Thermodynamics of Some Amphiphilic Drugs in Presence of Additives

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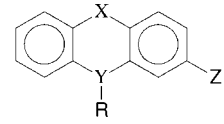
In the present paper, we report the thermodynamics of the four amphiphilic drugs (two antidepressants, amitriptyline hydrochloride and imipramine hydrochloride, and two phenothiazines, chlorpromazine hydrochloride and promethazine hydrochloride) in the presence of additives [NaCl, cetyltrimethylammonium bromide (CTAB), polyethylene glycol *t*-octylphenyl ether (TX-100)] and evaluated Gibbs energies [at the air/water interface ($G^{(s)}_{min}$), the standard Gibbs energy change of micellization ($\Delta_{mic}G^0$), the standard Gibbs energy change of adsorption ($\Delta_{ads}G^0$), and the excess free energy change of micellization (ΔG_{ex})].

Introduction

In aqueous solution, amphiphilic molecules (namely, surfactants, polymers, drugs, etc.) or ions are frequently assembled at interfaces and self-associate in an attempt to sequester their apolar regions from contact with the aqueous phase.[1] The surface-active behavior among many diverse classes of drugs has been reported, and attempts have been made to correlate surface activity and biological activity.[2−6] The aggregation of the above drugs follows the same principles as those of conventional surfactants.[2−6] Their "surfactant-like" behavior is due to the presence of an almost planar tricyclic ring system and a short hydrocarbon chain carrying a terminal nitrogen atom.[2,7] Like ionic surfactants, clouding phenomena of amphiphilic drugs are rare, but under special conditions, they show that clouding phenomenon and phase separation occur.[8−13] The pK_a values of these drugs lie between 9.1 and 9.4,[14] and depending upon the solution pH, the drug monomers may acquire cationic (i.e., protonated) or neutral (i.e., deprotonated) forms.[8] It is well-known that the critical micelle concentration (cmc) values of amphiphiles vary in the presence of additives, because the interfacial and micellar properties of these compounds in solutions are governed by a subtle balance of hydrophobic and hydrophilic interactions. As additives are known to modify those interactions and drugs are used in combination with additives (e.g., salts, surfactants, excipients, etc.), it is necessary to have a knowledge of the additive effect on the cmc and the thermodynamics of amphiphilic drugs. Surfactants have been widely used as a drug delivery vehicle or drug carrier because they have a long shelf-life and simple preparation.

In our previous study,[5] we reported surface properties [in water and in the presence of varying concentrations of sodium chloride (NaCl), cetyltrimethylammonium bromide (CTAB), and polyethylene glycol *t*-octylphenyl ether (TX-100)] of four amphiphilic drugs and their micellar and surface parameters. The work presented herein is aimed at obtaining a better understanding of the role of the presence of additives (NaCl, CTAB, and TX-100) on the thermodynamic quantities of micellization of the four amphiphilic drugs [two tricyclic antidepressants, 3-(10,11-dihydro-5*H*-dibenzo[*a*,*d*]cycloheptane-5-ylidene)-*N*,*N*-dimethyl-1-1-propanamine hydrochloride (amitriptyline hydrochloride, AMT) and 5-[3-(dimethylamino)propyl]-10,11-dihydro-5*H*-dibenz[*b*,*f*]azepine hydrochloride (imipramine hydrochloride, IMP), and two phenothiazines, 2-chloro-10-(3-dimethylaminopropyl)phenothiazine hydrochloride (chlorpromazine hydrochloride, CPZ) and 10-[2-(dimethylamino)propyl] phenothiazine hydrochloride (promethazine hydrochloride, PMT) drugs; see Scheme 1]. The Gibbs energies at the air−water interface ($G^{(s)}_{min}$), the standard Gibbs energy of micellization ($\Delta_{mic}G^0$), the standard Gibbs free energy change of adsorption ($\Delta_{ads}G^0$), and the excess Gibbs energy change of micellization (ΔG_{ex}) are evaluated and discussed.

