



# Thermodynamic Micellization of Cationic-Nonionic Surfactants in Aqueous Solution Using Conductivity and Surface Tension Measurements

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## Abstract

In the current work, we discuss the mixed micelles and thermodynamic micellization of aqueous binary mixture of polyoxyethylene - 20 sorbitan-monododecanoate (Tween 20) as nonionic surfactant and Benzyltrimethylhexadecyl ammonium chloride (HDBAC) as cationic surfactant using conductivity and surface tension ( $\gamma$ ) estimations in the temperatures range (288 -318K). Critical micelle concentration (CMC) and variables of micellization, like the standard thermodynamic functions: Gibbs free energy ( $\Delta G_m^0$ ), enthalpy ( $\Delta H_m^0$ ) and entropy ( $\Delta S_m^0$ ) were calculated using the variation of conductivity and  $\gamma$  with molar concentration and the variation of  $\ln X_{CMC}$  with temperature. The experimental CMC values were applied to calculate the mole fractions of surfactant in the mixed micelle ( $X_1^m$ ), the  $\beta$  parameter and the coefficient of activity  $f_1$  and  $f_2$ , using the equations proposed by Clint and Rubingh, which indicate the  $\beta$  parameter, is always negative. In addition, the results of thermodynamic parameters show that  $\Delta G_m^0$  are negative for both individual and mixture of HDBAC-Tween20 surfactants and the values negatively increased with increasing temperature while its negative values decreasing with decreasing initial mole fraction of HDBAC.

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**Key Words:** Thermodynamic Micellization, Tween 20, HDBAC, Conductivity Measurement.

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## Introduction

The researches on surface properties of the mixed surfactants system were thoroughly performed and its application is still growing widely and applied in field of dispersion, moistening, emulsification, and different orientations [Bunton, 1991]. These mixtures often have lower CMC (critical micelle concentration), have better solubility, improved surface activity and improved rheology [Lan, 2007]. Micelles may be viewed as a separate step within a surfactant solution and their physical and chemical properties change dramatically depending on the concentration of surfactant. These properties can be measured as a function of the concentration of surfactant concentration from sharp inconsistency

in the profile which leads to determine the CMC of the surfactant. Various techniques attempts were applied to determine the CMC values of different surfactant systems, like tensiometry [Fainerman, 2009; Inaam, 2019; Fouad, 2020], conductometry study [Akhtar, 2006; Pornpen, 2009; Zarganian, 2011], fluorescence techniques [Kabir, 2010], calorimetric study [Chakraborty, 2007], light scattering [Mohamed, 2015], etc.

The micelle behaviors and associated thermodynamics of the different surfactants depend on their compositions and the temperature play important role on self-assembled structures in solutions such as micellar composition and aggregation number formation in aqueous media.

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There are many attempts to examine the surface properties, micelle mole fractions of the mixed components containing cationic-nonionic surfactant [Rub, 2016; Sanjeev, 2012; Azum, 2013; Bagheri, 2017], which have been studied using a variety of theories like Clint [Clint, 1975] Motomura, [Motomura, 1984] Rosen [Rosen, 1993] Rubingh, [Rubingh, 1979]. These approaches suggest that the electrostatic interactions between cationic amphiphilic surfactant and the nonionic one lead to nonideality of the surfactant mixtures. Also, it predicts type of interactions in air - water interface and micelle between various compositions of surfactants which may be included many interactions such as: electrostatic; steric; van der Waals, and hydrogen bonding among surfactant molecules [Hadgiivanov, 2009].

In this study, thermodynamic functions and parameters were estimated from the interfacial interaction of binary mixture of the surfactant Tween 20 (polyoxyethylene - 20 sorbitan-monododecanoate with 12 Carbon atoms) and Benzyltrimethylhexadecylammonium chloride (HDBAC) using both surface tension and conductivity measurements.

