

Densities and Viscosities of 1-Pentanol Binary Mixtures at 293.15 K

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Densities (ρ) and viscosities (η) for binary mixtures of 1-pentanol with benzene, chlorobenzene, bromobenzene, iodobenzene, nitrobenzene, aniline, toluene, and *p*-xylene have been measured over the whole composition range at 293.15 K. The excess volumes V^E and the viscosity deviations $\Delta\eta$ were calculated, and the results were fitted to a Redlich–Kister type polynomial relation. The corresponding parameters have been derived. The resulting excess functions were interpreted in terms of the interactions between the molecules in the binary mixtures.

Introduction

A study of molecular interactions in binary liquid mixtures has been the subject of active investigation in our laboratories in recent years (Palaiologou and Molinou, 1995; Tsierkezos and Molinou, 1998).

Alcohols are interesting polar solvents, self-associated through hydrogen bonding, creating multimers of different degrees (Dewan et al., 1991; Dewan et al., 1992).

Recently, excess volumes and viscosity deviations of binary mixtures of alkanols have been extensively studied (Dewan et al., 1991; Dewan et al., 1992; Aminabhavi et al., 1993; Nikam et al., 1995; Bhardwaj et al., 1996; Garcia et al., 1997). Thermodynamic properties of binary mixtures containing substituted benzenes have also been reported (Qin et al., 1992; Yadava et al., 1994; Ramadevi and Prabhakara Rao, 1995; Ramachandran et al., 1995).

1-Pentanol in its pure form is assumed to create monomers and cyclic dimers (Dewan et al., 1992). A mixture of 1-pentanol with another organic compound will lead to specific interactions depending on the proton-accepting ability of the other molecule. This process will determine the volume deviation from ideality.

Regarding values of V^E of mixtures of lower alkanols with benzene and its substitutes, already measurements for methanol, ethanol, 1-propanol, 1-butanol with nitrobenzene at 298.15 and 303.15 K (Nikam et al., 1995), 1-butanol with benzene at 298.15 and 308.15 K (Yu and Tsai, 1994), and 1-butanol with *p*-xylene at 308.15 K (Bhardwaj et al., 1996) have been reported.

In this work we report the experimental densities and viscosities for binary mixtures of 1-pentanol with benzene, chlorobenzene, bromobenzene, iodobenzene, nitrobenzene, aniline, toluene, and *p*-xylene at 293.15 K. From these results the excess volumes and viscosity deviations were derived. The aim of this work is to compare the effect of the various substituents on the benzene molecule on the intermolecular interactions.

No literature data on excess molar volumes and viscosities are available for these mixtures except for benzene + 1-pentanol at 298.15 and 323.15 K (Ortega and Paz-Andrade, 1986) and for the same mixture at 298.15 and 308.15 K (Yu and Tsai, 1994).

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Table 1. Comparison of Experimental Densities ρ and Refractive Indices n_D of Pure Liquids with Literature Values at 293.15 K

	$\rho(\text{exp})/\text{g cm}^{-3}$	$\rho(\text{ref})/\text{g cm}^{-3}$	$n_D(\text{exp})$	$n_D(\text{ref})$
1-pentanol	0.814 68	0.814 80 ^a	1.4108	1.4100 ^a
benzene	0.878 91	0.879 32 ^b 0.879 20 ^f 0.878 66 ^k	1.5015	1.50113 ^j 1.50111 ^k
chlorobenzene	1.106 46	1.105 86 ^k 1.106 60 ^l 1.106 53 ^h	1.5247	1.5250 ^g 1.5244 ^h 1.52412 ^k 1.5248 ^l
bromobenzene	1.494 66	1.495 04 ^k 1.495 90 ^l	1.5597	1.5600 ^g 1.55971 ^k 1.5601 ^l
iodobenzene	1.825 92	1.831 60 ^l	1.6196	1.6201 ^l
nitrobenzene	1.203 29	1.202 60 ^l	1.5527	1.5525 ^g 1.5521 ^l
aniline	1.021 66	1.020 58 ⁱ	1.5860	1.5865 ^g
toluene	0.866 89	0.867 20 ^b 0.867 00 ^c 0.866 95 ^k	1.4971	1.4970 ^g 1.49677 ^j 1.49612 ^k
<i>p</i> -xylene	0.863 14	0.861 27 ^b 0.861 20 ^d 0.860 73 ^e 0.861 30 ^f	1.4962	1.4958 ^d

