



**Effect on CMC of sodium dodecyl benzene sulphate (SDBS) in the presence of alcohols and its derivatives at different temperatures**

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***Abstract***

Alcohols and its derivatives can be used to enhance the emulsifying properties of sodium dodecyl benzene sulfate (SDBS) based micellar system and thus making it appropriate for food applications. Exploration of interactions between the two species is the key constraint for execution of such ideas. In this work, various micellar and thermodynamic parameters of SDBS like critical micellar concentration (CMC), standard Gibbs free energy of micellization ( $\Delta G^0_{mic}$ ) etc. have been calculated in different concentrations of alcohols and its derivatives in the temperature range (298.15-318.15) K from the conductivity measurements. The parameters obtained from these studies reveal the competitive nature of the additives with SDBS for available positions at the air/water interface.

**Keywords:** Sodium dodecyl benzene sulphate; Alcohols; Conductivity, critical micelle concentration.

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### ***Introduction***

Surfactants play an essential role in the formation of microemulsions as the surfactant molecule has both hydrophilic and hydrophobic groups, that help in the stabilization of polar and non-polar groups, thereby reducing the interfacial tension between them [1]. Spontaneous emulsification occurs when the system attains ultra-low interfacial tension, as the energy required for the increase in surface area is also lowered [2]. They are gaining importance due to high stability, low interfacial tension, and low energy requirements. However, a high percentage of surfactant is required, limiting its usefulness as an economical alternative [3]. Amphi-solvents have a tendency to remain at interface since they miscible in both polar and non-polar phases of the microemulsion, and hence have been used for formulating surfactant-free microemulsions [4]. Alcohols have been usually used as amphi-solvents, replacing the need for additional surfactants [5, 6]. The concentration of surfactant needed to initiate micelle formation is called the critical micelle concentration (CMC). The value of the CMC can be determined by the change in the physicochemical properties of the surfactant solution as the concentration of the amphiphile is increased [7–10]. Some of the physical properties that have been studied for this purpose include solution detergency, viscosity, density, conductivity, surface tension, osmotic pressure, interfacial tension, refractive index and light scattering.

Mostly, surfactants are low-molecular weight compounds, so when dissolved, they form true solutions in concentration ranges below the CMC. Micelles are aggregates of a large number of simple molecules or ions of surfactants (e.g. several dozens), so the resulting size of such structures is in the colloidal range. For this reason, the micelle solutions of surfactants are regarded as association colloids. It is essential to employ physical methodologies which are highly sensitive to structural changes for determining the CMC. The existence of CMC indicates aggregation of amphiphilic molecules in solutions. The knowledge of the CMC is important for the calculation of the thermodynamic parameters, which confirms the scientific interest of a precise determination of the CMC [11]. The

CMC in aqueous solution is influenced by the degree of binding of counter ions to the micelles. For aqueous systems, the increased binding of the counter ion to the surfactant causes a decrease in the CMC and an increase in the aggregation number [12]. Since water alcohol-surfactant systems are frequently used as media in the studies of chemical equilibria and reaction rates, it is essential to investigate the effect of the nature of the alkyl groups in the alcohol on the CMC of the surfactants. Addition of alcohols to aqueous solutions of surfactants has allowed the investigation to be made of the effect of hydrophobic interactions on the micellar structure [13]. The effect of the presence of additives on the CMC of surfactants has been widely studied. It is generally accepted that the alcohol binds to the micelle in the surface region, leading to three principal effects: (a) The alcohol molecules intercalate between the surfactants ionic head groups to decrease the micelle surface area per head group and increase the ionization. This effect is correlated with modification of the growth and shape of the micelle. It seems to be a function of the mole fraction of alcohol at the micellar interface but is independent of the type of alcohol. (b) The dielectric constant at the micellar interface decreases probably due to the replacement of water molecules in the interface region by alcohol molecules. (c) The molecular order of the interface region of the micelle changes [14–19]. These were chosen because they are widely used and commercially available. Our work essentially involves the determination of the CMC by means of conductivity measurements. In the presence of various chain alcohols, the relationships among the CMC, thermodynamic functions, and the alcohol carbon number during the micellization process are discussed.

