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Isothermal vapour–liquid equilibria in cyclohexanone + dichloroalkane binary mixtures at temperatures from 298.15 to 318.15 K

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Abstract: The vapour pressures of binary mixtures of cyclohexanone + dichloroalkane (1,3-dichloropropane and 1,4-dichlorobutane) were measured at temperatures between 298.15 and 318.15 K. The vapour pressures vs. liquid phase composition data were used to calculate the activity coefficients of the two components and the excess molar Gibbs energies G^E for the mixtures, using the Barker method and the Redlich–Kister, Wilson, NRTL and UNIQUAC equations, taking into account the vapour phase imperfection in terms of the 2nd virial coefficient. No significant difference between the G^E values obtained with these equations was observed.

Keywords: vapour pressure; vapour–liquid equilibria; excess Gibbs energy; mixtures; cyclohexanone; dichloroalkanes.

INTRODUCTION

As part of a series of experimental vapour–liquid equilibria (VLE) studies on mixtures of cycloketones with chloroalkanes, measurements on (cyclohexanone + 1,3-dichloropropane and + 1,4-dichlorobutane) binary mixtures, for which no such experimental data are available,^{1,2} are reported herein.

In addition, no experimental excess Gibbs free energy, G^E , or excess enthalpy, H^E , data for these mixtures could be found in the literature.²

In previous papers, experimental VLE data for cyclopentanone + 1,2-dichloroethane and + 1,1,1-trichloroethane,³ 1,1,2,2-tetrachloroethane + cyclopentanone and + cyclohexanone,⁴ cyclopentanone + 1,3-dichloropropane, + 1,4-dichlorobutane, + 1-chlorobutane,⁵ 1,2-dichloroethane + cyclohexanone, chloro-

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form + cyclopentanone and chloroform + cyclohexanone⁶ and cyclohexanone + 1-chlorobutane, + 1,1,1-trichloroethane⁷ were reported.

To correlate the experimental VLE data, different expressions for excess Gibbs energy G^E , *i.e.*, Redlich–Kister,⁸ Wilson,⁹ NRTL¹⁰ and UNIQUAC¹¹ equations were used.

These studies will be used to estimate the interaction parameters for the group contribution methods DISQUAC, for a possible comparison with *n*-alkanone + chloroalkane systems in terms of molecular surface interaction, steric and ring strain effects, and electron-donor ability of the cyclo-carbonyl group with chloroalkanes.

EXPERIMENTAL

Materials

The chemicals used, of the highest available purity, were commercial products from Aldrich (cyclohexanone and 1,3-dichloropropane) and Merck (1,4-dichlorobutane). The purity of the substances, checked by gas chromatography, was not less than 99.8 mol %. Evidence of chemical purity was also provided by comparison of the measured refractive indices, $n_D(298.15\text{ K})$, densities, $\rho(298.15\text{ K})$ and vapour pressure with the literature values, given in Table I.

The liquids were dried and stored over 4Å molecular sieves and used without further purification.

TABLE I. Physical properties of the pure compounds at 298.15 K

Temperature, K	n_D		$\rho / \text{kg m}^{-3}$		p / kPa	
	This work	Literature	This work	Literature	This work	Literature
Cyclohexanone						
298.15	1.4482	1.4480 ¹²	943.0	942.5 ¹²	0.64	0.58 ¹³
308.15					1.16	1.05 ¹³
318.15					1.92	1.79 ¹³
1,3-Dichloropropane						
298.15	1.4459	1.4460 ¹⁴	1181.4	1181.8 ¹⁴	2.72	2.24 ¹⁵
308.15					4.40	3.82 ¹⁵
318.15					6.89	6.26 ¹⁵
1,4-Dichlorobutane						
298.15	1.4519	1.4518 ¹⁴ 1.4522 ¹⁶	1132.8	1134.02 ¹⁶ 1133.1 ¹⁷	0.78	0.56 ¹³ , 0.77 ¹⁸
308.15					1.38	1.00 ¹³ , 1.25 ¹⁸
318.15					2.10	1.74 ¹³ , 1.99 ¹⁸

Apparatus and procedure

The vapour pressure, p , of the pure compounds and the binary mixtures were measured by a static method, in which the total pressure was measured as a function of the overall composition in an equilibrium cell.

