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Thermodynamic study of 2-methyl-tetrahydrofuran with isomeric chlorobutanes

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Abstract

Excess molar volumes, V^E, isentropic compressibility deviations, $\Delta\kappa_S$, and excess molar enthalpies, H^E, for the binary mixtures 2-methyl-tetrahydrofuran with 1-chlorobutane, 2-chlorobutane, 2-methyl-1-chloropropane and 2-methyl-2-chloropropane have been determined at temperatures 298.15 and 313.15 K, excess molar enthalpies were only measured at 298.15 K. We have applied the Prigogine–Flory–Patterson (PFP) theory to these mixtures at 298.15 K.

Keywords: Thermodynamic properties; 2-Methyl-tetrahydrofuran; Isomeric chlorobutanes; PFP theory

1. Introduction

Here we present excess molar volumes, V^E, isentropic compressibility deviations, $\Delta\kappa_S$, and excess molar enthalpies, H^E, of the binary mixtures 2-methyl-tetrahydrofuran with isomeric chlorobutanes: 1-chlorobutane, 2-chlorobutane, 2-methyl-1-chloropropane and 2-methyl-2-chloropropane at temperatures 298.15 and 313.15 K, H^E were only measured at 298.15 K. This work follows our previous papers in which we have reported thermodynamic and transport properties for cyclic ethers with chlorinated compounds [1–5] in order to study the Cl–O specific interaction.

The Prigogine–Flory–Patterson (PFP) theory [6,7] has been used to analyze the V^E and H^E results at 298.15 K. Furthermore, we have also applied this theory to predict at 298.15 K the isentropic compressibilities from excess molar enthalpies with the aim to test the reliability of the model.

As far as we know mixing thermodynamic properties for these systems have not been reported in the literature.

2. Experimental

The liquids used were: 2-methyl-tetrahydrofuran, 1-chlorobutane, 2-chlorobutane and 2-methyl-2-chloropropane, (>99%) provided by Aldrich as well as 2-methyl-1-chloropropane (>99%) obtained from Fluka. The purities of these compounds were checked by gas chromatography and no further purification was considered necessary.

The pure compound properties, densities and speeds of sound, at 298.15 and 313.5 K are collected in Table 1, together with literature density values at 298.15 K [8–10].

Densities, ρ, of the pure compounds and their mixtures were determined with an Anton Paar DMA-58 vibrating tube densimeter whose temperature was controlled within ±0.01 K. The accuracy of the densimeter after proper calibration is $\pm 10^{-5}$ g cm^{-3} and the precision of the density measurements was $\pm 5 \times 10^{-6}$ g cm^{-3}.

Speeds of sound, u, were obtained with an Anton Paar DSA-48 density and sound analyzer automatically thermostated at ±0.01 K. The accuracy of the speed of sound measurements is ± 1 m s^{-1} and the corresponding precision ± 0.1 m s^{-1}. The calibration of both apparatus, vibrating tube densimeter and density and sound analyzer,

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Table 1
Densities, ρ, and speeds of sound, u, at 298.15 and 313.15 K of the pure compounds, together with literature density values at 298.15 K

Compound	$T = 298.15$ K			$T = 313.15$ K	
	ρ (g cm^{-3})		u (m s^{-1})	ρ (g cm^{-3})	u (m s^{-1})
	Experimental	Literature			
2-Methyl-tetrahydrofuran	0.84920	0.84882[a]	1202.6	0.83388	1136.2
1-Chlorobutane	0.88069	0.88095[b]	1118.8	0.86386	1057.1
2-Chlorobutane	0.86737	0.8671[c]	1066.8	0.85016	1006.9
2-Methyl-1-chloropropane	0.87089	0.8717[b]	1079.2	0.85356	1016.3
2-Methyl-2-chloropropane	0.83567	0.8361[b]	983.1	0.81843	920.3

[a] Ref. [8].
[b] Ref. [9].
[c] Ref. [10].

was carried out with deionized doubly distilled water and dry air. Mixtures were prepared by mass using a Sartorius semi-micro balance with a precision of $\pm 10^{-5}$ g. The possible error in the mole fractions is estimated to be less than 10^{-4}.

