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CETYLPIRIDINIUM CHLORIDE TETRADECYLTRIMETHYLAMMONIUM BROMIDE MIXED MICELLES IN POLYETHYLENE GLYCOL 1000 WATER MIXTURES

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CETYLPYRIDINIUM CHLORIDE + TETRADECYLTRIMETHYLAMMONIUM BROMIDE MIXED MICELLES IN POLYETH- YLENE GLYCOL 1000 + WATER MIXTURES

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ABSTRACT

The conductances of cetylpyridinium chloride (CPyCl) + tetradecyltrimethylammonium bromide (TTAB) mixtures over the entire mole fraction range of CPyCl (x_{CPyCl}) were measured in aqueous binary mixtures of polyethylene glycol 1000 (PEG 1000) containing 0.5, 1, 2, 5 and 10 wt% of PEG 1000 at 30°C. From the conductivity data, the critical micellar concentration (cmc), degree of counter ion association (χ) and the standard free energy of micelle formation (ΔG_M°) of CPyCl and TTAB were computed. The ΔG_M° value is further divided into the hydrophobic contribution of free energy of transfer of the surfactant hydrocarbon chain from the medium to the micelle (ΔG_{HP}°), and the energy associated with the surface contributions (ΔG_s°) consisting of electrostatic interactions between the head groups and counter ions. Both contributions show a linear dependence on the amount of PEG additive. These results have been explained on the basis of the medium effects of aqueous PEG.

The mixed micelle formation by CPyCl+TTAB show non-ideal behavior which is quite similar in the absence, as well as in the presence, of PEG additive as evaluated by using the regular solution theory. The interaction parameter, β , is always negative and remains almost constant with respect to the amount of PEG additive. This indicates that the mixed micelle formation occurs mainly due to the synergistic interactions between the unlike surfactant monomers only.

INTRODUCTION

Micelle formation, in the presence of additives, has been widely studied [1-3]. Recently, a lot of work has been done to understand the surfactant-polymer interactions. This kind of study can have many applications in industrial products such as paints, cosmetics, coating products, and tertiary oil recovery [4-6]. Generally, water soluble polymers such as polyethylene oxide, polyethylene glycol, polyvinyl pyrrolidone etc. are the ideal choice due to their high cohesive energies and considerable hydrogen bonding capabilities which favors the micelle formation in the aqueous media [7]. Different models have been suggested to explain such kinds of interactions e.g. Nikas and Blakshtein [8] proposed the necklace model to explain the various steps in surfactant-polymer interactions. Picullel *et al.* [9] suggested the "spaghetti and meat balls" model to explain the surfactant-polymer interactions. Borisov and Halperin [10] used the concept of polysoap micelles to explain the effect of bridging interactions in polymer-surfactant solutions. In all the theories, the micelle size and the length of the polymer chain are the main factors taken into consideration.

Apart from this, mixed micelles are considered to be more versatile than single surfactant [11-13] and have many applications in surface activity, detergency, wetting, spreading and foaming. Most of the work on the mixed micelle formation has been focused in pure water only [14-18]. However, it is also considered worthwhile to study the mixed micelle formation in the presence of water soluble polymer like polyethylene glycol (PEG). This is due to the fact that mixed micelles have considerable stronger hydrophobic environment rather than single surfactant micelle, therefore, it is expected that such assemblies would have favorable interactions with those of polymer macromolecules. However, the driving force responsible for surfactant-PEG interactions is still poorly understood [19]. Therefore, keeping the above facts in consideration, it was opted to study the micelle formation by single as well as by mixed cationic surfactants in the presence of polyethylene glycol 1000. The measurements have been performed by the conductivity technique which seemed to be the most useful tool in order to detect the micellar transitions accurately due to its high sensitivity and reproducibility. We have limited our conductivity measurements in the concentration region of critical micellar concentration so as to compare the additive effect on the micelle formation by single and mixed surfactants.

EXPERIMENTAL

Materials

Cetylpyridinium chloride (CPyCl), from Fluka, and tetradecyltrimethylammonium bromide (TTAB) from Sigma were recrystallized from ethanol+acetone and ethanol+ethyl acetate mixtures, respectively. Both surfactants were dried *in vacuo* at 60°C for two days.

