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Extended Verhoff-Banchero acid dew point model

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Abstract

This paper proposes a modification to the Verhoff-Banchero relation describing the acid dew point temperature, based on the evaluation of a generic equation that revealed two major limitations. First, the original relation's narrow application range was identified through comparison with the H_2O-H_2SO_4 vapour-liquid equilibrium dataset from the literature. Second, testing of the function's limits showed that the acid dew point temperature does not approach the water dew point temperature as the sulfuric acid concentration approaches zero. To address these limitations, the Verhoff-Banchero relation is extended into a bivariate quadratic function. Regression parameters, along with their estimation errors, are derived using the aforementioned dataset and the least squares method. Statistical test (p-value < 0.01) confirmed the significance of the introduced quadratic terms. The maximum estimation error for the modified relation is found to be ±10°C across a broad application range, with water partial pressures from 1013.25 Pa to 101325 Pa and sulfuric acid liquid mass fractions ranging from 0.1 to 0.98. Additionally, two semi-empirical relations are proposed, incorporating liquid-phase composition as an independent variable. The proposed extended Verhoff-Banchero model was evaluated using an experimental dataset for exhaust gas. Data points fall within an acceptable deviation range, with exceptions from two specific datasets. One of the observed deviations is explained by the partial derivative of the acid dew point temperature with respect to sulfuric acid partial pressure. The limitations of the proposed relations are discussed. Finally, the conclusions with potential future research directions in modelling the H_2O-H_2SO_4 system in exhaust gas are outlined.

Keywords: Acid dew point temperature; Vapour-liquid equilibrium; Sulfuric acid; Thermodynamic modelling

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1. Introduction

During the early stages of technological development aimed at meeting ecological and energy efficiency regulations (e.g. IMO (International Maritime Organization) Annex VI), there is a growing need to estimate exhaust gas acid dew point (ADP) temperature, especially at lower H_2SO_4 vapour pressures resulting from reduced sulfur concentrations in fuels.

The problem of ADP temperature calculation falls within the broader topic of H_2O-H_2SO_4 vapour-liquid equilibrium modelling, which is widely applied in various research areas. One key objective of ADP temperature estimation is to prevent low-temperature corrosion on heat transfer surfaces, a critical concern in sensible waste heat recovery systems, as discussed in [1,2]. This objective has evolved as new technologies operating below this limit, such as flue gas desulfurisation using the wet sulfuric acid method, studied by Nielsen et al. [3], latent waste heat recovery from exhaust gas [4] and exhaust gas recirculation (EGR) [5].

Many researchers focus on cylinder liner wear and tribology problems [6−8], where the partial pressures of water, sulfuric acid and sulfur trioxide are relatively high and influenced by engine cycle parameters.

Nomenclature

a – activity coefficient in the vapour phase
A – ratio of vaporisation entropy and universal gas constant
b – regression parameter in Verhoff-Banchero model
B – ratio of vaporisation enthalpy and universal gas constant
L – vaporisation enthalpy, J kmol^{-1}
p – partial pressure, Pa
P – saturation pressure, Pa
R – universal gas constant, J kmol^{-1} K^{-1}
S – vaporisation entropy, J kmol^{-1} K^{-1}
t – empirical acid dew point temperature, °C
T – absolute acid dew point temperature, K
w – mass fraction
x – mole fraction

Greek symbols

β, λ, ψ – regression parameters

Subscripts and Superscripts

1 – water
2 – sulfuric acid

Abbreviations and Acronyms

ADP – acid dew point
VB – Verhoff-Banchero
VLE – vapour-liquid equilibrium

One of the most commonly employed ADP models is the Verhoff-Banchero (VB) formulation [9] with regression parameters determined by Jia and Ling in [10]. This formulation is given by the formula:

$$\frac{1}{T} = b - b_1 \lg p_1 - b_2 \lg p_2 + b_{12} \lg p_1 \lg p_2, \tag{1}$$

where: p_1 – partial pressure of water, p_2 – partial pressure of sulfuric acid, $b = 2.9882 \times 10^{-3}$, $b_1 = 1.3761 \times 10^{-4}$, $b_2 = 2.6740 \times 10^{-4}$, $b_{12} = 3.2870 \times 10^{-5}$.

