Table VII.	Calculated	Average D	eviations	$(\Delta n)_{av}$ for
Different M	lixing Rule	s at Variou	18 Temper	atures

		(,	$(\Delta n)_{\rm av} \times 10$) ³		
t/°C	AB⁴	GD	LL	Wď	He	
5	0.87	0.87	0.87	0.87	0.87	
10	0.79	0.79	0.79	0.79	0.79	
15	1.03	1.03	1.03	1.03	1.03	
20	0.84	0.84	0.84	0.84	0.84	
25	0. 9 1	0.91	0. 9 1	0.91	0.91	
30	0.77	0.77	0.77	0.77	0.77	
35	1.05	1.05	1.05	1.05	1.05	
40	0.96	0.96	0.96	0.96	0.96	
45	0.95	0.95	0.95	0.95	0.96	
50	0.92	0.92	0.92	0.92	0.92	
55	1.10	1.10	1.10	1.10	1.10	
60	0.7 9	0.79	0.7 9	0.7 9	0.79	
65	0.81	0.81	0.81	0.81	0.81	
70	0.81	0.81	0.81	0.81	0.81	
75	0.7 9	0.79	0.7 9	0.7 9	0.79	
80	0.66	0.66	0.66	0.66	0.66	
av	0.89	0.89	0.89	0.89	0.89	

^aArago-Biot equation (7). ^bGladstone-Dale equation (8). ^cLorentz-Lorenz equation (9). ^dWiener equation (10). ^eHeller equation (11).

The results of this analysis are summarized in Table VII, where we have reported the average deviations of the refractive index differences, (Δn) , expressed as

$$\Delta n = n_{\text{exott}} - n_{\text{calcd}} \tag{12}$$

with n_{calcd} evaluated by means of different mixing rules. The five mixing rules tested are equivalent to each other, all reproducing the experimental data within the limits of experimental uncertainty over the range of temperature and composition studied.

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Glossary

A',B'	coefficients in eq 3
a,'	coefficients in eq 4
b,'	coefficients in eq 5
n	refractive index
t	Celsius temperature
_	

Т absolute temperature, K

Greek Letters

~	coefficiente	in	60	4
α_{i}	coefficients	IN	ea	

- β_{l} coefficients in eq 2
- dielectric constant E
- volume fraction of the /th component φ,
- standard deviation

Registry No. DMF, 68-12-2; DHE, 107-21-1.

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Densities and Excess Molar Volumes of the 1.2–Ethanediol +2–Methoxyethanol Solvent System at Various Temperatures

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Densities are reported for the binary mixture 1,2-ethanediol + 2-methoxyethanol in the whole composition range at 19 temperatures between -10 and +80 °C. The excess molar volumes, V^E, calculated from the density values, are negative at all concentrations and become more negative with increasing temperature.

Introduction

In recent papers we have reported some electroanalytical studies about acid-base equilibria which take place in solutions of 1,2-ethanediol and 2-methoxyethanol and in their binary mixtures operating at different temperatures in the range -10 to +80 °C (1-4). These alcohols appear very interesting from the point of view of practical and theoretical applications, being protic, protophilic, and dipolar, as well as potentially acidic, nonaqueous solvents. 1,2-Ethanediol is the simplest homologue of the diol series, largely utilized as thermoregulator fluid, as a controlling agent of density/viscosity reaction baths, and as emulsion coating owing to its unusual physicochemical properties (5).

On the other hand, 2-methoxyethanol is widely used for various industrial processes (6) and has unique solvating properties associated with its quasi-aprotic character (5). 2-Methoxyethanoi, an ether alcohol, shows physicochemical characteristics midway between protic and dipolar aprotic solvents. It is a quite-toxic solvent, a weak narcotic, and a

Table I. Experimental Densities ρ for 1,2-Ethanediol (1) + 2-Methoxyethanol (2) Mixtures at Temperature t and 1,2-Ethanediol Mole Fraction X_1