Scheme 1. Molecular Structure of Amphiphilic Drugs Used in the Present Studies



Amitriptyline (AMT) : X = CH_2CH_2, Y = C, Z = H, R = $C_3H_5N(CH_3)_2H^+Cl^-$
Imipramine (IMP) : X = CH_2CH_2, Y = N, Z = H, R = $C_3H_6N(CH_3)_2H^+Cl^-$
Chlorpromazine (CPZ) : X = S, Y = N, Z = Cl, R = $C_3H_6N(CH_3)_2H^+Cl^-$
Promethazine (PMT) : X = S, Y = N, Z = H, R = $C_3H_6N(CH_3)_2H^+Cl^-$

Materials and Methods

AMT ($\chi \geq 0.980$, CAS Registry No. 549-18-8, Sigma, USA), IMP ($\chi \geq 0.980$, CAS Registry No. 113-52-0, Sigma, USA), CPZ, ($\chi \geq 0.950$, CAS Registry No. 60-09-0, Fluka, Switzerland), PMT ($\chi \geq 0.980$, CAS Registry No. 58-33-3, Sigma, USA), NaCl ($\chi \geq 0.999$, CAS Registry No. 7647-14-5, BDH, England), CTAB ($\chi \geq 0.990$, CAS Registry No. 57-09-0, BDH, England), and TX-100 ($\chi \geq 0.990$, CAS Registry No. 9002-93-1, Fluka, Switzerland) were used as received. Doubly distilled and deionized water [sp. cond. = (1 to 2) $\mu S \cdot cm^{-1}$] was used as the solvent.

The cmc values of the drugs (in the absence or presence of additives) were obtained using surface tension (γ) measurements.[5] The γ−log[drug] isotherms were constructed, and the point of break, when the constancy of γ begins, was taken

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as the cmc of the drug (Figures 1 to 3; see Supporting Information (SI), Tables S1 to S3). The respective uncertainties on the cmc and Π_{cmc} (surface pressure at the cmc) were estimated to be less than (0.1 to 0.3)·10^{-4}, and (0.05 to 0.10)·10^{-4}, respectively. The γ values were measured by the ring detachment method using a S. D. Hardson tensiometer (Kolkata, India).

Results and Discussion

In our previous work,[5] we showed that the cmc values for pure drugs were found in good agreement with the literature values,[3] whereas the values decrease in the presence of additives (NaCl, CTAB, TX-100).

The surface tension values are given in Figures 1 to 3 (SI, Tables S1 to S3). The values of the surface pressure at the cmc (Π_{cmc}) were obtained by using the equation

$$\Pi_{cmc} = \gamma_0 - \gamma_{cmc} \quad (1)$$

where γ_0 and γ_{cmc} are the surface tension of the solvent and the surface tension of the mixture at the cmc, respectively. When increasing the additive concentration, the values of Π_{cmc} increase, indicating that the efficiency increases (Table 1).

The surface excess concentration is an effective measure of the Gibbs adsorption at the liquid−air interface which was calculated by applying the equation[14]

$$\Gamma_{max} = -\frac{1}{2.303nRT}(d\gamma/d\log c)_T \quad (2)$$

where γ, R, T, and c are the surface tension, gas constant, absolute temperature, and concentration, respectively. The variable n is introduced to allow for the simultaneous adsorption of cations and anions. The expression used in the calculation of n was that proposed by Matejevic and Pethica,[15] $n = 1 + m/(m + m_s)$, where m_s is the concentration of the added electrolyte. Thus, n has a value of 2 in water and approaches 1 in the presence of excess inert electrolyte. The slope of the tangent at the given concentration of the γ versus log c plot was used to obtain Γ_{max}, and A_{min} was evaluated using the relation[16]

$$A_{min} = 10^{16}/N_A\Gamma_{max} \quad (3)$$

where N_A is Avogadro's number.

Sugihara et al.[17,18] have proposed a thermodynamic quantity for the evaluation of synergism in mixing, that is, the free energy of the given air−water interface ($G^{(s)}_{min}$) which is defined as follows:

$$G^{(s)}_{min} = A_{min}\Pi_{cmc}N_A \quad (4)$$

$G^{(s)}_{min}$ is regarded as the work needed to make an interface per mole or the free energy change accompanied by the transition from the bulk phase to the surface phase of the solution components. In other words, the lower the values of $G^{(s)}_{min}$ are, the more thermodynamically stable surface is found. The $G^{(s)}_{min}$ values are found to decrease with increasing the additive concentrations/mole fractions (Figure 4A−C, Table 1).

