

❖ Debye-Huckel Limiting Law of Activity Coefficients and Its Limitations

So far in this chapter, the idea of ions in electrolytic solutions and their mutual interactions was taken for granted. However, the conceptual understanding of these electrolytic solutions thrived via a different route. Initially, the scientific community thought that the electrolytic solutions are the same as the non-electrolytic ones. In other words, they did not pay any special attention to the charged species, and therefore, treated them just like the non-electrolytes.

➤ Derivation of Debye-Huckel Limiting Law

Classically, the partial molar free energy (μ_i) of the i th species in a non-electrolyte is given by the following relation.

$$\mu_i = \mu_i^0 + RT \ln x_i \quad (91)$$

Where μ_i^0 is the partial molar free energy when the concentration (x_i) is unity (standard state). Mathematically, we can say that is if $x_i = 1$, $\mu_i = \mu_i^0$.

At this point, it worthy to point out that the Coulombic interactions (long-range) can be neglected in the case of non-electrolytic solutions, which is obviously due to the uncharged or neutral nature of the solute particles. However, some short-range electrostatic interactions like the London dispersion forces or the dipole-dipole may play a significant role if the average inter-particle distance is small enough. This means that if the dilution is large enough, the ion-ion interactions (long-range Coulombic effects) can simply be ignored in a non-electrolytic solution. Thus, the equation (91) is valid only for those solutions in which no long-range electrostatic interactions occur, and such solutions are called as ideal solutions. However, when the equation (91) was applied to the electrolytic solutions, it was found that

$$\mu_i - \mu_i^0 \neq RT \ln x_i \quad (92)$$

Such solutions which show deviation from equation (91) are called as non-ideal solutions. This means that unlike non-electrolytes, the ion-ion interactions must be considered before any theoretical treatment for the chemical potential of ionic solutions is carried out.

Therefore, to rationalize this deviation, the scientific community developed a unique approach even before the Debye-Huckel's ionic-cloud theory. They tried to quantify the deviation from "idealistic behavior" by incorporating an empirical parameter f_i in the equation (92) as

$$\mu_i - \mu_i^0 = RT \ln x_i f_i \quad (93)$$

Where μ_i^0 is the partial molar free energy when the concentration (x_i) and correction factor (f_i) both are unity. Mathematically, we can say that if $x_i = 1$ and $f_i = 1$, $\mu_i = \mu_i^0$; which is obviously a hypothetical situation in which standard state behaves ideally. Moreover, it should also be noted that term x_i represents the actual concentration of the ions of i th type which may or may not be equal to expected concentration because weak electrolytes do not dissociate completely like the strong one. After that, it was assumed that it is not the actual

concentration (x_i) but the effective concentration ($x_i f_i$) that dictates the chemical potential change. For simplicity, the effective concentration if i th species was simply labeled as the activity (a_i). Mathematically, we can say that

$$a_i = x_i f_i \quad (94)$$

The correction factor f_i is also called as the “activity coefficient” and has a value of unity for ideal solutions i.e. when $f_i = 1$, $a_i = x_i$.

Consequently, the partial molar free energy change in going from the ideal state to the real state in ionic solutions can be written as

$$\mu_i - \mu_i^0 = RT \ln x_i + RT \ln f_i \quad (95)$$

The above equation is an empirical formulation of the behavior of ionic solutions and cannot give any theoretical result for f_i . Therefore, in order to find out the physical significance of the activity coefficient, we need to assume an ionic solution that can be switched from ideal (no ion-ion interaction) to the real situation (with ion-ion interaction). For ideal solutions, we have

$$\mu_i(\text{ideal}) = \mu_i^0 + RT \ln x_i \quad (96)$$

For real solutions,

$$\mu_i(\text{real}) = \mu_i^0 + RT \ln x_i + RT \ln f_i \quad (97)$$

The chemical potential change from ion-ion interaction can be obtained by subtracting equation (96) from equation (97) i.e.

$$\mu_i(\text{real}) - \mu_i(\text{ideal}) = \Delta\mu_{i-I} = \mu_i^0 + RT \ln x_i + RT \ln f_i - \mu_i^0 - RT \ln x_i \quad (98)$$

$$\Delta\mu_{i-I} = RT \ln f_i \quad (99)$$

Hence, the activity coefficient is a measure of chemical potential change due to the interaction of i th types with the rest of the ionic species. Now, according to the Debye-Huckel theory of ion-ion interaction, the partial molar free energy change due to ion-ion interaction is

$$\Delta\mu_{i-I} = -\frac{N_A Z_i^2 e_0^2 \kappa}{2\varepsilon} \quad (100)$$

Where Z_i is the charge number on the i th type of ion while e_0 is the electronic charge. N_A is the Avogadro number and ε is the dielectric constant of the surrounding medium, i.e., solvent. The symbol κ with n_i^0 as the bulk concentration is

$$\kappa = \left(\frac{4\pi}{\varepsilon kT} \sum_i n_i^0 Z_i^2 e_0^2 \right)^{1/2} \quad (101)$$

