NOTATION

D = Hanning lag window
G = system transfer function
i = $+\sqrt{-1}$
k = integer
N = number of points sampled from continuous record
P_{xy} = even part of cross covariance, (in. of recorder deflection)^2
Q_{xy} = odd part of cross covariance, (in. of recorder deflection)^2
R_{fg} = covariance of functions f and g
R_{xx} = autocovariance of input signal, (in. or recorder deflection)^2
R_{xy} = cross covariance with input leading output, (in. of recorder deflection)^2
R_{yx} = cross covariance with input lagging output, (in. of recorder deflection)^2
S_{xx} = input power spectrum, (in. of recorder deflection)^2/(cycle) (sec.)
S_{yy} = output power spectrum, (in. of recorder deflection)^2/(cycle) (sec.)
S_{xy} = power spectrum of cross covariance, (in. of recorder deflection)^2/(cycle) (sec.)
T_m = maximum lag time, sec.

Greek Letters

γ^2 = coherence between input and output signals
Δt = sample interval, sec.
ν = integer assuming values between zero and maximum number of lags
τ = time lag, $|\nu|\Delta t$, sec.
ω = frequency, rad./sec.

Subscripts

f, g = general functions of time
x = system input signal
y = system output signal

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Activity Coefficients at Infinite Dilution for Ternary System

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A simple thermodynamic expression determining the infinite dilution ternary activity coefficients from the dew point isobar is presented. A flow method measuring the dew point temperature for ternary and binary isobaric systems is proposed. An analysis determining the infinite dilution ternary activity coefficients for ethanol-isopropyl alcohol-water system at 1 atm. is illustrated.

Knowledge of the infinite dilution ternary activity coefficients in the liquid phase is important for the design of distillation equipment, especially for the proper choice of solvent in distillation operation.

There have been several studies for experimentally determining the activity coefficients at infinite dilution. For binary systems the useful procedure, which is based on readily obtained data such as total pressure-concentration curve ($P - x$), bubble point-concentration curve ($T - x$) and dew point-concentration curve ($T - y$, or $P - y$), has been proposed by Carlson and Colburn (*5*), Redlich and Kister (*13*), Gautreaux and Coates (*7*), and Ellis and Jonah (*6*), and recently examined by Slocum and Dodge (*15*), and Kojima et al. (*9, 11*).

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But for the infinite dilution ternary activity coefficients work has been done only by Buchholz-Meisenheimer and Kortüm (3), who proposed an approximate expression by modifying the Carlson and Colburn method for binary systems.

This paper presents a thermodynamic expression for determining the infinite dilution ternary activity coefficients from $T - y$ data, and presents a new method for measuring the ternary $T - y$ data. This technique is illustrated by determination of the infinite dilution ternary activity coefficients for the ethanol-isopropyl alcohol-water system at 1 atm.

THEORY

According to the criterion of vapor-liquid equilibrium, the activity coefficient of a component at infinite dilution is represented as follows

$$\frac{\gamma^0_{iL}}{\gamma^0_{iV}} = \lim_{x_i \to 0} \frac{\gamma_{iL}}{\gamma_{iV}} = \left[\left(\frac{f_{iV}}{f_{iL}} \right) \left(\frac{dy_i}{dx_i} \right) \right]_{x_i=0} \tag{1}$$

Equation (1) indicates that γ^0_{iL} can be calculated by knowing the value of $(dy_i/dx_i)x_i = 0$ if the fugacities of the pure component i in the vapor and liquid phase f_{iV} and f_{iL} and the activity coefficient in the vapor phase γ^0_{iV} can be obtained by using the equations of state or from the experimental $p - v - T$ data.

