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CMC and solute – solvent interactions of copper surfactants derived from fried oils in benzene at 298.15 K

Renu Bhutra[1]
Rashmi Sharma[2]
Arun Kumar Sharma[3*]

Author Affiliations
[1]Department of Chemistry, Rajesh Pilot Govt. Polytechnic College, Dausa-303303, Rajasthan, India
[2]Department of Chemistry, S.P.C. Govt. College, Ajmer-305001 Rajasthan, India.
[3]Department of Chemistry, Govt. P.G. College, Jhalawar-326001, Rajasthan, India

***Corresponding Author**
Arun Kumar Sharma,
Department of Chemistry, Govt. P.G. College, Jhalawar-326001, Rajasthan, India

E- mail: sharmaarun423@gmail.com

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Abstract

The density, M.V., and A.M.V. of copper surfactant derived from fried and unfried sesame and soyabean oils in non-aqueous and non- polar solvent benzene has been determined at constant temperature 298.15 K. Three samples of each oil were taken fresh, after treating at high temperature for 15 minutes and 60 minutes. The results were used to determine to CMC, chain length effect of the surfactant molecule on CMC and solute-solvent interactions. The studies were used to analyze the effect of high temperature treatment on the structure of the oil and molecular changes. The conclusions have been discussed in terms of well-known Masson's. This helps us to the use of the oils after abuse of frying at high temperature as they have various industrial and biological applications. Colloidal behavior of copper surfactant used in to study of micellar features and their possible uses in agriculture and industries.

Keywords: Copper, Surfactant-solvent interaction, CMC, Density, Apparent molar volume

Abbreviation: M.V. = Molar Volume, A.M.V. = Apparent Molar Volume, CMC= Critical Micelle concentration.

1. Introduction

The surface active compounds have found wide applications in many industrial uses and play an important role in the colloid chemical phenomena [1]. The nature and structure of surfactants derived from transition metals are of great importance for determining their characteristics under various conditions [2-3]. Recently work on polymetallic complexes and transition metal complexes of heterocyclic ligands has been done, and their structure and biological characteristics have also been discussed [4].Various edible oils used in deep fat frying in Indian market are available easily and economically [5]. The use of fried oils as potential biocidal agents and wood preservatives and analysis of their physico-chemical studies will generate new hopes in biological and pharmacological fields [6-7]. In our earlier communication ultrasonic, antifungal, TGA, photochemical degradation of Cu (II) surfactants and their complexes derived from various oils have been reported [8-10]. In this paper we report studies of the newly synthesized and characterized surface active compounds derived from sesame and soyabean oils as unfried, fried at high temperature for 15 and 60 minutes. The parameters of these molecules such as density, M.V., and A.M.V. have been studied and analyzed in a binary system having non -aqueous, non -polar benzene as a solvent which has been chosen due to maximum solubility of the solute. The CMC of copper surfactants have been measured by density in different solvents. While information about solute-solute, solvent-solvent interactions in solutions have been obtain from measurement of A.M.V. Micellar characterizations of copper surfactants have been looked into with the aid of organic solvent (benzene). For this purpose, physical properties like density, M.V., A.M.V. have been enumerated.

2. Experimental

Three samples for each oil have been prepared as fresh (unfried), fried oil at high temperature for 15 minutes and fried oil at high temperature for 60 minutes.

2.1 Preparation of fried oils and surfactants:

The frying of chips experiment with both oil samples at 180-200^{0}C for 15 and 60 minutes were conducted. Copper surfactant was prepared by earlier reported methods [11]. The abbreviations of copper surfactants synthesized are as follows-

A.	From unfried oils-	
1.	Copper surfactant of sesame oil	CSe
2.	Copper surfactant of soyabean oil	CSo
B.	From fried oils for 15 minutes-	
1.	Copper surfactant of sesame oil	CSe_{15}
2.	Copper surfactant of soyabean oil	CSo_{15}
C.	From fried oils for 60 minutes-	
1.	Copper surfactant of sesame oil	CSe_{60}
2.	Copper surfactant of soyabean oil	CSo_{60}

2.2Measurement of Density:

For measuring the density of the complex solutions Ostwald's modification of the springel pyknometer was used.The density ρ were in good agreement to within ± 0.02 kg m^{-3}

$$\rho = \frac{\omega}{\omega_0} \qquad (1)$$

Where w= weights of the same volume of solution
w_0= weights of the water.

