

Effect of Streptomycin Drug on the Surface and Micellization Properties of Sodium Dodecylbenzyl Sulphate Surfactant

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Abstract: The effect of Streptomycin (STMY) drug on the surface and micellar properties of Sodium Dodecylbenzyl Sulphate Surfactant (SDBS) have been investigated by surface tension and conductivity measurements at the temperature range 293-323 K. From these measurements, the Critical Micelle Concentration (CMC) T_{max} (maximum surface excess) A_{min} (minimum surface area per molecule) and γ_{cmc} (surface tension at the CMC) have been determined. Thermodynamic parameters (ΔG°_m , ΔH°_m , ΔS°_m) of the micelle formation were calculated from the temperature dependence on the CMC. The standard Gibbs free energies of adsorption (ΔG°_{ads}) were also, evaluated and the results show that all the ΔG°_{ads} values are negative and their magnitudes reveal that micelle formation is less spontaneous than adsorption.

Key words: Critical micelle concentration, conductivity, surface tension, sodium dodecylbenzyl sulphate, streptomycin, adsorption

INTRODUCTION

Biological membranes are extremely complex system, so that, models of less complexity have been required to investigate different bilayer properties. One of these models is micelles which provide an attractive model system for biomembranes because of their relative simplicity, low toxicity, bioavailability and stability of the drug through micelle incorporation (Corrigan and Healy, 2002; Vermathen *et al.*, 2000; Rangel-Yagui *et al.*, 2005; Torchilin, 2001). Interaction of various drugs with the micellar phase and encapsulation of molecules inside micelle are studied as models to see the nature of interaction (Caetano and Tabak, 2000; Erdinc *et al.*, 2004; Rangel-Yagui *et al.*, 2005). Surfactants are also used as excipients and usually added to the formulation to facilitate the preparation, patient acceptability and functioning of the dosage form (Strickley, 2004). Surfactant micelles have been widely utilized as an approach to increase the water solubility of many pharmaceutical substances that represents a formidable problem in formulation of an acceptable dosage form (Sun *et al.*, 2003).

The interaction of chlorpromazine and trifluoperazine drugs with ionic micelles has been monitored by using electronic absorption and fluorescence spectroscopy (Caetano and Tabak, 1999). The apparent pKa changes of the drugs induced by the micelles were observed to be consistent with a strong interaction. The special characteristics of the drugs binding to anionic Sodium

Dodecyl Sulfate (SDS) monomers and or micellar aggregates were focused. The binding constants showed a significant hydrophobic contribution modulated by electrostatic interactions of the cationic drug with the micelle head group. Small angle X-ray scattering studies have been reported on the interaction of chlorpromazine and SDS micelles by Caetano *et al.* (2002). The influence of trifluoperazine dihydrochloride on the micellar properties of SDS and zwitterionic surfactant 3-(N-hexadecyl-N, N-dimethylammonium) propane sulfonate have been investigated (Caetano *et al.*, 2003). The properties on the surface and in the bulk of the solution of trifluoperazine and fluphenazine dihydrochloride have also been studied at different pH (Cheema *et al.*, 2007).

The interaction of Ciprofloxacin hydrochloride (Cpf) as an antibiotic in aqueous solution with SDS as an anionic surfactant was checked using steady state fluorescence spectroscopy. Binding was shown by Cpf and SDS in the pre-micellar region of SDS. The drug becomes free from the SDS monomers in the micellar and post-micellar regions of SDS due to the stronger organizational forces of micelle formation by the monomers of SDS (Khan and Shah, 2009). The interactions of promethazine and trifluoperazine hydrochloride with anionic surfactant sodium dodecyl sulfate in the absence and presence of various concentrations of cosolvents have been studied by absorption spectroscopy as a function of surfactant concentration ranging from the pre-micellar to post-micellar

region at 298 K. The addition of cosolvents increased the CMC of sodium dodecyl sulfate and at a certain concentration totally inhibited the micellization (Gokturk and Var, 2012).

