Densities, Viscosities, and Excess Molar Enthalpies of Propylene Carbonate + Anisole or + Phenetole at (293.15, 303.15, and 313.15) K

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Dynamic viscosities, η, densities, ρ, and enthalpies of mixing of binary mixtures containing propylene carbonate + anisole, or + phenetole have been determined at (293.15, 303.15, and 313.15) K and at atmospheric pressure over the whole range of composition. From these results, the deviations in viscosity, $\Delta\eta$, the excess molar volumes, V_m^E, and the excess molar enthalpies, H_m^E, have been calculated. Both $\Delta\eta$ and V_m^E are always negative, while positive values have been obtained for H_m^E. Minima of $\Delta\eta$ and V_m^E and maxima of H_m^E are the following: -0.248 mPa·s, -0.744 cm³·mol⁻¹, and 347 J·mol⁻¹ for the mixtures propylene carbonate + anisole; -0.248 mPa·s, -0.146 cm³·mol⁻¹, and 766 J·mol⁻¹ for the mixtures propylene carbonate + phenetole. The temperature effect has shown more marked differences for H_m^E. Results have been correlated using the Redlich−Kister polynomial.

Introduction

Experimental viscosity, η, density, ρ, and enthalpy of mixing results, H_m^E, in binary mixtures give information on the existence of specific interactions between the components, and they are very important in many practical problems concerning heat transport, mass transport, and fluid flows.

Furthermore, the importance of propylene carbonate, an aprotic solvent used for a variety of industrial applications, particularly in lithium batteries, has been widely stressed.[1−3] This solvent has been also the object of several previous studies.[4−6] In the present paper we report experimental dynamic viscosities, η, densities, ρ, and excess enthalpies, H_m^E of systems with two chemicals having different steric hindrance. Measurements of binary mixtures containing propylene carbonate + anisole or + phenetole have been performed at (293.15, 303.15, and 313.15) K and at atmospheric pressure. A literature survey showed that no measurements have been previously reported for the mixtures studied in this paper.

Experimental Section

Chemicals. All chemicals were from Aldrich Chemical Co. Propylene carbonate (stated purity 99.7%, water impurity < 0.02%) has been used without further purification. Anisole and phenetole, (both with stated purities of 99%, water impurity < 0.05%), have been purified following the procedure described elsewhere.[7] Purities of all products have been checked by using a Hewlett-Packard gas chromatograph model 5890 supplied with a HP (cross-linked 5% ME siloxane) capillary column. Purity values have confirmed the one stated for propylene carbonate. After purification the purities of anisole and phenetole were (99.8 and 99.7) mol % respectively.

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Table 1. Comparison of Experimental Densities, ρ, and Dynamic Viscosities, η, of Pure Components with Literature Values

compound	T/K	ρ/g·cm⁻³ this work	ρ/g·cm⁻³ lit.	η/mPa.s this work	η/mPa.s lit.
propylene carbonate	293.15	1.204 96		2.721	2.76[7]
	298.15	1.199 52	1.1995[8]	2.478	2.530[7]
			1.1998[9]		
			1.1988[10]		
			1.199 45[5]		
	303.15	1.194 30	1.1941[8]	2.237	
	313.15	1.183 67	1.1836[8]	1.863	
anisole	293.15	0.993 90	0.9940[7]	1.050	
	298.15	0.989 29	0.989 32[10]	0.969	
			0.989 32[11]		
			0.989 46[12]		
	303.15	0.984 51	0.9842[12]	0.915	0.920[11]
					0.907[12]
	313.15	0.975 06	0.9750[12]	0.793	
phenetole	293.15	0.964 85	0.965 14[7]	1.231	
	298.15	0.960 44	0.960 49[7]	1.130	
	303.15	0.955 50		1.036	1.040[7]
	313.15	0.964 85		0.894	

Before measurements, pure liquids have been degassed by ultrasound (ultrasonic bath, Hellma, type 460, Milan, Italy), kept in dark bottles, and dried over molecular sieves (Union Carbide, type 4A, $^1/_{16}$ in. pellets)

The measured densities and viscosities of pure components have been compared with literature values,[5,7−12] and results are reported in Table 1.

