

Supporting Information

Densities, heat capacities, viscosities, ^1H - and ^{13}C -NMR spectra, and solvatochromic parameters of binary mixtures of 1,3-dimethyl-1,3-diazinan-2-one (DMPU) and water

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Densities

Table S1. Density (ρ /g·cm⁻³) as function of temperature, T , and DMPU mole fraction, x_{DMPU} , for the binary mixtures of DMPU + water, at atmospheric pressure (0.1 MPa), determined at the University of Aveiro.

x_{DMPU}	T / K												
	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.9984	1.06290	1.05880	1.05460	1.05050	1.04630	1.04220	1.03800	1.03390	1.02970	1.02550	1.02140	1.01720	1.01290
0.9984	1.06373	1.05916	1.05493	1.05075	1.04658	1.04242	1.03825	1.03410	1.02995	1.02580	1.02183	1.01752	1.01337
0.9984	1.06368	1.05918	1.05495	1.05077	1.04660	1.04244	1.03827	1.03411	1.02996	1.02582	1.02167	1.01752	1.01337
0.9831	1.06387	1.05958	1.05534	1.05115	1.04698	1.04280	1.03863	1.03446	1.03046	1.02614	1.02198	1.01782	1.01365
0.9554	1.06568	1.06116	1.05691	1.05272	1.04852	1.04434	1.04015	1.03596	1.03178	1.02760	1.02343	1.01925	1.01507
0.9357	1.06619	1.06158	1.05732	1.05311	1.04890	1.04470	1.04050	1.03629	1.03210	1.02790	1.02371	1.01952	1.01532
0.8669	1.06787	1.06329	1.05900	1.05474	1.05049	1.04624	1.04199	1.03773	1.03346	1.02921	1.02495	1.02068	1.01640
0.7970	1.06973	1.06512	1.06078	1.05648	1.05217	1.04787	1.04355	1.03922	1.03489	1.03056	1.02622	1.02187	1.01751
0.6917	1.07265	1.06788	1.06345	1.05905	1.05466	1.05025	1.04583	1.04139	1.03694	1.03249	1.02805	1.02371	1.01921
0.6498	1.07351	1.06900	1.06454	1.06011	1.05568	1.05123	1.04682	1.04230	1.03780	1.03330	1.02893	1.02427	1.01970
0.5809	1.07546	1.07070	1.06619	1.06171	1.05721	1.05270	1.04816	1.04362	1.03903	1.03445	1.02985	1.02523	1.02058
0.5081	1.07702	1.07239	1.06782	1.06326	1.05869	1.05410	1.04948	1.04484	1.04018	1.03551	1.03083	1.02611	1.02137
0.4616	1.07793	1.07327	1.06867	1.06406	1.05945	1.05481	1.05015	1.04546	1.04075	1.03602	1.03128	1.02650	1.02171
0.4076	1.07870	1.07400	1.06940	1.06470	1.06000	1.05530	1.05060	1.04580	1.04100	1.03620	1.03140	1.02650	1.02160
0.3609	1.07860	1.07400	1.06930	1.06460	1.05990	1.05520	1.05050	1.04570	1.04090	1.03610	1.03130	1.02640	1.02140
0.2978	1.07740	1.07270	1.06810	1.06340	1.05880	1.05410	1.04940	1.04470	1.03990	1.03510	1.03020	1.02530	1.02040
0.2471	1.07470	1.07020	1.06550	1.06080	1.05620	1.05150	1.04680	1.04210	1.03730	1.03240	1.02760	1.02260	1.01770
0.2063			1.06230	1.05770	1.05320	1.04860	1.04400	1.03930	1.03460	1.02990	1.02510	1.02020	1.01530
0.1739	1.06670	1.06230	1.05800	1.05370	1.04930	1.04490	1.04040	1.03590	1.03130	1.02670	1.02200	1.01740	1.01260
0.1461	1.06190	1.05790	1.05380	1.04970	1.04550	1.04120	1.03690	1.03250	1.02810	1.02360	1.01890	1.01440	1.00970
0.1228	1.05520	1.05140	1.04750	1.04360	1.03960	1.03560	1.03160	1.02740	1.02320	1.01890	1.01460	1.01020	1.00570
0.08573	1.04300	1.03960	1.03630	1.03290	1.02940	1.02580	1.02210	1.01840	1.01460	1.01060	1.00660	1.00240	0.99840
0.05680	1.03020	1.02740	1.02460	1.02170	1.01860	1.01560	1.01220	1.00890	1.00540	1.00160	0.99790	0.99350	0.98910
0.02868			1.01150	1.00910	1.00670	1.00410	1.00150	0.99840	0.99490	0.99120	0.98750	0.98340	0.97860
0.01545	1.00710	1.00540	1.00360	1.00160	0.99960	0.99720	0.99460	0.99200	0.98910	0.98590	0.98220	0.97860	0.97440
0.007139	1.00380	1.00200	1.00030	0.99830	0.99620	0.99390	0.99130	0.98840	0.98570	0.98280	0.97960	0.97650	0.97230
0	0.99821	0.99705	0.99565	0.99403	0.99222	0.99021	0.98804	0.98569	0.98320	0.98055	0.97776	0.97484	0.97179

Standard uncertainties (u) and relative standard uncertainty (u_r) are: $u(\rho) = 0.00005$ g cm⁻³, $u(T) = 0.02$ K, $u(p) = 0.003$ MPa and $u_r(x) = 0.01\%$.

Table S2. Experimental relative volumetric heat capacities ($(\sigma - \sigma^0)/\sigma^0$), densities (ρ), specific heat capacities (c_p) and apparent molar isobaric heat capacities ($C_{p,\phi}$) of DMPU in water and of water in DMPU, DMPU mole fraction, x_{DMPU} , DMPU molality, m_{DMPU} , and the number of independent measurements, n , determined at Murdoch, at 298.15 K and atmospheric pressure ($p = 0.1$ MPa).

