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Stabilization of Sodium Dodecyl Sulphate Micelle with Prometizine Hydrochloride at Different Temperature

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ABSTRACT.

Micelles of sodium dodecylsulphate (SDS) were stabilized in the presence of Prometizine hydrochloride (PMZ) (at various concentration and temperature) mixed system to optimize their use as biochemical models and for the stabilization of feeble medicine. Conductometric titration technique was employed in this work. The specific conductivity—surfactant plots were analyzed for the determination of critical micelle concentration (CMC) of the SDS in distilled water at different temperature. The result was analyzed using differential conductivity method and method based on the fit of the experimental raw data to simple non-linear function obtained by direct integration of a Boltzman type sigmoid function. SDS micelles were micellized and stabilized earlier when PMZ concentration rose from 0.1mM to 0.25mM. The conductivity values of SDS solutions increase with increase in concentration of PMZ in water. Addition of drug leads to easy dissociation of surfactant molecule into ions hence increase conductivity and counter-ion binding values. Micellization of SDS in the presence of PMZ revealed that increasing the PMZ content in water caused CMC to decrease to an extent which indicate greater surfactant dissociation.

Keywords: Prometizine Hydrochloride, Micellization, solubilization. Critical micelle concentration

Introduction.

The surfactant-promazine hydrochloride PMZ system has historically generated a great deal of interest in both basic and applied research. The features of additive are expected to be of great relevance in the matter of their interactions with surfactant, particularly in relation to their effect on micellization of surfactant molecule which is largely responsible for the characteristics behaviour of such type of system .A chemical compound known as amphiphile possesses both hydrophilic and hydrophobic qualities. Their interactions with various pharmaceutical types are crucial to the biological, pharmacological, and clinical processes. A lots of insoluble drugs (hydrophobic)will require a carrier for its secure delivery to targeted organs, and the lower the critical micelle concentration- cmc, the better it is for the drug to get to its target, whereas, many membrane systems are encountered when drug make a way to its target site (1).

Distribution of drug in the body is majorly effected by molecular polarity(2). Drugs may be lipophilic or hydrophilic. Lipophilic drugs are non polar and distribute into adipose tissue as well as passing the brain blood barrier (3), whereas, polar drugs hydrophilic and are mainly distributed in lean body tissue. There are reportes that large stable bio molecules can reach their destination easily but small bio molecules (with an upper moleculer weight of approximately 900gmol^{-1}, are not able to reach their target in effective concentrations because of the dissociation in water/ acid prior to reaching the lipophilic target due to their high hydrophilicity.

Introduction of surfactant could anyway curb this challenge.

Drug is a substance which when taken into the body, alters the body function either physically and or physiologically. Drug may be legal (alcohol, caffeine, tobacco) or illegal (cannabis, ecstasy, cocaine, heroin) . Drug must partition to a well-defined and energetically favourable environment, orientation, and conformation in the membrane layer before diffusing into intrabilayer receptor binding cell (4). Some reports has shown that surface active drugs can be mixed with cationic surfactants to form cationic vesicles and micelles, which when incorporated into gels, were useful in slowing release rates of active pharmaceutical ingredient(5)

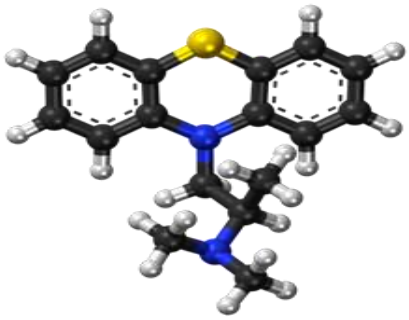
Promethazine drug PMZ is a neuroleptic medication and first generation antihistamine of the phenothiazine family which is used as sedative, to treat migraines, to reduce nervousness, restlessness and agitation caused by psychiatric conditions. Though the side effects could be drowsiness, dizziness, fatigue, constipation etc.

