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Volumetric and viscometric studies of copper soap urea complex derived from various vegetable oils in methanol-benzene mixture.

Abstract

The density and viscosity of Cu (II) soap urea complex derived from four edible oils (mustard, groundnut, sesame, soyabean) in non-aqueous solvents of varying composition has been determined at constant temperature. The results were used to determine the critical micelle concentration (CMC), soap-solvent interactions and the effect of chain length of the soap urea complex molecule on various parameters. The CMC values in higher volume % of methanol are higher those in higher volume % of benzene, with regards to chain length of the soap urea complex CMC value follow the order:
CSoU>CSeU> CGU > CMU
The conclusions with regard to soap- urea complex and soap- solvent interaction have been discussed in terms of well-known Masson's and Jones- Dole equations. This vital information plays an important role in various industrial and biological applications.

Keywords: Cu(II) soap urea complex, edible oils, soap- solvent interaction, CMC, density and viscosity.

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1. Introduction

Copper soaps have a tendency of complexion with the compounds containing donor atom like N, O, Cl and S etc. [1-4]. It is well known that compound containing N and S atoms play significant role in biological activities and have sufficient industrial and analytical applications [5-6]. Coherence of biological activities of copper soaps derived from various edible oils and their urea complexes in different methanol and benzene solvents stimulated our interest to extend the studies in their cumulative form. It is anticipated that it will generate a new hopes in various industrial and analytical applications. Many nitrogen and sulphur containing compounds were synthesized and investigated for a wide range of pharmacological and biological activities [7-8]. They have been used as tranquilizers, sedatives, anti-inflammatory agents, antiviral, anti-malarial fungicide, bactericide [9-12]. These compounds have found wide applications in many industrial uses and play an important role in the colloid chemical phenomena. The exact information on the nature and structure of these compounds are of great significance for explaining their characteristics under different conditions. 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 [13-15]. The present work deals with the study of viscosity of copper urea complex derived from various edible oils in a ternary system. The effect of soap urea complex concentration on viscosity of the solution in polar and Non- polar solvent has been discussed in terms of Jones- Dole equations [16-18].

2. Experimental

All chemicals used were of LR/AR grade. Oils were procured directly from the seeds of mustard, groundnut, sesame. Soyabean (Pure) oil was taken from the market of a reputed brand. The fatty acid composition of oils was confirmed by sending their methyl esters to RSIC, CDRI, Lucknow, U.P. India mentioned in Table 1. Copper soaps were prepared by refluxing the oils with 2N KOH solution and alcohol for about 3 h (direct metathesis). The excess of KOH was neutralized using 1N HCl. Saturated solution of copper sulphate was added to it. The copper soap obtained was filtered, washed with warm water followed by alcohol, dried at 50 ^{0}C and recrystallized with hot benzene. Copper soap-urea complexes were prepared by taking copper soap and urea in molar ratio (1:1). 0.005 moles of ligand (urea) was dissolved in 2-3 ml of ethyl alcohol and 0.005 moles of copper (II) soap derived from various edible oils were dissolved in 10-15 ml of benzene and solution of urea was added in it. The above reaction mixture was then heated for 1.5 h. Separated solid complex was filtered, washed with hot water and alcohol and dried in vacuum over fused calcium chloride. The dried sample was purified and recrystallized from hot benzene. These complexes are solids and green which are soluble in benzene and methanol-benzene solvent mixture but are insoluble in water, all the complexes are quite stable. The formation of copper soap and their urea complexes was confirmed by using IR, UV, NMR technique [19-20].The details of these complexes summarized in Table 2

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%
Sesame Oil	8	4	45	41	-	-

Table 2: Analytical and Physical Data of Copper (II) Soap Urea Complexes

Compound	Colour	M. P. (^{0}C)	Metal %		Mol. Wt.
			Found	Calculated	
CMU	Dark Green	72	8.46	8.35	759.72
CGU	Dark Green	85	9.31	8.86	716.08
CSeU	Dark Green	90	9.16	8.98	706.78
CSoU	Dark Green	100	9.31	9.10	697.17

2.1 Measurement of Density

Ostwald's modification of *Sprengel's* pyknometer 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:

$$\rho = \frac{w}{w_0} \qquad (1)$$

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

2.2 Evaluation of Apparent Molar Volume:

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

$$\phi_v = \frac{M}{\rho_0} + \frac{1000\ (\rho_0 - \rho)}{c.\rho_0} \qquad (2)$$

Where ρ_0 represents the density of the solvent, ρ is the density of the complex solution, M is the molecular weight of the complex and c is the concentration of solution.

