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Diffusivities in Aqueous Solutions of the Potassium Salt of Amino Acids

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Diffusivities of the potassium salt of taurine, sarcosine, L-proline, and glycine have been determined in aqueous solutions using the Taylor dispersion technique over a temperature range from (293 to 368) K and salt concentrations ranging from (0 to 3.0) mol·dm⁻³. The diffusion coefficients of absorbed N₂O in aqueous solutions of the potassium salt of taurine, sarcosine, L-proline, and glycine have been determined over a temperature range from (293 to 333) K and salt concentrations ranging from (1.0 to 3.0) mol·dm⁻³. The experimental results have been compared to estimated values from a modified Stokes–Einstein relation and available literature data. The diffusivities can be estimated with sufficient accuracy using a modified Stokes–Einstein relation. The diffusion coefficients of CO₂ in the aqueous amino acid salt solutions can be estimated by means of the N₂O analogy.

Introduction

Aqueous solutions of amines are frequently used for the removal of acid gases, such as CO₂ and H₂S, from a variety of gas streams. In particular, aqueous solutions of alkanolamines and blends of alkanolamines are widely applied in gas treating. Alkanolamines can undergo degradation in oxygen atmosphere, usually encountered in the treatment of flue gases. The relative high volatility of alkanolamines also leads to losses of the compound during commercial operation of CO₂ capture plants. Aqueous solutions of salts of amino acids might be an attractive alternative to alkanolamines. They have been found to have better resistance to oxidative degradation, and their reactivity with CO₂ is comparable to aqueous alkanolamines of related classes. The ionic nature of the aqueous solutions of salts of amino acids results in a negligible volatility.¹–⁵

Knowledge of diffusion coefficients is needed for the design of absorbers and desorbers in a commercial CO₂ capture plant, as they are related to the mass transfer coefficients. They are also essential for a correct and accurate interpretation of many (laboratory scale) absorption rate experiments, e.g., experiments aimed to determine the intrinsic kinetics in a gas–liquid process. Diffusivities of the potassium salt of taurine, sarcosine, L-proline, and glycine in aqueous solutions determined by the Taylor dispersion technique at temperatures from (293 to 368) K and salt concentrations ranging from (0 to 3.0) mol·dm⁻³ are presented in this work. Diffusivities of N₂O in the same solutions determined with the same technique at temperatures from (293 to 333) K and salt concentrations ranging from (1.0 to 3.0) mol·dm⁻³ are also presented. The results are correlated and compared to literature values.

Theory and Experimental Procedures

General Theory of the Taylor Dispersion Technique. The diffusivities were determined using the Taylor dispersion technique. A square pulse of a solute solution was injected into a solvent solution showing laminar flow through a capillary tube. The solute solution contained the same components as the solvent solution but with a slightly higher concentration of the compound of which diffusivities were to be determined (an addition of approximately 0.05 mol·dm⁻³). In the case of absorbed gas measurements, the solute solution contained the same amounts of liquid components as the solvent solution, but with additional N₂O gas absorbed. As an example of liquid measurements, for measurements of diffusivities in an approximately 1.00 mol·dm⁻³ solution of potassium taurate, the solvent solution contained 1.00 mol·dm⁻³ potassium taurate. The solute solution contained 1.05 mol·dm⁻³ potassium taurate, and a square pulse of this solution was injected into the flowing solvent solution. For gas measurements in aqueous solutions of potassium taurate at 1.00 mol·dm⁻³, both the solvent and solute solution contained 1.00 mol·dm⁻³ potassium taurate. The solute solution contained additional absorbed N₂O, and a square pulse of this solution was injected into the flowing solvent solution. The combined action of axial convection and radial and axial molecular diffusion eventually changed the pulse into a Gaussian curve. The theory, mathematical description, and procedure of such diffusivity measurements have been described in detail by Taylor,⁶,⁷ Aris,⁸ and Hamborg et al.⁹

The experimental setup used is shown schematically in Figure 1. Two vessels containing the solute solution and the solvent solution were kept under a constant 5·10¹² kPa pressure of saturated helium to create a constant flow of the solute and the solvent. Saturated helium was used to prevent any concentration changes in the solute and solvent solution as the liquid level in the two closed vessels decreased during a measurement due to the solution outflow. Introduction of a solute pulse was done by switching an air actuated six-way valve back and forth within a few seconds. The capillary tube was elliptical coiled and placed in a water bath for temperature control of ± 0.1 K. The flow velocity was controlled with mass flow controllers (Rosemount Flowmegas 5881), located behind the refractive index (RI) detector (Varian 350 RI) and the six-way pulse valve to obtain a constant pulsation-free solute and solvent solution flow throughout the measurement. To avoid bubble formation inside
Influence of the pressure on the diffusion coefficients can be additionally to the dispersion process. This topic has been extensively discussed by Alizadeh et al. and Snijder et al. Prior to each experiment, the pressure applied.10,11

