

Kinetics of the Gas–Liquid Reaction between Carbon Dioxide and Hydroxide Ions

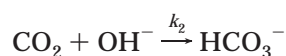
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Gas–liquid reaction kinetics of the reactions between carbon dioxide and hydroxide ions is obtained via dynamic experiments in a stirred cell reactor. The evaluation of experiments is performed using a new technique enabling experimental series without time-consuming solvent purification after each experiment. A new expression for the reaction rate constant at infinite dilution is obtained. Furthermore, a kinetic description of aqueous potassium hydroxide and aqueous sodium hydroxide solutions in the temperature range between 20 and 50 °C is proposed.

1. Introduction

Reactions of dissolved gases with liquid-phase components play an important role in the design of absorption equipment for the purification of gas streams.¹ The reaction between carbon dioxide and hydroxide ions is of special interest because it occurs in all caustic solutions:



This reaction is of second order; the reaction rate can be described by

$$r = k_2[\text{CO}_2][\text{OH}^-] \quad (1)$$

Numerous research works have been concerned with the study of this reaction, most of them for the case of alkaline solutions. The kinetics at infinite dilution was studied by Faurholt² and Brinkman et al.³ and later by Pinsent et al.,⁴ Himmelblau and Babb,⁵ and Sirs.⁶ In these works, experiments were performed in the range between 0 and 40 °C. The data reported in ref 5 should be considered questionable because they exceed the reaction rate constants published by the other authors by a factor of almost 50. Nijssing et al.⁷ and Astarita et al.⁸ extended the previous works to dilute alkaline solutions and described the influence of the ionic strength on the reaction rate constants by the following formula:

$$\log k_2 = \log k_2^\infty + aI \quad (2)$$

Nijssing et al.⁷ found out that different cations in the liquid phase, such as K⁺, Na⁺, or Li⁺, lead to different reaction rate constants; however, they only performed experiments at 20 °C and with ionic strengths above 0.44 kmol/m³. In opposition to ref 7, Astarita et al.⁸ do not consider the cation-type influence. The most recent approach is presented by Pohorecki and Moniuk,⁹ who also give a good review on this subject. They experimentally confirmed the influence of the cation type described by Nijssing et al.⁷ and thus call the approach

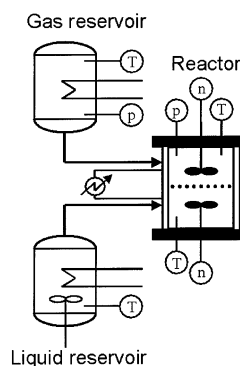


Figure 1. Experimental setup.

of Astarita et al.⁸ into question. Pohorecki and Moniuk⁹ suggest the following expression for the rate constant:

$$\log k_2 = \log k_2^\infty + \sum B_{\text{ion}} I_{\text{ion}} \quad (3)$$

Equation 3 has only been tested at 20 °C and is relevant exclusively for ionic strengths above 0.5 kmol/m³.

The aim of this work is to develop a method for the calculation of kinetic constants for dilute electrolyte solutions based on the infinite dilution kinetics. To develop a consistent approach valid in a broad temperature range, the ionic strength influence on the rate constants has to be considered as a temperature-dependent factor. For this purpose, two different caustic alkaline solutions, the first being aqueous potassium hydroxide and the second aqueous sodium hydroxide, are investigated in the temperature range between 20 and 50 °C. Because the new approach is based on the infinite dilution kinetics, an additional study of this subject is required.

