equilibrium ratios for each component in a binary system converge to unity at the critical pressure of the mixture that has any specified temperature as its critical temperature, it is possible to estimate the critical pressure from a graphical representation of equilibrium ratios as a function of pressure at fixed temperatures. This method was used to estimate the critical pressures at 37.8, 71.1, and 104.4 °C from the data of Besserer and Robinson (*1*, *3*).

The critical loci for the binary systems studied in this work were constructed by using the values given in Table III and are presented graphically in Figure 5. This shows the six binary critical points and the vapor pressure curves and critical point for each of the pure components.

Discussion

The completion of this study has filled the temperature gap that existed in the binary vapor-liquid equilibrium data for the *n*-pentane- and isopentane-carbon dioxide binary systems. The additional information will be useful for evaluating any possible temperature dependence of the binary interaction parameters for these systems. This may assist in improving the reliability of vapor-liquid equilibrium and fluid property predictions based on equations of state or other correlations requiring these parameters for multicomponent calculations.

Additional work is in progress on vapor-liquid equilibrium measurements in mixtures of neopentane with carbon dioxide. This will complete the vapor-liquid equilibrium data required for carbon dioxide with all the pentanes.

Glossary

K	equilibrium ratio, y/x
K_{CO_2}	equilibrium ratio for carbon dioxide
K_{nC_5}	equilibrium ratio for *n*-pentane
K_{iC_5}	equilibrium ratio for isopentane
P	pressure, MPa
T	temperature, °C
x	mole fraction of component in liquid phase
y	mole fraction of component in vapor phase

Registry No. *n*-C_5, 109-66-0; *i*-C_5, 78-78-4; CO_2, 124-38-9.

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Measurement of γ^∞ Using GLC. 2. Results for the Stationary Phases *N*-Formylmorpholine and *N*-Methylpyrrolidone

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Gas-liquid chromatography (GLC) has been used to determine activity coefficients at infinite dilution for a variety of classes of substances; the stationary phases employed were *N*-formylmorpholine (NFM) and *N*-methylpyrrolidone (NMP). These two substances are used technically as solvents in extractive distillation, particularly for the separation of alkanes from aromatics or alkanes from alkenes. In order to prevent the columns with these substances from "bleeding out" during the measurements, the apparatus was modified in such a way that the carrier gas is saturated with the liquid stationary phase before entering the column.

Table I. Vapor Pressures of NFM and NMP as a Function of Temperature

	Antoine constants		
	A	*B*	*C*
NFM	7.28945	2075.15	228.119
NMP	8.2789	2570.30	273.15

T, °C	P_{NFM}, mmHg	P_{NMP}, mmHg
25	0.12	0.46
50	0.67	2.11
75	2.78	7.87
100	9.23	24.59
125	25.87	66.57

Introduction

The measurement of activity coefficients at infinite dilution is an alternative to the conventional static and dynamic methods for the determination of vapor-liquid equilibria. The basic idea of the γ^∞ measurements is to use the information on the activity coefficients at infinite dilution to predict the behavior of the activity coefficients and the vapor-liquid equilibrium across the whole concentration range with the help of a suitable thermodynamic model. The obtained data were also stored in the Dortmund Data Bank (*1*) and, together with VLE (*2*) and h^E data (*3*), used for the further development of group contribution methods (*4*). The GLC method, together with the theoretical background to γ^∞ measurements carried out using gas chromatography, is described in detail by Weidlich and Gmehling (*5*); the stationary phases used there are octacosane, 1-docosanol, 10-nonadecanone, and 1-eicosene. The aim of the present work was to modify the apparatus in such a way that it is also possible to carry out measurements using stationary phases which have relatively high vapor pressures at the temperatures used.