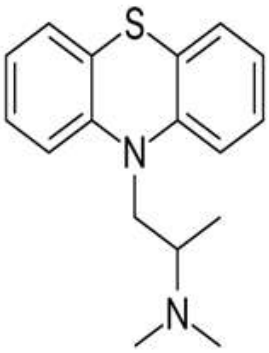
PMZ is a white to faint yellow odourless, crystalline powder.

According to some literatures earlier published, drug modifications includes micellar solubilization, use of co-solvent, addition of different salts, modification of crystal structure, construction of solid dispersion, particle size reduction, use of prodrug along with complex generation(6).

Model Structure of PMZ

Molecular formular $C_{17}H_20N_2S$ Molecular Weight 320.879g/mol





Critical Micelle Concentration

This is the concentration of surfactant above which the micelles form and all additional surfactants added to the system go to micelle. Before reaching the CMC, the surface tension changes strongly with concentration of the surfactant.

When the equivalent conductivity (specific conductance per gram-equivalent of solute) of an anionic surfactant of the type Na^+R^- in water is plotted against the square root of the normality of the solution, the curve obtained, instead of being the smoothly decreasing curve characteristics of ionic electrolytes of this type, has a sharp break in it at low concentration (7) . This break in curve, with its sharp reduction in the conductivity of solution, indicating a sharp increase in the mass unit charge of the material in solution, is interpreted as evidence of the formation of that point of micelles from the unassociated molecules of surfactant, with part of the charge of the micelle neutralised by the associated counterions., the concentration at which the phenomenon occur is called critical micelle concentration-CMC.

Similar breaks in almost every measurable physical property that depends on size or number of particles in solution, including micellar solubilization of solvent-insoluble material and reduction of surface or interfacial tension, are shown by all types of surfactant-nonionic, anionic, cationic and zwitterionic in aqueous media.

Constant value of the activity above the CMC goes into solution in the form of micelles. The determination of the value of the CMC can be use of any of these physical properties, but most commonly the breaks in the electrical conductivity, surface tension, light scattering, or fluorescence spectroscopy concentration curves have been used for this purpose. Critical micelle concentrations have also very frequently been determined from the change in the spectral characteristics of some dyestuff added to the surfactant solution when the CMC of the latter is reached.

Since the properties of solution of surface-active agent change markedly when micelle formation commences, many investigations have been concerned with determining value of CMC in various systems, and a great deal of work have been done on elucidating the various factor that determine the CMC at which micelle formation becomes significant, especially in aqueous media.

CMC value depend on temperature and pressure. When surface coverage by the surfactant increases, the surface free energy (surface tension) decreases and the surface starts aggregating into micelles, thus again decreasing the systems free energy by decreasing the contact area of hydrophobic part of the surfactant with water, any further addition of surfactant will just increase the number of micelles.

CMC values of surfactant decreases with drug's concentration in water. Because the drug (in the salt form is freely dissociated) has an opposite charge from that of surfactant alongside a tricyclic group imparting significant hydrophobic character to the molecule, the effect of such amphiphillic molecules on the micellization behavior of surfactants requires consideration of both the moieties related to the molecule.

Factors affecting the CMC in aqueous media are-Amphiphile chain length, Presence of added electrolyte in the solution, Structure of head group, Temperature of the solution and Presence of a second liquid phase

Surfactants

The surfactant which could be anionic, cationic or amphoteric are amphiphilic substances that reduce the surface tension of water and most other solvents (8). Surfactant as the name implies stand for surface active agent indicating that they preference for surfaces and interfaces.

Surfactant play a vital role in many processes of interest in both fundamental and applied science (9). Extensive studies have been carried out and reported in literature on various mixtures of surfactant systems. Majority of the reports were on anionic-anionic, anionic-cationic, anionic-nonionic, and cationic-nonionic . Physicochemical properties of these mixtures have been mostly investigated at room temperature in aqueous medium.

This work aims to investigate the micelle formation of SDS in the presence of different concentrations of promethazine hydrochloride (PMZ)

Micelle formation

Micelle formation can be envisioned as a stepwise process, characterised by a series of equilibria and equilibrium constants or as a phase separation process such that, once a critical concentration (the critical micellar concentration ,cmc) is reduced, further addition of the surfactant will result in aggregation.

