Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting information

Understanding of the interactions between azole-anion-based ionic liquids and 2-methyl-3-butyn-2-ol from the experimental perspective: the cage effect

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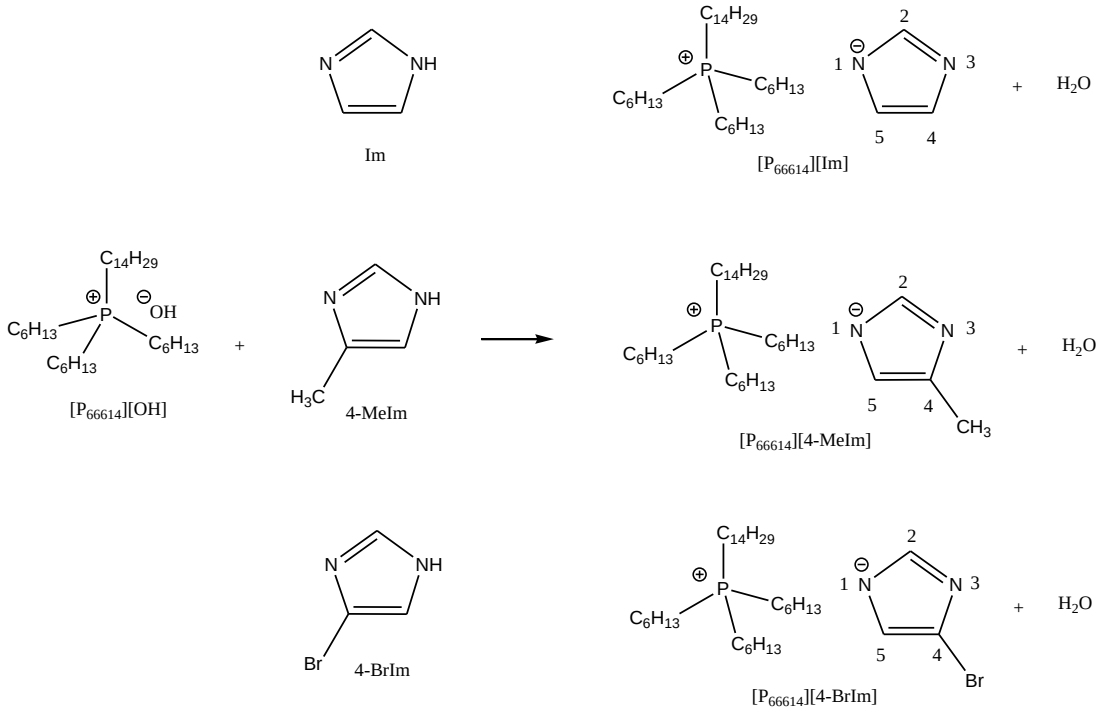

Scheme S1. Molecular structure and synthesis of AILs.

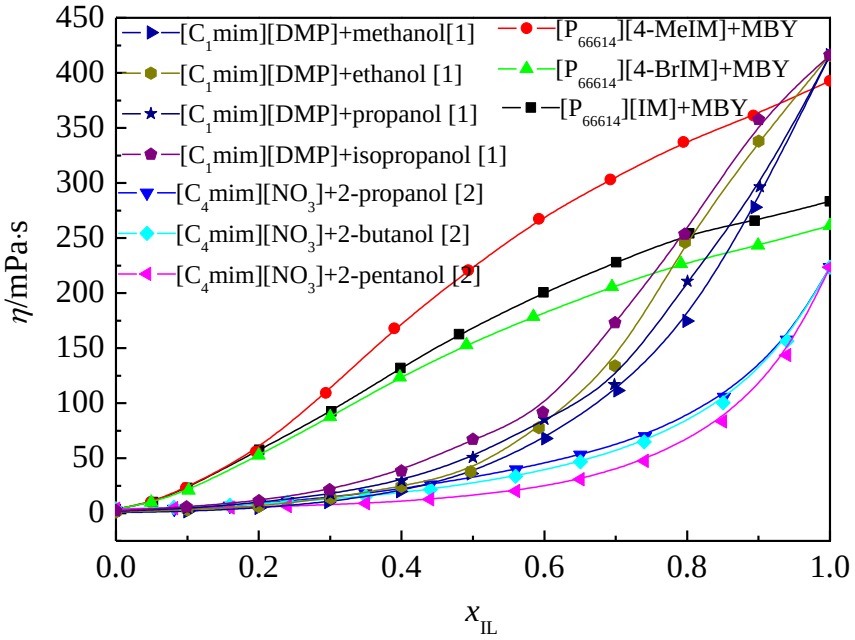

Fig. S1 Comparison of the concentration-dependent viscosity of ILs + alcohol mixtures at T = 293.15 K.

References

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[2] Heydarian, S.; Almasi, M.; Saadati, Z. Thermophysical study of binary mixtures of 1-butyl-3-methylimidazolium nitrate ionic liquid + alcohols at different temperatures. *J. Chem. Thermodyn.* **2019**, *135*, 345-351.