m_{DMPU}/m^0	x_{DMPU}	n^a	$10^3 (\sigma - \sigma^0)/\sigma^0$	$\rho / (\text{g}\cdot\text{cm}^{-3})$	$c_p / (\text{J}\cdot\text{K}^{-1}\cdot\text{g}^{-1})$	$C_{p,\phi,\text{DMPU}} / (\text{J}\cdot\text{K}^{-1}\cdot\text{mol}^{-1})$ DMPU in water	$C_{p,\phi,w} / (\text{J}\cdot\text{K}^{-1}\cdot\text{mol}^{-1})$ water in DMPU
0	0			0.997047	4.1819		
0.4101	0.007334	6	-8.09	1.0011	4.131 ^b	402.9	76.6
0.8643	0.01533	4	-18.39	1.0058	4.069 ^b	383.7	77.8
1.948	0.03390	2	-44.48	1.0164	3.920 ^b	365.1	80.2
3.334	0.05666	2	-75.76	1.0281	3.748 ^b	349.5	82.7
3.347	0.05687	2	-76.80	1.0281	3.744 ^b	349.6	82.7
5.212	0.08583	2	-114.57	1.0403	3.549 ^b	333.4	85.3
7.812	0.1234	2	-156.34	1.0522	3.343 ^b	315.9	87.8
11.33	0.1695	4	-221.87	1.0624 ^h	3.054 ^c	298.5	89.9
11.64	0.1733	4	-209.87	1.0629	3.037 ^c	297.2	90.0
11.67	0.1737	2	-212.00	1.0628	3.091 ^b	297.1	90.0
14.69	0.2093	8	-246.27	1.0678	2.94 ^b	286.4	91.0
14.69	0.2093	6	-260.10	1.0678	2.89 ^c	286.4	91.0
17.92	0.2441	2	-253.43	1.0707	2.91 ^b	277.7	91.7
18.19	0.2469	4	-293.96	1.0710	2.75 ^c	277.1	91.7
18.19	0.2469	4	-291.76	1.0710	2.76 ^d	277.1	91.7
18.36	0.2485	6	-288.14	1.0712	2.77 ^c	276.7	91.7
23.48	0.2973	8	-302.98	1.0734	2.71 ^b	267.3	92.3
23.48	0.2973	4	-328.44	1.0734	2.61 ^c	267.3	92.3
31.33	0.3608	3	-368.00	1.0742 ^h	2.45 ^c	258.4	93.0
40.00	0.4188	6	-363.50	1.0740	2.47 ^c	252.8	93.8
40.00	0.4188	6	-362.27	1.0740	2.48 ^e	252.8	93.8
70.30	0.5588	1	-454.87	1.0711 ^h	2.12 ^c	244.9	97.9
70.30	0.5588	4	-451.37	1.0711 ^h	2.14 ^f	244.9	97.9
70.30	0.5588	3	-443.89	1.0711 ^h	2.17 ^g	244.9	97.9
146.0	0.7245	4	-496.88	1.0666 ^h	1.97 ^f	238.8	106.0
475.6	0.8955	4	-527.42	1.0619	1.85 ^f	231.3	111.5
3555	0.9846	6	-549.42	1.0598	1.77 ^e	227.6	108.0
64474	0.9991	6	-548.95	1.0592	1.77 ^f	227.1	96.7

^a Number of independent measurements; ^b using peristaltic pump with water reference; ^c using syringe burettes with water reference; ^{d-g} using syringe burettes with (DMPU + water) reference with $w_{\text{DMPU}} = 0.60, 0.70, 0.80$ and, 0.84 , respectively, ^h interpolated. Standard uncertainties (u) and relative standard uncertainties (u_r) are: $u(\rho) = 0.0005 \text{ g}\cdot\text{cm}^{-3}$, $u(c_p) | x_{\text{DMPU}} < 0.15 = 0.006 \text{ J}\cdot\text{K}^{-1}\cdot\text{g}^{-1}$, $u(c_p) | x_{\text{DMPU}} > 0.15 = 0.05 \text{ J}\cdot\text{K}^{-1}\cdot\text{g}^{-1}$, $u_r((\sigma - \sigma^0)/\sigma^0) = 0.09 \times 10^{-3}$, $u_r(C_{p,\phi}) = 0.1 \text{ J}\cdot\text{K}^{-1}\cdot\text{mol}^{-1}$, $u_r(m_{\text{DMPU}}/m^0) = 0.0005$, $u(T) = 0.02 \text{ K}$, $u(p) = 0.003 \text{ MPa}$, $m^0 = 1 \text{ mol}\cdot\text{kg}^{-1}$ and $u_r(x) = 0.01\%$.

Table S3. Literature information regarding temperature (T), density (ρ), water content, purification procedure and method of density measurements, of pure DMPU.

Authors	T / K	$\rho / (\text{g} \cdot \text{cm}^{-3})$	Water content	Purification procedure	Method of density determination
Rosenfarb <i>et al.</i> , (1976) [1]	298.15	1.0596	not specified	distilled at 354 K (370 Pa) over BaO	pycnometry and dilatometry
	308.15	1.0512			
	318.15	1.0428			
	328.15	1.0344			
	348.15	1.0181			
	373.15	0.9970			
Kneisl and Zondlo (1987) [2]	298.15	1.0593	not specified	distilled at 660 Pa through a column of BaO	pycnometry
	308.15	1.0509			
	313.15	1.0467			
	318.15	1.0426			
	333.15	1.0301			
	353.15	1.0135			
373.15	0.9969				
Stroka <i>et al.</i> , (1990) [3]	298.15	1.0600	not specified	distilled at 368 K (550 Pa) over BaO	pycnometry
Lemos and Maestre (1995) [4]	278.15	1.0747	less than 0.03%	dried over activated 0.3 nm molecular sieves	vibrating tube densimetry
	288.15	1.0668			
	298.15	1.0592			
	308.15	1.0507			
	318.15	1.0425			
López <i>et al.</i> , (1999) [5]	298.15	1.0595	not specified	dried over activated 0.4 nm molecular sieves	vibrating tube densimetry
Székely and Jancsó (2009) [6]	288.15	1.0678	not specified	purity $\geq 99\%$; not further purified	vibrating tube densimetry
	298.15	1.0594			
	313.15	1.0470			
Batov and Ivanov (2011) [7]	298.15	1.0601	$\approx 0.005\%$	dried over activated 0.3 nm molecular sieves; distilled at 340 K (267 Pa)	vibrating tube densimetry
Ivanov <i>et al.</i> , (2011) [8]	278.15	1.0762	$\approx 0.005\%$	dried over activated 0.3 nm molecular sieves; distilled at 340 K (267 Pa)	vibrating tube densimetry
	288.15	1.0678			
	298.15	1.0595			
	308.15	1.0511			
	313.15	1.0470			
	318.15	1.0428			
Bai <i>et al.</i> , (2015) [9]	288.15	1.0696	less than 0.03%	distilled; dried over activated 0.4 nm molecular sieves	vibrating tube densimetry
	298.15	1.0612			
	308.15	1.0528			
	318.15	1.0444			
Li <i>et al.</i> , (2016) [10]	293.15	1.065	0.001	Not specified	vibrating tube densimetry

