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Thermochimica Acta 294 (1997) 179–186

thermochimica
acta

Prediction and experimental determination of thermodynamic properties of binary mixtures containing propylene carbonate +1,2-epoxybutane at (288.15, 298.15, and 313.15 K)

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Received 12 July 1996; accepted 12 December 1996

Abstract

This paper reports experimentally determined excess molar enthalpies, H_m^E, excess heat capacities, C_p^E, densities, ρ, and excess molar volumes, V_m^E, of propylene carbonate +1,2-epoxybutane at (288.15, 298.15 and 313.15 K) and atmospheric pressure. The results of H_m^E and V_m^E are compared with those predicted by an extended cell model elaborated by [I. Prigogine, The Molecular Theory of Solutions, North- Holland, Amsterdam (1957)]. Experimental H_m^E values are positive and show only small changes with the increase of temperature, while V_m^E data are always negative and decrease with the increase of temperature. Also C_p^E values are negative for all mixtures. Results have been qualitatively interpreted in terms of molecular interactions. © 1997 Elsevier Science B.V.

Keywords: Cell model; Densities; Excess heat capacities; Excess molar enthalpies; Excess molar volumes

1. Introduction

The peculiarity of recovering some classes of hydrocarbon compounds and sour gases and dissolving a variety of organic and inorganic substances can be ascribed to esters of carbonic acids. Particularly, propylene carbonate finds a wide range of applications in organic synthesis and electrochemical studies as important solvent for nonaqueous electrolytes in high-energy lithium batteries [1–3].

In spite of the great importance in industry, there are only a few thermodynamic studies of aqueous or nonaqueous solution mixtures containing propylene carbonate.

Some of our previous papers [4–6] have been concerned with the physical and the thermodynamic properties of binary mixtures containing esters of carbonic acid with a second component at various temperatures or with different functional groups.

Continuing our research program, we report in the present study the excess molar enthalpies, H_m^E, the heat capacities of mixtures, C_p, and the excess molar volumes, V_m^E, of propylene carbonate +1,2-epoxybutane, the latter compound being a strong proton acceptor monoether used as a solubilizing agent.

To our knowledge the data reported in this paper are not available in literature.

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PII S0040-6031(97)00049-X

2. Experimental

2.1. Chemicals

Both the chemicals used in this study were purchased from Aldrich(Milawaukee, WI): propylene carbonate (41,422-0; purity 99.7 mol%), 1,2-epoxybutane (24,134-2; purity +99 mol%). Propylene carbonate was used as received, without further purification owing to its high-purity grade, checked and confirmed by GCL analysis. 1,2-Epoxybutane was distilled with sodium in a Vigreux column and the 80% middle distilled product was collected. After purification, GCL purity was ascertained to be 99.8 mol%.

Both components, before measurements, were kept in dark bottles and stored over molecular sieves (Union Carbide type 4A,1/16 in. pellets) and degassed by ultrasound technique(ultrasonic bath, Hellma, type 460, Milan, Italy).

Densities, ρ, and refractive indices, $n(D)$, of pure components, determined at 298.15 K are listed in Table 1 and compared with literature data [3,7,8].

2.2. Calorimetric measurements

Excess molar enthalpies, H_m^E, were measured at (288.15, 298.15 and 313.15 K) and at atmospheric pressure using a flow microcalorimeter (LKB Produkter, model 2107, Bromma, Sweden): details and operating procedure of the apparatus have been described previously by Monk and Wadso and Francesconi and Comelli [9,10]. Temperature was kept constant within ±0.01 K by a circulating bath and digital instruments. Two automatic burets (ABU, Radiometer, Copenhagen, Denmark) were used to pump liquids into the mixing cell of the calorimeter. Mole fractions were determined from the calibrated flow rates with a precision of $\pm 1 \cdot 10^{-4}$ and two replicate measurements were made for each determination.

The apparatus was electrically calibrated before use and checked by measuring the H_m^E values of the test mixture cyclohexane + hexane [11]. The precision between our values and literature is better than 0.5% over the central range of mole fraction of cyclohexane.

