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Volumetric and viscometric studies of sodium dodecyl sulphate in aquous and in amino acid solutions at different temperatures

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Abstract: *The densities, ultrasonic velocities and viscosities of sodium dodecyl sulphate (SDS) in water and in aqueous L-cysteine solutions have been measured as a function of SDS concentration at five equidistant temperatures ranging from 293.15 K to 313.15 K. The density data were used to calculated the apparent molar volume, ϕ_v, the limiting apparent molar volume, ϕ_v^0 and experimental slopes, S_v, derived from the Masson equations. The apparent molar adiabatic compressibilies, β_s were also calculated by using both the density and ultrasonic velocity data. The viscosity data were employed to determine the viscosity B-coefficients, and the free energies, $\Delta G^{\neq}$, enthalpies, $\Delta H^{\neq}$ and entropies, $\Delta S^{\neq}$ of activation using the Nightingale and Benck, and Eyring equations. The structural properties of SDS in water and aqueous L-cysteine were studied by using* $\left[\frac{\partial^2 \varphi_v^0}{\partial T^2}\right]_p$ *and dB/dT parameters.*

Keywords: *Apparent molar volume, Adiabatic compressibility, Free energy, Viscosity coefficient, Critical micelle concentration, Sodium dodecyl sulphate, L-cysteine.*

I. Introduction

In recent years, there has been growing interest in the interactions between protein and surfactant due to their many applications in biosciences, foods and cosmetics, drug delivery, detergency, and biotechnological processes [1, 2]. Commonly used surfactant sodium dodecyl sulfate (SDS) is reported to act as a more potent protein denaturant than urea and guanidine hydrochloride [3]. It is commonly used to stabilize biological membranes and to isolate and purity membranes proteins and membrane lipid. In ionic surfactants, the repulsive forces originated primarily from electrostatic repulsion between the polar head groups [4] whereas, attractive interactions have generally been attributed to hydrophobic interactions [5] between the nonpolar tails of the surfactant monomers. Using various numbers of tools and techniques, these interactions have been studied and published in the past few years [6-19]. It has been proposed that hydrophobic and electrostatic interactions are the two main driving forces for the association between surfactants and proteins in aqueous solution. However, the study of protein-surfactant interactions is difficult because of the complexity of interactions in such a large molecule. Several details in the mode of these interactions remain unanswered. Therefore, it is very important to understand the origin and nature of these interactions both qualitatively and quantitatively. To understand the fine details, the interactions of the building blocks of the protein with surfactants must be studied. There have been some investigations on the interaction of surfactants with amino acids [20-28]. Singh *et al* [22] reported only volumetric properties of some amino acids and two peptides (diglycine and triglycine) in aqueous surfactant solutions at T = 298.15 K and Yan et al [28] reported interactions of glycyl dipeptides with sodiumdodecyl sulfate in aqueous solution by volumetric, conductometric, and fluorescence Probe Study in 298.15K to 313.15K with 5K intervals. Both have used aqueous sodiumdodecyl sulfate as solvent. However, to the best of our knowledge, no report is available in the literature on the physico-chemical properties of SDS in aqueous amino acid solutions at different temperatures.

Molecular interactions (*i.e*. solute-solvent, solute-solute, and solvent-solvent-solvent) have great importance in biological chemistry, physical chemistry, surface chemistry, environmental chemistry, and geochemistry. The partial molar volume, adiabatic compressibility and viscosity *B*-coefficient are the important physicochemical properties to understand the interactions between molecules in solution. Thus in continuation with our earlier work [29] in the present paper we report the study of the volumetric, viscometric and ultrasound

behavior of sodium dodecyl sulfate in water and in 0.1m L-cysteine. Using these data, infinite-dilution apparent molar volumes, Heplar constant, viscosity *B*-coefficients, and activation free-energy parameters are also calculated. Results are discussed in terms of the structural hydration interaction model and the transition-state theory.

II. Experimental

The Surfactant used in this study was sodium dodecyl sulphate (SDS), $C_{12}H_{25}SO_4Na$ (SDS) (purity. Mass fraction ≥0.98) and the amino acid was L-cysteine (purity. Mass fraction ≥0.99). Both of them were procured from Fluka Chemical Company, Switzerland. Supplied distilled water was redistilled and deionized by passing through two ion exchange columns. The deionized water was distilled again in alkaline $KMnO_4$ medium and used for preparation of solution. Conductivity of this water was found to be about 1.00 μS.

The conductivity measurements were carried out on a Laboratory Conductivity Meter (Model 4310 Jenway). The estimated values of critical micellar concentrations for SDS are 8.2×10^{-3}, 8.3×10^{-3}, 8.7×10^{-3}, 9.0×10^{-3}, 9.3×10^{-3} mol·kg^{-1} at T = (303.15, 308.15, 313.15, 318.15, 323,15 K) respectively and agrees with the reported values [30]. The CMC (critical micelle concentration) is determined by extrapolating the molar conductivity data in the premicellar region to intersect with a straight line drawn through the data in the postmicellar region. The molar conductivity decreases with increasing SDS concentration and show a sharp break in its value where micelle starts to form and is determined by extrapolating the molar conductivity data in the premicellar region to intersect with a straight line drawn through the data in the postmicellar region.

An electric balance with an accuracy of ± 0.0001 g was used for weighing. The flow time of solutions was recorded by an electronic stopwatch capable of reading up to 0.01 second. A constant temperature water thermostat was used for the measurements of viscosity of solution. The temperature of the thermostat was maintained constant to an accuracy of ± 0.1 K. The density meter (DMA-5000, Anton Paar, Austria) and the Cannon-Fenske Routine viscometer were used for the measurements of density and viscosity respectively.

