SIMPLE FUNCTIONS FOR PREDICTING THE THERMODYNAMIC PROPERTIES OF AMMONIA - WATER MIXTURE

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Abstract

The engineering calculation and simulation of absorption refrigeration systems require the availability of simple and efficient functions for the determination of thermodynamic property values of the operating fluid. This work presents a set of five simple and explicit functions for the determination of the vapor-liquid equilibrium properties of the ammonia-water mixture. The functions are constructed by the least square method for curve fitting using the valid available data in the literature. The presented functions are valid for $-20 \leq T \leq 140^\circ C$, $0.2 \leq P \leq 100$ bar, and the entire composition range (i.e. $0 \leq x \leq 1$), which cover the region within, and also by which absorption cycles commonly operate. Obtained results are compared in detail with other correlations in the literature in graphical and statistical forms and reasonable accuracy are observed. Standard deviation of presented functions are 3.44 kJ/kg for the enthalpy of saturated liquid mixture, 14.06 kJ/kg for the enthalpy of saturated vapor mixture, and 0.0123 kg - NH$_3$/kg - mix for ammonia mass fraction in vapor phase.

Keywords

Ammonia-Water Mixture, Thermodynamic Properties, Absorption Refrigeration Systems

1. INTRODUCTION

Due to the ozone depletion problem associated with the use of CFC and HCFC refrigerants, increasing interest in absorption heat pumps and refrigeration systems has taken place in recent years. More and more, they are regarded not only as environmentally friendly alternatives to CFC based systems, but also related to energy efficient heating and cooling technology. Recently, aqueous mixtures of natural substances have been considered as working fluids for various purposes because of the lack of negative effects on the natural environment and excellent materials suitable for future communities with most material recycled as desired in the 21st century.
For example, ammonia-water mixtures have attracted much attention due to its potential as a working fluid in a solar assisted absorption refrigeration system. Also, in conventional steam power cycles, water is used as the working fluid. One way to improve the thermal efficiency is to replace the one-component fluid with a binary fluid. The most well-known power cycle with a binary working fluid (an ammonia-water mixture) is the Kalina cycle. This cycle has been shown to be more efficient than conventional power cycles for several applications [1-2]. For theoretical performance simulation of ammonia-water power cycles and absorption refrigeration cycles, thermodynamics properties are necessary. For this purpose, correlations for calculating of thermodynamic properties of binary mixtures have been presented by researchers [3-5]. Recently, in 1998 the newly sophisticated correlation for the thermodynamic properties of ammonia-water mixture has been presented by Tillner-Roth and Friend [6]. As mentioned in the literature, and as far as the author has found in the literature, this correlation provides reliable information on the thermodynamic properties of an ammonia-water mixture for the entire composition range and in a wide range of temperatures and pressures and has the-state-of-the-art accuracy. From the viewpoint of mathematical structure, their formulation is very complicated and requires more computational time for calculation and is not suitable for simulation and engineering applications.

In the present work, based on the obtained numerical data from Tillner-Roth and Friend formulation and the best fitted equation procedure, a set of five simple and explicit equations describing the vapor-liquid equilibrium properties of the ammonia-water mixture necessary for absorption cycle design, have been presented. The set comprises of the functions for calculating: the enthalpy of saturated liquid of mixture, the enthalpy of saturated vapor of mixture, the mass fraction of ammonia in the vapor phase as a function of pressure and mass fraction of ammonia in the liquid phase. Also, a function is developed for calculating the saturated pressure of the mixture as a function of temperature and compositions which is necessary for construction of the Dühring diagram. The enthalpy of saturated ammonia-water mixture is explicitly expressed in terms of two independent variables. The validity of the obtained functions is suitable for operation conditions of absorption refrigeration cycles. The predicted results from the present work are compared with available correlations in the literature.

