

Scientific paper

# Osmotic Coefficient of Aqueous Solutions of Cyclohexylsulfamic Acid at the Freezing Point of Solutions

Marija Bešter-Rogač,<sup>1</sup> Cveto Klofutar<sup>2</sup>  
and Darja Rudan-Tasič<sup>2,3,\*</sup>

<sup>1</sup> Faculty of Chemistry and Chemical Technology, University of Ljubljana, SI-1000 Ljubljana, Slovenia.

<sup>2</sup> Biotechnical Faculty, University of Ljubljana, SI-1000 Ljubljana, Slovenia.

<sup>3</sup> Krog-MIT d. o. o., Tržaška 43, SI-1000 Ljubljana, Slovenia.

\* Corresponding author: E-mail: darja.rudan.tasic@bf.uni-lj.si  
darja.rudan.tasic@krog-mit.com

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## Abstract

The osmotic coefficient of aqueous solutions of cyclohexylsulfamic acid was determined by freezing point measurements up to the molality 0.65 mol kg<sup>-1</sup>. The osmotic coefficients were fitted to the *Pitzer* equation, and ion interaction parameters  $\alpha_1$ ,  $\beta^{(0)}$  and  $\beta^{(1)}$  were evaluated. The mean ion activity coefficient of the solute was calculated, and the non-ideal behaviour of the system investigated was characterized by calculation of the excess *Gibbs* energy of solution, as well as the respective partial molar functions of solute and solvent. The partial molar excess *Gibbs* energy of the solute is negative, like the excess *Gibbs* energy of its solution, while the partial molar excess *Gibbs* energy of the solvent is positive and increases with increasing concentration of the solute. The solvation ability of water was calculated from the difference between the *Gibbs* energy of solution of water in solution and that of pure water, and found to be positive and small for the solute investigated, throughout the concentration range studied.

**Keywords:** Osmotic coefficient, mean ion activity coefficient, cyclohexylsulfamic acid, aqueous solution

## 1. Introduction

The use of artificial sweeteners, which show greatly enhanced sweetness compared with sucrose and have very low caloric value, is steadily increasing.<sup>1</sup> Non-nutritive sweeteners represent a large commercial market in the field of pharmaceutical compounds and bulk chemicals. Among them cyclohexylsulfamic acid and its sodium or calcium salts are one of the most widely used artificial sweetening agents in foods, beverages and pharmaceuticals.

Although no plausible explanation of the molecular mechanism of taste chemoreceptor has been found, several models were proposed to describe the sweetener-receptor interaction, e. g.<sup>2-4</sup> The exceptional role of water in the sweetness mechanism which takes into account the solute-solvent interaction in aqueous solutions of sweete-

ners, as well as the effect of sweet molecules on water structure was first recognized by *Mathlouthi* et al.<sup>5</sup> In the past many of solution properties of artificial sweeteners were used in interpreting the ease of accession of sweet molecules or ions to the receptor side. The hydrophilic and hydrophobic balance as well as steric factors in the molecules of sweeteners is thought to affect the mobility of water in the vicinity of the solute. From this point of view it is surprisingly enough, that the data about thermophysical and thermochemical properties of cyclohexylsulfamic acid (on the contrary of its sodium salt) are very scarce in the literature.<sup>6, 7, 8, 9, 10</sup> Therefore, we focused a part of our recent studies on cyclohexylsulfamic acid itself.<sup>11, 12, 13, 14</sup>

A number of ionic interaction models provide the simple and most coherent procedure for calculating the properties of electrolyte compounds, e. g. *Pitzer* model,<sup>15</sup>

the Bromley model,<sup>16, 17</sup> the NRTL model,<sup>18</sup> and the SIT model.<sup>19</sup> The Pitzer model yields an extended Debye-Hückel formula using a virial expansion to account for the ionic strength dependence of the short-range forces in binary and ternary ion interactions.