The densities of binary mixtures (SDS + water and L-cysteine + water) and ternary mixtures (SDS + L-cysteine + water) were measured using high-precision vibrating tube digital density meter (DMA-5000, Anton, Paar, Austria). The DMA-5000 density measuring cell consists of a U-shaped oscillator glass cylinder. The temperature of the sample tube is controlled to ± 0.001 K and the accuracy in the density measurement was ± $5\cdot10^{-5}$ g·cm^{-3}. The apparatus was calibrated once a day with double-distilled water (deionized and degassed) and dry air for the temperature range investigated. Triplicate measurements were performed for each sample at each mentioned temperature. Density measurements were made on solutions of SDS in water, L-cysteine in water and also SDS in 0.1 mol kg^{-1} aqueous L-cysteine at five equidistant temperatures ranging from 293.15 K to 313.15 K.

An A-type Ostwald viscometer previously calibrated with redistilled water was used to measure the viscosities.An electronic digital stopwatch with an uncertainty of ±0.01 s was used for flow time measurements. A transparent glass walled thermostatic water bath was used, and the uncertainty in the temperature during the measurements was ±0.05 K. From the measurement of the time of fall, the viscosity coefficient, η was calculated by using the Poiseuille's equation,

$$\eta = A\rho t \qquad (1)$$

where $A = \pi r^4 hg/8lV$ is the viscometer constant at a particular temperature determined by measuring the efflux time of water. The uncertainty of η in the present experiments was less than 2×10^{-4} mPa.s. For all of the mixture compositions and the pure solvents, triplicate measurements were performed, and the average of these values was considered in all calculations.

III. Result and Discussion

The density data measured for L-cysteine in water and aqueous SDS solution at T = (293.15, 303.15, 308.15, and 313.15) K are listed in Table 1. The apparent molar volumes (φ_v) were calculated from solution densities,ρ, using the following equation

$$\varphi_v = \left[\frac{M}{\rho} - \frac{10^3(\rho-\rho_0)}{m\rho\rho_0}\right] \qquad (2)$$

Where ρ_0, M and m are the density of pure solvent, the molar mass of the solute, the molality of the solution in mol·kg^{-1}. The variation of apparent molar volumes of SDS in aqueous solutions and in 0.1m aqueous

Table 1. Concentration dependence of densities (ρ) and apparent molar volumes (φ_v) for L-cysteine, SDS and SDS in 0.1 m aqueous L-cysteine solutions at 293.15 K, 298.15 K, 303.15 K, 308.15 K, and 313.15 K.

m/mol·kg^{-1}	Density, ρ /kg m^{-3}·10^3					Apparent molal volume, φ_v /m^3·mol^{-1}·10^6				
	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K
					L-cysteine in water					
0.0500	1.000582	0.999400	0.997978	0.996347	0.994518	73.26	73.67	74.08	74.42	74.74
0.0990	1.002905	1.001705	1.000266	0.998620	0.996778	73.30	73.71	74.10	74.45	74.77
0.1496	1.005280	1.004061	1.002605	1.000944	0.999089	73.33	73.74	74.13	74.48	74.79
0.1999	1.007622	1.006386	1.004913	1.003236	1.001370	73.37	73.77	74.15	74.51	74.81
0.2499	1.009928	1.008673	1.007183	1.005493	1.003614	73.40	73.80	74.19	74.53	74.84
0.2983	1.012143	1.010869	1.009365	1.007661	1.005770	73.43	73.83	74.21	74.56	74.87
0.3500	1.014486	1.013197	1.011673	1.009957	1.008055	73.46	73.86	74.25	74.58	74.89
0.3982	1.016655	1.015350	1.013811	1.012082	1.010168	73.49	73.89	74.27	74.61	74.91
					SDS in water					
0.0003	0.998209	0.997045	0.995644	0.994026	0.992210	227.66	231.26	231.51	235.22	235.55
0.0005	0.998221	0.997057	0.995655	0.994037	0.992220	229.04	231.23	233.48	235.78	238.14
0.0020	0.998310	0.997144	0.995740	0.994120	0.992299	228.95	230.66	232.42	234.23	237.11
0.0040	0.998425	0.997255	0.995848	0.994224	0.992402	230.21	232.17	233.93	235.99	237.84
0.0060	0.998538	0.997365	0.995955	0.994327	0.992500	230.77	232.65	234.41	236.56	238.76
0.0080	0.998647	0.997472	0.996058	0.994429	0.992601	231.55	233.27	235.16	236.98	238.84
0.0100	0.998750	0.997569	0.996155	0.994524	0.992696	232.42	234.45	236.02	237.74	239.31
0.0120	0.998851	0.997670	0.996252	0.994619	0.992789	233.51	235.23	236.92	238.57	240.10
0.0140	0.998949	0.997768	0.996348	0.994715	0.992884	234.42	235.92	237.55	239.01	240.45
0.0160	0.999048	0.997867	0.996441	0.994806	0.992975	234.95	236.30	238.14	239.58	240.88
0.0180	0.999145	0.997957	0.996536	0.994898	0.993061	235.54	237.15	238.53	240.02	241.56
0.0200	0.999237	0.998048	0.996624	0.994988	0.993153	236.16	237.69	239.11	240.39	241.70
0.0220	0.999338	0.998142	0.996718	0.995076	0.993239	236.29	238.02	239.34	240.80	242.13
0.0240	0.999426	0.998232	0.996807	0.995164	0.993323	236.94	238.46	239.74	241.15	242.57
					SDS in 0.1 m aqueous L-cystene solution					
0.0003	1.002972	1.001769	1.000334	0.998687	0.996845	222.06	222.93	224.17	224.09	225.75
0.0005	1.002983	1.001780	1.000344	0.998697	0.996855	228.20	229.39	230.23	231.11	231.84
0.0020	1.003081	1.001876	1.000441	0.998791	0.996946	223.66	224.85	225.08	226.86	228.68
0.0040	1.003201	1.001994	1.000553	0.998899	0.997049	226.15	227.34	229.07	231.10	233.42
0.0060	1.003293	1.002079	1.000634	0.998979	0.997129	231.08	233.12	235.05	236.69	238.37
0.0080	1.003411	1.002196	1.000745	0.999089	0.997238	230.36	232.07	234.33	235.76	237.23
0.0100	1.003498	1.002284	1.000838	0.999180	0.997332	233.24	234.56	235.92	237.32	238.27
0.0120	1.003597	1.002381	1.000933	0.999272	0.997416	233.82	235.12	236.47	237.95	239.48
0.0140	1.003697	1.002478	1.001027	0.999365	0.997507	234.47	235.84	237.25	238.63	240.13
0.0160	1.003793	1.002572	1.001118	0.999454	0.997594	235.12	236.47	237.92	239.30	240.79
0.0180	1.003886	1.002663	1.001206	0.999540	0.997679	235.77	237.10	238.59	239.97	241.39
0.0199	1.003973	1.002747	1.001289	0.999619	0.997757	236.36	237.74	239.17	240.65	242.02
0.0220	1.004055	1.002827	1.001365	0.999695	0.997830	237.47	238.83	240.33	241.70	243.12
0.0239	1.004153	1.002917	1.001455	0.999782	0.997914	237.41	239.01	240.42	241.83	243.29