Micelles are formed by self-assembly of amphiphilic molecules. Micelles are formed in aqueous solution whereby the polar region faces the outside surface of the micelle and the nonpolar region forms the core.

The aim of this study was to investigate the effects of concentration of Promethazine hydrochloride (PMZ) and ethanol on the micellar properties of sodium dodecyl sulphate (SDS) at different temperature.

Materials

The anionic surfactant, sodium dodecyl sulphate (SDS) (MW =288g/mol) and promethazine hydrochloride (PMZ) were purchase from Fluka Switzerland and Aldrich (USA) and Sigma chemicals respectively. Sodium dodecyl sulphate are of the highest purity, commercially available, and they are therefore used without any further purification. The water used in this work was double distilled, with specific conductivity of between 1 and 3μScm-1 at room temperature. A digital conductivity meter was used to test the electrical conductivities of surfactants in pure water and aqueous PMZ (Hanna-H15521-02). The conductivity meter was calibrated before use by measuring the electrical conductivities of 0.01 N KCl solution (Merck, purity 99%) to give 1413μS cm-1 at 298.15K and a thermostatic water bath (Grant GD 120) to maintain the temperature within ±0.1K. All weights measurements were carried out using an electronic weighing balance (Mettler Toledo AB54, ±0.0001g.

Methods

Conductometric titration method involves the titration of a known volume of surfactant with a fixed volume of water in the presence of assumed concentration of drug in a thermostated beaker as employed. To observed the effect of drug on the micellization of surfactant, solution of surfactant will be prepared in aqueous solutions of drug having similar concentration. All measurements were within 25 -45 temperature range in a thermostated water bath to maintain a constant temperature. To fully ascertain that the solution mixture (s) have attained the temperature of the bath, the solution-mixture was allowed to equilibrate for at least 30 seconds, after which a thermometer was used to measure the temperature of the solution-mixture. Then after, the meter probe [after calibration] was dipped inside the test tubes and the readings were taken after the value displayed on the meter screen. Electrical conductivity was then recorded.

A plot of specific conductivity against surfactant concentration consists of two linear segments with different positive slopes (pre-CMC slope, and post CMC slope), that intercepted at break points (fig 4.1) which corresponds to the formation of micelles (CMC). The break is as a result of the binding of some of the counter-ion to the micellar surface. The CMC values reflects the degree of binding, an increase in binding causes a decrease in value of the CMC, and the extent of the binding can be obtained as follows:

$$\beta = 1 - \alpha$$

$$\alpha = \frac{S_2}{S_1}$$

where α is the degree of counter-ion dissociation, β is the extent of counter ion binding, S_2, is the post micellar slope, and S_1 is the pre-micellar slope (10). The conductivity measurement has been reported (11) to be one of the straight forward techniques for the determination of the critical micelle concentration (CMC) of the ionic surfactants and other micellar parameters such as degree of micellar ionization. This is due to high sensitivity and reproducibility. This methods has also been reported to be a better diagnostic tool for the measurement of CMC of ionic surfactant (12)

Samples preparation

Preparation of stock solution of promethazine hydrochloride (PMZ)

A stock solution of PMZ (10 mM) was prepared by carefully weighing 0.320g of PMZ in some quantity of distilled water in 100 ml standard volumetric flask.

Preparation of 0.10 mM solution of promethazine hydrochloride (PMZ)

0.10 mM of PMZ was prepared by taking 1ml from stock solution into a 100 ml standard volumetric flask and distilled water was added to make to volume.

Preparation of 0.15 mM solution of promethazine hydrochloride (PMZ)

0.15 mM of PMZ was prepared by taking 1.5 ml from stock solution into a 100 ml standard volumetric flask and distilled water was added to make to volume.

Preparation of 0.20 mM solution of promethazine hydrochloride (PMZ)

0.20 mM of PMZ was prepared by taking 2.0 ml from stock solution into a 100 ml standard volumetric flask and distilled water was added to make to volume.

Preparation of 0.25 mM solution of promethazine hydrochloride (PMZ)

0.25 mM of PMZ was prepared by taking 2.5 ml from stock solution into a 100 ml standard volumetric flask and distilled water was added to make to volume.