The phases chosen were *N*-methylpyrrolidone (NMP) and *N*-formylmorpholine (NFM). Both are used technically as sol-

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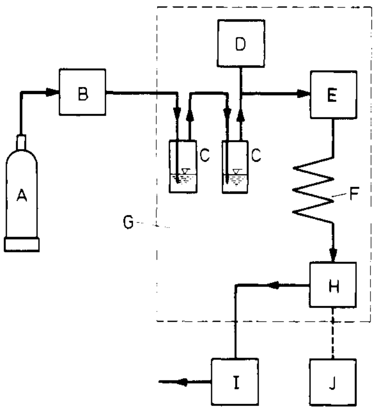

Figure 1. Schematic representation of the gas-chromatographic measurement setup: (A) source of carrier gas; (B) pressure adjustment; (C) saturation vessel; (D) pressure measurement; (E) injector block; (F) column; (G) oven; (H) detector; (I) flow indicator; (J) integrator.

vents in extractive distillation (e.g., for separation of alkanes from aromatics or of butadiene from a C_4 fraction) and are thus of particular interest.

Apparatus and Measurement Techniques

Table I shows the vapor pressures of NMF and NMP as a function of the temperature. The measurement of activity coefficients at infinite dilution on columns coated with these substances is possible at higher temperatures only when the carrier gas used is saturated with the relevant stationary phase before it enters the column. The saturation must be complete in order to avoid weight loss. Pescar and Martin (*6*) suggest passing the carrier gas through a precolumn, also coated with the stationary phase, before it enters the actual measurement column. Shaffer and Daubert (*7*) passed the carrier gas through a wash-bottle filled with the stationary phase. The first method has in comparison with the second the advantage that the distance which must be traversed by the carrier gas before it enters the column is longer and that a better saturation may thus be realized. The disadvantage is a rapid "bleeding out" of the column. There were additional technical problems due to lack of space in the oven of the gas chromatograph used; these led to the rejection of this proposed solution.

In order to prevent as far as possible the evaporation of the stationary phase, the carrier gas was first warmed to oven temperature and then passed through two wash-bottles filled with the stationary phase before it entered the column. The saturation was carried out directly in the oven of the gas chromatograph, thus ensuring that the temperature in the saturation vessel corresponded to the oven temperature used. The wash-bottles are in principle bubble columns with a gas distributor which causes a fine distribution of the gas within the liquid.

The gas-chromatographic experimental setup for the measurement of activity coefficients at infinite dilution is shown schematically in Figure 1.

Further details of the apparatus and the experimental procedure are given by Weidlich and Gmehling (*4*). At higher temperatures, where weight loss occurred in spite of presaturation (since it was not possible to achieve complete saturation of the carrier gas even with the present method) it was possible to determine the mass of the stationary phase present during a particular measurement using an interpolation method. This requires that the time corresponding to the total weight loss and the time from the beginning of the series to the measurement in question be known. In order to minimize the weight loss, the columns were changed after completion of each series of measurements (ca. 8 h).

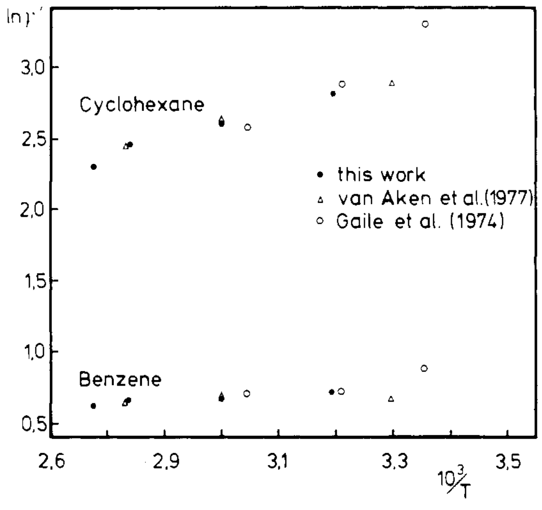

Figure 2. Comparison of activity coefficients at infinite dilution for cyclohexane and benzene in NFM obtained by different authors.

