

Ultrasonic Studies of Copper Soap-urea Complexes Derived from Mustard and Soybean Oils

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ABSTRACT: *Ultrasonic velocities and densities have been measured in ternary mixtures containing copper soap-urea complexes derived from mustard and soybean oils in 20% and 40% methanol-benzene mixture to assess the effect of polarity of solvent on the micellar features of surfactant. The studies suggest that predomination of benzene (higher percentage of non-polar solvent) plays a significant role in compacting the molecular organisation of the micelles. From these values, the ultrasonic velocity (u), the specific acoustic impedance (Z), adiabatic compressibility (β), intermolecular free length (L_f), apparent molar compressibility (f_k), molar sound velocity (R) and primary solvation number (S_n) have been calculated. The data clearly indicate that the values of u , Z , S_n and R increase whereas the values β and L_f decrease consistently with increase in the soap complex concentration. The critical micelle concentration (CMC) has also been determined and it has been found that CMC is dependent on the composition of the solvent mixture and composition of the edible oil. The results have been analysed in terms of Masson's equation. The results have been explained on the basis of intermolecular interactions between solute and solvent molecules and effect of polarity and concentration were accessed on various acoustic parameters.*

Keywords: Copper soap-urea complex, methanol, benzene, acoustic parameters, intermolecular interactions

1. INTRODUCTION

Study of propagation of ultrasonic waves and their absorption forms one of the most important methods of investigation of properties of matter in all the three states. It is well known that study of absorption of ultrasonic waves in a medium provides important information about various inter and intra-molecular processes such as relaxation of the medium or the existence of isomeric states or the exchange of energy between various molecular degrees of freedom.¹⁻⁶ Derived parameters from ultrasonic velocity measurements provide qualitative information regarding the nature and strength of molecular interactions in liquid mixtures.^{7,8} This has been studied for various binary and ternary mixtures with respect to variation in concentration of the liquids and temperatures.^{9,10} Ultrasonic velocity measurements have been extensively used to study the molecular interactions in electrolyte solutions and to get significant information on the arrangement of molecules in aqueous electrolyte solutions.¹¹⁻¹⁵ In the present investigation, we have studied various thermo-acoustical parameters, such as specific acoustic impedance (Z), adiabatic compressibility (β), intermolecular free length (L_f), apparent molar compressibility, (f_k), molar sound velocity (R) and primary solvation number (S_n) to examine the ion-solvent and ion-ion interactions in the aqueous solutions of soap urea complexes made by mustard and soybean oils which have wide applications in pharmaceutical, medicinal, agricultural, environmental, textile and leather industries.¹⁶⁻²⁰

The present work deals with the determination of ultrasonic velocity measurements which have been used to obtain information regarding various acoustic parameters and solute-solute and solute-solvent interactions of solutions of Cu(II) soap urea complex of edible oil of mustard and soybean in pure non-polar solvents, polar and non-polar solvent mixtures (benzene and its varying compositions with methanol) to assess the effect of polarity of solvent molecules on the micellar nature of solute molecule. The studies have also been done to understand the nature of the solution and observed that the increase in the internal pressure and in lowering the compressibility of the solution. Benzene as a solvent has been used due to maximum solubility of soaps in this non-aqueous solvent.

2. EXPERIMENTAL

All the chemicals used were of lab reagent/analytical reagent (LR/AR) grade. Copper soap was prepared by refluxing the mustard and soybean oils (mustard oil extracted from seeds and purified, and soybean oil available in market with reputed brand) with ethyl alcohol and 2N KOH solutions for 3-4 h (direct metathesis). The neutralisation of excess KOH present was done by slow addition

of 0.5N HCl. Saturated solution of copper sulphate was then added to it, for conversion of neutralised potassium soap into their corresponding copper soaps. Copper soap so obtained was then washed with warm water and 10% alcohol at 50°C and recrystallised using hot benzene. Molecular weights of copper soaps were determined from saponification values.²¹ The copper soap complexes are abbreviated as copper-mustard urea (CMU) and copper-soybean urea (CSoU). Fatty acid composition of oils used for synthesis of Cu(II) soaps and their analytical and physical data are summarised in Tables 1 and 2.

