Crystal Violet Partitioning in Anionic Surfactants Micellar Media: UV-Visible Spectroscopy Study

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Abstract

Simple and differential UV/Visible spectroscopic method were used to monitored the interaction of Sodium dodecyl sulphate (SDS) and Sodium – *N*- laouryl sarcosinate (SLS) with crystal violet (CV) in pre-micellar and post-micellar concentration range. The results showed that the peak of CV at 590nm was unperturbed in CV/SDS system, but the intensity increased as surfactant concentration increased, unlike in the CV/SLS system. This is an indication that strong interaction between occurred CV and surfactants molecule. Absorption data were also used to obtain partition coefficient (K_c) values and binding constant ($K_{\alpha\beta}$) values from which the free energy of partition (ΔG_c^0) and the free energy of binding ($\Delta G_{\alpha\beta}^0$) were estimated. From the partition coefficient (K_c) values, it could be inferred that the solubilization of CV is more pronounced and spontaneous in the SDS than in the SLS micellar media.

Keywords: Partitioning coefficient; surfactant; sarcosinate; binding constant; micelle.

1. Introduction

Investigations on dye–surfactant interactions in aqueous system have been the line of focus in the last few decades [1–7]. The general picture emerging from these studies is that the peculiar behavior in both absorption spectra and conductivity measurement of dyes [8–10] in the presence of surfactants of opposite charge could be attributed to the formation of a continuum of dye surfactant aggregates [8-10]. Various techniques, such as spectrophotometry [11–14], membrane selective electrode [15], polarography [16], potentiometry [17] and conductometry [18,19] have been used to study the equilibria between surfactant and dye so as to provide information on industrial application like solubilization and on how surfactant assist in fastening of dye in textile industry [20].

Solubilization has been treated as partitioning of additive molecules between a micellar phase and an intermolecular bulk phase [3-5]. Factors like partition coefficient, binding constant, free energies of partition and binding, plus the change in the critical micelle concentration (CMC) of the surfactant in the presence of organic additive, provide useful information relating to the solubilization of additives by micelles [8-10].

Structure of dye and surfactant is important for their mutual interaction as well as dye-fiber and surfactant– fiber interaction, hence, need for optimal surfactant structure for better interaction. Surfactants with different chemical structures give different partition coefficient values for a specific dye. The partition coefficient depends on the structure of the additive, the surfaces that constitute the micelles and other physicochemical variables such as the concentration and temperature.

Crystal violet (CV) is a well-known dye being used for various purposes: a biological stain, a dermatological agent, a veterinary medicine, an additive to poultry feed to inhibit propagation of mold, intestinal parasites and fungus etc. It is also extensively used in textile dying and paper printing [11]. As one of the most widely investigated dyes in terms of its molecular structure, electronic states and the relaxation dynamics of the electronic states [12] with a functional group that is capable of forming hydrogen and covalent bond when interacting with substrate, hence the need for its use as the organic additive in this study.

The surfactants under investigation, for the CV partitioning, are sodium dodecyl sulfate (SDS) and sodium laouryl *N*-sarcosinate (SLS), which are anionic in nature. As shown in Figure 1, both surfactants have 12-carbon atoms at the hydrophobic end. The main difference in their structure is that, Sodium-*N*- laouryl sarcosinate has amide bond connecting the hydrophobic tail and the polar head group while sodium lauryl sulfate has a sulfonic acid group instead.

In the present study, the presence of amide and carboxylate groups, at the hydrophilic end of SLS compared with SDS were investigated using spectrophotometric technique to obtain the optimal head group for interaction. The values of partition coefficients (K_c) and equilibrium binding constants ($K_{\alpha\beta}$) were compared when they both interact with CV in aqueous medium. Availability of amide group in SLS, coupled with the carboxylate group is expected to have either negative or positive effect on the interactive power of SLS over SDS. Observation from this study shall provide an insight into the
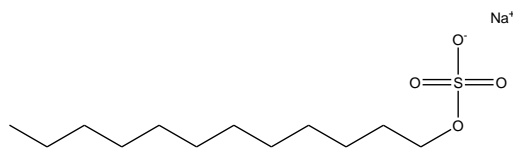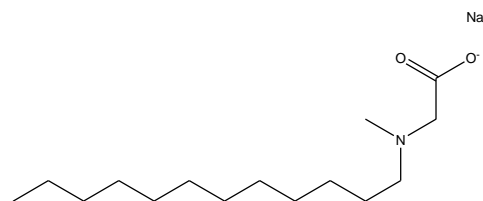
Figure1a. Sodium dodecyl sulfate

