J. Chem. Eng. Data **2003,** *48,* 535−540

Vapor−Liquid Equilibria and Excess Molar Enthalpies for *N*-Methyl-2-pyrrolidone with Chloroethanes and Chloroethenes

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Precise temperature−composition ($T-x$) data and excess molar enthalpies (H_m^E) of binary mixtures of *N*-methyl-2-pyrrolidone (NMP) with chloroethanes and ethenes were measured by using an ebulliometer and a solution calorimeter. The chloroethanes are 1,2-dichloroethane, 1,1,1-trichloroethane, and 1,1,2,2-tetrachloroethane. The chloroethenes are trichloroethene and tetrachloroethene. The $T-x$ data were used to compute vapor-phase composition and activity coefficients with the Wilson model. The experimental (H_m^E) values are negative for four binary systems over the entire range of composition. An inversion in sign from negative to positive for the system *N*-methyl-2-pyrrolidone + tetrachloroethene was observed.

Introduction

Separation of mixtures with components that are similar in volatility by simple distillation is difficult or even impossible, if the mixtures exhibit azeotropic points. To overcome this problem, solvents are added; volatile additives are used to form a low boiling azeotrope with one of the components or ternary azeotropes for the separation by azeotropic distillation. Selective solvents are required in extractive distillation processes. The task of the selective solvent is a strong alteration of the separation factor for the compounds to be separated. NMP is a solvent that is often used for separation processes in the petrochemical, chemical, and gas-processing industries because of its selective affinity for aromatics, unsaturated hydrocarbons, and sulfur or oxygen components and its unique physical and chemical properties, such as high polarity, low volatility, miscibility with water and organic solvents, and thermal and chemical stability. Several authors[1−4] have studied the optimization and design of appropriate solvents to be used as separating agents for extractive distillation and liquid−liquid extraction. Measurements on the systems *N*-methyl-2-pyrrolidone + benzene have been reported by Zaretskii et al.[5] and Gupta et al.[6] at 105 kPa, by Aim[7] at 287 K and 283 K, by Fabries et al.[8] at 293 K and 363 K, by Gierycz et al.[9] at 333 K and 354 K, and by Dojcansky et al.[10] at 343 K. Measurements on *N*-methyl-2-pyrrolidone + toluene have been reported by Zaretskii et al.[11] at 101 kPa and by Gupta and Rawat[12] at 101 kPa. Measurements on *N*-methyl-2-pyrrolidone + heptane have been reported by Gaube et al.[13] at 298 K and Fabries et al.[8] at 293 K and 371 K.

In this work, experimental isobaric 95.3 kPa (715 mmHg) vapor−liquid equilibria (VLE) of the binary systems formed by NMP with chloroethanes and chloroethenes are reported. Five binary systems have been experimentally determined: NMP with 1,2-dichloroethane, 1,1,1-trichloroethane, 1,1,2,2-tetrachloroethane, trichloroethene, and tetrachloroethene. Experimental excess molar enthalpies (H_m^E) for the above five binary systems of *N*-methyl-2-pyrrolidone at the temperature 298.15 K are presented here. The behavior of H_m^E is described in terms of specific group interactions. Koldobskaya *et al.*[14] reported values for (*N*-methyl-2-pyrrolidone + *N*,*N*-dimethylformamide + 1-hexyne) at low alkyne composition. However, no excess molar enthalpies or isobaric vapor−liquid equilibria have been reported for the above binary liquid mixtures.

Table 1. Densities (ρ) at 30 °C and Boiling Points (t) of the Pure Components at the Pressure 101.3 kPa (760 mmHg)

	ρ/g·cm^{-3}		t/°C	
component	expt	lit.	expt[d]	lit.
N-methyl-2-pyrrolidone	1.0234	1.023 70[a]	202.3	202.0[c]
1,2-dichloroethane	1.2383	1.238 43[b]	83.8	83.4[c]
1,1,1-trichloroethane	1.32094	1.320 94[b]	74.2	74.0[c]
1,1,2,2-tetrachloroethane	1.57858	1.578 57[b]	146.2	146.2[c]
trichloroethene	1.4512	1.451 39[b]	87.1	87.2[c]
tetrachloroethene	1.6064	1.606 41[b]	121.0	121.2[c]

[a] Reference 21. [b] Reference 15. [c] Reference 22. [d] Measured at 101.3 kPa (760 mmHg).

