$\sigma_H(X_{12})$ = standard deviation between theoretical and experimental molar excess enthalpies
σ_V = standard error of estimate for representation of molar excess volume results by Equation 1
θ = site fraction

Subscripts

1 = component of binary mixture with shorter hydrocarbon chain
2 = component of binary mixture with longer hydrocarbon chain

Superscripts

* = characteristic values of p, v, and T
$\tilde{}$ = reduced values of v and T

LITERATURE CITED

(1) Abe, A., Flory, P.J., *J. Amer. Chem. Soc.* **87**, 1838 (1965).
(2) Benson, G.C., Singh, J., *J. Phys. Chem.* **72**, 1345 (1968).
(3) Flory, P.J., *J. Amer. Chem. Soc.* **87**, 1833 (1965).
(4) Flory, P.J., Abe, A., *Ibid.*, **86**, 3563 (1964).
(5) Hutchinson, E., Bailey, L.G., *Z. Phys. Chem. (Frankfurt)* **21**, 30 (1959).
(6) "International Critical Tables," Vol. V, p. 108, McGraw-Hill, New York, 1928.
(7) Orwoll, R.A., Flory, P.J., *J. Amer. Chem. Soc.* **89**, 6822 (1967).
(8) Pflug, H.D., Benson, G.C., *Can. J. Chem.* **46**, 287 (1968).
(9) Pflug, H.D., Pope, A.E., Benson, G.C., J. Chem. Eng. Data **13**, 408 (1968).
(10) Pope, A.E., Pflug, H.D., Dacre, B., Benson, G.C., *Can. J. Chem.* **45**, 2665 (1967).
(11) Schaaffs, W., in "Molecular Acoustics," Vol. **5** of "Landolt-Börnstein New Series Group II," K.-H. Hellwege and A. M. Hellwege, Eds., p. 41, Springer Verlag, Berlin, 1967.
(12) Timmermans, J., "Physico-Chemical Constants of Pure Organic Compounds," Vols. **I** and **II**, Elsevier, New York, 1950 and 1965.

Received for review November 17, 1969. Accepted February 11, 1970. Issued as NRCC No. 11416.

Vapor-Liquid Equilibria of the Quinary System Hexane, Methylcyclopentane, Cyclohexane, Benzene, and Toluene

ROBERT M. WEATHERFORD[1] and MATTHEW VAN WINKLE
Department of Chemical Engineering, The University of Texas, Austin, Tex. 78712

Vapor-liquid equilibria were determined for the quinary system of hexane, methylcyclopentane, cyclohexane, benzene, and toluene at 760 mm of mercury. Ten data points were run for this system. Six additional points were taken on the five possible quaternaries. Nine of the required sets of binary equilibria were obtained from the literature, and the methylcyclopentane-cyclohexane data were experimentally determined by the authors. The Wilson equation was used to predict the experimental values; agreement was excellent.

BATCH FRACTIONATION to separate benzene and cyclohexane azeotrope indicated that the presence of the other three components enhanced the separation. Recent investigations (*6, 8, 16*) wherein accurate multicomponent polar and hydrogen-bonding data have been experimentally determined, have given opportunity to compare the capability of various two-parameter equations to predict highly nonideal multicomponent vapor-liquid equilibria data from binary data. A modified form of the Wilson (*17*) equation assuming ideal vapor mixtures and requiring only binary equilibrium and pure component data served this purpose satisfactorily. Because multicomponent data for different types of systems are not abundant, this investigation was initiated to determine experimental quinary data for a multitype hydrocarbon mixture. In addition, it was desirable to learn the capability of the equation to predict equilibrium data for multicomponent hydrocarbon mixtures at atmospheric pressure from binary data.

