



Apparent molar volume, compressibility and viscometric studies of sodium dodecyl benzene sulfonate (SDBS) and dodecyltrimethylammonium bromide (DTAB) in aqueous amino acid solutions: A thermo-acoustic approach



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ABSTRACT

In this paper, we examine density (d), ultrasonic velocity (u) and viscosity studies of SDBS and DTAB in water and in aqueous solutions of 0.01, 0.05, 0.10 mol kg⁻¹ L-glutamine, L-histidine and L-methionine at $T = 293.15, 298.15, 303.15, 308.15, 313.15$ K. The apparent molar volume (ϕ_v) dependence on SDBS and DTAB concentration reflects the modification of water–water interactions described as hydrophobic hydration of the surfactant molecules in the presence of amino acid. The trends of apparent molar adiabatic compression (ϕ_k) values of DTAB with surfactant concentration appear reasonable to suggest that hydrophilic hydration of DTAB–amino acid complex occurs at low surfactant concentration region rendering it to be negative. The gradual increase in viscous relaxation time (τ) with rise in concentration of surfactant and decrease with temperature seem to be happen due to the structural relaxation processes occurring as a result of re-arrangement of molecules.

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1. Introduction

The objective of this paper is to lift the basic knowledge pertaining to amino acid–surfactant interactions which is still a conjecture. Various biological processes involve change in volume along with the solvation of molecules. Their complete understanding needs a comprehensive notion for the state and the behavior of these molecules in the solvent medium. Since most of these reactions take place in aqueous environment, it is pertinent to know the effect of water as solvent on the volumetric and viscometric properties of such processes. As known, 20% of the human body is made up of proteins which play a decisive role in almost every biological process and more importantly, amino acids are the building blocks of it. Amino acids are organic compounds contain at least one $-\text{NH}_2$ and a $-\text{COOH}$ group. In the human genome, 20 amino acids are created to build proteins and therefore termed proteinogen. Besides this, there are approximately 250 amino acids which do not form proteins. These are generally used to form sugars. Amino acids have an

influence on the function of organs, glands, tendons and arteries. They are furthermore essential for healing wounds and repairing tissue, especially in the muscles, bones, skin and hair as well as for the removal of all kinds of waste deposits produced in connection with the metabolism [1–4]. Therefore, it is quite worthy to study such molecules in vitro having diverse biological importance.

Surfactants play a major role in the formulation of many chemical products. In the first place, they are used to stabilize emulsions and microemulsions. Secondly, surfactants are added in emulsifiable concentrates for their spontaneous dispersion on dilution. Surfactants also form a high class of industrial applications including food, pharmaceuticals, in petroleum and in oil recovery, etc. [5–9]. Their role as ingredient in many biochemical industries cannot also be ignored. On one hand, anionic surfactants are very soluble in water at room temperature and are used pharmaceutically as a preoperative skin cleaner, having bacteriostatic action against Gram-positive bacteria, and also in medicated shampoos and is a component of emulsifying wax. On the other hand, the quaternary ammonium and pyridinium cationic surfactants are important pharmaceutically because of their bactericidal activity against a wide range of Gram-positive and some Gram-negative organisms. They may be used on the skin, especially in the cleaning of wounds. Their aqueous solutions are used for cleaning contaminated utensils. Such a wide range of applications of such molecules (amino acids and surfactants) has forced us to study their

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Table 1
Specification and mass fraction purity of chemical samples.

Chemical name	Source	Purification method	Mass fraction purity
L-Glutamine (Gln)	Merck	None	0.99 ^a
L-Histidine (His)	Merck	None	0.99 ^a
L-Methionine (Met)	S. D. Fine	None	0.98 ^a
Sodium dodecyl benzene sulfonate (SDBS)	HIMEDIA	Recrystallized	0.97
Dodecyltrimethylammonium bromide (DTAB)	S. D. Fine	Recrystallized	0.98

^a Declared by supplier.

physico-chemical properties to make them reliable to be used in every chemical and industrial application.

Further, among micellization of amino acid–surfactant, it has been proposed that the hydrophobic and electrostatic interactions are the two main driving forces for the association between surfactants and amino acids in aqueous solution. The aggregation phenomena of amphiphilic molecules involve contributions from both repulsive and attractive interactions. Especially in ionic surfactants, the repulsive forces originated primarily from electrostatic repulsion between the polar head groups [10], whereas, attractive interactions have generally been attributed to hydrophobic interactions [11] between the non-polar tails of the surfactant monomers. The interactions of these two moieties with water and additives are an important cause for surfactants to aggregate into micelles and other nanometer scale structures in aqueous solution [12]. Since most applications depend on the formation and characteristics of micelles, it is necessary to understand the magnitude and nature of the additives as well as the mechanism involved as they provide vital information about the solute–solute and solute–solvent interactions in surfactant solutions. These additives can modify the micellization process either through specific interactions with surfactant molecules or by changing the solvent nature [13].

