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Ultrasonic and Viscosimetric Studies of Samarium Laurate in Benzene-Dimethylsulfoxide mixtures

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Summary. Ultrasonic and viscosity measurements of samarium laurate in benzene-*DMSO* mixtures of different compositions (7:3 and 1:1 V/V) have been used to determine the critical micelle concentration (CMC), soap-solvent interaction, and various acoustic parameters of the system. The values of critical micelle concentration increase with increasing amount of *DMSO* in the solvent mixtures. The viscosity results have been explained on the basis of equations proposed by *Einstein*, *Vand. Moulik*, and *Jones-Dole*. The values of CMC for samarium laurate obtained from the viscosity measurements are in agreement with the results obtained from ultrasonic measurements. The results show that the soap molecules do not aggregate appreciably below CMC; there is a marked change in the aggregation behaviour at CMC.

Keywords. Samarium laurate; Micellization; Ultrasonic velocity; Acoustic parameters; Viscosity.

Ultraschall- und Viskositätsmessungen an Samariumlaurat in Benzol-*DMSO*-Mischungen

Zusammenfassung. Mit Hilfe von Ultraschall- und Viskositätsmessungen an Samariumlaurat in Benzol-*DMSO* – Mischungen verschiedener Zusammensetzung (7:3 und 1:1 v/v) wurden die kritische Micellenkonzentration (CMC), Seife-Lösungsmittel-Wechselwirkungen und verschiedene akustische Parameter des Systems bestimmt. Die Werte für die kritische Micellenkonzentration steigen mit wachsendem Anteil von *DMSO* im Lösungsmittelgemisch. Die Ergebnisse der Viskositätsmessungen werden auf der Basis der Gleichungen von *Einstein*, *Vand*, *Moulik* und *Jones-Dole* erklärt. Die CMC-Werte für Samariumlaurat aus Viskositäts- und Ultraschallmessungen stimmen überein. Die Ergebnisse zeigen, daß die Seifenmoleküle bei Konzentrationen unter CMC nicht stark aggregieren; bei Erreichen des CMC-Werts tritt eine ausgeprägte Änderung im Aggregationsverhalten ein.

Introduction

Recently, the metallic soaps are becoming increasingly important in technological and academic fields. The alkali, alkaline earth, and transition metal soaps have been thoroughly investigated, but the lanthanide and actinide soaps have remained almost unexplored in spite of their widespread applications in various industries [1–11]. The physico-chemical characteristics and structures of soaps can be controlled up to a certain extent by the methods and conditions of their preparation.

The studies of metallic soaps are therefore of great significance for their applications in various industries under different conditions.

The present work deals with the ultrasonic velocity and viscosity measurements of solutions of samarium laurate in mixtures of benzene-dimethylsulfoxide of different compositions (7:3 and 1:1 V/V). The results have been used to determine the critical micelle concentration (CMC), soap-solvent and soap-soap interactions, and various parameters related to the acoustical properties of the solutions.

Results and Discussion

Micellization in organic solvents

The micellization process in organic solvents is somewhat different from that in aqueous solutions. The main cause of micellization in organic solvents is the energy change due to dipole-dipole interactions between the polar head groups of soap molecules. The aggregation begins at very low concentrations in organic solvents and results in the formation of very much smaller aggregates than in H_2O. The association in organic solvents can be described in terms of a stepwise association model [16–19]. The molecules of soap are characterized by the presence of both lyophilic and lyophobic moieties in the same molecule, and the micelles in organic solvents can be visualized as *Hartley's Inverted Micelles* in which polar head groups are present in the centre of the micelles with the hydrocarbon chains extending outwards into the solvent. The determination of CMC in organic solvents cannot be carried out by the methods commonly used for aqueous solutions as the association starts at very low concentrations. Therefore, the ultrasonic velocity and viscosity measurements were used to determine the CMC value and various other parameters.