^a TRC Databases for Chemistry and Engineering 1991–98. ^b Qin et al., 1992. ^c Palaiologou and Molinou, 1995. ^d Avraam et al., 1998. ^e Serrano et al., 1990. ^f Yadava et al., 1994. ^g Ramadevi and Prabhakara Rao, 1995. ^h Kalali et al., 1992. ⁱ Patil et al., 1999. ^j Fornfeld-Schwarz and Svejda, 1999. ^k Aminabhavi et al., 1982. ^l Korosi and Kovats, 1981.

Experimental Section

Materials. The reagents toluene (Merck p.a. >99.5%), aniline (Merck p.a. >99.8%), benzene (Merck p.a. >99.5%), chlorobenzene (Fluka p.a. >99.0%), bromobenzene (Fluka p.a. >99.0%), iodobenzene (Fluka p.a. >99.5%), 1-pentanol (Fluka p.a. >99.5%), nitrobenzene (Fluka p.a. >99.5%), and *p*-xylene (Merck >99.0%) were used without purification. The purity of the liquids was assessed by comparing the densities and refractive indices with the literature values (Table 1). The agreement was satisfactory.

The binary mixtures were prepared by mass (Mettler A210P, ± 0.01 mg). The mole fractions were known to ± 0.0001 in all cases.

Table 2. Experimental Densities ρ and Viscosities η of 1-Pentanol Binary Mixtures

