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Thermodynamic behaviour of the binary systems dimethyl carbonate + *n*-octane or *n*-nonane

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Abstract

Isobaric molar heat capacities, densities and speeds of sound were determined for the dimethyl carbonate + *n*-octane and *n*-nonane systems at atmospheric pressure covering the whole composition interval and in the temperature range (288.15–308.15 K). From these data, derived properties and excess magnitudes were obtained. A strongly temperature-dependent W-shaped curves against composition were observed for excess isobaric molar heat capacities in both system, showing the other excess magnitudes regular tendencies. From these results, a comparative analysis of the existing differences between the thermodynamic behaviour of the two systems was carried out.

Keywords: Dialkyl carbonate; *n*-Alkane; Heat capacity; Excess magnitudes

1. Introduction

The thermodynamic properties of dialkyl carbonate + alkane binary systems have been extensively studied in the last years [1–16]. They present a strong deviation from the ideal mixture, fact reflected in the highly positive values of excess enthalpies and excess volumes [4–11]. Besides, some of these systems, characterised by a high dipolar moment of the dialkyl carbonate and/or long alkyl chain of the alkane, present liquid–liquid equilibria near ambient temperature [12]. It is well known that at experimental conditions close to the critical point of this transition, long-range concentration fluctuations appears [17]. They affect slightly to first-order excess magnitudes, as excess volumes or excess enthalpies, but strongly to some second-order ones, as excess isobaric molar heat capacity. In this context, it has been found for this systems that this quantity shows, as for other polar + alkane systems, a W-shaped composition dependence, being the maximum of the curve highly enhanced as temperature is closer to the critical one [13–16]. Critical phenomena should have also strong influence in the composition dependence of other second-order excess derivatives [17], but, although some works have reported them [13–16], this fact has not been experimentally confirmed yet. In the present paper new isobaric molar heat capacity, density and speed of sound values were reported for the dimethyl carbonate + *n*-octane and *n*-nonane systems. From this data, excess isobaric molar heat capacities, excess molar volumes, excess isobaric thermal expansivities, excess isothermal compressibilities, excess isochoric molar heat capacities and excess isentropic compressibilities were obtained. These results allowed to make an analysis of the thermodynamic behaviour of the two systems in the frame of the above-mentioned general assertions for the dialkyl carbonate + *n*-alkane systems.

2. Experimental

Dimethyl carbonate (purity >99% mol) was supplied from Aldrich, *n*-octane from Fluka (purity >99.5%) and *n*-nonane (purity >99%) from Aldrich. All chemicals were degassed by an ultrasonic bath and dried by passage through molecular sieves prior to use. Mixtures were prepared by weighing

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using a Mettler AE-240 balance. The uncertainty in mole fraction was estimated in ±0.0001.Isobaric heat capacities per unit volume were measured with a Setaram micro DSCII differential scanning calorimeter, using the stepwise method. Vacuum and *n*-heptane from Fluka (purity >99.8%) were used as heat capacity standards, being the values of the latter taken from literature [18]. Density ρ and speed of sound u were measured using a densimeter and sound analyser DSA48. Density calibration was done using Milli-Q water and *n*-heptane and for speed of sound only the former was needed. The values of these calibration standards were taken from literature [19–22]. Isobaric molar heat capacities C_p were obtained from heat capacity and density data. The estimated uncertainty in C_p is 0.15 J mol^{-1} K^{-1}, in ρ, 0.0001 g cm^{-3} and in u, 0.1 m s^{-1}. A detailed description of the experimental procedure can be found elsewhere [23,24].

3. Results and discussion

Table 1 shows the data for isobaric molar heat capacity C_p, density ρ and speed of sound u of the pure compounds compared with literature. In Tables 2–4 the values for the mixtures are presented. From ρ and u, isentropic compressibility κ_S can be determined from Laplace's equation $\kappa_S = (\rho u^2)^{-1}$. In order to calculate isobaric thermal expansivities α_p from direct analytical derivation, density data were fitted to the next expression:

$$\rho = \sum_{i=1}^{5}\sum_{j=1}^{3} A_{ij} 10^{1-j} x^{i-1} (T - T_0)^{j-1} \qquad (1)$$