For absorbed gas diffusion measurements, N2O was absorbed extensively discussed by Alizadeh et al. and Snijder et al. and the secondary flows present in the flow can contribute to the elliptic coiling of the tube. The varying path lengths traversed by the fluid at different radial positions in the tube and the flow conditions are given in Table 1. The measurements had to be carried out at a value of (De)2Sc lower than the critical one.

To avoid this disturbance, the critical (De)2Sc was determined for each system. The dimensionless (De)2Sc number is defined where Re is the well-known Reynolds number and µ and ρ are the solvent viscosity and density. The other parameters are defined in Table 1. The measurements had to be carried out at a value of (De)2Sc lower than the critical one.

A disturbance of the laminar fluid flow profile can occur due to the elliptic coiling of the tube. The varying path lengths traversed by the fluid at different radial positions in the tube and the secondary flows present in the flow can contribute additionally to the dispersion process. This topic has been extensively discussed by Alizadeh et al. and Snijder et al. To avoid this disturbance, the critical (De)2Sc was determined for each system. The dimensionless (De)2Sc number is defined as

$$\text{De} = \frac{Re}{10^4}$$  \hspace{1cm} (1)

$$\text{Sc} = \frac{\mu}{\rho D}$$  \hspace{1cm} (2)

where Re is the well-known Reynolds number and µ and ρ are the solvent viscosity and density. The other parameters are defined in Table 1. The measurements had to be carried out at a value of (De)2Sc lower than the critical one.

The aqueous amino acid zwitterion will be deprotonated and form a potassium salt of the amino acid. The other parameters are defined in Table 1. The measurements had to be carried out at a value of (De)2Sc lower than the critical one.

$$\text{De} = \frac{Re}{10^4}$$  \hspace{1cm} (1)

$$\text{Sc} = \frac{\mu}{\rho D}$$  \hspace{1cm} (2)

The aqueous amino acid zwitterion will be deprotonated and form a potassium salt of the amino acid. The other parameters are defined in Table 1. The measurements had to be carried out at a value of (De)2Sc lower than the critical one.

**Chemicals.** Taurine [107-35-7], sarcosine [107-97-1], glycine [56-40-6], L-proline [147-85-3], KOH [1310-58-3] (Sigma-Aldrich), N2O (10024-97-2), and helium [7440-59-7] (Linde Gas) were used as supplied. Water was demineralized and further purified by vacuum distillation.

**Salt Preparation.** The respective aqueous solutions of the potassium salt of the amino acids were prepared by adding equimolar amounts of KOH and amino acid based on weight. The aqueous amino acid zwitterion will be deprotonated according to

$$\text{OOC} - \text{R} - \text{NH}_3^+ + \text{KOH} \rightarrow \text{K}^+ + \text{OOC} - \text{R} - \text{NH}_3 + \text{H}_2\text{O}$$  \hspace{1cm} (3)

The results of salt diffusivity measurements can be described by

$$c_m = \frac{N_{mij,SAA}}{2\pi R^2 \sqrt{\pi K_{SAA}t}} \exp\left(-\frac{(L - ut)^2}{4K_{SAA}t}\right)$$  \hspace{1cm} (4)

$$K_{SAA} = \frac{u^2 R^2}{48D_{SAA}} + D_{SAA}$$  \hspace{1cm} (5)

where $c_m$ is the measured excess concentration profile at the end of the capillary tube; $N_{mij,SAA}$ is the excess number of moles of the salt of amino acid injected; $t$ is the time; and $D_{SAA}$ is the binary diffusion coefficient. The subscript SAA refers to the salt of the amino acid. The other parameters are defined in Table 1. $N_{mij,SAA}$, $u$, and $D_{SAA}$ are the independent parameters used to fit eqs 4 and 5 to the experimentally recorded results. Since the concentration of the solute pulse decreases because of

![Page of a document with a table and text]
Table 3. Diffusion Coefficients, $D$, of Salts of Amino Acids in Aqueous Solutions at Various Temperatures, $T$, and Reference Concentrations, $c_{ref}$