Ultrasonic measurements

The variation of ultrasonic velocity with soap concentration follows the relationship

$$v = v_0 + GC,$$

where v_0 is the ultrasonic velocity in the solvent and G is *Garnsey*'s constant [20]. The plots (Fig. 1) of ultrasonic velocity *vs.* soap concentration are characterized by an intersection of two straight lines at a definite soap concentration which corresponds to the CMC value of the soap. These values were found to be 5.0×10^{-3} *M* and 6.8×10^{-3} *M* for samarium laurate in mixtures of benzene-*DMSO* of compositions of 7:3 and 1:1 (V/V), respectively. The values increase with increasing amount of *DMSO* in the solvent. The micellization is mainly caused by the energy change due to dipole-dipole interactions between the polar head groups of soap molecules. The micelles are held together by *Van der Waals* forces acting between hydrocarbon chains of parallel layers and by strong dipole-dipole interactions between polar heads. The extrapolated values of v_0 (1.2466×10^3 and 1.2729×10^3 $\mathrm{m \cdot s^{-1}}$) for solutions in benzene-*DMSO* mixtures of compositions of 7:3 and 1:1 (V/V) were found to be in close agreement with the velocities in the pure solvent mixtures (1.2475×10^3 and $1.2735 \times 10^3\,\mathrm{m \cdot s^{-1}}$, respectively), indicating that the soap molecules do not aggregate up to an appreciable extent below CMC. The values of

Garnsey's constant were found to be 1.36×10^3 and 1.13×10^3 for the solvent mixtures in question.

The values of the adiabatic compressibility (β) of the solutions of samarium laurate decrease with increasing soap concentration (Tables 1 and 2). The decrease in adiabatic compressibility is attributed to the fact that the soap molecules in dilute solutions are considerably ionized into samarium cation (Sm^{3+}) and laurate anions ($C_{11}H_{23}COO^-$). 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 attributed to the influence of the electrostatic field of the ions. Thus, the internal pressure increases which lowers the compressibility of the solutions, *i.e.* the solutions become harder to compress [21].

The adiabatic compressibility of the soap solutions is found to obey *Bachem*'s relationship [22]:

$$\beta = \beta_0 + AC - BC^{3/2}$$

A and B are constants, C is the molar concentration of the soap, and β_0 is the adiabatic compressibility of the solvent mixture. The values of constants A (-4.6×10^{-10} and -140×10^{-10}) and B (15.4×10^{-10} and 40.0×10^{-10}) were obtained from the intercept and the slope of the plots of $[\beta - \beta_0/C]$ *vs.* $C^{1/2}$ for the solutions investigated.

It follows from *Debye-Hückel*'s theory that the apparent molar compressibility, ϕ_k, is related to the molar concentration of the soap by the relationship

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

where ϕ_k^0 is the apparent molar compressibility of the solvent ($m^5N^{-1}kg^{-1}mol^{-1}$) and S_k is a constant. The values of ϕ_k^0 (-1.020×10^6 and -2.040×10^6) and S_k(-6.0×10^6 and $+10.6 \times 10^6$) for the solutions were evaluated from the intercept and the slope of the linear plots of ϕ_k *vs.* $C^{1/2}$ below CMC.

The intermolecular freelength (L_f) decreases, whereas the specific acoustic impedance (Z) increases with increasing soap concentration. This indicates that there is a significant interaction between soap and solvent molecules [23–27] due to which the structural arrangement is considerably affected. The increase in the

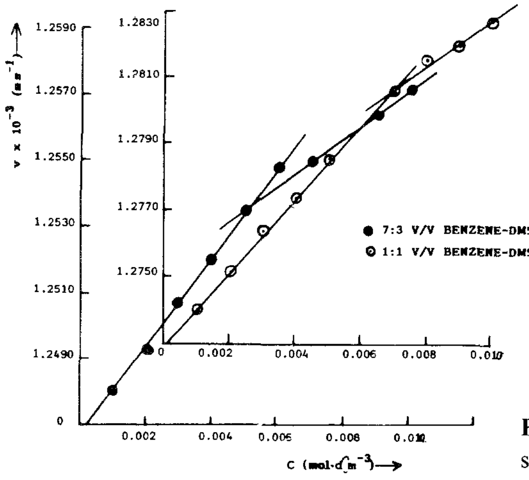

Fig. 1. Ultrasonic velocity *vs.* concentration of samarium laurate

Table 1. Ultrasonic velocity and acoustic parameters of samarium laurate in benzene-*DMSO* (7:3 V/V) at 40 ± 0.05 °C; for symbols, see text

