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Excess molar enthalpies, densities and excess molar volumes for binary mixtures containing dimethyl carbonate or diethyl carbonate + 2,2,4-trimethylpentane at 288.15, 298.15, and 313.5 K, and at atmospheric pressure. Application of an extended cell model

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Abstract

Excess molar enthalpies, H_m^E, densities, ρ, and excess molar volumes, V_m^E, of dimethyl carbonate or diethyl carbonate + 2,2,4-trimethylpentane have been determined at 288.15, 298.15 and 313.15 K and at atmospheric pressure, using an LKB flow microcalorimeter and an Anton Paar digital density meter. For all mixtures, the H_m^E and V_m^E values are positive and increase slowly as the temperature is increased. The experimental values have been correlated by the Redlich–Kister equation and the adjustable parameters have been evaluated by least-squares analysis. The results are discussed qualitatively in terms of molecular interactions and have been interpreted by an extended cell model.

Keywords: Density; Enthalpy; Excess molar volume; Extended cell model

1. Introduction

Recently, alkyl carbonates were recognized to have good properties as solvents for a variety of extractions of industrial interest, especially in the synthesis of pharmaceuticals and agricultural chemicals. Moreover, there is a continuous need for reliable values of the physical properties of organic solvents, particularly of those involved in

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industrial processes. Thus, continuing our studies on the thermodynamic properties of alkyl carbonates [1–3], we report in this paper measurements of excess molar enthalpies, H_m^E, densities, ρ, and excess molar volumes, V_m^E, of binary mixtures containing dimethyl carbonate or diethyl carbonate + 2,2,4-trimethylpentane (isooctane). Measurements were performed at atmospheric pressure at temperatures of 288.15, 298.15 or 313.15 K.

The aim of the present investigation was to study the effects of temperature, chain length, and structure of the alkyl carbonates and 2,2,4-trimethylpentane on the properties of their binary mixtures.

Garcia de la Fuente et al. [4] have determined the excess molar volume of dimethyl carbonate + 2,2,4-trimethylpentane at 298.15 K. The mean deviation between the two sets of data is of the order of ±0.5% over the entire range of mole fraction. To our knowledge, no other measurements on these mixtures have been reported in the literature.

2. Experimental

2.1. Chemicals

Dimethyl carbonate, diethyl carbonate and 2,2,4-trimethylpentane were purchased from Aldrich Chemicals with 99%, +99% and 99.8% purities, respectively. All chemicals, stored in dark bottles over molecular sieves (Union Carbide type 4A, 1/16-in. pellets), were used directly without further purification.

Table 1 lists the measured densities, ρ, and the corresponding literature values [4, 5].

2.2. Calorimetric measurements

Measurements of excess molar enthalpies, H_m^E, were carried out in an LKB flow microcalorimeter [6] (LKB Produkter, Bromma, Sweden) following the procedure described in a previous paper [7]. The performance of the apparatus was checked by measuring the H_m^E value of the well-investigated cyclohexane–hexane test mixture [8]. Deviations between our data and those in the literature were less than 0.5% over the central range of mole fractions. The temperature in the circulator bath was kept

Table 1
Densities, ρ, of pure chemicals at 298.15 K

Component	$\rho/g\ cm^{-3}$	
	Expt.	Lit.
Dimethyl carbonate	1.06328	1.0635 [4]
Diethyl carbonate	0.96925	0.96926 [5]
2,2,4-Trimethylpentane	0.68766	0.68769 [5]

constant to within ±0.005 K and the estimated error in the mole fraction is of the order $\delta x = \pm 0.0005$.

2.3. Volumetric measurements

Mole fractions of solutions were prepared by mass in stoppered bottles using a Mettler balance (model AE 160).

The heavier component was loaded first to minimize the errors in composition due to evaporation. This leads to an accuracy of the order of 2×10^{-4} in the mole fraction. All measurements were corrected for buoyancy and for evaporation of the components.

Excess molar volumes, $V^{\mathrm{E}}_{\mathrm{m}}$, were calculated from density measurements using a digital vibrating tube density meter (Anton Paar, model DMA 60/602, Graz, Austria) with a resolution of 1×10^{-6} g cm^{-3} and equipped with three temperature sensors (Anton Paar DT-100(15-25-40)). Temperature oscillations were kept within ±0.01 K. An external Hetotherm bath circulator (Heto, type 01 DBT 623) was used with a temperature control interval of ±0.005 K.

