$$-\frac{2\left(\frac{d\theta}{d\eta}\right)_0}{N_{Pr}\,Q_6}\left\{1-\frac{(e^{N_{Rew}N_{Pr}}-1)}{N_{Pr}N_{Rew}}+(e^{N_{Rew}}-1)\left(\frac{1}{2}-\frac{1}{N_{Rew}}\right)-\frac{(e^{N_{Rew}}-1)}{N^2{}_{Pr}N^2{}_{Rew}}\left[e^{N_{Rew}N_{Pr}}\,(N_{Rew}N_{Pr}-1)+1\right]+\frac{(e^{N_{Rew}(1+N_{Pr})}-1)}{N_{Rew}(1+N_{Pr})}\right\}$$

and the values of $Q_n$ ($n = 1, \ldots, 6$) are given by

$$Q_1=\frac{1}{N_{Rew}N^2{}_{Pr}}+\frac{(e^{N_{Rew}}-1)}{N^2{}_{Rew}N^3{}_{Pr}}+\frac{1}{N_{Rew}\,(1-N_{Pr})}+\frac{1}{N_{Rew}N_{Pr}}$$

$$Q_2=-\left(\frac{1}{N_{Pr}}+\frac{(e^{N_{Rew}}-1)}{N_{Rew}N^2{}_{Pr}}\right)$$

$$Q_3=-\left(\frac{1}{N_{Rew}N^2{}_{Pr}}+\frac{e^{N_{Rew}}-1}{N^2{}_{Rew}N^3{}_{Pr}}+\frac{1}{N_{Rew}N_{Pr}}\right)$$

$$Q_4=\frac{e^{N_{Rew}}-1}{2N_{Pr}}$$

$$Q_5=\frac{1}{N_{Rew}(1-N_{Pr})}$$

$$Q_6=(N_{Rew}+2)+e^{N_{Rew}}\,(N_{Rew}-2)$$

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# Vapor Phase Activity Coefficients and Standard State Hypothetical Vapor Fugacities for Hydrocarbons

**DWIGHT S. HOFFMAN, J. REED WELKER, V. N. P. RAO, and JAMES H. WEBER**
**University of Idaho, Moscow, Idaho**

In an earlier paper Hoffman et al. (*1*) discussed a method for predicting the vaporization equilibrium ratios, defined as $y/x$, for the components of a multicomponent mixture through use of standard state liquid fugacities, standard state vapor fugacities, liquid phase activity coefficients, and vapor phase activity coefficients. As used here and throughout this paper, standard state fugacity means the fugacity of a pure component at the temperature and pressure of the system. The relationship among these variables has been shown to be

