



Rheological Studies of Anionic Surfactants in Presence of Urea Derivatives and Monohydric Alcohols at Constant Temperature

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ABSTRACT: Surfactants are adaptable amphiphilic compounds have an especially distinctive polar hydrophilic head and non-polar hydrophilic tail group. The surfactant is extensively used in each walk of life such as care products, domestic cleaners, pharmaceuticals, oil recovery, food handling, and nanotechnologies due to decreasing the interfacial tension,. The Micellization of anionic surfactants such as potassium dodecyl sulphate (KDS), sodium dodecyl sulphate (NaDS), sodium dodecyl benzene sulphonate (NaDBS) in presence of urea derivatives (2.0 M) in 2.5% alkanol-water systems at constant temperature (35°C) are measured using the Agla micrometer syringe. The conclusion of this study gives the Critical Micelles Concentration (CMC) values of anionic surfactants decreased with the increase in methyl group of urea derivatives, monohydric alcohols (methanol, propanol butanol) and water systems at 35°C.

KEYWORDS: Alcohols, Critical Micelles Concentration (CMC), Surfactant, Surface Tension (γ), Urea, Urea derivatives.

INTRODUCTION

Surfactants molecules structurally consist of polar and non-polar atom (amphiphilic). These molecules occurs a particular type of self-assembly process and the occurrence are identified as micellization. The Anionic surfactants (surface active agents) are amphiphiles with a negatively charged head and are important molecules in detergents, emulsions and industrial applications [1-2]. Surfactants play an important part in preparation of diverse drug delivery systems. Compound, which are not completely soluble in aqueous medium, pharmaceutically acceptable surfactants are characteristically used to improve solubility. The micelles of polymer prepared throughout surfactants acquire a complete set of inimitable features that mark them right positive carriers of the drug for a broad variety of drugs. The important role of surfactants in controlling the particle size of the polymeric nan-particulate organization such as nanocapsules having penicillin-G in double emulsion synthesis; categorization and discharge of drug loaded poly-butyl adipate nanocapsules with penicillin-G.

Surfactants are used extensively in many industries such as food, cosmetics, detergents, paint, pharmaceuticals, petroleum, and more. A speedy development in the usage of surfactants is likely in the years to come. The physiochemical properties of surfactants are largely determined by micelization, and different properties are observed less and greater than the critical micelle concentration [3-6]. Recently, surfactants have been used for environmental remediation, for the elimination of various contaminants from polluted soils and aquifer sediments [7-11].

Urea is an organic compound considered as a renowned denaturant for proteins, polypeptides and biopolymers as it weakens the hydrophobic interactions in aqueous solution. The effect of urea is credited to two different planned mechanisms [12]. One is indirect mechanism, in which urea alters the structure of interfacial water neighboring the solute 13. Some water molecules in the hydration shell of the solute are replaces by urea [14]. Urea and its derivatives, which are well-organized as modifiers of the aqueous meium properties, have established substantial concentration because they are strong protein denaturants, and this result can be considered to be equivalent to the demicellization of micelles in aqueous of urea [15].

Urea is considered as a well-known denaturant for biomolecules such as proteins, polypeptides and biopolymers as it weakens the hydrophobic connections in aqueous solution. The outcome of urea is attributed to two different proposed mechanisms [16]. One is indirect mechanism, in which urea alters the construction of interfacial water surrounding the solute [17]. While in the direct mechanism the replacement of urea by water molecules in the hydration shell of the solute [18].



It is widely recognized that the property of micellar solution are analyzed by a fragile equilibrium of hydrophobic interactions, give a driving force and electrostatic repulsions, which provide an opposing force for micellization [19]. The major factors that affect the behavior of micelles as aggregation due to the nature of the polar head group, the surfactant counter ion, the length and structure of the hydrophobic chain, the presence of additives and the temperature [20, 21].

The formation of micelle in an aqueous solution is known to be affected by inorganic additives and there have been many investigations about the belongings of additives of organic lying on the Critical Micelles Concentration of anionic surfactants. Bahadur *et al.*, (1982-83) [22] observed the effect of additives of organic on the micellar behavior of ionic and non-ionic surfactants in water. Enea *et al.*, (1982) [23] studied the use of urea as a denaturant of proteins. Khurski *et al.*, (1984) [24] observed the change in properties of aqueous solutions in presence of urea and their derivatives. There are two diverse mechanisms have been projected to elucidate the action of urea on aqueous solutions; action of urea as water structure breaker and also participates in the solvation of hydrophobic chains in water due to replacing of water molecules in the hydration shell of the solute.