The purpose of this work was to obtain the Pitzer ion interaction parameters from osmotic coefficient data of aqueous solution of cyclohexylsulfamic acid at high concentration. The obtained parameters enable us to characterize the non-ideal behaviour of aqueous solutions of cyclohexylsulfamic acid by calculation of the excess Gibbs energy of the solution, as well as the difference in the Gibbs energy of solvation of water in the solution relative to pure water. In addition, this study represents a continuation of our previous work on this matter.<sup>11</sup>

## 2. Experimental

### 2.1. Materials

Commercially available cyclohexylsulfamic acid (HCy) was purchased from Sigma. The compound was used as delivered without further purification and stored in a desiccator over P<sub>2</sub>O<sub>5</sub>. The purity of the acid was checked by titration with sodium tetraborate and also by analysis of the elements C, H and N (Perkin Elmer, 2400 Series II CHNS/O Analyser); it was found to be at least 99.9 % purity.

### 2.2. Preparation of Solutions

The solutions investigated were prepared on a molal concentration scale (mol kg<sup>-1</sup>) by precise weighing with double distilled water, on a digital balance (Mettler Toledo, model AT201, Switzerland) that was accurate to within  $\pm 1 \times 10^{-5}$  g. Before use, the solutions were degassed by ultrasound (ultrasonic bath, Bandelin Sonorex, type TK 52, Berlin, Germany).

### 2.3. Freezing Point Measurements

The freezing point depression was measured with a Knauer cryoscopic unit (model 7312400000, equipped with a strip chart recorder, Knauer Model 733.41). The solvent and the solutions were supercooled and the formation of ice crystals was initiated by internal vibrations. The freezing-point depression was recorded as the difference in resistance of the thermistor,  $\Delta r$  (arbitrary scale) between solvent and solution. The reproducibility of these  $\Delta r$  measurements was better than 0.5 % of the measuring scale. The instrument was calibrated with sodium chloride solutions<sup>20</sup> of accurately known molality over the same freezing-point range as for the solutions investigated, and the calibration was checked before and after each run. The freezing-point data of the sodium chloride solution were smoothed using the Lagrange interpolation method. The

following relationship was obtained:  $\theta = k \cdot \Delta r$ , where  $\theta$  is the freezing point depression and  $k$  is the calibration constant. The overall accuracy of the freezing-point depression of sodium chloride solution was  $\pm 0.005$  K.

## 3. Results and Discussion

The freezing-point depression data,  $\theta$  (K), given in Table 1, were used to calculate the osmotic coefficient on molality basis,  $\phi_m$ , through the relation (1) where the higher terms were omitted due to the uncertainty of the osmotic coefficient:<sup>21</sup>

$$\phi_m = \frac{\Delta_{\text{fus}} H_{T_m}^{\circ}}{\nu m M_{\text{H}_2\text{O}} R T_m^2} \theta \quad (1)$$

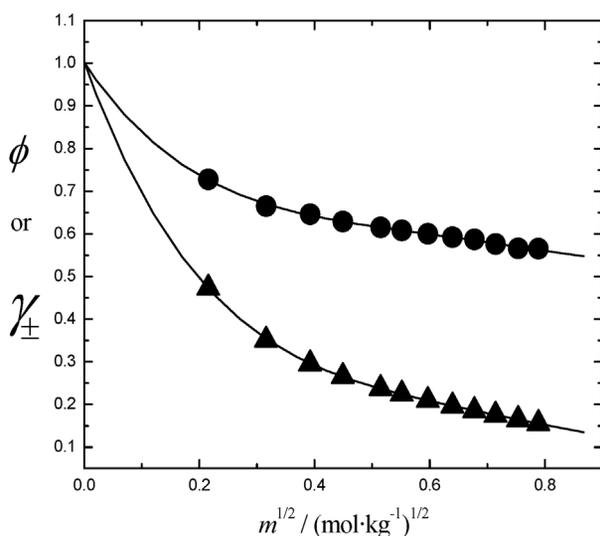
where  $M_{\text{H}_2\text{O}} R T_m^2 / \Delta_{\text{fus}} H_{T_m}^{\circ} = 1.8598$  K kg mol<sup>-1</sup>, the ideal freezing point depression constant,  $\nu$  is the number of ions into which the electrolyte dissociates, and  $m$  (mol kg<sup>-1</sup>) are the molalities of the aqueous solutions investigated. The osmotic coefficients  $\phi_m$  are given in Table 1 and shown in Fig. 1 where  $\phi_m$  is plotted against square root of the molality. The uncertainties of the osmotic coefficient,  $\delta\phi_m$ , were evaluated according to Eq. (2), and found to be smaller than 0.003.

$$(\delta\phi_m)^2 = \left( \frac{1}{1.8598\nu m} \right)^2 (\delta\theta)^2 \quad (2)$$

**Table 1.** The freezing point data, osmotic coefficients and mean ion activity coefficients of aqueous solutions of cyclohexylsulfamic acid at the freezing point of the solution.

