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VAPOR-LIQUID EQUILIBRIUM STUDIES FOR HYDROGEN-CONTAINING MIXTURES

BY

CHENG-TZE FU

A thesis submitted in partial fulfillment

of the requirement for the degree of

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ABSTRACT

Vapor-liquid equilibria (VLE) behavior of hydrogen-containing mixtures which also contain nitrogen, carbon monoxide, and methane have been investigated. The research work also encompassed the design, construction and testing of a new apparatus to improve the measurement of VLE properties. In addition, the VLE values from literature were analyzed and correlated to establish the temperature dependence of binary interaction parameters which are useful for predicting multicomponent VLE values.

In Part A of the presentation, the data on phase behavior and critical loci of binary, ternary, and quaternary hydrogen-containing systems are presented in the pressure-composition and/or temperature-composition diagrams. Mixtures of these compounds are of special importance for designing and evaluating the performance of coke oven gas, and petroleum cracking and gas reforming plants.

The design, construction and operation of the new laboratory equipment for measuring vapor-liquid equilibrium under high pressure and low temperature are described in Part B of the presentation. In this design, upper pressure limit was increased to 340 atm (34473.65 KPa) and dead space was reduced to the volume of the needle valves of the sampling lines.

The newly designed apparatus was used to measure the VLE properties for binary system nitrogen-carbon dioxide at 273.15 K. The experimental results were found in good agreement with those obtained from the literature. In addition, the VLE properties of the binary nitrogen-methane system at 169.9 K and the binary system methane-ethylene at 243.15 and 223.15 K were measured. The experimental results of these binary systems were successfully compared with those values calculated from the equation of state of Ishikawa, Chung and Lu (ICL).

The ICL equation was used to correlate and to predict the VLE values of mixtures containing hydrogen, nitrogen, carbon monoxide and methane at high pressures and low temperatures. Binary interaction coefficients describing the behavior of mixture of these constituents, C_{ij}, were established using the ICL Equation from binary experimental data available in the literature. A total of 553 data points taken at 84 isotherms was selected in the determination. The calculated C_{ij} values for each binary system were correlated linearly with temperatures. These coefficients which were obtained from the correlations were then used in the ICL Equation to predict VLE values for three hydrogen-containing ternary systems, hydrogen-nitrogen-carbon monoxide, hydrogen-nitrogen-methane, and hydrogen-carbon monoxide-methane, and for the quaternary system hydrogen-nitrogen-carbon monoxide-methane for a total of 12 isothermal conditions.

The calculated VLE values for the three hydrogen-containing ternary systems were also compared with those obtained from the modified Soave-Redlich-Kwong (SRK) equation. In addition, the calculated VLE compositions for the quaternary system at three isotherms using the Lee-Kesler correlation as applied by Plocker et al. were also compared with the experimental data and with those obtained from the ICL and the modified SRK equations.

It was found that the ICL equation and the modified SRK equation are adequate for predicting phase equilibrium properties as intended. Furthermore, the ICL and the modified SRK equations, and the method of Plocker et al. are all suitable for representing the VLE properties of the quaternary system hydrogen-nitrogen-carbon monoxide-methane at 103.15, 93.15 and 77.35 K. For the sake of simplicity, the ICL and the modified SRK equations are recommended for this quaternary system.

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NOMENCLATURE

a,b	= constants of the van der Waals equation.
a,b	= constants of the Redlich-Kwong equation.
a,b	= constants of the Soave Redlich-Kwong equation.
a,b	= constants of the Peng-Robinson equation.
a,b	= parameters of the Ishikawa-Chung-Lu equation.
$B,C,D,\ldots$	= virial coefficients of the virial equation.
$B,C,D,$ b,c,d,β,γ	= constants in the Lee-Kesler equation.
A_o,B_o,C_o a,b,c,α,γ	= constants in the Benedict-Webb-Rubin equation of state.
C_{ij}	= binary interaction coefficient for components i and j.
f	= fugacity.
G	= molar Gibbs free energy.
K	= equilibrium ratio, y/x.
K	= degree of Kelvin.
m	= slope of equation (III-1).
N	= number of components.
n	= number of moles.
P	= pressure.
R	= gas constant.
T	= absolute temperature.
V	= volume.
v	= molar volume.

x = mole fraction in the liquid phase.

y = mole fraction in the vapor phase.

Z = compressibility factor.

z = mole fraction of component either in the liquid phase or in the vapor phase.

GREEK LETTERS

Ω_a, Ω_b = parameters of the Ishikawa-Chung-Lu equation.

ϕ = fugacity coefficient.

Δ = difference.

∂ = partial differential.

$\sum$ = summation.

μ = chemical potential.

ω = acentric factor.

SUPERSCRIPTS

L = liquid phase.

V = vapor phase.

o = standard state.

r = reference value.

$\hat{}$ = property in mixture.

$-$ = partial molar quantity.

SUBSCRIPTS

AV	= average value.
c	= critical property.
calc	= calculated.
expl	= experimental.
i,j,k	= component identification.
M	= mixture.
r	= reduced property.
T	= total
1,2,3,4,11,22,33,44	= component identification.
12,23,31,14,24,34 123,124,134,234	= mixture identification.

PART A

VAPOR LIQUID EQUILIBRIA FOR HYDROGEN-CONTAINING MIXTURES

CHAPTER I

INTRODUCTION

Vapor-liquid equilibrium data for mixtures containing hydrogen, nitrogen, carbon monoxide, methane are of particular interest to the purification of coke-oven gas (COG) plants.

The phase behavior and probable critical loci of binary, ternary and quaternary mixtures of these components have not been thoroughly studied. Some of the critical loci for hydrogen containing binary and ternary system are neither agreeable nor complete in current literature (1,6,71,72,82,101-105). For example, information pertinent to critical points for binary systems hydrogen-nitrogen and hydrogen-carbon monoxide in the temperature range from 77.35 to 103.15 K are limited and some of these are not evidently available in public domain. Thus, a further investigation of PVT relations including determination of probable critical loci, and understanding of phase behavior for these major components of COG mixtures are needed.

VLE data available in the literature for mixtures containing hydrogen, nitrogen, carbon monoxide and methane have been studied and reported in Table I-1. Information required for the design of COG plants are not completely available in the literature. For example, experimental VLE values of the ternary system hydrogen-carbon monoxide-methane cannot be found in the literature at the region or vicinity of the critical point.

Table I-1 VLE Data Available in the Literature for Mixtures Containing Hydrogen, Nitrogen, Carbon Monoxide and Methane

	System	Reference
Binary	Hydrogen-Nitrogen	1,50,82,102
	Hydrogen-Carbon Monoxide	1,104
	Hydrogen-Methane	6,33,43,72,100
	Nitrogen-Carbon Monoxide	78,106
	Nitrogen-Methane	12,78,88
	Carbon Monoxide-Methane	12,78,88
Ternary	Hydrogen-Nitrogen-Carbon Monoxide	1,20,22,23,50,70,71,91
	Hydrogen-Nitrogen-Methane	20,52,81,100
	Hydrogen-Carbon Monoxide-Methane	17,31,100

It is interesting to note that the only available information for the quaternary system hydrogen-nitrogen-carbon monoxide-methane in the literature is an abstract translated from Russian which appeared in 1954 (87). Therefore, characterization of the quaternary system is of great interest, and is highly valuable for the design of COG plants.

There are numerous equations of state (3, 5, 9, 10, 14, 19, 21, 24-26, 29, 30, 37, 38, 45, 47-49, 54, 56, 59-61, 65, 68, 69, 74-76, 79, 80, 84-86, 90, 95, 96, 99, 108) available in the literature for calculating VLE values at high pressures and low temperatures. It is of importance to select the most suitable equation for representing VLE behavoir of mixtures because equations of state provide the only way to calculate the properties of compounds that are supercritical, subcooled, at conditions of interest. Excellent results can be obtained if the required binary interaction coefficients for the selected equation are available. In this study, the recently proposed Ishikawa-Chung-Lu (ICL) equation (37, 38, 49) was chosen for predicting VLE values at cryogenic conditions, because this equation had been found to be most suitable for representing the vapor-liquid equilibria of the hydrogen-containing mixtures. For the purpose of comparison, two recently revised equations of state, the modified Soave-Redlich-Kwong (SRK) equation (24) and the modified Benedict-Webb-Rubin (BWR) equation (61), were also used for VLE calculations in this study.

Although the two parameters of the ICL equation for pure component have been established and generalized for a number of nonpolar compounds (36-38), the nature of binary interaction coefficients of the

correlated with temperature. The temperature dependence of these coefficients has not been well established in the literature. For example, Graboski et al. (24) and Plocker et al. (61) considered these coefficients insensitive to the temperature change. On the other hand, Kato et al. (41) considered these coefficients temperature dependent. For this reason, the binary interaction coefficients of the ICL equation and the temperature dependence of these coefficients were investigated in this study.

CHAPTER II

THEORETICAL CONSIDERATIONS

The thermodynamic relationships required in the development of formulas for predicting VLE properties are discussed. Equations of state which are used in this investigation and the history of their development are briefly presented in this chapter.

II-A. THERMODYNAMIC RELATIONSHIPS USED IN PREDICTING VLE PROPERTIES BY MEANS OF AN EQUATION OF STATE

When a liquid mixture is in equilibrium with a vapor, the parameters of interest are the pressure, temperature, and the equilibrium compositions of both phases. Neglecting the surface, gravitational, magnetic, and other field effects, conditions under which thermodynamic equilibrium for each component i in the mixture can be achieved are:

$$T^L = T^V \qquad \text{(II-1)}$$

$$p^L = p^V \qquad \text{(II-2)}$$

$$\mu_i^L = \mu_i^V \qquad i = 1,2,\ldots\ldots N \qquad \text{(II-3)}$$

or

$$\hat{f}_i^L = \hat{f}_i^V \qquad \text{(II-4)}$$

where L, V, T, P, μ_i, $\hat{f}_i$, and N are the liquid phase, vapor phase, system temperature; total pressure, chemical potential of component i, fugacity of component i in the solution, and total number of components in the mixture, respectively.

In a homogeneous phase, the chemical potential of component i is defined as:

$$\mu_i = \left(\frac{\partial G}{\partial n_i}\right)_{T,P,n_j} \qquad \text{(II-5)}$$

where the G is the total Gibbs free energy of the mixture. The quantities n_i and n_j $(i \neq j)$ are the numbers of moles of components i and j in the mixture, respectively.

Let μ_i be the chemical potential of the component i in a gas mixture at temperature T and pressure P. For the component i of the gas mixture, the fugacity $\hat{f}_i$ is defined by the following relations in which μ_i° is a function of temperature only:

$$\mu_i = \mu_i^\circ + RT \ln \hat{f}_i \qquad \text{(II-6)}$$

$$\hat{f}_i / y_i P \to 1 \quad \text{as} \quad P \to 0 \qquad \text{(II-7)}$$

The fugacity of component i in the gas mixture depends on temperature, pressure, and composition of that mixture.

The fugacity coefficient of component i in an imperfect gas mixture is defined by:

$$\hat{\phi}_i = \frac{\hat{f}_i}{y_i P} \qquad \text{(II-8)}$$

The ratio $\hat{f}_i / y_i P$ is a dimensionless group. For a imperfect pure gas, Equation (II-8) becomes $\phi = f/P$.

The equation used for determining $\hat{\phi}_i$ (62) is given by:

$$\ln \hat{\phi}_i = \frac{1}{RT} \int_0^P \left[\left(\frac{\partial V}{\partial n_i} \right)_{T,P,n_j} - \frac{RT}{P} \right] dP \qquad \text{(II-9)}$$

or

$$\ln \hat{\phi}_i = \frac{1}{RT} \int_V^\infty \left(\left(\frac{\partial P}{\partial n_i} \right)_{T,V,n_j} - \frac{RT}{V} \right) dV - \ln Z \qquad \text{(II-10)}$$

where V is the total volume of the gas mixture, and Z is the compressibility factor of the gas mixture at T and P. Since most equations of state are explicit in pressure, Equation (II-10) is more convenient to use.

The fugacity coefficient of component i in a mixture can be calculated from Equation (II-9) or (II-10) if sufficient volumetric data are available. Since such data are generally not available, especially for multi-component systems, calculations of fugacity coefficients are most often performed by an equation of state.

For data reduction, it is also desirable to represent the experimentally determined VLE values by a suitable equation of state. All the equations mentioned in the next section have been widely used for such calculations.

II-B. EQUATIONS OF STATE

Numerous equations of state have been proposed in the literature. In this section, an attempt is made to describe the equations which are most suitable for representing the results of this investigation. Historical development of some important equations of state are also briefly presented.

II-B1. Van der Waals Equation of State

In 1873, Van der Waals (90) proposed the following equation of state:

$$P = \frac{RT}{v-b} - \frac{a}{v^2} \qquad \text{(II-11)}$$

Using the relationship $\left(\frac{\partial p}{\partial v}\right)_{T_c} = 0$ and $\left(\frac{\partial^2 p}{\partial v^2}\right)_{T_c} = 0$ at the critical point, one can express the quantities a and b in terms of P_c and T_c as follows:

$$a = \frac{27R^2T_c^2}{64P_c} \qquad \text{(II-12)}$$

$$b = \frac{RT_c}{8P_c} \qquad \text{(II-13)}$$

The equation can be reduced to the ideal gas expression if both a and b are set equal to zero. The first term of the right-hand side of Equation (II-11) accounts for the repulsive forces among molecules. The term a/v^2 represents the attractive forces. This volume-cubic

equation is capable of representing both vapor and liquid behavior though it is inaccurate over extended temperature ranges.

Equation (II-11) can be rewritten as:

$$P = \frac{n_T RT}{V-n_T b} - \frac{n_T^2 a}{V^2} \qquad \text{(II-14)}$$

where $v = V/n_T$ and n_T is the total number of moles.

To calculate the fugacity of component i in the mixture at given temperature, pressure, and composition, Equation (II-14) is differentiated with respect to n_i.

$$\left(\frac{\partial P}{\partial n_i}\right)_{T,V,n_j} = \frac{RT}{V-n_T b} + \frac{n_T RT\left(\frac{\partial (n_T b)}{\partial n_i}\right)}{(V-n_T b)^2} - \frac{1}{V^2}\frac{\partial (n_T^2 a)}{\partial n_i} \qquad \text{(II-15)}$$

Substituting Equation (II-15) into Equation (II-10) gives:

$$RT \ln\left(\frac{\hat{f}_i}{y_i P}\right) = RT \ln \frac{V-n_T b}{V}\Bigg]_V^\infty - n_T RT \left(\frac{\partial (n_T b)}{\partial n_i}\right)\frac{1}{(V-n_T b)}\Bigg]_V^\infty + \left(\frac{\partial (n_T^2 a)}{\partial n_i}\right)\frac{1}{V}\Bigg]_V^\infty - RT \ln Z. \qquad \text{(II-16)}$$

At the upper limit of integration, where $V \to \infty$,

$$\ln \frac{V-n_T b}{V} \to 0, \quad \frac{1}{V-n_T b} \to 0, \quad \frac{1}{V} \to 0, \qquad \text{(II-17)}$$

Equation (II-16) becomes

$$RT \ln \frac{\hat{f}_i}{y_i P} = RT \ln \frac{V}{V-n_T b} + n_T RT \left(\frac{\partial (n_T b)}{\partial n_i}\right)\left(\frac{1}{V-n_T b}\right) - \left(\frac{\partial (n_T^2 a)}{\partial n_i}\right)\frac{1}{V} - RT \ln Z. \qquad \text{(II-18)}$$

or

$$\ln \hat{\phi}_i = \ln \frac{\hat{f}_i}{y_i P} = \ln \frac{v}{v-b} - \frac{b_i}{v-b} - \frac{2\sqrt{a_i} \sum_i y_i \sqrt{a_j}}{vRT} - \ln Z \quad \text{(II-19)}$$

Equation (II-19) which was derived from rigorous thermodynamic relations gives the desired result. For a mixture, the quantities a and b are expressed as:

$$a = \sum_{i=1} \sum_{j=1} y_i y_j a_{ij} \quad \text{(II-20)}$$

$$b = \sum_{i=1} y_i b_i \quad \text{(II-21)}$$

where a is the average strength of attraction between all molecular pairs and b is the average volume of all molecules in the mixture. a_{ij} is a measure of the strength of attraction between a molecule i and a molecule j. Unfortunately, current molecular theory is insufficient to predict the properties of mixtures. For $i \neq j$, Berthelot (63) suggested, on strictly empirical grounds that a_{ij} could be obtained from the geometric mean of a_i and a_j,

$$a_{ij} = \sqrt{a_i a_j} \quad \text{(II-22)}$$

Equation (II-19) indicates that the fugacity of a component in the mixture at a given temperature, pressure, and composition, the constants a and b could be calculated using the mixing rules given by Equations (II-20), (II-21), and (II-22). In practice, the fugacities of the mixture are calculated by first obtaining the fugacity coefficients. For instance, $\hat{f}_i^L$ is a function of x_i, P, and T, and $\hat{f}_i^V$ is a function of y_i, P, and T. From Equation (II-14), the molar volume v

of the mixture can be obtained by a trial-and-error method. Once the molar volume is known, the compressibility factor Z is easily calculated and the fugacity is readily found from Equation (II-19).

II-B2. Virial Equation of State

The virial equation of state has been proposed for gases and has been explained with the help of the molecular theory. The compressibility factor, Z, of a gas can be expressed by an infinite series using either reciprocal molar volume, 1/v, or pressure, P, as the independent variable. The Leiden form gives:

$$Z = \frac{Pv}{RT} = 1 + \frac{B}{v} + \frac{C}{v^2} + \ldots. \qquad \text{(II-23)}$$

The term B/v accounts interactions between pairs of molecules; the C/v^2 term accounts three-body interactions; etc. Since two-body interactions are many times more common than three-body interactions, and three-body interactions are many times more common than four-body interactions, etc., the contributions to Z or successively higher-ordered terms fall off rapidly. In general practice, only the first three terms of the Equation (II-23) is used and the remaining terms are truncated at low and moderate pressures.

For an imperfect gas mixture of N components, virial coefficients of the mixture are defined as:

$$B_M = \sum_i \sum_j y_i y_j B_{ij} \qquad \text{(II-24)}$$

$$C_M = \sum_i \sum_j \sum_k y_i y_j y_k C_{ijk} \qquad \text{(II-25)}$$

where B_M and C_M are the second and third Virial coefficients of the mixture, respectively. B_{ij} and C_{ijk} are the cross Virial coefficients.

The fugacity coefficient for a component i in a mixture can be obtained for using a similar approach as described for the van der Waals equation:

$$\ln \hat{\phi}_i = \frac{2}{v} \sum_{j=i} y_i B_{ij} + \frac{3}{2} \frac{1}{v^2} \sum_{j=1} \sum_{k=1} y_j y_k C_{ijk} - \ln Z_M \qquad \text{(II-26)}$$

In equation (II-26), the summations are over all components in the mixture.

II-B3. Benedict-Webb-Rubin Equation of State and Its Modifications

In 1928, Beattie and Bridgeman (3) proposed a complicated equation of state having five adjustable parameters. The Beattie and Bridgeman equation fits experimental data for most gases above the critical temperature but becomes unreliable in the vicinity of the critical point or in the co-existent region. Benedict et al.(5) thus developed a modification to this equation. The new equation known as the Benedict-Webb-Rubin (BWR) equation of state has eight constants and is expressed as Equation (II-27).

$$P = \frac{RT}{v} + \frac{B_oRT - A_o - C_o/T^2}{v^2} + \frac{bRT - a}{v^3} + \frac{a\alpha}{v^6} + \frac{c}{v^3T^2}\left(1 + \frac{\gamma}{v^2}\right)\exp\frac{-\gamma}{v^2} \qquad \text{(II-27)}$$

where A_o, B_o, C_o, a,b,c,α, and γ are constants for a given fluid. In spite of its complexity, this equation has a number of applications in industry because of its greater overall accuracy. It is also the first expression which successfully predicts the vapor pressure and thermodynamic properties of the coexisting phases in pure fluids.

A number of modifications (21, 56, 79, 80, 84) had been proposed following the success of the original BWR equation. One was developed by Lee and Kesler (45) as shown below:

$$Z = \frac{P_rv_r}{RT_r} = 1 + \frac{B}{v_r} + \frac{C}{v_r^2} + \frac{D}{v_r^5} + \frac{c_4}{T_r^3v_r^2}\left(\beta + \frac{\gamma}{v_r^2}\right)\exp\left(-\frac{\gamma}{v_r^2}\right) \qquad \text{(II-28)}$$

with $B = b_1 - b_2/T_r - b_3/T_r^2 - b_4/T_r^3$

$C = c_1 - c_2/T_r + c_3/T_r^3$

$D = d_1 + d_2/T_r$

where b_1, b_2, b_3, b_4, c_1, c_2, c_3, c_4, d_1, d_2, β, and γ are constants.

While tested primarily on hydrocarbon components in the reduced temperature ranges of 0.3 to 4 and in the reduced pressure ranges of 0 to 10, average errors were reported to be less than two percent for both vapor and liquid phases.

In 1978, Plocker et al. (61) combined into the Lee-Kesler (45) form of the BWR equation (5) some empirical formulas of Prausnitz and co-workers (61,63). The new proposed mixing rules were given by:

$$T_{cM} = \frac{1}{v_{cM}^{\eta}} \sum_j \sum_k y_j y_k v_{cjk}^{\eta} T_{cjk} \qquad \text{(II-29)}$$

$$v_{cM} = \sum_j \sum_k y_j y_k v_{cjk} \qquad \text{(II-30)}$$

$$\omega_M = \sum_j y_j \omega_j \qquad \text{(II-31)}$$

$$T_{cjk} = (T_{cj} T_{ck})^{0.5} k_{jk} \qquad \text{(II-32)}$$

$$v_{cjk} = \frac{1}{8} (v_{cj}^{1/3} + v_{ck}^{1/3})^3 \qquad \text{(II-33)}$$

where v_c is the molar critical volume, k_{jk} is an adjustable binary parameter, characteristic of the binary mixture. The pseudo-critical pressure was reported as:

$$P_{cM} = (0.2905 - 0.085\omega_M) RT_{cM}/v_{cM} \qquad \text{(II-34)}$$

The binary interaction coefficients, k_{jk}, depends on binary system only.

For phase equilibrium calculations, the fugacity coefficient of the ith component in a mixture, liquid or vapor, can be calculated from:

$$\ln \hat{\phi}_i = \ln \phi_M - \frac{1}{T} \frac{\Delta h_M}{R \cdot T_{cM}} \sum_{j \neq i} z_j \left(\frac{dT_{cM}}{dz_j}\right)_{z_k} + \frac{Z_M - 1}{P_{cM}} \sum_{j \neq i} z_j \left(\frac{dP_{cM}}{dz_j}\right)_{z_k} - \left(\frac{\partial \ln \phi_M}{\partial \omega_M}\right)_{T_r P_r} \sum_{j \neq i} z_j \left(\frac{d\omega_M}{dz_j}\right)_{z_k} \quad (k \neq i, j) \qquad \text{(II-35)}$$

with

$$\left(\frac{\partial \ln\phi_M}{\partial \omega_M}\right)_{T_r P_r} = \frac{1}{\omega(r)} \cdot [(\ln\phi_M)(r) - (\ln\phi_M)(0) \qquad \text{(II-36)}$$

$$\left(\frac{dT_{cM}}{dz_j}\right)_{z_k} = [2 \sum_{1} z_l \, (v_{clj}{}^{n} \, T_{clj} - v_{cli}{}^{n} \, T_{cli})$$

$$- n v_{cM}{}^{n-1} \left(\frac{dv_{cM}}{dz_j}\right)_{z_k} T_{cM}]/v_{cM}{}^{n} \qquad \text{(II-37)}$$

$$\left(\frac{dv_{cM}}{dz_j}\right)_{z_k} = 2 \sum_{1} z_l \, (v_{clj} - v_{cli}) \qquad \text{(II-38)}$$

$$\left(\frac{dP_{cM}}{dz_J}\right)_{z_k} = P_{cM}\left[\left(\frac{dZ_{cM}}{dz_j}\right)_{z_k}/Z_{cM}\right.$$

$$\left. + \left(\frac{dT_{cM}}{dz_J}\right)_{z_k}/T_{cM} - \left(\frac{dv_{cM}}{dz_J}\right)_{z_k}/v_{cM}\right] \qquad \text{(II-39)}$$

$$\left(\frac{dZ_{cM}}{dz_j}\right)_{z_k} = -0.085 \left(\frac{d\omega_M}{dz_j}\right)_{z_k} \qquad \text{(II-40)}$$

$$\left(\frac{d\omega_M}{dz_j}\right)_{z_k} = \omega_j - \omega_i \qquad \text{(II-41)}$$

In Equation (II-35) to (II-41), subscript k is not equal to i or j.

A special procedure is required for calculating $\Delta h_M/RT_{cM}$ and $\ln\phi_M$. The quantity $\ln\phi_M$ is a function of T_r, P_r, and ω_M, and Δh_M is the residual enthalpy of the mixture. The quantity z_i refers to the mole fraction of component i either in the liquid phase or in the vapor phase. The calculation procedure for the liquid phase is identical to that of the vapor phase.

II-B4. Redlich-Kwong Equation of State and Its Modifications

A two-constant equation of state, which effectively modified the van der Waals equation and widely used for engineering calculations, was proposed by Redlich and Kwong in 1949 (65) as:

$$P = \frac{RT}{v-b} - \frac{a}{T^{1/2}v(v+b)} \qquad \text{(II-42)}$$

with

$$a = \frac{\Omega_a R^2 T_c^{2.5}}{P_c} \qquad \text{(II-43)}$$

$$b = \frac{\Omega_b R T_c}{P_c} \qquad \text{(II-44)}$$

Redlich and Kwong proposed the universal constants 0.42748 and 0.08664 for Ω_a and Ω_b, respectively. Chueh and Prausnitz (14) showed that the Redlich-Kwong equation predicts more acceptable values of density when the values of Ω_a and Ω_b are established from data. Later, Zudkevitch and Joffe (108), Chang and Lu (11) and Hamam and Lu (29) showed that Ω_a and Ω_b vary with the temperature.

To apply Equation (II-42) to mixtures, Redlich and Kwong used the same mixing rules as those of van der Waals.

The fugacity coefficient of component i in each phase of the mixture (62) is:

$$\ln \hat{\phi}_i = \ln \frac{v}{v-b} + \frac{b_i}{v-b} - \frac{2\sum_{j=1} y_i a_{ij}}{RT^{3/2}b} \ln \frac{v+b}{v} + \frac{ab_i}{RT^{3/2}\,b^2}\left(\ln \frac{v+b}{v} - \frac{b}{v+b}\right) - \ln Z \qquad \text{(II-45)}$$

II-B4a. Soave Redlich-Kwong Equation of State

Modifications to the Redlich-Kwong equation were made and appeared in the literature for many years. Soave (76) proposed a simplification to the rigorous procedures of Zudkevitch and Joffe (108), and Chang and Lu (11). The equation becomes:

$$P = \frac{RT}{v-b} - \frac{a(T)}{v(v+b)} \qquad \text{(II-46)}$$

with

$$a(T) = a\alpha \qquad \text{(II-47)}$$

$$a = \Omega_a R^2 T_c^{\,2}/P_c \qquad \text{(II-48)}$$

$$b = \Omega_b RT_c/P_c \qquad \text{(II-49)}$$

For hydrocarbons

$$\alpha = (1+m\ (1-T_r^{\,0.5}))^2 \qquad \text{(II-50)}$$

$$m = 0.480 + 1.57\omega - 0.176\omega^2 \qquad \text{(II-51)}$$

The Redlich-Kwong equation was modified by replacing the expression $a/T^{0.5}$ with a more general temperature-dependent term $a(T)$. In equation (II-47), a is an energy constant. The quantity α depends on temperature and varies with substances as shown in equation (II-50) and equation (II-51). Constants Ω_a and Ω_b have the same values as those of the original RK equation. ω is the acentric factor.

For mixtures of nonpolar fluids, such as hydrocarbons, nitrogen and carbon monoxide, a generalized mixing rule was adopted:

$$\alpha a = \sum_{i=1}^{n} \sum_{j=1}^{n} y_i y_j \alpha_{ij} a_{ij} \qquad \text{(II-52)}$$

$$b = \sum_{j=1}^{n} y_i b_i \qquad \text{(II-53)}$$

Hence

$$\alpha_{ij}a_{ij} = (\alpha_i a_i \alpha_j a_j)^{1/2} (1-C_{ij}) \qquad \text{(II-54)}$$

The quantity C_{ij} is the interaction coefficient as described by Chueh and Prausnitz (14). It corrects the deviation of a_{ij} from the geometric mean. Values of C_{ij} are determined from experimentally measured data for binary mixtures. For hydrocarbons, Soave suggested that the value of C_{ij} should be set equal to zero.

Based on the Soave-Redlich-Kwong equation of state, the fugacity coefficient of component i in a mixture, $\hat{\phi}_i$, is given by:

$$\ln \hat{\phi}_i = \frac{b_i}{b}(Z-1) - \ln(Z-B) - \frac{A}{B}\left(\frac{2\sum_{i=1}^{n} y_j \alpha_{ij} a_{ij}}{a}\right) - \frac{b_i}{b} \ln\left(1 + \frac{B}{Z}\right) \qquad \text{(II-55)}$$

where b_i is the volume constant of component i, $\alpha_{ij}a_{ij}$ is given by Equation (II-54). Furthermore,

$$A = \frac{\alpha a P}{R^2 T^2} \qquad \text{(II-56)}$$

$$B = \frac{bP}{RT} \qquad \text{(II-57)}$$

and Z is the compressibility factor.

Graboski and Daubert (24) modified the Soave Redlich-Kwong equation in 1978. For all fluids except hydrogen, the Equation (II-51) was

revised as:

$$m = 0.48508 + 1.55171\omega - 0.15613\omega^2 \qquad \text{(II-58)}$$

For hydrogen component, a new Equation (II-59) was derived:

$$\alpha_{H_2} = 1.202 \exp(-0.30228\ T_{r,H_2}) \qquad \text{(II-59)}$$

where

$$T_{r,H_2} = T/T_{c,H_2} = \text{reduced temperature of hydrogen} \qquad \text{(II-60)}$$

The binary interaction coefficients were used to improve the phase equilibrium predictions for systems containing H_2S, CO_2, N_2, CO, SO_2 or H_2O. For hydrocarbon mixtures, Soave suggested that C_{ij} values were set equal to zero. Graboski et al. found that this approximation was reasonable and used it in vapor-liquid equilibrium calculations for hydrocarbon mixtures. They also suggested that C_{ij} values for binary mixtures containing hydrogen and other components should be set equal to zero.

II-B4b. Peng-Robinson Equation of State

Another modification of Redlich-Kwong equation was made by Peng and Robinson (60) in 1976:

$$P = \frac{RT}{v-b} - \frac{a(T)}{v(v+b)+b(v-b)} \qquad \text{(II-61)}$$

in which

$$a(T) = a(T_c)\ \alpha(T_r,\omega) \qquad \text{(II-62)}$$

$$a(T_c) = 0.45724\ R^2 T_c^{\,2}/P_c \qquad \text{(II-63)}$$

$$b = b(T_c) = 0.07780\ RT_c/P_c \qquad \text{(II-64)}$$

For all substances,

$$\alpha = (1 + K\ (1-T_r^{\,0.5}))^2 \qquad \text{(II-65)}$$

$$K = 0.37464 + 1.54226\omega - 0.26992\omega^2 \qquad \text{(II-66)}$$

Although Equations (II-65) and (II-66) are similar to Equations (II-50), (II-51) and (II-58), they were derived from different conditions.

For mixtures, the mixing rules are given by

$$a = a(T) = \sum_i \sum_j y_i y_j a_{ij} \qquad \text{(II-67)}$$

$$b = \sum_i y_i b_i \qquad \text{(II-68)}$$

where

$$a_{ij} = (1-C_{ij})\ (a_i a_j)^{1/2} \qquad \text{(II-69)}$$

Equations (II-67) to (II-69) are the same as those used by Soave (1972) for the SRK equation, except the definition of a and a_{ij}.

The fugacity coefficient of component i in the vapor phase of a mixture can be obtained from Equation (II-70).

$$\ln \hat{\phi}_i = \frac{b_i}{b}(Z-1) - \ln(Z-B) - \frac{A}{2\sqrt{2}\,B} \cdot \left(\frac{2\sum_k y_k a_{ki}}{a} - \frac{b_i}{b}\right) \ln\left(\frac{Z+2.414B}{Z-0.414B}\right) \qquad \text{(II-70)}$$

where

$$A = aP/R^2T^2 \qquad \text{(II-71)}$$

$$B = bP/RT \qquad \text{(II-72)}$$

$$Z = Pv/RT \qquad \text{(II-73)}$$

This two-parameter cubic equation can be used to accurately predict the vapor pressure of pure substances and the equilibrium ratio of mixtures for nonpolar fluids.

II-B5. Ishikawa-Chung-Lu Equation of State

The two-parameter cubic equation of state proposed by Ishikawa et al. (36-38) in 1980 could represent properties of vapor-liquid equilibrium for multicomponent system containing hydrogen. It combines the hard-sphere compressibility factor of Scott (74) and the empirical attractive term of the Redlich-Kwong equation of state (65):

$$P = \frac{RT}{v}\frac{2v+b}{2v-b} - \frac{a}{T^{0.5}v(v+b)} \qquad \text{(II-74)}$$

The quantities a and b are related to the critical properties as follows:

$$a = \Omega_a R^2 T_c^{2.5}/P_c \qquad \text{(II-75)}$$

$$b = \Omega_b R T_c/P_c \qquad \text{(II-76)}$$

The pure-component parameters Ω_a and Ω_b are considered temperature dependent as well as substance dependent. The generalized correlations

developed by Ishikawa et al (36,37) were used in this study. For nitrogen, carbon monoxide, and methane, the values of Ω_a were obtained by the following equation:

$$\Omega_{a,i} = \sum_{j=0}^{2} (\alpha_{j,Ar} + \beta_j \omega_i) \; T_{r_i}^{-j} \qquad \text{(II-77)}$$

The following values of $\alpha_{j,Ar}$ and β_j were proposed by Ishikawa et al. (36):

$$\alpha_{0,Ar} = 0.351908$$
$$\alpha_{1,Ar} = 0.183860$$
$$\alpha_{2,Ar} = -0.066362 \qquad \text{(II-78)}$$

$$\beta_0 = -0.851174$$
$$\beta_1 = 1.001033$$
$$\beta_2 = -0.222659 \qquad \text{(II-79)}$$

and the values of Ω_b were calculated by the following equation:

$$\Omega_{b,i} = 0.034 - 0.025\omega_i + 0.1 \; T_{r_i}^{-1} - 0.025 \; T_{r_i}^{-2} \qquad \text{(II-80)}$$

For hydrogen, the values of Ω_a and Ω_b were obtained from Equations (II-81) and (II-82), respectively:

$$\Omega_{a,H_2} = 0.503476 - 0.008760 \; T_r^{-1} - 0.028115 \; T_r^{-2} \qquad \text{(II-81)}$$

$$\Omega_{b,H_2} = 0.100278 - 0.0233989 \; T_r^{-1} + 0.032129 \; T_r^{-2} \qquad \text{(II-82)}$$

The values of the acentric factor, ω, used in the calculation are 0.0000, 0.0400, 0.0490, and 0.0072 for hydrogen, nitrogen, carbon monoxide, and methane, respectively (58,62).

In applying Equation (II-74) to phase equilibrium calculations, mixing rules used for gas mixtures are the same as Equations (II-67) to (II-69).

The fugacity coefficient of a component i in the vapor phase, $\hat{\phi}_i^V$, is given by:

$$\ln \hat{\phi}_i^V = \ln \left(\frac{\hat{f}_i^V}{y_i P}\right) = \frac{2b_i}{2v-b_m} - 2\ln\left(1 - \frac{b_m}{2v}\right) - \frac{2\sum_{k=1}^{m} y_k a_{ki}}{RT^{1.5} b_m} \ln \left(\frac{v+b_m}{v}\right)$$

$$+ \frac{a_m b_i}{RT^{1.5} b_m^2} \left[\ln \left(\frac{v+b_m}{v}\right) - \frac{b_m}{v+b_m}\right] - \ln Z_m \qquad \text{(II-83)}$$

The same equations can be applied to calculate the liquid fugacity coefficient ($\hat{\phi}_i^l = \hat{f}_i^l / x_i P$) by replacing y in Equation (II-83) with x.

II-C. EQUATIONS OF STATE USED IN THIS INVESTIGATION

In this study, the ICL equation was selected because it has the following advantages:

1. It keeps the simplicity of a two-constant cubic equation.
2. It can represent VLE values for multicomponent system containing hydrogen and normal fluid.
3. It can be used to accurately predict vapor pressure of pure substances and equilibrium ratios for components in mixtures at low temperatures and high density regions.
4. It can be easily applied to the calculation and prediction of VLE values because the values of Ω_a and Ω_b are already correlated as a function of temperature and substance.

5. It gives accurate results for hydrogen containing systems because the binary interaction coefficients, C_{ij}, are considered to be temperature dependent.

Both the modified Redlich-Kwong equation (25, 48, 59, 76, 108) and the modified Benedict-Webb-Rubin equation (7, 16, 18, 46) have been widely applied in the calculation of vapor-liquid equilibria at high pressures and low temperatures. The modified BWR equation proposed by Lee and Kesler as applied by Plocker et al. (61) and the modified Soave-RK equation advanced by Graboski and Daubert (24) are selected for the purpose of comparison with the Ishikawa-Chung-Lu equation.

For the quaternary system hydrogen-nitrogen-carbon monoxide-methane, calculated values at the three temperatures are compared against the experimental data. The predicted values by the modified SRK method (24) for the three ternaries of hydrogen, those with nitrogen, carbon monoxide and methane, at three isothermal conditions are compared in Table II-9 with those calculated by the ICL equation.

CHAPTER III

RESULTS AND DISCUSSION

The results of Part A of this study may be summarized and discussed in three parts as follows:

III-A. PHASE BEHAVIOR FOR HYDROGEN-CONTAINING SYSTEMS

Available VLE data for systems containing hydrogen reported in the literature are listed in Table I-1. These data were studied and used to construct nine isothermal and twelve isobaric diagrams for the three hydrogen-containing ternary systems: hydrogen-nitrogen-methane, hydrogen-nitrogen-carbon monoxide and hydrogen-carbon monoxide-methane shown in Figures III-1 and III-2. In addition, the data reported by Akers and Eubanks (1) and Yorizane et al. (104) on the binary system hydrogen-carbon monoxide and the data reported by Streett and Calado (82) and Yorizane et al. (102) on the binary system hydrogen-nitrogen were used in estimating the binary critical points as depicted in Figures III-3 to III-5. These binary critical points are also indicated in Figures III-1 and III-2. The nine isothermal diagrams of Figure III-1 were then used to construct the temperature-composition diagrams for these three ternary systems at 125 atm, with the temperature varying from 77.35 to 103.15 K as shown in Figure III-6. The twelve isobaric diagrams of Figure III-2 were used to construct the pressure-composition diagrams at 93.15 K, with pressure varying from 75 to 150 atm, as shown in Figure III-7.

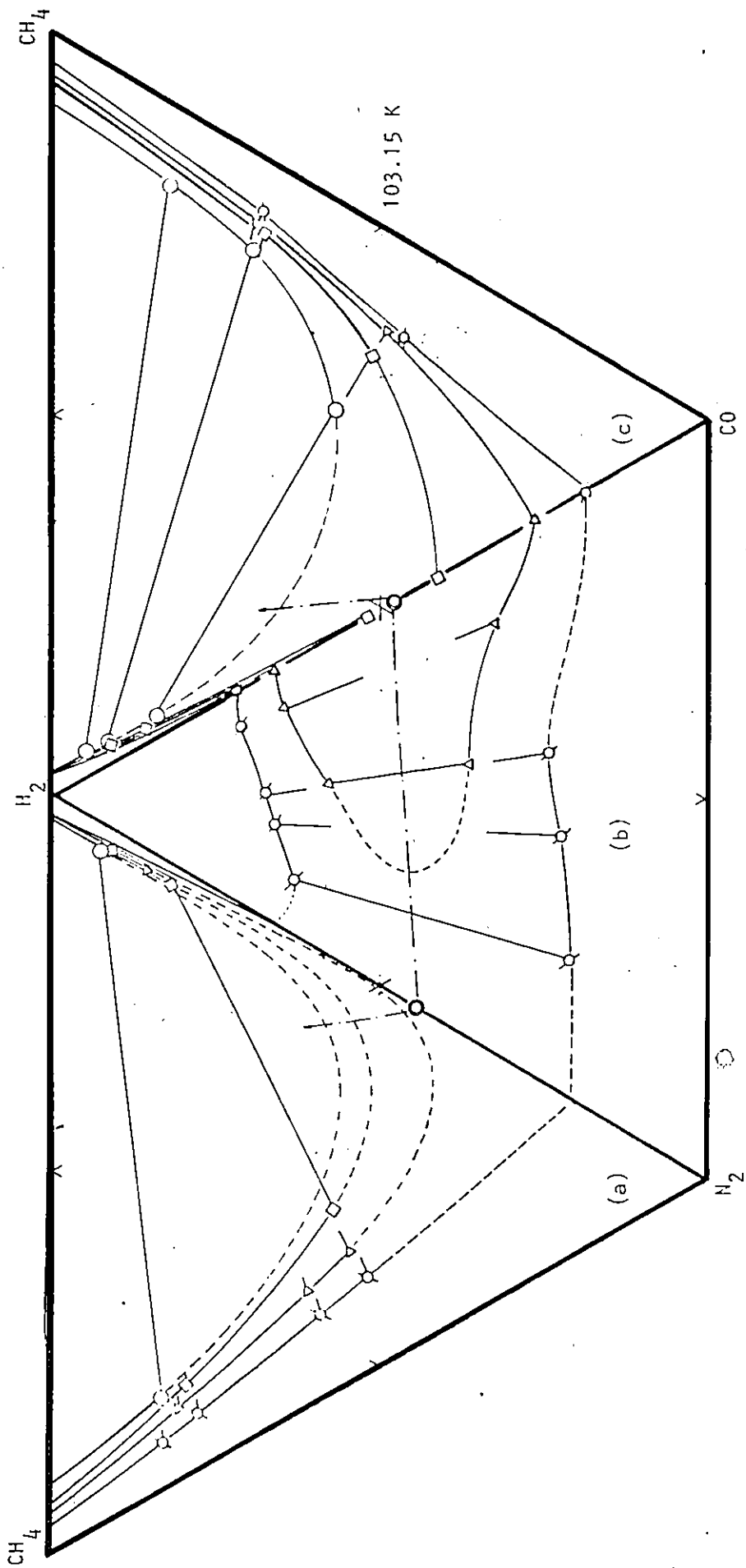


FIGURE III-1: VAPOR-LIQUID EQUILIBRIUM DATA FOR THE TERNARY SYSTEMS H_2-N_2-CH_4, H_2-N_2-CO AND H_2-CO-CH_4 AT THREE ISOTHERMAL CONDITIONS (103.15, 93.15, AND 77.35 K) (▲50 atm^{-1}, ○ 75 atm, △100 atm; □ 125 atm, ● 143 atm, ○ 150 atm, ---- ESTIMATED, -·- ESTIMATED TERNARY CRITICAL LOCUS, ◎ ESTIMATED BINARY CRITICAL POINT)

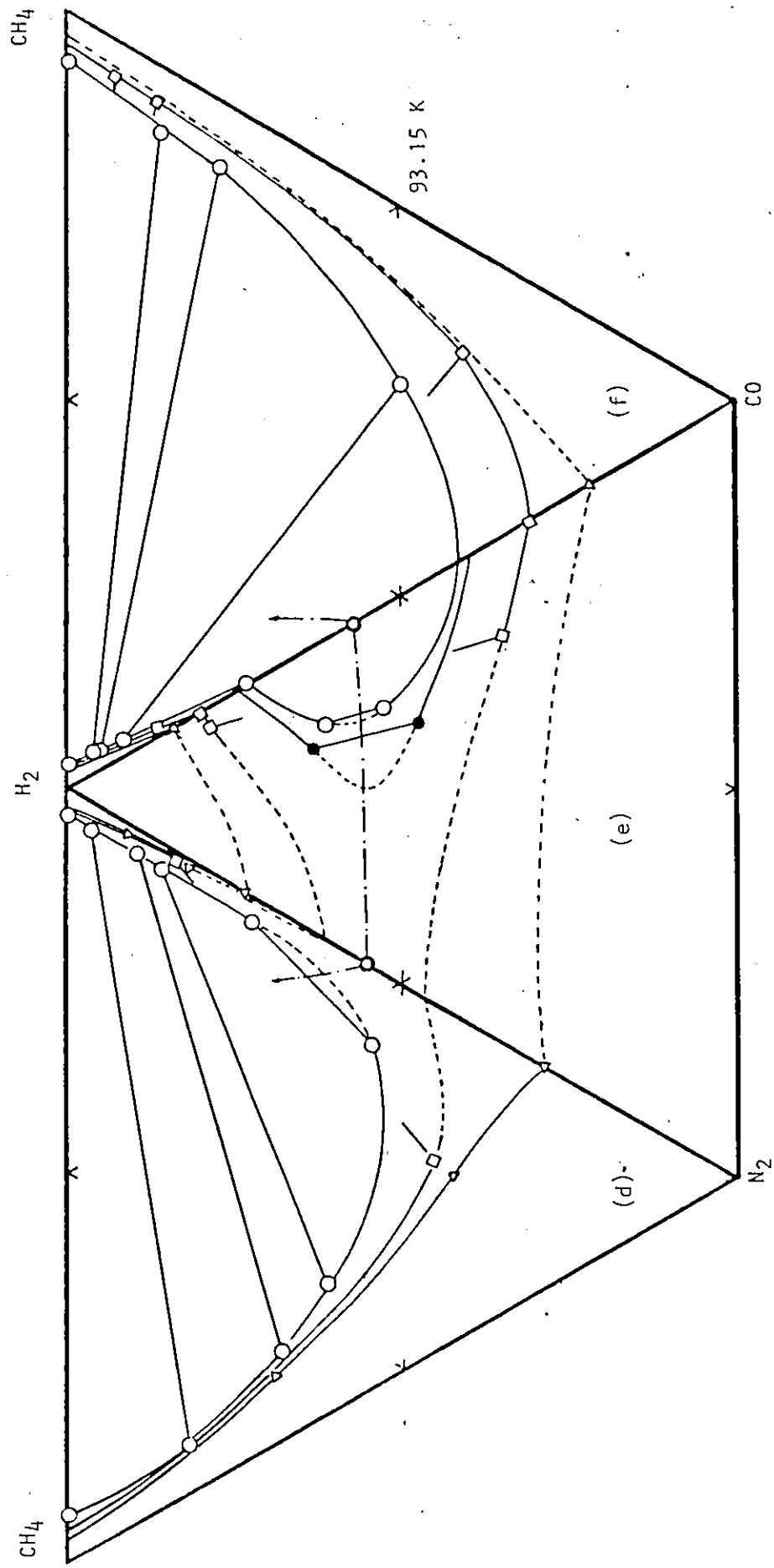


FIGURE III-1: (CONTINUED)

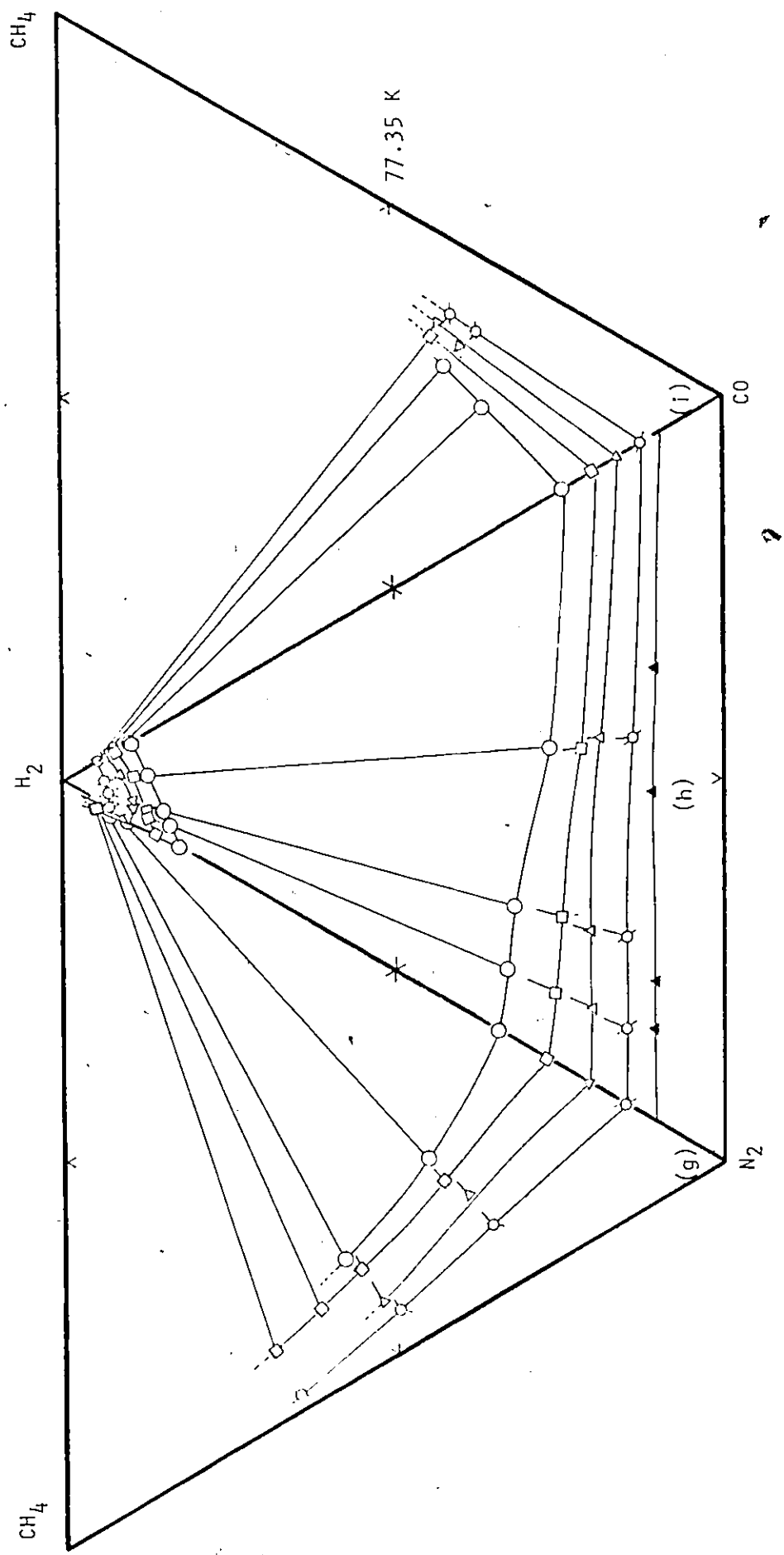


FIGURE III-1: (CONTINUED)

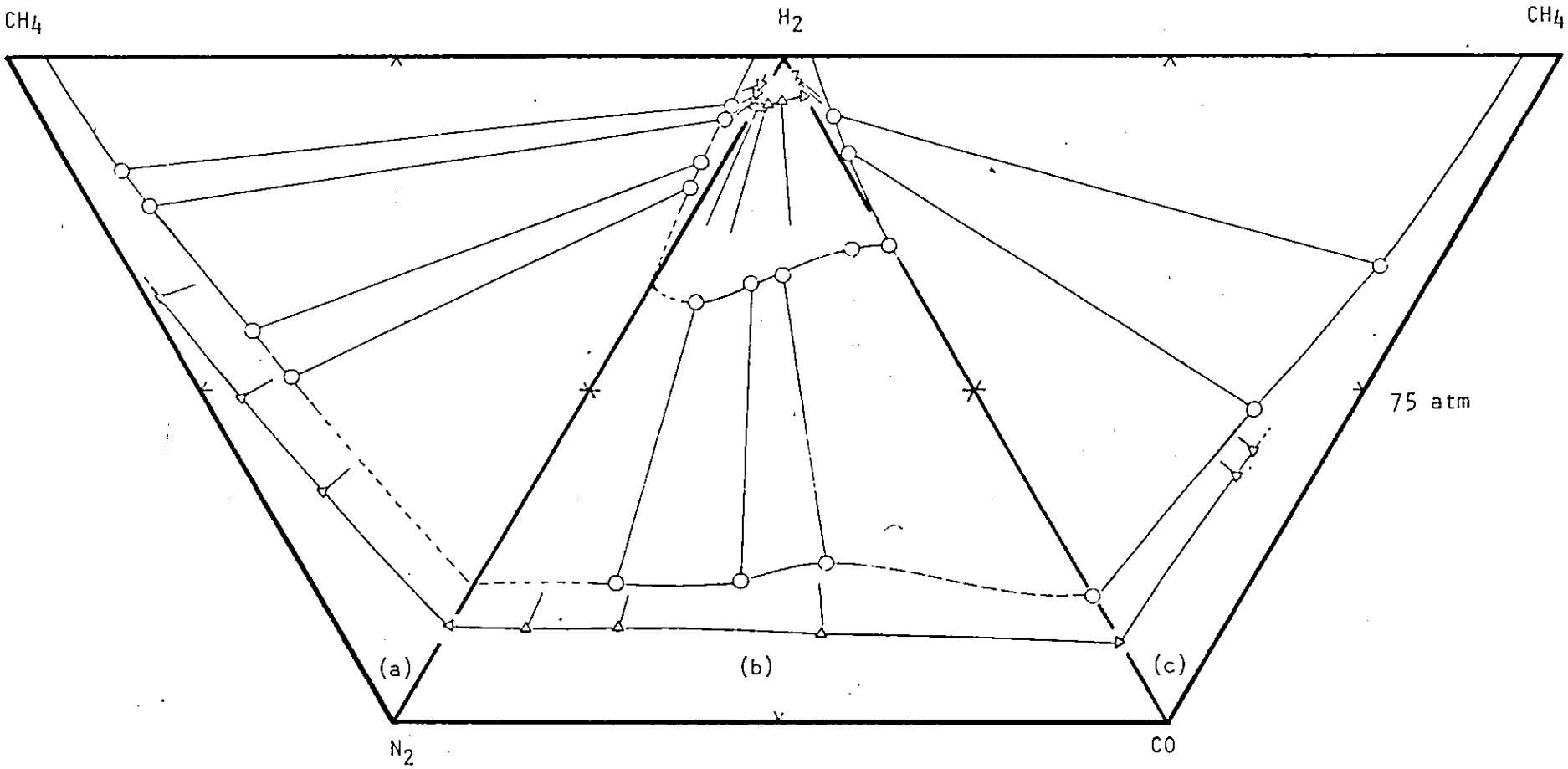


FIGURE III-2: VAPOR-LIQUID EQUILIBRIUM DATA FOR THE TERNARY SYSTEMS H_2-N_2-CH_4, H_2-N_2-CO AND H_2-CO-CH_4 AT FOUR ISOBARIC CONDITIONS (75, 100, 125, AND 150 atm) (O 103.15 K, □ 93.15 K, Δ 77.35 K, ---- ESTIMATED, -•- ESTIMATED TERNARY CRITICAL LOCUS, ◎ ESTIMATED BINARY CRITICAL POINT)

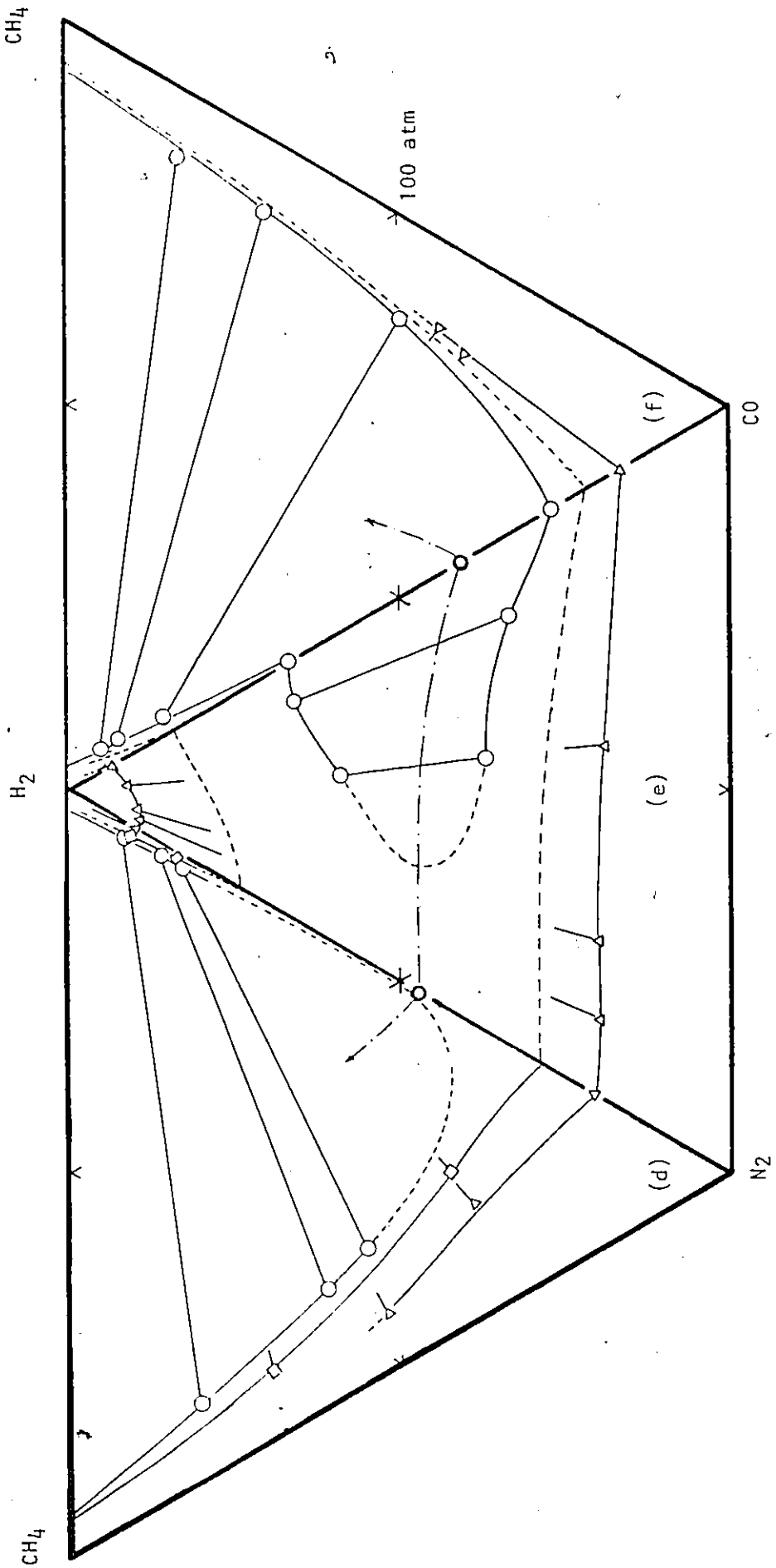


FIGURE III-2: (CONTINUED)

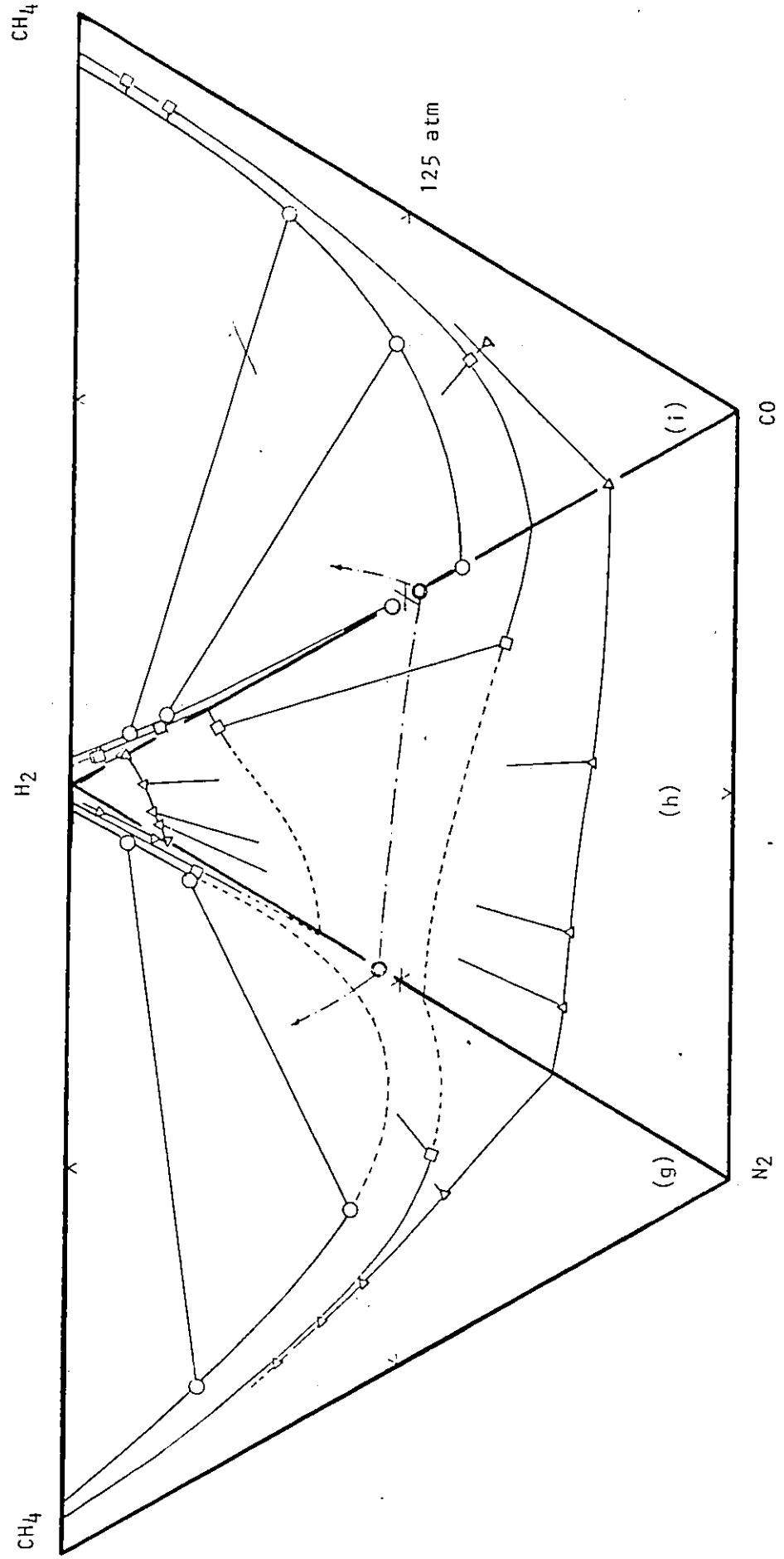


FIGURE III-2: (CONTINUED)

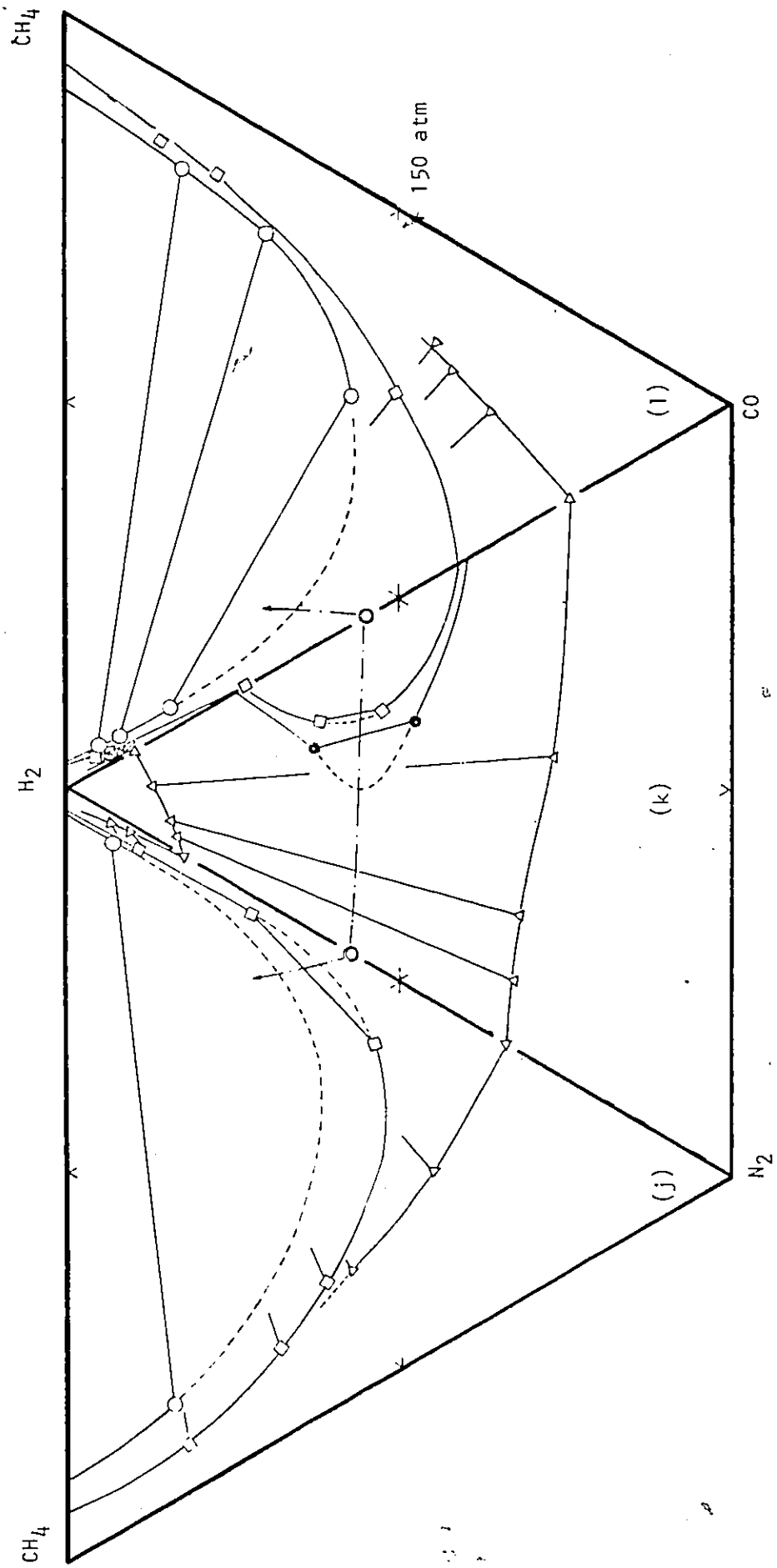


FIGURE III-2: (CONTINUED)

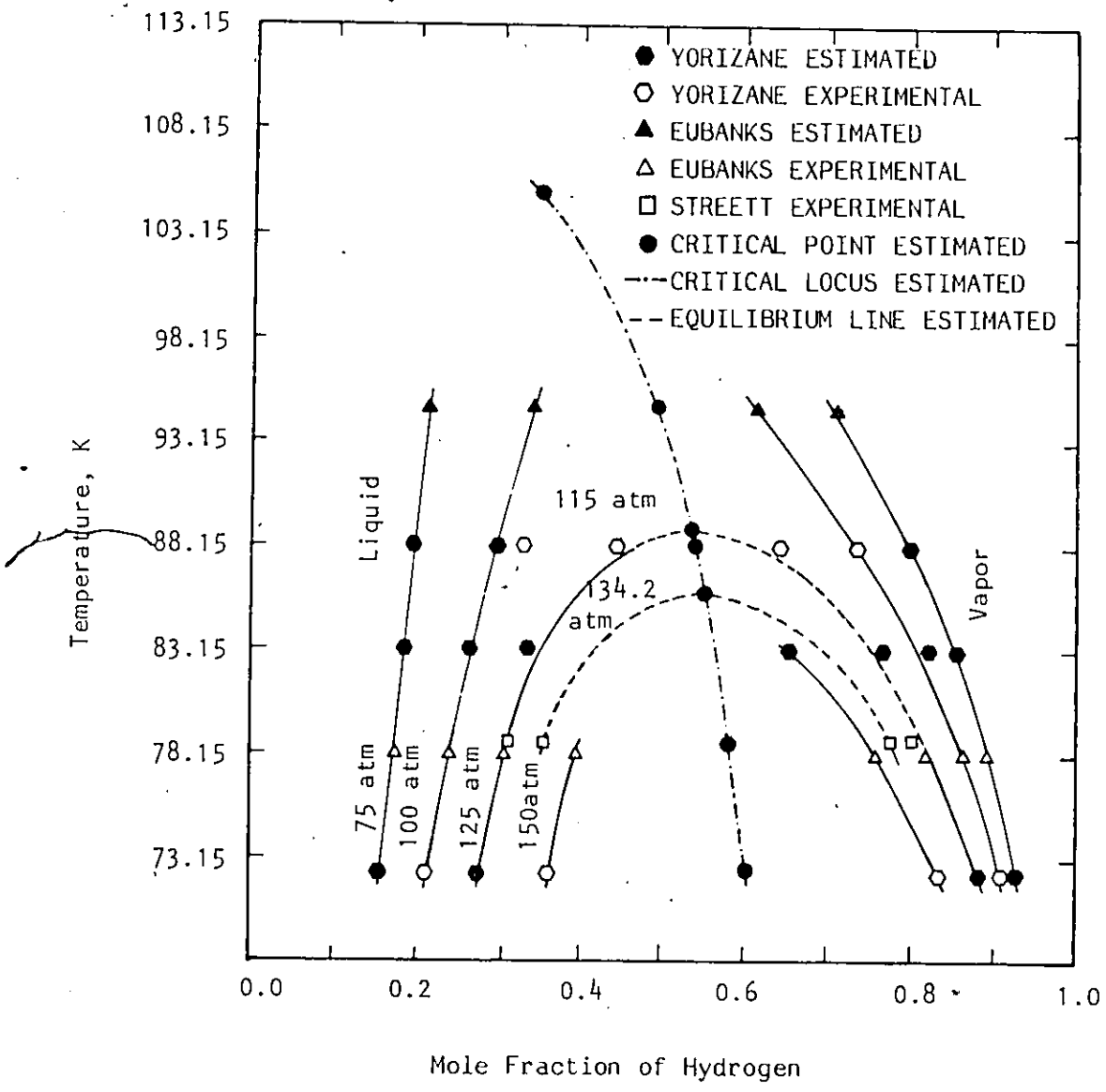


FIGURE III-3: TEMPERATURE-COMPOSITION AND CRITICAL LOCUS DIAGRAM FOR THE BINARY SYSTEM $N_2(1)$-$H_2(2)$ AT SIX ISOBARIC CONDITIONS

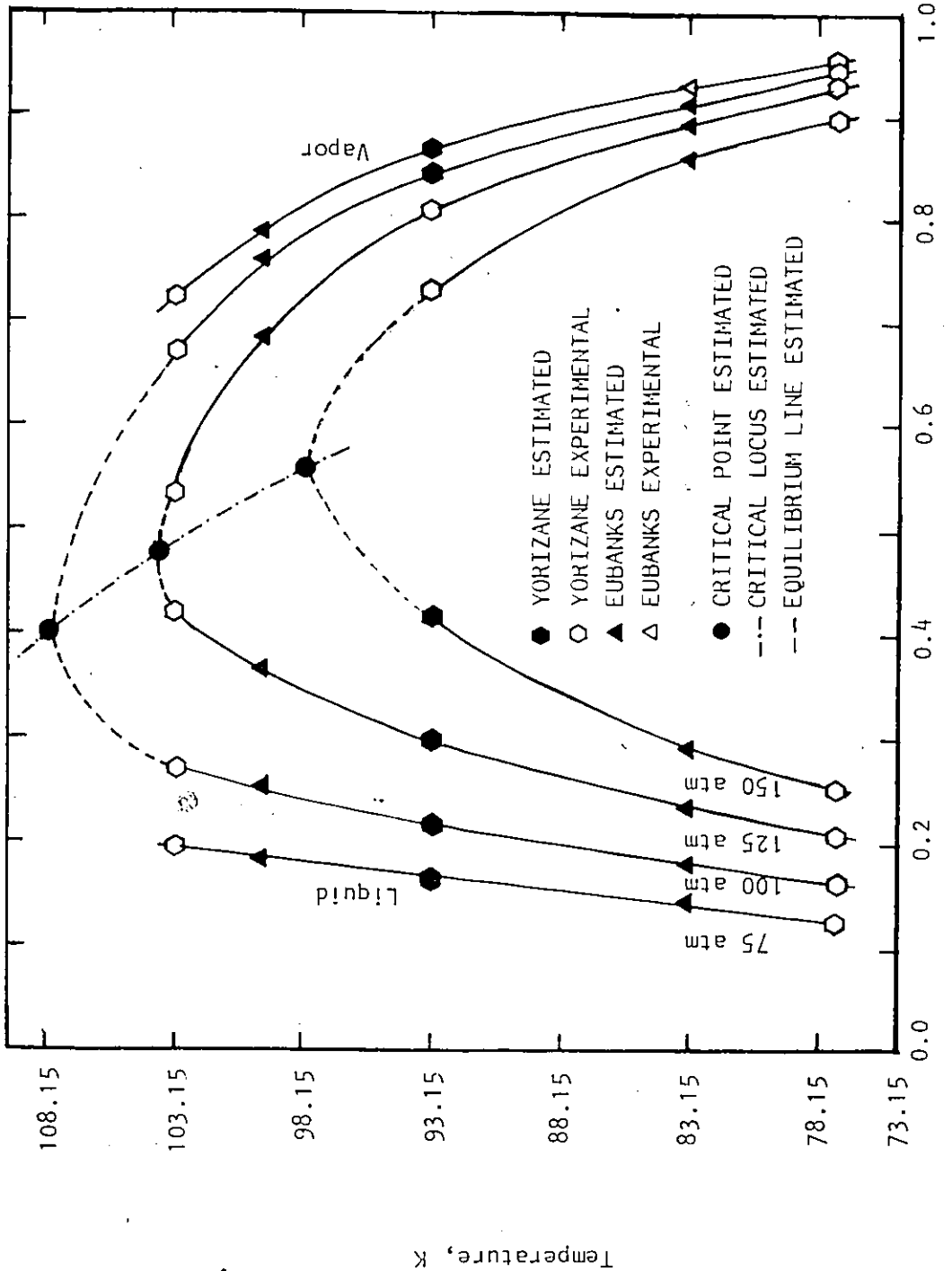


FIGURE III-4: TEMPERATURE-COMPOSITION AND CRITICAL LOCUS DIAGRAM FOR THE BINARY SYSTEM H_2-CO AT FOUR ISOBARIC CONDITIONS

