

Density and Relative Permittivity of 2-Methoxyethanol + Dipropylamine Mixtures at Various Temperatures

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Densities (ρ) and relative permittivities (ϵ) at five temperatures, between (291.15 and 303.15) K, of binary mixtures of 2-methoxyethanol (1) + dipropylamine (2), are reported. The excess molar volumes (V^E) and the relative permittivity deviations ($\Delta\epsilon$) were calculated from these experimental data. The results are discussed in terms of intermolecular interactions.

Introduction

The nature and type of intermolecular interactions in binary liquid mixtures containing alkoxyalcohols have been studied by us earlier in terms of the mixing parameters such as excess molar volume, excess molar refractivity, deviations in relative permittivity, deviations in viscosity and deviations in isentropic compressibility.^{1–4} Alkoxyalcohols and amines are the versatile organic liquids used in chemical industries as the solvents for oils, petrol, lacquers, and as an antifreeze for explosives. The literature review shows that the mixing properties of 2-methoxyethanol (ME) + dipropylamine (DPA) binary mixtures have not been measured. It was therefore decided to measure the density and the relative permittivity in these mixtures.

From these results, the excess molar volume V^E and deviation of relative permittivity ($\Delta\epsilon$) (at 291.15 K, 293.15 K, 298.15 K, 303.15 K, and 308.15 K) have been calculated. Furthermore, the experimental results are used to disclose the nature of intermolecular interactions in the bulk of the studied binary mixtures.

Experimental Section

Materials. ME and DPA, Merck pro-analysis, containing less than 0.05% (w/w) of water, respectively (determined by Karl Fischer method), were used.

ME and DPA were purified by the methods described by Riddick.⁸ The mixtures were prepared by mass, with weighings accurate 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 a drybox over phosphoric pentoxide and degassed by ultrasound just before the experiment.

Experimental data of densities and relative permittivities for the pure solvents, at 298.15 K, are compared with values available in the literature and listed in Table 1.

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 the C3 ($1 < \epsilon < 25$) type. The cell was calibrated with standard pure liquids, such

Table 1. Reference Density and Relative Permittivity Values of ME and DPA, at 298.15 K

solvent	$\rho/\text{g}\cdot\text{cm}^{-3}$		ϵ	
	this work	lit	this work	lit
ME	0.96029	0.960288 ¹² 0.9601 ¹⁴	16.96	16.94 ¹³
DPA	0.73256	0.73326 ¹⁵ 0.7329 ⁵	3.31 (at 293.15K)	3.068 ⁵ (at 293.15K)

as acetone, butan-1-ol, and dichloromethane. All these solvent were of spectrograde quality or higher. The relative permittivity for the standards were taken from the literature.^{5,6} Solvent densities were measured with a bicapillary-type Lipkin pycnometer with a capacity of ca. 90 cm³. The density and relative permittivity were measured with a precision of $\pm 1 \times 10^{-6}$ g·cm⁻³ and ± 0.01 . In all the physicochemical property measurements, an Haake model DC-30 thermostat was used. For temperatures from $T = 291.15$ to 308.15 K, the thermostat ensures a thermal stability of ± 0.01 K.

Results and Discussion

The experimental values of density (ρ) and relative permittivity (ϵ) at (291.15, 293.15, 298.15, 303.15, and 308.15) K are given in Tables 2 and 3.

In many articles, the dependence of density on temperature ($t/^\circ\text{C}$) is started on the basis of a high polynomial of the type¹²

$$\rho(t) = \sum_0^3 a_k t^k \quad (1)$$

where the a_k values are the adjustable parameters summarized in Table 4 along with the standard error of determination $\sigma(\rho)$.

Equation 1 reproduces the experimental density values with an average uncertainty, evaluated by means of the relation

$$\overline{\Delta\rho} = \frac{\sum |\rho_{\text{cal}} - \rho_{\text{exp}}|}{N} \quad (2)$$

where N is the number of experimental points.

The values of $\overline{\Delta\rho} = \pm 0.00007$ g·cm⁻³ for ME + DPA binary liquid mixtures.