Then we start with the derivation of the relationship between $(dy_i/dx_i)\ x_i = 0$ and $T - y$ data for a ternary system. Here γ_{iV} is treated as unity, so that the symbol γ_i is used to denote only the activity coefficient of a component i in the liquid phase. For this purpose we introduce the useful expression recently proposed by Tao (*16*), which rigorously describes the molar enthalpy difference between two equilibrium phases and $x - y - T$ data for an isobaric multicomponent system

$$\sum_i (x_i - y_i)\, d \ln \hat{f}_{iV} = \frac{\Delta H - \Delta H^*}{RT^2}\, dT \tag{2}$$

where $\hat{f}_{iV}$ is the fugacity of component i in the saturated vapor phase, ΔH is the difference between the saturated vapor enthalpy and the saturated liquid enthalpy, and ΔH^* is the difference between the ideal gas enthalpy with vapor phase composition and the ideal gas enthalpy with liquid phase composition.

For the ternary system, a convenient path to determine $(dy_1/dx_1)\ x_1 = 0$ from $T - y$ data is the constant $y_2 = y_2/(y_2 + y_3)$ line. Taking the limit of Equation (2) along the Y_2 path at $x_1 = 0$, whence $y_1 = 0$, $\lim_{x_1 \to 0} [(x_1 - y_1)/\hat{f}_{1V}]$ $d\hat{f}_{1V} = d(x_1 - y_1)$ (*16*). Then

$$\left[\left(\frac{dx_1}{dy_1} \right)_{Y_2} \right]_{x_1=0} = 1 - (x_2' - y_2') \left[\left(\frac{d \ln \hat{f}_{2V}/\hat{f}_{3V}}{dy_1} \right)_{Y_2} \right]_{y_1=0} + \left(\frac{\Delta H - \Delta H^*}{RT^2} \right)_{2\text{-}3} \left[\left(\frac{dT}{dy_1} \right)_{Y_2} \right]_{y_1=0} \tag{3}$$

where $[(dT/dy_1)Y_2]y_1 = 0$ is the limiting slope of $T - y$ curve for ternary system at $y_1 = 0$ along path Y_2, and x_2' and y_2' denote respectively mole fractions in the liquid and vapor phases of component 2 in the binary system 2-3.

The term $[(\Delta H - \Delta H^*)/RT^2]_{2\text{-}3}$ in Equation (3) is of the binary system 2-3 and is given in the form derived from Equation (2)

$$\left(\frac{\Delta H - \Delta H^*}{RT^2} \right)_{2\text{-}3} = (x_2' - y_2') \left(\frac{d \ln \hat{f}_{2V}/\hat{f}_{3V}}{dy_2'} \right)_{2\text{-}3} \Bigg/ \left(\frac{dT}{dy_2'} \right)_{2\text{-}3} \tag{4}$$

where $(dT/dy_2')_{2\text{-}3}$ is the slope of $T - y$ curve for binary system 2-3 at $Y_2 = y_2'$.

A combination of Equation (1) with Equations (3) and (4) shows that the infinite dilution ternary activity coefficients in liquid phase may be calculated from the $T - y$ data, if the vapor-liquid equilibrium data for the three associated binary systems and the equation of state or experimental $p - v - T$ data for determining the fugacities in the ternary system are available.

In the case of a sufficiently low pressure, where the vapor phase may be treated as an ideal gas, that is, $\hat{f}_{iV} = y_iP$, $f_{iV} = P$ and $f_{iL} = P_i$, we note that the equations $dy_2 = -Y_2\, dy_1$ and $dy_3 = -(1 - Y_2)dy_1$ may be applied along Y_2 constant path for the ternary $T - y$ data. Combining Equation (1) with Equation (3) gives Equation (5), and Equation (4) is reduced to Equation (6)

$$\gamma_1{}^0 = \left(\frac{P}{P_1} \right) \Bigg/ \left\{ 1 + \left(\frac{\Delta H - \Delta H^*}{RT^2} \right)_{2\text{-}3} \left[\left(\frac{dT}{dy_1} \right)_{Y_2} \right]_{y_1=0} \right\} \tag{5}$$

$$\left(\frac{\Delta H - \Delta H^*}{RT^2} \right)_{2\text{-}3} = \left(\frac{x_2'}{y_2'} - \frac{x_3'}{y_3'} \right) \Bigg/ \left(\frac{dT}{dy_2'} \right)_{2\text{-}3} \tag{6}$$

where the liquid phase composition x_2' may also be calculated from the $(T - y)$ curve for binary system 2-3 by Equation (7), proposed by Kojima (9), based on the Gibbs-Duhem equation assuming the ideal behavior of the vapor phase and no liquid heat of mixing

