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Conductivity Study of DTAB in Water and Ethanol-Water Mixture in the Presence and Absence of $ZnSO_4$

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Abstract:

The physicochemical behaviour of dodecyltrimethylammonium bromide (DTAB) in water and ethanol-water mixture in the presence and absence of $ZnSO_4$ were studied by measuring the conductivity at 298.15 K. The pre-micellar (S_1) and post-micellar slopes (S_2) were obtained and calculated the degree of dissociation (α) and the critical micelle concentration (cmc). With an increase in ethanol content, the cmc and α of DTAB increased whereas, in the presence of $ZnSO_4$, the cmc and α decreased. By using cmc and α, thermodynamic properties as the standard free energy of micellization ($\Delta G_\mathrm{m}^\mathrm{o}$) were evaluated. With an increase in ethanol content, the negative values of $\Delta G_\mathrm{m}^\mathrm{o}$ are decreased indicating less spontaneous in the micellization which makes micellization less favourable. The correlation of the pre-micellar and post-micellar slopes with the volume fraction of ethanol were discussed. DTAB micellization was tested in contexts of specific solvent parameters and solvophobic parameter.

Key words: DTAB, Ethanol-water, Solvent parameters, Solvophobic parameter, $ZnSO_4$

Introduction:

Surfactants have a hydrophilic head and hydrophobic tail and are vigorously used in most of the areas of applications (1). In a given concentration, surfactants display dramatic differences in behaviors, named cmc, which implies the basic micelle ability of amphiphilic molecules to self-aggregate in the solutions. In contrast, the surfactant composition, the solubilizing media, the concentration of surfactants and the procedure used to create self-assemblies are all highly important for the creation of micelle (2). The cmc is primarily dependent upon its chemical composition; however, this may even influence temperature & various materials (mostly heavy electrolytes). The conductance measurement of surfactant solutions is the principal way of calculating cmc (3,4). The conductivity methods are very popular nowadays to calculate the cmc of cationic surfactant (5), anionic surfactant (6), nonionic surfactant (7) and even in zwitterionic surfactant (8).

The study on the effects of salts in cationic surfactants by conductivity methods is still useful for the researchers (9-11). Such types of research results play a very important role in a broad area of biological and physicochemical phenomena from the electrolyte effects and surfactant solutions to the transport across biological membranes and enzyme activities which are widely used for the public benefits (12). The micellar solutions of alkyltrimethylammonium bromide surfactants in the presence of KBr are widely used in various household, industrial and research applications (13).

There were some works on the effects of salts in DTAB from the different methods (13-15). In 2016, Chauhan et al measured the density and speed of sound of DTAB in aqueous solutions of tetraalkylammonium bromides at (293.15-318.15) K to calculate the first critical micelle concentration and second critical micelle concentration (16). Recently, Sharma et al studied the effects of sodium salicylate on the dynamic behavior of DTAB micelles using dynamic light scattering and quasielastic neutron scattering techniques in the year 2017(17). But there were very few papers related to conductivity measurement of DTAB in the presence of salts. Pal et al used the conductivity technique to see the effects of salt in DTAB and did not notice the cmc (18). However, there was the study of the effect of salt on the cmc of DTAB in the aqueous medium (19) and found the cmc values of DTAB decline by adding tetra-alkylammonium bromide salts. The reason for decreasing the cmc values of DTAB on adding tetra-alkylammonium bromide salts may be due to the synergistic hydrophobic interactions between the non-polar hydrocarbon chains of DTAB and alkyl chains of tetra-alkylammonium bromide salts. The effects of sodium salts NaCl, NaH_2PO_4, Na_2HPO_4, Na_2CO_3, Na_2SO_4, Na_3PO_4 and Na_3Cit on the aggregation behavior of DTAB in aqueous solutions was studied and calculated the values of cmc as well as other parameters. The decrease of cmc was noticed by adding the salts and was found more pronounced for the anions with higher valency than that with lower valency (20). It was mentioned in the literature (14) that the conductance method had limitations to locate the cmc at increased NaBr concentration in comparison to the calorimetry method. Moreover, the cmc of DTAB decreased continuously with the addition of NaBr for both methods (21).

There is no conductivity study to calculate the cmc of DTAB in the presence of $ZnSO_4$. Our work aims is to see the effects of $ZnSO_4$ on the micellisation of DTAB in the ethanol-water mixture from the conductivity methods which is the noble study to calculate the various parameters as S_1, S_2, α, cmc, ΔG_m^o as well as the correlation of ΔG_m^o with solvent parameters and solvophobic parameter.

