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Excess Properties of Some Methanol + Amide Systems Proposed as Working Fluids for Absorption Machines

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Excess enthalpies and excess volumes of the binary systems methanol + *N*-formylmorpholine, methanol + *N*-methyl-2-pyrrolidine, and methanol + 1,3-dimethyl-3,4,5,6-tetrahydro-2(1*H*)-pyrimidinone at 298.15 K and atmospheric pressure have been measured. Excess enthalpies H^E were measured in a Calvet microcalorimeter and excess volume V^E in a vibrating-tube densimeter. All the V^E values were negative, and the H^E values were also negative except those of the methanol + *N*-formylmorpholine system. The results are discussed qualitatively in terms of molecular interactions. A version of the UNIFAC group contribution model has been used to compare the predicted and experimental H^E results.

Introduction

This work continues research (López et al., 1995, 1997) on the thermophysical properties of new working fluids for absorption machines, mainly addressed to the characterization of new absorbent−refrigerant pairs that could improve the cycle performance.

In the present work we have chosen some amides, *N*-formylmorpholine (NFM), *N*-methyl-2-pyrrolidine (NMP), and 1,3-dimethyl-3,4,5,6-tetrahydro-2(1*H*)-pyrimidinone also called 1,3-dimethylpropyleneurea (DMPU), as absorbents (Nowaczyk and Steimle, 1992), to study different thermodynamic properties of the mixtures formed by these compounds with methanol (as refrigerant). We report the excess molar enthalpies and excess molar volumes at 298.15 K and atmospheric pressure of these mixtures.

An absorption machine operates with a working fluid pair. One of these fluids is the refrigerant and consists of a mixture or pure highly volatile liquid. The other one is the absorbent, which must be practically involatile. Both fluids must be thermally and chemically stable, and the difference between their vaporization temperatures must be greater than 150 K. Other chief requirements are as follows: large vaporization enthalpy, low viscosity, high thermal conductivity, small excess enthalpy, low specific heat capacity, and negative deviations from Raoult's law. In particular, since these last requirements depend on the formation of a hydrogen bond between the refrigerant and the absorbent, it is advisable to choose as refrigerant a fluid with small polar molecules that can easily come near molecules of the absorbent.

Methanol, among other alcohols, has been proposed as a refrigerant (Moncada Lo Giudice and Cotana, 1994) because it presents several important advantages such as enthalpy of vaporization (1248 kJ/kg) and its ability to evaporate under temperatures less than 273.15 K (the triple point of methanol is at 206 K).

Amides are excellent solvents both for organic and inorganic compounds. They have properties that make them of particular interest to both research and production. NFM and NMP are widely used in industrial processes such as absorption columns, in the extraction of some monomers in the petrochemical industry, and in gas wash processes for the removal of desirable or undesirable components, etc. These amides are highly polar solvents with high electron density surrounding their acceptor atoms, which implies that effective hydrogen bonds are formed by the H atom of the methanol and the O amide atom. The excess enthalpies and the excess volumes give some information about these interactions. Also, the calorimetric and volumetric properties are necessary in the calculation of the performance of the absorption cycles.

Table 1. Densities of Pure Compounds at 298.15 K and Atmospheric Pressure

liquid	density/(g·cm^{-3}) exptl	density/(g·cm^{-3}) lit.
methanol	0.78658	0.78664[a] 0.78637[b]
NFM	1.14645	1.14648[c] 1.1463[d]
NMP	1.02811	1.02812[e] 1.02821[f] 1.0279[g]
DMPU	1.05953	1.059220[h] 1.0593[i] 1.0596[j]

[a] TRC, 1996. [b] Riddick et al., 1986. [c] Awwad, 1988. [d] Al-Azzawl and Allos, 1992. [e] Wu et al., 1985. [f] Uosaki et al., 1990. [g] Liu et al., 1996. [h] Lemos and Maestre, 1995. [i] Kneisl and Zondlo, 1987. [j] Stroka et al., 1990.

Additionally, H^E values can be used to verify the interaction parameters of the group contribution models. In this work we have applied the version of the UNIFAC model due to Gmehling et al. (1993) in order to check the goodness of the predictions of excess enthalpies, because it is the only model with available parameters in the literature for this type of mixtures.

