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## Interfacial And Thermodynamic Properties Of Sodium Dodecyl Benzyl Sulphate Surfactant-Furosemide Drug System.

Mena M Faris, and Sameer H Kareem\*.

Department of Chemistry- College of Science for Women- University of Baghdad.

### ABSTRACT

Interfacial and thermodynamic properties  $\Gamma_{\max}$ ,  $A_{\min}$ ,  $\Pi_{cmc}$ ,  $\Delta G_m^\circ$ ,  $\Delta H_m^\circ$ , and  $\Delta S_m^\circ$  of individual sodium dodecylbenzylsulphate (SDBS) surfactant and their mixture with furosemide (FUMD) drug at two concentrations (0.0001 and 0.00001 M) have been calculated. The calculation was performed using the surface tension and conductivity measurements with the variation of surfactant concentration to determine the cmc of all systems studied. The variation of critical micelle concentration (cmc) with concentration of furosemide drug and the temperature was used to calculate the parameters above. The results indicate that the cmc of SDBS decrease when the FUMD was added but when the temperature increased the cmc decreased at the whole temperature studied. The results obtained for interfacial properties show that the FUMD drug has no effect on the surface activity of SDBS but  $\Gamma_{\max}$  small increases with the addition of FUMD and decreases with temperature. The results of thermodynamic properties indicate that the micellization and adsorption at interface are spontaneous and the  $\Delta G_{ads}^\circ$  are more negative than  $\Delta G_m^\circ$  at all temperatures which reveal that micelle formation is less spontaneous than adsorption.

**Keywords:** sodium dodecylbenzylsulphate, furosemide, critical micelle concentration, interfacial properties

\*Corresponding author

## INTRODUCTION

Surfactants are widely used in pharmaceutical products to increase the solubility of medicinal materials, to reduce the interfacial tension between the medium and the drug, and to increase the rate of percutaneous absorption. Surfactants are also used in dental, oral, sublingual (under the tongue) to form ointments, creams, gels, patches, tapes, and liquids [1]. The study of drug-surfactant interactions [2-5] has received an increased attention in the last period of time because of widespread application of surfactants in the pharmaceutical field especially the interaction of Furosemide with drugs.

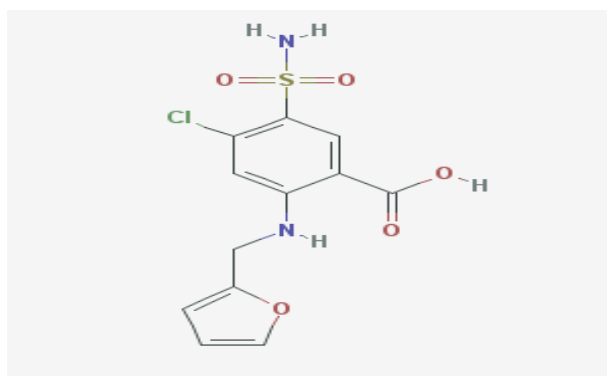
The interaction of Furosemide in aqueous solutions with the anionic surfactant SDS and the cationic surfactant CTAB have been carried out in the temperature range 20–40 °C using sound velocity and density measurements [6]. The cmc of surfactants in (0.002 and 0.02 mol dm<sup>-3</sup>) furosemide solutions have been determined and the dependence of cmc on temperature was used to calculate various thermodynamic parameters such as the standard enthalpy change ( $\Delta H_m^\circ$ ), standard entropy change ( $\Delta S_m^\circ$ ), and standard Gibbs energy change ( $\Delta G_m^\circ$ ) for micellization. Micellization behaviour of SDS in the presence of Furosemide at two concentrations 0.001 and 0.002 M in water – dimethylsulphoxide solution using conductance, densities, velocities of sound, and viscosities studies in the temperature range 20–40 °C [7]. The cmc of SDS was determined from the plots of specific conductance of SDS. The results showed that cmc values of surfactant decrease with increase in concentration of Furosemide and increase with the rise in temperature. By using cmc data, various thermodynamic parameters like ( $\Delta H_m^\circ$ ), ( $\Delta S_m^\circ$ ) and ( $\Delta G_m^\circ$ ), that have direct bearing on the consequences of such interactions at the molecular level have been calculated.