2. Experimental Section

A batch-operated stirred cell reactor (Figure 1) has been used for the experimental determination of the reaction rate constants. This easy-to-use device offers the advantage that no analysis of the liquid phase is required if the reaction mechanism is known because the pressure decrease is the only decisive factor for the evaluation of kinetic parameters. In each experiment, a certain amount of gas is rapidly introduced into the reaction chamber and the pressure decrease caused by

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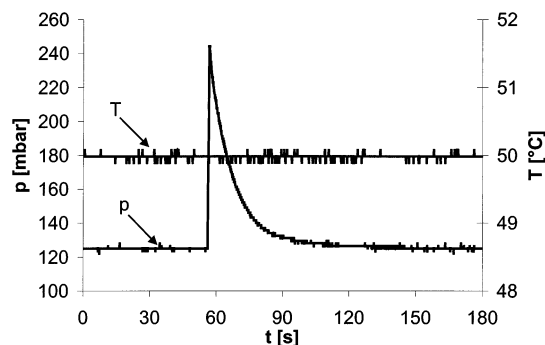


Figure 2. Experimental procedure.

absorption and reaction is recorded (Figure 2). From this record, the reaction velocity at a respective temperature and liquid-phase composition can be derived. A detailed description of the experimental device and procedure is given in ref 10.

The absorption of carbon dioxide with simultaneous reaction in the liquid phase can be described by the well-known enhancement factor. An accurate determination of the forward reaction rate constant with an analytical evaluation method is only possible for the first- or pseudo-first-order irreversible reactions because only for this case are exact analytical solutions for the enhancement factor available.¹¹ Furthermore, because for this type of reaction the application of the mass-transfer coefficient is not required, a possible additional error associated with the mass-transfer correlation is avoided.

Reactions of pseudo-first-order must obey the following criteria:¹²

$$Ha > 2 \quad (4)$$

$$E_{\infty}/Ha > 5 \quad (5)$$

The Hatta number for pseudo-first-order reactions is defined by¹³

$$Ha = \frac{\sqrt{k_{OV}D_{CO_2}}}{k_L} \quad (6)$$

with k_{OV} as the overall pseudo-first-order reaction rate constant:

$$k_{OV} = k_2 c_{OH^-} \quad (7)$$

The enhancement factor for instantaneous reactions in terms of the film theory is expressed as follows:¹¹

$$E_{\infty} = 1 + \frac{D_{OH^-} c_{OH^-}}{D_{CO_2} c_{CO_2}^i} \quad (8)$$

For the preliminary planning of experiments, ratios between the enhancement factor of an instantaneous reaction and the Hatta number are calculated for different partial pressures of carbon dioxide and concentrations of hydroxide ions in the liquid phase. From these results the initial partial pressure of carbon dioxide for a specified concentration of the reactants in the liquid phase can be chosen.

The mass-transfer coefficient required for the calculation of the Hatta number (eq 6) is correlated by the following reactor-specific equation:¹⁰

$$Sh = 0.7082 Re^{0.66} Sc^{0.11} \quad (9)$$

3. Determination Method

For the first-order irreversible reactions, the following expression is valid:¹¹

$$E = \frac{Ha}{\tanh(Ha)} \quad (10)$$

For the calculation of the physical gas solubility in the two different electrolyte solvents, Henry's law is used, with Henry's constant determined by¹²

$$\log\left(\frac{He_{CO_2,S}}{He_{CO_2,H_2O}}\right) = (h_{K^+,Na^+} + h_{OH^-} + h_{CO_2})I \quad (11)$$

The diffusion coefficient is estimated by a modified Stokes–Einstein relation:⁷

$$D_{CO_2,S} \eta_S^{0.85}|_{T=\text{constant}} = D_{CO_2,H_2O} \eta_{H_2O}^{0.85}|_{T=\text{constant}} \quad (12)$$

A description of the mass-transfer acceleration via the enhancement factor (eq 10), taking into account that $E \approx Ha$ for $Ha > 3$ and applying the ideal gas law, yields

$$\frac{dp_{CO_2}}{dt} = - \frac{ART\sqrt{k_{OV}D_{CO_2}}}{V_G} \left(\frac{p_{CO_2}}{He_{CO_2}} - c_{CO_2}^b \right) \quad (13)$$