Apparatus and Procedure. Binary mixtures were prepared from known masses ($\approx$20 g) of each component in airtight stoppered bottles, and the mass measurements were made using a digital balance (Mettler, model AE 160, Switzerland) showing an accuracy of $\pm$ 0.0001 g. The more volatile component (anisole and phenetole) was introduced directly in the bottle, and the stoppered bottle has been weighed. Propylene carbonate was injected into the bottle

Table 2. Activation Energies for Viscous Flow

compound	E/kJ.mol^{-1}	T range/K	σ
propylene carbonate	14.5	293.15–313.15	0.012
anisole	10.6	293.15–313.15	0.009
phenetole	12.3	293.15–313.15	0.006

though the stopper by means of a syringe. This method led to negligible vapor loss and contamination and the error in the mole fraction of propylene carbonate was calculated to be less than $\pm 1.5 \times 10^{-4}$. Densities, ρ, have been determined using a digital density meter (Anton Paar, DMA 60/602, Graz, Austria). The temperature of the measuring cell was controlled to within ± 0.005 K by circulation of thermostated water (Heto bath, type 01 DBT 623, Birkeròd, Denmark). Two precision digital thermometers (Anton Paar, DT-25 and DT-40) with a thermistor probe have been used to read the cell temperature to an accuracy of ± 0.01 K. Experimental densities, obtained from measurements of the oscillation period in the vibrating U-tube of the densimeter showed accuracies of $\pm 1 \times 10^{-5}$ g·cm^{-3} which lead to an estimated uncertainty in V_m^E's less than 3×10^{-3} cm^3· mol^{-1}.

Values of excess molar volumes, V_m^E, were determined from densities using the following relation

$$V_m^E = (x_1 M_1 + x_2 M_2)/\rho - x_1 M_1/\rho_1 - x_2 M_2/\rho_2 \quad (1)$$

where M_i and ρ_i are the molar mass and the density of component i. Corrections for buoyancy and evaporation of components were made.

Before measurements, the apparatus was calibrated with twice-distilled water and dry air whose density values are well-known in the literature[13,14] and has also been checked against literature data of the mixture benzene + cyclohexane.[15] An agreement of our data to better than 0.5% in the central range of mole fraction of benzene was found.

Dynamic viscosities, η, were determined using a Schott measuring system (Schott-Gerate GmbH, model AVS 310, Mainz, Germany), consisting of a digital control unit, a measuring stand, a constant-temperature bath, and a calibrated Schott KPG Ubbelohde suspended level viscometer.

The precision of the electronic stopwatch used to measure the liquid flow times was ± 0.01 s. Temperature control of the thermostatic bath was carried out at an accuracy of ± 0.01 K. At least triplicate measurements of flow times were taken and their reproducibility was within 0.2% or less. Viscosities are uncertain to less than 1%, and that leads to an estimated deviation, $\Delta \eta$, on the order of 1%. The viscometer has been calibrated with double-distilled water whose viscosity and density are well-known and reported in the literature.[7]

Values of the dynamic viscosity, η, have been calculated using the relation

$$\eta = \rho v \quad (2)$$

Dynamic viscosities, η, of pure liquids have been correlated to temperature by the Arrhenius equation

$$\eta = A e^{E/RT} \quad (3)$$

where A is a system specific constant, E the activation energy for viscous flow, R the gas constant, and T the temperature.

Values of E have been computed by an unweighted linear least-squares fitting and are reported in Table 2.