Materials and Methods:

Apparatus and chemicals

The digital conductivity meter systronics of cell constant 1.002 $cm^{-1}$was purchased from India. The digital balance was also purchased from India to get exact weights of different required experimental solutions of dodecyltrimethylammoniumbromide (DTAB) of molecular weight 308.3 g/mol. $ZnSO_4$ and DTAB (Figs.1 and 2) were purchased from SdFINE CHEM Ltd., India. Ethanol was obtained from E.Merck, India. The double-distilled water was taken for preparing the solvent composition of water and ethanol mixture.

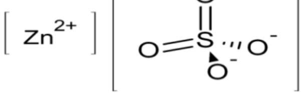

Figure 1. Structure of $ZnSO_4$

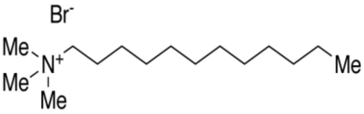

Figure 2. Structure of DTAB

Dodecyltrimethylammonium bromide and salt solution preparation:

Double distilled water containing a fair conductivity under 10^{-6} S/cm was utilized during the formulation of the mixture at 298.15 K. Double distilled water and 0.10 to 0.30 volume fraction containing ethanol were used to make 0.04 M dodecyltrimethylammonium bromide solution in the presence and absence of zinc sulphate.

Conductance measurement:

The specific conductance of dodecyltrimethylammonium bromide solution in the presence and absence of ZnSO4 was measured on the conductivity meter. The cell was calibrated with the help of aqueous KCl solution (0.1 Demal and 0.01 Demal) (22). Such type of calibration with the help of aqueous potassium chloride was also found in the literature (23).

Results and Discussion:

The specific conductance versus concentration can be shown if a molar conductance is replaced by the specific conductance ($\lambda = \kappa/c$). The cmc is the interaction point of two linear straight lines and can be calculated by solving two linear equations described simultaneously. The degree of dissociation (α) in the plot of the specific conductivity with the concentration of surfactant is derived from slopes over and under the cmc proportion:

$$\alpha = S_2/S_1 \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (1)$$

Where, S_1 is the slope of pre-cmc and S_2 is the slope of post-cmc (24-25). In the ethanol-water mixture, both slopes (S_1 and S_2) decline along with the rise of ethanol.

Determination of cmc of DTAB in the absence of salt:

The conductance of dodecyltrimethylammonium bromide in pure water as well as in three separate mixtures of ethanol-water (comprising 0.10, 0.20 and 0.30 proportions of the volume of ethanol) for measuring the cmc at 298.15 K was determined (Fig. 3).

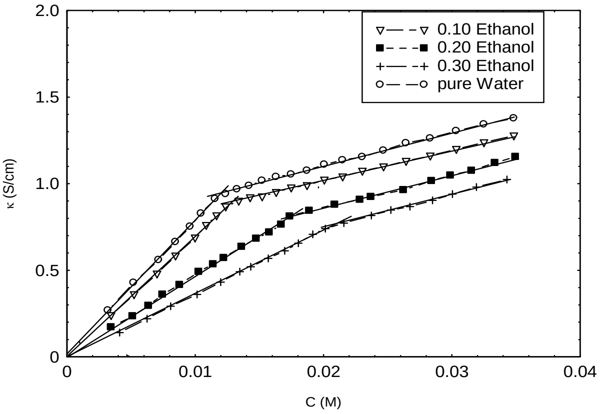

Figure 3. The plot of concentration vs conductance for DTAB in pure water and various ethanol-water mixtures

From Fig. 3, the conductance of dodecyltrimethylammonium bromide at 298.15 K in pure water as well as in three separate compositions of ethanol-water, the conductance reduces when alcohol concentration in mixed solvent systems increases. The pre-cmc slope (S_1) and post-cmc slope (S_2) decrease with the addition of ethanol (Table 1), cmc and α increases with the addition of ethanol (Table 2).

Determination of cmc of DTAB in the presence of 0.005 M salt:

The conductance of dodecyltrimethylammonium bromide in pure water as well as in three separate mixtures of ethanol-water in the presence of 0.005 M salt for measuring the critical micelle concentration at 298.15 K was determined (Fig. 4).