The characteristics of the density meter and the operating procedure were the same as reported by other authors [9]. Dry air and freshly bidistilled water were used as standards for calibration of the apparatus before each series of measurements. The densities, ρ, have an estimated accuracy of 1×10^{-5} g cm^{-3}.

The test system benzene + cyclohexane [10] at 298.15 K was used to evaluate the accuracy of the experimental technique and results were in agreement with data in the literature (discrepancy less than 0.5% over the entire range of mole fraction).

3. Correlation of the calorimetric data

The experimental values of $H^{\mathrm{E}}_{\mathrm{m}}$ and $V^{\mathrm{E}}_{\mathrm{m}}$ are listed in Tables 2 and 3 as a function of the mole fraction of alkyl carbonate (component 1) and are reported graphically in Figs. 1 and 2. The Redlich–Kister equation

$$Q^{\mathrm{E}}_{\mathrm{m}} = x_1 x_2 \sum_{k \geq 0} a_k (x_1 - x_2)^k \tag{1}$$

where $Q^{\mathrm{E}}_{\mathrm{m}} = H^{\mathrm{E}}_{\mathrm{m}}/(\mathrm{J\ mol^{-1}})$ or $Q^{\mathrm{E}}_{\mathrm{m}} = V^{\mathrm{E}}_{\mathrm{m}}/(\mathrm{cm^3\ mol^{-1}})$, was fitted to the results by a least-squares method, with all points weighted equally. Values of the adjustable parameters, a_k, are reported in Tables 2 and 3. These values correspond to the minimum of the standard deviations $\sigma(Q^{\mathrm{E}}_{\mathrm{m}})$

$$\sigma(Q^{\mathrm{E}}_{\mathrm{m}}) = |\phi/(N-n)|^{0.5} \tag{2}$$

where N is the number of experimental points and n is the number of adjustable parameters; ϕ is the objective function defined as

$$\phi = \sum_{k=1}^{N} \eta_k^2 \tag{3}$$

where $\eta = Q^{\mathrm{E}}_{\mathrm{calc}} - Q^{\mathrm{E}}$; $Q^{\mathrm{E}}_{\mathrm{calc}}$ was determined from the right-hand side of Eq. (1).

Table 2
Experimental excess molar enthalpies, H_m^E, adjustable parameters a_k, and standard deviation $\sigma(H_m^E)$, of dialkyl carbonates + 2,2,4-trimethylpentane at 288.15, 298.15 or 313, 15 K

x_1	H_m^E/J mol^{-1}	x_1	H_m^E/J mol^{-1}
Dimethyl carbonate (l) + 2,2,4-trimethylpentane (2)			
$T = 288.15$ K			
0.0756	453	0.7464	1460
0.1405	818	0.7969	1261
0.1969	1113	0.8547	954
0.2463	1313	0.8870	745
0.3290	1591	0.9217	492
0.3953	1727	0.9401	338
0.4951	1817	0.9593	209
0.5667	1807	0.9792	90
0.6623	1675		
$a_0 = 7307.8$; a_1, $a_2 = 0$; $a_3 = 3716.4$; $a_4 = 2905.9$; $a_5 = -4968.7$; $a_6 = -6021.4$; $\sigma(H_m^E)$/J mol^{-1} = 8.2			
$T = 298.15$ K			
0.0755	539	0.7463	1528
0.1404	861	0.7968	1327
0.1968	1124	0.8547	1021
0.2463	1326	0.8869	783
0.3289	1677	0.9217	535
0.3952	1852	0.9401	368
0.4950	1973	0.9592	224
0.5666	1942	0.9792	99
0.6622	1773		
$a_0 = 7917.4$; $a_1 = 0$; $a_2 = -2318.6$; $a_3 = 5790.8$; $a_4 = 6540.2$; $a_5 = -8680.1$; $a_6 = -5911.7$; $\sigma(H_m^E)$/J mol^{-1} = 8.9			
$T = 313.15$ K			
0.0754	490	0.7462	1572
0.1404	886	0.7967	1352
0.1967	1153	0.8546	1017
0.2462	1395	0.8869	785
0.3288	1688	0.9216	521
0.3947	1863	0.9400	370
0.4949	1972	0.9592	222
0.5664	1964	0.9792	96
0.6621	1840		
$a_0 = 7915.7$; $a_1 = 764.0$; a_2, $a_3 = 0$; $a_4 = 1079.9$; $a_5 = 1969.9$; $a_6 = -3783.5$; $a_7 = -4751.0$; $\sigma(H_m^E)$/J mol^{-1} = 8.4			
Diethyl carbonate(l) + 2,2,4-trimethylpentane(2)			
$T = 288.15$ K			
0.0537	203	0.6715	995
0.1019	385	0.7315	869
0.1455	582	0.8034	668
0.1850	725	0.8449	517
0.2541	920	0.8910	352
0.3123	1051	0.9160	234
0.4052	1164	0.9294	198