Table 2. Limiting apparent molar volumes (φ_v^0) and experimental slopes (S_v) for SDS + water, L-cysteine+water and SDS + 0.1 m aqueous L-cysteine solutions at different temperatures.

Compound	T / K	φ_v^0 /m^3·mol^{-1}·10^6	S_v /m^3· mol^{-1}·10^{-6}	$(\partial^2 \varphi_v^0 / \partial T^2)_p$
		Pre-micellar region		
SDS + Water	293.15	227.15(±0.3844)	49.55 (±5.8001)	-0.002
	298.15	230.07 (±0.5528)	36.68 (±8.3410)	
	303.15	231.28 (±0.6102)	43.43 (±9.2072)	
	308.15	234.40 (±0.6541)	28.35 (±9.8702)	
	313.15	236.01 (±0.6739)	32.48 (±10.168)	

		Post-micellar region		
	293.15	225.73 (±0.5065)	72.53 (±3.7754)	-0.0014
	298.15	227.30 (±0.3960)	72.48 (±2.9519)	
	303.15	230.22 (±0.3036)	61.89 (±2.2632)	
	308.15	232.27 (±0.1290)	57.49 (±0.9617)	
	313.15	234.04 (±0.3524)	54.79 (±2.6270)	
	293.15	73.12 (±0.0144)	0.57 (±0.0305)	
	298.15	73.54 (±0.0136)	0.53 (±0.0287)	
L-cystene	303.15	73.95 (±0.0162)	0.49 (±0.0343)	-0.0007
	308.15	74.31 (±0.0103)	0.46 (±0.0218)	
	313.15	74.63 (±0.0095)	0.43 (±0.0201)	
		Pre-micellar region		
	293.15	221.77 (±2.2082)	102.25 (±33.318)	
	298.15	222.66 (±2.3191)	110.26 (±34.991)	
	303.15	223.27 (±2.4268)	123.04 (±36.617)	0.0033
SDS + 0.10 m L-cystene + water	308.15	223.77 (±2.3342)	136.44 (±35.221)	
	313.15	225.29 (±2.0571)	136.50 (±31.039)	
		Post-micellar region		
	293.15	224.37 (±0.7369)	85.57 (±5.4939)	
	298.15	225.20 (±0.6180)	89.75 (±4.6072)	
	303.15	226.40 (±0.6587)	91.48 (±4.9107)	0.0063
	308.15	227.90 (±0.6147)	90.80 (±4.5829)	
	313.15	229.68 (±0.6450)	88.38 (±4.8087)	

Table 3. Concentration dependence of ultrasonic velocity (u)and adiabatic compressibility (β_s) for L-cysteine, SDS and SDS in 0.1 m aqueous L-cysteine solutions at 293.15 K, 298.15 K, 303.15 K, 308.15 K, and 313.15 K.

