

Investigating the Solubility of CO₂ in the Solution of Aqueous K₂CO₃ Using Wilson-NRF Model

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Abstract

Hot potassium carbonate (PC) solution in comparison with amine solution had a decreased energy of regeneration and a high chemical solubility of CO₂. To present vapor and liquid equation (VLE) of this system and predict CO₂ solubility, the ion specific non-electrolyte Wilson-NRF local composition model (isNWN) was used in this study; the framework of this model was molecular. Therefore, it was suitable for both electrolyte and non-electrolyte solutions. The present research employed the NWN model and the Pitzer-Debye-Hückel theory in order to assess the contribution of the excess Gibbs energy of electrolyte solutions in a short and long range. The data of CO₂ solubility in water and the system of aqueous K₂CO₃ were correlated in the model considering a temperature range of 10 – 170 °C and a pressure range of 0.5 – 141.1 bar. The average absolute error of (CO₂ – H₂O) and (CO₂ – H₂O – K₂CO₃) systems were 4.2% and 11.41% respectively. The results and comparisons with other models proved that the experimental data were exactly correlated in the model.

Keywords: CO₂ Capture, Solubility, Non-electrolyte Wilson-NRF, Thermodynamic Modeling, Potassium Carbonate, Ion-specific Parameters

1. Introduction

The global climate is changing due to the continuous increase in the temperature of the earth, which is of great concern for the environment. The major reason for the global warming is the increasing production of CO₂ more than the nature can accept. CO₂ is increasingly released into the atmosphere through using fossil fuels and industrial processes (Višković et al., 2014). There are different methods available for the elimination of CO₂ from flue gases of power plants and gas streams. CO₂ capture by using chemical absorption is a mature technology currently used (Zhenqi et al., 2012; Meihong et al., 2011; Zhao et al., 2012). Utilization of amine solution is the most popular method for chemical absorption (Afkhamipour et al., 2013; Austgen et al., 1991; Dang et al., 2003; Sidi-Boumedine et al., 2004). However, amines have some disadvantages; for instance, these solutions have shown excessive energy requirement for regeneration, easy degradation, and strong corrosion to equipment. Appropriate absorbers must have a fast reaction rate and a lower heat of regeneration. Hot potassium carbonate solution has a decreased energy of regeneration (Kohl et al., 1997) and a high chemical solubility of

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CO₂ (Todinca et al., 2007). An equilibrium thermodynamic model is required to anticipate the total pressure of the solution as well as the partial pressure of CO₂. Similar systems can be thermodynamically modeled through many available models to achieve acceptable simulation and optimization for a chemisorption process (Afkhamipour et al., 2013). These models can be categorized into three groups: 1) a state equation through phi-phi method as reported by Gubbins and Button (Button et al., 1999) and Huttenhuis et al. (Huttenhuis et al., 2008); 2) semi-empirical models such as the Kent-Eisenberg model (Kent et al., 1976) 3) excess Gibbs energy or gamma-phi method such as the works of Austgen et al. (Austgen et al., 1991) and Li and Mather (Li et al., 1998; Qian et al., 1995). This research has investigated the free energy of surplus Gibbs to demonstrate the systems VLE (CO₂ – H₂O) and (CO₂ – H₂O – K₂CO₃) through the thermodynamic modeling. In these systems, the local composition statement was used regarding the short-range contribution, and one of the Debye-Hückel (DH) equations was employed for long range electrostatic interactions (Zhao et al., 2000; Messnaoui et al., 2008). Haghtalab and Mazloumi (Haghtalab et al., 2009) applied Wilson-NRF model to electrolyte solutions, which were considered as an ion-pair. The model had a salt specific parameter as well as two adjustable parameters per each salt. However, Mazloumi (Mazloumi et al., 2015) had used this model using ion specific parameters for strong aqueous binary and ternary electrolytes. The results were highly correlated with the experimental data. The Wilson-NRF model was utilized as the thermodynamic model for non-electrolytes in the current work as presented in the study of Mazloumi (Mazloumi et al., 2015).

