

# THERMODYNAMIC PROPERTIES OF AMMONIA-WATER MIXTURES

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## ABSTRACT

*An equation of state for ammonia-water mixtures is presented. Separate equations of state for liquid and gas phases are provided for pure ammonia and pure water. In the gas phase, the mixture is assumed to behave as an ideal solution, while in the liquid phase, the Gibbs excess energy is used to allow departure from ideal solution behavior. An existing correlation for the liquid Gibbs excess energy is modified to include experimental data at higher temperatures and pressures. The new correlation covers vapor-liquid equilibrium pressures of 0.2 to 110 bar and temperatures of 230 to 600 K. Comparisons with reported experimental data show good agreement.*

## INTRODUCTION

Many studies of the vapor-liquid equilibrium and thermodynamic properties of ammonia-water mixtures are cited in the literature. The temperature and pressure ranges of thermodynamic properties of most of the literature data are suitable for absorption-refrigeration cycle applications. Institute of Gas Technology tables (Macriss et al. 1964) cover the range up to a pressure of 34 bar and its corresponding saturation temperature. Using the IGT data, Schultz (1972) developed equations of state for a pressure range of 0.01 to 25 bar and a temperature range of 200 to 450 K. Ziegler and Trepp (1984) presented a new correlation of equilibrium properties of ammonia-water mixtures. They used an equation of state that is based on that developed by Schultz and extended the range of applicability to 500 K and 50 bar.

A study of power cycles using ammonia-water mixtures was recently initiated by Kalina (1983). For power cycles, thermodynamic data of ammonia-water mixtures at higher temperatures and pressures than those presented by IGT are required. Gillespie et al. (1987) published vapor-liquid equilibrium measurements for five isotherms between 313 and 588 K. Corresponding pressures ranged from 0.1 to 210 bar. Herold et al. (1988) developed a computer program for calculating the thermodynamic properties of ammonia-water mixtures using the Ziegler and Trepp correlations. El-Sayed and Tribus (1986) presented a method for computing the thermodynamic properties of

mixtures from the properties of pure components to extend the property correlation to higher temperatures and pressures. Derived properties cover pressures of 0.1 to 110 bar and temperatures between 300 to 770 K. Kalina et al. (1986) presented a similar method to predict the thermodynamic properties of two miscible-component mixtures for purpose of power-cycle analysis. Park and Sonntag (1991) published a set of thermodynamic data of ammonia-water mixtures based on a generalized equation of state. The pressure and temperature ranges are extended to 200 bar, and 650 K, respectively. The data from Gillespie et al. (1987) are not included in their correlations.

In this study, thermodynamic properties for the ammonia-water mixture are correlated to the form of the equation of state given by Ziegler and Trepp (1984). Separate equations of state for liquid and the gas phases are provided for pure ammonia and pure water. The correlation given by Ziegler and Trepp for the Gibbs excess energy are modified to include Wiltec experimental data at higher temperatures and pressures. The new correlations cover vapor-liquid equilibrium pressures of 0.2 to 110 bar and temperature of 230 to 600 K.

## EQUATION OF STATE FOR PURE COMPONENTS

The fundamental equation of the Gibbs energy,  $G$ , of a pure component can be derived from known relations for volume and heat capacity as a function of temperature and pressure. The fundamental equation of the Gibbs energy is given in an integral form as

$$G = h_o - Ts_o + \int_{T_o}^T C_p dT + \int_{P_o}^P v dP - T \int_{T_o}^T \frac{C_p}{T} dT \quad (1)$$

where  $h_o$ ,  $s_o$ ,  $T_o$ , and  $P_o$  are the enthalpy, entropy, temperature, and pressure at the reference state. The volume,  $v$ , and the heat capacity at constant pressure,  $C_p$ , for liquid phase are assumed to fit the following empirical relations proposed by Ziegler and Trepp (1984):

$$v^L = a_1 + a_2 P + a_3 T + a_4 T^2 \quad (2)$$

$$C_p^L = b_1 + b_2 T + b_3 T^2 \quad (3)$$

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For the gas phase, the corresponding empirical relations are

$$v^g = RT/P + c_1 + c_2/T^3 + c_3 T^{11} + c_4 P^2/T^{11} \quad (4)$$

$$C_P^{g,o} = d_1 + d_2 T + d_3 T^2 \quad (5)$$

where the superscripts are *L* for liquid, *g* for gas, and *o* for the ideal gas state. Integration leads to the following equations for the Gibbs energy for the pure components.