2.3 Evaluation of M.V.:

The M.V. of the complex solution Vhas been calculated by the relationship.

$$\nabla = \frac{M_1X_1 + M_2X_2 + M_3X_3}{\rho} \qquad (2)$$

Where
X_1= Mole fraction of the surfactant
M_1= Molecular weight of surfactant
M_2=Molecular weight of benzene
X_2= Mole fraction of the solvent
ρ= density of the solution. The molar volume ∇ were in good agreement in± 0.02 m^{3}mol^{-1}

2.4 Evaluation of A.M.V.:

The A. M. V. has been calculated using the following equation:

$$\phi_v = \frac{M}{\rho_0} + \frac{1000\,(\rho_0 - \rho)}{c.\rho_0} \qquad (3)$$

Where

ρ_0 =density of the solvent,

ρ= density of the surfactant solution,

M= molecular weight of the surfactant

C=concentration of solution. The apparent molar volume ϕvwere in good agreement in ± 0.02 m^3mol^{-1}

3. Results and Discussion

3.1 Density

The density of copper surfactants of unfried oils first increases with increase in surfactant concentration but there is a slight decrease followed by linear increase in CSo solutions. The density of CSe solutions increases regularly. It is due to the increase in the shorter fatty acid content in the corresponding oil composition (Table 1-2).

Table 1: Composition of % fatty acid in oils used for copper surfactant preparation

Oil used	% Fatty Acids					
	16:0	18:0	18:1	18:2	18:3	Other Acids
Sesame Oil	8	4	45	41	-	-
Soyabean Oil	12	4	24	51	9	-

Table 2: Physical characteristic of copper surfactants derived from unfried and fried oils

Copper Surfactant	Color	M.P./^{0}C	%Yield	% Metal		S.V.	S.E.	Av. Mol. Wt.
				Found	Calculated			
CSe	Green	104	71	10.05	9.870	191.70	292.64	646.78
CSo	Green	108	70	10.80	10.303	194.90	287.83	637.17
CSe_{15}	Green	97	73	13.01	13.174	252.40	222.26	506.02
CSo_{15}	Green	101	72	15.98	16.116	336.60	166.66	394.82
CSe_{60}	Green	80	75	20.15	20.352	448.00	125.22	311.94
CSo_{60}	Green	70	77	10.52	10.672	210.38	266.66	594.82

The plots of densityρ against the surfactant concentration c for CSo surfactants are characterized by an intersection of a convex curve and a straight line (Figure 1) at a definite surfactant concentration which corresponds to the critical micelle concentration (CMC) of the surfactant whereas the plots of densityρ with the surfactant concentration c for CSe surfactant are characterized by an intersection of two straight lines at a point that corresponds to the CMC of the surfactant.

At the CMC there is a sudden change in the aggregation of the surfactant molecules. The plots of densityρ against the surfactant concentration c has been extrapolated to zero surfactant concentration. It is found that the extrapolated values of the density are in fairly close agreement with the density of solvent. It may be suggested that below CMC the surfactant molecules do not aggregate adequately whereas at and above CMC, suddenly there is marked enhancement in the degree of aggregation of the surfactant molecules in benzene [12-13].