Table 1: Fatty acid composition of oils used for copper urea complexes synthesis.

Name of oil	% Fatty acids					Other acids
	16:0	18:0	18:1	18:2	18:3	
Mustard oil	2	1	25	18	10	C ₂₀ –C ₂₂ 41%
Soybean oil	12	4	24	51	9	–

Table 2: Analytical and physical data of copper urea complexes derived from mustard and soybean oils.

Compound	Colour	Melting point (°C)	Metal %		Mol.wt.
			Found	Calculated	
CMU	Dark green	72	8.46	8.35	759.72
CSoU	Dark green	100	9.31	9.10	697.17

The ultrasonic velocity was determined with the help of a single crystal ultrasonic interferometer model M-82 (Mittal Enterprises, New Delhi) working at a fixed frequency of 2 MHz. The temperature of water was maintained at 30°C ± 0.1°C by a thermostat through the jacket of the cell before the measurement was actually made. The measured velocities have an uncertainty of 0.5 m s⁻¹. Further, the operative part of the measurement involves the slow movement of micrometer till the anode current meter shows a maximum. A number of maxima readings of anode current are passed on and their number (n) is counted. The total distance (d) thus moved by the micrometer gives the value of wavelength (λ) with the help of the following relation:

$$d = n \times \lambda \quad (1)$$

Once the wavelength is known, the velocity (u) in the liquid can be calculated with the help of the following relation:

$$u = \lambda \times f \quad (2)$$

where f is the frequency of instrument used, i.e., 2 MHz.

3. ACOUSTIC PARAMETERS

The values of Z , β , L_f , ϕ_k , R and S_n are calculated by using the following relationships:²²

$$Z = u\rho \quad (3)$$

$$\beta = \frac{1}{u^2\rho} \quad (4)$$

$$L_f = K\sqrt{\beta} \quad (5)$$

$$\phi_k = \left[1000 \cdot \frac{\beta\rho_0 - \beta_0\rho}{c\rho_0} \right] + \frac{\beta M}{\rho_0} \quad (6)$$

$$R = M \cdot (u)^{1/3} \rho_0 \quad (7)$$

$$M = X_1 M_1 + X_2 M_2 + X_3 M_3 \quad (8)$$

$$S_n = \left[\frac{n^0}{n} \right] \cdot \left[1 - \frac{V\beta}{n^0 V^0 \beta^0} \right] \quad (9)$$

where b_0 , β , r_0 , ρ , n_0 , n , V_0 and V are adiabatic compressibility, density, numbers of moles and molar volume of solvent and solute, respectively. The expression for molar volume of solvent V_0 and soap solution V may be written as:

$$V = X_1 M_1 + X_2 M_2 + X_3 M_3 / \rho \quad (10)$$

$$V_0 = X_1 M_1 + X_2 M_2 + X_3 M_3 / \rho_0 \quad (11)$$

In Equations 6, 8 and 9, M represent the molecular weight and X the mole fraction of the component. Here the subscripts 1, 2 and 3 refer to soap, methanol and benzene, respectively.

4. RESULTS AND DISCUSSION

4.1 Copper Soap-urea Complex Derived from Mustard and Soybean Oils in 20% Methanol-benzene Mixture

4.1.1 Acoustic parameters

After perusal of the observed data, it may be mentioned that the acoustic parameters and equations related to ultrasonic investigations are the same as has been enumerated for the previous soaps. The values of ultrasonic velocity and allied parameters for these complexes in methanol-benzene mixture are given in Tables 3 and 4.