## Experimental

### Materials

Tween 20 was purchased from Sigma Chemical Co., Benzyltrimethylhexadecylammonium chloride (HDBAC) purchased from (BDH) Chemicals Ltd. Both the individual and mixtures of HDBAC-Tween20 solutions are made using deionized water ( $2-3 \mu\text{S cm}^{-1}$ ). Before measuring conductivity and  $\gamma$ , the prepared solution were kept for about 30 minutes to attain equilibrium at certain temperature. Surfactant solution with different concentration and percentage of mixture were prepared by diluting certain amounts of stock

solution in 50 ml volumetric flask with fresh distilled water.

### Surface Tension Measurements

Du Nouys platinum ring on S.E.O. Co. Ltd, tension meter (Korea) was used to measure  $\gamma$ . This ring was cleaned before each measurement and the results were the average of three measurements. The CMC values were estimated from drawing a graph of surface tension against log C and were extrapolated from the break point of after and before micelle areas.

### Conductivity Measurements

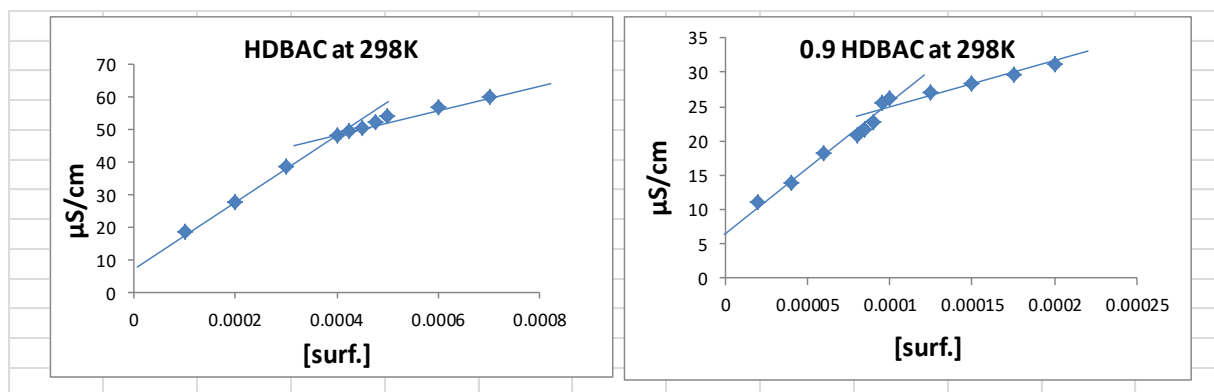
Electrical conductance measurements at four temperatures (288K, 298K, 308K and 318K) were carried with the help of conductmeter type (Cyber Scan 350) from Eutech Instruments equipped with platinum electrode which was washed with deionized water after each reading, the accuracy of conductivity measurements was 0.5%. The conductmeter cell was calibrated initially with standard potassium chloride solutions.

## Results and Discussion

### Conductivity and Surface Tension Results

The conductivity and  $\gamma$  measurements are useful techniques for studying association behavior of many combined systems. The specific conductance and  $\gamma$  for individual and mixtures of HDBAC - Tween20 were established at four different temperatures (288K, 298K, 308K and 318K). Figure 1 represents the relation between conductivity measurements and concentration of pure HDBAC and different mole fraction of binary mixture HDBAC-Tween20 at 298K while figure 2 shows the relation between  $\gamma$  and concentration of pure Tween 20.

