ABSTRACT

Experimental vapor-liquid equilibrium data for the binary mixture argon-ethane at 115.5°K and the ternary mixture argon-methane ethane at 115.5°K were obtained by means of a forced circulation method. In addition, a total pressure method was used to determine vapor-liquid equilibrium data for the binary system argon-methane at 115.22°K, nitrogen-argon at 110°K, and ethylene-propane at four isothermal conditions.

The Clausius equation of state has been modified by considering its parameters temperature-dependent. Vapor pressure and saturated molar volumes of liquid and vapor for pure substances were used for evaluating these parameters. The equation was successfully employed in calculation of compressibilities in the homogenous region for pure nitrogen, oxygen and argon. The absolute average deviation was found to be 0.07. The modified Clausius equation of state was successfully applied in calculation of phase equilibria, for systems at high pressure, from total pressure-liquid phase composition measurements. The absolute average deviation in the vapor phase composition (more volatile component) was found to be 0.1 mole percent. Also, the modified equation was used to predict phase equilibrium data in the binary and multicomponent system containing the components nitrogen, oxygen, argon, methane, carbon dioxide, propane, n-butane and hydrogen sulphide. The agreement obtained between the predicted and literature was good.

For systems at moderate pressure, (approx. 50-200 lb/sq. in.) a method was proposed for computing vapor phase composition from total pressure-liquid phase composition measurements. In this method the hypothetical component concept was employed and thus extended a method for binary systems to ternary systems at isothermal conditions.

The proposed method was successfully applied to the ternary system argon-methane-ethane.

An analysis technique was proposed for the evaluation of calibration constant using gas chromotography in the absence of pure components. This situation applies, for example, to the determination of equilibrium composition of liquid and vapor phase in a phase equilibrium study.

Moreover, vapor-liquid equilibrium data available in the literature for binary and ternary mixtures of nitrogen, argon, and oxygen were tested and screened over the range from 90 to 120°K. Activity coefficients for the consistent isothermal binary data were correlated by means of the Redlich-Kister equation. The binary constants were successfully employed for predicting the ternary data, without requiring a ternary constant.

ACKNOWLEDGEMENT

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The author also wishes to express his gratitude to Faculty members of the Department of Chemical Engineering for their helpful discussions and to Mr. G. Gasperetti for his assistance in constructing and improving the experimental apparatus.

Finally, I would like to express my most sincere appreciation to my wife Samia, whose faith and encouragement made this work possible.

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NOMENCLATURE

A, B, C, a, b, c = parameters in the Clausius equation of state

a, b = parameters in the R-K equation of state

B_{12}, C_{12}, D_{12}
B_{23}, C_{23}, D_{23} = constants in the Redlich-Kister equation for binary systems
B_{31}, C_{31}, D_{31}

f = fugacity

H, h = dummy parameters in the Clausius equation of state (Equation (4-37))

K = vaporization equilibrium ratio, y/x

k = temperature-independent parameter in Equation (4-54)

N = number of components in a system

n = total number of moles

P = pressure

p = vapor pressure of pure component

R = gas constant

T = temperature

v = volume

x = liquid mole fraction

y = vapor mole fraction

Z = compressibility factor

β = second virial coefficient

γ = activity coefficient

Δ = difference

φ = fugacity coefficient

μ = chemical potential

Ω_a, Ω_b = characteristic parameters of the R-K equation of state

Ω_a, Ω_b, Ω_c = characteristic parameters of the modified Clausius equation of state

ω = acentric factor

Superscripts

° = reference state

^ = fugacity of fugacity coefficient in the mixture

— = partial molal quantity

~ = molal quantity

E = excess property

Subscripts

c = critical property

cv = convergence

i, j, 1, 2, 3 = component identification

L = liquid phase

V = vapor phase

R = reduced state

CHAPTER I

INTRODUCTION

An adequate knowledge of the relevant equilibria data is a basic requirement for the successful design of separation equipment. Such equilibria data are also increasingly important in feasibility studies of separation and in the interpretation of the operation and performance of existing industrial plants.

At the present time with an ever increasing number of complex vapor-liquid equilibria problems arising from numerous new industrial processes there is a need for more rapid methods in the determination and prediction of vapor-liquid equilibria data. Such rapid evaluation are being greatly aided by the use of improved analytical techniques and by the increasing use of digital computers. Computers have removed the tedium from phase equilibria calculations and now make possible the rapid application of various consistency tests and correlating and predicting equations in the binary and multi-component systems. Further more, when only limited vapor-liquid equilibrium data or indirect physical measurements are available, computers can be of considerable value in searching for and finding the necessary parameters required in various predicting equations.

Phase equilibria data can be predicted using two approaches. First, the statistical thermodynamic approach which seeks to establish relations between macroscopic properties and intermolecular forces through partition functions. However, useful configurational partition functions have been constructed only for nearly ideal situations. Therefore, this approach is at present insufficient for many practical purposes.

Second, the empirical approach which is based on classical thermodynamic but relying on molecular physics and statistical thermodynamic to supply insight into the behavior of matter.

In this work we have modified the total pressure method, previously proposed by Ho(1), to predict vapor phase composition for ternary systems at moderate pressures using the hypothetical "pure" component approach.

Since our ultimate aim is to use our theoretical knowledge, to make predictions of phase equilibria of mixtures on the basis of those of pure components. For this purpose, an adequate equation of state must satisfy the three conditions at saturation. Therefore, an equation of state suitable for this kind of calculation should contain a minimum of three constants.

Although there are few three-constant equations of state(2, 3) available in the literature, none of them is capable of describing the behavior of pure substances perfectly. The Clausius equation of state(2) appears promising, because it does reproduce so perfectly the qualitative behavior of substances that the belief is justified that the basis upon which it rests is not wholly incorrect, but rather not sufficiently complete. Therefore, in this work, an attempt has been made to improve the Clausius equation of state by considering it's parameters temperature dependent. With this modification, the Clausius equation of state was found to be capable of predicting pure component compressibilities in the homogeneous region, as well as phase equilibria data for mixtures.

The experimental part of this work discusses the design of a low temperature phase equilibrium apparatus and determinations were made for the argon-methane, nitrogen-argon, and ethylene-propane systems. Also, a forced circulation apparatus was used for the determination of vapor-liquid equilibrium data for the binary system argon-ethane and the ternary system argon-methane-ethane.

CHAPTER II

LITERATURE REVIEW

2.1 VAPOR-LIQUID EQUILIBRIA DATA

In this work, new experimental data are reported for binary and ternary systems made of air components and hydrocarbon gases. For this reason a review of previous experimental determination was made for this class of mixtures.

A complete literature survey up to October 1959 was prepared by Flynn(4). It gave a bibliography of phase equilibria data of cryogenic fluids, such as helium, nitrogen, methane etc. Another review has been recently prepared by Ruhemann and Harmens(5). In this survey, which covers the period up to 1966, phase equilibria data of low boiling mixtures are classified into 8 groups. In addition, the authors of this survey draw a special attention to the extensive bibliography published in Russia in 1963 by Malkov et al.(6). This survey may be summarized in Table 2-1. More over, the book by Hala, Pick, Fried and Vilim(7) provides useful references. In the third part of this book a review of the literature is given which covers the period up to 1965. In view of the fact that these reviews are quite exhaustive and up to date, in this work a brief review has been done and the current publications will be quoted accordingly.

2.2 PREDICTION METHODS

One of the most practised applications of vapor-liquid equilibria calculations is in the prediction of binary vapor-liquid equilibria from indirect measurements or knowledge of other physical properties.

TABLE 2-1

SUMMARY OF THE BIBLIOGRAPHY PREPARED BY RUHEMANN AND HARMENS(5)

Systems Containing	No. of Ref.	Pressure Range in Bars	Temperature Range in °K
1. Atmospheric gas	26	0.55 - 26	54 - 139
2. Carbon dioxide	16	1 - 180	87 - 384
3. Hydrogen sulfide	8	1 - 130	90 - 422
4. Hydrocarbons only	31	1 - 690	89 - 510
5. Hydrogen and hydrocarbons	18	7 - 550	67 - 336
6. Nitrogen and/or carbon monoxide with hydrogen and/or hydrocarbons	22	0.5 - 200	25 - 363
7. Helium	18	0.07 - 300	15.5 - 358
8. Miscellaneous	8	0.12 - 1000	67 - 395

Such predicted data are not only important for binary separation studies, but also are the starting point for multi-component vapor-liquid equilibria calculations.

It is the custom to characterize vapor-liquid equilibrium (particularly for technically important systems) by the equilibrium ratio K, which is defined by the relation

$$K_i = \frac{y_i}{x_i} \tag{2-1}$$

In general K_i may be calculated from the following equation:

$$K_i = \frac{\gamma_i^L \; f_i^L}{\gamma_i^V \; f_i^V} \tag{2-2}$$

where:

x_i = mole fraction of component i in the liquid phase

y_i = mole fraction of component i in the vapor phase

f_i^L = liquid fugacity of pure component i of the temperature and pressure of the mixture

f_i^V = vapor fugacity of pure component i at the temperature and pressure of the mixture

γ_i^L = liquid phase activity coefficient

γ_i^V = vapor phase activity coefficient

Equation (2-2) is a general equation applicable to non-ideal systems at high pressure.

Since vapor-liquid phase equilibria ratios (K - values) were introduced over 30 years ago a large number of graphical correlations have been published to provide engineering estimates of the K - values to be expected under a wide range of operating conditions.

Computation methods may be divided into:

Composition-independent K - values

For composition-independent K - values the liquid phase activity coefficient is equal to unity and unless the pressure is greater than 500 lb. /sq. in. the vapor-phase activity coefficient is usually unity and consequently Equation (2-1) reduces to:

$$K_i = \frac{y_i}{x_i} = \frac{f_i^L}{f_i^V} \qquad (2\text{-}3)$$

At pressures less than 50 lb. /sq. in. it is usually to replace the fugacity by pressure terms such that:

$$K_i = \frac{y_i}{x_i} = \frac{p^\circ_i}{P} \qquad (2\text{-}4)$$

where:

p°_i = vapor pressure of the pure component

P = total pressure of the system

For pressures in the range of 50 lb. /sq. in. to 200 lb. /sq. in. Equation (2-4) may be modified as follows to take into account deviations from the ideal gas laws:(8)

$$K_i = \frac{y_i}{x_i} = \frac{p^\circ_i}{P} \cdot \text{Exp}\left[\frac{(V_{iL} - \beta_{ii})(P - p^\circ_i)}{RT}\right] \qquad (2\text{-}5)$$

where:

V_{iL} = molal liquid volume of pure component i

β_{ii} = second virial coefficient of pure component i

According to the theory of corresponding states for simple fluids, pure components manifest the same fugacity coefficient, (f/P), when measured at the same reduced conditions of pressure and temperature. Thus:

$$f/P = F(P_R, T_R) \qquad (2\text{-}6)$$

where:

P_R = reduced pressure

T_R = reduced temperature

Based on this principle, generalized charts have been developed in the form of (f/P) plotted against P_R and T_R by Lewis and Kay$^{(9)}$, Newton$^{(10)}$, and Souders, Selheimer, and Brown$^{(11)}$.

In these methods the vapor-phase fugacity is found by the equation:

$$\ln \frac{f_i^V}{P} = \int_0^{P_R} (Z - 1) \frac{dP_R}{P_R} \qquad (2\text{-}7)$$

where:

Z = compressibility factor.

Using an equivalent fugacity coefficient chart, while the fugacity of the liquid phase is found from the same generalized chart using the vapor pressure p_i° in the fugacity coefficient and in the definition of the reduced pressure. In this application of Souders, Selheimer, and Brown method$^{(11)}$ a correction is made for the variation of the liquid-phase fugacity with pressure between p_i° and P by the relationship:

$$(f_i^L)_P = (f_i^L)_{p_i^\circ} \cdot \text{Exp}\left[\frac{V_i^L (P - p_i^\circ)}{RT}\right] \qquad (2\text{-}8)$$

In many cases the two-parameter theorem of corresponding states is too much of an approximation, as (f/P) is not an exact function of P_R and T_R. Equation (2-6) is based on simple fluids but because of orientation effects and clustering of the molecules deviation can occur.

The above mentioned generalized charts are based on P-V-T data for low molecular weight hydrocarbons, with close boiling point, but some experimental equilibrium data for mixtures of hydrocarbons are included.

For application to wider range of hydrocarbon mixtures and to allow for deviation from ideal pure-component fugacities efforts have been made to introduce one or two additional parameters to improve the theorem of corresponding states$^{(12-15)}$. Hougen and Watson$^{(16)}$ introduced a critical compressibility factor, as a third parameter and with modification, the theorem of corresponding states becomes as follows:

$$f/P = F(P_R, T_R, Z_c) \qquad (2\text{-}9)$$

where:

Z_c = the compressibility factor at the critical state

Generalized tables and fugacity coefficients of pure components are given by Hougen et al. at $Z_c = 0.27$ as a function of T_R and P_R. The fugacity coefficients for substances whose compressibility is other than 0.27 may be calculated by the use of deviation or correction terms, D, which are also tabulated. The fugacity coefficient is then obtained from the following equation:

$$f/P = (f/P)_{0.27} \cdot 10^{D(Z_c - 0.27)} \qquad (2\text{-}10)$$

Another three parameter generalized correlation for pure substances has been used by Edmister(17, 18) and is of the form:

$$(f/P) = (f/P)^0 \left[(f/P)^1\right]^{\omega} \qquad (2\text{-}11)$$

where:

$(f/P)^0$ = values of (f/P) for a simple fluid

$(f/P)^1$ = correction for the deviation of values of (f/P) from that of a simple fluid

ω = acentric factor(18)

The acentric factor is defined by:

$$\omega = -\log P_R - 1.0 \qquad (2\text{-}12)$$

where:

P_R = reduced vapor pressure at the reduced temperature 0.7.

Generalized charts were prepared for $(f/P)^0$ and $(f/P)^1$ as function of P_R and T_R.

Several other third parameters have been tried(12) such as the reduced dipole moment, bond length, and normal heat of vaporization, but according to Hougen and Watson these parameters give less favourable results. Reidel(14) introduced the slope of vapor pressure at the critical temperature as the third parameter. Bloomer and Peck(19) used the slope of the critical isometric curve on a P-T chart as the third parameter. For polar substances, Black(20, 21) has shown that one additional parameter is required.

Composition-dependent K - values

The effect of composition can be incorporated into K - value definition (Equation 2-1) as follows:

$$K_i = \frac{y_i}{x_i} = \frac{\hat{\phi}_{iL}}{\hat{\phi}_{iV}} = \frac{\gamma_i \ f_i^\circ}{\hat{\phi}_{iV} \ P} \tag{2-13}$$

where:

$\hat{\phi}_{iL}$ and $\hat{\phi}_{iV}$ = fugacity coefficients of component i in liquid and vapor phase respectively

f_i° = fugacity of pure component i in the reference state

In the use of Equation (2-13) the effects of deviation from the ideal gas laws are incorporated in the vapor phase fugacity coefficient, $\hat{\phi}_{iV}$, whereas for the liquid phase these deviations are separated into the fugacity coefficient $\hat{\phi}_{iL}$ or fugacity $\hat{f}_{iL}$ and liquid phase activity coefficient, γ_i, depending on the method used for calculation.

Other methods of correlation and prediction include the use of convergence pressures and equations of state, but undoubtedly the most common methods involve the evaluation of liquid and vapor phase activity and fugacity coefficients.

Methods of calculation of the composition-dependent K-values may be conveniently classified into two main groups:

i) <u>Convergence Pressure Method</u>

The convergence pressure method(22-27) has its basis in the mathematical statement that the K - ratio at equilibrium is dependent upon four parameters:

$$K = F(x, T, P, P_{conv.}) \tag{2-14}$$

where:

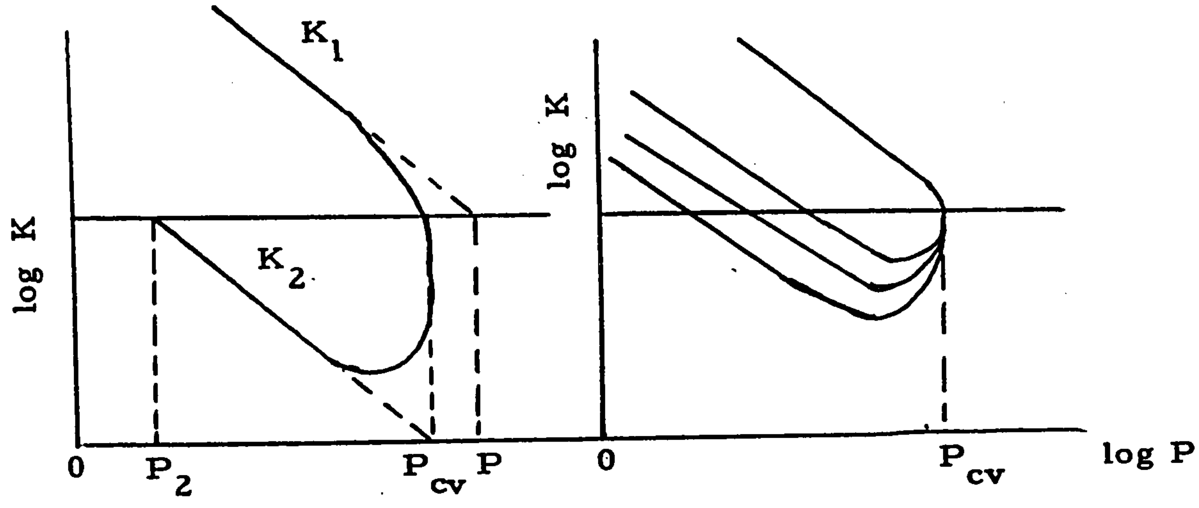
x = composition of component

T = system temperature

P = system pressure

$P_{conv.}$ = convergence pressure

The convergence pressure, $P_{conv.}$, is defined as the pressure at which the K - values for all components become unity simultaneously in isothermal plot of log K vs. log P as shown in Figure (2-1).



Binary system, T = constant Complex system, T = constant

Fig. 2-1

Hadden(23, 24), Lenoir(27) and White(25) have discussed methods for predicting the convergence pressure.

The National Gasoline Association of America(28), (N. G. A. A.), has proposed charts containing plots of log K against log P for each of several convergence pressures. There are a total of 135 charts with convergence pressure ranging from 600 to 20, 000 lb. /sq. in. . The good agreement between K - values based on the N. G. A. A. charts and those given by Kellogg charts(29) is taken as supporting evidence for the validity of the proposed method.

ii) Equation of State

A detailed review on the equations of state was prepared by many authors(30-35).

Of the many empirical equations of state, the Benedict-Webb-Rubin(36), B. W. R. and the Redlich-Kwong Equation(37) are the most widely used in the calculation of vapor-liquid equilibria.

The B. W. R. equation of state containing eight constants relating pressure and density applies to liquid and vapor phases and gives fairly accurate results for mixtures of similar compounds. This equation is widely applied in phase equilibrium studies for hydrocarbons of low and moderate molecular weight because of its reliability in this area. The results have been summarized in 350 charts for 12 hydrocarbons and are known as the Kellogg charts(28). The number of Kellogg charts required to present the data has been reduced by De Priester(38), and by Edmister and Ruby(15).

In addition to the B. W. R. equation of state the two-constant Redlich-Kwong equation of state(37), R-K, has been applied extensively to phase equilibria calculations. Numerous modifications on the R-K equation have been proposed(39-42) to improve the agreement between the experimental and predicted equilibria properties for mixtures.

The literature shows that, only two equations of state are widely used in the calculation of phase equilibria. They are the B. W. R. and R-K equations of state. A comparison between the two equations was prepared by Hsi(43) which was in favour of R-K equation of state. As a result, the R-K equation of state appears to possess a combination of advantages in simplicity and reasonable accuracy over wide ranges of temperature and pressure.

2. 3 EXPERIMENTAL DETERMINATION OF EQUILIBRIA DATA

Many types of equilibrium stills have been reported in the literature for determination of vapor-liquid equilibria. The most commonly used are the forced circulation and the static methods. A comprehensive and well illustrated review of equilibrium stills is given by Hala(7) et al. These methods of determination can be calssified into the following groups:

a) Forced Circulation Method

This is a common method for obtaining vapor-liquid equilibrium data. In this method the vapor phase is circulated through the liquid phase until no further change in the composition of vapor and liquid phases takes place. Circulation of the vapor phase is achieved by means of a mercury pump(44) or an electro-magnetic pump(45, 46).

b) Dew-Point and Bubble-Point Method(47-50)

This technique consists of introducing a mixture of known composition into an evacuated container of variable volume. The system is maintained at a constant temperature, and by varying the volume, the pressure is obtained at which condensation commences and completed. From such data, the saturation curves of two phases at constant temperature can be plotted against composition.

The main disadvantages are that the dew-point and bubble-point are not sharply defined, hence they must be made with highly refined precision instruments, and the materials used should be pure and free from traces of gases.

c) Flow Method(5, 51, 54)

Another method that has been widely used for the determination of vapor-liquid equilibrium data, is one which the vapor is passed through a series of vessels containing a liquid of suitable composition. The vapor entering the vessel may be of composition somewhat different from the equilibrium vapor, but as it passes through the system it tends to approach equilibrium. It is obvious that it cannot be an exact equilibrium because of the fact that a pressure drop is involved in passing the vapor through the system, ie. there are pressure variation which will effect the equilibrium. There is also the danger of entrainment.

d) Static and Total-Pressure Methods(55)

In these methods the liquid mixture is introduced into a closed evacuated vessel. It is then agitated at a constant temperature, either by shaking the vessel or using a magnetic stirrer, until equilibrium is attained between the vapor and liquid phases.

In the static method samples of the vapor and liquid phases are withdrawn and analysed, where in the total-pressure method only the liquid phase is analysed.

The selection of an equilibrium still for the mixture being investigated depends on the physical properties of the system, the quantities available, the operating conditions, the time available, and to some extent the method of analysis used. When the equilibrium still has been selected considerable care and experience is required in its operation.

CHAPTER III

EXPERIMENTAL STUDY OF VAPOR - LIQUID EQUILIBRIA

Experimental studies of phase equilibria of hydrocarbons and air component mixtures are of great help in the design of separation equipment. Although vapor-liquid equilibrium data are available for many binary combinations, for the binary systems argon-ethane and ethylene-propane and the ternary system argon-methane-ethane, there are no equilibrium data available in the literature. Therefore, it was decided to measure equilibrium data for the above mentioned binary and ternary systems and to remeasure the binary systems nitrogen-argon and argon-methane for the purpose of testing the new equipment.

3.1 FORCED CIRCULATION APPARATUS

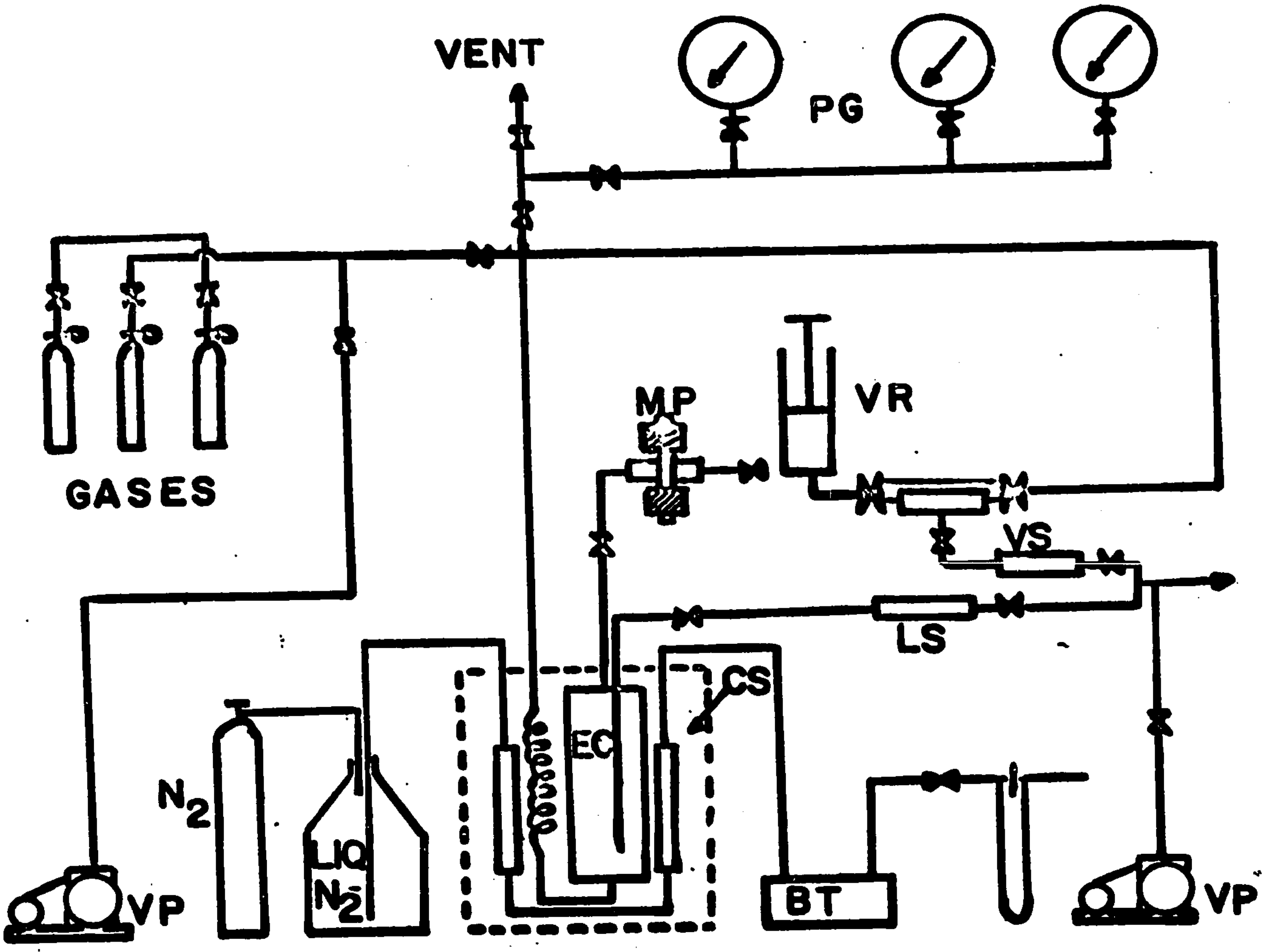
3.1.1. APPARATUS

A forced circulation apparatus was used for part of this work. Design details are available elsewhere(46, 56) and only a brief description is given here. A schematic diagram is shown in Figure (3-1).

The apparatus assembly consists of a feed charging unit, a transparent equilibrium cell, a closed circulation loop, an electro-magnetic pump, a cryostat and its temperature control system, sampling facilities, and temperature and pressure measuring devices.

a) Feed Charging Unit

A group of gas cylinders, each equipped with a pressure regulator, were connected to the system. The number and type of these cylinders will depend on the system under study.



BT	BUFFER TANK.
CS	CRYOSTAT.
EC	EQUILIBRIUM CELL.
PG	PRESSURE GAUGE.
VS	VAPOR SAMPLING BULB.
LS	LIQUID SAMPLING BULB.
MP	MAGNETIC PUMP.
VP	VACUUM PUMP.
VR	VOLUME REGULATOR.

Figure 3-1 Schematic Diagram of Experimental Apparatus

b) Closed Circulation Loop

The closed circulation loop consists of the equilibrium cell, the electro-magnetic pump, a volume regulator, a vapor sampling tube, and a cooling coil. A stainless steel 100 ml. Jerguson transparent level gauge was used as the equilibrium cell. A 1/2" NPT nipple was installed at the top of the cell, through which three 1/16" diameter liquid sampling tubes were inserted at different levels. The bottom of the cell was fitted with a 1/2" NPT connector and welded to it was a 1/8" autoclave adapter which was connected to the cooling coil of the vapor return line. A cross section view of the cell is shown in Figure (3-2).

The electric circuit of the electro-magnetic pump is shown in Figure (3-3).

c) Cryostat and Temperature Control System

A Dewar flask, of 18 liter capacity filled with isopentane, was used as the cryostat. The cryostat was equipped with two variable speed stirrers, a heating element, a refrigerating coil, and a resistant type temperature sensing probe connected to a proportional-type Bayley temperature controller, Model 250. The temperature of the cryostat was regulated by controlling the evaporation rate of liquid nitrogen in the refrigerating coil.

d) Temperature and Pressure Measuring Devices

The equilibrium temperature was measured by means of two calibrated thermocouples of the protected-type, one for the liquid phase and one for the vapor phase, in conjunction with a Leeds and Northrup K-3 poteniometer, and a Tinsely SRI galvanometer. The thermocouples were calibrated against the vapor pressure of pure methane(57).

The pressure of the system was measured by means of three pretested 12-inch certified Heise-Bourdon gauges (0-60, 0-250, & 0-500 psia.).

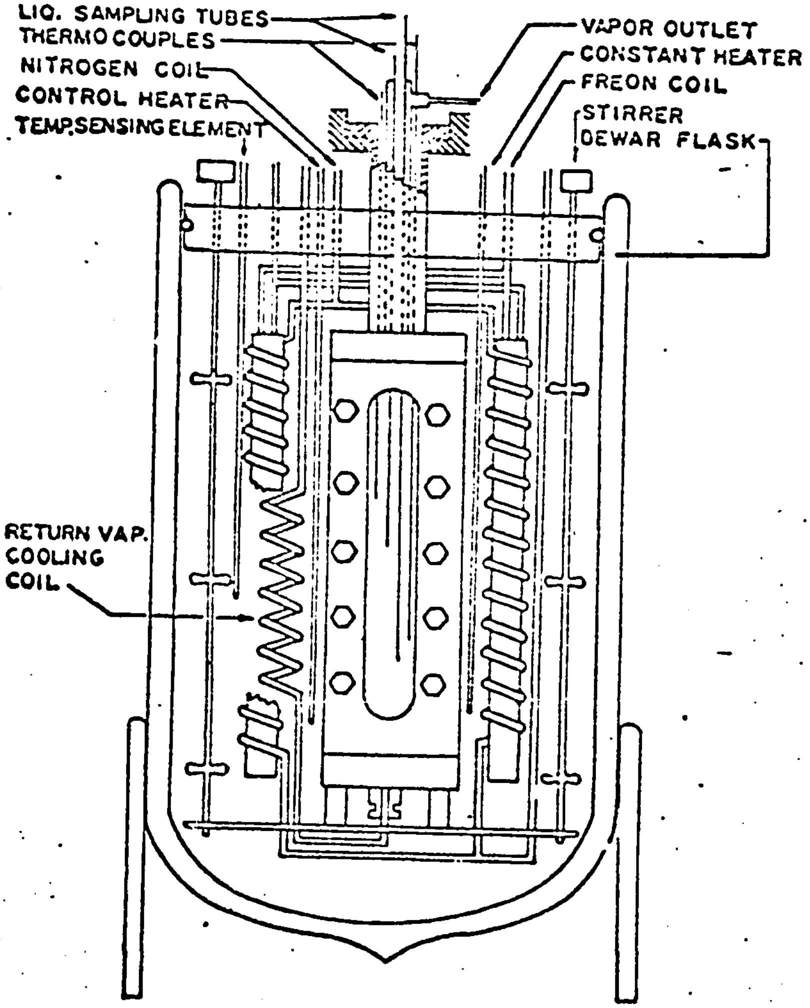


Fig. 3.2 Equilibrium cell and cryostat.

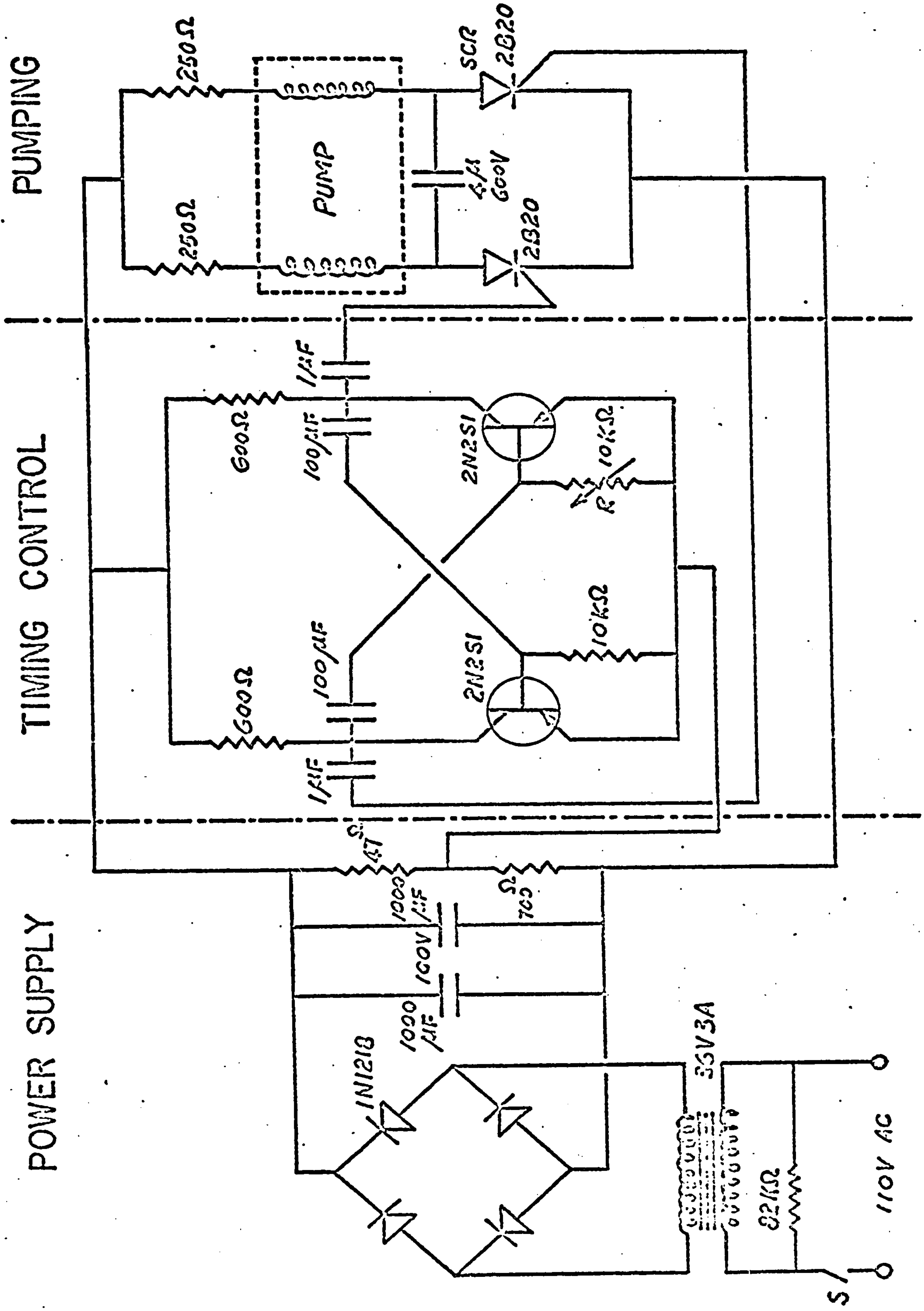


Figure 3-3 Electric Circuit of Electromagnetic Pump

e) Sampling

The liquid sample was withdrawn from a tube 1/16" in diameter. Vapor samples were trapped either in the volume regulator or in the vapor sampling tube. A schematic diagram of the sampling system is shown in Figure (3-4).

3.1.2 MATERIALS

Research grade gases, supplied by Matheson of Canada Ltd., were used without further purification. The specified minimum purities of these gases were as follows:

Argon	99.998	mole %
Methane	99.99	mole %
Ethane	99.9	mole %

3.1.3 EXPERIMENTAL PROCEDURE

The experimental procedure followed during this work can be summarized in the following steps:

1. The system was evacuated at room temperature, purged with the more volatile component of the system under study, evacuated, and maintained under vacuum.
2. The Dewar flask was filled with isopentane and the equilibrium cell was immersed into it.
3. The liquid nitrogen tank was pressurized with compressed nitrogen gas to about 10 psig., and the flow rate of the liquid nitrogen was adjusted.
4. The bath stirrers were turned on.
5. The temperature controller was turned on when the required temperature range was reached (± 10°K).

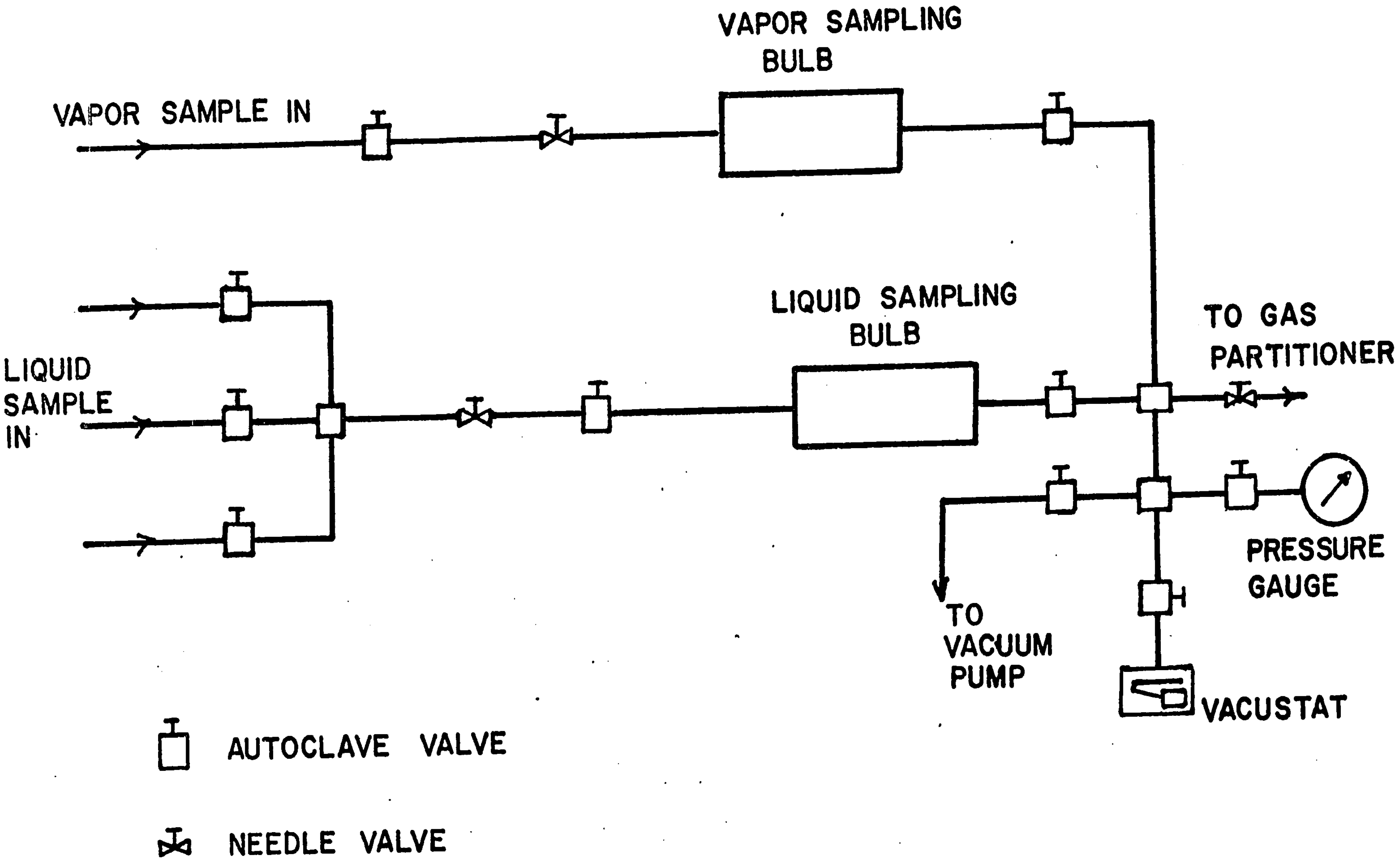


Figure 3-4 Schematic Diagram of Sampling System

6. After the desired temperature was achieved, the components of the system under study were fed into the cell, starting with the less volatile, until the liquid level had risen to about half of the equilibrium cell height.
7. The electro-magnetic pump was started and was kept on until equilibrium was attained.
8. As soon as the pump was stopped, pressure and temperature of the system was recorded; also, vapor phase sample was trapped.
9. Liquid sample was obtained by very slow vaporization of liquid phase through the sampling tube, and maintaining the system pressure by adjusting the volume regulator.
10. Analysis of the liquid and vapor phase samples followed using the gas partitioner.
11. After the first experimental point had been obtained, the above steps were repeated for different composition until the whole P - X - Y curve was obtained.

Analysis of the samples was made with a Fisher/Hamilton gas partitioner, model 29V, using silica gel (28-30 mesh) as the column (6 ft.) packing material, and helium as the carrier gas. The temperature of the gas partitioner cell was maintained at 70°C, and the flow rate of helium was maintained at 100 ml. /min. at 35 psig. A retention time of 6 min. was required for three component mixture.

The gas partitioner was calibrated using known composition samples. The result of the calibration was plotted as composition ratios versus peak height ratios of first component to the second component.

3.1.4 MEASUREMENT ERRORS

Measurement errors may be attributed to:

a) Temperature measurement error

The temperature of the cryostat was controllable to ± 0.02°C and the temperatures measured were probably accurate to about ± 0.01°C.

b) Pressure measurement error

The accuracy of the pressure gauges used was ± 0.1% of the full scale.

c) Composition analysis error

The analysis technique used in this work yielded analysis of binary mixtures reproducible to about 0.003-0.004 mole fraction. The average deviation obtained in the calibration was between 0.001-0.002 mole fraction.

3.1.5 EXPERIMENTAL RESULTS

Vapor-liquid equilibrium data for the binary system argon-ethane and the ternary system argon-methane-ethane were determined at 115.5°K as part of a continuing programme to investigate the non-ideal behaviour of mixtures of simple molecules. Phase equilibria of these systems are important for design purposes and for testing solution theories. Equilibrium data for the binary system argon-ethane was previously reported by Eckert and Prausnitz(58). However, there were only six experimental points, each measured at a different temperature. In this investigation, 38 experimental points were determined for the binary system, and 23 experimental points for the ternary system.

The experimental equilibrium temperature, pressure, and liquid composition determined in this investigation for the binary system argon-ethane are listed in Table 3.1, and shown in Figure (3-5). The concentration of ethane in the vapor phase was found to be extremely low, and

TABLE 3-1

Experimental Results for the Binary System Argon-Ethane at 115.5°K

P_{psia}	x_A
24.6	0.0690
25.2	0.0710
25.2	0.0712
29.7	0.0926
29.7	0.0928
30.8	0.0980
33.8	0.1119
34.1	0.1135
36.0	0.1222
41.2	0.1450
43.4	0.1546
48.4	0.1760
51.0	0.1873
57.8	0.2176
63.9	0.2470
70.7	0.2835
80.1	0.3422
81.8	0.3540
87.2	0.3946
87.4	0.3969
90.9	0.4259
94.3	0.4577
98.6	0.5046
100.9	0.5355
105.5	0.6120
108.1	0.6638
109.8	0.6978
110.8	0.7304
113.0	0.7581
115.2	0.8004
116.2	0.8210
116.3	0.8230
118.5	0.8619
123.1	0.9151
124.5	0.9265
129.1	0.9570
130.5	0.9654
133.1	0.9813

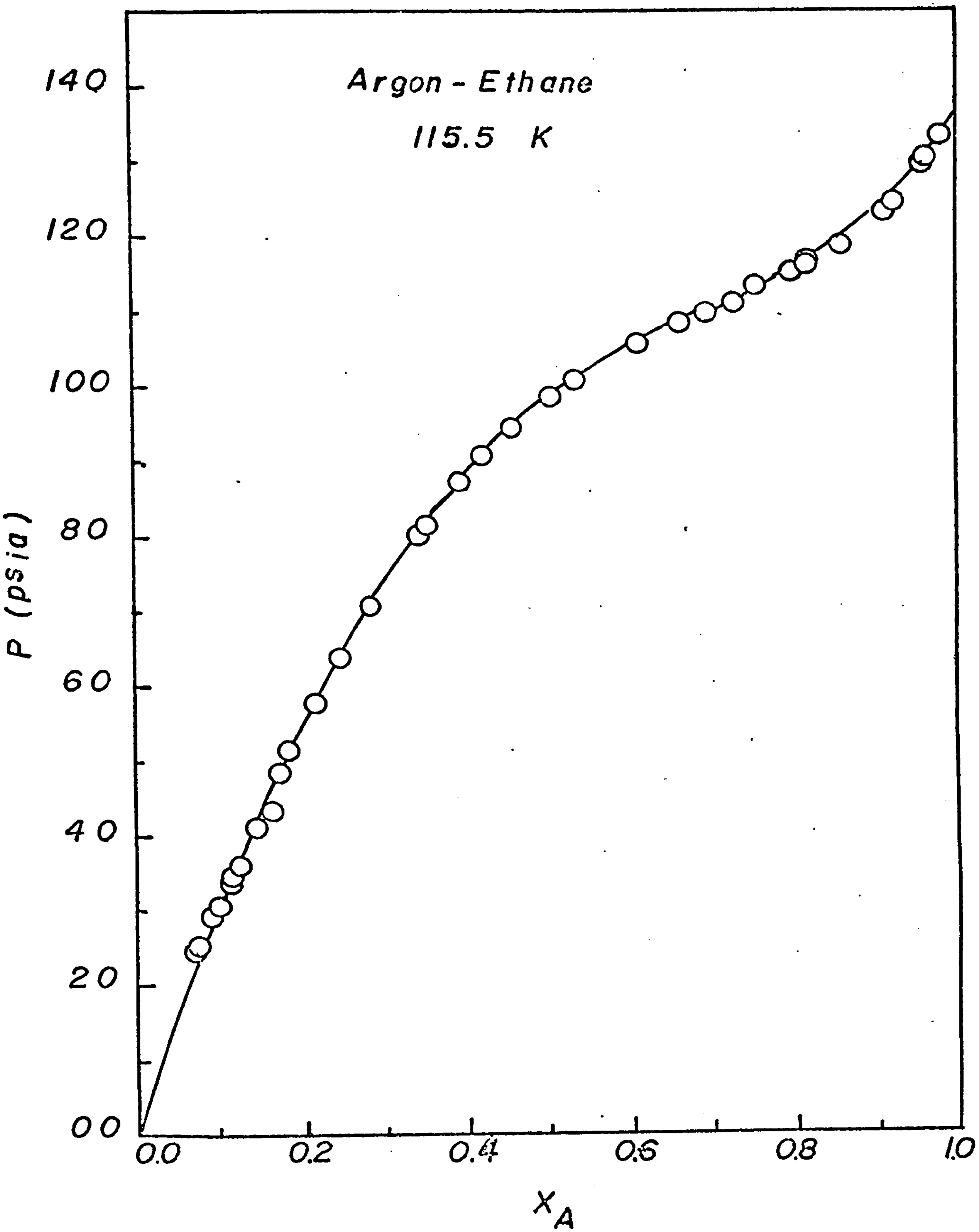


Figure 3-5 Total Pressure-Liquid Composition Diagram for the System Argon-Ethane at 115. 5°K

TABLE 3-2

Experimental Results for the Ternary System Argon-Methane-Ethane at 115.5°K

P_{sia}	x_A	x_{C_1}	y_A	y_{C_1}
59.8	0.2572	0.1859	0.9164	0.0836
71.1	0.3241	0.1743	0.9332	0.0668
80.0	0.4177	0.1521	0.9525	0.0475
89.5	0.5148	0.1271	0.9646	0.0354
94.4	0.5889	0.1079	0.9761	0.0279
98.5	0.6553	0.0980	0.9762	0.0238
51.0	0.1977	0.3081	0.8400	0.1600
64.6	0.3046	0.3066	0.8852	0.1148
70.7	0.3547	0.2737	0.9056	0.0944
79.1	0.4521	0.2351	0.9300	0.0700
84.4	0.4925	0.2187	0.9377	0.0623
90.4	0.5571	0.1887	0.9489	0.0511
99.5	0.6625	0.1481	0.9617	0.0383
107.5	0.7564	0.1071	0.9730	0.0270
51.8	0.2810	0.5164	0.8018	0.1982
59.8	0.3398	0.4746	0.8359	0.1641
69.5	0.4198	0.4143	0.8715	0.1285
79.5	0.5083	0.3551	0.8984	0.1016
89.2	0.6135	0.2803	0.9242	0.0758
98.0	0.6924	0.2044	0.9469	0.0531
78.3	0.4431	0.2317	0.9309	0.0691
81.0	0.4606	0.2096	0.9380	0.0620
84.0	0.4854	0.1675	0.4510	0.0490

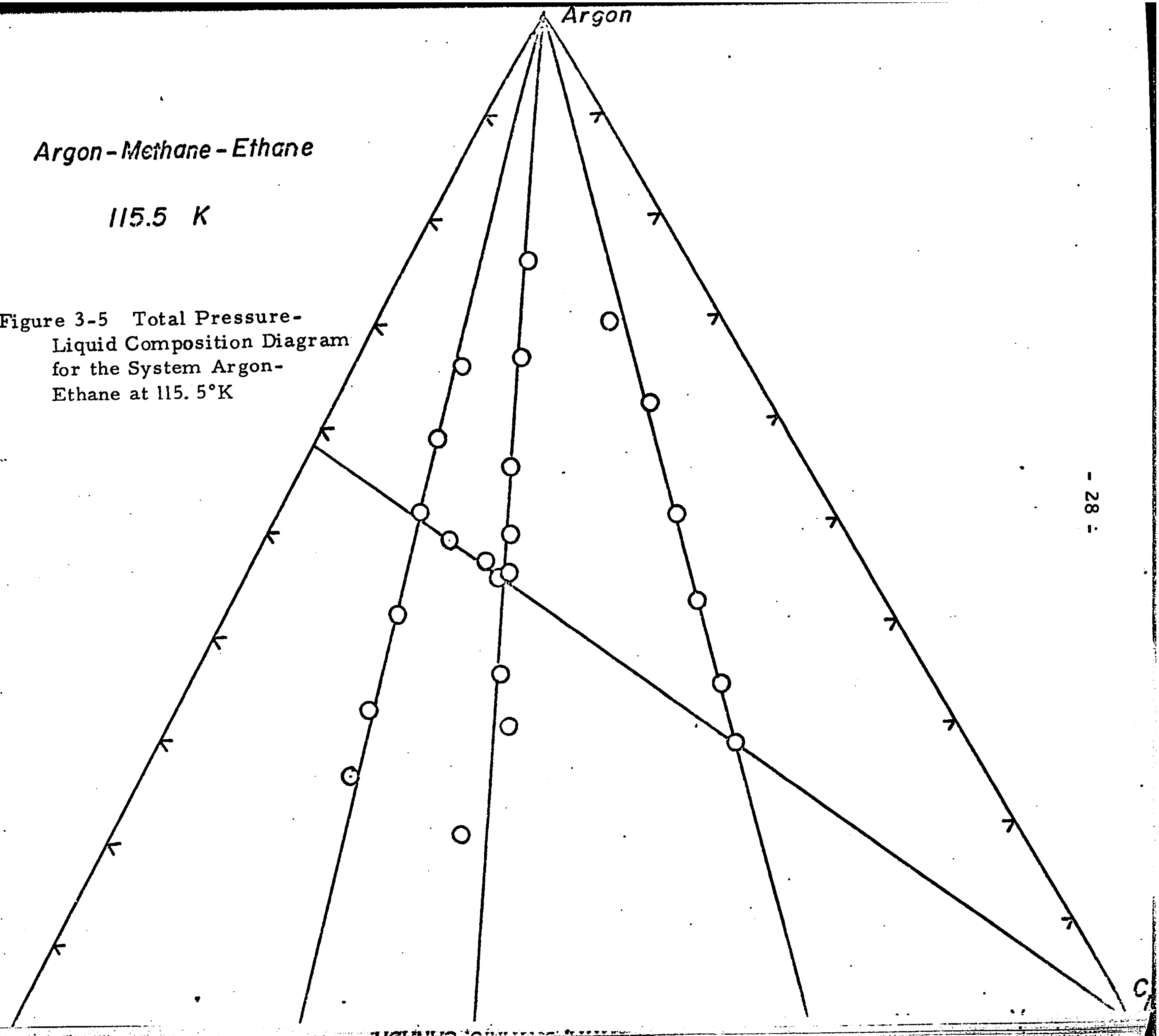


Figure 3-5 Total Pressure-Liquid Composition Diagram for the System Argon-Ethane at 115.5°K

the error in its determination was rather high. For this reason, it was decided to evaluate the ethane composition of the vapor from the total vapor pressure-liquid composition measurements. At 115.5°K the system pressure was relatively low, and the total pressure method previously proposed(1) was employed for this purpose.

The experimental points for the ternary system argon-methane-ethane were determined along constant x_{C_1}/x_{C_2} and constant x_A/x_{C_2} secants. This arrangement is shown in Figure (3-6) and listed in Table (3-2) and was planned for the purpose of reducing the ternary system to a hypothetical binary so that the total pressure method proposed for binary system(1) could be extended and applied.

3.2 TOTAL PRESSURE APPARATUS

3.2.1 APPARATUS

The apparatus consists mainly of an equilibrium cell, an electromagnetic stirrer, a cryostat and its temperature and pressure controllers, and temperature and pressure measuring devices. A schematic diagram is shown in Figure (3-7).

a) Equilibrium Cell

The equilibrium cell was constructed by drilling out on the top of a copper bar of 24" length and 5" diameter. The dimensions of the cell were 4" length and 3/4" diameter.

Two windows were made opposite to each other on the sides of the cell. The windows were covered with high pressure gauge glass and were held tight with two brass plates and a set of 8 bolts. This arrangement permits visual observation of the cell contents. A diagram of the cell is shown in Figure (3-8).

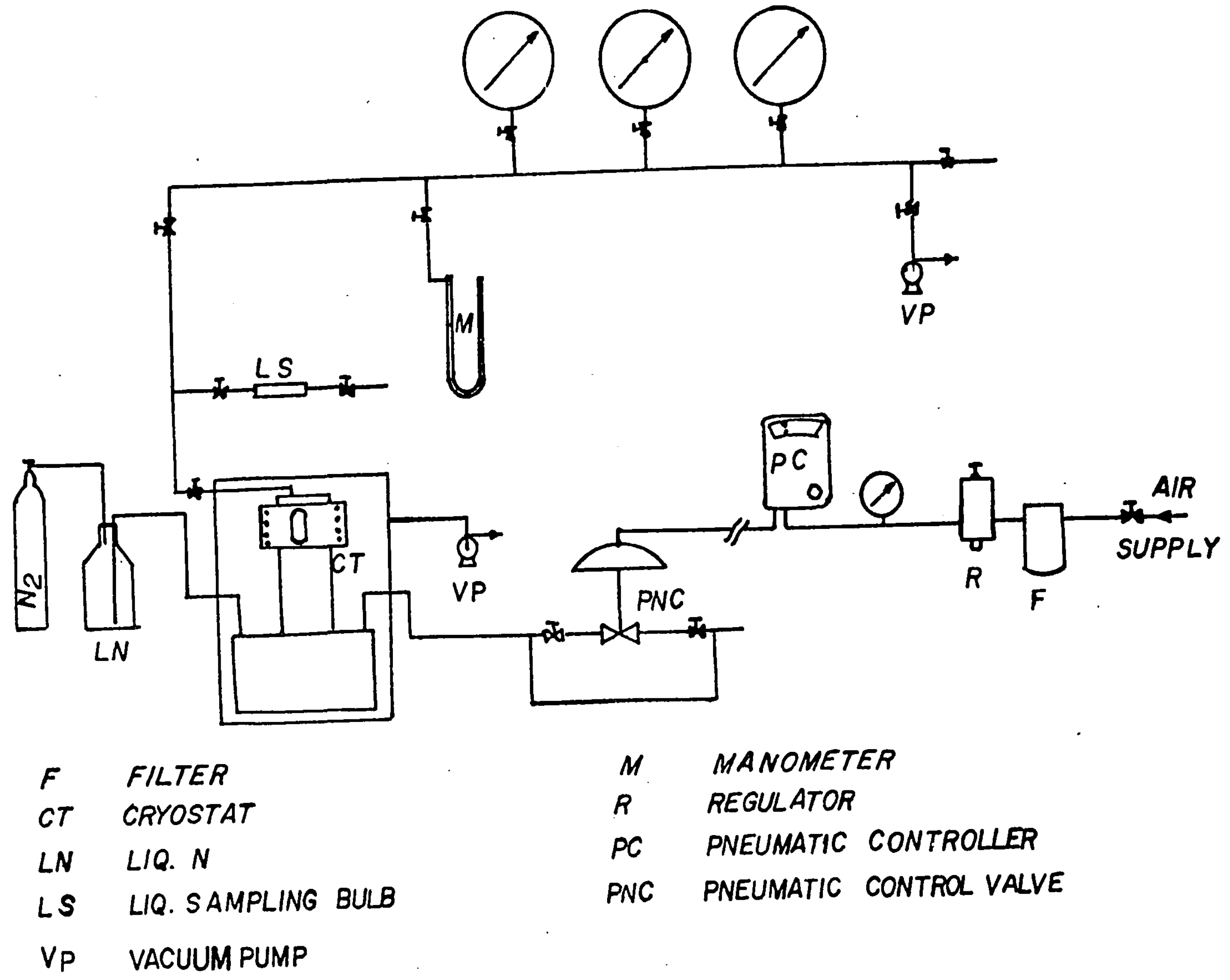
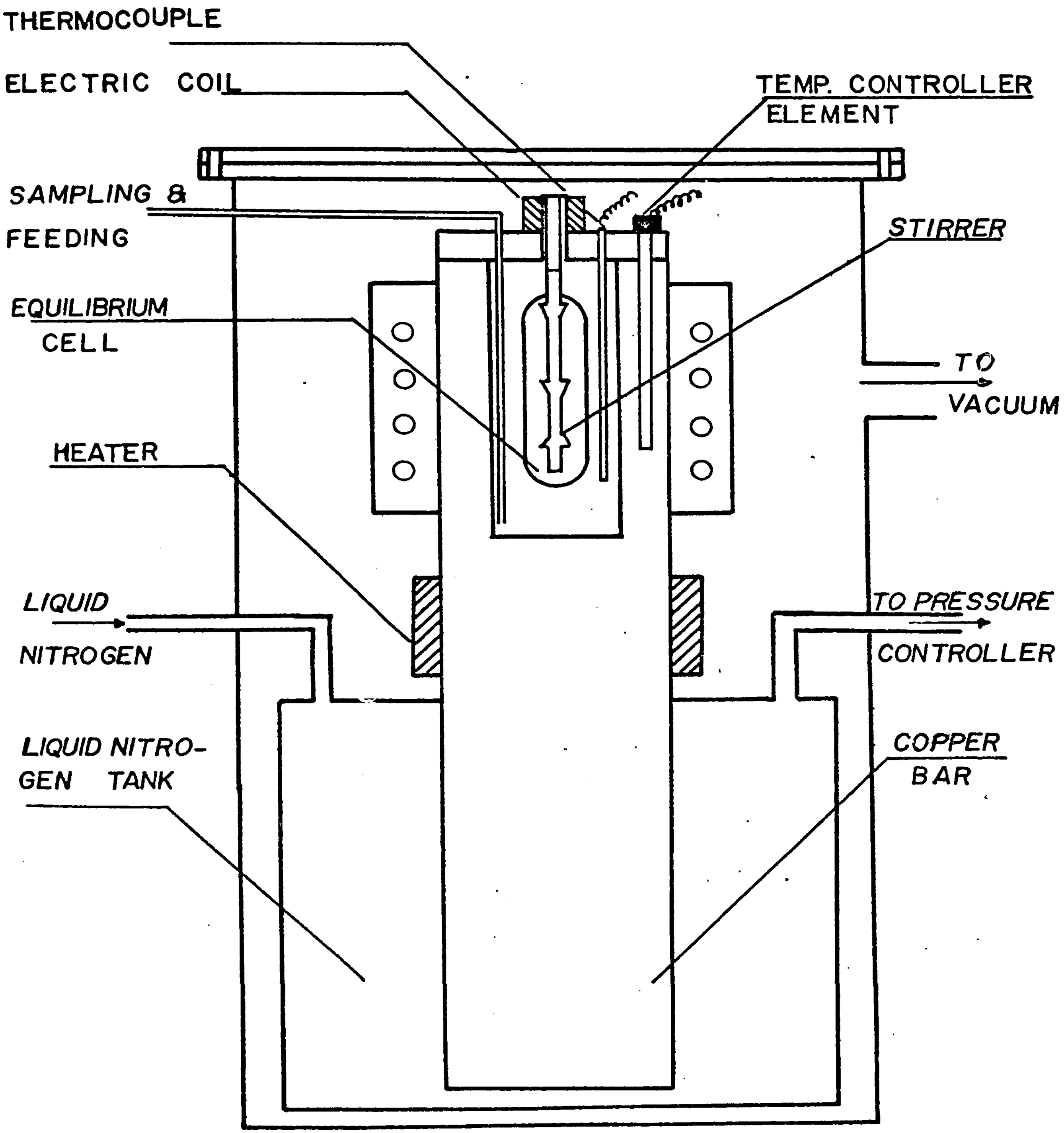


Figure 3-7 Schematic Diagram of Experimental Apparatus



TOTAL PRESSURE CRYOSTAT

Figure 3-8 Equilibrium Cell and Cryostat

Agitation of the liquid phase was performed by means of an electro-magnetic stirrer which moves upwards and downwards.

c) The Cryostat

Operation at low temperature was achieved by the use of a vacuum cryostat, Figure (3-8). The copper bar, that has the equilibrium cell at one end, was placed in a circular steel tank (15-lit.) which was filled with liquid nitrogen. The entire assembly was suspended inside a steel chamber. The temperature of the cell was controllable to ± 0.02°C using a Hallikainen controller (Thermtrol model-1109), and a heater placed at the middle portion of the copper bar. The controller element (model 1358-AS3, for solids) was placed in a hole beside the cell, which was filled with copper powder.

The vapor pressure of the liquid nitrogen was controlled by means of a pneumatic pressure controller supplied by Foxboro Co. of Canada.

Thermal insulation was achieved by evacuating the steel chamber to a pressure about 10^{-4} mm-Hg, and the outside parts of the cryostat were polished and painted with aluminum paint to minimize radiation.

Visual observation of the cell was made through two opposing windows in the outer shell with the corresponding windows of the equilibrium cell. A light bulb was placed near one window and the visibility through the other was excellent.

The cryostat was designed for low temperature operation, from the triple point of liquid nitrogen to 120°K, using the temperature controller and the pressure pneumatic controller (closed mode). This temperature range was extended to over 120°K using the temperature controller and controlling the flow rate of liquid nitrogen (open mode).

d) Temperature and Pressure Measurements

Equilibrium temperature was measured by means of two calibrated thermocouples one for the liquid and one for the vapor phase, in conjunction with a Leeds and Northrup K-3 potentiometer and a Tinsely SRI galvanometer.

The system pressure was measured by a pretested 12" Heise gauge (0-500 psia). The accuracy of this gauge is 0.1% of the full scale.

e) Feed Preparation

The mixture under study was prepared in advance in a separate gas tank. The less volatile component was introduced first. The gas tank was left overnight to insure good mixing and then analysed to determine its composition.

3.2.2 EXPERIMENTAL PROCEDURE

This apparatus was used to determine vapor-liquid equilibrium data for the system ethylene-propane. Before this study, a series of test runs were performed on the binary systems nitrogen-argon at 100°K and argon-methane at 115.22°K.

The experimental procedure may be summarized as follows:

1. The cryostat was purged with helium and evacuated.
2. The equilibrium cell was purged with the more volatile component of the system under study, and then evacuated.
3. The liquid nitrogen tank was connected to the cryostat which was operated either in the closed or in the open mode depending on the temperature of interest.
4. The heater was turned on until the required temperature range was reached, then the temperature controller was turned on to achieve the desired temperature exactly.
5. The premixed gas mixture was introduced into the equilibrium cell until the liquid level had risen to more than 3/4 of the cell height.

TABLE 3-3

Comparison Between the Premixed Gas Composition and the Condensed Phase Composition for the System Ethylene-Propane

Pressure atm.	Premixed gas composition mol. fr. (ethylene)	Liquid phase composition mol. fr. (ethylene)	Difference	y_{expt} mol. fr.	y (Chapter IV) mol. fr.
		Temp. = 273.06°K			
15.24	0.3402	0.3382	0.002	0.7325	0.7316
20.13	0.4915	0.4905	0.001	0.8220	0.8175
32.15	0.8238	0.8208	0.003	0.9371	0.9350
		Temp. = 255.38°K			
11.27	0.4000	0.3970	0.003	0.8221	0.8169
12.67	0.4630	0.4620	0.001	0.8500	0.8466
17.09	0.6560	0.6530	0.003	0.9138	0.9127
		Temp. = 227.9°K			
4.0	0.2755	0.2710	0.005	0.8259	0.8226
9.15	0.7485	0.7455	0.003	0.9649	0.9636
		Temp. = 199.83°K			
2.56	0.5220	0.5170	0.005	0.9592	0.9567
3.66	0.7999	0.7959	0.004	0.9848	0.9851

6. The electro magnetic stirrer was started and kept on until equilibrium was attained (5-6 hrs.).
7. After the stirrer was stopped, the pressure and the temperature of the system were recorded.
8. Analysis of phases followed, using the gas partitioner (for certain runs). This step was not necessary, but was performed to prove the following assumptions:

    i) If the liquid phase has much larger volume than the vapor phase, it can be considered to be of the same initial composition from which it was condensed i. e. analysis of evaporated sample of the liquid phase is then avoided. This assumption was found to be valid by Din(55) and by the results reported in Table (3-3).

    ii) Vapor phase composition can be evaluated from the method proposed in Chapter IV.

3. 2. 3 ANALYSIS

Analysis was done on a Fisher/Hamilton gas partitioner, model 29V, equipped with a 3 ft. DEHS (di-2 ethyl-hexylsebacate) and 6 ft. silica gel column supplied by Fisher.

Calibration procedure was the same as that reported in section 3. 1. 3.

3. 2. 4 MATERIALS

Research grade gases supplied by Matheson of Canada Ltd. , were used without further purification. Minimum purities are as follows:

Ethylene	99. 9	mole %
Propane	99. 99 +	mole %

3.2.5 EXPERIMENTAL RESULTS

Equilibrium T-P-x measurements were made for the following systems:

1. Argon-methane system at 115.22°K
2. Nitrogen-argon system at 100.0°K
3. Ethylene-propane system at 273.06, 255.38, 227.9, and 199.83°K

The results of these measurements are listed in Table (3-4) and shown in Figure (3-9) - (3-14).

The experimental T-P-x data for the system argon-methane and nitrogen-argon were compared with those of Gravelle(59), and Narinskii(60). Results of the comparison are shown in Figure (3-13) (2-14) and discussed in Chapter IV.