$$K_i' = \frac{\gamma_i^L}{\gamma_i^v}\frac{1}{Z_i}\frac{P_{vi}}{P} \tag{1}$$

where

$$\gamma_i^L = \frac{(f_i)_L}{(f_i^\circ)_L\,x_i} \tag{2}$$

and

$$\gamma_i^v = \frac{(f_i)_v}{(f_i^\circ)_v\,y_i} \tag{3}$$

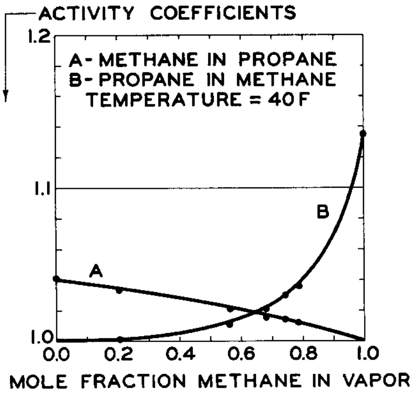

**Fig. 1. Vapor phase activity coefficients in the methane-propane system.**

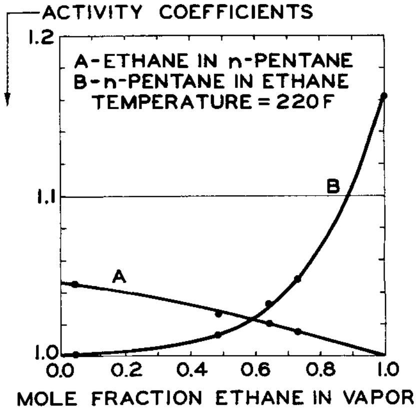

**Fig. 2. Vapor phase activity coefficients in the ethane-*n*-pentane system.**

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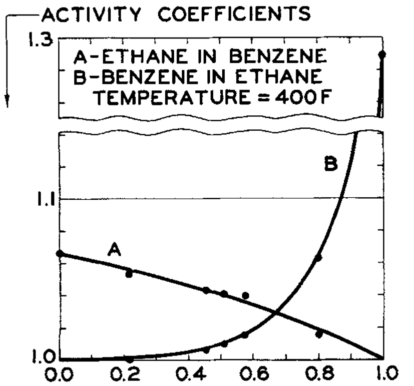

**Fig. 3. Vapor phase activity coefficients in the ethane-benzene system.**

$$Z_i = \frac{\nu_i^\circ}{(f_i^\circ)_L / P_{v_i}} \tag{4}$$

In Equation (4)

$$\nu_i^\circ = \frac{(f_i^\circ)_v}{P} \tag{5}$$

and it represents the fugacity coefficient of a pure vapor at standard state temperature and pressure.

In the paper by Hoffman et al. (*1*), methods were presented for evaluating the fugacities of the components of a multicomponent mixture and for evaluating the standard state fugacities of hypothetical liquids.

It is the purpose of this paper to present a method for evaluating the standard state fugacities of hypothetical vapors, through use of vapor phase activity coefficients, and to disclose these values for propane, *n*-pentane, and benzene; to present coincident values of the vaporization equilibrium constants; and to show correlations of these values by means of a generalized fugacity coefficient chart and generalized Z factors.

The use of vapor phase activity coefficients, obtained from the fugacities of the components in a mixture and the standard state fugacities of the components, is not limited to the prediction of vaporization equilibrium ratios. Bierlein and Kay (*2*) in discussing the testing of vapor-liquid equilibrium data for binary systems have warned that the Gibbs-Duhem relation must be applied simultaneously to both phases rather than the liquid phase alone. If not, more uncertainties will be created than will be eliminated; this is especially true at high pressures.

## VAPOR PHASE ACTIVITY COEFFICIENTS FOR THE MORE VOLATILE COMPONENT

In the work described here, use was made of the vapor-liquid equilibrium data of Sage, Lacey, and Schaafsma (*3*) and Akers, Burns, and Fairchild (*4*) for the methane-propane system; of Reamer, Sage, and Lacey (*5*) for the ethane-*n*-pentane system; of Kay and Nevens (*6*) for the ethane-benzene system; of Sage and Lacey (*7*) for the propane-*n*-pentane system; and of Glanville, Sage, and Lacey (*8*) for the propane-benzene system.

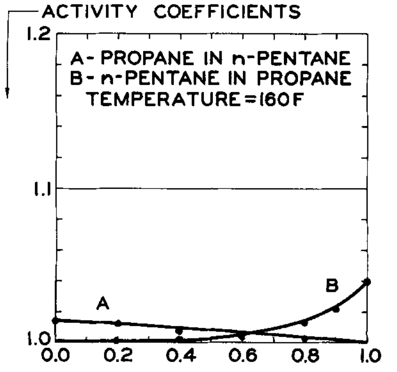

**Fig. 4. Vapor phase activity coefficients in the propane-*n*-pentane system.**

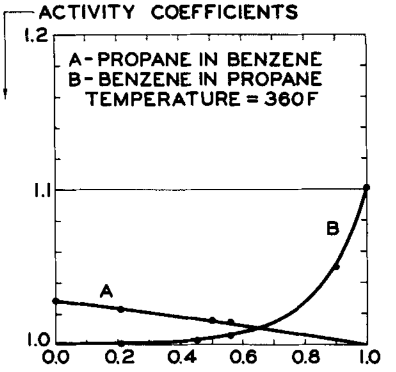

**Fig. 5. Vapor phase activity coefficients in the propane-benzene system.**

Hoffman et al. (*1*) have previously described the application of equations developed by Black (*9*) to give

$$\ln \frac{(f_i)_v}{P y_i} = \frac{P}{RT}\left(\tau_i^+ - \sigma_i\right) \tag{6}$$

for the evaluation of the fugacities of the components in a vapor mixture, and

$$\ln \nu_i^\circ = \frac{P}{RT}\left(-\sigma_i^\circ\right) \tag{7}$$

for the evaluation of the fugacity coefficient of a pure vapor. Combination of Equations (3), (5), (6), and (7) gives

$$\ln \gamma_i^v = \frac{P}{RT}\left(\tau_i^+ - \sigma_i + \sigma_i^\circ\right) \tag{8}$$

Equation (8) was used in this work to evaluate the vapor phase activity coefficients of the more volatile component of each of the binary mixtures studied.

