

Literature Cited

- (1) Peng, D.; Robinson, D. B. *Ind. Eng. Chem. Fundam.* **1976**, *15*, 59-64.
- (2) Loos, Th. W. de; Kooi, H. J. van der; Ott, P. L. *J. Chem. Eng. Data* **1986**, *31*, 166-168.
- (3) Aaldijk, L. Ph.D Thesis, Delft University of Technology, Delft, 1971.
- (4) Boublik, T. J.; Fried, V.; Hala, E. *The Vapour Pressures of Pure Substances*; Elsevier: Amsterdam, 1984.
- (5) Douslin, D. R.; Harrison, R. H. *J. Chem. Thermodyn.* **1973**, *5*, 491-512.
- (6) Deiters, U.; Schneider, G. M. *Ber. Bunsen-Ges. Phys. Chem.* **1976**, *80*, 1316-1321.
- (7) Loos, Th. W. de; Poot, W.; Lichtenthaler, R. N. *Ber. Bunsen-Ges. Phys. Chem.* **1984**, *88*, 855-859.

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Physical Behavior of Some Reaction Media. Density, Viscosity, Dielectric Constant, and Refractive Index Changes of Ethanol-Dioxane Mixtures at Several Temperatures

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Viscosities, densities, dielectric constants, and refractive indexes were determined for ethanol-dioxane mixtures at 15, 20, 25, 30, and 35 °C. These properties are represented by means of empirical relations wherein the composition and temperature effects are involved. The analysis of the dielectric constant data revealed experimental evidence of molecular interactions between the components of the system.

Introduction

This paper is part of our research concerning the kinetics of S_N2 reactions between ions and dipolar molecules in solvents where the ionic reactant associates to form ion pairs (1-6).

In the progress of these investigations we decided to extend our studies to binary solvent systems, where the mixture of the two organic cosolvents, in various proportions, could enable us to change the physical properties of the reaction media.

Thus, in a first attempt we studied the kinetics of the reaction of sodium ethoxide and methyl iodide in ethanol-dioxane mixtures (6). In this investigation, in agreement with analogous literature kinetic data (7, 8), we observed that the corresponding reaction is markedly accelerated as the dioxane content is progressively increased. This effect has been attributed to changes in the reactant-solvating ability of the medium. Consequently, taking into account this behavior, the ethanol-dioxane mixtures can be considered as very important reaction media for various studies of nucleophilic substitution reactions.

One problem, generally encountered in such studies, is to correlate reaction rate data with the physical properties of the corresponding media.

However, among the many intensive physical properties of liquids the dielectric constant (ϵ), viscosity (η), and index of refraction (n) remain the common solvent parameters used to interpret medium effects upon mechanisms of reactions for polar and ionic species (9), as well as upon electrochemical data and ionic equilibria (10, 11).

Although extensive tabulation of values of these properties for pure solvents are generally available, literature data for various binary solvent systems are often incomplete and may be reported only as empirical graphs.

This situation has encouraged us to make a series of systematic measurements of density, viscosity, dielectric constant, and refractive index of ethanol-dioxane mixtures over the whole composition range and temperatures from 15 to 35 °C.

Because plots of Y values (where $Y = \epsilon, \eta, \rho, n$) vs. solvent composition are nonlinear, an attempt has been made to express these properties by means of single equations wherein the temperature and composition effects are involved.

It is noted that these equations can be used as interpolation formulas allowing prediction of the data at any temperature and composition.

Experimental Section

Reagents. Absolute ethanol (Fluka, p.a.) was further purified according to a recommended method (12).

Carlo Erba 1,4-dioxane (99%, bp 101 °C) was further purified by refluxing with sodium metal for several days and redistilling; the whole procedure was repeated until the sodium remained bright after several hours. In all distillations only the middle fraction coming over at the reported boiling point and comprising about 75% was retained. Pure-component physical properties are listed and compared to average literature data in Table I. Mixtures were gravimetrically prepared on a Mettler analytical balance just before their use. The probable error in the dioxane mole fraction X_2 is estimated to be less than 10^{-4} .

