Thermodynamic Study of Osmotic Coefficient of Fatty acid salts

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Abstract: Fatty acid salts are used in numerous industrial applications. Understanding thermodynamics is crucial for achieving the desired quality of products in industries and for addressing a variety of environmental issues. we must understand thermodynamics. From sodium formate to sodium heptylate, we've taken a solution of seven sodium fatty acid salts and divided them into electrostatic and non-electrostatic depending on their constituents. An electrostatic contribution to the osmotic coefficient is given by the carboxylic group in the salt, while a non-electrostatic contribution is given by the hydrocarbon compounds in the salt. The hydrophobic part of hydration has received much less research attention than the hydrophilic part (Electrostatic contribution). To determine the non-electrostatic contribution, we employed the Flory-Huggins theory. For Long-chain electrolytes' hydrocarbon chains' miscibility is governed by the entropic force balances, which are typically negligible, and local interactions between various segments, which only favour miscibility in the presence of certain forces. We observe that for long-chain electrolyte solution, the hydration number is the same because the only carboxylic group is responsible for hydration, and the rest hydrocarbon chain will not contribute to hydration. The debey-Huckle model is used to calculate Electrostatic Contribution to the osmotic coefficient.

Keywords: Fatty acid salts, Osmotic Coefficient, Hydration number, Electrostatic, Non-Electrostatic.

Introduction:

Fatty acid salts are commonly used in food production, beverages, pharmaceuticals, cosmetics, agriculture/animal feed, and other industries as intermediate (manufacturing various organic chemicals). Bayer and Steiger [1] conducted research. Commonly discovered as efflorescence on museum items including vintage metals, stonework, and ceramic are acetates and formates. The organic salts combine to produce complicated mixed electrolyte systems that depend on temperature and relative humidity, together with inorganic salts like chlorides, nitrates, and sulfates that are frequently found in porous materials[2-3]. When it comes to the conservation of porous artifacts, the dynamics of the underlying phase equilibria are very important because crystal development within the pores creates stresses that are strong enough to lead to disintegration. Temperature and relative humidity must be maintained at the proper levels to stop crystal development and prevent damage. A thermodynamic study of fatty acid salts is necessary to anticipate the behaviour of a salt mixture.

To investigate the nature of different ionic interactions in numerous industrial and environmental processes, one has to know the thermodynamic characteristics of aqueous mixed electrolyte solutions.

Fatty acid anions like Formate, Acetate, Propionate, Butyrate, etc. are considered fatty acid salts because of their tendency to tighten up the water in their near vicinity. These ions are all negatively charged and are concentrated in the carboxylic group. Since they are all produced from weak acids that are the same strength, their carboxylate group contacts the adjacent water molecule strongly in the same manner and to the same extent.

Fatty acid Salts contain Hydrophilic (Carboxylic group) and Hydrophobic (Hydrocarbon tail) Components. Both Parts play their role significantly in a function. While the molecular structure's hydrocarbon constituents are primarily responsible for hydrophobic hydration, hydrophilic (nucleophilic and electrophilic) interactions with water are monopolized by atoms that act as electron donors or acceptors as well as any positively or negatively charged groups that may be present [4]. An acknowledged but poorly understood component of this scenario is that a molecule's presence can alter the structure of nearby solvent layers, which can then interact with the molecule and affect its structure and function. Numerous computer models, NMR, IR, X-ray, and neutron scattering techniques have been used to extensively study carboxylate hydration. However, these studies were primarily focused on the hydration of the COO group and only to a lesser extent on the impacts of their alkyl substituents. However, these findings imply that water is highly organized around the COO- group. Therefore, the primary emphasis of our work is the study of hydrophobic Characteristics of sodium fatty acid salts. Clusters of ion hydrates will appear when the electrolyte disintegrates in the water. It comprises both cations and anions. This category of hydrates is distinguished by the hydration numbers and the dispersions distribution for stoichiometric coefficient hydrates. The amount of interactions between cluster ion constituent elements determines these distributional characteristics. Dispersion forces drive hydrophobic hydration. The amount of the distribution's dispersion hydrate in this instance should be greater than hydrophilic hydration.

