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# Micellar Properties of Linear Alkyl Benzene Sulphonate in Aqueous Glucose Solution

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**ABSTRACT**

*The solution properties of linear alkyl benzene sulphonate (LABS) in water in presence of glucose have been investigated conductmetrically. The critical aggregation concentration (cac) and critical saturation concentration (csc) were determined. It has been observed that at low concentration of surfactant glucose interact with hydrophilic head group of surfactant molecules and form molecular aggregate/micelle like structure. The decrease in molar conductance, $\lambda_m$ , with increasing concentration of glucose in post saturation region confirms the formation of LABS – glucose aggregate and the reported structure maker character of glucose in aqueous solution. The structure making character of glucose becomes more effective above 0.5 mol dm$^{-3}$ concentration as the value of binding capacity tends to minimum value. The micellization of LABS in presence of aqueous glucose solutions has shown that the glucose behave as a weak electrolyte and can form aggregates through interactions with LABS. In aqueous solution, further increase in concentration of surfactant which corresponds to the saturation points beyond which micellization of surfactant follow a regular trend of post micellization like in water. It suggests the association and ionization of glucose – LABS aggregate / micelle, which is justified by the data of binding capacities at different concentration. The free energy of micellization in this study has same trend as it is reported for sodium dodecyl sulphate surfactant in glucose solution. The study may be a model to the surfactant systems which are applicable in interpretation of the biological, catalytic and industrial formulation of commercial importance.*

**Keywords:** Linear alkyl benzene sulphonate; glucose; surfactant; micellization; solute-solute interaction; binding capacity.

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## INTRODUCTION

The studies on aqueous micellar systems have received much attention in the literature due to their versatile and important role in all branches of science. The knowledge of the physical properties and thermodynamics of micellization of surfactant in solvent mixtures may be useful to select appropriate surfactant system of specific utility. The various systems have been investigated in recent past [1-15]. The critical micelle concentration of surfactant solutions and micellar structures with shape can be changed by additives in solutions. The micellar properties are influence by the presence of additives [16, 17]. The effect of additives on properties of surfactant has been a subject of great significance in a variety of industrial and technological fields [18]. The interactions of surfactants with additives in aqueous solutions have been studied during the past several years [19, 20]. These additives belongs to four classes namely electrolytes, polar organic compounds, non- polar organic compounds and other surfactants. In aqueous solution the addition of electrolytes causes a decrease in the critical micelle concentration, cmc, of the ionic surfactants [17, 21] whereas addition of non polar organic compounds results the increase in cmc value of the surfactant [22]. The presences of polar organic compounds have structure maker/ breaker character in water which influences the process of micellization. The solvent characteristic like dielectric constant, viscosity, density, hydrogen bond forming capacity produces a change in the formation of micelles in

presence of an additive. In presence of a second surfactant the micellization takes place by the synergistic interaction between both the surfactant molecules [23]. So it is difficult to generalize the effect of any class of additives on micellization of a surfactant. In the present study we present the effect of non ionic additive glucose in order to differentiate it from ionic additive [21, 24]. Glucose plays an essential role in the metabolism of living organism which may be useful to explore the utilization possibilities of surfactants in the biological process like drug delivery [25]. A study on intermolecular forces of glucose in water [26] predicted solute-solute interaction/ solute-solvent interaction are more significant and glucose behaves as structure maker. It belong to the class of sugars which are well known stabilizing agents for native states of proteins / enzymes due to their ability to enhance the structure of water.

## EXPERIMENTAL SECTION

De-ionized water twice distilled was used in all the studies. The specific conductance of the water prepared for the present study was of the order of $<2\times10\ \Omega^{-1}\ cm^{-1}$.The LABS used in the study were obtained in the laboratory by sulphonation of linear alkyl benzene (IPCL, $C_{10}$=14, $C_{11}$=31, $C_{12}$=37, $C_{13}$=16 and $C_{14}$=1%) having an average molecular weight of 343.Surface tension measurements were made on aqueous solution of LABS prepared by dissolving an accurately weighed sample in distilled water. No surface tension minima were found which implies that no surface-active impurities exist in the studied LABS samples. The techniques employed for test of impurities was same as reported in literature [19]. The solutions of glucose (Quligens, AR) were prepared by dissolving the calculated amount in distilled water.

*2.1 Conductance measurements*

Conductance measurement was carried out on a digital conductivity meter (Systronics type 306) with a sensitivity of 0.1% and a dipping type conductivity cell with a platinum electrode of cell constant 1.0. All the measurements were made at constant temperature using thermostatically controlled water bath (Tanco, Kanpur) capable of maintaining the temperature constant to $\pm0.1\ ^0C$.

