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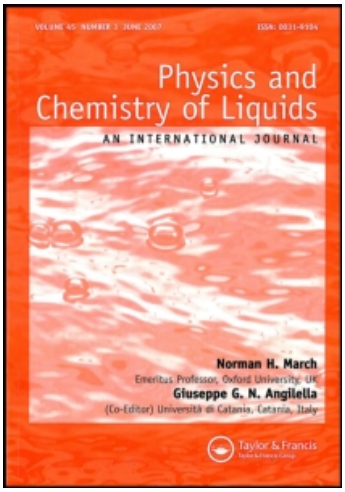

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Deviations in refractive indices and applicability of mixing rules in aniline + alkanol binary mixtures at different temperatures

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Deviations in refractive indices and applicability of mixing rules in aniline + alkanol binary mixtures at different temperatures

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The refractive indices, n of binary mixtures of aniline with 1-propanol, 2-propanol, 2-methyl-1-propanol, 2-methyl-2-propanol, including those of pure liquids, covering the whole composition range have been measured at 293.15, 298.15, 303.15, 308.15, 313.15 and 318.15 K. From the experimental data, the deviations in refractive index, Δn has been calculated. The variation of Δn with composition and temperature of the mixtures has been discussed in terms of molecular interaction in these mixtures. It is observed that the extent of the deviation Δn for these mixtures follows the sequence: 1-propanol < 2-propanol < 2-methyl-1-propanol < 2-methyl-2-propanol, indicating the presence of strong interactions in these mixtures in the same order. Further, the refractive indices of these binary mixtures were calculated theoretically from the refractive index data of pure components and densities of the mixtures by using various empirical and semi-empirical relations and the results were compared with the experimental findings.

Keywords: refractive index; aniline; alkanol; molecular interactions

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1. Introduction

The refractive index, n, is an important physical property, which affects the solution of different problems in chemical engineering in order to develop industrial processes. The knowledge of n of multicomponent mixtures provides substantial information about molecular influence on the intensity of the interactions in the mixtures [1–5]. Prediction of n of multicomponent liquid mixtures is essential for many physicochemical calculations involving multiphase systems [4]. In previous studies [6–12] we have reported the volumetric, acoustic, transport and refractive properties of non-aqueous binary mixtures. Here we report the results of our studies on refractive indices of the binary mixtures of aniline with 1-propanol, 2-propanol, 2-methyl-1-propanol, 2-methyl-2-propanol, over the entire composition range at various temperatures. Aniline molecules are polar ($\mu = 1.51$ D at 298.15 K) [13] and self-associated through hydrogen bonding of their amino groups [14], and alkanol molecules are polar and self-associated through hydrogen bonding of their hydroxyl groups [15].

Aniline is used in the manufacture of synthetic dyes, drugs and as an accelerator in vulcanisation of rubber; and the alkanols are of interest in their own right and serve as

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simple examples of biologically and industrially important amphiphilic materials [16]. Therefore, the study of intermolecular interactions in aniline + alkanol mixtures would be interesting owing to their industrial applications. Literature survey indicates that there has been no temperature-dependent study of these systems from the point of view of their refractive index behaviour.

The present article reports the refractive indices of aniline + 1-propanol/2-propanol/2-methyl-1-propanol/2-methyl-2-propanol binary mixtures, including the pure liquids, covering the entire composition range expressed by mole fraction, x_1 of aniline at 293.15, 298.15, 303.15, 308.15, 313.15 and 318.15 K. The experimental values of n have been used to calculate the deviations in refractive index, Δn; and the results were discussed in terms of molecular interactions in these mixtures. Further, the refractive indices of these binary mixtures were calculated theoretically from the refractive index data of pure components and densities of the mixtures [17] by using various empirical and semi-empirical relations [18–23] and the results are compared with the experimental findings.