Significant discrepancies from ideality are observed in the aqueous solutions of electrolytes. Ion hydration is the primary cause of these aberrations. It was discovered [7] that Our ability to characterize the thermodynamic characteristics of solutions at different salt concentrations, from a dilute to a saturated solution, is made possible by the concept of average ion hydration number. Ionic hydration is the process by which water molecules align according to the polarity of an ionic group, and hydrophobic hydration is the process by which water molecules form a hydrogen bond network similar to clathrate hydrates. In addition, there is a large amount of data about the odd patterns in which the osmotic coefficients change with salt concentration: first increasing, then passing through a maximum, and then decreasing, then going through a minimum. For aqueous solutions of different sodium carboxylates, this dependence was discovered using the isopiestic approach [8]. It was hypothesized that it was caused by the micelle production for longer chain lengths or branched chains [6-7]. Therefore, the ions exhibit slightly lower osmotic coefficients than the corresponding straight chain.

In accordance with the literature, electrolytes, where either one or both ions are subject to hydrophobic hydration, are where the abnormal dependences of the Osmotic Coefficient on molality are found. As a result, the goal of this research was to create a model for characterizing the thermodynamic characteristics of concentrated aqueous electrolyte solutions, which are non-ideal because of hydrophilic and hydrophobic hydrations. The Hydrophobic Hydration (Osmotic Coefficient Contributed Non-Electrostatically) is Hypothesized to be Calculated Using Flory-Huggins Theory.

In this work, the Flory-Huggins Interaction parameter for various fatty acid salts and the Hydration number of sodium fatty acid salts have been determined. These results provide information regarding the non-electrostatic interaction with the osmotic coefficient.

Methodology:

Electrostatic and non-electrostatic contributions are taken into consideration when calculating the total osmotic coefficient.

$$\varphi = 1 + \varphi_e^E + \varphi_e^{NE} \tag{1}$$

when there is no ionic interaction in the solution is represented by the first term on the right in this equation, which equals 1. (i.e. the ideal solution's osmotic coefficient). The contributions to the nonideal part of the osmotic coefficient that is denoted by the terms "φ_e^E" and "φ_e^{NE}" are electrostatic (superscript E) and nonelectrostatic (superscript NE). The subscript e is used to indicate that they are in the excess numbers.

The contribution of Electrostatic to Osmotic Coefficient:

The Extended Debey-Huckle theory is said to be employed in the literature [11] to determine the contribution of electrostatics to the osmotic coefficient. The Extended Debey-Huckle theory is applied as stated in this theory.

$$\varphi_e^E = \frac{1}{3}(Z_+Z_-)A_\varphi\sqrt{I}\sigma(\mathrm{Ka}) \tag{2}$$

Where

Z_+ denotes the cations charge and Z_- denotes the charge of the anion.

$$\sigma(x) = \frac{3}{(ka)^3}[1 + (ka) - \frac{1}{1 + (ka)} - \ 2\,ln(1 + (ka)] \tag{2.1}$$

And

$$A_\varphi = \frac{F^2 e\sqrt{2}}{8\Pi(\varepsilon\varepsilon_0 RT)^{\frac{3}{2}}\sqrt{1000}} \tag{2.2}$$

In this Equation, I is Ionic strength of the solution and k is an inverse Debey length, a is Ionic Size Parameter,

$$\mathrm{k} = \frac{\mathrm{F}\sqrt{2 * 10\text{^}3 * \ \mathrm{c}}}{\sqrt{2\varepsilon\varepsilon_0\mathrm{RT}}} \tag{2.3}$$

e is the charge of electrons, F is the Faraday's constant, ε is Dielectric constant of the pure water and ε0 is the permittivity of free space, R universal gas Constant, T is the Temperature of the Electrolyte.

