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## Physics and Chemistry of Liquids

Publication details, including instructions for authors and subscription information:

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### Densities and Viscosities of the Ternary Mixture (Benzene 1-Propanol Ethyl Acetate) at 298.15 K

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**To cite this Article** Mariano, A. , Postigo, M. , Artigas, H. , Pardo, J. and Royo, F. M.(2000) 'Densities and Viscosities of the Ternary Mixture (Benzene 1-Propanol Ethyl Acetate) at 298.15 K', *Physics and Chemistry of Liquids*, 38: 5, 567 — 581

**To link to this Article:** DOI: 10.1080/00319100008030304

**URL:** <http://dx.doi.org/10.1080/00319100008030304>

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## DENSITIES AND VISCOSITIES OF THE TERNARY MIXTURE (BENZENE + 1-PROPANOL + ETHYL ACETATE) AT 298.15 K

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(Received 14 July 1999)

Densities and viscosities of the ternary mixture (benzene + 1-propanol + ethyl acetate) and the corresponding binary mixtures (benzene + 1-propanol, benzene + ethyl acetate and 1-propanol + ethyl acetate) have been measured at the temperature 298.15 K. From these measurements excess volumes,  $V^E$ , excess viscosities,  $\eta^E$ , and excess Gibbs energies of activation for viscous flow,  $G^{*E}$ , have been determined. The equation of Redlich-Kister has been used for fitting the excess properties of binary mixtures. The excess properties of the ternary system were fitted to Cibulka's equation.

**Keywords:** Ternary mixtures; benzene; 1-propanol; ethyl acetate; excess properties

### INTRODUCTION

A significant part of recent bibliography shows a great interest in determining the viscosity of multicomponent liquid systems. Such interest is justified by the extensive applications of this transport property in the industrial field (for example, it is necessary in flow, mass transfer or heat transfer calculations). The study of viscosities implies

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\*Corresponding author.

on the one hand the performance of accurate measurements and on the other hand the development of empirical or semiempirical methods for the correlation of data (Katti-Chaudhri, Nissan-Grunberg, McAllister) [1–3] and also of predictive methods such as those based on the Cao's molecular approaching [4, 5] and the group contribution methods [6, 7]. In his turn, the development of these methods clearly requires to have an adequately broad and precise database.

In this work densities and viscosities of the binary mixtures (benzene + 1-propanol, benzene + ethyl acetate and 1-propanol + ethyl acetate) and the ternary one (benzene + 1-propanol + ethyl acetate) are reported at the temperature 298.15 K. These data have been used to calculate the corresponding excess volumes, excess viscosities and excess Gibbs energies of activation for viscous flow ( $V^E$ ,  $\eta^E$  and  $G^{*E}$  respectively). In the discussion, the results will be compared to those found in literature at the same temperature [8–12] and an interpretation of the obtained values will be carried out in terms of the interactions existing in the systems.

## EXPERIMENTAL

The liquids used were: benzene (better than 99.5 mol%), provided by Dorwill, 1-propanol (better than 99.5%) supplied by Merck and ethyl acetate (better than 99.9 mol%), provided by EM Science. The purity of chemicals used was checked by measuring his density and was considered sufficient, so no further purification was attempted. 1-Propanol was dried over activated molecular sieve type 0.5 nm from Anedra.

Table I shows the experimental values of density and viscosity for the pure compounds at 298.15 K in comparison with published values [13]. The densities of the pure components and mixtures were measured by

TABLE I Densities  $\rho$ , and viscosities  $\eta$ , of pure compounds at  $T = 298.15$  K compared with literature data [13]

Component	$\rho/\text{g} \cdot \text{cm}^{-3}$		$\eta/\text{mPa} \cdot \text{s}$	
	Lit.	This paper	Lit.	This paper
Benzene	0.87360	0.87341	0.6028	0.5865
1-Propanol	0.79960	0.79965	1.9430	2.0221
Ethyl acetate	0.89455	0.89444	0.426	0.4062

means of a Mettler DA-310 vibrating tube densimeter automatically thermostated at  $298.15 \pm 0.01$  K. Calibration was carried out with de-ionized doubly distilled water and dry air. The precision of the density measurements is estimated to be  $\pm 0.00001 \text{ g} \cdot \text{cm}^{-3}$ .

The viscosity measurements were obtained with an Ubbelohde viscosimeter and a Schott-Geräte automatic measuring unity model AVS-440. Molar fractions of the mixtures were determined by mass. Details of the procedure have been described previously [14].

## RESULTS AND DISCUSSION

The excess properties were calculated from our measurements according to the following equations:

$$V^E = \sum_{i=1}^n x_i M_i \left( \frac{1}{\rho} - \frac{1}{\rho_i} \right) \quad (1)$$

$$\eta^E = \eta - \sum_{i=1}^n x_i \eta_i \quad (2)$$

$$G^{*E} = RT \left( \ln(\eta V) - \sum_{i=1}^n x_i \ln(\eta_i V_i) \right) \quad (3)$$

where  $x_i$ ,  $M_i$ ,  $\rho_i$ ,  $\eta_i$  and  $V_i$  designate molar fraction, molecular weight ( $\text{g} \cdot \text{mol}^{-1}$ ), density ( $\text{g} \cdot \text{cm}^{-3}$ ), absolute viscosity ( $\text{mPa} \cdot \text{s}$ ) and molar volume ( $\text{cm}^3 \cdot \text{mol}^{-1}$ ) of pure component  $i$ , respectively, and  $V = \sum_{i=1}^n x_i V_i$  is the molar volume of the mixture,  $R$  is the gas constant ( $\text{J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$ ),  $T$  is the absolute temperature and  $n$  is the number of components in the mixture. Symbols without subscript refer to the mixture, subscript 1 is used for benzene; 2 for 1-propanol and 3 for ethyl acetate.