EXPERIMENTAL

Materials. The hexane and methylcyclopentane were Phillips pure grade (99 mole % minimum). The cyclohexane was obtained from the Phillips Petroleum Co. as the 99.5 weight % minimum purity grade. The benzene was certified ACS reagent grade solvent from the Fisher Scientific Co., which had been further purified by distillation by Hudson (*8*). The toluene was obtained from the Monsanto Co. Analysis by a GC-2A gas chromatograph indicated that the toluene and benzene exceeded 99.9 mole % purity, and these components were not further purified. The hex-

---

[1] Present address, Humble Oil and Refining Co., Baytown, Tex. 77520

ane, methylcyclopentane, and cylcohexane contained significant amounts of trace impurities, and were subjected to batch distillation at atmospheric pressure in a 30-plate Oldershaw column. Typically, 700 ml were charged to the still pot, and the column was operated at total reflux for 30 minutes, using a timed reflux of approximately 11 to 1. The overhead product was analyzed at various intervals by gas chromatography and the "heart" cut of the desired purity was taken for use in the equilibrium determinations. Composite sample analyses gave the following approximate purities: hexane, 99.74; methylcyclopentane, 99.86; cyclohexane, 99.98; benzene, 99.91; and toluene, 99.96 mole %. Table I compares the experimental and literature values for the boiling points and refractive indexes of the purified components.

Apparatus and Procedure. The pure component boiling points reported here were obtained by use of a modified Colburn still used by Willock (*16*). A similar still, but with slightly different heating coil arrangement and double capacity, was used in the determination of the vapor-liquid equilibrium data. This was the still described by Matocha (*9*) and Garrett (*3, 4*). Better flow control was obtained, which allowed more rapid attainment of equilibrium.

The experimental procedure consisted of weighing amounts of each component into a flask to give approximately the desired concentration. The total sample charged to the still amounted to 34 to 36 ml for each determination. After charging, the pressure was adjusted to 760 mm of mercury and the vaporizer chamber was heated to start the sample boiling. The other heaters were then slowly adjusted until the levels in the condensate receiver remained constant and steady-state conditions were reached. A few drops of liquid were maintained in the vaporizer tube and only vapor was introduced into the liquid in the residue chamber. When temperature, pressure, and constant flow rate were reached, the still was operated for 45 minutes to 1 hour before shutdown and sampling. The first few milliliters of both condensed vapor and liquid samples were discarded to prevent contamination with nonequilibrium material.

The samples were analyzed with a Beckman GC 2A gas chromatograph equipped with a 6-foot, general-purpose silicone column, a 1-mv Sargent SR recorder, and a Disc integrator.

The calibration technique described by Hudson (*7*) was followed in sample analyses, and his computer program for calculation of equilibrium values from chromatographic peak areas was used. The wide variation in composition of equilibrium samples required changes in attenuation and introduced some uncertainty in reproducibility. For this reason, both the equilibrium samples and their corresponding known samples were analyzed in duplicate. If any peak area did not reproduce within 2%, the sample was analyzed again. The mean of the two or three replicates was then used in calculating equilibrium values.

The detailed procedure used was designed to limit experimental error. In addition to error produced by unintentional deviation from this procedure, the estimated limits of error are:

Temperature.	±0.1° C
Pressure.	±0.2 mm of mercury
Composition.	±0.005 mole fraction

Table I. Comparison of Pure Component Properties

	Normal Boiling Points, ° C		Refractive Index (25° C)	
	Exptl.	Lit.	Exptl.	Lit.
Hexane	68.76	68.740 (*2*)	1.37238	1.37226 (*2*)
Methylcyclopentane	71.82	71.812 (*1*)	1.40700	1.40700 (*1*)
Cyclohexane	80.76	80.738 (*1*)	1.42355	1.42354 (*1*)
Benzene	80.06	80.100 (*1*)	1.4978	1.49792 (*1*)
Toluene	110.56	110.625 (*1*)	1.4941	1.49414 (*1*)