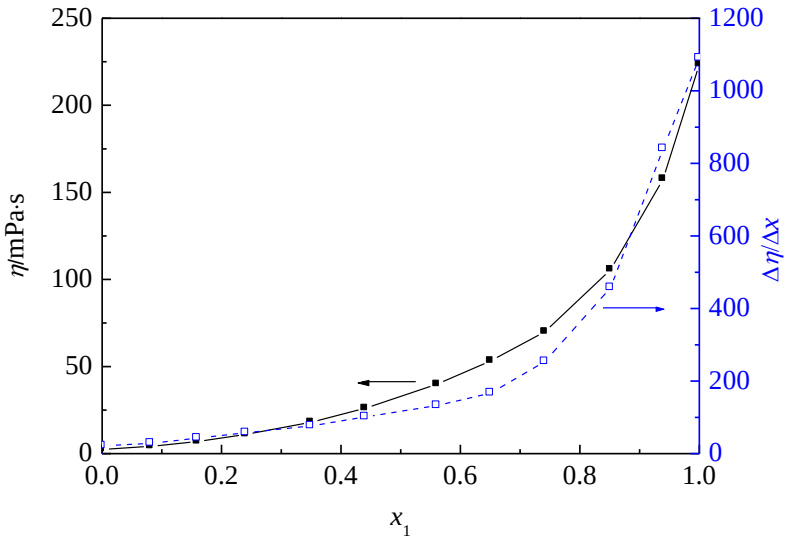

Fig. S2 Viscosity and their derivative of [Bmim][NO_3](1) + 2-propanol (2) mixture [2] with the mole fraction of IL at T = 293.15 K.

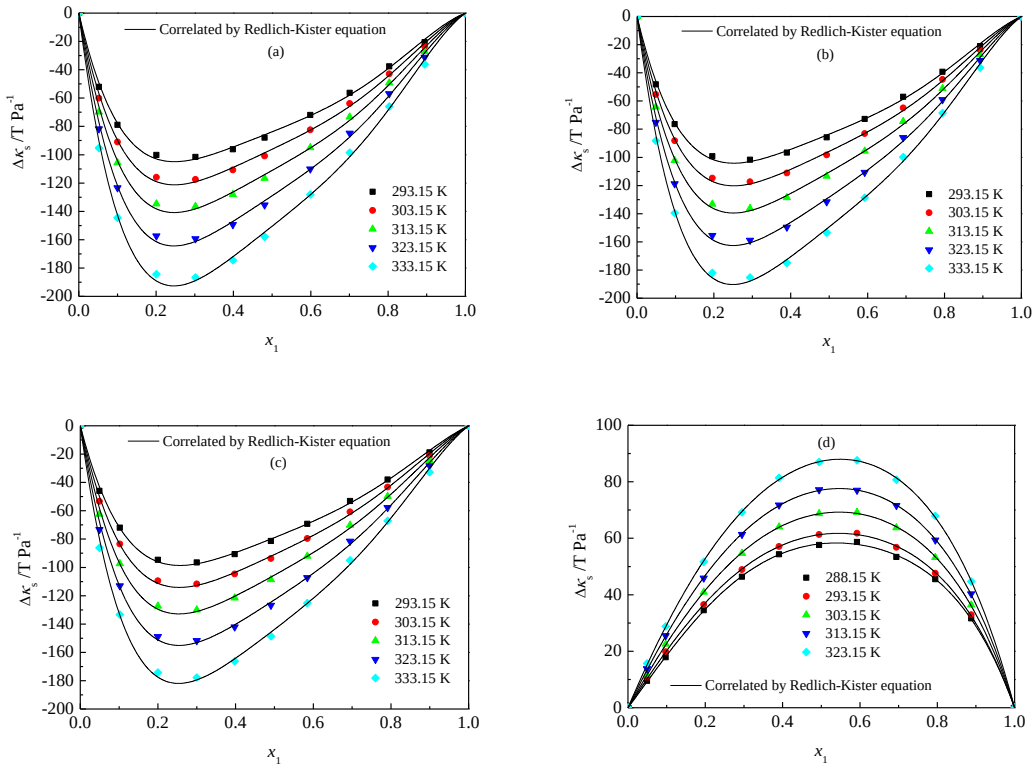

Fig. S3 Isentropic compressibility deviations $\Delta\kappa_s$ of AILs (1) + MBY (2) mixtures with the mole fraction of AIL, (a), $[P_{66614}][Im]$; (b), $[P_{66614}][4\text{-MeIm}]$; (c), $[P_{66614}][4\text{-BrIm}]$; (d), isentropic compressibility deviations $\Delta\kappa_s$ of CCl_4 (1) + MBY (2) mixture with the mole fraction of CCl_4.

Table S1. Comparison of experimental values of density (ρ), viscosity (η), and speed of sound (u) of the pure MBY and CCl_4 at various temperatures with corresponding literature values.