	Ultrasonic velocity, u/ms^{-1}·10^{-2}					Adiabatic compressibility, β_s /N^{-1}m^2.10^7				
m/mol·kg^{-1}	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K
					L-cystene in water					
0.0500	1486.15	1500.15	1512.31	1522.83	1531.71	4.5250	4.4462	4.3812	4.3280	4.2858
0.0990	1489.71	1503.59	1515.63	1526.01	1534.76	4.4930	4.4157	4.3521	4.3002	4.2591
0.1496	1493.39	1507.16	1519.06	1529.29	1537.89	4.4603	4.3845	4.3224	4.2718	4.2320
0.1999	1497.06	1510.71	1522.49	1532.54	1541.01	4.4282	4.3538	4.2930	4.2440	4.2053
0.2499	1500.71	1514.22	1525.88	1535.78	1544.10	4.3966	4.3239	4.2643	4.2166	4.1791
0.2983	1504.28	1517.61	1529.16	1538.95	1547.07	4.3662	4.2952	4.2369	4.1902	4.1541
0.3500	1508.05	1521.28	1532.65	1542.23	1550.18	4.3343	4.2647	4.2080	4.1629	4.1281
0.3982	1511.51	1524.69	1535.92	1545.35	1553.12	4.3053	4.2366	4.1812	4.1374	4.1039
					SDS in water					
0.0003	1482.91	1497.05	1509.37	1520.01	1529.05	4.5556	4.4752	4.4086	4.3542	4.3108
0.0005	1482.95	1497.08	1509.40	1520.04	1529.08	4.5554	4.4750	4.4084	4.3540	4.3105
0.0020	1483.16	1497.29	1509.61	1520.24	1529.28	4.5536	4.4733	4.4068	4.3525	4.3091
0.0040	1483.45	1497.57	1509.89	1520.52	1529.55	4.5513	4.4712	4.4047	4.3504	4.3071
0.0060	1483.72	1497.85	1510.17	1520.79	1529.82	4.5492	4.4690	4.4026	4.3484	4.3052
0.0080	1483.99	1498.12	1510.43	1521.05	1530.08	4.5470	4.4669	4.4006	4.3465	4.3033
0.0100	1484.24	1498.36	1510.66	1521.28	1530.30	4.5450	4.4650	4.3989	4.3448	4.3016
0.0120	1484.45	1498.56	1510.85	1521.47	1530.49	4.5433	4.4634	4.3973	4.3433	4.3001
0.0140	1484.61	1498.73	1511.02	1521.63	1530.65	4.5418	4.4619	4.3959	4.3419	4.2988
0.0160	1484.76	1498.88	1511.16	1521.77	1530.79	4.5405	4.4606	4.3947	4.3407	4.2976
0.0180	1484.90	1499.01	1511.30	1521.91	1530.92	4.5392	4.4594	4.3934	4.3395	4.2965
0.0200	1485.03	1499.14	1511.43	1522.04	1531.06	4.5380	4.4582	4.3923	4.3384	4.2954
0.0220	1482.89	1497.02	1509.34	1519.98	1529.02	4.5558	4.4754	4.4088	4.3544	4.3109
0.0240	1482.91	1497.05	1509.37	1520.01	1529.05	4.5556	4.4752	4.4086	4.3542	4.3108
					SDS in 0.1m L-cystene					

0.0003	1490.27	1504.06	1516.08	1526.42	1535.16	4.4893	4.4127	4.3492	4.2976	4.2566
0.0005	1490.32	1504.11	1516.12	1526.46	1535.20	4.4890	4.4124	4.3489	4.2973E	4.2564
0.0020	1490.60	1504.37	1516.37	1526.69	1535.42	4.4869	4.4104	4.3471	4.2956	4.2547
0.0040	1490.93	1504.70	1516.68	1526.99	1535.71	4.4843	4.4079	4.3448	4.2934	4.2527
0.0060	1491.20	1504.96	1516.95	1527.26	1535.97	4.4823	4.4060	4.3429	4.2916	4.2509
0.0080	1491.49	1505.25	1517.22	1527.52	1536.22	4.4800	4.4038	4.3409	4.2897	4.2491
0.0100	1491.62	1505.38	1517.36	1527.66	1536.37	4.4789	4.4027	4.3397	4.2885	4.2478
0.0120	1491.76	1505.52	1517.49	1527.79	1536.49	4.4776	4.4014	4.3385	4.2874	4.2468
0.0140	1491.91	1505.65	1517.63	1527.93	1536.63	4.4762	4.4002	4.3373	4.2862	4.2457
0.0160	1492.04	1505.79	1517.76	1528.05	1536.76	4.4750	4.3990	4.3362	4.2851	4.2446
0.0180	1492.18	1505.93	1517.89	1528.18	1536.88	4.4738	4.3978	4.3351	4.2840	4.2435
0.0199	1492.31	1506.05	1518.01	1528.29	1536.99	4.4726	4.3967	4.3340	4.2831	4.2426
0.0220	1492.42	1506.17	1518.12	1528.40	1537.10	4.4716	4.3957	4.3331	4.2821	4.2417
0.0239	1492.55	1506.28	1518.24	1528.53	1537.23	4.4704	4.3946	4.3320	4.2810	4.2406

Table 4. Concentration dependence of viscosities for L-cysteine, SDS and SDS in 0.1 m aqueous L-cysteine solutions at 293.15 K, 298.15 K, 303.15 K, 308.15 K, and 313.15 K.