The Contribution of Non-Electrostatic to Osmotic Coefficient:

For Non-electrostatic contributions, Generalized Flory – Huggins model is used, is given as[12]

$$\frac{\Delta G}{RT} = \frac{n_S}{r_p}\ln(\phi_S) + n_w \ln(1-\phi_S) + \chi n_w \phi_S + n_S \frac{\mu_S^0}{RT} + n_w \frac{\mu_w^0}{RT} \tag{3}$$

The moles of solute and water in the aqueous solution are represented in the equation above by the letters n_S and n_w . The chemical potentials of a pure solute (μ_S^0)and a pure liquid (μ_w^0), respectively, are given by these symbols. The term χ denotes the Flory-Huggins Interaction parameter and should be considered as a function of the temperature, T, and the volume percentage of the solute, ϕ_S. rp is the radius of gyration (i.e. rp = 1 for short chain length),

Differentiate Equation number 3 concerning moles of Water, To find Water activity

$$\left[\frac{\partial(\Delta G/RT)}{\partial n_w}\right]_{n_s} = \frac{\nu n_s}{\phi_s}\frac{\partial \phi_s}{\partial n_w} + \ln(1-\phi_s)\frac{\partial n_w}{\partial n_w} - \frac{n_w}{(1-\phi_s)}\frac{\partial \phi_s}{\partial n_w} + \chi\phi_s\frac{\partial n_w}{\partial n_w} + \chi n_w \frac{\partial \phi_s}{\partial n_w} + n_w\phi_s\frac{\partial \chi}{\partial n_w}$$

The Volume Fraction of Salt is given as

$$\phi_s = \frac{\nu n_s v_h}{v_w n_w + \nu n_s v_h} \tag{4}$$

Molality is given as

$$\mathrm{c} = \frac{\mathrm{\nu n_s}}{\mathrm{v_w n_w + \nu n_s v_h}} \tag{5}$$

Relation between volume fraction of solute and molarity, we get this relation by solving the above equation 4 and 5.

$$\varphi_s = c v_h \tag{6}$$

The rate at which the free energy of the thermodynamic system changes in response to the addition of more atoms or molecules of a certain species to the system is referred to as the chemical potential of that species in the mixture. The following is the expression for the Chemical Potential of Water and Solute in the Solution is given as

$$\left[\frac{\partial(\Delta G/RT)}{\partial n_w}\right]_{n_s} = \frac{\mu_w - \mu_w^0}{RT} \tag{7}$$

Chemical potential of water in a solution is denoted by μ_w , pure water by μ_w^0 , and a system's change in Gibbs free energy is denoted by ΔG.

The Water activity in the Fatty acid salt solution can be obtained using the below equation.

$$\ln a_w = \frac{\mu_w - \mu_w^0}{RT} \tag{8}$$

By solving the above equations, We get Water activity.

$$\ln a_w = \ln(1 - cv_h\ ) + cv_h\ \left(1 - \frac{vv_w}{v_h}\right) + \chi(cv_c\ )^2\ - (1 - cv_h)(cv_h\ )^2 \frac{\partial\chi}{\partial cv_h} \tag{9}$$

The osmotic Coefficient φ is related to the Chemical potential of water

$$\varphi = -\frac{\mu_w - \mu_w^0}{M_w RTvm} \tag{10}$$

On substitution of equation 8 and 10 in equation 9, We get

$$\varphi_e^{NE} = \frac{-1}{M_w vm}\left[\ln(1 - cv_h\ ) + c\ (v_h - vv_w) + \chi(cv_c\ )^2 - (1 - cv_h)(cv_h\ )^2 \frac{\partial\chi}{\partial cv_h}\right] \tag{11}$$

Where

v_h indicates the Hydrated Salt Molar Volume, v_c= Long Chain Electrolyte molar volume, M_w is Molecular Weight of water, m = molality , v= Stoichiometric Coefficient of Salt , c= Molarity. The Solution change of mixing due to entropy is shown in first term and The change in mixing caused by enthalpy is shown by the second term. The third and fourth terms describe how mixing has changed as a result of interactions between molecules of the solute and solvent (Flory-Higgins Interaction term).

Results and Discussion:

We have taken the Osmotic Coefficient data with changes in Concentration for different Sodium fatty acid salts from Literature [9,13]. plotted graph between Osmotic Coefficient and molality. Marked the linear region in the plots and calculated slopes for different sodium fatty acid salts.