## RESULTS AND DISCUSSION

The specific conductance values at constant concentration of glucose in aqueous solution were obtained by varying the concentration of LABS. The specific conductance data for different concentration are given in Table 1.

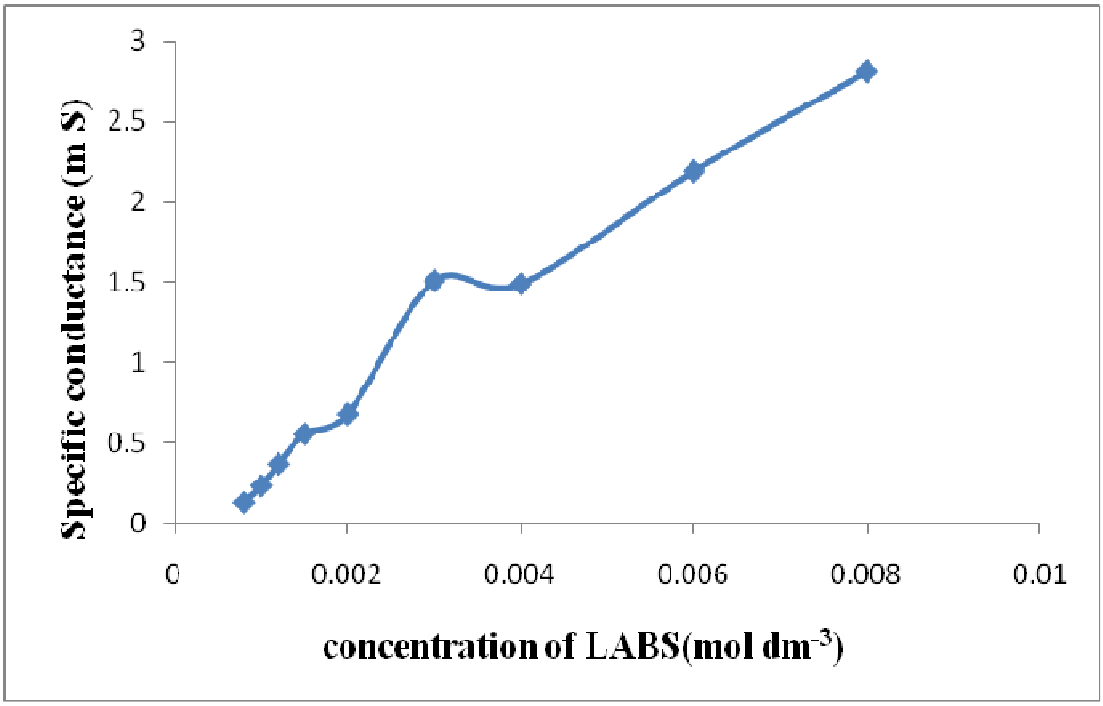


**Fig.1 Plot for 0.1mol $dm^{-3}$ glucose at 301K**

**Table 1. Variation of specific conductance of LABS with different concentration of glucose**

| S.No. | [LABS] | Temp | Specific conductance ( m S) | | | | | |
|---|---|---|---|---|---|---|---|---|
| | $10^{-3}$ moldm$^{-3}$ | K | [Glucose] 0.0 moldm$^{-3}$ | [Glucose] 0.1 moldm$^{-3}$ | [Glucose] 0.2 moldm$^{-3}$ | [Glucose] 0.3 moldm$^{-3}$ | [Glucose] 0.4 moldm$^{-3}$ | [Glucose] 0.5 moldm$^{-3}$ |
| 1 | 0.8 | 301 | 0.110 | 0.130 | 0.160 | 0.170 | 0.140 | 0.180 |
| | | 311 | | 0.190 | 0.220 | 0.190 | 0.150 | 0.190 |
| 2 | 1.0 | 301 | 0.273 | 0.234 | 0.268 | 0.235 | 0.235 | 0.254 |
| | | 311 | | 0.278 | 0.310 | 0.286 | 0.258 | 0.294 |
| 3 | 1.2 | 301 | 0.449 | 0.370 | 0.350 | 0.320 | 0.330 | 0.340 |
| | | 311 | | 0.410 | 0.401 | 0.390 | 0.360 | 0.401 |
| 4 | 1.5 | 301 | 0.588 | 0.557 | 0.503 | 0.442 | 0.469 | 0.462 |
| | | 311 | | 0.603 | 0.541 | 0.548 | 0.521 | 0.562 |
| 5 | 2.0 | 301 | 0.751 | 0.681 | 0.632 | 0.612 | 0.590 | 0.551 |
| | | 311 | | 0.862 | 0.776 | 0.793 | 0.746 | 0.736 |
| 6 | 3.0 | 301 | 1.16 | 1.51 | 1.09 | 1.04 | 1.00 | 0.997 |
| | | 311 | | 1.30 | 1.25 | 1.24 | 1.13 | 1.160 |
| 7 | 4.0 | 301 | 1.58 | 1.49 | 1.43 | 1.39 | 1.31 | 1.290 |
| | | 311 | | 1.74 | 1.63 | 1.62 | 1.51 | 1.470 |
| 8 | 6.0 | 301 | 2.24 | 2.19 | 2.09 | 2.03 | 1.91 | 1.870 |
| | | 311 | | 2.43 | 2.28 | 2.30 | 2.21 | 2.140 |
| 9 | 8.0 | 301 | 2.87 | 2.81 | 2.69 | 2.62 | 2.47 | 2.430 |
| | | 311 | | 3.16 | 3.00 | 2.97 | 2.82 | 2.750 |

