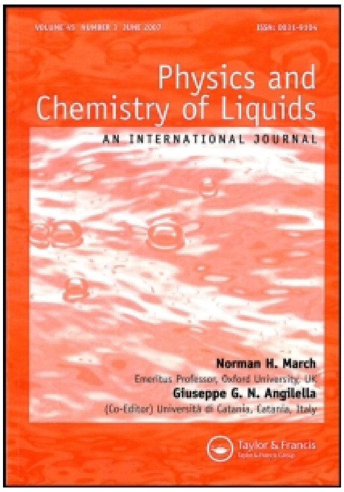

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Experimental results of and for binary mixtures (propylbenzene + an alkane or + an alkyl ethanoate)

J. Ortega[a]; G. Bolat[b]; E. Marrero[a]; I. M. Popa[b]
[a] Laboratorio de Termodinámica y Fisicoquímica de Fluidos, Parque Científico Tecnológico, 35071-Universidad de Las Palmas de Gran Canaria, Canary Islands, Spain [b] Faculty of Chemical Engineering, Department of Chemical Engineering, Technical University "Gh. ASACHI", Iasi 700050, Romania

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Experimental results of H_m^E and V_m^E for binary mixtures (propylbenzene + an alkane or + an alkyl ethanoate)

J. ORTEGA*†, G. BOLAT‡, E. MARRERO† and I. M. POPA‡

†Laboratorio de Termodinámica y Fisicoquímica de Fluidos, Parque Científico Tecnológico, 35071-Universidad de Las Palmas de Gran Canaria, Canary Islands, Spain
‡Faculty of Chemical Engineering, Department of Chemical Engineering, Technical University "Gh. ASACHI", Iasi 700050, Romania

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This article presents an experimental and theoretical contribution on binary mixtures alkylbenzene + alkanes or + alkylethanoates as a continuation of an investigation project on liquid mixtures containing aromatic compounds and a second component. So, excess molar enthalpies H_m^E at $T = 298.15$ K and excess molar volumes V_m^E at $T = 298.15$ and 318.15 K for a set of 11 binary mixtures formed by propylbenzene and alkanes (C_7 to C_{16}) and alkyl ethanoates (methyl to butyl) are presented. All systems studied showed endothermic effects increasing regularly the H_m^E and V_m^E in mixtures with alkanes as the aliphatic hydrocarbon chain-length increases, however, in mixtures with ethanoates both excess properties decrease with the increase of the ester molecular weight. The V_m^Es result positive in mixtures with all alkanes except with heptane and also with all alkylethanotes except with butyl ethanoate which present a sigmoidal representation. An increase in temperature produces $(\partial V_m^E/\partial T) < 0$ in mixtures containing heptane and hexadecane and a almost null slope in the remain mixtures. However, the variation in mixtures with alkyl ethanoates is positive for methyl ethanaote changing to negative from ethyl. Experimental values for all systems are correlated with a suitable parametric equation which is function of the so-called active fraction. Finally, two known versions of the UNIFAC group contribution model are applied to our mixtures, giving acceptable results for all mixtures studied when one of the versions is used.

Keywords: Excess enthalpy; Excess volumes; Alkane; Propylbenzene; Ethanoates; UNIFAC

1. Introduction

This article is the second of a series in which the behavior of binary mixtures containing aromatic hydrocarbons with alkanes and alkyl ethanoates is studied. In a previous paper [1] the experimental data of H_m^E and V_m^E for a set of nine mixtures of {butyl benzene + an alkane (C_7 to C_{12}), + an alkyl ethanoate (methyl to butyl)} were presented. Results were justified through an analysis of the interactions ocurring for this kind of mixtures and a model was proposed to explain their behavior. UNIFAC group

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*Corresponding author. Tel.: +34 928 459 548. Fax: +34 928 459 550. Email: jortega@dip.ulpgc.es

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contribution model in two of the versions [2,3] was also applied and the comparison with experimentals are shown in figure 1(a) and (b). The version by Gmehling *et al.* [3] gave values smaller than experimental; however, the version by Dang and Tassios [2], using new interaction parameters obtained previously [1] gave place to acceptable predictions. The utility of these new parameters will be checked here for the H_m^E values of the mixtures containing propylbenzene elected for this work.