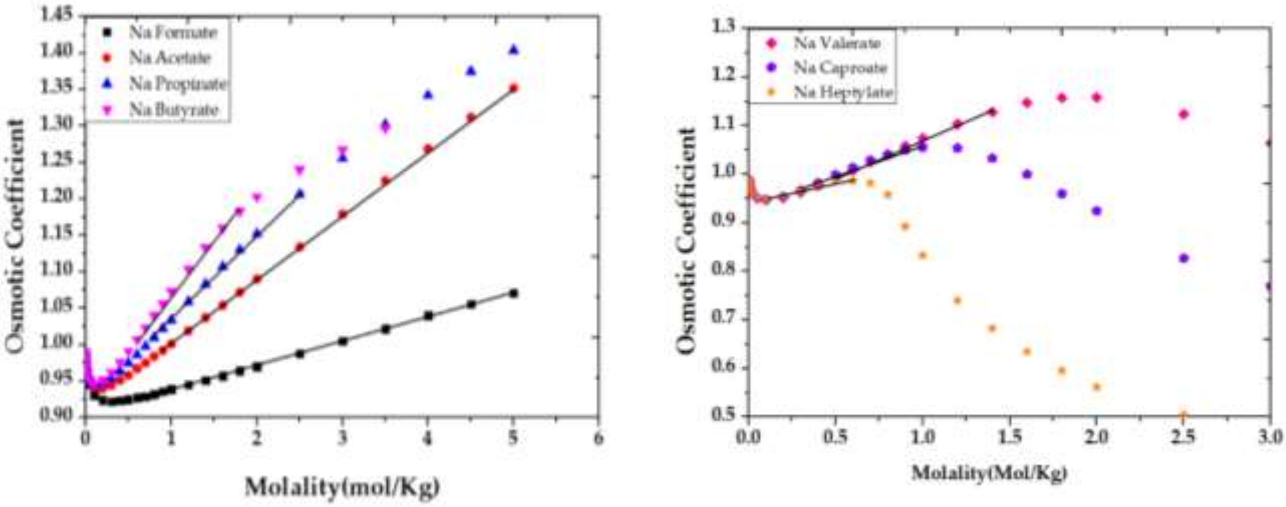

Figure 1 : *Osmotic Coefficient of Sodium fatty Acid Salt at different concentrations. The linear region is marked with Bold Line.*

The graphs show that sodium formate and sodium acetate are more prevalent in the linear zone because the hydrocarbon compound impact is less prevalent. The linear zone is contracting as we move toward higher order hydrocarbon molecules, which is caused by an increase in the hydrophobic effect. For sodium caproate, the slope is reducing as the CH2 chain length grows, and for sodium heptylate, it is even lowering as a result of an increase in the radius of gyration.

Table 1: *Slopes of experimentally determined osmotic coefficient and molality graphs for several sodium fatty acid salts.*

Compounds	**Slope (Experimental)**
Na Formate	3.39×10^{-2}
Na Acetate	8.80×10^{-2}
Na propinate	1.20563×10^{-1}
Na Butyrate	1.64×10^{-1}
Na Valerate	1.65×10^{-1}
Na Caproate	1.56×10^{-1}
Na Heptylate	1.04×10^{-1}

Molar Volume Calculations:

Using equation number 11, it is possible to calculate the contribution of Non-Electrostatic to the osmotic coefficient for various sodium fatty acid salts. Calculating the hydrated salts molar volume is one of the crucial variables. The bare electrolyte's molar volume is added along with the result of the hydration number and the molar volume of the pure solvent to calculate the molar volume of the hydrated salt.

$$v_h = v_s + hv_w \tag{12}$$

Where

v_h is the hydrated salt molar volume , v_s is the bare salt molar volume, h is the Hydration number, v_w is the pure water molar volume.

From the constituent ions Pauling radii, the bare salt molar volume v_s is estimated. applying the formula

$$v_s = \frac{4\pi}{3} N_{av} \Sigma r_i^3 \tag{13}$$

Where

N_{av} is Avogadro number and r_i is the pauling radius of constituent ions. The radii for sodium formate is taken from reference[8,14].

The density of sodium acetate salt is taken from the literature [16]. Converted into the molar volume of sodium acetate salt v_s. Similarly, the molar volume of the CH_2 Chain is Calculated by taking density data of alcohols like Methanol, Ethanol, and propanol from the literature [17]. Converted into molar volume. The difference in the molar volume of ethanol and the molar volume of methanol gives the molar volume of CH_2 Compound (v_c) or The difference in the molar volume Propanol and the molar volume ethanol. The addition of the molar volume of CH_2 to the molar volume of sodium acetate gives the molar volume of sodium propionate. Similarly, molar volume is calculated for the rest of the salts. The molar volume of CH_2 taken a constant value. Because it has negligible change with change in concentration.

