

Physicochemical Properties of Several Aqueous Potassium Amino Acid Salts

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In memory of Jacco van Holst who suddenly passed away at the young age of 28

Aqueous amino acid salt solutions might be an attractive alternative over traditional alkanolamines in the removal of CO₂ from flue gases especially because of their oxidative stability and negligible volatility. The density and viscosity of the aqueous solutions of the potassium salts of the following amino acids have been measured: β-alanine, 6-aminohexanoic acid, L-arginine, L-aspartic acid, L-glutamic Acid, DL-methionine, L-phenylalanine, L-proline, and sarcosine. Data are reported in the temperature range of (25 to 60) °C and concentration range of (0.25 to 3.5) mol·L⁻¹. The data could be correlated accurately with relatively simple relations. The physical solubility of N₂O in the same amino acid salt solutions was also measured at a temperature of 298 K and a concentration of 0.5 mol·L⁻¹ of the amino acid salt. Using the N₂O/CO₂ analogy on one hand and Schumpe's method on the other, the physical solubility of CO₂ in these solutions was estimated. Both methods yielded results very close to each other.

Introduction

One of the most alarming global environmental problems of today is the increase of global temperatures. This problem is most likely caused by the increasing atmospheric carbon dioxide concentration due to the burning of fossil fuels for power generation. To minimize these effects, the carbon dioxide emissions from combustion and gasification processes in power plants have to be decreased by efficiency improvements and carbon dioxide capture.

The removal of acid gases such as carbon dioxide, H₂S, or COS by absorption in aqueous alkanolamine solutions is widely used in the chemical industry.¹ This process is based on the reversible chemical absorption of CO₂ using an acid–base reaction. Typical alkanolamines used for this process are monoethanolamine, diethanolamine, or *N*-methyldiethanolamine. A problem with the use of alkanolamines for the CO₂ removal from flue gas is that they degrade as a result of long exposure or repeated use because of side reactions with CO₂, oxygen, and other contaminants. Because the desire to separate CO₂ from flue gas streams is gaining momentum as a result of environmental concerns, there is an urgent need to develop new solvent systems that are stable in the presence of oxygen. Amino acids have the same functional groups as alkanolamines and can be expected to behave similarly toward carbon dioxide but do not deteriorate in the presence of oxygen.² Kumar et al.³ have proven the first assumption to be valid. An example of a possibly suitable amino acid is sarcosine, which is shown in Figure 1.

An additional advantage of amino acids is the possibility of adding a salt function. The carboxylate group can be neutralized with potassium hydroxide, to produce the potassium salt of glycine. This salt function ensures the nonvolatility of the substance,⁴ which is helpful when working at stripper conditions (lowered pressure and elevated temperature). A third advantage is their high surface tension, which makes them interesting for gas–liquid membrane applications. In contrast with MEA, which

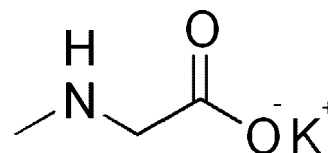


Figure 1. Potassium salt of sarcosine.

can only be used in combination with expensive membranes due to wetting problems, they can be used in conjunction with simple polyolefin membranes, like polypropylene.

Design of gas–liquid contactors, used in acid gas treating processes, requires information on mass-transfer coefficients, interfacial area, reaction kinetics, and physicochemical properties such as density, viscosity of the solvents, and the solubility of the relevant gases in the solvents. These data are also necessary to deduce chemical reaction kinetics from absorption rate experiments. The problem in measuring the physical solubility of CO₂ in reactive absorbents is that it cannot be measured directly because it will react with the absorbent. Therefore, it is necessary to measure this parameter indirectly, by using a gas that does not react with the absorbent but is chemically similar to CO₂, namely N₂O. The use of the nonreactive N₂O as a replacement for CO₂ to be able to measure the physical solubility has been introduced by Laddha et al.⁵ (N₂O/CO₂ analogy) and has been widely accepted since then.

This paper reports viscosity and density of water + amino acid salts and the solubility of N₂O in various amino acid salt solutions at 298 K.

Experimental Section

The amino acids dissolved in water exist as zwitterions with the amino group completely protonated. The ionic equilibrium of the amino acids exists as follows



Addition of KOH to the amino acid solution will result in the deprotonation of the zwitterion into the deprotonated amino

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acid salt solution KO_2CRNH_2 . This deprotonation step is necessary to make the amino group reactive toward CO_2 .