Measurements. Dielectric constant measurements were carried out at 2.0 MHz by the heterodyne beat method with a Wissenschaftliche-Technische Werkstätten DM 01 Dipolmeter. The thermostated measuring cells (MFL 1/S and MFL 2/S type) were adequate to cover the dielectric constant range of ethanol-dioxane mixtures at the temperatures used. The overall experimental uncertainty in ϵ values (standard deviation) was approximately equal to $\pm 0.2\%$. The cells were previously calibrated with standard pure liquids in accordance with the manufacturer's specifications and with National Bureau of Standards Circular 514 (13).

Density measurements were made in a capillary pycnometer of approximately 25 cm³ capacity. The pycnometer was calibrated with distilled water at each temperature. The standard deviations of the densities of the solutions used were less than 0.01%.

Viscosities were measured with an Ubbelohde capillary viscosimeter (Schott Mainz Jena Glas C 2023). In all determina-

Table I. Physical Properties of Pure Components

temp, °C	ρ , g cm ⁻³		η , cP		ϵ		n_D	
	exptl	lit.	exptl	lit.	exptl	lit.	exptl	lit.
Ethanol								
15	0.793 69	0.793 63 (15)	1.3182		26.24		1.363 62	1.363 30 (26)
20	0.789 22	0.789 40 (15)	1.2050		25.41		1.361 63	1.361 4 (15)
25	0.785 07	0.785 (16-18)	1.0710	1.073 (15, 19)	24.55	24.42 (13, 24)	1.359 72	1.359 5 (15)
30	0.780 76	0.780 79 (19)	0.9892	0.991 (21)	23.80	23.80 (24)	1.357 72	
35	0.776 81		0.8987	0.8683 (22)	23.06		1.355 71	
Dioxane								
15	1.038 24	1.039 22 (16)	1.4230	1.439 (23)	2.226		1.424 37	1.424 36 (23)
20	1.033 22	1.033 61 (15, 19)	1.2957		2.218	2.235 (25)	1.422 02	1.422 41 (27)
25	1.026 93	1.026 9 (20)	1.1969	1.192 (20)	2.209	2.209 (13)	1.420 60	1.420 2 (15)
30	1.022 62	1.022 30 (15, 19)	1.0891	1.087 (23)	2.201		1.417 42	
35	1.016 35		1.0313		2.192		1.415 43	

Table II. Experimental Density (ρ), Viscosity (η), Dielectric Constant (ϵ), and Refractive Index (n_D) Data for Ethanol-Dioxane Mixtures at 15, 20, 25, 30, and 35 °C and Dioxane Mole Fraction (X_2)