A perusal of the data suggests that the values of u , Z , β , ϕ_k , R and S_n increase with increasing complex concentration whereas β and L_r decrease with the increase in the concentration. Z has a greater significance and as a characteristic property of the medium than either density or ultrasonic velocity individually.²³

Table 3: Ultrasonic velocity and other acoustic parameter of copper urea complex derived from mustard oil in 20% methanol-benzene mixture (CMU₂₀).

c (mol l ⁻¹)	ρ (g dm ⁻³)	u (m s ⁻¹)	$Z \times 10^{-5}$ (g cm ⁻² s ⁻¹)	$b_{ad} \times 10^{11}$ (cm ² dyne ⁻¹)	$L_r \times 10^{10}$ (A ⁰)	$-\phi_k \times 10^8$	$R \times 10^{-2}$	$S_n \times 10^{-3}$
0.0003	0.9548	1538.5	1.4689	4.4250	1.3171	4220.7	34.700	-57.757
0.0004	0.9556	1544.4	1.4758	4.3874	1.3115	3270.5	34.719	-42.598
0.0006	0.9565	1545.6	1.4784	4.3765	1.3099	2205.9	34.701	-28.232
0.0008	0.9574	1548.6	1.4826	4.3555	1.3067	1686.2	34.696	-20.960
0.0010	0.9559	1563.1	1.4942	4.2816	1.2956	1413.0	34.864	-16.302
0.0011	0.9564	1565.6	1.4973	4.2660	1.2932	1301.0	34.867	-14.708
0.0012	0.9569	1569.2	1.5016	4.2438	1.2899	1213.2	34.879	-13.342
0.0013	0.9574	1571.1	1.5042	4.2316	1.2880	1131.2	34.877	-12.239
0.0014	0.9579	1574.2	1.5079	4.2128	1.2851	1065.7	34.885	-11.262
0.0016	0.9586	1579.2	1.5138	4.1832	1.2806	953.0	34.902	-9.714
0.0018	0.9595	1584.2	1.5200	4.1530	1.2760	866.4	34.912	-8.504
0.0020	0.9605	1588.6	1.5258	4.1257	1.2718	795.9	34.913	-7.546

Table 4: Ultrasonic velocity and other acoustic parameter of copper urea complex derived from soybean oil in 20% methanol-benzene mixture (CSoU₂₀).

c (mol l ⁻¹)	ρ (g dm ⁻³)	u (m s ⁻¹)	$Z \times 10^{-5}$ (g cm ⁻² s ⁻¹)	$b_{ad} \times 10^{11}$ (cm ² dyne ⁻¹)	$L_r \times 10^{10}$ (A ⁰)	$-\phi_k \times 10^8$	$R \times 10^{-2}$	$S_n \times 10^{-3}$
0.0003	0.9554	1533.2	1.4648	4.4529	1.3213	4140.1	34.638	-58.343
0.0004	0.9562	1535.5	1.4683	4.4355	1.3187	3159.3	34.629	-43.393
0.0006	0.9572	1539.1	1.4732	4.4105	1.3150	2156.5	34.625	-28.590
0.0008	0.9584	1540.8	1.4767	4.3948	1.3126	1644.9	34.600	-21.262
0.0010	0.9592	1548.6	1.4854	4.3473	1.3055	1367.4	34.634	-16.656
0.0011	0.9598	1551.6	1.4892	4.3278	1.3026	1263.7	34.637	-15.003
0.0012	0.9604	1553.4	1.4919	4.3150	1.3006	1171.6	34.632	-13.664
0.0013	0.9585	1559.5	1.4947	4.2901	1.2969	1091.7	34.748	-12.526
0.0014	0.9590	1561.3	1.4973	4.2778	1.2950	1024.3	34.746	-11.561
0.0016	0.9598	1564.3	1.5014	4.2576	1.2919	911.4	34.745	-10.014
0.0018	0.9606	1567.4	1.5056	4.2374	1.2889	823.5	34.744	-8.812
0.0020	0.9615	1570.5	1.5100	4.2169	1.2858	753.7	34.739	-7.847

The values of Z follow the order: CMU > CSoU.

