PVT Properties of Liquid *n*-Alkane Mixtures

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Four liquid-phase isotherms are measured for two binary and one ternary mixture made from four straight-chain hydrocarbons. The accuracy is estimated at ±0.06% in the volume. A brief description of the modified Bridgman-type sylphon-bellows piezometer used is given, followed by the results of the four isotherms between 25.00° and 85.00°C for equimolar binary mixtures of *n*-decane with *n*-tetradecane and *n*-dodecane with *n*-hexadecane. The ternary mixture is 0.6000 mole fraction *n*-decane with 0.2000 mole fraction *n*-tetradecane and 0.2000 mole fraction *n*-hexadecane. Excess volumes are calculated for the three mixtures at five representative pressures.

Liquid PVT properties of the three mixtures were determined by use of a modified Bridgman-type (*5*) sylphon-bellows piezometer. The data taken in this study represent the first high-accuracy mixture PVT data taken in which the pressure range extends from atmospheric pressure to just below the freezing pressure on all isotherms for liquid mixtures. The unsmoothed PVT data are tabulated as a function of temperature and pressure for the three mixtures. Pure component data over the same ranges of temperature and pressure have been reported elsewhere (*24*). However, the molecular weight of *n*-tetradecane was incorrectly taken as 196.37 g/g-mol. Thus, molar volume values must be multiplied by 198.40/196.37 to obtain the correct molar volumes for *n*-tetradecane.

Experimental

Apparatus. The operation of the PVT cell can be described with the aid of Figure 1. The two main components of the PVT cell are the bellows, marked B on the drawing, and the slide wire. S. The slide wire is a section of Karma wire approximately 1 in. in length and 0.010 in. in diameter. Karma is a trademark of the Driver-Harris Co., Harrison, N.J., and is an alloy of nickel, chromium, and aluminum. Karma wire was used as the slide wire because of its low-temperature coefficient of resistivity (0 ± 10 ppm/°C), its precision drawn diameter, and its highly uniform linear conductivity (experimentally determined as 3.87800 ± 0.00002 cm/ohm for the 0.010-in. diameter Karma wire) (*23*). There are three electrical contacts. labeled E_1, E_2, and E_3 on Figure 1, made to the Karma slide wire. The slide wire is held against the edge of contact C by a Teflon plunger-spring arrangement. Contact C is a piece of 0.010-in. diameter Karma wire soldered to a brass plate. The retainer. R, supports the bellows and the associated electrical components. The bellows is held in the retainer by three screws. marked P on the drawing.

As the bellows and sample compress longitudinally under hydrostatic load, the Karma slide wire is drawn past the fixed contact C of Figure 1. The change in length of the bellows is calculated from the change in electrical resistance across E_1, E_2, and E_3 after thermal equilibrium is attained.

A Leeds and Northrup Model G-2 Mueller bridge and a Model 2284d galvanometer and scale were used in conjunction with a four-position mercury contact commutator to make the necessary resistance measurements. The bridge had been calibrated by Leeds and Northrup using National Bureau of Standards calibrated reference resistors, and a set of correctional constants was provided with the bridge. The measuring circuit was wired to eliminate lead wire and contact resistance (*23*).

The pressure-generating system consisted of three hydraulic handjacks and a piston intensifier. The system is capable of pressures to 200,000 psi with a maximum temperature limit on the PVT cell of 150°C. Two Heise gages, one 0–1500-psi gage and one 0–50,000-psi gage, were used for pressure measurements below 50,000 psi. These were temperature compensated and accurate to 0.1% of full scale. The two Heise gages were calibrated by the Heise Bourdon Tube Co. using a National Bureau of Standards approved dead weight tester. A report of the calibration was supplied with each gage. In addition. the calibration of the two gages was rechecked in this laboratory with a Ruska dead weight tester for the 0–1500-psi gage and an Aminco 100,000-psi dead weight tester for the 0–50,000-psi gage. For pressures above 50.000 psi, a Manganin cell pressure transducer was used in conjunction with the Mueller G-2 bridge. The Manganin cell, calibrated at 25.00°C with the Aminco 100,000-psi dead weight tester, was maintained at 25.00 ± 0.01°C during each isotherm by an Aminco constant-temperature bath.