The effect of furosemide at two concentrations 0.01 and 0.001 M on micellization behavior of two ionic surfactants sodium dodecyl sulphate (SDS) and cetyltrimethylammonium bromide (CTAB) has been studied using conductance measurements in the temperature range 293 to 313 K [8]. Critical micelle concentration cmc of SDS and CTAB have been determined in these solutions and the thermodynamic parameters  $\Delta G_m^\circ$ ,  $\Delta H_m^\circ$ , and  $\Delta S_m^\circ$  was calculated. The results indicate that the process is spontaneous and exothermic in nature.

In the present work the interaction of Furosemide drug with SDBS surfactant was studied using surface tension and conductivity measurements in the temperature range 293 to 313 K. The cmc of surfactant was determined from the plots of surface tension or conductivity versus concentration and used to calculate various parameters of interfacial and thermodynamic properties.

## EXPERIMENTAL

The anionic surfactant, SDBS, was purchased from BDH and has 80% active constituent; the remainder being sodium sulphate; free from commercial detergent additive. The drug furosemide (4-chloro-N-(2-furylmethyl)-5-sulphamoyl-anthracitic acid) is supplied by Sanofi in injectable form, having a composition of 20 mg in 2 ml distilled water. All solutions were prepared in Deionized water (sp. conductivity =  $2 \times 10^{-6}$  S cm<sup>-1</sup>). The drug furosemide has the structure shown in structure 1.



Structure 1: The structure of Furosemide

The surface tension ( $\gamma$ ) values were measured by platinum ring Du Nouy method using tensiometer model DST 30 M, Surface and Electro Optics (SEO) Company- Korea. The ring was cleaned by immersed in 5M HCl solution after a each set of experiments. Each measurement was repeated three times to ensure the reproducibility of the results. Conductivity measurements were carried out using a precision conductivity meter (WTW – Germany). The conductivity meter was calibrated with a KCl standard solution of known conductivity.

### RESULTS AND DISCUSSION

Figures 1 and 2 show the variation of surface tension ( $\gamma$ ) and conductivity ( $\kappa$ ) of SDBS surfactant solutions in water and in furosemide solutions (0.0001 and 0.00001M) as a function of SDBS concentration at 293K. The CMC for the surfactant was then considered as the point of intersection between two continuous lines shown in Figures 1 and 2.

The cmc values obtained for SDBS/FUMD ( $10^{-5}$ M) system at 293 K are 0.0010 and 0.00092 M, while for SDBS/FUMD ( $10^{-4}$ M) system are 0.00074 and 0.00076 M using surface tension and conductivity measurements respectively. These values and the values of cmc obtained at the temperatures 303, 313 and 323 K are listed in Table 1. The results of Table 1 show that when FUMD drug was added to the SDBS solution, the cmc values decreased and the magnitude more decrease as the concentration of FUMD increased from  $10^{-5}$  to  $10^{-4}$  M. This means that the presence of FUMD facilitates the aggregation of SDBS surfactant at lower surfactant concentration due to the complex formation of drug-surfactant which cause the cmc values decrease [9,10]. The values of cmc of SDBS/ FUMD mixtures increased with temperature increased as expected, which indicate that when the temperature increased the surfactant solubility increased and increased repulsions between surfactant-charged head [11,12].

The surface properties, surface excess concentration ( $\Gamma_{max}$ ) was calculated from Gibb's equation:

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

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

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

Where C is the molar concentration of the surfactant in solution, n is the number of species constituting surfactant, and N is Avogadro's number. The  $d\gamma/d\ln C$  factor was obtained from the slopes of the linear plots of  $\gamma$  vs.  $\ln C$  (not shown).

$\Pi_{cmc}$  is calculated from the equation;  $\Pi_{cmc} = \gamma_o - \gamma_{cmc}$ . Where  $\gamma_o$  is the surface tension of pure solvent and  $\gamma_{cmc}$  is the surface tension at the cmc.

$\Delta G_{ad}^\circ$  at the air/water interface is calculated from the relation:

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

Thermodynamic parameters of micellization ( $\Delta G_m^\circ$ ,  $\Delta H_m^\circ$ , and  $\Delta S_m^\circ$ ) were calculated from the temperature dependence of the cmc from the following equations:

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

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

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

Where  $X_{cmc}$  is the critical micelle concentration of surfactant in mole fraction unit, R is gas constant, and T is the absolute temperature.  $(\partial \ln X_{cmc} / \partial T)$  was evaluated from the slope of the plot of  $\ln X_{cmc}$  versus temperature.