	Viscosity, η /kg·m^{-1}s^{-1}·10^{-2}				
m/mol·kg^{-1}	293.15K	298.15K	303.15K	308.15K	313.15K
			L-cystene in water		
0.0500	1.015020	0.903819	0.812177	0.731609	0.664225
0.0999	1.030023	0.921140	0.823511	0.741983	0.673175
0.1499	1.045178	0.930874	0.838590	0.750163	0.681399
0.2001	1.062423	0.943747	0.847599	0.758961	0.690620
0.2504	1.080073	0.958895	0.863352	0.771006	0.701086
0.2999	1.096212	0.971454	0.874410	0.781087	0.709719
0.3501	1.112552	0.987026	0.889210	0.792538	0.720576
0.3998	1.130826	1.000486	0.903641	0.802893	0.730355
			SDS in water		
0.0003	1.014340	0.895246	0.801506	0.723249	0.656635
0.0005	1.016031	0.895528	0.801785	0.723559	0.656983
0.0020	1.019894	0.896284	0.802356	0.724073	0.657565
0.0040	1.020387	0.896888	0.802791	0.724470	0.658467
0.0060	1.024493	0.897626	0.803187	0.724961	0.659895
0.0080	1.025870	0.898091	0.803656	0.725319	0.661553
0.0100	1.026983	0.898973	0.803851	0.725918	0.662791
0.0120	1.029241	0.900789	0.805398	0.726403	0.664558
0.0140	1.034363	0.907256	0.808531	0.727911	0.666743
0.0160	1.036165	0.909091	0.811372	0.730039	0.668851
0.0180	1.039626	0.911422	0.813480	0.734476	0.671751
0.0200	1.042431	0.913600	0.816067	0.738761	0.674655
0.0220	1.046886	0.919078	0.819278	0.742193	0.677519
0.0240	1.052416	0.921139	0.820550	0.746535	0.679395
			SDS in 0.1 m L-cystene		
0.0003	1.005455	0.894493	0.802634	0.722996	0.656491
0.0005	1.005882	0.894938	0.802965	0.723226	0.656719
0.0020	1.006302	0.895422	0.803442	0.723405	0.657019
0.0040	1.006839	0.895887	0.803873	0.723669	0.657437
0.0060	1.007216	0.896361	0.804376	0.723986	0.657950
0.0080	1.007751	0.896882	0.804769	0.724363	0.658428
0.0100	1.008444	0.897378	0.805319	0.724763	0.658895
0.0120	1.011272	0.899700	0.807258	0.725349	0.659393
0.0140	1.015560	0.902780	0.810717	0.726679	0.660928
0.0160	1.020148	0.905821	0.813871	0.729045	0.663825
0.0180	1.026781	0.910262	0.818087	0.731483	0.667422
0.0200	1.030528	0.914166	0.822033	0.736330	0.670240
0.0220	1.033872	0.917764	0.825652	0.740767	0.673572
0.0240	1.037537	0.921524	0.829358	0.743542	0.676580

Table 5. The viscosity coefficient values *A*, *B* and d*B*/d*T* for L-cysteine, SDS and SDS in 0.1 m aqueous L-cysteine solutions at 293.15 K, 298.15 K, 303.15 K, 308.15 K, and 313.15 K.

Compound	*T*/K	*A*-Coefficient	*B-Coefficient*	[d*B*/d*T*]

L-cystene + water	293.15	0.2867 (±0.0519)	0.2141 (±0.0131)	0.00127 (±0.00169)
	298.15	0.1214 (±0.0700)	0.2574 (±0.0176)	
	303.15	0.0932 (±0.0285)	0.2846 (±0.0072)	
	308.15	0.0660 (±0.0253)	0.2514 (±0.0064)	
	313.15	0.0903 (±0.0117)	0.2489 (±0.0030)	
		Pre-micellar region		
SDS + water	293.15	-3.8506 (±0.6553)	0.5573 (±0.0434)	-0.02127 (±0.01038)
	298.15	-0.5114 (±0.1253)	0.0994 (±0.0083)	
	303.15	-0.2564 (±0.0488)	0.0619 (±0.0032)	
	308.15	-0.2273 (±0.0700)	0.0638 (±0.0046)	
	313.15	0.5007 (±0.1546)	0.0434 (±0.0102)	
		Post-micellar region		
	293.15	1.5967 (±0.2126)	0.0469 (±0.0285)	-0.01137 (±0.00737)
	298.15	2.5034 (±0.3098)	-0.1864 (±0.0416)	
	303.15	2.3711 (±0.1353)	-0.2000 (±0.0181)	
	308.15	3.2189 (±0.4396)	-0.3278 (±0.0590)	
	313.15	2.5831 (±0.0977)	-0.1667 (±0.0131)	
		Pre-micellar region		
SDS + 0.10 m L-cystene + water	293.15	-0.0013 (±0.0600)	0.0310 (±0.0040)	0.000161(±0.00329)
	298.15	-0.2187 (±0.0726)	0.0578 (±0.0048)	
	303.15	-1.0022 (±0.2019)	0.1422 (±0.0134)	
	308.15	-0.1598 (±0.0727)	0.0416 (±0.0048)	
	313.15	-0.0394 (±0.0708)	0.0431 (±0.0047)	
		Post-micellar region		
	293.15	3.4772 (±0.2199)	-0.3192 (±0.0295)	-0.00164 (±0.00089)
	298.15	3.3755 (±0.1148)	-0.3190 (±0.0154)	
	303.15	3.5021 (±0.0556)	-0.3092 (±0.0075)	
	308.15	3.0876 (±0.4468)	-0.3277 (±0.0599)	
	313.15	3.6048 (±0.1447)	-0.3559 (±0.0194)	

Table 6. Concentration dependence of activation parameters for viscous flow of L-cysteine, SDS and SDS in 0.1 m aqueous L-cysteine solutions at 293.15 K, 298.15 K, 303.15 K, 308.15 K, and 313.15 K.