This indicates that lyophobic interaction between complex and solvent molecules is higher in CSoU than those in CMU. It may be suggested that as $>C=O$ and NH_2 groups of urea are capable of making intermolecular hydrogen bonding with other CSoU molecules, their interactions strength contribution is highest. For both complexes, the values of β and L_f decrease with increasing complex concentration which indicates that there are significant interactions between complex and solvent molecules. It is interesting to point out that the values of the parameters u , Z , β and L_f with respect to chain length of the complex follow the order:

$$[u]: \text{CMU} > \text{CSoU}$$

$$[Z]: \text{CMU} > \text{CSoU}$$

$$[b]: \text{CMU} < \text{CSoU}$$

$$[L_f]: \text{CMU} < \text{CSoU}$$

For both complexes, the S_n increases with the increase in complex concentration signifying the aggregation of solvent molecules around the solute moiety. Negative values of solvation number indicate that the solutions are more compressible than the solvent. Many authors in literature report negative solvation number.²⁴ These results indicate that strong solute-solvent interaction takes place in the solution. In general, one can consider two types of interactions between the component molecules such as physical and chemical interactions which are arising from dispersion forces or weak dipole-dipole interactions making a positive contribution and the chemical or specific interactions due to the charge transfer, hydrogen bond formation and other complex forming interaction results in negative values.²⁵

4.1.2 Micellar features

The plots of u , Z , β and L_f against complex concentration (c) are characterised by an intersection of two straight lines corresponding to the CMC of the complex (Figures 1 to 4). It reveals that CMC is dependent on the composition of the solvent mixture. Like copper soaps derived from various edible oils in methanol-benzene mixture, copper soap-urea complex too also obeys Equation 10, showing a linear variation of ultrasonic velocity with complex concentration. The values of ϕ_k increases with increasing complex concentration. The ϕ_k values are negative for all urea complexes of copper soap. Literature survey reveals that negative value of ϕ_k indicates electrostriction and hydrophobic interaction.²⁶ The negative values of ϕ_k are also attributed to the loss of structural compressibility of solute molecules. The plot of ϕ_k vs. \sqrt{c} is characterised by an intersection of two straight lines corresponding to the CMC of the complex (Figure 5). The ϕ_k data has also been

analysed in terms of Masson's equation. The parameters ϕ_{k2}^0 and S_k have been evaluated both below and above CMC. The order of this parameter is as follow:

$$\phi_{k1}^0 < \phi_{k2}^0 \quad \text{and} \quad S_{k1} > S_{k2}$$

From this order, it may be suggested that solute-solvent interaction is more pronounced in concentrate solution, i.e., above CMC as compared to below CMC, this observation is in good agreement with our apparent molar volume ϕ_v interpretations for complexes.

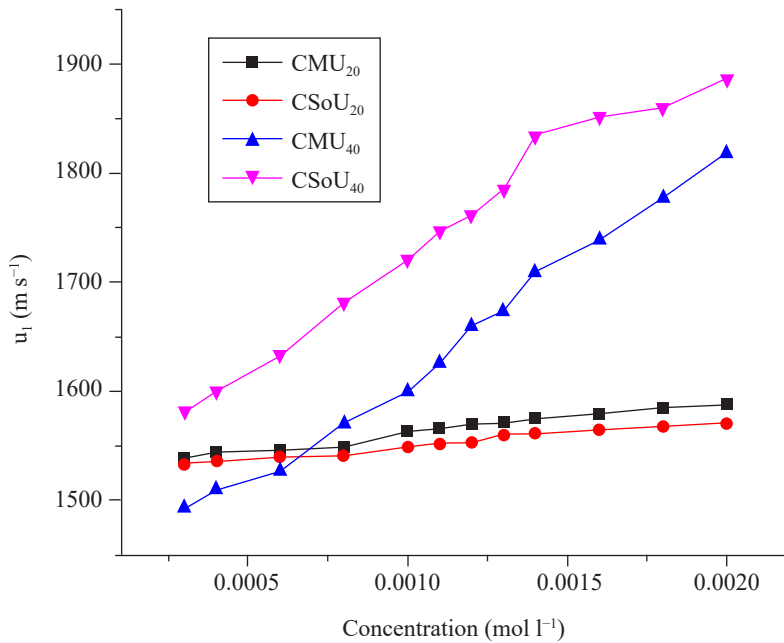


Figure 1: Plots of u vs. c of copper soap urea complex derived from mustard and soybean oil in 20% and 40% methanol-benzene mixture.