	C (mol·dm^{-3})	$\rho \times 10^{-3}$ (kg·m^{-3})	$v \times 10^{-3}$ (m.s^{-1})	$\beta \times 10^{9}$ (m^2·N^{-1})	$L_f \times 10^{10}$ (Å)	$Z \times 10^{-6}$ (kg·m^{-2}·s^{-1})	$-(\beta-\beta_0)/C \times 10^{10}$	S_n	$-\phi_k \times 10^{6}$ (m^5·N^{-1}·kg^{-1}·mol^{-1})	$R \times 10^{4}$ (m^3/mol·(N/m^2)$^{1/3}$)	$W \times 10^{3}$ (m^3/mol·(N/m^2)$^{1/7}$)
1	0.001	1.0569	1.2480	0.6075	0.3032	1.3190	9.0	19.98	1.449	7.9586	1.5323
2	0.002	1.0572	1.2492	0.6061	0.3029	1.3206	11.5	25.53	1.296	7.9572	1.5322
3	0.003	1.0577	1.2506	0.6045	0.3025	1.3227	13.0	28.86	1.350	7.9564	1.5321
4	0.004	1.0582	1.2520	0.6028	0.3021	1.3248	14.0	31.00	1.402	7.9556	1.5320
5	0.005	1.0587	1.2535	0.6011	0.3016	1.3271	14.6	32.41	1.433	7.9550	1.5319
6	0.006	1.0584	1.2548	0.6001	0.3014	1.3280	13.8	30.71	1.261	7.9600	1.5323
7	0.007	1.0581	1.2550	0.6000	0.3013	1.3279	12.0	26.64	1.001	7.9627	1.5332
8	0.008	1.0577	1.2556	0.5999	0.3013	1.3280	10.6	23.59	0.813	7.9670	1.5338
9	0.009	1.0574	1.2564	0.5991	0.3011	1.3285	10.3	22.94	0.744	7.9701	1.5345
10	0.010	1.0571	1.2572	0.5987	0.3010	1.3289	9.9	21.94	0.669	7.9749	1.5351

Table 2. Ultrasonic velocity and acoustic parameters of samarium laurate in benzene-*DMSO* (1:1 V/V) at 40 ± 0.05 °C; for symbols, see text

	C (mol·dm^{-3})	$\rho \times 10^{-3}$ (kg·m^{-3})	$v \times 10^{-3}$ (m.s^{-1})	$\beta \times 10^{9}$ (m^2·N^{-1})	$L_f \times 10^{10}$ (Å)	$Z \times 10^{-6}$ (kg·m^{-2}·s^{-1})	$-(\beta-\beta_0)/C \times 10^{10}$	S_n	$-\phi_k \times 10^{6}$ (m^5·N^{-1}·kg^{-1}·mol^{-1})	$R \times 10^{4}$ (m^3/mol·(N/m^2)$^{1/3}$)	$W \times 10^{3}$ (m^3/mol·(N/m^2)$^{1/7}$)
1	0.001	1.1085	1.2740	0.5558	0.2900	1.4122	14.0	35.59	2.030	7.6388	1.4797
2	0.002	1.1089	1.2752	0.5545	0.2897	1.4141	13.5	34.32	1.577	7.6385	1.4796
3	0.003	1.1094	1.2764	0.5533	0.2894	1.4160	13.0	33.05	1.410	7.6374	1.4795
4	0.004	1.1098	1.2774	0.5622	0.2891	1.4176	12.5	31.78	1.289	7.6367	1.4794
5	0.005	1.1102	1.2786	0.5610	0.2888	1.4195	12.4	31.52	1.236	7.6363	1.4793
6	0.006	1.1106	1.2798	0.5497	0.2884	1.4231	12.5	31.78	1.217	7.6359	1.4792
7	0.007	1.1109	1.2806	0.5489	0.2882	1.4226	11.9	30.14	1.125	7.6355	1.4791
8	0.008	1.1106	1.2816	0.5482	0.2881	1.4233	11.3	28.60	1.006	7.6395	1.4798
9	0.009	1.1101	1.2820	0.5481	0.2880	1.4231	10.1	25.70	0.8357	7.6438	1.4805
10	0.010	1.1098	1.2828	0.5476	0.2879	1.4236	9.6	24.40	0.7494	7.6474	1.4811