Heats of mixing have been obtained with a Thermometric 2277 thermal activity monitor maintained at $\pm 2 \times 10^{-4}$ K operating under constant flow conditions. Two Shimadzu LC-10ADVP HPLC pumps were used to drive the liquids. The pumps were calibrated for each liquid before the start of the calorimetric measurements. The uncertainty in the mole fractions of the mixtures, calculated from the uncertainty in the flow delivered by the pumps, is ±0.001. The calibration of the calorimeter was achieved with reference to the very accurate H^E of the mixture n-hexane with cyclohexane [11]. This calibration was checked by determining the excess molar enthalpies of the endothermic system benzene + cyclohexane [12] and of the exothermic system 1,4-dioxane + tetrachloromethane [13] and comparing the obtained results with well-established literature data, in both cases the deviations lie within ±1% over the whole composition range. The accuracy in the determination of the heats of mixing could be expected to be ±1%. More details of procedure and calibration can be found in a previous paper [14].

3. Results

Excess molar volumes, V^E, were calculated from the density of the mixture, ρ, densities, ρ_i, and molar masses, M_i, of the pure compounds, and the corresponding molar fractions, x_i, by using the equation:

$$V^E = x_1\left(\frac{M_1}{\rho} - \frac{M_1}{\rho_1}\right) + x_2\left(\frac{M_2}{\rho} - \frac{M_2}{\rho_2}\right) \quad (1)$$

The measured densities and calculated excess molar volumes can be found in the supplementary material, excess molar volumes are graphically represented in Figs. 1 and 2.

Isentropic compressibilities, κ_S, and isentropic compressibility deviations, $\Delta\kappa_S$, can be obtained, assuming that ultrasonic absorption is negligible, from densities, ρ, and speeds of sound, u, by means of the relations:

$$\kappa_S = \frac{1}{\rho u^2} \quad (2)$$

$$\Delta\kappa_S = \kappa_S - \sum_i x_i \kappa_{S,i} \quad (3)$$

where x_i and $\kappa_{S,i}$ are, respectively, the mole fraction and the isentropic compressibility of the component i in the mixture. The experimental speeds of sound together with the estimated quantities κ_S and $\Delta\kappa_S$ are given in the supplementary material, isentropic compressibility deviations are represented in Figs. 3 and 4.

Finally, excess molar enthalpies H^E at 298.15 K are listed in Table 2 and represented in Fig. 5.

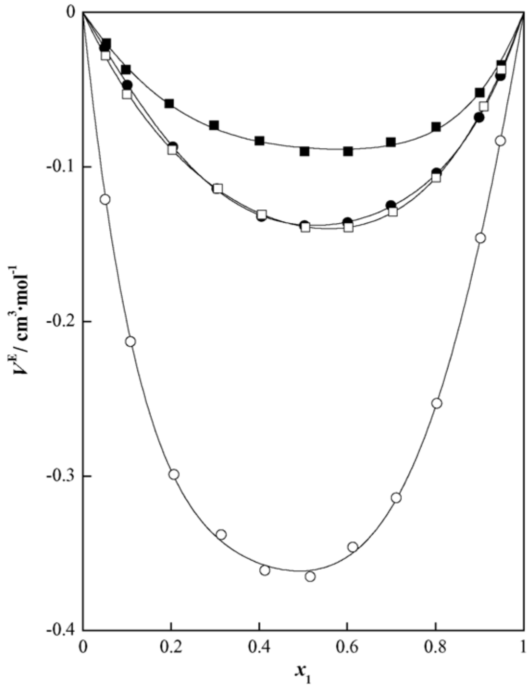

Fig. 1. Excess molar volumes, V^E, at 298.15 K for 2-methyl-tetrahydrofuran (1) + isomeric chlorobutanes (2): 1-chlorobutane (■); 2-chlorobutane (●); 2-methyl-1-chloropropane (□); 2-methyl-2-chloropropane (○).