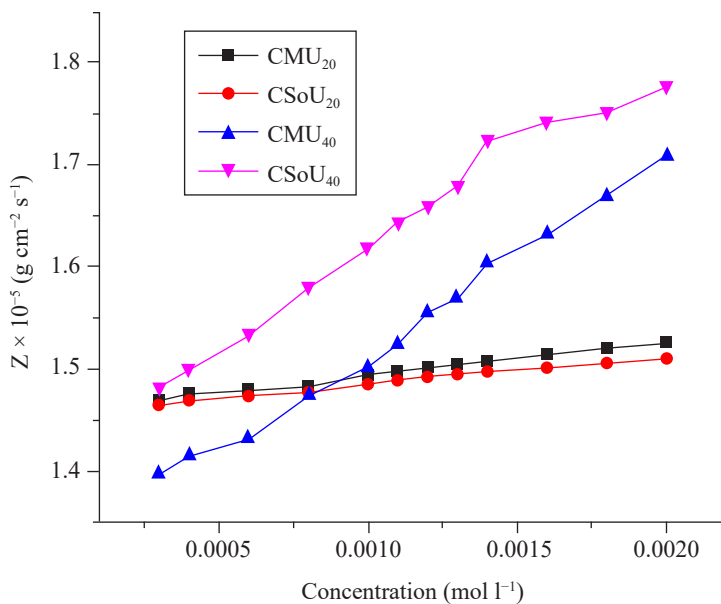


Figure 2: Plots of Z vs. c of copper soap urea complex derived from mustard and soybean in 20% and 40% methanol benzene mixture.

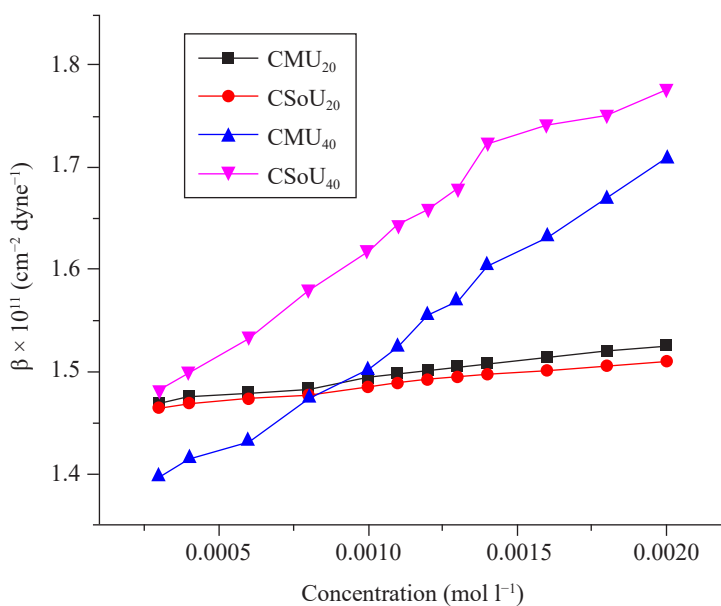


Figure 3: Plots of β_{ad} vs. c of copper soap urea complex derived from mustard and soybean in 20% and 40% methanol benzene mixture.

