



Carbohydrate-surfactant interactions in aqueous and mixed organic solvents at various temperatures: Volumetric, compressibility and acoustical studies



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ABSTRACT

Herein, we report the detailed volumetric, compressibility and acoustical studies of carbohydrate-surfactant interactions in aqueous and mixed organic solvents at various temperatures. The interactions between sugars (ribose, glucose, sucrose and raffinose) and sodium dodecyl sulfate (SDS, an anionic surfactant) were studied in aqueous solutions at various temperatures, i.e. 293.15, 303.15 and 313.15 K using density and speed of sound measurements. Further, to clearly understand the sugar-surfactant interactions, various parameters such as molar volume (V_ϕ), isentropic compressibility (κ_s) and apparent molar isentropic compressibility ($\kappa_{\phi,s}$) were calculated from the obtained data. The observed results revealed that different saccharides exhibit pronounced effect on micellization of SDS which might be due to the differences in number of -OH linkages. The increase in such linkages results in dominance of sugar-water interactions, thus favoring the micelle formation. Further, to support such conclusion, viscosity measurements (relative viscosity and viscous relaxation time) of SDS in aqueous solutions of sugars were also examined. The effect of different co-solvents viz. dimethyl sulfoxide (DMSO), methanol (MeOH) and dioxane on sugar-surfactant interactions has also been discussed and demonstrated in this paper.

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1. Introduction

Water-soluble biopolymers such as carbohydrates have recently gained much interest due to their unique rheological properties e.g. alteration of transition temperatures, formation of thermo-reversible gels or changes in the gel strength and solution viscosity [1–3]. The interactions of carbohydrates with food surfactants are extremely important in many food industries including chemical feed stocks, food production, preservation of processed foods, and so on [4–10]. The properties of saccharide solutions are of great interests in various aspects of basic research and applications as these poly-hydroxy compounds present complex three-dimensional arrays of functionality to their surroundings [11–13]. The peculiar nature of carbohydrates i.e. their pronouncedly hydrophilic character containing multiple hydroxyl groups with well-defined orientations are responsible for making strong cooperative hydrogen bonds with the surfactant molecules [14–16]. This fact, together

with the hydrophobic interactions between the long hydrocarbon chains, lead to spontaneous association of amphiphiles in water. The micellization behavior of surfactants is greatly influenced by the presence of co-solutes or co-solvents, and can be interpreted in terms of the structural consequences of intermolecular interactions, which provide a potential tool to investigate structural changes in these solutions [17–24].

The properties of ionic surfactant solutions in non-aqueous and mixed solvents have extensively studied in order to have a better understanding of their relative behavior in different solvents [25–35]. Such systems are important in fundamental research and technology as the properties such as viscosity, relative permittivity (dielectric constant), and molar volume are strongly dependent on the solvent composition. The nature of solvent affects the stability of system by distributing itself between aqueous and micellar phase or by accumulating both at polar head groups and inside the micelle hydrophobic core. Investigations involving effect of organic additives, such as dipolar protic/aprotic solvents on CMC in aqueous solutions are primarily dealt with their effect on bulk dielectric constant of the medium, which influences the state of solvation of both the hydrophilic and hydrophobic portion of surfactant molecule. Organic additives, which increase the CMC, decrease the aggregation number [36] are rationalized as being due to “structure-breaking effect” of the additive whereas

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a decrease in CMC is interpreted to mean “structure-making effect” [37]. The present work is not only expected to provide information on the interactions existing between sugars and sodium dodecyl sulfate (SDS), an anionic surfactant in water, but also enables us to understand the effect of co-solvents as additives (DMSO, MeOH and Dioxane) on surfactant aggregation in water by varying the solvent composition.

2. Experimental details

2.1. Materials

Double distilled water (Millipore-Elix system; conductivity = $1\text{--}2 \times 10^{-6} \text{ S}\cdot\text{cm}^{-1}$, and pH = 6.7–6.9) was used for all experiments. Sodium dodecyl sulfate (SDS), biochemical grade was procured from BDH and was further purified as suggested in literature [38]. Ribose and raffinose were obtained from Loba Chemie Pvt. Ltd. Sucrose and glucose were obtained from S.D. Fine-Chem. Pvt. Ltd. Table 1 demonstrate the specifications of the chemicals used as declared by the supplier.

2.2. Methods

Densities and sound velocities were measured from Density and Sound Velocity Analyzer (DSA-5000) supplied by Anton Paar GmbH, Graz, Austria. The DSA-5000 measures the speed of sound and density of the solution simultaneously with a resolution of $1 \times 10^{-1} \text{ m}\cdot\text{s}^{-1}$ and $1 \times 10^{-6} \text{ g}\cdot\text{cm}^{-3}$ respectively. The calibration of DSA was done over a temperature range 293.15–318.15 K. DSA was thermostated with in $\pm 0.002 \text{ K}$ with Peltier heating device. Viscosity measurements were carried out with a calibrated Ostwald viscometer. The precision achieved in viscosity measurements was $\pm 0.02\%$. A high precision water thermostat fitted with a digital temperature controlled device was supplied by NSW - New Delhi. The temperature of the thermostat was maintained within $\pm 0.1 \text{ K}$ over the entire temperature range studied. The density and sound velocity values of SDS were measured over a wide range of solvent concentrations (0.05, 0.10, 0.20) $\text{mol}\cdot\text{kg}^{-1}$ in aqueous solutions of ribose, glucose, sucrose and raffinose at different temperatures (293.15 to 318.15) K.