	$\rho(t)/(\text{g cm}^{-3})$										
t/°C	$X_1 = 1.0000$	$X_1 = 0.9272$	$X_1 = 0.8499$	$X_1 = 0.7676$	$X_1 = 0.6799$	$X_1 = 0.5861$	$X_1 = 0.4856$	$X_1 = 0.3776$	$X_1 = 0.2614$	$X_1 = 0.1359$	$X_1 = 0.0000$
-10	1.133 634	1.122 202	1.110813	1.098 209	1.085 729	1.070110	1.054 848	1.038 826	1.026 487	1.010 245	0.993 991
-5	1.130452	1.118673	1.107184	1.094 298	1.081503	1.065827	1.050744	1.034 589	1.022101	1.005586	0.989 269
0	1.127069	1.115021	1.103 481	1.090 306	1.077314	1.061 729	1.046656	1.030 608	1.017 692	1.000959	0.984 554
5	1.123664	1.111518	1.099 695	1.086543	1.073294	1.057585	1.042436	1.026387	1.013150	0.996487	0.979847
10	1.120 296	1.107882	1.095817	1.082441	1.069 197	1.053481	1.038353	1.022291	1.008704	0.991 703	0.975032
15	1.116 906	1.104475	1.091 976	1.078597	1.065028	1.049326	1.034117	1.018069	1.004 203	0.987067	0.970 166
20	1.113 440	1.100 808	1.088 293	1.074614	1.060 991	1.045274	1.030032	1.014071	0.999 655	0.982510	0.965 443
25	1.109 913	1.097382	1.084387	1.070649	1.056987	1.041097	1.025949	1.009 983	0.995 099	0.977821	0.960 490
30	1.106512	1.093 799	1.080547	1.066795	1.052926	1.037 069	1.021 698	1.005723	0.990612	0.973268	0.955845
35	1.103 089	1.090167	1.076791	1.062799	1.048845	1.033050	1.017505	1.001639	0.986 106	0 .968 765	0.951088
40	1.099747	1.086 603	1.072 955	1.058792	1.044743	1.029002	1.013318	0.997 490	0.981 608	0.964 084	0.946 292
45	1.096 312	1.082933	1.069052	1.054882	1.040518	1.024798	1.009 150	0.993172	0.977233	0.959 648	0.941 621
50	1.0 92 946	1.079 448	1.065 369	1.050 99 0	1.036511	1.020629	1.005091	0.989004	0.972 696	0.955005	0.936878
55	1.089 471	1.075813	1.061563	1.046907	1.032404	1.016613	1.000 986	0.984748	0.968288	0.950522	0.932133
60	1.086011	1.072201	1.057814	1.043059	1.028162	1.012369	0.996739	0.980629	0.963 869	0.945932	0.927387
65	1.082634	1.068671	1.053993	1.039 208	1.024105	1.008397	0. 992 61 5	0.976407	0.959 506	0.941 276	0.922714
70	1.079003	1.065177	1.050218	1.035 293	1.019847	1.004 246	0.988 319	0.972 290	0.955077	0.936777	0.917811
75	1.075369	1.061687	1.046572	1.031 403	1.015848	1.000332	0.984214	0.968195	0.950632	0.932055	0.913087
80	1.071 699	1.058169	1.042838	1.027627	1.011990	0.996274	0.980 113	0.964 025	0.946 094	0.927 455	0.908 144

systemic poison (7) and must be handled with care.

Attention is here devoted to some correlation procedures available for the thermomechanical parameters, such as density and related mixing properties. The literature data collections on these properties of binary, or multicomponent, solvent systems are scarce, and the data are often reported incompletely or only graphically. The fact that the extrathermodynamic properties of binary mixtures depend on temperature and on composition makes possible the variation "in continuum" of these properties and can provide more and useful structural information for liquid mixtures not otherwise available.

Experimental Section

Materials. The solvents, 1,2-ethanediol and 2-methoxyethanol (containing < 0.10% and < 0.05% water, respectively, found by Karl-Fischer titration), were Carlo Erba (Milan) highpurity grade.

Apparatus. The mixtures were prepared just before their use by weight through a Mettler PM 4800 Δ -range balance operating in a dry nitrogen atmosphere and then stored over molecular sieves. The estimated error in the 1,2-ethanediol mole fraction, X_1 , is less than 1.5×10^{-4} . The densities were determined with a digital density meter (Anton Paar DMA 60) equipped with a density-measuring cell (DMA 602), with a sensitivity up to 10^{-6} g cm⁻³. The temperature was maintained constant to ± 0.02 °C by means of two Lauda K2R thermostatic baths in cascade. The thermal control was checked by a thermoresistance Pt100 (Tersid, Milan) inserted into the jacket of the measuring cell and near the sample capillary tube. Karl-Fisher titrations were performed with an automatic titration system (Crison Model KF431) equipped with a digital buret (Crison Model 738).

Procedure. The density determination was performed by measuring the period of oscillation τ of a vibrating U-shaped sample tube, filled with the liquid sample. The following relationship exists between τ and the density ρ at the temperature t:

$$\rho(t) = (\tau^2 - B)/A$$

where A and B are instrumental constants which are determined by calibration with fluids of known density (dried air and bidistilled water) at each investigated temperature.

