Modelling of the Mixture critical locus with a modified Cubic Plus Association (CPA) EoS – Water, alkanols, amines and alkanes

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Supplementary Data

Amines parameters

Table A-1 Parameters for the amine sets used in this work

Compound	a_c (Pa.m^6.mol^{-2})	$b.10^5$ (m^3.mol^{-1})	c_1	c_2	c_3	c_4	c_5	$\beta.10^2$	c_{vs} (m^3.kmol^{-1})
diethylamine	1.95	9.74	0.75	1.25	-5.2	3.4	0	0.23	0.016
dipropylamine	2.84	12.69	1.44	-3.32	13.2	-29.8	23	0.11	0.009

Binary interaction parameter and %AAD tables for binary mixtures

Table A-2 Binary interaction parameters and %AAD for mixtures containing two associative compounds.

Comp. 1	Comp. 2	Data used	k_{ij}	%AAD T_c	%AAD P_c	Figure
methanol	1-propanol	no kij		0.10	2.14	1
methanol	1-butanol	no kij		0.11	3.25	1, 3
ethanol	2-propanol	VLE	-0.009	0.27	no data	-
methanol	water	VLE	-0.045	0.29	3.12	2, 3
ethanol	water	VLE	-0.004	0.44	2.02	2
1-propanol	water	VLE	$0.125-2.93\times10^{-4}T$	0.46	7.66	2

Table A-3 Binary interaction parameters and %AAD for mixtures containing methanol and an alkane

Comp. 1	Comp. 2	Data used	k_{ij}	%AAD T_c	%AAD P_c	Figure
methanol	hexane	LLE	0.036	0.58	8.66	4
methanol	hexane	VLE	0.052	0.38	8.70	5, 6
methanol	hexane	VLE	$-0.069+4.00\times10^{-4}T$	0.51	8.49	-
methanol	heptane	LLE	0.033	0.46	12.42	4
methanol	heptane	VLE	0.055	0.25	12.35	-
methanol	octane	LLE	0.028	0.41	12.71	4
methanol	nonane	LLE	0.024	0.31	12.22	-
methanol	decane	LLE	0.020	0.39	13.90	-
methanol	dodecane	LLE	0.009	0.43	11.42	-
methanol	tetradecane	LLE	0.009	0.30	11.32	-

Table A-4 Binary interaction parameters and %AAD for mixtures containing hexane and a primary alkanol (except methanol)

Comp. 1	Comp. 2	Data used	k_{ij}	%AAD T_c	%AAD P_c	Figure
ethanol	hexane	VLE	0.056	0.37	6.24	5
1-propanol	hexane	VLE	0.041	0.89	1.24	5
1-butanol	hexane	VLE	0.027	0.58	1.36	5, 6
1-pentanol	hexane	VLE	0.031	0.62	0.69	5
ethanol	hexane	VLE	$-0.068+3.74\times10^{-4}T$	0.55	6.44	-
1-propanol	hexane	VLE	$0.143-2.81\times10^{-4}T$	1.60	1.64	-

Table A-5 Binary interaction parameters and %AAD for mixtures containing a primary alkanol and an alkane (except methanol)

Comp. 1	Comp. 2	Data used	k_{ij}	%AAD T_c	%AAD P_c	Figure
ethanol	butane	VLE	0.047	0.73	1.48	7
ethanol	pentane	VLE	0.097	0.09	3.71	7
ethanol	heptane	VLE	0.046	0.25	6.03	7
ethanol	octane	VLE	0.046	0.37	6.78	7
ethanol	nonane	VLE	0.022	0.32	no data	-
ethanol	cyclohexane	VLE	0.071	0.37	3.44	-
1-propanol	butane	VLE	0.027	1.93	1.42	-
1-propanol	heptane	VLE	0.050	0.23	4.41	-
1-propanol	octane	VLE	0.040	0.40	4.47	-
1-propanol	nonane	VLE	0.006	1.38	no data	-
1-propanol	decane	VLE	0.015	1.25	6.52	-
1-propanol	cyclohexane	VLE	0.072	0.04	2.73	-
1-butanol	butane	VLE	0.027	2.04	2.14	-
1-butanol	pentane	VLE	0.032	0.56	no data	-
1-butanol	nonane	VLE	0.017	0.60	no data	-
1-butanol	decane	VLE	0.015	0.70	no data	-
1-pentanol	pentane	VLE	0.022	1.30	no data	-
1-pentanol	nonane	VLE	0.034	0.22	no data	-
1-hexanol	heptane	VLE	0.045	0.23	no data	-

Table A-6 Binary interaction parameters and %AAD for mixtures containing a secondary alkanol and an alkane

Comp. 1	Comp. 2	Data used	k_{ij}	%AAD T_c	%AAD P_c	Figure
2-propanol	hexane	VLE	0.073	0.33	3.44	-
2-butanol	hexane	VLE	0.039	0.42	no data	-
2-pentanol	hexane	VLE	0.030	0.33	no data	-
2-propanol	pentane	correlated	0.089	0.23	no data	8
2-propanol	heptane	VLE	0.064	0.67	6.21	8
2-propanol	octane	VLE	0.055	0.40	5.35	8
2-propanol	decane	VLE	0.021	0.60	6.37	8
2-propanol	cyclohexane	VLE	0.073	1.09	3.48	8
2-butanol	heptane	VLE	0.045	0.22	2.77	-
2-butanol	octane	VLE	0.043	0.63	3.94	-
2-butanol	nonane	correlated	0.041	1.40	3.69	-
2-butanol	decane	correlated	0.039	0.91	4.20	-
2-butanol	cyclohexane	VLE	0.070	0.11	1.70	-

Table A-7 Binary interaction parameters and %AAD for mixtures containing an amine

Comp. 1	Comp. 2	Data used	k_{ij}	%AAD T_c	%AAD P_c	Figure
diethylamine	hexane	no kij		0.47	0.77	9
dipropylamine	hexane	no kij		0.59	no data	9

Table A-8 Binary interaction parameters and %AAD for the mixtures methanol + methane and water + hexane

Comp. 1	Comp. 2	Data used	k_{ij}	Figure
methanol	methane	gas solubility	$0.074+2.18\times10^{-4}T$	10
methanol	methane	critical data	$0.194-4.67\times10^{-4}T$	10, 11
water	hexane	LLE	0.180	12
water	hexane	critical data	0.075	12

The data used in the above calculations and not present in the main document are from de Loos et al. [1], He et al. [2], Deák et al. [3], Hicks and Young [4] Christou et al. [5] and the TRC database [6]

Table A-9 Binary interaction parameters used with the SRK EoS.