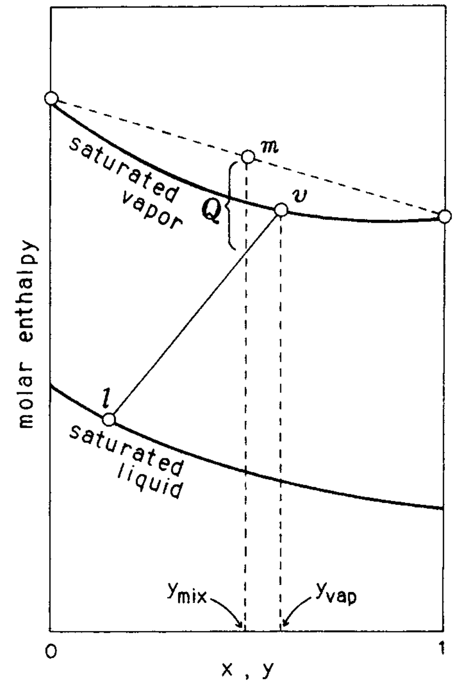

Fig. 1. Enthalpy versus concentration diagram.

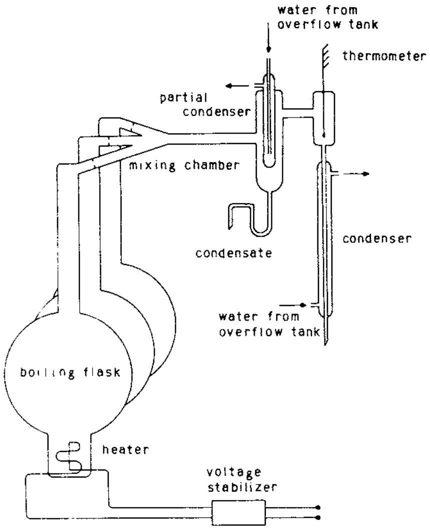

Fig. 2. Schematic diagram of experimental apparatus.

$$x_2' = \frac{y_2' + y_2'\, y_3' \left(\frac{dT}{dy_2'}\right)_{2\text{-}3} \frac{d \ln P_3}{dT}}{1 - y_2'\, y_3' \left(\frac{dT}{dy_2'}\right)_{2\text{-}3} \frac{d \ln P_2/P_3}{dT}} \qquad (7)$$

Equation (7) can also be directly obtained from Equation (2) by using $\Delta H - \Delta H^* = x_2'\lambda_2 + x_3'\lambda_3$ and $\lambda_i = RT^2\, d \ln P_i/dT$, that is, ideal behavior of the vapor phase and no liquid heat of mixing are assumed.

Combining Equation (5) with Equations (6) and (7) gives Equation (8)

$$\gamma_1{}^0 = \frac{P}{P_1} \Bigg/ \left\{ 1 + \frac{\left(\frac{d \ln P_3}{dT} + y_2' \frac{d \ln P_2/P_3}{dT}\right) \left[\left(\frac{dT}{dy_1}\right)_{Y_2} \right]_{y_1 = 0}}{1 - y_2'\, y_3' \frac{d \ln P_2/P_3}{dT} \left(\frac{dT}{dy_2'}\right)_{2\text{-}3}} \right\} \qquad (8)$$

Thus, the infinite dilution ternary activity coefficients are determined from the experimental $T - y$ data for the ternary and its three associated binary systems for low pressure systems with no liquid heat of mixing.