For the measurements, we have selected the period of 2000 oscillation cycles at each temperature and collected the values at least 2 h after the sample injection. Temperature equilibration was conveniently followed over ca. 30 readout cycles

Fable II .	Reference	Density	Values	ρof	1,2-Ethanediol	and
2-Methox;	yethanol at	Temper	ature t			

			,	
$ ho/(\mathrm{g~cm^{-3}})$	t/°C	measurement technique	ref	
	1,2-E	thanediol		
1.1099	25	pycnometer	17	
1.10982	25	pycnometer	10	
1.1098	25	pycnometer	18	
1.1096	25	oscillating tube	19	
1.10982	25	oscillating tube	20	
1.1063	30	oscillating tube	21	
1.109810	40	oscillating tube	9	
	2-Meth	oxyethanol		
0.96002	25	oscillating tube	20	
0.9605	25	oscillating tube	22	
0.946169	40	oscillating tube	23	
		-		

until the τ value becomes constant within ± 1 (8), and with a maximum variation between each series of τ of ± 3 .

Results and Discussion

The results of the experimental measurements of density for the two pure solvents and nine binary mixtures at 19 temperatures in the $-10 \le t/^{\circ}C \le +80$ range, with thermal scanning of 5 °C, are summarized in Table I.

For comparison purposes, the Table II shows some literature values relative to the two pure solvents and in particular to 1,2-ethanediol even if some authors reported many other data about the dependence of its density on the temperature (9). The discrepancies between these values are probably ascribable to the very different measurement methods and to the different procedures of samples purification adopted by the authors. These problems have been throughly discussed by Fogg et al. (10). Our density values were correlated as a function of temperature by the equation (11)

$$\rho(t)/(\text{g cm}^{-3}) = \sum_{i=0}^{4} \rho_i (t/^{\circ}\text{C})^i$$
 (1)

through a multilinear regression package (12), and the ρ_i coefficients are reported, together with the standard deviations $\sigma(\rho)$ for each solvent system, in Table 1 of supplementary material. The $\sigma(\rho)$ values were calculated with the equation

$$\sigma(\rho) = \left[\frac{\sum (\rho_{\text{calcd}} - \rho_{\text{exptl}})^2}{N - n}\right]^{1/2}$$
(2)

Table III.	Coefficients ρ_i	j of Equation	5 for 1,2-Ethanediol
$(1) + 2 \cdot M_0$	ethoxyethanol ((2) and Stand	iard Deviations $\sigma(\rho)$

	variable	ρ _{ij}		
ij	quantity	expanded eq	simplified eq	
00		0.969 361	0.969 169	
01	X_1	0.240275	0.238 802	
02	X_{1}^{-2}	$-3.56128 imes 10^{-2}$	-2.42431×10^{-2}	
03	X_{1}^{3}	0.218702	0.199 593	
04	X_{1}^{4}	-0.123 268	-0.113657	
10	t	-1.88166×10^{-3}	$-1.85622 imes 10^{-3}$	
20	t^2	$-2.65517 imes 10^{-7}$		
30	t ³	3.88003 × 10 ⁻⁸	1.390 47 × 10 ⁻⁸	
40	t4	-3.09306×10^{-10}	-8.43209×10^{-11}	
11	tX_1	2.015 02 × 10 ⁻⁴	2.080 18 × 10 ⁻⁴	
21	t^2X_1	-4.45378 × 10 ⁻⁶		
31	t^3X_1	4.68207×10^{-8}		
41	t^4X_1	2.31353×10^{-10}		
12	tX_1^2	1.38747×10^{-3}	9.817 46 × 10 ⁻⁴	
22	$t^2 X_1^2$	3.05681×10^{-5}		
32	$t^{3}X_{1}^{2}$	-6.64792×10^{-7}		
42	$t^{4}X_{1}^{2}$	2.20058×10^{-9}		
13	tX_1^3	-3.74338×10^{-3}	-2.77453×10^{-3}	
23	$t^2 X_1^3$	-3.31773×10^{-5}		
33	$t^{3}X_{1}^{3}$	7.15875 × 10 ⁻⁷		
43	$t^{4}X_{1}^{3}$	-8.40570×10^{-10}		
14	tX_1^4	2.54265×10^{-3}	1.90507×10^{-3}	
24	$t^{2}X_{1}^{4}$	5.71184×10^{-6}		
34	$t^{3}X_{1}^{4}$	-8.81639 × 10 ⁻⁸		
44	$t^{4}X_{1}^{4}$	-1.65321×10^{-9}		
σ(ρ)/(g ci	m⁻³)	0.000 94	0.000 93	

where N is the number of data points for each system and n is the number of parameters. Equation 1 reproduces the experimental values of Table I with a mean deviation $\Delta(\rho)$ equal to ± 0.00004 g cm⁻³, calculated as follows:

$$\Delta(\rho) = \frac{\sum |\rho_{calcd} - \rho_{exptl}|}{N}$$
(3)