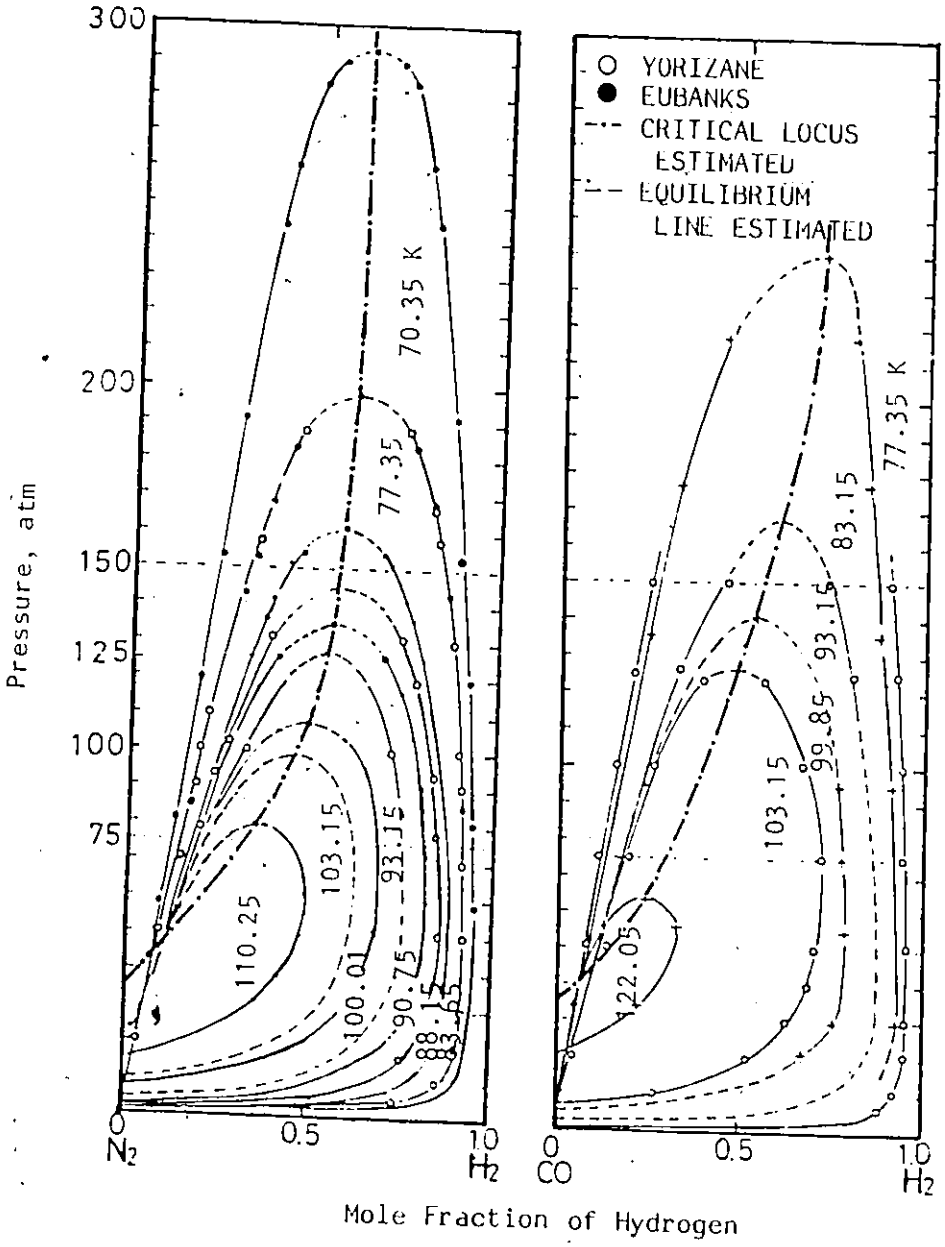


FIGURE III-5: PRESSURE-COMPOSITION AND CRITICAL LOCUS DIAGRAMS FOR THE BINARY SYSTEMS H_2-N_2 AND H_2-CO

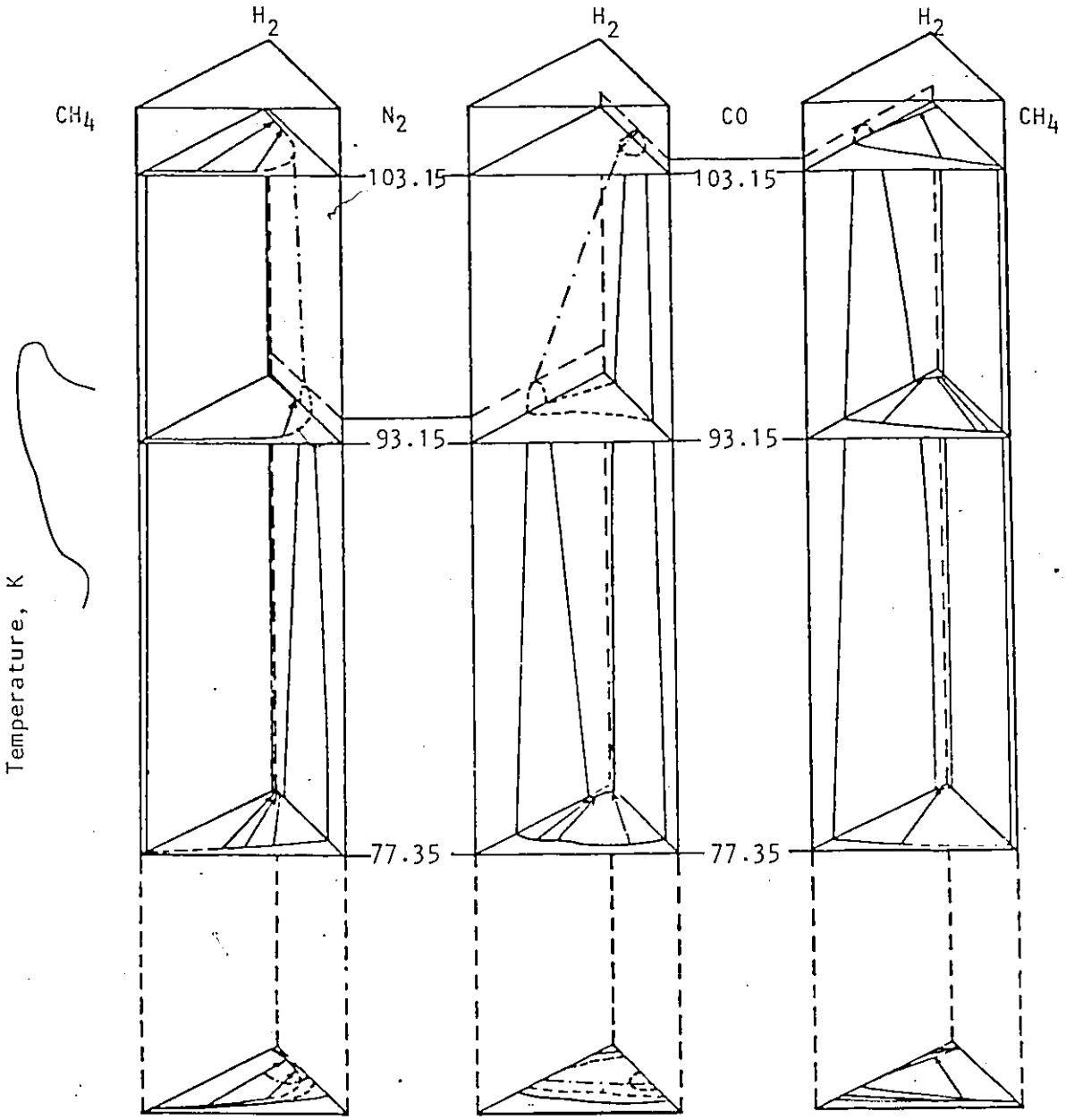


FIGURE III-6: SCHEMATIC THREE-DIMENSIONAL TEMPERATURE-COMPOSITION DIAGRAMS FOR THE TERNARY SYSTEMS H_2-N_2-CH_4, H_2-N_2-CO AND H_2-CO-CH_4 AT 125 atm (→ TIE LINE ---- ESTIMATED -•- ESTIMATED TERNARY CRITICAL LOCI)

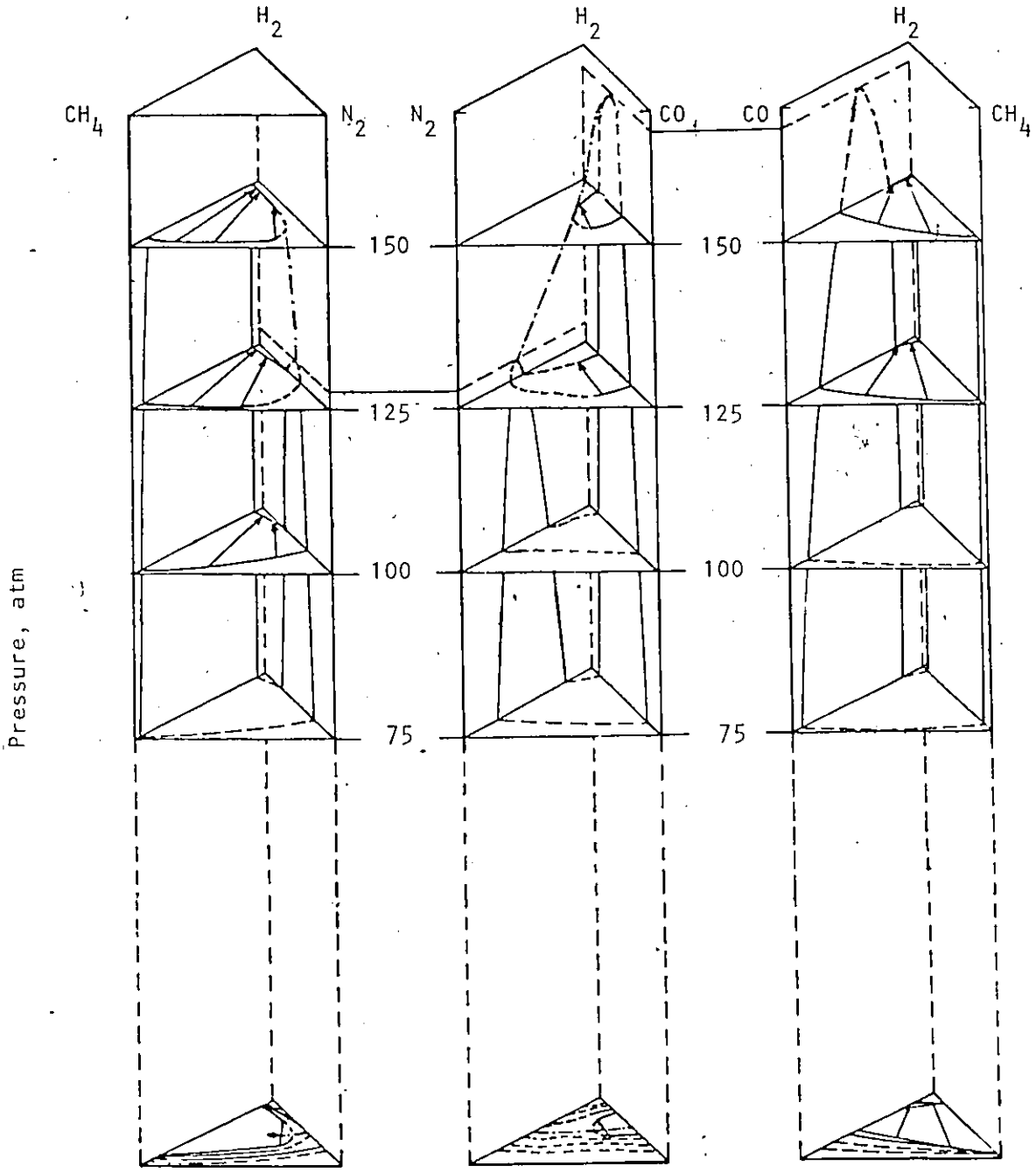


FIGURE III-7: SCHEMATIC THREE-DIMENSIONAL PRESSURE-COMPOSITION DIAGRAMS FOR THE TERNARY SYSTEMS H_2-N_2-CH_4, H_2-N_2-CO AND H_2-CO-CH_4 AT 93.15 K (→ TIE LINE ---- ESTIMATED -•- ESTIMATED TERNARY CRITICAL LOCUS)

Equilibrium vapor and liquid compositions are presented in these diagrams together with the estimated binary critical points and the ternary critical loci.

It can be seen from Figures III-1 to III-5 that binary critical points have been measured in the systems hydrogen-nitrogen and hydrogen-carbon monoxide in the pressure range between 100 and 150 atm and the temperature range between 103.15 and 93.15 K. Ternary critical points have also been established in all three hydrogen containing ternary systems. In the hydrogen-nitrogen-carbon monoxide system, the probable location of the ternary critical locus which passes through the binary critical points in the temperature and pressure region mentioned above, is illustrated in Figures III-1, III-2, III-6 and III-7. Binary critical points for the hydrogen-methane system have also been established, but at temperature and pressures higher than those depicted in Figures III-1 to III-7. Nevertheless, the probable ternary critical locus for the ternary system hydrogen-nitrogen-methane is indicated in these figures. In Figure III-2, it is also shown that there is a tendency for the ternary critical loci of the systems hydrogen-nitrogen-methane and hydrogen-carbon monoxide-methane to spread out as the system pressure decreases.

As shown in Figure III-1 to III-6, the phase behavior of these systems is inevitably affected by the conditions (P, T, x and y) of critical points. These observations are discussed below:

1. For the ternary system hydrogen-nitrogen-carbon monoxide and in the intermediate region of the ternary critical locus between the two binary critical points, the shape of the ternary bubble-point and dew-point curve is strongly influenced by the presence of binary critical points. The appearance and disappearance of ternary critical points are very sensitive to small changes in temperature and pressure.
2. In the vicinity of a binary critical point, the slopes of the tie lines are affected by the presence of the critical points, thus influencing the position of the ternary critical points.
3. In the temperature and pressure region away from the binary critical points, the influence of the critical points on the phase behavior of ternary mixtures is small but not necessarily negligible. For example, the curvature of the bubble-point curves for the ternary system hydrogen-nitrogen-carbon dioxide is affected from 150 atm down to 50 atm at 77.35 K, as shown in Figure III-1. However, this influence disappears for this system around 20 atm as shown graphically by Ruhemann and Zinn (71).

The phase behavior of the ternary system hydrogen-nitrogen-methane is very similar to that of the ternary system hydrogen-carbon monoxide-methane in the temperature and pressure ranges considered in this study. This observation can be attributed by the similarity in the critical properties of nitrogen and carbon monoxide, which are very close to each other.

It is also observed that in the temperature range considered, the solubility of hydrogen in liquid mixtures increases with the increase of temperature.

For the two ternary systems containing methane, the lack of data in the methane rich region at 77.35 K is due to the difficulties encountered in sampling. Liquid solutions are partially solidified at this low temperature.

In addition, phase behavior for the quaternary system hydrogen-nitrogen-carbon monoxide-methane is shown in a three dimensional triangular pyramid in Figure III-8. Estimated critical loci of the quaternary system at 150 atm and three isothermal conditions are also indicated in this figure.

III-B. BINARY INTERACTION COEFFICIENTS FOR VLE CALCULATIONS AT LOW TEMPERATURES

III-B1. Evaluation of C_{ij} Values

The binary interaction coefficients, C_{ij}, are needed for using the ICL equation of state to calculate VLE values. The optimum value of C_{ij} for a binary system at a specific temperature was obtained by minimizing the difference between the calculated and experimental values of equilibrium liquid phase composition. Six binary systems were considered for the mixtures containing hydrogen, nitrogen, carbon monoxide, and methane at cryogenic temperatures. The binary interaction coeffi-

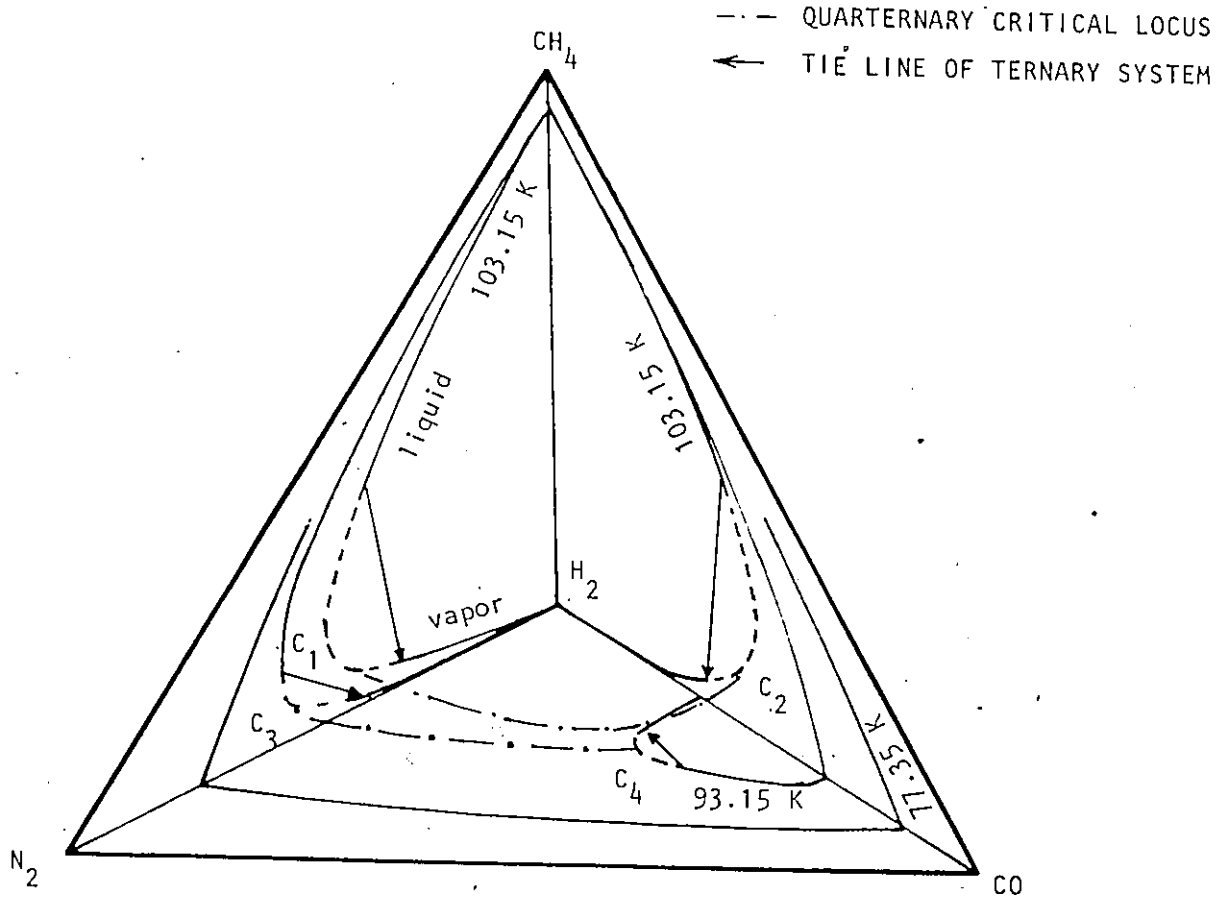


FIGURE III-8: CRITICAL POINTS AND CRITICAL LOCI OF THE TERNARY AND QUATERNARY SYSTEMS CONTAINING H_2, N_2, CO AND CH_4 AT 150 atm

cients for these systems have been determined using a total of 553 data points from 84 isotherms. The C_{ij} values thus obtained together with the average absolute deviations between the experimental and calculated equilibrium compositions are reported in Table III-1.

III-B2. Correlation of C_{ij} Values

It was observed that the C_{ij} values obtained for the six binary systems varied with temperature. Furthermore, the numerical values of C_{ij} depended on the data source. Therefore all the C_{ij} values obtained were then plotted against temperature as shown in Figure III-9, and a linear regression analysis was made to correlate the C_{ij} values with temperature. The equation obtained is:

$$C_{ij} = \ell + m\,T \qquad \text{(III-1)}$$

The results of the regression analysis are listed in Table III-2 and also plotted in Figure III-9. In the temperature range studied, the C_{ij} values for mixtures containing hydrogen decrease with increasing temperature. Furthermore, negative C_{ij} values are obtained for the hydrogen-containing mixtures. For the other three binary systems, the C_{ij} values increase with increasing temperature and they are positive. The values obtained for the coefficients ℓ and m of Equation III-1 are reported in Table III-2. In addition, the average absolute deviations between the calculated and correlated C_{ij} values are also included in this table.

Table III-1 Correlation of VLE Data for Six Binary Systems

System	T,K	N	C_{12}	$\|\Delta x\|_{av}$	$\|\Delta y\|_{av}$	Ref.
Hydrogen-Nitrogen	70.35	12	-0.0112	0.0223	0.0422	82
	77.55	12	-0.0237	0.0160	0.0216	82
	83.15	4	-0.0408	0.0068	0.0153	1
	83.67	5	-0.0609	0.0099	0.0593	82
	88.15	8	-0.0631	0.0114	0.0168	102
	90.03	8	-0.0272	0.0017	0.0008	50
	90.79	10	-0.0586	0.0185	0.0204	82
	95.00	3	-0.0485	0.0011	0.0010	50
	99.82	5	-0.0635	0.0076	0.0211	1
	100.00	10	-0.0641	0.0156	0.0281	82
	110.30	10	-0.0738	0.0103	0.0193	82
Hydrogen-Carbon Monoxide	77.35	6	-0.0101	0.0099	0.0105	104
	83.15	4	-0.0248	0.0040	0.0104	1
	99.82	5	-0.0625	0.0066	0.0098	1
	103.15	3	-0.0850	0.0030	0.0294	104
	122.04	2	-0.0986	0.0014	0.0192	1
	123.15	2	-0.1499	0.0226	0.0394	104
Hydrogen-Methane	90.72	9	-0.0679	0.0007	0.0007	43
	93.15	3	-0.0280	0.0087	0.0075	100
	103.07	5	-0.0925	0.0002	0.0010	43
	103.15	5	-0.0922	0.0060	0.0076	100
	103.15	5	-0.1266	0.0023	0.0083	72
	109.99	7	-0.1095	0.0004	0.0013	43
	116.48	4	-0.1656	0.0313	0.0052	6
	116.51	6	-0.1209	0.0008	0.0017	43
	123.15	6	-0.1272	0.0006	0.0210	72
	123.15	4	-0.1486	0.0038	0.0274	100
	143.05	6	-0.1653	0.0015	0.0088	72
	143.15	5	-0.0873	0.0117	0.0308	100
	144.26	5	-0.2174	0.0245	0.0147	6
	163.15	4	-0.2126	0.0171	0.0398	100
	172.04	4	-0.1119	0.0015	0.0081	6
	172.05	2	-0.2491	0.0047	0.0139	72
	173.65	9	-0.2132	0.0067	0.0172	72

Table III-1 (continued)

System	T,K	N	C_{12}	$\|\Delta x\|_{av}$	$\|\Delta y\|_{av}$	Ref.
Nitrogen-Carbon Monoxide	70.00	4	0.0007	0.0277	0.0279	106
	75.00	4	0.0027	0.0178	0.0176	106
	79.20	4	0.0046	0.0204	0.0277	106
	83.82	9	0.0153	0.0268	0.0355	78
	90.10	6	0.0157	0.0306	0.0411	106
	96.70	6	-0.0006	0.0421	0.0428	106
	100.00	7	0.0091	0.0182	0.0250	106
	105.10	5	0.0185	0.0230	0.0362	106
	110.00	10	0.0071	0.0099	0.0160	106
	116.60	6	0.0074	0.0076	0.0137	106
	121.80	3	0.0077	0.0021	0.0128	106
Nitrogen-Methane	90.67	9	0.0290	0.0086	0.0034	78
	91.65	4	0.0385	0.0019	0.0420	12
	95.00	7	0.0404	0.0056	0.0008	44
	97.15	4	0.0402	0.0028	0.0402	12
	99.82	11	0.0372	0.0073	0.0122	15
	100.00	9	0.0395	0.0030	0.0009	44
	105.00	7	0.0448	0.0079	0.0005	44
	105.15	4	0.0376	0.0014	0.0156	12
	110.00	9	0.0445	0.0079	0.0009	44
	110.93	14	0.0426	0.0067	0.0037	15
	113.71	8	0.0555	0.0084	0.0020	97
	114.57	4	0.0390	0.0002	0.0115	12
	115.00	8	0.0533	0.0115	0.0028	44
	120.00	8	0.0523	0.0112	0.0027	44
	122.04	14	0.0468	0.0042	0.0069	15
	122.04	11	0.0635	0.0123	0.0033	83
	126.04	15	0.0513	0.0100	0.0067	15
	127.59	15	0.0679	0.0080	0.0120	83
	133.15	14	0.0493	0.0034	0.0095	15
	138.46	9	0.0539	0.0079	0.0095	83
	144.26	12	0.0529	0.0056	0.0105	15
	149.82	8	0.0575	0.0056	0.0059	44
	160.93	12	0.0582	0.0020	0.0122	44
	172.04	8	0.0709	0.0012	0.0056	44

Table III-1 (continued)

System	T,K	N	C_{12}	$\|\Delta x\|_{av}$	$\|\Delta y\|_{av}$	Ref.
Nitrogen-Methane	177.59	10	0.0867	0.0017	0.0049	44
	183.15	8	0.0995	0.0009	0.0032	44
Carbon Monoxide-Methane	90.67	9	0.0172	0.0078	0.0057	78
	91.56	5	0.0351	0.0025	0.0610	12
	97.17	4	0.0299	0.0026	0.0523	12
	105.18	5	0.0250	0.0085	0.0200	12
	113.71	1	0.0210	0.0001	0.0014	88
	114.48	5	0.0287	0.0056	0.0213	12
	122.04	2	0.0404	0.0090	0.0149	88
	123.90	3	0.0303	0.0012	0.0126	12
	130.37	4	0.0382	0.0039	0.0090	88
	138.71	1	0.0544	0.0000	0.0075	88
	144.26	5	0.0207	0.0043	0.0087	88
	152.59	1	0.0324	0.0000	0.0046	88
	158.15	4	0.0298	0.0073	0.0295	88

Table III-2 Correlation of C_{ij} with Temperature by Means of Equation (III-1)

System	ℓ	m	$\lvert\Delta C_{ij}\rvert_{av}$	Temp. Range, K
Hydrogen-Nitrogen	0.0812	-0.001433	0.0091	70.35 - 110.30
Hydrogen-Carbon Monoxide	0.1868	-0.002549	0.0105	77.35 - 123.15
Hydrogen-Methane	0.0626	-0.001550	0.0281	90.72 - 173.65
Nitrogen-Carbon Monoxide	-0.0011	0.000096	0.0049	70.00 - 121.80
Nitrogen-Methane	-0.0128	0.000523	0.0061	90.67 - 183.15
Carbon Monoxide-Methane	0.0157	0.000126	0.0072	90.67 - 158.15

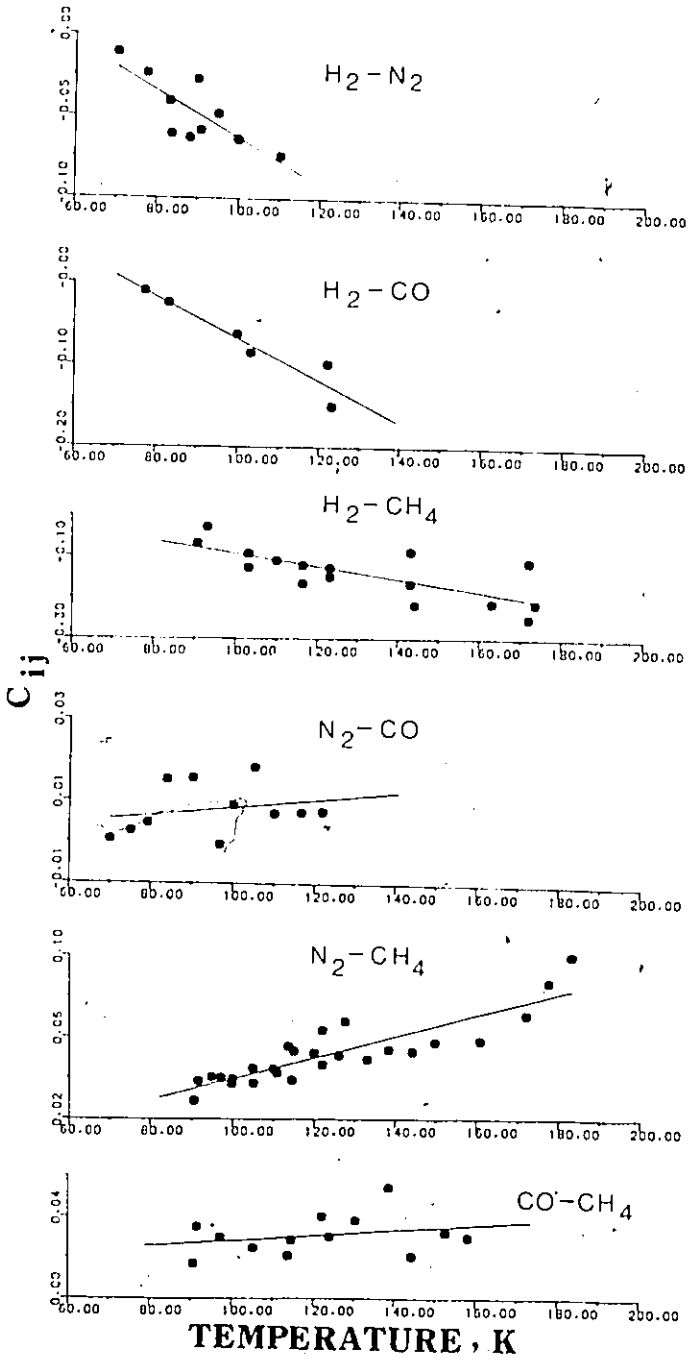


FIGURE III-9: VARIATION OF C_{ij} WITH TEMPERATURE

III-B3. Applicability of the Correlation

For testing the applicability of the proposed correlations, the calculated equilibrium compositions are compared with the experimental values for all the 553 data points of the six binary systems studied in Table A-1a to A-1f, in Appendix A.

The average absolute deviations obtained for Δx and Δy for the systems hydrogen-nitrogen, hydrogen-carbon monoxide, hydrogen-methane, nitrogen-carbon monoxide, nitrogen-methane and carbon monoxide and methane are 0.0130, 0.0218; 0.0075, 0.0163; 0.0063, 0.0117; 0.0209, 0.0274; 0.0061, 0.0075; and 0.0052, 0.0214; respectively. These deviations are only slightly higher than those reported in Table III-1. Hence the proposed correlations are considered acceptable.

An attempt was made to compare the calculated results obtained from the proposed correlations with those reported by Gray (25) for the binary systems hydrogen-methane and hydrogen-nitrogen using the same literature data as listed in Table III-3. A summary of the comparison for two systems, at nine isotherms is presented in Table III-4. Deviations, obtained by minimizing the absolute difference between experimental and calculated equilibrium liquid compositions ($|\Delta x|_{min}$) for calculated methane equilibrium ratios, are lower than those reported by Gray except nitrogen component. For nitrogen, the deviations obtained in this calculation are slightly higher than those reported by Gray. The obtained deviations for hydrogen from this work are lower than those reported by Gray by a factor of 3.42 and 2.30 for the system hydrogen-

Table III-3 Comparison of Experimental and Calculated Equilibrium Compositions and K Values for Two Binary Systems Obtained from the ICL Equation and the Gray's (25) Method

Temp. K	N	C_{12}		$\|\Delta x\|_{av}$		$\|\Delta y\|_{av}$		100 X $\|\Delta K/K\|_{av}$ %		Ref
		$\|\Delta x\|_{min}$	$\|\Delta K_t\|_{min}$	$\|\Delta x\|_{min}$	$\|\Delta K_t\|_{min}$	$\|\Delta x\|_{min}$	$\|\Delta K_t\|_{min}$	$\|\Delta x\|_{min}$	$\|\Delta K_t\|_{min}$	
					Hydrogen-Methane					
90.72	9	-0.0679	-0.0708	0.00065	0.0009	0.0007	0.0007	0.875	0.982	6
								2.915	2.843	
103.07	5	-0.0925	-0.0979	0.00023	0.0006	0.00099	0.0010	0.488	0.799	6
								1.747	1.727	
116.51	6	-0.1209	-0.1333	0.00077	0.0013	0.0017	0.0018	0.936	1.305	6
								1.357	1.261	
109.99	7	-0.1095	-0.1229	0.00038	0.0016	0.0013	0.0011	0.541	1.404	6
								1.531	1.073	
116.48	4	-0.1656	-0.1262	0.03125	0.0383	0.00516	0.0024	6.985	8.846	12
								4.095	1.722	
144.26	5	-0.2174	-0.2174	0.02451	0.0245	0.0147	0.0147	5.536	5.537	12
								3.375	3.378	
172.04	4	-0.11195	-0.1098	0.00151	0.0017	0.00813	0.0083	1.335	1.426	12
								0.855	0.896	
					Hydrogen-Nitrogen					
83.15	4	-0.0408	-0.0317	0.00684	0.0128	0.01528	0.0169	3.315	3.961	104
								6.361	7.481	
99.82	5	-0.0635	-0.0444	0.00757	0.0136	0.02105	0.0206	4.157	4.181	104
								3.639	3.831	

Table III-4 Comparison of Equilibrium Compositions and K Values Obtained from the ICL Equation with those Values Reported by Gray (25) for Two Binary Systems

Temperature - Range K	Number of Points	$\|\Delta x\|_{av}$		$\|\Delta y\|_{av}$		$\|(\Delta K)/K\|_{av}$ X 100% in RMS			Ref
		$\|\Delta x\|_{min}$	$\|\Delta K_t\|_{min}$	$\|\Delta x\|_{min}$	$\|\Delta K_t\|_{min}$	$\|\Delta K_{min}\|$*	$\|\Delta x\|_{min}$	$\|\Delta K_t\|_{min}$	
				Hydrogen-Methane					
90.72 - 172.04	40	0.0067	0.0078	0.0039	0.0037	6.9	2.017	2.482	6,12
						3.1	2.263	1.917	
				Hydrogen-Nitrogen					
83.15 - 99.82	9	0.0072	0.0132	0.0124	0.0190	8.7	3.783	4.083	104
						4.6	4.849	5.453	

*calculated values reported by Gray.

-methane and the system hydrogen-nitrogen, respectively. By the method of minimizing the absolute difference between experimental and calculated K values ($|\Delta K_t|_{min}$), the calculated deviations for hydrogen from this work are lower than those reported by Gray (25) by a factor of 2.78 and 2.13 for the binary system hydrogen-methane and the binary system hydrogen-nitrogen, respectively.

The binary system hydrogen-methane (32) at ten isothermal conditions was chosen to compare the difference between VLE compositions calculated (by the ICL equation) by using the evaluated binary interaction coefficients and those obtained by using correlated $C_{H_2-CH_4}$ values as represented by Equation (III-1). The results are reported in Tables A-2a and A-2b, in Appendix A. The absolute average deviations obtained for Δx and Δy for the binary system are summarized in Table III-5.

The numerical values given in the parenthesis in Table III-5 indicated the number of data points which were convergent in the calculation. The total number of data points, N, are also indicated in the table. It was apparent that the difference between the results obtained by using the evaluated C_{ij} values and those obtained by the correlated C_{ij} values were insignificant although the number of convergent data points by using the evaluated C_{ij} values in the calculation were slightly lower.

In addition, the C_{ij} values as represented by Equation (III-1) are used to predict equilibrium compositions for three hydrogen-containing ternary systems for a total of ten isotherms. The calculated

Table III-5 Comparison of Predicted and Experimental Equilibrium Compositions (32) for the Binary System $H_2(1)$-$CH_4(2)$ at Ten Isothermal Conditions

T,K	N	C_{ij}		$\|\Delta x\|_{av}$		$\|\Delta y\|_{av}$	
		Evaluated	Correlated	Evaluated	Correlated	Evaluated	Correlated
163.11	17	-0.2212	-0.1478	0.0058(14)	0.0038(17)	0.0134	0.0206
173.24	14	-0.2059	-0.2260	0.0184(14)	0.0116(14)	0.0304	0.0308
163.16	16	-0.1903	-0.2495	0.0398(14)	0.0327(16)	0.0192	0.0298
153.20	20	-0.1748	-0.2416	0.0595(17)	0.0259(20)	0.0456	0.0328
143.07	16	-0.1591	-0.1736	0.0747(16)	0.0670(14)	0.0597	0.0334
132.94	12	-0.1434	-0.1748	0.0110(11)	0.0122(12)	0.0086	0.0099
123.10	11	-0.1282	-0.1425	0.0070(11)	0.0036(11)	0.0040	0.0034
118.12	8	-0.1205	-0.1349	0.0075(8)	0.0026(8)	0.0051	0.0040
113.13	11	-0.1127	-0.1201	0.0069(11)	0.0045(11)	0.0053	0.0049
108.12	10	-0.1050	-0.0959	0.0037(10)	0.0040(10)	0.0016	0.0019

VLE results for ternary hydrogen-nitrogen-carbon monoxide system at three isotherms, hydrogen-nitrogen-methane system at one isotherm and hydrogen-carbon monoxide-methane system at six isotherms are all shown in Table B-1 in Appendix B. A brief summary of the satisfactory results obtained is presented in Table III-6.

III-C. CALCULATION AND PREDICTION OF VLE VALUES BY USING EQUATIONS OF STATE

III-C1. Hydrogen Containing Ternary Systems

The ICL equation was used to calculate and to predict vapor compositions as well as x_1 and x_2 by fixed T, P and x_3 for three hydrogen containing systems hydrogen(1)-nitrogen(2)-carbon monoxide(3), hydrogen(1)-nitrogen(2)-methane(3), and hydrogen(1)-carbon monoxide(2)-methane(3) at three isotherms in the pressure range from 75 atm to 150 atm (method 1). A comparison of experimental and calculated VLE compositions for these systems was made. The results are reported in Table B-2 in Appendix B. The values of C_{ij} for the six constituent binary systems were determined using experimental values reported in the literature and correlated in terms of a linear function of temperature as mentioned in section III-B. The C_{ij} values used in this study for the ICL equation are reported in Table III-7.

For the purpose of comparison, the Redlich-Kwong equation as modified by Soave (76) was also used for the same calculations.

Table III-6 Comparison of Experimental and Predicted Equilibrium Compositions for Three Hydrogen Containing Ternary Systems

T,K	N	Pressure Range, atm	$\|\Delta x\|_{av}$	$\|\Delta y\|_{av}$	Ref
		Hydrogen-Nitrogen-Carbon Monoxide			
83.15	17	21.43 - 136.08	0.0040	0.0088	1
99.82	29	21.43 - 95.26	0.0053	0.0093	1
122.04	6	34.02 - 54.43	0.0039	0.0162	1
		Hydrogen-Nitrogen-Methane			
144.26	8	34.02 - 68.05	0.0015	0.0182	17
		Hydrogen-Carbon Monoxide-Methane			
108.12	10	27.22 - 68.04	0.0060	0.0148	31
113.13	21	27.22 - 102.06	0.0060	0.0098	31
123.10	40	27.22 - 102.06	0.0033	0.0074	31
143.07	33	27.22 - 102.06	0.0039	0.0191	31
163.16	33	27.22 - 102.06	0.0039	0.0157	31
173.24	10	40.82 - 102.06	0.0025	0.0136	31

Table III-7 Values of Binary Interaction Coefficient for Mixtures of Hydrogen(1), Nitrogen(2), Carbon Monoxide(3), and Methane(4) Used in the ICL Method

Temp., K	C_{12}	C_{13}	C_{14}	C_{23}	C_{24}	C_{34}
77.35	-0.0307	-0.0104	-0.0473	0.0063	0.0276	0.0238
93.15	-0.0534	-0.0507	-0.0818	0.0078	0.0359	0.0262
103.15	-0.0677	-0.0762	-0.0973	0.0088	0.0411	0.0278

In the application of the Soave-RK equation, the method advanced by Graboski and Daubert (24) (method 2) was adopted, and the values of the interaction coefficients recommended by them were used in the calculations. Those interaction coefficient values are reported in Table III-8. The calculated results obtained by this method for three hydrogen containing systems are reported in Table B-3, in Appendix B.

The calculated results of these two methods for the three ternary systems are summarized in terms of average absolute deviations for Δx and Δy and reported in Table III-9. The numerical numbers given in the parenthesis indicate the number of data points which were convergent in the calculation. The total number of data points included in the calculation for the ternary system hydrogen-nitrogen-carbon monoxide at the temperature of 77.35 and 103.15 K are 11 and 3 respectively; for the ternary system hydrogen-nitrogen-methane at the temperature of 77.35, 93.15, and 103.5 K, are 11, 7, and 11 respectively; and for the ternary system hydrogen-carbon monoxide-methane at the temperature of 77.35, 93.15 and 103.15 K, are 10, 5, and 10 respectively.

The comparison indicates that not all the data points were convergent at the three temperatures by the two methods. However, it is seen clearly that the prediction of VLE compositions by the two methods are very successful.

III-C2. Quaternary System Hydrogen-Nitrogen-Carbon Monoxide-Methane

The ICL equation was used to predict the vapor compositions as well as liquid compositions of hydrogen and nitrogen for the quaternary

Table III-8 Values of Binary Interaction Coefficient Recommended by the Graboski and Daubert Method (24)

Components	Interaction Coefficient
H_2 - Others	0.0
N_2 - CO	0.0460
N_2 - CH_4	0.0319
CO - CH_4	0.0300

Table III-9 Comparison of VLE Compositions Obtained From Two Calculation Methods with Experimental Values for Three Hydrogen Containing Ternary Systems

Ave.Abs.Dev.	Method	Temperature, K		
		77.35	93.15	103.15
		Hydrogen-Nitrogen-Carbon Monoxide		
Δx	1	0.0076 (11)	-	0.0187 (3)
	2	0.0642 (3)	-	0.0115 (2)
Δy	1	0.0115	-	0.0129
	2	0.0056	-	0.0139
		Hydrogen-Nitrogen-Methane		
Δx	1	0.0115 (11)	0.0108 (6)	0.0130 (11)
	2	0.0240 (9)	0.0124 (6)	0.0070 (11)
Δy	1	0.0047	0.0143	0.0054
	2	0.0038	0.0268	0.0066
		Hydrogen-Carbon Monoxide-Methane		
Δx	1	0.0108 (10)	0.0339 (5)	0.0145 (10)
	2	0.0244 (8)	0.0306 (4)	0.0140 (10)
Δy	1	0.0047	0.0084	0.0066
	2	0.0045	0.0078	0.0095

system at 77.35, 93.15 and 103.15 K in the pressure range between 75 and 105 atm. This calculation was carried out by fixed system temperature, pressure and liquid compositions of carbon monoxide and methane. The values C_{ij} for the six constituent binary systems used in this calculation were the same as those reported in Table III-7. The calculated VLE compositions for the quaternary systems at three isotherms are reported in Table C-1, in Appendix C. The calculated hydrogen and nitrogen compositions are compared with the experimental values in Figure III-10 to III-13 to illustrate the agreement obtained. Similar results were obtained for carbon monoxide and methane of the quaternary system. In addition, a summary of the average absolute deviations between experimental and calculated equilibrium compositions from method 1 are reported in Table III-10.

For the purpose of comparison, the modified SRK equation (76) (method 2), and the generalized correlation of Lee and Kesler (45) as applied by Plocker et al. (61) to vapor-liquid equilibrium calculations (method 3) were also tested for their capability of representing the VLE data of the quaternary system. In the calculation procedure of Plocker et al., it was recommended that temperature-dependent effective critical constants be used for systems containing quantum gases. However, it was observed in this study that temperature-independent effective critical constants would yield better results for the quaternary system.

This was also true for the three binary systems containing hydrogen: hydrogen-nitrogen, hydrogen-carbon monoxide, and hydrogen-methane. For this reason, the calculated results used in the comparison were

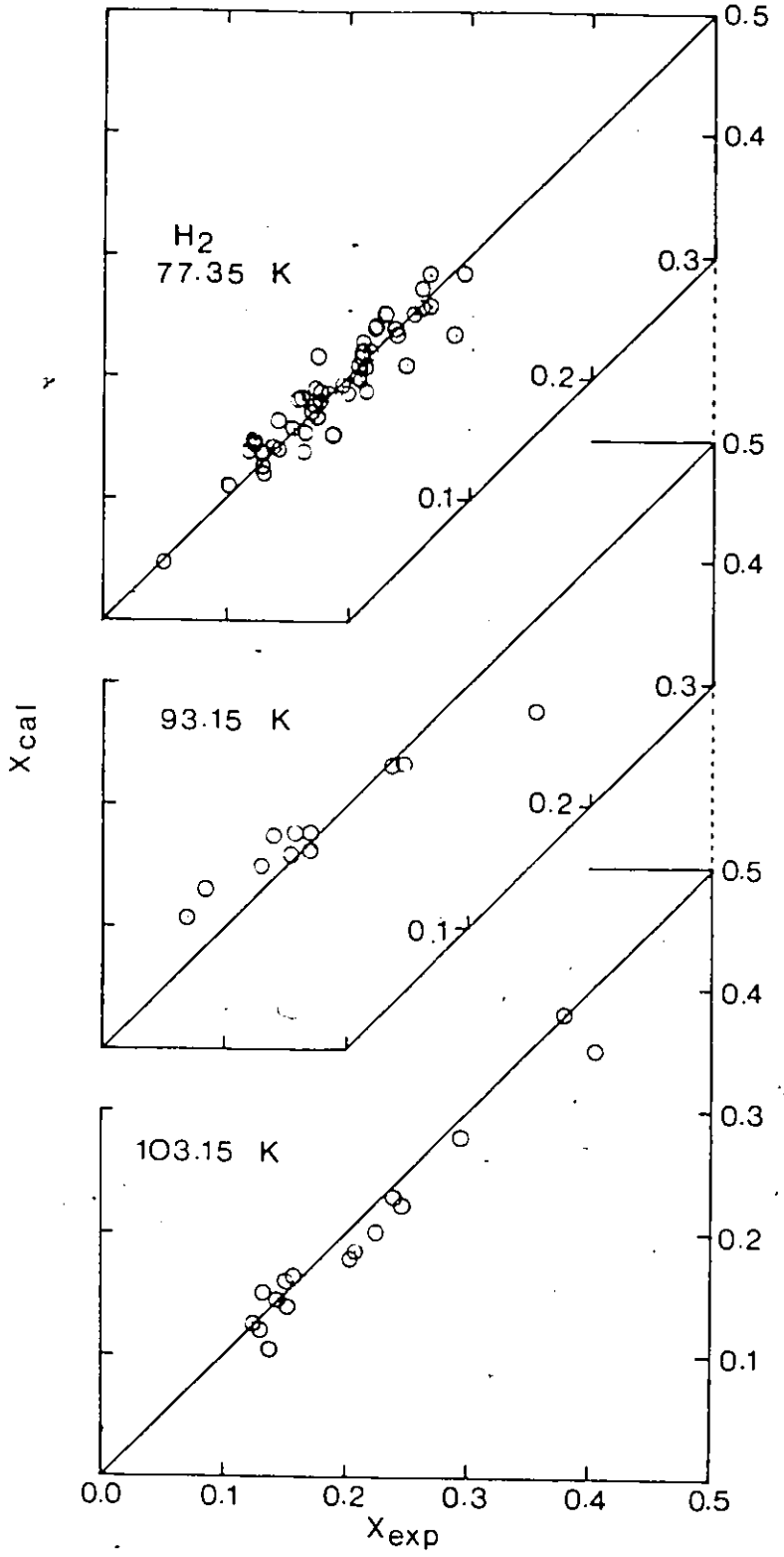


FIGURE III-10: COMPARISON OF CALCULATED AND EXPERIMENTAL LIQUID COMPOSITIONS OF H_2 IN THE QUATERNARY SYSTEM $H_2(1)$-$N_2(2)$-$CO(3)$-$CH_4(4)$

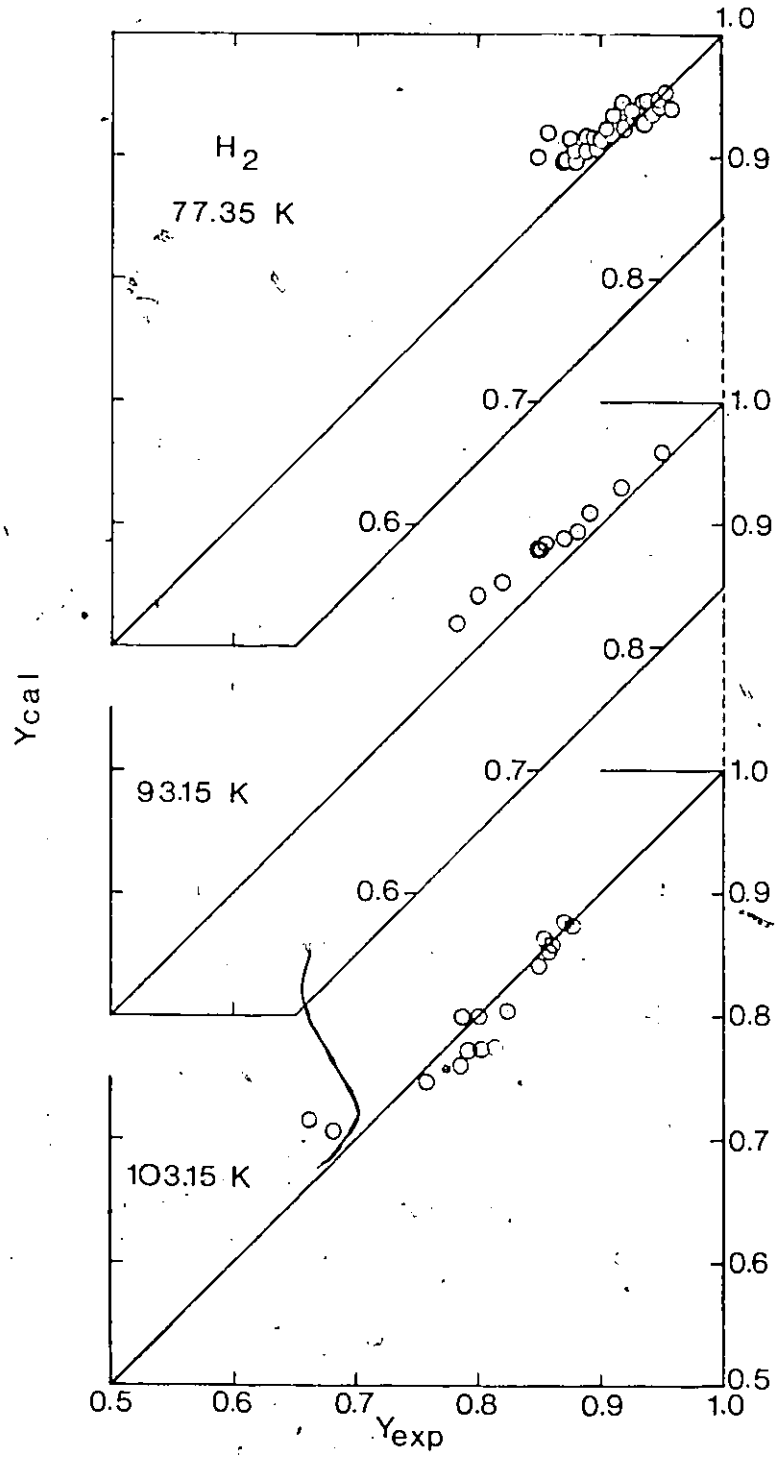


FIGURE III-11: COMPARISON OF CALCULATED AND EXPERIMENTAL VAPOR COMPOSITIONS OF H_2 IN THE QUATERNARY SYSTEM $H_2(1)$-$N_2(2)$-$CO(3)$-$CH_4(4)$

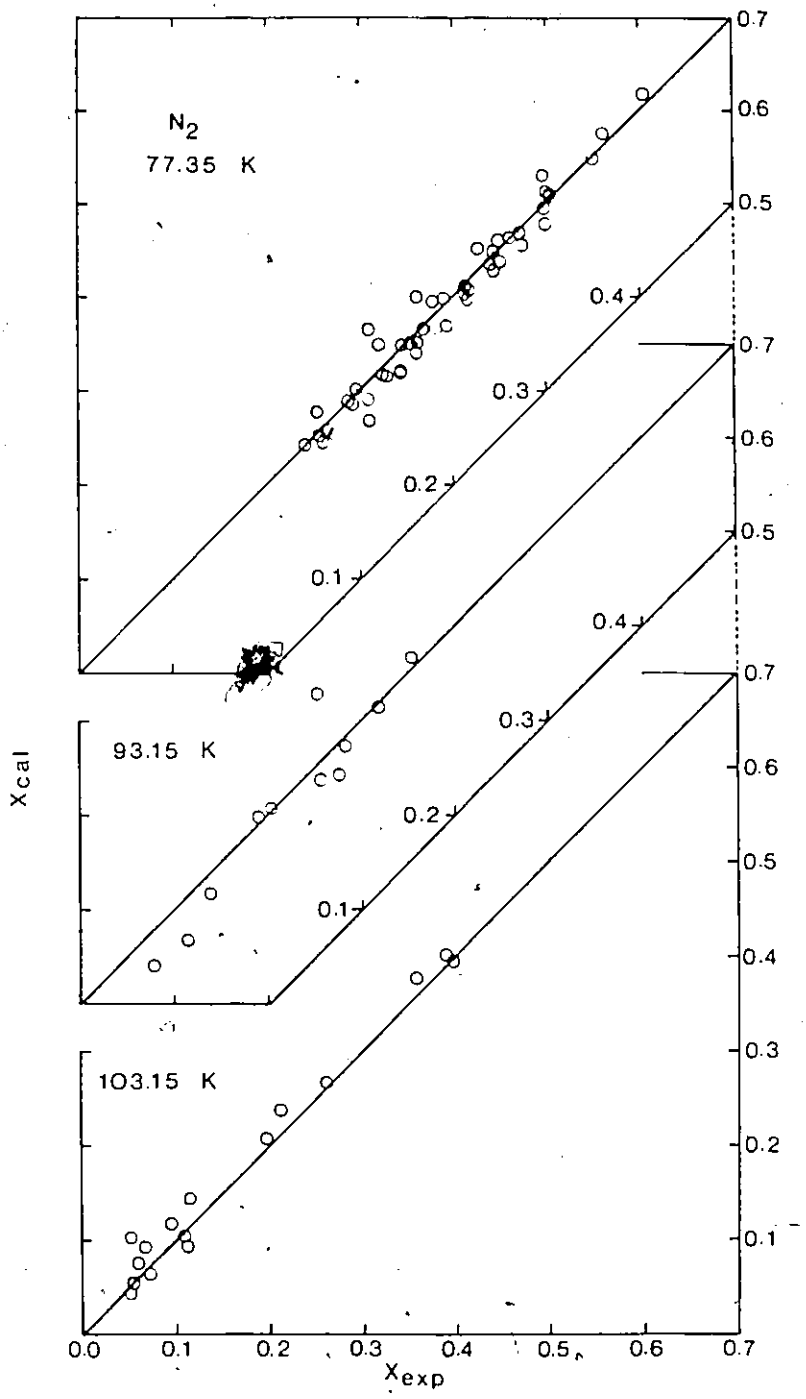


FIGURE III-12: COMPARISON OF CALCULATED AND EXPERIMENTAL LIQUID COMPOSITIONS OF N_2 IN THE QUATERNARY SYSTEM $H_2(1)$-$N_2(2)$-$CO(3)$-$CH_4(4)$

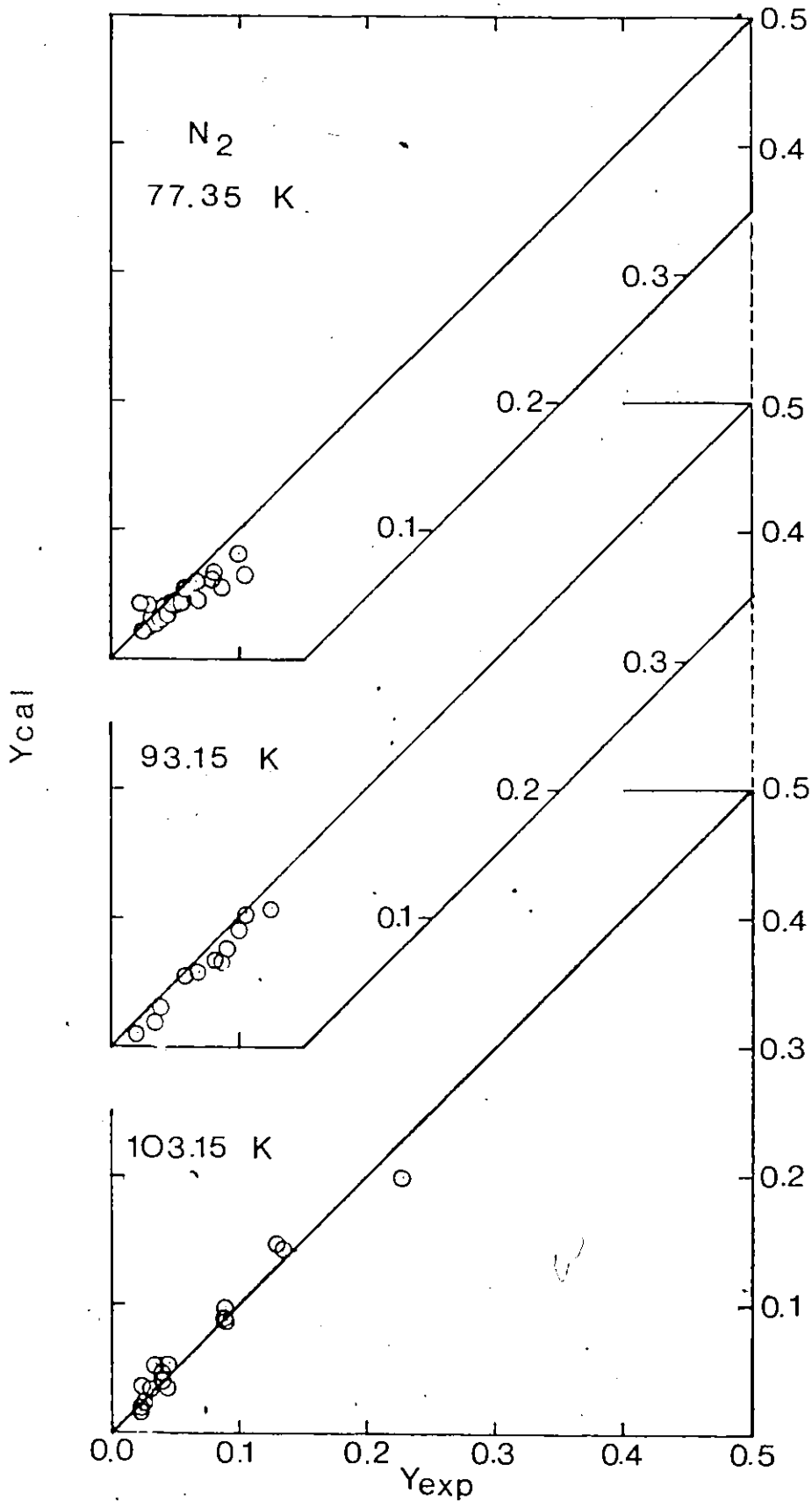


FIGURE III-13: COMPARISON OF CALCULATED AND EXPERIMENTAL VAPOR COMPOSITIONS OF N_2 IN THE QUATERNARY SYSTEM $H_2(1)$-$N_2(2)$-$CO(3)$-$CH_4(4)$

Table III-10 Comparison of Experimental and Calculated Vapor-Liquid Equilibrium Compositions of the Quaternary System Hydrogen(1)-Nitrogen(2)-Carbon Monoxide(3)-Methane(4)

Ave.Abs.Dev.	Method	Temperature, K		
		77.35	93.15	103.15
Δx_1 and Δx_2	1	0.0145 (48)	0.0250 (11)	0.0220 (17)
	2	0.0676 (21)	0,0310 (11)	0.0182 (19)
	3	0.0517 (47)	0.0509 (17)	0.0248 (16)
Δy_1	1	0.0132	0.0251	0.0170
	2	0.0462	0.0384	0.0162
	3	0.0170	0.0267	0.0119
Δy_2	1	0.0090	0.0105	0.0112
	2	0.0245	0.0222	0.0088
	3	0.0059	0.0200	0.0093
Δy_3	1	0.0053	0.0087	0.0072
	2	0.0251	0.0082	0.0066
	3	0.0109	0.0064	0.0061
Δy_4	1	0.0009	0.0077	0.0028
	2	0.0011	0.0091	0.0043
	3	0.0019	0.0038	0.0019

obtained by using the temperature-independent effective critical constants. On the other hand, the values of the binary interaction coefficients recommended by Plocker et al. (61) as shown in Table III-11 were used in method 3 without any modifications.

The calculated results by methods 2 and 3 are reported in Tables C-2 and C-3 in Appendix C and summarized in Table III-10. Average absolute deviations for Δx and Δy are also reported in Table III-10. The numerical numbers given in the parenthesis indicated the number of data points which were convergent in the calculation. The total number of data points included in the calculation at the temperatures of 77.35, 93.15 and 103.15 K are 48, 17 and 19, respectively.

The comparison indicates that not all the data points were convergent at the three temperatures by the three methods. The average absolute deviations obtained by methods 2 and 3 are approximately the same at the three temperatures.

At 103.15 K, all the data points were convergent by method 2. The overall average absolute deviation in the calculated x and y values obtained by method 1 is slightly higher than those obtained by the other two methods.

At 93.15 K, all the data points were convergent by method 3. the overall average absolute deviation in the calculated x and y values obtained by method 1 is lower than those obtained by methods 2 and 3.

Table III-11 Values of Binary Interaction Coefficients Recommended by Plocker et al. (61)

Component	H_2	N_2	CO	CH_4
H_2	-	-	-	-
N_2	1.080	-	-	-
CO	1.085	0.987	-	-
CH_4	1.216	0.977	0.974	-

At 77.35 K, method 2 is the least suitable one with less than half of the data points convergent in the calculation. All the data points were convergent by method 1, which also yielded the lowest average absolute deviations in the calculated x and y values.

CHAPTER IV

CONCLUSIONS

The existence of probable critical points of the binary systems H_2-N_2 and H_2-CO has been presented graphically at the temperatures of 93.15 and 103.15 K in the pressure range between 100 and 150 atm. The probable critical loci of three hydrogen containing ternary systems have been suggested and indicated in the schematic three dimensional temperature-composition and pressure-composition diagrams. The probable critical loci of the quaternary hydrogen-nitrogen-carbon monoxide-methane system are also suggested and indicated graphically.

It has been shown that the binary interaction coefficient, $^{\wedge}C_{i-j}$ can be correlated as a linear function of temperature. The proposed correlations for the six binary mixtures containing hydrogen, nitrogen, carbon monoxide and methane have been satisfactorily applied to the calculation and prediction of VLE values for mixtures containing these components. The average absolute deviations of liquid compositions $|\Delta x|$ obtained for the systems hydrogen-nitrogen, hydrogen-carbon monoxide, hydrogen-methane, nitrogen-carbon monoxide, nitrogen-methane and carbon monoxide-methane vary from 0.0052 to 0.0130. For vapor compositions, $|\Delta y|$, it varies from 0.0075 to 0.0274. These results are considered to be very satisfactory.

It has been found that the application of the ICL equation (method 1) is adequate for correlating and predicting phase equilibrium properties for the binary, ternary and quaternary systems containing hydrogen, nitrogen, carbon monoxide and methane. The application of the modified SRK equation (method 2) is adequate for representing the VLE properties for the hydrogen containing ternary systems consisting of hydrogen, nitrogen, carbon monoxide and methane. It has also been seen that the applications of the modified SRK equation and the modified BWR equation (method 3) are all suitable for representing and predicting phase equilibrium properties for the quaternary system hydrogen-nitrogencarbon monoxide-methane at the three temperatures investigated. For the sake of simplicity and accuracy, the ICL equation and the modified SRK equation are recommended for representing the VLE values of the systems. On the other hand, the ICL equation appears to represent the VLE data best among these three methods for the quaternary system H_2-N_2-CO-CH_4 at the lowest temperature of 77°K.

PART B

EXPERIMENTAL ASPECTS

CHAPTER V

INTRODUCTION

Numerous experimental methods, including static cell, dew and bubble point, flow separator, forced vapor recirculation, rocked cell, total pressure and packed column flow (11, 28, 34, 94, 98) have been used in the past for VLE measurements. A summary of advantages and disadvantages of these methods was discussed by Wilson (98). Among these, the forced vapor recirculation method is one of the most popular methods used at cryogenic conditions. However, the application of forced vapor recirculation method (11, 34, 94) involves the following experimental problems as stated by Wilson (98):

1. The cell often leaks around the window seals at cryogenic temperature due to the contraction of gasket material. When this occurs, the entire assembly must be warmed up and the seals retightened.
2. The vapor recirculation pump fails to function because the magnetically actuated plunger sometimes sticks and/or the check valves fail to operate. Moisture or other material which solidifies at cryogenic temperatures usually is the cause of the problem. Under these circumstances it is also necessary to warm up the entire assembly and dismantle the pump to clear out the obstruction.

In addition, apparatus dead volume, which exists between the sampling line and the equilibrium cell, contributes to significant deviation in VLE measurement. It is a major problem that needs to be overcome. Therefore, the design of a new apparatus to overcome the above mentioned

The VLE data on methane-ethylene system were required in the design of ethylene plant. However the binary data of the system were insufficient in the temperature range from 223 K to 273 K. At 223 K, only limited experimental data were reported by Sagara et al (75). Thus, further studies within this temperature range would be very desirable. The binary system methane-ethylene at 243 K was chosen to be one of the topics studied since desirable binary data was not available in the literature. In addition, more experimental data for the binary system methane-ethylene at 223 K were measured.

It was also found that the binary data of nitrogen-methane system at 103 K as reported by Wang (94) were questionable. Measurements for this system were also carried out in this work.

Table V-1 VLE Data Available in the Literature for Binary Systems Nitrogen-Carbon Dioxide and Methane-Ethylene at the Specified Conditions

System	Temperature range, K	Pressure range, atm	Reference
Nitrogen-Carbon Dioxide	273.15	35 ~ 118	8,40,52,101,107
Methane-Ethylene	100 ~ 273	0 ~ 65	8,27,35,51,72, 89,93

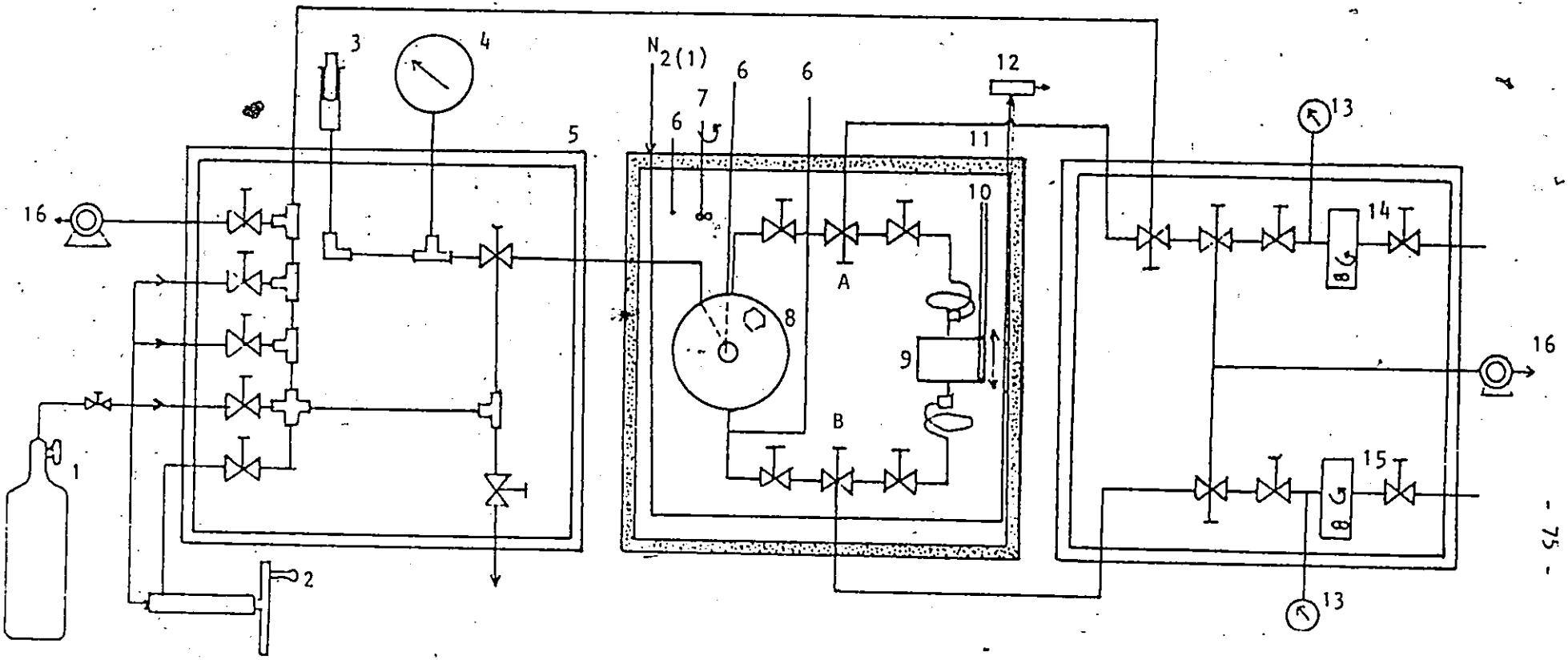
CHAPTER VI

EXPERIMENTAL DETAILS

In this chapter, the design of a new apparatus is presented. It consists of a charging system, two equilibrium cells, a cryostat and temperature control assembly, temperature and pressure measuring devices, a coolant supply system, and a sampling and analysis system. Experimental procedure, safety considerations, as well as the materials used are also described in detail.

VI-A. APPARATUS

A schematic flow diagram of the new apparatus is shown in Figure VI-1. It consisted of a charging system, a cryostat containing a static cell and a movable cell, and a sampling and analysis system. The equipment was tested at its working temperature up to 150 atm, while the charging system was operated at room temperature and the cryostat was maintained at 77.35 K. In this design, the dead volume between the sampling valves and the static equilibrium cell was reduced to the volume of the needle valves (A and B) as shown in Figure VI-1 . The level of the liquid and vapor mixture could be accomplished by moving a equal-volume cell which in turn changed the level of the liquid by a reciprocal motion device. This new design minimized mechnical problems which might be encountered by mechanical devices installed within the cryostat. Furthermore, the liquid level in the static equilibrium cell could be visibly observed during the operation through two glass windows.



1	Gas cylinder	9	Equilibrium cell (type B)
2	Proportioning pump	10	Up and down moving device
3	Pressure relief valve	11	Cryostat
4	Pressure gauge	12	Buffer tank
5	Water vessel	13	Compund gauge
6	Thermocouple	14	Vapor sample
7	Stirrer	15	Liquid sample
8	Equilibrium cell (type A)	16	Vacumm pump

FIGURE VI-1: SCHEMATIC FLOW DIAGRAM OF APPARATUS

VI-A1. Charging System

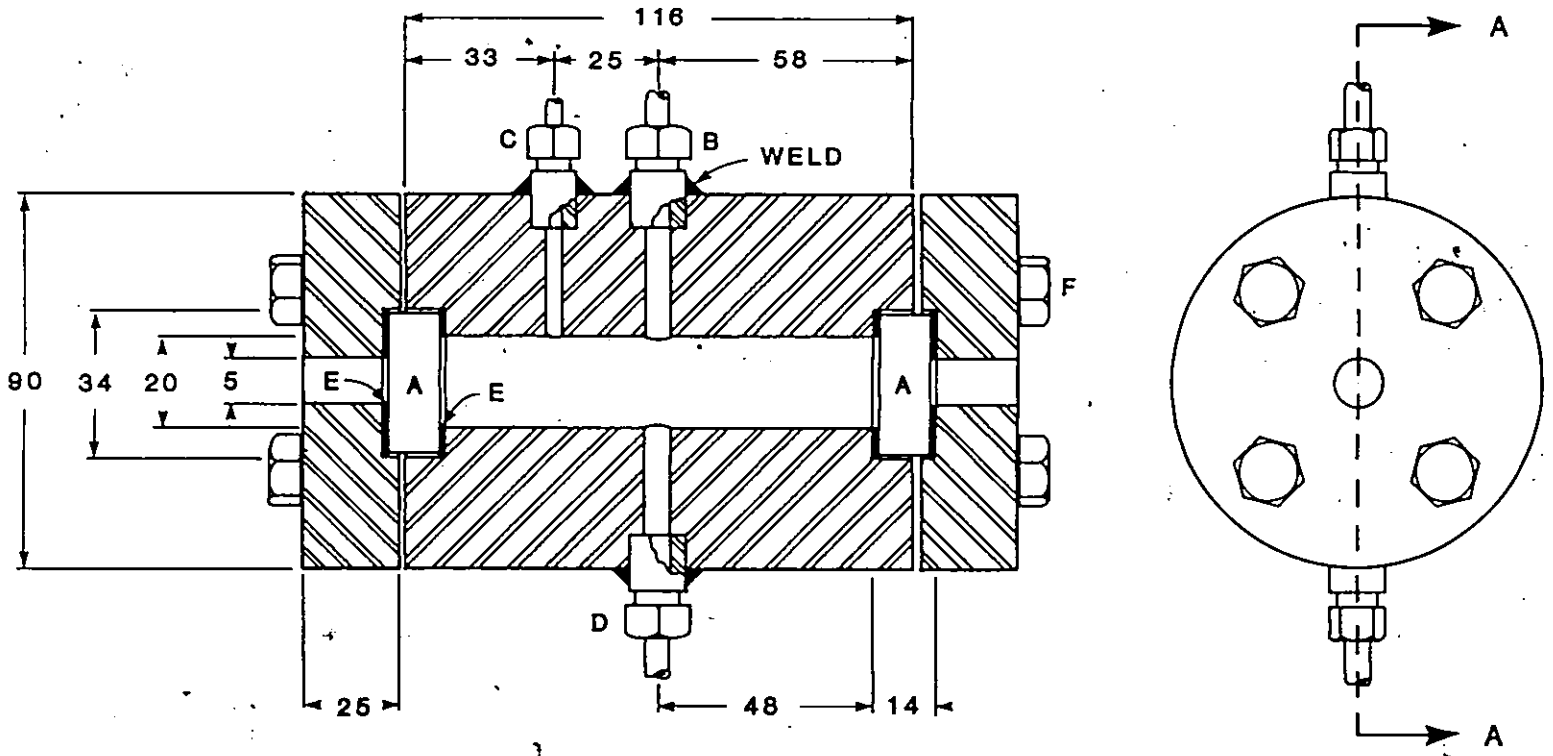
The charging system consisted of a Ruska proportioning pump, a pressure gauge, a pressure relief valve, and a vacuum pump. When the system pressure was low, the required gas was charged directly from the gas cylinder. But when the system pressure was higher than the cylinder pressure, the required gas was charged into the system by means of the Ruska proportioning pump.