Determination of the Critical Micelle Concentration of SDS in Distilled Water at 298K

About 25 ml of an approximately 0.0808 M aqueous stock solution of SDS was prepared using a 25 ml standard flask. 20ml of distilled water was carefully pipetted into the conductance cell and 200 µL of the stock solution of SDS was added. The conductivity was recorded after thorough mixing and temperature equilibration. Addition of 200 µL volume of the SDS was added until 30 additions has been done.

Determination of the Critical Micelle Concentration of SDS in the Presence of Different Concentration of Promethazine Hydrochloride in the Temperature range of 298.15 – 318.15K at 5K Intervals

Different concentration of promethazine hydrochloride 0.1mM, 0.15mM, 0.20mM, and 0.25mM were prepared using distilled water. Each of this solution were used as solvent for preparing stock solution of SDS while pipetting 20ml of the solvent into conductance cells. 200µL of the stock solution of SDS was added to the conductance cell using calibrated micro-pipette and the conductance values were recorded after thorough mixing and temperature equilibration. Addition of 200µL volume of SDS stock was added until 30 addition has been done. The determination was done at 298.15k. The procedure was repeated at 303.15K, 308.15K, 313.15K, and 318.15K

Result and Discussion.

Determination of the Critical Micelle Concentration of SDS in Distilled Water at 298K

The method used in this study was conductivity measurement, which is one of the best ways for determining the CMC of ionic surfactants (13). The values of the CMC of SDS, in the presence of different concentrations of PMZ and ethanol in aqueous systems were determined by conductometric method. The conductivity can be linearly correlated with the surfactant concentration in both the pre-micelle and post-micelle regions. The plot of specific conductivity against the SDS concentration in the absence of any additive at 298.15K is shown in Figure 4.1. In Figure 4.1, the critical micelle concentration (CMC) was graphically determined from the intersection point between two straight line that corresponded to the well-defined part in the conductivity-concentration curve. The slope in the pre-micelle region is greater than in the post micelle region and the ratio of these slopes gives the effective degree of counter-ion dissociation. The CMC value of SDS (8.32 x 10-3 dm-3) reasonably agrees with the range of value reported in the literature (14).

The CMC values of SDS in aqueous solution of promethazine hydrochloride (PMZ) in the range 0.10 – 0.25Mm were determined by conductivity method. Figure 4.2 shows the plots of the values of specific conductivity (k) against SDS concentrations in the presence of promethazine hydrochloride at different temperature while Figure 4.3 depicts the relationship between CMC of SDS at different concentration of PMZ.

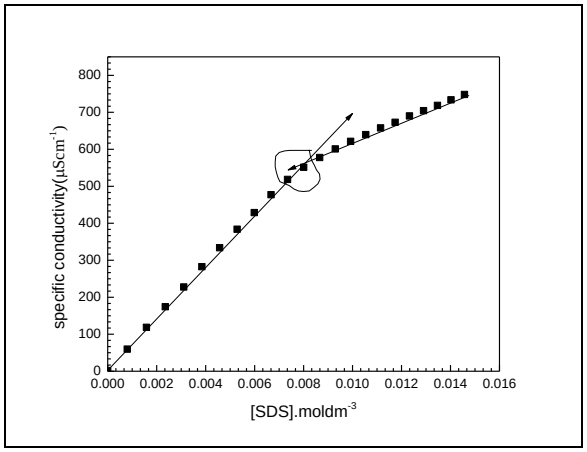


Figure 4.1: Plot of Specific (µScm-1) Conductivity value against the concentration of SDS at 298.15k.

