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# Comparison of Models for Calculation of the Thermodynamic Properties of NH<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O Mixture

Vilborg Gudjonsdottir

*Delft University of Technology, Section Engineering Thermodynamics, Netherlands, v.gudjonsdottir-1@tudelft.nl*

Carlos Infante Ferreira

*Delft University of Technology, Section Engineering Thermodynamics, Netherlands, C.A.InfanteFerreira@tudelft.nl*

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# Comparison of Models For Calculation of the Thermodynamic Properties of NH<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O Mixture

Vilborg GUDJONSDOTTIR<sup>1\*</sup>, Carlos A. INFANTE FERREIRA<sup>1</sup>

<sup>1</sup> Delft University of Technology, Section Engineering Thermodynamics,  
Leeghwaterstraat 39, 2628 CB Delft, The Netherlands,  
V.Gudjonsdottir@tudelft.nl

\* Corresponding Author

## ABSTRACT

In the past several attempts have been made to fit experimental data of the mixture NH<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O making use of thermodynamic models. The most commonly used model for process calculations is the electrolyte NRTL model. The model is available in the Aspen Plus software (version 8.8) both in its original form as well as modified versions where parameters have been fitted especially for the NH<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O system. In this paper these models and a new fit are compared to the Extended UNIQUAC model. Comparison of the models shows a clear improvement of the modified models over the original e-NRTL model. The better fitted models compare well to the extended UNIQUAC model which gives good results for its application range.

## 1. INTRODUCTION

The NH<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O mixture has been identified as a possible working fluid for compression-resorption heat pumps (CRHP). The advantages of CRHP for low grade waste heat recovery compared to alternative technologies have been reported by Van de Bor et al. (2015). In the research the working fluid for the CRHP was ammonia water. Preliminary calculations suggest that adding small amount of CO<sub>2</sub> to the system can increase the coefficient of performance (COP) of the heat pump. To further investigate the performance of the CRHP with NH<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O mixture as a working fluid an accurate thermodynamic model is needed.

Different thermodynamic models have been used and developed for calculating the thermodynamic properties of NH<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O. These models are normally activity coefficient models for the liquid phase and an equation of state (EOS) for the vapor phase calculations. The activity coefficient models that have been most commonly used are electrolyte models such as: the Pitzer model (Kurz et al., 1995), the extended UNIQUAC model originally developed by Thomsen and Rasmussen (1999) and the more commonly used e-NRTL model proposed by Chen et al. (1982).

Que and Chen (2011) deemed the e-NRTL model the most suitable for process modeling and simulations since it requires only binary interaction parameters and makes use of mole fraction concentration scale consistently for both the short range local composition interactions and the long range Debye-Huckel expression. Darde et al. (2012) compared a built in e-NRTL model from Aspen Plus to an upgraded version of the extended UNIQUAC model described by Darde et al. (2010). Their findings were that the extended UNIQUAC model generally performed better than the e-NRTL model from Aspen especially for the partial pressure of NH<sub>3</sub> and the solubility of ammonium bicarbonate. Darde (2011) mentions that if the binary interaction parameters were better fitted to experimental data for NH<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O mixture, the e-NRTL model might become more competitive with the extended UNIQUAC model. Since then the e-NRTL model has been modified in this way by a couple of authors, including Que and Chen (2011) and Niu et al. (2013). Both of their adjusted models have then been used by other authors for process modeling, for example Zhang and Guo (2014) used the model with adjusted parameters from Niu et al. (2013) and Liu et al. (2015) used the

modified model from Que and Chen (2011). A modified Pitzer activity coefficient model has been developed by Xu et al. (2014) which needs significantly fewer parameters to be fitted than the traditional models and still is reported to be accurate over wide range of temperatures and concentrations. The authors state that further tests of the model will be reported in the future. The model will therefore not be further discussed in this paper however it seems to show great potential.

The extended UNIQUAC model has previously not been compared to the modified model from Que and Chen (2011) over a large range of operating conditions. Therefore these models are compared together to see if a modified e-NRTL model can perform with similar accuracy as the extended UNIQUAC model. The e-NRTL models that are built into Aspen are used as a reference. Additionally a new fit of the e-NRTL model is developed and compared with the others.