<table>
<thead>
<tr>
<th>$T$</th>
<th>$D_{PT}$</th>
<th>$c_{ref,PT}$</th>
<th>$D_{PS}$</th>
<th>$c_{ref,PS}$</th>
<th>$D_{PP}$</th>
<th>$c_{ref,PP}$</th>
<th>$D_{PG}$</th>
<th>$c_{ref,PG}$</th>
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<tbody>
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<td>K</td>
<td>$10^{-5}\text{m}^2\text{s}^{-1}$</td>
<td>mol$\cdot$dm$^{-3}$</td>
<td>$10^{-5}\text{m}^2\text{s}^{-1}$</td>
<td>mol$\cdot$dm$^{-3}$</td>
<td>$10^{-5}\text{m}^2\text{s}^{-1}$</td>
<td>mol$\cdot$dm$^{-3}$</td>
<td>$10^{-5}\text{m}^2\text{s}^{-1}$</td>
<td>mol$\cdot$dm$^{-3}$</td>
</tr>
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<td>293.15</td>
<td>1.01</td>
<td>0.3$\cdot$10$^{-3}$</td>
<td>0.961</td>
<td>0.9$\cdot$10$^{-3}$</td>
<td>0.924</td>
<td>0.9$\cdot$10$^{-3}$</td>
<td>0.999</td>
<td>0.5$\cdot$10$^{-3}$</td>
</tr>
<tr>
<td>298.15</td>
<td>1.13</td>
<td>0.3$\cdot$10$^{-3}$</td>
<td>1.10</td>
<td>0.1$\cdot$10$^{-2}$</td>
<td>1.04</td>
<td>0.5$\cdot$10$^{-4}$</td>
<td>1.11</td>
<td>0.5$\cdot$10$^{-3}$</td>
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<tr>
<td>303.15</td>
<td>1.26</td>
<td>0.4$\cdot$10$^{-3}$</td>
<td>1.23</td>
<td>0.1$\cdot$10$^{-2}$</td>
<td>1.17</td>
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<td>1.25</td>
<td>0.6$\cdot$10$^{-3}$</td>
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<tr>
<td>313.15</td>
<td>1.56</td>
<td>0.4$\cdot$10$^{-3}$</td>
<td>1.54</td>
<td>0.1$\cdot$10$^{-2}$</td>
<td>1.48</td>
<td>0.5$\cdot$10$^{-4}$</td>
<td>1.56</td>
<td>0.6$\cdot$10$^{-3}$</td>
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<td>2.34</td>
<td>0.2$\cdot$10$^{-2}$</td>
<td>2.18</td>
<td>0.6$\cdot$10$^{-4}$</td>
<td>2.28</td>
<td>0.8$\cdot$10$^{-3}$</td>
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<tr>
<td>343.15</td>
<td>2.71</td>
<td>0.5$\cdot$10$^{-3}$</td>
<td>2.71</td>
<td>0.2$\cdot$10$^{-2}$</td>
<td>2.55</td>
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<td>2.70</td>
<td>0.9$\cdot$10$^{-3}$</td>
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<td>353.15</td>
<td>3.17</td>
<td>0.6$\cdot$10$^{-3}$</td>
<td>3.13</td>
<td>0.2$\cdot$10$^{-2}$</td>
<td>2.93</td>
<td>0.8$\cdot$10$^{-4}$</td>
<td>3.11</td>
<td>0.9$\cdot$10$^{-3}$</td>
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<td>363.15</td>
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<td>0.6$\cdot$10$^{-3}$</td>
<td>3.56</td>
<td>0.2$\cdot$10$^{-2}$</td>
<td>3.42</td>
<td>0.8$\cdot$10$^{-4}$</td>
<td>3.56</td>
<td>0.1$\cdot$10$^{-2}$</td>
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<td>3.36</td>
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<td>0.3$\cdot$10$^{-2}$</td>
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<td>1.0$\cdot$10$^{-4}$</td>
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<td>0.1$\cdot$10$^{-2}$</td>
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<td>1.0$\cdot$10$^{-3}$</td>
<td>3.64</td>
<td>0.1$\cdot$10$^{-2}$</td>
<td>3.19</td>
<td>1.0$\cdot$10$^{-4}$</td>
<td>3.11</td>
<td>1.0$\cdot$10$^{-2}$</td>
</tr>
<tr>
<td>403.15</td>
<td>3.40</td>
<td>0.6$\cdot$10$^{-3}$</td>
<td>3.49</td>
<td>0.2$\cdot$10$^{-2}$</td>
<td>2.84</td>
<td>2.9$\cdot$10$^{-4}$</td>
<td>3.95</td>
<td>3.0$\cdot$10$^{-3}$</td>
</tr>
</tbody>
</table>

$^a$ Subscripts: PT = potassium taurate; PS = potassium sarcosinate; PP = potassium proline; PG = potassium glycinate.