To quantify the effect of additives in the mixture on the micellization process, the standard Gibbs free energy change of micellization, $\Delta_{mic}G^0$, and the standard Gibbs energy of adsorption, $\Delta_{ads}G^0$, were calculated by using eqs 5 and 6,

$$\Delta_{mic}G^0 = RT\ln \mathrm{cmc}_m \quad (5)$$

(cmc_m is the cmc of the mixture of the two components at a given mole fraction)

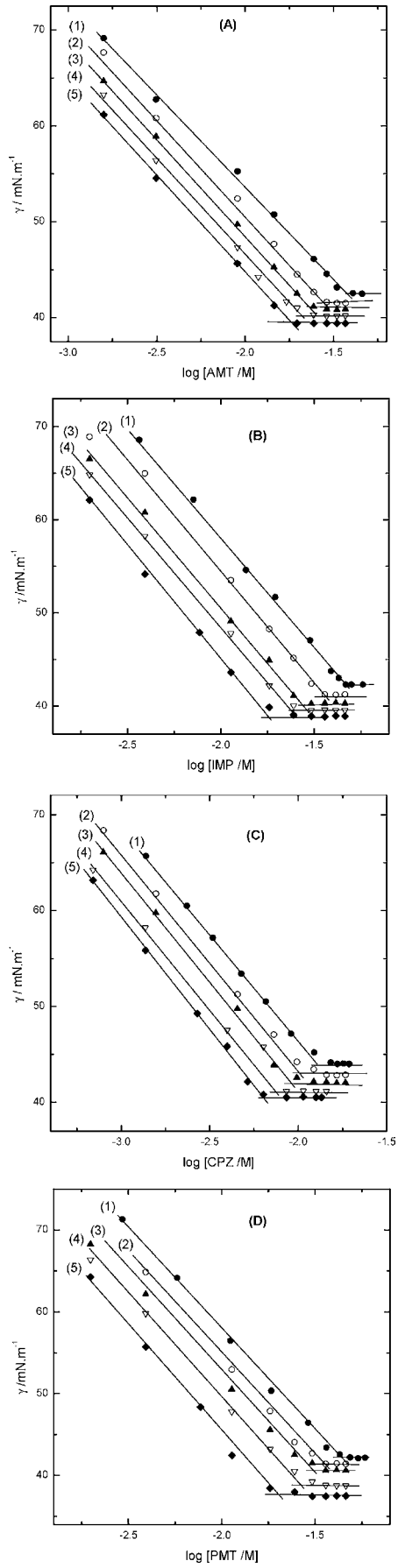
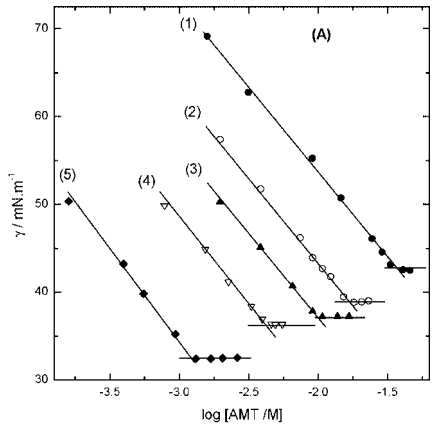
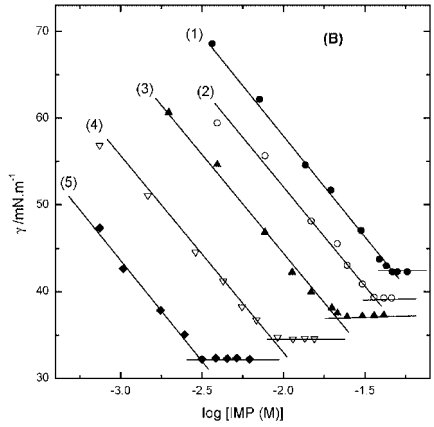
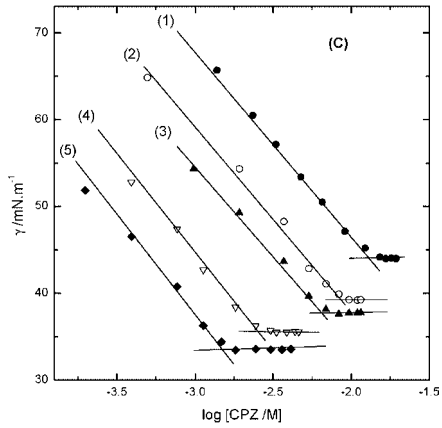


