J. Chem. Eng. Data **2004,** *49,* 923−927

pρTx Data for the Dimethyl Carbonate + Decane System

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Density data of the dimethyl carbonate + decane system are reported from $T = 283.15$ to 328.15 K and $p = 0.1$ to 40 MPa over the whole composition range. An automated measuring device, based on the vibrating tube principle, is used. Data are correlated through a temperature-dependent Tait equation that allows us to derive isothermal compressibilities and isobaric thermal expansivities. Excess quantities for molar volume, isothermal compressibility, and isobaric thermal expansivity are also given.

Introduction

Dialkyl carbonates have been proposed as alternative lubricants in low environmental impact air conditioners because of their miscibility with HFCs, favorable viscosity, and thermal stability.[1−3] Besides their intrinsic interest, thermodynamic properties of the dialkyl carbonate + *n*-alkane mixtures allow us to obtain parameters for molecular models that are useful to predict thermophysical properties for HFCs + carbonate systems.[4,5]

In this work, a study on the densities of the dimethyl carbonate + decane system from 283.15 to 328.15 K and 0.1 to 40 MPa is presented. The data were correlated as a function of temperature and pressure, allowing us to derive isobaric thermal expansivities and isothermal compressibilities. In addition, excess quantities for the above properties were calculated. The global volumetric behavior thus observed was compared to previous experimental *pρTx* studies on other dialkyl carbonate + alkane systems.[6−9]

Experimental Section

Materials. Dimethyl carbonate (purity, 99 mol %) was supplied from Aldrich and decane (purity, 99 mol %) from Sigma. The stated purities of the chemicals were checked by gas chromatography analysis. Both liquids were degassed and dried over Fluka 0.4-nm molecular sieves before use. Mixtures were prepared by weighing to an uncertainty in a mol fraction of ±0.0001 using a AE-240 balance.

Apparatus and Procedure. Densities were measured using an Anton-Paar DMA-512P high-pressure cell based on the vibrating-tube principle. Temperature and pressure control as well as data acquisition were fully automatized. The uncertainties in *T* and *p* were ±0.01 K and ±0.01 MPa, respectively. The details of the experimental device together with the calibration procedure have been explained in a previous paper.[10] The uncertainty in density data was estimated to be about $\pm 0.0003\ \text{g·cm}^{-3}$, whereas repeatability was quoted to $\pm 0.00003\ \text{g·cm}^{-3}$.

Results and Discussion

Experimental densities were obtained within the working range in pressure and temperature steps of 2.5 MPa and 5 K, respectively. Selected data for pure liquids can be found in Table 1. Comparison with literature-recommended data for decane[11] results in an absolute mean difference of $0.00015\ \text{g·cm}^{-3}$. For dimethyl carbonate, only two sources of data at high pressures are available.[6,7] Good agreement is found between the data of ref 6 and ours (absolute mean difference of $0.00018\ \text{g·cm}^{-3}$), whereas relatively large discrepancies are encountered with data of ref 7; therefore, we choose to take data of the former for further comparison on derived properties. In Table 2, data for the mixtures are shown. For each sample, they were fitted to the Tait equation with temperature-dependent parameters[11]

$$\rho(p,T) = \frac{\rho(0.1,T)}{1 - c_0 \ln\left(\dfrac{B(T) + p}{B(T) + 0.1\text{MPa}}\right)} \tag{1}$$

where c_0 is a temperature-independent parameter and $B(T)$ is a polynomial of the form

$$B(T) = \sum_{i=0}^{2} b_i \left(\frac{T - T_0}{100}\right)^i \tag{2}$$

$\rho(0.1,T)$ are the densities at 0.1 MPa, interpolated by a third-order polynomial form

$$\rho(0.1,T) = \sum_{i=1}^{4} a_i \left(\frac{T - T_0}{100}\right)^{i-1} \tag{3}$$

with $T_0 = 273.15$ K in all cases. The final values of the parameters together with standard deviations *s* of the fits are listed in Table 3.