where $T_0 = 283.15$ K. Coefficients A_{ij} are listed in Table 5. From all these magnitudes, isochoric molar heat capacities C_v and isothermal compressibilities κ_T were calculated using these well-known thermodynamic relations:

$$C_v = C_p \frac{\kappa_S}{\kappa_T} \qquad (2)$$

Table 2
Density of mixtures at several temperatures

x	ρ (g cm^{-3})			
	$T = 288.15$ K	$T = 293.15$ K	$T = 298.15$ K	$T = 308.15$ K
x(dimethyl carbonate) + (1 − x)(*n*-octane)				
0.05047	0.71514	0.71088	0.70669	0.69846
0.10722	0.72536	0.72113	0.71689	0.70837
0.19692	0.74356	0.73910	0.73468	0.72585
0.29575	0.76642	0.76179	0.75715	0.74786
0.40177	0.79460	0.78970	0.78490	0.77510
0.50113	0.82500	0.81990	0.81480	0.80450
0.59737	0.85880	0.85350	0.84810	0.83730
0.69901	0.90018	0.89447	0.88877	0.87752
0.79823	0.94783	0.94192	0.93598	0.92407
0.89916	1.00583	0.99955	0.99330	0.98086
0.94899	1.03895	1.03256	1.02612	1.01324
x(dimethyl carbonate) + (1 − x)(*n*-nonane)				
0.05335	0.72939	0.72545	0.72148	0.71345
0.10090	0.73694	0.73286	0.72879	0.72068
0.19983	0.75447	0.75026	0.74603	0.73748
0.29629	0.77422	0.76982	0.76540	0.75646
0.39646	0.79813	0.79341	0.78873	0.77936
0.49777	0.82630	0.82139	0.81645	0.80649
0.59827	0.85919	0.85392	0.84867	0.83824
0.69763	0.89772	0.89223	0.88673	0.87562
0.79818	0.94458	0.93872	0.93290	0.92126
0.89850	1.00230	0.99620	0.99010	0.97770

$$\kappa_T = \kappa_S + \frac{T v \alpha_p^2}{C_p} \qquad (3)$$

where v is the molar volume of the sample. Excess magnitudes for all quantities were determined using the Benson and Kiyohara's criterion of ideality [28], defined by the next equations:

$$C_p^{id} = x C_{p1} + (1 - x) C_{p2} \qquad (4)$$

$$v^{id} = x v_1 + (1 - x) v_2 \qquad (5)$$

$$\alpha_p^{id} = \phi \alpha_{p1} + (1 - \phi) \alpha_{p2} \qquad (6)$$

$$\kappa_T^{id} = \phi \kappa_{T1} + (1 - \phi) \kappa_{T2} \qquad (7)$$

Table 1
Properties of pure liquids at the temperature T

Liquid	T (K)	C_p (J mol^{-1} K^{-1})		ρ (g cm^{-3})		u (m s^{-1})	
		This work	Literature [18]	This work	Literature	This work	Literature
Dimethyl carbonate	288.15	164.03		1.07656	1.07646 [25]		
	293.15			1.07002	1.0700 [26]		
	298.15	165.02		1.06342	1.06338 [25]	1197.06	1196 [26]
	308.15	166.3		1.05011	1.05004 [25]		
n-Octane	288.15	250.12	250.86	0.70654	0.70670 [20]		
	293.15			0.70253	0.70268 [20]		
	298.15	253.94	255.12	0.69850	0.69864 [20]	1172.09	1172 [19]
	308.15	258.03	259.50	0.69034	0.69052 [20]		
n-Nonane	288.15	280.13	280.14	0.72166	0.72162 [20]		
	293.15			0.71775	0.71775 [20]		
	298.15	284.23	284.43	0.71385	0.71387 [20]	1206.35	1206 [27]
	308.15	288.8	288.67	0.70605	0.70607 [20]		