In order to carry out a systematic study on the systems indicated above in this work the mixing properties, H_m^E at 298.15 K and V_m^E at the temperatures of 298.15 and 318.15 K, for a set of 11 binary mixtures formed by {propylbenzene + an alkane (C_nH_{2n+2}, $n = 7$–10, 12, 14, 16), or + an alkyl ethanoate ($H_3CCOOC_vH_{2v+1}$, $v = 1$–4)} have been measured.

According to the results obtained in a previous paper [1] for butylbenzene we have also considered convenient present here the measurements of the above thermodynamic quantities in mixtures of that aromatic compound with tetradecane and hexadecane because we have checked that the estimations of the theoretical models are less precise when the hydrocarbon chain length increases, such as is shown in figure 1(a) and (b). For mixtures containing propylbenzene literature shows some data of H_m^E at several temperatures for propylbenzene $+C_7$ [4] or $+C_8$ [5,6] and of V_m^E for (propylbenzene + C_7, or $+C_8$, $+C_9$, $+C_{14}$) [7,8] which will be used for comparison. On the other hand, literature [9] contains V_m^E values measured at 303.15 K for (propylbenzene + ethyl, or +butyl ethanoate).

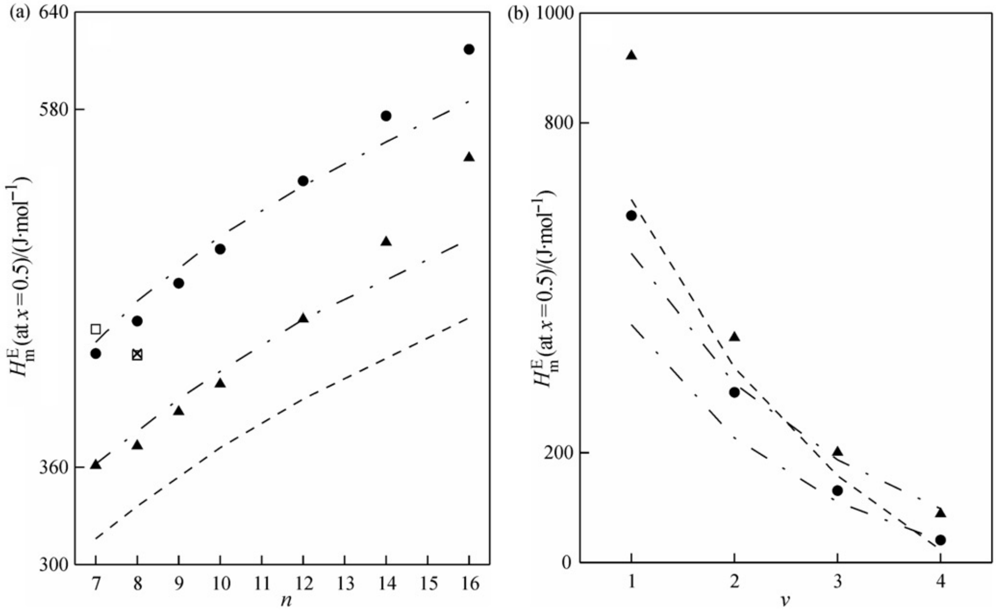

Figure 1. Comparison of equimolar (●) H_m^E values for binaries {$C_6H_5C_3H_7 + C_nH_{2n+2}$, in (a) or $+CH_3COOC_vH_{2v+1}$ in (b)} with the corresponding H_m^E values (▲) of the binaries {$C_6H_5C_4H_9 + C_nH_{2n+2}$, in (a) or $+CH_3COOC_vH_{2v+1}$ in (b)}. Values from literature, (☐) at 298.15 K [4,5], and (✕) at 308.15 K [6]. Straight lines correspond to values by application of UNIFAC method, (— · — ·) from [2], and (– – –) from [3].

Lastly, as mentioned earlier, another objective of this series of papers with aromatic compounds is to verify the predictive capacity of two of the more known versions of UNIFAC method, and to improve the application proposing better values for parameters corresponding to alkyl/aromatic and carboxylate/aromatic molecular interactions.