In Figure 1, the first break point occurs at 0.0015 mol dm$^{-3}$; this value is 25% higher from the reported literature value of cmc, 0.0012 mol dm$^{-3}$ for LABS [24]. It is clear from the Figure 1 and data of Table-1**.** The accuracy in the calculation of break points is based not only on graph but their value has been obtained from the linearity relation. The values of regression coefficient always greater than 0.99 and the gradient value 617 is obtained from the data up to 0.0015 mol dm$^{-3}$ concentration and the gradient value 319 is obtained for the data above 0.002 mol dm$^{3}$concentration. If we calculate the gradient between two break points, its valve is 248. The presences of break points were checked in all the sets by reproducibility in the conductance values. Thus the second break point at concentration 0.002 mol dm$^{-3}$ has been assessed in case of aqueous glucose solutions. In water a single break point in linear plot of specific conductance versus molar concentration of surfactant corresponds to the cmc value. In aqueous glucose solution a different trend in plots has been assessed from experimental data with linearity relation. The trend is independent of glucose concentration in the concentration range 0.1 to 0.5 mol dm$^{-3}$ in the same concentration region of LABS 0.0015 to 0.002 mol dm$^{-3}$. In Figure 1, the two concentration range 0.0008 mol dm$^{-3}$ to 0.0015 mol dm$^{-3}$ and 0.002 mol dm$^{-3}$ to 0.008 mol dm$^{-3}$ can be accessed from the experimental data of Table 1.The regression results for specific conductance and concentration of LABS consist linear relation for both the concentration region but the value of gradients are different. A comparison of the gradients calculated with the help of computer clearly indicates that for various systems reported in the Table 1, the gradient values in different concentration regions follows the order: 0.0008 mol dm$^{-3}$ to 0.0015 mol dm$^{-3}$ $\neq$ to 0.002 mol dm$^{-3}$ to 0.008 moldm$^{-3}$ $\neq$ 0.0015 mol dm$^{-3}$ to 0.002 moldm$^{-3}$. An important observation made from the infliction region 0.0015mol dm$^{-3}$ to 0.002 mol dm$^{-3}$ shows same pattern for different set of conductivity data obtained at different concentration of glucose. Therefore, the presence of infliction region has been assessed. The variation of gradient in three concentration regions has been confirmed by calculating the standard deviation in the values of specific conductance in concentration region up to 0.0015 mol dm$^{-3}$ and above 0.002 mol dm$^{-3}$.For example, the standard deviation was 0.155 up to 0.0015 mol dm$^{-3}$ and 0.79 above 0.002 mol dm$^{-3}$ for glucose solution of 0.2 mol dm$^{-3}$ at 301K. In order to ascertain these specific regions of concentration, the molar conductance, $\lambda_m$, of surfactant solution were calculated from the experimental data.

Figure 2 indicates the inflection in curve in the same concentration region of LABS as assessed from linearity relationship with concentration. The cmc however cannot be determined from these plots but the concave upward nature of these plots in lower concentration indicating that the surfactants behaves as weak electrolyte in lower concentration and Debye - Huckel - Onsager equation is not applicable to the surfactant in aqueous glucose solutions in this concentration region. For the concentration range above 0.002 mol dm$^{-3}$ the plot indicates the applicability of this equation. The value of limiting molar conductance has been obtained from the intercept of the linear plot from the data of higher concentration region. The value of limiting molar conductance, $\lambda_{0,}$ have been recorded in Table 2**.**