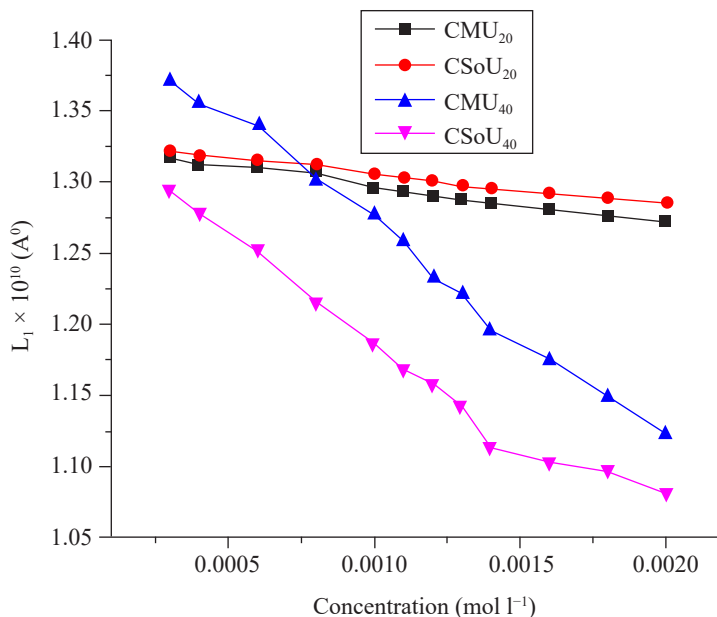


Figure 4: Plots of L_r vs. c of copper soap urea complex derived from mustard and soybean oil in 20 % and 40 % methanol benzene mixture.

The CMC value follows the order:

$$\text{CSoU} > \text{CMU}$$

The results are in agreement with the fact that there is decrease in the CMC value with the increase in average molecular weight of the complex.

4.2 Copper Soap-urea Complex Derived from Mustard and Soybean Oils in 40% Methanol-benzene Mixture

4.2.1 Acoustic parameters

At the very outset it may be mentioned that the acoustic parameters and related equations pertaining to ultrasonic investigations for the previously-referred system are almost the same as have been enumerated for the 40% methanol-benzene mixture and are recorded in Tables 5 and 6. The data clearly indicate that the values of u , Z , ϕ_k , R and S_n increase whereas the value of β and L_r decreases consistently with increase in the complex concentration in varying compositions of methanol-benzene mixture.^{27,28}

Table 5: Ultrasonic velocity and other acoustic parameter of copper urea complex derived from mustard oil in 40% methanol-benzene mixture (CMU₄₀).

c (mol l ⁻¹)	ρ (g dm ⁻³)	u (m s ⁻¹)	Z × 10 ⁻⁵ (g cm ⁻² s ⁻¹)	b _{ad} × 10 ¹¹ (cm ² dyne ⁻¹)	L _r × 10 ¹⁰ (A ⁰)	-φ _k × 10 ⁸	R × 10 ⁻²	S _n × 10 ⁻³
0.0003	0.9360	1492.5	1.3970	4.7959	1.3712	1936.8	28.787	-64.180
0.0004	0.9368	1509.4	1.4140	4.6852	1.3553	1739.9	28.873	-46.008
0.0006	0.9375	1526.7	1.4313	4.5763	1.3394	1346.7	28.966	-29.290
0.0008	0.9384	1570.5	1.4737	4.3207	1.3015	1334.9	29.217	-19.562
0.0010	0.9389	1600.0	1.5022	4.1605	1.2771	1230.1	29.388	-14.449
0.0011	0.9370	1626.0	1.5236	4.0366	1.2580	1220.6	29.609	-12.357
0.0012	0.9374	1659.8	1.5559	3.8725	1.2321	1257.2	29.803	-10.303
0.0013	0.9378	1673.6	1.5695	3.8068	1.2217	1212.4	29.875	-9.127
0.0014	0.9382	1709.4	1.6038	3.6477	1.1958	1240.8	30.076	-7.624
0.0016	0.9386	1739.1	1.6323	3.5225	1.1751	1164.8	30.242	-6.086
0.0018	0.9392	1777.8	1.6697	3.3689	1.1492	1122.2	30.450	-4.769
0.0020	0.9399	1818.2	1.7089	3.2184	1.1233	1086.8	30.661	-3.727