VI-A2. Equilibrium Cells

Two equilibrium cells were designed, constructed and used to measure VLE values in this study. One is a static equilibrium cell, and the other is a movable equilibrium cell.

VI-A2a. Static Equilibrium Cell

The 316 stainless steel static equilibrium cell with two transparent glass windows was designed and constructed. A diagram of the cell (Type A) is shown in Figure VI-2. To the top of the cell was welded a 1/4 inch stainless steel Autoclave joint, B, to which the vapor outlet tube was connected. Another 1/8 inch Autoclave joint, C, was welded to the top to permit two protected-type copper constantan thermocouples to be inserted for measuring the temperature of the liquid and the vapor phases. A 1/4 inch stainless steel Autoclave joint, D, was welded to the bottom of the cell. This served as inlet and outlet for circulating liquid as well as inlet for charging the feed.



SECTION A – A (DIMENSIONS IN mm)

A GLASS WINDOWS

B VAPOUR CONNECTION (AUTOCLAVE 1/4")

C THERMOCOUPLE CONNECTION (AUTOCLAVE 1/8")

D LIQUID CONNECTION (AUTOCLAVE 1/4")

E TEFLON GASKETS

F BOLTS (3/8" UNC)

FIGURE VI-2 STATIC EQUILIBRIUM CELL (TYPE A)

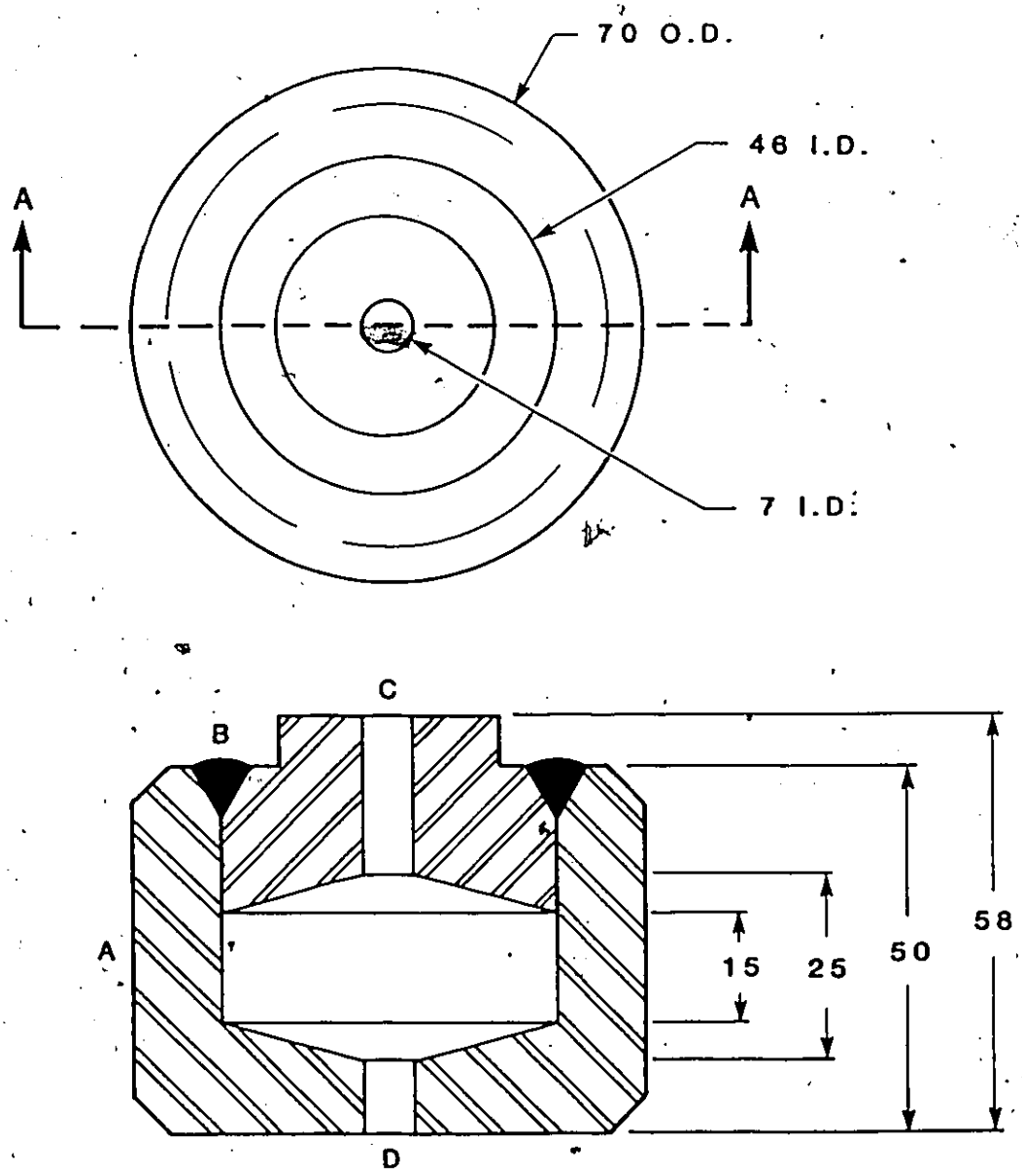
The maximum working pressure of the cell was 150 atm (2205 psia) and the lowest testing temperature was set at 77.35 K (-320.44°F). The cell has a capacity of 30.7 ml with a weight of 12.50 kg (27.50 lbm).

VI-A2b. Movable Equilibrium Cell

A drawing of the movable equlibrium cell (Type B) is shown in Figure VI-3. The body was made of 316 stainless steel with a wall thickness of 12 mm. To both ends of the cell were welded with 1/4 inch stainless steel Autoclave joints which were served as inlet and outlet for the circulated fluids. The capacity of the cell was identical to that of the static equilibrium cell but with a weight of 1.23 kg (2.71 lbm). The maximum test pressure was 150 atm at 77.35 K.

VI-A3. Cryostat and Temperature Control Assembly

The equipment shown in the center section of Figure VI-1 was mounted on a frame and submerged in the cryostat bath. The layout of this equipment is shown in Figure VI-4 and the photograph of Figure VI-4A. All needle valves in this assembly had valve extensions so that they could be operated from above the bath liquid. The movable cell was moved in an up and down motion using a motor driven Boston gear to accomplish mixing in the cell system. The liquid level in the static equilibrium cell could be observed through windows in the cryostat wall. A 48 liter stainless steel cylinder was used as the cryostat vessel. Liquefied 2-methylbutane was used as the bath liquid. The cryostat was equipped with a refrigeration coil (for liquid nitrogen),



SECTION A - A (DIMENSIONS IN mm)

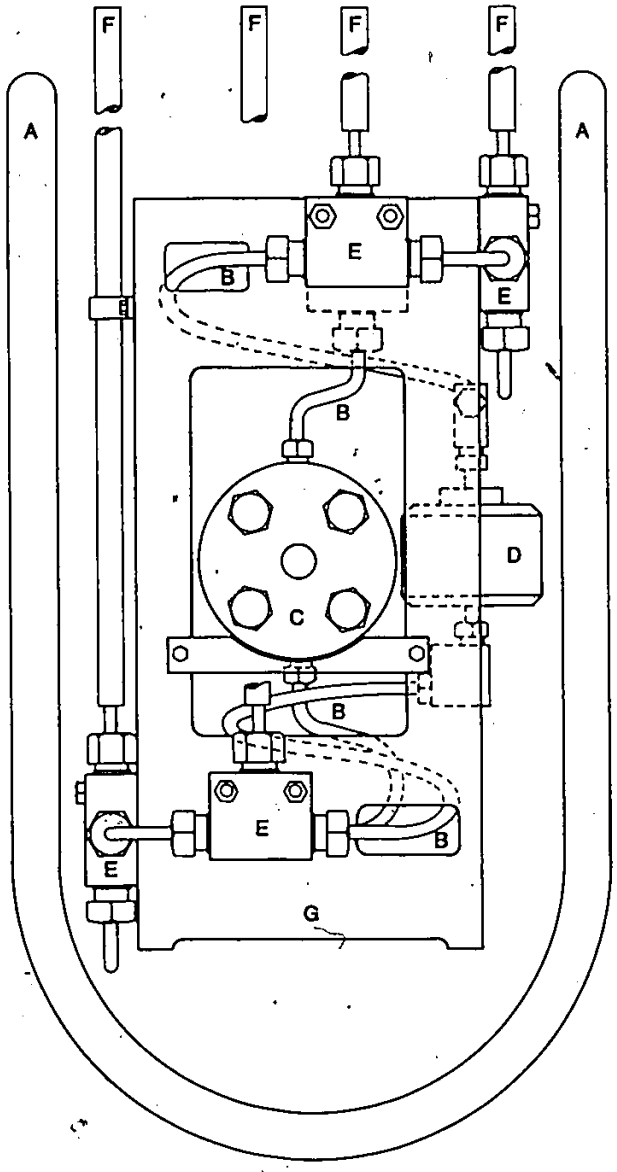
A BODY (316 STAINLESS STEEL)

B WELDED JOINT

C VAPOUR CONNECTION (AUTOCLAVE 1/4")

D LIQUID CONNECTION

FIGURE VI-3 MOVABLE EQUILIBRIUM CELL (TYPE B)



A CRYOSTAT

B CONNECTION TUBES

C STATIC CELL

D MOVABLE CELL

E VALVES

F VALVE HANDLE EXTENSIONS

G SUPPORT FRAME

FIGURE VI-4 EQUILIBRIUM CELLS INSTALLED INSIDE THE CRYOSTAT

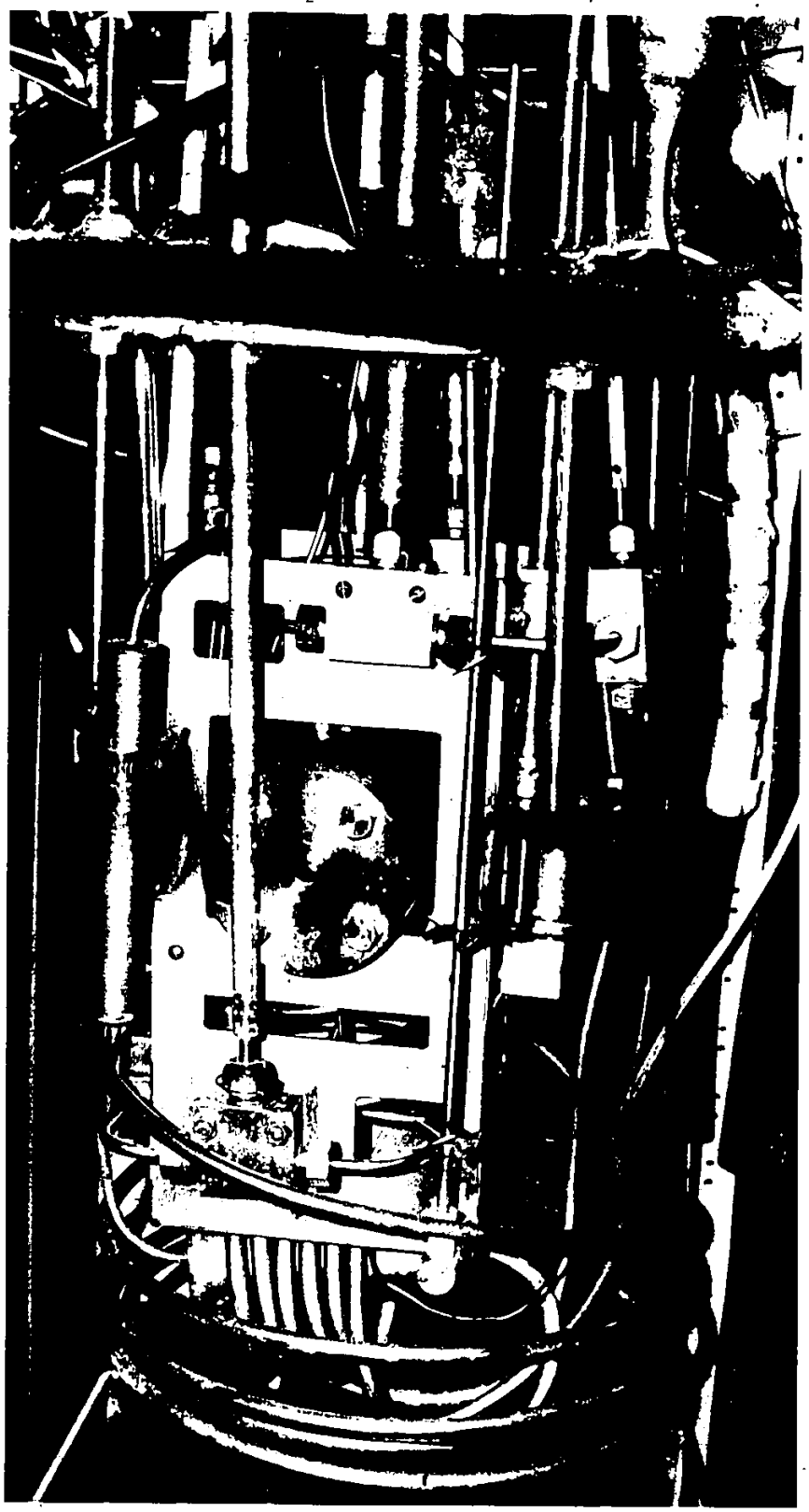


FIGURE VI-4A

two stirrers, one heating element, two copper-constantan thermocouples, and a resistance type temperature sensing element. The temperature of the cryostat was maintained by liquid nitrogen flowing through the refrigeration coil. At the downstream end of the coil, a buffer tank and a needle valve was located to keep the flow rate of nitrogen constant. The heating element was used to supply heat to keep the temperature constant in cryostat.

VI-A4. Temperature and Pressure Measuring Devices

Four protected type copper-constantan thermocouples were installed in this apparatus. Two of them were located in the static equilibrium cell (one in the vapor phase, and the other in the liquid phase). The other two were installed in the cryostat (one at one quarter height, and the other at three-quarters height of the bath). The system temperature was measured using a Leeds and Northrup 7555 type K-5 potentiometer. The temperature of the cryostat was measured with a platinum resistance thermometer and controlled through a Bayley precision temperature controller (Model 250) that manipulated the heat into the bath through the heating element. This system controlled the bath temperature with an accuracy of ± 0.1 K.

Two calibrated Heise gauges with ranges of 0 to 34.0 atm and 0 to 136.1 atm, and with 0.034 and 0.136 atm subdivisions, respectively, were used for measuring lower system pressures. In addition, a high pressure gauge with range of 0 to 340.2 atm and with 0.68 subdivision was also provided. The uncertainties involved in the pressure measure-

ments are + 0.1 atm below 136 atm, and + 0.7 atm between 150 and 350 atm.

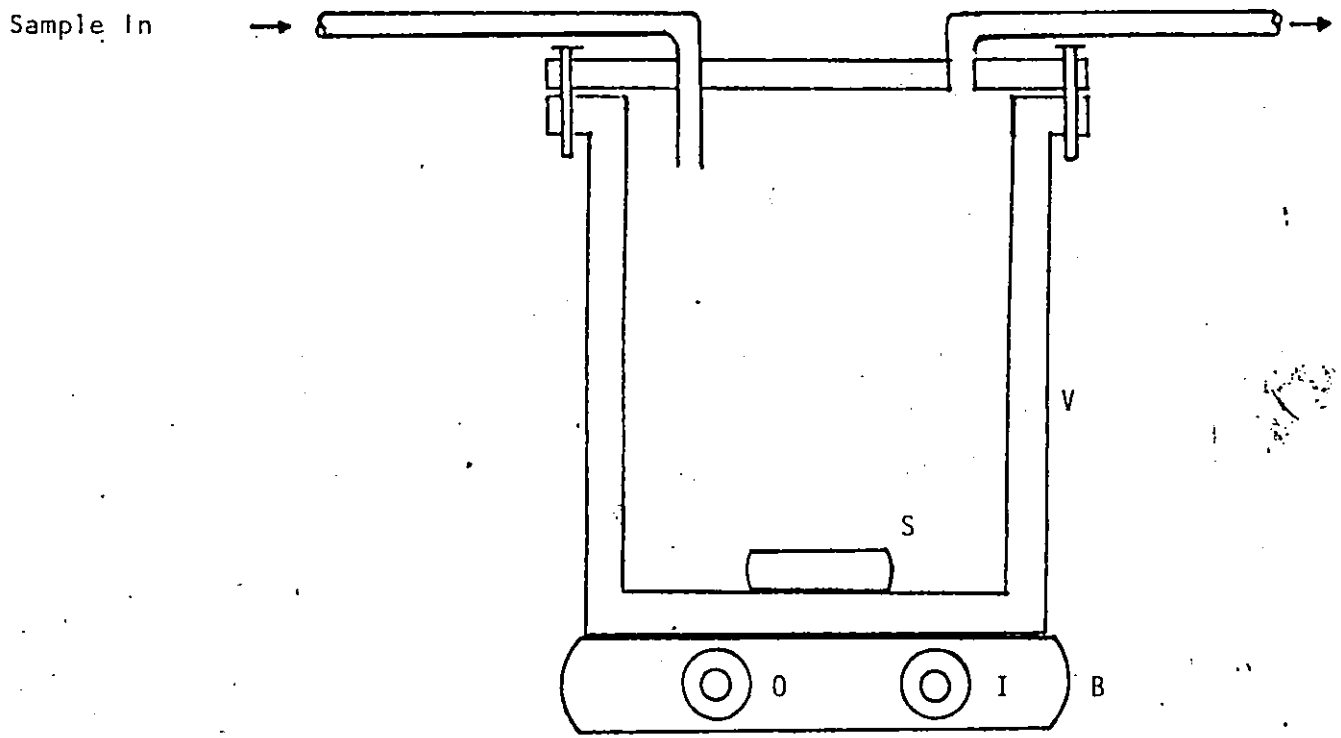
VI-A5. Coolant Supply System

Liquefied nitrogen was used as a refrigerant to maintain the system at the desired temperature. Compressed dry nitrogen was used to push the liquefied nitrogen through the refrigeration coil in the cryostat. The heat was removed from cryostat by the vaporization of liquid nitrogen in the coil.

VI-A6. Sampling and Analysis System

Two stainless steel 316 vessels, 14 and 15, were used as sample collectors, as shown in Figure VI-1, one of which served as a liquid sampler with a capacity of 1000 milliliters, and the other as a vapor sampler with a capacity of 175 milliliters. The design of vapor sampler is shown in Figure VI-5. To prevent any condensation during analysis, the vapor mixture was uniformly mixed by a magnetic stirrer, S, inside the sampler before each analysis was made by gas chromatography. The liquid sampler was similar in design to that of vapor sampler except its capacity.

A Hewlett Packard 5830A gas chromatograph was used for composition analyses. It was equipped with a Hewlett Pakard Accessory 18825A Valve Driver Board. A Hewlett Packard 18850A GC terminal was used to record and to integrate the chromatographic curves.



B Air Drive Magnet Base
I Compressed Air Inlet
O Compressed Air Inlet
S Magnetic Stirrer
V Vessel

FIGURE VI-5: SCHEMATIC DIAGRAM OF THE VAPOR SAMPLER

VI-B. MATERIALS USED

Research grade nitrogen, methane, ethylene, and carbon dioxide supplied by Matheson of Canada Limited, Whitby, Ontario were used in this investigation. Minimum purities of the gases were reported by the supplier as follows:

Component	Minimum Purity, mole %
Nitrogen	99.9995
Methane	99.99
Ethylene	99.98
Carbon Dioxide	99.99

These gases were used in this study without any further purification.

VI-C. EXPERIMENTAL PROCEDURE

Data could be taken with either ascending or descending system pressure. If descending pressure method were used, gases would have to be vented from the system to lower the pressure for the next experimental condition. After only a few such ventings, the amount of liquid phase in the equilibrium cell would be greatly reduced. Eventually, it would fall below an acceptable liquid level. It was thus more convenient to acquire data using the ascending pressure method since the amount of liquid phase increased when more gas was added to the cells. In this study, the ascending pressure method was used for all experiments. Detailed experimental procedures will be discussed in the following subsections.

VI-C1. Temperature and Pressure Control

As mentioned earlier, a Bayley temperature controller was used to maintain the temperature of the fluid in the cryostat.

The system temperature could be lowered by evaporating liquid nitrogen which was passed through the coil submerged in the cryostat liquid bath. The nitrogen evaporating rate was adjusted to bring the bath temperature just slightly below the desired temperature, to ensure that the heat removed by the nitrogen was just sufficient so that the bath temperature would decrease slowly. The temperature controller was turned on for about 20 minutes before being used to bring the unit to full sensitivity. In case of over cooling of the cryostat interior, the system temperature could be raised using an electric heating element which was controlled by the Bayley temperature controller. A good technique was required to maintain a delicate balance between the heat supplied by the heating element and the heat removed by the cooling coil.

The system temperature was measured by a Leeds and Northrup 7555 type potentiometer. The temperature of the constant-temperature bath was determined using two protected copper constantan thermocouples located at different positions. The temperature inside the cell was determined using two protected copper constantan termocouples located in the vapor and liquid phases. It was observed that the temperature difference between the inside of the cell and the bath was very small

at equilibrium and was insignificant. The thermocouples were calibrated using the vapor pressure of methane or ethane following the method of Prydz and Goodwin (64).

When the desired initial system pressure was to be below 10 atm, it was established manually by first adding the least volatile component from the high pressure supply cylinder. After the first gas supply was cut off, the other gas component was compressed to the desired pressure using the Ruska proportioning pump supplied by the Ruska Instrument Corporation. The pump could be either motor driven or hand operated depending on the pressure change desired. The total displacement of the pump was 500 ml. This pump was connected directly to the Heise pressure gauges.

VI-C2. Preparation of a Run

For precise measurement, the entire apparatus required thorough cleaning to remove oil, solid particles, and other contaminants before each investigation. The desired cleanness was achieved by rinsing the system with solvents and then by alternately evacuating and flushing with dry helium. Benzene, carbon tetrachloride, and petroleum ether were used as solvents. All pieces of the apparatus were individually cleaned. After the system was assembled, it was alternately flushed with dry helium and the main component of the mixture to be studied. A positive pressure (>3 atm) was always maintained in the system to minimize the contamination from the surrounding atmosphere.

VI-C3. Vapor-Liquid Equlibrium Measurements

To begin an experiment, the entire system was evacuated for at least 8 hours and then flushed with the main component of the desired mixture. This procedure was repeated several times when a new system was studied.

After the desired bath temperature was reached, gases were charged into the cells with the least volatile component introduced first, then followed by components in the order of increasing vapor pressure. The gas being charged was roughly estimated by observing the liquid level in the equilibrium cell. The desired pressure was set approximately by adding the most volatile component last using the proportioning pump. Further adjustments of the pressure could be made by changing a small amount of the most volatile gas during the early stage of the operation.

The level of the liquid phase in the static cell could be adjusted by changing the level of the movable equilibrium cell. This motion circulated the liquid mixture between the movable and the static equilibrium cells through a 1/4 inch stainless steel tube connected at the bottom of the cells. At the same time, the vapor mixture in the two cells were connected through a 1/4 inch stainless steel tube. The mixing motion was continued until the desired temperature and pressure were reached and remained constant. During the experiment, both the vapor and the liquid phases inside the submerged static equilibrium cell

could be visually observed through two glass windows in the cryostat wall.

When the pressure remained at a constant value, the system was stabilized for at least one hour to assure the equilibrium condition was reached. A small portion of vapor in the equilibrium cells was discharged to the sampler through the vapor sampling lines, and then a very small amount of liquid sample was discharged to the liquid sampler. The sampling valves were only partially opened to allow the withdrawal of an extremely small amount of the sample from the cells. With the slight but continuous withdrawal of sample, the compositions of the sample mixture in the sampling lines were assumed to be the same as those in the equilibrium cells.

In order to prevent any condensation, an initial mixing period of thirty minutes in the vapor sampler was used. After that, the vapor sample was analyzed using a Hewlett Packard 5830A gas chromatograph. The liquid sample was mixed in the liquid sampler for about 70 minutes before analysis. At least six analyses of each sample were made. After the vapor and the liquid samples were analyzed, the system pressure was adjusted for the next run.

The pressure was read, in psia, (pounds per square inch, absolute) using the Heise pressure gauges. This data can be converted to the SI unit, pascal (Pa = $N.m^{-2}$) by applying the multiplying by the conversion

factor of 6,894.757, or to values in atmospheres by dividing by the conversion factor of 14.696.

VI-C3a. Composition Analyses

All gas mixture compositions were analyzed by the gas-chromatograph equipped with a 6 or 10 ft long, 1/8 inch diameter stainless steel column, packed with Porapak Q, mesh 80-100. The gas chromatograph was equipped with a thermal conductivity detector. Helium was used as the carrier gas.

The results of gas chromatography analyses were calibrated against several synthetic samples of known compositions. The analytical results of the known compositions were expressed in terms of composition ratios and plotted against peak area ratios. The ratio plot was adopted instead of the absolute quantity plot, because it was relatively insensitive to any change in detector current, temperature, column pressure, carrier gas flow rate, sample quantity, recorder sensitivity, etc. These plots are shown in Appendix D.

VI-D. SAFETY CONSIDERATIONS

Safety is always a prime consideration in any high pressure work. The use of highly explosive gases in this study made safety considerations even more desirable. Some special precautions employed in this investigation are outlined below:

1. All parts of the apparatus subject to high pressures were directly connected to a blow out safety valve with a rupture disk manufactured by the Autoclave Engineers.
2. Major parts of the apparatus, including the valves, were subjected to commissioning tests at pressures up to the maximum pressure before the parts were assembled.
3. Special precautions were required when 2-methylbutane was used as bath fluid at cryogenic conditions. Positive ventilation was maintained through the measurement. Flame and sparks were prohibited in the laboratory.
4. All parts of the apparatus except those in the cryostat were submerged in water so that any small gas leak could be easily noticed during the measurement.
5. Before each experimental run, the system was thoroughly checked for gas leaks using a Matheson 8017 gas leak detector.

VI-E. ADVANTAGES OF THE NEW APPARATUS

This new apparatus, which is suitable for VLE measurements at cryogenic conditions, offers the following advantages:

1. It can be operated at relatively higher pressure. The maximum working pressure is 150 atm.
2. The dead volume in this apparatus is minimal being that of the two needle valves (Valves A and B). Thus, the deviation in VLE measurements is expected to be reduced significantly.

3. The mechanical problems have been minimized because no pump or other complicated mechanical mixing device has been installed inside the cryostat.

4. It is more economical since the inside volume of this equilibrium cell is smaller than those cells used previously in recirculating cell experiments. Hence, the quantity of gases needed for the operation is reduced in each experimental run.

CHAPTER VII

EXPERIMENTAL RESULTS AND DISCUSSION

In this study, vapor-liquid equilibrium data were obtained experimentally on three binary systems, nitrogen-carbon dioxide, nitrogen-methane and methane-ethylene. The results obtained from the experimental part of this study are presented in this chapter.

VII-A. NITROGEN-CARBON DIOXIDE SYSTEM

The capability of the new apparatus was first tested on the binary system nitrogen-carbon dioxide at 273.15 K. This system was selected because the data could be easily measured and because the two gases are not flammable. Comparisons of the experimental data for this system and those obtained from Canjar (8), Kaminishi (40), Muirbrook et al (53), Yorizane (104), and Zenner (107), are shown in Figure VII-1. The experimental data and the calculated values from the ICL equation are presented in Table VII-1. The excellent agreement shown in Figure VII-1 assures that the newly designed apparatus is capable of producing reliable results for the measurement of vapor-liquid equilibrium.

VII-B. METHANE-ETHYLENE SYSTEM

VLE values for the binary system methane-ethylene were measured at 223.15 K and 243.15 K. The VLE properties experimentally obtained for the system methane-ethylene at 243.15 K are reported in Table VII-2.

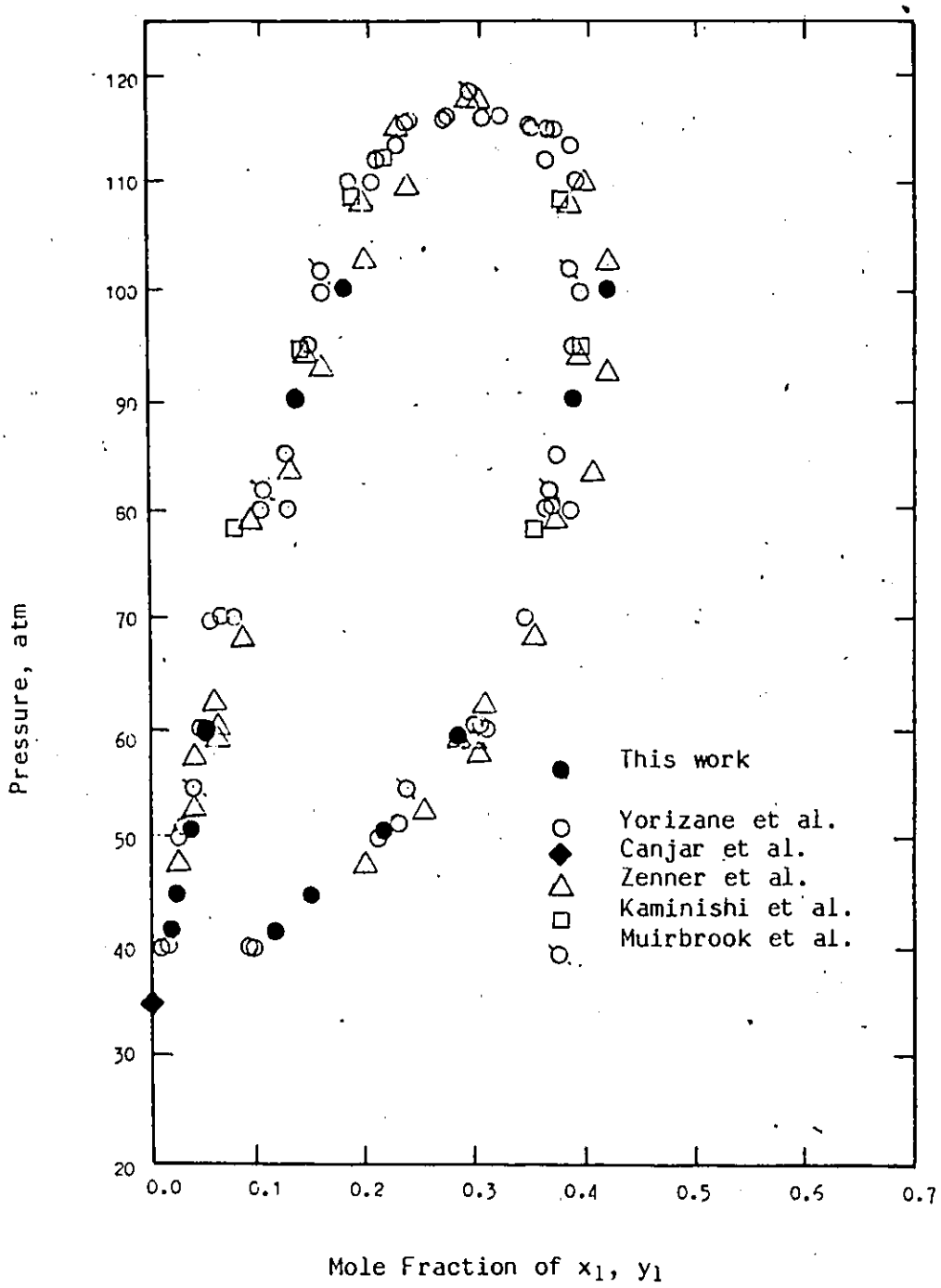


FIGURE VII-1: COMPARISON OF EXPERIMENTAL DATA WITH LITERATURE VALUES FOR THE $N_2(1)$-$CO_2(2)$ SYSTEM AT 273.15K

Table VII-1 Comparison of Experimental and Calculated VLE Compositions for the Binary System $N_2(1)$-$CO_2(2)$ at 273.15 K (ICL equation)

Pressure	x_1		y_1		Δx_1	Δy_1
atm.	Expl.	Calc.	Expl.	Calc.		
			$C_{12} = 0.04814$			
			$\lvert\Delta x\rvert_{av} = 0.0050$	$\lvert\Delta y\rvert_{av} = 0.0260$		
42.52	0.0225	0.0196	0.1189	0.1409	-0.0029	0.0220
45.10	0.0244	0.0258	0.1489	0.1744	0.0014	0.0255
50.82	0.0440	0.0396	0.2175	0.2362	-0.0044	0.0187
59.39	0.0584	0.0608	0.2780	0.3058	0.0024	0.0278
90.61	0.1379	0.1442	0.3965	0.4340	0.0063	0.0375
100.54	0.1810	0.1738	0.4266	0.4512	-0.0072	0.0246

Table VII-2 Comparison of Experimental and Calculated VLE Compositions for the Binary System $CH_4(1)$-$C_2H_4(2)$ at 243.15 K (ICL equation)

Pressure	x_1		y_1		Δx_1	Δy_1
atm.	Expl.	Calc.	Expl.	Calc.		
			C_{12} = 0.04588			
		$\lvert\Delta x\rvert_{av}$ = 0.0025		$\lvert\Delta y\rvert_{av}$ = 0.0026		
22.93	0.0431	0.0385	0.1478	0.1417	-0.0046	-0.0061
31.53	0.1289	0.1293	0.3460	0.3439	0.0004	-0.0021
39.87	0.2169	0.2205	0.4544	0.4560	0.0036	0.0016
47.90	0.3151	0.3123	0.5256	0.5234	-0.0028	-0.0022
54.84	0.4005	0.3973	0.5605	0.5597	-0.0032	-0.0008
56.88	0.4226	0.4242	0.5634	0.5664	0.0016	0.0030

The vapor and the liquid compositions calculated by using the ICL equation from fixed system temperature and pressure are also reported in the table. In addition, the binary interaction coefficient C_{12} and the average absolute deviations of the vapor and the liquid compositions obtained from the ICL equation at that isotherm are also presented in the table. The average absolute deviations obtained for $|\Delta x|$ and $|\Delta y|$ for the system are 0.0025 and 0.0026, respectively. The agreement obtained is considered as very satisfactory.

The critical pressure/composition point of the binary system methane-ethylene at 243.15 K was determined by extrapolation of the x-y data to the point of zero value of $(y_1-x_1)^2$. This is illustrated in Figure VII-2. The numerical values used in the preparation of this plot are reported in Table VII-3, and essentially the critical composition/pressure point of a binary system at a given temperature could be determined by the law of rectilinear diameters (52), when the pressure is plotted versus $(y_1+x_1)/2$. Thus, the critical pressure and composition point of this binary system at 243.15 K was determined to be P_c = 61.2 atm and x_c = 0.5075, respectively. This is shown in Figure VII-2.

To demonstrate the good agreement between the experimentally measured and the calculated values obtained from the ICL equation, a pressure-composition diagram for the binary system methane-ethylene at 243.15 K was plotted and shown in Figure VII-3.

VLE values for the binary system methane-ethylene at 223.15 K were also measured experimentally. The measured values for the system are reported in Table VII-4. A comparison of the experimental and the

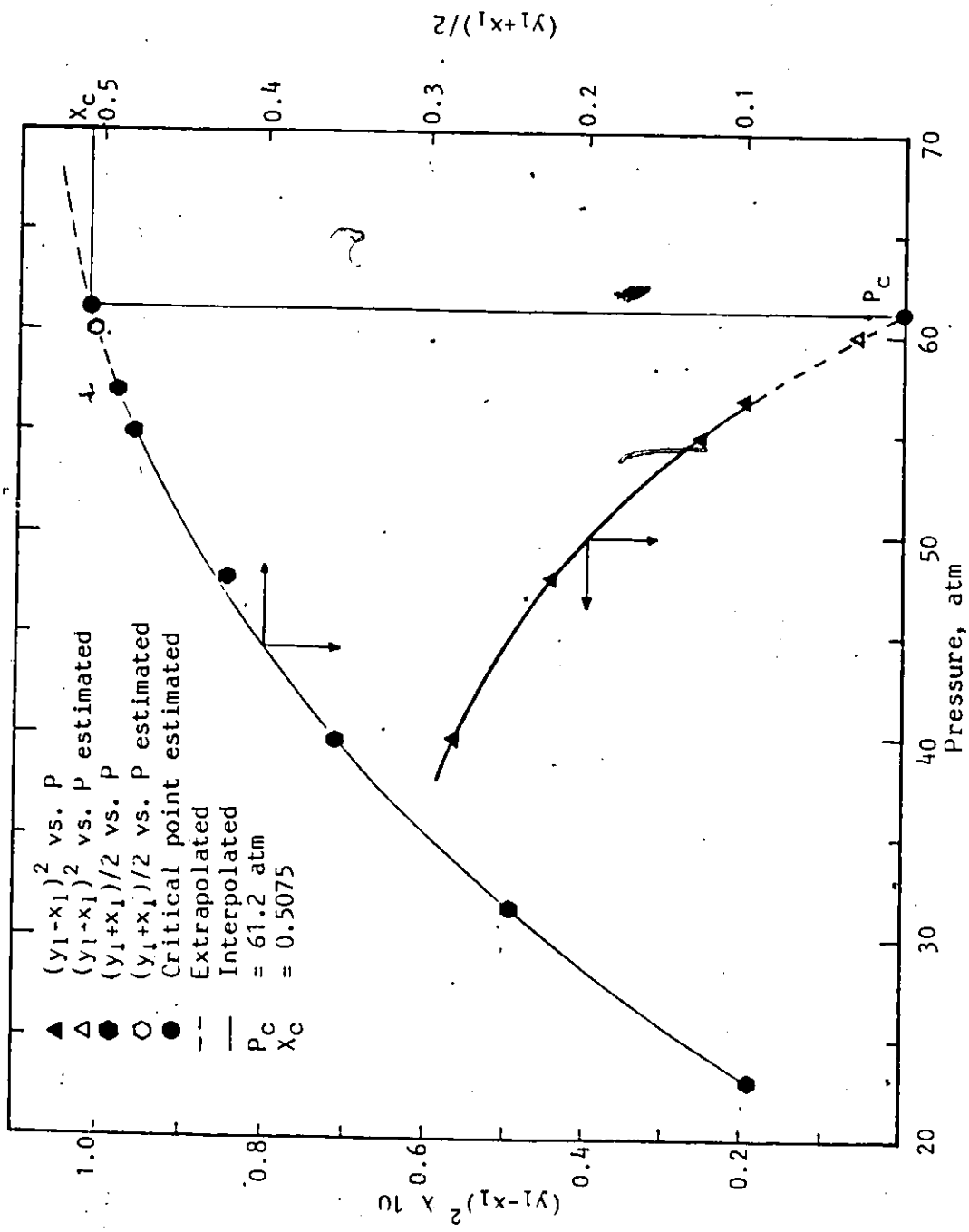


FIGURE VII-2: ESTIMATION OF CRITICAL PRESSURE AND CRITICAL COMPOSITION FOR THE BINARY SYSTEM $CH_4(1)$-$C_2H_4(2)$ AT 243.15 K

Table VII-3 Properties Used for Determining of Critical Pressure and Critical Compositions for the Binary System $CH_4(1)$-$C_2H_4(2)$ at 243.15 K

No.	P, atm	$(y_1-x_1)^2$	$(y_1+x_1)/2$	P_c, atm	$y_c = x_c$
1	22.93	0.01096	0.09545		
2	31.53	0.0471	0.23745		
3	39.87	0.0564	0.33565		
4	47.90	0.0443	0.42035		
5	54.84	0.0256	0.4805		
6	56.88	0.01982	0.4930		
critical point obtained by graphical solution				61.2	0.51

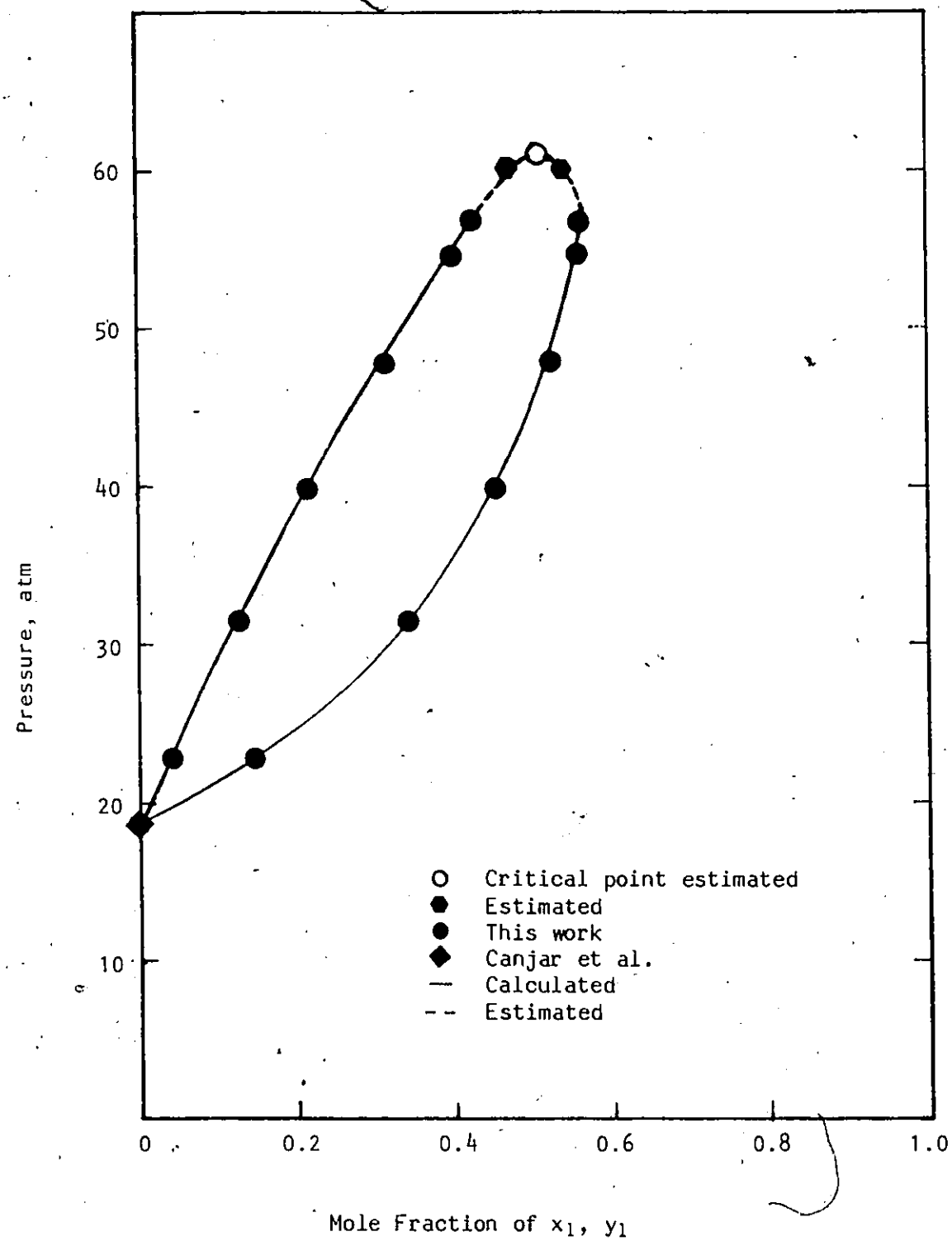


FIGURE VII-3: PRESSURE-COMPOSITION DIAGRAM FOR THE $CH_4(1)$-$C_2H_4(2)$ SYSTEM AT 243.15 K

Table VII-4 Comparison of Experimental and Calculated VLE Compositions for the Binary System $CH_4(1)$-$C_2H_4(2)$ at 243.15 K (ICL Equation)

Pressure	x_1		y_1		Δx_1	Δy_1
atm	Expl.	Calc.	Expl.	Calc.		
			$C_{12} = 0.03687$			
			$\lvert\Delta x\rvert_{av} = 0.0075$	$\lvert\Delta y\rvert_{av} = 0.0042$		
19.46	0.1197	0.1202	0.4312	0.4336	0.0005	0.0024
22.80	0.1708	0.1662	0.5147	0.5103	-0.0046	-0.0044
29.26	0.2658	0.2569	0.6120	0.6094	-0.0089	-0.0026
38.17	0.3939	0.3853	0.7042	0.6906	-0.0086	-0.0136
52.94	0.6060	0.6079	0.7610	0.7587	0.0019	-0.0023
44.91	0.4729	0.4851	0.7294	0.7295	0.0122	0.0

calculated results from the ICL equations for the system was also made. The results of the comparison are reported in Table VII-4. A pressure versus composition plot is shown in Figure VII-4. Data reported by Canjar et al. (8) on pure ethylene and on the mixture reported by Sagara et al. (72) are also included in this figure for comparison. The critical pressure of 57.2 atm and critical composition of 0.71 mole fraction of methane were determined by the same method described earlier. A pressure versus $(y_1-x_1)^2$ plot was used to extrapolate the critical pressure and a pressure versus $(y_1+x_1)/2$ plot was used to obtain the critical composition for the binary system at 223.15 K as shown in Figure VII-5. The values used for construction of Figure VII-5 are reported in Table VII-5. It is apparent that the calculated values by using the ICL equation for this binary system at 223.15 K are in good agreement with those experimentally determined in this study. In addition, the data reported by Sagara et al (72) are reasonably close to the measured data and the calculated values from the ICL equation as indicated in Figure VII-4.

VII-C. NITROGEN-METHANE SYSTEM

Vapor-liquid equlibria for the binary system nitrogen-methane were measured at 169.9 K. The predicated values using the ICL equation were compared with those measured data. Both the measured and the calculated VLE compositions for this system are reported in Table VII-6. The data obtained by Wang (94) and by Kidnay et al. (42) were also included in the comparison whereby values of y calculated with the ICL equation were evaluated. The absolute deviations of calculated versus experimental

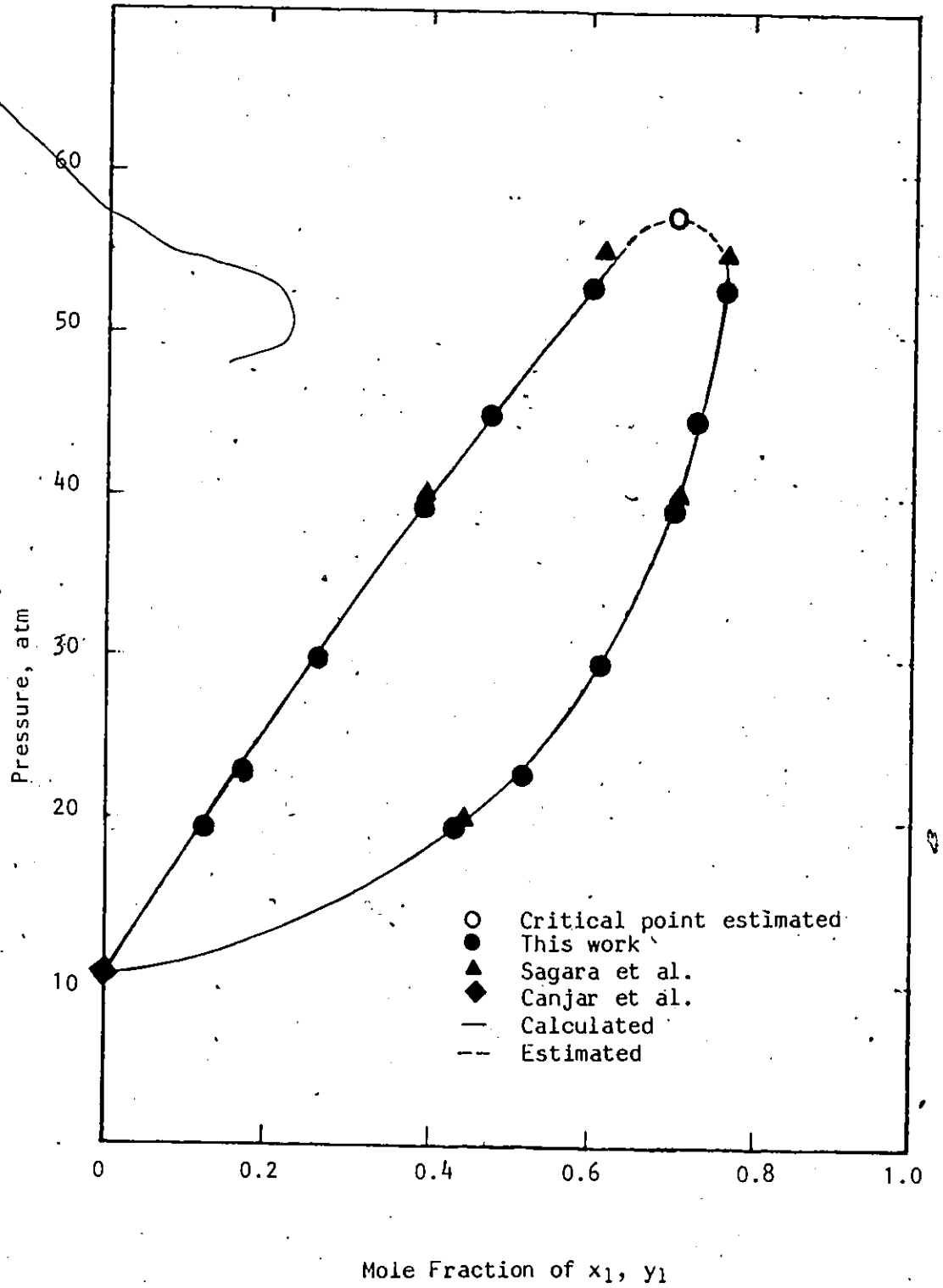


FIGURE VII-4: COMPARISON OF EXPERIMENTAL AND CALCULATED VALUES FOR THE BINARY SYSTEM $CH_4(1)$-$C_2H_4(2)$ AT 223.15 K

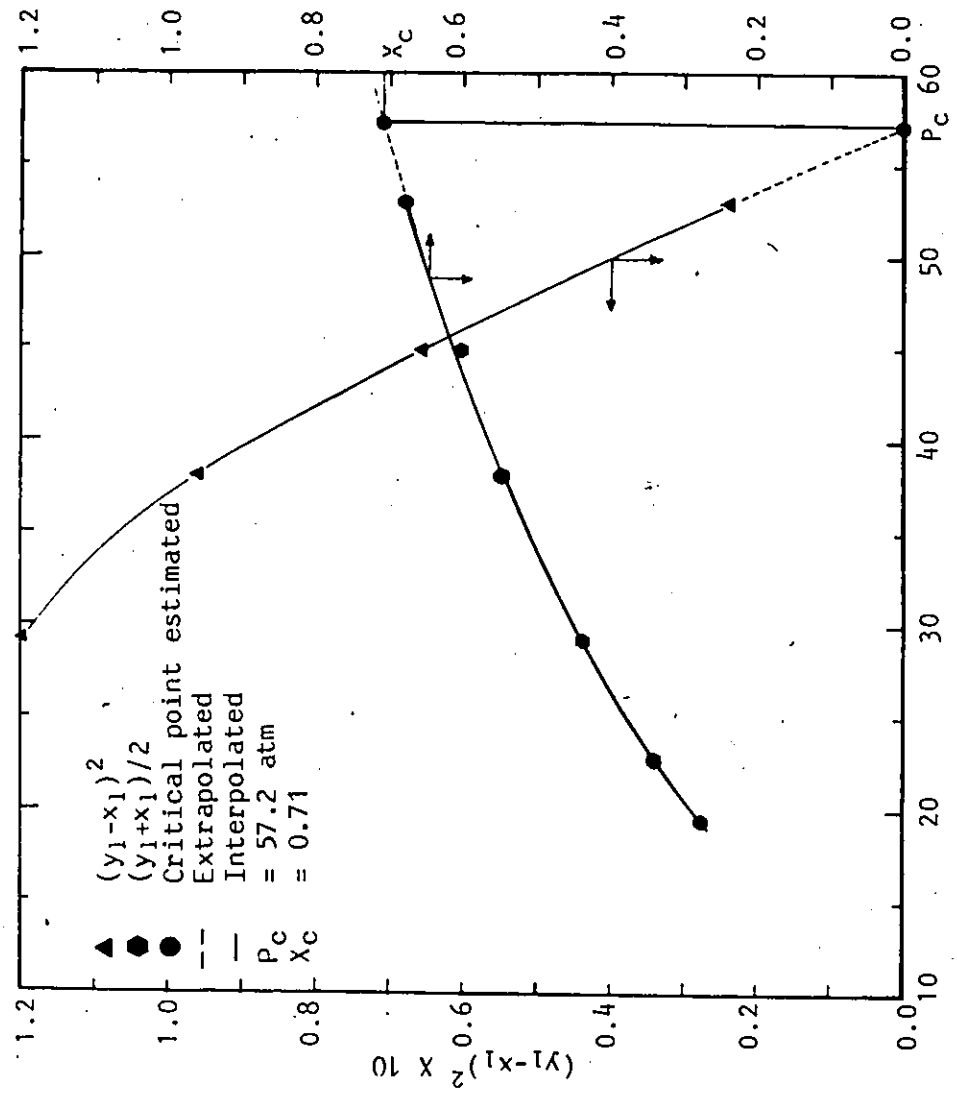


FIGURE VII-5: ESTIMATION OF CRITICAL PRESSURE AND CRITICAL COMPOSITION FOR THE BINARY SYSTEM $CH_4(1)$-$C_2H_4(2)$ AT 223.15 K

Table VII-5 Properties Used for Determining of Critical Pressure and Critical Compositions for the Binary System $CH_4(1)$-$C_2H_4(2)$ at 223.15 K

No.	P, atm	$(y_1-x_1)^2$	$(y_1+x_1)/2$	P_c, atm	$y_c=x_c$
1	19.46	0.0970	0.27545		
2	22.80	0.11827	0.34275		
3	29.26	0.11985	0.4389		
4	38.17	0.09629	0.54905		
5	44.91	0.06579	0.60115		
6	52.94	0.02402	0.6835		
				57.2	0.71

Table VII-6 Comparison of Experimental and Calculated VLE Compositions for the Binary System $N_2(1)$-$CH_4(2)$ (ICL Equation)

Pressure	x_1		y_1		Δx_1	Δy_1
atm	Expl.	Calc.	Expl.	Calc.		
Results Obtained at 169.9 K (C_{12} = 0.0495) in This Work						
	$\lvert\Delta x\rvert_{av}$ = 0.0057		$\lvert\Delta y\rvert_{av}$ = 0.0148			
22.85	0.0		0.0			
26.67	0.0499	0.0385	0.1152	0.1086	-0.0114	-0.0366
28.03	0.0601	0.0537	0.1640	0.1434	-0.0064	-0.0206
32.18	0.0948	0.1013	0.2513	0.2311	0.0065	-0.0202
39.26	0.1957	0.1873	0.3303	0.3341	-0.0084	0.0038
45.66	0.2715	0.2744	0.3890	0.3907	0.0029	0.0017
47.73	0.3040	0.3067	0.3961	0.4011	0.0027	0.0050
48.65	0.3167	0.3224	0.3872	0.4030	0.0057	0.0158
Data Reported by Wang (55) at 169.9 K (C_{12} = 0.05371)						
	$\lvert\Delta x\rvert_{av}$ = 0.0029		$\lvert\Delta y\rvert_{av}$ = 0.0148			
24.60	0.0231	0.0154	0.0770	0.0480	-0.0077	-0.0290
25.41	0.0301	0.0241	0.0993	0.0727	-0.0060	-0.0267
27.36	0.0579	0.0454	0.1485	0.1259	-0.0125	-0.0226
31.20	0.0888	0.0884	0.2298	0.2114	-0.0004	-0.0184
34.07	0.1183	0.1215	0.2752	0.2620	0.0032	-0.0132
38.09	0.1632	0.1700	0.3275	0.3185	0.0068	-0.0090
43.55	0.2352	0.2409	0.3790	0.3735	0.0057	-0.0055
45.66	0.2683	0.2708	0.3875	0.3885	0.0025	0.0010
46.74	0.2885	0.2871	0.4235	0.3944	-0.0014	-0.0291
47.76	0.3057	0.3041	0.3970	0.3994	-0.0016	0.0024
47.83	0.3032	0.3046	0.3962	0.3993	0.0014	0.0031
48.58	0.3159	0.3172	0.3836	0.4014	0.0013	0.0178
Data Reported by Kidnay et al (100) at 170 K (C_{12} = 0.07474)						
	$\lvert\Delta x\rvert_{av}$ = 0.0038		$\lvert\Delta y\rvert_{av}$ = 0.0064			
23.95	0.0082	0.0073	0.0291	0.0249	-0.0009	-0.0042
25.06	0.0189	0.0182	0.0652	0.0591	-0.0007	-0.0061
25.86	0.0276	0.0261	0.0889	0.0821	-0.0015	-0.0068
27.11	0.0399	0.0386	0.1219	0.1151	-0.0013	-0.0068
28.06	0.0495	0.0483	0.1458	0.1384	-0.0013	-0.0074
29.96	0.0688	0.0678	0.1875	0.1803	-0.0010	-0.0072
33.98	0.1115	0.1106	0.2593	0.2527	-0.0009	-0.0066
38.08	0.1562	0.1569	0.3135	0.3091	0.0007	-0.0044
41.96	0.2001	0.2037	0.3519	0.3493	0.0036	-0.0026
45.70	0.2469	0.2532	0.3743	0.3772	0.0063	0.0029
47.70	0.2729	0.2827	0.3784	0.3868	0.0098	0.0084
48.31	0.2915	0.2923	0.3798	0.3890	0.0008	0.0092
48.74	0.3066	0.2994	0.3801	0.3909	-0.0072	0.0108

vapor compositions and liquid compositions are also reported in the table. The binary interaction coefficient of the ICL equation, $C_{N_2-CH_4}$ was also calculated and included in the table. The comparison of experimental and calculated results is illustrated in Figure VII-6. The experimental data reported by Kidnay et al. and by Wang are also shown in this figure. As shown in Figure VII-6, the data reported for nitrogen-methane system at 170.0 K by Kidnay showed best agreement with calculated values by the ICL equation at 169.9 K among three measurements at pressure under 30 atm. However, disagreements grow gradually at pressure above 30 atm. At pressure above 35 atm, the agreement among calculated values and those reported by Wang and this work for the binary system became satisfactory with the exception of the point, y_1 = 0.4235 at P = 46.74 atm, reported by Wang.

Possible critical point for this binary system at 169.9 K was estimated by applying the rectilinear rule as mentioned earlier. The properties used in this estimation procedure are listed in Table VII-7. The critical pressure/composition point of the nitrogen-methane system at 169.9 K was graphically established as illustrated in Figure VII-7. That estimated critical point is also shown in Figure VII-6.

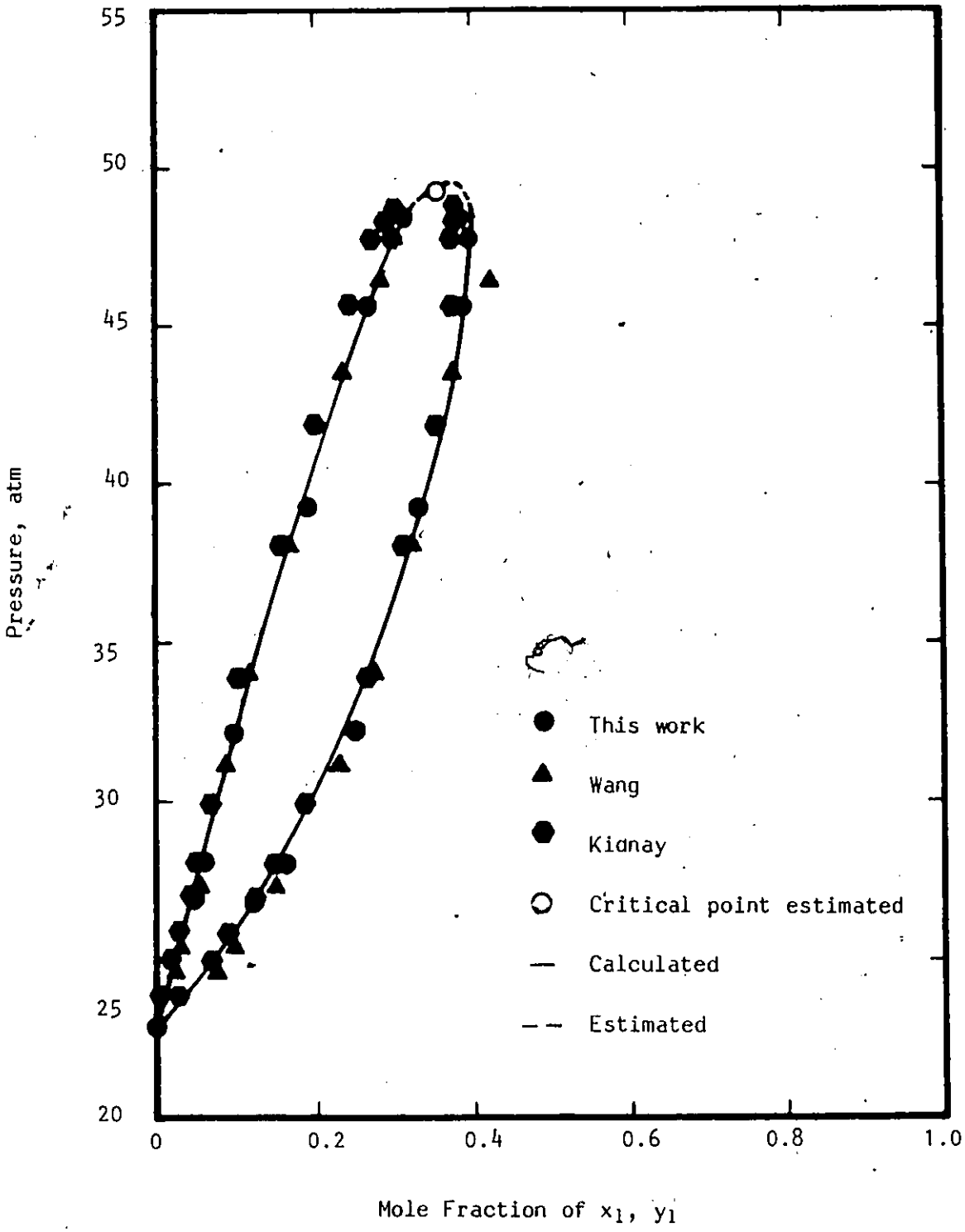


FIGURE VII-6: COMPARISON OF EXPERIMENTAL AND CALCULATED VALUES AT 169.9 K FOR THE BINARY SYSTEM $N_2(1)$-$CH_4(2)$ TOGETHER WITH THOSE REPORTED BY WANG (94) AT 169.9 K AND BY KIDNAY ET AL. (42) AT 170.0 K

Table VII-7 Properties Used for Determining of Critical Pressure and Critical Compositions for the Binary System $N_2(1)$-$CH_4(2)$ at 169.9 K

No.	P, atm	$(y_1-x_1)^2$	$(y_1+x_1)/2$	P_c, atm	$y_c = x_c$
1	26.67	0.0091	0.0976		
2	28.03	0.0108	0.1121		
3	32.18	0.0245	0.1731		
4	39.26	0.0155	0.2680		
5	45.66	0.0138	0.3303		
6	47.73	0.0085	0.3501		
7	48.65	0.0050	0.3520		
critical point obtained by graphical solution				49.25	0.36

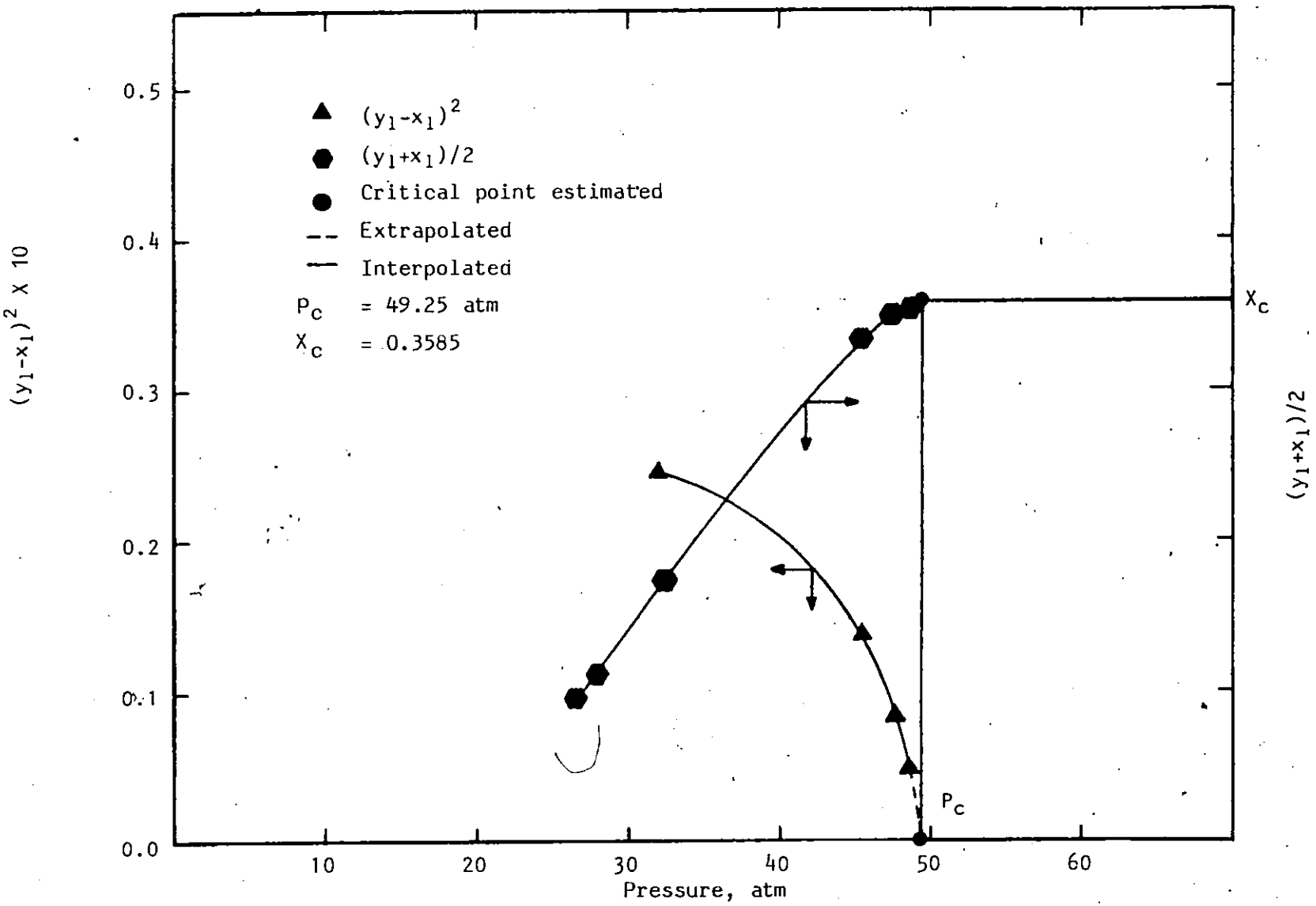


FIGURE VII-7: ESTIMATION OF CRITICAL PRESSURE AND CRITICAL COMPOSITION FOR THE BINARY SYSTEM $N_2(1)$-$CH_4(2)$ AT 169.9 K

CHAPTER VIII

SUMMARY

A new apparatus suitable for VLE measurements at cryogenic conditions was designed, constructed and used to measure data on binary systems of nitrogen-carbon dioxide, nitrogen-methane and methane-ethylene. System conditions, temperature, pressure, and phase compositions of vapor and liquid at equlibrium, were determined with this apparatus. The data have shown that the newly designed equipment provided with four major advantages: high working pressure, reduced dead volume, simple mechanical arrangement and economical operation.

The VLE properties of binary system nitrogen-carbon dioxide at 273.15 K were measured with this set up and the data were compared with those available in the literature (40,52,100,107). Good agreement between the new set and the available data was observed.

Also measured were data on the binary system methane-ethylene at 223.15 K and 243.15 K. The new experimental data obtained at these two temperatures were compared with values calculated by the ICL (36-38) correlation. The agreement between experimental and calculated results on this system at each of the temperatures was excellent, thus the validating the usefulness of the ICL correlation for describing the system and (indirectly) highlighting the smoothness of the data. In addition, both experimental and calculated results for the methane-ethylene system

at 243.15 K were compared with those reported by Sagara (72) and by Canjar et al. (8). Good agreement was also found in this comparison.

The VLE behavior of the binary system nitrogen-methane were experimentally determined at 169.9 K. The experimental results of the binary system were compared with those calculated from the ICL equation. The agreement was found satisfactory. In addition, both experimental and calculated results for the system were compared with those reported by Wang (94) and by Kidnay et al (42). One data point, $y = 0.4235$ at $P = 46.74$ atm, reported by Wang was found to deviate from the rest of data.

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APPENDICES

APPENDIX A

EXPERIMENTAL AND CALCULATED VLE VALUES FOR BINARY SYSTEMS

The ICL equation was used to calculate vapor and liquid compositions from fixed values of system temperature and pressure. In the calculation, the optimal value of C_{ij} for a given system and temperature was obtained by minimizing the difference between the calculated and experimental values of liquid composition.