The excess properties for binary mixtures were fitted to a Redlich-Kister polynomial equation [15]:

$$Y_{ij}^E = x_i x_j \sum_{p=0}^p A_p (x_i - x_j)^p \quad (4)$$

where  $Y_{ij}^E$  is  $V^E$ ,  $\eta^E$  or  $G^{*E}$ ,  $x_i$  denotes the molar fraction of component  $i$  in the  $i, j$  mixture with  $x_j = 1 - x_i$ , and  $A_p$  are adjustable parameters.

The excess volumes, excess viscosities and excess Gibbs energies of activation for viscous flow for the ternary mixture have been correlated to the Cibulka's equation [16]:

$$Y^E = Y_{\text{bin}}^E + x_1 x_2 (1 - x_1 - x_2) (B_1 + B_2 x_1 + B_3 x_2) \quad (5)$$

where

$$Y_{\text{bin}}^E = x_1 x_2 \sum_{p=0}^P A_{p,12} (x_1 - x_2)^p + x_1 x_3 \sum_{p=0}^P A_{p,13} (x_1 - x_3)^p + x_2 x_3 \sum_{p=0}^P A_{p,23} (x_2 - x_3)^p \quad (6)$$

where  $x_i$  is the molar fraction of the component  $i$  in the ternary data point, and  $B_1$ ,  $B_2$  and  $B_3$  are adjustable parameters.

The calculated values of the excess functions for the binary mixtures are presented in Tables II and III, and graphically shown in Figures 1–3. They are drawn taking into account that subscript  $i$  represents the first of the components of the corresponding mixture.

TABLE II Excess volumes  $V^E$  of the binary mixtures at  $T = 298.15 \text{ K}$

$x_1$	$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	$x_1$	$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	$x_1$	$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$
Benzene (1) + 1-Propanol (2)					
0.0690	-0.016	0.3972	0.051	0.7993	0.137
0.1016	-0.025	0.5009	0.088	0.8488	0.127
0.1403	-0.022	0.5531	0.103	0.8987	0.103
0.1869	-0.006	0.6930	0.141	0.9243	0.090
Benzene (1) + Ethyl acetate (3)					
0.0504	0.026	0.2922	0.100	0.7977	0.091
0.0719	0.034	0.3915	0.113	0.8525	0.075
0.0984	0.045	0.4966	0.122	0.8975	0.055
0.1476	0.059	0.5691	0.121	0.9250	0.042
0.1994	0.071	0.5961	0.119	0.9757	0.014
1-Propanol (2) + Ethyl acetate (3)					
0.0484	0.063	0.3958	0.235	0.8472	0.099
0.0700	0.081	0.4479	0.242	0.9211	0.054
0.0938	0.103	0.4991	0.242	0.9501	0.034
0.2033	0.173	0.5489	0.237		
0.2984	0.212	0.7010	0.191		

TABLE III Excess viscosities  $\eta^E$  and excess Gibbs energies of activation for viscous flow  $G^{*E}$  of the binary mixtures at  $T = 298.15\text{ K}$ 

$x_1$	$\eta^E/mPa \cdot s$	$G^{*E}/J \cdot mol^{-1}$	$x_1$	$\eta^E/mPa \cdot s$	$G^{*E}/J \cdot mol^{-1}$	$x_1$	$\eta^E/mPa \cdot s$	$G^{*E}/J \cdot mol^{-1}$
	Benzene (1) + 1-Propanol (2)							
0.0480	-0.0914	-120.2	0.4486	-0.2624	-624.1	0.8987	-0.0745	-288.5
0.0690	-0.1272	-174.0	0.5009	-0.2558	-652.5	0.9243	-0.0563	-221.7
0.1016	-0.1627	-234.4	0.5531	-0.2469	-674.8	0.9506	-0.0365	-145.6
0.1403	-0.1953	-298.5	0.5932	-0.2347	-673.6	0.9735	-0.0203	-82.3
0.2983	-0.2751	-534.2	0.6930	-0.1903	-609.9			
0.3972	-0.2671	-592.4	0.8488	-0.1100	-411.0			
	Benzene (1) + Ethyl acetate (3)							
0.0504	-0.0017	-8.9	0.1994	-0.0093	-49.5	0.5961	-0.0308	-150.1
0.0719	-0.0026	-13.8	0.2922	-0.0166	-87.8	0.8975	-0.0209	-91.0
0.0984	-0.0036	-19.0	0.4561	-0.0265	-134.6	0.9250	-0.0166	-71.2
0.1476	-0.0057	-30.4	0.4966	-0.0275	-138.1			
	1-Propanol (2) + Ethyl acetate (3)							
0.0194	-0.0121	-70.5	0.3958	-0.2394	-899.9	0.5968	-0.3633	-1013.3
0.0938	-0.0532	-286.3	0.4479	-0.2760	-967.8	0.7010	-0.4249	-1002.9
0.1457	-0.0872	-447.1	0.4991	-0.3068	-996.8	0.9000	-0.3500	-551.7
0.2033	-0.1187	-568.3	0.5489	-0.3345	-1005.3	0.9211	-0.2977	-444.4