Use was made of an isoteniscope based on the Surovy design.¹⁹ The working procedure and the performance of the apparatus were described in previous papers^{20,21} and detailed in other.^{6,7}

The equilibrium cell, of total volume of 80 cm³, was tightly connected with an Hg-filled U-tube as a null manometer surrounded by a thermostated jacket. The cell was equipped with other fittings-on the thermostated mantle. The temperature of this mantle was the actually equilibrium temperature, T , while the temperature of the jacket of the null manometer was maintained 1 to 2 K higher in order to prevent the partial condensation of vapours in the upper part of the apparatus. The isoteniscope was joined *via* the null manometer to an external mercury manometer, which enabled the pressure to be accurately measured within the range of 0.1–100 kPa.

After thermostating the equilibrium cell at the required temperature, the difference of the mercury levels in the null manometer was equalized with dry air and the pressure, p , was read on the precision Hg manometer connected to the apparatus. The manometric readings were performed with a Griffin and George Ltd. (London, UK), type 4214 cathetometer, to within ± 0.01 mm, equivalent to an uncertainty of ± 3 Pa. The measured equilibrium pressures were reproducible to better than 20 Pa. In order to avoid modifications of the cell volume, the level of the mercury in the null manometer was maintained always at the same position. In this way, the volume of the vapour space in the cell was kept nearly constant (70 cm³).

Mixtures of known composition of about 10 cm³ were prepared by weighing to within 10⁻⁷ kg and thorough degassing in the equilibrium cell by alternate freezing, high vacuum pumping and thawing, as described by Ronc²² and Young.²³ During the vapour pressure measurement, the liquid in the equilibrium cell was stirred by means of a magnetic stirrer.

The equilibrium temperature, T , was measured with an accuracy of 0.1 K against IST-90 (International Temperature Scale of 1990) by means of mercury thermometers previously checked at the National Institute of Metrology, Bucharest, Romania.

Finally, the experimental uncertainties were: $\sigma T = 0.1$ K, $\sigma p = 0.02$ kPa, $\sigma x_i = 0.001$ for temperature, pressure and molar fraction, respectively.

RESULTS

The vapour pressures of the pure components, cyclohexanone, 1,3-dichloropropane and 1,4-dichlorobutane, were measured in the same apparatus at the working temperatures and were in good agreement with the literature values (Table I).

The direct experimental values, x - p - T , and the calculated vapour phase compositions, y , for the binary systems cyclohexanone + 1,3-dichloropropane and + 1,4-dichlorobutane at temperatures from 298.15 to 318.15 K are presented in Tables II and III.

Figures 1 and 2 show the experimental and calculated isotherms fitted to the 3rd order Redlich–Kister Equation for G^E :

$$G^E = RTx_1x_2 \sum A_i (x_1 - x_2)^{i-1} \quad (1)$$

A good agreement between the data was observed.

TABLE II. Experimental VLE data for 1,3-dichloropropane (x) + cyclohexanone ($1-x$) mixtures