Now, from equation (99) and equation (100), we have

$$RT \ln f_i = -\frac{N_A Z_i^2 e_0^2 \kappa}{2\epsilon} \quad (102)$$

The above result implies that the Debye-Huckel's ionic cloud theory enables us to determine the activity coefficient in a theoretical framework.

Furthermore, if the actual concentration of the i th type of ion is represented in terms of molarity (c_i) or the molality (m_i), the equation (96) takes the form

$$\mu_i = \mu_i^0(c) + RT \ln c_i \quad (103)$$

and

$$\mu_i = \mu_i^0(m) + RT \ln m_i \quad (104)$$

Similarly, for real solutions, equation (97) takes the forms

$$\mu_i = \mu_i^0(c) + RT \ln c_i + RT \ln f_c \quad (105)$$

and

$$\mu_i = \mu_i^0(m) + RT \ln m_i + RT \ln f_m \quad (106)$$

Where $\mu_i^0(c)$ and $\mu_i^0(m)$ are corresponding standard chemical potential at a molarity c_i and molality m_i , respectively.

At this point, some experimental limitations must be discussed before any comparative analysis of the activity coefficient is carried out. It is a quite well-known fact that we cannot measure the hydration energy, i.e., ion-solvent interaction of individual ionic species because the addition of only cations or the anions is not possible practically. Even if it was possible, it would result in a negatively or positively charged solution depending upon the nature of the ions added, which eventually, would cause undesired interactions. Owing to similar arguments, it is also practically impossible to measure the activity coefficient f_i which is also a function chemical potential change arising from ion-ion interaction. The only way to avoid the situation is to add the electroneutral electrolytes to the solvent which would eventually produce the positive and negative ions simultaneously irrespective of whether it is strong or weak. Therefore, one can determine the activity of a net electrolyte consisted of minimum two ionic species, and we need something that can connect activity coefficient of electrolyte with the individual ionic species. In other words, the activity coefficient of a single ionic species can be determined only via a theoretical route. All this resulted in the idea of "mean ionic activity coefficient". To illustrate mathematically, consider a NaCl-type univalent electrolyte MA. The chemical potential of cations (M^+) and anions (A^-) can be written as

$$\mu_{M^+} = \mu_{M^+}^0 + RT \ln x_{M^+} + RT \ln f_{M^+} \quad (107)$$

and

$$\mu_{A^-} = \mu_{A^-}^0 + RT \ln x_{A^-} + RT \ln f_{A^-} \quad (108)$$

Combining the equation (107) with equation (108), we get

$$\mu_{M^+} + \mu_{A^-} = (\mu_{M^+}^0 + \mu_{A^-}^0) + RT \ln (x_{M^+}x_{A^-}) + RT \ln (f_{M^+}f_{A^-}) \quad (109)$$

The above equation gives the free energy of the system due to two moles of ions i.e. one mole of M^+ and mole of A^- ; or due to one mole of electroneutral electrolyte. Since we are interested in the average input to the total free energy due to one mole of ions only, therefore, we must divide the equation (109) by 2, i.e.,

$$\frac{\mu_{M^+} + \mu_{A^-}}{2} = \frac{\mu_{M^+}^0 + \mu_{A^-}^0}{2} + RT \ln (x_{M^+}x_{A^-})^{1/2} + RT \ln (f_{M^+}f_{A^-})^{1/2} \quad (110)$$

If we consider

$$\mu_{\pm} = \frac{\mu_{M^+} + \mu_{A^-}}{2} \quad (111)$$

$$\mu_{\pm}^0 = \frac{\mu_{M^+}^0 + \mu_{A^-}^0}{2} \quad (112)$$

and

$$x_{\pm} = (x_{M^+}x_{A^-})^{1/2} \quad (113)$$

$$f_{\pm} = (f_{M^+}f_{A^-})^{1/2} \quad (114)$$

Where μ_{\pm} and μ_{\pm}^0 are the mean chemical potential and standard mean chemical potential, respectively. The symbol x_{\pm} and f_{\pm} represent the mean ionic mole fraction and mean ionic activity coefficient, respectively. Here, it is also worthy to note that μ_{\pm} and μ_{\pm}^0 are simply the arithmetic means whereas x_{\pm} and f_{\pm} are the geometric mean quantities. Now using values from equations (111–114) into equation (110), we get

$$\mu_{\pm} = \mu_{\pm}^0 + RT \ln x_{\pm} + RT \ln f_{\pm} \quad (115)$$

Since it is for one mole instead two, we can write

$$\frac{1}{2}\mu_{MA} = \mu_{\pm} = \mu_{\pm}^0 + RT \ln x_{\pm} + RT \ln f_{\pm} \quad (116)$$

