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Published in:
Current Opinion in Colloid and Interface Science

DOI:
10.1016/j.cocis.2021.101440

Publication date:
2021

Citation for published version (APA):
Cox, S., & Davies, T. (2021). The evolution of numerical methods for predicting the distribution of surfactant in the bubble-scale dynamics of foams. *Current Opinion in Colloid and Interface Science*, *53*, [101440]. https://doi.org/10.1016/j.cocis.2021.101440

The evolution of numerical methods for predicting the distribution of surfactant in the bubble-scale dynamics of foams

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Abstract

Many numerical methods now exist to simulate the structure and dynamics of surface-tension dominated aqueous foams at the level of the individual films and the liquid structures where they meet. We review these methods, focusing in particular on bubble-scale simulations of foam rheology. We highlight methods that allow the distribution of surfactant during flow to be taken into account.

Keywords: foam rheology, numerical simulation

1. Introduction

Liquid foams, and their closely-related cousins highly-concentrated emulsions, constitute not only much of the food that we eat but find use in many of the industrial processes on which our society relies, such as enhanced oil recovery and metallic ore separation [1, 2]. Since Plateau's experiments on the way in which the interfaces of a foam fit together [3] and Kelvin's proposal that the aether consists of a foam-like packing [4], both working in the late 19th century, scientists from a broad array of disciplines have sought to describe and explain the properties of foams and emulsions.

The microstructure of a foam in itself is rich in geometry, with links to the honeycomb conjecture as well as Kelvin's partition of three-dimensional space. Of more interest in applications is the effect of gravity, which changes the local foam density via the process of drainage (or creaming) [1], with the denser liquid phase collecting towards the bottom of the material. Nonetheless this does not destroy the foam: it is when films thin that bubbles burst and the foam finally collapses.

In the 1980s, Princen used a model of a hexagonal ("ordered") foam [5] to predict some of the properties of a sheared foam, particularly the elastic shear modulus and yield stress. This triggered an extensive body of work over the last forty years on foam rheology [6] which shows no signs of stopping. Much of it is concerned with the development of continuum models of foam flow, using and extending the Herschel-Bulkley constitutive model [7, 8, 9] for a viscoplastic (yield stress) fluid to include elasticity.

Here we focus on the bubble scale, where the emphasis is on combining existing models, for example of foam rheology and gravity-driven liquid drainage [10] or including Marangoni surfactant redistribution with rheology [11]. This is a response to the recognition that while a model such as Herschel-Bulkley can be fitted to experimental data with high fidelity, the dependence of the fitting parameters (such as the power law exponent) on the material properties (bubble size and polydispersity, for example) is obscure.

Following in Princen's footsteps, we therefore survey existing methods for simulating foam structure and dynamics, emphasising recent developments in combining film-level redistribution of surfactant during flow of disordered foams.

2. Static Structure

In an aqueous foam in which the bubbles are at rest, liquid drains down through the foam under gravity to collect at its base. Over time, the top of the foam dries out, leaving what is known as a dry foam with vanishingly small liquid fraction ϕ_l. The foam structure in this limit consists of gas contained within thin curved films, a simple approximation that is attractive for theory. Further simplification is found in two-dimensional (2D) models of such dry foams as a collection of curved lines separating planar bubbles; these are approximately realizable in practice by trapping a foam between two closely-spaced parallel glass plates [2] (figure 1). 2D models, particularly of dry foams, have enjoyed great success in elucidating the structural (and dynamical) properties of foams, but foams in real applications are rarely dry (or 2D, but past experience suggests that there is much that can still be learned from the 2D case), and so it is important to consider "wet" foams in which the regions where films meet, known as Plateau borders, contain liquid.

The Young-Laplace law states that in a foam at equilibrium each interface balances the pressure difference $p_\pm$ on different sides by adjusting its mean

curvature C. The coefficient of proportionality is the interfacial tension γ:

$$p_{\pm} = \gamma C. \tag{1}$$

This law applies both to an interface that separates the liquid in the Plateau borders from a bubble and to an interface between two bubbles; in the latter case this is in practice treated as a double interface (gas – liquid – gas) and so the effective interfacial tension is doubled to 2γ. At equilibrium we also expect the interfacial tension to be constant, noting that this is the result not only of a cessation in any changes of area of each interface, but also of the movement of surfactant along and between interfaces in response to gradients in surfactant concentration. Then since the pressure in each bubble is constant on each side of an interface, the mean curvature of each interface is constant.