2. Thermodynamic framework

The CO₂ solubility and the other solute species quantity may be predicted by the thermodynamic modeling (Anderko et al., 2002). A few equations must be considered to model a system like potassium carbonate solution thermodynamically, and a thermodynamic approach should be utilized for the VLE calculations.

2.1. Standard state

The equilibrium of the system was defined through chemical potential. In this condition, each component of all the phases should have equal chemical potential values. These values of solvent (water) in an aqueous system can be defined by a mole fraction-symmetrical equation (Afkhamipour et al., 2013):

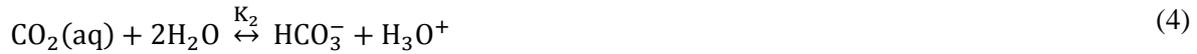
$$\mu_w = \mu_w^0 + RT \ln(\gamma_w x_w) \quad (1)$$

In which, μ_w^0 is the standard chemical potential of pure water. γ_w also represents symmetrical water activation coefficient, and x_w is the water mole fraction; T stands for absolute temperature, and R is the universal gas constant. The chemical potential values of the solute in the system is calculated by the following equation:

$$\mu_i = \mu_i^* + RT \ln(\gamma_i^* x_i) \quad (2)$$

2.2. Chemical equilibrium

The chemical reactions of the liquid phase must be taken into consideration for speciation calculations. The ionic complex species are produced when CO₂ reacts with PC in an aqueous solution. The subsequent reactions are given by:



The stoichiometric technique was utilized in this study to create the calculation. This technique was correctly investigated by several researchers (Al-Rashed et al., 2012; Cullinane et al., 2002).

Equation 6 calculates the chemical equilibrium constant, which is solved for each reaction mole fraction and for the balance of mass and charge equations (Al-Rashed et al., 2012).

$$K_j = \prod_{i=1}^n \beta_i^{\vartheta_i} \quad (6)$$

where, K_j is the constant of equilibrium in each chemical reaction j at a standard state ($T_0 = 298.15$ K), which was stated as the experimental function of the temperature in most of researches (Equation 6). Also, ϑ_i is the reaction stoichiometry of i species. This value is negative for the reactants, while it is positive for the products. β_i as the concentration of species i , can be based on the activity coefficient (γ_i) and mole fraction (x_i). In the following, the chemical equilibrium constant is expressed in Equation 7 as a function of temperature:

$$\ln K_j = A_j + B_j/T + C_j \ln T + D_j T \quad (7)$$

The liquid phase equations (charge equilibrium, component equilibrium, and overall equilibrium) must be solved along with the equations considered for equilibrium constants. The parameters of Equation 7 are reported for the reactions of PC solution in Table 1.

Table 1

Equilibrium equations based on mole fraction in equilibrium models (Višković et al., 2014; Niu et al., 2012).

Equation No.	Equilibrium constant	$\ln K_j = A + B/T + C \ln T + DT$			
		A	B	C	D
(3)	$K_w = \frac{\beta_{\text{H}_3\text{O}^+} \beta_{\text{OH}^-}}{\beta_{\text{H}_2\text{O}}^2}$	132.9	-13446	-22.48	0
(4)	$K_{\text{HCO}_3^-} = \frac{\beta_{\text{HCO}_3^-} \beta_{\text{H}_3\text{O}^+}}{\beta_{\text{CO}_2} \beta_{\text{H}_2\text{O}}^2}$	231.4	-12092	-36.78	0
(5)	$K_{\text{CO}_3^{2-}} = \frac{\beta_{\text{H}_3\text{O}^+} \beta_{\text{CO}_3^{2-}}}{\beta_{\text{HCO}_3^-} \beta_{\text{H}_2\text{O}}}$	216.0	-12432	-35.48	0