$m$ (mol kg <sup>-1</sup> )	$\theta$ (K)	$\phi_m$	$-\ln \gamma_{\pm}$
0.0466	0.126	0.728	0.749
0.0999	0.247	0.665	1.047
0.1540	0.370	0.646	1.219
0.2017	0.472	0.629	1.327
0.2649	0.606	0.615	1.437
0.3043	0.688	0.608	1.495
0.3563	0.795	0.600	1.564
0.4090	0.901	0.592	1.629
0.4590	1.002	0.587	1.686
0.5102	1.094	0.576	1.743
0.5680	1.196	0.566	1.805
0.6220	1.306	0.565	1.861

From Table 1 and Fig. 1, it can be seen that the osmotic coefficients  $\phi_m$  decrease with increasing molality. The cyclohexylsulfamic acid is a moderately strong acid with the dissociation constant of  $K_a = 0.0154 \pm 0.0005$  at 298.15 K and as such it was treated as 1:1 electrolyte.<sup>12</sup> The adjustable parameters of the Pitzer equation,<sup>15</sup> which for dilute aqueous solutions of 1:1 electrolyte takes the



**Fig. 1:** Osmotic coefficients (●) and mean ion activity coefficient (▲) of aqueous solutions of cyclohexylsulfamic acid at the freezing point of the solution

form shown in Eq. (3), were fitted to experimentally obtained osmotic coefficients:

$$\phi_m = 1 + f^{(\phi_m)} + mB^{(\phi_m)} \quad (3)$$

where:

$$f^{(\phi_m)} = -A^{(\phi_m)} \frac{m^{1/2}}{1 + bm^{1/2}} \quad (4)$$

and:

$$B^{(\phi_m)} = \beta^{(0)} + \beta^{(1)} \exp(-\alpha_1 m^{1/2}) \quad (5)$$

In Eq. (4),  $A^{(\phi_m)}$  is the *Debye-Hückel* constant, and  $b$  is a parameter of  $1.2 \text{ (kg mol}^{-1}\text{)}^{1/2}$  for aqueous solutions. The  $B^{(\phi_m)}$  term in Eq. (5) is a slowly varying function of molality that takes into account short-range ion-ion interactions. The parameter has an especially simple form having the desired general properties, namely it has a finite value at zero molality, a rapid change linear in  $m^{1/2}$  at low ionic strength and slow approach to an asymptotic value at high molality. The constant  $\beta^{(0)}$  in Eq. (5) is often correlated with structure-making and structure-breaking effects. The parameters  $\beta^{(0)}$ ,  $\beta^{(1)}$  and  $\alpha_1$  are obtained from experimental data.

In fitting procedures of osmotic coefficients with Eq. (3), we used a value of  $A^{(\phi_m)} = 0.37642 \text{ (kg mol}^{-1}\text{)}^{1/2}$  for the *Debye-Hückel* constant,<sup>22</sup> and for a temperature independent parameter,  $b = 1.2 \text{ (kg mol}^{-1}\text{)}^{1/2}$ , which was proposed by *Pitzer*<sup>15</sup> for aqueous 1:1 electrolyte solutions. The following values were received for the parameters:  $\beta^{(0)} = - (0.387 \pm 0.009) \text{ kg mol}^{-1}$ ,  $\beta^{(1)} = - (16.32 \pm 0.10) \text{ kg mol}^{-1}$ , and  $\alpha_1 = (6.484 \pm 0.005) \text{ (kg mol}^{-1}\text{)}^{1/2}$ . The value of parameter  $\alpha_1$  is substantially higher than 2.0, which

was the value used for simple 1:1 electrolyte solutions.<sup>23</sup> As was pointed out by *Pitzer*,<sup>23</sup> the value of parameter  $\alpha_1$  may be adjusted for each substance, if desired.

