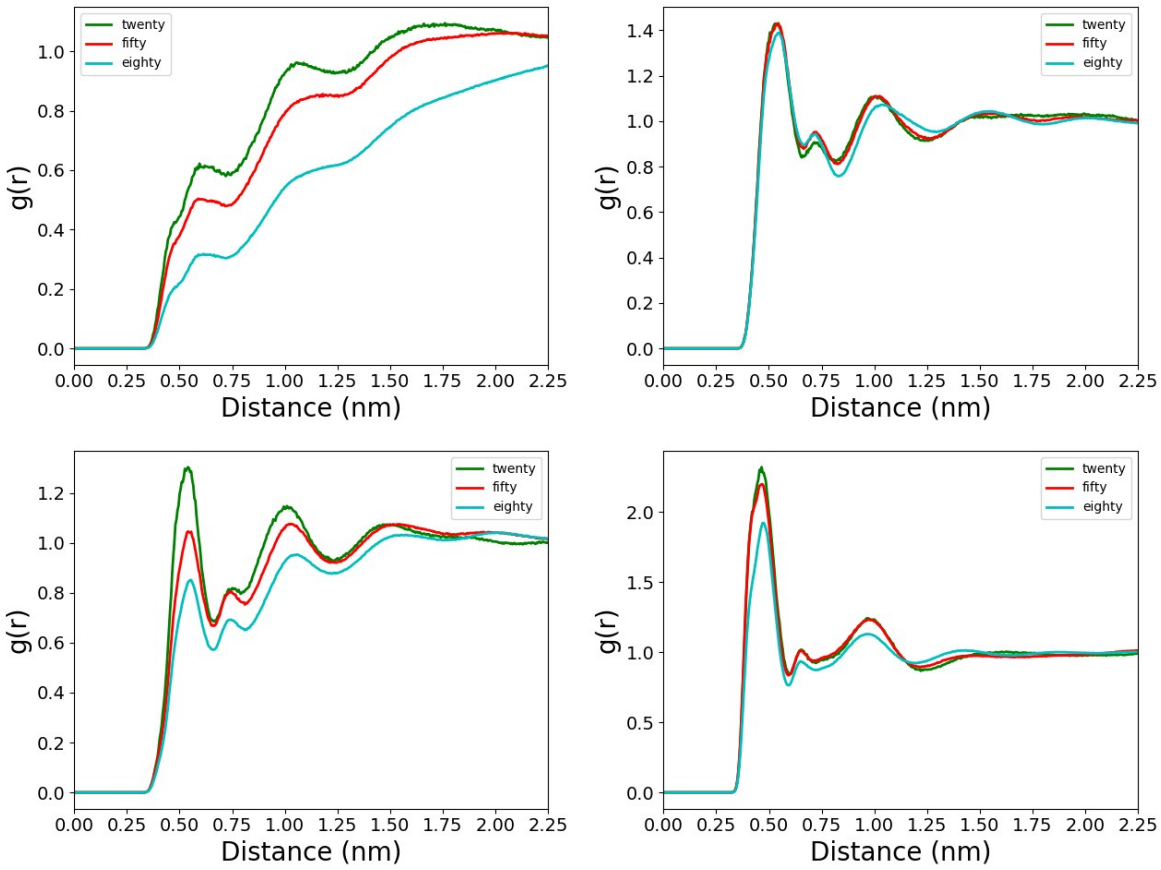

Figure S73. RDF of centre of mass of imidazolium ring in [$C_{10}C_1$im][OTf] to centre of mass of molecular solute for (left to right from top) CyOAc, CyOH, Cyone and DMSO respectively at 20 mol%, 50 mol% and 80 mol% of molecular solute.

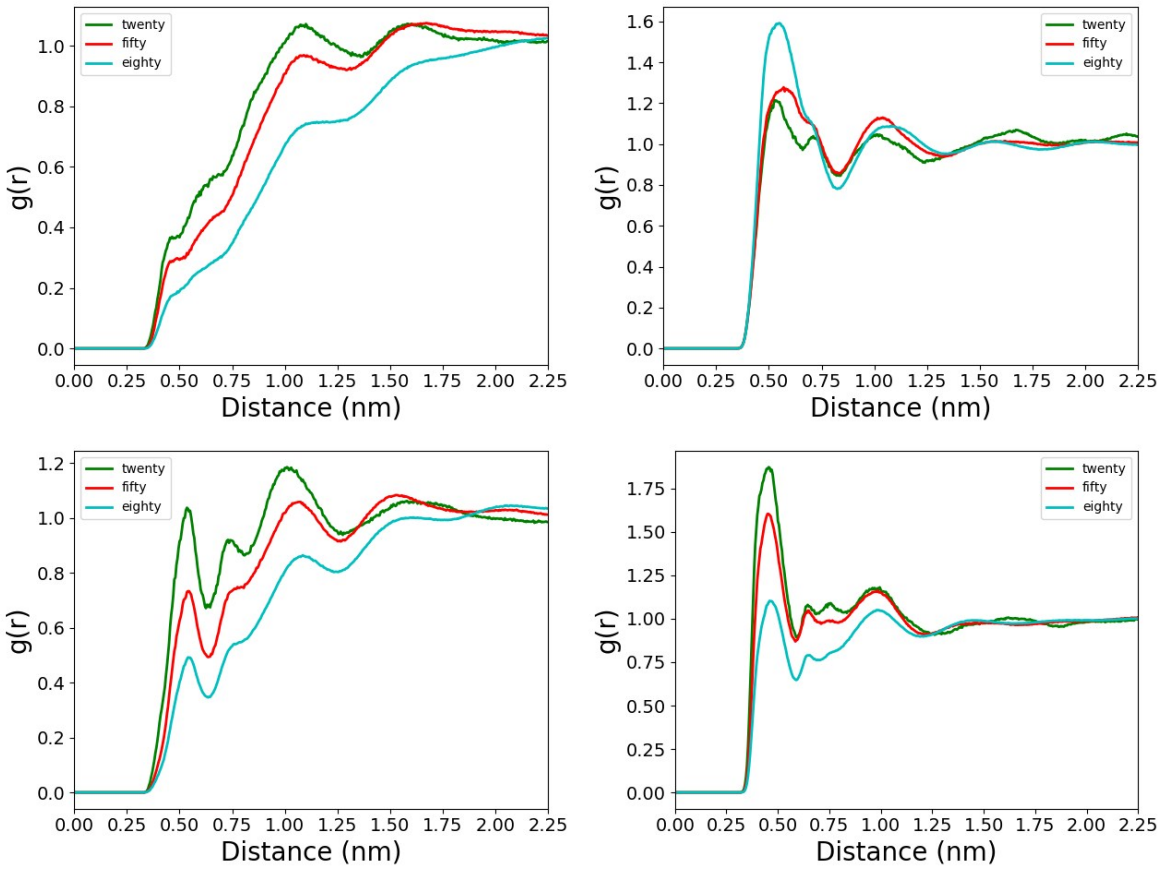

Figure S74. RDF of centre of mass of imidazolium ring in $[C_{10}C_1im][Me_2PO_4]$ to centre of mass of molecular solute for (left to right from top) CyOAc, CyOH, Cyone and DMSO respectively at 20 mol%, 50 mol% and 80 mol% of molecular solute.

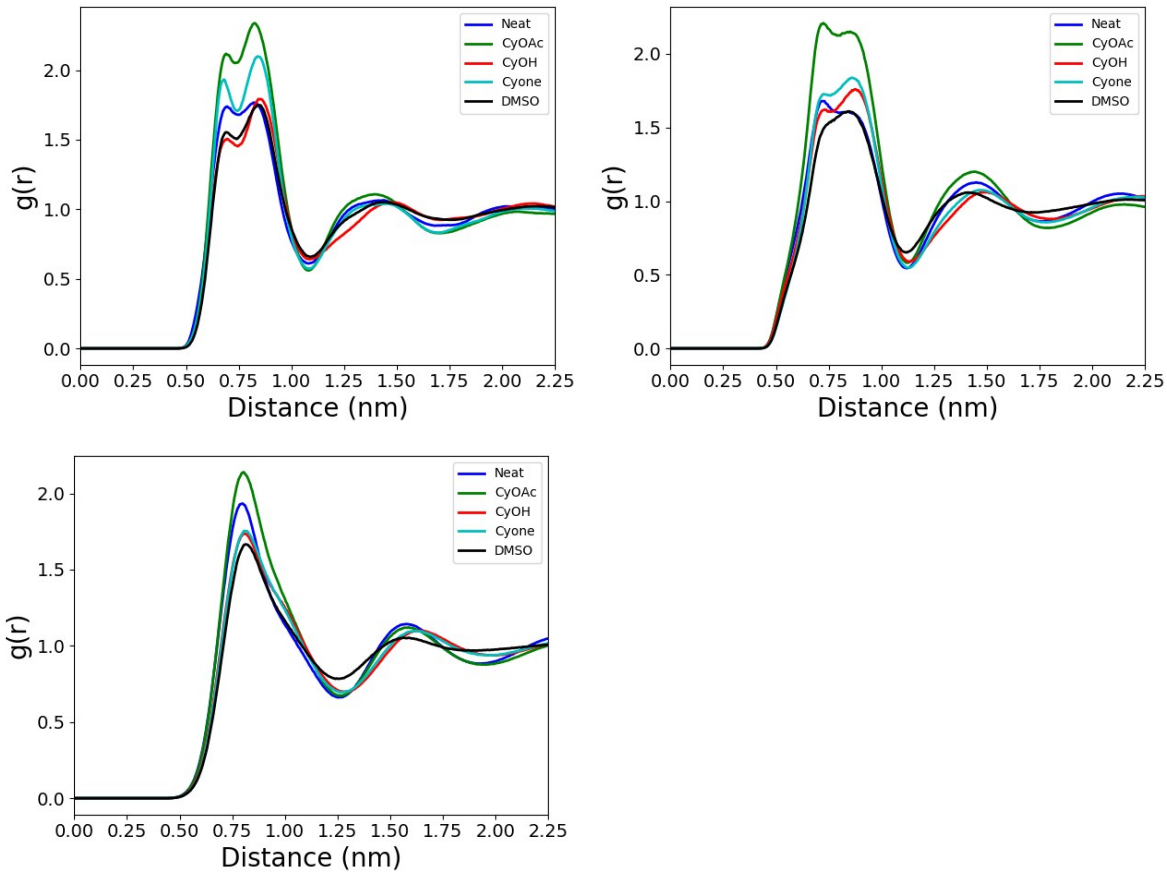

Figure S75. Anion-anion RDFs between the centre of mass of anions for (left to right from top): $[C_{10}C_1im][Me_2PO_4]$, $[C_{10}C_1im][OTf]$ and $[C_{10}C_1im][NTf_2]$ at 50 mol% of the molecular solute.