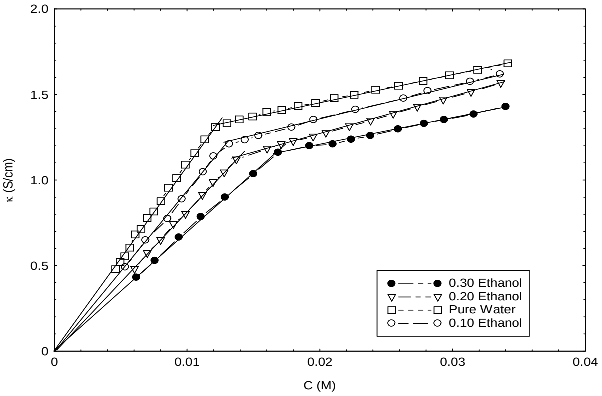

Figure 4. The plot of concentration vs conductance for DTAB in presence of 0.005M $ZnSO_4$, in pure water and various ethanol-water mixtures

From Fig. 4, the conductance of dodecyltrimethylammonium bromide in the presence of 0.005 M $ZnSO_4$ at 298.15 K in pure water as well as in three separate compositions of the ethanol-water binary mixture, the conductance reduces when alcohol concentration in mixed solvent systems increases. The S_1 and S_2 decrease with the addition of ethanol (Table 1) but higher values of S_1 and S_2 in comparison with the DTAB system.

Determination of cmc of DTAB in the presence of 0.01 M salt:

The conductance of dodecyltrimethylammonium bromide in pure water as well as in three separate mixtures of ethanol-water in the presence of 0.01M salt for measuring the critical micelle concentration at 298.15 K was determined (Fig. 5).

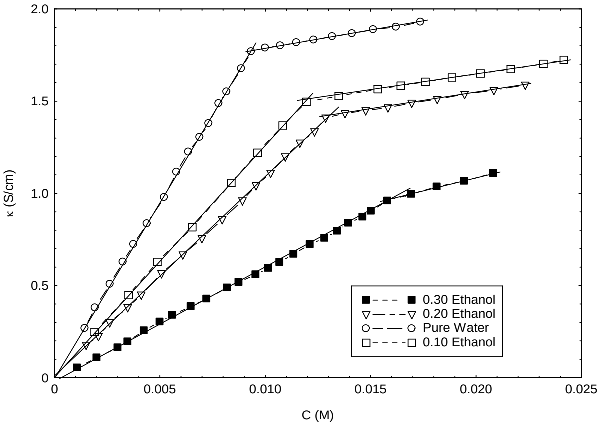

Figure 5. The plot of concentration vs conductance for DTAB in presence of 0.01M $ZnSO_4$, in pure water and various ethanol-water mixtures

From Fig. 5, the conductance of dodecyltrimethylammonium bromide in the presence of 0.01 M ZnSO4 at 298.15 K in pure water as well as in three separate compositions of ethanol-water, the conductance reduces when alcohol concentration in mixed solvent systems increases. The S_1 and S_2 decrease with the addition of ethanol (Table 1) but higher values of S_1 and S_2 in comparison with DTAB in 0.005 M ZnSO4 system. The cmc and α increase with the addition of ethanol but lesser values of cmc and α in comparison with DTAB in the presence of 0.005M ZnSO4 and 0.01M ZnSO4 system (Table 2).

The value of DTAB's degree of ionization 'α' is found to be higher in the presence of ethanol than in its absence. With a rise in the concentration of ethanol 'α' value continues to rise as a result of the rise in the surface area of the ionic head group facilitating counter ionizations (26). The evidence indicates that the cmc, as well as α, is clarified by the volume fraction of ethanol is firstly due to the binding counter ions of alcohol molecule between surfactant ions, which for steric reasons raises the average length among ionic head groups and the dielectric constant of the palisade layer is the second consequence. Here the cmc obtained in the present work for the investigated method is consistent with the literature values (5).

Moreover, the cmc of dodecyltrimethylammonium bromide (DTAB) in ethanol is higher in different concentrations of ethanol and pure water but this decreases concurrently in the presence of $ZnSO_4$ of 0.005 M and 0.01M respectively. The fall in cmc is due to the stronger synergistic influence when $ZnSO_4$ is added to the DTAB. With the increase in concentrations of salt, α as well as cmc also further decreases. Such types of results were reported in the previous literature (27).

Table 1. S_1 and S_2 for the DTAB-ethanol, DTAB-0.005 M $ZnSO_4$ and DTAB-0.01 M $ZnSO_4$

Vol. fraction of ethanol	without salt (S_1, S_2) (S/cm*M)	with 0.005M salt (S_1, S_2) (S/cm*M)	with 0.01 M salt (S_1, S_2) (S/cm*M)
0.00	77.0, 19.0	110, 26.3	173, 22.1
0.10	58.0, 15.2	108, 25.9	126, 17.5
0.20	49.0, 14.2	85.1, 22.3	108, 16.7
0.30	43.3, 14.0	44.5, 12.7	77.2, 12.4

Representation of variations of the slopes:

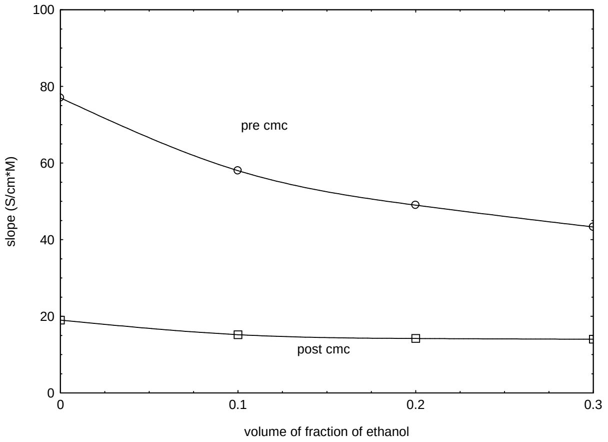

Figure 6. Slope variance with the volume fraction of ethanol of DTAB solutions

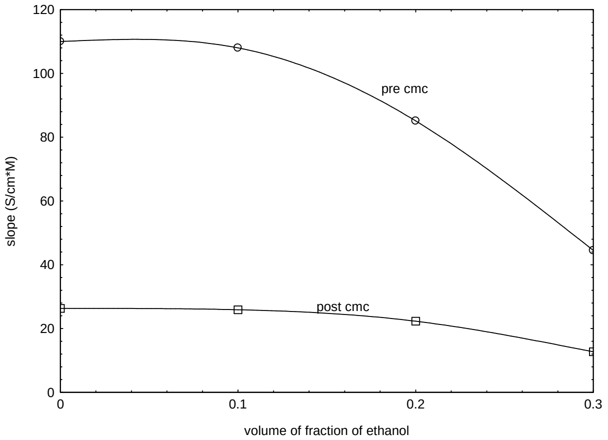

Figure 7. Slope variance with the volume fraction of ethanol of DTAB-0.005M $ZnSO_4$ solutions

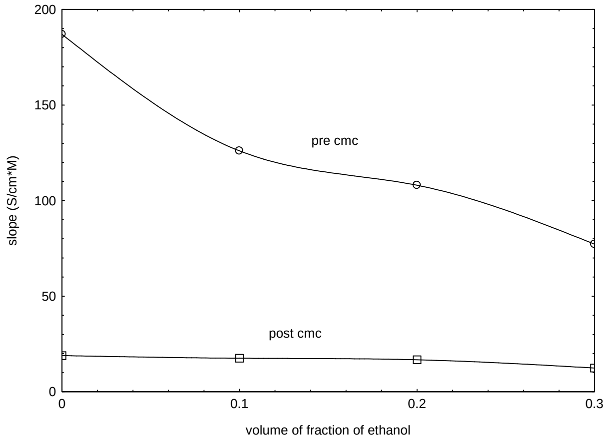

Figure 8. Slope variance with the volume fraction of ethanol of DTAB-0.01M $ZnSO_4$ solutions

From Figs.6, 7 and 8, the curves of the pre-cmc slopes are higher than the curves of the post-cmc slopes with the addition of ethanol in the absence and presence of $ZnSO_4$ in DTAB. The fitting of the curve of the pre-cmc slope was done by the polynomial equation having correlation coefficient, $r^2 = 1$ with the sharp decreases on the addition of ethanol whereas post-cmc slopes curve was also done by the polynomial equation having correlation coefficient, $r^2 = 1$ with the monotonous decrease on the addition of ethanol.

Table 2. cmc and α for the DTAB, DTAB-0.005 M $ZnSO_4$ and DTAB-0.01 M $ZnSO_4$ systems

vol. fraction of ethanol	without salt [cmc (mM), α]	with 0.005 M salt [cmc (mM), α]	with 0.01M salt [cmc (mM), α]
0.00	12.72, 0.24	10.26, 0.23	10.22, 0.13
0.10	14.89, 0.26	12.81, 0.24	11.92, 0.14
0.20	16.65, 0.29	13.74, 0.26	13.16, 0.15
0.30	21.15, 0.32	20.26, 0.28	14.33, 0.16

Calculations of thermodynamic parameters:

The micelle-forming free energies are determined using the following equation (28):

$$\Delta G_{m}^{o} = RT(2-\alpha)\ln X_{CMC} \ldots\ldots\ldots\ldots\ldots(2)$$

where R= gas constant and T= temperature

This is clear through Table 3; ΔG_m^o is negative in all cases but is less negative as the ethanol concentration in the mixture increases, suggesting that micelle development at a greater ethanol concentration is less spontaneous. It shows the accumulation of greater ethanol output is less favorable. Koya et. al (29) and many other study groupings have also identified related trends (30). It becomes less negative in 0.005 M and 0.01M respectively in the presence of salt $ZnSO_4$.