2. Experimental

All substances used were of the highest commercial purity between those manufactured by Fluka and Aldrich. Before use all them were degasified with ultrasound and then stored in the dark on a molecular sieve (Fluka, 0.3 nm) to remove the possible moisture. The quality of substances was checked measuring some physical properties, such as the density ρ and the refractive index n_D, at two temperatures of 298.15 and 318.15 K. The quality of the substances was verified with a GC model HP-6890, equipped with flame ionization detector (FID), and the resulting purity values for all the substances coincided with those indicated by the manufacturer. Experimental measurements obtained for propylbenzene, tetradecane, and hexadecane are shown in table 1 together with the values reported in literature, at the same conditions, and all of them presented a good agreement with those from literature [10–12]. Measurements for other hydrocarbons were almost coincident with those published previously [1] and therefore they have not been shown here.

Densities at the temperatures mentioned for pure compounds and mixtures were measured with a digital vibrating-tube densimeter Anton Paar, model DMA-58, with a reading error of $\pm 0.02\,\mathrm{kg\,m^{-3}}$. Apparatus was previously calibrated using double-distilled water, with an electrical conductivity smaller than 2 μS, and nonane (Fluka) according to the standard procedure used in our laboratory for years. The precision in the temperature control by a Peltier effect was of ±0.01 K. From density measurements and known the composition, x, of the samples prepared with an

Table 1. Physical Properties of pure substances measured at $T = 298.15$ and 318.15 K and atmospheric pressure, where ρ and n_D denote the density and refractive index, respectively.

		ρ (kg m^{-3})		n_D	
Compound	T (K)	Exp.	Lit.	Exp.	Lit.
Propylbenzene	298.15	857.55	857.51[a]	1.4896	1.4889[a]
			857.29[b]		1.4895[b]
	318.15	840.62	840.35[d]	1.4810	1.4795[d]
Tetradecane	298.15	759.13	760.03[a]	1.4270	1.4263[a]
			759.17[b]		1.4269[b]
	318.15		759.36[c]		1.4271[c]
		744.97	745.30[b]	1.4189	
Hexadecane	298.15	770.09	769.96[b]	1.4321	1.4325[b,c]
			770.10[c]		
	318.15	756.21	756.30[b]	1.4242	

[a] Ref. [10].
[b] Ref. [11].
[c] Ref. [12].
[d] Values interpolated from [11].

uncertainty better than $\pm 5 \cdot 10^{-5}$, the V_m^Es were indirectly calculated with an uncertainty of $\pm 2 \cdot 10^{-9}\,m^3\,mol^{-1}$.

Excess molar enthalpies H_m^E, were measured directly in a microcalorimeter Calvet, model MS80D, from Setaram, but only at $T = 298.15$ K. The apparatus was regularly calibrated with a Joule effect and using different values of electrical power. The uncertainty estimated for calorimetric measurements was of $\pm 0.2\,J\,mol^{-1}$, comparing the experimental results obtained with those from standard mixture (cyclohexane + benzene) [13] and of $\pm 2 \cdot 10^{-4}$ for mole fractions.

3. Results and discussion

Figure 2(a) and (b) present, respectively, the experimental H_m^E values obtained at a temperature of 298.15 K with the corresponding correlation curves for each of binary mixtures {$x$propyl benzene + $(1-x)$ alkanes (C_7 to C_{16})} and for binaries {$x$propyl benzene + $(1-x)$ alkyl (methyl to butyl) ethanoates}. Measurements for binaries {$x$butyl benzene + $(1-x)$alkanes (C_{14} to C_{16})} were also made but not included here to avoid confusion, however, the corresponding equimolar values are plotted in figure 1(a). In all cases the pairs (x, H_m^E) were correlated with a parametric equation whose form, for a excess generic function, is