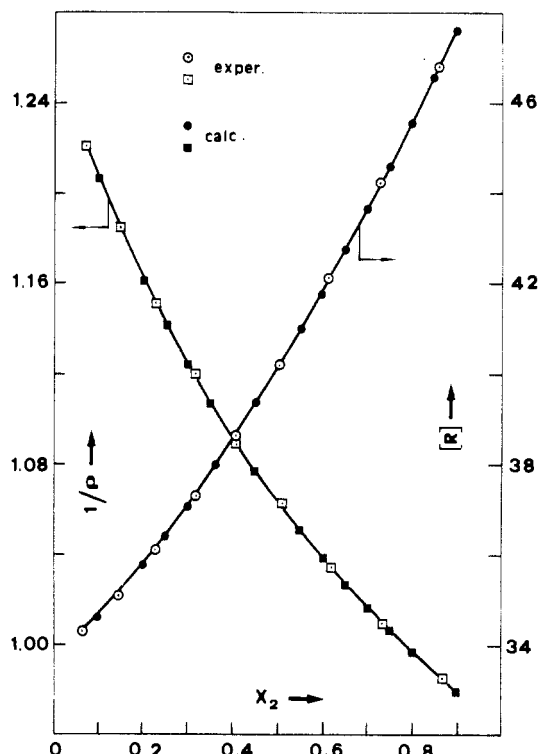
X_2	15 °C	20 °C	25 °C	30 °C	35 °C
ρ , g/cm ³					
0.0703	0.81907	0.81468	0.81264	0.80646	0.80153
0.1455	0.84425	0.83999	0.83599	0.83051	0.82547
0.2259	0.86821	0.86403	0.86000	0.85445	0.84883
0.3126	0.89255	0.88814	0.88303	0.87851	0.87302
0.4045	0.91825	0.91364	0.90864	0.90252	0.89683
0.5051	0.94059	0.93619	0.93182	0.92646	0.92034
0.6163	0.96589	0.96140	0.95287	0.94709	0.94396
0.7297	0.98974	0.98662	0.98063	0.97416	0.96800
0.8600	1.01413	1.00949	1.00288	0.99782	0.99160
η , cP					
0.0703	1.1640	1.0691	0.9600	0.8879	0.8147
0.1455	1.0757	0.9889	0.8885	0.8285	0.7595
0.2259	1.0264	0.9468	0.8529	0.7902	0.7308
0.3126	0.9996	0.9216	0.8387	0.7727	0.7174
0.4045	0.9999	0.9225	0.8391	0.7751	0.7182
0.5051	1.0059	0.9277	0.8469	0.7819	0.7278
0.6163	1.0453	0.9582	0.8803	0.8165	0.7622
0.7297	1.1197	1.0287	0.9425	0.8647	0.8101
0.8600	1.2343	1.1352	1.0436	0.9699	0.8938
ϵ					
0.0703	25.08	24.11	22.91	22.11	21.06
0.1455	20.70	19.90	18.97	18.23	17.46
0.2259	17.08	16.41	15.77	15.23	14.39
0.3126	13.33	12.87	12.35	11.81	11.47
0.4045	10.28	9.88	9.61	9.20	8.87
0.5051	7.92	7.63	7.43	7.16	6.93
0.6163	5.96	5.74	5.62	5.45	5.28
0.7297	4.348	4.238	4.163	4.072	3.988
0.8600	3.163	3.111	3.056	3.013	2.956
n_D					
0.0703	1.36970	1.36768	1.36597	1.36397	1.36246
0.1455	1.37592	1.37391	1.37181	1.36978	1.36781
0.2259	1.38187	1.37983	1.37775	1.37556	1.37374
0.3126	1.38805	1.38593	1.38382	1.38162	1.37959
0.4045	1.39418	1.39208	1.38987	1.38748	1.38598
0.5051	1.40101	1.39790	1.39603	1.39353	1.39147
0.6163	1.40729	1.40412	1.40227	1.39950	1.39808
0.7297	1.41234	1.41010	1.40801	1.40562	1.40427
0.8600	1.41812	1.41606	1.41382	1.41150	1.40954

tions the kinetic energy correction has been taken into account according to a recommended method (14). The standard deviation of the viscosity measurements at each composition was 0.2%.

Refractive indexes for the sodium D line were measured with a thermostated Pulfrich refractometer (Bellinghan and Stanley) with an error of less than ± 0.0001 units.

In all cases temperature was controlled within ± 0.01 °C by means of a Haake Ultrathermostat NBS.

Experiments were generally performed in five replicates for each composition and the results were averaged.

Figure 1. Variation of $1/\rho$ (cm³ g⁻¹) and $[R]$ (cm³ P^{1/8} mol⁻¹) with mole fraction X_2 of dioxane at 15 °C.

Results and Discussion

The experimental density, viscosity, dielectric constant, and refractive index data at 15, 20, 25, 30, and 35 °C for the ethanol-dioxane solvent system are listed in Table II. In all cases X_2 is the mole fraction of dioxane.

Densities. The polynomial equation

$$\frac{1}{\rho} = \sum_{i=0}^n d_i X_2^i \quad (1)$$

was fitted at each temperature by a least-squares technique. On the basis of careful examination of the results of the least-squares fitting and taking into account that as the degree, n , of the polynomial is increased the amount of smoothing is decreased, it was decided that the optimum degree n was equal to 4, namely much less than the number of data points. The values of the adjustable coefficients d_i are listed in Table III, along with the standard deviations σ . A comparison of experimental and calculated $1/\rho$ data at 15 °C is presented as example in Figure 1.