Table S4. Excess molar volumes ($\text{cm}^3 \cdot \text{mol}^{-1}$) of DMPU/water mixtures as function of DMPU mole fraction, x_{DMPU} , and temperature, T .

x_{DMPU}	T / K												
	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.9984	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
0.9984	-0.096	-0.043	-0.040	-0.031	-0.034	-0.027	-0.031	-0.025	-0.031	-0.038	-0.054	-0.041	-0.060
0.9984	-0.090	-0.045	-0.042	-0.033	-0.037	-0.030	-0.033	-0.026	-0.033	-0.040	-0.034	-0.041	-0.060
0.9831	-0.127	-0.106	-0.101	-0.091	-0.094	-0.085	-0.089	-0.081	-0.104	-0.090	-0.083	-0.089	-0.105
0.9554	-0.352	-0.306	-0.301	-0.291	-0.291	-0.283	-0.285	-0.275	-0.278	-0.282	-0.274	-0.278	-0.294
0.9357	-0.422	-0.367	-0.360	-0.348	-0.347	-0.336	-0.337	-0.325	-0.327	-0.327	-0.318	-0.320	-0.333
0.8669	-0.643	-0.593	-0.582	-0.564	-0.558	-0.541	-0.536	-0.519	-0.511	-0.506	-0.490	-0.483	-0.486
0.7970	-0.859	-0.807	-0.790	-0.768	-0.755	-0.734	-0.720	-0.697	-0.683	-0.670	-0.646	-0.632	-0.626
0.6917	-1.144	-1.081	-1.055	-1.024	-1.003	-0.973	-0.950	-0.918	-0.894	-0.870	-0.839	-0.826	-0.808
0.6498	-1.218	-1.176	-1.147	-1.114	-1.089	-1.056	-1.035	-0.997	-0.969	-0.942	-0.918	-0.877	-0.853
0.5809	-1.361	-1.302	-1.269	-1.233	-1.202	-1.165	-1.134	-1.096	-1.062	-1.029	-0.989	-0.954	-0.925
0.5081	-1.453	-1.404	-1.366	-1.324	-1.289	-1.248	-1.211	-1.168	-1.131	-1.094	-1.050	-1.010	-0.976
0.4616	-1.495	-1.444	-1.404	-1.360	-1.322	-1.279	-1.240	-1.195	-1.156	-1.115	-1.070	-1.028	-0.991
0.4076	-1.518	-1.465	-1.425	-1.376	-1.333	-1.287	-1.247	-1.197	-1.153	-1.111	-1.064	-1.016	-0.975
0.3609	-1.493	-1.446	-1.399	-1.351	-1.308	-1.263	-1.223	-1.175	-1.132	-1.091	-1.046	-1.001	-0.955
0.2978	-1.415	-1.362	-1.319	-1.271	-1.232	-1.188	-1.148	-1.107	-1.065	-1.025	-0.978	-0.935	-0.897
0.2471	-1.294	-1.248	-1.200	-1.152	-1.112	-1.067	-1.027	-0.986	-0.945	-0.901	-0.860	-0.814	-0.777
0.2063			-1.083	-1.037	-1.000	-0.959	-0.922	-0.881	-0.844	-0.809	-0.769	-0.729	-0.692
0.1739	-1.032	-0.986	-0.949	-0.912	-0.877	-0.842	-0.808	-0.773	-0.740	-0.708	-0.672	-0.643	-0.611
0.1461	-0.908	-0.872	-0.839	-0.807	-0.776	-0.744	-0.715	-0.683	-0.655	-0.626	-0.590	-0.565	-0.536
0.1228	-0.747	-0.715	-0.684	-0.656	-0.629	-0.603	-0.581	-0.555	-0.531	-0.507	-0.484	-0.460	-0.437
0.08573	-0.511	-0.484	-0.465	-0.445	-0.427	-0.409	-0.391	-0.375	-0.360	-0.341	-0.324	-0.305	-0.294
0.05680	-0.311	-0.293	-0.280	-0.268	-0.254	-0.246	-0.232	-0.222	-0.211	-0.195	-0.183	-0.157	-0.133
0.02868			-0.127	-0.119	-0.115	-0.111	-0.109	-0.099	-0.084	-0.068	-0.053	-0.032	0.002
0.01545	-0.054	-0.048	-0.046	-0.043	-0.044	-0.040	-0.035	-0.034	-0.029	-0.021	-0.005	0.007	0.028
0.007139	-0.049	-0.040	-0.037	-0.032	-0.028	-0.025	-0.018	-0.009	-0.007	-0.003	0.004	0.006	0.028

Standard uncertainties (u) and relative standard uncertainty (u_r) are: $u(T) = 0.02 \text{ K}$ and $u_r(x) = 0.01\%$