The liquid-phase balance of the reactor is as follows:

$$\frac{dc_{CO_2}^b}{dt} = \frac{A\sqrt{k_{OV}D_{CO_2}}}{V_L} \left(\frac{p_{CO_2}}{He_{CO_2}} - c_{CO_2}^b \right) - k_{OV}c_{CO_2}^b \quad (14)$$

The solution of eqs 13 and 14 yields

$$p_{CO_2} = C_1 \exp\left(\frac{-\kappa + \lambda}{2}t\right) + C_2 \exp\left(\frac{-\kappa - \lambda}{2}t\right) \quad (15)$$

with the parameters κ and λ containing the desired kinetic constants

$$\kappa = \left(\frac{A}{V_L} + \frac{ART}{V_G He_{CO_2}} \right) \sqrt{k_{OV}D_{CO_2}} + k_{OV} \quad (16)$$

$$\lambda = \sqrt{\kappa^2 - 4 \frac{ARTk_{OV}^{3/2} \sqrt{D_{CO_2}}}{V_G He_{CO_2}}} \quad (17)$$

The constants C_1 and C_2 can be determined using two boundary conditions, which are represented by two values of the measured partial pressure at two different times:

$$C_1 = p_{CO_2,0} - \left(\frac{p_{CO_2,1} - p_{CO_2,0} \exp\left(\frac{-\kappa + \lambda}{2}t_1\right)}{\exp\left(\frac{-\kappa - \lambda}{2}t_1\right) - \exp\left(\frac{-\kappa + \lambda}{2}t_1\right)} \right) \quad (18)$$

$$C_2 = \frac{p_{CO_2,1} - p_{CO_2,0} \exp\left(\frac{-\kappa + \lambda}{2}t_1\right)}{\exp\left(\frac{-\kappa - \lambda}{2}t_1\right) - \exp\left(\frac{-\kappa + \lambda}{2}t_1\right)} \quad (19)$$

The knowledge on the concentration of the dissolved gas in the liquid phase is advantageous because it allows a series of experiments to be realized without time-

consuming purification of the liquid solvent after each experiment which is necessary to fulfill the zero bulk loading assumption. The bulk load at the end of an experiment is calculated by inserting the expression for the partial pressure, eq 15, and its derivative into eq 13:

$$c_{\text{CO}_2}^b|_{t=t_e} = C_1 \exp\left(\frac{-\kappa + \lambda}{2} t_e\right) \left[\frac{1}{He_{\text{CO}_2}} + \frac{V_G}{ART\sqrt{k_{\text{OV}}D_{\text{CO}_2}}} \left(\frac{-\kappa + \lambda}{2}\right) \right] + C_2 \exp\left(\frac{-\kappa - \lambda}{2} t_e\right) \left[\frac{1}{He_{\text{CO}_2}} + \frac{V_G}{ART\sqrt{k_{\text{OV}}D_{\text{CO}_2}}} \left(\frac{-\kappa - \lambda}{2}\right) \right] \quad (20)$$

The constants C_1 and C_2 have to be recalculated for each experiment of a series using the bulk concentration of the previous experiment and the initial pressure of the current experiment

$$C_1 = p_{\text{CO}_2,0} \left[1 - \left(\frac{ART\sqrt{k_{\text{OV}}D_{\text{CO}_2}}}{He_{\text{CO}_2}V_G\lambda} - \frac{\kappa}{2\lambda} + 0.5 \right) \right] - c_{\text{CO}_2}^b|_{t=t_e}^{\text{prev}} \frac{ART\sqrt{k_{\text{OV}}D_{\text{CO}_2}}}{V_G\lambda} \quad (21)$$

$$C_2 = \frac{p_{\text{CO}_2,0}}{V_G\lambda} \left[\frac{ART\sqrt{k_{\text{OV}}D_{\text{CO}_2}}}{He_{\text{CO}_2}} + V_G \left(\frac{-\kappa + \lambda}{2} \right) \right] - c_{\text{CO}_2}^b|_{t=t_e}^{\text{prev}} \frac{ART\sqrt{k_{\text{OV}}D_{\text{CO}_2}}}{V_G\lambda} \quad (22)$$

The evaluation of the kinetic rate constants is performed by a least-squares optimization, in which the overall rate constant is varied to minimize the residuals between the calculated and experimental pressure values. After each evaluation, the criteria for the pseudo-first-order reaction domain (eqs 4 and 5) have to be checked.