The heat capacity measurements were carried out using a Perkin Elmer DSC 7 differential scanning calorimeter, driven by a Unix computer.

Dry nitrogen was used as purge gas, flowing at a rate of 30 ml min^{-1}. The external block temperature control was set at 238.15 K, permitting operation down to 273.15 K. The instrument was equipped with dry box assembly, through which dry nitrogen gas was kept flowing in order to avoid condensation of atmospheric moisture. The instrument was calibrated with high-purity standards (indium and cyclohexane) at 5 K min^{-1}. The temperature was known within ±0.1 K. The samples, approximately 10 mg, were weighed to ±0.01 mg and encapsulated in hermetic pans. The heat capacity of the samples was obtained by means of three consecutive DSC runs at a scanning rate of 5 K min^{-1}: the sample run, the blank run and the standard sample (sapphire) run [11]. Care was taken to ensure that for all the three scans, sample, blank and standard, similar initial and final isotherm levels were reached. The heat capacity data were obtained by means of the commercial software supplied by Perkin Elmer. The estimated error for repeated data is less than ±1%.

Before measurements we have checked the calorimeter by a test mixture water + methanol at 308.15 K [12]. The discrepancy of heat capacities between our values and the literature is less than 0.6% in the central range of mole fraction of water (our C_p values for pure components water and methanol are 75.410 $\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$ and 84.540 $\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$; literature values are 75.266 $\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$ and 83.740 $\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$, respectively).

Table 1
Densities, ρ, and refractive indices, $n(D)$, of pure chemicals in comparison with literature values at 298.15 K

Component	ρ/g·cm^{-3}		$n(D)$	
	Expt.	Lit.	Expt.	Lit.
Propylene carbonate	1.19957	1.1998 [3]	1.4195	1.4194 [7]
1,2-Epoxybutane	0.82463	0.8240 [8]	1.3815	1.381 [8]

Table 2
Heat capacities of mixing, C_p, of propylene carbonate(1) +1,2-epoxybutane(2) at (288.15, 298.15 and 313.15 K)

x_1	C_p/ J·mol^{-1}·K^{-1}	x_1	C_p/ J·mol^{-1}·K^{-1}
T=288.15 K			
0.0000	147.3	0.5468	155.8
0.2227	149.8	0.6424	158.3
0.2870	150.4	0.7427	159.3
0.3354	151.6	0.8099	160.8
0.3944	152.7	0.9164	164.2
0.5075	155.1	1.0000	166.4
T=298.15 K			
0.0000	149.8	0.5468	157.9
0.2227	152.2	0.6424	160.2
0.2870	152.4	0.7427	161.3
0.3354	154.4	0.8099	163.2
0.3944	154.9	0.9164	165.9
0.5075	156.8	1.0000	168.2
T=313.15 K			
0.0000	154.0	0.5468	161.1
0.2227	155.6	0.6424	163.7
0.2870	156.3	0.7427	164.4
0.3354	158.2	0.8099	166.2
0.3944	158.4	0.9164	168.8
0.5075	159.6	1.0000	170.9

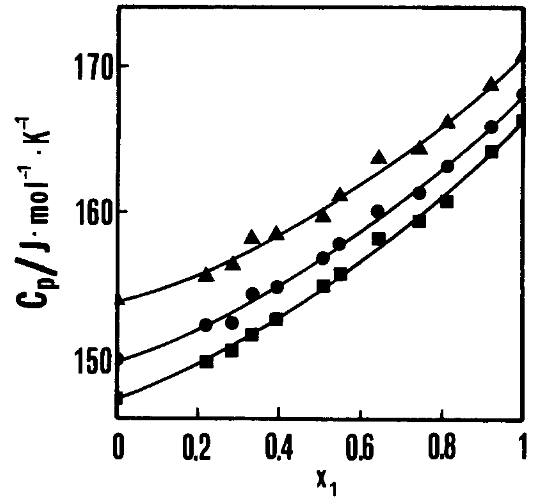

Fig. 1. Heat capacities of mixing, C_p, as a function of mole fraction x_1 of propylene carbonate for the mixtures propylene carbonate +1,2-epoxybutane. Solid symbols (■),(●),(▲) refer at (288.15, 298.15 and 313.15 K), respectively; solid curves, Eq. (1).