DISCUSSION AND CORRELATION OF EXPERIMENTAL RESULTS

Basically, the Wilson equation is a two-parameter model for predicting liquid phase activity coefficients. The general expression is

$$\ln \gamma_m = 1 - \ln\left[\sum_{j=1}^{k} x_j \Lambda_{mj}\right] - \sum_{i=1}^{k}\left[\frac{x_i \Lambda_{im}}{\sum_{j=1}^{k} x_j \Lambda_{ij}}\right] \qquad (1)$$

where the Λ's are defined by

$$\Lambda_{ij} = \frac{v_j}{v_i} \exp\left[\frac{-(\lambda_{ij} - \lambda_{ii})}{RT}\right] \qquad (2)$$

With the assumption of ideality in the gas phase, vapor and liquid equilibrium compositions may be calculated by

$$y_i = \frac{\gamma_i x_i P_i}{P_T} \qquad (3)$$

Multicomponent data in this study were predicted by use of a computer program, originally published by Prausnitz (*13*) and modified by Holmes (*6*) and Hudson (*7*). Gas phase ideality was assumed in these calculations.

Binary Data. The binary parameters expressed in Equation 2 were obtained from binary vapor-liquid equilibrium data using the computer method described by Holmes (*6*). The sources of the binary data and the resulting parameters are presented in Table II.

Data were available in the literature for all ten possible binaries. When data were reported on one system by more than one author, the data utilized in evaluating the parameters were selected, when possible, from the author who had reported data on other of the ten binary systems.

Table II. Binary Wilson Parameters

Binary	Binary Data Reference	Wilson Parameters	
		$\lambda_{12} - \lambda_{11}$	$\lambda_{12} - \lambda_{22}$
Hexane-methylcyclopentane	(*12*)	348.43	−219.53
Hexane-cyclohexane	(*12*)	496.96	−230.57
Hexane-benzene	(*10*)	279.52	134.35
Hexane-toluene	(*10*)	−121.00	383.89
Methylcyclopentane-cyclohexane	This study	327.06	−247.40
Methylcyclopentane-benzene	(*11*)	94.01	162.93
Methylcyclopentane-toluene	(*11*)	−465.89	1004.73
Cyclohexane-benzene	(*14*)	151.22	107.28
Cyclohexane-toluene	(*11*)	−446.10	981.96
Benzene-toluene	(*5*)	75.54	−88.83

Table III. Methylcyclopentane[a]-Cyclohexane Binary System

x_1	y_1	γ_1	γ_2	P, Mm Hg	T, ° C
0.102	0.131	1.015	0.999	760	79.68
0.477	0.541	0.994	1.009	760	76.17
0.771	0.814	0.998	1.010	760	73.65

[a] Component 1.

The data, with the exception of the cyclohexane-benzene binary at 759.9 mm of mercury, were reported at 760 mm of mercury. Parameters were calculated at both 759.9 and 760 mm of mercury for this binary; while the values of the parameters changed slightly, no difference resulted when both sets of parameters were used to predict a trial quinary point.

The data on the reported methylcyclopentane-cyclohexane binary system (*15*) appeared to be inconsistent and three points were experimentally determined in this laboratory (Table III). The reported data and experimental points were plotted (Figure 1). The equilibrium concentrations were plotted on the same curves, but our data showed a more consistent trend in activity coefficients. Therefore, they were used in determining the methylcyclopentane-cyclohexane parameters.

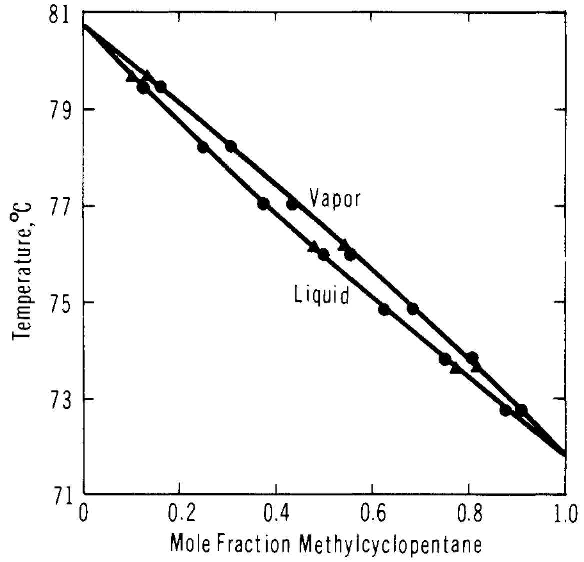

Figure 1. Methylcyclopentane-cyclohexane binary data

● (*15*)