EXPERIMENT

Basic Principle

The dew point isobar of a mixture can be determined essentially by measuring the temperatures and the compositions of saturated vapor mixtures at a constant pressure. The principle of this can be seen from Figure 1, which shows the enthalpy-concentration diagram for a binary mixture. That is, first the saturated vapor of the individual pure constituents 1 and 2, which are separately vaporized each at constant rate, are mixed under adiabatic isobaric condition to form the vapor mixture of the composition, for example y_{mix} (m) as shown in Figure 1. Then the saturated vapor mixture, as for example, y_{vap} (v) in Figure 1, can be formed by partially condensing the vapor mixture (m). That is, the heat Q is absorbed from the vapor mixture. In this case, if the partial condensation is carried out such that the condensate is in equilibrium with the uncondensed vapor, the saturated liquid (l) will be in equilibrium with the vapor (v). But in practice, it is difficult to obtain such a saturated liquid. It is, however, possible to obtain only the saturated vapor mixture (v) by a relatively simple condenser. Thus a dew point for a binary mixture can be simply determined by measuring the temperature and the composition of a saturated vapor mixture.

Apparatus

The dew point isobar of a ternary system was measured by a flow method, as shown schematically in Figure 2. The apparatus consists of the following parts: three boiling vessels, a mixing chamber, a partial condenser for the vapor mixture, a section for measuring the temperature of a saturated vapor mixture and a total condenser.

The boiling vessels, in which the vapors of the individual pure constituents are separately vaporized, have the capacity of 2*l* each. Each vessel is fitted with an internal heater and is packed with Raschig rings in order to ensure the steady generation of vapor bubbles.

The partial condenser is a cold finger type. A highly effective cooler, the lower end of which is fitted with a drop counter, is used for total condensation of the vapors passed through the thermometer well. All parts of the apparatus, except the boiling vessels and the total condenser, are vacuum-jacketed and silvered to ensure adiabatic operation.

Temperature measurements were made, after calibration, by using a Yokogawa P-7W potentiometer connected with a galvanometer and a platinum resistance thermometer with an accuracy of ± 0.01°C.

Procedure

About 1 liter each of the three pure liquids were initially placed in the separated boiling vessels. The liquids were brought to a boil and the saturated vapors from the boiling vessels were thoroughly mixed in the chamber to form the vapor mixture of a constant concentration. Then the vapor mixture was introduced to the partial condenser, in which the

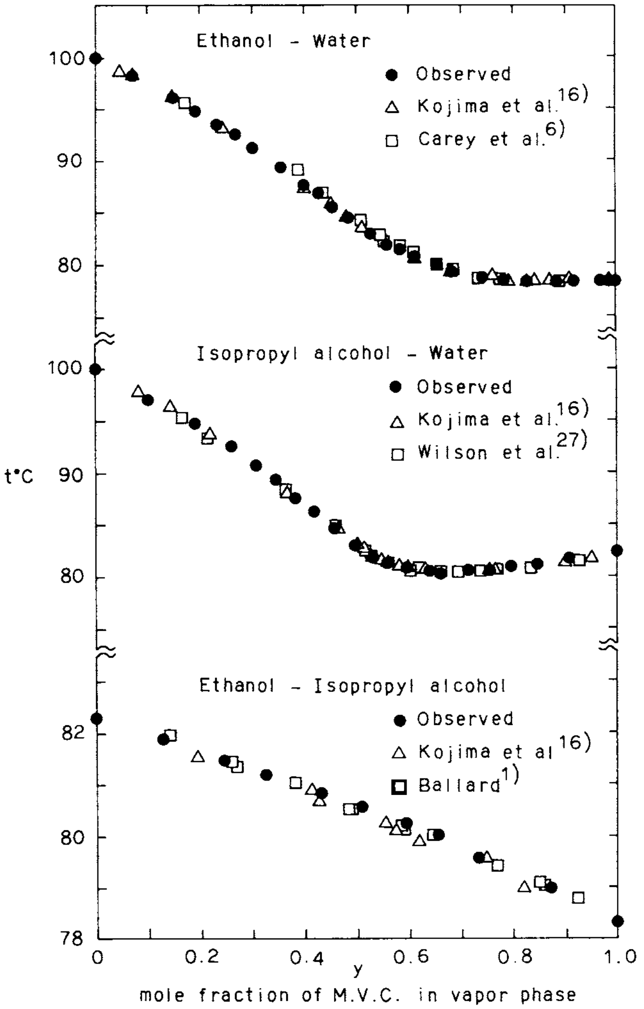

Fig. 3. Experimental dew point data for the binary systems at 760 mm Hg.