Another widely used empirical relation was published by Okkes and it can be found in [11]. Recently, the Okkes's regression parameters were recalculated by Tang et al. [12].

To evaluate these ADP models, a set of ADP temperatures and partial pressures of the constituents was interpolated using the tabulated data from Vermeulen et al. [13]. A simplified graphical representation of this dataset is shown in Fig. 1 and the deviations of the temperatures estimated using Eq. (1) and the equations given in [11] and [12] are presented in Table 1.

Two major drawbacks were observed from the results shown in Table 1:

- narrow application range of the tested models,
- ADP temperature does not approach the water dew point as the H_2SO_4 concentration tends to zero.

To address these limitations, a modification of the Verhoff-Banchero acid dew point relation is proposed, extending it into a bivariate quadratic function. Furthermore, two alternative semi-empirical relations are proposed, based on thermodynamic principles: the August relation derived from the Clausius-Clapeyron equation and a modified Raoult's law. The liquid-phase composition is added as an independent variable in these alternative relations due to its observed importance in previous studies.

The research objectives of this study are twofold:

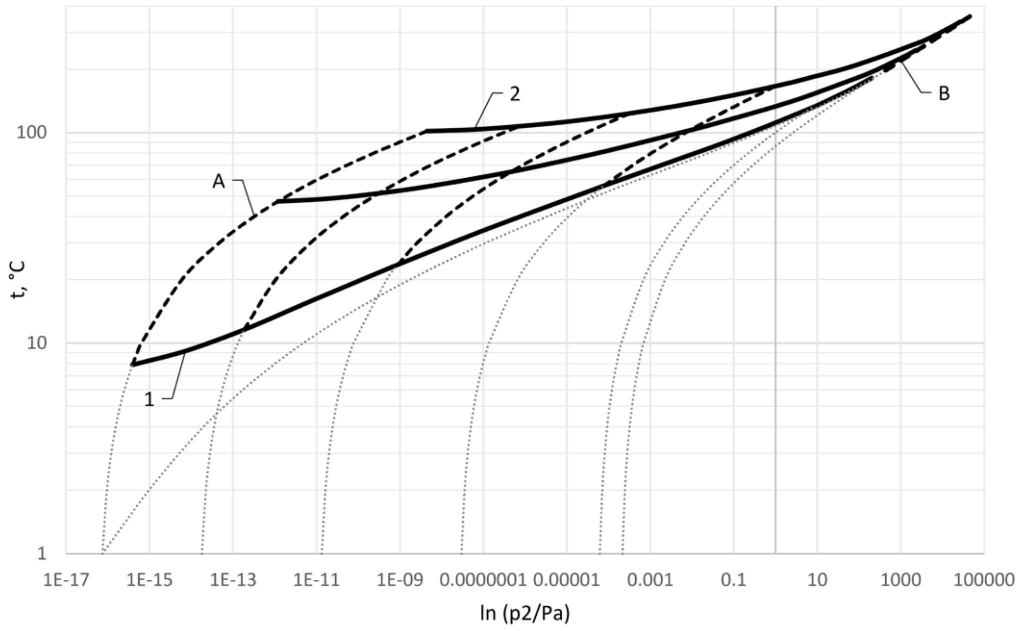

Fig. 1. Graphical representation of reference dataset interpolated from Vermeulen et al. [13]: 1, 2 – limitation curves computed at constant H_2O pressure equal to 1013.25 Pa and 101325 Pa, A-B – limitation curves computed at constant H_2SO_4 mass fraction equal to 0.1 and 0.98.

Table 1. Deviation of calculated acid dew point temperatures.

