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Enthalpies of Mixing, Densities, and Refractive Indices for Binary Mixtures of (Anisole or Phenetole) + Three Aryl Alcohols at 308.15 K and at Atmospheric Pressure

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Excess molar enthalpies, H_m^E, excess molar volumes, V_m^E, and refractive indices, n_D, for the binary mixtures anisole or phenetole + benzyl alcohol, + 2-phenylethanol, and + 3-phenyl-1-propanol have been determined at 308.15 K and at atmospheric pressure using an LKB flow microcalorimeter, an Anton Paar density meter, and an Abbe refractometer, respectively. A Redlich−Kister-type equation was used to correlate the experimental data and to derive the adjustable parameters and the standard deviations. The experimental and calculated quantities were used to study the nature of the mixing behavior between mixture components.

Introduction

Excess and bulk properties are fundamentally important in understanding the intermolecular interactions between dissimilar molecules and in developing the thermodynamic models.

In earlier papers, we have studied the thermodynamic and bulk properties of binary mixtures containing some aryl alcohols,[1−3] and in continuation of this research, we now present in this work additional data on density, heat of mixing, and refractive indices (for the sodium D line) of binary mixtures containing anisole or phenetole + benzyl alcohol, + 2-phenyl ethanol, and + 3-phenyl-1-propanol over the entire range of composition at 308.15 K and at atmospheric pressure. Anisole or phenetole is component 1.

The derived properties (excess molar enthalpies, H_m^E, excess molar volumes, V_m^E, and deviations in molar refraction, ΔR), in combination with other mixing properties, provide valuable information for qualitatively analyzing the molecular interactions between molecules.

The experimental data have been fitted to the Redlich−Kister polynomial to derive the adjustable parameters and to estimate the standard deviations. We could not find any information about these mixtures in the available literature.

Experimental Section

Anisole and phenetole were purchased from Aldrich (purity >99% for both products), benzyl alcohol (purity 99.8%) was from Riedel-de-Haën, and 2-phenyl ethanol (purity 99%) and 3-phenyl-1-propanol (purity 99%) were from Fluka. The purities of the products were checked on a Hewlett-Packard gas chromatograph model 5890 by using an HP (cross-linked 5% ME siloxane) capillary column, and the stated purities were confirmed. The liquids were used without further purification, other than being kept over molecular sieves (Union Carbide, type 4A 1/16 in. pellets) to remove water. For measurements, the liquids were also degassed by ultrasound (ultrasonic bath, type 400, Hellma, Milan, Italy) to avoid bubble formation. Experimental values of densities, ρ, and refractive indices, n_D, for the pure liquids were compared with the data found in the literature,[2,4−8] as shown in Table 1.

Table 1. Densities, ρ, and Refractive Indices of Pure Components at 308.15 K and Comparison with Literature Values

		ρ/g·cm^{-3}		n_D	
component	T/K	this paper	lit.	this paper	lit.
anisole	298.15	0.98930	0.98931[a]	1.5153	
	308.15	0.97971	0.9798[b]	1.5095	
phenetole	298.15	0.9690		1.5058	
	308.15	0.95086		1.5002	
benzyl alcohol	298.15	1.04165	1.0416[c]	1.5377	1.5378[c]
	308.15	1.03364	1.0330[d]	1.5350	
2-phenylethanol	298.15	1.20435	1.20439[e]	1.5321	
	308.15	1.00875	1.00915[e]	1.5269	
3-phenyl-1-propanol	298.15	1.03269		1.5259	
	308.15	0.99101	0.99081[f]	1.5214	

[a] Reference 4. [b] Reference 5. [c] Reference 6. [d] Reference 7. [e] Reference 8. [f] Reference 2.

Calorimetric Measurements. The excess molar enthalpies, H_m^E, were measured by a flow isothermal microcalorimeter (LKB, model 2107, Producer AB, Bromma, Sweden), which consists of a mixing cell, a reference cell, a thermostatic water bath, a data acquisition unit, and two liquid burets (pumps) (ABU Radiometer, Copenhagen, Denmark). The water bath controls the temperature of the heat sink where the mixing cell, reference cell, and thermoelectric sensors were located. The temperature of the bath was controlled to within ±0.01 K. The pure components were pumped into the mixing cell at selected flow rates using the two pumps. The mole fractions, x_1, of component 1 in the mixed stream were calculated from the densities and volumetric flow rates of components 1, the total flow rate being 0.4 cm^3·min^{-1} and the uncertainties in mole fraction kept to about 2×10^{-4}. Because the residence time in the mixing chamber, guaranteeing com-