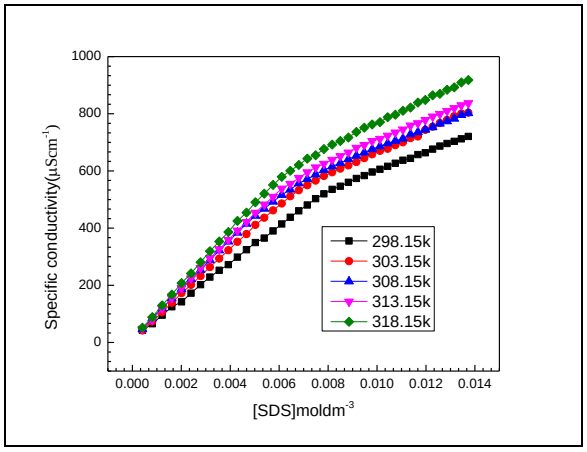


Figure 4.2: A typical plot of specific conductivity versus concentration of SDS in the presence of PMZ at different Temperature.

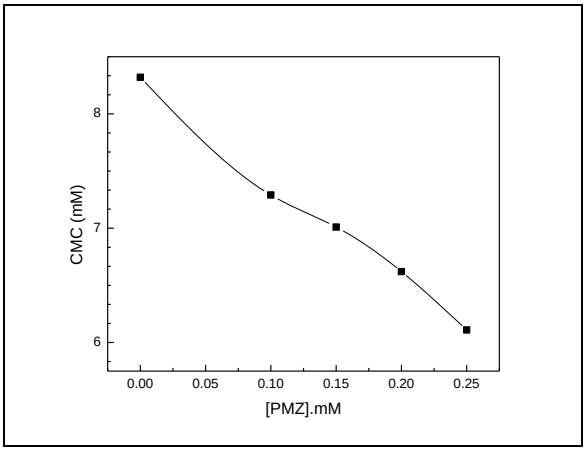


Figure 4.3: Graph of critical micelle concentration (CMC) versus content of promethazine hydrochloride (PMZ) of SDS+PMZ mixture in water at room temperature.

From these plots, significant decrease in the CMC of SDS with increase PMZ concentrations was observed. However, at higher temperatures, it was discovered that the micellization process became stepwise, this made the inflection point very difficult to locate. Similar shortcoming has been reported and solution has been proffered by different authors by an approach that is based on the analysis of the plots of the differential conductivity of first order $\frac{\partial k}{\partial c}$ or second order $\frac{\partial^2 k}{\partial c^2}$, versus surfactant concentrations. In the case of $\frac{\partial k}{\partial c}$ versus [SDS] at different concentration PMZ , it was observed that the curves obtained showed an abrupt fall, with a reverse sigmoid profile. The CMC value is given by the centre of the sigmoid which can be obtained from fitting of the data to a Boltzmann-type reverse sigmoid.

$$\frac{\partial k}{\partial C} = \frac{A_1 - A_2}{1 + \exp((C - C_0)/\Delta C} + A_2 \quad \text{……..4.1}$$

Where, k is the specific conductivity, c, is the total concentration of surfactant, A1, and A2 are the upper and lower limits of the sigmoid, respectively. The CMC value is the center of the sigmoid and Δc is called time constant, which is directly related to the independent variable range where the abrupt change of the dependent variable occurs. The CMC and the degree of counter-ion dissociation values (α = A2/A1) obtained directly from the fitting of conductivity versus concentration plot to Eq. 4.1

A direct integration of Equation 4.1 yields:

$$k(c) = k_{(c=0)} + A_1 C + \Delta C(A_2 - A_1) \ln\left[\frac{1 + e^{((C - C_0/\Delta C)}}{1 + e^{-(C_0/\Delta C)}}\right]$$

The CMC values of SDS, both in the presence and absence of promethazine are presented in Tables 4.1 – 4.5

Table 4. 1: Critical micelle concentration (CMC) and degree of counter ion binding (β) of SDS in the absence of PMZ at different temperature.

Surfactant	T/K	CMC(Mm)	χ_{cmc} (mM)	$\ln \chi_{cmc}$ (mM)	β	$\overline{\beta}$
SDS + H_2O	298.15	8.32	0.130	-8.946	0.589	
	303.15	7.90	0.124	-8.991	0.576	0.58
	308.15	7.10	0.113	-9.085	0.568	
	313.15	8.50	0.133	-8.927	0.555	
	318.15	8.90	0.138	-8.880	0.539	

Table 4.2: Critical micelle concentration (CMC) and degree of counter ion binding (β) of SDS in the presence 0.1mMkg^{-1} of PMZ at different temperature.