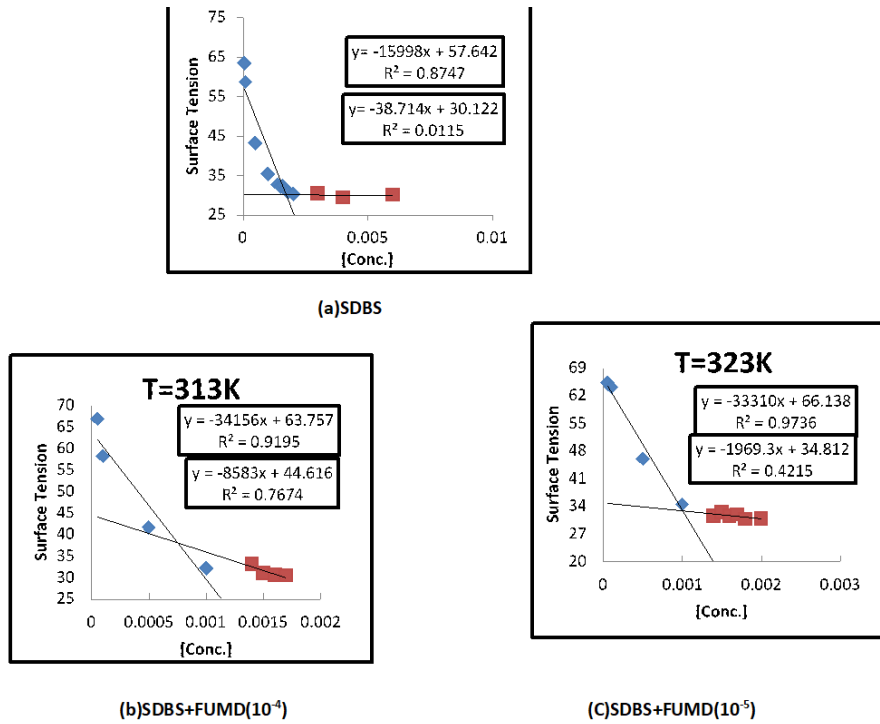


Figure 1. Surface tensions ( $\gamma$ ) versus [C] for SDBS surfactant and SDBS + FUMD drug systems

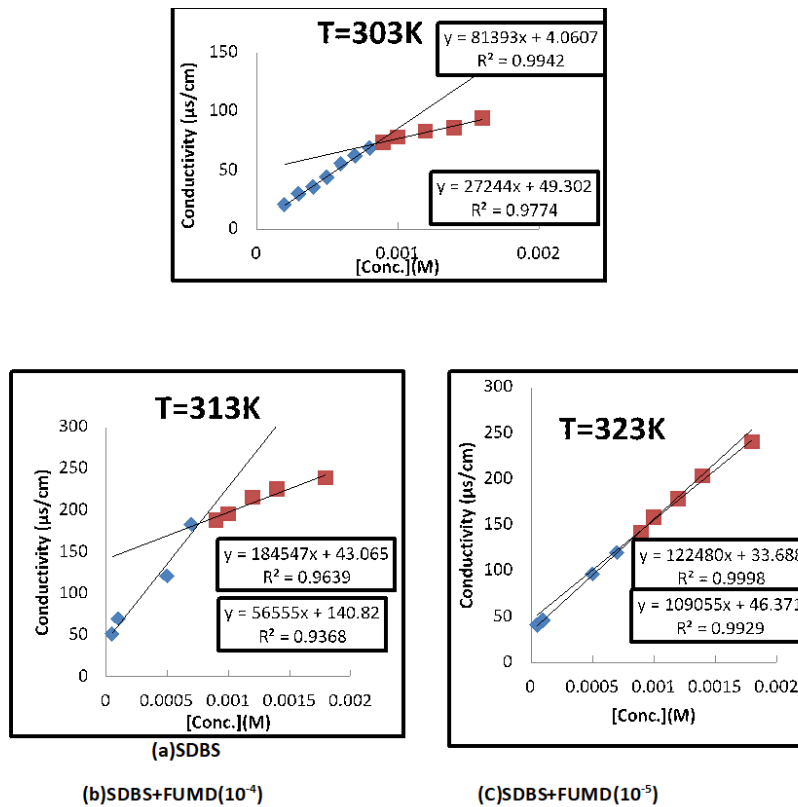


Figure 2. Conductivity ( $\kappa$ ) versus [C] for SDBS surfactant and SDBS + FUMD drug systems

The results of the surface properties and thermodynamic parameters obtained from the equations above are listed in Table 1.