2.3 Evaluation of Viscosity:

Ubbelohde type viscometer was used for measuring the viscosity of the solutions of varying concentrations of the soap

The viscosity of the soap solutions were calculated by the following relationship.

$$\frac{\eta_0}{\eta} = \frac{\rho_0 t_0}{\rho t} \qquad (3)$$

Where η_0, η, ρ_0, ρ, t_0 and t are the viscosity, density and time of flow for the known and unknown solutions respectively. The accuracy of the results was checked by determining the viscosity of known solutions and the agreement was found to be good and the difference was below 0.3%. All the measurements were made at a constant temperature 298.15K in a thermostat. The viscosity results are expressed in millipoise.

3. Results and Discussion

The complexes of Copper soaps synthesized with urea are abbreviated as in the following manner.

Complexes	Methanol-benzene	
	20%	40%
1. Copper Mustard soap- Urea Complex	CMU_{20}	CMU_{40}
2. Copper Groundnut soap- Urea Complex	CGU_{20}	CGU_{40}
3. Copper Sesame soap- Urea Complex	$CSeU_{20}$	$CSeU_{40}$
4. Copper Soyabean soap- Urea Complex	$CSoU_{20}$	$CSoU_{40}$

3.1 Density and apparent molar volume:

The density of copper urea soaps derived from four edible oils in 20% methanol-benzene mixture are higher than that of 40 % methanol-benzene mixture and follow the order (Table 3-6)

Table 3: Apparent Molar Volume, Viscosity and Specific Viscosity of Copper Mustard Urea Complex in Methanol Benzene Mixture at Constant Temperature 303+ 0.1 K

Complex Conc. g mol L^{-1}	A.M.V. (ϕ_v)		Viscosity (η)		Specific Viscosity (η_{sp})	
	20 %	40 %	20 %	40 %	20 %	40 %
0.0003	-375182.9	-368172	6.4952	6.5456	0.13553	0.1173
0.0004	-283496.8	-278234.2	6.5527	6.5767	0.14557	0.1226
0.0006	-190451	-186548.1	6.6109	6.6327	0.15575	0.1322
0.0008	-143928.1	-140996.4	6.6692	6.6773	0.16595	0.1398
0.0010	-113217.1	-113199.2	6.7108	6.7141	0.17322	0.1461
0.0011	-103373.9	-100813.1	6.8184	6.7566	0.19203	0.1533
0.0012	-95171.3	-92725.2	6.8741	6.7977	0.20176	0.1603
0.0013	-88230.6	-85881.7	6.9167	6.8389	0.20922	0.1674
0.0014	-82281.4	-80015.7	6.9594	6.8801	0.21668	0.1744
0.0016	-72395.4	-70192.2	7.0036	6.9009	0.22441	0.178
0.0018	-64835.9	-62681.2	7.0494	7.0152	0.23241	0.1975
0.0020	-58846.5	-56730.7	7.1874	7.0588	0.25654	0.2049

Table 4: Apparent Molar Volume, Viscosity and Specific Viscosity of Copper Groundnut Urea Complex In Methanol Benzene Mixture At Constant Temperature 303 + 0.1 K

Complex Conc. g mol L^{-1}	**A.M.V. (ϕ_v)**		**Viscosity (η)**		**Specific Viscosity (η_{sp})**	
	20 %	40 %	20 %	40 %	20 %	40 %
0.0003	-376010.8	-369389.4	6.4966	6.5605	0.13577	0.1198
0.0004	-284421.8	-278868.8	6.5235	6.6036	0.14048	0.1272
0.0006	-192055.9	-187377	6.5643	6.6738	0.14761	0.1392
0.0008	-145872.9	-141776.7	6.6261	6.7192	0.15841	0.1469
0.0010	-117463.8	-114300.1	6.6839	6.7896	0.16851	0.159
0.0011	-103848.6	-102772.2	6.7041	6.8207	0.17205	0.1643
0.0012	-95610.6	-92874.3	6.7727	6.8495	0.18404	0.1692
0.0013	-88550.4	-86112.9	6.799	6.8914	0.18864	0.1763
0.0014	-82582	-80483.9	6.8573	6.9477	0.19883	0.1859
0.0016	-72592	-70899.7	6.9399	7.0047	0.21327	0.1957
0.0018	-64886.7	-63445.4	7.0102	7.0874	0.22556	0.2098
0.0020	-58606	-57481.9	7.1053	7.1703	0.24219	0.2239