Figure 1b. Sodium laouryl N-sarcosinate.

role of hydrogen bonding on the self-assembly of SLS in comparison with SDS as condition for spontaneous solubilization of dye in micellar media.

2. Parameters Calculated
2.1 Calculation of Partition and Binding Parameters

The partition law is the basic law that governs partitioning of additive molecules (dye) between aqueous and micellar medium. The partition coefficient is determined by differential absorbance method reported by Kawamura et al [12]

$$\frac{1}{\Delta A} = \frac{1}{K_c \Delta A_\infty (C_a + C_s^{mo})} + \frac{1}{\Delta A_\infty} \tag{1}$$

C_s denotes dye (CV) in mol/dm^3, C_s^{mo} represents the analytical concentration of CV calculated as C_s-cmc_0 (cmc_0 is the CMC of the surfactant in water) while C_s is the total surfactant concentration. ΔA represents the differential absorbance while ΔA_∞ represents its value when C_s is at infinity i.e. when CV is completely solubilized. K_c is the partition constant having value in dm^3/mol. The dimensionless partition coefficient K_x is related to K_c as $K_x = K_c n_w$, where n_w is the number of mole of water per dm^3 (55.5 mol dm^{-3}).

The standard free energy change for the transfer of additive from bulk water to micellar phase can be calculated using the relation (28);

$$\Delta G_c^o = -2.302 RT \log K_c \tag{2}$$

In Eq. (2), R is the gas constant and T *is the absolute* temperature. Binding constant was calculated from

$$\frac{C_s C_d}{\Delta A} = \frac{C_s}{\Delta \varepsilon l} + \frac{1}{K_{\alpha\beta} \Delta \varepsilon l} \tag{3}$$

Here, C_d represents the concentration of additive (dye), likewise C_s, denotes the surfactant concentration. Also, ΔA and $\Delta \varepsilon$ represent the differential absorbance and the difference of absorption coefficient, l is path length, while $K_{\alpha\beta}$ stands for the binding constant [13-16]

The standard Gibb free energy of binding can be obtained from the relation;

$$\Delta G_{\alpha\beta}^o = -2.302 RT \log K_{\alpha\beta} \tag{4}$$

3. Experimental
3.1 Materials

Sodium dodecyl sulphate (SDS) and Sodium *N*-laouryl sarcosinate (SLS) were obtained from Sigma Chemical Company. Analytical grade CV was obtained from Sinopharm Chemical Reagent Co, Ltd. All the experiment solutions were prepared with double distilled water of specific conductance 2-4 μScm^{-1} at 298.15 K. All experiments were carried out at 298.15±0.2K, except otherwise stated.

3.2 Method

Stock solutions of CV (3.0×10^{-5} moldm^{-3}) were prepared by dissolving the dye in distilled water. Stock solution of SDS and SLS (1.0×10^{-2} moldm^{-3}) surfactants were prepared by dissolving the corresponding amount of each surfactants in distilled water. The absorption spectra of CV (concentration = 3.0×10^{-5} moldm^{-3}) were recorded in aqueous medium and in micellar media of different surfactants using a double beam UV-2100S spectrophotometer setting, having 0.5nm wavelength resolution with 1.0 cm quartz cell, at 298.15K. The additive- surfactants interactions, in terms of partition coefficient and binding constant, were quantified from the absorbance spectral data. For simple UV/visible spectra, distilled water was used as the reference, while in differential UV/visible spectroscopy dye solution of a particular concentration was used as the reference.