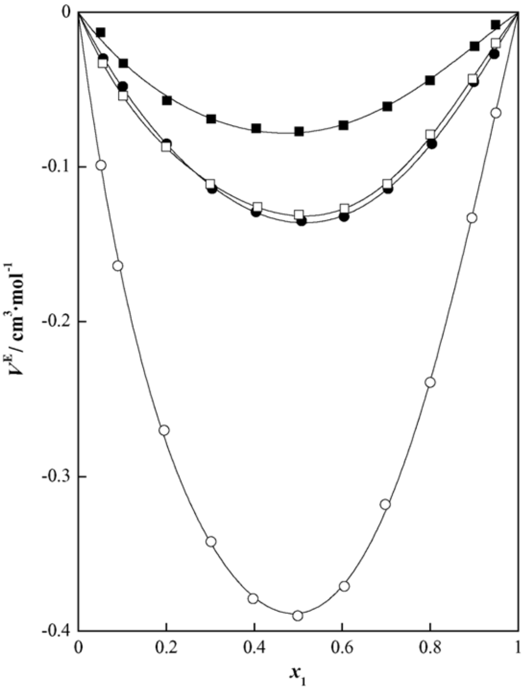

Fig. 2. Excess molar volumes, V^E, at 313.15 K for 2-methyl-tetrahydrofuran (1) + isomeric chlorobutanes (2): 1-chlorobutane (■); 2-chlorobutane (●); 2-methyl-1-chloropropane (□); 2-methyl-2-chloropropane (○).

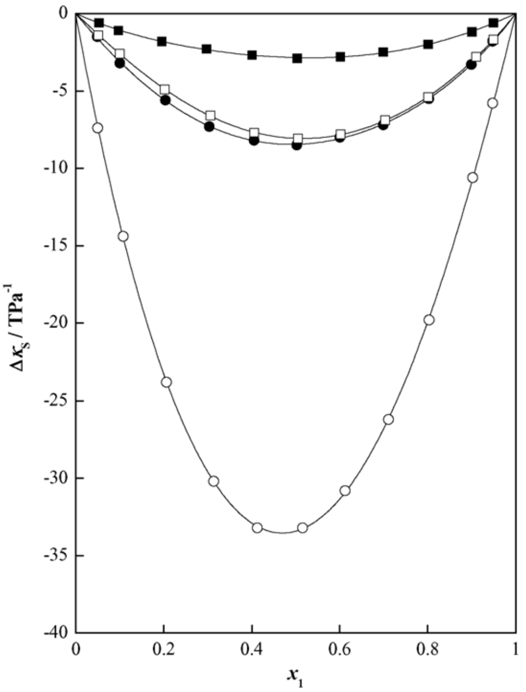

Fig. 3. Isentropic compressibility deviations, $\Delta\kappa_S$, at 298.15 K for 2-methyl-tetrahydrofuran (1) + isomeric chlorobutanes (2): 1-chlorobutane (■); 2-chlorobutane (●); 2-methyl-1-chloropropane (□); 2-methyl-2-chloropropane (○).

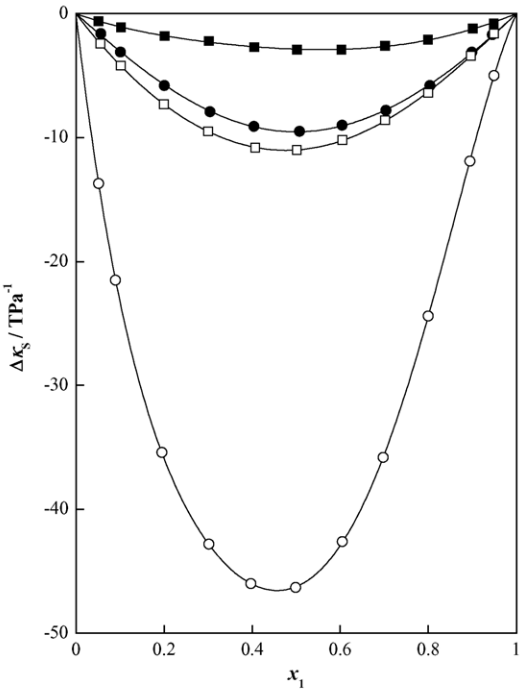

Fig. 4. Isentropic compressibility deviations, $\Delta\kappa_S$, at 313.15 K for 2-methyl-tetrahydrofuran (1) + isomeric chlorobutanes (2): 1-chlorobutane (■); 2-chlorobutane (●); 2-methyl-1-chloropropane (□); 2-methyl-2-chloropropane (○).