Experimental Section

All of the chemicals used were of analytical grade. *N*-Methyl-2-pyrrolidone (Merck, >99%) was distilled at low pressure and stored over freshly activated molecular sieves of type 3A (Union Carbide). 1,2-Dichloroethane, 1,1,1-trichloroethane, 1,1,2,2-tetrachloroethane, trichloroethene, and tetrachloroethene were purified by the methods described by Chandrasekhar *et al.*[15] The purities of the compounds were checked by measuring the densities with a bicapillary-type pycnometer of 12 cm^3 capacity that offers an accuracy of 2 parts in 10^5 and found to be in good agreement with literature values. Boiling points were measured using a Swietoslawski-type ebulliometer, giving an accuracy of ±0.2 °C. Boiling points corrected to 1 atm pressure and densities determined at 30 °C are presented in Table 1, along with the litereture values.[22]

The vapor−liquid equilibrium measurements were studied in a Swietoslawski-type ebulliometer[16] connected to a vacuum system. Pressures were measured using a mercury manometer which gave an accuracy of ±1 mmHg. The

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10.1021/je020065c CCC: $25.00

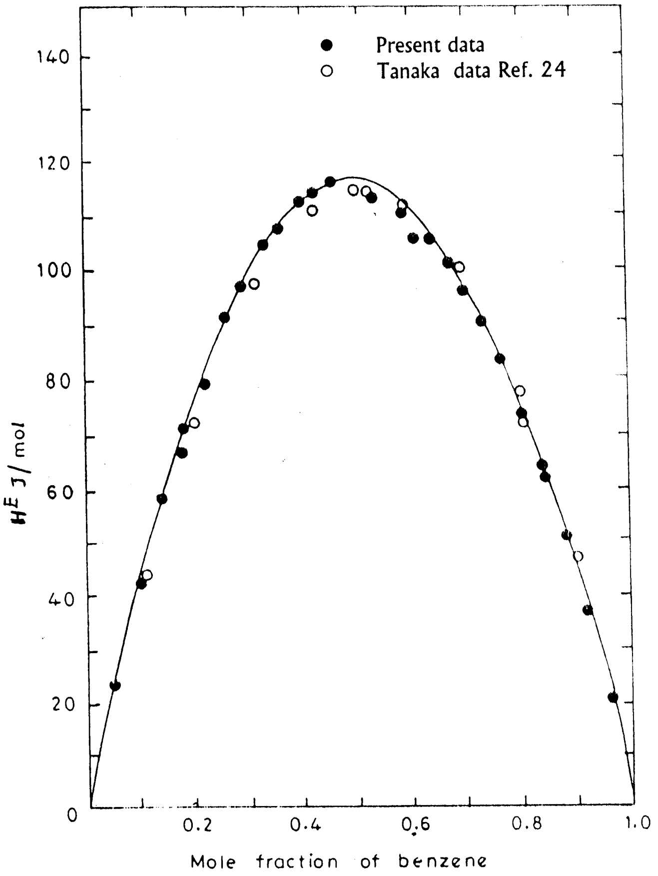

Figure 1. Heats of mixing for benzene + carbon tetrachloride.

Table 2. Wilson Parameters with Standard Deviation σ(*t*) at 95.3 kPa (715 mmHg)

N-methyl-2-pyrrolidone (B) +	Λ_{AB}	Λ_{BA}	σ
1,2-dichloroethane (A)	−432.36	−8.19	0.1
1,1,1-trichloroethane (A)	240.16	−346.77	0.1
1,1,2,2-tetrachloroethane (A)	−506.59	478.62	0.1
trichloroethene (A)	−148.59	−190.00	0.1
tetrachloroethene (A)	−314.84	1585.80	0.1

Table 3. Antoine Constants[a] of Pure Components

component	*A*	*B*	*C*
N-methyl-2-pyrrolidone	8.2910	2572.70	273.15
1.2-dichloroethane	7.0240	1271.02	222.93
1,1,1-trichloroethane	6.9483	1217.00	225.00
1,1,2,2-tetrachloroethane	6.9824	1465.10	211.00
trichloroethene	7.0268	1312.69	230.00
tetrachloroethene	7.0187	1415.23	221.00

[a] cgs units.

temperatures were measured using a PT 100 thermometer with an accuracy of ±0.1 °C. The detailed experimental procedures have been described in the literature.[16,17]

The boiling points of mixtures of *N*-methyl-2-pyrrolidone with 1,2-dichloroethane, 1,1,1-trichloroethane, 1,1,2,2-tetrachloroethane, trichloroethene, and tetrachloroethene were determined at 95.3 kPa (715 mmHg) over the entire range of compositions; mixtures of different composition were prepared by measuring the mass of the pure compounds.