4. Results and Discussion
4.1 Interaction of CV with SDS

The UV-Visible absorption spectrum of CV in aqueous medium (Figure 2a) displayed maximum absorbance at 590 nm.
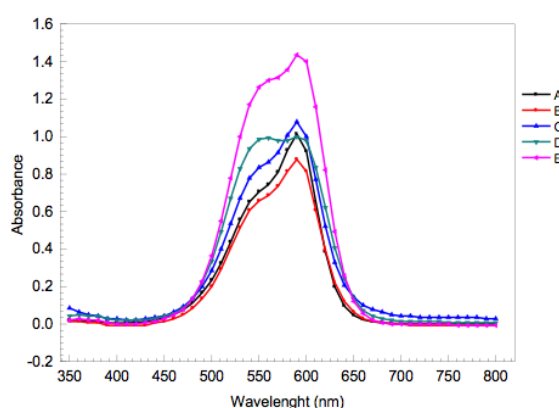

Figure 2a. Absorption spectral of 1.0×10^{-5} moldm^{-3} CV at various concentrations of SDS at 298.15K. Concentrations of SDS (moldm^{-3}): (A) 2.0×10^{-4}; (B) 4.0×10^{-4}; (C) 2.0×10^{-2}; (D) 4.0×10^{-2} and (E) 0.00.

The absorption spectrum of the dye – surfactant combination (CV/SDS system) was recorded from pre-micellar to post-micellar region of SDS concentrations (0.002 mol/dm^3 to 0.04 mol/dm^3) and at fixed concentration (1.0×10^5 mol/dm^3) of CV in aqueous solution (Figure 2a).

The exhibited maximum absorption at 590mn wavelength in the absence of SDS was unperturbed when SDS was added. The absorption spectral of CV with different concentrations of the surfactant (i.e SDS) provided insight into the CV-surfactants interactions. The absorbance intensity increased from a value of 0.5812 at 1.0×10^{-4} mol dm^{-3} concentration of SDS to a value of 2.104 at 1.0×10^{-2} mol dm^{-3} concentration of SDS (Figure 2b).

The increasing absorbance intensity ascribed to a manifest of host – guest relationship between CV and SDS. Incorporation of CV molecules into SDS micelle attracts strong electrostatic interaction between the positive charges on the nitrogen atom of CV with the ionic groups on the SDS. As shown in Figure 2b, the absorbance value increased with increasing concentration of SDS until the critical micelle concentration (CMC) is reached. As soon as the CMC is reached, this absorbance leveled off after reaching a certain value due to maximum incorporation of CV into the micelles [17-22]. In water, the CMC of SDS was 8.01 mM while in the presence of CV, the value decreased to 1.5 mM due to the modification of coulombic repulsions between the SDS charge head groups and possibility of CV aggregating to form micelles. These aftermaths made the micellization entropically more favorable and a decrease in the CMC value is expected.

The results showed that the values of partition coefficient (K_c) for the CV/SDS system was 13.32 dm^3 mol^{-1} while the binding constant $K_{\alpha\beta}$ was 8.30×10^5 dm^3 mol^{-1} (Table 1). Since electrostatic interaction between CV and anionic SDS molecules (head group) was unhindered, CV molecules were forced to prefer solubilized site in the palisade layer of the micelles hence possible large value of partition coefficient (K_c) and binding constant ($K_{\alpha\beta}$) were obtained. The values of $\Delta G_{\alpha\beta}^o$, ΔG_c^o and the related equilibrium constant are summarized in Table 1. The negative values of ΔG_c and $\Delta G_{\alpha\beta}$ (−33.2 and −8.42 $KJmol^{-1}$) for CV/SDS system indicated the spontaneous nature of the partitioning and binding phenomenal as well as the stability of the system [23-29].

Table 1. Partition coefficient K_c , free energy of partition ΔG_c, binding constant $K_{\alpha\beta}$ and free energy of binding $\Delta G_{\alpha\beta}$ for CV/SDS and CV/SLS systems.