Figure 1. Plots of surface tension (γ) vs the logarithm of AMT (A), IMP (B), CPZ (C), and PMT (D) concentrations at different fixed concentrations of NaCl: (1) 0, (2) 100, (3) 200, (4) 300, and (5) 400 mM.







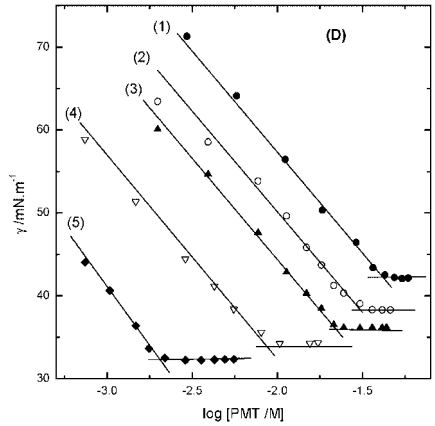
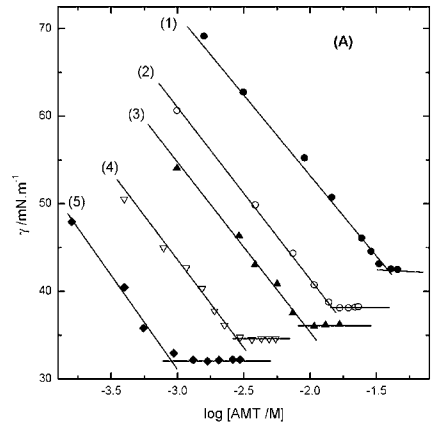
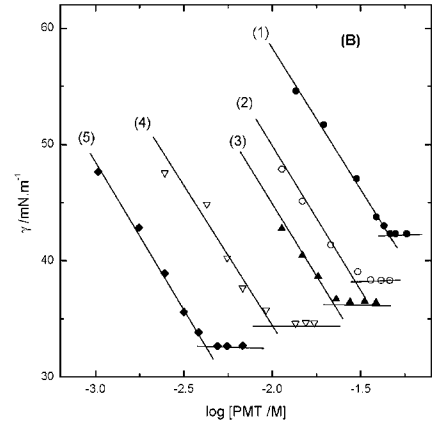
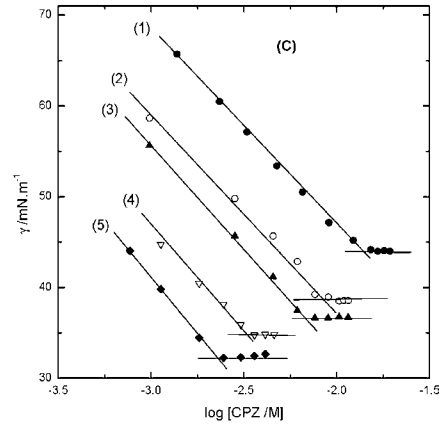


Figure 2. Plots of surface tension (γ) vs the logarithm of AMT (from ref 5) (A), IMP (B), CPZ (C), and PMT (D) concentrations at different fixed concentrations of CTAB: (1) 0, (2) 0.25, (3) 0.50, (4) 0.75, and (5) 1.00 mM.







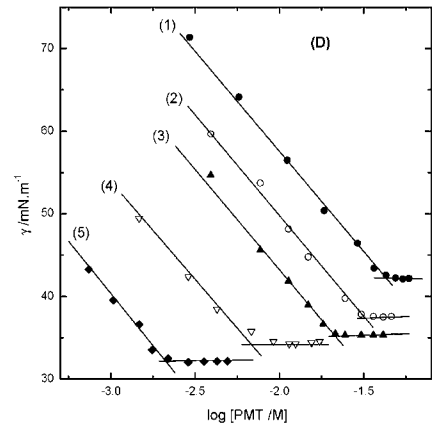


Figure 3. Plots of surface tension (γ) vs the logarithm of AMT (A), IMP (B), CPZ (C), and PMT (D) concentrations at different fixed concentrations of TX-100: (1) 0, (2) 0.075, (3) 0.150, (4) 0.225, and (5) 0.300 mM.