T (K)	ρ (g·cm^{-3})		η (mPa·s)		u (m·s^{-1})	
	Exp.	Lit.	Exp.	Lit.	Exp.	Lit.
			MBY			
293.15	0.86210	0.865 [3]	3.654	3.38 [3]	1263.39	
303.15	0.85219	0.8518 [4]	2.489		1223.66	
313.15	0.84205		1.769	1.7752 [5]	1183.87	
323.15	0.83163		1.322	1.3471 [5]	1143.96	
333.15	0.82094	0.821 [3]	1.022	1.04 [3]	1103.93	
			CCl_4			
288.15	1.60391		1.050		952.74	
293.15	1.59420		0.987		936.99	921.26 [6]
303.15	1.57470	1.57461 [7]	0.845		905.65	
313.15	1.55508	1.55496 [7]	0.742		874.77	
323.15	1.53530		0.659		844.32	

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Table S2. Mole fraction (x_1), density (ρ), viscosity (η), speed of sound (u), isentropic compressibility (κ_s), excess molar volume (V^E), and isentropic compressibility deviation ($\Delta\kappa_s$) of AILs (1) + MBY (2) and CCl_4 (1) + MBY (2) mixtures.

x_1	ρ (g·cm^{-3})	η (mPa·s)	u (m·s^{-1})	κ_s (T Pa^{-1})	V^E (cm^3·mol^{-1})	$\Delta\kappa_s$ (T Pa^{-1})
			[P_{66614}][Im] (1) + MBY (2)			
			T = 293.15 K			
0.0000	0.86210	3.654	1263.39	726.72	0.0000	0.0
0.0521	0.87556	10.13	1313.04	662.46	-0.3261	-52.1
0.1000	0.88330	23.02	1346.41	624.51	-0.5559	-78.9
0.2007	0.89193	57.57	1390.59	579.79	-0.7062	-100.2
0.3016	0.89632	92.58	1418.01	554.85	-0.6549	-101.6
0.3984	0.89913	132.0	1438.17	537.72	-0.6116	-96.2
0.4808	0.90090	162.7	1451.53	526.83	-0.5647	-87.9
0.5987	0.90266	200.7	1466.28	515.28	-0.4802	-72.0
0.7006	0.90377	227.9	1476.86	507.30	-0.3721	-56.3
0.8033	0.90463	254.3	1483.86	502.05	-0.2485	-37.6
0.8948	0.90524	265.7	1489.83	497.69	-0.1284	-20.6
1.0000	0.90584	283.2	1495.17	493.82	0.0000	0.0
			T = 303.15 K			
0.0000	0.85219	2.489	1223.66	783.69	0.0000	0.0
0.0521	0.86677	6.369	1274.87	709.85	-0.3608	-60.1
0.1000	0.87517	14.01	1309.56	666.28	-0.6146	-91.0
0.2007	0.88465	33.68	1355.84	614.91	-0.8015	-115.9
0.3016	0.88952	53.67	1384.23	586.72	-0.7735	-117.5
0.3984	0.89252	74.88	1404.56	567.94	-0.7117	-110.7
0.4808	0.89437	90.92	1418.00	556.07	-0.6538	-100.9
0.5987	0.89632	111.0	1432.82	543.44	-0.5552	-82.4
0.7006	0.89753	125.5	1442.86	535.18	-0.4415	-63.8
0.8033	0.89846	139.3	1450.47	529.03	-0.3043	-42.9
0.8948	0.89911	145.6	1456.48	524.30	-0.1633	-23.5
1.0000	0.89973	155.9	1461.85	520.09	0.0000	0.0
			T = 313.15 K			
0.0000	0.84205	1.769	1183.87	847.33	0.0000	0.0

0.0521	0.85784	4.432	1236.96	761.87	-0.4028	-69.8
0.1000	0.86695	8.974	1273.18	711.58	-0.6837	-105.7
0.2007	0.87736	20.95	1321.82	652.35	-0.9177	-134.7
0.3016	0.88270	33.95	1351.43	620.30	-0.9077	-136.5
0.3984	0.88593	45.76	1372.22	599.45	-0.8390	-128.2
0.4808	0.88791	54.92	1385.90	586.36	-0.7659	-116.6
0.5987	0.88999	66.42	1400.86	572.57	-0.6469	-95.0
0.7006	0.89128	74.49	1410.97	563.57	-0.5066	-73.4
0.8033	0.89227	82.21	1418.70	556.83	-0.3449	-49.3
0.8948	0.89298	86.62	1424.81	551.63	-0.1903	-27.0
1.0000	0.89364	92.00	1430.26	547.03	0.0000	0.0
			T = 323.15 K			
0.0000	0.83163	1.322	1143.96	918.86	0.0000	0.0
0.0521	0.84874	3.214	1199.22	819.27	-0.4530	-81.7
0.1000	0.85860	6.110	1237.02	761.12	-0.7636	-123.3
0.2007	0.87000	14.83	1288.28	692.56	-1.0489	-157.3
0.3016	0.87587	22.96	1319.41	655.84	-1.0661	-159.3
0.3984	0.87936	30.09	1340.81	632.56	-0.9936	-149.3
0.4808	0.88146	35.56	1354.78	618.10	-0.9011	-135.4
0.5987	0.88369	42.44	1369.97	602.95	-0.7576	-110.0
0.7006	0.88504	47.42	1380.18	593.15	-0.5828	-84.8
0.8033	0.88611	52.06	1388.06	585.73	-0.3985	-56.9
0.8948	0.88681	54.28	1394.27	580.06	-0.2370	-31.1
1.0000	0.88757	57.95	1399.81	574.99	0.0000	0.0
			T = 333.15 K			
0.0000	0.82094	1.022	1103.93	999.55	0.0000	0.0
0.0521	0.83945	2.421	1161.00	883.77	-0.5093	-95.2
0.1000	0.85012	4.403	1201.00	815.52	-0.8561	-144.5
0.2007	0.86259	10.56	1255.18	735.84	-1.2010	-184.4
0.3016	0.86902	16.31	1288.01	693.64	-1.2464	-186.7
0.3984	0.87283	20.78	1310.12	667.50	-1.1837	-174.5
0.4808	0.87504	24.41	1324.45	651.48	-1.0595	-157.9
0.5987	0.87741	28.76	1339.93	634.79	-0.8873	-128.0
0.7006	0.87888	32.05	1350.29	624.05	-0.7019	-98.5
0.8033	0.87997	34.88	1358.34	615.91	-0.4613	-66.0