4. Kinetics at Infinite Dilution

A precise description of the rate constants at infinite dilution is of crucial importance for the kinetic description of dilute solutions (see eq 3). However, a direct experimental determination of these data is not possible, and therefore, they have to be estimated by extrapolation of finite dilution kinetics. To reach a sufficient accuracy, more than 200 experiments with aqueous potassium hydroxide as the liquid solvent in the temperature range between 20 and 50 °C were performed. In these experiments, the ionic strength was varied between 0.075 and 0.85 kmol/m³. For each solvent concentration, an evaluation of the rate constants is performed, leading to a temperature-dependent exponential function (Figure 3). Such functions taken for different compositions are transferred into a plot of the reaction rate constant versus the solvent concentration (ionic strength) for the whole experimental region, allowing further determination of isothermal lines. The latter are described by polynomial functions of second

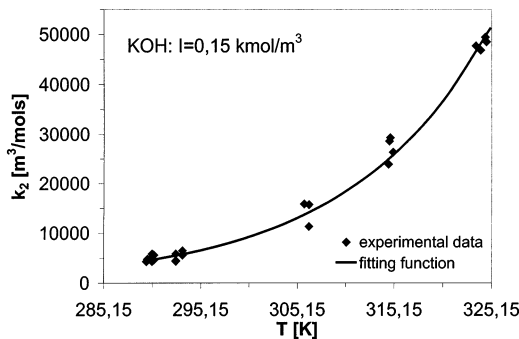


Figure 3. Reaction rate constants for a specific potassium hydroxide concentration.

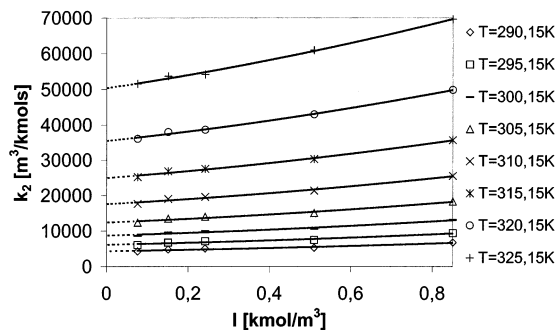


Figure 4. Reaction rate constants for potassium hydroxide solutions with extrapolation to zero ionic strength: points, experimental values; lines, polynomial fitting functions.

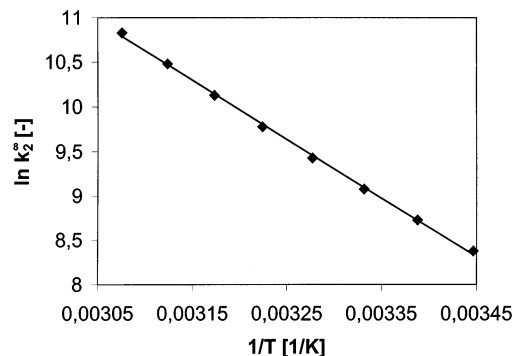


Figure 5. Arrhenius plot of infinite dilution reaction rate constants.

order, which were found to be best suited. It can be seen that the constants depend on the concentration of electrolytes present in the liquid phase.

Extrapolation of an isothermal line to zero ionic strength delivers the infinite dilution reaction rate constant at the respective temperature (Figure 4). Performing this extrapolation procedure for each temperature, we obtain the following Arrhenius expression (Figure 5):

$$k_2^\infty = 3.27869 \times 10^{13} \exp(-54971/RT) \quad (23)$$

Figure 6 demonstrates a good agreement between the new expression for the reaction rate constants and those predicted in refs 4, 6, 7, and 9 in the temperature range between 20 and 40 °C. Essentially, the new expression (eq 23) is valid in a wider range, up to 50 °C.