x_1	$\rho/\text{g cm}^{-3}$	$\eta/\text{mPa s}$	x_1	$\rho/\text{g cm}^{-3}$	$\eta/\text{mPa s}$
1-Pentanol (1) + Benzene (2)					
0.0000	0.878 91	0.657	0.5800	0.836 62	1.532
0.0659	0.872 67	0.671	0.6971	0.830 06	2.016
0.1351	0.866 85	0.715	0.7290	0.828 37	2.159
0.2180	0.860 34	0.791	0.8032	0.824 46	2.568
0.2928	0.854 89	0.876	0.8704	0.821 16	3.014
0.4304	0.845 57	1.127	0.9454	0.817 31	3.594
0.5061	0.840 99	1.307	1.0000	0.814 68	4.030
1-Pentanol (1) + Chlorobenzene (2)					
0.0000	1.106 46	0.806	0.4687	0.965 95	1.387
0.0511	1.090 35	0.816	0.5602	0.939 54	1.637
0.1017	1.074 81	0.852	0.5789	0.934 16	1.697
0.1449	1.061 66	0.892	0.7339	0.889 95	2.339
0.1980	1.045 63	0.936	0.8618	0.853 72	3.062
0.2665	1.025 21	1.017	0.9524	0.828 15	3.666
0.3688	0.994 94	1.195	1.0000	0.814 68	4.030
1-Pentanol (1) + Bromobenzene (2)					
0.0000	1.494 66	1.142	0.4182	1.206 17	1.570
0.0631	1.450 27	1.140	0.4909	1.156 86	1.716
0.1715	1.375 08	1.212	0.5969	1.085 19	2.011
0.2092	1.349 10	1.252	0.6886	1.023 43	2.346
0.2807	1.300 05	1.335	0.7544	0.979 11	2.636
0.3259	1.269 06	1.398	0.8796	0.895 26	3.294
0.3831	1.230 04	1.512	1.0000	0.814 68	4.030
1-Pentanol (1) + Iodobenzene (2)					
0.0000	1.825 92	1.736	0.7761	1.047 83	2.915
0.1000	1.727 34	1.665	0.8055	1.017 51	3.043
0.2000	1.628 59	1.694	0.8184	1.004 18	3.100
0.2500	1.579 12	1.709	0.8554	0.965 86	3.273
0.3834	1.446 38	1.864	0.9419	0.875 74	3.700
0.4289	1.400 74	1.940	0.9750	0.841 16	3.867
0.5101	1.318 99	2.098	1.0000	0.814 68	4.030
1-Pentanol (1) + Nitrobenzene (2)					
0.0000	1.203 29	2.024	0.2976	1.083 71	1.918
0.0258	1.192 45	1.964	0.3319	1.070 26	1.938
0.0556	1.180 36	1.924	0.3827	1.050 39	2.003
0.1040	1.160 78	1.867	0.4677	1.017 32	2.104
0.1800	1.130 32	1.856	0.6694	0.939 82	2.475
0.2040	1.120 74	1.860	0.8217	0.882 06	2.982
0.2635	1.097 18	1.899	1.0000	0.814 68	4.030
1-Pentanol (1) + Aniline (2)					
0.0000	1.021 66	4.404	0.7695	0.858 25	3.258
0.1207	0.993 72	3.714	0.8055	0.851 38	3.326
0.2883	0.956 50	3.273	0.8557	0.841 84	3.444
0.3221	0.949 18	3.227	0.9099	0.831 63	3.605
0.3789	0.937 03	3.161	0.9484	0.824 41	3.756
0.4074	0.931 06	3.138	0.9525	0.823 65	3.775
0.5661	0.898 30	3.087	1.0000	0.814 68	4.030
1-Pentanol (1) + Toluene (2)					
0.0000	0.866 89	0.592	0.5308	0.838 62	1.290
0.0767	0.862 35	0.623	0.6010	0.835 14	1.512
0.1550	0.858 03	0.678	0.6728	0.831 53	1.795
0.2336	0.853 82	0.747	0.7788	0.826 19	2.338
0.3074	0.849 96	0.834	0.8616	0.821 97	2.883
0.3459	0.847 97	0.888	0.8898	0.820 51	3.096
0.4529	0.842 54	1.094	1.0000	0.814 68	4.030
1-Pentanol (1) + <i>p</i> -Xylene (2)					
0.0000	0.863 14	0.655	0.5534	0.837 97	1.356
0.1458	0.856 50	0.727	0.6142	0.835 09	1.543
0.2275	0.852 85	0.792	0.7185	0.829 96	1.973
0.2722	0.850 84	0.836	0.7687	0.827 41	2.232
0.3455	0.847 53	0.925	0.8498	0.823 15	2.747
0.4093	0.844 64	1.022	0.9425	0.818 04	3.494
0.4819	0.841 31	1.164	1.0000	0.814 68	4.030

Measurements. A digital densimeter (Anton Paar, model DMA 58) was used for the determination of the densities of the pure components and the binary mixtures. The estimated uncertainty of the measured densities is $\pm 0.00001 \text{ g}\cdot\text{cm}^{-3}$. The DMA cell was calibrated with dry air and doubly distilled water at atmospheric pressure. The

sample size was 0.7 cm^3 , and the sample thermostat was controlled to $\pm 0.01 \text{ K}$.

The flow times of the mixtures and the pure liquids were measured with an automated Schott Geräte AVS 310 Unit, equipped with an Ubbelohde capillary viscometer. The viscometer was calibrated with doubly distilled water. The accuracy in the flow times was $\pm 0.01 \text{ s}$. The viscometer was placed in a thermostat (model Schott CT 050/2) maintained constant within $\pm 0.03 \text{ K}$. The absolute viscosities η were calculated from the kinematic viscosities ν using the density values ρ for each mixture ($\eta = \nu\rho$). The accuracy of the kinematic viscosities ($\pm 0.001 \text{ mm}^2\cdot\text{s}^{-1}$) and the densities ($\pm 0.00001 \text{ g}\cdot\text{cm}^{-3}$) gives the maximum uncertainty $\pm 0.001 \text{ mPa}\cdot\text{s}$ in the absolute viscosities.