The potassium salts of the selected amino acids were prepared by neutralizing the amino acid, with purities of at least 98 % (Sigma Aldrich) dissolved in demineralized water, with an equimolar amount of potassium hydroxide (Riedel-de Haën, ≥ 85 %, pellets). The total amount of impurities in the potassium hydroxide pellets other than water was around 1 % (mostly as K_2CO_3 , product analysis by Riedel-de Haën). The actual purity of the KOH pellets was determined by acid titration. This number was used for the determination of the weight of KOH pellets required for the deprotonation of the amino acids. The actual concentration of the aqueous amino acid salt solutions was measured potentiometrically with a standard $1 \text{ mol}\cdot\text{L}^{-1}$ hydrochloric acid solution (Merck). The experimentally determined concentrations were within 0.5 %.

Density. The densities of the potassium amino acid salt solutions were measured at different temperatures (25, 30, 40, 50, and 60) °C, and at concentrations between (0.25 and 3.5) $\text{mol}\cdot\text{L}^{-1}$. A commercial density meter (DMA 58, Anton Paar GmbH) was used.

Viscosity. The viscosity was measured using a Ubbelohde-type viscometer (size 0). The viscometer was immersed in a large water bath. The temperature was controlled with a constant-temperature circulator to within ± 0.05 K. From the efflux time, the kinematic viscosity was calculated from the equation

$$\nu = Ct \quad (2)$$

where ν is the kinematic viscosity; C is a constant specific to the viscometer; and t is the efflux time. End effect corrections were neglected in the calculation of the kinematic viscosity. The constant, C , in eq 2 was determined using distilled water and was $1.089 \cdot 10^{-2} \text{ mm}^2 \cdot \text{s}^{-2}$. Each reported measurement was the average of at least three runs with a maximum deviation in the kinematic viscosity of approximately ± 0.05 %.

Solubility. The physical solubility of N_2O in the aqueous amino acid solutions was measured in a thermostatted glass vessel with a volume of $1.539 \cdot 10^{-3} \text{ m}^3$ which was filled with a known volume ($1 \cdot 10^{-3} \text{ m}^3$) of solution. In each experiment, the solution was degassed by evacuation of the equilibrium vessel, and the contents were held under vacuum until there were no bubbles coming out of the liquid. Then the reactor was closed, and 30 min to 1 h was allowed for the temperature to equilibrate and vapor-liquid equilibrium was established. The equilibrium pressure was recorded by a pressure transducer (Heise DXD), which was connected to a computer with a RS232 connection. The pressure range of the transducer was (0 to 138) kPa with a full-scale uncertainty of ± 28 Pa. N_2O was then fed to the vessel until an arbitrary pressure was reached (with $P \leq 10^5$ Pa). Then the vessel was closed, and a gas-inducing stirrer was started to agitate the liquid. The pressure change in the gas phase was continuously recorded by the transducer and computer. For all experiments, it took about 10 min to reach equilibrium, but to be on the safe side, another 20 min was allowed. If the pressure was stable during this 20 min, it was assumed that equilibrium was reached. The physical solubility could be calculated, based on Henry's law according to

$$k_m = k_{\text{He}} \cdot RT = \frac{(P^{\text{int}} - P^{\text{eq}})}{P^{\text{eq}}} \cdot \frac{V_g}{V_l} \quad (3)$$

where k_m is the dimensionless physical solubility; k_{He} is the Henry coefficient; R is the gas constant; P^{int} is the initial pressure just after admittance of the gas; P^{eq} is the equilibrium pressure; and V_g and V_l are the volumes of the gas and liquid phase, respectively. The dimensionless physical solubility is defined as the ratio of the liquid-phase concentration to the gas-phase concentration of the solute at equilibrium conditions

$$k_m = \left(\frac{c_l}{c_g} \right) \quad (4)$$

with c_l being the concentration of the solute in the liquid phase and c_g the concentration of the solute in the gas phase.

Results and Discussion

Density. The measured density of the aqueous salt solutions studied is given in Table 1. The estimated uncertainty of the density measurements is $\pm 0.001 \text{ kg}\cdot\text{L}^{-1}$.

The experimental densities were fitted to a polynomial function that is temperature and concentration dependent, according to

$$\rho = k_1 T + k_2 c + k_3 \quad (5)$$

where ρ is the density in $\text{kg}\cdot\text{L}^{-1}$; T is the temperature in K; c is the concentration in $\text{mol}\cdot\text{L}^{-1}$; and k_i are the polynomial coefficients, which are presented in Table 2. The maximum deviation of the densities calculated using eq 5 and the data in Table 2 was never higher than 0.41 % from the experimental densities (the standard deviations are given in Table 2). An example of the polynomial fit for the potassium salt of β -alanine is given in Figure 2. This figure is illustrative for all other aqueous amino acid salt solutions.