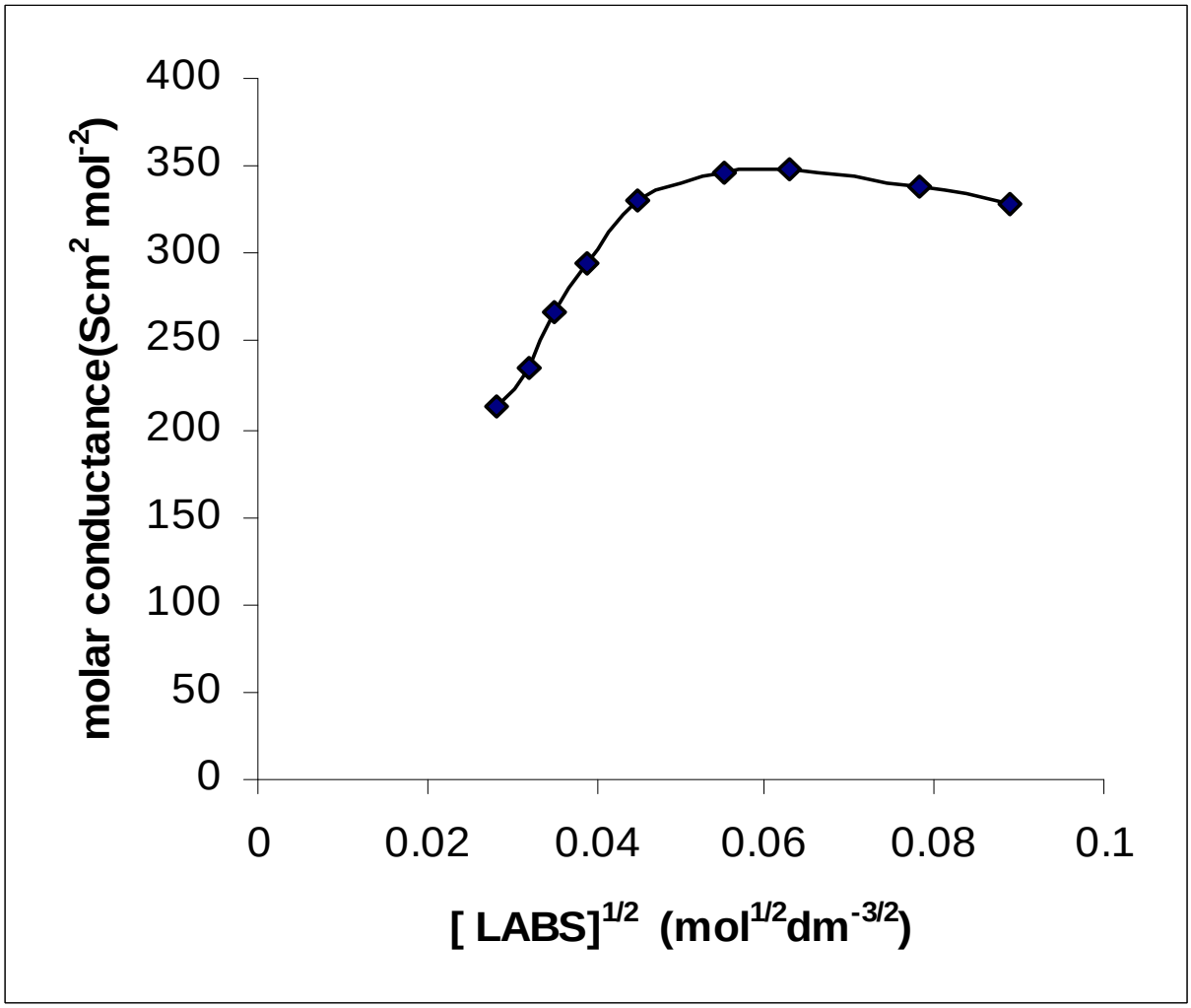


**Fig. 2 Plot for 0.3 mol dm$^{-3}$ glucose at 301 K**

**Table 2. Values of degree of ionization α for glucose- LABS complex/aggregate**

| S.No. | [glucose] moldm$^{-3}$ | csc mol dm$^{-3}$ | cac moldm$^{-3}$ | mole fraction of water | Degree of ionization( $\alpha$ ) at | | $\lambda_0$ Scm$^2$mol$^{-2}$ | |
|---|---|---|---|---|---|---|---|---|
| | | | | | 301K | 311K | 301K | 311K |
| 1 | 0.1 | 0.002 | 0.0015 | 54.94 | 0.989 | 0.729 | 429.98 | 508.09 |
| 2 | 0.2 | 0.002 | 0.0015 | 53.73 | 0.926 | 0.770 | 406.13 | 488.6 |
| 3 | 0.3 | 0.002 | 0.0015 | 53.51 | 0.737 | 0.730 | 381.56 | 483.76 |
| 4 | 0.4 | 0.002 | 0.0015 | 52.98 | 0.833 | 0.768 | 372.19 | 419.44 |
| 5 | 0.5 | 0.002 | 0.0015 | 52.21 | 1.00 | 0.950 | 375.59 | 446.43 |

