Liquid Densities at Elevated Pressures of 1-Alkanols from C_1 to C_{10}: A Critical Evaluation of Experimental Data

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The published experimental data for 1-alkanols are critically reviewed, and the parameters of the Tait equation are given. This equation allows the calculation of smoothed values of the liquid density of 1-alkanols from methanol to 1-decanol over a temperature and pressure range.

Introduction

This work, the results of which are summarized here, is a continuation of the critical evaluation of published $P-\varrho-T$ data for liquid 1-alkanols. Recently, the critical evaluation of the published density data obtained at atmospheric pressure or along a saturation curve for 1-alkanols and n-alkanes was performed and published (*58*) as a part of the IUPAC Project on Vapor-Liquid Equilibria in 1-Alkanol + n-Alkane Mixtures whose aim was to prepare a collection of critically evaluated experimental data on various properties of these mixtures and the respective pure components. In this paper published densities for 1-alkanols at pressures higher than either 0.1 MPa or the saturation pressure are reviewed and critically evaluated.

Sources of Data

The original experimental data (5558 data points) processed were extracted from a source database developed and installed under the FOXPRO 2.0 environment in our laboratory. The database contains published experimental data for several properties of pure liquids (density, volume, compression, compressibilities, expansivities, compressibility factor, speed of sound) as a function of temperature and pressure compiled from the literature. Besides the measured values and their uncertainties, a large amount of auxiliary information on experimental methods used, descriptions of experimental devices, and information on substances measured (sample source, purification, purity, etc.), which may also be displayed during the process of a critical assessment and evaluation of the data, are recorded in the database.

The characteristics of data that were available in the database for density and related quantities (molar and specific volumes, volume ratio, compression) of 1-alkanols are summarized in Table 1. The displayed temperature ranges and numbers of experimental values are restricted up to the critical temperature (for the critical temperatures selected see ref 58); i.e., only subcritical data were taken from the source database.

Treatment of Data

Since the experimental accuracy of liquid densities at elevated pressures is generally poorer than that at 0.1 MPa or at saturation, the influence of differences between temperature scales on the results was found to be negligible, and therefore no adjustments were made for the different temperature scales. Moreover, rarely have researchers indicated their temperature scale (see Table 1).

Density values were not corrected with respect to impurities (see Table 1); however, contributions reflecting the purity of the samples (generally higher for samples of unknown purity) were introduced into the estimations of the overall experimental uncertainties. The sources and quality of the materials and the methods of the purification were also taken into account.

Estimation of the overall uncertainties necessary for the evaluation of weighing factors (see eq A-2 in the Appendix) for all experimental density values was the final step of the treatment of the data. The uncertainties and other auxiliary information taken from the source database were employed. The uncertainties of the data given by the researchers were preferred; however, care was taken not to interpret the reproducibility of the measurements, often declared as the accuracy of data, as the overall experimental uncertainty. In cases where the uncertainties were not given by the researchers crude estimations were made taking into account the method of the measurements, the accuracy of the temperature and pressure measurements, and the purity of the samples. Unfortunately those sets of information are more or less incomplete in the literature, and moreover the influence of some of these factors (e.g., unspecified impurities) is not known in most cases. Therefore, the experimental uncertainties of a large number of experimental density values were estimated intuitively. Significantly smaller uncertainties were attributed to those measurements that were regarded as high quality (the careful experimental procedure, the samples of high purity) compared to those measurements reported without any additional information. During the process of evaluation (see below) it was, however, possible to adjust statistical weights through the parameter μ_j (see eq A-2 in the Appendix) and thus to correct the over- and underestimated uncertainties.

All available data as reported in Table 1 were taken into the evaluation procedure with the exception of data at extreme pressures obtained by a shock wave method (*10*).

Method of Data Evaluation

Available experimental data on the compressed liquid density of alkanols were fitted by a Tait equation with temperature-dependent parameters $C(T)$ and $B(T)$ written in the form

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Table 1. Characteristics of Data Sets: Overall Numbers of Data Points, N_p, Temperature and Pressure Ranges within the Liquid State, T_{min}, T_{max}, P_{min}, and P_{max}, Experimental Methods Used, Types of Data, and Purities of Measured Samples