TABLE 3-4

P-x Data for the System Argon (1) - Methane (2) at 115.22°K

P, atm.	x_1
1.87	0.0551
3.58	0.2492
4.06	0.3213
5.56	0.5075
7.45	0.7780
8.76	0.9561

P-x Data for the System Nitrogen (1) - Argon (2) at 100.0°K

3.89	0.1389
5.00	0.3792
5.36	0.4613
5.65	0.5287
6.28	0.6721
6.60	0.7459
7.27	0.8999

P-x Data for the System Ethylene (1) - Propane (2) at 199.83°K

1.41	0.2505
1.64	0.3025
1.84	0.3469
2.56	0.5220
3.66	0.7999
4.13	0.9159

P-x Data for the System Ethylene (1) - Propane (2) at 227.9°K

2.99	0.1856
4.00	0.2755
5.43	0.4065
7.07	0.5577
9.15	0.7405
11.34	0.9355

TABLE (cont'd)

P - x Data for the System Ethylene (1) - Propane (2) at 255.38°K

P, atm.	x_1
3.77	0.054
8.08	0.2540
11.27	0.4000
11.82	0.4250
12.67	0.4630
12.82	0.4070
17.09	0.6565
19.37	0.7505
24.78	0.9490

P - x Data for the System Ethylene (1) - Propane (2) at 273.06°K

7.72	0.0996
10.53	0.1905
11.79	0.2308
15.24	0.3402
17.66	0.4157
20.13	0.4915
22.00	0.5466
24.53	0.6212
25.96	0.6617
29.13	0.7478
32.15	0.8238
35.4	0.8980
38.69	0.9635

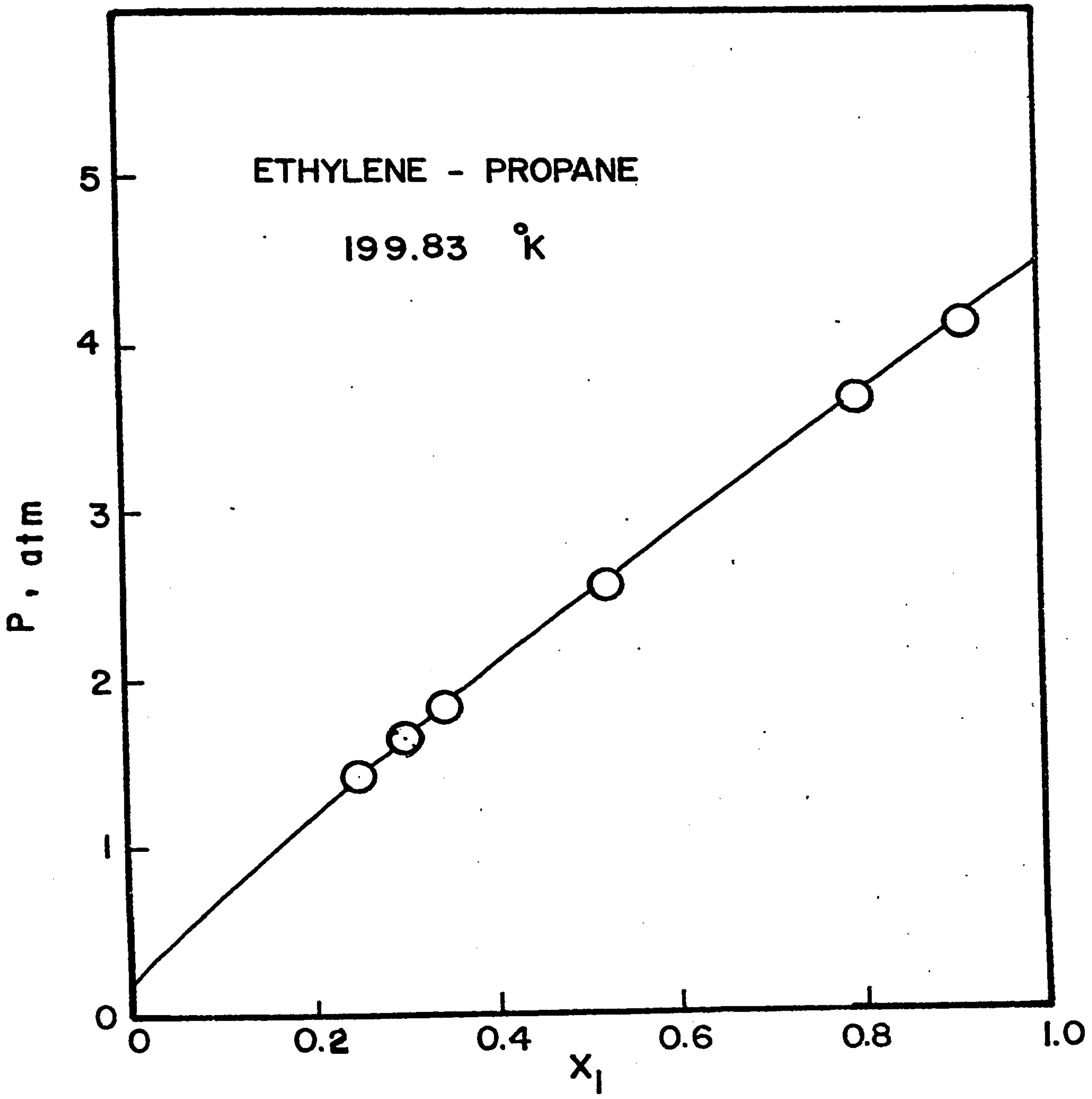


Figure 3-9 P-x Data for the System Ethylene-Propane at 199.83°K

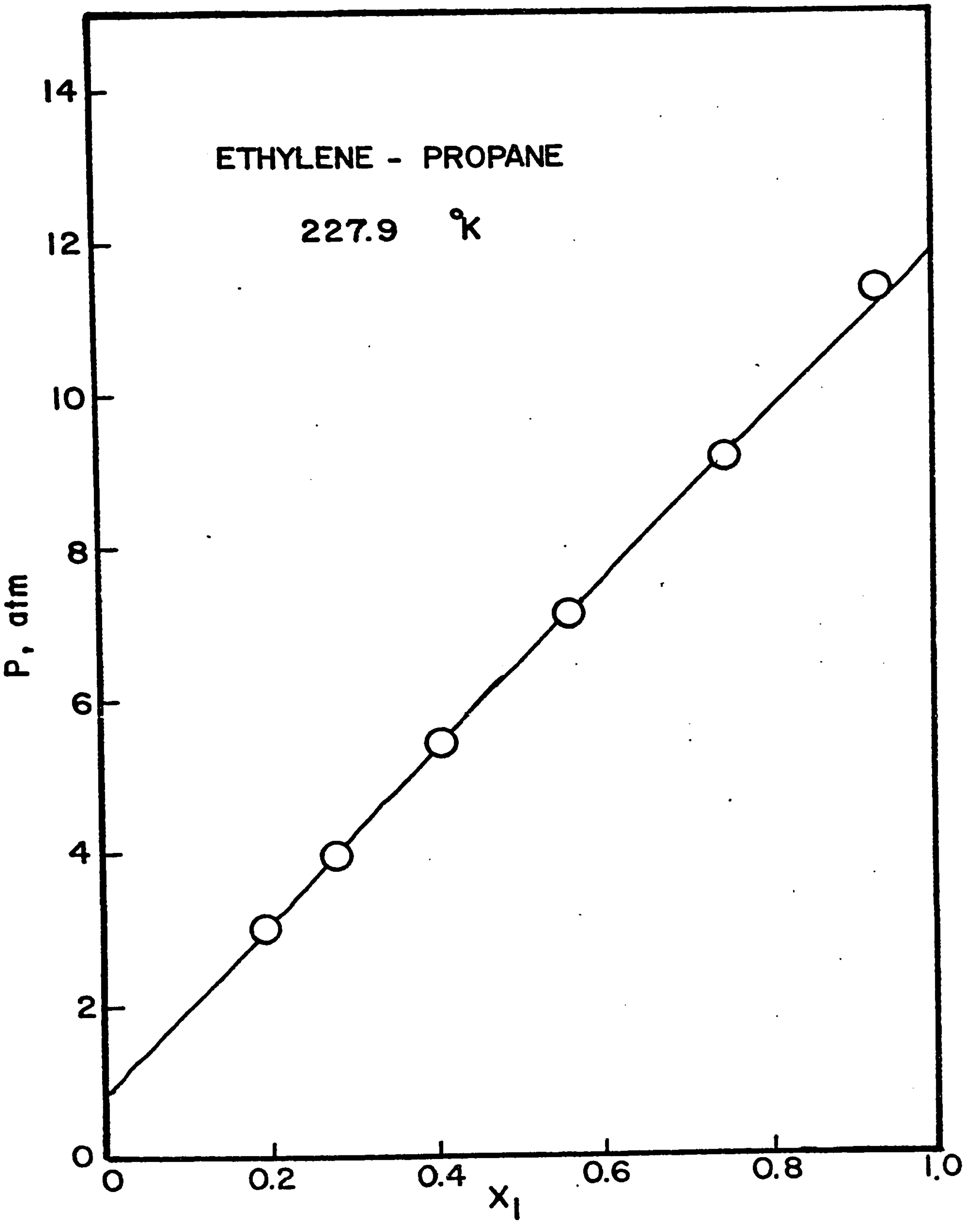


Figure 3-10 P-x Data for the System Ethylene-Propane at 227.9°K

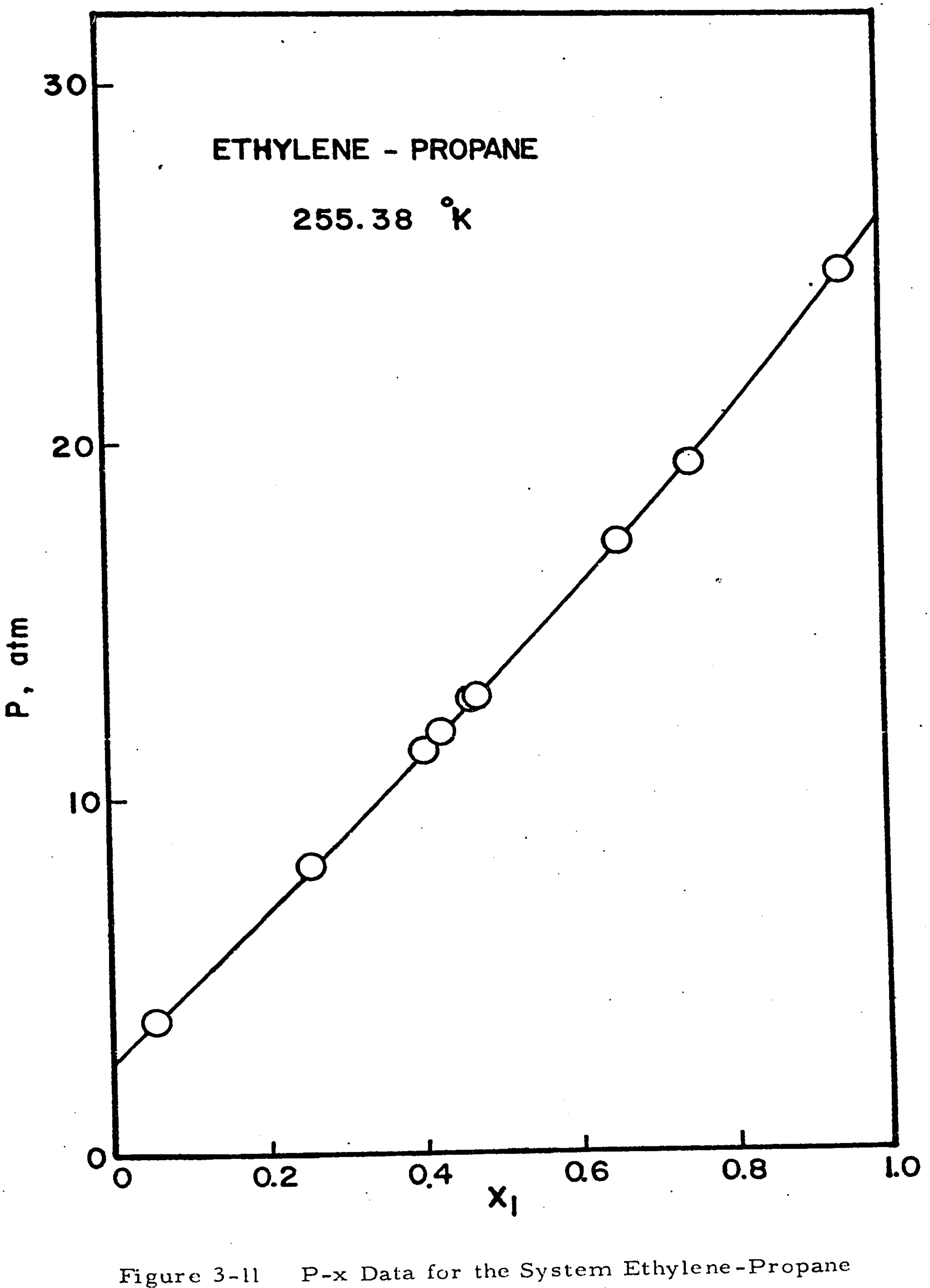


Figure 3-11 P-x Data for the System Ethylene-Propane at 255. 38°K

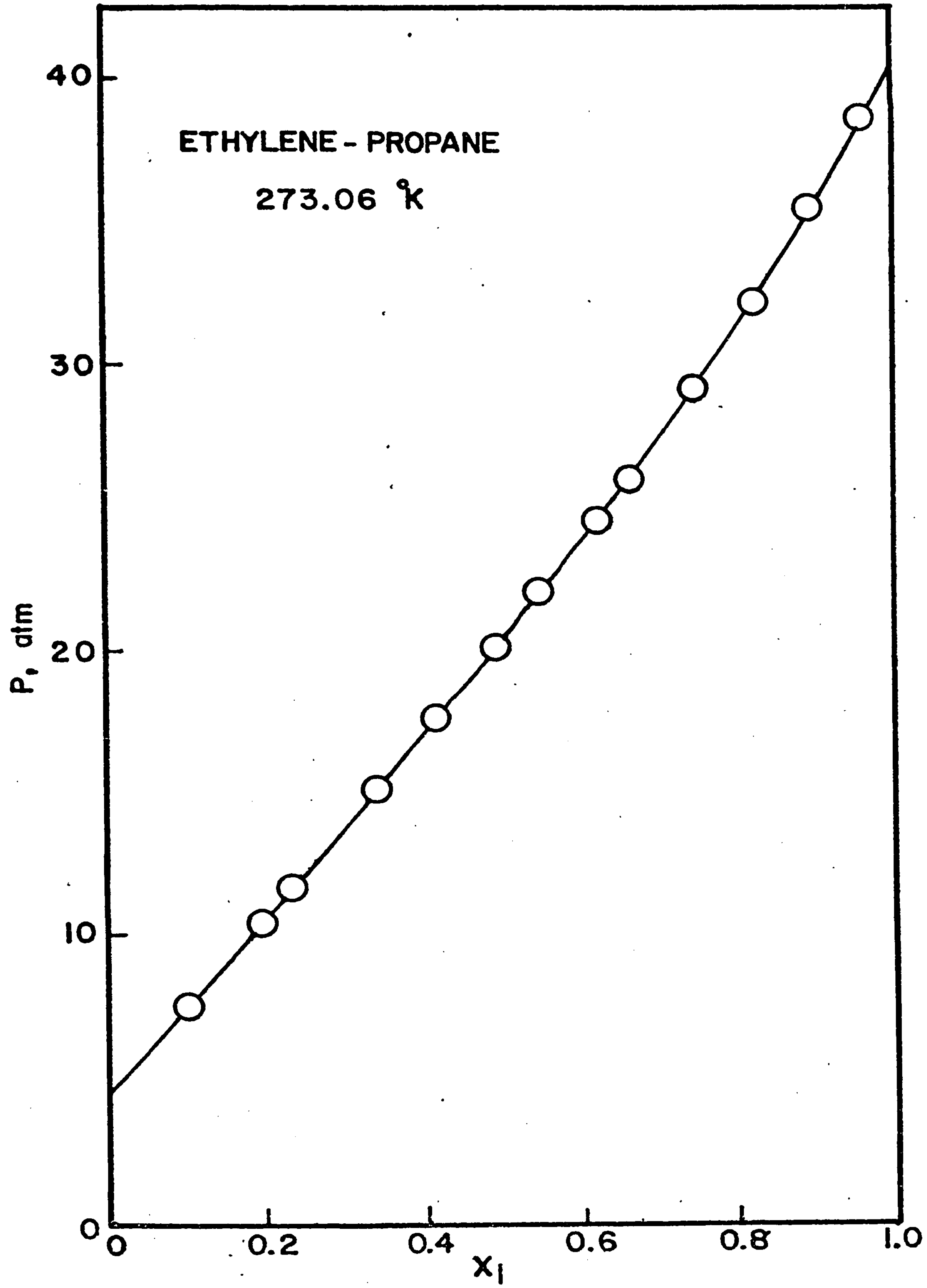


Figure 3-12 P-x Data for the System Ethylene-Propane at 273.06°K

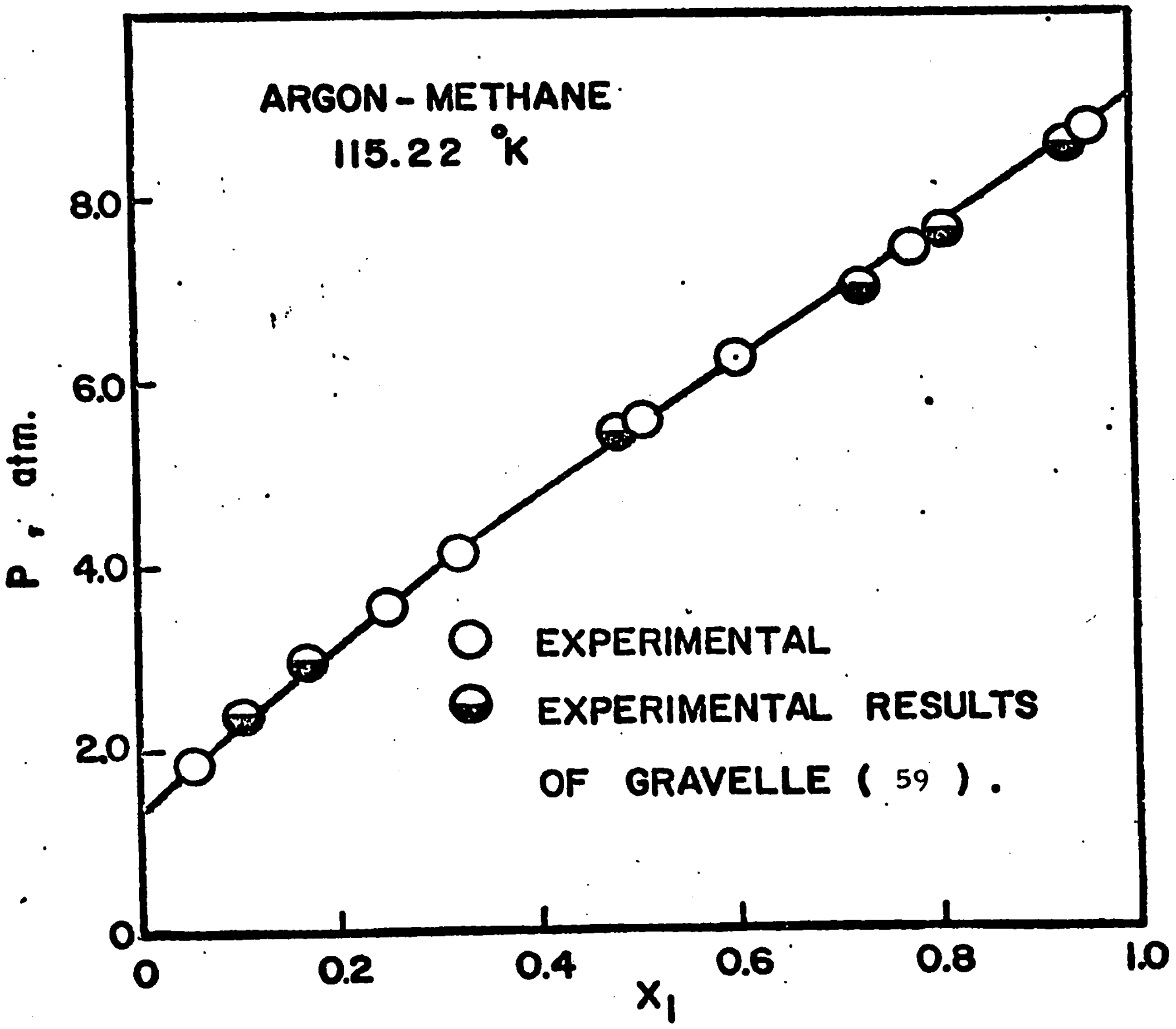


Figure 3-13 P-x Data for the System Argon-Methane at 115.22°K

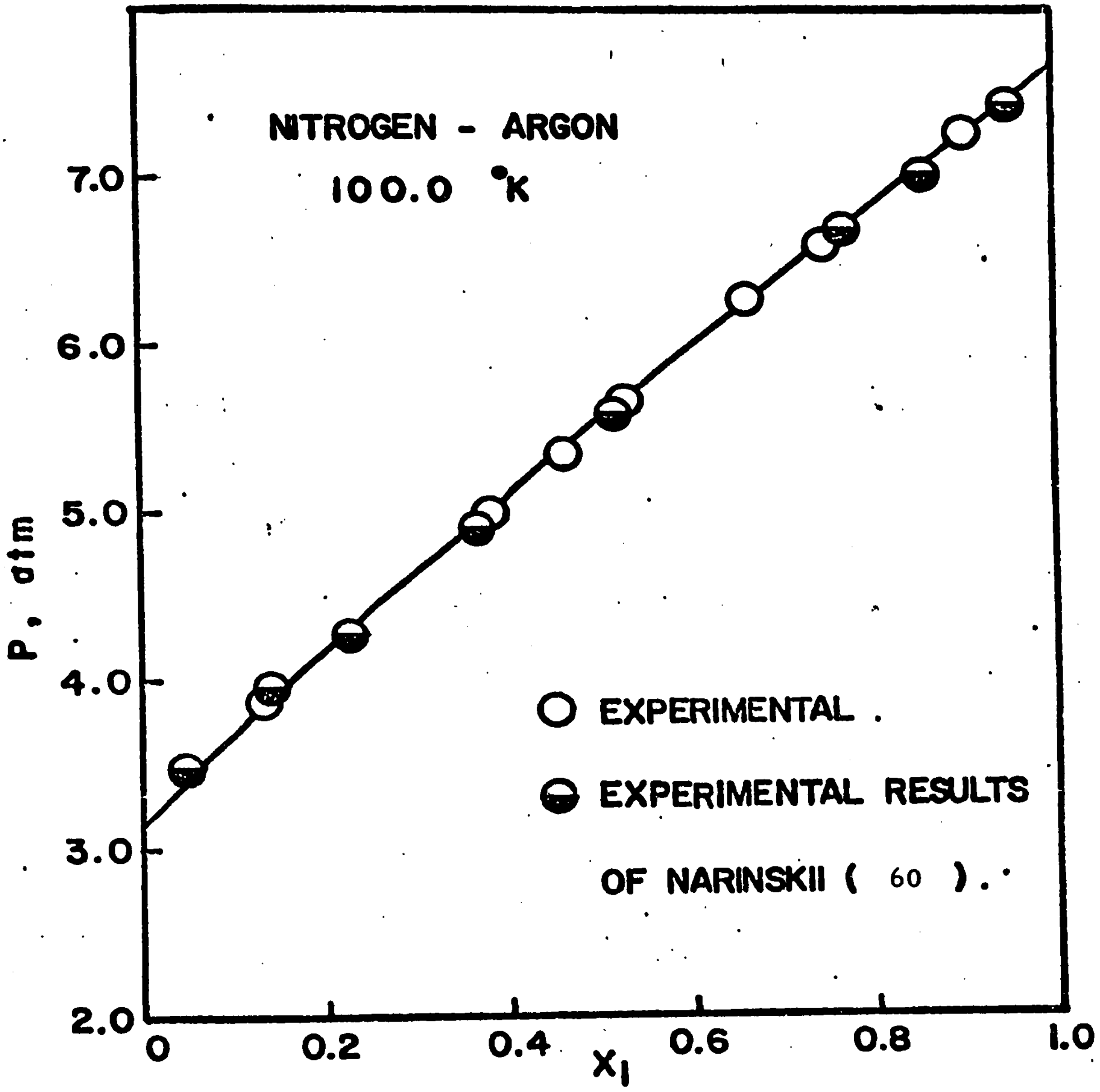


Figure 3-14 P-x Data for the System Nitrogen-Argon at 100.0°K

CHAPTER IV

CORRELATION AND PREDICTION OF PHASE EQUILIBRIA DATA USING A MODIFIED CLAUSIUS EQUATION OF STATE

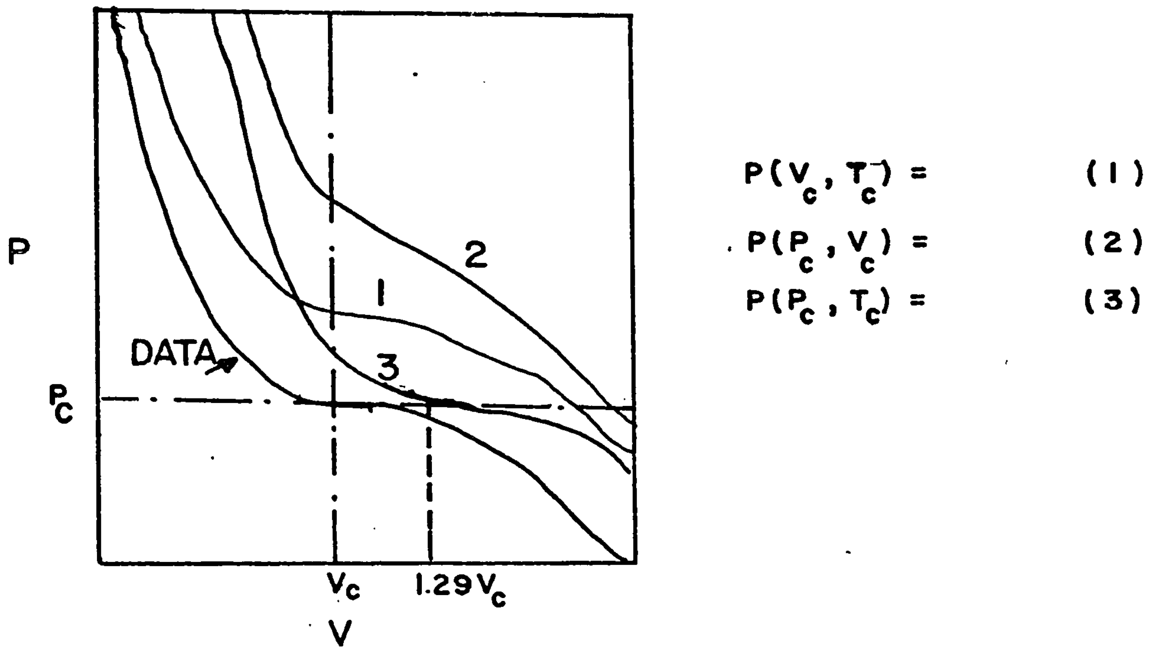
Two approaches to the development of an equation of state may be followed. One is the theoretical approach based on either kinetic theory or statistical mechanics involving intermolecular forces, for example Van der Waals equation etc... . The other approach is empirical or at best semitheoretical. This is examplified by equations such as Clausius, Redlich-Kwong etc... .

Equations of state serve essentially three purposes:

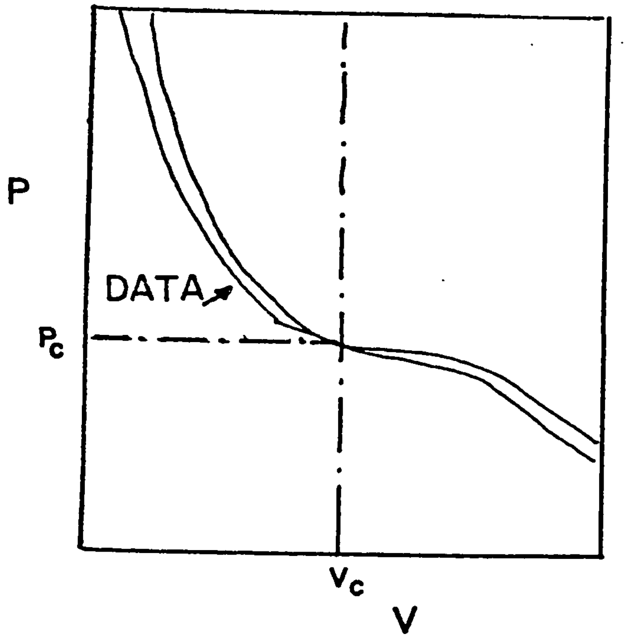
1. Representation of P-V-T data for data smoothing and interpolation, and integration of P-V-T data for calculation of derived thermodynamic properties.
2. Prediction of gas phase equilibria of pure fluids and their mixtures from a minimum experimental determination.
3. Prediction of vapor-liquid equilibrium of mixtures.

There is no single equation of state which can satisfactorily achieve all of these three purposes for a variety of fluids. Indeed there is not even one equation of state which can, with complete satisfaction, achieve just one of them over a wide ranges of conditions. There are numerous equations of state and we must remember that each has its limited usefulness; each has its strengths and its weaknesses. The choice of an equation of state must always be determined by the application for which it is intended.

Equations of state have two extremes, either an equation that has



a) TWO-CONSTANT EQUATION



b) THREE - CONSTANT EQUATION

Figure 4-1

multiconstant or an equation that has the minimum number of constants. A multiconstant-equation of state requires an extensive number of experimental data in order to evaluate the constants, also it requires an extensive number of mixing rules for the prediction of phase equilibrium in mixtures. Therefore, this type of equation of state is suitable for representing extensive experimental data over wide range of conditions i. e. its primary use is for smoothing and interpolation purposes. On the other hand a two-constant equation of state requires a minimum number of experimental data to determine its constants (for example P_c, v_c and T_c). Such an equation of state will produce inaccurate results, since its constants can be expressed in different combinations of the critical properties(32) i. e. (P_c, T_c), (P_c, v_c), and (T_c, v_c). The (P_c, v_c) and (T_c, v_c) combinations will give results which are completely far from the experimental data. On the other hand the (P_c, T_c) combination was so far the best but it is shifted towards the right as shown in Figure 4-1. a. Then, an equation of state expressed in terms of (P_c, T_c) combination has the general appearance of the true curve with the exception of a horizontal displacement in volume. Thus, a linear translation in the volume coordinate is indicated as a good possibility particularly because it has no effect on the two pressure-volume derivatives i. e.

$$\left(\frac{\partial P}{\partial v}\right)_{T=T_c} = 0 \quad , \quad \left(\frac{\partial^2 P}{\partial v^2}\right)_{T=T_c} = 0$$

This is accomplished by merely adding a constant to the volume term in equation of state. Now if this constant is set equal to $0.29\ v_c$, the calculated critical point will fall precisely on the true critical point. Unfortunately when this is done, the result is as shown in Figure 4-1.b, and it is seen that the translation is carried too far to the left. To overcome this difficulty in a two-constant equation of state some researchers tend to consider the equation parameters temperature dependent(41, 42). By doing so, one of the following important conditions must be sacrificed

$$f_i^L = f_i^V \quad \text{(at constant T)} \tag{4-1}$$

$$\Delta Z_i^L = 0 \quad \text{(at constant T)} \tag{4-2}$$

$$\Delta Z_i^V = 0 \quad \text{(at constant T)} \tag{4-3}$$

where:

f_i^L and f_i^V = fugacities of the pure liquid and vapor phase respectively.

$\Delta Z = Z_{expt.} - Z_{calc.}$

For example, Chang and Lu(41) sacrificed condition (4-3) and Joffe and Zudkevatich(42) sacrificed condition (4-1).

Therefore, a three constant equation of state is capable of satisfying conditions 4-1 to 4-3.

In 1880 Clausius(2, 30, 35, 61) proposed the following equation of state:

$$P = \frac{RT}{(v-b)} - \frac{a}{T(v+c)^2} \tag{4-4}$$

$$a = \frac{27}{64} S^2 v_c^2 P_c T_c \tag{4-5}$$

$$b = v_c (1 - S/4) \tag{4-6}$$

$$c = v_c (3 S/8 - 1) \tag{4-7}$$

$$S = R T_c / P_c V_c \tag{4-8}$$

These constants, a, b, and c, can be obtained from Equation (4-4) by equating the derivatives $(\partial P/\partial v)_T$ and $(\partial^2 P/\partial v^2)_T$, at the critical point, to zero. Clausius postulated the existence of clusters of two or more molecules which are formed by collisions. These clusters are formed at lower temperatures and break up more readily at higher ones. Clausius proposed his equation shortly after the appearance of Van der Waals equation, but his technique was not fully appreciated, possibly because he offered a kinetic explanation, rather than a mathematical explanation based on linear translation of the independent variable. It was found(30) that this equation is capable of predicting the critical isotherm up to the critical point. However, in the liquid phase region it fails to predict acceptable values.

In 1914 Wohl(3, 30, 35) suggested an equation of state which represents a modification of Van der Waals equation. The form of this equation is as follows:

$$P = \frac{RT}{(v-b)} - \frac{a}{T\,v(v-b)} + \frac{c}{T^2 v^3} \tag{4-9}$$

The constants a, b, and c are as follows:

$$a = 6\,P_c T_c v_c^2 \tag{4-10}$$

$$b = v_c/4 \tag{4-11}$$

$$c = 4\,P_c T_c^2 v_c^3 \tag{4-12}$$

Along the critical isotherm, the values predicted(30) by Equation (4-9) agree closely with the experimental values up to $P_R = 1.0$. The isotherms of this equation have a tendency to reach a peak and then decrease, never to rise again.

From the previously mentioned discussion, one may conclude that the three-constant Clausius equation of state is capable of satisfying the three conditions (4-1) - (4-3) and it possesses the ability to predict reasonably the P-V-T behavior of pure substances. In this work the Clausius equation of state was modified in order to obtain better agreement between the computed and experimental phase equilibria data. In the modification the three parameters of the equation were considered temperature dependent and may be computed from the following expressions:

$$a = \Omega_a S^2 v_c^2 P_c T_c \qquad (4\text{-}13)$$

$$b = \Omega_b v_c (4 - S) \qquad (4\text{-}14)$$

$$c = \Omega_c v_c (3S - 8) \qquad (4\text{-}15)$$

where Ω_a, Ω_b and Ω_c are the temperature-dependent parameters.

4.1 DERIVATION OF THERMODYNAMIC RELATIONSHIP

In any problem concerning the equilibrium distribution of a component i between two phases, the following condition should be satisfied

$$\mu_{iV} = \mu_{iL}$$

where μ is the chemical potential. For practical work it is convenient to introduce certain auxiliary functions such as fugacity rather than to use chemical potential directly. To predict phase equilibria data, we need to obtain an expression for the fugacity of pure components, and to evaluate the parameters of Equation (4-4) at the temperature of interest.

4.1.1 FUGACITY OF PURE SUBSTANCES

The fugacity coefficient of pure substances may be calculated by

$$R\,T\,\ln(f/P) = \int_0^P \alpha\, dP = \int_0^P \left(v - \frac{RT}{P}\right) dP$$

$$= \int_0^P V\, dP - RT \int_0^P d\ln P \qquad (4\text{-}16)$$

Differentiating Equation (4-4) with respect to volume at constant temperature results

$$dP = \left[- \frac{RT}{(v-b)^2} + \frac{2\,a}{T\,(v+c)^3} \right] dv \qquad (4\text{-}17)$$

Multiplying both sides of Equation (4-17) by v and performing integration from very low pressure to any pressure, P, then,

$$\int_0^P v\, dP = -RT \int_\infty^v \frac{v\, dv}{(v-b)^2} + \frac{2\,a}{T} \int_\infty^v \frac{v\, dv}{(v+c)^3} \qquad (4\text{-}18)$$

$$\int_0^P v\, dP = -RT \ln(v-b)\Big|_\infty^v - \frac{2\,a}{T(v+c)}\Big|_\infty^v + \frac{a\,c}{T\,(v+c)^2}\Big|_\infty^v \qquad (4\text{-}19)$$

By substituting the integration limits and subtracting the quantity $(RT \int_0^P d\ln P)$ from both sides of Equation (4-18). The following expression was obtained for the fugacity coefficient of a pure substance,

$$\ln\left(\frac{f}{P}\right) = \ln \varphi = \ln \frac{RT}{P(v-b)} + \frac{b}{(v-b)} - \frac{2a}{RT^2(v+c)} + \frac{ac}{RT^2(v+c)^2} \qquad (4\text{-}20)$$

4.1.2 FUGACITY OF A COMPONENT IN A MIXTURE

The fugacity of a component i in a mixture may be expressed as follows:

$$RT \ln \hat{\varphi}_i = \int_v^\infty \left[\left(\frac{\partial P}{\partial n_i}\right)_{T,V,n_j} - \frac{RT}{v} \right] dv - RT \ln Z \qquad (4\text{-}21)$$

The Clausius equation of state may be written as follows:

$$P = \frac{RT}{\tilde{v}-b} - \frac{a}{T(\tilde{v}+c)^2} \qquad (4\text{-}22)$$

In Equation (4-22), $\tilde{v}$ is the molar volume of the mixture and a, b and c are constants which depend on the composition.

In order to substitute Equation (4-22) into Equation (4-21), we must first transfer it from a molar basis to a total basis by the following definition:

$$\tilde{v} = V/n \qquad (4\text{-}23)$$

where n is the total number of moles. Equation (4-22) then becomes

$$P = \frac{nRT}{(V-nb)} - \frac{n^2 a}{T(V+nc)^2} \qquad (4\text{-}24)$$

By differentiating Equation (4-24) with respect to n_i at constant T, V, and n_j one obtains

$$\left(\frac{\partial P}{\partial n_i}\right)_{T,V,n_j} = \frac{RT}{(V-nb)} + \frac{n\,RT\,(\partial n\,b/\partial n_i)}{(V-nb)^2} - \left\{\frac{(\partial n^2 a/\partial n_i)}{T\,(V+nc)^2} - \frac{2n^2\,a\,(\partial n\,c/\partial n_i)}{(V+n\,c)^3}\right\} \tag{4-25}$$

If the parameters of the mixture are related to those of the pure components by the following relations:

$$a = \sum_i \sum_j x_i\, x_j\, a_{ij} = \frac{1}{n^2} \sum_i \sum_j n_i\, n_j\, a_{ij} \tag{4-26}$$

$$b = \sum_i x_i\, b_i = \frac{1}{n} \sum_i n_i\, b_i \tag{4-27}$$

$$c = \sum_i x_i\, c_i = \frac{1}{n} \sum_i n_i\, c_i \tag{4-28}$$

then,

$$\left(\frac{\partial n^2 a}{\partial n_i}\right) = 2 \sum_j n_j\, a_{ij} \tag{4-29}$$

$$\left(\frac{\partial n\,b}{\partial n_i}\right) = b_i \tag{4-30}$$

$$\left(\frac{\partial n\,c}{\partial n_i}\right) = c_i \tag{4-31}$$

Substituting Equations (4-29) - (4-31) into Equation (4-25), one obtains,

$$\left(\frac{\partial P}{\partial n_i}\right)_{T,V,n_j} = \frac{1}{n}\left[\frac{RT}{(\tilde{v}-b)}\left\{1+\frac{b_i}{(\tilde{v}-b)}\right\} - 2\,\frac{\Sigma\, x_j\, a_{ij} - a c_i/(\tilde{v}+c)}{T\,(\tilde{v}+c)^2}\right] \tag{4-32}$$

Substituting Equation (4-32) into Equation (4-21) and performing the integration yields the required expression for the fugacity coefficient for a component in a mixture,

$$\ln \hat{\varphi}_i = \ln\left(\frac{\tilde{v}}{\tilde{v}-b}\right) + \frac{b_i}{(\tilde{v}-b)} - \frac{2\sum_j x_j a_{ij}}{RT^2(\tilde{v}+c)} + \frac{a\,c_i}{RT^2(\tilde{v}+c)^2} - \ln Z \qquad (4\text{-}33)$$

4.1.3 EVALUATION OF THE PARTIAL MOLAL VOLUME IN A MIXTURE

The partial molal volume of a component in a mixture may be computed by the following relation:

$$\bar{V}_i = - \frac{(\partial P/\partial n_i)_{P,V,n_j}}{(\partial P/\partial V)_{T,n_i}} \qquad (4\text{-}34)$$

Differentiating Equation (4-24) with respect to V at constant T and n_i gives,

$$\left(\frac{\partial P}{\partial V}\right) = \frac{1}{n}\left[-\frac{RT}{(\tilde{v}-b)^2} + \frac{2a}{T(\tilde{v}+c)^3}\right] \qquad (4\text{-}35)$$

Substituting Equation (4-25) and (4-35) into Equation (4-34) yields the required equation for the partial molal volume for a component in a mixture,

$$\bar{V}_i = \frac{\dfrac{RT}{(\tilde{v}-b)}\left(1 + \dfrac{b_i}{(\tilde{v}-b)}\right) - 2\,\dfrac{\sum_j x_j a_{ij} - a\,c_i/(\tilde{v}+c)}{T(\tilde{v}+c)^2}}{\dfrac{RT}{(\tilde{v}-b)^2} - \dfrac{2a}{(T(\tilde{v}+c)}} \qquad (4\text{-}36)$$

4.2 EVALUATION OF THE CLAUSIUS EQUATION PARAMETERS

The primary information required for the evaluation of the temperature dependent parameters are the saturation pressure and the saturated vapor and liquid molar volumes of pure substances. For convenience Equation (4-4) was rearranged in the following form:

$$Z = \frac{1}{1-h} - \left(\frac{A}{B}\right) \frac{h}{(1+h)^2} \tag{4-37}$$

where:

$$Z = Pv/RT \tag{4-38}$$

$$A = a/R^2T^3 = \Omega_a S^2 v_c^2 T_c P_c/R^2 T^3 \tag{4-39}$$

$$B = b/RT = \Omega_b v_c (4 - S)/RT \tag{4-40}$$

$$C = c/RT = \Omega_c v_c (3S - 8)/RT \tag{4-41}$$

$$h = b/v = BP/Z \tag{4-42}$$

$$H = c/v = CP/Z \tag{4-43}$$

At equilibrium

$$f_v = f_L$$

or

$$\varphi_v = \varphi_L \tag{4-44}$$

From Equation (4-20), the fugacity coefficient for a pure substance in the pure liquid phase may be written in the following form:

$$\ell n\, \varphi_\ell = \frac{h_\ell}{(1-h_\ell)} - \ell n\, Z_\ell - \ell n\,(1-h_\ell) - \left(\frac{1}{1-h_\ell} - Z_\ell\right)(2+H_\ell) \tag{4-45}$$

Similarly, for the vapor phase,

$$\ell n\, \varphi_v = \frac{h_v}{(1 - h_v)} - \ell n\, Z_v - \ell n\,(1-h_v) - (\frac{1}{1-h_v} - Z_v)(2+H_v) \qquad (4\text{-}46)$$

By combining Equations (4-45) and (4-46) one obtains

$$\ell n\, \varphi_v\, Z_\ell = \frac{h_\ell}{1-h_\ell} + \ell n\,(1-h_\ell) + (\frac{1}{1-h_\ell} - Z_\ell)(2+H_\ell) \qquad (4\text{-}47)$$

These equations were used to evaluate the temperature dependent parameters Ω_a, Ω_b, and Ω_c. The calculation procedure contains two iteration loops, as shown in Figure (4-2) and may be summarized as follows:

1. Assume initial values of Ω_a, Ω_b and Ω_c ($\Omega_a = 0.42$, $\Omega_b = 0.25$, and $\Omega_c = 0.125$ which are the values obtained by Clausius).
2. For a given value of Z_L (experimental) and an assumed value of φ_v ($\varphi_v = 1.0$), calculate h_ℓ by means of Equation (4-47).
3. Calculate B, and A from Equation (4-42) and Equation (4-37).
4. Substitute the new values of A and B into Equation (4-37) and (4-42) to obtain values for Z_v and h_v.
5. Using the calculated values of Z_v and h_v from Equation (4-46).
6. Using the calculated values of φ_v, a new value of h_ℓ is obtained using Equation (4-47).
7. Compare the new value of h_ℓ with that of step 2. If disagreable repeat the calculation. (Specified tolerance, $\Delta h_\ell = 0.00005$).

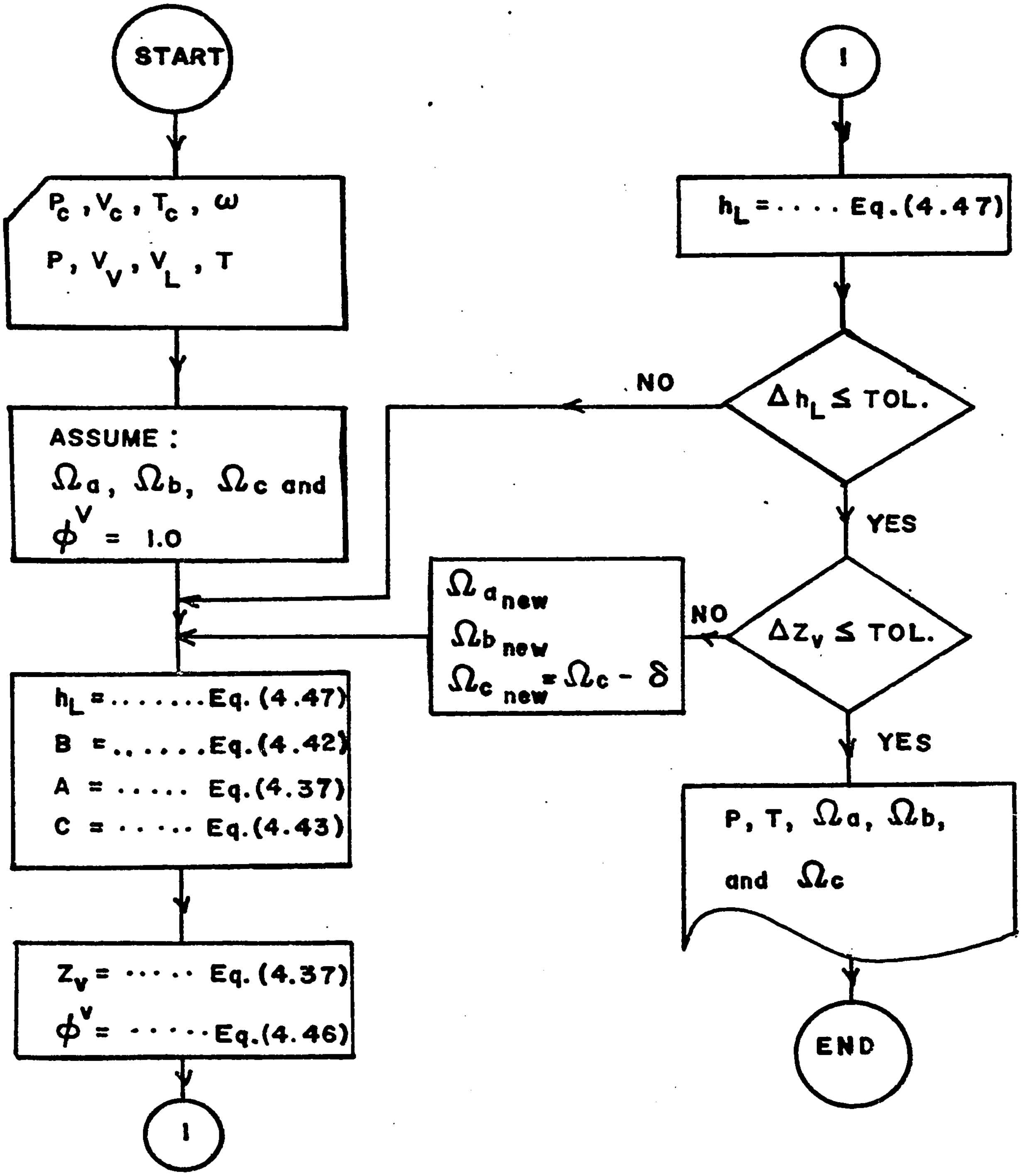


Figure 4-2 Schematic Diagram for the Evaluation of the Modified Clausius Equation Parameters

8. If the change in h_ℓ is less than the specified tolerance, the second iteration loop starts by comparing values of Z_v, from step 4, with the experimental ones.
9. If the comparisons of Z'_v are disagreable, start from the beginning with an incremented value of Ω_c, and the calculated values of Ω_a and Ω_b from the last step; step 8.
10. The second iteration loop is completed when the values of Z_v calculated agree with Z_v experimental.

4. 3 PREDICTION OF PHASE EQUILIBRIA DATA

The modified Clausius equation of state was used to predict equilibrium data using the mixing rules which were previously proposed by Chueh and Prausnitz(62).

The mixing rules were as follows:

$$a = \sum_i^N \sum_j^N x_i x_j a_{ij} \tag{4-48}$$

$$b = \sum_i^N x_i b_i \tag{4-49}$$

$$c = \sum_i^N x_i c_i \tag{4-50}$$

where:

$$a_{ij} = \Omega_{a_{ij}} (v_{c_{ij}} / Z_{c_{ij}})^2 P_{c_{ij}} T_{c_{ij}} \tag{4-51}$$

$$b_i = \Omega_{b_i} v_{c_i} (4 - 1/Z_{c_i}) \tag{4-52}$$

$$c_i = \Omega_{c_i} v_{c_i} (3/Z_{c_i} - 8) \qquad (4\text{-}53)$$

$$T_{c_{ij}} = (T_{c_{ii}} T_{c_{jj}})^{1/2} (1 - k_{ij}) \qquad (4\text{-}54)$$

$$\Omega_{a_{ij}} = 1/2 (\Omega_{a_i} + \Omega_{a_j}) \qquad (4\text{-}55)$$

$$P_{c_{ij}} = Z_{c_{ij}} R T_{c_{ij}} / v_{c_{ij}} \qquad (4\text{-}56)$$

$$V_{c_{ij}} = [1/2 (v_{c_i}^{1/3} + v_{c_j}^{1/3})]^3 \qquad (4\text{-}57)$$

$$Z_{c_{ij}} = 0.091 - 0.08\, \omega_{ij} \qquad (4\text{-}58)$$

$$\omega_{ij} = 1/2 (\omega_i + \omega_j) \qquad (4\text{-}59)$$

The k_{ij} is a constant characteristic of i-j interaction and was considered independent of temperature, pressure and composition(62). A minimum of one observation of a binary mixture property e.g. vapor-liquid equilibrium data, is required for the evaluation of k_{ij}.

For vapor-liquid equilibria of a N-component system, the following equation must be satisfied

$$\hat{f}_{iL} = \hat{f}_{iv} , \quad i = 1, 2, \ldots, N \qquad (4\text{-}60)$$

in which

$$\hat{f}_{iL} = \hat{\varphi}_{iL}\, x_i\, P \qquad (4\text{-}61)$$

$$\hat{f}_{iv} = \hat{\varphi}_{iv}\, y_i\, P \qquad (4\text{-}62)$$

Incorporating with Equations (4-48) - (4-59), and Equation (4-33), the fugacity coefficients for both phases may be expressed as follows:

$$\ln \hat{\varphi}_{iv} = \ln\left(\frac{1}{1-h_v}\right) - \ln Z_v + \left(\frac{b_i}{b}\right)\left(\frac{h_v}{1-h_v}\right) - (A/c)\left(\frac{H_v}{1+H_v}\right)$$

$$\left[\frac{2\sum_j^N x_j a_{ij}}{a} - \left(\frac{H_v}{1+H_v}\right)(c_i/c)\right] \qquad (4\text{-}63)$$

and similarly for the liquid phase

$$\ln \hat{\varphi}_{iL} = \ln\left(\frac{1}{1-h_L}\right) - \ln Z_L + \left(\frac{b_i}{b}\right)\left(\frac{h_L}{1-h_L}\right) - (A/c)\left(\frac{H_L}{1+H_L}\right)$$

$$\left[\frac{2\sum_j^N x_j a_{ij}}{a} - \left(\frac{H_L}{1+H_L}\right)(c_i/c)\right] \qquad (4\text{-}64)$$

If the temperature and (N-1) independent liquid phase mole fractions are specified, the total pressure P and (N-1) independent vapor phase mole fractions could be calculated with the aid of Equation (4-60). This is the so called bubble-point calculation. The N simultaneous equations are solved by a trial and error procedure, which is shown in Figure (4-3), and may be summarized as follows:

1. Initial guesses for P and $\hat{\varphi}_{iv}$ are made for the first iteration.
2. From the given x_i's and T, $\hat{\varphi}_{iL}$ is calculated by means of Equation (4-64).
3. Vapor phase compositions are then calculated by

$$y_i = (\hat{\varphi}_{iL}/\hat{\varphi}_{iv})\, x_i \qquad (4\text{-}65)$$

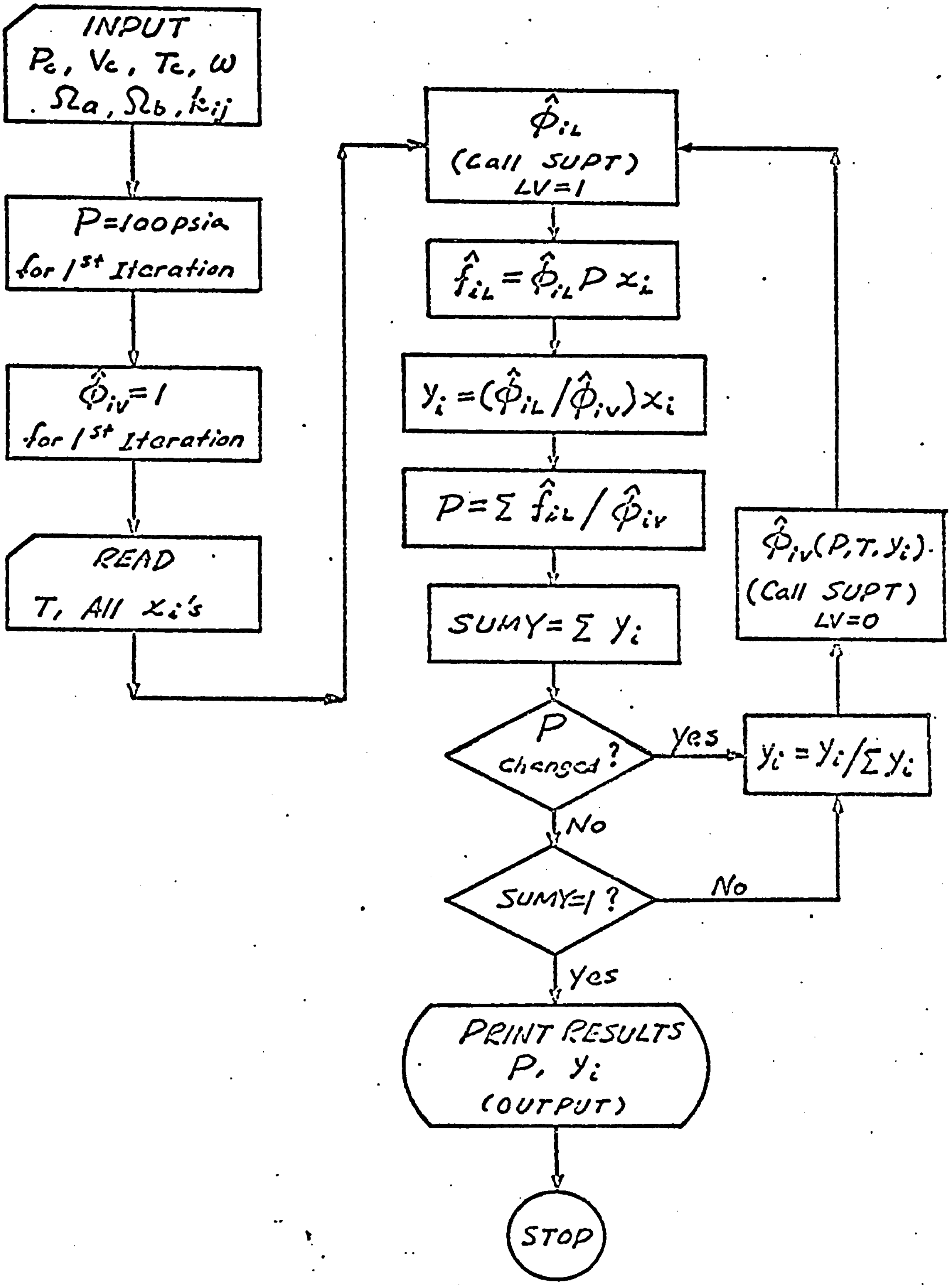


Figure 4-3 Schematic Diagram of Bubble Point Calculation

4. The total pressure, P, is calculated by

$$P = \sum_{i=1}^{N} \hat{f}_{iL}/\hat{\varphi}_{iv} \qquad (4\text{-}66)$$

5. The calculated total pressure is then compared with the assumed one, if it is different, $\hat{\varphi}_{iv}$'s are recalculated with the last obtained value of P and the normalized vapor phase composition.
6. $\hat{\varphi}_{iL}$'s are recalculated with the last obtained P.
7. As soon as the change in P satisfies the specified tolerance ($\Delta P/P = 0.0001$), the relation

$$\sum_{i}^{N} y_i = 1.0 \qquad (4\text{-}67)$$

is finally considered. Usually this is satisfied when the pressure has attained an unchanging value.

4.4 RESULTS AND DISCUSSIONS

The saturation properties (T, P, V_v, V_ℓ) available in the literature for methane(63), propane(64), n-butane(65), nitrogen(63), argon(64), oxygen(66), carbon dioxide(67) and hydrogen sulfide(67), were employed for the evaluation of the temperature dependent parameters Ω_a, Ω_b and Ω_c by means of the proposed procedure. The calculated values of these parameters are listed in Table A-I. 1.

The proposed method was used for the prediction of P-V-T data in the liquid and vapor phase region for pure oxygen, argon and nitrogen. The experimental(66) and calculated P-V-T data, for oxygen, from the proposed modification and from the modified R-K equation(41) were compared. Results of the comparison indicated that the modified Clausius equation of state is capable of predicting better results in the vapor phase

TABLE 4-1

Comparison of the Average Absolute Deviations From Various Equations of State for Argon

Equation	$\lvert \Delta Z \rvert$
Redlich-Kwong (37)	0.0133
Redlich et al. (73)	0.0086
Sugie-Lu (74)	0.0025
Modified Clausius	0.0040

region. The absolute average deviation in vapor phase compressibilities was found to be 0.0076 as shown in Table A-I. 2. In addition, the proposed method was tested for the prediction of the critical isotherm for argon and nitrogen. The average absolute deviation for argon is 0.058 and for nitrogen 0.083 as shown in Tables A-I. 3 and A-I.4. The calculated values using the proposed method, for argon as an example, were compared by those calculated by other equations of state in the range of $P_R = 0.15$ to $P_R = 0.90$. Results of comparison are listed in Table (4-1).

The proposed method was also employed for predicting system pressure, P, and equilibrium vapor phase compositions, y, from a given liquid phase compositions, x, and system temperature, T. The calculated results obtained from the proposed modification and from the modified R-K(41) equation were compared with the experimental literature values for several systems. These systems were for normal fluid mixtures and were arbitrarily chosen for the purpose of comparison.

Nitrogen-Argon

The predicted results for the nitrogen-argon system at 110.0°K were compared with the reported data of Narinskii(75). Results of comparison are listed in Tables A-I. 5 and A-I. 6 as shown in Figure (4-4) and (4-5).

Nitrogen-Oxygen

A good agreement was obtained between calculated and experimental(44) values of phase equilibrium data for the system nitrogen-oxygen at 110.05°K. Results of comparison are shown in Figure (4-6) and (4-7) and listed in Tables A-I. 7 and A-I. 8.

Nitrogen-Methane

The comparison of the calculated and experimental(76) phase equilibrium data for the system nitrogen-methane at 110.94°K is shown in Figure (4-8) and (4-9) and listed in Tables A-I. 9 and A-I. 10.