Table 3. ΔG^o_m of DTAB and DTAB-ZnSO$_4$ solutions in water and ethanol-water mixture

Vol. fraction of ethanol	ΔG^o_m (kJ/mol) (DTAB)	ΔG^o_m (kJ/mol) (DTAB+0.005 MZnSO$_4$)	ΔG^o_m (kJ/mol) (DTAB+0.01M ZnSO$_4$)
0.00	-16.56	-15.38	-13.23
0.10	-15.44	-14.44	-12.67
0.20	-14.43	-13.80	-12.16
0.30	-12.91	-12.11	-11.64

Variation of ΔG^o_m with solvent parameters:

The different physical quantities like a dielectric constant(*D*), viscosity(η), Reichardt's parameter (E_T), Gordon parameter(*G*) and solvophobic parameter (S_p) are very important physical parameters and their relation with the free energy change ΔG^o_m(Figs. 9-13) gives not only the stability of the solution but also relate micellization of molecular association, the fluidity, polarity, and solvent structure (31). The values of *D, G*, η, E_T and S_P were taken from Table 4.

Table 4. Various physicochemical parameters of ethanol-water mixture at 298.15 K [31]

vol. fraction of ethanol	Dielectric constant(D)	Gordon parameter (G/J m^{-3})	Coefficient of viscosity (η /mPa·s)
0.00	78.2473	2.74	0.8959
0.10	73.5294	1.72	1.1441
0.20	70.2247	1.39	1.4346
0.30	66.0066	1.16	1.7371

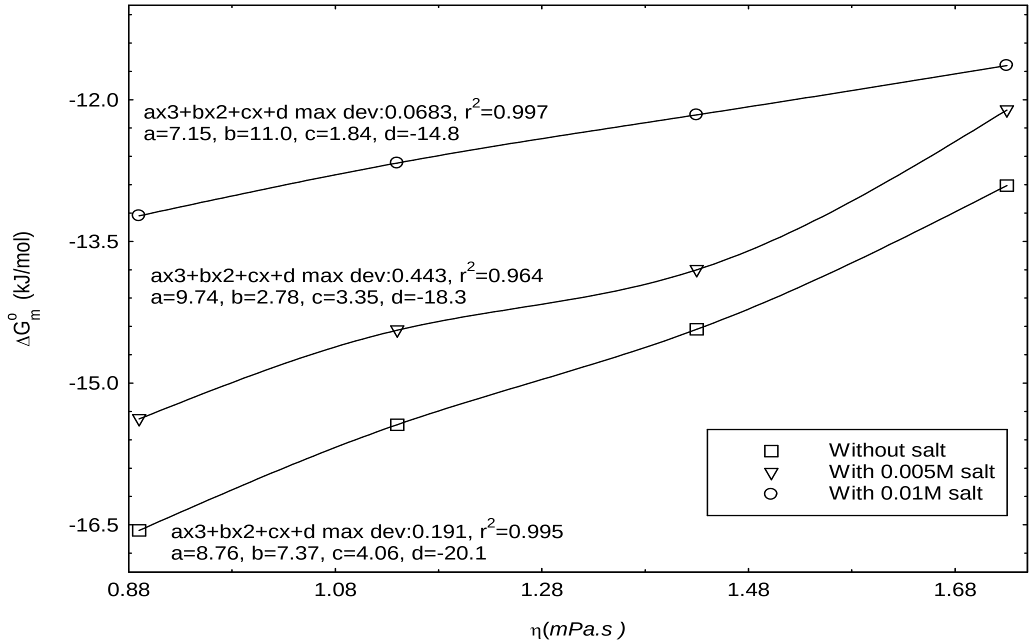

Figure 9. Graph of Gibbs free energy vs coefficient of viscosity in pure water and availability of salt concentrations

For ΔG^o_m vs η at 298.15 K, DTAB in 0.01M ZnSO$_4$ has a much high fitting curve (look linearly increase) with the polynomial equation of correlation coefficient, $r^2 = 0.997$ in comparison with DTAB in 0.005M ZnSO$_4$ where the fitting curve (monotonously increase) with the polynomial equation of correlation coefficient, $r^2 = 0.964$ with the addition of ethanol. The fitting curve for DTAB increases sharply on the addition of ethanol with the polynomial equation of the correlation coefficient, $r^2 = 0.995$. It is observed that both the curves are higher than the curve of DTAB only (Fig.9).