APPENDIX A

Table A-1a

Comparison of Experimental and Calculated VLE Values for the Binary System $H_2(1)$-$N_2(2)$

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
			70.35 K (C_{12} = -0.0112)				
14.31	0.0320	0.0331	0.9610	0.9573	0.0011	-0.0037	11
34.76	0.0680	0.0781	0.9670	0.9685	0.0101	0.0015	11
58.52	0.1050	0.1256	0.9620	0.9650	0.0206	0.0030	11
81.61	0.1380	0.1677	0.9540	0.9571	0.0297	0.0031	11
119.70	0.1970	0.2311	0.9360	0.9398	0.0341	0.0038	11
153.06	0.2420	0.2826	0.8970	0.9219	0.0406	0.0249	11
191.15	0.2900	0.3388	0.8740	0.8988	0.0488	0.0248	11
243.55	0.3830	0.4151	0.8080	0.8609	0.0321	0.0529	11
260.62	0.4140	0.4405	0.7830	0.8469	0.0265	0.0639	11
270.78	0.4470	0.4560	0.7710	0.8406	0.0090	0.0696	11
283.02	0.4870	0.4751	0.7250	0.8321	-0.0119	0.1071	11
294.07	0.5360	0.5307	0.6900	0.8401	-0.0053	0.1501	11
			77.55 K (C_{12} = -0.0237)				
10.16	0.0260	0.0227	0.8860	0.8730	-0.0033	-0.0130	11
20.43	0.0460	0.0476	0.9140	0.9194	0.0016	0.0054	11
33.95	0.0640	0.0792	0.9310	0.9338	0.0152	0.0028	11
44.21	0.0860	0.1025	0.9280	0.9362	0.0165	0.0082	11
69.37	0.1400	0.1574	0.9210	0.9313	0.0174	0.0103	11
85.06	0.1730	0.1902	0.9190	0.9245	0.0173	0.0055	11
103.42	0.2140	0.2277	0.9180	0.9143	0.0137	-0.0037	11
136.08	0.2860	0.2930	0.8730	0.8916	0.0070	0.0186	11
143.58	0.3060	0.3079	0.8620	0.8856	0.0019	0.0236	11
			77.55 K (C_{12} = -0.0237)				
153.75	0.3350	0.3281	0.8400	0.8773	-0.0069	0.0373	11
168.75	0.3730	0.3583	0.8200	0.8640	-0.0147	0.0440	11
183.75	0.4330	0.3891	0.7620	0.8485	-0.0439	0.0865	11

Table A-1a (continued)

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
			83.15 K (C_{12} = -0.0408)				
21.43	0.0487	0.0511	0.8655	0.8652	0.0024	-0.0003	12
34.02	0.0763	0.0832	0.8948	0.8905	0.0069	-0.0043	12
95.26	0.2300	0.2388	0.8700	0.8808	0.0038	0.0108	12
136.08	0.3446	0.3350	0.7977	0.8435	-0.0096	0.0458	12
			83.67 K (C_{12} = -0.0609)				
111.21	0.2730	0.2979	0.8260	0.8568	0.0249	0.0308	11
125.52	0.3250	0.3385	0.7980	0.8403	0.0135	0.0423	11
136.77	0.3600	0.3718	0.7650	0.8247	0.0118	0.0597	11
141.51	0.3840	0.3864	0.7470	0.8180	0.0024	0.0710	11
151.08	0.4290	0.4171	0.7110	0.8037	-0.0120	0.0927	11
			88.15 K (C_{12} = -0.0631)				
16.50	0.0490	0.0383	0.7650	0.7563	-0.0107	-0.0087	13
31.00	0.0760	0.0791	0.8470	0.8299	0.0031	-0.0171	13
50.00	0.1340	0.1324	0.8660	0.8515	-0.0017	-0.0145	13
77.40	0.2100	0.2097	0.8530	0.8466	-0.0003	-0.0064	13
93.40	0.2350	0.2559	0.8420	0.8348	0.0209	-0.0072	13
102.60	0.2710	0.2832	0.8080	0.8256	0.0122	0.0176	13
119.10	0.3540	0.3343	0.7810	0.8058	-0.0197	0.0248	13
131.10	0.3820	0.3741	0.7490	0.7873	-0.0079	0.0383	13
			90.03 K (C_{12} = -0.0272)				
13.39	0.0248	0.0253	0.6732	0.6726	0.0005	-0.0006	48
12.81	0.0234	0.0238	0.6619	0.6614	0.0004	-0.0005	48
11.70	0.0207	0.0210	0.6367	0.6369	0.0003	0.0002	48
10.07	0.0164	0.0168	0.5903	0.5914	0.0004	0.0011	48
8.32	0.0119	0.0123	0.5217	0.5224	0.0004	0.0007	48
9.93	0.0159	0.0164	0.5855	0.5867	0.0005	0.0012	48
24.10	0.0534	0.0526	0.7800	0.7797	-0.0008	-0.0003	48
45.33	0.1115	0.1061	0.8304	0.8319	-0.0054	0.0015	48

Table A-1a (continued)

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
			90.79 K (C_{12} = -0.0586)				
18.26	0.0340	0.0408	0.6850	0.7207	0.0068	0.0358	11
24.47	0.0470	0.0583	0.7480	0.7672	0.0113	0.0192	11
34.05	0.0720	0.0852	0.7940	0.8021	0.0132	0.0081	11
44.90	0.0990	0.1157	0.8160	0.8194	0.1670	0.0034	11
59.90	0.1440	0.1581	0.8230	0.8261	0.1412	0.0031	11
72.14	0.1780	0.1932	0.8220	0.8238	0.0152	0.0018	11
90.49	0.2370	0.2470	0.8030	0.8121	0.0100	0.0091	11
104.11	0.2890	0.2886	0.7790	0.7984	-0.0004	0.0194	11
117.04	0.3490	0.3302	0.7460	0.7819	-0.0188	0.0359	11
125.92	0.4070	0.3606	0.6980	0.7664	-0.0464	0.0684	11
			95.00 K (C_{12} = -0.0485)				
10.95	0.0149	0.0158	0.4492	0.4495	0.0009	0.0003	48
25.87	0.0566	0.0577	0.7005	0.6982	0.0011	-0.0023	48
44.73	0.1121	0.1110	0.7643	0.7638	-0.0011	-0.0005	48
			99.82 K (C_{12} = -0.0635)				
21.43	0.0377	0.0420	0.5509	0.5409	0.0043	-0.0100	12
34.02	0.0741	0.0806	0.6686	0.6471	0.0065	-0.0215	12
54.43	0.1384	0.1453	0.7279	0.6998	0.0069	-0.0282	12
74.85	0.2116	0.2140	0.7175	0.7042	0.0024	-0.0133	12
95.26	0.3030	0.2903	0.6530	0.6853	-0.0127	0.0323	12
			100.00 K (C_{12} = -0.0641)				
14.31	0.0180	0.0201	0.3380	0.3850	0.0021	0.0470	11
20.43	0.0310	0.0387	0.4900	0.5211	0.0077	0.0311	11
29.21	0.0560	0.0657	0.5870	0.6145	0.0097	0.0275	11
40.85	0.0900	0.1020	0.6560	0.6701	0.0120	0.0141	11
52.70	0.1280	0.1399	0.6820	0.6947	0.0119	0.0127	11
62.56	0.1580	~~0.1724~~	0.6880	0.7021	0.0144	0.0141	11
73.81	0.2070	0.2110	0.6890	0.7016	0.0040	0.0126	11
84.37	0.2480	0.2492	0.6740	0.6943	0.0012	0.0203	11
93.25	0.3010	0.2835	0.6450	0.6846	-0.0175	0.0396	11
102.04	0.3610	0.3203	0.6060	0.6679	-0.0407	0.0619	11

Table A-1a (continued)

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
			110.30 K (C_{12} = -0.0738)				
23.49	0.0290	0.0311	0.2530	0.2637	0.0021	0.0107	11
31.28	0.0550	0.0594	0.3610	0.3738	0.0044	0.0128	11
38.49	0.0800	0.0864	0.4260	0.4325	0.0064	0.0065	11
46.28	0.1070	0.1169	0.4630	0.4707	0.0099	0.0077	11
50.03	0.1250	0.1322	0.4770	0.4822	0.0072	0.0052	11
56.45	0.1530	0.1594	0.4890	0.4956	0.0064	0.0066	11
60.89	0.1720	0.1792	0.4910	0.4998	0.0072	0.0088	11
65.33	0.2020	0.2000	0.4860	0.5034	-0.0020	0.0174	11
74.50	0.2600	0.2481	0.4560	0.4989	-0.0119	0.0429	11
76.87	0.2860	0.2622	0.4210	0.4950	-0.0238	0.0740	11

Table A-1b

Comparison of Experimental and Calculated VLE Values for the Binary System $H_2(1)$-CO(2)

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
			77.35 K (C_{12} = -0.0101)				
46.00	0.0750	0.0820	0.9470	0.9601	0.0070	0.0132	14
51.00	0.0830	0.0903	0.9600	0.9599	0.0073	-0.0001	14
75.00	0.1130	0.1279	0.9440	0.9557	0.0149	0.0177	14
100.00	0.1600	0.1643	0.9410	0.9483	0.0043	0.0073	14
125.00	0.2000	0.1985	0.9290	0.9394	-0.0015	0.0104	14
150.00	0.2500	0.2310	0.9090	0.9295	-0.0190	0.0205	14
			83.15 K (C_{12} = -0.0248)				
21.43	0.0398	0.0412	0.9084	0.9115	0.0014	0.0031	12
34.02	0.0609	0.0657	0.9320	0.9279	0.0048	-0.0041	12
95.26	0.1745	0.1756	0.9143	0.9233	0.0011	0.0090	12
136.08	0.2490	0.2428	0.8790	0.9042	-0.0062	0.0252	12
			99.82 K (C_{12} = -0.0625)				
21.43	0.0389	0.0402	0.6803	0.6638	0.0013	-0.0165	12
34.02	0.0674	0.0716	0.7619	0.7426	0.0042	-0.0193	12
54.43	0.1163	0.1229	0.7844	0.7827	0.0066	-0.0017	12
74.85	0.1727	0.1749	0.7848	0.7891	0.0022	0.0043	12
95.26	0.2399	0.2285	0.7739	0.7811	-0.0114	0.0072	12
			103.15 K (C_{12} = -0.0850)				
50.00	0.1360	0.1186	0.7100	0.7281	-0.0174	0.0181	14
75.00	0.1920	0.1905	0.7260	0.7423	-0.0015	0.0164	14
100.00	0.2670	0.2680	0.6730	0.7268	0.0010	0.0538	14
			122.04 K (C_{12} = -0.0986)				
34.02	0.0449	0.0504	0.2260	0.2402	0.0055	0.0142	12
54.43	0.1382	0.1382	0.3349	0.3592	-0.0000	0.0243	12
			123.15 K (C_{12} = -0.1499)				
62.00	0.2000	0.2024	0.2910	0.3298	0.0024	0.0388	14
62.70	0.2470	0.2763	0.2930	0.3330	0.0293	0.0400	14

Table A-1c

Comparison of Experimental and Calculated VLE Values for the Binary System $H_2(1)$-$CH_4(2)$

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
		90.72 K (C_{12} = -0.0679)					
60.27	0.0394	0.0397	0.9910	0.9923	0.0003	0.0013	50
40.20	0.0275	0.0275	0.9919	0.9923	0.0	0.0004	50
20.05	0.0143	0.0142	0.9878	0.9900	-0.0001	0.0022	50
125.20	0.0748	0.0746	0.9888	0.9896	-0.0002	0.0008	50
100.00	0.0612	0.0618	0.9902	0.9909	0.0006	0.0007	50
79.93	0.0479	0.0510	0.9918	0.9917	0.0031	-0.0001	50
50.20	0.0336	0.0337	0.9926	0.9924	0.0001	-0.0002	50
30.12	0.0212	0.0210	0.9917	0.9917	-0.0002	0.0	50
10.12	0.0076	0.0073	0.9851	0.9843	-0.0003	-0.0008	50
		93.15 K (C_{12} = -0.0280)					
100.00	0.0460	0.0537	0.9865	0.9899	0.0077	0.0034	51
125.00	0.0801	0.0647	0.9810	0.9889	-0.0154	0.0079	51
150.00	0.0700	0.0749	0.9766	0.9877	0.0049	0.0111	51
		103.07 K (C_{12} = -0.0925)					
100.27	0.0789	0.0774	0.9757	0.9771	-0.0015	0.0014	50
80.13	0.0634	0.0632	0.9787	0.9780	-0.0002	-0.0007	50
60.00	0.0480	0.0484	0.9776	0.9779	0.0004	0.0003	50
30.03	0.0252	0.0250	0.9734	0.9726	-0.0002	-0.0008	50
15.10	0.0126	0.0126	0.9596	0.9580	0.0	-0.0016	50
		103.15 K (C_{12} = -0.0922)					
10.00	0.0040	0.0083	0.9300	0.9417	0.0043	0.0117	51
15.00	0.0100	0.0125	0.9530	0.9574	0.0025	0.0044	51
20.00	0.0130	0.0167	0.9620	0.9651	0.0037	0.0031	51
30.00	0.0300	0.0250	0.9680	0.9724	-0.0050	0.0044	51
50.00	0.0500	0.0408	0.9630	0.9771	-0.0092	0.0141	51
		103.15 K (C_{12} = -0.1266)					
19.70	0.0142	0.0185	0.9530	0.9644	0.0043	0.0114	15

Table A-1c (continued)

Pressure atm (psia)	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1	Ref
		103.15 K (C_{12} = -0.1266)					
40.80	0.0337	0.0378	0.9670	0.9751	0.0041	0.0081	15
60.40	0.0569	0.0549	0.9690	0.9770	-0.0020	0.0080	15
80.90	0.0708	0.0720	0.9700	0.9768	0.0012	0.0068	15
90.00	0.0819	0.0794	0.9690	0.9763	-0.0025	0.0073	15
		109.99 K (C_{12} = -0.1095)					
124.80	0.1080	0.1055	0.9588	0.9623	-0.0025	0.0035	50
100.14	0.0880	0.0865	0.9622	0.9640	-0.0015	0.0017	50
80.07	0.0711	0.0704	0.9634	0.9644	-0.0007	0.0010	50
60.14	0.0532	0.0537	0.9633	0.9633	0.0005	-0.0001	50
45.34	0.0407	0.0409	0.9605	0.9602	0.0002	-0.0003	50
30.08	0.0275	0.0273	0.9508	0.9520	-0.0002	0.0012	50
14.97	0.0134	0.0134	0.9247	0.9235	0.0	-0.0011	50
		116.48 K (C_{12} = -0.1656)					
33.34	0.0318	0.0363	0.9279	0.9265	0.0045	-0.0014	16
101.38	0.0950	0.1084	0.9466	0.9441	0.0135	-0.0025	16
156.49	0.1394	0.1627	0.9383	0.9376	0.0233	-0.0007	16
272.16	0.3170	0.2686	0.9289	0.9129	-0.0484	-0.0160	16
		116.51 K (C_{12} = -0.1209)					
124.67	0.1188	0.1150	0.9424	0.9458	-0.0038	0.0034	50
100.00	0.0967	0.0938	0.9467	0.9471	-0.0029	0.0004	50
79.90	0.0771	0.0759	0.9466	0.9467	-0.0012	0.0	50
40.14	0.0382	0.0388	0.9364	0.9343	0.0006	-0.0022	50
40.14	0.0383	0.0388	0.9360	0.9343	0.0005	-0.0018	50
20.17	0.0192	0.0191	0.9024	0.9000	-0.0001	-0.0024	50
		123.15 K (C_{12} = -0.1272)					
10.10	0.0073	0.0085	0.6830	0.7287	0.0012	0.0457	15
20.00	0.0192	0.0192	0.8180	0.8438	0.0	0.0258	15
39.80	0.0385	0.0401	0.8860	0.9000	0.0016	0.0140	15
59.80	0.0616	0.0606	0.9040	0.9164	-0.0010	0.0124	15
80.70	0.0816	0.0814	0.9110	0.9227	-0.0002	0.0117	15

Table A-1c (continued)

Pressure atm (psia)	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1	Ref
		123.15 K (C_{12} = -0.1272)					
100.50	0.1010	0.1005	0.9080	0.9245	-0.0005	0.0165	15
		123.15 K (C_{12} = -0.1486)					
20.00	0.0200	0.0202	0.8860	0.8432	0.0002	-0.0428	51
30.00	0.0380	0.0315	0.9310	0.8813	-0.0065	-0.0497	51
40.00	0.0380	0.0425	0.9090	0.8994	0.0045	-0.0096	51
50.00	0.0560	0.0535	0.9020	0.9094	-0.0025	0.0074	51
		143.05 K (C_{12} = -0.1653)					
10.60	0.0031	0.0043	0.2140	0.2484	0.0012	0.0344	15
19.90	0.0168	0.0174	0.5410	0.5461	0.0006	0.0051	15
40.20	0.0477	0.0455	0.7210	0.7213	-0.0022	0.0003	15
59.10	0.0684	0.0713	0.7790	0.7730	0.0029	-0.0060	15
78.70	0.0992	0.0978	0.8020	0.7968	-0.0014	-0.0052	15
100.20	0.1270	0.1265	0.8100	0.8084	-0.0005	-0.0016	15
		143.15 K (C_{12} = -0.0873)					
10.00	0.0030	0.0030	0.1520	0.2080	0.0	0.0560	51
20.00	0.0080	0.0152	0.5100	0.5476	0.0072	0.0376	51
30.00	0.0350	0.0272	0.6820	0.6651	-0.0078	-0.0169	51
40.00	0.0550	0.0391	0.6930	0.7236	-0.0159	0.0306	51
50.00	0.0640	0.0508	0.7450	0.7579	-0.0132	0.0129	51
		144.26 K (C_{12} = -0.2174)					
34.02	0.0343	0.0404	0.6392	0.6713	0.0061	0.0321	16
68.04	0.0781	0.0927	0.7618	0.7701	0.0146	0.0083	16
101.38	0.1310	0.1438	0.7931	0.7919	0.0128	-0.0012	16
135.40	0.1805	0.1963	0.7823	0.7928	0.0158	0.0105	16
204.12	0.3499	0.3067	0.7457	0.7671	-0.0432	0.0214	16
		163.15 K (C_{12} = -0.2126)					
30.00	0.0110	0.0229	0.3410	0.2892	0.0119	-0.0518	51
40.00	0.0300	0.0422	0.4560	0.4045	0.0122	-0.0515	51
140.00	0.2860	0.2525	0.5760	0.5920	-0.0355	0.0160	51

Table A-1c (continued)

Pressure atm (psia)	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1	Ref
		163.15 K (C_{12} = -0.2126)					
150.00	0.2860	0.2778	0.5500	0.5901	-0.0082	0.0401	51
		172.04 K (C_{12} = -0.1119)					
34.02	0.0170	0.0182	0.1583	0.1668	0.0012	0.0085	16
68.04	0.0877	0.0874	0.4060	0.4135	-0.0003	0.0075	16
87.09	0.1255	0.1277	0.4631	0.4615	0.0022	-0.0016	16
102.06	0.1622	0.1606	0.4678	0.4827	-0.0016	0.0149	16
		172.05 K (C_{12} = -0.2491)					
86.40	0.1360	0.1532	0.4350	0.4395	0.0172	0.0045	15
102.20	0.2010	0.2004	0.4290	0.4523	-0.0006	0.0233	15
		173.65 K (C_{12} = -0.2132)					
35.20	0.0168	0.0203	0.1480	0.1521	0.0035	0.0041	15
40.10	0.0257	0.0318	0.2060	0.2100	0.0061	0.0040	15
59.90	0.0709	0.0794	0.3620	0.3473	0.0085	-0.0147	15
70.10	0.0938	0.1051	0.4060	0.3846	0.0113	-0.0214	15
80.60	0.1240	0.1327	0.4240	0.4091	0.0087	-0.0149	15
82.70	0.1310	0.1384	0.4240	0.4124	0.0074	-0.0116	15
99.20	0.1900	0.1860	0.4200	0.4321	-0.0040	0.0121	15
102.00	0.2050	0.1948	0.4110	0.4337	-0.0103	0.0227	15
106.90	0.2250	0.2228	0.4010	0.4513	-0.0022	0.0503	15

Table A-1d

Comparison of Experimental and Calculated VLE Values for the Binary System $N_2(1)$-CO(2)

Pressure atm (psia)	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1	Ref
		70.00 K (C_{12} = 0.0007)					
0.36	0.8100	0.8356	0.9100	0.8956	0.0256	-0.0144	52
0.32	0.6000	0.6337	0.7280	0.7486	0.0337	0.0186	52
0.29	0.4000	0.4274	0.6000	0.5618	0.0274	-0.0382	52
0.25	0.2010	0.1753	0.3100	0.2695	-0.0257	-0.0405	52
		75.00 K (C_{12} = 0.0027)					
0.70	0.8010	0.8322	0.8750	0.8865	0.0312	0.0115	52
0.64	0.6000	0.6301	0.7000	0.7322	0.0301	0.0322	52
0.58	0.4000	0.4065	0.5200	0.5289	0.0065	0.0089	52
0.51	0.2010	0.1824	0.2900	0.2720	-0.0186	-0.0180	52
		79.20 K (C_{12} = 0.0046)					
1.16	0.8600	0.8460	0.8600	0.8910	-0.0140	0.0310	52
1.08	0.6000	0.6440	0.6900	0.7345	0.0440	0.0445	52
0.98	0.4000	0.4182	0.5000	0.5314	0.0182	0.0314	52
0.87	0.2010	0.1883	0.2700	0.2741	-0.0127	0.0041	52
		83.82 K (C_{12} = 0.0153)					
1.36	0.0991	0.0609	0.1600	0.1024	-0.0382	-0.0577	53
1.44	0.1920	0.1485	0.2830	0.2289	-0.0435	-0.0541	53
1.51	0.2680	0.2218	0.3780	0.3207	-0.0462	-0.0573	53
1.65	0.4450	0.3973	0.5600	0.5059	-0.0477	-0.0541	53
1.75	0.5910	0.5486	0.6850	0.6410	-0.0424	-0.0440	53
1.84	0.7180	0.6888	0.7870	0.7555	-0.0292	-0.0315	53
1.93	0.8580	0.8545	0.8950	0.8853	-0.0035	-0.0097	53
1.97	0.9320	0.9475	0.9500	0.9582	0.0155	0.0082	53
2.01	1.0000	1.0050	1.0000	0.9978	0.0050	-0.0022	53
		90.10 K (C_{12} = 0.0157)					
3.34	0.7920	0.7303	0.8310	0.7810	-0.0618	-0.0500	52
3.32	0.7590	0.7108	0.8030	0.7651	-0.0482	-0.0379	52

Table A-1d (continued)

Pressure atm (psia)	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1	Ref
			90.10 K (C_{12} = 0.0157)				
3.22	0.6500	0.6053	0.7100	0.6779	-0.0447	-0.0321	52
2.91	0.3750	0.3388	0.4520	0.4341	-0.0362	-0.0179	52
2.76	0.2240	0.2267	0.2880	0.3129	0.0027	0.0249	52
2.68	0.1350	0.1707	0.1620	0.2458	0.0357	0.0838	52
			96.70 K (C_{12} = -0.0006)				
5.57	0.8220	0.7585	0.8560	0.8088	-0.0635	-0.0472	52
5.34	0.6930	0.6339	0.7450	0.7002	-0.0591	-0.0448	52
5.30	0.6660	0.6497	0.7210	0.7222	-0.0163	0.0012	52
4.62	0.3280	0.2361	0.3840	0.2952	-0.0919	-0.0888	52
4.53	0.2060	0.1855	0.2600	0.2359	-0.0205	-0.0241	52
4.45	0.1070	0.1404	0.1350	0.1814	0.0334	0.0464	52
			100.00 K (C_{12} = 0.0091)				
6.97	0.7180	0.6516	0.7550	0.7065	-0.0664	-0.0485	52
6.87	0.6600	0.6034	0.7000	0.6637	-0.0566	-0.0363	52
6.24	0.3480	0.3217	0.3950	0.3917	-0.0263	-0.0033	52
6.15	0.3020	0.2843	0.3550	0.3518	-0.0177	-0.0032	52
6.06	0.2600	0.2476	0.3000	0.3115	-0.0124	0.0115	52
5.88	0.1700	0.1761	0.1980	0.2292	0.0061	0.0312	52
5.82	0.1360	0.1529	0.1600	0.2013	0.0169	0.0413	52
			105.10 K (C_{12} = 0.0185)				
9.70	0.6000	0.5428	0.6400	0.5988	-0.0572	-0.0412	52
9.66	0.5840	0.5285	0.6270	0.5859	-0.0555	-0.0411	52
9.44	0.4750	0.4534	0.5200	0.5166	-0.0216	-0.0034	52
8.73	0.2280	0.2414	0.2560	0.3030	0.0134	0.0470	52
8.60	0.1900	0.2066	0.2160	0.2643	0.0166	0.0483	52
			110.00 K (C_{12} = 0.0071)				
13.14	0.6300	0.6396	0.7180	0.6827	0.0096	-0.0353	52
13.07	0.6600	0.6203	0.7000	0.6650	-0.0397	-0.0350	52
12.68	0.5340	0.5150	0.5800	0.5661	-0.0190	-0.0139	52
12.49	0.4850	0.4648	0.5300	0.5176	-0.0202	-0.0124	52

Table A-1d (continued)

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
		110.00 K (C_{12} = 0.0071)					
12.34	0.4400	0.4257	0.4870	0.4792	-0.0143	-0.0078	52
12.20	0.3970	0.3897	0.4430	0.4431	-0.0073	0.0001	52
12.00	0.3500	0.3389	0.3930	0.3911	-0.0111	-0.0019	52
11.62	0.2460	0.2447	0.2800	0.2909	-0.0013	0.0109	52
		110.00 K (C_{12} = 0.0071)					
11.52	0.2150	0.2202	0.2450	0.2639	0.0052	0.0189	52
11.26	0.1490	0.1578	0.1700	0.1933	0.0088	0.0233	52
		116.60 K (C_{12} = 0.0074)					
18.54	0.5870	0.5577	0.6200	0.5957	-0.0293	-0.0243	52
18.40	0.5650	0.5300	0.6000	0.5692	-0.0350	-0.0308	52
17.81	0.4160	0.4157	0.4570	0.4576	-0.0003	0.0006	52
17.76	0.4100	0.4063	0.4450	0.4481	-0.0037	0.0031	52
17.52	0.3600	0.3610	0.3920	0.4024	0.0010	0.0104	52
16.94	0.2440	0.2539	0.2780	0.2910	0.0099	0.0130	52
		121.80 K (C_{12} = 0.0077)					
23.96	0.5420	0.5328	0.5700	0.5620	-0.0092	-0.0080	52
22.33	0.2800	0.2833	0.3000	0.3132	0.0033	0.0132	52
22.07	0.2450	0.2450	0.2560	0.2732	0.0	0.0172	52

Table A-1e

Comparison of Experimental and Calculated VLE Values for the Binary System $N_2(1)$-$CH_4(2)$

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
			90.67 K (C_{12} = 0.0290)				
0.95	0.1330	0.1395	0.8890	0.8813	0.0065	-0.0078	53
1.25	0.2000	0.2033	0.9200	0.9140	0.0033	-0.0060	53
1.40	0.2380	0.2381	0.9280	0.9251	0.0001	-0.0029	53
1.76	0.3410	0.3332	0.9470	0.9447	-0.0078	-0.0023	53
2.03	0.4290	0.4172	0.9590	0.9555	-0.0118	-0.0035	53
2.33	0.5310	0.5196	0.9680	0.9649	-0.0115	-0.0031	53
2.61	0.6280	0.6247	0.9750	0.9725	-0.0033	-0.0025	53
3.14	0.8110	0.8210	0.9850	0.9857	0.0100	0.0007	53
3.60	0.9520	0.9639	0.9950	0.9967	0.0199	0.0017	53
			91.65 K (C_{12} = 0.0385)				
0.21	0.0090	0.0087	0.2950	0.3414	-0.0003	0.0464	54
0.29	0.0190	0.0187	0.4760	0.5240	-0.0003	0.0480	54
0.44	0.0370	0.0384	0.6440	0.6911	0.0014	0.0471	54
1.13	0.1470	0.1438	0.8610	0.8875	-0.0032	0.0265	54
			95.00 K (C_{12} = 0.0404)				
2.22	0.2679	0.2610	0.9216	0.9215	-0.0069	-0.0001	57
2.70	0.3562	0.3544	0.9374	0.9393	-0.0018	0.0019	57
2.98	0.4249	0.4178	0.9460	0.9475	-0.0071	0.0015	57
3.39	0.5271	0.5236	0.9573	0.9578	-0.0035	0.0005	57
3.60	0.5889	0.5833	0.9618	0.9627	-0.0056	0.0009	57
4.19	0.7495	0.7518	0.9756	0.9756	0.0023	0.0	57
4.52	0.8271	0.8367	0.9823	0.9827	0.0096	0.0004	57
			97.15 K (C_{12} = 0.0402)				
0.36	0.0090	0.0084	0.2300	0.2707	-0.0006	0.0407	54
0.47	0.0180	0.0187	0.4050	0.4517	0.0007	0.0467	54
0.67	0.0360	0.0370	0.5700	0.6172	0.0010	0.0472	54
1.66	0.1460	0.1407	0.8270	0.8532	-0.0053	0.0262	54

Table A-1e (continued)

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
			99.82 K (C_{12} = 0.0372)				
1.36	0.0790	0.0857	0.7480	0.7551	0.0067	0.0071	58
1.70	0.1110	0.1179	0.7910	0.8072	0.0069	0.0162	58
2.04	0.1440	0.1523	0.8220	0.8422	0.0083	0.0202	58
2.38	0.1800	0.1890	0.8480	0.8675	0.0090	0.0195	58
2.72	0.2210	0.2285	0.8680	0.8867	0.0075	0.0187	58
3.40	0.3120	0.3172	0.8990	0.9143	0.0052	0.0153	58
4.08	0.4270	0.4210	0.9220	0.9337	-0.0060	0.0117	58
4.76	0.5600	0.5404	0.9400	0.9490	-0.0196	0.0090	58
5.44	0.6820	0.6687	0.9540	0.9621	-0.0134	0.0081	58
6.12	0.7930	0.7926	0.9680	0.9744	-0.0004	0.0064	58
6.80	0.9010	0.9017	0.9850	0.9866	0.0007	0.0016	58
			100.00 K (C_{12} = 0.0395)				
1.96	0.1329	0.1377	0.8336	0.8336	0.0048	0.0	57
2.92	0.2397	0.2424	0.8907	0.8944	0.0027	0.0037	57
3.57	0.3301	0.3281	0.9168	0.9182	-0.0020	0.0014	57
3.94	0.3831	0.3821	0.9277	0.9284	-0.0010	0.0007	57
4.34	0.4463	0.4475	0.9372	0.9382	0.0012	0.0010	57
4.64	0.5069	0.5012	0.9448	0.9448	-0.0057	0.0	57
4.97	0.5638	0.5605	0.9508	0.9513	-0.0033	0.0005	57
5.52	0.6671	0.6669	0.9618	0.9618	-0.0002	0.0	57
6.37	0.8133	0.8189	0.9764	0.9768	0.0056	0.0004	57
			105.00 K (C_{12} = 0.0448)				
3.60	0.2101	0.2030	0.8533	0.8541	-0.0071	0.0008	57
4.45	0.2869	0.2781	0.8855	0.8862	-0.0088	0.0007	57
5.59	0.4108	0.3971	0.9149	0.9154	-0.0137	0.0005	57
6.22	0.4836	0.4730	0.9275	0.9278	-0.0106	0.0003	57
7.15	0.6012	0.5972	0.9440	0.9440	-0.0040	0.0	57
8.31	0.7548	0.7539	0.9626	0.9623	-0.0009	-0.0003	57
9.68	0.8990	0.9119	0.9829	0.9840	0.0129	0.0011	57

Table A-1e (continued)

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
		105.15 K (C_{12} = 0.0376)					
0.69	0.0090	0.0068	0.2110	0.1634	-0.0022	-0.0476	54
0.86	0.0180	0.0171	0.3350	0.3297	-0.0009	-0.0053	54
1.19	0.0360	0.0372	0.5120	0.5144	0.0012	0.0024	54
2.71	0.1450	0.1435	0.7900	0.7969	-0.0015	0.0069	54
		110.00 K (C_{12} = 0.0445)					
4.75	0.2093	0.2061	0.8237	0.8227	-0.0032	-0.0010	57
5.84	0.2856	0.2800	0.8615	0.8603	-0.0056	-0.0012	57
7.35	0.4061	0.3986	0.8950	0.8957	-0.0075	0.0007	57
8.26	0.4843	0.4791	0.9113	0.9119	-0.0052	0.0006	57
8.37	0.5008	0.4895	0.9147	0.9138	-0.0113	-0.0009	57
9.48	0.6056	0.5965	0.9319	0.9307	-0.0091	-0.0012	57
10.44	0.6948	0.6909	0.9456	0.9443	-0.0039	-0.0013	57
11.64	0.7956	0.8015	0.9612	0.9608	0.0059	-0.0004	57
13.01	0.8978	0.9100	0.9790	0.9798	0.0122	0.0008	57
		110.93 K (C_{12} = 0.0426)					
1.70	0.0320	0.0336	0.4270	0.4355	0.0016	0.0085	58
2.04	0.0470	0.0497	0.5240	0.5313	0.0027	0.0073	58
2.38	0.0630	0.0662	0.5980	0.5999	0.0032	0.0019	58
2.72	0.0800	0.0832	0.6490	0.6515	0.0032	0.0025	58
3.40	0.1150	0.1186	0.7210	0.7241	0.0036	0.0031	58
4.08	0.1520	0.1561	0.7680	0.7729	0.0041	0.0049	58
4.76	0.1900	0.1960	0.8010	0.8081	0.0060	0.0071	58
5.44	0.2340	0.2384	0.8290	0.8349	0.0044	0.0059	58
6.12	0.2830	0.2839	0.8510	0.8561	0.0009	0.0051	58
6.80	0.3380	0.3325	0.8700	0.8735	-0.0055	0.0035	58
8.51	0.4860	0.4690	0.9070	0.9068	-0.0170	-0.0002	58
10.21	0.6380	0.6224	0.9330	0.9324	-0.0156	-0.0006	58
11.91	0.7800	0.7740	0.9550	0.9553	-0.0060	0.0003	58
13.61	0.9020	0.9040	0.9770	0.9780	0.0020	0.0010	58
		113.71 K (C_{12} = 0.0555)					
3.40	0.0894	0.0836	0.6491	0.6471	-0.0058	-0.0020	60

Table A-1e (continued)

Pressure atm (psia)	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1	Ref
		113.71 K (C_{12} = 0.0555)					
5.10	0.1671	0.1584	0.7660	0.7681	-0.0087	0.0021	60
6.80	0.2609	0.2453	0.8290	0.8301	-0.0157	0.0011	60
10.17	0.4984	0.4679	0.8990	0.8968	-0.0305	-0.0022	60
13.57	0.7491	0.7450	0.9443	0.9421	-0.0041	-0.0021	60
13.62	0.7495	0.7486	0.9450	0.9428	-0.0009	-0.0023	60
15.14	0.8580	0.8570	0.9645	0.9628	-0.0010	-0.0017	60
17.01	0.9558	0.9660	0.9874	0.9895	0.0102	0.0021	60
		114.57 K (C_{12} = 0.0390)					
1.50	0.0090	0.0083	0.1440	0.1387	-0.0007	-0.0053	54
1.74	0.0180	0.0179	0.2470	0.2570	-0.0001	0.0100	54
2.17	0.0360	0.0359	0.3940	0.4073	-0.0001	0.0133	54
4.58	0.1440	0.1442	0.7090	0.7264	0.0002	0.0174	54
		115.00 K (C_{12} = 0.0533)					
5.80	0.1891	0.1784	0.7782	0.7753	-0.0107	-0.0029	57
7.62	0.2868	0.2701	0.8363	0.8333	-0.0167	-0.0030	57
9.45	0.3989	0.3788	0.8739	0.8710	-0.0201	-0.0029	57
10.98	0.5027	0.4837	0.8983	0.8949	-0.0190	-0.0034	57
12.06	0.5789	0.5637	0.9129	0.9094	-0.0152	-0.0035	57
13.98	0.7110	0.7098	0.9377	0.9336	-0.0013	-0.0041	57
15.44	0.8043	0.8112	0.9538	0.9518	0.0069	-0.0020	57
17.24	0.9040	0.9172	0.9750	0.9754	0.0132	0.0004	57
		120.00 K (C_{12} = 0.0523)					
4.92	0.0977	0.0939	0.6122	0.6074	-0.0038	-0.0048	57
7.45	0.1938	0.1851	0.7458	0.7437	-0.0087	-0.0021	57
9.85	0.2990	0.2847	0.8125	0.8103	-0.0143	-0.0022	57
11.94	0.3978	0.3843	0.8512	0.8483	-0.0135	-0.0028	57
13.76	0.4959	0.4811	0.8773	0.8738	-0.0148	-0.0035	57
15.71	0.5938	0.5928	0.9000	0.8972	-0.0010	-0.0028	57
17.79	0.7055	0.7118	0.9225	0.9202	0.0063	-0.0023	57
19.62	0.7877	0.8074	0.9414	0.9402	0.0197	-0.0012	57

Table A-1e (continued)

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
		122.04 K (C_{12} = 0.0468)					
3.40	0.0360	0.0348	0.3240	0.3450	-0.0012	0.0210	58
4.08	0.0560	0.0556	0.4450	0.4543	-0.0004	0.0093	58
4.76	0.0770	0.0769	0.5290	0.5327	-0.0001	0.0037	58
5.44	0.0990	0.0988	0.5860	0.5918	-0.0002	0.0058	58
6.12	0.1210	0.1214	0.6300	0.6379	0.0004	0.0079	58
6.80	0.1440	0.1446	0.6680	0.6750	0.0006	0.0070	58
8.51	0.2030	0.2059	0.7370	0.7425	0.0029	0.0055	58
10.21	0.2720	0.2722	0.7820	0.7884	0.0002	0.0064	58
11.91	0.3480	0.3443	0.8180	0.8222	-0.0037	0.0042	58
13.61	0.4320	0.4223	0.8460	0.8488	-0.0097	0.0028	58
17.01	0.6000	0.5923	0.8940	0.8902	-0.0077	-0.0038	58
20.41	0.7630	0.7609	0.9340	0.9256	-0.0021	-0.0084	58
23.81	0.8930	0.9009	0.9690	0.9615	0.0079	-0.0075	58
27.22	0.9890	0.9939	0.9970	0.9917	0.0049	-0.0053	58
		122.04 K (C_{12} = 0.0635)					
2.76	0.0165	0.0135	0.1963	0.1903	-0.0030	-0.0060	59
3.40	0.0351	0.0304	0.3466	0.3422	-0.0048	-0.0044	59
5.10	0.0860	0.0770	0.5588	0.5597	-0.0090	0.0009	59
6.79	0.1411	0.1272	0.6655	0.6689	-0.0139	0.0034	59
13.61	0.4283	0.3879	0.8411	0.8409	-0.0404	-0.0002	59
19.37	0.7093	0.6924	0.9109	0.9069	-0.0169	-0.0041	59
20.43	0.7524	0.7467	0.9225	0.9181	-0.0057	-0.0044	59
21.16	0.7837	0.7823	0.9308	0.9260	-0.0014	-0.0048	59
24.09	0.8962	0.9053	0.9589	0.9600	0.0091	0.0011	59
25.86	0.9514	0.9653	0.9785	0.9831	0.0140	0.0046	59
27.10	0.9880	0.9930	0.9946	0.9923	0.0049	-0.0024	59
		126.04 K (C_{12} = 0.0513)					
3.40	0.0140	0.0127	0.1630	0.1472	-0.0013	-0.0158	58
4.08	0.0310	0.0299	0.2950	0.2876	-0.0011	-0.0074	58
4.76	0.0490	0.0475	0.3890	0.3883	-0.0015	-0.0007	58
5.44	0.0660	0.0655	0.4610	0.4641	-0.0005	0.0031	58
6.12	0.0850	0.0839	0.5190	0.5233	-0.0011	0.0043	58

Table A-1e (continued)

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
		126.04 K (C_{12} = 0.0513)					
6.80	0.1040	0.1028	0.5660	0.5709	-0.0012	0.0049	58
8.51	0.1530	0.1519	0.6570	0.6570	-0.0011	0.0	58
10.21	0.2070	0.2043	0.7170	0.7152	-0.0027	-0.0018	58
11.91	0.2660	0.2603	0.7600	0.7574	-0.0057	-0.0026	58
13.61	0.3340	0.3203	0.7930	0.7898	-0.0138	-0.0032	58
17.01	0.4800	0.4528	0.8460	0.8377	-0.0272	-0.0083	58
20.41	0.6180	0.5981	0.8880	0.8740	-0.0199	-0.0141	58
23.81	0.7440	0.7409	0.9220	0.9061	-0.0031	-0.0159	58
27.22	0.8540	0.8634	0.9520	0.9388	0.0094	-0.0132	58
30.62	0.9440	0.9588	0.9800	0.9754	0.0148	-0.0046	58
		127.59 K (C_{12} = 0.0679)					
3.48	0.0097	0.0061	0.1144	0.0817	-0.0036	-0.0327	59
5.10	0.0471	0.0410	0.3786	0.3664	-0.0061	-0.0122	59
6.79	0.0912	0.0795	0.5318	0.5202	-0.0117	-0.0116	59
13.61	0.3040	0.2638	0.7662	0.7579	-0.0402	-0.0083	59
20.28	0.5475	0.5099	0.8564	0.8438	-0.0376	-0.0126	59
26.67	0.7825	0.7749	0.9129	0.9033	-0.0076	-0.0096	59
28.37	0.8353	0.8345	0.9263	0.9200	-0.0008	-0.0063	59
30.14	0.8822	0.8893	0.9410	0.9385	0.0071	-0.0024	59
31.30	0.9122	0.9213	0.9521	0.9516	0.0090	-0.0006	59
31.98	0.9308	0.9387	0.9604	0.9596	0.0079	-0.0007	59
32.66	0.9412	0.9365	0.9644	0.9365	-0.0047	-0.0278	59
33.27	0.9541	0.9493	0.9709	0.9493	-0.0047	-0.0216	59
33.95	0.9674	0.9625	0.9761	0.9625	-0.0048	-0.0135	59
34.16	0.9724	0.9676	0.9781	0.9676	-0.0049	-0.0105	59
34.22	0.9741	0.9692	0.9779	0.9692	-0.0049	-0.0087	59
		133.15 K (C_{12} = 0.0493)					
4.76	0.0080	0.0067	0.0790	0.0655	-0.0013	-0.0135	58
5.44	0.0210	0.0210	0.1780	0.1792	0.0	0.0012	58
6.12	0.0360	0.0354	0.2600	0.2680	-0.0006	0.0080	58
6.80	0.0500	0.0501	0.3300	0.3394	0.0001	0.0094	58
8.51	0.0860	0.0879	0.4600	0.4686	0.0019	0.0086	58

Table A-1e (continued)

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
		133.15 K (C_{12} = 0.0493)					
10.21	0.1250	0.1271	0.5500	0.5554	0.0021	0.0054	58
11.91	0.1640	0.1679	0.6180	0.6178	0.0039	-0.0002	58
13.61	0.2050	0.2105	0.6680	0.6651	0.0055	-0.0029	58
17.01	0.3010	0.3013	0.7390	0.7324	0.0003	-0.0066	58
20.41	0.4060	0.4003	0.7890	0.7789	-0.0057	-0.0101	58
23.81	0.5130	0.5064	0.8290	0.8142	-0.0066	-0.0148	58
27.22	0.6210	0.6160	0.8600	0.8433	-0.0050	-0.0167	58
30.62	0.7250	0.7226	0.8880	0.8694	-0.0024	-0.0186	58
34.02	0.8220	0.8196	0.9120	0.8942	-0.0024	-0.0178	58
		138.46 K (C_{12} = 0.0539)					
6.82	0.0165	0.0157	0.1499	0.1235	-0.0008	-0.0264	59
8.10	0.0411	0.0388	0.2724	0.2557	-0.0022	-0.0167	59
10.21	0.0810	0.0783	0.4183	0.4031	-0.0028	-0.0152	59
13.61	0.1490	0.1453	0.5568	0.5460	-0.0037	-0.0108	59
20.41	0.3037	0.2939	0.6983	0.6914	-0.0098	-0.0069	59
26.67	0.4593	0.4502	0.7677	0.7623	-0.0091	-0.0054	59
33.88	0.6430	0.6464	0.8146	0.8157	0.0034	0.0011	59
37.29	0.7295	0.7382	0.8367	0.8367	0.0087	-0.0020	59
38.10	0.7466	0.7600	0.8392	0.8383	0.0134	-0.0009	59
		144.26 K (C_{12} = 0.0529)					
8.51	0.0090	0.0084	0.0700	0.0590	-0.0006	-0.0110	58
10.21	0.0360	0.0357	0.2100	0.2066	-0.0003	-0.0034	58
11.91	0.0630	0.0636	0.3170	0.3130	0.0006	-0.0040	58
13.61	0.0890	0.0922	0.3950	0.3933	0.0032	-0.0017	58
17.01	0.1450	0.1515	0.5120	0.5064	0.0065	-0.0056	58
20.41	0.2060	0.2139	0.5920	0.5823	0.0079	-0.0097	58
23.81	0.2770	0.2799	0.6510	0.6368	0.0029	-0.0142	58
27.22	0.3520	0.3495	0.6920	0.6779	-0.0025	-0.0141	58
30.62	0.4310	0.4229	0.7260	0.7098	-0.0081	-0.0162	58
34.02	0.5090	0.4999	0.7520	0.7352	-0.0092	-0.0168	58
37.42	0.5880	0.5798	0.7700	0.7553	-0.0082	-0.0147	58
40.82	0.6680	0.6644	0.7830	0.7689	-0.0036	-0.0141	58

Table A-1e (continued)

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
		149.82 K (C_{12} = 0.0575)					
12.18	0.0284	0.0256	0.1529	0.1386	-0.0028	-0.0143	57
13.78	0.0514	0.0483	0.2404	0.2299	-0.0031	-0.0105	57
16.98	0.0985	0.0951	0.3683	0.3620	-0.0034	-0.0063	57
20.34	0.1503	0.1465	0.4579	0.4568	-0.0038	-0.0011	57
26.98	0.2579	0.2549	0.5779	0.5742	-0.0030	-0.0037	57
33.68	0.3778	0.3757	0.6480	0.6443	-0.0021	-0.0037	57
40.42	0.5034	0.5112	0.6867	0.6864	0.0078	-0.0003	57
44.16	0.5866	0.5969	0.6878	0.6954	0.0103	0.0076	57
		160.93 K (C_{12} = 0.0582)					
17.01	0.0098	0.0056	0.0445	0.0247	-0.0041	-0.0198	57
18.81	0.0297	0.0268	0.1221	0.1057	-0.0029	-0.0164	57
20.48	0.0490	0.0466	0.1835	0.1682	-0.0024	-0.0153	57
24.36	0.0957	0.0939	0.2986	0.2810	-0.0018	-0.0176	57
26.84	0.1257	0.1252	0.3471	0.3360	-0.0005	-0.0111	57
30.91	0.1774	0.1790	0.4130	0.4071	0.0016	-0.0059	57
35.79	0.2457	0.2456	0.4671	0.4679	-0.0001	0.0008	57
40.69	0.3128	0.3183	0.5131	0.5111	0.0055	-0.0020	57
44.02	0.3683	0.3722	0.5284	0.5313	0.0039	0.0029	57
47.70	0.4382	0.4400	0.5296	0.5413	0.0018	0.0117	57
48.24	0.4514	0.4494	0.5181	0.5388	-0.0020	0.0207	57
48.31	0.4527	0.4531	0.5178	0.5409	0.0004	0.0231	57
		172.04 K (C_{12} = 0.0709)					
25.21	0.0060	0.0027	0.0194	0.0087	-0.0032	-0.0107	57
26.54	0.0169	0.0157	0.0525	0.0476	-0.0012	-0.0049	57
27.28	0.0245	0.0231	0.0740	0.0680	-0.0014	-0.0060	57
30.82	0.0576	0.0588	0.1532	0.1506	0.0012	-0.0026	57
34.02	0.0924	0.0923	0.2157	0.2099	-0.0001	-0.0058	57
37.42	0.1287	0.1296	0.2646	0.2607	0.0009	-0.0039	57
40.69	0.1657	0.1672	0.3009	0.2996	0.0015	-0.0013	57
47.83	0.2599	0.2613	0.3445	0.3545	0.0014	0.0100	57

Table A-1e (continued)

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
		177.59 K (C_{12} = 0.0867)					
30.62	0.0041	0.0048	0.0120	0.0129	0.0007	0.0009	57
34.05	0.0326	0.0360	0.0794	0.0847	0.0034	0.0054	57
36.33	0.0555	0.0574	0.1266	0.1244	0.0020	-0.0022	57
39.06	0.0828	0.0841	0.1693	0.1649	0.0013	-0.0044	57
41.44	0.1097	0.1086	0.2023	0.1948	-0.0012	-0.0075	57
44.23	0.1391	0.1390	0.2286	0.2238	0.0	-0.0048	57
46.00	0.1613	0.1598	0.2459	0.2387	-0.0015	-0.0072	57
47.63	0.1816	0.1807	0.2495	0.2497	-0.0009	0.0002	57
48.24	0.1929	0.1889	0.2488	0.2532	-0.0040	0.0044	57
48.99	0.2026	0.2014	0.2458	0.2576	-0.0012	0.0118	57
		183.15 K (C_{12} = 0.0995)					
39.19	0.0254	0.0283	0.0493	0.0544	0.0029	0.0051	57
40.69	0.0416	0.0419	0.0774	0.0759	0.0003	-0.0016	57
42.39	0.0568	0.0579	0.0963	0.0978	0.0011	0.0014	57
44.50	0.0785	0.0789	0.1251	0.1214	0.0004	-0.0037	57
45.86	0.0911	0.0934	0.1357	0.1343	0.0023	-0.0014	57
46.88	0.1060	0.1054	0.1425	0.1432	-0.0006	0.0007	57
47.63	0.1165	0.1157	0.1449	0.1497	-0.0008	0.0048	57
47.83	0.1175	0.1169	0.1425	0.1495	-0.0006	0.0070	57

Table A-1f

Comparison of Experimental and Calculated VLE Values for the Binary System CO(1)-CH_4(2)

Pressure atm (psia)	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1	Ref
		90.67 K (C_{12} = 0.0172)					
0.28	0.0422	0.0439	0.6080	0.5781	0.0017	-0.0299	53
0.53	0.1130	0.1196	0.8000	0.7902	0.0066	-0.0098	53
0.73	0.1780	0.1877	0.8590	0.8567	0.0097	-0.0023	53
0.94	0.2530	0.2643	0.8980	0.8958	0.0113	-0.0022	53
1.38	0.4440	0.4519	0.9430	0.9416	0.0079	-0.0014	53
1.52	0.5170	0.5208	0.9530	0.9517	0.0038	-0.0013	53
1.81	0.6710	0.6671	0.9700	0.9684	-0.0039	-0.0016	53
2.06	0.8010	0.7931	0.9800	0.9805	-0.0079	0.0005	53
2.25	0.8920	0.8800	0.9910	0.9885	-0.0120	-0.0025	53
		91.56 K (C_{12} = 0.0351)					
0.26	0.0310	0.0256	0.4320	0.4913	-0.0054	0.0593	54
1.13	0.2460	0.2548	0.8630	0.8995	0.0088	0.0365	54
0.41	0.0590	0.0578	0.6050	0.6826	-0.0012	0.0776	54
0.83	0.1650	0.1629	0.7940	0.8539	-0.0021	0.0599	54
0.66	0.1140	0.1151	0.7360	0.8075	0.0011	0.0715	54
		97.17 K (C_{12} = 0.0299)					
1.23	0.1650	0.1610	0.7760	0.8102	-0.0040	0.0342	54
0.46	0.0310	0.0292	0.3850	0.4407	-0.0018	0.0557	54
0.64	0.0590	0.0575	0.5420	0.6067	-0.0015	0.0647	54
1.00	0.1140	0.1176	0.7030	0.7577	0.0036	0.0547	54
		105.18 K (C_{12} = 0.0250)					
0.86	0.0300	0.0275	0.3530	0.3337	-0.0025	-0.0193	54
2.08	0.1640	0.1601	0.7350	0.7463	-0.0039	0.0113	54
1.73	0.1130	0.1191	0.6640	0.6853	0.0061	0.0214	54
1.19	0.0580	0.0605	0.4970	0.5248	0.0025	0.0278	54
2.64	0.2450	0.2283	0.7890	0.8092	-0.0167	0.0202	54

Table A-1f (continued)

Pressure atm (psia)	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1	Ref
		113.71 K (C_{12} = 0.0210)					
6.80	0.4445	0.4444	0.8670	0.8684	-0.0001	0.0014	18
		114.48 K (C_{12} = 0.0287)					
1.79	0.0300	0.0306	0.2660	0.2907	0.0006	0.0247	54
2.27	0.0570	0.0606	0.4150	0.4483	0.0036	0.0333	54
3.70	0.1640	0.1566	0.6670	0.6777	-0.0074	0.0107	54
4.78	0.2450	0.2360	0.7440	0.7618	-0.0090	0.0178	54
3.10	0.1120	0.1152	0.5870	0.6068	0.0032	0.0198	54
		122.04 K (C_{12} = 0.0404)					
13.61	0.5910	0.6116	0.9065	0.8858	0.0206	-0.0207	18
6.80	0.2100	0.2049	0.6850	0.6941	-0.0051	0.0091	18
		123.90 K (C_{12} = 0.0303)					
3.16	0.0300	0.0264	0.2040	0.2085	-0.0036	0.0045	54
3.86	0.0570	0.0560	0.3420	0.3579	-0.0010	0.0159	54
5.14	0.1110	0.1119	0.5090	0.5263	0.0009	0.0173	54
		130.37 K (C_{12} = 0.0382)					
20.41	0.6160	0.6467	0.8830	0.8655	0.0307	-0.0175	18
13.61	0.3590	0.3540	0.7575	0.7528	-0.0050	-0.0047	18
7.48	0.1220	0.1203	0.5075	0.5052	-0.0017	-0.0023	18
6.80	0.0960	0.0972	0.4430	0.4529	0.0012	0.0099	18
		138.71 K (C_{12} = 0.0544)					
6.80	0.0171	0.0171	0.1055	0.1130	0.0	0.0075	18
		144.26 K (C_{12} = 0.0207)					
38.78	0.7875	0.7841	0.8365	0.8429	-0.0034	0.0064	18
34.02	0.6715	0.6666	0.8060	0.8062	-0.0049	0.0002	18
27.22	0.4750	0.4901	0.7315	0.7349	0.0151	0.0034	18
20.41	0.3110	0.3111	0.6345	0.6262	0.0001	-0.0083	18
13.61	0.1390	0.1375	0.4462	0.4207	-0.0016	-0.0255	18

Table A-1f (continued)

Pressure atm (psia)	x_1		y_1		Δx_1	Δy_1	Ref
	Expl	Calc	Expl	Calc			
		152.59 K (C_{12} = 0.0324).					
13.61	0.0352	0.0352	0.1405	0.1359	0.0	-0.0046	18
		158.15 K (C_{12} = 0.0298)					
45.59	0.5510	0.5483	0.6220	0.5474	-0.0027	-0.0746	18
40.82	0.4560	0.4743	0.6300	0.6132	0.0183	-0.0168	18
27.22	0.2090	0.2155	0.4500	0.4401	0.0065	-0.0099	18
20.41	0.0992	0.0954	0.2805	0.2639	-0.0038	-0.0166	18

Table A-2a

Comparison of Experimental and Calculated VLE Composition for the Binary System $H_2(1)$-$CH_4(2)$ (33), (Optimal C_{ij} Obtained for Individual Isotherms, ICL Equation)

Pressure atm	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1
		183.11 K (C_{12} = -0.22118)				
		$\lvert\Delta x\rvert_{av}$ = 0.0058		$\lvert\Delta y\rvert_{av}$ = 0.0134		
38.64	0.0068	0.0082	0.0338	0.0334	0.0014	-0.0004
39.63	0.0107	0.0111	0.0479	0.0443	0.0004	-0.0036
39.63	0.0107	0.0111	0.0489	0.0443	0.0004	-0.0046
41.44	0.0117	0.0166	0.0589	0.0630	0.0049	0.0041
42.87	0.0173	0.0209	0.0789	0.0767	0.0036	-0.0022
44.29	0.0203	0.0253	0.0907	0.0895	0.0050	-0.0012
48.61	0.0328	0.0388	0.1292	0.1236	0.0060	-0.0056
55.45	0.0529	0.0612	0.1706	0.1654	0.0083	-0.0052
68.11	0.0994	0.1080	0.2028	0.2077	0.0086	0.0049
70.11	0.1084	0.1161	0.1990	0.2131	0.0077	0.0141
72.45	0.1234	0.1263	0.1993	0.2221	0.0029	0.0228
73.14	0.1269	0.1294	0.1896	0.2238	0.0025	0.0342
73.35	0.1257	0.1304	0.1885	0.2225	0.0047	0.0340
74.53	0.1768	0.1870	0.1768	0.2269	0.0102	0.0501
		173.24 K (C_{12} = -0.20589)				
		$\lvert\Delta x\rvert_{av}$ = 0.0184		$\lvert\Delta y\rvert_{av}$ = 0.0304		
30.24	0.0087	0.0096	0.0882	0.0843	0.0009	-0.0039
35.96	0.0196	0.0226	0.1755	0.1698	0.0030	-0.0057
41.81	0.0327	0.0360	0.2422	0.2347	0.0033	-0.0075
48.41	0.0442	0.0514	0.2951	0.2896	0.0072	-0.0055
54.60	0.0590	0.0660	0.3364	0.3285	0.0070	-0.0079
61.23	0.0745	0.0820	0.3656	0.3604	0.0075	-0.0052
69.03	0.0973	0.1012	0.3966	0.3884	0.0039	-0.0082
82.64	0.1358	0.1362	0.4260	0.4211	0.0004	-0.0049
96.27	0.1685	0.1738	0.4229	0.4348	0.0053	0.0119
102.06	0.1941	0.1909	0.4163	0.4424	-0.0032	0.0261
108.87	0.2272	0.2122	0.3949	0.4448	-0.0150	0.0499
112.50	0.2487	0.2243	0.3621	0.4453	-0.0244	0.0832
113.32	0.2623	0.2271	0.3629	0.4416	-0.0352	0.0787
114.72	0.3082	0.3607	0.3082	0.4356	0.0525	0.1274

Table A-2a (continued)

Pressure atm	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1
		163.16 K (C_{12} = -0.19027)				
		$\lvert\Delta x\rvert_{av}$ = 0.0398		$\lvert\Delta y\rvert_{av}$ = 0.0292		
21.39	0.0063	0.0063	0.1192	0.1104	0.0	-0.0088
24.12	0.0111	0.0113	0.1918	0.1794	0.0002	-0.0124
28.34	0.0188	0.0192	0.2686	0.2625	0.0004	-0.0061
39.87	0.0403	0.0407	0.4152	0.4039	0.0004	-0.0114
55.42	0.0688	0.0700	0.5131	0.5015	0.0012	-0.0116
69.03	0.0970	0.0960	0.5580	0.5482	-0.0010	-0.0098
82.64	0.1222	0.1223	0.5825	0.5758	0.0001	-0.0067
96.24	0.1495	0.1492	0.5947	0.5925	-0.0003	-0.0022
109.85	0.1802	0.1767	0.5962	0.6019	-0.0035	0.0057
123.46	0.2155	0.2051	0.5924	0.6054	-0.0104	0.0130
136.38	0.2506	0.2333	0.5745	0.6045	-0.0173	0.0300
156.80	0.3661	0.4965	0.4827	0.5625	0.1304	0.0798
157.48	0.3800	0.4948	0.4733	0.5658	0.1148	0.0925
157.81	0.4465	0.4937	0.4465	0.5650	0.0472	0.1185
		153.20 K (C_{12} = -0.17483)				
		$\lvert\Delta x\rvert_{av}$ = 0.0595		$\lvert\Delta y\rvert_{av}$ = 0.0456		
35.68	0.0357	0.0376	0.5565	0.5456	0.0019	-0.0109
40.82	0.0443	0.0457	0.5954	0.5821	0.0014	-0.0133
40.82	0.0446	0.0457	0.5916	0.5821	0.0011	-0.0095
51.68	0.0622	0.0629	0.6430	0.6345	0.0007	-0.0085
61.23	0.0744	0.0779	0.6732	0.6638	0.0035	-0.0094
62.90	0.0795	0.0806	0.6752	0.6680	0.0011	-0.0072
74.91	0.0974	0.0995	0.6973	0.6909	0.0021	-0.0064
81.65	0.1067	0.1101	0.7015	0.6999	0.0034	-0.0016
88.45	0.1202	0.1209	0.7105	0.7071	0.0007	-0.0034
102.06	0.1417	0.1424	0.7177	0.7167	0.0007	-0.0010
115.66	0.1674	0.1641	0.7198	0.7222	-0.0033	0.0024
136.07	0.2071	0.1969	0.7114	0.7238	-0.0102	0.0124
196.29	0.3773	0.3792	0.6143	0.7548	0.0019	0.1405
200.04	0.3984	0.3984	0.5973	0.7510	0.0020	0.1537
202.42	0.4131	0.7439	0.5770	0.7364	0.3308	0.1594
204.46	0.4434	0.5907	0.5568	0.6613	0.1473	0.1045
205.48	0.5157	0.5880	0.5157	0.6624	0.0723	0.1467

Table A-2a (continued)

Pressure atm	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1
		143.07 K (C_{12} = -0.15914)				
		$\lvert\Delta x\rvert_{av}$ = 0.0747		$\lvert\Delta y\rvert_{av}$ = 0.0597		
13.68	0.0079	0.0086	0.3988	0.3893	0.0007	-0.0095
34.06	0.0366	0.0366	0.6962	0.6908	0.0	-0.0054
40.99	0.0461	0.0460	0.7307	0.7247	-0.0001	-0.0060
54.53	0.0657	0.0643	0.7697	0.7644	-0.0014	-0.0053
68.00	0.0842	0.0824	0.7909	0.7864	-0.0018	-0.0045
81.64	0.1019	0.1005	0.8062	0.7996	-0.0014	-0.0067
85.04	0.1067	0.1050	0.8033	0.8018	-0.0017	-0.0015
102.06	0.1283	0.1273	0.8083	0.8097	-0.0010	0.0014
136.07	0.1794	0.1716	0.8063	0.8137	-0.0078	0.0074
170.10	0.2366	0.2156	0.7901	0.8090	-0.0210	0.0189
204.11	0.2983	0.2596	0.7609	0.7993	-0.0387	0.0384
221.13	0.3327	0.2818	0.7394	0.7928	-0.0510	0.0534
238.14	0.3788	0.2227	0.7102	0.2230	-0.1561	-0.4872
255.24	0.4608	0.6668	0.6447	0.7248	0.2060	0.0801
259.56	0.4892	0.6651	0.6167	0.7269	0.1759	0.1102
260.59	0.6046	0.6679	0.6046	0.7237	0.0633	0.1191
		132.94 K (C_{12} = -0.14344)				
		$\lvert\Delta x\rvert_{av}$ = 0.0110		$\lvert\Delta y\rvert_{av}$ = 0.0086		
27.18	0.0268	0.0278	0.7800	0.7798	0.0010	-0.0002
35.04	0.0376	0.0372	0.8161	0.8138	-0.0004	-0.0023
40.85	0.0436	0.0441	0.8312	0.8301	0.0005	-0.0011
54.46	0.0604	0.0600	0.8543	0.8534	-0.0004	-0.0009
67.70	0.0738	0.0753	0.8663	0.8656	0.0015	-0.0007
68.61	0.0776	0.0764	0.8658	0.8663	-0.0012	0.0005
95.15	0.1068	0.1064	0.8751	0.8768	-0.0004	0.0017
135.74	0.1488	0.1509	0.8711	0.8786	0.0021	0.0075
170.10	0.1972	0.1874	0.8589	0.8744	-0.0098	0.0155
204.66	0.2385	0.2233	0.8410	0.8673	-0.0152	0.0263
238.14	0.2959	0.2575	0.8201	0.8585	-0.0384	0.0384

Table A-2a (continued)

Pressure atm	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1
		123.10 K (C_{12} = -0.12819)				
		$\lvert\Delta x\rvert_{av}$ = 0.0070		$\lvert\Delta y\rvert_{av}$ = 0.0040		
44.56	0.0439	0.0452	0.9088	0.9058	0.0013	-0.0030
54.43	0.0554	0.0553	0.9152	0.9137	-0.0001	-0.0015
58.17	0.0611	0.0591	0.9178	0.9158	-0.0020	-0.0020
68.04	0.0691	0.0690	0.9206	0.9198	-0.0001	-0.0007
68.04	0.0693	0.0690	0.9211	0.9198	-0.0003	-0.0013
85.04	0.0841	0.0858	0.9217	0.9235	0.0017	0.0018
102.06	0.1037	0.1022	0.9233	0.9246	-0.0015	0.0014
136.62	0.1411	0.1345	0.9193	0.9232	-0.0066	0.0039
136.75	0.1402	0.1346	0.9183	0.9232	-0.0056	0.0049
169.75	0.1724	0.1642	0.9098	0.9192	-0.0082	0.0094
203.43	0.2146	0.1934	0.8995	0.9138	-0.0212	0.0143
		118.12 K (C_{12} = -0.12048)				
		$\lvert\Delta x\rvert_{av}$ = 0.0075		$\lvert\Delta y\rvert_{av}$ = 0.0051		
41.16	0.0386	0.0400	0.9284	0.9280	0.0014	-0.0004
54.70	0.0518	0.0530	0.9354	0.9360	0.0012	0.0006
68.31	0.0662	0.0659	0.9397	0.9399	-0.0003	0.0002
102.06	0.0975	0.0966	0.9410	0.9423	-0.0009	0.0013
136.42	0.1304	0.1262	0.9353	0.9402	-0.0042	0.0050
170.10	0.1611	0.1540	0.9285	0.9363	-0.0071	0.0078
204.11	0.1925	0.1810	0.9187	0.9313	-0.0115	0.0127
203.78	0.1941	0.1807	0.9181	0.9314	-0.0134	0.0133
		113.13 K (C_{12} = -0.11274)				
		$\lvert\Delta x\rvert_{av}$ = 0.0069		$\lvert\Delta y\rvert_{av}$ = 0.0053		
27.83	0.0240	0.0258	0.9340	0.9336	0.0018	0.0026
41.09	0.0352	0.0382	0.9490	0.9484	0.0030	-0.0006
54.57	0.0459	0.0504	0.9543	0.9535	0.0045	-0.0008
68.18	0.0594	0.0624	0.9572	0.9559	0.0030	-0.0013
84.78	0.0752	0.0766	0.9569	0.9568	0.0014	-0.0001

Table A-2a (continued)

Pressure atm	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1
		113.13 K (C_{12} = -0.11274)				
		$\lvert\Delta x\rvert_{av}$ = 0.0069		$\lvert\Delta y\rvert_{av}$ = 0.0053		
102.19	0.0900	0.0911	0.9557	0.9565	0.0011	0.0008
135.53	0.1189	0.1177	0.9502	0.9542	-0.0012	0.0040
170.43	0.1485	0.1442	0.9448	0.9504	-0.0043	0.0056
204.11	0.1792	0.1686	0.9361	0.9460	-0.0106	0.0099
242.90	0.2074	0.1955	0.9265	0.9404	-0.0119	0.0139
280.32	0.2335	0.2203	0.9160	0.9347	-0.0132	0.0187
		108.12 K (C_{12} = -0.10498)				
		$\lvert\Delta x\rvert_{av}$ = 0.0037		$\lvert\Delta y\rvert_{av}$ = 0.0016		
35.38	0.0280	0.0314	0.9631	0.9617	0.0034	-0.0014
41.24	0.0327	0.0365	0.9647	0.9641	0.0038	-0.0006
54.63	0.0435	0.0478	0.9668	0.9672	0.0043	0.0003
68.38	0.0546	0.0591	0.9682	0.9684	0.0045	0.0002
77.90	0.0622	0.0667	0.9687	0.9686	0.0045	-0.0001
84.57	0.0684	0.0720	0.9690	0.9685	0.0036	-0.0005
95.26	0.0762	0.0803	0.9681	0.9682	0.0041	0.0001
136.07	0.1056	0.1105	0.9630	0.9654	0.0049	0.0024
168.06	0.1345	0.1328	0.9582	0.9622	-0.0017	0.0040
202.04	0.1591	0.1554	0.9515	0.9583	-0.0037	0.0068

Table A-2b

Comparison of Experimental and Calculated VLE Compositions for the Binary System $H_2(1)$-$CH_4(2)$ (33), (Correlated C_{ij} Obtained from Equation (III-1), ICL Equation)

Pressure atm	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1
		183.11 K (C_{12} = -0.14777)				
		$\lvert\Delta x\rvert_{av}$ = 0.0038		$\lvert\Delta y\rvert_{av}$ = 0.0206		
38.64	0.0068	0.0077	0.0338	0.0331	0.0009	-0.0007
39.63	0.0107	0.0105	0.0479	0.0439	-0.0002	-0.0040
39.63	0.0107	0.0105	0.0489	0.0439	-0.0002	-0.0050
41.44	0.0117	0.0156	0.0589	0.0625	0.0039	0.0036
42.87	0.0173	0.0197	0.0789	0.0762	0.0024	-0.0027
44.29	0.0203	0.0238	0.0907	0.0890	0.0035	-0.0017
48.61	0.0328	0.0365	0.1292	0.1235	0.0037	-0.0057
55.45	0.0529	0.0572	0.1706	0.1660	0.0043	-0.0046
62.26	0.0740	0.0791	0.1968	0.1965	0.0051	-0.0003
62.59	0.0734	0.0803	0.1907	0.1958	0.0069	0.0051
68.11	0.0994	0.0990	0.2028	0.2201	-0.0004	0.0173
70.11	0.1084	0.1062	0.1990	0.2254	-0.0022	0.0264
72.45	0.1234	0.1222	0.1993	0.2431	-0.0012	0.0438
73.14	0.1269	0.1256	0.1896	0.2453	-0.0013	0.0557
73.35	0.1257	0.1245	0.1885	0.2442	-0.0012	0.0557
74.40	0.1487	0.1472	0.1817	0.2506	-0.0015	0.0689
74.53	0.1768	0.1913	0.1768	0.2279	0.0145	0.0511
		173.24 K (C_{12} = -0.22596)				
		$\lvert\Delta x\rvert_{av}$ = 0.0116		$\lvert\Delta y\rvert_{av}$ = 0.0308		
30.24	0.0087	0.0098	0.0882	0.0844	0.0011	-0.0038
35.96	0.0196	0.0231	0.1755	0.1699	0.0035	-0.0056
41.81	0.0327	0.0368	0.2422	0.2346	0.0041	-0.0076
48.41	0.0442	0.0526	0.2951	0.2892	0.0084	-0.0059
54.60	0.0590	0.0676	0.3364	0.3278	0.0086	-0.0086
61.23	0.0745	0.0840	0.3656	0.3592	0.0095	-0.0064
69.03	0.0973	0.1038	0.3966	0.3864	0.0065	-0.0102
82.64	0.1358	0.1400	0.4260	0.4174	0.0042	-0.0086
96.27	0.1685	0.1793	0.4229	0.4282	0.0108	0.0053
102.06	0.1914	0.1973	0.4163	0.4344	0.0032	0.0181

Table A-2b (continued)

Pressure atm	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1
		173.24 K (C_{12} = -0.22596)				
		$\lvert\Delta x\rvert_{av}$ = 0.0116		$\lvert\Delta y\rvert_{av}$ = 0.0308		
108.87	0.2272	0.2200	0.3949	0.4375	-0.0072	0.0426
112.50	0.2487	0.2464	0.3621	0.4546	-0.0023	0.0925
113.32	0.2623	0.2597	0.3629	0.4582	-0.0026	0.0953
114.72	0.3082	0.3580	0.3082	0.4299	0.0498	0.1217
		163.16 K (C_{12} = -0.24949)				
		$\lvert\Delta x\rvert_{av}$ = 0.0327		$\lvert\Delta y\rvert_{av}$ = 0.0298		
21.39	0.0063	0.0067	0.1192	0.1106	0.0004	-0.0086
24.12	0.0111	0.0122	0.1918	0.1796	0.0011	-0.0122
28.34	0.0188	0.0207	0.2686	0.2623	0.0019	-0.0063
39.87	0.0403	0.0440	0.4152	0.4023	0.0037	-0.0129
55.42	0.0688	0.0760	0.5131	0.4979	0.0072	-0.0152
69.03	0.0970	0.1046	0.5580	0.5423	0.0076	-0.0157
82.64	0.1222	0.1338	0.5825	0.5678	0.0116	-0.0145
96.24	0.1495	0.1641	0.5947	0.5811	0.0146	-0.0136
109.85	0.1802	0.1955	0.5962	0.5874	0.0153	-0.0088
123.46	0.2155	0.2288	0.5924	0.5839	0.0133	-0.0085
136.38	0.2506	0.2626	0.5745	0.5780	0.0120	0.0035
149.99	0.3095	0.3020	0.5290	0.5731	-0.0075	0.0441
153.39	0.3283	0.3129	0.5156	0.5575	-0.0154	0.0419
156.80	0.3661	0.4667	0.4827	0.5671	0.1006	0.0844
157.48	0.3800	0.4784	0.4733	0.5517	0.0984	0.0784
157.81	0.4465	0.4764	0.4465	0.5541	0.0299	0.1076
		153.20 K (C_{12} = -0.24157)				
		$\lvert\Delta x\rvert_{av}$ = 0.0259		$\lvert\Delta y\rvert_{av}$ = 0.0328		
35.68	0.0357	0.0417	0.5565	0.5431	0.0060	-0.0134
40.82	0.0443	0.0508	0.5954	0.5790	0.0065	-0.0164
40.82	0.0446	0.0508	0.5916	0.5790	0.0062	-0.0126
51.68	0.0622	0.0700	0.6430	0.6300	0.0078	-0.0130
61.23	0.0744	0.0870	0.6732	0.6582	0.0126	-0.0150
62.90	0.0795	0.0900	0.6752	0.6618	0.0105	-0.0134
74.91	0.0974	0.1115	0.6973	0.6829	0.0141	-0.0144

Table A-2b (continued)

Pressure atm	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1
		153.20 K (C_{12} = -0.24157)				
		$\lvert\Delta x\rvert_{av}$ = 0.0259		$\lvert\Delta y\rvert_{av}$ = 0.0328		
81.65	0.1067	0.1237	0.7015	0.6917	0.0170	-0.0098
88.45	0.1202	0.1360	0.7105	0.6978	0.0158	-0.0127
102.06	0.1417	0.1610	0.7177	0.7054	0.0193	-0.0123
115.66	0.1674	0.1863	0.7198	0.7083	0.0189	-0.0115
136.07	0.2071	0.2254	0.7114	0.7057	0.0183	-0.0057
153.09	0.2425	0.2593	0.6976	0.6993	0.0168	0.0017
170.10	0.2843	0.2951	0.6776	0.6875	0.0108	0.0099
187.58	0.3385	0.3347	0.6406	0.6763	-0.0038	0.0357
196.29	0.3773	0.3560	0.6143	0.6674	-0.0213	0.0531
200.04	0.3964	0.3925	0.5973	0.6841	-0.0039	0.0868
202.42	0.4131	0.4090	0.5770	0.6838	-0.0041	0.1068
204.46	0.4434	0.5644	0.5568	0.6454	0.1210	0.0886
205.48	0.5157	0.5642	0.5157	0.6425	0.0485	0.1268
		143.07 K (C_{12} = -0.17364)				
		$\lvert\Delta x\rvert_{av}$ = 0.0670		$\lvert\Delta y\rvert_{av}$ = 0.0334		
13.68	0.0079	0.0088	0.3988	0.3892	0.0009	-0.0096
34.06	0.0366	0.0376	0.6962	0.6901	0.0010	-0.0061
40.99	0.0461	0.0473	0.7307	0.7238	0.0012	-0.0069
54.53	0.0657	0.0661	0.7697	0.7634	0.0004	-0.0063
68.00	0.0842	0.0847	0.7909	0.7851	0.0005	-0.0058
81.65	0.1019	0.1035	0.8062	0.7979	0.0016	-0.0083
85.04	0.1067	0.1081	0.8033	0.8002	0.0014	-0.0031
102.06	0.1283	0.1313	0.8083	0.8076	0.0030	-0.0007
136.07	0.1794	0.1773	0.8063	0.8109	-0.0021	0.0046
170.10	0.2366	0.2232	0.7901	0.8050	-0.0134	0.0149
221.13	0.3327	0.2931	0.7394	0.7867	-0.0396	0.0473
255.24	0.4608	0.7454	0.6447	0.7577	0.2846	0.1130
259.56	0.4892	0.6452	0.6167	0.7353	0.1560	0.1186
260.59	0.6046	0.6588	0.6046	0.7264	0.0542	0.1218

Table A-2b (continued)

Pressure atm	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1
		132.94 K (C_{12} = -0.17484)				
		$\lvert\Delta x\rvert_{av}$ = 0.0122		$\lvert\Delta y\rvert_{av}$ = 0.0099		
27.18	0.0268	0.0297	0.7800	0.7786	0.0029	-0.0014
35.04	0.0376	0.0398	0.8161	0.8124	0.0022	-0.0037
40.85	0.0436	0.0472	0.8312	0.8285	0.0036	-0.0027
54.46	0.0604	0.0644	0.8543	0.8514	0.0039	-0.0029
67.70	0.0738	0.0809	0.8663	0.8633	0.0071	-0.0030
68.61	0.0776	0.0820	0.8658	0.8639	0.0044	-0.0019
95.15	0.1068	0.1146	0.8751	0.8736	0.0078	-0.0016
135.74	0.1488	0.1632	0.8711	0.8739	0.0144	0.0028
170.10	0.1972	0.2036	0.8589	0.8680	0.0064	0.0091
204.66	0.2385	0.2438	0.8410	0.8587	0.0053	0.0177
238.14	0.2959	0.2827	0.8201	0.8477	-0.0132	0.0276
272.16	0.3565	0.3225	0.7894	0.8340	-0.0340	0.0446
		123.10 K (C_{12} = -0.14249)				
		$\lvert\Delta x\rvert_{av}$ = 0.0036		$\lvert\Delta y\rvert_{av}$ = 0.0034		
44.56	0.0439	0.0468	0.9088	0.9052	0.0029	-0.0036
54.43	0.0554	0.0574	0.9152	0.9130	0.0020	-0.0022
58.17	0.0611	0.0613	0.9178	0.9150	0.0002	-0.0028
68.04	0.0691	0.0717	0.9206	0.9190	0.0026	-0.0016
68.04	0.0693	0.0717	0.9211	0.9190	0.0024	-0.0021
85.04	0.0841	0.0892	0.9217	0.9225	0.0051	0.0008
102.06	0.1037	0.1063	0.9233	0.9235	0.0026	0.0002
136.62	0.1411	0.1401	0.9193	0.9215	-0.0010	0.0022
136.75	0.1402	0.1402	0.9183	0.9215	0.0	0.0032
169.75	0.1724	0.1713	0.9098	0.9171	-0.0011	0.0073
203.43	0.2146	0.2021	0.8995	0.9110	-0.0125	0.0115
		118.12 K (C_{12} = -0.13485)				
		$\lvert\Delta x\rvert_{av}$ = 0.0026		$\lvert\Delta y\rvert_{av}$ = 0.0040		
41.16	0.0386	0.0416	0.9284	0.9275	0.0030	-0.0009
54.70	0.0518	0.0552	0.9354	0.9354	0.0034	0.0
68.31	0.0662	0.0686	0.9397	0.9392	0.0024	-0.0005
102.06	0.0975	0.1007	0.9410	0.9413	0.0032	0.0003

Table A-2b (continued)

Pressure atm	x_1 Expl	x_1 Calc	y_1 Expl	y_1 Calc	Δx_1	Δy_1
		118.12 K (C_{12} = -0.13485)				
		$\lvert\Delta x\rvert_{av}$ = 0.0026		$\lvert\Delta y\rvert_{av}$ = 0.0040		
136.42	0.1304	0.1319	0.9353	0.9388	0.0015	0.0035
170.10	0.1611	0.1612	0.9285	0.9344	0.0001	0.0059
204.11	0.1925	0.1897	0.9187	0.9290	-0.0028	0.0103
203.78	0.1941	0.1894	0.9181	0.9290	-0.0047	0.0109
		113.13 K (C_{12} = -0.12014)				
		$\lvert\Delta x\rvert_{av}$ = 0.0045		$\lvert\Delta y\rvert_{av}$ = 0.0049		
27.83	0.0240	0.0264	0.9340	0.9364	0.0024	0.0024
41.09	0.0352	0.0390	0.9490	0.9481	0.0038	-0.0009
54.57	0.0459	0.0515	0.9543	0.9533	0.0056	-0.0010
68.18	0.0594	0.0638	0.9572	0.9556	0.0044	-0.0016
84.78	0.0752	0.0783	0.9569	0.9564	0.0031	-0.0005
102.19	0.0900	0.0932	0.9557	0.9561	0.0032	0.0004
135.53	0.1189	0.1206	0.9502	0.9536	0.0017	0.0034
170.43	0.1485	0.1479	0.9448	0.9496	-0.0006	0.0048
204.11	0.1792	0.1730	0.9361	0.9450	-0.0062	0.0089
242.90	0.2074	0.2008	0.9265	0.9391	-0.0066	0.0126
280.32	0.2335	0.2265	0.9160	0.9330	-0.0070	0.0170
		108.12 K (C_{12} = -0.09593)				
		$\lvert\Delta x\rvert_{av}$ = 0.0040		$\lvert\Delta y\rvert_{av}$ = 0.0019		
35.38	0.0280	0.0305	0.9631	0.9619	0.0025	-0.0012
41.24	0.0327	0.0354	0.9647	0.9643	0.0027	-0.0004
54.63	0.0435	0.0464	0.9668	0.9674	0.0029	0.0006
63.38	0.0546	0.0574	0.9682	0.9687	0.0028	0.0005
77.90	0.0622	0.0648	0.9687	0.9689	0.0026	0.0002
84.57	0.0684	0.0699	0.9690	0.9689	0.0015	-0.0001
95.26	0.0762	0.0779	0.9681	0.9686	0.0017	0.0005
136.07	0.1056	0.1071	0.9630	0.9660	0.0015	0.0030
168.06	0.1345	0.1286	0.9582	0.9630	-0.0059	0.0048
202.04	0.1591	0.1503	0.9515	0.9593	-0.0088	0.0078

APPENDIX B

EXPERIMENTAL AND CALCULATED VLE VALUES FOR TERNARY SYSTEMS

The ICL and the modified SRK equations were used to calculate the vapor and liquid compositions by fixing system temperature, pressure, and the liquid composition of the least volatile component of that system.