$$Y_m^E = z(1-z)\sum_{i=0} a_i z^i, \tag{1}$$

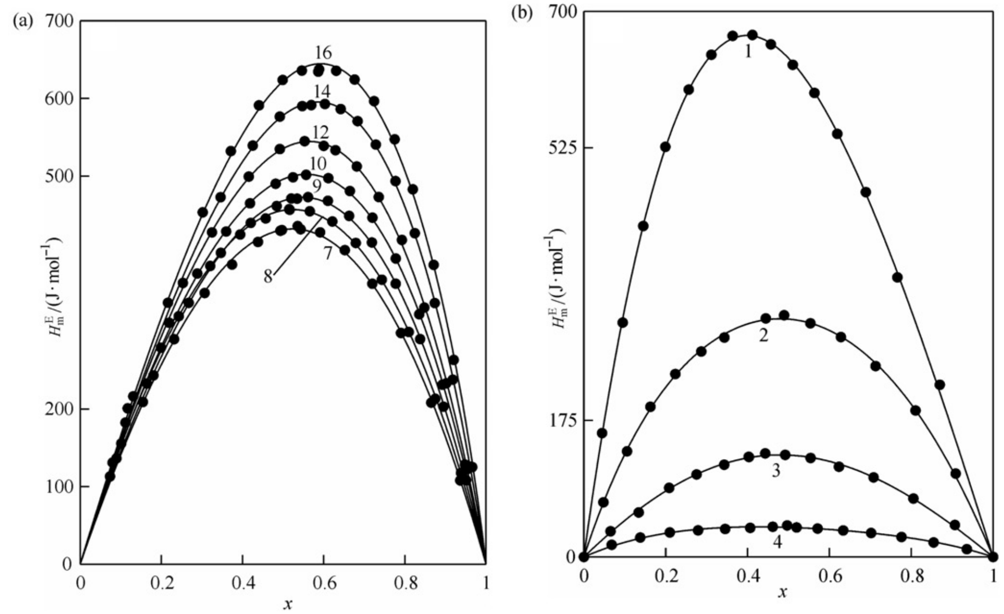

Figure 2. Experimental H_m^E values at 298.15 K and correlation curves for binaries: (a), {$xC_6H_5C_3H_7 + (1-x)\ C_nH_{2n+2}$} and (b), {$xC_6H_5C_3H_7 + (1-x)\ CH_3CO_2C_vH_{2v+1}$} at 298.15 K. Corresponding labels indicate, respectively, the values of n and v.

where:

$$z = \frac{x}{x + k(1 - x)} \tag{2}$$

where Y_m^E would be either H_m^E ($\mathrm{J\,mol^{-1}}$) or V_m^E ($\mathrm{m^3\,mol^{-1}}$). The optimum coefficients found for the set of mixtures using a least-squares method are shown in table 2.

Table 2. Coefficients a_i and k, and standard deviation SD, obtained in the correlations of experimental values using equation (1).