A new Parr [45] solution calorimeter was used in the measurements of H^E of binary liquid mixtures. The detailed procedures and apparatus used have been described in our earlier publications.[18−20] In the measurement of excess enthalpies, a known mass of 100 mL of component A is charged into the Dewar vessel and 20 mL of component B is weighed in a sealed glass rotating cell. Both the components are kept in thermal contact with each other in the Dewar mixing chamber at 298.15 K. After attaining thermal equilibrium, component B in the rotating cell is mixed with component A in the Dewar vessel by pushing the glass rod. The temperature change accompanying the mixing process is sensed by the thermistor and recorded on the strip chart recorder. These experiments were continued until the concentrations reached (50 to 60)%.

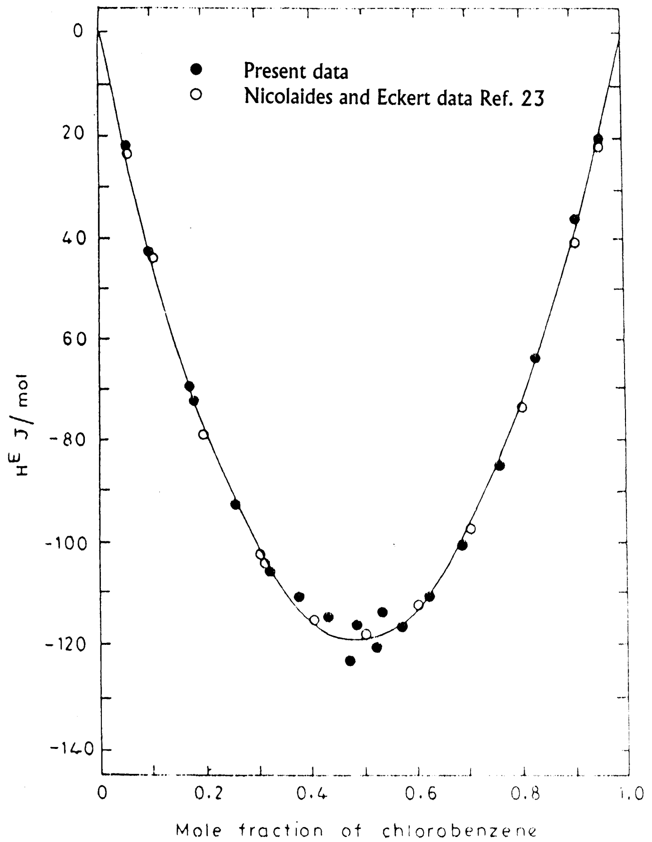

Figure 2. Heats of mixing for chlorobenzene + toluene.

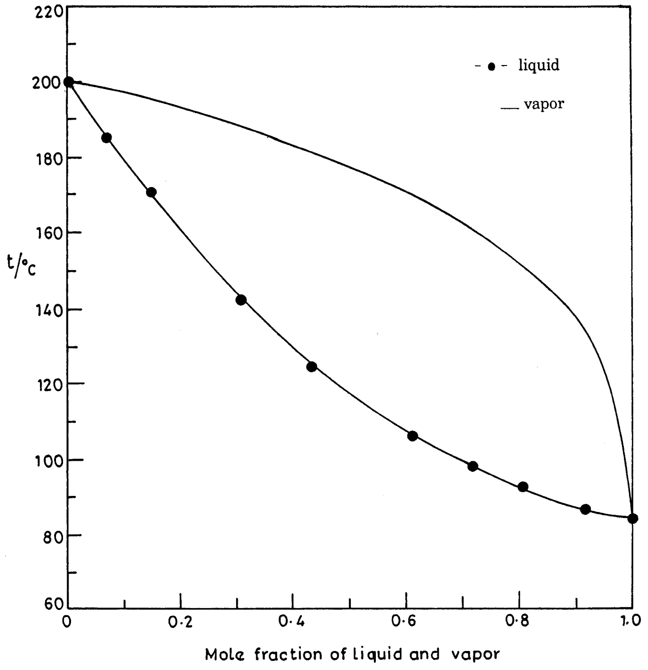

Figure 3. 1,2-Dichloroethane + *N*-methyl-2-pyrrolidone.