2. AVAILABLE CORRELATIONS

The availability of new experimental data, especially of the thermodynamic properties of the mixture, has allowed the formulation of unified and conceptually simple equations based upon the Helmholtz or Gibbs free energies. A correlation for calculation of the thermodynamic properties of the ammonia-water mixture was developed by Stecco and Desideri [7]. Their correlation is based on work presented by Ziegler and Trepp [8]. Expressions for the Gibbs free energy, for which pressure, temperature, and mole fraction of ammonia are independent variables, are used as the fundamental function. Different equations are used for the vapor and liquid phases. The vapor is assumed to be an ideal mixture of real gases, while the properties of the liquid phase are corrected by a term calculated from the Gibbs excess energy. The same correlations as suggested by Stecco and Desideri have also been presented and used by Xu and Goswami [9]. Another correlation, also used in power cycle simulations, is the one presented by Ibrahim and Klein [10]. They use the same equations as Ziegler and Trepp. The constants in the function for the Gibbs excess energy have, however, been recalculated with experimental data at higher pressures and temperatures.

The presented correlation by Tillner-Roth and Friend [6] is based on a fundamental equation of state for the Helmholtz free energy. In this new correlation, the entire thermodynamic space of the mixture is described by one single equation, and the independent variables are volume, temperature, and a mole fraction of ammonia. The importance of the development work done in the new formulations is excellently demonstrated by the International Association for the Properties of Water and Steam (IAPWS) [11] for the calculation of power cycles operating with the ammonia-water mixture. The formulation of the equation is...
illustrated by Equation 1, where \( a \) is the molar Helmholtz free energy and \( v \) is the molar volume. The superscript \( r \) stands for residual.

\[
a(T, v, \zeta) = (1 - \zeta)a_{\text{H,O}}(T, v) + \zeta a_{\text{NH}_3}(T, v) + RT\left((1 - \zeta)\ln(1 - \zeta) + \zeta \ln \zeta + \Delta a^r(T, v, \zeta)\right)
\]  

(1)

The functions for the pure components, \( a_{\text{H,O}} \) and \( a_{\text{NH}_3} \), consist of a function for ideal-gas properties and a residual part correcting to the real-component behavior. The term for the departure from non-ideal mixture behavior, \( \Delta a^r(T, v, \zeta) \), is correlated with the most reliable, available experimental data for the ammonia-water mixture. Reduced properties are used in the correlations for the ammonia-water mixture as well. For a mixture liquid and vapor phase in equilibrium, the chemical potential of each component is the same in both the liquid and the vapor phase. By setting up equations for this equality and solving them iteratively, the saturation properties for the mixture can be calculated. In the correlations both by Ibrahim and Klein and by Tillner-Roth and Friend, this procedure for calculating the saturation properties is used. It should be noted that by Tillner-Roth and Friend, a different reference state for ammonia is used than in the other correlations. Therefore enthalpies calculated from the different correlations cannot be directly compared. Finally a set of five simple functions for fast calculations of selected thermodynamic properties of the ammonia-water mixture have been presented by Patek and Klomfar [12]. Their equations are suitable for the industrial design of absorption refrigeration equipment.

3. DERIVATION OF FUNCTIONS

The task of finding suitable equations to represent the thermodynamic properties of the ammonia-water mixture are a simple analytical form, fast evaluation, and a reasonable accuracy for industrial calculations. The vapor-liquid equilibrium data for the ammonia-water mixture from Tillner-Roth and Friend [6] have been taken into consideration and calculation in the present work. Based on this data, the functions with two variables are constructed for the calculation of some thermodynamic properties of the ammonia-water mixture by the least squares method. A mathematical procedure for finding the best fitting equation to a give a set of points are constructed by minimizing the sum of the squares of the offsets (the residuals) of the points from the fitted equation. While there are methodical procedures for fitting equations to data, the process is also an art. The art of intuition is particularly needed in deciding upon the form of the equation, namely, the choice of independent variables to be included and the form in which these variables should appear. There are no fixed rules for knowing the variables’ form in the equation, but making at least a rough plot of the data will often provide some insight. A requirement for keeping the equation simple is to choose the proper terms (polynomial, exponential, etc.) to include in the equations. It is possible, of course, to include all the terms that could possibly be imagined and evaluate the coefficient by the method of least squares.

