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Dielectric properties of 2-methoxyethanol + isobutylamine, 2methoxyethanol + *sec*-butylamine, and 2-methoxyethanol + *tert*butylamine binary mixtures

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DIELECTRIC PROPERTIES OF 2-METHOXYETHANOL + ISOBUTYLAMINE, 2-METHOXYETHANOL + sec-BUTYLAMINE, AND 2-METHOXYETHANOL + tert-BUTYLAMINE BINARY MIXTURES

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Relative permittivities of numerous binary mixtures of 2-methoxyethanol (1) + isobutylamine (2), 2-methoxyethanol (1) + *sec*-butylamine (2), and 2-methoxyethanol (1) + *tert*-butylamine (2), between 291.15 and 313.15 K, are reported. These results are used to calculate deviations in the relative permittivities. The results are fitted to the Redlich–Kister polynomial equation to estimate the binary coefficients and standard errors. Furthermore, the experimental results are used to expose the nature of binary interactions in the bulk of the binary mixtures studied.

Keywords: 2-Methoxyethanol; Butylamines; Relative permittivity; Binary liquid mixtures

1. INTRODUCTION

Physicochemical and thermodynamic investigations play an important role in helping to understand the nature and the extent of the patterns of molecular aggregation that exist in liquid binary mixtures and their sensitivities to variations in composition and the molecular structure of the pure components [1-3].

As a part of our experimental program on the measurements of physicochemical properties and studies on internal structures of binary liquid mixtures, in which 2-methoxyethanol is one of the two constituents [4–8], we present here the relative permittivities for the 2-methoxyethanol+isobutylamine, 2-methoxyethanol+*sec*-butylamine, and 2-methoxyethanol+*tert*-butylamine, at different temperatures. We calculated the deviations in the relative permittivities, which were fitted to the Redlich–Kister equation [9].

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2. EXPERIMENTAL SECTION

Materials

2-Methoxyethanol (ME), isobutylamine (*i*-BA), *sec*-butylamine (*sec*-BA), and *tert*-butylamine (*tert*-BA) Merck, pro-analysis, containing less than 0.05% (w/w) of water, respectively (determined by Karl–Fischer method), were used.

2-Methoxyethanol and amines were further purified by the methods described by Riddick [10]. The mixtures were prepared by mass, with weighing accuracy equal 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 1×10^{-4} . Liquids were stored in dry-box over phosphoric pentoxide and degassed by ultrasound just before the experiment.

Measurements

The relative permittivity measurements were carried out at 3 MHz using a bridge of the type OH – 301 (made in Radelcis, Hungary). The thermostatic stainless steel measuring cell was of C3 ($1 < \varepsilon < 25$) type. The cell was calibrated with standard pure liquids, such as acetone, butan-1-ol, and dichloromethane. All these solvent were of spectrograde quality or higher. The relative permittivities for the standards were taken from the literature [11]. The accuracy in the relative permittivity measurements was ± 0.02 .

Each temperature was maintained with an accuracy of ± 0.01 K.

3. RESULTS AND DISCUSSION

The experimental data of relative permittivities (ε) obtained from the measurements for the pure solvents and the analyzed binary mixtures, at all the temperatures studied, are summarized in Tables I.

In our studies on intermolecular interactions in liquid mixtures of ME with butylamines, we have carried out the analysis of changes of deviations from ideality values of ε_{12} as a function of the composition of the mixed solvents. The study of this extra-thermodynamic structural parameter of liquid binary mixtures represents a unique tool for investigating the formation of intermolecular complexes, and provides a valuable aid for determining their stoichiometric composition [1–3].

The values of $\Delta \varepsilon$ have been evaluated by means of the equation [12–14]:

$$\Delta \varepsilon = \varepsilon - (\varepsilon_1 x_1 + \varepsilon_2 x_2) \tag{1}$$

where ε_1 , ε_2 , and ε are the relative permittivities of the ME, amine, and the mixtures, respectively.