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Table 2. Experimental Density (ρ) and Excesses Molar Volume (V^E) for ME (1) + DPA (2) Binary Mixtures

x_1	$\rho/\text{g}\cdot\text{cm}^{-3}$					$V^E/\text{cm}^3\cdot\text{mol}^{-1}$				
	291.15K	293.15K	298.15K	303.15K	308.15K	291.15K	293.15K	298.15K	303.15K	308.15K
0.0000	0.73847	0.73720	0.73256	0.72820	0.72364	0.000	0.000	0.000	0.000	0.000
0.0501	0.74683	0.74526	0.74037	0.73586	0.73107	-0.298	-0.246	-0.206	-0.179	-0.144
0.1000	0.75498	0.75342	0.74852	0.74391	0.73904	-0.491	-0.446	-0.409	-0.374	-0.326
0.1502	0.76340	0.76190	0.75690	0.75227	0.74736	-0.657	-0.625	-0.577	-0.543	-0.496
0.2001	0.77206	0.77051	0.76550	0.76085	0.75594	-0.801	-0.765	-0.720	-0.688	-0.647
0.2508	0.78114	0.77961	0.77459	0.76992	0.76498	-0.923	-0.894	-0.851	-0.821	-0.779
0.2999	0.79026	0.78866	0.78368	0.77897	0.77407	-1.021	-0.985	-0.953	-0.922	-0.891
0.3502	0.79992	0.79835	0.79328	0.78857	0.78365	-1.098	-1.071	-1.029	-1.001	-0.971
0.4000	0.80992	0.80821	0.80320	0.79845	0.79351	-1.164	-1.121	-1.091	-1.061	-1.032
0.4500	0.82034	0.81867	0.81361	0.80882	0.80385	-1.208	-1.175	-1.141	-1.109	-1.079
0.4999	0.83106	0.82940	0.82430	0.81953	0.81450	-1.220	-1.191	-1.154	-1.129	-1.094
0.5507	0.84247	0.84075	0.83560	0.83080	0.82578	-1.216	-1.184	-1.142	-1.115	-1.085
0.5982	0.85345	0.85169	0.84660	0.84177	0.83669	-1.178	-1.144	-1.111	-1.083	-1.047
0.6502	0.86602	0.86426	0.85911	0.85424	0.84917	-1.119	-1.089	-1.050	-1.020	-0.987
0.6999	0.87841	0.87659	0.87147	0.86658	0.86150	-1.024	-0.991	-0.956	-0.925	-0.891
0.7489	0.89121	0.88934	0.88421	0.87930	0.87418	-0.915	-0.880	-0.843	-0.811	-0.774
0.8015	0.90566	0.90370	0.89854	0.89364	0.88851	-0.781	-0.741	-0.701	-0.671	-0.633
0.8496	0.91922	0.91732	0.91215	0.90723	0.90213	-0.611	-0.580	-0.538	-0.506	-0.471
0.8997	0.93442	0.93235	0.92721	0.92230	0.91711	-0.449	-0.406	-0.365	-0.334	-0.291
0.9497	0.95019	0.94822	0.94310	0.93830	0.93324	-0.251	-0.221	-0.180	-0.158	-0.125
1.0000	0.96652	0.96488	0.96029	0.95576	0.95110	0.000	0.000	0.000	0.000	0.000

Table 3. Experimental Relative Permittivity (ϵ) and Deviations Relative Permittivity ($\Delta\epsilon$) for ME (1) + DPA (2) Binary Mixtures

