

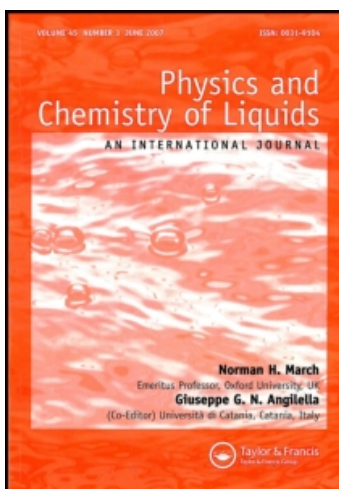
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### Density, viscosity and refractive index for ethyl *tert*-butyl ether + 2-butoxyethanol mixtures

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## Density, viscosity and refractive index for ethyl *tert*-butyl ether + 2-butoxyethanol mixtures

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Densities ( $\rho$ ) at 293.15, 298.15, 303.15, 308.15, and 313.15 K, viscosities ( $\eta$ ) at 293.15, 298.15, and 303.15 K and refractive indexes ( $n$ ) at 298.15 K of binary mixtures of ethyl *tert*-butyl ether (**1**) + 2-butoxyethanol (**2**), are reported. The excess molar volumes ( $V^E$ ) and the viscosities, and refractive index deviations ( $\Delta \ln \eta$  and  $\Delta n$ ) were calculated from these experimental data. The results are discussed in terms of intermolecular interactions.

**Keywords:** Density; Viscosity; Refractive index; 2-Butoxyethanol; Ethyl *tert*-butyl ether; Intermolecular interactions

### 1. Introduction

In continuation of our program on studies of the structural properties of some mixtures of alkoxyethanols with ethyl *tert*-butyl ether (ETBE) [1–3], the present article reports densities at 293.15, 298.15, 303.15, 308.15, and 313.15 K, viscosities at 293.15, 298.15, and 303.15 K, and refractive indexes at 298.15 K for ETBE + 2-butoxyethanol (BE) binary mixtures. From these results, the excess molar volumes ( $V^E$ ) and the deviations of the viscosity ( $\Delta \ln \eta$ ) from a mole fraction ( $x$ ) and refractive index ( $\Delta n$ ) from a volume fraction ( $\phi$ ) average have been calculated. These quantities have been fitted to the Redlich–Kister equation [4], to obtain the binary coefficients and standard errors, between the calculated and the experimental parameters.

Furthermore, the experimental results are used to disclose the nature of binary interactions in the bulk of studied the binary mixtures.

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Table 1. Reference density, viscosity and refractive index values of ethyl *tert*-butyl ether (ETBE) and 2-butoxyethanol (BE), at 298.15 K.

Solvent	$\rho$ (g cm <sup>-3</sup> )		$\eta$ (mPa s)		$n$	
	This work	Literature	This work	Literature	This work	Literature
ETBE	0.73559	0.73557 [10] 0.7353 [11]	0.504	–	1.37322	1.3731 [16]
BE	0.89629	0.89580 [12] 0.89581 [13] 0.89621 [14]	2.825	2.836 [15]	1.41753	1.4175 [17]

## 2. Experimental section

### 2.1. Materials

2-Butoxyethanol and ETBE, Merck, pro-analysis, containing <0.05% (w/w) of water, respectively (determined by Karl–Fischer method), were used.

Ethyl *tert*-butyl ether and BE were further purified by the methods described by Riddick [5]. The mixtures were prepared by mass, with weightings accuracy to  $\pm 1 \times 10^{-4}$  g. Conversion to molar quantities was based on the relative atomic mass table of 1985 issued by IUPAC in 1986. The uncertainty in the mole fractions is less than  $1 \times 10^{-4}$ . Liquids were stored in dry box over phosphoric pentoxide and degassed by ultrasound just before the experiment.

Experimental data of densities, viscosities and refractive indexes for the pure solvents, at 298.15 K, are compared with values available in the literature and listed in table 1.

### 2.2. Measurements

Solvent densities were measured with a bicapillary type Lipkin pycnometer, with a capacity of ca 90 cm<sup>3</sup>. The maximum error in the density measurements was  $3 \times 10^{-5}$  g cm<sup>-3</sup>.

