



Foamability of fatty acid solutions and surfactant transfer between foam and solution phases



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ABSTRACT

The foaming properties of aqueous solutions of fatty acids with different hydrocarbon chain lengths - hexanoate, laurate, and oleate - were assessed through measurements of foam volume and growth rate under continuous aeration. Simultaneous measurements of the partition of each surfactant between the bulk solution and foam phases were also carried out. Although bubble size measurements in bulk solution pointed towards quite similar gas dispersing abilities of the tested surfactants, their foamabilities under the same conditions were remarkably enhanced with increasing chain length of the hydrocarbon tail. Shorter chain surfactants - hexanoate and laurate - generated wet foams and the accumulation of these surfactants in the foam phase was very low, indicative of a low adsorption density of the reagents at the gas-liquid interface. In contrast, sodium oleate produced stable dry foams, and the amount of oleate transferred to the foam was much higher. The formation of these different types of foam was attributed to the higher adsorption of oleate at the gas-liquid interface compared to the lower adsorption of hexanoate or laurate, and to different mechanisms of foam stabilization. Regardless of the interfacial mechanism of the foaming action of the tested surfactants, it was generally found that strong foaming was possible only from surfactant solutions whose bulk concentration was higher than the critical coalescence concentration of a given surfactant. It was concluded that bubbles entering the foam phase from the bulk solution phase must already be stable against coalescence to induce foam formation and impart stability to the entire foam column. Foaming was observed only in true solutions of the tested surfactants, while precipitation of colloidal acid species completely depressed foam formation.

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1. Introduction

A froth flotation system can be divided into a bulk solution phase and a foam/froth phase. The effect of a reagent on the behavior of gas bubbles in the bulk solution phase is most often assessed through bubble size and gas hold-up measurements. Such tests are collectively known as gas dispersion measurements. One of the key quantities characterizing the gas dispersion capabilities of a surfactant is the critical coalescence concentration, which depending on the method through which it is obtained, can be designated as CCC (Cho and Laskowski, 2002) or CCC95 (Finch et al., 2008). The critical coalescence concentration can be viewed as such a surfactant concentration above which the coalescence of bubbles in bulk solution is prevented and gas bubbles attain their lowest sizes for a given set of hydrodynamic conditions.

The vast majority of studies on foamability from surfactant solutions have been carried out under steady-state foaming condi-

tions, or when the foam initially grows in height at a given gas flow rate, and then reaches a steady state height/volume when the foam growth and foam breakage processes cancel out. Steady-state foams are often referred as wet foams and the relatively high water content in such foams results from slow drainage of water from the foam phase. As gas bubbles enter the foam phase, their movement induces a concentration gradient of surfactant molecules between the top and the bottom sections of the bubble. Viscous drag forces surfactant molecules to accumulate at the bottom surface of the moving bubble generating a surface tension gradient. However, the natural tendency of surfactant molecules to diffuse to offset concentration gradients will tend to redistribute the molecules around the bubble surface, and this surfactant diffusion opposes the downward flow of water through the films separating the gas bubbles, a phenomenon known as the Marangoni-Gibbs effect. As a result, these competing processes retain a portion of water that would otherwise drain freely from the foam and thus the water content in wet steady-state foams is relatively high, usually around 30% (Małysa, 1992). As stressed in several reviews of the subject (Sheludko, 1967; Małysa, 1992; Pugh, 1996; Małysa and

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Lunkenheimer, 2008), the above phenomena can take place only under dynamic conditions, so as soon as bubbling stops the entire column of wet steady-state foam collapses very quickly, often within seconds.

The dynamic behavior of wet steady-state foams can be quantified in terms of the water overflow rate, foam volume and growth rate, the dynamic frothability index (*DFI*) and the retention time (*rt*), just to name the most common parameters (Bikerman, 1953; Czarnecki et al., 1982). The work of Małysa et al. (1991) on fatty acids of shorter chain lengths (C_5 – C_9) showed that those surfactants formed steady-state foams from dilute HCl solutions (0.005 mol/L). However, oleic acid (C_{18}) solutions at low pH do not foam, while oleate solutions at high pH typically do not reach steady state conditions (Atrafi et al., 2012). Oleate foams grow continuously with time, and it can actually be observed during the test that no air escapes from the foam column, which is also consistent with reports of froth stability issues during the fatty acid flotation of salt-type minerals in the presence of excess fatty acid (Vilinska et al., 2011). As a result, a comparison of foaming from oleate solutions with the behavior of shorter chain homologues becomes very difficult using the tests applicable strictly to steady-state foams. The foam formed by sodium oleate appears to be composed of large polyhedral bubbles, and remains stable for a long of time even after aeration stops (Atrafi et al., 2012). These characteristics are typical of dry foams (Małysa, 1992; Pugh, 1996).

