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Free-standing thin film interactions with small particles



Georgina Zimbitas *, Peter J. Fryer, Zhibing Zhang, Serafim Bakalis

University of Birmingham, Birmingham, UK

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ABSTRACT

In order to investigate foam stability and coating ability via interactions with particles a free-standing film of initial thickness 150 µm to 350 µm was created within a metallic ring of 16 mm diameter. The weight change of the film is monitored and attributed to water loss with time. The effects of film drying are investigated with respect to the film's interaction with a solid, spherical, 3 mm diameter glass bead. This particle was dropped onto the film and high-speed camera imaging was used to examine the moment it interacts with the film. In addition to the water content of the film, the relative humidity of the environment, the initial film thickness of the film and the moment the bead interacts with the film are all factors that influence the quality of the film and hence how it interacts with the glass bead. The bead would cause defect points in the film to turn into tears. Films with a higher final water content presented with less tears and gave the particle a more homogeneous coating.

Industrial relevance: Thin films are often used in experimental and theoretical studies as a basis to investigate foam behavior (Bergeron & Radke, 1992; Myers, 1998; Stubenrauch & Von Klitzing, 2003; Weaire & Kermode, 1983). The importance of foam interaction with small solids spans a wide range of industrial and scientific fields such as agriculture, food science, pharmaceuticals, drilling fluids, household & personal care products (Dickinson, 2010; Eisner, Jeelani, Bernhard, & Windhab, 2007; Hunter, Pugh, Franks, & Jameson, 2008; Mason, Wilking, Meleson, Chang, & Graves, 2006; Murray, 2007; Quebaud, Sibai, & Henry, 1998). Liquid coatings are often used within the food industry, most notably for snack foods and animal feeds (Baldwin, Hagenmaier, & Bai, 2011; Beckett, 2008; Rockey, Plattner, & Souza, 2010). What is under investigation here is how a foam, represented via a liquid thin film, can be used to coat a small spherical particle. In addition the drying of the thin, surfactant-based films is taken under consideration with respect to the interaction of such films with solid particles.

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1. Introduction

Foams are considered as being gas bubble dispersions within a continuous matrix, the latter being either solid or liquid (Bikerman, 2013). Such systems are considered to comprise of gas bubbles separated by a series of interconnected thin, liquid films and as such thin films have been, from early on, extensively studied (Bikerman, 2013; Gibbs, 1928; Hooke, 1757; Myers, 1998; Mysels, Shinoda, & Frankel, 1959; Newton, 1704; Stubenrauch & Von Klitzing, 2003; Weaire & Kermode, 1983) often in order to get a better understanding of foam systems. Thin liquid film dynamics can give insight in understanding the foam structure and stability (Wasan, Nikolov, Lobo, Koczo, & Edwards, 1992), whereas the thickness of the film within a foam is critical to controlling foam destabilization (Stevenson, 2012). Overall, stability of thin films - and in extension foams - plays an important role in many technology processes, from the medical and biochemical field in the form of the preparation of nanostructures (Xu, Xia, & Lin, 2007), modification

* Corresponding author. *E-mail address:* G.Zimbitas@bham.ac.uk (G. Zimbitas). of DNA structures (Abramchuk, Khokhlov, Iwataki, Oana, & Yoshikawa, 2001), and medical diagnostics (Tarasevich, Isakova, Kondukhov, & Savitskaya, 2010), to more industrial processes such as in coatings and the food industry (Baldwin, Hagenmaier, & Bai, 2011; Beckett, 2008; Rockey, Plattner, & Souza, 2010; Zhang et al., 2012). Surfactants are often used in thin film technology due to their ability to lower the surface tension of various liquids and in general aid in film system minimization of undesirable effects associated with surface phenomena (Danov, Kralchevsky, & Ivanov, 1999). Additives such as salts are known to alter phases of the system (Blesic et al., 2009; Stewart, Saiani, Bayly, & Tiddy, 2011). Other surface active agents such as proteins or other small particles are often employed to aid in the stabilization of foam structures (Pickering, 1907). These macromolecules are found suspended in the film or partially embedded in the surfactant covered interface, acting to modify the interface properties. Polymer additives, in particular, have long been known to have an effect on the dynamic and thermodynamic stability of surfactant-based liquid films (Grant, 1966), and thus also on the mechanical properties of the films near interfaces and within thin films (Jabbari & Peppas, 1995; Lo & Narasimhan, 2005; Schnell, Stamm, & Creton, 1998). Studies of films

