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Volumetric Studies of Copper Soap Derived from Treated and Untreated Oils in Benzene at 298.15 K

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Abstract

The density, molar volume, and apparent molar volume of Cu (II) soap derived from mustard and groundnut oils treated and untreated in non-aqueous solvent benzene has been determined at constant temperature 298.15 K. The results were used to determine the critical micelle concentration (CMC), the effect of chain length of the soap molecule on CMC and soap-solvent interactions. The conclusions with regard to solute-solute and solute- solvent interaction have been discussed in terms of well-known Masson's. This vital information plays an important role in various industrial and biological applications.

Keywords: Cu (II), mustard and ground nut oils, soap- solvent interaction, CMC, density molar volume, and apparent molar volume

1. INTRODUCTION

Copper soaps play significant role in biological activities and have sufficient industrial and analytical applications [1-2]. Coherence of biological activities of copper soaps derived from various edible and non-edible oils and their complexes in different methanol and benzene solvents stimulated our interest to extend the studies in their cumulative form. Studies have also been done by various workers who studied the molar volume and other parameters of various soaps[3-4].The phenomenon of micellization of surfactants in the bulk phase, as well as their ability to be accumulated at an interface are of immense theoretical, applied and biological interests as indicated by large number of publication of papers and reviews in last three decades [5-6].The present work deals with the study of density,molar volume, and apparent molar volume of copper soap derived from mustard and groundnut oilsin a binary system. The density measurements have been used to determine the critical micelle concentrations (CMC) of Cu (II) soaps in non-aqueous media and apparent molar volume data have been employed to obtain information about solute-solute, solvent-solvent interactions in solutions [7-8]. The effect of soap concentration on density and apparent molar volume of the solution in non-polar solvent has been discussed in terms of Masson's equation [9-10].

2. EXPERIMENTAL

All the chemicals used were of LR/AR grade. Copper soap was prepared by refluxing the mustard and groundnut oils (extracted from kernels and purified) with ethyl alcohol and 2N KOH solutions for 3-4 h (Direct Metathesis), copper soap so obtained was then washed with warm water and 10% alcohol at 50°C and recrystallized using hot benzene. Molecular weight of copper soap was determined from saponification value [11]. The copper soaps are abbreviated as copper – mustard (CM), copper – groundnut (CG):

2.1 Measurement of Density:

Ostwald's modification of Sprengel'spyknometer with a volume of about 10 ml was used for measuring the density of the soap solution in the thermo stated bath at 298.15 K. The density of the solutions was calculated by the following relationship.The possible error in the density was estimated to be less than ± 1. The density ρwere reproducible to within ± 0.020 kg m^{-3}

$$\rho = \frac{w}{w0} \qquad (1)$$

Where w and w_0 are the weights (same volume) of solution and water respectively.

2.2 Evaluation of Molar Volume:

The molar volume of the soap solution V has been calculated by the relationship;

$$\nabla = M_1X_1 + M_2X_2 + M_3X_3/\rho \quad (2)$$

Where X_1 is the mole fraction of the soap of molecular weight M_1 whereas X_2 is the mole fraction of solvent of molecular weight M_2 and 'ρ' stands for density of the solution. The molar volume∇were reproducible to within ± 0.020 m^{3}mol^{-1}

2.3 Evaluation of Apparent Molar Volume:

The apparent molar volume has been calculated from the density data using the following equation;

$$\phi v = \frac{M}{\rho^0} + \frac{1000(\rho^0 - \rho)}{c.\rho_0} \quad (3)$$

Where 'ρ_0' represents the density of the solvent,'ρ' is the density of the soap solution, 'M' is the molecular weight of the soap and 'c' is the concentration of solution in mol L^{-1}.The AMV ϕvwere reproducible to within ± 0.020 m^{3}mol^{-1}

3. RESULTS AND DISCUSSION

3.1 Density

The density of copper soaps of untreated oils first increases with increase in soap concentration but there is a slight decrease followed by linear increase in CG solutions. The density of CM solutions increases regularly. It is due to the increase in the shorter fatty acid content in the corresponding oil composition (Table1-2). The plots of density 'ρ' against the soap concentration 'c' for CG soaps are characterized by an intersection of a convex curve and a straight line (Fig. 1) at a definite soap concentration which corresponds to the critical micelle concentration (CMC) of the soap whereas the plots of density 'ρ' with the soap concentration 'c' for CM soap are characterized by an intersection of two straight lines at a point that corresponds to the CMC of the soap (Fig. 1). At the CMC there is a sudden change in the aggregation of the soap molecules. The plots of density 'ρ' against the soap concentration 'c' has been extrapolated to zero soap concentration. It is found that the extrapolated values of the density are in fairly close agreement with the density of solvent (benzene). It is therefore concluded that the soap molecules do not aggregate adequately below CMC whereas at this definite soap concentration there is marked enhancement in the degree of aggregation of the soap molecules in benzene [12].