The values of each property at each temperature were correlated with a Redlich–Kister polynomial equation, in this equation $Y = V^E$ or $\Delta\kappa_S$ or H^E, and A_i are adjustable parameters.

Table 2
Excess molar enthalpies, H^E, of the binary mixtures

x_1	H^E (J mol^{-1})	x_1	H^E(J mol^{-1})	x_1	H^E (J mol^{-1})
2-Methyl-tetrahydrofuran + 1-chlorobutane at 298.15 K					
0.057	−28	0.398	−195	0.799	−147
0.104	−58	0.497	−210	0.901	−85
0.201	−116	0.597	−207	0.954	−43
0.298	−159	0.696	−188		
2-Methyl-tetrahydrofuran + 2-chlorobutane at 298.15 K					
0.057	−19	0.398	−138	0.799	−105
0.104	−42	0.497	−149	0.901	−59
0.201	−83	0.597	−150	0.954	−30
0.298	−116	0.696	−135		
2-Methyl-tetrahydrofuran + 2-methyl-1-chloropropane at 298.15 K					
0.060	−33	0.411	−211	0.806	−151
0.109	−67	0.511	−224	0.904	−84
0.210	−132	0.609	−220	0.954	−43
0.309	−174	0.707	−196		
2-Methyl-tetrahydrofuran + 2-methyl-2-chloropropane at 298.15 K					
0.057	−14	0.398	−101	0.799	−77
0.104	−30	0.497	−109	0.901	−44
0.201	−61	0.597	−106	0.954	−23
0.298	−83	0.696	−95		

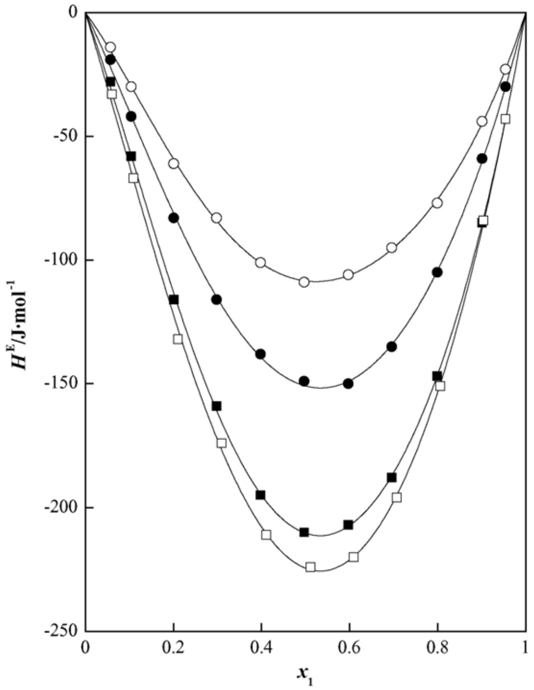

Fig. 5. Excess molar enthalpies, H^E, at 298.15 K for 2-methyl-tetrahydrofuran (1) + isomeric chlorobutanes (2): 1-chlorobutane (■); 2-chlorobutane (●); 2-methyl-1-chloropropane (□); 2-methyl-2-chloropropane (○).

$$Y = x_1(1-x_1)[A_0 + A_1(2x_1-1) + A_2(2x_1-1)^2 + A_3(2x_1-1)^3] \tag{4}$$

The values of the parameters A_i together with the standard deviations $\sigma(Y)$ are given in Table 3.

Excess molar volumes are negative over the whole composition range for all the systems at both temperatures 298.15 and 313.15 K. The system containing 1-chlorobutane shows the smallest negative V^E values while the mixture with 2-methyl-2-chloropropane presents the biggest negative V^E values, on the other hand the mixtures containing 2-chlorobutane and 2-methyl-1-chloropropane exhibit a similar volumetric behaviour and their values are intermediate between the above-mentioned systems. The effect of the temperature is not very marked, for the mixtures containing 1-chlorobutane, 2-chlorobutane or 2-methyl-1-chloropropane the excess molar volumes become a little less negative with temperature while for 2-methyl-2-chloropropane the V^E values become slightly more negative when the temperature rises.