Table 4. Vapor−Liquid Equilibrium Data for Binary Systems with *N*-Methyl-2-pyrrolidone (NMP) at 95.3 kPa (715 mmHg): Liquid-Phase Mole Fraction (X_A), Vapor-Phase Mole Fraction y_A, Temperature *t*, and Activity Coefficients γ_i

		t/°C				
x_A	y_A	expt	cal[a]	γ_A	γ_B	Δt[b]/°C
1,2-Dichloroethane (A) + *N*-Methyl-2-pyrrolidone (B)						
0.0000		200.1	200.1	0.3927	1.0000	0.0
0.0512	0.2619	190.5	190.8	0.4085	0.9985	0.3
0.1082	0.4835	180.5	180.5	0.4273	0.9927	0.0
0.2667	0.8196	154.2	154.0	0.4896	0.9488	−0.2
0.4000	0.9287	135.1	135.1	0.5573	0.8704	0.0
0.4590	0.9537	127.7	127.7	0.5927	0.8208	0.0
0.7752	0.9974	95.8	95.9	0.8444	0.3931	0.1
0.8898	0.9995	87.4	87.4	0.9464	0.2067	0.0
0.9225	0.9998	85.4	85.4	0.9704	0.1593	0.0
1.0000	1.0000	81.9	81.9	1.0000	0.0690	0.0
1,1,1-Trichloroethane (A) + *N*-Methyl-2-pyrrolidone (B)						
0.0000		200.1	200.1	0.6248	1.0000	0.0
0.1005	0.6595	166.2	166.2	0.6818	0.9920	0.0
0.1937	0.8633	142.2	142.2	0.7406	0.9706	0.0
0.3592	0.9641	114.0	113.9	0.8423	0.9085	−0.1
0.4683	0.9829	101.7	101.7	0.8972	0.8593	0.0
0.6059	0.9928	90.5	90.6	0.9483	0.7958	0.1
0.7193	0.9965	83.7	83.7	0.9758	0.7455	0.0
0.8367	0.9985	78.2	78.2	0.9925	0.6968	0.0
0.9275	0.9995	74.7	74.7	0.9986	0.6620	0.0
1.0000	1.0000	72.3	72.3	1.0000	0.6359	0.0
1,1,2,2-Tetrachloroethane (A) + *N*-Methyl-2-pyrrolidone (B)						
0.0000		200.1	200.1	0.6044	1.0000	0.0
0.0755	0.1607	196.2	196.5	0.6242	0.9985	0.3
0.1319	0.2717	193.5	193.7	0.6399	0.9951	0.2
0.3474	0.6197	182.2	182.0	0.7094	0.9564	−0.2
0.5776	0.8652	168.4	168.3	0.805	0.8389	−0.1
0.6458	0.9016	164.1	164.0	0.8384	0.7797	−0.1
0.7848	0.9644	155.1	155.4	0.9127	0.6105	0.3
0.9162	0.9932	147.7	147.7	0.9796	0.3858	0.0
0.9623	0.9978	145.4	145.5	0.9951	0.2970	0.1
1.0000	1.0000	143.9	143.9	1.0000	0.2249	0.0
Trichloroethene (A) + *N*-Methyl-2-pyrrolidone (B)						
0.0000		200.1	200.1	0.4658	1.0000	0.0
0.0725	0.3780	185.4	185.6	0.5052	0.9959	0.2
0.1507	0.6439	170.0	169.9	0.5501	0.9819	−0.1
0.3078	0.8878	142.5	142.4	0.6495	0.9212	−0.1
0.4447	0.9582	123.8	123.8	0.7431	0.8344	0.0
0.6156	0.9878	106.5	106.6	0.8572	0.6921	0.1
0.7193	0.9945	98.6	98.6	0.9173	0.5944	0.0
0.8102	0.9975	93.0	93.0	0.9596	0.5065	0.0
0.9275	0.9994	87.3	87.3	0.9936	0.3950	0.0
1.0000	1.0000	85.1	85.1	1.0000	0.3298	0.0
Tetrachloroethene (A) + *N*-Methyl-2-pyrrolidone (B)						
0.0000		200.1	200.1	1.2181	1.0000	0.0
0.1002	0.5034	178.4	178.6	1.1969	0.9995	0.2
0.1899	0.7053	164.7	164.8	1.1863	0.9980	0.1
0.4386	0.9091	140.4	140.3	1.1734	0.9890	−0.1
0.4839	0.9252	137.2	137.2	1.1711	0.9876	0.0
0.6251	0.9591	129.2	129.1	1.1579	0.9916	−0.1
0.7500	0.9766	123.9	123.9	1.1319	1.0350	0.0
0.8869	0.9881	120.1	120.1	1.0740	1.3382	0.0
0.9313	0.9908	119.4	119.4	1.0458	1.7586	0.0
1.0000	1.0000	118.8	118.8	1.0000	1.4430	0.0

[a] From the Wilson equation. [b] $\Delta t = t_{cal} - t_{expt}$.