TABLE 1. SMOOTHED DEW POINT DATA FOR THREE BINARY SYSTEMS AT 760 mm. Hg

Ethanol-water		Isopropyl alcohol-water	Ethanol-isopropyl alcohol
*y**	*t*, °C.	*t*, °C.	*t*, °C.
0.00	100.00	100.00	82.26
0.05	98.62	98.68	82.08
0.10	97.21	97.34	81.91
0.15	95.79	95.85	81.75
0.20	94.26	94.17	81.57
0.25	92.63	92.42	81.40
0.30	90.97	90.60	81.24
0.35	89.31	88.78	81.08
0.40	87.48	86.80	80.92
0.45	85.69	84.68	80.75
0.50	83.83	82.54	80.57
0.55	82.07	81.22	80.37
0.60	80.80	80.54	80.17
0.65	79.82	80.27	79.96
0.70	79.07	80.24	79.74
0.75	78.61	80.37	79.52
0.80	78.38	80.63	79.29
0.85	78.24	80.97	79.06
0.90	78.15	81.37	78.83
0.95	78.17	81.80	78.58
1.00	78.32	82.26	78.32

* More volatile component in vapor phase.

TABLE 2. EXPERIMENTAL DEW POINT DATA FOR ETHANOL (1), ISOPROPYL ALCOHOL (2), AND WATER (3) SYSTEM AT 760 mm. Hg

y_1	y_2	*t*, °C.	y_1	y_2	*t*, °C.	y_1	y_2	*t*, °C.
0.053	0.361	85.85	0.206	0.259	84.31	0.433	0.277	79.35
0.062	0.513	80.82	0.225	0.137	88.22	0.461	0.255	79.31
0.071	0.158	92.98	0.235	0.161	87.22	0.472	0.149	80.23
0.071	0.857	81.40	0.235	0.493	79.80	0.474	0.086	82.14
0.072	0.240	90.05	0.243	0.687	80.91	0.479	0.248	79.23
0.076	0.563	80.22	0.248	0.441	79.82	0.481	0.383	79.64
0.093	0.110	93.94	0.253	0.371	80.27	0.506	0.444	80.17
0.094	0.637	80.12	0.254	0.516	79.91	0.509	0.259	79.14
0.098	0.729	80.59	0.259	0.279	81.87	0.527	0.291	79.21
0.103	0.748	80.64	0.268	0.546	79.98	0.531	0.081	80.38
0.107	0.353	84.21	0.284	0.220	83.08	0.545	0.322	79.46
0.109	0.522	80.18	0.297	0.465	79.72	0.557	0.148	79.23
0.110	0.820	81.32	0.308	0.376	79.76	0.583	0.344	79.64
0.119	0.416	81.74	0.313	0.605	80.55	0.611	0.067	79.38
0.136	0.624	80.12	0.325	0.393	79.56	0.628	0.150	78.88
0.144	0.475	80.38	0.336	0.548	80.29	0.649	0.284	79.48
0.145	0.577	80.01	0.354	0.108	84.80	0.654	0.210	79.16
0.154	0.238	87.21	0.383	0.230	80.28	0.688	0.142	78.66
0.155	0.538	80.00	0.386	0.521	80.28	0.720	0.055	78.59
0.175	0.124	90.89	0.391	0.315	79.52	0.797	0.174	79.07
0.194	0.107	90.72	0.395	0.472	79.93	0.827	0.046	78.35
0.200	0.342	81.61	0.420	0.122	82.26	0.946	0.016	78.31
0.201	0.654	80.39	0.420	0.315	79.38			

condensation was carried out such that uncondensed vapor and the condensate were formed. As the condensate was formed, it was continuously removed from the partial condenser and thus was not in equilibrium with the uncondensed vapor. The uncondensed vapor was introduced into the section where its temperature was measured. The vapor leaving this part was condensed in the total condenser. After steady state had been attained, that is, the temperature of the uncondensed vapor reached a constant value, the condensate from the total condenser was collected for analysis. The time required to attain steady state was about 10 min. The composition of the vapor mixture was controlled by changing the heating rate in the separate boiling vessels.

