ature, the complete system can be described up to the critical point.

Figure 1 illustrates the $P-T$ relations for the system. The solid line for n-pentane was drawn by using literature data while the points on that line correspond to experimental check data on that component.

Figure 2 illustrates the $P-T$ projections of the bubble points of the three mixtures calculated with the Peng–Robinson (2) equation and experimental measurements from this study. As can be seen, the Peng–Robinson equation is able to predict bubble points of this system satisfactorily. No binary interaction coefficients were used in this study.

Literature Cited

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Viscosities and Densities of Binary Mixtures of 1-Alkanols from 15 to 55 °C

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Viscosities of binary mixtures of 1-decanol, 1-nonanol, 1-octanol, and 1-heptanol with 1-butanol, 1-propanol, ethanol, and methanol were measured at 15, 25, 35, 45, and 55 °C. Viscosities η are presented as a polynomial in mole fraction X_1 of the low molecular weight component of binary mixture: $\eta = A + BX_1 + CX_1^2 + DX_1^3 + EX_1^4$. Molar volumes of binary mixtures are given by the sums of the products of molar volume and mole fraction of components involved. Viscous heats and preexponential factors are also reported. These data were also fitted to the empirical equation of McAllister, Herric, Auslander, and two additional polynomials.

Introduction

The literature dealing with the viscosity of mixtures of 1-alkanols is rather limited. While a number of empirical relations (1–6) have been proposed to predict the viscosity of a mixture from that of pure components, we did not trust the extrapolation to higher alkanols. In the present work, we have measured the viscosity of 16 binary mixtures comprising 1-decanol, 1-nonanol, 1-octanol, 1-heptanol, 1-butanol, 1-propanol, ethanol, and methanol. These data were fitted to a five-parameter polynomial with a goodness of fit approximating experimental error. In addition, these binary data were fitted to the empirical relations of McAllister (1), Herric (2), and Auslander (3) and also to two additional polynomial equations using a multiparametric curve-fitting program. In order to obtain viscosity from kinematic viscosity, densities of all mixtures were also determined. The viscous heat and the preexponential factor of the mixtures were also calculated.

Models and Equations

Several equations have been put forward to calculate the viscosities of binary mixtures.

McAllister's Formula (1). McAllister derived the following relation for a liquid mixture on the basis of the absolute reaction rate theory of Eyring:

$$\ln \eta = X_1^3 \ln \eta_1 + 3X_1^2 X_2 \ln \eta_{12} + 3X_1 X_2^2 \ln \eta_{21} + X_2^3 \ln \eta_2 + D' \quad (1)$$

where

$$D' = -\ln (X_1 + X_2 M_2/M_1) + 3X_1^2 X_2 \ln (\tfrac{2}{3} + M_2/3M_1) + 3X_1 X_2^2 \ln (\tfrac{1}{3} + 2M_2/3M_1) + X_2^3 \ln (M_2/M_1)$$

The equation contains two adjustable parameters, ln η_{12} and ln η_{21}.

Herric's Formula (2). The following relation was proposed by Herric to correlate viscosities of binary mixture:

$$\ln \eta = X_1 \ln \eta_1 + X_2 \ln \eta_2 + X_1 \ln M_1 + X_2 \ln M_2 - \ln (X_1 M_1 + X_2 M_2) + \Delta 12 \quad (2)$$

where $\Delta 12 = \alpha_{12} X_1 X_2$ is a function representing molecular interaction and $\alpha_{12} = \alpha_{21}$ is the interaction parameter. Herric expressed α_{12} or α_{21} as a linear function of composition

$$\alpha_{12} = \beta_{12} + \beta_{21}(X - X_2) \quad (3)$$

The coefficients β_{12} and β_{21} may be determined from a least-squares method.