Note that Equation (7), and hence Equation (8), is applicable to the more volatile component of a binary vapor mixture which is in equilibrium with the corresponding liquid phase, but it is not applicable to the less volatile component of the vapor mixture. This is because for these systems where no azeotropes exist, the more vola-

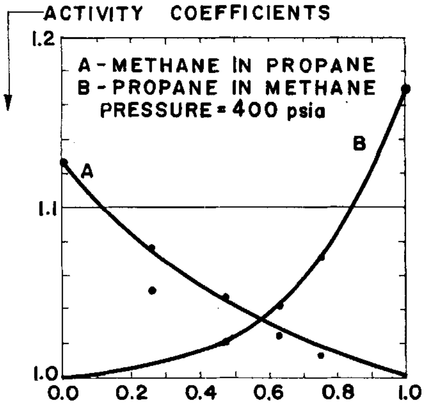

**Fig. 1*a*. Vapor phase activity coefficients in the methane-propane system.**

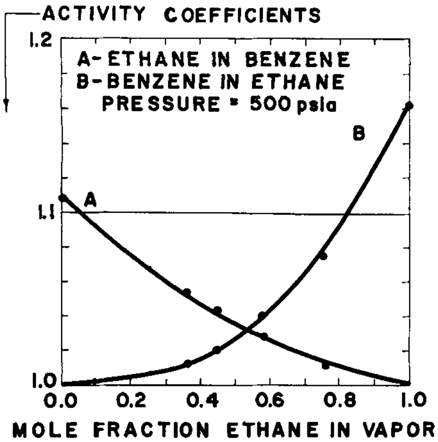

Fig. 3*a*. Vapor phase activity coefficients in the ethane-benzene system.

tile component can exist as a pure vapor at the temperature and pressure of the system; on the contrary, the less volatile component in the vapor mixture cannot exist as a pure vapor at the temperature and pressure of the system.

The vapor phase activity coefficients for the more volatile components of all of the systems studied were plotted against vapor-phase composition, with the temperature held constant and the pressure allowed to vary from the vapor pressure of the less volatile component to the vapor pressure of the more volatile component. For each system, several isothermal plots were prepared.

## VAPOR PHASE ACTIVITY COEFFICIENTS FOR THE LESS VOLATILE COMPONENT

The vapor phase activity coefficients of the less volatile components of the mixtures were evaluated through use of the van Laar equation:

$$\log \gamma_i^v = \frac{a^2_{ij}}{(1 + a^2_{ij} y_i / a^2_{ji} y_j)^2} \tag{9}$$

The van Laar equation is, of course, one solution of the Gibbs-Duhem equation for two components

$$\left[ y_i \frac{d \ln \gamma_i^v}{dy_i} = y_j \frac{d \ln \gamma_j^v}{dy_j} \right]_{T,P} \tag{10}$$

wherein both temperature and pressure are constant. For the case of variable pressure and constant temperature the Gibbs-Duhem equation must be modified as follows:

$$\left[ y_i \frac{d \ln \gamma_i^v}{dy_i} = y_j \frac{d \ln \gamma_j^v}{dy_j} + \frac{\Delta v}{RT} \frac{dP}{dy_i} \right]_T \tag{11a}$$

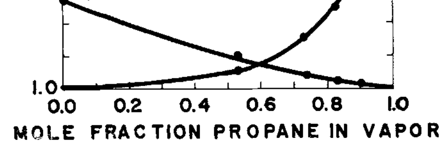

Fig. 4*a*. Vapor phase activity coefficients in the propane-*n*-pentane system.