Hydration Number Calculation:

For sodium formate, the hydration number (h) is calculated By comparing the slope of the experimentally discovered Osmotic coefficient graph with the slope of the Osmotic coefficient calculated owing to Non-Electrostatic Contribution. By ignoring the χ interaction terms in equation number 11, because the sodium formate formula only contains the carboxylic group. Hydrophobes have no impact on the osmotic coefficient.

$$\varphi_e^{NE} = \frac{-1}{M_w v m}[\ln(1 - c v_h\ ) + c\ (v_h - v v_w)] \tag{13}$$

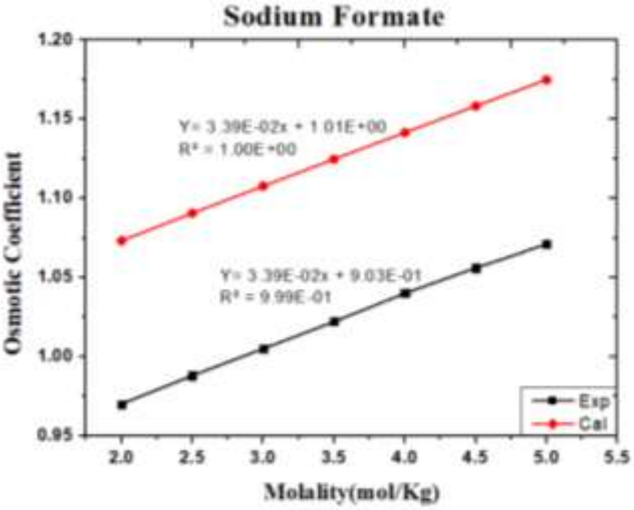

Figure 2: *The Contribution of Non-Electrostatic to Osmotic Coefficient estimated for sodium formate matched the experimentally measured osmotic coefficient line slope.*

The Hydration number calculated for sodium formate h = 3.664. This is used for the rest of the salts because only the Carboxylic group is responsible for the hydration number and the Carboxylic group (COO^- Na^+) is the same in all salts.

Calculation of the osmotic coefficient using the Flory-Huggins theory:

By using Equation number 11. We can Calculate Non electrostatic osmotic coefficient for different sodium fatty acid salts. non-electrostatic interaction includes the hydration number and Flory-Huggins interactions parameter χ. The first type of chain in this long chain electrolyte is a carboxylic group, which contributes to hydration but has no Flory-Huggins interaction χ ; the second type of chain is a hydrocarbon chain, which contributes nothing to hydration but has a total contribution to Flory-Huggins interaction χ.