$\Delta\kappa_S$ is negative over the whole composition range showing the mixture containing 1-chlorobutane the lowest negative $\Delta\kappa_S$ values and the mixture with 2-methyl-2-chloropropane the biggest ones, the mixtures containing 2-chlorobutane and 2-methyl-1-chloropropane present similar $\Delta\kappa_S$ values and these values are again intermediate between those of 1-chlorobutane and 2-methyl-2-chloropropane. Isentropic compressibility deviations of all mixtures increase slightly in absolute value when the temperature increases, although for the mixture 2-methyl-tetrahydrofuran with 1-chlorobutane the change with the temperature is negligible.

Excess molar enthalpies are negative for the four systems wherever mole fraction. Absolute H^E values follow the sequence: 2-methyl-2-chloropropane < 2-chlorobutane < 1-chlorobutane < 2-methyl-1-chloropropane. The minimum H^E values are slightly shifted towards high molar fractions of 2-methyl-tetrahydrofuran.

4. Prigogine–Flory–Patterson theory

The Prigogine–Flory–Patterson theory considers excess thermodynamic properties of binary mixtures to be the sum of different contributions. The excess molar volume can be expressed as: an interactional term which is proportional to the interaction parameter, χ_{12}, a free volume contribution which arises from the dependence of the reduced volume upon the reduced temperature as a result of the differences between the degrees of thermal expansion of the components and an internal pressure contribution which depends both in the difference of internal pressures and of the reduced volumes of the components:

$$\frac{V^E}{x_1V_1^* + x_2V_2^*} = \frac{(\tilde{V}^{1/3}-1)\,\tilde{V}^{2/3}\psi_1\theta_2\chi_{12}}{[(4/3)\,\tilde{V}^{-1/3}-1]P_1^*} - \frac{(\tilde{V}_1-\tilde{V}_2)^2[(14/9)\,\tilde{V}^{1/3}-1]\psi_1\psi_2}{[(4/3)\tilde{V}^{-1/3}-1]\tilde{V}} + \frac{(\tilde{V}_1-\tilde{V}_2)(P_1^*-P_2^*)\psi_1\psi_2}{P_1^*\psi_1+P_2^*\psi_2} \tag{5}$$

With respect to the excess molar enthalpy, it can be divided into an interactional contribution and a free volume contribution:

$$\frac{H^E}{x_1U_1^* + x_2U_2^*} = \frac{\left[-\tilde{U} + \tilde{T}\tilde{C}_P\right]\,\psi_1\theta_2\chi_{12}}{P_1^*} + \tilde{C}_P[\psi_1\tilde{T}_1 + \psi_2\tilde{T}_2 - \tilde{T}] \tag{6}$$

In these equations the reduced magnitudes for the mixture: $\tilde{V}$, $\tilde{T}$ and $\tilde{C}_P$ can be obtained from the reduced configurational energy, $\tilde{U}$, that was calculated, according to Barbe and Patterson [15], in terms of the reduced configurational energies of the pure components by:

$$\tilde{U} = \psi_1\tilde{U}_1 + \psi_2\tilde{U}_2 \tag{7}$$

where the contact energy fraction, ψ_i, is defined as:

$$\psi_1 = 1 - \psi_2 = \frac{x_1P_1^*V_1^*}{x_1P_1^*V_1^* + x_2P_2^*V_2^*} \tag{8}$$

All other parameters in the above equations can be obtained using Flory's theory [16–19].

Table 3
Parameters, A_i, and standard deviations, $\sigma(Y)$, for Eq. (4)