Binding and distribution properties of Trimethoprim (TMP) in the presence of various anionic surfactants; Sodium octyl sulfate (C_8SO_4Na) sodium decyl sulfate ($C_{10}SO_4Na$) sodium lauryl sulfate ($C_{12}SO_4Na$) and sodium tetradecyl sulfate ($C_{14}SO_4Na$) has been studied by conductivity, spectrophotometry and surface tension measurements. The surface properties of anionic surfactants, maximum surface excess concentration (Γ_{max}) and minimum area per surfactant molecule (A_{min}) at the air/water interface have been evaluated in the absence and presence of TMP using Gibbs adsorption isotherm (Gokturk and Aslan, 2014).

Micellar solubilization of drug furocemide at concentrations 0.001 and 0.01 M in surfactants; Sodium Dodecyle Sulfate (SDS) and Cetyltrimethylammonium Bromide (CTAB) has been carried out by measuring different transport properties. Conductance measurements of anionic surfactant SDS and cationic surfactant CTAB have been measured in solutions of aqueous Furosemide at above said concentrations in the temperature range 293-313 K. From these measurements, CMC 's of SDS and CTAB have been determined in these solutions. From CMC data, various thermodynamic parameters such as ΔG°_m , ΔH°_m and ΔS°_m have been evaluated (Kaushal *et al.*, 2015). The interaction of amphiphilic antidepressant drug Nortriptyline hydrochloride (NOT) with nonionic surfactant Triton X-100 (TX-100) was studied using tensiometry in aqueous and in urea solutions at 303 K. Various models such as Clint, Rubingh and Rosen were employed to get the information regarding the nature of interaction between the compounds in bulk and at the interface. The CMC values of the mixed systems of NOT and TX-100 were found to be in between the CMC value of pure components which suggest nonideal mixed system having attractive interactions in the absence and presence of urea (Rub *et al.*, 2016).

In this study, the interaction of streptomycin with anionic surfactant SDBS was studied using surface tension and conductivity measurements. Γ_{max} was calculated from Gibb 's equation while A_{min} was computed from surface excess. Thermodynamic parameters ΔG°_m , ΔH°_m and ΔS°_m of the micelle formation were calculated from the temperature dependence on the CMC.

MATERIALS AND METHODS

Experimental

Material and experimental: The anionic surfactant, SDBS was BDH product has 80% active constituent; the

remainder being sodium sulphate; free from commercial detergent additive and a Streptomycin drug (STMY) was product of Merck with purity 98%. All solutions were prepared in Deionized water (sp. conductivity = 2×10^{-6} S cm^{-1}).

Equipment and measurements: The surface tension (γ) values were measured using Du Nouys platinum ring method using tensiometer Model DST 30 M, Surface and Electro Optics (SEO) Company-Korea. For each set of experiments, the ring was cleaned by immersed in 5M HCl solution. Each measurement was repeated 3 times to ensure the reproducibility of the results. The mixed solutions were prepared by mixing 2 pure solutions and were kept for at least 30 min for equilibration before measuring the surface tension. Conductivity measurements were carried out using a high precision Conductivity Meter (WTW-Germany). The conductivity meter was calibrated with a KCl standard solution of known conductivity.

RESULTS AND DISCUSSION

Surface properties: The surface tension (γ) and conductivity (κ) of SDBS surfactant solutions in water and in streptomycin solutions (0.0001 and 0.00001M) were measured as a function of SDBS concentration at 293, 303, 313 and 323 K and the CMC for the surfactant was then considered as the point of intersection between two continuous lines obtained by plotting (γ) vs. (surf) and κ vs. (surf) as shown in Fig. 1 and 2. The surface excess concentration, Γ_{max} was calculated from Gibb's Eq. 1:

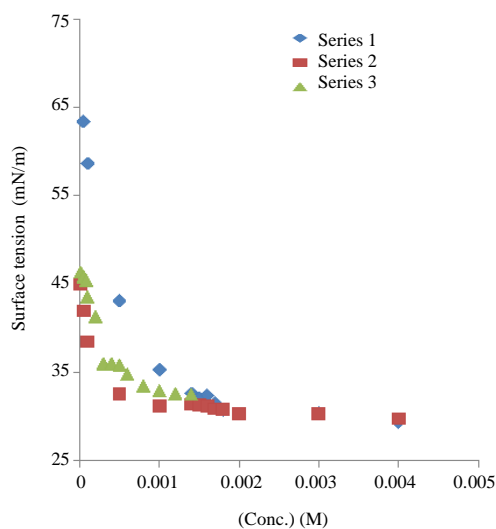


Fig. 1: Surface tensions (γ) (versus) (C) for SDBS surfactant and SDBS+STPY drug systems