The specific conductance data is linearly related, to the concentration of LABS in aqueous glucose solution, with two break points, in presence of glucose. There is only one break point in the plot at the cmc of LABS in water. The first break point in the plots corresponds to the concentration of LABS at 0.0015 mol dm$^{-3}$ as assessed from gradient values obtained from the data at concentration 0.0008, 0.001, 0.0012, 0.0015 mol dm$^{-3}$ and gradient from data 0.002, 0.003, 0.004, 0.006, 0.008 moldm$^{-3}$. This concentration indicates the critical aggregation concentration, cac, of glucose surfactant, where the formation of micelle like aggregate takes place in presence of glucose.

The addition of glucose, which have shown rise in critical micelle concentration values of LABS, is in agreement of the result obtained in case of anionic surfactant sodium lauryl sulphate [22] whereas the second break point observed in the conductance versus surfactant concentration plot is also reported as cmc of polymer surfactant complex or polymer saturation point in aqueous system [27]. The behavior of LABS micelles in aqueous glucose solution is somewhat similar to the observed behavior in aqueous glycine solution [28].

The micellization of LABS under experimental conditions can be explained by the following considerations:

**(i)** The glucose behave as a weak electrolyte as shown by molecular conductance and concentration in the study and as reported [29] in the literature. The ionization of glucose can takes place and it acts as weak acid.

$$\underset{\text{glucose}}{C_6H_{12}O_6} \xrightarrow[12.87]{pk_a} C_6H_{11}O_6^- + H^+ \text{.....} \textbf{(1)}$$

The plot between molecular conductance and square root of concentration, √c, of surfactant also supports the above fact that the glucose and LABS both are weak electrolyte in low concentration region and also indicated by values of $\lambda_0$ Table 2, which decrease with increase in concentration of glucose.

**(ii)** The specific conductivity versus concentration plot between the concentration 0.0015 to 0.002 mol $dm^{-3}$ of LABS are indicative of the interaction between glucose molecule and LABS in this concentration region, which is independent of glucose concentration. The occurrence of this concentration region is only possible when there is a dynamic equilibrium between transient micellar structure and glucose molecules.

**(iii)** The oxygen atom of glucose molecule having partially negative charge may establish interaction with surfactant molecules. The presence of ions gives an increase of the electrostatic field in the solution whereas the solvent water is strongly polar. Hence, the ionization of glucose cannot be explained only by assuming an increase of hydration of glucose. The electrostatic field combined with intra-molecular hydrogen bonding must have a dominating interaction effect. This interaction corresponds to the first break point and can be defined as, cmc 1 or critical aggregation concentration, cac, of glucose – LABS system in specific conductance and concentration plot.

**(iv)** The second break point corresponds to cmc 2 or critical saturation concentration, csc, of glucose – LABS system. Beyond this point additional glucose molecules around aggregate/ structure creates the negative charge which is responsible for the repulsion between micelles and glucose molecules. In this region the equation of strong electrolyte is applicable and the values of molecular conductance decreases with increasing concentration of glucose.

**(v)** The value of, cac, is 25% higher from, cmc, of LABS in aqueous glucose solution. The hydration of glucose can be considered as hydration of hydroxyl group, involving hydrogen bonding as well as further hydration of hydrate water molecules. After ionization, the hydroxyl groups are oriented and established by intra-molecular hydrogen bonding. Consequently, no hydroxyl groups of glucose are then available for bonding with water molecules which leads to hydrophobic interactions. It can be explained in terms of hydrogen bonding ability of glucose molecule through -OH groups it leads to weak contribution to the hydrophobic interaction, which is main driving force for the formation of micelle and hence the value of cac > cmc value. The structure maker property of glucose molecule in water has less significant influence due to the aggregation. Therefore hydrogen bonding ability is an entropy directed process. In order to verify this specific effect of glucose on micellization of LABS in aqueous solution, the value of cac and csc measured in the aqueous solution of D- mannose, the observed values are 0.002 and 0.0025 $moldm^{-3}$.The plot between specific conductance versus concentration of D- mannose which is an epimer of glucose. It is indicative of the explanation for increase in cmc of LABS on addition of glucose is justified. For D- mannose the higher value of cac and cmc is due to change in hydrogen bonding ability and orientation at second carbon of the molecule.