Table 2 (*Continued*)

x_1	H_m^E/J mol^{-1}	x_1	H_m^E/J mol^{-1}
0.4759	1175	0.9703	68
0.5767	1143		
$a_0 = 4703.1$; $a_1 = -416.8$; $a_2 = 454.5$; $a_3 = -281.1$; $a_4 = -2723.5$; $\sigma(H_m^E)$/J mol$^{-1} = 9.2$			
$T = 298.15$ K			
0.0537	195	0.6715	1047
0.1020	371	0.7316	929
0.1456	561	0.8035	711
0.1851	705	0.8450	546
0.2541	932	0.8911	359
0.3124	1043	0.9160	258
0.4053	1170	0.9424	158
0.4760	1220	0.9703	71
0.5768	1189		
$a_0 = 4854.7$; $a_1 = 0$; $a_2 = 311.7$; $a_3 = -594.3$; $a_4 = -2805.3$; $\sigma(H_m^E)$/J mol$^{-1} = 9.0$			
$T = 313.15$ K			
0.0538	205	0.6717	1068
0.1020	389	0.7317	933
0.1456	568	0.8036	716
0.1852	703	0.8451	552
0.2453	902	0.8911	378
0.3125	1063	0.9160	262
0.4055	1206	0.9424	160
0.4762	1252	0.9704	74
0.5769	1223		
$a_0 = 4968.7$; $a_1, a_2 = 0$; $a_3 = -631.0$; $a_4 = -2385.3$; $\sigma(H_m^E)$/J mol$^{-1} = 9.2$			

Table 3

Densities, ρ, excess molar volumes, V_m^E, adjustable parameters, a_k, and standard deviation, $\sigma(V_m^E)$, of diethyl carbonate + 2,2,4-trimethylpentane at 288.15, 298.15 or 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}
Dimethyl carbonate (1) + 2,2,4-trimethylpentane(2)					
$T = 288.15$ K					
0.0032	0.70156	0.159	0.6991	0.89704	0.618
0.1274	0.71984	0.510	0.7345	0.91360	0.564
0.2937	0.75807	0.807	0.7676	0.93004	0.501
0.3714	0.77897	0.854	0.8078	0.95115	0.434
0.4452	0.80100	0.851	0.8753	0.99019	0.288
0.5206	0.82596	0.815	0.9313	1.02646	0.157
0.5901	0.85139	0.757	0.9619	1.04784	0.091
0.6230	0.86442	0.720	0.9853	1.06503	0.041
0.6638	0.88144	0.663			

Table 3 (*Continued*)