H_2SO_4 mass fraction	p_1 = 1013.25 Pa				p_1 = 10132.5 Pa				p_1 = 101325 Pa			
	ADP (°C)	Deviation Eq. (1)	Deviation Eq. from [11]	Deviation Eq. from [12]	ADP (°C)	Deviation Eq. (1)	Deviation Eq. from [11]	Deviation Eq. from [12]	ADP (°C)	Deviation Eq. (1)	Deviation Eq. from [11]	Deviation Eq. from [12]
0	7	[A]	[A]	[A]	46	[A]	[A]	[A]	100	[A]	[A]	[A]
0.1	8	88[B]	[A]	[C]	47	74[B]	[A]	[C]	102	58[B]	[A]	[C]
0.2	9	81[B]	[A]	[C]	49	65[B]	[A]	[C]	104	48[B]	[A]	[C]
0.3	12	73[B]	[A]	[C]	52	57[B]	[A]	[C]	107	39[B]	25[B]	[C]
0.4	16	64[B]	[A]	[C]	57	47[B]	[A]	[C]	113	30[B]	17[B]	–86[B]
0.5	24	53[B]	[A]	[C]	65	35[B]	12[B]	[C]	123	21[B]	10	–21[B]
0.6	36	40[B]	[A]	[C]	79	22[B]	3	–74[B]	140	11[B]	5	23[B]
0.7	56	23[B]	5	[C]	103	8	–1	6	166	5	6	37[B]
0.8	88	4	2	14[B]	137	–5	2	24[B]	204	5	18[B]	19[B]
0.9	132	21[B]	29[B]	47[B]	188	–4	24[B]	7	266	27[B]	53[B]	–10
0.92	142	–8	19[B]	19[B]	199	–2	30[B]	2	278	32[B]	61[B]	–14[B]
0.94	154	–7	25[B]	16[B]	214	3	39[B]	-2	296	42[B]	75[B]	–16[B]
0.96	171	–5	35[B]	13[B]	233	9	52[B]	-6	319	57[B]	93[B]	–17[B]
0.98	198	2	53[B]	10[B]	267	24[B]	77[B]	-7	364	86[B]	[C]	–13[B]

[A] Numerical solution does not exist.

[B] Deviation of ADP temperature exceeding the assumed application limit equal to ±10°C.

[C] Deviation of ADP temperature exceeding ±100°C omitted in the table.

1) To improve the accuracy of ADP temperature estimation across a broad range of H_2O and H_2SO_4 partial pressures by developing a model that meets these criteria.
2) To create ADP temperature relations that are reliable and can be employed in ongoing studies on low-temperature EGR cooling systems for dual-fuel marine engines installed on LNG carriers and LNG-fuelled ships.

The importance of the developed ADP model is significant, especially for marine dual-fuel engines, due to the low sulfur content in LNG and specifically in pilot fuel oil.

2. Materials and methods

The ternary system of H_2O-SO_3-H_2SO_4 in vapour-liquid equilibrium was studied, as this is a typical configuration of components in fuel combustion products formed in the oxidation of hydrogen and sulfur. In the acid dew point model, the focus was on the binary system of H_2O (1)-H_2SO_4 (2), considering that SO_3 has not been converted into sulfuric acid as one of the remaining constituents of dry exhaust gas.

To describe the relationship between the observed variable (ADP temperature) and the independent variables (partial pressures of H_2O and H_2SO_4, regression analysis using the least squares method was employed, along with two-sided interval estimation. These methods were used to determine the regression function parameters and their standard errors. A p-value test of the regression parameters was performed, with a significance level set below 0.01 to ensure statistical robustness.

For the estimation of the regression parameters of the H_2O-H_2SO_4 system, a dataset consisting of 576 observations was used. The regression parameters for each pure component were determined from separate datasets. Specifically, for pure sulfuric acid, 36 observations were taken from the dataset of Vermeulen et al. [13]. The water input dataset, consisting of 177 observations, was sourced from the work of Wagner and Pruß [14].

3. Acid dew point model

The generic Verhoff-Banchero acid dew point model, presented in Eq. (1), can be considered a purely empirical relation, represented as a bivariate quadratic regression function with quadratic terms initially set to zero.