At atmospheric pressure, *x*dimethyl carbonate + (1 − *x*)decane presents, below ambient temperature, partial miscibility of the upper critical solution temperature type, i.e., the *Tx* liquid−liquid equilibrium curve presents a maximum, corresponding to a critical point. The critical coordinates are $x_c = 0.668$ and $T_c = 287.27$ K, whereas the slope of the critical line $\mathrm{d}T_c/\mathrm{d}p$ is equal to 0.176 K/MPa.[12] This implies that, for some mixtures, liquid−liquid-phase separation can be found within the working range. Unfor-

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10.1021/je0342320

Table 1. Densities ρ of the Pure Liquids at Selected Pressures p and Temperatures T

	ρ/g·cm^{-3}									
p/MPa	283.15 K	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15K	323.15 K	328.15 K
					Dimethyl Carbonate					
0.1	1.08317	1.07643	1.06976	1.06310	1.05641	1.04966	1.04286	1.03604	1.02921	1.02224
5	1.08733	1.08080	1.07422	1.06770	1.06117	1.05459	1.04794	1.04130	1.03463	1.02792
10	1.09139	1.08502	1.07858	1.07221	1.06582	1.05939	1.05291	1.04644	1.03995	1.03345
15	1.09534	1.08907	1.08276	1.07652	1.07029	1.06400	1.05767	1.05136	1.04504	1.03869
20	1.09913	1.09300	1.08683	1.08070	1.07459	1.06843	1.06222	1.05608	1.04991	1.04371
25	1.10279	1.09679	1.09073	1.08474	1.07874	1.07265	1.06660	1.06060	1.05458	1.04853
30	1.10639	1.10048	1.09450	1.08862	1.08275	1.07680	1.07082	1.06495	1.05907	1.05314
35	1.10985	1.10403	1.09819	1.09239	1.08660	1.08078	1.07493	1.06915	1.06339	1.05757
40	1.11317	1.10750	1.10174	1.09604	1.09035	1.08463	1.07887	1.07321	1.06756	1.06187
					Decane					
0.1	0.73740	0.73369	0.72994	0.72619	0.72246	0.71865	0.71479	0.71099	0.70717	0.70330
5	0.74081	0.73719	0.73353	0.72988	0.72623	0.72252	0.71883	0.71508	0.71140	0.70767
10	0.74417	0.74063	0.73708	0.73352	0.72998	0.72636	0.72276	0.71913	0.71555	0.71197
15	0.74739	0.74393	0.74050	0.73700	0.73355	0.73001	0.72652	0.72299	0.71949	0.71603
20	0.75049	0.74714	0.74376	0.74037	0.73698	0.73355	0.73014	0.72669	0.72328	0.71987
25	0.75352	0.75022	0.74692	0.74360	0.74029	0.73701	0.73361	0.73022	0.72690	0.72357
30	0.75643	0.75319	0.74998	0.74672	0.74346	0.74013	0.73695	0.73365	0.73037	0.72710
35	0.75927	0.75611	0.75294	0.74972	0.74654	0.74326	0.74017	0.73693	0.73371	0.73051
40	0.76202	0.75892	0.75579	0.75267	0.74950	0.74632	0.74325	0.74005	0.73693	0.73379

Table 2. Densities ρ of xDimethyl Carbonate + (1 − x)Decane at Selected Pressures p and Temperatures T

