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

Measuring and PC-SAFT modelling three-phase behaviour

Iago Rodríguez-Palmeiro[1], Oscar Rodríguez[1], Ana Soto[11], Christoph Held[2]
[1]Department of Chemical Engineering, University of Santiago de Compostela, E-15782, Santiago de Compostela (Spain)
[2]Laboratory of Thermodynamics, Department of Biochemical and Chemical Engineering, Technische Universität Dortmund, Emil-Figge-Str. 70, D-44227 Dortmund (Germany)

SUPPLEMENTARY MATERIAL

[1] Corresponding author. E-mail: ana.soto@usc.es

Table S1. Density data (ρ) for IL [C_{12}mim][NTf_2] in the temperature range from 283.15 to 348.15 K and atmospheric pressure

T/K	ρ (g·cm^{-3})
288.15	1.25268
298.15	1.24431
308.15	1.23593
318.15	1.22757
328.15	1.21924
338.15	1.21096
348.15	1.20273

Table S2. Absolute relative deviations (ARD) between PC-SAFT calculations (with parameters used in this work) and experimental vapour pressures (p^S) and liquid densities (ρ^L)

Compound	ARD* p^S	Data Ref.	ARD* ρ^L	T-range (K)	Data Ref.
Water	1.88	[1]	6.83	273-647	[1]
n-Dodecane	2.10	[2]	0.93	263-658	[3]
[C_{10}mim][NTf_2]	-	-	1.62	293-343	[4]
[C_{12}mim][NTf_2]	-	-	1.51	293-343	[This work]

$$* \text{ARD} = 100 \cdot \frac{1}{\text{NP}} \sum_{k=1}^{\text{NP}} \left| \left(1 - \frac{y_k^{\text{PC-SAFT}}}{y_k^{\text{exp}}} \right) \right|$$

Literature

[1] VCI-Wärmeatlas; VDI-Gesellschaft Verfahrenstechnik und Chemieingenieure (GVC): Düsseldorf, Germany, 1994.

[2] N.B. Vargaftik, Tables of Thermophysical Properties of Liquids and Gases, John Wiley and Sons, New York, 1975

[3] American Petroleum Institute Research Project 44. Selected Values of Properties of Hydrocarbons and Related Compounds. Texas A&M University, College Station, Texas, 1973; Table 23-2-(1.101)-d.

[4] A. Nann, J. Mündges, C. Held, S.P. Verevkin G. Sadowski, J. Phys. Chem. B 117 (2013) 3173-3185.

Table S3. CAS number, water content (ω_{H2O}), and experimental and literature values for density (ρ), viscosity (η) and surface tension (γ) of the pure components at 298.15 K and atmospheric pressure.

Compound	CAS number	ω_{H2O} (ppm)	ρ (g·cm^{-3})		η (mPa·s)		γ (mN·m^{-1})	
		Exp	Exp	Lit	Exp	Lit	Exp	Lit
Water	7732-18-5	---	0.99704	0.99705[1]	0.904	0.890[1]	72.0	71.8[1]
n-Dodecane	112-40-3	74	0.74527	0.74518[1]	1.356	1.378[1]	24.9	24.9[1]
[C_{10}mim][NTf_2]	433337-23-6	64	1.27800	1.27830[2]	119.71	119.85[3]	31.1	29.5[3]
								32.2[2]
[C_{12}mim][NTf_2]	404001-48-5	32	1.24404	1.24470[2]	151.42	151.37[3]	31.2	29.8[3]
								32.3[2]

[a] Standard uncertainty: $u(P)$= 5 kPa. For density measurements: $u(T)$ = 0.01 K, $u(\rho)$ = 0.00003 g·cm^{-3}. For viscosity measurements: $u(T)$ = 0.01 K, $u(\eta)$= 0.5%. For surface tension measurements: $u(T)$ = 0.05 K, $u(\gamma)$= 0.1.

Literature

[1] J.A. Riddick, W.B. Bunger, T.K. Sakano, Organic Solvents. Physical Properties and Methods of Purification, 4th ed., John Wiley and Sons, New York, 1986.

[2] H.F.D. Almeida, M.G. Freire, A.M. Fernandes, J.A. Lopes da Silva, P. Morgado, K. Shimizu, E.J.M. Filipe, J.N. Canongia Lopes, L.M.N.B.F. Santos, J.A.P. Coutinho. *Langmuir* 30 (2014) 6408-6418.

[3] C. Kolbeck, J. Lehmann, K. R. J. Lovelock, T. Cremer, N. Paape, P. Wasserscheid, A. P. Fröba, F. Maier, H.P. Steinrück. *J. Phys. Chem. B.* 114 (2010) 17025–17036.

Table S4. Densities and viscosities of involved phases in water/brine + [C_{10}mim][NTf_2] + *n*-dodecane equilibrium at 298.15 K

		H_2O		Brine	
	Phase	ρ (g/cm^3)	η (mPa.s)	ρ (g/cm^3)	η (mPa.s)
Water/brine + ionic liquid	Upper	0.99712	0.893	1.02620	0.950
	Lower	1.27455	83.93	1.27474	90.60
Ionic liquid + *n*-dodecane	Upper	0.74520	1.354	0.74520	1.354
	Lower	1.24173	95.15	1.24173	95.15
Triphasic system	Upper	0.74517	1.398	0.74653	1.394
	Middle	0.99711	0.894	1.02554	0.943
	Lower	1.24003	68.93	1.24010	69.46

Standard uncertainty: $u(P)$ = 5 kPa. For density measurements: $u(T)$ = 0.01 K. $u(\rho)$ = 0.00003 g · cm^{-3}. For viscosity measurements: $u(T)$ = 0.01 K. $u(\eta)$ = 0.5%.