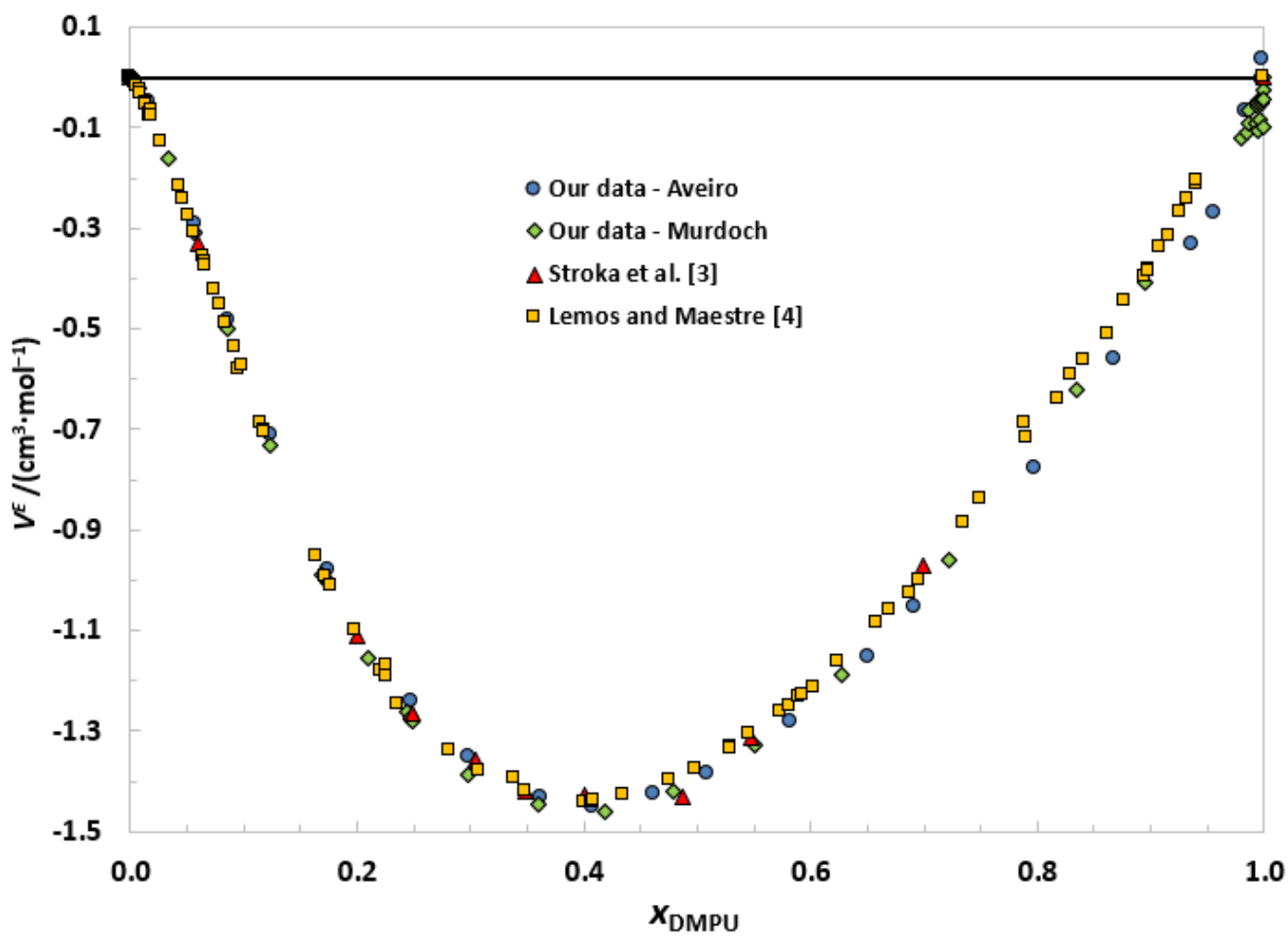


Figure S1. Comparison of the excess molar volumes of DMPU/water mixtures obtained in this work and reported in the literature [3,4] at 298.15 K.

Table S5. Apparent molar volumes ($V_{\phi,\text{DMPU}}/\text{cm}^3\cdot\text{mol}^{-1}$) of DMPU (solute) in water as a function of DMPU mole fraction, x_{DMPU} , and temperature, T .

x_{DMPU}	T / K												
	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.9984	120.5	121.0	121.5	122.0	122.5	123.0	123.5	123.9	124.5	125.0	125.5	126.0	126.5
0.9831	120.5	120.9	121.4	121.9	122.4	122.9	123.4	123.9	124.4	124.9	125.4	125.9	126.4
0.9554	120.2	120.7	121.2	121.7	122.2	122.7	123.2	123.7	124.2	124.7	125.2	125.7	126.2
0.9357	120.1	120.7	121.1	121.6	122.1	122.6	123.1	123.6	124.1	124.6	125.1	125.7	126.2
0.8669	119.8	120.4	120.9	121.4	121.9	122.4	122.9	123.4	123.9	124.4	124.9	125.4	126.0
0.7970	119.5	120.0	120.5	121.0	121.6	122.1	122.6	123.1	123.6	124.1	124.7	125.2	125.8
0.6917	118.9	119.5	120.0	120.5	121.0	121.6	122.1	122.6	123.2	123.7	124.3	124.8	125.4
0.6498	118.7	119.2	119.8	120.3	120.8	121.4	121.9	122.4	123.0	123.5	124.1	124.7	125.2
0.5809	118.2	118.8	119.3	119.9	120.4	121.0	121.5	122.1	122.6	123.2	123.8	124.4	124.9
0.5081	117.7	118.3	118.8	119.4	120.0	120.5	121.1	121.7	122.2	122.8	123.4	124.0	124.6
0.4616	117.3	117.9	118.5	119.1	119.6	120.2	120.8	121.4	122.0	122.6	123.2	123.8	124.4
0.4076	116.9	117.5	118.0	118.6	119.2	119.8	120.4	121.0	121.6	122.3	122.9	123.5	124.1
0.3609	116.4	117.0	117.7	118.3	118.9	119.5	120.1	120.7	121.3	122.0	122.6	123.2	123.9
0.2978	115.8	116.5	117.1	117.7	118.4	119.0	119.6	120.3	120.9	121.5	122.2	122.9	123.5
0.2471	115.3	116.0	116.7	117.3	118.0	118.7	119.3	120.0	120.6	121.3	122.0	122.7	123.4
0.2063			116.3	117.0	117.7	118.3	119.0	119.7	120.4	121.1	121.8	122.5	123.2
0.1739	114.7	115.4	116.1	116.8	117.5	118.1	118.8	119.5	120.2	120.9	121.6	122.3	123.0
0.1461	114.4	115.1	115.8	116.5	117.2	117.9	118.6	119.3	120.0	120.7	121.4	122.1	122.9
0.1228	114.5	115.2	116.0	116.7	117.4	118.1	118.7	119.5	120.1	120.9	121.5	122.3	123.0
0.08573	114.6	115.4	116.1	116.8	117.5	118.2	118.9	119.6	120.3	121.0	121.7	122.4	123.1
0.05680	115.1	115.9	116.6	117.3	118.0	118.6	119.4	120.1	120.8	121.6	122.3	123.2	124.2
0.02868			117.1	117.9	118.5	119.1	119.7	120.5	121.5	122.6	123.6	124.9	126.6
0.01545	117.1	117.9	118.6	119.2	119.7	120.4	121.2	121.8	122.6	123.6	125.2	126.4	128.4
0.007139	113.6	115.4	116.4	117.5	118.5	119.5	120.9	122.7	123.5	124.5	126.0	126.9	130.5

Standard uncertainties (u) and relative standard uncertainty (u_r) are: $u(T) = 0.02 \text{ K}$ and $u_r(x) = 0.01\%$.