Table 1. Effect of Additive Concentrations on the Surface Pressure at the cmc (Π_{cmc}), the Gibbs Energy at the Air−Water Interface ($G^{(s)}_{min}$), the Standard Gibbs Energy Change of Micellization ($\Delta_{mic}G^0$), the Standard Gibbs Energy Change of Adsorption ($\Delta_{ads}G^0$), and the Excess Energy Change of Micellization (ΔG_{ex}) of Four Amphiphilic Drugs in Aqueous Solutions at 300 K

[additive] / mM	Π_{cmc} / mN·m^{-1}	$G^{(s)}_{min}$ / kJ·mol^{-1}	$\Delta_{mic}G^0$ / kJ·mol^{-1}	$\Delta_{ads}G^0$ / kJ·mol^{-1}	ΔG_{ex} / kJ·mol^{-1}
Amitriptyline Hydrochloride					
NaCl					
0	29.35	26.92			
100	30.40	11.97	−11.836	−20.607	
200	31.05	11.67	−12.276	−21.149	
300	31.75	11.35	−12.556	−21.531	
400	32.50	10.98	−12.951	−22.009	
CTAB					
0	29.35	26.92			
0.25	33.05	11.80	−17.549	−27.588	−1.717
0.50	34.70	11.11	18.790	−29.155	−2.499
0.75	35.50	10.56	−21.010	−31.308	−4.371
1	39.45	8.94	−24.174	−35.048	−7.273
TX-100					
0	29.35	26.92			
0.075	33.80	24.53	−18.853	−40.606	−2.084
0.150	35.90	22.22	−19.979	−42.140	−2.727
0.225	37.40	19.60	−22.883	−44.132	−5.246
0.300	39.75	16.87	−25.684	−46.537	−7.750
Imipramine Hydrochloride					
NaCl					
0	29.60	21.69			
100	30.65	9.92	−11.241	−18.608	
200	31.65	9.90	−11.733	−19.519	
300	32.35	9.32	−12.018	−19.641	
400	33.00	9.01	−12.582	−20.220	
CTAB					
0	29.60	21.69			
0.25	32.65	10.02	−15.582	−23.918	−0.024
0.50	34.75	9.42	−16.821	−25.632	−0.883
0.75	37.35	8.69	−19.122	−28.524	−2.802
1.00	39.70	8.01	−21.701	−31.574	−5.099
TX-100					
0	29.60	21.69			
0.075	33.55	18.75	−16.868	−33.271	−0.366
0.150	35.45	17.51	−17.834	−34.862	−0.909
0.225	37.30	16.15	−19.774	−37.187	−2.476
0.300	39.25	14.53	−21.93	−39.402	−4.319
Chlorpromazine Hydrochloride					
NaCl					
0	27.75	23.27			
100	29.05	10.83	−14.124	−21.469	
200	29.75	10.59	−14.665	−22.142	
300	30.75	10.10	−15.198	−22.744	
400	31.40	9.38	−15.573	−22.843	
CTAB					
0	27.75	23.27			
0.25	32.65	9.93	−19.189	−27.448	−2.199
0.50	34.30	9.39	−19.863	−28.437	−2.502
0.75	36.40	8.82	−22.264	−31.304	−4.835
1.00	38.45	8.19	−23.547	−32.966	−5.615
TX-100					
0	27.75	23.27			
0.075	33.40	19.56	−19.964	−36.936	−1.955
0.150	35.25	18.49	−20.691	−38.469	−2.329
0.225	37.15	17.28	−22.560	−41.029	−6.652
0.300	39.65	15.88	−23.568	−43.095	−4.735
Promathiazine Hydrochloride					
NaCl					
0	29.70	20.11			
100	30.50	9.73	−11.305	−18.471	
200	31.30	9.34	−11.808	−19.009	
300	33.10	8.78	−12.339	−19.825	
400	34.45	8.26	−12.986	−20.583	
CTAB					
0	29.70	20.11			
0.25	33.55	8.86	−16.159	−23.905	−0.528
0.50	35.75	8.11	−16.863	−24.879	−0.812
0.75	37.65	7.51	−19.283	−27.539	−2.932
1.00	39.65	6.72	−22.757	−31.016	−6.078
TX-100					
0	29.70	20.11			
0.075	34.35	17.77	−17.043	−33.297	−0.419
0.150	36.60	16.24	−17.967	−34.801	−0.937
0.225	37.65	15.54	−20.783	−37.867	−3.354
0.300	39.80	14.10	−23.557	−41.044	−5.886