2.3. Phase equilibria

The liquid-vapor phase equilibrium states that the vapor phase composition follows the concentration of the liquid phase components using the speciation calculation. The chemical potential should be

typical at temperature T and pressure P regarding each component in the equilibrium between the gas phase and liquid phase as follows (Cullinane et al., 2002):

$$\mu_i^L = \mu_i^G \quad (8)$$

where, μ_i^L and μ_i^G are the chemical potential of component i in the liquid and vapor phase. Using gamma-phi approach, the vapor-liquid equilibrium calculation is given by:

$$y_i \phi_i P = x_i \gamma_i^* H_i \exp\left(\frac{V_i^\infty (P - P_0)}{RT}\right) \quad (9)$$

where, i stands for molecular solute species. For solvent (water), the VLE is given by:

$$y_w \phi_w P = x_w \gamma_w P_w \phi_w^0 \exp\left(\frac{V_w (P - P_0)}{RT}\right) \quad (10)$$

where, γ_i^* and γ_w are the activity coefficients of the unsymmetrical ion and the symmetrical water respectively. H_i is the Henry's constant which calculates the partial pressure of a gas with regard to its solubility in pure water. Henry's constant is the symbol of physical solubility of a solute in the solvent expressed by several investigators and has the overall format as defined below (Wang et al., 2011):

$$\ln H = D_1 + D_2/T + D_3 \ln(T) + D_4 T \quad (11)$$

For CO₂ in pure water, the values of D_1 - D_4 are listed in Table 2.

Table 2

Adjustable parameters of the Henry's law (Wang et al., 2011).

Compound	D_1	D_2	D_3	D_4	Henry's constant unit
CO ₂	110.03	-6789.04	-11.452	-0.0105	Pa

In this study, the equation of virial state is applied to calculating the vapor-phase fugacity coefficients of component i (ϕ_i) and pure and saturated gaseous water (ϕ_w) (Posey et al., 1998).

2.4. Coefficient of activity

As mentioned before, the Wilson-NRF model is utilized in the present study to calculate the activity coefficient of the electrolyte solution. The unsymmetrical surplus Gibbs energy was presumed in an electrolyte system consisting of short-range and long-range expressions as follows:

$$\frac{g^{E,*}}{RT} = \frac{g_{SR}^{E,*}}{RT} + \frac{g_{LR}^{E,*}}{RT} \quad (12)$$

where, SR and LR subscripts stand for short-range and long-range contribution respectively. The non-electrolyte Wilson-NRF (Haghtalab et al., 2009) was applied to the short-range interactions, and the Pizer-Debye-Hückel theory was used for long-range interactions. For non-electrolyte Wilson-NRF model (Haghtalab et al., 2009), the symmetrical excess Gibbs energy is expressed by:

$$g_{SR}^E/RT = Z \left[\sum_i x_i \left(\ln \Gamma_{ii} + \sum_j x_j \ln \beta_{ij} \right) \right] \quad (13)$$

$$\Gamma_{ij} = \frac{\beta_{ij}}{\sum_{k=1}^n x_k \beta_{kj}} \quad (14)$$

$$\beta_{ij} = \exp\left(-\frac{h_{ij} - h_{jj}}{ZRT}\right) \quad (15)$$

where, h_{ij} is enthalpic interaction energy parameter, and Z is coordinate number (herein, $Z = 8$). Considering the disparity of Equation 13, the activity coefficient of the symmetrical component i for short-range contributions is expressed in (Messnaoui et al., 2008):

$$\ln \gamma_{i,SR} = Z \left[1 + \ln \Gamma_{ii} - \sum_j x_j \left(\Gamma_{ij} - \ln \beta_{ij} \beta_{ji} + \sum_k x_k \ln \beta_{kj} \right) \right] \quad (16)$$

The ion activity coefficient of the unsymmetrical short-range can be calculated as follows:

$$\ln \gamma_{ion,SR}^* = \ln \gamma_{ion,SR} - \ln \gamma_{ion,SR}^\infty \quad (17)$$

The Pitzer-Debye-Hückel equation (Posey et al., 1998) was applied to long-range interactions due to the electrostatic forces (Messnaoui et al., 2008) :

$$g_{LR}^E = -A_\phi \left(\frac{1000g/Kg}{M_w} \right)^{.5} \frac{4I_x}{\rho} \ln(1 + \rho I_x^{.5}) \quad (18)$$

where, I_x is the strength of ionic mole fraction, and A_ϕ is the parameter of Debye-Hückel.