This procedure was repeated by taking component A as component B and component B as component A. The excess molar enthalpy was measured, knowing the energy equivalent of the system and the equation

$$H_M^E = (W_A C_{pA} + W_B C_{pB} + E)\Delta T/n \qquad (1)$$

where W_i is the weight of liquid *i* (g), C_{pi} is the heat capacity of liquid *i* (J·g^{-1}·K^{-1}), *E* is the energy equivalent of the calorimeter (J·K^{-1}), ΔT is the temperature change (K), and *n* is the number of moles of mixture. The reliability of the apparatus and the method was established by measuring H^E values of standard systems, that is, benzene with carbon tetrachloride and chlorobenzene with toluene, throughout the concentration range at 298.15 K. Mole fraction versus excess enthalpy curves of the above standard systems are presented graphically in Figures 1 and 2. The agreement between the measured values and literature data[23,24] is within 2% and satisfactory.

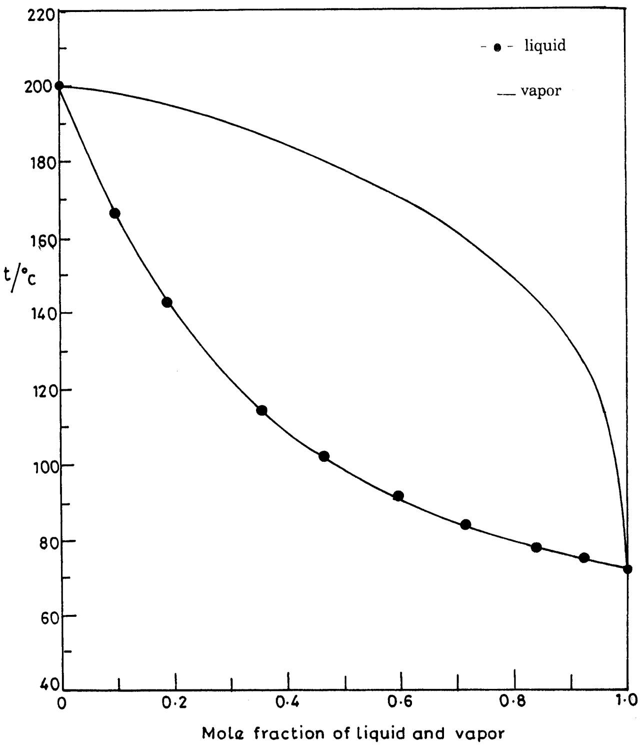

Figure 4. 1,1,1-Trichloroethane + *N*-methyl-2-pyrrolidone.

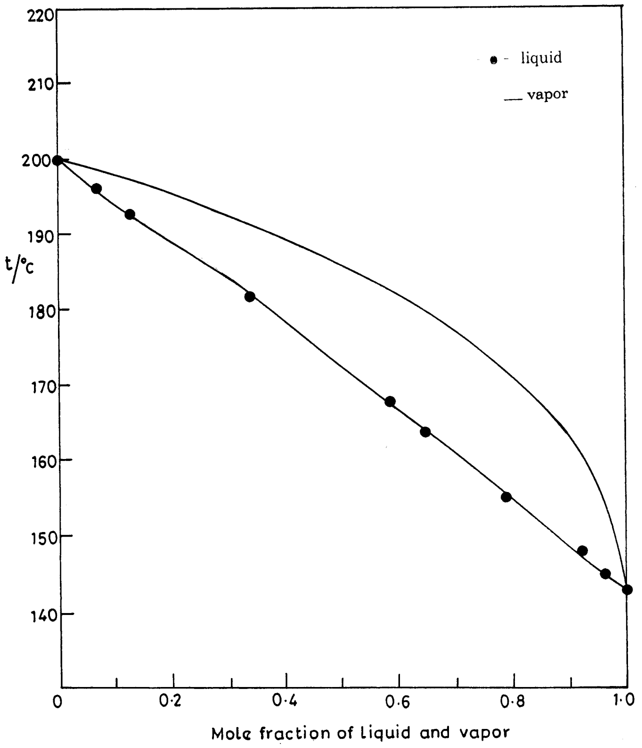

Figure 5. 1,1,2,2-Tetrachloroethane + *N*-methyl-2-pyrrolidone.