Results and Discussion

The experimental values of density and viscosity for all mixtures at different compositions and at 293.15 K are listed in Table 2.

The values of excess molar volumes V^E and viscosity deviations $\Delta\eta$ were calculated from the experimental data according to the following equations:

$$V^E = V_m - \sum_{i=1}^2 V_i x_i \quad (1)$$

$$\Delta\eta = \eta_m - \sum_{i=1}^2 \eta_i x_i \quad (2)$$

where x_i , V_i , and η_i represent the mole fraction, the molar volume, and the viscosity of the i th pure component of the mixture. V_m and η_m are the molar volume and the viscosity of the mixture, respectively. The molar volume V_m of the mixture was calculated from the following equation:

$$V_m = \sum_{i=1}^2 x_i M_i / \rho_m \quad (3)$$

where ρ_m is the mixture density and M_i is the molecular weight of component i in the mixture.

The experimental values of V^E and $\Delta\eta$ are fitted to the Redlich–Kister equation:

$$Y = x_1 x_2 \sum_{k=0}^n A_k (2x_1 - 1)^k \quad (4)$$

where Y represents the excess volume or the viscosity deviations and A_k represents the parameters.

The number of parameters A_k used was obtained from the calculated and experimental values according to the equation

$$\sigma = \left[\frac{\sum (Y_{\text{expt}} - Y_{\text{calcd}})^2}{(n - p)} \right]^{1/2} \quad (5)$$

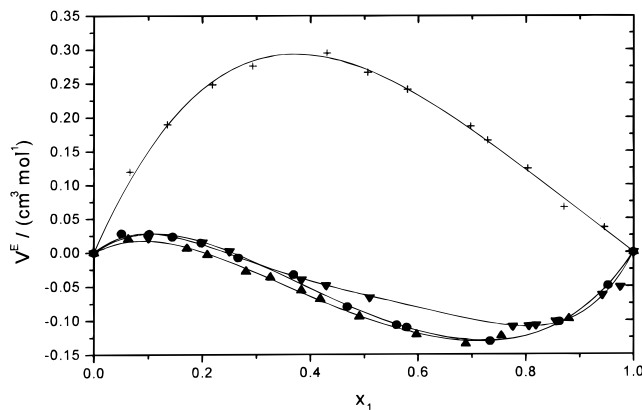
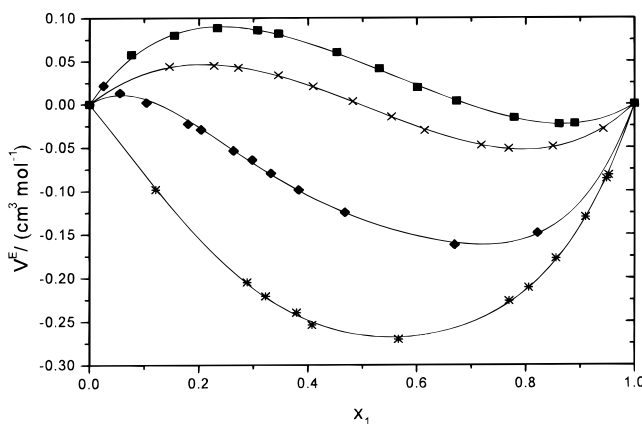
where n and p are the number of experimental points and parameters, respectively, and σ is the standard deviation. The values of A_k and σ are presented in Table 3.

In Figures 1 and 2 the V^E values are positive for a 1-pentanol + benzene mixture, change to sigmoidal for aryl halides, toluene, and *p*-xylene, and are negative for nitrobenzene and aniline.