Experimental Section

Materials. Methanol (Aldrich, mole fraction >0.999), *N*-formylmorpholine (Aldrich, mole fraction >0.99), *N*-meth-

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Table 2. Excess Molar Enthalpies, H^E, at 298.15 K of the Methanol + Amide Systems

x	$H^E/(J\cdot mol^{-1})$	x	$H^E/(J\cdot mol^{-1})$	x	$H^E/(J\cdot mol^{-1})$	x	$H^E/(J\cdot mol^{-1})$
			xMethanol + $(1 - x)$NMP				
0.0707	−142.4	0.3725	−598.5	0.5676	−649.4	0.8169	−400.5
0.0771	−158.7	0.4022	−631.8	0.5878	−637.9	0.8496	−331.7
0.1539	−296.6	0.4666	−651.7	0.6647	−592.8	0.9045	−212.3
0.1715	−342.0	0.4726	−658.1	0.7466	−498.3	0.9221	−188.2
0.2719	−488.8						
			xMethanol + $(1 - x)$NFM				
0.0910	97.1	0.3740	399.5	0.5638	550.1	0.7874	518.6
0.1361	150.3	0.4272	454.1	0.5764	547.7	0.8114	488.6
0.2085	222.1	0.4783	493.7	0.6113	568.1	0.8593	415.1
0.2675	299.5	0.5117	526.2	0.6790	565.4	0.9170	292.4
0.2685	291.3	0.5371	527.8	0.6952	555.5	0.9400	222.2
0.3569	393.4	0.5418	536.0	0.7133	554.9		
			xMethanol + $(1 - x)$DMPU				
0.0887	−299.8	0.3143	−906.7	0.5769	−1129.8	0.8382	−632.2
0.1328	−451.8	0.3855	−1043.2	0.6614	−1062.2	0.8452	−611.5
0.2266	−714.3	0.4437	−1112.7	0.6938	−998.3	0.8766	−480.5
0.2293	−719.2	0.5369	−1134.5	0.7232	−920.4	0.9501	−215.1
0.2686	−817.8	0.5569	−1144.7	0.7944	−745.9	0.9620	−166.1

yl-2-pyrrolidinone (Aldrich, mole fraction >0.99), and 1,3-dimethyl-3,4,5,6-tetrahydro-2(1*H*)-pyrimidinone (Fluka, mole fraction >0.99) were subjected to no further purification other than drying with Union Carbide 0.4 nm molecular sieves. Chemical purities were checked by gas chromatography, and they were found to be as follows: 99.99%, 99.9%, 99.3%, and 99.3% for methanol, NFM, NMP, and DMPU, respectively. In Table 1 we present the experimental densities of all the pure liquids.

Apparatus. Excess molar enthalpies were determined with a standard Calvet microcalorimeter linked to a Philips voltimeter PM2525. The microcalorimeter was calibrated electrically using a stabilized current source (EJP-30, Setaram). The experimental technique due to Paz-Andrade et al. (1972) was used to determine the enthalpy of mixing over the entire mole fraction range. The temperature is regulated to better than ±0.005 using an electronic controller. The mole fractions of the binary mixtures were determined by mass using a Mettler balance H51 (precision, 10^{-5} g). The possible error in the mole fractions is estimated to be less than $\pm 10^{-4}$. The performance of the apparatus was checked by determining at 298.15 K the H^E of hexane + cyclohexane and of water + methanol. Our results differ by less than 1% from the literature values (Marsh, 1973; Gmehling, 1992; Abello, 1973; Keller et al., 1992).

Excess molar volumes were measured using a vibrating-tube densimeter (Anton Paar DMA60/602) thermostated to within ±0.01 K in a Polyscience circulating water bath. The cell temperature was measured with an Anton Paar CKT 100 precision thermometer. The products were partially degassed before use with a Branson 2210 ultrasonic bath. Before each series of measurements the instrument was calibrated with heptane and water (Milli-Q and Milli-ρ). Binary mixtures were prepared by mass using a precision digital Sartorius 210-P balance with an accuracy of $\pm 10^{-5}$ g. To reduce the uncertainties in the mole fraction due to vaporization, airtight stoppered bottles, with a small vapor phase, were used. The possible error in the mole fractions for these types of mixtures is estimated to be less than $\pm 10^{-4}$.