5. Kinetics for Dilute Solutions

Brönsted¹⁴ and Bjerrum¹⁵ proposed an equation for the calculation of reaction rate constants of electrolyte

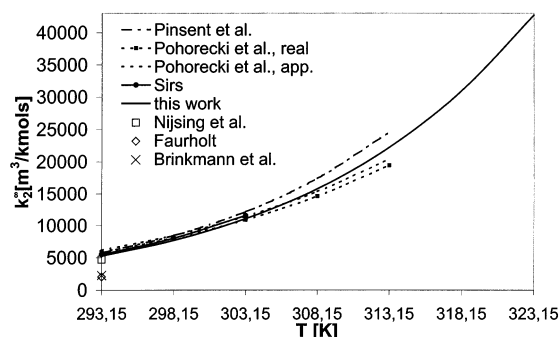


Figure 6. Comparison of the reaction rate constants at infinite dilution.

systems. For the investigated reaction, this equation reads as follows:

$$k_2 = k_2^\infty \frac{\gamma_{\text{CO}_2} \gamma_{\text{OH}^-}}{\gamma_{\text{HCO}_3^-}} \quad (24)$$

Brönsted¹⁴ postulated that the activity coefficient of an ion is determined by its charge and that species with identical electrical charges have equal activity coefficients. Application of this assumption and the Debye–McAuley expression for the activity coefficients of a neutral molecule in an electrolyte solution¹⁶

$$\gamma_{\text{CO}_2} = \exp(\beta_{\text{CO}_2} I) \quad (25)$$

yields

$$k_2 = k_2^\infty \exp(\beta_{\text{CO}_2} I) \quad (26)$$

Because the nonideal behavior of electrolyte systems is influenced by temperature, β_{CO_2} is temperature-dependent. Application of eq 26 enables a determination of reaction kinetic constants for various ionic strengths and temperatures.

5.1. Potassium Hydroxide Solutions. For the investigation of the influence of the ionic strength on the reaction rate constants, the results obtained for the infinite dilution kinetics (Figure 5) are used. The temperature dependence of β_{CO_2} is derived via a least-squares minimization. Fitting β_{CO_2} for potassium hydroxide solutions in the temperature range between 20 and 50 °C, with the ionic strength varying between 0.075 and 0.85 kmol/m³, gives

$$\beta_{\text{CO}_2} = 3.3968 \times 10^{-4} T^2 - 2.1215 \times 10^{-1} T + 33.506 \quad (27)$$

The deviations between the experimental and calculated reaction rate constants are less than 4% (Figure 7). The temperature dependence of β_{CO_2} is similar to that of the temperature-dependent contribution parameter h_{CO_2} in eq 11.¹² This was expected because both parameters describe the deviation from ideal behavior due to the electrolyte nature of the solution.

Because in the literature only data measured at 20 °C are available, a comparison of the results calculated by eqs 26 and 27 can only be made at this temperature. The new approach shows a good agreement with the data suggested by Nijsing et al.⁷ and Pohorecki and Moniuk⁹ (Figure 8). The rate constants calculated by eqs 26 and 27 at higher temperatures are also demonstrated in Figure 8.

