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### Conductometric, Viscometric and Ultrasonic Studies of Terbium Laurate in Benzene-Methanol Mixture

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# CONDUCTOMETRIC, VISCOMETRIC AND ULTRASONIC STUDIES OF TERBIUM LAURATE IN BENZENE-METHANOL MIXTURE

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The conductometric, viscometric and ultrasonic velocity measurements of the solutions of terbium laurate in benzene-methanol mixture (7:3 v/v) were carried out at  $40 \pm 0.05^\circ\text{C}$ . The conductivity results showed that the soap behaves as a weak electrolyte in dilute solutions. The viscosity results were explained on the basis of equations proposed by Einstein, Vand, Moulik and Jones-Dole and the values of molar volume evaluated by using Einstein's and Vand's equations were in close agreement. The ultrasonic velocity measurements were used to evaluate various acoustic parameters. The results showed that the soap-soap interactions are weaker than soap-solvent interactions in dilute solutions and the soap molecules do not aggregate below the critical micelle concentration in dilute solutions.

KEY WORDS: Terbium laurate, viscosity, conductivity, acoustic parameters, CMC.

## INTRODUCTION

Recently, the study of metallic soaps is becoming increasingly important in technological as well as in academic fields. While major developments have taken place in the studies of alkali, alkaline earth and transition metal soaps, the investigations on lanthanide and actinide soaps have remained almost untouched with the result that only few references are available in this relatively unexplored field<sup>1-26</sup>.

The present work deals with the conductometric, viscometric and ultrasonic measurements of terbium laurate in benzene-methanol (7:3 v/v) mixture. The selection of the solvent mixture is based on the fact that the soap possesses maximum solubility in the solvent mixture of this composition.

## EXPERIMENTAL

All the chemicals used were of BDH/AR grade. Terbium laurate was prepared by direct metathesis of potassium laurate with slight excess of required amount of aqueous solution of terbium acetate at  $50-60^\circ\text{C}$  under vigorous stirring. The excess of

potassium laurate, terbium acetate or lauric acid was removed by washing with distilled water and acetone. The soap was recrystallized with benzene-methanol mixture and dried under reduced pressure. The purity of soap was confirmed by determination of its melting point, 108°C and elemental analysis. The absence of hydroxyl group in the soap molecule was confirmed by the absence of absorption maxima in the region of 3500–3000 cm<sup>-1</sup> in its IR spectrum.

The conductances of the soap solutions were measured with a digital conductivity meter (Toshniwal CL.01.10A) and a dipping type conductivity cell with platinized electrodes. The accuracy of the results was ± 0.5%. The densities of the solutions were measured with a dilatometer calibrated with pure benzene. The volume of the dilatometer was about 15 ml and the accuracy of the density results was ± 0.0001 g ml<sup>-1</sup>. The viscosity of the soap solutions were measured with an Ostwald's type viscometer. The probable error of viscosity results was below ± 0.3%. A multifrequency interferometer M-83 (Mittal-Enterprises, New Delhi) was used to measure the ultrasonic velocity in the solvent mixture and soap solutions. The uncertainty of velocity measurements was ± 0.2%. All measurements were made at a constant temperature (40 ± 0.05°C) in a thermostat.

The viscosity ( $\eta$ ), adiabatic compressibility ( $\beta$ ), specific acoustic impedance ( $Z$ ), intermolecular free length ( $L_f$ ) and apparent molar compressibility ( $\phi_k$ ) were calculated by using the relationships:

$$\eta/\eta_0 = \rho \cdot t / \rho_0 t_0 \quad (1)$$

$$\beta = v^{-2} \cdot \rho^{-1} \quad (2)$$

$$Z = v \cdot \rho \quad (3)$$

$$L_f = (\beta/K)^{1/2} \quad (4)$$

$$\phi_k = \frac{1000}{C \cdot \rho_0} (\rho_0 \beta - \beta_0 \rho) + \frac{M \beta_0}{\rho_0} \quad (5)$$

where  $\eta_0, \eta, \rho_0, \rho, t_0, t$  and  $\beta_0, \beta$  are viscosity, density, time of flow and compressibility for solvent and solution, respectively;  $K, v$  and  $M$  are the temperature dependent Jacobson's constant, ultrasonic velocity and molecular weight of the soap, respectively.