## 2. MODELS

In the subsections below the extended UNIQUAC and the e-NRTL models are described in more detail.

### 2.1 Extended UNIQUAC model

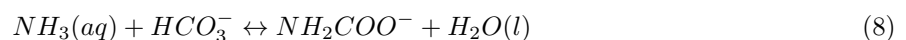
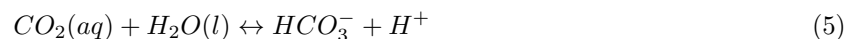
The Extended UNIQUAC model was developed by Thomsen and Rasmussen (1999). The model uses the extended UNIQUAC model to calculate activity coefficients for the liquid phase and the Soave-Redlich-Kwong (SRK) EOS for vapor phase calculations. The model was further developed and described by Darde et al. (2010) and implemented as a Fortran subroutine in Aspen Plus by Maribo-Mogensen (2014). The original model describes accurately the thermodynamic properties of the  $\text{NH}_3\text{-CO}_2\text{-H}_2\text{O}$  mixture for ammonia concentrations up to 80 molal  $\text{NH}_3$ , temperature of 0-110 °C and pressure up to 10 MPa. The newer version of the model describes the thermodynamic properties accurately up to 150 °C. Additionally the model parameters have been fitted to more experimental data to increase accuracy. The extrapolation of equilibrium constants into the supercritical range was also improved using Henry's law instead of the Gibbs-Helmholtz equation. Darde (2011) describes the mathematical model in detail.

The chemical equilibria that are taken into account in the model are stated below

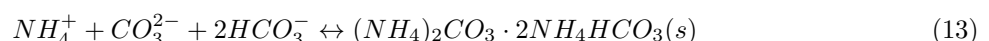
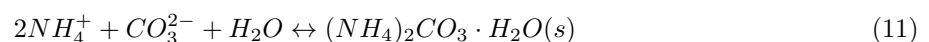
Vapor-liquid equilibrium



Speciation equilibrium



Liquid-solid equilibrium



**Table 1:** Experimental data for regression of the NH<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O system

Data Type	T, K	wt% NH <sub>4</sub> HCO <sub>3</sub>				Source
SLE	273 - 363	2.4 - 14.4				Janecke (1929)
Data Type	T, K	mol% NH <sub>3</sub>	mol% CO <sub>2</sub>	P (MPa)	Deviation (%)	Source
VLE	393.15	1.2 - 17.7	0.3 - 9.9	0.3 - 5	6.6	Göppert and Maurer (1988)
VLE	393.15	4 - 18	0.4 - 7.6	0.1 - 1.3	7.1	Müller et al. (1988)

## 2.2 E-NRTL model

The e-NRTL model is built into the Aspen Plus software (Aspen Physical Property System, 2015). In this study the version 8.8 of Aspen Plus is used. A data package for NH<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O mixture using the e-NRTL method and the Redlich-Kwong (RK) EOS for the vapor phase is included in the software. This model will be called e-NRTL1 from here on. A modified version of the model and the one that Darde et al. (2012) used for their comparison is included in a carbon capture example (Aspen Physical Property System, 2011) also included in the Aspen Plus v. 8.0 software. Additionally this model has been regressed to VLE, SLE, speciation and heat capacity data, this model will be called e-NRTL2.

The thermodynamic model proposed by Que and Chen (2011) is included in another carbon capture example available in the Aspen Plus software (Aspen Physical Property System, 2012). The main difference between that model and the e-NRTL2 model is that the PC-SAFT (Perturbed Chain Statistical Association Fluid Theory) EOS is used for vapor phase calculations instead of the RK EOS. As mentioned in the introduction the e-NRTL model has also been modified by other authors like Niu et al. (2014). The model modified by Que and Chen (2011) was however chosen since more experimental data are used for data regression of the model parameters. The model by Que and Chen (2011) is reported to be accurate for systems with temperatures up to 473 K, pressures up to 7 MPa, NH<sub>3</sub> concentration up to 30 wt%, and CO<sub>2</sub> loading up to unity. Que and Chen (2011) also give a good overview of the mathematical model.