Comp. 1	Comp. 2	Data used	k_{ij}	Figure
methanol	1-butanol	No k_{ij}		3
ethanol	water	VLE	-0.079	3
methanol	hexane	VLE	0.064	6
methanol	hexane	critical data	0.156	6
ethanol	pentane	VLE	0.125	-
ethanol	hexane	VLE	0.126	-
1-butanol	hexane	VLE	0.062	6
methanol	methane	gas solubility	$-0.327 + 9.14 \times 10^{-4} T$	10,11

Table A-10 Binary interaction parameters used for the LLE calculations

Comp. 1	Comp. 2	Fit	k_{ij}	Figure
methanol	hexane	0.1 MPa	0.033	15
methanol	hexane	1 MPa	0.031	15
methanol	methylcyclopentane	LLE	0.052	16
methanol	methylcyclopentane	LLCP	0.050	16
methanol	decane	LLCP	0.017	17
ethanol	hexadecane	LLCP	-0.037	17

VLE results using the k_{ij} values presented in the previous tables (results not available here are presented in previous works [7–9])

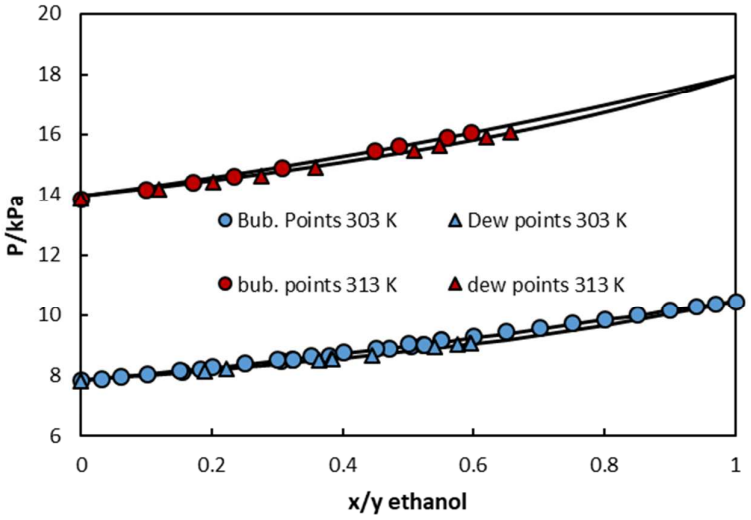

Figure A-1 VLE of 2-propanol + ethanol at two temperatures. Data from Pradhan et al. [10] and Zielkiewicz. [11]

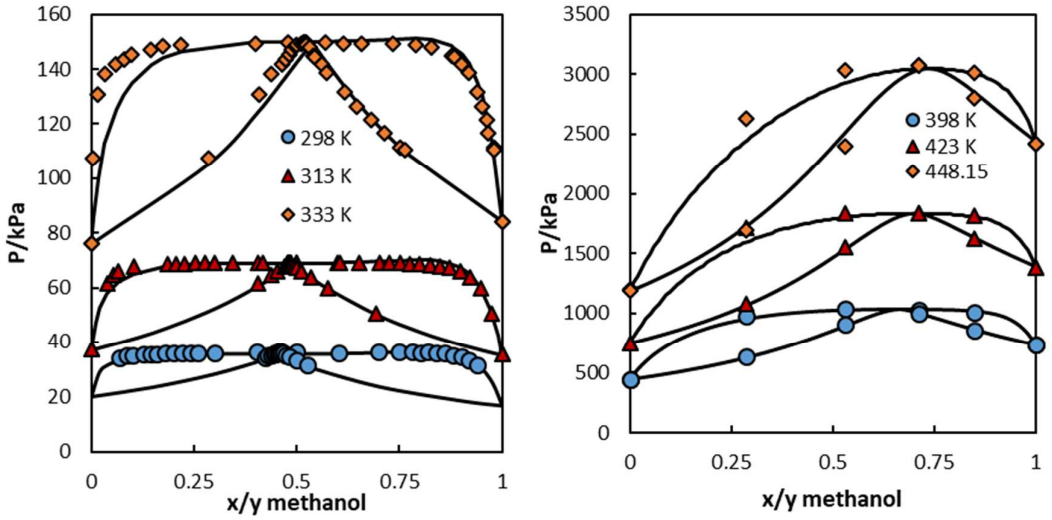

Figure A-2 VLE of hexane + methanol at six temperatures, using the temperature dependent k_{ij} from table 6.3. Data from Alonso et al. [12], Hongo et al. [13], Zawisza [14] and Scheller et al. [15]

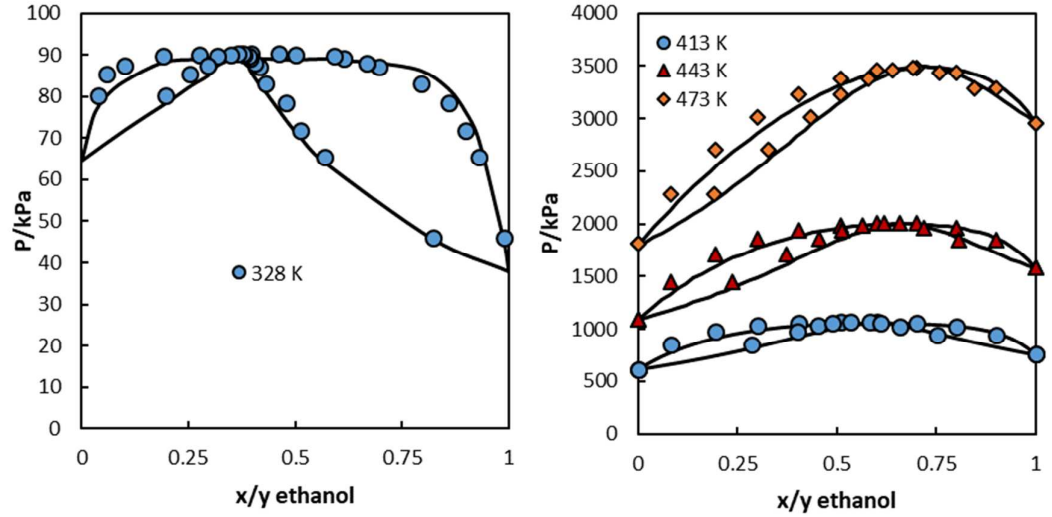

Figure A-3 VLE of hexane + ethanol at six temperatures, using the temperature dependent k_{ij} from table 6.3. Data from Nhu et al. [16] and Yuan et al. [17]

