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Vapor–Liquid Equilibrium Data for the Methane–Dimethyl Ether and Methane–Diethyl Ether Systems between 282 and 344 K

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A static method described in a previous paper has been used to obtain vapor–liquid equilibrium data for the methane–dimethyl ether and methane–diethyl ether systems at three temperatures. Experimental data are fitted with the Soave, Peng and Robinson, and Mathias cubic equations of state. Adjusting two parameters instead of one allows a slight improvement of the data representation.

Introduction

Knowledge of methane solubility in several organic compounds is very useful in relation to methane and energy transportation. Solubilities of methane in methanol (*1–4*), ethanol (*5*, *6*), and acetone (*7*) have already been studied, but a lot of other mixtures remain to be investigated. To increase the experimental data base, we decided to use a recently developed fast apparatus (*8*) and began with ether compounds. This apparatus was used previously to obtain vapor–liquid equilibrium data for the propane–methanol–carbon dioxide mixtures (*9*). The two selected ethers are dimethyl ether and diethyl ether.

Experimental Section

Apparatus. It is based on a static method with sampling and analysis of vapor and liquid phases. The equilibrium cell is maintained at constant temperature in a liquid bath. It contains a very efficient magnetic stirrer to ensure fast equilibrium and is equipped with two sampling systems that allow withdrawal of microsamples of a diphasic mixture (for example, liquid sample volumes are about 10^{-3} cm^3), which are swept directly into a gas chromatograph. In this way, samples which are representative of the coexisting phases are analyzed in totality. Pressures are measured by means of a pressure transducer calibrated at each working temperature. Temperatures are determined within 0.2 K through calibrated thermocouples. For more details it is recommended to see ref *8*.

Chemicals. Methane was supplied by Messer Griesheim with a certified minimum purity of 99.995 vol %. Dimethyl ether was obtained from Fluka and diethyl ether from Merck; they respectively have GLC minimum purities of 99.8 and 99.5%. They were used without any further purification except for careful degassing of the diethyl ether.

Results and Discussion

The experimental procedure used to carry out the experimental work is the same as that described previously (*8*). In the case of the methane–dimethyl ether system, the "heavy" component, dimethyl ether, which is a gas at room temperature, was introduced into the cell directly from its container through the tubing circuit which was evacuated.

Isothermal vapor–liquid equilibrium results are reported in Table I for methane–dimethyl ether and Table II for methane–diethyl ether systems and plotted in Figures 1 and 3. The partition coefficients are plotted in Figures 2 and 4 as a function of pressure. Temperatures are given in the first columns of Tables I and II, pressures in the second, methane liquid and vapor mole fractions in the third and fifth, partition coefficients of methane and ether in seventh and ninth columns, and respective errors on x_1, y_1, K_1, and K_2 in fourth, sixth, eight, and tenth columns.

Error calculations were performed in the following way: from the expression 1 relating mole fractions z_i to chromatographic measurements

$$z_i = \frac{1}{1 + \sum\limits_{j \neq i} A_{ji} R_{ij}} \qquad z_i = x_i \text{ or } y_i \tag{1}$$

we have the errors given by

$$\Delta z_i = z_i^2 \sum_{j \neq i} (R_{ij} \Delta A_{ji} + A_{ji} \Delta R_{ij}) \tag{2}$$

with

$$\frac{\Delta R_{ij}}{R_{ij}} = \frac{\Delta R_i}{R_i} + \frac{\Delta R_j}{R_j} \tag{3}$$

ΔA_{ji} is estimated from the dispersion of A_i and A_j values obtained by analyses on at least five samples. Each chromatographic analysis needs about 5 min and one sample is taken every 8 min. ΔR_i and ΔR_j are the mean quadratic deviations resulting from a data fitting on the results of the chromatograph detector calibration.

Data Representation. Representation of the experimental data was carried out using three cubic equations of state (see Appendix).

Critical parameters, T_c and P_c, and acentric factors used in this work, are reported in Table III.