Liquid phase:

$$\begin{aligned} G_r^L = & h_{r,o}^L - T_r s_{r,o}^L + B_1 (T_r - T_{r,o}) \\ & + \frac{B_2}{2} (T_r^2 - T_{r,o}^2) + \frac{B_3}{3} (T_r^3 - T_{r,o}^3) \\ & - B_1 T_r \ln(T_r/T_{r,o}) - B_2 T_r (T_r - T_{r,o}) \\ & - \frac{B_3}{2} T_r (T_r^2 - T_{r,o}^2) \\ & + (A_1 + A_3 T_r + A_4 T_r^2) (P_r - P_{r,o}) \\ & + \frac{A_2}{2} (P_r^2 - P_{r,o}^2) \end{aligned} \quad (6)$$

Gas phase:

$$\begin{aligned} G_r^g = & h_{r,o}^g - T_r s_{r,o}^g + D_1 (T_r - T_{r,o}) \\ & + \frac{D_2}{2} (T_r^2 - T_{r,o}^2) + \frac{D_3}{3} (T_r^3 - T_{r,o}^3) \\ & - D_1 T_r \ln(T_r/T_{r,o}) - D_2 T_r (T_r - T_{r,o}) \\ & - \frac{D_3}{2} T_r (T_r^2 - T_{r,o}^2) \\ & + T_r \ln(P_r/P_{r,o}) + C_1 (P_r - P_{r,o}) \\ & + C_2 (P_r/T_r^3 - 4 P_{r,o}/T_r^3 + 3 P_{r,o} T_r/T_{r,o}^4) \\ & + C_3 (P_r/T_r^{11} - 12 P_{r,o}/T_{r,o}^{11} + 11 P_{r,o} T_r/T_{r,o}^{12}) \\ & + \frac{C_4}{3} (P_r^3/T_r^{11} - 12 P_{r,o}^3/T_{r,o}^{11} + 11 P_{r,o}^3 T_r/T_{r,o}^{12}) \end{aligned} \quad (7)$$

where the reduced thermodynamic properties are defined as

$$\begin{aligned} T_r &= T/T_B, \\ P_r &= P/P_B, \\ G_r &= G/RT_B, \\ h_r &= h/RT_B, \\ s_r &= s/R, \text{ and} \\ v_r &= vP_B/RT_B. \end{aligned} \quad (8)$$

The reference values for the reduced properties are  $R = 8.314 \text{ kJ/kmole}\cdot\text{K}$ ,  $T_B = 100 \text{ K}$ , and  $P_B = 10 \text{ bar}$ .

The molar specific enthalpy, entropy, and volume are related to Gibbs free energy by

$$h = -T^2 \left[ \frac{\partial}{\partial T} (G/T) \right]_P \quad (9)$$

$$s = - \left[ \frac{\partial G}{\partial T} \right]_P \quad (10)$$

TABLE 1  
Coefficients for Equations 6 and 7  
for Pure Water and Pure Ammonia  
(from Ziegler and Trepp [1984])

Coefficient	Ammonia	Water
A <sub>1</sub>	3.971423 10 <sup>-2</sup>	2.748796 10 <sup>-2</sup>
A <sub>2</sub>	-1.790557 10 <sup>-5</sup>	-1.016665 10 <sup>-5</sup>
A <sub>3</sub>	-1.308905 10 <sup>-2</sup>	-4.452025 10 <sup>-3</sup>
A <sub>4</sub>	3.752836 10 <sup>-3</sup>	8.389246 10 <sup>-4</sup>
B <sub>1</sub>	1.634519 10 <sup>+1</sup>	1.214557 10 <sup>+1</sup>
B <sub>2</sub>	-6.508119	-1.898065
B <sub>3</sub>	1.448937	2.911966 10 <sup>-1</sup>
C <sub>1</sub>	-1.049377 10 <sup>-2</sup>	2.136131 10 <sup>-2</sup>
C <sub>2</sub>	-8.288224	-3.169291 10 <sup>+1</sup>
C <sub>3</sub>	-6.647257 10 <sup>+2</sup>	-4.634611 10 <sup>+4</sup>
C <sub>4</sub>	-3.045352 10 <sup>+3</sup>	0.0
D <sub>1</sub>	3.673647	4.019170
D <sub>2</sub>	9.989629 10 <sup>-2</sup>	-5.175550 10 <sup>-2</sup>
D <sub>3</sub>	3.617622 10 <sup>-2</sup>	1.951939 10 <sup>-2</sup>
h <sub>r,o</sub> <sup>L</sup>	4.878573	21.821141
h <sub>r,o</sub> <sup>g</sup>	26.468873	60.965058
s <sub>r,o</sub> <sup>L</sup>	1.644773	5.733498
s <sub>r,o</sub> <sup>g</sup>	8.339026	13.453430
T <sub>r,o</sub>	3.2252	5.0705
P <sub>r,o</sub>	2.0000	3.0000

$$v = \left[ \frac{\partial G}{\partial P} \right]_T \quad (11)$$

In terms of reduced variables, Equations 9 through 11 become

$$h = -RT_B T_r^2 \left[ \frac{\partial}{\partial T_r} (G_r/T_r) \right]_P \quad (12)$$

$$s = -R \left[ \frac{\partial G_r}{\partial T_r} \right]_P \quad (13)$$

$$v = \frac{RT_B}{P_B} \left[ \frac{\partial G_r}{\partial P_r} \right]_T \quad (14)$$