Results

The quantity actually measured is the specific retention volume V_g^0 corrected to 0 °C.

$$V_g^0 = (t_R - t_A)\frac{273.15F}{T_{fm}m_L}\left[\frac{p_{fm}-p_w}{p_{fm}}\right]\frac{3}{2}\left[\frac{(p_E/p_A)^2-1}{(p_E/p_A)^3-1}\right] \quad (1)$$

where V_g^0 is the specific retention volume (corrected to 0 °C), t_R retention time, t_A dead time (retention time of an inert gas), F gas flow, T_{fm} temperature at the flow meter in K, p_{fm} pressure at the flow meter, p_E pressure at column entrance, p_A pressure at column exit, m_L mass of stationary phase, and p_w vapor pressure of water at T_{fm}. The activity coefficient at infinite dilution can be determined from the specific retention volume V_g^0 by using the equation

$$\gamma^\infty = \frac{273.15R}{V_g^0\varphi_i^s P_i^s M_L} \quad (2)$$

where γ^∞ is the activity coefficient at infinite dilution, M_L molecular weight of the stationary phase, P_i^s vapor pressure of component i, R gas constant, and φ_i^s fugacity of the component i in the saturated state. The fugacity coefficients in the saturated state φ_i^s were obtained in this work from the Soave equation (*8*) as modified by Graboski and Daubert (*9*).

Activity coefficients at infinite dilution of substances of various types were determined on NFM and NMP at up to three temperatures. Because of a particular interest with regard to the separation of alkanes and aromatics by extractive distillation, the measurements of these on NFM were carried out at four temperatures. The choice of the substances studied was determined in part by the temperature range usable for the stationary phase concerned: thus some high-boiling compounds could not be measured at low temperatures since the expected retention time would have been too long. The results of the measurement are collected in Tables II–IV. Apart from activity coefficients at infinite dilution, these tables also contain the specific retention volumes corrected to 0 °C, since the retention volume is the actual parameter measured: the limiting activity coefficients are determined from these values.

The results for cyclohexane and benzene for the solvent *N*-formylmorpholine as function of the temperature are shown in Figure 2 together with the results of other authors. As can be seen there is good agreement between our results and the results of van Aken (*10*) and Gaile (*11*).

Table II. Experimentally Determined Retention Volumes and Activity Coefficients at Infinite Dilution on *N*-Formylmorpholine ($C_5H_9NO_2$)

	40.1 °C		59.5 °C		79.3 °C		100.2 °C	
	V_g^0	γ^∞	V_g^0	γ^∞	V_g^0	γ^∞	V_g^0	γ^∞
n-pentane	7.5	23.6	5.11	19.3	3.61	16.4	2.75	13.5
n-hexane	16.8	32.1	11.0	24.6	7.19	20.6	5.05	17.0
n-heptane	37.8	42.3	23.3	31.4	13.8	26.1	9.0	21.5
n-octane	87.0	54.2	48.2	40.7	26.3	33.7	16.5	26.3
isooctane	31.6	48.2	19.8	36.0	11.9	30.7	8.2	24.4
n-decane	434.1	90.1	201.1	65.8	95.6	53.3	49.5	42.2
cyclopentane	23.2	11.7	14.7	9.92	9.55	8.76	6.65	7.60
methylcyclopentane	33.6	17.5	21.0	14.2	12.9	12.5	8.66	10.7
cyclohexane	48.7	16.7	29.5	13.5	17.9	11.7	11.7	10.0
methylcyclohexane	71.3	22.8	41.0	18.5	24.8	15.6	15.7	13.2
ethylcyclohexane	178.1	29.6	94.5	23.3	53.1	19.1	32.2	15.5
trans-1,4-dimethylcyclohexane	104.4	30.5	56.8	24.3	32.5	20.7		
cis-1,4-dimethylcyclohexane	139.6	28.2	73.2	22.7	42.6	18.4		
trans-1,2-dimethylcyclohexane	128.0	28.7	68.6	23.1	40.3	18.7	25.7	15.1
cis-1,2-dimethylcyclohexane	182.3	26.2	97.2	20.6	53.5	17.6	33.1	14.1
benzene	394.1	2.05	196.7	1.96	104.6	1.94	60.5	1.86
toluene	854.9	2.78	412.3	2.65	203.1	2.58	109.4	2.46
o-xylene	2757.0	3.53	1106.0	3.35	498.9	3.23	251.2	2.98
m-xylene	1925.0	4.10	802.3	3.81	368.3	3.68	190.2	3.34
p-xylene	1712.0	4.07	767.7	3.84	355.3	3.71	175.5	3.56
ethylbenzene	1794.0	3.88	773.8	3.60	344.6	3.59	170.9	3.44
isopropylbenzene	2699.0	4.95	1107.0	4.54	476.2	4.44	223.6	4.29