$$I_x = \frac{1}{2} \sum_{ion} (z_{ion}^2 x_{ion}) \quad (19)$$

where, z_{ion} is the charge number of ionic components, and x_{ion} is the ionic mole fraction; M_w is the solvent molecular weight. The mentioned coefficient of the long-range contribution is calculated as follows:

$$\ln \gamma_{ion,LR}^* = -A_\phi \left(\frac{1000g/kg}{M_w} \right)^{.5} \left(\frac{2z_{ion}^2}{\rho} \ln(1 + \rho I_x^{.5}) + \frac{z_{ion}^2 I_x^2 - 2I_x^{1.5}}{1 + \rho I_x^{.5}} \right) \quad (20)$$

The water activity coefficient in long-range contribution, i.e. Pitzer-Debye-Hückel equation, is given by:

$$\ln \gamma_{w,LR} = -A_\phi \left(\frac{1000g/kg}{M_w} \right)^{.5} \left(-\frac{2I_x^{1.5}}{1 + \rho I_x^{.5}} \right) \quad (21)$$

Thus, the water activity coefficients as well as the ion species are defined by:

$$\ln \gamma_w = \ln \gamma_{w,SR} + \ln \gamma_{w,LR} \quad (22)$$

$$\ln \gamma_{ion}^* = \ln \gamma_{ion,SR}^* + \ln \gamma_{ion,LR}^* \quad (23)$$

3. Results and discussion

As noted, the non-electrolyte Wilson NRF and Pitzer-Debye-Hückel equation (PDH) equations are formulated to predict the solubility of CO₂ in multicomponent systems. The present study considered two systems of (CO₂ – H₂O) and (CO₂ – H₂O – K₂CO₃). Four species of CO₂ (c), Water (w), anions (a), and cations (C) are available in a CO₂+salt+water system (Messnaoui et al., 2008). Therefore, the energy parameters of the enthalpic interaction in the non-electrolyte Wilson-NRF model for salt systems include h_{ww} , h_{cc} , $h_{wc} = h_{cw}$, $h_{wa} = h_{aw}$, $h_{cc} = h_{cC}$, $h_{ac} = h_{Ca}$, h_{aa} , and h_{CC} . To cover the entire experimental data, the parameters were assumed temperature-dependent.

$$\frac{h_{ij}}{RT} = a(T - T^0)^2 + b(T - T^0) + c \quad (24)$$

where, T^0 is the referenced temperature equal to 298.15 K.

Due to the strong repulsive electrostatic interaction between ions or cations with equal signs and since the anion or cation assumptions are supposed to have a lower number of parameters, their local mole fractions need to be around zero. To this end, h_{aa}/RT and h_{CC}/RT were set at 100 (Pitzer et al., 1983). The interaction parameters of the NWN model were calculated using 1400 data points covering a large temperature and pressure range (Messnaoui et al., 2008). The objective function of optimizing the interaction parameters is defined by:

$$OF = \frac{1}{n} \sum_n \left| \frac{(P_{CO_2})_{CAL} - (P_{CO_2})_{EXP}}{(P_{CO_2})_{EXP}} \right| \quad (25)$$

In which, the number of data points is shown by n .

3.1. (CO₂ – H₂O) system

The absorption of gaseous carbon dioxide in water has been measured by several research groups (Valtz et al., 2004; Li et al., 2004). Their results indicated that higher pressures and temperatures increase the CO₂ solubility. The applied pressure was up to 14.1 MPa, and the applied temperature ranged from 283 to 448 K. This system does not have any chemical reactions, so it does not contain any inions or cations. However, (CO₂ – H₂O) system has three enthalpic interaction energy parameters: h_{ww} , h_{cw} , and h_{cc} . h_{ww} is identical in all the salt/water systems ($h_{ww}/RT = -82.846$). The optimized binary interaction parameters of this system (CO₂ – H₂O) are tabulated in Table 3.

Table 3

The adjusted interaction energy parameters, $\frac{h_{ij}}{RT}$, of CO₂-H₂O system (average absolute deviation (ADD) = 4.2%).

	CO ₂	Water
CO ₂	5.77161	
Water	97.49000	-82.84600

Figure 1 compares the experimental and calculated values of CO₂ pressure versus the gas loading (mol CO₂/mol H₂O) (Austgen et al., 1991).