#### Ammonia-Water Liquid Mixtures

The Gibbs excess energy for liquid mixtures allows for deviation from ideal solution behavior. The Gibbs excess energy of the liquid mixture is expressed by the relation proposed by Redlich and Kister (Reid and Prausnitz 1987;

TABLE 2  
Coefficients for Gibbs Excess Energy Function

E <sub>1</sub>	-41.733398	E <sub>9</sub>	0.387983
E <sub>2</sub>	0.02414	E <sub>10</sub>	-0.004772
E <sub>3</sub>	6.702285	E <sub>11</sub>	-4.648107
E <sub>4</sub>	-0.011475	E <sub>12</sub>	0.836376
E <sub>5</sub>	63.608967	E <sub>13</sub>	-3.553627
E <sub>6</sub>	-62.490768	E <sub>14</sub>	0.000904
E <sub>7</sub>	1.761064	E <sub>15</sub>	24.361723
E <sub>8</sub>	0.008626	E <sub>16</sub>	-20.736547

Ziegler and Trepp 1984), which is limited to three terms and is given by

$$G_r^E = \{F_1 + F_2(2x - 1) + F_3(2x - 1)^2\} \times (1 - x) \quad (15)$$

where

$$F_1 = E_1 + E_2 P_r + (E_3 + E_4 P_r) T_r + E_5 / T_r + E_6 / T_r^2 \quad (16)$$

$$F_2 = E_7 + E_8 P_r + (E_9 + E_{10} P_r) T_r + E_{11} / T_r + E_{12} / T_r^2 \quad (17)$$

$$F_3 = E_{13} + E_{14} P_r + E_{15} / T_r + E_{16} / T_r^2 \quad (18)$$

The objective is to extend Ziegler and Trepp correlations to include the experimental data reported by Gillespie et al (1987). All of the experimental data reported in Gillespie et al. were used to determine the activity coefficients and excess volume. Ziegler and Trepp correlations were used to generate approximately 4,000 points of activity coefficients, excess volume, and excess enthalpy in the range of their accuracy. Based on these data, a weighted least-squares linear regression was employed to best fit coefficients  $E_1$  through  $E_{16}$  appearing in the relations shown below for excess volume, excess enthalpy, and activity coefficients.

The excess enthalpy, entropy, and volume for the liquid mixtures are given as

$$h^E = -RT_B T_r^2 \left[ \frac{\partial}{\partial T_r} (G_r^E / T_r) \right]_{P_r, x} \quad (19)$$

$$s^E = -R \left[ \frac{\partial G_r^E}{\partial T_r} \right]_{P_r, x} \quad (20)$$

$$v^E = \frac{R T_B}{P_B} \left[ \frac{\partial G_r^E}{\partial P_r} \right]_{T_r, x} \quad (21)$$

The enthalpy, entropy, and volume of a liquid mixture are computed by Equations 22 through 25.

$$h_m^L = x h_a^L + (1 - x) h_w^L + h^E, \quad (22)$$

$$s_m^L = x s_a^L + (1 - x) s_w^L + s^E + s^{mix}, \quad (23)$$

$$s^{mix} = -R \{x \ln(x) + (1 - x) \ln(1 - x)\}, \quad (24)$$

$$v_m^L = x v_a^L + (1 - x) v_w^L + v^E. \quad (25)$$

### Ammonia-Water Vapor Mixtures

Ammonia-water vapor mixtures are assumed to be ideal solutions. The enthalpy, entropy, and volume of the vapor mixture are computed by Equations 26, 27, and 28:

$$h_m^g = y h_a^g + (1 - y) h_w^g, \quad (26)$$

$$s_m^g = y s_a^L + (1 - y) s_w^g + s^{mix}, \quad (27)$$

$$v_m^g = y v_a^L + (1 - y) v_w^g. \quad (28)$$

### Pressure-Temperature-Composition Behavior

At equilibrium, binary mixtures must have the same temperature and pressure. Moreover, the partial fugacity of each component in the liquid and gas mixtures must be equal. These conditions of phase equilibrium of mixtures can be written as four equations:

$$T^L = T^g = T, \quad (29)$$

$$P^L = P^g = P, \quad (30)$$

$$\hat{f}_a^L = \hat{f}_a^g, \quad (31)$$

$$\hat{f}_w^L = \hat{f}_w^g, \quad (32)$$

where  $P$  and  $T$  are the equilibrium pressure and temperature of the mixture, and  $\hat{f}$  is the fugacity of each component in the mixture at equilibrium.