## ***2. Experimental***

### **2.1. Materials and methods**

SDBS was purchased from Sigma-Aldrich; Ethanol, 2-ethoxy ethanol and 2-methoxy ethanol supplied by Sisco Research Laboratories (SRL, India). All these chemicals were having purity higher than 99%. SDBS was dried over anhydrous  $\text{CaCl}_2$  in vacuum desiccator before use. Solutions were prepared in second stage Milli-Q water having specific resistance of 18.2 mega ohm cm. The solutions were prepared using Mettler

balance having an accuracy of  $\pm 0.01$  mg. The standard uncertainty in the molarity on an average is  $\pm 1.2 \times 10^{-5}$  mol.L<sup>-1</sup>.

## *2.2. Equipments and procedure*

### *2.2.1. Conductivity measurements*

Conductivity measurements were carried out with a digital conductivity meter (Systronics-306) at a fixed frequency of 50 Hz using a dip type cell with double walled glass jacket. The conductivity meter was calibrated using KCl conductivity standard solution ([KCl] = 0.1000 mol.L<sup>-1</sup>). The temperature of the cell was maintained using a thermostat bath (Model F25 Julabo, Germany) having a temperature control of  $\pm 0.01$  K. Sample solutions were homogenised by stirring with a magnetic bar. The reproducibility of the conductivity measurements was within  $\pm 0.2\%$ .



*Digital conductivity meter (Model: Systronics-306) connected with a temperature bath used for conductivity measurements.*

## **3. Results and Discussion**

The electrical conductances of Sodium Dodecyl Benzene Sulfate (SDBS) in water and in aqueous ethanol, 2- ethoxy ethanol and 2- methoxy ethanol solutions having concentrations 0.5, 1, 2 and 5 (%) (v/v) have been measured at 298.15, 308.15 and 318.15 K as a function of concentration. Conductance is linearly co-related to the surfactant concentrations in both the pre micellar and in post micellar regions. The

intersection of two straight line parts of the plot between conductance and concentration of SDBS determines the critical micelle concentration (CMC) of anionic surfactant SDBS [20] (Fig. 1), and the ratio between the slopes of post-micellar region to that of the pre micellar region gives the degree of counter ion dissociation ( $\alpha$ ), (Table 2). Degree of ionisation of surfactants micelles in water and in aqueous solutions of various additives has been determined by a number of workers [21-25]. The major driving force for the micelle formation is hydrophobic interactions. At room temperature, there is large gain in entropy when water molecules in hydration shell around the hydrophobic tails of the monomeric amphiphiles are released during micellization.

With increasing temperature, CMC values of SDBS increase. As the temperature increases, the degree of hydration of the hydrophilic group decreases, which favors micellization [26], however an increase in temperature also causes the disruption of water structure surrounding the hydrophobic group and this is unfavorable to micellization. In this case, second effect is predominant as CMC (Table 2) values are increasing with an increase in temperature. A decrease in the CMC values is observed when the concentration of ethanol, 2-Ethoxy ethanol and 2-Methoxy ethanol are increased. Plots of CMC values of SDBS at various temperatures vs. concentration (%) of ethanol, 2-Ethoxy ethanol and 2-Methoxy ethanol are given in Figs. 2 to 4.

The total effect of such additives on the micellization behaviour of surfactant is considered to be approximately the sum of its effects on various parts of surfactant molecule in aqueous phase. Interesting thing on the part of surfactant molecules is that the hydrophilic part of these molecules is in contact with aqueous phase both in the monomeric as well as in micellar form, while the hydrophobic groups are in contact with aqueous phase only in the monomeric form. So the hydrophobic groups of surfactants are the moieties most likely to be affected by the addition.

