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GRAPHICAL ABSTRACT

# Evaporating foam films of pure liquid stabilized via the thermal Marangoni effect

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## HIGHLIGHTS

- Evaporation stabilizes foam films made of pure liquids.
- Evaporation induces a temperature difference between the film and the meniscus.
- The temperature gradient induces a stabilizing thermal Marangoni flow.
- The Marangoni flow sustains a stationary thickness of the foam film.
- As the meniscus cools down, the film thins until its rupture at a critical thickness.

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## ABSTRACT

A foam film made of pure liquid can be stabilized by evaporation. This is demonstrated experimentally for water and alkane films formed in a Scheludko cell at controlled saturation of the ambient air. A mechanism of the stabilization is proposed: evaporation leads to a local decrease of the temperature in the centre of the film; the meniscus acts as a thermostat and maintains a higher temperature at the film periphery. The resulting temperature gradient brings about a surface tension gradient causing a stabilizing thermal Marangoni flow that carries fluid from the meniscus to the interior of the film. The film thickness is quasi-stationary and gradually decreases as the meniscus cools due to the evaporation. At a certain critical meniscus temperature, the film reaches a critical thickness at which the Marangoni effect can no longer counteract the combined action of the capillary pressure and the van der Waals attraction, and the film breaks. The lifetime of the film is estimated as a function of the film size and the experimental conditions (temperature, saturation, vapour pressure, capillary pressure). The theoretical and the experimental results for the lifetime and the critical thickness are in qualitative agreement for films at moderate saturation.

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## 1. Introduction

The drainage and stability of thin liquid films determine the properties of a broad range of objects, including foam, bakery yeast

\* Corresponding author. E-mail address: ris26@cam.ac.uk (R.I. Slavchov). (Exerowa and Kruglyakov, 1998), cement (Du and Folliard, 2005), water-crude oil emulsions (Sullivan and Kilpatrick, 2002), and many others. The process of evaporation is known to influence the stability of these films. In particular, there are numerous studies on evaporating wetting films on solid substrates due to their importance for certain technological processes, such as fibre optic coating, semiconductor chip deposition (Danov et al., 1998), and





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fuel atomization in engines (Baumgarten, 2006). Evaporation interferes with these processes primarily by causing Marangoni flows (Baumgarten, 2006; Danov et al., 1998; Sultan et al., 2005; Yiantsios and Higgins, 2006; Zuiderweg and Harmens, 1958). In contrast, the research on evaporating *foam films* is quite limited (Manev and Nguyen, 2005) and to our knowledge, only a handful of studies devote attention to evaporating thin films containing no surfactant, e.g. (Yaminsky et al., 2010a). The related problem for evaporation of sessile drops from pure liquids has been studied in greater depth (Chandramohan et al., 2016; Hu and Larson, 2006, 2005).

Evaporation has a twofold effect on the stability of thin films by taking away heat and mass from the film, it brings about thermal Marangoni flows (driven by a temperature gradient) and Gibbs-Marangoni flows (driven by concentration gradients in multicomponent liquids). In the case of a wetting film, the solid substrate acts as a thermostat and reduces the magnitude of the thermal Marangoni effect. Due to the absence of this thermostating effect, it can be expected that the Marangoni flow is stronger in evaporating foam films. There are hints in the literature in support of this hypothesis. For example, Li et al. (2012) reported that bubbles formed from surfactant solutions burst under non-uniform evaporation. In their experiment, evaporation leads to a local thinning of the liquid films, as well as to a local increase of the surfactant concentration, resulting in a gradient of the surface tension  $\sigma$ . This gradient causes a Marangoni flux away from the thinner region of more intense evaporation and leads to the bursting of the bubble. In contrast, Manev et al. studied evaporating foam films formed from surfactant solutions in a Scheludko cell and found no significant effect of evaporation in these systems (Manev, 1975; Manev and Nguyen, 2005). Yaminsky et al. hypothesized that evaporation-driven Marangoni effects were the cause of the stabilization of the foam films they observed in a variety of experimental setups. Their surfactant-free films reacted particularly strongly to evaporation (Yaminsky 2006; Yaminsky et al. 2010a, 2010b). Yaminsky (2006) reported that when a small air bubble attaches to the interface between air and a pure liquid containing no surfactant, its behaviour is dependent upon the saturation - if the air is saturated with vapour, the bubble coalesces immediately. Conversely, in an unsaturated environment, the air liquid|air film formed upon contact between the bubble and the surface remains stable over an indefinite period of time. Another interesting phenomenon occurring in evaporating films free of surfactant was reported by Karakashev et al. (2008). Contrary to expectations based on the Derjaguin-Landau-Verwey-Overbeek theory, their foam films exhibited significant stability (lifetime  $\sim 100$  s) and a complex dynamic behaviour despite being formed from concentrated solutions of inorganic electrolytes. This anomalous stability was only displayed when the surrounding gas was undersaturated with respect to the vapour pressure of the salt solution (Karakashev et al., 2008).