presenting similar surface energies but different internal structures have recently been gaining interest, especially in the form of studies of diblock copolymers (Bermdez, Aranda-Espinoza, Hammer, & Discher, 2003; Discher et al., 1999; Fredrickson & Bates, 1996). The mixing of polymer and surfactant solution leads to different forms of segregation (Bergfeldt & Piculell, 1996; Goddard, 2016; Lindman, Antunes, Aidarova, Miguel, & Nylander, 2014; Piculell, Bergfeldt, & Gerdes, 1996; Thalberg & Lindman, 1993; Thalberg, Lindman, & Karlström, 1990), depending on the chain length, size and charge of the molecules present. Oppositely charged surfactant-polymer mixes lead to what is referred to as associative phase separation, whereby a water phase depleted of macromolecules is in equilibrium with a concentrated surfactant-polymer phase (Goddard, 2016; Thalberg et al., 1990). In the case of similarly charged systems the phase separation that occurs is referred to as segregative (Bergfeldt & Piculell, 1996; Nilsson, Blokhus, Hellebust, & Glomm, 2002; Piculell & Lindman, 1992; Thalberg & Lindman, 1993), leading to phases enriched with their respective macromolecule, polymer or surfactant micelles. Kuzma, Wedler, Saupe, Shin, and Kumar (1992) used microscopy and calorimetry to show how the addition of polyethylene oxide proved to be a disruptive influence to the lamellar order of the various surfactant-based systems. X-ray scattering was used by Ligoure, Bouglet, and Porte (1993) in order to see the phase separation of lyotropic phases occurring due to polymer presence. Surface active polymers influence the rheological properties of films altering the rate of draining (Bruinsma, Di Meglio, Quere, & Cohen-Addad, 1992; Lionti-Addad & Di Meglio, 1992). Bergeron, Hanssen, and Shoghl (1997) used foam-film disjoining pressure and surface tension isotherms in order to observe how the addition of certain polymers to liquids containing charged surfactants lead to the formation of strong polymer-surfactant interactions, which in turn created an enhanced adsorption of both materials to the airliquid interface. In general, the addition of soluble polymers to liquids result in an increase in the bulk viscosity which in turn can significantly slow down the drainage of the thin-liquid films and foams. Critical micelle concentration (cmc) plays an important role in how these macromolecules self-assemble within the liquid structure. Surfactant molecules and amphiphilic polymers in aqueous solutions at concentrations above the cmc assemble into micellar structures, having the hydrophilic part of the surfactant in contact with the water while simultaneously shielding the hydrophobic part within the inner part of the structure. Further increase in the cmc concentration results in the formation of periodic hexagonal, cubic or lamellar mesophases (Broze, 1999). Studies by Krichevsky and Stavans (1993, 1994) revealed a correlation between the critical micelle concentration and the thinning of surfactant-based films, whereby once above the cmc non-ionic polymers can interact with the surfactant aggregates present in the bulk and thus influence the driving influence for thinning by modifying, for example, the depletion forces.

Drying of polymer films has been under investigation for many years (Okazaki, Shioda, Masuda, & Toei, 1974). Solvent casting involves the drying of polymer solutions and is used in the manufacture of many devices, from (micro)electronics (de Gans, Duineveld, & Schubert, 2004; Minemawari et al., 2011) to biomaterials (Dugas, Broutin, & Souteyrand, 2005). Ordered mesoporous materials are often manufactured by solvent evaporation of surfactant-polymer mixes (Bruinsma, Kim, Liu, & Baskaran, 1997; Lu et al., 1997) at temperatures well above room temperature. Despite the extensive research into drying of polymer-surfactant mixes the drying process near the air-liquid interface of films is, however, still not well understood. The effect of relative humidity is even less understood with regards to how it influences the drying of surfactant and surfactant-polymer films (Alexandridis, Munshi, & Gu, 2010), with the majority of the studies focusing on polymer solutions and plasticization effects (Fernández-Pan, Ziani, Pedroza-Islas, & Maté, 2010; Kayserilioğlu, Bakir, Yilmaz, & Akkaş, 2003; Tapia-Blácido, Sobral, & Menegalli, 2005).