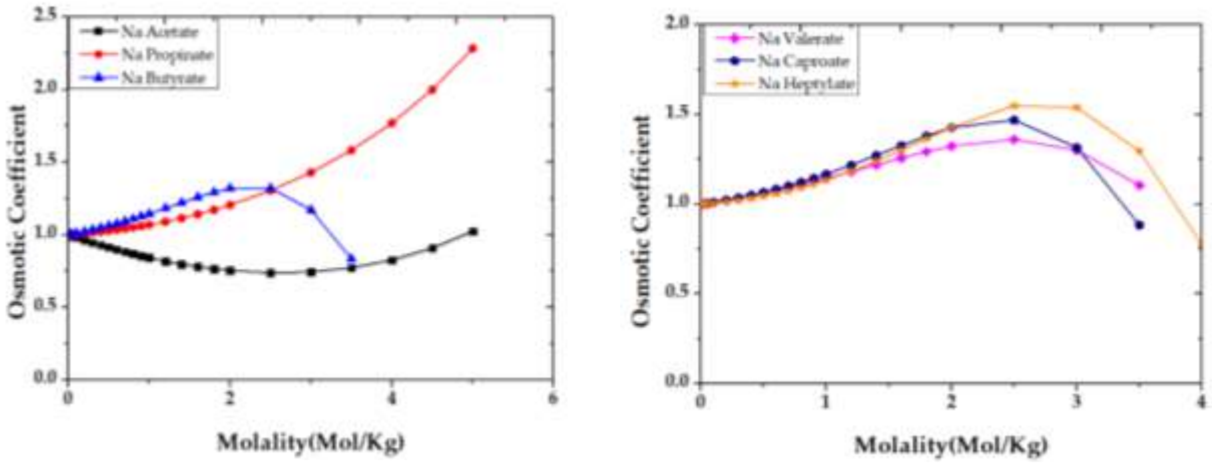

Figure 3: *The Contribution of Non-Electrostatic to Osmotic coefficient of sodium fatty acid salts.*

The above graphs depict The contribution of Non-Electrostatic to the osmotic coefficient Calculated from a developed model from Flory-Huggins Theory. Due to the high influence of χ interactions Osmotic coefficient is increasing for sodium Acetate and Sodium propionate. Whereas for remaining salts Osmotic Coefficient increases reaches a maximum and decreases same behaviour observed by smith and Robinson in 1942 by isopiestic method as we discussed above. It is because they assumed that due to formation of micelles. Campbell and Lakshminarayanan [15]. have broken this myth by performing surface tension experiments for Sodium octanoate, sodium decanoate, sodium laurate and sodium myristate. Their surface tension of sodium carboxylates, measured by stalagmometer method educes that sodium carboxylates having carbon chain length less than eight do not show any type of micellar formation but Sodium carboxylates having carbon chain length greater that eight, confirm their micellar formations. So, we assumed that the decrease in osmotic coefficient is due to the decrease in χ interactions influence at higher concentration for long chain sodium fatty acid salts. And the Flory-Huggins interaction becomes constant for all long chain salts. The Contribution of Electrostatic to the osmotic coefficient is calculated by differencing Contribution of non-Electrostatic to the osmotic coefficient from the total Osmotic coefficient.

$$\varphi^E = \varphi - \varphi^{NE} \qquad (14)$$

And these Electro static contribution is compared with the Debey-Huckle Theory.

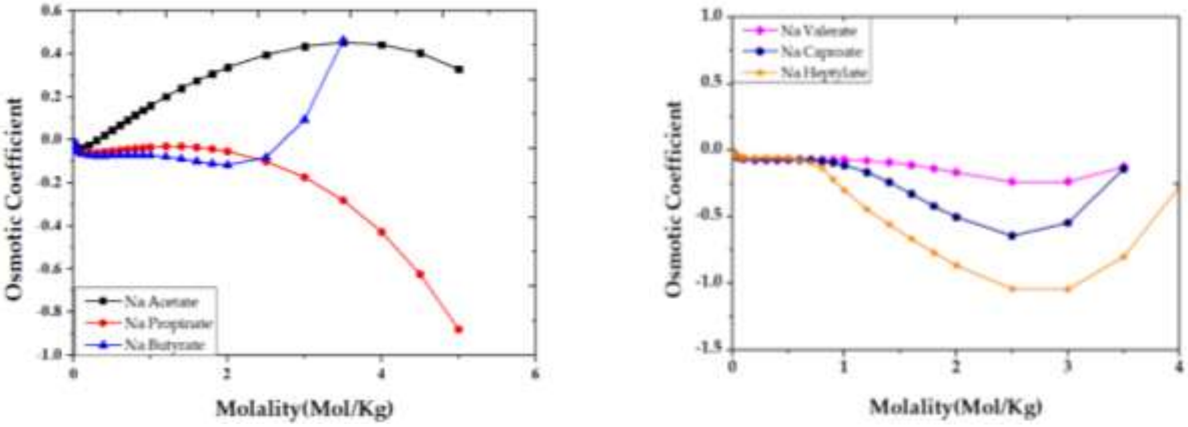

Figure 4: *The contribution of Electrostatic to the osmotic coefficient of sodium fatty acid salts. (From Flory-Huggins Theory).*

The above graphs depict The contribution of Electrostatic to osmotic coefficient of different fatty acid salts.