values of Z with increasing C can be explained on the basis of a lyophobic interaction between soap and solvent molecules which increases the intermolecular distance, opening relatively wide gaps between the molecules and becoming the main cause of impedance in the propagation of ultrasound waves. The plots of L_f *vs.* C and Z *vs.* C show a break at the CMC value of the soap. The results of adiabatic compressibility and intermolecular freelength are in agreement with the results reported for yttrium soaps [28].

The solvation number S_n decreases above CMC with increasing soap concentration which may be attributed to an enhanced incorporation of solvent molecules above CMC, reducing the repulsive forces acting between the heads of the ionic micelles. The molar sound velocity (R) and molar sound compressibility (W) increase above CMC with increasing soap concentrations.

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

Viscosity measurements

Viscosity (η) and specific viscosity (η_{sp}) of the solutions of samarium laurate in mixtures of benzene-*DMSO* of compositions of 7:3 and 1:1 (V/V) increase with increasing soap concentration (Tables 3 and 4). This may be due to the increasing tendency of the soap molecules to form aggregates with increasing soap concentration. The plots of η *vs.* C (Fig. 2) are characterized by an intersection of two straight lines at a definite soap concentration which corresponds to the critical micelle concentration of the soap. The values of CMC were found to be 5.0×10^{-3} *M* and 6.9×10^{-3} *M* at 40 ± 0.05 °C, respectively, and are in agreement with the values obtained from other micellar properties. The viscosity measurement results confirm that there is no appreciable aggregation of the soap molecules in dilute solutions, whereas there is a sudden increase in the aggregation at the CMC of the soap.

Table 3. Density and viscosity of samarium laurate in benzene-*DMSO* (7:3 V/V) at 40 ± 0.05 °C; for symbols, see text

	C (mol·dm^{-3})	$\rho \times 10^{-3}$ (kg·m^{-3})	$(\rho - \rho_0)/C$	$\eta \times 10^3$ (Pa·s)	$\eta_{sp} \times 10^3$	$\eta_{sp}/C^{1/2}$	$(\eta/\eta_0)^2$	$1/\log(\eta/\eta_0)$
1	0.001	1.0569	1.70	0.7670	6.56	0.2074	0.013	352.06
2	0.002	1.0572	1.00	0.7720	13.10	0.2930	1.026	176.60
3	0.003	1.0577	0.83	0.7770	19.68	0.3591	1.039	118.11
4	0.004	1.0582	0.75	0.7818	25.98	0.4110	1.052	89.76
5	0.005	1.0587	0.70	0.7870	32.80	0.4639	1.066	71.32
6	0.006	1.0584	0.53	0.7980	37.79	0.4876	1.077	62.06
7	0.007	1.0581	0.41	0.7935	41.33	0.4938	1.084	56.84
8	0.008	1.0577	0.31	0.7970	45.93	0.5137	1.093	51.27
9	0.009	1.0574	0.24	0.8005	50.52	0.5323	1.103	46.72
10	0.010	1.0571	0.19	0.8034	54.33	0.5933	1.112	43.52