## RESULTS AND DISCUSSION

### Conductivity

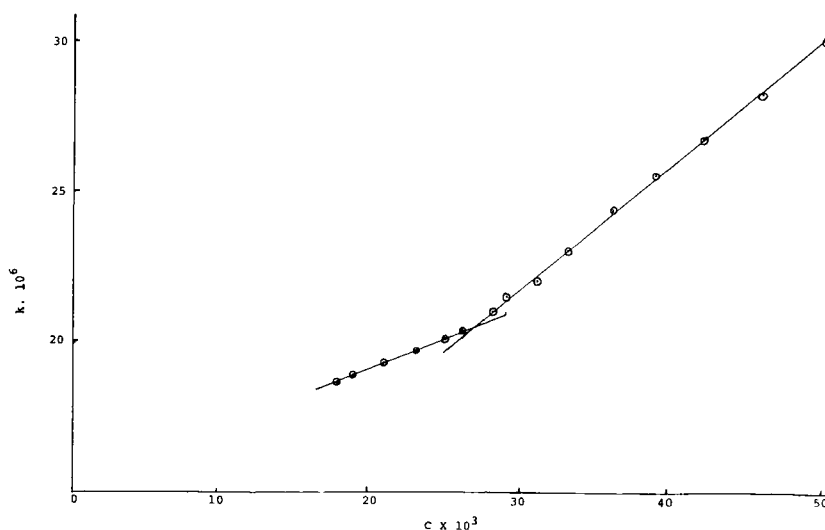
The specific conductance ( $k$ ) of the solutions of terbium laurate in benzene-methanol mixture (7:3 v/v) increases with increasing soap concentration (Tab. 1) which may be due to the ionization of the soap into simple metal cations, ( $Tb^{3+}$ ) and fatty acid anions, ( $C_{11}H_{23}COO^-$ ) in dilute solutions and due to the aggregation of ionic micelles at

**Table 1** Conductivity of Terbium Laurate in Benzene-Methanol Mixture (7:3 v/v) at  $40 \pm 0.05^\circ\text{C}$ .

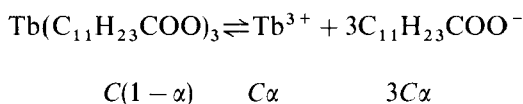
S.No.	$C \cdot 10^3$ ( $\text{mol l}^{-1}$ )	$k \cdot 10^6$ ( $\text{mho cm}^{-1}$ )	$\mu$ ( $\text{mho cm}^2 \text{mol}^{-1}$ )	$\mu^3 C^3 \cdot 10^6$	$1/\mu$	$\alpha$	$k \cdot 10^5$
1.	50.0	30.0	0.600	27.0	1.66	0.323	5.43
2.	45.6	28.3	0.618	22.4	1.62	0.332	4.54
3.	41.7	26.8	0.642	19.5	1.55	0.345	4.24
4.	38.5	25.6	0.657	16.2	1.52	0.353	3.69
5.	35.7	24.4	0.681	14.4	1.47	0.366	3.46
6.	33.3	23.0	0.691	12.2	1.45	0.371	3.00
7.	31.3	22.0	0.706	10.8	1.42	0.380	2.80
8.	29.4	21.5	0.751	9.9	1.37	0.393	2.70
9.	27.8	21.0	0.752	9.1	1.33	0.404	2.60
10.	26.3	20.4	0.776	8.5	1.29	0.417	2.54
11.	25.0	20.1	0.804	8.1	1.24	0.432	2.58
12.	22.7	19.7	0.868	7.7	1.15	0.467	2.82
13.	20.8	19.3	0.928	7.2	1.08	0.498	2.97
14.	19.2	18.9	0.984	6.7	1.02	0.529	3.18
15.	17.9	18.6	1.040	6.5	0.96	0.559	3.43

higher soap concentrations. The plot of specific conductance Vs soap concentration shows a break at a definite soap concentration, ( $0.027 \text{ mol l}^{-1}$ ) which corresponds to the CMC (Fig. 1). The value is in agreement with the values of CMC ( $0.034$  and  $0.033 \text{ mol l}^{-1}$ ) for cerium caprylate and samarium caprylate, respectively in benzene-methanol mixture (6:4 v/v) at  $40 \pm 0.05^\circ\text{C}$ <sup>26</sup>.