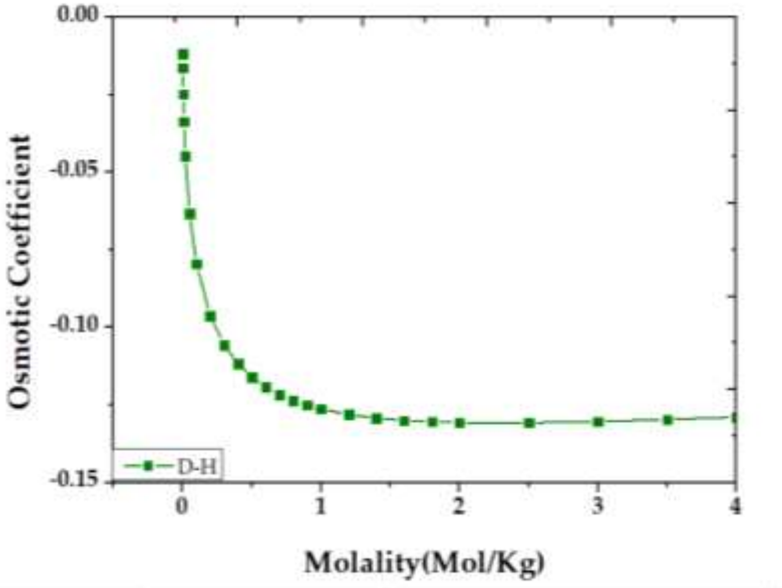

Figure 5: *Variation of Osmotic coefficient by varying of molality. (Debey-Huckle Theory)*

The above graph depicts The Change in osmotic coefficient by change in molality. Calculated from Debey-Huckle Theory. It would be compared with the contribution of electrostatic to osmotic coefficient Calculated from flory-Huggins theory. The change in osmotic Coefficient is same for all salts because the carboxylic group is same in all sodium fatty acid salts. The contribution of Electrostatic to osmotic coefficient (From Flory-Huggins) for most of the salts is similar to that of change in osmotic Coefficient from Debey-Huckle.

The Flory-Huggins interaction parameter is calculated by comparing the slope of the experimentally discovered Osmotic coefficient graph with the slope of the Osmotic coefficient calculated owing to Non-Electrostatic Contribution. By including the χ interaction terms in equation number 11.

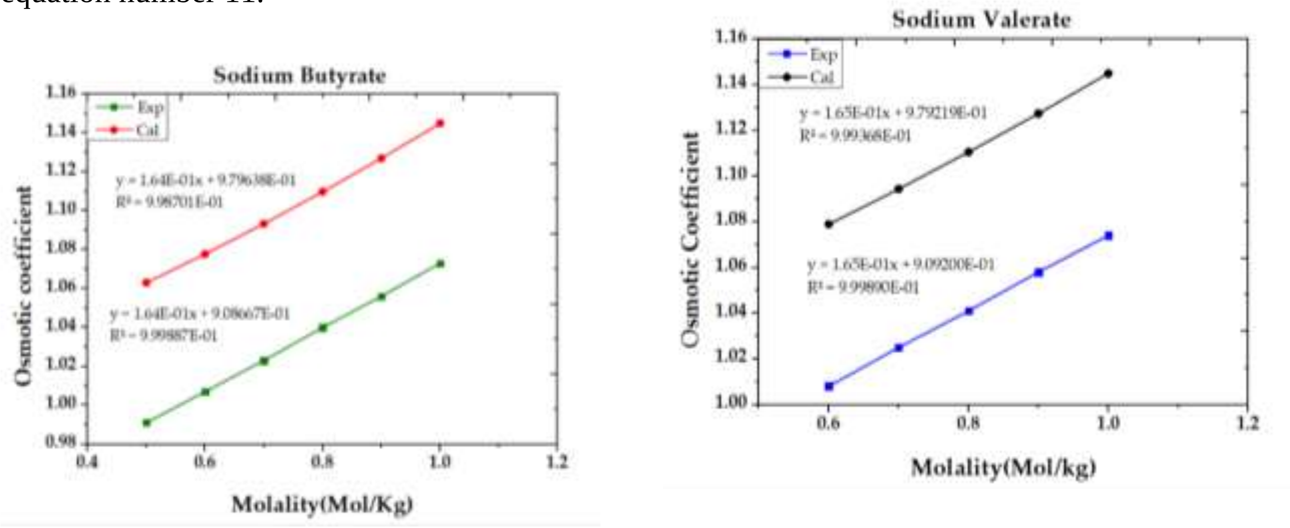

Figure 6: *The slope of Experimentally determined osmotic coefficient graph is matched with the Osmotic coefficient calculated by Flory-Huggins Theory. (For sodium Butyrate and Sodium valerate)*