ref	N_p	T_{min}/K	T_{max}/K	P_{min}/MPa	P_{max}/MPa	meth[a]	data type[b]	sample purity[c]/%
				Methanol				
1	119	293.15	353.15	49.0	1176.8	vs	D	
3	1	298.15	298.15	100.0	100.0	nd	D	
4	29	298.15	448.15	980.7	4903.3	vs	D	
7	8	303.15	303.15	10.1	202.6	va	D	
8	61	293.85	508.35	1.0	100.0	bu	D	
8	68	293.15	508.15	2.0	101.3	bu	S	
10	2	288.15	297.15	4660.0	10950.0	sw	D	
12	13	453.15	509.53	2.7	21.8	cl	D	99.95[e]
15	1	303.15	303.15	101.3	101.3	va	D	>99[e]
17	125	298.15	473.15	50.0	800.0	cl	D	
19	9	298.15	298.15	10.1	141.9	va	D	
20	1	298.15	298.15	101.3	101.3	va	D	
29	59	283.15	348.15	2.1	207.5	cl	D	>99.5v[d]
32	236	223.10	298.10	10.0	300.0	vs	D	
35	164	298.12	488.86	0.5	103.8	hp	D[f]	99.98[d]
39	12	398.15	448.15	1.1	4.0	nd	D	99.66w[d]
40	56	378.16	508.19	2.5	34.2	hp	D[f]	99.9[d]
42	8	323.00	398.00	50.0	200.0	vs	D	
43	59	283.15	348.15	2.1	207.5	cl	D	>99.9w[e]
44	98	273.15	333.15	20.0	280.0	ul	C	99.8m[e]
50	67	183.20	213.20	10.0	300.0	vs	S	99.9a[e]
51	98	203.15	263.15	20.0	280.0	ul	C	99.8m[e]
57	56	207.36	297.30	0.4	51.5	cs	D[g]	99.997a[e]
60	64	205.09	321.07	0.5	51.3	cs	D[g]	99.97a[e]
total:	1414	183.20	509.53	0.4	10950.0			
				Ethanol				
1	119	293.15	353.15	49.0	1176.8	vs	D	
4	8	298.15	348.15	980.6	2814.5	vs	D	
5	1	298.15	298.15	101.3	101.3	va	D	
7	1	303.15	303.15	101.3	101.3	va	D	
9	4	298.15	298.15	88.2	386.3	va	D	
10	2	294.15	299.15	4730.0	11040.0	sw	D	
13	66	473.15	513.15	8.2	68.9	cl	S	99.95[e]
15	1	303.15	303.15	101.3	101.3	va	D	>99[e]
18	4	273.15	363.15	2.7	22.0	vs	D	
19	9	298.15	298.15	10.1	141.8	va	D	
20	1	298.15	298.15	101.3	101.3	va	D	
21	44	298.15	323.15	7.0	310.3	va	D	99.5v[d]
22	6	298.15	298.15	36.9	203.8	vs	D	99.9[d]
28	74	200.00	500.00	1.0	50.0	bu	D	99.84[e]
30	41	298.20	348.20	9.9	196.2	vl	D	99.9m[e]
36	5	298.15	298.15	50.0	250.0	vb	D	
43	44	298.15	323.15	7.0	310.4	cl	D	>99.9w[e]
45	98	273.15	333.15	20.0	280.0	ul	C	>99.7v[e]
52[h]	9	298.15	298.15	2.0	33.8	mo	D	>99.8[d]
53[h]	9	298.15	298.15	2.0	33.8	mo	D	>99.5
54	112	193.15	263.15	20.0	280.0	ul	C[f]	>99.7v[e]
total:	678	193.15	513.15	1.0	11040.0			
				1-Propanol				
1	119	293.15	353.15	49.0	1176.8	vs	D	
4	18	298.15	348.15	980.6	4903.3	vs	D	
5	1	298.15	298.15	101.3	101.3	va	D	
11	83	297.15	535.45	0.5	51.6	bu	D	
11	95	293.15	533.15	2.0	50.6	bu	S	
23	123	289.87	531.49	1.0	49.1	bu	D	
27	21	194.45	273.15	1.0	49.1	bu	D	
28	83	200.00	520.00	1.0	50.0	bu	D	99.84[e]
33	120	150.00	370.00	2.5	80.0	nd	S	99.55[d]
34	17	423.15	523.15	0.6	5.1	nd	D	99.99w[d]
43	48	283.15	348.15	15.6	207.6	cl	D	>99.9w[e]
49	8	298.15	298.15	20.0	200.0	va	D	
52	9	298.15	298.15	2.0	33.8	mo	D	>99.5[e]
total:	745	150.00	535.45	0.5	4903.3			
				1-Butanol				
2	30	273.15	368.15	49.0	1176.8	vb	D	
4	5	298.15	348.15	980.6	1863.2	vs	D	
11	87	292.75	553.85	0.2	50.0	bu	D	
11	84	293.15	553.15	2.0	50.6	bu	S	
16	90	298.15	333.15	1.0	150.0	vl	S	99.9m[d]
25	92	297.31	558.47	1.0	49.1	bu	D	99.84[e]
27	15	194.61	236.36	1.0	49.1	bu	D	

Table 1. (Continued)

ref	N_p	T_{min}/K	T_{max}/K	P_{min}/MPa	P_{max}/MPa	meth[a]	data type[b]	sample purity[c]/%
				1-Butanol				
28	103	200.00	560.00	1.0	50.0	bu	D	99.84[e]
37	18	298.14	399.81	0.2	20.5	mo	D	
43	48	283.15	348.15	15.5	206.1	cl	D	>99.9w[e]
total:	572	194.61	560.00	0.2	1863.2			
				1-Pentanol				
4	18	298.15	348.15	980.6	4903.3	vs	D	
10	2	292.15	296.15	5090.0	11590.0	sw	D	
24	89	292.30	499.50	4.9	58.8	bu	D	99.5[e]
26	97	232.94	581.53	1.0	49.1	bu	D	99.9[e]
59	50	323.15	373.15	1.0	10.0	mo	D[f]	>99m[d]
total:	256	232.94	581.53	1.0	11590.0			
				1-Hexanol				
2	22	273.15	368.15	49.0	686.4	vb	D	
24	107	290.10	588.10	4.9	58.8	bu	D	99.5[e]
47	36	298.15	348.15	0.15	40.0	mo	D	>99[d]
59	50	323.15	373.15	1.0	10.1	mo	D[f]	>99m[d]
total:	215	273.15	588.10	0.15	686.4			
				1-Heptanol				
31	80	273.15	575.07	1.1	49.1	bu	D	99.43[e]
59	50	323.15	373.15	1.0	10.0	mo	D[f]	>99.5m[d]
total:	130	273.15	575.07	1.0	49.1			
				1-Octanol				
41	138	283.15	623.15	5.0	78.8	bu	S	99.62[e]
47	34	298.15	348.15	0.15	40.2	mo	D	97[d]
56	4	298.15	298.15	50.0	200.0	va	D	
59	50	323.15	373.15	1.0	10.0	mo	D[f]	>99.5m[d]
total:	226	283.15	623.15	0.15	200.0			
				1-Nonanol				
14	190	288.65	624.75	5.1	50.6	bu	D	
14	920	293.15	623.15	0.2	50.6	bu	S	
59	50	323.15	373.15	1.0	10.0	mo	D[f]	>98m[d]
total:	1160	288.65	624.75	0.2	50.6			
				1-Decanol				
6	22	298.15	353.15	19.6	137.3	nd	D	
47	28	298.15	348.15	1.0	40.2	mo	D	>99[d]
48	112	283.15	623.15	5.0	78.8	bu	D	98.2[e]
total:	162	283.15	623.15	1.0	137.3			

