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Micellar and Viscometric Properties of Some Bioactive Copper Soaps Derived From Karanj Oil in Non - Polar Solvent

Abstract

The viscosity, specific viscosity, and fluidity of Cu (II) surfactant derived from karanj oil in non-polar and polar solvent mixture have been determined at constant temperature 303.15 K. The results were used to determine the critical micelle concentration (CMC), solute-solvent interactions and the effect of chain length of the surfactant molecule on different parameters. The conclusions have been discussed in terms of well-known Moulik's and Jones- Dole equations. The effect of surfactant concentration on viscosity of the solution in solvent mixtures with different polarityhas been discussed. The observations suggested that the structure breaking effect by the solute on the solvent molecules is more prominent above CMC as compared to below CMC after the formation of the micelles. These study helps in interpretation of solute solvent interactions, structural insight of micelle and colloidal chemical behavior in binary/ternary systems. Study of these factors is essential as it indirectly makes them responsible for their immense applicability in wide areas of field. The vital information plays an important role for the selection of the synthesized bio-molecule in various research and development fields.

Keywords: Cu(II) surfactant, karanj oil, Soap-solvent interaction, CMC, Viscosity.

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1. INTRODUCTION

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 [1-4]. Colloidal behavior of Cu (II) soaps in non-aqueous solvent has been studied in our reported papers [5-6]. The studies of various parameter derived from viscometric equations provide revealing interpretations of solute – solvent interaction and structural insight of micelle in binary and ternary system, which plays a vital role in its selection for industrial and biological applications [7-8]. Recently, the deeper understanding of the role of metal ion in bio-system had led to the awareness that metal complexing is useful in the treatment of bacterial, fungal and viral diseases [9-10]. Karanj oil has insecticidal, antiseptic, anti-parasitic and cleansing properties. It has been used historically in the treatment of scabies, itch, herpes, eczema and sores [11]. The oil known as "Pongam Oil" is used for leather dressing, soap and candle-making, lubrications and illumination. It is applied in scabies, rheumatism, herpes, and leucoderma and also given as stomachic and cholagogue [12]. All these important applications led us to investigate the micellar features of copper (II) soaps derived from karanj oil in different solvents.

The copper soaps derived from karanj (*Pongamia Pinnata*) oil has been synthesized and studied for structural aspects, which were confirmed using various techniques like IR and NMR. The purity of these soaps was confirmed by M.P. and elemental analysis. Benzene-methanol has been selected as co- solvents since mixed solvents show tendency to interact with complex molecules and result in affecting the aggregation of complex molecule. Viscosity of soap solution has been deeply investigated in order to understand the nature of critical micelle concentration and miceller characterization. The study of functions such as specific viscosity and fluidity etc. of binary/ternary mixture are useful in understanding of nature and strength of molecular interactions between the components molecules. The importance of solute-solvent interaction and nature of solvation has been widely emphasized. Since, most of the reactions of chemicals and biological interest occur in solution; understanding of solvation phenomenon demands extensive studies of physical properties of electrolyte and non electrolytes in aqueous, mixed and non aqueous solvents.

2. EXPERIMENTAL

All the chemicals used were of AR grade. Karanj oil was extracted from their kernel using petroleum ether and then purified. The fatty acid composition of karanj oils was confirmed through GLC [13]. (Table 1) Copper karanj soap was synthesized as by earlier reported methods [14] (Direct Metathesis).

Table 1: % fatty acid composition of karanj oil used for synthesis of soap

Name of oil	% fatty acid (Carbon Number)							
	16:0	18:0	18:1	18:2	20:0	20:1	22:0	24:0
Karanj Oil	5.2	5.0	57.3	13.8	2.7	10.3	4.1	1.6

Molecular weights of copper soaps were determined from saponification value.

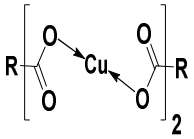
$$S.E. = \frac{56100}{S.V.} \qquad (1)$$

The value of S.V. and molecular weights are recorded in Table 2.

Table 2: Physical properties of copper karanj soap

Soap	Colour	M.P. (^{0}C)	Metal %		S.V.	S.E.	Av.
			Observed	Calculated			
CK	Dark green	51	9.4996	9.3426	181.5	309.091	679.682

The copper soap is abbreviated as follows:
Copper – Karanj soap (CK).