x_1	ϵ					$\Delta\epsilon$				
	291.15K	293.15K	298.15K	303.15K	308.15K	291.15K	293.15K	298.15K	303.15K	308.15K
0.0000	3.34	3.31	3.24	3.18	3.11	0.00	0.00	0.00	0.00	0.00
0.0501	3.30	3.28	3.26	3.23	3.20	-0.75	-0.73	-0.67	-0.62	-0.56
0.1000	3.39	3.37	3.33	3.29	3.26	-1.37	-1.34	-1.28	-1.23	-1.16
0.1502	3.63	3.58	3.51	3.44	3.38	-1.85	-1.84	-1.79	-1.75	-1.69
0.2001	3.92	3.87	3.81	3.74	3.66	-2.27	-2.25	-2.18	-2.12	-2.06
0.2508	4.25	4.20	4.13	4.05	3.94	-2.66	-2.63	-2.55	-2.48	-2.44
0.2999	4.67	4.61	4.48	4.40	4.32	-2.94	-2.91	-2.87	-2.79	-2.70
0.3502	5.14	5.07	4.92	4.82	4.73	-3.19	-3.16	-3.12	-3.04	-2.95
0.4000	5.68	5.61	5.46	5.33	5.21	-3.36	-3.32	-3.27	-3.20	-3.12
0.4500	6.28	6.20	6.02	5.89	5.75	-3.47	-3.43	-3.39	-3.31	-3.23
0.4999	6.93	6.84	6.67	6.51	6.37	-3.53	-3.49	-3.43	-3.35	-3.26
0.5507	7.67	7.57	7.39	7.21	7.06	-3.51	-3.47	-3.41	-3.33	-3.24
0.5982	8.42	8.30	8.10	7.91	7.73	-3.44	-3.41	-3.35	-3.27	-3.19
0.6502	9.32	9.20	8.97	8.74	8.56	-3.28	-3.24	-3.19	-3.13	-3.04
0.6999	10.27	10.12	9.88	9.65	9.42	-3.04	-3.02	-2.96	-2.89	-2.82
0.7489	11.27	11.14	10.87	10.63	10.40	-2.73	-2.68	-2.64	-2.56	-2.48
0.8015	12.44	12.27	12.02	11.76	11.51	-2.31	-2.29	-2.22	-2.14	-2.06
0.8496	13.58	13.42	13.15	12.87	12.61	-1.86	-1.82	-1.75	-1.67	-1.59
0.8997	14.87	14.69	14.36	14.06	13.78	-1.28	-1.25	-1.22	-1.15	-1.07
0.9497	16.21	16.00	15.66	15.34	15.04	-0.65	-0.64	-0.61	-0.54	-0.46
1.0000	17.58	17.35	16.96	16.55	16.16	0.00	0.00	0.00	0.00	0.00

The variation of the density with binary composition was studied by using the following equation¹²

$$\rho(x_1) = \sum_0^6 b_j x_1^j \quad (3)$$

which could be fitted to the experimental data at each temperature using a least-squares method.¹² The values of b_j coefficients and the standard deviations $\sigma(\rho)$ are summarized in Table 5. The goodness-of-fit of this procedure is ascertained by a mean deviation $\overline{\Delta\rho} = \pm 0.00003 \text{ g}\cdot\text{cm}^{-3}$ for ME + DPM binary liquid mixtures.

In the same way, we investigated the dependencies of the relative permittivity and refractive index of the analyzed binary mixtures.

Many investigations of the dielectric behavior of liquids have been reported in the literature, and the best representation of the results as a function of temperature is given by the eq 4¹³

$$\ln \epsilon = \alpha_0 + \alpha_1 T \quad (4)$$

where α_i ($i = 0$ and 1) are empirical constants and T is the absolute temperature.

Table 4. Coefficients a_k and Standard Deviations $\sigma(\rho)$ of Eq 1 for ME + DPA Binary Mixtures