Table 4. Diffusivity of N$_2$O, $D_{N_2O}$, in Aqueous Solutions of the Potassium Salt of Amino Acids at Various Temperatures, $T$, and Solvent Concentrations, $c$, determined. Alizadeh et al.$^{12}$ referred to this as the reference concentration $c_{ref}$ given by

$$c_{ref} = c_{sol} + \frac{N_{inj,SAA}}{\pi R^2 \sqrt{2c_{N_2O}}} \left( \frac{5}{16} - \frac{1}{8\sqrt{\pi}} \right)$$

where $c_{sol}$ is the solvent concentration and $t'$ is the time at which the measurement ends.

Absorbed Gas Diffusivity Measurements. The experimental procedure used to determine diffusivities of absorbed N$_2$O in aqueous solutions by the Taylor dispersion technique has been given in detail by Hamborg et al.$^9$ The recorded pulses are described by eqs 7 to 9

$$c_m = \frac{N_{inj,N_2O}}{2\pi R^2 \sqrt{\pi K_{N_2O} t}} \exp \left( -\frac{(L - ut)^2}{4K_{N_2O}} \right) + \frac{N_{inj,SAA}}{2\pi R^2 \sqrt{\pi K_{N_2O}} t} \exp \left( -\frac{(L - ut)^2}{4K_{N_2O}} \right)$$

$$K_{N_2O} = \frac{\mu^2 R^2}{48D_{N_2O}} + D_{N_2O}$$

$^a$ Subscripts: PT = potassium taurate; PS = potassium sarcosinate; PP = potassium proline; PG = potassium glycinate.
\[ K_{\text{SAA}} = \frac{u^2 R^2}{48 D_{\text{SAA}}} + D_{\text{SAA}} \]  

(9)

where \( c_m \) is the measured concentration profile; \( N_{\text{inj}, \text{N2O}} \) and \( N_{\text{inj, SAA}} \) are the respective numbers of moles of absorbed N\(_2\)O and excess number of moles of salt of amino acid injected; \( t \) is the time; and \( D_{\text{N2O}} \) and \( D_{\text{SAA}} \) are the respective diffusion coefficients of absorbed N\(_2\)O and the potassium salt of amino acid. The other parameters are defined in Table 1. The parameters \( N_{\text{inj}, \text{N2O}}, N_{\text{inj, SAA}}, u, \) and \( D_{\text{N2O}} \) are the independent variables used to fit the equations to the experimental data. The value of \( D_{\text{SAA}} \) is from the previous measurement as aforementioned, and the influence of absorbed N\(_2\)O present in the current measurement on this value is neglected.

**Results and Discussion**

**Diffusivity of the Potassium Salt of Amino Acids in Aqueous Solutions.** Values of the diffusivities of the potassium salt of taurine, sarcosine, L-proline, and glycine in aqueous solutions were measured and averaged over at least three measurements at temperatures from (293 to 368) K and salt concentrations ranging from (0 to 3.0) mol·dm\(^{-3}\). The results are presented in Table 3. The experimental uncertainty is estimated to be 5 % at temperatures less than 333 K and 10 % at higher temperatures. The reproducibility is within 2 %. The critical (De)\(^2\)Sc number was found to be about 100 for these systems.

The theory of diffusion of aqueous salts at low concentration is well developed.\(^{14}\) For dilute solutions of a single salt, the diffusion coefficient can be estimated by the Nernst–Haskell equation

\[ D^0 = \frac{RT[(1/z_+)+(1/z_-)]}{F^2[(1/\lambda_0^0)+(1/\lambda_0^0)]} \]  

(10)

where \( D^0 \) is the diffusion coefficient at infinite dilution; \( R \) is the gas constant; \( T \) is the temperature; \( F \) is the faraday constant; \( \lambda_0^0 \) and \( \lambda_0^0 \) are the limiting ionic conductivities; and \( z_+ \) and \( z_- \) are the respective valences of the cation and anion. In the case of the potassium salt of glycine at 298.15 K, limiting ionic conductivities have been reported by D'yachkova et al.,\(^{15}\) Miyamoto & Schmidt,\(^{16}\) and Mehl & Schmidt\(^{17}\) for glycinate. These values are listed in Table 2. The limiting ionic conductivity of the potassium ion is reported by Poling et al.\(^{14}\) to be 73.5 cm\(^2\)·S·mol\(^{-1}\). By the use of these values and eq 10, the respective estimated diffusivities at infinite dilution, \( D_{\text{est, potassium glycinate}} \), are calculated and listed in Table 2. From Table 3, the experimentally determined value at low salt concentrations, \( D_{\text{potassium glycinate}} \), is 1.1×10\(^{-9}\) m\(^2\)·s\(^{-1}\) at 298.15 K. The relative deviation between the estimated and the experimental values is listed in Table 2. The limiting ionic conductivity of 39.2 cm\(^2\)·S·mol\(^{-1}\) from Mehl & Schmidt\(^{17}\) is most likely too high for an ion of the size of glycinate. This can explain the relative deviation of 22.5 %. In the absence of information on limiting ionic conductivities of taurate, sarcosinate, and prolinate, diffusivities at infinite dilution are not estimated for salts of these ions. At higher concentrations, salt diffusivities can be estimated\(^{18}\) by the additional use of viscosities, solvent partial molar volume, and molar density and mean ionic activity coefficients. As some of these properties are not readily available for the considered salts, such estimations are not further investigated in the present work.