▲ This investigation

Since the Wilson equation is consistent with the Gibbs-Duhem relationship, the agreement between experimental vapor compositions and those predicted through the use of the equation should indicate the consistency of the data. Using this criterion, the data used in determining binary parameters appear to be good and no other consistency tests were performed.

Quaternary Data. Since an extremely large number of data points would be necessary to describe a quinary system completely, selected points were chosen that were significant in describing the system and were capable of accurate analysis. One point for each of the five possible quaternary systems was determined and then a quantity of the fifth component was added to determine its effect on experimental and predicted vapor-liquid equilibrium values. The quaternary data are presented in Table IV. With the exception of one point, QUAT 3, disagreement between predicted and experimental equilibrium values was not significantly greater than the estimated cumulative limits of experimental error of 0.010 mole fraction. Since the reproducibility in previous analyses had been excellent, the usual procedure of analyzing duplicates was not used for this point. As the agreement between predicted and experimental values was significantly less for QUAT 3 than for any of the other quaternaries, another point, QUAT 6, was run with approximately the same charge composition as the deviant. The usual duplicate analyses procedure was used, and the

Table IV. Hexane (Hex)-Methylcyclopentane (Mcp)-Cyclohexane (Ch) Benzene (Bz)-Toluene (Tol) Quaternary Data

		Experimental Values				Wilson Eq. Predictions		
Component	Run No.	T, °C	x	y	γ	T, °C	y	γ
Hex	QUAT 1	80.88	0.181	0.273	1.047	80.32	0.281	1.096
Mcp			0.223	0.297	1.015		0.301	1.047
Ch			0.263	0.278	1.055		0.277	1.065
Tol			0.333	0.152	1.153		0.141	1.093
Hex	QUAT 2	81.65	0.134	0.235	1.183	81.90	0.246	1.235
Mcp			0.196	0.290	1.106		0.290	1.096
Bz			0.278	0.307	1.052		0.303	1.031
Tol			0.392	0.169	1.062		0.161	1.006
Hex	QUAT 3	83.24	0.115	0.207	1.168	83.54	0.227	1.270
Ch			0.208	0.271	1.210		0.250	1.106
Bz			0.325	0.382	1.066		0.371	1.029
Tol			0.352	0.140	0.931		0.152	0.995
Hex	QUAT 4	73.60	0.152	0.191	1.080	73.71	0.195	1.098
Mcp			0.208	0.227	1.034		0.233	1.055
Ch			0.267	0.228	1.061		0.220	1.024
Bz			0.373	0.355	1.167		0.353	1.156
Mcp	QUAT 5	84.15	0.142	0.234	1.141	85.00	0.234	1.116
Ch			0.196	0.244	1.125		0.247	1.110
Bz			0.303	0.356	1.040		0.357	1.016
Tol			0.359	0.166	1.047		0.162	0.994
Hex	QUAT 6	82.61	0.123	0.228	1.222	83.25	0.240	1.264
Ch			0.208	0.252	1.142		0.248	1.105
Bz			0.320	0.365	1.056		0.364	1.032
Tol			0.348	0.155	1.063		0.149	0.998

Δy_H av. dev. = 0.011 Δy_H max. dev. = 0.020
Δy_M av. dev. = 0.003 Δy_M max. dev. = 0.006
Δy_C av. dev. = 0.007 Δy_C max. dev. = 0.021
Δy_B av. dev. = 0.005 Δy_B max. dev. = 0.009
Δy_T av. dev. = 0.008 Δy_T max. dev. = 0.012
ΔT av. dev. = 0.47° C ΔT max. dev. = 0.85° C

difference between experimental and predicted values was no greater than that of the other quaternaries. In all points other than QUAT 3, duplicate analyses were made.