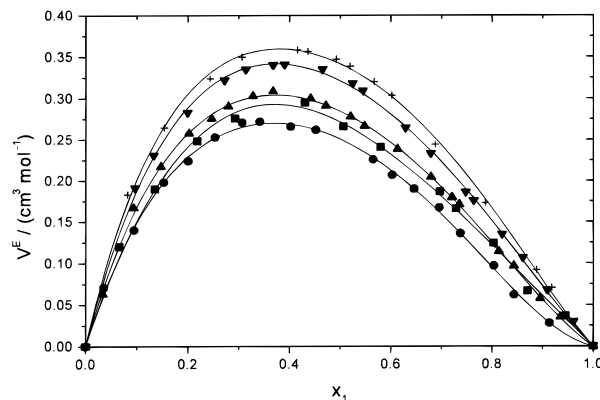
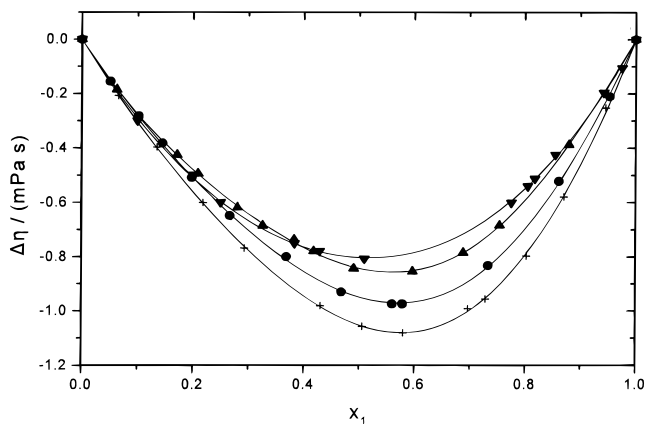
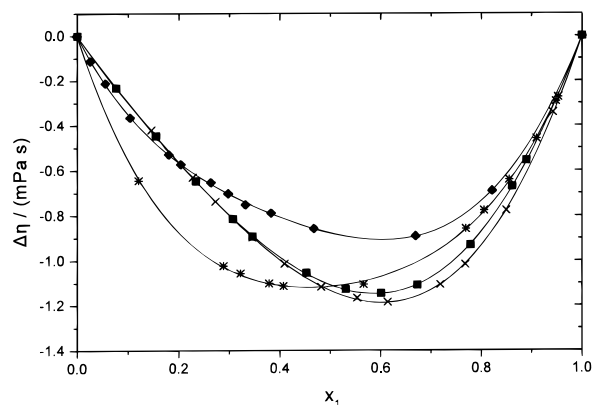
The behavior of the 1-pentanol + benzene mixture may be attributed to the disruption of the hydrogen-bonded alcohol by the nonpolar benzene (Yu and Tsai, 1994).

Table 3. Parameters and Standard Deviations of Excess Functions of 1-Pentanol Binary Mixtures