Viscosity. The measured values of the viscosity of the aqueous solutions of the potassium salts of the amino acids are given in Table 3.

The effect of temperature on the viscosity of fluids can be expressed by an Arrhenius-type eq 6

$$\eta = \eta_0 \exp\left(\frac{E_a}{RT}\right) \quad (6)$$

where η is the viscosity of the solution; η_0 is a pre-exponential factor; and E_a is the activation energy of flow.

Variation of viscosity of fluids with concentration can be described by an exponential-type relationship⁶

$$\eta = \eta'_0 \exp(k_4 c) \quad (7)$$

where η'_0 is a pre-exponential factor and k_4 is a constant. A temperature- and concentration-dependent relation is formed, when eqs 6 and 7 are combined

$$\eta = k_1 \exp\left[\frac{k_2 \exp(k_3 c)}{RT}\right] \exp(k_4 c) \quad (8)$$

Here the activation energy of flow (E_a) has been replaced by an exponential-type equation dependent on the concentration, and the pre-exponential factors η_0 and η'_0 have been lumped together into k_1 . The experimental viscosities were fitted to eq 8, and the coefficients k_i are presented in Table 4. The maximum deviation from the experimental viscosities was never higher than 2.5 % (the standard deviations are given in Table 4). An example of the polynomial fit for the potassium salt of β -alanine is given in Figure 3. This figure is illustrative for all other aqueous amino acid salt solutions.

Solubility. The experimental setup and procedure were validated by measuring the physical solubility of N_2O in water at 298 K. The measured physical N_2O solubility in water was $k_{m,\text{N}_2\text{O}} = (0.591 \pm 0.028)$ which is in good agreement with the solubility data reported in the literature (for example, Versteeg and van Swaaij⁷ report a solubility of $k_{m,\text{N}_2\text{O}} = 0.618$).

The physical solubility of N_2O was measured for aqueous solutions of $0.5 \text{ mol}\cdot\text{L}^{-1}$ of the potassium salts of 6-aminohexanoic acid, β -alanine, DL-methionine, L-arginine, L-aspartic acid, L-glutamic acid, L-phenylalanine, L-proline, and sarcosine at 298 K. All measurements were done in triplicate at partial N_2O pressures

Table 1. Density of Aqueous Solutions of Potassium Salts

$c_2/\text{mol}\cdot\text{L}^{-1}$	$T/^\circ\text{C}$				
	25 °C	30 °C	40 °C	50 °C	60 °C
	$\rho/\text{kg}\cdot\text{L}^{-1}$				
	potassium salt of β -alanine				
0.496	1.026	1.024	1.020	1.017	1.011
1.003	1.053	1.053	1.048	1.043	1.038
1.492	1.079	1.078	1.074	1.070	1.065
2.019	1.107	1.106	1.100	1.096	1.090
2.492	1.133	1.131	1.126	1.121	1.116
2.964	1.157	1.155	1.150	1.145	1.140
3.417	1.181	1.179	1.174	1.169	1.162
	potassium salt of 6-aminohexanoic acid				
0.497	1.024	1.022	1.018	1.014	1.009
1.002	1.049	1.048	1.044	1.038	1.034
1.429	1.072	1.070	1.067	1.061	1.057
1.982	1.098	1.095	1.089	1.084	1.078
2.458	1.119	1.117	1.111	1.105	1.098
2.942	1.141	1.139	1.132	1.126	1.118
	potassium salt of L-aspartic acid				
0.255	1.029	1.027	1.024	1.020	1.015
0.470	1.059	1.057	1.053	1.049	1.045
0.708	1.089	1.087	1.083	1.079	1.074
0.979	1.119	1.117	1.113	1.109	1.105
1.208	1.147	1.145	1.141	1.137	1.133
1.449	1.176	1.173	1.169	1.166	1.162
	potassium salt of L-proline				
0.507	1.028	1.027	1.023	1.019	1.014
1.010	1.059	1.057	1.053	1.048	1.043
1.523	1.089	1.087	1.082	1.077	1.072
2.010	1.118	1.115	1.110	1.105	1.100
2.521	1.146	1.144	1.138	1.133	1.127
2.983	1.171	1.168	1.162	1.156	1.150
3.448	1.197	1.194	1.188	1.182	1.176
	potassium salt of L-glutamic acid				
0.248	1.029	1.027	1.024	1.019	1.017
0.493	1.059	1.057	1.054	1.049	1.047
0.733	1.088	1.086	1.083	1.078	1.076
1.000	1.117	1.116	1.112	1.107	1.105
1.240	1.145	1.143	1.139	1.134	1.132
1.469	1.173	1.171	1.167	1.162	1.160
	potassium salt of sarcosine				
0.510	1.024	1.023	1.019	1.015	1.010
1.010	1.051	1.049	1.045	1.041	1.036
1.534	1.077	1.074	1.070	1.065	1.060
2.019	1.103	1.100	1.096	1.091	1.085
2.445	1.124	1.121	1.117	1.111	1.106
2.987	1.150	1.148	1.143	1.138	1.132
	potassium salt of DL-methionine				
0.252	1.016	1.014	1.010	1.006	1.003
0.503	1.033	1.032	1.027	1.023	1.020
0.732	1.051	1.049	1.044	1.039	1.037
0.994	1.067	1.065	1.060	1.055	1.053
1.252	1.084	1.083	1.077	1.072	1.069
1.479	1.100	1.098	1.093	1.087	1.085
	potassium salt of L-arginine				
0.238	1.021	1.019	1.016	1.011	1.007
0.479	1.043	1.041	1.037	1.032	1.027
0.725	1.064	1.062	1.058	1.053	1.048
0.971	1.087	1.085	1.080	1.075	1.069
	potassium salt of L-phenylalanine				
0.250	1.015	1.014	1.010	1.005	1.001
0.496	1.032	1.031	1.027	1.022	1.017
0.745	1.051	1.049	1.044	1.039	1.034
0.996	1.067	1.065	1.060	1.055	1.049
1.226	1.084	1.081	1.077	1.071	1.066
1.453	1.100	1.097	1.092	1.087	1.081