Hence, the experimental value of f_{\pm} can be obtained just by knowing the free energy of one mole of electrolytic solution at a particular concentration. Once the value of f_{\pm} is known, the product of individual activity coefficients can be obtained using equation (114). The individual values of activity coefficients obtained from equation (102) can be put into equation (114) to compare with experimentally observed value so that the Debye-Huckel model can be tested.

Furthermore, if one mole of electroneutral electrolyte generates ν_+ and ν_- moles of cations and anions, then equation (107) and equation (108) will become

$$\nu_+\mu_+ = \nu_+\mu_+^0 + \nu_+RT \ln x_+ + \nu_+RT \ln f_+ \quad (117)$$

and

$$\nu_-\mu_- = \nu_-\mu_-^0 + \nu_-RT \ln x_- + \nu_-RT \ln f_- \quad (118)$$

To find the free energy change due to per mole of cation and anion, add equation (117) to equation (118) and then divide by $\nu = \nu_+ + \nu_-$.

$$\frac{\nu_+\mu_+ + \nu_-\mu_-}{\nu} = \frac{\nu_+\mu_+^0 + \nu_-\mu_-^0}{\nu} + RT \ln (x_+^{\nu_+} x_-^{\nu_-})^{1/\nu} + RT \ln (f_+^{\nu_+} f_-^{\nu_-})^{1/\nu} \quad (119)$$

If we consider

$$\mu_{\pm} = \frac{\nu_+\mu_+ + \nu_-\mu_-}{\nu} \quad (120)$$

$$\mu_{\pm}^0 = \frac{\nu_+\mu_+^0 + \nu_-\mu_-^0}{\nu} \quad (121)$$

and

$$x_{\pm} = (x_+^{\nu_+} x_-^{\nu_-})^{1/\nu} \quad (122)$$

$$f_{\pm} = (f_+^{\nu_+} f_-^{\nu_-})^{1/\nu} \quad (123)$$

Now using values from equations (120–123) into equation (119), we get

$$\mu_{\pm} = \mu_{\pm}^0 + RT \ln x_{\pm} + RT \ln f_{\pm} \quad (124)$$

Taking logarithm both side of equation (123), we have

$$\ln f_{\pm} = \frac{1}{\nu} (\nu_+ \ln f_+ + \nu_- \ln f_-) \quad (125)$$

Now putting the value of $\ln f_+$ and $\ln f_-$ using equation (102), we get

$$\ln f_{\pm} = -\frac{1}{\nu} \left[\frac{N_A e_0^2 \kappa}{2\epsilon RT} (\nu_+ Z_+^2 + \nu_- Z_-^2) \right] \quad (126)$$

Now owing to the electroneutrality of the solution, $\nu_+ Z_+$ must be equal to $\nu_- Z_-$, i.e.,

$$\nu_+ Z_+^2 + \nu_- Z_-^2 = \nu_+ Z_+ Z_- + \nu_- Z_- Z_+ \quad (127)$$

$$\nu_+ Z_+^2 + \nu_- Z_-^2 = Z_+ Z_- (\nu_+ + \nu_-) \quad (128)$$

$$v_+ Z_+^2 + v_- Z_-^2 = Z_+ Z_- \nu \quad (129)$$

After putting the value of equation (129) into equation (126), we have

$$\ln f_{\pm} = -\frac{N_A e_0^2 \kappa}{2\epsilon RT} Z_+ Z_- \quad (130)$$

Furthermore, using the value of κ using equation (101), the above equation takes the form

$$\ln f_{\pm} = -\frac{N_A e_0^2 Z_+ Z_-}{2\epsilon RT} \left(\frac{4\pi}{\epsilon kT} \sum_i n_i^0 Z_i^2 e_0^2 \right)^{1/2} \quad (131)$$

Since $n_i^0 = c_i N_A / 1000$, the equation (131) becomes

$$\ln f_{\pm} = -\frac{N_A e_0^2 Z_+ Z_-}{2\epsilon RT} \left(\frac{4\pi}{\epsilon kT} \sum_i \frac{c_i N_A Z_i^2 e_0^2}{1000} \right)^{1/2} \quad (132)$$