Binary interaction parameter adjustments were carried out with temperature and liquid composition chosen as independent variables and by minimizing the objective function Q:

$$Q = \sum_{j=1}^{n} [(P_{j,\text{exptl}} - P_{j,\text{calcd}})/P_{j,\text{exptl}}]^2 + \sum_{j=1}^{n} [(y_{j,\text{exptl}} - y_{j,\text{calcd}})/y_{j,\text{exptl}}]^2 \tag{4}$$

All the results of modelization are reported in Table IV. The standard relative deviations given in this table were calculated through eq 5 and 6.

$$\sigma^{R}{}_{P} = 100 \left[\sum_{i=1}^{n} \left[\frac{P_{i,\text{exptl}} - P_{i,\text{calcd}}}{P_{i,\text{exptl}}} \right]^2 / (n-1) \right]^{1/2} \tag{5}$$

$$\sigma^{R}{}_{y} = 100 \left[\sum_{i=1}^{n} \left[\frac{y_{i,\text{exptl}} - y_{i,\text{calcd}}}{y_{i,\text{exptl}}} \right]^2 / (n-1) \right]^{1/2} \tag{6}$$

Cubic equations of state used in a predictive way, that is to say with zero interaction parameter, are not very accurate (see Table IV). However, the Mathias (*10*) equation appears as the best especially for the methane–diethyl ether system.

Adjusting one binary interaction parameter allows us to decrease the relative standard deviations by a factor of about 2. Mathias' equation of state with two adjusted parameters gives

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Table I. Vapor-Liquid Equilibrium Data for the Methane (1)-Dimethyl Ether (2) System at Different Temperatures[a]

		mole fraction							
		liquid		vapor					
temp, K	press., MPa	x_1	$\pm\Delta x_1$	y_1	$\pm\Delta y_1$	K_1	$\pm\Delta K_1$	K_2	$\pm\Delta K_2$
282.9	1.97	0.062	0.003	0.774	0.007	12.5	0.7	0.241	0.008
282.9	3.96	0.151	0.006	0.858	0.006	5.7	0.3	0.167	0.008
282.9	6.01	0.249	0.006	0.879	0.005	3.53	0.10	0.161	0.008
282.9	7.89	0.339	0.008	0.878	0.005	2.59	0.08	0.185	0.010
282.9	10.47	0.476	0.008	0.856	0.005	1.80	0.04	0.275	0.014
282.9	12.36	0.633	0.008	0.782	0.008	1.24	0.03	0.59	0.03
313.3	2.02	0.044	0.002	0.515	0.007	11.7	0.7	0.507	0.008
313.3	3.03	0.083	0.003	0.645	0.006	7.8	0.4	0.387	0.008
313.3	4.03	0.121	0.004	0.702	0.008	5.8	0.3	0.339	0.011
313.3	6.00	0.208	0.007	0.760	0.007	3.7	0.16	0.303	0.012
313.3	8.00	0.285	0.007	0.774	0.007	2.72	0.09	0.316	0.013
313.3	10.00	0.384	0.008	0.759	0.007	1.98	0.06	0.391	0.016
313.3	11.51	0.486	0.009	0.721	0.006	1.48	0.04	0.54	0.02
313.3	12.16	0.561	0.009	0.659	0.008	1.17	0.03	0.78	0.03
343.8	3.10	0.050	0.002	0.362	0.007	7.2	0.4	0.672	0.009
343.8	4.06	0.084	0.003	0.455	0.007	5.4	0.3	0.595	0.010
343.8	5.03	0.120	0.004	0.523	0.008	4.4	0.2	0.542	0.012
343.8	6.03	0.160	0.005	0.566	0.007	3.54	0.15	0.517	0.011
343.8	7.01	0.199	0.006	0.589	0.008	2.96	0.14	0.513	0.014
343.8	8.01	0.242	0.006	0.603	0.007	2.49	0.09	0.524	0.013
343.8	8.92	0.286	0.007	0.600	0.006	2.10	0.07	0.560	0.014
343.8	10.06	0.354	0.007	0.576	0.006	1.63	0.05	0.656	0.016
343.8	10.66	0.413	0.008	0.532	0.007	1.29	0.04	0.80	0.02

[a] $\Delta P = \pm 0.02$ MPa; $\Delta T = \pm 0.2$ K.