From the variation of CMC with temperature, thermodynamic parameters of micellization can be calculated. The standard Gibb's free energy of micellization per mole,  $\Delta G_{mic}^0$  (Table 2) at constant temperature and pressure can be calculated as:

$$\Delta G_{mic}^0 = (2-\alpha) RT \ln X_{cmc} \quad (1)$$

Where  $R$  is the gas constant and  $T$  being the absolute temperature and  $X_{\text{cmc}}$  stands for the CMC in terms of mole fraction.  $\Delta G_{\text{m}}^0$  values are negative in all cases which show that process of micellization is spontaneous.  $\Delta G_{\text{m}}^0$  values become more negative with an increase in temperature means process becomes more feasible with an increase in temperature.

The standard Enthalpy of micellization per mole,  $\Delta H_{\text{mic}}^0$  (Table 2) at constant temperature and pressure can be calculated with the help of equation:

$$\Delta H_{\text{mic}}^0 = - (2-\alpha)RT^2(d \ln X_{\text{cmc}})/dT \quad (2)$$

$\Delta H_{\text{mic}}^0$  values are negative in all cases. The standard entropy of micellization, per mole,  $\Delta S_{\text{mic}}^0$  (Table 2) at constant temperature and pressure can be calculated with the help of equation given below using the predetermined values of  $\Delta G_{\text{m}}^0$  and  $\Delta H_{\text{mic}}^0$ .

$$\Delta G_{\text{mic}}^0 = \Delta H_{\text{mic}}^0 - T\Delta S_{\text{mic}}^0. \quad (3)$$

Entropy of micellization values are positive in all cases which shows that entropy favors the process of micellization, which indicates that more structured water molecules around the hydrophobic tails of surfactant are set free when latter are involved in micelle formation and are no longer in contact with bulk water [28]. This process is termed as the release of hydrophobic hydration of ionic surfactant monomers. The aqueous region around the non polar tails is also known as ‘‘icebergs’’ or flickering clusters. Additionally, the effect on the head groups as a result of transition from monomeric form to micellar form is also accountable and is accompanied with positive entropy change. This favours the non-random enhancement of the packing of hydrocarbon chains within the micellar core. Smaller values of  $\Delta S_{\text{mic}}^0$  in the presence of different alcohol concentrations as compared to those in water and also a decrease with increasing concentration support relatively less contribution of above mentioned two factors to  $\Delta S_{\text{mic}}^0$ .

Considering the effect of temperature on the various thermodynamic parameters of micellization, it may be noted that although CMC increases with temperature but  $\Delta G_{\text{mic}}^0$  values become more negative indicating facilitated micellization. This may be due to the reason that both the amount of water structured by hydrophobic chains and amount of water bound by hydrophilic groups decrease with increase in temperature, this results in a

decrease in  $\Delta S^0_{mic}$  and  $\Delta H^0_{mic}$ , respectively. As is clear from the equation, these two parameters contribute to  $\Delta G^0_{mic}$  in opposite manner, so increase or decrease of  $\Delta G^0_{mic}$  values with temperature depends upon the predominance of one of these two factors.

**TABLE 1: Specific conductance of SDBS in water at temperatures 298.15, 308.15 and 318.15 K.**

Concentration of SDBS (M)	Specific conductance (mS) 298.15 K	Specific conductance (mS) 308.15K	Specific Conductance (mS) 318.15 K
0	.0010	.0008	.0007
.0001	.0035	.0043	.0049
.0003	.0061	.0078	.0088
.0004	.0088	.0112	.0118
.0006	.0114	.0145	.0163
.0007	.0139	.018	.020
.0009	.0163	.021	.023
.0011	.019	.024	.027
.0012	.021	.027	.030
.0013	.023	.030	.034
.0014	.025	.033	.038
.0016	.027	.036	.041
.0017	.029	.039	.045
.0018	.032	.041	.048
.0020	.034	.044	.051
.0021	.036	.047	.054
.0022	.038	.050	.057
.0024	.041	.052	.060
.0025	.043	.054	.063
.0026	.044	.056	.066
.0027	.046	.058	.069
.0029	.047	.060	.070