These reports suggest that evaporation plays a key role in the stabilization of surfactant-free foam films against the capillary pressure-driven drainage and generated our interest in exploring this topic. The salt films studied by Karakashev et al. (2008) are complex systems - the observed intense fluxes must be affected by the evaporation-induced gradient of the electrolyte concentration and the effect of the electrolyte on surface tension (Slavchov Novev, 2012). Evidently, evaporation-driven Gibbsand Marangoni flows are also present in surfactant solutions; for this reason, trace amounts of surfactant have a destabilizing effect on evaporating films of pure liquid (Yaminsky et al., 2010a). Instead of tackling the complicated problem of two-component films, in this study we explore the simpler case of an evaporating film comprised of pure liquid, where only the thermal Marangoni effect is present.

Even in this case, however, the behaviour of the film is influenced by many factors other than the relative saturation, complicating the interpretation of the experimental data. There is no consensus in the literature about the stability of pure water foam films even in the absence of evaporation (Peng and Chang, 2014): while some authors state that they rupture instantly and even use this as a criterion for the purity of their experimental setup (Karakashev et al., 2008; Nguyen and Nguyen, 2010), others report stable films of this kind (Exerowa, 1969; Exerowa and Kruglyakov, 1998; Yaminsky et al., 2010a). The reported lifetime of such films varies by orders of magnitude depending on the method used for their preparation. Karakashev and Firouzi (Karakashev and Firouzi, 2014) observed stable water films; however, they became unstable following a 200-fold flushing of the entire cell with water. In a saturated environment, after the flushing, pure water films were found to have a lifetime of  $\sim$ 5 s, reaching a thickness of  $\sim$ 30 nm before rupture. In an undersaturated environment, such films ruptured instantly.

Several studies report that the stability of pure water films is also dependent upon the approach velocity of the two water|air interfaces (Del Castillo et al., 2011; Firouzi and Nguyen, 2014; Wang and Qu, 2012; Yaminsky, 2006; Yaminsky et al., 2010a, 2010b). In a saturated environment, if the velocity is above a certain critical value (35 µm/s (Firouzi and Nguyen, 2014), 200 µm/s (Yaminsky et al., 2010b) or less than  $11 \,\mu$ m/s (Wang and Qu, 2012)), the two surfaces coalesce instantly. Firouzi and Nguyen (Firouzi and Nguyen, 2014) found that films live for several seconds at an approach velocity of under 35 µm/s. Yaminsky et al. reported a similar finding at both saturated (Yaminsky et al., 2010b) and undersaturated (Yaminsky et al., 2010a) conditions: for approach velocities between  $\sim 1$  and  $\sim 100 \,\mu m/s$ , the average lifetime of their films was about a minute, while below 1 µm/s, it was of the order of hours. Numerous studies devoted to the coalescence of gas bubbles with the water|air interface show a similar dependence on the approach velocity (Del Castillo et al., 2011; Katsir and Marmur, 2014a, 2014b). The stability at low approach rates is attributed to electrostatic repulsion between the two interfaces (Katsir and Marmur, 2014b; Yaminsky et al., 2010b). It is not entirely clear what the cause for this electrostatic repulsion is or by what mechanism it is overcome at high approach rates. Finally, a persistent reason for contradicting reports is the presence of surface-active impurities. Such impurities generally stabilize films under saturated conditions; however, evaporating films made of pure liquid can in fact be destabilized by trace quantities of surfactants (Yaminsky et al., 2010a).

The short review above demonstrates that, though it may not be entirely consistent, there is a body of evidence indicating that *films* formed from single-component liquids may be stabilized by evaporation. Our aim in this study is to clarify the question by (i) considering the possible mechanism of stabilization theoretically and (ii) investigating evaporating films experimentally under controlled saturation of the ambient air. The basic hypothesis we will test is that evaporation gives rise to a thermal Marangoni effect - an idea formulated by Yaminsky et al. (2010a). The model we will present in Section 2 generalizes the classical Maxwell-Langmuir-Fuchs theory (Fuchs, 1959) of the evaporation of a spherical droplet to an approximate geometry mimicking the biconcave droplet in the Scheludko cell (Sheludko, 1967); it can also be viewed as a thermal-Marangoni-effect analogue of Marrucci's theory of the stabilization of films by the Gibbs-Marangoni effect (Marrucci, 1969). We use this model to analyse the behaviour of several measurable quantities with respect to changes in the experimentally controllable parameters - particularly interesting are: the evolution of the film thickness; the film lifetime as a function of its size and the ambient vapour pressure; the critical parameters of film rupture - film thickness, radius, saturation - as a function of the Download English Version:

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