In addition, the volumetric and viscometric properties are important tools to study the solution behavior of solutes and display valuable information about solute–solute and solute–solvent interactions present in the mixture. These include apparent molar volume, compressibility, apparent molar isentropic compression and relative viscosity, etc. [14–16]. Although, volumetric and viscometric properties of aqueous solutions of surfactants with or without additives have been studied extensively in the recent years [17–19], there is hardly any information regarding these properties in case of L-glutamine, L-histidine and L-methionine with ionic surfactants, SDBS and DTAB in aqueous solutions. Therefore, in the present work, we tried to rejuvenate the type of intermolecular interaction present in such system which can prove its efficacy both in biological and technical applications. For this, we choose two industrially important surfactants, SDBS and DTAB as solute moieties and three biologically active amino acids at three different molal concentrations (0.01, 0.05 and 0.10 mol kg⁻¹) as a solvent medium. The temperature variance is also in compliance with the biological system i.e. 293.15–313.15 K.

2. Experimental

2.1. Material

SDBS and DTAB, both were of A.R. grade obtained from HIMEDIA and S.D. Fine-Chem. Ltd., respectively. However, a pure sample of SDBS and DTAB was obtained by giving the following additional treatment as reported in literature [20,21]; 50 g of SDBS or DTAB was dissolved in about 500 ml A.R. grade ethanol. Suspension in the solution was removed by filtration. After filtration, the solution was heated on a steam bath as long as the volume of the solution was reduced to one-fourth of its original volume. The solution was left to cool at room temperature for about 2 h. Appearance of white

colored needles of pure surfactant was observed soon after the liquid attained the room temperature. The solution was decanted and the crystals were allowed to dry at room temperature. The sample was however, re-crystallized twice from ethanol and finally dried in vacuum oven in the presence of P₂O₅ for 24 h at ~60 °C. L-Histidine and L-glutamine were obtained from MERCK (Germany) and L-methionine from S. D. Fine-Chem. Ltd. (India), all were of A.R. grade and were used as received. The provenance and final mass fraction purity of the sample used have been provided in Table 1.

2.2. Method

Density and speed of sound measurements were performed with a high-precision digital Density and Sound Analyzer-5000 (DSA-5000) supplied by Anton Paar (Austria). The instrument was further calibrated periodically with distilled water over a temperature range 293.15–313.15 K. The two in one instrument has been equipped with a density cell and a sound velocity cell. Both cells are temperature-controlled by a built-in Peltier thermostat. The sample is introduced into a U-shaped glass tube that is being excited to vibrate at its characteristic frequency electronically. The characteristic frequency changes depending on the density of sample. The precision in the density data was found to be better than $\pm 2 \times 10^{-6}$ g cm⁻³ and that of velocity data it was better than ± 0.20 m s⁻¹. The precision in temperature of the DSA-5000 is ± 0.001 K. The uncertainties in the density measurements were well within $\pm 2 \times 10^{-4}$ kg m⁻³ and the uncertainties in the speed of sound measurements were found to be better than ± 0.2 m s⁻¹.

Viscosity measurements were carried out using Man Singh Survismeter, an inexpensive device for measuring viscosity and surface tension of the solution simultaneously [22,23]. It was supplied by Spectro Lab. Equip. Pvt. Ltd. (India). The Survismeter was periodically cleaned by treating with chromic acid and distilled water and finally washed with alcohol and dried in oven for 3–4 h. After drying, the Survismeter was filled with fixed volume of the test solution and was clamped in the thermostated paraffin bath in a vertical position. The bath used for measurements of viscosity values was maintained at a desired temperature ± 0.01 K for about 30 min prior to recording of readings at each temperature of study. The solution was sucked into the measuring bulb and was allowed to stand there for about 5 min by closing the corks and then the corks were removed for recording the time of flow of solution from the upper to lower end of the bulb through the fine capillary. After taking 2–3 readings at the desired temperature, the average of the very close values of time of fall was taken. The average deviation for these measurements of a single concentration of a solution did not exceed ± 0.03 s. The efflux time for viscosity measurements was in between 300 and 395 s, therefore, no kinetic energy correction was made [24]. The temperature of Survismeter was maintained constant to ± 0.1 K. However, the viscometer was calibrated before use with DMSO and MeOH (both of A.R. grade) at 298.15 K using viscosity coefficient, $\eta_0 = 0.008903$ poise and density, $d = 0.99707$ g cm⁻³ for water [25]. The viscosity coefficients of DMSO and MeOH were found to be 0.02 and 0.0055 poise respectively which were found in good agreement with the literature values [26,27]. The precision

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