The fugacities of ammonia and water in liquid mixtures are given by Walas (1985):

$$\hat{f}_a^L = \gamma_a f_a^\circ x \delta_a, \quad (33)$$

$$\hat{f}_w^L = \gamma_w f_w^\circ (1 - x) \delta_w, \quad (34)$$

where

$\gamma$  = activity coefficient,  
 $f^\circ$  = standard state fugacity of pure liquid component corrected to zero pressure,

- $x$  = liquid mole fraction,  
 $\delta_i$  = Poynting correction factor from zero pressure to saturation pressure of mixture.

Assuming an ideal mixture in the vapor phase, the fugacities of the pure components in the vapor mixtures are given by

$$\hat{f}_a^s = \phi_a P y, \quad (35)$$

$$\hat{f}_w^s = \phi_w P(1 - y), \quad (36)$$

where

- $\phi$  = fugacity coefficient,  
 $y$  = vapor mole fraction,  
 $P$  = saturation pressure of mixture.

The standard state fugacities of pure liquid ammonia and water are

$$f_a^o = \phi_a^{sat} P_a^{sat} / \delta_a^{sat}, \quad (37)$$

$$f_w^o = \phi_w^{sat} P_w^{sat} / \delta_w^{sat}, \quad (38)$$

where  $P^{sat}$  is the saturation pressure of the pure component at the temperature of the system.  $\phi^{sat}$  and  $\delta^{sat}$  are the fugacity coefficient and the Poynting factor for each pure component's saturation pressure. Gillespie et al. (1987) derived an equation for the pure ammonia fugacity that extends beyond the critical point of ammonia.

$$f_a^o = 10.7215 - 4.9319(1000/T) + 1.4992(1000/T)^2 - 0.236202(1000/T)^3. \quad (39)$$

The activity coefficients are related to the total excess Gibbs free energy and are given by Reid et al. (1987):

$$T_r \ln(\gamma_a) = (F_1 + 3F_2 + 5F_3)(1-x)^2 - 4(F_2 + 4F_3)(1-x)^3 + 12F_3(1-x)^4, \quad (40)$$

$$T_r \ln(\gamma_w) = (F_1 - 3F_2 + 5F_3)x^2 + 4(F_2 - 4F_3)x^3 + 12F_3x^4 + 12F_3(1-x)^4. \quad (41)$$

The fugacity coefficient is calculated from the following equations:

$$\phi_a = \frac{P^o}{P} \exp \{ (G_{r,a} - G_{r,a}^o) / T_r \}, \quad (42)$$

$$\phi_w = \frac{P^o}{P} \exp \{ (G_{r,w} - G_{r,w}^o) / T_r \}, \quad (43)$$

where  $P^o$  is a low pressure at which the ideal gas assumption is valid.  $\phi_a = \phi_w = 1$  for an ideal gas.

The Poynting factor, which is important at high pressure (greater than 30 bar), is given by

$$\delta_a = \exp \{ (\bar{v}_a^L P - \bar{v}_a^L P_a^{sat}) / RT \}, \quad (44)$$

$$\delta_w = \exp \{ (\bar{v}_w^L P - \bar{v}_w^L P_w^{sat}) / RT \}, \quad (45)$$

where  $\bar{v}$  is the partial molar volume. At moderate pressure (less than 30 bar), the Poynting factor is very close to unity.

## Comparison with Literature Data

Figure 1 shows comparisons of calculated and experimental activity coefficients of different isotherms of ammonia and water, respectively. In general, the results show good agreement with the experimental data. Below temperatures of 406 K, the model is almost perfect. At temperatures higher than 406 K, the calculated values show an average deviation of less than 5%.

Calculated bubble and dew-point temperatures at constant pressures are compared to the IGT experimental

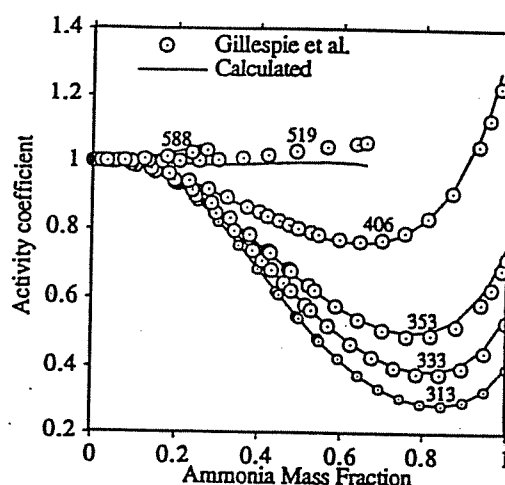


Figure 1a Activity coefficients of water as a function of liquid ammonia mass fraction.