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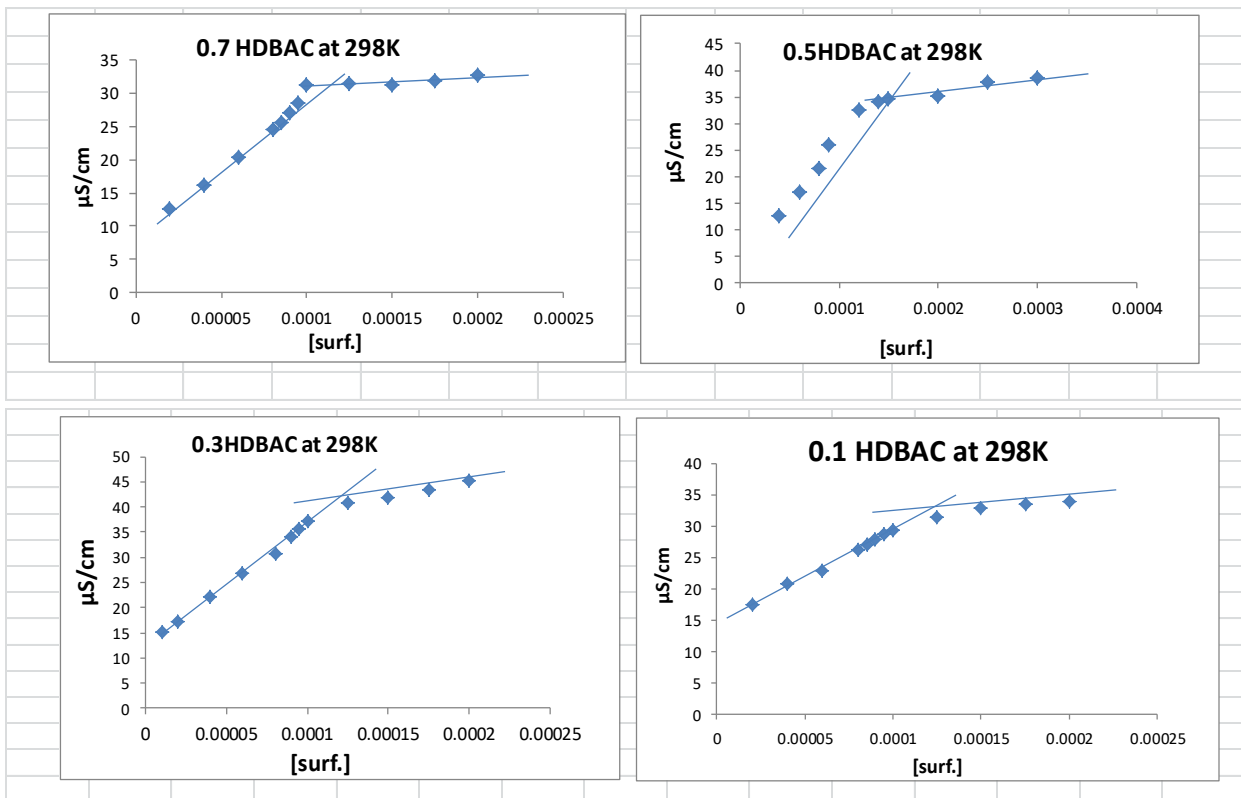


Figure 1. Conductivity measurement against concentration of different mole fraction for HDBAC and Tween 20 at 298K

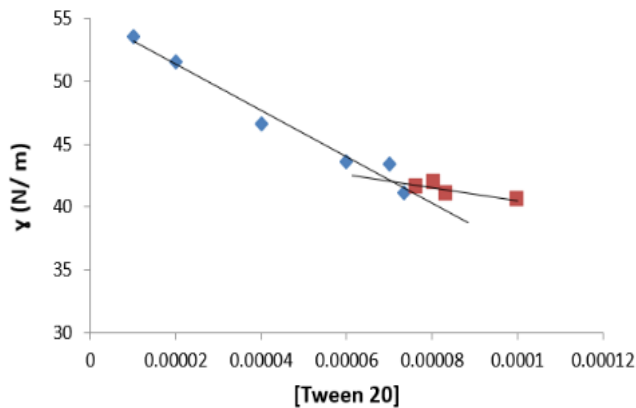


Fig. 2. Surface tension measurements against [Tween 20] at 298K

From crossing point of the two above and below straightforward lines, CMC values can be estimated, and show from the figure, the CMC of HDBAC ( $4 \times 10^{-4}$  mol/L) was drop to around  $1 \times 10^{-4}$  mol/L for different mole ratio of mixture. The magnitude of CMC for single and different mole ratio of mixture at four temperature (288K-318K) was tabulated in Table (1) revealing increasing in the magnitude of CMC with decreasing in the initial mole ratio of HDBAC and also the CMC increasing as the temperature increased. This rise in temperature makes it difficult to form the micelles, owing to the greatly increased hydration of the ion. This may be making the electrostatic interaction of micellization weak [Li, 2017].