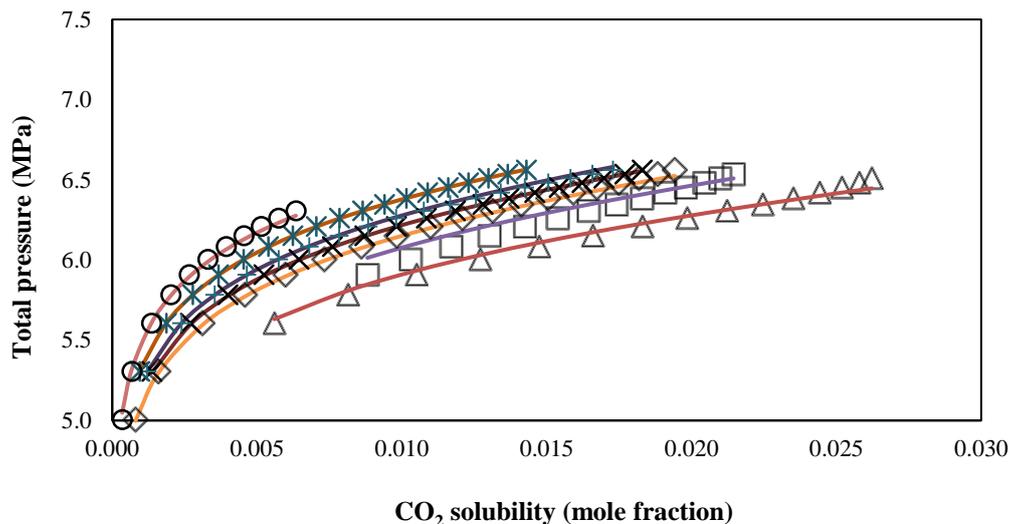


Figure 1

Total pressure as a function of carbon dioxide mole fraction in the water-rich phase at various temperatures; Literature data: (Δ) 273.15 K, (\diamond) 288.15 K, (\bullet) 293.15 K, (\square) 298.15 K, ($+$) 308.15 K, and (\circ) 323.15 K by (Afkhamipour et al., 2013).

The variation of total pressure versus carbon dioxide mole fraction for the (CO₂ – H₂O) system is displayed in Figure 2. The small value of average absolute deviation (ADD = 4.2%) indicates the high accuracy of the model for the CO₂ – H₂O system.

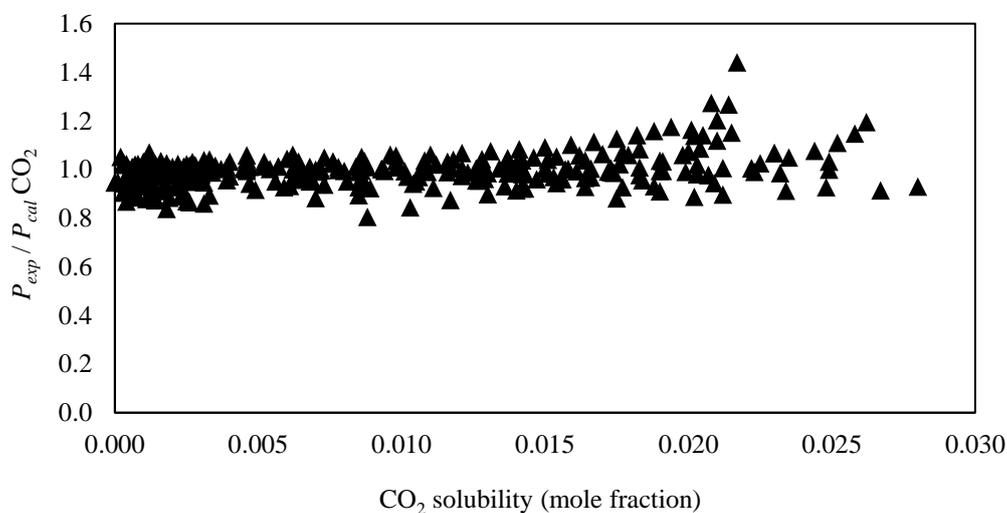


Figure 2

Comparison between the experimental data and model results of total pressure versus mole fraction of CO₂ for the (CO₂ – H₂O) system at various temperatures.