In general, an approximating function of a thermodynamic quantity \( z \) depending on two independent variables \( u \) and \( v \) was sought in the form of:

\[
z(u, v) = \sum_{i=0}^{n} \sum_{j=0}^{m} \psi_j(\alpha_{i,j}, v) \phi_i(u)
\]  

(2)

Where \( \phi_i(u) \) and \( \psi_j(\alpha_{i,j}, v) \) are suitable functions which describes mathematical the behavior of data points. For the data points \( z(u, v_k) \) with \( v_k = \text{cte} \) it is assumed that the best fitted equations has the form of:

\[
z(u, v_k) = \sum_{i=0}^{n} \beta_i \phi_i(u) \quad \text{for } v_k = \text{cte}
\]  

(3)

Where the \( \beta \) constants are calculated from the nonlinear least squares fitting method using respective thermodynamic properties data from Ref. 6. For \( m \) pairs of data points the sum of the squares of the offsets and minimization relations can be written as:

\[
s_k = \sum_{m} \left[z(u_m, v_k)|_{\text{Ref.6}} - \sum_{i=0}^{n} \beta_i \phi_i(u_m)\right]^2
\]  

(4)
\[
\frac{\partial s_k}{\partial \beta_i} = 0 \quad i = 0, 1, \ldots, n
\]  
(5)

A similar procedure can be followed for the thermodynamic properties values for \( v_k = \text{cte} \) \((k = 1, 2, \ldots, l)\) and then the corresponding \( \beta \) constants are calculated. Next the \( b \) constants are fitted as a function in terms of \( v \) and could have the form of:

\[
b_i = \sum_{j=0}^{m_i} y_j (\alpha_{i,j} v)
\]  
(6)

Finally, the \( \alpha \) constants are calculated by applying the nonlinear least squares method again. The obtained functions are arranged for the ammonia-water mixture in the next section.

### 4. RESULTING FUNCTIONAL FORMS

The resulting functions for fast calculation of the thermodynamic properties of the ammonia-water mixture have the following structure:

- The enthalpy of saturated liquid of ammonia-water mixture (mixture bubble point enthalpy), \( h_L(x,T) \), as a function of ammonia mass fraction, \( x \), and temperature, \( T \):

\[
h_L(x,T) = \sum_{i=0}^{4} \left( a_i + b_i T + c_i T^2 + d_i T^3 \right) x^i
\]  
(7)

Range of validity: \(-20 \leq T \leq 140^\circ C\) and \( 0 \leq x \leq 1 \)
- The enthalpy of saturated liquid of the ammonia-water mixture, \( h_L(x,P) \), as a function of ammonia mass fraction, \( x \), and pressure, \( P \):

\[
h_L(x,P) = a_0 P^{b_0} + \sum_{i=1}^{4} \left( a_i + b_i P + c_i P^2 + d_i P^3 \right) x^i
\]  
(8)

Range of validity corrected to range of validity: \( 0.2 \leq P \leq 100 \text{ bar} \) and \( 0 \leq x \leq 1 \)
- The mass fraction of ammonia in vapor phase, \( y(x,P) \), as a function of ammonia mass fraction in liquid phase, \( x \), and pressure, \( P \):

\[
y(x,P) = 1 - \exp\left[ a(P) x + \left( c + \frac{d}{P} \right) x^2 \right]
\]  
(11)

Range of validity: \( 0.2 \leq P \leq 100 \text{ bar} \) and \( 0 \leq x \leq 1 \)

The respective coefficient of Equations 7-11 are given in Tables 1-5.

### 5. DISCUSSION

The presented functions provide reliable information on the thermodynamic properties of the ammonia-water mixture for the entire composition range and in a wide range of temperature and pressure. Specifically, the functions provide property data for all compositions from pure water to pure ammonia, vapor-liquid equilibrium properties between \((-20\) to \(140^\circ C\)) and up to 100 bar.

Although the presented functions are of completely mathematical nature, they allow the determination of the property of the ammonia-water mixture with accuracy acceptable for industrial use. The numerical solution of the present equations is simpler than that of the formulation recently adopted by the IAPWS based on the fundamental equations of state for Helmholtz free energy developed by Tillner - Roth and Friend. These equations also easily solved.
TABLE 1. Coefficients of Equation 7.

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<tr>
<th>i</th>
<th>a_i</th>
<th>b_i</th>
<th>c_i</th>
<th>d_i</th>
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<tr>
<td>4</td>
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TABLE 2. Coefficients of Equation 8.