Table 1. Values of CMC for individual HDBAC and different molar ratio of binary mixture of (HDBAC + Tween20)

Surfactant	CMC			
	288K	298K	308K	318K
HDBAC	0.00038	0.0004	0.00043	0.00045
0.9 HDBAC	0.000087	0.00009	0.00012	0.00014
0.7 HDBAC	0.000093	0.00015	0.00015	0.00021
0.5 HDBAC	0.000103	0.00022	0.00061	0.00021
0.3 HDBAC	0.000126	0.00030	0.00088	0.0006
0.1 HDBAC	0.000143	0.00035	0.00093	0.0009
Tween 20	0.000076	0.000070	0.000064	0.000073

**Mixed Micelles Formation**

In the mixture of two surfactants, two critical micelle concentrations were existing, one is the ideal value ( $CMC_{ideal}$ ), which is predicted if no interaction between the components was present, and the other is real value ( $CMC_{Exp}$ ) which depends on the form of interactions between the components. For non-



interacting mixed system the ideal critical micelle concentrations can be predicted using Clint model:

$$\frac{1}{CMC_{ideal}} = \frac{\alpha_1}{CMC_1} + \frac{(1-\alpha_1)}{CMC_2} \quad (1)$$

$CMC_1$  and  $CMC_2$  are CMC values of the two pure surfactants;  $\alpha_1$  is Tween 20 mole ratio.  $CMC_{Exp}$  values of HDBAC and its mixtures at various initial mole fractions were calculated using conductivity measurements while CMC values for Tween 20 was extracted using surface tension measurements. The results show negative deviation from ideality where the  $CMC_{Exp}$  values is lower than  $CMC_{ideal}$  calculated as in Fig 3, indicating that a mutual attractive interaction exists in micellization process of HDBAC - Tween 20 surfactants mixture. This is owing to the existence of cationic head groups in conjunction with bulky nonionic surfactant, which reduce the Columbic repulsion forces among them [Malik, 2019].

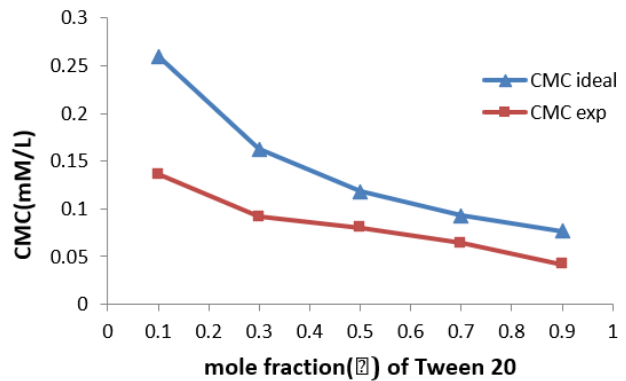


Figure 3. Variation of  $CMC_{ideal}$  and  $CMC_{Exp}$  values with  $\alpha_1$  of Tween 20

The interacting parameter ( $\beta$ ) and the micellar composition of the HDBAC surfactant in the mixed micelle ( $X_1^m$ ) were estimated by equations 2 and 3 and the obtained results are given in table (2):

$$\left(\frac{X_1^m}{1-X_1^m}\right)^2 \frac{\ln\left(\frac{\alpha_1 X_1^m CMC}{(1-X_1^m) CMC_1}\right)}{\ln\left(\frac{(1-\alpha_1) CMC}{(1-X_1^m) CMC_2}\right)} = 1 \quad (2)$$

$$\beta = \frac{\ln\left(\frac{\alpha_1 X_1^m CMC}{(1-X_1^m) CMC_1}\right)}{(1-X_1^m)^2} \quad (3)$$

Table 2. The interaction micellization parameters values for HDBAC-Tween 20 mixed surfactant systems

System (α)	T(K)	$X_1^m$	$\beta^m$	$f_1$	$f_2$	$\Delta G_{ex} \text{ J.mole}^{-1}$
0.1 Tween 20 0.9 HDBAC	288	0.455	-4.665	0.251	0.379	-2.770
	298	0.464	-4.505	0.274	0.378	-2.776
	308	0.472	-3.322	0.396	0.476	-2.120
	318	0.451	-3.293	0.371	0.510	-2.156
0.3 Tween 20 0.7 HDBAC	288	0.589	-2.635	0.640	0.401	-1.527
	298	0.620	-1.647	0.788	0.530	-0.961
	308	0.718	-0.274	0.978	0.867	-0.133
	318	0.738	0.406	1.028	1.247	0.194

The Rubingh model is insolvable up to  $\alpha_1=0.3$  due to the large divergence in CMC magnitude of the two surfactants (13).