Structure of copper soap CK), where-R Mixed fatty acid chain as per given table-1

Ubbelohde type viscometer was used for measuring the viscosity of the solutions of varying concentrations of the soap. The densities were determined by means of a *Sprengel's* pyknometer. The viscosity of the soap solutions was calculated by the following relationship [15].

$$\rho = \frac{w}{w_0} \qquad (2)$$

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

Where η_0,η,$\rho_0\rho$, 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 (25 ± 0.1^0C) in a thermostat. The viscosity results are expressed in millipoise.

3. RESULTS AND DISCUSSION

Flow characterization of solutions of soaps in terms of viscometric measurements has been employed as tool for finding out the CMC of copper soaps in benzene and two compositions of benzene-methanol systems.

3.1 Viscosity

The effect of soap concentration on the viscosity of CK soap solution in pure benzene, 80% benzene-methanol and 60% benzene-methanol is observed. It is observed that the viscosities of CK soap solutions are of the order of viscosity of solvents [16-17].

The viscosity of CK soap solutions also initially increases then decreases with the increase in concentration and then suddenly increases after a definite concentration. The plot of viscosity η against the concentration c is characterized by an intersection of possible two straight lines (of two convex curves) with respect to x-axis at a definite concentration (Figure-1). And the intersection of the two possible straight lines of the curves corresponds to the CMC of the soap. This plot also shows curve nature before CMC and after CMC viscosity increases in a linearly curved fashion. It is observed that the viscosity of CK soap solutions is of the order of the viscosity of the solvents. The viscosity is:

$$CK_{100} > CK_{80} > CK_{60}$$

It is observed by data that CMC value of CK soap also decreases with the increase in polar solvent methanol in the dominance of non-polar solvent benzene. The CMC of the CK soap solution is:

$$CK_{100} > CK_{80} > CK_{60}$$

Hence the CMC data shows that it is dependent on the solvent composition and the values are in complete agreement with those obtained from other physical properties [18-19].

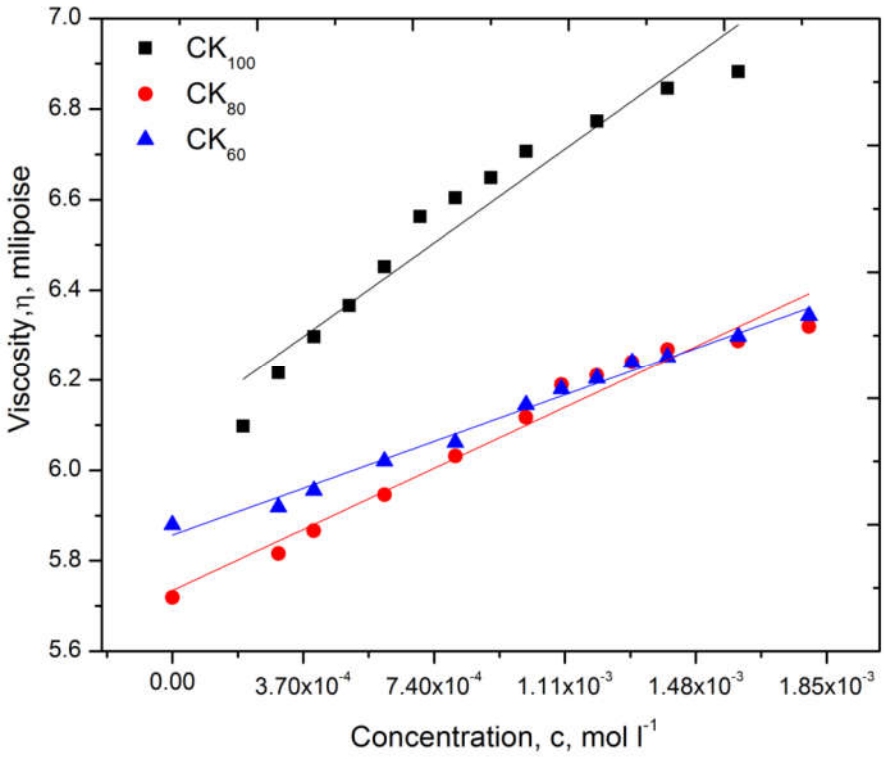


Figure 1: Plots of viscosity v/s concentration of copper soaps derived from karanjoil in 100 % benzene, 80 % benzene -methanol and 60 % benzene- methanol system at 298.15 K