A Hallikainen constant-temperature bath and Hallikainen Thermitrol controller were used to control the temperature of the PVT cell. A Sola constant voltage transformer was used as the Thermitrol's power supply. The temperature of the bath was measured to ±0.01°C with a platinum resistance thermometer previously calibrated by the National Bureau of Standards on the 1948 International Practical Temperature Scale. All isotherms were run at the set point temperature with measured variations of ±0.003°C about the set point.

Data reduction. The change in volume of the sample in the bellows relative to the volume at atmospheric pressure as pressure is applied to the system is a function of the following: The vacuum corrected weight of the sample in the bellows, W_{VC}, the atmospheric pressure density

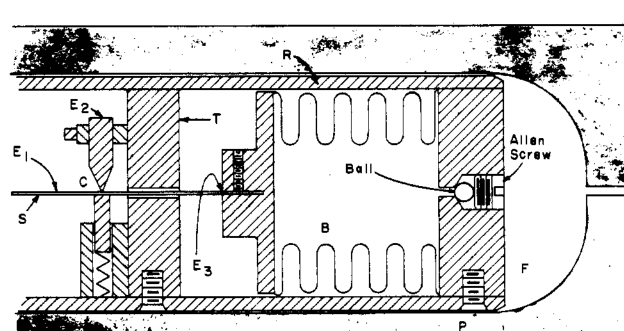

Figure 1. Detail of bellows-slide wire arrangement

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of the sample, $\rho_{o,T}$, the temperature and pressure corrected cross-sectional area of the bellows, $A_{P,T}$, and the change in length of the bellows with pressure, ΔL_B. Appropriate temperature and pressure corrections were applied to ΔL_B and $A_{P,T}$ to obtain the true compression of the sample as represented by Equation 1

$$(v_o - v_i)/v_o = (\Delta L_B \cdot A_{P,T} \cdot \rho_{o,T})/W_{vc} \qquad (1)$$

The quantity $(v_o - v_i)/v_o$ is the compression of the sample where v_i is the specific volume at temperature T and pressure P_i, and v_o is the specific volume at temperature T and atmospheric pressure P_o.

With the exception of the atmospheric pressure density, $\rho_{o,T}$, all terms on the right-hand side of Equation 1 were obtained during the course of this study. The atmospheric pressure density, $\rho_{o,T}$, for the mixtures were determined from pycnometric measurements (*12, 15, 22*). For the binary system at temperatures from 25° to 65°C, inclusively, and the ternary system at 25° and 45°C, the experimental excess volume at each temperature for each exact mole fraction was estimated from a curve fit to the data. This value was then used in combination with the pure component densities reported (*12, 15, 22*) to determine the mixture density. For isotherms at other temperatures, pure component densities and mixture excess volume were obtained by linear temperature interpolation or extrapolation of experimental results for each system. The mixture densities were then calculated as above. The estimated maximum error introduced was 0.0001 g/cm³ in the density at 85°C.

An error analysis technique used by the National Bureau of Standards and detailed by Mickley et al. (*18*), indicates that the errors in the PVT measurements made with this system are no greater than 0.0006 cm³/cm³.

A direct comparison of these results with those previously reported for some of the pure components (*4, 6, 8, 9*) was not possible since the investigations were not conducted at the same temperatures. However, Snyder and Winnick (*24*) have fitted the isothermal compressibilities of Boelhouwer (*4*), Bridgman (*6*), and Cutler et al. (*8, 9*) with temperature and then interpolated to the temperature of the present study. The relative volume of decane, dodecane, and hexadecane then calculated as a function of pressure agrees with our results within the precision allowed by the temperature interpolation, generally to ±0.2% at the highest pressure.