x_1	ρ/g cm^{-3}	V^E_m/cm^3 mol^{-1}	x_1	ρ/g cm^{-3}	V^E_m/cm^3 mol^{-1}
$a_0 = 3.3167$; $a_1 = -1.1227$; $a_2 = 0.5364$; $a_3 = -0.3324$; $\sigma(V^E_m)$/cm^3 mol^{-1} = 0.0036					
$T = 298.15$ K					
0.0047	0.69597	0.242	0.6967	0.88491	0.671
0.1460	0.71501	0.602	0.7329	0.90161	0.609
0.2840	0.74639	0.863	0.7701	0.91985	0.534
0.3534	0.76447	0.914	0.8194	0.94590	0.427
0.4561	0.79456	0.917	0.8777	0.97967	0.293
0.5306	0.8192	0.873	0.9323	1.01467	0.159
0.5757	0.83546	0.833	0.9602	1.03396	0.096
0.6277	0.85558	0.772	0.9857	1.05240	0.038
0.6623	0.86985	0.726			
$a_0 = 3.5939$; $a_1 = -1.18109$; $a_2 = 0.4453$; $a_3 = -0.5051$; $\sigma(V^E_m)$/cm^3 mol^{-1} = 0.0030					
$T = 313.15$ K					
0.0032	0.68058	0.205	0.6991	0.86909	0.749
0.1275	0.69800	0.630	0.7345	0.88516	0.682
0.2937	0.73462	0.999	0.7676	0.90117	0.602
0.3714	0.75475	1.056	0.8078	0.92180	0.504
0.4452	0.77593	1.065	0.8753	0.95970	0.334
0.5206	0.80008	1.014	0.9313	0.99503	0.171
0.5901	0.82474	0.934	0.96190	1.01576	0.098
0.6231	0.83736	0.890	0.9853	1.03245	0.040
0.6638	0.85389	0.816			
$a_0 = 4.1229$; $a_1 = -1.4312$; $a_2 = 0.4006$; $a_3 = -0.6311$; $\sigma(V^E_m)$/cm^3 mol^{-1} = 0.0043					
Diethyl carbonate (1) + 2,2,4-trimethylpentane(2)					
$T = 288.15$ K					
0.0213	0.70011	0.050	0.6049	0.84451	0.312
0.0785	0.71185	0.168	0.6690	0.86403	0.276
0.1972	0.73781	0.319	0.6966	0.87277	0.251
0.3030	0.76276	0.380	0.7676	0.89593	0.198
0.3694	0.77936	0.389	0.8250	0.91553	0.148
0.4282	0.79466	0.386	0.9007	0.94255	0.089
0.4946	0.81268	0.372	0.9432	0.95836	0.055
0.5397	0.82540	0.348	0.9797	0.97235	0.027
0.5719	0.83475	0.327			
$a_0 = 1.4584$; $a_1 = -0.7794$; $a_2 = 0.2495$; $\sigma(V^E_m)$/cm^3 mol^{-1} = 0.0036					
$T = 298.15$ K					
0.4694	0.69703	0.107	0.6126	0.83678	0.352
0.1210	0.71241	0.245	0.6548	0.84955	0.320
0.2145	0.73300	0.356	0.7100	0.86676	0.276
0.2706	0.74600	0.396	0.7471	0.87875	0.243
0.37006	0.77027	0.427	0.8301	0.90669	0.164
0.4393	0.78817	0.424	0.9130	0.93623	0.085
0.4979	0.80397	0.411	0.9381	0.94551	0.066
0.5365	0.81474	0.392	0.9890	0.96488	0.019
0.5760	0.82607	0.369			

Table 3 (*Continued*)

x_1	ρ/g cm^{-3}	V^E_m/cm^3 mol^{-1}	x_1	ρ/g cm^{-3}	V^E_m/cm^3 mol^{-1}
$a_0 = 1.6326$; $a_1 = -0.7634$; $a_2 = 0.1296$; $\sigma(V^E_m)$/cm^3 mol^{-1} = 0.0032					
$T = 313.15$ K					
0.0213	0.67932	0.060	0.6049	0.81941	0.407
0.0785	0.69064	0.205	0.6690	0.83850	0.356
0.1972	0.71576	0.391	0.6966	0.84698	0.333
0.3030	0.73992	0.477	0.7676	0.86962	0.263
0.3694	0.75600	0.497	0.8250	0.88879	0.199
0.4282	0.77087	0.496	0.9007	0.91526	0.117
0.4946	0.78842	0.475	0.9432	0.93074	0.069
0.5397	0.80080	0.450	0.9797	0.94446	0.029
0.5719	0.80986	0.434			
$a_0 = 1.8864$; $a_1 = -0.8827$; $a_2 = 0.2030$; $\sigma(V^E_m)$/cm^3 mol^{-1} = 0.0026					