0.8948	0.88081	36.65	1364.62	609.67	-0.2761	-36.1
1.0000	0.88153	39.06	1370.29	604.14	0.0000	0.0
			$[P_{66614}][4\text{-MeIm}]$ (1) + MBY (2)			
			T = 293.15 K			
0.0000	0.86210	3.654	1263.39	726.72	0.0000	0.0
0.0488	0.87490	10.05	1308.67	667.39	-0.2454	-48.2
0.0980	0.88321	22.97	1342.84	627.90	-0.4648	-76.4
0.1959	0.89260	55.88	1386.50	582.78	-0.7226	-99.2
0.2942	0.89752	109.4	1413.34	557.78	-0.7975	-101.7
0.3900	0.90039	168.0	1432.74	541.05	-0.7678	-96.6
0.4936	0.90251	220.7	1448.38	528.18	-0.7098	-85.8
0.5923	0.90401	267.5	1460.45	518.62	-0.6541	-72.8
0.6928	0.90513	303.1	1469.71	511.48	-0.5690	-57.0
0.7951	0.90597	337.2	1477.26	505.79	-0.4472	-39.3
0.8931	0.90650	361.3	1482.82	501.71	-0.2509	-21.0
1.0000	0.90692	392.8	1487.61	498.26	0.0000	0.0
			T = 303.15 K			
0.0000	0.85219	2.489	1223.66	783.69	0.0000	0.0
0.0488	0.86603	6.354	1270.21	715.68	-0.2718	-55.4
0.0980	0.87504	14.04	1305.83	670.19	-0.5171	-88.2
0.1959	0.88527	33.15	1351.45	618.48	-0.8149	-114.7
0.2942	0.89063	61.19	1379.08	590.37	-0.9043	-117.4
0.3900	0.89375	92.40	1398.59	572.01	-0.8782	-111.1
0.4936	0.89602	119.3	1414.18	558.05	-0.8047	-98.3
0.5923	0.89763	143.5	1426.10	547.78	-0.7345	-83.1
0.6928	0.89882	161.6	1435.31	540.05	-0.6288	-64.9
0.7951	0.89972	177.9	1442.83	533.90	-0.4865	-44.7
0.8931	0.90030	192.0	1448.39	529.47	-0.2710	-23.8
1.0000	0.90077	207.1	1453.20	525.70	0.0000	0.0
			T = 313.15 K			
0.0000	0.84205	1.769	1183.87	847.33	0.0000	0.0
0.0488	0.85700	4.352	1232.03	768.73	-0.3038	-64.3
0.0980	0.86677	9.001	1269.16	716.25	-0.5797	-102.3
0.1959	0.87793	21.28	1317.11	656.59	-0.9278	-133.2
0.2942	0.88373	37.74	1345.88	624.69	-1.0322	-136.3

0.3900	0.88711	55.03	1365.89	604.21	-1.0027	-128.6
0.4936	0.88953	69.85	1381.54	589.00	-0.9072	-113.4
0.5923	0.89124	83.22	1393.47	577.84	-0.8190	-95.6
0.6928	0.89251	93.85	1402.71	569.44	-0.6884	-74.5
0.7951	0.89349	102.2	1410.29	562.72	-0.5277	-51.2
0.8931	0.89412	109.8	1415.90	557.88	-0.2878	-27.2
1.0000	0.89465	118.5	1420.80	553.71	0.0000	0.0
			T = 323.15 K			
0.0000	0.83163	1.322	1143.96	918.86	0.0000	0.0
0.0488	0.84780	3.125	1194.01	827.35	-0.3442	-75.1
0.0980	0.85843	6.118	1232.34	767.07	-0.6634	-118.8
0.1959	0.87053	14.57	1283.27	697.56	-1.0602	-155.4
0.2942	0.87683	24.97	1313.47	661.07	-1.1876	-158.9
0.3900	0.88048	35.41	1334.09	638.13	-1.1601	-149.6
0.4936	0.88305	43.94	1350.04	621.33	-1.0396	-131.6
0.5923	0.88486	51.79	1362.10	609.13	-0.9269	-110.6
0.6928	0.88620	58.39	1371.44	599.95	-0.7647	-86.0
0.7951	0.88725	62.77	1379.09	592.61	-0.5794	-59.0
0.8931	0.88790	67.58	1384.78	587.32	-0.2966	-31.3
1.0000	0.88851	72.43	1389.77	582.71	0.0000	0.0.0
			T = 333.15 K			
0.0000	0.82094	1.022	1103.93	999.55	0.0000	0.0
0.0488	0.83842	2.326	1156.00	892.53	-0.3927	-88.2
0.0980	0.84989	4.368	1196.37	822.07	-0.7486	-139.6
0.1959	0.86305	10.28	1249.82	741.77	-1.2097	-182.0
0.2942	0.86990	17.35	1281.00	700.54	-1.3659	-185.3
0.3900	0.87385	23.88	1303.25	673.76	-1.3443	-175.0
0.4936	0.87658	29.57	1319.41	655.31	-1.1987	-153.4
0.5923	0.87850	34.39	1331.63	641.94	-1.0580	-128.6
0.6928	0.87991	38.96	1341.12	631.87	-0.8598	-99.8
0.7951	0.88103	41.34	1349.10	623.62	-0.6444	-68.5
0.8931	0.88176	43.99	1354.68	617.98	-0.3453	-36.3
1.0000	0.88239	47.11	1359.77	612.93	0.0000	0.0
			$[P_{66614}]$[4-BrIm] (1) + MBY (2)			
			T = 293.15K			