3.2 Specific Viscosity

The ratio of change in viscosity to the original viscosity of the solvent is called the specific viscosity η_{sp}[20]

$$\eta_{sp} = \frac{\eta}{\eta_0} - 1 \qquad (4)$$

Where η and η_0 are the viscosities of solution and solvent respectively.

The specific viscosity η_{sp} of solutions is recorded in Table 3. The effect of the soap concentration on specific viscosity of copper (II) solution of variable concentrations i.e. pure benzene, 80% benzene–methanol, 60% benzene-methanol was found initially increases then decrease with the increase in the concentration, and after a definite concentration corresponding to the CMC of the soap solution shows increases again. The plot of specific viscosity η_{sp} against the concentration c of the soap is characterized by an intersection of possible two straight lines (of two convex curves) with respect to x-axis. The change in the trend of the plot below and above CMC however suggests that there is phenomenal change in the micellar agglomeration below and above CMC. The value of CMC so obtained is in good agreement with the value obtained from the other physical parameters like viscosity. It is observed that the viscosities of CK soap solutions are of the order of specific viscosity of solvents [21].

Table 3: Specific viscosity data for copper-karanj solution in pure benzene, 80% benzene-methanol and 60% benzene-methanol mixture at 25°C

Concentration (mol L^{-1})	CK_{100}	CK_{80}	CK_{60}
0.0002	0.0473	0.0194	0.0593
0.0003	0.0661	0.0574	0.0889
0.0004	0.1221	0.0394	0.0698
0.0005	0.1025	0.0297	0.0594
0.0006	0.0372	0.0098	0.0391
0.0007	0.0932	0.0196	0.0489
0.0008	0.1120	0.0482	0.0686
0.0009	0.1402	0.0673	0.0884
0.0010	0.1684	0.0868	0.1178
0.0012	0.2064	0.1153	0.1383
0.0014	0.2624	0.1529	0.1874
0.0016	0.1794	0.1609	0.1971

3.3 Fluidity

The fluidity ϕ which is defined as reciprocal of viscosity [22]

$$\phi = \frac{1}{\eta} \qquad (5)$$

The fluidity ϕ of CK_{100}, CK_{80} and CK_{60} initially decrease than increase and then decreases again with increase in concentration after a definite concentration corresponding to CMC of the soap. The plot of fluidity ϕ against concentration C of the soap, show an abrupt change at a point corresponding to the CMC of the soap change at a point corresponding to the CMC of the soap solution (Figure 2). The plot is characterized by an intersection of two possible straight lines of two concave curves at CMC of the soap [23]. A perusal of CMC data shows that it is in good agreement with the values obtained from η v/s c and η_{sp} v/s c plots.