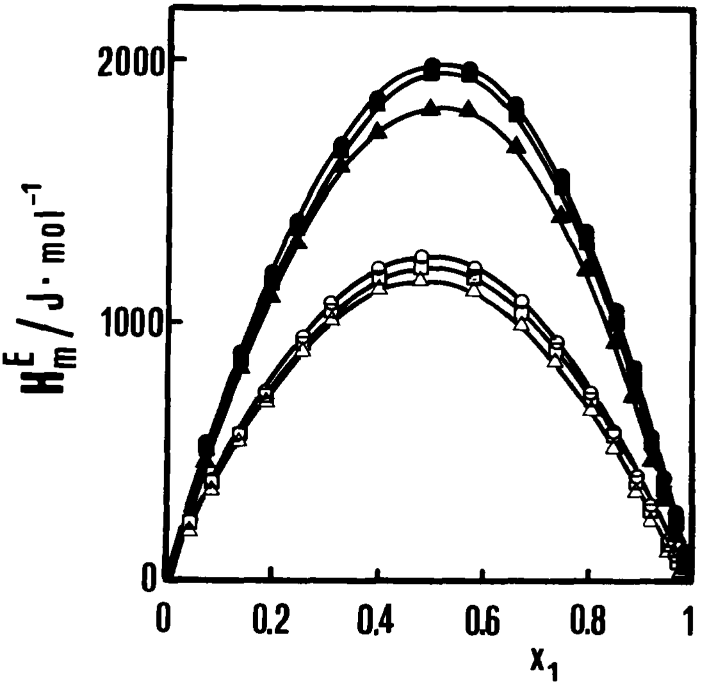

Fig. 1. Excess molar enthalpies, H^E_m, of dialkyl carbonates + 2,2,4-trimethylpentane. Solid symbols (▲), (■), (●) refer to dimethyl carbonate + 2,2,4-trimethylpentane at 288.15, 298.15 or 313, 15 K; open symbols (△), (□), (○) refer to diethyl carbonate + 2,2,4-trimethylpentane at 288.15, 298.15 or 313.15 K, respectively. Solid curves are least-squares representations of results by Eq. (1).

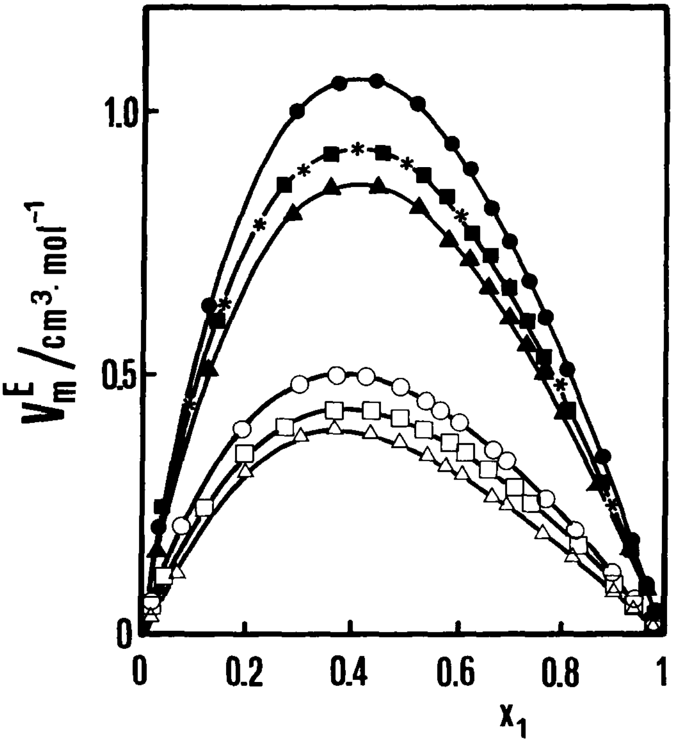

Fig. 2. Excess molar volumes, V_m^E, of dialkyl carbonates + 2,2,4-trimethylpentane. Solid symbols (▲), (■), (●) refer to dimethyl carbonate + 2,2,4-trimethylpentane at 288.15, 298.15 or 313.15 K; open symbols (△), (□), (○) refer to diethyl carbonate + 2,2,4-trimethylpentane at 288.15, 298.15 or 313, 15 K, respectively; (*) refers to Garcia de la Fuente et al. Ref. [4]; solid curves are least-squares representations of results by Eq. (1).