0.0000	0.86210	3.654	1263.39	726.72	0.0000	0.0
0.0495	0.89371	9.832	1292.50	669.80	-0.1734	-46.0
0.1019	0.91474	20.82	1314.99	632.20	-0.2923	-72.0
0.1998	0.93836	52.58	1346.35	587.92	-0.4876	-94.6
0.3000	0.95174	87.63	1365.22	563.74	-0.4675	-96.5
0.3975	0.96020	123.6	1378.49	548.06	-0.4185	-90.6
0.4909	0.96598	152.9	1388.94	536.62	-0.3700	-81.4
0.5845	0.97032	178.5	1397.14	527.97	-0.3060	-69.3
0.6949	0.97424	205.6	1405.41	519.67	-0.2302	-53.1
0.7914	0.97695	226.5	1412.04	513.37	-0.1701	-38.0
0.8992	0.97939	243.3	1416.65	508.77	-0.1001	-18.8
1.0000	0.98122	261.3	1420.33	505.19	0.0000	0.0
			T = 303.15 K			
0.0000	0.85219	2.489	1223.66	783.69	0.0000	0.0
0.0495	0.88467	6.363	1254.90	717.80	-0.1965	-53.4
0.1019	0.90630	12.89	1278.93	674.58	-0.3314	-83.5
0.1998	0.93062	31.46	1312.26	624.00	-0.5536	-109.5
0.3000	0.94442	51.69	1332.42	596.42	-0.5499	-111.7
0.3975	0.95312	71.89	1346.17	578.97	-0.5033	-104.6
0.4909	0.95904	88.22	1356.84	566.38	-0.4455	-93.7
0.5845	0.96346	102.1	1365.19	556.90	-0.3643	-79.6
0.6949	0.96746	117.0	1373.42	547.97	-0.2690	-60.8
0.7914	0.97024	127.4	1380.03	541.18	-0.1997	-43.3
0.8992	0.97272	137.5	1384.80	536.09	-0.1045	-21.2
1.0000	0.97461	147.3	1388.90	531.90	0.0000	0.0
			T = 313.15 K			
0.0000	0.84205	1.769	1183.87	847.33	0.0000	0.0
0.0495	0.87546	4.337	1217.39	770.73	-0.2242	-62.4
0.1019	0.89773	9.413	1243.15	720.79	-0.3803	-97.3
0.1998	0.92282	20.13	1278.82	662.62	-0.6344	-127.3
0.3000	0.93709	32.79	1300.36	631.09	-0.6553	-130.0
0.3975	0.94604	44.89	1314.85	611.42	-0.6158	-121.6
0.4909	0.95209	54.49	1325.85	597.49	-0.5476	-108.7
0.5845	0.95662	62.96	1334.37	587.09	-0.4533	-92.1
0.6949	0.96070	71.83	1342.74	577.34	-0.3362	-70.2

0.7914	0.96353	76.98	1349.43	569.95	-0.2488	-49.8
0.8992	0.96605	83.08	1354.34	564.35	-0.1238	-24.4
1.0000	0.96798	88.91	1358.55	559.73	0.0000	0.0
			T = 323.15 K			
0.0000	0.83163	1.322	1143.96	918.86	0.0000	0.0
0.0495	0.86607	3.134	1179.95	829.32	-0.2576	-73.2
0.1019	0.88905	5.670	1206.86	772.25	-0.4376	-113.0
0.1998	0.91494	13.52	1245.78	704.25	-0.7247	-148.8
0.3000	0.92971	21.94	1268.84	668.10	-0.7734	-151.7
0.3975	0.93896	29.79	1284.31	645.67	-0.7458	-142.0
0.4909	0.94518	36.02	1295.82	630.08	-0.6705	-126.7
0.5845	0.94980	41.23	1304.48	618.72	-0.5581	-107.2
0.6949	0.95396	46.82	1313.04	608.01	-0.4159	-81.5
0.7914	0.95685	50.10	1319.84	599.95	-0.3062	-57.7
0.8992	0.95941	53.50	1324.83	593.85	-0.1453	-28.2
1.0000	0.96138	57.36	1329.17	588.77	0.0000	0.0
			T = 333.15 K			
0.0000	0.82094	1.022	1103.93	999.55	0.0000	0.0
0.0495	0.85648	2.353	1142.48	894.51	-0.2964	-86.2
0.1019	0.88021	4.160	1171.71	827.51	-0.5029	-133.3
0.1998	0.90698	9.530	1213.01	749.33	-0.8274	-174.3
0.3000	0.92231	15.30	1237.79	707.67	-0.9088	-177.8
0.3975	0.93187	20.90	1254.37	682.01	-0.8932	-166.3
0.4909	0.93827	24.70	1266.73	664.21	-0.8117	-148.6
0.5845	0.94301	28.25	1275.36	651.96	-0.6827	-125.2
0.6949	0.94725	31.99	1284.12	640.21	-0.5112	-95.0
0.7914	0.95017	34.18	1291.05	631.41	-0.3536	-67.1
0.8992	0.95279	36.54	1296.16	624.72	-0.1727	-32.8
1.0000	0.95481	38.72	1300.59	619.16	0.0000	0.0
			CCl_4 (1) + MBY (2)			
			T = 288.15 K			
0.0000	0.86695	4.501	1282.55	701.22	0.0000	0.0
0.0495	0.90252	3.935	1249.23	710.00	0.1376	9.5
0.0978	0.93725	3.516	1219.27	717.70	0.2588	17.9
0.1962	1.00809	2.830	1163.39	732.91	0.4596	34.5