Function	T (K)	A_0	A_1	A_2	A_3	$\sigma(Y)$
2-Methyl-tetrahydrofuran + 1-chlorobutane						
V^E (cm^3 mol^{-1})	298.15	−0.350	−0.047	−0.211	−0.102	0.001
	313.15	−0.312	0.037	0.023	0.053	0.002
κ_S (TPa^{-1})	298.15	−11.4	−1.3	−1.6	1.1	0.0
	313.15	−11.4	−2.4	−2.1	1.7	0.1
H^E (J mol^{-1})	298.15	−842	−122	87	−144	2
2-Methyl-tetrahydrofuran + 2-chlorobutane						
V^E (cm^3 mol^{-1})	298.15	−0.550	−0.041	−0.126	−0.166	0.001
	313.15	−0.544	−0.031	0.029	0.074	0.001
κ_S (TPa^{-1})	298.15	−33.8	1.2	−2.3	−3.4	0.1
	313.15	−38.1	0.0	0.6	−0.1	0.1
H^E (J mol^{-1})	298.15	−604	−93	70	−78	2
2-Methyl-tetrahydrofuran + 2-methyl-1-chloropropane						
V^E (cm^3 mol^{-1})	298.15	−0.554	−0.097	−0.158	−0.004	0.001
	313.15	−0.526	−0.029	0.004	0.184	0.001
κ_S (TPa^{-1})	298.15	−32.3	−2.1	0.9	−2.1	0.0
	313.15	−44.0	4.5	4.0	2.2	0.1
H^E (J mol^{-1})	298.15	−898	−136	103	−76	2
2-Methyl-tetrahydrofuran + 2-methyl-2-chloropropane						
V^E (cm^3 mol^{-1})	298.15	−1.445	0.023	−0.772	0.537	0.003
	313.15	−1.554	0.059	−0.163	0.421	0.003
κ_S (TPa^{-1})	298.15	−133.7	16.0	−3.3	4.1	0.1
	313.15	−184.9	28.8	−9.9	86.8	0.1
H^E (J mol^{-1})	298.15	−434	−4	37	−111	1

Oswal [20] extended the Prigogine–Flory–Patterson theory to estimate the isentropic compressibilities and speeds of sound of liquid mixtures. At a given temperature, T, the PFP theory can be used to calculate the molar volumes, V, the molar heat capacities, C_P of a liquid mixture if the interaction parameter, χ_{12}, is known. The terms $(\partial V/\partial T)_P$ and $(\partial V/\partial P)_T$ can be also calculated by means of the following equations:

$$\left(\frac{\partial V}{\partial P}\right)_T = \frac{-\tilde{V}^{7/3}+2\tilde{V}^{2}-2\tilde{V}^{5/3}}{(4/3)-\tilde{V}^{1/3}}\cdot\frac{V^*T^*}{P^*T} \qquad (9)$$

$$\left(\frac{\partial V}{\partial T}\right)_P = \frac{\tilde{V}(\tilde{V}^{1/3}-1)}{(4/3)-\tilde{V}^{1/3}}\cdot\frac{V^*}{T} \qquad (10)$$

where V^*, P^* and T^* are the characteristic volume, pressure and temperature of the mixture, respectively.

From all these quantities the isentropic compressibility, $\kappa_S = -V^{-1}(\partial V/\partial P)_S$, can be obtained using the following thermodynamic relation:

$$\left(\frac{\partial V}{\partial P}\right)_S = \left(\frac{\partial V}{\partial P}\right)_T + TC_P^{-1}\left(\frac{\partial V}{\partial T}\right)_P^2 \qquad (11)$$

Flory parameters of the pure compounds along with their physical properties are gathered in Table 4. Thermal expansion coefficients, α, were derived from measured densities in this laboratory. Isothermal compressibilities, κ_T, were calculated from thermal expansion coefficients, experimental isentropic compressibilities and molar heat capacities [1,21]. The number of contact sites per segment of a molecule, s, has been estimated using the Bondi's method [22].

In this work, the interaction parameter for each mixture was obtained by fitting the PFP theory to the corresponding experimental equimolar H^E values. Once the interaction parameter is obtained the thermodynamic properties of the mixture can be estimated.

Table 4
Physical properties and Flory parameters of the pure compounds at 298.15 K

Compound	α (kK^{-1})	$C_{P,m}$ (J mol^{-1})	κ_T (TPa^{-1})	$\tilde{V}$	P^* (J cm^{-3})	s (Å^{-1})
2-Methyl-tetrahydrofuran	1.239	161.6[a]	1110.1	1.295	558	1.25
1-Chlorobutane	1.216	158.9[b]	1200.7	1.290	502	1.42
2-Chlorobutane	1.313	160.9[b]	1355.5	1.309	495	1.41
2-Methyl-1-chloropropane	1.183	158.7[b]	1269.2	1.284	458	1.42
2-Methyl-2-chloropropane	1.437	162[b]	1656.2	1.331	458	1.45

[a] Ref. [1].
[b] Ref. [21].