around 1 bar, and the results are presented in Table 5. The uncertainty in the measurements was 0.002 [–].

Now that the N_2O physical distribution coefficients are known, a way to correlate them to the physical CO_2 distribution

Table 2. Polynomial Coefficients for Density (Equation 5)

potassium salt of amino acid	$k_1 \cdot 10^4$ $\text{kg}\cdot\text{L}^{-1}\cdot\text{K}^{-1}$	$k_2 \cdot 10^2$ $\text{kg}\cdot\text{mol}^{-1}$	k_3 $\text{kg}\cdot\text{L}^{-1}$	σ $\text{kg}\cdot\text{L}^{-1}$
β -alanine	-4.837	5.238	1.146	0.0008
6-aminohexanoic acid	-5.363	4.633	1.165	0.0019
L-arginine	-4.761	8.774	1.143	0.0007
L-aspartic acid	-3.973	12.14	1.119	0.0014
L-glutamic acid	-3.780	11.62	1.114	0.0011
DL-methionine	-4.361	6.714	1.130	0.0009
L-phenylalanine	-4.857	6.837	1.144	0.0007
L-proline	-5.258	5.602	1.160	0.0015
sarcosine	-4.862	5.000	1.146	0.0009

coefficients is needed. For alkanolamines, it is well-known that the N_2O analogy^{5,8} is a proficient way to estimate the physical solubility of CO_2 from physical N_2O solubility data. N_2O is similar to CO_2 with regard to configuration, molecular volume, and electronic structure, but does not react with the absorbents. The CO_2 - N_2O analogy states that the ratio of the physical solubilities of CO_2 and N_2O in water at identical partial pressure is the same as the ratio of the physical solubilities of both gases in the amino acid salt solution

$$\left(\frac{k_{m,\text{CO}_2}}{k_{m,\text{N}_2\text{O}}}\right)_{\text{AmA}} = \left(\frac{k_{m,\text{CO}_2}}{k_{m,\text{N}_2\text{O}}}\right)_{\text{H}_2\text{O}} = C_1 \quad (9)$$

where C_1 is the ratio between the dimensionless physical solubilities of CO_2 and N_2O in water. C_1 can be calculated by the following temperature-dependent relation⁷

$$C_1 = 3.04 \exp\left(\frac{-240}{T/\text{K}}\right) \quad (10)$$

Based on the $\text{N}_2\text{O}/\text{CO}_2$ analogy, the physical distribution coefficient of CO_2 in amino acid salt solutions can thus be derived from the measured distribution coefficient of N_2O in these solutions using

$$k_{m,\text{CO}_2,\text{AmA}} = C_1 \cdot k_{m,\text{N}_2\text{O},\text{AmA}} = 3.04 \cdot \exp\left(\frac{-240}{T/\text{K}}\right) \cdot k_{m,\text{N}_2\text{O},\text{AmA}} \quad (11)$$

The estimated physical distribution coefficients of CO_2 as predicted by eq 11 are given in Table 6.