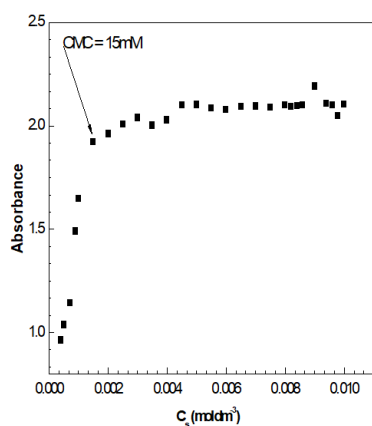| Surfactant	$K_{\alpha\beta}$	$\Delta G_{\alpha\beta}$ (kJ/mol)	K_c	ΔG_c (kJ/mol)
SDS	8.30×10^5	−8.42	13.32	−33.2
SLS	1.96×10^3	−6.03	2.86	−18.8

Figure 2b. Plot of simple absorbance of CV as a function of SDS concentration.

The changes in the differential absorbance of CV with varying SDS concentrations are presented in Fig 3a. The values of partition coefficient (K_c) and the binding constant ($K_{\alpha\beta}$) for the CV/SDS system were obtained from Fig. 3b and c, respectively.
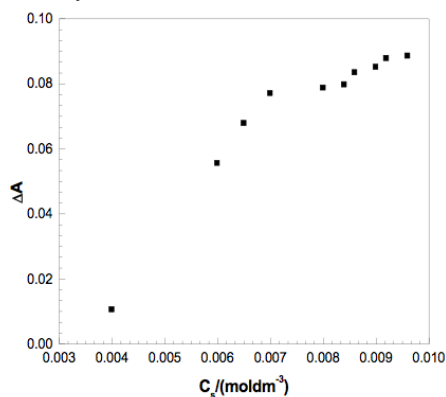

Figure 3a. Plot of differential absorbance of CV as a function of SDS concentrations.
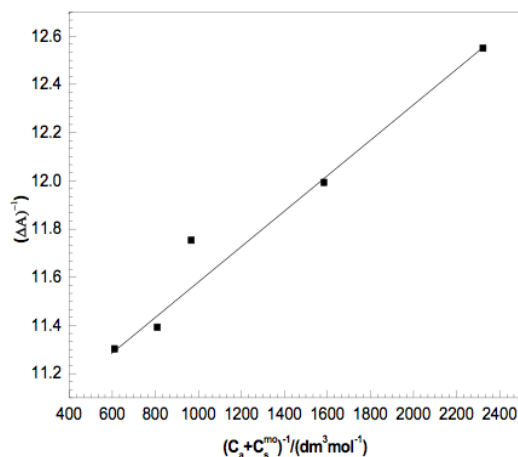

Figure 3b. Relationship between $(\Delta A)^{-1}$ and $(C_s + C_s^{mo})^{-1}$ for the calculation of partition coefficient (K_c) for CV/SDS system.
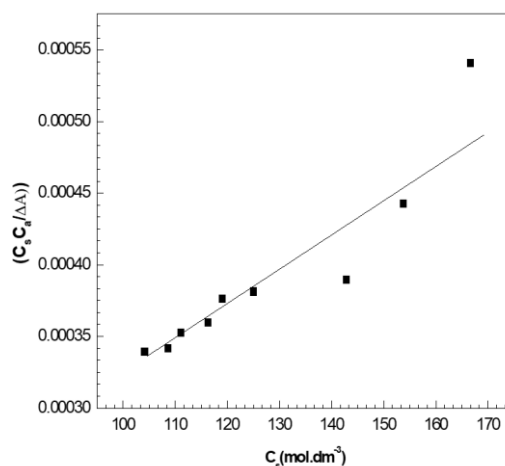

Figure 3c. Plot for calculation of binding constant ($K_{\alpha\beta}$) for CV/SDS system.

4.2 Interaction of CV with SLS

The UV/visible spectrum of CV in the presence of SLS is shown in Figure 4a. It was observed that a red shift in the peak at 590 nm to 55 3nm occurred. The peak at 553nm appeared as a result of the formation of the dimer of the type of the dye which can better be explained in term of dye – surfactant interaction [30].