Viscosities. In an attempt to establish useful correlation functions relating the viscosity to the solvent composition, the

Table III. Coefficients and Standard Deviations σ for Representation of $1/\rho$, $[R]$, ϵ , and n_D of Ethanol-Dioxane Mixtures by Eq 1, 4, 10, and 20 at 15, 20, 25, 30, and 35 °C

	15 °C	20 °C	25 °C	30 °C	35 °C
Eq 1					
d_0	1.25885	1.26735	1.26739	1.27839	1.28549
$-d_1$	0.57599	0.61118	0.55087	0.57727	0.57094
d_2	0.51542	0.66459	0.44589	0.49108	0.47653
$-d_3$	0.34125	0.59724	0.26129	0.27366	0.29267
d_4	0.10666	0.25056	0.07218	0.05621	0.08701
$\sigma_{\rho^{-1}}$	± 0.00063	± 0.00068	± 0.00139	± 0.00115	± 0.00036
Eq 4					
d_0'	33.66	33.54	33.08	33.06	32.84
d_1'	8.024	6.897	8.490	7.843	8.810
d_2'	14.67	20.48	15.31	16.23	13.07
$-d_3'$	12.30	23.51	15.21	15.35	11.11
d_4'	5.891	12.56	7.862	7.494	5.410
$\sigma_{[R]}$	± 0.014	± 0.016	± 0.015	± 0.011	± 0.009
Eq 10					
D_0	29.56	28.39	26.88	25.94	24.76
$-D_1$	67.71	64.78	59.96	58.04	56.14
D_2	52.45	49.52	44.15	41.34	45.18
D_3	-0.6271	0.4597	2.333	6.819	-3.897
$-D_4$	12.02	11.88	11.72	14.52	8.162
σ_ϵ	± 0.09	± 0.08	± 0.09	± 0.11	± 0.06
Eq 20					
D_0'	1.36425	1.36143	1.36029	1.35816	1.35678
$10^2 D_1'$	7.925	9.283	8.245	8.498	8.182
$-10^2 D_2'$	-0.1529	5.731	2.264	3.780	3.772
$10^2 D_3'$	-3.823	4.192	-0.1023	2.007	3.905
$10^4 D_4'$	161.4	-173.2	0.3289	-82.56	-265.0
σ_{n_D}	$\pm 1.95 \times 10^{-4}$	$\pm 7.7 \times 10^{-5}$	$\pm 4.0 \times 10^{-7}$	$\pm 8.1 \times 10^{-5}$	$\pm 2.02 \times 10^{-4}$

rheochor $[R]$ (28, 29) for the various mixtures was calculated from the following equation by using smoothing density data

$$[R] = \frac{\bar{M}}{\rho} \eta^{1/8} \quad (2)$$

where

$$\bar{M} = (1 - X_2)M_1 + X_2M_2 \quad (3)$$

M_1 and M_2 being the molecular weights of ethanol and dioxane, respectively.

It was found that plots of $[R]$ vs. X_2 (see Figure 1) are not linear and that the corresponding data at each temperature can be fitted, by least-squares, to a polynomial-type equation

$$[R] = \sum_{i=0}^4 d_i' X_2^i \quad (4)$$

whose coefficients d_i' are summarized in Table III. The fidelity of these fits is also graphically represented by the example in Figure 1.

By combining eq 1 and 2, one obtains the following rational function (i.e., a ratio of two polynomials):

$$\eta^{1/8} = \frac{1}{\bar{M}} \frac{d_0' + d_1' X_2 + \dots + d_4' X_2^4}{d_0 + d_1 X_2 + \dots + d_4 X_2^4} \quad (5)$$

It should be noted that the advantages which can result from the use of rational functions rather than linear polynomials for fitting data have been discussed in the literature (30, 31). In accordance with this discussion, it was found in this investigation that, in all cases, eq 5 represents much better the reported viscosity data than a simple polynomial. The overall uncertainty in the calculated values of η is $\pm 2.4 \times 10^{-5}$ P (standard deviation). Figure 2 provides two examples of fitting the viscosity η as a function of X_2 at 25 and 35 °C.