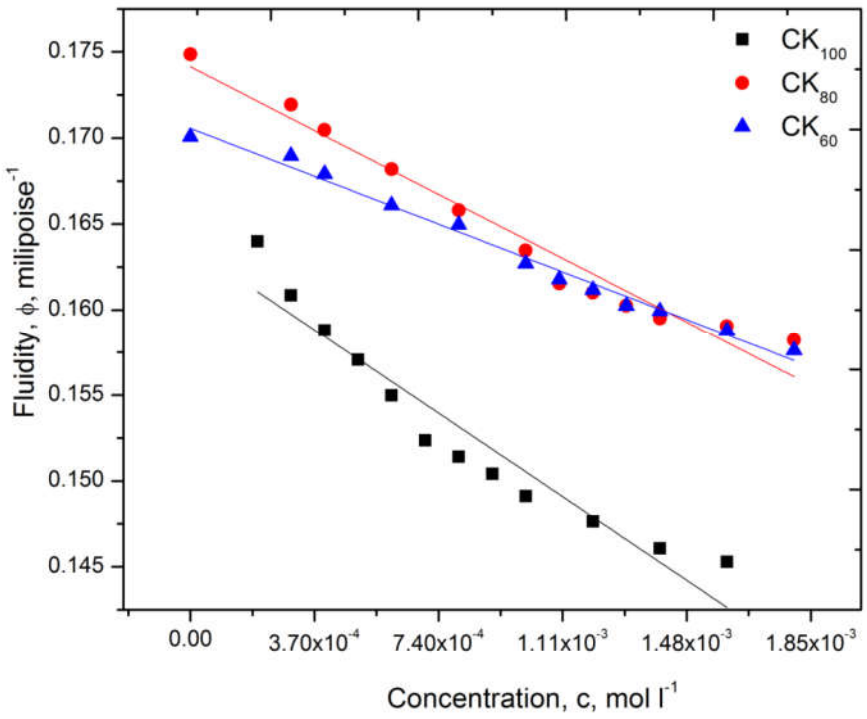


Figure 2: Plots of Fluidity v/s concentration of copper soaps derived from karanj oil in 100% benzene, 80% benzene -methanol and 60% benzene- methanol system at 298.15 K

3.4 Important Equations:

The results of the viscosities of the CN_{100}, CN_{80} and CN_{60} can be satisfactory explained when applied to the various equations discussed below.

Einstein Equation: [24]

$$\eta_{sp} = 2.5\ \nabla .\ c \qquad (6)$$

Thomas equation:[25]

$$\frac{(\frac{\eta}{\eta_0} - 1)}{c} = 2.5\ \nabla + (10.05\ \nabla)^2 .\ c \qquad (7)$$

Vand equation[26]

$$\frac{1}{c} = \left(\frac{0.921}{\nabla}\right)^{-1} \frac{1}{\log\left(\frac{\eta}{\eta_0}\right)} + Q\ \nabla \qquad (8)$$

Where $\bar{V}$, C, Q, η and η_0 are the molar volume of the soap solution, concentration of the soap solution, interaction coefficient, viscosity of the solution and solvent respectively.

The viscometric data has been interpreted in the light of Einstein, Thomas and Vand's equation for the solutions of CK_{100}, CK_{80} and CK_{60} (Figures 3-5) respectively. The molar volume values obtained from these equations are recorded in Table 4.

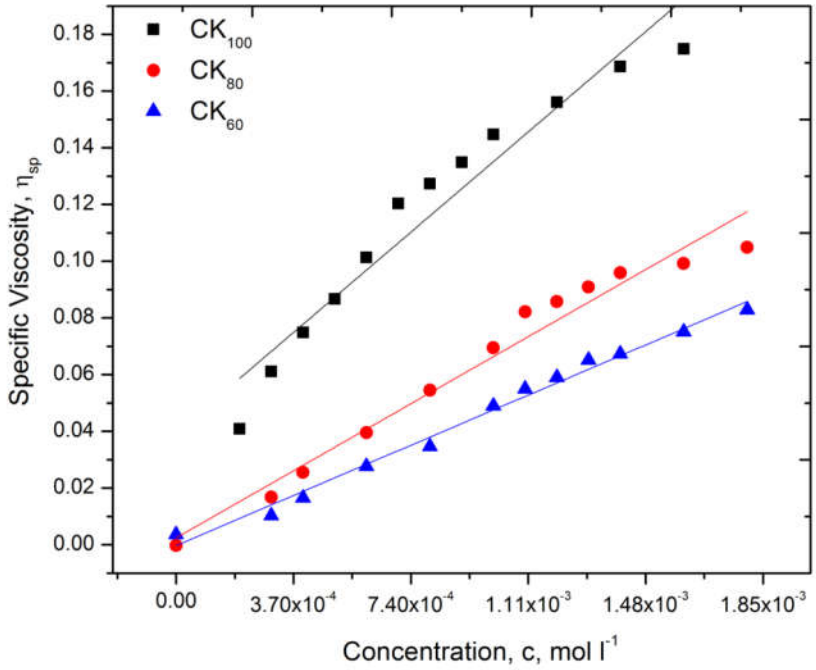


Figure 3: Plots of η_{sp} v/s concentration of copper soaps derived from karanj oil in 100% benzene, 80% benzene -methanol and 60 % benzene- methanol system at 298.15 K (Einstein Equation)