The dependence of the experimental densities on the composition of the binary mixture was represented by the polynomial equation (13)

$$\rho(X_1)/(\text{g cm}^{-3}) = \sum_{j=0}^{4} \rho_j X_1^{j}$$
 (4)

whose coefficients are listed in the Table 2 (supplementary material) at each investigated temperature. Equation 4 fits the experimental values (Table I) with a mean deviation $\Delta(\rho) = \pm 0.00026 \text{ g cm}^{-3}$. This relatively large value does not signifi-

icantly diminish when higher polynomials are used. Even for a polynomial of degree 9, that is the maximum degree compatible with the limited number of points available throughout the entire composition range, the above difference does not become comparable with the experimental uncertainty.

The two eqs 1 and 4 are now available for the dependence of ρ on t and X₁, respectively. They can be combined to give a single function $\rho = \rho(t, X_1)$ of the type

$$\rho(t,X_1)/(\text{g cm}^{-3}) = [\rho(t) \ \rho(X_1)] = [\sum_{i,j=0}^{4} \rho_{i,j}(t/^{\circ}\text{C})'X_1^{i}]^{1/2}$$
 (5)

Eq 5 in its complete form reproduces the experimental ρ values with a mean deviation of ± 0.00030 g cm⁻³.

In this way, eq 5 provides the lowest difference between calculated and experimental values. Now, another and perhaps more useful strategy is to reduce as much as possible the number of terms, having always acceptable mean relative deviation between calculated and observed values. This is obtained by omitting in successive steps all the terms having statistical weights less than unity, thus emphasizing the importance of the remaining ones (12). Table III contains the ρ_{ij} coefficients of eq 5 in its complete form and in the simplified one, together with the corresponding variable quantities and the standard deviations $\sigma(\rho)$.

The simplified equation reproduces the experimental values within $\Delta(\rho) = \pm 0.00031 \text{ g cm}^{-3}$ (practically the same uncertainty found with the 25-terms complete form equation) and shows the advantage to contain only 12 terms, having eliminated all the t^2 terms and all the cross terms tX_1^{f} with $l \ge 2$.

Excess Volumes. The experimental ρ values at each temperature were converted into excess molar volumes of mixing $V^{\rm E}$

$$V^{\rm E} = \frac{(X_1M_1 + X_2M_2)}{\rho} - (X_1V_1 + X_2V_2) \tag{6}$$

where X_1 , X_2 are the mole fractions, V_1 , V_2 the molar volumes of pure components, and M_1 and M_2 the molar masses of the solvents, respectively (Table IV). The system exhibits negative deviations from ideality at all the investigated temperatures. The experimental V^E data were fitted by least-squares to a smoothing equation of the Redlich-Kister (14) type, as employed by several authors (15, 16).

$$V^{\rm E}/({\rm cm}^3 {\rm mol}^{-1}) = X_1 X_2 \sum_{k=0}^3 a_k (X_1 - X_2)^k$$
 (7)

Table IV. Excess Molar Volumes V^{E} of 1,2-Ethanediol (1) + 2-Methoxyethanol (2) at Temperature t and 1,2-Ethanediol Mole Fraction X_{1}