Table 5: Apparent Molar Volume, Viscosity And Specific Viscosity Of Copper Seasam Urea Complex In Methanol Benzene Mixture At Constant Temperature 303 + 0.1 K

Complex Conc. (g mol L^{-1})	**A.M.V. (ϕ_v)**		**Viscosity (η)**		**Specific Viscosity (η_{sp})**	
	20 %	40 %	20 %	40 %	20 %	40 %
0.0003	-376798.6	-369789	6.524	6.5739	0.14055	0.1221
0.0004	-284724	-279462.6	6.5502	6.6178	0.14514	0.1296
0.0006	-191872.4	-187582.3	6.5897	6.6872	0.15205	0.1415
0.0008	-145009.6	-141642.1	6.635	6.7313	0.15997	0.149
0.0010	-116891.9	-113961.5	6.6934	6.7874	0.17018	0.1586
0.0011	-105130.9	-104054.7	6.7256	6.8422	0.1758	0.1679
0.0012	-95330.1	-96284.7	6.7576	6.8879	0.1814	0.1757
0.0013	-88650.9	-85855	6.8154	6.902	0.1915	0.1781
0.0014	-82842.6	-80078.7	6.8725	6.9567	0.20149	0.1875
0.0016	-72748.5	-70546.5	6.9413	7.0265	0.21352	0.1994
0.0018	-64897.6	-63132.7	7.0363	7.122	0.23013	0.2157
0.0020	-58966.5	-57143.3	7.1359	7.2169	0.24753	0.2319

Table 6: Apparent Molar Volume, Viscosity and Specific Viscosity of Copper Soyabean Urea Complex in Methanol Benzene Mixture at Constant Temperature 303 + 0.1 K

Complex Conc. (g mol L^{-1})	A.M.V. (ϕ_v)		Viscosity (η)		Specific Viscosity (η_{sp})	
	20 %	40 %	20 %	40 %	20 %	40 %
0.0003	-377587	-372131	6.5513	6.5909	0.14533	0.125
0.0004	-285318	-280640	6.5776	6.6334	0.14993	0.1323
0.0006	-191884	-188954	6.6548	6.7178	0.16343	0.1467
0.0008	-145458	-142819	6.7153	6.7626	0.17401	0.1543
0.0010	-117136	-115138	6.7731	6.8202	0.18411	0.1642
0.0011	-107049	-105020	6.8035	6.8487	0.18942	0.169
0.0012	-98644	-96782	6.8469	6.9043	0.19702	0.1785
0.0013	-89290	-88377	6.8725	6.9354	0.20149	0.1838
0.0014	-83270	-81089	6.9152	6.9784	0.20896	0.1912
0.0016	-73343	-71651	6.9994	7.0506	0.22366	0.2035
0.0018	-65621	-64051	7.0705	7.1582	0.23611	0.2219
0.0020	-59502	-57912	7.2211	7.2781	0.26242	0.2423

CSoU>CSeU>CGU>CMU

This is due to the fact that the density increases with the increase in the shorter fatty acid content in the corresponding oil composition. The CMC of complexes obtain from density v/s concentration follows the order Fig 1-2