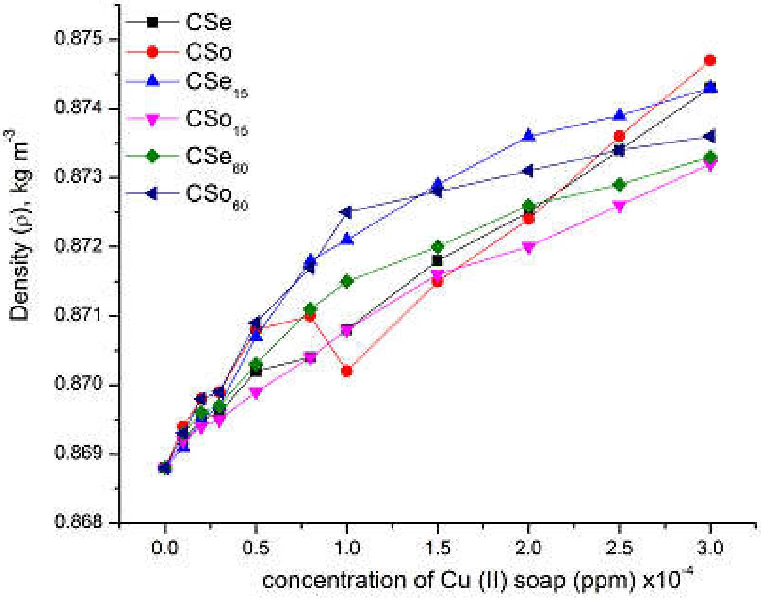

Figure 1: Plots of ρ v/s c of copper surfactants (derived from fried and unfried oils) solution in benzene.

The density and CMC of copper surfactants derived from unfried oils follows the order:

CSo>CSe

The above trend clearly demonstrates that association of surfactant molecules do not occurs at lower concentration in both surfactants in benzene [14]. The density of copper surfactants derived from fried oils for 15 and 60 minutes' increases with increase in the surfactant concentration. The plots of the densityρ with the surfactant concentration are characterized by an intersection of two straight lines at a definite surfactant concentration which corresponds to the CMC of the surfactant. From the values of CMC it is observed that values of CMC of copper surfactants derived from fried oils for 15 and 60 minutes are higher than the values of CMC of copper surfactants of unfried oils. It is also found that the values of CMC follow the order:

CSo_{15} >CSe_{15}
CSe_{60}> CSo_{60}

This may be attributed to the fact that there is decrease in CMC values with the increase of the average molecular weight of the surfactant. It is also known that the CMC decrease by about one-third per methylene group in aqueous solution but this change is smaller in non -aqueous solutions because the energy required to transfer a methylene group from micelle to bulk is small as intermolecular forces are stronger in polar solvents like water and methanol whereas intermolecular forces are weaker in non- polar solvents like benzene [15]. The values of density follow the order:

$CSe_{15} > CSo_{15}$

From comparison of copper surfactants derived from differently fried oils, the values of CMC follow the order:

$CSe_{60} > CSe_{15} > CSe$
$CSo_{15} > CSo_{60} > CSo$

From this trend, it is obvious that in copper surfactant of sesame oil, CMC increases with heating the oil but in copper surfactants of soyabean oil, CMC increases first, but after more heating, CMC decreases. That means, the effect of heating in soyabean oil is different from other oils.

3.2 M. V.:

The M.V. of copper surfactants (derived from unfried oils) solution is shows in Fig-2. From the data, it is clear that the M.V. increases with the increase in concentration of surfactant solution. The plots of the M. V. against the surfactant concentration c are characterized by an intersection of two straight lines at a definite surfactant concentration corresponding to CMC of the surfactant. At the CMC, hydrocarbon chain structure of complex molecules allows extensive contact between adjacent chains, possibly accompanied by changes in the vibrational and rotational degree of freedom of methylene group [16].

The M.V. of copper surfactants (derived from unfried oils) solutions follows the order:

$CSe \approx CSo$

The effect of the surfactant concentration on the M.V. of copper surfactants (derived from fried oils for 15 and 60 minutes) solutions in benzene is similar to that of above surfactants. The values of CMC follow the order:

$CSo_{15} > CSe_{15}$
$CSe_{60} > CSo_{60}$

The order of CMC is follows that there is decrease in CMC with the increase in M.W. of the surfactants. The values of M.V. follow the order:

$CSe_{15} > CSo_{15}$
$CSo_{60} > CSe_{60}$

The values of M.V. derived from differently fried oils are in the order:

$CSe > CSe_{15} > CSe_{60}$
$CSo > CSo_{15} < CSo_{60}$

From these trends, it is apparent that in copper surfactants derived from sesame oils, molar volume decreases with the increase of time of heating of the but in copper surfactants of soyabean oil, molar volume decreases first but after more heating molar volume increases. That means, the effect of heating in soyabean oil is different from other oils [17].