Table 4. Density and viscosity of samarium laurate in benzene-*DMSO* (1:1 V/V) at 40 ± 0.05 °C; for symbols, see text

	C (mol·dm^{-3})	$\rho \times 10^{-3}$ (kg·m^{-3})	$(\rho - \rho_0)/C$	$\eta \times 10^3$ (Pa·s)	$\eta_{sp} \times 10^3$	$\eta_{sp}/C^{1/2}$	$(\eta/\eta_0)^2$	$1/\log(\eta/\eta_0)$
1	0.001	1.1085	2.00	0.9275	2.70	0.0854	1.005	853.10
2	0.002	1.1089	1.20	0.9306	6.05	0.1353	1.012	381.48
3	0.003	1.1094	0.96	0.9340	9.73	0.1775	1.019	237.80
4	0.004	1.1098	0.83	0.9370	12.97	0.2052	1.026	178.63
5	0.005	1.1102	0.74	0.9400	16.22	0.2294	1.033	143.15
6	0.006	1.1106	0.68	0.9430	19.46	0.2511	1.039	119.47
7	0.007	1.1109	0.62	0.9462	22.92	0.2738	1.046	101.61
8	0.008	1.1106	0.51	0.9486	25.51	0.2813	1.052	91.39
9	0.009	1.1101	0.40	0.9502	27.24	0.2870	1.055	85.66
10	0.010	1.1098	0.33	0.9530	30.24	0.3027	1.061	77.21

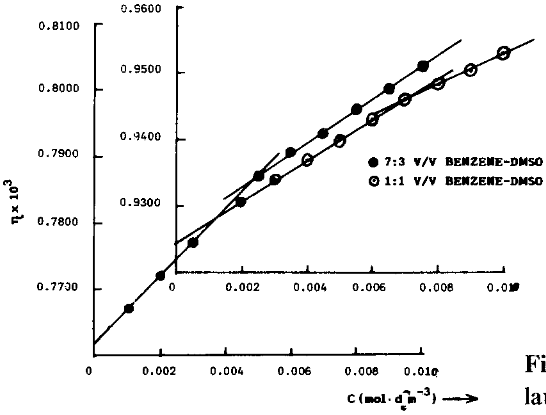

Fig. 2. Viscosity *vs.* concentration of samarium laurate

The viscosity results have been interpreted in the light of equations proposed by *Einstein* [29], *Vand* [30], *Moulik* [31], and *Jones-Dole* [32].

Einstein: $\eta_{sp} = 2.5\bar{V}C$
Vand: $1/C = (0.921/\bar{V})^{-1} \cdot 1/\log(\eta/\eta_0) + \phi\bar{V}$
Moulik: $(\eta/\eta_0)^2 = \mathrm{M} + \mathrm{K}'C^2$
Jones-Dole: $\eta_{sp}/\sqrt{C} = \mathrm{A} + \mathrm{B}\sqrt{C}$

$\bar{V}$, C, ϕ, η, η_0, and η_{sp} are the molar volume (l·mol^{-1}) and concentration (mol·dm^{-3}) of the soap, interaction coefficient, viscosity of solution, viscosity of solvent, and specific viscosity, respectively. M and K′ are *Moulik*'s constants. The constants A and B of *Jones-Dole*'s equation refer to soap-soap and soap-solvent interactions, respectively.

The plots of η_{sp} *vs.* C are linear below CMC with intercepts almost equal to zero which shows that *Einstein*'s equation is applicable to dilute solutions of samarium laurate. The value of the molar volume of the soap obtained from the slope of the plot of *Vand*'s equation (2.63 l·mol^{-1}) is in close agreement with the value obtained from *Einstein*'s equation ($2.61\,\mathrm{l\cdot mol^{-1}}$) for the solution of samarium laurate in benzene-*DMSO* 7:3.