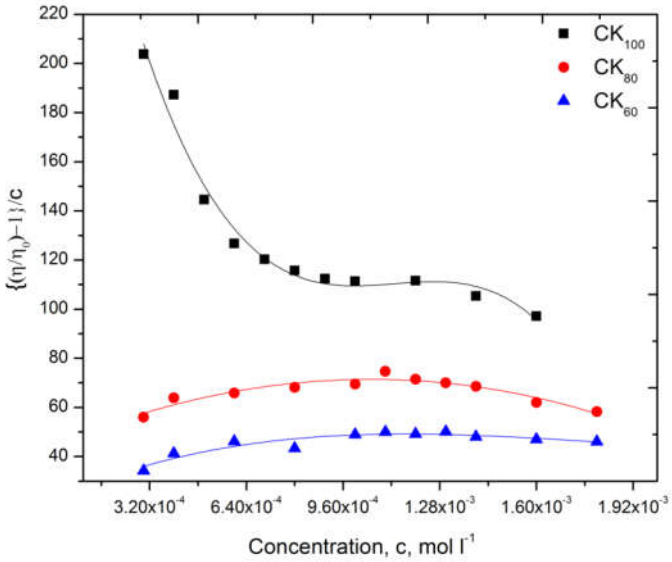


Figure 4: Plots of $\{(\eta/\eta_0)-1\}/c$ v/s concentration of copper soaps derived from karanjoil in 100% benzene, 80% benzene -methanol and 60% benzene- methanol system at 298.15 K (Thomas Equation)

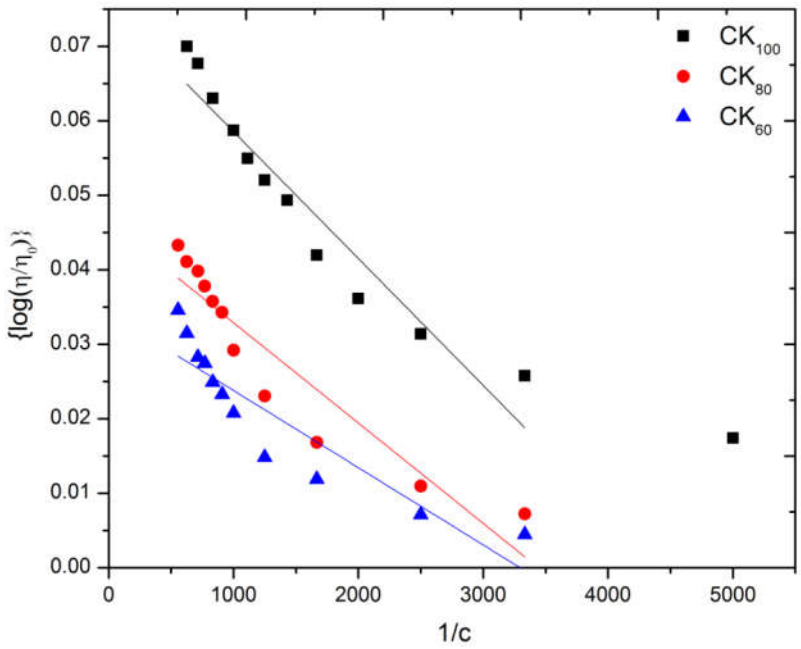


Figure 5: Plots of 1/ {log (η/η_0)}v/s 1/c of copper soaps derived from karanj oil in 100% benzene, 80% benzene -methanol and 60% benzene- methanol system at 298.15 K (Vand Equation)

Table 4: Molar volume derived from different equations for copper-karanj solution in pure benzene, 80% benzene-methanol and 60% benzene-methanol mixture at 25°C

Soap	Einstein Equation		Vand Equation		Thomas Equation	
	$\bar{V}_1$	$\bar{V}_2$	$\bar{V}_1$	$\bar{V}_2$	$\bar{V}_1$	$\bar{V}_2$
CK_{100}	-169.5300	132.8000	-13.6796	10.5570	316.4600	-49.9400
CK_{80}	-61.3100	54.7800	-3.3080	16.5473	105.6700	12.1000
CK_{60}	-61.4100	39.5100	-25.8671	31.4614	157.1100	30.2800

Interestingly Moulik's equation also fits equally well to our referred systems. The equation is [27]

$$\left(\frac{\eta}{\eta_0}\right)^2 = M + K\ c^2 \qquad (9)$$

Where M and K are constants. The values of M and K are evaluated from the plot of $(\eta/\eta_0)^2$ v/s c^2, which are recorded in Table 5. The plot of $(\eta/\eta_0)^2$ v/s c^2 is characterized by an intersection of two straight lines (Figure 6). Thus the value of M and K could be evaluated both below and above CMC and are designated as M_1, M_2 and K_1, K_2 respectively.