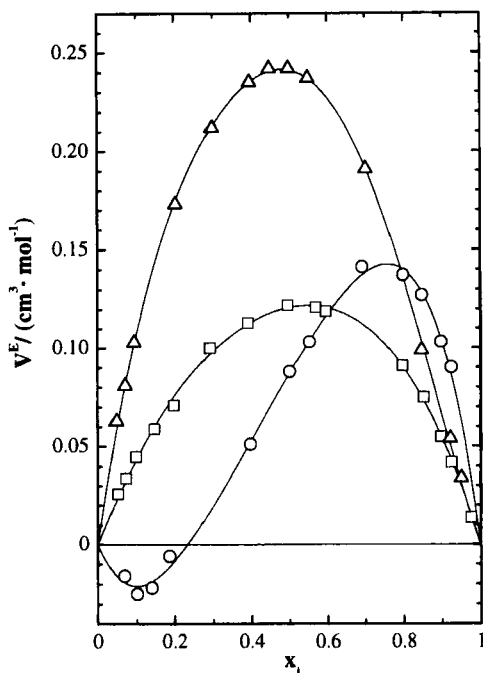


FIGURE 1 Excess volumes,  $V^E$ , of binary mixtures at 298.15 K as a function of molar fraction  $x_1$ : benzene (1)+1-propanol (2) (o); benzene (1)+ethyl acetate (3) (□); 1-propanol (2)+ethyl acetate (3) (Δ).

The calculated values of the excess functions for the ternary system are given in Tables IV and V. Tridimensional surfaces of  $V^E$ ,  $\eta^E$  and  $G^{*E}$  calculated from Cibulka's equation (5) have been plotted in Figures 4–6. The isolines at constant values of  $V^E$ ,  $\eta^E$  and  $G^{*E}$  have been drawn in Figures 7–9. The coefficients  $A_p$  (Eq. (4)),  $B_p$  (Eq. (5)) and the standard deviations  $\sigma$  obtained by the least squares method are given in Table VI.

The behaviour exhibited by both the ternary system and the corresponding binary mixtures can be interpreted through the interactions present in the pure liquids and in the mixtures.

In that respect and first of all, the main characteristics of the pure compounds to be taken into account are: (1) the orientational order of aromatic hydrocarbons [8], (2) the hydrogen bond associated structure and the dipole–dipole interactions existing between alcohol

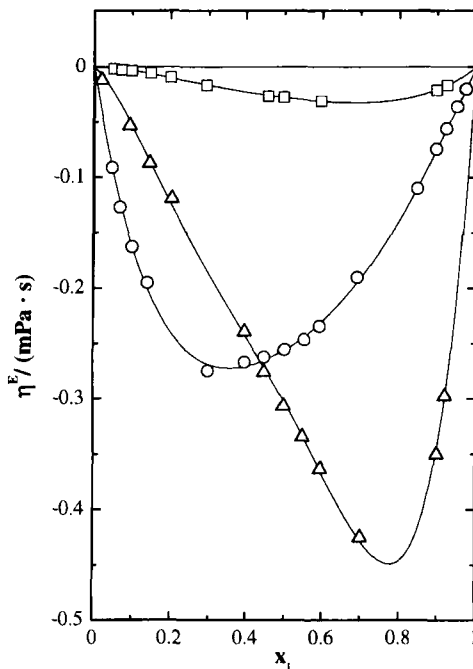


FIGURE 2 Excess viscosities,  $\eta^E$ , of binary mixtures at 298.15 K as a function of molar fraction  $x_1$ : benzene (1) + 1-propanol (2) ( $\circ$ ); benzene (1) + ethyl acetate (3) ( $\square$ ); 1-propanol (2) + ethyl acetate (3) ( $\Delta$ ).

monomers and multimers in the 1-propanol [8], and (3) the dipole–dipole interactions established between carboxylic groups of the ethyl acetate [9].