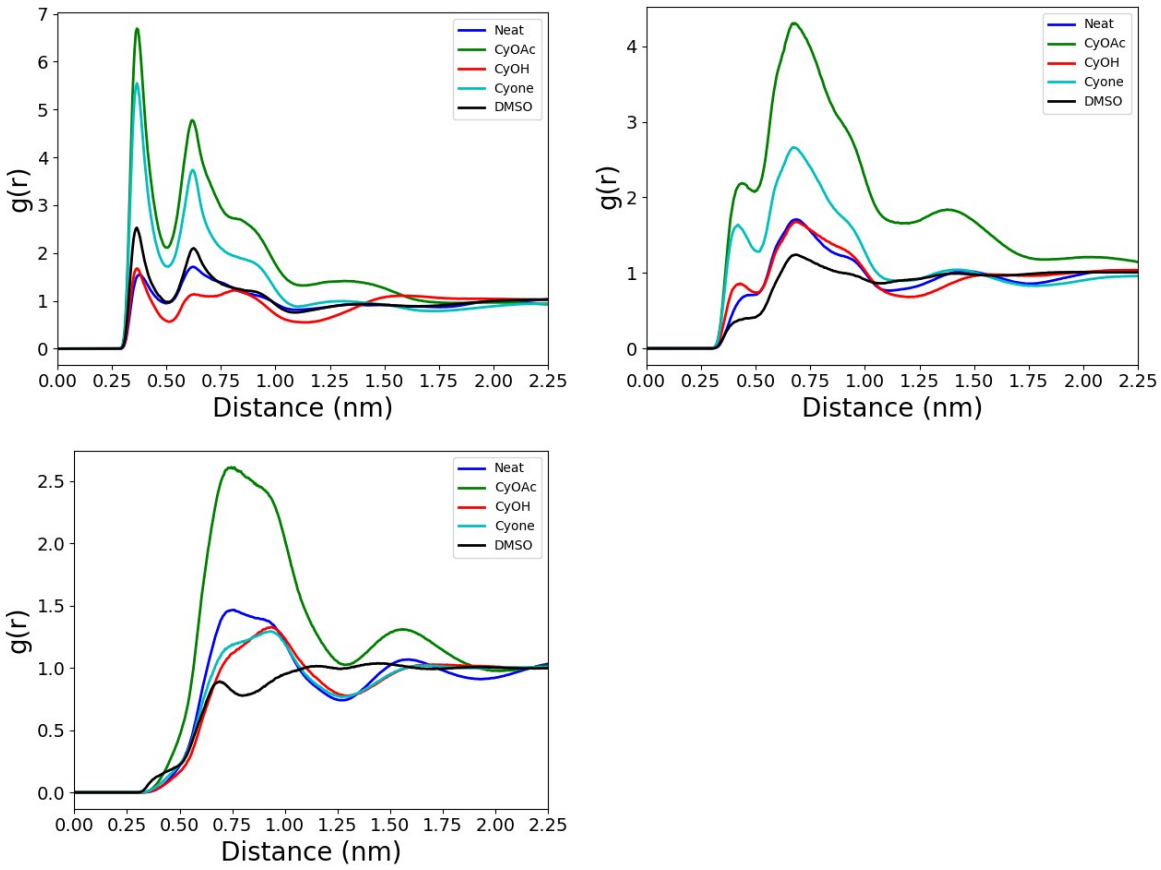

Figure S76. Cation-cation RDFs between the centre of mass of anions for (left to right from top): $[C_{10}C_1im][Me_2PO_4]$, $[C_{10}C_1im][OTf]$ and $[C_{10}C_1im][NTf_2]$ at 80 mol% of the molecular solute.

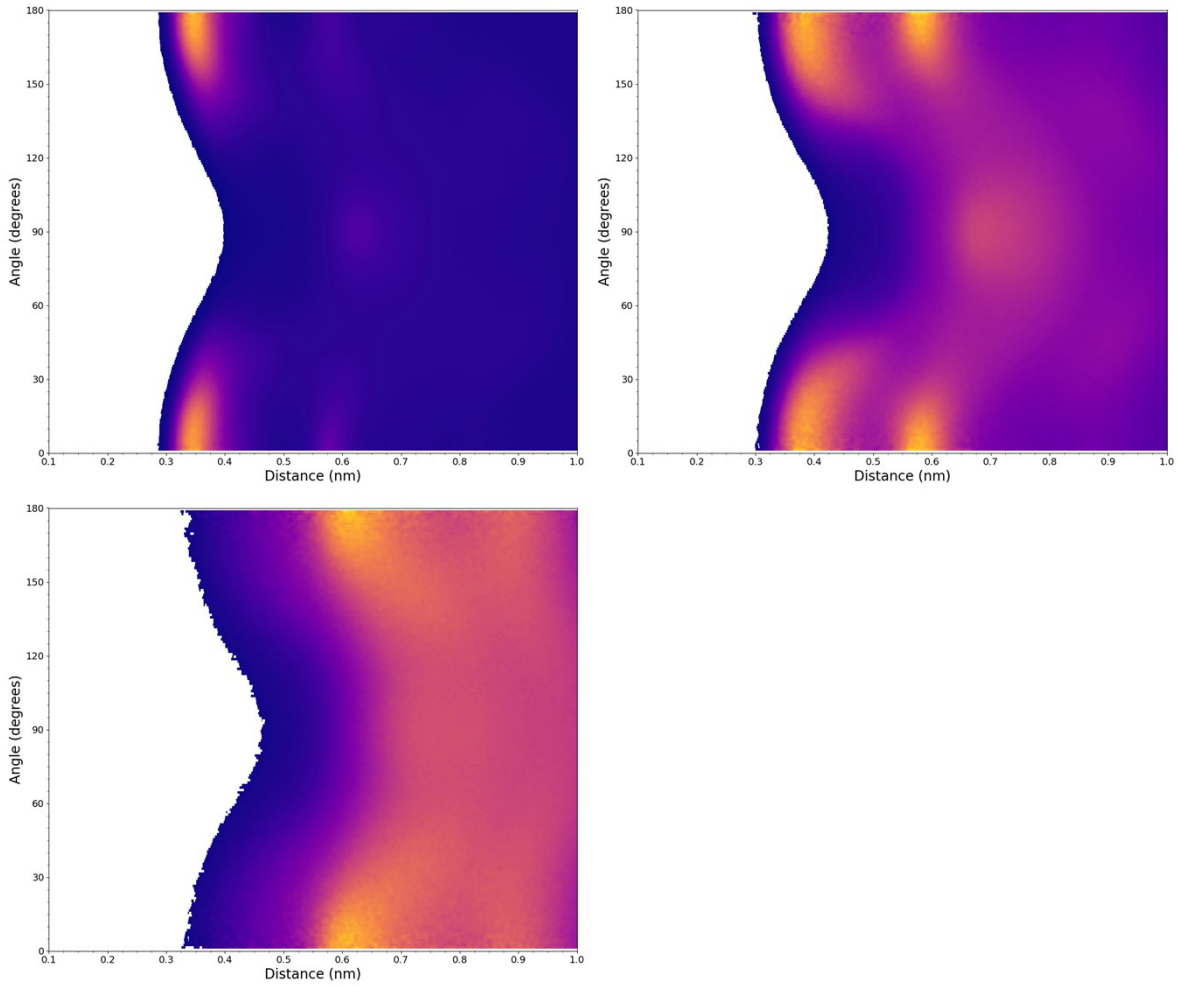

Figure S77. CDFs displaying the distance between centre of mass for imidazolium rings on the x axis with the angle between the normals of these rings as the y axis for 80 mol% CyOAc-IL mixtures for (left to right from top): $[C_{10}C_1im][Me_2PO_4]$, $[C_{10}C_1im][OTf]$ and $[C_{10}C_1im][NTf_2]$.

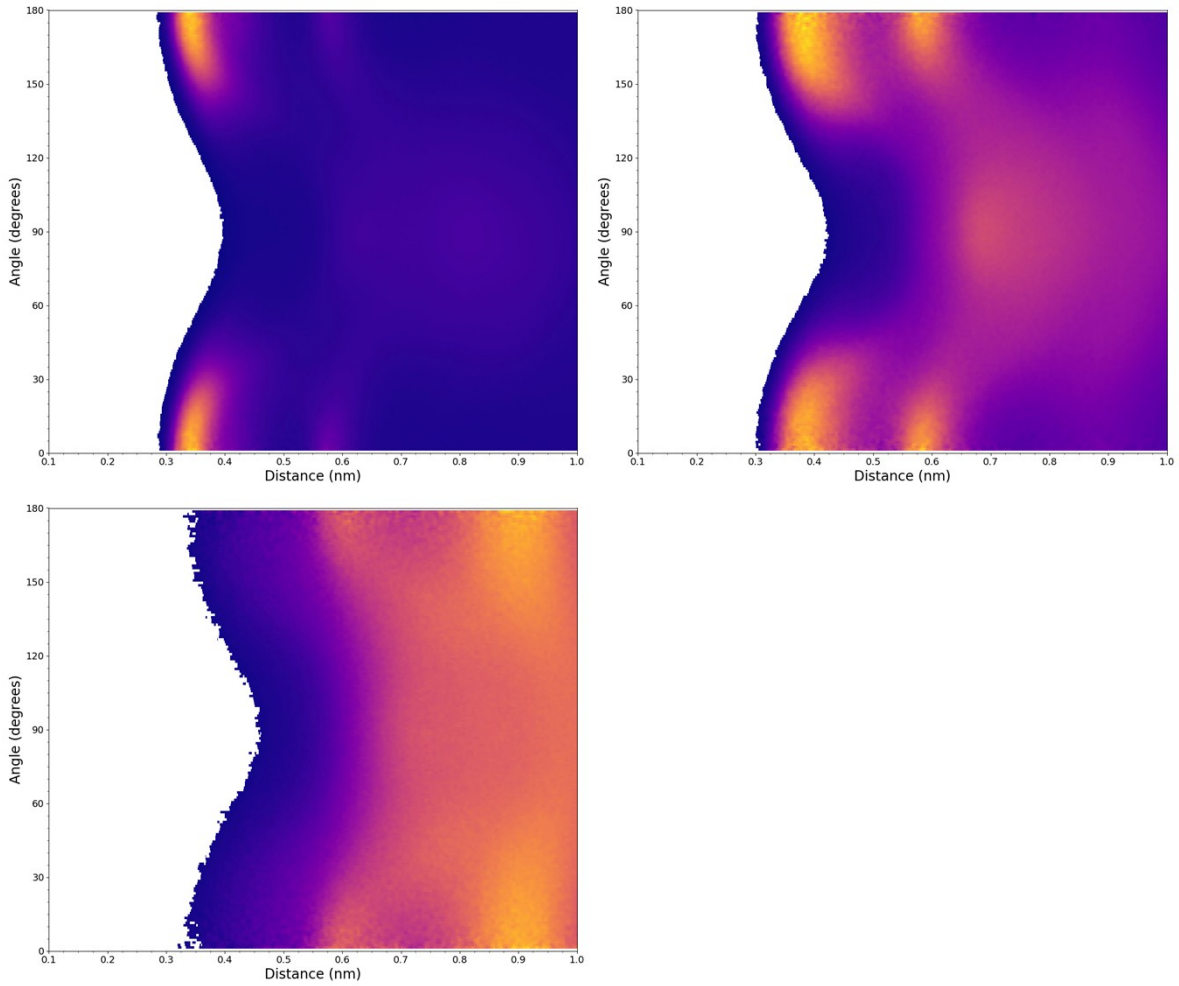

Figure S78. CDFs displaying the distance between centre of mass for imidazolium rings on the x axis with the angle between the normals of these rings as the y axis for 80 mol% CyOH-IL mixtures for (left to right from top): $[C_{10}C_1im][Me_2PO_4]$, $[C_{10}C_1im][OTf]$ and $[C_{10}C_1im][NTf_2]$.