	ρ/g·cm^{-3}								
p/MPa	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15K	323.15 K	328.15 K
					$x = 0.0676$				
0.1	0.74197	0.73816	0.73431	0.73048	0.72653	0.72258	0.71868	0.71471	0.71069
10	0.74910	0.74545	0.74180	0.73815	0.73445	0.73076	0.72704	0.72336	0.71966
20	0.75573	0.75225	0.74875	0.74530	0.74178	0.73828	0.73475	0.73125	0.72774
30	0.76188	0.75856	0.75523	0.75190	0.74849	0.74523	0.74184	0.73846	0.73512
40	0.76771	0.76448	0.76128	0.75805		0.75165	0.74836	0.74515	0.74193
					$x = 0.2303$				
0.1	0.76795	0.76384	0.75970	0.75557	0.75135	0.74710	0.74290	0.73881	0.73447
10	0.77545	0.77154	0.76762	0.76368	0.75973	0.75575	0.75173	0.74790	0.74393
20	0.78240	0.77869	0.77495	0.77121	0.76745	0.76369	0.75987	0.75620	0.75249
30	0.78887	0.78531	0.78173	0.77815	0.77452	0.77099	0.76735	0.76383	0.76025
40	0.79493	0.79149	0.78805	0.78461	0.77852	0.77772	0.77420	0.77085	0.76741
					$x = 0.2934$				
0.1	0.78023	0.77599	0.77164	0.76736	0.76300	0.75866	0.75431	0.74994	0.74553
10	0.78782	0.78379	0.77968	0.77558	0.77151	0.76750	0.76340	0.75930	0.75520
20	0.79487	0.79101	0.78710	0.78323	0.77937	0.77554	0.77165	0.76780	0.76394
30	0.80143	0.79771		0.79027	0.78653	0.78296	0.77922	0.77553	0.77185
40	0.80754	0.80397	0.80040	0.79682		0.78979	0.78619	0.78266	0.77913
					$x = 0.4055$				
0.1	0.80452	0.79994	0.79537	0.79078	0.78620	0.78163	0.77708	0.77241	0.76774
10	0.81234	0.80799	0.80365	0.79932	0.79499	0.79074	0.78645	0.78210	0.77774
20	0.81959	0.81546	0.81130	0.80720	0.80310	0.79907	0.79497	0.79090	0.78678
30	0.82632	0.82234	0.81838	0.81445	0.81051	0.80672	0.80280	0.79887	0.79498
40	0.83265	0.82880	0.82500	0.82119		0.81378	0.80998	0.80624	0.80250
					$x = 0.5020$				
0.1	0.82972	0.82483	0.81995	0.81509	0.81029	0.80545	0.80057	0.79568	0.79073
10	0.83773	0.83307	0.82844	0.82386	0.81930	0.81481	0.81024	0.80566	0.80104
20	0.84478	0.84072	0.83629	0.83194	0.82762	0.82333	0.81901	0.81472	0.81036
30		0.84780	0.84357	0.83939	0.83523	0.83119	0.82705	0.82291	0.81878
40		0.85444	0.85035	0.84629		0.83842	0.83442	0.83050	0.82654
					$x = 0.5476$				
0.1	0.84328	0.83817	0.83319	0.82819	0.82317	0.81821	0.81318	0.80816	0.80307
10	0.85140	0.84656	0.84177	0.83703	0.83233	0.82767	0.82296	0.81826	0.81356
20		0.85435	0.84977	0.84525	0.84077	0.83631	0.83186	0.82741	0.82299
30		0.86157	0.85718	0.85282	0.84852	0.84429	0.84000	0.83574	0.83150
40		0.86838	0.86411	0.85991		0.85166	0.84753	0.84343	0.83936
					$x = 0.5883$				
0.1		0.85108	0.84593	0.84082	0.83571				
10		0.85946	0.85459	0.84974	0.84495				
20		0.86728	0.86262	0.85802	0.85346				
30		0.87453	0.87004	0.86563	0.86127				
40			0.87698	0.87272					
					$x = 0.7101$				
0.1	0.90171	0.89639	0.89082	0.88521	0.87957	0.87400	0.86841	0.86269	0.85701
10		0.90511	0.89975	0.89446	0.88919	0.88394	0.87862	0.87327	0.86793
20		0.91324	0.90810	0.90307	0.89805	0.89308	0.88803	0.88295	0.87785
30		0.92082	0.91587	0.91102	0.90621	0.90147	0.89664	0.89178	0.88687
40			0.92309	0.91842		0.90927	0.90462	0.89997	0.89522

Table 2 (Continued)

	ρ/g·cm^{-3}								
p/MPa	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	318.15K	323.15 K	328.15 K 0
					$x = 0.7993$				
0.1		0.93674	0.93088	0.92509	0.91930	0.91346	0.90756	0.90172	0.89568
10		0.94544	0.93991	0.93441	0.92893	0.92344	0.91790	0.91240	0.90686
20		0.95346	0.94828	0.94303	0.93783	0.93263	0.92738	0.92217	0.91693
30			0.95606	0.95101	0.94602	0.94103	0.93602	0.93108	0.92610
40				0.95841		0.94881	0.94400	0.93925	0.93448
					$x = 0.9062$				
0.1	1.00468	0.99835	0.99218	0.98593	0.97975	0.97348	0.96713	0.96079	0.95430
10	1.01319	1.00719	1.00124	0.99533	0.98947	0.98352	0.97756	0.97162	0.96560
20	1.02119	1.01543	1.00974	1.00409	0.99848	0.99281	0.98716	0.98153	0.97583
30	1.02863	1.02308	1.01762	1.01220	1.00683	1.00136	0.99597	0.99060	0.98517
40	1.03561	1.0308	1.02501	1.01978		1.00935	1.00415		0.99379