Materials. Decane, dodecane, tetradecane, and hexadecane were obtained from Humphrey Chemical Co., North Haven, Conn. They were manufactured from naturally occurring, even-numbered, straight-chain fatty alcohols. Their purity was at least 99%, as determined from a chromatographic analysis. The most probable impurities would be the adjacent even-numbered *n*-alkanes. The sample was degassed before the bellows was filled. These were the same materials as used in the atmospheric pressure density determinations (*12, 15, 22*).

Results and Discussion

Experimental data. The relative volumes for each of the three mixtures studied as a function of pressure at 25.00°, 45.00°, 65.00°, and 85.00°C are presented in Table I. The relative volume, $\bar{v}_i$, is defined as 1.0000 −

Table I. Mole Fraction Mixtures

25.00°C		45.00°C		65.00°C		85.00°C	
Press, atm	Rel vol, cc/cc	Press, atm	Rel vol, cc/cc	Press, atm	Rel vol, cc/cc	Press, atm	Rel vol, cc/cc
0.5000 Mole fraction mixture of *n*-decane *n*-tetradecane							
$\rho_{o,T} = 0.74488$ g/cm³		$\rho_{o,T} = 0.73052$ g/cm³		$\rho_{o,T} = 0.71560$ g/cm³		$\rho_{o,T} = 0.70067$ g/cm³	
1.0	1.0000	1.0	1.0000	1.0	1.0000	1.0	1.0000
17.3	0.9992	17.0	0.9984	17.9	0.9980	9.9	0.9991
70.5	0.9949	33.3	0.9962	32.6	0.9960	15.4	0.9985
99.1	0.9920	68.1	0.9926	70.2	0.9916	18.1	0.9977
137.1	0.9894	98.4	0.9892	101.4	0.9879	20.9	0.9976
176.1	0.9858	137.8	0.9854	135.1	0.9849	26.9	0.9963
208.6	0.9838	210.6	0.9792	214.0	0.9764	35.4	0.9954
239.5	0.9808	304.5	0.9718	518.8	0.9510	42.0	0.9940
273.9	0.9790	411.3	0.9630	614.1	0.9441	48.8	0.9930
312.7	0.9752	504.5	0.9568	722.3	0.9373	55.7	0.9922
377.3	0.9712	612.7	0.9492	823.0	0.9306	63.9	0.9907
514.1	0.9623	723.7	0.9431	927.8	0.9255	70.9	0.9903
585.5	0.9572	823.0	0.9370	1026.5	0.9198	71.0	0.9900
646.1	0.9546	932.6	0.9316	1131.2	0.9151	86.3	0.9876
717.5	0.9497	1018.3	0.9269	1234.7	0.9092	101.4	0.9857
786.9	0.9467	1130.6	0.9219	1500.7	0.8976	137.1	0.9822
852.3	0.9427	1230.6	0.9169	1720.5	0.8901	208.5	0.9739
921.0	0.9398	1431.3	0.9090	1901.5	0.8830	307.9	0.9638
991.1	0.9357	1632.7	0.9009	2322.7	0.8692	417.4	0.9528
1058.4	0.9331	1838.2	0.8942	2725.6	0.8585	613.4	0.9379
1131.2	0.9294	2047.8	0.8862	2927.0	0.8539	728.4	0.9303
1183.0	0.9271			3131.1	0.8481	824.4	0.9235
1257.8	0.9241					926.4	0.9175
						1031.9	0.9114
						1226.5	0.9014
						1373.5	0.8953
						1510.3	0.8882
						1635.5	0.8842
						1770.9	0.8781
						1990.0	0.8714