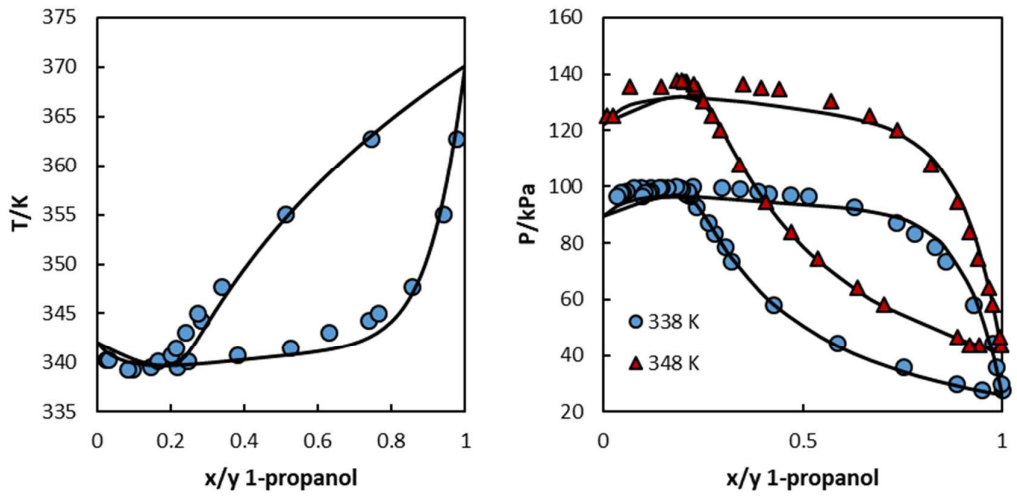

Figure A-4 VLE of hexane + 1-propanol at two temperatures (right) and at 1 bar (left), using the temperature dependent k_{ij} from table 6.3. Data from Maciel and Francesconi [18] and the TRC database. [6]

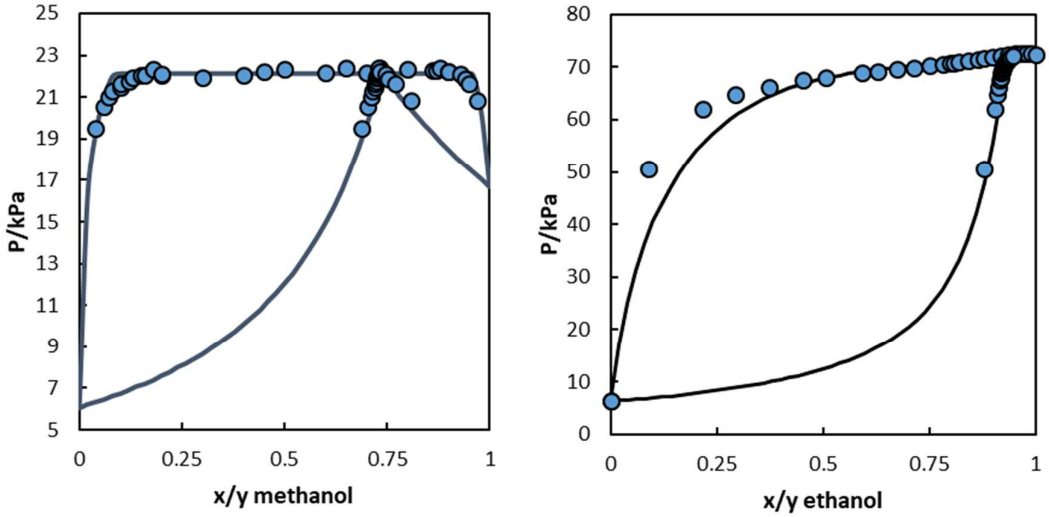

Figure A-5 VLE results of methanol + heptane at 298.15 K (left) and for ethanol + nonane at 343.21 K (right). Data from Hongo et al. [13] and Berro et al. [19]

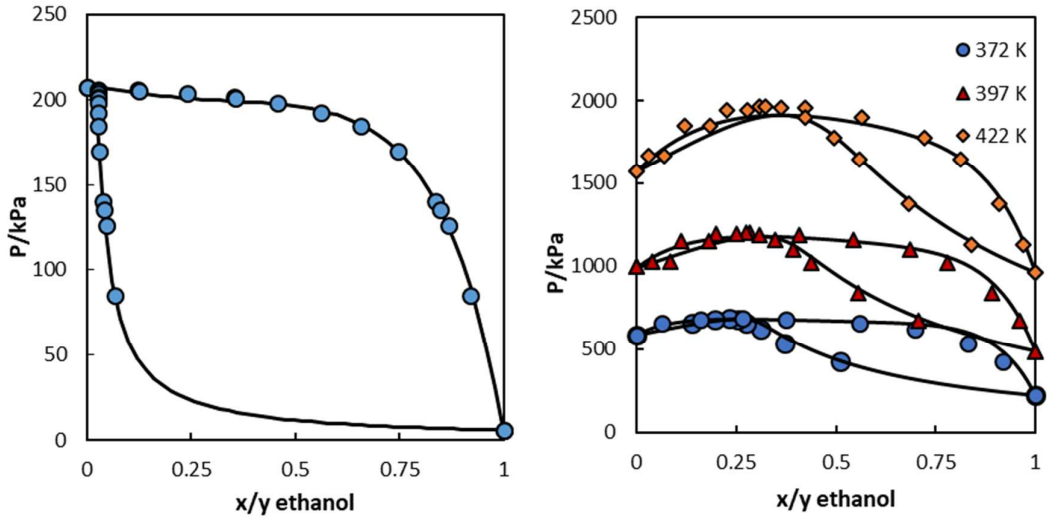

Figure A-6 VLE of butane + ethanol at 293.15 K (left) and for pentane + ethanol at three temperatures (right). Data from Dahlhoff et al. [20] and Campbell et al. [21]

