



Competitive adsorption of surfactant–protein mixtures in a continuous stripping mode foam fractionation column



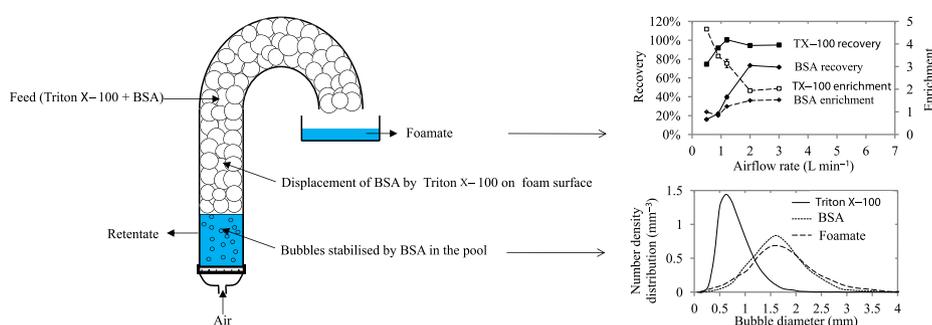
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HIGHLIGHTS

- Competitive adsorption shown in a binary stripping mode foam fractionation column.
- Analytical characterisation of protein (BSA) and surfactant (Triton X–100) system.
- Foam stabilised by BSA with negligible Triton X–100 in bottom product.
- Triton X–100 has higher surface activity than BSA and displaced it from interface.

GRAPHICAL ABSTRACT



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ABSTRACT

In this paper a detailed experimental study on the application of continuous stripping mode foam fractionation to separate a model surfactant–protein mixture was performed with emphasis on the competitive adsorption behaviour and transport processes of surfactant–protein mixtures in the rising foam column. Bubble size measurements of the foamate showed that at steady state conditions the bubbles rising from the liquid pool were stabilised by BSA. However at the top of the column the recovery of Triton X–100 in the foamate (75–100%) was always greater than the recovery of BSA (13–76%) for all foam fractionation experiments. The enrichment of BSA remained at almost unity for experiments with high feed concentrations of both components and low air flow rates, and only increased when the recovery of Triton X–100 reached 100%. Thus it was concluded that Triton X–100 displaced the adsorbed BSA from the surface. The surface activity and diffusivity of the two components was determined from surface tension and nuclear magnetic resonance (NMR) measurements. These results illustrated that competitive adsorption behaviour was due to the greater maximum surface pressure (2.05 times) and diffusivity (19.6 times) of Triton X–100 than BSA. In addition to investigating the effect of foam fractionation process parameters on the separation of mixed systems, the results from the characterisation studies of surface adsorption and foam properties provided insight and deeper understanding of the competitive adsorption behaviour of surfactants and proteins in a foam fractionation process.

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1. Introduction

Mixed surfactant protein systems are an important class of chemical in industrial products and processes. In the food industry surfactant–protein mixtures such as fatty acids and whey proteins are widely used for the stabilisation of foams and emulsions

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(Miller et al., 2005). Surfactant–protein mixtures are particularly significant in bioproducts such as biosurfactants and biopolymers where fermented products tend to be dilute mixtures of surface active components such as surfactant and protein (Winterburn and Martin, 2012). This complicates the downstream process and makes a universalised approach difficult. Currently a major barrier to commercialisation of bioproducts is expensive and inefficient downstream processing, for example Burghoff (2012) estimates these as 80 per cent of production costs. Thus there is a need to understand the separation science of such mixtures to improve the efficiency and effectiveness of downstream processing of such systems.

Foam fractionation has drawn most attention due to its low cost, mild conditions, sustainable approach where solvents are not required, the ability to process dilute solutions with high separation efficiency and the possibility for batchwise or continuous operation (Stevenson, 2012). It is a foam based separation process based on the preferential adsorption of surface active molecules on to an air–water surface (Lemlich, 1968). In a foam fractionation process, foam is created and stabilised at the bottom of a vertical column. Depending on factors such as pool volume, residence time of bubbles in pool and air flow rate, the foam may or may not be in equilibrium. The foam generated in a foam fractionation process consists of gas bubbles surrounded by liquid film lamellae. The lamellae intersect to form a network of Plateau borders and these connect at vertices. As the foam rises up the column, downwards drainage of interstitial liquid occurs in the Plateau border and vertices due to gravity and capillary forces. The contribution of capillary drainage typically becomes negligible by a foam height of around 50 mm such that the rising foam approaches a constant liquid hold up with column height (Martin et al., 2010). This foam product often called the foamate is enriched in surfactant and is collapsed to form a surfactant rich solution (Lemlich, 1972).