In the e-NRTL model only the formation of ammonium bicarbonate ( $NH_4HCO_3$ ) is considered for SLE and not ammonium carbonate ( $(NH_4)_2CO_3 \cdot H_2O$ ), ammonium carbamate ( $NH_2COONH_4$ ) and ammonium sesqui-carbonate ( $(NH_4)_2CO_3 \cdot 2NH_4HCO_3$ ). Researches have shown however, that ammonium bicarbonate is dominant in the total amount of ammonium salts once the CO<sub>2</sub> absorption reaches steady state (Kim et al., 2008; Park et al., 2008). Therefore the e-NRTL model might still be a good option.

These versions of the e-NRTL models are compared to the extended UNIQUAC model as well as a new fit in the following chapter. In the next chapter it is demonstrated that the model developed by Que and Chen shows satisfactory results except for SLE at temperatures above 50 °C. The new fit is therefore based on the model from Que and Chen (2011) except the NRTL model binary interaction parameters  $\tau_{1,ij}$  (see equation 14), associated with the major species of the electrolyte, is refitted to additional SLE ternary NH<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O experimental data. During the fitting the initial values were kept as the same ones developed by Que and Chen (2011). Additionally the parameters were fitted at the same time to VLE data at 393.15 K. This was done in order to keep the accuracy of the VLE equilibrium. Fitting to additional VLE data at lower temperatures did not further increase the accuracy of the fit. As explained by Que and Chen (2011) the NRTL model requires a non randomness factor  $\alpha_{ij}$  and asymmetric binary interaction energy parameters  $\tau_{ij}$  calculated with the following equation

$$\tau_{ij} = \tau_{1,ij} + \frac{\tau_{2,ij}}{T} \quad (14)$$

Where i and j stand for the components, either ionic species, water, ammonia or carbon dioxide. An overview of the experimental data is listed in Table 1 and the refitted parameters are listed in table 2.

**Table 2:** Adjusted NRTL binary interaction parameters

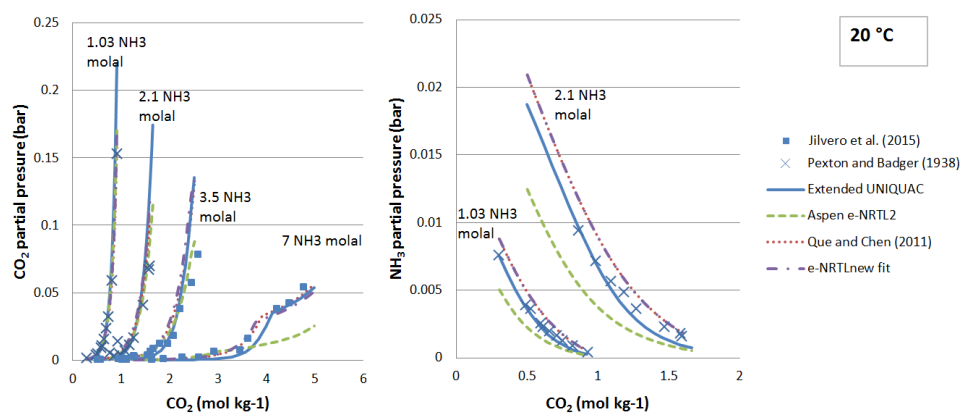
Component i	Component j	$\tau_{1,ij}$
H <sub>2</sub> O	(NH <sub>4</sub> <sup>+</sup> , HCO <sub>3</sub> <sup>-</sup> )	-4.18753
H <sub>2</sub> O	(NH <sub>4</sub> <sup>+</sup> , CO <sub>3</sub> <sup>-2</sup> )	3.46678
(NH <sub>4</sub> <sup>+</sup> , CO <sub>3</sub> <sup>-2</sup> )	H <sub>2</sub> O	-2.6869
H <sub>2</sub> O	(NH <sub>4</sub> <sup>+</sup> , NH <sub>2</sub> COO <sup>-</sup> )	9.542
(NH <sub>4</sub> <sup>+</sup> , NH <sub>2</sub> COO <sup>-</sup> )	H <sub>2</sub> O	-4.3115
NH <sub>3</sub>	(NH <sub>4</sub> <sup>+</sup> , NH <sub>2</sub> COO <sup>-</sup> )	7.45449
(NH <sub>4</sub> <sup>+</sup> , NH <sub>2</sub> COO <sup>-</sup> )	NH <sub>3</sub>	-4.82636