x_1	$10a_0$	10^4a_1	10^5a_2	10^7a_3	$10^5\sigma(\rho)$
0.0000	7.42428	4.99493	-5.03573	5.93307	19.02
0.0501	7.60478	-5.72994	-1.26230	1.42915	15.84
0.1000	7.66773	-3.55609	-2.05964	2.32320	14.92
0.1502	7.73626	-1.57116	-2.84977	3.30342	18.46
0.2001	7.83426	-2.82083	-2.42202	2.81632	17.12
0.2508	7.91760	-1.96425	-2.72758	3.15511	17.63
0.2999	8.01458	-2.56840	-2.54500	2.98384	14.05
0.3502	8.10746	-1.97452	-2.82508	3.36798	16.91
0.4000	8.24263	-6.11405	-1.27733	1.47090	11.54
0.4500	8.32929	-3.97450	-2.10306	2.47423	13.16
0.4999	8.44747	-5.29671	-1.58184	1.79248	14.94
0.5507	8.57393	-6.52557	-1.21826	1.43800	13.93
0.5982	8.69415	-7.92324	-0.61591	0.59803	11.42
0.6502	8.81062	-6.63439	-1.18408	1.36364	12.04
0.6999	8.94887	-8.33334	-0.54483	0.57480	9.52
0.7489	9.09329	-10.23710	0.16216	-0.30237	8.37
0.8015	9.27188	-14.11650	1.56080	-1.95493	7.06
0.8496	9.37956	-10.79480	0.29759	-0.39186	8.23
0.8997	9.59922	-18.82250	3.34271	-4.16950	4.05
0.9497	9.73582	-16.38990	2.43317	-2.97979	7.33
1.0000	9.79128	-4.93383	-1.41905	1.54662	6.29

The α_i coefficients of this fitting procedure are listed in Table 6, along with the standard deviations $\sigma(\ln \epsilon)$ for each of the binary mixtures.

Table 5. Coefficients b_j and Standard Deviations $\sigma(\rho)$ of Eq 3 for ME + DPA Binary Mixtures, at Different Temperatures

T/K	$10b_0$	$10b_1$	$10b_2$	$10b_3$	$10b_4$	$10b_5$	$10b_6$	$10^5\sigma(\rho)$
291.15	7.38477	1.68074	-6.08985	4.08458	-6.67308	5.47223	-1.67463	5.94
293.15	7.37182	1.61355	-0.01499	1.75679	-2.29492	1.54117	0.32426	4.68
298.15	7.32547	1.54225	0.54479	-0.34796	1.63382	-1.99904	0.90355	2.82
303.15	7.28190	1.49813	0.82923	-1.33382	3.39920	-3.56965	1.45255	3.01
308.15	7.23639	1.42466	1.39479	-3.37797	7.07383	-6.78747	2.54647	4.06

Table 6. Coefficients α_0 and α_1 and Standard Deviations $\sigma(\epsilon)$ of Eq 4 for ME + DPA Binary Mixtures

x_1	α_0	α_1	$10^3\sigma(\ln \epsilon)$
0.0000	2.413086	-0.004148	0.92
0.0501	1.698282	-0.001735	1.25
0.1000	1.892271	-0.002310	1.21
0.1502	2.481016	-0.004105	2.79
0.2001	2.490572	-0.003869	2.36
0.2508	2.687467	-0.004263	3.19
0.2999	2.861690	-0.004549	4.51
0.3502	3.052252	-0.004874	4.65
0.4000	3.209395	-0.005064	1.99
0.4500	3.331660	-0.005141	2.81
0.4999	3.367250	-0.004924	2.05
0.5507	3.445577	-0.004844	2.05
0.5982	3.567476	-0.004944	2.18
0.6502	3.686954	-0.005004	2.26
0.6999	3.774143	-0.004972	2.14
0.7489	3.787340	-0.004695	1.55
0.8015	3.817774	-0.004463	2.04
0.8496	3.857726	-0.004296	1.42
0.8997	3.985926	-0.004427	1.94
0.9497	4.042652	-0.004327	2.20
1.0000	4.285505	-0.004879	1.42

Equation 4 reproduces the experimental relative permittivity values with an average uncertainty, evaluated by means of the relation

$$\overline{\Delta\epsilon} = \frac{\sum |\epsilon_{\text{cal}} - \epsilon_{\text{exp}}|}{N} \quad (5)$$

where N is the number of experimental points.