Mixture	k_h	a_0	a_1	a_2	$\mathrm{SD}(H_\mathrm{m}^\mathrm{E})\ \mathrm{J\,mol^{-1}}$
$Y_\mathrm{m}^\mathrm{E} = H_\mathrm{m}^\mathrm{E}(\mathrm{J\,mol^{-1}})$					
$T = 298.15\,\mathrm{K}$					
$x\mathrm{C_6H_5C_3H_7}+$					
$(1-x)\ \mathrm{C_7H_{16}}$	1.138	1728.6	112.9	−232.8	3.8
$(1-x)\ \mathrm{C_8H_{18}}$	1.260	2096.9	−631.8	153.4	2.9
$(1-x)\ \mathrm{C_9H_{20}}$	1.386	2175.0	−596.7	96.6	2.9
$(1-x)\ \mathrm{C_{10}H_{22}}$	1.519	2463.7	−1158.4	428.9	2.1
$(1-x)\ \mathrm{C_{12}H_{26}}$	1.760	2980.4	−1914.5	593.8	4.1
$(1-x)\ \mathrm{C_{14}H_{30}}$	2.009	3345.5	−2537.3	932.3	4.3
$(1-x)\ \mathrm{C_{16}H_{34}}$	2.463	4171.8	−4725.0	2298.0	5.9
$x\mathrm{C_6H_5C_3H_7}+$					
$(1-x)\ \mathrm{CH_3CO_2CH_3}$	0.665	2527.4	617.1	−639.3	4.0
$(1-x)\ \mathrm{CH_3CO_2C_2H_5}$	0.802	1290.9	−579.2	862.3	3.0
$(1-x)\ \mathrm{CH_3CO_2C_3H_7}$	0.931	469.8	223.2	−232.7	2.1
$(1-x)\ \mathrm{CH_3CO_2C_4H_9}$	1.060	245.2	−298.9	218.8	1.2
	k_v	a_0	a_1	a_2	$10^9\mathrm{SD}(V_\mathrm{m}^\mathrm{E})\ \mathrm{m^3\,mol^{-1}}$
$Y_\mathrm{m}^\mathrm{E} = 10_\mathrm{m}^9\mathrm{E}(\mathrm{m^3\,mol^{-1}})$					
$T = 298.15\,\mathrm{K}$					
$x\mathrm{C_6H_5C_3H_7}+$					
$(1-x)\ \mathrm{C_7H_{16}}$	1.052	−202	−245	154	2
$(1-x)\ \mathrm{C_8H_{18}}$	1.167	369	−27	−151	2
$(1-x)\ \mathrm{C_9H_{20}}$	1.282	801	124	−339	5
$(1-x)\ \mathrm{C_{10}H_{22}}$	1.398	1266	−381	−39	4
$(1-x)\ \mathrm{C_{12}H_{26}}$	1.630	1970	−1001	265	4
$(1-x)\ \mathrm{C_{14}H_{30}}$	1.865	2783	−2792	1622	10
$(1-x)\ \mathrm{C_{16}H_{34}}$	2.098	3052	−2714	1221	6
$x\mathrm{C_6H_5C_3H_7}+$					
$(1-x)\ \mathrm{CH_3CO_2CH_3}$	0.570	1279	333	407	3
$(1-x)\ \mathrm{CH_3CO_2C_2H_5}$	0.703	430	435	−482	2
$(1-x)\ \mathrm{CH_3CO_2C_3H_7}$	0.826	159	−80	−30	0
$(1-x)\ \mathrm{CH_3CO_2C_4H_9}$	0.946	81	−211	63	0
$T = 318.15\,\mathrm{K}$					
$x\mathrm{C_6H_5C_3H_7}+$					
$(1-x)\ \mathrm{C_7H_{16}}$	1.059	−380	−465	405	2
$(1-x)\ \mathrm{C_8H_{18}}$	1.171	336	−240	175	1
$(1-x)\ \mathrm{C_9H_{20}}$	1.285	924	−517	463	3
$(1-x)\ \mathrm{C_{10}H_{22}}$	1.400	1351	−716	328	3
$(1-x)\ \mathrm{C_{12}H_{26}}$	1.630	2010	−1286	584	7
$(1-x)\ \mathrm{C_{14}H_{30}}$	1.863	2296	−1168	219	8
$(1-x)\ \mathrm{C_{16}H_{34}}$	2.094	2816	−2237	772	9
$x\mathrm{C_6H_5C_3H_7}+$					
$(1-x)\ \mathrm{CH_3CO_2CH_3}$	0.576	1339	303	469	5
$(1-x)\ \mathrm{CH_3CO_2C_2H_5}$	0.709	528	9	−28	3
$(1-x)\ \mathrm{CH_3CO_2C_3H_7}$	0.831	48	260	−418	1
$(1-x)\ \mathrm{CH_3CO_2C_4H_9}$	0.969	−16	−19	−94	0

Specifically, the k parameter in equation (2) was taken as a fix value, identified as k_h and calculated such as was indicated in a previous paper [1] by the following expression

$$k_h = \left(\frac{q_2}{q_1}\right)\left(\frac{V^o_{m,2}}{V^o_{m,1}}\right)^{2/3}\left(\frac{r_1}{r_2}\right)^{2/3} = \left(\frac{q_2}{q_1}\right)\left(\frac{k_v r_1}{r_2}\right)^{2/3} \tag{3}$$

where q_i and r_i can be obtained, respectively, by $q_i = \sum \upsilon_k^{(i)} Q_k$ and $r_i = \sum \upsilon_k^{(i)} R_k$, produced by a group contribution method of area parameters and volume obtained by weighting the values corresponding to each group Q_k and R_k, of van der Waals, see Bondi [14]. The integer $\upsilon_k^{(i)}$ is the whole number corresponding to the number of the k-type groups in a molecule of the i-component. For our mixtures literature only presents H^E_m values for (propylbenzene + heptane) at 298.15 K by Kehiaian *et al.* [4] and they are slightly higher than ours such as is shown in figure 1(a) using equimolar values. On the contrary the corresponding value obtained at the same temperature by Paul *et al.* [5] for (propylbenzene + octane) is lower and almost identical to that measured at 308.15 K [6]. In this last case the discrepancies found could be due to the different sensitivity of the two calorimeters used.