Table S6. Apparent molar volumes ($V_{\phi,\text{water}}/\text{cm}^3\cdot\text{mol}^{-1}$) of water (solute) in DMPU as function of water mole fraction, x_{water} , and temperature, T .

x_{water}	T / K												
	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.9929	18.00	18.03	18.06	18.09	18.13	18.17	18.21	18.27	18.32	18.37	18.43	18.49	18.57
0.9845	17.99	18.02	18.05	18.08	18.11	18.15	18.20	18.24	18.29	18.35	18.42	18.49	18.57
0.9713			17.96	18.00	18.04	18.08	18.12	18.17	18.24	18.30	18.37	18.45	18.54
0.9432	17.72	17.76	17.80	17.84	17.89	17.93	17.99	18.04	18.10	18.17	18.23	18.32	18.40
0.9143	17.49	17.54	17.59	17.64	17.69	17.75	17.81	17.87	17.93	18.00	18.07	18.15	18.22
0.8772	17.20	17.26	17.32	17.38	17.44	17.51	17.57	17.65	17.72	17.80	17.88	17.96	18.05
0.8539	16.99	17.05	17.12	17.18	17.25	17.33	17.40	17.48	17.56	17.64	17.74	17.82	17.92
0.8261	16.81	16.88	16.95	17.02	17.10	17.18	17.26	17.34	17.43	17.52	17.62	17.71	17.81
0.7937			16.74	16.82	16.90	16.99	17.08	17.17	17.26	17.36	17.46	17.57	17.68
0.7529	16.35	16.42	16.51	16.60	16.69	16.78	16.88	16.97	17.07	17.18	17.29	17.41	17.52
0.7022	16.06	16.14	16.23	16.32	16.41	16.51	16.61	16.71	16.81	16.92	17.04	17.16	17.28
0.6391	15.75	15.82	15.92	16.02	16.12	16.23	16.33	16.45	16.56	16.68	16.80	16.93	17.07
0.5924	15.53	15.61	15.71	15.81	15.92	16.03	16.14	16.27	16.39	16.51	16.65	16.78	16.92
0.5384	15.32	15.41	15.51	15.61	15.72	15.83	15.95	16.07	16.19	16.32	16.46	16.59	16.73
0.4919	15.16	15.24	15.34	15.45	15.56	15.68	15.79	15.92	16.05	16.18	16.32	16.45	16.59
0.4191	14.88	15.00	15.10	15.21	15.32	15.44	15.56	15.68	15.82	15.95	16.11	16.24	16.39
0.3502	14.68	14.76	14.87	14.98	15.09	15.21	15.32	15.46	15.59	15.73	15.86	16.02	16.17
0.3083	14.47	14.62	14.73	14.85	14.95	15.08	15.20	15.34	15.47	15.61	15.77	15.86	16.01
0.2030	14.06	14.20	14.31	14.42	14.53	14.65	14.77	14.91	15.04	15.17	15.35	15.47	15.61
0.1331	13.61	13.80	13.89	14.02	14.12	14.24	14.34	14.49	14.62	14.73	14.93	15.02	15.14
0.06433	12.37	12.77	12.87	13.00	13.09	13.23	13.30	13.46	13.54	13.65	13.90	13.88	13.93
0.04461	11.47	11.81	11.92	12.03	12.11	12.24	12.30	12.46	12.53	12.60	12.89	12.81	12.79
0.01686	14.07	13.45	13.62	13.91	13.89	14.20	14.18	14.46	13.33	14.48	15.15	14.76	14.60
0.001574	16.94	17.01	17.08	17.15	17.21	17.28	17.35	17.42	17.49	17.56	17.63	17.71	17.78

Standard uncertainties (u) and relative standard uncertainty (u_r) are: $u(T) = 0.02 \text{ K}$ and $u_r(x) = 0.01\%$.

Viscosity

Table S7. Dynamic viscosities (η /mPa·s) of the mixtures of DMPU (x_{DMPU}) and water, at atmospheric pressure (0.1 MPa) and temperatures (T) from 293.15 to 353.15 K.

x_{DMPU}	T / K												
	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15	338.15	343.15	348.15	353.15
0.9984	3.2780	2.9170	2.6090	2.3480	2.1260	1.9370	1.7770	1.6340	1.5100	1.3990	1.2980	1.2120	1.1350
0.9984	3.300	2.937	2.628	2.365	2.141	1.950	1.784	1.641	1.516	1.407	1.312	1.228	1.152
0.9831	3.401	3.022	2.700	2.425	2.191	1.992	1.819	1.670	1.536	1.423	1.324	1.236	1.159
0.9554	3.604	3.187	2.834	2.533	2.283	2.071	1.886	1.726	1.585	1.463	1.355	1.259	1.174
0.9357	3.761	3.326	2.957	2.647	2.384	2.160	1.968	1.803	1.658	1.531	1.420	1.321	1.233
0.8669	4.170	3.644	3.203	2.837	2.531	2.274	2.055	1.868	1.704	1.564	1.441	1.332	1.237
0.7970	4.745	4.102	3.571	3.134	2.772	2.468	2.219	2.003	1.817	1.657	1.517	1.396	1.289
0.6917	5.824	4.954	4.245	3.672	3.202	2.817	2.499	2.234	2.013	1.820	1.654	1.507	1.380
0.6498	6.342	5.325	4.532	3.892	3.370	2.946	2.593	2.306	2.063	1.857	1.674	1.526	1.394
0.5809	7.345	6.105	5.129	4.357	3.750	3.257	2.855	2.522	2.243	2.009	1.812	1.643	1.497
0.5081	8.518	6.965	5.768	4.838	4.110	3.531	3.064	2.684	2.371	2.110	1.890	1.705	1.546
0.4616	9.277	7.512	6.161	5.130	4.326	3.692	3.185	2.776	2.440	2.162	1.930	1.735	1.568
0.3968	10.078	7.993	6.483	5.332	4.450	3.744	3.215	2.780	2.425	2.126	1.895	1.697	1.521
0.3486	10.683	8.424	6.796	5.568	4.626	3.900	3.327	2.867	2.481	2.180	1.931	1.726	1.550
0.2961	10.817	8.492	6.828	5.569	4.616	3.878	3.296	2.831	2.456	2.150	1.897	1.688	1.514
0.2453	10.242	8.068	6.471	5.287	4.386	3.685	3.139	2.706	2.351	2.055	1.817	1.616	1.450
0.2062	9.318	7.376	5.943	4.870	4.051	3.414	2.910	2.509	2.168	1.897	1.705	1.522	1.366
0.1745	8.375	6.615	5.342	4.401	3.676	3.114	2.651	2.290	1.998	1.762	1.555	1.480	1.305
0.1461	6.909	5.559	4.568	3.799	3.201	2.728	2.350	2.042	1.787	1.581	1.411	1.267	1.146
0.1227	5.656	4.614	3.836	3.224	2.741	2.354	2.041	1.787	1.577	1.405	1.260	1.141	1.042
0.08573	3.965	3.285	2.774	2.371	2.050	1.791	1.579	1.398	1.256	1.137	1.038	0.953	0.872
0.05680	2.715	2.295	1.979	1.733	1.538	1.379	1.250	1.134	1.024	0.934	0.853	0.742	0.691
0.02970			1.386	1.236	1.112	1.002	0.902	0.827	0.763	0.679	0.633	0.569	0.530
0.01508	1.435	1.267	1.127	1.012	0.919	0.843	0.776	0.715	0.637	0.587	0.546	0.511	0.484
0.007139	1.207	1.066	0.942	0.848	0.769	0.699	0.639	0.586	0.540	0.503	0.468	0.462	0.430