Polyethylene glycol with average molecular weight of 1000 (PEG 1000) from BDH, England, was used as received.

Methods

Conductivity water having a specific conductance of $4-8 \times 10^{-7} \text{ S cm}^{-1}$ was used in the preparation of all solutions.

The precise conductances of CPyCl+TTAB mixtures over the entire mole fraction range of CPyCl (x_{CPyCl}) in PEG 1000+water (W) containing 0.5-10 wt% of PEG 1000 in their respective binary mixtures were measured at $30(\pm 0.01)^\circ\text{C}$ as explained earlier [20]. The data in pure water have been taken from our earlier work [20]. The error in the conductance measurements was $\pm 0.5\%$.

RESULTS AND DISCUSSION

Figure 1 shows the typical conductivity (k) plots of CPyCl, TTAB and their mixtures in aqueous 2 wt% PEG 1000 over the entire mole fraction range of CPyCl (x_{CPyCl}). Similar plots were also observed in other aqueous PEG mixtures. It can be seen that all curves are tracing the same path in the pre-micellar region whereas distinct curves can be observed in the post micellar region. This is a general behavior of mixed surfactants when they are not associated with each other in the pre-micellar region and a different degree of association in post-micellar region [20]. Single break is evident from each k curve in the concentration range studied for both the single and the mixed surfactants in the present work. However, it is to be noted that in most of the surfactant-polymer studies [7, 22] two breaks were observed in the k curve. The second break is generally observed at comparatively higher concentration of the surfactant. Since the aim of the present work is to study the additive effect of PEG on the micelle formation by single and mixed surfactants, therefore, the k measurements have been

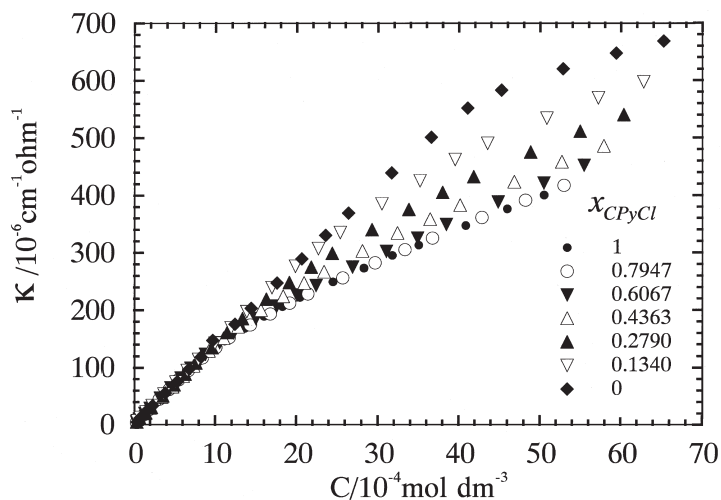


Figure 1. Plot of conductivity (κ) versus concentration (C) of mixed CPyCl+TTAB in 2 wt% PEG 1000.

restricted to the critical micellar concentration region only. From the single break in the κ curve, the critical micelle concentration (cmc) and from the ratio between the slopes of the post micellar region to that in the pre-micellar region, the degree of counter ion association (χ) can be calculated [20, 21].

Micelle Formation by Single Surfactant

In the case of single surfactant, the free energy of micelle formation in the presence of additives, consists of surfactant-surfactant interactions, additive-surfactant interactions and additive-additive interactions. These interactions can be divided into hydrophobic and hydrophilic contributions. In order to estimate both kind of contributions, the thermodynamics of micelle formation, proposed by Evans and Ninham [23], has been taken into account, which gives

$$RT \ln X_{cmc} = \Delta G_{HP}^o + \Delta G_S^o \quad (1)$$

where ΔG_{HP}^o is the hydrophobic free energy of transfer of the surfactant hydrocarbon chain from the medium to the interior of the micelle, and ΔG_S^o corresponds to the energy associated with the surface contributions consisting of electrostatic interactions between the head groups and counter ions and all other contributions due to specific interactions. The sum of these two terms is equivalent to the total Gibbs energy per surfactant molecule ($\Delta G_M^o = RT \ln X_{cmc}$) associated with forming micelles. Both ΔG_{HP}^o and ΔG_S^o were computed by the method reported elsewhere [21, 23].