### 3. MODELS COMPARISON

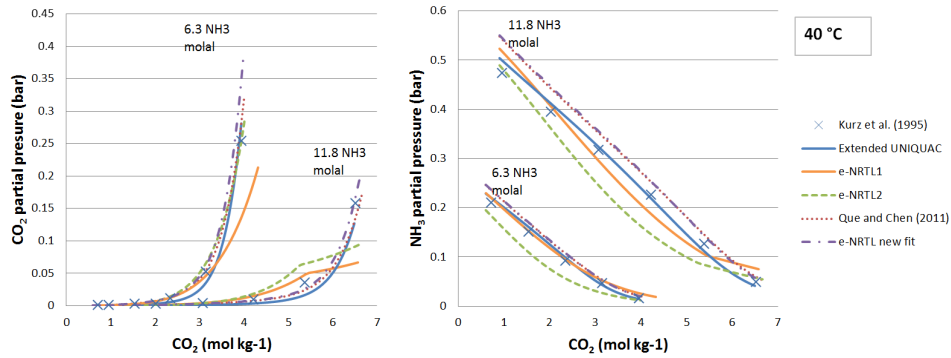
The chosen model should be able to accurately describe the vapor-liquid equilibrium (VLE), the solid-liquid equilibrium (SLE), speciation and enthalpy change over a large range of temperatures and concentrations of NH<sub>3</sub> and CO<sub>2</sub>. Comparison of the models mentioned in the previous chapter are discussed in the following sections.

#### 3.1 VLE

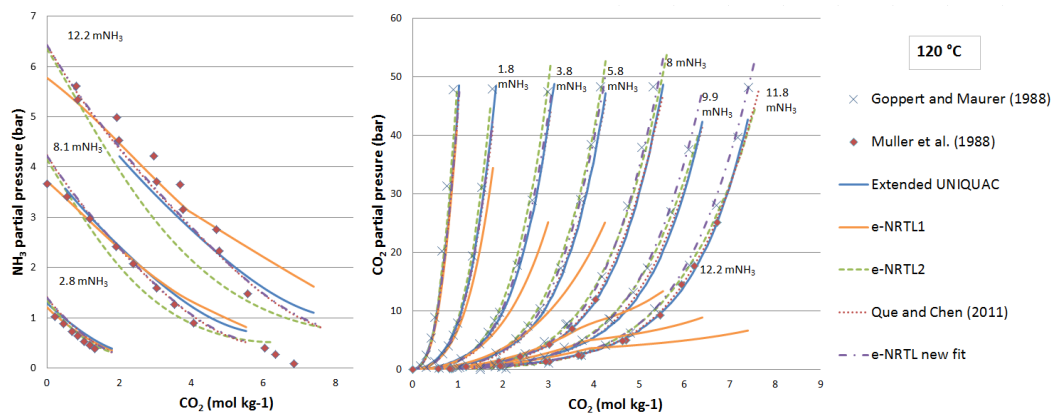
In figures 1 - 4 the partial bubble point pressures of CO<sub>2</sub> and NH<sub>3</sub> versus the molality of CO<sub>2</sub> are compared for different temperatures. The e-NRTL1 model is generally inaccurate at high temperatures and high loadings, as previously reported by Darde (2011). The e-NRTL2 model is in most cases an improvement from the e-NRTL1 model however it generally underestimates the partial bubble point pressure of NH<sub>3</sub> as well as inaccurately portray the CO<sub>2</sub> pressure at high loadings at 20 and 40 °C. The model by Que and Chen (2011), the new fit and the extended UNIQUAC model accurately portray the partial pressures. Jilvero et al. (2015) even reported that the model by Que and Chen (2011) fit their experimental data of CO<sub>2</sub> partial bubble point pressures, for 10- 40 °C, even more accurately than the Extended UNIQUAC model. The only exception is for the partial bubble point pressure of CO<sub>2</sub> at 150 °C. However since all the models deviate from the experimental data from Pawlikowski et al. (1982) in that case it is considered to be rather the data it self than the models that is inaccurate.