As the SLS concentrations increase, the dimer disappeared and the peak finally appeared at 596nm. This indicated strong electrostatic interaction between oppositely charged molecule of SLS [31] and compartmentalization of all CV molecules into normal micelles [32]. The result presented in Figure 4b depicted increase in the absorbance of CV as the concentration of SLS increased.
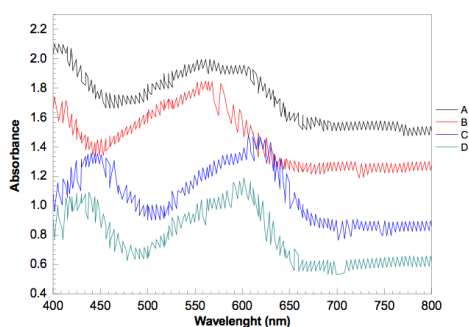

Figure 4a. Absorption spectral of 1.0×10^{-5} moldm^{-3} CV at various concentrations of SLS at 298.15K. Concentrations of SLS (moldm-3): (A) 2.0 X 10^{-4}; (B) 4.0 × 10^{-4}; (C) 2.0 X 10^{-2}; 4.0 × 10^{-2}.
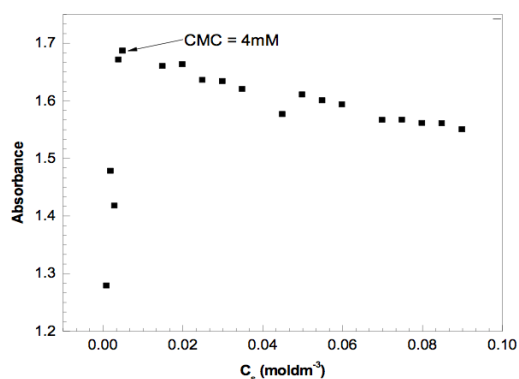

Figure 4b. Plot of absorbance value of CV as a function of SLS concentration.
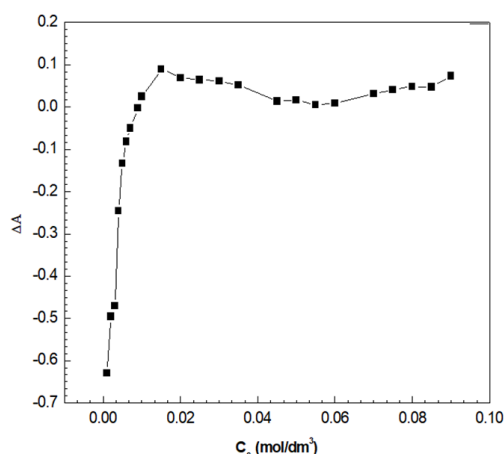

Figure 4c. Plot of differential absorbance of CV as a function of SLS concentrations.

This indicated that incorporation of CV molecules into the micellar core of SLS occurred. Figure 4c showed variation in the differential absorbance of CV with increasing SLS concentrations.

Partition coefficient (K_c) and binding constant ($K_{\alpha\beta}$) values were obtained from Figures 5a and 5b, respectively.
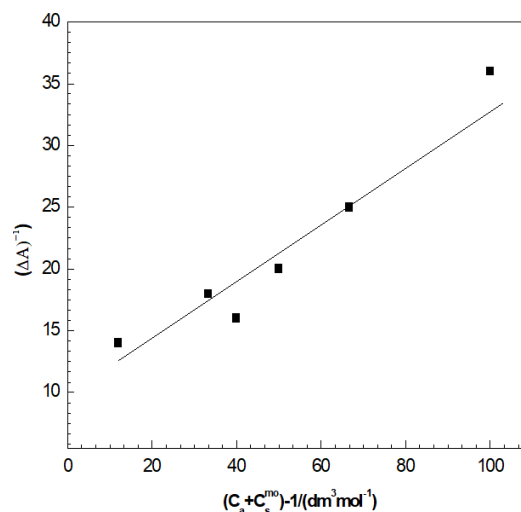

Figure 5a. Relationship between $(\Delta A)^{-1}$ and $(C_s+C_s^{mo})^{-1}$ for the calculation of partition coefficient (K_c) for CV/SLS system.