The Values of $\overline{\Delta\epsilon} = \pm 0.01$ Units for ME + DPM Binary Liquid Mixtures. The variation of relative permittivity with binary composition was studied by using the following equation¹³

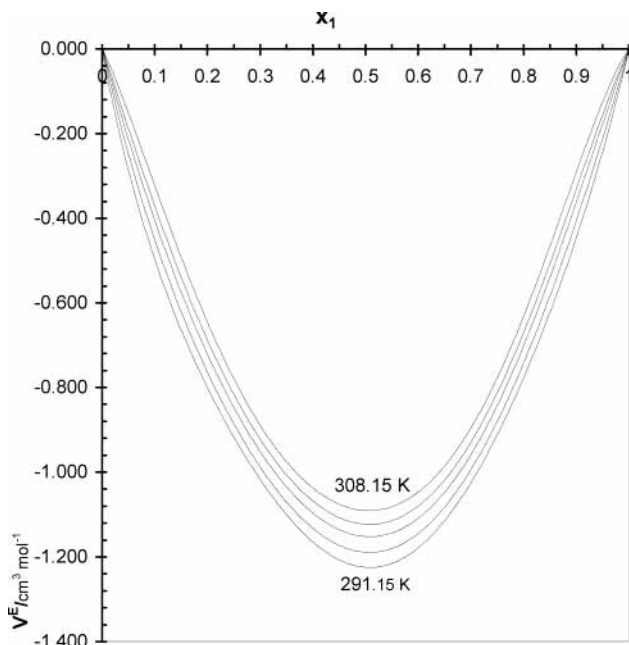
$$\ln \epsilon = \sum_{j=0}^5 \beta_j x_1^j \quad (6)$$

which could be fitted to the experimental data at each temperature using a least-squares method.¹³ The values of β_j coefficients and the standard deviations $\sigma(\ln \epsilon)$ are summarized in Table 7. The goodness-of-fit of this procedure is ascertained by a mean deviation $\overline{\Delta\epsilon} = \pm 0.02$ units for ME + DPM binary mixtures.

From the measured densities, the excess values of molar volumes V^E of the mixtures, at the each investigated temperature, were fitted to the equation⁷

$$V^E = x_1 M_1 (\rho^{-1} - \rho_1^{-1}) + x_2 M_2 (\rho^{-1} - \rho_2^{-1}) \quad (7)$$

where M_1 and M_2 are the molar masses of the pure components and ρ_1 , ρ_2 , and ρ are the densities of the pure

**Figure 1.** Isothermal best-fit curves of the excess molar volume for ME + DPA mixtures calculated by eq 9, at various temperatures.

species (1 and 2) and that of the mixtures at different temperatures, respectively.

The deviations of the relative permittivity from a mole fraction (x) were calculated from the equation^{7,8}

$$\Delta\epsilon = \epsilon - (x_1\epsilon_1 + x_2\epsilon_2) \quad (8)$$

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

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

The variations of V^E vs the mole fraction of ME (x_1), at all measured temperatures, are presented in Figure 1, respectively. The curves have been obtained by fitting the V^E with a Redlich-Kister equation of the type⁹

$$V^E/\text{cm}^3 \cdot \text{mol}^{-1} = x_1(1 - x_1) \sum_{j=0}^4 a_j(2x_1 - 1)^j \quad (9)$$

Figure 1 shows that the excesses of molar volumes (V_m^E) are negative over the whole composition range and that they become less negative when temperature increases, with the minimums lying always nearly $x_1 \approx 0.50$.

Two effects possibly influence the volume concentrations. The negative values of V^E over the whole mole fraction may

Table 7. Coefficients β_j and Standard Deviations $\sigma(\epsilon)$ of Eq 6 for ME + DPA Binary Mixtures, at Different Temperatures

T/K	β_0	β_1	β_2	β_3	β_4	β_5	$10^3\sigma(\ln \epsilon)$
291.15	1.202042	-0.500135	9.119721	-15.952448	13.412235	-4.416277	3.41
293.15	1.194748	-0.515824	9.056763	-15.695993	13.121801	-4.309252	2.23
298.15	1.176462	-0.335171	7.572965	-11.990494	9.372959	-2.966007	3.40
303.15	1.160867	-0.299750	7.267827	-11.435502	9.039081	-2.925353	4.27
308.15	1.143009	-0.206099	6.496145	-9.514874	7.100082	-2.232805	5.95