Table 3. Experimental Densities, ρ, Dynamic Viscosities, η, Excess Molar Volumes, V_m^E, and Viscosity Deviations, $\Delta\eta$, of Binary Mixtures Containing Propylene Carbonate + Anisole or Phenetole

x_1	ρ/g.cm^{-3}	η/mPa.s	V_m^E/cm^3.mol^{-1}	$\Delta\eta$/mPa.s
	Propylene Carbonate(1) + Anisole(2)			
		T/K = 293.15		
0.0	0.993 90	1.050	0.0	0.0
0.0794	1.009 58	1.142	−0.254	−0.041
0.1323	1.020 00	1.199	−0.383	−0.073
0.2487	1.043 07	1.316	−0.583	−0.150
0.3578	1.065 01	1.449	−0.682	−0.199
0.4971	1.093 67	1.640	−0.705	−0.241
0.5795	1.111 01	1.773	−0.670	−0.245
0.6669	1.129 75	1.917	−0.596	−0.248
0.7278	1.142 77	2.031	−0.503	−0.236
0.8350	1.166 69	2.246	−0.354	−0.199
0.9009	1.181 90	2.406	−0.225	−0.150
1.0	1.204 96	2.721	0.0	0.0
		T/K = 303.15		
0.0	0.984 51	0.915	0.0	0.0
0.0794	1.000 14	0.971	−0.262	−0.050
0.1323	1.010 50	1.013	−0.394	−0.077
0.2487	1.033 45	1.101	−0.599	−0.143
0.3578	1.055 27	1.212	−0.702	−0.176
0.4971	1.083 76	1.372	−0.723	−0.201
0.5795	1.101 00	1.479	−0.688	−0.203
0.6669	1.119 61	1.591	−0.612	−0.206
0.7278	1.132 81	1.682	−0.537	−0.196
0.8350	1.156 51	1.851	−0.363	−0.168
0.9009	1.171 41	1.978	−0.231	−0.129
1.0	1.194 30	2.237	0.0	0.0
		T/K = 313.15		
0.0	0.975 06	0.793	0.0	0.0
0.0794	0.990 64	0.849	−0.271	−0.029
0.1323	1.000 95	0.889	−0.406	−0.045
0.2487	1.023 77	0.972	−0.614	−0.087
0.3578	1.045 52	1.066	−0.723	−0.110
0.4971	1.073 84	1.177	−0.744	−0.148
0.5795	1.090 99	1.260	−0.709	−0.153
0.6669	1.109 47	1.347	−0.628	−0.160
0.7278	1.122 59	1.423	−0.551	−0.149
0.8350	1.146 14	1.569	−0.373	−0.118
0.9009	1.160 95	1.671	−0.237	−0.086
1.0	1.183 67	1.863	0.0	0.0
	Propylene Carbonate(1) + Phenetole(2)			
		T/K = 293.15		
0.0	0.964 85	1.231	0.0	0.0
0.1218	0.985 91	1.338	−0.082	−0.074
0.2226	1.004 34	1.436	−0.105	−0.127
0.3591	1.031 29	1.586	−0.107	−0.181
0.4874	1.059 09	1.739	−0.090	−0.219
0.5257	1.067 93	1.789	−0.084	−0.226
0.5674	1.077 84	1.837	−0.074	−0.239
0.6723	1.104 32	1.985	−0.051	−0.248
0.7807	1.134 31	2.170	−0.029	−0.224
0.8898	1.167 56	2.405	−0.010	−0.152
0.9431	1.185 12	2.544	−0.004	−0.092
1.0	1.204 96	2.721	0.0	0.0
		T/K = 303.15		
0.0	0.955 50	1.231	0.0	0.0
0.1218	0.976 51	1.119	−0.093	−0.063
0.2226	0.994 90	1.202	−0.124	−0.101
0.3591	1.021 70	1.316	−0.125	−0.151
0.4874	1.049 34	1.440	−0.107	−0.181
0.5257	1.058 14	1.474	−0.100	−0.194
0.5674	1.068 00	1.516	−0.091	−0.201
0.6723	1.094 33	1.630	−0.067	−0.213
0.7807	1.124 14	1.773	−0.041	−0.200
0.8898	1.157 15	1.960	−0.015	−0.145
0.9431	1.174 59	2.078	−0.006	−0.091
1.0	1.194 30	2.237	0.0	0.0
		T/K = 313.15		
0.0	0.964 85	0.894	0.0	0.0
0.1218	0.967 06	0.945	−0.100	−0.067
0.2226	0.985 37	1.013	−0.135	−0.097
0.3591	1.012 11	1.110	−0.146	−0.132
0.4874	1.039 60	1.215	−0.126	−0.151
0.5257	1.048 35	1.246	−0.119	−0.158
0.5674	1.058 15	1.278	−0.108	−0.166
0.6723	1.084 33	1.373	−0.081	−0.172
0.7807	1.113 96	1.495	−0.051	−0.155
0.8898	1.146 79	1.651	−0.022	−0.105
0.9431	1.164 12	1.742	−0.010	−0.066
1.0	1.183 67	1.863	0.0	0.0