Table 1: Fatty Acid Composition Of Oils Used For Copper Soap Synthesis

Name of oil	**% Fatty Acids**					
	16:0	**18:0**	**18:1**	**18:2**	**18:3**	**Other Acids**
Groundnut Oil	10	4	61	18	-	C_{20}-C_{24} 7%
Mustard Oil	2	1	25	18	10	C_{20}-C_{22} 41%

Table 2: Aanalytical and physical data of copper soaps derived from Untreated and treated oils

Name of Copper Soap	**Color**	**Melting point (in ^{0}C)**	**Yield %**	**Metal %**		**S.V.**	**S.E.**	**Average Mol. Wt.**
				Found	**Calculated**			
CG	Green	98	70	10.15	9.724	188.70	297.29	656.08
CM	Green	92	72	9.58	9.110	175.80	319.10	699.72
CG_{15}	Green	105	75	11.90	11.936	238.40	235.31	532.12
CM_{15}	Green	84	74	12.48	12.549	266.47	210.53	482.56
CG_{60}	Green	68	76	13.59	13.744	280.00	200.35	462.20
CM_{60}	Green	61	78	12.96	13.138	260.00	215.76	493.03

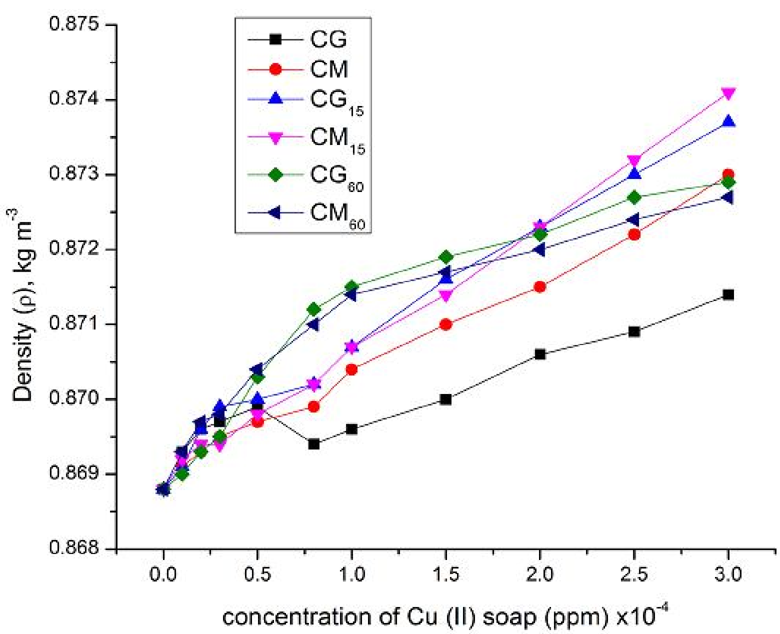

Figure 1: Plot of density v/s conc. Of copper soap (derived from treated and untreated oils) solutions in benzene

The density and CMC of copper soaps derived from untreated oils follows the order:

CG >CM

The above trend clearly demonstrates that association of soap molecules occurs at lower concentration in mustard and groundnut soaps in benzene. The density of copper soaps derived from treated oils for 15 and 60 minutes increases with increase in the soap concentration (Table-3). The plots of the density 'ρ' with the soap concentration are characterized by an intersection of two straight lines at a definite soap concentration which corresponds to the CMC of the soap (Fig-1).From the values of CMC it is observed that values of CMC of copper soaps derived from treated oils for 15 and 60 minutes are higher than the values of CMC of copper soaps of untreated oils [14].

