Isobaric Vapor–Liquid Equilibria of the Water + 1-Propanol System at 30, 60, and 100 kPa

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Isobaric vapor—liquid equilibria for the water + 1-propanol system are reported at 30, 60, and 100 kPa. The results were found to be thermodynamically consistent according to Van Ness—Byer—Gibbs, Kojima, and Wisniak methods. The system shows a minimum boiling azeotrope, and the azeotropic composition is scarcely shifted with pressure. Results were compared with literature values. The data were correlated with Margules, Van Laar, Wilson, NRTL, and UNIQUAC liquid-phase activity coefficient models.

Introduction

Distillation is the most common operation for the separation of liquid binary and multicomponent mixtures. The correct design of distillation columns requires the availability of accurate and, if possible, thermodynamically consistent vapor—liquid equilibria (VLE) data and the use of generalized methods to predict the properties of the mixtures.

The present work is part of a project studying the effect of pressure on the behavior of the azeotropic point in mixtures in which at least one component is an alcohol. For this purpose, the azeotropic system water + 1-propanol was selected. For this system, isobaric and isothermal VLE data sets have been found in the literature, the majority of these included in the DECHEMA Chemistry Data Series (Gmehling and Onken, 1977, 1981) VLE compilation, and other more recently presented sources (Munday et al., 1980; Morrison et al., 1990; Zielkiewicz and Konitz, 1991). Among these, seven isobaric and six isothermal sets of data have been reported in the DECHEMA as consistent according to the point-to-point thermodynamic consistency test of Van Ness-Byer-Gibbs (1973), modified by Fredenslund (1977). However, a detailed study on these data reveals differences with respect to the azeotropic values. The azeotropic points are plotted in Figure 1, showing how azeotropic compositions tend to be scattered. The object of our study is the accurate measurement of VLE and the determination of azeotropic data for the isobaric system at three pressures (30, 60, and 100 kPa) not previously reported. The contribution presented in this paper includes VLE data that fulfill the thermodynamic consistency tests (Van Ness-Byer-Gibbs, 1973; Kojima et al. 1990; Wisniak, 1993).

Experimental Section

Chemicals. Milli-Q water and 1-propanol high-purity grade purchased from Aldrich Chemical were used without further purification. The purities of all chemicals, checked by gas chromatography (GC), were as follows: water, 100.00 mass %; 1-propanol, 99.80 mass %. Experimental densities of the pure liquids at 293.15 K were measured in a digital precision densimeter, Anton Paar DMA55, with a thermostated bath controlled to 0.01 K. The refractive indexes at 293.15 K were measured using an Abbe refractometer (ATAGO 3T). The accuracies in density and refractive index measurements were 0.000 01 g cm⁻³ and ± 0.0002 , respectively. The boiling points were determined



Figure 1. Variation of the azeotropic composition for the system water (1) + 1-propanol (2): (\bigcirc) this work; (\bigcirc) literature data (points numbered as indicated in Table 6).

Table 1. Physical Properties of Chemicals: Densities d,Refractive Indexes n, and Boiling Points $T_{\rm b}$

	d(293.15	/(293.15 K)/g cm ⁻³		<i>n</i> (293.15 K)		T _b (100.00 kPa)/K	
compd	exptl	lit.	exptl	lit.	exptl	lit.	
water	0.998 06	0.998 20 ^a	1.3334	1.3330 ^a	372.78	372.77 ^a	
1-propanol	0.803 66	0.803 75 ^a	1.3853	1.3855 ^a	369.75	369.95 ^a	

^a TRC Thermodynamic Tables Hydrocarbons, 1994.

using the apparatus described below. The experimental values of these properties are listed in Table 1 together with the literature values (TRC, 1994).

Apparatus and Procedure. The equilibrium vessel used in this work is an all-glass, dynamic recirculating still described by Walas (1985), equipped with a Cottrell pump. The still (Labodest model), manufactured by Fischer Labor und Verfahrenstechnik (Bonn, Germany) is capable of handling pressures from 0.25 to 400 kPa and temperatures up to 523.15 K. In the boiler, vapor is generated by external heating. The Cottrell pump ensures the intimate contact between the liquid and vapor phases and also with the temperature sensing element. The equilibrium temperature is measured with a digital Ditel thermometer with an accuracy of ± 0.01 K. For the pressure measurement,