Values of the deviations in viscosity have been calculated from dynamic viscosities according to the following equation

$$\Delta\eta = \eta - \sum_i x_i \eta_i^\circ \qquad (4)$$

where η_i° is the dynamic viscosity of pure liquid i.

Excess molar enthalpies, H_m^E, have been determined using a flow microcalorimeter (LKB, model 2107, Bromma, Sweden) working in the absence of vapor phase and equipped with two automatic burets (ABU, Radiometer, Copenhagen, Denmark) necessary to pump pure liquids into the mixing cell of the calorimeter. Temperature has been kept constant to ±0.01 K and electronically controlled inside the apparatus. Electrical calibration and operating procedure have been described elsewere.[16,17] Mole fractions x_1 of propylene carbonate (component 1) have been determined from flow rates with a precision of 1×10^{-4}, which leads to an error on H_m^E of 0.5−1 J·mol^{-1}. Two replicate measurements of H_m^E have been made for each point. Before measurements, the apparatus has been checked using the test mixture cyclohexane + hexane for which literature values are well-known,[17] and the agreement with literature values was less than 0.5% over the central range of mole fraction of cyclohexane.

Results and Discussion

Values of V_m^E and $\Delta\eta$ are reported in Table 3, while excess molar enthalpies, H_m^E, are listed in Table 4.

To each set of experimental values, a Redlich−Kister polynomial of the type

$$Q_m^E = x_1 x_2 \sum_{k \geq 0} a_k (x_1 - x_2)^k \qquad (5)$$

has been fitted by an unweighted least-squares method, where $Q_m^E = H_m^E/\text{J.mol}^{-1}$, $V_m^E/\text{cm}^3.\text{mol}^{-1}$, or $\Delta\eta/\text{mPa·s}$, x_1, x_2 are the molar fractions of propylene carbonate and component 2, and a_k are the adjustable parameters. The a_k values are given in Table 5 together with the standard deviation $\sigma(Q_m^E)$, defined as

$$\sigma(Q_m^E) = |\phi_{min}/(N - n)|^{0.5} \qquad (6)$$

In eq 6, N and n are the number of experimental points and of parameters respectively, whereas ϕ_{min} is the minimum value of the objective function ϕ defined as

$$\phi = \sum_{k=1}^{N} \omega_k^2 \qquad (7)$$

where $\omega_k = Q_{m,calcd}^E - Q_m^E$; Q_m^E is the experimental value, and $Q_{m,calcd}^E$ is evaluated through eq 5.

Deviations in viscosities, excess molar volumes, V_m^E, and excess molar enthalpies, H_m^E, vs mole fraction, x_1, of propylene carbonate are listed in Table 5 and represented in Figures 1−3.