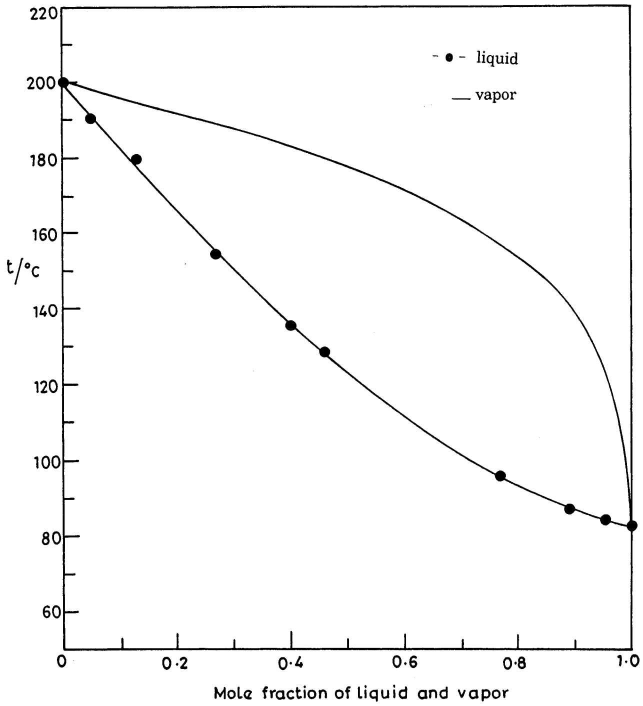

Figure 6. Trichloroethene + *N*-methyl-2-pyrrolidone.

Results and Discussion

The boiling points t of the binary systems of known composition x have been measured using an ebulliometer technique over the entire range of composition at 95.3 kPa (715 mmHg). The $T-x$ measurements were used to compute vapor-phase composition, activity coefficients, and excess Gibbs free energies on the basis of the Wilson model. The Wilson equation was chosen to represent the composition and temperature dependence of liquid-phase activity coefficients γ_A and γ_B. γ_A and γ_B can be calculated using eqs 2 and 3.

$$l_n\gamma_A = -l_n[x_A + \Lambda_{AB}x_B] + x_B\left[\frac{\Lambda_{AB}}{x_A + x_B\Lambda_{AB}} - \frac{\Lambda_{BA}}{x_B + x_A\Lambda_{BA}}\right] \tag{2}$$

$$l_n\gamma_B = -l_n[x_B + \Lambda_{BA}x_A] + x_A\left[\frac{\Lambda_{BA}}{x_B + x_A\Lambda_{BA}} - \frac{\Lambda_{AB}}{x_A + x_B\Lambda_{AB}}\right] \tag{3}$$

Λ_{AB} and Λ_{BA} are Wilson parameters which are temperature dependent. The temperature dependence of these parameters is given by

$$\Lambda_{AB} = \frac{V_B^{\,l}}{V_A^{\,l}} \exp\left[\frac{\Lambda_{AB} - \Lambda_{AA}}{RT}\right] \tag{4}$$

$$\Lambda_{BA} = \frac{V_A^{\,l}}{V_B^{\,l}} \exp\left[\frac{\Lambda_{AB} - \Lambda_{BB}}{RT}\right] \tag{5}$$

where $V_A^{\,l}$ and $V_B^{\,l}$ are the liquid molar volumes at temperature T of the pure liquids A and B. $[\Lambda_{AB} - \Lambda_{AA}]$ and $[\Lambda_{AB} - \Lambda_{BB}]$ are taken as constants over a limited temperature range. For a wide temperature range these also depend on temperature. Λ's are the energies of interaction between the molecules designated in the subscripts. The optimum Wilson constants $((\Lambda_{AB} - \Lambda_{AA})/R)$ and $((\Lambda_{AB} - \Lambda_{BB})/R)$ are obtained by minimizing the objective function, ø,

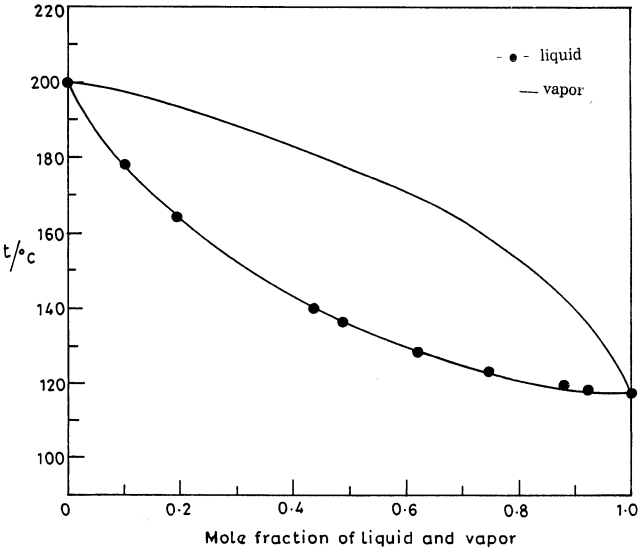

Figure 7. Tetrachloroethene + *N*-methyl-2-pyrrolidone.