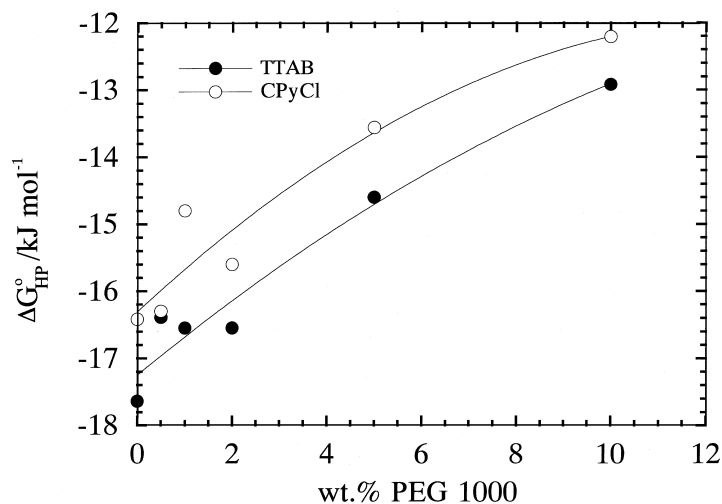


Figure 2. Plot of ΔG_{HP}^o versus wt% PEG 1000 for CPyCl and TTAB.

Figures 2 and 3 show the variation in ΔG_{HP}^o and ΔG_S^o values with the increase in the amount of PEG additive respectively. The increase (less negative) in the ΔG_{HP}^o value (Figure 2) can be explained on the basis of the fact that the transfer of the hydrophobic tail from the medium to the micelles with the

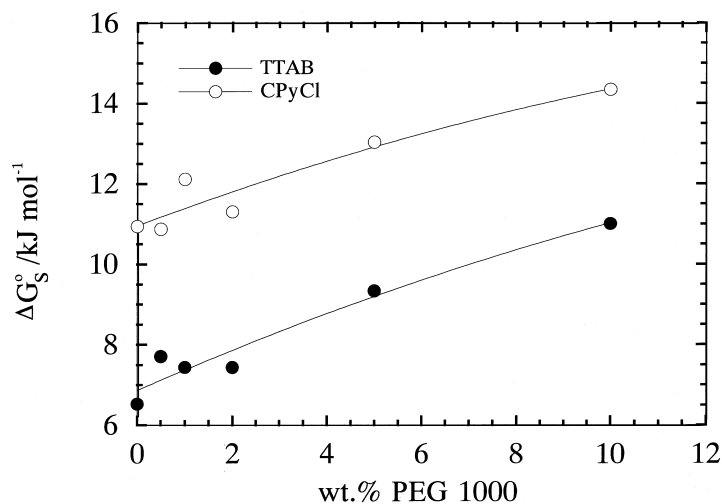


Figure 3. Plot of ΔG_S^o versus wt% PEG 1000 for CPyCl and TTAB.

TABLE 1. Values of Mixed $\text{cmc}/10^{-4} \text{ mol dm}^{-3}$, χ and β of CPyCl+TTAB in PEG 1000+W Systems at 30°C.