Table 2. Antoine Coefficients A, B, and C

		Ant	Antoine coefficients				
component	temp range/K	A	В	С			
water 1-propanol	$274 - 373 \\ 303 - 370$	16.5700 16.0353	$3984.92 \\ 3415.56$	$-39.724 \\ -70.733$			

Table 3. Vapor–Liquid Equilibrium Data, Liquid-Phase Mole Fraction x_1 , Vapor-Phase Mole Fraction y_1 , Temperature *T*, and Activity Coefficients γ_i for the Water (1) + 1-Propanol (2) System at 30 kPa

<i>X</i> ₁	y_1	<i>T</i> /K	γ1	Y2
0.000	0.000	341.08		
0.008	0.027	340.49	3.658	1.008
0.066	0.184	337.88	3.393	1.016
0.137	0.315	335.92	3.057	1.015
0.210	0.405	334.33	2.757	1.041
0.304	0.481	332.82	2.425	1.110
0.377	0.514	332.26	2.145	1.194
0.434	0.545	332.06	1.994	1.243
0.510	0.568	331.93	1.780	1.371
0.578	0.583	331.93	1.613	1.537
0.659	0.597	331.84	1.453	1.848
0.725	0.606	331.94	1.335	2.229
0.779	0.612	331.99	1.252	2.725
0.823	0.615	332.08	1.186	3.360
0.857	0.619	332.17	1.141	4.098
0.889	0.622	332.27	1.100	5.211
0.914	0.629	332.51	1.070	6.523
0.936	0.638	332.83	1.044	8.418
0.954	0.661	333.43	1.033	10.646
0.964	0.691	334.27	1.028	11.896
0.977	0.739	335.90	1.007	14.523
0.983	0.796	337.06	1.023	14.519
0.989	0.851	338.46	1.020	15.326
0.994	0.891	339.50	1.015	19.565
0.997	0.935	340.52	1.015	22.240
1.000	1.000	342.33		

a digital manometer with an accuracy of ± 0.01 kPa is used. The temperature probe was calibrated against the ice and steam points of distilled water. High-purity (>99.9 mass %) hexane vapor pressures were used for the manometer calibration.

In each VLE experiment, the pressure was fixed and remained constant using a vacuum pump, and the heating and shaking systems of the liquid mixture were turned on. The system was kept at the boiling point at least for 30 min to ensure that the steady state was reached. At this moment, 0.2 cm³ samples of liquid and condensed vapor of the Cottrell pump were taken for analysis.

Analysis. All the samples were analyzed by using a Varian Star 3400 CX gas chromatograph with a thermal conductivity detector. The GC response was treated with a Star Chromatography Station. The chromatographic column (2 m × $^{1}/_{8}$ in.) was packed with Porapak P. The carrier gas was helium flowing at 50 cm³ min⁻¹, and the column temperature was 383.15 K. The GC was calibrated with gravimetrically prepared standard solutions. The uncertainty of composition measurements was estimated to be ±0.001. At least two analyses were made for each liquid and each condensed vapor sample.

Results and Discussion

Vapor pressures P_i^0 were calculated with the Antoine equation

$$\ln(P_i^0/kPa) = A - \frac{B}{(C + T/K)}$$
(1)

The Antoine constants for water were those given in DECHEMA *Chemistry Data Series* (Gmehling and Onken, 1977). For 1-propanol, the values given in Aucejo et al.

Table 4. Vapor–Liquid Equilibrium Data, Liquid-Phase Mole Fraction x_1 , Vapor-Phase Mole Fraction y_1 , Temperature *T*, and Activity Coefficients γ_i for the Water (1) + 1-Propanol (2) System at 60 kPa

<i>X</i> 1	y_1	<i>T</i> /K	γ1	γ_2
0.000	0.000	356.78		
0.009	0.031	356.35	3.845	0.995
0.045	0.123	354.53	3.318	1.009
0.103	0.245	352.44	3.107	1.011
0.168	0.340	350.70	2.839	1.028
0.231	0.408	349.51	2.603	1.051
0.294	0.461	348.63	2.397	1.083
0.361	0.501	347.63	2.212	1.158
0.417	0.527	347.74	2.005	1.197
0.487	0.551	347.50	1.814	1.306
0.557	0.571	347.52	1.642	1.443
0.621	0.585	347.53	1.508	1.631
0.686	0.594	347.55	1.385	1.925
0.747	0.602	347.73	1.279	2.323
0.796	0.605	347.80	1.203	2.850
0.833	0.609	347.91	1.152	3.430
0.869	0.612	348.14	1.099	4.294
0.892	0.620	348.28	1.078	5.070
0.925	0.630	348.51	1.046	7.036
0.946	0.646	348.99	1.028	9.154
0.960	0.672	349.92	1.014	10.992
0.970	0.704	350.93	1.009	12.655
0.981	0.760	352.78	0.998	14.956
0.988	0.806	354.43	0.983	17.840
0.993	0.872	356.06	0.992	18.838
0.996	0.923	357.30	0.996	18.830
0.999	0.974	358.02	1.019	24.684
1.000	1.000	359.14		

(1995) were used. Antoine constants for both compounds are summarized in Table 2.