Table 6: Ultrasonic velocity and other acoustic parameter of copper urea complex derived from soybean oil in 40% methanol-benzene mixture (CSoU₄₀).

c (mol l ⁻¹)	ρ (g dm ⁻³)	u (m s ⁻¹)	Z × 10 ⁻⁵ (g cm ⁻² s ⁻¹)	b _{ad} × 10 ¹¹ (cm ² dyne ⁻¹)	L _r × 10 ¹⁰ (A ⁰)	-φ _k × 10 ⁸	R × 10 ⁻²	S _n × 10 ⁻³
0.0003	0.9370	1581.0	1.4814	4.2695	1.2938	3711.0	29.313	-51.011
0.0004	0.9376	1600.0	1.5002	4.1662	1.2780	3049.1	29.413	-36.295
0.0006	0.9387	1632.7	1.5326	3.9966	1.2517	2324.8	29.582	-22.046
0.0008	0.9396	1680.7	1.5792	3.7678	1.2154	2034.9	29.845	-14.385
0.0010	0.9405	1720.4	1.6181	3.5923	1.1867	1807.9	30.055	-10.185
0.0011	0.9409	1746.7	1.6435	3.4834	1.1686	1744.2	30.196	-8.518
0.0012	0.9415	1762.1	1.6590	3.4207	1.1580	1653.7	30.268	-7.409
0.0013	0.9405	1785.7	1.6795	3.3344	1.1433	1588.1	30.437	-6.370
0.0014	0.9394	1834.9	1.7237	3.1619	1.1134	1593.1	30.752	-5.023
0.0016	0.9405	1851.9	1.7417	3.1005	1.1025	1435.8	30.815	-4.096
0.0018	0.9412	1860.5	1.7511	3.0696	1.0970	1295.2	30.845	-3.506
0.0020	0.9418	1886.8	1.7770	2.9826	1.0813	1210.5	30.975	-2.828

4.2.2 Micellar features

The plots of acoustic parameters viz. u, Z, β and L_r against complex concentration are characterised by an intersection of two straight lines (Figures 1 to 4). Before CMC, there is slight curvature and after CMC, the value of acoustic parameters are

in a linear manner which corresponds to the CMC of complex. It is obvious from Table 7 that the CMC is depending upon the composition of the solvent mixture. Like other copper soap and their urea complexes, these complex also obeys the equation $u = u_0 + Gc$. The values of G and u_0 were evaluated from the u vs. c plots and are recorded in Table 8.

Table 7: CMC values of copper urea complexes derived from mustard and soybean oils in 20% and 40% methanol-benzene mixture.

Plot	CMU ₂₀	CSoU ₂₀	CMU ₄₀	CSoU ₄₀
u vs. c	0.0010	0.0013	0.0011	0.0014
Z vs. c	0.0010	0.0013	0.0011	0.0014
b_{ad} vs. c	0.0010	0.0013	0.0011	0.0014
L_f vs. c	0.0010	0.0013	0.0011	0.0014
f_k vs. ϕ_c	0.0010	0.0013	0.0011	0.0014

Table 8: Values of various constants obtained from Masson's equation for copper urea complexes derived from mustard and soybean oils in 20% and 40% methanol-benzene mixture.

Name of the soap	$U \text{ m s}^{-1}$	$G \times 10^{-5}$	$f_{k1}^0 \times 10^8$	$f_{k2}^0 \times 10^8$	$S_{k1} \times 10^8$	$S_{k2} \times 10^8$
CMU ₂₀	1533.5	0.3838	-39.0	-23.8	1.8040	0.9004
CSoU ₂₀	1526.5	0.4452	-35.0	-21.0	1.8807	0.8390
CMU ₄₀	1435.0	0.2867	-23.2	-14.6	1.0355	0.1853
CSoU ₄₀	1515.0	0.3639	-31.2	-30.2	1.3763	1.1106