m/mol·kg^{-1}	Free energy, $\Delta G^{\neq}$/J·mol^{-1}·10^3					$\Delta S^{\neq}$/ J· mol^{-1}·K^{-1}	$\Delta H^{\neq}$/ J· mol^{-1}·K^{-1}
	293.15K	298.15K	303.15K	308.15K	313.15K		
			L-cystene in water				
0.0500	9.3311	9.2055	9.0940	8.9806	8.8795	22.6764	16.0055
0.0999	9.3737	9.2596	9.1361	9.0239	8.9218	22.8111	16.0583
0.1499	9.4163	9.2928	9.1892	9.0595	8.9610	22.8926	16.1236
0.2001	9.4631	9.3339	9.2234	9.0968	9.0035	23.1628	16.2459
0.2504	9.5101	9.3804	9.2769	9.1444	9.0502	23.1462	16.2893
0.2999	9.5529	9.4195	9.3159	9.1848	9.0892	23.2734	16.3678
0.3501	9.5960	9.4661	9.3656	9.2296	9.1364	23.1412	16.3739
0.3998	9.6423	9.5064	9.4130	9.2698	9.1786	23.3050	16.4669
			SDS in water				
0.0003	9.3226	9.1749	9.0536	8.9439	8.8422	23.9086	16.3154
0.0005	9.3268	9.1758	9.0546	8.9451	8.8437	24.0142	16.3490
0.0020	9.3368	9.1787	9.0571	8.9477	8.8468	24.3035	16.4411
0.0040	9.3389	9.1813	9.0595	8.9501	8.8514	24.2112	16.4158
0.0060	9.3496	9.1843	9.0617	8.9528	8.8581	24.3941	16.4764
0.0080	9.3538	9.1865	9.0641	8.9550	8.8655	24.2694	16.4423
0.0100	9.3572	9.1898	9.0656	8.9580	8.8713	24.1896	16.4214
0.0120	9.3635	9.1956	9.0713	8.9607	8.8792	24.1929	16.4282
0.0140	9.3764	9.2142	9.0820	8.9668	8.8886	24.5795	16.5568
0.0160	9.3814	9.2200	9.0916	8.9751	8.8977	24.3633	16.4990
0.0180	9.3902	9.2271	9.0989	8.9915	8.9098	24.0541	16.4155
0.0200	9.3975	9.2338	9.1077	9.0071	8.9217	23.6849	16.3135
0.0220	9.4086	9.2493	9.1183	9.0197	8.9335	23.7115	16.3340
0.0240	9.4221	9.2555	9.1229	9.0354	8.9415	23.7548	16.3567
			SDS in 0.1 m L-cystene				
0.0003	9.3151	9.1871	9.0717	8.9578	8.8567	22.9574	16.0372
0.0005	9.3163	9.1884	9.0728	8.9587	8.8578	22.9708	16.0419
0.0020	9.3180	9.1906	9.0751	8.9602	8.8598	22.9741	16.0453

0.0040	9.3204	9.1929	9.0776	8.9622	8.8626	22.9616	16.0440
0.0060	9.3224	9.1954	9.0803	8.9645	8.8658	22.9159	16.0325
0.0080	9.3247	9.1978	9.0826	8.9670	8.8688	22.8901	16.0273
0.0100	9.3275	9.2004	9.0855	8.9696	8.8719	22.8801	16.0270
0.0120	9.3354	9.2078	9.0926	8.9728	8.8750	23.1537	16.1157
0.0140	9.3468	9.2174	9.1046	8.9786	8.8822	23.3931	16.1976
0.0160	9.3589	9.2269	9.1155	8.9881	8.8948	23.3781	16.2039
0.0179	9.3758	9.2401	9.1297	8.9979	8.9101	23.5220	16.2615
0.0200	9.3857	9.2518	9.1429	9.0159	8.9222	23.3000	16.2070
0.0220	9.3948	9.2628	9.1552	9.0326	8.9364	22.9774	16.1220
0.0240	9.4045	9.2740	9.1676	9.0433	8.9491	22.8627	16.0986

solutions in the low concentration region ($\varphi_v - \sqrt{m}$ plots) shows sudden change in φ_v value at particular molality (figure 1). The apparent molar volume of SDS in aqueous and in aqueous L-cysteine may have two components, viz. true size of the molecule and the free space between the molecules. In premicellar region, the molecules exist as monomer and the monomer interaction may account for φ_v with concentration having the free space between the molecules [31].

The apparent molar volume at infinite dilution, (φ_v^0) was calculated using least square fit to the linear plots of experimental values of φ_v versus square root of molal concentration ($\sqrt{m}$) using the Masson equation [32]:

$$\varphi_v = \varphi_v^0 - S_v\sqrt{m} \qquad (3)$$

where S_v is the experimental slope, which is sometimes considered to be the volumetric pairwise interaction coefficient [33, 34]. The limiting apparent molar volume (φ_v^0) and S_v values along with standard error, are given in Table 2. For SDS in aqueous L-cysteine solution, the values of S_v are positive which indicates the very strong ion-ion interactions in this region. In the case of premicellar region in aqueous L-cysteine the ion-ion interaction is greater than post-micellar region.

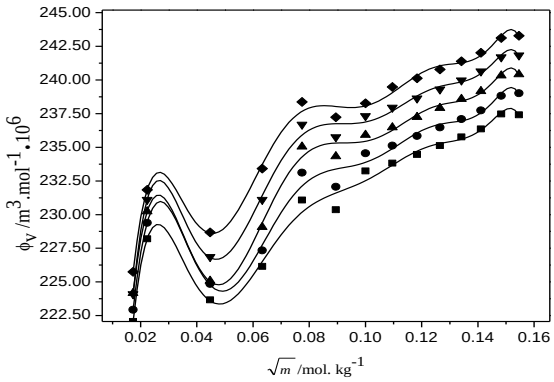

Figure 1. Plots of φ_v/m^3.mol^{-1}.10^6 versus $\sqrt{m}$ /mol. kg^{-1} for SDS in 0.1 m L-cysteine solution at different temperatures (indicated by symbols: ■ 293.15 K, ● 298.15 K, ▲ 303.15 K, ▼ 308.15 K, ◆313.15 K) .