x	y_{calc}	$p_{\text{exp}} / \text{kPa}$	$(p_{\text{exp}} - p_{\text{cal}}) / \text{kPa}$	$G^E / \text{J mol}^{-1}$
$T = 298.15 \text{ K}$				
0.000	0.000	0.64	0	0
0.100	0.210	0.72	0.00	-163
0.176	0.368	0.79	-0.02	-259
0.267	0.533	0.92	-0.02	-343
0.348	0.654	1.12	0.03	-390
0.430	0.751	1.28	0.00	-414
0.565	0.861	1.62	0.03	-403
0.667	0.915	1.88	0.02	-356
0.729	0.940	2.01	-0.02	-312
0.825	0.968	2.22	-0.06	-223
0.909	0.986	2.43	-0.06	-124
0.958	0.994	2.58	-0.03	-60
1.000	1.000	2.72	0	0
$T = 308.15 \text{ K}$				
0.000	0.000	1.16	0	0
0.100	0.207	1.18	-0.12	-135
0.176	0.352	1.40	-0.05	-221
0.267	0.504	1.62	-0.03	-303
0.348	0.620	1.94	0.08	-357
0.430	0.718	2.19	0.07	-391
0.565	0.838	2.61	0.02	-402
0.667	0.901	3.01	0.01	-370
0.729	0.930	3.21	-0.05	-332
0.825	0.964	3.58	-0.10	-246
0.909	0.985	3.90	-0.13	-142
0.958	0.994	4.16	-0.07	-71
1.000	1.000	4.40	0	0
$T = 318.15 \text{ K}$				
0.000	0.000	1.92	0	0
0.100	0.218	2.11	-0.09	-100
0.176	0.361	2.40	-0.05	-168
0.267	0.504	2.76	-0.01	-233
0.348	0.613	3.16	0.05	-279
0.430	0.707	3.62	0.12	-311
0.565	0.825	4.13	-0.06	-327
0.667	0.890	4.84	0.04	-306
0.729	0.921	5.12	-0.07	-278
0.825	0.959	5.75	-0.04	-209
0.909	0.982	6.31	-0.03	-122
0.958	0.993	6.55	-0.09	-61
1.000	1.000	6.89	0	0

TABLE III. Experimental VLE data for 1,4-dichlorobutane (x) + cyclohexanone ($1-x$) mixtures

x	y_{calc}	$p_{\text{xp}} / \text{kPa}$	$(p_{\text{exp}} - p_{\text{calc}}) / \text{kPa}$	$G^E / \text{J mol}^{-1}$
$T = 308.15 \text{ K}$				
0.000	0.000	1.16	0.00	0
0.067	0.086	1.19	0.02	14
0.121	0.149	1.20	0.01	24
0.200	0.241	1.23	0.00	38
0.296	0.344	1.25	-0.00	51
0.418	0.467	1.27	-0.01	61
0.522	0.566	1.30	-0.01	64
0.634	0.669	1.33	-0.00	62
0.712	0.739	1.35	0.00	56
0.812	0.828	1.37	0.01	42
0.898	0.905	1.38	0.01	26
1.000	1.000	1.38	0.00	0
$T = 318.15 \text{ K}$				
0.0000	0.000	1.92	0.00	0
0.067	0.075	1.93	0.00	4
0.121	0.133	1.94	0.00	7
0.200	0.218	1.95	-0.00	10
0.296	0.318	1.99	0.00	13
0.418	0.442	2.00	0.00	16
0.522	0.544	2.03	0.00	16
0.634	0.653	2.04	-0.01	15
0.712	0.728	2.06	0.00	13
0.812	0.823	2.08	0.00	10
0.898	0.904	2.09	0.00	6
1.000	1.000	2.10	0.00	0

CORRELATION AND DISCUSSION

The vapour pressures of the pure component agree fairly well with literature data in the range of the VLE measurements (Table I). The literature values for vapour pressure shown in Table I were calculated using specific equations, as given in the mentioned references.

The isothermal (vapour-liquid) equilibrium data of the mixtures were correlated by Barker's method²⁴ using well-known expressions for G^E , *i.e.*, the Redlich-Kister,⁸ Wilson,⁹ NRTL¹⁰ and UNIQUAC Equations.¹¹

The model coefficients were determinate by regression through minimization of the objective function Q , Eq. (2):

$$Q = \sum_j^N \left[\frac{p_{\text{calc},j} - p_{\text{exp},j}}{p_{\text{exp},j}} \right]^2 \quad (2)$$

where the subscripts calc and exp denote the calculated and experimental values of the pressure of an experimental point j , N being the total number of experimental points for one isotherm.