[a] Methods used for measurements: bu, buoyancy methods; cl, constant-volume cell with liquid piston; cs, constant-volume cell with solid piston; hp, high-pressure pycnometer; mo, mechanical oscillator methods; nd, not described or stated in the reference; sw, shock wave method; ul, ultrasound velocity method; va, Aime's method; vb, variable-volume cell with bellows; vl, variable-volume cell with liquid piston; vs, variable-volume cell with solid piston. For the classification and description of the methods, see ref 38. [b] D, direct experimental data; S, smoothed data; C, densities calculated from ultrasound velocities. [c] No letter, unspecified percent; a, mass percent assuming water as an impurity; m, mole percent; v, volume percent; w, mass percent. [d] Purity of source material is given only. [e] Final purity of the sample. [f] IPTS-68 declared by the researchers. [g] ITS-90 declared by the researchers. [h] These two references presented the same data.

$$\varrho(T,P,\vec{c},\vec{b}) = \frac{\varrho(T,P_{\mathrm{ref}}(T))}{1 - C(T,\vec{c})\ln\left[\dfrac{B(T,\vec{b}) + P}{B(T,\vec{b}) + P_{\mathrm{ref}}(T)}\right]} \quad (1)$$

where

$$C(T,\vec{c}) = \sum_{i=0}^{N_C} c_i[(T - T_0)/100]^i, \quad \vec{c} = \{c_i\} = \{c_0, ..., c_{N_C}\} \quad (2)$$

$$B(T,\vec{b}) = \sum_{i=0}^{N_B} b_i[(T - T_0)/100]^i, \quad \vec{b} = \{b_i\} = \{b_0, ..., b_{N_B}\} \quad (3)$$

and T_0 is a parameter with a preselected fixed value for which $C(T_0) = c_0$ and $B(T_0) = b_0$ hold.

The Tait equation describes a compression only, i.e., a change in density or volume relative to a selected reference point defined by a reference pressure at the same temperature,

$$1 - \frac{\varrho(T,P_{\mathrm{ref}}(T))}{\varrho(T,P)} = 1 - \frac{V(T,P)}{V(T,P_{\mathrm{ref}}(T))} = C(T)\ln\left[\frac{B(T) + P}{B(T) + P_{\mathrm{ref}}(T)}\right] \quad (4)$$

and thus, to calculate density at a given T and P, the values of the density at the same temperature and the reference pressure, $\varrho(T,P_{\mathrm{ref}}(T))$, and a reference pressure itself, $P_{\mathrm{ref}}(T)$, are therefore required. Within the work the reference lines, $\varrho(T,P_{\mathrm{ref}}(T))$ and $P_{\mathrm{ref}}(T)$, were selected as follows. At temperatures below the normal boiling temperature and densities at atmospheric pressure ($P_{\mathrm{ref}} = 0.101\ 325$ MPa) were used, while for higher temperatures the values along the saturation curve, i.e., saturated liquid densities and saturated vapor pressures, were employed.

If the densities at atmospheric pressure or at saturation for the same sample were also reported along with com-

Table 2. Parameters c_i, b_i, and T_0 of Eq 1, Temperature and Pressure Ranges[a] of Validity, T_{min}, T_{max}, P_{min}, and P_{max}, Absolute, RMSD, and Relative, $RMSD_r$, Root Mean Square Deviations, Biases, bias, Numbers of Data Points, N_p, ±, and Weighted Standard Deviations, s_W[b]

	methanol(I)	methanol(II)	ethanol(I)	ethanol(II)		methanol(I)	methanol(II)	ethanol(I)	ethanol(II)
c_0	0.099374	0.097871	0.095279	0.095027	T_{max}/K	342.83	483.18	333.15	473.15
c_1/K^{-1}		−0.005322	−0.004757	−0.005735	P_{min}/MPa	0.5	0.5	20.0	1.0
b_0/MPa	67.4341	66.0258	84.8525	84.7620	P_{max}/MPa	280.0	280.0	280.0	280.0
b_1/(MPa·K^{-1})	−49.6697	−54.4554	−64.7513	−66.0861	RMSD/(kg·m^{-3})	0.345	0.440	0.203	0.678
b_2/(MPa·K^{-2})	18.5137	12.0902	16.8197	13.5273	$RMSD_r$/%	0.040	0.059	0.023	0.099
b_3/(MPa·K^{-3})	16.6876	−4.4449	1.9753	−1.8708	bias/(kg·m^{-3})	−0.024	−0.019	0.034	−0.046
b_4/(MPa·K^{-4})	11.3084	1.2057		0.2375	N_p	336	471	209	259
T_0/K	323.15	323.15	298.15	298.15	±	2	−5	23	35
T_{min}/K	183.20	183.20	193.15	193.15	s_W	0.792	1.022	0.954	1.106

	1-propanol(I)	1-propanol(II)	1-butanol	1-pentanol		1-propanol(I)	1-propanol(II)	1-butanol	1-pentanol
c_0	0.092058	0.086148	0.090982	0.095200	T_{max}/K	348.15	524.16	523.88	549.51
c_1/K^{-1}		−0.003058	−0.003268		P_{min}/MPa	2.0	2.0	0.2	1.0
b_0/MPa	80.6058	71.5896	84.0516	80.5273	P_{max}/MPa	207.6	1176.8	1863.3	4903.3
b_1/(MPa·K^{-1})	−52.8723	−56.4067	−62.3764	−55.8129	RMSD/(kg·m^{-3})	0.423	0.938	0.457	1.235
b_2/(MPa·K^{-2})	5.5466	7.9726	11.4866	6.1070	$RMSD_r$/%	0.049	0.109	0.053	0.149
b_3/(MPa·K^{-3})		0.7782	−1.3657	1.1574	bias/(kg·m^{-3})	0.003	0.006	0.118	0.301
b_4/(MPa·K^{-4})			0.4606		N_p	66	285	124	216
T_0/K	323.15	323.15	323.15	348.15	±	−6	−3	10	42
T_{min}/K	283.15	170.00	194.61	232.94	s_W	0.862	0.855	0.734	1.039