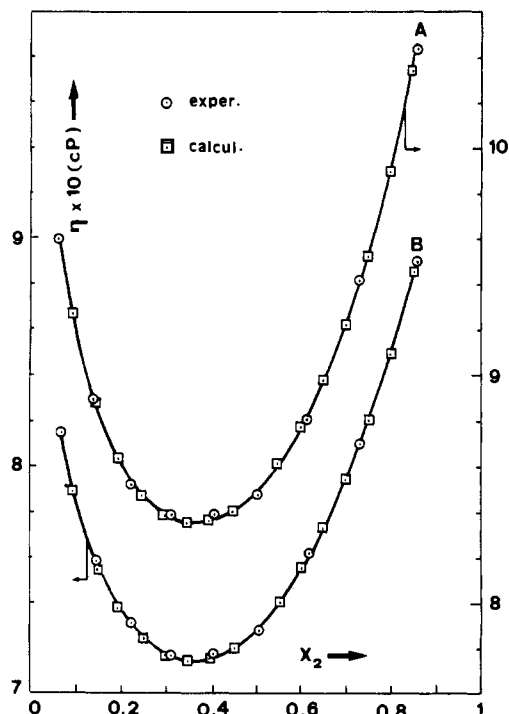


Figure 2. Variation of experimental (O) and calculated (□) viscosities with mole fraction X_2 of dioxane for the ethanol-dioxane systems at (A) 25 °C and (B) 35 °C.

The effect of temperature on the viscosity of the mixtures was examined by assuming the validity of the following equation advanced by Arrhenius

$$\eta = A \exp(B/RT) \quad (6)$$

R being the gas constant and T the absolute temperature.

Calculated viscosity data, by use of eq 5, was used in the plots of $\ln \eta$ against $1/T$. The mole fraction of dioxane was kept as a constant parameter. Straight lines were obtained for

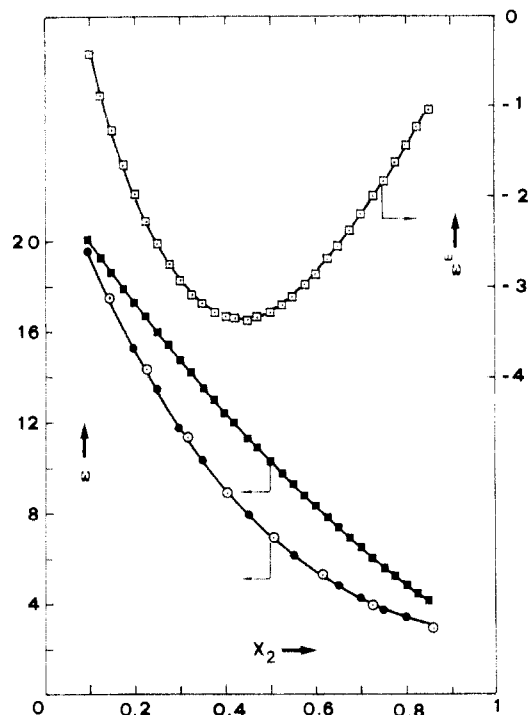


Figure 3. Variation of experimental (○), calculated (●), ideal (■), and excess (□) dielectric constants with mole fraction X_2 of dioxane at 35 °C.

each composition with a correlation coefficient greater than 0.999. The corresponding constants A and B were calculated by a least-squares method. Based on these data, the following relations were found:

$$10^5 A = \sum_{i=0}^5 a_i X_2^i \equiv 4.56 + 0.699X_2 + 3.27X_2^2 + 37.4X_2^3 - 87.2X_2^4 + 53.1X_2^5 \quad (7)$$

$$\sigma_A = 2.0 \times 10^{-8}$$

$$B = \sum_{i=0}^5 b_i X_2^i \equiv 3226.21 - 915.42X_2 + 506.51X_2^2 - 1675.14X_2^3 + 4602.6X_2^4 - 3025.51X_2^5 \quad (8)$$

$$\sigma_B = 0.045$$

It follows that the equation relating the viscosity to the temperature, T , and the mole fraction of dioxane, X_2 , is

$$\eta = \left(\sum_{i=0}^5 a_i X_2^i \right) \exp \left(\frac{\sum_{i=0}^5 b_i X_2^i}{RT} \right) \quad (9)$$

This equation fits the experimental data with an uncertainty of ± 0.0043 cP over the specified range of temperature (15–35 °C) and compositions ($0.07 \leq X_2 \leq 0.86$).