The values of constants A (0.0 and -4.0) and B (6.5 and 3.7) of *Jones-Dole*'s equation have been calculated from the intercept and slope of the plots of $\eta_{sp}/\sqrt{C}$ *vs.* $\sqrt{C}$ below CMC, respectively. The smaller values of constant A (soap–soap interaction) with respect to B (soap-solvent iteraction) confirm that the soap molecules do not aggregate appreciably below CMC and that there is a sudden change in the aggregation behaviour at CMC.

The values of *Moulik*'s constant (1.011 and 1.003) obtained from the intercept of the plots of $(\eta/\eta_0)^2$ *vs.* C^2 are approximately constant, whereas the values of constant K′ (2.85×10^3 and 1.82×10^3), calculated from the slope of the plots of $(\eta/\eta_0)^2$ *vs.* C^2, vary with the change in the composition of the solvent mixture. The values of CMC obtained from the viscosity and ultrasonic measurements are in close agreement. It is therefore concluded that the viscosity results for dilute solutions of samarium laurate may be satisfactorily explained in terms of the equations proposed by *Einstein*, *Vand*, *Moulik*, and *Jones-Dole*.

Experimental

The chemicals used were of AR grade (E. Merck). Samarium laurate was prepared by direct metathesis of potassium laurate with a slight excess of samarium acetate solution at 50–60 °C under vigorous stirring. The precipitated soap was washed several times with distilled water and finally with acetone. The soap was recrystallized from benzene-methanol and dried under reduced pressure. The purity of the soap was checked by elemental analysis. The results agreed with the theoretically calculated values. The absence of the hydroxyl group in the soap molecules was confirmed by the absence of any absorption maxima in the region of 3500–$3000\,\mathrm{cm^{-1}}$ in the infrared spectrum of the soap. The melting point of the purified samarium laurate was 94 °C.

The freshly prepared solutions of samarium laurate containing different concentrations of soap in mixtures of benzene and *DMSO* of different compositions (7:3 and 1:1 V/V) were kept for 2 hours in a thermostat at constant temperature (40 ± 0.05 °C). The ultrasonic velocity measurements of the solutions were performed with a multifrequency Ultrasonic Interferometer (M-83 Mittal Enterprises, New Delhi) at a frequency of 1 MHz at 40 ± 0.05 °C. The uncertainty of the velocity measurements was $\pm 0.2\%$. The densities were measured with a dilatometer. The volume of the dilatometer was 15 ml and the accuracy of density results was ± 0.0001. An *Ostwald* type viscometer was used for measuring the viscosity; the accuracy of the results was $\pm 0.3\%$.

Calculations

Various acoustic parameters such as adiabatic compressibility (β), intermolecular freelength (L_f) [12], specific acoustic impedance (Z) [13], apparent molar compressibility (ϕ_k), molar sound velocity (R), molar sound compressibility (W) [14], and solvation number (S_n) [15] were calculated using the following relationships:

$$\beta = v^{-2} \cdot \rho^{-1} \qquad 1$$

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

$$Z = v \cdot \rho \qquad 3$$

$$\phi_k = \frac{1000}{C \cdot \rho_0} \cdot (\rho_0 \beta - \beta_0 \rho) + \frac{M \beta_0}{\rho_0} \qquad 4$$

$$R = \frac{\bar{M}}{\rho}\cdot v^{1/3} \quad \left(\bar{M} = \frac{n_0 M_0 + nM}{n_0 + n}\right) \qquad 5$$

$$W = \frac{\bar{M}}{\rho}\cdot \beta^{-1/7} \qquad 6$$

$$S_n = \frac{n_0}{n}\left(1 - \frac{\bar{V}\beta}{n_0 \bar{V}_0 \beta_0}\right) \qquad 7$$

v, v_0; ρ, ρ_0; β, β_0; and $\bar{V}$, $\bar{V}_0$ are the ultrasonic velocity, density, adiabatic compressibility and molar volume of solution and solvent, respectively; n, n_0 and M, M_0 are number of mole and molecular mass of the solute and solvent, respectively; K and C are the temperature dependent *Jacobson's* constant and concentration.

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