According to Kabir-ud-din *et al.*, (1996) [25] the critical micelle concentrations (CMC) of ionic and non-ionic surfactants increases significantly by the addition of urea in aqueous solutions. Bahadur *et al.*, (2003) [26] explain the effect of polymer as additives on SDS. Varade *et al.*, (2005) [27] noticed that the micellar behavior of combination of sodium dodecyl sulfate and dodecyl dimethyl amine oxide in aqueous solution. The physicochemical properties of hydroxyl mixed ether HME surfactants and their interaction with sodium dodecyl sulphate proposed by Abdul-Rahem *et al.*, (2009) [28]. The effect of calcium ions concentration on the foaming power of anionic surfactants studied by the Cohen *et al.*, (2009) [29]. The micellization of sodium dodecyl sulfate and polyoxyethylene dodecyl ether in solution noticed by Patel *et al.*, (2009) [30]. Urea induced demicellization of pluronic L-64 in water was observed by Bharatiya *et al.*, (2009) [31]. The anionic-cationic surfactants systems of sodium dodecyl trioxyethylene sulfate with cationic surfactants was noticed by Parekh *et al.*, (2011) [32]. The effect of urea, its derivatives and monohydric alcohol on the micelle formation of anionic surfactants at different temperature as well as constant temperature has been studied by Kumar *et al.*, (2014-2015) [33]. Our studies helped identify the effect of anionic surfactants and urea derivatives in alcohols on surface tension.

EXPERIMENTAL PROCEDURE

Materials and Methods: Sodium dodecyl sulphate, Urea and Ethyl Alcohol (after recrystallization) was used for the preparation of Potassium Dodecyl Sulphate (KDS). It was prepared by direct metathesis and used for physical properties. Double distill water obtained from all Pyrex glass assembly was used throughout studies.

2.5% mixtures of Alcohol and water of several compositions of urea derivatives (2.0 M) were prepared by mixing essential amount of alcohol in water. Reserve solution of surfactants was prepared by weighing. The surface tensions (γ) of surfactant were measured by Agla micrometer syringe (Burroughs Wellcome Co. Ltd. England). The values of CMC were measured at the split point of nearly two in a direct line portions in the γ vs log C plots. The CMC value of the surfactant in presence of urea derivatives, monohydric alcohols in water at stable temperature (35°C) is observed 8.39.

Results and Discussion: The surface tension (γ) of the surfactants in presence of urea derivatives (2.0M) in 2.5% alkanol-water systems at constant temperature of 35°C was measured. Studies however could not be made for higher concentrations of alkanols due to their low solubility of KDS, NaDS and NaDBS in the presence of urea derivatives (2.0M) in 2.5% alkanol-water systems. The CMC values of KDS, NaDS, NaDBS in pure water at constant temperature are reported in Table (1.0 & 2.0). The surface tension (γ) values of KDS, NaDS, NaDBS, using concentration of urea derivatives (2.0M) in 2.5% butanol-water system at constant temperature (35°C) are reported in Table (3.0-5.0) respectively. The surface tension (γ) of all systems decreases with increasing the concentration of surfactants due to increasing the tendency to form aggregates with increasing surfactants concentration. The decreasing order of surface tension (γ -values) of the system in presence of urea derivatives and in different alkanol-water systems studied as:

- 1. Effect of temperature:** The surface tension was decreases with temperature are due to the expansion of surface. The net attractive forces of bulk liquid such that cohesive forces between water molecules are decreased due to increased kinetic energy which ultimately favors aggregation with increasing surfactants concentration.
- 2. Effect of alkanols:** The addition of alcohols as derivatives in surfactants illustrates a decrease in surface tension for a given surfactant concentration which may be due to increase in dimension of the micelle as alcohol is also incorporated into the micelle.

These observations are in close agreement with text.

3. Effect of urea and its derivatives: The lowering in surface tension (γ -values) with rising concentration of urea may be explained in terms of power of cohesive forces. Stronger hydrophobic interactions construct higher cohesive forces for urea due to precious caging of bulk water around them. Hence it develops higher included molecular forces among urea with stronger bond with glass. The rheological properties of surfactants in presence of an organic solvent greatly affected. Lower value of CMC of surfactants in small amount of alcohol might be owing to their straight action on water structure and the subsequent addition may origin secondary effect as their solubilization in micelle and decrease of hydrophobic effect. Urea is a strong water structure breaker; in presence of alcohols it may be destroy the cavity of ordered of water structure whereas CMC should increase with increases in the concentration of urea.