Dielectric Constants. The smoothing function

$$\epsilon = \sum_{i=0}^4 D_i X_2^i \quad (10)$$

was fitted at each temperature by the least-squares method. Values of the coefficients D_i and the standard deviations σ are summarized at Table III. The results of fitting the data for 35 °C are presented as an example in Figure 3.

The dielectric constant deviations from ideality were also considered because they give an adequate approach to estimate the intermolecular interaction between the components of the mixture.

These deviations were determined by

$$\epsilon^E = \epsilon - \epsilon^{id} \quad (11)$$

Where ϵ^E and ϵ^{id} are the excess and the ideal dielectric constant, respectively. The ideal dielectric constants of the mixtures were computed by using Decroocq's formula (32) in the most general form

$$\epsilon^{id} = (1 - Y_2)\epsilon_1 + Y_2\epsilon_2 + 2 \left(\frac{1}{\epsilon^{id}} - \frac{(1 - Y_2)}{\epsilon_1} - \frac{Y_2}{\epsilon_2} \right) \quad (12)$$

where ϵ_1 and ϵ_2 are the dielectric constants of ethanol and dioxane, respectively, Y_2 is the volume fraction defined on the partial molar volume basis (33)

$$Y_2 = \frac{X_2 \bar{V}_2}{X_2 \bar{V}_2 + (1 - X_2) \bar{V}_1} \quad (13)$$

\bar{V}_2 and \bar{V}_1 being the dioxane and methanol partial molar volumes, respectively. The partial molar volumes \bar{V}_1 and \bar{V}_2 were determined by means of the following equations (33) modified in the mole fraction scale

$$\bar{V}_1 = M_1 \frac{1}{\rho} - \bar{M} X_2 \frac{d(1/\rho)}{dX_2} \quad (14)$$

$$\bar{V}_2 = M_2 \frac{1}{\rho} - \bar{M}(1 - X_2) \frac{d(1/\rho)}{dX_2} \quad (15)$$

It was found that ϵ^E values are generally negative and the curves $\epsilon^E = f(X_2)$ corresponding at each temperature present a pronounced minimum as it is shown in the example of Figure 3.

At the molecular level, it has been argued (31, 33–35) that the negative values of ϵ^E for various systems are a consequence of associations between the components of the mixture. Another point of interest is that the maximum deviations from ideality occur for all temperatures around the composition $X_2 \approx 0.45$. This behavior could be attributed to the formation of two complexes between dioxane and ethanol at molar ratios of 1:1 and 1:2. It should be noted that this assumption is in agreement with the possibility that dioxane can be associated through H bonds to either of its oxygen atoms.

The variation of dielectric constant with temperature was studied by using the following equation (13, 36)

$$\epsilon = A' \exp(-B'/T) \quad (16)$$

A similar form of calculation, as described before in the case of eq 6, was applied. The obtained results are graphically represented in Figure 4. The following equations were obtained:

$$A' = \sum_{i=0}^5 \alpha_i X_2^i \equiv 381.40 - 1053.46X_2 + 673.74X_2^2 + 648.56X_2^3 - 940.98X_2^4 + 290.03X_2^5 \quad (17)$$

$$\sigma_{A'} = 0.036$$

$$10^3 B' = \sum_{i=0}^5 \beta_i X_2^i \equiv 8.971 - 3.817X_2 + 11.164X_2^2 - 46.797X_2^3 + 52.845X_2^4 - 20.928X_2^5 \quad (18)$$

$$\sigma_{B'} = 6.01 \times 10^{-3}$$

Consequently, the relation of dielectric constant with composition and temperature is

$$\epsilon = \left(\sum_{i=0}^5 \alpha_i X_2^i \right) \exp \left(-T \sum_{i=0}^5 \beta_i X_2^i \right) \quad (19)$$

This equation predicts the experimental data with an overall uncertainty of ± 0.1 units.