x_{CPyCl}	cmc	χ	β	x_{CPyCl}	cmc	χ	β
0.5 wt.% PEG 1000				5 wt.% PEG 1000			
0.000	39.07	0.68		0.000	41.61	0.61	
0.1475	24.63	0.64	-0.76	0.1322	31.46	0.60	-0.32
0.3019	19.81	0.56	-0.63	0.2759	25.01	0.57	-0.40
0.4638	14.58	0.53	-1.2	0.4324	20.31	0.56	-0.60
0.6337	13.04	0.58	-1.4	0.6038	16.63	0.55	-1.0
0.8122	11.98	0.59	-1.8	0.7921	15.47	0.56	-1.0
1.000	11.53	0.60	(-1.1)*	1.000	14.54	0.51	(-0.67)*
1 wt.% PEG 1000				10 wt.% PEG 1000			
0.000	40.70	0.69		0.000	41.91	0.54	
0.1385	27.95	0.64	-0.26	0.1399	29.87	0.46	-0.51
0.2868	22.20	0.53	-0.50	0.2892	23.78	0.48	-0.61
0.4457	16.76	0.56	-1.1	0.4487	20.60	0.47	-0.67
0.6166	15.03	0.56	-1.2	0.6194	16.76	0.50	-1.1
0.8008	12.91	0.55	-1.6	0.8027	15.83	0.50	-0.94
1.000	12.82	0.55	(-0.92)*	1.000	14.84	0.46	(-0.77)*
2 wt.% PEG 1000							
0.000	40.91	0.69					
0.1340	29.24	0.60	-0.41				
0.2790	21.79	0.56	-0.63				
0.4363	17.39	0.56	-0.97				
0.6067	14.02	0.56	-1.4				
0.7947	12.48	0.58	-1.9				
1.000	12.86	0.58	(-1.0)*				

*Average β value

increase in the amount of PEG additive become unfavorable. This can be attributed to a large increase in the concentration of PEG in the aqueous phase (i.e. from 0.5 to 10 wt%). This will certainly influence the medium properties such as relative permittivity (ϵ) and the viscosity (η). Hence, the decrease in ϵ and increase in η value of the medium with the increase in the amount of PEG will result in the hydrophobic hydration of the surfactant monomer with the increase in the solvophobic interactions with the solvent. On the other hand, ΔG_3^0 depends on the electrostatic interactions between the counter ions and the polar head groups on the surface of the micelles. Therefore, any factor which will reduce the binding of the counter ions, will destabilize the micelles by increasing the polar head group repulsions. Our results show that the χ value for CPyCl and TTAB somewhat decreases with the increase in the amount of additive (Table 1). As far as the solubilization of PEG is concerned, there may be two possibilities for the additive macromolecules to accommodate themselves. First, the additive penetrates into the region occupied by the surfactant ionic head groups or the palisade layer which has partially hydrophilic and partially hydrophobic characters; and the second, the additive adsorbs onto the micellar surface or partition itself between the aqueous and the micellar phases which can change the micro environment surrounding the micelle. However, earlier studies [7] have shown that the PEG of moderate molecular weight interacts with the cationic surfactants as the concentration of PEG is increased. Therefore, in the present study, it can reasonably be believed that with the increase in the amount of PEG, it may partition between the aqueous and the micellar phases. Therefore, the second possibility may be working in the present study. This will remove some of the water molecules already bound to the polar head groups and will increase the interfacial free energy between the surfactant monomers and the solvent.

Micelle Formation by Binary Surfactant Mixtures

In the mixed state, the k plots for CPyCl+TTAB mixtures show only single break (Figure 1) which is assigned to the mixed micelle formation by the unlike monomers. The critical micellar concentration (cmc) and the degree of counter ion association (χ) in the mixed state are also calculated in a similar way [20] as explained for pure surfactants.

In CPyCl+TTAB mixtures, the deviation in the mixed micelle formation from the ideal behavior can be evaluated by using the following equation (1) [24, 25]

$$\frac{1}{cmc^*} = \frac{\alpha_1}{f_1 cmc_1} + \frac{(1-\alpha_1)}{f_2 cmc_2} \quad (1)$$

where α_1 is the mole fraction of surfactant 1 in total mixed solute, f_1 and f_2 are the activity coefficients and cmc_1 and cmc_2 are the critical micellar concentrations of components 1 and 2, respectively. The use of this equation is subject to the conditions that the cmc_1 and cmc_2 are independent of the composition so that the ionic strength can be assumed to be constant. Therefore, the cmc_1 and cmc_2 can be defined as the cmc of single surfactants at constant temperature, pressure and ionic strength and that of the mixtures i.e. cmc^* at the same conditions. Hence, the f_1 and f_2 should be equal to 1 in Equation 1. This demonstrates that in fact the CPyCl and TTAB are expected to behave ideally in the mixed state since they have structurally similar hydrophobic tails which will be having the similar kind of interactions as in the homomicelles.