*3.1 Thermodynamic quantities of micellization*

In the present study, it is concluded that micelle/ micellar aggregate is formed by the interaction between glucose molecule and surfactant monomer. The csc value is independent of the concentration of glucose and temperature but the free energy of micellization will be different with the change in mole fraction of surfactant in different concentration of glucose. At the csc the degree of ionization, α, of glucose-LABS complex is computed from the slopes of conductance-concentration plot below and above csc. The obtained α value of glucose-LABS complex is found in the range 74% to 99%, when independently concentration ranges were taken with the elimination of common readings in the plot. The free energy of micellization depends upon the various factors related to the orientation of the molecule with micro environmental conditions. The degree of ionization, α, value were used to calculate the free energy of micellization $\Delta G°_{mic}$ at csc using the relation

$$\Delta G°_{mic} = (2- \alpha)\, RT \ln csc \text{ ..... } \textbf{(2)}$$

where csc is in the mole fraction scale. The calculated values of α at 301K and 311K are given in Table-2. The values of $\Delta G°_{mic}$ at different concentrations of glucose are present in Table-3.

**Table 3. Thermodynamic parameters at csc in glucose –LABS system**

| S.No. | [Glucose] moldm$^{-3}$ | - Δ G$^0_{mic}$ kJmol$^{-1}$ | | - Δ S$^0_{mic}$ kJmol$^{-1}$K$^{-1}$ | - Δ H$^0_{mic}$ kJmol$^{-1}$ | | Binding capacity m.mol/mol |
|---|---|---|---|---|---|---|---|
| | | 301K | 311K | | 301K | 311K | |
| 1 | 0.1 | 25.86 | 33.59 | 0.773 | 206.81 | 208.81 | 50 |
| 2 | 0.2 | 27.41 | 32.43 | 0.502 | 123.69 | 123.69 | 25 |
| 3 | 0.3 | 32.23 | 33.43 | 0.125 | 5.39 | 5.44 | 17 |
| 4 | 0.4 | 29.75 | 32.46 | 0.271 | 51.82 | 51.82 | 12 |
| 5 | 0.5 | 25.47 | 27.63 | 0.216 | 39.54 | 39.54 | 10 |

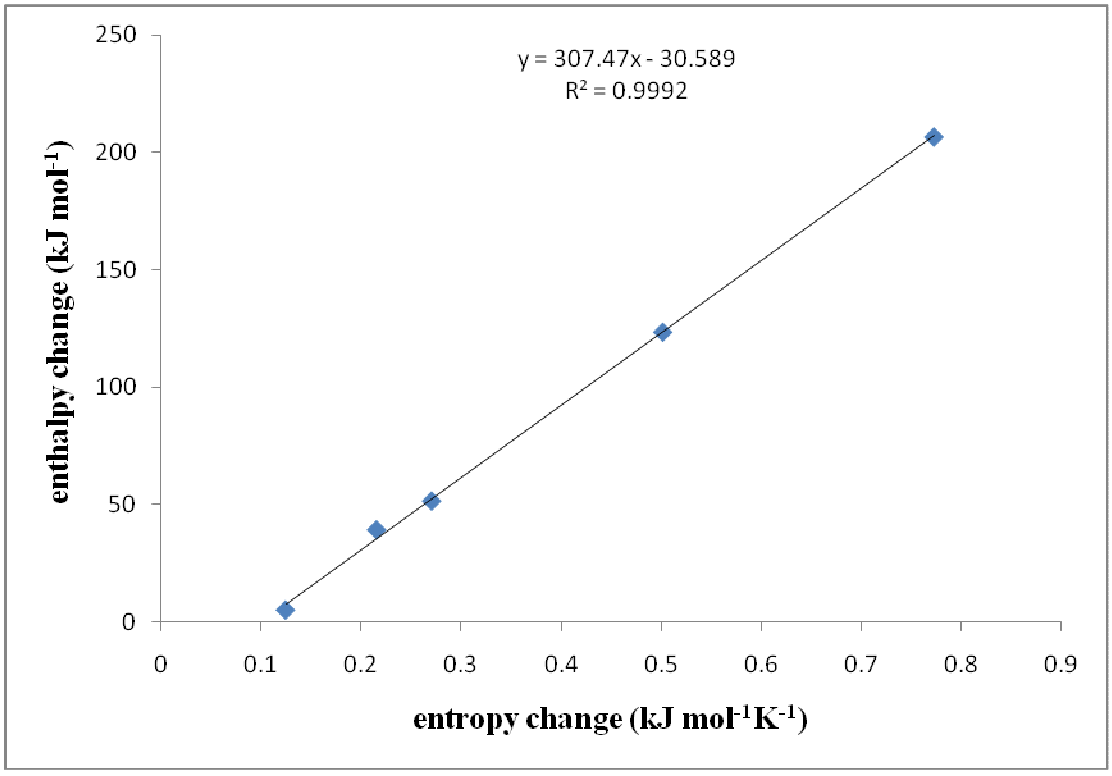


**Fig. 3 Plot of enthalpy versus entropy change with concentration of glucose 0.1moldm$^{-3}$ to 0.5 moldm$^{-3}$ at 301K**