Table 3
Isobaric molar heat capacity of mixtures at several temperatures

x	C_p (J mol^{-1} K^{-1})		
	T = 288.15 K	T = 298.15 K	T = 308.15 K
x(dimethyl carbonate) + (1 − x)(n-octane)			
0.05306	245.12	248.71	252.62
0.10162	240.86	244.21	248.11
0.14956		240.18	243.91
0.20120	232.99	235.89	239.35
0.29994	225.81	228.26	231.22
0.40121	219.02	220.47	222.77
0.45492	215.27	216.32	218.33
0.50368	211.61	212.23	213.92
0.55610	207.58	207.88	209.21
0.59980		200.61	205.31
0.64825	199.24	199.37	200.70
0.70753	192.93		
0.79818	182.94	183.94	185.39
0.84841	177.49	178.74	180.34
0.89685			175.52
0.94934	167.98	169.06	170.50
x(dimethyl carbonate) + (1 − x)(n-nonane)			
0.05262	273.59	277.51	281.91
0.10119	267.97	271.68	275.81
0.15139	262.27	265.73	269.66
0.20012	256.97	260.11	263.87
0.29702	247.10	249.53	252.77
0.40176	237.24	238.51	241.06
0.45197	232.75	233.42	235.55
0.50142	228.40	228.22	230.02
0.55135	223.85	222.78	224.27
0.60237	218.72	217.10	218.19
0.64770	213.45	211.64	212.66
0.69785	206.69	205.13	206.01
0.79444	191.68	191.78	192.97
0.84499	183.74	184.48	185.93
0.90159	175.65	176.75	178.33
0.94857	169.80	170.87	172.40

Table 4
Speed of sound of mixtures at 298.15 K

x(dimethyl carbonate) + (1 − x) (n-octane)		x(dimethyl carbonate) + (1 − x) (n-nonane)	
x	u (m s^{-1})	x	u (m s^{-1})
0.05047	1164.98	0.05335	1198.49
0.10722	1158.10	0.10090	1191.76
0.19692	1148.90	0.19983	1180.00
0.29575	1141.50	0.29629	1170.49
0.40177	1136.06	0.39646	1162.42
0.50113	1133.74	0.49777	1156.83
0.59737	1134.25	0.59827	1153.43
0.69901	1138.73	0.69763	1153.52
0.79823	1149.09	0.79818	1158.15
0.89916	1167.73	0.89850	1171.20
0.94899	1180.93		

where superscript id denotes the ideal magnitude, subscript 1 and 2 denotes properties of pure compounds and ϕ is the volume fraction, defined by:

$$\phi = \frac{xv_1}{v^{\mathrm{id}}} \tag{8}$$

C_v^{id} and κ_v^{id} are obtained from the ideal magnitudes defined by Eqs. (4)–(7) using the thermodynamic relations given by Eqs. (2) and (3). The excess values for a magnitude X is defined, as usual, by:

$$X^{\mathrm{E}} = X - X^{\mathrm{id}} \tag{9}$$

v^{E} were fitted to the next Redlich–Kister polynomial:

$$v^{\mathrm{E}} = x(1-x)\sum_{i=1}^{5}\sum_{j=1}^{3} A_{ij}10^{1-j}x^{i-1}(T-T_0)^{j-1} \tag{10}$$

Table 5
Parameters A_{ij} for the magnitudes fitted against composition and temperature (Eqs. (1) and (10))

j	i				
	1	2	3	4	5
x(dimethyl carbonate) + (1 − x)(n-octane)					
ρ(g cm^{-3}) (s = 0.00008)					
1	1.40721	−0.32406	−0.17999	0.06192	−0.04176
2	0.01598	0.00426	−0.02969	0.04302	−0.0226
3	0.00022	−0.00162	0.00897	−0.01493	0.00759
v^{E}(cm^3 mol^{-1}) (s = 0.007)					
1	4.593	−0.934	1.527	0.435	−1.248
2	0.449	0.210	0.053	−0.920	1.166
3	−0.012	−0.053	−0.037	0.244	−0.349
x(dimethyl carbonate) + (1 − x)(n-nonane)					
ρ(g cm^{-3}) (s = 0.0001)					
1	1.37817	−0.262	−0.20528	0.08605	−0.07354
2	0.01484	−0.00369	0.01509	−0.02446	0.00921
3	0.00017	0.00088	−0.0033	0.00407	−0.00159
v^{E} (cm^3 mol^{-1}) (s = 0.007)					
1	5.018	−0.824	1.220	0.541	0.384
2	0.436	0.308	−0.232	−0.241	−0.203
3	0.006	−0.056	0.035	0.012	0.057

Table 6
Parameters A_i for the magnitudes fitted only against composition (Eq. (11))