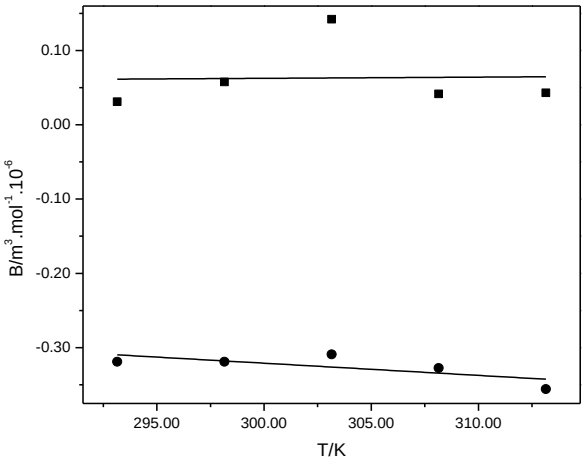

Figure 2. Plots of viscosity B-coefficients of SDS in 0.1 m aqueous L-cysteine solution versus temperature (indicated by symbols ■ pre-miceller region, ● post-miceller region).

The limiting apparent molar volumes (φ_v^0) which is taken to be the partial molar volume at infinite dilution of SDS in aqueous and in aqueous L-cysteine solutions reflect the true volume of the solute and the volume change arising from solute-solvent interactions. The variation of φ_v^0 with the molality of SDS can be rationalized in terms of cosphere overlap model [35]. According to the model, the overlap of the cospheres of two ions or polar groups or an ion with that of a hydrophilic group always produces a positive volume change. On the other hand, the overlap of the co-spheres of an ion with that of hydrophobic groups results in a negative volume change. By the overlap of the co-spheres of SDS-SDS and SDS-hydrophilic groups, zwitterion interactions may take place. The overlap of the co-spheres of SDS gives a positive change in volume due to the relaxation of the electrostricted water molecules due to strongly localized ion-zwitterion interactions from the co-spheres of amino acid and SDS may cause an increase in volume [36].

The positive volume change due to the overlap of the co-spheres of amino acid with those of the hydrophilic groups of SDS outweighs the negative volume change due to the overlap of co-spheres of amino acid and hydrophobic group of SDS (negligible), giving a greater φ_v^0 value in amino acid compared to that in water in this region (Table 2). The water-water and water-amino acid interaction are assumed to be the same and do not produce a considerable change in volume. An increase in the molality of SDS increases the ion-zwitterion and also the SDS-SDS interactions giving rise to increased φ_v^0 values.

The increase of φ_v^0 with temperature may be due to the result of the following effects [37].

i) At higher temperature the thermal energy of the water molecule is increased causing fast movement of the bulk electrostricted water molecules from the interaction region of $-{}^{+}NH_3$ and $-COO^-$groups results in a positive volume change.

ii) An increase in temperature renders the SDS-SDS interaction giving rise to small negative volume change.

iii) A decrease in SDS-water interactions causing a positive volume change.

iv) The water-water interactions decreasing with increasing in temperature giving rise to a small negative change in volume.

SDS in aqueous and in aqueous amino acid solutions, the interactions of SDS-SDS and SDS-Zwitterions interaction increase giving rise to an increased φ_v^0 value.

The φ_v^0 value of SDS in ternary solution can alternatively be thought of arising from four constituents, as [38]:

$$\varphi_v^0 = V_w + V_f + V_n + V_s \quad (4)$$

where V_w and V_f are vander Waals volume and volume of empty spaces present there in respectively[39]. The V_n and V_s represent the contributions due to the hydrophobic and hydrophilic hydration.

The V_w and V_f are assumed to be the same in aqueous amino acids as in water. The variation of φ_v^0 is therefore due to the change in (V_n+V_s) resulting from SDS-amino acid, amino acid-amino acid and amino acid-water interactions, the contribution from water-water is assumed to be negligible.

The temperature dependence of limiting apparent molal volume φ_v^0 for binary and ternary solution can be expressed by the expression

$$\varphi_v^0 = a + bT + cT^2 \tag{5}$$

The sign of $\left[\frac{\partial^2 \varphi_v^0}{\partial T^2}\right]_p$ *i.e.* second derivative of limiting apparent molar volume of the solution with respect to temperature at constant pressure which corresponds to structure making or breaking properties of solution were determined. From Table 2 L-cysteine and SDS in aqueous solutions, show negative value of $\left[\frac{\partial^2 \varphi_v^0}{\partial T^2}\right]_p$ indicating that L-cysteine and SDS act as a structure breaker for water solvent systems [40]. Similar information was reported by Devine and Lowe [41]. Again the value of $\left[\frac{\partial^2 \varphi_v^0}{\partial T^2}\right]_p$ was found to be positive for SDS in aqueous L-cysteine solution corresponding to structure making property of water [42].

The concentration dependence of speed of sound of aqueous solution of SDS are listed in Table 3 at studied temperatures. The values of ultrasonic velocity are found to increase with increases in temperature as well as solute concentration. Such an increase may be attributed to an increase in the intermolecular interaction between the SDS–aqueous L-cysteine.

