



Surface and foaming properties of polyoxyethylene glycerol ester surfactants



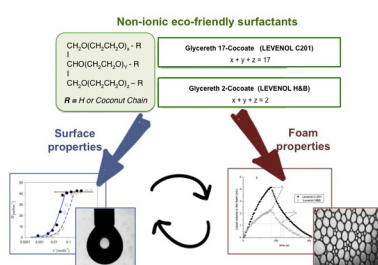
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HIGHLIGHTS

- The surface properties of two environment-friendly surfactants were studied.
- A Frumkin model assuming the compressibility of the adsorbed film was used.
- The diffusion coefficient of Levenol C-201 is higher than Levenol H&B.
- Foam stability correlates with surface dilatational elasticity.

GRAPHICAL ABSTRACT



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ABSTRACT

Polyoxyethylene glycerol esters derived from cocoa oil are non-ionic surfactants obtained from a renewable source which fulfil the environmental and toxicological requirements to be used as ecofriendly foaming and/or emulsifying agents. This paper reports a study on the equilibrium adsorption, surface rheology and foaming properties of two commercial polyoxyethylene glycerol ester surfactants which differ in the number of ethylene oxide (EO) groups. Dynamic and equilibrium surface pressure values were obtained with a drop profile tensiometer. The oscillating drop technique was used to study the dilatational rheology of adsorbed surfactant layers. The foaming properties of aqueous solutions of these surfactants were characterized in a commercial foam scan column. Dynamic surface pressure measurements showed two adsorption processes in which a slower condensation of the adsorbed film followed a fast diffusion step. Surface rheology and equilibrium surface pressure data fitted an extended Frumkin model which takes into account the compressibility of the adsorbed layer. The polyoxyethylene glycerol ester with the highest number of EO groups turned out to be more surface active, leading to lower cmc and higher adsorption constant. Both surfactants exhibited similar overall foam capacity at a given concentration. However, the time evolution of the liquid volume in the foam and the size of the air bubbles pointed to the formation of a more homogeneous and stable foam with smaller bubbles for the most hydrophilic polyoxyethylene glycerol ester studied. This is considered to be related to its surface dilatational rheology properties.

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1. Introduction

Most of the surfactants currently used in the food and cosmetic industries are produced by the chemical transformation of animal and plant fats or petroleum derivatives [1]. Nevertheless, surfactants obtained directly from renewable natural materials are gaining more and more attention since they are more eco-friendly than traditional ones [2–5].

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Polyoxyethylene-based non-ionic surfactants are widely used in industrial applications such as detergents, food dispersions, cosmetics and personal care products, agricultural formulations, petroleum industry applications, and in general to stabilize foams and emulsions [6,7].

Polyoxyethylene glycerol esters derived from cocoa oil are non-ionic surfactants obtained from a renewable source which fulfil the environmental and toxicological requirements to be used as foaming and/or emulsifying agents [8]. These surfactants are fully innocuous for human skin and hair [9] and their properties are adequate in order to design eco-friendly products [10]. Therefore, they have been used for the formulation of detergents, personal care products and cosmetics [11–14]. At present, there is a marked trend towards the use of eco-friendly surfactants derived from inexpensive renewable resources in detergent formulation [15].

Several works have related the surface properties of polyoxyethylene-based non-ionic surfactants with technical applications as foam and emulsion stabilizers [16–18]. The influence of the hydrocarbon chain length as well as of the number of ethylene oxide (EO) groups on the adsorption of these non-ionic surfactants at the air/water [19–21] and oil/water [22,23] interfaces has been previously reported. However, the surface properties of polyoxyethylene glycerol esters derived from cocoa oil, namely equilibrium adsorption, dynamic surface tension and surface rheology, have not yet been studied. Taking into account the fact that most of the technical applications of these surfactants involve events occurring at the interfaces, gaining a deeper knowledge of the interfacial properties is of key importance in order to rationally design new formulations.

Foams are important in many products and process technologies, especially for food dispersions and detergents [24]. A foam is a dispersion of gas bubbles separated by thin liquid films and its formation and stability depends on the properties of the surface-active components in the system, especially on the dilatational properties of the adsorbed film [25,26].