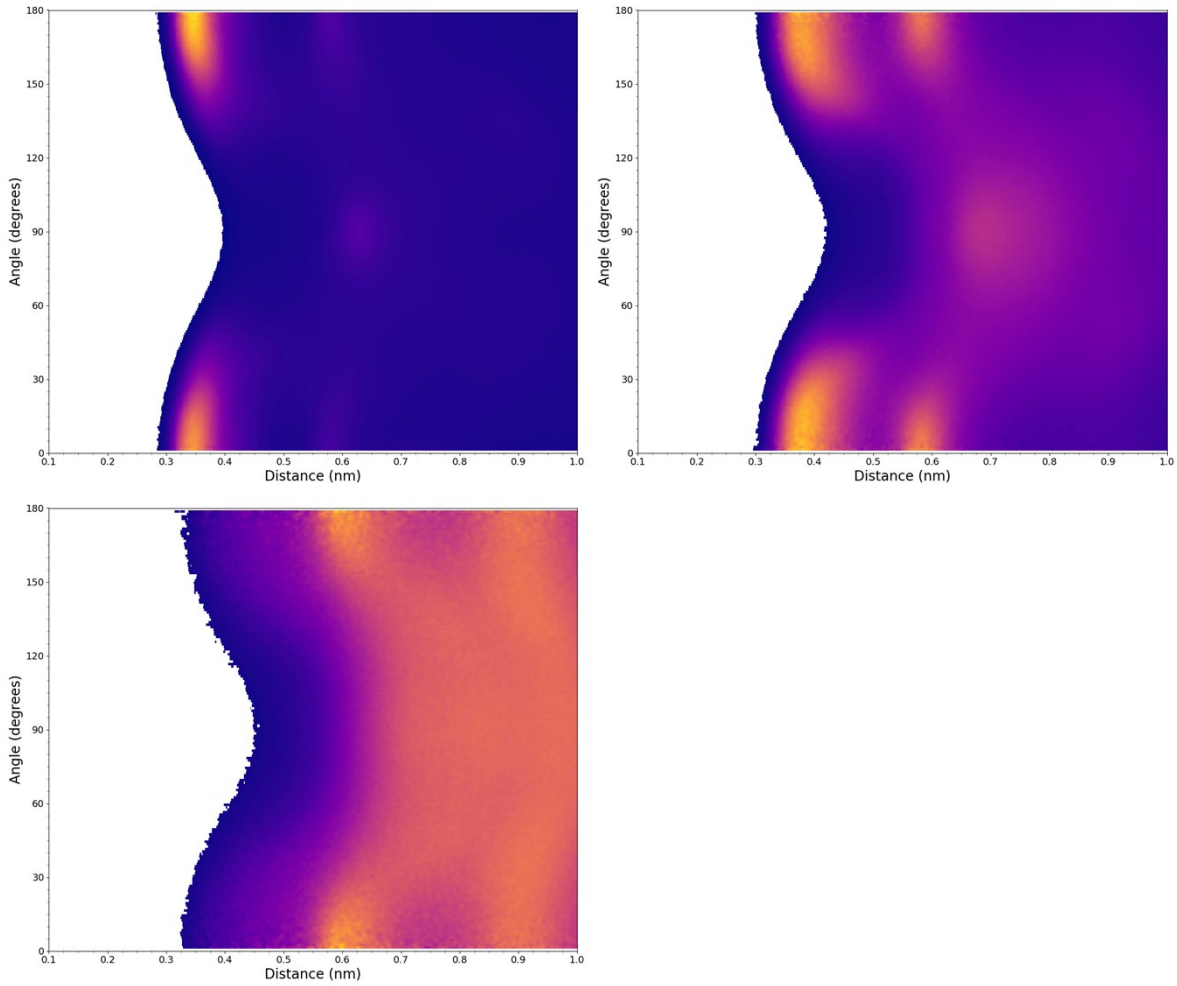

Figure S79. CDFs displaying the distance between centre of mass for imidazolium rings on the x axis with the angle between the normals of these rings as the y axis for 80 mol% Cyone-IL mixtures for (left to right from top): $[C_{10}C_1im][Me_2PO_4]$, $[C_{10}C_1im][OTf]$ and $[C_{10}C_1im][NTf_2]$.

Convergence of Simulations

To assess whether the simulations converged after the 10 ns of equilibration time and 100 ns production runs we examined the outputs of cation-anion RDFs for the triplicate experiments across all solute-IL mixtures. The shape and relative order of these curves are consistent across all 3 replicates for each of the 12 mixtures. Some minor variations in the quantitative detail can be seen, for example in Fig. S73, however the relative trends and details remain the same across all 3 simulations. As these replicates were simulated from randomised initial conditions this implies convergence of these simulations to the level required for the qualitative analysis we have performed.

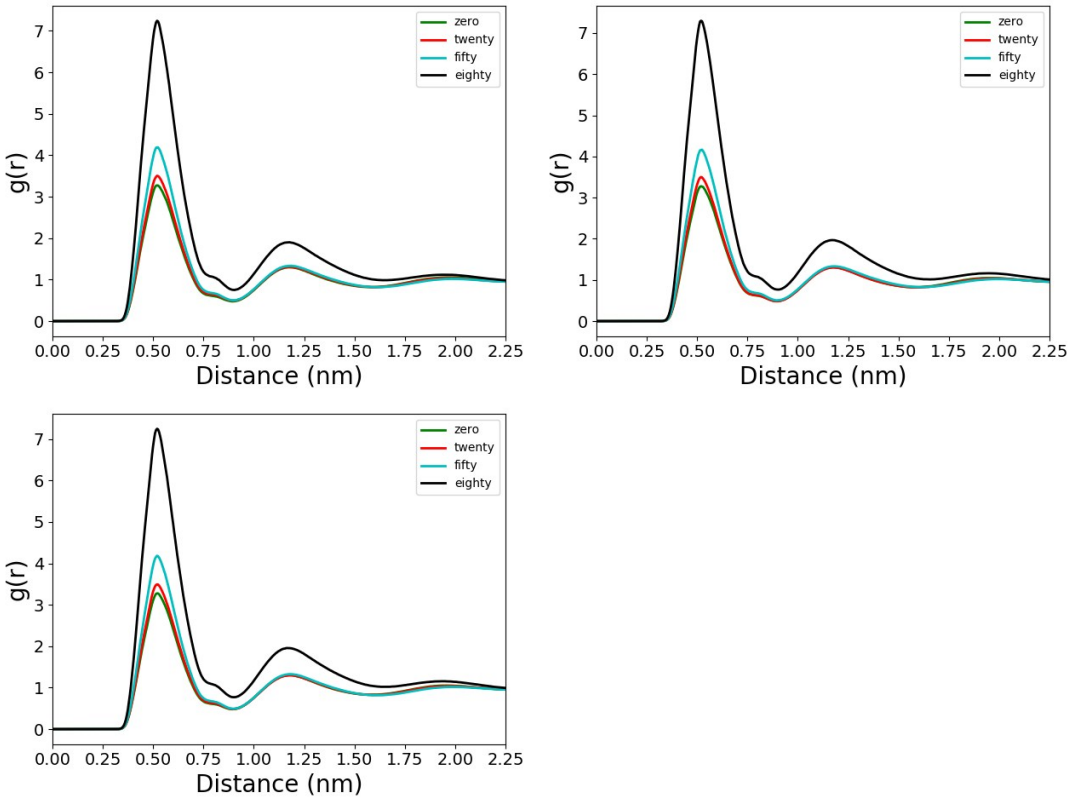

Figure S80. RDF of centre of mass of imidazolium ring to centre of mass of anion in CyOAc-[$C_{10}C_1$im][NTf_2] mixtures for each of the triplicate simulations.

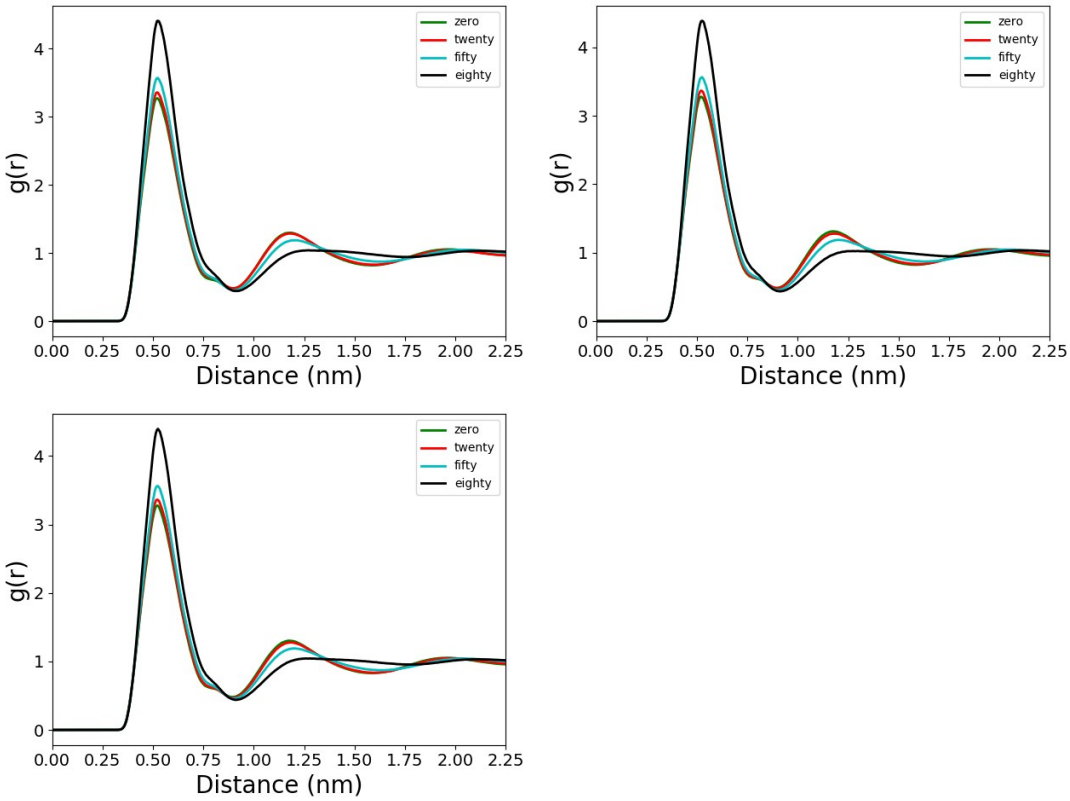

Figure S81. RDF of centre of mass of imidazolium ring to centre of mass of anion in CyOH-[$C_{10}C_1$im][NTf_2] mixtures for each of the triplicate simulations.