	1-hexanol	1-heptanol	1-octanol(I)	1-octanol(II)		1-hexanol	1-heptanol	1-octanol(I)	1-octanol(II)
c_0	0.109989	0.098887	0.089655	0.093973	T_{max}/K	490.20	575.03	373.15	623.15
c_1/K^{-1}					P_{min}/MPa	0.2	1.0	0.2	0.2
b_0/MPa	117.5853	142.2713	102.0489	108.0594	P_{max}/MPa	58.8	49.1	200.0	200.0
b_1/(MPa·K^{-1})	−73.9423	−73.8798	−61.7326	−69.2343	RMSD/(kg·m^{-3})	0.842	0.617	0.129	1.205
b_2/(MPa·K^{-2})	9.0136	11.1140	14.2678	14.6384	$RMSD_r$/%	0.115	0.091	0.016	0.177
b_3/(MPa·K^{-3})	3.5726	−0.5325		−1.1325	bias/(kg·m^{-3})	0.026	0.083	0.035	0.246
b_4/(MPa·K^{-4})					N_p	157	124	87	202
T_0/K	323.15	273.15	323.15	323.15	±	15	44	37	68
T_{min}/K	290.10	273.15	298.15	283.15	s_W	1.010	1.076	1.082	0.943

	1-nonanol(I)	1-nonanol(II)	1-decanol		1-nonanol(I)	1-nonanol(II)	1-decanol
c_0	0.043024	0.055031	0.114405	T_{max}/K	373.15	431.15	623.15
c_1/K^{-1}				P_{min}/MPa	1.0	1.0	1.0
b_0/MPa	48.3935	64.0089	160.9127	P_{max}/MPa	10.0	50.6	78.8
b_1/(MPa·K^{-1})	−16.2848	−54.4799	−88.8559	RMSD/(kg·m^{-3})	0.139	1.086	2.572
b_2/(MPa·K^{-2})	−93.6748	58.6667	19.6783	$RMSD_r$/%	0.017	0.137	0.348
b_3/(MPa·K^{-3})	154.3116	−42.4205	−1.9272	bias/(kg·m^{-3})	0.005	0.390	0.279
b_4/(MPa·K^{-4})				N_p	49	118	122
T_0/K	323.15	323.15	298.15	±	3	52	14
T_{min}/K	323.15	288.85	283.15	s_W	2.065	1.554	0.881

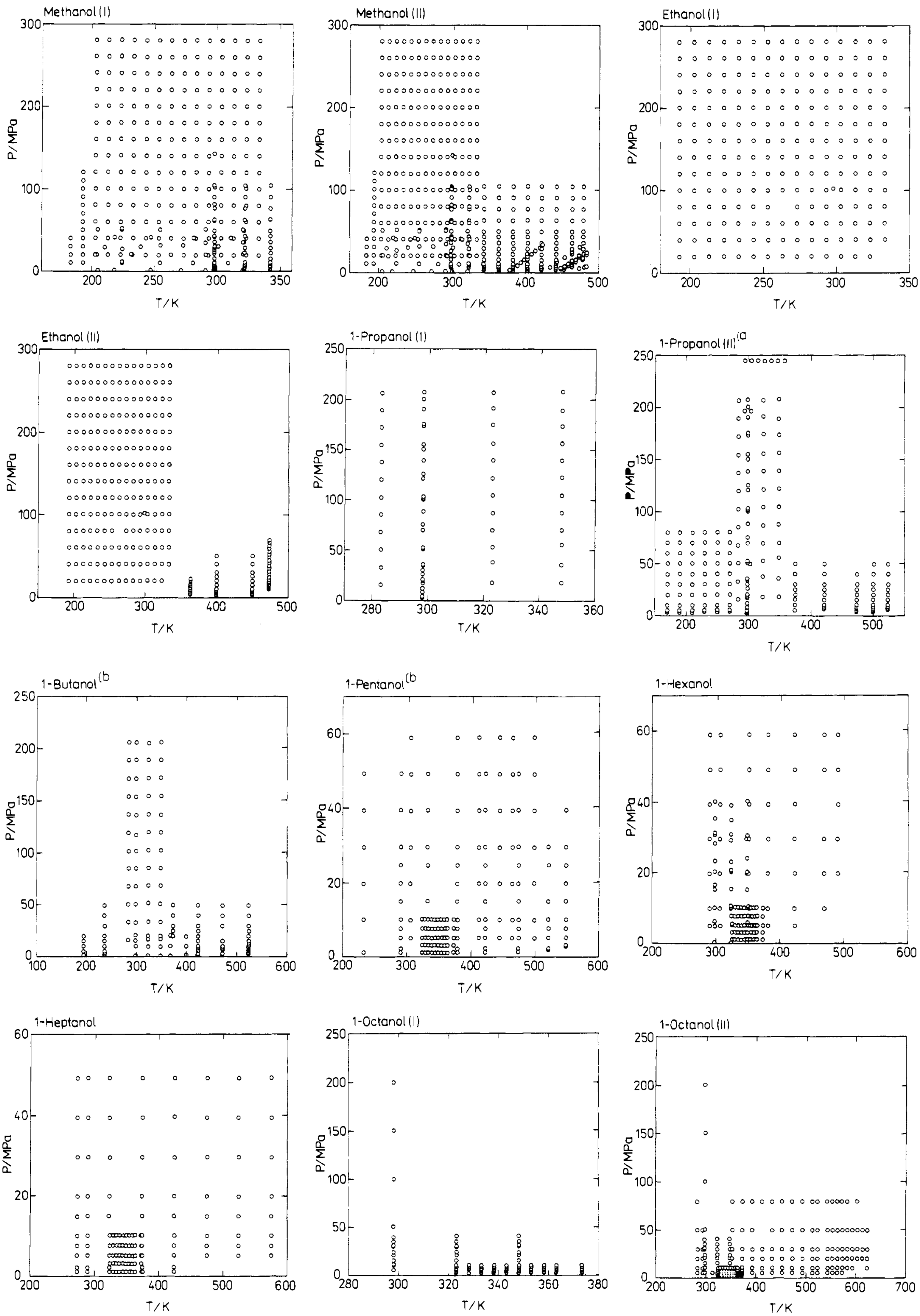
[a] The low limit of pressure ranges is 0.1 MPa or a saturation pressure (whichever is higher) for all fits; P_{min} is the lowest pressure in a particular set of compressed liquid density data retained for the correlation. [b] The fits marked I cover limited $T-P$ ranges; the fits II and nonmarked ones cover as large a $T-P$ area as possible (see also Table 3 and Figure 1).

pressed liquid density data, then those values were preferably used for the reference density, $\varrho(T,P_{ref})$. Their use substantially decreases the influence of both systematic errors and sample impurities since the compression values as reported by the researchers are then smoothed by eq 1. If the reference values were not available in the original source, then densities obtained from the critical evaluation (*58*) were employed in the correlations. Saturated vapor pressures were calculated from the smoothing functions given by Ambrose and Walton (*46*).