With regard to the mixtures, a decrease of these effects occurs and yields an expansion or positive contribution to  $V^E$ . On the other hand, several interactions appear as consequence of the mixing process. They are the following: (1) electron donor-acceptor interactions between alcohols and aromatic hydrocarbons (donor) [8], (2) formation of  $n$ - $\pi$  complexes between free electrons of the carboxylic group and the aromatic ring [9], (3) induced dipole–dipole interactions between benzene and ethyl acetate [9, 10], and finally (4) dipole–dipole interactions between 1-propanol and ethyl acetate. All of them give rise to negative contributions to excess volume. The effect of interactions



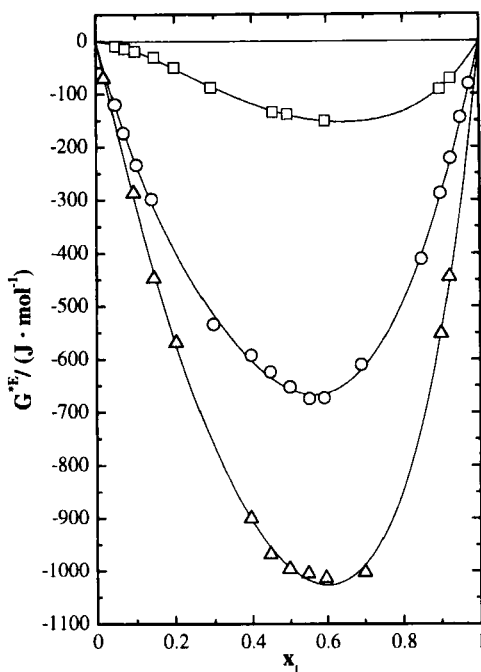


FIGURE 3 Excess Gibbs energies of activation for viscous flow,  $G^{*E}$ , of binary mixtures at 298.15 K as a function of molar fraction  $x_1$ : benzene (1) + 1-propanol (2) (o); benzene (1) + ethyl acetate (3) (□); 1-propanol (2) + ethyl acetate (3) (Δ).

weakening or formation on excess viscosity and  $G^{*E}$  will be opposite to that described for  $V^E$  as several authors have pointed out [17, 18].

From the analysis of the excess properties studied for the binary mixtures is remarkable the fact that a region of negative  $V^E$  is obtained for the mixture (benzene + 1-propanol) just in the zone with a high concentration in alcohol. This could be explained considering that the planar structure of benzene accommodates itself among the alcohol multimers. When the concentration of benzene rises, the effect of breaking of association in 1-propanol prevails and excess volume rises becoming positive. This same region of negative  $V^E$  has been observed by Mato and Coca [11] although in two other papers [8, 12] does not appear. Nevertheless, both the maximum value and the maximum position obtained in our work coincide with those reported in the three cited publications.

The other binary mixtures show always positive  $V^E$  values, higher in the case of the mixture (1-propanol + ethyl acetate). For this last

TABLE IV Excess volumes  $V^E$  of the ternary mixture benzene(1) + 1-propanol (2) + ethyl acetate (3) at  $T = 298.15$  K

$x_1$	$x_2$	$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	$x_1$	$x_2$	$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$
0.0050	0.0957	0.103	0.1886	0.7616	0.047
0.0123	0.2345	0.196	0.0454	0.1055	0.118
0.0173	0.3311	0.223	0.0594	0.1380	0.142
0.0225	0.4301	0.252	0.0904	0.2102	0.187
0.0272	0.5191	0.240	0.1209	0.2811	0.212
0.0322	0.6154	0.217	0.1489	0.3461	0.215
0.0373	0.7121	0.179	0.1790	0.4162	0.208
0.0474	0.9059	0.036	0.2103	0.4889	0.187
0.0203	0.1816	0.160	0.2250	0.5231	0.172
0.0302	0.2707	0.204	0.2550	0.5927	0.127
0.0398	0.3565	0.221	0.2859	0.6647	0.065
0.0513	0.4600	0.227	0.1029	0.1527	0.152
0.0598	0.5355	0.216	0.1401	0.2081	0.181
0.0702	0.6288	0.185	0.1796	0.2667	0.200
0.0854	0.7656	0.107	0.2210	0.3282	0.216
0.0954	0.8547	0.030	0.2627	0.3900	0.201
0.0200	0.0807	0.091	0.3010	0.4469	0.173
0.0500	0.2020	0.178	0.3217	0.4776	0.155
0.0739	0.2982	0.221	0.3623	0.5380	0.115
0.0897	0.3620	0.223	0.3822	0.5675	0.088
0.1088	0.4394	0.220	0.0271	0.0278	0.041
0.1294	0.5223	0.200	0.0993	0.1017	0.127
0.1488	0.6007	0.168	0.1476	0.1512	0.165
0.1589	0.6414	0.150	0.1987	0.2035	0.192
0.1783	0.7201	0.087	0.2347	0.2405	0.210
0.2940	0.3012	0.215	0.4418	0.1081	0.167
0.3471	0.3556	0.180	0.5226	0.1279	0.166
0.3970	0.4068	0.158	0.5636	0.1379	0.166
0.4199	0.4302	0.149	0.5799	0.1419	0.164
0.4688	0.4803	0.109	0.7228	0.1769	0.142
0.0916	0.0604	0.094	0.7681	0.1880	0.132
0.1516	0.1000	0.138	0.1832	0.0198	0.082
0.2116	0.1395	0.170	0.2774	0.0300	0.115
0.2970	0.1959	0.198	0.3633	0.0394	0.137
0.3316	0.2187	0.197	0.4498	0.0487	0.148
0.3916	0.2583	0.191	0.5428	0.0588	0.149
0.4510	0.2974	0.177	0.5841	0.0633	0.149
0.4840	0.3192	0.166	0.6768	0.0733	0.139
0.5426	0.3578	0.100	0.7213	0.0781	0.128
0.5724	0.3775	0.126	0.7658	0.0829	0.116
0.2810	0.1200	0.168	0.1386	0.0073	0.059
0.3527	0.1506	0.174	0.2372	0.0125	0.091
0.4215	0.1800	0.186	0.3304	0.0174	0.109
0.4900	0.2092	0.178	0.4248	0.0223	0.121
0.5238	0.2236	0.174	0.5215	0.0274	0.126
0.5962	0.2546	0.152	0.5733	0.0301	0.126
0.6654	0.2841	0.133	0.6578	0.0346	0.122
0.0358	0.0088	0.024	0.8074	0.0424	0.095
0.2028	0.0496	0.114	0.8557	0.0450	0.084
0.2835	0.0694	0.140	0.8990	0.0472	0.071
0.3607	0.0883	0.156			