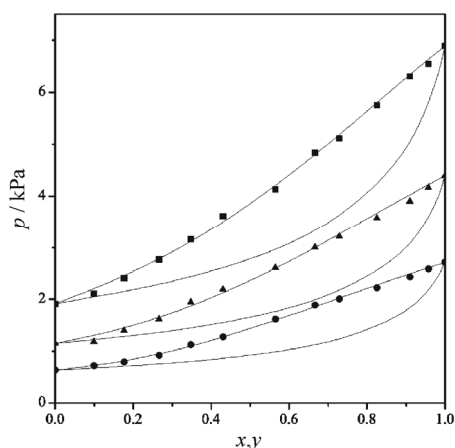


Fig. 1. Isothermal VLE for 1,3-dichloropropane (x) + cyclohexanone ($1-x$) mixtures. The symbols represent the experimental data at $T = 298.15$ (●), 308.15 (▲) and 318.15 (■) K and the curves were obtained from a 3rd-order Redlich–Kister Equation.

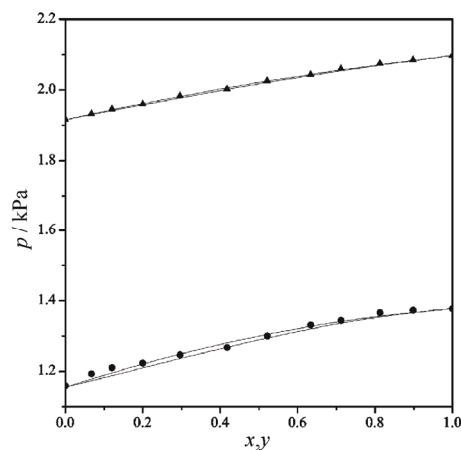


Fig. 2. Isothermal VLE for 1,4-dichlorobutane (x) + ($1-x$) cyclohexanone mixture. The symbols represent the experimental data at $T = 308.15$ (●), 318.15 (▲) K and the curves were obtained from a 3rd-order Redlich–Kister Equation.

Vapour phase imperfection was accounted for in terms of the 2nd molar virial coefficient, estimated by the method of Tsonopoulos²⁵, while the molar volumes were calculated by the Rackett²⁶ Equation with coefficients taken from Lide and Kehiaian.²⁷ Critical properties of substances were used as founded in the report of Ambrose.²⁸

For the mixtures under study, the standard deviation σp values were between 3.0 and 76 Pa (Table IV).

In this Table, A_1 , A_2 and A_3 denote the binary parameters of the models: ($k_{ij} - k_{ji}$) for the Wilson Equation, ($g_{ij} - g_{ji}$) for the NRTL equation and ($u_{ij} - u_{ji}$) for the UNIQUAC Equation and they represent the energies of interactions between unlike molecules 1–2 and 2–1 (being expressed in J mol^{-1}). For the Redlich–Kister Equation, they are without physical significance. The third NRTL parameter, α , is related to the non-randomness in the mixtures that for the studied class of systems had a fixed value of 0.3.

For the mixture 1,3-dichloropropane + cyclohexanone, the correlation with NRTL and UNIQUAC models failed, probably due to some numerical problems in the fitting procedure.

The results presented in Figs. 1 and 2 show that the studied systems behave differently. For the mixture 1,3-dichloropropane + cyclohexanone, the deviations from the Raoult Law were negative, while the mixture 1,4-dichlorobutane + cyclohexanone showed small positive deviations from the Raoult Law.

TABLE IV. Parameters of the equations used to correlate the VLE data for 1,3-dichloropropane (x) + cyclohexanone ($1-x$) and 1,4-dichlorobutane (x) + cyclohexanone ($1-x$) binary mixtures and the standard deviation σp at 298.15, 308.15 and 318.15 K