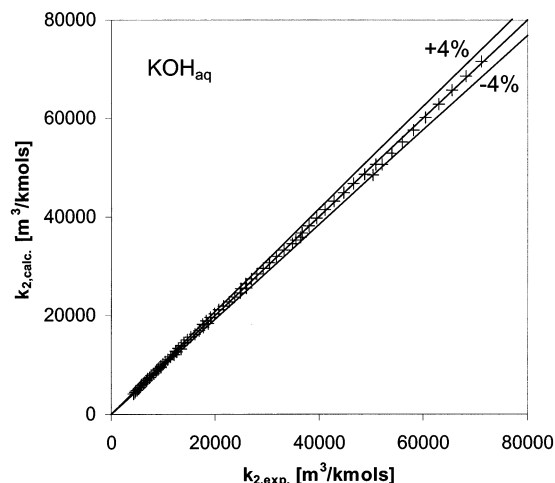


Figure 7. Calculated vs experimental reaction rate constants for potassium hydroxide: $T = 20\text{--}50$ °C; $I = 0.075\text{--}0.85$ kmol/m³.

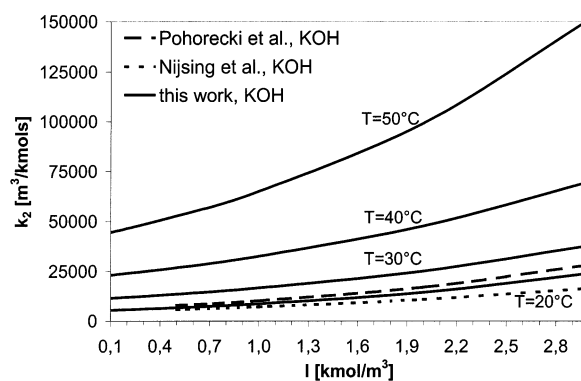


Figure 8. Calculated reaction rate constants and comparison with the literature data for potassium hydroxide solutions: $T = 20\text{--}50$ °C; $I = 0.075\text{--}3.0$ kmol/m³.

To determine rate constants at higher solvent concentrations, an extrapolation of the polynomial functions for the isotherms is performed beyond the range of experimental measurements, up to an ionic strength of 3.0 kmol/m³. From Figure 8 it can be seen that the deviations between the proposed approach and the data suggested by Nijsing et al.⁷ and Pohorecki and Moniuk⁹ remain almost constant and a good agreement is preserved. This leads to the conclusion that the suggested approach is applicable to electrolyte solutions with higher concentrations.

5.2. Sodium Hydroxide Solutions. Similar experimental investigations were performed for the system of aqueous sodium hydroxide with an ionic strength varying between 0.3 and 1.25 kmol/m³. For infinite dilution the experimental results according to eq 23 are applied. Similar to the experimental method for aqueous potassium hydroxide, reaction rate constants are obtained for each solvent composition. These individual evaluations lead to exponential expressions for the description of the reaction rate constants, similar to Figure 3. Plotting the calculated rate constants versus the ionic strength for the whole experimental region yields isothermal lines (Figure 9). In this case the constants also depend on the amount of electrolytes present in the liquid phase, but at identical temperatures and ionic strengths, they are lower than those for the previous system. This is in line with the results of Nijsing et al.⁷ and Pohorecki and Moniuk,⁹ and thus the application of a kinetic description neglecting the influence of ions⁸ cannot be recommended.

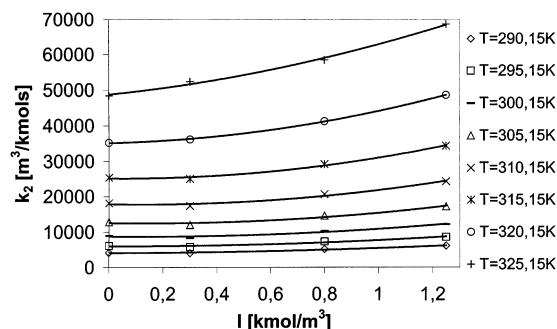


Figure 9. Reaction rate constants for sodium hydroxide solutions: points, experimental values; lines, polynomial fitting functions.