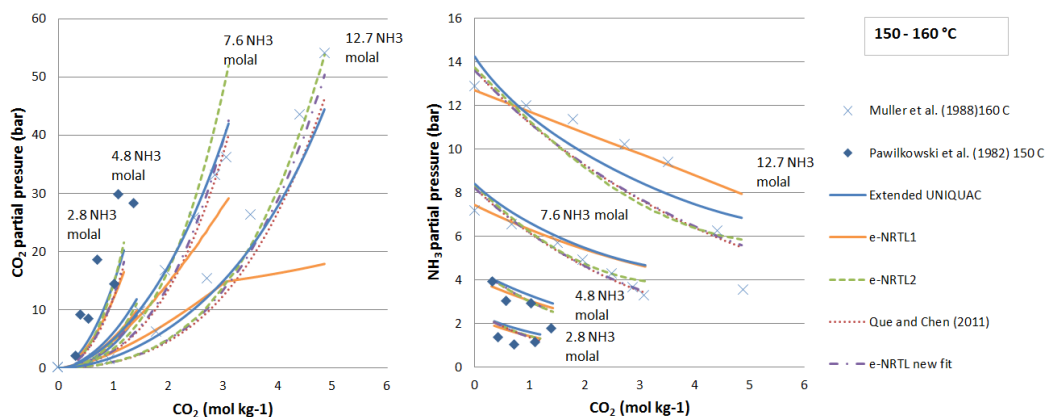
**Figure 1:** Comparison of the experimental data from Pexton and Badger (1938), and Jilvero et al. (2015) for partial bubble point pressures of CO<sub>2</sub> and NH<sub>3</sub> at 20 °C and the model correlations.



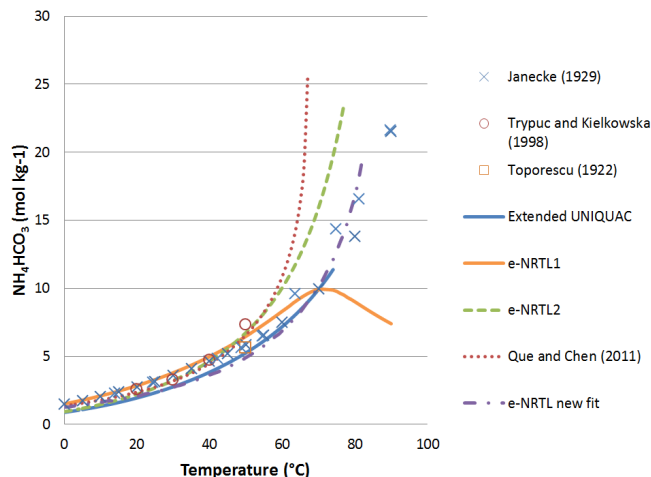
**Figure 2:** Comparison of the experimental data from Kurz et al. (1995) for partial bubble point pressure of  $\text{CO}_2$  and  $\text{NH}_3$  at  $40^\circ\text{C}$  and the model correlations.



**Figure 3:** Comparison of the experimental data from Göppert and Maurer (1988) and Müller et al. (1988) for partial bubble point pressure of  $\text{NH}_3$  and  $\text{CO}_2$  at  $120^\circ\text{C}$  and the model correlations.



**Figure 4:** Comparison of the experimental data from Müller et al. (1988) and Pawlikowski et al. (1982) for partial bubble point pressure of  $\text{NH}_3$  and  $\text{CO}_2$  at  $150$  and  $160^\circ\text{C}$  and the model correlations.