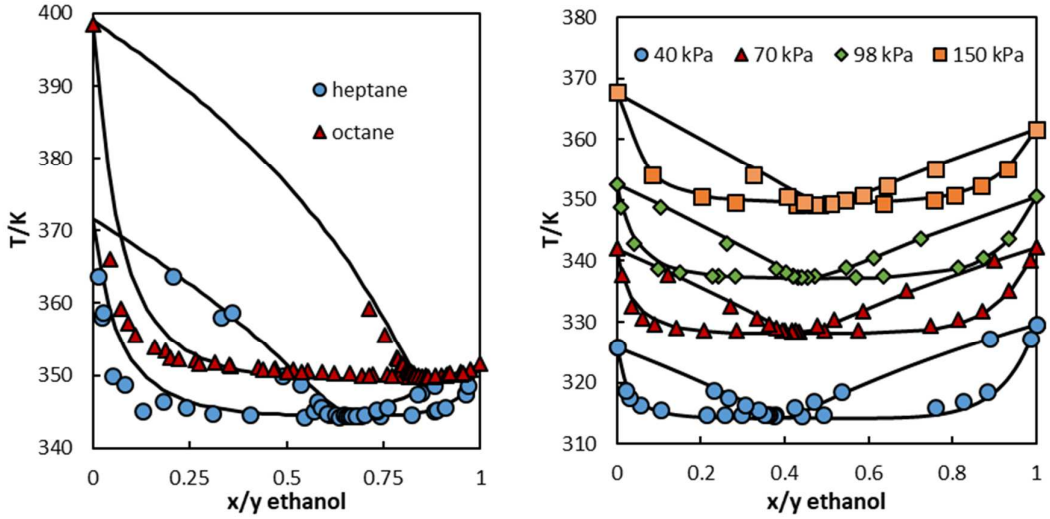

Figure A-7 VLE of heptane + ethanol and octane + ethanol at 1 bar (left) and for cyclohexane + ethanol at four pressures (right). Data from Raal et al. [22] Li and Li [23], Hiaki et al. [24] and Reddy et al. [25]

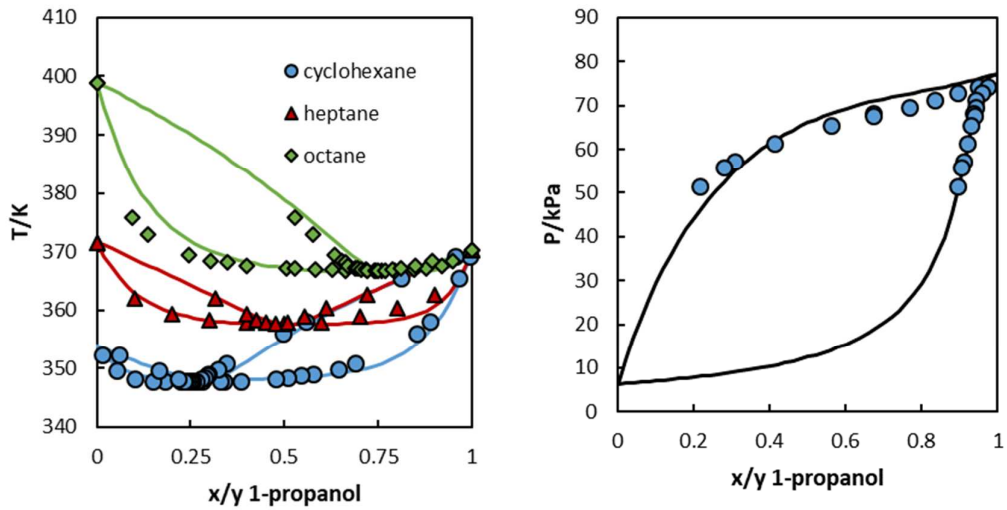

Figure A-8 VLE of heptane + 1-propanol, octane + 1-propanol and cyclohexane + 1-propanol at 1 bar (left) and for decane + 1-propanol at 363 K (right). Data from Hiaki et al. [26,27], Gurukul and Raju [28] and Ratcliff and Chao. [29]

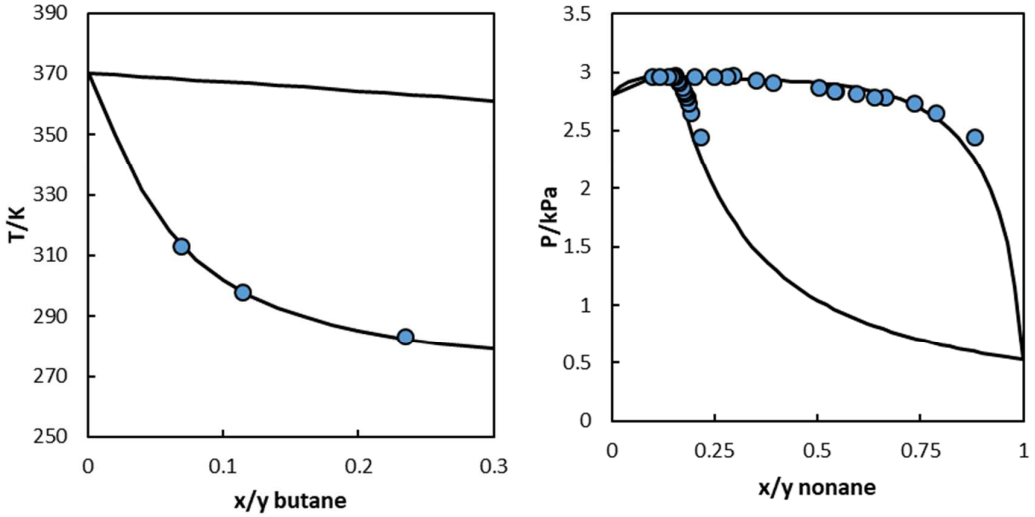

Figure A-9 VLE results of butane + 1-propanol at 1 bar (left) and for 1-propanol + nonane at 298.15 K (right). Data from Mlyano and Hayduk [30] and Heintz et al. [31]