This obviously suggests that the behaviour of complex in benzene-dominated environment and methanol dominated environment is quite different as they occupy different position in palisade layers of soap and complex agglomerations.^{29,30}

With regards to solvent mixture u , Z and ϕ_k increases with the predominance of methanol in 40% methanol-benzene solvent mixture whereas decreases with the predominance of benzene in 20% methanol-benzene mixture. The results are vice-versa for β and L_f for copper soap-urea complex system (Tables 3 to 6).^{31,32}

The values of CMC of ultrasonic parameters are higher in 40% methanol-benzene mixture as compared to 20% methanol-benzene mixture. With regards to chain length of the soap segment of the complex molecules, the CMC follows the order:

$$\text{CSoU} > \text{CMU}$$

The plots of apparent molar compressibility ϕ_k against square root of complex concentration \sqrt{c} are characterised by an intersection of two straight lines followed by convex curve (Figure 5) and after CMC, the value of ϕ_k increases near the CMC. The values of ϕ_k^0 are evaluated from the intercept of the plots of ϕ_k vs. \sqrt{c} and values of S_k are calculated from the slope of these plots and are recorded in (Table 8).

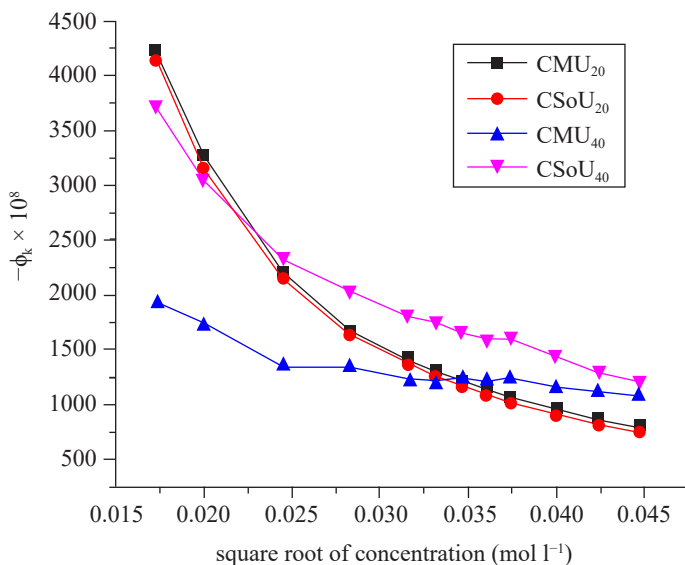


Figure 5: Plots of ϕ_k vs. \sqrt{c} of copper soap urea complex derived from mustard and soybean in 20% and 40% methanol-benzene mixture.

The order of these parameters is as follows:

$$\phi_{k1}^0 > \phi_{k2}^0 \quad \text{and} \quad S_{k2} < S_{k1}$$

This suggests that solute-solvent interaction is more prominent in concentrate solution, i.e., above CMC as compared to below CMC. The value of ϕ_k is higher in 40% methanol-benzene mixture as compared to 20% methanol-benzene mixture. The value of ϕ_k is negative for all the copper urea complexes.

5. CONCLUSION

The nonlinear variation of apparent molar compressibility and other thermoacoustical parameters with molar concentration of copper soap-urea complex derived from mustard and soybean oils with 20% and 40% methanol-benzene mixture shows the intermolecular interaction occurs in constituent molecules.

This provides useful information about the nature of intermolecular forces existing in the ternary mixture. The change in the behaviour below and above the CMC observed is attributed to the fact that aggregation of the soap-complex molecules is different in the ternary mixture of various concentrations which suggests that the solute-solvent interaction is more prominent in concentrate solution, i.e., above CMC as compared to below CMC. The value of ϕ_k is higher in 40% methanol-benzene mixture as compared to 20% methanol-benzene mixture. The negative ϕ_k values may be due to the loss of compressibility of solvent due to electrostatic forces in the vicinity of ions.

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