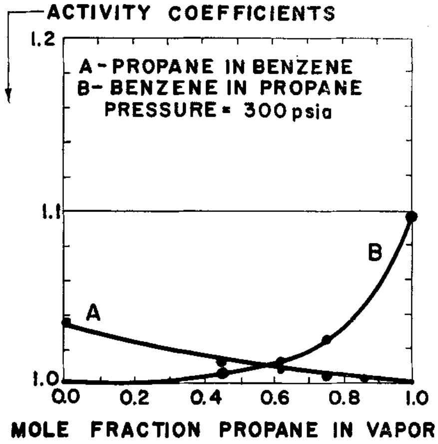

Fig. 5*a*. Vapor phase activity coefficients in the propane-benzene system.

where $\Delta v$ is the volume change upon mixing the separate pure components, all at the same temperature and pressure corresponding to the composition of the mixture, to form 1 mole of solution. For the binary systems studied in this work, use was made of Equation (11*a*) by neglecting the volume change upon mixing. Consequently, Equation (9) was fit to the isothermal vapor phase activity coefficients of the more volatile component of each of the binary mixtures studied. The $a^2_{ij}$ constants were then used to calculate the isothermal vapor phase activity coefficients of the less volatile component of the binary mixtures. Figures 1, 2, 3, 4, and 5 show isothermal vapor phase activity coefficients for both components of the mixtures studied.

## ISOBARIC VAPOR PHASE ACTIVITY COEFFICIENTS

In order to substantiate the neglect of $\Delta v$ in the use of Equation (11*a*), the vapor-liquid data for the methane-propane system, the ethane-benzene system, the propane-*n*-pentane system, and the propane-benzene system were treated as constant pressure, variable temperature data.

The Gibbs-Duhem relationship between the activity coefficients for the case of a constant pressure and variable pressure is

$$\left[ y_i \frac{d \ln \gamma_i^v}{dy_i} = y_j \frac{d \ln \gamma_j^v}{dy_j} + \frac{\Delta H}{RT^2} \frac{dT}{dy_i} \right]_P \tag{11b}$$

where $\Delta H$ is the enthalpy change upon mixing the separate pure components, all at the same temperature and pressure corresponding to the composition of the mixture, to form 1 mole of solution.

In treating the data isobarically, the same sort of procedure was followed as was used in the isothermal treatment described above. The vapor phase activity coefficients of the more volatile component of a binary mixture were

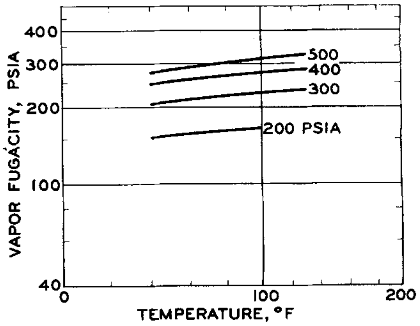

Fig. 6 Standard state hypothetical vapor fugacity of propane.

Table 1. Standard State Hypothetical Vapor Fugacities

| | Pressure | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | 200 lb./sq. in. abs. | | | 300 lb./sq. in. abs. | | | 400 lb./sq. in. abs. | | | 500 lb./sq. in. abs. | | |
| T, °F.* | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD |
| | Methane (1)—propane (2) | | | | | | | | | | | |
| 130 | | | | 231.7 | 231.6 | 0.0 | 283.8 | 283.0 | 0.3 | 323.4 | 320.9 | 0.8 |
| 100 | 165.0 | 165.0 | 0.0 | 227.1 | 222.7 | 2.0 | 270.4 | 267.5 | 1.1 | 309.5 | 303.6 | 1.9 |
| 70 | 158.9 | 158.3 | 0.4 | 214.0 | 209.8 | 2.0 | 256.3 | 249.8 | 2.6 | 290.5 | 280.0 | 3.7 |
| 40 | 153.9 | 152.3 | 1.0 | 204.8 | 197.9 | 3.5 | 245.0 | 235.0 | 4.2 | 274.7 | 259.8 | 5.7 |