The experimental VLE data for the binary system at (30, 60, and 100) kPa are given in Tables 3-5. The T-x-y diagrams at the three pressures are shown in Figure 2. It can be observed that the system presents a minimum boiling azeotrope and the azeotropic composition is scarcely shifted with pressure.

The azeotropic composition, pressure, and temperature are listed in Table 6 along with thermodynamically consistent literature data from the DECHEMA *Chemistry Data Series* (Gmehling and Onken, 1977, 1981). In all cases, the azeotropic point has been calculated from the experimental data by fitting the function $z = x_1/y_1$ to a polynomial function of x_1 and solving for x_1 at z = 1. Variation of azeotropic pressure with composition is shown in Figure 1. Azeotropic pressure is shifted to lower values as the water mole fraction increases. The values obtained in this work are in agreement with the tendency shown by the majority of the consistent data previously reported, contributing to reduce the moderate divergence among the different data.

Comparison of VLE data can be also performed by representing the value of $G^{\rm E}/RT$ at constant composition vs 1/T. Such a plot for 0.5 mole fraction is presented in Figure 3. The temperature dependence of $G^{\rm E}$ may be established by means of $H^{\rm E}$ data according to the Gibss–Helmholtz equation

$$\left(\frac{\partial (G^{\rm E}/T)}{\partial (1/T)}\right)_{P,n} = H^{\rm E}$$
(2)

 $H^{\rm E}$ being the molar excess enthalpy. The $H^{\rm E}$ variation with temperature at equimolar composition has been adjusted from literature $H^{\rm E}$ data (Christensen et al., 1984). In Figure 3, two solid lines showing this tendency have been plotted, delimiting the experimental data.

To calculate the liquid-phase activity coefficients, γ_{i} , the Poynting factor was considered as unity at the experimen-



Figure 2. Temperature *T* vs composition (x_1 , y_1) for the system water (1) + 1-propanol (2) at constant pressure, P = 30, 60, and 100 kPa: (\bigcirc) experimental; (-) NRTL (α_{12} adjustable) model.

Table 5. Vapor-Liquid Equilibrium Data, Liquid Phase Mole Fraction x_1 , Vapor Phase Mole Fraction y_1 , Temperature *T*, and Activity Coefficients γ_i for the Water (1) + 1-Propanol (2) System at 100 kPa

<i>X</i> 1	<i>y</i> 1	<i>T</i> /K	γ1	γ2
0.000	0.000	369.75		
0.013	0.025	369.40	2.174	0.994
0.038	0.097	367.84	3.056	1.003
0.083	0.194	366.07	2.989	1.006
0.160	0.317	363.80	2.760	1.018
0.226	0.390	362.46	2.530	1.041
0.283	0.437	361.63	2.337	1.072
0.356	0.487	360.82	2.136	1.124
0.426	0.522	360.47	1.939	1.192
0.500	0.550	360.33	1.750	1.295
0.583	0.572	360.28	1.564	1.480
0.648	0.585	360.27	1.440	1.701
0.708	0.595	360.32	1.338	1.997
0.763	0.603	360.44	1.252	2.400
0.817	0.608	360.62	1.171	3.047
0.853	0.613	360.78	1.124	3.720
0.883	0.619	360.93	1.090	4.574
0.911	0.627	361.12	1.062	5.841
0.926	0.632	361.43	1.041	6.844
0.945	0.643	361.77	1.024	8.812
0.957	0.663	362.41	1.018	10.369
0.967	0.683	363.30	1.003	12.265
0.978	0.739	365.06	1.004	14.126
0.985	0.794	366.87	1.000	15.232
0.991	0.854	368.71	0.999	16.756
0.996	0.905	370.23	0.996	23.145
0.999	0.984	371.95	1.015	24.348
1.000	1.000	372.78		

tal conditions. The fugacity coefficients were calculated on the basis of the virial equation of state, with the second virial coefficient being estimated by means of the Pitzer