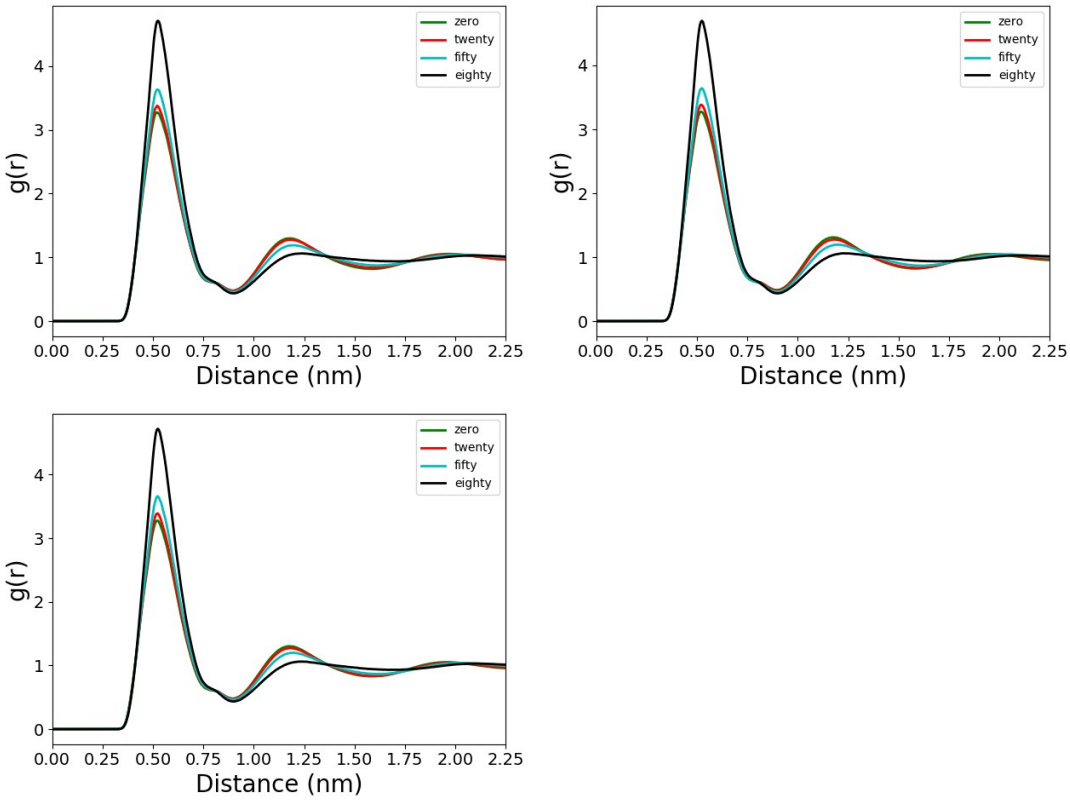

Figure S82. RDF of centre of mass of imidazolium ring to centre of mass of anion in Cyone-$[C_{10}C_1im][NTf_2]$ mixtures for each of the triplicate simulations.

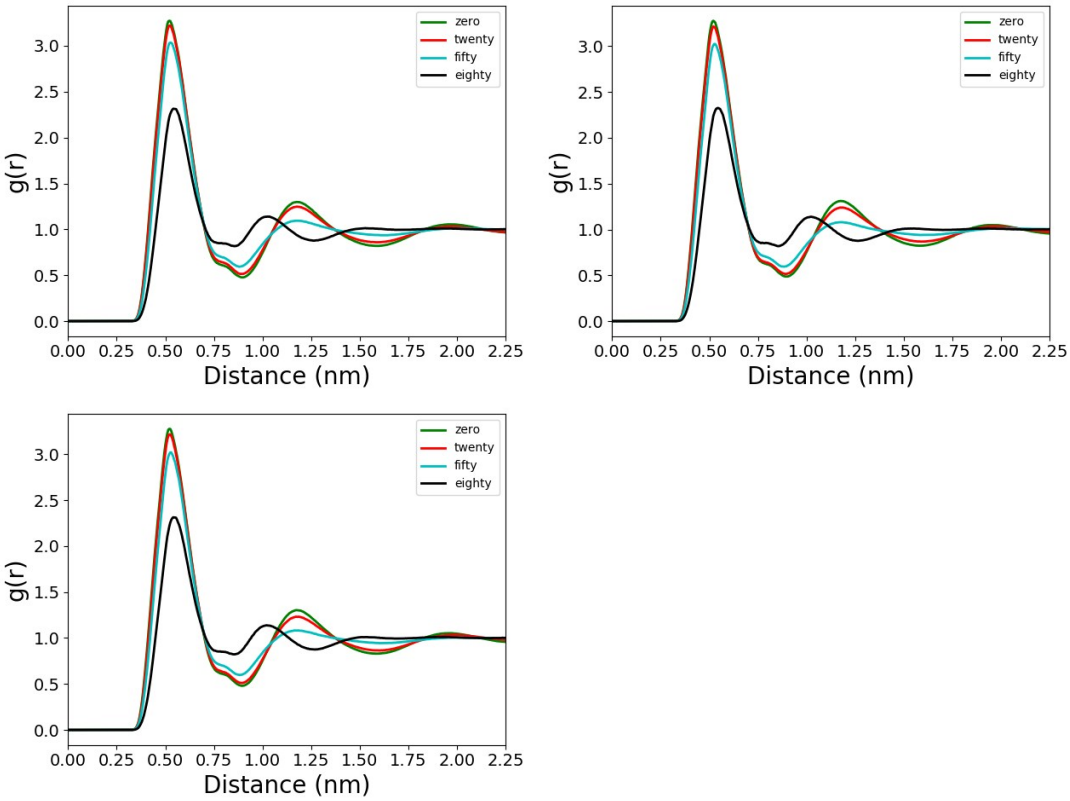

Figure S83. RDF of centre of mass of imidazolium ring to centre of mass of anion in DMSO-$[C_{10}C_1im][NTf_2]$ mixtures for each of the triplicate simulations.

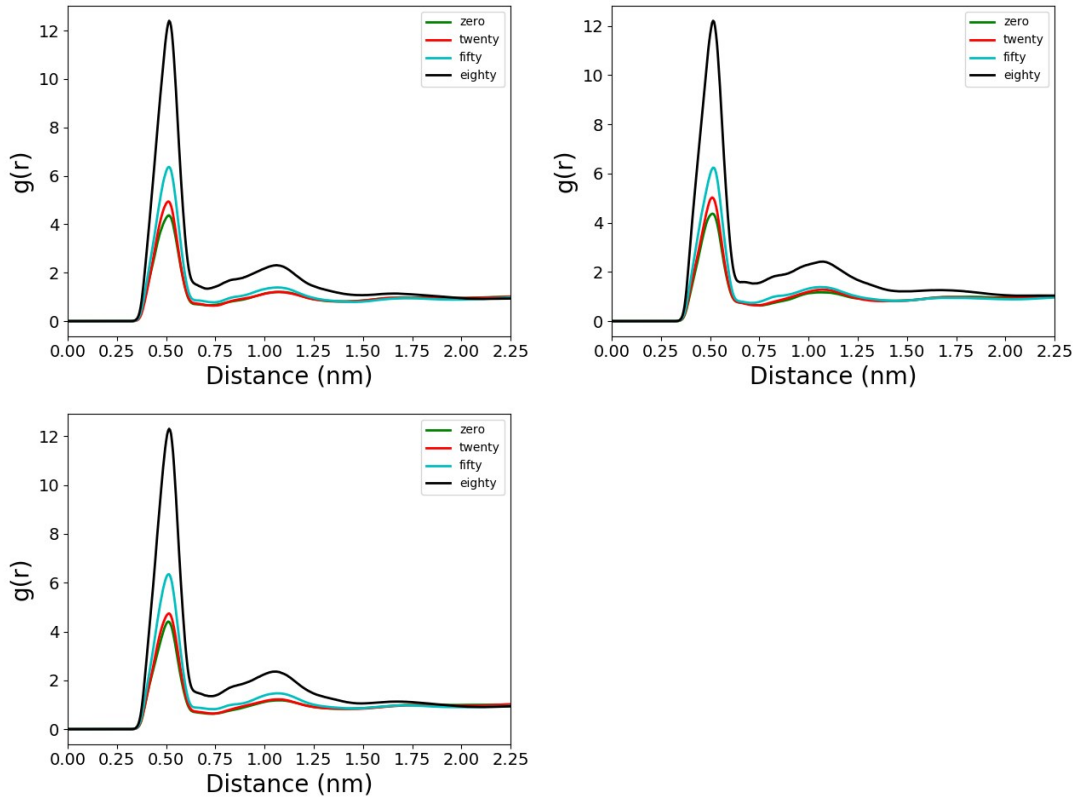

Figure S84. RDF of centre of mass of imidazolium ring to centre of mass of anion in CyOAc-$[C_{10}C_1im][Me_2PO_4]$ mixtures for each of the triplicate simulations.