The flow times of the mixtures and pure liquids were measured in a ViscoClock (made by Schott), equipped with an Ubbelohde capillary viscometer. The double distilled, deionized and degassed water with a specific conductance of  $1 \times 10^{-7}$   $\Omega^{-1}$  cm<sup>-1</sup> was used for the calibration. The time measurement tolerance was  $\pm 0.005\%$ , and the display accuracy was  $\pm 0.01$  s. The accuracy in the viscosity measurements was  $\pm 0.001$  mPa s.

The refractive indexes  $n_D$  (Na-D line, at  $\lambda = 589$  nm) were measured by an automatic refractometer DR 5000 Krüss, with a resolution  $1 \times 10^{-5}$  and an accuracy of  $2 \times 10^{-5}$ .

In all the physicochemical properties measurements, an Haake model DC-30 thermostat was used at a constant digital temperature control of  $\pm 0.01$  K.

## 3. Results and discussion

The experimental values of density ( $\rho$ ), viscosity ( $\eta$ ) and refractive index ( $n$ ) at all measured temperatures are given in tables 2 and 3. From the measured densities the

Table 2. Experimental density ( $\rho$ ) and excess molar volume ( $V^E$ ) for ethyl tert-butyl ether (1) + 2-butoxyethanol (2) binary mixtures.

$x_1$	$\rho$ (g cm <sup>-3</sup> )					$V^E$ (cm <sup>3</sup> mol <sup>-1</sup> )				
	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K
0.0000	0.90119	0.89629	0.89235	0.88818	0.88349	0.000	0.000	0.000	0.000	0.000
0.0499	0.89430	0.88909	0.88487	0.88043	0.87548	-0.218	-0.182	-0.150	-0.119	-0.088
0.1000	0.88702	0.88170	0.87732	0.87271	0.86760	-0.382	-0.338	-0.292	-0.244	-0.195
0.1511	0.87951	0.87413	0.86966	0.86494	0.85967	-0.534	-0.489	-0.438	-0.381	-0.317
0.2007	0.87220	0.86676	0.86223	0.85742	0.85207	-0.675	-0.628	-0.577	-0.515	-0.445
0.2489	0.86507	0.85960	0.85499	0.85014	0.84475	-0.805	-0.760	-0.706	-0.645	-0.575
0.2999	0.85749	0.85196	0.84731	0.84241	0.83700	-0.933	-0.887	-0.834	-0.772	-0.706
0.3421	0.85116	0.84559	0.84088	0.83597	0.83054	-1.027	-0.980	-0.925	-0.868	-0.804
0.4018	0.84209	0.83644	0.83167	0.82674	0.82131	-1.138	-1.085	-1.030	-0.977	-0.920
0.4500	0.83463	0.82893	0.82412	0.81916	0.81373	-1.200	-1.145	-1.090	-1.038	-0.988
0.5002	0.82671	0.82096	0.81612	0.81115	0.80872	-1.236	-1.179	-1.124	-1.076	-1.031
0.5521	0.81840	0.81261	0.80775	0.80274	0.79730	-1.247	-1.187	-1.135	-1.087	-1.044
0.5998	0.81066	0.80485	0.79994	0.79490	0.78943	-1.234	-1.175	-1.119	-1.071	-1.026
0.6538	0.80194	0.79599	0.79103	0.78594	0.78039	-1.219	-1.139	-1.081	-1.027	-0.973
0.6987	0.79446	0.78858	0.78356	0.77840	0.77281	-1.161	-1.096	-1.030	-0.969	-0.908
0.7501	0.78600	0.78007	0.77497	0.76974	0.76404	-1.104	-1.032	-0.957	-0.885	-0.808
0.8000	0.77775	0.77174	0.76657	0.76125	0.75550	-1.033	-0.950	-0.866	-0.780	-0.693
0.8492	0.76947	0.76343	0.75819	0.75283	0.74702	-0.930	-0.841	-0.747	-0.655	-0.559
0.8997	0.76068	0.75464	0.74941	0.74406	0.73830	-0.761	-0.672	-0.581	-0.492	-0.404
0.9493	0.75149	0.74558	0.74048	0.73528	0.72966	-0.484	-0.416	-0.349	-0.286	-0.224
1.0000	0.74111	0.73559	0.73085	0.72598	0.72069	0.000	0.000	0.000	0.000	0.000

excess values of molar volumes  $V^E$  of the mixtures, at the each investigated temperature, were fitted to the equation:

$$V^E = x_1 M_1 (\rho^{-1} - \rho_1^{-1}) + x_2 M_2 (\rho^{-1} - \rho_2^{-1}) \quad (1)$$

where  $M_1$  and  $M_2$  are the molar masses of the pure components and  $\rho_1$ ,  $\rho_2$ , and  $\rho$  are the densities of the pure species (1) and (2) and that of the mixtures at different temperatures, respectively.