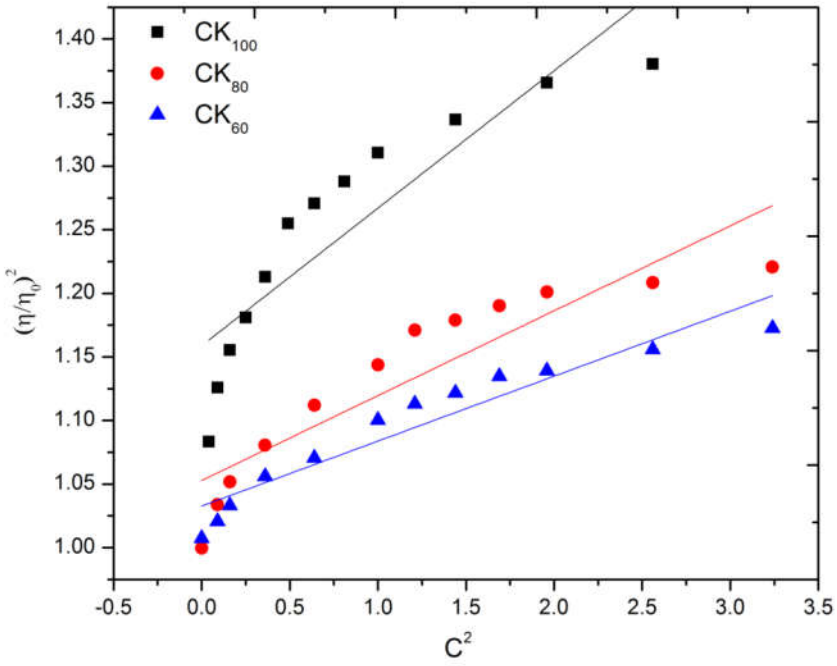


Figure 6: Plots of $(\eta/\eta_0)^2$ v/s c^2 of copper soaps derived from karanj oil in 100% benzene, 80% benzene -methanol and 60% benzene- methanol system at 298.15 K (Moulik's equation)

Table 5: Computed parameters derived from different equations for copper-karanj solution in pure benzene, 80% benzene-methanol and 60 % benzene-methanol mixture at 25°C

Soap	Moulik's Equation				Jones – Dole Equation			
	M_1	M_2	K_1	K_2	A_1	A_2	B_1	B_2
CK_{100}	1.0406	0.9107	-0.9152	0.4639	26.4840	-12.0320	-1019.0000	551.9000
CK_{80}	1.1513	1.0281	-0.3660	0.1077	10.0700	-3.5260	-395.3300	189.9100
CK_{60}	1.2136	1.0740	-0.3746	0.1390	13.6690	-2.5400	-492.7500	186.3300

The intrinsic viscosity [η] = limit (c→ 0) [η_{sp}] of the soap solution according to Gray and Alexander can be represented by the equation: [28]

$$\frac{c}{\eta_{sp}} = \frac{1}{\eta} - K.c \qquad (10)$$

The values of intrinsic viscosity and coefficient K′ are determined from the plot of reduced viscosity (c/η_{sp}) against the concentration c of the soap. The plot of (c/η_{sp}) v/s c is characterized by an intersection of two straight lines (Figure 7), thus Gray and Alexander equation fits well both below and above CMC. The values of intrinsic viscosity [η] and K′ evaluated from the plot is recorded in Table 6. The sudden change in the trend of the plot at CMC clearly indicated that the change in the behaviour of the soap takes place in the periphery of this concentration.