Experimental Results

In Tables 2 and 3 we present the experimental values of H^E and V^E for the three binary mixtures studied in the present work. For each binary mixture, a Redlich−Kister function was used to correlate the experimental values:

$$Y^E = x(1 - x)\sum_i A_i(2x - 1)^i \qquad (1)$$

where Y^E is either $H^E/(J\cdot mol^{-1})$ or $V^E/(cm^3.mol^{-1})$.

The parameters A_i, obtained by a linear least-squares fitting procedure, are given in Tables 4 and 5 together with the standard deviations, s. The number of coefficients was determined in each case using an F-test. Figures 1 and 2 show the measured values of H^E and V^E, respectively, plotted against x, the mole fraction of methanol, together with the fitted curves.

Figure 3 shows that our experimental H^E results for methanol + NMP are in agreement with those already reported by Murakami et al. (1974), by Zivny et al. (1991), and Sitnyakovskii et al. (1991). Our H^E values present mean deviations of 1.4% and 2.8% respectively from the H^E measurements of the two first papers in which data were collected in the entire composition range. On the contrary, Sitnyakovskii et al. (1991) have presented H^E data for the methanol dilute range. Our data present a mean absolute deviation of 10.6 $J\cdot mol^{-1}$ and a percentage mean deviation of 8.0% from the Sitnyakovskii et al. results. All these H^E values are negative. On the contrary, the excess enthalpies of Joly and Mermet-Dupin (1975) are dramatically different, because they are positive. This may be due to the fact that these last authors have forgotten the negative sign in the calculations of the excess enthalpies.

For the methanol + NFM system, the H^E values in the alkanol dilute region of Sitnyakovskii et al. (1991) present a mean deviation of 7.7% from our values with a mean absolute deviation of 5.0 $J\cdot mol^{-1}$ (see Figure 3). For this system the V^E values obtained in this work present a deviation of 13% from those reported by Awwad et al. (1984) for the equimolecular mixture, being the absolute deviation of 0.16 $cm^3\cdot mol^{-1}$.

There is not unique behavior for the $H^E(x)$ curves of the three mixtures: methanol + NFM is endothermic, $H^E(x)$ being slightly asymmetrical and with the maximum in the methanol rich region. The mixtures methanol + DMPU and methanol + NMP are exothermic, and their $H^E(x)$ curves are symmetrical. All the $V^E(x)$ curves present a small asymmetry with respect to the equimolecular composition, with minima in the alcohol rich region.

Discussion

Methanol + NFM. In their pure state both components of this mixture are strongly associated through hydrogen