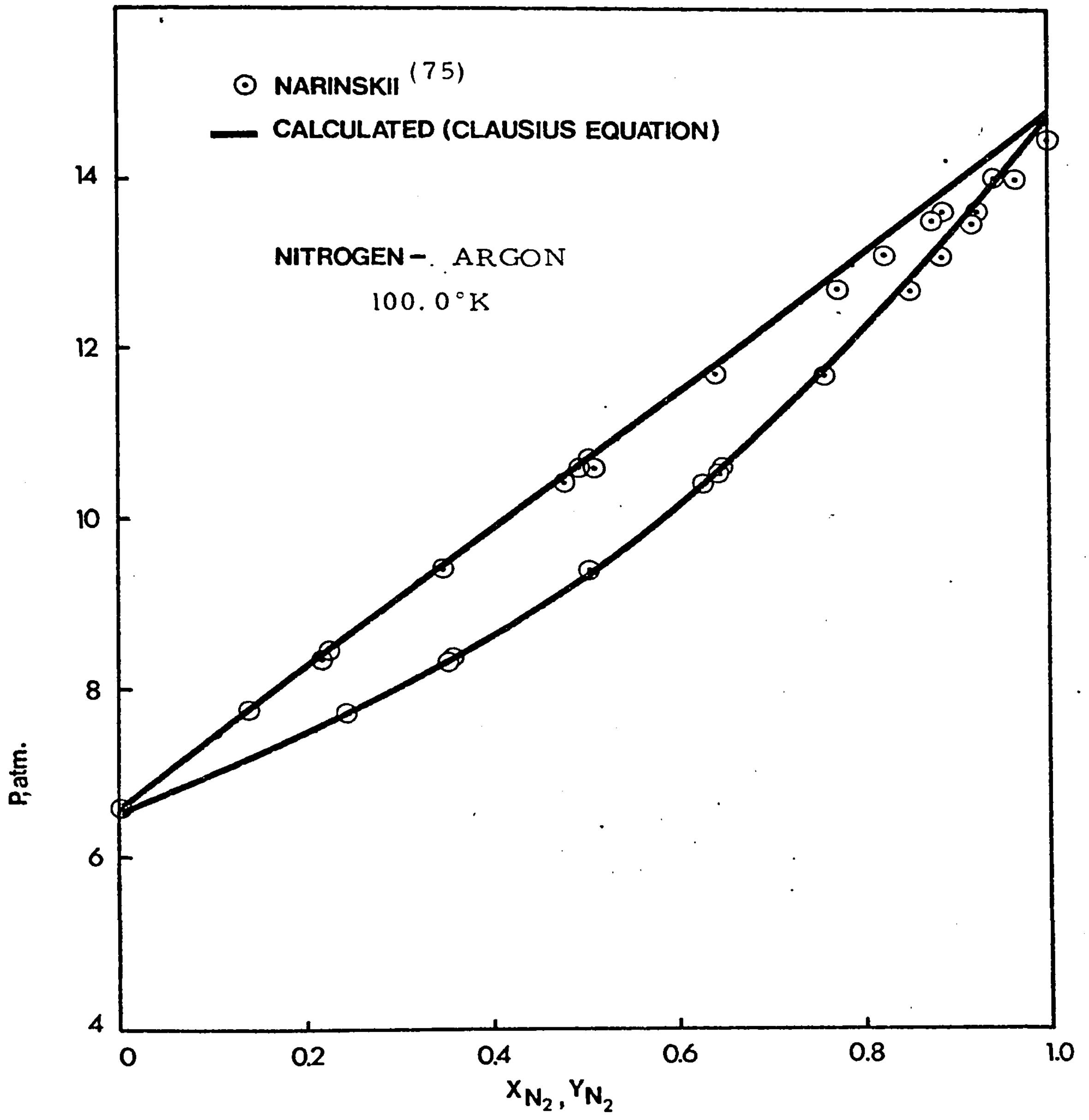


Figure 4-4 Comparison of Calculated and Experimental Vapor-Liquid Equilibrium Data for the System Nitrogen-Argon at 100.0°K

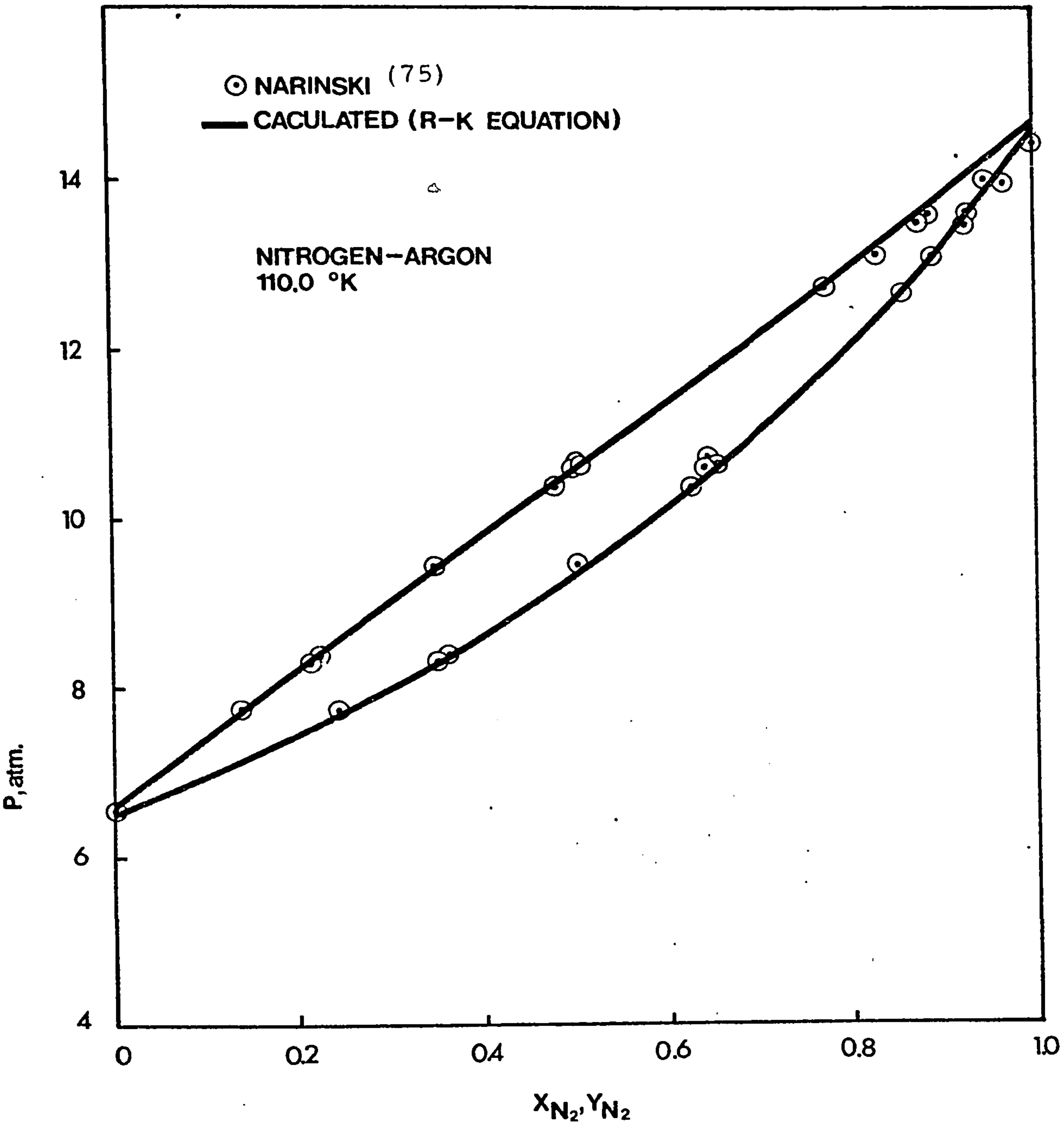


Figure 4-5 Comparison of Calculated and Experimental Vapor-Liquid Equilibrium Data for the System Nitrogen-Argon at 110.0°K

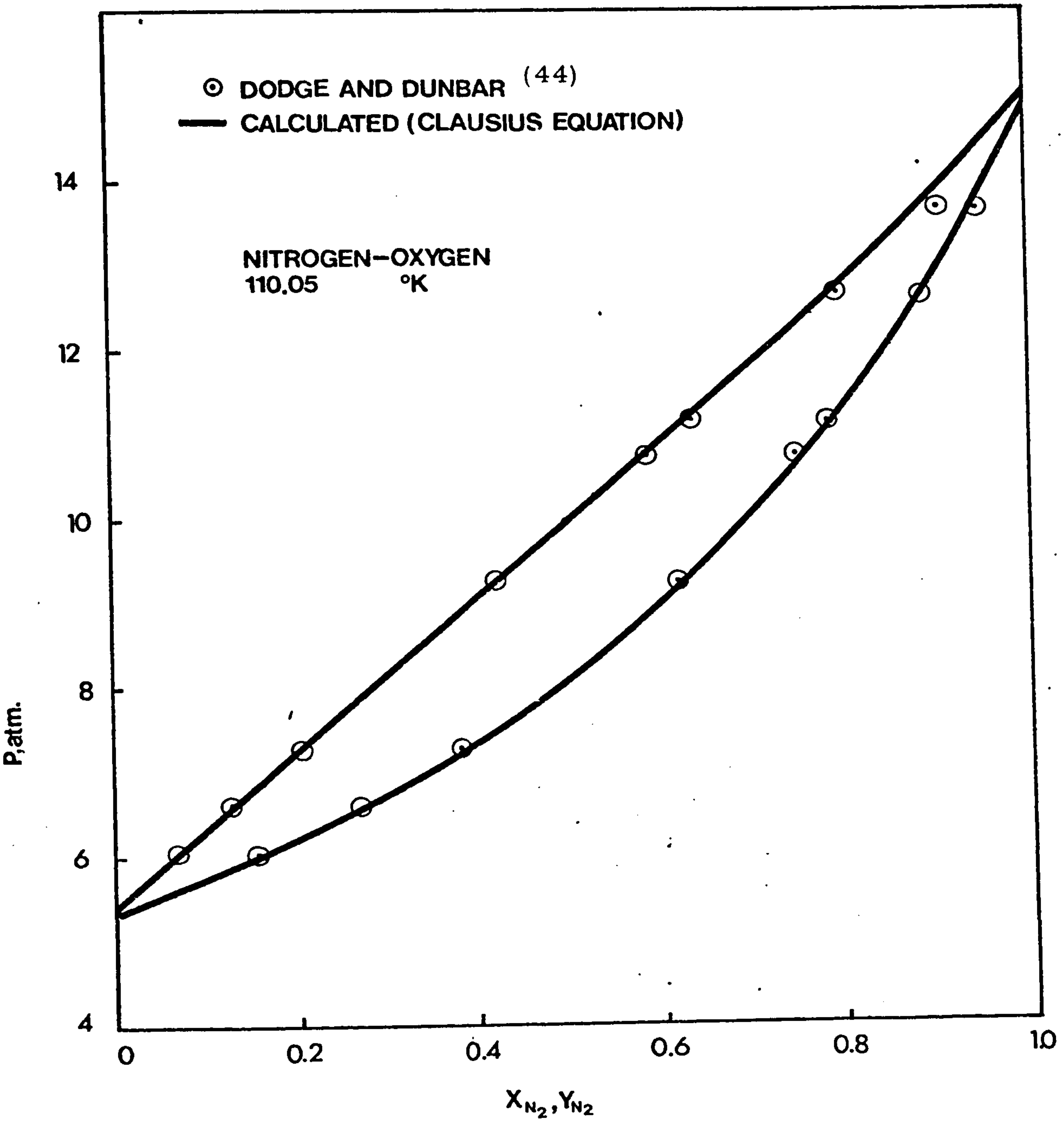


Figure 4-6 Comparison of Calculated and Experimental Vapor-Liquid Equilibrium Data for the System Nitrogen-Oxygen at 110.05°K

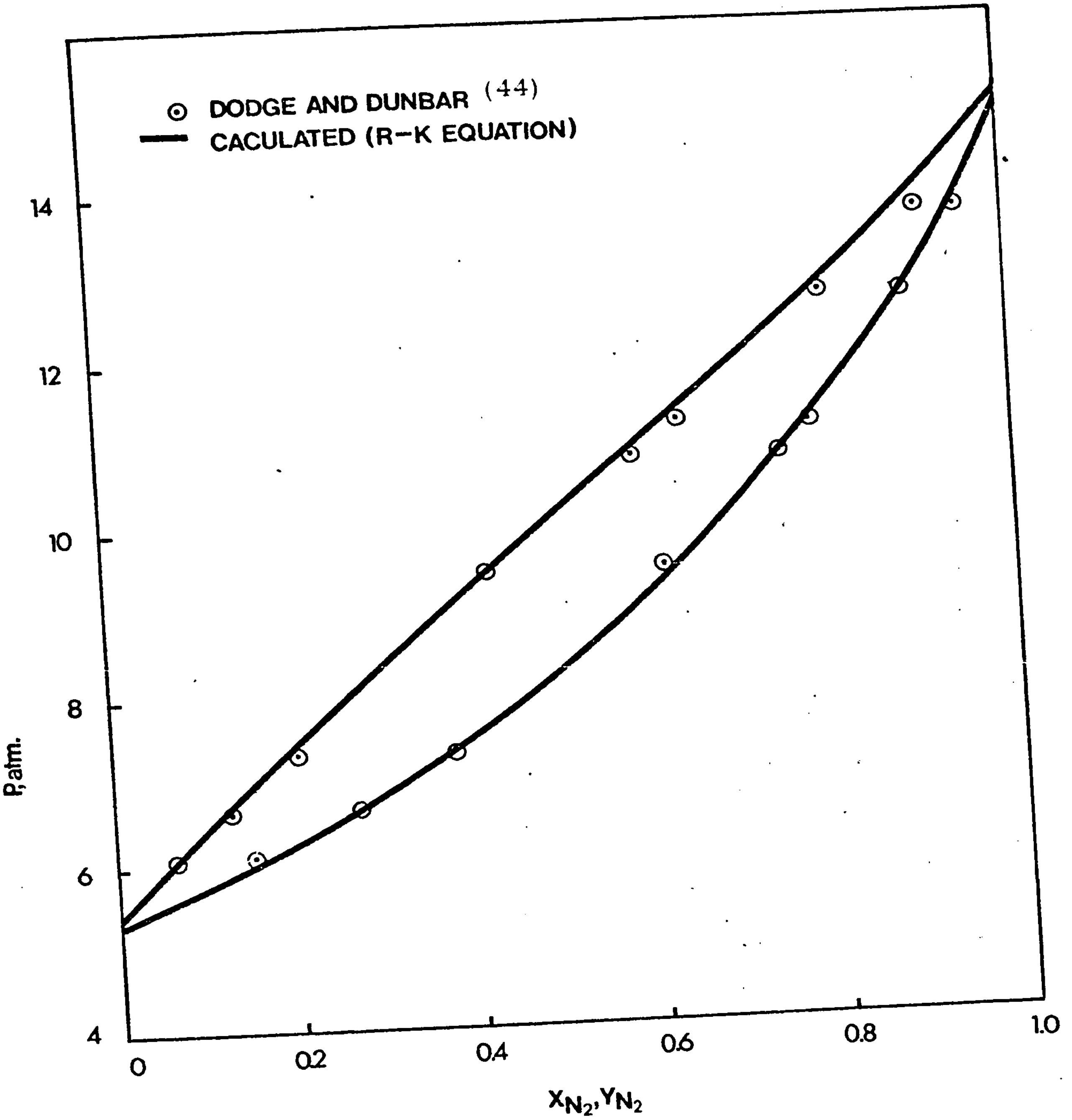


Figure 4-7 Comparison of Calculated and Experimental Vapor-Liquid Equilibrium Data for the System Nitrogen-Oxygen at 110.05°K

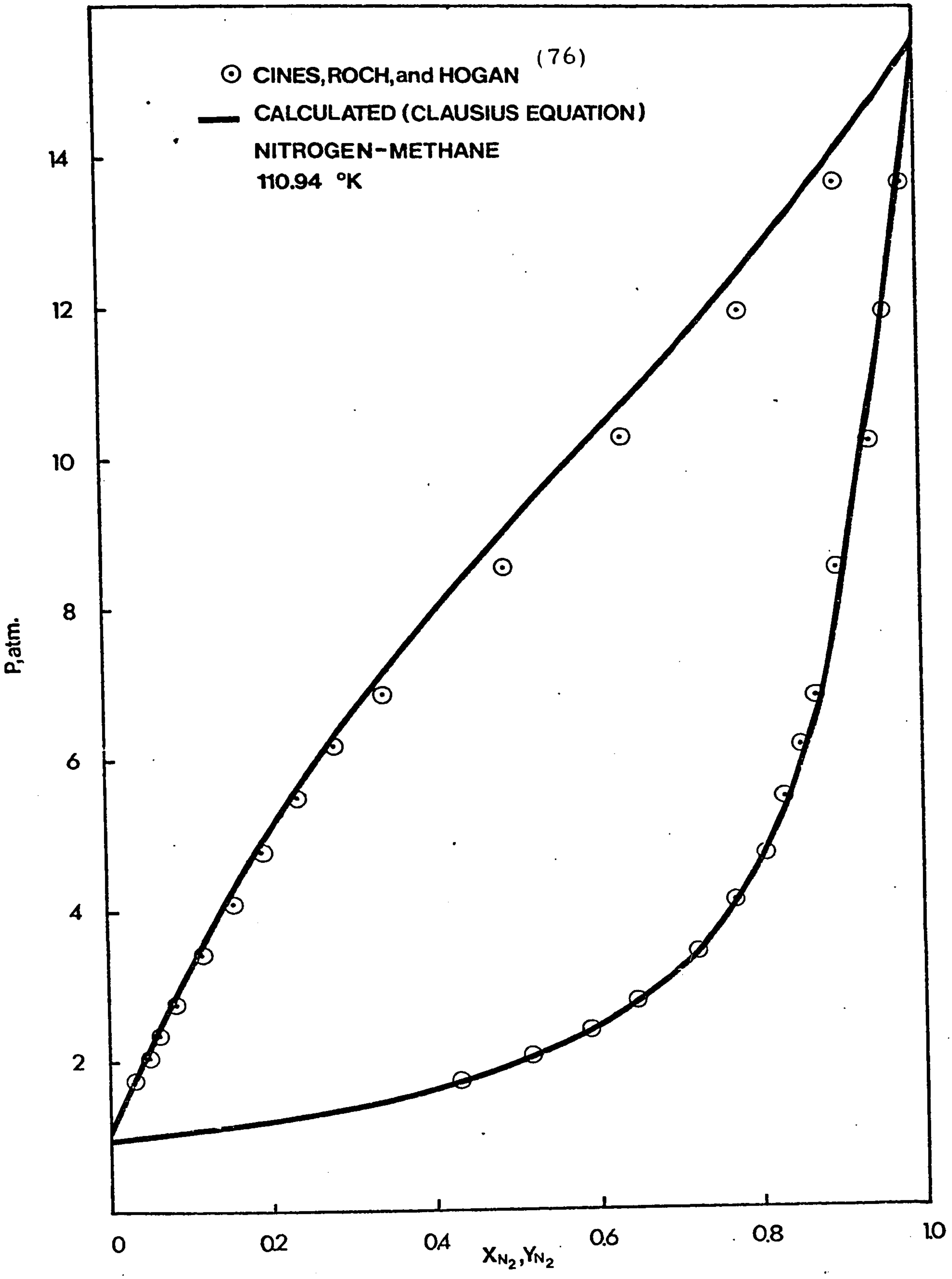


Figure 4-8 Comparison of Calculated and Experimental Vapor-Liquid Equilibrium Data for the System Nitrogen-Methane at 110.94°K

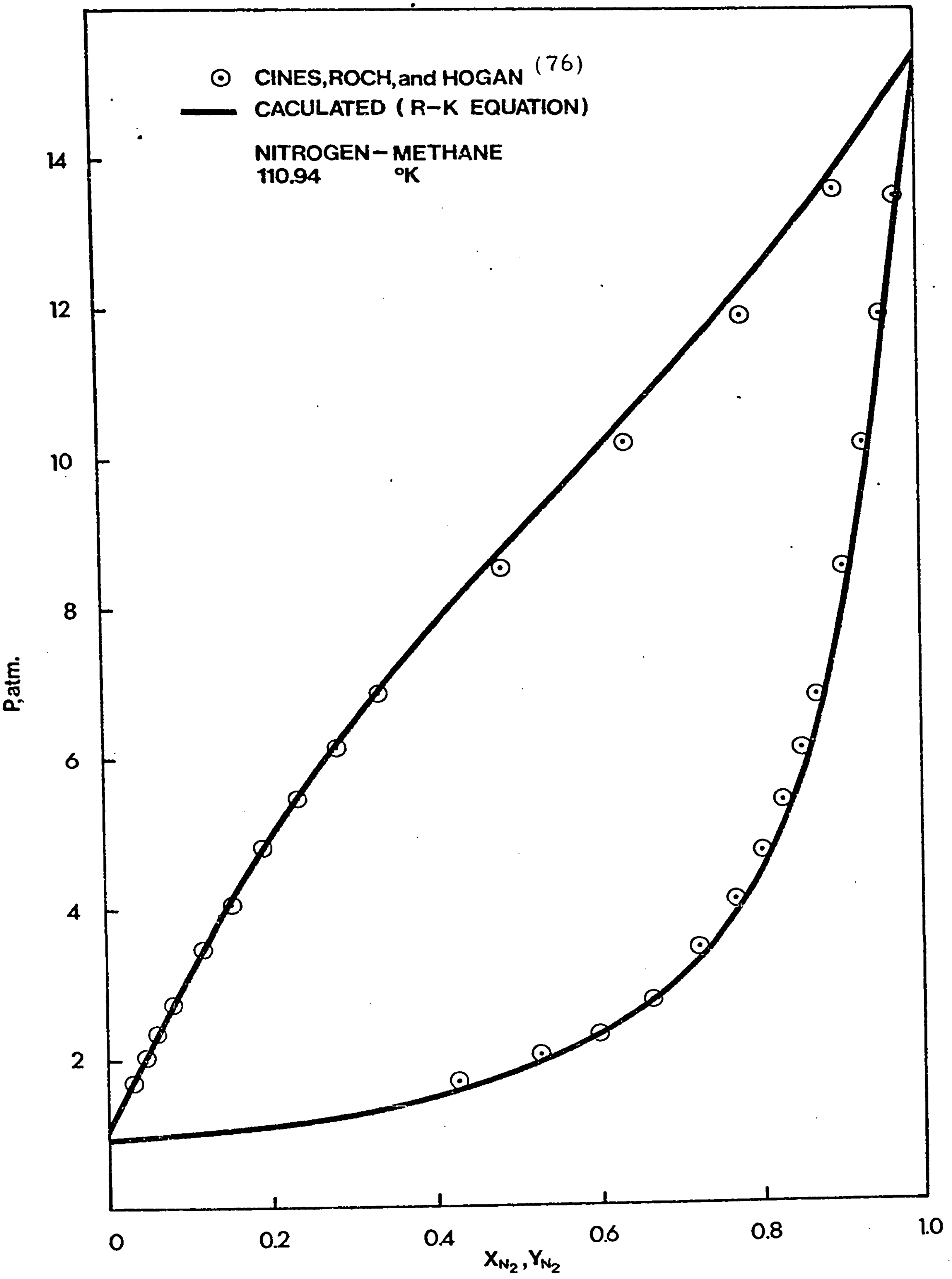


Figure 4-9 Comparison of Calculated and Experimental Vapor-Liquid Equilibrium Data for the System Nitrogen-Methane at 110.94°K

Argon-Oxygen

The experimental data of Narinskii(77) were taken and compared with calculated values for the argon-oxygen system at 110.0°K. Results of comparison are listed in Tables A-I. 11 and A-I. 12 and are shown in Figure (4-10) and (4-11).

Argon-Methane

The prediction of phase equilibrium data for the system argon-methane at 115.22°K was tested against the data of Gravelle(78). Results of the comparison are listed in Tables A-I. 13 and A-I. 14 and shown in Figure (4-12) and (4-13).

Systems containing carbon dioxide and/or hydrogen sulfide as one of the constituents deserve special attention since these mixture exhibit highly non-ideal behavior. In this work the binary systems carbon dioxide-hydrogen sulfide, carbon dioxide-propane and hydrogen sulfide-n-butane were studied for this purpose.

Carbon Dioxide-Hydrogen Sulfide

The experimental data of Sobocinski and Kurata(79) at five isothermal condition, 40°F, 50°F, 60°F, 70°F and 80°F were used for the purpose of comparison. Results of comparison are listed in Tables A-I. 15 to A-I. 17. It was observed that the interaction binary constant, k_{ij}, are not the same at these five isothermal conditions. Then, the calculation was repeated with an average value of k_{ij} equal to 0.072. Results of comparison are given in Table A-I. 16.

Carbon Dioxide-Propane

The predicted results for the carbon dioxide-propane system at 40°F were compared with the experimental data of Reamer et al.(80).

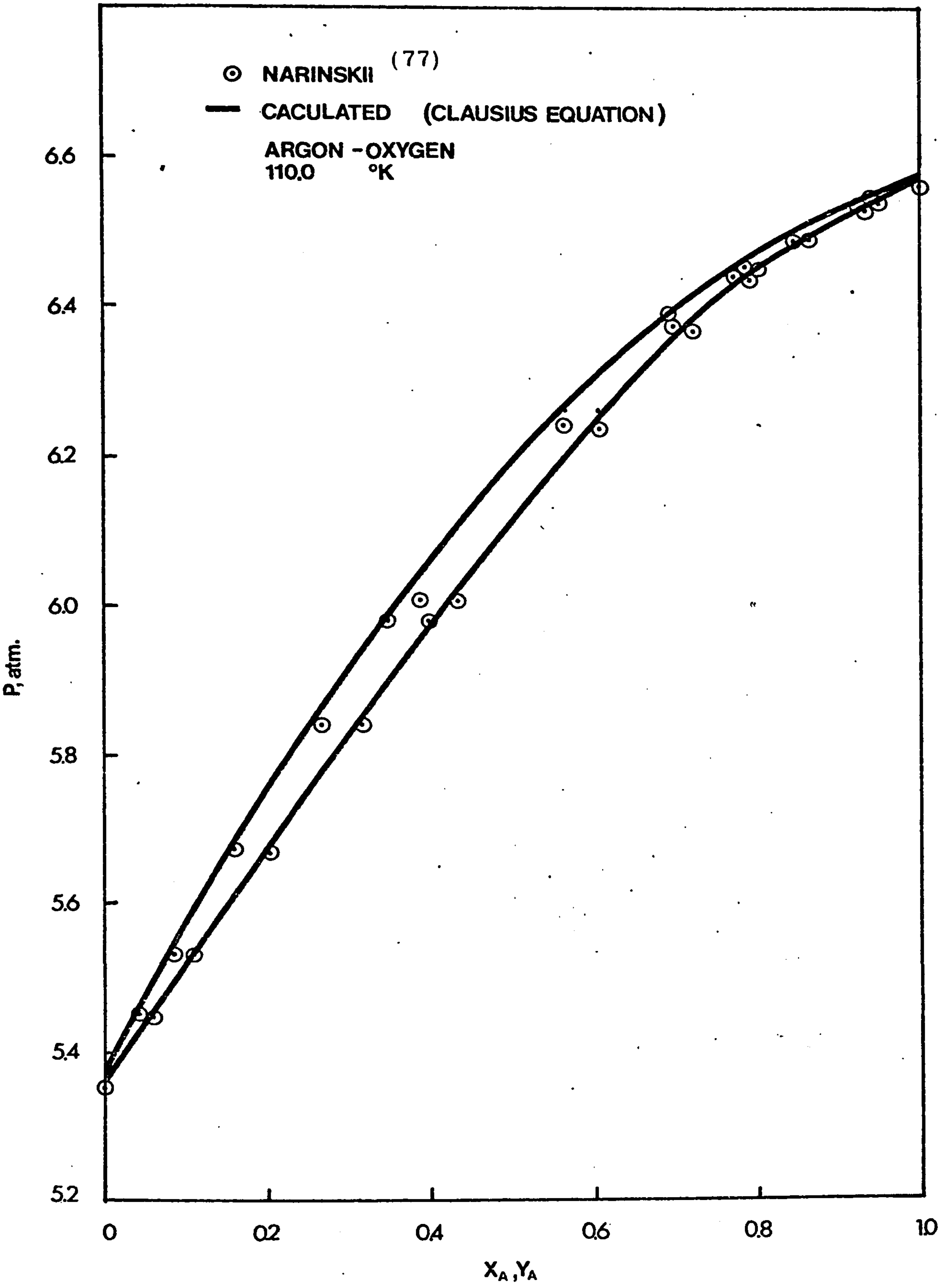


Figure 4-10 Comparison of Calculated and Experimental Vapor-Liquid Equilibrium Data for the System Argon-Oxygen at 100.°K

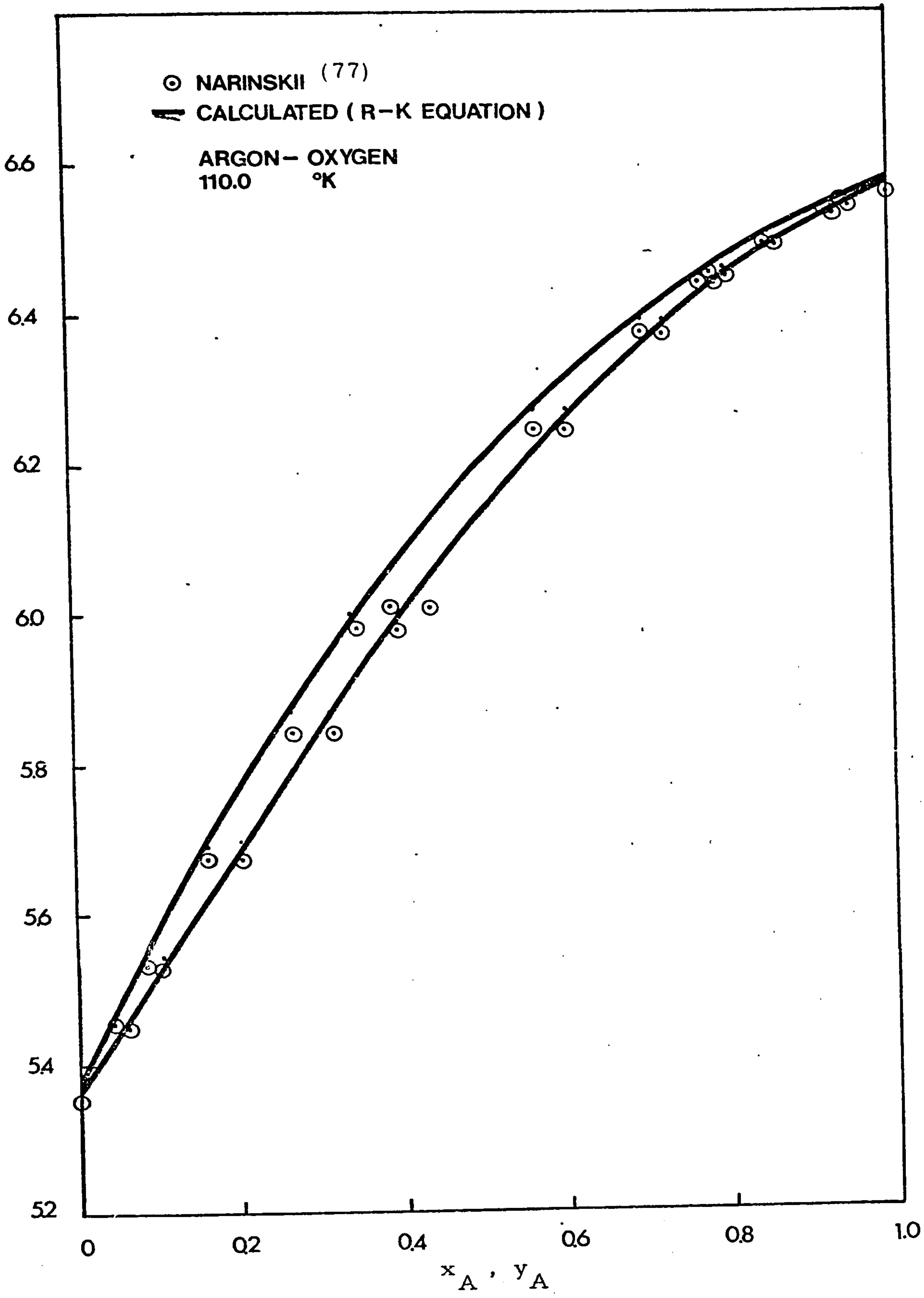


Figure 4-11 Comparison of Calculated and Experimental Vapor-Liquid Equilbirium Data for the System Argon-Oxygen at 110.0°K

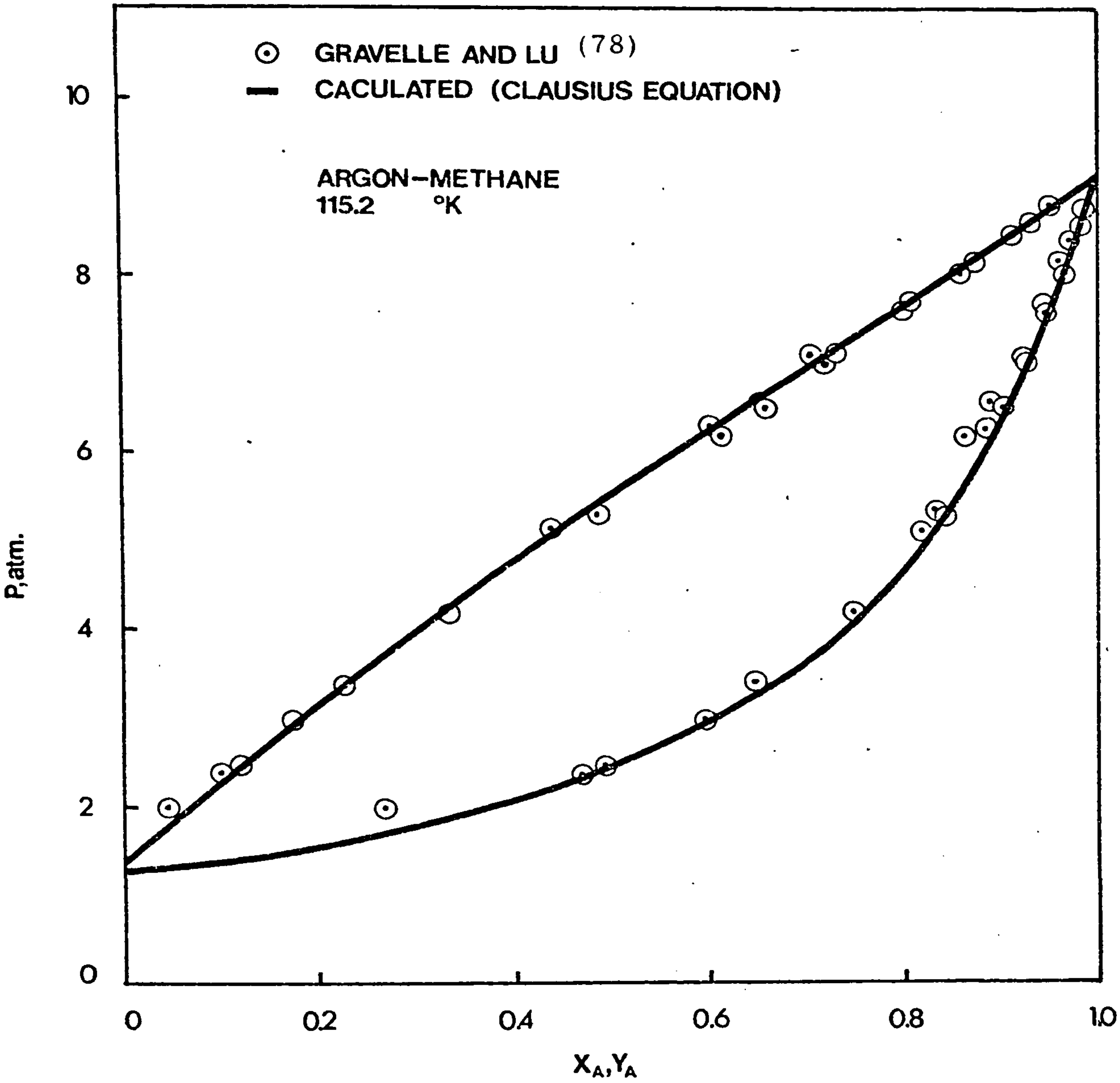


Figure 4-12 Comparison of Calculated and Experimental Vapor-Liquid Equilibrium Data for the System Argon-Methane at 115.2°K

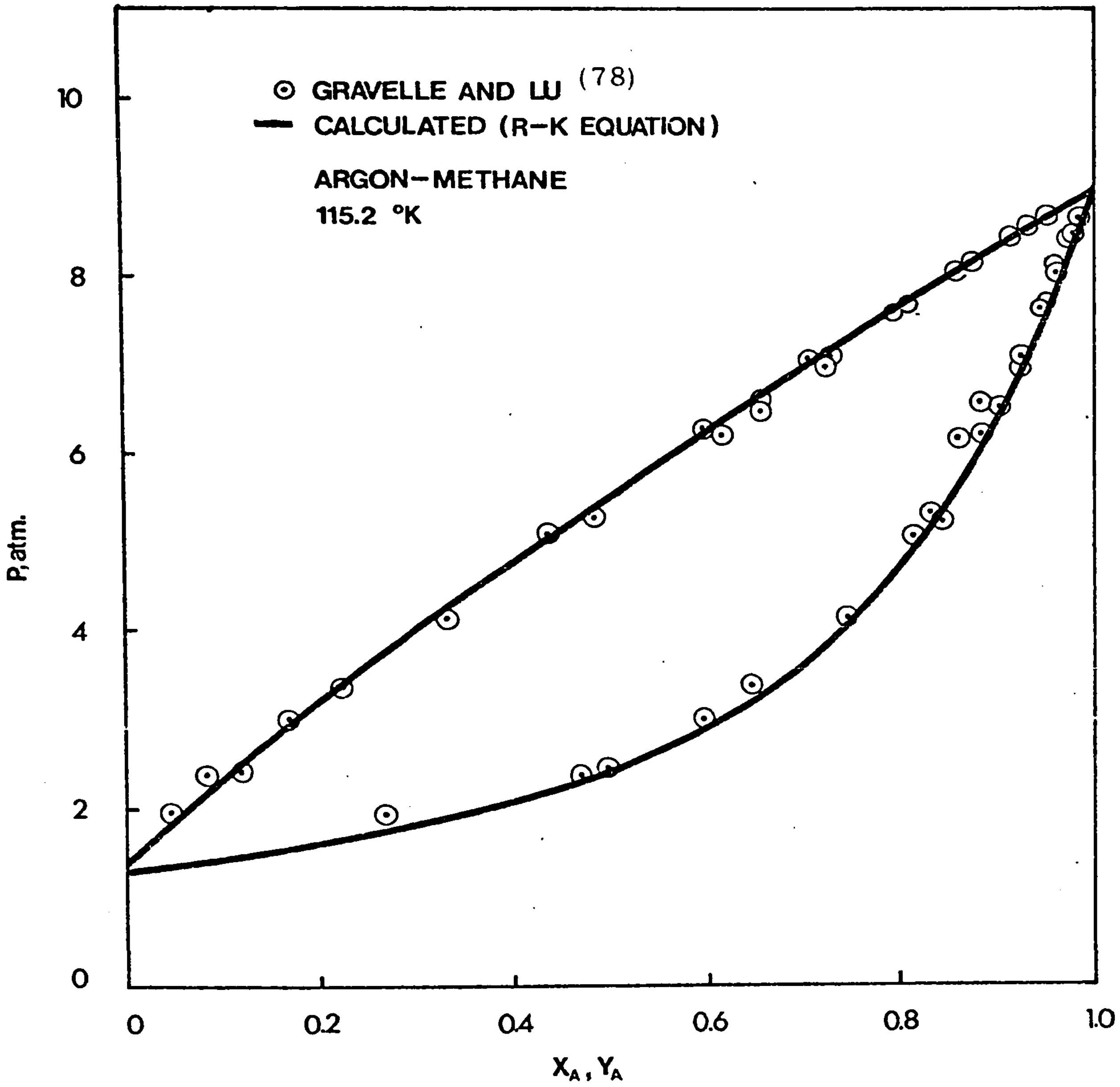


Figure 4-13 Comparison of Calculated and Experimental Vapor-Liquid Equilibrium Data for the System Argon-Methane at 115. 2°K

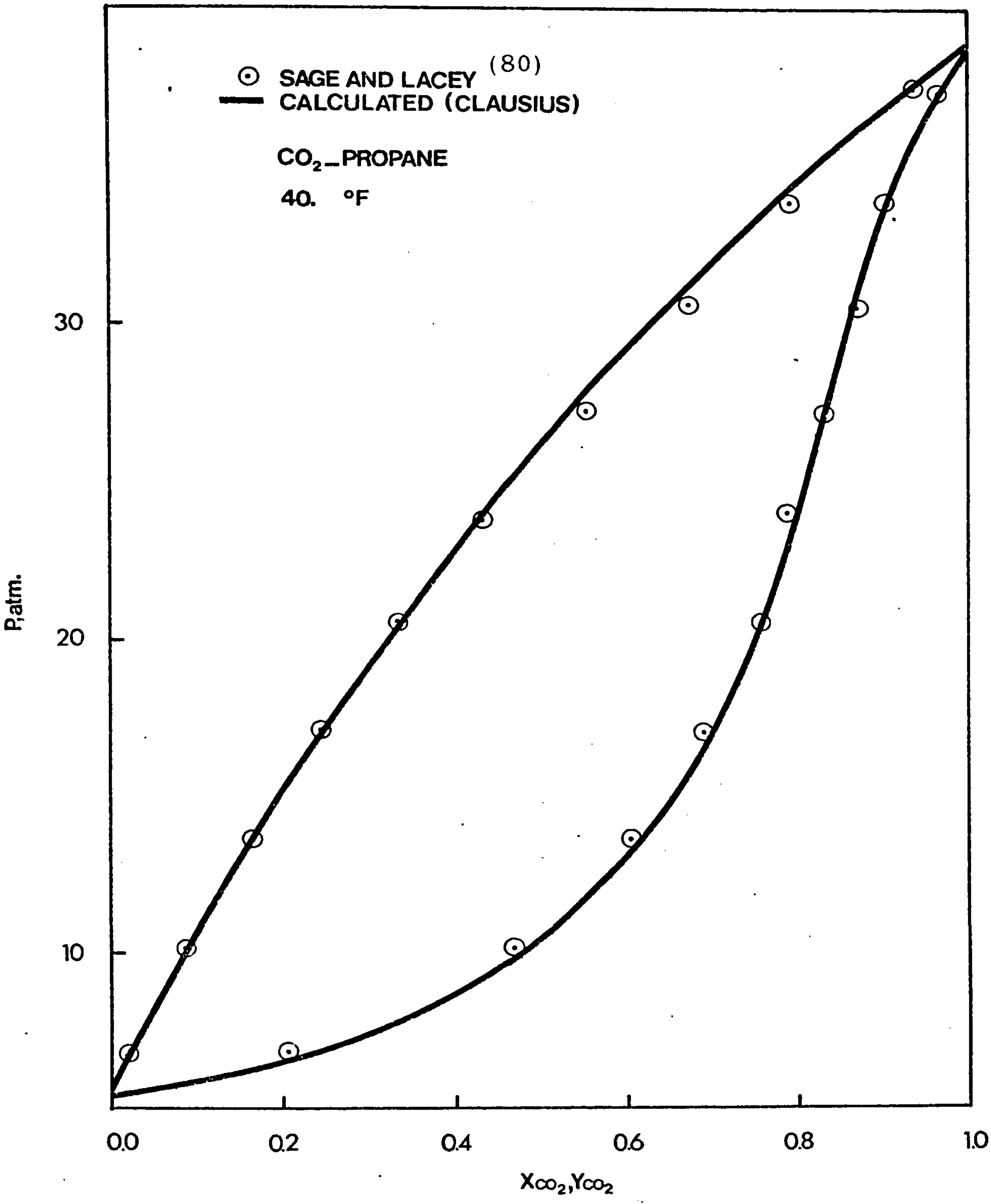


Figure 4-14 Comparison of Calculated and Experimental Vapor-Liquid Equilibrium Data for the System Carbon Dioxide-Propane at 40° F.

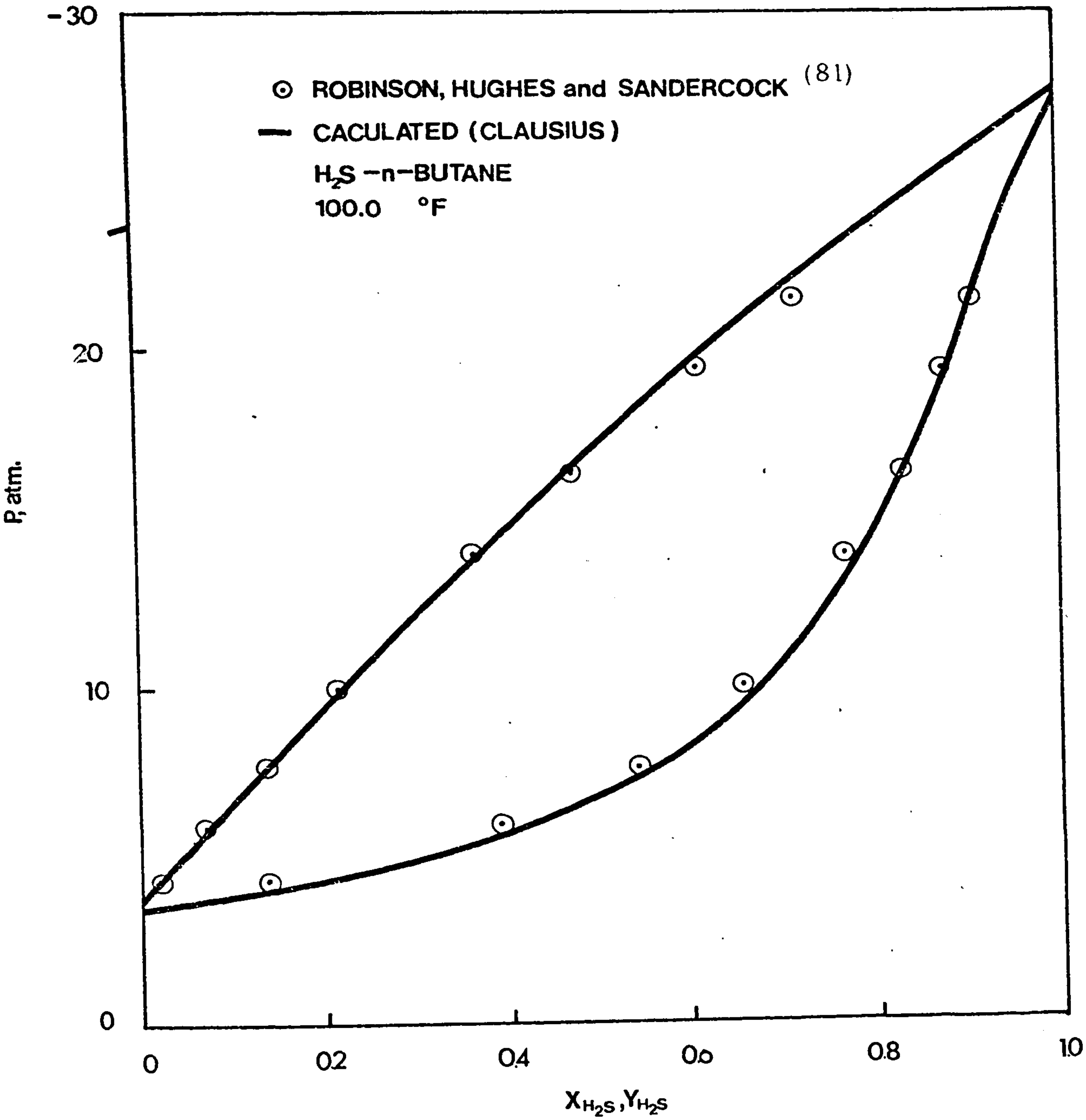


Figure 4-15 Comparison of Calculated and Experimental Vapor-Liquid Equilibrium Data for the System Hydrogen Sulfide-n-Butane at 100°F.

Results of the comparison are shown in Figure (4-14) and listed in Tables A-I. 18 and A-I. 19.

Hydrogen Sulfide-n-Butane

The experimental data of Robinson et al (81) were taken and compared with calculated values for the hydrogen sulfide-n-butane system at 100° F. Results of the comparison are listed in Tables A-I. 20 and A-I. 21 and also shown in Figure (4-15).

Nitrogen-Argon-Oxygen

The binary interaction constants, k_{ij}, computed from experimental phase equilibrium data for the binaries nitrogen-argon, nitrogen-oxygen and argon-oxygen systems were employed to predict vapor-liquid equilibria data for the ternary system nitrogen-argon-oxygen at 110. 0°K. Using the proposed method and the modified R-K equation. The calculated results were compared with the experimental data of Narinskii (82) and Wilson (83) in Tables A-I. 22 to A-I. 25.

4. 5 A TOTAL PRESSURE METHOD FOR SYSTEMS AT HIGH PRESSURES

The theoretical correlation of activity coefficients for any multicomponent mixture derived from the Gibbs-Duhem equation

$$\sum_{i=1}^{N} x_i \, d\,(\ln \gamma_i) = \frac{v^E}{RT} \, dP - \frac{H^E}{RT^2} \, dT \qquad (4\text{-}68)$$

At constant temperature Equation (4-68) reduces to

$$\sum_{i=1}^{N} x \, d\,(\ln \gamma_i) = \frac{v^E}{RT} \, dP \qquad (4\text{-}69)$$

At low pressure various workers(84, 85) have suggested that for many isothermal systems the right hand side of Equation (4-69) is small and can be taken as zero. This implies that we are using for isothermal conditions the isothermal-isobaric form of Gibbs-Duhem equation.

At high pressure this assumption is not valid. Due to this reason, a method has been proposed to calculate vapor-phase composition from total pressure-liquid phase composition measurements for binary systems at high pressures.

4.5.1 DERIVATION OF EQUATIONS

For vapor-liquid equilibria of a binary system the following equation must be satisfied

$$\hat{f}_i^L = \hat{f}_i^v \qquad i = 1, 2 \tag{4-70}$$

where,

$$\hat{f}_i^v = y_i \hat{\varphi}_i^v P \tag{4-71}$$

$$\hat{f}_i^L = x_i \hat{\varphi}_i^L P = \gamma_i x_i f_{iL}^\circ \tag{4-72}$$

The activity coefficient of component i in the liquid-phase(86) is given by

$$\gamma_i (P^\circ, T, x_i) = \frac{\hat{f}_i^L (P^\circ, T, x_i)}{f^\circ_{iL} (P^\circ, T)\, x_i} \tag{4-73}$$

where f_i^L and x_i are the fugacity and composition of component i in the liquid mixtures, and f°_{iL} is the reference fugacity of component i at the reference pressure P°. The pressure effect on the fugacity coefficient is

$$\left(\frac{\partial \ln f^\circ_{iL}}{\partial P}\right)_T = \frac{\bar{v}_i}{RT} \tag{4-74}$$

then,

$$\gamma_i (P^\circ, T, x_i) = \frac{\hat{f}_{iL} (P, T, x_i)}{f^\circ_{iL} (P^\circ, T) x_i} \exp \int_P^{P^\circ} \frac{\bar{v}_i}{RT} d P \tag{4-75}$$

For convenience Equation (4-75) may be written in the following form:

$$\gamma_i = \frac{\hat{f}_{iL}}{f^\circ_{iL} \; x_i} \exp \int_P^{P^\circ} \frac{\bar{v}_i}{RT} d P \tag{4-76}$$

The following convention was used to normalize the activity coefficient

$$\gamma_i \longrightarrow 1.0 \quad , \quad \text{as} \quad x_i \longrightarrow 1.0 \tag{4-77}$$

Combining Equations (4-70) to (4-72), the total pressure and the vapor-phase composition are given by the following equations:

$$P = \sum_{i=1}^{N} \frac{\gamma_i f^\circ_{iL} x_i}{\hat{\varphi}_{iv}} \tag{4-78}$$

and

$$y_i = (\hat{\varphi}_{iL} / \hat{\varphi}_{iv}) \; x_i \tag{4-79}$$

Equations (4-78) and (4-79) are the general and basic equations for computing vapor phase compositions from total pressure-liquid phase compositions data of isothermal conditions. The supporting properties such as $\hat{\varphi}_i$ and $\bar{v}_i$ are calculated using the modified Clausius equation of state.

4. 5. 2 RESULTS AND DISCUSSIONS

The modified total pressure method was used to predict vapor-phase composition for the binary systems ethylene-propane, argon-methane, and nitrogen-argon. The fugacity coefficients, and partial molal volumes were calculated using the modified Clausius equation of state. As mentioned previously, the temperature dependent parameters Ω_a, Ω_b, and Ω_c were evaluated using vapor pressure and saturated liquid and vapor molar volumes of pure components(64, 87, 88). Values of the parameters are listed in Table .

The numerical routine for determining vapor-phase composition is shown in Figure (4-16) and may be summarized as follows:

1. Initial values of y_i's and $\hat{\varphi}_{iv}$'s ($\hat{\varphi}_{iv} = 1.0$) are assumed for the first iteration.
2. Liquid-phase fugacity coefficients, $\hat{\varphi}_{iL}$'s, and activity coefficients, y_i's, are calculated using Equations (4-64) and (4-79).
3. The total pressure, P, is calculated from Equation (4-78) and vapor-phase composition using Equation (4-79).
4. The calculated and experimental total pressure values are then compared. If they are disagreeable, y_i's are used to calculate new values of $\hat{\varphi}_{iv}$ and the whole procedure is repeated until a good agreement is obtained between experimental and calculated values of total pressure.

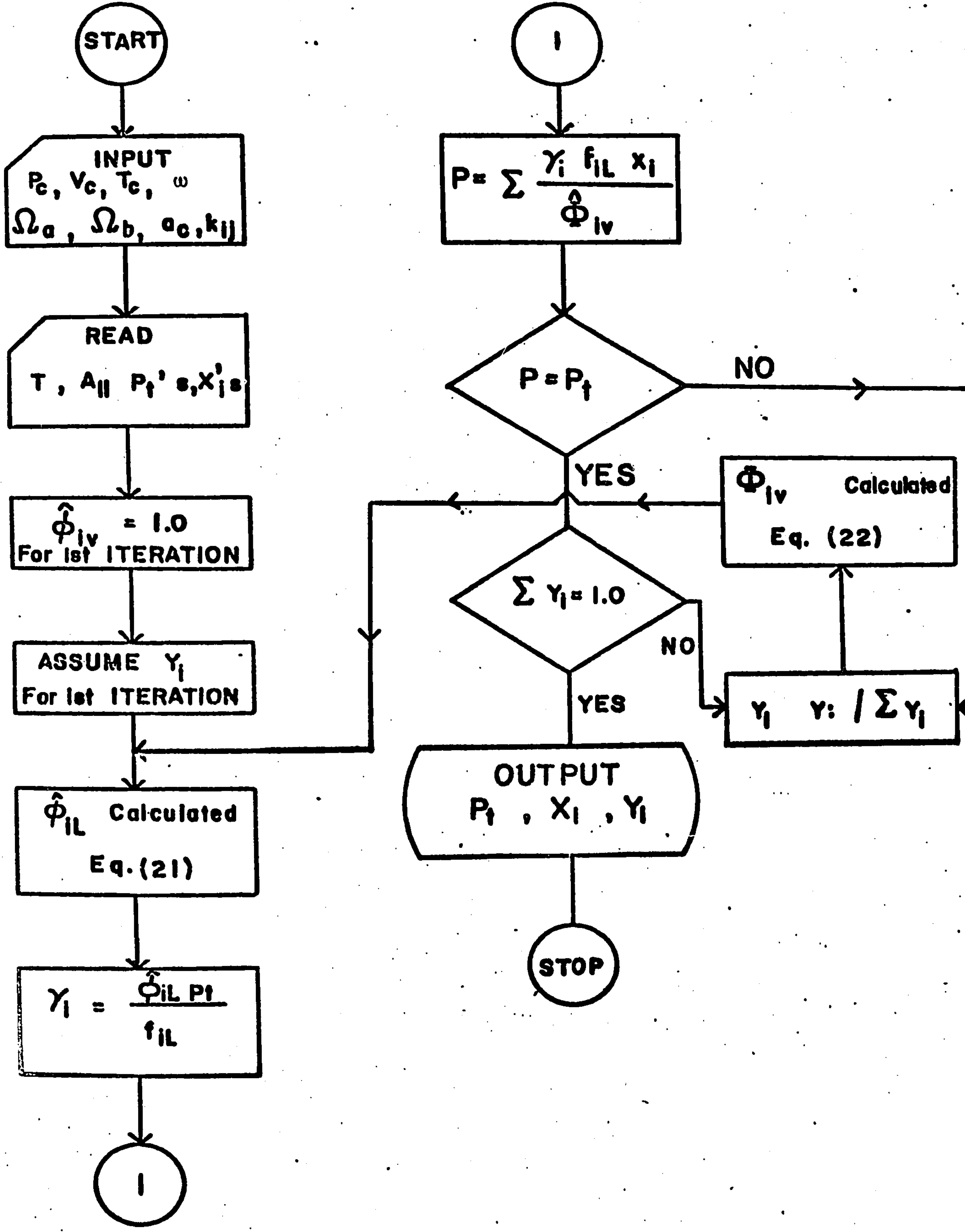


Figure 4-16 Schematic Diagram of Vapor-Phase Composition Calculation

TABLE 4-2

Values of Redlich-Kister Equation Constants

System Ethylene-Propane

Temp. °K	B	C	D
199.83	0.3103	0.0553	0.009
277.9	0.2086	0.0334	0.0004
255.35	0.1129	0.0347	0.0160
173.06	0.2509	0.0698	0.0420
System Argon - Methane			
115.22	0.3477	0.0420	0.0045
System Nitrogen-Argon			
100	0.1235	-0.0090	0.0011

The modified equation of state was tested for the prediction of vapor-phase composition from total pressure-liquid phase composition measurements. The calculated values were compared with experimental data for several system.

Argon-Methane

The predicted results for the argon-methane system at 115.22°K were compared with the reported experimental results in Chapter III. Results of comparison are listed in Table A-I. 26 and shown in Figure (4-17). The maximum average absolute deviation in vapor phase composition is 0.19%.

Nitrogen-Argon

A comparison of calculated and experimental results reported in Chapter III for the system nitrogen-argon at 100.0°K was made. A good agreement was obtained. Results of comparison are listed in Table A-I. 27 and shown in Figure (4-18).

Ethylene-Propane

The experimental results reported in Chapter III were used to predict vapor phase-composition for the system ethylene propane at four isothermal conditions namely, 273.06°K, 255.38°K, 227.9°K , and 199.83°K. Results of calculation are shown in Figure (4-19) to Figure (4-22) and listed in Table A-I. 28.

The activity coefficients of these systems were also correlated by means of Redlich-Kister equation. Values of these constants are listed in Table (4-2). It was found that the two isotherms, 273.06°K and 255.38°K, exhibit a slight negative deviation from Raoult's law which is in agreement with the conclusions established by King(89).

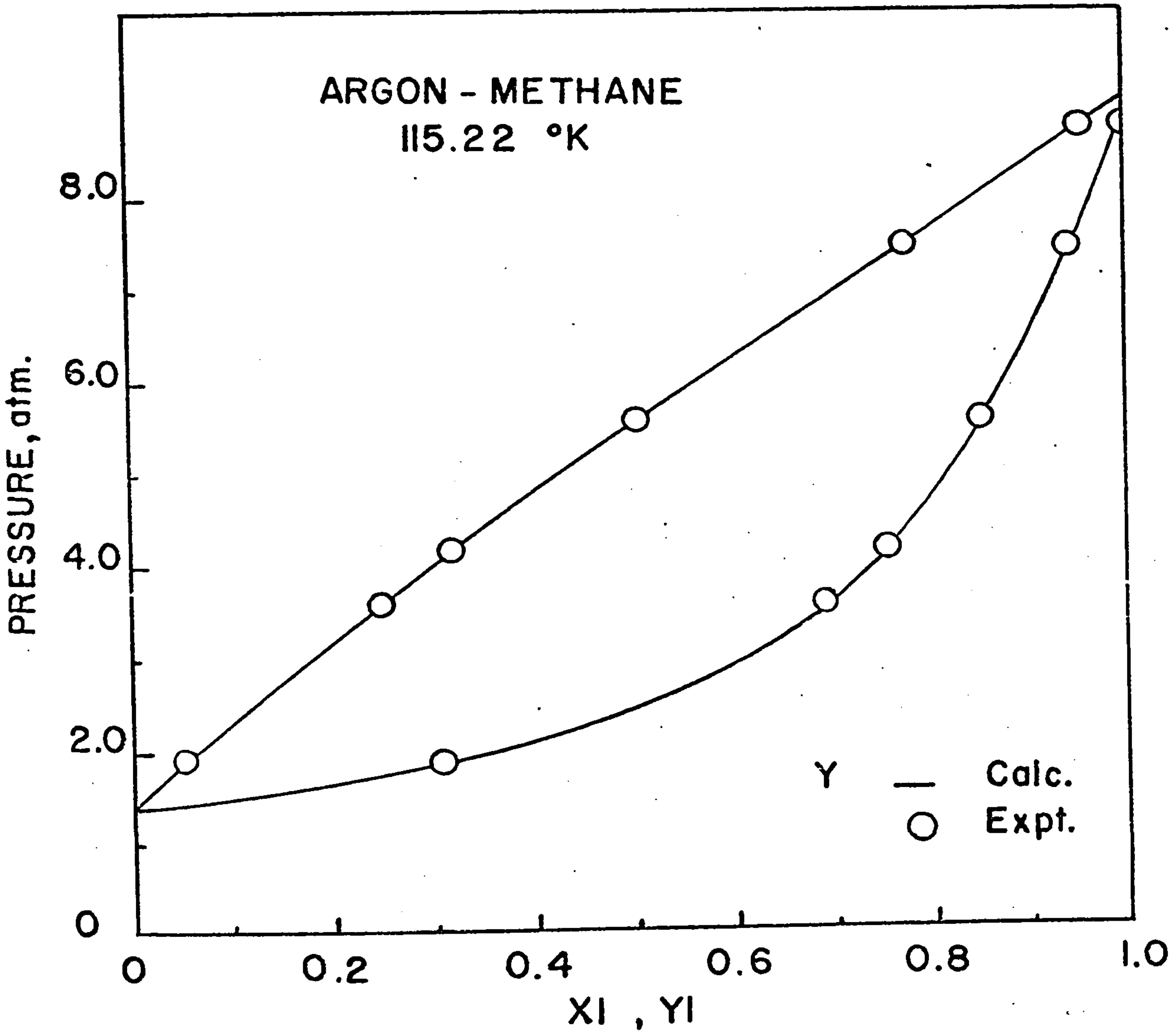


Figure 4-17 Comparison of Calculated and Experimental Vapor-Phase Compositions of Argon-Methane System at 115.22°K

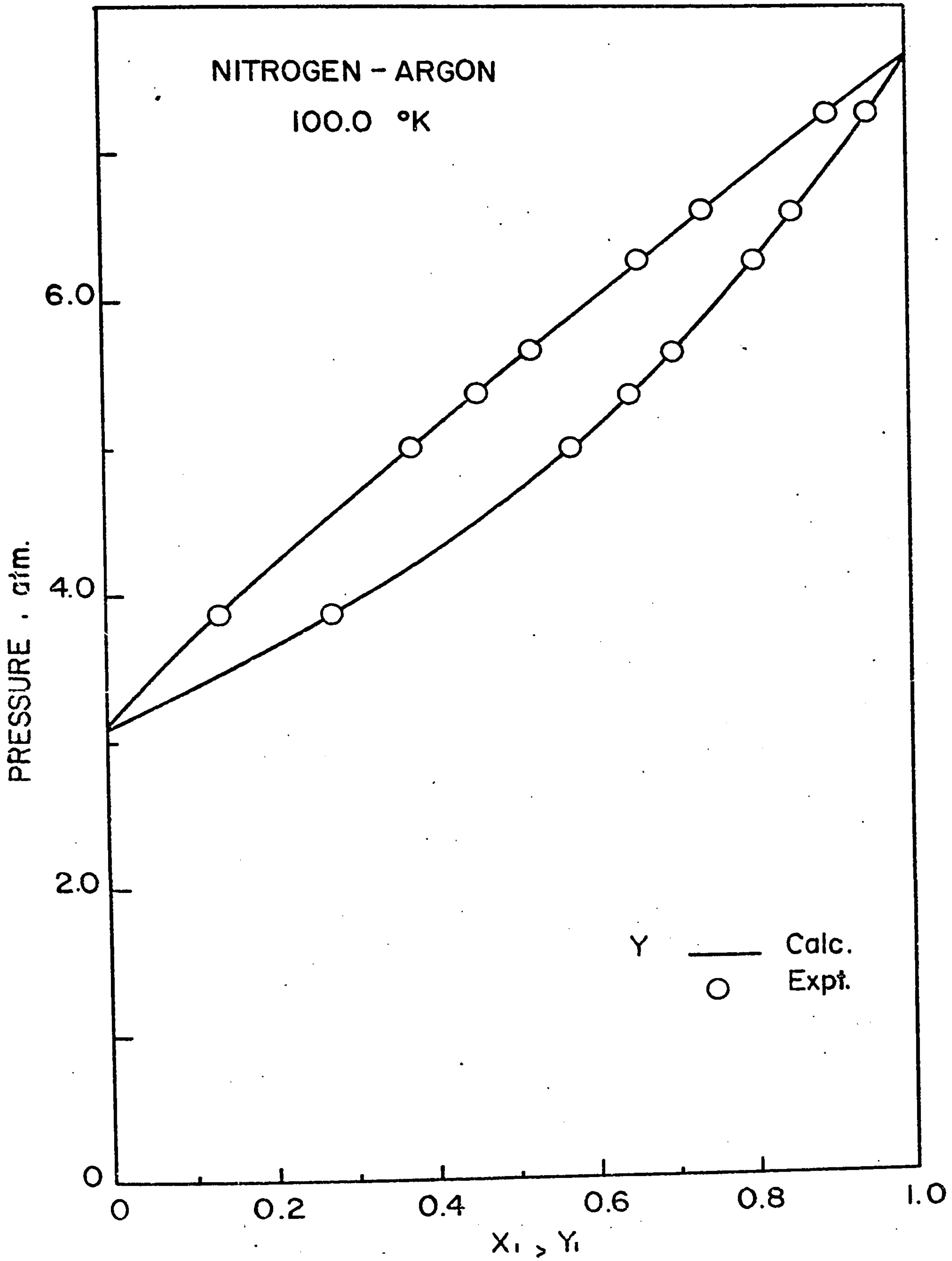


Figure 4-18 Comparison of Calculated and Experimental Vapor-Phase Compositions of Nitrogen-Argon System at 100.0°K

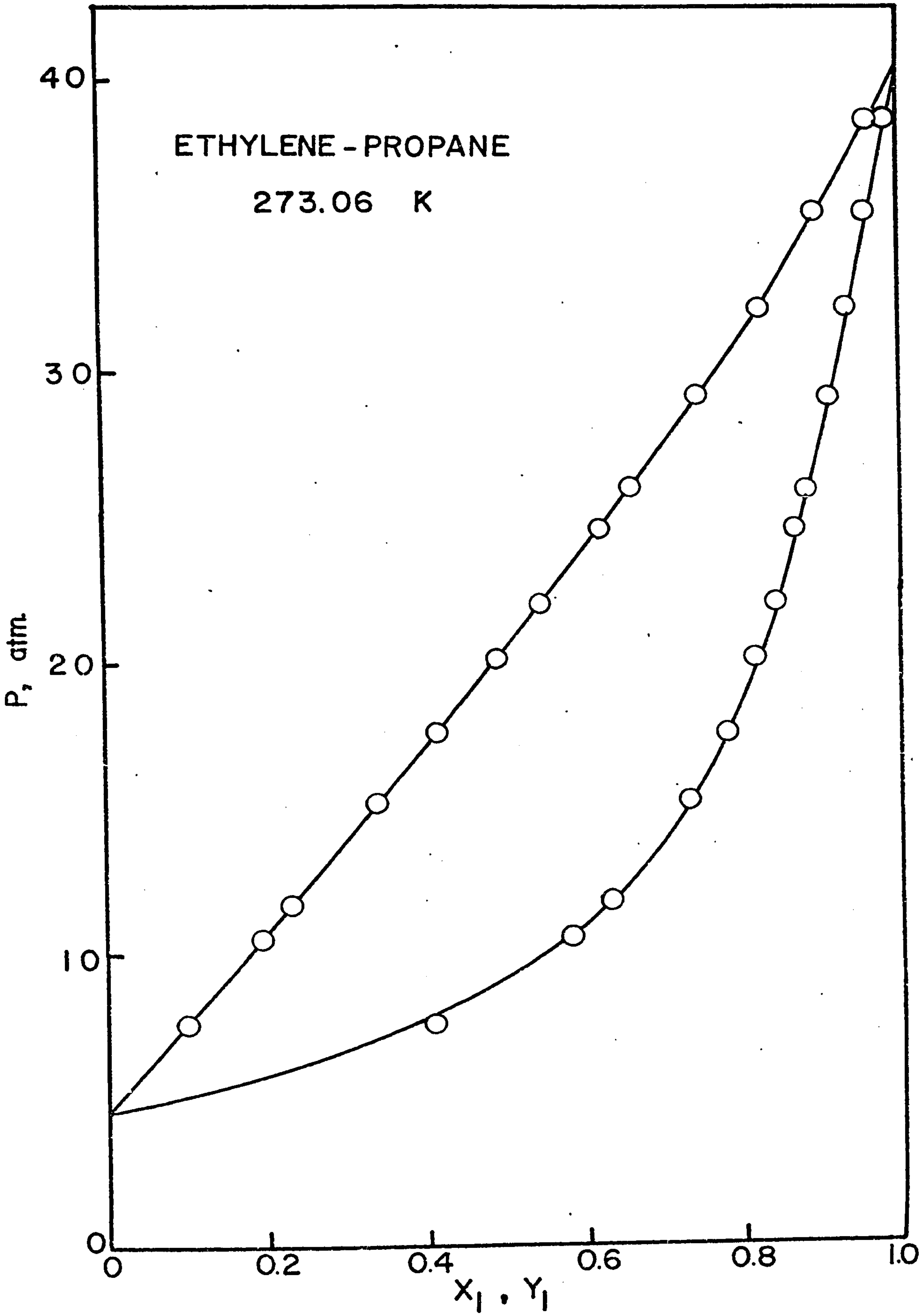


Figure 4-19 Calculated Vapor-Phase Compositions of Ethylene-Propane System at 273.06°K

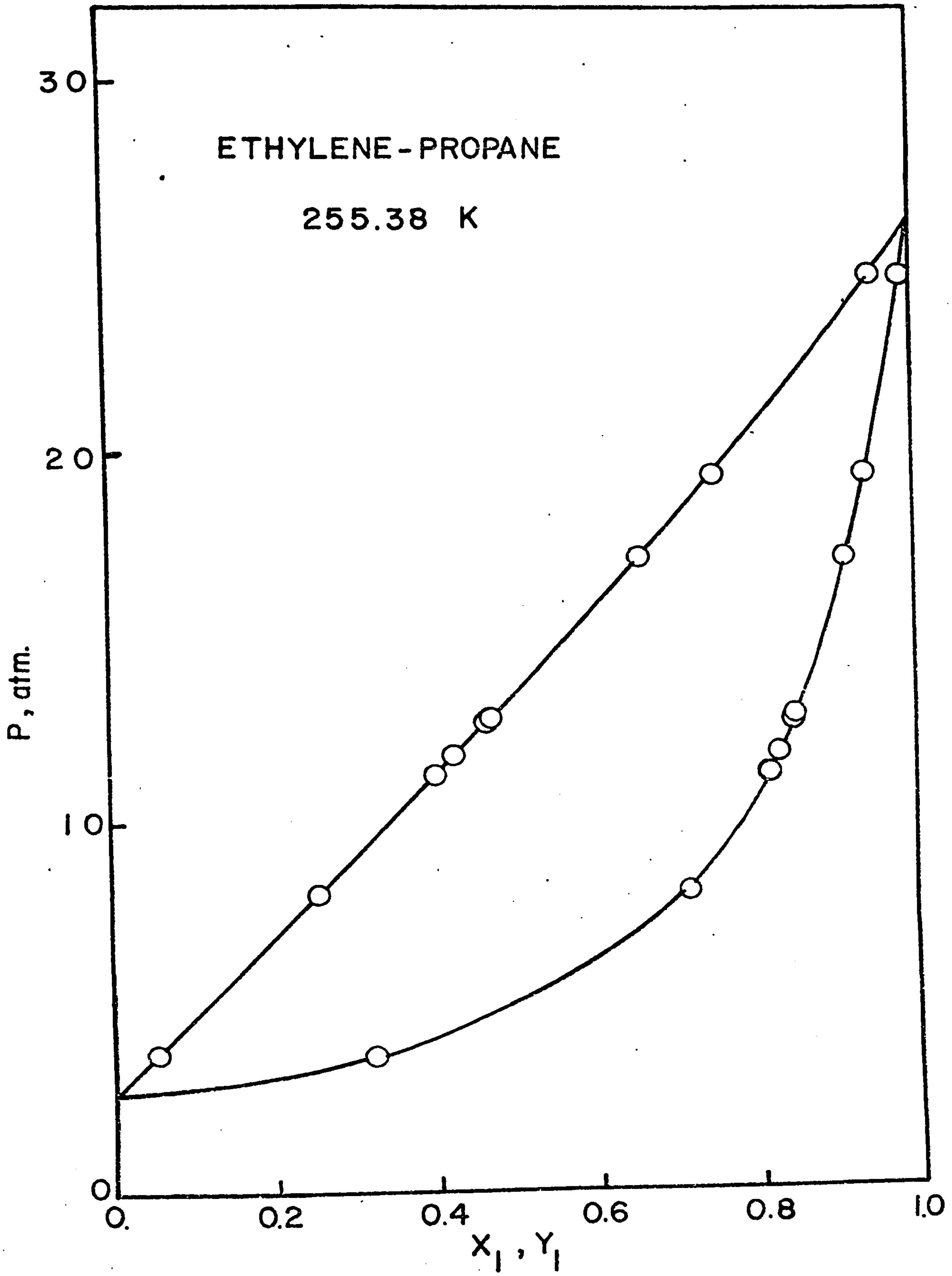


Figure 4-20 Calculated Vapor-Phase Compositions of Ethylene-Propane System at 255.38°K

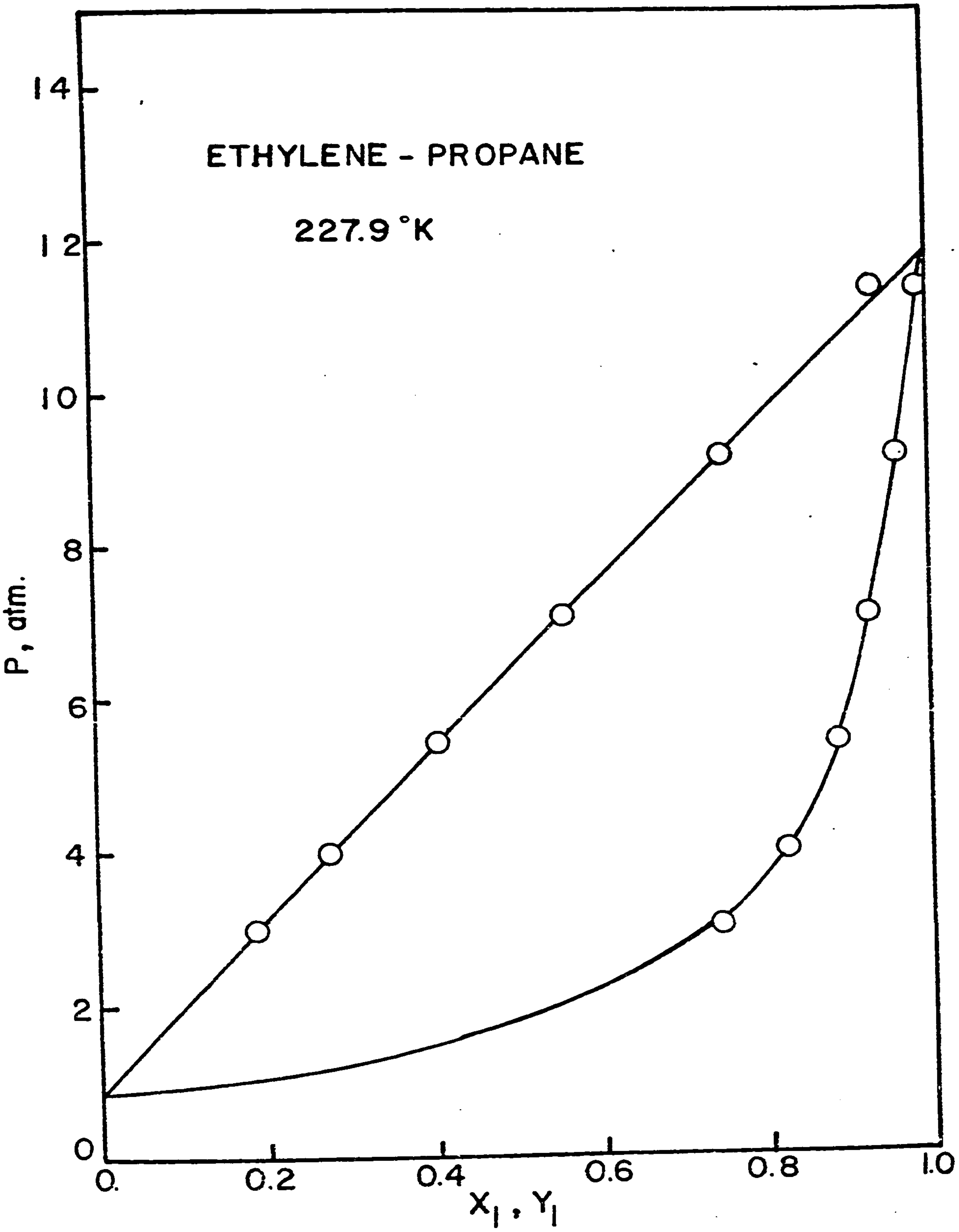


Figure 4-21 Calculated Vapor-Phase Compositions of Ethylene-Propane System at 227.9°K

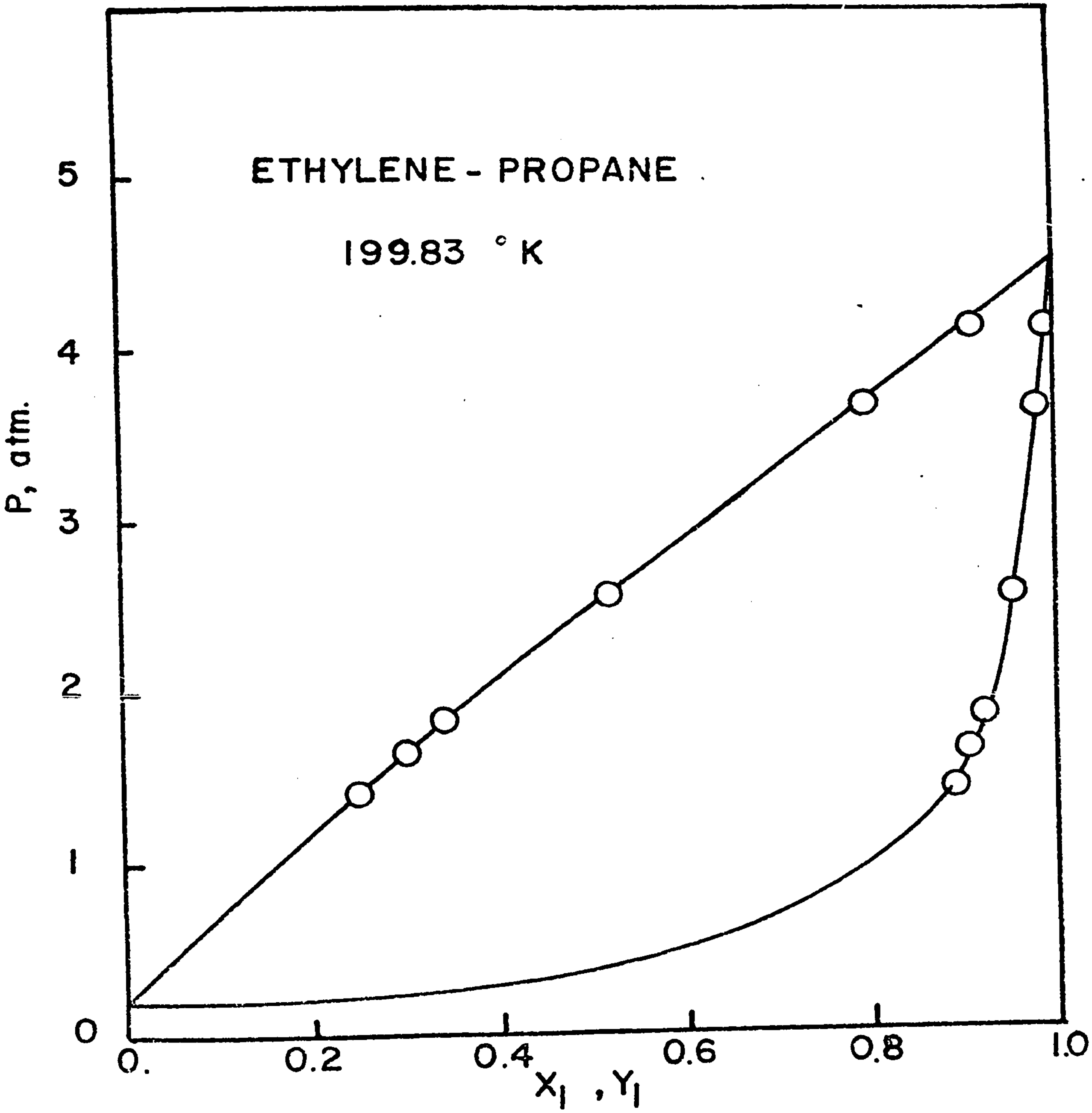


Figure 4-22 Calculated Vapor-Phase Compositions of Ethylene-Propane System at 199.83°K

4.6 CONCLUSIONS

An attempt has been made in this study to improve the Clausius equation of state by considering its parameters temperature dependent. With this modification, the Clausius equation of state was found to be capable of predicting the volumetric data of pure substances in the homogenous region. In order to illustrate the capabilities of the modified equation, the calculated results were compared with those of the modified R-K(41) equation of state. In the liquid phase region results of comparison indicated that the modified Clausius equation of state is comparable with the modified R-K equation. In the vapor phase region, the modified Clausius equation was found to be superior than the modified R-K equation of state. At saturation the modified Clausius equation satisfies the three conditions (4-1) - (4-3) while the modified R-K equation of state relaxes the third condition (4-3). This relaxation in condition (4-3) will produce high error in the calculation of the compressibility factor, for example, the error may be as high as 100% as in the case of pure carbon dioxide at 80° F.