The values of $\Delta \varepsilon$ of the analyzed binary mixtures, at 298.15 K, are shown graphically in Fig. 1. Deviations of relative permittivity were fitted by a Redlich-Kister type equation [9]:

$$\Delta \varepsilon = x_1 \cdot (1 - x_1) \sum_{j=0}^{k} a_j \cdot (2x_1 - 1)^j$$
(2)

2-Methoxyethanol + isobutylamine						$\frac{2-Methoxyethanol+sec-butylamine}{\varepsilon}$					
Ξ											
x_I	293.15 [K]	298.15 [K]	303.15 [K]	308.15 [K]	313.15 [K]	X_I	293.15 [K]	298.15 [K]	303.15 [K]	308.15 [K]	<i>313.15</i> [K]
0.0000	4.51	4.43	4.31	4.16	3.98	0.0000	4.82	4.51	4.38	4.22	4.09
0.0500	4.77	4.71	4.59	4.46	4.30	0.0509	5.10	4.85	4.78	4.69	4.61
0.1001	5.12	5.05	4.96	4.82	4.67	0.1001	5.43	5.21	5.17	5.11	5.06
0.1511	5.56	5.48	5.38	5.24	5.08	0.1489	5.80	5.60	5.58	5.52	5.47
0.2000	6.03	5.95	5.84	5.69	5.53	0.1999	6.25	6.04	6.00	5.94	5.89
0.2513	6.59	6.48	6.36	6.20	6.03	0.2506	6.73	6.50	6.45	6.36	6.30
0.2996	7.15	7.02	6.90	6.72	6.54	0.3131	7.36	7.11	7.02	6.90	6.81
0.3519	7.82	7.66	7.52	7.32	7.12	0.3511	7.77	7.50	7.40	7.25	7.14
0.4002	8.46	8.28	8.12	7.91	7.70	0.3999	8.31	8.03	7.88	7.71	7.59
0.4491	9.13	8.94	8.76	8.53	8.31	0.4509	8.91	8.60	8.44	8.24	8.09
0.4989	9.86	9.63	9.44	9.19	8.96	0.5000	9.50	9.18	8.99	8.78	8.62
0.5501	10.61	10.36	10.16	9.89	9.64	0.5489	10.10	9.78	9.57	9.35	9.18
0.6000	11.35	11.09	10.87	10.59	10.33	0.5999	10.76	10.44	10.21	9.99	9.80
0.6521	12.14	11.86	11.63	11.33	11.05	0.6502	11.44	11.11	10.89	10.66	10.46
0.6999	12.87	12.57	12.32	12.02	11.73	0.7000	12.13	11.82	11.59	11.36	11.16
0.7502	13.64	13.33	13.06	12.74	12.44	0.7492	12.86	12.55	12.32	12.09	11.89
0.8000	14.40	14.06	13.78	13.45	13.15	0.7995	13.65	13.33	13.10	12.87	12.65
0.8512	15.18	14.83	14.52	14.17	13.85	0.8493	14.46	14.14	13.91	13.66	13.43
0.9000	15.92	15.54	15.22	14.85	14.51	0.9000	15.36	15.03	14.77	14.50	14.24
0.9492	16.65	16.24	15.91	15.51	15.15	0.9499	16.31	15.95	15.66	15.33	15.03
1.0000	17.41	16.96	16.59	16.16	15.78	1.0000	17.41	16.96	16.59	16.16	15.78

TABLE I Experimental relative permittivity for 2-methoxyethanol (1)+isobutylamine (2), 2-methoxyethanol (1)+sec-butylamine (2), and 2-methoxyethanol (1)+tert-butylamine (2) binary mixtures

(continued)