.0030	.049	.062	.073
.0031	.050	.064	.075
.0032	.052	.066	.077
.0033	.053	.068	.079
.0035	.055	.070	.081
.0036	.056	.072	.083
.0037	.057	.074	.086
.0038	.059	.076	.088
.0039	.060	.077	.089
.0040	.061	.079	.090
.0041	.062	.081	.092
.0042	.064	.082	.094
.0044	.065	.084	.096
.0045	.066	.086	.098
.0046	.067	.087	.100
.0047	.068	.090	.102
.0048	.069	.092	.104
.0049	.071	.093	.106
.0050	.072	.095	.107

**Table 2: Values of CMC,  $X_{CMC}$ ,  $\alpha$ ,  $\Delta G^0_{mic}$ ,  $\Delta H^0_{mic}$  and  $\Delta S^0_{mic}$  of SDBS in water and various concentrations of ethanol, 2-Ethoxy ethanol and 2-Methoxy ethanol.**

Temp.(K)	CMC(M)	$X_{cmc}$	$\alpha$	$\Delta G^0_{mic}$ (kJ/mol)	$\Delta H^0_{mic}$ (kJ/mol)	$\Delta S^0_{mic}$ (KJ/K/mol)
<b>SDBS In Water</b>						
298.15	.0026	.00004	.4909	-37.30	-6.803	.1022



308.15	.0028	.00005	.883	-28.22	-5.379	.0741
318.15	.0029	.00006	.662	-34.85	-6.868	.0879
<b>SDBS In Ethanol (0.5%)</b>						
298.15	.0024	.00004	.779	-30.35	-6.136	.0812
308.15	.0027	.00005	.914	-27.65	-5.830	.0708
318.15	.0028	.00005	.883	-29.23	-6.391	.0717
<b>SDBS In Ethanol (1%)</b>						
298.15	.0023	.00004	.662	-33.42	-8.306	.0842
308.15	.0025	.00005	.883	-28.60	-7.407	.0687
318.15	.0027	.00005	.883	-29.27	-7.895	.0672
<b>SDBS In Ethanol (2%)</b>						
298.15	.0021	.00004	.602	-35.15	-6.612	.0957
308.15	.0023	.00004	.883	-28.84	-5.643	.0752
318.15	.0024	.00004	.662	-35.44	-7.206	.0887
<b>SDBS In Ethanol (5%)</b>						
298.15	.0021	.00004	.685	-33.11	-5.928	.0911
308.15	.0022	.00004	.773	-31.74	-5.908	.0838
318.15	.0024	.00004	.662	-35.51	-6.868	.0900
<b>SDBS In 2-Ethoxy ethanol (0.5%)</b>						
298.15	.0024	.00004	.8115	-29.56	-5.094	.0820

308.15	.0025	.00005	.662	-34.23	-6.126	.0912
318.15	.0027	.00005	.794	-31.64	-5.886	.0809
<b>SDBS In 2- Ethoxy ethanol (1%)</b>						
298.15	.0023	.00004	.795	-30.07	-5.343	.0829
308.15	.0024	.00004	.662	-34.40	-6.337	.0910
318.15	.0026	.00005	.964	-27.26	-5.230	.0692
<b>SDBS In 2- Ethoxy ethanol (2%)</b>						
298.15	.0023	.00004	.736	-31.61	-3.923	.0928
308.15	.0024	.00004	.707	-33.27	-4.287	.0940
318.15	.0025	.00005	.745	-33.22	-4.435	.0904
<b>SDBS In 2- Ethoxy ethanol (5%)</b>						
298.15	.0021	.00004	.927	-27.06	-5.233	.0732
308.15	.0022	.00004	.713	-33.33	-6.705	.0864
318.15	.0024	.00004	.745	-33.34	-6.957	.0829
<b>SDBS In 2- Methoxy ethanol (0.5%)</b>						
298.15	.0024	.00004	.795	-36.15	-3.384	.1099
308.15	.0024	.00004	.707	-33.17	-3.878	.0950
318.15	.0026	.00005	.736	-33.33	-4.042	.0920
<b>SDBS In 2- Methoxy ethanol (1%)</b>						
298.15	.0022	.00004	.662	-33.49	-3.461	.1007