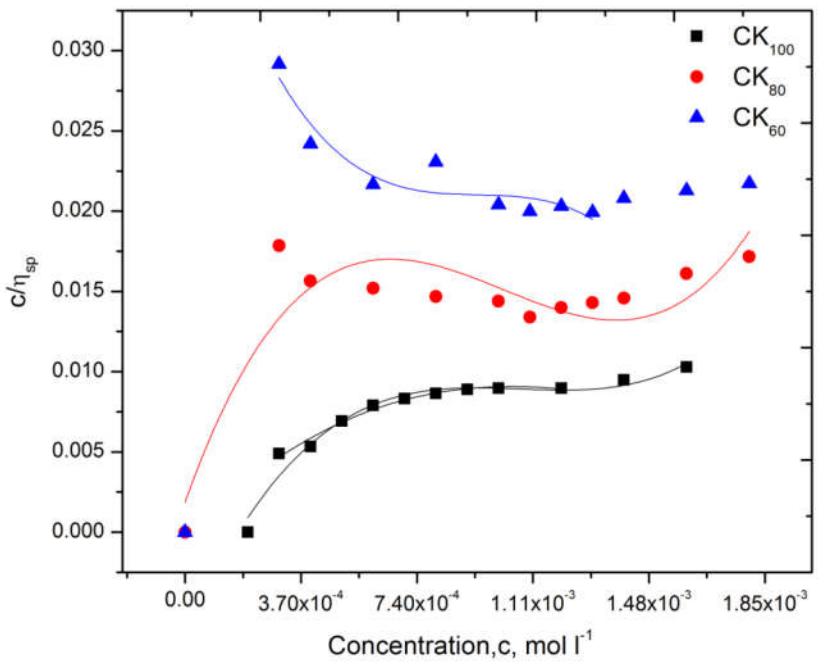


Figure 7: Plots of (c/η_{sp}) v/s c of copper soaps derived from karanj oil in 100% benzene, 80% benzene -methanol and 60% benzene- methanol system at 298.15 K (Gray and Alexander equation)

Table 6: Computed parameters for Gray-Alexander equation for copper-karanj solution in pure benzene, 80% benzene-methanol and 60% benzene-methanol mixture at 25°C

Soap	Gray – Alexander's Equation			
	$[\eta_1]$	$[\eta_2]$	K`1	K`2
CK_{100}	-41.6667	119.0476	-64.0570	2.0340
CK_{80}	-80.0000	76.3359	-58.1590	3.9482
CP_{60}	-74.0741	71.4286	-48.0950	4.5510

The viscosity data have also been interpreted in the light of Jones – Dole Equation [29]

$$\frac{\left(\frac{\eta}{\eta_0}\right)^2 - 1}{\sqrt{c}} = A + B\sqrt{c} \qquad (11)$$

For convenience, the equation may be expressed as:

$$\frac{\psi}{\sqrt{c}} = A + B\sqrt{c} \qquad (12)$$

Where the coefficient A and B refer to the solute–solute and solute–solvent interaction respectively. The values of A and B are evaluated from the plot of ($\psi/\sqrt{c}$) v/s $\sqrt{c}$, which is characterized by an intersection of a two straight lines (Figure 8). Thus Jones–Dole equations fit well both below and above CMC. The value of CMC so obtained is in close agreement with the CMC obtained from η v/s c, η_{sp} v/s c, and ϕ v/s c plots.