The experimental C_p data of the mixtures, reported in Table 2 and graphically represented in Fig. 1 show a non linear behaviour and the model used to fit the heat capacities C_p is

$$C_p/\mathrm{J\cdot mol^{-1}\cdot K^{-1}} = a_0 + a_1x_1 + a_2x_1^2 \qquad (1)$$

where a_k are temperature dependent parameters and x_1 is the mole fraction of propylene carbonate. Values of a_k, correlation coefficients $|R|$ and standard deviation $\sigma(C_p)$ are listed in Table 3. The uncertainties of C_p are

Table 3
Parameters, a_k, correlation coefficients, $|R|$, and standard deviations, $\sigma(C_p)$, $\sigma(C_p^E)$, Eq. (1), of heat capacities C_p and excess molar heat capacities C_p^E for the mixtures propylene carbonate(1) +1,2-epoxybutane(2) at (288.15, 298.15, and 313.15 K)

Function\ J·mol^{-1}·K^{-1}	T/K	a_0	a_1	a_2	$\lvert R\rvert$	$\Sigma(C_p)$, $\sigma(C_p^E)$/ J·mol^{-1}·K^{-1}
C_p	288.15	147.30	10.144	8.9442	0.9997	0.44
C_p^E	288.15	−0.0162	−8.9511	8.9579	0.9678	0.44
C_p	298.15	149.79	9.2301	9.2153	0.9997	0.43
C_p^E	298.15	−0.0123	−9.2218	9.2261	0.9716	0.43
C_p	313.15	153.93	7.7055	9.2339	0.9993	0.55
C_p^E	313.15	−0.0218	−9.2075	9.2399	0.9542	0.55

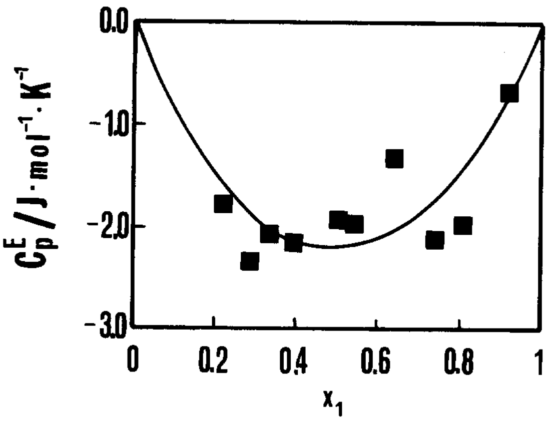

Fig. 2. Excess molar heat capacities, C_p^E, of propylene carbonate +1,2-epoxybutane at 288.15 K and at different mole fraction of propylene carbonate (the solid curve is according to Eq. (1) and solid squares represent the experimental points).

estimated to be less than 1%, which leads to an accuracy of the excess molar heat capacities, C_p^E, of ±1 J·mol^{-1}·K^{-1}.C_p^E values were evaluated by the formula

$$C_p^E/\mathrm{J\cdot mol^{-1}\cdot K^{-1}} = C_p - x_1 C_{p1} - (1 - x_1)C_{p2} \tag{2}$$

where C_{p1} and C_{p2} are the heat capacities of pure components.

The C_p^E values at 298.15 K are shown in Fig. 2.

2.3. Volumetric measurements

Mixtures necessary to determine both densities, ρ, and excess molar volumes, V_m^E, were prepared by mass in air-tight glass bottles of nominal volume ≈20 cm^3 and weighed in a single-pan balance (Mettler, model AE 160, Switzerland) to an accuracy of $1\cdot10^{-4}$ g.

We have charged the heavier component first to prevent evaporation and to minimize errors in composition: error in mole fraction is estimated to be $1\cdot10^{-4}$.

Densities, ρ, were measured using a digital vibrating density meter (Anton Paar, model 60, Graz, Austria) equipped with a density measuring cell (model 602) with an accuracy of the measured period of $\pm1\cdot10^{-6}$ s. We have selected the period of 10 000 oscillation cycles, keeping 20 measurements for every samples.