TABLE V Excess viscosities  $\eta^E$  and excess Gibbs energies of activation for viscous flow  $G^{*E}$  of the ternary mixture benzene(1) + 1-propanol(2) + ethyl acetate(3) at  $T = 298.15$  K

$x_1$	$x_2$	$\eta^E/\text{mPa}\cdot\text{s}$	$G^{*E}/\text{J}\cdot\text{mol}^{-1}$	$x_1$	$x_2$	$\eta^E/\text{mPa}\cdot\text{s}$	$G^{*E}/\text{J}\cdot\text{mol}^{-1}$
0.0050	0.0957	-0.0462	-257.9	0.1783	0.7201	-0.3444	-723.7
0.0123	0.2345	-0.1299	-619.8	0.1886	0.7616	-0.3128	-598.8
0.0173	0.3311	-0.1951	-827.2	0.0454	0.1055	-0.0629	-344.8
0.0225	0.4301	-0.2668	-993.0	0.0594	0.1380	-0.0779	-409.1
0.0272	0.5191	-0.3241	-1056.8	0.0904	0.2102	-0.1286	-618.7
0.0322	0.6154	-0.3725	-1034.7	0.1209	0.2811	-0.1762	-770.1
0.0373	0.7121	-0.4136	-969.8	0.1489	0.3461	-0.2184	-867.1
0.0398	0.7598	-0.4194	-897.7	0.1790	0.4162	-0.2625	-932.4
0.0445	0.8515	-0.3601	-631.4	0.2103	0.4889	-0.3027	-948.9
0.0049	0.0438	-0.0171	-101.2	0.2250	0.5231	-0.3193	-941.2
0.0157	0.1404	-0.0749	-398.4	0.2550	0.5927	-0.3293	-844.5
0.0203	0.1816	-0.1018	-516.9	0.2859	0.6647	-0.2962	-645.1
0.0302	0.2707	-0.1597	-728.9	0.0417	0.0619	-0.0328	-188.1
0.0398	0.3565	-0.2140	-871.6	0.1029	0.1527	-0.0925	-472.0
0.0513	0.4600	-0.2847	-1001.5	0.1401	0.2081	-0.1301	-616.4
0.0598	0.5355	-0.3341	-1046.5	0.1796	0.2667	-0.1709	-744.2
0.0702	0.6288	-0.3670	-973.2	0.2210	0.3282	-0.2146	-850.1
0.0200	0.0807	-0.0427	-241.6	0.2627	0.3900	-0.2322	-816.4
0.0500	0.2020	-0.1173	-576.1	0.3010	0.4469	-0.2625	-833.4
0.0739	0.2982	-0.1910	-836.5	0.3217	0.4776	-0.2793	-838.4
0.0897	0.3620	-0.2281	-909.7	0.3623	0.5380	-0.2900	-768.8
0.1088	0.4394	-0.2746	-972.1	0.3822	0.5675	-0.2858	-709.6
0.1294	0.5223	-0.3252	-1006.4	0.0271	0.0278	-0.0122	-72.8
0.1488	0.6007	-0.3615	-973.5	0.0993	0.1017	-0.0649	-351.0
0.1589	0.6414	-0.3699	-920.7	0.1476	0.1512	-0.0986	-498.1
0.1987	0.2035	-0.1346	-629.3	0.6654	0.2841	-0.1959	-665.4
0.2347	0.2405	-0.1522	-666.7	0.0358	0.0088	-0.0068	-25.1
0.2940	0.3012	-0.2009	-801.2	0.2028	0.0496	-0.0423	-224.2
0.3471	0.3556	-0.2324	-839.7	0.2835	0.0694	-0.0638	-324.7
0.3970	0.4068	-0.2528	-823.9	0.3607	0.0883	-0.0798	-380.3
0.4199	0.4302	-0.2412	-811.0	0.4418	0.1081	-0.0958	-439.7
0.4688	0.4803	-0.2677	-744.2	0.5226	0.1279	-0.1115	-484.0
0.0916	0.0604	-0.0384	-216.5	0.5636	0.1379	-0.1201	-500.2
0.1516	0.1000	-0.0691	-368.3	0.5799	0.1419	-0.1227	-500.1
0.2116	0.1395	-0.1000	-502.4	0.7228	0.1769	-0.1423	-528.2
0.2970	0.1959	-0.1506	-694.5	0.7681	0.1880	-0.1455	-519.0
0.3316	0.2187	-0.1523	-666.1	0.1832	0.0198	-0.0223	-126.2
0.3916	0.2583	-0.1787	-725.5	0.2774	0.0300	-0.0368	-201.1
0.4510	0.2974	-0.1998	-748.8	0.3633	0.0394	-0.0478	-252.0
0.4840	0.3192	-0.2109	-755.2	0.4498	0.0487	-0.0577	-292.4
0.5426	0.3578	-0.2216	-726.4	0.5428	0.0588	-0.0658	-319.3
0.5724	0.3775	-0.2325	-730.6	0.5841	0.0633	-0.0690	-327.9
0.0741	0.0316	-0.0224	-122.8	0.6768	0.0733	-0.0768	-348.9
0.1434	0.0612	-0.0473	-253.3	0.7213	0.0781	-0.0800	-355.3
0.2100	0.0897	-0.0672	-343.4	0.7658	0.0829	-0.0819	-355.3
0.2810	0.1200	-0.0933	-448.0	0.8576	0.0929	-0.0850	-350.8
0.3527	0.1506	-0.1126	-514.6	0.1386	0.0073	-0.0111	-63.8
0.4215	0.1800	-0.1381	-596.8	0.2372	0.0125	-0.0218	-121.9
0.4900	0.2092	-0.1577	-638.8	0.3304	0.0174	-0.0299	-161.8