2. Experimental details

Aniline, 1-propanol, 2-propanol, 2-methyl-1-propanol and 2-methyl-2-propanol used in the study were the products from S. D. Fine-Chem Ltd. (India) and were purified by using the methods described in the literature [24,25]; the mass fraction purities of chemicals used, as determined by gas chromatography are: aniline > 0.996, 1-propanol > 0.996, 2-propanol > 0.995, 2-methyl-1-propanol > 0.995 and 2-methyl-2-propanol > 0.993. Before use, the chemicals were stored over 0.4 nm molecular sieves for 72 h to remove water content, as far as possible, and were degassed at low pressure. The mixtures were prepared by mass and were kept in special airtight stopper glass bottles to avoid evaporation. The weightings were done with an electronic balance with a precision of ±0.1 mg. The average uncertainty in the mole fraction was estimated to be less than ±0.0001.

The refractive indices of pure liquids and their binary mixture were measured by using a thermostated Abbe refractometer. The refractometer was calibrated by measuring the refractive indices of triply distilled water and toluene at desired temperatures. The values of refractive index were obtained using sodium D light. The reproducibility of refractive index measurements was within ±0.0001. The temperature of the test liquid sample in the prisms was maintained at the desired value by circulating water through the jacket around the prisms from a thermostatic water bath (JULABO, Model ME-31A, Germany) with an accuracy of ±0.01 K. The reliability of experimental measurements of n was ascertained by comparing the experimental data of pure liquids with the corresponding values available in the literature [24] at 298.15 K. The experimental values of n of pure aniline, 1-propanol, 2-propanol, 2-methyl-1-propanol, 2-methyl-2-propanol obtained at 298.15 K are 1.5839, 1.3836, 1.3750, 1.3941 and 1.3852, respectively; and the corresponding literature [24] values are 1.58364, 1.3837, 1.3752, 1.3939 and 1.3850, respectively.

3. Results and discussion

The experimental values of refractive index, n of the binary mixtures of aniline with 1-propanol, 2-propanol, 2-methyl-1-propanol, 2-methyl-2-propanol, as a function of mole

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fraction, x_1 of aniline at different temperatures are listed in Table 1. The deviations in refractive index, Δn have been calculated by using the following relation [1]:

$$\Delta n = n - (\phi_1 n_1 + \phi_2 n_2), \tag{1}$$

where ϕ is the volume fraction (calculated using the molar volumes of the pure components obtained from the density data from our earlier work [17]) and the subscripts 1 and 2 represent pure components, aniline and alkanol, respectively. The values of Δn were fitted to a Redlich–Kister [26] type polynomial equation of the form

$$\Delta n = \phi_1(1 - \phi_1) \sum_{i=1}^{5} A_i (1 - 2\phi_1)^{i-1}. \tag{2}$$

The values of coefficients, A_i evaluated by the method of least-squares, with all points weighted equally, together with the corresponding standard deviation, σ calculated by using the relation

$$\sigma = \left[\frac{\sum (\Delta n_{\text{Expt.}} - \Delta n_{\text{Calcd}})^2}{(m-k)} \right]^{1/2}, \tag{3}$$

where m is the number of experimental data points and k is the number of coefficients considered ($k = 5$ in the present calculation), are listed in Table 2. The values of Δn_{Cal} were obtained from Equation (2) by using the best-fit values of coefficient A_i. The variations of Δn with composition (in terms of mole fraction of aniline) of the mixture along with smoothed Δn values by using the Equation (2), at 298.15 and 318.15 K are shown in Figure 1.

A perusal of Figure 1 indicates that Δn values are positive over the entire composition range and at all temperatures investigated for all the four binary mixtures (aniline + 1-propanol/2-propanol/2-methyl-1-propanol/2-methyl-2-propanol). In general, the positive deviations Δn (on volume fraction dependence basis) is considered due to the presence of significant interactions in the mixtures, whereas negative deviations Δn indicates weak interactions between the components of the mixture [1,5–7]. The extent of the positive deviation Δn from linear dependence on composition follows the sequence: 1-propanol < 2-propanol < 2-methyl-1-propanol < 2-methyl-2-propanol. The observed trends (Figure 1) of Δn values indicate the presence of significant interactions in these mixtures, which follow the order: 1-propanol < 2-propanol < 2-methyl-1-propanol < 2-methyl-2-propanol. This further reinforces our earlier conclusions regarding the intermolecular interactions from the variations of V_{m}^{E} values [17] of these mixtures. Also, the deviations Δn are found opposite to the sign of excess molar volumes V_{m}^{E} for all the four binary mixtures, which is in agreement with the view proposed by others [1,5].