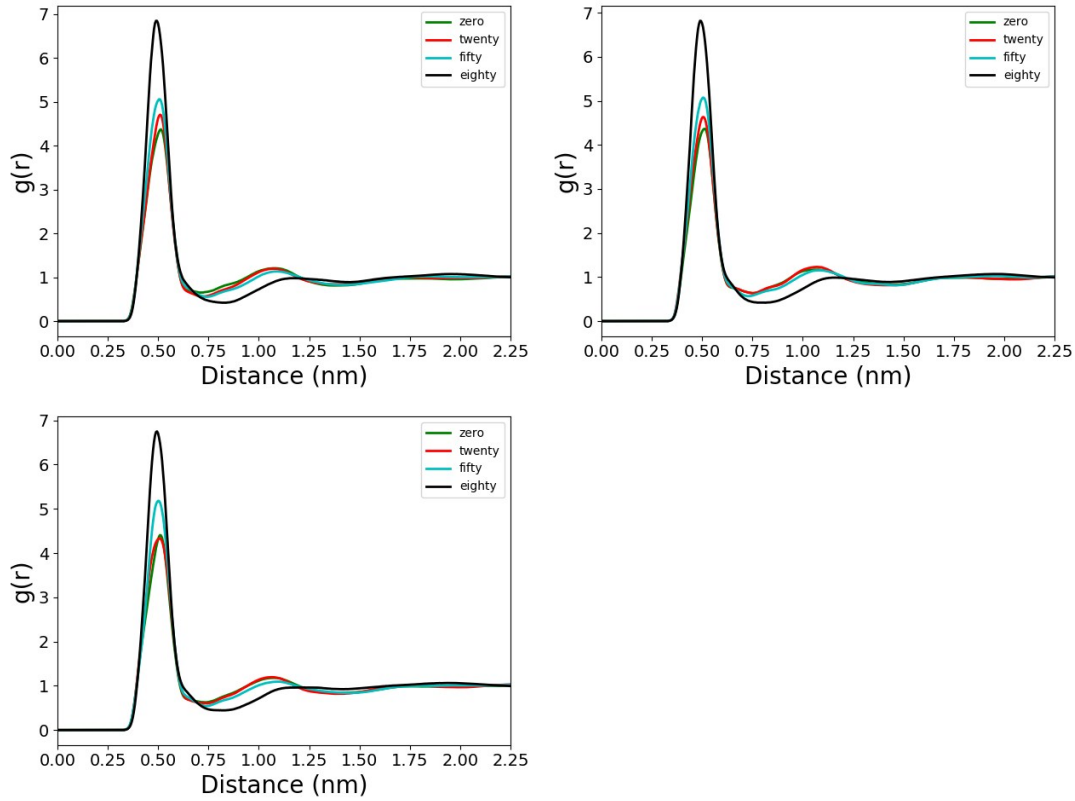

Figure S85. RDF of centre of mass of imidazolium ring to centre of mass of anion in CyOH-$[C_{10}C_1im][Me_2PO_4]$ mixtures for each of the triplicate simulations.

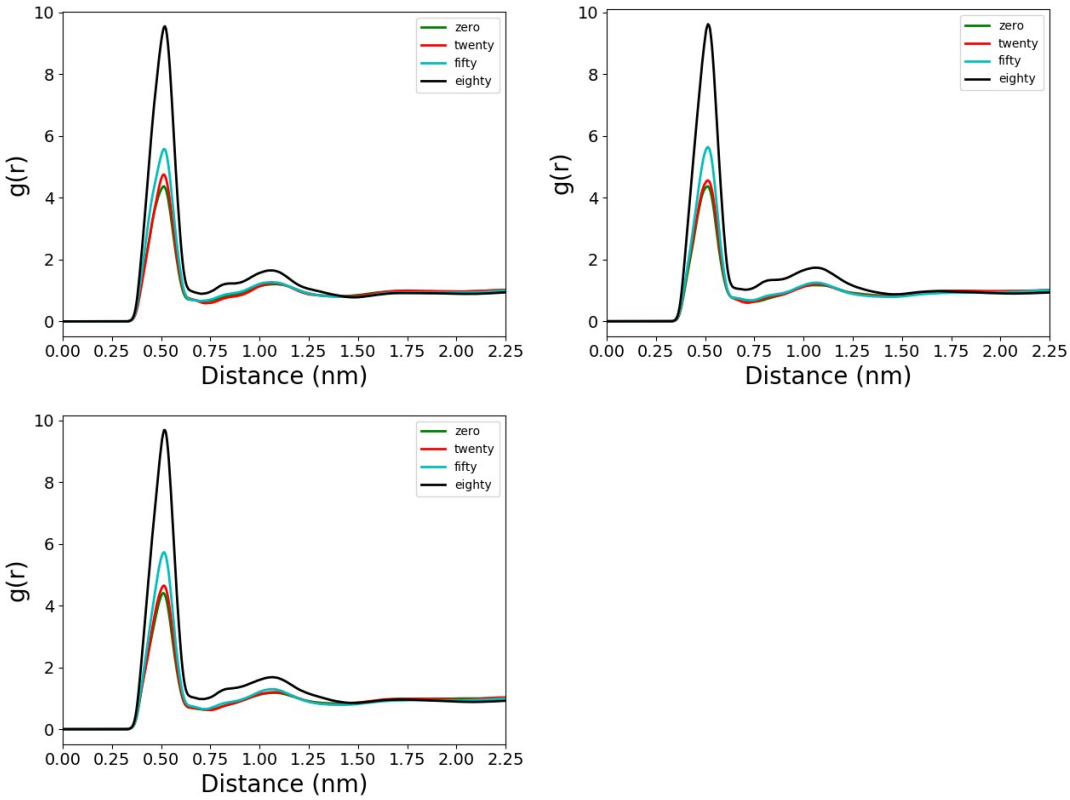

Figure S86. RDF of centre of mass of imidazolium ring to centre of mass of anion in Cyone-$[C_{10}C_1im][Me_2PO_4]$ mixtures for each of the triplicate simulations.

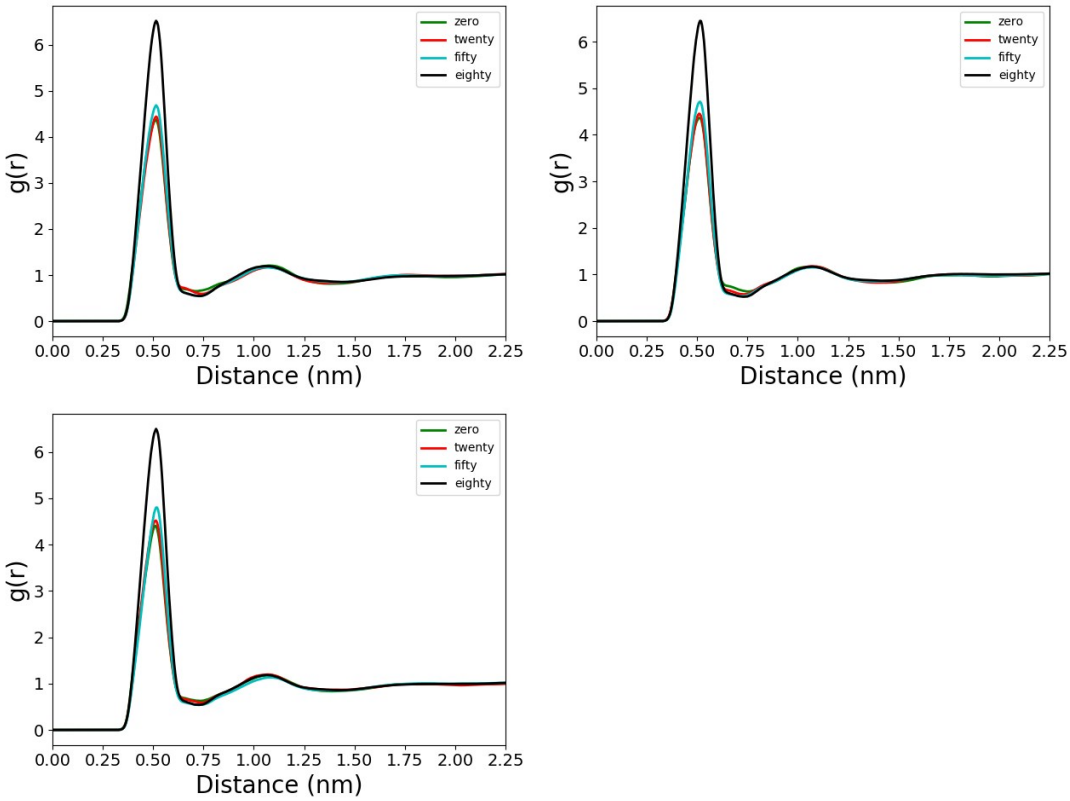

Figure S87. RDF of centre of mass of imidazolium ring to centre of mass of anion in DMSO-$[C_{10}C_1im][Me_2PO_4]$ mixtures for each of the triplicate simulations.

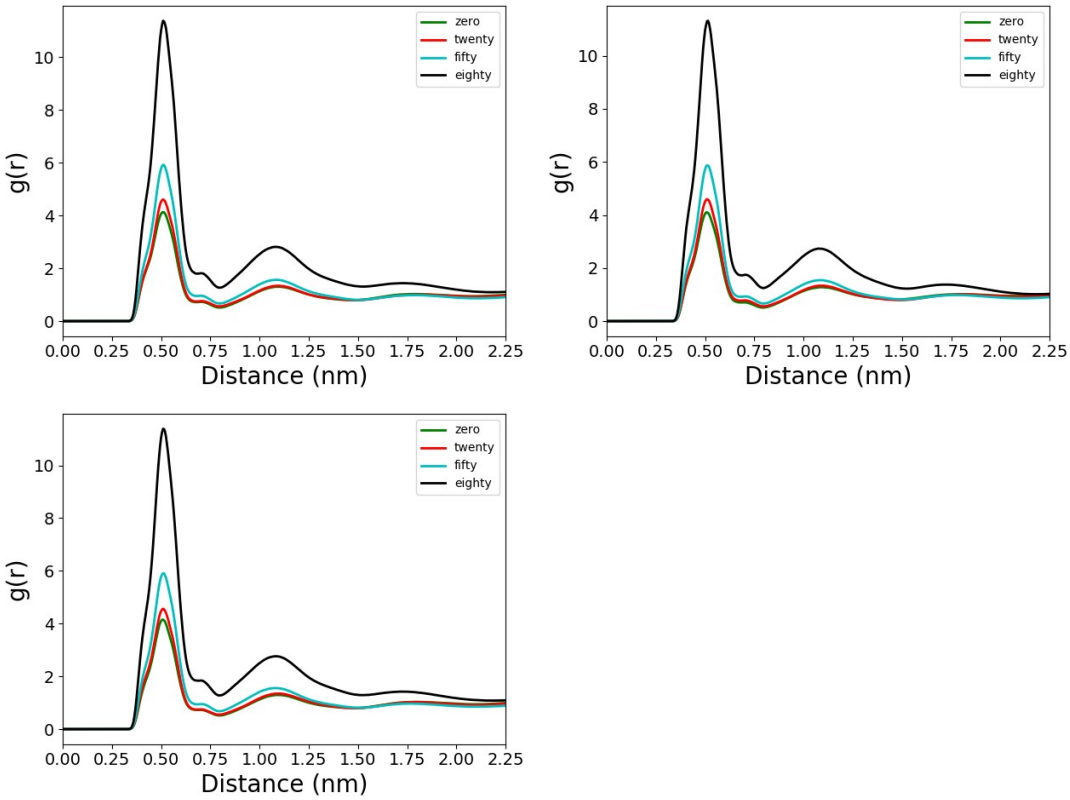

Figure S88. RDF of centre of mass of imidazolium ring to centre of mass of anion in CyOAc-[$C_{10}C_1$im][OTf] mixtures for each of the triplicate simulations.