Table 5
Comparison between experimental and theoretical V^E and κ_S at equimolar composition together with calculated contributions to V^E and H^E at equimolar composition at 298.15 K

Function	Experimental	PFP	Interactional	Free volume	Internal pressure
2-Methyl-tetrahydrofuran + 1-chlorobutane ($\chi_{12} = -9.4\ \mathrm{J\,cm^{-3}}$)					
V^E ($\mathrm{cm^3\,mol^{-1}}$)	−0.088	−0.168	−0.178	−0.001	0.011
H^E ($\mathrm{J\,mol^{-1}}$)	−210	−210	−210	0	
κ_S ($\mathrm{TPa^{-1}}$)	857.8	860.5			
2-Methyl-tetrahydrofuran + 2-chlorobutane ($\chi_{12} = -6.7\ \mathrm{J\,cm^{-3}}$)					
V^E ($\mathrm{cm^3\,mol^{-1}}$)	0.138	−0.172	−0.132	−0.006	−0.034
H^E ($\mathrm{J\,mol^{-1}}$)	−149	−149	−149	−1	
κ_S ($\mathrm{TPa^{-1}}$)	905.2	908.9			
2-Methyl-tetrahydrofuran + 2-methyl-1-chloropropane ($\chi_{12} = -9.8\ \mathrm{J\,cm^{-3}}$)					
V^E ($\mathrm{cm^3\,mol^{-1}}$)	−0.139	−0.154	−0.193	−0.004	0.044
H^E ($\mathrm{J\,mol^{-1}}$)	−222	−222	−221	−1	
κ_S ($\mathrm{TPa^{-1}}$)	892.0	897.7			
2-Methyl-tetrahydrofuran + 2-methyl-2-chloropropane ($\chi_{12} = -4.5\ \mathrm{J\,cm^{-3}}$)					
V^E ($\mathrm{cm^3\,mol^{-1}}$)	−0.362	−0.281	−0.099	−0.038	−0.143
H^E ($\mathrm{J\,mol^{-1}}$)	−108	−108	−103	−5	
κ_S ($\mathrm{TPa^{-1}}$)	992.7	998.1			

Table 5 provides the comparison between experimental and theoretical V^E and κ_S at equimolar composition together with calculated contributions to V^E and H^E at equimolar composition, in this table the interaction parameter for each system are also shown.

The estimation of excess molar volumes from H^E values can be considered, in a general way, satisfactory and this prediction show good quantitative agreement for the mixtures containing 2-chlorobutane and 2-methyl-1-chloropropane. The interactional contribution is negative for all the systems, showing the existence of a specific interaction between the mixed molecules, and this contribution is the dominant term for the mixtures containing 1-chlorobutane, 2-chlorobutane and 2-methyl-1-chloropropane. The free volume contribution is negative for the four systems and very small except for the mixture with 2-methyl-2-chloropropane due to the high thermal expansion coefficient of this compound. The third contribution, that is the internal pressure contribution, is positive for the mixtures with 1-chlorobutane and 2-methyl-1-chloropropane and negative for the mixtures with 2-chlorobutane and 2-methyl-2-chloropropane in which is the dominant term.

In the case of excess molar enthalpies the interactional contribution is negative and is the dominant term. This contribution depends, apart other factors, of the interaction parameter, χ_{12}, which characterizes the difference between the energy of interaction among sites on neighbouring molecules of different species and the average of the interaction energies in the pure components; the sign and magnitude of the interactional contribution for the mixtures studied here reflects the existence of a specific interaction among unlike molecules [23]. This interaction takes place between a lone pair of electrons of the oxygen atom in the cylic ether [24] and the Cl atom that is the acceptor atom. For the mixture 2-methyl-tetrahydrofuran with 2-methyl-2-chloropropane is less negative probably due to steric hindrance to establish the Cl–O interaction. On the other hand the free volume contribution is negligible for the mixtures containing 1-chlorobutane, 2-chlorobutane and 2-methyl-1-chloropropane and very small for 2-methyl-2-chloropropane.

With respect to the isentropic compressibilities it can be seen in supplementary data that there is excellent agreement between estimated and experimental values, for all the mixtures the deviation from experimental data are less than 0.6%. It can be pointed out that for these mixtures the theory slightly overestimates the isentropic compressibilities.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.tca.2004.11.034.

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