In the present work adsorption, surface rheology and foaming properties of two eco-friendly polyoxyethylene glycerol ester surfactants with different numbers of EO groups have been studied. An extended Frumkin model was used to describe the adsorption process and surface rheology of both surfactants. The combination of measurements along with the theoretical fitting of the experimental curves provides a thorough characterization, enabling the interpretation of interfacial conformation at a molecular level.

2. Materials and methods

2.1. Materials

Two commercial polyoxyethylene glycerol ester surfactants derived from cocoa oil have been studied. These surfactants differ in the number of EO groups. Namely Levenol® C-201 (Glycereth-17 cocoate) and Levenol® H&B (Glycereth-2 cocoate), which are technical grade surfactants with 17 and 2 average EO groups and HLB numbers 13 and 11.3, respectively. They were kindly provided by KAO and used as received. The solutions were prepared with Milli-Q water. All glassware was cleaned with sulfuric acid (98%) containing 8 g L⁻¹ of ammonium disulfure.

2.2. Drop profile tensiometry

Surface tension measurements were performed with a drop profile analysis tensiometer (CAM200, KSV, Finland). The drop was formed inside a thermostatted cuvette at 20 °C and controlled using a custom-built control unit consisting of a syringe with a piston that is driven by a stepper motor. The control procedure was as follows:

once the drop was formed the contour of the drop was acquired and then the drop initial area calculated. Every 10 s the area was calculated and the actual and initial value were compared. Whenever the values differed, the stepper motor drove the piston in the respective direction to correct the difference.

Oscillation perturbations were made using a PD 100 module from KSV. This module consists of a chamber filled with the solution to be measured in which there is a piezoelectric device calibrated with the pc sound card, which produces harmonic oscillations.

Prior to each measurement the surface tension of the water, γ_0 , was measured to check that its value was constant and between 72 and 73 mN m⁻¹. Surface pressure values are calculated as: $\Pi = \gamma_0 - \gamma$.

2.3. Foaming properties

The foaming properties of Levenol® C-201 and Levenol® H&B solutions were measured using a foam scan column (Foamscan IT Concept, Longessaigne, France). This device, based on an idea by Popineau et al. [27,28], allowed us to determine the foam formation, the foam stability and the drainage of the liquid from the foam by conductimetric and optical measurements [25]. The foam was generated by blowing gas (nitrogen) at a flow of 45 mL min⁻¹ through a porous glass filter with a pore diameter of 0.2 μ m. This was located at the bottom of a glass tube where 20 mL of the aqueous surfactant solution was placed. The foam was allowed to reach 120 mL of volume. Then, the bubbling was stopped and the evolution of the foam was monitored. The amount of liquid incorporated into the foam, the foam homogeneity, and the liquid drainage from the foam were followed by measuring the electrical conductivity in the cuvette containing the liquid sample and at different heights in the glass column by means of suitable electrodes.

Two parameters were determined as a measure of foaming capacity, the overall foaming capacity (OFC, mL s⁻¹) and the foam capacity (FC). The former is a measure of the rate of foam formation and was determined from the slope of the foam volume curve while bubbling. The latter is a measure of the gas retention in the foam and was determined by Eq. (1)

$$FC = \frac{V_{\text{foam}(f)}}{V_{\text{gas}(f)}} \quad (1)$$

where $V_{\text{foam}(f)}$ is the final foam volume and $V_{\text{gas}(f)}$ is the final gas volume injected.

The half-life time ($t_{1/2}$) is the time needed to drain half of the volume of the liquid contained in the foam. It was used as a measure of foam stability. This was also determined by following the development of bubbles in the foam. The Foamscan instrument is equipped with a CCD camera set that has a macro objective to capture the variation of air bubble sizes in the foam at 5 s intervals and at a foam height of 10 cm.

Foaming properties were measured at 20 °C from aqueous solutions at a concentration of 1 g L⁻¹. All experiments were performed three times. The reported values are the average values and the corresponding standard deviation.

3. Theory

3.1. Equilibrium isotherm

The theoretical model used to fit the experimental data of adsorption was developed by Fainerman and co-workers and it is referred to as the Frumkin-A model. This model combines the Frumkin-type adsorption mechanism with the assumption that the area occupied by the adsorbed molecules, Ω , is a function of surface pressure, Π . This model has been successfully applied to explain the adsorption isotherm of several surfactants [29–31]. Hence, the

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