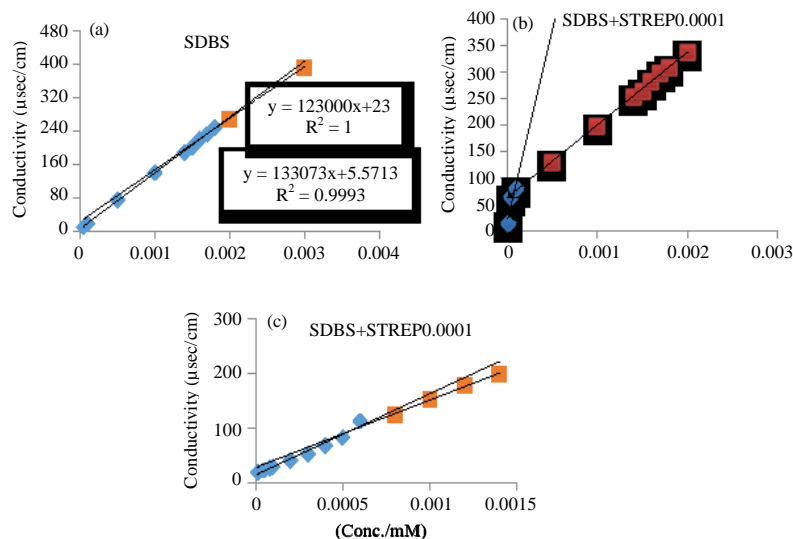


Fig. 2: Conductivity (γ) versus (C) for SDBS surfactant and SDBS+STPY drug systems

Table 1: Interfacial and thermodynamic parameters for SDBS surfactant and SDBS+STPY drug System

| T/K | CMC/mM tension | CMC/mM conduc | Π_{CMC} mN m ⁻¹ | $\Gamma_{MAX} \times 10^6$ (mol/m ²) | A_{min} Å ² /molecule | $-\Delta G_{ad}^{\circ}$ (kJ/mol) | $-\Delta G_m^{\circ}$ (kJ/mol) | $-\Delta H_m^{\circ}$ (kJ/mol) | ΔS_m° (J/mol K) |
|-----|----------------|---------------|--------------------------------|--|------------------------------------|-----------------------------------|--------------------------------|--------------------------------|--------------------------------|
| 293 | 1.85 | 1.60 | 41.481 | 1.914 | 86.750 | 49.478 | 28.076 | 1.3820 | 91.100 |
| 303 | 1.90 | 1.70 | 40.481 | 1.174 | 141.400 | 57.455 | 22.974 | 1.2180 | 71.861 |
| 313 | 1.95 | 1.90 | 39.981 | 1.208 | 137.400 | 60.959 | 27.863 | 1.5306 | 84.130 |
| 323 | 1.97 | 2.00 | 39.481 | 1.089 | 147.600 | 49.425 | 28.532 | 1.6190 | 83.321 |
| 293 | 0.09 | 0.08 | 38.481 | 0.543 | 305.900 | 126.360 | 55.454 | 11.8780 | -107.300 |
| 303 | 0.10 | 0.09 | 40.481 | 0.511 | 324.839 | 139.265 | 60.077 | 12.6610 | -160.456 |
| 313 | 0.11 | 0.10 | 39.381 | 0.399 | 415.769 | 160.756 | 62.156 | 13.6300 | -110.539 |
| 323 | 0.12 | 0.12 | 40.481 | 0.435 | 382.005 | 156.735 | 63.612 | 14.4910 | -108.996 |
| 293 | 0.33 | 0.58 | 35.481 | 0.328 | 506.582 | 142.531 | 34.292 | 8.1830 | 89.109 |
| 303 | 0.23 | 0.55 | 36.481 | 0.429 | 386.700 | 122.171 | 37.213 | 8.9160 | 93.389 |
| 313 | 0.25 | 0.53 | 39.481 | 0.440 | 377.061 | 128.967 | 38.184 | 10.1320 | 89.623 |
| 323 | 0.28 | 0.55 | 41.281 | 0.302 | 549.600 | 175.459 | 38.813 | 10.0730 | 88.981 |

$$\Gamma_{max} = -1/nRT [d\gamma/d \ln C] \quad (1)$$

The minimum area occupied by surfactant molecule, A_{min} was computed from surface excess using Eq. 2:

$$A_{min} = 1/N \Gamma_{max} \quad (2)$$

Where:

- C = The molar concentration of the surfactant in solution
- n = The number of species constituting surfactant
- N = Avogadro's number

The $d\gamma/d \ln C$ factor was obtained from the slopes of the linear plots of γ vs. $\ln C$ (not shown) (Table 1). The results obtained for CMC are 1.85, 1.90, 1.95 and 1.97 mM at temperatures 293, 303, 313 and 323 K, respectively. It should be noted that the present results are in the range of the results previously reported (Zhao *et al.*, 2013; Azuma *et al.*, 2014). The cmc for SDBS-STMY

system was decreased extensively to 0.33 and 0.09 mM at 10⁻⁵ and 10⁻⁴ M of STMY, respectively. The decrease in CMC values indicates that the micellization of SDBS becomes highly cooperative in the presence of STMY drug (Kaushal *et al.*, 2015).

The values of Γ_{max} decrease while the A_{min} value obtained increase when the STMY was added in the 2 concentrations studied which indicates that when STMY drug was added the mixture have a greater tendency to be adsorbed at the air/water interface compared to a pure SDBS surfactant solution. The presence of STMY may be increases the repulsion among head groups and low surfactant mixture molecules can be adsorbed at the interface (Khan *et al.*, 2010; Ali, 2017).

Thermodynamic of micelle formation: Thermodynamic parameters of micellization (ΔG_m° , ΔH_m° , ΔS_m°) were calculated from the temperature dependence of the CMC from the following Eq. 3-5:

$$\Delta G_m^\circ = RT \ln X_{CMC} \quad (3)$$

$$\Delta H_m^\circ = -RT^2 (\partial \ln X_{CMC} / \partial T) \quad (4)$$

$$\Delta G_m^\circ = \Delta H_m^\circ - T \Delta S_m^\circ \quad (5)$$

Where:

X_{CMC} = The Critical Micelle Concentration of surfactant in mole fraction unit

R = Gas constant

T = The absolute temperature

$\partial \ln X_{CMC} / \partial T$ = Evaluated from the slope of the plot of $\ln X_{CMC}$ versus temperature

ΔG_{ad}° = The air/water interface is calculated from the relation

$$\Delta G_{ad}^\circ = \Delta G_m^\circ - (\Pi_{CMC} / \Gamma_{max}) \quad (6)$$

Π_{CMC} is calculated from the Equation; $\Pi_{CMC} = \gamma_0 - \gamma_{CMC}$. Where, γ_0 is the surface tension of pure solvent and γ_{CMC} is the surface tension at the CMC.

The results of the thermodynamic parameters obtained from the Eq. 3-6 are listed in Table 1. The results of this table show that ΔG_m° is negative for the 3 systems in the temperature range studied which indicates that the micellization processes are spontaneous. The values of ΔH_m° are negative and increased as temperature increased which indicates the micellization is exothermic. The entropy of micellization, ΔS_m° for SDBS and SDBS+10⁻⁵ M STMY are positive in all temperature range. This is due to the fact that the head group is more hydrated than the hydrophobic tail which leads to an ordering of the system (Owoyomi *et al.*, 2011). The values of ΔG_{ad}° are all negative and more than ΔG_m° (in magnitude). This shows that surfactant first try to adsorb at the interface and then form micelles. The values of pure component are a smaller amount negative than those of the mixtures, showing that the mixed systems are more active than those of the pure components.

CONCLUSION

From surface tension and conductivity measurements it can be inferred that the presence of streptomycin drug as well its concentration affect the surface and micellization properties of SDBS surfactant.

Value of Γ_{max} decreases and A_{min} value increases when STMY was added, this indicates the mixtures SDBS-STMY have a greater tendency to be adsorbed at the air/water interface, compared to a pure SDBS surfactant solution.

Thermodynamic adsorption data showed that the adsorption and micelle formation of the surfactant and their mixtures with STMY occurs spontaneously and

ΔG_{ads}° magnitudes for the three systems reveal that micelle formation is less spontaneous than adsorption.

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