In a 2D foam this means that interfaces are arcs of circles. In a dry 3D foam at equilibrium, Plateau's rules [3] dictate the local structure: interfaces meet in threes along Plateau borders and Plateau borders meet in fours. The interfacial tensions then dictate the angles between interfaces and between Plateau borders. Any imbalance in the tensions between Plateau border surfaces and thin film surfaces may lead to a finite contact angle where the two meet; this may trigger flocculation of the structure, in which the liquid is no longer uniformly distributed through the foam or emulsion [12].

Numerical techniques for predicting foam structure often rely either on satisfying Plateau's laws [13, 14] or, more usually, minimizing the free energy of the foam (which is equivalent to Plateau's laws [15], but computationally more straightforward). High precision can be achieved with Brakke's Surface Evolver [16], particularly for small foams (less than about 1000 bubbles) with low liquid fraction, providing information about quantities including film curvature and bubble pressure [17] and, for example, the actual 3D liquid structure in a quasi-2D foam [18] (figure 1) in a deterministic way. With many bubbles, this level of precision means that the simulation is slow, while at high liquid fraction any significant departure from the proposed initial foam structure during relaxation may require changes in the topology of the liquid network, which are difficult to perform. In these cases the Potts model [19] provides similar (but less accurate) information about the structure, again using minimization of energy, in a stochastic manner, and hence offers an easier route to simulating many realisations of a disordered foam.

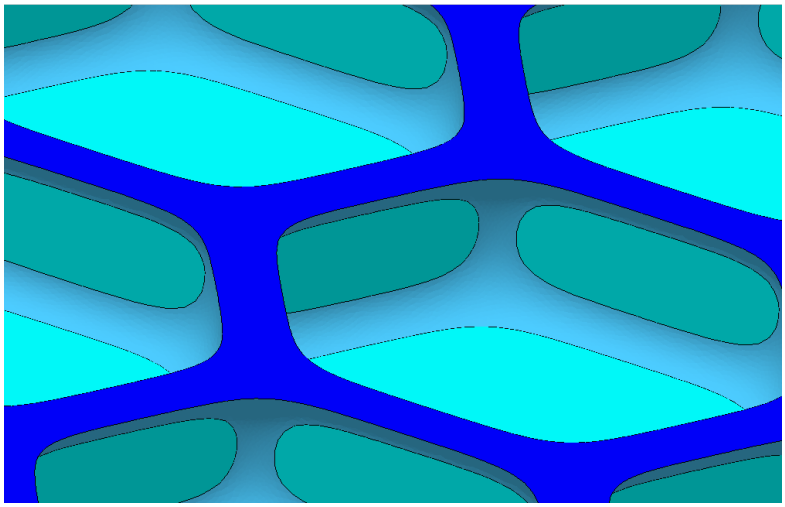

Figure 1: The liquid distribution in a layer of hexagonal bubbles squeezed between parallel plates. The vertical interfaces between bubbles are shown in cyan, the Plateau border interfaces in light blue, and the regions where the liquid touches the upper plate in dark blue.

Recently, a "Morse-Witten" theory [20] for the way in which compressed droplets (or bubbles in this case) deform has been used to generate equilibrium structures of monodisperse [21, 22] and polydisperse foams in which bubble deformation is small [23], up to 10% of the bubble diameter. It remains to be seen whether this method can provide a route to predicting foam flow.

3. Foam flow with constant interfacial tension

Both the Surface Evolver and the Potts model (distributed as Compucell) provide information about *slow* foam flows by assuming that the structure never deviates from an equilibrium structure while changing the boundary conditions (for example simple shear). Such quasi-static simulations, in which the interfacial tension is generally considered constant, are appropriate for predicting the evolution of foam structure when the mechanical relaxation of this structure is much faster than, for example, the shear-rate, the rate of gas diffusion between bubbles (coarsening, or Ostwald ripening) and the rate of surfactant redistribution.

At finite strain-rate there are other possible numerical methods. For dry foams, the *vertex model* assumes planar films and viscous dissipation at the

vertices [24, 25], representing the effect of the liquid that would be present there in a wetter foam. The simpler polygonal structure makes the numerics faster, allowing more than 1000 bubbles to be simulated, and a simulation proceeds by seeking to minimize the dissipation rate as well as a free energy similar to the Surface Evolver one.