The molar conductance ( $\mu$ ) of the soap solutions decreases with increasing soap concentration in dilute solutions due to the combined effects of ionic atmosphere,


**Figure 1** Specific Conductance ( $k$ ) Vs Soap Concentration ( $C$ ).

solvation of ions, decrease of mobility and ionization and formation of micelles. The plot of  $\mu$  Vs  $C^{1/2}$  is concave upwards indicating that the soap behaves as a weak electrolyte in dilute solutions. Hence, an expression for dissociation of terbium laurate can be developed using Ostwald's dilution law:



where  $C$  and  $\alpha$  are the concentration and degree of dissociation of soap, respectively. The dissociation constant ( $K$ ) can be written as:

$$K = \frac{\{\text{Tb}^{3+}\}\{\text{C}_{11}\text{H}_{23}\text{COO}^-\}^3}{\{\text{Tb}(\text{C}_{11}\text{H}_{23}\text{COO})_3\}} = \frac{27C^3 \cdot \alpha^4}{1 - \alpha} \quad (6)$$

Since the ionic concentrations are low and the interionic effects being almost negligible, the solutions will not deviate appreciably from the ideal behaviour and so the activities of ions can be taken as almost equal to their concentrations. The degree of dissociation, ( $\alpha$ ) may be replaced by conductance ratio,  $\mu/\mu_0$  where  $\mu$  and  $\mu_0$  are the molar conductances at finite and infinite dilutions, respectively. On substituting the value of  $\alpha$  and rearranging, Eqn. (6) can be written as:

$$\mu^3 C^3 = \frac{K\mu_0^4}{27\mu} - \frac{K\mu_0^3}{27} \quad (7)$$

The values of  $K$  and  $\mu_0$  were obtained from the slope ( $K\mu_0^4/27$ ) and intercept ( $-K\mu_0^3/27$ ) of the linear portion of the plot of  $\mu^3 C^3$  Vs  $1/\mu$  and were found to be  $2.3 \times 10^{-5}$  and  $1.86 \text{ mho cm}^2 \text{ mol}^{-1}$ , respectively.

The values of degree of dissociation ( $\alpha$ ) lie in the range of 0.32 to 0.56 indicating that the soap behaves as a weak electrolyte in dilute solutions.

## DENSITY AND VISCOSITY

The density of soap solutions increases with the increase in soap concentration and the results have been explained in terms of Root's equation:

$$\rho = \rho_0 + AC - BC^{3/2} \quad (8)$$

where  $\rho_0$  and  $\rho$  are the densities of the solvent and soap solution, respectively. The constants  $A$  and  $B$  refer to soap-solvent and soap-soap interactions, respectively. The values of constants  $A$  (0.24) and  $B$  (0.16) were obtained from the intercept and slope of the plot  $(\rho - \rho_0)/C$  Vs  $C^{1/2}$ . The results confirm that the soap-solvent interaction is larger than the soap-soap interaction in dilute soap solutions.

The viscosity ( $\eta$ ) and specific viscosity ( $\eta_{sp}$ ) of the soap solutions in benzene-methanol mixture (7:3 v/v) increase with the increase in soap concentration. The plot of  $\eta$  Vs  $C$  is characterized by an intersection of two lines at a definite soap concentration, ( $0.028 \text{ mol l}^{-1}$ ) which corresponds to the CMC of soap and the value is in agreement with that obtained from the conductivity measurements. The viscosity results show that the soap molecules do not aggregate appreciably below the CMC whereas there is a marked change in the aggregation at the CMC of the soap.

The viscosity results have been interpreted on the basis of equations proposed by Einstein<sup>27</sup>, Vand<sup>28</sup>, Moulik<sup>29</sup> and Jones-Dole<sup>30</sup>. The values, 0.54 and  $0.51 \text{ l mol}^{-1}$  for molar volume ( $\bar{V}$ ) of the soap evaluated from the equations of Einstein and Vand were found to be consistent indicating the applicability of both of these equations below the CMC. The linearity of the plot of  $(\eta/\eta_0)^2$  Vs  $C^2$  below the CMC suggests that the Moulik's equation holds good for dilute soap solutions. The values of Moulik's constants,  $M$  and  $K$  obtained from the plot were found to be 1.003 and 17.11, respectively. The constants  $A$  and  $B$  of Jones-Dole's equation (plot of  $\eta_{sp}/C^{1/2}$  Vs  $C^{1/2}$ ) signify soap-soap and soap-solvent interactions, respectively and the larger value of  $B$  (0.47) as compared to  $A$  (0.018) for dilute soap solutions below the CMC substantiates the fact that the soap-solvent interaction is larger in dilute solutions and the soap molecules begin to aggregate at the CMC. The results are in agreement with those obtained for other lanthanide soaps<sup>31,32</sup>. The viscosity and density results were also found to be in accordance with each other.