Standard uncertainties (u) and relative standard uncertainties (u_r) are: $u_r(\eta) = 0.01$, $u(T) = 0.02$ K, $u(p) = 0.003$ MPa and $u_r(x) = 0.01\%$.

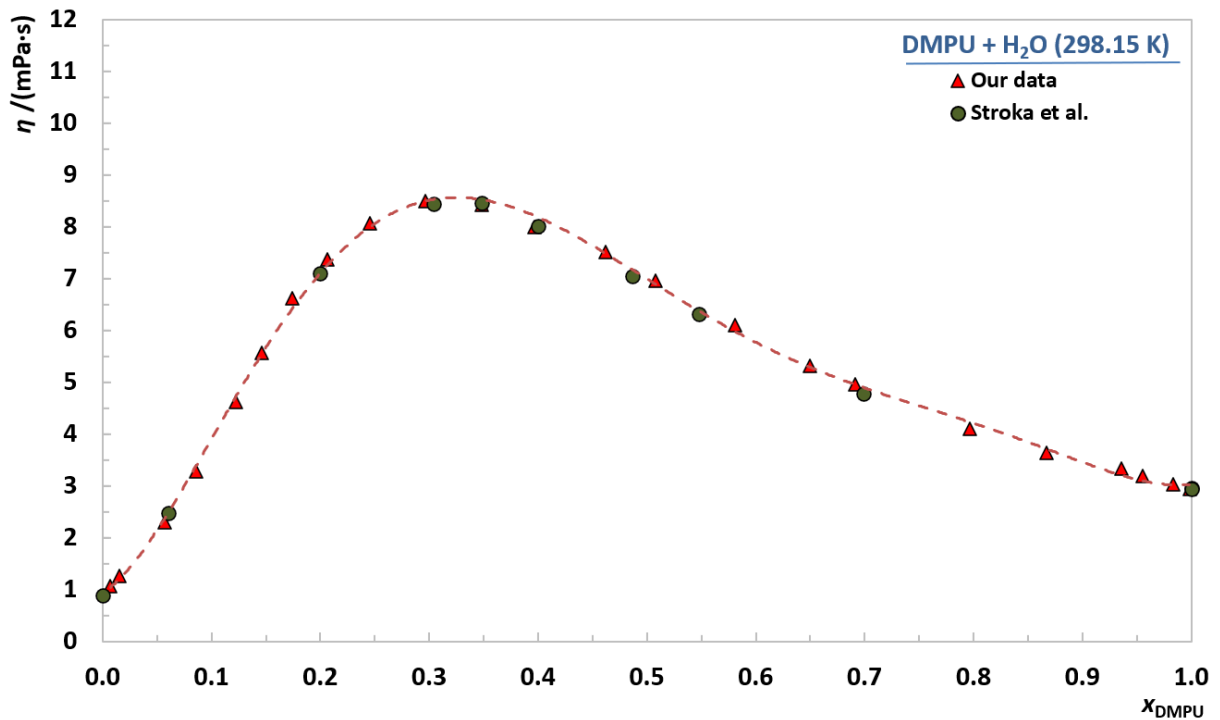


Figure S2. Dynamic viscosities of DMPU + water mixtures at 298.15 K as a function of composition: comparison of present data with those of Stroka *et al.* [3]. The dashed line is a guide for the eye only.

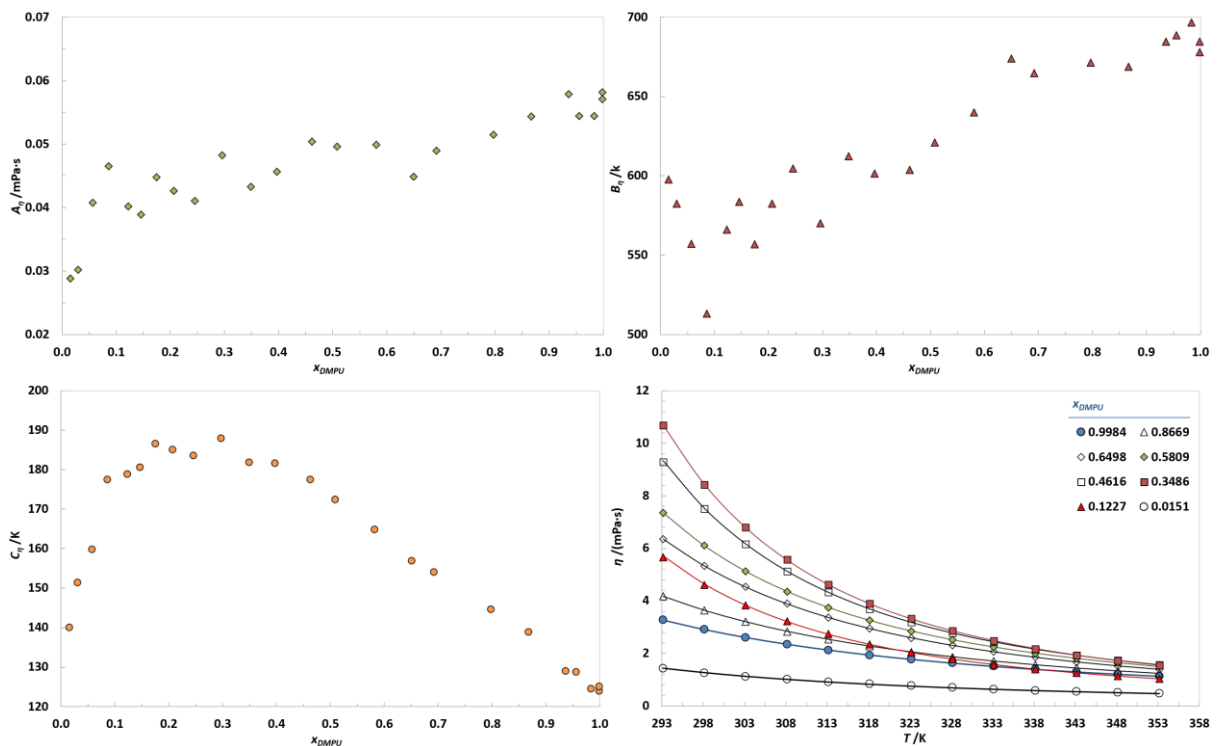


Figure S3. VTF model parameters A_η , B_η and C_η adjusted freely to fit the experimental viscosities of DMPU + water mixtures as a function of temperature.