Table I. Continued

25.00°C		45.00°C		65.00°C		85.00°C	
Press, atm	Rel vol, cc/cc	Press, atm	Rel vol, cc/cc	Press, atm	Rel vol, cc/cc	Press, atm	Rel vol, cc/cc
						$\rho_{o,T}$ = 0.70067 g/cm³	
						2178.5	0.8642
						2388.0	0.8589
						2795.6	0.8477
						3195.1	0.8377
						3597.5	0.8288
						3809.9	0.8236
						4000.3	0.8199
						4244.8	0.8156
0.5000 mole fraction mixture of *n*-dodecane and *n*-hexadecane							
$\rho_{o,T}$ = 0.75891 g/cm³		$\rho_{o,T}$ = 0.74490 g/cm³		$\rho_{o,T}$ = 0.73062 g/cm³		$\rho_{o,T}$ = 0.71628 g/cm³	
1.0	1.0000	1.0	1.0000	1.0	1.0000	1.0	1.0000
17.6	0.9984	18.1	0.9977	17.2	0.9985	36.4	0.9953
72.6	0.9934	85.9	0.9912	35.0	0.9961	97.6	0.9879
84.7	0.9926	98.7	0.9901	68.7	0.9925	137.8	0.9835
135.7	0.9885	135.1	0.9864	97.1	0.9896	205.8	0.9763
175.9	0.9851	275.2	0.9756	137.1	0.9858	403.8	0.9579
205.1	0.9831	339.2	0.9707	208.5	0.9786	517.5	0.9489
239.2	0.9808	410.0	0.9652	307.9	0.9700	616.8	0.9414
271.1	0.9783	484.1	0.9596	409.3	0.9617	718.9	0.9347
308.6	0.9757	548.8	0.9561	515.4	0.9541	825.0	0.9281
344.6	0.9732	618.9	0.9518	617.5	0.9470	929.1	0.9222
380.0	0.9707	684.9	0.9476	812.1	0.9354	1018.3	0.9173
411.3	0.9684	754.3	0.9439	924.4	0.9297	1134.0	0.9118
446.7	0.9664	822.3	0.9400	1018.3	0.9248	1227.9	0.9069
480.7	0.9641	889.0	0.9367	1130.6	0.9188	1369.4	0.9008
512.0	0.9625	958.4	0.9332	1221.7	0.9146	1497.3	0.8952
550.1	0.9602	1019.6	0.9302	1368.7	0.9083	1646.4	0.8891
		1082.9	0.9273	1498.0	0.9032	1770.2	0.8844
		1159.8	0.9240	1640.2	0.8979	1913.8	0.8796
		1225.8	0.9212	1753.2	0.8937	2045.1	0.8751
				1971.6	0.8862	2249.2	0.8686
				2168.9	0.8798	2443.8	0.8628
						2664.3	0.8571
						3076.7	0.8464
						3269.7	0.8421
Mixture of 0.6000 mole fraction *n*-decane and 0.2000 mole fraction each *n*-tetradecane and *n*-hexadecane							
$\rho_{o,T}$ = 0.74503 g/cm³		$\rho_{o,T}$ = 0.73048 g/cm³		$\rho_{o,T}$ = 0.71571 g/cm³		$\rho_{o,T}$ = 0.70085 g/cm³	
1.0	1.0000	1.0	1.0000	1.0	1.0000	1.0	1.0000
16.7	0.9982	18.6	0.9981	17.8	0.9974	17.1	0.9976
35.0	0.9968	97.5	0.9893	31.6	0.9957	40.4	0.9944
68.9	0.9931	137.8	0.9860	68.2	0.9912	67.8	0.9906
100.3	0.9910	210.6	0.9793	201.1	0.9767	201.7	0.9745
137.1	0.9876	308.6	0.9704	308.6	0.9672	318.8	0.9622
206.5	0.9819	518.2	0.9547	411.3	0.9588	410.6	0.9537
275.9	0.9762	618.2	0.9487	497.7	0.9513	517.5	0.9447
352.1	0.9711	723.7	0.9419	616.1	0.9432	624.3	0.9367
410.6	0.9662	817.6	0.9369	717.5	0.9363	720.9	0.9296
472.6	0.9631	931.9	0.9305	926.4	0.9242	824.4	0.9231
550.8	0.9573	1021.7	0.9263	1021.7	0.9193	1021.7	0.9117
616.8	0.9538	1130.6	0.9206	1130.6	0.9139	1129.2	0.9058
688.3	0.9494	1225.8	0.9169	1223.1	0.9093	1232.6	0.9009
752.2	0.9463	1499.4	0.9052	1372.8	0.9027	1368.7	0.8944
820.3	0.9420	1636.8	0.8998	1497.3	0.8974	1502.8	0.8885
953.6	0.9356	1779.7	0.8955	1645.0	0.8913	1644.3	0.8827
1025.8	0.9330	1899.5	0.8912	1763.4	0.8873	1771.9	0.8778
1076.1	0.9302	2041.7	0.8872	1919.9	0.8817	1991.3	0.8700
1154.4	0.9266			2039.7	0.8777	2178.5	0.8637
				2265.6	0.8708	2584.0	0.8521
				2450.7	0.8659	2794.3	0.8463
				2663.0	0.8595	2939.6	0.8415
				2881.4	0.8544	3186.9	0.8366
				2981.4	0.8515	3386.3	0.8321
				3126.3	0.8484	3543.7	0.8286