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Table 2. Excess Molar Enthalpies, H_m^E, for Binary Mixtures Containing Anisole or Phenetole + Three Aryl Alcohols at 308.15 K

x_1	H_m^E/J·mol^{-1}	x_1	H_m^E/J·mol^{-1}	x_1	H_m^E/J·mol^{-1}	x_1	H_m^E/J·mol^{-1}	x_1	H_m^E/J·mol^{-1}	x_1	H_m^E/J·mol^{-1}
Anisole (1) + Benzyl Alcohol (2)											
0.0380	92.1	0.1364	367.0	0.3216	761.5	0.5872	1040.8	0.7913	1003.4	0.9191	610.1
0.0732	192.8	0.1916	507.9	0.3871	849.0	0.6547	1064.3	0.8505	881.6	0.9578	367.0
0.1059	274.1	0.2401	619.9	0.4866	968.8	0.7398	1053.9	0.8835	775.7		
Anisole (1) + 2-Phenyl Ethanol (2)											
0.0437	95.1	0.1546	396.6	0.3543	867.7	0.6221	1124.8	0.8145	1049.1	0.9294	613.3
0.0838	200.1	0.2153	559.2	0.4224	952.0	0.6870	1142.9	0.8682	907.7	0.9634	361.3
0.1206	304.7	0.2678	688.0	0.5232	1045.7	0.7670	1115.4	0.9006	775.7		
Anisole (1) + 3-Phenyl-1-propanol(2)											
0.0493	126.6	0.1719	446.8	0.3838	855.7	0.6514	1091.3	0.8328	1000.6	0.9373	555.2
0.0940	246.7	0.2374	607.6	0.4536	937.3	0.7136	1108.8	0.8770	868.1	0.9676	325.3
0.1347	361.1	0.2934	717.3	0.5547	1029.3	0.7889	1079.1	0.9088	733.9		
Phenetole (1) + Benzyl Alcohol (2)											
0.0328	103.3	0.1195	357.5	0.2893	739.3	0.5499	1056.1	0.7651	1083.0	0.9070	707.0
0.0636	190.1	0.1691	495.0	0.3517	856.1	0.6196	1095.9	0.8301	971.3	0.9513	433.2
0.0924	283.3	0.2135	615.8	0.4488	971.8	0.7095	1113.9	0.8669	874.5		
Phenetole (1) + 2-Phenylethanol (2)											
0.0378	112.8	0.1358	409.8	0.3203	805.7	0.5857	1122.6	0.7904	1102.5	0.9188	674.4
0.0728	223.3	0.1907	564.3	0.3858	911.5	0.6534	1149.2	0.8498	971.4	0.9577	403.6
0.1054	322.8	0.2391	666.9	0.4852	1024.9	0.7388	1142.7	0.8829	853.2		
Phenetole (1) + 3-Phenyl-1-propanol (2)											
0.0427	137.8	0.1513	476.2	0.3485	806.2	0.6162	1074.1	0.8106	1065.1	0.9277	634.8
0.0818	264.2	0.2110	610.6	0.4162	886.1	0.6815	1115.4	0.8596	942.3	0.9625	374.7
0.1180	368.0	0.2629	700.0	0.5169	989.0	0.7625	1116.4	0.8954	812.9		

Table 3. Densities, ρ, Excess Molar Volumes, V_m^E, and Refractive Indices, n_D, for Binary Mixtures Containing Anisole or Phenetole + Three Aryl Alcohols at 308.15 K