APPENDIX B

Table B-1

Comparison of Experimental and Calculated VLE Compositions for Three Hydrogen Containing Ternary Systems (31), (Correlated C_{ij} Obtained from Equation (III-1), ICL Equation)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-N_2(2)-CO(3) at 83.15 K (C_{12}=-0.03902, C_{13}=-0.02517, C_{23}=-0.00686)							
		$\lvert\Delta x\rvert_{av}$ = 0.0040		$\lvert\Delta y\rvert_{av}$ = 0.0088			
21.43	1	0.0404	0.0436	0.8865	0.8993	0.0032	0.0128
	2	0.2186	0.2154	0.0416	0.0311	-0.0032	-0.0105
21.43	1	0.0460	0.0498	0.8662	0.8706	0.0038	0.0044
	2	0.8270	0.8232	0.1169	0.1164	-0.0038	-0.0005
34.02	1	0.0658	0.0681	0.9250	0.9224	0.0023	-0.0026
	2	0.1165	0.1142	0.0158	0.0137	-0.0023	-0.0021
34.02	1	0.0672	0.0701	0.9198	0.9179	0.0029	-0.0019
	2	0.2170	0.2141	0.0285	0.0255	-0.0029	-0.0030
34.02	1	0.0726	0.0752	0.9073	0.9069	0.0026	-0.0004
	2	0.4784	0.4758	0.0583	0.0562	-0.0026	-0.0021
34.02	1	0.0745	0.0777	0.9018	0.9015	0.0032	-0.0003
	2	0.6195	0.6163	0.0710	0.0727	-0.0032	0.0017
34.02	1	0.0755	0.0795	0.8987	0.8978	0.0040	-0.0009
	2	0.7185	0.7146	0.0819	0.0845	-0.0040	0.0026
34.02	1	0.0761	0.0811	0.8964	0.8943	0.0050	-0.0021
	2	0.8176	0.8126	0.0932	0.0964	-0.0050	0.0032
95.26	1	0.1815	0.1826	0.9080	0.9178	0.0011	0.0098
	2	0.0940	0.0929	0.0168	0.0127	-0.0011	-0.0041
95.26	1	0.1828	0.1840	0.9067	0.9168	0.0012	0.0101
	2	0.1122	0.1110	0.0201	0.0152	-0.0012	-0.0049
95.26	1	0.1950	0.1969	0.8950	0.9071	0.0019	0.0121
	2	0.2860	0.2841	0.0483	0.0398	-0.0019	-0.0085
95.26	1	0.2094	0.2114	0.8831	0.8965	0.0020	0.0134
	2	0.4796	0.4776	0.0810	0.0691	-0.0020	-0.0119
95.26	1	0.2190	0.2217	0.8765	0.8890	0.0027	0.0125
	2	0.6212	0.6185	0.1049	0.0923	-0.0027	-0.0126
95.26	1	0.2231	0.2263	0.8731	0.8856	0.0032	0.0125
	2	0.6864	0.6832	0.1159	0.1034	-0.0032	-0.0125
136.08	1	0.2815	0.2725	0.8508	0.8841	-0.0090	0.0333
	2	0.2270	0.2360	0.0440	0.0447	0.0090	0.0007
136.08	1	0.3000	0.2892	0.8356	0.8733	-0.0108	0.0377

Table B-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-N_2(2)-CO(3) at 83.15 K (C_{12}=-0.03902, C_{13}=-0.02517, C_{23}=-0.00686)							
			x Π• = 0.0040		y Π• = 0.0088		
136.08	2	0.3500	0.3608	0.0743	0.0717	0.0108	-0.0026
136.08	1	0.3317	0.3193	0.8090	0.8532	-0.0124	0.0442
	2	0.5644	0.5768	0.1550	0.1276	0.0124	-0.0274
System H_2(1)-N_2(2)-CO(3) at 99.82 K (C_{12}=-0.06291, C_{13}=-0.06767, C_{23}=-0.00846)							
			$\lvert\Delta x\rvert_{av}$ = 0.0053		$\lvert\Delta y\rvert_{av}$ = 0.0093		
21.43	1	0.0393	0.0410	0.6682	0.6509	0.0017	-0.0174
	2	0.0811	0.0794	0.0392	0.0386	-0.0017	-0.0006
21.43	1	0.0398	0.0413	0.6504	0.6333	0.0015	-0.0171
	2	0.1970	0.1955	0.0958	0.0943	-0.0015	-0.0015
21.43	1	0.0400	0.0415	0.6357	0.6190	0.0015	-0.0167
	2	0.2980	0.2965	0.1390	0.1423	-0.0015	0.0033
21.43	1	0.0397	0.0419	0.5919	0.5864	0.0022	-0.0055
	2	0.5493	0.5471	0.2563	0.2607	-0.0022	0.0044
21.43	1	0.0392	0.0420	0.5680	0.5675	0.0028	-0.0005
	2	0.7102	0.7074	0.3315	0.3369	-0.0028	0.0054
21.43	1	0.0385	0.0420	0.5563	0.5507	0.0035	-0.0056
	2	0.8651	0.8616	0.4037	0.4116	-0.0035	0.0079
21.43	1	0.0380	0.0419	0.5519	0.5429	0.0039	-0.0090
	2	0.9413	0.9374	0.4393	0.4489	-0.0039	0.0096
34.02	1	0.0689	0.0735	0.7483	0.7331	0.0046	-0.0152
	2	0.0766	0.0720	0.0335	0.0273	-0.0046	-0.0062
34.02	1	0.0707	0.0748	0.7318	0.7193	0.0041	-0.0125
	2	0.1908	0.1867	0.0797	0.0705	-0.0041	-0.0092
34.02	1	0.0720	0.0759	0.7190	0.7073	0.0039	-0.0117
	2	0.2968	0.2929	0.1133	0.1104	-0.0039	-0.0029
34.02	1	0.0744	0.0791	0.6868	0.6705	0.0047	-0.0163
	2	0.6576	0.6529	0.2352	0.2473	-0.0047	0.0121
34.02	1	0.0745	0.0792	0.6851	0.6688	0.0047	-0.0163
	2	0.6761	0.6714	0.2422	0.2544	-0.0047	0.0122
34.02	1	0.0744	0.0801	0.6740	0.6548	0.0057	-0.0193
	2	0.8342	0.8285	0.2985	0.3162	-0.0057	0.0177
34.02	1	0.0744	0.0802	0.6732	0.6538	0.0058	-0.0194
	2	0.8455	0.8397	0.3023	0.3207	-0.0058	0.0184

Table B-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-N_2(2)-CO(3) at 99.82 K (C_{12}=-0.06291, C_{13}=-0.06767, C_{23}=0.00846)							
			$\lvert\Delta x\rvert_{av}$ = 0.0053		$\lvert\Delta y\rvert_{av}$ = 0.0093		
54.43	1	0.1281	0.1285	0.7708	0.7671	0.0004	-0.0037
	2	0.1342	0.1338	0.0492	0.0443	-0.0004	-0.0049
54.43	1	0.1342	0.1334	0.7539	0.7489	-0.0008	-0.0050
	2	0.3074	0.3082	0.1099	0.1030	0.0008	-0.0069
54.43	1	0.1366	0.1378	0.7398	0.7320	0.0012	-0.0078
	2	0.4826	0.4814	0.1627	0.1628	-0.0012	0.0001
54.43	1	0.1378	0.1416	0.7290	0.7166	0.0038	-0.0012
	2	0.6556	0.6518	0.2120	0.2239	-0.0038	0.0119
54.43	1	0.1381	0.1429	0.7256	0.7109	0.0048	-0.0147
	2	0.7230	0.7182	0.2340	0.2484	-0.0048	0.0144
54.43	1	0.1383	0.1446	0.7213	0.7022	0.0063	-0.0191
	2	0.8313	0.8250	0.2693	0.2887	-0.0063	0.0194
74.85	1	0.1860	0.1832	0.7760	0.7846	-0.0028	0.0086
	2	0.0717	0.0745	0.0257	0.0247	0.0028	-0.0010
74.85	1	0.2030	0.1955	0.7448	0.7478	-0.0075	0.0030
	2	0.3452	0.3527	0.1237	0.1223	0.0075	-0.0014
74.85	1	0.2074	0.2027	0.7317	0.7320	-0.0047	0.0003
	2	0.4995	0.5042	0.1790	0.1794	0.0047	0.0004
74.85	1	0.2102	0.2100	0.7208	0.7141	-0.0002	-0.0067
	2	0.6817	0.6819	0.2443	0.2511	0.0002	0.0068
74.85	1	0.2114	0.2128	0.7181	0.7068	0.0014	-0.0113
	2	0.7611	0.7597	0.2727	0.2841	-0.0014	0.0114
95.26	1	0.2527	0.2389	0.7578	0.7684	-0.0139	0.0106
	2	0.0663	0.0802	0.0330	0.0292	0.0139	-0.0038
95.26	1	0.2662	0.2474	0.7357	0.7543	-0.0188	0.0186
	2	0.1634	0.1822	0.0813	0.0681	0.0188	-0.0132
95.26	1	0.2792	0.2589	0.7090	0.7365	-0.0203	0.0275
	2	0.2965	0.3168	0.1476	0.1229	0.0203	-0.0247
95.26	1	0.3001	0.2868	0.6565	0.6908	-0.0133	0.0343
	2	0.6522	0.6655	0.3247	0.2904	0.0133	-0.0344
System H_2(1)-N_2(2)-CO(3) at 122.04 K (C_{12}=-0.09475, C_{23}=-0.12432, C_{23}=0.01059)							
			$\lvert\Delta x\rvert_{av}$ = 0.0039		$\lvert\Delta y\rvert_{av}$ = 0.0162		
34.02	1	0.0422	0.0501	0.1974	0.2121	0.0079	0.0147
	2	0.1481	0.1402	0.1395	0.1306	-0.0079	-0.0089

Table B-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-N_2(2)-CO(3) at 122.04 K (C_{12}=-0.09475, C_{23}=-0.12432, C_{23}=0.01059)							
		$\|\Delta x\|_{av}$ = 0.0039		$\|\Delta y\|_{av}$ = 0.0162			
34.02	1	0.0395	0.0472	0.1660	0.1799	0.0077	0.0140
	2	0.3279	0.3202	0.3090	0.2973	-0.0077	-0.0117
34.02	1	0.0356	0.0441	0.1357	0.1520	0.0085	0.0163
	2	0.5083	0.4998	0.4791	0.4637	-0.0085	-0.0154
34.02	1	0.0332	0.0423	0.1204	0.1381	0.0091	0.0177
	2	0.6088	0.5997	0.5738	0.5569	-0.0091	-0.0169
34.02	1	0.0288	0.0287	0.0945	0.0286	-0.0001	-0.0659
	2	0.8242	0.8243	0.7768	0.8244	0.0001	0.0476
54.43	1	0.1387	0.1486	0.3095	0.3266	0.0099	0.0171
	2	0.1330	0.1231	0.1120	0.1047	-0.0099	-0.0073
System H_2(1)-N_2(2)-CH_4(3) at 144.26 K (C_{12}=-0.12659, C_{13}=-0.16098, C_{23}=0.06317)							
		$\|\Delta x\|_{av}$ = 0.0015		$\|\Delta y\|_{av}$ = 0.0182			
34.02	1	0.0286	0.0309	0.4337	0.4689	0.0023	0.0352
	2	0.0998	0.0975	0.2191	0.2111	-0.0023	-0.0080
34.02	1	0.0155	0.0195	0.1677	0.1995	0.0040	0.0318
	2	0.2739	0.2699	0.5087	0.4986	-0.0040	-0.0101
34.02	1	0.0030	0.0075	0.0236	0.0529	0.0045	0.0293
	2	0.4176	0.4131	0.6793	0.6648	-0.0045	-0.0145
68.05	1	0.0826	0.0835	0.6735	0.6881	0.0009	0.0146
	2	0.0617	0.0608	0.0838	0.0846	-0.0009	0.0008
68.05	1	0.0849	0.0853	0.5002	0.5373	0.0004	0.0371
	2	0.1790	0.1787	0.2377	0.2315	-0.0004	-0.0062
68.05	1	0.0915	0.0935	0.3130	0.3370	0.0020	0.0240
	2	0.3733	0.3713	0.4291	0.4256	-0.0020	-0.0035
68.05	1	0.0932	0.0949	0.3005	0.3203	0.0017	0.0198
	2	0.3912	0.3895	0.4413	0.4414	-0.0017	0.0001
68.05	1	0.0926	0.0966	0.2758	0.3020	0.0040	0.0262
	2	0.4137	0.4097	0.4637	0.4585	-0.0040	-0.0052
System H_2(1)-CO(2)-CH_4(3) at 173.24 K (C_{12}=-0.25484, C_{13}=-0.20589, C_{23}=0.03749)							
		$\|\Delta x\|_{av}$ = 0.0025		$\|\Delta y\|_{av}$ = 0.0136			
40.82	1	0.0286	0.0297	0.1818	0.1829	0.0011	0.0011
	2	0.0301	0.0290	0.0594	0.0543	-0.0011	-0.0051
40.82	1	0.0059	0.0110	0.0320	0.0471	0.0051	0.0151
	2	0.1503	0.1452	0.2528	0.2389	-0.0051	-0.0139

Table B-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System $H_2(1)$-$CO(2)$-$CH_4(3)$ at 173.24 K (C_{12}=-0.25484; C_{13}=-0.20589, C_{23}=0.03749)			$\lvert\Delta x\rvert_{av}$ = 0.0025		$\lvert\Delta y\rvert_{av}$ = 0.0136		
40.82	1	0.0020	0.0044	0.0099	0.0169	0.0024	0.0070
	2	0.1820	0.1796	0.2848	0.2831	-0.0024	-0.0017
54.43	1	0.0578	0.0632	0.2871	0.2916	0.0054	0.0045
	2	0.0322	0.0268	0.0517	0.0421	-0.0054	-0.0096
54.43	1	0.0300	0.0369	0.0300	0.0502	0.0069	0.0202
	2	0.2900	0.2831	0.2900	0.3096	-0.0069	0.0196
68.04	1	0.0935	0.0980	0.3485	0.3525	0.0045	0.0040
	2	0.0304	0.0259	0.0427	0.0357	-0.0045	-0.0070
68.04	1	0.0877	0.0972	0.1540	0.1949	0.0095	0.0409
	2	0.1750	0.1655	0.2038	0.1967	-0.0095	-0.0071
68.04	1	0.0916	0.0986	0.1340	0.1725	0.0070	0.0385
	2	0.1913	0.1843	0.2086	0.2119	-0.0070	0.0033
102.06	1	0.1976	0.1957	0.3828	0.4168	-0.0019	0.0340
	2	0.0168	0.0187	0.0188	0.0212	0.0019	0.0024
102.06	1	0.1271	0.1265	0.1271	0.1264	-0.0006	-0.0007
	2	0.0461	0.0467	0.0461	0.0467	0.0006	0.0006
System $H_2(1)$-$CO(2)$-$CH_4(3)$ at 163.16 K (C_{12}=-0.22914, C_{13}=-0.19027, C_{23}=0.03622)			$\lvert\Delta x\rvert_{av}$ = 0.0039		$\lvert\Delta y\rvert_{av}$ = 0.0157		
27.22	1	0.0131	0.0154	0.2050	0.2109	0.0023	0.0059
	2	0.0179	0.0156	0.0500	0.0394	-0.0023	-0.0106
27.22	1	0.0096	0.0094	0.1063	0.1144	-0.0002	0.0081
	2	0.0670	0.0672	0.1716	0.1615	0.0002	-0.0102
27.22	1	0.0089	0.0083	0.0933	0.0993	-0.0006	0.0060
	2	0.0754	0.0760	0.1846	0.1810	0.0006	-0.0036
27.22	1	0.0070	0.0052	0.0700	0.0589	-0.0018	-0.0111
	2	0.0988	0.1006	0.2323	0.2340	0.0018	0.0017
40.82	1	0.0350	0.0411	0.3760	0.3836	0.0061	0.0076
	2	0.0234	0.0173	0.0448	0.0327	-0.0061	-0.0121
40.82	1	0.0350	0.0411	0.3490	0.3836	0.0061	0.0346
	2	0.0234	0.0173	0.0422	0.0327	-0.0061	-0.0095
40.82	1	0.0336	0.0394	0.3517	0.3512	0.0058	-0.0005
	2	0.0436	0.0378	0.0812	0.0704	-0.0058	-0.0108
40.82	1	0.0330	0.0351	0.2871	0.2777	0.0021	-0.0094
	2	0.0896	0.0875	0.1578	0.1572	-0.0021	-0.0006

Table B-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-CO(2)-CH_4(3) at 163.16 K (C_{12}=-0.22914, C_{13}=-0.19027, C_{23}=0.03622)			$\lvert\Delta x\rvert_{av}$ = 0.0039		$\lvert\Delta y\rvert_{av}$ = 0.0157		
40.82	1	0.0322	0.0315	0.2335	0.2272	-0.0007	-0.0063
	2	0.1244	0.1251	0.2174	0.2181	0.0007	0.0007
40.82	1	0.0242	0.0262	0.1429	0.1648	0.0020	0.0219
	2	0.1787	0.1767	0.3159	0.2952	-0.0020	-0.0207
40.82	1	0.0198	0.0207	0.1042	0.1139	0.0009	0.0097
	2	0.2254	0.2245	0.3681	0.3598	-0.0009	-0.0083
54.43	1	0.0614	0.0660	0.4340	0.4315	0.0046	-0.0025
	2	0.0512	0.0466	0.0782	0.0719	-0.0046	-0.0063
54.43	1	0.0572	0.0632	0.3603	0.3614	0.0060	0.0011
	2	0.1068	0.1008	0.1576	0.1499	-0.0060	-0.0077
54.43	1	0.0549	0.0614	0.3125	0.3213	0.0065	0.0088
	2	0.1407	0.1342	0.2067	0.1951	-0.0065	-0.0116
54.43	1	0.0458	0.0581	0.2275	0.2591	0.0123	0.0316
	2	0.2030	0.1907	0.2861	0.2661	-0.0123	-0.0200
54.43	1	0.0449	0.0551	0.1833	0.2120	0.0102	0.0287
	2	0.2484	0.2382	0.3380	0.3204	-0.0102	-0.0176
54.43	1	0.0495	0.0527	0.1672	0.1792	0.0032	0.0120
	2	0.2775	0.2743	0.3644	0.3582	-0.0032	-0.0062
54.43	1	0.0408	0.0506	0.1403	0.1544	0.0098	0.0141
	2	0.3140	0.3042	0.3963	0.3872	-0.0098	-0.0091
54.43	1	0.0405	0.0487	0.1206	0.1341	0.0082	0.0135
	2	0.3384	0.3302	0.4177	0.4103	-0.0082	-0.0074
54.43	1	0.0340	0.0341	0.0593	0.0345	0.0001	-0.0025
	2	0.4227	0.4226	0.4541	0.4238	-0.0001	-0.0303
68.04	1	0.0919	0.0937	0.4814	0.4768	0.0018	-0.0046
	2	0.0545	0.0527	0.0722	0.0711	-0.0018	-0.0011
68.04	1	0.0874	0.0937	0.3199	0.3255	0.0063	0.0056
	2	0.1912	0.1849	0.2342	0.2303	-0.0063	-0.0039
68.04	1	0.0854	0.0947	0.2465	0.2642	0.0093	0.0177
	2	0.2548	0.2455	0.3007	0.2930	-0.0093	-0.0077
68.04	1	0.0852	0.0949	0.2398	0.2569	0.0097	0.0171
	2	0.2628	0.2531	0.3065	0.3004	-0.0097	-0.0061
68.04	1	0.0904	0.0964	0.1999	0.2214	0.0060	0.0215
	2	0.2968	0.2908	0.3368	0.3347	-0.0060	-0.0021
68.04	1	0.0979	0.0990	0.1651	0.1926	0.0011	0.0275
	2	0.3252	0.3241	0.3561	0.3622	-0.0011	0.0061

Table B-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-CO(2)-CH_4(3) at 163.16 K (C_{12}=-0.22914, C_{13}=-0.19027, C_{23}=0.03622)							
			|Δx| av = 0.0039		|Δy| av = 0.0157		
68.04	1	0.1072	0.1061	0.1551	0.1941	-0.0011	0.0390
	2	0.3372	0.3383	0.3571	0.3717	0.0011	0.0146
68.04	1	0.1163	0.1047	0.1421	0.1735	-0.0116	0.0314
	2	0.3449	0.3565	0.3564	0.3863	0.0116	0.0299
102.06	1	0.1717	0.1703	0.5009	0.5124	-0.0014	0.0115
	2	0.0689	0.0703	0.0768	0.0775	0.0014	0.0007
102.06	1	0.1795	0.1789	0.4402	0.4572	-0.0006	0.0170
	2	0.1152	0.1158	0.1217	0.1255	0.0006	0.0038
102.06	1	0.1959	0.1867	0.4078	0.4275	-0.0092	0.0197
	2	0.1370	0.1462	0.1443	0.1560	0.0092	0.0117
102.06	1	0.2146	0.1990	0.3485	0.3794	-0.0157	0.0309
	2	0.1666	0.1822	0.1731	0.1914	0.0157	0.0183
102.06	1	0.2760	0.2735	0.2760	0.3926	-0.0025	0.1166
	2	0.1830	0.1855	0.1830	0.1860	0.0025	0.0030
System H_2(1)-CO(2)-CH_4(3) at 143.07 K (C_{12}=-0.17793, C_{13}=-0.15914, C_{23}=0.0337)							
			|Δx| av = 0.0039		|Δy| av = 0.0191		
27.22	1	0.0160	0.0198	0.3088	0.3351	0.0038	0.0263
	2	0.1912	0.1874	0.3721	0.3375	-0.0038	-0.0346
27.22	1	0.0083	0.0132	0.1609	0.1726	0.0049	0.0117
	2	0.3217	0.3168	0.5495	0.5278	-0.0049	-0.0217
27.22	1	0.0078	0.0094	0.1174	0.1074	0.0016	-0.0100
	2	0.3803	0.3787	0.6089	0.6074	-0.0016	0.0005
40.82	1	0.0373	0.0446	0.6118	0.6143	0.0073	0.0025
	2	0.0890	0.0817	0.1082	0.1167	-0.0073	0.0085
40.82	1	0.0380	0.0332	0.5293	0.1713	-0.0048	-0.3580
	2	0.5293	0.5341	0.2007	0.6225	0.0048	0.4218
40.82	1	0.0380	0.0410	0.3841	0.3890	0.0030	0.0049
	2	0.2820	0.2790	0.3569	0.3644	-0.0030	0.0075
40.82	1	0.0329	0.0405	0.3539	0.3633	0.0076	0.0094
	2	0.3125	0.3050	0.3916	0.3936	-0.0076	0.0020
40.82	1	0.0272	0.0331	0.1711	0.1693	0.0059	-0.0018
	2	0.5426	0.5367	0.6279	0.6249	-0.0059	-0.0030
40.82	1	0.0196	0.0291	0.1121	0.1181	0.0095	0.0060
	2	0.6224	0.6129	0.7000	0.6913	-0.0095	-0.0087

Table B-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-CO(2)-CH_4(3) at 143.07 K (C_{12}=-0.17793, C_{13}=-0.15914, C_{23}=0.0337)							
		$\lvert\Delta x\rvert_{av}$ = 0.0039		$\lvert\Delta y\rvert_{av}$ = 0.0191			
40.82	1	0.0214	0.0276	0.0991	0.1034	0.0062	0.0043
	2	0.6426	0.6364	0.7194	0.7109	-0.0062	-0.0085
40.82	1	0.0219	0.0274	0.0959	0.1011	0.0055	0.0052
	2	0.6456	0.6401	0.7180	0.7140	-0.0055	-0.0040
40.82	1	0.0115	0.0115	0.0323	0.0115	0.0	-0.0208
	2	0.7542	0.7542	0.8060	0.7545	0.0	-0.0515
40.82	1	0.0394	0.0426	0.5091	0.4769	0.0032	-0.0322
	2	0.1995	0.1963	0.2228	0.2662	-0.0032	0.0434
54.43	1	0.0651	0.0660	0.5712	0.5586	0.0009	-0.0126
	2	0.1901	0.1892	0.2083	0.2162	-0.0009	0.0079
54.43	1	0.0692	0.0662	0.5610	0.5434	-0.0030	-0.0176
	2	0.2016	0.2046	0.2193	0.2325	0.0030	0.0132
54.43	1	0.0630	0.0664	0.5436	0.5315	0.0034	-0.0121
	2	0.2201	0.2167	0.2379	0.2453	-0.0034	0.0074
54.43	1	0.0704	0.0703	0.3444	0.3458	-0.0001	0.0014
	2	0.4262	0.4263	0.4545	0.4501	0.0001	-0.0044
54.43	1	0.0673	0.0745	0.2405	0.2440	0.0072	0.0035
	2	0.5624	0.5552	0.5704	0.5672	-0.0072	-0.0032
54.43	1	0.0787	0.0842	0.1366	0.1457	0.0055	0.0091
	2	0.6792	0.6737	0.6767	0.6762	-0.0055	-0.0005
54.43	1	0.0941	0.0932	0.1161	0.1525	-0.0009	0.0365
	2	0.0834	0.6843	0.6838	0.6811	0.0009	-0.0027
68.04	1	0.0837	0.0854	0.6972	0.6937	0.0017	-0.0035
	2	0.0924	0.0907	0.0921	0.0946	-0.0017	0.0025
68.04	1	0.0856	0.0859	0.6817	0.6792	0.0003	-0.0025
	2	0.1059	0.1056	0.1015	0.1097	-0.0003	0.0082
68.04	1	0.0880	0.0908	0.5863	0.5736	0.0028	-0.0128
	2	0.2200	0.2172	0.2100	0.2191	-0.0028	0.0091
68.04	1	0.0839	0.0926	0.5453	0.5434	0.0093	-0.0019
	2	0.2598	0.2505	0.2499	0.2507	-0.0093	0.0008
68.04	1	0.0980	0.0997	0.4495	0.4510	0.0017	0.0015
	2	0.3578	0.3561	0.3451	0.3483	-0.0017	0.0032
68.04	1	0.1147	0.1212	0.2747	0.2995	0.0065	0.0248
	2	0.5355	0.5290	0.5247	0.5093	-0.0065	-0.0154

Table B-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-CO(2)-CH_4(3) at 143.07 K (C_{12}=-0.17793, C_{13}=-0.15914, C_{23}=0.0337)							
			$\lvert\Delta x\rvert_{av}$ = 0.0039		$\lvert\Delta y\rvert_{av}$ = 0.0191		
68.04	1	0.1251	0.1263	0.2646	0.2780	0.0012	0.0134
	2	0.5522	0.5510	0.5335	0.5315	-0.0012	-0.0020
68.04	1	0.1561	0.1371	0.2344	0.2478	-0.0190	0.0134
	2	0.5638	0.5828	0.5565	0.5643	0.0190	0.0078
102.06	1	0.1393	0.1359	0.7364	0.7326	-0.0034	-0.0038
	2	0.0820	0.0854	0.0677	0.0745	0.0034	0.0068
102.06	1	0.1564	0.1503	0.6394	0.6367	-0.0061	-0.0027
	2	0.1862	0.1923	0.1552	0.1670	0.0061	0.0118
102.06	1	0.1628	0.1624	0.5701	0.5759	-0.0004	0.0058
	2	0.2581	0.2585	0.2171	0.2250	0.0004	0.0079
102.06	1	0.2056	0.1986	0.4336	0.4619	-0.0070	0.0283
	2	0.3703	0.3773	0.3329	0.3345	0.0070	0.0016
102.06	1	0.2311	0.2132	0.3991	0.4253	-0.0179	0.0262
	2	0.3863	0.4042	0.3600	0.3649	0.0179	0.0049
System H_2(1)-CO(2)-CH_4(3) at 123.10 K (C_{12}=-0.12702, C_{13}=-0.12819, C_{23}=0.03119)							
			$\lvert\Delta x\rvert_{av}$ = 0.0033		$\lvert\Delta y\rvert_{av}$ = 0.0074		
27.22	1	0.0233	0.0270	0.8100	0.8117	0.0037	0.0017
	2	0.0611	0.0574	0.0690	0.0660	-0.0037	-0.0030
27.22	1	0.0256	0.0273	0.6693	0.6720	0.0017	0.0027
	2	0.2005	0.1988	0.2089	0.2143	-0.0017	0.0054
27.22	1	0.0230	0.0279	0.5458	0.5351	0.0049	-0.0107
	2	0.3672	0.3623	0.3512	0.3634	-0.0049	0.0122
27.22	1	0.0244	0.0282	0.4828	0.4633	0.0038	-0.0195
	2	0.4636	0.4598	0.4267	0.4437	-0.0038	0.0170
27.22	1	0.0248	0.0286	0.3938	0.3851	0.0038	-0.0087
	2	0.5780	0.5742	0.5295	0.5337	-0.0038	0.0042
27.22	1	0.0253	0.0283	0.2914	0.2822	0.0030	-0.0092
	2	0.7357	0.7327	0.6512	0.6582	-0.0030	0.0070
27.22	1	0.0208	0.0278	0.2341	0.2512	0.0070	0.0171
	2	0.7877	0.7807	0.7070	0.6976	-0.0070	-0.0094
40.82	1	0.0397	0.0422	0.8596	0.8561	0.0025	-0.0035
	2	0.0569	0.0544	0.0468	0.0472	-0.0025	0.0004
40.82	1	0.0423	0.0440	0.7925	0.7838	0.0017	-0.0087
	2	0.1486	0.1469	0.1180	0.1229	-0.0017	0.0049

Table B-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-CO(2)-CH_4(3) at 123.10 K (C_{12}=-0.12702, C_{13}=-0.12819, C_{23}=0.03119)							
		$\|\Delta x\|_{av}$ = 0.0033			$\|\Delta y\|_{av}$ = 0.0074		
40.82	1	0.0495	0.0482	0.6840	0.6709	-0.0013	-0.0131
	2	0.3071	0.3084	0.2334	0.2434	0.0013	0.0100
40.82	1	0.0496	0.0494	0.6621	0.6455	-0.0002	-0.0166
	2	0.3477	0.3479	0.2567	0.2710	0.0002	0.0143
40.82	1	0.0503	0.0524	0.6022	0.5906	0.0021	-0.0116
	2	0.4390	0.4369	0.3244	0.3315	-0.0021	0.0071
40.82	1	0.0538	0.0572	0.5321	0.5216	0.0034	-0.0105
	2	0.5566	0.5532	0.3956	0.4094	-0.0034	0.0138
40.82	1	0.0539	0.0572	0.5303	0.5211	0.0033	-0.0092
	2	0.5575	0.5542	0.4012	0.4100	-0.0033	0.0088
40.82	1	0.0667	0.0708	0.3714	0.3636	0.0041	-0.0078
	2	0.8106	0.8065	0.5952	0.6020	-0.0041	0.0068
54.43	1	0.0583	0.0586	0.8509	0.8467	0.0003	-0.0042
	2	0.0962	0.0959	0.0668	0.0691	-0.0003	0.0023
54.43	1	0.0686	0.0651	0.7642	0.7533	-0.0035	-0.0109
	2	0.2386	0.2421	0.1576	0.1669	0.0035	0.0093
54.43	1	0.0790	0.0769	0.6581	0.6434	-0.0021	-0.0148
	2	0.4303	0.4324	0.2727	0.2853	0.0021	0.0127
54.43	1	0.0889	0.0891	0.5816	0.5653	0.0002	-0.0163
	2	0.5705	0.5703	0.3574	0.3728	-0.0002	0.0154
54.43	1	0.1039	0.1068	0.4922	0.4768	0.0029	-0.0154
	2	0.7124	0.7095	0.4634	0.4774	-0.0029	0.0140
54.43	1	0.1064	0.1127	0.4569	0.4509	0.0063	-0.0060
	2	0.7498	0.7435	0.5046	0.5094	-0.0063	0.0048
68.04	1	0.0664	0.0710	0.8917	0.8945	0.0046	0.0028
	2	0.0440	0.0394	0.0296	0.0258	-0.0046	-0.0038
68.04	1	0.0681	0.0732	0.8650	0.8684	0.0051	0.0034
	2	0.0861	0.0810	0.0565	0.0525	-0.0051	-0.0040
68.04	1	0.0784	0.0771	0.8290	0.8283	-0.0013	-0.0007
	2	0.1459	0.1472	0.0947	0.0938	0.0013	-0.0009
68.04	1	0.0771	0.0810	0.7967	0.7950	0.0039	-0.0017
	2	0.2078	0.2039	0.1243	0.1283	-0.0039	0.0040
68.04	1	0.0873	0.0876	0.7593	0.7434	0.0003	-0.0119
	2	0.2884	0.2881	0.1678	0.1783	-0.0003	0.0105

Table B-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-CO(2)-CH_4(3) at 123.10 K (C_{12}=-0.12702, C_{13}=-0.12819, C_{23}=0.03119)							
			$\lvert\Delta x\rvert_{av}$ = 0.0033		$\lvert\Delta y\rvert_{av}$ = 0.0074		
68.04	1	0.0990	0.0965	0.7074	0.6960	-0.0025	-0.0114
	2	0.3787	0.3812	0.2226	0.2328	0.0025	0.0103
68.04	1	0.1217	0.1197	0.6095	0.5988	-0.0020	-0.0107
	2	0.5503	0.5523	0.3303	0.3396	0.0020	0.0093
68.04	1	0.1470	0.1450	0.5134	0.5202	-0.0020	0.0068
	2	0.6633	0.6653	0.4327	0.4304	0.0020	-0.0023
68.04	1	0.1734	0.1694	0.4523	0.4577	-0.0040	0.0054
	2	0.7240	0.7280	0.5079	0.5064	0.0040	-0.0015
68.04	1	0.1708	0.1738	0.4390	0.4451	0.0030	0.0061
	2	0.7386	0.7356	0.5244	0.5212	-0.0030	-0.0032
68.04	1	0.1879	0.1898	0.3946	0.4104	0.0019	0.0158
	2	0.7568	0.7549	0.5788	0.5650	-0.0019	-0.0138
68.04	1	0.1840	0.1883	0.3985	0.4122	0.0043	0.0138
	2	0.7578	0.7535	0.5719	0.5622	-0.0043	-0.0097
102.06	1	0.1137	0.1121	0.8686	0.8682	-0.0016	-0.0004
	2	0.0987	0.1003	0.0548	0.0561	0.0016	0.0013
102.06	1	0.1167	0.1185	0.8350	0.8382	0.0018	0.0032
	2	0.1565	0.1547	0.0854	0.0862	-0.0018	-0.0008
102.06	1	0.1403	0.1394	0.7674	0.7609	-0.0009	-0.0065
	2	0.2939	0.2948	0.1521	0.1644	0.0009	0.0123
102.06	1	0.1854	0.1767	0.6729	0.6663	-0.0087	-0.0066
	2	0.4416	0.4503	0.2497	0.2618	0.0087	0.0121
102.06	1	0.2184	0.2132	0.5947	0.5988	-0.0052	0.0041
	2	0.5288	0.5340	0.3215	0.3333	0.0052	0.0118
102.06	1	0.2412	0.2303	0.5530	0.5718	-0.0109	0.0188
	2	0.5473	0.5582	0.3670	0.3625	0.0109	-0.0045
102.06	1	0.2918	0.2704	0.4637	0.5170	-0.0214	0.0533
	2	0.5657	0.5871	0.4516	0.4220	0.0214	-0.0296
System H_2(1)-CO(2)-CH_4(3) at 175.13 K (C_{12}=-0.10160, C_{13}=-0.11274, C_{23}=0.02993)							
			$\lvert\Delta x\rvert_{av}$ = 0.0060		$\lvert\Delta y\rvert_{av}$ = 0.0098		
27.22	1	0.0277	0.0263	0.8800	0.8689	-0.0014	-0.0111
	2	0.0878	0.0892	0.0574	0.0696	0.0014	0.0123
27.22	1	0.0309	0.0277	0.8111	0.7978	-0.0032	-0.0133
	2	0.1921	0.1953	0.1347	0.1444	0.0032	0.0097

Table B-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-CO(2)-CH_4(3) at 113.13 K (C_{12}=-0.10160, C_{13}=-0.11274, C_{23}=0.02993)			$\lvert\Delta x\rvert_{av}$ = 0.0060		$\lvert\Delta y\rvert_{av}$ = 0.0098		
27.22	1	0.0341	0.0376	0.5784	0.5566	0.0035	-0.0218
	2	0.6601	0.6566	0.3870	0.4084	-0.0035	0.0214
27.22	1	0.0395	0.0392	0.5425	0.5295	-0.0003	-0.0130
	2	0.7122	0.7125	0.4238	0.4398	0.0003	0.0160
40.82	1	0.0404	0.0419	0.8725	0.8668	0.0015	-0.0057
	2	0.1463	0.1448	0.0794	0.0846	-0.0015	0.0052
40.82	1	0.0440	0.0459	0.8225	0.8095	0.0019	-0.0130
	2	0.2628	0.2609	0.1325	0.1450	-0.0019	0.0125
40.82	1	0.0424	0.0459	0.8221	0.8100	0.0035	-0.0121
	2	0.2633	0.2598	0.1327	0.1444	-0.0035	0.0117
40.82	1	0.0698	0.0662	0.6696	0.6470	-0.0036	-0.0226
	2	0.6406	0.6442	0.2946	0.3232	0.0036	0.0286
40.82	1	0.0729	0.0708	0.6264	0.6207	-0.0021	-0.0057
	2	0.7024	0.7045	0.3397	0.3537	0.0021	0.0140
54.43	1	0.0485	0.0559	0.8815	0.8883	0.0074	0.0068
	2	0.1408	0.1334	0.0752	0.0673	-0.0074	-0.0079
54.43	1	0.0953	0.0916	0.7080	0.6993	-0.0037	-0.0087
	2	0.5967	0.6004	0.2615	0.2707	0.0037	0.0092
54.43	1	0.1074	0.1033	0.0712	0.6607	-0.0041	-0.0105
	2	0.6857	0.6898	0.3031	0.3151	0.0041	0.0120
68.04	1	0.0666	0.0657	0.9268	0.9266	-0.0009	-0.0002
	2	0.0628	0.0637	0.0288	0.0300	0.0009	0.0012
68.04	1	0.0819	0.0756	0.8723	0.8639	-0.0063	-0.0084
	2	0.2048	0.2111	0.0870	0.0947	0.0063	0.0077
68.04	1	0.1259	0.1166	0.7289	0.7246	-0.0093	-0.0043
	2	0.5618	0.5711	0.2391	0.2444	0.0093	0.0053
58.04	1	0.1467	0.1364	0.6903	0.6791	-0.0103	-0.0112
	2	0.6602	0.6705	0.2842	0.2964	0.0103	0.0122
102.06	1	0.0989	0.0999	0.9093	0.9176	0.0010	0.0083
	2	0.0952	0.0942	0.0460	0.0392	-0.0010	-0.0068
102.06	1	0.1113	0.1125	0.8588	0.8747	0.0012	0.0159
	2	0.2031	0.2019	0.0960	0.0826	-0.0012	-0.0134
102.06	1	0.1651	0.1573	0.7467	0.7766	-0.0078	0.0299
	2	0.4396	0.4474	0.2143	0.1845	0.0078	-0.0298
102.06	1	0.1858	0.1838	0.7211	0.7352	-0.0120	0.0141
	2	0.5212	0.5332	0.2410	0.2291	0.0120	-0.0119

Table B-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-CO(2)-CH_4(3) at 113.13 K (C_{12}=-0.10160, C_{13}=-0.11274, C_{23}=0.02993)			$\lvert\Delta x\rvert_{av}$ = 0.0060		$\lvert\Delta y\rvert_{av}$ = 0.0098		
102.06	1	0.3075	0.2775	0.5663	0.6129	-0.0300	0.0467
	2	0.6285	0.6585	0.4048	0.3686	0.0300	-0.0362
System H_2(1)-CO(2)-CH_4(3) at 108.12 K (C_{12}=-0.08883, C_{13}=-0.10498, C_{23}=0.02930)			$\lvert\Delta x\rvert_{av}$ = 0.0060		$\lvert\Delta y\rvert_{av}$ = 0.0148		
27.22	1	0.0366	0.0355	0.7457	0.7062	-0.0011	-0.0395
	2	0.5197	0.5208	0.2242	0.2646	0.0011	0.0404
27.22	1	0.0492	0.0477	0.6444	0.5713	-0.0015	-0.0731
	2	0.8590	0.8605	0.3372	0.4190	0.0015	0.0818
40.82	1	0.0474	0.0559	0.7836	0.7763	0.0085	-0.0073
	2	0.4910	0.4825	0.1902	0.1978	-0.0085	0.0076
40.82	1	0.0757	0.0826	0.6873	0.6560	0.0069	-0.0313
	2	0.8336	0.8267	0.2997	0.3351	-0.0069	0.0354
54.43	1	0.0707	0.0764	0.8129	0.8036	0.0057	-0.0093
	2	0.4750	0.4693	0.1633	0.1717	-0.0057	0.0084
54.43	1	0.0864	0.0940	0.7520	0.7521	0.0076	0.0001
	2	0.6430	0.6354	0.2245	0.2288	-0.0076	0.0043
54.43	1	0.1147	0.1170	0.7161	0.6398	0.0023	-0.0223
	2	0.7915	0.7892	0.2730	0.2968	-0.0023	0.0238
68.04	1	0.0804	0.0939	0.8181	0.8235	0.0135	0.0054
	2	0.4498	0.4363	0.1543	0.1511	-0.0135	-0.0032
68.04	1	0.1067	0.1183	0.7644	0.7707	0.0116	0.0063
	2	0.6175	0.6059	0.2121	0.2092	-0.0116	-0.0029
68.04	1	0.1403	0.1520	0.7160	0.7091	0.0117	-0.0069
	2	0.7676	0.7559	0.2730	0.2807	-0.0117	0.0077

Table B-2

Comparison of Experimental and Calculated VLE Compositions for Three Hydrogen Containing Ternary Systems (100), (Correlated C_{ij} Obtained from Equation (III-1), ICL Equation)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System $H_2(1)$-$N_2(2)$-CO(3) at 103.15 K (C_{12}=-0.06768, C_{13}=-0.07616, C_{23}=-0.00878)			$\|\Delta x\|_{av}$ = 0.0187		$\|\Delta y\|_{av}$ = 0.0129		
75.00	1	0.2210	0.2165	0.6350	0.6667	-0.0045	0.0317
	2	0.6000	0.6045	0.2910	0.2642	0.0045	-0.0027
75.00	1	0.2280	0.2096	0.6730	0.6842	-0.0184	0.0112
	2	0.4360	0.4544	0.2000	0.1927	0.0184	-0.0073
75.00	1	0.2500	0.2050	0.6810	0.6960	-0.0045	0.0150
	2	0.3170	0.3620	0.1550	0.1506	0.0045	-0.0044
System $H_2(1)$-$N_2(2)$-CO(3) at 77.35 K (C_{12}=-0.03071, C_{13}=-0.01038, C_{23}=0.00631)			$\|\Delta x\|_{av}$ = 0.0076		$\|\Delta y\|_{av}$ = 0.0115		
75.00	1	0.1380	0.1488	0.9380	0.9432	0.0108	0.0052
	2	0.3790	0.3682	0.0323	0.0300	-0.0108	-0.0023
75.00	1	0.1500	0.1633	0.9283	0.9353	0.0133	0.0070
	2	0.6350	0.6217	0.0581	0.0518	-0.0133	-0.0063
75.00	1	0.1476	0.1697	0.9224	0.9319	0.0221	0.0095
	2	0.7580	0.7359	0.0687	0.0622	-0.0221	-0.0065
100.00	1	0.1930	0.1924	0.9227	0.9334	-0.0006	0.0107
	2	0.3470	0.3476	0.0403	0.0345	0.0006	-0.0058
100.00	1	0.2130	0.2131	0.9007	0.9232	0.0001	0.0225
	2	0.5940	0.5939	0.0768	0.0618	-0.0001	-0.0150
100.00	1	0.2126	0.2207	0.9024	0.9194	0.0081	0.0171
	2	0.6930	0.6849	0.0873	0.0729	-0.0081	-0.0144
125.00	1	0.2190	0.2338	0.9052	0.9211	0.0148	0.0159
	2	0.3430	0.3282	0.0461	0.0402	-0.0148	-0.0059
125.00	1	0.2530	0.2597	0.8811	0.9086	0.0067	0.0275
	2	0.5560	0.5493	0.0928	0.0722	-0.0067	-0.0206
125.00	1	0.2654	0.2709	0.8820	0.9032	0.0055	0.0212
	2	0.6480	0.6425	0.1060	0.0876	-0.0055	-0.0184
150.00	1	0.2550	0.2732	0.8799	0.9073	0.0182	0.0274
	2	0.3240	0.3058	0.0584	0.0459	-0.0182	-0.0125
150.00	1	0.3250	0.3073	0.8639	0.8905	-0.0177	0.0266
	2	0.5040	0.5217	0.1070	0.0871	0.0177	-0.0199

Table B-2 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-N_2(2)-CH_4(3) at 103.15 K (C_{12}=-0.06768, C_{13}=-0.09728, C_{23}=-0.04110)			$\|\Delta x\|_{av}$ = 0.0130		$\|\Delta y\|_{av}$ = 0.0054		
75.00	1	0.1430	0.1156	0.8030	0.7970	-0.0274	-0.0060
	2	0.4670	0.4944	0.1820	0.1832	0.0274	0.0012
75.00	1	0.0830	0.0730	0.9130	0.9046	-0.0100	-0.0084
	2	0.1590	0.1690	0.0670	0.0733	0.0100	0.0063
75.00	1	0.0970	0.0788	0.8970	0.8817	-0.0182	0.0153
	2	0.2120	0.2302	0.0850	0.0963	0.0182	0.0114
75.00	1	0.1360	0.1035	0.8220	0.8181	-0.0325	-0.0039
	2	0.3930	0.4255	0.1520	0.1613	0.0325	0.0093
100.00	1	0.1820	0.1530	0.8070	0.8016	-0.0290	-0.0054
	2	0.4390	0.4680	0.1710	0.1753	0.0290	0.0043
100.00	1	0.1080	0.1004	0.8950	0.8937	-0.0076	-0.0013
	2	0.1950	0.2026	0.0810	0.0824	0.0076	0.0014
100.00	1	0.1620	0.1370	0.8360	0.8227	-0.0250	-0.0133
	2	0.3820	0.4070	0.1420	0.1537	0.0250	0.0117
125.00	1	0.1530	0.1232	0.8900	0.8929	-0.0299	0.0029
	2	0.1680	0.1978	0.0780	0.0804	0.0299	0.0024
125.00	1	0.1340	0.1236	0.8910	0.8919	-0.0104	0.0009
	2	0.1900	0.2004	0.0800	0.0813	0.0104	0.0013
125.00	1	0.2460	0.1985	0.8020	0.7873	-0.0475	-0.0147
	2	0.4190	0.4665	0.1730	0.1852	0.0475	0.0122
150.00	1	0.1350	0.1366	0.8990	0.9066	0.0016	0.0076
	2	0.1550	0.1534	0.0670	0.0639	-0.0016	-0.0031
System H_2(1)-N_2(2)-CH_4(3) at 93.15 K (C_{12}=-0.05335, C_{13}=-0.08178, C_{23}=0.03587)			$\|\Delta x\|_{av}$ = 0.0108		$\|\Delta y\|_{av}$ = 0.0143		
100.00	1	0.1107	0.1042	0.8965	0.9096	-0.0065	0.0131
	2	0.2993	0.3058	0.0870	0.0792	0.0065	-0.0078
100.00	1	0.2296	0.1880	0.8350	0.8416	-0.0416	0.0066
	2	0.5737	0.6153	0.1628	0.1499	0.0416	-0.0129
125.00	1	0.2598	0.2298	0.8120	0.8301	-0.0300	0.0181
	2	0.5468	0.5768	0.1761	0.1586	0.0300	-0.0175
150.00	1	0.0694	0.1134	0.9470	0.9458	0.0440	-0.0012
	2	0.1711	0.1271	0.0326	0.0389	-0.0440	0.0063
150.00	1	0.1295	0.1471	0.8668	0.9002	0.0176	0.0334
	2	0.3061	0.2885	0.1068	0.0837	-0.0176	-0.0231

Table B-2 (continued)

Pressure		x		y			
atm	Comp.	Expl	Calc	Expl	Calc	Δx	Δy
System $H_2(1)$-$N_2(2)$-$CH_4(3)$ at 93.15 K (C_{12}=-0.05335, C_{13}=-0.08178, C_{23}=0.03587)							
		$\lvert\Delta x\rvert_{av}$ = 0.0108		$\lvert\Delta y\rvert_{av}$ = 0.0143			
150.00	1	0.1836	0.1771	0.8286	0.8737	-0.0065	0.0451
	2	0.3792	0.3857	0.1359	0.1100	0.0065	-0.0259
System $H_2(1)$-$N_2(2)$-$CH_4(3)$ at 77.35 K (C_{12}=-0.03071, C_{13}=-0.05730, C_{23}=0.02761)							
		$\lvert\Delta x\rvert_{av}$ = 0.0115		$\lvert\Delta y\rvert_{av}$ = 0.0047			
75.00	1	0.0610	0.0863	0.9505	0.9557	0.0253	0.0052
	2	0.5010	0.4757	0.0495	0.0425	-0.0253	-0.0070
75.00	1	0.1020	0.1148	0.9430	0.9473	0.0128	0.0043
	2	0.6390	0.6262	0.0570	0.0512	-0.0128	-0.0058
75.00	1	0.0940	0.0775	0.9620	0.9589	-0.0165	-0.0031
	2	0.4010	0.4175	0.0380	0.0392	0.0165	0.0012
100.00	1	0.0780	0.1080	0.9450	0.9498	0.0301	0.0048
	2	0.4870	0.4570	0.0550	0.0477	-0.0301	-0.0073
125.00	1	0.1340	0.1303	0.9380	0.9418	-0.0038	0.0038
	2	0.4470	0.4508	0.0620	0.0547	0.0038	-0.0073
125.00	1	0.2010	0.1799	0.9260	0.9283	-0.0211	0.0023
	2	0.5690	0.5901	0.0740	0.0687	0.0211	-0.0053
125.00	1	0.1190	0.1152	0.9460	0.9469	-0.0038	0.0009
	2	0.3880	0.3918	0.0500	0.0495	0.0038	-0.0005
125.00	1	0.1020	0.0993	0.9490	0.9537	-0.0027	0.0047
	2	0.3150	0.3177	0.0470	0.0426	0.0027	-0.0044
150.00	1	0.2390	0.2107	0.9030	0.9165	-0.0283	0.0135
	2	0.5450	0.5733	0.0910	0.0795	0.0283	-0.0115
150.00	1	0.1160	0.1287	0.9305	0.9411	0.0127	0.0106
	2	0.3840	0.3713	0.0630	0.0541	-0.0127	-0.0089
150.00	1	0.0750	0.1063	0.9400	0.9517	0.0313	0.0117
	2	0.3090	0.2777	0.0540	0.0434	-0.0313	-0.0106
System $H_2(1)$-$CO(2)$-$CH_4(3)$ at 103.15 K (C_{12}=-0.07616, C_{13}=-0.09728, C_{23}=0.02778)							
		$\lvert\Delta x\rvert_{av}$ = 0.0145		$\lvert\Delta y\rvert_{av}$ = 0.0066			
75.00	1	0.0810	0.0843	0.9030	0.8953	0.0033	-0.0077
	2	0.3070	0.3037	0.0800	0.0851	-0.0033	0.0051
75.00	1	0.1390	0.1158	0.8490	0.8384	-0.0232	-0.0106
	2	0.5220	0.5452	0.1360	0.1461	0.0232	0.0101
100.00	1	0.1100	0.1105	0.9070	0.8977	0.0005	-0.0093
	2	0.2940	0.2935	0.0730	0.0809	-0.0005	0.0079

Table B-2 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System $H_2(1)$-CO(2)-$CH_4(3)$ at 103.15 K (C_{12}=-0.07616, C_{13}=-0.09728, C_{23}=0.02778)			$\lvert\Delta x\rvert_{av}$ = 0.0145		$\lvert\Delta y\rvert_{av}$ = 0.0066		
100.00	1	0.1450	0.1482	0.8410	0.8458	0.0032	0.0048
	2	0.5010	0.4978	0.1400	0.1359	-0.0032	-0.0041
100.00	1	0.1080	0.0957	0.9300	0.9274	-0.0123	-0.0026
	2	0.1650	0.1773	0.0480	0.0503	0.0123	0.0023
125.00	1	0.1080	0.1351	0.8790	0.8962	0.0271	0.0172
	2	0.3100	0.2829	0.0880	0.0798	-0.0271	-0.0082
125.00	1	0.1910	0.1850	0.8340	0.8390	-0.0060	0.0050
	2	0.4750	0.4810	0.1410	0.1396	0.0060	-0.0014
125.00	1	0.1350	0.1582	0.8610	0.8930	0.0232	0.0320
	2	0.2950	0.2718	0.0950	0.0798	-0.0232	-0.0152
150.00	1	0.2830	0.2290	0.8210	0.8216	-0.0540	0.0006
	2	0.4250	0.4790	0.1490	0.1533	0.0540	0.0043
150.00	1	0.1220	0.1346	0.9230	0.9276	0.0126	0.0046
	2	0.1660	0.1534	0.0470	0.0450	-0.0126	-0.0020
System $H_2(1)$-CO(2)-$CH_4(3)$ at 93.15 K (C_{12}=-0.05066, C_{13}=-0.08178, C_{23}=0.02624)			$\lvert\Delta x\rvert_{av}$ = 0.0339		$\lvert\Delta y\rvert_{av}$ = 0.0084		
125.00	1	0.0620	0.0857	0.9862	0.9790	0.0238	-0.0072
	2	0.0644	0.0406	0.0	0.0083	-0.0238	0.0083
125.00	1	0.0569	0.0917	0.9595	0.9685	0.0348	0.0090
	2	0.1295	0.0947	0.0262	0.0189	-0.0348	-0.0073
125.00	1	0.1396	0.1816	0.8703	0.8893	0.0420	0.0190
	2	0.5978	0.5558	0.1170	0.1016	-0.0420	-0.0154
150.00	1	0.0952	0.1215	0.9283	0.9471	0.0263	0.0188
	2	0.2175	0.1912	0.0512	0.0387	-0.0263	-0.0125
150.00	1	0.2424	0.1545	0.9207	0.9170	-0.0879	-0.0037
	2	0.2663	0.3542	0.0614	0.0694	0.0879	0.0080
System $H_2(1)$-CO(2)-$CH_4(3)$ at 77.35 K (C_{12}=-0.01038, C_{13}=-0.05730, C_{23}=0.02383)			$\lvert\Delta x\rvert_{av}$ = 0.0108		$\lvert\Delta y\rvert_{av}$ = 0.0047		
75.00	1	0.1082	0.0944	0.9700	0.9664	-0.0139	-0.0036
	2	0.6270	0.6408	0.0300	0.0323	0.0139	0.0023
75.00	1	0.1120	0.0898	0.9710	0.9677	-0.0222	-0.0033
	2	0.5830	0.6052	0.0290	0.0309	0.0222	0.0019
100.00	1	0.1450	0.1198	0.9610	0.9614	-0.0252	0.0004
	2	0.5960	0.6212	0.0390	0.0369	0.0252	-0.0021

Table B-2 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-CO(2)-CH_4(3) at 77.35 K (C_{12}=-0.01038, C_{13}=-0.05730, C_{23}=0.02383)			$\lvert\Delta x\rvert_{av}$ = 0.0108		$\lvert\Delta y\rvert_{av}$ = 0.0047		
100.00	1	0.1190	0.1123	0.9690	0.9634	-0.0067	-0.0056
	2	0.5710	0.5777	0.0310	0.0347	0.0067	0.0037
125.00	1	0.1560	0.1365	0.9590	0.9572	-0.0195	-0.0018
	2	0.5540	0.5735	0.0410	0.0403	0.0195	-0.0007
125.00	1	0.1110	0.1396	0.9530	0.9564	0.0286	0.0034
	2	0.6160	0.5874	0.0470	0.0412	-0.0286	-0.0058
125.00	1	0.1310	0.0786	0.9500	0.9765	-0.0524	0.0265
	2	0.1730	0.2254	0.0500	0.0200	0.0524	-0.0300
150.00	1	0.1790	0.1843	0.9390	0.9440	0.0053	0.0050
	2	0.6540	0.6487	0.0610	0.0535	-0.0053	-0.0075
150.00	1	0.1770	0.1644	0.9450	0.9493	-0.0126	0.0043
	2	0.5720	0.5846	0.0550	0.0476	0.0126	-0.0074
150.00	1	0.1440	0.1542	0.9520	0.9519	0.0102	-0.0001
	2	0.5580	0.5478	0.0480	0.0448	-0.0102	-0.0032

Table B-3

Comparison of Experimental and Calculated VLE Compositions for Three Hydrogen Containing Ternary Systems (100), (Modified SRK Equation)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System $H_2(1)$-$N_2(2)$-CO(3) at 103.15 K (C_{12} = 0.0, C_{13} = 0.0, C_{23} = 0.046)							
		$\lvert\Delta x\rvert_{av}$ = 0.0115		$\lvert\Delta y\rvert_{av}$ = 0.0139			
75.00	1	0.2280	0.2598	0.6730	0.6893	0.0318	0.0163
	2	0.4360	0.4042	0.2000	0.1803	-0.0318	-0.0197
75.00	1	0.2500	0.2528	0.6810	0.7030	0.0028	0.0220
	2	0.3170	0.3142	0.1550	0.1390	-0.0028	-0.0160
System $H_2(1)$-$N_2(2)$-CO(3) at 77.35 K (C_{12} = 0.0, C_{13} = 0.0, C_{23} = 0.046)							
		$\lvert\Delta x\rvert_{av}$ = 0.0642		$\lvert\Delta y\rvert_{av}$ = 0.0056			
75.00	1	0.1380	0.2329	0.9380	0.9412	0.0949	0.0032
	2	0.3790	0.2841	0.0323	0.0283	-0.0949	-0.0040
75.00	1	0.1500	0.2471	0.9283	0.9348	0.0971	0.0065
	2	0.6350	0.5379	0.0581	0.0484	-0.0971	-0.0097
75.00	1	0.1476	0.2446	0.9224	0.9341	0.0970	0.0117
	2	0.7580	0.6610	0.0687	0.0577	0.0970	-0.0110
System $H_2(1)$-$N_2(2)$-$CH_4(3)$ at 103.15 K (C_{12} = 0.0, C_{13} = 0.0, C_{23} = 0.0319)							
		$\lvert\Delta x\rvert_{av}$ = 0.0070		$\lvert\Delta y\rvert_{av}$ = 0.0066			
75.00	1	0.1430	0.1318	0.8030	0.8153	-0.0112	0.0123
	2	0.4670	0.4782	0.1820	0.1685	0.0112	-0.0135
75.00	1	0.0830	0.0802	0.9130	0.9138	-0.0028	0.0008
	2	0.1590	0.1618	0.0670	0.0679	0.0028	0.0009
75.00	1	0.0970	0.0873	0.8970	0.8925	-0.0097	-0.0045
	2	0.2120	0.2217	0.0850	0.0894	0.0097	0.0044
75.00	1	0.1360	0.1173	0.8220	0.8343	-0.0187	0.0123
	2	0.3930	0.4117	0.1520	0.1487	0.0187	-0.0033
100.00	1	0.1820	0.1733	0.8070	0.8210	-0.0087	0.0140
	2	0.4390	0.4477	0.1710	0.1600	0.0087	-0.0110
100.00	1	0.1080	0.1103	0.8950	0.9043	0.0023	0.0093
	2	0.1950	0.1927	0.0810	0.0760	-0.0023	-0.0050
100.00	1	0.1620	0.1542	0.8360	0.8398	-0.0078	0.0038
	2	0.3820	0.3898	0.1420	0.1408	0.0078	-0.0012
125.00	1	0.1530	0.1344	0.8900	0.9039	-0.0186	0.0139
	2	0.1680	0.1866	0.0780	0.0740	0.0186	-0.0040
125.00	1	0.1340	0.1349	0.8910	0.9030	0.0009	0.0120
	2	0.1900	0.1891	0.0800	0.0749	-0.0009	-0.0051

Table B-3 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System $H_2(1)$-$N_2(2)$-$CH_4(3)$ at 103.15 K (C_{12} = 0.0, C_{13} = 0.0, C_{23} = 0.0319)							
		$\lvert\Delta x\rvert_{av}$ = 0.0070		$\lvert\Delta y\rvert_{av}$ = 0.0066			
150.00	1	0.1836	0.2046	0.8286	0.8798	0.0210	0.0512
	2	0.3792	0.3582	0.1359	0.1056	-0.0210	-0.0303
150.00	1	0.4226	0.3985	0.7164	0.7907	-0.0241	0.0743
	2	0.4823	0.5064	0.2562	0.1965	0.0241	-0.0597
System $H_2(1)$-$N_2(2)$-$CH_4(3)$ at 77.35 K (C_{12} = 0.0, C_{13} = 0.0, C_{23} = 0.0319)							
		$\lvert\Delta x\rvert_{av}$ = 0.0240		$\lvert\Delta y\rvert_{av}$ = 0.0038			
75.00	1	0.0610	0.1154	0.9505	0.9591	0.0544	0.0086
	2	0.5010	0.4466	0.0495	0.0396	-0.0544	-0.0099
75.00	1	0.1020	0.1545	0.9430	0.9517	0.0525	0.0087
	2	0.6390	0.5865	0.0570	0.0471	-0.0525	-0.0099
75.00	1	0.0940	0.1035	0.9620	0.9619	0.0095	-0.0001
	2	0.4010	0.3915	0.0380	0.0366	-0.0095	-0.0014
125.00	1	0.1340	0.1689	0.9380	0.9409	0.0349	0.0029
	2	0.4470	0.4121	0.0620	0.0559	-0.0349	-0.0061
125.00	1	0.2010	0.2365	0.9260	0.9274	0.0355	0.0014
	2	0.5690	0.5335	0.0740	0.0697	-0.0355	-0.0043
125.00	1	0.1190	0.1489	0.9460	0.9462	0.0299	0.0002
	2	0.3880	0.3581	0.0500	0.0506	-0.0299	0.0006
125.00	1	0.1020	0.1280	0.9490	0.9533	0.0260	0.0043
	2	0.3150	0.2890	0.0470	0.0434	-0.0260	-0.0036
150.00	1	0.2390	0.2734	0.9030	0.9108	0.0344	0.0078
	2	0.5450	0.5106	0.0910	0.0848	-0.0344	-0.0062
150.00	1	0.1160	0.1636	0.9305	0.9377	0.0476	0.0072
	2	0.3840	0.3364	0.0630	0.0577	-0.0476	-0.0053
System $H_2(1)$-$CO(2)$-$CH_4(3)$ at 103.15 K (C_{12} = 0.0, C_{13} = 0.0, C_{23} = 0.0300)							
		$\lvert\Delta x\rvert_{av}$ = 0.0140		$\lvert\Delta y\rvert_{av}$ = 0.0095			
75.00	1	0.0810	0.0950	0.9030	0.9024	0.0140	-0.0006
	2	0.3070	0.2930	0.0800	0.0812	-0.0140	0.0012
75.00	1	0.1390	0.1343	0.8490	0.8522	-0.0047	0.0032
	2	0.5220	0.5267	0.1360	0.1346	0.0047	-0.0014
100.00	1	0.1100	0.1237	0.9070	0.9055	0.0137	-0.0015
	2	0.2940	0.2803	0.0730	0.0766	-0.0137	0.0036

Table B-3 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System H_2(1)-CO(2)-CH_4(3) at 103.15 K (C_{12} = 0.0, C_{13} = 0.0, C_{23} = 0.0300)							
		$\lvert\Delta x\rvert_{av}$ = 0.0140		$\lvert\Delta y\rvert_{av}$ = 0.0095			
100.00	1	0.1450	0.1706	0.8410	0.8596	0.0256	0.0186
	2	0.5010	0.4754	0.1400	0.1247	-0.0256	-0.0153
100.00	1	0.1080	0.1051	0.9300	0.9333	-0.0029	0.0033
	2	0.1650	0.1679	0.0480	0.0482	0.0029	0.0002
125.00	1	0.1080	0.1504	0.8790	0.9042	0.0424	0.0252
	2	0.3100	0.2676	0.0880	0.0755	-0.0424	-0.0125
125.00	1	0.1940	0.2118	0.8340	0.8532	0.0208	0.0192
	2	0.4750	0.4542	0.1410	0.1281	-0.0208	-0.0129
150.00	1	0.1350	0.1749	0.8610	0.9009	0.0399	0.0399
	2	0.2950	0.2551	0.0950	0.0758	-0.0399	-0.0192
150.00	1	0.2830	0.2611	0.8210	0.8381	-0.0219	0.0171
	2	0.4250	0.4469	0.1490	0.1398	0.0219	-0.0092
150.00	1	0.1220	0.1457	0.9230	0.9340	0.0237	0.0010
	2	0.1660	0.1423	0.0470	0.0431	-0.0237	-0.0039
System H_2(1)-CO(2)-CH_4(3) at 93.15 K (C_{12} = 0.0, C_{13} = 0.0, C_{23} = 0.03)							
		$\lvert\Delta x\rvert_{av}$ = 0.0306		$\lvert\Delta y\rvert_{av}$ = 0.0078			
125.00	1	0.0619	0.0951	0.9862	0.9828	0.0332	-0.0034
	2	0.0644	0.0312	0.0	0.0068	-0.0332	0.0068
125.00	1	0.0569	0.1028	0.9595	0.9719	0.0459	0.0124
	2	0.1295	0.0836	0.0262	0.0178	-0.0459	-0.0084
150.00	1	0.0952	0.1377	0.9283	0.9499	0.0425	0.0216
	2	0.2175	0.1750	0.0512	0.0379	-0.0425	-0.0133
150.00	1	0.2424	0.1806	0.9207	0.9205	-0.0618	-0.0002
	2	0.2663	0.3281	0.0614	0.0675	0.0618	0.0061
System H_2(1)-CO(2)-CH_4(3) at 77.35 K (C_{12} = 0.0, C_{13} = 0.0, C_{23} = 0.03)							
		$\lvert\Delta x\rvert_{av}$ = 0.0244		$\lvert\Delta y\rvert_{av}$ = 0.0045			
75.00	1	0.1082	0.1359	0.9700	0.9683	0.0277	-0.0017
	2	0.6270	0.5993	0.0300	0.0307	-0.0277	0.0007
75.00	1	0.1120	0.1285	0.9710	0.9695	0.0165	-0.0015
	2	0.5830	0.5665	0.0290	0.0294	-0.0165	0.0004
100.00	1	0.1450	0.1709	0.9610	0.9615	0.0259	0.0005
	2	0.5960	0.5701	0.0390	0.0371	-0.0259	-0.0019

Table B-3 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
System $H_2(1)$-$CO(2)$-$CH_4(3)$ at 77.35 K (C_{12} = 0.0, C_{13} = 0.0, C_{23} = 0.03)			$\lvert\Delta x\rvert_{av}$ = 0.0244		$\lvert\Delta y\rvert_{av}$ = 0.0045		
100.00	1	0.1190	0.1585	0.9690	0.9635	0.0395	-0.0055
	2	0.5710	0.5315	0.0310	0.0349	-0.0395	0.0039
125.00	1	0.1560	0.1909	0.9590	0.9546	0.0349	-0.0044
	2	0.5540	0.5191	0.0410	0.0430	-0.0349	0.0020
125.00	1	0.1310	0.1032	0.9500	0.9765	-0.0278	0.0265
	2	0.1730	0.2008	0.0500	0.0205	0.0278	-0.0295
150.00	1	0.1770	0.2289	0.9450	0.9430	0.0519	-0.0020
	2	0.5720	0.5201	0.0550	0.0537	-0.0519	0.0013
150.00	1	0.1440	0.2122	0.9520	0.9462	0.0682	-0.0058
	2	0.5580	0.4898	0.0480	0.0504	-0.0682	0.0024

APPENDIX C

EXPERIMENTAL AND CALCULATED VLE VALUES FOR THE QUATERNARY SYSTEM H_2-N_2-CO-CH_4

The ICL equation, the modified SRK equation and the modified BWR equation had been used to calculate the vapor and liquid compositions by fixed system temperature, pressure and liquid compositions of two least-volatile components.