$(v_o - v_i)/v_o$. The compression $(v_o - v_i)/v_o$ is defined by Equation 1. No smoothing of the data has been done in Table I.

Representation of experimental data by Tait equation. An empirical equation usually known as the "Tait" equation has been used to represent the PVT behavior for a number of liquids (*2, 3, 8–11, 14, 17, 19–21, 23–25*). The "usual" Tait equation has been commonly written as

$$v_i = v_o - J \ln [(P_i + L)/(P_o + L)] \quad (2)$$

where $v_o = v_o(P_o)$, a reference volume, the parameters J and L are taken pressure-independent. Macdonald (*16*) suggested that Equation 2 is better written in the form involving two physical quantities, K_o and K_o', where K_o is the bulk modulus at reference pressure, $K_o = K|_{P=P(o)}$, and $K_o' = (\partial K/\partial P)_T|_{P=P(o)}$. This form of the UTE may be written in terms of the relative volume as

$$\bar{v}_i = 1 - 1/(K_o' + 1) \ln [1 + (K_o' + 1)(P_i - P_o)/K_o] \quad (3)$$

where $\bar{v}_i$ is the relative volume at pressure P_i.

The two parameters, K_o and K_o', were evaluated from the experimental PVT data on each of the mixtures for each of the isotherms by a new generalized least-squares regression technique (*1, 7*). The results are shown in Table II. The pure component data tabulated in ref. *24* were reevaluated by this new technique. The best-fit values for K_o and K_o' are also reported in Table II. The worst deviation of any data point from the smooth curve was 0.0011 cm^3/cm^3.

The curves shown in Figure 2 are the results by using Equation 3 with the parameters as given in Table II. The raw experimental data for the 0.5000 mole fraction *n*-decane and *n*-tetradecane mixture are also shown. In order to see the difference in the fit of Equation 3 to the experimental mixture PVT data, it was necessary to plot differences in compression as a function of pressure at constant temperature. Figure 3 presents such a representative plot for the 0.5000 mole fraction *n*-dodecane and *n*-hexadecane mixture at 85.00°C.

The original Tait equation (*13*), $(v_o - v)/v_o = AP/(B + P)$, and the equation referred to by Macdonald as 3BE (*16*), a series expansion of $\bar{v}_i$ in terms of $(P - P_o)$ up to third degree, were also tested. However, neither the pure component nor the mixture data were represented as well as with Equation 3, even though only two parameters are required by Equation 3.