In this study, the model was extended by adding quadratic terms to improve its accuracy. The extended model is given by

$$1/T = \beta - \beta_1 \ln p_1 - \beta_2 \ln p_2 + \beta_{12} \ln p_1 \ln p_2 - \beta_{11}(\ln p_1)^2 - \beta_{22}(\ln p_2)^2, \qquad (2)$$

where: p_1 – partial pressure of water, p_2 – partial pressure of sulfuric acid, with the coefficients $\beta = (2.776 \pm 0.01) \times 10^{-3}$, $\beta_1 = (1.150 \pm 0.17) \times 10^{-5}$, $\beta_2 = (9.500 \pm 0.15) \times 10^{-5}$, $\beta_{12} = (4.300 \pm 0.16) \times 10^{-6}$, $\beta_{11} = (2.930 \pm 0.16) \times 10^{-6}$, $\beta_{22} = (1.100 \pm 0.04) \times 10^{-6}$.

These quadratic terms are crucial for improving the model's performance, especially in regions of the parameter space where the relationship between the ADP temperature and partial pressures deviates from a simple linear approximation.

3.1. Alternative semi-empirical relations

An alternative acid dew point temperature relation was derived in the specific form for each component. Both presented formulas are based on the well-known August equation with a regression parameter newly introduced in this paper, denoted by ψ for water and λ for sulfuric acid.

For water, the following formula was derived

$$\frac{1}{T} = (A_1 - \ln p_1)\left[B_1 + R\left(\psi - \frac{1}{2}x_1\right)\right]^{-1}, \qquad (3)$$

where: A_1= 24.58 ± 0.046, B_1= 4902 ± 20, $\psi = \begin{cases} 0.456 \pm 0.01 \\ x_1/2 \end{cases}$.

The first condition of the empirical parameter ψ is applied in the concentration region $0.1 < x_1 \leq 0.9$, where interaction between components becomes significant. The second condition eliminates the last term from the denominator, which leads to the rearranged August equation and is valid for the H_2O mole fraction range $0.9 < x_1 \leq 1$.

A similar formula was derived for sulfuric acid:

$$\frac{1}{T} = (A_2 - \ln p_2)[B_2 + R(1 - \lambda w_2)]^{-1}, \tag{4}$$

where: $A_2 = 25.65 \pm 0.20$, $B_2 = 9204 \pm 80$, $\lambda = \begin{cases} 1.055 \pm 0.018 \\ 1.024 \pm 0.020 \end{cases}$. The first condition of the empirical parameter lambda should be applied in the region $0.1 \leq w_2 < 0.94$, while the second one for $0.94 \leq w_2 < 1$.

In Eqs. (3) and (4), the universal gas constant, equal to $R = 8314.5$ J kmol^{-1}K^{-1}, was used and considered free of error.

Alternative ADP temperature relation was derived by means of a binary vapour-liquid equilibrium condition having the form of modified Raoult's rule

$$p_i = a_i P_i, \tag{5}$$

where: p_i – i-th constituent partial pressure, a_i – activity coefficient of the i-th component in vapour phase, P_i – i-th pure component vapour pressure over saturated liquid.

Activity coefficient of constituents in the vapour phase was based on integrated vapour-liquid equilibrium (VLE) of Clausius-Clapeyron with respect to temperature

$$\ln a_i = -\frac{\Delta L_i}{R}\frac{1}{T}, \tag{6}$$

where ΔL_i is the difference between the vaporisation enthalpy of a given component as a pure constituent and constituent of the considered two-phase system. It is dependent on the composition and temperature. Here, averaged values over the considered temperature range were used.

Pure component vapour pressure over saturated liquid was described similarly:

$$\ln P_i = \frac{S_i}{R} - \frac{L_i}{R}\frac{1}{T}. \tag{7}$$

Using Eqs. (5)–(7), the following relation for the ith component was derived:

$$\frac{1}{T} = \left(\frac{S_i}{R} - \ln P_i\right)\left(\frac{L_i}{R} + \frac{\Delta L_i}{R}\right)^{-1}, \tag{8}$$

where the well-known notation was added: $A_i = S_i/R$ – ratio of the pure component vaporisation entropy and universal gas constant, $B_i = L_i/R$ – ratio of the pure component vaporisation enthalpy and universal gas constant.