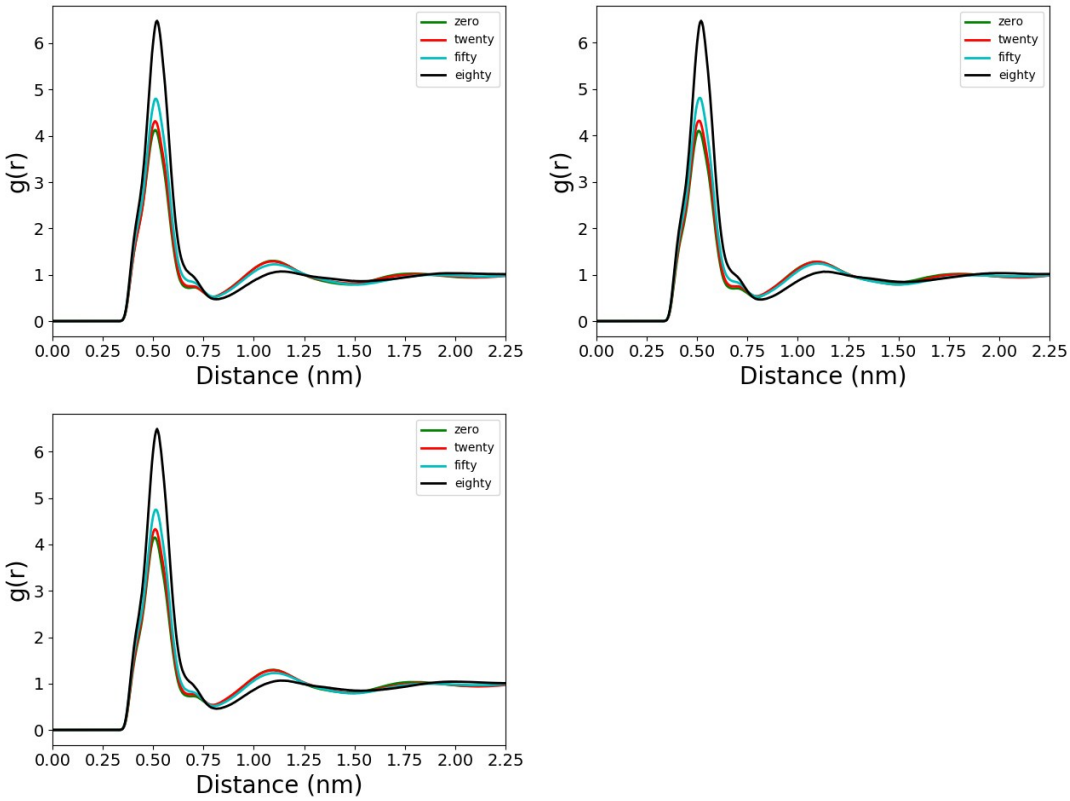

Figure S89. RDF of centre of mass of imidazolium ring to centre of mass of anion in CyOH-[$C_{10}C_1$im][OTf] mixtures for each of the triplicate simulations.

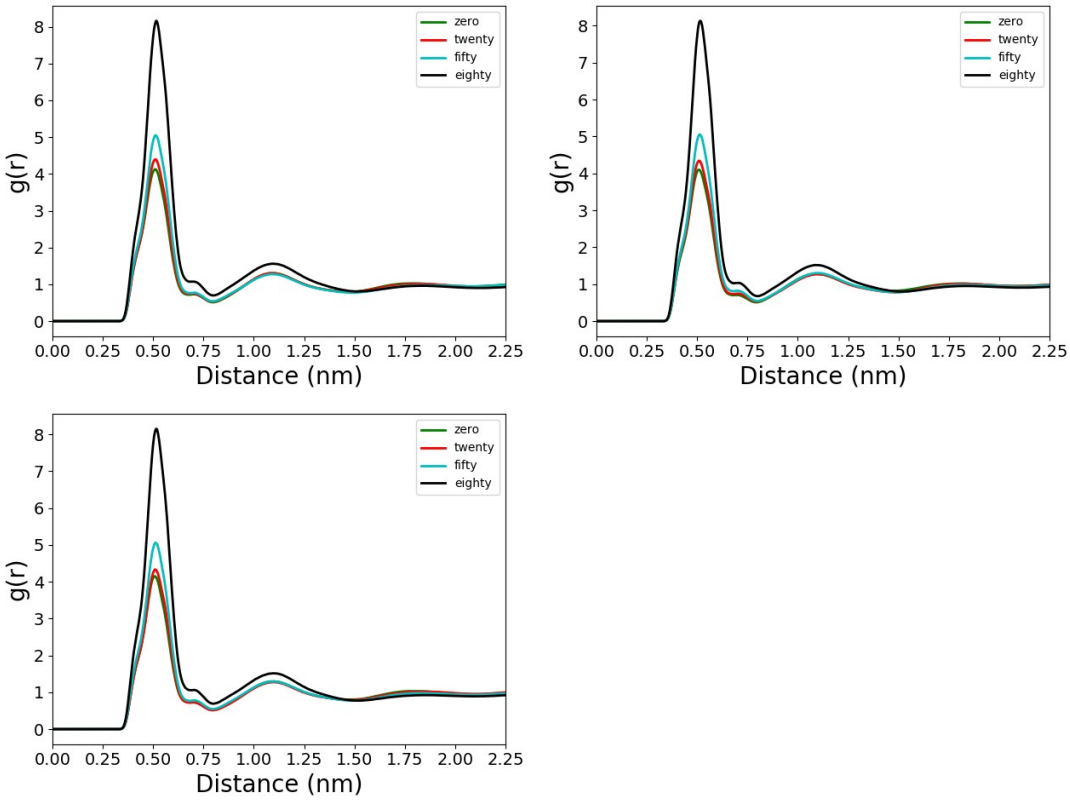

Figure S90. RDF of centre of mass of imidazolium ring to centre of mass of anion in Cyone-[$C_{10}C_1$im][OTf] mixtures for each of the triplicate simulations.

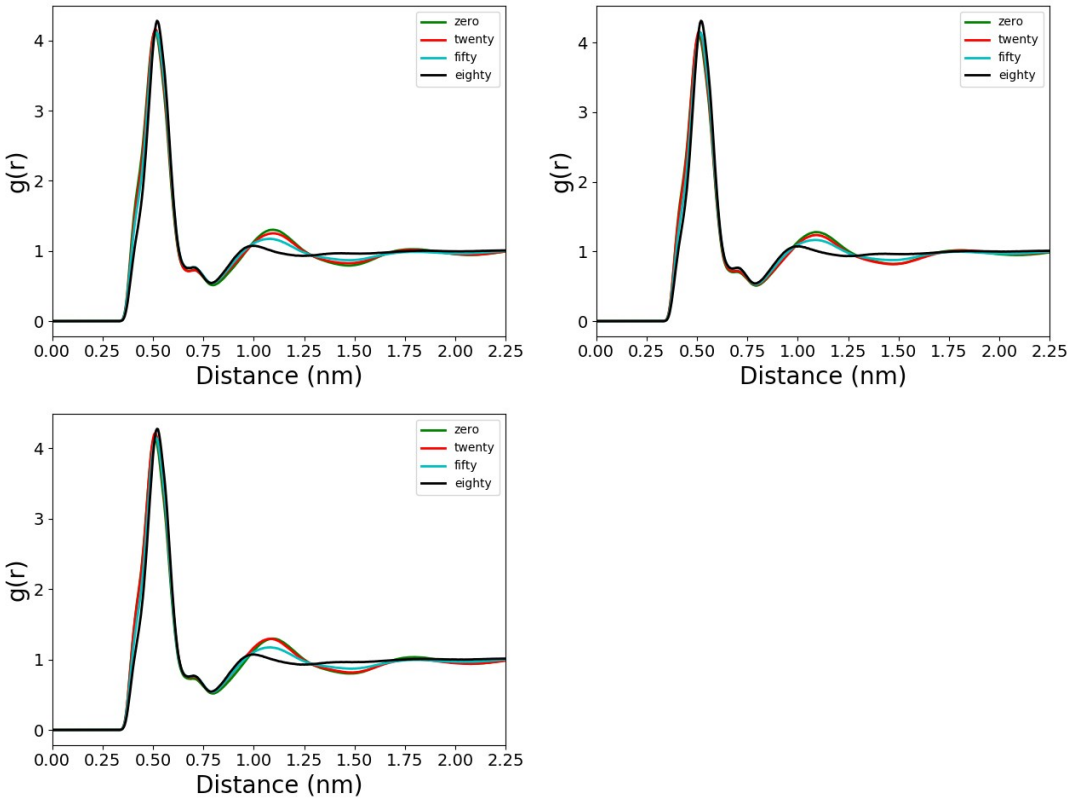

Figure S91. RDF of centre of mass of imidazolium ring to centre of mass of anion in DMSO-[$C_{10}C_1$im][OTf] mixtures for each of the triplicate simulations.