Table 3: Values of Density of Copper Soap (Derived From Treated and Untreated Oils) Solutions in Benzene

c (ppm) x10^{-4}	**Density**					
	CG	**CM**	**CG_{15}**	**CM_{15}**	CG_{60}	CM_{60}
0.0	0.8688	0.8688	0.8688	0.8688	0.8688	0.8688
0.1	0.8693	0.8691	0.8691	0.8692	0.8690	0.8693
0.2	0.8696	0.8693	0.8696	0.8694	0.8693	0.8697
0.3	0.8697	0.8695	0.8699	0.8694	0.8695	0.8698
0.5	0.8699	0.8697	0.8700	0.8698	0.8703	0.8704
0.8	0.8694	0.8699	0.8702	0.8702	0.8712	0.8710
1.0	0.8696	0.8704	0.8707	0.8707	0.8715	0.8714
1.5	0.8700	0.8710	0.8716	0.8714	0.8719	0.8717
2.0	0.8706	0.8715	0.8723	0.8723	0.8722	0.8720
2.5	0.8709	0.8722	0.8730	0.8732	0.8727	0.8724
3.0	0.8714	0.8730	0.8737	0.8741	0.8729	0.8727

It is also found that the values of CMC follow the order:

$CM_{15} > CG_{15}$
$CG_{60} > CM_{60}$

This observation is in agreement with the fact that there is decrease in CMC values with the increase of the average molecular weight of the soap. 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 [16]. The values of density follow the order:

$CG_{15} > CM_{15}$

From comparison of copper soaps derived from differently treated oils, the values of CMC follow the order:

$CG_{60} > CG_{15} > CG$

$CM_{60} \approx CM_{15} > CM$

From this trend, it is obvious that in copper soap of groundnut, mustard oils, CMC increases with heating the oil.

3.2 Molar Volume:

The molar volume V of copper soaps (derived from untreated oils) solution is recorded in Table-4. From the data, it is clear that the molar volume increases with the increase in concentration of soap solution. The plots of the molar volume V against the soap concentration c are characterized by an intersection of two straight lines at a definite soap concentration corresponding to CMC of the soap (Fig-2). 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. The values of CMC, so determined, are in good agreement with the values obtained from the density measurements [17].

Table 4: Values of Molar Volume of Copper Soap (Derived From Treated and Untreated Oils) Solutions in Benzene

c (ppm) x10^{-4}	Molar Volume					
	CG	**CM**	**CG$_{15}$**	**CM$_{15}$**	CG$_{60}$	CM$_{60}$
0.0	89.778	89.778	89.778	89.778	89.778	89.778
0.1	89.818	89.843	89.836	89.823	89.844	89.818
0.2	89.881	89.912	89.878	89.895	89.901	89.865
0.3	89.912	89.940	89.886	89.931	89.918	89.893
0.5	90.114	90.143	90.092	90.112	90.055	90.049
0.8	90.394	90.348	90.296	90.284	90.177	90.205
1.0	90.606	90.529	90.467	90.449	90.352	90.381
1.5	91.021	90.928	90.811	90.802	90.737	90.779
2.0	91.407	91.322	91.170	91.139	91.134	91.181
2.5	91.829	91.702	91.534	91.471	91.510	91.570
3.0	89.778	89.778	91.894	91.803	91.912	91.963