0.2949	1.07909	2.271	1116.53	743.36	0.6270	46.4
0.3908	1.14857	1.935	1077.51	749.89	0.7117	54.3
0.4945	1.22444	1.650	1042.35	751.68	0.7206	57.6
0.5923	1.29616	1.450	1013.26	751.45	0.7010	58.7
0.6945	1.37163	1.305	989.46	744.67	0.6308	53.4
0.7948	1.44605	1.191	969.76	735.34	0.5363	45.5
0.8879	1.51625	1.120	957.02	720.09	0.3739	31.6
1.0000	1.60391	1.050	952.74	686.86	0.0000	0.0
			T = 293.15 K			
0.0000	0.86210	3.654	1263.39	726.72	0.0000	0.0
0.0495	0.89738	3.204	1229.98	736.59	0.1447	10.5
0.0978	0.93186	2.896	1199.94	745.30	0.2711	19.8
0.1962	1.00219	2.381	1145.22	760.80	0.4781	36.5
0.2949	1.07267	1.963	1098.86	772.06	0.6516	48.9
0.3908	1.14163	1.680	1060.35	779.07	0.7417	57.1
0.4945	1.21692	1.435	1025.12	781.97	0.7549	61.3
0.5923	1.28817	1.288	996.81	781.27	0.7337	61.8
0.6945	1.36307	1.184	972.92	775.05	0.6660	56.8
0.7948	1.43704	1.082	954.03	764.55	0.5662	47.6
0.8879	1.50684	1.028	941.43	748.78	0.3961	32.9
1.0000	1.59420	0.987	936.99	714.48	0.0000	0.0
			T = 303.15 K			
0.0000	0.85219	2.489	1223.66	783.69	0.0000	0.0
0.0495	0.88693	2.222	1190.98	794.88	0.1581	11.7
0.0978	0.92088	2.043	1161.26	805.26	0.2956	22.5
0.1962	0.99016	1.736	1107.95	822.72	0.5192	40.9
0.2949	1.05959	1.488	1062.73	835.63	0.7066	54.7
0.3908	1.12752	1.318	1025.13	843.95	0.8075	64.0
0.4945	1.20169	1.170	990.79	847.70	0.8289	68.7
0.5923	1.27191	1.051	963.29	847.28	0.8109	69.2
0.6945	1.34576	0.971	940.08	840.82	0.7425	63.7
0.7948	1.41878	0.907	921.89	829.33	0.6353	53.2
0.8879	1.48781	0.863	909.97	811.71	0.4489	36.4
1.0000	1.57470	0.845	905.65	774.25	0.0000	0.0
			T = 313.15 K			

0.0000	0.84205	1.769	1183.87	847.33	0.0000	0.0
0.0495	0.87624	1.618	1151.41	860.83	0.1722	13.8
0.0978	0.90966	1.509	1122.71	872.14	0.3213	25.5
0.1962	0.97787	1.321	1070.78	891.90	0.5652	45.9
0.2949	1.04623	1.165	1026.74	906.68	0.7692	61.4
0.3908	1.11311	1.050	990.14	916.37	0.8829	71.8
0.4945	1.18617	0.958	956.71	921.07	0.9131	77.2
0.5923	1.25535	0.879	930.08	920.86	0.8995	77.0
0.6945	1.32818	0.823	907.58	914.06	0.8296	71.6
0.7948	1.40026	0.773	890.19	901.21	0.7138	59.4
0.8879	1.46858	0.752	878.94	881.42	0.5072	40.3
1.0000	1.55508	0.742	874.77	840.35	0.0000	0.0
			T = 323.15 K			
0.0000	0.83163	1.322	1143.96	918.86	0.0000	0.0
0.0495	0.86527	1.229	1112.21	934.28	0.1879	15.7
0.0978	0.89816	1.155	1084.19	947.19	0.3499	28.8
0.1962	0.96528	1.034	1033.67	969.58	0.6169	51.7
0.2949	1.03255	0.932	990.83	986.49	0.8399	69.2
0.3908	1.09838	0.855	955.00	998.25	0.9684	81.4
0.4945	1.17031	0.785	922.88	1003.25	1.0092	87.0
0.5923	1.23847	0.742	897.08	1003.35	1.0002	87.6
0.6945	1.31029	0.703	875.38	995.95	0.9278	80.7
0.7948	1.38143	0.671	858.30	982.64	0.8042	67.9
0.8879	1.44914	0.661	848.30	958.94	0.5705	44.7
1.0000	1.53530	0.659	844.32	913.68	0.0000	0.0

Table S3. Physical quantities of AILs, MBY, and CCl_4 in Eyring-UNIQUAC equation.

