



Effect of tartarate and citrate based food additives on the micellar properties of sodium dodecylsulfate for prospective use as food emulsifier



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ABSTRACT

Citrate and tartarate based food preservatives can be used to enhance the emulsifying properties of sodium dodecylsulfate (SDS) based micellar system and thus making it appropriate for food applications. Exploration of interactions between the two species is the key constraint for execution of such ideas. In this work various micellar and thermodynamic parameters of SDS like critical micellar concentration (CMC), standard Gibbs free energy of micellization (ΔG_{mic}^0) etc. have been calculated in different concentrations of disodium tartarate (DST) and trisodium citrate (TSC) in the temperature range (288.15–318.15) K from the conductivity and surface tension measurements. The parameters obtained from these studies reveal the competitive nature of both the additives with SDS for available positions at the air/water interface. TSC is found to be more effective additive in order to make SDS micellar system better for its potential applications as food emulsifier.

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1. Introduction

Citric acid is an important intermediary compound of a chain of biochemical reactions, used by all aerobic organisms to generate energy from carbohydrates, proteins and fats, known as Krebs' cycle/tricarboxylic acid cycle (Fisher, 1977). In contrast to it, tartaric acid is an inhibitor of Krebs' cycle and is purely yeast byproduct. Although citric and tartaric acid have different biological activities but the metal ion complexing properties of these are similar and make them useful as sequestrants, antioxidants and preservatives resulting in some common applications in food and agriculture industry (Shibata, Yamaguchi, Takeda, & Masai, 1980; Wassay, Barrington, & Tokunaga, 1998). U. S. Food and Drug Administration has reviewed the safety of trisodium citrate (TSC) and disodium tartarate (DST) as food additives and these are reported as GRAS materials (Generally Regarded as Safe) (Sec.

582.1751, Sec. 184.1801). European Food Safety Authority has also approved the use of DST and TSC as permitted food additives and is added to the list of antioxidants with E numbers 335 and 331, respectively. It has been reported that sodium salts of citrate and tartarate are used as acidity regulators, food preservatives, anticaking agents, flavour enhancers, colour retention agents as well as stabilizers (Arslan, Ilhan, & Karabulut, 2006; Vibhkara, Gupta, Jayaraman, & Mohan, 2006). Another method for preventing food materials from putrefaction comprises the admixing of acetic acid and salts of citric, tartaric, malic as well as lactic acid (Shibata et al., 1980). Positive antimicrobial effects of sodium citrate plus sodium diacetate or buffered vinegar against *Escherichia coli* 0157:H7 when included in beef enhancement solutions have been reported by Ponrajan et al. (2012). It is clear that in many instances the use of citrate and tartarate salts along with other additives is must to make flavours sharper and making preservation and stability of food products even better.

Numerous examples in literature can be found where surfactants have been used as emulsifiers (Giannoulis et al., 2013; Ravi,

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Manohar, & Rao, 2000). Sodium dodecylsulfate (SDS) is one of the most commonly studied surface active agents which find their applications as emulsifiers by allowing the coexistence of isotropic and thermodynamically stable mixture of water and oil. SDS has been regarded as multipurpose additive in food products for human consumption by the U. S. Food and Drug Administration (FDA, 2012) although its use as a food additive is not permitted in European legislation (Cid et al., 2014). Emulsification properties of any surfactant are mainly determined on the basis of Hydrophile – Lipophile Balance which is also a measure of surfactant partitioning tendency between oil and water (Guo, Rong, & Xugen, 2006). SDS is the surfactant which is used for the preparation of oil in water type emulsions on the basis of the HLB values calculated by Davies rules (Davies & Rideal, 1961).

By having an idea about the nature of interactions existing between food additives and surface active agents from the thermodynamic profile of micellization, the microstructure and properties of surfactants can be tuned for various applications by changing the concentration of added salts. The broad spectrum microbial or germ killing properties of SDS when combined with these salts provide additional benefits and biodegradable compositions of these have been used for cleaning and disinfecting nasal tract and sinus cavity (Lopes, 2008).

Effects of various bioactive molecules such as amino acids and butylated hydroxytoluene (a phenol derivative added to many food materials to prevent food spoilage) have been studied on the micellar properties of anionic and cationic surfactants. (Bhardwaj, Chauhan, & Sharma, 2014; Yu, Lu, Luan, Liu, & Xu, 2005). Thermodynamic properties of SDS-Salicylic acid based micellar system have been studied (Cid et al., 2014) for potential uses in fruits postharvest. Recently, a report on binding characteristics of food additive curcumin (E number 100) with sodium bis (2-ethylhexyl) sulfosuccinate (AOT) based on spectrometric studies has been presented (Zhou, Yang, & Wang, 2014).