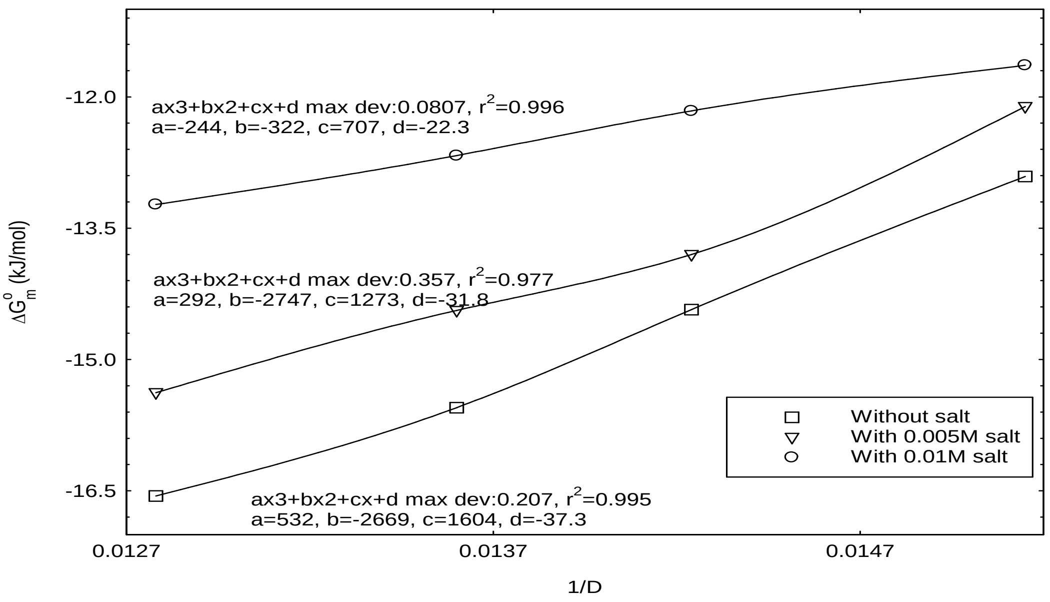

Figure 10. Graph of Gibbs free energy versus inverse dielectric constant in pure water and availability of salt concentrations

For ΔG_m^o vs 1/D at 298.15 K, DTAB in 0.01M $ZnSO_4$ has a much high fitting curve (look linearly increase) with the polynomial equation of correlation coefficient, $r^2 = 0.996$ in comparison with DTAB in 0.005M $ZnSO_4$ where the fitting curve (monotonously increase) with the polynomial equation of correlation coefficient, $r^2 = 0.977$ with the addition of ethanol. The fitting curve for DTAB increases sharply on the addition of ethanol with the polynomial equation of the correlation coefficient, $r^2 = 0.995$. It is observed that both the curves are higher than the curve of DTAB only (Fig. 10).

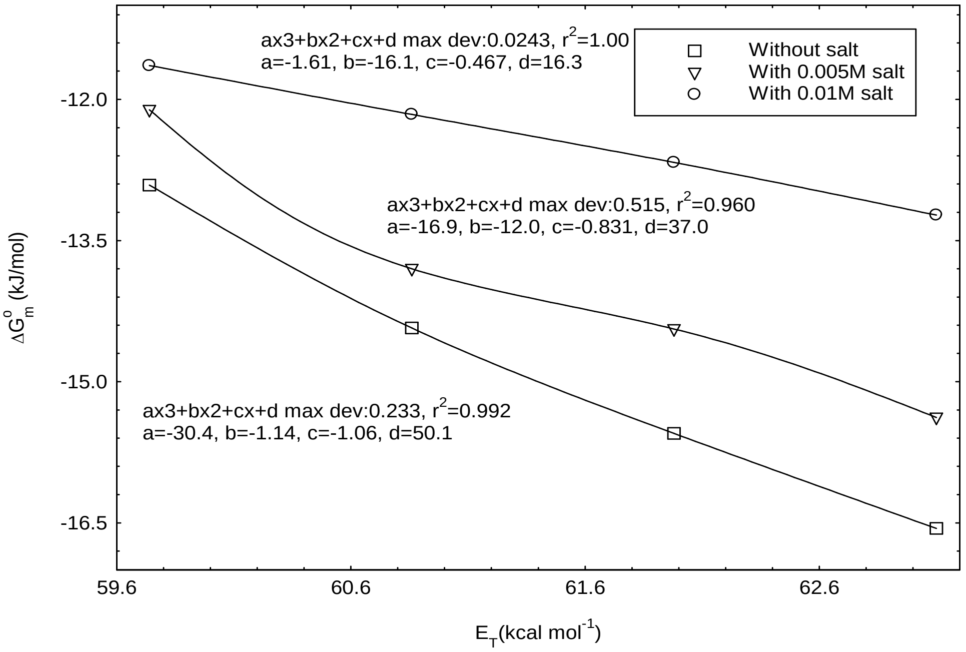

Figure 11. Graph of Gibbs free energy vs parameter of Reichardt in pure water and the presence of salt concentrations