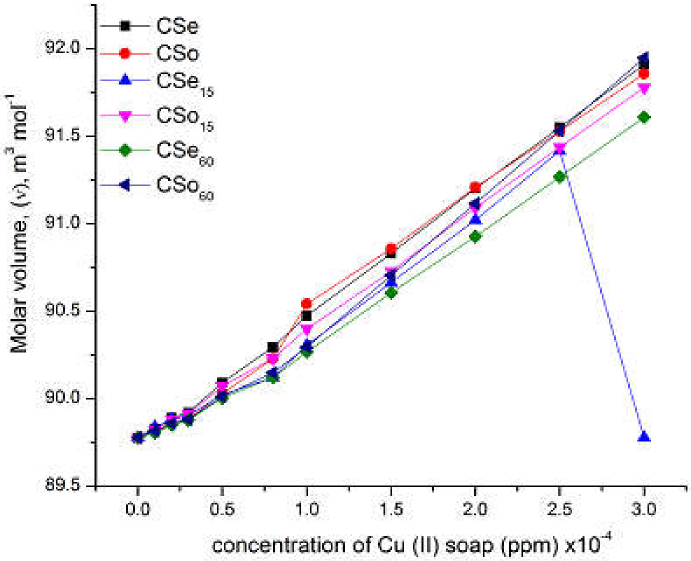

Figure 2: Plots of M.V. v/s c of copper surfactants (derived from fried and unfried oils) solution in benzene.

3.3 A. M.V.:

The micellar features of copper surfactants have been interpreted in terms of apparent molar volume. The A.M.V. (ϕ_v) of copper surfactants (derived from unfried oils) solutions in benzene, calculated by using equation 3, are shows in Fig.-3. The values of A.M.V. of all surfactants decrease sharply below the CMC with the increase in surfactant concentration. Above CMC, the values of ϕ_v are nearly constant for CSe solutions but the values of ϕ_v increase for CSo solutions (Table3).

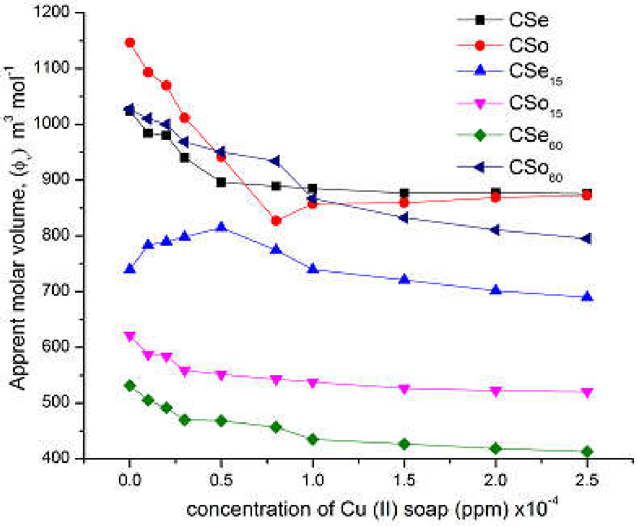

Figure 3: Plots of A.M.V. v/s c of copper surfactants (derived from fried and unfried oils) solution in benzene.

Table 3: Values of CMC of copper surfactants derived from fried and unfried oils

Parameter	**CMC**					
	CSe	**CSo**	**CSe$_{15}$**	**CSo$_{15}$**	**CSe$_{60}$**	**CSo$_{60}$**
Density	0.0117	0.0155	0.0145	0.0192	0.0242	0.0147
M. V.	0.0115	0.0155	0.0147	0.0190	0.0240	0.0150
A. M. V.	0.0116	0.0158	0.0148	0.0190	0.0237	0.0144

The data have been interpreted by using Masson's equation [18].

$$\phi_v = \phi_v^0 + S\sqrt{c} \qquad (4)$$

The plots of ϕ_v versus √c are shown in Figure 4. The plots are characterized by an intersection of two straight lines at a definite surfactant concentration, which corresponds to the CMC of the surfactant. Of course, this is the maximum concentration of molecular dispersion at which there is balancing of internal forces causing the formation of micelles.

