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Effect of tetrabutylammonium hydrogen sulphate on the Solution thermodynamics of thiamine hydrochloride in aqueous solutions

Abhijit Sarkar *, Biswajit Sinha

Department of Chemistry, University of North Bengal, Darjeeling 734013, India

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ABSTRACT

The densities and viscosities of thiamine hydrochloride (vitamin-B_1) in aqueous tetrabutylammonium hydrogen sulphate (Bu_4NHSO_4) solutions with several molal concentrations ($m_{Bu_4NHSO_4}$ = 0.000, 0.005, 0.010, 0.015 and 0.020mol·kg^{-1}) of Bu_4NHSO_4 were determined at T = (298.15, 308.15 and 318.15) K under atmospheric pressure. Using experimental data apparent molar volume (φ_V), standard partial molar volume (φ_V^0), the slope (S_V), standard isobaric partial molar expansibility (φ_E^0) and its temperature dependence$(\partial\varphi_E^0/\partial T)_P$, the viscosity *B*-coefficient and solvation number (S_n), etc., were determined. Viscosity *B*-coefficients were further utilized to obtain the free energies of activation of viscous flow per mole of the solvents ($\Delta\mu_1^{0\neq}$) and of the solute ($\Delta\mu_2^{0\neq}$). Effects of molality, solute structure and temperature on all these parameters were analyzed in term of ion-ion and ion-solvent interactions; results revealed that the solutions are characterized predominantly by ion-solvent interactions and thiamine hydrochloride behaves as a long-range structure maker.

1. Introduction

Studies on volumetric, viscometric and acoustic properties of food components in solution phase have been of much interest in recent years. For example, Rudan-Tasic et al. [1,2] studied the correlation between the volumetric properties of some cyclohexylsulfamates with their sweetening effects. C. Zhu et al. [3,4] studied the volumetric and viscometric properties of amino acids in aqueous solutions of different vitamins; similar works with food additives or food components are available in the literature [5–7]. Anyway, vitamins are organic compounds required by organisms as a vital nutrient to maintain their functions and developments. Thiamine or vitamin B_1 is a water-soluble vitamin. Being a coenzyme or their precursor for several major enzyme complexes (such as α–ketoglutarate dehydrogenase, pyruvate dehydrogenase and transketolase, etc.), it is crucial for carbohydrate metabolism and genetic regulatory processes [8–9]. It is also necessary for normal development and function of the brain and nerves [10]. Thus vitamins such as vitamin B_1 are essential nutrients for human body. But vitamins are not synthesized within the body and thus are must be supplemented in the food staffs as a food additive [11]. The recommended daily intake of thiamine for a healthy adult is 1.5 mg [12]. Phosphate derivatives of thiamine are involved in many cellular processes. It is required for the biosynthesis of neurotransmitter acetylcholine and γ-amino butyric acid. It is also used for digestive problem including poor appetite and ulcerative colitis. People also take thiamine for conditions related to low levels of thiamine including beriberi and inflammation of the nerves. Hence it finds widespread application as an additive in food and drug.

The stabilisation of bioactive molecules in solution phase is governed by a combination of several non-covalent interactions such as ion-dipolar, hydrophobic-hydrophobic and hydrophilic-hydrophobic group interactions, etc. These interactions are greatly affected by the surrounding solvent and cosolute molecules; that is why the physicochemical behaviours of vitamins are strongly influenced by the presence of cosolutes specially salts in aqueous phase. Tetraalkylammonium salts are bulky in nature and are known as water orienting water molecules depending on their alkyl chain lengths [13–14]. Therefore aqueous solution of tetraalkylammonium cations can stand as model systems for the study of hydrophobic hydration and salts like tetrabutylammonium hydrogen sulphate (Bu_4NHSO_4), an ion-pairing reagent [15], can provide better information about the effect of different non-covalent interactions on the stabilities of vitamins.

Although few studies are available on various properties of thiamine hydrochloride in aqueous solutions [16–18], such a study on solution properties of thiamine hydrochloride in aqueous Bu_4NHSO_4 solution is not available so far in the literature. Hence the present work aims to study the various interactions interplaying in the aqueous solutions of thiamine hydrochloride and Bu_4NHSO_4 in terms of apparent molar

* Corresponding author.
E-mail addresses: abhijitslg4@gmail.com (A. Sarkar), biswachem@gmail.com (B. Sinha).

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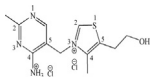
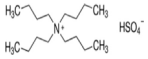
Table 1
Provenance and purity of the chemicals used.

Chemical	Structure	Source	Purification method	Mass fraction purity	CAS No
Thiamine hydrochloride		Sigma-Aldrich, Germany	none	0.990	67-03-8
Tetrabutylammonium hydrogen sulphate		S. D. Fine Chemicals, India	none	0.985	32503-27-8

volumes, standard partial molar volumes and viscosity *B*-coefficients, etc., at $T = (298.15, 308.15 \text{ and } 318.15)$ K under atmospheric pressure.

2. Experimental

2.1. Chemicals

Reagent grade thiamine hydrochloride (CAS: 67-03-8; Sigma-Aldrich, mass fraction purity > 0.990) and A. R grade Bu_4NHSO_4 (CAS: 32503-27-8, S. D. Fine Chemicals, India; mass fraction purity > 0.985) were used for present study (Table 1).