Also for dry foams, but for fast-flowing ones, the 2D Viscous Froth Model (VFM) [26] includes friction along the length of each film to supplement the Young-Laplace balance between curvatures and pressures. This is to represent the external dissipation that occurs when a soap film moves between parallel plates, as in a typical 2D experiment in a Hele-Shaw cell. The model has successfully explained the motion of bubbles in microfluidic devices [27], for which the 2D approximation is often appropriate, and recent work [28] indicates how the VFM might be extended to wet foams.

Inspired by the other extreme of liquid fraction, Durian's bubble model [29] approximates a wet foam as a collection of spheres [30]. Now referred to more generally as soft-disk models [31], the idea is that the overlap of two neighbouring spheres represents the elastic repulsion between them, while any lateral motion is opposed by a linear friction force. Adaptations of Durian's model also allow for short-range interactions between non-overlapping disks [32, 33], which models an attractive contribution to the disjoining pressure and a shear flow in the thin interfaces. Although the soft-disk models have been criticised [34] as unrealistic since they do not include bubble deformation, their computational simplicity and ability to effectively capture important aspects of the rheological behaviour of a wet foam make them valuable [31, 33].

Other simulation techniques that are used for wet foams include two-phase Lattice Boltzmann methods [35, 36], which are not well suited to resolving thin films, a "Soft dynamics" model [37] in which the bubbles are deformable spheres, and Saye and Sethian's level set approach [10], which includes structural relaxation and flow, drainage, coarsening, and film rupture, but as with the two preceding methods, has not been widely taken up by the community, perhaps due to its complexity.

Finally in this section we describe recent work [38] that seeks to include a disjoining pressure in a model of a wet 2D foam. In the sense that this "DySMaL" method [38] treats the motion of each point of the circumference of each (deformable) bubble using a balance of forces on a discretised line, it is related to the VFM. The differences are that each side of a film is treated independently, so the liquid fraction is an additional parameter, and the two sides of the film are held apart by a disjoining pressure (see figure 2) which

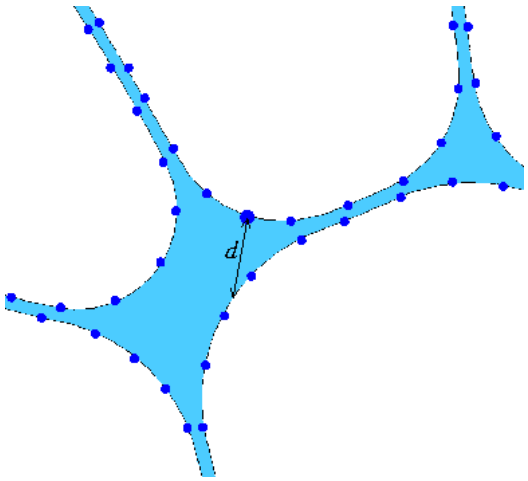

Figure 2: The DySMaL method for simulating the dynamics of a 2D foam uses discretised straight interfaces (meeting at the vertices shown as dots). The force on a vertex on one interface due to the disjoining pressure in the (shaded) liquid region between the bubbles is determined by the distance d to the nearest interface in a perpendicular direction. Any stretching of the interfaces can be related to local changes in interfacial tension via eq. (2).

could be taken to be proportional to e^{-d}, where d is the distance between the interfaces, as suggested by DLVO theory [39]. Moreover, the source of dissipation is chosen to be due to the liquid flow around the bubbles, although in a 2D setting it is natural and straightforward to replace this with a VFM-like dissipation from external friction, as in our own work (see figure 3). We find that simulations of the flow of around 100 bubbles can take days to weeks of computer time. So while the model could be extended to 3D in a natural way, the computations are likely to be very slow and would require highly-parallel computations to provide information about the flow of a bulk foam.

4. Accounting for surfactant motion

The variety of numerical methods that now exist to simulate foam flow in the presence of various sources of dissipation, in both 2D and 3D and for a range of liquid fractions, is evidence of remarkable progress in the field. Examples include the differential flow of bubbles of different sizes [25] and the prediction of bubble deformation in foam flow through convoluted channels [27] and past obstacles [40].