x_1	ρ g·cm^{-3}	V_m^E cm^3·mol^{-1}	n_D	x_1	ρ g·cm^{-3}	V_m^E cm^3·mol^{-1}	n_D	x_1	ρ g·cm^{-3}	V_m^E cm^3·mol^{-1}	n_D	x_1	ρ g·cm^{-3}	V_m^E cm^3·mol^{-1}	n_D
Anisole (1) + Benzyl Alcohol (2)															
0.0519	1.03085	−0.016	1.5335	0.2883	1.01800	−0.053	1.5270	0.5916	1.00147	−0.046	1.5193	0.9039	0.98473	−0.010	1.5118
0.0898	1.02881	−0.025	1.5324	0.3766	1.01319	−0.057	1.5247	0.6747	0.99696	−0.037	1.5173	0.9448	0.98258	−0.005	1.5108
0.1854	1.02362	−0.043	1.5298	0.4911	1.00694	−0.055	1.5218	0.7745	0.99159	−0.025	1.5149	0.9723	0.98114	−0.003	1.5102
Anisole (1) + 2-Phenylethanol (2)															
0.0521	1.00744	−0.009	1.5260	0.3153	1.00043	−0.031	1.5217	0.6170	0.99155	−0.008	1.5165	0.8110	0.9855	0.012	1.5127
0.1223	1.00563	−0.019	1.5248	0.4172	0.99753	−0.028	1.5200	0.7479	0.98750	0.007	1.5140	0.9073	0.98255	0.012	1.5106
0.2121	1.00326	−0.027	1.5233	0.5245	0.99436	−0.019	1.5182	0.7673	0.98690	0.009	1.5136	0.9513	0.98120	0.008	1.5096
Anisole (1) + 3-Phenyl-1-propanol (2)															
0.0579	0.99046	−0.003	1.5208	0.3840	0.98728	−0.009	1.5173	0.6495	0.98407	0.020	1.5142	0.8652	0.98126	0.033	1.5113
0.1201	0.98991	−0.007	1.5201	0.44751	0.98657	−0.004	1.5166	0.7477	0.98278	0.031	1.5129	0.91296	0.98067	0.027	1.5108
0.2636	0.98855	−0.013	1.5186	0.5551	0.98527	0.008	1.5154	0.8274	0.98173	0.035	1.5118	0.9588	0.98014	0.016	1.5101
Phenetole (1) + Benzyl Alcohol (2)															
0.0642	1.02761	−0.041	1.5322	0.1751	1.01738	−0.092	1.5275	0.4598	0.99251	−0.139	1.5172	0.7572	0.96865	−0.086	1.5071
0.0826	1.02586	−0.047	1.5313	0.2585	1.00989	−0.117	1.5244	0.5440	0.98553	−0.133	1.5142	0.8621	0.96077	−0.051	1.5039
0.1663	1.01818	−0.088	1.5279	0.3363	1.00306	−0.132	1.5216	0.6369	0.97804	−0.117	1.5111	0.9361	0.95537	−0.024	1.5018
Phenetole (1) + 2-Phenylethanol (2)															
0.0656	1.00496	−0.024	1.5252	0.3803	0.98652	−0.072	1.5166	0.5904	0.97422	−0.059	1.5108	0.8881	0.95700	0.001	1.5029
0.1079	1.00250	−0.037	1.5240	0.4826	0.98037	−0.070	1.5137	0.6819	0.96887	−0.041	1.5083	0.9469	0.95374	0.005	1.5014
0.1726	0.99871	−0.052	1.5223	0.5243	0.97809	−0.067	1.5126	0.7929	0.96244	−0.018	1.5054	0.9881	0.95150	0.003	1.5005
0.3041	0.99099	−0.069	1.5187												
Phenetole (1) + 3-Phenyl-1-propanol (2)															
0.0636	0.98877	−0.022	1.5203	0.3100	0.97965	−0.071	1.5153	0.6168	0.96729	−0.056	1.5086	0.8977	0.9552	0.003	1.5023
0.1067	0.98722	−0.034	1.5194	0.4170	0.97546	−0.075	1.5130	0.7074	0.96343	−0.037	1.5066	0.9488	0.95299	0.005	1.5012
0.2164	0.98320	−0.059	1.5173	0.5150	0.97151	−0.070	1.5109	0.7975	0.95953	−0.016	1.5045	0.9596	0.95253	0.005	1.5010

plete mixing, may vary considerably with the type of mixture, the optimum flow rate has to be determined experimentally. Details and operating procedure of the apparatus have been reported elsewere.[9,10] The uncertainty of the reported H_m^E values (about 1%) was verified on the three standard systems, cyclohexane + hexane, benzene + hexane, and methanol + water at 298.15 K, that are known in the literature.[11] Agreement with literature data was better than 0.5% at the maximum of the thermal effect.