The entropy and enthalpy have same negative sign and free energy is also negative. It indicates that in the process bonds are formed. The process is spontaneous depending upon the magnitude of temperature, entropy and enthalpy. The minimum value of enthalpy at 0.3 mol dm$^{-3}$ indicates the formation of more stable aggregate/micelle or optimal condition for the micellization or more hydrophobic bonding. The values of Δ G°$_{mic}$ increases (becomes less negative) on increasing the concentration of glucose up to 0.3mol dm$^{-3}$. This indicates that the strong interaction between LABS monomers and glucose molecules in this concentration region at 301K. At higher temperature the magnitude of these interactions is less effective due to increase in thermal energy. The standard entropies were calculated from the Δ G°$_{mic}$ for two temperatures using the relation given by equation (3) and the Δ H°$_{mic}$ values were calculated at each concentration of glucose using equation(4) .The values of cac and csc remain same with increase of temperature.

$$\Delta S^0_{mic} = \frac{-\delta(\Delta G^0_{mic})}{\delta T} \quad \text{..... (3)}$$

$$\Delta H°_{mic} = \Delta G°_{mic} + T\,\Delta S°_{mic} \quad \text{..... (4)}$$

The Δ H°$_{mic}$ values are positive indicates the interactions seems to be endothermic in nature. On plotting enthalpy versus entropy change at concentration of glucose 0.1 moldm$^{-3}$ to 0.5 mol dm$^{-3}$, a linear co- relation was obtained having linearity coefficient 0.99 in Figure 3. At around Δ S°$_{mic}$ value of 0.111 Jmol$^{-1}$K$^{-1}$, Δ H°$_{mic}$ becomes zero in this condition Δ G°$_{mic}$ = - TΔS°$_{mic}$ , this indicates that the glucose surfactant interaction in this condition favoured only by the entropy change. Such enthalpy, entropy compensations effect is reported for many physicochemical

processes [30]. From this plot, the slope value is 311.02 K. At 311K the micellization process is totally independent of structure changes of the system and depends on the enthalpic forces [27].

The $\Delta H°_{mic}$ values remain constant with the increase of temperature for each concentration of glucose. This indicates that glucose surfactant interactions are more favoured by entropy change. The more positive value of entropy of micellization below 0.2 mol $dm^{-3}$ of glucose is confirmed by the plot between specific conductivity versus concentration of glucose having maxima at concentration 0.2 mol $dm^{-3}$ of glucose, the illustrative plot is given in Figure 4.