308.15	.0023	.00004	.713	-33.17	-3.554	.0961
318.15	.0024	.00004	.745	-27.98	-3.107	.0782
<b>SDBS In 2- Methoxy ethanol (2%)</b>						
298.15	.0022	.00004	.883	-28.05	-3.384	.0827
308.15	.0022	.00004	.815	-30.61	-3.835	.0868
318.15	.0024	.00004	.745	-33.36	-4.330	.0912
<b>SDBS In 2- Methoxy ethanol (5%)</b>						
298.15	.0021	.00004	.924	-27.16	-4.453	.0761
308.15	.0022	.00004	.906	-28.34	-4.836	.0762
318.15	.0023	.00004	.995	-26.77	-4.736	.0692

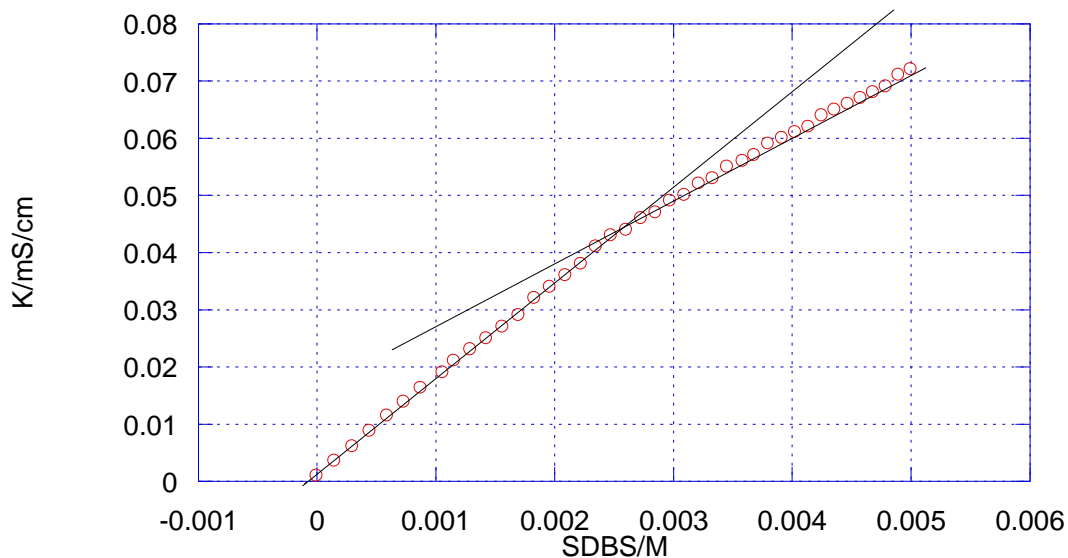


Fig 1. Specific conductance vs. concentration plot of SDBS in water at 298.15 K.