The refractive indices of all the binary liquid mixtures have been theoretically calculated from the refractive index data of pure components and densities of the mixtures by using various mixing rules [18–24].

Lorentz–Lorenz (L-L) relation

$$\left[\frac{(n^2-1)}{(n^2+2)}\right] = \left[\frac{(n_1^2-1)}{(n_1^2+2)}\right]\varphi_1 + \left[\frac{(n_2^2-1)}{(n_2^2+2)}\right]\varphi_2, \tag{4}$$

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Table 1. Values of refractive index, n as a function of mole fraction, x_1 of aniline for the aniline + alkanol mixtures at different temperatures.

	T (K)					
x_1	293.15	298.15	303.15	308.15	313.15	318.15
Aniline + 1-propanol						
0.0000	1.3855	1.3836	1.3817	1.3799	1.3781	1.3763
0.1177	1.4141	1.4121	1.4100	1.4081	1.4062	1.4043
0.2056	1.4345	1.4324	1.4303	1.4283	1.4262	1.4243
0.3156	1.4589	1.4567	1.4545	1.4524	1.4504	1.4483
0.4112	1.4791	1.4769	1.4747	1.4725	1.4703	1.4682
0.5106	1.4993	1.4970	1.4947	1.4924	1.4902	1.4880
0.6141	1.5193	1.5170	1.5146	1.5123	1.5100	1.5078
0.7219	1.5391	1.5367	1.5343	1.5319	1.5295	1.5272
0.8114	1.5550	1.5526	1.5501	1.5477	1.5452	1.5428
0.9041	1.5707	1.5682	1.5657	1.5632	1.5608	1.5584
1.0000	1.5864	1.5839	1.5814	1.5789	1.5764	1.5740
Aniline + 2-propanol						
0.0000	1.3770	1.3750	1.3730	1.3709	1.3689	1.3668
0.1201	1.4068	1.4047	1.4026	1.4004	1.3983	1.3961
0.2094	1.4281	1.4259	1.4237	1.4215	1.4193	1.4171
0.3017	1.4493	1.4471	1.4449	1.4426	1.4403	1.4381
0.4167	1.4746	1.4723	1.4700	1.4677	1.4654	1.4631
0.5163	1.4956	1.4933	1.4909	1.4885	1.4862	1.4838
0.6090	1.5144	1.5120	1.5096	1.5072	1.5048	1.5024
0.7047	1.5331	1.5307	1.5283	1.5258	1.5234	1.5210
0.8149	1.5538	1.5513	1.5488	1.5463	1.5438	1.5414
0.9060	1.5701	1.5676	1.5651	1.5626	1.5601	1.5577
1.0000	1.5864	1.5839	1.5814	1.5789	1.5764	1.5740
Aniline + 2-methyl-1-propanol						
0.0000	1.3961	1.3941	1.3920	1.3899	1.3878	1.3857
0.1093	1.4173	1.4152	1.4130	1.4108	1.4086	1.4065
0.2145	1.4377	1.4355	1.4333	1.4310	1.4288	1.4266
0.3099	1.4560	1.4538	1.4515	1.4493	1.4470	1.4447
0.4058	1.4744	1.4721	1.4698	1.4674	1.4651	1.4629
0.5038	1.4930	1.4907	1.4883	1.4860	1.4836	1.4813
0.6061	1.5124	1.5101	1.5076	1.5052	1.5028	1.5005
0.7087	1.5318	1.5294	1.5270	1.5245	1.5221	1.5197
0.8090	1.5507	1.5482	1.5458	1.5433	1.5408	1.5384
0.9051	1.5687	1.5662	1.5637	1.5612	1.5587	1.5563
1.0000	1.5864	1.5839	1.5814	1.5789	1.5764	1.5740
Aniline + 2-methyl-2-propanol						
0.0000	1.3879	1.3852	1.3825	1.3798	1.3770	1.3742
0.1082	1.4096	1.4069	1.4041	1.4013	1.3985	1.3957
0.2066	1.4292	1.4265	1.4238	1.4210	1.4182	1.4154
0.3075	1.4493	1.4466	1.4439	1.4412	1.4384	1.4356
0.4074	1.4691	1.4664	1.4637	1.4610	1.4582	1.4555
0.5064	1.4887	1.4860	1.4834	1.4807	1.4779	1.4752
0.6076	1.5088	1.5061	1.5035	1.5008	1.4981	1.4954
0.7028	1.5277	1.5251	1.5224	1.5197	1.5170	1.5144
0.8062	1.5482	1.5456	1.5430	1.5403	1.5377	1.5351
0.9012	1.5670	1.5644	1.5618	1.5592	1.5566	1.5541
1.0000	1.5864	1.5839	1.5814	1.5789	1.5764	1.5740