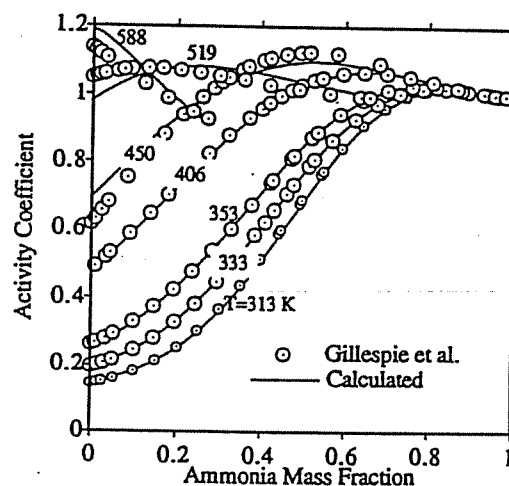


Figure 1b Activity coefficients of ammonia as a function of liquid ammonia mass fraction.

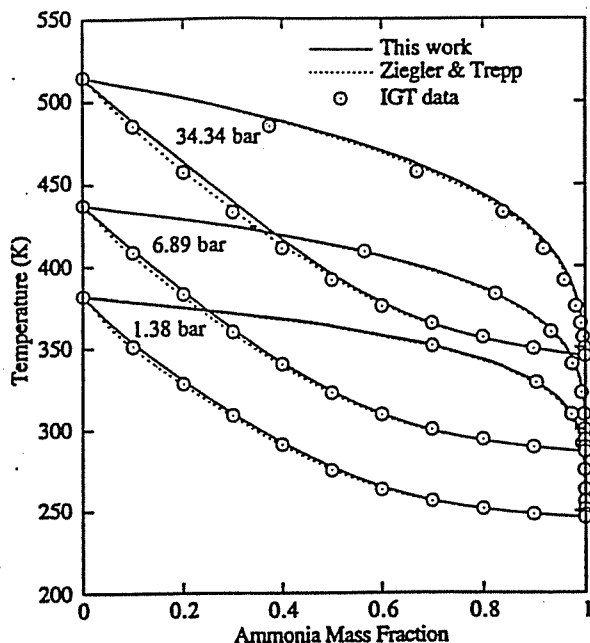


Figure 2 Bubble and dew points of ammonia water mixtures.

data. The Ziegler and Trepp correlation and the correlation of this work agree very well with the IGT experimental data, with a difference of less than 2%, as shown in Figure 2.

Pressure-concentration relationships for four isotherms are compared with the Wiltec experimental data, as shown in Figure 3. For temperatures less than 406 K, the Ziegler and Trepp correlation and this work predict values that compare to the experimental data with an average deviation of less than 3%. At higher temperatures, the results from the Ziegler and Trepp correlation start to deviate, with a difference of more than 15%. The revised correlation presented here shows much less deviation. At pressures of less than 110 bar, the maximum deviation is about 5%.

In Figure 4, the specific volume of saturated liquid is plotted as a function of the ammonia mass fraction for different isotherms. The curves show reasonable agreement with the Wiltec data. The deviations between the calculated and the experimental data are within the experimental error.

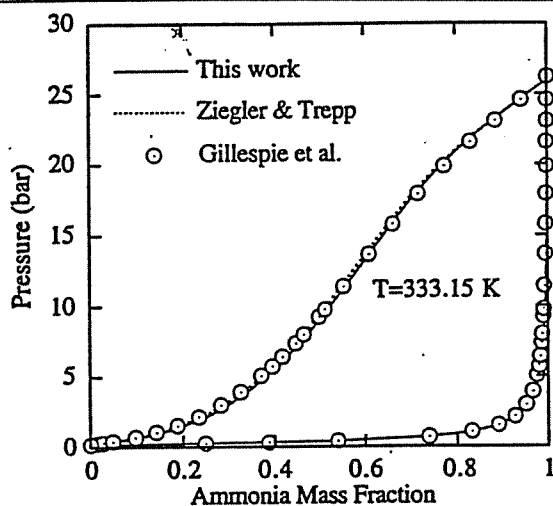


Figure 3a Saturation pressure of ammonia-water mixtures at a temperature of 333.15 K.