Auslander's Formula (3). Auslander's equation has the following three-parameter form:

$$X_1(X_1 + B_{12} X_2)(\eta - \eta_1) + A_{21} X_2(B_{21} X_1 + X_2)(\eta - \eta_2) = 0 \quad (4)$$

Here, A_{21}, B_{12}, and B_{21} are the parameters representing binary interactions.

Polynomial Functions. Viscosity data were fitted to three different polynomials of the following form, the first of which was chosen as the form of data presentation in this paper; note that polynomial I is the only equation which does not require fixed values of the viscosities of the pure components and allow them to share in the goodness of fit:

polynomial I

$$\eta = A + BX_1 + CX_1^2 + DX_1^3 + EX_1^4 \quad (5)$$

where A, B, C, D, and E are adjustable parameters.

polynomial II

$$\eta = \eta_1 X_1 + \eta_2 X_2 + AX_1 + BX_2 + X_1 X_2 A + B(X_1 - X_2) + C(X_1 - X_2)^2 \quad (6)$$

polynomial III

$$\eta = \eta_1 X_1 + \eta_2 X_2 + X_1 X_2 A + B(X_1 - X_2) + C(X_1 - X_2)^2 \quad (7)$$

Table I. Densities (g/mL) of Different 1-Alkanols

	15.0 °C		25.0 °C		35.0 °C		45.0 °C,	55.0 °C
	obsd	lit.	obsd	lit.	obsd	lit.	obsd	obsd
C_1	0.7959	0.7960[e]	0.7861	0.7865[a] 0.7864[b] 0.7866[c]	0.7755		0.7693	0.7586
C_2	0.7934	0.7933[e]	0.7845	0.7850[c]	0.7767	0.7764[e]	0.7681	0.7599
C_3	0.8077	0.8075[e]	0.7994	0.8023[d] 0.7993[e]	0.7920		0.7841	0.7758
C_4	0.8132	0.8133[e]	0.8056	0.8085[d] 0.8057[a]	0.7981		0.7907	0.7829
C_7	0.8257		0.8190		0.8112		0.8041	0.7871
C_8	0.8276	0.8320[e]	0.8211		0.8142		0.8078	0.8010
C_9	0.8310		0.8244		0.8177		0.8111	0.8043
C_{10}	0.8327		0.8254		0.8197		0.8127	0.8064

[a] Reference 8. [b] Reference 9. [c] Reference 10. [d] Reference 11. [e] Reference 12.

Table II. Viscosities (cP) of 1-Alkanols

	15.0 °C		25.0 °C		35.0 °C		45.0 °C		55.0 °C,
	obsd	lit.	obsd	lit.	obsd	lit.	obsd	lit.	obsd
C_1	0.6305		0.547	0.546[a] 0.545[b,c]	0.4745		0.4194		0.3722
C_2	1.280		1.056	1.078[d] 1.068[e]	0.8818	0.8683[e]	0.7461	0.7292[e]	0.6302
C_3	2.3179	2.522[d]	1.898	1.966[f] 2.577[h]	1.436		1.223		0.9908
C_4	3.159	3.379[e]	2.524	2.564[f] 2.463[g]	1.969	2.00[i]	1.558		1.246
C_7	8.302		5.774		4.263		3.156		2.364
C_8	10.662	10.640[e]	7.363	7.21[d]	5.256		3.868		2.890
C_9[j]	13.146		9.101		6.522		4.677		3.476
C_{10}[j]	16.465		10.974		7.509		5.358		3.957

[a] Reference 13. [b] Reference 14. [c] Reference 15. [d] Reference 16. [e] Reference 12. [f] Reference 11. [g] Reference 17.
[h] Reference 18. [i] Reference 19. [j] The interpolated values of viscosities at 11 and 32 °C from the present data are in good agreement with ref 20.

Table III. Averages of Rms for All Mixtures at All Temperatures of 1-Alkanol

	McAllister[a]	Auslander	Herric[a]	polynomial		
				I	II	III
C_{10} mixtures	0.0258	0.0410	0.0378	0.0417	0.2442	0.0576
C_7 mixtures	0.0200	0.0118	0.0138	0.0130	0.1161	0.0122

[a] Rms in logarithm form.