On the other hand, excess volumes obtained at the two temperatures of 298.15 and 318.15 K, are graphically shown in figure 3(a) and (b), respectively, for binaries of $\{x$propyl benzene + $(1-x)$ alkanes (C_7 to C_{16}) and + $(1-x)$ alkyl ethanoates$\}$ along to the corresponding correlation curves using the same equation (1). Now the k parameters, identified as $k_v = V^o_{m,2}/V^o_{m,1}$, are presented along with the a_i coefficients for each mixture in table 2. A comparison is made in figure 4(a) between our calculated V^E_m values at equimolar concentration and those from literature for (propylbenzene + alkanes) [7,8]

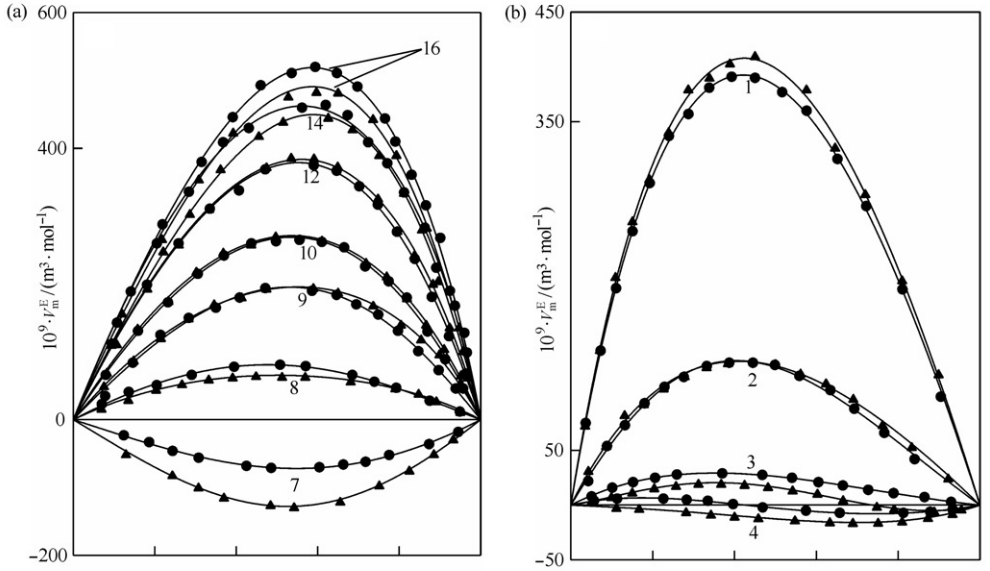

Figure 3. Experimental V^E_m values at two temperatures of 298.15 K (●) and 318.15 K (▲), and the corresponding correlation curves for binaries: (a), $\{xC_6H_5C_3H_7 + (1-x)\ C_nH_{2n+2}\}$ and (b), $\{xC_6H_5C_3H_7 + (1-x)\ CH_3CO_2C_vH_{2v+1}\}$. Labels correspond to values of n and v.

at 298.15 K. Figure 4(b) plots the corresponding experimental and correlated values for the mixtures containing alkylethanoates, the comparison in this case with literature [9] for V_m^E is carried out with values at an intermediate temperature of 303.15 K. The concordance appears to be acceptable in all cases.

Taking into account the graphical representations of the thermodynamic properties H_m^E and V_m^E for this set of mixtures the behavior is similar to that of the binaries containing butylbenzene published previously [1]. So, both properties are positive for the set $\{C_6H_5C_3H_7 + C_nH_{2n+2}$ ($n = 7$–10, 12, 14, 16) increasing regularly with the hydrocarbon chain-length increases. However, in those mixtures where the second component is an alkylethanoate the variation of the excess properties with v, $CH_3COOC_vH_{2v+1}$, is negative, decreasing quasi-exponentially with the increase of number of –CH_2– groups in ethanoate. The differences between the results obtained here and those from previous paper are explained according to the behavior model already described and where were considered several kinds of interactions. The interactions more relevant for mixtures with hydrocarbons were those corresponding to π-electrons of the benzene ring and the aromatic–apliphatic interactions although also should be taken into account the apliphatic–aliphatic interactions weaker than the two latter. Likewise in addition to the interactions mentioned we must further consider the possibility of dipole–dipole interaction in the mixtures with esters in spite of the small dipole moment of propylbenzene, $\mu \cdot 10^{30}/(\mathrm{Cm}) = 2.17$ [15] in comparison with those of ethanoates, > 5.5 which would explain the fact that the mixing enthalpies and excess volumes of ethanoates with propylbenzene diminish with the ethanoate alkanolic part increases, such as is shown in figures 2(b) and 3(b). Changes in the mixing effect are