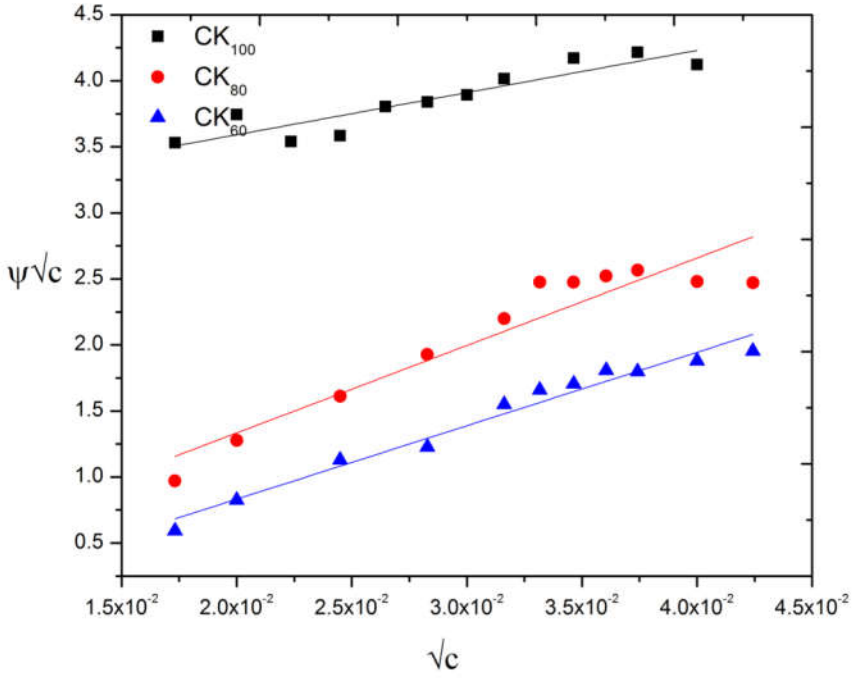


Figure 8: Plots of (ψ/√c) v/s √c of copper soaps derived from karanj oil in 100% benzene, 80% benzene -methanol and 60% benzene- methanol system at 298.15 K (Jones – Dole equation)

The values of A and B so evaluated are recorded in Table 5. Literature survey reveals that positive value of A suggests a strong solute–solute interaction [30].

B co-efficient measures the structural modification induced by solute-solvent interaction. Earlier studies done show that, the increase in concentration of solute in solution contributes positively to the viscosity of B-coefficient [31-32]. Therefore the systems/molecule exhibiting negative B-coefficient have been assumed to exert a structure breaking effect by the solute on the solvent molecules [33-34].

Thus it is suggested that solute-solute interaction is greater below CMC as compared to above CMC for our referred CK_{100}, CK_{80} and CK_{60} system as $A_1 > A_2$ and A_1 is positive while $A_2$is negative for all the systems (Table 7) Literature survey [35-36] also reveals that B-coefficient measures the structural modification induced by solute-solvent interaction, and the molecules exhibiting negative B co-efficient have been assumed to exert a structure–breaking effect by the solute on the solvent molecules and it has been observed for our CK_{100}, CK_{80} and CK_{60} system that the B-coefficient below CMC i.e. B_1 is negative. This suggests that structure-breaking effect is more below CMC as $B_1 < B_2$. It can be suggested that the structure breaking effect by the solute on the solvent molecules is more prominent below CMC as compared to above CMC i.e. after the formation of the micelles.

Table 7: Values of CMC for copper-karanj solution in pure benzene, 80% benzene-methanol and 60% benzene-methanol mixture at 25°C

Plot	CK_{100}	CK_{80}	CK_{60}
η v/s c	0.00058	0.00053	0 .00051
η_{sp} v/s c	0.00058	0.00053	0 .00051
ϕ v/s c	0.00058	0.00053	0 .00051

(η_{sp}/c) v/s c	0.00058	0.00053	0 .00051
$(\psi/\sqrt{c})$ v/s $\sqrt{c}$	0.00058	0.00053	0 .00051
$(\eta/\eta_0)^2$ v/s c^2	0.00058	0.00053	0 .00051
(1/c) v/s $\{1/\log(\eta/\eta_0)\}$	0.00058	0.00053	0 .00051
(C/η_{sp}) v/s c	0.00058	0.00053	0 .00051

4. CONCLUSION

Colloidal behaviour of copper(II) soap complexes play a significant role in their selection in various fields like preservatives, herbicidal, pesticidal activities, detergency, paints, lubrication. In biological systems, these agents are vital components and useful in many industrial process. The viscosity data are important to relate the micellar features, CMC and nature of the synthesized bioactive molecule in different solvents. The studies done will contribute in identifying the structural, physical and biochemical nature of copper soaps. Above study definitely generates a new hope in pharmaceutical, agrochemical and therapeutic applications of complex. The present research work makes an attempt to prepare surface active compounds from metal and natural oils. It is found that the beneficial effects of the synthesized biologically active molecules, is yet to be explore in the field of agrochemicals and pharmaceuticals. 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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