T / K	A_1 or A_{12}	A_2 or A_{21}	A_3	$\sigma p^a / \text{kPa}$
1,3-Dichloropropane (x) + cyclohexanone ($1-x$)				
Redlich–Kister Equation (3 rd -order)				
298.15	-0.67100	0.007450	–	0.033
308.15	-0.63092	-0.05575	–	0.076
318.15	-0.49198	-0.08619	–	0.068
Redlich–Kister Equation (4 th -order)				
298.15	-0.69235	0.21735	-0.27949	0.027
308.15	-0.65264	0.21043	-0.56714	0.040
318.15	-0.49341	0.02619	-0.26056	0.049
Wilson Equation				
298.15	-176.1683	-0.9786	–	0.032
308.15	7.0019	-172.6808	–	0.075
318.15	82.1212	-199.5170	–	0.065
1,4-Dichlorobutane (x) + cyclohexanone ($1-x$)				
Redlich–Kister Equation (3 rd order)				
308.15	0.10047	0.01354	–	0.010
318.15	0.02447	0.0004	–	0.004
Redlich–Kister Equation (4 th order)				
308.15	0.06521	0.01080	0.21734	0.003
318.15	0.02113	0.00023	0.02081	0.004
Wilson Equation				
308.15	188.0812	-118.4118	–	0.009
318.15	5.3689	2.4429	–	0.004
NRTL Equation ($\alpha = 0.3$)				
308.15	195.9862	-137.5274	–	0.010
318.15	6.0756	1.7278	–	0.004
UNIQUAC Equation				
308.15	-86.2824	47.8833	–	0.010
318.15	-79.6637	31.8715	–	0.004

^athe average standard deviation of the total vapour pressure: $\sigma p = (\sum (p_{\text{exp}} - p_{\text{calc}})^2 / (N - m))^{1/2}$, where N is the number of experimental points and m is the number of equation parameters

The calculated excess Gibbs energy G^E increases with increasing temperature, for the first mixture and decreases with increasing temperature, for the second mixture, as is observed in Figs. 3 and 4.

The equimolar excess enthalpies, H^E , estimated from the temperature dependence of G^E , for the 1,3-dichloropropane + cyclohexanone mixture are: 1761 J mol⁻¹, at an average temperature of 308.15 K, and 1543 J mol⁻¹ for the 1,4-dichlorobutane + cyclohexanone mixture, at an average temperature of 313.15 K. However, it is well known that the calculation of excess enthalpy from vapour pressure data implies a great uncertainty,²⁹ which was also mentioned by other authors, also very recently³⁰; hence, in the absence of the calorimetric data, the calculated H^E values are qualitative only.

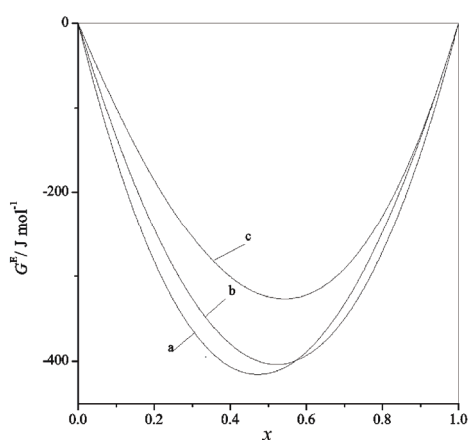


Fig. 3. Molar excess Gibbs energies for 1,3-dichloropropane (x) + cyclohexanone ($1-x$) mixtures at $T = 298.15$ (a), 308.15 (b), 318.15 (c) K.

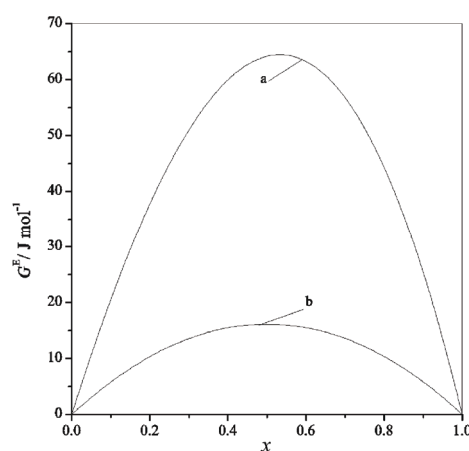


Fig. 4. Molar excess Gibbs energies for 1,4-dichlorobutane (x) + ($1-x$) cyclohexanone mixtures at $T = 308.15$ (a), 318.15 (b) K.