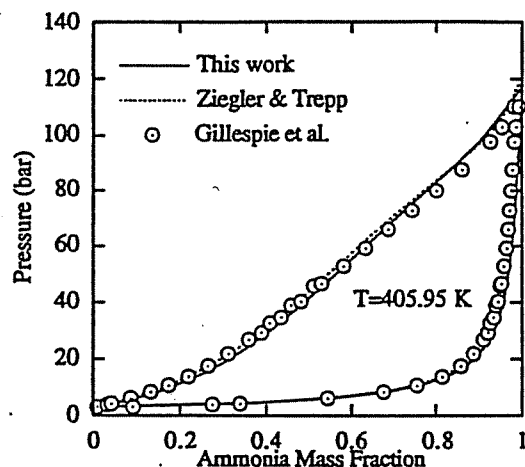


Figure 3b Saturation pressure of ammonia-water mixtures at a temperature of 405.95 K.

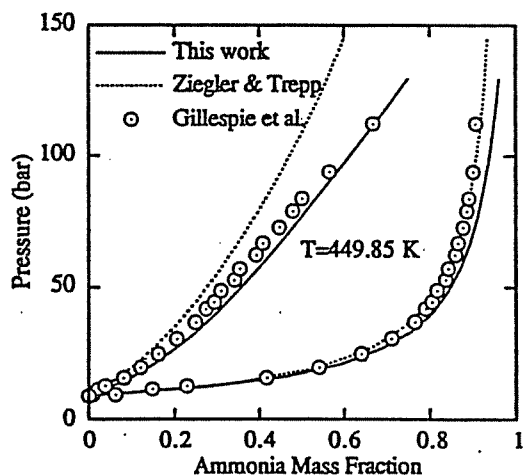


Figure 3c Saturation pressure of ammonia-water mixtures at a temperature of 449.85 K.

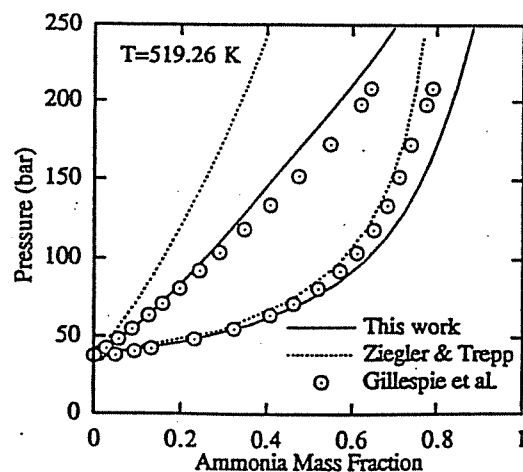


Figure 3d Saturation pressure of ammonia-water mixtures at a temperature of 519.26 K.

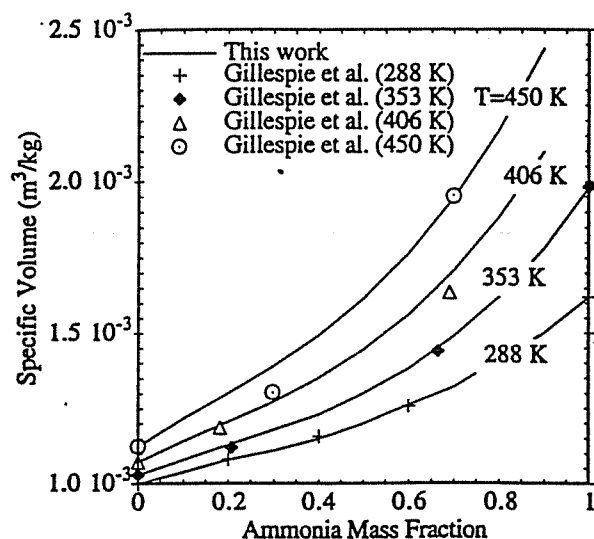


Figure 4 Specific volume of liquid ammonia-water mixtures at different isotherms.

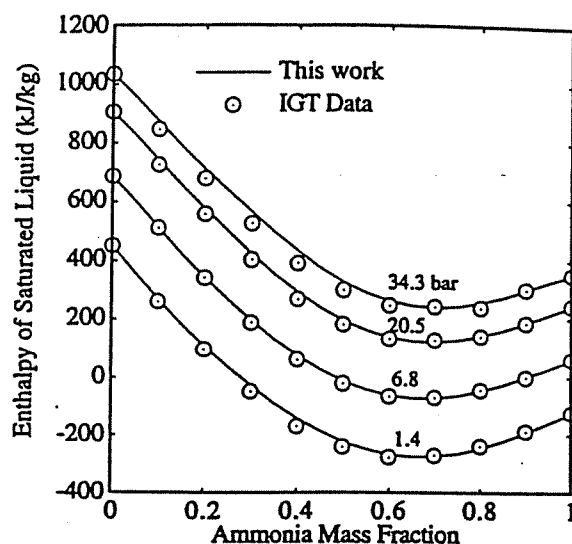


Figure 5 Enthalpy of saturated liquid ammonia-water mixtures at constant pressures.