3.2. (CO₂ – H₂O – K₂CO₃) system

The equilibrium chemical reaction should be considered in this system. By combining the reactions (Equations 3-5), one can obtain:



It is obvious that potassium carbonate is able to be highly dissolved in water (Bamberger et al., 2000). The considered concentration of each molecular and ion species in VLE calculations can be obtained by the mass and electro-neutrality condition balance as given below:

$$C_{H_2O} = C_{H_2O}^0 - 2C_{HCO_3^-} \quad (27)$$

$$C_{CO_2} = C_{CO_2}^0 - 2C_{HCO_3^-} \quad (28)$$

$$C_{CO_3^{2-}} = C_{CO_3^{2-}}^0 - 2C_{HCO_3^-} \quad (29)$$

The CO_2 solubility in aqueous hot potassium carbonate has been investigated by other authors, including Tosh et al. (Moore et al., 1997). They concluded the experimental data on CO_2 system, water, potassium carbonate, and bicarbonate. The equilibrium data on $CO_2 - H_2O - K_2CO_3$ system includes 20 – 40 wt. % K_2CO_3 at various conversions of K_2CO_3 to $KHCO_3$ at 70 – 140 °C.

Table 4 presents the interaction parameters which are optimized by the NWN model.

Table 4

The adjusted interaction energy parameters, h_{ij}/RT , of the $K_2CO_3 - CO_2 - H_2O$ system ($ADD = 11.41\%$).

	Water	CO_2	CO_3^{2-}	HCO_3^-	K^+
Water	-82.84600				
CO_2	97.49000	5.77161			
CO_3^{2-}	-2.03258	356.46371	100		
HCO_3^-	40.37018	166.50527	100	100	
K^+	-16.21400	-10.05434	-15.30410	96.94629	100

Figures 3, 4, and 5 display the pressure against CO_2 loading at 20, 30, and 40 wt.% of K_2CO_3 respectively and at a temperature range of 70 – 140 °C.

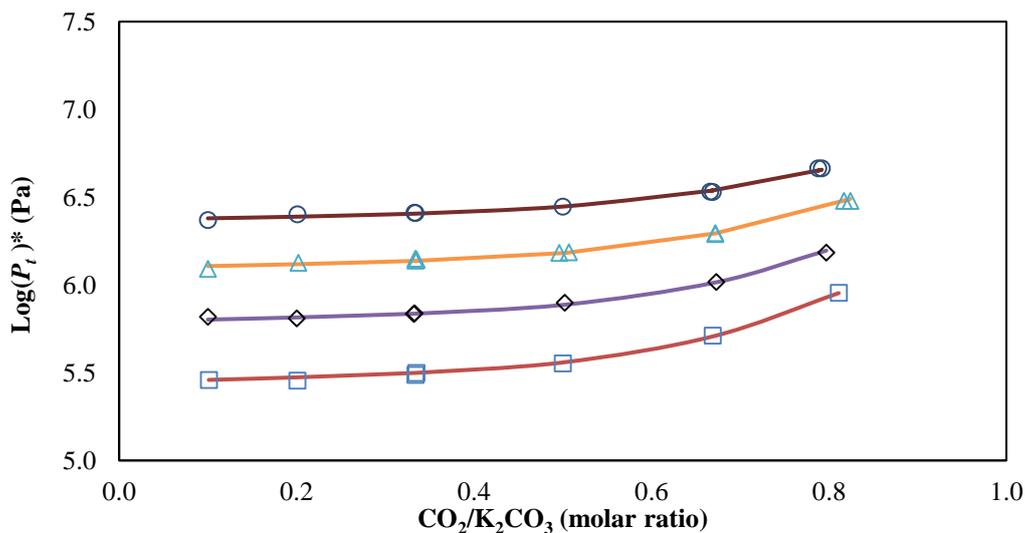


Figure 3

Total pressure of the aqueous solution of ($K_2CO_3 + CO_2$; K_2CO_3 at 20 wt.%): (□) 343.15 K, (◇) 363.15 K, (△) 383.15 K, (○), and 403.15 K (Tosh et al., 1959; Button et al., 1999).