Dry foams tend to form from solutions of highly surface active substances characterized by a high adsorption density at the gas-liquid interface (Pugh, 1996). The high adsorption density of the surfactant leads to a fast relaxation of surface tension gradients around gas bubbles and to weakening of the Marangoni effect. Consequently, the drainage of water from the foam occurs unopposed. For wet steady-state foams, the thickness of the liquid films separating gas bubbles is relatively large, typically more than 100 nm. However, the high concentrations of the surfactant at the bubble surfaces of a well-drained foam can facilitate the formation of a different type of much thinner films, provided that the foam can withstand the initial drainage process. At such short separation distances, the stability of the film against thinning and rupture is influenced by surface forces (electrostatic, van der Waals, structural) and by specific intermolecular interactions between the adsorbed surfactant layers. In the extreme, very thin films (4–10 nm) of exceptional stability may form, whose structure can be represented as a thin layer of a few water molecules sandwiched between two layers of the adsorbed surfactant molecules on the adjacent gas bubbles (Gamba et al., 1992). These films are commonly known as “black films” from their appearance as nearly black areas on bubbles surfaces, representing the thinnest domains of the film, when optical methods are used to measure the film thickness (Pugh, 1996). As noted by Małysa (1992), dry stable foams initially form as wet foams, and then become dry as a result of drainage processes, so in real foams the water content decreases with the foam height.

In the present study, the foamability of solutions of several fatty acids of different chain lengths - hexanoic, lauric, and oleic - is investigated in more detail. The gas dispersion properties of the same surfactants were already reported in an earlier contribution (Atrafi and Pawlik, 2016). Although it is recognized that bubble “history” has a major impact on the properties of the resulting foam (Jachimska et al., 2001; Małysa and Lunkenheimer, 2008), no direct data have been provided for continuously aerated systems to assess that connection. It is the main objective of this work to probe the role of gas dispersion processes in the bulk solution phase in the foaming phenomena from fatty acid solutions.

The term foamability, as opposed to frothability, is used throughout this work to denote the behavior of a two-phase system (air-solution) since no solids were used in the experimental

program. Any data and comments used in the discussion regarding experimental conditions, surface tension, gas bubble sizes, CCC95 values, or reagent speciation are based on our earlier paper (Atrafi and Pawlik, 2016), unless indicated otherwise.

2. Materials and methods

2.1. Reagents

Sodium oleate powder, (>82% fatty acids as oleic acid) ($C_{18}H_{33}O_2Na$) was obtained from Sigma Life Science. Sodium laurate (dodecanoate), 98% pure ($C_{12}H_{23}O_2Na$), and sodium hexanoate, 99% pure ($C_6H_{11}O_2Na$) were obtained from Acros Organics as white powders. Sodium dodecyl sulfate, 99% ($C_{12}H_{25}SO_4Na$) was obtained from Acros Organics, while sodium *n*-octadecyl sulfate ($C_{18}H_{37}SO_4Na$), 99% (dry weight) was obtained from Alfa Aesar. A sample of MIBC, (methyl isobutyl carbinol, $C_6H_{14}O$) or 4-methyl-2-pentanol, 98% pure, was obtained from Sigma Aldrich. All the surfactants were introduced as aqueous solutions. The pH of tested solutions was adjusted using analytical grade sodium hydroxide and hydrochloric acid solutions. Analytical grade sodium chloride (Fisher Scientific) was used as a background electrolyte.

In contrast to the carboxylic acids, the tested alkyl sulfates are strong electrolyte-type anionic surfactants fully dissociated in aqueous solutions regardless of pH. As such, their effect on foaming properties provides a baseline measure of the behavior of true solutions of surfactants of the same chain length, i.e., in the absence of colloidal species forming when the solubility of the associated acid species is exceeded.

2.2. Foamability measurements

The measuring set up for foamability studies consisted of three glass columns, each 1-meter high with an internal diameter of 4.3 cm. Each column had a porous glass plate at the bottom for producing air bubbles. Air was independently supplied to each column through a separate digital mass flow meter (ProStar PRS10FRC). Each flow meter was calibrated for airflow at a temperature of 25 °C and pressure of 0.1013 MPa (1 atm.). The airflow rates were controlled by a digital flow controller (ProStar CM4) and the flow rate could be adjusted and maintained between 100 and 1000 cm³/min. Since the columns were connected in-parallel to the air supply system, three different sample solutions could simultaneously be tested at different gas flow rates. This digital gas flow control system generated flow rates reproducible within 1% of the pre-set value.

The approach presented by Atrafi et al. (2012) was used for foamability measurements, which involved a continuous measurement of the foam height as a function of time at a given air flow rate. Then, the total gas volume contained in the system was determined and recorded as a function of time. The slope of such a plot is a measure of the froth growth rate. The total gas volume was calculated as the difference between the initial solution volume and the combined volumes of solution and foam at a given time after turning on the gas flow. The procedure was to introduce 200 mL of a test solution into a column, and the air flow rate was adjusted to 400 mL/min, equivalent to a superficial gas velocity, J_g , of about 0.5 cm/s. The same superficial gas velocity was used in our earlier work on gas dispersion phenomena in fatty acid solutions so that correlations can be established between the gas dispersion and foamability results.

The standard deviation of foamability measurements was 15–25 mL at a given measuring time, and the size of the data symbols is an accurate measure of the experimental errors.

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