Table S8. VTF model parameters (A_η , B_η and C_η), %ARD and energy barrier E (at 298.15 K) of the studied DMPU + water mixtures (x_{DMPU}).

x_{DMPU}	$A_\eta / \text{mPa}\cdot\text{s}$	B_η / K	C_η / K	%ARD / %	$E / \text{kJ}\cdot\text{mol}^{-1}$
0.9984	0.0480	617.9	150.5	6.23	18.00
0.9984			150.5	6.39	18.00
0.9831			151.7	6.11	18.24
0.9554			153.4	5.79	18.61
0.9357			155.7	6.76	19.13
0.8669			157.6	5.05	19.57
0.7970			160.9	4.35	20.38
0.6917			166.3	3.37	21.82
0.6498			167.8	2.20	22.24
0.5809			171.4	2.35	23.29
0.5081			174.3	0.97	24.23
0.4616			175.9	0.06	24.73
0.3968			176.6	2.55	24.98
0.3486			177.7	2.82	25.36
0.2961			177.5	3.88	25.27
0.2453			175.8	4.60	24.71
0.2062			172.5	3.92	23.63
0.1745			170.1	6.00	22.90
0.1461			165.2	7.00	21.51
0.1227			159.4	7.31	20.00
0.08573			147.3	7.30	17.34
0.05680			134.0	6.89	14.99
0.02970			111.1	6.77	11.93
0.01508			101.1	8.78	10.88

Standard uncertainty (u) and relative standard uncertainties (u_r) are: $u_r(x) = 0.01\%$, $u(A_\eta) = 0.005 \text{ mPa}\cdot\text{s}$, $u(B_\eta) = 72.8 \text{ K}$, $u(C_\eta) = 8.3 \text{ K}$ and $u(E) = 2.8 \text{ kJ}\cdot\text{mol}^{-1}$

Nuclear magnetic resonance

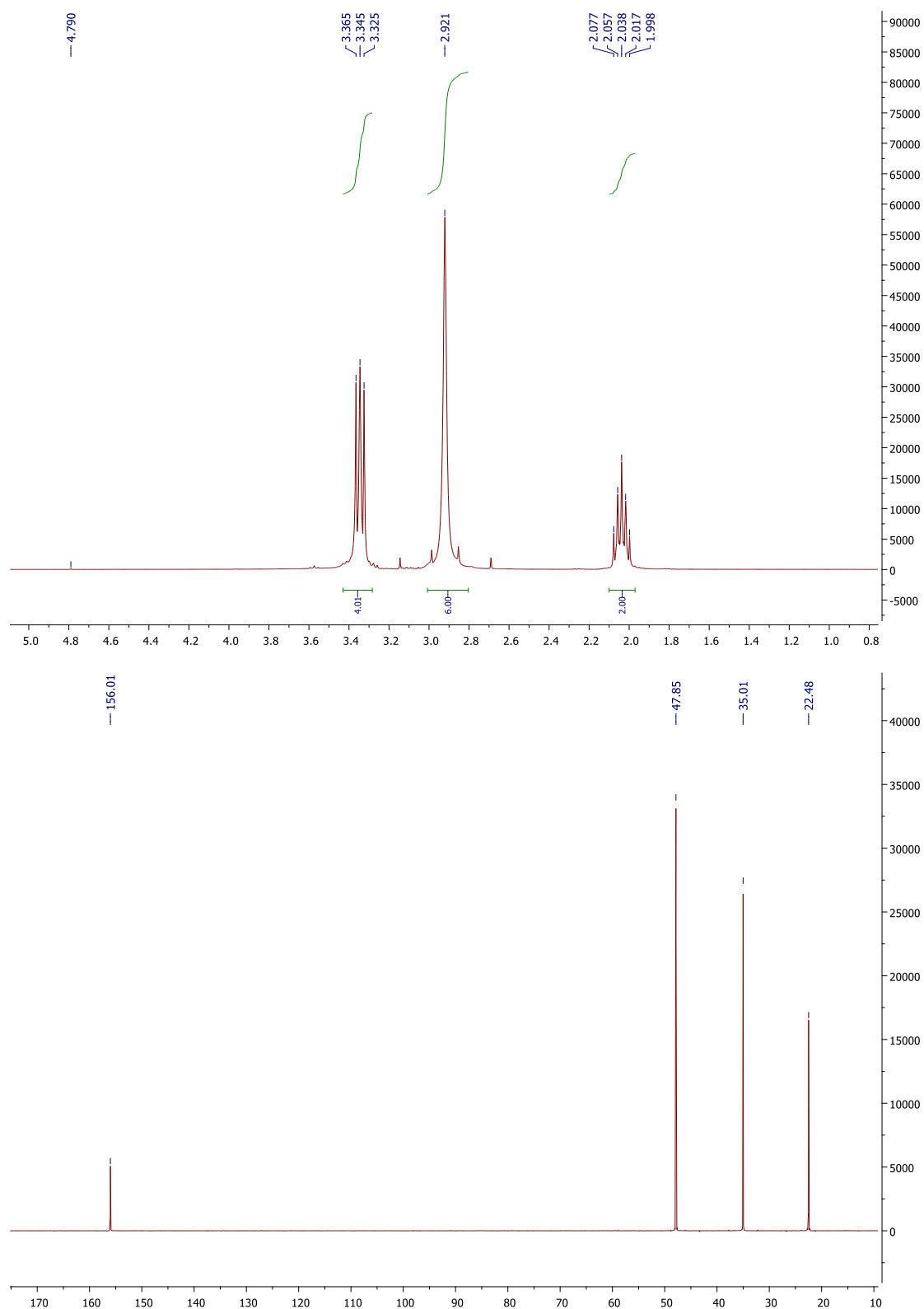


Figure S4. Proton (^1H , top box) and carbon (^{13}C , (bottom box) nuclear magnetic resonance spectra of DMPU (mass fraction purity > 0.99).