Quinary Data. Information derived from the quinary data and a summary of the deviations between experimental and predicted vapor composition for these data are given in Table V. Agreement was within the estimated limits of experimental error. As in the quaternary data, the predicted equilibrium temperature was consistently higher than that observed. The average temperature difference was approximately 0.5° C.

Points QUIN 1 through QUIN 5 were determined to obtain the effect of the fifth component on the quaternaries. These points indicate that no loss in the accuracy of the predictive equation resulted from expansion to the quinary system.

QUIN 7 contained approximately the relative amounts of methylcyclopentane and benzene reported in the binary

Table V. Hexane (Hex)-Methylcyclopentane (Mcp)-Cyclohexane (Ch)-Benzene (Bz)-Toluene (Tol) Quinary Data

		Experimental Values				Wilson Eq. Predictions		
Component	Run No.	T, ° C	x	y	γ	T, ° C	y	γ
Hex	QUIN 1	78.27	0.186	0.274	1.105	78.53	0.284	1.135
Mcp			0.257	0.328	1.049		0.333	1.058
Ch			0.118	0.118	1.081		0.118	1.066
Bz			0.153	0.167	1.153		0.156	1.067
Tol			0.286	0.113	1.094		0.110	1.055
Hex	QUIN 2	84.41	0.129	0.243	1.183	84.64	0.249	1.200
Mcp			0.159	0.224	0.972		0.252	1.086
Ch			0.132	0.168	1.138		0.162	1.095
Bz			0.100	0.128	1.124		0.116	1.011
Tol			0.480	0.237	1.107		0.221	1.026
Hex	QUIN 3	80.45	0.132	0.219	1.164	81.09	0.232	1.209
Mcp			0.095	0.134	1.093		0.135	1.073
Ch			0.218	0.241	1.115		0.239	1.086
Bz			0.253	0.284	1.110		0.273	1.046
Tol			0.302	0.122	1.039		0.122	1.013
Hex	QUIN 4	80.50	0.069	0.112	1.133	81.32	0.124	1.233
Mcp			0.166	0.238	1.104		0.239	1.085
Ch			0.215	0.241	1.128		0.238	1.088
Bz			0.261	0.290	1.097		0.282	1.042
Tol			0.288	0.119	1.062		0.116	1.008
Hex	QUIN 5	76.58	0.132	0.192	1.146	77.28	0.202	1.178
Mcp			0.155	0.202	1.123		0.194	1.060
Ch			0.228	0.213	1.061		0.220	1.072
Bz			0.333	0.336	1.129		0.331	1.086
Tol			0.152	0.057	1.096		0.053	0.998
Hex	QUIN 6	77.98	0.067	0.105	1.180	78.68	0.111	1.222
Mcp			0.081	0.106	1.087		0.106	1.064
Ch			0.360	0.362	1.092		0.365	1.078
Bz			0.359	0.374	1.111		0.371	1.081
Tol			0.132	0.053	1.121		0.047	0.977
Hex	QUIN 7	75.71	0.066	0.087	1.070	75.93	0.089	1.084
Mcp			0.583	0.676	1.030		0.689	1.042
Ch			0.077	0.070	1.064		0.069	1.035
Bz			0.089	0.095	1.225		0.085	1.082
Tol			0.186	0.072	1.174		0.069	1.114
Hex	QUIN 8	78.83	0.098	0.153	1.145	79.45	0.162	1.196
Mcp			0.149	0.206	1.115		0.200	1.065
Ch			0.269	0.272	1.072		0.278	1.075
Bz			0.265	0.282	1.104		0.277	1.065
Tol			0.219	0.088	1.100		0.083	1.013
Hex	QUIN 9	78.54	0.186	0.273	1.088	78.96	0.273	1.132
Mcp			0.255	0.339	1.084		0.335	1.058
Ch			0.127	0.126	1.060		0.128	1.067
Bz			0.128	0.137	1.125		0.131	1.061
Tol			0.304	0.125	1.131		0.119	1.059
Hex	QUIN 10	76.02	0.130	0.178	1.092	76.61	0.182	1.099
Mcp			0.471	0.563	1.052		0.572	1.051
Ch			0.060	0.055	1.052		0.055	1.048
Bz			0.109	0.115	1.197		0.105	1.079
Tol			0.230	0.089	1.166		0.085	1.089