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<td>4.0425×10^2</td>
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<table>
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<td>0</td>
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<td>-7.5905×10^{-4}</td>
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<td>+2.7155×10^{-5}</td>
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Equation 7 reproduces the data of Tillner-Roth and Friend for the specific enthalpy of the liquid phase with root mean square deviation 3.44 kJ/kg. The comparison of the predicted saturation properties for the liquid ammonia-water mixture is shown in Figures 1-4. Figure 1 shows the Enthalpy-Concentration diagram for the saturated liquid mixture of ammonia-water at various temperatures while Figure 2 illustrates various isobars on the Enthalpy-Concentration diagram. As it is evident from Figures 1 and 2, the comparison of predicted data from the present work and other correlations

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<th>c_i</th>
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<td>-</td>
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<td>3</td>
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<td>+1.0632 × 10^{-3}</td>
<td>-1.3020 × 10^{-4}</td>
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<td>-1.4100 × 10^2</td>
<td>-1.8000 × 10^{-2}</td>
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TABLE 5. Coefficients of Equation 11.

<table>
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<th>b</th>
<th>c</th>
<th>d</th>
</tr>
</thead>
<tbody>
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<td>-2.6700 × 10^{-1}</td>
<td>-2.2106 × 10^0</td>
<td>+2.7246 × 10^0</td>
</tr>
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</table>

show reasonable accuracy.

The predicted enthalpy for the mixture have been compared at a temperature of 177 [°C] and pressure of 108.2 bar with different correlations. As shown in Figure 3, at higher temperatures and pressures, the presented equations still show good agreement with other correlations. Figure 4 depicts the Dühring diagram i.e. ln P vs. -1/T, (also called PTX diagram) for the ammonia-water mixture for a pressure of up to 100 bar which is suitable for the presentation of the processes of absorption refrigeration cycles. Figure 5 shows the vapor-liquid equilibrium curves at 1, 20 and 60 bar.

To obtain a simple functional form for the y(x,P) relation, its region of validity has to be curtailed to the ammonia mass fraction in the liquid phase greater than 0.02 and a pressure above 0.2 bar. However, this limitation is not very onerous, since on the low temperature side of refrigeration systems a pressure above atmospheric is preferred to avoid leakage of air into them, and to increase the capacity of the cycle. The numerical consistency of the equations for P(x,T) and y(x,P) has been checked and found to be better than 1 % in y for x above 0.2 and not worse than 4 % for a concentration of less than 0.2. The Patek and Klomfar function for predicting vapor-liquid equilibrium conditions is suitable for 0.2 < P < 20 bar, while the presented equation is applicable for 0.2 < P < 100 bar. Values of enthalpy of the gas phase have been computed from Equation 10. The resultant values have been compared with those calculated from the formulation of Tillner-Roth and Friend and Patek and Klomfar. In both cases the differences are less than 2 % over the entire region from 0.2 to 100 bar. The part of the resultant enthalpy-concentration diagram concerning the gas phase is shown in Figures 6 and 7. The enthalpy of saturated ammonia vapor shows a maximum near 20 bar. This is why all isobars corresponding to a higher pressure intersect those of lower pressure, as seen on the isobars for 20 bar in Figure 7.

Figures 8-12 show the deviation between the calculated data for the fitted equations and values calculated from helmholtz free energy formulation by Tillner-Roth and Friend [6]. To prove that these assessments are still conservative, Figure 8 shows deviations between...
the calculated enthalpy of saturated liquid mixture from Equation 7 and highly accurate results from Ref. 6 which cover the range \(-20 \leq T \leq 140^o\) C and \(0 \leq x \leq 1\). With very few exceptions, all of these data are represented within ± 10 kJ/kg. Deviation between the calculated enthalpy of the saturated liquid mixture from Equation 8 and those values from Ref. 6 are depicted in Figure 9. Deviations for this enthalpy remain within - 15 to 25 kJ/kg. In Figures 10, 11, and 12 deviations between the calculated saturation pressure, enthalpy of saturated vapor, and ammonia mass fraction in the vapor phase from the fitted Equations 9-11 with those values from Ref. 6 are demonstrated respectively.

After using graphical methods to evaluate the goodness of fit, the goodness-of-fit statistics should be examined. For this purpose the root mean square error (RMSE) has been used to obtain data from the fitted equations. The RMSE is an estimate of the standard deviation of the calculated values from the fit to the values of Ref. 6 and is defined as:

\[
RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (z_{\text{cal},i} - z_{\text{Ref.6},i})^2}
\]  
(12)

Figure 1. Enthalpy - Concentration Diagram for saturated liquid of ammonia-water mixture at various temperatures.