TABLE 1 Continued									
2-Met	hoxyethanol + tert-	butylamine		2-Methoxyethanol + tert-butylamine					
E					ε				
291.15 [K]	293.15 [K]	298.15 [K]	303.15 [K]	x_I	291.15 [K]	293.15 [K]	298.15 [K]	303.15 [K]	
4.63	4.36	4.04	3.77	0.5510	6.33	6.15	5.91	5.67	
5.01	4.79	4.55	4.30	0.6009	6.50	6.31	6.08	5.82	
5.29	5.12	4.92	4.69	0.6491	6.64	6.44	6.24	5.98	
5.46	5.33	5.18	4.94	0.7004	6.75	6.59	6.39	6.14	
5.54	5.45	5.31	5.09	0.7495	6.95	6.79	6.59	6.38	
5.58	5.51	5.37	5.14	0.8003	7.34	7.20	6.99	6.83	
5.62	5.55	5.39	5.18	0.8500	8.13	8.03	7.77	7.65	
5.69	5.61	5.42	5.23	0.9007	9.79	9.66	9.38	9.24	
5.79	5.71	5.49	5.29	0.9498	12.68	12.49	12.17	11.93	
5.94	5.83	5.60	5.39	1.0000	17.58	17.35	16.96	16.55	
6.14	5.99	5.74	5.52						
	2-Met 291.15 [K] 4.63 5.01 5.29 5.46 5.54 5.58 5.62 5.69 5.79 5.94 6.14	2-Methoxyethanol + tert- ε 291.15 [K] 293.15 [K] 4.63 4.36 5.01 4.79 5.29 5.12 5.46 5.33 5.54 5.45 5.58 5.51 5.62 5.55 5.69 5.61 5.79 5.71 5.94 5.83 6.14 5.99	2-Methoxyethanol + tert-butylamine ε 291.15 [K] 293.15 [K] 298.15 [K] 4.63 4.36 4.04 5.01 4.79 4.55 5.29 5.12 4.92 5.46 5.33 5.18 5.54 5.45 5.31 5.58 5.51 5.37 5.62 5.55 5.39 5.69 5.61 5.42 5.79 5.71 5.49 5.94 5.83 5.60 6.14 5.99 5.74	2-Methoxyethanol + tert-butylamine \mathcal{E} 291.15 [K] 293.15 [K] 298.15 [K] 303.15 [K] 4.63 4.36 4.04 3.77 5.01 4.79 4.55 4.30 5.29 5.12 4.92 4.69 5.46 5.33 5.18 4.94 5.54 5.51 5.37 5.14 5.62 5.55 5.39 5.18 5.69 5.61 5.42 5.23 5.79 5.71 5.49 5.29 5.94 5.83 5.60 5.39 6.14 5.99 5.74 5.52	ε x _I 2-Methoxyethanol + tert-butylamine ε 291.15 [K] 293.15 [K] 298.15 [K] 303.15 [K] x _I 4.63 4.36 4.04 3.77 0.5510 5.01 4.79 4.55 4.30 0.6009 5.29 5.12 4.92 4.69 0.6491 5.46 5.33 5.18 4.94 0.7004 5.54 5.51 5.37 5.14 0.8003 5.62 5.55 5.39 5.18 0.8003 5.69 5.61 5.42 5.23 0.9007 5.79 5.71 5.49 5.29 0.9498 5.94 5.83 5.60 5.39 1.0000 6.14 5.99 5.74 5.52 1.0000	$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	2-Methoxyethanol + tert-butylamine 2-Methoxyethanol + tert-butylamine ε ε 291.15 [K] 293.15 [K] 298.15 [K] 303.15 [K] x_I 291.15 [K] 293.15 [K] 298.15 [K] 4.63 4.36 4.04 3.77 0.5510 6.33 6.15 5.91 5.01 4.79 4.55 4.30 0.6009 6.50 6.31 6.08 5.29 5.12 4.92 4.69 0.6491 6.64 6.44 6.24 5.46 5.33 5.18 4.94 0.7004 6.75 6.59 6.39 5.58 5.51 5.37 5.14 0.8003 7.34 7.20 6.99 5.62 5.55 5.39 5.18 0.8500 8.13 8.03 7.77 5.69 5.61 5.42 5.23 0.9007 9.79 9.66 9.38 5.79 5.71 5.49 5.29 0.9498 12.68 12.49 12.17 5.94	

TABLE I Continued



FIGURE 1 Plot of $\Delta \varepsilon$ as a function of composition for ME+*iso*-BA (\blacktriangle), ME+*sec*-BA (\blacksquare), and ME+*tert*-BA (\bigcirc), at 298.15 K.

The parameters a_j of Eq. (2) were evaluated by the least-squares method. The values of these parameters, at each studied temperature, with standard deviation σ , are summarized in Table II.

Standard deviation values were obtained from

$$\sigma = \left[\frac{\sum \left(X_{\text{exptl}} - X_{\text{calcd}}\right)^2}{n - p}\right]^{1/2}$$
(3)

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

As we can see, the deviations of this extra-thermodynamic parameter ($\Delta \varepsilon$) are always negative over the whole composition range for all studied mixtures. As a major feature, these plots show the minimums centered at *ca*. $x_1 \approx 0.35$ for ME+*i*-BA, $x_1 \approx 0.50$ for ME+*sec*-BA, and $x_1 \approx 0.80$ for ME+*tert*-BA.