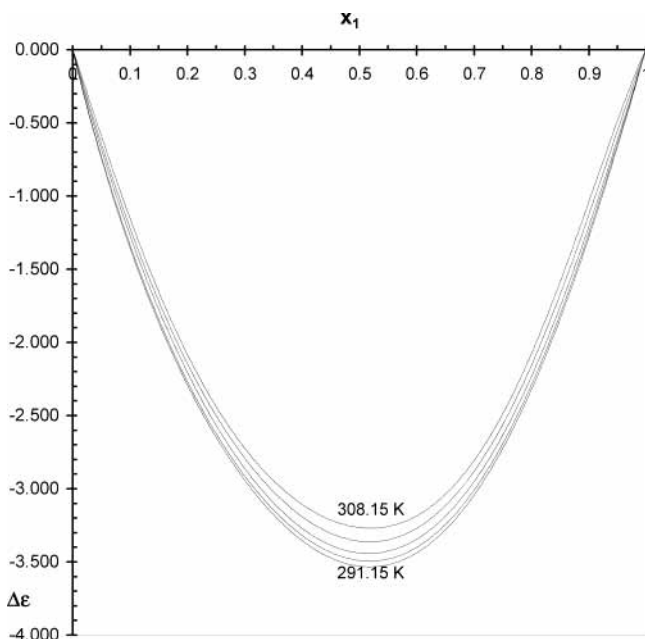


Figure 2. Isothermal best-fit curves of the deviations relative permittivity for ME + DPA mixtures calculated by eq 10, at various temperatures.

be attributed mainly to the association through *intermolecular hydrogen bonds* between the $-OH$ groups in ME and the nitrogen atom in DPA. Another effect, which would give a negative contribution to the excess volumes, is the difference in molecular sizes between the two components in the binary mixtures (this is a geometrical effect). As these differences increase, the more negative would be the contribution to V^E . In the studied mixtures, the ME and DPA molecules have different molar volumes as pure species

$$V(\text{DPA}) = 138.137 \text{ cm}^3 \cdot \text{mol}^{-1} < V(\text{ME}) = 79.242 \text{ cm}^3 \cdot \text{mol}^{-1}$$

Therefore, this effect should be significant and of great importance. The difference in the free volumes between ME and DPA would facilitate the penetration of one component (ME) into the other (DPA).

After these considerations, we may assume that in the ME + DPA binary liquid mixtures all the effects discussed above should be taken into account and may contribute to V^E values.^{6,10–12}

The variation of $\Delta\epsilon$ vs the mole fraction of ME (x_1), at all measured temperatures, is presented in Figure 2, respectively. The curves have been obtained by fitting the $\Delta\epsilon$ with a Redlich–Kister equation of the type⁹

$$\Delta\epsilon = x_1(1 - x_1) \sum_{j=0}^4 a_j(2x_1 - 1)^j \quad (10)$$

Figure 2 shows that the relative permittivity deviations are always negative for the ME + DPA binary system and that they become less negative when temperature increases, with a minimum lying always nearly $x_1 \approx 0.5$.

Tassi has been arguing that the small negative $\Delta\epsilon$ value ($-5 \leq \Delta\epsilon \leq 0$) for various binary mixtures may be attributed either to homoconjugation phenomena of the more polar component induced by the presence of an apolar or quasiapolar one and/or to associations between unlike molecules in the mixtures.^{13,16,17}

After taking into consideration the polarity ($\mu_{\text{ME}} = 2.08 \times 10^{-30} \text{ C} \cdot \text{m}$ and $\mu_{\text{DPA}} = 1.03 \times 10^{-30} \text{ C} \cdot \text{m}$)⁵ and the internal structure of studied solvents, the second hypothesis seems to be more probable.

As suggested by other authors,^{13,16,17} the study of this structural parameter for binary liquid systems represents a unique tool for investigating the formation of intermolecular complexes and provides a valuable aid for determining their stoichiometry.

Results obtained in this paper seem to indicate that the stable intermolecular complexes of the ME·DPA types are respectively formed in the studied ME + DPA binary mixtures.

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