It has been noted previously on the bases of our viscosity studies that cyclohexylsulfamic acid is a weak structure-maker<sup>14</sup> and the results here agree. The negative value of  $\beta^{(0)}$  cyclohexylsulfamic acid (between the values of sodium saccharin and potassium acesulfame, both with evident structure-breaking effects<sup>24</sup>) merely confirms that it is necessary to assume incomplete dissociation of the acid<sup>14, 25</sup> and that important structure differences exist among these large organic ions. In zwitterion form of cyclohexylsulfamic acid the balance between hydrophilicity, hydrophobicity, anionic and cationic characteristics ensures only weak net interactions to surrounding water. The dissociation process results in the disappearance of the zwitterion structure and formation of hydrogen and cyclohexylsulfamate ions; the latter was already found as positive-solvating ion (structure-making ion) from the volumetric properties,<sup>24, 26</sup> opposite to the nitranions of saccharin (sodium salt) and acesulfame (potassium salt) which were classified previously as negative-solvating ions or structure-breaking ions.<sup>27</sup> However, the observed difference in parameters  $\beta^{(0)}$  and  $\beta^{(1)}$  relative to other 1:1 electrolytes can be attributed by the incomplete dissociation of cyclohexylsulfamic acid in aqueous solution and by hydrophobic hydration. Very large values of parameter  $\beta^{(1)}$  were also found for some higher carboxylate salts such as sodium octanoate ( $-7.73638 \text{ kg mol}^{-1}$ ), manolate ( $-10.3798 \text{ kg mol}^{-1}$ ), and decanoate ( $-7.40138 \text{ kg mol}^{-1}$ ).<sup>28</sup> The high values observed were described to the well known tendency of these salts to form micellar aggregates, especially at higher concentrations. The overall quality of the fit was expressed as the corresponding standard deviation shown in Eq. (6):

$$s = \pm \left[ \frac{\sum_{i=1}^n (\phi_{m,i(\text{exp})} - \phi_{m,i(\text{cal})})^2}{n} \right]^{1/2} \quad (6)$$

where  $\phi_{m,i(\text{exp})}$  and  $\phi_{m,i(\text{cal})}$  denote the experimental and calculated osmotic coefficients, and  $n$  is the number of experimental data. The standard deviation of the fit amounts to  $\pm 0.008$ . It was shown by *Marshall* et al.<sup>28</sup> that for a small molality range the experimental data by the *Pitzer* relation provides the best fit although the parameters obtained for small molality ranges do not necessarily result in good prediction at higher molality. They concluded that the *Pitzer* model does not appear to have good extrapolative capacity. Furthermore, the *Pitzer* model assumes the solution consists only of solvent, anions and cations. Incomplete dissociation, micellar aggregation or hydrolysis of electrolyte solutes are left out of account.

The mean ion activity coefficient of the solute,  $\gamma_{\pm}$ , was calculated from Eq. (7):<sup>15</sup>

$$\ln \gamma_{\pm} = f^{(\gamma)} + mB^{(\gamma)} \quad (7)$$

where:

$$f^{(\gamma)} = -A^{(\phi_m)} \left[ \frac{m^{1/2}}{1+bm^{1/2}} + \frac{2}{b} \ln(1+bm^{1/2}) \right] \quad (8)$$

and:

$$B^{(\gamma)} = 2\beta^{(0)} + \frac{2\beta^{(1)}}{\alpha_1^2 m} \left[ 1 - \exp(-\alpha_1 m^{1/2}) \left( 1 + \alpha_1 m^{1/2} - \frac{1}{2} \alpha_1^2 m \right) \right] \quad (9)$$

The calculated values of  $\ln \gamma_{\pm}$  are given in Table 1 and shown as  $\gamma_{\pm}$  in Fig. 1.