As suggested by Tassi and other authors, the small negative $(\Delta \varepsilon)$ value for various solvent systems, may be attributed either to homoconjugation phenomena of the more polar component induced by the presence of an apolar or quasi-apolar one, and/or to associations between unlike molecules in the mixtures [2,12–14].

In the analyzed ME-butylamines mixtures, we have to vary polar components $(\mu_{ME}^{298,15K} = 2.04 \text{ D}, \mu_{i\text{-}BA}^{298,15K} = 1.27 \text{ D}, \mu_{sec-BA}^{298,15K} = 1.28 \text{ D}, \text{ and } \mu_{tert-BA}^{298,15K} = 1.29 \text{ D})$ [10], therefore it is likely that there are homoconjugations between similar molecules but more likely that there are weak dipolar associations between unlike molecules and/or appropriate negative contributions may be expected to originate in hydrogen bonding between ME and butylamine molecules and formation of the ME · 2*i*-BA, ME · sec-BA, and 4ME · tert-BA adducts in these binary solvent systems.

	-	a_0	a_1	<i>a</i> ₂	<i>a</i> ₃	<i>a</i> ₄	$\sigma(\Delta \varepsilon)$
2-N	lethorvet	thanol + isobutyle	amine		-		. ,
2 1.	T/K	inanoi isooutyn		293.15			
$\Delta \varepsilon$	1	-4.3685	3.2366	-0.7061	0.1249	-0.1987	0.0036
	T/K			298.15			
$\Delta \varepsilon$	T /IZ	-4.1998	3.1601	-0.2671	0.0077	-0.1328	0.0028
10	1/K	2 0782	2 0270	303.15	0.0656	0 1272	0.0024
$\Delta \varepsilon$	T/K	-3.9783	5.0279	-0.0033	0.0050	-0.1372	0.0034
$\Delta \varepsilon$	1/11	-3.8209	2.9271	0.4041	0.0712	-0.1056	0.0033
	T/K			313.15			
$\Delta \varepsilon$		-3.6404	2.8158	0.6465	-0.0187	0.0062	0.0029
2-N	lethoxye	thanol $\pm sec$ -buty	lamine				
2 10	T/K	inanor see outy		293.15			
$\Delta \varepsilon$,	-6.4871	-0.6453	-1.19773	-0.5231	-0.6969	0.0068
	T/K			298.15			
$\Delta \varepsilon$	TIZ	-6.2256	-0.7714	-0.7638	-0.3794	-0.3644	0.0042
٨٥	1/K	5 0028	1 1624	303.15	0.0275	0.2126	0.0020
Δc	T/K	-5.5928	-1.1024	308.15	-0.0275	-0.2120	0.0029
$\Delta \varepsilon$	-/	-5.6351	-1.2207	1.9297	0.1153	0.2425	0.0033
	T/K			313.15			
$\Delta \varepsilon$		-5.2747	-1.3578	3.0145	0.2932	0.4181	0.0049
2-M	lethoxve	thanol + <i>tert</i> -buty	lamine				
	T/K			291.15			
$\Delta \varepsilon$		-19.8925	-18.0585	-18.6008	-37.7657	-22.2135	0.0040
	T/K	10,40,40	10 4500	293.15	22.12.55	10 4020	0.0000
$\Delta \varepsilon$	T/V	-19.4849	-19.4530	-17.5731	-33.4257	-19.4030	0.0033
Λc	1/K	_19.0189	-19 4514	_15.0115	_33 8398	_21 2953	0 0090
Δc	T/K	-17.0109	-17.4214	303.15	55.0570	-21.2755	0.0090
$\Delta \varepsilon$, –	-18.5602	-19.7493	-14.8537	-31.0194	-18.6207	0.0077

TABLE II Parameters a_j of Eq. (2), and standard deviations $\sigma(\Delta \varepsilon)$ for 2-methoxyethanol + isobutylamine, 2-methoxyethanol + *sec*-butylamine, and 2-methoxyethanol + *tert*-butylamine binary mixtures

On the basis of comparison of $\Delta \varepsilon$ values in studied liquid mixtures in the composition regions corresponding to their maximum deviation from ideality, it is possible to deduced that:

$$\Delta \varepsilon_{\mathrm{ME}-i-\mathrm{BA}} \approx \Delta \varepsilon_{\mathrm{ME}-sec-\mathrm{BA}} < \Delta \varepsilon_{\mathrm{ME}-tert-\mathrm{AB}}$$

Therefore, it is necessary to assume that the energetic stability of intermolecular complexes ME \cdot 2*i*-BA, ME \cdot sec-BA, and 4ME \cdot tert-BA changes in the identical way.