function	A_0	A_1	A_2	A_3	σ
1-Pentanol (1) + Benzene (2)					
$V^E/\text{cm}^3 \text{ mol}^{-1}$	1.0857	-0.6211	0.1417		0.0065
$\Delta\eta/\text{mPa s}$	-4.2109	-1.4039	-0.0845	0.4951	0.0088
1-Pentanol (1) + Chlorobenzene (2)					
$V^E/\text{cm}^3 \text{ mol}^{-1}$	-0.3516	-0.6410	0.0518	-0.2230	0.0040
$\Delta\eta/\text{mPa s}$	-3.7985	-1.2033	0.0536	0.5878	0.0078
1-Pentanol (1) + Bromobenzene (2)					
$V^E/\text{cm}^3 \text{ mol}^{-1}$	-0.3830	-0.5602	0.0069	-0.2360	0.0041
$\Delta\eta/\text{mPa s}$	-3.3715	-0.9160	0.0445	0.8927	0.0064
1-Pentanol (1) + Iodobenzene (2)					
$V^E/\text{cm}^3 \text{ mol}^{-1}$	-0.2490	-0.3792	-0.1517	-0.6929	0.0075
$\Delta\eta/\text{mPa s}$	-3.2162	-0.2121	-0.2819		0.0082
1-Pentanol (1) + Nitrobenzene (2)					
$V^E/\text{cm}^3 \text{ mol}^{-1}$	-0.5317	-0.4622	-0.0960	-0.5766	0.0029
$\Delta\eta/\text{mPa s}$	-3.5097	-1.0677	-1.5524	0.4790	0.0053
1-Pentanol (1) + Aniline (2)					
$V^E/\text{cm}^3 \text{ mol}^{-1}$	-1.0625	-0.1678	-0.2529	-0.3953	0.0026
$\Delta\eta/\text{mPa s}$	-4.4614	0.4033	-2.1165		0.0084
1-Pentanol (1) + Toluene (2)					
$V^E/\text{cm}^3 \text{ mol}^{-1}$	0.1905	-0.5031	0.0740	-0.1559	0.0017
$\Delta\eta/\text{mPa s}$	-4.4324	-1.7122	-0.1572	0.3270	0.0040
1-Pentanol (1) + <i>p</i> -Xylene (2)					
$V^E/\text{cm}^3 \text{ mol}^{-1}$	-0.0065	-0.4934	-0.0339	-0.0589	0.0011
$\Delta\eta/\text{mPa s}$	-4.5335	-2.0052	-0.3895	0.2037	0.0054

**Figure 1.** Excess molar volumes for 1-pentanol (1) + benzene (2) (+), chlorobenzene (2) (●), bromobenzene (2) (▲), and iodobenzene (2) (▼).**Figure 2.** Excess molar volumes for 1-pentanol (1) + toluene (2) (■), *p*-xylene (2) (×), nitrobenzene (2) (◆), and aniline (2) (*).

The sigmoidal curves show that the negative values are at the 1-pentanol-rich region. The breaking up of the self-associated 1-pentanol, which leads to a volume increase,

**Figure 3.** Excess molar volumes for 1-pentanol (1) + benzene (2) at different temperatures. This work at 293.15 K (■); Yu and Tsai at 298.15 K (●); Ortega and Paz-Andrade at 308.15 K (▲), 318.15 K (▼), and 323.15 K (+).**Figure 4.** Viscosity deviations of 1-pentanol (1) + benzene (2) (+), chlorobenzene (2) (●), bromobenzene (2) (▲), and iodobenzene (2) (▼).**Figure 5.** Viscosity deviations of 1-pentanol (1) + toluene (2) (■), *p*-xylene (2) (×), nitrobenzene (2) (◆), and aniline (2) (*).

and the specific interactions, such as hydrogen bond formation between the 1-pentanol and the substituted benzene (Crabtree, 1998; Rastogi et al., 1967; Garcia et al., 1997), which lead to a volume reduction, seem to contribute to the sigmoidal curves.

The strong negative values for the mixtures 1-pentanol + aniline can be interpreted as a consequence of the strong attractions appearing between the hydrogen of the OH group and the lone pair of electrons in the NH_2 group. Furthermore, the high dipole moment of nitrobenzene (3.96 D at 20 °C) (Daniels and Alberty, 1975) favors dipole-dipole interactions, leading also to a volume reduction

the 1-pentanol + nitrobenzene mixture (Nikam et al., 1995).

In Figure 3 we compare our V^E values of the binary 1-pentanol + benzene mixture with the reference values. The agreement is quite satisfactory.

The results for $\Delta\eta$ are represented graphically in Figures 4 and 5. The $\Delta\eta$ values are negative for all systems investigated. For the mixtures 1-pentanol + benzene, + chlorobenzene, + bromobenzene, and + iodobenzene (Figure 4), the algebraic values of $\Delta\eta$ fall in the order iodobenzene > bromobenzene > chlorobenzene > benzene, reflecting an easier flow of the mixture at the direction of decreasing molecular weight of the aryl halide.

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