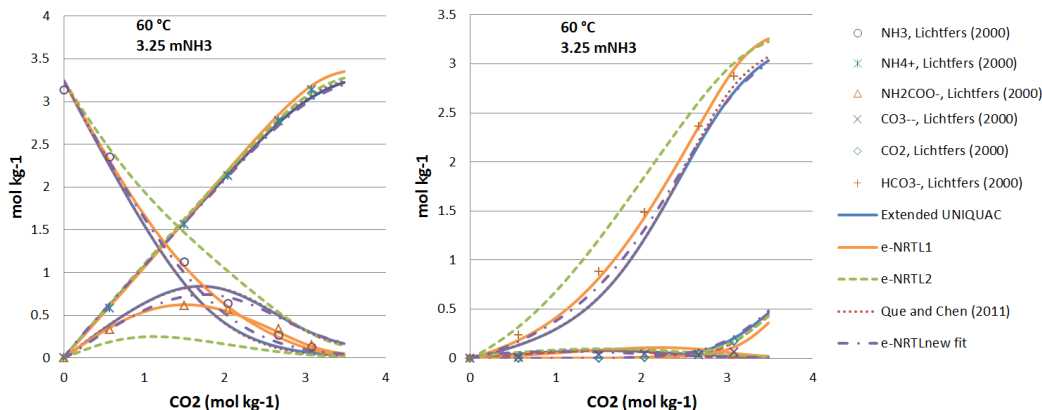
**Figure 5:** Comparison of the experimental data from Janecke (1929), Trypuc and Kielkowska (1998) and Toporescu (1922) for solubility of  $\text{NH}_4\text{HCO}_3$  in water and the model predictions.

### 3.2 SLE

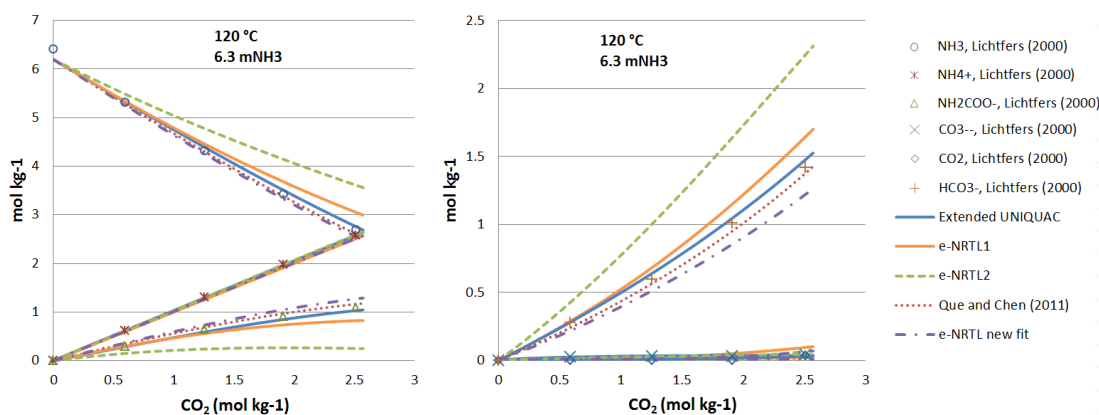
The comparison of the models for solubility of ammonium bicarbonate ( $\text{NH}_4\text{HCO}_3$ ) in water versus temperature is shown in figure 5. The models are compared to experimental data from Janecke (1929), Trypuc and Kielkowska (1998) and Toporescu (1922). Assuming that the experimental data from Janecke (1929) is accurate, the extended UNIQUAC model as well as the new fit are the most accurate at high temperatures or above approximately 50 °C. At higher temperatures the e-NRTL2 and the model proposed by Que and Chen (2011) overestimate the solubility of  $\text{NH}_4\text{HCO}_3$  while the e-NRTL1 model underestimates the solubility. In the case of e-NRTL2 and the model from Que and Chen (2011) the reason for this difference can be easily explained since the experimental data used for the regression for both models was the one from Trypuc and Kielkowska (1988). The experimental data from them reaches to temperatures of 50 °C. Also their value at 50 °C is slightly higher than the one from Janecke (1929) and Toporescu (1922). Since more experimental data at high temperatures was not found in literature it is questionable which of the data sets correspond best to reality. Additionally in practice for the CRHP application the concentration of  $\text{NH}_3$  and  $\text{CO}_2$  is unlikely to come close to the concentration necessary for salt formation at high temperatures. For example the reported concentration by Janecke et al. (1929) of  $\text{CO}_2$  at approximately 60°C is around and above 30 wt% (depending on the  $\text{NH}_3$  concentration). The data from Trypuc and Kielkowska (1998) suggest that this limit might be even higher and therefore either model should give satisfying results if the application concentration does not reach this limit.