Here again, A, B, and C are adjustable parameters and could be determined by a weighted least-squares method.

The viscous heat and the preexponential factor of the mixtures were also calculated from the equation

$$\eta = \eta_0 \exp(E_v/RT) \qquad (8)$$

where η is viscosity, η_0 is the preexponential factor, E_v is viscous heat, R is the gas constant, and T is the absolute temperature.

Experimental Section

All 1-alkanols used in this work were of analytical grade (Fluka AG, Buchs SG) and had a purity of 99.9%. Mixtures of methanol, ethanol, 1-propanol, and 1-butanol with 1-heptanol, 1-octanol, 1-nonanol, and 1-decanol were prepared in all compositions used by weighing an appropriate volume of each solvent. Density and viscosity measurements were carried out at five different temperatures, i.e., 15.0–55.0 °C with a regular interval of 10 °C.

Density measurements were carried out by using a densitometer which was calibrated with mercury. The densitometer was a specific gravity bottle ($\sim$ 10 mL) which had a graduated capillary neck. Viscosities of mixtures were determined by KPG-Ubbelhode viscometers, Schott-Gerate Nos. 2450113, 2452610, and 2452613. Time of flow was noted by using the

Model AVS viscosity measuring instrument of Schott-Gerate, which uses a modulated light transmitter/receiver. Efflux times are noted by a digital counter of 0.01-s resolution.

A temperature stability of ±0.1 °C was recorded by using a platinum resistance thermometer. Computation of the kinematic viscosity is done by using the relation

$$\alpha = kt$$

where α = kinematic viscosity of a sample in cSt, k = capillary constant of the viscometer in cSt/s, and, t = time of flow in seconds.

The viscometer was calibrated with water at all working temperatures and the mean value of k was used in calculations.

The kinematic viscosities were adjusted for kinetic energy by using the Hagenbach correction (7). Absolute viscosity values were then determined by using the relation

$$\eta = \alpha\rho$$

where η = absolute viscosity of a sample in cP and ρ = density of a sample in g/mL.

Results and Discussion

Densities were determined for pure 1-alkanols C_1, C_2, C_3, C_4, C_7, C_8, C_9, and C_{10} at 15, 25, 35, 45, and 55 °C. The values are listed in Table I along with literature values and there is

Table IV.   Values of Various Parameters for the Equation $\eta = A + BX_1 + CX_1^2 + DX_1^3 + EX_1^4$