GROMACS Topology Parameters

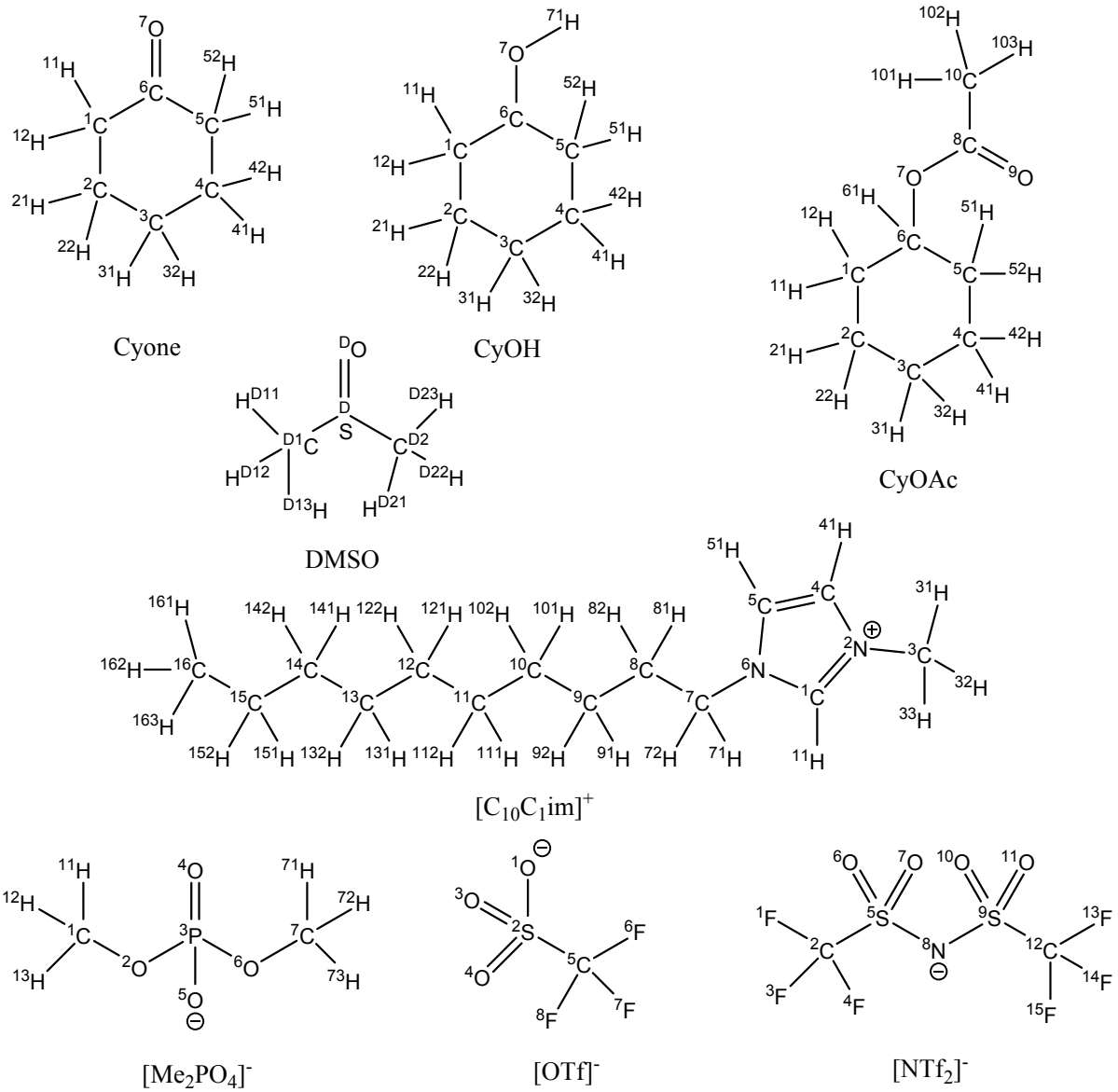

Figure S92. Atom numbering as used for the MD simulations.