The non-ideal behaviour of the aqueous solutions of cyclohexylsulfamic acid was further characterized by the excess *Gibbs* energy of solution,  $G^E$ . From the mean ion activity coefficient and osmotic coefficient, the excess *Gibbs* energy of solution per kilogram of solvent is given by Eq. (10):<sup>15, 23, 29</sup>

$$G^E = 2mRT(1 - \phi_m + \ln \gamma_{\pm}) = RT \left( f^{(G^E)} + 2m^2 B^{(G^E)} \right) \quad (10)$$

where:

$$f^{(G^E)} = -A^{(\phi_m)} \left( \frac{4m}{b} \right) \ln(1+bm^{1/2}) \quad (11)$$

and:

$$B^{(G^E)} = \beta^{(0)} + \frac{2\beta^{(1)}}{\alpha_1^2 m} \left[ 1 - \exp(-\alpha_1 m^{1/2}) \left( 1 + \alpha_1 m^{1/2} \right) \right] \quad (12)$$

The calculated values of  $G^E$  are given in Table 2. The values of  $G^E$  are negative and show the same tendency for non-ideal behaviour as the osmotic and mean ion activity coefficient. The negative sign of the excess *Gibbs* energy may come from non-structural interactions, through hydrophobic interactions which are largely responsible for this behaviour.

The excess *Gibbs* energy of solution can also be given by Eq. (13):<sup>29</sup>

$$G^E = n_1 G_1^E + m G_2^E \quad (13)$$

where  $n_1$  represents the number of moles of solvent per kilogram of solvent,  $G_1^E$  ( $\text{J mol}^{-1}$ ) is the partial specific excess *Gibbs* energy of the solvent, and  $G_2^E$  ( $\text{J mol}^{-1}$ ) is the partial molar excess *Gibbs* energy of the solute.  $G_2^E$  can be calculated according to Eq.(14):<sup>29, 30</sup>

**Table 2.** Excess *Gibbs* energies of solution, partial specific excess *Gibbs* energies of the solvent and the solute, and the differences in the *Gibbs* energy of solvation of water molecules at the freezing point of the solution.

$m$ (mol kg <sup>-1</sup> )	$-G^E$ (J kg <sup>-1</sup> )	$n_1 G_1^E$ (J kg <sup>-1</sup> )	$-m G_2^E$ (J kg <sup>-1</sup> )	$\Delta \Delta G_1^*$ (J mol <sup>-1</sup> )
0.04655	101	55.5	158	5
0.09987	323	150	474	17
0.15403	602	250	852	30
0.20173	878	339	1214	41
0.26492	1274	461	1725	56
0.30426	1535	538	2061	65
0.35631	1895	644	2627	77
0.40904	2276	755	3122	90
0.45895	2649	860	3503	101
0.51019	3046	977	4023	113
0.56796	3508	1116	4635	127
0.62201	3954	1221	5232	139

$$G_2^E = \left( \frac{\partial G^E}{\partial m} \right)_{n_1, T, p} = 2RT \ln \gamma_{\pm} = 2RT \left[ f^{(\gamma)} + mB^{(\gamma)} \right] \quad (14)$$

and  $G_1^E$  according to Eq. (15):

$$G_1^E = 2M_1 m RT (1 - \phi_m) = -2M_1 m RT \left[ f^{(\phi_m)} + mB^{(\phi_m)} \right] \quad (15)$$

where  $M_1$  ( $\text{g mol}^{-1}$ ) is the molar mass of the solvent. The values of the products  $n_1 G_1^E$  and  $m G_2^E$  are given in Table 2. As can be seen the  $G_2^E$  values are negative, like  $G^E$ , i.e. they decrease with increasing concentration of the solute; in contrast, the  $n_1 G_1^E$  values are positive, and increase with increasing concentration of the solute. Cyclohexylsulfamic acid can exist in aqueous solution as a neutral molecule or in the zwitterionic form.<sup>12, 31</sup> The positive value of the product  $n_1 G_1^E$  can be ascribed to the hydrophobic solvation of solute molecules. The negative sign of the product  $m G_2^E$  comes from non-structural interaction and hydrophobic interaction of zwitterionic structure with solvent molecules.