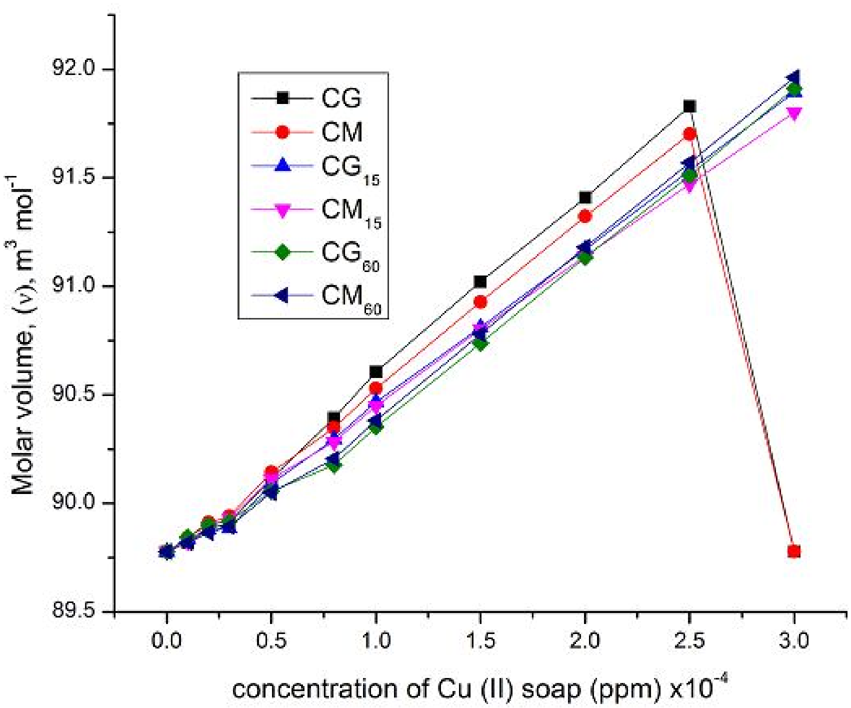

Figure 2: Plot of molar volume v/s conc. Of copper soap (derived from treated and untreated oils) solutions in benzene

The molar volume of copper soaps (derived from untreated oils) solutions follows the order:

CG ≈CM

The effect of the soap concentration on the molar volume of copper soaps (derived from treated oils for 15 and 60 minutes) solutions in benzene is similar to that of above soaps [18]. The values of CMC follow the order:

$CM_{15} > CG_{15}$
$CG_{60} > CM_{60}$

The order of CMC is according to the fact that there is decrease in CMC with the increase in molecular weight of the soaps [19]. The values of molar volume follow the order:

$CG_{15} > CM_{15}$

$CM_{60} > CG_{60}$

The values of molar volume derived from differently treated oils are in the order:

$CG > CG_{15} > CG_{60}$

$CM > CM_{15} > CM_{60}$

From these trends, it is apparent that in copper soaps derived from groundnut and mustard oils, molar volume decreases with the increase of time of heating.

3.3 Apparent Molar Volume:

It has been propounded that any physical property, which is able to conveniently demonstrate the deviation in its normal trend, can be used to determine the CMC of the surfactants. Keeping this in mind, it has been thought worthwhile to interpret the micellar features of copper soaps in terms of apparent molar volume. The apparent molar volume (ϕ_v) of copper soaps (derived from untreated oils) solutions in benzene, calculated by using equation 3, is recorded in Table-5. The values of apparent molar volume of all soaps decrease sharply below the CMC with the increase in soap concentration. Above CMC, the values of ϕ_v are nearly constant for CG and CM. The data have been analyzed in terms of Masson equation [20].

$$\phi v = \phi^0 v + S\sqrt{C} \quad (4)$$

The plots of ϕ_v versus $\sqrt{c}$ are shown in Fig-3. The plots are characterized by an intersection of two straight lines at a definite soap concentration, which corresponds to the CMC of the soap. Of course, this is the maximum concentration of molecular dispersion at which there is balancing of internal forces causing the formation of micelles. The CMC obtained from the plots of ϕ_v v/s $\sqrt{c}$ 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 soaps obtained from extrapolation of ϕ_v v/s $\sqrt{c}$ plots to $c \rightarrow 0$ 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 [21-22].

Table 5: Values of Apparent Molar Volume of Copper Soap (Derived From Treated and Untreated Oils) Solutions in Benzene

c (ppm) x10^{-4}	Apparent Molar Volume					
	CG	**CM**	**CG_{15}**	**CM_{15}**	CG_{60}	CM_{60}
0.0	1119.89	1014.20	789.91	776.69	635.00	822.79
0.1	1037.21	994.74	838.89	704.57	656.72	805.77
0.2	1022.18	1010.64	879.31	690.15	684.91	792.15
0.3	922.97	946.72	761.74	660.19	688.10	746.75
0.5	814.73	923.12	724.18	656.12	698.38	730.86
0.8	809.47	929.77	725.00	657.97	676.40	712.14
1.0	808.70	916.70	724.18	650.94	642.37	676.01
1.5	818.12	911.76	719.18	651.31	621.89	656.81
2.0	812.93	912.62	714.47	652.20	613.44	647.79
2.5	816.74	913.49	711.94	652.61	603.37	640.83
3.0	1119.89	1014.20	789.91	776.69	635.00	822.79