| | Pressure | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | 200 lb./sq. in. abs. | | | 300 lb./sq. in. abs. | | | 400 lb./sq. in. abs. | | | 500 lb./sq. in. abs. | | |
| T, °F.* | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD |
| | Propane (1)—*n*-pentane (2) | | | | | | | | | | | |
| 280 | 157.1 | 157.2 | 0.1 | 212.0 | 204.3 | 3.7 | 252.3 | 250.8 | 0.6 | 283.9 | 278.4 | 1.9 |
| 240 | 152.1 | 151.7 | 0.2 | 202.8 | 201.2 | 0.8 | 239.2 | 235.0 | 1.8 | 267.5 | 256.9 | 4.1 |
| 220 | 149.6 | 148.8 | 0.5 | 196.9 | 195.5 | 0.7 | 232.0 | 226.5 | 2.4 | 255.8 | 244.1 | 4.8 |
| 200 | 147.3 | 146.0 | 0.9 | 191.9 | 188.5 | 1.8 | 224.2 | 216.2 | 3.7 | 245.5 | 231.0 | 6.3 |

| | Pressure | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | 200 lb./sq. in. abs. | | | 500 lb./sq. in. abs. | | | 600 lb./sq. in. abs. | | | 800 lb./sq. in. abs. | | |
| T, °F.* | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD |
| | Ethane (1)—benzene (2) | | | | | | | | | | | |
| 420 | 289.7 | 288.4 | 0.5 | 331.0 | 328.3 | 0.9 | 369.8 | 363.7 | 1.7 | 409.4 | 398.3 | 2.8 |
| 400 | 287.0 | 285.2 | 0.6 | 323.4 | 319.1 | 1.3 | 356.7 | 348.4 | 2.3 | 388.6 | 375.4 | 3.5 |
| 360 | 275.9 | 271.0 | 1.8 | 308.0 | 298.8 | 3.1 | 340.9 | 326.3 | 4.5 | 346.6 | 325.9 | 6.4 |
| 300 | 251.4 | 243.3 | 3.3 | 278.6 | 265.4 | 5.0 | 298.9 | 280.9 | 6.4 | 291.9 | 267.8 | 9.0 |

| | Pressure | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | 400 lb./sq. in. abs. | | | 300 lb./sq. in. abs. | | | 400 lb./sq. in. abs. | | | 500 lb./sq. in. abs. | | |
| T, °F.* | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD | $(f^\circ_2)^T_v$ | $(f^\circ_2)^P_v$ | o/oD |
| | Propane (1)—benzene (2) | | | | | | | | | | | |
| 360 | 163.5 | 163.5 | 0.0 | 224.2 | 223.7 | 0.5 | 272.3 | 270.8 | 0.6 | 309.8 | 307.0 | 0.9 |
| 320 | 160.8 | 160.2 | 0.3 | 215.1 | 213.6 | 1.6 | 257.9 | 254.4 | 1.4 | 289.7 | 283.7 | 2.1 |
| 280 | 153.0 | 151.9 | 0.8 | 201.0 | 198.3 | 2.7 | 236.4 | 231.0 | 2.3 | 261.0 | 252.0 | 3.5 |
| 240 | 145.4 | 141.8 | 2.6 | 187.7 | 183.0 | 4.6 | 218.2 | 210.4 | 3.7 | 236.1 | 216.2 | 9.2 |

* $(f^\circ_2)^T_v$ = standard state hypothetical vapor fugacity from isothermal data.
$(f^\circ_2)^P_v$ = standard state hypothetical vapor fugacity from isobaric data.
o/oD = percentage difference based on $(f^\circ_2)^P_v$.

plotted as constant pressure lines; Equation (9) was used to evaluate the isobaric van Laar constants; Equation (11*b*) was then used, with $\Delta H$ neglected, to calculate the vapor phase activity coefficients of the less volatile component. Figures 1*a*, 3*a*, 4*a*, and 5*a* show the isobaric vapor phase activity coefficients for both components of the mixtures studied.

## HYPOTHETICAL VAPOR FUGACITIES

The term *standard state hypothetical vapor* is used to designate the less volatile component of a binary mixture as a pure vapor at the temperature and pressure of the system, where the pure vapor cannot actually exist.