Magnitude	T (K)	A_1	A_2	A_3	A_4	A_5	A_6	A_7	s
x(dimethyl carbonate) + (1 − x)(n-octane)									
C_p^E (J mol^{-1} K^{-1})	288.15	19.6	22.11	−39.94	−54.26	−6.33	33.8	15.41	0.07
	298.15	12.62	11.34	−28.04	−14.55				0.07
	308.15	8.76	7.24	−21.01	−8.24				0.09
α_p^E (M K^{-1})	298.15	269.4	94.9						0.3
κ_T^E (T Pa^{-1})	298.15	286.1	−12.4	35.8					0.5
C_v^E(J mol^{-1} K^{-1})	298.15	1.33	6.71	−26.04	−14.27				0.02
κ_S^E(T Pa^{-1})	298.15	173.46	−26.08	25.75	−32.74	−36.11			0.14
x(dimethyl carbonate) + (1 − x)(n-nonane)									
C_p^E J mol^{-1} K^{-1})	288.15	25.97	55.42	−13.79	−135.73	−105.44	85.92	92.26	0.07
	298.15	14.95	27.91	−17.48	−53.41	−29.85	24.5	21.52	0.06
	308.15	10.54	19.27	−20.02	−33.7	−2.44	15.88		0.05
α_p^E (K^{-1})	298.15	290.0	150.3	−67.3					0.6
κ_T^E (T Pa^{-1})	298.15	313.7	47.7	23.1	32.2				0.3
C_v^E (J mol^{-1} K^{-1})	298.15	2.49	18.14	−11.97	−31.84	−11.75			0.09
κ_S^E (T Pa^{-1})	298.15	200.67	14.14	26.77					0.2

where $T_0 = 283.15$ K. Coefficients A_{ij} and standards deviations are listed in Table 5. The other excess magnitudes, since there are not enough data to perform a simultaneous fit against composition and temperature, were fitted at each temperature to the next expression:

$$X^E = x(1-x)\sum_{i=0}^{n} A_i(2x-1)^i \tag{11}$$

The A_i coefficients for the studied magnitudes, as well as standard deviations of the fits, are listed in Table 6. There are literature values for the studied mixtures to perform a comparison with the data of this work. Tojo et al. [8] have measured v^E for both systems. Their values present a mean absolute deviation from those of this work of 0.03 cm^3 mol^{-1} for the n-octane system and of 0.018 cm^3 mol^{-1} for the n-nonane one. Also, Lugo et al. [7] have measured v^E and α_p for the dimethyl carbonate + n-octane system. The comparison with these values yields a mean absolute deviation of 0.02 cm^3 mol^{-1} for v^E and 0.014 kK^{-1} for α_p.

Fig. 1 presents excess isobaric molar heat capacities C_p^E for the two studied systems. A clear W-shaped composition dependence can be observed, with the maximum located near equimolar composition. Besides, it is strongly enhanced as temperature is decreased, being this effect more pronounced for the n-nonane system. As has been said in the introductory section, polar + n-alkane systems often present this surprising W-shaped composition dependence. Since C_p^E is a measure of the degree of non-randomness in the mixture, these curves have been explained as a manifestation of a change of the non-randomness character of the mixtures as composition is varied [29]. At extreme values of mole fraction, the order of the dilute component is fully destroyed, due to the interactions between its molecules are hindered by the molecules of the other component, reverting in a negative value of C_p^E. At intermediate composition there are not enough molecules of the solvent to avoid interactions between the molecules of the diluted component, thus they can take place, resulting in non-randomness in the mixture and then, in a positive contribution to C_p^E. This situation is more pronounced as temperature is lowered, provoking the phase separation at temperatures enough low. For values near critical composition these increment in C_p^E is highly enhanced as temperature is closer to the critical one, tending to infinity at the critical point, fact explained in the context of critical phenomena, since close to critical point, long range composition fluctuations appears, reverting in a very strong character of non-randomness of the mixture, and then in a very high value of C_p^E [30]. Using previous data of liquid–liquid equilibria for dimethyl carbonate + n-alkane systems [12] and extrapolating them for the studied systems, values for critical temperatures can be estimated, being for n-octane system $T_c = 277$ K, and for n-nonane one $T_c = 282$ K. These values explain their different behaviour, since at the lower working temperature, dimethyl

