Determination of the mean activity coefficient of hydrochloric acid by electromotive force measurement

— written by: Mária Ujvári —

1.) Introduction

The task is the determination of the mean activity coefficient of hydrochloric acid by electromotive force measurement. This experiment is related to basic electrochemistry concepts, like electromotive force, potential of the cell reaction, Nernst equation, Debye-Hückel theory.

2.) Basics

2.a) Definition of activity

For the thermodynamic characterization of an \( i \) component of a system, the chemical potential is used which can be derived from the Gibbs function: the chemical potential is the partial molar Gibbs free energy:

\[
\mu_i = \left( \frac{\partial G(T, p, n)}{\partial n_i} \right)_{T, p, n_{-i}} \tag{2.1}
\]

The absolute activity (\( \lambda \)) is:

\[
\lambda_i = \exp \left( \frac{\mu_i}{RT} \right) \tag{2.2}
\]

The absolute activities – and the chemical potentials - of the components of a system are not independent of the composition. It’s worth introducing reference systems in which species have fixed absolute activities – therefore fixed chemical potentials: e.g. the “absolute pure material” or the “infinitely diluted solution”. We refer to the activity and chemical potential in such systems as standard absolute activity (\( \lambda_i^\circ \)) and standard chemical potential (\( \mu_i^\circ \)).

By the help of the absolute activity and the standard absolute activity of the \( i^{th} \) component of our system, the relative activity (\( a_i \)) can be written as:

\[
a_i = \frac{\lambda_i}{\lambda_i^\circ} \tag{2.3}
\]

Comparing equations 2.2 and 2.3 we arrive at the well-known thermodynamic expression:

\[
\mu_i = \mu_i^\circ + RT \ln a_i \tag{2.4}
\]

Taking an absolute dilute solution (where \( x_i = 0 \)) as a reference system:

\[
\lim_{x_i \to 0} \lambda_i = \lambda_i^\circ \tag{2.5}
\]

and the mole fraction dependence of the relative activity is:

\[
a_i = f_i x_i \tag{2.6}
\]
where \( f_i \) is dimensionless and it is called the activity coefficient of the \( i \text{th} \) component. The value of the activity coefficient depends on the composition and the natural variables of the Gibbs free energy function:

\[
   f_i = f_i(T, p, x)
\]  

(2.7)

Systems in which \( f_i = 1 \) for all components at any composition are called ideal systems.

The choice of the standard system is arbitrary and its composition can be given by any variable, e.g. molarity or mole fraction.

The relative activity can be written as follows:

\[
   a_i = y_i \frac{c_i}{c^o}
\]

(2.8)

where \( c^o = 1 \text{ mol} \cdot \text{dm}^{-3} \) is the standard molarity, and \( y_i \) is the relative activity coefficient expressed by the molarity.

2.b) Activity, mean activity and activity coefficients of dissolved electrolytes

Dissolving material \( B \) in a solvent, it usually dissociates, forms ions having a hydration shell which stabilizes the ions. These materials are called electrolytes. The driving force of the dissociation of the electrolytes is the decreasing of the Gibbs free energy caused by the hydration.

The dissolved \( B \) electrolyte can be thermodynamically characterized by the method written above. The relative activity based on the molarity is:

\[
   a_{\text{c,B}} = \frac{\lambda_{\text{c,B}}}{\mu_{\text{c,B}}}
\]

(2.9)

The chemical potential:

\[
   \mu_{\text{c,B}} = \mu_{\text{c,B}}^{\text{B}} + RT \ln a_{\text{c,B}} = \mu_{\text{c,B}}^{\text{B}} + RT \ln \left( y_B \frac{c_B}{c^o} \right) = \mu_{\text{c,B}}^{\text{B}} + RT \ln \frac{c_B}{c^o} + RT \ln y_B
\]

(2.10)

The last term of expression 2.10 \((RT \ln y_B)\) is the „excess” chemical potential, which describes the deviation of the behaviour of the electrolyte from the ideal. Its value is equal to the value of the virtual work which we have to do if we take 1 mole of electrolyte from the ideal state to the real state. In the solution of totally dissociating (called strong) electrolytes, the only cause of the deviation from ideal behaviour is the electrostatic interaction between the dissolved ions.

Above we have given formulas corresponding to the activity and chemical potential of the electrolytes, but we cannot determine the individual chemical potentials and activity of the formed ions. Since the anions and cations in a system are present always together, we can estimate only an average activity.

Assume that during the dissociation of the \( B \) electrolyte, the formed \( K \) cation’s number is \( \nu_+ \) and the formed \( A \) anion’s number is \( \nu_- \) based on the following reaction:

\[
   B \rightarrow \nu_+ K + \nu_- A
\]

(2.11)

Introduce quantity

\[
   \nu = \nu_+ + \nu_-
\]

(2.12)

for the denotation of the overall number of ions.