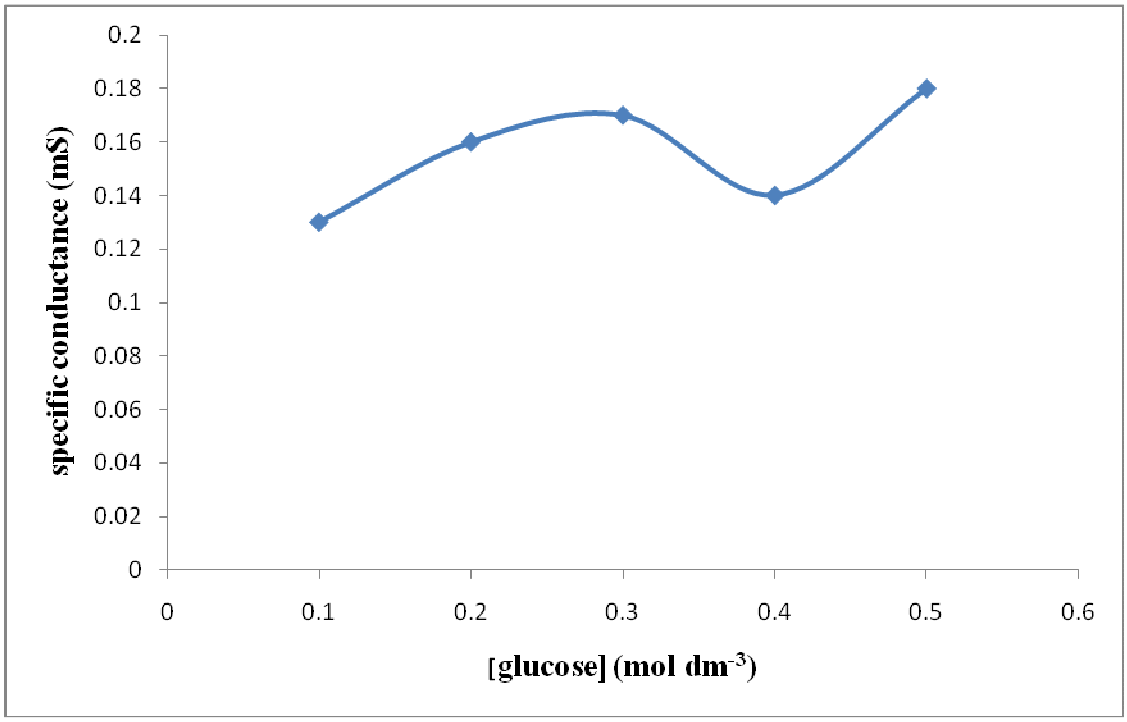


**Fig. 4 Plot between [glucose] and specific conductance with LABS at 0.0008 mol $dm^{-3}$ at 301K**

It can be concluded that the glucose molecules are participating in micelle / aggregate formation with LABS and acting as co-surfactant. The glucose concentration 0.2 mol $dm^{-3}$ is the concentration required to form stable aggregate where as below this concentration a dynamic equilibrium is possible between LABS and glucose molecules. This nature of curve changes by increase the concentration of glucose, the maxima of the curve disappear and the plot tends to follow a linear trend with a point of inflection at maxima. The standard Gibbs free energy of transfer of surfactant from pure aqueous solution to glucose rich aqueous phase is calculated using the relation (5).

$$\Delta G_{tr}^{0} = RT \ln \frac{cmc}{cac} + RT \ln \frac{f}{f^{a}} \quad \text{.....(5)}$$

The value of cmc for LABS is taken 0.0012 literature value [24]. The value of $f/f^{a}$ ratio of activity coefficients of surfactant in lower concentration can be taken as unity. The calculated value of standard free energy of transfer is 1.94 kJ $mol^{-1}$ at 301K. The positive magnitude indicates the non feasibility of transfer of LABS from pure water to glucose rich water phase. This indicates the micellar formation in the presence of glucose is taking place due to the formation of LABS- glucose complex / aggregate. At csc, the adsorption at charged sides of glucose molecules by LABS monomers and csc shows the formation of independent micelles after saturation point. A limited number of LABS- glucose aggregate / complex micelle can be formed through the interactions. The binding capacity of LABS is calculated [28] at different concentrations of glucose. The values are given in Table-3.

*3.2 Temperature effect*

The examination of the data in Table-3 reveals that the increase in temperature results in the lowering of the standard free energy of micellization $\Delta G°_{mic.}$ It is due to water structure breaking at high temperature and hence the decrease in hydration of surfactant head groups. The $\Delta H°_{mic}$ values are negative and $\Delta S°_{mic}$ values are positive at saturation. Thus the micellization / aggregation is favoured both by entropy gain as well as enthalpy effect. At the

saturation point $\Delta H°_{mic}$ values are positive, $\Delta S°_{mic}$ value are positive. Thus the aggregation is favored only by the entropy gain, due to the saturation by the glucose molecule.

## CONCLUSION

The decrease in molecular conductance with increasing concentration of glucose in post saturation region confirms the formation of LABS – glucose aggregate and the reported structure maker character [26] of glucose in aqueous solution. The structure making character of glucose becomes more effective above 0.5 mol $dm^{-3}$ concentration as the value of binding capacity tends to minimum value.

The micellization of LABS in presence of aqueous glucose solutions has shown that the glucose behave as a weak electrolyte and can form aggregates through interactions with LABS. In aqueous solution further increase in concentration of surfactant which corresponds to the saturation points, beyond which micellization of surfactant followed a regular trend of post micellization like in water. It suggests the association and ionization of glucose – LABS aggregate / micelle, which is justified by the data of binding capacities at different concentration. The free energy of micellization in this study has same trend as it is reported for non ionic surfactant in glucose solution [22]. The study may be a model to the surfactant systems which are applicable in interpretation of the biological, catalytic and industrial formulation of commercial importance. The results obtained in the study indicates that the transition in the micellization occur over a very small concentration region rather than at a specific concentration as mentioned in the literature [31].

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