## ULTRASONIC MEASUREMENTS

The ultrasonic-velocity ( $v$ ) of the solutions of terbium laurate in benzene-methanol mixture (7:3 v/v) increases with the increase in the soap concentration. The plot of  $v$  Vs  $C$  indicates a break at a definite soap concentration, ( $0.027 \text{ mol l}^{-1}$ ) which corresponds to the CMC of the soap. The plot of  $v$  Vs  $C$  was extrapolated to the zero soap concentration and the extrapolated value of ultrasonic velocity,  $v_0$

**Table 2** Density and Viscosity of Terbium Laurate in Benzene-Methanol (7:3 v/v) at  $40 \pm 0.05^\circ\text{C}$ .

S.No.	$C \cdot 10^3$ ( $\text{mol l}^{-1}$ )	$\rho$ ( $\text{g. ml}^{-1}$ )	$\eta$ ( $\text{Pa.s.}$ )	$\eta_{sp} \cdot 10^3$	$l$	$(\eta/\eta_0)^2$
					$\log(\eta/\eta_0)$	
1.	5.0	0.8002	0.4809	3.3	690.39	1.007
2.	10.0	0.8004	0.4819	5.6	414.70	1.011
3.	15.0	0.8014	0.4832	8.4	276.85	1.017
4.	20.0	0.8018	0.4848	10.6	218.80	1.022
5.	25.0	0.8022	0.4853	12.8	181.80	1.026
6.	30.0	0.8030	0.4865	16.1	193.74	1.032
7.	35.0	0.8039	0.4889	20.2	116.02	1.040
8.	40.0	0.8042	0.4908	23.4	99.61	1.047
9.	45.0	0.8060	0.4923	27.3	85.54	1.055
10.	50.0	0.8071	0.4952	31.2	74.99	1.063

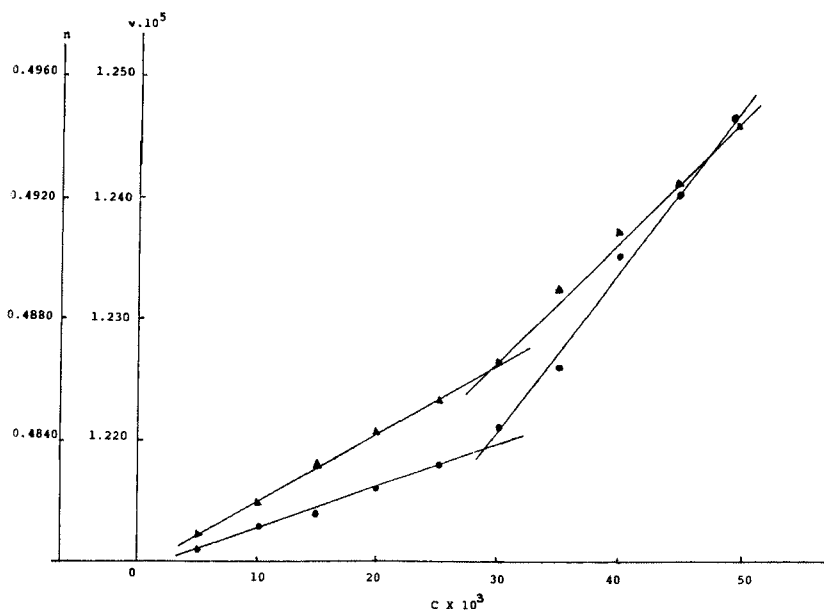


Figure 2 Viscosity,  $\blacktriangle$  ( $n$ ) and Ultrasonic velocity,  $\bullet$  ( $v$ ) Vs Soap Concentration ( $C$ ).

Table 3 Ultrasonic Velocity and Acoustic Parameters of Terbium Laurate in Benzene-Methanol (7:3 v/v) at  $40 \pm 0.05^\circ\text{C}$ .