APPENDIX C

Table C-1

Comparison of Experimental and Calculated VLE Compositions for the Quaternary System $H_2(1)$-$N_2(2)$-$CO(3)$-$CH_4(4)$ (100), (Correlated C_{ij} Obtained from Equation (III-1), ICL Equation)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
			103.15 K (-170°C)				
			$\|\Delta x\|_{av}$ = 0.0165		$\|\Delta y\|_{av}$ = 0.0308		
75.00	1	0.1380	0.1048	0.8510	0.8407	-0.0332	-0.0103
	2	0.1150	0.1482	0.0440	0.0557	0.0332	0.0117
	3	0.3110	0.3110	0.0860	0.0855	0.0	-0.0005
75.00	1	0.1300	0.1219	0.8230	0.8063	-0.0081	-0.0167
	2	0.2600	0.2681	0.0890	0.0981	0.0081	0.0091
	3	0.2820	0.2820	0.0730	0.0787	0.0	0.0057
75.00	1	0.3050	0.1962	0.7530	0.7051	-0.1088	-0.0479
	2	0.2970	0.4058	0.1190	0.1645	0.1088	0.0455
	3	0.3700	0.3700	0.1280	0.1271	0.0	-0.0009
75.00	1	0.1520	0.1605	0.8040	0.7732	0.0085	-0.0308
	2	0.0730	0.0645	0.0260	0.0240	-0.0085	-0.0020
	3	0.6710	0.6710	0.1640	0.1949	0.0	0.0309
75.00	1	0.1540	0.1396	0.7930	0.7746	-0.0144	-0.0184
	2	0.3890	0.4034	0.1320	0.1475	0.0144	0.0155
	3	0.2150	0.2150	0.0610	0.0624	0.0	0.0014
100.00	1	0.3800	0.3819	0.6630	0.7160	0.0019	0.0530
	2	0.3980	0.3961	0.2280	0.1992	-0.0019	-0.0288
	3	0.2110	0.2110	0.1090	0.0968	0.0	-0.0122
100.00	1	0.1260	0.1287	0.8600	0.8592	0.0027	-0.0008
	2	0.1090	0.1063	0.0400	0.0400	-0.0027	0.0
	3	0.2880	0.2880	0.0790	0.0797	0.0	0.0007
100.00	1	0.2470	0.2227	0.7850	0.7623	-0.0243	-0.0227
	2	0.0680	0.0923	0.0250	0.0367	0.0243	0.0117
	3	0.5910	0.5910	0.1820	0.1911	0.0	0.0091
100.00	1	0.2070	0.1867	0.7900	0.7761	-0.0203	-0.0139
	2	0.3580	0.3783	0.1330	0.1433	0.0203	0.0103
	3	0.2000	0.2000	0.0590	0.0616	0.0	0.0026
125.00	1	0.1330	0.1525	0.8560	0.8640	0.0195	0.0080
	2	0.1120	0.0925	0.0430	0.0357	-0.0195	-0.0073
	3	0.2640	0.2640	0.0750	0.0759	0.0	0.0009

Table C-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		103.15 K (-170°C) $\lvert\Delta x\rvert_{av}$ = 0.0165 $\lvert\Delta y\rvert_{av}$ = 0.0308					
125.00	1	0.2260	0.2020	0.8000	0.8022	-0.0240	0.0022
	2	0.2130	0.2370	0.0880	0.0927	0.0240	0.0047
	3	0.2600	0.2600	0.0830	0.0813	0.0	-0.0017
125.00	1	0.1450	0.1459	0.8780	0.8758	0.0009	-0.0022
	2	0.0560	0.0551	0.0230	0.0213	-0.0009	-0.0017
	3	0.2760	0.2760	0.0770	0.0786	0.0	0.0016
125.00	1	0.2940	0.2798	0.7590	0.7493	-0.0142	-0.0097
	2	0.0620	0.0762	0.0290	0.0336	0.0142	0.0046
	3	0.5500	0.5500	0.1990	0.2036	0.0	0.0046
150.00	1	0.2040	0.1806	0.8580	0.8547	-0.0234	-0.0033
	2	0.0950	0.1184	0.0390	0.0474	0.0234	0.0084
	3	0.2260	0.2260	0.0720	0.0692	0.0	-0.0028
150.00	1	0.2410	0.2305	0.7880	0.8007	-0.0105	0.0127
	2	0.1980	0.2085	0.0900	0.0866	0.0105	-0.0034
	3	0.2490	0.2490	0.0880	0.0838	0.0	-0.0042
150.00	1	0.1580	0.1661	0.8700	0.8785	0.0081	0.0085
	2	0.0520	0.0439	0.0220	0.0175	-0.0081	-0.0045
	3	0.2560	0.2560	0.0740	0.0762	0.0	0.0062
150.00	1	0.4050	0.3532	0.6810	0.7090	-0.0518	0.0280
	2	0.0520	0.1038	0.0340	0.0552	0.0518	0.0212
	3	0.4620	0.4620	0.2570	0.2167	0.0	-0.0403
		93.15 K (-180°C) $\lvert\Delta x\rvert_{av}$ = 0.0337 $\lvert\Delta y\rvert_{av}$ = 0.1272					
150.00	1	0.3545	0.2792	0.7839	0.8213	-0.0753	0.0374
	2	0.2540	0.3293	0.1052	0.1031	0.0753	-0.0021
	3	0.2483	0.2483	0.0905	0.0639	0.0	-0.0027
150.00	1	0.2368	0.2340	0.8202	0.8539	-0.0028	0.0337
	2	0.2051	0.2079	0.0679	0.0594	0.0028	-0.0085
	3	0.3325	0.3325	0.0928	0.0742	0.0	-0.0186
150.00	1	0.1703	0.1793	0.8523	0.8800	0.0090	0.0277
	2	0.2829	0.2739	0.0913	0.0769	-0.0090	-0.0144
	3	0.1322	0.1322	0.0319	0.0279	0.0	-0.0040

Table C-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		93.15 K (-180°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0337		$\lvert\Delta y\rvert_{av}$ = 0.1272			
150.00	1	0.1415	0.1742	0.8566	0.8852	0.0327	0.0286
	2	0.2752	0.2425	0.0813	0.0680	-0.0327	-0.0133
	3	0.1519	0.1519	0.0389	0.0317	0.0	-0.0072
150.00	1	0.0705	0.1085	0.9500	0.9600	0.0381	0.0099
	2	0.0791	0.0411	0.0205	0.0126	-0.0381	-0.0079
	3	0.0619	0.0619	0.0090	0.0128	0.0	0.0038
150.00	1	0.1545	0.1608	0.8697	0.8887	0.0063	0.0190
	2	0.3207	0.3144	0.0997	0.0899	-0.0063	-0.0098
	3	0.0251	0.0251	0.0054	0.0053	0.0	-0.0001
150.00	1	0.1711	0.1625	0.8812	0.8961	-0.0086	0.0149
	2	0.1895	0.1981	0.0587	0.0556	0.0086	-0.0031
	3	0.1622	0.1622	0.0391	0.0333	0.0	-0.0058
150.00	1	0.1306	0.1514	0.8935	0.9100	0.0209	0.0165
	2	0.1394	0.1186	0.0386	0.0333	-0.0209	-0.0053
	3	0.2092	0.2092	0.0464	0.0421	0.0	-0.0043
150.00	1	0.0841	0.1321	0.9167	0.9305	0.0480	0.0138
	2	0.1161	0.0681	0.0332	0.0196	-0.0480	-0.0136
	3	0.1758	0.1758	0.0296	0.0353	0.0	0.0057
150.00	1	0.1577	0.1784	0.8505	0.8830	0.0207	0.0325
	2	0.2579	0.2372	0.0849	0.0663	-0.0207	-0.0186
	3	0.1710	0.1710	0.0396	0.0357	0.0	-0.0039
150.00	1	0.2479	0.2354	0.8007	0.8431	-0.0125	0.0424
	2	0.3562	0.3687	0.1269	0.1075	0.0125	-0.0194
	3	0.1505	0.1505	0.0507	0.0351	0.0	-0.0156
		77.35 K (-195.8°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0087		$\lvert\Delta y\rvert_{av}$ = 0.0081			
75.00	1	0.1880	0.1548	0.9590	0.9392	-0.0333	-0.0198
	2	0.4970	0.5303	0.0240	0.0434	0.0333	0.0194
	3	0.2990	0.2990	0.0170	0.0172	0.0	0.0002
75.00	1	0.1220	0.1470	0.9460	0.9438	0.0250	-0.0022
	2	0.3940	0.3690	0.0320	0.0299	-0.0250	-0.0021
	3	0.4740	0.4740	0.0220	0.0262	0.0	0.0042

Table C-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		77.35 K (-195.8°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0087		$\lvert\Delta y\rvert_{av}$ = 0.0081			
75.00	1	0.1300	0.1414	0.9430	0.9451	0.0114	0.0021
	2	0.4140	0.4026	0.0330	0.0323	-0.0114	-0.0067
	3	0.4060	0.4060	0.0240	0.0222	0.0	-0.0018
75.00	1	0.1620	0.1391	0.9390	0.9477	-0.0229	0.0087
	2	0.2550	0.2779	0.0260	0.0223	0.0229	-0.0037
	3	0.5570	0.5570	0.0350	0.0298	0.0	-0.0052
75.00	1	0.1310	0.1218	0.9470	0.9492	-0.0092	0.0022
	2	0.4520	0.4612	0.0370	0.0369	0.0092	-0.0001
	3	0.2380	0.2380	0.0160	0.0128	0.0	-0.0032
75.00	1	0.1300	0.1247	0.9430	0.9476	-0.0053	0.0046
	2	0.5050	0.5103	0.0440	0.0409	0.0053	-0.0031
	3	0.1920	0.1920	0.0130	0.0105	0.0	-0.0025
75.00	1	0.1190	0.1350	0.9360	0.9463	0.0160	0.0103
	2	0.4530	0.4370	0.0390	0.0349	-0.0160	-0.0041
	3	0.3330	0.3330	0.0250	0.0181	0.0	-0.0069
75.00	1	0.1660	0.1548	0.9270	0.9380	-0.0112	0.0110
	2	0.6060	0.6172	0.0580	0.0505	0.0112	-0.0075
	3	0.1900	0.1900	0.0150	0.0111	0.0	-0.0039
75.00	1	0.1390	0.1430	0.9390	0.9454	0.0040	0.0064
	2	0.3540	0.3500	0.0330	0.0282	-0.0040	-0.0048
	3	0.4810	0.4810	0.0280	0.0262	0.0	-0.0018
75.00	1	0.1250	0.1482	0.9330	0.9419	0.0233	0.0089
	2	0.5000	0.4768	0.0470	0.0386	-0.0233	-0.0084
	3	0.3410	0.3410	0.0200	0.0192	0.0	-0.0008
75.00	1	0.1230	0.1444	0.9450	0.9450	0.0215	0.0
	2	0.3620	0.3406	0.0310	0.0275	-0.0215	-0.0035
	3	0.5000	0.5000	0.0240	0.0273	0.0	0.0033
75.00	1	0.1030	0.1096	0.9520	0.9556	0.0066	0.0036
	2	0.2930	0.2864	0.0260	0.0231	-0.0066	-0.0029
	3	0.3880	0.3880	0.0220	0.0202	0.0	-0.0018
100.00	1	0.2160	0.1876	0.9270	0.9348	-0.0284	0.0078
	2	0.3210	0.3494	0.0380	0.0344	0.0284	-0.0036
	3	0.4450	0.4450	0.0350	0.0306	0.0	0.0044

Table C-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		77.35 K (-195.8°C)					
		$\|\Delta x\|_{av}$ = 0.0087		$\|\Delta y\|_{av}$ = 0.0081			
100.00	1	0.1770	0.1889	0.9240	0.9344	0.0119	0.0104
	2	0.3640	0.3521	0.0390	0.0347	-0.0119	-0.0043
	3	0.4450	0.4450	0.0370	0.0307	0.0	-0.0063
100.00	1	0.2000	0.1847	0.9230	0.9349	-0.0153	0.0119
	2	0.3790	0.3943	0.0420	0.0385	0.0153	-0.0035
	3	0.3790	0.3790	0.0350	0.0260	0.0	-0.0090
100.00	1	0.1720	0.1785	0.9370	0.9391	0.0065	0.0021
	2	0.2650	0.2585	0.0290	0.0250	-0.0065	-0.0040
	3	0.5370	0.5370	0.0340	0.0355	0.0	0.0015
100.00	1	0.1810	0.1566	0.9410	0.9407	-0.0244	-0.0003
	2	0.4290	0.4534	0.0440	0.0433	0.0024	-0.0007
	3	0.2180	0.2180	0.0150	0.0144	0.0	-0.0006
100.00	1	0.1550	0.1589	0.9330	0.9393	0.0039	0.0063
	2	0.4990	0.4952	0.0530	0.0474	-0.0039	-0.0056
	3	0.1740	0.1740	0.0140	0.0116	0.0	-0.0024
100.00	1	0.1700	0.1730	0.9330	0.9337	0.0030	0.0047
	2	0.4150	0.4120	0.0450	0.0396	-0.0030	-0.0054
	3	0.3230	0.3230	0.0220	0.0216	0.0	-0.0004
100.00	1	0.2100	0.1978	0.9150	0.9280	-0.0122	0.0130
	2	0.5630	0.5752	0.0690	0.0578	0.0122	-0.0112
	3	0.1840	0.1840	0.0160	0.0135	0.0	-0.0025
100.00	1	0.1590	0.1828	0.9250	0.9367	0.0238	0.0118
	2	0.3450	0.3212	0.0390	0.0313	-0.0238	-0.0077
	3	0.4680	0.4680	0.0360	0.0316	0.0	-0.0044
100.00	1	0.1950	0.1933	0.9210	0.9312	-0.0017	0.0102
	2	0.4630	0.4647	0.0510	0.0462	0.0017	-0.0048
	3	0.3120	0.3120	0.0280	0.0222	0.0	-0.0058
100.00	1	0.1570	0.1846	0.9240	0.9362	0.0277	0.0122
	2	0.3460	0.3184	0.0380	0.0311	-0.0277	-0.0069
	3	0.4770	0.4770	0.0380	0.0324	0.0	-0.0056
100.00	1	0.1440	0.1409	0.9430	0.9481	-0.0031	0.0051
	2	0.2880	0.2911	0.0320	0.0276	0.0031	-0.0044
	3	0.3600	0.3600	0.0250	0.0226	0.0	-0.0024

Table C-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		77.35 K (-195.8°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0087		$\lvert\Delta y\rvert_{av}$ = 0.0081			
125.00	1	0.2220	0.2417	0.8750	0.9161	0.0197	0.0411
	2	0.4750	0.4553	0.0880	0.0569	-0.0197	-0.0311
	3	0.2870	0.2870	0.0370	0.0266	0.0	-0.0104
125.00	1	0.2890	0.2342	0.8580	0.9203	-0.0548	0.0623
	2	0.3110	0.3658	0.0700	0.0448	0.0548	-0.0252
	3	0.3900	0.3900	0.0720	0.0347	0.0	-0.0373
125.00	1	0.2200	0.2216	0.9130	0.9240	0.0016	0.0111
	2	0.3700	0.3684	0.0540	0.0439	-0.0016	-0.0101
	3	0.3650	0.3650	0.0330	0.0312	0.0	-0.0018
125.00	1	0.2120	0.2200	0.9170	0.9256	0.0080	0.0086
	2	0.3250	0.3170	0.0440	0.0376	-0.0080	-0.0064
	3	0.4280	0.4280	0.0390	0.0361	0.0	-0.0029
125.00	1	0.2130	0.2166	0.9210	0.9281	0.0036	0.0071
	2	0.2570	0.2534	0.0340	0.0298	-0.0036	-0.0042
	3	0.5040	0.5040	0.0450	0.0416	0.0	-0.0034
125.00	1	0.1630	0.1821	0.9240	0.9334	0.0191	0.0094
	2	0.4170	0.3979	0.0550	0.0452	-0.0191	-0.0098
	3	0.2400	0.2400	0.0210	0.0191	0.0	-0.0019
125.00	1	0.1860	0.1885	0.9230	0.9300	0.0025	0.0070
	2	0.4710	0.4685	0.0610	0.0538	-0.0025	-0.0072
	3	0.1690	0.1690	0.0160	0.0138	0.0	-0.0022
125.00	1	0.2490	0.2100	0.9170	0.9265	-0.0390	0.0095
	2	0.3620	0.4010	0.0550	0.0468	0.0390	-0.0082
	3	0.3010	0.3010	0.0280	0.0251	0.0	-0.0029
125.00	1	0.2390	0.2403	0.8960	0.9146	0.0013	0.0186
	2	0.5500	0.5487	0.0820	0.0686	-0.0013	-0.0134
	3	0.1680	0.1680	0.0220	0.0158	0.0	-0.0062
125.00	1	0.1760	0.2182	0.9090	0.9273	0.0422	0.0183
	2	0.3110	0.2688	0.0470	0.0318	-0.0422	-0.0153
	3	0.4870	0.4870	0.0440	0.0405	0.0	-0.0003
125.00	1	0.2410	0.2362	0.9090	0.9180	-0.0048	0.0090
	2	0.4440	0.4488	0.0600	0.0553	0.0048	-0.0047
	3	0.2870	0.2870	0.0310	0.0260	0.0	-0.0050

Table C-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		77.35 K (-195.8°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0087		$\lvert\Delta y\rvert_{av}$ = 0.0081			
125.00	1	0.2140	0.2273	0.9040	0.9235	0.0133	-0.0195
	2	0.3290	0.3157	0.0460	0.0381	-0.0133	-0.0079
	3	0.4430	0.4430	0.0500	0.0382	0.0	-0.0118
125.00	1	0.1430	0.1640	0.9370	0.9419	0.0210	0.0049
	2	0.2740	0.2530	0.0340	0.0282	-0.0210	-0.0058
	3	0.3670	0.3670	0.0290	0.0275	0.0	-0.0015
150.00	1	0.2680	0.2841	0.8490	0.9006	0.0161	0.0516
	2	0.4450	0.4289	0.1060	0.0667	-0.0161	-0.0394
	3	0.2730	0.2730	0.0450	0.0323	0.0	-0.0127
150.00	1	0.2620	0.2573	0.8910	0.9116	-0.0047	0.0206
	2	0.3450	0.3497	0.0630	0.0504	0.0047	-0.0126
	3	0.3470	0.3470	0.0460	0.0368	0.0	-0.0092
150.00	1	0.2320	0.2539	0.8960	0.9142	0.0219	0.0182
	2	0.3120	0.2901	0.0550	0.0414	-0.0219	-0.0136
	3	0.4190	0.4190	0.0490	0.0434	0.0	-0.0056
150.00	1	0.2550	0.2535	0.8980	0.9156	-0.0015	0.0176
	2	0.2420	0.2436	0.0430	0.0347	0.0015	-0.0083
	3	0.4790	0.4790	0.0590	0.0491	0.0	-0.0099
150.00	1	0.2160	0.2079	0.9190	0.9233	-0.0081	0.0043
	2	0.3910	0.3991	0.0530	0.0536	0.0081	0.0006
	3	0.2060	0.2060	0.0240	0.0197	0.0	-0.0043
150.00	1	0.2100	0.2125	0.9080	0.9209	0.0025	0.0129
	2	0.4400	0.4375	0.0700	0.0593	-0.0025	-0.0107
	3	0.1680	0.1680	0.0180	0.0164	0.0	-0.0016
150.00	1	0.2230	0.2392	0.8990	0.9163	0.0162	0.0173
	2	0.3830	0.3668	0.0640	0.0511	-0.0162	-0.0129
	3	0.3010	0.3010	0.0370	0.0304	0.0	-0.0066
150.00	1	0.2970	0.2861	0.8720	0.8980	-0.0109	0.0260
	2	0.5030	0.5139	0.1020	0.0806	0.0109	-0.0214
	3	0.1680	0.1680	0.0260	0.0203	0.0	-0.0057
150.00	1	0.2670	0.2613	0.8920	0.9116	-0.0057	0.0196
	2	0.2970	0.3027	0.0530	0.0440	0.0057	-0.0090
	3	0.4120	0.4120	0.0550	0.0437	0.0	-0.0113

Table C-1 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		77.35 K (-195.8°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0087		$\lvert\Delta y\rvert_{av}$ = 0.0081			
150.00	1	0.2620	0.2736	0.8810	0.9048	0.0116	0.0238
	2	0.4190	0.4074	0.0800	0.0614	-0.0116	-0.0186
	3	0.2910	0.2910	0.0390	0.0329	0.0	-0.0061
150.00	1	0.1740	0.1902	0.9130	0.9331	0.0162	0.0201
	2	0.2610	0.2448	0.0430	0.0319	-0.0162	-0.0111
	3	0.3580	0.3580	0.0440	0.0319	0.0	-0.0121

Table C-2

Comparison of Experimental and Calculated VLE Compositions for the Quaternary System H_2(1)-N_2(2)-CO(3)-CH_4(4) (100), (Modified SRK Equation)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
			103.15 K (-170°C)				
			$\|\Delta x\|_{av}$ = 0.0182		$\|\Delta y\|_{av}$ = 0.0009		
75.00	1	0.1380	0.1206	0.8510	0.8490	-0.0174	-0.0020
	2	0.1150	0.1324	0.0440	0.0506	0.0174	0.0066
	3	0.3110	0.3110	0.0860	0.0852	0.0	-0.0008
75.00	1	0.1300	0.1421	0.8230	0.8147	0.0121	-0.0083
	2	0.2600	0.2479	0.0890	0.0913	-0.0121	0.0023
	3	0.2820	0.2820	0.0730	0.0797	0.0	0.0067
75.00	1	0.3050	0.2405	0.7530	0.7143	-0.0645	-0.0387
	2	0.2970	0.3615	0.1190	0.1519	0.0645	0.0329
	3	0.3700	0.3700	0.1280	0.1306	0.0	0.0026
75.00	1	0.1520	0.1883	0.8040	0.7937	0.0363	-0.0103
	2	0.0730	0.0367	0.0260	0.0151	-0.0363	-0.0109
	3	0.6710	0.6710	0.1640	0.1842	0.0	0.0202
75.00	1	0.1540	0.1640	0.7930	0.7848	0.0100	-0.0082
	2	0.3890	0.3790	0.1320	0.1372	-0.0100	0.0052
	3	0.2150	0.2150	0.0610	0.0647	0.0	0.0037
100.00	1	0.3800	0.3733	0.6630	0.6706	-0.0067	0.0076
	2	0.3980	0.4047	0.2280	0.2194	0.0067	-0.0086
	3	0.2110	0.2110	0.1090	0.1072	0.0	-0.0018
100.00	1	0.1260	0.1461	0.8600	0.8688	0.0201	0.0088
	2	0.1090	0.0889	0.0400	0.0340	-0.0201	-0.0060
	3	0.2880	0.2880	0.0790	0.0795	0.0	0.0005
100.00	1	0.2470	0.2603	0.7850	0.7841	0.0133	-0.0009
	2	0.0680	0.0547	0.0250	0.0237	-0.0133	-0.0013
	3	0.5910	0.5910	0.1820	0.1834	0.0	0.0014
100.00	1	0.2070	0.2177	0.7900	0.7878	0.0107	-0.0022
	2	0.3580	0.3473	0.1330	0.1315	-0.0107	-0.0015
	3	0.2000	0.2000	0.0590	0.0642	0.0	0.0052
125.00	1	0.1330	0.1713	0.8560	0.8737	0.0383	0.0177
	2	0.1120	0.0737	0.0430	0.0291	-0.0383	-0.0139
	3	0.2640	0.2640	0.0750	0.0764	0.0	0.0014

Table C-2 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
			103.15 K (-170°C)				
		$\|\Delta x\|_{av}$ = 0.0182		$\|\Delta y\|_{av}$ = 0.0009			
125.00	1	0.2260	0.2320	0.8000	0.8123	0.0060	0.0123
	2	0.2130	0.2070	0.0880	0.0831	-0.0060	-0.0049
	3	0.2600	0.2600	0.0830	0.0835	0.0	0.0005
125.00	1	0.3360	0.3246	0.7370	0.7492	-0.0114	0.0122
	2	0.2650	0.2764	0.1300	0.1269	0.0114	-0.0031
	3	0.2670	0.2670	0.0110	0.1057	0.0	-0.0053
125.00	1	0.1450	0.1633	0.8780	0.8859	0.0183	0.0079
	2	0.0560	0.0377	0.0230	0.0150	-0.0183	-0.0080
	3	0.2760	0.2760	0.0770	0.0786	0.0	0.0016
125.00	1	0.2940	0.3234	0.7590	0.7709	0.0294	0.0119
	2	0.0620	0.0326	0.0290	0.0158	-0.0294	-0.0132
	3	0.5500	0.5500	0.1990	0.2006	0.0	0.0016
150.00	1	0.2040	0.2015	0.8580	0.8651	-0.0025	0.0071
	2	0.0950	0.0975	0.0390	0.0399	0.0025	0.0009
	3	0.2260	0.2260	0.0720	0.0705	0.0	-0.0015
150.00	1	0.2410	0.2619	0.7880	0.8086	0.0209	0.0206
	2	0.1980	0.1771	0.0900	0.0771	-0.0209	-0.0129
	3	0.2490	0.2490	0.0880	0.0877	0.0	-0.0003
150.00	1	0.3980	0.3990	0.6760	0.7250	0.0010	0.0490
	2	0.2470	0.2460	0.1510	0.1336	-0.0010	-0.0174
	3	0.2390	0.2390	0.1360	0.1160	0.0	-0.0200
150.00	1	0.1580	0.1841	0.8700	0.8885	0.0261	0.0185
	2	0.0520	0.0259	0.0220	0.0107	-0.0261	-0.0113
	3	0.2560	0.2560	0.0740	0.0771	0.0	0.0031
150.00	1	0.4050	0.4060	0.6810	0.7448	0.0010	0.0638
	2	0.0520	0.0510	0.0340	0.0285	-0.0010	-0.0055
	3	0.4620	0.4620	0.2570	0.2094	0.0	-0.0476
			93.15 K (-180°C)				
		$\|\Delta x\|_{av}$ = 0.0310		$\|\Delta y\|_{av}$ = 0.0195			
150.00	1	0.4418	0.4254	0.7107	0.7837	-0.0164	0.0730
	2	0.2560	0.2724	0.1547	0.1172	0.0164	-0.0375
	3	0.2339	0.2339	0.1159	0.0887	0.0	-0.0272

Table C-2 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		93.15 K (-180°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0310		$\lvert\Delta y\rvert_{av}$ = 0.0195			
150.00	1	0.4120	0.3698	0.7548	0.8040	-0.0422	0.0492
	2	0.4080	0.4502	0.1979	0.1635	0.0422	-0.0344
	3	0.0547	0.0547	0.0215	0.0192	0.0	-0.0023
150.00	1	0.3686	0.3470	0.7860	0.8202	-0.0216	0.0342
	2	0.2508	0.2724	0.1088	0.0967	0.0216	-0.0121
	3	0.2365	0.2365	0.0852	0.0708	0.0	-0.0144
150.00	1	0.3545	0.3469	0.7839	0.8210	-0.0076	0.0371
	2	0.2540	0.2616	0.1052	0.0930	0.0076	-0.0122
	3	0.2483	0.2483	0.0905	0.0739	0.0	-0.0166
150.00	1	0.1703	0.2103	0.8523	0.8806	0.0400	0.0283
	2	0.2829	0.2429	0.0913	0.0735	-0.0400	-0.0178
	3	0.1322	0.1322	0.0319	0.0320	0.0	0.0001
150.00	1	0.0705	0.1207	0.9500	0.9641	0.0502	0.0141
	2	0.0791	0.0289	0.0205	0.0094	-0.0502	-0.0111
	3	0.0619	0.0619	0.0090	0.0142	0.0	0.0052
150.00	1	0.1545	0.1850	0.8697	0.8931	0.0305	0.0234
	2	0.3207	0.2902	0.0997	0.0864	-0.0305	-0.0133
	3	0.0251	0.0251	0.0054	0.0062	0.0	0.0008
150.00	1	0.1711	0.1894	0.8812	0.8970	0.0183	0.0158
	2	0.1895	0.1712	0.0587	0.0521	-0.0183	-0.0066
	3	0.1622	0.1622	0.0391	0.0374	0.0	-0.0017
150.00	1	0.1306	0.1758	0.8935	0.9119	0.0452	0.0184
	2	0.1394	0.0942	0.0386	0.0290	-0.0452	-0.0096
	3	0.2092	0.2092	0.0464	0.0462	0.0	-0.0002
150.00	1	0.2479	0.2837	0.8007	0.8414	0.0358	0.0407
	2	0.3562	0.3204	0.1269	0.1030	-0.0358	-0.0239
	3	0.1505	0.1505	0.0507	0.0414	0.0	-0.0093
150.00	1	0.4007	0.4335	0.6761	0.7645	0.0328	0.0884
	2	0.4312	0.3984	0.2462	0.1802	-0.0328	-0.0660
	3	0.0985	0.0985	0.0552	0.0428	0.0	-0.0124
		77.35 K (-195.8°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0676		$\lvert\Delta y\rvert_{av}$ = 0.0242			
75.00	1	0.1880	0.2376	0.9590	0.9378	0.0496	-0.0212
	2	0.4970	0.4474	0.0240	0.0408	-0.0496	0.0168
	3	0.2990	0.2990	0.0170	0.0212	0.0	0.0042

Table C-2 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		77.35 K (-195.8°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0676		$\lvert\Delta y\rvert_{av}$ = 0.0242			
75.00	1	0.1220	0.1230	0.9460	0.1232	0.0010	-0.8228
	2	0.3940	0.3930	0.0320	0.3933	-0.0010	0.3613
	3	0.4740	0.4740	0.0220	0.4735	0.0	0.4515
75.00	1	0.1300	0.2168	0.9430	0.9435	0.0868	0.0005
	2	0.4140	0.3272	0.0330	0.0306	-0.0868	-0.0024
	3	0.4060	0.4060	0.0240	0.0255	0.0	0.0015
75.00	1	0.1620	0.2143	0.9390	0.9465	0.0523	0.0075
	2	0.2550	0.2027	0.0260	0.0207	-0.0523	-0.0053
	3	0.5570	0.5570	0.0350	0.0326	0.0	-0.0024
75.00	1	0.1310	0.1767	0.9470	0.9489	0.0457	0.0019
	2	0.4520	0.4063	0.0370	0.0348	-0.0457	-0.0022
	3	0.2380	0.2380	0.0160	0.0154	0.0	-0.0006
75.00	1	0.1300	0.1794	0.9430	0.9479	0.0494	0.0049
	2	0.5050	0.4556	0.0440	0.0383	-0.0494	-0.0057
	3	0.1920	0.1920	0.0130	0.0129	0.0	-0.0001
75.00	1	0.1190	0.2027	0.9360	0.9453	0.0837	0.0093
	2	0.4530	0.3963	0.0390	0.0330	-0.0837	-0.0060
	3	0.3330	0.3330	0.0250	0.0212	0.0	-0.0038
75.00	1	0.1660	0.2296	0.9270	0.9383	0.0636	0.0113
	2	0.6060	0.5424	0.0580	0.0470	-0.0636	-0.0110
	3	0.1900	0.1900	0.0150	0.0144	0.0	-0.0006
75.00	1	0.1390	0.2214	0.9390	0.9437	0.0824	0.0047
	2	0.3540	0.2716	0.0330	0.0266	-0.0824	-0.0064
	3	0.4810	0.4810	0.0280	0.0295	0.0	0.0015
75.00	1	0.1250	0.2275	0.9330	0.9404	0.1025	0.0074
	2	0.5000	0.3975	0.0470	0.0364	-0.1025	-0.0106
	3	0.3410	0.3410	0.0200	0.0229	0.0	0.0029
75.00	1	0.1230	0.2245	0.9450	0.9433	0.1015	-0.0017
	2	0.3620	0.2605	0.0310	0.0259	-0.1015	-0.0051
	3	0.5000	0.5000	0.0240	0.0307	0.0	0.0067
75.00	1	0.1030	0.1603	0.9520	0.9548	0.0573	0.0028
	2	0.2930	0.2357	0.0260	0.0220	-0.0573	-0.0040
	3	0.3880	0.3880	0.0220	0.0223	0.0	0.0003

Table C-2 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		77.35 K (-195.8°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0676		$\lvert\Delta y\rvert_{av}$ = 0.0242			
100.00	1	0.2160	0.2913	0.9270	0.9288	0.0753	0.0018
	2	0.3210	0.2457	0.0380	0.0323	-0.0753	-0.0057
	3	0.4450	0.4450	0.0350	0.0387	0.0	0.0037
100.00	1	0.2000	0.2834	0.9230	0.9291	0.0834	0.0061
	2	0.3790	0.2956	0.0420	0.0370	-0.0834	-0.0050
	3	0.3790	0.3790	0.0350	0.0333	0.0	-0.0017
100.00	1	0.1810	0.2246	0.9410	0.9375	0.0436	-0.0035
	2	0.4290	0.3854	0.0440	0.0423	-0.0436	-0.0017
	3	0.2180	0.2180	0.0150	0.0187	0.0	0.0037
100.00	1	0.1550	0.2256	0.9330	0.9367	0.0706	0.0037
	2	0.4990	0.4284	0.0530	0.0463	-0.0706	-0.0067
	3	0.1740	0.1740	0.0140	0.0155	0.0	0.0015
100.00	1	0.1700	0.2586	0.9330	0.9329	0.0886	-0.0001
	2	0.4150	0.3264	0.0450	0.0384	-0.0886	-0.0066
	3	0.3230	0.3230	0.0220	0.0277	0.0	0.0057
100.00	1	0.2100	0.2939	0.9150	0.9238	0.0839	0.0088
	2	0.5630	0.4791	0.0690	0.0564	-0.0839	-0.0126
	3	0.1840	0.1840	0.0160	0.0191	0.0	0.0031
100.00	1	0.1440	0.2034	0.9430	0.9446	0.0594	0.0016
	2	0.2880	0.2286	0.0320	0.0266	-0.0594	-0.0054
	3	0.3600	0.3600	0.0250	0.0272	0.0	0.0022
125.00	1	0.2890	0.3660	0.8580	0.9053	0.0770	0.0473
	2	0.3110	0.2340	0.0700	0.0431	-0.0770	-0.0269
	3	0.3900	0.3900	0.0720	0.0513	0.0	-0.0207
125.00	1	0.2490	0.3112	0.9170	0.9153	0.0622	-0.0017
	2	0.3620	0.2998	0.0550	0.0472	-0.0622	-0.0078
	3	0.3010	0.3010	0.0280	0.0358	0.0	0.0078

Table C-3

Comparison of Experimental and Calculated VLE Compositions for the Quaternary System $H_2(1)$-$N_2(2)$-$CO(3)$-$CH_4(4)$ (Modified BWR Equation)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
			103.15 K (-170.0°C)				
		$\|\Delta x\|_{av}$ = 0.0248		$\|\Delta y\|_{av}$ = 0.0007			
75.00	1	0.1380	0.1163	0.8510	0.8579	-0.0217	0.0069
	2	0.1150	0.1367	0.0440	0.0472	0.0217	0.0032
	3	0.3110	0.3110	0.0860	0.0789	0.0	-0.0071
75.00	1	0.1300	0.1330	0.8230	0.8280	0.0030	0.0050
	2	0.2600	0.2570	0.0890	0.0854	-0.0030	-0.0036
	3	0.2820	0.2820	0.0730	0.0719	0.0	-0.0011
75.00	1	0.3050	0.1845	0.7530	0.7411	-0.1205	-0.0119
	2	0.2970	0.4175	0.1190	0.1480	0.1205	0.0290
	3	0.3700	0.3700	0.1280	0.1084	0.0	-0.0196
75.00	1	0.1520	0.1510	0.8040	0.8012	-0.0010	-0.0028
	2	0.0730	0.0740	0.0260	0.0250	0.0010	-0.0010
	3	0.6710	0.6710	0.1640	0.1672	0.0	0.0032
75.00	1	0.1540	0.1493	0.7930	0.8006	-0.0047	0.0076
	2	0.3590	0.3937	0.1320	0.1295	0.0047	-0.0025
	3	0.2150	0.2150	0.0610	0.0568	0.0	-0.0042
100.00	1	0.1260	0.1463	0.8600	0.8733	0.0203	0.0133
	2	0.1090	0.0887	0.0400	0.0314	-0.0203	-0.0086
	3	0.2880	0.2880	0.0790	0.0756	0.0	-0.0034
100.00	1	0.2470	0.2169	0.7850	0.7893	-0.0301	0.0043
	2	0.0680	0.0981	0.0250	0.0356	0.0301	0.0106
	3	0.5910	0.5910	0.1820	0.1667	0.0	-0.0154
100.00	1	0.2070	0.2052	0.7900	0.8008	-0.0018	0.0108
	2	0.3580	0.3598	0.1330	0.1250	0.0018	-0.0080
	3	0.2000	0.2000	0.0590	0.0573	0.0	-0.0017
125.00	1	0.1330	0.1764	0.8560	0.8752	0.0434	0.0192
	2	0.1120	0.0686	0.0430	0.0256	-0.0434	-0.0174
	3	0.2640	0.2640	0.0750	0.0745	0.0	-0.0005
125.00	1	0.2260	0.2304	0.8000	0.8203	0.0044	0.0203
	2	0.2130	0.2086	0.0880	0.0779	-0.0044	-0.0102
	3	0.2600	0.2600	0.0830	0.0784	0.0	-0.0046

Table C-3 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		103.15 K (-170.0°C)					
		$\|\Delta x\|_{av}$ = 0.0248		$\|\Delta y\|_{av}$ = 0.0007			
125.00	1	0.3360	0.2985	0.7370	0.7569	-0.0375	0.0199
	2	0.2650	0.3025	0.1300	0.1277	0.0375	-0.0023
	3	0.2670	0.2670	0.1110	0.0970	0.0	-0.0140
125.00	1	0.1450	0.1683	0.8780	0.8858	0.0233	0.0078
	2	0.0560	0.0327	0.0230	0.0123	-0.0233	-0.0107
	3	0.2760	0.2760	0.0770	0.0774	0.0	0.0004
125.00	1	0.2940	0.2825	0.7590	0.7726	-0.0115	0.0136
	2	0.0620	0.0735	0.0290	0.0305	0.0115	0.0015
	3	0.5500	0.5500	0.1990	0.1846	0.0	-0.0144
150.00	1	0.2040	0.2130	0.8580	0.8636	0.0090	0.0056
	2	0.0950	0.0860	0.0390	0.0345	-0.0090	-0.0045
	3	0.2260	0.2260	0.0720	0.0707	0.0	-0.0013
150.00	1	0.2410	0.2692	0.7880	0.8140	0.0282	0.0260
	2	0.1980	0.1698	0.0900	0.0702	-0.0282	-0.0198
	3	0.2490	0.2490	0.0830	0.0846	0.0	-0.0034
150.00	1	0.1580	0.1941	0.8700	0.8853	-0.0361	0.0153
	2	0.0520	0.0159	0.0220	0.0064	-0.0361	-0.0156
	3	0.2560	0.2560	0.0740	0.0780	0.0	0.0040
		93.15 K (-180°C)					
		$\|\Delta x\|_{av}$ = 0.0509		$\|\Delta y\|_{av}$ = 0.0142			
150.00	1	0.4410	0.4075	0.7100	0.7652	-0.0335	0.0552
	2	0.2560	0.2895	0.1550	0.1290	0.0335	-0.0260
	3	0.2340	0.2340	0.1160	0.0925	0.0	-0.0235
150.00	1	0.4120	0.3823	0.7540	0.7822	-0.0297	0.0282
	2	0.4080	0.4377	0.1980	0.1783	0.0297	-0.0197
	3	0.0550	0.0550	0.0220	0.0203	0.0	-0.0017
150.00	1	0.3680	0.3447	0.7860	0.8073	-0.0233	0.0213
	2	0.2510	0.2743	0.1090	0.1013	0.0233	-0.0077
	3	0.2370	0.2370	0.0850	0.0744	0.0	-0.0106
150.00	1	0.3550	0.3438	0.7840	0.8080	-0.0112	0.0240
	2	0.2540	0.2652	0.1050	0.0978	0.0112	-0.0072
	3	0.2480	0.2480	0.0910	0.0775	0.0	-0.0135

Table C-3 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		93.15 K (-180°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0509		$\lvert\Delta y\rvert_{av}$ = 0.0142			
150.00	1	0.2370	0.2845	0.8200	0.8453	0.0475	0.0253
	2	0.2050	0.1575	0.0680	0.0515	-0.0475	-0.0165
	3	0.3320	0.3320	0.0930	0.0861	0.0	-0.0069
150.00	1	0.0640	0.1670	0.8890	0.9177	0.0830	0.0287
	2	0.2270	0.1440	0.0760	0.0472	-0.0830	-0.0288
	3	0.0630	0.0630	0.0120	0.0154	0.0	0.0034
150.00	1	0.1120	0.1974	0.8670	0.8934	0.0854	0.0264
	2	0.2930	0.2076	0.0890	0.0659	-0.0854	-0.0231
	3	0.0840	0.0840	0.0180	0.0205	0.0	0.0025
150.00	1	0.1700	0.2255	0.8520	0.8759	0.0555	0.0239
	2	0.2830	0.2275	0.0910	0.0714	-0.0555	-0.0196
	3	0.1320	0.1320	0.0320	0.0326	0.0	0.0006
150.00	1	0.1420	0.2185	0.8570	0.8810	0.0765	0.0240
	2	0.2750	0.1985	0.0810	0.0622	-0.0765	-0.0188
	3	0.1520	0.1520	0.0390	0.0369	0.0	-0.0021
150.00	1	0.0710	0.1317	0.9500	0.9600	0.0607	0.0100
	2	0.0790	0.0183	0.0200	0.0064	-0.0607	-0.0136
	3	0.0620	0.0620	0.0090	0.0153	0.0	0.0063
150.00	1	0.1550	0.2044	0.8700	0.8865	0.0494	0.0165
	2	0.3210	0.2716	0.1000	0.0863	-0.0494	-0.0137
	3	0.0250	0.0250	0.0050	0.0063	0.0	0.0013
150.00	1	0.1710	0.2028	0.8810	0.8921	0.0318	0.0111
	2	0.1900	0.1582	0.0590	0.0497	-0.0318	-0.0093
	3	0.1620	0.1620	0.0390	0.0386	0.0	-0.0004
150.00	1	0.1310	0.1864	0.8940	0.9063	0.0554	0.0124
	2	0.1390	0.0836	0.0390	0.0264	-0.0554	-0.0126
	3	0.2090	0.2090	0.0460	0.0484	0.0	0.0024
150.00	1	0.0840	0.1614	0.9160	0.9279	0.0774	0.0119
	2	0.1160	0.0386	0.0330	0.0126	-0.0774	-0.0204
	3	0.1760	0.1760	0.0300	0.0410	0.0	0.0110
150.00	1	0.1580	0.2233	0.8510	0.8785	0.0653	0.0275
	2	0.2580	0.1927	0.0850	0.0603	-0.0653	-0.0247
	3	0.1710	0.1710	0.0390	0.0415	0.0	0.0025

Table C-3 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		93.15 K (-180°C)					
		$\|\Delta x\|_{av}$ = 0.0509		$\|\Delta y\|_{av}$ = 0.0142			
150.00	1	0.2480	0.2957	0.8000	0.8354	0.0477	0.0354
	2	0.3560	0.3063	0.1270	0.1028	-0.0477	-0.0242
	3	0.1510	0.1510	0.0510	0.0421	0.0	-0.0089
150.00	1	0.4010	0.4328	0.6760	0.7477	0.0318	0.0717
	2	0.4310	0.3992	0.2460	0.1924	-0.0318	-0.0536
	3	0.0990	0.0990	0.0550	0.0440	0.0	-0.0110
		77.35 K (-195.8°C)					
		$\|\Delta x\|_{av}$ = 0.0517		$\|\Delta y\|_{av}$ = 0.0009			
75.00	1	0.1880	0.1818	0.9590	0.9338	-0.0062	-0.0252
	2	0.4970	0.5032	0.0240	0.0459	0.0062	0.0219
	3	0.2990	0.2990	0.0170	0.0201	0.0	0.0031
75.00	1	0.1220	0.1664	0.9460	0.9385	0.0444	-0.0075
	2	0.3940	0.3496	0.0320	0.0320	-0.0444	0.0
	3	0.4740	0.4740	0.0220	0.0295	0.0	0.0075
75.00	1	0.1940	0.2001	0.9340	0.9266	0.0061	-0.0074
	2	0.7660	0.7599	0.0660	0.0709	-0.0061	0.0049
	3	0.0310	0.0310	0.0	0.0024	0.0	0.0024
75.00	1	0.1410	0.1768	0.9000	0.9357	0.0358	0.0357
	2	0.4950	0.4592	0.0100	0.0417	-0.0358	0.0317
	3	0.3410	0.3410	0.0100	0.0223	0.0	0.0123
75.00	1	0.1300	0.1659	0.9430	0.9401	0.0359	-0.0029
	2	0.4140	0.3781	0.0330	0.0341	-0.0359	0.0011
	3	0.4060	0.4060	0.0240	0.0253	0.0	0.0013
75.00	1	0.1620	0.1549	0.9390	0.9427	-0.0071	0.0037
	2	0.2550	0.2621	0.0260	0.0240	0.0071	-0.0020
	3	0.5570	0.5570	0.0350	0.0331	0.0	-0.0019
75.00	1	0.1310	0.1553	0.9470	0.9457	0.0243	-0.0013
	2	0.4520	0.4277	0.0370	0.0381	-0.0243	0.0011
	3	0.2380	0.2380	0.0160	0.0150	0.0	-0.0010
75.00	1	0.1300	0.1594	0.9430	0.9441	0.0294	0.0011
	2	0.5050	0.4756	0.0440	0.0423	-0.0294	-0.0017
	3	0.1920	0.1920	0.0130	0.0123	0.0	-0.0007

Table C-3 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		77.35 K (-195.8°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0517		$\lvert\Delta y\rvert_{av}$ = 0.0009			
75.00	1	0.1190	0.1639	0.9360	0.9419	0.0499	0.0059
	2	0.4530	0.4081	0.0390	0.0365	-0.0449	-0.0025
	3	0.3330	0.3330	0.0250	0.0208	0.0	-0.0042
75.00	1	0.1660	0.1858	0.9270	0.9332	0.0198	0.0062
	2	0.6060	0.5862	0.0580	0.0532	-0.0198	-0.0048
	3	0.1900	0.1900	0.0150	0.0132	0.0	-0.0018
75.00	1	0.1390	0.1629	0.9390	0.9402	0.0239	0.0012
	2	0.3540	0.3301	0.0330	0.0301	-0.0239	-0.0030
	3	0.4810	0.4810	0.0280	0.0295	0.0	0.0015
75.00	1	0.1250	0.1748	0.9330	0.9368	0.0498	0.0038
	2	0.5000	0.4502	0.0470	0.0408	-0.0498	-0.0062
	3	0.3410	0.3410	0.0200	0.0221	0.0	0.0021
75.00	1	0.1230	0.1628	0.9450	0.9398	0.0398	-0.0052
	2	0.3620	0.3222	0.0310	0.0294	-0.0398	-0.0016
	3	0.5000	0.5000	0.0240	0.0306	0.0	0.0066
75.00	1	0.1030	0.1375	0.9520	0.9523	0.0345	0.0003
	2	0.2930	0.2585	0.0260	0.0235	-0.0345	-0.0025
	3	0.3880	0.3880	0.0220	0.0229	0.0	0.0009
100.00	1	0.2160	0.2218	0.9270	0.9179	0.0058	-0.0091
	2	0.3210	0.3152	0.0380	0.0407	-0.0058	0.0027
	3	0.4450	0.4450	0.0350	0.0411	0.0	0.0061
100.00	1	0.1770	0.2230	0.9240	0.9172	0.0460	-0.0068
	2	0.3640	0.3180	0.0390	0.0412	-0.0460	0.0022
	3	0.4450	0.4450	0.0370	0.0413	0.0	0.0043
100.00	1	0.2000	0.2246	0.9230	0.9185	0.0246	-0.0045
	2	0.3790	0.3544	0.0420	0.0453	-0.0246	0.0033
	3	0.3790	0.3790	0.0350	0.0353	0.0	0.0003
100.00	1	0.1720	0.2048	0.9370	0.9236	0.0328	-0.0135
	2	0.2650	0.2322	0.0290	0.0295	-0.0328	0.0005
	3	0.5370	0.5370	0.0340	0.0465	0.0	0.0125
100.00	1	0.1810	0.2044	0.9410	0.9284	0.0234	-0.0126
	2	0.4290	0.4056	0.0440	0.0495	-0.0234	0.0055
	3	0.2180	0.2180	0.0150	0.0197	0.0	0.0047

Table C-3 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		77.35 K (-195.8°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0517		$\lvert\Delta y\rvert_{av}$ = 0.0009			
100.00	1	0.1550	0.2085	0.9330	0.9270	0.0535	-0.0060
	2	0.4990	0.4455	0.0530	0.0545	-0.0535	0.0015
	3	0.1740	0.1740	0.0140	0.0161	0.0	0.0021
100.00	1	0.1700	0.2167	0.9330	0.9231	0.0467	-0.0099
	2	0.4150	0.3683	0.0450	0.0459	-0.0467	0.0009
	3	0.3230	0.3230	0.0220	0.0294	0.0	0.0074
100.00	1	0.2100	0.2498	0.9150	0.9107	0.0398	-0.0043
	2	0.5630	0.5232	0.0690	0.0690	-0.0398	0.0
	3	0.1840	0.1840	0.0160	0.0193	0.0	0.0033
100.00	1	0.1590	0.2153	0.9250	0.9205	0.0563	-0.0045
	2	0.3450	0.2887	0.0390	0.0369	-0.0563	-0.0021
	3	0.4680	0.4680	0.0360	0.0421	0.0	0.0061
100.00	1	0.1950	0.2381	0.9210	0.9138	0.0432	-0.0072
	2	0.4630	0.4199	0.0510	0.0548	-0.0432	0.0038
	3	0.3120	0.3120	0.0280	0.0308	0.0	0.0028
100.00	1	0.1570	0.2162	0.9240	0.9197	0.0592	-0.0043
	2	0.3460	0.2868	0.0380	0.0368	-0.0592	-0.0012
	3	0.4770	0.4770	0.0380	0.0431	0.0	0.0051
100.00	1	0.1440	0.1798	0.9430	0.9373	0.0358	-0.0057
	2	0.2880	0.2522	0.0320	0.0305	-0.0358	-0.0015
	3	0.3600	0.3600	0.0250	0.0297	0.0	0.0047
125.00	1	0.2220	0.3122	0.8750	0.8750	0.0902	0.0
	2	0.4750	0.3848	0.0880	0.0777	-0.0902	-0.0104
	3	0.2870	0.2870	0.0370	0.0465	0.0	0.0095
125.00	1	0.2890	0.2910	0.8580	0.8813	0.0020	0.0233
	2	0.3110	0.3090	0.0700	0.0600	-0.0020	-0.0100
	3	0.3900	0.3900	0.0720	0.0582	0.0	-0.0138
125.00	1	0.2200	0.2792	0.9130	0.8892	0.0592	-0.0238
	2	0.3700	0.3108	0.0540	0.0575	-0.0592	0.0035
	3	0.3650	0.3650	0.0330	0.0515	0.0	0.0185
125.00	1	0.2120	0.2701	0.9170	0.8913	0.0581	-0.0257
	2	0.3250	0.2669	0.0440	0.0490	-0.0581	0.0050
	3	0.4280	0.4280	0.0390	0.0390	0.0	0.0194

Table C-3 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		77.35 K (-195.8°C) $\lvert\Delta x\rvert_{av}$ = 0.0517 $\lvert\Delta y\rvert_{av}$ = 0.0009					
125.00	1	0.2130	0.2567	0.9210	0.8948	0.0437	-0.0262
	2	0.2570	0.2133	0.0340	0.0386	-0.0437	0.0046
	3	0.5040	0.5040	0.0450	0.0656	0.0	0.0206
125.00	1	0.1630	0.2406	0.9240	0.9084	0,0776	-0.0156
	2	0.4170	0.3394	0.0550	0.0567	-0.0776	0.0017
	3	0.2400	0.2400	0.0210	0.0302	0.0	0.0092
125.00	1	0.1860	0.2521	0.9230	0,9041	0.0661	-0.0189
	2	0.4710	0.4049	0.0610	0.0687	-0.0661	0.0077
	3	0.1690	0.1690	0.0160	0.0224	0.0	0.0064
125.00	1	0.2490	0.2711	0.9170	0.8951	0.0221	-0.0219
	2	0.3620	0.3399	0.0550	0.0606	-0.0221	0.0056
	3	0.3010	0.3010	0.0280	0.0412	0.0	0.0132
125.00	1	0.2390	0.3195	0.8960	0.8753	0.0805	-0.0207
	2	0.6500	0.4695	0.0820	0.0943	-0.0805	0.0123
	3	0.1680	0.1680	0.0220	0.0282	0.0	0.0062
125.00	1	0.1760	0.2606	0.9090	0.8935	0.0846	-0.0156
	2	0.3110	0.2264	0.0470	0.0412	-0.0846	-0.0058
	3	0.4870	0.4870	0.0440	0.0643	0.0	0.0203
125.00	1	0.2410	0.3051	0.9090	0.8788	0.0641	-0.0302
	2	0.4440	0.3799	0.0600	0.0747	-0.0641	0.0148
	3	0.2870	0.2870	0.0310	0.0451	0.0	0.0141
125.00	1	0.2140	0.2762	0.9040	0.8868	0.0622	-0.0172
	2	0.3290	0.2668	0.0460	0.0503	-0.0622	0.0043
	3	0.4430	0.4430	0.0500	0.0623	0.0	0.0123
125.00	1	0.1430	0.2101	0.9370	0.9206	0.0672	-0.0165
	2	0.2740	0.2069	0.0340	0.0335	-0.0672	-0.0005
	3	0.3670	0.3670	0.0290	0.0414	0.0	0.0124
150.00	1	0.2680	0.3877	0.8490	0.8186	0.1197	-0.0304
	2	0.4450	0.3253	0.1060	0.1049	-0.1197	-0.0011
	3	0.2730	0.2730	0.0450	0.0747	0.0	-0.0297
150.00	1	0.2620	0.3350	0.8910	0.8480	0.0730	-0.0430
	2	0.3450	0.2720	0.0630	0.0734	-0.0730	0.0104
	3	0.3470	0.3470	0.0460	0.0748	0.0	0.0288

Table C-3 (continued)

Pressure atm	Comp.	x Expl	x Calc	y Expl	y Calc	Δx	Δy
		77.35 K (-195.8°C)					
		$\lvert\Delta x\rvert_{av}$ = 0.0517		$\lvert\Delta y\rvert_{av}$ = 0.0009			
150.00	1	0.2550	0.3084	0.8980	0.8549	0.0534	-0.0431
	2	0.2420	0.1886	0.0430	0.0489	-0.0534	0.0059
	3	0.4790	0.4790	0.0590	0.0943	0.0	0.0353
150.00	1	0.2160	0.2788	0.9190	0.8808	0.0628	-0.0382
	2	0.3910	0.3282	0.0530	0.0739	-0.0628	0.0210
	3	0.2060	0.2060	0.0240	0.0364	0.0	0.0125
150.00	1	0.2100	0.2873	0.9080	0.8771	0.0773	-0.0309
	2	0.4400	0.3627	0.0700	0.0831	-0.0773	0.0131
	3	0.1680	0.1680	0.0180	0.0308	0.0	0.0128
150.00	1	0.2230	0.3151	0.8990	0.8620	0.0921	-0.0370
	2	0.3830	0.2909	0.0640	0.0724	-0.0921	0.0084
	3	0.3010	0.3010	0.0370	0.0593	0.0	0.0223
150.00	1	0.2970	0.4035	0.8720	0.8161	0.1065	-0.0558
	2	0.5030	0.3965	0.1020	0.1310	-0.1065	0.0290
	3	0.1680	0.1680	0.0260	0.0486	0.0	0.0226
150.00	1	0.2670	0.3301	0.8920	0.8458	0.0631	-0.0462
	2	0.2970	0.2339	0.0530	0.0638	-0.0631	0.0108
	3	0.4120	0.4120	0.0550	0.0883	0.0	0.0333
150.00	1	0.2620	0.3675	0.8810	0.8310	0.1055	-0.0500
	2	0.4190	0.3135	0.0800	0.0937	-0.1055	0.0137
	3	0.2910	0.2910	0.0390	0.0723	0.0	0.0333
150.00	1	0.1740	0.2444	0.9130	0.8968	0.0704	-0.0162
	2	0.2610	0.1906	0.0430	0.0405	-0.0704	-0.0025
	3	0.3580	0.3580	0.0440	0.0550	0.0	0.0110