For ΔG_m^o vs E_T at 298.15 K, DTAB in 0.01M $ZnSO_4$ has a high curve with the correlation coefficient, $r^2 = 1$ in comparison with DTAB in 0.005M $ZnSO_4$ with a monotonous increase having polynomial equation of correlation coefficient, $r^2 = 0.960$ on the addition of ethanol. The fitting curve for DTAB increases sharply on the addition of ethanol with the polynomial equation of the correlation coefficient, $r^2 = 0.992$. It is observed that both the curves are higher than the curve of DTAB only (Fig. 11).

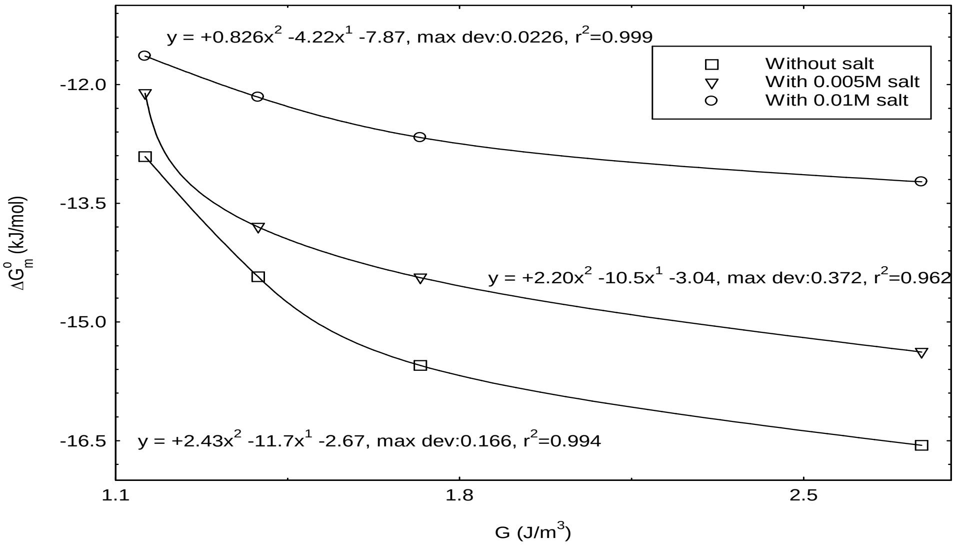

Figure 12. The plot of Gibbs free energy vs Gordon parameter in pure water and the presence of salt concentrations

Some researchers (32-33) reported linear relations between ΔG^o_m and G for several surfactants in mixed aqueous-organic solvents. In our case ΔG^o_m vs *G* at 298.15 K, DTAB in 0.01M $ZnSO_4$ has a higher somehow convex curve with correlation coefficient, $r^2 = 0.999$ in comparison with DTAB in 0.005M $ZnSO_4$ with a monotonous decrease (convex curve) having polynomial equation of correlation coefficient, $r^2 = 0.962$ on addition of ethanol. The convex fitting curve for DTAB decreases sharply on the addition of ethanol with the polynomial equation of correlation coefficient, $r^2 = 0.994$. It is observed that both the curves are higher than the curve of DTAB only (Fig. 12).

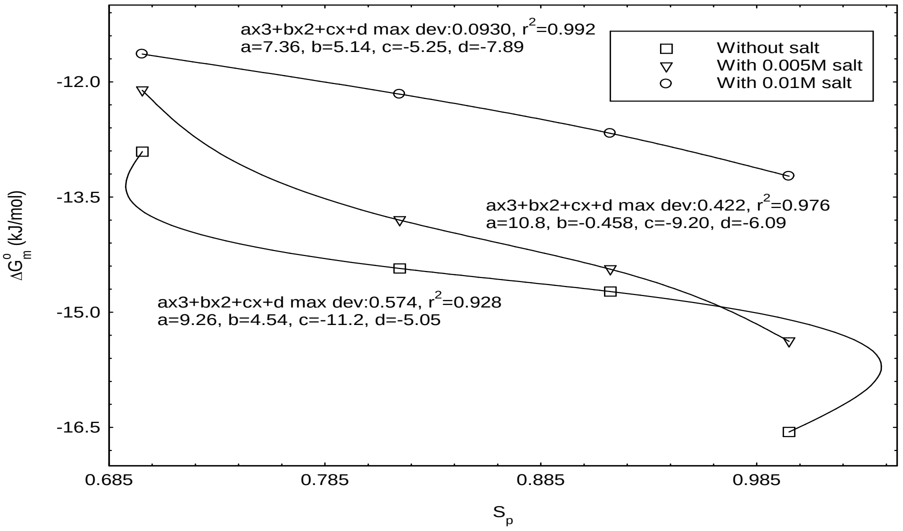

Figure 13. The plot of Gibbs free energy versus solvophobic parameter (S_p) at room temperature in pure water and the presence of salt concentrations