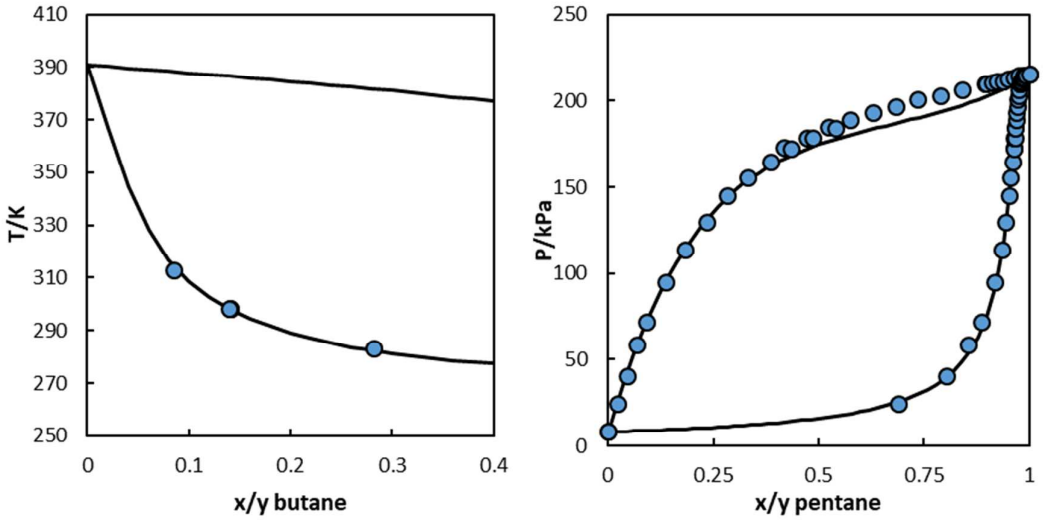

Figure A-10 VLE results of butane + 1-butanol at 1 bar (left) and for 1-butanol + pentane at 333.15 K (right). Data from Mlyano and Hayduk [30] and McDougal et al. [32]

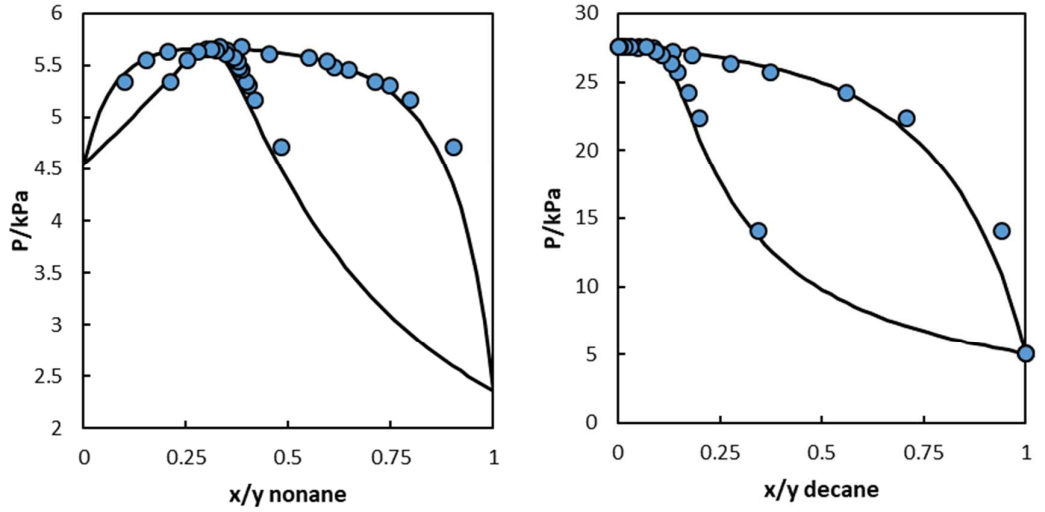

Figure A-11 VLE results of nonane + 1-butanol at 1 323.14 K (left) and for 1-butanol + decane at 358.15 K (right). Data from Heintz et al. [31] and Bernatová et al. [33]

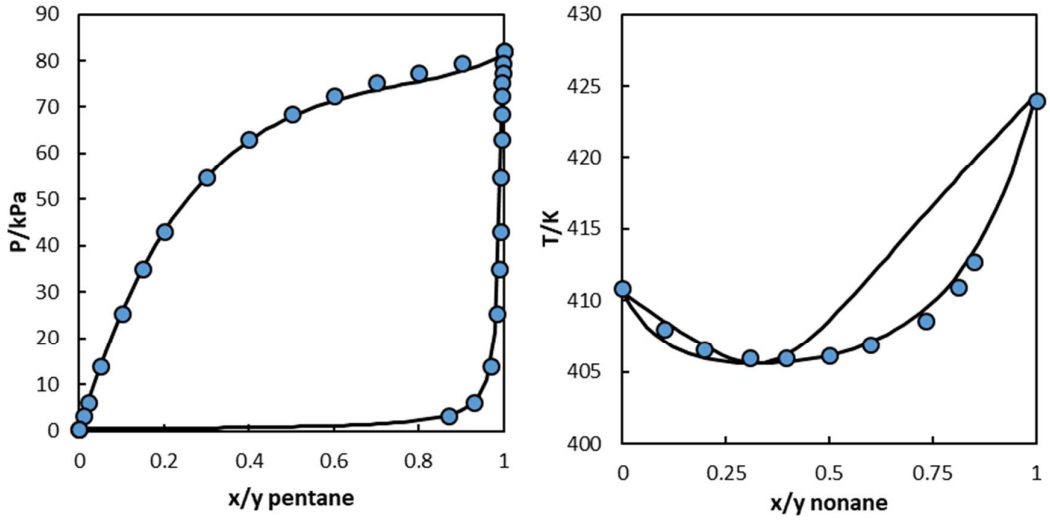

Figure A-12 VLE results of pentane + 1pentanol at 1 303.15 K (left) and for 1-pentanol + nonane at 1 bar (right). Data from Ronc and Ratcliff [34] and Kirss et al. [35]