The major goal of the present work would be to choose suitable concentrations of the added salts so that the micellar combination of SDS provides better emulsification properties. So, an attempt has been made to have an insight in the micellization behaviour of SDS in water as well as in various concentrations of sodium salts of citrate and tartarate (0.0005, 0.0010, 0.0015, 0.0020, 0.0050, 0.0075 and 0.0100 M) at different temperatures. Further the limited use of SDS in food additives can be overcome by increasing the effective concentration of it when used in combination with the permitted edible salts.

2. Experimental

2.1. Materials and methods

SDS was purchased from Sigma–Aldrich; TSC and DST were supplied by Sisco Research Laboratories (SRL, India). All these chemicals were having purity higher than 99%. These were dried over anhydrous CaCl_2 in vacuum desiccator before use. Solutions were prepared in second stage Milli-Q water having specific resistance of 18.2 mega ohm cm. The solutions were prepared using Mettler balance having an accuracy of ± 0.01 mg. The standard uncertainty in the molarity on an average is $\pm 1.2 \cdot 10^{-5}$ mol L^{-1} .

2.2. Equipments and procedure

2.2.1. Conductivity measurements

Conductivity measurements were carried out with a digital conductivity meter (Systronics-306) at a fixed frequency of 50 Hz using a dip type cell with double walled glass jacket. The conductivity meter was calibrated using KCl conductivity standard

solution ($[\text{KCl}] = 0.1000$ mol L^{-1} , $\kappa = 12.97$ mS cm^{-1}). The temperature of the cell was maintained using a thermostat bath (Model F25 Julabo, Germany) having a temperature control of ± 0.01 K. Sample solutions were homogenised by stirring with a magnetic bar. The reproducibility of the conductivity measurements was within $\pm 0.2\%$.

2.2.2. Surface tension measurements

The ring detachment method was used to measure the surface tension (γ) values of the aqueous solutions of surfactants with a Kruss Easy Dyne Tensiometer from Kruss GmbH (Hamburg, Germany). The Platinum ring used in the measurements was cleaned by washing with Milli-Q water followed by drying with acetone. Surface tension of Milli-Q water = 72 mN m^{-1} at 298.15 ± 0.1 K was used for calibrating the instrument. Each surface tension value reported was an average of at least 3 measurements. The accuracy in the measurement of surface tension with tensiometer is ± 0.15 mN m^{-1} .

3. Results and discussion

3.1. Conductivity measurements

Micellization behaviour of sodium dodecylsulfate (SDS) has been studied in water as well as in aqueous solutions of various concentrations of disodium tartarate (DST) and trisodium citrate (TSC) over a temperature range (288.15–318.15) K. Plots for specific conductance of SDS versus concentration in the presence of various concentrations of DST and TSC at 298.15 K are shown in Fig. 1 and the break in the curve represents the CMC of the surfactant. CMC values of SDS in water at different temperatures are in good agreement with those reported in literature (Banipal, Kaur, Banipal, & Sood, 2014) (Table 1 and S1) (as Supplementary information). An increase in CMC values is observed with temperature in the studied range. Temperature influences the micellization behaviour of surfactants in two ways. Increase in temperature decreases the degree of hydration of hydrophilic groups of surfactant which favours micellization, but it also breaks water structure around the hydrophobic groups which opposes the tendency of micellization (Varade et al., 2005). For ionic surfactants CMC decreases in the low temperature region, reaches a minimum and then increases. However in the present studied range, a continuous increase in the CMC values of SDS in water is observed which indicates the predominance of second effect on the micellization behaviour of SDS. The CMC values of SDS decrease with concentration of DST and TSC (Table 1 and S1). These salts behave as electrolytes and electrolyte induced changes in the micellization behaviour of ionic surfactants are more pronounced as compared to those of zwitter ionic and nonionics (Rosen, 2004). Therefore decrease in the CMC of surfactants in the presence of electrolytic solutions is mainly due to the decrease in the thickness of ionic atmosphere surrounding the ionic head groups and consequently decreased electrostatic repulsions between them. Additional effect of such additives on the hydrophobic parts of surfactants is their ability, to make the water structure and salting out the hydrophobic groups of monomeric form of surfactants.

A study of micellization properties of AOT in aqueous solutions of sodium chloride (NaCl), sodium acetate (NaAc), sodium propionate (NaPr) and sodium butyrate (NaBu) at 298.15 K reveals that co-anion of electrolyte does not affect the CMC values and these are dependent on Na^+ concentration in the solution (Umlong, Dey, Chanda, & Ismail, 2007). For SDS, CMC variation in aqueous NaAc, NaPr and NaCl solutions shows that at lower concentrations, values are independent of co-anion's nature; however at higher concentrations, different behaviour for different electrolytes is

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