**Table 1. Adsorption at interface and thermodynamic parameters for SDBS surfactant and SDBS + FUMD drug**

	T (K)	cmc/ST (M)	cm/CT (M)	$\Pi_{cmc}$ (mN/m)	$\Gamma_{max} \cdot 10^{-6}$ (mol/m <sup>2</sup> )	$A_{min}$ (Å <sup>2</sup> /molecule)	$S_2/S_1$	$-\Delta G_{ad}^{\circ}$ (kJ/mol)	$-\Delta G_m^{\circ}$ (kJ/mol)	$-\Delta H_m^{\circ}$ (kJ/mol)	$\Delta S_m^{\circ}$ (J/mol.K)
SDBS	293	0.00185	0.0016	41.481	1.914	86.75	0.924	49.478	28.076	1.382	91.1
	303	0.0019	0.0017	40.481	1.174	141.4	1.113	57.455	22.974	1.218	719
	313	0.00195	0.0019	39.981	1.208	137.4	0.956	60.959	27.863	1.5306	84.1
	323	0.00197	0.0020	39.481	1.089	147.6	0.963	49.425	28.532	1.619	83.3
SDBS+FUMD (10 <sup>-4</sup> )	293	0.00089	0.00076	33.481	2.315	71.1	0.306	60.023	45.561	11.849	115
	303	0.00091	0.00085	37.481	0.911	182.2	0.454	84.039	42.910	11.564	103.4
	313	0.00099	0.00088	41.481	1.301	127.6	0.492	73.419	41.536	12.037	97.5
	323	0.00120	0.00093	37.581	1.269	130.8	0.513	72.505	42.891	12.640	93.6
SDBS+FUMD (10 <sup>-5</sup> )	293	0.0010	0.00092	38.481	2.153	77.1	15.325	47.411	29.538	19.093	35.6
	303	0.0017	0.00111	41.481	1.141	145.5	26.286	66.925	30.571	21.430	30.1
	313	0.0020	0.00115	42.481	0.798	207.9	36.307	83.884	30.697	22.633	25.7
	323	0.0021	0.00132	42.481	0.649	255.8	44.672	96.326	30.870	23.600	22.5

From the results of Table 1, one can see that  $\Pi_{cmc}$  values of (SDBS+FUMD)(10<sup>-4</sup>+10<sup>-5</sup>) systems were approximately the same  $\Pi_{cmc}$  values of SDBS system which indicate that the FUMD was no effect on the surface activity of SDBS surfactant.  $\Gamma_{max}$  decreased with temperatures for two systems (SDBS+FUMD) (10<sup>-4</sup>+10<sup>-5</sup>) and  $A_{min}$ , as expected presents an inverse trend with temperatures which indicate that the SDBS/FUMD systems behave as SDBS system behaves to the temperature increasing. This means that the temperature was affected on the packing of molecules of the surfactants [13, 14]. It can be generalized  $\Delta G_m^{\circ}$  and  $\Delta G_{ad}^{\circ}$  are negative in whole temperatures range studied and no effect of temperature which indicates that the micellization process and adsorption at Air-water interface are spontaneous for SDBS+FUMD (10<sup>-4</sup>M) and SDBS+FUMD(10<sup>-5</sup>M) systems. It can also be seen from Table 1 that  $\Delta G_{ads}^{\circ}$  values are more than  $\Delta G_m^{\circ}$  values (with their sign) at all temperatures, which suggest that when micelles are formed, work must be performed to transfer the surfactant molecules from the surface to the micellar state through the solvent [15].  $\Delta H_m^{\circ}$  is negative which indicates that the micellization process is exothermic. The entropy of micellization is positive in all temperature range studied and the values decreased when the temperature increases.

### CONCLUSIONS

From the foregoing results and discussions, the following conclusions can be drawn:

- The cmc of SDBS surfactant decreases in the presence of FUMD drug and more decreases as the concentration of FUMD grows from  $10^{-5}$  to  $10^{-4}$  M. In addition, the cmc of the two systems of SDBS-FUMD increase when the temperature increases from 293 to 323 K.
- The  $\Pi_{cmc}$  values of individual SDBS and SDBS-FUMD systems indicate that FUMD drug has no effect on the surface activity of SDBS.  $\Gamma_{max}$  small increases with the addition of FUMD and decreases with temperature.
- The values of  $\Delta G_m^\ddagger$  and  $\Delta G_{ads}^\ddagger$  estimated for individual surfactant and surfactant- drug systems show that the micelle formation process and adsorption at interface are spontaneous and the  $\Delta G_{ads}^\ddagger$  values are more negative than  $\Delta G_m^\ddagger$  at all temperatures which reveal that micelle formation is less spontaneous than adsorption.

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