Having determined the vapor phase activity coefficients of the less volatile component of each of the binary mixtures, as described above, and having determined the fugacity coefficients of these components in the vapor mixtures, one can determine the fugacity coefficients of these components in the standard state hypothetical vapor state by

$$\nu_i^\circ = \frac{(f_i)_v}{Py_i} \Big/ \gamma_i^v \qquad (12)$$

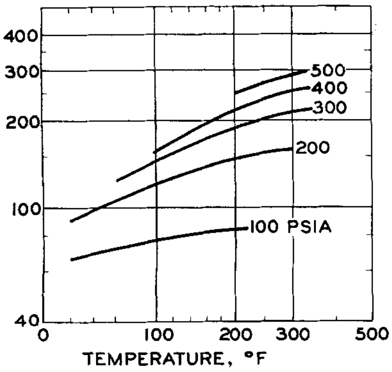

Fig. 7. Standard state hypothetical vapor fugacity of *n*-pentane.

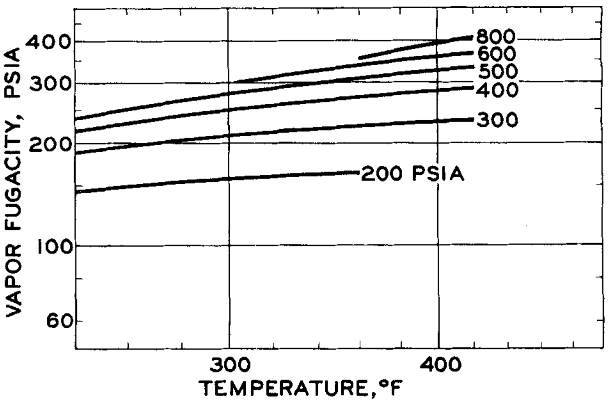

Fig. 8. Standard state hypothetical vapor fugacity of benzene.

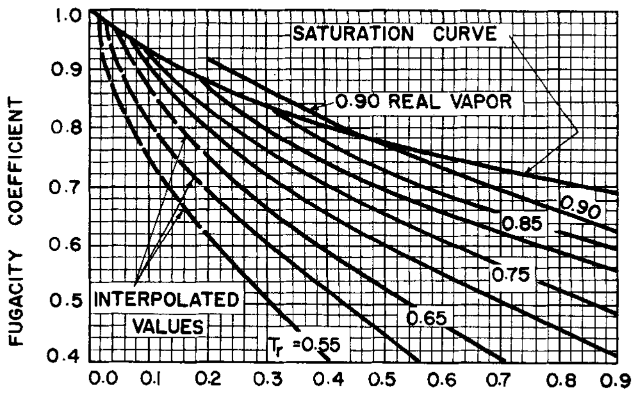

Fig. 9. Generalized correlation of fugacity coefficients.

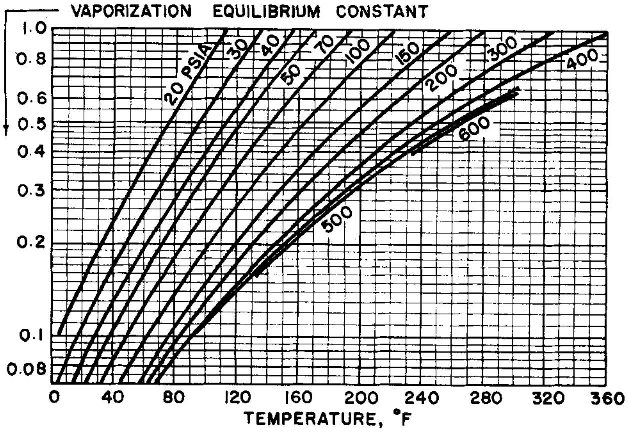

Fig. 11. Vaporization equilibrium constants for *n*-pentane.

The fugacity coefficients for propane, *n*-pentane, and benzene in the hypothetical standard state were converted to standard state hypothetical fugacities.

The values, which were obtained through the isobaric treatment of the binary systems studied, for the standard state hypothetical vapor fugacities for ethane, pentane, and benzene are shown in Table 1. A comparison of fugacity values obtained for the same components from the same systems with the isothermal treatment is also shown in Table 1.

Average values of the fugacities obtained are shown in Figures 6, 7, and 8 and reported in tabular form in Tables 2, 3, and 4.* The corresponding fugacity coefficients for propane, *n*-pentane, and benzene in the hypothetical vapor state were correlated, through use of a generalized plot, as a function of reduced pressure and reduced temperature. Figure 9 shows the generalized correlation.