The analysis of the condensate was made at 30°C. by density and refractive index measurements, using Ostwald type pycnometers and an Atago-Abbe refractometer, respectively. For this purpose, calibration charts were made for the ternary system and its three associated binary systems.

Materials

Special grade ethanol and isopropyl alcohol (Wako pure chem. Ind. Ltd.) were used directly. The purity of the ethanol was greater than 99.80 wt. % and that of the isopropyl alcohol 99.98 wt. %.

EXPERIMENTAL RESULTS AND DISCUSSION

For three binary systems: ethanol-water, isopropyl alcohol-water and ethanol-isopropyl alcohol, the experimental dew points at 1 atm. are shown in Figure 3 and the smoothed values are presented in Table 1. For comparison, the literature values, all of which have been measured with ordinary vapor-liquid equilibrium equipment, are also shown in this figure.

For the ethanol-isopropyl alcohol-water system, the experimental dew-point data at 1 atm. is shown in Table 2.

The ternary $T - y$ data and its three associated binary $T - y$ data were then used to calculate the infinite dilution ternary activity coefficients in the liquid phase by Equation (8), under the assumption of an ideal behavior of the vapor phase and no liquid heat of mixing. The calculated values of γ_1^0 and the auxiliary calculation for ethanol are presented in Table 3. In the present calculation the binary slope $(dT/dy_2')_{2\text{-}3}$ was determined by graphical differentiation, and the ternary limiting slope $[(dT/dy_1)_{Y_2}]_{y_1=0}$ was determined from Equation (9) which is similar to that proposed by Ellis and Jonah (6) for the binary system.

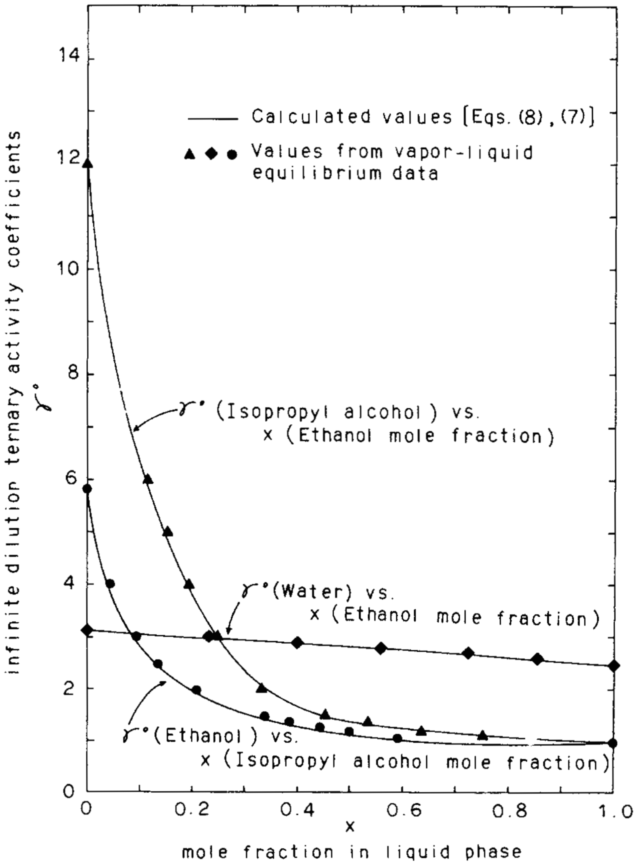

Fig. 4. The infinite dilution ternary activity coefficients versus liquid concentration.