Compounds	r	q	M (g mol^{-1})	T (K)	ρ (g cm^{-3})	V (cm^3 mol^{-1})
CCl_4	3.33 [8]	2.82 [8]	153.82	288.15	1.60391	95.90
				293.15	1.59420	96.49
				303.15	1.57470	97.68
				313.15	1.55508	98.91
				323.15	1.53530	100.19
MBY	4.3137 [9]	3.984 [9]	84.12	293.15	0.86210	97.58
				303.15	0.85219	98.71
				313.15	0.84205	99.90
				323.15	0.83163	101.15
				333.15	0.82094	102.47
$[P_{66614}][Im]$	27.7312[a]	23.0796[a]	550.94	293.15	0.90584	608.21
				303.15	0.89973	612.34
				313.15	0.89364	616.51
				323.15	0.88757	620.73
				333.15	0.88153	624.98
$[P_{66614}][4\text{-MeIm}]$	28.6263[a]	23.3948[a]	564.96	293.15	0.90692	622.94
				303.15	0.90077	627.20
				313.15	0.89465	631.49
				323.15	0.88851	635.85
				333.15	0.88239	640.26
$[P_{66614}][4\text{-BrIm}]$	28.6804[a]	23.3788[a]	629.83	293.15	0.98122	641.88
				303.15	0.97461	646.24
				313.15	0.96798	650.66
				323.15	0.96138	655.13
				333.15	0.95481	659.64

[a] Volume parameter r and the surface area parameter q of three AILs ($[P_{66614}][Im]$, $[P_{66614}][4\text{-MeIm}]$, and $[P_{66614}][4\text{-BrIm}]$) cannot be obtained directly from the literature. Through literature research, it is found that the volume and surface area parameters of [P][Cl] have been reported in the literature [10], but there is no the related parameters of azole-based anions. Therefore, in order to obtain the volume and surface area parameters of AILs, we adopted the following strategies: first, based on the parameters r and q of [P][Cl] and [Cl] reported in the literature[9,10], the parameters r and q of $[P]^+$ are obtained by subtracting the parameter of [Cl]

from the parameters of [P][Cl], respectively, and then the *r* and *q* of the alkyl group are combined to obtain the parameters of $[P_{66614}]^+$. Secondly, use pyridine instead of $[Im]^-$ to obtain *r* and *q* of $[Im]^-$, similarly, 4-methylpyridine and *p*-bromopyridine instead of $[4\text{-MeIm}]^-$ and $[4\text{-BrIm}]^-$ to obtain *r* and *q* of $[4\text{-MeIm}]^-$ and $[4\text{-BrIm}]^-$, respectively. Finally, based on the parameters of cations and anions, according to the principle of group contribution method, the parameters of three AILs are obtained, which are listed in the Table S4.

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Table S4. Coefficient a_l of Redlich-Kister type polynomial equation and standard deviation σ of AILs (1) + MBY (2) and CCl_4 (1) + MBY (2) mixtures.

Y	systems	T/K	a_0	a_1	a_2	a_3	σ
	$[P_{66614}][Im]$ (1) + MBY (2)	293.15	-2.1636	1.3449	-2.2668	2.5539	0.0121
		303.15	-2.5272	1.6443	-2.5675	2.3357	0.0091
		313.15	-2.9727	2.0699	-2.7071	2.1816	0.0089
		323.15	-3.5018	2.5942	-2.9216	1.8089	0.0010
		333.15	-4.1571	3.0542	-3.0471	1.7170	0.0009
	$[P_{66614}][4\text{-MeIm}]$ (1) + MBY (2)	293.15	-2.8868	1.3538	-1.8562	0.2996	0.0148
		303.15	-3.2741	1.6795	-1.8972	0.1597	0.0176
		313.15	-3.6974	2.1297	-1.9902	-0.0375	0.0206
		323.15	-4.2399	2.6112	-2.0007	-0.0454	0.0191
V^E		333.15	-4.8726	3.2037	-2.1470	-0.4825	0.0098
($cm^3 \cdot mol^{-1}$)	$[P_{66614}][4\text{-BrIm}]$ (1) + MBY (2)	293.15	-1.4499	1.5112	-1.3329	-0.0929	0.1368
		303.15	-1.7461	1.7708	-1.2875	-0.1584	0.0130
		313.15	-2.1582	2.0051	-1.2506	-0.2325	0.0120
		323.15	-2.6422	2.2315	-1.1746	-0.2301	0.0100
		333.15	-3.2024	2.4926	-1.0215	-0.1704	0.0057
	CCl_4 (1) + MBY (2)	288.15	2.8942	-0.1860	0.7090	1.2020	0.0060
		293.15	3.0255	-0.1348	0.7794	1.2100	0.0232
		303.15	3.3178	-0.0244	0.9425	1.2908	0.0070
		313.15	3.6537	0.1017	1.1021	1.3882	0.0252
		323.15	4.0364	0.2513	1.2651	1.4775	0.0148
	$[P_{66614}][Im]$ (1) + MBY (2)	293.15	-335.2242	219.1452	-302.1621	312.3125	1.8
		303.15	-384.5375	261.1295	-351.1402	349.9049	2.0
		313.15	-444.3628	307.9305	-409.8591	402.2742	2.3
		323.15	-515.9839	364.8399	-480.4435	465.7558	2.7
		333.15	-601.8864	434.4542	-564.2577	535.9752	3.1
$\Delta\kappa_s$ ($T\ Pa^{-1}$)	$[P_{66614}][4\text{-MeIm}]$ (1) + MBY (2)	293.15	-334.1512	223.3245	-294.7163	286.5856	1.5
		303.15	-382.9518	264.2071	-342.1041	324.1170	1.7