Table 3. Fitting Coefficients of Equation 1 for xDimethyl Carbonate + (1 − x)Decane and Standards Deviations s

	dimethyl carbonate	decane	$x = 0.0676$	$x = 0.2303$
c_0	0.088445	0.095624	0.093003	0.091719
b_0/MPa	120.2580	105.7950	101.3620	98.7657
b_1/MPa K^{-1}	−93.1567	−68.1881	−63.7453	−66.6930
b_2/MPa·K^{-2}	23.1718	16.0410	10.5807	15.0099
a_1/g·cm^{-3}	1.09659	0.74478	0.75330	0.77990
a_2/g·cm^{-3}·K^{-1}	−0.13506	−0.07341	−0.07465	−0.07762
a_3/g·cm^{-3}·K^{-2}	0.00778	−0.00367	−0.00511	−0.01540
a_4/g·cm^{-3}·K^{-3}	−0.01452			0.01147
s/g·cm^{-3}	0.00007	0.00003	0.00003	0.00003

	$x = 0.2934$	x=0.4055	$x = 0.5020$	$x = 0.5476$
c_0	0.090817	0.090562	0.090689	0.091337
b_0/MPa	98.2891	98.4643	100.2940	99.2257
b_1/MPa K^{-1}	−67.9574	−70.4616	−75.7084	−66.6482
b_2/MPa·K^{-2}	15.0873	17.2183	21.0724	10.3786
a_1/g·cm^{-3}	0.79300	0.81856	0.84486	0.85888
a_2/g·cm^{-3}·K^{-1}	−0.08459	−0.09511	−0.10361	−0.10666
a_3/g·cm^{-3}·K^{-2}	−0.00311	0.01262	0.02125	0.02013
a_4/g·cm^{-3}·K^{-3}		−0.01398	−0.02150	−0.01947
s/g·cm^{-3}	0.00003	0.00002	0.00007	0.00003

	$x = 0.5883$	$x = 0.6650$	$x = 0.7101$	$x = 0.7993$	$x = 0.9062$
c_0	0.090128	0.090241	0.093183	0.088321	0.088788
b_0/MPa	100.9348	100.4480	106.1020	104.4432	111.2940
b_1/MPa K^{-1}	−76.0982	−68.3245	−81.0830	−76.6368	−81.1356
b_2/MPa·K^{-2}	18.1894	9.1196	22.9774	13.3895	12.9184
a_1/g·cm^{-3}	0.87157	0.90178	0.91755	0.96072	1.02398
a_2/g·cm^{-3}·K^{-1}	−0.10248	−0.12800	−0.10221	−0.12393	−0.13223
a_3/g·cm^{-3}·K^{-2}		0.05962	−0.02313	0.02528	0.02779
a_4/g·cm^{-3}·K^{-3}		−0.05459	0.01617	−0.02718	−0.03224
s/g·cm^{-3}	0.00006	0.00007	0.00007	0.00003	0.00003

tunately, reliable values for density in the heterogeneous region cannot be obtained with our experimental equipment.

Differentiation of eq 1 with respect to pressure and temperature yields isothermal compressibilities κ_T and isobaric thermal expansivities α_p, respectively. The details of the calculation have been explained previously.[10] The uncertainty in the derived properties is estimated to be ±40 TPa^{-1} for κ_T and ± 0.03 kK^{-1} for α_p. The values of κ_T and α_p for the pure liquids at selected pressures and temperatures are given in Table 4. Comparison with literature data for decane[11] results in an absolute mean deviation of 7 TPa^{-1} for κ_T and 0.009 kK^{-1} for α_p. The deviations with respect to literature[6] for dimethyl carbonate were 11 TPa^{-1} for κ_T and 0.012 kK^{-1} for α_p.

Figure 1 shows excess molar volumes v^E at various temperatures and pressures obtained from the densities of pure liquids and mixtures. The uncertainty in this quantity is estimated to be about ±0.03 cm^3·mol^{-1}. Solid lines correspond to the fitted values against composition at the given [p,T] conditions. A Redlich−Kister polynomial was used to this end

$$Y^E = x(1 - x)\sum_{i=1}^{n} A_i(2x - 1)^{i-1} \tag{4}$$

where x denotes the mol fraction of dimethyl carbonate, A_i values are the fitting parameters, and Y is equal to v in this case. Table 5 shows the final values of the parameters together with the standard deviations.