TABLE V (Continued)

$x_1$	$x_2$	$\eta^E/mPa \cdot s$	$G^{*E}/J \cdot mol^{-1}$	$x_1$	$x_2$	$\eta^E/mPa \cdot s$	$G^{*E}/J \cdot mol^{-1}$
0.5238	0.2236	-0.1668	-652.6	0.4248	0.0223	-0.0393	-205.8
0.5962	0.2546	-0.1826	-662.2	0.5215	0.0274	-0.0453	-227.8
0.5733	0.0301	-0.0482	-237.6	0.8557	0.0450	-0.0463	-199.7
0.6578	0.0346	-0.0496	-235.1	0.8990	0.0472	-0.0412	-173.7
0.8074	0.0424	-0.0485	-214.5				

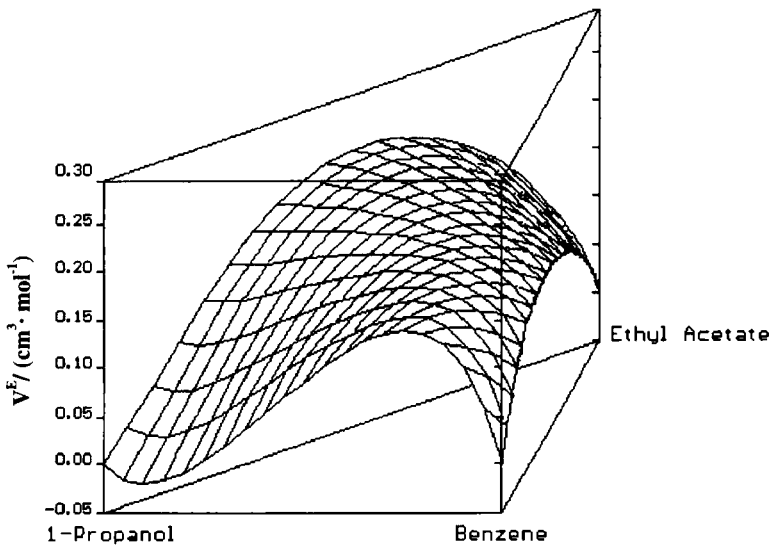


FIGURE 4 Tridimensional surface of  $V^E$  at 298.15 K for the ternary system benzene (1) + 1-propanol (2) + ethyl acetate (3) correlated with the Cibulka's equation (5).

mixture two references have been found in literature [9, 10], and our values are in good agreement with theirs.

The  $\eta^E$  and  $G^{*E}$  are negative for all of the studied mixtures, corresponding the more negative to the mixture (1-propanol + ethyl acetate). The interpretation of these results can be made in the same manner considered for  $V^E$ .

In the ternary system,  $V^E$  is positive while both  $\eta^E$  and  $G^{*E}$  are negative over the entire composition range. The highest values of excess volumes are reached in zones with high concentration of ethyl acetate and the smallest values in regions rich in 1-propanol, where the