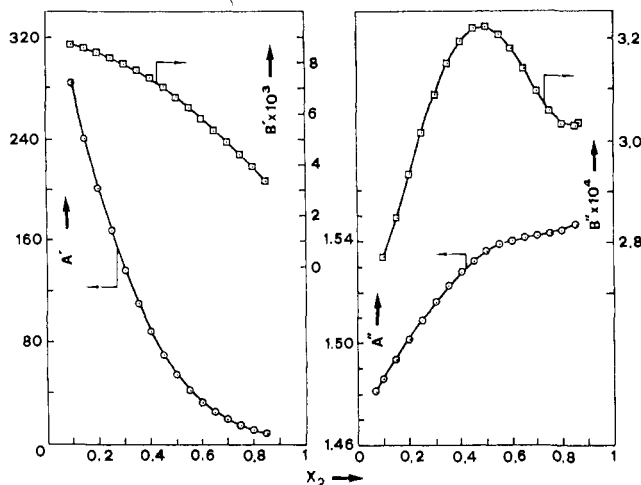


Figure 4. Variation of the constants A' , B' (eq 16) and A'' , B'' (eq 21) with mole fraction X_2 of dioxane.

Refractive Indexes. A similar form of calculation, as before, was applied. Thus, the smoothing function

$$n_D = \sum_{i=0}^4 D_i' X_2^i \quad (20)$$

was fitted at each temperature. The coefficients D_i' , obtained by least squares, are listed in Table III.

The effect of temperature on n_D was studied by means of the following equation:

$$n_D = A'' \exp(-B''/T) \quad (21)$$

The obtained values of A'' and B'' for various compositions are graphically shown in Figure 4 and the corresponding curves were fitted to the following equations:

$$A'' = 1.47329 + 0.09968X_2 + 0.362706X_2^2 - 0.870236X_2^3 + 0.504727X_2^4 - 0.00449X_2^5 \quad (22)$$

$$\sigma_{A''} = 5 \times 10^{-6}$$

$$B'' = 2.6774 + 0.23033X_2 + 8.7186X_2^2 - 19.9722X_2^3 + 12.2269X_2^4 - 0.58118X_2^5 \quad (23)$$

$$\sigma_{B''} = 1.2 \times 10^{-4}$$

By combining eq 21, 22, and 23 it is possible to predict experimental data with an average uncertainty ± 0.00020 units.

Glossary

ρ	density of the mixture, g cm^{-3}
η	viscosity of the mixture, cP
ϵ	dielectric constant of the mixture
ϵ^E	excess dielectric constant of the mixture
ϵ^{id}	ideal dielectric constant of the mixture
n_D	refractive index of the mixture for sodium light
T	absolute temperature
$[R]$	rheochor of the mixture defined by eq 2

\bar{V}_1, \bar{V}_2	partial molar volumes of ethanol and dioxane
X_2	mole fraction of dioxane
Y_2	volume fraction of dioxane
A, B	constants in eq 6
A', B'	constants in eq 16
A'', B''	constants in eq 21
M_1, M_2	molecular weights of ethanol and dioxane
$d_0 \dots d_4$	coefficients in eq 1
$d'_0 \dots d'_4$	coefficients in eq 4
$D_0 \dots D_4$	coefficients in eq 10
$D'_0 \dots D'_4$	coefficients in eq 20
σ	standard deviation