From the experimental values of relative permittivities (ε_{12}) (see Table I), the temperature coefficients of the relative permittivity, denoted α_{12} , viz.:

$$\alpha_{12} = \frac{1}{\varepsilon} \cdot \left[\frac{d\varepsilon}{d(1/T)} \right] \tag{4}$$

at 298.15 K, were calculated (see Fig. 2 and Table III).



FIGURE 2 Plot of α_{12} as a function of composition for ME+*iso*-BA (\blacktriangle), ME+*sec*-BA (\blacksquare), and ME+*tert*-BA (\bigcirc), at 298.15 K.

$\overline{x_I}$	α ₁₂ [K]						
	ME+i-BA	ME+sec-BA	ME+tert-BA				
0.0000	477.80	784.15	1496.42				
0.0500	402.41	490.92	1088.22				
0.1000	353.23	351.10	854.15				
0.1510	348.80	289.11	699.32				
0.2000	342.25	292.92	600.01				
0.2510	359.53	325.28	585.74				
0.3000	362.87	376.76	601.22				
0.3520	387.39	403.22	652.17				
0.4000	395.83	442.37	691.35				
0.4490	401.00	459.66	741.17				
0.4990	414.09	466.28	777.15				
0.5500	412.91	457.89	790.41				
0.6000	408.57	442.71	771.18				
0.6520	406.49	419.04	732.21				
0.7000	403.88	390.29	670.77				
0.7500	403.89	367.32	609.66				
0.8000	403.72	349.77	523.72				
0.8510	408.41	337.09	464.54				
0.9000	412.04	342.39	435.20				
0.9490	418.92	366.99	429.11				
1.0000	441.01	441.01	441.01				

TABLE III Values of temperature coefficients of the relative permittivity for 2-methoxyethanol + isobutyloamine, 2-methoxyethanol + *sec*-butylamine, and 2-methoxyethanol + *tert*-butylamine binary mixtures at 298.15 K

The composition range of liquid binary mixtures within which α_{12} or its excess attain their highest values should be interpreted (as shown in Räetzsch *et al.*'s thermodynamic considerations [15]) as a region characterized by maximal intermolecular interactions between two different components of the given binary liquid mixture.

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For all studied mixtures, the α_{12} vs x_1 curves are W-shaped, being negative at the ends and positive at x_1 from about 0.20 to 0.85. In the case of all studied mixtures, we observed the tendency to achieve the maximum by the function α_{12} at $ca. x_1 \approx 0.50$. This effect can be accounted for by maximal intermolecular interactions between ME and *i*-BA, sec-BA, and tert-BA, which lead to the formation of stable ME · *i*-BA, ME · sec-BA, and ME · tert-BA intermolecular complexes [1–3,12–14].

By comparing values of α_{12} in studied liquid mixtures, it is possible to conclude that:

$$\alpha_{12(\text{ME}+i\text{-BA})}^{\text{max}} \approx \alpha_{12(\text{ME}+sec\text{-BA})}^{\text{max}} < \alpha_{12(\text{ME}+tert\text{-BA})}^{\text{max}}$$

Thus also in this case, the intermolecular complexes formed by ME and *tert*-BA should be assumed as energetically the most stable.

The conclusions to be drawn from the presented results are that:

- The molecules of studied binary mixtures may be joined, by a network of interactions such as dipolar and/or hydrogen bonds, to form stable intermolecular complexes.
- The probable compositions of the intermolecular complexes are in ME + *i*-BA = 1 : 1 and 1 : 2; in ME + *sec*-BA = 1 : 1; and in ME + *tert*-BA = 1 : 1 and 4 : 1 mole reactions, over the measured temperature range.
- Most likely, complexes of ME with tert-BA are energetically the most stables.

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