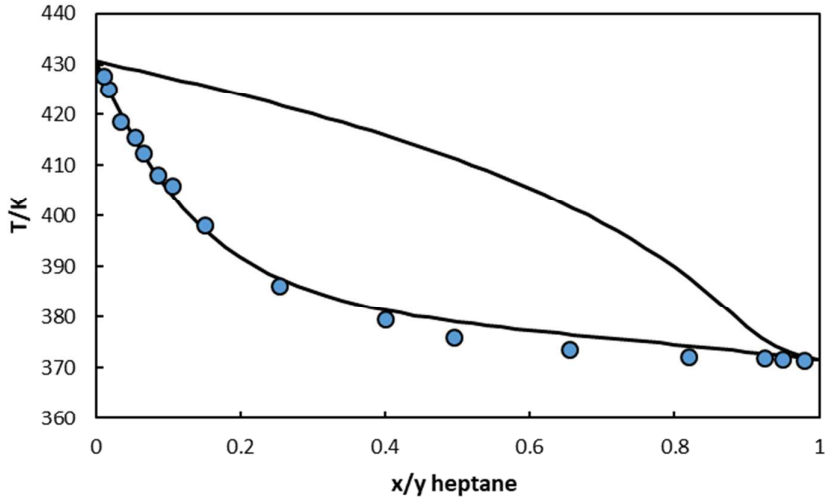

Figure A-13 VLE results of heptane + 1-hexanol at 1 bar. Data from Rao et al. [36]

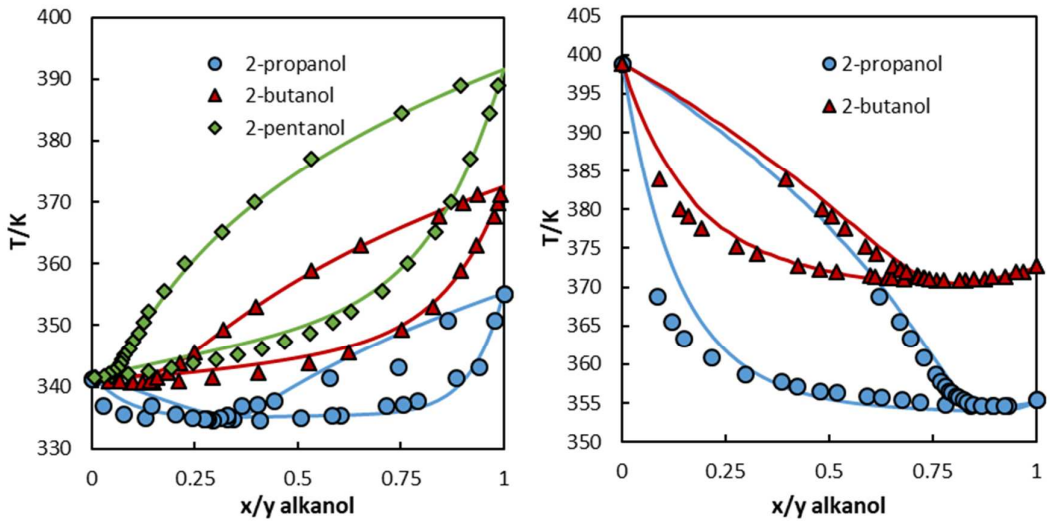

Figure A-14 VLE of hexane + 2-propanol, hexane + 2-butanol and hexane + 2-pentanol at 1 bar (left) and for octane + 2-propanol and octane + 2-butanol at 1 atm (right). Data from Rotter et al. [37], Domínguez et al. [38], Marrufo et al. [39] and Hiaki et al. [27,40]

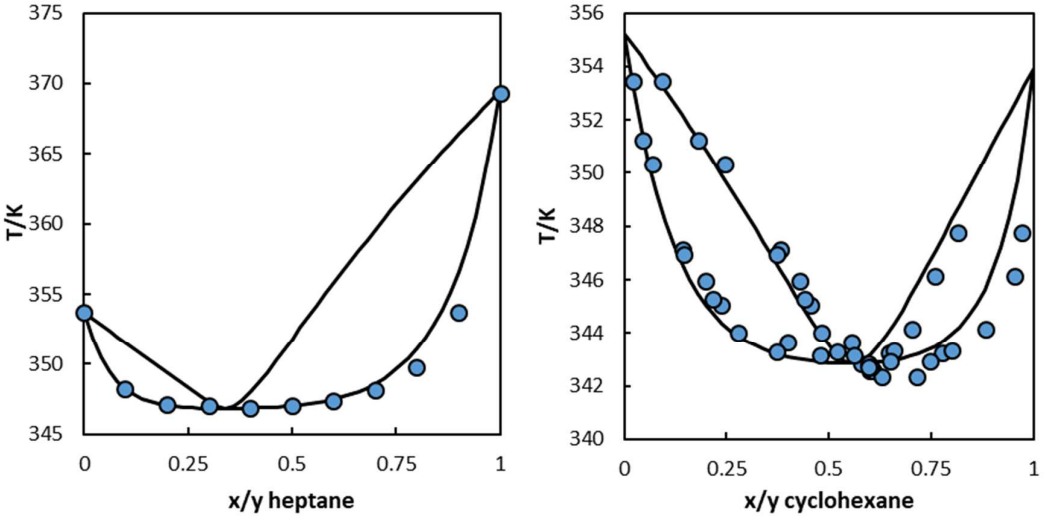

Figure A-15 VLE results of 2-propanol + heptane at 0.95 bar (left) and 2-propanol + cyclohexane at 1 bar (right). Data from Prasad et al. [41], Verhoeye [42] and Nagata. [43]