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Table 2. Coefficients, A_i of Equation (2) of Δn (10^{-2}) and standard deviations, σ (Δn) for aniline + alkanol mixtures at different temperatures.

T (K)	A_1	A_2	A_3	A_4	A_5	σ (Δn)
Aniline + 1-propanol						
293.15	0.5170	−0.1626	0.1021	0.1039	−0.2731	0.0011
298.15	0.4977	−0.1630	0.0893	0.0861	−0.3277	0.0014
303.15	0.4811	−0.1740	0.0140	0.0984	−0.2634	0.0021
308.15	0.4634	−0.1652	−0.0257	0.1200	−0.2394	0.0016
313.15	0.4481	−0.1542	−0.0723	0.1059	−0.2551	0.0013
318.15	0.4280	−0.1431	−0.0877	0.1083	−0.2257	0.0015
Aniline + 2-propanol						
293.15	0.5799	−0.1182	0.1327	−0.1022	−0.2327	0.0021
298.15	0.5575	−0.1051	0.0497	−0.0945	−0.1535	0.0025
303.15	0.5265	−0.1295	0.0355	−0.0225	−0.1586	0.0026
308.15	0.5019	−0.1407	0.0211	0.0102	−0.1832	0.0023
313.15	0.4801	−0.1460	−0.0656	0.0774	−0.0724	0.0016
318.15	0.4502	−0.1089	−0.0282	−0.0075	−0.1807	0.0009
Aniline + 2-methyl-1-propanol						
293.15	0.6981	0.0848	0.2112	−0.1513	−0.3573	0.0011
298.15	0.6795	0.0841	0.1391	−0.1520	−0.3260	0.0006
303.15	0.6531	0.0867	0.1106	−0.1528	−0.3051	0.0003
308.15	0.6242	0.0885	0.1183	−0.1655	−0.3653	0.0011
313.15	0.6028	0.0851	0.0540	−0.1291	−0.2960	0.0005
318.15	0.5826	0.0642	−0.0426	−0.0720	−0.1457	0.0004
Aniline + 2-methyl-2-propanol						
293.15	0.8462	0.0626	0.2316	−0.0681	−0.0864	0.0011
298.15	0.8115	0.0786	0.2613	−0.0973	−0.2438	0.0008
303.15	0.7788	0.1063	0.2065	−0.1424	−0.2773	0.0017
308.15	0.7439	0.1245	0.1222	−0.1594	−0.2815	0.0015
313.15	0.7090	0.1363	0.1199	−0.1719	−0.3776	0.0007
318.15	0.6770	0.1169	0.0619	−0.1259	−0.3410	0.0008

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where the quantity φ_i has been calculated, from the molar volume, V_m and partial molar volumes, $\overline{V}_{m,i}$ of the pure components, using the following relation:

$$\varphi_i = \frac{x_i V_{m,i}}{x_1 \overline{V}_{m,1} + x_2 \overline{V}_{m,2}}. \tag{5}$$

The $\overline{V}_{m,i}$ values of the components have been calculated from the density data of the mixtures [17].