## VAPORIZATION EQUILIBRIUM CONSTANTS

The vaporization equilibrium constant is defined as

$$K_i = \frac{(f_i°)_L}{(f_i°)_v} \tag{13}$$

Values for the vaporization equilibrium constants of the three components studied, in the hypothetical vapor regions, were evaluated through use of Equation (13). These values are shown in Figures 10, 11, and 12 and reported tabular form in Tables 2, 3, and 4.* Values for the standard state real liquid fugacities which were used in Equation (13) were obtained from the generalized data of Lydersen, Greenkorn, and Hougen (*10*).

Combination of Equations (4), (5), and (13) gives

$$Z_i = \frac{1}{K_i}\frac{P_{v_i}}{P} \tag{14}$$

Ehrett, Weber, and Hoffman (*11*) have shown that Z factors defined by Equation (14) may be correlated, in a generalized plot, as a function of reduced temperature and reduced pressure. Z factors for propane, *n*-pentane, and benzene, in the hypothetical vapor regions, were evaluated through use of Equation (14). The generalized correlation of these values is shown in Figure 13.

## DISCUSSION AND CONCLUSION

Previous evaluations of standard state hypothetical vapor fugacities, and correlations thereof, have been made by Gamson and Watson (*12*), Smith and Watson (*13*), and Prausnitz (*14*).

Gamson and Watson (*12*) obtained a generalized correlation of the fugacity coefficients of several gases in the real state and extrapolated the fugacity coefficient curves into the hypothetical region. Smith and Watson (*13*) revised the extrapolations of Gamson and Watson (*12*).

Hoffman and Weber (*15*) have previously evaluated fugacities and fugacity coefficients for benzene, using the generalized fugacity coefficients of Gamson and Watson (*12*) for the hypothetical vapor region. The values of fugacity and fugacity coefficients for benzene in the hypothetical vapor region obtained in the present work are larger than those previously obtained. The difference be-

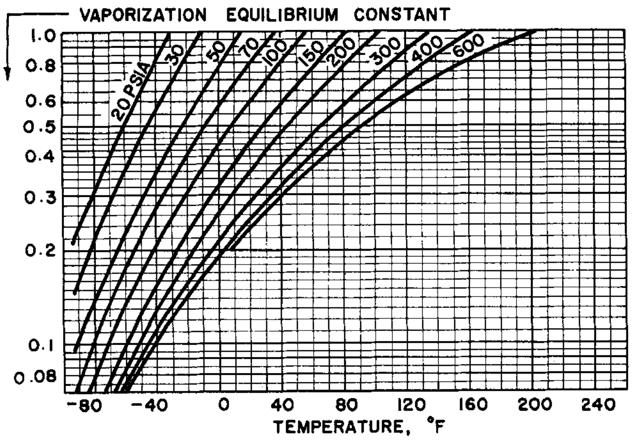

Fig. 10. Vaporization equilibrium constants for propane.

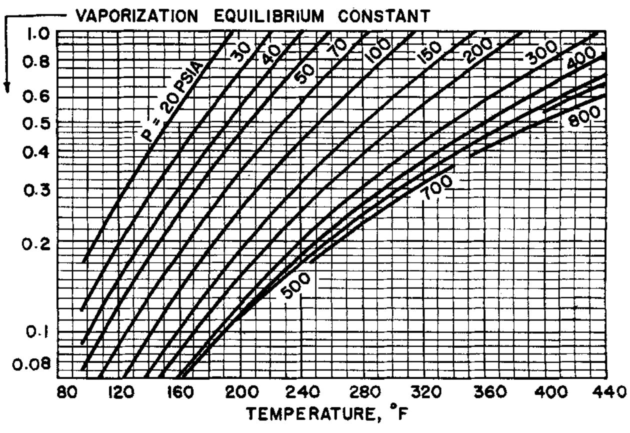

Fig. 12. Vaporization equilibrium constants for benzene.

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* Tabular material has been deposited as document 8084 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for $1.25 for photoprints or 35-mm. microfilm.