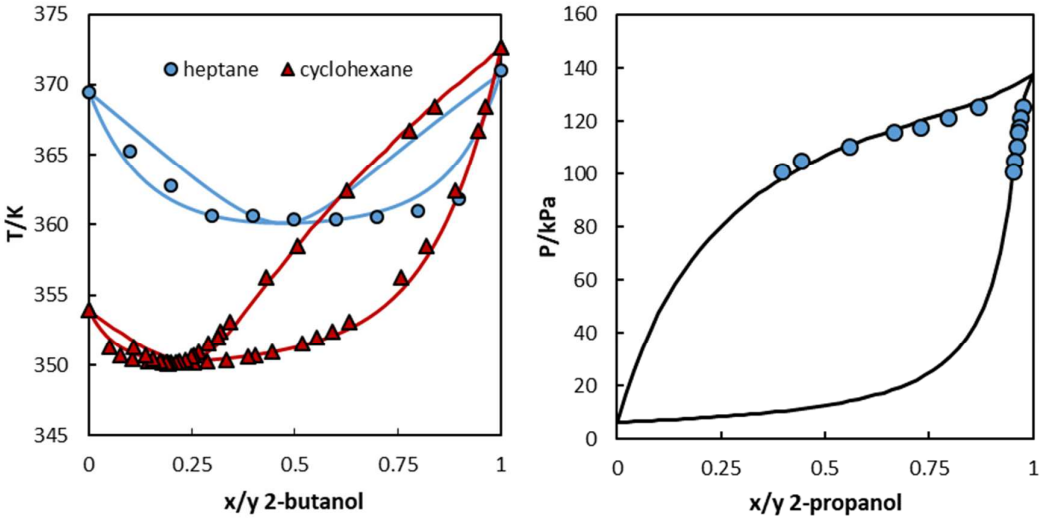

Figure A-16 VLE of heptane + 2-butanol (at 0.95 bar), cyclohexane + 2-butanol (at 1 bar) (left) and for decane + 2-propanol at 363 K (right). Data from Prasad et al. [41], Feng et al. [44] and Ratcliff and Chao. [29]

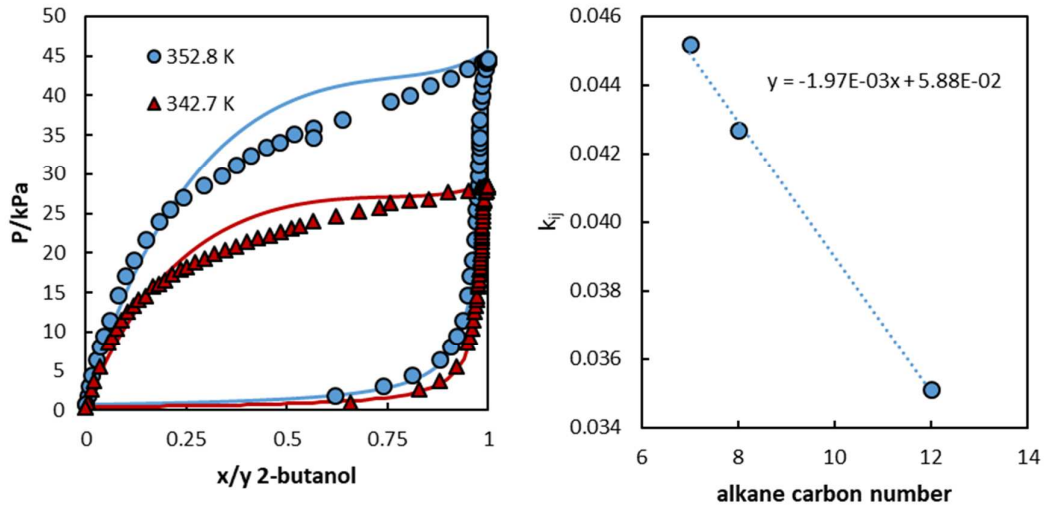

Figure A-17 VLE of dodecane + 2-butanol at two temperatures (left) and correlation applied for the k_{ij} of 2-butanol + nonane and 2-butanol + decane. Data from Raal et al.[45]

Example of the results of T_c vs P_c for some mixtures

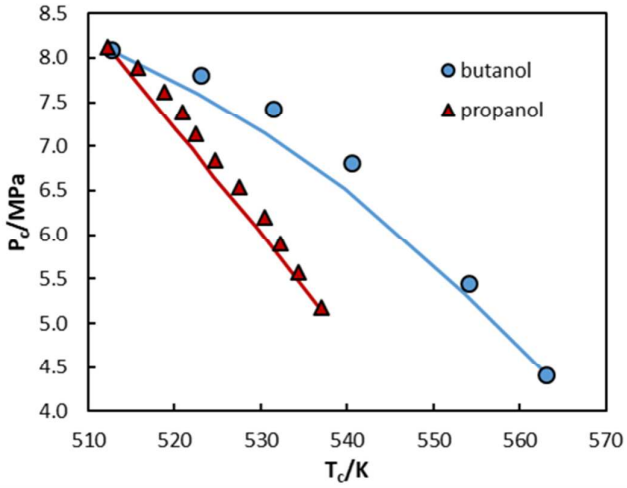

Figure A-18 Curve of the predicted P_c in relation to predicted T_c for methanol + 1-propanol and methanol + 1-butanol mixture, compared to experimental data for both properties. Experimental data from Nazmutdinov et al. [46] and Wang et al. [47].

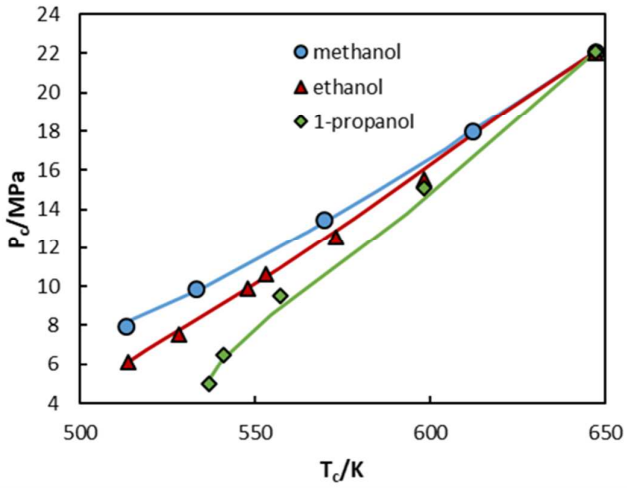

Figure A-19 Results for P_c as function of T_c for water + methanol, water + ethanol and water + 1-propanol mixtures. Experimental data from Hicks and Young [4] and Bazaev et al. [48].