In case of totally dissociating electrolyte \( B \), the mean chemical potential for 1 mol ion in the solution is defined by the following equation:

\[
   \mu_\nu = \frac{\mu_B}{\nu} = \frac{\mu_B^O}{\nu} + RT \ln \sqrt[a_B]{\lambda_B} = \frac{\mu_B^O}{\nu} + RT \ln a_\nu
\]

(2.13)
where $\mu_\pm$ is the **mean chemical potential** of the dissolved electrolyte and $a_\pm$ is the mean activity of the electrolyte which can be defined as follows:

$$a_\pm = \sqrt{a_B} = \sqrt{\frac{\lambda_B}{\lambda_B^0}} = \exp \left[ \frac{\mu_B - \mu_B^0}{\nu RT} \right]$$

(2.14)

The mean chemical potential and activity of the ions formed during the dissolution is based on the well-defined activity and chemical potential of the B electrolyte. We can compose the activity of the electrolyte from the individual activities of the ions:

$$\lambda_B = \lambda_B^+ \lambda_B^-$$

(2.15)

and

$$\mu_B = \nu \mu_\pm = \nu \mu_+ + \nu \mu_-$$

(2.16)

The equation 2.15 is valid for the standard absolute activities. Also, we can easily validate the following relation corresponding to the mean relative activity:

$$a_\pm = \sqrt{a_B^+ a_B^-} = \sqrt{\nu \nu^+ \nu^- \frac{c_B}{c_B^0}} \cdot \sqrt{y^+ y^-}$$

(2.17)

Let’s introduce $Q$ as a simplification:

$$Q = \sqrt{\nu \nu^+ \nu^-}$$

(2.18)

Equation 2.17 can be written as:

$$a_\pm = Q \cdot \frac{c_B}{c_B^0} \cdot y_\pm$$

(2.19)

This is the definition of the main activity coefficient $y_\pm$ of the electrolyte which can be expressed with the individual activity coefficients in the following way:

$$y_\pm = \sqrt{y^+ y^-}$$

(2.20)

2.c) The Debye–Hückel-theory of the strong electrolytes

As it has been mentioned before, the individual activities of the ions in an electrolyte solution are not determinable by measurement; we can get information only about the mean activities. The subject of our measurement is this mean activity. In contrast to the undeterminable activities, these can be estimated using theoretical considerations. The most fundamental consideration is the Debye-Hückel theory of strong electrolytes. This theory is based on the following assumptions:

i.) Only coulombic interactions are between the ions (e.g. van der Waals interactions are neglected).

ii.) The permittivity of the solution is equal to the permittivity of the solvent. The influence of the solute dissolution is neglected.

iii.) The ions are point charges, (uniformly charged, non-polarizable spheres).

iv.) The dissociation is total.

v.) The potential between the ions is negligible to the thermal energy of the ions.

Based on these assumptions, taking into account the thermal distribution and the electrostatic interactions for the mean activity coefficient of strong electrolytes in very dilute solution we get:

$$\ln y_\pm = - \frac{A z_+ z_- \sqrt{T}}{1 + B a \sqrt{T}}$$

(2.21)
In this equation $I$ is called the ionic strength, which can be calculated knowing the $c_i$ concentrations and $z_i$ charge numbers:

$$I = \frac{1}{2} \sum_i c_i z_i^2$$  \hspace{1cm} (2.22)

The $A$ and $B$ constants in equation 2.21 at 25 °C can be given by:

$$A = \sqrt{2N_A} \frac{e^3}{8\pi(\varepsilon kT)^{1/2}} = 1.173 \text{ mol}^{-1/2}\text{dm}^{3/2}$$  \hspace{1cm} (2.23)

and

$$B = \sqrt{\frac{2N_A e^2}{\varepsilon kT}} = 3.29 \cdot 10^8 \text{ mol}^{-1/2}\text{dm}^{3/2}$$  \hspace{1cm} (2.24)

Parameter $a$ represents the distance of closest approach of ions. In very diluted solutions, where

$$Ba\sqrt{I} \ll 1$$  \hspace{1cm} (2.25)

is true, equation 2.21 can be simplified:

$$\ln y_z = -A|z_z z_i|\sqrt{I}$$  \hspace{1cm} (2.26)

This equation is called Debye–Hückel limiting law.

3.) General information about the measurement

3.a) Theory

For the determination of the mean activity and mean activity coefficient of an electrolyte (in our case HCl), we must have a galvanic cell which contains the given electrolyte and the electrode potentials of the electrodes depends only on the activity of the anions and the cations respectively at a given temperature. Knowing the concentration of the electrolyte using the measured electromotive force values as potentials of the cell reaction, the mean activity coefficients can be calculated. In our work we assemble galvanic cells with different electrolyte concentrations and measuring the electromotive force we can calculate the mean activity of the electrolyte.