				1	$V^{\mathbf{E}}/(\mathrm{cm}^3 \mathrm{mol}^{-1})$	•)			
t/°C	$X_1 = 0.9272$	$X_1 = 0.8499$	$X_1 = 0.7676$	$X_1 = 0.6799$	$X_1 = 0.5861$	$X_1 = 0.4856$	$X_1 = 0.3776$	$X_1 = 0.2614$	$X_1 =$
-10	-0.1194	-0 2537	-0.3322	-0 4285	-0.3485	-0.2867	0 1689	-0 2957	-0.1554
-5	-0.1110	-0.2492	-0.3238	-0.4108	-0.3385	-0.2921	-0.1795	-0.3028	-0.1526
0	-0.1076	-0.2463	-0.3192	-0.4013	-0.3359	-0.3009	-0.1964	-0.3098	-0.1532
5	-0.1078	-0.2443	-0.3174	-0.3977	-0.3387	-0.3121	-0.2175	-0.3166	-0.1563
10	-0.1102	-0.2427	-0.3171	-0.3980	-0.3451	-0.3249	-0.2412	-0.3232	-0.1613
10	-0.1137 -0.1174	-0.2411	-0.3175	-0.4006		-0.3384	-0.2662	-0.3295	-0.1677
25	-0.1206	-0.2369	-0.3179	-0.4037	-0.3631	-0.3522	-0.2913	-0.3359	-0.1750
30	-0.1229	-0.2340	-0.3172	-0.4078	-0.3812	-0.3785	-0.3383	-0.3484	-0.1907
35	-0.1241	-0.2307	-0.3155	-0.4075	-0.3888	-0.3908	-0.3593	-0.3550	-0.1986
40	-0.1242	-0.2271	-0.3131	-0.4052	-0.3951	-0.4023	-0.3785	-0.3620	-0.2063
45	-0.1236	-0.2237	-0.3104	-0.4012	-0.4005	-0.4134	-0.3964	-0.3698	-0.2138
50 55	-0.1228 -0.1926	-0.2207	-0.3078	-0.3960	-0.4054	-0.4244	-0.4135	-0.3786	-0.2212
60	-0.1242	-0.2191	-0.3065	-0.3907	-0.4106	-0.4308	-0.4309	-0.3667	-0.2260
65	-0.1291	-0.2228	-0.3102	-0.3857	-0.4278	-0.4634	-0.4724	-0.4146	-0.2446
70	-0.1388	-0.2303	-0.3188	-0.3900	-0.4434	-0.4818	-0.5006	-0.4313	-0.2544
75	-0.1556	-0.2431	-0.3341	-0.4024	-0.4666	-0.5050	-0.5368	-0.4512	-0.2663
80	-0.1817	-0.2627	-0.3583	-0.4259	-0.5003	-0.5347	-0.5844	-0.4751	-0.2810

Table V. Coefficients a, of Equation 7 for 1,2-Ethanediol (1) + 2-Methoxyethanol (2) at Temperature t

					$\sigma(V^{\mathbf{E}})/$
t/°C	a_0	a_1	a_2	a_3	$(10^{-2} \text{ cm}^3 \text{ mol}^{-1})$
-10	-1.2087	-1.4023	-1.2252	2.2868	4.8
-5	-1.2134	-1.2110	-1.1365	1.9737	4.7
0	-1.2401	-1.0456	-1.0491	1.6945	4.5
5	-1.2823	-0.9024	-0.9630	1.4467	4.2
10	-1.3344	-0.7743	-0.8774	1.2269	3.9
15	-1.3917	-0.6583	-0.7931	1.0345	3.5
20	-1.4504	-0.5466	-0.7114	0.8592	3.1
25	-1.5073	-0.4404	-0.6316	0.7063	2.8
30	-1.5601	-0.3333	-0.5553	0.5635	2.4
35	-1.6084	-0.2243	-0.4825	0.4295	2.1
40	-1.6515	-0.1115	-0.4151	0.2980	1.8
45	-1.6909	0.0073	-0.3539	0.1598	1.6
50	-1.7284	0.1323	-0.2995	0.0109	1.3
55	-1.7672	0.2636	-0.2534	-0.1604	1.1
60	-1.8121	0.3988	-0.2170	-0.3562	0.9
65	-1.8686	0.5394	-0.1927	-0.5937	0.7
70	-1.9442	0.6808	-0.1807	-0.8753	0.6
75	-2.0466	0.8196	-0.1852	-1.2106	0.8
80	-2.1862	0.9544	-0.2060	-1.6158	1.3



Figure 1. Excess molar volumes V^{E} vs mole fraction X_{2} of 2-methoxyethanol at various temperatures ranging from -10 to +80 °C.

whose coefficients a_k are listed in Table V, along with the standard deviations $\sigma(V^E)$. This function reproduces the experimental V^E with a mean deviation $\Delta(V^E)$ of ±0.051 cm³ mol⁻¹.

The trend of eq 7 is shown in Figure 1, where, for the sake of clarity, the experimental points were not represented.

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Supplementary Material Available: Tables 1 and 2, listing the ρ_i and ρ_j coefficients of eqs 1 and 2, respectively (2 pages). Ordering information is given on any current masthead page.