Some information about the solvation ability of water can be obtained by the approach of *Ben-Naim*.<sup>32</sup> According to this theory, the difference in the *Gibbs* energy of solvation of water molecules in solution relative to pure water,  $\Delta \Delta G_1^*$  ( $\text{J mol}^{-1}$ ), is given by Eq. (16):

$$\Delta \Delta G_1^* = \Delta G_1^{*l} - \Delta G_1^{*p} = RT \ln \left( \frac{\rho_1^l a_1}{\rho_1^p} \right) \quad (16)$$

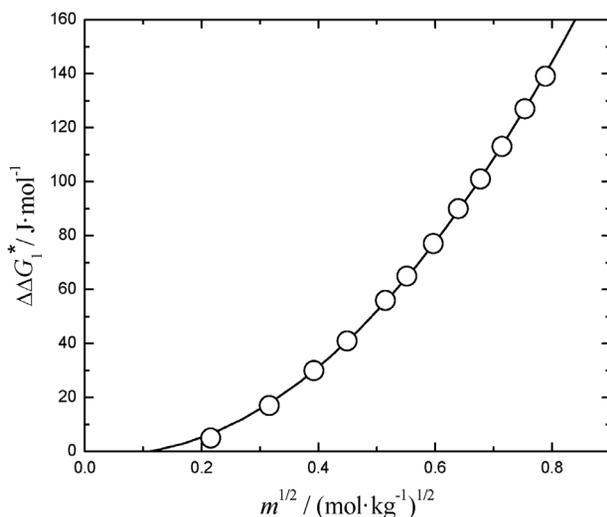
where  $\Delta G_1^{*l}$  and  $\Delta G_1^{*p}$  are the *Gibbs* energy of solution  $l$  relative to that of pure water  $p$ ,  $\rho_1$  is the number density of water in a pure solvent ( $\text{molecules/cm}^3$ ),  $\rho_1^l$  is the number density of water, and  $\alpha_1$  is the activity of water in solution. The term  $\rho_1^l/\rho_1^p$  was obtained through the respective densities and expansibility of water<sup>33</sup> and aqueous solution of

cyclohexylsulfamic acid.<sup>13</sup> The values of  $\ln a_1$  were calculated from Eq. (17):

$$\ln a_1 = -\nu m M_1 \phi_m \quad (17)$$

where  $\nu = 2$ . The calculated values of  $\Delta\Delta G_1^*$  (J mol<sup>-1</sup>) are given in Table 2. From Table 2 and Fig. 2, it follows that the  $\Delta\Delta G_1^*$  values are positive and small, and increase with increasing concentrations of the solute. It is interesting to note that  $\Delta\Delta G_1^*$  values of some strong acids, e. g. hydrochloric acid are small and negative.<sup>30</sup> Contrary, values of  $\Delta\Delta G_1^*$  of aqueous solutions of sucrose and glucose are positive and high.<sup>30</sup>

According to Ben-Naim,<sup>32</sup> it is not possible to state categorically whether the observed effects on  $\Delta\Delta G_1^*$  are due to direct solute-solvent interactions or to indirect changes in the structure of the solvent environment that are induced by the addition of the solute. Since the  $\Delta G_1^{*p}$  of water as a pure liquid at 0 °C is negative,  $\Delta G_1^{*p} = -27.792$  kJ mol<sup>-1</sup>,<sup>32</sup> we can conclude that the addition of cyclohexylsulfamic acid makes the Gibbs energy of solvation of water in solution,  $\Delta G_1^{*s}$ , less negative than in pure water. The major reason for this appears to be the overall weakening of the average binding energy of water molecules to their surroundings when cyclohexylsulfamic acid is added to the solvent.



**Fig. 2:** Concentration dependence of the Gibbs energy of solvation of water molecules in solution relative to pure water,  $\Delta\Delta G_1^*$ , for aqueous solution of cyclohexylsulfamic acid at the freezing point of solution

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## Povzetek

Na osnovi znižanja zmrzišča smo določili osmozni koeficient vodnih raztopin cikloheksilsulfaminske kisline. Koncentracijsko odvisnost osmoznega koeficienta smo izrazili s *Pitzerjevo* enačbo in ovrednotili interakcijske koeficiente  $\alpha_1$ ,  $\beta^{(0)}$  and  $\beta^{(1)}$ . Neidealno obnašanje vodnih raztopin cikloheksilsulfaminske kisline smo okarakterizirali s presežno *Gibbsovo* energijo raztopine, topljenca in topila ter pojasnili z nepopolno disociacijo in hidrofobno hidratacijo topljenca.