Therefore, in case of the ideal behaviour, Equation 1 can be written as

$$\frac{1}{cmc^*} = \frac{\alpha_1}{cmc_1} + \frac{(1-\alpha_1)}{cmc_2} \quad (2)$$

By using Equation 2, the values for mixed critical micellar concentration (cmc^*) in ideal state were calculated in various PEG+W systems. The extent of non-ideality in the mixed micelle formation between CPyCl+TTAB in the presence of PEG additive can be estimated from regular solution theory [25] by evaluating the interaction parameter, β . The value of β can be computed by using the following Equations 3 and 4

$$\frac{x_1^2 \ln(cmc^* \alpha_1 / cmc_1 x_1)}{(1-x_1)^2 \ln(cmc^* (1-\alpha_1) / cmc_2 (1-x_1))} = 1 \quad (3)$$

where x_1 is the mole fraction of surfactant 1 in the mixed micelle. Equation 3 can be solved iteratively to obtain the value of x_1 , from which β , can be computed using the following equation (4).

$$\beta = \frac{\ln\left(\frac{cmc^* \alpha_1}{cmc_1 x_1}\right)}{(1-x_1)^2} \quad (4)$$

The β values thus computed are also listed in Table 1 along with those for cmc.

When $\beta=0$, the two surfactants form an ideal mixture. The negative β values have generally been ascribed to the synergistic interactions responsible for the mixed micelle formation. The positive β values indicate the incompatibility of surfactant species and thus represent a measure of the antagonistic behavior of the concerned surfactants [26-28].

From the regular solution theory, it is also possible to calculate the excess free energy of mixing (G^E) by using the following equation (5);

$$G^E = \beta x_1 (1-x_1) RT \quad (5)$$

A variation in the mixed cmc versus x_{CPyCl} for CPyCl+TTAB mixtures in the presence of different amounts of PEG 1000 is shown in Figure 4. All the curves show a non-linear variation. A cmc curve in a particular system is gradually shifted towards the higher values with the increase in the amount of additive. Such results can be explained better in terms of the stability of the micelles. Figure 5 shows the variation in G^E . Like cmc, G^E also shows a non-linear variation. However, there is no clear systematic trend shown by the variation in G^E , but it can be seen that the curves are shifted towards the less negative value with the increase in the amount of additive. This suggests that the mixed micelles become less stable as the amount of additive increases. This behavior of mixed

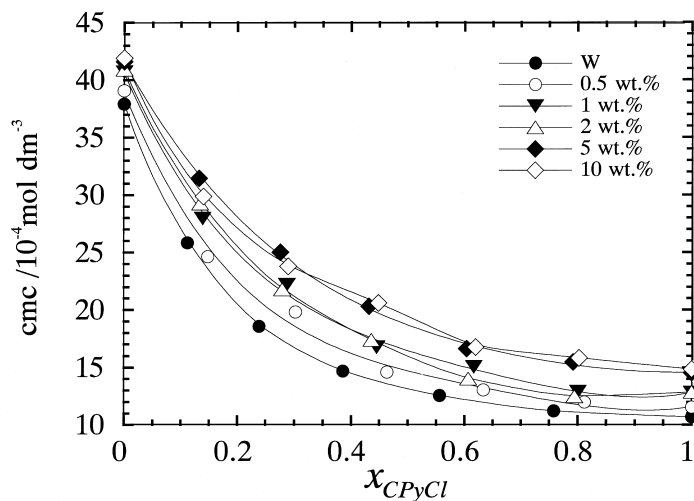


Figure 4. Plot of cmc versus x_{CPyCl} for CPyCl+TTAB in PEG 1000+W systems.