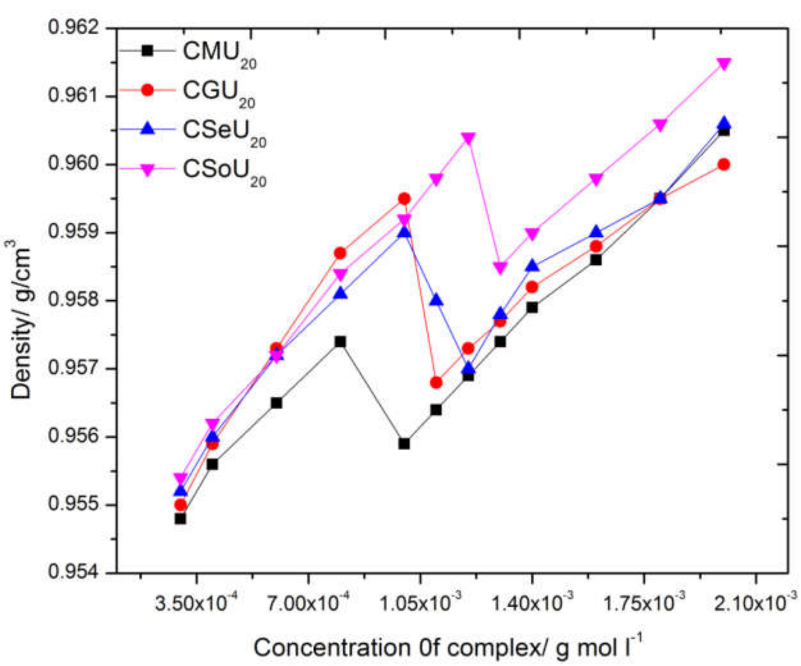

Figure 1: Plots of Density V/S Concentration of Copper Soap-Urea Complex In 20 % Methanol-Benzene Mixture

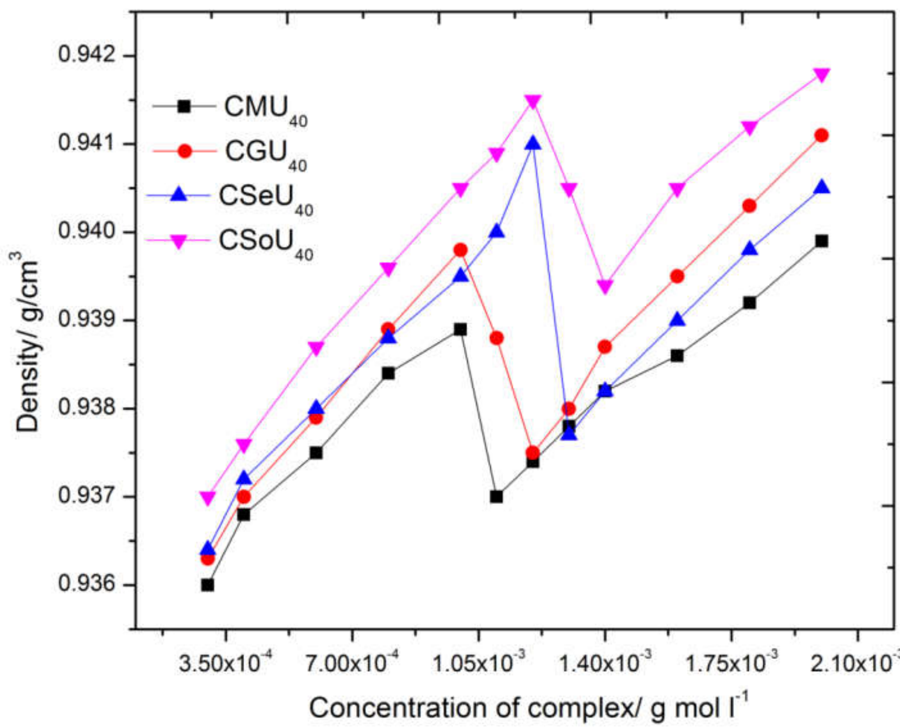

Figure 2: Plots of Density V/S Concentration of Copper Soap-Urea Complex In 40 % Methanol-Benzene Mixture

CSoU>CSeU>CGU>CMU

This observation is in the agreement with the fact that there is a decrease in CMC values with the increase of average molecular weight of the complex and presence of longer fatty acid content in oil composition. According to our observation of ϕ_v values for copper soap complexes in 20% and 40% methanol-benzene mixture are negative. Their negative values could be ascribed on the basis of group additively, which shows that different groups will display increasingly negative values according to their increasing polarity and hydrogen bonding capability.

The plots of ϕ_v v/s $\sqrt{C}$ are characterized by an intersection of curve and a straight line, in each case which corresponds to the CMC of soap complex. The value of apparent molar volume of CGU and CSoU increases sharply below CMC and gradually above CMC (Figures 3-4). The CMC obtain from the plots of ϕ_v v/s $\sqrt{C}$ also follow the same order as for density (Table 7). The CMC obtain in this study is also confirmed by other physical properties like viscosity and ultrasound velocity. This is in agreement with the observation that there is decrease in CMC value with the increase in the average molecular with of the soap complexes.