Δy_H av. dev. = 0.007
Δy_M av. dev. = 0.006
Δy_C av. dev. = 0.003
Δy_B av. dev. = 0.008
Δy_T av. dev. = 0.005
ΔT av. dev. = 0.52° C

Δy_H max. dev. = 0.013
Δy_M max. dev. = 0.013
Δy_C max. dev. = 0.007
Δy_B max. dev. = 0.012
Δy_T max. dev. = 0.016
ΔT max. dev. = 0.82° C

azeotrope (0.125 mole fraction benzene). A mixture of methylcyclopentane and benzene of the azeotropic composition was made and a still charge composed of 71% by volume of this mixture and 29% of the other three components was then run. The resulting normalized (based on the sum of the azeotropic components' compositions) liquid and vapor benzene compositions were 0.132 and 0.123 mole fraction, respectively. These relative concentrations are not significantly different from those of the azeotrope when the estimated errors are considered.

Three points—QUIN 6, QUIN 8, and QUIN 9—were run in a similar manner to examine the cyclohexane-benzene azeotrope. These points contained total azeotrope charges of 71, 50, and 22 volume %, respectively. For QUIN 6, the resulting normalized liquid and vapor compositions were 0.499 and 0.508 mole fraction as compared to the azeotropic benzene concentration of 0.498 mole fraction. The corresponding liquid and vapor values were 0.496 and 0.509 mole fraction for QUIN 8. In QUIN 9, the resulting benzene liquid concentration was 0.502 mole fraction, the vapor being 0.521 mole fraction. These values represent a relative volatility of the benzene and cyclohexane of 1.04 for QUIN 6, 1.05 for QUIN 8, and 1.08 for QUIN 9. These points also indicate that the charge compositions were slightly different from those exhibited by the azeotrope. When this difference is considered with the possibility of experimental error, it is again difficult to evaluate the significance of the effect of the addition of the other system components on the separability of this azeotrope.

Batch Distillation of Quinary System. Since the three quinary equilibrium points did not significantly demonstrate the effect of the other components on the separability of the cyclohexane-benzene binary azeotrope by distillation, an atmospheric batch distillation was performed with a 30-plate Oldershaw column with the charge containing the approximate relative azeotropic concentrations of benzene and cyclohexane. The charge consisted of 100 ml each of hexane, methylcyclopentane, and benzene; 121 ml of cyclohexane; and 200 ml of toluene. After a 15-minute period of total reflux, fractions containing volumes equivalent to the charge quantities of the four lightest components were taken. Cuts of 40, 30, 20, and 10 ml were taken, using an operating reflux ratio of 10 to 1 based on an internal timer, and were combined to make the first three fractions. For the fourth fraction, the corresponding cuts contained 50, 40, 20, and 11 ml. Ten-minute intermediate periods of total reflux operation were provided between each cut. The four fractions were then analyzed by gas chromatograph (Table VI). On Figure 2, the compositions of benzene and cyclohexane in the fractions are presented. While these results are probably less accurate than the equilibrium data, because only single analyses were made, it is apparent that the benzene-cyclohexane separation by batch fractionation is greatly enhanced by the addition of the other quinary components.

CONCLUSIONS

The Wilson equation accurately predicts multicomponent vapor-liquid equilibria at atmospheric pressure in the five possible quaternaries and the quinary system of hexane, methylcyclopentane, cyclohexane, benzene, and toluene in the concentration ranges examined.