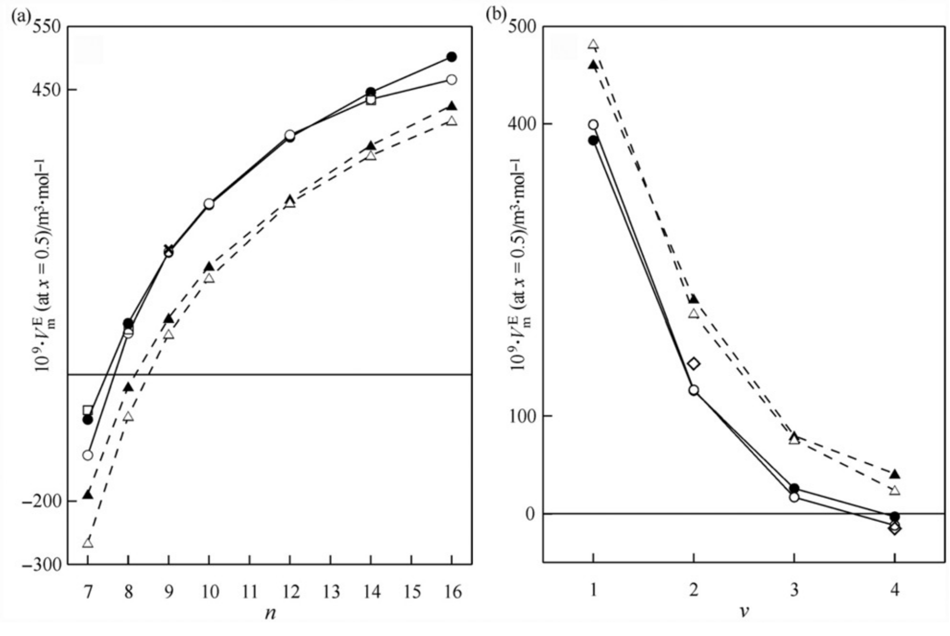

Figure 4. Comparison between equimolar V_m^E values at two temperatures, closed symbols at 298.15 K and open symbol at 318.15 K and those from literature for: (a), circle for $(C_6H_5C_3H_7 + C_nH_{2n+2})$, triangle for $(C_6H_5C_4H_9 + C_nH_{2n+2})$, and literature values (□), [7,8]; (✕), [8]; (b), circle for $(C_6H_5C_3H_7 + CH_3CO_2C_vH_{2v+1})$, triangle for $(C_6H_5C_4H_9 + CH_3CO_2C_vH_{2v+1})$, and literature values (◇), [9].

negative in the case of saturated hydrocarbon, with $(\partial V_m^E/\partial T)_{p,x} < 0$, because of the relative decreasing of empty spaces between molecules. However, in the mixtures (aromatic + ethanoates) the global volumetric effect is small and present a change of sign depending of the ethanoate chain. So, the volumetric coefficient $(\partial V_m^E/\partial T)_{p,x}$ is positive in the case of methyl ethanoate but become negative for butyl ethanoate due to the increase of the dipole moment of ethanoates, the same variation that ocurrs for butylbenzene mixtures, see figure 4(b).