Thiamine hydrochloride and Bu_4NHSO_4 were used without further purification but it was dried in vacuo over anhydrous $CaCl_2$ for several hours before use. Doubly distilled de-ionized water with a specific conductance $< 1\cdot10^{-6}$ S cm^{-1} at 298.15 K was used to prepare different aqueous solutions of Bu_4NHSO_4. Various mixed solvents were prepared by mass and necessary adjustments were done to achieve exact molal concentrations ($m_{Bu_4NHSO_4}$ = 0.005, 0.010, 0.015 and 0.020 mol·kg^{-1}) of Bu_4NHSO_4 in the mixed solvents at 298.15 K. The physical properties of aqueous solutions of Bu_4NHSO_4 are found same as our previously published paper [19]. Stock solutions of thiamine hydrochloride in different solvent mixtures were prepared by mass and all the working solutions were prepared afresh before use by mass dilution. A Comparison of density and viscosity data of aqueous thiamine hydrochloride with available literature data has given graphically in Fig. 1 and Fig. 2. The mass measurements were made on a digital electronic analytical balance (Mettler, AG 285, Switzerland) with an uncertainty of $\pm 1\cdot10^{-4}$ g. The conversion of molalities into molarities was accomplished by using experimental density data whenever needed [20]. Estimated uncertainty in molality of thiamine hydrochloride solutions was evaluated to $\pm 1\cdot10^{-4}$ mol·kg^{-1}.

2.2. Apparatus and procedure

The densities were measured with a vibrating-tube density meter (Anton Paar, DMA 4500 M). The densitometer was calibrated at the experimental temperatures with doubly distilled, degassed water and dry air at atmospheric pressure. The temperature was automatically kept constant with an accuracy of $\pm 1\cdot10^{-2}$K using the built–in Peltier technique. The stated repeatability and accuracy of the densities were $\pm 1\cdot10^{-5}$g· cm^{-3} and $\pm 5\cdot10^{-5}$g· cm^{-3}, respectively. However, the standard uncertainty of the density measurements for most of the solutions was found to be within the range $\pm$ 0.1 kg·m^{-3}.

The viscosity was measured by means of a suspended Canon-type Ubbelohde viscometer thoroughly cleaned, dried and calibrated at the experimental temperatures with triply distilled, degassed water and purified methanol [21,22]. It was filled with an experimental liquid and placed vertically in a glass sided thermostatic bath (Julabo, Germany) maintained constant to $\pm$0.01 K. After attainment of thermal equilibrium, the efflux times of flow of the liquid samples were recorded with a digital stopwatch correct to $\pm$0.01 s. Adequate precautions were adopted to minimize evaporation loses during the viscosity measurements and an average of triplicate measurements was taken into account. Standard uncertainty in viscosity measurements was within $\pm$0.003 mPa s.

The absorption spectra of thiamine hydrochloride in aqueous solutions of Bu_4NHSO_4 were recorded on Jasco V-530 double beam UV-VIS Spectrophotometer. It was coupled with thermostatic arrangement and maintained at 298.15 K. A quartz cell of 1 cm path length was used and spectroscopic grade water was used as the reference solvent for all the absorption measurements. During spectrophotometric measurements, 2 mL of either thiamine hydrochloride or Bu_4NHSO_4 solution ($1\cdot10^{-4}$mol·L^{-1}) was placed in the quartz cell and the absorbance of

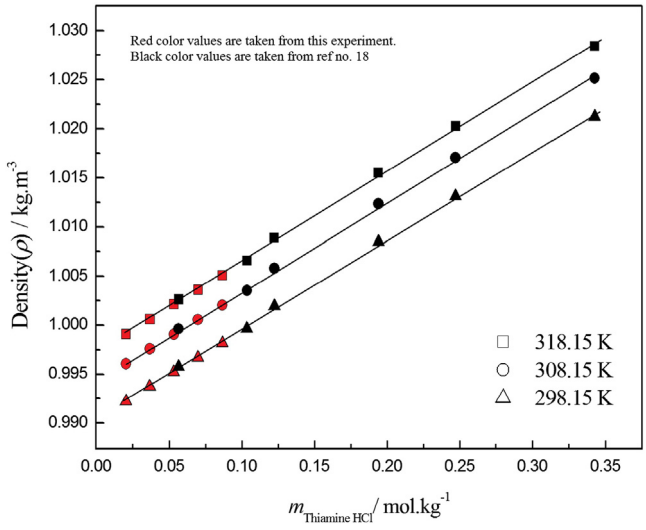


Fig. 1. Comparison of density values of aqueous thiamine hydrochloride solutions with ref. number 18.

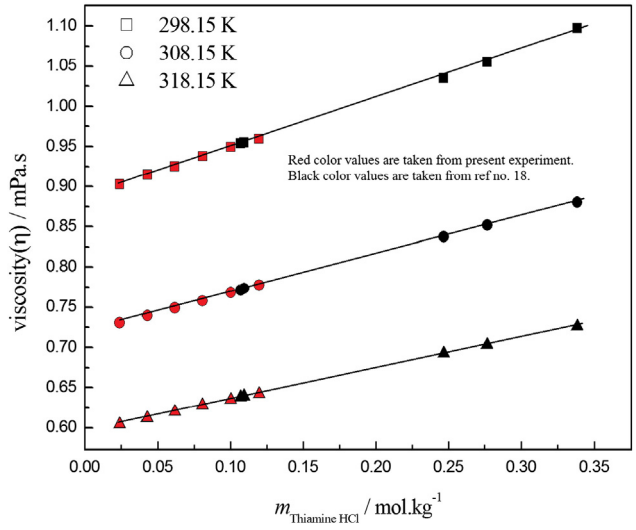


Fig. 2. Comparison of viscosity values of aqueous thiamine hydrochloride solutions with reference number 18.