The value of the partition coefficient (K_c) was 2.86, while the value of binding constant ($K_{\alpha\beta}$) was 1.96×10^3 (Table 1). Since the amide nitrogen in SLS is methylated, thus, the amide bond cannot be a hydrogen bond donor (i.e. no formation of hydrogen bonding) which inhibit intermolecular attraction between the amphiphiles aligned at the palisade region and CV molecules. In addition, the steric hindrance of N- methyl group and the possibility of conformation, *cis* and *trans* of the amide bond [33] of SLS may render close alignment of the amphiphiles difficult. All this behavior ascribed to amide bond, *N*-methylated or not, increased the solubility of SLS in water, hence less values of partition coefficient (K_c) and binding constant ($K_{\alpha\beta}$) as compared with the CV/SDS system. As a resemblance of CV/SDS system, negative values of ΔG_c and $\Delta G_{\alpha\beta}$ is an indication of spontaneous nature of both partitioning and binding process in this study [23-29].
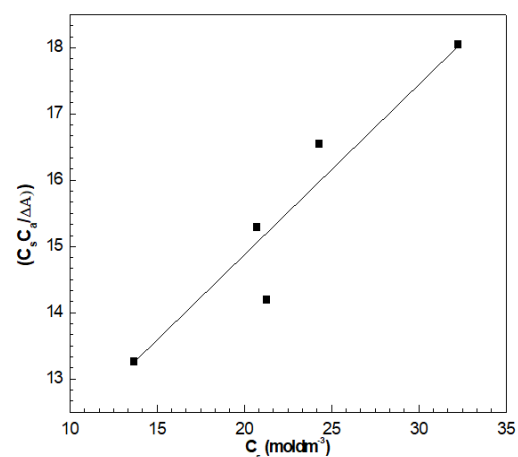

Figure 5b. Plot for calculation of binding constant ($K_{\alpha\beta}$) for CV/SDS system.
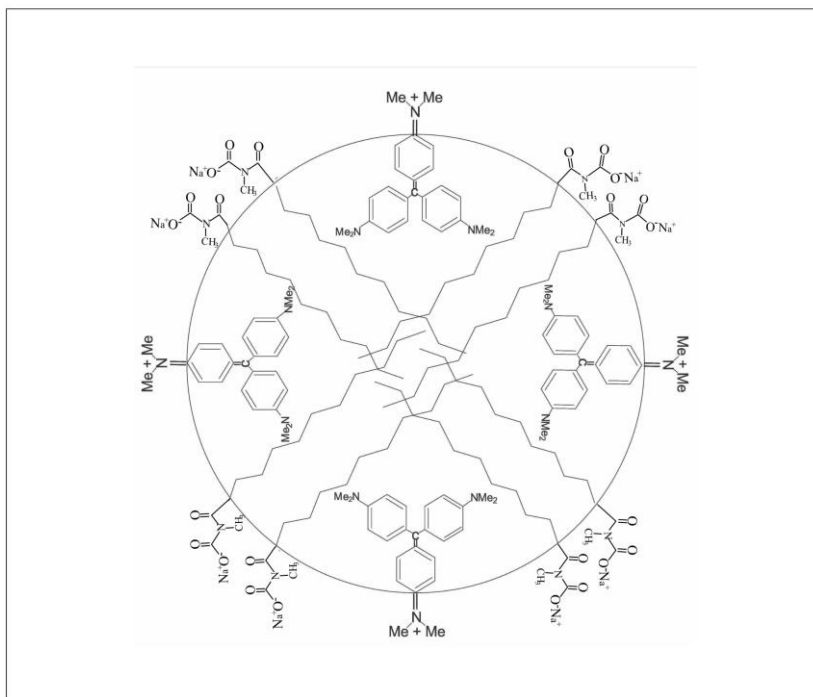
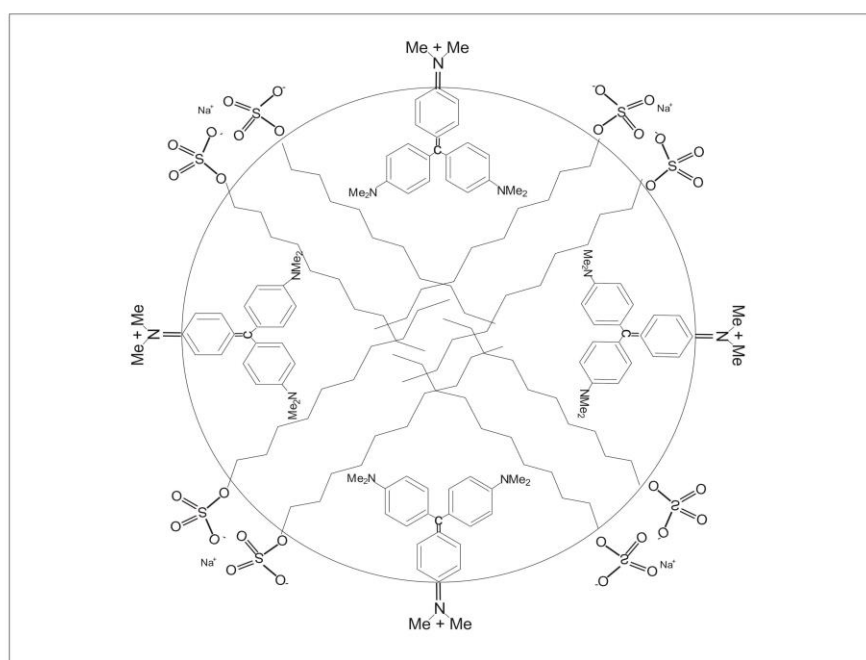*Figure 6a. Proposed locus of CV in SDS micelle.*