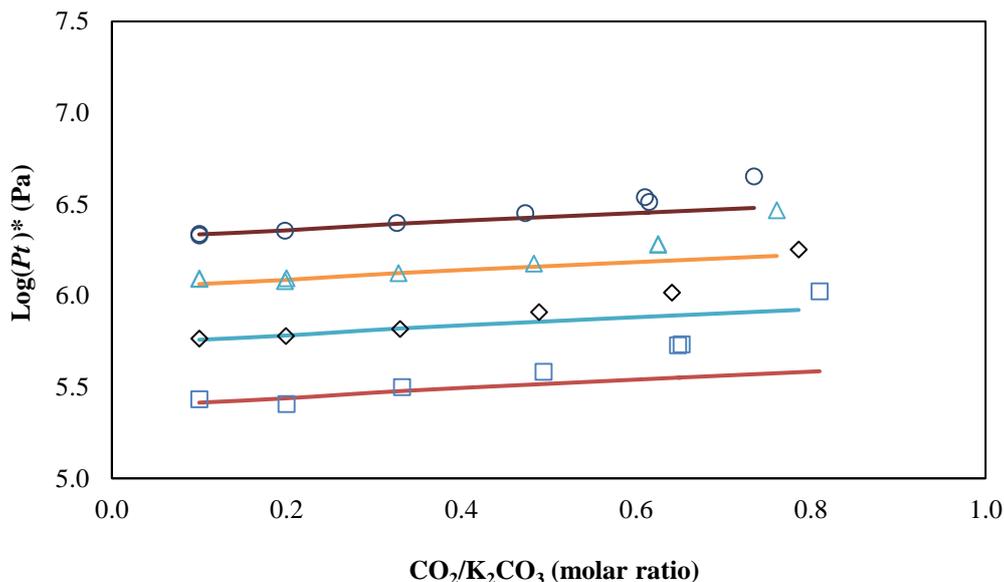


Figure 4

Total pressure of the aqueous solution of (K₂CO₃ + CO₂; K₂CO₃ at 30 wt.%): (□) 343.15 K, (◇) 363.15 K, (Δ) 383.15 K, (○), and 403.15 K (Tosh et al., 1959; Button et al., 1999).

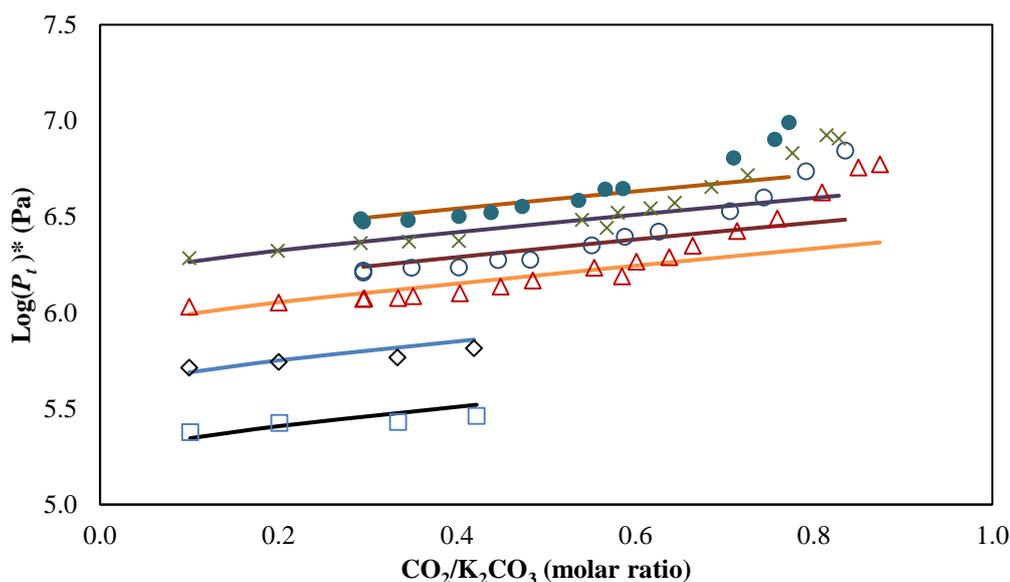


Figure 5

Total pressure of the aqueous solution of (K₂CO₃ + CO₂; K₂CO₃ at 40 wt.%): (□) 343.15 K, (◇) 363.15 K, (Δ) 383.15 K, (○) 393.15 K, (+) 403.15 K, and (●) 413.15 K (Tosh et al., 1959; Button et al., 1999).