Table 3. Experimental Densities, ρ, at 298.15 K of the Methanol + Amide Systems

x	ρ/(g·cm^{-3})	x	ρ/(g·cm^{-3})	x	ρ/(g·cm^{-3})	x	ρ/(g·cm^{-3})
xMethanol + (1 $-x$)NMP							
0.0469	1.02387	0.3489	0.98941	0.6532	0.93050	0.9119	0.83695
0.0482	1.02374	0.3660	0.98688	0.6831	0.92244	0.9288	0.82833
0.0718	1.02162	0.4046	0.98096	0.7135	0.91367	0.9301	0.82759
0.1416	1.01463	0.4444	0.97443	0.7443	0.90421	0.9379	0.82345
0.1725	1.01139	0.4992	0.96458	0.7888	0.88910	0.9399	0.82239
0.1783	1.01080	0.5188	0.96081	0.8123	0.88044	0.9559	0.81349
0.2469	1.00289	0.5430	0.95593	0.8403	0.86941	0.9944	0.79029
0.3168	0.99389	0.6360	0.93480	0.8689	0.85718		
xMethanol + (1 $-x$)NFM							
0.0882	1.13508	0.3541	1.08950	0.5771	1.03090	0.8054	0.93296
0.1405	1.12751	0.3643	1.08729	0.5854	1.02810	0.8285	0.91970
0.1840	1.12069	0.3893	1.08182	0.6219	1.01550	0.8719	0.89210
0.2060	1.11714	0.4294	1.07241	0.6825	0.99218	0.9153	0.86098
0.2253	1.11392	0.4439	1.06888	0.6941	0.98732	0.9339	0.84622
0.2640	1.10711	0.5029	1.05335	0.7479	0.96295	0.9475	0.83499
0.2759	1.10488	0.5277	1.04622	0.7821	0.94564	0.9776	0.80828
0.3074	1.09893	0.5681	1.03375				
xMethanol + (1 $-$ x)DMPU							
0.0563	1.05527	0.4003	1.01677	0.6380	0.96831	0.8907	0.86697
0.1030	1.05128	0.4272	1.01255	0.6888	0.95362	0.9056	0.85790
0.1329	1.04859	0.4470	1.00928	0.7174	0.94438	0.9241	0.84601
0.1841	1.04360	0.4610	1.00686	0.7599	0.92914	0.9335	0.83961
0.2393	1.03775	0.5136	0.99712	0.7907	0.91680	0.9647	0.81647
0.2713	1.03409	0.5562	0.98827	0.8350	0.89675	0.9759	0.80750
0.2903	1.03180	0.5686	0.98551	0.8616	0.88322	0.9804	0.80378
0.3388	1.02556						

Table 4. Parameters A_i and Standard Deviation, s, for the Redlich–Kister Equation Corresponding to the Excess Enthalpies, H^E, for Methanol + Amide Systems

system	A_0/(J·mol^{-1})	A_1/(J·mol^{-1})	A_2/(J·mol^{-1})	A_3/(J·mol^{-1})	s
methanol + NMP	−2638.7	−188.9	358.1		5.8
methanol + NFM	2036.7	1322.6	593.6	400.9	5.3
methanol + DMPU	−4561.1	−528.6	−667.5		9.6

Table 5. Parameters A_i and Standard Deviation, s, for the Redlich–Kister Equation Corresponding to the Excess Volumes, V^E, for Methanol + Amide Systems

system	A_0/(cm^3·mol^{-1})	A_1/(cm^3·mol^{-1})	A_2/(cm^3·mol^{-1})	A_3/(cm^3·mol^{-1})	A_4/(cm^3·mol^{-1})	s
methanol + NMP	−2.298 538	−1.044 055	−0.494 960	−0.364 207		0.002
methanol + NFM	−3.086 413	−1.432 011	−0.445 138	−0.427 132	−0.697 473	0.002
methanol + DMPU	−2.937 684	−1.216 855	−0.210 889	−0.255 147	−0.753 796	0.002

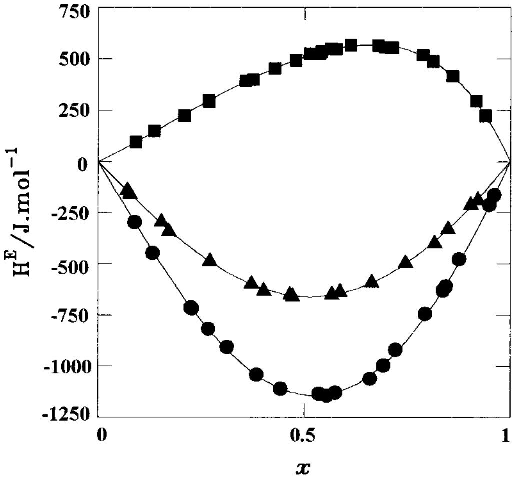

Figure 1. Experimental excess enthalpies, H^E, at 298.15 K of the methanol + amide systems: ■, $x$methanol + (1 − x)NFM; ▲, $x$methanol + (1 − x)NMP; ●, $x$methanol + (1 − x) DMPU. Solid lines: Redlich–Kister correlations.