Surfactant	T/K	CMC (mM)	χ_{cmc} (mM)	$\ln \chi_{cmc}$	β	$\overline{\beta}$
SDS + 0.1(mM) PMZ	298.15	7.29	0.131	-8.939	0.585	
	303.15	7.20	0.130	-8.951	0.578	0.594
	308.15	7.21	0.125	-8.965	0.565	
	313.15	7.25	0.130	-8.944	0.575	
	318.15	7.59	0.137	-8.899	0.671	

Table 4.3: Critical micelle concentration (CMC) and degree of counter ion binding (β) of SDS in the presence 0.15mMkg^{-1} of PMZ at different temperature

Surfactant	T/K	CMC (mM)	χ_{cmc} (mM)	$\ln \chi_{cmc}$	β	$\overline{\beta}$
SDS + 0.15(mM) PMZ	298.15	7.01	0.126	-8.978	0.512	
	303.15	6.73	0.121	-9.019	0.534	0.577
	308.15	6.22	0.112	-9.098	0.642	
	313.15	6.92	0.124	-8.994	0.584	
	318.15	7.43	0.134	-8.920	0.615	

Table 4.4: Critical micelle concentration (CMC) and degree of counter ion binding (β) of SDS in the presence 0.20mMkg^{-1} of PMZ at different temperature

Surfactant	T/K	CMC (mM)	χ_{cmc} (mM)	$\ln \chi_{cmc}$	β	$\overline{\beta}$
SDS + 0.20(mM) PMZ	298.15	6.62	0.119	-9.035	0.589	
	303.15	6.57	0.118	-9.043	0.581	0.584
	308.15	5.83	0.105	-9.162	0.550	
	313.15	6.81	0.123	-9.007	0.599	
	318.15	7.41	0.133	-8.923	0.601	

Table 4.5: Critical micelle concentration (CMC) and degree of counter ion binding (β) of SDS in the presence 0.25mMkg^{-1} of PMZ at different temperature

Surfactant	T/K	CMC (mM)	χ_{cmc} (mM)	$\ln \chi_{cmc}$	β	$\overline{\beta}$
SDS + 0.25(mM) PMZ	298.15	6.11	0.110	-9.115	0.556	
	303.15	5.57	0.100	-9.208	0.617	0.593
	308.15	5.48	0.010	-9.224	0.581	
	313.15	6.86	0.123	-8.999	0.632	
	318.15	7.05	0.127	-8.972	0.580	