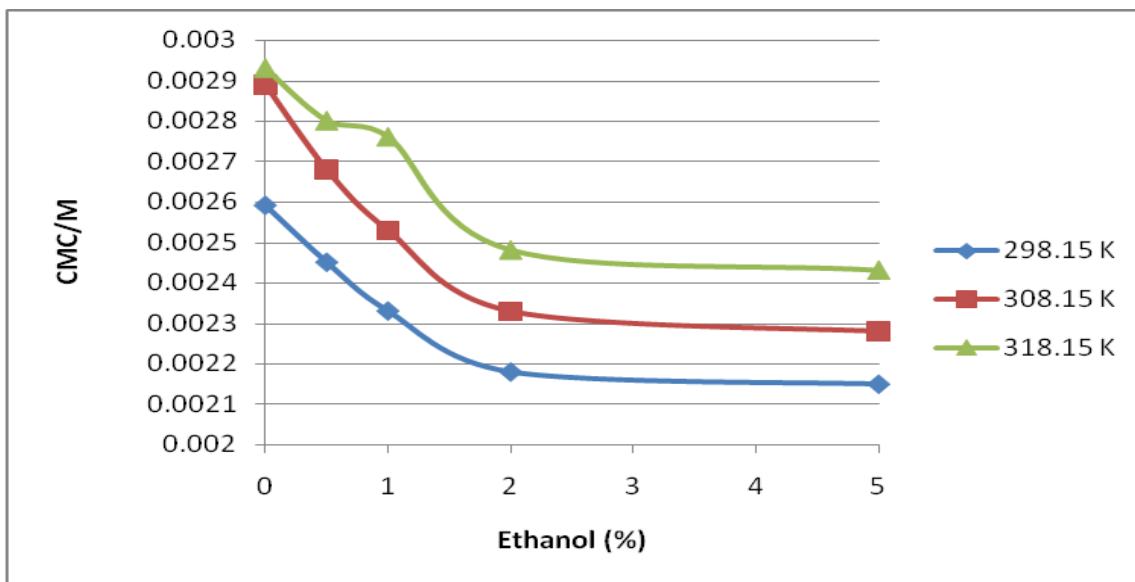


Fig 2. Plots of CMC values of SDBS vs. ethanol concentration at  $\diamond$  298.15,  $\square$  308.15,  $\Delta$  318.15 K.

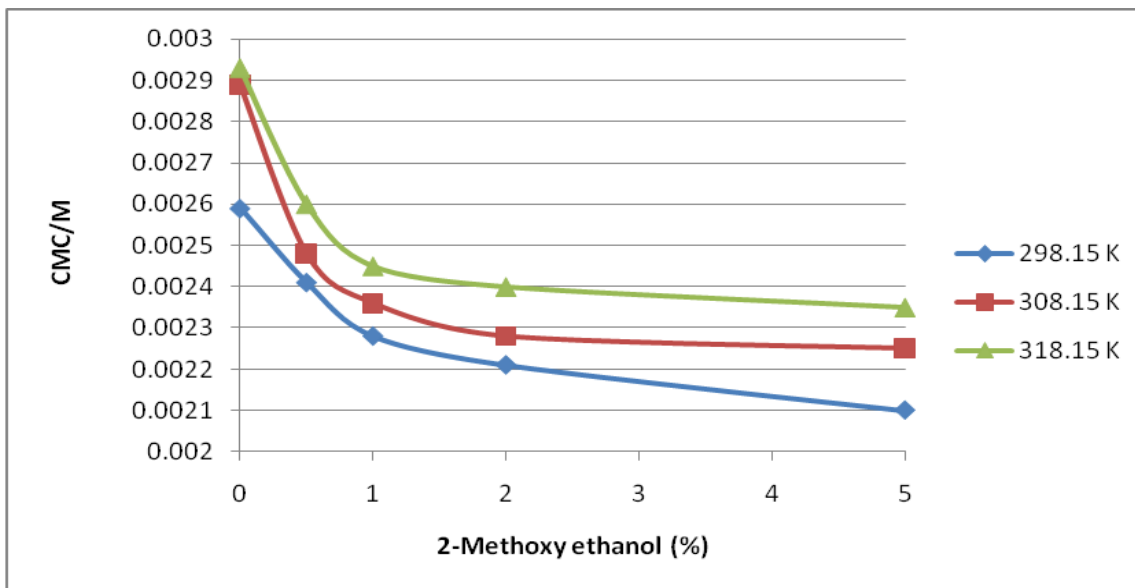


Fig 3. Plots of CMC values of SDBS vs. 2-methoxy ethanol concentration at  $\diamond$  298.15,  $\square$  308.15,  $\Delta$  318.15 K.