4. The cell model

An extended cell model, elaborated by Prigogine et al. [11–13], Salsburg and Kirkwood [14] and Rowlinson [15, 16], was used to compute the excess molar enthalpies, H_m^E, and the excess molar volumes, V_m^E. According to the cell model, the theoretical expression for H_m^E is

$$H_m^E = x_1 x_2 E_{11} z[-1.44\theta + 10.76(RT/zE_{11})^2(-2\theta - \delta^2 + 4\delta\theta x_2 + 4x_1 x_2 \theta^2)] \quad (4)$$

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

$$\delta = (E_{22} - E_{11})/E_{11} \quad (5)$$

$$\theta = [E_{12} - (E_{11} + E_{22})/2]/E_{11} \quad (6)$$

$$E = E_{11} + E_{22} - 2E_{12} \quad (7)$$

and E_{11}, E_{22}, E_{12} are the interaction energies between molecules. To test the experimental calorimetric data of this paper by Eq. (4), a value of $z=8$ was chosen and δ calculated from Eq. (5), with E_{11} and E_{22} evaluated by heat of vaporization [5]. The heat of vaporization λ of dimethyl carbonate is not quoted in the literature and it has been calculated from vapor–liquid measurements [17] using the Clausius–Clapeyron expression

$$\log P^0 = C + D/T \tag{8}$$

where $\lambda = -DR$ and R being the gas constant. Constants C and D were obtained by a least-squares method.

The values of θ were then obtained from Eq. (7) 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 4 reports the interaction energies between molecules for the mixtures obtained from this theory.

According to the extended cell model, the expression for V_m^E is

$$V_m^E = 2.03\,V^* x_1 x_2 RT/E_{11}(-2\theta - \delta^2 + 4\theta\delta x_2 + 4x_1 x_2 \theta^2) \tag{9}$$

with

$$V^* = (cr_{av})^3 \tag{10}$$

where c is an empirical factor accounting for approximations involved in evaluating r_{av}. V^* was obtained by the approximate Eq. (10) 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

$$r_i = (\gamma_i/N_a)^{1/3} \tag{11}$$

where γ is the molar liquid volume and N_a is Avogadro's constant. This procedure was used by Kohler [18] 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.

Values of parameters obtained by the cell model theory at the temperature of 298.15 K are reported in Table 4.

Figs. 3 and 4 show the comparison between the Redlich–Kister fits and the cell model for dialkyl carbonates + 2,2,4-trimethylpentane at 298.15 K. Similar plots are obtained for the other temperatures.

Table 4
Interaction energies E_{ij}/J mol^{-1} between molecules for mixtures, Eqs. (5)–(7), and empirical factors c, Eq. (10) at 298.15 K

Mixture	E_{11}	E_{22}	E_{12}	$E_{11}+E_{22}-2\,E_{12}$	c
Dimethyl carbonate + 2,2,4-trimethylpentane	2094.1 [a]	2007.2 [b]	2011.5	78.3	1.8
Diethyl carbonate + 2,2,4-trimethylpentane	2489.6 [b]	2007.2 [b]	2224.2	48.4	1.7

[a] Obtained from Clausius–Clapeyron, Ref. [17].
[b] Ref. [5].