The temperature of the U-shaped tube was checked using calibrated digital thermometers (Anton Paar, DT-15; DT-25;DT-40) with an accuracy of ±0.01 K, while a Hetotherm bath circulator (Heto type 01 DBT 623, Birkerød, Denmark) kept the temperature constant within ±0.005 K.

Acetone was used for rinsing the cell tube after each run.

The operating procedure as described by Fermeglia and Lapasin [13] was followed to prepare mole fractions. The accuracies in densities and excess molar volumes are $5\cdot10^{-5}$ g·cm^{-3} and $3\cdot10^{-3}$ cm^3·mol^{-1}, respectively.

Before measurements, the density meter was calibrated by using dry air and doubly distilled water and checked with the test mixture benzene + cyclohexane [14]. Our results agree with literature, showing a discrepancy of less than 0.5% over the central range of the mole fraction of benzene.

All measurements were corrected for buoyancy and for evaporation of components but only a negligible contribution to the mole fraction was detected with a variance of $1\cdot10^{-4}$.

Excess molar volumes, V_m^E, were computed from the density and composition using the equation

$$V_m^E/\mathrm{cm^3\cdot mol^{-1}} = (x_1M_1 + (1 - x_1)M_2)/\rho - (x_1M_1/\rho_1 + (1 - x_1)M_2/\rho_2) \tag{3}$$

where M_1 and M_2 are the molecular masses of pure components, and ρ, ρ_1, ρ_2 are the densities of the mixture, propylene carbonate and 1,2-epoxybutane.

3. Correlation of the experimental values

The experimental results of H_m^E, and V_m^E as a function of mole fraction x_1 of propylene carbonate, reported in Tables 4 and 5, were fitted by unweighed least-squares method to the Redlich–Kister equation

$$Q_m^E = x_1x_2\sum_{k\geq0} a_k(x_1 - x_2)^k \tag{4}$$

where $Q_m^E = H_m^E/\mathrm{J\cdot mol^{-1}}$ and $V_m^E/\mathrm{cm^3\cdot mol^{-1}}$, respectively.

The number of adjustable parameters a_k are listed in Table 6 with the standard deviations $\sigma(Q_m^E)$ defined as

$$\sigma(Q_m^E) = |\Phi/(N - n)|^{0.5} \tag{5}$$

Table 4
Excess molar enthalpies, H_m^E, of propylene carbonate (1) +1,2-epoxybutane (2) at (288.15, 298.15 and 313.15 K)

x_1	H_m^E/J·mol^{-1}	x_1	H_m^E/J·mol^{-1}
T=288.15 K			
0.0409	30	0.6055	210
0.0786	54	0.6717	195
0.1134	78	0.7543	157
0.1457	93	0.8036	128
0.2037	124	0.8599	86
0.2543	150	0.8911	63
0.3384	180	0.9247	39
0.4055	201	0.9609	17
0.5057	214		
T =298.15 K			
0.0489	29	0.6065	201
0.0788	51	0.6726	184
0.1138	73	0.7550	150
0.1462	92	0.8043	119
0.2043	121	0.8604	82
0.2551	147	0.8915	55
0.3394	176	0.9250	32
0.4065	195	0.9610	12
0.5067	207		
T =313.15 K			
0.0413	23	0.6082	200
0.0794	45	0.6742	185
0.1145	70	0.7563	152
0.1470	89	0.8054	123
0.2055	117	0.8613	84
0.2564	142	0.8922	62
0.3409	174	0.9247	39
0.4082	194	0.9613	18
0.5085	208		

where N is the number of experimental points and n the number of adjustable parameters.The objective function Φ is defined as

$$\Phi = \sum_{k=1}^{N} \eta_k^2 \tag{6}$$

where $\eta = Q_{m,calcd}^E - Q_m^E$, $Q_{m,calcd}^E$ being determined from the right hand side of Eq. (4).

Experimental values of H_m^E and V_m^E are listed in Tables 4 and 5, while Figs. 3 and 4 show their graphical representations.