Adjustable parameters $\vec{c}$ and $\vec{b}$ were obtained by the weighted least-squares method. A brief summary of the mathematical procedure along with definitions of some statistical quantities is presented in the Appendix. The calculation of the parameters was repeated several times for each compound, and the results obtained were critically reviewed after each calculation. Between the consecutive calculations it was possible to reject and to reinclude individual experimental density values and whole density data subsets (i.e., the sets of values taken from individual literature sources), and to change statistical weights of the retained data. The calculations were repeated until the final fit was obtained where the deviations between retained experimental and smoothed values were roughly equal to the experimental uncertainties modified by means of the parameter μ_j (see eq A-2 in the Appendix), i.e., where the weighted standard deviation was close to unity.

In some original sources, the data are available in two forms (see Table 1), i.e., as direct experimentally observed data (D) given at experimental temperatures and pressures and smoothed values (S) which are usually presented at round values of temperature and pressure. In such cases both data sets were extracted from the source database and taken into the procedure; however, the direct data were always preferred for the critical evaluation, while smoothed values representing the direct ones were excluded from the correlations (see Table 3 below). Smoothed values were retained in those cases only where the direct data were not available.

Generally those data that were randomly scattered along a smoothing function, exhibited deviations within the experimental uncertainty, and were in agreement with other reasonably reliable data were retained in the correlations while others were rejected. If in a particular temperature and pressure range, several sets of data were available, then even those with either large random or systematic deviations but still within the experimental uncertainty were usually rejected while the data of better


Methanol (I)
Methanol (II)
Ethanol (I)
Ethanol (II)
1-Propanol (I)
1-Propanol (II)(a
1-Butanol(b
1-Pentanol(b
1-Hexanol
1-Heptanol
1-Octanol (I)
1-Octanol (II)
P/MPa
T/K

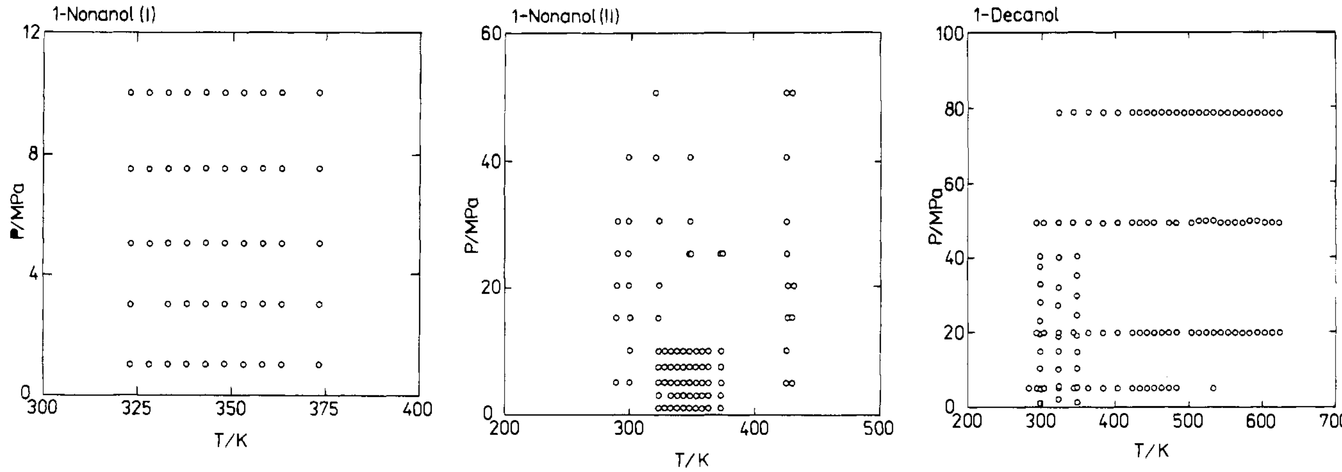


Figure 1. Temperature and pressure coordinates of data points retained for the correlations: (a) Data of ref 1 over 250 MPa are not displayed, (b) Data of ref 4 are not displayed.

quality were retained. The consistency of density values at 0.1 MPa or at saturation (if reported by the researchers) with critically evaluated data (*58*) was also taken into account during the evaluation where possible (the origin of the reference density values used in the correlation is indicated in column RD of Table 3).

Results

Table 2 records the values of the parameters of eq 1 for each alkanol along with some statistical information in the fits. Temperature and pressure ranges of validity of the fits given in the table are informative only and allow extrapolation using eq 1 with the parameters from Table 2 beyond $P-T$ areas shown in Figure 1 to be avoided. That plot provides crude information on the distribution of the retained data points in the $P-T$ area for each fit performed.

For several alkanols the results of two fits (denoted as I and II) are presented. Fit I covers a limited temperature range, mostly from low temperature to temperatures not far from ambient. Fit II was obtained for as wide a temperature interval as possible. In several cases the pressure range of fit I was also less than that for fit II. Two fits were performed for those alkanols for which the accuracy and reliability of available data at higher temperatures were significantly poorer than at lower temperatures, and thus the inclusion of high-temperature data influenced the quality of the correlation at lower temperatures. However, the maximum differences in density between fits I and II were found to be less than 0.4 $kg{\cdot}m^{-3}$ in the overlapping $T-P$ range, and most of the differences were less than 0.2 $kg{\cdot}m^{-3}$ (0.03%), which is less than the experimental uncertainty of most of the data in the lower temperature range.

Table 3 summarizes some statistical information derived from the fits. Only those data subsets for which the temperature and pressure ranges are displayed in the table were retained in the correlations. The statistical characteristics of these subsets refer only to the data points retained in the correlation. On the other hand, the characteristics of the rejected data subsets, i.e., those for which no T and P ranges are given in the table, illustrate the deviations of the rejected data points from eq 1, but only for those values within $T-P$ areas of the retained data (see Figure 1).