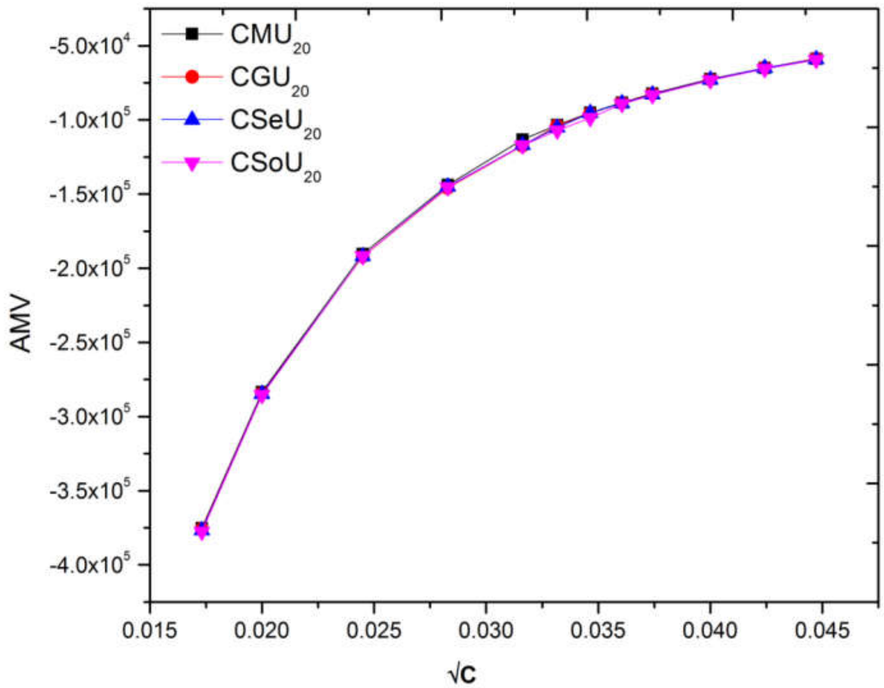

Figure 3: Plots of AMV Versus √C Of Copper Soap Urea Complex In 20% Methanol- Benzene Mixture

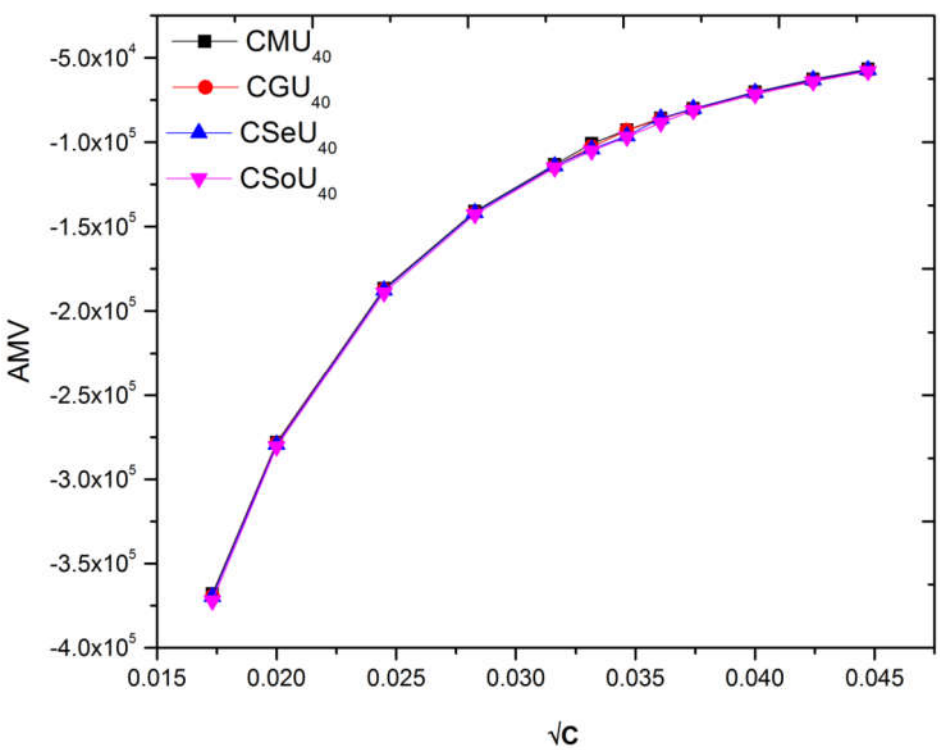

Figure 4: Plots of AMV Versus √C Of Copper Soap Urea Complex In 40 % Methanol- Benzene Mixture