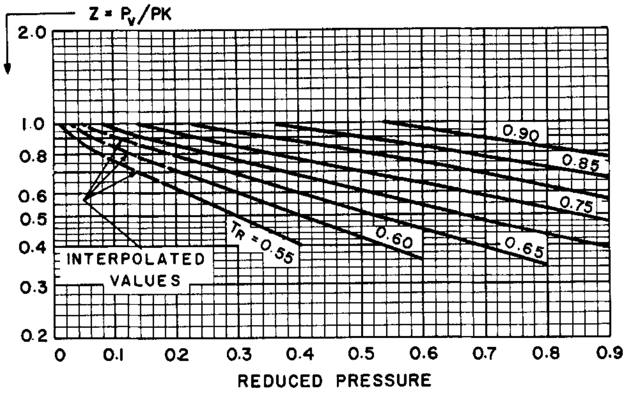

Fig. 13. Generalized correlation of Z values.

tween the two sets of values becomes greater at higher pressures.

Prausnitz (*14*) extrapolated pressure-volume isotherms into the hypothetical vapor region and then calculated hypothetical vapor fugacity coefficients from the hypothetical pressure-volume-temperature data. By this method, the extrapolations were limited to pure components, and effects of composition were avoided.

In this work the van Laar equation was used to extend data into the hypothetical region, and composition effects were considered. In the evaluation of the fugacity coefficients for *n*-pentane and benzene, with the isothermal treatment, two independent binary systems were used for each component. In both cases the fugacity coefficients were found to be a function of temperature and pressure only, regardless of the second component of the binary systems. Likewise, in the isobaric treatment two sets of data were used to evaluate the fugacity coefficients for benzene, and the values were found to be a function of temperature and pressure but independent of the second component in the binary mixtures.

The determinations of the vapor fugacity coefficients for propane from methane-propane data, for *n*-pentane from propane-*n*-pentane data, for benzene from ethane-benzene data, and for benzene from propane-benzene data were done through both an isothermal and isobaric treatment of the data. Both approaches gave very nearly the same values, and the deviations are shown in Table 1. This offers considerable evidence that for the binary systems studied here the changes in volume upon mixing of vapors and the corresponding heats of mixing are small enough to make the correction terms in Equations (11*a*) and (11*b*) practically negligible.

The conclusions are:

1. Vapor phase activity coefficients do actually exist, and they may be represented by an integrated form of the Gibbs-Duhem equation.
2. Standard state hypothetical vapor fugacities for propane, *n*-pentane, and benzene have been evaluated, and they have been correlated as fugacity coefficients by means of a generalized plot in terms of reduced temperature and reduced pressure.
3. Through use of vapor phase activity coefficients and standard state hypothetical vapor fugacities, vapor liquid equilibrium data for multicomponent systems may be predicted.
4. The method disclosed here may be used to evaluate the standard state hypothetical vapor fugacities for additional substances.

## ACKNOWLEDGMENT

The authors thank Mr. Chester James who performed the calculations for the isobaric treatment of the vapor liquid equilibrium as a part of his Master's degree thesis work at the University of Idaho.

## NOTATION

$a^2$ = coefficient in van Laar equation
$f$ = fugacity of a component in a mixture
$f^\circ$ = fugacity of a pure component at a given temperature and pressure (standard state fugacity)
$K$ = vaporization equilibrium constant, defined by Equation (11)
$K'$ = $y/x$, vaporization equilibrium ratio
$P$ = total pressure of a system
$P_v$ = vapor pressure
$R$ = gas law constant
$T$ = absolute temperature
$x$ = mole fraction in liquid phase
$y$ = mole fraction in vapor phase
$Z$ = vaporization equilibrium constant coefficient, defined by Equations (4) and (14)

### Greek Letters

$\Delta H$ = enthalpy change upon mixing pure components
$\Delta v$ = volume change upon mixing pure components
$\gamma^L$ = liquid phase activity coefficient
$\gamma^v$ = vapor phase activity coefficient
$\nu^\circ$ = fugacity coefficient for a pure component, defined by Equation (5)
$\sigma$ = composition parameter dependent upon temperature and pressure
$\sigma^\circ$ = attraction coefficient for a pure substance dependent upon temperature and pressure
$\tau^+$ = composition parameter dependent upon temperature

### Subscripts

$i, j$ = components in a mixture
$L$ = liquid phase
$r$ = reduced property
$v$ = vapor phase
1, 2 = specific components in a mixture

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