These outcomes indicate that the addition of urea, breaking the water structure event at several concentrations of alcohols and the partition of additives among the solution and micelles may be responsive to the structure of the urea (polar compound) and the temperature. In surfactants solutions, the presence of urea derivatives breaks the hydrogen-hydrogen bonds, thereby wrecking the cohesive forces existing connecting the water molecules. The approaching out tendency for the hydrophobic portion is thus lessened with the result that larger portion of single surfactant molecule can remain in solution. Micelle formation would, therefore take place at higher concentration of surfactants.

In particular, the addition of urea to surfactant solutions leads to an increase in the CMC values. Jencks et al., have proposed that the increased solubility of hydrocarbons in aqueous results primarily from a smaller free energy of cavity formation in the mixed solvent, resulting in the placement of water by a larger urea molecule in the salvation [34, 35].

The degree of contention dissociation of micelles increases with the addition of urea [36]. Consequently the increase in the micelle hydration would increase, which would contribute to the CMC increase. This can be understood from the discussion of the CMC decreasing with addition of urea where the presence of $\text{H}_2\text{N-CO-NH}_2$ group enables the micelle to interact specifically with the compound.

The CMC value of various ionic surfactants such as SDS and HTAB solutions increases with the increasing temperature. The positive temperature coefficient of CMC for ionic surfactants might be due to dehydration of surfactant ionic head groups at elevated temperature. Resulting a stronger repulsion of their ionic head and shifting of monomer to micelle equilibrium in good turn of monomer at higher temperature [36].

Since the dielectric constant of urea is lower than water, urea addition decreases the dielectric constant of the aqueous system [37]. It is due to decreasing the columbic force between the ions of opposite charged Na^+ ion and DS decreasing the dielectric constant of the polar solvent. As a result, the ion pair formation (i.e. micelle) is hindered. This effect is dominating when the concentration of co-solute (urea) is higher (0.2M) or more. Thus, for a particular surfactant, the CMC increases with decreasing the dielectric constant of the polar solvents [38]. Furthermore, micelle formation depends not only on the hydrogen bonding capability but also on the dispersion forces among the alkyl chains of the surfactant [39].

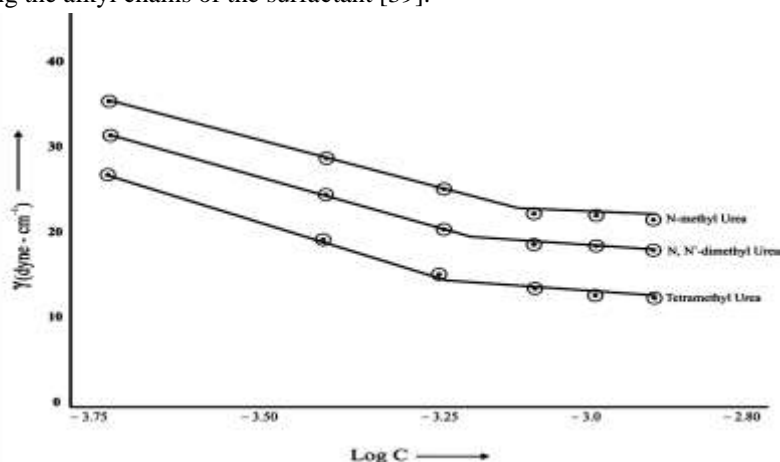


Figure 1: Curve of surface tension vs log concentrations of NaDBs in presence of urea derivatives (2.0 m) in 2.5% butanol-water system at 35 °c.