Table III. Determination of the Specific Retention Volumes and Activity Coefficients at Infinite Dilution on *N*-Formylmorpholine ($C_5H_9NO_2$)

	30.3 °C		50.0 °C		69.6 °C	
	V_g^0	γ^∞	V_g^0	γ^∞	V_g^0	γ^∞
1-hexene	37.0	17.5	21.6	14.5	14.2	11.9
1-octene	203.3	31.5	99.9	24.6	57.3	19.0
methanol	827.0	1.07	336.0	1.05	156.4	1.04
ethanol	1221	1.52	427.2	1.41	207.2	1.35
1-propanol			1040	1.56	424.1	1.46
2-propanol	1286	1.94	513.3	1.70	227.5	1.52
2-methyl-2-propanol	1103	2.35	452.5	1.89	199.5	1.75
methyl acetate	298.4	1.85	146.2	1.76	79.1	1.69
ethyl acetate	491.2	2.48	223.8	2.36	118.4	2.18
propyl acetate	1081	3.18	452.7	2.97	211.9	2.84
ethyl propionate	891.9	3.38	375.3	3.18	180.6	3.01
ethyl butyrate			640.4	3.88	303.1	3.65
vinyl acetate	530.5	1.90	244.0	1.85	123.4	1.81
acetone	331.6	1.57	162.5	1.52	88.6	1.48
2-butanone	665.6	1.93	308.2	1.84	159.6	1.75
2-pentanone	1289	2.48	557.0	2.31	267.5	2.21
3-pentanone	1152	2.79	517.9	2.48	248.2	2.38
4-methyl-2-pentanone			660.3	3.23	319.9	2.93
n-butanal	500.5	2.09	234.6	2.01	122.5	1.96
2-methylpropanal	297.4	2.32	144.6	2.25	80.2	2.13
n-pentanal	1221	2.68	520.2	2.56	246.2	2.47
crotonaldehyde			785.6	1.61	387.8	1.51
thiophene	1062	1.38	478.2	1.34	246.2	1.30

Discussion

The results of the measurements with NFM and NMP as the stationary phase make clear the different influences which these two substances have on the value of the activity coefficients at infinite dilution. Table V contains the results for *n*-heptane, methylcyclohexane, cyclohexane, toluene, and benzene on NFM in order to make such influences clear. The activity coefficients of *n*-heptane at infinite dilution are up to 15 times larger and those of methylcyclohexane up to 8 times larger than those of toluene. The temperature dependence is also different: while the value of *n*-heptane decreases between 40 and 100 °C by ca. 50% and that of methylcyclohexane by ca. 40%, the value for toluene decreases by only ca. 10%. The limiting activity coefficients of cyclohexane in NFM are also up to 8 times larger than those of benzene; the temperature dependence is similar to that for methylcyclohexane and toluene.