Table II. Vapor-Liquid Equilibrium Data for the Methane (1)-Diethyl Ether (2) System at Different Temperatures[a]

		mole fraction							
		liquid		vapor					
temp, K	press., MPa	x_1	$\pm\Delta x_1$	y_1	$\pm\Delta y_1$	K_1	$\pm\Delta K_1$	$10K_2$	$\pm 10\Delta K_2$
284.3	2.07	0.102	0.003	0.967	0.003	9.5	0.3	0.37	0.04
284.3	3.99	0.189	0.005	0.977	0.002	5.17	0.15	0.284	0.018
284.3	5.94	0.279	0.007	0.979	0.001	3.51	0.09	0.291	0.012
284.3	8.03	0.358	0.008	0.977	0.002	2.73	0.07	0.36	0.03
284.3	10.02	0.437	0.008	0.969	0.002	2.22	0.05	0.55	0.03
284.3	11.99	0.512	0.008	0.960	0.003	1.87	0.04	0.82	0.05
284.3	14.00	0.596	0.008	0.945	0.003	1.59	0.03	1.36	0.07
284.3	14.76	0.628	0.008	0.932	0.003	1.48	0.02	1.83	0.08
284.3	16.30	0.707	0.008	0.896	0.004	1.27	0.02	3.55	0.16
284.3	17.00	0.771	0.007	0.841	0.005	1.09	0.02	6.9	0.3
313.3	2.47	0.098	0.002	0.928	0.004	9.5	0.2	0.80	0.05
313.3	4.95	0.197	0.004	0.949	0.003	4.82	0.11	0.63	0.03
313.3	6.90	0.269	0.005	0.949	0.003	3.53	0.08	0.70	0.03
313.3	10.05	0.384	0.006	0.941	0.003	2.45	0.05	0.96	0.04
313.3	12.25	0.471	0.007	0.927	0.005	1.97	0.04	1.38	0.08
313.3	15.05	0.557	0.007	0.902	0.004	1.62	0.03	2.21	0.09
313.3	15.92	0.590	0.006	0.887	0.005	1.50	0.02	2.76	0.16
313.3	17.04	0.644	0.006	0.868	0.005	1.35	0.02	3.71	0.14
342.5	2.13	0.073	0.002	0.817	0.005	11.2	0.4	0.197	0.006
342.5	4.11	0.149	0.004	0.888	0.006	6.0	0.2	0.132	0.008
342.5	6.11	0.217	0.005	0.902	0.005	4.16	0.12	0.125	0.007
342.5	8.10	0.285	0.006	0.907	0.004	3.18	0.08	0.130	0.007
342.5	10.07	0.348	0.006	0.906	0.004	2.60	0.06	0.144	0.007
342.5	12.08	0.420	0.007	0.897	0.004	2.14	0.05	0.178	0.009
342.5	14.69	0.516	0.006	0.859	0.004	1.66	0.03	0.291	0.012
342.5	16.42	0.602	0.006	0.802	0.005	1.33	0.02	0.50	0.02
342.5	17.06	0.647	0.006	0.763	0.005	1.18	0.02	0.67	0.03

[a] $\Delta P = \pm 0.02$ MPa; $\Delta T = \pm 0.2$ K.

Table III. Critical Parameters and Acentric Factors[a]

component	crit press., MPa	crit temp, K	acentric factor
methane	4.60	190.6	0.008
dimethyl ether	5.37	400.0	0.192
diethyl ether	3.64	466.7	0.281

[a] From ref *14*.

results slightly improved with respect to those from one-parameter Soave (*11*) and Peng-Robinson (*12*) equations of state.