Experimental excess volumes. The excess volume at any temperature, pressure, and composition is defined as

$$V^E(T, P, x) = V_m(T, P, x) - \Sigma x_i V_i(T, P) \quad (4)$$

where V_m is the molar volume of the mixture, and V_i and x_i are the molar volume and mole fraction of the ith component, respectively. The molar volumes of the three mixtures and the four pure components are calculated by

Table II. Compression Parameters K_o and K_o'

System		Temp, °C 25.00	45.00	65.00	85.00
C_{10}	K_o =	9161 ± 23	7825 ± 31	6778 ± 27	6002 ± 24
	K_o' =	10.4 ± 0.05	10.5 ± 0.06	10.4 ± 0.05	10.3 ± 0.04
C_{12}		10701 ± 100	8885 ± 26	8008 ± 58	6595 ± 25
		9.3 ± 0.2	10.4 ± 0.06	10.1 ± 0.1	10.5 ± 0.05
C_{14}		11114 ± 235	9751 ± 84	8632 ± 22	7512 ± 22
		9.0 ± 1.1	9.9 ± 0.6	10.0 ± 0.04	10.1 ± 0.04
C_{16}		(8735 ± 426)[a]	10264 ± 176	9215 ± 65	8058 ± 65
		(31.0 ± 6.7)[a]	10.9 ± 0.6	10.0 ± 0.1	10.1 ± 0.1
$C_{10} + C_{14}$		11604 ± 164	8932 ± 75	7908 ± 38	6703 ± 42
		7.6 ± 0.4	10.3 ± 0.2	10.1 ± 0.07	10.5 ± 0.07
$C_{12} + C_{16}$		10781 ± 88	9140 ± 87	8619 ± 40	7441 ± 15
		11.0 ± 0.5	10.8 ± 0.3	10.1 ± 0.08	10.3 ± 0.03
$C_{10} + C_{14} +$ C_{16}		10115 ± 105	8718 ± 54	7640 ± 42	6722 ± 18
		10.3 ± 0.3	10.6 ± 0.1	10.5 ± 0.08	10.3 ± 0.03

[a] Pressure range of data insufficient to accurately determine parameters.

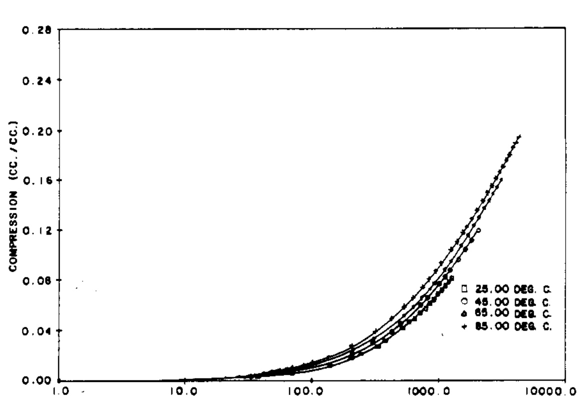

Figure 2. Compression results for decane–tetradecane equimolar mixture curves are calculated from Equation 3 by using parameters listed in Table II

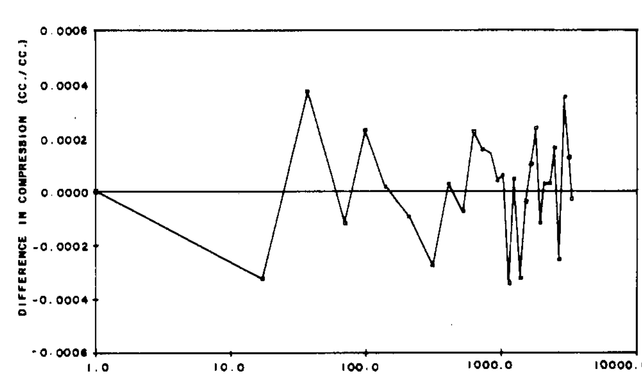

Figure 3. Difference between calculated (Equation 3) and experimental compression for dodecane–hexadecane equimolar mixture at 85°C