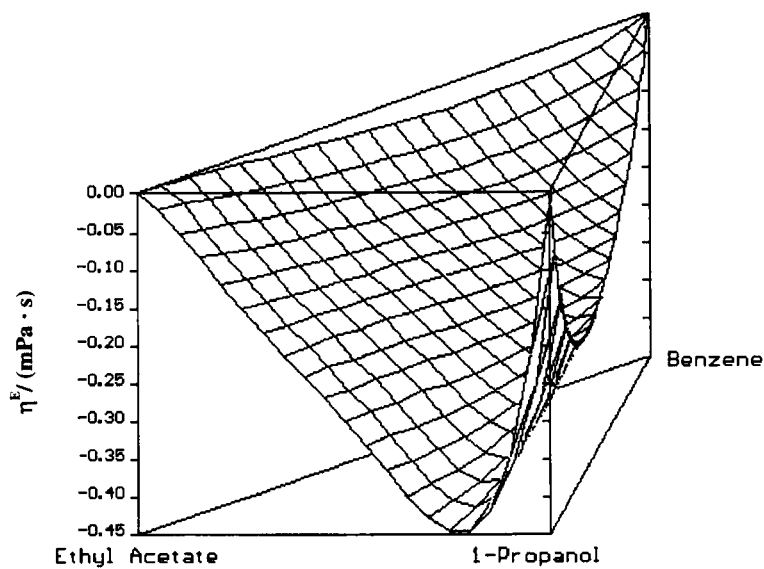


FIGURE 5 Tridimensional surface of  $\eta^E$  at 298.15 K for the ternary system benzene (1) + 1-propanol (2) + ethyl acetate (3) correlated with the Cibulka's equation (5).

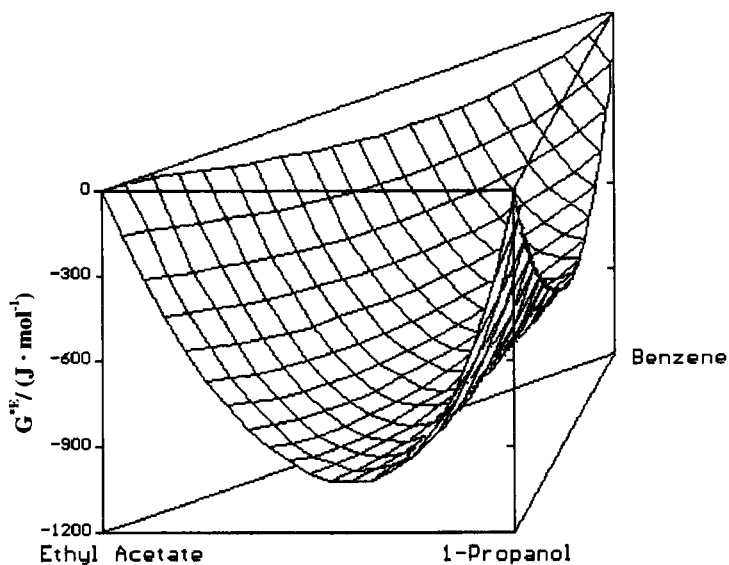


FIGURE 6 Tridimensional surface of  $G^{*E}$  at 298.15 K for the ternary system benzene (1) + 1-propanol (2) + ethyl acetate (3) correlated with the Cibulka's equation (5).

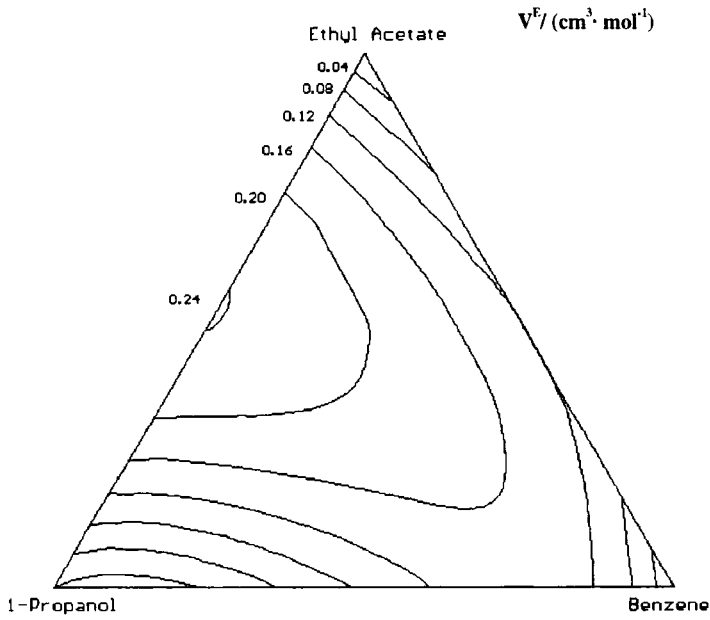


FIGURE 7 Isolines at constant  $V^E$  at 298.15 K for the ternary system benzene (1) + 1-propanol (2) + ethyl acetate (3) correlated with the Cibulka's equation (5).