S.No.	$C \cdot 10^3$ ( $\text{moll}^{-1}$ )	$v \cdot 10^{-5}$ ( $\text{cm s}^{-1}$ )	$\beta \cdot 10^{11}$ ( $\text{cm}^2 \text{dyne}^{-1}$ )	$L_f$ ( $\text{\AA}$ )	$Z \cdot 10^{-5}$ (C. G. S.)	$-\phi_k \cdot 10^7$ ( $\text{cm}^2 \text{dyne}^{-1}$ )
1.	5.0	1.211	8.53	11.36	9.69	29.40
2.	10.0	1.213	8.50	11.34	9.71	14.73
3.	15.0	1.214	8.48	11.32	9.72	9.84
4.	20.0	1.216	8.43	11.30	9.75	7.52
5.	25.0	1.218	8.40	11.27	9.77	6.03
6.	30.0	1.221	8.35	11.24	9.80	5.13
7.	35.0	1.240	8.28	11.19	9.85	4.55
8.	40.0	1.235	8.15	11.11	9.93	4.10
9.	45.0	1.240	8.07	11.05	9.99	3.95
10.	50.0	1.246	7.98	11.00	10.06	3.71

( $1.209 \times 10^5 \text{ cm s}^{-1}$ ) was found to be in close agreement with the experimental value of the velocity of the solvent mixture, ( $1.199 \times 10^5 \text{ cm s}^{-1}$ ). The variation of ultrasonic velocity, ( $v$ ) with the soap concentration ( $C$ ) for dilute soap solutions below the CMC follows the relationship:

$$v = v_0 + GC$$

where  $G$  is Garnsey's constant<sup>33</sup>. The value of Garnsey's constant obtained from the slope of the plot of  $v$  Vs  $C$  below the CMC was found to be  $4.0 \times 10^6 \text{ cm}^2 \text{mol}^{-1} \text{ s}^{-1}$ .

The adiabatic compressibility ( $\beta$ ) of the solutions of terbium laurate decreases with increasing soap concentration and the decrease may be attributed to the fact that the soap molecules in dilute solutions behave as weak electrolyte and are ionised into terbium cations ( $\text{Tb}^{3+}$ ) and fatty acid anions ( $\text{C}_{11}\text{H}_{23}\text{COO}^-$ ) as indicated by the conductivity measurements. The ions are surrounded by a layer of solvent molecules firmly bound and oriented towards the ions. The orientation of solvent molecules around the ions is due to the influence of the electrostatic field of ions and results in the increase in internal pressure and decrease in the compressibility ( $\beta$ ) of the solutions i.e. the solutions become harder to compress<sup>34</sup>.

The adiabatic compressibility ( $\beta$ ) of the soap solutions is found to obey Bachem's relationship<sup>35</sup>:

$$\beta = \beta_0 + AC + BC^{3/2} \quad (8)$$

where  $\beta_0$  is the compressibility of the solvent,  $C$  is the soap concentration and  $A$  and  $B$  are constants. The values of  $A$  ( $-5.3 \times 10^{11}$ ) and  $B$  ( $19.0 \times 10^{11}$ ) were obtained from the intercept and slope of the plot of  $(\beta - \beta_0)/C$  Vs  $C^{1/2}$ .

The values of the specific acoustic impedance ( $Z$ ) increase<sup>36</sup> while of intermolecular free length, ( $L_f$ ) decrease<sup>37</sup> with increasing soap concentration for the solutions of terbium laurate in benzene-methanol mixture. The plots of  $Z$  Vs  $C$  and  $L_f$  Vs  $C$  were extrapolated to zero soap concentration and the extrapolated values obtained for  $Z$  and  $L_f$  were found to be  $9.45 \times 10^5$  and  $36.10 \text{ \AA}$ , respectively. The increase in the values of specific acoustic impedance ( $Z$ ) with soap concentration ( $C$ ) can be explained on the basis of lyophobic interaction between the soap and solvent molecules which increases the intermolecular distance making relatively wider gaps between the molecules and thus becoming the main cause of impedance in propagation of ultrasound waves.

The apparent molal compressibility ( $\phi_k$ ) decreases with increase in soap concentration. It follows from the Debye-Huckel's theory that the apparent molal compressibility ( $\phi_k$ ) is related to the concentration ( $C$ ) by the relationship:

$$\phi_k = \phi_k^0 + S_k C^{1/2}$$

where  $\phi_k^0$  is the apparent molal compressibility of the solvent and  $S_k$  is a constant. The values of the  $S_k$  and  $\phi_k$  were obtained from the slope and intercept of the plot of  $\phi_k$  Vs  $C^{1/2}$  and were found to be  $1.8 \times 10^{-5}$  and  $-38.40 \times 10^7 \text{ cm}^2 \text{ dyne}^{-1}$ , respectively.

The results of ultrasonic velocity measurements show that the soap behaves as a simple weak electrolyte in dilute solutions. The results confirm that there is a significant interaction between the soap and solvent molecules in dilute soap solutions and the soap molecules do not aggregate appreciably in dilute solutions below the CMC.

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