Table 6. Variation of the Azeotropic Composition andTemperature with Pressure for the System Water (1) +1-Propanol (2)

	-			
	<i>P</i> /kPa	<i>X</i> 1	<i>T</i> /K	ref
1	6.4	0.604	303.15	Udovenko and Mazanko, 1972 ^a
2	18.7	0.591	323.07	Vrevskii, 1910 ^a
3	26.7	0.583	329.83	Smirnova, 1959 ^a
4	30.0	0.585	331.90	this work
5	31.2	0.574	333.15	Schreiber et al., 1971 ^a
6	40.1	0.583	339.09	Vrevskii, 1910 ^a
7	53.3	0.576	345.07	Smirnova, 1959 ^a
8	60.0	0.575	347.52	this work
9	66.7	0.550	350.66	Goelles and Still, 1979 ^b
10	73.3	0.571	352.95	Vrevskii, 1910 ^a
11	80.0	0.567	354.83	Smirnova, 1959 ^a
12	100.0	0.569	360.29	this work
13	101.1	0.579	360.57	Dawe et al., 1973 ^a
14	101.3	0.567	360.81	Smirnova, 1959 ^a
15	101.3	0.565	360.77	Kojima et al., 1968 ^b
16	110.1	0.567	363.15	Ratcliff and Chao, 1969 ^a

^a Data from the DECHEMA *Chemistry Data Series* (Gmehling and Onken, 1977). ^b Data from the DECHEMA *Chemistry Data Series* (Gmehling and Onken, 1981).



Figure 3. Comparison of $G^{\mathbb{E}}(x = 0.5)/RT$ at various temperatures: (\bigcirc) this work; (\bullet) literature data (points numbered as indicated in Table 6).

and Curl equations (1957) with the correction proposed by Tsonopoulos (1974). For every experimental pressure– temperature–composition condition, the calculated fugacity coefficients were close to unity (0.9998 maximum, 0.9666 minimum) and were not considered in the analysis of the VLE data. So, the experimental liquid-phase activity coefficients γ_i were calculated from

$$\gamma_i = \frac{P y_i}{x_i P_i^0} \tag{3}$$

The thermodynamic consistency of the VLE experimental data was checked by the following methods: the point-topoint test of Van Ness–Byer–Gibbs (1973), the infinite dilution test proposed by Kojima et al. (1990), modified by Jackson and Wilsak (1995), and the L-W method of Wisniak (1993).

For the point-to-point test of Van Ness-Byer-Gibbs a four-parameter Legendre polynomial was used for the excess Gibbs free energy. The temperature dependence of the excess Gibbs free energy was allowed for by the term $\Delta H/RT^2$ dT from the Gibbs-Duhem equation. Heat of mixing data were taken from the literature (Christensen et al., 1984). The selected objective function to minimize was the sum of the squared relative deviations in the total pressure. The consistency criteria in this test are that the mean absolute deviation between calculated and measured mole fractions of component 1 in the vapor phase, MAD-(y), is less than 0.01, and that deviations scatter randomly about zero. To check the absence of bias in the data, a graphical inspection of the error in y_1 is performed, y_1

Kojima							
Van Ness-Byer-Gibbs		error for dilute	error for dilute	or for dilute Wisr		niak	
<i>P</i> /kPa	MAD(y)	BIAS	component 1 (%)	component 2 (%)	L	W	D
30	0.0098	-0.0054	20.4	21.4	8.23	8.66	2.58
60	0.0071	-0.0007	6.1	1.8	8.79	9.27	2.65
100	0.0070	0.0003	5.4	0.2	9.16	9.67	2.74

Table 8. Correlation Parameters for Activity Coefficients, Activity Coefficients at Infinite Dilution γ_{i}^{ω} , and Mean Absolute Deviations MAD(y) and MAD(T)