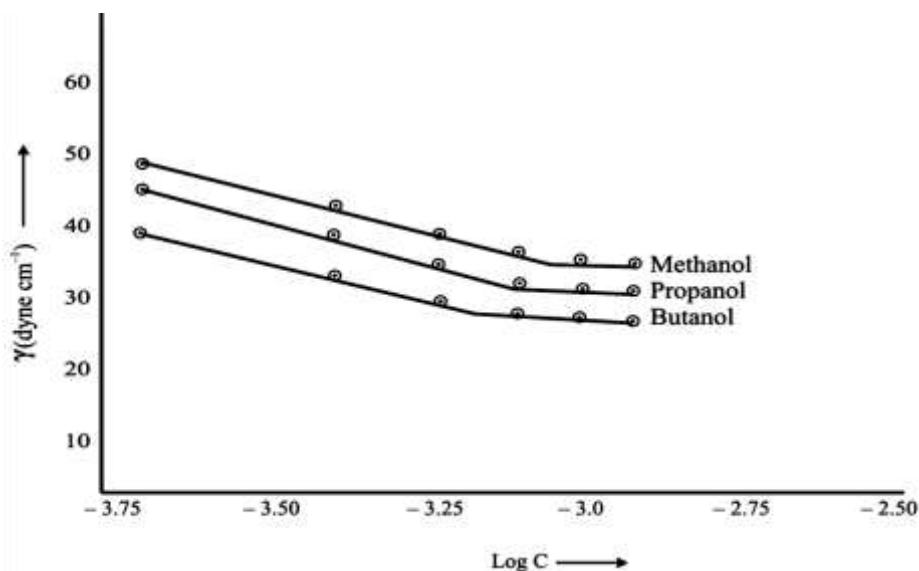


Figure 2: Curve between surface tension and log concentration of NaDBs in presence of urea (2.0 m) in 2.5% alkanol-water systems at 35°C.

Table-1: The values of Critical Micelles Concentration for KDS, NaDS and NaDBS in pure water at different temperatures (30-45°C).

Temperature (°C)	CMC x 10 ³		CMC x 10 ⁴
	KDS	NaDS	NaDBS
30	8.53	8.35	10.0
35	9.00	8.50	12.5
40	9.25	8.65	13.8
45	9.50	8.80	15.0

Table 2: Values of Critical Micelles Concentration for KDS, NaDS and NaDBS in presence of urea (2.0 M) and 2.5% alkanol-water systems at 35°C obtained by γ vs log C plots.

2.5% alkanol-water systems	CMC × 10 ³		CMC × 10 ⁴
	KDS	NaDS	NaDBS
Methanol	5.50	7.10	9.50
Propanol	5.30	6.50	9.00
Butanol	4.95	6.00	8.39

Table 3: Values of CMC for KDS, NaDS and NaDBS in presence of urea and its derivatives (2.0 M) and 2.5% butanol-water system at 35°C obtained by γ vs. log C plots

2.5% butanol-water system	CMC × 10 ³		CMC × 10 ⁴
	KDS	NaDS	NaDBS
Urea	4.95	6.00	8.39
N-methyl urea	4.20	4.93	7.65
N, N ¹ dimethyl urea	3.40	4.66	6.60
Tetramethyl urea	2.50	4.39	6.00



Table 4: Values of surface tension (γ) for KDS in the presence of urea derivatives (2.0 M) and 2.5% butanol-water system at 35°C.

2.5% butanol-water system	Concentration of KDS in Mole litre ⁻¹	Concentration of urea derivatives (2.0 M)		
		N-methyl urea	N, N ¹ dimethyl urea	Tetramethyl urea
		γ (dyne cm ⁻¹)	γ (dyne cm ⁻¹)	γ (dyne cm ⁻¹)
	0.001	24.0	20.5	18.0
	0.002	17.5	16.0	14.8
	0.003	15.0	13.3	11.8
	0.004	14.0	12.3	10.8
	0.005	13.5	11.8	10.0
	0.006	13.0	11.5	9.5
	0.007	12.3	11.3	8.8

Table 5: Values of surface tension (γ) for NaDS in the presence of urea derivatives (2.0 M) and 2.5% butanol-water system at 35°C.

2.5% butanol-water system	Concentration of NaDS in Mole litre ⁻¹	Concentration of urea derivatives (2.0 M)		
		N-methyl urea	N, N ¹ dimethyl urea	Tetramethyl urea
		γ (dyne cm ⁻¹)	γ (dyne cm ⁻¹)	γ (dyne cm ⁻¹)
	0.002	39.3	37.5	35.7
	0.004	36.2	34.6	33.2
	0.006	34.9	33.8	32.7
	0.008	34.7	33.5	32.5
	0.010	34.5	33.3	32.4
	0.012	34.2	33.1	32.2

Table 6: Values of surface tension (γ) for NaDBS in presence of urea derivatives (2.0 M) and 2.5% butanol-water system at 35°C.