Table S5. Densities and viscosities of involved phases in water/brine + [C_{10}mim][NTf_2] + dodecane equilibrium at 348.15 K

		H_2O		Brine	
	Phase	ρ (g/cm^3)	η (mPa.s)	ρ (g/cm^3)	η (mPa.s)
Water/brine + ionic liquid	Upper	0.97500	0.381	1.00575	0.420
	Lower	1.23022	13.37	1.20062	15.25
Ionic liquid + *n*-dodecane	Upper	0.70852	0.682	0.70852	0.682
	Lower	1.19666	14.91	1.19666	14.91
Triphasic system	Upper	0.70846	0.662	0.70857	0.662
	Middle	0.97470	0.381	1.00416	0.418
	Lower	1.19603	10.68	1.18684	12.15

Standard uncertainty: $u(P)$ = 5 kPa. For density measurements: $u(T)$ = 0.01 K. $u(\rho)$ = 0.00003 g · cm^{-3}. For viscosity measurements: $u(T)$ = 0.01 K. $u(\eta)$ = 0.5%.

Table S6. Densities and viscosities of involved phases in water/brine + [C_{12}mim][NTf_2] + dodecane equilibrium at 298.15 K

		H_2O		Brine	
	Phase	ρ (g/cm^3)	η (mPa.s)	ρ (g/cm^3)	η (mPa.s)
Water/brine + ionic liquid	Upper	0.99706	0.897	1.02696	0.958
	Lower	1.24066	109.2	1.24088	111.4
Ionic liquid + *n*-dodecane	Upper	0.74536	1.350	0.74536	1.350
	Lower	1.17744	103.7	1.17744	103.7
Triphasic system	Upper	0.74563	1.348	0.74518	1.347
	Middle	0.99706	0.893	1.02617	0.950
	Lower	1.17591	79.98	1.17606	80.09

Standard uncertainty: $u(P)$ = 5 kPa. For density measurements: $u(T)$ = 0.01 K. $u(\rho)$ = 0.00003 g · cm^{-3}. For viscosity measurements: $u(T)$ = 0.01 K. $u(\eta)$ = 0.5%.

Table S7. Densities and viscosities of involved phases in (water/brine + [C_{12}mim][NTf_2] + dodecane) equilibrium at 348.15 K

		H_2O		Brine	
	Phase	ρ (g/cm^3)	η (mPa.s)	ρ (g/cm^3)	η (mPa.s)
Water/brine + ionic liquid	Upper	0.97490	0.379	1.00921	0.425
	Lower	1.19508	15.09	1.19518	15.64
Ionic liquid + *n*-dodecane	Upper	0.70866	0.662	0.70866	0.662
	Lower	1.13559	15.33	1.13559	15.33
Triphasic system	Upper	0.70903	0.663	0.70854	0.667
	Middle	0.97174	0.380	1.00409	0.417
	Lower	1.12766	11.73	1.13463	12.31

Standard uncertainty: $u(P)$ = 5 kPa. For density measurements: $u(T)$ = 0.01 K. $u(\rho)$ = 0.00003 g · cm^{-3}. For viscosity measurements: $u(T)$ = 0.01 K. $u(\eta)$ = 0.5%.

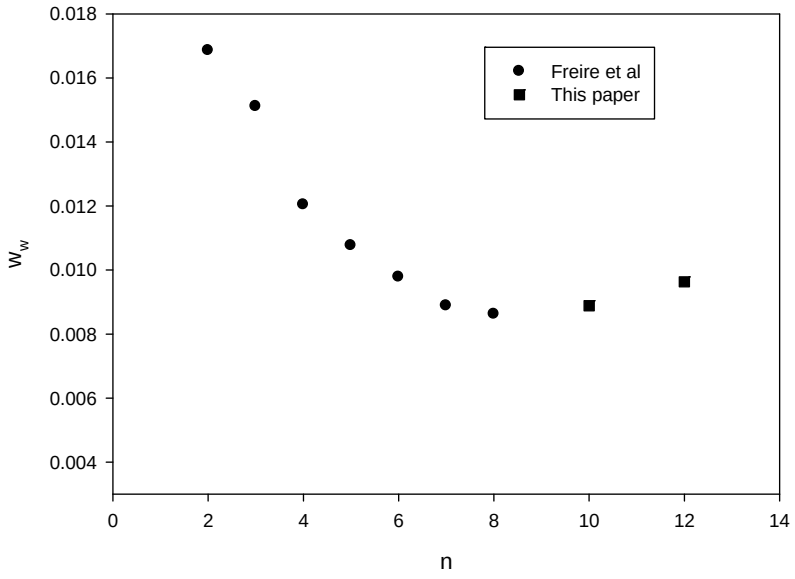

Figure S1. Solubility (mass fraction) of water in $[C_n mim][NTf_2]$ at 298.15 K

Literature

M.G. Freire, P.J. Carvalho, R.L. Gardas, I.M. Marrucho, L.M.N.B.F. Santos, J.A. P. Coutinho, *J. Phys. Chem. B* **2008,** *112,* 1604-1610