<i>P</i> /kPa	model	A_{12}	A_{21}	α ₁₂	γ_1^{∞}	γ_2^{∞}	MAD(y)	MAD(7)
30.00	Margules	0.9388 ^a	2.6214 ^a		2.56	13.75	0.0291	0.64
	Van Laar	1.2133 ^a	2.9060 ^a		3.36	18.28	0.0109	0.33
	Wilson	5451.42^{b}	4019.48 ^b		4.19	25.24	0.0107	0.37
	NRTL ^c	8103.12 ^b	-5.61^{b}	0.30 ^a	3.35	17.19	0.0115	0.35
	$NRTL^{d}$	1048.05 ^b	2190.39 ^b	-1.56^{a}	4.16	18.91	0.0062	0.29
	UNIQUAC	1867.76 ^b	194.98 ^b		3.12	19.00	0.0117	0.39
60.00	Margules	0.9000 ^a	2.6011 ^a		2.46	13.48	0.0297	0.74
	Van Laar	1.1578 ^a	2.9373^{a}		3.18	18.86	0.0101	0.31
	Wilson	5442.05 ^b	4521.85 ^b		3.85	26.47	0.0088	0.31
	NRTL ^c	8738.57 ^b	-198.20^{b}	0.30 ^a	3.16	17.42	0.0120	0.35
	$NRTL^{d}$	977.32^{b}	2313.63 ^b	-1.56^{a}	3.76	19.29	0.0064	0.21
	UNIQUAC	2127.32^{b}	37.79^{b}		3.00	19.11	0.0113	0.37
100.00	Margules	0.8164 ^a	2.5933^{a}		2.26	13.37	0.0299	0.86
	Van Laar	1.1010 ^a	2.9219 ^a		3.01	18.58	0.0099	0.31
	Wilson	5457.66 ^b	4505.32 ^b		3.60	23.93	0.0109	0.34
	NRTL ^c	9218.80 ^b	-414.77^{b}	0.30 ^a	2.96	17.00	0.0117	0.34
	$NRTL^{d}$	874.58 ^b	2399.76 ^b	-1.57^{a}	3.39	18.78	0.0062	0.21
	UNIQUAC	2132.80 ^b	37.71^{b}		2.95	17.98	0.0109	0.33

^{*a*} Dimensionless. ^{*b*} J mol⁻¹. ^{*c*} With recommended α_{12} value. ^{*d*} With adjustable α_{12} .



Figure 4. Deviation between calculated and measured vaporphase mole fraction (y_1) vs liquid-phase mole fraction (x_1) for the water (1) + 1-propanol (2) system at constant pressure, P = 60 kPa.

residuals being plotted vs x_1 . This plot for the 60 kPa data is shown, as an example, in Figure 4. Similar plots were obtained at 30 kPa and at 100 kPa. Experimental VLE data were found thermodynamically consistent according to this test, with the values of MAD(y) and sum of y_1 residuals (BIAS) as listed in Table 7.

The application of the infinite dilution test of Kojima (1990) includes the calculation of the excess Gibbs free energy from the experimental data, and the extrapolation to infinite dilution using smoothing functions. The extrapolated values are then compared with those extrapolated of $\ln \gamma_1$ and $\ln \gamma_2$ at infinite dilution, and thermodynamic consistency is achieved if the values agree to within 30%. The results at 60 kPa are shown in Figure 5 where $G^{\rm E}/RTx_1x_2$, $\ln \gamma_1$, and $\ln \gamma_2$ have been plotted vs x_1 . Deviations of the extrapolated values at the three pressures (Table 7) were within the method tolerance.



Figure 5. Infinite dilution test for the water (1) + 1-propanol (2) binary system at constant pressure, P = 60 kPa: (\bigcirc) ln γ_1 ; (\diamondsuit) ln γ_2 ; (\square) G^{E}/RTx_1x_2 ; (-) NRTL (α_{12} adjustable) model; (- - -) polynomial regression; (+) excluded points.

The L-W method of Wisniak requires the evaluation of the integrals L and W as described in the paper by Wisniak (1993), and values of the deviation D defined as

$$D = 100 \frac{|L - W|}{L + W} \tag{4}$$

less than 3-5 indicate thermodynamic consistency. As can be observed in Table 7, experimental VLE data were also found consistent according to the Wisniak test.

The activity coefficients were correlated with the Margules, Van Laar, Wilson, NRTL, and UNIQUAC equations (Gmehling and Onken, 1977). To fit the binary parameters, a nonlinear optimization method was used to minimize the function

Table 7. Thermodynamic Consistency Tests

$$F = \sum_{n} \sum_{i} \left(\frac{\gamma_{\text{exptl}} - \gamma_{\text{calcd}}}{\gamma_{\text{exptl}}} \right)_{i,n}^{2}$$
(5)

where *n* is the number of data points. For the NRTL model, two alternatives were selected: using the recommended value for α_{12} of 0.30 (Renon and Prausnitz, 1968) or adjusting α_{12} in the fitting process. The parameters A_{12} , A_{21} , and α_{12} for the correlation equations, mean absolute deviations, and activity coefficients at infinite dilution γ_i° are given in Table 8. The systems show positive deviations from ideality, as can be observed from the activity coefficients at infinite dilution. At the three pressures, Van Laar, Wilson, NRTL with $\alpha_{12} = 0.30$, and UNIQUAC models yield similar deviations between experimental and calculated vapor compositions and temperatures, while Margules shows greater deviations in both variables. Better results were obtained with the NRTL equation using adjustable α_{12} .