Table III. Experimental Excess Volumes

	25°C		45°C
P, atm	V^E, cc/mol	P, atm	V^E, cc/mol
	$C_{10} + C_{14}$ ($x_1 = 0.5000$)		
1	−0.04 ± 0.04	1	−0.10 ± 0.04
100	0.16 ± 0.04	400	0.00 ± 0.05
300	0.42 ± 0.05	800	0.07 ± 0.06
500	0.54 ± 0.05	1200	0.13 ± 0.08
600	0.58 ± 0.06	1600	0.18 ± 0.09
	65°C		85°C
1	−0.09 ± 0.04	1	−0.09 ± 0.04
700	0.10 ± 0.06	1000	−0.07 ± 0.07
1300	0.12 ± 0.09	2000	0.08 ± 0.09
1900	0.12 ± 0.09	3000	0.21 ± 0.11
2400	0.11 ± 0.11	3600	0.28 ± 0.12
	$C_{12} + C_{16}$ ($x_1 = 0.5000$)		
	45°C		65°C
1	−0.03 ± 0.04	1	−0.06 ± 0.04
200	−0.12 ± 0.04	500	−0.09 ± 0.05
500	−0.18 ± 0.05	1000	−0.11 ± 0.07
800	−0.21 ± 0.06	1500	−0.12 ± 0.09
1000	−0.21 ± 0.07	2000	−0.13 ± 0.09
	85°C		
1	−0.06 ± 0.04		
800	0.10 ± 0.06		
1600	0.14 ± 0.09		
2400	0.16 ± 0.10		
2900	0.17 ± 0.11		
	$C_{10} + C_{14} + C_{16}$ ($x_1 = 0.6000$, $x_2 = 0.2000$)		
	45°C		65°C
1	−0.11 ± 0.04	1	−0.16 ± 0.04
200	−0.13 ± 0.04	500	−0.18 ± 0.05
500	−0.14 ± 0.05	1000	−0.15 ± 0.07
800	−0.13 ± 0.06	1500	−0.11 ± 0.09
1000	−0.13 ± 0.07	2000	−0.07 ± 0.09
	85°C		
1	−0.20 ± 0.04		
800	−0.24 ± 0.06		
1600	−0.22 ± 0.09		
2400	−0.19 ± 0.10		
2900	−0.17 ± 0.11		

using Equation 3 with the parameters as given in Table II, the standard molecular weights, and the densities at pressure P_o (1 atm).

The experimental excess volumes of the two binary mixtures and one ternary mixture at five characteristic pressures are presented in Table III. The experimental uncertainties in the calculated excess volumes are also reported.

Acknowledgment

Most of the computing was done with the help of the University of Missouri Research Center and the Engineering Computer Center.

Nomenclature

$A_{P,T}$ = cross-sectional area of the sylphon-bellows, cm^2
J = coefficient in Equation 2, cm^3/g
K = isothermal bulk modulus, atm
K_o = isothermal bulk modulus at $P = P_o$, atm
K_o' = $(\partial K/\partial P)_T|_{P=P(o)}$, dimensionless
L = coefficient in Equation 2, atm
ΔL_B = change in length of the sylphon-bellows as a function of P and T, cm
P_o = atmospheric pressure, atm
P_i = pressure, atm
t = temperature, °C
T = temperature, K
v_o = atmospheric pressure specific volume, cm^3/g
v_i = specific volume at ith pressure, cm^3/g
$\bar{v}_i$ = relative volume at ith pressure, dimensionless
V_i = molar volume of ith component, cc/mol
V_m = molar volume of mixture, cc/mol
V^E = excess volume, cc/mol
x = mole fraction
W_{vc} = vacuum corrected weight of sample in bellows, grams
$\rho_{o,T}$ = atmospheric pressure density at temperature T, g/cm^3

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