Added to atomtypes.atp

```
kpl_001      12.01100
kpl_002      12.01100
kpl_003      12.01100
kpl_004      12.01100
kpl_005      12.01100
kpl_006      12.01100
kpl_007       1.00800
kpl_008       1.00800
kpl_009       1.00800
kpl_010       1.00800
kpl_011      14.00670
kpl_012      12.01100
kpl_013      18.99840
```

```
kpl_014      32.06000
kpl_015      15.99940
kpl_016      15.99940
kpl_017      14.00670


;Additional nonbonded parameters

;Methylimidazolium atom types

kpl_001    KPLC1    6   12.01100    -0.170      A       3.5e-01
2.7614e-01
kpl_002    KPLC2    6   12.01100     0.010      A       3.5e-01
2.7614e-01
kpl_003    KPLCR    6   12.01100    -0.110      A       2.13e-01
4.3932e-01
kpl_004    KPLCS    6   12.01100    -0.120      A       3.5e-01
2.7614e-01
kpl_005    KPLCT    6   12.01100    -0.180      A       3.5e-01
2.7614e-01
kpl_006    KPLCW    6   12.01100    -0.130      A       3.0175e-01
2.0501e-01
kpl_007    KPLHA    1    1.00800     0.210      A       1.452e-01
1.8828e-01
kpl_008    KPLHB    1    1.00800     0.210      A       2.057e-01
8.79e-02
kpl_009    KPLHC    1    1.00800     0.060      A       2.5e-01
1.2552e-01
kpl_010    KPLH1    1    1.00800     0.130      A       2.5e-01
1.2552e-01
kpl_011    KPLNA    7   14.00670     0.150      A       3.25e-01
7.1128e-01
; combined triflate, bistriflyimide atom types
kpl_012    KPL_C    6   12.01100     0.494      A       3.150e-01
8.282e-02
kpl_013    KPL_F    9   18.99840    -0.189      A       2.655e-01
6.651e-02
kpl_014    KPL_S    16  32.06000     1.076      A       4.0825e-01
3.1372e-01
; triflate only atom types
kpl_015    TF_O     8   15.99940    -0.668      A       3.4632e-01
2.6353e-01
; bistriflyimide only atom types
kpl_016    NTF2_O   8   15.99940    -0.579      A       3.4632e-01
2.6353e-01
kpl_017    NTF2_N   7   14.00670    -0.690      A       3.25e-01
2.1333e-01


; Addtional bonded parameters
[ bondtypes ]
; imidazolium bond types
KPLCR   KPLHA   1   0.10800     284512.0
KPLCW   KPLHB   1   0.10800     284512.0
KPLC1   KPLH1   1   0.10900     284512.0
KPLC2   KPLHC   1   0.10900     284512.0
```

```
KPLCS   KPLHC   1   0.10900     284512.0
KPLCT   KPLHC   1   0.10900     284512.0
KPLCR   KPLNA   1   0.13150     199600.0
KPLCW   KPLNA   1   0.13780     178700.0
KPLCW   KPLCW   1   0.13410     217600.0
KPLNA   KPLC1   1   0.14660     141000.0
KPLC1   KPLC2   1   0.15290     112100.0
KPLC2   KPLCS   1   0.15290     112100.0
KPLCS   KPLCS   1   0.15290     112100.0
KPLCS   KPLCT   1   0.15290     112100.0
;triflate/bistriflate bond types
KPL_C   KPL_F   1   0.13230     369700.0
KPL_C   KPL_S   1   0.18180     197000.0
KPL_S   TF_O    1   0.14420     533100.0
KPL_S   NTF2_O  1   0.14420     533100.0
KPL_S   NTF2_N  1   0.15700     311300.0

[ angletypes ]
; imidazolium angle types
KPLCW   KPLNA   KPLCR   1   108.0   292.6
KPLCW   KPLNA   KPLC1   1   125.6   292.6
KPLCR   KPLNA   KPLC1   1   126.4   292.6
KPLNA   KPLCR   KPLHA   1   125.1   146.3
KPLNA   KPLCR   KPLNA   1   109.8   292.6
KPLNA   KPLCW   KPLCW   1   107.1   292.6
KPLNA   KPLCW   KPLHB   1   122.0   146.3
KPLCW   KPLCW   KPLHB   1   130.9   146.3
KPLNA   KPLC1   KPLH1   1   110.7   156.6
KPLC2   KPLC1   KPLH1   1   110.7   156.6
KPLC1   KPLC2   KPLHC   1   110.7   156.6
KPLCS   KPLC2   KPLHC   1   110.7   156.6
KPLC2   KPLCS   KPLHC   1   110.7   156.6
KPLCS   KPLCS   KPLHC   1   110.7   156.6
KPLCS   KPLCT   KPLHC   1   110.7   156.6
KPLCT   KPLCS   KPLHC   1   110.7   156.6
KPLH1   KPLC1   KPLH1   1   107.8   138.1
KPLHC   KPLC2   KPLHC   1   107.8   138.1
KPLHC   KPLCS   KPLHC   1   107.8   138.1
KPLHC   KPLCT   KPLHC   1   107.8   138.1
KPLNA   KPLC1   KPLC2   1   112.7   418.4
KPLC1   KPLC2   KPLCS   1   112.7   418.4
KPLC2   KPLCS   KPLCS   1   112.7   418.4
KPLCS   KPLCS   KPLCS   1   112.7   418.4
KPLCS   KPLCS   KPLCT   1   112.7   418.4
; triflate/bistriflate angle types
KPL_F   KPL_C   KPL_F   1   107.1   781.0
KPL_S   KPL_C   KPL_F   1   111.8   694.0
KPL_C   KPL_S   TF_O    1   102.6   870.0
KPL_C   KPL_S   NTF2_O  1   102.6   870.0
TF_O    KPL_S   TF_O    1   115.3   969.0
NTF2_O  KPL_S   NTF2_O  1   118.5   969.0
NTF2_O  KPL_S   NTF2_N  1   113.6   789.0
KPL_C   KPL_S   NTF2_N  1   100.2   816.0
KPL_S   NTF2_N  KPL_S   1   125.6   671.0

[ dihedraltypes ]
```

```
; imidazolium proper dihedral types
KPLCW   KPLNA   KPLC1   KPLH1   3    0.25950   0.77850  -0.00000  -
1.03800   0.00000   0.00000
KPLCR   KPLNA   KPLC1   KPLH1   3    0.00000   0.00000  -0.00000  -
0.00000   0.00000   0.00000
KPLCW   KPLNA   KPLC1   KPLC2   3    2.92600   4.76800  -6.10600  -
1.58800   0.00000   0.00000
KPLCR   KPLNA   KPLC1   KPLC2   3   -2.63450   2.63450  -0.00000  -
0.00000   0.00000   0.00000
KPLNA   KPLC1   KPLC2   KPLCS   3   -1.17750   1.93550  -3.16400
2.40600   0.00000   0.00000
KPLNA   KPLC1   KPLC2   KPLHC   3    0.00000   0.00000  -0.00000  -
0.00000   0.00000   0.00000
KPLH1   KPLC1   KPLC2   KPLCS   3    0.76550   2.29650  -0.00000  -
3.06200   0.00000   0.00000
KPLHC   KPLC2   KPLCS   KPLCS   3    0.76550   2.29650  -0.00000  -
3.06200   0.00000   0.00000
KPLHC   KPLCS   KPLC2   KPLC1   3    0.76550   2.29650  -0.00000  -
3.06200   0.00000   0.00000
KPLC2   KPLCS   KPLCS   KPLHC   3    0.76550   2.29650  -0.00000  -
3.06200   0.00000   0.00000
KPLCS   KPLCS   KPLCS   KPLHC   3    0.76550   2.29650  -0.00000  -
3.06200   0.00000   0.00000
KPLCS   KPLCS   KPLCT   KPLHC   3    0.76550   2.29650  -0.00000  -
3.06200   0.00000   0.00000
KPLCT   KPLCS   KPLCS   KPLHC   3    0.76550   2.29650  -0.00000  -
3.06200   0.00000   0.00000
KPLH1   KPLC1   KPLC2   KPLHC   3    0.66550   1.99650  -0.00000  -
2.66200   0.00000   0.00000
KPLHC   KPLC2   KPLCS   KPLHC   3    0.66550   1.99650  -0.00000  -
2.66200   0.00000   0.00000
KPLHC   KPLCS   KPLCS   KPLHC   3    0.66550   1.99650  -0.00000  -
2.66200   0.00000   0.00000
KPLHC   KPLCS   KPLCT   KPLHC   3    0.66550   1.99650  -0.00000  -
2.66200   0.00000   0.00000
KPLC1   KPLC2   KPLCS   KPLCS   3    3.56650  -1.88950   0.65700  -
2.33400   0.00000   0.00000
KPLC2   KPLCS   KPLCS   KPLCS   3    3.56650  -1.88950   0.65700  -
2.33400   0.00000   0.00000
KPLCS   KPLCS   KPLCS   KPLCS   3    3.56650  -1.88950   0.65700  -
2.33400   0.00000   0.00000
KPLCS   KPLCS   KPLCS   KPLCT   3    3.56650  -1.88950   0.65700  -
2.33400   0.00000   0.00000
X       KPLNA   KPLCR   X       3   19.46000   0.00000 -19.46000  -
0.00000   0.00000   0.00000
X       KPLCW   KPLCW   X       3   44.98000   0.00000 -44.98000  -
0.00000   0.00000   0.00000
X       KPLNA   KPLCW   X       3   12.55000   0.00000 -12.55000  -
0.00000   0.00000   0.00000
; triflate proper dihedral types
KPL_F   KPL_C   KPL_S   TF_O    3    0.72550   2.17650  -0.00000  -
2.90200   0.00000   0.00000
; bistriflate proper dihedral types in rtp file to override default
RB?
KPL_F   KPL_C   KPL_S   NTF2_N  9   0.0     2.0401      3
KPL_S   NTF2_N   KPL_S   KPL_C   9   0.0     23.7647     1
```

```
KPL_S   NTF2_N   KPL_S   KPL_C   9   0.0     6.2081      2
KPL_S   NTF2_N   KPL_S   KPL_C   9   0.0     -2.3684     3
KPL_S   NTF2_N   KPL_S   KPL_C   9   0.0     -0.0298     4
KPL_S   NTF2_N   KPL_S   KPL_C   9   0.0     0.6905      5
KPL_S   NTF2_N   KPL_S   KPL_C   9   0.0     1.0165      6


; Ions
; dimethyl phosphate, anion
[ DMPO ]
[ atoms ]
C1      opls_443        0.300       1
H11     opls_444       -0.030       1
H12     opls_444       -0.030       1
H13     opls_444       -0.030       1
O2      opls_442       -0.600       2
O6      opls_442       -0.600       2
P3      opls_440        1.620       2
O4      opls_441       -0.920       2
O5      opls_441       -0.920       2
C7      opls_443        0.300       3
H71     opls_444       -0.030       3
H72     opls_444       -0.030       3
H73     opls_444       -0.030       3

[ bonds ]
C1  H11
C1  H12
C1  H13
C1  O2
O2  P3
P3  O4
P3  O5
P3  O6
O6  C7
C7  H71
C7  H72
C7  H73

; triflate anion
[ TFO ]
[ atoms ]
O1      kpl_015     -0.630      1
S2      kpl_014      1.020      1
O3      kpl_015     -0.630      1
O4      kpl_015     -0.630      1
C5      kpl_012      0.350      1
F6      kpl_013     -0.160      1
F7      kpl_013     -0.160      1
F8      kpl_013     -0.160      1

[ bonds ]
O1  S2
S2  O3
S2  O4
S2  C5
```

```
C5  F6
C5  F7
C5  F8

; methyl deca imidazolium cation
[ C10M ]
[ atoms ]
 C1     kpl_003     -0.110      1
H11     kpl_007      0.210      1
 N2     kpl_011      0.150      1
 C3     kpl_001     -0.170      1
H31     kpl_010      0.130      1
H32     kpl_010      0.130      1
H33     kpl_010      0.130      1
C4      kpl_006     -0.130      1
H41     kpl_008      0.210      1
C5      kpl_006     -0.130      1
H51     kpl_008      0.210      1
N6      kpl_011      0.150      1
C7      kpl_001     -0.170      1
H71     kpl_010      0.130      1
H72     kpl_010      0.130      1
C8      kpl_002      0.010      1
H81     kpl_009      0.060      1
H82     kpl_009      0.060      1
C9      kpl_004     -0.120      2
H91     kpl_009      0.060      2
H92     kpl_009      0.060      2
C10     kpl_004     -0.120      3
H101    kpl_009      0.060      3
H102    kpl_009      0.060      3
C11     kpl_004     -0.120      4
H111    kpl_009      0.060      4
H112    kpl_009      0.060      4
C12     kpl_004     -0.120      5
H121    kpl_009      0.060      5
H122    kpl_009      0.060      5
C13     kpl_004     -0.120      6
H131    kpl_009      0.060      6
H132    kpl_009      0.060      6
C14     kpl_004     -0.120      7
H141    kpl_009      0.060      7
H142    kpl_009      0.060      7
C15     kpl_004     -0.120      8
H151    kpl_009      0.060      8
H152    kpl_009      0.060      8
C16     kpl_005     -0.180      9
H161    kpl_009      0.060      9
H162    kpl_009      0.060      9
H163    kpl_009      0.060      9

[ bonds ]
C1  H11
C1  N2
C1  N6
N2  C3
```

```
N2  C4
C3  H31
C3  H32
C3  H33
C4  H41
C4  C5
C5  H51
C5  N6
N6  C7
C7  H71
C7  H72
C7  C8
C8  H81
C8  H82
C8  C9
C9  H91
C9  H92
C9  C10
C10 H101
C10 H102
C10 C11
C11 H111
C11 H112
C11 C12
C12 H121
C12 H122
C12 C13
C13 H131
C13 H132
C13 C14
C14 H141
C14 H142
C14 C15
C15 H151
C15 H152
C15 C16
C16 H161
C16 H162
C16 H163

[ impropers ]
C1  N2  C4  C3  improper_Z_N_X_Y
C1  N6  C5  C7  improper_Z_N_X_Y
H11 C1  N6  N2  improper_Z_CA_X_Y
N2  C4  C5  H41 improper_Z_CA_X_Y
N6  C5  H51 C4  improper_Z_CA_X_Y

; bistrifylimide anion
[ NTF2 ]
[ atoms ]
F1      kpl_013     -0.189      1
C2      kpl_012      0.494      1
F3      kpl_013     -0.189      1
F4      kpl_013     -0.189      1
S5      kpl_014      1.076      1
N8      kpl_017     -0.690      1
```

```
O6      kpl_016     -0.579      1
O7      kpl_016     -0.579      1
S9      kpl_014      1.076      1
C12     kpl_012      0.494      1
O10     kpl_016     -0.579      1
O11     kpl_016     -0.579      1
F13     kpl_013     -0.189      1
F14     kpl_013     -0.189      1
F15     kpl_013     -0.189      1

[ bonds ]
F1  C2
C2  F3
C2  F4
C2  S5
S5  O6
S5  O7
S5  N8
N8  S9
S9  O10
S9  O11
S9  C12
C12 F13
C12 F14
C12 F15

; Solutes
; cyclohexanol
 [ HAOL ]
[ atoms ]
  C1     opls_136   -0.120     1
  H11    opls_140    0.060     1
  H12    opls_140    0.060     1
  C2     opls_136   -0.120     2
  H21    opls_140    0.060     2
  H22    opls_140    0.060     2
  C3     opls_136   -0.120     3
  H31    opls_140    0.060     3
  H32    opls_140    0.060     3
  C4     opls_136   -0.120     4
  H41    opls_140    0.060     4
  H42    opls_140    0.060     4
  C5     opls_136   -0.120     5
  H51    opls_140    0.060     5
  H52    opls_140    0.060     5
  C6     opls_158    0.205     6
  H61    opls_140    0.060     6
  O7     opls_154   -0.683     7
  H71    opls_155    0.418     8

[ bonds ]
 H31    C3
 H11    C1
 H21    C2
 C3     H32
 C1     H12
```

```
 C2     H22
 H51    C5
 H41    C4
 C5     H52
 C4     H42
 C6     H61
 C3     C2
 C3     C4
 C1     C2
 C1     C6
 C5     C4
 C5     C6
 O7     C6
 O7     H71

; cyclohexanone
[ HONE ]
[ atoms ]
  C1     opls_136   -0.120     1
  H11    opls_282    0.060     1
  H12    opls_282    0.060     1
  C2     opls_136   -0.120     2
  H21    opls_140    0.060     2
  H22    opls_140    0.060     2
  C3     opls_136   -0.120     3
  H31    opls_140    0.060     3
  H32    opls_140    0.060     3
  C4     opls_136   -0.120     4
  H41    opls_140    0.060     4
  H42    opls_140    0.060     4
  C5     opls_136   -0.120     5
  H51    opls_282    0.060     5
  H52    opls_282    0.060     5
  C6     opls_280    0.470     6
  O7     opls_281   -0.470     7

[ bonds ]
 H11    C1
 H31    C3
 H21    C2
 C3     H32
 C1     H12
 C2     H22
 H51    C5
 C5     H52
 C4     H41
 C4     H42
 C3     C2
 C3     C4
 C1     C2
 C5     C4
 C1     C6
 C6     C5
 C6     O7

[ impropers ]
```

```
  C1   C5   C6   O7 improper_O_C_X_Y

; cyclohexylacetate
[ HACT ]
[ atoms ]
  C1     opls_136   -0.120     1
  H11    opls_140    0.060     1
  H12    opls_140    0.060     1
  C2     opls_136   -0.120     2
  H21    opls_140    0.060     2
  H22    opls_140    0.060     2
  C3     opls_136   -0.120     3
  H31    opls_140    0.060     3
  H32    opls_140    0.060     3
  C4     opls_136   -0.120     4
  H41    opls_140    0.060     4
  H42    opls_140    0.060     4
  C5     opls_136   -0.120     5
  H51    opls_140    0.060     5
  H52    opls_140    0.060     5
  C6     opls_491    0.220     6
  H61    opls_469    0.030     6
  O7     opls_467   -0.330     7
  C8     opls_465    0.510     7
  O9     opls_466   -0.430     7
  C10    opls_135   -0.180     8
  H101   opls_140    0.060     8
  H102   opls_140    0.060     8
  H103   opls_140    0.060     8

[ bonds ]
 H101   C10
 H102   C10
 C10    H103
 H61    C6
 H21    C2
 H11    C1
 H41    C4
 C1     H12
 C2     H22
 H51    C5
 C5     H52
 C4     H42
 C3     H31
 C3     H32
 C10    C8
 C8     O9
 C8     O7
 O7     C6
 C6     C1
 C6     C5
 C1     C2
 C2     C3
 C5     C4
 C4     C3
```

```
[ impropers ]
  O7  C10   C8   O9 improper_O_C_X_Y

DMSO
[ DMSO  ]
 [ atoms ]
OD      opls_125    -0.459      1
SD      opls_124     0.139      1
CD1     opls_139    -0.020      1
HD11        opls_140     0.060      1
HD12        opls_140     0.060      1
HD13        opls_140     0.060      1
CD2     opls_139    -0.020      1
HD21        opls_140     0.060      1
HD22        opls_140     0.060      1
HD23        opls_140     0.060      1

 [ bonds ]
    OD     SD
    SD     CD1
    SD     CD2
    CD1    HD11
    CD1    HD12
    CD1    HD13
    CD2    HD21
    CD2    HD22
    CD2    HD23
```


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