The deviations of the viscosity from a mole fraction ( $x$ ) and refractive index from a volume fraction ( $\phi$ ) average were calculated from the equations:

$$\Delta \ln \eta = \ln \eta - (x_1 \ln \eta_1 + x_2 \ln \eta_2) \quad (2)$$

$$\Delta n = n - (\phi_1 n_1 + \phi_2 n_2) \quad (3)$$

where  $\eta_1$ ,  $\eta_2$ ,  $n_1$ ,  $n_2$ , and  $\eta$ , and  $n$  are the viscosities and refractive indexes of the ETBE, BE and the mixtures, respectively.

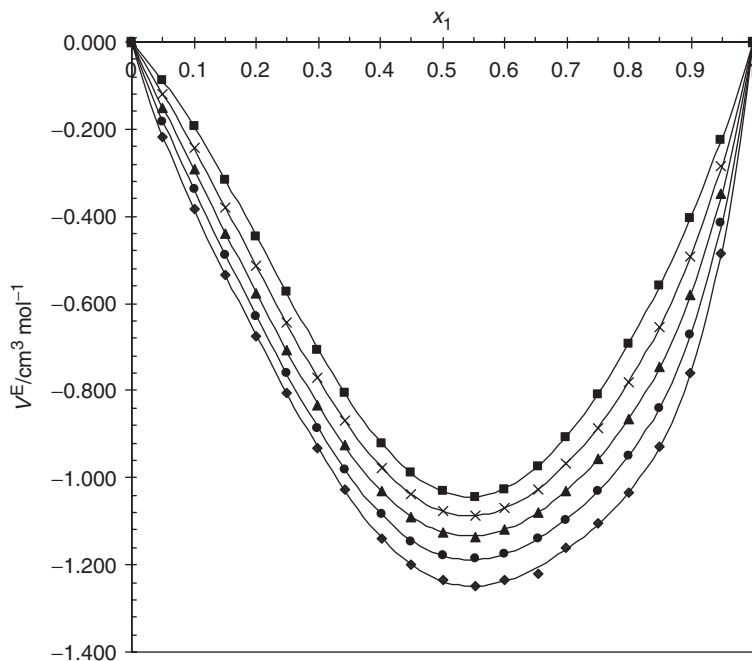
The values of  $V^E$ ,  $\Delta \ln \eta$  and  $\Delta n$  of the studied binary mixtures, at all measured temperatures, are given in tables 2 and 3.

The variations of  $V^E$  and  $\Delta \ln \eta$  versus the mole fraction of 2-methoxyethanol ( $x_1$ ), and  $\Delta n$  versus the volume fraction of 2-methoxyethanol ( $\phi_1$ ), at all measured temperatures, are presented in Figures 1–3, respectively. The curves have been obtained by fitting the  $V^E$ ,  $\Delta \ln \eta$  and  $\Delta n$  with Redlich–Kister equations of the type [4]:

$$V^E \text{ (cm}^3 \text{ mol}^{-1}) \quad \text{or} \quad \Delta \ln \eta = x_1(1 - x_1) \sum_{j=0}^4 a_j(2x_1 - 1)^j \quad (4)$$

Table 3. Experimental viscosity ( $\eta$ ), refractive index ( $n$ ) and deviations viscosity ( $\Delta \ln \eta$ ), and refractive index ( $\Delta n$ ) for ethyl *tert*-butyl ether (**1**) + 2-Butoxyethanol (**2**) binary mixtures.