From the data of table (2) one can prove that the micellar composition ( $X_1^m$ ) rises with the mole fraction of Tween 20 increase from 0.1 to 0.3 indicating the enrichments in HDBAC molecules. This is owing to the higher hydrophobicity of Tween 20 nonionic surfactant than the cationic surfactant HDBAC and the contribution of the Tween 20 surfactant to the micelle formation is significant contrast to the cationic surfactant HDBAC.

Positive (anti synergism) and negative (synergism) values of  $\beta$  were used to demonstrate mixed micelle formation, whereas a value close to zero corresponded to ideal action [Azum,2017; Rub,2017]. For all mole ratio studied, the values of  $\beta$  are negative meaning the interactions between HDBAC and Tween 20 in the micelle phase are more

attractive than the interactions take place between the single surfactants. Also, the negative value of  $\beta$  for the mixed surfactants system reduces by growing the mole ratio of Tween 20 and this may inform about strong synergism in the forming of mixed micelles [Soheila, 2008; Renu, 2014].

The activity coefficients  $f_1$  and  $f_2$ , which were obtained using the following equations, are the most powerful parameters for describing the degree of interaction between any surfactants present in the mixed micelles. The results of activity coefficients are given in table (2).

$$f_1 = \exp(\beta(1-X_1^m)^2) \quad (4)$$

$$f_2 = \exp(\beta(X_1^m)^2) \quad (5)$$

$f_1$  and  $f_2$  are substituted in equation (6) to estimate  $\Delta G_{ex}$  (the excess Gibbs free energy of mixing) [Motomura, 1993], the results are given in table (2):

$$\Delta G_{ex} = [X_1 \ln f_1 + (1-X_1) \ln f_2] RT \quad (6)$$



The magnitude of activity coefficients,  $f_1$  and  $f_2$  obtained are  $< 1$  which meaning the non-ideal manner of the studied binary systems except for  $\alpha_1$  system which shows a value greater than unity [Patel,2015]. The table also shows the estimated  $\Delta G_{ex}$  values are less than 0 which indicate that the micelles of mixed surfactants studied are more stable than the micelles of HDBAC and Tween 20 in individual forms and the maximum value are observed in case of  $\alpha_1 = 0.3$  system at 298K.

**Micellization Thermodynamic Functions**

As the CMC is temperature dependence, various thermodynamics parameters can be gained from the temperature dependent of surfactants CMC, such as, standard Gibbs free energy of micellization,  $\Delta G_m^0$ , enthalpy of micellization,  $\Delta H_m^0$  and entropy of micellization,  $\Delta S_m^0$ . These functions help us

understand the surfactant behavior and the related importance of the hydrophobic interactions, the contact of water-surfactant and the repulsion of head-group. The  $\Delta G_m^0$  was calculated according to the following equation:

$$\Delta G_m^0 = (2 - \alpha)RT \ln \chi_{CMC} \tag{7}$$

Where R, T and  $\chi_{CMC}$  are gas constant, temperature and CMC in mole fraction.

Also,  $\Delta H_m^0$  was obtained by:

$$\Delta H_m^0 = -(2 - \alpha)RT^2 \left( \frac{\partial \chi_{CMC}}{\partial T} \right) \tag{8}$$

Where  $\left( \frac{\partial \chi_{CMC}}{\partial T} \right)$  was estimated from the plot slope of  $\ln \chi_{CMC}$  against T.