3. Results and discussion

Speed of sound in combination with density and viscosity furnish information that takes into account the contributions arising from different kind of interactions with respect to the behavior of solute species in solutions [39–43]. Therefore, density, ρ and speed of sound, u for SDS in aqueous solutions of ribose, glucose, sucrose and raffinose have been measured at different temperatures and are listed in Tables S1–S2.

A perusal of speed of sound data shows a considerable concentration dependence of all the studied sugars. Similar results for carbohydrates in binary mixtures of DMSO + pure water at 298.15 K have been

reported in literature [44,45]. The data has been further used to derive the isentropic compressibility, κ_s by using the equation:

$$\kappa_s = 1/u^2\rho. \quad (1)$$

Fig. 1(a–c) shows the dependence of isentropic compressibility, κ_s on [SDS] in various sugars at different temperatures. It is observed that isentropic compressibility, κ_s tends to decrease linearly with [SDS]. The decrease of κ_s values is due to the influence of solute on the surrounding solvent molecules leading to increase in internal pressure and thus solution becomes harder to compress which is indicative of the presence of solute–solvent interactions [46,47].

The density and speed of sound data were however, used to calculate the apparent molar volume, V_ϕ and apparent molar isentropic compressibility, $\kappa_{\phi,s}$ of SDS from the following relations [48].

$$V_\phi = \frac{M}{\rho} + \frac{(\rho_o - \rho)}{m\rho\rho_o} \quad (2)$$

$$\kappa_{\phi,s} = V_\phi\kappa_s + \frac{(\kappa_s - \kappa_{s,o})}{m\rho_o} \quad (3)$$

where m is the molality ($\text{mol}\cdot\text{kg}^{-1}$) of solution, M is the molar mass of SDS ($\text{kg}\cdot\text{mol}^{-1}$), and ρ_o and ρ are the densities of the solvent and solution ($\text{kg}\cdot\text{m}^{-3}$), respectively. κ_s and $\kappa_{s,o} = 1/u_o^2\rho_o$ are the isentropic compressibilities of the solution and solvent (TPa^{-1}), respectively. The values of V_ϕ and $\kappa_{\phi,s}$ have been reported in Tables 2 and 3 with an uncertainty of $\pm 0.2 \times 10^{-5} \text{ m}^3\cdot\text{mol}^{-1}$ and $\pm 5 \times 10^{-3} \text{ m}^3\cdot\text{mol}^{-1}\cdot\text{TPa}^{-1}$ respectively.

The data could not be analyzed in terms of Masson's equation ($V_\phi = V_\phi^o + S_v m^{1/2}$), for the reason that V_ϕ dependence on SDS concentration was not found to be linear. However, an attempt was made to derive information as regards to sugar–surfactant interaction from the dependence of V_ϕ and $\kappa_{\phi,s}$ on surfactant concentration. The solution behavior of such molecules as bearing both hydrophobic and hydrophilic groups, can be expected to be quite complex because it is likely to be governed by a delicate balance of both hydrophobic and hydrophilic interactions [49,50]. The possibilities of various types of interactions occurring in the ternary saccharide–surfactant–water system are:

- (1) hydrophilic–ionic interactions between the –OH groups of saccharides and the $-\text{OSO}_3^-$ group of surfactant i.e. SDS,
- (2) hydrophilic–hydrophobic interactions between –OH groups of saccharides and the non-polar groups of surfactant and
- (3) hydrophobic–hydrophobic interactions between the non-polar groups of saccharides and non-polar groups of surfactant.

The dependence of V_ϕ values of SDS in aqueous solutions of different sugars at 293.15, 303.15 and 313.15 K has been shown in Fig. 2(a–c). The V_ϕ values are positive for all sugars over the entire [SDS] as well

Table 1
Specification of chemicals used.

Chemical name	Source	M·Wt./kg·mol ⁻¹	Purification method	Mass fraction purity ^a
D (–) Ribose (C ₅ H ₁₀ O ₅)	Loba Chemie Pvt. Ltd.	0.150	Recrystallization	>0.97
D (–) Raffinose (C ₁₈ H ₃₂ O ₁₆ ·5H ₂ O)	Loba Chemie Pvt. Ltd.	0.594	Recrystallization	>0.97
Sucrose	S.D. Fine-Chem. Pvt. Ltd.	0.342	Recrystallization	>0.98
D (+) Glucose	S.D. Fine-Chem. Pvt. Ltd.	0.180	Recrystallization	>0.98
Sodium dodecyl sulfate (C ₁₂ H ₂₅ NaO ₄ S)	Biochemical Grade	0.288	Recrystallization	>0.98

^a Declared by the supplier.

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