$$\Delta_{ads}G^0 = \Delta_{mic}G^0 - \Pi_{cmc}/\Gamma_{max} \quad (6)$$

Figures 5A−C and 6A−C illustrate that $\Delta_{mic}G^0$ and $\Delta_{ads}G^0$ decrease with the increase in the additive concentrations. The standard state for the adsorbed surfactant is a hypothetical monolayer at its minimum surface area per molecule, but at zero surface pressure. The last term in eq 6 expresses the work involved in transferring the surfactant molecule from a monolayer at zero surface pressure to the micelle. In all of the cases (in absence or presence of additives), $\Delta_{mic}G^0$ values are negative and decrease with increasing additive concentration/mole fraction. This indicates that the micellization is more spontaneous in the presence of the additives (NaCl, CTAB, TX-100; Figure 5A−C). Also, all of the $\Delta_{ads}G^0$ values are negative (Table 1), implying that the adsorption of the surfactants at the air−mixture interface takes place spontaneously (see Figure 6A−C).

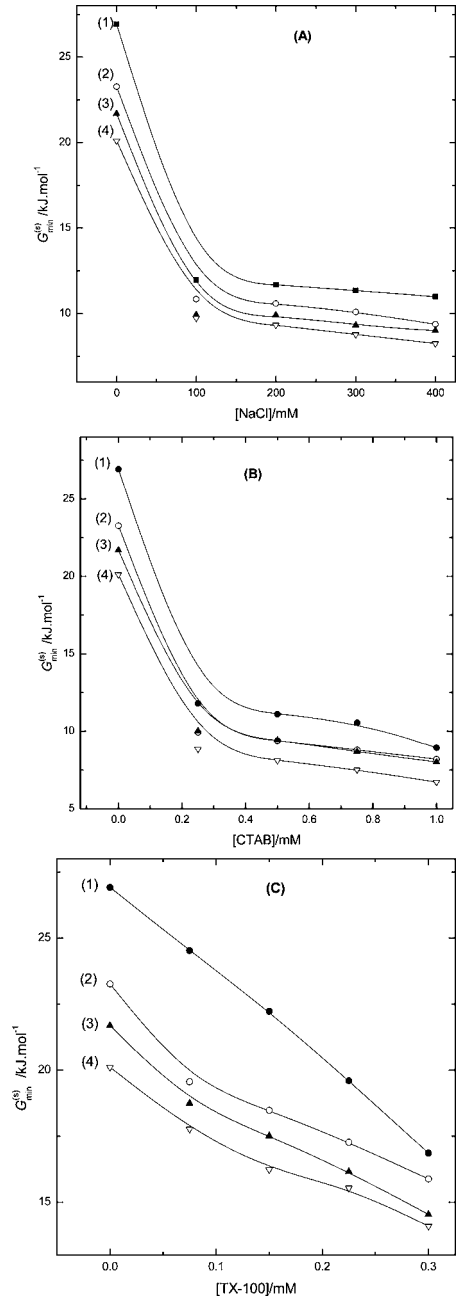


Figure 4. Variation of Gibbs free energy at the air−water interface, $G^{(s)}_{min}$, of the amphiphilic drugs [(1) AMT, (2) CPZ, (3) IMP, and (4) PMT] at different concentrations of additives: (A) NaCl, (B) CTAB, and (C) TX-100.