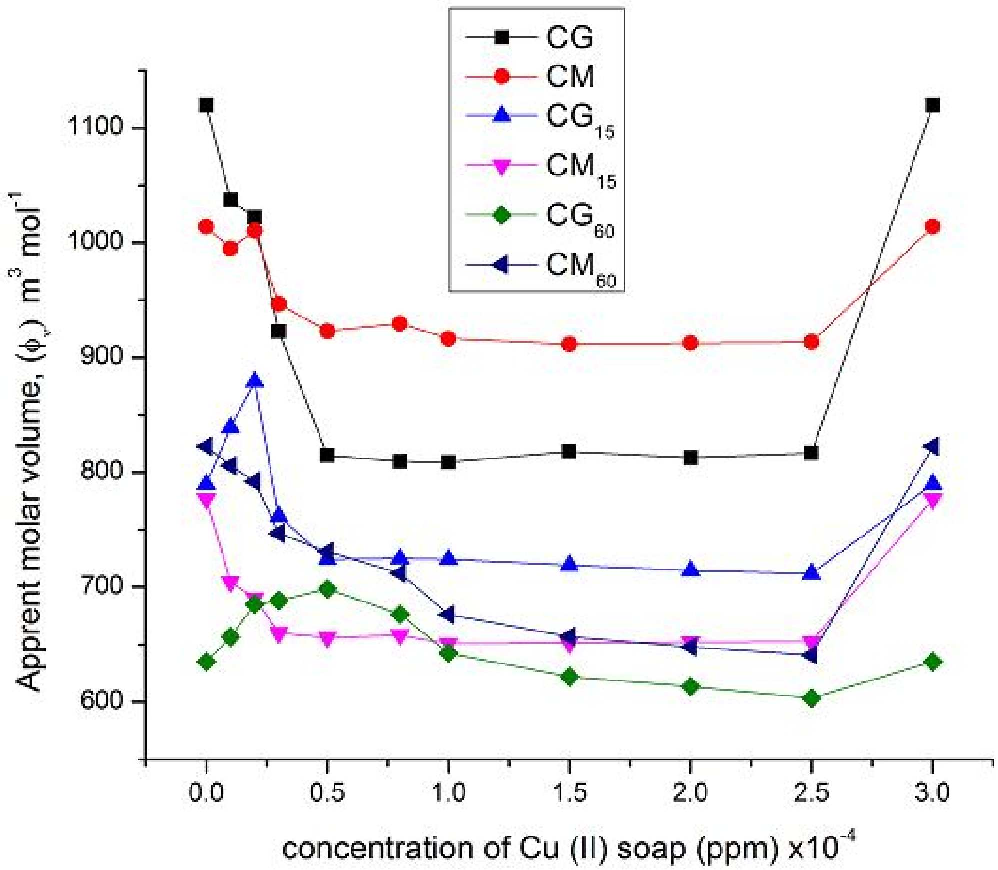

Figure 3: Plot of Apparent Molar Volume v/s conc. of Copper Soap (Derived From Treated And Untreated Oils) Solutions in Benzene

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 soap-solvent interaction [23].

It is obvious that for copper soaps derived from untreated oils $\phi^o_{v1} > \phi^o_{v2}$. That means, for all the soap solutions, soap- solvent interaction is more pronounced in dilute solutions. This may be ascribed due to the favorable interaction between soap and solvent molecules at the premicellar concentration. Above CMC, soap-soap interaction becomes more prominent than soap- solvent interaction. The results suggest that the soap- solvent interaction below CMC (ϕ^o_{v1}) follow the order:

CG > CM

The compositions of oils reveal that the soap-solvent interaction increases with the increase in the shorter fatty acid content in the oil. A perusal of Table-6 shows that there is a greater decrease of solute-solvent interaction in copper soaps of groundnut oil as compared to copper soaps of mustard oil suggesting that as the molecular weight decreases, the solvent-solute interaction decreases with the increase in the solute concentration in the solution, the difference between ϕ^o_{v1} and ϕ^o_{v2} thus gets enhanced.

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 is obvious from Table-6 that the values of S_{v1} follow the order:

CM > CG

Table 6: Computed Parameters of Masson's Equation for Copper Soaps Derived from Treated and Untreated Oils.

Name of the soap	ϕ^o_{v1}	ϕ^o_{v2}	S_{v1}	S_{v2}
CG	1278	814	-4.51	+0.0260
CM	1070	931	-1.40	-0.0875
CG_{15}	962	737	-0.7954	-0.0437
CM_{15}	740	665	-0.2679	-0.0175
CG_{60}	615	805	+0.2679	-0.3541
CM_{60}	882	785	-0.5317	-0.2493

The data reveals that within the micellar palisade layer, solute- solute interactions are greater for copper soap of mustard oil due to presence of higher fatty acid (C_{20}- C_{24}) content in oil composition. A perusal of the results also shows that as the concentration of the soap increases, solute-solute interaction become greater due to micellization process. The data also suggests that there is a greater increase in the interaction for soaps of lower molecular weight resulting greater difference in S_{v1} and S_{v2} values.