Table IV. Determination of the Specific Retention Volumes and Activity Coefficients at Infinite Dilution on *N*-Methylpyrrolidone (C_5H_9NO)

	50.2 °C		60.0 °C		70.2 °C	
	V_g^0	γ^∞	V_g^0	γ^∞	V_g^0	γ^∞
n-pentane	13.2	11.5	11.1	10.2	9.21	9.35
n-hexane	31.8	13.5	24.6	12.7	19.5	11.6
n-heptane	75.5	16.3	57.0	14.7	43.2	13.4
n-octane	180.	18.9	134.9	16.8	91.1	16.1
isooctane	66.7	17.9	51.2	16.0	39.8	14.4
cyclopentane	35.	6.41	27.6	6.07	21.3	5.84
cyclohexane	78.7	8.18	59.7	7.66	44.2	7.34
methylcyclopentane	56.4	8.41	43.1	7.92	32.8	7.50
methylcyclohexane	129.9	9.67	95.7	9.14	70.	8.64
benzene	497.6	1.27	362.5	1.23	246.7	1.28
toluene	1121.	1.66	735.3	1.67	509.2	1.66
1-hexene	49.2	7.44	38.5	6.86	29.7	6.47
1-octene	266.9	10.7	190.6	9.87	135.	9.10
methanol	784.1	0.527	521.8	0.521	360.7	0.517
ethanol	1168.	0.661	776.8	0.639	500.0	0.643
2-propanol	1308.	0.754	805.0	0.755	533.1	0.737
2-methyl-2-propanol	1236.	0.803	774.0	0.801	472.7	0.834
methyl acetate	181.4	1.62	132.8	1.59	95.2	1.59
propyl acetate	709.6	2.16	469.1	2.14	323.6	2.12
ethyl propionate	591.4	2.32	401.9	2.26	280.0	2.18
ethyl butyrate	1193.0	2.41	817.7	2.31	566.9	2.22
vinyl acetate	351.1	1.47	254.0	1.44	172.7	1.46
acetone	214.8	1.32	159.6	1.29	116.4	1.29
2-butanone	447.4	1.45	309.6	1.45	221.0	1.42
2-pentanone	885.9	1.67	595.5	1.64	416.0	1.62
3-pentanone	881.9	1.66	615.9	1.60	407.1	1.65
n-butanal	360.0	1.51	260.8	1.48	179.3	1.51
2-methylpropanal	221.5	1.68	166.4	1.62	120.0	1.62
n-pentanal	868.5	1.74	610.3	1.65	434.4	1.59
crotonaldehyde	1328.	1.08	902.9	1.07	609.5	1.10
thiophene	898.1	0.823	630.9	0.804	457.5	0.794

Table V. Comparison of the Activity Coefficients at Infinite Dilution for Five Different Alkanes and Aromatics with NFM as the Stationary Phase

	40.1 °C	59.5 °C	79.3 °C	100.2 °C
n-heptane	42.3	31.4	26.1	21.5
methylcyclohexane	22.8	18.5	15.6	13.2
cyclohexane	16.7	13.5	11.7	10.0
toluene	2.78	2.65	2.58	2.46
benzene	2.05	1.96	1.94	1.86

The effect of the additives in extractive distillation becomes clear when the dependence of the separation factor α_{12} on the activity coefficients of the substances involved is considered.

$$\alpha_{12} = \frac{y_1x_2}{x_1y_2} = \frac{\gamma_1P_1^s}{\gamma_2P_2^s}$$

The basis for the separation is either differences in the vapor presence or in the activity coefficients or in both. In extractive distillation the separation of substances of similar volatility is made possible by differing influence on activity coefficients due to the pressure of additive such as NFM and NMP. A detailed treatment of possible errors involved in the measurements described can be found in part I of this work (*4*).

Such a discussion is much more difficult for measurements using NFM and NMP, since the weight losses of the stationary phase can (in spite of presaturation of the carrier gas) no longer be neglected; it is difficult to assess the exactness of the interpolation made to compensate for this weight loss. However, in order to be able to estimate the error a substance with a medium retention time was chosen and measured at definite intervals. The weight of the stationary phase was corrected as a function of time and the standard deviation of the measured limiting activity coefficients calculated. The deviations obtained by using this procedure were as follows: NFM, T = 69.6 °C, 4.0%; NFM, T = 79.3 °C, 4.0%; NFM, T = 100.2 °C, 5.5%; NMP, T = 50.2 °C, 4.5%; NMP, T = 60.0 °C, 5.5%; NMP, T = 70.2 °C, 6.5%. A standard deviation of 3% was calculated for the measurements on NFM at 30.3, 40.1, 50.0, and 59.9 °C. At these temperatures the weight loss was practically negligible or could be extremely exactly corrected for by interpolation. Another aspect which must be considered is that even with very good presaturation solvent bleed will still occur because of the pressure drop across the column. In designing the experiment the constraint is that one must keep the pressure drop across the column as low as possible in order to minimize the solvent bleed. In general it is advisable to use short columns or coarse supports.