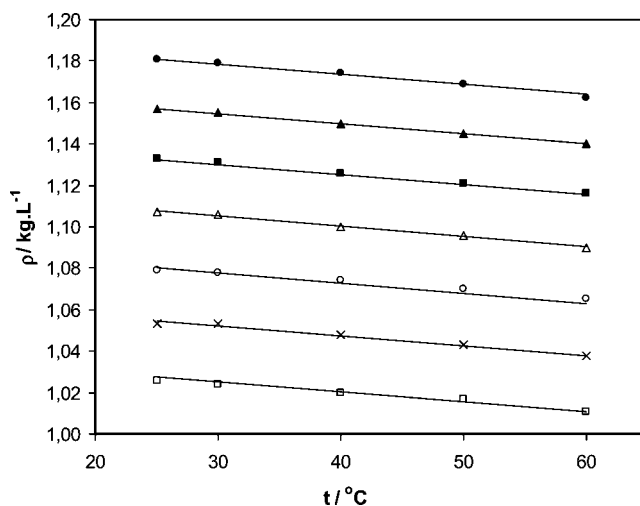


Figure 2. Liquid densities in $\text{kg}\cdot\text{L}^{-1}$ of aqueous potassium salt of β -alanine at different temperatures and concentrations. \square , 0.496 $\text{mol}\cdot\text{L}^{-1}$; \times , 1.003 $\text{mol}\cdot\text{L}^{-1}$; \circ , 1.492 $\text{mol}\cdot\text{L}^{-1}$; \triangle , 2.019 $\text{mol}\cdot\text{L}^{-1}$; \blacksquare , 2.492 $\text{mol}\cdot\text{L}^{-1}$; \blacktriangle , 2.964 $\text{mol}\cdot\text{L}^{-1}$; \bullet , 3.417 $\text{mol}\cdot\text{L}^{-1}$. Continuous lines give predictions according eq 5.

Table 3. Dynamic Viscosity of Aqueous Potassium Salt Solutions at Concentration c_s

$c_s/\text{mol}\cdot\text{L}^{-1}$	$T/^\circ\text{C}$				
	25 °C	30 °C	40 °C	50 °C	60 °C
	mPa·s				
	potassium salt of β -alanine				
0.496	1.050	0.944	0.762	0.638	0.546
1.003	1.230	1.097	0.892	0.744	0.634
1.492	1.453	1.293	1.049	0.871	0.737
2.019	1.759	1.548	1.250	1.027	0.866
2.492	2.131	1.881	1.497	1.223	1.022
2.964	2.609	2.293	1.808	1.464	1.217
3.417	3.245	2.832	2.209	1.770	1.459
	potassium salt of 6-aminohexanoic acid				
0.497	1.191	1.058	0.854	0.706	0.599
1.002	1.608	1.418	1.126	0.913	0.762
1.429	2.152	1.882	1.497	1.199	0.987
1.982	3.091	2.669	2.035	1.608	1.307
2.458	4.445	3.796	2.850	2.211	1.768
2.942	6.601	5.573	4.112	3.145	2.458
	potassium salt of L-aspartic acid				
0.255	0.992	0.887	0.725	0.613	0.525
0.470	1.104	0.998	0.816	0.693	0.593
0.708	1.239	1.115	0.914	0.771	0.663
0.979	1.419	1.269	1.041	0.865	0.743
1.208	1.615	1.450	1.185	0.994	0.836
1.449	1.856	1.667	1.353	1.142	0.957
	potassium salt of L-proline				
0.507	1.107	0.986	0.799	0.664	0.565
1.010	1.400	1.242	0.992	0.816	0.685
1.523	1.811	1.590	1.251	1.017	0.846
2.010	2.365	2.061	1.599	1.286	1.061
2.521	3.163	2.721	2.076	1.636	1.331
2.983	4.112	3.516	2.664	2.068	1.658
3.448	5.732	4.837	3.587	2.767	2.181
	potassium salt of L-glutamic acid				
0.248	1.011	0.908	0.744	0.623	0.535
0.493	1.163	1.041	0.854	0.713	0.611
0.733	1.344	1.199	0.982	0.817	0.699
1.000	1.579	1.404	1.143	0.953	0.809
1.240	1.844	1.644	1.336	1.101	0.933
1.469	2.194	1.962	1.570	1.295	1.089
	potassium salt of sarcosine				
0.510	1.018	0.913	0.748	0.627	0.538
1.019	1.185	1.060	0.876	0.759	0.614
1.534	1.408	1.252	1.005	0.833	0.703
2.019	1.663	1.466	1.180	0.968	0.811
2.445	1.959	1.707	1.356	1.098	0.919
2.987	2.493	2.185	1.721	1.395	1.115
	potassium salt of DL-methionine				
0.252	0.987	0.884	0.722	0.608	0.519
0.503	1.109	0.987	0.806	0.675	0.575
0.732	1.263	1.125	0.909	0.754	0.640
0.994	1.425	1.264	1.019	0.840	0.708
1.252	1.654	1.478	1.161	0.952	0.799
1.479	1.883	1.657	1.316	1.071	0.893
	potassium salt of L-arginine				
0.238	1.026	0.917	0.750	0.630	0.539
0.479	1.215	1.098	0.886	0.738	0.632
0.725	1.431	1.276	1.035	0.856	0.723
0.971	1.882	1.553	1.244	1.019	0.848
	potassium salt of L-phenylalanine				
0.250	1.059	0.938	0.769	0.641	0.549
0.496	1.224	1.092	0.884	0.738	0.631
0.746	1.441	1.279	1.032	0.852	0.720
0.996	1.695	1.495	1.194	0.980	0.823
1.227	2.030	1.782	1.410	1.150	0.963
1.453	2.483	2.169	1.702	1.377	1.139