APPENDIX D

CALIBRATION RESULTS OF THE GAS CHROMATOGRAPH

APPENDIX D

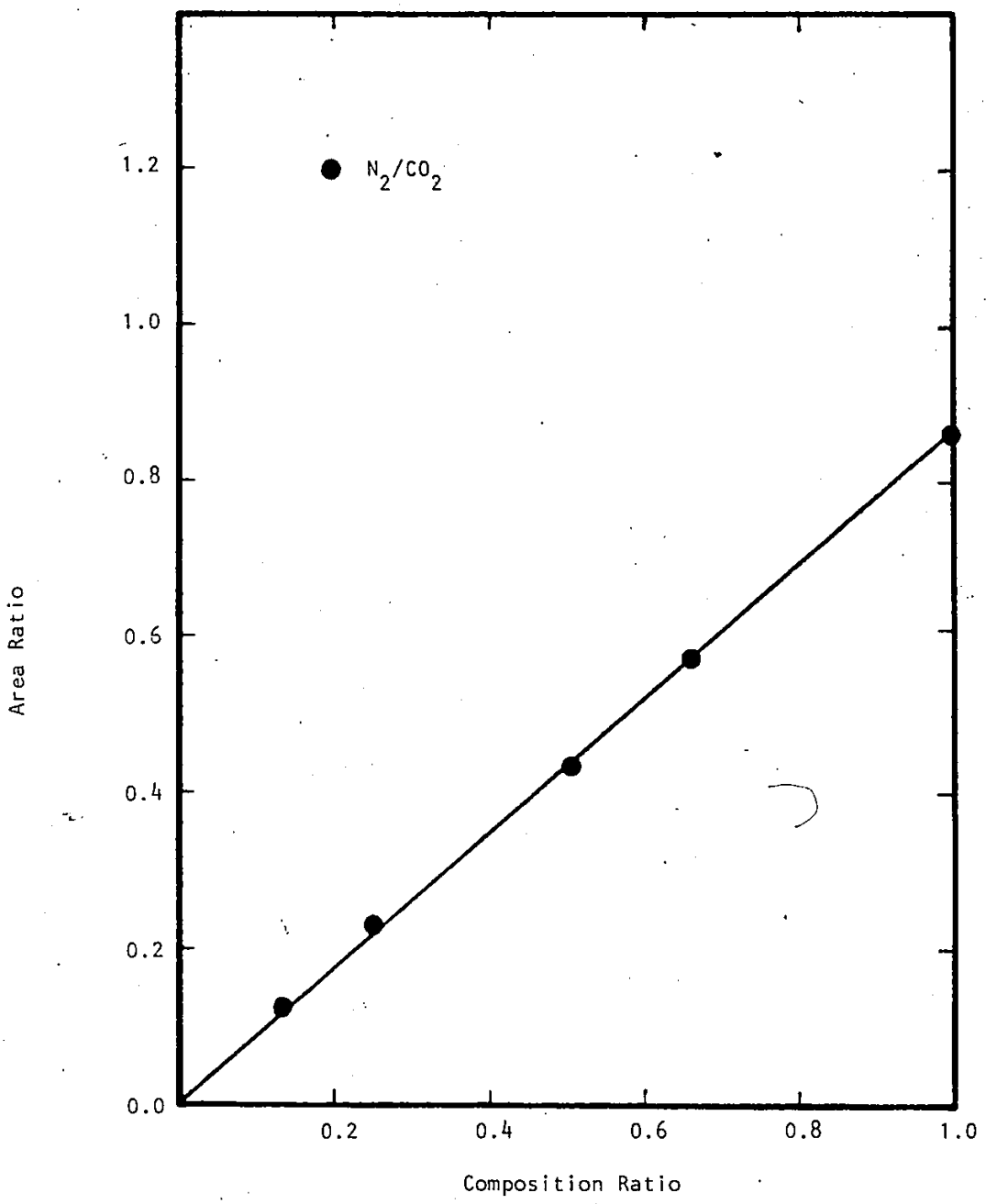

FIGURE D-1: CALIBRATION CURVE FOR NITROGEN AND CARBON DIOXIDE MIXTURES

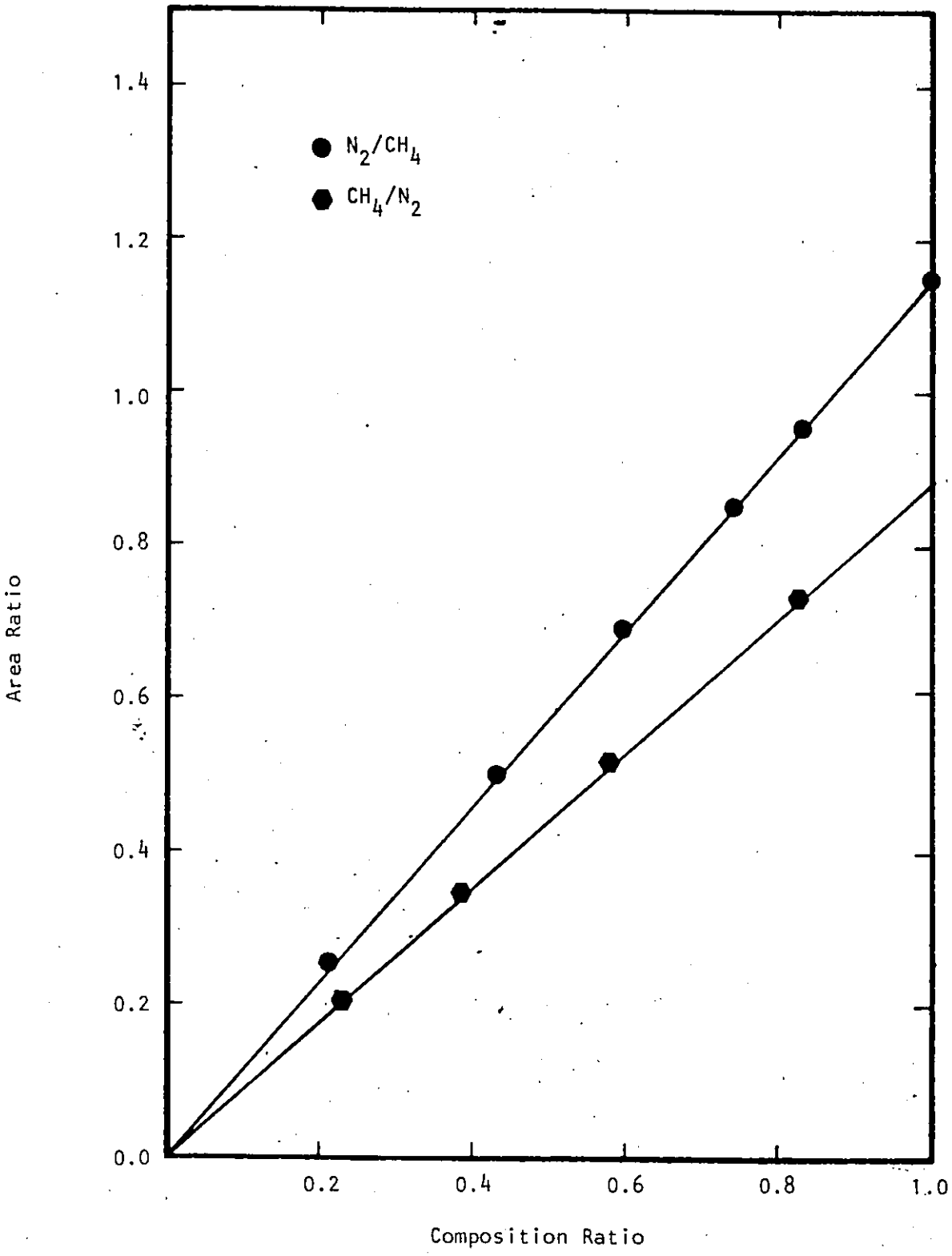

FIGURE D-2: CALIBRATION CURVE FOR NITROGEN AND METHANE MIXTURES

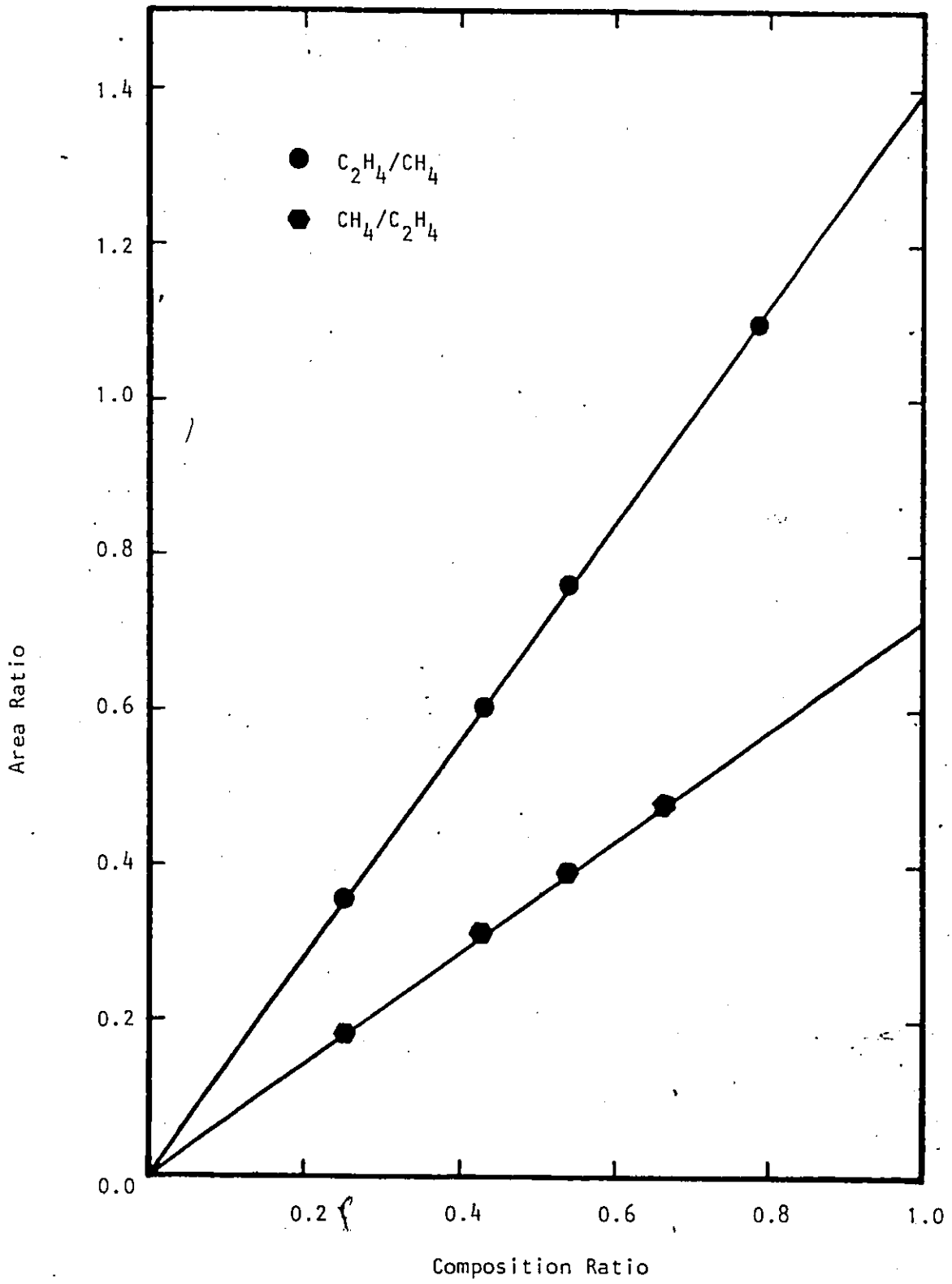

FIGURE D-3: CALIBRATION CURVE FOR METHANE AND ETHYLENE MIXTURES

APPENDIX E

COMPUTER PROGRAMS USED IN THIS INVESTIGATION

APPENDIX E

PROGRAM E-1

Computer Program Used to Predict VLE Properties and C_{ij} Values for Binary Systems by Using the ICL Equation of State

```
C ******************************************************************
C
C PROGRAM E-1
C
C COMPUTER PROGRAM USED TO PREDICT VLE PROPERTIES AND
C CIJ VALUES  FOR BINARY SYSTEMS BY USING THE ICL
C EQUATION OF STATE
C
C ******************************************************************
C
C     THIS PROGRAM IS USED TO FIND THE VALUE OF K12 FOR BINARY SYSTEM
C     SUCH THAT THE DEVIATION OF PRESSURE, PCALC. - PEXP., IS MINIMUM
C     THIS PROGRAM IS A COLLECTIVE VERSION OF THE PREVIOS ONES
C     MODIFIED BY C.-T. FU
C
C
C     DEPARTMENT OF CHEMICAL ENGINEERING
C
C     GRADUATE SCHOOL
C
C     STUDENT NUMBER 039129
C
C     FU CNENG TZE
C
C     MAY 24, 1979
C
C     ****************************************************************
C
C
C
C     SUBROUTINES PYINIT, PCALC, YVCALC, SUPT, CUBEQN AND EQNRK ARE
C     REQUIRED
C
C     ****************************************************************
      DIMENSION TITLE(20), PCII(5), VCII(5), TCII(5), W(5),
     1WAII(5), WBII(5), GOMMA(5), COMPB(5), COMPC(5),
     2FREFER(5), PHI(5), PHIV(5), PHIJ(5), PHIK(5), P1(5),
     3PTVOL(5), PPVOL(5), VVV(5), CORRL(5,5), X(5), Y(5),
     4DX(5), VC(5), TC(5), EXG(5), EXGF(5), YX(5), VVL(5),
     5DY(5), PVOL(5), GAMMB(5), GAMMA(5), AMOLWT(5), PC(5),
     6XX(50,5), YY(50,5), PP(50), EXG1(5), COMPD(5),
     7SSDK(5), ESSDK(5), SADK(5)
      DIMENSION A0(20), A1(20), A2(20), A3(20), B(20),
     1TR(5), S(5)
      DIMENSION EDYAV(5), DYAV(5), EDY(5)
      DIMENSION P00(5)
      COMMON /FIRST/ TC, PC, VCII, W, AMOLWT, T, P, R,
     1NCOMP, NONTUM /SECOND/ Z, A, MTYPE /THIRD/ PCII, TCII,
     2WAII, WBII, PSI, PP, XX, YY, IJK, KIJ, NOI, ITER, K,
     3CORRL, EXGT, PPVOL, PHI, PHIV, VV, VVL1, DV, P1, PPP,
     4Y /FOURTH/ SDP, EP, DP, SSDY, SDY, SDV, PHIK, GAMMA,
     5EXG, X, SUMY, GAMMB, PG, RT, PF, FREFER, YX, DY, EXG1
     6/FIFTH/ DPAV, EDYAV, EDY, EDP, EDPAV, DYAV, SSDK,
     7SADY, SADK /SIXTH/ PN /SEVEN/ P00, KPRINT
      NONH2 = 10
C      FOR NON-HYDROGEN SYSTEM SET NONH2=100
C      FOR HYDROGEN SYSTEM SET NONH2 =10
      READ (1,610) M
      M4 = 2 * (M + 1) + 2 * (M + 2)
      M1 = M + 2
```

```
C
      DO 10 I = 1, M4
      READ (1,770) A0(I), A1(I), A2(I), A3(I)
   10 WRITE (3,620) A0(I), A1(I), A2(I), A3(I)
C
   20 READ (1,630,END=600) (TITLE(I),I=1,20)
      WRITE (3,640) (TITLE(I),I=1,20)
      READ (1,650) NCOMP, NQNTUM, IJK, R
C      IF NQNTU IS 1 XX IS K VALVE, XX = YY/XX
C      NQNTUM = 2 , XX AND YY ARE GIVEN IN K1 AND K2, RESPECTIVELY
      IF (NCOMP .LE. 0) GO TO 20
C      NCOMP MUST BE GREATER THAN ZERO
C      NCOMP: NO. OF COMPONENTS IN THE MIXTURE
C      R: GAS CONSTANT
C      IJK: CONTROL VARIABLE
C      WRITE HEADINGS
      WRITE (3,660)
      WRITE (3,670)
C
      DO 30 I = 1, NCOMP
      READ (1,680) PCII(I), VCII(I), TCII(I), W(I), WAII(I),
     1WBII(I), COMPA(I), COMPB(I), COMPC(I), COMPD(I)
   30 WRITE (3,690) PCII(I), VCII(I), TCII(I), W(I),
     1WAII(I), WBII(I), COMPA(I), COMPB(I), COMPC(I),
     2COMPD(I)
C      IT            T
C      0        DEGREE K
C      1        DEGREE F
C      2        DEGREE C
      READ (1,700) T, PSI, IT
      IF (IT .EQ. 1) T = (T - 32.) * 5. / 9. + 273.15
      IF (IT .EQ. 2) T = T + 273.15
   40 WRITE (3,710) T, PSI
C      IJK           P
C      0          PSIA
C      1          ATM
C      2      MPA
C      3           MMHG
      READ (1,720) IJK, KIJ, CINI, CFIN
C      PP IS EXPERIMENTAL VALUE OF PRESSURE
      NCOMP1 = NCOMP - 1
      NOI = 1
   50 READ (1,730) PP(NOI), (XX(NOI,J),J=1,NCOMP1),
     1(YY(NOI,J),J=1,NCOMP1)
C  105 FORMAT(F14.8,F10.9,F14.13)
      IF (NQNTUM .EQ. 1) XX(NOI,1) = YY(NOI,1) / XX(NOI,1)
      IF (NQNTUM .EQ. 2) GO TO 90
   60 CONTINUE
      IF (IJK .EQ. 4) PP(NOI) = PP(NOI) / 76.0 * 14.697
      IF (IJK .EQ. 3) PP(NOI) = PP(NOI) / 760.0 * 14.697
      IF (IJK .EQ. 2) PP(NOI) = PP(NOI) * 1000. / 6.895
      IF (IJK .EQ. 1) PP(NOI) = PP(NOI) * 14.697
      SX = 0.0
      SY = 0.0
      IF (PP(NOI)) 100, 70, 70
C
   70 DO 80 J = 1, NCOMP1
      SY = SY + YY(NOI,J)
   80 SX = SX + XX(NOI,J)
C
```

```
      XX(NOI,NCOMP) = 1.0 - SX
      YY(NOI,NCOMP) = 1.0 - SY
      PP(NOI) = PP(NOI) / 14.697
      WRITE (3,740) NOI, PP(NOI), (XX(NOI,J),J=1,NCOMP),
     1(YY(NOI,J),J=1,NCOMP)
      PP(NOI) = PP(NOI) * 14.697
      NOI = NOI + 1
      GO TO 50
   90 CONTINUE
      TEMP = (1. - YY(NOI,1)) / (XX(NOI,1) - YY(NOI,1))
      YY(NOI,1) = TEMP * XX(NOI,1)
      XX(NOI,1) = TEMP
      GO TO 60
  100 NOI = NOI - 1
      BK4 = 0.0
  110 READ (1,750) BK4, BK5, BK6, BK7, BK8, BK9
C
      DO 120 I = 1, NCOMP
  120 TR(I) = T / TCII(I)
C
      WRITE (3,760) (TR(I),I=1,NCOMP)
  130 READ (1,770) W(1), W(2), W(3), W(4)
C      IVPE = 0 VAPOR PRESSURE IS THE ANTOINE EQ.
C      IVPE = 1 VAPOR PRESSURE IS GOODWIN'S EQ.
C      IVPE = 2 FOR GOODWIN V.P.EQ. ( CH4)
C    IVPE=3 FOR V.P. IN ATM. AT THE SPECIFIED TEMP.
      DO 180 I = 1, NCOMP
      READ (1,650) IVPE
      IVPE = IVPE + 1
      GO TO (140, 150, 160, 170), IVPE
  140 READ (1,730) AAP, BBP, CCP
      WRITE (3,740) I, AAP, BBP, CCP
      P00(I) = AAP - BBP / (CCP + T - 273.15)
      P00(I) = 10. ** P00(I) / 760.
      IF (TR(I) .GT. 1.0) P00(I) = 10. ** (ALOG10(PCII(I))*
     1TR(I))
      WRITE (3,740) IVPE, P00(I)
      GO TO 180
  150 READ (1,730) AAP, BBP, CCP, DDP
      XP = (1. - 63.14/T) / (1. - 63.14/TCII(I))
      P00(I) = AAP * XP + BBP * XP * XP + CCP * XP ** 3
      FX = 0.
      IF (XP .LT. 1.) FX = -(1. - XP) ** 2 * ALOG(1. - XP)
      P00(I) = P00(I) + DDP * FX
      P00(I) = EXP(P00(I)) * .1233
      IF (XP .GT. 1.0) P00(I) = 10. ** (ALOG10(PCII(I))*TR(
     1I))
      WRITE (3,740) I, AAP, BBP, CCP, DDP
      WRITE (3,740) IVPE, P00(I)
      GO TO 180
  160 READ (1,730) AAP, BBP, CCP, DDP
      XP = (1. - 90.68/T) / (1. - 90.68/TCII(I))
      XXP = ABS(1. - XP)
      IF (XP .GE. 1.) DDP = 0.0
      P00(I) = AAP * XP + BBP * XP * XP + CCP * XP ** 3 +
     1DDP * XP * XXP ** 1.5
      P00(I) = EXP(P00(I)) * .1159
      IF (XP .GT. 1.0) P00(I) = 10. ** (ALOG10(PCII(I))*TR(
     1I))
      WRITE (3,740) I, AAP, BBP, CCP, DDP
```

```
      WRITE (3,740) IVPE, P00(I)
      GO TO 180
  170 READ (1,730) P00(I)
      WRITE (3,740) IVPE, P00(I)
  180 CONTINUE
C
      READ (1,780) BW, WA10, WA11, WA12, WA13
      READ (1,780) BW, WA20, WA21, WA22, WA23
      WRITE (3,620) WA10, WA11, WA12, WA13, WB10, WB11,
     1WB12, WB13
      WRITE (3,620) WA20, WA21, WA22, WA23, WB20, WB21,
     1WB22, WB23
C
      DO 190 I = 1, NOI
      IF (XX(NOI,1) .LT. .0 .OR. XX(NOI,1) .GT. 1.0)
     1GO TO 20
  190 CONTINUE
C
      DO 200 I = 1, NCOMP
      S(I) = 10. ** ((1. - SQRT(TR(I)))*(.028 + .665*W(I) -
     1.2*(1. - TR(I))))
      WAII(I) = .42748 * S(I) ** 2.5
      WBII(I) = .08664 * S(I)
      WRITE (3,790) I, S(I), I, WAII(I), I, WBII(I)
  200 CONTINUE
C
      DO 250 JJJ = 1, NCOMP
      IF (TR(JJJ) .LT. .85) GO TO 220
      II = 1
      IFF = M1
C
      DO 210 IN = II, IFF
      I = IN - II + 1
      B(I) = A0(IN) + A1(IN) * W(JJJ) + A2(IN) * W(JJJ) * W(
     1JJJ) + A3(IN) * W(JJJ) * W(JJJ) * W(JJJ)
      JN = M + 2 + IN
      J = M + 2 + I
  210 B(J) = A0(JN) + A1(JN) * W(JJJ) + A2(JN) * W(JJJ) * W(
     1JJJ) + A3(JN) * W(JJJ) * W(JJJ) * W(JJJ)
C
      IF (TR(JJJ) .GE. 1.0) WAII(JJJ) = B(1)
      IF (TR(JJJ) .GE. 1.0) WBII(JJJ) = B(5)
      IF (TR(JJJ) .GE. 1.0) GO TO 250
      WAII(JJJ) = B(1) + B(2) * (1. - TR(JJJ)) + B(3) * (1.
     1- TR(JJJ)) ** (1./3.) + B(4) * (1. - TR(JJJ)) ** (2./
     23.)
      WBII(JJJ) = B(5) + B(6) * (1. - TR(JJJ)) + B(7) * (1.
     1- TR(JJJ)) ** (1./3.) + B(8) * (1. - TR(JJJ)) ** (2./
     23.)
      GO TO 250
  220 II = 2 * (M + 2) + 1
      IFF = 2 * (M + 2) + (M + 1)
      WAII(JJJ) = 0.0
      WBII(JJJ) = 0.0
C
      DO 230 IN = II, IFF
      I = IN - II + 1
      B(I) = A0(IN) + A1(IN) * W(JJJ) + A2(IN) * W(JJJ) * W(
     1JJJ)
      JN = M + 1 + IN
```

```
      J = M + 1 + I
  230 B(J) = A0(JN) + A1(JN) * W(JJJ) + A2(JN) * W(JJJ) * W(
     1JJJ)
C
      M2 = M + 1
C
      DO 240 I = 1, M2
      JJ = I - 1
      J = M + 1 + I
      WAII(JJJ) = WAII(JJJ) + B(I) * TR(JJJ) ** JJ
  240 WBII(JJJ) = WBII(JJJ) + B(J) * TR(JJJ) ** JJ
C
  250 CONTINUE
C
      WBII(1) = 0.034 - .025 * W(1) + 0.1 / TR(1) - 0.025 /
     1TR(1) ** 2M - SRK1P
      WBII(2) = 0.034 - 0.025 * W(2) + 0.1 / TR(2) - 0.025 /
     1 TR(2) ** 2M - SRK1P
      WAII(1) = WA10 + WA11 / TR(1) + WA12 / TR(1) ** 2M -
     1SRK1P
      WAII(2) = WA20 + WA21 / TR(2) + WA22 / TR(2) ** 2M -
     1SRK1P
C
      DO 260 I = 1, NCOMP
      WAII(I) = ((1.12961961 - .7846527*W(I)) + (-.13989729
     1+ 1.0449394*W(I))/TR(I) + (.012089291 - .24821323*W(I)
     2)/TR(I)**2) ** 2 * .467123
      WBII(I) = .108762
      WAII(I) = (.35190808 - W(I)*.79946286) + (.18385964 +
     1W(I)*.92295131) / TR(I) + (-.066361596 - W(I)*.
     2194016S3) / TR(I) ** 2
      WAII(I) = (.35190808 - W(I)*.85117382) + (.18385964 +
     1W(I)*1.0010328) / TR(I) + (-.066361596 - W(I)*.
     2222659421 / TR(I) ** 2
      WBII(I) = .034 - .025 * W(I) + .1 / TR(I) - .025 / TR(
     1I) ** 2
  260 CONTINUE
C
      IF (NONH2 .EQ. 100) GO TO 270
      WAII(1) = 0.503476 - 0.00876 / TR(1) - 0.028115 / TR(
     11) ** 2
      WBII(1) = 0.100278 - 0.0233989 / TR(1) + 0.032129 /
     1TR(1) ** 2
  270 WRITE (3,800) (WAII(I),I=1,NCOMP), (WBII(I),I=1,NCOMP)
     1, (S(I),I=1,NCOMP)
  280 CONTINUE
  290 KIJ = KIJ + 2
      IF (KIJ .GE. 3) GO TO 20
  300 WRITE (3,810) IJK, KIJ, CINI, CFIN
C      GOLDEN SECTION SEARCH TECHNIQUE FOR FINDING THE VALUE K12 SUCH
      KPRINT = 0
C      THAT SDP IS MINIMUM
      TOL = .00005
      TOL = .0001
      I1 = 0
      I2 = 0
      I3 = 0
      I4 = 0
      BK4 = CFIN
      BK1 = CINI
```

```
      NOS = 1
      IF (I1 .EQ. 1) GO TO 320
      PPP = PSI
      CALL PYINIT(BK1)
C
      DO 310 K = 1, NOI
      CALL XCALC
      CALL YVCALC
  310 CONTINUE
C
      PSI = PPP
      DP1 = SDP
      DY1 = DP1
      DY1 = SSDY
      I1 = 1
  320 IF (I2 .EQ. 1) GO TO 340
      BK2 = BK1 + (3.0 - SQRT(5.0)) / 2.0 * (BK4 - BK1)
      PPP = PSI
      SDP = 0.0
      SDV = 0.0
      SSDY = 0.0
      SADY = 0.0
      SSDK(1) = 0.0
      SSDK(2) = 0.0
      CALL PYINIT(BK2)
C
      DO 330 K = 1, NOI
      CALL XCALC
      CALL YVCALC
  330 CONTINUE
C
      PSI = PPP
      DP2 = SDP
      DY2 = DP2
      DY2 = SSDY
      I2 = 1
C3002 IF (ABS(DP2 - DP1).LE. TOL .OR. ABS(BK2-BK1) .LE. TOL) GO TO 7001
C3002 IF (ABS(DY2 - DY1).LE. TOL .OR. ABS(BK2-BK1) .LE. TOL) GO TO 7001
  340 IF (ABS(DY2 - DY1) .LE. TOL .OR. ABS(BK2 - BK1) .LE.
     1TOL) GO TO 440
C5000 IF (DP2 .GT. DP1) GO TO 4001
  350 IF (DY2 .GT. DY1) GO TO 410
  360 IF (I3 .EQ. 1) GO TO 380
      BK3 = BK4 - (3.0 - SQRT(5.0)) / 2.0 * (BK4 - BK1)
      PPP = PSI
      SDP = 0.0
      SDV = 0.0
      SSDY = 0.0
      SADY = 0.0
      SSDK(1) = 0.0
      SSDK(2) = 0.0
      CALL PYINIT(BK3)
C
      DO 370 K = 1, NOI
      CALL XCALC
      CALL YVCALC
  370 CONTINUE
C
      PSI = PPP
      DP3 = SDP
```

```
      DY3 = DP3
      DY3 = SSDY
      I3 = 1
C3003 IF (ABS(DP3 - DP2).LE. TOL .OR. ABS(BK3-BK2) .LE. TOL) GO TO 7002
C3003 IF (ABS(DY3 - DY2).LE. TOL .OR. ABS(BK3-BK2) .LE. TOL) GO TO 7002
 380 IF (ABS(DY3 - DY2) .LE. TOL .OR. ABS(BK3 - BK2) .LE.
     1TOL) GO TO 470
C     IF (DP3 .GT. DP2) GO TO 4003
      IF (DY3 .GT. DY2) GO TO 420
      IF (I4 .EQ. 1) GO TO 400
      PPP = PSI
      SDP = 0.0
      SDV = 0.0
      SSDY = 0.0
      SADY = 0.0
      SSDK(1) = 0.0
      SSDK(2) = 0.0
      CALL PYINIT(BK4)
C
      DO 390 K = 1, NOI
      CALL XCALC
      CALL YVCALC
 390 CONTINUE
C
      PSI = PPP
      DP4 = SDP
      DY4 = DP4
      DY4 = SSDY
C3004 IF (ABS(DP4 - DP3).LE. TOL .OR. ABS(BK4-BK3) .LE. TOL) GO TO 7003
C3004 IF (ABS(DY4 - DY3).LE. TOL .OR. ABS(BK4-BK3) .LE. TOL) GO TO 7003
 400 IF (ABS(DY4 - DY3) .LE. TOL .OR. ABS(BK4 - BK3) .LE.
     1TOL) GO TO 480
      IF (DY3 .GT. DY4) GO TO 430
      BK1 = BK2
      DP1 = DP2
      DY1 = DY2
      BK2 = BK3
      DP2 = DP3
      DY2 = DY3
      I3 = 0
      IF (NOS .GE. 30) GO TO 490
      NOS = NOS + 1
      GO TO 360
 410 BK4 = BK2
      DP4 = DP2
      DY4 = DY2
      I4 = 1
      I2 = 0
      I3 = 0
      IF (NOS .GE. 30) GO TO 490
      NOS = NOS + 1
      GO TO 320
 420 BK4 = BK3
      DP4 = DP3
      DY4 = DY3
      BK3 = BK2
      DP3 = DP2
      DY3 = DY2
      I4 = 1
      I2 = 0
```

```
      IF (NOS .GE. 30) GO TO 490
      NOS = NOS + 1
      GO TO 320
  430 BK1 = BK3
      DP1 = DP3
      DY1 = DY3
      I2 = 0
      I3 = 0
      IF (NOS .GE. 30) GO TO 490
      NOS = NOS + 1
      GO TO 320
  440 BK4 = BK2
      DP4 = DP2
      DY4 = DY2
      I4 = 1
      I3 = 0
  450 BK2 = BK1 + (3.0 - SQRT(5.0)) / 2.0 * (BK4 - BK1)
      PPP = PSI
      SDP = 0.0
      SDV = 0.0
      SSDY = 0.0
      SADY = 0.0
      SSDK(1) = 0.0
      SSDK(2) = 0.0
      CALL PYINIT(BK2)
C
      DO 460 K = 1, NOI
      CALL XCALC
      CALL YVCALC
  460 CONTINUE
C
      PSI = PPP
      DP2 = SDP
      DY2 = DP2
      DY2 = SSDY
      I2 = 1
C      IF (ABS(DP2 - DP1).LE. TOL .OR. ABS(BK2-BK1) .LE. TOL) GO TO 7000
C      IF (ABS(DY2 - DY1).LE. TOL .OR. ABS(BK2-BK1) .LE. TOL) GO TO 7000
      IF (ABS(DY2 - DY1) .LE. TOL .OR. ABS(BK2 - BK1) .LE.
     1TOL) GO TO 500
      IF (NOS .GE. 30) GO TO 490
      NOS = NOS + 1
      GO TO 350
  470 BK1 = BK2
      DP1 = DP2
      DY1 = DY2
      I1 = 1
      BK4 = BK3
      DP4 = DP3
      DY4 = DY3
      I4 = 1
      I3 = 0
      GO TO 450
  480 BK1 = BK3
      DP1 = DP3
      DY1 = DY3
      I1 = 1
      I3 = 0
      GO TO 450
  490 WRITE (3,820) BK1, BK2, BK3, BK4, DP1, DP2, DP3, DP4,
```

```
     1DY1, DY2, DY3, DY4
  500 WRITE (3,830) BK4, DP4, DY4, NOS
      CALL PYINIT(BK4, BK5, BK6, BK7, BK8, BK9)
      WRITE (3,840) ((CORRL(I,J),I=1,NCOMP),J=1,NCOMP)
      WRITE (3,850)
C
      DO 510 J = 1, NCOMP
      EDYAV(J) = 0.0
      DYAV(J) = 0.0
      SSDK(J) = 0.0
  510 CONTINUE
C
      EDPAV = 0.0
      DPAV = 0.0
      SSDY = 0.0
      SADY = 0.0
      SDV = 0.0
      KPRINT = 1
C
      DO 580 K = 1, NOI
      CALL XCALC
      WRITE (3,860) VV, (X(I),I=1,NCOMP), (XX(K,I),I=1,
     1NCOMP)
C      IF ( YY(K, 1) .LE. 0.0) GO TO 500
      DO 520 I = 1, NCOMP
  520 WRITE (3,870) I, PHIV(I), I, Y(I)
C
      DO 530 I = 1, NCOMP
  530 WRITE (3,880) I, PHIK(I), I, P1(I)
C
      WRITE (3,540) T, P, K, PP(K), DP, EP
  540 FORMAT(3X,'T=',F10.4,3X,'P=',F10.4,3X,'PP(',I3,')=',
     1F10.4,3X'DX=',F10.4,20X,'EX=',F10.4,'%')
      CALL YVCALC
      WRITE (3,890) (X(I),I=1,3), (Y(I),I=1,3),
     1(EDY(I),I=1,3), DP, EDP
      WRITE (3,900) DY(1), DY(2), DY(3)
C
      DO 550 J = 1, NCOMP
  550 WRITE (3,910) COMPA(J), COMPB(J), COMPC(J), COMPD(J),
     1PG, J, X(J), J, GAMMB(J)
C
      WRITE (3,920) (EXG1(I),I=1,NCOMP)
      WRITE (3,930) EXGT
C
      DO 560 J = 1, NCOMP
  560 WRITE (3,940) COMPA(J), COMPB(J), COMPC(J), COMPD(J),
     1P, J, YX(J), J, GAMMA(J)
C
      WRITE (3,920) (EXG(I),I=1,NCOMP)
C
      DO 570 J = 1, NCOMP
      DX(J) = X(J) - XX(K,J)
      WRITE (3,950) COMPA(J), COMPB(J), COMPC(J), COMPD(J),
     1J, X(J), K, J, XX(K,J), J, DX(J)
  570 WRITE (3,960) COMPA(J), COMPB(J), COMPC(J), COMPD(J),
     1J, X(J), J, Y(J), K, J, YY(K,J), J, DY(J), J,
     2PPVOL(J), J, PHI(J), J, PHIV(J), SUMY, ITER
C
      WRITE (3,970) VV, VVL1, DV
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```
 580 CONTINUE
C
     WRITE (3,980) DPAV, EDPAV, (DYAV(J),J=1,3),
    1(EDYAV(J),J=1,3)
     SADY = SADY / NOI / NCOMP
     SSDYA = SADY * NOI
     WRITE (3,990) BK4, SDP, SADY, SSDYA
C
     DO 590 J = 1, NCOMP
     ESSDK(J) = SADK(J) * 100.0
 590 CONTINUE
C     ESSDK(J) = SQRT (SUM OF (DK/K)**2 ) *100./NO OF COMPONENT
     WRITE (3,1000) (ESSDK(J),J=1,NCOMP)
     PSI = PPP
     GO TO 290
 600 RETURN
 610 FORMAT (2I5)
 620 FORMAT (1H0, 4D20.8)
 630 FORMAT (20A4)
 640 FORMAT (1H1, 20A4)
 650 FORMAT (3I5, F10.4)
 660 FORMAT (1H0, 'INPUT DATA:'//)
 670 FORMAT (1H0, 6X, 'PCII', 8X, 'VCII', 8X, 'TCII', 8X,
    1'W', 11X, 'WAII', 8X, 'WBII', 4X, 'COMPONENT', 3X,
    2'AMOLWT'//)
 680 FORMAT (4F8.4, F8.5, F9.6, 4A4)
 690 FORMAT (1H , 6F12.6, 3X, 4A4)
 700 FORMAT (2F10.4, 5I2)
 710 FORMAT (1H0, 'T=', F10.4, 3X, 'DEG. OF K', 3X, 'P=',
    1F10.4, 3X, ' ATM')
 720 FORMAT (2I5, 2F10.4)
 730 FORMAT (8F10.0)
 740 FORMAT (1H , I3, 1F12.5, ' P IN ATM ', 8F12.5)
 750 FORMAT (8F10.4)
 760 FORMAT (1H0, 'TR(1)=', F6.4, '  TR(2)=', F6.4, 5X,
    1'TR(3)=',*F6.4, 5X, 'TR(4)=', F6.4)
 770 FORMAT (4D20.8)
 780 FORMAT (6F13.8)
 790 FORMAT (1H0, 'S(', I2, ')=', F10.5, 'WAII(', I2, ')=',
    1F10.7, 'WBII(', I2, ')=', F10.7)
 800 FORMAT (1H0, 9F12.8)
 810 FORMAT (1H0, 3X, 'IJK=', I5, 3X, 'KIJ=', I5, 3X,
    1'CINI=', F10.4, 3X, 'CFIN=', F10.4//)
 820 FORMAT (1H0, 'K12 DID NOT CONVERGE,  THE FOLLOWING RESULT IS NOT O
    1PTIMUM '/8(2X,E13.7)/4(2X,E13.7))
 830 FORMAT (1H0, ' THE VALUE OF K12 IS ', F10.8, '    THE CORRESPONDIN
    1GD.P. IS ', E14.8/' AND THE CORRESPONDING D.Y. IS ',
    2E14.8, ' NO. OF ITERATIONS = ', I4)
 840 FORMAT (1H0, 9(3X,F10.5))
 850 FORMAT (1H0, 'RESULT:')
 860 FORMAT (1H0, 3X, 'VV=', F14.4, 3X, 'X=', 5F10.6, //,
    119X, '  XX=  ', 5F10.6)
 870 FORMAT (1H , 3X, 'PHIV(', I2, ')=', F10.4, 3X, 'Y(',
    1I2, ')=', F10.4)
 880 FORMAT (1H , 3X, 'PHIK(', I2, ')=', F10.4, 3X, 'P1(',
    1I2, ')=', F10.4)
 890 FORMAT (1H , '  X(1)=', F10.8, '  X(2)=', F10.8,
    1'  X(3)=', F10.8, '  Y(1)=', F10.8, '  Y(2)=', F10.8,
    2'  Y(3)=', F10.8/'  EDY(1)=', E12.6, '%  EDY(2)=',
    3E12.6, '%  EDY(3)=', E12.6, '%  DP=', E12.6, '  EDP=',
```

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     4E12.6, '%')
  900 FORMAT (1H , '  DY(1)=', E12.6, '  DY(2)=', E12.6,
     1'  DY(3)=', E12.6)
  910 FORMAT (10X, 4A4, 3X, 'P=', F10.4, 3X, 'X(', I2, ')=',
     1F10.4, 3X, 'GAMMB(', I2, ')=', F10.4)
  920 FORMAT (10X, 'LN @     EXG(I)=', 3F10.4)
  930 FORMAT (110X, 'EXGT=', F10.4)
  940 FORMAT (10X, 4A4, 3X, 'P=', F10.4, 3X, 'YX(', I2,
     1')=', F10.4, 3X, 'GAMMA(', I2, ')=', F10.4)
  950 FORMAT (1H0, 3X, 4A4, 3X, 'X(', I2, ')=', F10.6, 3X,
     13X, 'XX(', I4, I2, ')=', F10.8, 3X, 'DX(', I2, ')=',
     2E12.4)
  960 FORMAT (1H0, 3X, 4A4, 3X, 'X(', I2, ')=', F10.6, 3X,
     1'Y(', I2, ')=', F10.8, 3X, 'YY(', I4, I2, ')=', F10.8,
     23X, 'DY(', I2, ')=', E12.4/21X, 'PPVOL(', I2, ')=',
     3F10.4, 3X, 'PHI(', I2, ')=', F10.4, 3X, 'PHIV(', I2,
     4')=', F10.4, 3X, 'SUMY=', F10.4, 3X, 'ITER=', I4)
  970 FORMAT (15X, 'VV=', F10.4, 3X, 'VVL1=', F10.4, 3X,
     1'DV=', E12.4)
  980 FORMAT (1H0, '  DPAV=', E12.6, 'EDPAV=', E12.6,
     1'%  DYAV(1)=', E12.6, '  DYAV(2)=', E12.6,
     2'  DYAV(3)=', E12.6/'  EDYAV(1)=', E12.6,
     3'%  EYDAV(2)=', E12.6, '%  EDYAV(3)=', E12.6, '%')
  990 FORMAT (1H0, 3X, ' K12= ', E14.8, 3X, ' SDP= ', E14.8,
     13X, ' DYAV= ', E14.8, 3X, 'SUM OF DY=', E14.8)
 1000 FORMAT (5X, '  % OF K1 IN RMS = ', F10.4, 10X,
     1' % OF K2 IN RMS = ', F10.4, 5F10.4//)
      END
C
C
C
C
C
C
C
C     ******************************************************************
      SUBROUTINE PYINIT(BK, BK1, BK2, BK3, BK10, BK11)
C
C     ******************************************************************
C     PREDICT P-X-Y CURVE
C     MODIFIED REDLICH-KWONG EQUATION OF STATE IS USED FOR MULTI-
C     COMPONENT SYSTEMS CALCULATION FROM THE PURE COMPONENT PROPERTIES
C     ONE OF THE COMPONENTS IS AT ITS HYPERCRITICAL STATE
C     ******************************************************************
      DIMENSION TITLE(20), PCII(5), VCII(5), TCII(5), W(5),
     1WAII(5), WBII(5), COMPA(5), COMPB(5), COMPC(5),
     2FREFER(5), PHI(5), PHIV(5), PHIJ(5), PHIK(5), P1(5),
     3PTVOL(5), PPVOL(5), VVV(5), CORRL(5,5), X(5), Y(5),
     4SSDK(5), VC(5), TC(5), SADK(5), EXG(5), EXGF(5),
     5YX(5), VVL(5), DY(5), PVOL(5), GAMMB(5), GAMMA(5),
     6AMOLWT(5), PC(5), XX(50,5), YY(50,5), PP(50), EXG1(5),
     7COMPD(5)
      COMMON /FIRST/ TC, PC, VCII, W, AMOLWT, T, P, R,
     1NCOMP, NQNTUM /SECOND/ Z, A, MTYPE /THIRD/ PCII, TCII,
     2WAII, WBII, PSI, PP, XX, YY, IJK, KIJ, NOI, ITER, K,
     3CORRL, EXGT, PPVOL, PHI, PHIV, VV, VVL1, DV, P1, PPP,
     4Y /FOURTH/ SDP, EP, DP, SSDY, SDY, SDV, PHIK, GAMMA,
     5EXG, X, SUMY, GAMMB, PG, RT, PF, FREFER, YX, DY, EXG1
     6/FIFTH/ DPAV, EDYAV, EDY, EDP, EDPAV, DYAV, SSDK,
     7SADY, SADK
```

```
      SDP = 0.0
      SDV = 0.0
      SSDY = 0.0
      SADY = 0.0
C
      DO 10 J = 1, NCOMP
   10 SSDK(J) = 0.0
C      PF = REFERENCE PRESSURE OF 1000 LB OR 1000./14.697 ATMOSPHERE
C      SINCE ONE ATMOSPHERE = 14.697 LB
      PF = 1000 / 14.697
      PPP = PSI
      PSI = PSI / 14.697
C
      DO 20 I = 1, NCOMP
   20 CORRL(I,I) = 0.0
C
      CORRL(1,2) = BK
      CORRL(2,1) = CORRL(1,2)
      CORRL(2,3) = BK1
      CORRL(3,2) = CORRL(2,3)
      CORRL(1,3) = BK2
      CORRL(3,1) = CORRL(1,3)
      CORRL(1,4) = BK3
      CORRL(4,1) = BK3
      CORRL(2,4) = BK10
      CORRL(4,2) = CORRL(2,4)
      CORRL(3,4) = BK11
      CORRL(4,3) = CORRL(3,4)
C      SET UP THE NECESSARY INFORMATION FOR THE SUPPORT SUBROUTINE
      RETURN
      END
C
C ***********************************************************************
      SUBROUTINE XCALC
C
C ***********************************************************************
      DIMENSION TITLE(20), PCII(5), VCII(5), TCII(5), W(5),
     1WAII(5), WBII(5), COMPA(5), COMPB(5), COMPC(5),
     2FREFER(5), PHI(5), PHIV(5), PHIJ(5), PHIK(5), P1(5),
     3PTVOL(5), PPVOL(5), VVV(5), CORRL(5,5), X(5), Y(5),
     4SSDK(5), VC(5), TC(5), SADK(5), EXG(5), EXGF(5),
     5YX(5), VVL(5), DY(5), PVOL(5), GAMMB(5), GAMMA(5),
     6AMOLWT(5), PC(5), XX(50,5), YY(50,5), PP(50), EXG1(5),
     7COMPD(5)
      DIMENSION EDYAV(5), DYAV(5), EDY(5), SDK(5)
      DIMENSION P00(5)
      COMMON /FIRST/ TC, PC, VCII, W, AMOLWT, T, P, R,
     1NCOMP, NQNTUM /SECOND/ Z, A, MTYPE /THIRD/ PCII, TCII,
     2WAII, WBII, PSI, PP, XX, YY, IJK, KIJ, NOI, ITER, K,
     3CORRL, EXGT, PPVOL, PHI, PHIV, VV, VVL1, DV, P1, PPP,
     4Y /FOURTH/ SDP, EP, DP, SSDY, SDY, SDV, PHIK, GAMMA,
     5EXG, X, SUMY, GAMMB, PG, RT, PF, FREFER, YX, DY, EXG1
     6/FIFTH/ DPAV, EDYAV, EDY, EDP, EDPAV, DYAV, SSDK,
     7SADY, SADK /SIXTH/ PN /SEVEN/ P00, KPRINT
      IPP = 0
      ITER = 0
C
      DO 10 I = 1, NCOMP
      PC(I) = PCII(I)
      VC(I) = VCII(I)
```

```
   10 TC(I) = TCII(I)
C
      RT = R * T
      P = PF
C      USE TO CALCULATE THE MOLAL VOLUMN VL, FUGACITY COEFFICIENT PHI,
C      PARTIAL MOLAL VOLUMN PPVOL AND FUGACITY FREFER OF LIQUID PHASE
C      FOR PURE COMPONENT AT REFERENCE PRESSURE
   20 DO 40 I = 1, NCOMP
C
      DO 30 J = 1, NCOMP
   30 X(J) = 0.0
C
      X(I) = 1.0
      CALL SUPT(VL, PHI, PPVOL, WAII, WBII, CORRL, X, 1,
     1KIJ)
      IF (PHI(1) .GE. 10000.) WRITE (3,420)
      IF (PHI(1) .GE. 10000.) GO TO 40
      FREFER(I) = PHI(I) * P
   40 CONTINUE
C      ****************************
C      USE TO CALCULATE VAPORMOLE FRACTIONS Y(I) FOR MIXTURE ASSUMING
C       CORRESPONDING FUGACITY COEFFICIENTS PHIV(I) ARE 1. ( INITIAL
C GUESS)
      P = 0.
C
      DO 50 I = 1, NCOMP
   50 P = P00(I) * XX(K,I) + P
C
      P = PP(K) / 14.697
      LLL = 1
      X(1) = XX(K,1) * 0.995
      GO TO 70
   60 X(1) = XX(K,1) * 1.005
      LLL = LLL + 1
      IF (LLL .EQ. 3) GO TO 400
   70 IF (NCOMP .EQ. 3) GO TO 80
      IF (NCOMP .EQ. 4) GO TO 90
      X(2) = 1 - X(1)
      GO TO 100
   80 X(3) = XX(K,3)
      X(2) = 1 - X(1) - X(3)
      GO TO 100
   90 X(3) = XX(K,3)
      X(4) = XX(K,4)
      X(2) = 1 - X(1) - X(4) - X(3)
  100 NCOMP1 = NCOMP - 1
C
      DO 120 J = 1, NCOMP1
      I = J + 1
C
      DO 110 I = J, NCOMP
      IF (TC(J) .GT. TC(I)) GO TO 120
  110 CONTINUE
C
      GO TO 130
  120 CONTINUE
C
      J = NCOMP
C
  130 DO 140 II = 1, NCOMP
```

```
 140 Y(II) = .0001 / NCOMP1
C
     Y(1) = 0.9999
     CALL SUPT(VL, PHI, PPVOL, WAII, WBII, CORRL, X, 1,
    1KIJ)
     IF (PHI(1) .GE. 10000.) GO TO 320
     SUMY1 = 1.
 150 CALL SUPT(VV, PHIV, PVOL, WAII, WBII, CORRL, Y, 0,
    1KIJ)
     IF (PHI(1) .GE. 10000.) GO TO 330
C     SET UP THE LOOP TO FIND Y(I), SUCH THAT SUMY IS EQUAL TO SUMY1
C     ,THE SUMY OBTAINED IN IMMEDIATE PRECEDING CALCULATION, I. E. TEST
 160 CALL SUPT(VL, PHI, PPVOL, WAII, WBII, CORRL, X, 1,
    1KIJ)
     IF (PHI(1) .GE. 10000.) GO TO 340
C     FOR STABILITY BY CHANGING Y(I) AND TEST FOR SUMY = 1.0
 170 SUMY = 0.0
C
     DO 180 J = 1, NCOMP
     Y(J) = PHI(J) * X(J) / PHIV(J)
 180 SUMY = SUMY + Y(J)
C
     IF (KPRINT .EQ. 0) GO TO 190
C     WRITE(3, 900) P,Y(1),Y(2),SUMY,(PHI(J),J=1,2),(PHIV(J),J=1,2)
 190 CONTINUE
     IF (ABS(SUMY1 - SUMY) - 1.0E-4) 230, 230, 200
C     FIND A NEW VALUE OF Y(I) AND SUMY USING Y(I) IF SUMY1 IS DIFFERENT
C     FROM SUMY
 200 IF (ITER - 100) 210, 360, 360
C  15 IF (ITER - 50) 16, 21, 21
 210 ITER = ITER + 1
C
     DO 220 I = 1, NCOMP
 220 Y(I) = Y(I) / SUMY
C
     SUMY1 = SUMY
     CALL SUPT(VV, PHIV, PVOL, WAII, WBII, CORRL, Y, 0,
    1KIJ)
     IF (PHI(1) .GE. 10000.) GO TO 350
     GO TO 170
C     Y(I) IS STABLE. NOW WE HAVE TO TEST FOR SUMY = 1. BY CHANGING P
C     CHANGE X
 230 CONTINUE
     IF (IPP .NE. 0) GO TO 250
     SUMY00 = SUMY
     IPP = IPP + 1
     X0 = X(1)
     X(1) = X0 * 0.995
     ITER = ITER + 1
C
     DO 240 I = 1, NCOMP
 240 Y(I) = Y(I) / SUMY
C
     GO TO 150
C     ************************************
 250 IF (ABS((X0 - X(1))/X(1)) .LE. .0000001)
    1GO TO 360
     SUMY1 = SUMY00
     IF (ABS(SUMY - SUMY1) .LE. 1.0E-5) GO TO 360
     XN = X(1) - (X0 - X(1)) / (SUMY - SUMY1) * (1. - SUMY)
```

```
      IF (XN .LT. 0.0 .OR. XN .GT. 1.0) GO TO 60
  260 IF (ABS((XN - X(1))/XN) .LE. .0000001) GO TO 360
      X0 = X(1)
      X(1) = XN
      IF (NCOMP .EQ. 3) GO TO 270
      IF (NCOMP .EQ. 4) GO TO 280
      X(2) = 1. - X(1)
      GO TO 290
  270 X(2) = XX(K,2)
      X(3) = 1. - X(1) - X(2)
      GO TO 290
  280 X(2) = XX(K,2)
      X(3) = XX(K,3)
      X(4) = 1. - X(1) - X(2) - X(3)
  290 SUMY00 = SUMY
      ITER = ITER + 1
C
      DO 300 I = 1, NCOMP
  300 Y(I) = Y(I) / SUMY
C
      GO TO 150
  310 IF (KPRINT .EQ. 0) GO TO 380
      WRITE (3,440) PHI(1), PHI(2), PHIV(1), PHIV(2), P1(1),
     1P1(2), SUMP, P
      GO TO 380
  320 WRITE (3,450)
      GO TO 380
  330 WRITE (3,460)
      GO TO 380
  340 WRITE (3,470)
      GO TO 380
  350 WRITE (3,480)
      GO TO 380
  360 SUMP = 0.0
C
      DO 370 I = 1, NCOMP
      P1(I) = PHI(I) * X(I) * P / PHIV(I)
  370 SUMP = SUMP + P1(I)
C
      IF (ABS(P - SUMP) .GT. .1) GO TO 310
      IF (ABS(SUMY - 1.0) .GT. 1.0E-4) GO TO 310
C     GATHER STATISTICS AND PRINT OUT RESULTS
  380 P = P * 14.697
C     DP=DX
      DO 390 KKK = 1, NCOMP
      DP = X(KKK) - XX(K,KKK)
      EDP = (DP/XX(K,KKK)) * 100.
C     SDP = SDP + ABS(DP)
      SDP = ((SDP*(K - 1)*SDP + ABS(EDP)**2)/K) ** .5
      DPAV = (DPAV*(K - 1) + ABS(DP)) / K
      EDPAV = (EDPAV*(K - 1) + ABS(EDP)) / K
      EP = (DP/XX(K,KKK)) * 100.
  390 CONTINUE
C
      IF (LLL .EQ. 1) GO TO 410
  400 WRITE (3,490) LLL
  410 RETURN
  420 FORMAT (1H , 'FREFER')
  430 FORMAT (1H , 8F14.7)
  440 FORMAT (1H , ' THIS METHOD DID NOT  PRODUCE ANSWER'/1H0,
```

```
   18E13.7)
 450 FORMAT (1H , 'ZL =NEGATIVE VALUE')
 460 FORMAT (1H , 'ZV =NEGATIVE VALUE')
 470 FORMAT (1H , 'ZL1=NEGATIVE VALUE')
 480 FORMAT (1H , 'ZV1=NEGATIVE VALUE')
 490 FORMAT (5X, I2, 'COMPONENT 1 LESS THAN ZERO OR GREATER THAN 1'
    1)
     END
C
C ***************************************************************************
     SUBROUTINE YVCALC
C
C ***************************************************************************
     DIMENSION TITLE(20), PCII(5), VCII(5), TCII(5), W(5),
    1WAII(5), WBII(5), COMPA(5), COMPB(5), COMPC(5),
    2FREFER(5), PHI(5), PHIV(5), PHIJ(5), PHIK(5), P1(5),
    3PTVOL(5), PPVOL(5), VVV(5), CORRL(5,5), X(5), Y(5),
    4SSDK(5), VC(5), TC(5), SADK(5), EXG(5), EXGF(5),
    5YX(5), VVL(5), DY(5), PVOL(5), GAMMB(5), GAMMA(5),
    6AMOLWT(5), PC(5), XX(50,5), YY(50,5), PP(50), EXG1(5),
    7COMPD(5), SDK(5), RK(5), RKK(50,5)
     DIMENSION EDYAV(5), DYAV(5), EDY(5)
     COMMON /FIRST/ TC, PC, VCII, W, AMOLWT, T, P, R,
    1NCOMP, NQNTUM /SECOND/ Z, A, MTYPE /THIRD/ PCII, TCII,
    2WAII, WBII, PSI, PP, XX, YY, IJK, KIJ, NOI, ITER, K,
    3CORRL, EXGT, PPVOL, PHI, PHIV, VV, VVL1, DY, P1, PPP,
    4Y /FOURTH/ SDP, EP, DP, SSDY, SDY, SDV, PHIK, GAMMA,
    5EXG, X, SUMY, GAMMB, PG, RT, PF, FREFER, YX, DY, EXG1
    6/FIFTH/ DPAV, EDYAV, EDY, EDP, EDPAV, DYAV, SSDK,
    7SADY, SADK
     P = P / 14.697
     PP(K) = PP(K) / 14.697
  10 CONTINUE
C     GATHER ANOTHER STATISTICS
  20 CONTINUE
C
     DO 40 J = 1, NCOMP
     SDY = 0.0
     SDK(J) = 0.0
     DY(J) = Y(J) - YY(K,J)
     SDY = SDY + ABS(DY(J))
     IF (YY(K,J) .LE. 0.0) EDY(J) = 0.0
     IF (YY(K,J) .LE. 0.0) GO TO 30
     EDY(J) = (DY(J)/YY(K,J)) * 100.
     RK(J) = Y(J) / X(J)
     RKK(K,J) = YY(K,J) / XX(K,J)
     SDK(J) = SDK(J) + (RK(J)/RKK(K,J) - 1.0) ** 2
     SSDK(J) = SSDK(J) + SDK(J)
  30 CONTINUE
     EDYAV(J) = (EDYAV(J)*(K - 1) + ABS(EDY(J))) / K
     DYAV(J) = (DYAV(J)*(K - 1) + ABS(DY(J))) / K
     SADK(J) = (SSDK(J)**0.5) / K
  40 SADY = SADY + SDY
C
     SSDY = ((2*(K - 1)*SSDY)**2 + SADK(1)**2 + SADK(2)**2)
    1 ** 0.5 / (2*K)
C     SSDY = SADK (1)
     PSI = P
     PP(K) = PP(K) * 14.697
     GO TO 100
```

```
   50 WRITE (3,110)
      GO TO 100
   60 WRITE (3,120)
      GO TO 100
   70 WRITE (3,130)
      GO TO 100
   80 WRITE (3,140)
      GO TO 100
   90 WRITE (3,150)
  100 RETURN
  110 FORMAT (1H , 'ZL = NEGATIVE')
  120 FORMAT (1H , 'ZV = NEGATIVE')
  130 FORMAT (1H , 'ZL1= NEGATIVE')
  140 FORMAT (1H , 'ZV1= NEGATIVE')
  150 FORMAT (1H , 'ZVF=NEGATIVE')
      END
C     ***********************************************************************
      SUBROUTINE CUBEQN
C    SOLVES CUBIC EQUATION OF REDLICH-KWONG EQUATION
C      FOR COMPRESSIBILITY FACTORS
C     ***********************************************************************
      IMPLICIT REAL*8(A - H,O - Z)
      DIMENSION A(4), Z(3), B(3)
      COMMON /SECOND/ Z, A, MTYPE
   10 CONTINUE
      B(1) = A(2) / A(1)
      B1OV3 = B(1) / 3.0
      B(2) = A(3) / A(1)
      B(3) = A(4) / A(1)
      ALF = B(2) - B(1) * B1OV3
      ALFOV3 = ALF / 3.0
      BET = 2.0 * B1OV3 ** 3 - B(2) * B1OV3 + B(3)
      BETOV2 = BET / 2.0
      CUAOV3 = ALFOV3 ** 3
      SQBOV2 = BETOV2 ** 2
      DEL = SQBOV2 + CUAOV3
      IF (DEL) 100, 20, 50
   20 MTYPE = 0.0
      GAM = DSQRT(-ALFOV3)
      IF (BET) 40, 40, 30
   30 Z(1) = -2.0 * GAM - B1OV3
      Z(2) = GAM - B1OV3
      Z(3) = Z(2)
      GO TO 140
   40 Z(1) = 2.0 * GAM - B1OV3
      Z(2) = -GAM - B1OV3
      Z(3) = Z(2)
      GO TO 140
   50 MTYPE = 1
      EPS = DSQRT(DEL)
      TAU = -BETOV2
      RCU = TAU + EPS
      SCU = TAU - EPS
      SIR = 1.0
      SIS = 1.0
      IF (RCU) 60, 70, 70
   60 SIR = -1.0
   70 IF (SCU) 80, 90, 90
   80 SIS = -1.0
   90 R = SIR * (SIR*RCU) ** 0.33333333
```

```
      S = SIS * (SIS*SCU) ** 0.33333333
      Z(1) = R + S - B1OV3
      Z(2) = -(R + S) / 2.0 - B1OV3
      Z(3) = 0.86602540 * (R - S)
      GO TO 140
  100 MTYPE = -1
      QUOT = SQBOV2 / CUAOV3
      ROOT = DSQRT(-QUOT)
      IF (BET) 120, 110, 110
  110 PEI = (1.5707963 + DATAN(ROOT/DSQRT(1.0 - ROOT**2))) /
     1 3.0
      GO TO 130
  120 PEI = DATAN(DSQRT(1.0 - ROOT**2)/ROOT) / 3.0
  130 FACT = 2.0 * DSQRT(-ALFOV3)
      Z(1) = FACT * DCOS(PEI) - B1OV3
      PEI2 = PEI + 2.0943951
      Z(2) = FACT * DCOS(PEI2) - B1OV3
      PEI4 = PEI + 4.1887902
      Z(3) = FACT * DCOS(PEI4) - B1OV3
  140 RETURN
      END
C
C *********************************************************************
      SUBROUTINE SUPT(VL, PHI, PVOL, C1RKL, C2RKL, CORRL, X,
     1LV, KIJ)
C
C
C *********************************************************************
C      EVALUTION OF SUPPORTING PROPERTIES,THE FUGACITY AND THE
C      PARTIAL MOLAL VOLUME OF FLUID MIXTURES.
C      SHINN-DER CHANG, DEPT.OF CHEM.ENG., UNIV. OF OTTAWA.
      DOUBLE PRECISION A, Z
      DIMENSION TC(5), PC(5), VCO(5), W(5), AMOLWT(5),
     1ARKL(5,5), BRKL(5), WIJ(5,5), TCOIJ(5,5), TCIJ(5,5),
     2A(4), Z(3), C1RKL(5), C2RKL(5), PVOL(5), PHI(5),
     3CORRL(5,5), X(5), A2I(5), BI(5), AIRKL(5)
      DIMENSION ZZ(3)
      DIMENSION TC1(5), VC1(5), PC1(5), TC11(5,5),
     1VC11(5,5), WAII(5), WBII(5), PCII(5), TCII(5)
      DIMENSION P00(5)
      COMMON /FIRST/ TC, PC, VCO, W, AMOLWT, T, P, R, NCOMP,
     1NQNTUM /SECOND/ Z, A, MTYPE /THIRD/ PCII, TCII, WAII,
     2WBII /SEVEN/ P00, KPRINT
C      LV=0, FOR VAPOR PHASE.
C      LV=1, FOR LIQUID PHASE.
C      KIJ=1, TCIJ=SQRT(TCI*TCJ)*(1.-KIJ)
C      KIJ=2, AIJ=SQRT(AI*AJ)*(1.-KIJ)
C      KIJ=3, AIJ=(AI*KIJ+(1.-KIJ)*AJ)/2.
      RT = R * T
      WA0 = 0.42748
      WB0 = 0.08664
C
      DO 10 I = 1, NCOMP
      ARKL(I,I) = C1RKL(I) * R * R * (TC(I)**2.5) / PC(I)
      BRKL(I) = C2RKL(I) * R * TC(I) / PC(I)
   10 CONTINUE
C
   20 DO 130 I = 1, NCOMP
      IF (I .EQ. NCOMP) GO TO 130
      I1 = I + 1
```

```
C
      DO 130 J = I1, NCOMP
      IF (KIJ .GT. 1) GO TO 60
      TCIJ(I,J) = (TC(I)*TC(J)) ** 0.5 * (1.0 - CORRL(I,J))
      TCIJ(J,I) = TCIJ(I,J)
      WIJ(I,J) = (W(I) + W(J)) * 0.5
      ZCOIJ = 0.291 - 0.08 * WIJ(I,J)
   30 VCOIJ = (VCO(I)**(1./3.) + VCO(J)**(1./3.)) ** 3 / 8.0
   40 PCOIJ = ZCOIJ * R * TCIJ(I,J) / VCOIJ
      PCIJ = PCOIJ
   50 ARKL(I,J) = (C1RKL(I) + C1RKL(J)) * 0.5 * R ** 2 *
     1TCIJ(I,J) ** 2.5 / PCIJ
      GO TO 120
   60 IF (KIJ .GT. 2) GO TO 70
      ARKL(I,J) = (ARKL(I,I)*ARKL(J,J)) ** 0.5 * (1.0 -
     1CORRL(I,J))
      GO TO 120
   70 IF (KIJ .GT. 3) GO TO 80
      ARKL(I,J) = (ARKL(I,I)*CORRL(I,J) + (1. - CORRL(I,J))*
     1ARKL(J,J))
      GO TO 120
C
   80 DO 90 II = 1, NCOMP
      TC1(II) = (WAII(II)/WAO*WBO/WBII(II)) ** (2./3.) *
     1TCII(II)
   90 PC1(II) = (WAII(II)/WAO) ** (2./3.) * (WBO/WBII(II)) *
     1* (5./3.) * PCII(II)
C
      VC1(I) = (1./3.) * R * TC1(I) / PC1(I)
      VC1(J) = (1./3.) * R * TC1(J) / PC1(J)
      TC11(I,J) = (1. - CORRL(I,J)) * (TC1(I)*TC1(J)) ** 0.5
      IF (KIJ .GT. 4) GO TO 110
      VC11(I,J) = (1./2.) * (VC1(I) + VC1(J))
  100 ARKL(I,J) = WAO * R * R * TC11(I,J) ** 2.5 / ((1./3.)*
     1R*TC11(I,J)/VC11(I,J))
      GO TO 120
  110 VC11(I,J) = (1./8.) * (VC1(I)**(1./3.) + VC1(J)**(1./
     13.)) ** 3.0
      GO TO 100
  120 ARKL(J,I) = ARKL(I,J)
  130 CONTINUE
C
      AMRKL = 0.0
      BMRKL = 0.0
C
      DO 140 I = 1, NCOMP
      A2I(I) = ARKL(I,I) / RT ** 2 / T ** 0.5
      BI(I) = BRKL(I) / RT
      BI(I) = BRKL(I) / RT
      BMRKL = BMRKL + X(I) * BRKL(I)
      AIRKL(I) = 0.0
C
      DO 140 J = 1, NCOMP
      AIRKL(I) = AIRKL(I) + X(J) * ARKL(I,J)
  140 AMRKL = AMRKL + X(I) * X(J) * ARKL(I,J)
C
      A2 = AMRKL / RT ** 2 / T ** 0.5
      B = BMRKL / RT
      A(1) = 1.0
      A(2) = B * P / 2. - 1.0M - SCOTT
```

```
      A(3) = A2 * P - 3.0 * B * P / 2. - (B*P) ** 2 / 2.M -
     1SCOTT
      A(4) = -(B*B + A2*B) * P * P / 2.M - SCOTT
C      IF ( KPRINT .EQ. 1) WRITE(3, 593) A2,B,A(1),A(2),A(3),A(4)
      CALL CUBEQN
C      IF (KPRINT .EQ. 1)  WRITE(3, 591) MTYPE,Z(1),Z(2),Z(3)
      IF (MTYPE) 150, 180, 180
  150 IF (LV .EQ. 0) GO TO 170
C      ZL=AMIN1(Z(1),Z(2),Z(3))
      ZL = DMIN1(Z(1),Z(2),Z(3)) DOUBLEP
      GO TO 190
C 301 IF (KPRINT .EQ. 1) WRITE(3, 333) ZL,P,CORRL(1,2)
  160 CONTINUE
      PHI(1) = 20000.
      GO TO 250
  170 CONTINUE
C      ZL=AMAX1(Z(1),Z(2),Z(3))
      ZL = DMAX1(Z(1),Z(2),Z(3))
      GO TO 190
  180 ZL = Z(1)
  190 H = B * P / ZL
C      WRITE (3,591) H,ZL,B,Z(1),Z(2),Z(3),LV
C 591 FORMAT(5X,6E12.4,I2)
C       CALL EQNRK(A2,B,P,H,ZL,LV)
C  RK ONLY
      VL = RT * B / H
      IF (ZL .LE. 0.0 .OR. VL .LE. 0.0) GO TO 160
  200 CONTINUE
      IF (LV .EQ. 0) GO TO 210
C
  210 DO 240 I = 1, NCOMP
      YSCOTT = B * RT / 4.0 / VLM - SCOTT
C      IF (KPRINT .EQ.1) WRITE(3,334) ZL, H ,YSCOTT
      PHILN = BI(I) / B * (4.0*YSCOTT/(1. - 2.0*YSCOTT)) -
     12.0 * ALOG(1. - 2.0*YSCOTM - SCOTT
   90 T) - (A2 / B)*(2.0 * AIRKL(I) / AMRKL - BI(I) / B)*
     1ALOG(1.0 + H) M - SCOTT
C
  230 -A2*BI(I)/B*RT/(VL+B*RT) -ALOG(ZL)
     1M-SCOTT
      PHI(I) = EXP(PHILN)
      IF (LV .EQ. 0) GO TO 240
      QE1 = 0.0
  240 CONTINUE
C
  250 RETURN
  260 FORMAT (1H , 'A(1)=', 6F12.8)
  270 FORMAT (1H , 'Z', I6, 8F10.6)
  280 FORMAT (1H0, 'ZL=', 5F10.5)
  290 FORMAT (1H , ' ZL=', F13.7, ' H=', F14.8, ' YSCOTT=',
     1F15.9)
      END
```

PROGRAM E-2

Computer Program Used to Predict VLE Properties for Multicomponent Systems by Using the ICL Equation of State

```
C ******************************************************************
C
C PROGRAM E-2
C
C COMPUTER PROGRAM USED TO PREDICT VLE PROPERTIES FOR
C MULTICOMPONENT SYSTEM BY USING THE ICL EQ UATION OF STATE
C
C ******************************************************************
C
C     THIS PROGRAM IS A COLLECTIVE VERSION OF THE PREVIOS ONES
C     MODIFIED BY C.-T. FU
C
C
C     DEPARTMENT OF CHEMICAL ENGINEERING
C
C     GRADUATE SCHOOL
C
C     STUDENT NUMBER 039129
C
C     MAY 24, 1979
C
C     ******************************************************************
C
C
C                                                    )
C     SUBROUTINES PYINIT, PCALC, YVCALC, SUPT, CUBEQN AND EQNRK ARE
C     REQUIRED
C
C     ******************************************************************
      DIMENSION TITLE(20), PCII(5), VCII(5), TCII(5), W(5),
     1WAII(5), WBII(5), COMPA(5), COMPB(5), COMPC(5),
     2FREFER(5), PHI(5), PHIV(5), PHIJ(5), PHIK(5), P1(5),
     3PTVOL(5), PPVOL(5), VVV(5), CORRL(5,5), X(5), Y(5),
     4DX(5), VC(5), TC(5), CK(5), EXG(5), EXGF(5), YX(5),
     5VVL(5), DY(5), PVOL(5), GAMMB(5), GAMMA(5), AMOLWT(5),
     6PC(5), XX(60,5), YY(60,5), PP(60), EXG1(5), COMPD(5),
     7SSDK(5), ESSDK(5), SADK(5)
      DIMENSION A0(20), A1(20), A2(20), A3(20), B(20),
     1TR(5), S(5)
      DIMENSION EDYAV(5), DYAV(5), EDY(5)
      DIMENSION P00(5)
      COMMON /FIRST/ TC, PC, VCII, W, AMOLWT, T, P, R,
     1NCOMP, NQNTUM /SECOND/ Z, A, MTYPE /THIRD/ PCII, TCII,
     2WAII, WBII, PSI, PP, XX, YY, IJK, KIJ, NOI, ITER, K,
     3CORRL, EXGT, PPVOL, PHI, PHIV, VV, VVL1, DV, P1, PPP,
     4Y /FOURTH/ SDP, EP, DP, SSDY, SDY, SDV, PHIK, GAMMA,
     5EXG, X, SUMY, GAMMB, PG, RT, PF, FREFER, YX, DY, EXG1
     6/FIFTH/ DPAV, EDYAV, EDY, EDP, EDPAV, DYAV, SSDK,
     7SADY, SADK /SIXTH/ PN /SEVEN/ P00, KPRINT
      NONH2 = 10
C      FOR NON-HYDROGEN SYSTEM SET NONH2=100
C      FOR HYDROGEN SYSTEM SET NONH2 =10
      READ (1,420) M
      M4 = 2 * (M + 1) + 2 * (M + 2)
      M1 = M + 2
C
      DO 10 I = 1, M4
      READ (1,580) A0(I), A1(I), A2(I), A3(I)
   10 WRITE (3,430) A0(I), A1(I), A2(I), A3(I)
C
```

```
   20 READ (1,440,END=410) (TITLE(I),I=1,20)
      WRITE (3,450) (TITLE(I),I=1,20)
      READ (1,460) NCOMP, NQNTUM, IJK, R
C      IF NQNTU IS 1 XX IS K VALVE, XX = YY/XX
C      NQNTUM = 2 , XX AND YY ARE GIVEN IN K1 AND K2, RESPECTIVELY
      IF (NCOMP .LE. 0) GO TO 20
C      NCOMP MUST BE GREATER THAN ZERO
C      NCOMP: NO. OF COMPONENTS IN THE MIXTURE
C      R: GAS CONSTANT
C      IJK: CONTROL VARIABLE
C      WRITE HEADINGS
      WRITE (3,470)
      WRITE (3,480)
C
      DO 30 I = 1, NCOMP
      READ (1,490) PCII(I), VCII(I), TCII(I), W(I), WAII(I),
     1WBII(I), COMPA(I), COMPB(I), COMPC(I), COMPD(I)
   30 WRITE (3,500) PCII(I), VCII(I), TCII(I), W(I),
     1WAII(I), WBII(I), COMPA(I), COMPB(I), COMPC(I),
     2COMPD(I)
C      IT              T
C      0          DEGREE K
C      1          DEGREE F
C      2          DEGREE C
      READ (1,510) T, PSI, IT
      IF (IT .EQ. 1) T = (T - 32.) * 5. / 9. + 273.15
      IF (IT .EQ. 2) T = T + 273.15
   40 WRITE (3,520) T, PSI
C      IJK             P
C      0           PSIA
C      1           ATM
C      2      MPA
C      3              MMHG
      READ (1,530) IJK, KIJ, CINI, CFIN
C      PP IS EXPERIMENTAL VALUE OF PRESSURE
      NCOMP1 = NCOMP - 1
      NOI = 1
   50 READ (1,540) PP(NOI), (XX(NOI,J),J=1,NCOMP1),
     1(YY(NOI,J),J=1,NCOMP1)
C  105 FORMAT(F14.8,F10.9,F14.13)
      IF (NQNTUM .EQ. 1) XX(NOI,1) = YY(NOI,1) / XX(NOI,1)
      IF (NQNTUM .EQ. 2) GO TO 90
   60 CONTINUE
      IF (IJK .EQ. 4) PP(NOI) = PP(NOI) / 76.0 * 14.697
      IF (IJK .EQ. 3) PP(NOI) = PP(NOI) / 760.0 * 14.697
      IF (IJK .EQ. 2) PP(NOI) = PP(NOI) * 1000. / 6.895
      IF (IJK .EQ. 1) PP(NOI) = PP(NOI) * 14.697
      SX = 0.0
      SY = 0.0
      IF (PP(NOI)) 100, 70, 70
C
   70 DO 80 J = 1, NCOMP1
      SY = SY + YY(NOI,J)
   80 SX = SX + XX(NOI,J)
C
      XX(NOI,NCOMP) = 1.0 - SX
      YY(NOI,NCOMP) = 1.0 - SY
      PP(NOI) = PP(NOI) / 14.697
      WRITE (3,550) NOI, PP(NOI), (XX(NOI,J),J=1,NCOMP),
     1(YY(NOI,J),J=1,NCOMP)
```

```
      PP(NOI) = PP(NOI) * 14.697
      NOI = NOI + 1
      GO TO 50
   90 CONTINUE
      TEMP = (1. - YY(NOI,1)) / (XX(NOI,1) - YY(NOI,1))
      YY(NOI,1) = TEMP * XX(NOI,1)
      XX(NOI,1) = TEMP
      GO TO 60
  100 NOI = NOI - 1
      BK4 = 0.0
  110 READ (1,560) BK4, BK5, BK6, BK7, BK8, BK9
C
      DO 120 I = 1, NCOMP
  120 TR(I) = T / TCII(I)
C
      WRITE (3,570) (TR(I),I=1,NCOMP)
  130 READ (1,580) W(1), W(2), W(3), W(4)
C      IVPE = 0 VAPOR PRESSURE IS THE ANTOINE EQ.
C      IVPE = 1 VAPOR PRESSURE IS GOODWIN'S EQ.
C      IVPE = 2 FOR GOODWIN V.P.EQ. ( CH4)
C    IVPE=3 FOR V.P. IN ATM. AT THE SPECIFIED TEMP.
      DO 180 I = 1, NCOMP
      READ (1,460) IVPE
      IVPE = IVPE + 1
      GO TO (140, 150, 160, 170), IVPE
  140 READ (1,540) AAP, BBP, CCP
      WRITE (3,550) I, AAP, BBP, CCP
      P00(I) = AAP - BBP / (CCP + T - 273.15)
      P00(I) = 10. ** P00(I) / 760.
      IF (TR(I) .GT. 1.0) P00(I) = 10. ** (ALOG10(PCII(I))*
     1TR(I))
      WRITE (3,550) IVPE, P00(I)
      GO TO 180
  150 READ (1,540) AAP, BBP, CCP, DDP
      XP = (1. - 63.14/T) / (1. - 63.14/TCII(I))
      P00(I) = AAP * XP + BBP * XP * XP + CCP * XP ** 3
      FX = 0.
      IF (XP .LT. 1.) FX = -(1. - XP) ** 2 * ALOG(1. - XP)
      P00(I) = P00(I) + DDP * FX
      P00(I) = EXP(P00(I)) * .1233
      IF (XP .GT. 1.0) P00(I) = 10. ** (ALOG10(PCII(I))*TR(
     1I))
      WRITE (3,550) I, AAP, BBP, CCP, DDP
      WRITE (3,550) IVPE, P00(I)
      GO TO 180
  160 READ (1,540) AAP, BBP, CCP, DDP
      XP = (1. - 90.68/T) / (1. - 90.68/TCII(I))
      XXP = ABS(1. - XP)
      IF (XP .GE. 1.) DDP = 0.0
      P00(I) = AAP * XP + BBP * XP * XP + CCP * XP ** 3 +
     1DDP * XP * XXP ** 1.5
      P00(I) = EXP(P00(I)) * .1159
      IF (XP .GT. 1.0) P00(I) = 10. ** (ALOG10(PCII(I))*TR(
     1I))
      WRITE (3,550) I, AAP, BBP, CCP, DDP
      WRITE (3,550) IVPE, P00(I)
      GO TO 180
  170 READ (1,540) P00(I)
      WRITE (3,550) IVPE, P00(I)
  180 CONTINUE
```

```
C
      READ (1,590) BW, WA10, WA11, WA12, WA13
      READ (1,590) BW, WA20, WA21, WA22, WA23
      WRITE (3,430) WA10, WA11, WA12, WA13, WB10, WB11,
     1WB12, WB13
      WRITE (3,430) WA20, WA21, WA22, WA23, WB20, WB21,
     1WB22, WB23
C
      DO 190 I = 1, NOI
      IF (XX(NOI,1) .LT. .0 .OR. XX(NOI,1) .GT. 1.0)
     1GO TO 20
  190 CONTINUE
C
      DO 200 I = 1, NCOMP
      S(I) = 10. ** ((1. - SQRT(TR(I)))*(.028 + .665*W(I) -
     1.2*(1. - TR(I))))
      WAII(I) = .42748 * S(I) ** 2.5
      WBII(I) = .08664 * S(I)
      WRITE (3,600) I, S(I), I, WAII(I), I, WBII(I)
  200 CONTINUE
C
      DO 250 JJJ = 1, NCOMP
      IF (TR(JJJ) .LT. .85) GO TO 220
      II = 1
      IFF = M1
C
      DO 210 IN = II, IFF
      I = IN - II + 1
      B(I) = A0(IN) + A1(IN) * W(JJJ) + A2(IN) * W(JJJ) * W(
     1JJJ) + A3(IN) * W(JJJ) * W(JJJ) * W(JJJ)
      JN = M + 2 + IN
      J = M + 2 + I
  210 B(J) = A0(JN) + A1(JN) * W(JJJ) + A2(JN) * W(JJJ) * W(
     1JJJ) + A3(JN) * W(JJJ) * W(JJJ) * W(JJJ)
C
      IF (TR(JJJ) .GE. 1.0) WAII(JJJ) = B(1)
      IF (TR(JJJ) .GE. 1.0) WBII(JJJ) = B(5)
      IF (TR(JJJ) .GE. 1.0) GO TO 250
      WAII(JJJ) = B(1) + B(2) * (1. - TR(JJJ)) + B(3) * (1.
     1- TR(JJJ)) ** (1./3.) + B(4) * (1. - TR(JJJ)) ** (2./
     23.)
      WBII(JJJ) = B(5) + B(6) * (1. - TR(JJJ)) + B(7) * (1.
     1- TR(JJJ)) ** (1./3.) + B(8) * (1. - TR(JJJ)) ** (2./
     23.)
      GO TO 250
  220 II = 2 * (M + 2) + 1
      IFF = 2 * (M + 2) + (M + 1)
      WAII(JJJ) = 0.0
      WBII(JJJ) = 0.0
C
      DO 230 IN = II, IFF
      I = IN - II + 1
      B(I) = A0(IN) + A1(IN) * W(JJJ) + A2(IN) * W(JJJ) * W(
     1JJJ)
      JN = M + 1 + IN
      J = M + 1 + I
  230 B(J) = A0(JN) + A1(JN) * W(JJJ) + A2(JN) * W(JJJ) * W(
     1JJJ)
C
      M2 = M + 1
```

```
C
      DO 240 I = 1, M2
      JJ = I - 1
      J = M + 1 + I
      WAII(JJJ) = WAII(JJJ) + B(I) * TR(JJJ) ** JJ
  240 WBII(JJJ) = WBII(JJJ) + B(J) * TR(JJJ) ** JJ
C
  250 CONTINUE
C
      WBII(1) = 0.034 - .025 * W(1) + 0.1 / TR(1) - 0.025 /
     1TR(1) ** 2M - SRK1P
      WBII(2) = 0.034 - 0.025 * W(2) + 0.1 / TR(2) - 0.025 /
     1 TR(2) ** 2M - SRK1P
      WAII(1) = WA10 + WA11 / TR(1) + WA12 / TR(1) ** 2M -
     1SRK1P
      WAII(2) = WA20 + WA21 / TR(2) + WA22 / TR(2) ** 2M -
     1SRK1P
C
      DO 260 I = 1, NCOMP
      WAII(I) = ((1.12961961 - .7846527*W(I)) + (-.13989729
     1+ 1.0449394*W(I))/TR(I) + (.012089291 - .24821323*W(I)
     2)/TR(I)**2) ** 2 * .467123
      WBII(I) = .108762
      WAII(I) = (.35190808 - W(I)*.79946286) + (.18385964 +
     1W(I)*.92295131) / TR(I) + (-.066361596 - W(I)*.
     219401653) / TR(I) ** 2
      WAII(I) = (.35190808 - W(I)*.85117382) + (.18385964 +
     1W(I)*1.0010328) / TR(I) + (-.066361596 - W(I)*.
     222265942) / TR(I) ** 2
      WBII(I) = .034 - .025 * W(I) + .1 / TR(I) - .025 / TR(
     1I) ** 2
  260 CONTINUE
C
      IF (NONH2 .EQ. 100) GO TO 270
      WAII(1) = 0.503476 - 0.00876 / TR(1) - 0.028115 / TR(
     11) ** 2
      WBII(1) = 0.100278 - 0.0233989 / TR(1) + 0.032129 /
     1TR(1) ** 2
  270 WRITE (3,610) (WAII(I),I=1,NCOMP), (WBII(I),I=1,NCOMP)
     1, (S(I),I=1,NCOMP)
  280 CONTINUE
  290 KIJ = KIJ + 2
      IF (KIJ .GE. 3) GO TO 20
  300 WRITE (3,620) IJK, KIJ, CINI, CFIN
      CALL PYINIT(BK4, BK5, BK6, BK7, BK8, BK9)
      WRITE (3,630) ((CORRL(I,J),I=1,NCOMP),J=1,NCOMP)
      WRITE (3,640)
C
      DO 310 J = 1, NCOMP
      EDYAV(J) = 0.0
      DYAV(J) = 0.0
      SSDK(J) = 0.0
  310 CONTINUE
C
      EDPAV = 0.0
      DPAV = 0.0
      SSDY = 0.0
      SADY = 0.0
      SDV = 0.0
      KPRINT = 1
```

```
C
      DO 390 K = 1, NOI
      CALL XCALC
      WRITE (3,650) VV, (X(I),I=1,NCOMP), (XX(K,I),I=1,
     1NCOMP)
C       IF ( YY(K, 1) .LE. 0.0) GO TO 500
      DO 320 I = 1, NCOMP
  320 WRITE (3,660) I, PHIV(I), I, Y(I)
C
      DO 330 I = 1, NCOMP
  330 WRITE (3,670) I, PHIK(I), I, P1(I)
C
      WRITE (3,340) T, P, K, PP(K), DP, EP
  340 FORMAT(3X,'T=',F10.4,3X,'P=',F10.4,3X,'PP(',I3,')=',
     1F10.4,3X'DX=',F10.4,20X,'EX=',F10.4,'%')
      CALL YVCALC
      WRITE (3,680) (X(I),I=1,4), (Y(I),I=1,4),
     1(EDY(I),I=1,4), DP, EDP
      WRITE (3,690) DY(1), DY(2), DY(3), DY(4)
C
      DO 350 J = 1, NCOMP
  350 WRITE (3,700) COMPA(J), COMPB(J), COMPC(J), COMPD(J),
     1PG, J, X(J), J, GAMMB(J)
C
      WRITE (3,710) (EXG1(I),I=1,NCOMP)
      WRITE (3,720) EXGT
C
      DO 360 J = 1, NCOMP
  360 WRITE (3,730) COMPA(J), COMPB(J), COMPC(J), COMPD(J),
     1P, J, YX(J), J, GAMMA(J)
C
      WRITE (3,710) (EXG(I),I=1,NCOMP)
C
      DO 380 J = 1, NCOMP
      IF (X(J) .LE. 0.000001) CK(J) = 1000000.0
      IF (X(J) .LE. 0.000001) GO TO 370
      CK(J) = Y(J) / X(J)
  370 DX(J) = X(J) - XX(K,J)
      WRITE (3,740) COMPA(J), COMPB(J), COMPC(J), COMPD(J),
     1J, X(J), K, J, XX(K,J), J, DX(J), J, CK(J)
  380 WRITE (3,750) COMPA(J), COMPB(J), COMPC(J), COMPD(J),
     1J, X(J), J, Y(J), K, J, YY(K,J), J, DY(J), J,
     2PPVOL(J), J, PHI(J), J, PHIV(J), SUMY, ITER
C
      WRITE (3,760) VV, VVL1, DV
  390 CONTINUE
C
      WRITE (3,770) DPAV, EDPAV, (DYAV(J),J=1,4),
     1(EDYAV(J),J=1,4)
      SADY = SADY / NOI / NCOMP
      SSDYA = SADY * NOI
      WRITE (3,780) BK4, SDP, SADY, SSDYA
C
      DO 400 J = 1, NCOMP
      ESSDK(J) = SADK(J) * 100.0
  400 CONTINUE
C      ESSDK(J) = SQRT (SUM OF (DK/K)**2 ) *100./NO OF COMPONENT
      WRITE (3,790) (ESSDK(J),J=1,NCOMP)
      PSI = PPP
      GO TO 290
```

```
  410 RETURN
  420 FORMAT (2I5)
  430 FORMAT (1H0, 4D20.8)
  440 FORMAT (20A4)
  450 FORMAT (1H1, 20A4)
  460 FORMAT (3I5, F10.4)
  470 FORMAT (1H0, 'INPUT DATA:'//)
  480 FORMAT (1H0, 6X, 'PCII', 8X, 'VCII', 8X, 'TCII', 8X,
     1'W', 11X, 'WAII', 8X, 'WBII', 4X, 'COMPONENT', 3X,
     2'AMOLWT'//)
  490 FORMAT (4F8.4, F8.5, F9.6, 4A4)
  500 FORMAT (1H , 6F12.6, 3X, 4A4)
  510 FORMAT (2F10.4, 5I2)
  520 FORMAT (1H0, 'T=', F10.4, 3X, 'DEG. OF K', 3X, 'P=',
     1F10.4, 3X, ' ATM')
  530 FORMAT (2I5, 2F10.4)
  540 FORMAT (8F10.0)
  550 FORMAT (1H , I3, 1F12.5, ' P IN ATM ', 8F12.5)
  560 FORMAT (8F10.4)
  570 FORMAT (1H0, 'TR(1)=', F6.4, '  TR(2)=', F6.4, 5X,
     1'TR(3)=', F6.4, 5X, 'TR(4)=', F6.4)
  580 FORMAT (4D20.8)
  590 FORMAT (6F13.8)
  600 FORMAT (1H0, 'S(', I2, ')=', F10.5, 'WAII(', I2, ')=',
     1F10.7, 'WBII(', I2, ')=', F10.7)
  610 FORMAT (1H0, 9F12.8)
  620 FORMAT (1H0, 3X, 'IJK=', I5, 3X, 'KIJ=', I5, 3X,
     1'CINI=', F10.4, 3X, 'CFIN=', F10.4//)
  630 FORMAT (1H0, 9(3X,F10.5))
  640 FORMAT (1H0, 'RESULT:')
  650 FORMAT (1H0, 3X, 'VV=', F14.4, 3X, 'X=', 5F10.6, //,
     1 19X, '  XX=  ', 5F10.6)
  660 FORMAT (1H , 3X, 'PHIV(', I2, ')=', F10.4, 3X, 'Y(',
     1I2, ')=', F10.4)
  670 FORMAT (1H , 3X, 'PHIK(', I2, ')=', F10.4, 3X, 'P1(',
     1I2, ')=', F10.4)
  680 FORMAT (1H , '  X(1)=', F10.8, '  X(2)=', F10.8,
     1'  X(3)=', F10.8, '  X(4)=', F10.8, /, '  Y(1)=',
     2F10.8, '  Y(2)=', F10.8, '  Y(3)=', F10.8, '  Y(4)=',
     3F10.8, /'  EDY(1)=', E12.6, '%  EDY       (2)=',
     4E12.6, '%  EDY(3)=', E12.6, '%  EDY(4)=', E12.6,
     5'%  DP=', E12.6, '  EDP=', E12.6, '%')
  690 FORMAT (1H , '  DY(1)=', E12.6, '  DY(2)=', E12.6,
     1'  DY(3)=', E12.6, '  DY(4)=', E12.6)
  700 FORMAT (10X, 4A4, 3X, 'P=', F10.4, 3X, 'X(', I2, ')=',
     1F10.4, 3X, 'GAMMB(', I2, ')=', F10.4)
  710 FORMAT (10X, 'LN @     EXG(I)=', 3F10.4)
  720 FORMAT (110X, 'EXGT=', F10.4)
  730 FORMAT (10X, 4A4, 3X, 'P=', F10.4, 3X, 'YX(', I2,
     1')=', F10.4, 3X, 'GAMMA(', I2, ')=', F10.4)
  740 FORMAT (1H0, 3X, 4A4, 3X, 'X(', I2, ')=', F10.6, 3X,
     13X, 'XX(', I4, I2, ')=', F10.8, 3X, 'DX(', I2, ')=',
     2E12.4, 3X, 'CK(', I2, ')=', F8.4)
  750 FORMAT (1H0, 3X, 4A4, 3X, 'X(', I2, ')=', F10.6, 3X,
     1'Y(', I2, ')=', F10.8, 3X, 'YY(', I4, I2, ')=', F10.8,
     23X, 'DY(', I2, ')=', E12.4/21X, 'PPVOL(', I2, ')=',
     3F10.4, 3X, 'PHI(', I2, ')=', F10.4, 3X, 'PHIV(', I2,
     4')=', F10.4, 3X, 'SUMY=', F10.4, 3X, 'ITER=', I4)
  760 FORMAT (15X, 'VV=', F10.4, 3X, 'VVL1=', F10.4, 3X,
     1'DV=', E12.4)
```