4. Application of UNIFAC

For this work the prediction of H_m^E data was done using two versions of the UNIFAC group contribution model, the original one by Dang and Tassios [2] incorporating the parameters proposed in a previous paper [1], and another by Gmehling *et al.* [3]. This latter has a higher prediction capacity and can estimate VLE data and mixing enthalpies using the same set of interaction parameters. For the mixtures in this work, application of the method using both versions gave the results recorded graphically in figure 1(a) and 1(b), respectively, for alkanes and ethanoates with propylbenzene. The version of Gmehling *et al.* [3] produces values below real values, however, in order to afraid a recalculus of interaction parameters it would be necessary to have values of other thermodynamic quantities such as vapour–liquid equilibrium (VLE), liquid–liquid equilibrium (LLE), and others. On the other hand the version by Dang and Tassios [2] using the parameters obtained previously [1] in mixtures containing butylbenzene gave place to acceptable estimations for our mixtures although, such as is observed in

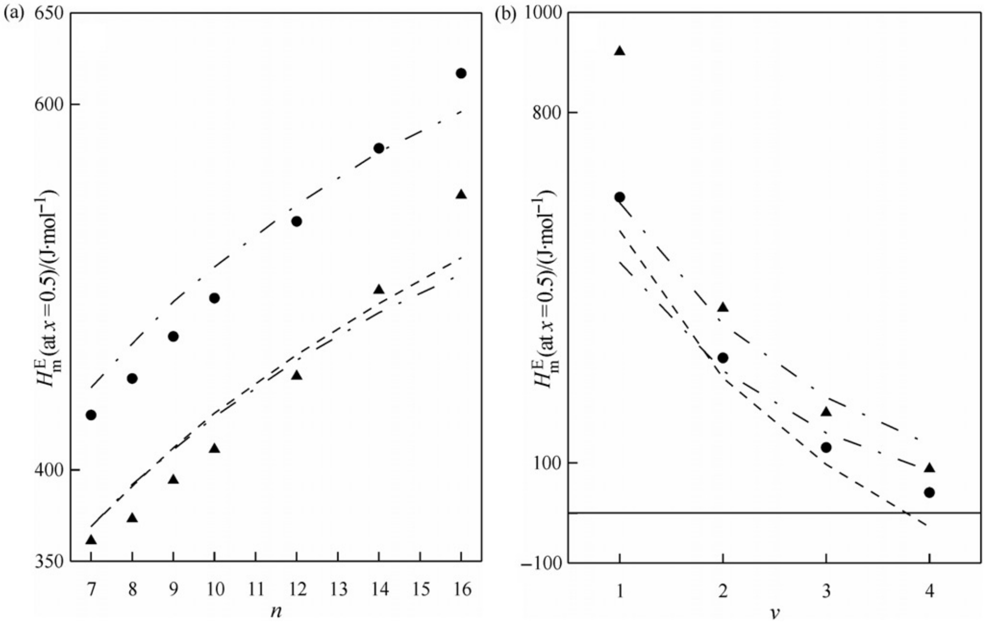

Figure 5. Plots of equimolar H_m^E values along to the predictions by two versions of UNIFAC, (— · — ·), [2] (– – –) [3]. (a), ($C_6H_5C_3H_7 + C_nH_{2n+2}$) (●), ($C_6H_5C_4H_9 + C_nH_{2n+2}$) (▲); (b), ($C_6H_5C_3H_7 + CH_3CO_2C_vH_{2v+1}$) (●), ($C_6H_5C_4H_9 + CH_3CO_2C_vH_{2v+1}$) (▲).

figure 1(a) and (b), the method is less precise for mixtures containing tetradecane and hexadecane. Therefore, in order to improve the application of this version of the method we have prepared a more wide database with enthalpies of those binary systems containing propyl and butyl benzene with alkanes and also other with data of the same aromatic hydrocarbons but with alkyl ethanoates. So, implementing the version of Dang and Tassios [2] in a least-squares procedure for non-linear function, Marquardt [16], a regression was carried out on each of the databases mentioned above, obtaining the following set of parameters: $a_{\mathrm{CH_2/ACH}} = -0.86$, $a_{\mathrm{ACH/CH_2}} = 19.20$; $a_{\mathrm{ACH/COOC}} = -1.64$, $a_{\mathrm{COOC/ACH}} = 32.65$. These values are very similar to those obtained previously [1] for binaries formed by butylbenzene. The representation in figure 5(a) and (b) shows that the estimations of the model are average values which present high discrepances in the extreme mixtures, (alkyl benzene + hexadecane) or (alkylbenzene + methyl ethanoate). Predictions with the version of Gmehling *et al.* [3] for the mixtures of this work are acceptable for alkyl ethanoates although predict negative values for (propylbenzene + butylethanoate) mixture but they are unacceptable for alkanes.

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