It should be noted though that the absorbents used in this study are not alkanolamines but amino acid salt solutions. The presence of salts can initiate salting out effects at higher ionic strengths, which means that the gas solubility in the salt solution is lower than compared to what it would be if there were no salts present in the solution. This effect of salt concentration,

c_s , on the gas solubility, c_g , of a barely soluble gas as compared to that in pure water ($c_{g,\text{H}_2\text{O}}$) was described by Sechenov⁹ in the following form

$$\log\left(\frac{c_{g,\text{H}_2\text{O}}}{c_{g,\text{L}}}\right) = Kc_s \quad (12)$$

In 1993, Schumpe¹⁰ proposed an empirical model which could be used to estimate the Sechenov constant, K , for salt solutions at different ionic strengths, and could even be extended to mixed electrolyte solutions. Schumpe proposed the following relation

$$\log\left(\frac{c_{g,\text{H}_2\text{O}}}{c_{g,\text{L}}}\right) = \sum (h_i + h_g)c_i \quad (13)$$

where h_i is the ion-specific parameter; h_g is the gas-specific parameter; and c_i is the concentration of ion i . In 1996, Weissenberger and Schumpe¹¹ extended the model to a wider temperature range. The temperature dependency was found gas-specific, and the gas-specific constant was assumed to be a linear function of the temperature:¹¹

$$h_g = h_{g,0} + h_T(T/\text{K} - 298.15) \quad (14)$$

Schumpe's method can be used to justify the use of the $\text{N}_2\text{O}/\text{CO}_2$ analogy as presented in eq 9 in these salt solutions. For the potassium amino acid salt solutions as used in this work, eq 13 can be written as

$$\frac{k_{m,\text{N}_2\text{O},\text{H}_2\text{O}}}{k_{m,\text{N}_2\text{O},\text{AmA}}} = 10^{(h_{\text{K}^+} + h_{\text{AmA}^-} + 2h_{\text{N}_2\text{O},g})c_s} \quad (15)$$

and

$$\frac{k_{m,\text{CO}_2,\text{H}_2\text{O}}}{k_{m,\text{CO}_2,\text{AmA}}} = 10^{(h_{\text{K}^+} + h_{\text{AmA}^-} + 2h_{\text{N}_2\text{O},g})c_s} \quad (16)$$

Both relations can be combined to give

$$\left(\frac{k_{m,\text{CO}_2}}{k_{m,\text{N}_2\text{O}}}\right)_{\text{AmA}} = \left(\frac{k_{m,\text{CO}_2}}{k_{m,\text{N}_2\text{O}}}\right)_{\text{H}_2\text{O}} \cdot 10^{2(h_{\text{N}_2\text{O},g} - h_{\text{CO}_2,g})c_s} = C_1 \cdot 10^{2(h_{\text{N}_2\text{O},g} - h_{\text{CO}_2,g})c_s} \quad (17)$$

The constant C_1 in eq 17 should match that of eqs 9 and 10, while the last term in eq 17 can be seen as a kind of corrective term for the $\text{N}_2\text{O}/\text{CO}_2$ analogy for these salt solutions. For the conditions in this work ($T = 298 \text{ K}$, $c_s = 0.5 \text{ mol}\cdot\text{L}^{-1}$), this corrective term can be calculated to be 1.02 (see Table 7 for the Schumpe model parameters), which seems to justify the use of the $\text{N}_2\text{O}/\text{CO}_2$ analogy in this case.