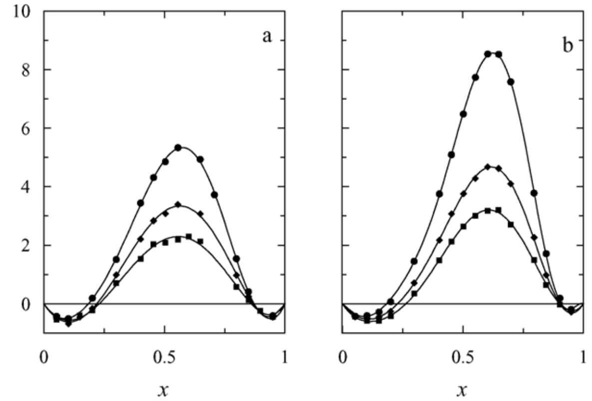

Fig. 1. Excess isobaric molar heat capacities C_p^E for (a) x(dimethyl carbonate) + (1 − x)(n-octane) and (b) x(dimethyl carbonate) + (1 − x)(n-nonane) at the temperatures: (●) 288.15 K, (◆) 298.15 K and (■) 308.15 K. (—) fitted values from Eq. (11).

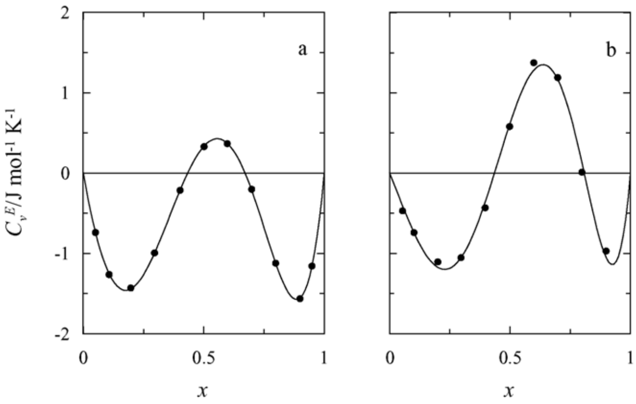

Fig. 2. Excess isochoric molar heat capacities C_v^{E} for (a) x(dimethyl carbonate) + (1 − x)(n-octane) and (b) x(dimethyl carbonate) + (1 − x)(n-nonane) at 298.15 K. (—) fitted values from Eq. (11).

carbonate + n-nonane system are closer to the critical point. Fig. 2 presents C_v^{E}. The W-shaped curves are a consequence of the strong influence of C_p in C_v, as can be deduced from Eqs. (2) and (3).

Figs. 3–6 show the experimental volumetric excess magnitudes (v^{E}, α_p^{E}, $\kappa_{\mathrm{T}}^{\mathrm{E}}$ and $\kappa_{\mathrm{s}}^{\mathrm{E}}$) for both systems. They are all strongly positive, with a composition dependence almost parabolic, and higher for n-nonane system. Besides, excess molar volumes increase as temperature is raised. Excess volumetric properties for polar + n-alkane systems are usually positives, fact due to the rupture of both dipolar interactions of the polar compound and dispersive interactions of the n-alkane during the mixing process [31,32]. Negative contributions to these magnitudes are related with dispersive interactions between unlike molecules, and due to the dissimilarity on the chemical nature of the two compounds, they often are small. This implies that as the polar character of the polar component and the length of the n-alkane are higher, the difference between the interactions strength of the like and unlike molecules is more marked, and then, volumetric excess magnitudes are higher. The temperature dependence of these properties is usually positive since as temperature is increased the structure of both liquids is easily broken by the mixing process. Taking into account all these considerations, the higher values obtained for the n-nonane

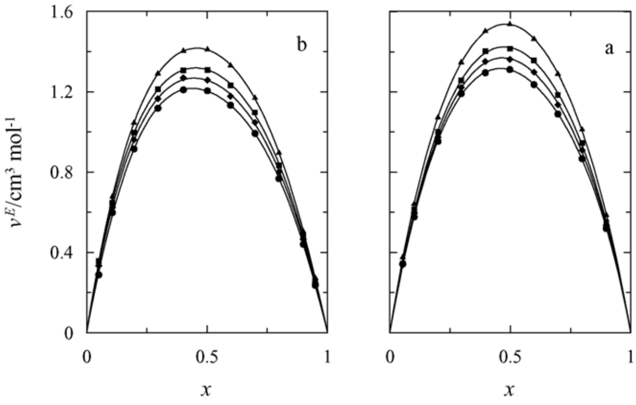

Fig. 3. Excess molar volumes v^{E} for (a) x(dimethyl carbonate) + (1 − x)(n-octane) and (b) x(dimethyl carbonate) + (1 − x)(n-nonane) at the temperatures: (●) 288.15 K, (⧫) 293.15 K, (■) 298.15 K and (▲) 308.15 K. (—) fitted values from Eq. (10).