Appendix: Data Treatment

The three modified forms of the Redlich-Kwong cubic equation of state used in this work are issued from generic expression

$$P = \frac{RT}{v - b} - \frac{a(T)}{bv} f(\beta) \tag{A1}$$

$$\text{where } \beta = b/v \tag{A2}$$

***Soave's Equation* (*11*).** It is obtained by writing

Table IV. Vapor-Liquid Equilibrium Data Representation

mixture	Soave			Peng-Robinson			Mathias[a]			
	k_{12}	σ^R_p	σ^R_y	δ_{12}	σ^R_p	σ^R_y	k^0_{a12}	k^0_{b12}	σ^R_p	σ^R_y
methane-dimethyl ether	0	6.0	5.5	0	7.7	5.9	0	0	4.2	4.2
	0.03	3.5	3.9	0.036	3.6	3.9	-0.017	-0.033	2.7	3.5
methane-diethyl ether	0	7.5	3.2	0	9.7	2.7	0	0	3.2	2.9
	0.024	3.7	2.7	0.031	3.6	2.0	-0.003	-0.011	2.2	2.7

[a] k^1_{a12} and k^1_{b12} have null values.

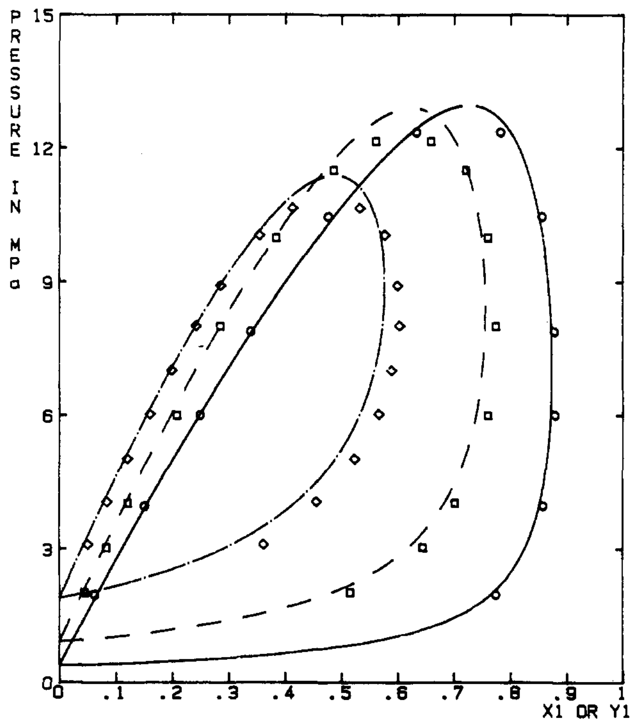

Figure 1. Pressure as a function of methane mole fraction in the system methane-dimethyl ether at 282.9 K (○), 313.3 K (□), and 343.8 K (◇). The curves were calculated with the Mathias equation of state and adjusted parameters given in Table IV.

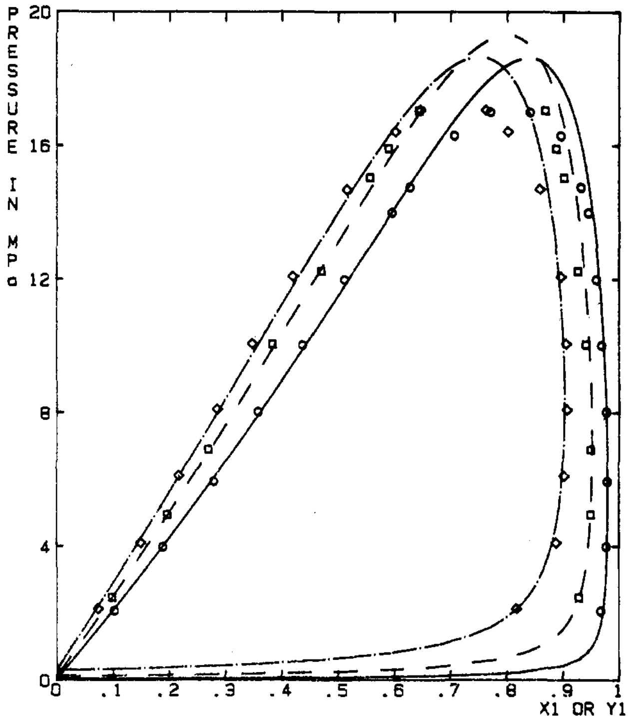

Figure 3. Pressure as a function of methane mole fraction in the system methane-diethyl ether at 284.3 K (○), 313.3 K (□), and 342.5 K (◇). The curves were calculated with the Mathias equation of state and adjusted parameters given in Table IV.