In order to describe $\Delta L_i/R$, a specific one-parameter empirical relation was introduced for each component, as can be seen in Eqs. (3) and (4).

3.2. Error of estimation

The general regression function that was selected is suitable for the expression of Eqs. (2), (3) and (4):

$$Y = f(X, \theta) \pm \epsilon, \tag{9}$$

where: Y – dependent variable, X – set of independent variables, θ – regression parameters, ϵ – error of estimated value.

Employing the given standard error of regression parameters, one can calculate the estimation error of ADP temperature using a widely employed statistical methodology. This problem is omitted here and will be examined in the next paper.

4. Results and discussion

In Table 2, the deviations of acid dew point temperature calculated using the proposed Eqs. (2), (3) and (4) are presented. From the observed deviations, it is clear that the best fit to the curves presented in Fig. 1 is obtained for Eqs. (2) and (3). The deviations of Eq. (4) show good accuracy in the highest partial pressure of water.

Acceptable deviations for ten percent of sulfuric acid mass fraction are observed for all of the relations. This indicates that the calculated temperature tends to the water dew point as the sulfuric mass fraction tends to zero.

The superior feature of Eq. (4) is the possibility of water dew point calculation at zero mass fraction of sulfuric acid.

Table 2. Deviation of acid dew point temperature calculated using the proposed Eqs. (2), (3) and (4).

H_2SO_4 mass fraction	**p_1=1013.25 Pa**			**p_1=10132.5 Pa**			**p_1=101325 Pa**		
	Deviation Eq. (2)	**Deviation Eq. (3)**	**Deviation Eq. (4)**	**Deviation Eq. (2)**	**Deviation Eq. (3)**	**Deviation Eq. (4)**	**Deviation Eq. (2)**	**Deviation Eq. (3)**	**Deviation Eq. (4)**
0	A	3	A	A	**0**	A	A	−2	A
0.1	−6	3	9	−1	**1**	7	7	−1	4
0.2	−2	5	11 B	1	**3**	9	7	1	4
0.3	1	7	14 B	3	**6**	12 B	7	5	6
0.4	4	7	15 B	4	**5**	13 B	6	4	8
0.5	6	3	14 B	4	**1**	12 B	4	0	7
0.6	6	1	12 B	3	**−2**	8	1	−3	3
0.7	4	2	5	0	**0**	2	−4	−4	−4
0.8	−3	4	−4	−7	**0**	−6	−10	−6	−8
0.9	A	1	36 B	−9	**−3**	−2	−7	−7	0
0.92	−8	−2	1	−9	**−7**	1	−5	−12 B	5
0.94	−8	−4	−6	−6	**−9**	−5	0	−15 B	−2
0.96	−6	−5	−3	−3	**−10**	−1	7	−14 B	6
0.98	−1	2	3	7	**1**	7	A	3	17 B

A Numerical solution does not exist.

B Deviation of ADP temperature exceeding the assumed application limit equal to ±10°C.

4.1. Model evaluation with empirical data

In Fig. 2, the experimental data on the acid dew point in exhaust gas reported by selected authors at a partial pressure of water equal to 10132.5 Pa are presented. The solid curve is computed using the extended VB model. Significant deviation of data points marked with numbers 2 and 3 from the computed curve can be observed in the figure.

This discrepancy was identified as potentially stemming from measurement challenges reported in [16] and [17]. This measurement problem by means of a conductive dew point meter in the situation of sulfuric acid low concentration in exhaust gas is described by Vasanthakumar [18].

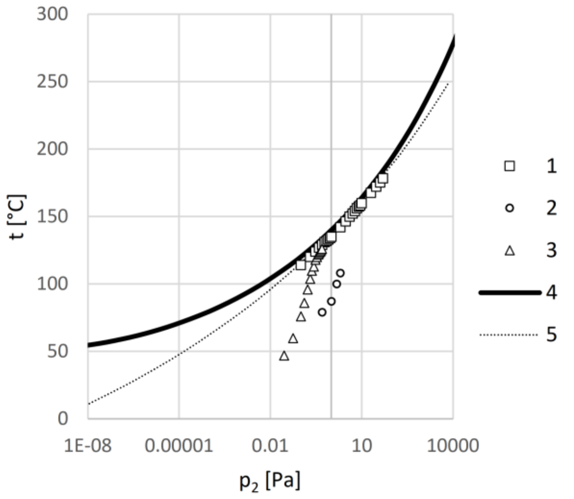

Fig. 2. Comparison of experimental data with evaluated acid dew point model: 1 – ZareNezhad et al. [15], 2 – Xiang et al. [16], 3 – Blanco et al. [17], 4 – extended VB (Eq. (2)), 5 – VB (Eq. (1)).