In several cases the data with systematic deviations or data of poorer accuracy, but with deviations well below 10 $kg{\cdot}m^{-3}$, were retained to widen the temperature and pressure range of the correlation. It concerned particularly data sets that comprised data points in either the low- or high-temperature regions and at high pressures for which no other data were available. The effect of these data was, however, always checked, and their inclusion into a correlated data set was allowed only in those cases where there was a negligible change (with respect to experimental uncertainties) of the representation of data in the other ranges. Examples of such inclusion of data are from the following references (see Table 3): 12 and 50 for methanol(II), 13 and 28 for ethanol(II), 1 for 1-propanol(II), 4 and 26 for 1-pentanol, 24 for 1-hexanol, 41 for 1-octanol(II), 14 for 1-nonanol(II), and 48 for 1-decanol.

Data points at high temperatures of some retained data sets were, however, also rejected in those cases where deviations from the Tait equation exceeded 10 $kg{\cdot}m^{-3}$ and it was not possible to improve the fit by additional parameters b_i and c_i. Thus, the $P-T$ ranges of some fits do not cover the entire original range of retained data sets. There might be several reasons for those large deviations: lower accuracy of the data and systematic errors in the measured variables at very high temperatures and pressures, decomposition of the substance at high temperatures, and at last a poorer performance of the Tait equation in the vicinity of the gas–liquid critical point.

De Reuck and Craven (*55*) have developed an equation of state for methanol which is based on available data on thermodynamic properties. The density values calculated from the Tait equation (Table 2, methanol(II)) using the reference density line from ref 58 and saturated vapor pressures from ref 46 are in very good accordance with values presented in part 2 of the report by de Reuck and Craven. The deviations are below 0.1% in the entire $T-P$ area of validity of the Tait equation (see the plot for methanol(II) in Figure 1), mostly lower than 0.05%, except for temperatures and pressures above 480 K and 50 MPa where the Tait equation yields density values which are lower by up to 0.3% than those by de Reuck and Craven. The comparison also revealed relatively good agreement in the rectangular $T-P$ area where no experimental $P-\varrho-T$ data were available, i.e., in the range from 330 to 480 K and from 100 to 280 MPa (see the plot for methanol(II) in Figure 1). Here the deviations between densities calculated from the Tait equation and those reported in ref 55 are positive, being the highest (0.5%) in the area around 460 K and pressures close to 280 MPa and negative along the isotherms around 480 K increasing up to −0.4% at 250 MPa. The extrapolation ability of the Tait equation in other $T-P$ areas, i.e., above 480 K and 280 MPa, has not been examined.

Appendix

Adjustable parameters $\vec{c}$ and $\vec{b}$ of function 1 were obtained by minimizing the objective function

$$\phi(\vec{c},\vec{b}) = \sum_{j=1}^{N_P} w_j[\varrho_j - \varrho(T_j,P_j,\vec{c},\vec{b})]^2 \qquad \text{(A-1)}$$

where ϱ_j, T_j, P_j is the jth experimental data point, $\varrho(T_j,P_j,\vec{c},\vec{b})$ is the value calculated from function 1 with parameters $\vec{c}$ and $\vec{b}$ for the values T_j and P_j, and N_P is the number of experimental values of density taken into the correlation. Adjustable parameters were calculated by the Marquardt algorithm in double precision to minimize the influence of rounding errors.

Statistical weights, w_j, in eq A-1 were defined as

$$w_j = \mu_j/(\delta\varrho_j)^2 \qquad \text{(A-2)}$$

where $\delta\varrho_j$ is the experimental uncertainty taken from the source database and either given by the researchers (preferably) or estimated by a compiler for the jth density value in a correlated data set and μ_j is a parameter the value of which can be changed between the consecutive correlations to adjust the statistical weights (initial values of all μ_j are set to unity). Thus, the original uncertainties may be changed so that the modified uncertainty of the jth data point is equal to $\delta\varrho_j/\mu_j^{1/2}$. This allows statistical weights of individual data points to be changed while the values of the experimental uncertainties as stored in the source database of original experimental data remain unchanged during the correlations.

The weighted standard deviations, s_w, of the fit is given by

$$s_w = [\phi/(N_p - N_C - N_B - 2)]^{1/2} \qquad \text{(A-3)}$$

Standard deviations of the values of density, $s(\varrho)$, calculated from eq 1 may be estimated for each selected

Table 3. Statistical Characteristics of Individual Data Sets for the Fits in Table 2: Temperature and Pressure Ranges Taken into the Correlations, T_{min}, T_{max}, P_{min}, and P_{max}, Absolute, RMSD, and Relative, RMSD$_r$, Root Mean Square Deviations, Biases, bias, Numbers of Data Points, N_p, ±, and Origin of the Reference Density Values Used in the Correlations, RD[a]