For ΔG^o_m vs S_p at 298.15 K, DTAB in 0.01M $ZnSO_4$ has a high somehow linear increase curve with the correlation coefficient, $r^2 = 0.992$ in comparison with DTAB in 0.005M $ZnSO_4$ with an increase curvilinear having polynomial equation of correlation coefficient, $r^2 = 0.976$ on the addition of ethanol. The curvilinear fitting curve for DTAB increases sharply on the addition of ethanol with the polynomial equation of correlation coefficient, $r^2 = 0.928$. It is observed that both the curves are higher than the curve of DTAB only (Fig. 13).

Besides it can be seen from Fig. 13 that the ΔG^o_m values decrease with increasing Sp values on ethanol-water. This indicates that the ΔG^o_m values depend strongly on the solvophobic power of hydrocarbon in water-ethanol mixtures.

Conclusions:

The conductivity of dodecyltrimethylammonium bromide was found to increase in pure water including in three separate mixtures of ethanol-water in the presence of zinc sulphate (0.005 and

0.01) M with concentration. All pre-cmc and post-cmc slopes decline with the rise in ethanol. The cmc of dodecyltrimethylammonium bromide increases with the addition of ethanol but decreases simultaneously in 0.005M and 0.01M $ZnSO_4$. The degree of dissociation was obtained for pure water including in three separate mixtures of ethanol-water as well as in presence of zinc sulphate (0.005 and 0.01) M. It was apparent that the standard free energy of micelle formation (ΔG_m^o) in most situations is negative as the ethanol concentration in the mixture rise, it appears slightly negative, suggesting that micellization development gets less spontaneous at greater rates ethanol levels. This behaviour suggests that accumulation on low ethanol content becomes less desirable. The physical quantities like a dielectric constant(D), viscosity(η), Reichardt's parameter, Gordon parameter relation with the free energy change(ΔG_m^o) give not only the stability of the solution but also relate micellization of molecular association, fluidity, polarity and solvent structure.

Data Availability:

The authors share the data underlying the findings of the manuscript.

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Authors' declaration:

- Conflicts of Interest: None.
- We hereby confirm that all the Figures and Tables in the manuscript are mine ours. Besides, the Figures and images, which are not mine ours, have been given permission for re-publication attached to the manuscript.
- Ethical Clearance: The project was approved by the local ethical committee of Tribhuvan University.

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في الماء ومزيج الماء والإيثانول في DTAB دراسة الموصلية لـدوديسيل ثلاثي ميثيل بروميد الأمونيوم ZnSO4 وجود وغياب كبريتات الزنك

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الخلاصة:

تمت دراسة السلوك الفيزيائيى و الكيميائي لدوديسيل ثلاثي ميثيل بروميد الأمونيوم(DTAB) في الماء ومزيج الماء والإيثانول في وجود وغياب كبريتات الزنك ZnSO4 عن طريق قياس الموصلية عند 298.15 كلفن . تم الحصول على الميل قبل التركيز الحرج للمايسيل (S1) و بعده (S2) وحسبت درجة التفكك (α) وتركيز الحرج للمايسيل (cmc). مع زيادة محتوى الإيثانول وجد cmcو α ل DTAB تزداد بغياب ، في ZnSO4 بينما تنخفض بوجودها. وباستخدام cmc و α تم تقييم الخواص الديناميكية الحرارية كطاقة الحرة القياسية للمايسيل ($\Delta G_{\mathrm{m}}^{\mathrm{o}}$). مع زيادة محتوى الإيثانول ، تنخفض القيم السالبة لـ$\Delta G_{\mathrm{m}}^{\mathrm{o}}$مما يشير الى انخفاض التلقائية في تكوين المايسيل وجعلها اقل ملائمة . تمت مناقشة العلاقة بين الميل ما قبلالمايسل وما بعدالمايسيلمع نسبة حجم الإيثانول. تم اختبارتكوين المايسيل الدوديسيل ثلاثي ميثيل بروميد الأمونيوم DTABمن خلال دوال خاصة للمذيب ودالة الصفة الكارهة للمذيب

الكلمات المفتاحية: الدوديسيل ثلاثي ميثيل بروميد الأمونيوم، الإيثانول-ماء، دوال المذيب، دالة الصفة الكارهة للمذيب،كبريتات الزنك