Literature Cited

- (1) Cayzergues, P.; Georgoullis, C.; Papanastasiou, G. *J. Chim. Phys.* **1977**, *74*, 1103.
- (2) Cayzergues, P.; Georgoullis, C.; Papanastasiou, G. *J. Chim. Phys.* **1977**, *74*, 1112.
- (3) Cayzergues, P.; Georgoullis, C.; Papanastasiou, G. *C. R. Acad. Sci. Paris* **1977**, *C183*, 285.
- (4) Papanastasiou, G.; Papoutsis, A.; Jannakoudakis, D. *Chim. Chron., New Ser.* **1984**, *73*, 99.
- (5) Papanastasiou, G.; Papoutsis, A.; Jannakoudakis, D.; Georgoullis, C. *J. Chim. Phys.* **1985**, *83*, 907.
- (6) Papoutsis, A.; Papanastasiou, G.; Jannakoudakis, D.; Georgoullis, C. *J. Chim. Phys.* **1985**, *83*, 913.
- (7) Tchoubar, B. *Bull. Soc. Chim. Fr.* **1984**, 2069 and references cited therein.
- (8) Guibe, F.; Bram, G. *Bull. Soc. Chim. Fr.* **1975**, 933 and references cited therein.
- (9) Amls, E. S. *Solvent Effects on Reaction Rates and Mechanisms*; Academic: New York, 1966; Chapters 1 and 2.
- (10) Lagowski, J. J., Ed. *The Chemistry of Nonaqueous Solvents*; Academic: New York, 1978; Vol. VA, pp 121-178.
- (11) King, E. J. *Acid-Base Equilibria*; Pergamon: Oxford, U.K., 1965; Chapters 7, 8, and 10.
- (12) Czernecki, S.; Georgoullis, C.; Prevosts, Ch. *Bull. Soc. Chim. Fr.* **1970**, 3088.
- (13) Maryott, A. A.; Smith, E. R. "Table of Dielectric Constants of Pure Liquids; *Natl. Bur. Stand. Circ.* **1951**, No. 514.
- (14) Cannon, M. R.; Monning, R. E.; Bell, J. D. *Anal. Chem.* **1960**, *32*, 355.
- (15) Timmermans, J. *Physical-Chemical Constants of Pure Organic Compounds*; Elsevier: New York, 1964; Vol. II.
- (16) Riddick, J. A.; Bunger, W. B. *Organic Solvents. Techniques of Chemistry*; Wiley-Interscience: New York, 1970; Vol. II.
- (17) Mussche, M. J.; Verhoeve, L. A. *J. Chem. Eng. Data* **1975**, *20*, 46.
- (18) Wel, C.; Rowley, R. L. *J. Chem. Eng. Data* **1984**, *29*, 332.
- (19) Janz, G. J.; Tomkins, R. P. T. *Nonaqueous Electrolytes Handbook*; Academic: New York, 1972; Vol. 1.
- (20) Stokes, R. H.; Mills, R. *Viscosity of Electrolytes and Related Properties*; Pergamon: Oxford, U.K., 1965; Vol. 3, p 76.
- (21) Titanl, T. *Bull. Inst. Res. Jpn.* **1927**, 671.
- (22) Guy, J. S.; Jones, H. C. *Am. Chem. J.* **1911**, *46*, 131.
- (23) Timmermans, J. *J. Chim. Phys.* **1937**, *34*, 693.
- (24) Dannhauser, W.; Bahe, L. W. *J. Chem. Phys.* **1964**, *40*, 3058.
- (25) Ulich, H.; Nesjulat, W. *Z. Phys. Chem.* **1932**, *16B*, 221.
- (26) Dorochewsky, A. G. *J. Russ. Phys. Chem. Soc.* **1911**, *43*, 66.
- (27) Hess, K.; Frahm, H. *Ber.* **1938**, *71*, 2627.
- (28) Reynaud, R. *Bull. Soc. Chim. Fr.* **1971**, 4269.
- (29) Partington, J. R. *An Advanced Treatise on Physical Chemistry*; Longmans: London, 1951; Vol. 2, pp 94, 119.
- (30) King, M. B.; Queen, N. M. *J. Chem. Eng. Data* **1979**, *24*, 176.
- (31) Koelling, O. W. *Anal. Chem.* **1985**, *57*, 1721.
- (32) Decroocq, D. *Bull. Soc. Chim. Fr.* **1963**, 127.
- (33) Reynaud, R. *Bull. Soc. Chim. Fr.* **1972**, 532.
- (34) Hoover, T. B. *J. Phys. Chem.* **1969**, *73*, 57.
- (35) Payne, R.; Theodorou, I. J. *J. Phys. Chem.* **1972**, *76*, 2892.
- (36) Jannakoudakis, D.; Papanastasiou, G.; Mavridis, P. G. *J. Chim. Phys.* **1976**, *73*, 156.

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