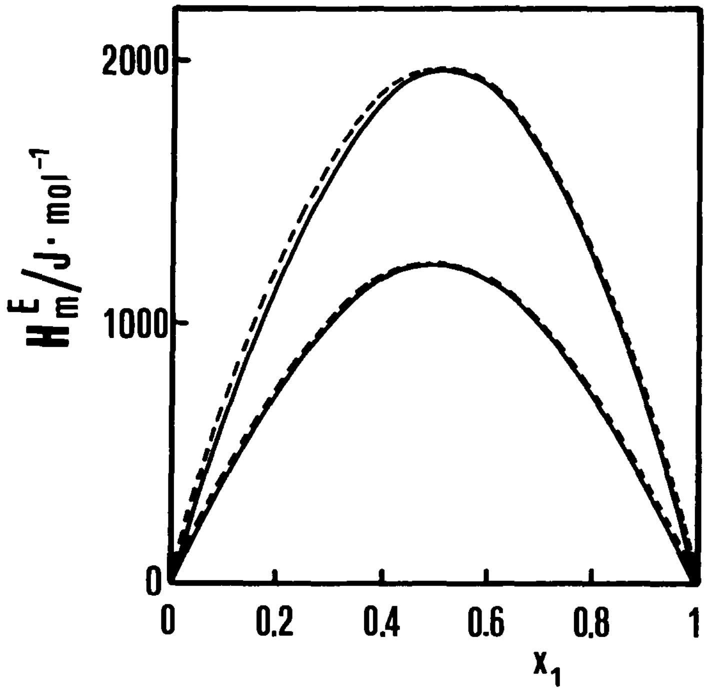

Fig. 3. Comparison between the Redlich–Kister fit (——), Eq. (1), and the cell model (- - -), Eq. (4), for H^E_m at 298.15 K for dialkyl carbonates + 2,2,4-trimethylpentane.

5. Conclusions

Figs. 1 and 2 point out the temperature behavior of H^E_m and V^E_m for the mixtures dimethyl carbonate or diethyl carbonate + 2,2,4-trimethylpentane. All mixtures show an increase of both H^E_m and V^E_m as temperature is increased. More significant changes with temperature are obtained for dimethyl carbonate + 2,2,4-trimethylpentane and for values of V^E_m.

Assuming that H^E_m is approximately proportional to $E_{11} + E_{22} - 2E_{12}$, the calculated values of the interaction parameters in Table 4 can reproduce the different trends of H^E_m for the mixtures.

In going from dimethyl carbonate to diethyl carbonate the value of E_{11} is increased, since the OC_2H_5 group has a stronger electron-donor character than OCH_3, which leads to higher polarization of the double bond in the C=O group and hence to stronger interactions between molecules. However, this effect gives rise to an increase in $2E_{12}$ that is larger than the corresponding increase in E_{11}. Thus H^E_m is lower for the diethyl carbonate + 2,2,4-trimethylpentane mixtures.

Analogously, positive values of $E_{11} + E_{22} - 2E_{12}$ indicate a stronger interaction energy in the pure state than in the mixture. The values are in agreement with the sequence of positive V^E_m, reported in Fig. 2.

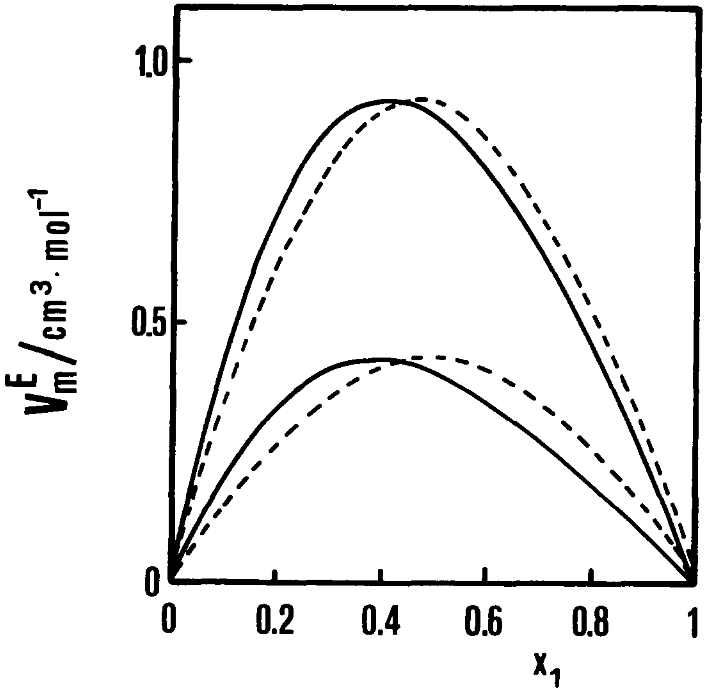

Fig. 4. Comparison between the Redlich–Kister fit (——), Eq. (1), and the cell model (– – –), Eq. (9), for V_m^E at 298.15 K for dialkyl carbonates + 2,2,4-trimethylpentane.

Finally, the values of H_m^E and V_m^E predicted by the cell model are in good agreement with the experimental data, if allowance is made for the approximations involved in the calculations.

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