In the relatively ideal hydrocarbon quinary system of this study, the temperatures predicted by the Wilson equation were consistently high by approximately 0.5° C. The addition of hexane, methylcyclopentane, and toluene significantly enhances the separation of the cyclohexane-benzene azeotrope by fractional distillation.

Table VI. Batch Distillation Data of Quinary System

Volume Collected Overhead, Ml	Mole Fraction				
	Hexane	Mcp	Benzene	Ch	Toluene
100	0.631	0.241	0.126	0.002	0.000
200	0.216	0.547	0.205	0.032	0.000
300	0.004	0.145	0.474	0.377	0.000
420	0.002	0.003	0.253	0.525	0.218

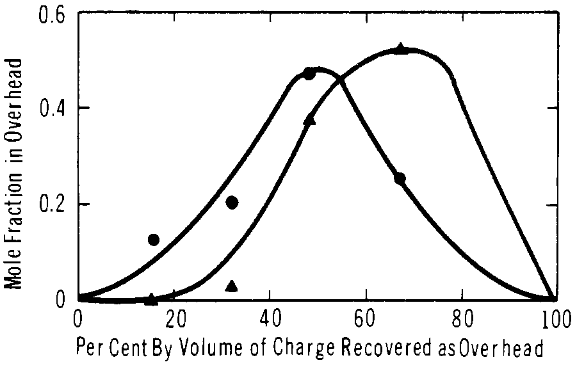

Figure 2. Overhead compositions of benzene and cyclohexane in batch fractionation of quinary system

▲ Cyclohexane
● Benzene

ACKNOWLEDGMENT

The authors express appreciation for the financial support contributed by the Engineering Foundation at the University of Texas at Austin and to the Monsanto Co. for materials.

NOMENCLATURE

k = number of components in system
m = general component specification
P_i = vapor pressure of component i, mm of mercury
P_T = system pressure, mm of mercury
R = gas constant
T = absolute temperature, ° K
v_i = molar volume of component i, cc/gram-mole
x_i = mole fraction of i in liquid
Y_i = mole fraction of i in vapor
γ_i = liquid phase activity coefficient of i
λ = Wilson equation interaction parameter

LITERATURE CITED

(1) Dreisbach, R.R., *Advan. Chem. Ser.* No. **15** (1955).
(2) *Ibid.*, No. **22** (1959).
(3) Garrett, G.R., M.S. thesis, University of Texas, Austin, 1968.
(4) Garrett, G.R., Van Winkle, M., J. CHEM. ENG. DATA **14**, 302 (1969).
(5) Heertjies, P.M., *Chem. Proc. Eng.* **41**, 385 (1960).
(6) Holmes, M.J., Van Winkle, M., *Ind. Eng. Chem.* **62**, 21 (1970).
(7) Hudson, J.W., Ph.D. dissertation, University of Texas, Austin, 1969.
(8) Hudson, J.W., Van Winkle, M., J. CHEM. ENG. DATA **14**, 310 (1969).
(9) Matocha, I.R., unpublished M.S. thesis, University of Texas, Austin, 1969.
(10) Myers, H.S., *Ind. Eng. Chem.* **47**, 2215 (1955).
(11) *Ibid.*, **48**, 1104 (1956).
(12) Myers, H.S., *Petrol. Refiner* **36**, No. 3, 175 (1957).
(13) Prausnitz, J.M., Eckert, C.A., Orye, R.V., O'Connell, J.P., "Computer Calculations of Multicomponent Vapor-Liquid Equilibria," Prentice-Hall, Englewood Cliffs, N. J. 1967.
(14) Richards, A.R., Hargreaves, E., *Ind. Eng. Chem.* **36**, 805 (1944).
(15) Susarev, M.P., Lyzlova, R.V., *Zh. Fiz. Khim.* **36**, 437 (1962).
(16) Willock, J.M., M.S. thesis, University of Texas, Austin, 1969.
(17) Wilson, G.M., *J. Amer. Chem. Soc.* **86**, 127 (1964).

RECEIVED for review January 7, 1970. Accepted April 27, 1970.