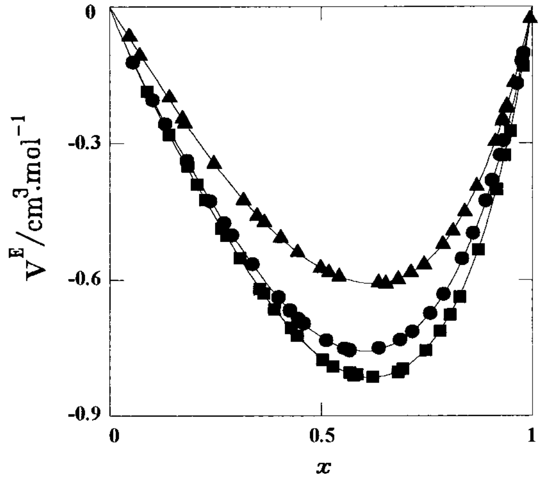

Figure 2. Experimental excess volumes, V^E, at 298.15 K of the methanol + amide systems: ■, $x$methanol + (1 − x)NFM; ▲, $x$methanol + (1 − x) NMP; ●, $x$methanol + (1 − x) DMPU. Solid lines: Redlich–Kister correlations.

bonds. Moreover, as both molecules are dipolar, dipole–dipole interactions are also expected. In addition, in the mixtures there are interactions between the hydroxyl group of the methanol and the O=C−H and the ether groups of the NFM. Therefore, the experimental endothermicity of this mixture shows that the main contribution is the breaking of the interactions in the pure liquids.

Methanol + NMP and Methanol + DMPU. For both mixtures the excess molar enthalpies are negative, indicating that the contribution due to complex formation between

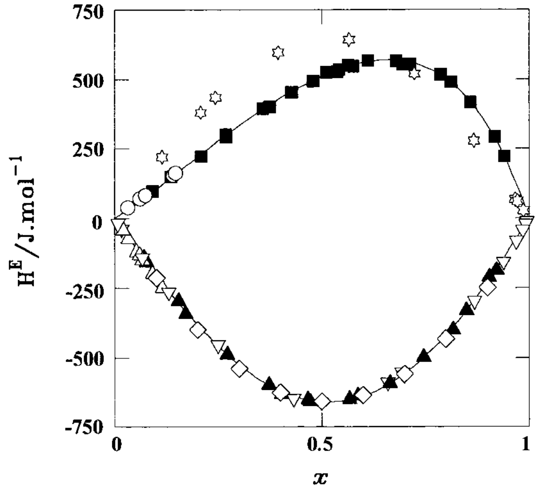

Figure 3. Excess enthalpies at 298.15 K of the system *x*methanol + (1 − *x*)NMP [▲, this work; ▽, Murakami et al. (1974); □, Sitnyakovskii et al. (1991); ◇, Zivny et al. (1991); ✡, Joly and Mermet-Dupin (1975)] and H^E of the system *x* methanol + (1 − *x*) NFM [■, this work; ○, Sitnyakovskii et al. (1991)].

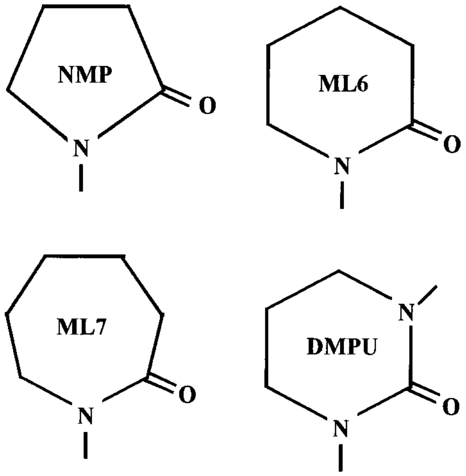

Figure 4. Molecular structures of different lactams.

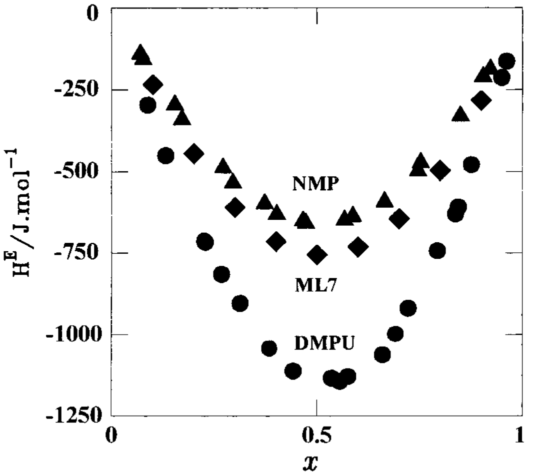

Figure 5. Excess enthalpies of system methanol + lactam: ●, DMPU; ▲, NMP; ♦, ML7 (Zivny et al., 1991).