$\Delta S_m^0$  was determined from the next relation between free energy of micellization and the enthalpy of micellization:

$$\Delta S_m^0 = \frac{\Delta H_m^0 - \Delta G_m^0}{T} \tag{9}$$

**Table 3.** Thermodynamic parameters for individual and different mole fraction of HDBAC-Tween20 surfactants mixture

Thermodynamic parameters		Surfactant mole fraction					
		HDBAC	0.9 HDBAC	0.7 HDBAC	0.5 HDBAC	0.3 HDBAC	0.1 HDBAC
288	$\Delta G_m^0$ kJ/mole	-12.083	-10.401	-11.321	-10.752	-9.632	-7.532
	$\Delta H_m^0$ kJ/mole	-5.403	-3.912	-2.786	-4.476	-3.882	-3.880
	$\Delta S_m^0$ J/mole.K	23.191	22.530	29.637	21.790	19.963	12.679
298	$\Delta G_m^0$ kJ/mole	-11.867	-10.257	-10.309	-9.951	-9.486	-7.755
	$\Delta H_m^0$ kJ/mole	-5.785	-4.189	-2.983	-4.793	-4.157	-4.155
	$\Delta S_m^0$ J/mole.K	20.38	20.362	24.585	17.310	17.885	12.082
308	$\Delta G_m^0$ kJ/mole	-8.562	-9.031	-9.043	-8.639	-7.771	-6.371
	$\Delta H_m^0$ kJ/mole	-6.180	-4.474	-3.186	-5.119	-4.440	-4.438
	$\Delta S_m^0$ J/mole.K	7.723	14.794	19.014	11.426	10.814	6.275
318	$\Delta G_m^0$ kJ/mole	-9.504	-4.726	-7.236	-3.627	-3.863	-4.274
	$\Delta H_m^0$ kJ/mole	-6.588	-4.770	-3.396	-5.457	-4.733	-4.731
	$\Delta S_m^0$ J/mole.K	20.440	11.336	12.711	6.052	4.811	0.198

The results of thermodynamic parameters were given in Table 3 which shows the  $\Delta G_m^0$  values are negative for both single and mixture of HDBAC-

Tween20 surfactants indicating the indicating the spontaneity of the micelle formation. Also, the values of  $\Delta G_m^0$  become more negative with



increasing temperature for HDBAC and mixture, while its negative values decreasing with decreasing initial mole fraction of HDBAC. The results of Table 3 show the process of micellization of HDBAC and Tween 20 is spontaneous and there is decreasing in  $\Delta G_m^0$  with rise in temperature which belongs to the desolvation of the hydrophilic groups of these surfactants [Wu, 2013].

The lowering of  $\Delta G_m^0$  with lowering HDBAC mole ratio shows the forming of mixed micelles is decreased. This behavior can be owing to the electrostatic attractions among the charge of head groups so the mixed micelle became unstable [Ren, 2014].

The values of  $\Delta H_m^0$  are negative and increased negatively with increasing in temperature, suggesting the forming of mixed micelle was more proper and micellization process of individual and mixture surfactants is exothermic.

The entropy of micellization  $\Delta S_m^0$  for HDBAC and its mixtures with Tween 20 of all mole fraction ratios are positive, implying that entropy gain promotes the micellization process [Islam, 2003]. Also, the entropy of micellization values decrease with increasing temperature because of increased water structure in its presence as a result of intermolecular hydrogen bonding [Homendra, 2006].

## Conclusions

In this work, we present a focused study on mixed micelles formation and thermodynamic of micellization parameters of dual mixture for cationic surfactant HDBAC and nonionic surfactant Tween 20. The values of CMC of mixtures rise with decreasing in the initial mole ratio of HDBAC, and with an increasing in temperature. The whole  $CMC_{Exp}$  values are less than  $CMC_{ideal}$  values which indicate the presence of attractive interactions resulting in a nonideal behavior. This result is confirmed by the negative values of  $\beta$  parameter over the various mole ratios. This negative variation signals that there are many attractive interactions between the implying strong synergism in the formation of mixed micelles.  $\Delta G_m^0$  values display the micellization of the studied mixtures are spontaneous.  $\Delta G_{ex}$  values obtained suggest that the micelles of studied mixtures are most stable than the micelles of HDBAC and Tween 20 in individual forms.

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