temp, °C	A	B	C	D	E	rms error
			Methanol–Decanol			
15.0	16.487	−19.236	−16.217	32.563	−12.990	0.098 41
25.0	10.980	−11.592	−8.454	14.123	−4.506 6	0.040 11
35.0	7.509 3	−7.267 8	−3.647 7	4.390 7	−0.500 8	0.037 20
45.0	5.358 6	−4.303 0	−3.214 0	2.498 1	0.086 15	0.030 35
55.0	3.972 9	2.772 4	−28.990	41.400	−19.002	0.090 52
			Ethanol–Decanol			
15.0	16.466 0	−18.337 8	−9.422 5	19.874 1	−7.312 9	0.022 571 1
25.0	10.974 8	−10.879 5	−5.451 22	8.967 3	−2.557 45	0.010 266 9
35.0	7.512 60	−3.271 9	−21.275 0	31.917 0	−14.023 9	0.058 139
45.0	5.356 76	−11.294 4	23.619 3	−29.004 1	12.135 0	0.052 669
55.0	3.959 76	−3.156 47	−3.347 79	6.794 59	−3.621 97	0.040 676
			Propanol–Decanol			
15.0	16.466 6	−13.713 8	−12.577 8	19.426 3	−7.269 92	0.022 20
25.0	10.975 3	−8.852 44	−4.571 48	5.456 26	−1.111 85	0.008 225
35.0	7.510 60	−6.248 76	3.247 05	−4.249 88	2.165 10	0.052 86
45.0	5.355 82	−4.322 80	0.206 155	0.905 682	0.910 139	0.028 29
55.0	3.954 61	−2.885 33	−0.425 44	0.087 336	0.276 58	0.023 85
			Butanol–Decanol			
15.0	16.465 1	−13.571 9	−11.071 8	19.102 3	−7.747 6	0.022 732
25.0	10.974 4	−6.269 33	−17.152 2	26.269 7	−11.336 5	0.076 374
35.0	7.514 70	−7.722 69	6.320 51	−8.304 82	4.167 57	0.799 77
45.0	5.360 10	−4.495 91	1.422 37	−2.393 71	1.650 94	0.024 030
55.0	3.958 59	−3.218 39	1.153 52	−1.857 40	1.193 89	0.017 058
			Methanol–Nonanol			
15.0	13.158 3	−16.689 1	−6.424 56	17.718 2	−7.144 16	0.027 524
25.0	9.108 03	−11.941 1	1.387 29	2.333 26	−0.346 57	0.027 829
35.0	6.526 32	−7.611 65	−0.185 159	2.139 81	−0.405 364	0.022 096 8
45.0	4.679 21	−4.759 38	−0.415 014	0.649 477	0.260 209	0.007 096 6
55.0	3.468 63	−2.374 48	−3.275 72	3.546 74	−0.999 072	0.028 679
			Ethanol–Nonanol			
15.0	13.143 9	−13.152 6	−4.828 37	8.450 35	−2.303 47	0.379 53
25.0	9.098 84	−9.345 24	0.342 402	0.307 139	0.678 855	0.351 205
35.0	6.521 40	−6.667 31	1.976 85	−2.473 60	1.543 13	0.023 070
45.0	4.676 38	−4.086 19	0.643 39	−1.538 51	1.060 3	0.014 490 5
55.0	3.475 87	−2.855 5	0.503 70	−1.121 62	0.633 199	0.007 500 2
			Propanol–Nonanol			
15.0	13.148 9	−10.404 7	−9.725 2	15.471 7	−6.157 07	0.029 722
25.0	9.102 25	−8.395 02	−0.172 42	2.081 26	−0.780 816	0.024 729
35.0	6.522 03	−6.207 87	2.048 91	−1.601 89	0.696 724	0.018 149 8
45.0	4.678 49	−4.004 09	1.806 39	−2.882 77	1.621 30	0.007 182 6
55.0	3.476 72	−2.762 31	1.184 16	−2.012 91	1.102 64	0.004 329 0
			Butanol–Nonanol			
15.0	13.135 6	−11.952 8	0.576 935	1.719 58	−0.277 45	0.045 578
25.0	9.095 89	−9.341 59	6.364 09	−7.030 12	3.443 02	0.020 389
35.0	6.515 51	−6.991 90	7.033 72	−8.556 63	3.977 49	0.027 730
45.0	4.669 03	−3.670 17	−0.048 921	1.101 71	−0.493 12	0.056 225
55.0	3.473 77	−3.083 0	2.945 13	−3.974 66	1.887 66	0.008 629
			Methanol–Octanol			
15.0	10.663 0	−17.009 2	7.911 30	−1.573 4	0.627 403	0.014 548
25.0	7.363 81	−10.330 0	3.658 87	−0.644 49	0.493 51	0.008 161 5
35.0	5.256 29	−6.360 68	1.267 23	−0.040 657	0.349 462	0.006 075
45.0	3.868 80	−4.104 05	0.484 82	−0.337 46	0.505 059	0.004 896 8
55.0	2.890 99	−2.632 68	−0.010 37	−0.332 29	0.453 40	0.003 431 1
			Ethanol–Octanol			
15.0	10.662 3	−14.180 1	3.340 31	3.322 0	−1.856 42	0.010 532
25.0	7.361 45	−9.256 48	4.077 24	−2.095 89	0.987 38	0.018 673
35.0	5.256 00	−5.217 16	−0.686 48	2.576 46	−1.041 93	0.004 136 8
45.0	3.868 60	−3.590 05	−0.054 27	0.749 961	−0.225 139	0.002 436 7
55.0	2.890 76	−2.369 19	−0.253 64	0.459 664	−0.096 460	0.001 383 0
			Propanol–Octanol			
15.0	10.665 0	−10.461 7	−0.532 237	5.438 25	−2.782 78	0.049 887
25.0	7.361 28	−6.794 71	1.172 85	0.443 472	−0.343 69	0.022 588
35.0	5.252 59	−7.220 41	12.082 2	−14.803 9	6.147 86	0.029 603
45.0	3.871 93	−3.309 94	1.569 08	−1.631 67	0.717 563	0.033 235
55.0	2.892 64	−3.393 71	5.906 74	−7.860 73	3.446 90	0.027 647
			Butanol–Octanol			
15.0	10.660 4	−9.851 35	2.013 69	1.401 64	−1.038 86	0.028 599
25.0	7.363 70	−6.147 10	1.604 14	−0.805 49	0.505 984	0.003 685
35.0	5.256 78	−3.981 03	0.862 344	−0.498 34	0.326 184	0.003 818 2