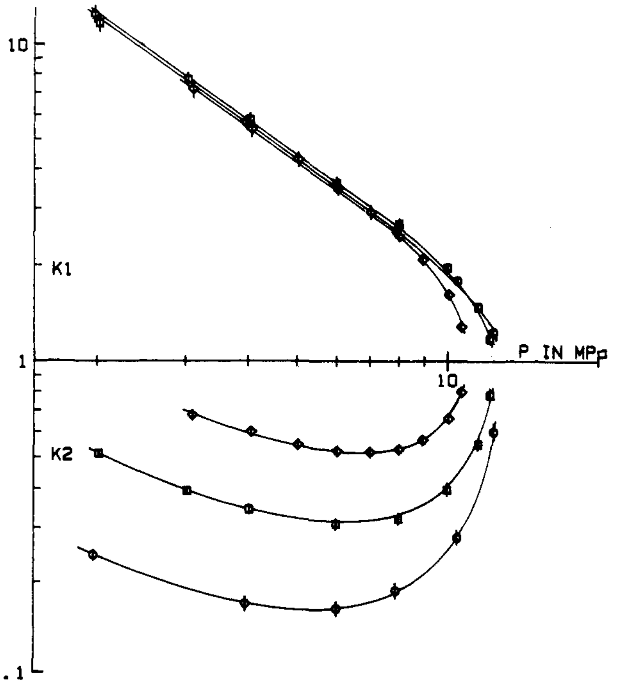

Figure 2. Partition coefficients as a function of pressure in the methane-dimethyl ether system at 282.9 K (○), 313.3 K (□), and 343.8 K (◇).

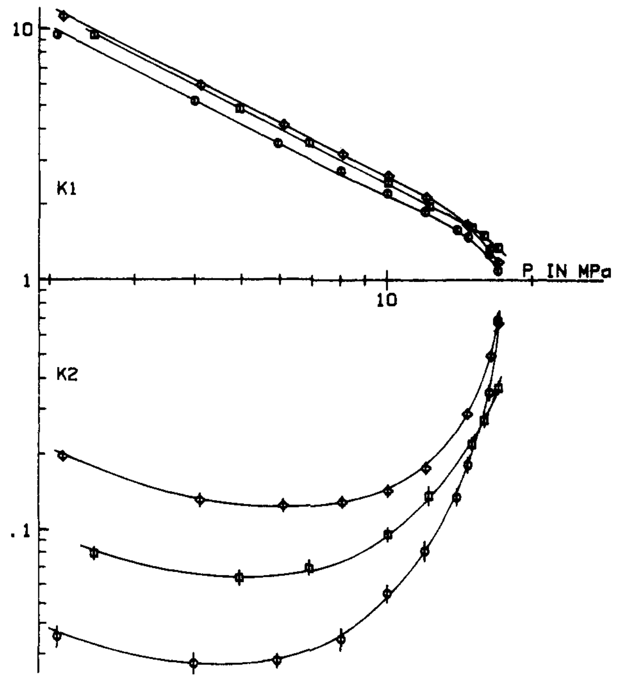

Figure 4. Partition coefficients as a function of pressure in the methane-diethyl ether system at 284.3 K (○), 313.3 K (□), and 342.5 K (◇).

$$f(\beta) = \beta/(1 + \beta) \qquad \text{(A3)}$$

i.e.

$$P = \frac{RT}{v - b} - \frac{a(T)}{v(v + b)} \qquad \text{(A4)}$$

For pure components

$$a(T) = 0.42747 \frac{R^2T_c^2}{P_c} \alpha(T) \qquad \text{(A5)}$$

$$b = 0.08664 \frac{RT_c}{P_c} \qquad \text{(A6)}$$

$$\alpha(T) = [1 + m(1 - T_R^{1/2})]^2 \qquad \text{(A7)}$$

$$m = 0.48 + 1.574w - 0.176w^2 \qquad \text{(A8)}$$

w is the acentric factor.