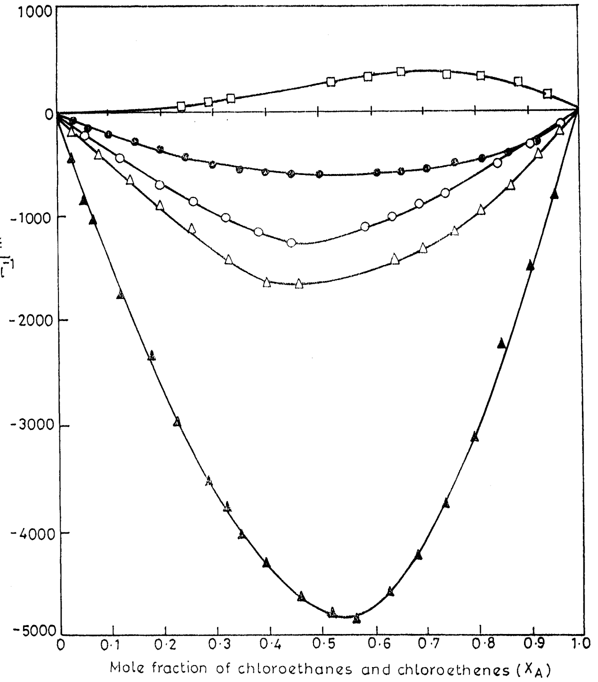

Figure 8. Excess enthalpies (H^E) as a function of mole fraction (X_A) for chloroethanes and chloroethenes (A) with *N*-methyl-2-pyrrolidone (B) at 298.15 K: 1,2-dichloroethane (○); 1,1,1-trichloroethane (●); 1,1,2,2-tetrachloroethane (▲); trichloroethene (△); tetrachloroethene (□).

$$\phi = \sum\left[\frac{P_{cal} - 1.0}{P_{exp}}\right]^2 \tag{6}$$

using the Nelder Mead optimization technique, where P_{cal} is given by

$$P_{cal} = \sum x_i\gamma_i P_i^{\,s} \tag{7}$$

The vapor composition was calculated using the following

Table 5. Experimental Excess Molar Enthalpies of Chloroethanes and Chloroethenes (A) + *N*-Methyl-2-pyrrolidone (B) at 298.15 K

X_A	H^E/J·mol^{-1}	X_A	H^E/J·mol^{-1}
1,2-Dichloroethane (A) + *N*-Methyl-2-pyrrolidone (B)			
0.0658	−236.5	0.6421	−1027.3
0.1316	−458.6	0.6925	−908.3
0.1959	−664.9	0.7469	−811.8
0.2633	−858.7	0.8024	−678.5
0.3310	−1036.6	0.8560	−513.6
0.3947	−1178.4	0.9111	−335.5
0.4525	−1257.7	0.9608	−168.5
0.5920	−1115.3		
1,1,1-Trichloroethane (A) + *N*-Methyl-2-pyrrolidone (B)			
0.0303	−60.5	0.5576	−571.3
0.0662	−135.0	0.6104	−589.8
0.1039	−208.0	0.6634	−588.2
0.1525	−293.9	0.7153	−563.8
0.2006	−374.5	0.7694	−510.0
0.2534	−442.9	0.8172	−490.5
0.3046	−509.0	0.8714	−432.9
0.3555	−554.9	0.9228	−327.7
0.4018	−582.3	0.9649	−182.7
0.4556	−583.1		
0.5054	−581.6		
1,1,2,2-Tetrachloroethane (A) + *N*-Methyl-2-pyrrolidone (B)			
0.0331	−473.6	0.4675	−4567.0
0.0582	−859.6	0.5226	−4749.8
0.0765	−1071.3	0.5688	−4841.4
0.1279	−1783.4	0.5953	−4853.1
0.1292	−1727.0	0.6265	−4547.2
0.1796	−2322.6	0.6815	−4186.7
0.2318	−2945.0	0.7384	−3722.1
0.2344	−2908.0	0.7933	−3104.7
0.2880	−3505.3	0.8536	−2231.8
0.3219	−3736.2	0.9099	−1480.3
0.3400	−4027.8	0.9574	−750.2
0.4028	−4233.1		
Trichloroethene (A) + *N*-Methyl-2-pyrrolidone (B)			
0.0381	−190.0	0.7095	−1316.4
0.0886	−443.0	0.7643	−1170.6
0.1426	−662.9	0.8186	−962.6
0.2005	−902.6	0.8730	−734.9
0.2675	−1153.8	0.9254	−469.4
0.3373	−1433.1	0.9678	−211.0
0.4001	−1647.3		
0.6547	−1434.3		
Tetrachloroethene (A) + *N*-Methyl-2-pyrrolidone (B)			
0.0390	−10.0	0.6006	296.7
0.0887	−12.2	0.6688	336.1
0.1357	−5.0	0.7420	324.2
0.2415	29.3	0.8130	295.3
0.2932	57.4	0.8822	236.7
0.3454	89.5	0.9441	142.9
0.5360	260.7		