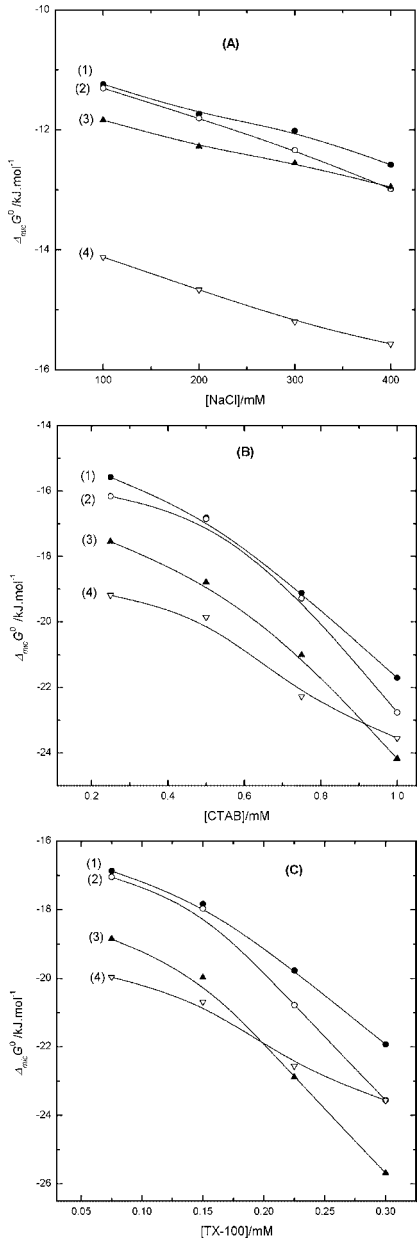


Figure 5. Variation of the standard Gibbs energy change of micellization, $\Delta_{mic}G^0$, of the amphiphilic drugs [(1) IMP, (2) PMT, (3) AMT, and (4) CPZ] at different concentrations of additives: (A) NaCl, (B) CTAB, and (C) TX-100.

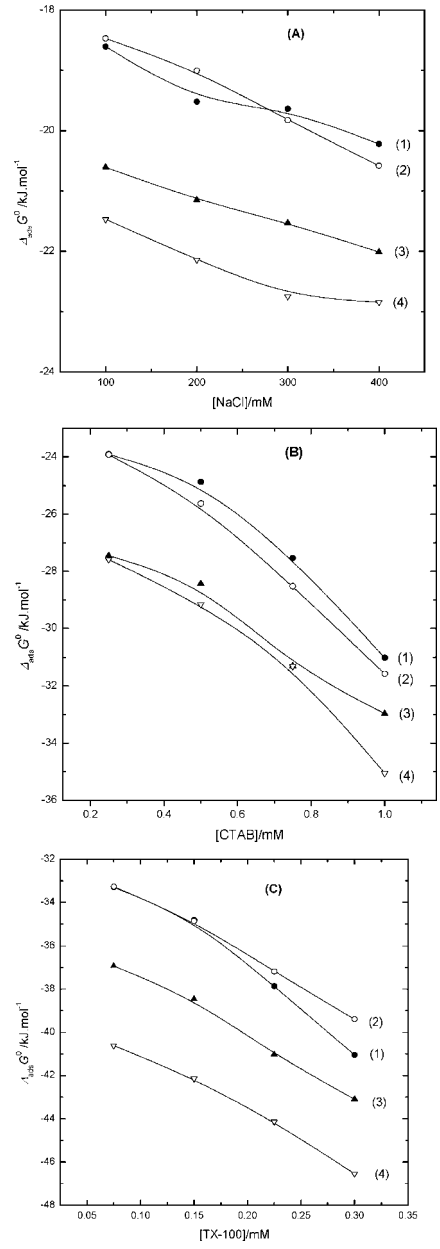


Figure 6. Variation of the standard Gibbs free energy change of adsorption, $\Delta_{ads}G^0$, of the amphiphilic drugs [(1) IMP, (2) PMT, (3) AMT, and (4) CPZ] at different concentrations of additives: (A) NaCl, (B) CTAB, and (C) TX-100.