Multiply and divide the equation (132) by 2, and then put $c_i Z_i^2 / 2 = I$, i.e., ionic strength, we get

$$\ln f_{\pm} = -\frac{N_A e_0^2 Z_+ Z_-}{2\epsilon RT} \left(\frac{8\pi N_A e_0^2}{1000 \epsilon kT} \right)^{1/2} \sqrt{I} \quad (133)$$

$$\ln f_{\pm} = -\frac{N_A e_0^2 Z_+ Z_-}{2\epsilon RT} B \sqrt{I} \quad (134)$$

Where the constant B is defined as

$$B = \left(\frac{8\pi N_A e_0^2}{1000 \epsilon kT} \right)^{1/2} \quad (135)$$

Converting the natural logarithm to the common logarithm, the equation (134) becomes

$$\log f_{\pm} = -\frac{1}{2.303} \frac{N_A e_0^2 Z_+ Z_-}{2\epsilon RT} B \sqrt{I} \quad (136)$$

After a new constant A as

$$A = \frac{1}{2.303} \frac{N_A e_0^2}{2\epsilon RT} B \quad (137)$$

The equation (136) can be further simplified as given below.

$$\log f_{\pm} = -A(Z_+ Z_-) \sqrt{I} \quad (138)$$

Which is the Debye-Huckel limiting law of activity coefficients.

➤ **Limitations of Debye-Huckel Limiting Law**

The negative sign in the Debye-Huckel limiting law implies the fact that the activity coefficient is always less than unity. In order to discuss the limitations of this law, recall the popular relationship, i.e.,

$$\log f_{\pm} = -A(Z_+Z_-)\sqrt{I} \quad (139)$$

Since A is constant and the product Z_+Z_- is also constant for a particular electrolyte, the logarithm of activity coefficient must decrease linearly with the square root of ionic strength. In other words, the slope $\log f_{\pm}$ and \sqrt{I} can be determined simply by knowing the valences of the ions involved and by the knowledge of some physical constants. Furthermore, it should also be noted that the slope is independent of the very nature of the electrolyte and is a function of valences of cations and anions only. For instance, since the value of A for the water as solvent is 0.509, the slope of for NaCl as well as for KCl must be equal to 1×0.509 only. For uni-bivalent and the bi-bivalent electrolyte is should be 2×0.509 and 4×0.509 . The logarithmic variation of mean ionic activity coefficient (f_{\pm}) with the square root of the ionic strength (\sqrt{I}) for electrolytes of different valences follows the equation (139) pretty strictly.

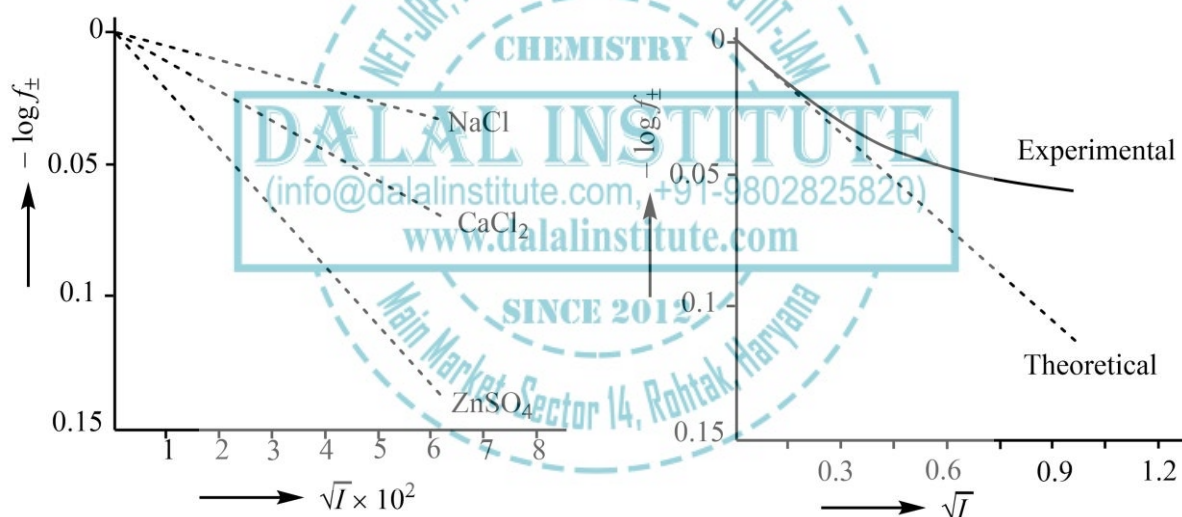


Figure 12. The logarithmic variation of f_{\pm} with the square root of the ionic strength (\sqrt{I}) for electrolytes of different valences in very dilute (left) and in solutions up to large concentration (right).