Table 6. $H^E(x=0.5)$ at 298.15 K of the Systems Methanol + Amide, Predicted by the Gmehling Version of the UNIFAC Model

	H^E/(J·mol^{-1})	
system	expt	Gmehling
methanol + NFM	509	338
methanol + NMP	−660	−2087
methanol + DMPU	−1140	−2870

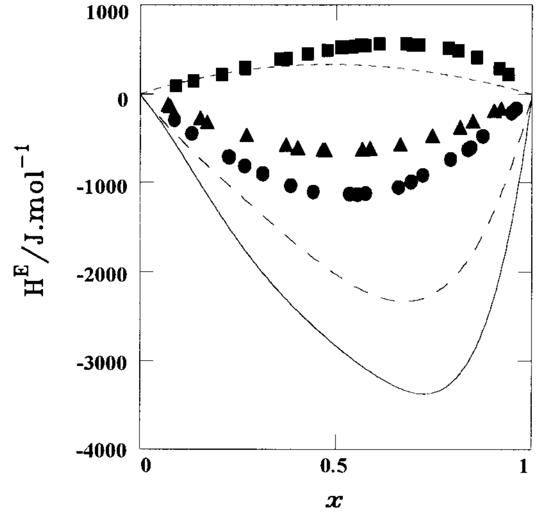

Figure 6. Experimental excess molar enthalpies and predicted ones by using the Gmehling et al. version of the UNIFAC model, of the following systems: methanol + NFM, (■), experimental points and (- -) prediction; methanol + NMP, (▲) experimental points and (— —) prediction; methanol + DMPU, (●), experimental points and (—) prediction.

methanol and the amide is the more important. The methanol + DMPU mixture is more exothermic than the NMP + methanol system. Note that this behavior is in agreement with the observed dependence of the excess enthalpies with the size of the lactam ring (Zivny et al., 1991) and with the fact that DMPU has one methylated amide group more than NMP, as can be seen in Figures 4 and 5.

The obtained sequence of the excess enthalpies (NFM, NMP, DMPU) is different from that of the excess volumes (NMP, DMPU, NFM). This means that the packing effect is much bigger in the case of NFM (H^E positive, V^E negative).

Taking into account the boiling points of the present amides (NFM, 509 K; NMP, 475 K; DMPU, 503 K) and that the exothermic behavior favors the absorption process, we can conclude that the mixture of methanol with DMPU could be more useful in absorption machines than those of the other amides, although other properties must be examined to conclusively determine their reliability.

Comparison with UNIFAC Predictions. In Table 6 we present the theoretical predictions together with the experimental data at equimolecular composition, and in Figure 6 we can see the $H^E(x)$ curves. For the systems methanol + DMPU and methanol + NMP the predictions are not acceptable, with deviations for equimolecular mixtures of 150% and 215%, respectively. In the case of methanol + NFM the predictions are better, with a deviation of 34%. This is due to the fact that the parameters existing in the literature are adequate for the groups of the NFM: DMF ($HCON(CH_2)_2$) and c-CH_2O (cyclic ether). On the contrary, for the other two systems the parameters corresponding to the group interactions $N{-}CH_3$ and $C{=}O$ with c-CH_2 are not available in the literature; for this reason we have utilized the parameters corresponding to these interactions with linear CH_2.

In all cases, the model predicts the sign of the excess enthalpies, but as can be seen in Figure 4, the symmetries of both calculated and experimental $H^E(x)$ curves are all quite different.

Recently, Gmehling (1995) has proposed c-CONC as a new group with c-$CONCH_3$ as subgroup. Its interaction parameters with methanol have not yet been derived. The present experimental H^E values for NMP + methanol might be very useful in determining them.

Registry Numbers Supplied by the Author: Methanol, [67-56-1]; NFM, [4394-85-8]; NMP, [872-50-4]; DMPU, [7226-23-5].

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