### 3.3 Speciation

Comparison of speciation calculations of the models and experimental data from Lichtfers (2000) is shown in figures 6 and 7. All the models are able to quite accurately describe the speciation at both temperatures (60 and 120 °C, respectively) except the e-NRTL2 model. The e-NRTL2 model overestimates the concentration of ammonia and bicarbonate and underestimates the carbamate concentrations. In the two previous subsections the e-NRTL2 model was in general an improvement of the original model (e-NRTL1). This shows the importance of using a wide range of experimental data for parameter fitting for the  $\text{NH}_3\text{-CO}_2\text{-H}_2\text{O}$  system.



**Figure 6:** Comparison of the experimental data from Lichtfers (2000) for speciation at 60 °C and molality of 3.25 mol/kg NH<sub>3</sub> and the model correlations.

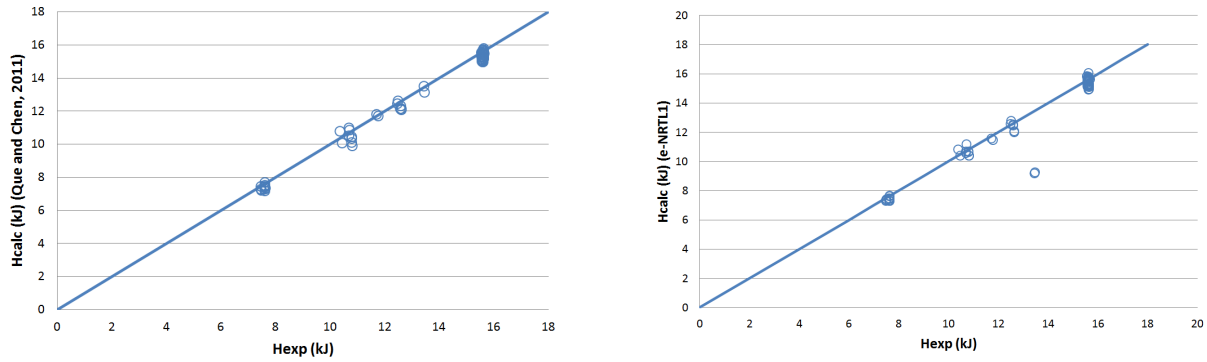


**Figure 7:** Comparison of the experimental data from Lichtfers (2000) for speciation at 120 °C and molality of 6.3 mol/kg NH<sub>3</sub> and the model correlations.