The statistical data on as the analysis of the deviation of the total pressure and CO₂ loading for the (CO₂ – K₂CO₃ – H₂O) system are presented in Figure 6. As can be seen, the model agrees well with the data of Tosh et al. (Moore et al., 1997); also, in comparison with pervious researches (Tosh et al., 1959; Cullinane et al., 2005; Álvaro et al., 2007), the average deviation is acceptable (ADD = 11.41%) over the entire range of conditions.

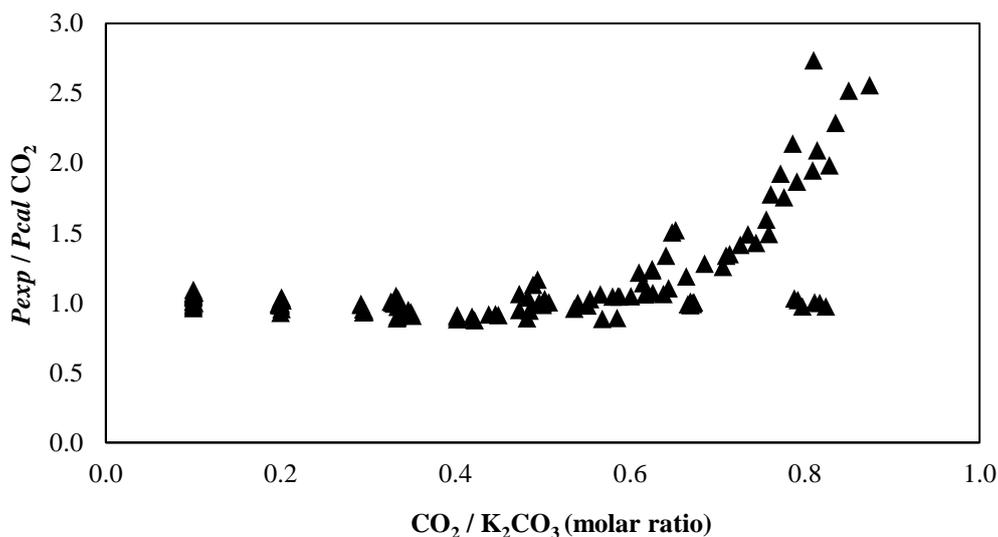


Figure 6

Comparison between the experimental data and model results of total pressure versus the molar ratio of CO_2 to K_2CO_3 at various temperatures.

The results showed that N-Wilson-NRF model is appropriate to design CO_2 capture and gas sweetening processes because it is simple and straightforward; additionally, it has been confirmed herein and in previous investigations that it has high accuracy (Fosbøl et al., 2013).

4. Conclusions

The present research chose the N-Wilson-NRF model to calculate the activity coefficient of short-range interactions. All the species were used in a wide range of temperature and concentration in the aqueous electrolyte system of ($\text{CO}_2 - \text{K}_2\text{CO}_3 - \text{H}_2\text{O}$). CO_2 solubility of the systems was correlated using the molecular framework concept. The results confirmed that the model deviation is less than the other models studied so far, and the model was reasonably utilized to predict solubility data and to design the equilibrium stage of the absorption columns of CO_2 capture processes. Although CO_2 capture using potassium carbonate solutions attracted a wide attention in recent years, and exciting results have been reported in this area, some efforts are required to obtain a deeper understanding of the kinetics and thermodynamics of processes based on potassium carbonate solutions.

Nomenclature

NWN	Non-electrolyte Wilson NRF
PC	Potassium carbonate solution
PDH	Pitzer-Debye-Hückel equation
VLE	Vapor and liquid equation

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