Table IV (Continued)

temp, °C	A	B	C	D	E	rms error
45.0	3.868 97	−2.733 36	0.689 260	−0.629 644	0.359 875	0.005 133 5
55.0	2.892 96	−1.714 08	−0.213 536	0.338 578	−0.062 032	0.009 402 6
			Methanol–Heptanol			
15.0	8.302 12	−14.153 2	9.853 54	−5.098 60	1.719 48	0.007 265
25.0	5.774 25	−8.080 30	3.318 36	−0.941 969	0.472 38	0.005 676
35.0	4.263 39	−5.457 85	1.966 28	−0.649 72	0.349 635	0.002 859
45.0	3.145 70	−3.453 83	0.973 97	−0.871 64	0.623 356	0.006 198 7
55.0	2.364 67	−2.060 66	−0.268 275	0.202 856	0.130 148	0.003 283
			Ethanol–Heptanol			
15.0	8.299 35	−11.351 2	5.343 21	−0.508 66	−0.477 709	0.023 642
25.0	5.773 04	−6.373 35	0.598 299	2.406 72	−1.331 62	0.013 434
35.0	4.262 34	−4.527 16	0.949 560	0.710 248	−0.498 834	0.012 120 2
45.0	3.153 95	−2.479 82	−2.022 38	4.220 71	−2.105 31	0.046 015
55.0	2.364 65	−1.662 82	−0.871 664	1.366 76	−0.561 34	0.006 303 3
			Propanol–Heptanol			
15.0	8.299 93	−8.385 26	3.450 22	−0.714 618	−0.307 20	0.027 027
25.0	5.773 56	−4.319 71	−1.102 53	3.084 36	−1.604 27	0.016 135 3
35.0	4.261 94	−3.509 08	1.275 57	−0.549 53	−0.026 697	0.016 629 7
45.0	3.157 26	−2.315 46	1.105 16	−1.507 96	0.782 215	0.004 578 7
55.0	2.366 87	−1.169 39	−0.874 57	1.000 99	−0.339 387	0.009 434 8
			Butanol–Heptanol			
15.0	8.295 47	−7.045 88	2.743 06	−0.324 91	−0.501 93	0.023 43
25.0	5.775 01	−3.781 99	0.058 004	0.762 847	−0.290 86	0.003 152 3
35.0	4.264 05	−3.198 57	2.344 56	−2.512 75	1.072 89	0.012 013 6
45.0	3.157 40	−1.937 82	0.866 140	−0.962 614	0.434 913	0.012 062 8
55.0	2.366 47	−1.169 18	0.074 282	−0.129 261	0.100 427	0.009 142 0