Figure 2. Enthalpy-Concentration Diagram for saturated liquid of ammonia-water mixture at various pressures.

Figure 3. Enthalpy of saturated liquid of ammonia-water mixture at pressure 108.2 bar and temperature 177 [°C].

Figure 4. Dühring (PTX) diagram of ammonia-water mixture.
The number of data points, $n$, used for the equation fitting, together with the root mean square error are summarized in Table 6. These deviations enable one to estimate the accuracy of the equations. The precision of a particular set of data is evident from its band of deviations.

![Figure 5. Vapor - liquid equilibrium curves at 1, 20 and 60 bar.](image)

**Figure 5.** Vapor - liquid equilibrium curves at 1, 20 and 60 bar.

![Figure 6. Enthalpy-Concentration Diagram for saturated vapor of ammonia - water mixture at various pressures.](image)

**Figure 6.** Enthalpy-Concentration Diagram for saturated vapor of ammonia - water mixture at various pressures.

![Figure 7. Enlargement of region "A" in Figure 6.](image)

**Figure 7.** Enlargement of region "A" in Figure 6.

![Figure 8. Deviations between the calculated enthalpy of saturated liquid mixture from Equation 7 and those values from Ref. 6.](image)

**Figure 8.** Deviations between the calculated enthalpy of saturated liquid mixture from Equation 7 and those values from Ref. 6.

![Figure 9. Deviations between the calculated enthalpy of saturated liquid mixture from Equation 8 and those values from Ref. 6.](image)

**Figure 9.** Deviations between the calculated enthalpy of saturated liquid mixture from Equation 8 and those values from Ref. 6.

![Figure 10. Deviations between the calculated saturated pressure of mixture from Equation 9 and those values from Ref. 6.](image)

**Figure 10.** Deviations between the calculated saturated pressure of mixture from Equation 9 and those values from Ref. 6.

<table>
<thead>
<tr>
<th>Eqs.</th>
<th>n</th>
<th>RMSE</th>
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<tbody>
<tr>
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<td>349</td>
<td>3.44 kJ/kg</td>
</tr>
<tr>
<td>(8) for 0.2 &lt; P &lt; 100</td>
<td>132</td>
<td>11.75 kJ/kg</td>
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<td>(8) for 0.2 &lt; P &lt; 60</td>
<td>132</td>
<td>8.45 kJ/kg</td>
</tr>
<tr>
<td>(9)</td>
<td>396</td>
<td>0.991 bar</td>
</tr>
<tr>
<td>(10)</td>
<td>190</td>
<td>14.06 kJ/kg</td>
</tr>
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<td>(11)</td>
<td>190</td>
<td>0.0123 kg/kg</td>
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6. CONCLUSIONS

The fitted equations for calculation of the thermodynamic properties of the ammonia-water mixture are intended for use in the design of absorption refrigeration cycles. Using dependent variables of technical relevance, the equations make it possible to avoid iterative evaluations. The presented equations have a simple mathematical form, while also having relevant accuracy in comparison with other complicated correlations. These models can be implemented with easiness in all types of language and program that are required of the calculation of these properties. As well as avoiding iterative calculation, the merit of the fast approximation functions consists in their simplicity, which facilitates their incorporation in any computer code.

7. NOMENCLATURE

- \( a \) Molar Helmholtz free energy [kJ/kmol]
- \( a, b, c, d \) coefficient
- \( h \) Specific enthalpy of mixture [kJ/kg]
- \( n \) number of data points
- \( p \) pressure [bar]
- \( R \) gas constant [kJ/kg.K]
- \( \text{RMSE} \) Root Mean Square Error
- \( T \) temperature [°C]
- \( u, \nu \) independent variables
- \( x \) Mass fraction of ammonia in liquid phase
- \( y \) Mass fraction of ammonia in vapor phase
- \( z \) Thermodynamic property quantity
- \( \alpha, \beta \) Coefficient
- \( \zeta \) Mole fraction of ammonia in liquid phase
- \( \nu \) Molar volume [m³/kmol]

Subscripts:

- \( \text{cal} \) calculated value from present work
- \( G \) Saturated vapor
- \( L \) Saturated liquid
- \( r \) residual
- Ref. 6 Calculated value from Reference [6]
8. REFERENCES