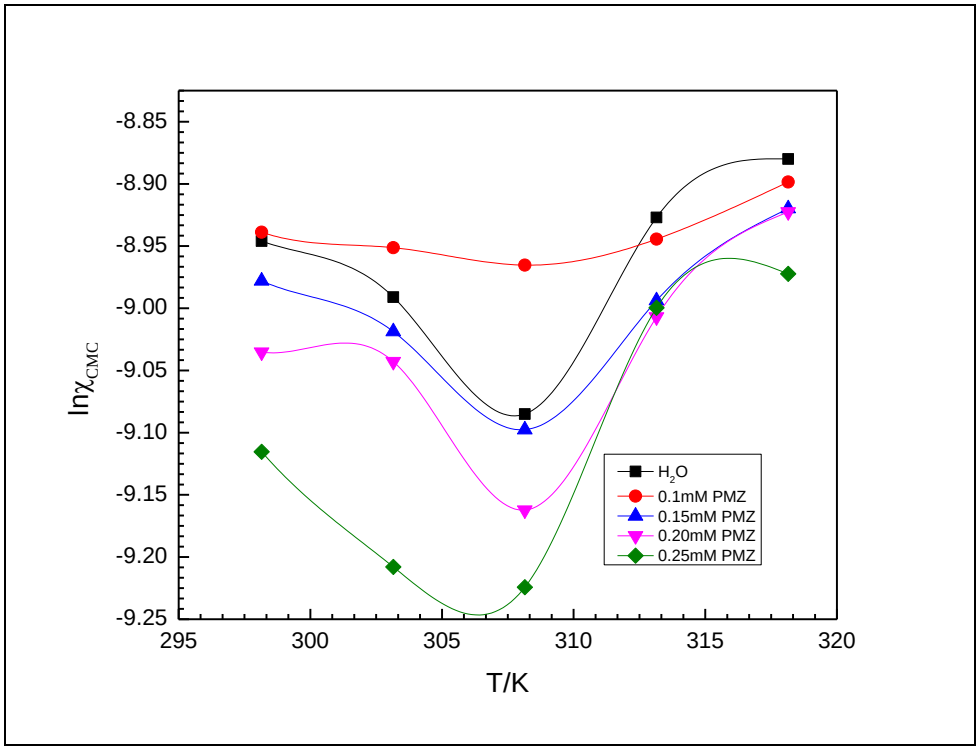


Figure 4.4: Plot of $\ln \chi_{cmc}$ versus temperature in the presence of different concentration of PMZ

The conductivity values of SDS solutions increase with increase in concentration of PMZ in water. Addition of drug leads to easy dissociation of surfactant molecule into ions hence increase conductivity and counter-ion binding values. This might result from the adsorption of drug molecules mainly in outer portion of micelle within the vicinity of micelle–water interface as these are polar organic molecules . Existence of additive PMZ within the external portion of micelle provides steric hindrance to the binding of counter ions. Additionally, the rise within the area per ionic head group occurring as drug enters within the palisade layer of the micelle also assists dissociation of counter ions (15). Variation of β values for SDS within the presence of PMZ at different temperatures is shown in Table 4.1-4.5. The CMC values of SDS (see Figure 4.3) showed endless decrease with PMZ concentration in water. Because the drug (in the salt form is freely dissociated) has an opposite charge from that of surfactant alongside a tricyclic group imparting significant hydrophobic character to the molecule, the effect of such amphiphillic molecules on the micellization behaviour of surfactants requires consideration of both the moieties related to the molecule (16). The CMC value of SDS shifts to lower concentration in solution of PMZ (0.1 and 0.25 mM) as compared thereto in water. Electrostatic interactions between the polar chain of drug and head groups of surfactant molecules in micelle decrease the repulsions among head groups facilitating micellization phenomena. However, at higher concentrations of PMZ, alongside the coulombic interactions, the incorporation of PMZ within the interior of micelle leads to generation of hydrophobic interactions (orientation of molecule is such ionic part is heading towards the outer portion of micelle and rest within the inner). Additionally, H-bonding and dipole–dipole interactions occur between water molecules of medium and polar groups of PMZ. These interactions prevent the incorporation of drug molecules deep within the micellar core, leading to hindrance in the micellization. Thus, polarity of medium features a key role within the probable location of drug molecule within the vicinity of micelle.

Table 4.1-1.5 explain the usefulness of temperature on counter ion binding, which is an important property associated with the micellization process of an ionic surfactant. Counter ion binding is influenced by temperature, nature and concentration of additives, and solvent properties.

As the temperature rises to an extent in all the concentrations, it was seen that the CMC reduces monotonically before increasing again. During the concentration range examined in the current study, because of the interaction between the drug and micelle, which tends to neutralize the charge on the micelle surface and reduce the thickness of ionic compound surrounding the surfactant, as well as the electrostatic repulsion between them that aids in the micelization process, it is evident that CMC decreases as PMZ concentration rises., also increasing the chain length decreases the CMC by increasing the hydrophobic nature of the surfactant

For non-ionic surfactant, CMC decreases with increasing temperature due to an increase in hydrophobicity caused by the destruction of hydrogen bonds between water molecule and hydrophilic group

In conclusion, In aqueous and PMZ/water mixed solvent systems, micellization of SDS in the presence of PMZ revealed that increasing the PMZ content in water caused CMC to decrease to an extent which indicate greater surfactant dissociation. The effect of the PMZ drug on SDS CMC values at different temperatures is not linear. This is due to the intricate interactions between surfactants and PMZ drug molecules caused by altering solvent polarity.

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