The galvanic cell we use is composed with a silver/silver chloride and a hydrogen electrode. The Ag/AgCl system is an electrode of the second kind. Its electrode potential, based on the reaction

$$\text{AgCl} + e^- \rightarrow \text{Ag} + \text{Cl}^-$$  \hspace{1cm} (3.1)

is determined by only the activity of the chloride ions in the solution:

$$e_{\text{Ag/AgCl}} = e_{\text{Ag/AgCl}}^\circ - \frac{RT}{F} \ln a_{\text{Cl}^-}$$  \hspace{1cm} (3.2)

The hydrogen electrode is a gas electrode of the first kind consisting of hydrogen saturated H\(^+\) containing solution and platinum plate. Based on the electrode reaction

$$\text{H}^+ + e^- \rightarrow \frac{1}{2}\text{H}_2$$  \hspace{1cm} (3.3)

the electrode potential of the hydrogen electrode can be given as:

\* We used natural logarithm; therefore these values can differ from other sources.
The standard potential of the electrode reaction is not known accurate enough; it can be estimated by extrapolation.

In equation 2.26 of the Debye-Hückel limiting law, substituting the ionic strength with the concentration, the concentration dependence of the mean activity coefficient can be described as:

\[ \ln y_{±} = -A \sqrt{c_{HCl}} \]  

(3.11)

From the known quantities of the equation 3.10 introduce \( \zeta \) as follows:

\[ \zeta = \frac{E_{cell}}{F} + \frac{2RT}{F} \ln \tilde{c}_{HCl} \]  

(3.12)

From equations 3.10, 3.11 and 3.12:

\[ \zeta = E_{cell}^{o} - \frac{2RT}{F} \ln y_{±} = E_{cell}^{o} + \frac{2RT}{F} A \sqrt{c_{HCl}} \]  

(3.13)
Drawing the calculated $\zeta$ quantities as the function of the square root of the acid concentration and linear fitting to the linear part corresponding to low concentrations, the intercept gives $E_{\text{cell}}^\infty$ and constant $A$ can be calculated from the slope. (Comparing it to the theoretical value, $A = 1.173 \text{ mol}^{-1/2} \text{ dm}^{1/2}$, we can conclude some information about the goodness of our measurement).

Using equation 3.10 knowing the $E_{\text{cell}}^\infty$ value, we can calculate the mean activity coefficients for all solutions.

The theoretical value of the mean activity coefficients can be estimated based on the equation 3.14 which is resulting from the Debye–Hückel-theory of the strong electrolytes and is an applicable form of the equation 2.21 for hydrochloric acid (if $B = a^{-1}$ is an acceptable approximation for HCl).

$$\ln y_x = -A \frac{\sqrt{c_{\text{HCl}}}}{1 + \sqrt{c_{\text{HCl}}}}$$

(3.14)

Drawing the activity coefficients calculated from the measured data and from the theory as the function of the acid concentration, we can give an explanation for the deviation of the measured and the calculated values.

3.2) Practice

Cell assembling:

Put the Ag/AgCl electrode into water to desorb the chloride ions coming from the 0.1 mol·dm$^{-3}$ sodium chloride solution in which the electrodes are saved.

Assemble the electrochemical cell using the appropriate solutions with the help of the instructor. Begin with the most diluted solution! The sematic picture of the cell can be seen in Fig 3.1. Fill the hydrogen electrode and rinse the Ag/AgCl rod with the given solution! Immerse them to the same small beaker containing the same solution. Maximum the 3/4 part of the electrode should be immersed to the solution to avoid preparation of a mixed electrode (under the glass there is copper!).

The stopcock should be closed during the measurements, otherwise the solution would flow out to the beaker! Wet the stopcock well to decrease its resistance! We measure the electromotive force with a digital voltmeter having large internal resistance. The time of saturation of the solution with hydrogen takes about 15 minutes in each case. We begin the experiment with the most diluted solution and we go to the direction of the more concentrated ones, therefore we do not rinse the equipment and the electrodes with water but with the next solution after measurement.

The cell is not thermostated during the measurements. Read the temperature of the laboratory and write it to your lab report! Use this data for the calculations.

After finishing all measurements, rinse the electrodes with double distilled water and put back to their reservoir: Ag/AgCl to the KCl solution, Pt to water.

Some solutions are ready and preparation of the others is subject of the present work. Ask for the solutions and the factor of the stock solution from the technician.