Conclusion

Activity coefficients at infinite dilution can be determined rapidly and to a satisfactory degree of exactitude by using GLC. For solvents with nonnegligible vapor pressures, the error in the measurement is caused mainly by loss of stationary phase. This error can however, be decreased by even more careful presaturation and thermostating, so that this technique may well be applicable to the measurement of mixtures for which up until now ebulliometry has been used. Thus a rapid method for assessing the suitability of additives is available; at the same time the foundation for a preliminary estimative separation column dimensioning is laid.

Acknowledgment

We thank Prof. Dr. U. Onken for his supporting interest in this work.

Glossary

A, B, C	Antoine constants
F	gas flow
h_i^E	partial molar excess enthalpy of component i at infinite dilution
M_L	molecular weight of the stationary phase
m_L	mass on stationary phase
p_A	pressure at column exit
p_E	pressure at column entrance
p_{fm}	pressure at the flow meter
P_i^s	vapor pressure of component i
p_w	vapor pressure of water
R	gas constant
T	temperature
t_A	dead time
T_{fm}	temperature at flow meter, K
t_R	retention time
V_g^0	specific retention volume (corrected to 0 °C)
VLE	vapor–liquid equilibrium
x_i	mole fraction of component i in the liquid phase
y_i	mole fraction of component i in the vapor phase
α_{ij}	separation factor
γ_i	activity coefficient of component i
γ_i^∞	activity coefficient at infinite dilution of component i
φ_i^s	fugacity coefficient of component i in the saturation state

Registry No. *n*-Pentane, 109-66-0; *n*-hexane, 110-54-3; *n*-heptane, 142-82-5; *n*-octane, 111-65-9; isooctane, 540-84-1; cyclopentane, 287-92-3; methylcyclopentane, 96-37-7; cyclohexane, 110-82-7; methylcyclohexane, 108-87-2; benzene, 71-43-2; toluene, 108-88-3; 1-hexene, 592-41-6; 1-octene, 111-66-0; methanol, 67-56-1; ethanol, 64-17-5; 2-propanol, 67-63-0; *tert*-butanol, 75-65-0; methyl acetate, 79-20-9; propyl acetate, 109-60-4; ethyl propionate, 105-37-3; Ethyl butyrate, 105-54-4; vinyl acetate, 108-05-4; acetone, 67-64-1; 2-butanone, 78-93-3; 2-pentanone, 107-87-9; 3-pentanone, 96-22-0; *n*-butanal, 123-72-8; 2-methylpropanal, 78-84-2; *n*-pentanal, 110-62-3; crotonaldehyde, 4170-30-3; thiophene, 110-02-1; *n*-decane, 124-18-5; ethylcyclohexane, 1678-91-7; *trans*-1,4-dimethylcyclohexane, 2207-04-7; *cis*-1,4-dimethylcyclohexane, 624-29-3; *trans*-1,2-dimethylcyclohexane, 6876-23-9; *cis*-1,2-dimethylcyclohexane, 2207-01-4; *o*-xylene, 95-47-6; *m*-xylene, 108-38-3; *p*-xylene, 106-42-3; ethylbenzene, 100-41-4; isopropylbenzene, 98-82-8; 1-propanol, 123-38-6; ethyl acetate, 141-78-6; 4-methyl-2-pentanone, 108-10-1; *N*-formylmorphine, 4394-85-8; *N*-methylpyrrolidone, 872-50-4.

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