A detailed calculation procedure has been developed for the calculation of system pressure, P, and vapor phase composition, y, from a given liquid phase composition, x, and system temperature, T. The only requirements for the calculation were the modified Clausius equation of state with a single set of parameters established at the temperature of interest, and a minimum of one observation of a binary mixture property. These parameters were determined from vapor pressure and saturated molar volumes of liquid and vapor phase of pure substances.

The applicability of the proposed method was tested on binary and ternary systems. It was also compared with the results obtained from the modified R-K equation of state as well as the experimental values reported in the literature. Results of the comparison showed that the proposed method is comparable to the modified R-K equation. A summary of the comparison, in terms of average absolute deviations for ΔP and Δy for the arbitrarily chosen systems is given in Table 4-3.

The absolute deviation for ΔP (at $x_1 = 1.0$ and $x_1 = 0.00$) were found not equal to zero as expected. This can be attributed to the difference between the vapor pressure values of the pure components reported with the volumetric data used in the evaluation of the parameters and those reported with the experimental phase equilibrium data at the same temperature.

The calculated results for the systems nitrogen-methane and argon-methane indicate the highest deviation in Δy_2 (3.6 and 5.01 respectively). This is partly due to the low relative volatility of methane. When the difference between the calculated and experimental y_2 is expressed in terms of mole fractions, the average absolute deviations are only 0.004 and 0.005 respectively.

The binary interaction constant k_{ij} was involved in the calculations. It was considered to be independent of temperature, pressure and composition. As has been demonstrated in the present work, the method based on this assumption yields satisfactory results.

However, from the present work, it has been found that the binary interaction constants, k_{ij}'s, for the system carbon dioxide-hydrogen sulfide are different at different isotherms. Since there is no definite trend indicated, it is difficult to make any conclusions at this moment. In addition, these changes in the values of k_{ij} with temperature may be due to the uncertainty of the data for this system.

The numerical values obtained from phase equilibrium data are found to be close to those calculated from the ionization potential of pure substances as suggested by Hiza and Duncan(90).

TABLE 4-3

A Summary of the Comparison Between the Calculated and Experimental Results Using Clausius and R-K Eq. of State

System	Modified Clausius Average Abs. Dev. %				Modified R - K An. Ab. Dev. %			
	ΔP	Δy_1	Δy_2	Δy_3	ΔP	Δy_1	Δy_2	Δy_3
Nitrogen-argon	0.88	0.73	1.86		0.65	0.37	1.65	
Nitrogen-oxygen	0.58	1.39	1.65		1.12	1.34	1.30	
Argon-oxygen	0.20	0.30	0.22		0.30	0.23	0.23	
Nitrogen-methane	2.30	0.54	3.60		2.3	0.64	4.88	
Argon-methane	1.1	0.6	5.01		1.3	0.70	4.99	
Carbon dioxide-hydrogen sulfide	2.98	2.31	2.4		2.97	3.06	2.50	
Nitrogen-argon-oxygen (N)	0.9	1.33	2.06	0.54	0.6	1.16	2.11	0.45
Nitrogen-argon-oxygen (W)	1.80	1.27	1.34	1.12	1.59	1.14	1.9	0.59
Propane-carbon dioxide	1.75	5.03	1.37		1.85	5.54	1.6	
Hydrogen sulfide-n-butane	3.41	3.22	4.67		2.09	3.60	6.36	

In conclusion, it may be stated that the satisfactory results obtained in this investigation indicate the applicability of the proposed procedure for calculating phase equilibrium by means of the modified Clausius equation of state.

CHAPTER VI

MISCELLANEOUS WORKS

5.1 CALCULATION OF PHASE EQUILIBRIUM DATA FROM TOTAL PRESSURE-LIQUID PHASE COMPOSITION DATA

The usual technique for obtaining vapor-liquid equilibrium data is by direct measurement ie., equilibrium is established and phases are analysed. The experimental technique used in such experiments must be delicate and accurate in order to ensure meaningful results. This fact, coupled with the necessity for much analytical work tends to enhance interest in methods for determination of equilibrium data that do not involve sampling and analysis of phases.

Ljunglin and VanNess[91] have suggested the classification of these methods into two groups, direct and indirect methods. The direct methods [92, 93] involve calculation of vapor phase composition by integration of the coexistence equation, a first order differential equation derived from the Gibbs-Duhem[1, 94-103] equation relating phases. The indirect methods involve first the calculation, by some appropriate form of the integrated Gibbs-Duhem equation, of the liquid phase activity coefficients and subsequent calculation of vapor phase therefrom. These methods usually involve ascertaining of which selected solution equations to the Gibbs-Duhem equation lead to the best fit of the experimental vapor pressure data, and of the determination of the parametric values producing the best fit, Barker[94], for example has developed a procedure based on the assumption that the excess Gibbs free energy can be represented as a polynomial function of composition.

There is a basic difference in the degree of rigor associated with the direct and the indirect methods. In the former, one makes no assumptions about the solution behavior. Solution behavior is determined directly from the experimental vapor pressure data.

The indirect method necessitates the assumption of a particular model and the estimation of its parameters. This deficiency in the indirect methods has been recognized by Tao(103) who has presented another indirect method in which the necessity for a priori assumption of a particular functional form for the excess Gibbs free energy has been removed. Tao's procedure involves calculation of the activity coefficients essentially by integration of an equation resembling the coexistence equation. His procedure, though indirect, retains the rigor usually associated with direct method.

Virtually all of the methods for calculating vapor phase compositions from total pressure measurements have been applied mostly to binary systems. If one considers the application of the direct method to ternary systems, one finds that the problem increases in complexity from the solution of one first-order differential equation to the simultaneous solution of two first-order partial differential equations. Not only are two independent variables involved (the two independent liquid phase compositions) but also two dependent variables (the two independent vapor phase compositions).

The work in this chapter presents the modification and extension of an indirect method, previously proposed by Ho et al.(1), to evaluate the liquid phase compositions for ternary systems at moderate pressures and binary systems at high pressures.

5.1.1 EVALUATION OF TERNARY VAPOR PHASE COMPOSITION FROM P-T-x DATA FOR SYSTEMS AT MODERATE PRESSURES

When the components of a ternary solution vary along a constant

x_1/x_2 secant, Figure (5.1), the mole fractions of components 1 and 2 depend on that of component 3 as follows:

$$x_1 = x' (1 - x_3) \tag{5-1. a}$$

$$x_2 = (1 - x')(1 - x_3) \tag{5-1. b}$$

where x' represents the mole fraction of component 1 in the initial binary solution 1-2. The mole fraction of the third component along the secant, is taken as the independent variable, and the ternary mixture may therefore be regarded as a binary one. Assuming that the vapor phase is an ideal solution, but not an ideal gas, the total pressure of the system may be represented by the following equation:

$$P = \frac{x_3 P^\circ{}_3 \gamma_3}{K_3} + \frac{(1-x_3) P^\circ{}_{12} \gamma_{12}}{K_{12}} \tag{5-2}$$

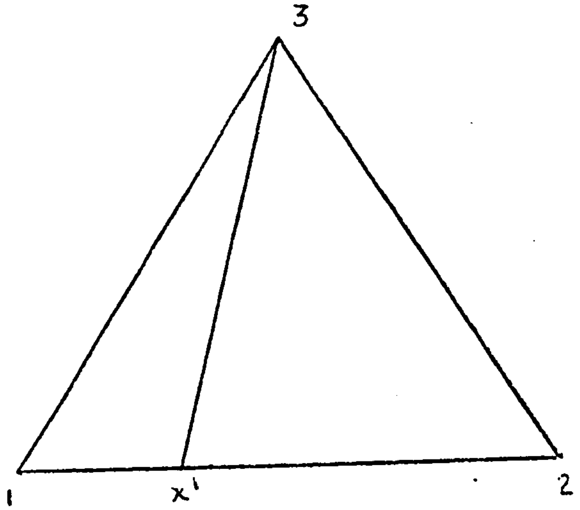


Figure 5-1

where

$$K_3 = \exp\left[\frac{(\beta_{33} - v_3)(P - P^\circ_3)}{RT}\right]$$

$$K_{12} = \exp\left[\frac{(\beta_{12} - v_{12})(P - P^\circ_{12})}{RT}\right]$$

In this investigation liquid phase activity coefficients γ, were correlated by means of the Redlich-Kister equation as follows:

$$\ln \gamma_3 = (1 - x_3)^2 \left[B + C(4x_3 - 1) + D(2x_3 - 1)(6x_3 - 1) \right] \qquad (5\text{-}3.\ a)$$

$$\ln \gamma_{12} = x_3^2 \left[B + C(4x_3 - 3) + D(2x_3 - 1)(6x_3 - 5) \right] \qquad (5\text{-}3.\ b)$$

Equations (5-2) and (5-3) indicate that the total pressure is a function of the Redlich-Kister constants and the liquid composition x_3. Hence at constant temperature,

$$P = P(B, C, D, x_3) \qquad (5\text{-}4)$$

Assuming a set of initial values for the constants B, C and D, then

$$\delta P = \frac{\partial P}{\partial B}\,\delta B + \frac{\partial P}{\partial C}\,\delta C + \frac{\partial P}{\partial D}\,\delta D \qquad (5\text{-}5)$$

where

$$\delta P = P_{expt} - P_{calc} \qquad (5\text{-}6)$$

and

$$\frac{\partial P}{\partial B} = \frac{\partial P}{\partial \gamma_3}\,\frac{\partial \gamma_3}{\partial B} + \frac{\partial P}{\partial \gamma_{12}}\,\frac{\partial \gamma_{12}}{\partial B}\,. \qquad (5\text{-}7)$$

Similar equations may be written for $\frac{\partial P}{\partial C}$ and $\frac{\partial P}{\partial D}$. Furthermore from equations (5-3. a) and (5-3. b)

$$\frac{\partial \gamma_3}{\partial B} = \gamma_3 (1 - x_3)^2 \qquad (5\text{-}8.\,a)$$

$$\frac{\partial \gamma_{12}}{\partial B} = \gamma_{12}\, x^2_3 \qquad (5\text{-}8.\,b)$$

and

$$\frac{\partial P}{\partial \gamma_3} = \frac{x_3\, P^\circ_3}{B_3} \qquad (5\text{-}9.\,a)$$

$$\frac{\partial P}{\partial \gamma_{12}} = \frac{(1 - x_3)\, P^\circ_{12}}{B_{12}} \qquad (5\text{-}9.\,b)$$

By substituting equations (5-8) and (5-9) into equation (5-7), the following expression is obtained

$$\frac{\partial P}{\partial B} = \frac{x_3\,(1-x_3)^2\;\gamma_3\, P^\circ_3}{K_3} + \frac{x^2_3\,(1-x_3)\,\gamma_{12}\, P^\circ_{12}}{K_{12}} \qquad (5\text{-}10)$$

Similar expressions for $\frac{\partial P}{\partial C}$ and $\frac{\partial P}{\partial D}$ may be obtained in the same manner.

Substituting $\frac{\partial P}{\partial B}$, $\frac{\partial P}{\partial C}$ and $\frac{\partial P}{\partial D}$ into equation (5-5) for all the experimental points give the following set of normal equations:

$$\delta B \sum_{i}^{n} \left(\frac{\partial P}{\partial B}\right)^2 + \delta C \sum_{i=1}^{n} \left(\frac{\partial P}{\partial C}\right)\left(\frac{\partial P}{\partial B}\right) + \delta D \sum_{i=1}^{n} \left(\frac{\partial P}{\partial D}\right)\left(\frac{\partial P}{\partial B}\right) = \sum_{i=1}^{n} \delta P \left(\frac{\partial P}{\partial B}\right) \qquad (5\text{-}11)$$

$$\delta B \sum_{i=1}^{n} \left(\frac{\partial P}{\partial B}\right)\left(\frac{\partial P}{\partial C}\right) + \delta C \sum \left(\frac{\partial P}{\partial C}\right) + \delta D \sum_{i=1}^{n} \left(\frac{\partial P}{\partial D}\right)\left(\frac{\partial P}{\partial C}\right) = \sum_{i=1}^{n} \delta P \left(\frac{\partial P}{\partial C}\right) \qquad (5\text{-}12)$$

and

$$\delta B \sum_{i=1}^{n} \left(\frac{\partial P}{\partial B}\right)\left(\frac{\partial P}{\partial D}\right) + \delta C \sum_{i=1}^{n} \left(\frac{\partial P}{\partial C}\right)\left(\frac{\partial P}{\partial D}\right) + \delta D \sum_{i=1}^{n} \delta P \left(\frac{\partial P}{\partial D}\right)^{2} = \sum_{i=1}^{n} \delta P \left(\frac{\partial P}{\partial D}\right) \quad (5\text{-}13)$$

These corrections for B, C and D are employed to calculate the actual values of B, C and D which in turn are used to evaluate the liquid phase activity coefficients, γ_3 and γ_{12}. Using the liquid phase activity coefficients, equation (5-14) may be applied to evaluate the vapor phase compositions.

$$y_i = \frac{(x_i P_i^\circ \gamma_i)/K_i}{[x_i P_i^\circ \gamma_i)/K_i] + [(x_{jk} P^\circ_{jk} \gamma_{jk})/K_{jk}]} \quad (5\text{-}14)$$

5.1.2 RESULTS AND DISCUSSIONS

The experimental equilibrium temperature, pressure and liquid composition, determined earlier in Chapter III for the binary system argon-ethane and the ternary system argon-methane-ethane, were utilized in this chapter. In the binary system argon-ethane, the concentration of ethane in the vapor phase was found to be extremely low, and the error in its determination was rather high. For this reason, it was decided to evaluate the ethane composition of the vapor from the total vapor pressure-liquid composition measurements. At 115.5°K the system pressure is relatively low, and the total pressure method previously proposed$^{(1)}$ was employed for this purpose. The calculated values of liquid phase activity coefficient, γ, and the vapor phase composition, y, are listed in Table 5-1. In Figure 5-2, ln γ values were plotted against liquid composition, x, to illustrate the non-ideal behavior of the binary system.

TABLE 5-1

Experimental and Calculated Results for the Binary System Argon - Ethane at 115.5°K

P lb in^{-2} (1 lb in^{-2} 6.9 kN m^{-2}	x_A	$(y_A)_{calc}$	γ_A	γ_{C_2}
24.6	0.0690	0.9990	2.7434	1.0421
25.2	0.0710	0.9990	2.7343	1.0419
25.2	0.0712	0.9990	2.7334	1.0424
29.7	0.0926	0.9992	2.6368	1.0469
29.7	0.0928	0.9992	2.6362	1.0476
30.8	0.0980	0.9993	2.6127	1.0475
33.8	0.1119	0.9993	2.5550	1.0513
34.1	0.1135	0.9993	2.5480	1.0509
36.0	0.1222	0.9994	2.5131	1.0529
41.2	0.1450	0.9995	2.4247	1.0589
43.4	0.1546	0.9995	2.3891	1.0619
48.4	0.1760	0.9995	2.3129	1.0691
51.0	0.1873	0.9995	2.2749	1.0735
57.8	0.2176	0.9996	2.1770	1.0859
63.9	0.2470	0.9996	2.0886	1.1006
70.7	0.2835	0.9997	1.9867	1.1216
80.1	0.3422	0.9997	1.8387	1.1635
81.8	0.3540	0.9997	1.8096	1.1775
87.2	0.3946	0.9997	1.7175	1.2109
87.4	0.3969	0.9997	1.7125	1.2131
90.9	0.4259	0.9997	1.6509	1.2436
94.3	0.4577	0.9997	1.5866	1.2853
98.6	0.5046	0.9998	1.4980	1.3587
100.9	0.5355	0.9998	1.4432	1.4110
105.5	0.6120	0.9998	1.3200	1.5923
108.1	0.6638	0.9998	1.2468	1.7624
109.8	0.6978	0.9998	1.2033	1.9054
110.8	0.7304	0.9998	1.1649	2.0600
113.0	0.7581	0.9998	1.1351	2.2326
115.2	0.8004	0.9998	1.0946	2.5291
116.2	0.8210	0.9998	1.0772	2.7114
116.3	0.8230	0.9998	1.0757	2.7267
118.5	0.8619	0.9999	1.0474	3.1422
123.1	0.9151	0.9999	1.0187	3.9233
124.5	0.9265	0.9999	1.0142	4.1604
129.1	0.9570	0.9999	1.0050	4.8089
130.5	0.9654	0.9999	1.0032	4.9761
133.1	0.9813	1.0000	1.0009	5.4854

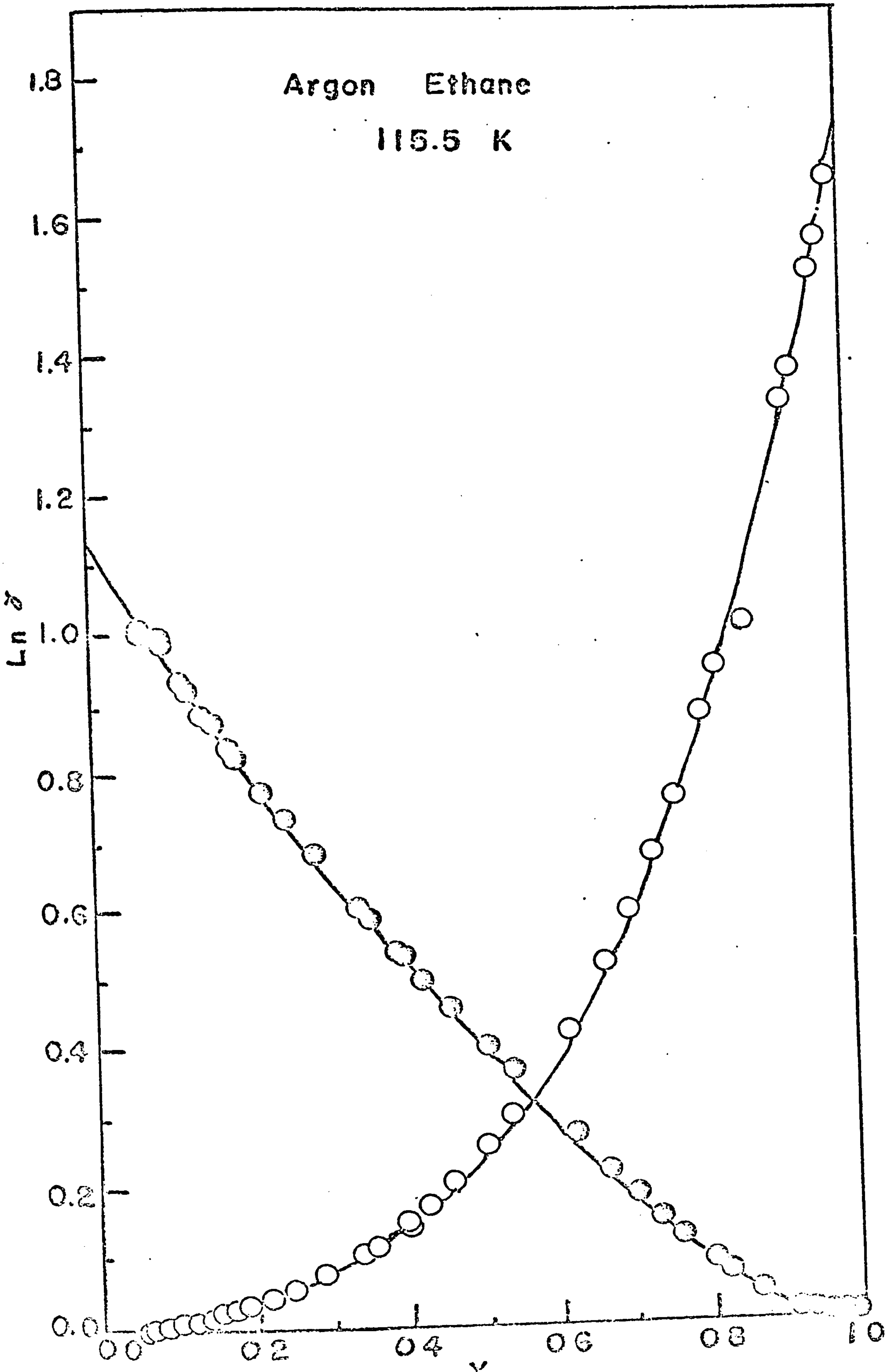


Figure 5-2 Liquid Phase Activity Coefficient-Composition Curve for the Binary Argon-Ethane at 115.5°K

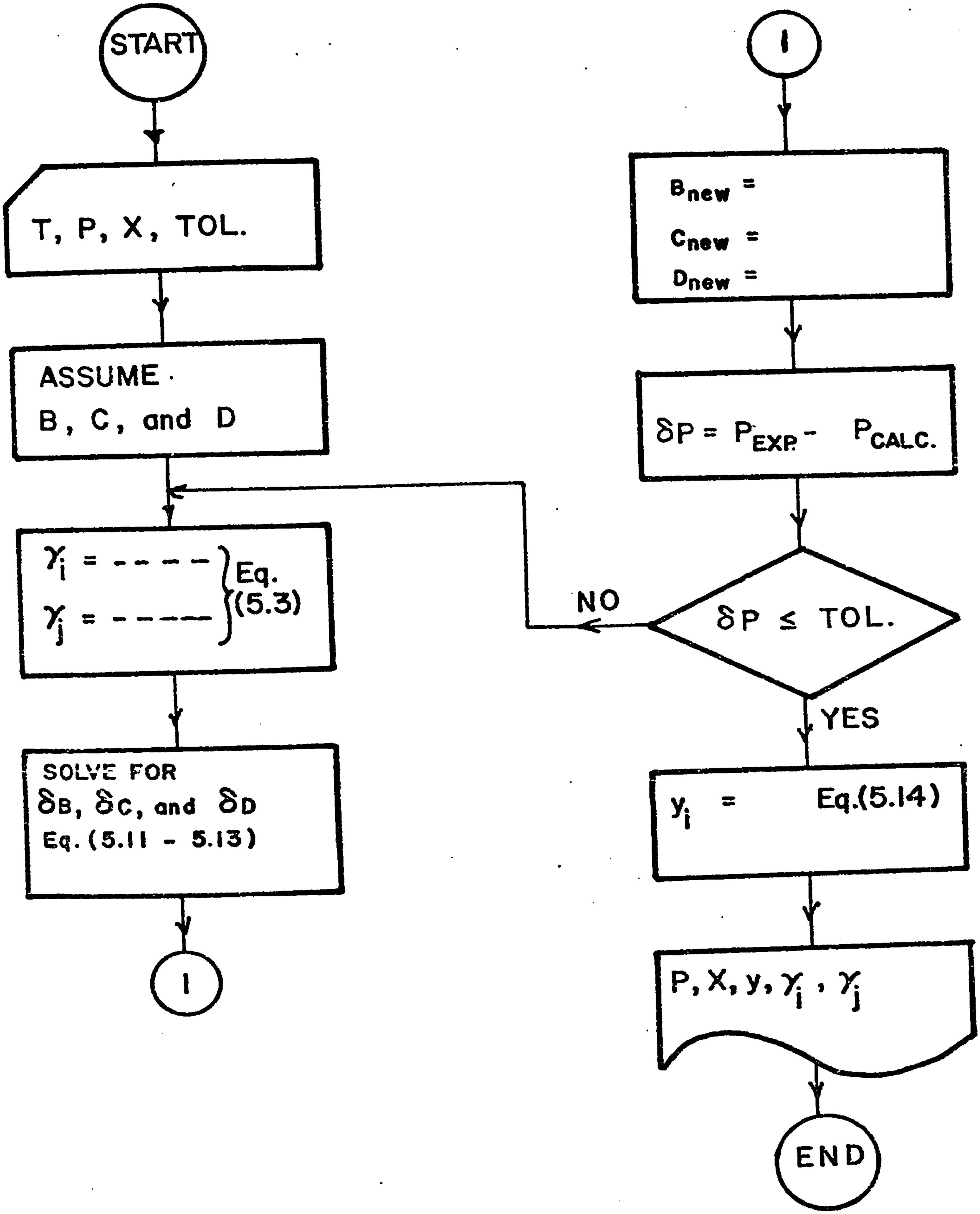


Figure 5-3 Schematic Diagram of Vapor-Phase Calculation

The experimental points for the ternary system argon-methane-ethane were determined along constant x_{C_1}/x_{C_2} and constant x_A/x_{C_2} secants. This arrangement was planned for the purpose of reducing the ternary system to a hypothetical binary system so that the total pressure method proposed for binary systems$^{(1)}$ could be extended and applied.

The modified total pressure method was used to predict the vapor phase composition of the ternary system argon-methane-ethane.

The numerical routine for determining the vapor phase composition is shown in Figure 5-3 and may be summarized as follows:

1. Initial values of B, C and D were assumed.
2. Liquid phase activity coefficients values were calculated from Equation (5-3. a) and (6-3. b).
3. Total pressure of the system was calculated at each experimental point, using the pure components properties. Second virial coefficients values for pure components and the hypothetical binaries were evaluated using the method of O'Connell and Prausnitz$^{(104)}$.
4. δB, δC, δD were evaluated from Equations (5-11) - (5-13).
5. New values of B, C and D were obtained.
6. The process was repeated until the tolerance of pressure was satisfied.

This procedure was first applied to the secants of constant x_{C_1}/x_{C_2} for the evaluation of y_A and then applied to the secants of constant x_A/x_{C_2}, which were fixed in such a manner that they intercept the secants of constant x_{C_1}/x_{C_2} at each of the experimental points, for the evaluation of y_{C_1}. The vapor phase composition of the least volatile component, y_{C_2} was determined by Equation (5-15).

TABLE 5-2

Experimental and Calculated Results for the Ternary System Argon-Methane-Ethane at 115.5°K

P lb in^{-2} (1 lb in^{-2} 6.9 kN m^{-2})	x_A	x_{C_1}	y_A Expt	y_A Calc	y_{C_1} Expt	y_{C_1} Calc	y_{C_2} Calc	γ_A	γ_{C_1}	γ_{C_2}
59.8	0.2572	0.1859	0.9164	0.9158	0.0836	0.0840	0.0002	3.5694	2.5350	1.5119
71.1	0.3241	0.1743	0.9332	0.9338	0.0668	0.0660	0.0002	3.0153	2.1964	1.7420
80.0	0.4177	0.1521	0.9525	0.9518	0.0475	0.0480	0.0002	2.4247	1.8479	2.2033
89.5	0.5148	0.1271	0.9646	0.9645	0.0354	0.0353	0.0002	1.9559	1.5827	3.0046
94.4	0.5889	0.1079	0.9761	0.9717	0.0279	0.0280	0.0003	1.6738	1.4331	4.0395
98.5	0.6553	0.0980	0.9762	0.9751	0.0238	0.0245	0.0003	1.4469	1.3198	5.7975
51.0	0.1977	0.3081	0.8400	0.8395	0.1600	0.1603	0.0003	3.0720	2.1149	1.7569
64.6	0.3046	0.3066	0.8852	0.8839	0.1148	0.1158	0.0003	2.2713	1.6439	2.4917
70.7	0.3547	0.2737	0.9056	0.9059	0.0944	0.0938	0.0003	2.1390	1.5893	2.6935
79.1	0.4521	0.2351	0.9300	0.9302	0.0700	0.0695	0.0003	1.7973	1.4205	3.5983
84.4	0.4925	0.2187	0.9377	0.9378	0.0623	0.0619	0.0003	1.6778	1.3654	4.1301
90.4	0.5571	0.1887	0.9489	0.9492	0.0511	0.0505	0.0003	1.5214	1.3017	5.1739
99.5	0.6625	0.1481	0.9617	0.9620	0.0383	0.0376	0.0004	1.2942	1.2233	8.4816
107.5	0.7564	0.1071	0.9730	0.9726	0.0270	0.0269	0.0005	1.1556	1.2113	4.0292
51.8	0.2810	0.5164	0.8018	0.8026	0.1982	0.1970	0.0005	1.5470	1.1582	5.9968
59.8	0.3398	0.4746	0.8359	0.8363	0.1641	0.1632	0.0005	1.4628	1.1386	6.8501
69.5	0.4198	0.4143	0.8715	0.8716	0.1285	0.1279	0.0005	1.3673	1.1247	8.1797
79.5	0.5083	0.3551	0.8984	0.8984	0.1016	0.1010	0.0005	1.2604	1.1136	10.7773
89.2	0.6135	0.2803	0.9242	0.9244	0.0758	0.0751	0.0005	1.1625	1.1264	15.2237
98.0	0.6924	0.2044	0.9469	0.9465	0.0531	0.0580	0.0005	1.1288	1.1597	17.1085
78.3	0.4431	0.2317	0.9309	0.9304	0.0691	0.0693	0.0003	1.1855	1.4547	3.3821
81.0	0.4606	0.2096	0.9380	0.9383	0.0620	0.0614	0.0003	1.8630	1.4730	3.3345
84.0	0.4854	0.1675	0.4510	0.9519	0.0490	0.0478	0.0002	1.9237	1.5354	3.1154

Root mean square deviation in vapor phase compositions of argon and methane = 0.002

$$y_{C_2} = 1 - (y_A + y_{C_1}) \qquad (5\text{-}15)$$

An IBM 360 computer program was written to perform the above calculations. The required binary equilibrium data of the system methane-ethane were interpolated from those of Chang Lu and Canfield(46, 105). The calculated results for the ternary system are given in Table 5-2. The root mean square deviation, which is defined by

$$\Delta = \left(\frac{\sum_{i=1}^{N} (y_{expt} - y_{calc})}{N} \right)^{0.5} \qquad (5\text{-}16)$$

is only 0.002 in the vapor phase compositions of argon and methane, indicating the good agreement obtained between the experimental and calculated results, and the validity of the proposed calculation procedure.

5.2 EVALUATION AND CORRELATION OF VAPOR-LIQUID EQUILIBRIUM DATA FOR AIR COMPONENTS MIXTURES

In the low temperature separation of air constituents, vapor-liquid equilibria for mixtures of nitrogen, argon and oxygen play an important role in the design of the fractionating columns. Although equilibrium data for the binary and the ternary systems have been previously investigated experimentally, the agreement between the numerous set of data is not always good. Latimer(107) in 1957, correlated the liquid activity coefficient data by means of the regular solution equations over the pressure range from 0.2 to 20 atmospheres, using published binary and ternary experimental values together with the plant performance data. Wilson(83) in 1964 made a literature survey for all existing data relating to vapor-liquid equilibrium and enthalpy of the nitrogen-argon-oxygen system up to 1963, and experimentally determined 1962 data points over the pressure range from 1 to 26 atmospheres. The liquid activity coefficient data calculated from the experimental data were correlated by means of the Van Laar equation. Since the publication of Wilson's data, additional experimental investigations have been reported in the literature. In this investigation, binary vapor-liquid equilibrium data compiled from different sources over the temperature range from 90° to 120°K, were first tested for their consistency, then correlated by means of the Redlich-Kister equation. In order to avoid the uncertainty due to the temperature effect on liquid activity coefficients, only isothermal data were tested for thermodynamic consistency. The binary constants obtained in this manner were then used to evaluate the ternary liquid activity coefficients for the purpose of determining whether ternary constants are required in the correlation.

5.2.1 EVALUATION OF LIQUID PHASE ACTIVITY COEFFICIENTS

In the thermodynamic treatment of the data, the constant-pressure constant-temperature liquid activity coefficient is defined by

$$\gamma_i(P^\circ, T, x) = \frac{f_{iL}(P^\circ, T, x)}{x_i \, f^\circ_{iL}(P^\circ, T)} \exp \int_P^{P^\circ} \frac{\overline{V}_{iL}}{RT} \, dP \qquad (5\text{-}17)$$

and

$$\gamma_i \rightarrow 1 \quad \text{as} \quad x_i \rightarrow 1 \qquad (5\text{-}18)$$

In Equation (5-17), $f_{iL}(P, T, x)$ represents the fugacity of component i in the liquid mixture at system temperature, T, and pressure, P and $f^\circ_{iL}(P^\circ, T)$ represents the fugacity of pure liquid i at the system temperature, T, and the reference pressure P°. In this investigation, P°, was arbitrarily taken to be 500 psia, which is above the highest saturation pressure encountered in this work, hence the hypothetical liquid state was avoided. The exponential term represents the pressure correction of γ from P to P°. The fugacities and the partial molal volumes were obtained from the Redlich-Kwong equation of state by means of a modified procedure(41). In the equation

$$P = \frac{RT}{v-b} - \frac{a}{T^{0.5}\, v(v+b)} \tag{5-19}$$

the parameters Ω_a and Ω_b, which are related to the quantities a and b as follows

$$a = \Omega_a R^2 Tc^{2.5}/Pc \tag{5-20}$$

$$b = \Omega_b R\, Tc/Pc \tag{5-21}$$

were considered temperature-dependent. The parameters were evaluated using molal volumes and vapor pressures of pure saturated liquids. This modification has been recently presented(41). Values of Ω_a and Ω_b used in this work are listed in Table A-I. 29. The mixing rules employed in this investigation are identical to those reported earlier(41). Values of the binary interaction constant k_{ij}, which represents the deviation from the geometric mean of the critical temperatures as defined by Chueh and Prausnitz(86), were obtained from the vapor-liquid equilibrium data of the binary systems.

5.2.2 CONSISTENCY TEST

Thermodynamic consistency of binary isothermal data were tested by means of the Redlich-Kister relationship

$$\int_{x_1=0}^{x_1=1} \ln \frac{\gamma_1}{\gamma_2} \, dx_1 = 0 \tag{5-22}$$

in which γ values were those obtained from Equation (5-17) at constant T and P°.

For ternary isothermal data the method proposed by Chang and Lu(108) was employed. A brief description of the method is presented below.

At constant temperature and pressure, the Gibbs-Duhem equation is:

$$\sum_{i=1}^{n} x_i \, d \ln \gamma_i = 0 \tag{5-23}$$

Integrating from point a to its adjacent point b by the trapezoidal rule gives

$$\sum_{i=1}^{n} (x_{ia} + x_{ib}) (\ln \gamma_{ib} - \ln \gamma_{ia}) = 0 \tag{5-24}$$

The assumption involved here is that all the $\ln \gamma$ vs. x curves can be approximated by segments of straight lines connecting the points. This approach is identical to that employed by Li and Lu(109) and by McDermott and Ellis(110). At equilibrium

$$\hat{f}_{iL} = \hat{f}_{iV}$$

and in the vapor phase

$$\hat{f}_{iV} = y_i \hat{\phi}_i P$$

Hence

$$\gamma_i(P^\circ, T, x) = \frac{y_i \hat{\varphi}_i P}{x_i f^\circ_{iL}} \exp \int_{P}^{P^\circ} \frac{\bar{V}_{iL}}{RT} dP \qquad (5\text{-}25)$$

Substituting Equation (5-25) into Equation (5-24)

$$\sum_{i=1}^{n} (x_{ib} + x_{ia}) \ln \frac{K_{ib}}{K_{ia}} = \sum_{i=1}^{n} (x_{ib} + x_{ia}) \ln \frac{\hat{\varphi}_{ia} P_a}{\hat{\varphi}_{ib} P_b}$$

$$+ \sum_{i=1}^{n} (x_{ib} + x_{ia}) \int_{P_a}^{P_b} \frac{\bar{V}_{iL}}{RT} dP \qquad (5\text{-}26)$$

in which $K_i = y_i / x_i$. The left-hand side of Equation (5-26) contains only the experimental equilibrium data while the right-hand side contains the quantities which can be evaluated from the Redlich-Kwong equation of state as modified above. In order to make a test conclusive, maximum experimental error bounds must be established. For isothermal data, the overall deviation resulting from experimental errors of the pair tested may be obtained by rearranging Equation (5-26) as follows

$$\Delta = \sum_{i=1}^{n} (x_{ib} + x_{ia}) \left[\ln \frac{K_{ib}}{K_{ia}} - \ln \frac{\hat{\varphi}_{ia} P_a}{\hat{\varphi}_{ib} P_b} - \int_{P_a}^{P_b} \frac{\bar{V}_{iL}}{RT} dP \right] \qquad (5\text{-}27)$$

Differentiating Equation (5-27) and replacing dP, dT and dx by the measuring errors, E(P), E(T) and E(x) respectively, the maximum experimental error bounds are established as follows

$$
\begin{aligned}
|D|_{max} = & \sum_{i=1}^{n} (x_{ib} + x_{ia}) \left[\left(\frac{1}{y_{ib}} + \frac{1}{y_{ia}} + \frac{1}{x_{ib}} + \frac{1}{x_{ia}} \right) \right. \\
& \left. + 2 \left| \left(\ln \frac{K_{ib}}{K_{ia}} - \ln \frac{\hat{\phi}_{ia} P_a}{\hat{\phi}_{ib} P_b} - \int_{P_a}^{P_b} \frac{\bar{V}_{iL}}{RT} dP \right) \right| \right] E(x) \\
& + \sum_{i=1}^{n} (x_{ib} + x_{ia}) \left(\frac{1}{P_a} + \frac{1}{P_b} + \left| \frac{\partial}{\partial P} \int_{P_a}^{P_b} \frac{\bar{V}_{iL}\, dP}{RT} \right| \right) E(P) \\
& + \sum_{i=1}^{n} (x_{ib} + x_{ia}) \left| \left(\frac{\partial}{\partial T} \int_{P_a}^{P_b} \frac{\bar{V}_{iL}\, dP}{RT} \right) \right| E(T) \qquad (5\text{-}28)
\end{aligned}
$$

where

$E(x)$ = error in composition x or y measurement
$E(P)$ = error in pressure measurement
$E(T)$ = error in temperature measurement

Experimental data which are widely separated are arranged along any arbitrary path, and tested in pairs by means of Equation (5-27) and (5-28). Each experimental point appears twice in the test, with the exceptions of the first and the last points of the chosen path. An experimental point is considered inconsistent if Δ is greater than $|D|_{max}$. In doing so, a point-by-point evaluation of the data is achieved.

5.2.3 RESULTS AND DISCUSSIONS

Nitrogen-Argon Mixtures

Most of the equilibrium data reported in the literature for the

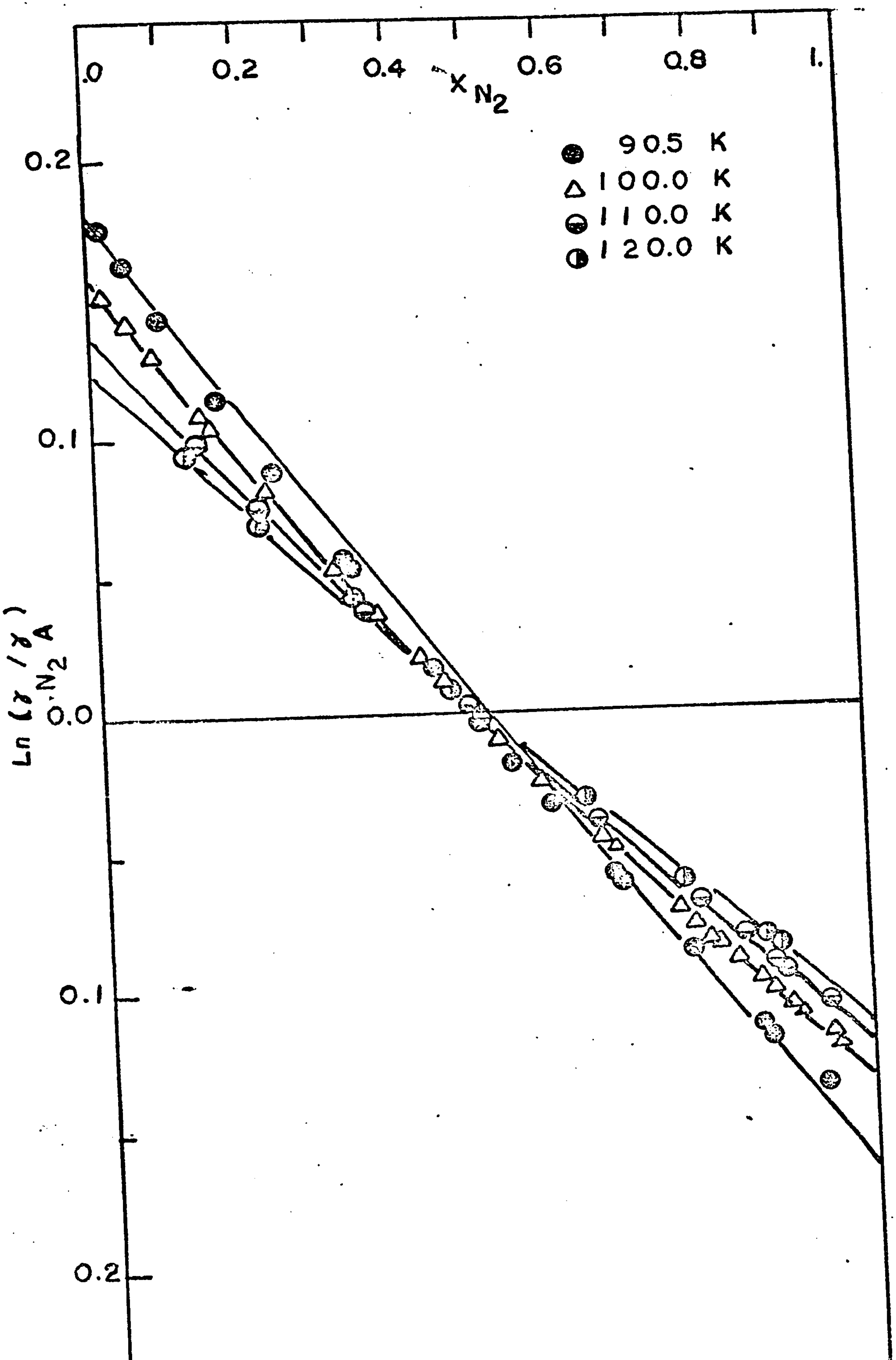


Figure 5-4 Area Test of Narinskii's Data for Nitrogen-Argon System

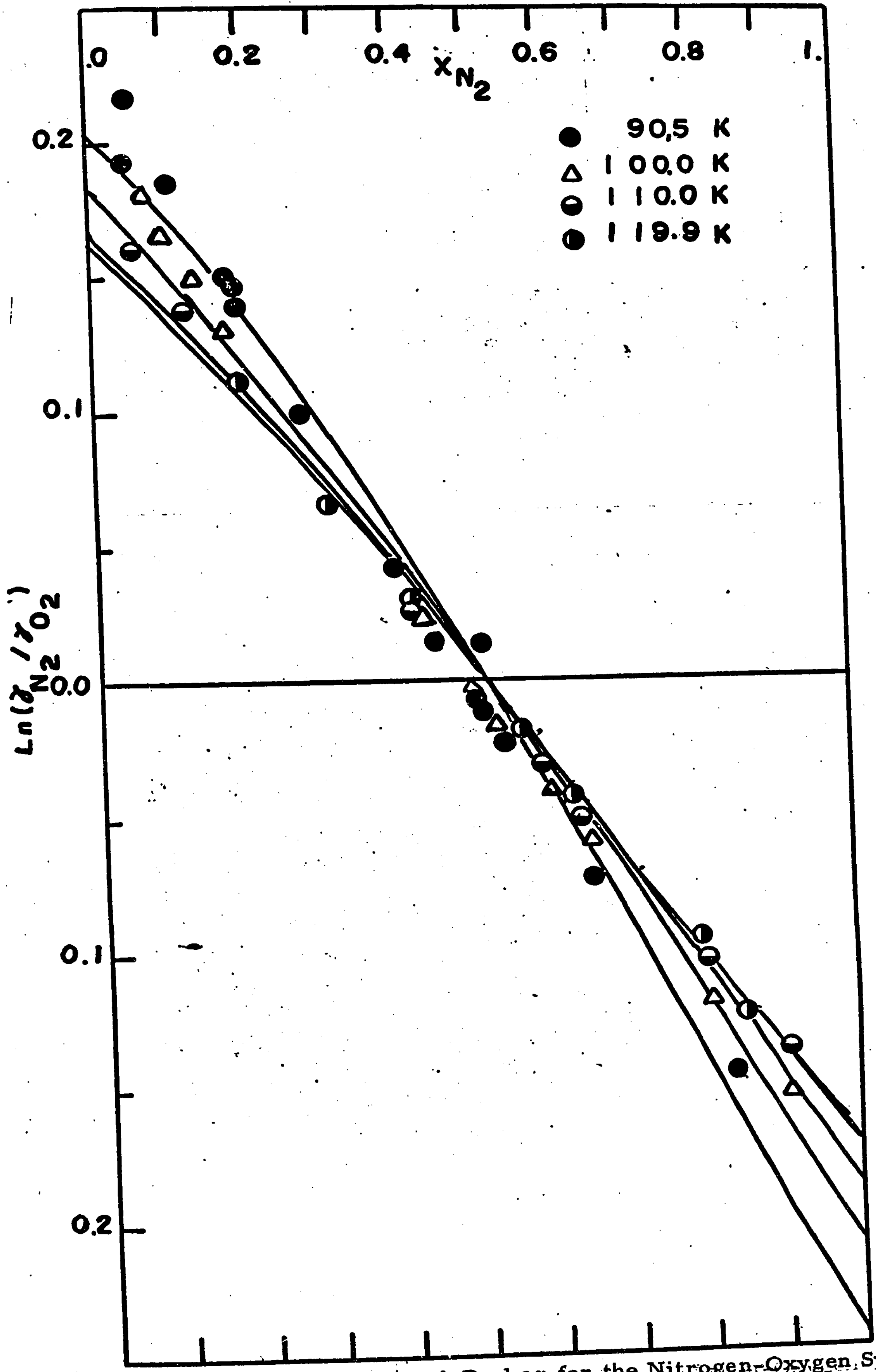


Figure 5-5 Area Test of the Data of Dodge & Dunbar for the Nitrogen-Oxygen System

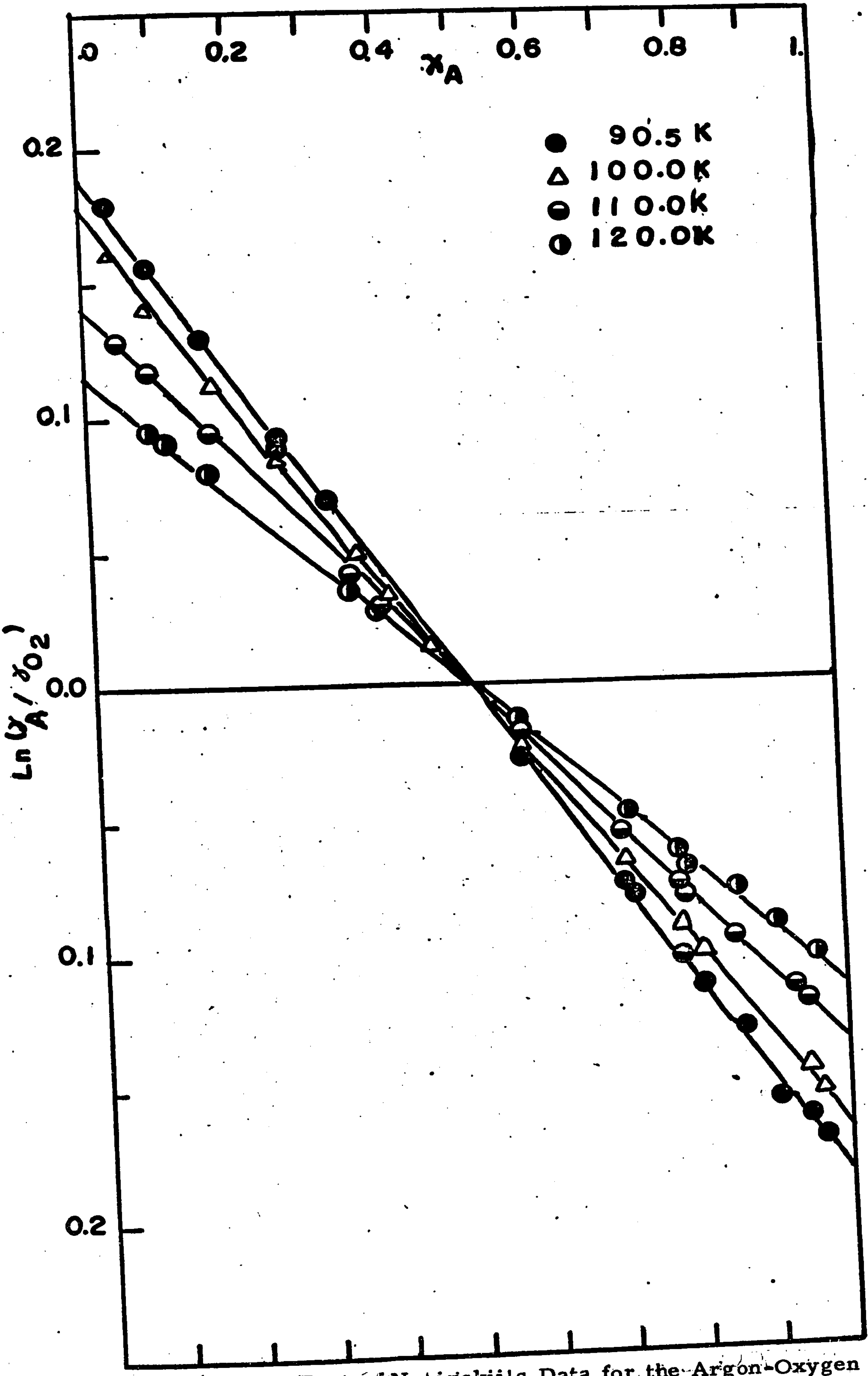


Figure 5-6 Area Test of Natiriskii's Data for the Argon-Oxygen System

binary system nitrogen-argon were restricted to the low temperature region. Since the publication of Wilson's data, additional experimental values were reported by Narinskii(75) and by Thorpe(111). A static method was used by Thorpe to determine vapor pressures and compositions of this system at pressures up to 10 atm. Unfortunately, isothermal data interpolated from his results did not cover the complete concentration range. On the other hand, the experimental results of Narinskii were reported over the complete concentration range at four isothermal conditions, namely 90.5°, 100°, 110° and 120°K. Liquid activity coefficient values obtained from Equation (5-17) for Narinskii's data satisfy the area-test as shown in Figure (5-4).

Argon-Oxygen Mixtures

For the binary system argon-oxygen, available literature data include those of Brube and Ishkin(112), Sagenkahn and Fink(113), Clark et al.(114), Fastovski and Petrovski(115, 116), Narinskii(82) and Burn and Din(117). Results of the three investigations(82, 114, 117) which cover the complete concentration range were tested for their thermodynamic consistency by means of the area-test method. The results of the test indicate that the data of Clark et al.(114) are inconsistent. Burn and Din(117) studied this system over the complete concentration range and over a pressure range of 1 to 10 atm. using a static method, and stated that their data agree well with the isothermal data of Narinskii(82) who reported the data of 90.5°, 100°, 110°, and 120°K. The results of the area-test as shown in Figure (5-5) indicate that the data of Narinskii are thermodynamically consistent.

Nitrogen-Oxygen Mixtures

In the temperature range of this investigation, literature data for the binary system nitrogen-oxygen system include those of Dodge and Dunbar(44) and Din(55).

TABLE 5-3

Values of Binary Redlich-Kister Constants

(Ref. Pressure = 500 psia)

System	T, °K	B_{ij}	C_{ij}	$\ln \gamma_a^\infty$	$\ln \gamma_b^\infty$
Nitrogen-Argon	90.5	0.1655	-0.0144	0.1829	0.1541
(k_{12} = 0.0)	100.	0.1459	-0.0155	0.1574	0.1344
	110.	0.1295	-0.0080	0.1375	0.1215
	120.	0.1266	-0.0009	0.1275	0.1257
Argon-Oxygen	90.5	0.1878	-0.0027	0.1905	0.1851
(k_{23} = 0.016)	100.	0.1681	-0.0027	0.1708	0.1654
	110.	0.1385	-0.0026	0.1411	0.1359
	120	0.1139	-0.0025	0.1164	0.1114
Nitrogen-Oxygen	90.5	0.2267	0.0198	0.2069	0.2965
(k_{13} = 0.001)	99.94	0.1988	0.0155	0.1833	0.2143
	110.05	0.1787	0.0113	0.1674	0.1900
	119.92	0.1715	0.0046	0.1669	0.1761

Results of the area-test indicate that the isothermal data of Din are thermodynamically inconsistent, while the isothermal data of Dodge and Dunbar satisfy the test. The area-test of the data of Dodge and Dunbar(44) is shown in Figure (5-6).

Correlation of Binary Liquid Activity Coefficients

The thermodynamically consistent liquid activity coefficients for the three binary systems were successfully correlated using the least square method by a two-constant Redlich-Kister(118) equation as follows:

$$\ln \gamma_i = x_j^2 \left[B_{ij} + C_{ij} (3x_i - x_j) \right] \qquad (5\text{-}29)$$

and

$$\ln \gamma_j = x_i^2 \left[B_{ij} + C_{ij} (x_i - 3x_j) \right] \qquad (5\text{-}30)$$

Values of B_{ij} and C_{ij} obtained at four temperatures for the three binary systems are listed in Table 5-3 . Values of $\ln \gamma$ at infinite dilutions are also listed in this table.

The calculated γ values agree well with the experimental data. A comparison at 100°K for the three binary systems is given in Tables A-II. 1 to A-II. 12.

Comparison With Binary Data of Wilson

Wilson studied the three binary systems at isobaric conditions and graphically reported $\ln \gamma$ values at 20°R intervals. The binary constants obtained in this investigation were used to obtain the $\ln \gamma$ curves at constant temperatures which were then compared with the isobaric data reported by Wilson over the appropriate intervals. The reference pressure used in this comparison is identical to that employed by Wilson. The agreement obtained appears to be as satisfactory as Wilson's own

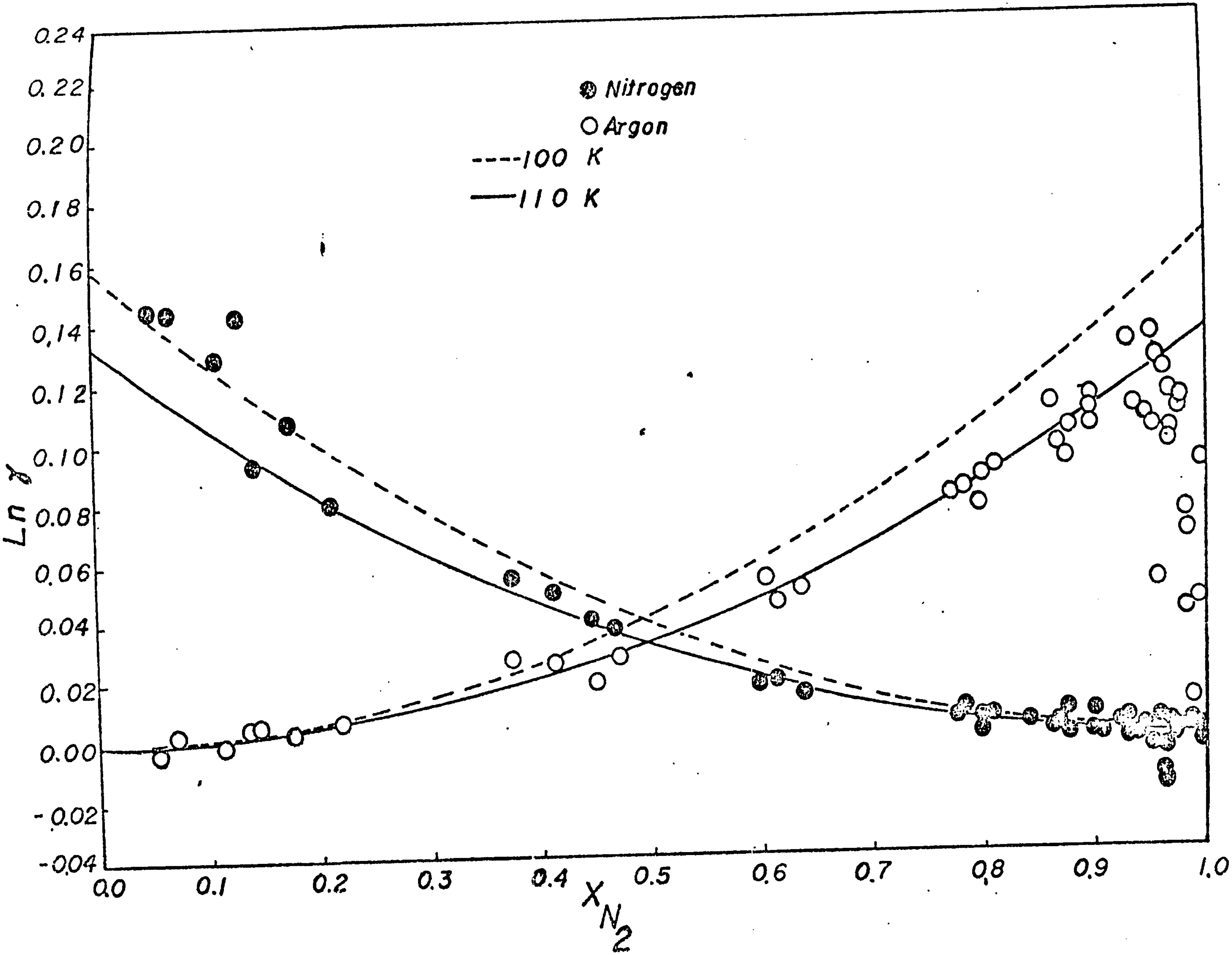


Figure 5-7 Comparison of Wilson's Experimental γ and the Calculated γ Values for the Nitrogen-Argon System at the Interval Between 100° and 110°K

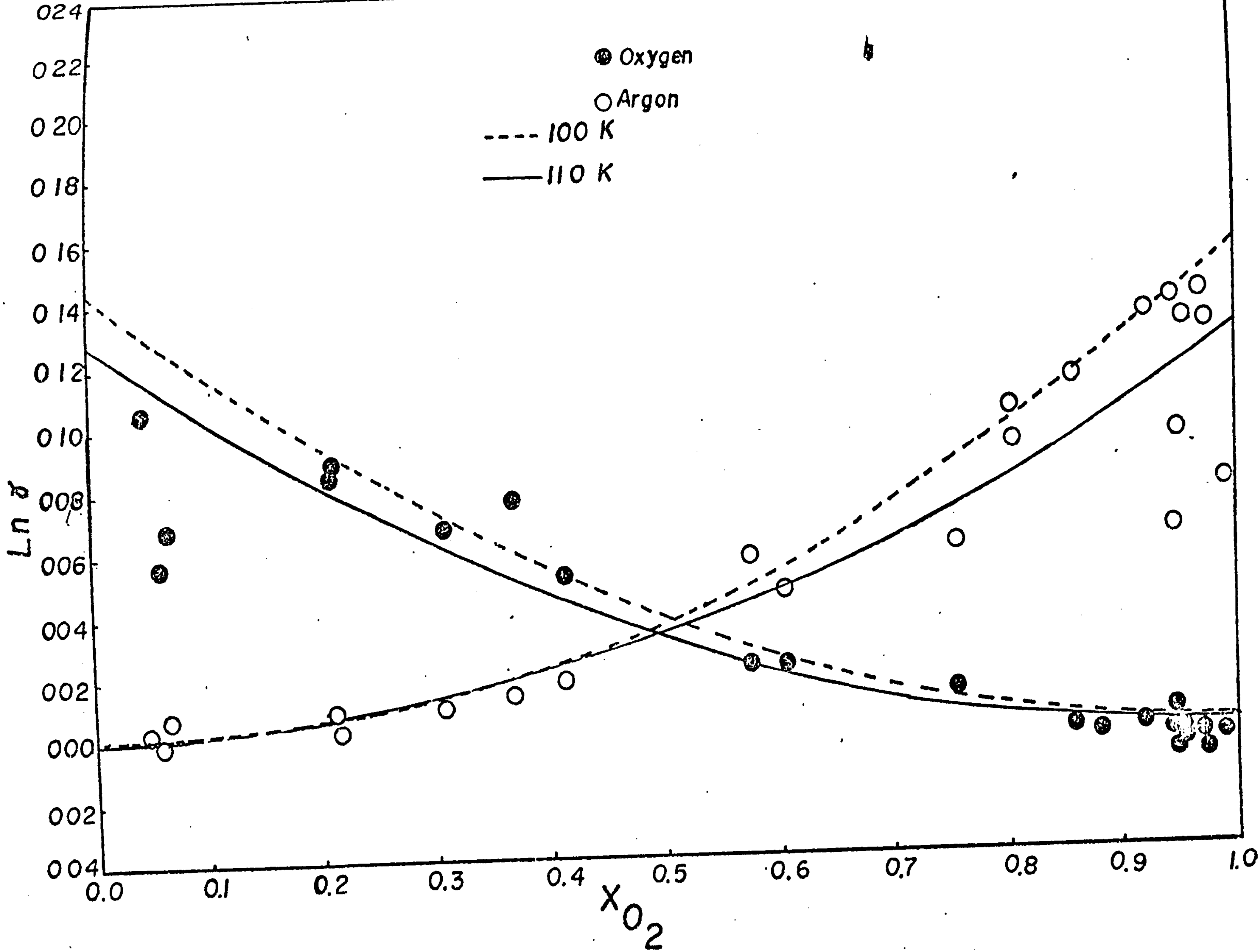


Figure 5-8 Comparison of Wilson's Experimental γ and the Calculated γ Values for the Argon-Oxygen System at the Interval Between 100° and 110°K

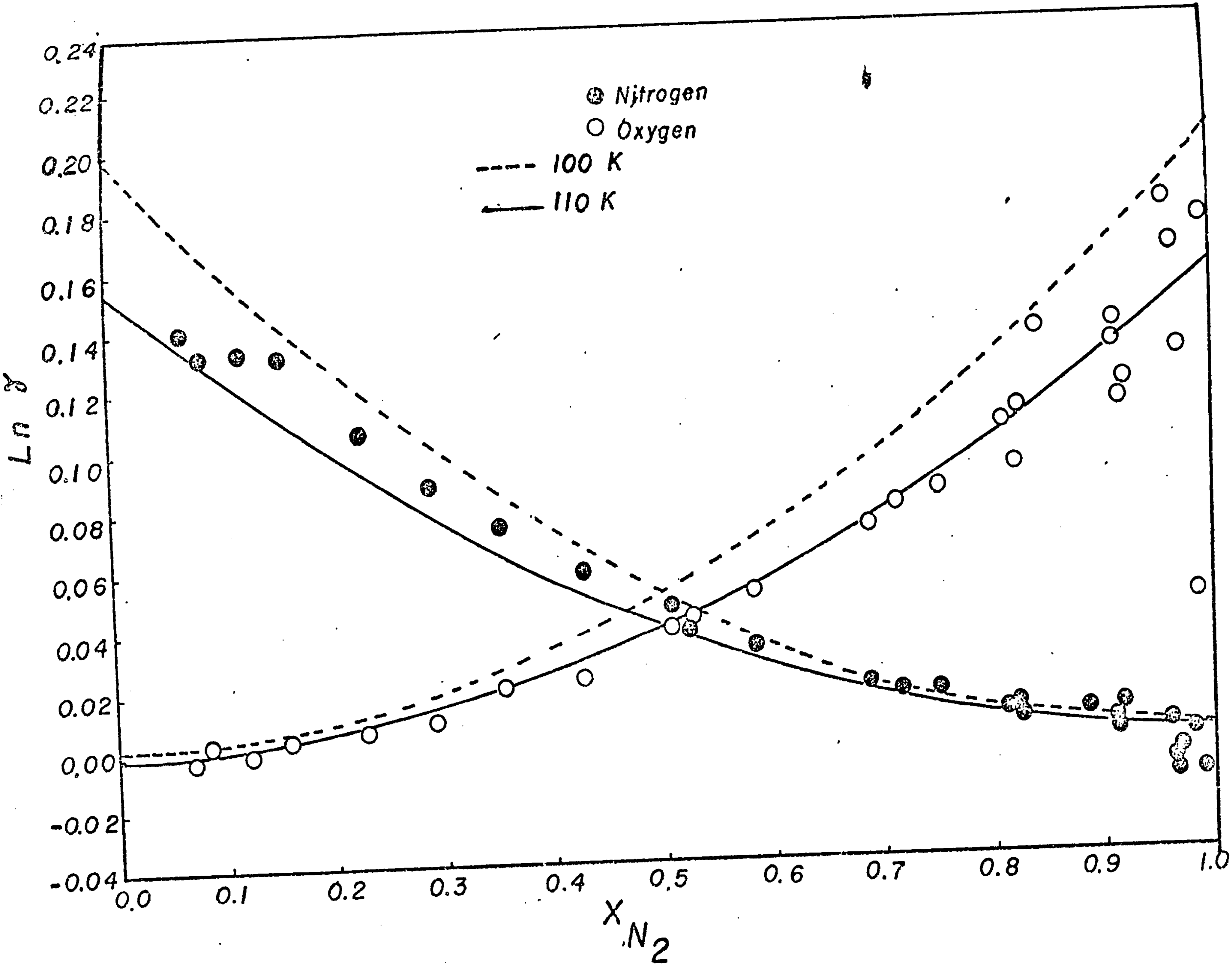


Figure 5-9 Comparison of Wilson's Experimental γ and the Calculated γ Values for the Nitrogen-Oxygen System at the Interval Between 100° and 110°K

correlation(83). The comparisons made at the interval between 100° and 110°K for the three binary systems are shown in Figures (5-7 - 5-9) as examples.