The workplace is usually used by a group of students; the freshly prepared solutions are used at two workplaces (in this case use volumetric flasks having 200 cm$^3$ volume). If only one workplace is used, please use 100 cm$^3$ volumetric flasks.

For the preparation of the solutions, use calibrated burettes (with “A” or “B” sign on it) to measure the volumes from the stock solution (nominal concentration 0.1 mol·dm$^{-3}$) written in table 3.1 into calibrated volumetric flasks and adjust the meniscus to the calibration mark with twice distilled water. When calculating, use the real concentrations, not the nominal ones; use the calibration of the burette and the flasks! In table 3.1, volumes for 100 cm$^3$ solution are written. Do not forget to multiply it by 2 if you are preparing 200 cm$^3$! Write the sign of the workplace, flasks, and burettes in your lab report! Below volumes of 2 cm$^3$, use the micro burette!
Solution | $c_{\text{nominal}}$ (mol·dm$^{-3}$) | volume of the stock solution for dilution to 100 cm$^3$
---|---|---
1 | 0.0004 | This solution is ready.
2 | 0.0009 | 0.90
3 | 0.0012 | 1.20
4 | 0.0016 | 1.60
5 | 0.0020 | 2.00
6 | 0.0026 | 2.60
7 | 0.0036 | 3.60
8 | 0.0064 | 6.40
9 | 0.0100 | 10.0
10 | 0.0144 | These solutions are ready.
11 | 0.0196 | 
12 | 0.0225 | 

Tab. 3.1. Data for solution preparation.

Calibration data of the volumetric flask and burettes can be seen in table 3.2 – 3.4:

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<th>Sign of the flask</th>
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</tr>
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</tr>
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<td>100</td>
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Tab. 3.2. Calibration data of the volumetric flasks.

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<th>$V_{\text{nominal}}$ (cm$^3$)</th>
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<th>Workplace B</th>
</tr>
</thead>
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<td>$V_{\text{actual}}$ (cm$^3$)</td>
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Tab. 3.3. Calibration data of the burettes having 25 cm$^3$ volume.
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<th>workplace B $V_{\text{actual}}$ / cm$^3$</th>
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Tab. 3.4. Calibration data of the burettes having 2 cm$^3$ volume.

4.) Calculations

1. From the exact volume data (read from the burette), calculate the concentration of the solutions! Do not forget to use the factor of the stock solution and the calibration of the burettes.

2. Calculate $\zeta$ based on equation 3.12 for all solutions. Draw $\zeta$ as the function of the square route of the concentration of the hydrochloric acid. Perform linear fitting for the low concentration part of the data based on the equation 3.13; calculate $E_{\text{cell}}^\varphi$ and $A$! Give the confidence intervals for these values based on 95% level. Be aware of the number of significant digits and use the appropriate units! Report the number of fitted points.

3. Calculate the practical values of the mean activity coefficient based on the equation 3.10 using $E_{\text{cell}}^\varphi$, and estimate the theoretical values from equation 3.14. Draw the practical and the theoretical coefficients on the same diagram as the function of the acid concentration.

4. Summarize the data in a table: real concentration of the solutions, electromotive forces, and the calculated $\zeta$ values, practical ($y_\pm$) and theoretical ($y_{\pm,\text{DH}}$) activity coefficients.

5. Prepare your lab report using computer in printed form.
5.) Questions

1. What kind of electrodes do you use for the assembling of the cell? Write at least one other cell in which we can also measure the mean activity coefficient of the hydrochloric acid!
2. What method do you use for the measuring of the electromotive force?
3. Draw the cell diagram of the galvanic cell you use!
4. What can be the problem if the voltmeter does not show any voltage, shows abnormal or non-constant values?
5. If we measure the electromotive force with a digital voltmeter what is the most important requirement of the voltmeter?
6. Why could it be a problem if the solution is in connection with the copper contact at the Ag/AgCl electrode?
7. Why can we assume that the diffusion potential negligible small in the used galvanic cell?
8. What happens if we leave open the stopcock of the hydrogen electrode?
9. What types of electrodes are: hydrogen electrode, Ag/AgCl electrode?
10. Define the mean activity and the mean activity coefficient of the electrolytes! Why is the ions individual activity, activity coefficient non determinable?

6.) Literature

Determination of the mean activity coefficient of hydrochloric acid by electromotive force measurement

— Appendix: measurement datasheet —

Name of the student: __________________________________________
Name of the mates measured together: ____________________________
Date of measurements: ___/___/___
Sign of the workplace: ______
Temperature of the laboratory during the measurements: ______
Factor of the used stock solution: ______

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<th>Sign of the burette</th>
<th>Volume from the burette / cm$^3$</th>
<th>$E_{\text{me}}$ / V</th>
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