TABLE 3. CALCULATION RESULTS ON THE INFINITE DILUTION TERNARY ACTIVITY COEFFICIENTS OF ETHANOL (1)

Y_2* (or y_2')	t, °C.	$\left(\frac{dT}{dy_2'}\right)_{2\text{-}3}$	x_2'**	$\left[\left(\frac{dT}{dy_1'}\right)_{Y_2}\right]_{y_1} = 0$	γ_1^0
0.0	100.00	−26.69	0.0000	−25.84	5.80
0.1	97.34	−29.14	0.0048	−25.11	5.57
0.2	94.17	−31.99	0.0106	−24.13	5.15
0.3	90.60	−35.33	0.0192	−22.97	4.79
0.4	86.80	−39.14	0.0358	−21.31	4.15
0.5	82.54	−36.44	0.1369	−16.72	2.54
0.6	80.54	−8.74	0.5152	−4.06	1.10
0.7	80.24	1.05	0.7089	−1.39	0.983
0.8	80.63	5.86	0.8379	−1.51	0.973
0.9	81.37	8.65	0.9313	−2.32	0.979
1.0	82.26	10.56	1.0000	−3.52	0.998

Vapor pressure data: Ethanol (*8*) (*12*) (*14*): $\log P_1 = 8.11576 - 1595.76/(t + 226.5)$
Isopropylalcohol (*2*): $\log P_2 = 7.72589 - 1351.627/(t + 196.726)$
Water (*8*): $\log P_3 = 7.9392 - 1650.4/(t + 226.27)$

* Component 2 = Isopropyl alcohol.
Component 3 = Water.

** Calculated values by Equation (7).

$$\left[\left(\frac{dT}{dy_1}\right)_{Y_2}\right]_{y_1=0} = \left[\left(\frac{\Delta T}{y_1(1-y_1)}\right)_{Y_2}\right]_{y_1=0} + (T - T_{2\text{-}3}) \quad (9)$$

where $\Delta T = T - [y_1T_1 + (1 - y_1)T_{2\text{-}3}]$, T = ternary dew point temperature along constant path Y_2, $T_{2\text{-}3}$ = dew point temperature of binary system 2-3 at $Y_2 = y_2'$, and T_1 = dew point temperature of pure component 1. The vapor pressure data for all the pure components were calculated by Antoine-type equations as shown in the footnote of Table 3.

The calculated γ^0's for ethanol are further shown in Figure 4, including more of the results for isopropyl alcohol and water. For comparison we show in Figure 4 the experimental γ^0's obtained by extrapolating the activity coefficient-concentration plots, which were constructed from the vapor-liquid equilibrium data for the ethanol-isopropyl alcohol-water system at 1 atm. reported by Kojima et al. (*10*). It can be seen that the agreement of the calculated and experimental values is satisfactory.

CONCLUSION

A thermodynamic expression for low pressure systems with no liquid heat of mixing was derived to determine the infinite dilution ternary activity coefficients from $T - y$ data, and a flow method for measuring the $T - y$ data was proposed. This technique was illustrated on the ethanol-isopropyl alcohol-water system at 1 atm.

NOTATION

f_i = fugacity of pure component i
f_i = fugacity of component i in a mixture
ΔH = enthalpy difference, cal./g.-mole
P = total pressure, mm. Hg
P_i = vapor pressure of component i, mm. Hg.
R = gas constant, 1.987 cal./g.-mole, °K.
t = temperature, °C.
T = temperature, °K.
x = mole fraction in liquid phase
y = mole fraction in vapor phase
Y_2 = $y_2/(y_2 + y_3)$
x_2' = mole fraction in liquid phase of component 2 in the binary system 2-3
y_2' = mole fraction in vapor phase of component 2 in the binary system 2-3

Greek Letters

γ = activity coefficient
γ^0 = activity coefficient at infinite dilution

Superscripts

0 = at infinite dilution
L = liquid phase
V = vapor phase
* = ideal gas state

Subscripts

1, 2, 3, i, = component 1, 2, 3, i
2-3 = 2-3 binary system

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