McAllister's multibody interaction model[18] is widely used for correlating the kinematic viscosity of liquid mixtures with mole fraction. The three-body model is defined as

$$\ln \nu = x_1^3 \ln \nu_1 + 3x_1^2 x_2 \ln \nu_{12} + 3x_1 x_2^2 \ln \nu_{21} +$$
$$x_2^3 \ln \nu_2 - \ln(x_1 + x_2 M_2/M_1) +$$
$$3x_1^2 x_2 \ln [(2 + M_2/M_1)/3] + 3x_1 x_2^2 \ln [(1 + 2M_2/M_1)/3] +$$
$$x_2^3 \ln(M_2/M_1) \qquad (8)$$

**Table 4. Excess Molar Enthalpies, H_m^E, for Binary Mixtures Containing Propylene Carbonate + Anisole or Phenetole**

x_1	$H_m^E/\text{J·mol}^{-1}$	x_1	$H_m^E/\text{J·mol}^{-1}$
	Propylene Carbonate(1) + Anisole(2)		
	$T/\text{K} = 293.15$		
0.0508	63	0.6584	292
0.0967	112	0.7198	262
0.1383	155	0.7939	211
0.1763	189	0.8370	175
0.2430	238	0.8851	139
0.2997	273	0.9113	107
0.3910	309	0.9391	73
0.4611	323	0.9686	39
0.5622	311		
	$T/\text{K} = 303.15$		
0.0508	75	0.6585	299
0.0967	136	0.7199	266
0.1381	182	0.7940	220
0.1762	211	0.8371	183
0.2431	264	0.8852	135
0.2999	293	0.9113	106
0.3912	319	0.9391	80
0.4613	328	0.9686	43
0.5624	326		
	$T/\text{K} = 313.15$		
0.0508	80	0.6587	309
0.0968	142	0.7200	280
0.1385	186	0.7941	224
0.1765	230	0.8372	190
0.2433	278	0.8853	136
0.3000	313	0.9114	106
0.3913	342	0.9391	78
0.4615	347	0.9686	42
0.5625	340		
	Propylene Carbonate(1) + Phenetole(2)		
	$T/\text{K} = 293.15$		
0.0586	178	0.6910	619
0.1107	305	0.7493	569
0.1574	401	0.8176	466
0.1994	485	0.8567	386
0.2720	582	0.8997	290
0.3325	646	0.9228	226
0.4277	698	0.9472	161
0.4990	708	0.9729	88
0.5991	690		
	$T/\text{K} = 303.15$		
0.0587	188	0.6918	647
0.1108	323	0.7495	581
0.1575	426	0.8178	474
0.1995	513	0.8568	403
0.2722	606	0.8998	297
0.3327	665	0.9229	243
0.4279	723	0.9472	173
0.4992	729	0.9729	92
0.5993	716		
	$T/\text{K} = 313.15$		
0.0587	180	0.6920	673
0.1109	318	0.7496	590
0.1576	423	0.8179	484
0.1997	505	0.8569	399
0.2724	613	0.8998	298
0.3329	686	0.9229	237
0.4281	752	0.9473	168
0.4994	766	0.9729	89
0.5995	774		

and the four-body model is given by

$$\ln \nu = x_1^4 \ln \nu_1 + 4x_1^3 x_2 \ln \nu_{1112} + 6x_1^2 x_2^2 \ln \nu_{1122} +$$
$$4x_1 x_2^3 \ln \nu_{2221} + x_2^4 \ln \nu_2 - \ln(x_1 + x_2 M_2/M_1) +$$
$$4x_1^3 x_2 \ln [(3 + M_2/M_1)/4] + 6x_1^2 x_2 2 \ln [(1 +$$
$$M_2/M_1)/2] + 4x_1 x_2^3 \ln [(1 + 3M_2/M_1)/4] +$$
$$x_2^4 \ln(M_2/M_1) \qquad (9)$$