In all cases (all excess properties at all [T,p] conditions), an almost parabolic dependence against composition is observed with the maximum being located around equimolar composition. As for other strongly polar (nonassociated) + long-chain alkane systems,[13,14] both dipolar order of dimethyl carbonate and the orientational order of the alkane are broken upon mixing, resulting in a positive contribution to v^E. Since, normally, dispersive interactions between unlike molecules are weaker than those between like molecules, it is reasonable that they contribute positively to v^E.[15] It appears that these two positive contributions largely overcome the negative ones arising from packing effects. In respect to pressure and temperature dependencies, it is found that v^E increases with an increase

Table 4. Isothermal Compressibilities κ_T and Isobaric Thermal Expansivities α_p for the Pure Liquids

κ_T/TPa^{-1}

p/MPa	T = 283.15	T = 293.15 K	T = 303.15 K	T = 313.15 K	T = 323.15 K
		Dimethyl Carbonate			
10	741	799	862	932	1009
20	694	745	800	860	925
30	654	698	747	799	856
		Decane			
10	892	948	1009	1073	1142
20	831	880	932	987	1045
30	779	822	867	915	965

α_p/kK^{-1}

p/MPa	T = 293.15 K	T = 303.15 K	T = 313.15 K	T = 323.15 K
		Dimethyl Carbonate		
0.1	1.250	1.271	1.302	1.343
10	1.186	1.201	1.224	1.256
20	1.131	1.140	1.158	1.184
30	1.083	1.089	1.102	1.124
40	1.041	1.044	1.055	1.073
		Decane		
0.1	1.026	1.047	1.068	1.090
10	0.964	0.980	0.996	1.013
20	0.911	0.924	0.937	0.950
30	0.867	0.877	0.887	0.898
40	0.828	0.836	0.845	0.854

in temperature and decreases with a increase in pressure (see Figure 1). These trends are in agreement with those previously found for this sort of system.[8]

Excess isothermal compressibilities κ_T^E and excess isobaric thermal expansivities α_p^E were calculated using the Benson and Kiyohara criterion[16]

$$\varsigma^E = \varsigma - \phi_1\varsigma_1 + (1 - \phi_1)\varsigma_2 \qquad (5)$$

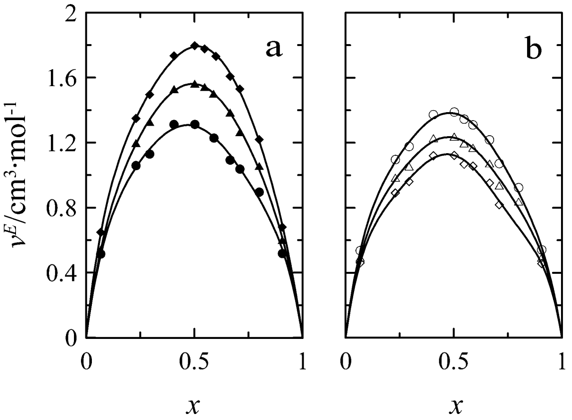

Figure 1. Excess molar volumes v^E for $x$dimethyl carbonate + (1 − x)decane at temperature T and pressure p. (a) p = 0.1 MPa and ●, T = 288.15 K; ▲, T = 308.15 K; ◆, T = 328.15 K. (b) T = 293.15 K and ○, p = 0.1 MPa; △, p = 15 MPa; ◇, p = 30 MPa. The solid line indicates fitted values from eq 4.

where ϕ_1 is the volume fraction of dimethyl carbonate and ζ can be κ_T or α_p. Both were fitted to eq 4, the values of A_i and s being given in Table 5. Experimental data together with the fitted curves are represented in Figures 2 and 3. The uncertainty in these magnitudes was estimated in ±6 TPa^{-1} and ±10 MK^{-1}, respectively.

As usual, the behavior of κ_T^E is very similar to that of v^E; they are always positive, increasing with temperature and decreasing with pressure, in agreement with literature data.[9] The excess isobaric thermal expansivity vs composition plot of Figure 3 reveals that this quantity is positive over the whole composition range, showing a parabolic shape. The pressure and temperature dependencies of α_p^E cannot be obtained with the present experimental device owing to the limited precision in the determination of α_p.