agreement to three significant figures. The estimated error in densities was $\pm 2 \times 10^{-5}$ g/mL. The good agreement of the experimental values of density with literature values allows the report of absolute viscosities instead of kinematic viscosities. The experimental values of absolute viscosities for the pure alkanols are reported at five temperatures in Table II, along with literature values for comparison. Observed viscosity values fall within the ranges of those reported in the literature. Literature values of C_7, C_9, and C_{10} were not available for these temperatures; however, interpolated data were in good agreement with the values reported in the literature (see footnotes to Table II). The estimated error of measurement in viscosities was 1×10^{-4} cP.

Viscosities of binary mixtures of 1-alkanols were fitted to McAllister, Auslander, and Herric's equations and also to three polynomials. The average values for the root mean square (rms) for all mixtures of C_{10} and C_7 investigated are given in Table III. The rms values of McAllister and Herric equations are for the natural logarithm of the rms, whereas that of Auslander's is in terms of viscosities and thus cannot be compared directly. A reasonable fit was found for all equations. A detailed table of rms values is available as supplementary material (see paragraph at end of text regarding supplementary material). It should be noted that all equations tested except polynomial I included fixed end values such that the experimental values of the pure components do not participate in the error determination.

The viscosities of binary mixtures at each temperature are reported in terms of the parameters of polynomial I in Table IV. It should be noted that parameter A is the best fit absolute viscosity for the high molecular weight component of the binary mixture whereas the sum of the other four parameters is the best fit value of the absolute viscosity of the low molecular weight component. This polynomial allows the calculation of viscosities of any mole fraction of the mixture within the rms error reported.

The ideal behavior of the molar volumes of the binary mixture of 1-alkanols as reported by $\bar{V} = X_1\bar{V}_1 + X_2\bar{V}_2$ is illustrated for two binary mixtures in Figure 1, where the molar volume is plotted vs. mole fraction. Viscous heats and preexponential factors as derived from the viscosities given by eq 8 are
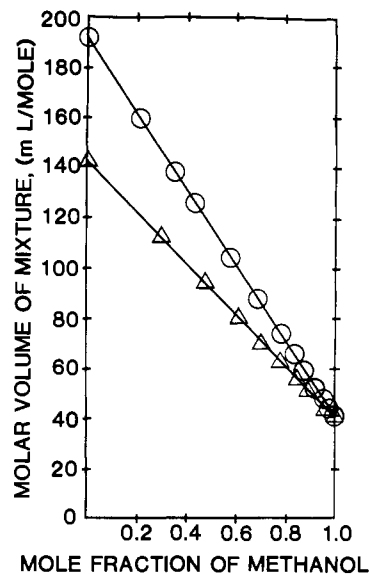

Figure 1. Molar volume of mixture vs. mole fraction of C_1 in binary mixtures of 1-alkanols: (O) C_1–C_{10}; (△) C_1–C_7.

Table V. Viscous Heats (E_V) and Preexponential Factors (η_0) of Various 1-Alkanols

	E_V, kcal/mol		$10^3\eta_0$, cP	
	obsd	lit.[a]	obsd	lit.[a]
C_1	2.50 ± 0.01	2.46 ± 0.04	8.2 ± 0.3	8.65
C_2	3.29 ± 0.01	3.22 ± 0.13	4.2 ± 0.4	4.65
C_3	3.91 ± 0.08	4.27 ± 0.28	2.56 ± 0.25	1.46
C_4	4.37 ± 0.05		1.64 ± 0.23	
C_7	5.85 ± 0.03		0.38 ± 0.08	
C_8	6.04 ± 0.02		0.276 ± 0.044	
C_9	6.17 ± 0.02		0.273 ± 0.025	
C_{10}	6.67 ± 0.04		0.153 ± 0.017	

[a] Reference 21.