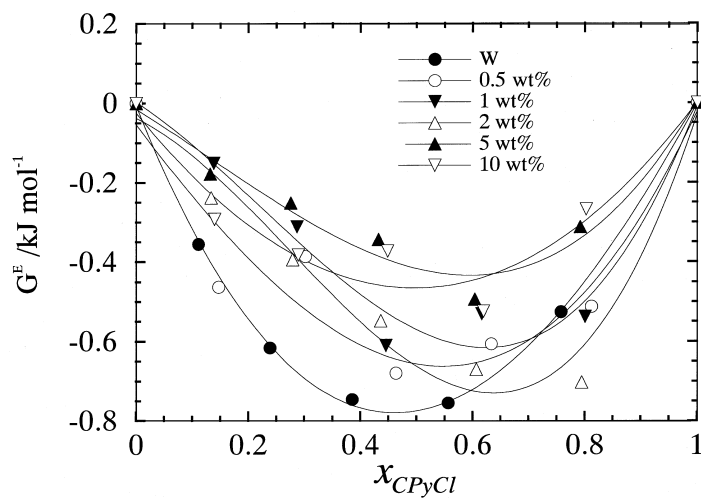


Figure 5. Plot of G^E versus x_{CPyCl} for CPyCl+TTAB in PEG 1000+W systems.

micelles is quite similar to that of the micelles of single surfactants and can be attributed primarily to the same reasons discussed in the previous sections. Similar results were also observed for the same surfactant mixture in the presence of ethylene glycol and diethylene glycol [20].

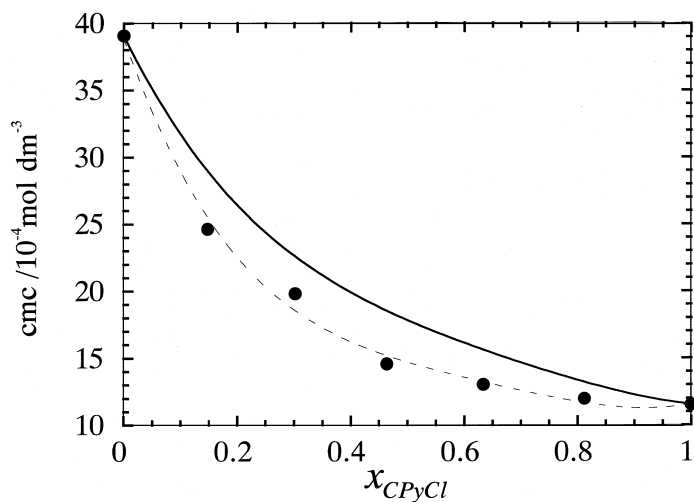


Figure 6. Plot of cmc versus x_{CPyCl} for CPyCl+TTAB in aqueous 0.5 wt% PEG 1000 system. cmc (points); cmc* (solid line).

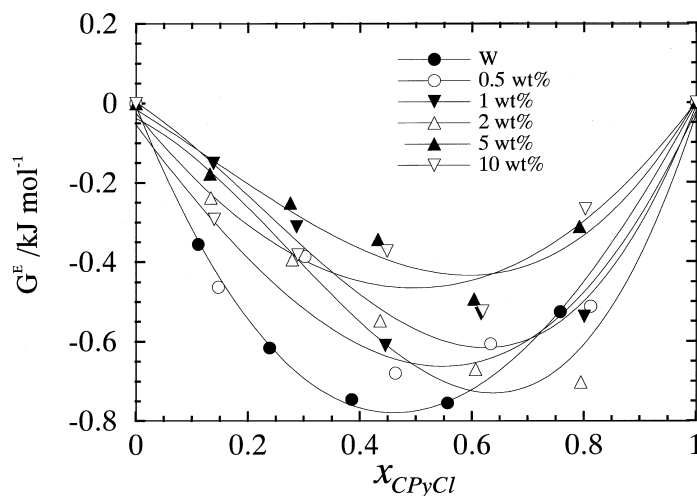


Figure 7. Plot of cmc versus x_{CPyCl} for CPyCl+TTAB in aqueous 10 wt% PEG 1000 system. cmc (points); cmc* (solid line).