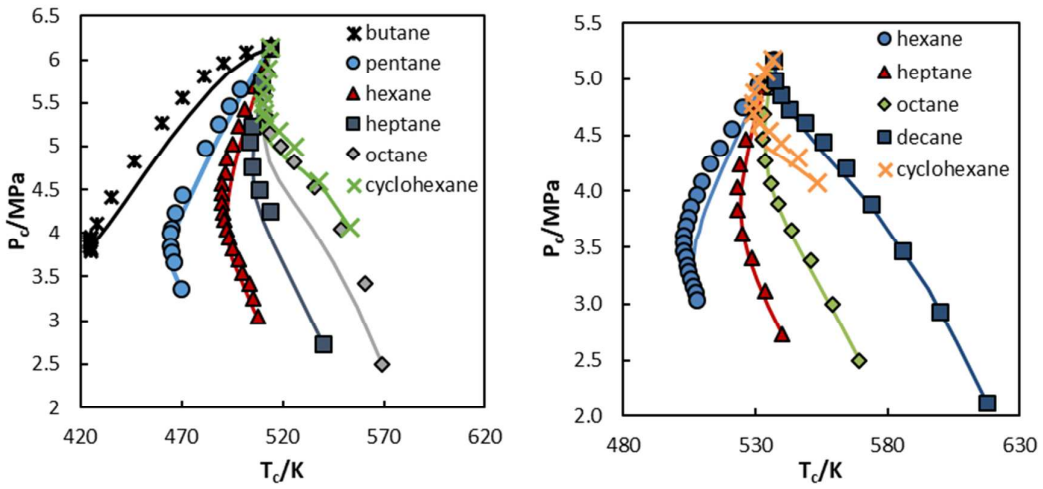

Figure A-20 Results for P_c as function of T_c for ethanol + alkanes (left) and 1-propanol + alkanes (right). Data from He et al. [49], Soo et al. [50] and Xin et al. [51].

P_c results for mixtures where only T_c was presented in the main document

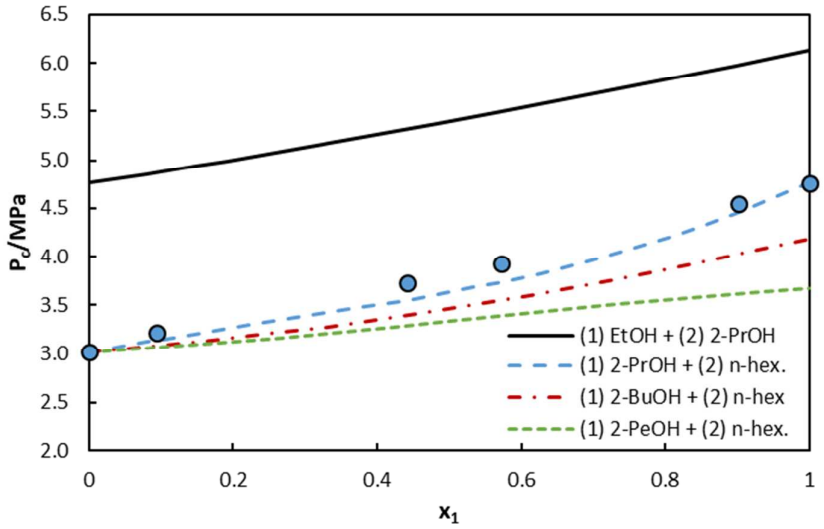

Figure A-21 Predictions of P_c for the mixtures ethanol + 2-propanol, n-hexane + 2-propanol, n-hexane + 2-butanol and n-hexane + 2-pentanol. Data for 2-propanol + n-hexane are from Seo et al. [52]

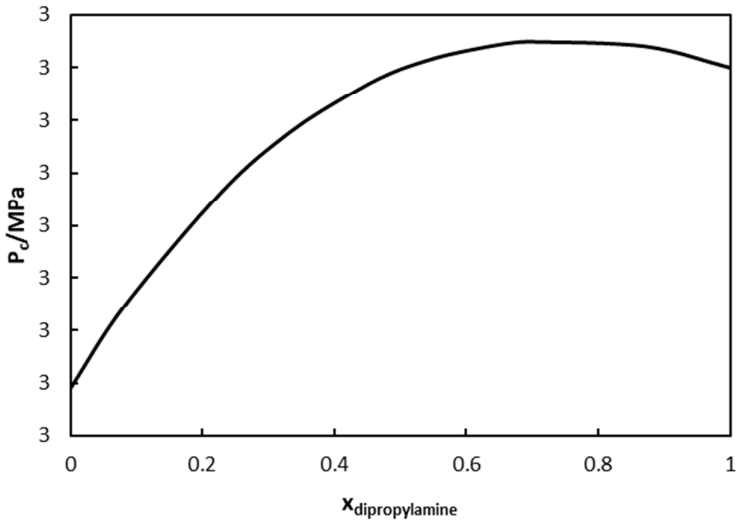

Figure A-22 Predictions of P_c for the mixture dipropylamine + hexane.

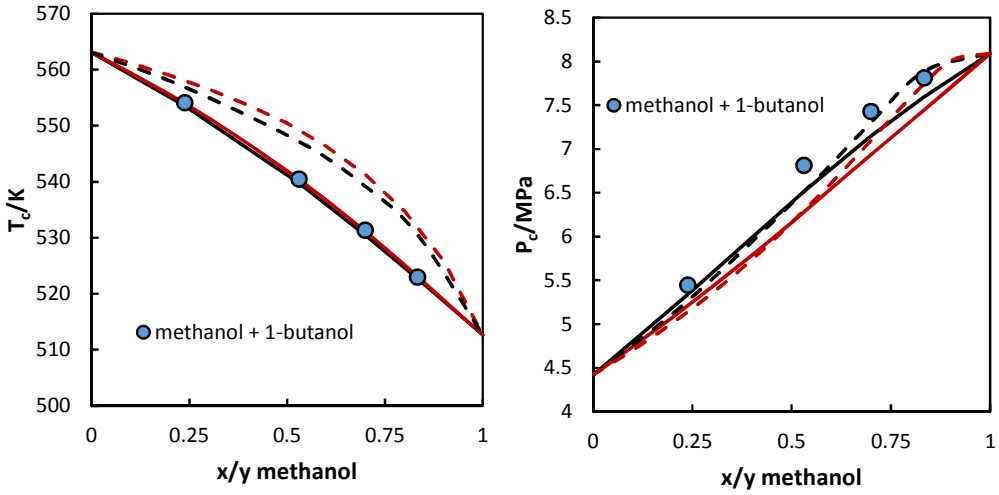

Figure A-23 Results for T_c (left) and P_c (right) for methanol + 1-butanol (top). Black Lines - modified CPA; Red lines - SRK. Dashed lines are obtained by introducing k_{ij} = -0.05 and -0.09 for CPA and SRK respectively.

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