Nitrogen-Argon-Oxygen Mixtures

Very limited data for the ternary system were available in the literature prior to Wilson's experimental investigation, which includes a total of 1962 data points. Recently, Narinskii(82) studied the ternary mixtures by means of a circulation method at temperatures of 90.5°, 100°, 110° and 120°K.

In this investigation, the binary constants obtained at isothermal conditions were used to evaluate the ternary liquid activity coefficients by means of the following expressions

$$\begin{aligned}\ln \gamma_1 = \quad & B_{12}\, x_2(1-x_1) + C_{12}x_2\,(2x_1 - x_2 - 2x_1^2 + 2x_1x_2) \\ & - B_{23}x_2x_3 - 2C_{23}x_2x_3(x_2 - x_3) \\ & + B_{31}x_3\,(1-x_1) + C_{31}x_3\,(x_3 - 2x_1x_3 + 2x_1^2 - 2x_1)\end{aligned} \tag{5-31}$$

$$\begin{aligned}\ln \gamma_2 = \quad & B_{12}x_1(1-x_2) + C_{12}x_1(x_1 - 2x_2 - 2x_1x_2 + 2x_2^2 \\ & + B_{23}x_3(1-x_2) + C_{23}x_3(2x_2 - x_3 - 2x_2^2 + 2x_2x_3) \\ & - B_{13}x_1x_3 - 2C_{31}x_1x_3(x_3 - x_1)\end{aligned} \tag{5-32}$$

and

$$\begin{aligned}\ln \gamma_3 = \; & - B_{12}x_1x_2 - 2C_{12}x_1x_2(x_1 - x_2) \\ & + B_{23}x_2(1-x_3) + C_{23}x_2(x_2 - 2x_3 - 2x_2x_3 + 2x_3^2 \\ & + B_{31}x_1(1-x_3) + C_{31}x_1(2x_3 - x_1 - 2x_3^2 + 2x_1x_3)\end{aligned} \tag{5-33}$$

In these expressions the ternary constants were neglected and only binary constants were involved.

The thermodynamic consistency of the isothermal data of Narinskii and Wilson were tested by means of the test method described in 5.2.2. Results of the test are listed in Tables A-II. 13 to A-II. 20. The calculated γ values obtained were then compared with the consistent data of Narinskii. The comparison is shown in Tables A-II. 21 to A-II. 24.

The calculated γ values obtained from Equations (5-31) - (5-33) were then compared with the consistent data of Wilson. Although Wilson studied the ternary system at isobaric conditions, it was possible to select a sufficient number of data points at constant temperatures. These values were tested for consistency and screened. In this comparison, γ values of Wilson's data were obtained at the reference pressure of this investigation. A summary of the comparison is given in Table 5-4. The average absolute deviation is generally less than 2%.

In conclusion, it may be stated that available vapor-liquid equilibrium data for binary and ternary mixtures of nitrogen, argon and oxygen in the temperature range from 90° to 120°K have been tested and screened. The binary Redlich-Kister constants obtained from consistent isothermal data at four temperatures have been successfully employed for predicting the ternary data without including any ternary constants. The calculated ternary γ values agree well with the data of Narinskii and Wilson.

TABLE 5-4

Summary of Comparison of Calculated and Experimental γ Values for the Ternary System

Source	Temp. °K	No. of Expt points	Average Absolute Deviation, %		
			γ_A	γ_{O_2}	γ_{N_2}
Narinskii (82)	90.5	21	1.14	2.04	1.24
	100.	21	1.45	1.42	0.95
	110	27	1.25	1.26	0.97
	120	25	1.3	1.23	0.83
Wilson (83)	90.5	13	0.73	0.96	0.82
	100.	24	0.30	0.39	0.41
	110	41	0.30	0.4	0.59
	120	22	1.09	1.05	0.43

5.3 EVALUATION OF CALIBRATION CONSTANT FOR BINARY MIXTURES IN THE ABSENCE OF PURE COMPONENTS

In the determination of mixture compositions the gas chromatographic technique has been used extensively. A calculation procedure has been developed in this investigation for the evaluation of calibration constant when two mixtures of a binary system are available, but pure components of the binary are absent. This situation applies, for example, to the determination of equilibrium compositions of liquid and vapor phases in a phase equilibrium study. Normally, mixtures of known compositions, prepared from pure components, are required to establish the calibration constant. Furthermore, it is generally necessary to check the calibration from time to time. The proposed method would eliminate this time-consuming procedure. The simple relation developed in this investigation is however restricted to the situation, when the peak height or peak area ratios (P.R) are directly proportional to the mole fraction ratios (x_1/x_2)(119). Namely,

$$(x_1/x_2) = M\,(P.R) \tag{5-34}$$

Let the peak height or peak area ratios obtained for mixtures a and b are A and B respectively,

$$x_{1a}/x_{2a} = K.A \tag{5-35}$$

$$x_{1b}/x_{2b} = K.B \tag{5-36}$$

where

K = calibration constant

A third mixture C may be prepared by combining mixtures a and b in an arbitrary but known proportions, say m parts of a with n parts of b, and the P, R value obtained for the mixture is C. Then

$$x_{1c}/x_{2c} = K . C \tag{5-37}$$

Furthermore,

$$K . C = \frac{mx_{1a} + nx_{1b}}{mx_{2a} + nx_{2b}} \tag{5-38}$$

By combining Equations (5-35) - (5-38) together with the relationships that

$$x_i + x_j = 1.0 \tag{5-39}$$

it may be readily shown that

$$K = \frac{m\ (A-C) - n\ (C-B)}{mB\ (C-A) - n\,A\ (B-C)} \tag{5-40}$$

In Equation (5-40) the calibration constant K can be evaluated by knowing the P. R. values of A, B, and C, and the arbitrary manner the c mixture is prepared.

The system ethylene-propane was used to test the validity of the proposed method. The calibration curve obtained in Chapter III was used for the purpose of comparison. Two known composition mixtures were prepared. A third mixture was prepared by combining the two mixtures in a known proportion, for example 13 parts of propane to 17 parts of ethylene. The peak height ratios of these mixture were obtained and substituted in Equation (5-40) to obtain the calibration constant K. A comparison between the experimental and calculated results is shown in Figure (5-10).

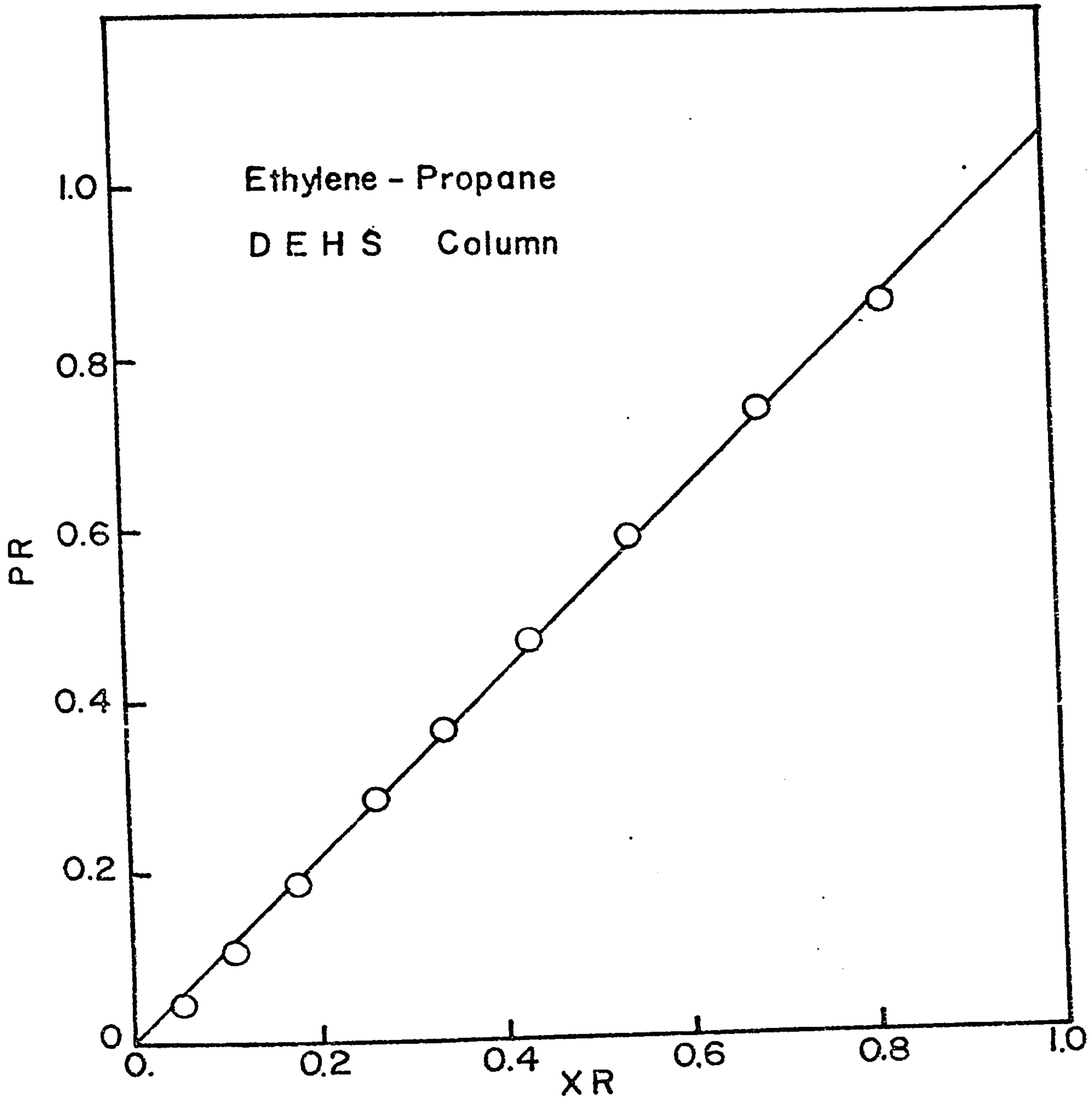


Figure 5-10 Calibration of the Gas Partitioner for the Ethylene-Propane System

CHAPTER VI

SUMMARY AND CONCLUSIONS

In this work some new proposals were suggested for the determination and calculation of phase equilibria data.

A summary and conclusions are as follows:

Experimental Work

A total pressure apparatus was designed and constructed for the experimental study of phase equilibria.

Experimental total pressure-liquid phase composition data for the binaries argon-methane, nitrogen-argon and ethylene-propane were obtained by means of the total pressure apparatus.

In addition a forced circulation apparatus was used to determine vapor-liquid equilibrium data for the binary system argon-ethane and the ternary system argon-methane-ethane.

From the nature of the present experimental work it may be concluded that the forced circulation apparatus is the most suitable and practical for obtaining vapor-liquid equilibrium data in this range of study for the following reasons:

1. It requires shorter time to reach equilibrium.
2. There are no preparations required prior to the experiment.

Calculation of Phase Equilibria Data

The Clausius equation of state was modified for the calculation of phase equilibria in the binary and multicomponent systems. In the modification, the three parameters of the equation were considered temperature dependent. Molal volumes of saturated liquid and vapor together with the condition that the fugacities of the vapor and the liquid phase are equal at the condition of equilibrium were used for evaluating the parameters. The equation was tested successfully for the calculation of volumetric data of pure substances in the liquid and vapor phases. Applications were also made successfully to calculate phase equilibria data for the systems nitrogen-argon, nitrogen-oxygen, argon-oxygen, argon-methane, nitrogen-methane, carbon dioxide-hydrogen sulfide, propane-carbon dioxide, hydrogen sulfide-n-butane and nitrogen-argon-oxygen.

The results obtained from the modified Clausius equation of state were compared with those obtained from the modified R-K equation. Results of the comparison for the pure components showed that the modified Clausius is superior in the vapor phase and the saturation region. In addition the author feels that the modified Clausius equation of state is more adequate since it satisfies the three requirements at saturation.

It may be suggested that in order to improve the quality of the calculated results using the proposed method the following points may be considered:

1. Accurate volumetric data should be used for evaluation of the equation parameters.
2. Vapor pressure of the pure components should match with those reported with the experimental volumetric data used for the evaluation of the equation parameters.

Moreover, the author wishes to stress that, although progress has been made, further improvement can be expected. Such improvement may be directed toward predicting the binary interaction constant, k_{ij}, from pure molecular properties rather than from experimental binary equilibrium data.

The Total Pressure Method for Systems at High Pressures

At high pressure the isothermal isobaric Gibbs-Duhem equation is not valid due to the appreciable effect of pressure on activity coefficient. To overcome this problem an adjusted activity coefficient was introduced with appropriate symmetric convention. As a result, a rigorous method has been proposed for calculating vapor phase compositionss from total pressure-liquid phase composition measurements at high pressure and isothermal conditions.

Data of the three systems argon-methane, nitrogen-argon and ethylene-propane have been successfully used to show the applicability of the proposed procedure.

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APPENDIX I

Tables A-I. 1 to A-I. 31 for Chapter IV

TABLE A-I. 1

Values of Clausius Parameters

	T_R	Ω_a	Ω_b	Ω_c
Methane	0. 576	0. 30007	0. 56108	0. 05300
	0. 602	0. 31820	0. 55191	0. 06130
	0. 628	0. 33023	0. 54607	0. 06450
	0. 654	0. 33241	0. 54851	0. 05940
	0. 680	0. 34742	0. 53663	0. 06630
	0. 707	0. 35507	0. 53093	0. 06710
	0. 733	0. 37371	0. 50851	0. 07900
	0. 759	0. 36986	0. 51720	0. 06940
	0. 785	0. 38229	0. 49915	0. 07660
	0. 811	0. 38708	0. 49191	0. 07665
	0. 837	0. 39100	0. 48340	0. 07655
	0. 890	0. 39879	0. 45815	0. 07900
	0. 916	0. 39653	0. 46259	0. 07280
	0. 942	0. 40186	0. 42894	0. 08019
	0. 968	0. 40289	0. 42928	0. 07710
Argon	0. 556	0. 23080	0. 55792	0. 00010
	0. 579	0. 23996	0. 56098	0. 00060
	0. 627	0. 27104	0. 55304	0. 01390
	0. 658	0. 29431	0. 54139	0. 02600
	0. 703	0. 32740	0. 52164	0. 04400
	0. 736	0. 34305	0. 51346	0. 04980
	0. 775	0. 36456	0. 48861	0. 06290
	0. 824	0. 38045	0. 46868	0. 06930
	0. 895	0. 40307	0. 42240	0. 08390
	0. 922	0. 40760	0. 41079	0. 08540
Nitrogen	0. 570	0. 26704	0. 54517	0. 02200
	0. 590	0. 32048	0. 51468	0. 06214
	0. 613	0. 33304	0. 50938	0. 06690
	0. 664	0. 36835	0. 48610	0. 08520
	0. 698	0. 36268	0. 49463	0. 07230
	0. 746	0. 33307	0. 53679	0. 03350
	0. 781	0. 33619	0. 54600	0. 02719
	0. 822	0. 34813	0. 54370	0. 02861
	0. 876	0. 37782	0. 49982	0. 05024
	0. 917	0. 39325	0. 45918	0. 06435
	0. 952	0. 40291	0. 41302	0. 07761

TABLE A-I.1(cont'd)

	T_R	Ω_a	Ω_b	Ω_c
Oxygen	0.518	0.34172	0.53264	0.10370
	0.582	0.35124	0.53301	0.09183
	0.647	0.36452	0.52796	0.08610
	0.712	0.37469	0.52215	0.08014
	0.776	0.38500	0.51144	0.07681
	0.841	0.39525	0.50859	0.07300
	0.906	0.40262	0.46447	0.07760
	0.388	0.19923	0.58056	0.02440
	0.453	0.27932	0.55868	0.07170
	0.518	0.34170	0.53265	0.10368
	0.582	0.35121	0.53303	0.09181
	0.647	0.36450	0.52798	0.08608
	0.712	0.37470	0.52214	0.08015
	0.776	0.38500	0.51145	0.07680
	0.841	0.39525	0.50859	0.07300
	0.906	0.40262	0.46448	0.07760
Carbon dioxide	0.913	0.41836	0.61937	0.08154
	0.928	0.41732	0.56350	0.08636
	0.931	0.41734	0.59541	0.08308
	0.949	0.41678	0.55101	0.08750
	0.950	0.41636	0.55355	0.08713
	0.953	0.41640	0.54220	0.08825
	0.958	0.41589	0.52933	0.08949
	0.962	0.41541	0.51680	0.09067
	0.968	0.41504	0.50705	0.09125
	0.986	0.42072	0.66165	0.06900
Hydrogen sulfide	0.744	0.43557	0.49964	0.11640
	0.759	0.41734	0.48189	0.12123
	0.774	0.39416	0.55633	0.07761
	0.788	0.43795	0.47828	0.11443
	0.803	0.42308	0.49895	0.10039
	0.832	0.42590	0.48195	0.10132
Propane	0.324	0.37460	0.89480	0.08553
	0.616	0.37476	0.90440	0.07128
	0.690	0.40602	0.85685	0.08179
	0.738	0.40081	0.86115	0.07267

TABLE A-I.1 (cont'd)

	T_R	Ω_a	Ω_b	Ω_c
Ethylene	0.707	0.22911	0.98547	0.03451
	0.806	0.39087	0.74881	0.06473
	0.903	0.41023	0.64375	0.07741
	0.966	0.42009	0.57983	0.11107
Butane	0.731	0.47501	0.65018	0.12505

TABLE A-I.2

Comparison of Calculated and Experimental Compressibilities for Pure Oxygen

Experimental (66)			Clausius	R-K	Clausius	R-K
T, K	P, atm.	Z	Z	Z	ΔZ	ΔZ
60	1.0	0.00507	0.00507	evaluation of parame-ters do not converge	0.0	
	9.87	0.04999	0.05002		0.00001	
	49.35	0.24907	0.24950		0.00043	
	98.69	0.49593	0.49757		0.00164	
	197.38	0.98343	0.98994		0.00651	
70	1.0	0.00450	0.00450	0.00450	0.0	0.0
	9.87	0.04440	0.04438	0.04439	0.00002	0.00001
	49.35	0.22095	0.22060	0.22094	0.00036	0.00001
	98.69	0.43951	0.43822	0.43949	0.00129	0.00002
	197.38	0.87009	0.86619	0.87064	0.00390	0.00055
80	1.0	0.00409	0.00409	0.00490	0.000	0.00000
	9.87	0.04035	0.04029	0.04034	0.00006	0.00001
	49.35	0.20063	0.19992	0.20038	0.00143	0.00025
	98.69	0.39855	0.39346	0.39776	0.00597	0.00081
	197.38	0.78689	0.77067	0.78525	0.01622	0.00164
90	1.0	0.00379	0.00379	0.00379	0.000	0.0000
	9.87	0.03738	0.03731	0.03736	0.00006	0.00002
	49.35	0.18555	0.18391	0.18505	0.00164	0.00050
	98.69	0.36790	0.36216	0.36626	0.00573	0.00164
	197.38	0.72431	0.70619	0.71979	0.01812	0.00452
100	1.0	0.97530(v)	0.97492	0.97756	0.00031	0.00226
	9.87	0.03522	0.03515	0.03519	0.00006	0.00003
	49.35	0.17433	0.17235	0.17360	0.00198	0.00073
	98.69	0.34482	0.33775	0.34221	0.00709	0.00262
	197.38	0.67617	0.65408	0.66849	0.02209	0.00768
110	1.0	0.92602(v)	0.98106	0.98259	0.05504	0.05657
	9.87	0.03373	0.03367	0.03370	0.00006	0.00003
	49.35	0.16636	0.16387	0.16520	0.00249	0.00157
	98.69	0.32780	0.31899	0.32373	0.00881	0.00407
	197.38	0.63941	0.61241	0.62732	0.02700	0.01209

TABLE A-I. 2 (cont'd)

Experimental(66)			Clausius	R-K	Clausius	R-K
T, K	P, atm.	Z	Z	Z	ΔZ	ΔZ
120	1.0	0.98564(v)	0.98531	0.98628	0.00034	0.00064
	9.87	0.83528(v)	0.83514	0.84717	0.00014	0.01189
	49.35	0.16132	0.15815	0.15969	0.00317	0.00162
	98.69	0.31592	0.30460	0.31013	0.01132	0.00579
	197.58	0.61139	0.57754	0.59432	0.03385	0.01706
130	1.0	0.98869(v)	0.98839	0.98896	0.00029	0.00027
	9.87	0.87625(v)	0.87462	0.88103	0.00163	0.00478
	49.35	0.15955	0.15831	0.16005	0.00124	0.00050
	98.69	0.30910	0.29955	0.30585	0.00955	0.00325
	197.38	0.59137	0.55805	0.57632	0.03332	0.01505
140	1.0	0.99092(v)	0.99061	0.99120	0.00031	0.00027
	9.87	0.90358(v)	0.90123	0.90744	0.00235	0.00387
	49.35	0.16254	0.15727	0.16024	0.00527	0.00230
	98.69	0.31913	0.28644	0.29774	0.02163	0.01033
	197.38	0.57817	0.51756	0.54832	0.06061	0.02985

TABLE A-I. 3

Comparison of Calculated and Experimental Compressibility Factors for Argon at $T_R = 1.0$

P_R	(68-71) Z_{expt}	Z_{calc}	$\lvert \Delta Z \rvert$
0.15	0.94449	0.94475	0.00026
0.2	0.92530	0.92547	0.00017
0.25	0.90709	0.90570	0.00139
0.30	0.88149	0.88540	0.00390
0.40	0.84981	0.84283	0.00698
0.6	0.75245	0.74672	0.00480
0.7	0.69245	0.69228	0.00220
0.8	0.61896	0.62789	0.00893
0.9	0.53950	0.54643	0.00692
1.2	0.22906	0.21999	0.00907
1.6	0.27562	0.25204	0.02357
2.0	0.32645	0.29032	0.03610
2.5	0.39210	0.33850	0.05360
3.0	0.46833	0.38577	0.08260
4.0	0.58099	0.47700	0.10399
5.0	0.70449	0.56437	0.14012
6.0	0.82090	0.64854	0.17236
7.0	0.93760	0.73034	0.20726
8.0	1.05065	0.80997	0.24068

TABLE A-I. 4

Comparison of Calculated and Experimental Compressibility Factors for Nitrogen at $T_R = 1.0$

P_R	$Z_{expt}^{(72)}$	Z_{calc}	$\lvert \Delta Z \rvert$
0.4	0.85072	0.84168	0.00904
0.6	0.76914	0.74568	0.02350
0.8	0.63163	0.62493	0.00670
1.2	0.22620	0.22869	0.0025
1.4	0.24677	0.238160	0.0086
1.6	0.27316	0.25247	0.0207
1.6	0.29944	0.27126	0.025
2.0	0.32572	0.29041	0.03530
3.0	0.45587	0.35210	0.07020
4.0	0.57919	0.47581	0.10340
6.0	0.82158	0.64613	0.17540
8.0	1.04650	0.80614	0.24030
10.0	1.26730	0.95900	0.30830

TABLE A-I. 5

Comparison of Calculated and Experimental Results for the System Nitrogen (1) - Argon (2) at 110. 0°K

$k_{ij} = 0.000$

Experimental (75)			Clausius		Deviations			
x_1	P, atm.	y_1	P, atm.	y_1	ΔP		Δy_1	
					atm.	%	mol. fr.	%
0	6.56	0	6.57	0	0.01	0.2	0	0
0.1365	7.74	0.2420	7.74	0.2468	0.00	0.0	0.005	2.1
0.2134	8.35	0.3499	8.38	0.3548	0.02	0.2	0.005	1.4
0.2156	8.38	0.3539	8.39	0.3577	0.01	0.1	0.004	1.1
0.3469	9.42	0.5033	9.45	0.5092	0.04	0.4	0.006	1.2
0.4785	10.41	0.6283	10.50	0.6336	0.09	0.9	0.005	0.8
0.4959	10.61	0.6411	10.64	0.6485	0.03	0.3	0.007	1.1
0.4988	10.67	0.6458	10.66	0.6510	0.01	0.1	0.005	0.8
0.5021	10.64	0.6486	10.69	0.6538	0.05	0.5	0.005	0.8
0.6472	11.67	0.7618	11.84	0.7677	0.16	1.4	0.006	0.8
0.7750	12.69	0.8519	12.86	0.8571	0.17	1.3	0.005	0.6
0.8296	13.09	0.8900	13.31	0.8931	0.22	1.7	0.003	0.3
0.8747	13.48	0.9195	13.68	0.9221	0.20	1.5	0.003	0.3
0.8854	13.56	0.9257	13.77	0.9289	0.22	1.6	0.003	0.3
0.9442	14.00	0.9643	14.27	0.9657	0.26	1.9	0.001	0.1
1.000	14.45	1.0	14.75	1.0	0.29	2.0	0.0	0.0

TABLE A-I. 6

Comparison of Calculated and Experimental Results for the System Nitrogen (1) - Argon at 110.0°K

$k_{ij} = 0.000$

Experimental(75)			R - K		Deviations			
x_1	P, atm.	y_1	P, atm.	y_1	ΔP		Δy_1	
					atm.	%	mol. fr.	%
0	6.56	0	6.57	0	0.01	0.2	0	0
0.1365	7.74	0.2420	7.68	0.2417	0.06	0.8	0.0003	0.12
0.2134	8.35	0.3499	8.30	0.3495	0.06	0.7	0.0004	0.11
0.2156	8.38	0.3539	8.31	0.3523	0.07	0.8	0.0016	0.45
0.3469	9.42	0.5033	9.35	0.5048	0.07	0.7	0.0015	0.30
0.4785	10.41	0.6283	10.31	0.6307	0.04	0.4	0.0024	0.38
0.4959	10.61	0.6411	10.52	0.6459	0.11	1.0	0.0048	0.75
0.4988	10.67	0.6458	10.53	0.6484	0.13	1.2	0.0026	0.40
0.5021	10.64	0.6486	10.56	0.6512	0.08	0.8	0.0026	0.40
0.6472	11.67	0.7618	11.70	0.7665	0.03	0.3	0.0047	0.62
0.775	12.69	0.8519	12.72	0.8569	0.03	0.2	0.0048	0.56
0.8296	13.09	0.8900	13.16	0.8929	0.08	0.6	0.0029	0.33
0.8747	13.48	0.9195	13.53	0.9220	0.05	0.4	0.0025	0.27
0.8854	13.56	0.9257	13.62	0.9288	0.06	0.4	0.0031	0.33
0.9442	14.00	0.9643	14.12	0.9657	0.12	0.9	0.0014	0.15
1.0	14.45	1.	14.60	1.0	0.15	1.0	0	0

TABLE A-I. 7

Comparison of Calculated and Experimental Results for the System Nitrogen (1) - Oxygen (2) at 110. 05°K

$$k_{ij} = 0.0016$$

Experimental[44]			Clausius		Deviations			
x_1	P, atm.	y_1	P, atm.	y_1	ΔP		Δy_1	
					atm.	%	mol. fr.	%
0.0616	6.03	0.1517	5.97	0.1413	0.06	0.99	0.9104	6.86
0.1266	6.61	0.2626	6.58	0.2631	0.03	0.45	0.0005	0.19
0.2017	7.29	0.3750	7.27	0.3787	0.02	0.27	0.0037	0.99
0.4169	9.23	0.6144	9.24	0.6187	0.01	0.11	0.0043	0.69
0.5808	10.73	0.7436	10.74	0.7498	0.01	0.09	0.0062	0.83
0.6309	11.14	0.7787	11.2	0.7845	0.06	0.54	0.0058	0.74
0.7982	12.65	0.8839	12.78	0.8888	0.13	1.03	0.0049	0.55
0.9045	13.66	0.9463	13.82	0.9486	0.16	1.17	0.0023	0.24

TABLE A-I. 8

Comparison of Calculated and Experimental Results for the System Nitrogen (1) - Oxygen (2) at 110.05°K

$k_{ij} = 0.0014$

Experimental (44)			R-K		Deviations			
					ΔP		Δy_1	
x_1	P, atm.	y_1	P, atm.	y_1	atm.	%	mol. fr.	%
0.0616	6.03	0.1517	6.04	0.1503	0.01	0.17	0.0014	0.92
0.1266	6.61	0.2626	6.70	0.2744	0.09	1.36	0.0118	4.49
0.2017	7.29	0.3750	7.43	0.3886	0.14	1.92	0.0136	3.63
0.4169	9.23	0.6144	9.40	0.6195	0.17	1.84	0.0051	0.83
0.5808	10.73	0.7436	10.84	0.7459	0.11	1.02	0.0023	0.31
0.6309	11.14	0.7787	11.28	0.7799	0.14	1.26	0.0012	0.15
0.7982	12.65	0.8839	12.76	0.8842	0.11	0.87	0.0003	0.03
0.9045	13.64	0.9463	13.73	0.9459	0.07	0.51	0.0004	0.04

TABLE A-I. 9

Comparison of Calculated and Experimental Results for the System Nitrogen(1) - Methane (2) at 110. 94°K

$k_{ij} = 0.025$

Experimental (76)			Clausius		Deviations			
x_1	P, atm.	y_1	P, atm.	y_1	ΔP		Δy_1	
					atm.	%	mol. fr.	%
0. 0320	1. 70	0. 4270	1. 72	0. 4283	0. 02	1. 2	0. 0013	0. 30
0. 0470	2. 04	0. 5240	2. 04	0. 5221	0. 0	0. 0	0. 0019	0. 36
0. 0630	2. 33	0. 5980	2. 39	0. 5925	0. 06	2. 6	0. 0055	0. 92
0. 0800	2. 72	0. 6490	2. 74	0. 6470	0. 02	0. 7	0. 0020	0. 31
0. 1150	3. 40	0. 7210	3. 43	0. 7219	0. 03	0. 9	0. 0009	0. 12
0. 1520	4. 08	0. 7680	4. 13	0. 7719	0. 84	0. 9	0. 0039	0. 51
0. 1900	4. 76	0. 8010	4. 80	0. 8068	0. 04	0. 8	0. 0058	0. 72
0. 2340	5. 44	0. 8290	5. 52	0. 8355	0. 08	1. 5	0. 0065	0. 78
0. 2830	6. 12	0. 8510	6. 28	0. 8587	0. 15	2. 5	0. 0077	0. 90
0. 3380	6. 80	0. 8700	7. 06	0. 8780	0. 26	3. 8	0. 0080	0. 92
0. 4860	8. 51	0. 9070	8. 93	0. 9127	0. 42	4. 9	0. 0057	0. 63
0. 6380	10. 21	0. 9330	10. 65	0. 9374	0. 45	4. 4	0. 0044	0. 47
0. 7800	11. 91	0. 9550	12. 31	0. 9586	0. 40	3. 4	0. 0036	0. 38
0. 9020	13. 61	0. 9770	13. 95	0. 9792	0. 34	2. 5	0. 0022	0. 23

TABLE A-I. 10

Comparison of Calculated and Experimental Results for the System Nitrogen (1) - Methane (2) at 110. 94°K

$k_{ij} = 0.026$

Experimental (76)			R-K		Deviations			
x_1	P, atm.	y_1	P, atm.	y_1	ΔP		Δy_1	
					atm.	%	mol. fr.	%
0. 0320	1. 70	0. 4270	1. 65	0. 4267	0. 05	2. 9	0. 0003	0. 07
0. 0470	2. 04	0. 5240	1. 96	0. 5207	0. 08	3. 9	0. 0032	0. 61
0. 0630	2. 33	0. 5980	2. 29	0. 5914	0. 04	1. 7	0. 0066	1. 10
0. 0800	2. 72	0. 6490	2. 63	0. 6462	0. 09	3. 3	0. 0008	0. 35
0. 1150	3. 40	0. 7210	3. 30	0. 7218	0. 10	2. 9	0. 0008	0. 11
0. 1520	4. 08	0. 7680	3. 98	0. 7722	0. 11	2. 7	0. 0042	0. 55
0. 1900	4. 76	0. 8010	4. 63	0. 8075	0. 13	2. 7	0. 0065	0. 81
0. 2340	5. 44	0. 8290	5. 34	0. 8365	0. 10	1. 8	0. 0075	0. 90
0. 2830	6. 12	0. 8510	6. 08	0. 8599	0. 04	0. 7	0. 0089	1. 05
0. 3380	6. 80	0. 8700	6. 86	0. 8795	0. 06	0. 9	0. 0095	1. 09
0. 4860	8. 51	0. 9070	8. 73	0. 9145	0. 22	2. 6	0. 0075	0. 82
0. 6380	10. 21	0. 9330	10. 47	0. 9392	0. 27	2. 6	0. 0062	0. 67
0. 7800	11. 91	0. 9550	12. 14	0. 9600	0. 23	1. 9	0. 0050	0. 52
0. 9020	13. 61	0. 9770	13. 79	0. 9800	0. 18	1. 3	0. 0030	0. 31

TABLE A-I. 11

Comparison of Calculated and Experimental Results for the System Argon (1) - Oxygen (2) at 110.0°K

$k_{ij} = 0.0077$

Experimental(77)			Clausius		Deviations			
x_1	P, atm.	y_1	P, atm.	y_1	ΔP		Δy_1	
					atm.	%	mol. fr.	%
0	5.35	0	5.36	0	0.01	0.2	0	0
0.0452	5.45	0.0602	5.46	0.0593	0.01	0.2	0.0069	1.50
0.0841	5.53	0.1098	5.54	0.1081	0.01	0.2	0.0017	1.55
0.1624	5.67	0.2006	5.68	0.2010	0.01	0.2	0.0004	0.20
0.2668	5.84	0.3163	5.86	0.3155	0.02	0.3	0.0008	0.25
0.3457	5.98	0.3960	5.98	0.3964	0.00	0.0	0.0004	0.10
0.3885	6.04	0.4372	6.04	0.4387	0.00	0.0	0.0015	0.34
0.5664	6.24	0.6050	6.26	0.6058	0.02	0.3	0.0008	0.13
0.6986	6.37	0.7230	6.39	0.7247	0.02	0.3	0.0017	0.24
0.7734	6.44	0.7907	6.45	0.7915	0.01	0.2	0.0008	0.10
0.7855	6.45	0.8025	6.46	0.8024	0.01	0.2	0.0001	0.01
0.8504	6.49	0.8608	6.50	0.8608	0.01	0.2	0.0000	0.00
0.9251	6.53	0.9296	6.54	0.9293	0.01	0.2	0.0003	0.03
0.9425	6.54	0.9460	6.55	0.9455	0.01	0.2	0.0005	0.05
1.0	6.56	1.0	6.57	1.0	0.01	0.2	0.0	0

TABLE A-I. 12

Comparison of Calculated and Experimental Results for the System Argon (1) - Oxygen (2) at 110. 0°K

$k_{ij} = 0.0104$

Experimental (77)			R-K		Deviations			
					ΔP		Δy_1	
x_1	P, atm.	y_1	P, atm.	y_1	atm.	%	mol. fr.	%
0.0	5.35	0.0	5.36	0.0	0.01	0.2	0	0
0.0452	5.45	0.0620	5.46	0.0596	0.01	0.2	0.0006	0.97
0.0841	5.53	0.1098	5.54	0.1086	0.01	0.2	0.0012	1.09
0.1624	5.67	0.2006	5.69	0.2014	0.02	0.4	0.0008	0.40
0.2668	5.84	0.3163	5.87	0.3156	0.03	0.5	0.0007	0.22
0.3457	5.98	0.3960	5.99	0.3962	0.01	0.2	0.0002	0.05
0.3885	6.04	0.4372	6.05	0.4384	0.01	0.2	0.0012	0.27
0.5664	6.24	0.6050	6.27	0.6051	0.03	0.5	0.0001	0.02
0.6986	6.37	0.7230	6.39	0.7241	0.02	0.3	0.0011	0.15
0.7734	6.44	0.7907	6.45	0.7911	0.01	0.2	0.0004	0.05
0.7855	6.45	0.8025	6.46	0.8020	0.01	0.2	0.0005	0.06
0.8504	6.49	0.8608	6.50	0.8606	0.01	0.2	0.0002	0.02
0.9251	6.53	0.9296	6.54	0.9293	0.01	0.2	0.0003	0.03
0.9425	6.54	0.9460	6.55	0.9455	0.01	0.2	0.0005	0.05
1.0	6.56	1.	6.57	1.0	0.01	0.2	0.	0

TABLE A-I. 13

Comparison of Calculated and Experimental Results for the System Argon (1) - Methane (2) at 115. 2°K

$k_{ij} = 0.0184$

Experimental (78)			Clausius		Deviations			
					ΔP		Δy_1	
x_1	P, atm.	y_1	P, atm.	y_1	atm.	%	mol. fr.	%
0.044	1.95	0.2640	1.77	0.2629	0.18	9.2	0.0011	0.41
0.1050	2.37	0.4690	2.33	0.4666	0.04	1.7	0.0024	0.51
0.1170	2.45	0.4930	2.44	0.4950	0.01	0.4	0.0020	0.41
0.1700	2.99	0.5940	2.91	0.5941	0.08	2.7	0.0001	0.02
0.2210	3.38	0.6470	3.34	0.6621	0.04	1.2	0.0151	2.33
0.3330	4.15	0.7470	4.24	0.7614	0.09	2.2	0.0144	1.93
0.4350	5.10	0.8180	5.01	0.8204	0.09	1.8	0.0024	0.29
0.4760	5.31	0.8310	5.31	0.8394	0.00	0.0	0.0084	1.01
0.4810	5.29	0.8430	5.35	0.8416	0.06	1.1	0.0014	0.17
0.6020	6.26	0.8850	6.21	0.8874	0.05	0.8	0.0024	0.27
0.6150	6.19	0.8630	6.30	0.8917	0.11	1.8	0.0287	3.33
0.656	6.53	0.8870	6.58	0.9048	0.05	0.8	0.0178	2.01
0.658	6.51	0.8960	6.60	0.9054	0.09	1.4	0.0094	1.05
0.7082	7.04	0.9240	6.95	0.9204	0.09	1.3	0.0036	0.39
0.7250	7.00	0.9230	7.07	0.9253	0.07	1.0	0.0023	0.25
0.7290	7.07	0.9240	7.09	0.9264	0.02	0.3	0.0024	0.26
0.8020	7.62	0.9470	7.61	0.9467	0.01	0.1	0.0003	0.03
0.8070	7.68	0.9480	7.65	0.9481	0.03	0.4	0.0001	0.01
0.8610	8.03	0.9620	8.04	0.9626	0.01	0.1	0.0006	0.06
0.8760	8.14	0.9620	8.15	0.9666	0.01	0.1	0.0046	0.48
0.9160	8.45	0.9780	8.45	0.9773	0.0	0.0	0.0007	0.07
0.9310	8.57	0.9830	8.56	0.9813	0.01	0.1	0.0017	0.17
0.9530	8.73	0.9860	8.73	0.9872	0.01	0.1	0.0012	0.12

TABLE A-I. 14

Comparison of Calculated and Experimental Results for the System Argon (1) - Methane (2) at 115. 2°K

k_{ij} = 0.0229

Experimental (78)			R-K		Deviations			
					ΔP		Δy_1	
x_1	P, atm.	y_1	P, atm.	y_1	atm.	%	mol. fr.	%
0. 0440	1. 95	0. 2640	1. 76	0. 2623	0. 18	9. 2	0. 0017	0. 64
0. 1085	2. 37	0. 4690	2. 33	0. 4661	0. 04	1. 7	0. 0029	0. 62
0. 1170	2. 45	0. 4930	2. 44	0. 4946	0. 01	0. 4	0. 0016	0. 32
0. 1700	2. 99	0. 5940	2. 90	0. 5939	0. 09	3. 0	0. 0091	0. 02
0. 2210	3. 38	0. 6470	5. 33	0. 6621	0. 05	1. 5	0. 0151	2. 33
0. 3330	4. 15	0. 7470	4. 24	0. 7617	0. 09	2. 2	0. 0147	1. 97
0. 4350	5. 10	0. 8180	5. 01	0. 8208	0. 09	1. 8	0. 0028	0. 34
0. 4760	5. 31	0. 8310	5. 31	0. 8398	0. 0	0. 0	0. 0088	1. 06
0. 4810	5. 29	0. 8430	5. 35	0. 8420	0. 06	1. 1	0. 0010	0. 12
0. 6020	6. 26	0. 8850	6. 21	0. 8878	0. 05	0. 8	0. 0028	0. 32
0. 6150	6. 19	0. 8630	6. 30	0. 8921	0. 11	1. 8	0. 0291	3. 37
0. 6560	6. 53	0. 8870	6. 58	0. 9052	0. 05	0. 8	0. 0182	2. 05
0. 6580	6. 51	0. 8960	6. 60	0. 9058	0. 09	1. 4	0. 0098	1. 09
0. 7080	7. 04	0. 9240	6. 95	0. 9207	0. 09	1. 3	0. 0033	0. 36
0. 7250	7. 00	0. 9230	7. 07	0. 9256	0. 07	1. 0	0. 0026	0. 28
0. 7290	7. 07	0. 9240	7. 09	0. 9268	0. 02	0. 3	0. 0028	0. 30
0. 8020	7. 62	0. 9470	7. 61	0. 9470	0. 01	0. 1	0. 0	0. 0
0. 8070	7. 68	0. 9480	7. 65	0. 9483	0. 04	0. 5	0. 0003	0. 03
0. 8610	8. 03	0. 9620	8. 04	0. 9628	0. 01	0. 1	0. 0008	0. 08
0. 8760	8. 14	0. 9620	8. 15	0. 9668	0. 01	0. 1	0. 0048	0. 50
0. 9160	8. 45	0. 9780	8. 45	0. 9774	0. 0	0. 0	0. 0006	0. 06
0. 9310	8. 57	0. 9830	8. 56	0. 9814	0. 01	0. 1	0. 0016	0. 16
0. 9530	8. 73	0. 9860	8. 73	0. 9872	0. 0	0. 0	0. 0013	0. 13

TABLE A-I. 15

Comparison of Calculated and Experimental Results for the System Carbon Dioxide (1) - Hydrogen Sulfide (2)

	Experimental[(79)]			Clausius		Deviations			
						ΔP		Δy_1	
T °K	x_1	P, atm.	y_1	P, atm.	y_1	atm.	%	mol. fr.	%
277. 50	0. 033	13. 61	0. 145	13. 27	0. 1452	0. 34	2. 49	0. 0002	0. 14
	0. 150	20. 41	0. 460	18. 86	0. 4235	1. 5	7. 35	0. 0366	7. 96
k_{ij} = 0. 0586	0. 395	27. 22	0. 645	27. 08	0. 6510	0. 14	0. 51	0. 0060	0. 93
	0. 710	34. 02	0. 815	34. 03	0. 8132	0. 01	0. 03	0. 0018	0. 22
283. 00	0. 093	20. 41	0. 349	20. 47	0. 3489	0. 05	0. 24	0. 0001	0. 03
	0. 275	27. 22	0. 551	30. 08	0. 5773	2. 87	10. 54	0. 0263	4. 78
k_{ij} = 0. 0851	0. 503	34. 02	0. 697	35. 20	0. 6974	1. 18	3. 47	0. 0004	0. 06
	0. 791	40. 83	0. 860	41. 98	0. 8351	1. 15	2. 82	0. 0249	2. 89
288. 62	0. 052	20. 41	0. 230	19. 80	0. 2088	0. 61	2. 99	0. 0212	9. 22
	0. 185	27. 22	0. 460	27. 96	0. 4601	0. 74	2. 72	0. 001	0. 02
k_{ij} = 0. 0613	0. 360	34. 02	0. 605	35. 41	0. 6069	1. 38	4. 06	0. 0019	0. 31
	0. 581	40. 83	0. 728	42. 13	0. 7255	1. 30	3. 18	0. 0025	0. 34
294. 27	0. 022	20. 41	0. 106	19. 60	0. 0946	0. 81	3. 97	0. 0114	10. 75
	0. 116	27. 22	0. 327	26. 67	0. 3414	0. 56	2. 06	0. 0144	4. 40
k_{ij} = 0. 07	0. 250	34. 02	0. 511	34. 64	0. 511	0. 67	1. 97	0. 0	0.
	0. 423	40. 83	0. 631	40. 41	0. 6316	0. 42	1. 03	0. 0006	0. 09
	0. 850	54. 44	0. 888	54. 90	0. 8501	0. 54	0. 99	0. 0379	4. 27
299. 73	0. 067	27. 22	0. 245	27. 20	0. 2389	0. 02	0. 07	0. 0061	2. 48
	0. 171	34. 02	0. 417	35. 48	0. 4217	1. 45	4. 26	0. 0047	1. 12
k_{ij} = 0. 0856	0. 300	40. 83	0. 540	43. 04	0. 5391	2. 21	5. 41	0. 0009	0. 17
	0. 650	54. 44	0. 755	55. 91	0. 7795	1. 48	2. 72	0. 0255	3. 37

TABLE A-I. 16

Comparison of Calculated and Experimental Results for the System Carbon Dioxide (1) - Hydrogen Sulfide (2)

$k_{ij} = 0.0721$

	Experimental$^{(79)}$			Clausius		Deviations			
						ΔP		Δy_1	
T, °K	x_1	P, atm.	y_1	P, atm.	y_1	atm.	%	mol. fr.	%
277.5	0.033	13.61	0.145	13.62	0.1633	0.01	0.07	0.0183	12.62
	0.150	20.41	0.460	20.06	0.4486	0.35	1.71	0.0114	2.48
	0.395	27.22	0.645	28.59	0.6575	1.37	5.03	0.0125	1.94
	0.710	34.02	0.815	35.22	0.8044	1.20	3.52	0.0106	1.30
283.0	0.093	20.41	0.349	19.51	0.3252	0.90	4.40	0.0238	6.82
	0.275	27.22	0.551	28.34	0.5638	1.12	4.11	0.0128	2.32
	0.503	34.02	0.697	35.53	0.6984	1.51	4.44	0.0014	0.20
	0.791	40.83	0.860	41.84	0.8439	1.01	2.47	0.0161	1.87
288.6	0.052	20.41	0.230	20.38	0.2265	0.03	0.15	0.0035	1.52
	0.185	27.22	0.460	29.38	0.4769	2.16	7.93	0.0169	3.67
	0.360	34.02	0.605	37.04	0.6125	3.02	8.88	0.0075	1.24
	0.581	40.83	0.728	43.54	0.7210	2.72	6.66	0.0070	0.96
294.3	0.022	20.41	0.106	19.65	0.0962	0.75	3.67	0.0098	9.24
	0.116	27.22	0.327	26.86	0.3446	0.35	1.29	0.0176	5.38
	0.250	34.02	0.511	34.99	0.5131	0.97	2.85	0.0021	0.41
	0.423	40.83	0.631	41.74	0.6319	0.91	2.23	0.0009	0.14
	0.850	54.44	0.888	55.34	0.8500	0.90	1.65	0.0380	4.28
299.7	0.067	27.22	0.245	26.30	0.2206	0.92	3.38	0.0244	9.96
	0.171	34.02	0.417	33.70	0.4039	0.32	0.94	0.0131	3.14
	0.300	40.83	0.540	40.82	0.5297	0.01	0.02	0.0103	1.91
	0.650	54.44	0.755	53.92	0.7371	0.52	0.96	0.0179	2.37

TABLE A-I. 17

Comparison of Calculated and Experimental Results for the System Carbon Dioxide (1) - Hydrogen Sulfide (2)

	Experimental$^{(79)}$			R-K		Deviations			
						ΔP		Δy_1	
T, °K	x_1	P, atm.	y_1	P, atm.	y_1	atm.	%	mol. fr.	%
277.5	0.033	13.61	0.145	13.56	0.1630	0.05	0.37	0.0180	12.41
k_{ij} = 0.0738	0.150	20.41	0.460	19.65	0.4488	0.76	3.72	0.0112	2.43
	0.395	27.22	0.645	27.78	0.6620	0.56	0.94	0.0170	2.64
	0.710	34.02	0.815	34.28	0.815	0.26	0.76	0.0	0.
283.00	0.093	20.41	0.349	20.08	0.3483	0.34	1.67	0.0007	0.20
k_{ij} = 0.0856	0.275	27.22	0.551	28.91	0.5795	1.69	6.21	0.0285	5.17
	0.503	34.02	0.697	35.55	0.7058	1.53	4.50	0.0088	1.26
	0.791	40.83	0.860	41.38	0.8473	0.56	1.37	0.0127	1.48
288.62	0.052	20.41	0.230	19.67	0.2054	0.74	3.63	0.0246	10.70
k_{ij} = 0.0726	0.185	27.21	0.460	27.22	0.4601	0.36	1.32	0.0001	0.02
	0.360	34.02	0.605	34.97	0.6126	0.94	2.76	0.0076	1.26
	0.581	40.83	0.728	41.75	0.7347	0.92	2.25	0.0067	0.92
294.27	0.022	20.41	0.106	19.39	0.0887	1.03	5.05	0.0173	16.32
k_{ij} = 0.0596	0.116	27.22	0.327	25.60	0.3312	1.62	5.95	0.0042	1.28
	0.250	34.02	0.511	32.78	0.5094	1.24	3.64	0.0016	0.31
	0.423	40.83	0.631	40.10	0.6424	0.73	1.79	0.0114	1.81
	0.850	54.44	0.888	54.31	0.8913	0.12	0.22	0.0033	0.37
299.73	0.067	27.22	0.245	27.86	0.2400	0.64	2.35	0.0050	2.04
k_{ij} = 0.0896	0.171	34.02	0.417	35.91	0.4220	1.89	5.56	0.0050	1.20
	0.300	40.83	0.540	43.15	0.5400	2.32	5.68	0.0	0.0
	0.650	54.44	0.755	55.83	0.7366	1.40	2.57	0.0184	2.44

TABLE A-I. 18

Comparison of Calculated and Experimental Results for the System Propane (1) - Carbon Dioxide (2) at 40. 0F

$k_{ij} = 0.0762$

Experimental(80)			Clausius		Deviations			
x_1	P, atm.	y_1	P, atm.	y_1	ΔP		Δy_1	
					atm.	%	mol. fr.	%
0. 9753	6. 80	0. 7944	6. 55	0. 8061	0. 25	3. 67	0. 0117	1. 47
0. 9116	10. 21	0. 5324	9. 80	0. 5412	0. 41	4. 01	0. 0088	1. 65
0. 8398	13. 61	0. 3964	13. 24	0. 3981	0. 37	2. 72	0. 0017	0. 42
0. 7598	17. 01	0. 3136	16. 79	0. 3092	0. 22	1. 29	0. 0044	1. 40
0. 6684	20. 41	0. 2569	20. 48	0. 2469	0. 06	0. 29	0. 0100	3. 89
0. 5639	23. 82	0. 2124	24. 20	0. 1998	0. 38	1. 60	0. 0125	5. 88
0. 4468	27. 22	0. 1691	27. 83	0. 1615	0. 61	2. 24	0. 0076	4. 49
0. 3286	30. 62	0. 1312	31. 04	0. 1289	0. 42	1. 37	0. 0023	1. 75
0. 2044	34. 02	0. 0898	34. 12	0. 0936	0. 09	0. 26	0. 0038	4. 23
0. 0599	37. 43	0. 0298	37. 41	0. 0373	0. 02	0. 05	0. 0075	25. 17

TABLE A-I. 19

Comparison of Calculated and Experimental Results for the System Propane (1) - Carbon Dioxide (2) at 40. 0° F

$k_{ij} = 0.095$

Experimental(80)			R-K		Deviations			
					ΔP		Δy_1	
x_1	P, atm.	y_1	P, atm.	y_1	atm.	%	mol. fr.	%
0.9753	6.80	0.7944	6.70	0.8069	0.10	1.53	0.0125	1.57
0.9116	10.21	0.5324	9.98	0.5414	0.23	2.23	0.0090	1.69
0.8398	13.61	0.3964	13.46	0.3969	0.15	1.08	0.0005	0.13
0.7598	17.01	0.3136	17.06	0.3069	0.05	0.30	0.0067	2.14
0.6684	20.41	0.2569	20.80	0.2436	0.38	1.88	0.0133	5.18
0.5639	23.82	0.2124	24.58	0.1957	0.76	3.67	0.0167	7.86
0.4468	27.22	0.1691	28.24	0.1569	1.02	3.77	0.0122	7.21
0.3286	30.62	0.1312	31.44	0.1245	0.82	2.68	0.0067	5.11
0.2044	34.02	0.0898	34.42	0.0904	0.40	1.18	0.0006	0.67
0.0599	37.43	0.0298	37.49	0.0369	0.08	0.22	0.0071	23.83

TABLE A-I. 20

Comparison of Calculated and Experimental Results for the System Hydrogen Sulfide (1) - n-Butane (2) at 100° F

$k_{ij} = 0.0489$

Experimental (81)			Clausius		Deviations			
x_1	P, atm.	y_1	P, atm.	y_1	ΔP		Δy_1	
					atm.	%	mol. fr.	%
0.021	4.11	0.133	4.08	0.1438	0.03	0.73	0.0108	8.12
0.066	5.77	0.386	5.34	0.3492	0.43	7.45	0.0368	9.53
0.131	7.55	0.539	7.15	0.5222	0.41	5.43	0.0168	3.12
0.209	9.99	0.655	9.31	0.6436	0.68	6.81	0.0114	1.74
0.355	13.84	0.763	13.29	0.7691	0.55	3.97	0.0061	0.79
0.468	16.28	0.818	16.26	0.827	0.02	0.12	0.0090	1.10
0.607	19.31	0.870	19.71	0.8775	0.40	2.07	0.0075	0.86
0.716	21.49	0.904	22.17	0.9089	0.68	3.16	0.0049	0.54
0.805	23.34		23.93	0.932	0.59	2.53		
0.858	24.41		24.86	0.9457	0.44	1.80		

TABLE A-I. 21

Comparison of Calculated and Experimental Results for the System Hydrogen Sulfide (1) - n-Butane (2) at 100° F

$k_{ij} = 0.062$

Experimental (81)			R-K		Deviations			
					ΔP		Δy_1	
x_1	P, atm.	y_1	P, atm.	y_1	atm.	%	mol. fr.	%
0.021	4.11	0.133	4.13	0.152	0.02	0.49	0.019	14.29
0.066	5.77	0.386	5.43	0.366	0.34	5.89	0.02	5.18
0.131	7.55	0.539	7.29	0.540	0.26	3.44	0.001	0.19
0.209	9.99	0.655	9.48	0.661	0.51	5.11	0.006	0.92
0.355	13.84	0.763	13.41	0.783	0.43	3.11	0.020	2.62
0.468	16.28	0.818	16.26	0.838	0.02	0.12	0.020	2.44
0.607	19.31	0.870	19.49	0.886	0.17	0.88	0.016	1.84
0.716	21.49	0.904	21.76	0.916	0.27	1.26	0.012	1.33
0.805	23.34		23.45	0.938	0.11	0.47		
0.858	24.41		24.39	0.951	0.02	0.08		

TABLE A-I. 22

Comparison of Calculated and Experimental Results for the System Nitrogen (1) - Argon (2) - Oxygen (3) at 110. 0°K

Experimental (82)					Clausius			Deviations					
								ΔP		Δy_1		Δy_2	
x_1	x_2	P, atm.	y_1	y_2	P, atm.	y_1	y_2	atm.	%	mol. fr.	%	mol. fr.	%
0.8384	0.0427	13.01	0.9060	0.0273	13.20	0.0075	0.0274	0.19	1.5	0.0015	0.17	0.0001	0.37
0.6429	0.0904	11.29	0.7792	0.0630	11.43	0.7824	0.0624	0.13	1.2	0.0032	0.41	0.0006	0.95
0.4357	0.1156	9.54	0.6195	0.0921	9.58	0.6218	0.0905	0.04	0.4	0.0027	0.44	0.0016	1.73
0.2821	0.1180	8.17	0.4642	0.1059	8.20	0.4681	0.1051	0.03	0.4	0.0039	0.84	0.0008	0.76
0.2521	0.1187	7.91	0.4314	0.1100	7.93	0.4325	0.1088	0.02	0.3	0.0011	0.25	0.0011	1.00
0.1715	0.0792	7.04	0.3256	0.0792	7.13	0.3282	0.0837	0.09	1.3	0.0026	0.80	0.0079	9.97
0.1079	0.0548	6.45	0.2273	0.0605	6.49	0.2275	0.0685	0.04	0.6	0.0002	0.09	0.0003	0.50
0.0454	0.1897	6.14	0.0997	0.2169	6.14	0.1004	0.2169	0.00	0.0	0.0007	0.70	0.0	0.
0.1046	0.2029	6.69	0.2105	0.2133	6.72	0.2113	0.2133	0.03	0.4	0.0008	0.38	0.0	0.
0.7758	0.1157	12.55	0.8600	0.0780	12.71	0.8646	0.0747	0.16	1.3	0.0046	0.53	0.0032	4.10
0.5530	0.1985	10.69	0.7046	0.1425	10.76	0.7087	0.1411	0.08	0.7	0.0042	0.60	0.0014	0.98
0.4583	0.2006	9.63	0.5972	0.1815	9.91	0.6329	0.1514	0.28	2.9	0.0357	5.98	0.0301	16.58
0.3304	0.2120	8.75	0.5078	0.1769	8.78	0.5116	0.1760	0.03	0.3	0.0038	0.75	0.0009	0.51
0.5535	0.2965	10.77	0.6948	0.2128	10.90	0.7023	0.2069	0.13	1.2	0.0075	1.08	0.0059	2.77
0.4852	0.3459	10.25	0.6411	0.2529	10.35	0.6452	0.2491	0.10	1.0	0.0041	0.64	0.0038	1.50
0.2599	0.3689	8.36	0.4165	0.3173	8.36	0.4215	0.3130	0.00	0.	0.0050	1.20	0.0043	1.36
0.2072	0.3029	7.45	0.3564	0.2750	7.80	0.3595	0.2748	0.35	4.7	0.0031	0.87	0.0002	0.07
0.0758	0.3680	6.61	0.1473	0.3825	6.70	0.1529	0.3788	0.09	1.4	0.0056	3.80	0.0037	0.97
0.0374	0.0460	6.45	0.0806	0.4797	6.47	0.0781	0.4832	0.02	0.3	0.0025	3.10	0.0035	0.73
0.4381	0.3883	9.92	0.5981	0.2899	9.98	0.6026	0.2860	0.05	0.5	0.0045	0.75	0.0039	1.35
0.3329	0.4550	9.04	0.4917	0.3589	9.11	0.4985	0.3567	0.07	0.8	0.0068	1.38	0.0022	0.61
0.3581	0.4080	9.20	0.5255	0.3152	9.28	0.5263	0.3167	0.08	0.9	0.0008	0.15	0.0015	0.48
0.1669	0.4586	7.56	0.2938	0.4176	7.64	0.2958	0.4159	0.07	0.9	0.0020	0.68	0.0017	0.41
0.3160	0.5560	9.03	0.4435	0.4663	9.08	0.4774	0.4337	0.05	0.6	0.0339	7.64	0.0326	6.99
0.2321	0.6109	8.36	0.3758	0.5076	8.39	0.3758	0.5076	0.02	0.2	0.0030	0.80	0.0022	0.43
0.1982	0.5659	8.01	0.3311	0.4885	8.04	0.3358	0.4865	0.02	0.2	0.0047	1.42	0.0020	0.41
0.1244	0.7794	7.55	0.2262	0.6953	7.57	0.2270	0.6940	0.02	0.3	0.0008	0.35	0.0013	0.19

TABLE A-I. 23

Comparison of Calculated and Experimental Results for the System Nitrogen (1) - Argon (2) - Oxygen (3) at 110.0°K

Experimental (82)					R.-K			Deviations					
								ΔP		Δy_1		Δy_2	
x_1	x_2	P, atm.	y_1	y_2	P, atm.	y_1	y_2	atm.	%	mol. fr.	%	mol. fr.	%
0.8384	0.0427	13.01	0.9060	0.0273	13.07	0.9068	0.0271	0.07	0.5	0.0008	0.09	0.0002	0.73
0.6429	0.0904	11.29	0.7792	0.0630	11.33	0.7813	0.0620	0.04	0.4	0.0021	0.27	0.0010	1.59
0.4357	0.1156	9.54	0.6195	0.0921	9.51	0.6205	0.0896	0.03	0.3	0.0010	0.16	0.0025	2.71
0.2821	0.1180	8.17	0.4642	0.1059	8.15	0.4669	0.1037	0.01	0.1	0.0027	0.58	0.0022	2.08
0.2521	0.1187	7.91	0.4314	0.1100	7.89	0.4314	0.1073	0.02	0.3	0.0	0.0	0.0027	2.45
0.1715	0.0792	7.04	0.3256	0.0792	7.10	0.3272	0.0789	0.06	0.9	0.0016	0.49	0.0003	0.38
0.1079	0.0548	6.45	0.2273	0.0605	6.48	0.2266	0.0596	0.03	0.5	0.0007	0.31	0.0009	1.49
0.0454	0.1897	6.14	0.0997	0.2169	6.12	0.1003	0.2138	0.02	0.3	0.0006	0.60	0.0031	1.43
0.1046	0.2029	6.69	0.2105	0.2133	6.68	0.2110	0.2106	0.01	0.1	0.0005	0.24	0.0027	1.27
0.7758	0.1157	12.55	0.8600	0.0780	12.58	0.8641	0.0746	0.03	0.2	0.0041	0.48	0.0034	4.36
0.5530	0.1985	10.69	0.7046	0.1425	10.66	0.7079	0.1406	0.02	0.2	0.0033	0.45	0.0019	1.33
0.4583	0.2006	9.63	0.5972	0.1815	9.82	0.6320	0.1506	0.19	2.0	0.0348	5.83	0.0309	17.02
0.3304	0.2120	8.75	0.5078	0.1769	8.71	0.5108	0.1746	0.04	0.5	0.0030	0.59	0.0023	1.30
0.5535	0.2965	10.77	0.6948	0.2128	10.79	0.7014	0.2070	0.01	0.1	0.0066	0.95	0.0058	2.73
0.4852	0.3459	10.25	0.6411	0.2529	10.24	0.6441	0.2494	0.01	0.1	0.0030	0.47	0.0035	1.38
0.2599	0.3689	8.36	0.4165	0.3173	8.28	0.4207	0.3122	0.08	1.1	0.0042	1.01	0.0051	1.61
0.2072	0.3029	7.45	0.3564	0.2750	7.74	0.3590	0.2731	0.28	4.2	0.0026	0.72	0.0019	0.64
0.0758	0.3680	6.61	0.1473	0.3825	6.65	0.1527	0.3767	0.04	0.6	0.0054	3.66	0.0058	1.52
0.0374	0.0460	6.45	0.0806	0.4797	6.42	0.0779	0.4819	0.03	0.3	0.0027	3.35	0.0022	0.46
0.4381	0.3883	9.92	0.5981	0.2899	9.87	0.6013	0.2865	0.05	0.6	0.0032	0.32	0.0036	1.24
0.3329	0.4550	9.04	0.4917	0.3589	9.02	0.4969	0.3576	0.02	0.2	0.0052	1.06	0.0013	0.36
0.3581	0.4080	9.20	0.5255	0.3152	9.19	0.5251	0.3171	0.02	0.2	0.0004	0.08	0.0019	0.60
0.1669	0.2950	7.56	0.2938	0.4176	7.57	0.2950	0.4156	0.01	0.1	0.0012	0.41	0.0020	0.48
0.3160	0.5560	9.03	0.4435	0.4663	8.98	0.4749	0.4360	0.05	0.6	0.0314	7.08	0.0303	6.50
0.2321	0.6109	8.36	0.3758	0.5076	8.30	0.3742	0.5081	0.06	0.7	0.0004	0.11	0.0005	0.10
0.1982	0.5659	8.01	0.3311	0.4885	7.96	0.3340	0.4881	0.05	0.6	0.0029	0.88	0.0036	0.74
0.1244	0.7790	7.55	0.2262	0.6953	7.51	0.2236	0.6982	0.04	0.5	0.0026	1.15	0.0029	0.42

TABLE A-I. 24

Comparison of Calculated and Experimental Results for the System Nitrogen (1) - Argon (2) - Oxygen (3) at 110.0°K