$x_1$	$\eta$ (mPa s)			$n$	$\Delta \ln \eta$			$\Delta n$
	293.15 K	298.15 K	303.15 K	298.15 K	293.15 K	298.15 K	303.15 K	298.15 K
0.0000	3.239	2.825	2.449	1.41753	0.0000	0.0000	0.0000	0.00000
0.0499	2.901	2.558	2.239	1.41572	-0.0202	-0.0134	-0.0083	0.00051
0.1000	2.603	2.314	2.038	1.25808	-0.0381	-0.0270	-0.0205	0.00102
0.1511	2.336	2.092	1.850	1.41215	-0.0541	-0.0400	-0.0335	0.00153
0.2007	2.107	1.899	1.687	1.40968	-0.0676	-0.0512	-0.0448	0.00218
0.2489	1.910	1.731	1.545	1.40844	-0.0787	-0.0605	-0.0537	0.00246
0.2999	1.725	1.573	1.412	1.40653	-0.0883	-0.0686	-0.0608	0.00284
0.3421	1.589	1.455	1.312	1.40464	-0.0945	-0.0739	-0.0650	0.00312
0.4018	1.417	1.305	1.186	1.40244	-0.1007	-0.0794	-0.0689	0.00335
0.4500	1.296	1.198	1.094	1.40037	-0.1034	-0.0821	-0.0706	0.00347
0.5002	1.183	1.097	1.007	1.39832	-0.1040	-0.0834	-0.0715	0.00354
0.5521	1.079	1.004	0.925	1.39614	-0.1024	-0.0830	-0.0715	0.00353
0.5998	0.993	0.926	0.857	1.39390	-0.0991	-0.0811	-0.0704	0.00347
0.6538	0.906	0.847	0.786	1.39151	-0.0934	-0.0771	-0.0677	0.00335
0.6987	0.840	0.788	0.734	1.38920	-0.0870	-0.0720	-0.0638	0.00317
0.7501	0.773	0.727	0.679	1.38686	-0.0781	-0.0644	-0.0571	0.00295
0.8000	0.714	0.673	0.632	1.38442	-0.0677	-0.0549	-0.0480	0.00267
0.8492	0.661	0.626	0.590	1.38176	-0.0555	-0.0436	-0.0366	0.00225
0.8997	0.612	0.581	0.550	1.37921	-0.0406	-0.0303	-0.0232	0.00174
0.9493	0.570	0.541	0.514	1.37640	-0.0228	-0.0159	-0.0101	0.00104
1.0000	0.532	0.504	0.478	1.37322	0.0000	0.0000	0.0000	0.00000

Figure 1. Plot of excess molar volume ( $V^E$ ) as a function of composition for ETBE (**1**) + BE (**2**) binary liquid mixtures, at 293.15, 298.15, 303.15, 308.15, and 313.15 K.

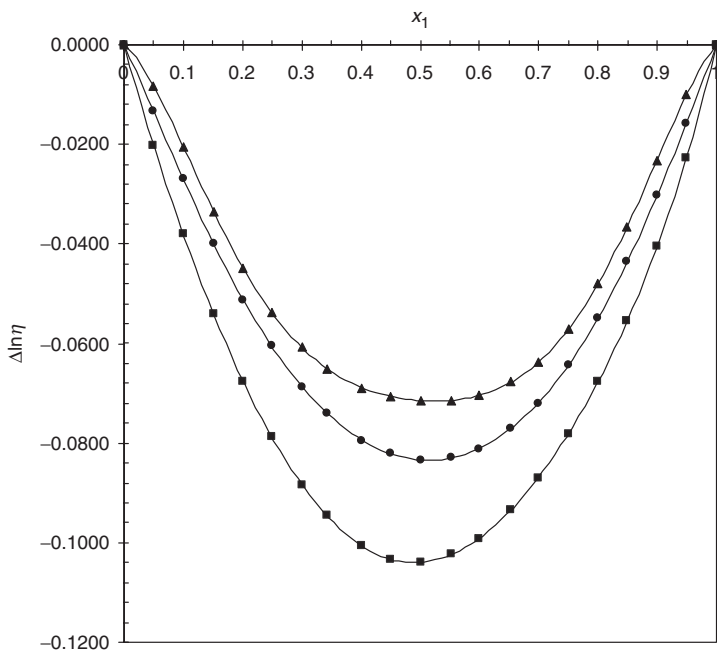


Figure 2. Plot of excess  $\Delta \ln \eta$  against mole fraction of ETBE for ETBE (1) + BE (2) binary liquid mixtures, at 293.15, 298.15, and 303.15 K.