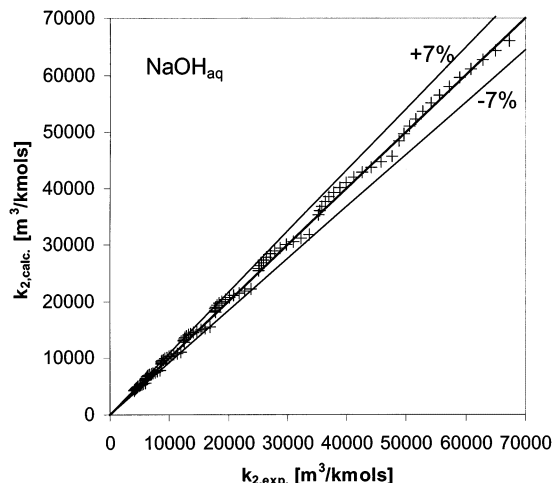


Figure 10. Calculated vs experimental reaction rate constants for sodium hydroxide solutions ($T = 20\text{--}50\text{ }^{\circ}\text{C}$; $I = 0.3\text{--}1.25\text{ kmol/m}^3$).

For the temperature range from 20 to 50 $^{\circ}\text{C}$ and the ionic strength varying between 0.3 and 1.25 kmol/m^3 , the following expression is obtained for sodium hydroxide solutions via a least-squares minimization:

$$\beta_{\text{CO}_2} = 2.8300 \times 10^{-4} T^2 - 1.7367 \times 10^{-1} T + 26.809 \quad (28)$$

In this range the relative deviations between the experimental and calculated reaction rate constants are less than 7% (Figure 10). The comparison of the data calculated by eqs 26 and 28 in the evaluation region with those available from the literature at 20 $^{\circ}\text{C}$ shows a very good agreement with the results obtained by Nijsing et al.;⁷ however, they are lower than the reaction rate constants predicted by Pohorecki and Moniuk⁹ (Figure 11).

This tendency holds also when extending the polynomial fits from the experimental region up to an ionic strength of 3.0 kmol/m^3 . While the deviations between the new results and the reaction rates predicted by Pohorecki and Moniuk⁹ at 20 $^{\circ}\text{C}$ are visible, the agreement with the approach of Nijsing et al.⁷ is very good in the entire range (Figure 11). Therefore, it can be concluded that the new approach for aqueous sodium hydroxide is also applicable to electrolyte solutions with higher concentrations.

6. Conclusions

In this paper the gas–liquid reaction kinetics of the reaction between carbon dioxide and hydroxide ions is

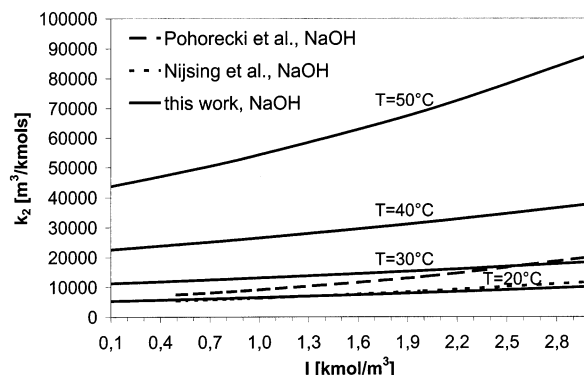


Figure 11. Calculated reaction rate constants and comparison with the literature data for sodium hydroxide solutions: $T = 20\text{--}50\text{ }^{\circ}\text{C}$; $I = 0.3\text{--}3.0\text{ kmol/m}^3$.

derived based on dynamic experiments in a stirred cell reactor. The experiments have been performed in the temperature range between 20 and 50 $^{\circ}\text{C}$.

A detailed description of the new evaluation technique taking into account the liquid bulk load is presented. This method allows experiments to be performed in series because the liquid solvent does not have to be purged of contaminants after each experimental cycle.

A new Arrhenius expression for the kinetics at infinite dilution is derived by extrapolation of kinetic rate constant isotherms obtained from experiments with aqueous potassium hydroxide. This kinetic approach shows a good agreement with the literature data.