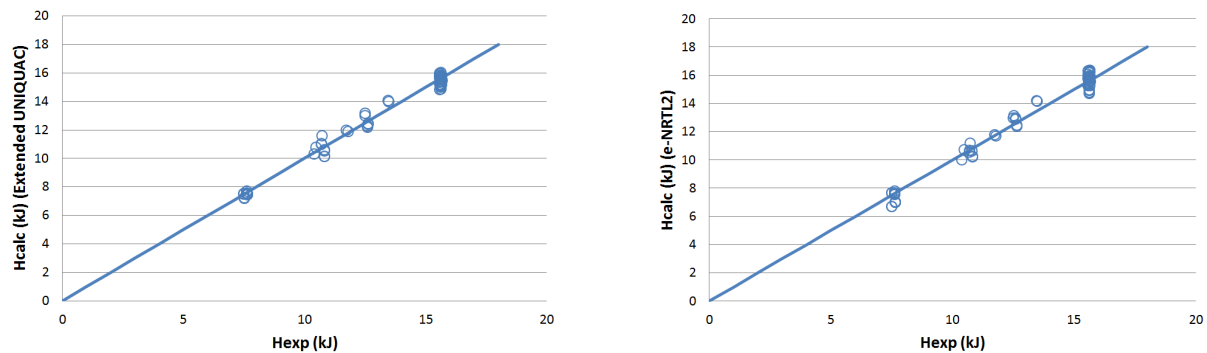
### 3.4 Enthalpy change

In figures 8, 9 and 10 the models are compared to experimental data from Rumpf et al. (1998) for enthalpy change upon partial evaporation of the NH<sub>3</sub>-CO<sub>2</sub>-H<sub>2</sub>O mixture. The temperature range of the experiments was from 40 to 137 °C with a typical temperature increase of 5-15 °C. The concentration range for NH<sub>3</sub> was up to 12 molal and up to 6 molal for CO<sub>2</sub>. The reported temperatures and pressures are used to calculate the inlet enthalpy. At the outlet however the reported vapor fraction is used instead of the pressure since the accuracy of the weight of the liquid and vapor part is higher than that of the measured pressure. All correlations show good matches to the experimental data with the only exception of two points for the e-NRTL1 correlation. These two points were at the highest reported temperature and CO<sub>2</sub> loading. This deviation corresponds to the previous shown results of VLE and SLE data.

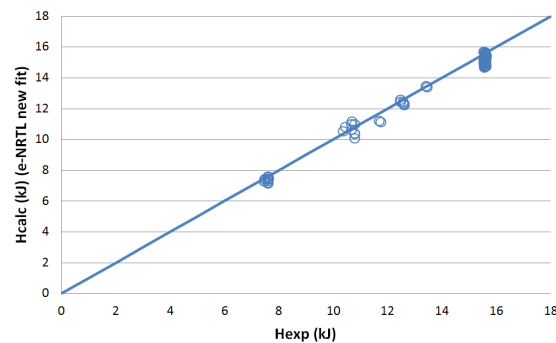




**Figure 8:** Comparison of the experimental data from Rumpf et al. (1998) for heat of partial evaporation and left the Que and Chen (2011) and right e-NRTL1 correlations.



**Figure 9:** Comparison of the experimental data from Rumpf et al. (1998) for heat of partial evaporation and left the extended UNIQUAC and right e-NRTL2 correlations.



**Figure 10:** Comparison of the experimental data from Rumpf et al. (1998) for heat of partial evaporation and the e-NRTL new fit correlation.

#### 4. CONCLUSIONS

From the model comparison it is clear that the modified e-NRTL models are in general an improvement of the original model. The model modified by Que and Chen (2011) and the new fit are especially compatible with the Extended UNIQUAC model. The model developed by Que and Chen (2011) is even more accurate

for the partial pressure of CO<sub>2</sub> at low temperatures (10-40 °C) as reported by Jilvero et al. (2015). Their model also improves the partial pressure of NH<sub>3</sub> and speciation compared to the e-NRTL2 model. The only exception is the SLE. The new fit that was developed solves this problem and the new model is able to represent the experimental data satisfactorily. The computational time with the model by Que and Chen (2011) and the new fit is also significantly better than when the extended UNIQUAC model is applied, which tended to freeze regularly during the calculations. On the same computer the e-NRTL models did not have that problem likely due to less complexity. Overall the modified e-NRTL model by Que and Chen (2011) and the new fit based on their model can be recommended for its application range. However the desired NH<sub>3</sub> concentration for the CRHP case is in some cases higher than the 30 wt% limit of the Que and Chen (2011) model (depending on the application case). The limit of the original extended UNIQUAC model was reported by Darde (2011) as 80 molal NH<sub>3</sub>. The newer model is however refitted with data that does not come close to that limit. Since limited data is available with NH<sub>3</sub> concentrations over 30 wt% further experiments should be performed to check the accuracy of the models at a higher range.

## NOMENCLATURE

$\alpha$	Nonrandomness factor		<b>Abbreviations</b>
$aq$	aqueous		COP      Coefficient of Performance
$g$	gas		CRHP     Compression-resorption heat pump
$l$	liquid		e-NRTL    Electrolyte Non Random Two Liquid
$m$	molality		EOS       Equation of State
mol%	mole percent		PC-SAFT   Perturbed Chain Statistical
$P$	pressure	bar	Association Fluid Theory
$T$	temperature	K	RK        Redlich-Kwong
$\tau$	Asymmetric binary interaction energy parameter		SLE       Solid-liquid equilibrium
			SRK       Soave-Redlich-Kwong
wt%	weight percent		UNIQUAC   Universal Quasi Chemical
<b>Subscript</b>			VLE       Vapor-liquid equilibrium
$i, j$	component		

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