	313.15	-441.9541	312.3271	-398.3531	371.0765	1.9
	323.15	-512.8983	370.3049	-464.3581	425.0854	2.2
	333.15	-597.4178	435.4577	-547.6517	502.4170	2.7
$[P_{66614}][4\text{-BrIm}]$ (1) + MBY (2)	293.15	-315.8316	221.3862	-276.3503	241.2339	1.3
	303.15	-363.6954	259.5190	-319.7078	281.5303	1.6
	313.15	-421.7489	305.1432	-372.3452	327.5370	1.9
	323.15	-491.6803	359.7076	-432.4266	378.6683	2.3
	333.15	-575.1890	422.8147	-508.7166	451.8303	2.8
CCl_4 (1) + MBY (2)	288.15	232.2399	28.2108	48.2129	67.9694	0.4
	293.15	245.4634	34.7951	50.4404	50.1849	0.3
	303.15	275.1638	40.5481	54.4101	48.1314	0.3
	313.15	308.2508	46.3127	61.1880	43.6741	0.3
	323.15	349.1144	56.7876	65.3604	38.2224	0.4

Table S5. The interaction parameters (g_{ji}-g_{ii}) of binary systems obtained from VLE by UNIQUAC equation and from viscosity by Eyring-UNIQUAC equation.

Systems	Data source	T (K)	p (kPa)	(g_{21}-g_{11}) (J mol^{-1})	(g_{12}-g_{22}) (J mol^{-1})	Refs.
[C_8mim][BTI] (1) + ethyl acetate (2)	viscosity	298.15		362.0	2685.7	[11]
[C_8mim][BTI] (1) + ethanol (2)	viscosity	298.15		1659.8	3930.5	[11]
[C_8mim][BTI] (1) + methyl acetate (2)	viscosity	298.15		796.4	3519.2	[11]
[C_8mim][BTI] (1) + methanol (2)	viscosity	298.15		5360.7	8532.8	[11]
[C_8mim][BTI] (1) + isopropyl acetate (2)	viscosity	298.15		349.8	2131.7	[11]
[C_8mim][BTI] (1) + 2-propanol (2)	viscosity	298.15		554.4	2491.6	[11]
[C_4mim][BTI] (1) + ethyl acetate (2)	viscosity	298.15		434.1	2031.6	[11]
[C_4mim][BTI] (1) + ethanol (2)	viscosity	298.15		1161.8	3150.1	[11]
[C_2mim][$EtSO_4$] (1) + ethanol (2)	viscosity	298.15		-35.5	2866.2	[11]
[C_2mim][$EtSO_4$] (1) + 1-propanol (2)	viscosity	298.15		-1227.6	2772.7	[11]
[C_2mim][$EtSO_4$] (1) + 2-propanol (2)	viscosity	298.15		-646.4	2256.0	[11]
[C_2mim][Tf_2N] (1) + water (2)	VLE	353.15		2903.7	67.1	[12]
[C_2mim][Tf_2N] (1) + 2-propanol (2)	VLE	353.15		-292.1	1534.9	[12]
[C_2mim][Tf_2N] (1) + acetone (2)	VLE	353.15		1392.5	-1392.5	[12]
[C_4mim][Tf_2N] (1) + water (2)	VLE	353.15		3054.2	345.0	[12]
[C_4mim][Tf_2N] (1) + 2-propanol (2)	VLE	353.15		-156.1	1199.6	[12]
[C_4mim][Tf_2N] (1) + acetone (2)	VLE	353.15		1367.5	-1367.5	[12]
Dimethyl carbonate (1) + methanol (2)	VLE		101.3	2192.62	150.27	
Dimethyl carbonate (1) + ethanol (2)	VLE		101.3	570.86	768.20	
Dimethyl carbonate (1) + 1-propanol (2)	VLE		101.3	1032.59	3.18	
Dimethyl carbonate (1) + 1-butanol (2)	VLE		101.3	1083.42	-560.83	
Dimethyl carbonate (1) + 1-pentanol (2)	VLE		101.3	740.15	-148.47	

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