ref	T_{min}/K	T_{max}/K	P_{min}/MPa	P_{max}/MPa	RMSD/(kg·m^{-3})	RMSD$_r$/%	bias/(kg·m^{-3})	N_p	±	RD[a]
					Methanol(I)					
1					7.990	0.925	−7.778	25	−25	o
3	298.15	298.15	100.0	100.0	0.141	0.016	0.141	1	1	o
4								0	0	e
7					3.322	0.396	−3.136	8	−8	o
8					2.711	0.323	−1.354	11	−1	e
8[b]					2.962	0.352	−2.133	13	−11	(o)
10								0	0	o
12								0	0	e
15					3.227	0.380	−3.227	1	−1	o
17					3.895	0.447	−3.799	20	−20	e
19	298.15	298.15	10.1	141.9	0.210	0.026	0.046	9	1	o
20	298.15	298.15	101.3	101.3	0.164	0.019	−0.164	1	−1	o
29					0.637	0.075	−0.189	44	−12	(o)
32					4.910	0.537	−4.371	224	−218	e
35	298.12	342.83	0.5	103.8	0.289	0.035	−0.023	49	7	e
39								0	0	o
40								0	0	e
42					4.121	0.484	−3.977	4	−4	e
43					0.636	0.075	−0.188	44	−12	(o)
44[c]	273.15	333.15	20.0	280.0	0.259	0.029	−0.029	98	−8	e
50[b]	183.20	193.20	10.0	120.0	0.775	0.084	−0.666	16	−16	o
51[c]	203.15	263.15	20.0	280.0	0.200	0.021	0.046	98	14	o
57					3.479	0.416	2.800	55	49	e
60	205.09	321.07	0.5	51.3	0.490	0.059	0.033	64	4	e
					Methanol(II)					
1					9.004	1.064	−8.697	35	−35	o
3	298.15	298.15	100.0	100.0	0.066	0.008	0.066	1	1	o
4								0	0	e
7					3.342	0.399	−3.157	8	−8	o
8					2.389	0.347	−0.067	39	−1	e
8[b]					2.187	0.293	−0.663	48	−12	(o)
10								0	0	o
12	453.15	473.15	2.7	20.1	1.700	0.287	−1.078	3	−1	e
15					3.259	0.384	−3.259	1	−1	o
17					2.831	0.334	−1.488	50	−22	e
19	298.15	298.15	10.1	141.9	0.222	0.027	−0.001	9	1	o
20	298.15	298.15	101.3	101.3	0.239	0.028	−0.239	1	−1	o
29					1.708	0.204	−0.898	59	−23	(o)
32					4.999	0.546	−4.406	224	−218	e
35	298.12	478.62	0.5	103.8	0.423	0.060	−0.137	147	−53	e
39					1.084	0.167	0.331	12	0	o
40	378.16	483.18	2.5	33.2	0.619	0.097	0.153	34	8	e
42					3.679	0.453	−3.421	8	−8	e
43					1.710	0.204	−0.900	59	−21	(o)
44[c]	273.15	333.15	20.0	280.0	0.473	0.052	0.015	98	14	e
50[b]	183.20	193.20	10.0	120.0	0.675	0.073	−0.538	16	−16	o
51[c]	203.15	263.15	20.0	280.0	0.170	0.019	0.099	98	30	o
57[c]					3.539	0.422	2.872	55	49	e
60	205.09	321.07	0.5	51.3	0.410	0.050	0.111	64	12	e

Table 3. (Continued)

ref	T_{min}/K	T_{max}/K	P_{min}/MPa	P_{max}/MPa	RMSD/(kg·m^{-3})	RMSD$_r$/%	bias/(kg·m^{-3})	N_p	±	RD[a]
					Ethanol(I)					
1					1.135	0.135	−1.008	25	−25	o
4								0	0	e
5					1.648	0.195	−1.648	1	−1	o
7					5.345	0.636	−5.345	1	−1	o
9					7.519	0.872	−7.390	2	−2	o
10								0	0	o
13[b]								0	0	e
15					3.235	0.384	−3.235	1	−1	o
18					1.640	0.205	1.615	2	2	e
19					0.367	0.045	0.181	7	5	o
20	298.15	298.15	101.3	101.3	0.172	0.020	−0.172	1	−1	o
21					1.873	0.219	−1.674	39	−39	o
22					1.025	0.120	0.100	6	0	o
28					2.794	0.321	2.500	15	15	e
30					1.102	0.130	1.026	23	23	o
36					0.559	0.065	0.555	5	5	o
43					1.894	0.222	−1.699	39	−39	o
45[c]	273.15	333.15	20.0	280.0	0.234	0.028	0.091	97	31	e
52					0.389	0.048	−0.386	3	−3	o
53					0.389	0.048	−0.386	3	−3	o
54[c]	193.15	263.15	20.0	280.0	0.171	0.019	−0.014	111	−7	o
					Ethanol(II)					
1					2.205	0.268	−1.710	35	−35	o
4								0	0	e
5					1.517	0.179	−1.517	1	−1	o
7					5.220	0.621	−5.220	1	−1	o
9					7.340	0.851	−7.219	2	−2	o
10								0	0	o
13[b]	473.15	473.15	9.7	68.9	2.022	0.302	−1.761	21	−19	e
15					3.110	0.369	−3.110	1	−1	o
18	363.15	363.15	2.8	22.1	0.294	0.040	−0.259	8	−6	e
19					0.393	0.049	0.299	9	7	o
20	298.15	298.15	101.3	101.3	0.041	0.005	−0.041	1	−1	o
21					1.791	0.211	−1.564	42	−42	o
22					1.112	0.129	0.249	6	0	o
28	400.00	450.00	1.0	50.0	0.972	0.146	0.908	21	21	e
30					1.510	0.186	−0.053	41	13	o
36					0.739	0.085	0.735	5	5	o
43					1.811	0.213	−1.588	42	−42	o
45[c]	273.15	333.15	20.0	280.0	0.309	0.036	0.126	97	45	e
52					0.307	0.038	−0.299	9	−9	o
53					0.307	0.038	−0.299	9	−9	o
54[c]	193.15	263.15	20.0	280.0	0.173	0.019	−0.036	111	−5	o
					1-Propanol(I)					
1					4.013	0.468	1.752	24	20	o
4								0	0	e
5	298.15	298.15	101.3	101.3	0.323	0.038	0.323	1	1	o
11					1.060	0.132	−0.520	20	−6	e
11[b]					0.862	0.106	−0.690	22	−18	(o)
23					1.509	0.187	1.470	23	23	e
27								0	0	e
28					1.349	0.166	1.344	10	10	e
33[b]					1.248	0.153	1.159	30	30	o
34								0	0	o
43	283.15	348.15	15.6	207.6	0.433	0.050	−0.020	48	0	o
49	298.15	298.15	20.0	200.0	0.556	0.063	0.269	8	2	o
52	298.15	298.15	2.0	33.8	0.150	0.018	−0.143	9	−9	o
					1-Propanol(II)					
1	293.15	353.15	49.0	1176.8	1.263	0.130	−0.009	92	−6	o
4					1.612	0.160	0.419	2	0	e
5	298.15	298.15	101.3	101.3	0.030	0.003	0.030	1	1	o
11					2.430	0.372	0.001	53	−15	e
11[b]					2.105	0.380	−0.190	85	−31	(o)
23	374.85	524.16	2.1	49.1	0.814	0.122	0.452	67	37	e
27					0.971	0.112	0.915	19	17	e
28					1.737	0.211	1.139	78	48	e
33[b]	170.00	270.00	2.5	80.0	0.330	0.037	0.010	60	4	o
34					3.002	0.510	0.305	12	0	o
43	283.15	348.15	15.6	207.6	0.982	0.117	−0.546	48	−30	o
49	298.15	298.15	20.0	200.0	0.765	0.086	0.177	8	0	o
52	298.15	298.15	2.0	33.8	0.443	0.054	−0.411	9	−9	o