2.5% butanol-water system	Concentration of NaDBS in Mole litre ⁻¹	Concentration of urea derivatives (2.0 M)		
		N-methyl urea	N, N ¹ dimethyl urea	Tetramethyl urea
		γ (dyne cm ⁻¹)	γ (dyne cm ⁻¹)	γ (dyne cm ⁻¹)
	0.0002	39.3	37.5	35.7
	0.0004	38.3	34.6	33.2
	0.0006	37.9	33.8	32.7
	0.0008	36.8	33.7	32.5



	0.0010	34.7	33.6	32.4
	0.0012	34.5	33.4	32.2

Table 7: Values of surface tension (γ) for KDS in presence of urea (2.0 M) and 2.5% alkanol-water systems at 35°C.

	Concentration of KDS in Mole litre ⁻¹	Concentration of urea (2.0 M)		
		Methanol	Propanol	Butanol
		γ (dyne cm ⁻¹)	γ (dyne cm ⁻¹)	γ (dyne cm ⁻¹)
2.5% alkanol-water systems	0.001	44.0	36.5	25.0
	0.002	37.5	30.5	22.5
	0.003	36.5	29.0	20.0
	0.004	34.0	27.0	18.5
	0.005	32.0	25.5	17.5
	0.006	31.0	24.5	17.0
	0.007	30.5	24.0	16.5

Table 8.0: Values of surface tension (γ) for NaDS in presence of urea (2.0 M) and 2.5% alkanol-water systems 35°C.

	Concentration of NaDS in Mole litre ⁻¹	Concentration of urea (2.0M)		
		Methanol	Propanol	Butanol
		γ (dyne cm ⁻¹)	γ (dyne cm ⁻¹)	γ (dyne cm ⁻¹)
2.5% alkanol-water systems	0.002	42.5	38.5	33.5
	0.004	38.0	32.5	27.0
	0.006	32.0	29.0	25.5
	0.008	31.0	28.0	25.0
	0.010	30.5	27.3	24.0
	0.012	30.0	27.0	23.5



Table 9:

Table 9.0: Values of surface tension (γ) for NaDBS in the presence of urea (2.0 M) and 2.5% alkanol-water systems at 35°C.

	Concentration of NaDBS in Mole litre ⁻¹	Concentration of urea (2.0M)		
		Methanol	Propanol	Butanol
		γ (dyne cm ⁻¹)	γ (dyne cm ⁻¹)	γ (dyne cm ⁻¹)
2.5% alkanol-water systems	0.0002	47.5	42.5	36.5
	0.0004	40.5	36.5	30.5
	0.0006	37.5	32.5	27.0
	0.0008	34.0	29.0	25.0
	0.0010	32.5	28.5	24.5
	0.0012	32.0	28.0	23.0

SUMMARY CONCLUSION

In this study, comparisons of physico-chemical properties of ionic and nonionic surfactant solutions were systematically made in various compositions of water- Urea mixed system at three different temperatures with the literature values. From the obtained results, it is concluded that both the addition of co-solute and rise in temperature results in an increase in the value of CMC and degree of counter ion dissociation (β) for ionic surfactants.

As the increases the concentration of urea, CMC value increases for studied systems. The values of CMC for pure surfactant systems obtained from experimental result were similar to that of the corresponding literature values. The micellization free energy for the studied surfactants with or without the co-solute (urea) is negative suggesting the feasibility of micellization in the studied system. As the increase in concentration of co-solute (urea) in the $G^0\Delta$ mixed medium, micellization becomes less favorable, as there is less negative micelles.

For the studied ionic surfactant solutions, with or without a co-solute, micellization in the bulk are favored by exothermic enthalpy change as well as entropy gain. For non-ionic surfactant though the enthalpy of micellization $H^0\Delta$ ($S\Delta$ mic) being endothermic, opposes the micelle formation yet predominant positive entropy change (0 mic) is the driving force of micelle formation. Therefore, on the basis of CMC, surface physico-chemical properties and thermodynamic property results in correspondence to literatures, it is concluded that, addition of co-solute (Urea) would not be beneficial for improving detergency and solubilizing capacity of aqueous surfactant solutions for organic compounds.

Surfactants are the surface active agents hold an important role in the established and development of various pharmaceutical products by acting as dispersants, detergents, foaming agents, wetting agents and emulsifiers. Surfactants also perform antimicrobial functions as they stop the nourishment of various pathogenic microbes such like bacteria, fungi, algae, and virus etc and make the pharmaceutical preparations free of harmful microbes.

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