Experimental (83)					Clausius			Deviations					
								ΔP		Δy_1		Δy_2	
x_1	x_2	P, atm.	y_1	y_2	P, atm.	y_1	y_2	atm.	%	mol. fr.	%	mol. fr.	%
0.0434	0.0307	6.00	0.1037	0.0379	5.84	0.1023	0.0379	0.16	2.7	0.0014	1.35	0.0	0.0
0.0408	0.0474	6.00	0.0965	0.0583	5.85	0.0959	0.0582	0.15	2.5	0.0006	0.62	0.0001	0.17
0.0261	0.0623	6.00	0.0879	0.0750	5.74	0.0652	0.0777	0.26	4.3	0.0254		0.0027	3.60
0.0354	0.0794	6.00	0.0819	0.0953	5.86	0.0829	0.0968	0.14	2.3	0.0010	1.20	0.0015	1.57
0.0286	0.1000	6.00	0.0665	0.1243	5.83	0.0672	0.1220	0.17	2.8	0.0007	1.05	0.0023	1.85
0.0190	0.1561	6.00	0.0441	0.1941	5.84	0.0444	0.1883	0.16	2.7	0.0003	0.68	0.0058	2.98
0.0440	0.0191	6.00	0.1039	0.0234	5.82	0.1041	0.0236	0.18	3.0	0.0002	0.19	0.0003	1.28
0.0368	0.0636	6.00	0.0874	0.0769	5.84	0.0865	0.0780	0.16	2.7	0.0009	1.03	0.0011	1.43
0.0464	0.0120	6.00	0.1097	0.0151	5.83	0.1097	0.0149	0.17	2.8	0.00	0.0	0.0002	1.32
0.2869	0.0336	8.00	0.4816	0.0304	8.11	0.4835	0.0306	0.11	1.4	0.0019	0.39	0.0002	0.65
0.2853	0.2853	8.00	0.4779	0.0488	8.13	0.4792	0.0481	0.13	1.6	0.0013	0.27	0.0007	1.46
0.2829	0.0703	8.00	0.4729	0.0620	8.13	0.4744	0.0635	0.13	1.6	0.0015	0.32	0.0015	2.42
0.2796	0.0859	8.00	0.4672	0.0785	8.13	0.4688	0.0775	0.13	1.6	0.0015	0.32	0.0010	1.27
0.2749	0.1214	8.00	0.4572	0.1091	8.14	0.4594	0.1087	0.14	1.8	0.0022	0.48	0.0004	0.37
0.2656	0.1761	8.00	0.4403	0.1577	8.14	0.4430	0.1565	0.14	1.8	0.0027	0.61	0.0012	0.76
0.2154	0.4646	8.00	0.3615	0.4007	8.08	0.3617	0.4015	0.08	1.0	0.0002	0.06	0.0009	0.22
0.1971	0.6106	8.00	0.3343	0.5191	8.07	0.3334	0.5211	0.07	0.9	0.0009	0.27	0.0020	0.39
0.1842	0.7776	8.00	0.3102	0.6603	8.11	0.3153	0.6549	0.11	1.4	0.0051	1.64	0.0054	0.82
0.2801	0.0685	8.00	0.4706	0.0621	8.11	0.4714	0.0621	0.11	1.4	0.0008	0.17	0.0	0.0
0.2901	0.0201	8.00	0.4850	0.0189	8.12	0.4889	0.0183	0.12	1.5	0.0039	0.80	0.0006	3.17
0.5152	0.0624	10.00	0.6903	0.0464	10.22	0.6933	0.0468	0.22	2.2	0.0030	0.43	0.0004	0.86
0.5171	0.0566	10.00	0.6926	0.0430	10.23	0.6954	0.0425	0.23	2.3	0.0028	0.40	0.0005	1.16
0.5154	0.0722	10.00	0.6888	0.0547	10.24	0.6923	0.0540	0.24	2.4	0.0035	0.51	0.0007	1.28
0.5117	0.0890	10.00	0.6850	0.0668	10.23	0.6875	0.0665	0.23	2.3	0.0025	0.36	0.0003	0.45
0.5054	0.1278	10.00	0.6753	0.0949	10.23	0.6785	0.0950	0.23	2.3	0.0032	0.47	0.0001	0.11
0.4974	0.1828	10.00	0.6644	0.1363	10.24	0.6668	0.1350	0.24	2.4	0.0024	0.36	0.0013	0.95
0.5209	0.0117	10.00	0.7017	0.0092	10.20	0.7038	0.0089	0.20	2.0	0.0021	0.30	0.0003	3.26
0.5195	0.0317	10.00	0.6955	0.0243	10.22	0.7002	0.0239	0.22	2.2	0.0048	0.69	0.0004	1.65

TABLE A-I. 24 (cont'd)

Experimental[83]					Clausius			Deviations					
x_1	x_2	P, atm.	y_1	y_2	P, atm.	y_1	y_2	ΔP		Δy_1		Δy_2	
								atm.	%	mol. fr.	%	mol. fr.	%
0.5163	0.0263	10.00	0.6930	0.0198	10.19	0.6984	0.0198	0.10	1.9	0.0054	0.78	0.00	0.0
0.5092	0.0500	10.00	0.6859	0.0375	10.15	0.6900	0.0378	0.15	1.5	0.0041	0.60	0.0003	0.80
0.4730	0.3088	10.00	0.6337	0.2275	10.19	0.6372	0.2257	0.19	1.9	0.0035	0.55	0.0018	0.79
0.4624	0.3994	10.00	0.6243	0.2918	10.21	0.6233	0.2890	0.21	2.1	0.0010	0.16	0.0028	0.95
0.5209	0.0104	10.00	0.7001	0.0078	10.20	0.7040	0.0078	0.20	2.0	0.0039	0.56	0.00	0.0
0.7024	0.0614	12.00	0.8192	0.0423	11.94	0.8240	0.0414	0.06	0.5	0.0048	0.59	0.0009	2.13
0.6954	0.1093	12.00	0.8097	0.0749	11.94	0.8151	0.0733	0.06	0.5	0.0054	0.67	0.0016	2.13
0.6886	0.1510	12.00	0.8019	0.1031	11.94	0.8072	0.1008	0.06	0.5	0.0053	0.67	0.0023	2.23
0.6788	0.2164	12.00	0.7903	0.1476	11.94	0.7961	0.1435	0.06	0.5	0.0058	0.73	0.0041	2.77
0.7069	0.0190	12.00	0.8265	0.0126	11.92	0.8312	0.0129	0.08	0.7	0.0047	0.56	0.0003	2.38
0.7021	0.0435	12.00	0.8230	0.0300	11.91	0.8256	0.0294	0.09	0.8	0.0026	0.32	0.0006	2.0
0.6730	0.2711	12.00	0.7845	0.1825	11.97	0.7890	0.1786	0.03	0.3	0.0045	0.57	0.0039	2.13

TABLE A-I. 25

Comparison of Calculated and Experimental Results for the System Nitrogen (1) - Argon (2) - Oxygen (3) at 110.0°K

Experimental (83)					R-K			Deviations					
								ΔP		Δy_1		Δy_2	
x_1	x_2	P, atm.	y_1	y_2	P, atm.	y_1	y_2	atm.	%	mol. fr.	%	mol. fr.	%
0.0434	0.0307	6.00	0.1037	0.0379	5.84	0.1019	0.0370	0.16	2.67	0.0018	1.74	0.0009	2.37
0.0408	0.0474	6.00	0.0965	0.0583	5.84	0.0955	0.0569	0.16	2.67	0.0010	1.04	0.0014	2.40
0.0261	0.0623	6.00	0.0879	0.0750	5.73	0.0623	0.0760	0.27	4.50	0.0256	29.12	0.0010	1.33
0.0354	0.0794	6.00	0.0819	0.0953	5.85	0.0826	0.0948	0.15	2.50	0.0007	0.85	0.0005	0.52
0.0286	0.1000	6.00	0.0665	0.1243	5.82	0.0700	0.1196	0.18	3.00	0.0005	0.75	0.0047	3.78
0.0190	0.1561	6.00	0.0441	0.1941	5.82	0.443	0.1852	0.18	3.00	0.0002	0.45	0.0089	4.59
0.0440	0.0191	6.00	0.1039	0.0234	5.82	0.1036	0.0231	0.18	3.00	0.0003	0.29	0.0003	1.28
0.0368	0.0636	6.00	0.0874	0.0769	5.83	0.0862	0.0763	0.17	2.83	0.0012	1.49	0.0006	2.78
0.0464	0.0120	6.00	0.1097	0.0151	5.83	0.1092	0.0145	0.17	2.83	0.0005	0.46	0.0006	3.97
0.2869	0.0336	8.00	0.4816	0.0304	8.08	0.4817	0.0301	0.08	1.00	0.0001	0.02	0.0003	0.99
0.2853	0.0531	8.00	0.4779	0.0488	8.09	0.4776	0.0473	0.09	1.13	0.0003	0.06	0.0015	3.07
0.2829	0.0703	8.00	0.4729	0.0620	8.09	0.4729	0.0625	0.09	1.13	0.0	0.0	0.0005	0.81
0.2796	0.0859	8.00	0.4672	0.0785	8.09	0.4674	0.0763	0.09	1.13	0.0002	0.04	0.0022	2.80
0.2749	0.1214	8.00	0.4572	0.1091	8.10	0.4583	0.1073	0.10	1.25	0.0011	0.24	0.0018	1.65
0.2656	0.1761	8.00	0.4403	0.1577	8.09	0.4421	0.1548	0.09	1.13	0.0018	0.41	0.0029	1.84
0.2154	0.4646	8.00	0.3615	0.4007	8.00	0.3605	0.4017	0.0	0.0	0.0010	0.28	0.0010	0.25
0.1971	0.6106	8.00	0.3343	0.5191	7.99	0.3311	0.5235	0.01	0.13	0.0032	0.96	0.0044	0.85
0.1842	0.7776	8.00	0.3102	0.6603	8.03	0.3106	0.6599	0.03	0.38	0.0004	0.13	0.0004	0.06
0.2801	0.0685	8.00	0.4706	0.0621	8.07	0.4699	0.0611	0.07	0.88	0.0007	0.15	0.0010	1.61
0.2901	0.0201	8.00	0.4850	0.0189	8.09	0.4869	0.0180	0.09	1.13	0.0019	0.39	0.0009	4.76
0.5152	0.0624	10.00	0.6903	0.0464	10.15	0.6916	0.0463	0.15	1.50	0.0013	0.19	0.0001	0.22
0.5171	0.0566	10.00	0.6926	0.0430	10.16	0.6938	0.0420	0.16	1.60	0.0012	0.17	0.0010	2.33
0.5154	0.0722	10.00	0.6888	0.0547	10.16	0.6908	0.0535	0.16	1.60	0.0020	0.29	0.0012	2.19
0.5117	0.0890	10.00	0.6850	0.0668	10.15	0.6861	0.0659	0.15	1.50	0.0011	0.16	0.0010	1.50
0.5054	0.1278	10.00	0.6573	0.0949	10.15	0.6773	0.0943	0.15	1.50	0.0020	0.30	0.0006	0.63
0.4974	0.1828	10.00	0.6644	0.1363	10.15	0.6659	0.1342	0.15	1.50	0.0015	0.23	0.0021	1.54
0.5209	0.0117	10.00	0.7017	0.0092	10.13	0.7017	0.0087	0.13	1.30	0.00	0.0	0.0005	5.43
0.5195	0.0317	10.00	0.6955	0.0243	10.15	0.6984	0.0236	0.15	1.50	0.0029	0.42	0.0007	2.88

TABLE A-I. 25 (cont'd)

Experimental (83)					R-K			Deviations					
								ΔP		Δy_1		Δy_2	
x_1	x_2	P, atm.	y_1	y_2	P, atm.	y_1	y_2	atm.	%	mol. fr.	%	mol. fr.	%
0.5163	0.0263	10.00	0.6930	0.0198	10.11	0.6965	0.0196	0.11	1.10	0.0035	0.51	0.0002	1.01
0.5092	0.0500	10.00	0.6859	0.0375	10.08	0.6883	0.0373	0.08	0.8	0.0024	0.35	0.0002	0.53
0.4730	0.3088	10.00	0.6337	0.2275	10.09	0.6362	0.2255	0.09	0.9	0.0025	0.39	0.0020	0.88
0.4624	0.3994	10.00	0.6243	0.2918	10.10	0.6219	0.2898	0.10	1.0	0.0024	0.38	0.0020	0.69
0.5209	0.0140	10.00	0.7001	0.0078	10.13	0.7019	0.0078	0.13	1.3	0.0018	0.26	0.0	0.0
0.7024	0.0641	12.80	0.8192	0.0423	11.83	0.8229	0.0411	0.17	1.42	0.0037	0.45	0.0019	4.49
0.6954	0.1093	12.00	0.8097	0.0749	11.83	0.8143	0.0729	0.17	1.42	0.0046	0.57	0.0020	2.67
0.6886	0.1510	12.00	0.8019	0.1031	11.82	0.8060	0.1005	0.18	1.50	0.0047	0.59	0.0026	2.52
0.6788	0.2164	12.00	0.7903	0.1476	11.82	0.7955	0.1434	0.18	1.50	0.0052	0.66	0.0042	2.85
0.7069	0.0190	12.30	0.8265	0.0126	11.82	0.8298	0.0128	0.18	1.50	0.0033	0.40	0.0002	1.59
0.7021	0.0435	12.00	0.8230	0.0300	11.81	0.8244	0.0292	0.19	1.58	0.0014	0.17	0.0008	2.67
0.6730	0.2711	12.00	0.7845	0.1825	11.83	0.7883	0.1790	0.17	1.42	0.0038	0.48	0.0035	1.92

TABLE A-I. 26

Comparison of Calculated and Experimental Results for the System Argon (4) - Methane (2) at 115. 22°K

k_{ij} = 0.025

Experimental				Calculated		Deviations	
P, atm.	x_1	y_1	y_1	$\ln \gamma_1$	$\ln \gamma_2$	$\lvert \Delta y_1 \rvert$ mol. fr.	%
1.87	0.0551	0.3107	0.3101	0.2828	0.0	0.0006	0.19
3.58	0.2492	0.6930	0.6934	0.1954	0.0169	0.0004	0.06
4.06	0.3213	0.7549	0.7543	0.1650	0.0289	0.0006	0.08
5.56	0.5075	0.8532	0.8536	0.0951	0.0793	0.0004	0.05
7.45	0.7780	0.9410	0.9405	0.0221	0.2128	0.0005	0.05
8.76	0.9561	0.9889	0.9880	0.0010	0.3523	0.0004	0.09

TABLE A-I. 27

Comparison of Calculated and Experimental Results for the System Nitrogen (1) - Argon (2) at 100.0°K

$k_{ij} = 0.0$

Experimental				Calculated		Deviations	
P, atm.	x_1	y_1	y_1	$\ln \gamma_1$	$\ln \gamma_2$	$\lvert \Delta y_1 \rvert$ mol. fr.	%
3.89	0.1389	0.2780	0.2789	0.0970	0.0022	0.0009	0.32
5.00	0.3792	0.5771	0.5768	0.0467	0.0201	0.0007	0.12
5.36	0.4613	0.6518	0.6512	0.0346	0.0295	0.0006	0.09
5.65	0.5287	0.7066	0.7060	0.0258	0.0378	0.0006	0.08
6.28	0.6721	0.8083	0.8087	0.0119	0.0585	0.0004	0.05
6.60	0.7459	0.8554	0.8559	0.0074	0.0710	0.0005	0.06
7.27	0.8999	0.9469	0.9460	0.0011	0.0987	0.0009	0.10

TABLE A-I. 28

Vapor-Liquid Equilibrium Data for the System Ethylene(1)-Propane(2)

$k_{ij} = 0.0$

P, atm.	x_1	y_1	ln γ_1	ln γ_2
		T = 273.06 °K		
7.72	0.0996	0.4101	0.1898	0.0004
10.53	0.1905	0.5818	0.1479	0.0025
11.79	0.2308	0.6329	0.1410	0.0043
15.24	0.3402	0.7316	0.1190	0.0133
17.66	0.4157	0.7793	0.1017	0.0239
20.13	0.4915	0.3175	0.0334	0.0391
22.00	0.5466	0.3410	0.0700	0.0536
24.53	0.6212	0.3693	0.0524	0.0784
25.96	0.6617	0.8832	0.0433	0.0946
29.13	0.7478	0.9111	0.0259	0.0136
32.15	0.8238	0.9350	0.0135	0.1823
35.41	0.8980	0.9593	0.0047	0.2363
38.69	0.9635	0.9836	0.0006	0.2921
		255.38 °K		
3.77	0.0540	0.3198	0.1393	0.6003
8.08	0.2540	0.7147	0.1021	0.0070
11.27	0.4000	0.8169	0.0730	0.0215
11.82	0.4250	0.8294	0.0685	0.0249
12.67	0.4630	0.8466	0.0613	0.0307
12.82	0.4702	0.8495	0.0599	0.0318
17.09	0.6565	0.9127	0.0282	0.0732
19.37	0.7505	0.9377	0.0157	0.1030
24.78	0.9490	0.9865	0.0007	0.1895
		227.9°K		
2.99	0.1856	0.7444	0.1326	0.0046
4.00	0.2755	0.8226	0.1113	0.0110
5.43	0.4065	0.8858	0.0808	0.0269
7.07	0.5577	0.9284	0.0488	0.0569
9.15	0.7485	0.9636	0.0174	0.1167
11.34	0.9355	0.9909	0.0010	0.2042

TABLE A-I. 28 (cont'd)

T = 199.83°K

P, atm.	x_1	y_1	ln γ_1	ln γ_2
1.41	0.2505	0.8890	0.1743	0.0126
1.64	0.3025	0.9098	0.1565	0.0194
1.84	0.3469	0.9233	0.1414	0.0267
2.56	0.522	0.9567	0.0847	0.0708
3.66	0.7999	0.9851	0.0173	0.2656
4.13	0.9159	0.9924	0.0033	0.2913

TABLE A-I. 29

R-K Equation of State Parameters

	T, °K	Ω_a	Ω_b
Argon	90.5	0.40582	0.08702
	100.0	0.41063	0.08634
	110.0	0.41316	0.08541
	115.2	0.41296	0.08465
	120.0	0.41399	0.08426
Nitrogen	90.5	0.42325	0.08607
	100	0.42088	0.08457
	110	0.41397	0.08247
	110.05	0.413797	0.08242
	110.94	0.41331	0.08224
	120	0.40826	0.08026
Oxygen	70.0	0.38983	0.08659
	80.0	0.40077	0.08666
	90.0	0.40850	0.08648
	90.5	0.41029	0.08653
	100.0	0.41358	0.08605
	110.0	0.41606	0.08531
	120.0	0.41618	0.08427
	130.0	0.41843	0.08410
	140.0	0.41004	0.08131
Methane	110.94	0.40324	0.08649
	115.20	0.40541	0.08632
Carbon dioxide	277.5	0.40737	0.07776
	283.0	0.40453	0.07751
	288.6	0.40110	0.07721
	294.27	0.39792	0.07701
	299.8	0.41730	0.08105
Hydrogen sulfide	277.5	0.42311	0.08319
	283.0	0.42216	0.08290
	288.6	0.41840	0.08239
	294.3	0.42129	0.08239
	299.	0.41872	0.08204

TABLE A-I. 30

Critical Constants and Acentric Factor of Pure Fluids

Component	Pc, atm	Vc, ml/g-mole	Tc, °K	ω
Methane	45.8	98.72	191.06	0.013
Ethylene	49.98	127.64	282.66	0.085
Ethane	48.50	141.72	305.50	0.105
Propane	42.10	195.70	370.00	0.152
n-Butane	37.47	254.71	425.17	0.201
Carbon dioxide	72.90	94.00	304.20	0.225
Hydrogen sulfide	88.90	97.70	373.60	0.100
Nitrogen	33.5	90.09	126.2	0.04
Oxygen	49.77	73.38	154.58	0.021
Argon	48.0	75.2	150.72	-0.002

TABLE A-I. 31

Binary Interaction Constants k_{12}

System		k_{12}				
		This work	Hsi and Lu[(43)]	Chueh and Prausnitz[(62)]	Hiza et al.[(90)]	
(1)	(2)				Eq. 5	Eq. 7
Nitrogen	- Argon	0.0	0.0	0.0	0.005	0.0
Nitrogen	- Oxygen	0.0016	0.0014		0.007	0.064
Nitrogen	- Methane	0.025	0.026	0.03		0.046
Argon	- Oxygen	0.0077	0.0104	0.010	0.007	0.06
Argon	- Methane	0.0184	0.0229	0.02	0.013	0.043
Carbon dioxide	- Hydrogen sulfide	0.07	0.07	0.08		0.1106
Carbon dioxide	- n-propane	0.0762	0.95	0.11 ± .01		0.0764
Hydrogen sulfide	- n-butane	0.048	0.062	0.09		0.004

APPENDIX II

Tables A-II. 1 to A-II. 28 for Chapter V

TABLE A-II. 1

Comparison of Calculated and Experimental γ Values for the System Nitrogen - Argon at 90. 5°K

		γ_{N_2}		γ_A	
P_{psia}	x_{N_2}	Expt (75)	Calc	Expt (75)	Calc
20.98	0.0197	0.9964	1.000	1.1709	1.1723
22.33	0.0496	1.0014	1.0003	1.1781	1.1618
24.09	0.0967	1.0065	1.0015	1.1210	1.1445
27.00	0.1688	1.0103	1.0047	1.1242	1.1208
29.36	0.2344	1.0137	1.0090	1.1022	1.1019
32.47	0.3199	1.0245	1.0170	1.0827	1.0800
32.63	0.3268	1.0224	1.0182	1.0788	1.0779
36.25	0.4344	1.0383	1.0315	1.0530	1.0544
39.68	0.5353	1.0517	1.0481	1.0403	1.0367
40.93	0.5811	1.0629	1.0576	1.0256	1.0294
43.78	0.6657	1.0814	1.0757	1.0190	1.0182
43.99	0.6718	1.0932	1.0779	1.0168	1.0182
46.77	0.7569	1.1064	1.0997	1.0120	1.0101
47.65	0.7873	1.1178	1.1086	1.0077	1.0074
49.95	0.8580	1.1376	1.1298	1.0050	1.0033
50.58	0.8764	1.1673	1.1354	1.0036	1.0025
52.34	0.9340	1.1474	1.1560	1.0020	1.0006

TABLE A-II. 2

Comparison of Calculated and Experimental γ Values for the System Nitrogen - Argon at 100°K

		γ_{N_2}		γ_A	
P_{psia}	x_{N_2}	Expt $^{(75)}$	Calc	Expt $^{(75)}$	Calc
48.56	0.0195	1.0001	1.0001	1.1635	1.1624
50.90	0.0480	1.0005	1.0004	1.1518	1.1510
53.07	0.0813	1.0012	1.0012	1.1389	1.1385
57.40	0.1400	1.0035	1.0034	1.1182	1.1181
58.58	0.1565	1.0044	1.0043	1.1128	1.1128
62.87	0.2222	1.0086	1.0034	1.0930	1.0931
68.98	0.3092	1.0161	1.0160	1.0706	1.0707
72.12	0.3618	1.0217	1.0217	1.0590	1.0590
75.84	0.4142	1.0281	1.0281	1.0487	1.0486
77.79	0.4421	1.0318	1.0318	1.0437	1.0436
82.27	0.5169	1.0426	1.0427	1.0319	1.0317
85.45	0.5703	1.0512	1.0514	1.0248	1.0245
90.69	0.6483	1.0650	1.0653	1.0163	1.0159
91.92	0.6597	1.0672	1.0675	1.0152	1.0148
92.27	0.7485	1.0849	1.0852	1.0082	1.0078
98.28	0.7691	1.0893	1.0896	1.0069	1.0065
99.45	0.7888	1.0936	1.0938	1.0057	1.0054
100.20	0.7982	1.0957	1.0959	1.0052	1.0049
101.91	0.8230	1.1012	1.1014	1.0041	1.0038
103.47	0.8514	1.1078	1.1078	1.0029	1.0026
104.78	0.8685	1.1119	1.1118	1.0023	1.0020
105.93	0.8903	1.1171	1.1169	1.0016	1.0014
106.83	0.9001	1.1195	1.1192	1.0014	1.0012
106.69	0.9444	1.1307	1.1300	1.0006	1.0004
109.56	0.9470	1.1313	1.1306	1.0005	1.0003

TABLE A-II. 3

Comparison of Calculated and Experimental γ Values for the System Nitrogen - Argon at 110. 0°K

		γ_{N_2}		γ_A	
P_{psia}	x_{N_2}	Expt (75)	Calc	Expt (75)	Calc
113.93	0.1365	1.0091	1.0024	1.1030	1.1030
122.89	0.2134	1.0101	1.0060	1.0366	1.0854
123.31	0.2156	1.0095	1.0061	1.0910	1.0844
138.39	0.3469	1.0203	1.0161	1.0591	1.0576
153.18	0.4785	1.0322	1.0304	1.0383	1.0366
155.74	0.4959	1.0437	1.0336	1.0354	1.0346
156.60	0.4988	1.0401	1.0337	1.0413	1.0336
156.31	0.5021	1.0373	1.0339	1.0375	1.0328
171.81	0.6472	1.0644	1.0566	1.0151	1.0161
186.46	0.7750	1.0936	1.0822	1.0057	1.0070
192.58	0.8296	1.0952	1.0953	1.0040	1.0040
198.13	0.8747	1.1098	1.1063	1.0032	1.0020
199.12	0.8854	1.1235	1.1085	1.0012	1.0017
205.95	0.9442	1.1319	1.1252	1.0005	1.0004

TABLE A-II. 4

Comparison of Calculated and Experimental γ Values for the System Nitrogen - Argon at 120.0°K

P_{psia}	x_{N_2}	γ_{N_2} Expt (75)	γ_{N_2} Calc	γ_A Expt (75)	γ_A Calc
199.69	0.1275	1.0014	1.0020	1.1086	1.0030
216.05	0.2169	1.0069	1.0060	1.0798	1.0822
216.47	0.2201	1.0067	1.0061	1.0782	1.0811
237.52	0.3352	1.0183	1.0141	1.0531	1.0586
260.14	0.4536	1.0306	1.0274	1.0413	1.0398
261.55	0.4661	1.0314	1.0284	1.0376	1.0377
265.26	0.4829	1.0325	1.030	1.0375	1.0346
293.14	0.6316	1.0596	1.0523	1.0200	1.0182
315.89	0.7603	1.0770	1.0779	1.0084	1.0070
336.09	0.8677	1.1063	1.1019	1.0015	1.0020
338.65	0.8800	1.1293	1.1052	0.9996	1.0018

TABLE A-II. 5

Comparison of Calculated and Experimental γ Values for the System Oxygen - Argon at 90.5°K

		γ_{O_2}		γ_A	
P_{psia}	x_{O_2}	Expt(117)	Calc	Expt(117)	Calc
19.99	0.100	1.0004	1.0017	1.1585	1.1505
19.74	0.200	1.0078	1.0068	1.1248	1.1179
19.43	0.300	1.0182	1.0154	1.0982	1.0896
19.27	0.400	1.0432	1.0277	1.0854	1.0655
18.56	0.500	1.0463	1.0440	1.0508	1.0453
18.03	0.600	1.0662	1.0643	1.0336	1.0289
18.47	0.700	1.0922	1.0891	1.0216	1.0163
16.83	0.800	1.1141	1.1186	1.0166	1.0072
16.17	0.900	1.1623	1.1534	1.0133	1.0018

TABLE A-II. 6

Comparison of Calculated and Experimental γ Values for the System Oxygen - Argon at 100.0°K

		γ_{O_2}		γ_A	
P_{psia}	x_{O_2}	Expt (117)	Calc	Expt (117)	Calc
46.36	0.100	0.9971	1.0014	1.1303	1.1257
45.84	0.200	1.0022	1.0056	1.1024	1.0992
45.20	0.300	1.0104	1.0128	1.0784	1.0759
44.41	0.400	1.0208	1.0231	1.0571	1.0559
43.44	0.500	1.0333	1.0368	1.0388	1.0389
42.38	0.600	1.0487	1.0540	1.0248	1.0250
41.22	0.700	1.0692	1.0751	1.0138	1.0141
39.93	0.800	1.0936	1.1003	1.0062	1.0063
38.33	0.900	1.1172	1.1301	0.9983	1.0016

TABLE A-II. 7

Comparison of Calculated and Experimental γ Values for the System Oxygen - Argon at 110.0°K

P_{psia}	x_{O_2}	γ_{O_2} Expt (117)	γ_{O_2} Calc	γ_A Expt (117)	γ_A Calc
96.02	0.100	1.0038	1.0013	1.1148	1.1093
95.09	0.200	1.0078	1.0053	1.0903	1.0848
93.93	0.300	1.0142	1.0118	1.0699	1.0639
92.50	0.400	1.0226	1.0200	1.0519	1.0463
90.73	0.500	1.0325	1.0327	1.0354	1.0317
88.80	0.600	1.0507	1.0472	1.0185	1.0200
86.73	0.700	1.0628	1.0643	1.0131	1.0112
84.42	0.800	1.0822	1.0843	1.0067	1.0049
82.08	0.900	1.1008	1.1071	1.0054	1.0012

TABLE A-II. 8

Comparison of Calculated and Experimental γ Values for the System Oxygen - Argon at 120°K

		γ_{O_2}		γ_A	
P_{psia}	x_{O_2}	Expt (117)	Calc	Expt (117)	Calc
176.13	0.100	1.0088	1.0011	1.1042	1.0913
174.68	0.200	1.0120	1.0045	1.0830	1.0708
172.81	0.300	1.0218	1.0100	1.0521	1.0532
170.50	0.400	1.0240	1.0178	1.0491	1.0385
167.64	0.500	1.0324	1.0276	1.0345	1.0263
164.49	0.600	1.0431	1.0396	1.0232	1.0166
161.19	0.700	1.0579	1.0538	1.0148	1.0092
157.53	0.800	1.0750	1.0701	1.0091	1.0040
152.87	0.900	1.09	1.0887	1.0027	1.0010

TABLE A-II. 9

Comparison of Calculated and Experimental γ Values for the System Nitrogen - Oxygen at 90. 5°K

		γ_{N_2}		γ_A	
P_{psia}	x_{N_2}	Expt (44)	Calc	Expt (44)	Calc
17.25	0.9458	1.1056	1.0010	0.9883	1.2956
19.91	0.8900	1.1135	1.0040	1.0069	1.2586
22.78	0.8241	1.1163	1.0090	1.0088	1.2361
23.00	0.8212	1.1205	1.0091	1.0129	1.2165
23.53	0.8060	1.1089	1.0111	1.0122	1.2068
26.57	0.7287	1.0868	1.0212	1.0151	1.1653
31.27	0.6097	1.0641	1.0450	1.0272	1.1140
33.36	0.5511	1.0465	1.0597	1.0359	1.0920
35.29	0.5043	1.0391	1.0736	1.0604	1.0768
36.12	0.4931	1.0467	1.0768	1.0838	1.0736
36.62	0.4964	1.0338	1.0855	1.0707	1.0661
41.84	0.3521	1.0407	1.1298	1.1034	1.0359
49.10	0.1626	1.0247	1.2251	1.1623	1.0076

TABLE A-II.10

Comparison of Calculated and Experimental γ Values for the System Oxygen - Nitrogen at 99.94°K

		γ_{O_2}		γ_{N_2}	
P psia	x_{O_2}	Expt (44)	Calc	Expt (44)	Calc
40.11	0.9500	1.2110	1.2099	1.0006	1.0006
42.66	0.9299	1.1997	1.1990	1.0012	1.0012
44.41	0.9005	1.841	1.1838	1.0025	1.0024
47.85	0.8640	1.1660	1.1661	1.0045	1.0044
50.73	0.8209	1.1464	1.1467	1.0077	1.0075
56.51	0.7488	1.0428	1.0426	1.0593	1.0596
69.95	0.5752	1.0641	1.0642	1.0403	1.0403
74.11	0.5125	1.0496	1.0495	1.0523	1.0524
82.64	0.4103	1.0307	1.0307	1.0748	1.0753
85.84	0.3624	1.0236	1.0232	1.0866	1.0872
97.58	0.1944	1.0066	1.0062	1.1346	1.1352
105.52	0.0914	1.0016	1.0016	1.1681	1.1688

TABLE A-II. 11

Comparison of Calculated and Experimental γ Values for the System Oxygen - Nitrogen at 110.05°K

		γ_{O_2}		γ_{N_2}	
P_{psia}	x_{O_2}	Expt (44)	Calc	Expt (44)	Calc
88.54	0.9348	1.1655	1.1445	0.9957	1.0010
97.46	0.8734	1.0292	1.1252	1.0056	1.0020
107.00	0.7983	1.0960	1.1030	1.0034	1.0060
135.75	0.5831	1.0563	1.0534	1.0315	1.0274
157.56	0.4192	1.0303	1.0274	1.0678	1.0408
163.55	0.3691	1.0215	1.0212	1.0756	1.0629
185.92	0.2018	1.0059	1.0060	1.1291	1.1030
200.79	0.0955	1.0022	1.0010	1.1610	1.1343

TABLE A-II. 12

Comparison of Calculated and Experimental γ Values for the System Oxygen - Nitrogen at 119.92°K

		γ_{O_2}		γ_{N_2}	
P_{psia}	x_{O_2}	Expt (44)	Calc	Expt (44)	Calc
174.04	0.8798	1.1276	1.1096	1.0022	1.0020
189.18	0.8082	1.1153	1.0920	1.0056	1.0050
213.50	0.6910	1.0756	1.0661	1.0254	1.0131
234.43	0.5889	1.0568	1.0471	1.0388	1.0233
264.95	0.4474	1.0388	1.0274	1.0644	1.0408
277.75	0.3751	1.0248	1.0192	1.0701	1.0534
315.54	0.2100	1.0108	1.0060	1.1197	1.0876
337.39	0.1162	1.0035	1.0020	1.1649	1.1107

TABLE A-II-13. A

TESTED DATA OF OXYGEN ARGON NITROGEN AT 90.5 K NARNISIKI DATA

ID	P, atm	x_1	x_2	y_3	y_1	y_2	y_3
1	3.5	0.0990	0.0373	0.8637	0.0389	0.0194	0.9417
2	3.2	0.1512	0.0868	0.7620	0.0616	0.0466	0.8918
3	3.0	0.2296	0.0861	0.6843	0.0869	0.0482	0.8550
4	2.5	0.4202	0.1122	0.4676	0.2041	0.0747	0.7212
5	2.1	0.4965	0.1704	0.3331	0.2674	0.1286	0.6040
6	2.0	0.5931	0.1179	0.2849	0.3406	0.0963	0.5631
7	1.7	0.6089	0.2033	0.1878	0.3927	0.1955	0.4218
8	1.6	0.6754	0.2046	0.1200	0.4805	0.2079	0.3116
9	1.4	0.7388	0.1924	0.0688	0.5793	0.2195	0.2011
10	1.4	0.7599	0.1698	0.0703	0.5970	0.1951	0.2079
11	2.6	0.2939	0.2049	0.5012	0.1368	0.1294	0.7338
12	2.2	0.4212	0.2271	0.3517	0.2233	0.1623	0.6144
13	1.7	0.5892	0.2554	0.1554	0.3932	0.2408	0.3660
14	2.8	0.1322	0.2754	0.5914	0.0585	0.1565	0.7850
15	2.3	0.3257	0.3060	0.3683	0.1706	0.2111	0.6183
16	2.3	0.1952	0.4317	0.3731	0.1023	0.2853	0.6124
17	2.4	0.2175	0.3887	0.3938	0.1099	0.2552	0.6349
18	1.8	0.4013	0.4172	0.1815	0.2573	0.3514	0.3913
19	1.9	0.2294	0.5548	0.2158	0.1400	0.4276	0.4324
20	2.1	0.1517	0.5977	0.2506	0.0905	0.4338	0.4757
21	1.8	0.0938	0.7677	0.1385	0.0645	0.6247	0.3108

TABLE A-II-13.B

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 90.5 K NARNISIKI DATA

EX= 0.0005 FP= 1.0000 ET= 0.2000

PAIR NO.	P(av.)	I	II	III	D	DMAX	DX	DP	DT
1- 2	49.1	0.1281	0.1487	-0.0026	-0.0180	0.1010	0.0181	0.0829	0.2646
2- 3	45.8	0.0738	0.1957	-0.0017	-0.0302	0.1056	0.0168	0.0887	0.2646
3- 4	40.4	0.4391	0.3720	-0.0049	0.0719	0.1179	0.0166	0.1013	0.2646
4- 5	33.8	0.2597	0.2775	-0.0029	-0.0149	0.1346	0.0144	0.1202	0.2646
5- 6	30.1	0.1460	0.1446	-0.0013	0.0027	0.1473	0.0134	0.1338	0.2646
6- 7	27.3	0.2355	0.2337	-0.0019	0.0036	0.1611	0.0131	0.1480	0.2646
7- 8	24.2	0.2146	0.2160	-0.0015	0.0001	0.1789	0.0121	0.1668	0.2646
8- 9	21.6	0.2061	0.2118	-0.0013	-0.0045	0.1980	0.0115	0.1865	0.2646
9-10	20.4	0.0069	0.0014	-0.0000	0.0055	0.2078	0.0110	0.1968	0.2646
10-11	29.3	-1.1777	-1.1744	0.0099	-0.0132	0.1698	0.0182	0.1515	0.2646
11-12	35.3	0.2971	0.2970	-0.0033	0.0034	0.1300	0.0147	0.1153	0.2646
12-13	28.5	0.5177	0.5282	-0.0044	-0.0061	0.1579	0.0136	0.1443	0.2646
13-14	33.2	-0.9934	-0.9993	0.0100	-0.0040	0.1503	0.0199	0.1304	0.2646
14-15	37.7	0.4154	0.4147	-0.0050	0.0057	0.1252	0.0165	0.1087	0.2646
15-16	33.9	-0.0481	-0.0501	0.0005	0.0015	0.1337	0.0146	0.1191	0.2646
16-17	34.6	-0.0342	-0.0260	0.0093	-0.0085	0.1310	0.0142	0.1167	0.2646
17-18	30.7	0.5153	0.5176	-0.0047	0.0025	0.1486	0.0145	0.1341	0.2646
18-19	27.5	-0.1465	-0.1473	0.0012	-0.0004	0.1599	0.0132	0.1467	0.2646
19-20	29.4	-0.1031	-0.1070	0.0009	0.0029	0.1505	0.0133	0.1371	0.2646
20-21	28.3	0.2562	0.2594	-0.0022	-0.0011	0.1564	0.0134	0.1430	0.2646

TABLE A-II-13.C

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 90.5 K NARNISIKI DATA

EX= 0.0010 EP= 1.0000 FT= 0.2000

PAIR NO	P (av.)	I	II	III	D	DMAX	DX	DP	DT
1- 2	49.1	0.1281	0.1487	-0.0026	-0.0180	0.1191	0.0362	0.0829	0.2646
2- 3	45.8	0.0738	0.1057	-0.0017	-0.0302	0.1224	0.0337	0.0887	0.2646
3- 4	40.4	0.4391	0.3720	-0.0049	0.0719	0.1345	0.0332	0.1013	0.2646
4- 5	33.8	0.2597	0.2775	-0.0029	-0.0149	0.1490	0.0298	0.1202	0.2646
5- 6	30.1	0.1460	0.1446	-0.0013	0.0027	0.1607	0.0269	0.1338	0.2646
6- 7	27.3	0.2355	0.2337	-0.0019	0.0036	0.1742	0.0261	0.1480	0.2646
7- 8	24.2	0.2146	0.2160	-0.0015	0.0001	0.1910	0.0241	0.1669	0.2646
8- 9	21.6	0.2061	0.2118	-0.0013	-0.0045	0.2095	0.0230	0.1865	0.2646
9-10	20.4	0.0069	0.0014	-0.0000	0.0055	0.2188	0.0220	0.1968	0.2646
10-11	29.3	-1.1777	-1.1744	0.0099	-0.0132	0.1880	0.0365	0.1515	0.2646
11-12	35.3	0.2971	0.2970	-0.0033	0.0034	0.1447	0.0294	0.1153	0.2646
12-13	28.5	0.5177	0.5282	-0.0044	-0.0061	0.1716	0.0273	0.1443	0.2646
13-14	33.2	-0.9934	-0.9993	0.0100	-0.0040	0.1703	0.0399	0.1304	0.2646
14-15	37.7	0.4154	0.4147	-0.0050	0.0057	0.1417	0.0330	0.1087	0.2646
15-16	33.9	-0.0481	-0.0501	0.0005	0.0015	0.1483	0.0291	0.1191	0.2646
16-17	34.6	-0.0342	-0.0260	0.0003	-0.0085	0.1452	0.0284	0.1167	0.2646
17-18	30.7	0.5153	0.5176	-0.0047	0.0025	0.1631	0.0290	0.1341	0.2646
18-19	27.5	-0.1465	-0.1473	0.0012	-0.0004	0.1731	0.0264	0.1467	0.2646
19-20	29.4	-0.1031	-0.1070	0.0009	0.0029	0.1638	0.0266	0.1371	0.2646
20-21	28.3	0.2562	0.2594	-0.0022	-0.0011	0.1697	0.0267	0.1430	0.2646

TABLE A-II. 13. D

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 90.5 K NARNISIKI DATA

EX= 0.0020 EP= 1.0000 FT= 0.2000

PAIR NC	P(av.)	I	II	III	D	DMAX	DX	DP	DT
1- 2	49.1	0.1281	0.1487	-0.0026	-0.0180	0.1553	0.0724	0.0829	0.2646
2- 3	45.8	0.0738	0.1057	-0.0017	-0.0302	0.1561	0.0673	0.0887	0.2646
3- 4	40.4	0.4391	0.3720	-0.0049	0.0719	0.1677	0.0653	0.1013	0.2646
4- 5	33.8	0.2597	0.2775	-0.0029	-0.0149	0.1778	0.0576	0.1202	0.2646
5- 6	30.1	0.1460	0.1446	-0.0013	0.0027	0.1876	0.0538	0.1338	0.2646
6- 7	27.3	0.2355	0.2337	-0.0019	0.0036	0.2003	0.0523	0.1480	0.2646
7- 8	24.2	0.2146	0.2160	-0.0015	0.0001	0.2151	0.0483	0.1668	0.2646
8- 9	21.6	0.2061	0.2118	-0.0013	-0.0045	0.2326	0.0461	0.1865	0.2646
9-10	20.4	0.0069	0.0014	-0.0000	0.0055	0.2409	0.0440	0.1968	0.2646
10-11	29.3	-1.1777	-1.1744	0.0099	-0.0132	0.2245	0.0730	0.1515	0.2646
11-12	35.3	0.2971	0.2970	-0.0033	0.0034	0.1741	0.0588	0.1153	0.2646
12-13	28.5	0.5177	0.5282	-0.0044	-0.0061	0.1989	0.0546	0.1443	0.2646
13-14	33.2	-0.9934	-0.9993	0.0100	-0.0040	0.2101	0.0797	0.1304	0.2646
14-15	37.7	0.4154	0.4147	-0.0050	0.0057	0.1747	0.0660	0.1087	0.2646
15-16	33.9	-0.0481	-0.0501	0.0005	0.0015	0.1774	0.0582	0.1191	0.2646
16-17	34.6	-0.0342	-0.0260	0.0003	-0.0085	0.1736	0.0569	0.1167	0.2646
17-18	30.7	0.5153	0.5176	-0.0047	0.0025	0.1921	0.0580	0.1341	0.2646
18-19	27.5	-0.1465	-0.1473	0.0012	-0.0004	0.1994	0.0529	0.1467	0.2646
19-20	29.4	-0.1031	-0.1070	0.0009	0.0029	0.1904	0.0532	0.1371	0.2646
20-21	28.3	0.2562	0.2594	-0.0022	-0.0011	0.1964	0.0534	0.1430	0.2646

TABLE A-II-14. A

TESTED DATA OF OXYGEN ARGON NITROGEN AT 100.0 K NARNISIKI DATA

ID	P, atm	x_1	x_2	x_3	y_1	y_2	y_3
1	7.1	0.1073	0.0393	0.8534	0.0501	0.0229	0.9270
2	6.7	0.1666	0.0929	0.7405	0.0804	0.0551	0.8645
3	6.2	0.2627	0.0896	0.6477	0.1326	0.0558	0.8146
4	6.3	0.2315	0.0865	0.6820	0.1146	0.0541	0.8313
5	4.6	0.5019	0.1711	0.3270	0.3105	0.1383	0.5512
6	3.8	0.6869	0.1130	0.2001	0.4842	0.1075	0.4083
7	3.5	0.6757	0.2045	0.1198	0.5196	0.2110	0.2694
8	3.2	0.7654	0.1683	0.0663	0.6386	0.1921	0.1693
9	5.4	0.3020	0.2086	0.4894	0.1658	0.1451	0.6881
10	4.7	0.4601	0.2058	0.3341	0.2828	0.1612	0.5560
11	5.9	0.1417	0.2896	0.5587	0.0758	0.1854	0.7388
12	5.6	0.1598	0.3326	0.5076	0.0857	0.2201	0.6942
13	5.4	0.1681	0.3859	0.4461	0.0953	0.2613	0.6434
14	4.8	0.3368	0.3105	0.3527	0.1997	0.2326	0.5677
15	4.1	0.4806	0.3008	0.2186	0.3260	0.2624	0.4116
16	3.4	0.5414	0.3750	0.0836	0.4293	0.3806	0.1901
17	3.5	0.4617	0.4501	0.0882	0.3610	0.4397	0.1993
18	4.0	0.3672	0.4566	0.1762	0.2599	0.3997	0.3404
19	4.9	0.1234	0.5474	0.3292	0.0778	0.3997	0.5325
20	4.2	0.1224	0.6964	0.1712	0.0876	0.5703	0.3421
21	3.9	0.0955	0.7759	0.1285	0.0728	0.6641	0.2631

TABLE A-II-14. B

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 100.0 K NARNISIKI DATA

FX= 0.0005 FP= 1.0000 ET= 0.2000

PAIR NC	P(av.)	I	II	III	D	DMAX	DX	DP	DT
1- 2	101.1	0.1263	0.1092	-0.0041	0.0213	0.0579	0.0169	0.0409	0.2646
2- 3	94.1	0.1315	0.1325	-0.0044	0.0034	0.0593	0.0154	0.0438	0.2646
3- 4	91.7	-0.0505	-0.0452	0.0015	-0.0068	0.0593	0.0149	0.0449	0.2646
4- 5	79.9	0.5567	0.5703	-0.0148	0.0012	0.0688	0.0162	0.0526	0.2646
5- 6	61.3	0.3022	0.3342	-0.0060	-0.0261	0.0798	0.0129	0.0669	0.2646
6- 7	53.5	0.1754	0.1458	-0.0022	0.0318	0.0882	0.0123	0.0759	0.2646
7- 8	49.0	0.1788	0.1740	-0.0023	0.0072	0.0943	0.0115	0.0827	0.2646
8- 9	63.1	-0.9586	-0.9540	0.0174	-0.0220	0.0869	0.0178	0.0591	0.2646
9-10	73.9	0.2695	0.2600	-0.0061	0.0156	0.0695	0.0139	0.0555	0.2646
10-11	77.8	-0.3888	-0.4174	0.0103	0.0183	0.0695	0.0164	0.0531	0.2646
11-12	84.8	0.0573	0.0896	-0.0024	-0.0299	0.0626	0.0144	0.0481	0.2646
12-13	80.7	0.0854	0.0830	-0.0022	0.0046	0.0649	0.0141	0.0508	0.2646
13-14	74.9	0.1807	0.1835	-0.0043	0.0015	0.0592	0.0144	0.0547	0.2646
14-15	65.6	0.2927	0.2931	-0.0058	0.0053	0.0757	0.0132	0.0625	0.2646
15-16	55.5	0.3189	0.3133	-0.0049	0.0105	0.0864	0.0127	0.0737	0.2646
16-17	51.1	-0.0467	-0.0318	0.0004	-0.0154	0.0908	0.0115	0.0792	0.2646
17-18	55.1	-0.2234	-0.2332	0.0036	0.0061	0.0862	0.0123	0.0739	0.2646
18-19	65.0	-0.3286	-0.3542	0.0069	0.0187	0.0780	0.0148	0.0632	0.2646
19-20	66.4	0.2796	0.2733	-0.0055	0.0118	0.0746	0.0132	0.0614	0.2646
20-21	59.6	0.0861	0.1025	-0.0017	-0.0147	0.0802	0.0123	0.0679	0.2646

TABLE A-II-14. C

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 100.0 K NARNISKI DATA

FX= 0.0010 FP= 1.0000 FT= 0.2000

PAIR NO	P(av.)	I	II	III	D	DMAX	DX	DP	DT
1- 2	101.1	0.1263	0.1092	-0.0041	0.0213	0.0747	0.0338	0.0409	0.2646
2- 3	94.1	0.1315	0.1325	-0.0044	0.0034	0.0747	0.0308	0.0433	0.2646
3- 4	91.7	-0.0505	-0.0452	0.0015	-0.0068	0.0747	0.0298	0.0449	0.2646
4- 5	79.9	0.5567	0.5703	-0.0148	0.0012	0.0349	0.0323	0.0526	0.2646
5- 6	61.3	0.3022	0.3342	-0.0060	-0.0261	0.0928	0.0259	0.0669	0.2646
6- 7	53.5	0.1754	0.1458	-0.0022	0.0318	0.1005	0.0246	0.0759	0.2646
7- 8	49.0	0.1788	0.1740	-0.0023	0.0072	0.1058	0.0231	0.0827	0.2646
8- 9	63.1	-0.9586	-0.9540	0.0174	-0.0220	0.1047	0.0356	0.0691	0.2646
9-10	73.9	0.2695	0.2600	-0.0061	0.0156	0.0834	0.0278	0.0555	0.2646
10-11	77.8	-0.3988	-0.4174	0.0103	0.0183	0.0859	0.0327	0.0531	0.2646
11-12	84.8	0.0573	0.0896	-0.0024	-0.0299	0.0770	0.0288	0.0481	0.2646
12-13	80.7	0.0854	0.0830	-0.0022	0.0046	0.0791	0.0283	0.0508	0.2646
13-14	74.9	0.1807	0.1835	-0.0043	0.0015	0.0836	0.0289	0.0547	0.2646
14-15	65.6	0.2927	0.2931	-0.0058	0.0053	0.0889	0.0264	0.0625	0.2646
15-16	55.5	0.3139	0.3133	-0.0049	0.0105	0.0990	0.0253	0.0737	0.2646
16-17	51.1	-0.0467	-0.0318	0.0004	-0.0154	0.1023	0.0231	0.0792	0.2646
17-18	55.1	-0.2234	-0.2332	0.0036	0.0061	0.0985	0.0246	0.0739	0.2646
18-19	65.0	-0.3286	-0.3542	0.0069	0.0187	0.0928	0.0295	0.0632	0.2646
19-20	66.4	0.2796	0.2733	-0.0055	0.0118	0.0878	0.0264	0.0614	0.2646
20-21	59.6	0.0861	0.1025	-0.0017	-0.0147	0.0925	0.0245	0.0679	0.2646

TABLE A-II-14. D

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 100.0 K NARNISIKI DATA

EX= 0.0020 EP= 1.0000 ET= 0.2000

PAIR NO	P(av.)	I	II	III	D	DMAX	DX	DP	DT
1- 2	101.1	0.1263	0.1092	-0.0041	0.0213	0.1085	0.0675	0.0409	0.2646
2- 3	94.1	0.1315	0.1325	-0.0044	0.0034	0.1054	0.0616	0.0438	0.2646
3- 4	91.7	-0.0505	-0.0452	0.0015	-0.0068	0.1045	0.0596	0.0449	0.2646
4- 5	79.9	0.5567	0.5703	-0.0148	0.0012	0.1173	0.0647	0.0526	0.2646
5- 6	61.3	0.3022	0.3342	-0.0060	-0.0261	0.1186	0.0517	0.0669	0.2646
6- 7	53.5	0.1754	0.1458	-0.0022	0.0318	0.1251	0.0492	0.0759	0.2646
7- 8	49.0	0.1788	0.1740	-0.0023	0.0072	0.1289	0.0461	0.0827	0.2646
8- 9	63.1	-0.9586	-0.9540	0.0174	-0.0220	0.1404	0.0713	0.0691	0.2646
9-10	73.9	0.2695	0.2600	-0.0061	0.0156	0.1112	0.0557	0.0555	0.2646
10-11	77.8	-0.3888	-0.4174	0.0103	0.0183	0.1186	0.0655	0.0531	0.2646
11-12	84.8	0.0573	0.0896	-0.0024	-0.0299	0.1053	0.0576	0.0481	0.2646
12-13	80.7	0.0854	0.0830	-0.0022	0.0046	0.1074	0.0565	0.0508	0.2646
13-14	74.9	0.1807	0.1835	-0.0043	0.0015	0.1125	0.0578	0.0547	0.2646
14-15	65.6	0.2927	0.2931	-0.0058	0.0053	0.1153	0.0528	0.0625	0.2646
15-16	55.5	0.3189	0.3133	-0.0049	0.0105	0.1244	0.0597	0.0737	0.2646
16-17	51.1	-0.0467	-0.0318	0.0004	-0.0154	0.1254	0.0461	0.0792	0.2646
17-18	55.1	-0.2234	-0.2332	0.0036	0.0061	0.1231	0.0492	0.0739	0.2646
18-19	65.0	-0.3286	-0.3542	0.0069	0.0187	0.1223	0.0591	0.0632	0.2646
19-20	66.4	0.2796	0.2733	-0.0055	0.0118	0.1142	0.0527	0.0614	0.2646
20-21	59.6	0.0861	0.1025	-0.0017	-0.0147	0.1170	0.0491	0.0679	0.2646

TABLE A-II-15. A

TESTED DATA OF OXYGEN ARGON NITROGEN AT 110.0 K NARNISIKI DATA

ID	P, atm	x_1	x_2	x_3	y_1	y_2	y_3
1	13.4	0.1189	0.0427	0.8384	0.0667	0.0273	0.9060
2	11.7	0.2667	0.0904	0.6429	0.1578	0.0630	0.7792
3	9.9	0.4487	0.1155	0.4357	0.2884	0.0921	0.6195
4	8.4	0.5999	0.1180	0.2821	0.4299	0.1059	0.4642
5	8.2	0.6292	0.1184	0.2521	0.4586	0.1100	0.4314
6	7.3	0.7493	0.0792	0.1710	0.5952	0.0792	0.3256
7	6.7	0.8373	0.0548	0.1079	0.7122	0.0605	0.2273
8	6.3	0.7649	0.1897	0.0454	0.6834	0.2169	0.0997
9	6.9	0.6925	0.2029	0.1045	0.5762	0.2133	0.2105
10	13.0	0.1085	0.1167	0.7748	0.0620	0.0780	0.8600
11	11.0	0.2485	0.1935	0.5579	0.1529	0.1425	0.7046
12	9.9	0.3411	0.2006	0.4583	0.2213	0.1815	0.5972
13	9.0	0.4576	0.2120	0.3304	0.3153	0.1769	0.5078
14	11.1	0.1500	0.2965	0.5535	0.0924	0.2128	0.6948
15	10.6	0.1689	0.3459	0.4852	0.1060	0.2529	0.6411
16	8.6	0.3712	0.3689	0.2599	0.2662	0.3173	0.4165
17	7.7	0.4899	0.3029	0.2072	0.3686	0.2750	0.3564
18	6.8	0.5562	0.3680	0.0758	0.4702	0.3825	0.1473
19	6.7	0.5025	0.4601	0.0374	0.4397	0.4797	0.0806
20	10.3	0.1736	0.3883	0.4881	0.1120	0.2899	0.5981
21	9.3	0.2121	0.4550	0.3329	0.1494	0.3589	0.4918
22	9.5	0.2339	0.4080	0.3581	0.1593	0.3152	0.5255
23	7.8	0.3745	0.4586	0.1669	0.2886	0.4176	0.2938
24	9.3	0.1280	0.5560	0.3160	0.0902	0.4363	0.4735
25	8.6	0.1570	0.6109	0.2321	0.1160	0.5076	0.3768
26	8.3	0.2359	0.5659	0.1982	0.1894	0.4895	0.3311
27	7.8	0.0966	0.7790	0.1244	0.0735	0.6953	0.2262

TABLE A-II-15. B

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 110.0 K NARNISIKI DATA

EX= 0.0005 EP= 1.0000 ET= 0.2000

PAIR NO	P(av.)	I	II	III	D	±DMAX	DX	DP	DT
1- 2	184.5	0.2020	0.2134	-0.0161	0.0047	0.0393	0.0161	0.0230	0.2546
2- 3	158.2	0.2592	0.2665	-0.0153	0.0080	0.0408	0.0142	0.0266	0.2646
3- 4	134.5	0.2466	0.2559	-0.0112	0.0019	0.0440	0.0129	0.0310	0.2646
4- 5	122.1	0.0499	0.0546	-0.0021	-0.0026	0.0460	0.0122	0.0338	0.2646
5- 6	113.5	0.1783	0.1978	-0.0067	-0.0128	0.0486	0.0122	0.0364	0.2646
6- 7	102.4	0.1500	0.1514	-0.0044	0.0030	0.0518	0.0117	0.0401	0.2646
7- 8	95.6	0.0937	0.0854	-0.0023	0.0105	0.0561	0.0133	0.0428	0.2646
8- 9	97.4	-0.1497	-0.1482	0.0040	-0.0055	0.0538	0.0117	0.0421	0.2646
9-10	146.1	-0.9698	-1.0233	0.0489	0.0046	0.0553	0.0240	0.0313	0.2646
10-11	176.4	0.2399	0.2461	-0.0173	0.0111	0.0395	0.0155	0.0240	0.2645
11-12	154.2	0.1349	0.1651	-0.0089	-0.0212	0.0407	0.0136	0.0271	0.2545
12-13	139.5	0.1449	0.1567	-0.0073	-0.0045	0.0431	0.0132	0.0299	0.2646
13-14	148.2	-0.3236	-0.3348	0.0171	-0.0059	0.0438	0.0154	0.0284	0.2646
14-15	159.6	0.0711	0.0783	-0.0046	-0.0026	0.0400	0.0136	0.0262	0.2646
15-16	141.3	0.3320	0.3309	-0.0156	0.0166	0.0439	0.0142	0.0297	0.2546
16-17	120.1	0.1107	0.1944	-0.0071	-0.0766	0.0470	0.0125	0.0345	0.2645
17-18	106.7	0.2472	0.2071	-0.0064	0.0465	0.0513	0.0126	0.0386	0.2646
18-19	99.1	0.0507	0.0422	-0.0012	0.0097	0.0531	0.0118	0.0413	0.2645
19-20	124.3	-0.7861	-0.7411	0.0295	-0.0745	0.0580	0.0223	0.0356	0.2646*
20-21	143.9	0.2338	0.1542	-0.0090	0.0886	0.0432	0.0134	0.0297	0.2646*
21-22	138.5	-0.0376	-0.0294	0.0014	-0.0096	0.0429	0.0129	0.0300	0.2645
22-23	127.3	0.3132	0.3265	-0.0131	-0.0003	0.0461	0.0133	0.0328	0.2646
23-24	126.0	-0.2737	-0.2955	0.0116	0.0102	0.0474	0.0143	0.0330	0.2645
24-25	132.0	0.1242	0.1267	-0.0054	0.0028	0.0442	0.0127	0.0314	0.2645
25-26	124.3	0.0731	0.0711	-0.0027	0.0047	0.0457	0.0125	0.0332	0.2646
26-27	118.1	0.0897	0.1010	-0.0036	-0.0077	0.0482	0.0132	0.0349	0.2646

* inconsistent

TABLE A-II-15. C

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 110.0 K NARNISIKI DATA

EX= 0.0010 EP= 1.0000 ET= 0.2000

PAIR NO	P(av.)	I	II	III	D	±DMAX	DX	DP	DT
1- 2	184.5	0.2020	0.2134	-0.0161	0.0047	0.0554	0.0322	0.0230	0.2646
2- 3	158.2	0.2592	0.2665	-0.0153	0.0080	0.0550	0.0283	0.0266	0.2646
3- 4	134.5	0.2466	0.2559	-0.0112	0.0019	0.0569	0.0259	0.0310	0.2646
4- 5	122.1	0.0499	0.0546	-0.0021	-0.0026	0.0582	0.0244	0.0338	0.2646
5- 6	113.5	0.1783	0.1978	-0.0067	-0.0128	0.0607	0.0243	0.0364	0.2646
6- 7	102.4	0.1500	0.1514	-0.0044	0.0030	0.0635	0.0234	0.0401	0.2646
7- 8	95.6	0.0937	0.0854	-0.0023	0.0105	0.0694	0.0266	0.0428	0.2646
8- 9	97.4	-0.1497	-0.1482	0.0040	-0.0055	0.0655	0.0233	0.0421	0.2646
9-10	146.1	-0.9698	-1.0233	0.0489	0.0046	0.0793	0.0480	0.0313	0.2646
10-11	176.4	0.2399	0.2461	-0.0173	0.0111	0.0550	0.0399	0.0240	0.2646
11-12	154.2	0.1349	0.1651	-0.0089	-0.0212	0.0543	0.0271	0.0271	0.2646
12-13	139.5	0.1449	0.1567	-0.0073	-0.0045	0.0562	0.0263	0.0298	0.2646
13-14	148.2	-0.3236	-0.3348	0.0171	-0.0059	0.0592	0.0307	0.0284	0.2646
14-15	159.6	0.0711	0.0783	-0.0046	-0.0026	0.0536	0.0273	0.0262	0.2646
15-16	141.3	0.3320	0.3309	-0.0156	0.0166	0.0581	0.0283	0.0297	0.2646
16-17	120.1	0.1107	0.1944	-0.0071	-0.0766	0.0594	0.0249	0.0345	0.2646
17-18	106.7	0.2472	0.2071	-0.0064	0.0465	0.0639	0.0253	0.0386	0.2646
18-19	99.1	0.0507	0.0422	-0.0012	0.0097	0.0649	0.0235	0.0413	0.2646
19-20	124.3	-0.7861	-0.7411	0.0295	-0.0745	0.0803	0.0446	0.0356	0.2646 *
20-21	143.9	0.2338	0.1542	-0.0090	0.0886	0.0566	0.0268	0.0297	0.2646 *
21-22	138.5	-0.0376	-0.0294	0.0014	-0.0096	0.0558	0.0257	0.0300	0.2646
22-23	127.3	0.3132	0.3265	-0.0131	-0.0003	0.0593	0.0265	0.0328	0.2646
23-24	126.0	-0.2737	-0.2955	0.0116	0.0102	0.0617	0.0286	0.0330	0.2646
24-25	132.0	0.1242	0.1267	-0.0054	0.0028	0.0569	0.0254	0.0314	0.2646
25-26	124.3	0.0731	0.0711	-0.0027	0.0047	0.0582	0.0250	0.0332	0.2646
26-27	118.1	0.0897	0.1010	-0.0036	-0.0077	0.0614	0.0265	0.0349	0.2646

TABLE A-II-15. D

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 110.0 K NARNISIKI DATA

EX= 0.0020 EP= 1.0000 ET= 0.2000

PAIR NO	P(av.)	I	II	III	D	±DMAX	DX	DP	DT
1- 2	184.5	0.2020	0.2134	-0.0161	0.0047	0.0876	0.0645	0.0230	0.2645
2- 3	158.2	0.2592	0.2665	-0.0153	0.0080	0.0833	0.0566	0.0266	0.2646
3- 4	134.5	0.2466	0.2559	-0.0112	0.0019	0.0828	0.0517	0.0310	0.2646
4- 5	122.1	0.0499	0.0546	-0.0021	-0.0026	0.0825	0.0488	0.0338	0.2646
5- 6	113.5	0.1783	0.1978	-0.0067	-0.0129	0.0850	0.0486	0.0364	0.2646
6- 7	102.4	0.1500	0.1514	-0.0044	0.0030	0.0869	0.0467	0.0401	0.2646
7- 8	95.6	0.0937	0.0854	-0.0023	0.0105	0.0960	0.0531	0.0428	0.2646
8- 9	97.4	-0.1497	-0.1482	0.0040	-0.0055	0.0888	0.0467	0.0421	0.2646
9-10	146.1	-0.9698	-1.0233	0.0489	0.0046	0.1273	0.0959	0.0313	0.2646
10-11	176.4	0.2399	0.2461	-0.0173	0.0111	0.0959	0.0618	0.0240	0.2646
11-12	154.2	0.1349	0.1651	-0.0089	-0.0212	0.0814	0.0543	0.0271	0.2646
12-13	139.5	0.1449	0.1567	-0.0073	-0.0045	0.0826	0.0527	0.0298	0.2646
13-14	148.2	-0.3236	-0.3348	0.0171	-0.0059	0.0899	0.0615	0.0284	0.2646
14-15	159.6	0.0711	0.0783	-0.0046	-0.0026	0.0809	0.0546	0.0262	0.2646
15-16	141.3	0.3320	0.3309	-0.0156	0.0166	0.0864	0.0567	0.0297	0.2646
16-17	120.1	0.1107	0.1944	-0.0071	-0.0766	0.0844	0.0499	0.0345	0.2646
17-18	106.7	0.2472	0.2071	-0.0064	0.0465	0.0892	0.0505	0.0386	0.2646
18-19	99.1	0.0507	0.0422	-0.0012	0.0097	0.0884	0.0471	0.0413	0.2646
19-20	124.3	-0.7861	-0.7411	0.0295	-0.0745	0.1249	0.0892	0.0356	0.2646 *
20-21	143.9	0.2339	0.1542	-0.0090	0.0886	0.0834	0.0537	0.0297	0.2646 *
21-22	138.5	-0.0376	-0.0294	0.0014	-0.0096	0.0815	0.0515	0.0300	0.2646
22-23	127.3	0.3132	0.3265	-0.0131	-0.0003	0.0859	0.0530	0.0329	0.2646
23-24	126.0	-0.2737	-0.2955	0.0116	0.0102	0.0903	0.0572	0.0330	0.2646
24-25	132.0	0.1242	0.1267	-0.0054	0.0028	0.0823	0.0508	0.0314	0.2646
25-26	124.3	0.0731	0.0711	-0.0027	0.0047	0.0832	0.0499	0.0332	0.2646
26-27	118.1	0.0897	0.1010	-0.0036	-0.0077	0.0879	0.0529	0.0349	0.2646

TABLE A-II.16. A

TESTED DATA OF OXYGEN ARGON NITROGEN AT 120.0 K NAPNISIKI DATA

ID	P, atm	x_1	x_2	x_3	y_1	y_2	y_3
1	22.9	0.1201	0.0432	0.8367	0.0802	0.0318	0.8890
2	19.7	0.3857	0.0950	0.6193	0.1954	0.0736	0.7310
3	16.8	0.4601	0.1176	0.4223	0.3342	0.1006	0.5652
4	14.6	0.6145	0.1182	0.2673	0.4821	0.1094	0.4085
5	14.4	0.5399	0.2356	0.2245	0.4308	0.2205	0.3487
6	13.1	0.6518	0.2034	0.1448	0.5489	0.2035	0.2476
7	12.4	0.7090	0.1992	0.0918	0.6227	0.2108	0.1665
8	13.3	0.7133	0.1105	0.1762	0.5950	0.1140	0.2910
9	12.1	0.8887	0.0554	0.1059	0.7434	0.0608	0.1958
10	22.0	0.1207	0.1274	0.7519	0.0805	0.0912	0.8283
11	19.7	0.2365	0.1509	0.6126	0.1610	0.1167	0.7223
12	18.6	0.2623	0.2074	0.5303	0.1857	0.1632	0.6511
13	16.0	0.4260	0.2292	0.3448	0.3221	0.1986	0.4793
14	17.8	0.1779	0.3611	0.4610	0.1239	0.2886	0.5875
15	13.4	0.5768	0.2668	0.1569	0.4862	0.2597	0.2541
16	18.9	0.1559	0.3039	0.5342	0.1097	0.2417	0.6486
17	15.8	0.3572	0.3250	0.3178	0.2716	0.2807	0.4477
18	13.8	0.4894	0.3017	0.2089	0.3935	0.2814	0.3251
19	17.4	0.1822	0.4046	0.4132	0.1328	0.3263	0.5379
20	16.6	0.2345	0.4056	0.3599	0.1745	0.3366	0.4889
21	13.8	0.3791	0.4573	0.1636	0.3118	0.4278	0.2584
22	13.7	0.4238	0.4278	0.1484	0.3537	0.4054	0.2409
23	16.1	0.1324	0.5703	0.2973	0.1024	0.4790	0.4186
24	14.4	0.2441	0.5731	0.1828	0.1993	0.5184	0.2823
25	14.3	0.1280	0.7170	0.1550	0.1073	0.6469	0.2458