Gladstone–Dale (G-D) relation

$$(n-1) = \varphi_1(n_1 - 1) + \varphi_2(n_2 - 1). \tag{6}$$

Arago–Biot (A-B) relation

$$n = \varphi_1 n_1 + \varphi_2 n_2. \tag{7}$$

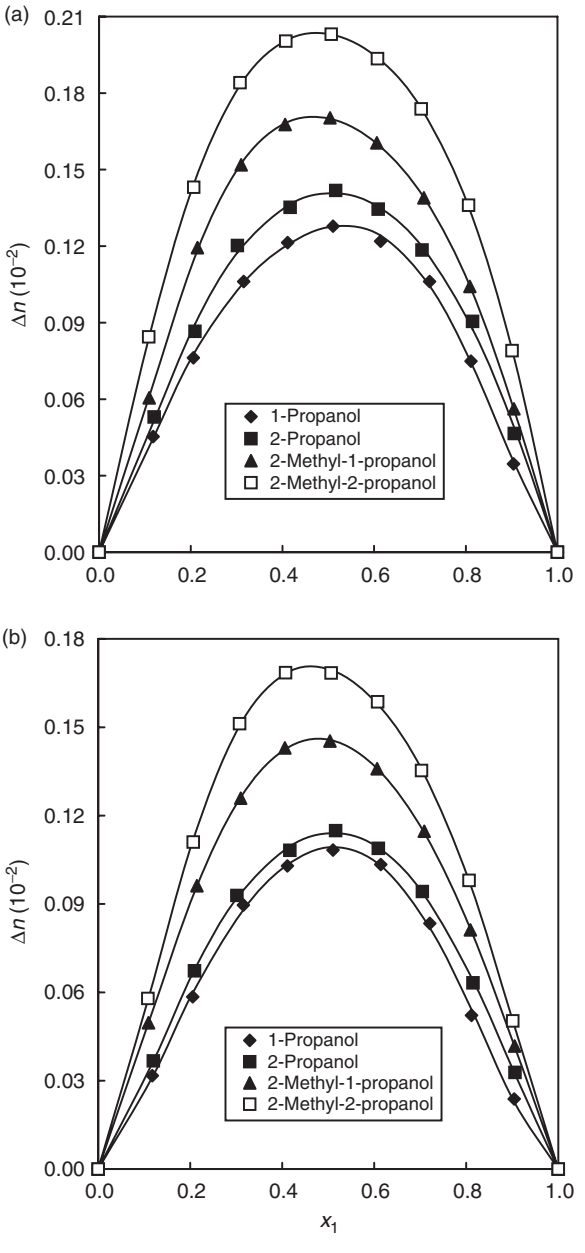

Figure. 1. Plots of deviations in refractive index, Δn *vs.* mole fraction, x_1 of aniline for the binary mixtures at (a) 298.15 K and (b) 318.15 K. The points show experimental values and curves show smoothed values using Equation (2).

Heller's (H) relation

$$\frac{(n-n_1)}{n_1} = \frac{3}{2}\left[\frac{(m^2-1)}{(m^2+2)}\right]\phi_2, \tag{8}$$

where $m = n_2/n_1$.

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Table 3. Average percentage deviations in theoretically calculated refractive indices for aniline + alkanol mixtures at different temperatures.