Table 7: Values Of CMC of Copper Soap Urea Complexes In 20% And 40% Methanol-Benzene Mixture Obtained From Various Parameters

Plot	**Volume % of Methanol in Solvent mixture**							
	20%				**40%**			
	CMU	**CGU**	**CSeU**	**CSoU**	**CMU**	**CGU**	**CSeU**	**CSoU**
η v/s c	0.0010	0.0011	0.0012	0.0013	0.0011	0.0012	0.0013	0.0014
η_{sp} v/s c	0.0010	0.0011	0.0012	0.00125	0.0011	0.0012	0.0013	0.0014
ϕ v/s c	0.0010	0.0011	0.0012	0.0013	0.0011	0.0012	0.00124	0.0014
$\psi / \sqrt{c}$ v/s $\sqrt{c}$	0.0010	0.0011	0.0012	0.0013	0.0011	0.0012	0.00131	0.0014

The data has been analyzed in terms of Masson equations [21].

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

The values of limiting apparent molar volume ϕ^0_v and constant S_v are a measure of solute-solvent and solute –solute interaction respectively. A perusal of Table 8 show that values of apparent molar volume ϕ_v for all the copper soap urea complexes follows the order:

$$\phi_{v40} > \phi_{v20}$$

For copper (II) soap urea complexes, the solute-solvent interactions increase with the predominance of methanol concentration in ternary system: Complex + Methanol + Benzene. This may be attributed to the favorable interactions between polar groups of complex molecules and hydrogen bonded alcohol molecules. In general, the studies suggest those solute-solvent interactions below and above CMC in 20% and 40% Methanol-benzene mixture follows the order:

For 20%
CSeU>CSoU≈ CGU > CMU (Below CMC)
CSeU> CGU>CSoU> CMU (Above CMC)

For 40%
CSeU> CGU>CSoU> CMU (Below CMC)
CSoU>CGU >CSeU> CMU (Above CMC)

In copper (II) soap urea complexes, the solute-solute interactions decrease with the predominance of Methanol in the solvent mixture (Table 8). It follows the order:

$S_v20 > S_v40$
In general we can observed that for both the solvent the value of $S_{v1} > S_{v2}$ which is due to hydrophobic long chain of the solute molecules with increase in the polarity of solvent mixture.

It is interesting to point out that solute-solute interaction is rather lucid in alcohol as compared to hydrocarbon, this may be attributed to the fact that change in the mobility of complex molecule due to change in the dielectric constant of the solvent.

Table 8: Values of Viscometric Parameters Obtained From Different Equations Of Copper Soap Urea Complexes In 20% And 40% Methanol-Benzene Mixture

Name of the Complex	Masson's Equation				Jones-Dole Equation			
	ϕ^0_{v1}	ϕ^0_{v2}	S_{v1}	S_{v2}	A_1	A_2	B_1	B_2
CMU_{20}	-55.2	-20.6	+2.4750	+0.4040	8.20	7.18	-1.9626	-1.1503
CGU_{20}	-56.0	-18.2	+2.7474	+0.2867	8.58	4.82	-2.6050	0.2125
$CSeU_{20}$	-54.0	-17.6	+2.9042	+0.4040	8.90	4.78	-2.4740	0.2679
$CSoU_{20}$	-56.0	-18.4	+2.7474	+0.3639	9.16	5.74	-2.1445	0.0069
CMU_{40}	-56.0	-21.4	+2.3558	+0.4452	6.96	4.47	-2.7474	0.1763
CGU_{40}	-54.2	-18.8	+2.2460	+0.3639	7.15	4.68	-2.4750	0.2125
CSeU40	-53.4	-18.9	+2.3558	+0.3738	7.62	4.60	-3.7320	0.4452
CSoU40	-55.2-	-16.0	+2.3558	+0.2679	7.35	4.71	-3.0776	0.1943