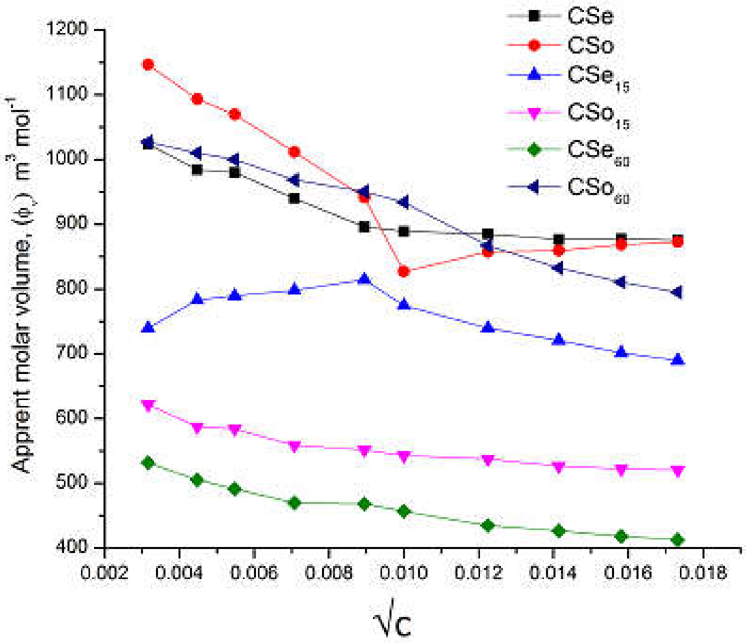

Figure 4: Plots of A.M.V. v/s $\sqrt{c}$ of copper surfactants (derived from fried and unfried oils) solution in benzene.

The CMC obtained from the plots of ϕ_v v/s √c is almost same as obtained from density and molar volume. At the CMC, the environment such as micellar clustering, solvation of ions, diminutions of mobility are entirely different below and above CMC. The Masson equation fits well in both below and above CMC. The values of limiting apparent molar volume for these surfactants obtained from extrapolation of ϕ_v v/s √c plots to c→0 Naturally, there are two values of ϕ^o_v referred as ϕ^o_{v1} (below CMC) and ϕ^o_{v2} (above CMC) as the Masson equation is equally applicable to the two intersecting straight lines. The change in the value of ϕ^o_v and S_v below and above CMC suggests that there is a phenomenal change in the micellar agglomeration below and above CMC [19]. Since ϕ^o_v has been regarded as a measure of solute-solvent interaction by earlier workers, therefore it is reasonable that greater magnitude of ϕ^{o}_{v} may be regarded as a measure of greater surfactant-solvent interaction [20].It is obvious from for copper surfactants derived from unfried oils $\phi^{o}_{v1}>\phi^{o}_{v2}$. That means, for all the surfactant solutions, surfactant- solvent interaction is more pronounced in dilute solutions [21-22]. This may be ascribed due to the favorable interaction between surfactant and solvent molecules at the premicellar concentration. Above CMC, surfactant-surfactant interaction becomes more prominent than surfactant- solvent interaction (Table4).

Table 4: Calculated parameters of Masson's equation for copper surfactants derived from fried and unfried oils

Name of the copper surfactant	ϕ^{00}_{v1}	ϕ^{0}_{v2}	S_{v1}	S_{v2}
CSe	1090	917	-1.80	-0.0194
CSo	1293	810	-3.37	+0.2686
CSe_{15}	747	955	+0.2309	-0.4452
CSo_{15}	635	565	-0.2586	-0.0699
CSe_{60}	582	490	-0.3640	-0.1051
CSo_{60}	1185	970	-0.9823	-0.3346

The results suggest that the surfactant- solvent interaction below CMC (ϕ^{o}_{v1}) follow the order:

CSo>CSe

It reveals that the surfactant-solvent interaction increases with the increase in the shorter fatty acid content in the oil. The parameter S_v in Masson equation represents the limiting apparent slope and indicates the existence of solute-solute interactions· It is reasonable to determine two values of parameter S_v below and above CMC designated as S_{v1} and S_{v2} respectively. It reveals the values of S_{v1} follow the order:

CSe>CSo

A perusal of the results also shows that as the concentration of the surfactant increases, solute-solute interaction become greater due to micellization process. The data also suggests that there is a greater increase in the interaction for surfactants of lower molecular weight resulting greater difference in S_{v1} and S_{v2} values [23]. The CMC values are in good agreement with the values of CMC obtained from other physical properties and follow the order:

CSo>CSe

Further on comparing the CMC results, it can be concluded that the value of the CMC of the surfactant solution in hydrocarbon are affected by the increase in the higher chain length content present in the oil composition and also with the unsaturation in the side chain [24]. The values of A.M.V. of CSo_{15} decreases with the increase in surfactant concentration and after CMC, the values of ϕ_v are almost constant. In CSe_{15}, the values of ϕ_v increases with the increase in concentration and in higher concentration, the value of ϕ_v decreases. The plots of ϕ_v v/s $\sqrt{c}$ show an intersection of two straight lines at definite surfactant concentration which corresponds to the CMC of surfactant. Here too, it is not unreasonable to stipulate two values of parameters below and above CMC designated as ϕ^{0}_{v1}, S_{v1} and ϕ^{0}_{v2}, S_{v2}. Moreover, the trends in the parameters are not different than those of copper surfactants of unfried oils. It is obvious from that

$\phi^{0}_{v1} > \phi^{0}_{v2}$ and $S_{v2} > S_{v1}$

The change in the values of ϕ_v and S_v below and above CMC suggests that there is a phenomenal change in the micellar clustering. The data suggest that the surfactant- solvent interaction below CMC (ϕ^o_{v1}) follows the order:

$CSe_{15} > CSo_{15}$
$CSo_{60} > CSe_{60}$

This trend reveals that there is decrease in surfactant-solvent interaction (below CMC) with the decrease in Av. M.W. of copper surfactant [25-26]. It is obvious that the values of S_{v1} follow the order:
$CSe_{15} > CSo_{15}$

The CMC values obtained from ϕ_v v/s √c plots are follow the order:

$CSo_{15} > CSe_{15}$
$CSe_{60} > CSo_{60}$

From comparison of the copper surfactants of differently fried oils, it is observed that the apparent molar volume follows the order:

$CSe > CSe_{15} > CSe_{60}$
$CSo > CSo_{15} < CSo_{60}$

These results reveal that the values of A.M.V. decrease with the decrease in Av. M.W. of the copper surfactants.

3.4 A General profile of Masson's equation with reference to copper surfactants -

A perusal of the results shows that in all copper surfactants, the solute-solvent interaction decreases with the increase in the concentration of surfactant solution i.e.
$\phi^0_{v1} > \phi^o_{v2}$

ϕ^0_{v1}, the solute–solvent interaction, is found to be higher in copper surfactants of unfried oils and decreases as the time increase for treating at higher temperature. Further the solute–solvent interaction ϕ^o_{v1} decreases for CSe_{60} but increases for CSo_{60}. The values of ϕ^o_{v1} for copper surfactants derived from differently fried oils follow the order:

$CSe > CSe_{15} > CSe_{60}$ (5)
$CSo > CSo_{15} < CSo_{60}$ (6)

These results show that with the decrease in the Av. M.W., the solute- solvent interaction decreases below CMC [27-28]. It is evident that difference between the values of limiting A.M.V. (below CMC) of copper surfactants of unfried oils and copper surfactants of fried oils for 15 minutes follows the order:

CSo>CSe

From the above observations, it is clear that maximum change in surfactant- solvent interaction takes place in surfactant derived from Soyabean oil. This may be due to higher unsaturation present in the oil which leads to maximum changes in the chains of the surfactant segment (higher fatty acid present in oil) during frying at high temperature for longer period [29]. These findings may be understand on the basis of that soyabean (84% unsaturation) and sesame (86% unsaturation) show maximum denaturing due to autoxidation and other chemical reactions during frying process [30-31].

A perusal of the order (5) and (6) show that interaction between solvent and solute molecules increase for CSo_{60}. This may be attributed to the fact that as the average molecular weight increases due to thermal abuse and other chemical reactions, interaction also increases [32-35]. The observations suggest that there is a marked difference between the molecular weights of the CSo_{60} and CSo_{15}, hence there is larger change in various interactions.

4. Conclusion

The paper attempt to prepare some new surfactants from abused edible oils due to heating and to understand their miceller features in non -aqueous, non- polar solvent to better understand the solution behavior and various interactions present in the system for their biological and industrial applications.

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