Density Measurements. Excess molar volumes, V_m^E, have been determined from density measurements with the help of a digital density meter (Anton Paar, model DMA 60, Graz, Austria) equipped with a measuring cell (Anton Paar, type 602) whose operating procedure has been described elsewhere.[12]

All measurements have been determined at a constant temperature using an external ultrathermostat bath circulator (Heto, type 01 DTB 623, Birkeròd, Denmark, precision ±0.005 K), and temperatures have been detected by a digital thermometer (Anton Paar, type CTK 100). Mole fractions, x_1, of anisole or phenetole were obtained by mass using a digital balance (Mettler, model AE 160, Switzerland) with an accuracy of ±0.0001 g. All masses have been corrected for buoyancy and evaporation of components. All molar quantities have been based on the relative atomic mass table by IUPAC.[13]

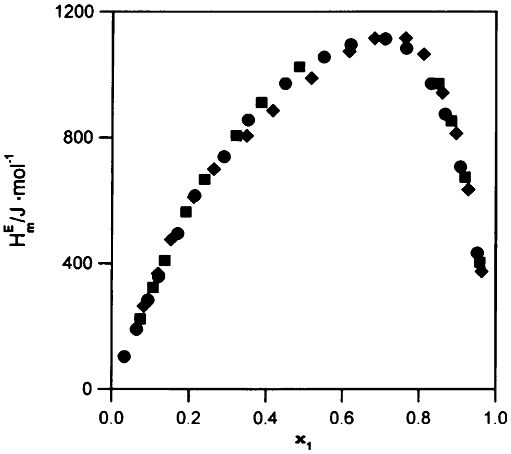

Figure 1. Experimental excess molar enthalpies, H^E_m, for binary mixtures of anisole (1) + aryl alcohols (2) at 308.15 K. ●, ■, and ◆ refer to mixtures containing benzyl alcohol, 2-phenylethanol, and 3-phenyl-1-propanol, respectively.

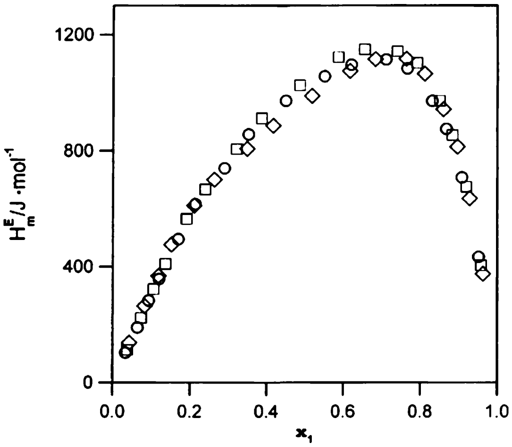

Figure 2. Experimental excess molar enthalpies, H^E_m, for binary mixtures of phenetole (1) + aryl alcohols (2) at 308.15 K. ○, □, and ◇ refer to mixtures containing benzyl alcohol, 2-phenylethanol, and 3-phenyl-1-propanol, respectively.

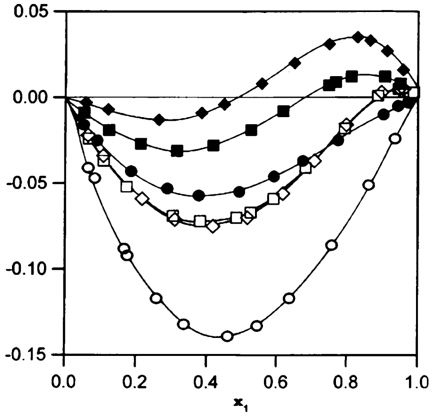

Figure 3. Experimental excess molar volumes, V^E_m, for binary mixtures of anisole (closed symbols) or phenetole (open symbols) + aryl alcohols at 308.15 K. ●, ■, ◆, ○, □, and ◇ refer to mixtures containing benzyl alcohol, 2-phenyl ethanol, and 3-phenyl-1-propanol, respectively. Solid curves, Redlich–Kister equation.

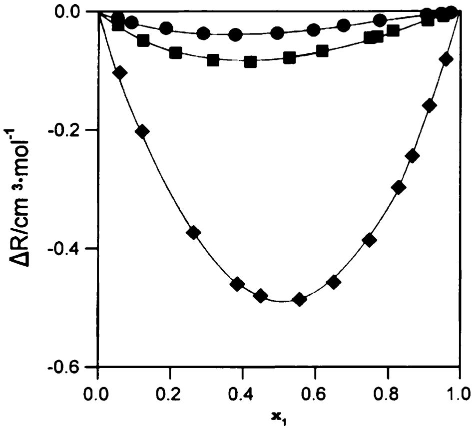

Figure 4. Experimental deviation in molar refraction, ΔR, for binary mixtures of anisole (1) + aryl alcohols (2) at 308.15 K. ●, ■, and ◆ refer to mixtures containing benzyl alcohol, 2-phenylethanol, and 3-phenyl-1-propanol, respectively. Solid curves, Redlich–Kister equation.