Foam fractionation can be operated in batch or continuous modes. The simplest mode of operation is the batch mode where an aqueous surfactant solution is sparged with gas until the surfactant concentration in the pool falls below the foaming concentration. However for steady state conditions to be achieved the process has to be run in continuous mode. There are two modes of continuous operation; simple and stripping mode. In a simple continuous process, feed is continuously fed into the bottom liquid pool whilst foam and a fraction of the bottom liquid pool are removed from the top and bottom of the column respectively. In a continuous stripping mode foam fractionation process, feed is injected near the top of column into the rising foam. Below the feed point a relatively wet rising foam is created; above the feed point the liquid in the foam drains and the foam becomes drier.

Bubbles are stabilised in the bottom pool and rise as a foam up the column. The feed will predominantly drain downwards through the centres of the Plateau border channels of the foam with some mixing at the vertices. Surfactant in the feed has the opportunity to diffuse from the centre of the Plateau border channels to their subsurface, and then adsorb to the interface. From the Plateau border–air interface, surfactant transport onto and throughout the lamellae films can occur through diffusive and Marangoni surface effect, which are coupled with the film drainage. The relative diffusive, adsorptive and interface properties of the different surfactant species can result in varying ratios of surface excess and therefore different separation effects. The transport and flow of surfactant through Plateau borders and on to foam lamellae have been described in previous theoretical studies (Vitasari et al., 2013b; Grassia et al., 2016; Vitasari et al., 2016).

The adsorptive separation of multicomponent systems in a foam fractionation process has been widely reported in the literature. The reported multicomponent systems include protein mixtures commonly found in milk and egg (Brown et al., 1999;

Lockwood et al., 2000; Saleh and Hossain, 2001; Linke and Berger, 2011), surfactant mixtures, surfactant/metal ion mixtures and metal ion mixtures found in waste water (Qu et al., 2008; Rujirawanich et al., 2012; Micheau et al., 2015). Surfactants such as cetyl trimethyl ammonium bromide (CTAB) have been used to boost and assist the recovery of algae, enzymes, proteins, textile dyes and metal ions (Walkowiak and Grieves, 1976; Lockwood et al., 1997; Gerken et al., 2006; Lu et al., 2010; Xu et al., 2010; Coward et al., 2014). Components such as enzymes and proteins generally have lower surface activity compared to surfactants. For example Xu et al. (2010) investigated the use of nonionic surfactant Tween 20 to boost the foaming of a mixture of Bovine serum albumin (BSA) with antifoam agent polyoxypropylene polyoxyethylene glycerin ether (PGE). BSA reduces the surface tension of water from 73 mN m^{-1} to 53 mN m^{-1} (Makievski et al., 1998) whilst Tween 20 reduces the surface tension of water to 35 mN m^{-1} (Niño and Patino, 1998). Therefore Tween 20 was used to increase the foam generation and stability of BSA in the foam fractionation process. More recently Liu et al. (2015) reported on the use of biosurfactant rhamnolipid as a foam stabiliser to assist in the foam fractionation separation of carotenoid lycopene from tomato-based processing wastewater.

Most of the aforementioned foam fractionation studies were performed in batch mode. The exceptions are Brown et al. (1999), performed in continuous simple mode, Gerken et al. (2006) and Qu et al. (2008), performed in continuous stripping mode and Rujirawanich et al. (2012), performed in a continuous multistage foam fractionation system. Brown et al. (1999), Gerken et al. (2006), Qu et al. (2008) and Rujirawanich et al. (2012) investigated the effect of process and design parameters such as air flow rate, feed flow rate, feed concentration and column height on the separation of multicomponent systems. These studies reported that the component with the highest surface activity was found to have greatest recovery and enrichment. These results are indicative of competitive adsorption processes, but this aspect was not explored in any further detail.

Studies considering the competitive adsorption of multicomponent systems in a foam fractionation process are relatively uncommon. Rujirawanich et al. (2012) applied the concept of competitive adsorption to its study of the separation of surfactant mixtures of cationic cetylpyridinium chloride (CPC) and nonionic Triton X–100 using a continuous multistage foam fractionation system. However the complexity of the multistage system limited the depth of insight into the fundamentals of the process. Conclusions generally empirically correlated the separation efficiency to operating parameters.

A previous theoretical study (Vitasari et al., 2013a) investigated the competitive adsorption of mixed surfactant–protein systems at an air–water surface. The simulation results illustrated how the competitive adsorption between the nonionic surfactant decyl dimethyl phosphine oxide (C_{10}DMPO) and protein Bovine beta lactoglobulin ($\beta\text{-LG}$) depended on the relative diffusivity and surface affinity of the two components. Initially the surfactant with the higher diffusivity and lower surface affinity was found to arrive at air bubble surface. The surfactant was then displaced from the surface once the protein arrived at a later time. The study concluded that surfactants with high diffusivity and low surface affinity relative to proteins were more likely to be displaced by high surface affinity proteins.

The objective of this paper is to perform a detailed experimental study on the application of foam fractionation to separate a model surfactant–protein mixture and elucidate the competitive adsorption process. The foam fractionation experiments were performed with an emphasis of gaining a deeper understanding of the competitive adsorption behaviour and transport processes of surfactant–protein mixtures in a foam fractionation column. The

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