For the description of the influence of the ionic strength on the reaction rate constants, a new expression is derived from the Brönsted–Bjerrum equation, taking the infinite dilution kinetics into consideration. This expression allows for the determination of the reaction kinetic constants at arbitrary ionic strengths and temperatures. Because the real infinite dilution reaction rate constants are used in the calculations, prediction of rate constants for electrolyte solutions, both with low and higher concentrations, can be made. Experiments are performed with two different alkaline solutions, aqueous potassium hydroxide and aqueous sodium hydroxide, in the temperature range from 20 to 50 $^{\circ}\text{C}$. It can be shown that at equal temperatures and ionic strengths the rate constants of the reaction for the systems containing sodium ions are lower than those for the systems containing potassium ions. This means that the influence of the cations present in the solution on the reaction rate constant cannot be neglected.

For the investigated systems, two new kinetic expressions taking into account the influence of temperature and ionic strength are suggested, with an accuracy of 4% for potassium hydroxide and 7% for sodium hydroxide, in the temperature range from 20 to 50 $^{\circ}\text{C}$. The comparison of the reaction rate constants at 20 $^{\circ}\text{C}$ predicted by the new approach with the expressions available from the literature^{7,9} for aqueous potassium hydroxide shows a good agreement. Furthermore, for aqueous sodium hydroxide, the new expression is consistent with the values predicted in ref 7 at 20 $^{\circ}\text{C}$.

The comparisons of the extrapolated data obtained with the new approach in an extended range of the ionic strength with the values from literature allow the conclusion that both suggested kinetic expressions are also applicable to higher solvent concentrations.

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Notation

A = interfacial area (m^2)
 a = constant in eq 2
 B_{ion} = ion contribution parameter in eq 3 (m^3/kmol)
 c = molar concentration (kmol/m^3)
 $c_{\text{CO}_2}^{\text{b}}|_{t=t_e}^{\text{prev}}$ = CO_2 bulk concentration at the end of the previous experiment (kmol/m^3)
 d = stirrer diameter (m)
 D = diffusion coefficient (m^2/s)
 E = enhancement factor
 E_{∞} = enhancement factor of the instantaneous reaction
 h = contribution parameter in eq 11
 Ha = Hatta number
 He = Henry's constant ($\text{Pa}\cdot\text{m}^3/\text{kmol}$)
 $I (=0.5\sum c_i z_i^2)$ = ionic strength (kmol/m^3)
 I_{ion} = contribution of an ion to the ionic strength (kmol/m^3)
 k_L = mass-transfer coefficient (m/s)
 k_2 = second-order reaction rate constant ($\text{m}^3/\text{kmol}\cdot\text{s}$)
 k_2^{∞} = second-order infinite dilution reaction rate constant ($\text{m}^3/\text{kmol}\cdot\text{s}$)
 k_{OV} = overall reaction rate constant ($\text{m}^3/\text{kmol}\cdot\text{s}$)
 n = stirrer speed ($1/\text{s}$)
 p = partial pressure (Pa)
 r = reaction rate ($\text{kmol}/\text{m}^3\cdot\text{s}$)
 R = gas constant ($8.3144 \text{ kJ}/\text{kmol}\cdot\text{K}$)
 $Re (= \rho n d^2 / \eta)$ = Reynolds number
 $Sc (= \eta / \rho D)$ = Schmidt number
 $Sh (= k_L d / D)$ = Sherwood number
 t = time (s)
 T = temperature (K)
 V = volume (m^3)
 z = electrical charge

Greek Letters

β = contribution factor in eq 26
 γ = activity coefficient
 κ = parameter defined by eq 16
 λ = parameter defined by eq 17
 η = viscosity ($\text{Pa}\cdot\text{s}$)
 ρ = density (kg/m^3)

Subscripts

e = end of the experiment
 G = gas phase

i = component index
 L = liquid phase
 S = solution
 0 = start of the experiment
 1 = end of the evaluation time

Superscripts

b = bulk phase
 i = interface

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