	Average percentage deviations						
T (K)	L–L Equation	G–D Equation	A–B Equation	H Equation	EYK Equation	OS Equation	W Equation
Aniline + 1-propanol							
293.15	0.364	−0.027	−0.210	0.441	6.444	6.118	−8.027
298.15	0.371	−0.025	−0.201	0.437	6.469	6.143	−7.929
303.15	0.380	−0.022	−0.190	0.433	6.494	6.169	−7.847
308.15	0.388	−0.019	−0.180	0.429	6.519	6.194	−7.752
313.15	0.395	−0.016	−0.170	0.425	6.543	6.219	−7.656
318.15	0.405	−0.013	−0.159	0.422	6.566	6.242	−7.588
Aniline + 2-propanol							
293.15	0.352	−0.045	−0.285	0.186	0.006	−0.501	−2.516
298.15	0.366	−0.041	−0.272	0.182	0.019	−0.487	−2.407
303.15	0.378	−0.037	−0.258	0.177	0.032	−0.475	−2.303
308.15	0.393	−0.032	−0.244	0.174	0.047	−0.460	−2.215
313.15	0.407	−0.028	−0.229	0.170	0.062	−0.446	−2.110
318.15	0.422	−0.023	−0.215	0.166	0.077	−0.431	−2.022
Aniline + 2-methyl-1-propanol							
293.15	0.268	−0.040	−0.301	0.192	−0.010	−0.414	−2.669
298.15	0.282	−0.036	−0.284	0.188	0.004	−0.400	−2.577
303.15	0.297	−0.031	−0.268	0.184	0.018	−0.387	−2.488
308.15	0.310	−0.027	−0.252	0.180	0.031	−0.375	−2.397
313.15	0.325	−0.022	−0.235	0.177	0.046	−0.360	−2.306
318.15	0.318	−0.025	−0.238	0.173	0.061	−0.346	−2.227
Aniline + 2-methyl-2-propanol							
293.15	0.263	−0.053	−0.379	0.219	−0.040	−0.480	−2.984
298.15	0.293	−0.044	−0.356	0.215	−0.012	−0.455	−2.853
303.15	0.320	−0.036	−0.334	0.210	0.014	−0.433	−2.708
308.15	0.346	−0.029	−0.312	0.204	0.038	−0.412	−2.547
313.15	0.375	−0.021	−0.291	0.200	0.064	−0.389	−2.416
318.15	0.404	−0.012	−0.269	0.196	0.092	−0.365	−2.298

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Eykman's (EYK) relation

$$\left[\frac{(n^2-1)}{(n+0.4)}\right]V_m = \left[\frac{(n_1^2-1)}{(n_1+0.4)}\right]x_1 V_{\mathrm{m},1} + \left[\frac{(n_2^2-1)}{(n_2+0.4)}\right](1-x_1)V_{\mathrm{m},2}. \tag{9}$$

Oster's (OS) relation

$$\left[\frac{(n^2-1)(2n^2+2)}{n^2}\right]V_{\mathrm{m}} = \left[\frac{(n_1^2-1)(2n_1^2+2)}{n^2}\right]V_{\mathrm{m},1} + \left[\frac{(n_2^2-1)(2n_2^2+2)}{n^2}\right]V_{\mathrm{m},2}. \tag{10}$$

Weiner (W) relation

$$\left[\frac{(n^2-n_1^2)}{(n^2+2n_1^2)}\right] = \phi_2\left[\frac{(n_2^2-n_1^2)}{(n_2^2+2n_1^2)}\right], \tag{11}$$

where V_m $(=x_1V_1+x_2V_2)$ is the molar volume. The results obtained from Equations (4)–(11) have been analysed in terms of average percentage deviation (APD) calculated using the relation

$$\mathrm{APD} = \frac{1}{m}\left[\sum \frac{(n_{\mathrm{Expt.}} - n_{\mathrm{Calcd}})}{n_{\mathrm{Expt.}}} \times 100\right]. \tag{12}$$

The APDs for all the four binary systems at the investigated temperatures are presented in Table 3. The results presented in Table 3 indicate that for aniline + 1-propanol mixtures, G-D and A-B relations provide best results with small APDs, followed by L-L and H relations, whereas OS, EYK and W relations could not predict the n values well and show relatively large deviations. In case of aniline + 2-propanol/2-methyl-1-propanol/2-methyl-2-propanol mixtures, G-D and EYK relations provide best results with small APDs, followed by H, A-B, L-L and OS relations; while W relation could not predict the n values well and show relatively large APD for all the binary systems. Also, for aniline + 1-propanol mixtures, the APD values from L-L, EYK and OS relations increase, whereas those from H, A-B, L-L and W relations decrease with a rise in temperature. For the remaining three systems, the APD values from L-L and EYK relations increase, whereas those from G-D, A-B, H, OS and W relations decrease with a rise in temperature.

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