The enthalpy of saturated liquid of this work is compared with IGT tables, as shown in Figure 5. At a pressure of 34.34 bar, the model predicts values that are about 2% high. At lower pressures, the predicted values agree with the IGT data with a deviation of less than 2%.

Figure 6 shows the enthalpy of saturated vapor ammonia-water mixtures at constant pressures. At pressures of 1.4 and 6.9 bar, the calculated enthalpy of saturated vapor is in fair agreement with IGT data. However, the calculated enthalpy tends to be consistently high as the pressure increases. The difference increases as the pressure increases. At a pressure of 34.34 bar (500 psia), the maximum difference is about 5%.

In Figures 7 through 11, the results of this study are compared with the generalized correlation proposed by Park and Sonntag (1991). Figure 7 shows the bubble and dew-point temperatures of ammonia-water mixtures for constant pressures of 50 and 100 bar. The bubble-point temperature (from Park and Sonntag) is consistently lower compared to that in this work. However, the differences are less than 5%.

The enthalpy and entropy of saturated vapor are compared with the Park and Sonntag tables in Figures 8 and 9. The enthalpy of saturated vapor of this study is in good agreement with the Park and Sonntag tables for pressures of 50 and 100 bar. Figure 9 shows a maximum difference of 10% between the entropy of saturated vapor in this study and the Park and Sonntag tables. The enthalpy and entropy of saturated liquid at 50 and 100 bar are compared with the Park and Sonntag tables in Figures 10 and 11. The comparison shows a difference of more than 25% at these high pressures. Park and Sonntag's data can be significantly in error at pressures higher than the critical pressure of the ammonia, where nonexistent equilibrium data are tabulated for pure ammonia without paying attention to the critical properties of the mixtures.

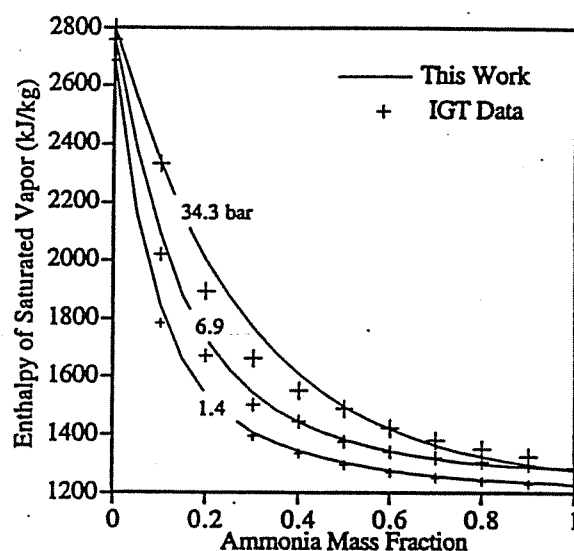


Figure 6 Enthalpy of saturated vapor ammonia-water mixtures at constant pressures.

## CONCLUSIONS

A new correlation of the equilibrium properties of ammonia-water mixtures is presented that extends the applicable range of pressure and temperature. The new correlations are useful for designing and analyzing power absorption cycles. The correlation covers vapor-liquid equilibrium pressures of 0.2 to 110 bar and temperatures of 230 to 600 K. Comparisons with reported experimental data show good agreement, with an average deviation of less than 5%. The correlation can be used to pressures of 150 bar with a maximum deviation of around 10%.

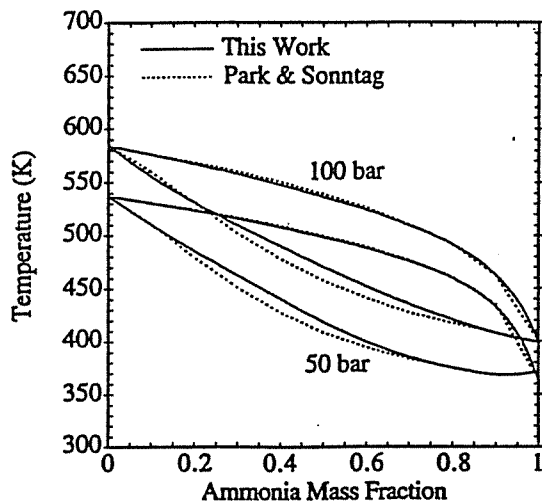


Figure 7 Bubble and dew points of ammonia-water mixtures.