The nature and strength of the interactions between the drugs and the surfactants can be determined by finding the values of their β^m parameters.[19]

The intermicellar interaction coefficient in the mixed micelles is calculated from:

$$\frac{[(x_1^m)^2 \ln(\mathrm{cmc}\cdot\alpha_1/\mathrm{cmc}_1\cdot x_1^m)]}{[(1-x_1^m)^2 \ln\{(\mathrm{cmc}(1-\alpha_1)/\mathrm{cmc}_2(1-x_1^m)\}} = 1 \tag{7}$$

and

$$\beta^m = \ln(\mathrm{cmc}\cdot\alpha_1\cdot x_1^m)/(1-x_1^m)^2 \tag{8}$$

where x_1^m is the mole fraction of component 1 in the micelles and cmc_1, cmc_2, and cmc are the cmc's for component 1, component 2, and their mixture at mole fraction of component 1, α_1, in the solution.

Equation 7 was solved iteratively for x_1^m, which was then substituted into eq 8 to obtain the β^m values.

The activity coefficients f_1 and f_2 are related to β^m as

$$f_1 = \exp\{\beta^m(1-x_1^m)^2\} \tag{9}$$

$$f_2 = \exp\{\beta^m(x_1^m)^2\} \tag{10}$$

In our previous paper,[5] the significance of β^m values were discussed in detail.

The excess free energy change of micellization, ΔG_{ex}, calculated by using eq 11,

$$\Delta G_{ex} = [x_1^m \ln f_1 + (1-x_1^m)\ln f_2]RT \tag{11}$$

is listed in Table 1 (see Figure 7). On addition, first the surfactants (CTAB/TX-100) get adsorbed and then form mixed micelles. The negative ΔG_{ex} indicates positive synergism.[17] The values of ΔG_{ex} are negative for all mole fractions/concentrations

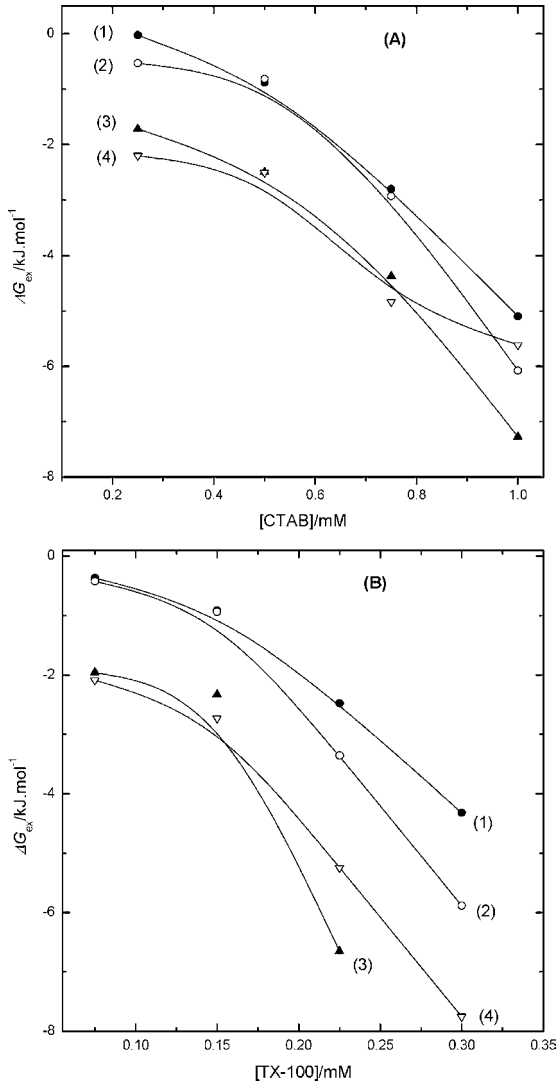


Figure 7. Variation of the excess free energy change of micellization, ΔG_{ex}, of the amphiphilic drugs [(1) IMP, (2) PMT, (3) AMT, (4) CPZ] different concentration of additives: (A) CTAB and (B) TX-100.

of additives, and the magnitude increases (ΔG_{ex} values become more negative) with the increasing additive mole fractions/concentrations, indicating the stability of the micelles as well as more effective (positive) synergism (Figure 7A,B).

Supporting Information Available:

Surface tension values of the four amphiphilic drugs in the absence and presence of NaCl, CTAB, and TX-100 (Tables S1, S2, and S3, respectively). This material is available free of charge via the Internet at http://pubs.acs.org.

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