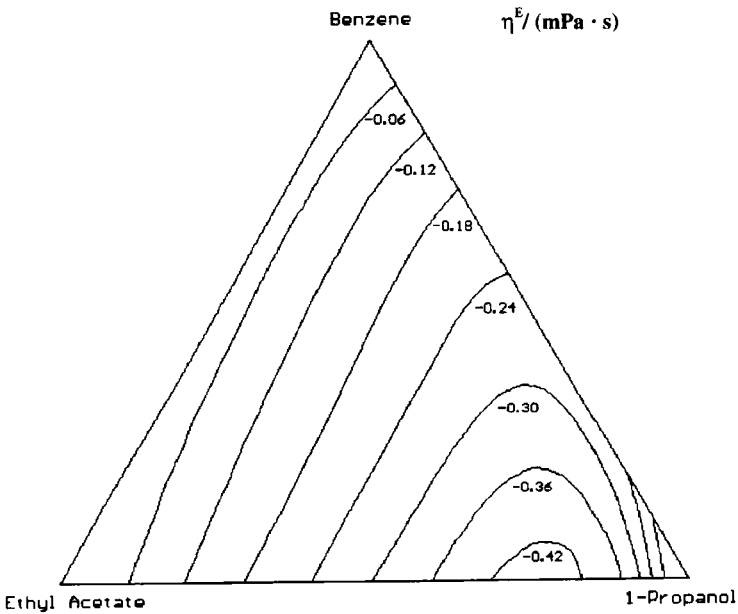


FIGURE 8 Isolines at constant  $\eta^E$  at 298.15 K for the ternary system benzene (1) + 1-propanol (2) + ethyl acetate (3) correlated with the Cibulka's equation (5).

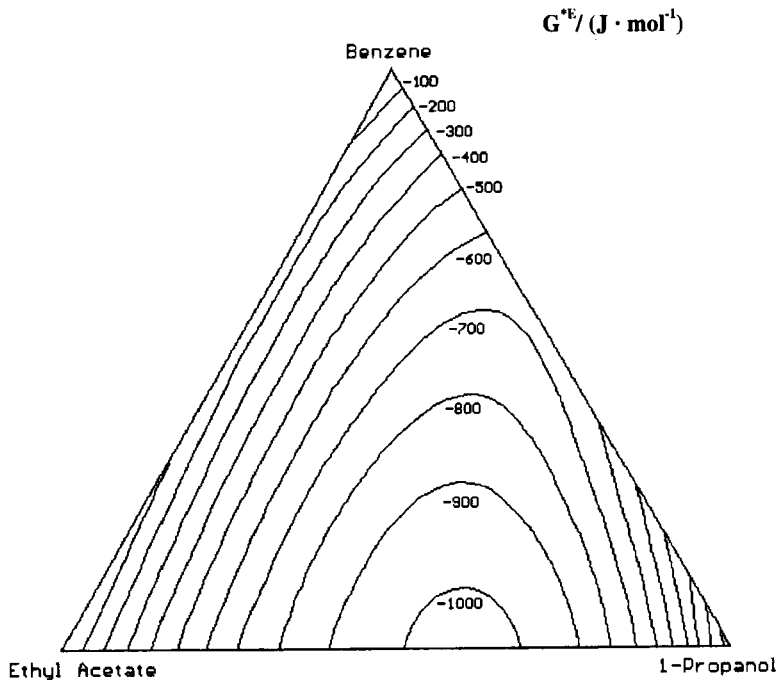


FIGURE 9 Isolines at constant  $G^E$  at 298.15 K for the ternary system benzene (1) + 1-propanol (2) + ethyl acetate (3) correlated with the Cibulka's equation (5).

TABLE VI Coefficients  $A_p$  of Eq. (4) and  $B_p$  of Eq. (5) together with standard deviations  $\sigma(Y^E)$  determined by the least squares method

Function	$A_0$	$A_1$	$A_2$	$A_3$	$\sigma$
Benzene (1) + 1-propanol (2)					
$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	0.349	0.651	0.171	0.336	0.004
$\eta^E/\text{mPa} \cdot \text{s}$	-1.0250	0.4163	-0.4417	0.3033	0.0036
$G^E/\text{J} \cdot \text{mol}^{-1}$	-2625.2	-634.9	-409.7	411.0	9.2
Benzene (1) + ethyl acetate (3)					
$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	0.484	0.077	0.097	-0.007	0.002
$\eta^E/\text{mPa} \cdot \text{s}$	-0.1115	-0.0838	-0.0335	-0.0529	0.0053
$G^E/\text{J} \cdot \text{mol}^{-1}$	-560.6	-334.2	-55.4	-237.7	2.7
1-Propanol (2) + ethyl acetate (3)					
$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	0.964	-0.076	0.002	-0.339	0.003
$\eta^E/\text{mPa} \cdot \text{s}$	-1.2122	-1.2369	-1.5774	-1.2378	0.0045
$G^E/\text{J} \cdot \text{mol}^{-1}$	-3976.1	-1234.6	-1243.8	-612.5	12.0
Function	$B_1$	$B_2$	$B_3$	$\sigma$	
Benzene (1) + 1-Propanol (2) + ethyl acetate (3)					
$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	0.019	-1.621	0.976	0.007	
$\eta^E/\text{mPa} \cdot \text{s}$	0.1313	0.2867	3.3963	0.0070	
$G^E/\text{J} \cdot \text{mol}^{-1}$	-2580.3	4013.0	1488.2	21.3	

interstitial accommodation is favoured (changes in free volumes). In these same regions  $\eta^E$  and  $G^{*E}$  show the most negative values.

However the existence in the mixtures of the interactions mentioned before, it can be seen that both  $V^E$  and  $\eta^E$  ( $G^{*E}$ ) results clearly indicate that the behaviour of the binary and ternary systems is mainly determined by the breaking of association in the alcohol.

### Acknowledgements

The authors are grateful for financial assistance from Universidad de Zaragoza (Proyecto UZ: 96-232-47) and Diputación General de Aragón (Proyecto PCB-0894).

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