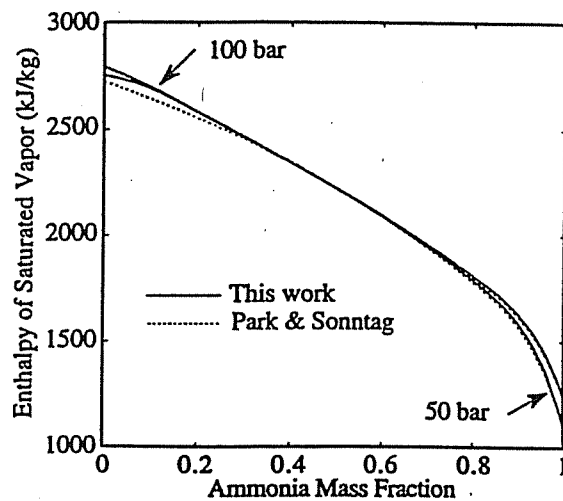


Figure 8 Enthalpy of saturated vapor.

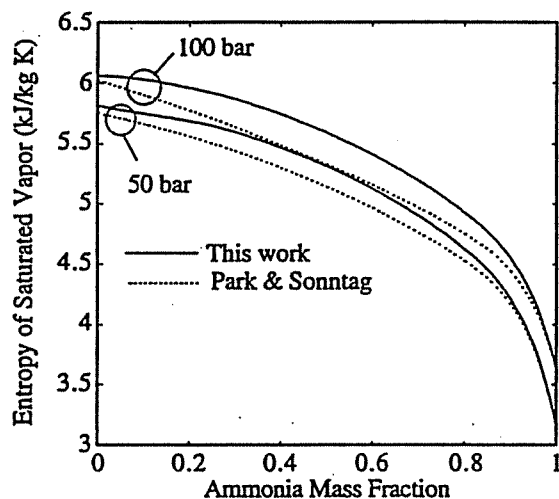


Figure 9 Entropy of saturated vapor.

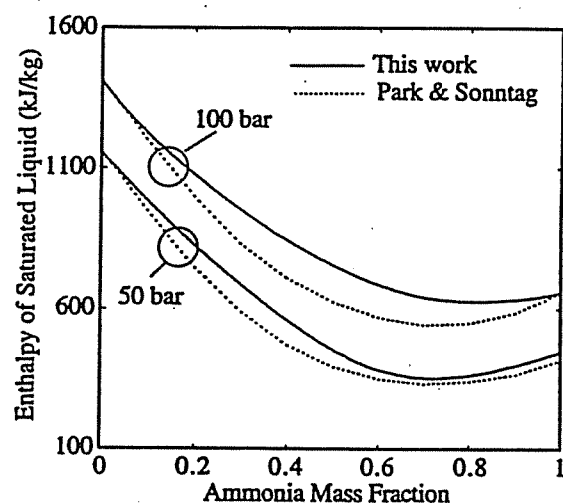


Figure 10 Enthalpy of saturated liquid.

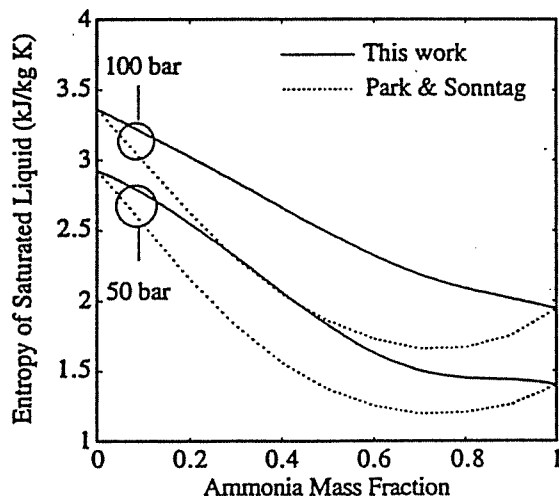


Figure 11 Entropy of saturated liquid.

## NOMENCLATURE

$a, b, c, d$	= coefficients
$A, B, C, D, E, F$	= dimensionless coefficients
$C_p$	= heat capacity at constant pressure, kJ/kmole K
$G$	= Gibbs free energy, kJ/kmole
$h$	= molar enthalpy, kJ/kmole
$f$	= fugacity, bar
$\hat{f}$	= fugacity of each component in the mixture at equilibrium, bar
$p$	= pressure, bar
$R$	= gas constant 8.314, kJ/kmole K
$s$	= molar entropy, kJ/kmole K
$T$	= temperature, K
$v$	= molar volume, m <sup>3</sup> /kmole

$\bar{v}$	= partial molar volume
$x$	= ammonia mole fraction
$y$	= water mole fraction

#### Greek Symbols

$\gamma$	= activity coefficient
$\phi$	= fugacity coefficient
$\delta$	= Poynting factor

#### Subscripts

$a$	= ammonia
$o$	= reference state
$r$	= reduced, dimensionless
$w$	= water

#### Superscripts

$g$	= gas phase
$L$	= liquid phase
$o$	= ideal gas state
$sat$	= saturation

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