The behaviour of the system 1,4-dichlorobutane + cyclohexanone, with small positive values of G^E , unlike the negative values for cyclohexanone + 1,2-dichloroethane⁶ and + 1,3-dichloropropane shows a decrease in specific inter-

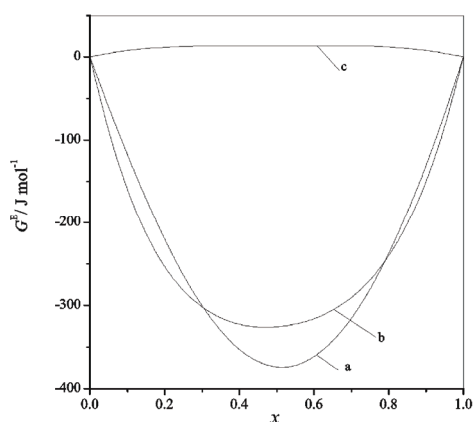


Fig. 5. Molar excess Gibbs energies for dichloroalkane ($1-x$) + cyclohexanone($1-x$) mixtures at $T = 318.15$ K (a – 1,2-dichloroethane; b – 1,3-dichloropropane; c – 1,4-dichlorobutane).

actions between the Cl groups of the chloroalkanes and the CO group of the cycloketone (Fig. 5), similarly to linear ketone + α,ω -dichloroalkane mixtures.³¹ This behaviour agrees with that expected from the proximity effect: the more the two Cl groups are separated, the less they influence each other and the behaviour of these systems is the same of systems with 1-chloroalkane, for which both G^E and H^E are positive.^{32,33}

CONCLUSIONS

The experimental vapour–liquid equilibrium data for cyclohexanone + dichloroalkane binary mixtures at temperatures from 298.15 to 318.15 K are reported. For the mixture 1,3-dichloropropane + cyclohexanone, the deviations from the Raoult Law are negative, while the mixture 1,4-dichlorobutane + cyclohexanone shows small positive deviations from this Law.

The vapour pressures vs. liquid phase composition data were used to calculate the activity coefficients of the two components, and the excess molar Gibbs energies G^E for the mixtures, using the Barker method. The G^E values were represented by the well-known Redlich–Kister, Wilson, NRTL and UNIQUAC Equations. No significant difference between the G^E values obtained with these equations was observed.

The calculated excess Gibbs energy G^E increases with increasing temperature for the first mixture and decreases with increasing temperature, for the second mixture.

NOMENCLATURE

A_i	– Redlich–Kister parameters
A_{12}, A_{21}	– Binary parameters of the Wilson, NRTL and UNIQUAC Equations
G^E	– Excess Gibbs free energy
H^E	– Excess enthalpy
m	– Number of equation parameters
$n_D(298.15\text{ K})$	– Refractive index at 298.15 K
N	– Total number of experimental points for one isotherm
p	– Vapour pressure
Q	– Objective function in Eq. (2)
T	– Thermodynamic temperature
x_i	– Liquid-phase mole fraction
y	– Vapour-phase mole fraction
<i>Subscripts</i>	
calc	– calculated
exp	– experimental
j	– j^{th} experimental point
<i>Greek letters</i>	
α	– NRTL parameter
$\rho(298.15\text{ K})$	– Liquid density at 298.15 K

σ – Standard deviation

ИЗВОД

ИЗОТЕРМСКА РАВНОТЕЖА ПАРА–ТЕЧНОСТ БИНАРНИХ СМЕША
ЦИКЛОХЕКСАНОН + ДИХЛОРАЛКАН У ТЕМПЕРАТУРНОМ ИНТЕРВАЛУ
ОД 298,15 ДО 318,15 К

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Извршена су мерења напона пара бинарних смеша циклохексанон + дихлоралкан (1,3-дихлорпропан, 1,4-дихлорбутан) у температурном интервалу од 298,15 до 318,15 К. Коefицијенти активности компонената и допунске моларне Gibbs-ове енергије (G^E) смеша, израчунати су Баркером методом коришћењем експерименталних података зависности напона паре од састава течне фазе. G^E вредности су израчунаване и помоћу Redlich–Kister-ове, Wilson-ове, NRTL и UNIQUAC једначина, при чему је неидеалност гасовите фазе одређена преко другог виријалног коefицијената. Нису добијене значајне разлике при прорачуну G^E помоћу различитих једначина.

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