presented in Table V. Viscous heats (SD = ±0.03 kcal/mol) and preexponential factors (SD = ±1.6 × 10^{-4} cP) as a function of mole fraction of lower alkanol in a typical binary mixture of 1-alkanols are depicted in Figures 2 and 3. Such figures
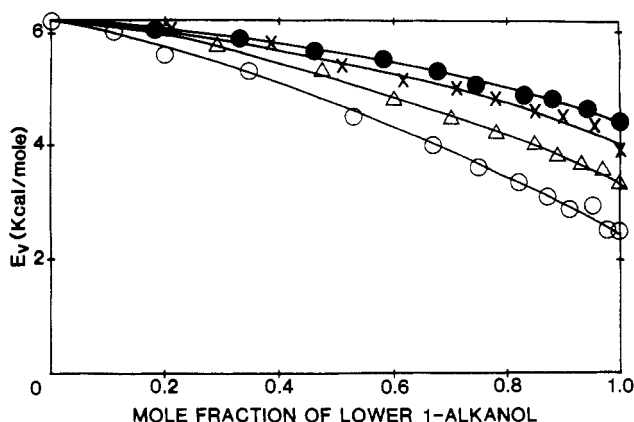
Figure 2. Viscous heat vs. mole fraction of lower 1-alkanol for binary mixtures of C_9 with C_1, C_2, C_3, and C_4: (O) C_1–C_9; (△) C_2–C_9; (X) C_3–C_9; (●) C_4–C_9.
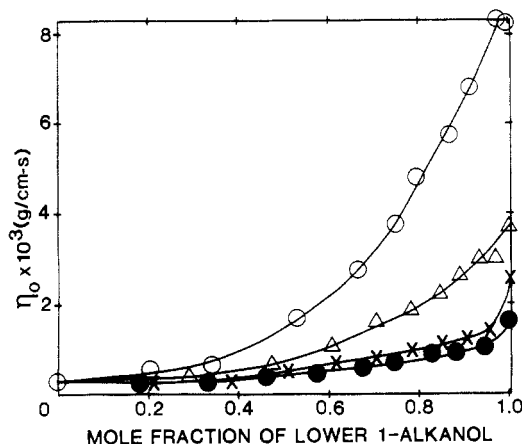
Figure 3. Preexponential factor vs. mole fraction of lower 1-alkanol for binary mixtures of C_9 with C_1, C_2, C_3, and C_4: (O) C_1–C_9; (△) C_2–C_9; (X) C_3–C_9; (●) C_4–C_9.

for all mixtures investigated are available as supplementary material.

Glossary

A, B, C, D, E	polynomial parameters
M_1	molecular weight of the lower component in a binary mixture
M_2	molecular weight of the higher component in a binary mixture
$\bar{V}$	molar volume of a sample
$\bar{V}_1$, $\bar{V}_2$	molar volumes of lower and higher components, respectively, in a binary mixture
X_1	mole fraction of the lower component in a binary mixture
X_2	mole fraction of the higher component in a binary mixture
α_{12}, α_{21}	interaction parameters
β_{12}, β_{21}	coefficients which need to be determined by eq 2
η	viscosity of a sample
η_1, η_2	viscosity of lower and higher components, respectively, in a binary mixture
ρ	density of a sample

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Solubility of Solid Acetic Acid in Liquid Organic Solvents

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The solubilities of acetic acid in some organic solvents (*n*-heptane, cyclopentane, carbon tetrachloride, toluene, chloroform, ethyl acetoacetate, methanol, ethanol, and acetone) have been measured for temperatures ranging from melting point to –30 °C. The results are compared with those predicted with UNIFAC and those calculated with the NRTL equation (vapor–liquid equilibrium (VLE) parameters).

Introduction

The experimental measurements of vapor–liquid (VLE), liquid–liquid (LLE), and liquid–solid equilibria available provide an abundance of information both theoretical and of interest in process design. While numerous experimental data are readily available for the first two types of equilibria (VLE and LLE), this is not so for liquid–solid equilibria (*1*, *2*).