Before each series of measurements, the apparatus was calibrated at atmospheric pressure using double-distilled water and dry air, whose densities were taken from the literature.[14,15] The uncertainty in the density was $\pm 1.5 \times 10^{-5}$ g·cm^{-3}. Correspondingly, the uncertainty in V^E_m is estimated to be on the order of 1×10^{-3} cm^3·mol^{-1}. Before the measurements, the apparatus was checked by determining V^E_m using the test mixture benzene + cyclohexane at 298.15 K.[16] Our results showed a difference of ±0.5% in the central range of the mole fraction of benzene.

Refractive Index Measurements. Refractive indices at the sodium D line, n_D, were measured using a thermostated Abbe refractometer (Carl Zeiss, model G, Jena, Switzerland) with uncertainties of less than ±0.0001 units. Water was circulated into the prism of the refractometer by a circulation pump connected to an external thermostated water bath. Calibration was performed by measuring the refractive indices of double-distilled water, toluene, cyclohexane, and carbon tetrachloride at defined temperatures.[17] The sample mixtures were directly injected into the prism assembly of the instrument using an airtight hypodermic syringe, and an average of four measurements was taken for each mixture.

The solutions were pre-thermostated at 308.15 before the experiences to achieve quick thermal equilibrium. A minimum of three independent readings was taken for each composition, and their average value was considered in all of the calculations.

Results and Discussion

Tables 2 and 3 report experimental excess molar enthalpies, H^E_m, experimental densities, ρ, excess molar volumes, V^E_m, and refractive indices, n_D. These values are also graphically represented in Figures 1–5.

The excess molar volumes were computed by the following equation

$$V^E_m = \frac{x_1 M_1 + x_2 M_2}{\rho} - \frac{x_1 M_1}{\rho_1} - \frac{x_2 M_2}{\rho_2} \qquad (1)$$

where x_i, M_i, and ρ_i (i = 1, 2) are the mole fractions, molecular weights, and densities of anisole or phenetole (component 1) and component 2, respectively.

The molar refraction deviations, ΔR, were calculated from the Lorentz–Lorenz equation

$$\Delta R = R_m - \sum_{i=1}^{2} R_i \varphi_\iota \qquad (2)$$

where R_i and R_m are the molar refraction of the pure

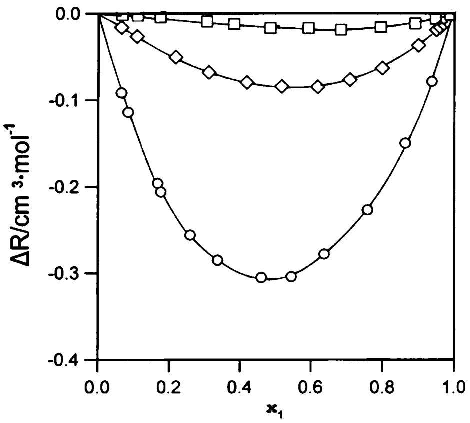

Figure 5. Experimental deviation in molar refraction, ΔR, for binary mixtures of phenetole (1) + aryl alcohols (2) at 308.15 K. ○, □, and ◇ refer to mixtures containing benzyl alcohol, 2-phenylethanol, and 3-phenyl-1-propanol, respectively. Solid curves, Redlich−Kister equation.

components and of the mixture, respectively, and φ_i is the volume fraction of the ith component, given as

$$\varphi_i = \frac{x_i V_i}{\sum_{i=1}^{2} x_i V_i} \tag{3}$$

where $V_i = M_i/\rho_i$.

The molar refraction, R_i, was obtained from the equation (Lorentz−Lorenz)

$$R_i = \left(\frac{{n_{\mathrm{D}(i)}}^2 - 1}{{n_{\mathrm{D}(i)}}^2 + 2}\right) V_i \tag{4}$$

where $n_{\mathrm{D}(i)}$ is the refractive index for the pure ith component.

The variations of H_m^E, V_m^E, and ΔR with composition are expressed by the Redlich−Kister polynomial

$$Q = x_1 x_2 \sum_{k \geq 0} a_k (x_1 - x_2)^k \tag{5}$$

where Q refers to H_m^E, V_m^E, or ΔR.