TABLE A-II-16. B

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 120.0 K NARNISIKI DATA

EX= 0.0005 FP= 1.0000 ET= 0.2000

PAIR NC	P(av.)	I	II	III	D	± DMAX	DX	DP	DT
1-	312.8	0.0722	0.2016	-0.0210	-0.1585	0.0320	0.0169	0.0149	0.2646 *
2- 3	268.0	0.4567	0.2298	-0.0354	0.2622	0.0312	0.0140	0.0170	0.2646 *
3- 4	230.9	0.1928	0.2054	-0.0172	0.0046	0.0312	0.0126	0.0185	0.2645
4- 5	213.0	0.0314	0.0278	-0.0021	0.0057	0.0324	0.0125	0.0198	0.2646
5- 6	202.1	0.1290	0.1382	-0.0090	-0.0002	0.0329	0.0120	0.0208	0.2546
6- 7	187.4	0.0937	0.0973	-0.0056	0.0020	0.0340	0.0116	0.0223	0.2646
7- 8	188.7	-0.1063	-0.1183	0.0069	0.0051	0.0344	0.0122	0.0222	0.2646
8- 9	187.1	0.0466	0.1514	-0.0081	-0.0966	0.0353	0.0122	0.0230	0.2646 *
9-10	250.6	-0.7510	-0.8775	0.0805	0.0461	0.0433	0.0242	0.0190	0.2646 *
10-11	306.3	0.1216	0.1397	-0.0213	0.0031	0.0287	0.0141	0.0143	0.2646
11-12	281.6	0.0720	0.0777	-0.0099	0.0042	0.0288	0.0133	0.0154	0.2646
12-13	254.3	0.1960	0.2155	-0.0217	0.0023	0.0303	0.0132	0.0169	0.2646
13-14	248.7	-0.1503	-0.1580	0.0151	-0.0075	0.0313	0.0139	0.0172	0.2646
14-15	229.6	0.3913	0.4279	-0.0349	-0.0017	0.0342	0.0153	0.0188	0.2646
15-16	237.3	-0.4467	-0.5045	0.0432	0.0145	0.0347	0.0162	0.0184	0.2646
16-17	255.3	0.2183	0.2489	-0.0248	-0.0058	0.0309	0.0139	0.0168	0.2646
17-18	218.2	0.1479	0.2049	-0.0154	-0.0416	0.0320	0.0125	0.0194	0.2646 *
18-19	229.6	-0.2774	-0.3397	0.0278	0.0345	0.0329	0.0141	0.0187	0.2646
19-20	249.9	0.0619	0.0650	-0.0063	0.0032	0.0309	0.0128	0.0171	0.2646
20-21	223.2	0.2437	0.2841	-0.0221	-0.0182	0.0323	0.0131	0.0191	0.2646
21-22	201.8	0.0317	0.0069	-0.0004	0.0253	0.0327	0.0119	0.0208	0.2646
22-23	218.7	-0.2262	-0.2409	0.0179	-0.0032	0.0339	0.0144	0.0194	0.2646
23-24	224.1	0.1495	0.1593	-0.0125	0.0027	0.0319	0.0129	0.0189	0.2646
24-25	210.9	0.0155	0.0192	-0.0014	-0.0023	0.0325	0.0125	0.0200	0.2646

TABLE A-II-16. C

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 120.0 K NARNISIKI DATA

EX= 0.0010 EP= 1.0000 ET= 0.2000

PAIR NO	P(av.)	I	II	III	D	± DMAX	DX	DP	DT
1- 2	312.9	0.0722	0.2016	-0.0210	-0.1585	0.0489	0.0338	0.0149	0.2646 *
2- 3	268.0	0.4567	0.2298	-0.0354	0.2622	0.0452	0.0280	0.0170	0.2646 *
3- 4	230.9	0.1928	0.2054	-0.0172	0.0046	0.0439	0.0253	0.0185	0.2646
4- 5	213.0	0.0314	0.0278	-0.0021	0.0057	0.0450	0.0251	0.0198	0.2646
5- 6	202.1	0.1290	0.1382	-0.0090	-0.0002	0.0449	0.0240	0.0208	0.2646
6- 7	187.4	0.0937	0.0973	-0.0056	0.0020	0.0456	0.0232	0.0223	0.2646
7- 8	188.7	-0.1063	-0.1183	0.0069	0.0051	0.0466	0.0243	0.0222	0.2646 *
8- 9	187.1	0.0466	0.1514	-0.0081	-0.0966	0.0475	0.0245	0.0230	0.2646 *
9-10	250.6	-0.7510	-0.8775	0.0805	0.0461	0.0675	0.0485	0.0190	0.2646
10-11	306.3	0.1216	0.1397	-0.0213	0.0031	0.0428	0.0283	0.0143	0.2646
11-12	281.6	0.0720	0.0777	-0.0099	0.0042	0.0421	0.0266	0.0154	0.2646
12-13	254.3	0.1960	0.2155	-0.0217	0.0023	0.0435	0.0264	0.0169	0.2646
13-14	248.7	-0.1503	-0.1580	0.0151	-0.0075	0.0452	0.0279	0.0172	0.2646
14-15	229.6	0.3913	0.4279	-0.0349	-0.0017	0.0495	0.0306	0.0188	0.2646
15-16	237.3	-0.4467	-0.5045	0.0432	0.0145	0.0509	0.0325	0.0184	0.2646
16-17	255.3	0.2183	0.2489	-0.0248	-0.0053	0.0448	0.0278	0.0168	0.2646
17-18	218.2	0.1479	0.2049	-0.0154	-0.0416	0.0444	0.0249	0.0194	0.2646 *
18-19	229.6	-0.2774	-0.3397	0.0278	0.0345	0.0471	0.0283	0.0187	0.2646
19-20	249.9	0.0619	0.0650	-0.0063	0.0032	0.0429	0.0255	0.0171	0.2646
20-21	223.2	0.2437	0.2841	-0.0221	-0.0182	0.0454	0.0262	0.0191	0.2646
21-22	201.8	0.0317	0.0069	-0.0004	0.0253	0.0446	0.0237	0.0208	0.2646
22-23	218.7	-0.2262	-0.2409	0.0179	-0.0032	0.0482	0.0287	0.0194	0.2646
23-24	224.1	0.1495	0.1593	-0.0125	0.0027	0.0447	0.0257	0.0189	0.2646
24-25	210.9	0.0155	0.0192	-0.0014	-0.0023	0.0450	0.0250	0.0200	0.2646

* inconsistent

TABLE A-II-16. D

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 120.0 K NARNISIKI DATA

FX= 0.0020 EP= 1.0000 ET= 0.2000

PAIR NO	P (av.)	I	II	III	D	± DMAX	DX	DP	DT
1- 2	312.8	0.0222	0.2016	-0.0210	-0.1585	0.0827	0.0676	0.0149	0.2646 *
2- 3	268.0	0.4567	0.2298	-0.0354	0.2622	0.0731	0.0560	0.0170	0.2646 *
3- 4	230.9	0.1929	0.2054	-0.0172	0.0046	0.0691	0.0506	0.0185	0.2646
4- 5	213.0	0.0314	0.0278	-0.0021	0.0057	0.0700	0.0502	0.0198	0.2646
5- 6	202.1	0.1290	0.1382	-0.0090	-0.0002	0.0688	0.0480	0.0208	0.2645
6- 7	187.4	0.0937	0.0973	-0.0056	0.0020	0.0688	0.0464	0.0223	0.2646
7- 8	188.7	-0.1063	-0.1183	0.0069	0.0051	0.0709	0.0487	0.0222	0.2546
8- 9	187.1	0.0466	0.1514	-0.0081	-0.0966	0.0720	0.0490	0.0230	0.2646 *
9-10	250.6	-0.7510	-0.8775	0.0805	0.0461	0.1160	0.0969	0.0190	0.2646 *
10-11	306.3	0.1216	0.1397	-0.0213	0.0031	0.0711	0.0566	0.0143	0.2646
11-12	281.6	0.0720	0.0777	-0.0099	0.0042	0.0687	0.0532	0.0154	0.2646
12-13	254.3	0.1960	0.2155	-0.0217	0.0023	0.0699	0.0529	0.0169	0.2646
13-14	248.7	-0.1503	-0.1580	0.0151	-0.0075	0.0731	0.0558	0.0172	0.2646
14-15	229.6	0.3913	0.4279	-0.0349	-0.0017	0.0801	0.0612	0.0188	0.2646
15-16	237.3	-0.4467	-0.5045	0.0432	0.0145	0.0834	0.0649	0.0184	0.2646
16-17	255.3	0.2183	0.2489	-0.0248	-0.0058	0.0726	0.0556	0.0168	0.2646
17-18	218.2	0.1479	0.2049	-0.0154	-0.0416	0.0694	0.0498	0.0194	0.2646 *
18-19	229.6	-0.2774	-0.3397	0.0278	0.0345	0.0754	0.0566	0.0187	0.2646
19-20	249.9	0.0619	0.0650	-0.0063	0.0032	0.0683	0.0511	0.0171	0.2646
20-21	223.2	0.2437	0.2841	-0.0221	-0.0182	0.0715	0.0523	0.0191	0.2646
21-22	201.8	0.0317	0.0069	-0.0004	0.0253	0.0683	0.0474	0.0209	0.2646
22-23	218.7	-0.2262	-0.2409	0.0179	-0.0032	0.0769	0.0574	0.0194	0.2646
23-24	224.1	0.1495	0.1593	-0.0125	0.0027	0.0704	0.0514	0.0189	0.2646
24-25	210.9	0.0155	0.0192	-0.0014	-0.0023	0.0700	0.0500	0.0200	0.2646

* inconsistent

TABLE A-II-17. A

TESTED DATA OF OXYGEN ARGON NITROGEN AT 90.5 K WILSON DATA

ID	P, atm	x_1	x_2	x_3	y_1	y_2	y_3
1	2.0	0.6506	0.0131	0.3364	0.3565	0.0129	0.6306
2	2.0	0.6397	0.0286	0.3317	0.3502	0.0225	0.6273
3	2.0	0.6267	0.0402	0.3331	0.3403	0.0315	0.6282
4	2.0	0.6181	0.0531	0.3288	0.3361	0.0411	0.6228
5	2.0	0.6010	0.0666	0.3324	0.3260	0.0515	0.6225
6	2.0	0.5795	0.0965	0.3240	0.3054	0.0701	0.6245
7	2.0	0.5410	0.1370	0.3220	0.2982	0.1048	0.5970
8	2.0	0.4761	0.2092	0.3146	0.2636	0.1577	0.5787
9	2.0	0.3302	0.3768	0.2930	0.1861	0.2757	0.5382
10	2.0	0.1792	0.5469	0.2740	0.1039	0.3903	0.5058
11	2.0	0.0859	0.6540	0.2601	0.0530	0.4610	0.4860
12	2.0	0.6485	0.0150	0.3365	0.3527	0.0119	0.6354
13	2.0	0.5778	0.0952	0.3271	0.3152	0.0745	0.6103

TABLE A-II-17.B

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 90.5 K WILSON DATA

EX= 0.0005 FP= 1.0000 FT= 0.2000

PAIR NO	P(av.)	I	II	III	D	± DMAX	DX	DP	DT
1- 2	29.4	-0.0047	-0.0000	0.0000	-0.0046	0.1511	0.0138	0.1372	0.2646
2- 3	29.4	-0.0124	-0.0000	-0.0000	-0.0124	0.1507	0.0134	0.1372	0.2646
3- 4	29.4	0.0035	-0.0000	0.0000	0.0035	0.1507	0.0134	0.1372	0.2646
4- 5	29.4	-0.0106	-0.0000	-0.0000	-0.0106	0.1507	0.0134	0.1372	0.2646
5- 6	29.4	-0.0253	-0.0001	0.0000	-0.0253	0.1510	0.0137	0.1372	0.2646
6- 7	29.4	0.0373	-0.0001	-0.0000	0.0374	0.1510	0.0138	0.1372	0.2646
7- 8	29.4	-0.0056	-0.0001	-0.0000	-0.0054	0.1508	0.0136	0.1372	0.2646
8- 9	29.4	-0.0040	-0.0002	-0.0000	-0.0038	0.1513	0.0140	0.1372	0.2646
9-10	29.4	-0.0058	-0.0001	-0.0000	-0.0057	0.1514	0.0141	0.1372	0.2646
10-11	29.4	0.0081	-0.0000	0.0000	0.0081	0.1513	0.0141	0.1372	0.2646
11-12	29.4	-0.0073	0.0007	0.0000	-0.0081	0.2051	0.0678	0.1372	0.2646
12-13	29.4	-0.0057	-0.0002	-0.0000	-0.0055	0.1557	0.0184	0.1372	0.2646

TABLE A-II-17.C

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 90.5 K WILSON DATA

EX= 0.0010 EP= 1.0000 ET= 0.2000

PAIR NO	P(av.)	I	II	III	D	±DMAX	DX	DP	DT
1- 2	29.4	-0.0047	-0.0000	0.0000	-0.0046	0.1649	0.0276	0.1372	0.2546
2- 3	29.4	-0.0124	-0.0000	-0.0000	-0.0124	0.1641	0.0268	0.1372	0.2646
3- 4	29.4	0.0035	-0.0000	0.0000	0.0035	0.1541	0.0268	0.1372	0.2546
4- 5	29.4	-0.0196	-0.0000	-0.0000	-0.0196	0.1641	0.0268	0.1372	0.2646
5- 6	29.4	-0.0253	-0.0001	0.0000	-0.0253	0.1647	0.0275	0.1372	0.2646
6- 7	29.4	0.0373	-0.0001	-0.0000	0.0374	0.1648	0.0275	0.1372	0.2546
7- 8	29.4	-0.0056	-0.0001	-0.0000	-0.0054	0.1644	0.0272	0.1372	0.2646
8- 9	29.4	-0.0040	-0.0002	-0.0000	-0.0038	0.1653	0.0280	0.1372	0.2646
9-10	29.4	-0.0058	-0.0001	-0.0000	-0.0057	0.1654	0.0282	0.1372	0.2546
10-11	29.4	0.0081	-0.0000	0.0000	0.0081	0.1654	0.0282	0.1372	0.2646
11-12	29.4	-0.0073	0.0007	0.0000	-0.0081	0.2729	0.1357	0.1372	0.2546
12-13	29.4	-0.0057	-0.0002	-0.0000	-0.0055	0.1741	0.0368	0.1372	0.2646

TABLE A-II-17.D

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 90.5 K WILSON DATA

EX= 0.0020 FP= 1.0000 FT= 0.2000

PAIR NO	P(av.)	I	II	III	D	± DMAX	DX	DP	DT
1- 2	29.4	-0.0047	-0.0000	0.0000	-0.0046	0.1925	0.0552	0.1372	0.2646
2- 3	29.4	-0.0124	-0.0000	-0.0000	-0.0124	0.1909	0.0537	0.1372	0.2646
3- 4	29.4	0.0035	-0.0000	0.0000	0.0035	0.1909	0.0536	0.1372	0.2646
4- 5	29.4	-0.0106	-0.0000	-0.0000	-0.0106	0.1909	0.0536	0.1372	0.2646
5- 6	29.4	-0.0253	-0.0001	0.0000	-0.0253	0.1922	0.0549	0.1372	0.2646
6- 7	29.4	0.0373	-0.0001	-0.0000	0.0374	0.1923	0.0550	0.1372	0.2646
7- 8	29.4	-0.0056	-0.0001	-0.0000	-0.0054	0.1915	0.0544	0.1372	0.2646
8- 9	29.4	-0.0040	-0.0002	-0.0000	-0.0038	0.1933	0.0560	0.1372	0.2645
9-10	29.4	-0.0058	-0.0001	-0.0000	-0.0057	0.1936	0.0564	0.1372	0.2646
10-11	29.4	0.0081	-0.0000	0.0000	0.0081	0.1936	0.0563	0.1372	0.2646
11-12	29.4	-0.0073	0.0007	0.0000	-0.0081	0.4086	0.2713	0.1372	0.2646
12-13	29.4	-0.0057	-0.0002	-0.0000	-0.0055	0.2109	0.0736	0.1372	0.2646

TABLE A-II-18. A

TESTED DATA OF OXYGEN ARGON NITROGEN AT 100.0 K WILSON DATA

ID	P, atm	x_1	x_2	x_3	y_1	y_2	y_3
1	4.0	0.7255	0.0276	0.2469	0.4924	0.0255	0.4821
2	4.0	0.6977	0.0689	0.2334	0.4742	0.0633	0.4625
3	4.0	0.6463	0.1275	0.2262	0.4407	0.1183	0.4410
4	4.0	0.5095	0.2884	0.2021	0.3636	0.2475	0.3889
5	4.0	0.3127	0.5136	0.1738	0.2204	0.4451	0.3345
6	4.0	0.1957	0.6464	0.1579	0.1456	0.5457	0.3087
7	4.0	0.0870	0.7674	0.1447	0.0632	0.6488	0.2880
8	4.0	0.0328	0.8257	0.1415	0.0245	0.6971	0.2784
9	4.0	0.5055	0.2954	0.1991	0.3538	0.2630	0.3831
10	4.0	0.0937	0.7618	0.1445	0.0692	0.6459	0.2849
11	6.0	0.2875	0.0355	0.6770	0.1427	0.0222	0.8351
12	6.0	0.2696	0.0574	0.6730	0.1346	0.0358	0.8296
13	6.0	0.2536	0.0755	0.6710	0.1169	0.0433	0.8398
14	6.0	0.2368	0.0953	0.6679	0.1173	0.0591	0.8236
15	6.0	0.2038	0.1332	0.6630	0.1020	0.0831	0.8149
16	6.0	0.1558	0.1906	0.6536	0.0639	0.0957	0.8404
17	6.0	0.3095	0.0136	0.6769	0.1522	0.0082	0.8396
18	6.0	0.2584	0.0731	0.6685	0.1294	0.0450	0.8257
19	6.0	0.1561	0.1921	0.6518	0.0781	0.1181	0.3038
20	6.0	0.2987	0.0171	0.6843	0.1495	0.0101	0.8404
21	6.0	0.2874	0.0317	0.6809	0.1437	0.0198	0.8365
22	6.0	0.2842	0.0423	0.6735	0.1430	0.0255	0.8315
23	6.0	0.0844	0.2702	0.6454	0.0431	0.7922	0.0844
24	6.0	0.2929	0.0236	0.6835	0.1444	0.0147	0.8409

TABLE A-II-18.B

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 100.0 K WILSON DATA

EX= 0.0005 EP= 1.0000 ET= 0.2000

PAIR NO	P(av.)	I	II	III	D	± DMAX	DX	DP	DT
1- 2	58.8	0.0085	-0.0002	-0.0000	0.0087	0.0822	0.0131	0.0691	0.2646
2- 3	58.8	-0.0012	-0.0002	-0.0000	-0.0009	0.0817	0.0126	0.0691	0.2646
3- 4	58.8	0.0146	-0.0004	-0.0000	0.0150	0.0822	0.0131	0.0691	0.2646
4- 5	58.8	-0.0023	-0.0004	-0.0000	-0.0019	0.0820	0.0129	0.0691	0.2646
5- 6	58.8	0.0023	-0.0001	-0.0000	0.0024	0.0817	0.0125	0.0691	0.2646
6- 7	58.8	-0.0022	0.0000	-0.0000	-0.0022	0.0821	0.0130	0.0691	0.2646
7- 8	58.8	-0.0010	0.0000	-0.0000	-0.0010	0.0825	0.0134	0.0691	0.2646
8- 9	58.8	0.0170	0.0004	0.0000	0.0165	0.0784	0.0293	0.0691	0.2646
9-10	58.8	-0.0111	-0.0005	-0.0000	-0.0106	0.0865	0.0175	0.0691	0.2646
10-11	73.5	-0.7794	-0.7154	0.0165	-0.0806	0.1006	0.0427	0.0578	0.2646
11-12	88.2	0.0021	-0.0001	-0.0000	0.0022	0.0618	0.0152	0.0466	0.2646
12-13	88.2	-0.0325	-0.0000	-0.0000	-0.0325	0.0621	0.0154	0.0466	0.2646
13-14	88.2	0.0288	-0.0001	-0.0000	0.0288	0.0621	0.0154	0.0466	0.2646
14-15	88.2	0.0016	-0.0001	-0.0000	0.0017	0.0617	0.0151	0.0466	0.2646
15-	88.2	-0.0825	0.0000	-0.0000	-0.0926	0.0630	0.0164	0.0466	0.2646 *
16-17	88.2	0.0739	0.0002	0.0000	0.0737	0.0796	0.0330	0.0466	0.2646
17-18	88.2	0.0065	-0.0001	-0.0000	0.0066	0.0664	0.0198	0.0466	0.2646
18-19	88.2	-0.0028	-0.0002	-0.0000	-0.0026	0.0632	0.0166	0.0466	0.2646
19-20	88.2	-0.0137	0.0003	0.0000	-0.0141	0.0749	0.0282	0.0466	0.2646
20-21	88.2	0.0026	-0.0000	-0.0000	0.0026	0.0622	0.0155	0.0466	0.2646
21-22	88.2	0.0077	-0.0000	-0.0000	0.0077	0.0617	0.0150	0.0466	0.2646
22-23	88.2	-2.4612	-0.0011	-0.0001	-2.4600	0.0801	0.0335	0.0466	0.2646 *
23-24	88.2	2.5104	0.0012	0.0001	2.5092	0.0867	0.0400	0.0466	0.2646 *

* inconsistent

TABLE A-II-18. C

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 100.0 K WILSON DATA

EX= 0.0010 EP= 1.0000 FT= 0.2000

PAIR NC	P (av.)	I	II	III	D	± DMAX	DX	DP	DT
1- 2	58.8	0.0085	-0.0002	-0.0000	0.0087	0.0953	0.0262	0.0691	0.2646
2- 3	58.8	-0.0012	-0.0002	-0.0000	-0.0009	0.0943	0.0251	0.0691	0.2646
3- 4	58.8	0.0146	-0.0004	-0.0000	0.0150	0.0952	0.0261	0.0691	0.2646
4- 5	58.8	-0.0023	-0.0004	-0.0000	-0.0019	0.0949	0.0258	0.0691	0.2646
5- 6	58.8	0.0023	-0.0001	-0.0000	0.0024	0.0942	0.0251	0.0691	0.2646
6- 7	58.8	-0.0022	0.0000	-0.0000	-0.0022	0.0952	0.0261	0.0691	0.2645
7- 8	58.8	-0.0010	0.0000	-0.0000	-0.0010	0.0959	0.0268	0.0691	0.2646
8- 9	58.8	0.0170	0.0004	0.0000	0.0165	0.1276	0.0585	0.0691	0.2646
9-10	58.8	-0.0111	-0.0005	-0.0000	-0.0106	0.1041	0.0350	0.0691	0.2645
10-11	73.5	-0.7794	-0.7154	0.0165	-0.0806	0.1433	0.0854	0.0578	0.2646
11-12	88.2	0.0021	-0.0001	-0.0000	0.0022	0.0770	0.0303	0.0466	0.2646
12-13	88.2	-0.0325	-0.0000	-0.0000	-0.0325	0.0775	0.0308	0.0466	0.2646
13-14	88.2	0.0288	-0.0001	-0.0000	0.0288	0.0775	0.0308	0.0466	0.2646
14-15	88.2	0.0016	-0.0001	-0.0000	0.0017	0.0768	0.0301	0.0466	0.2646
15-16	88.2	-0.0825	0.0000	-0.0000	-0.0826	0.0794	0.0327	0.0466	0.2646 *
16-17	88.2	0.0739	0.0002	0.0000	0.0737	0.1126	0.0659	0.0466	0.2646
17-18	88.2	0.0065	-0.0001	-0.0000	0.0066	0.0862	0.0395	0.0466	0.2646
18-19	88.2	-0.0028	-0.0002	-0.0000	-0.0026	0.0798	0.0332	0.0466	0.2646
19-20	88.2	-0.0137	0.0003	0.0000	-0.0141	0.1031	0.0565	0.0466	0.2646
20-21	88.2	0.0026	-0.0000	-0.0000	0.0026	0.0777	0.0311	0.0466	0.2645
21-22	88.2	0.0077	-0.0000	-0.0000	0.0077	0.0767	0.0300	0.0466	0.2646
22-23	88.2	-2.4612	-0.0011	-0.0001	-2.4600	0.1136	0.0669	0.0466	0.2646 *
23-24	88.2	2.5104	0.0012	0.0001	2.5092	0.1267	0.0800	0.0466	0.2646 *

* inconsistent

TABLE A-II-18. D

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 100.0 K WILSON DATA

EX= 0.0020 EP= 1.0000 ET= 0.2000

PAIR NO	P(av.)	I	II	III	D	± DMAX	DX	DP	DT	
1- 2	58.8	0.0085	-0.0002	-0.0000	0.0087	0.1215	0.0524	0.0691	0.2646	
2- 3	58.8	-0.0012	-0.0002	-0.0000	-0.0009	0.1194	0.0503	0.0691	0.2646	
3- 4	58.8	0.0146	-0.0004	-0.0000	0.0150	0.1214	0.0522	0.0691	0.2646	
4- 5	58.8	-0.0023	-0.0004	-0.0000	-0.0019	0.1207	0.0515	0.0691	0.2646	
5- 6	58.8	0.0023	-0.0001	-0.0000	0.0024	0.1193	0.0502	0.0691	0.2646	
6- 7	58.8	-0.0022	0.0000	-0.0000	-0.0022	0.1212	0.0521	0.0691	0.2646	
7- 8	58.8	-0.0010	0.0000	-0.0000	-0.0010	0.1226	0.0535	0.0691	0.2646	
8- 9	58.8	0.0170	0.0004	0.0000	0.0165	0.1862	0.1170	0.0691	0.2646	
9-10	58.8	-0.0111	-0.0005	-0.0000	-0.0106	0.1392	0.0700	0.0691	0.2646	
10-11	73.5	-0.7794	-0.7154	0.0165	-0.0806	0.2287	0.1708	0.0578	0.2646	
11-12	88.2	0.0021	-0.0001	-0.0000	0.0022	0.1073	0.0606	0.0466	0.2646	
12-13	88.2	-0.0325	-0.0000	-0.0000	-0.0325	0.1083	0.0616	0.0465	0.2646	
13-14	88.2	0.0288	-0.0001	-0.0000	0.0288	0.1083	0.0616	0.0456	0.2646	
14-15	88.2	0.0016	-0.0001	-0.0000	0.0017	0.1069	0.0602	0.0466	0.2646	
15-16	88.2	-0.0825	0.0000	-0.0000	-0.0826	0.1121	0.0655	0.0466	0.2646	*
16-17	88.2	0.0739	0.0002	0.0000	0.0737	0.1785	0.1318	0.0465	0.2546	
17-18	88.2	0.0065	-0.0001	-0.0000	0.0066	0.1257	0.0791	0.0466	0.2646	
18-19	88.2	-0.0028	-0.0002	-0.0000	-0.0026	0.1130	0.0663	0.0466	0.2646	
19-20	88.2	-0.0137	0.0003	0.0000	-0.0141	0.1596	0.1129	0.0466	0.2646	
20-21	88.2	0.0026	-0.0000	-0.0000	0.0026	0.1088	0.0621	0.0466	0.2646	
21-22	88.2	0.0077	-0.0000	-0.0000	0.0077	0.1067	0.0601	0.0456	0.2646	
22-23	88.2	-2.4612	-0.0011	-0.0001	-2.4600	0.1805	0.1338	0.0466	0.2646	*
23-24	83.2	2.5104	0.0012	0.0001	2.5092	0.2067	0.1601	0.0466	0.2646	*

* inconsistent

TABLE A-II. 19. A

TESTED DATA OF OXYGEN ARGON NITROGEN AT 110.0 K WILSON DATA

ID	P, atm	x_1	x_2	x_3	y_1	y_2	y_3
1	6.0	0.9259	0.0307	0.0441	0.8584	0.0379	0.1037
2	6.0	0.9118	0.0474	0.0408	0.8452	0.0583	0.0966
3	6.0	0.9116	0.0623	0.0261	0.8371	0.0750	0.0879
4	6.0	0.8852	0.0794	0.0354	0.8228	0.0953	0.0819
5	6.0	0.8714	0.1000	0.0287	0.8092	0.1243	0.0666
6	6.0	0.8249	0.1561	0.0190	0.7618	0.1941	0.0441
7	6.0	0.9369	0.0191	0.0440	0.8727	0.0234	0.1039
8	6.0	0.8996	0.0636	0.0365	0.8357	0.0769	0.0875
9	6.0	0.9416	0.0120	0.0464	0.8752	0.0151	0.1097
10	8.0	0.6795	0.0336	0.2870	0.4880	0.0304	0.4816
11	8.0	0.6616	0.0531	0.2853	0.4733	0.0488	0.4779
12	8.0	0.6468	0.0703	0.2829	0.4651	0.0620	0.4728
13	8.0	0.6345	0.0859	0.2797	0.4543	0.0785	0.4572
14	8.0	0.6037	0.1214	0.2749	0.4337	0.1091	0.4572
15	8.0	0.5583	0.1761	0.2656	0.4020	0.1577	0.4402
16	8.0	0.3200	0.4646	0.2154	0.2378	0.4007	0.3615
17	8.0	0.1923	0.6106	0.1972	0.1466	0.5191	0.3343
18	8.0	0.0382	0.7776	0.1841	0.0295	0.6603	0.3102
19	8.0	0.6514	0.0685	0.2801	0.4673	0.0621	0.4706
20	8.0	0.6898	0.0201	0.2901	0.4961	0.0189	0.4850
21	8.0	0.6898	0.0201	0.2901	0.4961	0.0189	0.4850
22	10.0	0.4224	0.0624	0.5153	0.2633	0.0464	0.6903
23	10.0	0.4263	0.0566	0.5171	0.2644	0.0430	0.6927

	P, atm	x_1	x_2	x_3	y_1	y_2	y_3
24	10.0	0.4124	0.0722	0.5155	0.2555	0.0547	0.6888
25	10.0	0.3993	0.0890	0.5117	0.2482	0.0668	0.6850
26	10.0	0.3668	0.1278	0.5055	0.2298	0.0949	0.6753
27	10.0	0.3198	0.1828	0.4974	0.1993	0.1363	0.6644
28	10.0	0.4674	0.0117	0.5209	0.2891	0.0092	0.7018
29	10.0	0.4488	0.0317	0.5195	0.2802	0.0243	0.6955
30	10.0	0.4574	0.0263	0.5163	0.2872	0.0198	0.6931
31	10.0	0.4408	0.0500	0.5092	0.2766	0.0375	0.6860
32	10.0	0.2182	0.3088	0.4720	0.1388	0.2275	0.6337
33	10.0	0.1382	0.3994	0.4624	0.0839	0.2918	0.6188
34	10.0	0.4687	0.0104	0.5210	0.2921	0.0078	0.4687
35	12.0	0.2362	0.0614	0.7024	0.1385	0.0423	0.8193
36	12.0	0.1953	0.1093	0.6954	0.1154	0.0749	0.8098
37	12.0	0.1604	0.1510	0.6885	0.0950	0.1031	0.8020
38	12.0	0.1048	0.2164	0.6789	0.0621	0.1476	0.7904
39	12.0	0.2741	0.0190	0.7069	0.1609	0.0126	0.8266
40	12.0	0.2544	0.0435	0.7021	0.1470	0.0300	0.8231
41	12.0	0.0559	0.2711	0.6730	0.0330	0.1825	0.7845

TABLE A-II-19. B

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 110.0 K WILSON DATA

EX= 0.0005 EP= 1.0000 FT= 0.2000

PAIR NC	P (av.)	I	II	III	D	± DMAX	DX	DP	DT
1- 2	88.2	0.0000	-0.0000	-0.0000	0.0000	0.0572	0.0108	0.0463	0.2645
2- 3	83.2	0.0041	-0.0000	-0.0000	0.0041	0.0574	0.0111	0.0463	0.2646
3- 4	88.2	-0.0017	-0.0000	0.0000	-0.0016	0.0574	0.0110	0.0463	0.2646
4- 5	88.2	0.0048	-0.0000	-0.0000	0.0048	0.0571	0.0108	0.0463	0.2646
5- 6	88.2	-0.0093	-0.0001	-0.0000	-0.0092	0.0573	0.0109	0.0463	0.2646
6- 7	88.2	0.0136	0.0003	0.0000	0.0134	0.0633	0.0169	0.0463	0.2646
7- 8	89.2	-0.0048	-0.0001	-0.0000	-0.0047	0.0585	0.0122	0.0463	0.2646
8- 9	88.2	0.0029	0.0001	0.0000	0.0028	0.0601	0.0138	0.0463	0.2646
9-10 ~	102.9	-0.5474	-0.4983	0.0146	-0.0638	0.0564	0.0157	0.0407	0.2646*
10-11	117.6	-0.0049	-0.0001	-0.0000	-0.0048	0.0475	0.0124	0.0351	0.2646
11-12	117.6	0.0004	-0.0001	-0.0000	0.0004	0.0474	0.0123	0.0351	0.2646
12-13	117.6	-0.0003	-0.0001	-0.0000	-0.0002	0.0474	0.0123	0.0351	0.2645
13-14	117.6	-0.0017	-0.0002	-0.0000	-0.0016	0.0475	0.0123	0.0351	0.2645
14-15	117.6	-0.0003	-0.0003	-0.0000	-0.0000	0.0475	0.0124	0.0351	0.2646
15-16	117.6	0.0096	-0.0008	-0.0000	0.0105	0.0489	0.0138	0.0351	0.2646
16-17	117.6	0.0018	-0.0001	-0.0000	0.0019	0.0477	0.0126	0.0351	0.2546
17-18	117.6	-0.0010	0.0000	-0.0000	-0.0010	0.0510	0.0159	0.0350	0.2646
18-19	117.6	0.0032	0.0015	0.0001	0.0016	0.0747	0.0396	0.0351	0.2646
19-20	117.6	0.0038	0.0003	0.0000	0.0035	0.0490	0.0139	0.0351	0.2646
20-21	117.6	0.0	0.0	0.0	0.0	0.0472	0.0121	0.0351	0.2646
21-22	132.3	-0.3569	-0.3696	0.0160	-0.0033	0.0466	0.0149	0.0317	0.2646

* inconsistent

22-23	147.0	-0.0017	0.0000	-0.0000	-0.0017	0.0418	0.0134	0.0284	0.2646
23-24	147.0	-0.0006	-0.0001	-0.0000	-0.0006	0.0419	0.0134	0.0284	0.2646
24-25	147.0	-0.0001	-0.0001	-0.0000	0.0000	0.0419	0.0134	0.0284	0.2646
25-26	147.0	0.0016	-0.0002	-0.0000	0.0018	0.0420	0.0136	0.0284	0.2646
26-27	147.0	-0.0025	-0.0002	-0.0000	-0.0022	0.0420	0.0136	0.0283	0.2646
27-28	147.0	0.0132	0.0007	0.0001	0.0124	0.0575	0.0291	0.0283	0.2646
28-29	147.0	0.0009	-0.0001	-0.0000	0.0010	0.0430	0.0145	0.0283	0.2646
29-30	147.0	0.0069	0.0000	0.0000	0.0069	0.0418	0.0134	0.0283	0.2646
30-31	147.0	0.0028	-0.0001	-0.0000	0.0029	0.0423	0.0138	0.0283	0.2646
31-32	147.0	-0.0008	-0.0008	-0.0001	0.0002	0.0475	0.0191	0.0283	0.2646
32-33	147.0	-0.0256	-0.0002	-0.0000	-0.0254	0.0423	0.0139	0.0283	0.2646
33-34	147.0	-0.3639	-0.0296	0.0002	-0.3344	0.0874	0.0590	0.0283	0.2646
34-35	161.7	0.2687	-0.2586	0.0172	0.5101	0.0463	0.0200	0.0261	0.2646*
35-36	176.4	0.0001	-0.0001	-0.0000	0.0003	0.0385	0.0145	0.0239	0.2646*
36-37	176.4	0.0003	-0.0001	-0.0000	0.0004	0.0382	0.0142	0.0239	0.2646
37-38	176.4	-0.0010	-0.0001	-0.0000	-0.0009	0.0384	0.0144	0.0239	0.2646
38-39	176.4	-0.0041	0.0005	0.0001	-0.0047	0.0514	0.0274	0.0239	0.2646
39-40	176.4	-0.0023	-0.0001	-0.0000	-0.0021	0.0391	0.0151	0.0239	0.2646
40-41	176.4	-0.0088	-0.0004	-0.0002	-0.0083	0.0472	0.0232	0.0239	0.2646

* inconsistent

TABLE A-II-19. C

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 110.0 K WILSON DATA

FX= 0.0010 EP= 1.0000 ET= 0.2000

PAIR NO	P(av.)	I	II	III	D	± DMAX	DX	DP	DT
1- 2	88.2	0.0000	-0.0000	-0.0000	0.0000	0.0680	0.0216	0.0463	0.2646
2- 3	88.2	0.0041	-0.0000	-0.0000	0.0041	0.0685	0.0222	0.0463	0.2646
3- 4	88.2	-0.0017	-0.0000	0.0000	-0.0016	0.0684	0.0221	0.0463	0.2646
4- 5	88.2	0.0048	-0.0000	-0.0000	0.0048	0.0679	0.0216	0.0463	0.2646
5- 6	88.2	-0.0093	-0.0001	-0.0000	-0.0092	0.0682	0.0219	0.0463	0.2646
6- 7	88.2	0.0136	0.0003	0.0000	0.0134	0.0802	0.0339	0.0463	0.2646
7- 8	88.2	-0.0048	-0.0001	-0.0000	-0.0047	0.0706	0.0243	0.0463	0.2646
8- 9	88.2	0.0029	0.0001	0.0000	0.0028	0.0740	0.0277	0.0463	0.2646
9-10	102.9	-0.5474	-0.4983	0.0146	-0.0638	0.0722	0.0314	0.0407	0.2646 *
10-11	117.6	-0.0049	-0.0001	-0.0000	-0.0048	0.0600	0.0249	0.0351	0.2646
11-12	117.6	0.0004	-0.0001	-0.0000	0.0004	0.0598	0.0247	0.0351	0.2646
12-13	117.6	-0.0003	-0.0001	-0.0000	-0.0002	0.0597	0.0246	0.0351	0.2646
13-14	117.6	-0.0017	-0.0002	-0.0000	-0.0016	0.0598	0.0247	0.0351	0.2646
14-15	117.6	-0.0003	-0.0003	-0.0000	-0.0000	0.0599	0.0248	0.0351	0.2646
15-16	117.6	0.0096	-0.0008	-0.0000	0.0105	0.0626	0.0275	0.0351	0.2646
16-17	117.6	0.0018	-0.0001	-0.0000	0.0019	0.0603	0.0252	0.0351	0.2646
17-18	117.6	-0.0010	0.0000	-0.0000	-0.0010	0.0670	0.0319	0.0350	0.2646
18-19	117.6	0.0032	0.0015	0.0001	0.0016	0.1144	0.0793	0.0351	0.2646
19-20	117.6	0.0038	0.0003	0.0000	0.0035	0.0630	0.0279	0.0351	0.2646
20-21	117.6	0.0	0.0	0.0	0.0	0.0593	0.0242	0.0351	0.2646
21-22	132.3	-0.3569	-0.3696	0.0150	-0.0033	0.0615	0.0297	0.0317	0.2646

* inconsistent

22-23	147.0	-0.0017	0.0000	-0.0000	-0.0017	0.0552	0.0268	0.0284	0.2646
23-24	147.0	-0.0006	-0.0001	-0.0000	-0.0006	0.0553	0.0269	0.0284	0.2646
24-25	147.0	-0.0001	-0.0001	-0.0000	0.0000	0.0553	0.0269	0.0284	0.2646
25-26	147.0	0.0016	-0.0002	-0.0000	0.0018	0.0556	0.0271	0.0284	0.2646
26-27	147.0	-0.0025	-0.0002	-0.0000	-0.0022	0.0556	0.0271	0.0283	0.2646
27-28	147.0	0.0132	0.0007	0.0001	0.0124	0.0866	0.0582	0.0283	0.2546
28-29	147.0	0.0009	-0.0001	-0.0000	0.0010	0.0575	0.0291	0.0283	0.2646
29-30	147.0	0.0069	0.0000	0.0000	0.0069	0.0552	0.0268	0.0283	0.2646
30-31	147.0	0.0028	-0.0001	-0.0000	0.0029	0.0561	0.0277	0.0283	0.2646
31-32	147.0	-0.0008	-0.0008	-0.0001	0.0002	0.0666	0.0382	0.0283	0.2646
32-33	147.0	-0.0256	-0.0002	-0.0000	-0.0254	0.0561	0.0277	0.0283	0.2646
33-34	147.0	-0.3639	-0.0296	0.0002	-0.3344	0.1464	0.1180	0.0283	0.2546*
34-35	161.7	0.2687	-0.2586	0.0172	0.5101	0.0663	0.0401	0.0261	0.2646*
35-36	176.4	0.0001	-0.0001	-0.0000	0.0003	0.0530	0.0290	0.0239	0.2646
36-37	176.4	0.0003	-0.0001	-0.0000	0.0004	0.0524	0.0284	0.0239	0.2646
37-38	176.4	-0.0010	-0.0001	-0.0000	-0.0009	0.0529	0.0289	0.0239	0.2646
38-39	176.4	-0.0041	0.0005	0.0001	-0.0047	0.0787	0.0547	0.0239	0.2546
39-40	176.4	-0.0023	-0.0001	-0.0000	-0.0021	0.0542	0.0302	0.0239	0.2646
40-41	176.4	-0.0088	-0.0004	-0.0002	-0.0083	0.0704	0.0464	0.0239	0.2646

* inconsistent

TABLE A-II-19. D

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 110.0 K WILSON DATA

EX= 0.0020 EP= 1.0000 ET= 0.2000

PAIR NO	P(av.)	I	II	III	D	± DMAX	DX	DP	DT
1- 2	88.2	0.0000	-0.0000	-0.0000	0.0000	0.0896	0.0433	0.0463	0.2646
2- 3	88.2	0.0041	-0.0000	-0.0000	0.0041	0.0907	0.0444	0.0463	0.2646
3- 4	88.2	-0.0017	-0.0000	0.0000	-0.0016	0.0905	0.0442	0.0463	0.2646
4- 5	88.2	0.0048	-0.0000	-0.0000	0.0048	0.0894	0.0431	0.0463	0.2646
5- 6	88.2	-0.0093	-0.0001	-0.0000	-0.0092	0.0901	0.0438	0.0463	0.2646
6- 7	88.2	0.0136	0.0003	0.0000	0.0134	0.1141	0.0678	0.0463	0.2646
7- 8	88.2	-0.0048	-0.0001	-0.0000	-0.0047	0.0950	0.0487	0.0463	0.2646
8- 9	88.2	0.0029	0.0001	0.0000	0.0028	0.1016	0.0553	0.0463	0.2646
9-10	102.9	-0.5474	-0.4983	0.0146	-0.0638	0.1036	0.0629	0.0407	0.2646 *
10-11	117.6	-0.0049	-0.0001	-0.0000	-0.0048	0.0848	0.0497	0.0351	0.2646
11-12	117.6	0.0004	-0.0001	-0.0000	0.0004	0.0844	0.0493	0.0351	0.2646
12-13	117.6	-0.0003	-0.0001	-0.0000	-0.0002	0.0843	0.0492	0.0351	0.2646
13-14	117.6	-0.0017	-0.0002	-0.0000	-0.0016	0.0845	0.0494	0.0351	0.2646
14-15	117.6	-0.0003	-0.0003	-0.0000	-0.0000	0.0846	0.0495	0.0351	0.2646
15-16	117.6	0.0096	-0.0008	-0.0000	0.0105	0.0901	0.0550	0.0351	0.2646
16-17	117.6	0.0018	-0.0001	-0.0000	0.0019	0.0856	0.0505	0.0351	0.2646
17-18	117.6	-0.0010	0.0000	-0.0000	-0.0010	0.0988	0.0637	0.0350	0.2646
18-19	117.6	0.0032	0.0015	0.0001	0.0016	0.1937	0.1586	0.0351	0.2646
19-20	117.6	0.0038	0.0003	0.0000	0.0035	0.0909	0.0557	0.0351	0.2646
20-21	117.6	0.0	0.0	0.0	0.0	0.0835	0.0484	0.0351	0.2646
21-22	132.3	-0.3569	-0.3696	0.0160	-0.0033	0.0912	0.0594	0.0317	0.2646

* inconsistent

22-23	147.0	-0.0017	0.0000	-0.0000	-0.0017	0.0821	0.0535	0.0284	0.2646
23-24	147.0	-0.0006	-0.0001	-0.0000	-0.0006	0.0821	0.0537	0.0284	0.2646
24-25	147.0	-0.0001	-0.0001	-0.0000	0.0000	0.0821	0.0537	0.0284	0.2646
25-26	147.0	0.0016	-0.0002	-0.0000	0.0018	0.0827	0.0542	0.0284	0.2646
26-27	147.0	-0.0025	-0.0002	-0.0000	-0.0022	0.0827	0.0543	0.0283	0.2646
27-28	147.0	0.0132	0.0007	0.0001	0.0124	0.1448	0.1164	0.0283	0.2646
28-29	147.0	0.0009	-0.0001	-0.0000	0.0010	0.0866	0.0532	0.0283	0.2646
29-30	147.0	0.0069	0.0000	0.0000	0.0069	0.0820	0.0536	0.0283	0.2646
30-31	147.0	0.0028	-0.0001	-0.0000	0.0029	0.0838	0.0554	0.0283	0.2646
31-32	147.0	-0.0008	-0.0008	-0.0001	0.0002	0.1048	0.0764	0.0283	0.2646
32-33	147.0	-0.0256	-0.0002	-0.0000	-0.0254	0.0839	0.0555	0.0283	0.2646
33-34	147.0	-0.3639	-0.0296	0.0002	-0.3344	0.2645	0.2360	0.0283	0.2646*
34-35	161.7	0.2687	-0.2586	0.0172	0.5101	0.1064	0.0802	0.0261	0.2646*
35-36	176.4	0.0001	-0.0001	-0.0000	0.0003	0.0820	0.0580	0.0239	0.2646
36-37	176.4	0.0003	-0.0001	-0.0000	0.0004	0.0808	0.0568	0.0239	0.2646
37-38	176.4	-0.0010	-0.0001	-0.0000	-0.0009	0.0818	0.0577	0.0239	0.2646
38-39	176.4	-0.0041	0.0005	0.0001	-0.0047	0.1335	0.1095	0.0239	0.2646
39-40	176.4	-0.0023	-0.0001	-0.0000	-0.0021	0.0844	0.0604	0.0239	0.2646
40-41	176.4	-0.0088	-0.0004	-0.0002	-0.0083	0.1169	0.0928	0.0239	0.2646

* inconsistent

TABLE A-II-20. A

TESTED DATA OF OXYGEN ARGON NITROGEN AT 120.0 K WILSON DATA

ID	P, atm	X_1	X_2	x_3	y_1	y_2	y_3
1	16.0	0.5480	0.0767	0.3752	0.4070	0.0657	0.5273
2	16.0	0.5034	0.1322	0.3644	0.3731	0.1136	0.5133
3	16.0	0.3192	0.3391	0.3417	0.2401	0.2890	0.4709
4	16.0	0.1311	0.5548	0.3140	0.1009	0.4651	0.4341
5	16.0	0.0414	0.6581	0.3005	0.0317	0.5492	0.4192
6	18.0	0.4425	0.0260	0.5315	0.3120	0.0213	0.6667
7	18.0	0.3886	0.0922	0.5193	0.2727	0.0749	0.6524
8	18.0	0.3667	0.1402	0.4931	0.2499	0.1082	0.6418
9	18.0	0.2365	0.2732	0.4902	0.1684	0.2184	0.6132
10	18.0	0.1091	0.4188	0.4721	0.0794	0.3326	0.5880
11	18.0	0.3558	0.1342	0.5099	0.2502	0.1093	0.6404
12	23.0	0.1105	0.0273	0.8622	0.0744	0.0201	0.9054
13	23.0	0.0580	0.1216	0.8503	0.0172	0.0919	0.8909
14	23.0	0.0708	0.0743	0.8550	0.0488	0.0559	0.8954
15	23.0	0.0669	0.0076	0.9255	0.0454	0.0054	0.9492
16	23.0	0.0532	0.0226	0.9242	0.0366	0.0168	0.9466
17	23.0	0.0431	0.0347	0.9223	0.0295	0.0243	0.9462
18	23.0	0.0349	0.0467	0.9184	0.0235	0.0336	0.9429
19	23.0	0.0210	0.0604	0.9187	0.0148	0.0438	0.9414
20	23.0	0.0075	0.0767	0.9158	0.0049	0.0562	0.9390
21	23.0	0.0411	0.0377	0.9212	0.0280	0.0273	0.9447
22	23.0	0.0653	0.0757	0.8591	0.0451	0.0572	0.8977

TABLE A-II-20.B

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 120.0 K WILSON DATA

FX= 0.0005 FP= 1.0000 ET= 0.2000

PAIR NO	P(av.)	I	II	III	D	± DMAX	DX	DP	DI
1- 2	235.1	0.0002	-0.0004	-0.0000	0.0006	0.0310	0.0128	0.0181	0.2646
2- 3	235.1	-0.0072	-0.0009	-0.0001	-0.0061	0.0320	0.0138	0.0181	0.2646
3- 4	235.1	-0.0023	-0.0003	-0.0012	-0.0019	0.0319	0.0137	0.0181	0.2646
4- 5	235.1	-0.0008	0.0001	-0.0001	-0.0009	0.0324	0.0142	0.0181	0.2646
5- 6	249.8	-0.1410	-0.1681	0.0172	0.0099	0.0663	0.0490	0.0172	0.2646
6- 7	264.5	-0.0033	-0.0004	-0.0001	-0.0028	0.0313	0.0149	0.0163	0.2646
7- 8	264.5	0.0018	-0.0002	-0.0001	0.0021	0.0297	0.0133	0.0163	0.2646
8- 9	264.5	0.0020	-0.0006	-0.0001	0.0027	0.0303	0.0139	0.0163	0.2646
9-10	264.5	-0.0012	-0.0002	-0.0012	-0.0009	0.0303	0.0139	0.0163	0.2646
10-11	264.5	0.0062	0.0007	0.0003	0.0052	0.0328	0.0164	0.0163	0.2646
11-12	301.3	-0.2822	-0.3202	0.0470	-0.0089	0.0341	0.0193	0.0147	0.2646
12-13	338.0	-0.1381	-0.0002	0.0048	-0.1427	0.0345	0.0209	0.0134	0.2646 *
13-14	338.0	0.1069	0.0001	-0.0049	0.1118	0.0304	0.0168	0.0134	0.2646 *
14-15	338.0	-0.0440	0.0000	0.0009	-0.0449	0.0367	0.0232	0.0132	0.2646 *
15-16	338.0	0.0005	-0.0000	-0.0000	0.0006	0.0288	0.0154	0.0132	0.2646
16-17	338.0	-0.0009	-0.0000	-0.0000	-0.0009	0.0274	0.0140	0.0132	0.2646
17-18	338.0	0.0023	-0.0000	-0.0001	0.0024	0.0274	0.0139	0.0132	0.2646
18-19	338.0	-0.0001	-0.0000	-0.0000	-0.0001	0.0275	0.0141	0.0132	0.2646
19-20	338.0	0.0004	0.0000	-0.0001	0.0005	0.0544	0.0409	0.0132	0.2646
20-21	338.0	0.0010	0.0000	0.0001	0.0009	0.0762	0.0627	0.0132	0.2646
21-	338.0	0.0397	-0.0000	-0.0007	0.0404	0.0280	0.0145	0.0132	0.2646

* inconsistent

TABLE A-II-20. C

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 120.0 K WILSON DATA

FX= 0.0020 EP= 1.0000 ET= 0.2000

PAIR NO	P (av.)	I	II	III	D	± DMAX	DX	DP	DT
1- 2	235.1	0.0002	-0.0004	-0.0000	0.0006	0.0693	0.0512	0.0181	0.2646
2- 3	235.1	-0.0072	-0.0009	-0.0001	-0.0061	0.0733	0.0551	0.0181	0.2646
3- 4	235.1	-0.0023	-0.0003	-0.0002	-0.0019	0.0731	0.0549	0.0181	0.2646
4- 5	235.1	-0.0008	0.0001	-0.0001	-0.0009	0.0750	0.0568	0.0181	0.2646
5- 6	249.8	-0.1410	-0.1681	0.0172	0.0099	0.2135	0.1962	0.0172	0.2646
6- 7	264.5	-0.0033	-0.0004	-0.0001	-0.0028	0.0762	0.0598	0.0163	0.2646
7- 8	264.5	0.0018	-0.0002	-0.0001	0.0021	0.0596	0.0532	0.0163	0.2646
8- 9	264.5	0.0020	-0.0006	-0.0001	0.0027	0.0719	0.0555	0.0163	0.2646
9-10	264.5	-0.0012	-0.0002	-0.0002	-0.0009	0.0719	0.0555	0.0163	0.2646
10-11	264.5	0.0062	0.0007	0.0003	0.0052	0.0820	0.0656	0.0163	0.2646
11-12	301.3	-0.2822	-0.3202	0.2470	-0.0089	0.0919	0.0770	0.0147	0.2646
12-13	338.0	-0.1381	-0.0002	0.0048	-0.1427	0.0971	0.0834	0.0134	0.2645 *
13-14	338.0	0.1069	0.0001	-0.0049	0.1118	0.0808	0.0671	0.0134	0.2646 *
14-15	338.0	-0.0440	0.0000	0.0009	-0.0449	0.1063	0.0928	0.0132	0.2646 *
15-16	338.0	0.0005	-0.0000	-0.0000	0.0006	0.0749	0.0614	0.0132	0.2646
16-17	338.0	-0.0009	-0.0000	-0.0000	-0.0009	0.0693	0.0559	0.0132	0.2646
17-18	338.0	0.0023	-0.0000	-0.0001	0.0024	0.0692	0.0557	0.0132	0.2646
18-19	338.0	-0.0001	-0.0000	-0.0000	-0.0001	0.0697	0.0563	0.0132	0.2646
19-20	338.0	0.0004	0.0000	-0.0001	0.0005	0.1770	0.1636	0.0132	0.2646
20-21	338.0	0.0010	0.0000	0.0001	0.0009	0.2642	0.2507	0.0132	0.2646
21-22	338.0	0.0397	-0.0000	-0.0007	0.0404	0.0715	0.0581	0.0132	0.2646

* inconsistent

TABLE A-II-20. D

TESTED RESULTS OF OXYGEN ARGON NITROGEN AT 120.0 K WILSON DATA

EX= 0.0010 EP= 1.0000 ET= 0.2000

PAIR NC	P (av.)	I	II	III	D	±DMAX	DX	DP	DT
1- 2	235.1	0.0002	-0.0004	-0.0000	0.0006	0.0438	0.0256	0.0181	0.2646
2- 3	235.1	-0.0072	-0.0009	-0.0001	-0.0061	0.0457	0.0276	0.0181	0.2646
3- 4	235.1	-0.0023	-0.0003	-0.0002	-0.0019	0.0456	0.0275	0.0181	0.2646
4- 5	235.1	-0.0008	0.0001	-0.0001	-0.0009	0.0466	0.0284	0.0181	0.2646
5- 6	249.8	-0.1410	-0.1681	0.0172	0.0099	0.1154	0.0981	0.0172	0.2646
6- 7	264.5	-0.0033	-0.0004	-0.0001	-0.0028	0.0463	0.0299	0.0163	0.2646
7- 8	264.5	0.0018	-0.0002	-0.0001	0.0021	0.0430	0.0266	0.0163	0.2646
8- 9	264.5	0.0020	-0.0005	-0.0001	0.0027	0.0441	0.0277	0.0163	0.2646
9-10	264.5	-0.0012	-0.0002	-0.0002	-0.0009	0.0441	0.0278	0.0163	0.2646
10-11	264.5	0.0062	0.0007	0.0003	0.0052	0.0492	0.0328	0.0163	0.2646
11-12	301.3	-0.2822	-0.3202	0.0470	-0.0089	0.0534	0.0385	0.0147	0.2646 *
12-13	338.0	-0.1381	-0.0002	0.0048	-0.1427	0.0554	0.0417	0.0134	0.2646 *
13-14	338.0	0.1069	0.0001	-0.0049	0.1118	0.0472	0.0336	0.0134	0.2646 *
14-15	338.0	-0.0440	0.0000	0.0009	-0.0449	0.0599	0.0464	0.0132	0.2646
15-16	338.0	0.0005	-0.0000	-0.0000	0.0006	0.0442	0.0307	0.0132	0.2646
16-17	338.0	-0.0009	-0.0000	-0.0000	-0.0009	0.0414	0.0279	0.0132	0.2646
17-18	338.0	0.0023	-0.0000	-0.0001	0.0024	0.0413	0.0279	0.0132	0.2646
18-19	338.0	-0.0001	-0.0000	-0.0000	-0.0001	0.0416	0.0281	0.0132	0.2646
19-20	338.0	0.0004	0.0000	-0.0001	0.0005	0.0953	0.0818	0.0132	0.2646
20-21	338.0	0.0010	0.0000	0.0001	0.0009	0.1388	0.1254	0.0132	0.2646
21-22	338.0	0.0397	-0.0000	-0.0007	0.0404	0.0425	0.0290	0.0132	0.2646

* inconsistent

TABLE A-II. 21

Comparison of Calculated and Experimental γ Values for the System Nitrogen - Argon - Oxygen at 90.5°K

P_{psia}	x_A	x_{O_2}	γ_A		γ_{O_2}		γ_{N_2}	
			Expt$^{(82)}$	Calc	Expt$^{(82)}$	Calc	Expt$^{(82)}$	Calc
51.16	0.0990	0.0373	1.1435	1.2037	1.1842	1.1532	1.0023	1.0040
47.12	0.1512	0.0868	1.1219	1.1646	1.1594	1.1245	1.0069	1.0110
44.46	0.2296	0.0861	1.1012	1.1290	1.1514	1.1146	1.0127	1.0204
36.31	0.4202	0.1122	1.0561	1.0624	1.1326	1.0995	1.0400	1.0582
31.29	0.4965	0.1704	1.0396	1.0418	1.1169	1.0901	1.0643	1.0853
28.97	0.5931	0.1179	1.0270	1.0270	1.1343	1.1088	1.0794	1.1041
25.59	0.6089	0.2033	1.0234	1.0233	1.1140	1.0955	1.1019	1.1231
22.84	0.6754	0.2046	1.0165	1.0160	1.1212	1.1046	1.1250	1.1446
20.44	0.7388	0.1924	1.0112	1.0106	1.1320	1.1169	1.1465	1.1639
20.43	0.7599	0.1698	1.0094	1.0881	1.1398	1.1239	1.1489	1.1672
38.09	0.2939	0.2049	1.0765	1.0912	1.1105	1.0780	1.0321	1.0442
32.46	0.4212	0.2271	1.0506	1.0555	1.1016	1.0750	1.0578	1.0751
24.53	0.5892	0.2554	1.0262	1.0261	1.1021	1.0853	1.1085	1.1266
41.88	0.1322	0.2754	1.1109	1.1449	1.1022	1.0713	1.0229	1.0278
33.46	0.3257	0.3060	1.0669	1.0764	1.0832	1.0577	1.0540	1.0662
34.37	0.1952	0.4317	1.0946	1.1121	1.0592	1.0375	1.0586	1.0623
34.86	0.2175	0.3887	1.0890	1.1058	1.0676	1.0438	1.0526	1.0587
26.45	0.4013	0.4172	1.0568	1.0592	1.0604	1.0458	1.0981	1.1052
28.60	0.2294	0.5548	1.0956	1.1032	1.0354	1.0229	1.0992	1.0940
30.27	0.1517	0.5977	1.1146	1.1275	1.0303	1.0178	1.0973	1.0879
26.40	0.0938	0.7677	1.1515	1.1551	1.0104	1.0057	1.1514	1.1172

TABLE A-II. 22

Comparison of Calculated and Experimental γ Values for the System Nitrogen - Argon - Oxygen at 100.0°K

P_{psia}	x_A	x_{O_2}	γ_A		γ_{O_2}		γ_{N_2}	
			Expt(82)	Calc	Expt(82)	Calc	Expt(82)	Calc
104.34	0.1073	0.0393	1.1201	1.1713	1.1582	1.1317	1.0025	1.0039
97.77	0.1666	0.0929	1.0991	1.1355	1.1342	1.1056	1.0075	1.0112
90.44	0.2627	0.0896	1.0784	1.1007	1.1259	1.0977	1.0145	1.0220
92.86	0.2315	0.0865	1.0854	1.1118	1.1295	1.1008	1.0117	1.0178
66.97	0.5019	0.1711	1.0323	1.0362	1.0974	1.0805	1.0577	1.0752
55.64	0.6869	0.1130	1.0132	1.0134	1.1176	1.1078	1.0926	1.1151
51.36	0.6757	0.2045	1.0136	1.0143	1.0997	1.0932	1.1085	1.1259
46.73	0.7654	0.1683	1.0074	1.0076	1.1154	1.1112	1.1299	1.1472
79.37	0.3020	0.2086	1.0622	1.0778	1.0925	1.0686	1.0304	1.0399
68.48	0.4601	0.2058	1.0370	1.0425	1.0896	1.0722	1.0550	1.0705
87.03	0.1417	0.2896	1.0894	1.1221	1.0838	1.0595	1.0233	1.0267
82.61	0.1598	0.3326	1.0843	1.1130	1.0725	1.0501	1.0309	1.0343
78.76	0.1681	0.3859	1.0820	1.1079	1.0608	1.0409	1.0403	1.0429
70.95	0.3368	0.3105	1.0535	1.0650	1.0692	1.0512	1.0508	1.0600
60.22	0.4806	0.3008	1.0338	1.0380	1.0714	1.0599	1.0772	1.0897
50.70	0.5414	0.3750	1.0299	1.0315	1.0626	1.0578	1.1116	1.1181
51.58	0.4617	0.4501	1.0420	1.0444	1.0478	1.0431	1.1107	1.1118
58.62	0.3672	0.4566	1.0529	1.0597	1.0438	1.0351	1.0890	1.0909
71.45	0.1234	0.5474	1.0944	1.1197	1.0342	1.0212	1.0681	1.0628
61.25	0.1224	0.6964	1.1086	1.1250	1.0149	1.0090	1.1115	1.0917
57.86	0.0955	0.7759	1.1246	1.1375	1.0081	1.0048	1.1135	1.1047

TABLE A-II. 23

Comparison of Calculated and Experimental γ Values for the System Nitrogen - Argon - Oxygen at 110.0°K

P_{psia}	x_A	x_{O_2}	γ_A Expt(82)	γ_A Calc	γ_{O_2} Expt(82)	γ_{O_2} Calc	γ_{N_2} Expt(82)	γ_{N_2} Calc
197.51	0.1189	0.0427	1.1023	1.1459	1.1399	1.1106	1.0032	1.0042
171.50	0.2667	0.0904	1.0670	1.0883	1.1098	1.0823	1.0143	1.0201
144.90	0.4487	0.1155	1.0363	1.0435	1.0934	1.0728	1.0372	1.0505
124.03	0.5999	0.1180	1.0186	1.0207	1.0925	1.0791	1.0641	1.0829
120.07	0.6292	0.1184	1.0158	1.0174	1.0933	1.0813	1.0705	1.0900
106.90	0.7493	0.0792	1.0075	1.0077	1.1064	1.0983	1.0934	1.1161
97.95	0.8373	0.0548	1.0032	1.0031	1.1181	1.1127	1.1146	1.1381
93.26	0.7649	0.1897	1.0064	1.0066	1.0914	1.0882	1.1205	1.1376
101.52	0.6925	0.2029	1.0102	1.0109	1.0833	1.0778	1.1019	1.1190
190.61	0.1085	0.1167	1.0951	1.1374	1.1199	1.0920	1.0059	1.0069
162.24	0.2485	0.1935	1.0618	1.0880	1.0867	1.0624	1.0215	1.0267
146.23	0.3411	0.2006	1.0469	1.0599	1.0797	1.0584	1.0325	1.0411
132.85	0.4576	0.2120	1.0311	1.0374	1.0744	1.0584	1.0513	1.0639
163.57	0.1500	0.2965	1.0727	1.1028	1.0719	1.0499	1.0239	1.0254
155.63	0.1689	0.3459	1.0679	1.0941	1.0605	1.0410	1.0321	1.0333
126.97	0.3712	0.3689	1.0400	1.0487	1.0489	1.0369	1.0636	1.0693
113.16	0.4899	0.3029	1.0268	1.0312	1.0595	1.0494	1.0733	1.0840
100.33	0.5562	0.3680	1.0231	1.0245	1.0532	1.0492	1.1035	1.1109
97.92	0.5025	0.4601	1.0319	1.0326	1.0404	1.0382	1.1141	1.1144
150.63	0.1736	0.3883	1.0664	1.0908	1.0524	1.0348	1.0390	1.0393
137.26	0.2121	0.4550	1.0613	1.0796	1.0395	1.0261	1.0557	1.0545
139.76	0.2339	0.4080	1.0576	1.0754	1.0460	1.0310	1.0498	1.0509
114.80	0.3745	0.4586	1.0424	1.0488	1.0367	1.0291	1.0839	1.0848
137.11	0.1280	0.5560	1.0763	1.0993	1.0284	1.0176	1.0656	1.0581
126.97	0.1570	0.6109	1.0759	1.0928	1.0207	1.0129	1.0832	1.0719
121.68	0.2359	0.5659	1.0633	1.0751	1.0242	1.0167	1.0846	1.0770
114.61	0.0966	0.7790	1.1013	1.1128	1.0068	1.0041	1.1238	1.0949

TABLE A-II. 24

Comparison of Calculated and Experimental γ Values for the System Nitrogen - Argon - Oxygen at 120.0°K

P_{psia}	x_A	x_{O_2}	γ_A		γ_{O_2}		γ_{N_2}	
			Expt$^{(82)}$	Calc	Expt$^{(82)}$	Calc	Expt$^{(82)}$	Calc
336.54	0.1201	0.0432	1.0494	1.13607	1.1351	1.1019	1.0048	1.0039
289.07	0.3857	0.0950	1.0421	1.0556	1.0900	1.0668	1.0284	1.0350
246.89	0.4601	0.1176	1.0306	1.0398	1.0809	1.0619	1.0409	1.0503
215.00	0.6145	0.1182	1.0148	1.0182	1.0780	1.0656	1.0674	1.0838
211.03	0.5399	0.2356	1.0183	1.0228	1.0598	1.0495	1.0721	1.0842
193.11	0.6518	0.2034	1.0106	1.0125	1.0666	1.0596	1.0903	1.1076
181.64	0.7090	0.1992	1.0076	1.0085	1.0699	1.0650	1.1037	1.1233
195.75	0.7133	0.1105	1.0079	1.0093	1.0816	1.0731	1.0877	1.1085
178.41	0.8887	0.0554	1.0012	1.0013	1.1009	1.0971	1.1253	1.1529
322.87	0.1207	0,1274	1.0831	1.1220	1.1114	1.0825	1.0089	1.0077
289.66	0.2365	0.1509	1.0596	1.0847	1.0905	1.0656	1.0189	1.0199
273.49	0.2623	0.2074	1.0509	1.0726	1.0755	1.0542	1.0271	1.0284
235.14	0.4260	0.2292	1.0291	1.0621	1.0471	1.0510	1.0510	1.0582
262.32	0.1779	0.3611	1.0550	1.0808	1.0523	1.0367	1.0383	1.0349
196.93	0.5768	0.2668	1.0155	1.0184	1.0562	1.0489	1.0857	1.0989
277.61	0.1559	0.3039	1.0606	1.0907	1.0649	1.0463	1.0282	1.0254
232.93	0.3572	0.3250	1.0343	1.0459	1.0489	1.0360	1.0558	1.0589
203.54	0.4894	0.3017	1.0220	1.0274	1.0504	1.0504	1.0745	1.0834
255.56	0.1822	0.4046	1.0535	1.0772	1.0446	1.0310	1.0456	1.0413
244.25	0.2345	0.4056	1.0472	1.0658	1.0415	1.0289	1.0523	1.0494
202.22	0.3791	0.4573	1.0339	1.0404	1.0313	1.0248	1.0860	1.0866
201.34	0.4238	0.4278	1.0294	1.0346	1.0347	1.0285	1.0880	1.0912
236.02	0.1324	0.5703	1.0616	1.0827	1.0238	1.0160	1.0714	1.0600
212.21	0.2441	0.5731	1.0508	1.0613	1.0202	1.0145	1.0892	1.0806
209.56	0.1280	0.7170	1.0723	1.0843	1.0097	1.0064	1.1084	1.0882