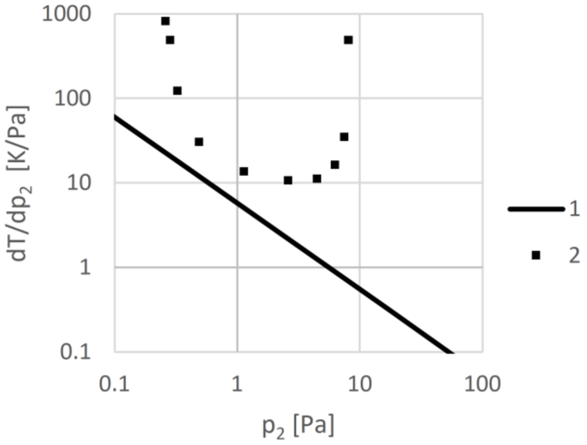

Fig. 3. Derivative of acid dew point temperature with respect to H_2SO_4 partial pressure: 1 – extended Verhoff-Banchero model (Eq.(2)), 2 – Li et al. [19].

In Fig. 3, the derivative of ADP temperature with respect to sulfuric acid partial pressure is displayed. The points were calculated using data published in [19] and the black solid curve was computed based on the modified VB model (Eq. (2)). From the graph, we can conclude that during the experiment reported in [19], the condensation did not proceed according to the equilibrium curve that represents a reversible process.

4.2. Model limitation

Despite satisfactory estimation accuracy of acid dew point temperature, the calculated partial pressure from rearranged Eq. (2) exhibits excessive deviation from the data points of Vermeulen et al. [13], which were taken as an input to build the model. Moreover, the behaviour of the rearranged function indicates numerical instability. Thus, it is advised to use the extended VB equation only for ADP temperature estimation. The problem of estimation error of partial pressure in the considered system was studied and described in the published note by Verhoff and Banchero [20], thus, it is omitted here.

Equation (2) is the recommended primary model. The limitations of the derived Eqs. (3) and (4) require further validation.

5. Conclusions

This study presents a significant advancement in the modelling of the acid dew point temperature by extending the Verhoff-Banchero equation to include quadratic terms. The proposed modification enhances the accuracy of ADP temperature estimations, particularly in the range of sulfuric acid mass fractions where previous models showed limitations. The introduction of semi-empirical relations for water and sulfuric acid provides further flexibility and applicability across a wider range of operating conditions.

The primary contribution of this research lies in the improved understanding of ADP temperature behaviour, especially in systems with low sulfuric acid concentrations. The extended Verhoff-Banchero model addresses the issue of ADP temperature approaching the water dew point as sulfuric acid concentration decreases.

While the extended model demonstrates improved accuracy for ADP temperature estimation, challenges remain in the numerical instability and deviations in partial pressure calculations.

The results of this work contribute to the development of more reliable models for the H_2O-H_2SO_4 system, with potential applications in environmental engineering, energy recovery and industrial processes that involve sulfuric acid and water vapour.

The VLE (vapour-liquid equilibrium) modelling of the H_2O-H_2SO_4 system in exhaust gases offers several avenues for future research. This study has focused on ADP temperature as a function of partial pressures and liquid phase composition. Future work should investigate the following relations with two independent variables:

- liquid phase composition as a function of ADP temperature and vapour phase composition,
- vapour phase composition as a function of ADP temperature and liquid phase composition.

By developing these relations and incorporating the impact of condensation nuclei on mist formation, a more realistic H_2O-H_2SO_4 condensation model in exhaust gas can be created for practical applications.

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