In this work, we used the expression given by Graboski and Daubert (*13*), instead of eq A8:

$$m = 0.48508 + 1.55171w - 0.15613w^2 \qquad \text{(A9)}$$

For mixtures, the following mixing rules were used

$$a = \sum_i \sum_j x_i x_j (1 - k_{ij})(a_i a_j)^{1/2} \qquad \text{(A10)}$$

$$b = \sum_i x_i b_i \qquad \text{(A11)}$$

where a_i and b_i are computed from eq A5 and A6 using the values of critical pressure, critical temperature, and acentric factor of each component; k_{ij} is an adjustable binary interaction parameter.

The fugacity coefficient ϕ_i is expressed by

$$\ln \phi_i = \ln\left(\frac{f_i}{Px_i}\right) = \ln \frac{v}{v - b} - \ln \frac{Pv}{RT} - \frac{2u}{RTb} \ln \frac{v + b}{v} + b_i\left[\frac{a}{RTb^2}\left(\ln \frac{v + b}{v} - \frac{b}{v + b}\right) + \frac{1}{v - b}\right] \qquad \text{(A12)}$$

where

$$u = \sum_j (1 - k_{ij})(a_i a_j)^{1/2} x_j \qquad \text{(A13)}$$

Peng and Robinson Equation of State (12). For this equation

$$f(\beta) = \frac{\beta}{1 + 2\beta - \beta^2} \qquad \text{(A14)}$$

which leads to

$$P = \frac{RT}{v - b} - \frac{a(T)}{v(v + b) + b(v - b)} \qquad \text{(A15)}$$

For pure components

$$a(T) = 0.45724 \frac{R^2T_c^2}{P_c} \alpha(T) \qquad \text{(A16)}$$

$$b = 0.0778 \frac{RT_c}{P_c} \qquad \text{(A17)}$$

$\alpha(T)$ is given by eq (A7), but

$$m = 0.37464 + 1.54226w - 0.26992w^2 \qquad \text{(A18)}$$

Mixing rules are given by eq A10 and A11; there are identical with those used by Soave. In the text we shall write δ_{ij} instead of k_{ij} when dealing with Peng–Robinson's data treatment.

Fugacity coefficient has the following form

$$\ln \phi_i = \frac{b_i}{b}\left(\frac{Pv}{RT} - 1\right) - \ln\left[\frac{Pv}{RT}\left(1 - \frac{b}{v}\right)\right] - \frac{1}{2bRT(2)^{1/2}}\left[2u - \frac{ab_i}{b}\right] \ln \frac{v + b(1 + 2^{1/2})}{v + b(1 - 2^{1/2})} \qquad \text{(A19)}$$

with u given by eq A13 where k_{ij} is replaced by δ_{ij}.

Mathias's Equation (10). For this equation we have

$$f(\beta) = \beta/(1 + \beta) \qquad \text{(A20)}$$

An essential difference from Soave's equation of state is provided by the way chosen to write $\alpha(T)$.

When $T < T_c$

$$\alpha(T) = [1 + m(1 - T_R^{1/2}) - p(1 - T_R)(0.7 - T_R)]^2 \qquad \text{(A21)}$$

with

$$m = 0.48508 + 1.55171w - 0.15613w^2 \qquad \text{(A22)}$$

And when $T > T_c$ then

$$\alpha(T) = \exp[d(1 - T_R^c)] \qquad \text{(A23)}$$

with

$$c = 1 + m/2 + 0.3p \qquad \text{(A24)}$$

$$d = (c - 1)/c \qquad \text{(A25)}$$

P is an adjustable parameter for pure polar components. In this work a null value for P has been used. $\alpha(T)$ is given by eq A7 for $T < T_c$.

Mixing rules proposed by Mathias are

$$a = \sum_i \sum_j x_i x_j (a_i a_j)^{1/2}\left(1 - k^0{}_{aij} - \frac{T}{1000} k^1{}_{aij}\right) \qquad \text{(A26)}$$

$$b = \sum_i \sum_j x_i x_j \frac{bi + bj}{2}\left(1 - k^0{}_{bij} - \frac{T}{1000} k^1{}_{bij}\right) \qquad \text{(A27)}$$