TABLE A-II. 25

Comparison of Calculated and Experimental γ Values for the System Nitrogen - Argon - Oxygen at 90.5°K

P_{psia}	x_{O_2}	x_A	γ_{O_2} Expt (83)	γ_{O_2} Calc	γ_A Expt (83)	γ_A Calc	γ_{N_2} Expt (83)	γ_{N_2} Calc
29.39	0.6506	0.0131	1.0310	1.0233	1.1401	1.1377	1.0981	1.1061
29.39	0.6397	0.0286	1.0319	1.0240	1.1356	1.1329	1.0966	1.1049
29.39	0.6267	0.0402	1.0334	1.0253	1.1318	1.1289	1.0939	1.1026
29.39	0.6181	0.0531	1.0341	1.0259	1.1283	1.1251	1.0930	1.1019
29.39	0.6010	0.0666	1.0364	1.0279	1.1237	1.1204	1.0894	1.0988
29.39	0.5795	0.0969	1.0386	1.0300	1.1157	1.1119	1.0871	1.0969
29.39	0.5410	0.1370	1.0438	1.0349	1.1045	1.1002	1.0817	1.0922
29.39	0.4761	0.2092	1.0537	1.0445	1.0867	1.0814	1.0749	1.0861
29.39	0.3302	0.3768	1.0826	1.0742	1.0530	1.0468	1.0690	1.0799
29.39	0.1792	0.5469	1.1231	1.1175	1.0287	1.0229	1.0743	1.0821
29.39	0.0859	0.6540	1.1540	1.1513	1.0184	1.0135	1.0844	1.0887
29.39	0.6485	0.0150	1.0313	1.0235	1.1394	1.1370	1.0976	1.1057
29.39	0.5778	0.0952	1.0390	1.0303	1.1158	1.1120	1.0864	1.0962

TABLE A-II. 26

Comparison of Calculated and Experimental γ Values for the System Nitrogen - Argon - Oxygen at 100.0°K

P_{psia}	x_{O_2}	x_A	γ_{O_2} Expt$^{(83)}$	γ_{O_2} Calc	γ_A Expt$^{(83)}$	γ_A Calc	γ_{N_2} Expt$^{(83)}$	γ_{N_2} Calc
58.78	0.7255	0.0276	1.0160	1.0118	1.1212	1.1274	1.1124	1.1146
58.78	0.6977	0.0687	1.0176	1.0133	1.1114	1.1165	1.1093	1.1121
58.78	0.6463	0.1275	1.0220	1.0337	1.0968	1.1006	1.1018	1.1058
58.78	0.5095	0.2884	1.0380	1.0337	1.0629	1.0639	1.0890	1.0947
58.78	0.3127	0.5136	1.0736	1.0719	1.0283	1.0271	1.0857	1.0901
58.78	0.1957	0.6464	1.1022	1.1035	1.0147	1.0131	1.0923	1.0938
58.78	0.0879	0.7674	1.1337	1.1390	1.0067	1.0052	1.1041	1.1016
58.78	0.0328	0.8257	1.1515	1.1595	1.0044	1.0030	1.1113	1.1064
58.78	0.5055	0.2954	1.0386	1.0343	1.0617	1.0627	1.0893	1.0949
58.78	0.0937	0.7618	1.1319	1.1370	1.0070	1.0050	1.1036	1.1013
88.18	0.2875	0.0355	1.1017	1.0996	1.1020	1.1108	1.0163	1.0212
88.18	0.2696	0.0574	1.1042	1.1026	1.0985	1.1059	1.0156	1.0204
88.18	0.2536	0.0755	1.1066	1.1056	1.0957	1.1020	1.0150	1.0197
88.18	0.2368	0.0953	1.1092	1.1088	1.0927	1.0979	1.0145	1.0191
88.18	0.2038	0.1332	1.1146	1.1156	1.0873	1.0906	1.0137	1.0182
88.18	0.1558	0.1906	1.1227	1.1264	1.0795	1.0805	1.0135	1.0179
88.18	0.3095	0.0136	1.0983	1.0955	1.1054	1.1156	1.0176	1.0228
88.18	0.2548	0.0731	1.1056	1.1043	1.0958	1.1023	1.0153	1.0201
88.18	0.1561	0.1921	1.1225	1.1262	1.0792	1.0801	1.0136	1.0180
88.18	0.2987	0.0171	1.1008	1.0985	1.1052	1.1153	1.0166	1.0215
88.18	0.2874	0.0317	1.1022	1.1002	1.1028	1.1119	1.0161	1.0210
88.18	0.2842	0.0423	1.1019	1.0998	1.1007	1.1091	1.0163	1.0213
88.18	0.0844	0.2702	1.1368	1.1457	1.0704	1.0689	1.0142	1.0185
88.18	0.2929	0.0236	1.1016	1.0995	1.1042	1.1138	1.0162	1.0212

TABLE A-II. 27

Comparison of Calculated and Experimental γ Values for the System Nitrogen - Argon - Oxygen at 110.0°K

P_{psia}	x_{O_2}	x_A	γ_{O_2} Expt(83)	γ_{O_2} Calc	γ_A Expt(83)	γ_A Calc	γ_{N_2} Expt(83)	γ_{N_2} Calc
88.18	0.9259	0.0307	1.0009	1.0006	1.1332	1.1291	1.1802	1.1622
88.18	0.9118	0.0474	1.0012	1.0009	1.1286	1.1249	1.1772	1.1602
88.18	0.9116	0.0623	1.0011	1.0009	1.1276	1.1232	1.1805	1.1624
88.18	0.8852	0.0794	1.0019	1.0015	1.1203	1.1170	1.1720	1.1566
88.18	0.8714	0.1000	1.0023	1.0019	1.1158	1.1127	1.1704	1.1555
88.18	0.8249	0.1561	1.0044	1.0038	1.1024	1.1000	1.1625	1.1501
88.18	0.9369	0.0191	1.0007	1.0005	1.1367	1.1323	1.1829	1.1640
88.18	0.8996	0.0636	1.0015	1.0012	1.1247	1.1211	1.1751	1.1588
88.18	0.9416	0.0120	1.0007	1.0004	1.1364	1.1339	1.1837	1.1645
117.57	0.6795	0.0336	1.0189	1.0150	1.0921	1.0977	1.0884	1.0916
117.57	0.6616	0.0531	1.0201	1.0161	1.0881	1.0932	1.0860	1.0895
117.57	0.6468	0.0703	1.0211	1.0170	1.0847	1.0895	1.0843	1.0880
117.57	0.6345	0.0789	1.0219	1.0178	1.0818	1.0862	1.0830	1.0870
117.57	0.6037	0.1214	1.0244	1.0201	1.0752	1.0789	1.0798	1.0842
117.57	0.5583	0.1761	1.0283	1.0241	1.0658	1.0684	1.0760	1.0810
117.57	0.3000	0.4640	1.0594	1.0578	1.0273	1.0264	1.0704	1.0749
117.57	0.1923	0.6106	1.0834	1.0851	1.0147	1.0131	1.0752	1.0776
117.57	0.0382	0.7776	1.1992	1.1270	1.0061	1.0046	1.0874	1.0859
117.57	0.6514	0.0685	1.0206	1.0166	1.0854	1.0901	1.0853	1.0889
117.57	0.6898	0.0201	1.0183	1.0145	1.0948	1.1007	1.0896	1.0925

TABLE A-II. 27 (cont'd)

P_{psia}	x_{O_2}	x_A	γ_{O_2} Expt	γ_{O_2} Calc	γ_A Expt	γ_A Calc	γ_{N_2} Expt	γ_{N_2} Calc
146.96	0.4224	0.0624	1.0578	1.0527	1.0767	1.0824	1.0350	1.0404
146.96	0.4263	0.0566	1.0575	1.0524	1.0776	1.0835	1.0352	1.0405
146.96	0.4124	0.0722	1.0590	1.0539	1.0753	1.0806	1.0343	1.0396
146.96	0.3993	0.0890	1.0602	1.0553	1.0729	1.0775	1.0337	1.0390
146.96	0.3668	0.1278	1.0638	1.0593	1.0675	1.0706	1.0322	1.0375
146.96	0.3198	0.1828	1.0694	1.0659	1.0605	1.0618	1.0306	1.0357
146.96	0.4674	0.0117	1.0533	1.0480	1.0844	1.0924	1.0383	1.0437
146.96	0.4488	0.0317	1.0552	1.0499	1.0813	1.0883	1.0368	1.0422
146.96	0.4574	0.0263	10540	1.0487	1.0822	1.0894	1.0378	1.0432
146.96	0.4408	0.0500	1.0552	1.0499	1.0785	1.0847	1.0370	1.0424
146.96	0.2182	0.3088	1.0832	1.0830	1.0462	1.0494	1.0303	1.0353
146.96	0.1382	0.3994	1.0965	1.1000	1.0381	1.0349	1.0316	1.0363
146.96	0.4687	0.0104	1.0532	1.0478	1.0846	1.0926	1.0384	1.0438
176.35	0.2362	0.0614	1.1006	1.1012	1.0848	1.0914	1.0121	1.0146
176.35	0.1953	0.1093	1.1058	1.1078	1.0794	1.0838	1.0113	1.0136
176.35	0.1604	0.1510	1.1105	1.1140	1.0750	1.0778	1.0110	1.0132
176.35	0.1084	0.2164	1.1186	1.1252	1.0686	1.0692	1.0111	1.0134
176.35	0.2741	0.0190	1.0959	1.0954	1.0878	1.0986	1.0134	1.0162
176.35	0.2544	0.0435	1.0980	1.0980	1.0867	1.0942	1.0128	1.0155
176.35	0.0559	0.2711	1.1266	1.1366	1.0641	1.0634	1.0118	1.0142

TABLE A-II. 28

Comparison of Calculated and Experimental γ Values for the System Nitrogen - Argon - Oxygen at 120.0°K

P_{psia}	x_{O_2}	x_A	γ_{O_2} Expt (83)	γ_{O_2} Calc	γ_A Expt (83)	γ_A Calc	γ_{N_2} Expt (83)	γ_{N_2} Calc
235.14	0.5480	0.0767	1.0316	1.0288	1.0625	1.0684	1.0620	1.0629
235.14	0.5034	0.1322	1.0348	1.0319	1.0554	1.0601	1.0593	1.0606
235.14	0.3192	0.3391	1.0535	1.0521	1.0334	1.0347	1.0520	1.0540
235.14	0.1311	0.5548	1.0799	1.0833	1.0180	1.0174	1.0557	1.0571
235.14	0.0414	0.6581	1.0953	1.1024	1.0132	1.0124	1.0613	1.0621
264.53	0.4425	0.0260	1.0533	1.0509	1.0682	1.0767	1.0373	1.0376
264.53	0.3886	0.0922	1.0576	1.0553	1.0607	1.0671	1.0348	1.0352
264.53	0.3667	0.1402	1.0576	1.0555	1.0551	1.0601	1.0360	1.0366
264.53	0.2365	0.2732	1.0730	1.0731	1.0436	1.0456	1.0316	1.0321
264.53	0.1091	0.4188	1.0898	1.0943	1.0335	1.0335	1.0331	1.0336
264.53	0.3558	0.1342	1.0603	1.0582	1.0562	1.0615	1.0339	1.0343
338.01	0.1105	0.0273	1.1366	1.1424	1.0971	1.1074	1.0045	1.0029
338.01	0.0580	0.1216	1.1379	1.1447	1.0872	1.0929	1.0052	1.0039
338.01	0.0708	0.0743	1.1407	1.1484	1.0937	1.1022	1.0043	1.0027
338.01	0.0669	0.0076	1.1361	1.1629	1.0948	1.1202	1.0043	1.0009
338.01	0.0532	0.0226	1.1362	1.1652	1.0927	1.1187	1.0043	1.0008
338.01	0.0431	0.0347	1.1369	1.1668	1.0916	1.1174	1.0043	1.0008
338.01	0.0349	0.0467	1.1389	1.1676	1.0916	1.1157	1.0043	1.0008
338.01	0.0210	0.0604	1.1381	1.1705	1.0889	1.1148	1.0044	1.0008
338.01	0.0075	0.0767	1.1393	1.1727	1.0876	1.1131	1.0045	1.0009
338.01	0.0411	0.0377	1.1375	1.1670	1.0916	1.1169	1.0043	1.0008
338.01	0.0653	0.0757	1.1418	1.1502	1.0940	1.1028	1.0042	1.0025
338.01								

APPENDIX III

Computer Programs

A-III - 1 Evaluation of Ω_a, Ω_b and Ω_c

```
  C   EVALUATION OF THE ADJUSTABLE PARAMETER OF THR CLAUSIUS EQUATION
  C  OF STATE DATA USING THE MAXWELL CONDITION.
  C*************************************************************************
  C*****
  C*****
  C*************SF795154  I.M.ELSHAYAL*************************************
  C*****
  C*****
  C*************************************************************************
  C
  C
  C      I.M. ELSHAYAL
  C
  C      CHEM. ENG. DEPT.
  C
  C
  C      SUBROUTINE FUG IS REQUIRED.
         IMPLICIT REAL*8(A-H,O-Z)
         DIMENSION TITLE(20),X(8)
         DIMENSION TT(50)
       1 READ(1,300)(TITLE(I),I=1,15)
     300 FORMAT(20A4)
         WRITE(3,400)(TITLE(I),I=1,15)
   C     WRITE(2,402) (TITLE(I),I=1,15)
     400 FORMAT(1H1,10X, ' EVALUATION OF ADJUSTABLE PARAMETERS OF CLAUSIUS
        2 EQUATION FOR',15A4///)
     402 FORMAT(            ' EVALUATION OF ADJUSTABLE PARAMETERS OF CLAUSIUS
        2 EQUATION FOR',15A4///)
          EL = 1.0D0
          LL = 1
   C      WC = 0.125D0
          NOBS=3
      10 READ(1,1111) PC,VC,TC,W,R,TOL
   C  10 READ(1,100)PC,VC,TC,W,R,TOL
         IF(PC.LE.0.0D0) GO TO 800
     100 FORMAT(8F10.4)
         SDTHI=0.0D0
         SDZV=0.0D0
      20 READ(1,1111) P,V,T,VL
    1111 FORMAT(6F10.0)
   C  20 READ(1,100)P,V,T,VL
         PR=P/PC
         TR=T/TC
```

A-III - 1 (cont'd)

```
G LEVEL  19                        MAIN                  DATE = 71169          12/31/
        BATA = (DLOG10(PR)-DLOG10(TR)*8.0D0/3.0D0-0.1832D0*(PR/TR/TR-
       2 1.0D0))/((1.0D0-TR)/TR+DLOG10(TR)*1.8D0)
C       READ(1,100)P,V,T,VL
        READ(1,1111) P,V,T,VL
        TR=T/TC
        ALFA = (VC/VL-1.0D0-(1.0D0-TR)**(1./3.))/((1.0D0-TR)+(1.0D0-TR)
       2**(1./3.))
        READ(1,100) (TT(I),I=1,NOBS)
        READ(1,1111) WC,DELWC
        WRITE(3,100)ALFA,BATA
  999   CONTINUE
        DO 80 I=1,NOBS
        T=TT(I)
        RT=R*T
        TR=T/TC
        CALL PFK(BATA,TR,PR)
        P=PR*PC
        VL = VC/(1.0D0+ALFA*(1.0D0-TR)+(1.0D0+ALFA)*(1.0D0-TR)**(1./3.))
C       V=R*T/P
        ZL=P*VL/R/T
        ZV=P*V/R/T
        SSS =(R*TC)/(PC*VC)
        WA = 0.4220D0
        WB = 0.25000
        EL2 = EL + 2.0D0
        A2 = (WA*SSS**2*VC**2*TC**EL*PC)/
       2 (R**2*T**EL2)
C       A2 = (WA*SSS**2*VC**2*TC*PC) /(R**2*T**3)
        B = ((WB*VC)/(R*T))*(4.0D0 - SSS)
        CCC = (WC*VC*(3.0D0*SSS - 8.0D0)) / (R*T)
C       CCC = VC*((3.0D0*SSS/8.0D0) - 1.0D0) /(R*T)
C       WRITE(3,1001) ZL,ZV,SSS,A2,B,CCC
 1001   FORMAT('ZL',F10.5,'ZV',F10.5,4E15.8)
        H=B*P/ZL
        E=B*P/ZV
        HHL = CCC*P/ZL
        EEV = CCC * P/ZV
C       WRITE(3,1002)H,E,HHL,EEV
 1002   FORMAT(1H ,'H',4E15.8)
        CALL FUG(W,PR,TR,THI)
        THI = DEXP(2.30259D0*THI)
C       WRITE(3,100)T,P,VL,V,THI
        ZLL=ZL
```

A-III - 1 (cont'd)

```
G LEVEL  19                    MAIN                 DATE = 71169             12/31
          ZVV=ZV
          THIL=THI
          THIV=THI
          HOLD=H
          IJK=0
       39 IJK=IJK+1
          IF(IJK.GE.30) GO TO 49
          J=0
       40 CONTINUE
          IH=0
       41 IF(H.LT.1.0D0) GO TO 42
          H = H - 0.05D0
          IH=IH+1
          IF(IH.LT.20) GO TO 41
       43 H = 0.98D0
       42 J=J+1
          FFH = DLOG(THIV*ZL) + DLOG(1.0D0 - H) -
         1 H/(1.0D0-H) + (HHL + 2.0D0) *
         2  ((1.0D0/(1.0D0-H)) - ZL)
          DFFH = (HHL+1.0D0)/(1.0D0-H)**2 -(1.0D0/(1.0D0-H))
          TEST=FFH/DFFH
C         WRITE(3,1003) FFH,DFFH,TEST
     1003 FORMAT(1H ,'FFH',3E15.8)
          H=H-TEST
          IF(J.GT.40)GO TO 45
          IF(DABS(TEST/H).GT.TOL) GO TO 40
       45 B=H*ZL/P
          A2 = B*((1.0D0/(1.0D0-H)) - ZL)*(1.0D0+HHL)**2/H
C         WRITE(3,1004) B,A2
     1004 FORMAT(1H ,'B',E15.8,10X,'A2',E15.8)
          IJ=0
       47 IF(E.GT.0.0D0) GO TO 46
          E = E + 0.01D0
          IH=IH+1
          IF(IH.LT.20)GO TO 47
          E = 0.02D0
       46 IJ=IJ+1
          IH=0
          FE = -B*P/E+1./(1.-E)-(A2/B)*E/(1.+EEV)**2
          DFE = B*P/E**2 + 1./(1.-E)**2 - (A2/B)/(1.+EEV)**2
          TEST=FE/DFE
C         WRITE(3,1005) FE,DFE,TEST
     1005 FORMAT(1H ,5X,'FE',3E15.8)
```

A-III - 1 (cont'd)

```
G LEVEL  19                     MAIN                    DATE = 71169              12/3

        E=E-TEST
        IF(IJ.GT.40)GO TO 48
        IF(DABS(TEST/E).GT.TOL) GO TO 47
     48 ZVV=B*P/E
        THIV = E/(1.0D0-E) - DLOG(ZVV) - DLOG(1.0D0-E) -
       2 (A2/B)*((2.0D0+EEV)/((1.0D0+EEV)**2)) *E
  C     WRITE(3,1006)THIV
   1006 FORMAT(1H , 30X,'THIV',E15.8)
        THIV = DEXP(THIV)
        WRITE(3,501) J,IJ,IJK
    501 FORMAT(3I5)
        IF(DABS(H-HOLD)/H.LE.TOL*10.0D0) GO TO 49
  C     WRITE(3,200)P,T,ZV,ZVV,VL,VLL,THIV,THIL,WA,WB,THI,H,E
        HOLD=H
        GO TO 39
     49 ZLL=B*P/H
        VLL=R*T*ZLL/P
        THIL = H/(1.0D0-H) - DLOG(ZLL) - DLOG(1.0D0-H) -
       2 (A2/B)*H*((2.0D0+HHL)/((1.0D0+HHL)**2))
   1007 FORMAT(1H , 50X,'THIL',E15.8)
        THIL = 1.0D0/DEXP(-THIL)
        WB = (B*R*T/VC)*(1.0D0/(4.0D0-SSS))
        WA = (A2*R**2*T**EL2) /(SSS**2*VC**2*TC**EL*PC)
  C     WA = A2*R**2*T**3 / (SSS*VC)**2*TC*PC
  C     WRITE(3,1008) WB,WA
   1008 FORMAT(1H , 20X,'WB',E15.8,5X,'WA',F15.8)
    500 FORMAT(I5,3F10.4,5F10.6)
        DTHI=THI-THIV
        DZV=ZV-ZVV
        SDTHI = SDTHI + DABS(DTHI)
        SDZV = SDZV + DABS(DZV)
  C  51 WRITE(3,200) P,T,ZV,WA,WB
     51 WRITE(3,200)P,T,ZV,ZVV,VL,VLL,THIV,THIL,WA,WB,THI,H,E
  C 200 FORMAT(1H ,5X,5(D15.8,5X))
    200 FORMAT(1H ,D11.4,F8.2,6F9.5,2F10.6,2F8.4,D11.4)
  C     WRITE(2,1000) EL
  C     WRITE(2,1000) T,WA,WB
   1000 FORMAT(3D15.8)
     80 CONTINUE
        ADTHI=SDTHI/NOBS
        ADZV=SDZV/NOBS
        WRITE(3,311) ADTHI,ADZV
  C     WRITE(3,3111) EL
```

A- III - 1 (cont'd)

```
G LEVEL  19                     MAIN                    DATE = 71169           12/31

          WRITE(3,3111) WC
   C      TOLZV = 0.005D0
          TOLZV = 0.00000D0
   C      TOLZV = 0.0005D0
   C      TOLZV = 0.001D0
          IF(ADZV.LE.TOLZV) GO TO 9999
   C      WC = WC - 0.001
          WC = WC - DELWC
   C      IF(WC.LT.0.0D0) GO TO 9999
          IF(LL.EQ.20) GO TO 9999
          LL = LL + 1
   C      EL = EL - 0.250D0
   C      IF(EL.EQ.0.0D0) GO TO 9999
          SDTHI=0.0D0
          SDZV=0.0D0
          GO TO 999
    9999  GO TO 1
    3111  FORMAT(///25X,' WC = ',5X,D14.8)
   C3111  FORMAT(///25X,'TEMP. EXPN=',2X,F10.5)
     311  FORMAT(//20X,'AV. DEV. IN FUGACITY = 'E14.8
         1  ,'AV. DEV. IN ZV = ' E14.8/)
     800  STOP
          END
```

A-III - 1 (cont'd)

```
G LEVEL  19                    FUG                  DATE = 71169           12/31.
        SUBROUTINE FUG(W,PR,TR,THI)
C       GENERALIZED CORRELATION OF FUGACITY COEFFICIENT OF PURE
C       COMPONENTS AT SATURATION.
        IMPLICIT REAL*8(A-H,O-Z)
     10 IF(TR.GT.0.56D0) GO TO 20
        THI=(PR/2.303)*((.1445-(.330+(.1385+(.0121)/TR)/TR)/TR)/TR
       1+W*(.073+(.460-(.5+(.097+.0073/TR**5)/TR)/TR)/TR)/TR)
        RETURN
     20 IF(TP.GT.0.70D0) GO TO 30
        THI=(-.53746+(.58798-(.18226-.009499/TR)/TR)/TR)
       1+W*(.11821-(.006542+(.045992-.0116504/TR)/TR)/TR)
        RETURN
     30 IF(TR.GT.0.84D0) GO TO 40
        THI=(-.65625+(.47890+(.13943-.12167/TR)/TR)/TR)
       1+W*(-.47986+(.53604+(.08951-.14448/TR)/TR)/TR)
        RETURN
     40 THI=(-0.87471+(0.73459-(0.11130-(0.27772-0.20204/TR)/TR)/TR)/TR)
       1+W*(-1.15987+(1.25872-(.13670-(.35086-.34700/TR)/TR)/TR)/TR)
        RETURN
        END
```

A-III - 1 (cont'd)

```
G LEVEL  19                    PFK                     DATE = 71169            12/31
          SUBROUTINE PFK(BATA,TR,PR)
          IMPLICIT REAL*8(A-H,O-Z)
          ITER=0
          TOL=0.00001D0
          PRC=-0.1832D0
       30 PR=BATA*(1.-TR)/TR+(1.8*BATA+8./3.)*DLOG10(TR)+PRC
          PRC=0.1832D0*(DEXP(PR*2.30259D0)/TR/TR-1.0D0)
          ITER=ITER+1
          IF(ITER.GT.20) GO TO 32
          IF(ITER.EQ.1) GO TO 31
          IF(DABS((PR-PROLD)/PR).LT.TOL) GO TO 32
       31 PROLD=PR
          GO TO 30
       32 PR = DEXP(PR*2.30259D0)
          RETURN
          END
```

```
C
C      PREDICT P-X-Y CURVE
C  MODIFIED CLAUSIUS EQUATION OF STATE IS USED
C       THREE PARAMETERS ARE FUNCTION OF TEMP.
C      FOR MULTICOMPONENT  SYSTEMS CALCULATION
C      FROM THE PURE COMPONENT PROPERTIES
C
C
C
C
C
C      I.M,  ELSHAYAL
C
C
C      CHEM, ENG, DFPT,
C
C
       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),C3RKV(5),
      A C1RKL(5),C2RKL(5),C3RKL(5),
      2 AMOLWT(5),COMPA(5),COMPB(5),CORRV(5,5),CORRL(5,5),GAMALN(5),
      3 A(4),Z(3),TITLE(20),D(5),PCO(5),VCO(5),TCO(5),PHIV(5),FREF(5),
      4 PS(5),PP(80),XX(80,5),YY(80,5),YCAL(5),PPVOL(5),DC1DT(5),DC2DT(5),
      5,YD(5),RATIO(5),VVL(80)
       COMMON  /FIRST/ TC,PC,VCO,W,AMOLWT,T,P,R,NCOMP,N0NTUM
      1   /SECOND/ Z,A,MTYPE
  800 READ(1,5)(TITLE(I),I=1,19)
    6 FORMAT(1H1,19A4)
    5 FORMAT(     19A4)
       WRITE(3,6)(TITLE(I),I=1,19)
       READ(1,500)NCOMP,N0NTUM,IJK,R
       IF(NCOMP.LE.0) GO TO 900
       WRITE(3,505)
       DO 210 I=1,NCOMP
       READ(1,510)PCO(I),VCO(I),TCO(I),W(I),C1RKL(I),C2RKL(I),C3RKL(I),
      1                                 COMPA(I),COMPB(I)
       C1RKV(I)=C1RKL(I)
       C2RKV(I)=C2RKL(I)
       C3RKV(I) = C3RKL(I)
  210 WRITE(3,515)PCO(I),VCO(I),TCO(I),W(I),C1RKL(I),C2RKL(I),C3RKL(I),
      1                                 COMPA(I),COMPB(I)
       CORRL(NCOMP,NCOMP)=0,0
```

A-III - 2 (cont'd)

```
 LEVEL  20                    MAIN                 DATE = 72202          10/02/39
      NCOMP1=NCOMP-1
      WRITE(3,519)
  519 FORMAT(/10X,12HINPUT DATA ://4X,1HP,8X,2HX1,8X,2HX2,9X,1HT,7X,3HRA
     11,7X,3HRB1,7X,3HRA2,7X,3HRB2//)
      DO 9 I=1,NCOMP1
      I1=I+1
      DO 9 J=I1,NCOMP
    9 READ(1,520)CORRL(I,J)
      READ(1,520) TTT,PSI
      I=0
   10 I=I+1
      SX=0.0
      SY=0.0
C INPUT FOR MULTI COMPONENT SYSTEMS
C
C
C     READ(1,666)    PP(I),(XX(I,J),J=1,NCOMP),(YY(I,J),J=1,NCOMP)
C
C
C INPUT FOR BINARY SYSTEMS
C
C
      READ( 1,666) PP(I),(XX(I,J),J=1,NCOMP1),(YY(I,J),J=1,NCOMP1)
C
C
C     READ( 1,520) PP(I),(XX(I,J),J=1,NCOMP1),(YY(I,J),J=1,NCOMP1)
  666 FORMAT(8F10.0)
C    1 ,VVL(I)
      DO 15 J=1,NCOMP1
      SY=SY+YY(I,J)
   15 SX=SX+ XX(I,J)
      XX(I,NCOMP)=1.0-SX
      YY(I,NCOMP)=1.0-SY
      IF(R.LT.11.0) GO TO 16
      PP(I) = PP(I) /14.696
C     PP(I) = PP(I) * (735.56/760.0)
C     PP(I)=PP(I)/760.
C     PSI=PSI/14.697
C     PSI=PSI/760.
   16 WRITE(3,520) PP(I),(XX(I,J),J=1,NCOMP ),(YY(I,J),J=1,NCOMP )
C    1 ,VVL(I)
      IF(PP(I).GE.0) GO TO 10
   20 NOBS=I-1
```

A-III - 2 (cont'd)

```
EVEL  20                    MAIN                    DATE = 72202             10/02/39

      READ(1,525)IJK,KIJ,CINI,CFIN
  525 FORMAT(2I5,2F10.4)
   30 KIJ=KIJ+1
      IF(KIJ.GE.2) GO TO 800
      WRITE(3,525)IJK,KIJ,CINI,CFIN
      K=0
      KK=20
      KK=0
  280 CONTINUE
      WRITE(3,6)(TITLE(I),I=1,19)
      KK=KK+1
      WRITE(3,516)
      DO 220 I=1,NCOMP1
      CORRL(I,I)=0.0
      I1=I+1
      DO 220 J=I1,NCOMP
      IF(KK.GE.20) GO TO 214
      CORRL(I,J)=CINI+(KK-1)*(CFIN-CINI)/10.
  214 WRITE(3,530)I,J,CORRL(I,J)
      CORRL(J,I)=CORRL(I,J)
  220 CONTINUE
      K=0
      SDP=0.0
      SDV=0.0
      SSYD=0.0
      IF(NONTUM.GE.1) GO TO 223
      DO 221 I=1,NCOMP
      PC(I)=PCO(I)
      VC(I)=VCO(I)
  221 TC(I)=TCO(I)
  120 T=TTT
      RT=R*T
      WRITE(3,535)
  535 FORMAT(/10X,9HCOMPONENT,1X,1HX,7X,1HY,7X,2HYY,7X,2HYD,4X,4HV(P),6X
     1 ,5HV(PO),5X,4HPHIL,6X,4HPHIV,6X,4HSUMY,6X,4HITER,4X,3HNIY/)
  223 CONTINUE
      NIY=0
      ITER=0
      P=PSI
      K=K+1
      DO 40 J=1,NCOMP
      PHIV(J)=1.0
C     PHIV(J)=0.85
```

A-III - 2 (cont'd)

```
.VEL  20                    MAIN                    DATE = 72202

 40 X(J)=XX(K,J)
 42 CALL SUPT(VL,PHI,PPVOL,C1RKL,C2RKL,C3RKL,CORRL,X,1,KIJ)
 41 SUMY=0.0
    SUMP=0.0
    DO 50 J=1,NCOMP
    Y(J)=PHI(J)*X(J)/PHIV(J)
    SUMY=SUMY+Y(J)
 50 SUMP=SUMP+P*Y(J)
    DO 52 J=1,NCOMP
    Y(J)=Y(J)/SUMY
    IF(X(J).EQ.0.0) GO TO 51
    RATIO(J)=Y(J)/X(J)
    GO TO 52
 51 RATIO(J)=0.0
 52 CONTINUE
    DY=SUMY-1.0
    POLD=P
    P=SUMP
    ITER=ITER+1
    IF(DABS((POLD-P)/P)-1.0D-4) 90,90,56
 56 IF(ITER.GT.150)GO TO 105
    IF(ITER.LE.10) GO TO 80
    IF(DABS(DY).GT.DABS(DYOLD)) GO TO 105
 80 DYOLD=DY
    CALL SUPT(VL,PHIV,PVOL,C1RKL,C2RKL,C3RKL,CORRL,Y,0,KIJ)
    GO TO 42
 90 IF(DABS(SUMY-1.0)-1.0D-4) 110,110,100
100 NIY=NIY+1
    IF(NIY-1) 91,91,92
 91 P=P+0.01
    GO TO 93
 92 SLOPE=(P-PO)/(SUMY-SYO)
    DELP=SLOPE*DDY
    IF(DABS(DELP)-1.0) 79,79,78
 78 DELP=DSIGN(1.0D0,DELP)
 79 P=P-DELP
 93 DO 94 J=1,NCOMP
 94 Y(J)=Y(J)/SUMY
    PO=P
    SYO=SUMY
    DDY=SUMY-1.0
    IF(NIY.LE.30) GO TO 95
    IF(DABS(DDY).LE.0.005) GO TO 110
```

A-III - 2 (cont'd)

```
LEVEL  20                    MAIN                    DATE = 72202              10/02/39

      GO TO 96
   95 CALL SUPT(VL,PHIV,PVOL,C1RKL,C2RKL,C3RKL,CORRL,Y,0,KIJ)
      GO TO 42
  105 IF(DABS(DY).LE.0.005) GO TO 110
   96 WRITE(3,601)
  110 DP=P-PP(K)
      SDP = SDP + DABS(DP)
C     P=P*14.697
C     P=P*760.
C     PP(K)=PP(K)*14.697
C     PP(K)=PP(K)*760.
      WRITE(3,600) T,P,PP(K),DP
C     PP(K)=PP(K)/760.
C     PP(K)=PP(K)/14.697
  600 FORMAT(3X,3HT= ,F8.2,5H  P= ,F10.4,7HPP(K)= ,F10.4,4HDP= ,F10.4)
      SYD=0.0
      DO 111 J=1,NCOMP
      YD(J)=Y(J)-YY(K,J)
      SYD = SYD + DABS(YD(J))
      SSYD=SSYD+SYD
C     WRITE(2,1111) P,X(J),Y(J)
 1111 FORMAT(3F10.5)
  111 WRITE(3,610)COMPA(J),COMPB(J),X(J),Y(J),YY(K,J),YD(J),PPVOL(J),
     1PVOL(J),PHI(J),PHIV(J),SUMY,ITER,NIY
      DV=VL-VVL(K)
      SDV = SDV + DABS(DV)
      WRITE(3,901)VL,VVL(K),DV
  901 FORMAT(15X,4HVL= ,F10.4,5HVVL= ,F10.4,4HDV= ,F10.4)
  610 FORMAT(10X,2A4,4D12.4,4F10.4,F8.4,2I5)
  601 FORMAT(1H0,87HFAILED TO CONVERGE, RESULTS BELOW ARE NOT RELIABLE I
     1F SUMY IS MUCH DIFFERENT FROM UNITY)
C     PSI=P
      IF(K-NOBS) 223, 850,850
  850 CONTINUE
      WRITE(3,620)SDP,SSYD,SDV
  620 FORMAT(/10X,'ACCUM. SUM OF DEV. IN P IS',D12.4,'ACCUM. SUM OF DEV.
     1IN Y IS', D12.4,'SDV IS',D12.4/)
      IF(KK-11)280,30,30
  900 STOP
  500 FORMAT (3I5,F10.4)
  505 FORMAT(1H0,4X,2HPC,10X,2HVC,6X,2HTC,10X,1HW,7X,5HC1RKL,5X,5HC2RKL,
     15X,5HMOLWT,4X,9HCOMPONENT//)
  510 FORMAT(7F10.0,2X,2A4)
```

A-III - 2 (cont'd)

```
G LEVEL  20                    MAIN                    DATE = 72202

  C 510 FORMAT(4F8.4,2F8.4,2A4)
    515 FORMAT(1H ,7D12.5,3X,2A4)
  C 515 FORMAT(1H ,6E12.5,3X,2A4)                                 / )
    516 FORMAT(1H , /1H ,16X,3HKIJ
    520 FORMAT(14F10.4)
    530 FORMAT(1H ,6X,2I2,F10.4,2F10.4)
        END
```

A-III - 3

```
LEVEL  19                  MAIN                 DATE = 71176          11/01/01

C
C EVALUATION OF ACTIVITY COEFFIECIENTS FROM LIQUID  PROPERTIES OF M/XITURES
C  MODIFIED CLUSIUS EQUATION OF STATE IS USED
C      THREE PARAMETERS ARE FUNCTION OF TEMP.
C
C      I.M.  ELSHAYAL
C
C
C      CHEM. ENG. DEPT.
C
C
       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),C3RKV(5),
      A C1RKL(5),C2RKL(5),C3RKL(5),
      2 AMOLWT(5),COMPA(5),COMPB(5),CORRV(5,5),CORRL(5,5),GAMALN(5),
      3 A(4),Z(3),TITLE(20),D(5),PCO(5),VCO(5),TCO(5),PHIV(5),FREF(5),
      4 PS(5),PP(80),XX(80,5),YY(80,5),YCAL(5),PPVOL(5),DC1DT(5),DC2DT(5),
       COMMON  /FIRST/ TC,PC,VCO,W,AMOLWT,T,P,R,NCOMP,NONTUM
      1   /SECOND/ Z,A,MTYPE
  800 READ(1,5)(TITLE(I),I=1,19)
    5 FORMAT(     19A4)
    6 FORMAT(1H1,19A4)
      WRITE(3,6)(TITLE(I),I=1,19)
      READ(1,500)NCOMP,NONTUM,IJK,R
      IF(NCOMP.LE.0) GO TO 900
      WRITE(3,505)
      DO 210 I=1,NCOMP
      READ(1,4567)
      A              PCO(I),VCO(I),TCO(I),W(I),C1RKL(I),C2RKL(I),C3RKL(I),
      1                                  COMPA(I),COMPB(I)
 4567 FORMAT(7F10.0,2A4)
      C1RKV(I)=C1RKL(I)
      C2RKV(I)=C2RKL(I)
  210 WRITE(3,515)PCO(I),VCO(I),TCO(I),W(I),C1RKL(I),C2RKL(I),C3RKL(I),
      1                                  COMPA(I),COMPB(I)
      CORRL(NCOMP,NCOMP)=0.0
      NCOMP1=NCOMP-1
```

A-III - 3 (cont'd)

```
       WRITE(3,519)
   519 FCRMAT(/10X,12HINPUT DATA ://4X,1HP,8X,2HX1,8X,2HX2,9X,1HT,7X,3H
     11,7X,3HRB1,7X,3HRA2,7X,3HRB2//)
       CC 9 I=1,NCCMP1
       I1=I+1
       CC 9 J=I1,NCCMP
     9 READ(1,8910) CORRL(I,J)
  8910 FCRMAT(8F10.C)
       I=0
    10 I=I+1
       SX=0.0
       SY=0.0
C INPLT FOR MULTI CCMPCNENT SYSTEMS
C
C
C      READ(1,666)    PP(I),(XX(I,J),J=1,NCCMP),(YY(I,J),J=1,NCOMP)
C
C
C INPLT FOR BINARY SYSTEMS
C
C
       READ( 1,666) PP(I),(XX(I,J),J=1,NCOMP1),(YY(I,J),J=1,NCOMP1)
C
C
C      READ( 1,520) PP(I),(XX(I,J),J=1,NCOMP1),(YY(I,J),J=1,NCOMP1)
   666 FCRMAT(8F10.0)
C    1 ,VVL(I)
       CC 15 J=1,NCCMP1
       SY=SY+YY(I,J)
    15 SX=SX+ XX(I,J)
       XX(I,NCCMP)=1.0-SX
       YY(I,NCCMP)=1.0-SY
       IF(R.LT.11.0) GO TO 16
C      PP(I) = PP(I) * (735.56/760.)
       PP(I) = PP(I) / 14.6970
C      PP(I)=PP(I)/760.
C      PSI=PSI/14.697
C      PSI=PSI/760.
    16 WRITE(3,52C) PP(I),(XX(I,J),J=1,NCOMP ),(YY(I,J),J=1,NCOMP )
C    1 ,VVL(I)
       IF(PP(I).GE.C) GC TO 10
    20 NCBS=I-1
       READ(1,8910) TTT,PSO,(PS(I),I=1,NCOMP)
```

A-III - 3 (cont'd)

```
LEVEL  19                    MAIN                DATE = 71176          11/01/
      PSC = PSO / 14.697
      DO 5555 I=1,NCOMP
 5555 PS(I) = PS(I) / 14.697
      IF(R.LT.11.0) GO TO 21
   21 WRITE(3,520)TTT,PSO,(PS(I),I=1,NCOMP)
      READ(1,525)IJK,KIJ,CINI,CFIN
  525 FORMAT(2I5,2F10.4)
   30 KIJ=KIJ+1
      IF(KIJ.GE.2) GO TO 800
      WRITE(3,525)IJK,KIJ,CINI,CFIN
      K=0
      KK=20
      KK=0
  280 CONTINUE
      WRITE(3,6)(TITLE(I),I=1,19)
      KK=KK+1
      WRITE(3,516)
      DO 220 I=1,NCOMP1
      CORRL(I,I)=0.0
      I1=I+1
      DO 220 J=I1,NCOMP
      IF(KK.GE.20) GO TO 214
      CORRL(I,J)=CINI+(KK-1)*(CFIN-CINI)/10.
  214 WRITE(3,530)I,J,CORRL(I,J)
      CORRL(J,I)=CORRL(I,J)
  220 CONTINUE
      K=0
      SSDY = 0.0
  219 CONTINUE
      IF(NONTUM.GE.1) GO TO 223
      DO 221 I=1,NCOMP
      PC(I)=PCC(I)
      VC(I)=VCC(I)
  221 TC(I)=TCC(I)
  120 T=TTT
      RT=R*T
      DO 226 I=1,NCOMP
      DO 224 J=1,NCOMP
      X(J)=0.0
  224 CONTINUE
      X(I)=1.0
      P=PSO
      CALL SLPT(VL,PHI,PVOL,C1RKL,C2RKL,C3RKL,CORRL,X,1,KIJ)
```

A-III - 3 (cont'd)

```
       FREFER(I)=PHI(I)*X(I)*PSO
   226 D(I)=PVOL(I)
       WRITE(3,521)T,PSO,(FREFER(I),I=1,NCOMP),(D(I),I=1,NCOMP)
   521 FORMAT(//5X,2F10.2,11F 9.4//)
       WRITE(3,535)
   535 FORMAT(/26X,9HCOMPONENT,1X,1HX,7X,4HYCAL,4X,4HYOBS,4X,
      1  6HGAMALN,2X,5HGAMMA,5X,4HV(P),6X,5HV(PO),5X,4HPHIL,6X,
      1  4HPHIV,6X,5HPHILL/)
   223 CONTINUE
       K=K+1
       DO 234 J=1,NCOMP
       Y(J)=YY(K,J)
   234 X(J)=XX(K,J)
       III=0
   230 III=III+1
       IF(III.GT.1)GO TO 236
       P=PSO
       CALL SLPT(VL,PHIRF,PVOL,C1RKL,C2RKL,C3RKL,CORRL,X,1,KIL)
       DO 233 J=1,NCOMP
       PPVOL(J)=PVOL(J)
   233 CONTINUE
       GO TO 230
   236 P=PP(K)
       CALL SLPT(VL,PHI,PVOL,C1RKL,C2RKL,C3RKL,CORRL,X,1,KIJ)
       DO 235 J=1,NCOMP
       GAMALN(J)=ALOG(PHI(J)*P/FREFER(J)        )
      1   +(PPVOL(J)+PVOL(J))*(PSO-P)/2.0/RT
       PHIV(J)=1.0
   235 GAMMA(J)= EXP(GAMALN(J) )
       ITER=0
       RATIO=PHIV(1)*Y(1)/PHI(1)/X(1)
   240 ITER=ITER+1
       SDY=0.0
       IF(ITER-20)242,242,260
   242 DO 245 J=1,NCOMP
       YCAL(J)=PHI(J)/PHIV(J)*X(J)
       SDY=SDY+YCAL(J)
   245 CONTINUE
       DO 246 J=1,NCOMP
       IF(ITER.LE.1) GO TO 250
   246 YCAL(J)=YCAL(J)/SDY
       DO 247 J=1,NCOMP
       IF(ABS(YCAL(J)-Y(J)) .GT.0.00001) GO TO 250
```

A-III - 3 (cont'd)

```
LEVEL  19                    MAIN                    DATE = 71176              11/01
  247 CONTINUE
      GO TO 260
  250 DO 255 J=1,NCOMP
  255 Y(J)=YCAL(J)
      CALL SUPT(VL,PHIV,PVOL,C1RKL,C2RKL,C3RKL,CORRL,Y,0,KIJ)
      GO TO 240
  260 SDY=0.0
      DO 265 J=1,NCOMP
  265 SDY    =SDY    +ABS(YCAL(J)-YY(K,J))
      SSDY=SSDY+SDY
      IF(R.LT.11.) GO TO 266
C 266 WRITE(2,540)P,(X(J),J=1,NCOMP1),(YY(K,J),J=1,NCOMP1)
C 266 WRITE(2,540)P,(X(J),J=1,NCOMP1),(YCAL(J),J=1,NCOMP1)
C    1   ,(GAMALN(J),J=1,NCOMP)
  266 CONTINUE
  540 FORMAT(8F10.4)
      WRITE(3,600)T,P ,RATIO
      DO 300 J=1,NCOMP
  300 WRITE(3,610)COMPA(J),COMPB(J),X(J),YCAL(J),YY(K,J),GAMALN(J),
     1   GAMMA(J),PVOL(J),PPVOL(J),PHI(J),PHIV(J),F(J),ITER
  600 FORMAT(3X,3HT= ,F8.2,5H  P= ,F10.4,8H  RATIO=,F8.4)
  610 FORMAT(27X,2A4,5F8.4,5F10.4,I5)
  270 IF(K-NOBS)223,850,850
  850 CONTINUE
      WRITE(3,620)SSDY
  620 FORMAT (/ 10X,'ACCUM. SUM OF DEV. IN Y IS 'E12.4/)
      IF(KK-10)280,30,30
  900 STOP
  500 FORMAT (3I5,F10.4)
  505 FORMAT(1H0,4X,2HPC,10X,2HVC,6X,2HTC,10X,1HW,7X,5HC1RKL,5X,5HC2RK
     15X,5HMOLWT,4X,9HCOMPONENT//)
  510 FORMAT(4F8.4,      5F8.4,2A4)
  515 FORMAT(1H ,4F10.3,3F10.4,3X,2A4)
  516 FORMAT(1H , /1H ,16X,3HKIJ                                 / )
  520 FORMAT(F10.2,12F10.4)
  530 FORMAT(1H ,6X,2I2,F10.3,2F10.4)
  562 FORMAT(45X,2A4,3F10.4)
      END
```

A- III - 4

```
C   COMPARISON OF ZV,ZL EXPT. VS. ZV,ZL CALC. FROM MODIFIED CLAUSIUS
C     EQUATION OF STATE
C
C      I.M.ELSHAYAL
C
C      CHEM ENG. DEPT.
C
C
C      M = 1 FOR VAPOR PHASE
C      M = 2 FOR LIQUID PHASE
C
C
      IMPLICIT REAL*8(A-H,O-Z)
      DIMENSION TITLE(80)
      DIMENSION PP(50),VV(50),TT(50)
      DIMENSION VL(50),VAP(50),DVL(50),DVAP(50),DV(50)
      DIMENSION A(4),Z(3)
    1 READ (1,10) (TITLE(I),I=1,80)
      WRITE(3,20) (TITLE(I),I=1,80)
   10 FORMAT(80A1)
   20 FORMAT(1H1,80A1)
      READ(1,30) M,N
      DO 3 I=1,N
    3 DV(I) = 0.0
      DZ = 0.0
      IF(N.EQ.0) GO TO 400
   30 FORMAT(2I5)
      READ(1,40) PC,TC,VC,W,WA,WB,WC,R
      WRITE(3,41) PC,VC,TC,W,WA,WB,WC,R
   40 FORMAT(8F10.0)
   41 FORMAT(///5X,8(F10.5))
      DO 51 I=1,N
      READ(1,50) PP(I),VV(I),TT(I)
   51 WRITE(3,52) PP(I),VV(I),TT(I)
   50 FORMAT(3F10.0)
   52 FORMAT(///20X,3(F10.5,10X))
      S = (R*TC)/(PC*VC)
      AA =(WA*(S**2)*(VC**2)*PC*TC)
      B  = WB*VC*(4.-S)
      C  = WC*VC* ((3.*S)-8.)
      DO100 I=1, N
      RT = R*TT(I)
      A(1) = 1.0
```

A-III - 4 (cont'd)

```
G LEVEL  20                    MAIN                    DATE = 72207          11/36/01
      A(2) = (2.0*C) - B - (RT/PP(I))
      A(3) = (C**2) - (2.0*B*C) - (2.0*RT*C/PP(I)) +(AA/(PP(I)*TT(I)))
      A(4) = (-B*(C**2))- ((RT*(C**2))/PP(I)) -(AA*B/(PP(I)*TT(I)))
      CALL CUBEQN(INDEX,A,Z)
      WRITE(3,300) Z(1),Z(2),Z(3)
  300 FORMAT(10X,3D15.8)
      IF(M.EQ.1) GO TO 60
      IF(M.EQ.2) GO TO 70
   60 VAP(I) =DMAX1(Z(1),Z(2),Z(3))
      DVAP(I) = VV(I) - VAP(I)
      WRITE(3,80) PP(I),TT(I),VV(I),VAP(I),DVAP(I)
   80 FORMAT(5X,'P= ',F10.5,2X,'T= ',F10.5,2X,'V= ',F10.5,2X,'VCAL= ',
     2 F10.5,2X,'DEV= ',D15.8)
      DV(I) = DV(I) + DABS(DVAP(I))
      ZV = (PP(I)*VV(I))/RT
      ZVCAL = (PP(I)*VAP(I))/RT
      DZV = ZV - ZVCAL
      WRITE(3,90) ZV,ZVCAL,DZV
   90 FORMAT(5X,'Z= ',D15.8,2X,'ZCAL= ',D15.8,2X,'DZ= ',D15.8)
      DZ = DZ + DABS(DZV)
      GO TO 101
   70 VL(I) = DMIN1(Z(1),Z(2),Z(3))
      DVL(I) =VV(I) - VL(I)
      WRITE(3,80) PP(I),TT(I),VV(I),VL(I),DVL(I)
      DV(I)=  DV(I) + DABS(DVL(I))
      ZL = (PP(I)*VV(I))/RT
      ZLCAL = (PP(I)*VL(I))/RT
      DZL = ZL - ZLCAL
      WRITE(3,90) ZL,ZLCAL,DZL
      DZ = DZ + DABS(DZL)
      GO TO 101
  101 CONTINUE
  100 CONTINUE
      DDV=0.0
      DO433 K=1,N
  433 DDV=DDV+DV(K)
      DDV  =DDV/N
      DZ = DZ / N
      WRITE(3,200) DDV  ,DZ
  200 FORMAT(////25X,'AV. DEV. IN MOL. VOLUME = ',2X,D15.8///
     2 25X ,'AV. DEV. IN COMPRESSIBILITY = ',2X,D15.8)
      GO TO 1
  400 RETURN

      END
```

A-III - 4 (cont'd)

```
G LEVEL  20                       MAIN                    DATE = 72207            11/36/
 C    SOLVING EQN F(Z)=A(1)*Z**3+A(2)*Z**2+A(3)*Z+A(4)=0
        SUBROUTINE CUBEQN(INDEX,A,Z)
        IMPLICIT REAL*8(A-H,O-Z)
        DIMENSION A(4),Z(3)
        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)**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 5
        THI=DARCOS(-(Q/2.0D0)/DSQRT(-(P/3.0D0)**3))
        R=2.0D0*DSQRT(-P/3.0D0)*DCOS(THI/3.0D0)
        GO TO 8
     5  S=(-(Q/2.0D0)+DSQRT(DE))
        U=(-Q/2.0D0-DSQRT(DE))
        IF(S) 6,7,6
     6  S=S/DABS(S)*(S*S)**(1.0D0/6.0D0)
     7  IF(U) 9,11,9
     9  U=U/DABS(U)*(U*U)**(1.0D0/6.0D0)
    11  R=S+U
     8  DISC=R*R-4.0D0*(P+R*R)
        IF(DISC)40,10,30
    10  INDEX=0
        Z(1)=R-T
        Z(2)=-R/2.0D0-T
        Z(3)=Z(2)
        RETURN
    30  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
        RETURN
    40  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)
        RETURN
        END
```

A-III - 5 Evaluation of Supporting Properties (Subroutine SUBT)

```
LEVEL  20                      SUPT                   DATE = 72202              10/02/39

      SUBROUTINE SUPT(VL,PHI, PVOL ,C1RKL,C2RKL,C3RKL,CORRL,X,LV,KIJ)
C     EVALUTION OF SUPPORTING PROPERTIES,THE FUGACITY AND THE
C     PARTIAL MOLAL VOLUME OF FLUID MIXTURES.
      IMPLICIT REAL*8(A-H,O-Z)
      DIMENSION TC(5),PC(5),VCO(5),W(5),AMOLWT(5)  ,ARKL(5,5),BRKL(5),
     A CRKL(5),
     1  WIJ(5,5),TCOIJ(5,5),TCIJ(5,5),A(4),Z(3),C1RKL(5),C2RKL(5),
     BC3RKL(5),
     1  PVOL(5),PHI(5),CORRL(5,5),X(5),A2I(5),BI(5),AIRKL(5),
     CCI(5)
      DIMENSION ZZ(3)
      COMMON  /FIRST/ TC,PC,VCO,W,AMOLWT,T,P,R,NCOMP,NQNTUM
     1   /SECOND/ Z,A,MTYPE
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
   91 DO 100 I=1,NCOMP
      ZC = (PC(I)*VCO(I))/(R*TC(I))
      ARKL(I,I) = C1RKL(I)*(VCO(1)**2)*PC(I)*TC(I)/(ZC**2)
      BRKL(I) = C2RKL(I)*      (4.0 - (1.0/ZC))*VCO(I)
      CRKL(I) = C3RKL(I)*      ((3./ZC)-8.)*VCO(I)
      IF(I.EQ.NCOMP) GO TO 100
      I1=I+1
      DO 100 J=I1,NCOMP
      IF(KIJ.GT.1) GO TO 97
      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)
   94 VCOIJ=(VCO(I)**(1./3.)+VCO(J)**(1./3.))**3/8.0
   96 PCOIJ=ZCOIJ* R    *TCIJ( I,J)/VCOIJ
      PCIJ=PCOIJ
   95 ARKL(I,J)=(C1RKL(I)+C1RKL(J))*0.5*
     2(VCOIJ)**2*PCOIJ*TCIJ(I,J)/((ZCOIJ)**2)
      GO TO 98
   97 IF(KIJ.GT.2) GO TO 99
      ARKL(I,J)=(ARKL(I,I)*ARKL(J,J))**0.5*(1.0-CORRL(I,J))
      GO TO 98
   99 ARKL(I,J)=(ARKL(I,I)*CORRL(I,J)+(1.-CORRL(I,J))*ARKL(J,J))
   98 ARKL(J,I)=ARKL(I,J)
```

A-III - 5 (cont'd)

```
.EVEL  20                    SUPT                    DATE = 72202              10/02/39

 100 CONTINUE
     AMRKL=0.0
     BMRKL=0.0
     CMRKL = 0.0
     DO 120 I=1,NCOMP
     A2I(I) = ARKL(I,I)/((R**2)*(T**3))
     BI(I)=BRKL(I)/RT
     CI(I) = CRKL(I)/RT
     BMRKL=BMRKL+X(I)*BRKL(I)
     CMRKL = CMRKL + X(I)*CRKL(I)
     AIRKL(I)=0.0
     DO 120 J=1,NCOMP
     AIRKL(I)=AIRKL(I)+X(J)*ARKL(I,J)
 120 AMRKL=AMRKL+X(I)*X(J)*ARKL(I,J)
     A2 = AMRKL/((R**2)*(T**3))
     B=BMRKL/RT
     C = CMRKL/RT
     A(1)=1.0
     A(2) = (C*P*2.0) - (B*P) - 1.0
     A(3) = ((C**2)*(P**2)) - (2.0*C*B*(P**2)) - (2.0*C*P)
    2 + ((A2/B)*B*P)
     A(4) = (-B*(C**2)*(P**3)) - ((C**2)*(P**2)) - ((A2/B)*(B**2)*(P**2
    2))
     CALL CUBEQN
     IF(MTYPE)115,140,140
 115 IF(LV.EQ.0) GO TO 145
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 150
 145 ZL = DMAX1(Z(1),Z(2),Z(3))
     GO TO 150
 140 ZL=Z(1)
 150 H=B*P/ZL
     HH = C*P/ZL
C    WRITE(3,591) H,HH,P,A2,B,ZL,LV
```

A-III - 5 (cont'd)

```
                                                                  DATE = 72202
LEVEL  20                          SUPT

  591 FORMAT(5X,6D12,4,I2)
      CALL EQNRK(A2,B,P,H,HH,ZL,LV)
      VL=RT*B/H
  123 CONTINUE
      IF(LV,EQ,0) GO TO 135
      QD = T*(VL+CMRKL)**2
      QH = (2.0*AMRKL)/(T*(VL+CMRKL)**3)
      QK = RT/((VL-BMRKL)**2)
  135 DO 130 I=1,NCOMP
C     WRITE(3,591)ZL,H
      PHILN = DLOG(1.0/(1.0-H)) - DLOG(ZL) +
     2 (H/(1.0-H))*(BRKL(I)/BMRKL)-(A2/C)*(HH/(1.0+HH))*
     3((2.0*AIRKL(I)/AMRKL)-((HH/(1,0+HH))*(CRKL(I)/CMRKL)))
      PHI(I)=DEXP(PHILN )
      IF(LV.EQ.0) GO TO 130
      QE1 = 0.0
      DO 125 J=1, NCOMP
  125 QE1 = QE1+ X(J)*ARKL(I,J)
      QE = 2.0* (QE1 - ((AMRKL*CRKL(I))/(VL+CMRKL)))
      QG = (RT/(VL-BMRKL)) * (1,0 + (BRKL(I)/(VL-BMRKL)))
      PVOL(I) = ((QE/QD) - QG) / (QH-QK)
  130 CONTINUE
  300 RETURN
      END
```

```
      SUBROUTINE CUBEQN
C     SOLVES CUBIC EQUATION OF CLUAISUS EQUATION OF STATE FOR COMPRESSEIBILITY
C     FACTORS
      IMPLICIT REAL*8(A-H,O-Z)
      DIMENSION A(4),Z(3) ,B(3)
      COMMON /SECOND/ Z,A,MTYPE
    1 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
      BET=2.0*B1OV3**3  -B(2)*B1OV3+B(3)
      BETOV2=BET/2.0
      ALFOV3=ALF/3.0
      CUAOV3=ALFOV3**3
      SQBOV2=BETOV2**2
      DEL=SQBOV2+CUAOV3
      IF(DEL)40,20,30
   20 MTYPE=0.0
      GAM = DSQRT(-ALFOV3)
      IF(BET)22,22,21
   21 Z(1)=-2.0*GAM-B1OV3
      Z(2)=GAM-B1OV3
      Z(3)=Z(2)
      GO TO 50
   22 Z(1)=2.0*GAM-B1OV3
      Z(2)=-GAM-B1OV3
      Z(3)=Z(2)
      GO TO 50
   30 MTYPE=1
      EPS = DSQRT(DEL)
      TAU=-BETOV2
      RCU=TAU+EPS
      SCU=TAU-EPS
      SIR=1.0
      SIS=1.0
       IF(RCU)31,32,32
   31 SIR=-1.0
   32 IF(SCU)33,34,34
   33 SIS=-1.0
   34 R=SIR*(SIR*RCU)**0.33333333                  99
      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 50
   40 MTYPE=-1
      QUOT=SQBOV2/CUAOV3
      ROOT = DSQRT(-QUOT)
      IF(BET)42,41,41
   41 PEI = (1.5707963 + DATAN(ROOT/DSQRT(1.0-ROOT**2)))/3.0
      GO TO 43
   42 PEI = DATAN(DSQRT(1.0-ROOT**2)/ROOT)/3.0
   43 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
   50 RETURN
      END
```

```
LEVEL  20                          EQNRK                    DATE = 72202
        SUBROUTINE EQNRK(A2,B,P,E,FE,Z,LV)
        IMPLICIT REAL*8(A-H,O-Z)
        TOL=1,0D-6
        IH=0
        IJ=0
    45  IF(LV.LT.1) GO TO 47
        IF(E.LT,1,0) GO TO 46
        E=E-0.02
        IH=IH+1
        IF(IH.LT,20) GO TO 45
        E=0,98
        GO TO 46
    47  IF(E.GT.0,0) GO TO 46
        E=E+0.01
        IH=IH+1
        IF(IH,LT.20)GO TO 47
        E=0.02
    46  IJ=IJ+1
        FE=-B*P /E+1./(1.0-E)-(A2/B)*E/(1.+EE)**2
        DFE=B*P /E**2 +1./(1.-E)**2-(A2/B)/(1.+EE)**2
C       WRITE(3,592)E,FE,DFE,LV
C  592  FORMAT(20X,3E12,4,5X,I2)
C       IF(ABS(DFE),LT.1.0E-20) DFE=DFE/ABS(DFE)*1,0E-20
        TEST=FE/DFE
        E=E-TEST
        IF(IJ.GT.60)GO TO 48
        IF(DABS(TEST/E),GT.TOL) GO TO 45
    48  Z  =B*P/E
        RETURN
        END
```

A-III - 6 Fitting Redlich-Kister Equation for Binary System

```
∇ RKFIT
 [.5] ' WHAT IS THE SYSTEM NAME '
 [.75] NAME←⍞
 [1] 'TYPE IN PHASE EQUILIBRIUM DATA ON THE FOLLOWING ORDER:P,X,Y,GAMA1,G
 [2] PP←⎕
 [2.05] XX←⎕
 [2.1] YY←⎕
 [2.2] GI←⎕
 [2.3] GII←⎕
 [5] F←1.-(2.×XX)
 [6] G←XX×(1.-XX)
 [7] H←(6.×G)-1.
 [8] G←1.-(8.×G)
 [9] E←GI-GII
 [10] A11←+/F×F
 [11] A12←+/F×H
 [12] A13←+/F×F×G
 [13] A14←+/F×E
 [14] A22←+/H×H
 [15] A23←+/F×H×G
 [16] A24←+/H×E
 [17] A33←+/F×F×G×G
 [18] A34←+/F×G×E
 [19] A21←A12
 [20] A31←A13
 [21] A32←A23
 [22] A11,A12,A13,A14,A21,A22,A23,A24,A31,A32,A33,A34
 [23] A←3 4⍴A11,A12,A13,A14,A21,A22,A23,A24,A31,A32,A33,A34
 [24] 3 SOLVE A
 [26] GAMC1←(1.-XX)×(1.-XX)×(B++C×((4.×XX)-1.)+D×((6.×XX)-1.)×((2.×XX)-1.
 [27] GAMC2←XX×XX×(B+C×((4.×XX)-3.)+D×((2.×XX)-1.)×((6.×XX)-5.))
 [28] DGI←GI-GAMC1
 [28.5] DGI←|DGI
 [29] DGII←GII-GAMC2
 [29.5] DGII←|DGII
 [31] K←0
 [31.5] NAME
 [32] LOOP:K←K+1                                     ';GI[K];'      ';GAMC1[K];'
 [33] ' ';PP[K];'   ';XX[K];'      ';YY[K];'
 [34] →(K<⍴PP)/LOOP
 [35] SUMI←(+/DGI)÷⍴PP
 [36] SUMII←(+/DGII)÷⍴PP
 [37] SUMI
 [38] SUMII
∇
```

A - III - 6 (cont'd)

```
∇ NA SOLVE A
 [1] NA1←NA+1
 [2] I←0
 [3] LOOP1:I←I+1
 [4] →(A[I,I]=0)/ST206
 [5] ST200:P←1.÷A[I,I]
 [6] J←I
 [7] LOOP2:J←J+1
 [8] A[I,J]←P×A[I,J]
 [9] →(J<NA1)/LOOP2
 [10] K←0
 [11] LOOP3:K←K+1
 [12] →((I-K)=0)/ST202
 [13] Q←A[K,I]
 [14] J←I
 [15] LOOP4:J←J+1
 [16] A[K,J]←A[K,J]-(Q×A[I,J])
 [17] →(J<NA1)/LOOP4
 [18] ST202:→(K<NA)/LOOP3
 [19] →(I<NA)/LOOP1
 [19.1] I←0
 [19.2] LOOP8:I←I+1
 [19.3] 'SOLUTION OF GIVEN EQUATIONS:      ';A[I,NA1]
 [19.31000306] →(I=1)/BB
 [19.32000613] →(I=2)/CC
 [19.32999387] →(I=3)/DD
 [19.33999694] BB:B←A[I,NA1]
 [19.35] CC:C←A[I,NA1]
 [19.35998775] DD:D←A[I,NA1]
 [19.5] →(I<NA)/LOOP8
 [19.6] INDIC←0
 [20] →(INDIC=0)/END
 [21] ST206:→((NA-1)=0)/ST304
 [22] I1←I+1
 [23] L←I1
 [24] LOOP5:L←L+1
 [25] →(A[L,I]=0)/ST303
 [26] J←I
 [27] LOOP6:J←J+1
 [28] HOLD←A[I,J]
 [29] A[I,J]←A[L,J]
 [30] A[L,J]←HOLD
 [31] →(J<NA1)/LOOP6
 [32] →ST200
 [33] ST303:→(L<NA)/LOOP5
 [34] INDIC←1
 [35] ST304:'DIAGONAL ELEMENT IN EQN.';I;'IS ZERO TRANSPOSE EQN. WITH P
 [36] END:
 ∇
```

A - III - 7 Lagrang Interpolation Method for Volumetric Data

```
∇ LAGRANG
[1]  'TYPE IN NO. OF TABULATED POINTS'
[2]  L←⎕
[3]  'TYPE IN NO. OF GIVEN ARGUMENTS'
[4]  LL←⎕
[5]  'TYPE IN TABULATED VALUES OF X'
[6]  ZX←⎕
[7]  ZX
[8]  'TYPE IN TABULATED VALUES OF Y'
[9]  ZT←⎕
[10] ZT
[11] 'TYPE IN GIVEN ARGUMENTS OF X'
[12] XK←⎕
[13] XK
[14] K←0
[15] LOOP1:K←K+1
[16] →(K>LL)/OUT
[17] C←0.00
[18] I←0
[19] LOOP2:I←I+1
[20] →(I>L)/TEST1
[21] P←1.00
[22] J←0
[23] LOOP3:J←J+1
[24] →(J>L)/TESTJ
[25] M←I-J
[26] →(M=0)/OUT3
[27] A←(XK[K]-ZX[J])÷(ZX[I]-ZX[J])
[28] P←P×A
[29] OUT3:→(J≤L)/LOOP3
[30] TESTJ:J←L
[31] B←P×ZT[I]
[32] C←C+B
[33] →(I≤L)/LOOP2
[34] TEST1:I←L
[35] YY←XK[K]
[36] E←YY-C                                                                        E=
[37] '   XK=    ';XK[K];'    C=          ';C;'         YY=      ';YY;'
[38] →(K≤LL)/LOOP1
[39] →(K>LL)/OUT
[40] OUT:
∇
```