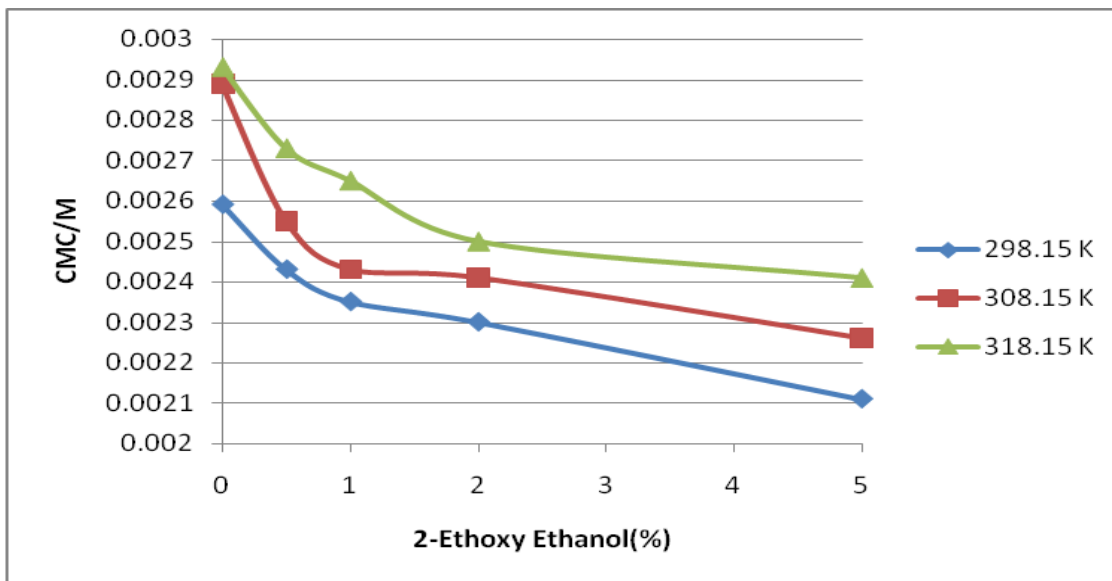


Fig 4. Plots of CMC values of SDBS vs. 2-ethoxy ethanol concentration at  $\diamond$  298.15,  $\square$  308.15,  $\Delta$  318.15 K.

**Conclusion:**

The study of surfactants is of immense importance as they are often used in colloidal chemistry because of their amphiphilic nature. They stabilize naturally occurring dispersions of different types such as emulsions, suspensions, and foams etc. which find wide applications in different fields. Therefore, an understanding of surfactant systems is important from both theoretical and practical point of view.

The electrical conductance of SDBS in aqueous solutions of ethanol, 2-ethoxy ethanol and 2-methoxy ethanol having concentration 0.5, 1, 2 and 5% (v/v) have been determined at 298.15, 308.15 and 318.15K. Specific conductivity,  $\kappa$  is linearly correlated to the surfactant concentration in both pre-micellar and post-micellar regions. These data have been used to calculate the thermodynamic parameters such as  $\Delta G_{mic}^0$ ,  $\Delta H_{mic}^0$ ,  $\Delta S_{mic}^0$ . The results have been attributed to the stability of micelles with the increasing amount of each additive upto a certain concentration of solvent. A complete profile of the effects of these additives on the thermodynamic, micellar as well as surface properties of surfactant is required for such applications. In the present report, from the variation of CMC values for SDBS in the presence of studied alcohols, it is clear that micellization of SDBS is facilitated by alcohols at a constant temperature. Temperature has a negative influence on the micellization tendency of SDBS in water as well as in the presence of additives. Enthalpy of micellization  $\Delta H_{mic}^0$  is found to be negative and entropy of micellization  $\Delta S_{mic}^0$  is positive in all cases, the major contribution of positive entropy to the negative value of  $\Delta G_{mic}^0$  indicates that hydrophobic interactions play a major role in the micellization process.  $\Delta G_{mic}^0$  values are negative in all cases which shows that process of micellization is spontaneous.

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