Table S9. ^1H NMR chemical shifts (δ), integrals (normalized to the H-5 peak) and coupling constants (J^3) of DMPU + water mixtures at different mole fractions.

x_{DMPU}	H-water (s)		H-1',1'' (s)		H-4,6 (t)			H-5 (p)		
	δ (ppm)	\int (H)	δ (ppm)	\int (H)	δ (ppm)	\int (H)	J (Hz)	δ (ppm)	\int (H)	J (Hz)
0.9984	-	-	2.92	6.06	3.35	3.99	6.04	2.04	2.00	5.97
0.8745	3.66	0.31	2.91	6.09	3.33	3.98	6.04	2.02	2.00	5.97
0.7690	3.76	0.62	2.90	6.05	3.32	3.96	6.06	2.00	2.00	5.97
0.6903	3.84	0.88	2.89	6.14	3.31	4.09	6.04	1.99	2.00	5.97
0.6458	3.88	1.03	2.88	6.33	3.30	4.09	6.05	1.99	2.00	5.97
0.6133	3.94	1.26	2.88	6.21	3.29	4.06	6.08	1.98	2.00	5.95
0.5842	4.01	1.57	2.87	6.19	3.29	4.02	6.05	1.97	2.00	5.96
0.5143	4.07	1.88	2.86	6.31	3.28	4.08	6.05	1.96	2.00	5.96
0.4644	4.15	2.31	2.85	6.25	3.27	4.11	6.05	1.95	2.00	5.96
0.2471	4.50	5.85	2.81	5.71	3.22	3.85	6.04	1.90	2.00	5.95
0.2063	4.56	7.18	2.81	5.78	3.22	3.88	6.03	1.90	2.00	5.95
0.1739	4.66	9.86	2.84	6.11	3.25	4.00	6.03	1.92	2.00	5.95
0.1228	4.66	14.27	2.76	6.06	3.17	3.95	6.01	1.84	2.00	5.96
0.08573	4.66	20.89	2.71	6.08	3.13	3.97	6.00	1.79	2.00	5.97
0.05680	4.66	31.36	2.69	5.93	3.11	4.09	5.99	1.77	2.00	5.97
0.02970	4.62	54.21	2.64	5.73	3.06	3.88	5.99	1.72	2.00	5.97
0.01545	4.66	122.28	2.68	5.79	3.10	3.88	5.98	1.76	2.00	5.97
0.007455	4.66	266.32	2.68	5.79	3.10	4.11	5.97	1.77	2.00	5.97

Relative standard uncertainty (u_r): $u_r(x) = 0.01\%$

Table S10. ^{13}C NMR chemical shifts (δ/ppm) of DMPU + water mixtures at different mole fractions.

x_{DMPU}	C-2	C-4,6	C-1',1''	C-5
0.9984	156.0	47.8	35.0	22.5
0.8745	156.1	47.8	35.0	22.4
0.7690	156.3	47.8	35.1	22.3
0.6903	156.4	47.8	35.1	22.3
0.6458	156.4	47.8	35.1	22.2
0.6133	156.5	47.8	35.1	22.2
0.5842	156.6	47.8	35.2	22.1
0.5143	156.7	47.8	35.2	22.1
0.4644	156.8	47.8	35.2	22.0
0.2471	157.3	47.7	35.3	21.6
0.2063	157.4	47.7	35.3	21.6
0.1739	157.5	47.7	35.3	21.5
0.1228	157.7	47.6	35.3	21.4
0.08573	157.9	47.6	35.3	21.3
0.05680	158.0	47.6	35.3	21.3
0.02970	158.1	47.6	35.3	21.2
0.01545	158.2	47.6	35.3	21.1
0.007455	158.3	47.6	35.3	21.1

Relative standard uncertainty (u_r): $u_r(x) = 0.01\%$

Solvatochromic parameters

The Kamlet–Taft solvatochromic parameters were calculated using the following equations [11]:

$$\pi^* = \frac{\nu_{N,N}(\text{sample}) - \nu_{N,N}(\text{cyclohexane})}{\nu_{N,N}(\text{DMSO}) - \nu_{N,N}(\text{cyclohexane})} \quad (1)$$

$$\beta = \frac{0.76 (\Delta\nu_{N,N}(\text{sample}) - \Delta\nu_{N,N}(\text{cyclohexane}))}{\Delta\nu_{N,N}(\text{DMSO}) - \Delta\nu_{N,N}(\text{cyclohexane})} \quad (2)$$

$$\Delta\nu = \nu_{N,N} - \nu_{4N} \quad (3)$$

$$\nu = 10^{-4} \frac{1}{\lambda_{\text{maxprobe}}} \quad (4)$$

$$\alpha = -0.15 (\delta_4 - \delta_2) + 2.32 \quad (5)$$

where ν is the experimental wavenumber and $\lambda_{\text{maxprobe}}$ is the maximum wavelength of the probe. Subscripts N,N and $4N$ represent the probes N,N -diethyl-4-nitroaniline and 4-nitroaniline, respectively. The subscripts cyclohexane and DMSO indicate the corresponding reference values for these solvents. α was estimated using the ^{13}C NMR chemical shifts, $\delta(\text{C}_i)$ (in ppm), of the carbons atoms of pyridine- N -oxide in positions $i = 2$ and 4 [12].

Table S11. Kamlet-Taft solvatochromic parameters (π^* , β and α) for DMPU + water mixtures (x_{DMPU}), at room temperature and atmospheric pressure (0.1 MPa), along with the standard deviations.

x_{DMPU}	π^*	β	α
0.0000	1.14 [13]	0.49 [13]	1.23 [13]
0.8205	0.91 ± 0.003	0.86 ± 0.004	0.48
0.6633	0.93 ± 0.004	0.84 ± 0.005	0.51
0.4954	0.98 ± 0.001	0.80 ± 0.002	0.60
0.3335	1.04 ± 0.001	0.73 ± 0.003	0.83
0.7891	0.89 ± 0.003	0.86 ± 0.004	
1.0000	0.86 ± 0.002	0.87 ± 0.003	0.46

Standard uncertainty (u) and relative standard uncertainty (u_r) are: $u(p) = 0.003$ MPa and $u_r(x) = 0.01\%$.

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