The Flory-Huggins interaction parameter Values are given as

Table 2: *Flory-Huggins Interaction Parameter for different sodium fatty acid salts.*

Compound	**F-H Interaction Parameter (χ)**	**No. Of CH2 Compounds Present**
Na Acetate	78.06226	1
Na Propinate	11.76333	2
Na Butyrate	5.12368	3
Na Valerate	2.56323	4
Na Caproate	2.9211	5
Na Heptylate	2.704734	6

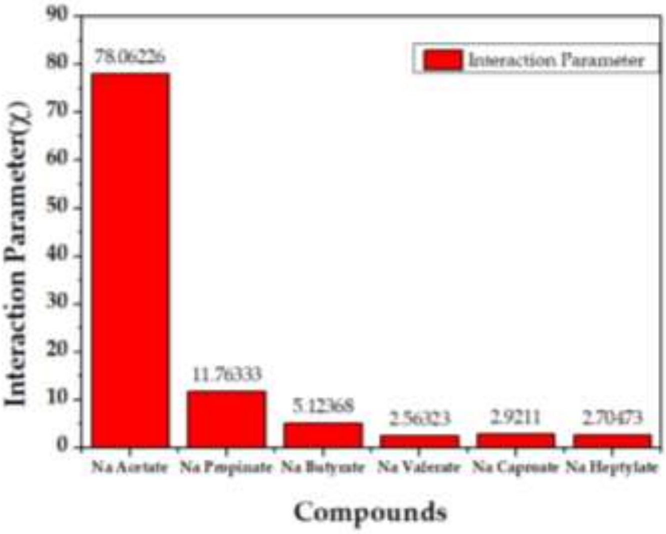

Figure 6: *The graphical representation of Flory-Huggins interaction parameter values.*

For sodium Acetate the Flory-Huggins interactions parameter is more due to the free energy available is less. So, the Chain will be straight. This is not the case with other salts, As number of CH_2 Compounds increasing All the carbon compound try to aggregate together form a clusters. Small surface area available per volume. So, For long chain fatty acid interaction parameter become almost simillar.

Conclusion:

The long hydrocarbon chain electrolyte has certain nonpolar carbon chains that do not mix with water molecules and have a hydrophobic effect. The phrase "hydrophobic effect" describes how poorly nonpolar compounds dissolve in water when compared to polar chemicals and organic solvents. Hydrogen bonds between the water molecules will be broken to make space for a hydrophobe when it is dropped into an aqueous solution, but water molecules do not interact with the hydrophobe. Because The breaking of bonds is referred to as an enthalpic reaction since heat is generated in the system.Water molecules that are deformed by the hydrophobe will create new hydrogen bonds and an ice-like cage structure known as a clathrate cage around the hydrophobe. The Change in entropy is negative because this orientation increases the structure of the system (hydrophobe) while reducing its overall entropy. As a result of the new hydrogen bonds being able to partially, entirely, or excessively make up for the hydrogen bonds that the hydrophobe's entry destroyed, the change in enthalpy (ΔH) of the system can be negative, zero, or positive. However, because there has been a significant increase in entropy (ΔS), the change in enthalpy is not significant in determining the spontaneity of the reaction (the mixing of hydrophobic molecules and water).

In our work we estimated the Flory-Huggins interaction parameter χ Contribution to osmotic coefficient for different sodium fatty acid salts. We observed The balance of entropic forces, which are typically negligible, as well as the local interaction between various segments, which only favours miscibility of hydrocarbon chain of long chain electrolyte in the presence of particular forces. Such as Columbic, Vander walls and hydrogen bond forces etc , Typically, the Flory-Huggins parameter χ is used to express the factor in a mean-field approximation. The value of χ regulates the phase structure; samples have a miscibility gap for χ values greater than a critical value χ_c, and are homogenous for χ values below a critical value χ_c.

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