The CMC values (Table-7) are in good agreement with the values of CMC obtained from other physical properties and follow the order:

CG > CM

Table 7: Values of CMC of Copper Soaps Derived from treated and untreated oils.

Parameter	**Name of the soap (CMC)**					
	CG	**CM**	**CG_{15}**	**CM_{15}**	**CG_{60}**	**CM_{60}**
Density	0.0115	0.0105	0.0140	0.0155	0.0162	0.0155
Molar volume	0.0110	0.0105	0.0140	0.0155	0.0160	0.0155
Apparent molar volume	0.0113	0.0108	0.0139	0.0149	0.0159	0.0154

This is in agreement that there is decrease in CMC value with the increase in the average molecular weight of the soap. Further on comparing the CMC results, it can be concluded that the value of the CMC of the soap 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.

The values of apparent molar volume (ϕ_v) enumerated with the help of equation (3) for copper soaps derived from treated oils for 15 and 60 minutes are recorded in Table-5.

The values of apparent molar volume of CG_{15}, and CM_{15} decreases with the increase in soap concentration and after CMC, the values of ϕ_v are almost constant.

The plots of ϕ_v v/s$\sqrt{C}$ show an intersection of two straight lines at definite soap concentration which corresponds to the CMC of soap (Fig-3). Masson equation is applicable to both straight lines. Here too, it is not unreasonable to stipulate two values of parameters below and above CMC designated asϕ^o_{v1}, S_{v1} and ϕ^o_{v2}, S_{v2}. Moreover, the trends in the parameters are not different than those of Copper soaps of untreated oils. It is obvious that [24]

$\phi^o_{v1} > \phi^o_{v2}$ 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 soap- solvent interaction below CMC (ϕ^o_{v1}) follows the order:

$CG_{15} > CM_{15}$
$CM_{60} > CG_{60}$

This trend reveals that there is decrease in soap-solvent interaction (below CMC) with the decrease in average molecular weight of Copper soap. It is obvious from Table 5 that the values of S_{v1} follow the order:

$CM_{15} > CG_{15}$

The CMC values obtained from ϕ_v v/s$\sqrt{C}$ plots are follow the order:

$CM_{15} > CG_{15}$
$CG_{60} > CM_{60}$

This is in agreement with the values of CMC obtained from other physical properties and according to the fact that there is increase in value of CMC with the decrease in average molecular weight of the soap [25-26].

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

$$CG > CG_{15} > CG_{60}$$

$$CM > CM_{15} < CM_{60}$$

These results reveal that the values of apparent molar volume decrease with the decrease in average molecular weight of the copper soaps

4. A GENERAL PROFILE OF MASSON EQUATION WITH REFERENCE TO COPPER SOAPS

A perusal of the results shows that in all copper soaps, the solute-solvent interaction decreases with the increase in the concentration of soap solution i.e.

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

ϕ^0_{v1}, the solute–solvent interaction, is found to be higher in Copper soaps of untreated oils and decreases as the time increase for treating at higher temperature. Further the solute–solvent interaction ϕ^0_{v1}decreases for CG_{60}butincreases for CM_{60}. The values of $\phi^0_{v1}$for copper soaps derived from differently treated oils follow the order: [27]

$CG > CG_{15} > CG_{60}$ (1) $CM > CM_{15} < CM_{60}$ (2)

These results show that with the decrease in the average molecular weight, the solute- solvent interaction decreases below CMC. From Tables (5), it is evident that difference between the values of limiting apparent molar volume (below CMC) of copper soaps of untreated oils and copper soaps of treated oils for 15 minutes follows the order:[28]

CM > CG

A perusal of the order (1) and (2) show that interaction between solvent and solute molecules increase for CM_{60}. This may be attributed to the fact that as the average molecular weight increase due to thermal abuse and other chemical reactions, interaction also increases [29-30]

5. CONCLUSION

The present research work makes an attempt to prepare surface active compounds from metal and natural edible oils. It is found that the beneficial effects of the synthesized biologically active molecules, agrochemicals and pharmaceuticals are still open for further research. The current topic will not only strengthen relation between industries, private sectors and research laboratories on the focal theme of biology, physics, and environment but also will also play a significant role in forth coming scientific development.

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