Table 5. Fitting Parameters A_i of Equation 4 and Standard Deviations s at Selected pressures p and Temperatures T for xDimethyl Carbonate + (1−x)Decane

v^E/cm^3·mol^{-1}

	T = 288.15 K	T = 308.15 K	T = 328.15 K	T = 293.15 K	T = 293.15 K	T = 293.15 K
	p = 0.1 MPa	p = 0.1 MPa	p = 0.1 MPa	p = 0.1 MPa	p = 15 MPa	p = 30 MPa
A_1/cm^3·mol^{-1}	5.211	6.245	7.159	5.526	4.927	4.493
A_2/cm^3·mol^{-1}	−0.644	−0.208	0.415	−0.428	−0.500	−0.733
A_3/cm^3·mol^{-1}	−0.020	0.086	0.306	−0.374	−0.478	−1.518
A_4/cm^3·mol^{-1}	−0.119	−0.527	−1.968	−0.610	−0.339	−0.155
A_5/cm^3·mol^{-1}	4.048	3.109	3.386	4.430	4.612	5.821
s/cm^3·mol^{-1}	0.03	0.015	0.015	0.03	0.03	0.02

$\kappa_T{}^E$/TPa^{-1}

	T = 293.15 K	T = 303.15 K	T = 313.15 K	T = 293.15 K	T = 293.15 K
	p = 5 MPa	p = 5 MPa	p = 5 MPa	p = 17.5 MPa	p = 30 MPa
A_1/TPa^{-1}	278	343.3	431.6	210	167
A_2/TPa^{-1}	119	107.4	206.7	86	79
A_3/TPa^{-1}	179	125.7	235.7	133	99
A_4/TPa^{-1}	−114	−38.6	−128.1	−47	−33
A_5/TPa^{-1}	−204	−112.2	−185.4	−156	−133

$\alpha_p{}^E$/10^{-3}kK^{-1}

	T = 308.15 K
	p = 0.1 MPa
A_1/MK^{-1}	271
A_2/MK^{-1}	155
A_3/MK^{-1}	−52
A_4/MK^{-1}	−130
A_5/MK^{-1}	78
s/MK^{-1}	5

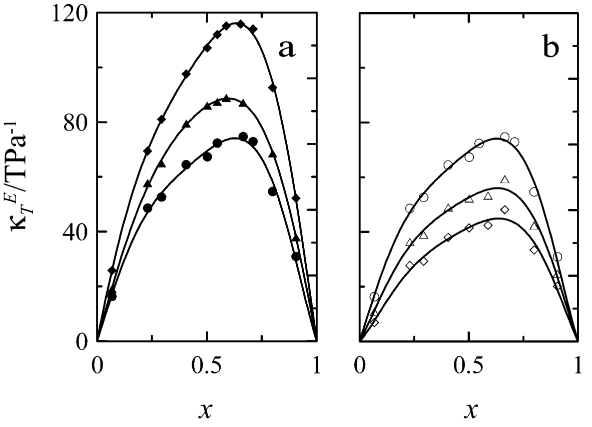

Figure 2. Excess isothermal compressibilities κ_T^E for $x$dimethyl carbonate + (1 − x)decane at temperature T and pressure p. (a) p = 5 MPa and ●, T = 293.15 K; ▲, T = 308.15 K; ◆, T = 323.15 K. (b) T = 293.15 K and ○, p = 5 MPa; △, p = 17.5 MPa; ◇, p = 30 MPa. The solid line indicates fitted values from eq 4.

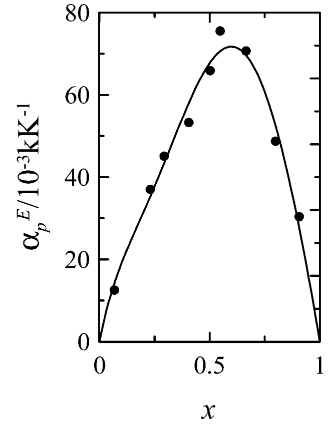

Figure 3. Excess isobaric thermal expansivities α_p^E at 308.15 K and 0.1 MPa for the system $x$dimethyl carbonate + (1 − x)decane. The solid line indicates fitted values from eq 4.

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Received for review November 12, 2003. Accepted April 21, 2004. This work is part of Research Project PGIDT00PXI38305PR. The authors are indebted to the Secretaría Xeral de Investigación e Desenvolvemento (Xunta de Galicia) for financial support.

JE0342320