With the measured N_2O solubility data of the several amino acid salt solutions given in Table 5 and the Schumpe model parameters given in Table 7, together with the N_2O solubility in water given by Versteeg and Van Swaaij,⁷ the ion-specific parameters for the anions h_i can be estimated. The solubility of N_2O in water is given by the following relation

$$k_{\text{He},\text{N}_2\text{O}}/\text{mol}\cdot\text{m}^{-3}\cdot\text{Pa}^{-1} = 1.17 \cdot 10^{-7} \exp\left(\frac{2284}{T/\text{K}}\right) \quad (18a)$$

where k_{He} is the Henry's law constant in $\text{mol}\cdot\text{m}^{-3}\cdot\text{Pa}^{-1}$. Using the dimensionless distribution coefficient k_m as applied in the present work, eq 18a gives

$$k_{m,\text{N}_2\text{O}} = 1.17 \cdot 10^{-7} \cdot RT/(J\cdot\text{mol}^{-1}) \exp\left(\frac{2284}{T/\text{K}}\right) \quad (18b)$$

The resulting Schumpe parameters for h_i are given in Table 7. It should be noted that for each amino acid salt solution the

Table 4. Coefficients for Viscosity (Equation 8)

potassium salt of amino acid	concentration range	$k_1 \cdot 10^3$	$k_2 \cdot 10^{-3}$	k_3	k_4	σ
	$\text{mol} \cdot \text{L}^{-1}$	$\text{mPa} \cdot \text{s}$	$\text{J} \cdot \text{mol}^{-1}$	$\text{L} \cdot \text{mol}^{-1}$	$\text{L} \cdot \text{mol}^{-1}$	$\text{mPa} \cdot \text{s}$
β -alanine	0.5 to 3.5	3.249	13.91	0.0947	-0.2513	0.013
6-aminohexanoic acid	0.5 to 3.0	1.354	16.22	0.1242	-0.3234	0.029
L-arginine	0.25 to 1	3.103	14.04	0.2747	-1.060	0.024
L-aspartic acid	0.25 to 1.5	2.890	14.10	0.0855	0.0159	0.010
L-glutamic acid	0.25 to 1.5	3.320	13.78	0.1441	-0.2608	0.012
DL-methionine	0.25 to 1.5	2.109	14.94	0.1178	-0.2626	0.001
L-phenylalanine	0.25 to 1.5	3.198	14.01	0.2035	-0.6571	0.014
L-proline	0.5 to 3.5	1.534	15.85	0.1071	-0.3025	0.031
sarcosine	0.5 to 3.0	2.950	14.15	0.1047	-0.3124	0.015

Table 5. Experimental Dimensionless Solubility of N_2O in Several Aqueous Potassium Amino Acid Salt Solutions at 298 K

potassium salt of amino acid	c_s	$k_{m,\text{N}_2\text{O}}$		
	$\text{mol} \cdot \text{L}^{-1}$	[-]		
β -alanine	0.496	0.521	0.524	0.523
6-aminohexanoic acid	0.502	0.509	0.506	0.504
L-arginine	0.503	0.478	0.479	0.479
L-aspartic acid	0.506	0.437	0.437	0.434
L-glutamic acid	0.492	0.444	0.447	0.441
DL-methionine	0.498	0.515	0.514	0.513
L-phenylalanine	0.498	0.513	0.511	0.513
L-proline	0.495	0.522	0.522	0.519
sarcosine	0.501	0.519	0.514	0.513

solubility was measured at only one concentration, $0.5 \text{ kmol} \cdot \text{m}^{-3}$, and temperature, 298 K, making the uncertainty in the estimated parameters h_i relatively high. Nevertheless, for the purpose of correlating the CO_2 solubility to the N_2O solubility in the salt solution at one concentration and temperature, it should be sufficient. With the ion-specific parameters known, the CO_2 solubility according to Schumpe's method can be estimated and is given in Table 8.

The data in Table 6 are well in line with the data in Table 8, with the largest difference of only 1.71 % for 6-aminohexanoic acid. This good agreement was already expected based on the value of the correction factor of 1.02 as determined in eq 16. Schumpe's method seems therefore a good way to estimate the solubility of N_2O in these solutions not only at other concentrations but also at other temperatures as used in this study.