Table 5. Least-Squares Parameters, a_k, Eq 5, and Standard Deviations, $\sigma(Q_m^E)$, Eq 6, of Propylene Carbonate + Anisole or Phenetole at (298.15 and 313.15) K

| function	a_0	a_1	a_2	a_3	$\sigma(Q_m^E)$
Propylene Carbonate(1) + Anisole(2)					
T/K = 293.15					
$\Delta\eta$/mPa·s	−0.955	−0.304	−0.263	−0.575	0.002
V_m^E/cm³·mol⁻¹	−2.8116	0.5444	−0.2102		0.0066
H_m^E/J·mol⁻¹	1293.9	3.1			3.6
T/K = 303.15					
$\Delta\eta$/mPa·s	−0.800	−0.159	−0.386	−0.490	0.002
V_m^E/cm³mol⁻¹	−2.8932	0.5328	−0.2504		0.0033
H_m^E/J mol⁻¹	1325.0	−101.2	190.6		2.6
T/K = 313.15					
$\Delta\eta$/mPa.s	−0.580	−0.332	−0.138	−0.009	0.003
V_m^E/cm³·mol⁻¹	−2.9762	0.5525	−0.2532		0.0040
H_m^E/J·mol⁻¹	1400.2	−143.5	152.0		2.6
Propylene Carbonate(1) + Phenetole(2)					
T/K = 293.15					
$\Delta\eta$/mPa·s	−0.892	−0.505	−0.402	−0.065	0.002
V_m^E/cm³·mol⁻¹	−0.3488	0.3611	−0.1499	0.1110	0.0008
H_m^E/J·mol⁻¹	2845.9	39.1	516.8		4.5
T/K = 303.15					
$\Delta\eta$/mPa·s	−0.746	−0.453	−0.456	−0.213	0.002
V_m^E/cm³·mol⁻¹	−0.4197	0.3794	−0.1794	0.1555	0.0006
H_m^E/J·mol⁻¹	2936.6	9.1	627.1		3.6
T/K = 313.15					
$\Delta\eta$/mPa·s	−0.621	−0.305	−0.383		0.002
V_m^E/cm³·mol⁻¹	0.4972	0.4193	−0.1475	0.0787	0.0008
H_m^E/J·mol⁻¹	3072.1	53.7	300.8		2.6
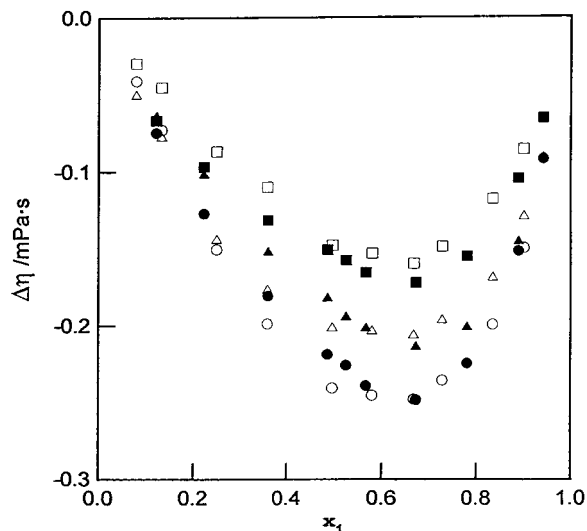
Figure 1. Deviations in viscosity, $\Delta\eta$, vs the mole fraction x_1 of propylene carbonate + anisole (open points) or phenetole (closed points) mixtures. Circles, triangles, and squares refer to (293.15, 303.15, and 313.15) K, respectively.

where ν_{12}, ν_{21}, ν_{1112}, ν_{1122}, and ν_{2221} are the model parameters. Table 6 records the calculated results, showing that the four-body model is more adequate for these systems.

As can be seen from the figures, the main features of the systems propylene carbonate + anisole or + phenetole are as follows: volume contraction and adsorption of enthalpy of mixing and negative values of the excess viscosity $\Delta\eta$.

Moreover, the small temperature dependence of V_m^E and H_m^E contrasts with the marked change of $\Delta\eta$ as the temperature increases.