Figure 6b. Proposed locus of CV in SLS micelle.

4.3 Comparison of Interaction of CV with SDS and SLS

SDS has a single functional group while the SLS has double functional groups. Experimental evidence showed that the interaction between the localized positive charge on the N – atom on the CV and anionic surfactants (SDS and SLS) enhanced electrostatic attraction between the guest and the host.

Indication from Table 1 showed that partition coefficient for both CV/SDS and CV/SLS system were greater than one. This is an indication that CV is more soluble in the micelle medium than in aqueous medium. This observation is more pronounced in CV/SDS system. This was ascribed to the fact that the electrostatic interaction between CV and SDS were unhindered at the palisade layer, where the SDS

molecules are anchored with their head groups at the interface toward water. Figure 6a shows proposed locus of CV in SDS micelle. In the case of CV/SLS system, there is difficulty in alignment of the amphiphiles because of the *N*-methylated amide bond, hence some CV molecule were forced to be solubilized in the aqueous system which reduced the quantity of CV solubilizing at the palisade layer, resulting in reduction in the partition coefficient value. Figure 6b shows proposed locus of CV in SLS micelle. Higher binding constant values were obtained in the CV/SDS system relative to the CV/SLS system, where the value was small. This observation was ascribed to the negative inductive effect of the alkyl chain of both surfactants, leading to a decrease in electron density on –

SO$_4^-$ group in the SDS than it can do on –COO$^-$ in the case of SLS. This action on CV/SDS system in palisade layer enhanced strong electrostatic interaction between CV and SDS molecules. Hence, a high value of binding constant was obtained for the CV/SDS system (Table 1)

5. Conclusions

In this study, the determination of the level of interaction between CV and anionic surfactants (SDS and SLS) as indicated by absorption spectrum, showed that there was interaction between the dye and the surfactant molecule. Despite the fact that SDS and SLS have the same hydrophobic tail, it was revealed that the charge head group of the surfactant molecule played a prominent role in the dye – surfactant interaction and dye solubilization. Results from this study showed that under the same experimental condition, CV/SDS system has much higher partition coefficient and binding constant values relative to the CV/SLS system. The interpretation of this is that, the degree of solubilization of CV/SDS system was greater than that of CV/SLS system. Partitioning was spontaneous in both systems but it is well pronounced in the case of the SDS micelles.

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