In order to explore the extent of non-ideality, in the mixed micelle formation, cmc and cmc* values in the presence of 0.5 and 10 wt% additive have been plotted in Figures 6 and 7, respectively. Contrary to the expectations, in both cases, the experimental mixed cmc values are lying lower to those for cmc* thus indicating a small degree of non-ideality in each case. Similar behavior can also be observed in other aqueous PEG systems. On the other hand, the average β values are always negative (Table 1) and there is no significant dependence on the amount of additive. Interestingly, the average β value in the presence of PEG additive in most of the cases are somewhat close to the value in pure water ($\beta \approx 1.2$) [20] within the experimental uncertainties. However, negative β values suggest that the synergistic interactions are mainly responsible for the mixed micelle formation. The non-ideality is also evident from the variation in surfactant mole fraction (x_1) in the mixed micelle versus stoichiometric mole fraction plot in Figure 8. The x_1 values are significantly deviating from the ideal behaviour. Similar results for another structurally similar cationic binary mixture [29] consisting of dodecylammonium chloride and tetradecylammonium chloride have also been reported earlier and discussed in detail. Since the aim of the present work is to explore the surfactant-polymer interactions, therefore, these results can be better explained from the viewpoint of hydrophobic characters of the micelles and the additive. This is due to the fact that the nature of interactions

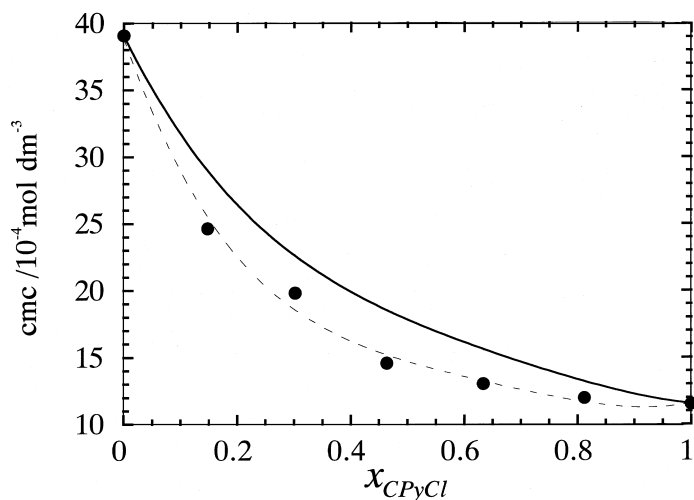


Figure 8. Plot of x_1 versus x_{CPyCl} for CPyCl+TTAB in pure water and aqueous 0.5 wt% PEG 1000. Pure water (filled circles); 0.5 wt% PEG 1000 (empty circles).

between the micelle and the polymer complex formation has not yet been clearly elucidated. Generally, it has been thought [30-32] that the hydrophobic interactions are predominantly playing part in surfactant-polymer interactions. Therefore, considering the above facts, it can be said that the mixed micelles should be the more ideal place for hydrophobic entity to interact rather than the micelles of single surfactants. However, our results show that the mixed micelles are believed to be made up of only CPyCl and TTAB monomers as evident from the nonsignificant variation in the β values with respect to the increase in the amount of PEG (Table 1). This is due to the fact that if PEG macromolecules penetrate into the mixed micelles then it should certainly have a significant influence on the value of interaction parameter, β . Therefore, it seems that though mixed micelle are more hydrophobic than micelles of single surfactant, but they are made up of only surfactant monomers without additive as observed by other authors in the case of single cationic surfactant-polymer systems [6, 7, 3]. Apart from this, the shift in the cmc and G^E curves can be ascribed predominantly to the medium effects as it was observed for single surfactants in previous section since the micelles of single and mixed surfactants are equally affected by the presence of PEG. However, an unsystematic decrease in the χ values (Table 1) with the increase in the amount of PEG additive may suggest the adsorption of

PEG macromolecules at the interface due to which the χ value decreases. Therefore, in such a case, the additive may be staying in the medium or at the interface but does not penetrate into the micelles. Hence, in either case, the additive does not change the shape of the original micelles, but makes the environment different which results in a change in the micellar properties.

CONCLUSION

It has been concluded that the CPyCl, TTAB and CPyCl+TTAB form independent micelles even in the presence of PEG. The micellar properties of single and mixed surfactants are equally affected due to the presence of PEG. This has been attributed to a change in the medium properties. Apart from this, at higher concentration of PEG, PEG is believed to be adsorb at the interface which results in the decrease in counter ion binding significantly especially in the case of mixed micelles. Overall, it can be said that PEG does not have significant interactions with either the micelle of single or mixed surfactants even though the mixed micelles are considered to be more hydrophobic than micelles of the single surfactants.

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