3.2 Viscosity, and Specific viscosity:

Viscosity (η), Specific viscosity (η_{sp}) are considered as important rheological parameters. The viscosity of the copper soap urea complex derived from four edible oils in methanol-benzene mixture (20 % - 40 %) increases with the increases the soap-complex concentration (Table 3-6). The increase the viscosity with increase in soap urea complex concentration may be due to the increasing tendency of copper soap molecules to form aggregates in the form of micelles. The existence of micelles of surfactants in organic solvents have been reported by various workers [22-24]. Viscosity of copper urea soap in 40 % is slightly higher than that of 20 % methanol-benzene mixture due to the difference in the viscosities of solvent mixture. The plots of viscosity, η against complex concentration, c are characterized by an intersection of two straight lines corresponding to CMC of the complex. Of course this is the maximum concentration of molecular dispersion where balancing of internal forces is causing the formation of complex molecule aggregates [25-27]. It is clear from the CMC are dependent on the composition of solvent mixture. The values of the CMC in the higher volume percent of methanol are higher than that of lower volume percent of methanol. It is suggested that methanol takes quite different position in the micelles and the soap exhibit different degree of aggregation in the mixed solvent of varying compositions. The value of CMC is in close agreement with those obtained from other physical properties and follows the order:

CSoU>CSeU> CGU > CMU

This is in agreement with the fact that there is a decrease in the CMC values with the increase of average molecular weight of the complex. The values of specific viscosity are recorded in (Table 3-6) specific viscosity of the Copper soap-urea complexes increase with the increase in concentration. The variation of specific viscosity and fluidity against the complex concentration are characterized by an intersection of two straight lines at a definite complex concentration corresponding to the CMC of the complex. The values of CMC for complex solutions are recorded in (Table 7). The change in the curve spells out that there is a change in the pattern of complex agglomeration below and above CMC. The values of CMC are in good agreement with the CMC values obtained from other physical properties.
Jones Dole equation has also been applied to these complexes in methanol-benzene mixture [28].

$$\frac{\left(\frac{\eta}{\eta_0}\right)^2 - 1}{\sqrt{c}} = A + B\sqrt{c} \qquad (5)$$

For convenience, the equation may be expressed as

$$\frac{\psi}{\sqrt{c}} = A + B\sqrt{c} \qquad (6)$$

Where the coefficients A and B refer to solute –solute and solute-solvent interactions respectively. The plots of $\psi/\sqrt{c}$ v/s $\sqrt{c}$ are characterized by an intersection of two straight lines at a slight curvature at the point corresponding to the CMC of the complex. In view of the two intersecting straight lines for $\psi/\sqrt{c}$ v/s $\sqrt{c}$ plots, it is reasonable to evaluate two values of each coefficient below and above CMC designated as A_1, A_2, and B_1, B_2 respectively and are given in (Table 8). A perusal of (Table 8) shows that the coefficient A_1, A_2, and B_1, B_2 for the above referred system follows the order:

$A_1 > A_2$ & $B_2 > B_1$

On the basis of the above observation it may be suggested that the positive values of A suggest a strong solute-solute interactions and is greater in below CMC rather than the above CMC. Negative values of B_1 suggest weak solute-solvent interaction before CMC and above CMC the positive B_2 value indicate structure making tendency of the solute on the solvent molecules. The CMC value obtained for the above referred system is in close agreement to the value obtained from other physical properties and follows the order:

CSoU>CSeU> CGU > CMU

Negative value of B for copper soap-urea complexes for both the solvent i.e. 20% and 40% in methanol-benzene mixture reveals that dispersion interaction are dominating over solute-solvent specific interaction. On the basis of the results obtained it can fairly be concluded that the above treatment gives satisfactory explanation of clustering profile and confirm the existence of complex aggregation in non-aqueous mixed solvents. These studies have been supported by various works [29-30]. At the end it would not be out of place to point out the fact that the above treatment gives a phenomenological description that micellization and aggregation exist in non-aqueous mixed solvent [31-32].

4. Conclusion

The present research work makes an attempt to prepare complex compounds from metal and N or S containing organic compounds. It is found that the beneficial effects of the synthesized pharmaceuticals and can be used as antimalarial, antifungal sickness, antihistaminic and analgesics. 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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