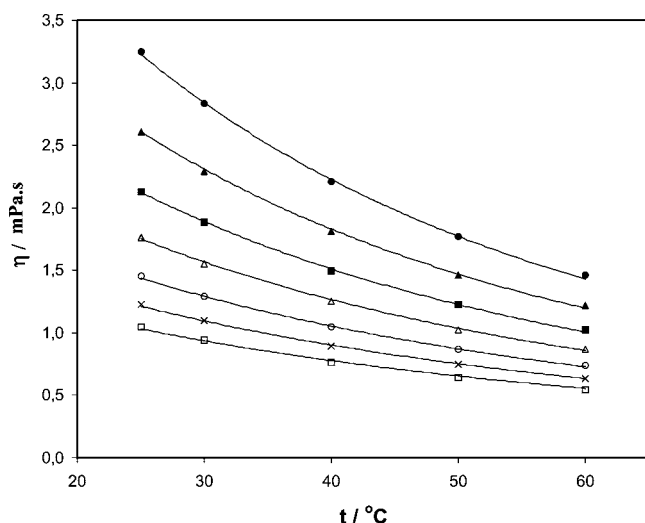


Figure 3. Dynamic viscosity of aqueous potassium salt of β -alanine at different temperatures and concentrations. \square , $0.496 \text{ mol} \cdot \text{L}^{-1}$; \times , $1.003 \text{ mol} \cdot \text{L}^{-1}$; \circ , $1.492 \text{ mol} \cdot \text{L}^{-1}$; \triangle , $2.019 \text{ mol} \cdot \text{L}^{-1}$; \blacksquare , $2.492 \text{ mol} \cdot \text{L}^{-1}$; \blacktriangle , $2.964 \text{ mol} \cdot \text{L}^{-1}$; \bullet , $3.417 \text{ mol} \cdot \text{L}^{-1}$. Continuous lines give predictions according eq 8.

Table 6. Estimated Solubility of CO_2 in Several Aqueous Potassium Salts of Amino Acids at 298 K by $\text{N}_2\text{O}/\text{CO}_2$ Analogy

potassium salt of amino acid	c_s	estimated CO_2 distribution coefficient k_m
	$\text{mol} \cdot \text{L}^{-1}$	[-]
β -alanine	0.496	0.709
6-aminohexanoic acid	0.502	0.687
L-arginine	0.503	0.651
L-aspartic acid	0.506	0.592
L-glutamic acid	0.492	0.603
DL-methionine	0.498	0.698
L-phenylalanine	0.498	0.697
L-proline	0.495	0.718
sarcosine	0.501	0.708

Table 7. Schumpe Model Parameters¹¹

cation	h_i	gas	h_G	$10^3 \cdot h_T$	temp. range
	$\text{m}^3 \cdot \text{kmol}^{-1}$		m^3/kmol	$\text{m}^3 \cdot \text{kmol}^{-1} \cdot \text{K}^1$	K
K^+	0.0922	N_2O	-0.0085	-0.479	273 to 313
		CO_2	-0.0172	-0.338	273 to 313

Table 8. Estimated Ion-Specific Schumpe Parameters and Estimated CO_2 Solubility m for Several Amino Acid Salts at 298 K

potassium salt of amino acid	c_s	h_i	estimated CO_2 distribution coefficient
	$\text{kmol} \cdot \text{m}^{-3}$	$\text{m}^3 \cdot \text{kmol}^{-1}$	k_m
β -alanine	0.496	0.0715	0.721
6-aminohexanoic acid	0.502	0.0968	0.699
L-arginine	0.503	0.1452	0.660
L-aspartic acid	0.506	0.2236	0.602
L-glutamic acid	0.492	0.2162	0.612
DL-methionine	0.498	0.0846	0.709
L-phenylalanine	0.498	0.0875	0.707
L-proline	0.495	0.0740	0.719
sarcosine	0.501	0.0819	0.711

Conclusions

The viscosity and density of amino acid salt solutions has been determined in the temperature range of (25 to 60) $^\circ\text{C}$ and concentration range of (0.25 to 3.5) $\text{mol} \cdot \text{L}^{-1}$. It was possible to accurately correlate the density for all solutions with the following simple relation: $\rho = k_1 T + k_2 c + k_3$. The viscosity was measured in the same temperature and concentration range. The experimental data could be represented by the following relation: $\eta = k_1 \exp[k_2 \exp(k_3 c) / RT] \exp(k_4 c)$. The predicted viscosity never differed by more than 2.5 % from the experimentally measured values.

Besides this, also the solubility of N_2O was measured in the same amino acid salt solutions; however, only for one concentration ($0.5 \text{ mol} \cdot \text{L}^{-1}$) and one temperature (298 K). By using either the $\text{N}_2\text{O}/\text{CO}_2$ analogy or the method of Schumpe, it was possible to predict the physical solubility of CO_2 in these amino acid salt solutions. Both methods yielded the same results within

1.7 %. Schumpe's method therefore seems suitable to be used for the prediction of the physical solubility of CO₂ in these solutions at other concentrations and temperatures.

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