equation

$$Y_A = \frac{x_A \gamma_A p_A^{\,s}}{P_{cal}} \qquad (8)$$

The Wilson parameters so obtained, with standard deviations, are presented in Table 2.

Since the prediction of the vapor−liquid equilibrium data involves measurement of the vapor pressures of the pure compounds, these were measured for *N*-methyl-2-pyrrolidone, 1,2-dichloroethane, 1,1,1-trichloroethane, 1,1,2,2-tetrachloroethane, trichloroethene, and tetrachloroethene. The Antoine constants for the pure components were obtained by a nonlinear regression analysis of the experimental data and are presented in Table 3. The measured boiling points, calculated boiling points from the Wilson equation, calculated vapor composition, activity coefficients, and Δt values are presented in Table 4.

The temperature versus mole fraction curves are presented graphically in Figures 3−7. The activity coefficients presented in Table 4 reveal that all the systems are nonideal.

Experimental excess molar enthalpies at 298.15 K of the five binary mixtures of *N*-methyl-2-pyrrolidone with chloroethanes and chloroethenes are reported in Table 4 and are graphically presented in Figure 8 in the form of H^E versus the mole fraction (x_A) of chloroethanes and -ethenes. The experimental H^E values may be represented by

$$E^H = x_A x_B \sum_i h_1 (x_A - x_B)^i \qquad (9)$$

where x_A and x_B are mole fraction of chloroethanes and ethenes and *N*-methyl-2-pyrrolidone. The h_1 binary parameters have been evaluated by the method of least squares and are listed in Table 6 along with the percentage standard deviation. The values of the percentage standard deviation are computed from the relation

$$\sigma(H^E)/\% = \left[\frac{\Sigma(((H^E_{exp} - H^E_{cal})/(H^E_{exp})) \times 100)^2}{n - p}\right]^{1/2} \qquad (10)$$

where n and p are the number of results and parameters, respectively. The data included in Table 5 show that the H^E values are negative (exothermic) for four binary systems of NMP with 1,2-dichloroethane, 1,1,1-trichloroethane, 1,1,2,2-tetrachloroethane, and trichloroethene at 298.15 K over the whole range of composition. An inversion in sign from negative to positive is observed with decreasing *N*-methyl-2-pyrrolidone concentration for the systems of *N*-methyl-2-pyrrolidone + tetrachloroethane. The exothermic nature of these results is a reflection of the dominance of the strong association between *N*-methyl-2-pyrrolidone and chloroethanes. The strongest association was between *N*-methyl-2-pyrrolidone and 1,1,2,2-tetrachloroethane and may be due to the stabilization of NMP by the ring structure, thereby increasing the electron donor capacity of the oxygen atom. The association effect decreases as substitution on the ethane molecule decreases. If this were correct, then the least exothermic effect should be occur for (NMP + 1,2-dichloroethane). In our work 1,1,1-trichloroethane shows the least exothermic effect.

Table 6. Estimated Parameters of Eq 8 and Percent Standard Deviation $\sigma(H^E)$ for the Mixtures of *N*-Methyl-2-pyrrolidone with Chloroethanes and Chloroethenes at 298.15 K

N-methyl-2-pyrrolidone (B) +	a_0 / J·mol^{-1}	a_1 / J·mol^{-1}	a_2 / J·mol^{-1}	a_3 / J·mol^{-1}	a_4 / J·mol^{-1}	a_5 / J·mol^{-1}	$\sigma(H^E)$ / %
1,2-dichloroethane (A)	−4871.4	467.9	2605.0	−995.8	−2048.6		2.4
1,1,1-trichloroethane (A)	−2380.0	35.7	−659.3	−2066.7	−1050.1		2.1
1,1,2,2-tetrachloroethane (A)	−19035	−298.3	5661.1	1777.7	−3346.0		2.3
trichloroethene (A)	−6657.8	330.6	2230.8	−3659.6	−1800.7		2.8
tetrachloroethene (A)	917.3	1750.8	1.8	−1046.3	523.2	1164.2	4.6

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Received for review April 8, 2002. Accepted February 15, 2003.

JE020065C