```
  770 FORMAT (1H0, '   DPAV=', E12.6, 'EDPAV=', E12.6, '%', /
     1, 'DYAV(1)=', E12.6, '   DYAV(2)=', E12.6,
     2'  DYAV(3)=', E12.6/'  DYAV(4)=', E12.6, /,
     3'  EDYAV(1)=', E12.6, '%  EYDAV(2)=', E12.6,
     4'%  EDYAV(3)=', E12.6, '%', '   EYDAV(4)=', E12.6, '%')
  780 FORMAT (1H0, 3X, ' K12= ', E14.8, 3X, ' SDP= ', E14.8,
     13X, ' DYAV= ', E14.8, 3X, 'SUM OF DY=', E14.8)
  790 FORMAT (2X, ' % OF K1 IN RMS = ', F10.4, 5X, ' % OF K2 IN RMS = '
     1, F10.4, /, 2X, ' % OF K3 IN RMS = ', F10.4, 5X,
     2' % OF K4 IN RMS = ', F10.4, //)
      END
C
C
C
C
C
C
C
C     ******************************************************************
      SUBROUTINE PYINIT(BK, BK1, BK2, BK3, BK10, BK11)
C
C     ******************************************************************
C     PREDICT P-X-Y CURVE
C     MODIFIED REDLICH-KWONG EQUATION OF STATE IS USED FOR MULTI-
C     COMPONENT SYSTEMS CALCULATION FROM THE PURE COMPONENT PROPERTIES
C     ONE OF THE COMPONENTS IS AT ITS HYPERCRITICAL STATE
C     ******************************************************************
      DIMENSION TITLE(20), PCII(5), VCII(5), TCII(5), W(5),
     1WAII(5), WBII(5), COMPA(5), COMPB(5), COMPC(5),
     2FREFER(5), PHI(5), PHIV(5), PHIJ(5), PHIK(5), P1(5),
     3PTVOL(5), PPVOL(5), VVV(5), CORRL(5,5), X(5), Y(5),
     4SSDK(5), VC(5), TC(5), SADK(5), EXG(5), EXGF(5),
     5YX(5), VVL(5), DY(5), PVOL(5), GAMMB(5), GAMMA(5),
     6AMOLWT(5), PC(5), XX(60,5), YY(60,5), PP(60), EXG1(5),
     7COMPD(5)
      COMMON /FIRST/ TC, PC, VCII, W, AMOLWT, T, P, R,
     1NCOMP, NQNTUM /SECOND/ Z, A, MTYPE /THIRD/ PCII, TCII,
     2WAII, WBII, PSI, PP, XX, YY, IJK, KIJ, NOI, ITER, K,
     3CORRL, EXGT, PPVOL, PHI, PHIV, VV, VVL1, DV, P1, PPP,
     4Y /FOURTH/ SDP, EP, DP, SSDY, SDY, SDV, PHIK, GAMMA,
     5EXG, X, SUMY, GAMMB, PG, RT, PF, FREFER, YX, DY, EXG1
     6/FIFTH/ DPAV, EDYAV, EDY, EDP, EDPAV, DYAV, SSDK,
     7SADY, SADK
      SDP = 0.0
      SDV = 0.0
      SSDY = 0.0
      SADY = 0.0
C
      DO 10 J = 1, NCOMP
   10 SSDK(J) = 0.0
C     PF = REFERENCE PRESSURE OF 1000 LB OR 1000./14.697 ATMOSPHERE
C     SINCE ONE ATMOSPHERE = 14.697 LB
      PF = 1000 / 14.697
      PPP = PSI
      PSI = PSI / 14.697
C
      DO 20 I = 1, NCOMP
   20 CORRL(I,I) = 0.0
C
      CORRL(1,2) = BK
```

```
      CORRL(2,1) = CORRL(1,2)
      CORRL(2,3) = BK1
      CORRL(3,2) = CORRL(2,3)
      CORRL(1,3) = BK2
      CORRL(3,1) = CORRL(1,3)
      CORRL(1,4) = BK3
      CORRL(4,1) = BK3
      CORRL(2,4) = BK10
      CORRL(4,2) = CORRL(2,4)
      CORRL(3,4) = BK11
      CORRL(4,3) = CORRL(3,4)
C      SET UP THE NECESSARY INFORMATION FOR THE SUPPORT SUBROUTINE
      RETURN
      END
C
C ***********************************************************************
      SUBROUTINE XCALC
C
C ***********************************************************************
      DIMENSION TITLE(20), PCII(5), VCII(5), TCII(5), W(5),
     1WAII(5), WBII(5), COMPA(5), COMPB(5), COMPC(5),
     2FREFER(5), PHI(5), PHIV(5), PHIJ(5), PHIK(5), P1(5),
     3PTVOL(5), PPVOL(5), VVV(5), CORRL(5,5), X(5), Y(5),
     4SSDK(5), VC(5), TC(5), SADK(5), EXG(5), EXGF(5),
     5YX(5), VVL(5), DY(5), PVOL(5), GAMMB(5), GAMMA(5),
     6AMOLWT(5), PC(5), XX(60,5), YY(60,5), PP(60), EXG1(5),
     7COMPD(5)
      DIMENSION EDYAV(5), DYAV(5), EDY(5), SDK(5)
      DIMENSION P00(5)
      COMMON /FIRST/ TC, PC, VCII, W, AMOLWT, T, P, R,
     1NCOMP, NQNTUM /SECOND/ Z, A, MTYPE /THIRD/ PCII, TCII,
     2WAII, WBII, PSI, PP, XX, YY, IJK, KIJ, NOI, ITER, K,
     3CORRL, EXGT, PPVOL, PHI, PHIV, VV, VVL1, DV, P1, PPP,
     4Y /FOURTH/ SDP, EP, DP, SSDY, SDY, SDV, PHIK, GAMMA,
     5EXG, X, SUMY, GAMMB, PG, RT, PF, FREFER, YX, DY, EXG1
     6/FIFTH/ DPAV, EDYAV, EDY, EDP, EDPAV, DYAV, SSDK,
     7SADY, SADK /SIXTH/ PN /SEVEN/ P00, KPRINT
      IPP = 0
      ITER = 0
C
      DO 10 I = 1, NCOMP
      PC(I) = PCII(I)
      VC(I) = VCII(I)
   10 TC(I) = TCII(I)
C
      RT = R * T
      P = PF
C      USE TO CALCULATE THE MOLAL VOLUMN VL, FUGACITY COEFFICIENT PHI,
C      PARTIAL MOLAL VOLUMN PPVOL AND FUGACITY FREFER OF LIQUID PHASE
C      FOR PURE COMPONENT AT REFERENCE PRESSURE
   20 DO 40 I = 1, NCOMP
C
      DO 30 J = 1, NCOMP
   30 X(J) = 0.0
C
      X(I) = 1.0
      CALL SUPT(VL, PHI, PPVOL, WAII, WBII, CORRL, X, 1,
     1KIJ)
      IF (PHI(1) .GE. 10000.) WRITE (3,420)
      IF (PHI(1) .GE. 10000.) GO TO 40
```

```
      FREFER(I) = PHI(I) * P
   40 CONTINUE
C     ******************************
C     USE TO CALCULATE VAPORMOLE FRACTIONS Y(I) FOR MIXTURE ASSUMING
C      CORRESPONDING FUGACITY COEFFICIENTS PHIV(I) ARE 1. ( INITIAL
C GUESS)
      P = 0.
C
      DO 50 I = 1, NCOMP
   50 P = P00(I) * XX(K,I) + P
C
      P = PP(K) / 14.697
      LLL = 1
      X(1) = XX(K,1) * 0.995
      GO TO 70
   60 X(1) = XX(K,1) * 1.005
      LLL = LLL + 1
      IF (LLL .EQ. 3) GO TO 400
   70 IF (NCOMP .EQ. 3) GO TO 80
      IF (NCOMP .EQ. 4) GO TO 90
      X(2) = 1 - X(1)
      GO TO 100
   80 X(3) = XX(K,3)
      X(2) = 1 - X(1) - X(3)
      GO TO 100
   90 X(3) = XX(K,3)
      X(4) = XX(K,4)
      X(2) = 1 - X(1) - X(4) - X(3)
  100 NCOMP1 = NCOMP - 1
C
      DO 120 J = 1, NCOMP1
      I = J + 1
C
      DO 110 I = J, NCOMP
      IF (TC(J) .GT. TC(I)) GO TO 120
  110 CONTINUE
C
      GO TO 130
  120 CONTINUE
C
      J = NCOMP
C
  130 DO 140 II = 1, NCOMP
  140 Y(II) = .0001 / NCOMP1
C
      Y(1) = 0.9999
      CALL SUPT(VL, PHI, PPVOL, WAII, WBII, CORRL, X, 1,
     1KIJ)
      IF (PHI(1) .GE. 10000.) GO TO 320
      SUMY1 = 1.
  150 CALL SUPT(VV, PHIV, PVOL, WAII, WBII, CORRL, Y, 0,
     1KIJ)
      IF (PHI(1) .GE. 10000.) GO TO 330
C      SET UP THE LOOP TO FIND Y(I), SUCH THAT SUMY IS EQUAL TO SUMY1
C     ,THE SUMY OBTAINED IN IMMEDIATE PRECEDING CALCULATION, I. E. TEST
  160 CALL SUPT(VL, PHI, PPVOL, WAII, WBII, CORRL, X, 1,
     1KIJ)
      IF (PHI(1) .GE. 10000.) GO TO 340
C      FOR STABILITY BY CHANGING Y(I) AND TEST FOR SUMY = 1.0
  170 SUMY = 0.0
```

```
C
      DO 180 J = 1, NCOMP
      Y(J) = PHI(J) * X(J) / PHIV(J)
  180 SUMY = SUMY + Y(J)
C
      IF (KPRINT .EQ. 0) GO TO 190
C      WRITE(3, 900) P,Y(1),Y(2),SUMY,(PHI(J),J=1,2),(PHIV(J),J=1,2)
  190 CONTINUE
      IF (ABS(SUMY1 - SUMY) - 1.0E-4) 230, 230, 200
C      FIND A NEW VALUE OF Y(I) AND SUMY USING Y(I) IF SUMY1 IS DIFFERENT
C      FROM SUMY
  200 IF (ITER - 100) 210, 360, 360
C   15 IF (ITER - 50) 16, 21, 21
  210 ITER = ITER + 1
C
      DO 220 I = 1, NCOMP
  220 Y(I) = Y(I) / SUMY
C
      SUMY1 = SUMY
      CALL SUPT(VV, PHIV, PVOL, WAII, WBII, CORRL, Y, 0,
     1KIJ)
      IF (PHI(1) .GE. 10000.) GO TO 350
      GO TO 170
C      Y(I) IS STABLE. NOW WE HAVE TO TEST FOR SUMY = 1. BY CHANGING P
C      CHANGE X
  230 CONTINUE
      IF (IPP .NE. 0) GO TO 250
      SUMY00 = SUMY
      IPP = IPP + 1
      X0 = X(1)
      X(1) = X0 * 0.995
      ITER = ITER + 1
C
      DO 240 I = 1, NCOMP
  240 Y(I) = Y(I) / SUMY
C
      GO TO 150
C      ***********************************
  250 IF (ABS((X0 - X(1))/X(1)) .LE. .0000001)
     1GO TO 360
      SUMY1 = SUMY00
      IF (ABS(SUMY - SUMY1) .LE. 1.0E-5) GO TO 360
      XN = X(1) - (X0 - X(1)) / (SUMY - SUMY1) * (1. - SUMY)
      IF (XN .LT. 0.0 .OR. XN .GT. 1.0) GO TO 60
  260 IF (ABS((XN - X(1))/XN) .LE. .0000001) GO TO 360
      X0 = X(1)
      X(1) = XN
      IF (NCOMP .EQ. 3) GO TO 270
      IF (NCOMP .EQ. 4) GO TO 280
      X(2) = 1. - X(1)
      GO TO 290
  270 X(3) = XX(K,3)
      X(2) = 1. - X(1) - X(3)
      GO TO 290
  280 X(4) = XX(K,4)
      X(3) = XX(K,3)
      X(2) = 1. - X(1) - X(4) - X(3)
  290 SUMY00 = SUMY
      ITER = ITER + 1
C
```

```
      DO 300 I = 1, NCOMP
  300 Y(I) = Y(I) / SUMY
C
      GO TO 150
  310 IF (KPRINT .EQ. 0) GO TO 380
      WRITE (3,440) PHI(1), PHI(2), PHIV(1), PHIV(2), P1(1),
     1P1(2), SUMP, P
      GO TO 380
  320 WRITE (3,450)
      GO TO 380
  330 WRITE (3,460)
      GO TO 380
  340 WRITE (3,470)
      GO TO 380
  350 WRITE (3,480)
      GO TO 380
  360 SUMP = 0.0
C
      DO 370 I = 1, NCOMP
      P1(I) = PHI(I) * X(I) * P / PHIV(I)
  370 SUMP = SUMP + P1(I)
C
      IF (ABS(P - SUMP) .GT. .1) GO TO 310
      IF (ABS(SUMY - 1.0) .GT. 1.0E-4) GO TO 310
C      GATHER STATISTICS AND PRINT OUT RESULTS
  380 P = P * 14.697
C      DP=DX
      DO 390 KKK = 1, NCOMP
      DP = X(KKK) - XX(K,KKK)
      EDP = (DP/XX(K,KKK)) * 100.
C      SDP = SDP + ABS(DP)
      SDP = ((SDP*(K - 1)*SDP + ABS(EDP)**2)/K) ** .5
      DPAV = (DPAV*(K - 1) + ABS(DP)) / K
      EDPAV = (EDPAV*(K - 1) + ABS(EDP)) / K
      EP = (DP/XX(K,KKK)) * 100.
  390 CONTINUE
C
      IF (LLL .EQ. 1) GO TO 410
  400 WRITE (3,490) LLL
  410 RETURN
  420 FORMAT (1H , 'FREFER')
  430 FORMAT (1H , 8F14.7)
  440 FORMAT (1H , ' THIS METHOD DID NOT  PRODUCE ANSWER'/1H0,
     18E13.7)
  450 FORMAT (1H , 'ZL =NEGATIVE VALUE')
  460 FORMAT (1H , 'ZV =NEGATIVE VALUE')
  470 FORMAT (1H , 'ZL1=NEGATIVE VALUE')
  480 FORMAT (1H , 'ZV1=NEGATIVE VALUE')
  490 FORMAT (5X, I2, 'COMPONENT 1 LESS THAN ZERO OR GREATER THAN 1'
     1)
      END
C
C ***************************************************************************
      SUBROUTINE YVCALC
C
C ***************************************************************************
      DIMENSION TITLE(20), PCII(5), VCII(5), TCII(5), W(5),
     1WAII(5), WBII(5), COMPA(5), COMPB(5), COMPC(5),
     2FREFER(5), PHI(5), PHIV(5), PHIJ(5), PHIK(5), P1(5),
     3PTVOL(5), PPVOL(5), VVV(5), CORRL(5,5), X(5), Y(5),
```

```
     4SSDK(5), VC(5), TC(5), SADK(5), EXG(5), EXGF(5),
     5YX(5), VVL(5), DY(5), PVOL(5), GAMMB(5), GAMMA(5),
     6AMOLWT(5), PC(5), XX(60,5), YY(60,5), PP(60), EXG1(5),
     7COMPD(5), SDK(5), RK(5), RKK(50,5)
      DIMENSION EDYAV(5), DYAV(5), EDY(5)
      COMMON /FIRST/ TC, PC, VCII, W, AMOLWT, T, P, R,
     1NCOMP, NQNTUM /SECOND/ Z, A, MTYPE /THIRD/ PCII, TCII,
     2WAII, WBII, PSI, PP, XX, YY, IJK, KIJ, NOI, ITER, K,
     3CORRL, EXGT, PPVOL, PHI, PHIV, VV, VVL1, DV, P1, PPP,
     4Y /FOURTH/ SDP, EP, DP, SSDY, SDY, SDV, PHIK, GAMMA,
     5EXG, X, SUMY, GAMMB, PG, RT, PF, FREFER, YX, DY, EXG1
     6/FIFTH/ DPAV, EDYAV, EDY, EDP, EDPAV, DYAV, SSDK,
     7SADY, SADK
      P = P / 14.697
      PP(K) = PP(K) / 14.697
   10 CONTINUE
C     GATHER ANOTHER STATISTICS
   20 CONTINUE
C
      DO 40 J = 1, NCOMP
      SDY = 0.0
      SDK(J) = 0.0
      DY(J) = Y(J) - YY(K,J)
      SDY = SDY + ABS(DY(J))
      IF (YY(K,J) .LE. 0.0) EDY(J) = 0.0
      IF (YY(K,J) .LE. 0.0) SDK(J) = 0.0
      IF (YY(K,J) .LE. 0.0) GO TO 30
      EDY(J) = (DY(J)/YY(K,J)) * 100.
      RK(J) = Y(J) / X(J)
      RKK(K,J) = YY(K,J) / XX(K,J)
      SDK(J) = SDK(J) + (RK(J)/RKK(K,J) - 1.0) ** 2
   30 CONTINUE
      SSDK(J) = SSDK(J) + SDK(J)
      EDYAV(J) = (EDYAV(J)*(K - 1) + ABS(EDY(J))) / K
      DYAV(J) = (DYAV(J)*(K - 1) + ABS(DY(J))) / K
      SADK(J) = (SSDK(J)**0.5) / K
   40 SADY = SADY + SDY
C
      SSDY = ((2*(K - 1)*SSDY)**2 + SADK(1)**2 + SADK(2)**2)
     1 ** 0.5 / (2*K)
C     SSDY = SADK (1)
      PSI = P
      PP(K) = PP(K) * 14.697
      GO TO 100
   50 WRITE (3,110)
      GO TO 100
   60 WRITE (3,120)
      GO TO 100
   70 WRITE (3,130)
      GO TO 100
   80 WRITE (3,140)
      GO TO 100
   90 WRITE (3,150)
  100 RETURN
  110 FORMAT (1H , 'ZL = NEGATIVE')
  120 FORMAT (1H , 'ZV = NEGATIVE')
  130 FORMAT (1H , 'ZL1= NEGATIVE')
  140 FORMAT (1H , 'ZV1= NEGATIVE')
  150 FORMAT (1H , 'ZVF=NEGATIVE')
      END
```

```
C     ***********************************************************************
      SUBROUTINE CUBEQN
C    SOLVES CUBIC EQUATION OF REDLICH-KWONG EQUATION
C      FOR COMPRESSIBILITY FACTORS
C     ***********************************************************************
      IMPLICIT REAL*8(A - H,O - Z)
      DIMENSION A(4), Z(3), B(3)
      COMMON /SECOND/ Z, A, MTYPE
   10 CONTINUE
      B(1) = A(2) / A(1)
      B1OV3 = B(1) / 3.0
      B(2) = A(3) / A(1)
      B(3) = A(4) / A(1)
      ALF = B(2) - B(1) * B1OV3
      ALFOV3 = ALF / 3.0
      BET = 2.0 * B1OV3 ** 3 - B(2) * B1OV3 + B(3)
      BETOV2 = BET / 2.0
      CUAOV3 = ALFOV3 ** 3
      SQBOV2 = BETOV2 ** 2
      DEL = SQBOV2 + CUAOV3
      IF (DEL) 100, 20, 50
   20 MTYPE = 0.0
      GAM = DSQRT(-ALFOV3)
      IF (BET) 40, 40, 30
   30 Z(1) = -2.0 * GAM - B1OV3
      Z(2) = GAM - B1OV3
      Z(3) = Z(2)
      GO TO 140
   40 Z(1) = 2.0 * GAM - B1OV3
      Z(2) = -GAM - B1OV3
      Z(3) = Z(2)
      GO TO 140
   50 MTYPE = 1
      EPS = DSQRT(DEL)
      TAU = -BETOV2
      RCU = TAU + EPS
      SCU = TAU - EPS
      SIR = 1.0
      SIS = 1.0
      IF (RCU) 60, 70, 70
   60 SIR = -1.0
   70 IF (SCU) 80, 90, 90
   80 SIS = -1.0
   90 R = SIR * (SIR*RCU) ** 0.33333333
      S = SIS * (SIS*SCU) ** 0.33333333
      Z(1) = R + S - B1OV3
      Z(2) = -(R + S) / 2.0 - B1OV3
      Z(3) = 0.86602540 * (R - S)
      GO TO 140
  100 MTYPE = -1
      QUOT = SQBOV2 / CUAOV3
      ROOT = DSQRT(-QUOT)
      IF (BET) 120, 110, 110
  110 PEI = (1.5707963 + DATAN(ROOT/DSQRT(1.0 - ROOT**2))) /
     1 3.0
      GO TO 130
  120 PEI = DATAN(DSQRT(1.0 - ROOT**2)/ROOT) / 3.0
  130 FACT = 2.0 * DSQRT(-ALFOV3)
      Z(1) = FACT * DCOS(PEI) - B1OV3
      PEI2 = PEI + 2.0943951
```

```
      Z(2) = FACT * DCOS(PEI2) - B1OV3
      PEI4 = PEI + 4.1887902
      Z(3) = FACT * DCOS(PEI4) - B1OV3
  140 RETURN
      END
C
C *********************************************************************
      SUBROUTINE SUPT(VL, PHI, PVOL, C1RKL, C2RKL, CORRL, X,
     1LV, KIJ)
C
C
C *********************************************************************
C     EVALUTION OF SUPPORTING PROPERTIES,THE FUGACITY AND THE
C     PARTIAL MOLAL VOLUME OF FLUID MIXTURES.
C     SHINN-DER CHANG, DEPT.OF CHEM.ENG., UNIV. OF OTTAWA.
      DOUBLE PRECISION A, Z
      DIMENSION TC(5), PC(5), VCO(5), W(5), AMOLWT(5),
     1ARKL(5,5), BRKL(5), WIJ(5,5), TCOIJ(5,5), TCIJ(5,5),
     2A(4), Z(3), C1RKL(5), C2RKL(5), PVOL(5), PHI(5),
     3CORRL(5,5), X(5), A2I(5), BI(5), AIRKL(5)
      DIMENSION ZZ(3)
      DIMENSION TC1(5), VC1(5), PC1(5), TC11(5,5),
     1VC11(5,5), WAII(5), WBII(5), PCII(5), TCII(5)
      DIMENSION P00(5)
      COMMON /FIRST/ TC, PC, VCO, W, AMOLWT, T, P, R, NCOMP,
     1NQNTUM /SECOND/ Z, A, MTYPE /THIRD/ PCII, TCII, WAII,
     2WBII /SEVEN/ P00, KPRINT
C     LV=0, FOR VAPOR PHASE.
C     LV=1, FOR LIQUID PHASE.
C     KIJ=1, TCIJ=SQRT(TCI*TCJ)*(1.-KIJ)
C     KIJ=2, AIJ=SQRT(AI*AJ)*(1.-KIJ)
C     KIJ=3, AIJ=(AI*KIJ+(1.-KIJ)*AJ)/2.
      RT = R * T
      WA0 = 0.42748
      WB0 = 0.08664
C
      DO 10 I = 1, NCOMP
      ARKL(I,I) = C1RKL(I) * R * R * (TC(I)**2.5) / PC(I)
      BRKL(I) = C2RKL(I) * R * TC(I) / PC(I)
   10 CONTINUE
C
   20 DO 130 I = 1, NCOMP
      IF (I .EQ. NCOMP) GO TO 130
      I1 = I + 1
C
      DO 130 J = I1, NCOMP
      IF (KIJ .GT. 1) GO TO 60
      TCIJ(I,J) = (TC(I)*TC(J)) ** 0.5 * (1.0 - CORRL(I,J))
      TCIJ(J,I) = TCIJ(I,J)
      WIJ(I,J) = (W(I) + W(J)) * 0.5
      ZCOIJ = 0.291 - 0.08 * WIJ(I,J)
   30 VCOIJ = (VCO(I)**(1./3.) + VCO(J)**(1./3.)) ** 3 / 8.0
   40 PCOIJ = ZCOIJ * R * TCIJ(I,J) / VCOIJ
      PCIJ = PCOIJ
   50 ARKL(I,J) = (C1RKL(I) + C1RKL(J)) * 0.5 * R ** 2 *
     1TCIJ(I,J) ** 2.5 / PCIJ
      GO TO 120
   60 IF (KIJ .GT. 2) GO TO 70
      ARKL(I,J) = (ARKL(I,I)*ARKL(J,J)) ** 0.5 * (1.0 -
     1CORRL(I,J))
```

```
      GO TO 120
   70 IF (KIJ .GT. 3) GO TO 80
      ARKL(I,J) = (ARKL(I,I)*CORRL(I,J) + (1. - CORRL(I,J))*
     1ARKL(J,J))
      GO TO 120
C
   80 DO 90 II = 1, NCOMP
      TC1(II) = (WAII(II)/WA0*WB0/WBII(II)) ** (2./3.) *
     1TCII(II)
   90 PC1(II) = (WAII(II)/WA0) ** (2./3.) * (WB0/WBII(II)) *
     1* (5./3.) * PCII(II)
C
      VC1(I) = (1./3.) * R * TC1(I) / PC1(I)
      VC1(J) = (1./3.) * R * TC1(J) / PC1(J)
      TC11(I,J) = (1. - CORRL(I,J)) * (TC1(I)*TC1(J)) ** 0.5
      IF (KIJ .GT. 4) GO TO 110
      VC11(I,J) = (1./2.) * (VC1(I) + VC1(J))
  100 ARKL(I,J) = WA0 * R * R * TC11(I,J) ** 2.5 / ((1./3.)*
     1R*TC11(I,J)/VC11(I,J))
      GO TO 120
  110 VC11(I,J) = (1./8.) * (VC1(I)**(1./3.) + VC1(J)**(1./
     13.)) ** 3.0
      GO TO 100
  120 ARKL(J,I) = ARKL(I,J)
  130 CONTINUE
C
      AMRKL = 0.0
      BMRKL = 0.0
C
      DO 140 I = 1, NCOMP
      A2I(I) = ARKL(I,I) / RT ** 2 / T ** 0.5
      BI(I) = BRKL(I) / RT
      BI(I) = BRKL(I) / RT
      BMRKL = BMRKL + X(I) * BRKL(I)
      AIRKL(I) = 0.0
C
      DO 140 J = 1, NCOMP
      AIRKL(I) = AIRKL(I) + X(J) * ARKL(I,J)
  140 AMRKL = AMRKL + X(I) * X(J) * ARKL(I,J)
C
      A2 = AMRKL / RT ** 2 / T ** 0.5
      B = BMRKL / RT
      A(1) = 1.0
      A(2) = B * P / 2. - 1.0M - SCOTT
      A(3) = A2 * P - 3.0 * B * P / 2. - (B*P) ** 2 / 2.M -
     1SCOTT
      A(4) = -(B*B + A2*B) * P * P / 2.M - SCOTT
C     IF ( KPRINT .EQ. 1) WRITE(3, 593) A2,B,A(1),A(2),A(3),A(4)
      CALL CUBEQN
C     IF (KPRINT .EQ. 1)  WRITE(3,-591) MTYPE,Z(1),Z(2),Z(3)
      IF (MTYPE) 150, 180, 180
  150 IF (LV .EQ. 0) GO TO 170
C     ZL=AMIN1(Z(1),Z(2),Z(3))
      ZL = DMIN1(Z(1),Z(2),Z(3)) DOUBLEP
      GO TO 190
C 301 IF (KPRINT .EQ. 1) WRITE(3, 333) ZL,P,CORRL(1,2)
  160 CONTINUE
      PHI(1) = 20000.
      GO TO 250
  170 CONTINUE
```

```
C      ZL=AMAX1(Z(1),Z(2),Z(3))
       ZL = DMAX1(Z(1),Z(2),Z(3))
       GO TO 190
  180 ZL = Z(1)
  190 H = B * P / ZL
C      WRITE (3,591) H,ZL,B,Z(1),Z(2),Z(3),LV
C  591 FORMAT(5X,6E12.4,I2)
C       CALL EQNRK(A2,B,P,H,ZL,LV)
C   RK ONLY
       VL = RT * B / H
       IF (ZL .LE. 0.0 .OR. VL .LE. 0.0) GO TO 160
  200 CONTINUE
       IF (LV .EQ. 0) GO TO 210
C
  210 DO 240 I = 1, NCOMP
       YSCOTT = B * RT / 4.0 / VLM - SCOTT
C      IF (KPRINT .EQ.1) WRITE(3,334) ZL, H ,YSCOTT
      PHILN = BI(I) / B * (4.0*YSCOTT/(1. - 2.0*YSCOTT)) -
     12.0 * ALOG(1. - 2.0*YSCOTM - SCOTT
     90 T) - (A2 / B)*(2.0 * AIRKL(I) / AMRKL - BI(I) / B)*
     1ALOG(1.0 + H) M - SCOTT
C
  230 -A2*BI(I)/B*RT/(VL+B*RT) -ALOG(ZL)
     1M-SCOTT
       PHI(I) = EXP(PHILN)
       IF (LV .EQ. 0) GO TO 240
       QE1 = 0.0
  240 CONTINUE
C
  250 RETURN
  260 FORMAT (1H ,,'A(1)=', 6F12.8)
  270 FORMAT (1H , 'Z', I6, 8F10.6)
  280 FORMAT (1H0, 'ZL=', 5F10.5)
  290 FORMAT (1H , ' ZL=', F13.7, ' H=', F14.8, ' YSCOTT=',
     1F15.9)
       END
C
  250 RETURN
  260 FORMAT (1H , 'A(1)=', 6F12.8)
  270 FORMAT (1H , 'Z', I6, 8F10.6)
  280 FORMAT (1H0, 'ZL=', 5F10.5)
  290 FORMAT (1H , ' ZL=', F13.7, ' H=', F14.8, ' YSCOTT=',
     1F15.9)
       END
       //GO.FT01F001 DD *
       0.42693395D00-0.40525654D-010.47840597D-010.
      130832269D000.23069780D01-0.57327191D010.19114806D000.
      241568655D00-0.14805542D01
       0.47833642D00-0.18450430D010.56833325D01
       0.86358627D-01 - 0.11047428D-010.12190902D-010.
      113797069D000.10831474D01-0.25186462D010.68997410D-010.
      223526222D00-0.60612746D00
       0.19217689D00-0.10650643D010.24621194D01
       0.27047169D000.17543951D000.19940365D00
       0.37561136D000.13735650D00-0.67950561D000.24335111D00-
      10.39910050D000.53413329D00
       0.84489292D-01 - 0.17053425D000.27681320D00
       0.18618565D-010.33391897D00-0.68448992D000.
      123386940D-01 - 0.19347031D000.43969793D00
       //GO.FT03F001  DD SYSOUT=A,DCB=(RECFM=FBA,BLKSIZE=399,
```

```
1LRECL=133)
/*
END
```

PROGRAM E-3

Computer Program Used to Predict VLE Properties for Multicomponent Systems by Using the Modified SRK Equation of State

```
C ******************************************************
C
C PROGRAM E-3
C
C COMPUTER PROGRAM USED TO PREDICT VLE PROPERTIES FOR
C MULTICOMPONENT SYSTEMS BY USING THE MODIFIED SRK
C EQUATION OF STATE
C
C ******************************************************
C
C     PREDICT P-X-Y CURVE
C     MODIFIED REDLICH-KWONG EQUATION OF STATE IS USED
C     FOR MULTICOMPONENT  SYSTEMS CALCULATION
C     FROM THE PURE COMPONENT PROPERTIES
C     SAOVE'S METHOD IS USED, SOAVEX IS NAMED IN FILE STATUS
C     MODIFIED BY CHENG TZE FU
C     THE ORIGINAL PROGRAM SUPPLIED BY C.HSI
C     SUBROUTINES SUPT,EQNRK, CUBEQN ARE SUPPLIED BY S.D.CHANG
C     FALSI-POSITION METHOD IS EMPLOYED
      IMPLICIT REAL*8(A - H,O - Z)
      DIMENSION PHI(5), X(5), F(5), GAMMA(5), FREFER(5), PVOL(5), Y(5),
     1PC(5), VC(5), TC(5), W(5), C1RKV(5), C2RKV(5), C1RKL(5), C2RKL(5),
     2AMOLWT(5), COMPA(5), COMPB(5), CORRV(5,5), CORRL(5,5), GAMALN(5),
     3A(4), Z(3), TITLE(20), D(5), PCO(5), VCO(5), TCO(5), PHIV(5),
     4FREF(5), PS(5), PP(80), XX(80,5), YY(80,5), YCAL(5), PPVOL(5),
     5DC1DT(5), DC2DT(5), YD(5), RATIO(5), VVL(80), ZZL(80), ZZV(80),
     6YI(5), G(5), YO(5), YPD(5), AM(5), ALF(5), TR(5), EK(5), SEK(5),
     7COMPC(5), COMPD(5), XD(5)
      COMMON /FIRST/ TC, PC, VCO, W, AMOLWT, T, P, R, NCOMP, NQNTUM,
     1ZZZL, ZZZV /SECOND/ Z, A, MTYPE /THIRD/ ALF
   10 READ (5,560,END=550) (TITLE(I),I=1,20)
      WRITE (6,570) (TITLE(I),I=1,20)
C      NQNTUM EQUAL 1  XX IS GIVEN IS K1 VALUE
C      NQNTUM EQUAL 2 XX AND YY ARE GIVEN IN K1 AND K2 VALUES
      READ (5,580) NCOMP, NQNTUM, IJK, R
      IF (NCOMP .LE. 0) GO TO 540
      WRITE (6,590)
C
      DO 20 I = 1, NCOMP
      READ (5,600) PCO(I), VCO(I), TCO(I), W(I), C1RKL(I), C2RKL(I),
     1COMPA(I), COMPB(I), COMPC(I), COMPD(I)
      PCO(1) = 20.75253
      TCO(1) = 41.666666666
      VCO(1) = R * TCO(1) * 0.291 / PCO(1)
      C1RKL(I) = .42747
      C2RKL(I) = .08664
      C1RKV(I) = C1RKL(I)
      C2RKV(I) = C2RKL(I)
   20 WRITE (6,610) PCO(I), VCO(I), TCO(I), W(I), C1RKL(I), C2RKL(I),
     1COMPA(I), COMPB(I), COMPC(I), COMPD(I)
C
      CORRL(NCOMP,NCOMP) = 0.0
      NCOMP1 = NCOMP - 1
C      READ(5,520) TTT,PSI,(YI(J),J=1,NCOMP)
C      IT = 0 , TTT IN DEGREE K
C      IT = 1 TTT IN DEGREE F
C      IT = 2, TTT IN DEGREE C
      READ (5,620) TTT, PSI, IT, (YI(J),J=1,NCOMP)
      IF (IT .EQ. 1) TTT = (TTT - 32.) * 5. / 9. + 273.15
      IF (IT .EQ. 2) TTT = TTT + 273.15
```

```
C      IJK = 0 PP IN PSIA
C      IJK = 1, PP IN ATM.
C      IJK = 2 PP IN MPA
       READ (5,630) IJK, KIJ, CINI, CFIN
       KIJ = KIJ + 1
C
       DO 30 MK = 1, NCOMP
       AM(MK) = .480 + 1.574 * W(MK) - .176 * W(MK) ** 2
       AM(MK) = 0.48508 + 1.55171 * W(MK) - 0.15613 * W(MK) ** 2
       TR(MK) = TTT / TCO(MK)
       ALF(MK) = 1.0 + AM(MK) * (1.0 - (TR(MK)**.5))
       ALF(MK) = ALF(MK) * ALF(MK)
       ALF(1) = 1.202 * DEXP(-0.30228*TR(1))
   30  WRITE (6,640) AM(MK), TR(MK), ALF(MK)
C      PSI=PSI/14.697
       PSII = PSI
C      PSI=PSI/760.
       WRITE (6,650) TTT, PSI
       I = 0
   40  I = I + 1
C      YPD=PERCENTAGE DEVIATION IN Y
       SX = 0.0
       SY = 0.0
C      READ(5,520)TTT,PP(I),(XX(I,J),J=1,NCOMP),(YY(I,J),J=1,NCOMP)
C     1   ,ZZL(I),ZZV(I)
C 520 FORMAT(2F10.4,F11.4,4F10.4)
C      READ( 5,520) PP(I),(XX(I,J),J=1,NCOMP1),(YY(I,J),J=1,NCOMP1)
C     1 ,VVL(I)
       READ (5,660) PP(I), (XX(I,J),J=1,NCOMP1), (YY(I,J),J=1,NCOMP1)
       IF (PP(I) .LE. 0.0) GO TO 90
       IF (NQNTUM .EQ. 1) XX(I,1) = YY(I,1) / XX(I,1)
       IF (NQNTUM .EQ. 2) GO TO 80
   50  CONTINUE
       IF (IJK .EQ. 1) PP(I) = PP(I) * 14.697
       IF (IJK .EQ. 2) PP(I) = PP(I) * 1000. / 6.895
C
       DO 60 J = 1, NCOMP1
       SY = SY + YY(I,J)
   60  SX = SX + XX(I,J)
C
       XX(I,NCOMP) = 1.0 - SX
       YY(I,NCOMP) = 1.0 - SY
       IF (R .LT. 11.0) GO TO 70
C      PP(I)=PP(I)/760.
C      PP(I)=PP(I)/14.697
       PP(I) = PP(I) / 14.697
   70  WRITE (6,670) I, PP(I), (XX(I,J),J=1,NCOMP), (YY(I,J),J=1,NCOMP)
       IF (PP(I) .GE. 0) GO TO 40
   80  CONTINUE
       TEMP = (1. - YY(I,1)) / (XX(I,1) - YY(I,1))
       YY(I,1) = TEMP * XX(I,1)
       XX(I,1) = TEMP
       GO TO 50
   90  NOBS = I - 1
C
       DO 100 I = 1, NOBS
  100  VVL(I) = 0.0
C
       DO 110 I = 1, NCOMP1
       I1 = I + 1
```

```
C
      DO 110 J = I1, NCOMP
  110 READ (5,660) CORRL(I,J)
C
      IIJJKK = 2 * NCOMP + 3
C
      DO 120 IJKL = 1, IIJJKK
      READ (5,660) DUMMY
  120 CONTINUE
C
  130 KIJ = KIJ + 1
      IF (KIJ .GE. 3) GO TO 10
C      IF(KIJ.GT.2) GO TO 800
C      IF(KIJ.GE.2) GO TO 800
      WRITE (6,630) IJK, KIJ, CINI, CFIN
      K = 0
      KK = 0
      KK = 20
  140 CONTINUE
      KK = KK + 1
      WRITE (6,680)
C
      DO 160 I = 1, NCOMP1
      CORRL(I,I) = 0.0
      I1 = I + 1
C
      DO 160 J = I1, NCOMP
      IF (KK .GE. 20) GO TO 150
      CORRL(I,J) = CINI + (KK - 1) * (CFIN - CINI) / 10.
      PSI = PSII
  150 WRITE (6,690) I, J, CORRL(I,J)
      CORRL(J,I) = CORRL(I,J)
  160 CONTINUE
C
      K = 0
C
      DO 170 J = 1, NCOMP
      SEK(J) = 0.0
  170 CONTINUE
C
      SDP = 0.0
      SDV = 0.0
      SSXD = 0.0
      SSYD = 0.0
      SPDPP = 0.0
      RMSYD1 = 0.0
      RMSYD2 = 0.0
      SRMPD = 0.0
      IF (NQNTUM .GE. 10) GO TO 200
C
      DO 180 I = 1, NCOMP
      PC(I) = PCO(I)
      VC(I) = VCO(I)
  180 TC(I) = TCO(I)
C
  190 T = TTT
      RT = R * T
  200 CONTINUE
      NIP = 0
      MIP = 0
```

```
      P = PSI
      K = K + 1
      P = PP(K)
      ZZZV = ZZV(K)
      ZZZL = ZZL(K)
C
      DO 210 J = 1, NCOMP
  210 PHIV(J) = 1.0
C
      DO 220 J = 1, NCOMP
  220 X(J) = XX(K,J)
C
      X1 = X(1)
      XNN = 0.0
      IF (NCOMP .EQ. 2) GO TO 250
C
      DO 230 I = 3, NCOMP
      XNN = XNN + X(I)
  230 CONTINUE
C
  240 CONTINUE
      X(1) = X1
  250 X(2) = 1 - X1 - XNN
  260 CALL SUPT(VL, PHI, PPVOL, C1RKL, C2RKL, CORRL, X, 1, KIJ)
      IF (VL .LE. - 10.) GO TO 130
      JY = 0
  270 IF (JY .GT. 20) GO TO 320
      SUMY = 0.
C
      DO 280 J = 1, NCOMP
      Y(J) = PHI(J) * X(J) / PHIV(J)
  280 SUMY = SUMY + Y(J)
C
      IF (JY) 290, 300, 290
C  60 IF(ABS((SUMYO-SUMY)/SUMY).LE.0.0005) GO TO 61
  290 IF (DABS((SUMYO - SUMY)/SUMY) .LE. .0001) GO TO 320
  300 SUMYO = SUMY
      JY = JY + 1
C
      DO 310 J = 1, NCOMP
  310 Y(J) = Y(J) / SUMY
C
      CALL SUPT(VL, PHIV, PVOL, C1RKL, C2RKL, CORRL, Y, 0, KIJ)
      IF (VL .LE. - 10.) GO TO 130
      GO TO 270
  320 CONTINUE
      DY = SUMY - 1.0
C      IF(ABS(DY).LE.0.0001) GO TO 110
      IF (DABS(DY) .LE. .0001) GO TO 480
      IF (NIP .GT. 100) GO TO 460
      IF (MIP .GT. 20) GO TO 460
      IF (MIP) 330, 330, 390
  330 IF (NIP .LE. 0) GO TO 340
      IF ((DY*DDY) .LT. 0.0) GO TO 400
  340 NIP = NIP + 1
      IF (NIP - 1) 350, 350, 360
  350 X1OLD = X1
      DDY = DY
      X1 = X1 + 0.001
      GO TO 440
```

```
 360 SLOPE = (X1 - X1OLD) / (DY - DDY)
     DELX = SLOPE * DY
C     IF(ABS(DELP)-1.0)79,79,78
     IF (DABS(DELX) - .01) 380, 380, 370
C  78 DELP=SIGN(1.0,DELP)
 370 DELX = DSIGN(1.0D-3,DELX)
 380 X1OLD = X1
     DDY = DY
     X1 = X1 - DELX
     GO TO 440
 390 IF ((DY*DDY) .LT. 0.0) GO TO 400
     IF ((DY*DYO) .LT. 0.0) GO TO 410
C  55 IF(ABS((P-POLD)/P).LE.0.0001) GO TO 105
 400 IF (DABS((X1 - X1OLD)/X1) .LE. 0.0001) GO TO 460
     X1N = (DY*X1OLD - DDY*X1) / (DY - DDY)
     GO TO 420
C  57 IF(ABS((P-PO)/P).LE.0.0001) GO TO 105
 410 IF (DABS((X1 - XO)/X1) .LE. .0001) GO TO 460
     X1N = (DY*XO - DYO*X1) / (DY - DYO)
     GO TO 430
 420 XO = X1OLD
     DYO = DDY
 430 X1OLD = X1
     DDY = DY
     X1 = X1N
     MIP = MIP + 1
C
 440 DO 450 J = 1, NCOMP
 450 Y(J) = Y(J) / SUMY
C
     CALL SUPT(VL, PHIV, PVOL, C1RKL, C2RKL, CORRL, Y, 0, KIJ)
     IF (VL .LE. 10.) GO TO 130
     GO TO 240
C 105 IF(ABS(DY).LE.0.005) GO TO 110
 460 IF (DABS(DY) .LE. .005) GO TO 480
 470 WRITE (6,700)
C     WHEN ATM. UNIT ARE USED THE NEXT TWO CARDS ARE COMMENTED AND 11/ G
C     THE NEXT CARD
 480 P = P * 14.697
C 110 P=P*14.697
C     PP(K)=PP(K)*14.697
     PP(K) = PP(K) * 14.697
     DP = P - PP(K)
C 110 DP=P-PP(K)
C     SDP=SDP+ABS(DP)
     SDP = SDP + DABS(DP)
C     PDPP=ABS(DP)/PP(K)
     PDPP = DABS(DP) / PP(K)
C     SPDPP = SPDPP + ABS(P4PP)
     SPDPP = SPDPP + DABS(PDPP)
     WRITE (6,710) T, P, PP(K), DP, PDPP, K
C    COMMENT FOR ATMOS UNIT
C     PP(K)=PP(K)/14.697
     PP(K) = PP(K) / 14.697
     SXD = 0.0
     SYD = 0.0
C
     DO 500 J = 1, NCOMP
     XD(J) = X(J) - XX(K,J)
     YD(J) = Y(J) - YY(K,J)
```

```
      IF (YY(K,J) .LE. .000005) YPD(J) = 0.0
      IF (YY(K,J) .LE. .000005) EK(J) = 0.0
      IF (YY(K,J) .LE. 0.000005) GO TO 490
      YPD(J) = YD(J) / YY(K,J)
      EK(J) = YD(J) / YY(K,J) * 100.
  490 CONTINUE
      SEK(J) = SEK(J) + DABS(EK(J))
      SXD = SXD + DABS(XD(J))
  500 SYD = SYD + DABS(YD(J))
C
      SSXD = SSXD + SXD
      SSYD = SSYD + SYD
      RMSYD1 = RMSYD1 + YPD(1) ** 2
      RMSYD2 = RMSYD2 + YPD(2) ** 2
      SRMPD = SRMPD + PDPP ** 2
      WRITE (6,720)
C
      DO 510 J = 1, NCOMP
  510 WRITE (6,730) J, COMPA(J), COMPB(J), COMPC(J), COMPD(J), X(J),
     1Y(J), YY(K,J), YD(J), XX(K,J), XD(J), PHI(J), PHIV(J), SUMY, NIP,
     2MIP
C
      DV = VL - VVL(K)
C      SDV=SDV+ABS(DV)
      SDV = SDV + DABS(DV)
      WRITE (6,750) VL, VVL(K), DV
      WRITE (6,740) (EK(J),J=1,NCOMP)
      PSI = P
C      COMMENT THE NEXT CARD IF ATMOS. IS USED
      PSI = PSI / 14.697
C      PSI=PSI/14.697
      IF (K - NOBS) 200, 520, 520
  520 CONTINUE
      DPAV = SDP / NOBS
      DXAV = SSXD / NOBS / NCOMP
      DYAV = SSYD / NOBS / NCOMP
C
      DO 530 J = 1, NCOMP
      SEK(J) = SEK(J) / NOBS
  530 CONTINUE
C
      DVAV = SDV / NOBS
      PDPPAV = SPDPP / NOBS
      RMSYD1 = (RMSYD1/NOBS) ** .5 * 100.
      RMSYD2 = (RMSYD2/NOBS) ** .5 * 100.
      RMSPD = (SRMPD/NOBS) ** .5 * 100.
      WRITE (6,760) NOBS, DPAV, PDPPAV, DYAV, RMSYD1, RMSYD2, RMSPD
      WRITE (6,740) (SEK(J),J=1,NCOMP)
      WRITE (6,780) SDP, SSYD, SDV
      WRITE (6,770) DXAV, DYAV
      IF (KK - 11) 140, 130, 130
  540 STOP
  550 RETURN
  560 FORMAT (20A4)
  570 FORMAT (1H1, 20A4)
  580 FORMAT (3I5, F10.4)
  590 FORMAT (1H0, 6X, 'PC  ', 8X, 'VC  ', 8X, 'TC  ', 8X, 'W', 11X,
     1'WAII', 8X, 'WBII', 7X, 'COMPONENT', //)
  600 FORMAT (6F8.4, 4A4)
  610 FORMAT (1H , 6F12.5, 3X, 4A4)
```

```
  620 FORMAT (2F10.0, I2, 5F10.0)
  630 FORMAT (2I5, 2F10.4)
  640 FORMAT (6(F10.6), 5X)
  650 FORMAT (1H0, 'T=', F10.4, 3X, 'DEG. OF K', 3X, 'P=', F10.4, 3X,
     1' ATM')
  660 FORMAT (2F10.4, F10.4, 5F10.4)
  670 FORMAT (1H , I3, 1F12.5, ' P IN ATM ', 8F12.5)
  680 FORMAT (1H , 1H , 16X, 3HKIJ/)
  690 FORMAT (1H , 6X, 2I2, F10.4, 2F10.4)
  700 FORMAT (1H0, 87HFAILED TO CONVERGE, RESULTS BELOW ARE NOT RELIABLE
     1 IF SUMY IS MUCH DIFFERENT FROM UNITY)
  710 FORMAT (//, 3X, 'T=', F8.2, ' P=', F10.4, ' PP(K)=', F10.4,
     1' DP=', F10.4, 5HPDPP=, F10.4, 15X, 'DATA NO. = ', I4)
  720 FORMAT (/10X, 9HCOMPONENT, 9X, 1HX, 9X, 1HY, 9X, 2HYX, 9X, 2HYD,
     19X, 4HXEPX, 6X, 5H XD  , 6X, 4HPHIL, 7X, 4HPHIV, 2X, 4HSUMY, 3X,
     24HITER, 2X, 3HNIY/)
  730 FORMAT (I2, 4A4, 4F12.4, 4F10.4, F8.4, 2I5)
  740 FORMAT (1H0, 'DEVIATION OF K IN %=', 8F8.3)
  750 FORMAT (15X, 4HVL=  , F10.4, 5HVVL=  , F10.4, 4HDV=  , F10.4)
  760 FORMAT (1H0, I3, 6E16.8)
  770 FORMAT (/10X, 'DXAV(ABS. AVE. DEV.) =', F10.6, 20X,
     1'DYAV(ABS.      AVE. DEV.) =', F10.6///)
  780 FORMAT (/10X, 'ACCUM. SUM OF DEV. IN P IS ', E12.4,
     1'ACCUM. SUM OF     DEV. IN Y IS ', E12.4, 'SDV IS ', E12.4/)
  790 FORMAT (F14.4, 6F10.4)
      END
      SUBROUTINE SUPT(VL, PHI, PVOL, C1RKL, C2RKL, CORRL, X, LV, KIJ)
C      EVALUTION OF SUPPORTING PROPERTIES,THE FUGACITY AND THE
C      PARTIAL MOLAL VOLUME OF FLUID MIXTURES.
C      SHINN-DER CHANG, DEPT.OF CHEM.ENG., UNIV. OF OTTAWA.
      IMPLICIT REAL*8(A - H,O - Z)
      DIMENSION TC(5), PC(5), VCO(5), W(5), AMOLWT(5), ARKL(5,5),
     1BRKL(5), WIJ(5,5), TCOIJ(5,5), TCIJ(5,5), A(4), Z(3), C1RKL(5),
     2C2RKL(5), PVOL(5), PHI(5), CORRL(5,5), X(5), A2I(5), BI(5),
     3AIRKL(5), ALF(5)
      DIMENSION ZZ(3)
      COMMON /FIRST/ TC, PC, VCO, W, AMOLWT, T, P, R, NCOMP, NQNTUM,
     1ZZZL, ZZZV /SECOND/ Z, A, MTYPE /THIRD/ ALF
C      LV=0, FOR VAPOR PHASE.
C      LV=1, FOR LIQUID PHASE.
C      KIJ=1, TCIJ=SQRT(TCI*TCJ)*(1.-KIJ)
C      KIJ=2, AIJ=SQRT(AI*AJ)*(1.-KIJ)
C      KIJ=3, AIJ=(AI*KIJ+(1.-KIJ)*AJ)/2.
      RT = R * T
C
   10 DO 20 I = 1, NCOMP
      ARKL(I,I) = C1RKL(I) * R * R * (TC(I)**2.5) / PC(I) * ALF(I) * (T/
     1TC(I)) ** .5
      BRKL(I) = C2RKL(I) * R * TC(I) / PC(I)
   20 CONTINUE
C
      I = 1
   30 I1 = I + 1
C
      DO 100 J = I1, NCOMP
      IF (KIJ .GT. 1) GO TO 70
      TCIJ(I,J) = (TC(I)*TC(J)) ** 0.5 * (1.0 - CORRL(I,J))
      TCIJ(J,I) = TCIJ(I,J)
      WIJ(I,J) = (W(I) + W(J)) * 0.5
      ZCOIJ = 0.291 - 0.08 * WIJ(I,J)
```

```
   40 VCOIJ = (VCO(I)**(1./3.) + VCO(J)**(1./3.)) ** 3 / 8.0
   50 PCOIJ = ZCOIJ * R * TCIJ(I,J) / VCOIJ
      PCIJ = PCOIJ
   60 ARKL(I,J) = (C1RKL(I) + C1RKL(J)) * 0.5 * R ** 2 * TCIJ(I,J) ** 2.
     15 / PCIJ
      GO TO 90
   70 IF (KIJ .GT. 2) GO TO 80
      ARKL(I,J) = (ARKL(I,I)*ARKL(J,J)) ** 0.5 * (1.0 - CORRL(I,J))
      GO TO 90
   80 ARKL(I,J) = (ARKL(I,I)*CORRL(I,J) + (1. - CORRL(I,J))*ARKL(J,J))
   90 ARKL(J,I) = ARKL(I,J)
  100 CONTINUE
C
      I = I + 1
      IF (I .NE. NCOMP) GO TO 30
      AMRKL = 0.0
      BMRKL = 0.0
C
      DO 110 I = 1, NCOMP
      A2I(I) = ARKL(I,I) / RT ** 2 / T ** 0.5
      BI(I) = BRKL(I) / RT
      BMRKL = BMRKL + X(I) * BRKL(I)
      AIRKL(I) = 0.0
C
      DO 110 J = 1, NCOMP
      AIRKL(I) = AIRKL(I) + X(J) * ARKL(I,J)
  110 AMRKL = AMRKL + X(I) * X(J) * ARKL(I,J)
C
      A2 = AMRKL / RT ** 2 / T ** 0.5
      B = BMRKL / RT
C      WRITE(6,711)BMRKL,     B,R,T,P
      A(1) = 1.0
      A(2) = -1.0
      A(3) = B * P * (A2/B - 1.0 - B*P)
      A(4) = -(A2/B) * (B*P) ** 2
      CALL CUBEQN(MTYPE, A, Z)
      IF (MTYPE) 120, 140, 140
  120 IF (LV .EQ. 0) GO TO 130
C      ZL=AMIN1(Z(1),Z(2),Z(3))
C     ZL=AMIN1(Z(1),Z(2),Z(3))
      ZL = DMIN1(Z(1),Z(2),Z(3))
C      JJ=0
C      DO 146 I=1,3
C      ZZ(I)=9999.
C      JJ=JJ+1
C      IF(Z(I).LT.0.0) GO TO 146
C      ZZ(JJ)=Z(I)
C  146 CONTINUE
C     ZL=AMIN1(ZZ(1),ZZ(2),ZZ(3))
      GO TO 160
C  145 ZL=AMAX1(Z(1),Z(2),Z(3))
  130 ZL = DMAX1(Z(1),Z(2),Z(3))
      GO TO 160
  140 ZL = Z(1)
C      IF(ZL.GT.0.0) GO TO 126
C      ZL=ZZZL
C  126 IF(ZL.LE.1000.) GO TO 123
C      ZL=ZZZV
  150 CONTINUE
  160 H = B * P / ZL
```

```
      IF (1.0 - H) 170, 170, 190
  170 IF (LV .EQ. 0) GO TO 180
      H = 0.98
      GO TO 190
  180 H = 0.02
  190 IF (1.0 + H) 200, 200, 220
  200 IF (LV .EQ. 0) GO TO 210
      H = 0.98
      GO TO 220
  210 H = 0.02
  220 CONTINUE
      VL = RT * B / H
C      WRITE(6,710)LV,ZL,VL,B,H,MTYPE
C      CALL EQNRK(A2,B,P,H,ZL,LV)
      IF (LV .EQ. 0) GO TO 230
      QD = (T**0.5) * VL * (VL + BMRKL)
      QH = (AMRKL/(T**0.5)) * ((2.*VL + BMRKL)/(VL**2*(VL + BMRKL)**2))
      QK = RT / ((VL - BMRKL)**2)
C
  230 DO 250 I = 1, NCOMP
C      WRITE(6,591)ZL,H
      IF (ZL .LT. 0.0) VL = -100.
      IF (ZL .LT. 0.0) WRITE (6,290) ZL, H
      IF (ZL .LT. 0.0) GO TO 260
C      PHILN =(ZL-1.0)*BI(I)/B-ALOG(ZL)-ALOG(1.0-H)
C     1-(A2/B)*(2.* AIRKL(I)/AMRKL      -BI(I)/B)*ALOG(1.0+H)
      PHILN = (ZL - 1.0) * BI(I) / B - DLOG(ZL) - DLOG(1.0 - H) - (A2/B)
     1 * (2.*AIRKL(I)/AMRKL - BI(I)/B) * DLOG(1.0 + H)
C      PHI(I)= EXP(PHILN )
      PHI(I) = DEXP(PHILN)
C      WRITE(6,592)PHI(I),LV
      IF (LV .EQ. 0) GO TO 250
      QE1 = 0.0
C
      DO 240 J = 1, NCOMP
  240 QE1 = QE1 + X(J) * ARKL(I,J)
C
      QE = 2.0 * QE1 - AMRKL * BRKL(I) / (VL + BMRKL)
      QG = (RT/(VL - BMRKL)) * (1.0 + BRKL(I)/(VL - BMRKL))
      PVOL(I) = ((QE/QD) - QG) / (QH - QK)
  250 CONTINUE
C
  260 RETURN
  270 FORMAT (3X, 5E12.5)
  280 FORMAT (3X, 'LV= ', I3, 2X, 'ZL= ', E12.5, 2X, 'VL= ', E12.5, 2X,
     1'B= ', E12.5, 2X, 'H= ', E12.5, 2X, 'MTYPE= ', I3)
  290 FORMAT (1H0, 'ZL = ', 2F16.8)
  300 FORMAT (/3X, E12.5, 2X, I3)
      END
      SUBROUTINE CUBEQN(INDEX, LI, LJ)
C    SOLVING EQN F(Z)=A(1)*Z**3+A(2)*Z**2+A(3)*Z+A(4)=0
C    BY SHINN-DER CHANG ( DEPT OF CHEM ENG , UNIV OF OTTAWA )
      IMPLICIT REAL*8(A - H,O - Z)
C      REAL*4 LI(4),LJ(3)
      REAL*8 LI(4), LJ(3)
      DIMENSION A(4), Z(3)
C
      DO 10 I = 1, 4
   10 A(I) = LI(I)
C
```

```
      P = (A(3) - A(2)**2/A(1)/3.0D0) / A(1)
      Q = 2.0D0 * (A(2)/3.0D0/A(1)) ** 3 - A(3) * A(2) / 3.0D0 / A(1) **
     1 2 + A(4) / A(1)
      DE = (P/3.0D0) ** 3 + (Q/2.0D0) ** 2
      T = A(2) / 3.0D0 / A(1)
      IF (DE .GE. 0.0D0) GO TO 20
      THI = DARCOS(-(Q/2.0D0)/DSQRT(-(P/3.0D0)**3))
      R = 2.0D0 * DSQRT(-P/3.0D0) * DCOS(THI/3.0D0)
      GO TO 70
   20 S = (-(Q/2.0D0) + DSQRT(DE))
      U = (-Q/2.0D0-DSQRT(DE))
      IF (S) 30, 40, 30
   30 S = S / DABS(S) * (S*S) ** (1.0D0/6.0D0)
   40 IF (U) 50, 60, 50
   50 U = U / DABS(U) * (U*U) ** (1.0D0/6.0D0)
   60 R = S + U
   70 DISC = R * R - 4.0D0 * (P + R*R)
      IF (DISC) 100, 80, 90
   80 INDEX = 0
      Z(1) = R - T
      Z(2) = -R / 2.0D0-T
      Z(3) = Z(2)
      GO TO 110
   90 INDEX = -1
      Z(1) = R - T
      Z(2) = -R / 2.0D0+DSQRT(DISC) / 2.0D0-T
      Z(3) = -R / 2.0D0-DSQRT(DISC) / 2.0D0-T
      GO TO 110
  100 INDEX = 1
      Z(1) = R - T
      DB = DSQRT(DABS(DISC)) / 2.0D0
      DA = -R / 2.0D0-T
      Z(2) = Z(1)
      Z(3) = Z(1)
C      WRITE(6,100)DA,DB
  110 DO 120 I = 1, 3
  120 LJ(I) = Z(I)
C
      RETURN
  130 FORMAT (/' THERE ARE TWO CONJUGATE ROOTS , REAL PART= ', E15.6/27X,
     1' IMAG. PART= ', E15.6/)
      END
```

PROGRAM E-4

Computer Program Used to Correlate

Binary Interaction Coefficient C_{ij}

```
C ***************************************************************
C
C PROGRAM E-4
C
C COMPUTER PROGRAM USED TO CORRELATE BINARY INTERACTION
C COEFFICIENT, CIJ VALUES
C
C ***************************************************************
C
      IMPLICIT REAL*8(A - H,O - Z)
      DIMENSION A(40,40), B(40,40), X(100), Y(100,1),
     1C(40,40), YC(100), DY(100), YCAL(100), YORIG(100,1),
     2DYO(100), XX(100), TITLE(20), EDY(100), WW(100)
C     ***************************************************************
   10 READ (5,240,END=230) W, XK
C     ***************************************************************
      IF (W .LT. 0.) GO TO 230
      L = 1
      M = 5
      M = 6
      M = 11
      M = 9
      M = 4
      M = 3
      M = 0
      M = 2
      M = 1
C     ***************************************************************
      READ (5,250) (TITLE(I),I=1,19)
C     ***************************************************************
      WRITE (6,260) (TITLE(I),I=1,19)
C     ***************************************************************
C     ***************************************************************
      WRITE (6,270) W
      READ (5,280) T, R, NI, NNI, NT
C     NT = 0 TEMP. IN DEGREE K
C     NT = 1 TEMP. IN DEGREE C
C     NT = 2 TEMP IN DEGREE F
C     NT = 3 TEMP IN DEGREE R
      NT = NT + 1
      GO TO (50, 40, 30, 20), NT
   20 T = T / 1.8
      GO TO 50
   30 T = (T - 32.) * 5. / 9. + 273.15
      GO TO 50
   40 T = T + 273.15
   50 CONTINUE
      NI = NI + 1
C     ***************************************************************
C     NI = 0 INPUT IS GE, X(1)
C     NI = 1 INPUT IS ACTIVITY COEFFICIENTS AND X(1)
C     NI = 2 INPUT IS LOG10(ACTIVITY COEF.|S) AND X(1)
C     NI = 3 INPUT IS NATURAL LOG(ACTIVITY COEF.|S) AND X(1)
C     NI = 4 INPUT IS SMOOTHED DATA OF LOG10(ACTIVITY COEF.'S) IN THE
C FORM OF
C     A EQ. WITH COEFFICIENTS B12, C12, D12 AND X(1)
C     NNI = 0      INPUT GE IS GE
C     NNI = 1      INPUT GE IS GE/R/T
C     R IS 1.987 IF GE IN CAL/MOLE
C     R IS 8.314 IF GE IN J./MOLE
```

```
      IF (NI .EQ. 5) READ(5,240) B12, C12, D12
C     ****************************************************************
      IF (NI .EQ. 5) WRITE (6,310) B12, C12, D12
      WRITE (6,290) T, R
C     ****************************************************************
      IF (XK .LT. .0) READ (5,240) TC
C     ****************************************************************
      I = 1
   60 GO TO (70, 80, 80, 80, 80), NI
C     ****************************************************************
   70 READ (5,300) X(I), (Y(I,J),J=1,L)
C     ****************************************************************
      IF (XK .LT. .0) X(I) = X(I) / TC
      IF (X(I) .LT. - 3.0) GO TO 110
C     X(I) = 1.0/X(I)
      WW(I) = 1.
      GO TO 100
C     ****************************************************************
   80 READ (5,240) E, F, X(I)
C     ****************************************************************
      IF (XK .LT. .0) X(I) = X(I) / TC
      IF (X(I) .EQ. 1.0 .OR. X(I) .EQ. 0.0) GO TO 60
      IF (X(I) .LT. .0) GO TO 110
      WW(I) = 1.
      IF (NI .EQ. 2) E = DLOG(E)
      IF (NI .EQ. 2) F = DLOG(F)
      IF (NI .EQ. 5) E = X(I) ** 2 * (B12 + C12*(4.*X(I)
     11.) + D12*(2.*X(I) - 1.)*(6.*X(I) - 1.))
      IF (NI .EQ. 5) F = X(I) ** 2 * (B12 + C12*(4.*X(I)
     13.) + D12*(2.*X(I) - 1.)*(6.*X(I) - 5.))
C
      DO 90 J = 1, L
      Y(I,J) = (X(I)*E + F*(1. - X(I))) * R * T
      IF (NI .EQ. 3) Y(I,J) = Y(I,J) * 2.303
      IF (NI .EQ. 5) Y(I,J) = Y(I,J) * 2.303
      Y(I,J) = Y(I,J) / R / T / X(I) / (1. - X(I))
   90 CONTINUE
C
  100 WRITE (6,310) X(I), Y(I,1)
      I = I + 1
      GO TO 60
  110 N = I - 1
C
      DO 120 I = 1, N
  120 C(I,1) = 1.0
C
      MP1 = M + 1
C
      DO 130 J = 2, MP1
C
      DO 130 I = 1, N
  130 C(I,J) = C(I,J - 1) * X(I)
C
      DO 140 I = 1, MP1
C
      DO 140 J = 1, MP1
      A(I,J) = 0.
C
      DO 140 K = 1, N
  140 A(I,J) = A(I,J) + C(K,I) * C(K,J) * WW(K)
```

```
C
      DO 150 J = 1, L
C
      DO 150 I = 1, MP1
      B(I,J) = 0.
C
      DO 150 K = 1, N
  150 B(I,J) = B(I,J) + C(K,I) * Y(K,J) * WW(K)
C
      CALL CHAPB(A, MP1, B, L, DET)
      WRITE (6,320)
      WRITE (6,330) N
      WRITE (6,340) M
C
      DO 160 J = 1, L
C
      DO 160 I = 1, MP1
      II = I - 1
  160 WRITE (6,360) II, B(I,J)
C
      SEDY = 0.
      SDY = 0.0
      XMEDY = 0.
      N3 = N + 3
      X(N + 1) = 273.15 - 90.04
      X(N + 2) = 273.15 - 99.91
      X(N + 3) = 273.15 - 109.99
C
      DO 200 J = 1, N3
      YC(J) = 0.
C
      DO 170 IL = 1, L
C
      DO 170 I = 1, MP1
      II = I - 1
      IF (X(J) .EQ. 0.0) GO TO 210
      YC(J) = YC(J) + B(I,IL) * X(J) ** II
  170 CONTINUE
C
  180 CONTINUE
      DY(J) = YC(J) - Y(J,IL)
      DY(N + 1) = 0.0
      DY(N + 2) = 0.0
      DY(N + 3) = 0.0
      IF (DABS(Y(J,IL)) .LE. 0.0000001) GO TO 220
      EDY(J) = (DY(J)/Y(J,IL)) * 100.
  190 IF (XMEDY .LT. DABS(EDY(J))) XMEDY = DABS(EDY(J))
      SEDY = SEDY + DABS(EDY(J))
      SDY = SDY + DABS(DY(J))
  200 WRITE (6,370) J, YC(J), J, DY(J), J, EDY(J), J, X(J)
C
      EDYAV = SEDY / N
      SDYAV = SDY / N
      WRITE (6,380) SDY, EDYAV, SDYAV
      WRITE (6,390) XMEDY
      GO TO 10
  210 YC(J) = B(1,IL)
      GO TO 180
  220 EDY(J) = 0.0
      GO TO 190
```

```
  230 RETURN
  240 FORMAT (8F10.4)
  250 FORMAT (19A4)
  260 FORMAT (1H1, 19A4)
  270 FORMAT (1H0, '   W=', D16.10)
  280 FORMAT (2F10.4, 5I2)
  290 FORMAT (1H0, '   T=', F16.8, '   R=', F16.10)
  300 FORMAT (4D20.8)
  310 FORMAT (1H0, 3D20.14)
  320 FORMAT (1H0, '-------------------------------------------'
     1)
  330 FORMAT (1H , ' THE NUMBER OF GIVEN DATA POINTS=', I3)
  340 FORMAT (1H , ' THE DEGREE OF POLYNOMIAL=', I3)
  350 FORMAT (4F20.8)
  360 FORMAT (1H0, I3, '   DEGREE COEFFICIENT= ', D20.14)
  370 FORMAT (1H0, '   YC(', I2, ')=', D20.14, '   DY(', I2,
     1')=', D20.14, '   EDY(', I2, ')=', D20.14, '%', '   X(',
     2I2, ')=', D10.5, /)
  380 FORMAT (1H0, ' SUM OF DY=', D20.14, '   AVE. D.Y.=',
     1D20.14, '%,   AVE. D.Y.=', D20.14)
  390 FORMAT (1H0, ' MAX. DEVIATION = ', D20.14)
      END
C THIS SUBROUTINE IS FOR MATRIX INVERSION AND SIMULT. LINEAR EQS.
      SUBROUTINE CHAP8(A, N, B, M, DET)
      IMPLICIT REAL*8(A - H,O - Z)
      DIMENSION A(40,40), B(40,40), IPVOT(40), INDEX(40,2),
     1PIVOT(40)
      COMMON IPVOT, INDEX, PIVOT
      EQUIVALENCE (IROW,JROW), (ICOL,JCOL)
C     FOLLOWING 3 STATEMENTS FOR INITIALIZATION
   10 DET = 1.
C
      DO 20 J = 1, N
   20 IPVOT(J) = 0
C
      DO 210 I = 1, N
C       FOLLOWING 12 STATEMENTS FOR SEARCH FOR PIVOT ELEMENT
      T = 0.
C
      DO 70 J = 1, N
      IF (IPVOT(J) - 1) 30, 70, 30
C
   30 DO 60 K = 1, N
      IF (IPVOT(K) - 1) 40, 60, 260
   40 IF (DABS(T) - DABS(A(J,K))) 50, 60, 60
   50 IROW = J
      ICOL = K
      T = A(J,K)
   60 CONTINUE
C
   70 CONTINUE
C
      IPVOT(ICOL) = IPVOT(ICOL) + 1
C     FOLLOWING 15 STATEMENT TO PUT PIVOT ELEMENT ON DIAGONAL
      IF (IROW - ICOL) 80, 120, 80
   80 DET = -DET
C
      DO 90 L = 1, N
      T = A(IROW,L)
      A(IROW,L) = A(ICOL,L)
```

```
  90 A(ICOL,L) = T
C
      IF (M) 120, 120, 100
C
 100 DO 110 L = 1, M
      B(IROW,L) = B(ICOL,L)
      T = B(IROW,L)
 110 B(ICOL,L) = T
C
 120 INDEX(I,1) = IROW
      INDEX(I,2) = ICOL
      PIVOT(I) = A(ICOL,ICOL)
      DET = DET * PIVOT(I)
C       FOLLOWING 6 STATEMENTS TO DIVIDE PIVOT ROW BY PIVOT ELEMENT
      A(ICOL,ICOL) = 1.
C
      DO 130 L = 1, N
 130 A(ICOL,L) = A(ICOL,L) / PIVOT(I)
C
      IF (M) 160, 160, 140
C
 140 DO 150 L = 1, M
 150 B(ICOL,L) = B(ICOL,L) / PIVOT(I)
C     FOLLOWING 10 STATEMENTS TO REDUCE NON-PIVOT ROWS
 160 DO 210 LI = 1, N
      IF (LI - ICOL) 170, 210, 170
 170 T = A(LI,ICOL)
      A(LI,ICOL) = 0.
C
      DO 180 L = 1, N
 180 A(LI,L) = A(LI,L) - A(ICOL,L) * T
C
      IF (M) 210, 210, 190
C
 190 DO 200 L = 1, M
 200 B(LI,L) = B(LI,L) - B(ICOL,L) * T
C
 210 CONTINUE
C      FOLLOWING 11 STATEMENTS TO INTERCHANG5&3oLUMNS
 220 DO 250 I = 1, N
      L = N - I + 1
      IF (INDEX(L,1) - INDEX(L,2)) 230, 250, 230
 230 JROW = INDEX(L,1)
      JCOL = INDEX(L,2)
C
      DO 240 K = 1, N
      T = A(K,JROW)
      A(K,JROW) = A(K,JCOL)
      A(K,JCOL) = T
 240 CONTINUE
C
 250 CONTINUE
C
 260 RETURN
      END
```