Table 3. (Continued)

ref	T_{min}/K	T_{max}/K	P_{min}/MPa	P_{max}/MPa	RMSD/(kg·m^{-3})	RMSD$_r$/%	bias/(kg·m^{-3})	N_p	±	RD[a]
					1-Butanol					
2					7.219	0.836	−6.395	30	−30	o
4	298.15	348.15	980.7	1863.3	0.808	0.078	−0.006	5	1	e
11					3.133	0.462	1.926	59	39	e
11[b]					4.239	0.677	2.127	68	8	(o)
16[b]					0.907	0.106	0.610	90	46	o
25	373.09	523.88	1.1	49.1	0.222	0.033	−0.017	38	−4	e
27	194.61	236.36	1.1	49.1	0.907	0.104	0.880	15	15	e
28					1.029	0.118	0.100	85	−31	e
37	298.14	399.81	0.2	20.5	0.187	0.024	0.071	18	8	e
43	283.15	348.15	15.5	206.1	0.404	0.047	0.018	48	−10	o
					1-Pentanol					
4	298.15	348.15	980.7	4903.3	2.852	0.238	1.920	18	10	e
10								0	0	o
24	306.50	499.50	4.9	58.8	0.953	0.132	−0.310	70	−24	(o)
26	232.94	549.51	1.1	49.1	1.230	0.181	0.662	78	44	e
59	323.15	373.15	1.0	10.0	0.164	0.021	0.012	50	12	o
					1-Hexanol					
2					37.892	4.791	−37.892	1	−1	(o)
24	290.10	490.20	4.9	58.8	1.243	0.170	0.028	71	1	(o)
47	298.15	348.15	0.2	40.0	0.136	0.017	0.030	36	8	o
59	323.15	373.15	1.0	10.1	0.135	0.017	0.019	50	6	o
					1-Heptanol					
31	273.15	575.03	1.1	49.1	0.790	0.117	0.135	74	34	e
59	323.15	373.15	1.0	10.0	0.136	0.017	0.005	50	10	o
					1-Octanol(I)					
41[b]					0.704	0.086	0.203	24	12	(o)
47	298.15	348.15	0.2	40.2	0.124	0.015	0.089	34	30	o
56	298.15	298.15	50.0	200.0	0.105	0.012	−0.024	4	0	o
59	323.15	373.15	1.0	10.0	0.134	0.017	0.002	49	7	o
					1-Octanol(II)					
41[b]	283.15	623.15	5.0	78.8	1.589	0.234	0.404	115	41	(o)
47	298.15	348.15	0.2	40.2	0.190	0.023	0.126	34	20	o
56	298.15	298.15	50.0	200.0	0.363	0.041	−0.114	4	0	o
59	323.15	373.15	1.0	10.0	0.138	0.017	−0.012	49	7	o
					1-Nonanol(I)					
14					4.025	0.514	3.409	5	3	e
14[b]					0.376	0.047	−0.295	60	−56	(o)
59	323.15	373.15	1.0	10.0	0.139	0.017	0.005	49	3	o
					1-Nonanol(II)					
14	288.85	431.15	5.1	50.6	1.415	0.179	0.658	69	43	e
14[b]					0.967	0.121	−0.054	434	−32	(o)
59	323.15	373.15	1.0	10.0	0.154	0.019	0.012	49	9	o
					1-Decanol					
6					4.843	0.577	−4.580	16	−16	o
47	298.15	348.15	1.0	40.2	0.119	0.015	0.054	27	9	o
48	283.15	623.15	5.0	78.8	2.914	0.394	0.343	95	5	(o)

[a] o, (o), from the same source as the compressed liquid density data, available for a part of the temperature range only, respectively; e, from the smoothing equations given in ref 58. [b] Smoothed data. [c] Densities calculated from ultrasound velocities.

temperature and pressure from the relation

$$s(\varrho) = s_w[\sum_{k=1}^{N}\sum_{m=1}^{N}(\partial f/\partial a_k)(\partial f/\partial a_m)c_{km}]^{1/2} \qquad \text{(A-4)}$$

where

$$\vec{a} = \{a_1, ..., a_N\} = \{c_0, ..., c_{N_C}, b_0, ..., b_{N_B}\} \qquad \text{(A-5)}$$

$$N = N_C + N_B + 2$$

$f = \varrho(T,P,\vec{a})$, and c_{km} are the elements of the matrix inverse to the matrix of the set of linear equations of the last iteration within the Marquardt procedure. The matrix was stored for each final fit in the database of the results.

The other statistical characteristics are defined as follows:

$$\mathrm{RMSD} = \{\sum_{j=1}^{N_P}[\varrho_j - \varrho(T_j,P_j,\vec{c},\vec{b})]^2/N_P\}^{1/2} \qquad \text{(A-6)}$$

$$\mathrm{RMSD}_r/\% = 100\{\sum_{j=1}^{N_P}[1 - \varrho(T_j,P_j,\vec{c},\vec{b})/\varrho_j]^2/N_P\}^{1/2} \qquad \text{(A-7)}$$

$$\mathrm{bias} = \sum_{j=1}^{N_P}[\varrho_j - \varrho(T_j,P_j,\vec{c},\vec{b})]/N_P \qquad \text{(A-8)}$$

$$\pm = \sum_{j=1}^{N_P}\mathrm{sign}[\varrho_j - \varrho(T_j,P_j,\vec{c},\vec{b})]\cdot 1 \qquad \text{(A-9)}$$

where N_P is either the overall number of experimental data points retained for the correlation (Table 2) or the number of either retained (retained data subsets) or rejected (rejected data subsets) experimental data points in a data subset taken from the particular literature reference (Table 3).

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