$$\ln \phi_i = \ln\left(\frac{fi}{Pxi}\right) = \ln \frac{v}{v - b} - \ln \frac{Pv}{RT} - \frac{2u}{RTb} \ln\left(\frac{v + b}{v}\right) + (2u' - b)\left[\frac{a}{RT^2b^2}\left(\frac{v + b}{v} - \frac{b}{v + b}\right) + \frac{1}{v - b}\right] \qquad \text{(A28)}$$

with u and u' given by

$$u = \sum_j (a_i a_j)^{1/2}\left(1 - k^0{}_{aij} - \frac{T}{1000} k^1{}_{aij}\right) x_j \qquad \text{(A29)}$$

$$u' = \sum_j \frac{bi + bj}{2}\left(1 - k^0{}_{bij} - \frac{T}{1000} k^1{}_{bij}\right) x_j \qquad \text{(A30)}$$

Glossary

a	energy parameter
A_i	chromatographic peak area for component i
A_{ij}	ratio of chromatographic peak areas: Ai/Aj
b	covolume parameter
K_i	partition coefficient (= y_i/x_i)
k_{ij}	binary interaction parameter in Soave's equation of state
$k^0{}_{aij}$, $k^1{}_{aij}$, $k^0{}_{bij}$, $k^1{}_{bij}$	binary interaction parameters in Mathias' equation of state
n	number of experimental data
Q	objective function
R	gas constant
R_i	response coefficient to component i
R_{ij}	ratio of chromatographic response coefficients: Ri/Rj
p	polar parameter in eq A21 and A24

P	pressure, MPa
T	temperature, K
x	liquid mole fraction
y	vapor mole fraction
v	molar volume, $cm^3 \cdot mol^{-1}$
w	acentric factor

Subscripts

c	critical property
R	reduced property
exptl	experimental property
calcd	calculated property
P	pressure
y	vapor mole fraction

Superscripts

c	parameter in equations A23 to A25
R	relative quantity

Greek Letters

δ	binary interaction parameter in Peng-Robinson equation of state
ϕ	fugacity coefficient
σ	standard deviation

Registry No. Methane, 74-82-8; dimethyl ether, 115-10-6; diethyl ether, 60-29-7.

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Vapor-Liquid Equilibrium Data for the Methane-Methylal Binary Mixture between 273 and 432 K up to the Critical Region

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Three static apparatuses were used to obtain vapor-liquid equilibrium data for the methane-methylal system at five different temperatures. Experimental data were represented by several models. The Soave equation used in combination with an excess Gibbs free energy equation for mixing rules was found the best. Similar calculations have been performed with the Soave, Peng-Robinson, and Mathias equations of state and with the Skjold-Jorgensen equation.

Introduction

Cubic equations of state have been extensively used for the prediction and correlation of high-pressure vapor-liquid equilibrium (*1*). Legret et al. (*2*) have shown that it is possible to correlate a large number of methane-hydrocarbon experimental binary data sets using the Soave (*3*) and Peng-Robinson (*4*) equations of state. Data related to methane with polar or slightly polar compounds are scarce. For this reason, we have measured vapor-liquid equilibria of the methane-methylal mixture. These data make possible the investigation of the ability of cubic equations of state with classical mixing rules to correlate the data. In the present work, more advanced equations recently proposed for very nonideal mixtures at high pressures are tested: Mathias (*5*), Soave with nonclassical mixing rules (*6*), and Skjold-Jorgensen (*7*).

Experimental Section

Apparatus. The two first apparatuses A (*8*) and B (*9*) are based on a static method with sampling and analysis of both phases. The third apparatus (*10*) has a variable-volume cell which makes it possible to record pressure vs. volume data and then identify bubble pressures very accurately. Composition of the liquid phase is determined in the latter apparatus by a very accurate weighing of the components introduced separately inside the equilibrium cell. The third apparatus has been essentially used to test reliability of data obtained with the two other apparatuses.

Chemicals. Methane was supplied by Messer Griesheim with a certified minimum purity of 99.995% by volume. Methylal [$CH_2(OCH_3)_2$] was provided by Fluka with a GLC certified minimum purity of 99%. The chemicals were used without any further purification except for the careful degassing of methylal.

Results and Discussion

Experimental procedures used to carry out all the experimental work were described in previous papers for apparatus

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