**Diffusivity of N\(_2\)O in Aqueous Solutions of the Potassium Salt of Amino Acids.** The experimental determined diffusivities of N\(_2\)O in aqueous solutions of the potassium salt of taurine, sarcosine, L-proline, and glycine were measured and averaged over at least three measurements at temperatures from (293 to 333) K and salt concentrations ranging from (1.0 to 3.0) mol·dm\(^{-3}\). The results are presented in Table 4. The experimental uncertainty is estimated to be 10 %, and the reproducibility is within 2 %. The critical (De)\(^2\)Sc number was found to be about 80 for these systems.

Several investigators have correlated the diffusivity of an absorbed gas in an aqueous salt solution to the viscosity or the salt concentration.\(^{19–23}\) Ratcliff & Holdcroft,\(^{19}\) Brilman et al.,\(^{22}\) and Kumar et al.\(^{23}\) correlated the diffusivities to the viscosities by a modified Stokes–Einstein relation

\[ \frac{D_{\text{N2O in H2O}}}{D_{\text{N2O in H2O}}} = \left( \frac{\mu_{\text{H2O}}}{\mu_{\text{SAA}}} \right)^{\alpha} = \text{constant} \]  

(11)

where \( \alpha \) is a constant. Ratcliff & Holdcroft\(^{19}\) found this value to be 0.637, whereas Brilman et al.\(^{22}\) found it to be 0.6. Kumar et al.\(^{23}\) found the value to be 0.74. An \( \alpha \) value of 0.6 was found to predict the diffusivities within an average and maximum absolute relative deviation of 5.6 % and 15.1 %, respectively. The viscosities were taken from Kumar et al.,\(^{23}\) van Holst et al.,\(^{24}\) Portugal et al.,\(^{25}\) and Lide.\(^{26}\) The diffusivity of N\(_2\)O in H\(_2\)O was taken from Versteeg & van Swaaij.\(^{27}\) The correlation is presented in Figure 2 for the aqueous solutions of the potassium salt of the amino acids considered. Diffusivities of CO\(_2\) in aqueous solutions of the potassium salt of amino acids can be estimated by the N\(_2\)O analogy.\(^{27}\)

Kumar et al.\(^{23}\) reported diffusivities of N\(_2\)O in aqueous solutions of the potassium salt of taurine at 293 K using a diaphragm cell technique. The results\(^{23}\) are compared to the results from the present work at 293.15 K in Figure 3 and the estimated values from the modified Stokes–Einstein relation at 293.15 K. The values from the present work have an absolute average and maximum deviation of 4.6 % and 6.1 % from the estimated values at 293.15 K, respectively. The results of Kumar et al.\(^{23}\) deviate by 4.8 % and 10.5 % from the estimated values.

**Conclusion**

Diffusivities of the potassium salt of taurine, sarcosine, L-proline, and glycine have been determined in aqueous solutions at temperatures from (293 to 368) K and salt concentrations...
Concentrations ranging from (1.0 to 3.0) mol·dm\(^{-3}\) determined at temperatures from (293 to 333) K and salt concentrations ranging from (1.0 to 3.0) mol·dm\(^{-3}\). The diffusivities can be estimated using a modified Stokes–Einstein relation:

\[
D = \frac{kT}{6\pi\eta R}
\]

where \(D\) is the diffusion coefficient, \(k\) is the Boltzmann constant, \(T\) is the temperature, \(\eta\) is the viscosity, and \(R\) is the radius of the solute molecule.

The authors also wish to acknowledge H. F. G. Moed and B. Knaken for the construction of the experimental setup and P. H. M. Feron and F. Geuzebroek for their valuable discussions regarding the experimental technique and results.

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