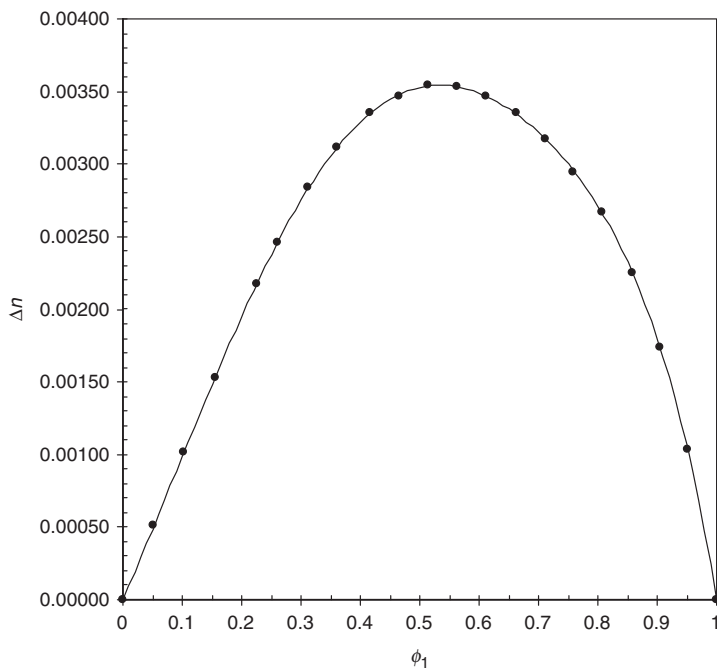


Figure 3. Plot of excess  $\Delta n$  against the volume fraction of ETBE for ETBE (1) + BE (2) binary liquid mixtures, at 298.15 K.

Table 4. Parameters  $a_j$  of equations (4) and (5) and SD  $\sigma(V^E)$ ,  $\sigma(\Delta \ln \eta)$  and  $\sigma(\Delta n)$  for ethyl *tert*-butyl ether (1) + 2-butoxyethanol (2) binary mixtures.

$T$	$a_0$	$a_1$	$a_2$	$a_3$	$a_4$	$\sigma(V^E)$ (cm <sup>3</sup> mol <sup>-1</sup> )
Ethyl <i>tert</i> -butyl ether (1) + 2-butoxyethanol (2)						
293.15 K	-4.9520	-0.9662	-4.0760	-2.5602	-4.0633	0.005
298.15 K	-4.7156	-0.8733	-0.4419	-2.2485	-2.8756	0.001
303.15 K	-4.4983	-0.8898	0.7016	-1.7280	-1.9541	0.001
308.15 K	-4.3059	-0.9725	1.2282	-1.1597	-1.3896	0.001
313.15 K	-4.1255	-1.1008	2.0056	-0.5394	-1.1546	0.001
						$\sigma(\Delta \ln \eta) \times 10^5$
293.15 K	-0.4160	0.0176	0.0026	-0.0534	-0.0547	3.7
298.15 K	-0.3336	-0.0182	-0.0152	-0.0064	0.0615	7.5
303.15 K	-0.2860	-0.0167	-0.1096	-0.0023	0.2779	2.6
						$\sigma(\Delta n) \times 10^6$
298.15 K	0.0141	0.0018	0.0001	0.0060	0.0032	7.3

$$\Delta n = \phi_1(1 - \phi_1) \sum_{j=0}^4 a_j(2\phi_1 - 1)^j \quad (5)$$

The values of these parameters, at each studied temperature, with SD  $\sigma(V^E)$ , are summarized in table 4.

Standard deviation values were obtained from:

$$\sigma = \left[ \frac{\sum (X_{\text{exptl}} - X_{\text{calcd}})^2}{n - p} \right]^{1/2} \quad (6)$$

where  $n$  is the number of experimental points,  $p$  is the number of parameters,  $X_{\text{exptl}}$  and  $X_{\text{calcd}}$  are the experimental and calculated properties.

Figures 1 and 2 shows that the excess of molar volumes ( $V^E$ ) and deviation of the viscosities ( $\Delta \ln \eta$ ) are negative over the whole composition range, and that they become less negative when temperature increases, with the minimums lying always nearly at  $x_1 \approx 0.50$ .

Figure 3 shows that the deviation of refractive indices ( $\Delta n$ ) are positive over the whole composition range, with the maximums lying nearly at  $\phi_1 \approx 0.51$  (corresponding to  $x_1 \approx 0.50$ ).

The course of changes of these structural parameters may be attributed mainly to the association through intermolecular hydrogen bonds between oxygen of the -O- group of the ETBE and hydrogen atom of the H-O- group of BE [1-3]. The addition of pure ETBE to BE would disrupt the self-associated structure in this alkoxyethanol causing the appearance in the solution of free BE molecules [6-9]. These free BE molecules may interact by dipole-dipole forces and/or intermolecular hydrogen bonds with ETBE molecules.

Obtained results seem to indicate that the stable intermolecular complexes of the ETBE-BE types are respectively formed in the studied binary mixtures.

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