The experimental values of density and ultrasonic velocity, *u* were combined to calculate adiabatic compressibility, β_s using the Laplace equation [28-33]:

$$\beta_S = \frac{1}{\rho u^2} \tag{6}$$

The results of adiabatic compressibility of the systems under investigation are presented in Table 3. The adiabatic compressibility is found to decrease with increases in temperature and concentration. Such a decrease may be attributed to an increase in the ion–ion interaction as well as to the corresponding increase in the number of incompressible ions with an increase in solute concentration. Another reason for the decrease in the adiabatic compressibility may be due to a change in the structure of water around the ions. The ionic part of SDS holds the water molecule through hydrophilic hydration, while the hydrophobic moiety of SDS holds the water molecule through hydrophobic hydration. The hydrophilic hydration shell of ionic group of SDS is controlled predominately by electrostatic interactions between water molecules and the ionic group of SDS. Strong electrostatic force between ionic group of SDS and water causes electrostriction. The hydrophobic moiety of SDS forms hydration shell around it through hydrophobic hydration. Thus hydrophilic as well as hydrophobic solute-solvent interactions determine the hydration sphere of SDS. As the concentration of SDS increases, the solute-solute interaction relaxes water molecules from both hydrophilic and hydrophobic hydration zone and hydrophobic zone to the bulk render the solution more compact and imparts a decrease in molality of SDS.

Viscometric properties of SDS and L-cysteine in aqueous and SDS in aqueous L-cysteine solutions were measured at 293.15, 298.15, 303.15, 308.15 and 313.15 K respectively. The relevant data were shown in Table 4.

The values of η increase with increase in molality and decrease with temperature. According to the Flickering cluster' [43] models of water, there are large void spaces within the hydrogen-bonded framework of water structure. The linear increase of η with concentration may be interpreted by the fact that the molecules may have penetrated in the void spaces and may have a positive interaction with water.

To calculate the viscosity coefficient *B* values, the viscosity data were analyzed in terms of the semi-empirical Jones-Dole equation [44].

$$\eta_{rel} = 1 + A\sqrt{C} + BC \tag{7}$$

The viscosity coefficient *B* represents information regarding solute-solvent interaction and shape and size effect on the solvent structure [45, 46]. The calculated values of the B-coefficient are represented in Table

5. The *B*-coefficient values of the above electrolyte in aqueous solutions are based on the fact that there exists around an ion, a region of modified solvent differing from the bulk in structure and in properties. Gurney's [47] co-sphere, Frank and Wens [43] A, B, and C zones and Nightinagle's hydrated radius are recent reflection of this idea. From the above approaches Kaminsky indicated that the observed viscosity changes results from competition between various effects occurring in the neighborhood ion.

The sign of $\frac{dB}{dT}$ i.e. first derivative of the viscosity coefficient of *B* with respect to temperature which corresponds to structure making or breaking properties of solutes were determined [48]. The values of $\frac{dB}{dT}$ are presented in Table 5.

For L-cysteine in aqueous solutions, the values of $\frac{dB}{dT}$ is positive which corresponds to structure breaking behaviour [49]. L-cysteine and alanine have high value of *B*, the simplest amino acids are classified as structure breaker [50-52]. It is seen that SDS in aqueous solutions and SDS in aqueous L-cysteine solutions for postmiceller region, the values of $\frac{dB}{dT}$ are negative which corresponds to structure making behaviour. But $\frac{dB}{dT}$ is positive for SDS in aqueous L-cysteine solutions for pre-miceller region which indicates that SDS acts as structure breaker (Figure 2) in aqueous L-cysteine solvent system [53].

The viscosity coefficient A represents the ion-ion interactions coupled with the size and shape effect of the solute and to some extent, solute-solvent interactions. In this study, an irregular variation in the values of A is found which may be due to

i) incomplete dissociation and ion association of electrolyte in aqueous and in aqueous L-cysteine solvent and
ii) the size of the ions which differs in the degree of hydration or solvation [54].

The decrease of *A* with rising temperature is probably due to the greater thermal agitation and reduction of attractive forces between the ions. The increase in *A* value can be explained by the interpenetration effect, which brings the ions closer together [55]. On the otherhand the viscosity coefficient *D* also represents the solute-solute interaction. But it is related to the non-electrolyte solutions.

The thermodynamic properties of aqueous SDS, aqueous L-cysteine and SDS in 0.1m aqueous L-cysteine solutions were calculated at the mentioned temperatures and are shown in Tables 6. The $\Delta G^{\neq}$ value is positive for all the studied systems. The positive free energy of activation for viscous flow can be interpreted with the help of Furth model [56] which states that the kinetic species involved in forming cavities or holes in the liquid medium is given by the work required in forming the hole against surface tension of the solution. The solute-solvent interaction, interstitial incorporation, hydrophilic hydration interaction renders the binary and ternary aqueous systems more structured. This is reflected the positive $\Delta G^{\neq}$ value.

The variations of entropy of activation ($\Delta S^{\neq}$) with the concentration of binary and ternary systems are noted in Table 6. The $\Delta S^{\neq}$ values for the flow process are positive in all cases but do not follow any specific pattern. The positive values of $\Delta H^{\neq}$ indicate that positive work has to be done to overcome the energy barrier for the flow process. That is, the viscous flow is not thermodynamically favoured for the systems studied.

IV. Conclusions

The densities, ultrasonic velocities, and viscosities of SDS and L-cysteine in water and SDS in 0.1m aqueous L-cysteine solutions were measured at different temperatures. Using the experimental results, various parameters were calculated. The results indicate that the behaviour of SDS in aqueous solution is a temperature dependent property. In pre-micellar and post-micellar region, it appears to be a structure breaker for water solvent system at the temperatures studied from 293.15 K to 313.15 K. Moreover, L-cysteine also shows the structure breaking properties in aqueous system at the studied temperature ranges. SDS in 0.1m aqueous L-cysteine solution acts as structure breaker in pre-micellar region and structure maker in post-micellar region at the studied temperatures.

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