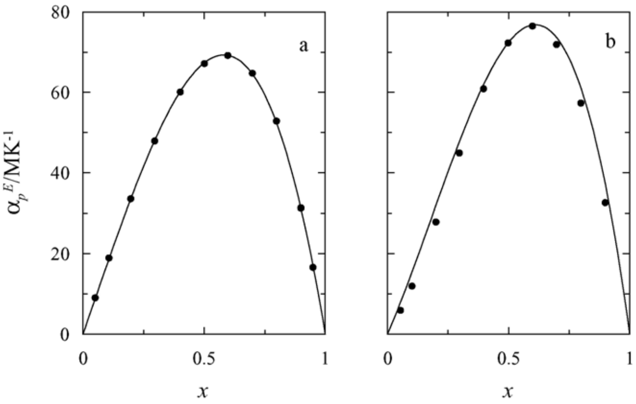

Fig. 4. Excess isobaric thermal expansivities α_p^{E} for (a) x(dimethyl carbonate) + (1 − x)(n-octane) and (b) x(dimethyl carbonate) + (1 − x)(n-nonane) at 298.15 K. (—) fitted values from Eq. (11).

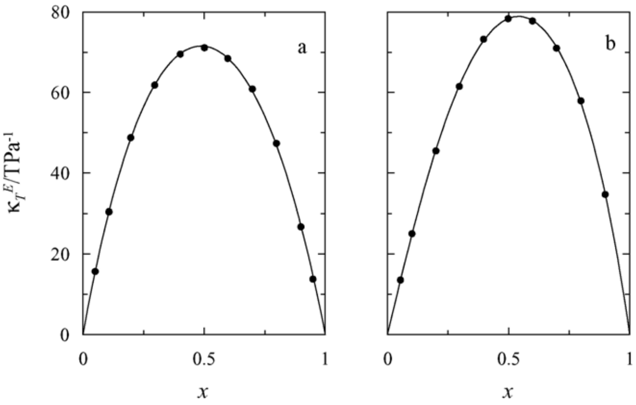

Fig. 5. Excess isothermal compressibilities $\kappa_{\mathrm{T}}^{\mathrm{E}}$ for (a) x(dimethyl carbonate) + (1 − x)(n-octane) and (b) x(dimethyl carbonate) + (1 − x)(n-nonane) at 298.15 K. (—) fitted values from Eq. (11).

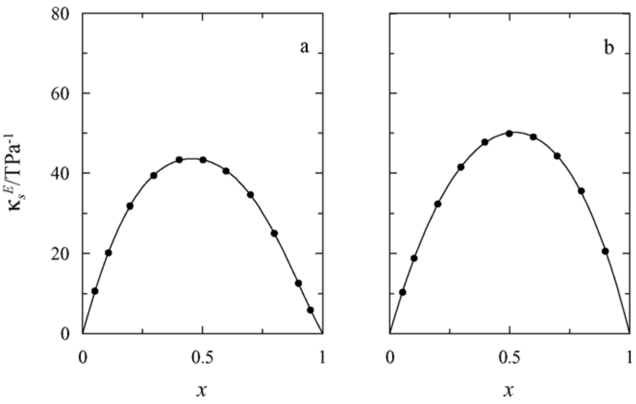

Fig. 6. Excess isentropic compressibilities $\kappa_{\mathrm{s}}^{\mathrm{E}}$ for (a) x(dimethyl carbonate) + (1 − x)(n-octane) and (b) x(dimethyl carbonate) + (1 − x)(n-nonane) at 298.15 K. (—) fitted values from Eq. (11).

system can be explained by the fact that the dispersive interactions between *n*-nonane molecules are higher than those of *n*-octane due to its longer alkyl chain, which implies that the mixing process break a more structured liquid and provokes a higher contribution to these excess magnitudes.

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