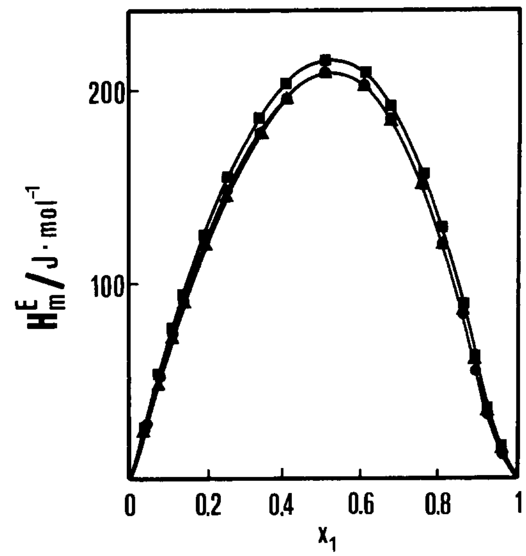

Fig. 3. Excess molar enthalpies, H_m^E, of propylene carbonate +1,2-epoxybutane. Solid symbols (■),(●),(▲) refer at (288.15, 298.15 and 313.15 K), respectively; solid curves are least-squares representations of results by Eq. (3).

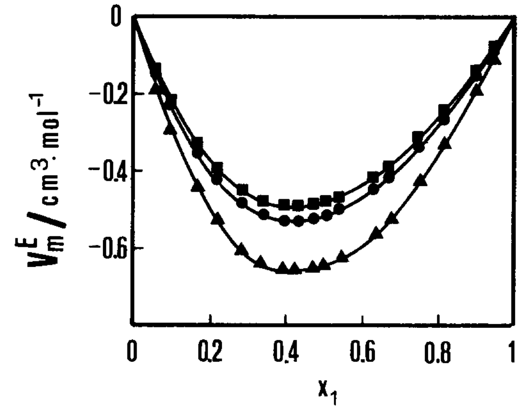

Fig. 4. Excess molar volumes, V_m^E, of propylene carbonate +1,2-epoxybutane. Solid symbols (■),(●),(▲) refer at (288.15, 298.15 and 313.15 K), respectively; solid curves are least-squares representations of results by Eq. (3).

4. The cell model

We have used the extended cell model proposed by Prigogine et al. [15,16], Salsburg and Kirkwood [17] and Rowlinson [18,19] to compute the excess molar enthalpies, H_m^E, and the excess molar volumes, V_m^E for the propylene +1,2-epoxybutane mixture. Looking to

Table 5

Densities, ρ, and excess molar volumes, V_m^E, of propylene carbonate(1) +1,2-epoxybutane(2) at (288.15, 298.15 and 313.15 K)

x_1	ρ/g·cm^{-3}	V_m^E/cm^3·mol^{-1}	x_1	ρ/g·cm^{-3}	V_m^E/cm^3·mol^{-1}
T =288.15 K					
0.0545	0.85691	−0.144	0.5023	1.02740	−0.483
0.0917	0.87143	−0.226	0.5487	1.04475	−0.469
0.1611	0.89824	−0.337	0.6352	1.07684	−0.420
0.2125	0.91799	−0.401	0.6728	1.09080	−0.395
0.2834	0.94498	−0.455	0.7572	1.12187	−0.319
0.3352	0.96458	−0.478	0.8159	1.14334	−0.252
0.3955	0.98732	−0.491	0.9019	1.17468	−0.144
0.4266	0.99904	−0.494	0.9402	1.18852	−0.086
0.4757	1.01744	−0.488			
T =298.15 K					
0.0545	0.84603	−0.154	0.5023	1.01657	−0.522
0.0917	0.86052	−0.238	0.5487	1.03391	−0.505
0.1611	0.88734	−0.360	0.6352	1.06606	−0.455
0.2125	0.90702	−0.422	0.6728	1.08000	−0.425
0.2834	0.93408	−0.486	0.7572	1.11112	−0.345
0.3352	0.95368	−0.512	0.8159	1.13260	−0.272
0.3955	0.97648	−0.530	0.9019	1.16393	−0.151
0.4266	0.98819	−0.532	0.9402	1.17783	−0.092
0.4757	1.00664	−0.530			
T =313.15 K					
0.0545	0.82956	−0.193	0.5023	1.00082	−0.647
0.0917	0.84418	−0.299	0.5487	1.01821	−0.628
0.1611	0.87120	−0.453	0.6352	1.05034	−0.562
0.2125	0.89098	−0.530	0.6728	1.06430	−0.527
0.2834	0.91817	−0.610	0.7572	1.09537	−0.424
0.3352	0.93783	−0.641	0.8159	1.11681	−0.334
0.3955	0.96067	−0.660	0.9019	1.14814	−0.191
0.4266	0.97240	−0.662	0.9402	1.16197	−0.116
0.4757	0.99087	−0.656			