Literature Cited

- Aucejo, A.; González-Alfaro, V.; Montón J. B.; Vázquez, M. I. Isobaric Vapor-Liquid Equilibria of Trichloroethylene with 1-Propanol and 2-Propanol at 20 and 100 kPa. *J. Chem. Eng. Data* **1995**, *40*, 332– 335.
- Christensen, C.; Gmehling, J.; Rasmussen, P.; Weidlich, U. *Heats of Mixing Data Collection*, Chemistry Data Series; DECHEMA: Frank-furt/Main, 1984; Vol. III, Part 1.
- Fredenslund, Aa.; Gmehling, J.; Rasmussen, P. Vapor-Liquid Equilibria Using UNIFAC; Elsevier: Amsterdam, 1977.
- Gmehling, J., Onken, U., Eds. Vapor-Liquid Equilibrium Data Collection; Chemistry Data Series; DECHEMA: Frankfurt/Main, 1977; Vol. 1, Part 1.
- Gmehling, J., Onken, U., Eds. Vapor-Liquid Equilibrium Data Collection, Chemistry Data Series; DECHEMA: Frankfurt/Main, 1981; Vol. 1, Part 1a.
- Jackson, P. L.; Wilsak, R. A. Thermodynamic Consistency Tests Based on the Gibbs-Duhem Equation Applied to Isothermal, Binary Vapor-

Liquid Equilibrium Data: Data Evaluation and Model Testing. *Fluid Phase Equilib.* **1995**, *103*, 155–197.

- Kojima, K.; Moon, H. M.; Ochi, K. Thermodynamic Consistency Test of Vapor-Liquid Equilibrium data. *Fluid Phase Equilib.* 1990, 56, 269–284.
- Morrison, J. F.; Baker, J. C.; Meredith, H. C., III; Newman, K. E.; Walter, T. D.; Massie, J. D.; Perry, R. L.; Cummings, P. T. Experimental Measurements of Vapor-Liquid Equilibrium in Alcohol/ Water/Salt Systems. *J. Chem. Eng. Data* **1990**, *35*, 395–404.
- Munday, E. B.; Mullins, J. C.; Edie, D. D. Vapor Pressure Data for Toluene, 1-Pentanol, 1-Butanol, Water, and 1-Propanol and for the Water and 1-Propanol System from 273.15 to 323.15 K. J. Chem. Eng. Data 1980, 25, 191–194.
- Pitzer, K. S.; Curl, R. F. The Volumetric and Thermodynamic Properties of Fluids III. Empirical Equation for the Second Virial Coefficient. J. Am. Chem. Soc. 1957, 79, 2369–2370.
 Renon, H.; Prausnitz, J. M. Local Compositions in Thermodynamic
- Renon, H.; Prausnitz, J. M. Local Compositions in Thermodynamic Excess Functions for Liquid Mixtures. AIChE J. 1968, 14, 134– 144.
- TRC Thermodynamic Tables Hydrocarbons; Thermodynamic Research Center, The Texas A&M University System: College Station, TX, 1994.
- Tsonopoulos, C. An Empirical Correlation of the Second Virial Coefficients. AIChE J. 1974, 20, 263–272.
- Van Ness, H. C.; Byer, S. M.; Gibbs, R. E. Vapor-Liquid Equilibrium: Part I. An Appraisal of Data Reduction Methods. AIChE J. 1973, 19, 238–244.
- Walas, S. M. Phase Equilibria in Chemical Engineering, Butterworth: London, 1985.
- Wisniak, J. A New Test for the Thermodynamic Consistency of Vapor-Liquid Equilibrium. Ind. Eng. Chem. Res. 1993, 32, 1531–1533.
- Zielkiewicz, J.; Konitz, A. (Vapor + liquid) equilibria of (N,N-dimethylformamide + water + propan-1-ol) at the temperature 313.15 K. *J. Chem. Thermodyn.* **1991**, *23*, 59–65.

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