Finally in going from anisole to phenetole, i.e., in the presence of an increase of the steric hindrance of the
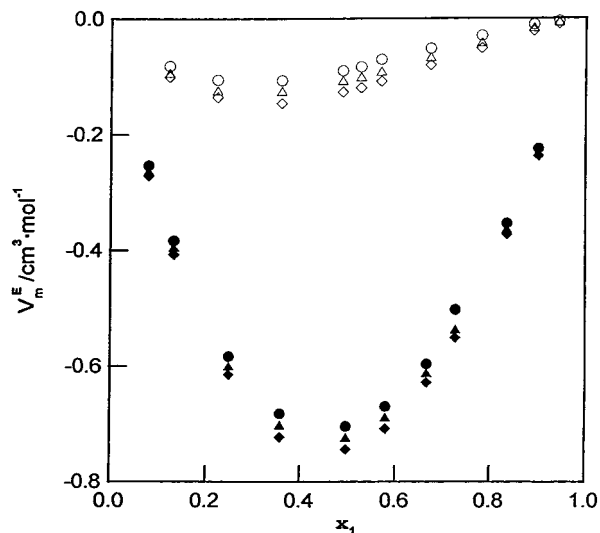
Figure 2. Excess molar volumes, V_m^E, vs the mole fraction x_1 of propylene carbonate + anisole (open points) or phenetole (closed points) mixtures. Circles, triangles, and squares refer to (293.15, 303.15, and 313.15) K, respectively.
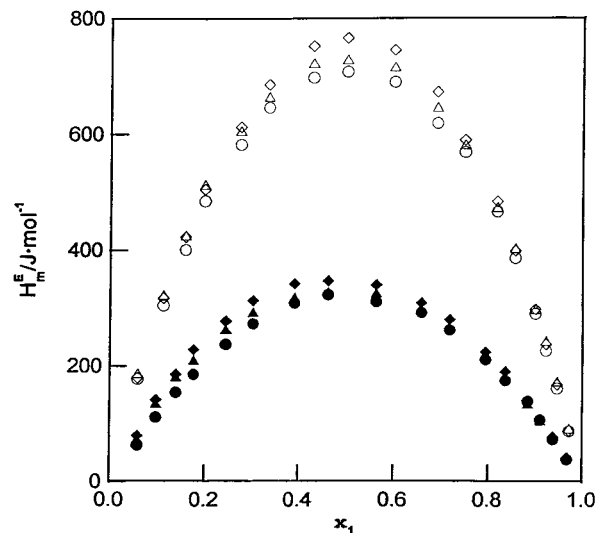
Figure 3. Excess molar enthalpies, H_m^E, vs the mole fraction x_1 of propylene carbonate + anisole (open points) or phenetole (Closed points) mixtures. Circles, triangles, and squares refer to (293.15, 303.15, and 313.15) K, respectively.

Table 6. Coefficients of McAllister's Models and Standard Deviations for Kinematic Viscosities

	three-body model			four-body model			
T/K	ν_{12}	ν_{21}	$\sigma/(10^{-6}$ m² s⁻¹)	ν_{1112}	ν_{1122}	ν_{2221}	$\sigma/(10^{-6}$ m² s⁻¹)
Propylene Carbonate(1) + Anisole(2)							
293.15	1.602	1.386	0.005	1.736	1.504	1.694	0.005
303.15	1.365	1.128	0.007	1.419	1.332	1.360	0.005
313.15	1.151	1.031	0.002	1.237	1.102	1.270	0.002
Propylene Carbonate(1) + Phenetole(2)							
293.15	1.705	1.558	0.004	1.777	1.711	1.838	0.001
303.15	1.387	1.322	0.007	1.433	1.457	1.537	0.001
313.15	1.215	1.087	0.008	1.232	1.254	1.272	0.002

volatile component, we note marked variations of both V_m^E and H_m^E while $\Delta\eta$ results are nearly unchanged.

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