Table 6

Least-squares parameters, a_k, Eq. (4), and standard deviation, $\sigma(Q_m^E)$, Eq. (5), of propylene carbonate(1) +1,2-epoxybutane(2) at (288.15, 298.15 and 313.15)K

Function	a_0	a_1	a_2	a_3	a_4	$\sigma(Q_m^E)$
T=288.15 K						
H_m^E/J·mol^{-1}	849.30	158.83	0	−367.36	−388.72	1.9
V_m^E/cm^3·mol^{-1}	−1.9362	0.5016	−0.3259	0.2611		0.0015
T=298.15 K						
H_m^E/J·mol^{-1}	821.22	125.73	0	−338.68	−467.80	1.6
V_m^E/cm^3·mol^{-1}	−2.0921	0.5129	−0.2775	0.2907		0.0014
T=313.15 K						
H_m^E/J·mol^{-1}	817.44	131.39	0	−232.32	−427.15	1.7
V_m^E/cm^3·mol^{-1}	−2.5958	0.6939	−0.3928	0.3043		0.0015

the cell model, the theoretical expression for H_m^E is

$$H_m^E = x_1x_2E_{11}z[-1.44\theta + 10.76(RT/zE_{11})^2 \cdot (-2\theta - \delta^2 + 4\delta\theta x_2 + 4x_1x_2\theta^2)] \quad (7)$$

where z is the number of the nearest neighbours in the quasi chemical-lattice model

$$\delta = (E_{22} - E_{11})/E_{11} \quad (8)$$

$$\theta = [E_{12} - (E_{11} + E_{22})/2]/E_{11} \quad (9)$$

$$E = E_{11} + E_{22} - 2E_{12} \quad (10)$$

and E_{11}, E_{22}, E_{12} are the interaction energies between molecules.

To test the experimental calorimetric data of this paper by Eq. (7), a value of z =8 was chosen and δ calculated from Eq. (8), with E_{11} and E_{22} evaluated by heat of vaporization reported in literature [8].

The values of θ is then obtained from Eq. (9) by substituting the excess enthalpy H_m^E and the mole fraction x_1 corresponding to the maximum in the H_m^E vs. x_1 curve.

Table 7 reports the interaction energies at 298.15 K between molecules for the mixtures obtained from this theory.

We have reported in this table only the results referring to the curve at 298.15 K E_{12} remains practically unchanged with temperature.

The expression for V_m^E, according to the cell model theory is

$$V_m^E = 2.03V^*x_1x_2RT/E_{11}(-2\theta - \delta^2 + 4\theta\delta x_2 + 4x_1x_2\theta^2) \quad (11)$$

with

$$V^* = (cr_{av})^3 \quad (12)$$

where c is an empirical factor accounting for approximations involved in evaluating r_{av}. V^* was obtained by the approximate Eq. (13) where r_{av} is the arithmetical mean of the distances r_1 and r_2 of molecules in pure liquids 1 and 2, as determined by the formula

$$r_1 = (\gamma_i/N_a)^{1/3} \quad (13)$$

where γ is the molar liquid volume and N_a is Avogadro's constant. This procedure was used by Kohler [20] in the calculation of the interchange energy in the lattice theory of solutions. The factor c was chosen so as to match the calculated and experimental values of V_m^E at the maximum.

Figs. 5 and 6 show the comparison between the Redlich–Kister fits and the cell model propylene carbonate +1,2-epoxybutane at 298.15 K.

Similar plots are obtained for the other temperatures.