The adjustable parameters, a_k, were determined by least-squares methods, fitting the experimental values to eq 6, and the results are given in Table 4. The standard deviations, $\sigma(Q)$, reported in Table 4 were defined as

$$\sigma(Q) = \left|\frac{\phi_{\min}}{N - n}\right|^{0.5} \tag{6}$$

with N and n being the number of experimental points and parameters, respectively, whereas $\phi_{\min}$ is the minimum value of the objective function ϕ defined as

$$\phi = \sum_{k=1}^{N} {\eta_k}^2 \tag{7}$$

where $\eta_k = Q_\mathrm{calcd} - Q$. Q is the experimental value, and Q_calcd is evaluated through eq 5.

Figures 1 and 2 show values of H_m^E that are positive and large because of the dominant interaction energy of hydrogen bonding in alcohols. What is striking is the close crowding of H_m^E's in going from benzyl alcohol to 3-phenyl-1-propanol (that is, with the increasing of the alkyl chain connecting the phenyl group to the OH) with the ether anisole or phenetole. The difference in H_m^E never exceeds 8% for the larger values of H_m^E.

Table 4. Adjustable Parameters, a_k, from Equation 5 and Standard Deviations $\sigma(Q)$ from Equation 6 of Anisole or Phenetole + Aryl Alcohols at 308.15 K

function	a_0	a_1	a_2	a_3	$\sigma(Q)$
		Anisole (1) + Benzyl Alcohol (2)			
H^E/J·mol^{-1}	3904.7	1672.6	2316.2	2146.2	5.0
V^E/cm^3·mol^{-1}	−0.21220	0.11843			0.0061
ΔR/cm^3·mol^{-1}	−0.14287	0.08956			0.0009
		Anisole (1) + 2-Phenylethanol (2)			
H^E/J·mol^{-1}	4139.7	1804.7	2392.6	2966.6	4.9
V^E/cm^3·mol^{-1}	−0. 08590	0.19518	0.10700		0.0002
ΔR/cm^3·mol^{-1}	−0.31988	0.16913			0.0004
		Anisole (1) + 3-Phenyl-1-propanol (2)			
H^E/J·mol^{-1}	3922.6	1841.7	2854.6	2663.4	3.6
V^E/cm^3·mol^{-1}	0.00328	0.23466	0.19878		0.0007
ΔR/cm^3·mol^{-1}	−1.97444	−0.11670			0.0031
		Phenetole (1) + Benzyl Alcohol (2)			
H^E/J·mol^{-1}	4063.8	1697.8	2688.1	2047.2	6.7
V^E/cm^3·mol^{-1}	−0.54466	0.14257			0.0011
ΔR/cm^3·mol^{-1}	−1.21932	0.11711	−0.26666		0.0018
		Phenetole (1) + 2-Phenylethanol (2)			
H^E/J·mol^{-1}	4160.6	1916.1	2885.6	2063.0	5.7
V^E/cm^3·mol^{-1}	−0.27664	0.15652	0.14760	0.15980	0.0007
ΔR/cm^3·mol^{-1}	−0.06750	−0.05979			0.0004
		Phenetole (1) + 3-Phenyl-1-propanol (2)			
H^E/J·mol^{-1}	3884.0	1840.0	3601.3	2157.3	7.0
V^E/cm^3·mol^{-1}	−0.28705	0.16501	0.18620	0.13043	0.0006
ΔR/cm^3·mol^{-1}	−0.34113	−0.07660			0.0008

This insensibility of the H_m^E (nearly the same value of H_m^E is observed at any composition for mixtures with the same molar fraction, independent of the alcohol and ether) seems to be ascribed to the peculiar properties of the mixtures with alcohols and ethers. In fact, starting from the approximate relation $H_\mathrm{m}^\mathrm{E} \approx E_{11} + E_{22} - 2E_{12}$ between the interaction energies of components, we may first point out the dominant interaction energy of the alcohol, E_{22}, and the relatively smaller value of E_{11} for the ether. Second, the decrease of E_{22} with the increase of the alkyl chain length in the three alcohols must couple with a decrease of the interaction energy E_{12} between the OH of the alcohol and the ethereal group of anisole or phenetole. This balancing of interactions leads to the observed pattern of the experimental H_m^E values.

The absolute values of V_m^E are small, not exceeding 0.15 cm^3·mol^{-1}, but are larger for the mixture with phenetole, with negative V_m^E, whereas anisole−alcohol mixtures give complicated patterns with sign variation of V_m^E (maximum absolute values on the order of 5×10^{-2} cm^3·mol^{-1}).

The values of ΔR, finally, are negative, but no particular trend can be derived from the curves.

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