It is obvious from both the graphs that $\log f_{\pm}$ becomes zero when the dilution is very large indicating that the activity coefficient is unity, which is according to the Debye-Huckel limiting law. However, it should also be remembered that any theory is always a simplification of the real problem, and therefore, some deviations are expected. The same has been observed when the ionic strength is increased. It can be clearly seen that the deviation of the experimental result increases with the rise in the square root of the ionic strength.

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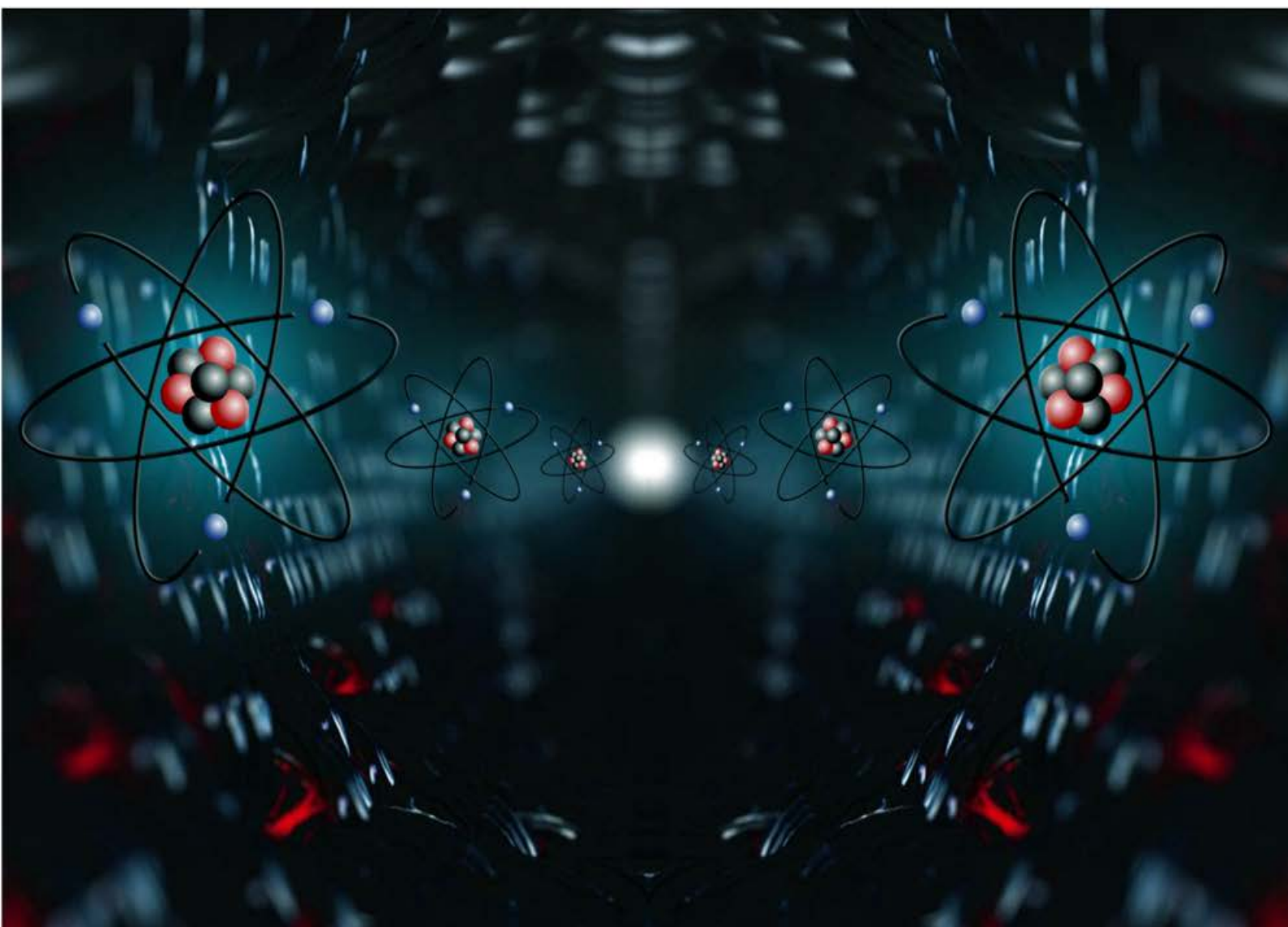
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MANDEEP DALAL



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