Interactions of films with particles of different sizes are of significance in many scientific and industrial fields (Dickinson, 2010; Eisner, Jeelani, Bernhard, & Windhab, 2007; Hunter, Pugh, Franks, & Jameson, 2008; Mason, Wilking, Meleson, Chang, & Graves, 2006; Murray, 2007; Quebaud, Sibai, & Henry, 1998). Interaction of thin films with small, colloidal particles of 1–1000 nm diameter is of interest in areas ranging from biology (Kralchevsky & Nagayama, 2000) to the food industry (Eric, 1992; McClements, 2004; Spyropoulos et al., 2011) due to particle presence being of significance in, for example, modifying the stability of foam and emulsion systems (Bergeron & Radke, 1992). Under certain conditions small particles within the film can come together forming larger agglomerates that ultimately destabilize the film (Li, Yin, Yang, Tang, & Wei, 2012). It is common in the pharmaceutical industry to use thin films to coat digestible pills. In this work thin films were created from a high content polymersurfactant mix and the drying of the films at room temperature was investigated and taken under consideration in how it affects the stability of the film. The initial thickness of the film (app. 100 μ m-350 μ m) and the relative humidity (app. 30%-60%) of the surrounding atmosphere were monitored and shown to influence the evaporation kinetics of water from the film for the first 10-15 min after the film formation. A small, solid particle was used mainly as a means to measure the stability of these films.

2. Materials & methods

Copolymer solution w/w 40% active and NaLAS w/w 45% active solution (referred here to as NaLAS paste) were provided by Procter & Gamble, Newcastle UK. The former is a transparent, yellowish liquid copolymer mix of acrylic acid and maleic acid, whereas the latter is a white paste consisting of Sodium Linear Alkylbenzene Sulphonate (NaLAS) with alkyl chain lengths varying from C10 to C16. A 70.4:29.6 copolymer to NaLAS w/w ratio mix was prepared. A Krüss Processor K 100 Tensiometer was used to acquire density and surface tension measurements for the starting materials and the mix, the results of which are tabulated in Table 1. Specifically for 100 g of sample 1, whereby the NaLAS paste is 45% active and the copolymer solution 40%, the dissolved dry mass (referred to in this work as the solid content) is of (29.6 * 0.45 + 70.4 * 0.4 =) 41.5 g and the water content would thus be (100-41.5) = 58.5 g.

A schematic of the setup is presented in Fig. 1. A metal ring of 16 mm inner diameter was manufactured from silver wire of 0.5 mm diameter. This ring was attached to a holder which was set on a Precisa XB 220 A precision balance in order to monitor the weight change of the films during the experiments conducted. The liquid mix was placed in a small petri dish under the ring. The petri dish was either lifted by hand into the ring, or a Laser 2000 Zaber 25 mm linear travel actuator (resolution of 0.05 µm) was used to move the liquid petri dish perpendicular to the ring allowing for it to be submerged and then retracted from the liquid at a steady speed ranging from 0.001 mm/s to 0.1 mm/s, thus facilitating the reproducibility of the thickness/weight of the thin films formed. The petri dish was removed from the chamber immediately after film formation. A plastic tube of 21 cm height and 0.5 cm inner diameter was placed approximately 2-5 cm above the ring. This tube was used as a channel by which the particle was dropped through, allowing for the particle to land in the center of the metallic

Table 1
Density and Surface Tension measurements for materials used.

Copolymer solution: NaLAS paste w/w ratio measured	Density (g/mL)	Mean Surface Tension (mN/m)
70.4: 29.6 (Sample 1) Copolymer solution NaLAS paste	$\begin{array}{c} 1.459 \pm 0.006 \\ 1.322 \pm 0.005 \\ 1.451 \pm 0.1 \end{array}$	$\begin{array}{c} 19.38 \pm 0.7 \\ 25.37 \pm 0.1 \\ 23.76 \pm 0.6 \end{array}$

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