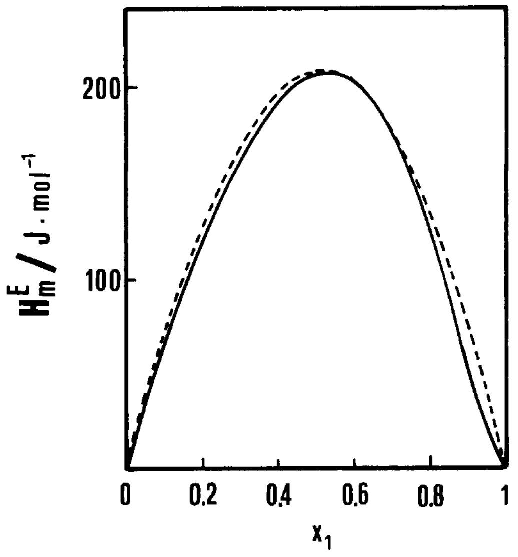

Fig. 5. Comparison between the Redlich–Kister fit (———), Eq. (3), and the cell model (- - -), Eq. (6), for H_m^E at 298.15 K of propylene carbonate +1,2-epoxybutane.

Table 7

Interaction energies E_{ij}/J·mol^{-1} between molecules for propylene carbonate +1,2-epoxybutane, Eq. (8)–(10), and empirical factor c, Eq. (13), at 298.15 K

E_{11}	E_{22}	E_{12}	E_{11}+E_{22}−2 E_{12}	c^a
3154	1825	2485	36	1.55

[a] at (288.15 and 313.15 K), c=1.53 and 1.87, respectively.

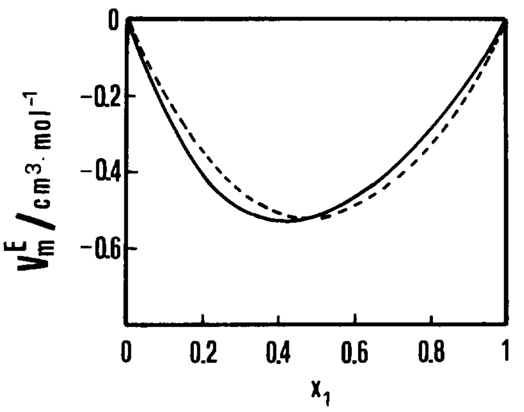

Fig. 6. Comparison between the Redlich–Kister fit (———), Eq. (3), and the cell model (- - -), Eq. (11), for V_m^E at 298.15 of propylene carbonate +1,2-epoxybutane.

5. Conclusions

Fig. 3 shows positive values for H_m^E for all temperatures.

This trend may be interpreted as the prevailing of intermolecular forces between similar molecules over the forces between the different molecules in the mixtures.

In fact, the structure of both propylene carbonate and 1,2-epoxybutane excludes the presence of hydrogen bonding between them, which is also confirmed by the results of the cell model, see Table 6, stressing the relatively high value of the interaction energy between ester molecules, E_{11}, and the positive value of $E_{11} + E_{22} - 2E_{12}$, which is, almost approximatively, proportional to H_m^E.

The values obtained for C_p^E are negative. Absolute values are very small, not exceeding 2 J·mol^{-1}·K^{-1}, which explain the scattering curve, see Fig. 2, for data at 288.15 K, since values of C_p^E compare with uncertainty on C_p.

This result agrees with the H_m^E vs x_1 curves at different temperatures, see Fig. 3, showing small decreases of H_m^E with the increase of temperature, with a maximum value of $\partial H_m^E/\partial T$ around 1 J·mol^{-1}·K^{-1}.

As to the V_m^E curves, it must be recalled that the excess molar volume depends on two distinct effects connected to energy interactions and geometric packaging of molecules after mixing, respectively.

The only energy effect would lead to volume increase after mixing, since $E_{11} + E_{22} - 2E_{12}$ is positive, that is pure components have stronger interactions than molecules in the mixture. Hence, packaging effect must predominate, with the reduction of volume after mixing, probably due to a possible overlapping of the three-atom ring of 1,2-epoxybutane with the four-atom ring of propylene carbonate.

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