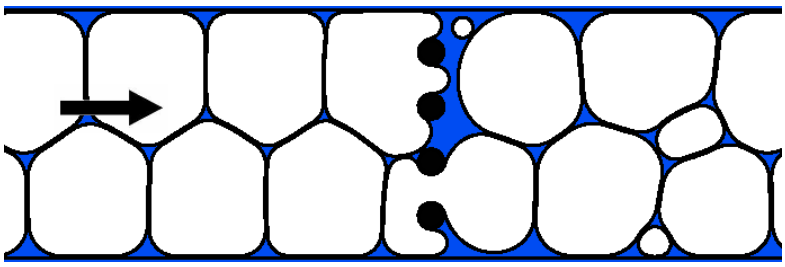

Figure 3: In our version of the DySMaL method the dissipation is due to friction with the upper and lower plates that contain the foam. In this example a staircase structure flows from left to right past an array of four discs, which leads to stretching and break-up of the bubbles to create a finer foam.

However, there are still shortcomings, for example that many methods require several days, weeks or even months of computer time and therefore rely on rather small foam samples of not much more than 1000 bubbles. More importantly, the methods described above all assume that the surface tension γ in all the films of a given foam is a fixed constant γ_0.

The local surface tension of a film is determined by the concentration Γ of surfactant molecules there; hence sufficiently rapid flow of the foam may induce currents within films or stretching of films, both of which are likely to lead to changes in the distribution of surfactant.

A first-order correction to the surface tension of a film (or part of a film) is given by the Langmuir expression

$$\gamma = \gamma_0 - G \ln \left(\frac{\Gamma}{\Gamma_0} \right), \tag{2}$$

where Γ_0 is the equilibrium surfactant concentration and G is the Gibbs elasticity of the film. Simply put, as a film is stretched, the concentration of surfactant within the film decreases below the equilibrium value and so the surface tension rises, cf. figure 2. (If the rise is extreme this can lead to rupture.)

Any non-uniformity in surfactant concentration will be compensated by a flow of surfactant molecules that seeks to re-equilibrate the distribution, known as the Marangoni effect [2]. This may occur within each film, or over a wider region of the foam if flow across the Plateau borders where the films meet is possible. To this can be added further effects, for example due to adsorption of surfactant into the film [41], the effects of surface viscosity

in resisting flow at the interface [42], and surfactant solubility, making the determination of the relevant dynamics remarkably complex [2].

For a single film, Vitasari *et al.* [43] studied the balance between the drainage of liquid from within the film (towards the Plateau borders) with a surface flux of surfactant towards the depleted region at the centre, extending an already extensive body of work on foam films, the Marangoni effect and marginal regeneration [44, 45]. Critical to predictions of the importance of Marangoni effects is the mobility of the interface: in this case, the dynamics of a soap film with a *rigid* interface (in, for example, a foam stabilized by proteins) is dominated by the surfactant flow and the drainage of liquid from within the film is largely suppressed. As with simulations of several films using the VFM [11], diffusion of surfactant from the film to the interface is neglected.

Since in the DySMaL model [38] each bubble consists of a single closed interface (figure 3), it is straightforward to implement a simple surfactant balance in the model, since movement of surfactant molecules at vertices is accomplished naturally. Moreover, as the local distance between interfaces is part of the calculation, diffusion of surfactant molecules from one interface to another could be included.

For foams, not only is it of interest to consider a collection of films and to ask how each film is affected by the re-distribution of surfactant during flow, and the extent to which there is transfer of surfactant between films, but in particular we seek to determine the effects of topological changes (T1s) in the foam structure. The paradigmatic geometry for this problem is a system of five films connecting four pins (figure 4), in which the pins can be moved to trigger a topological change in which one of the films shrinks to zero length and reforms in a new orientation [42, 46, 47, 48].

The five film geometry has been analysed as a 2D problem in which every film remains straight and in which the surfactant molecules cannot cross vertices [42, 46]. This work was later extended to curved films with transfer across vertices using the VFM [47]. Similarly, Titta *et al.* [48] have considered a T1 in a wet foam using a level-set method. In their comprehensive calculations they also include many more facets of the relevant surfactant dynamics.

What is lacking from analyses of the five film geometry is any discussion of the effect of bubble pressures. An extension of the VFM attempts to rectify this by considering a single 2D bubble being pushed along channels of various geometries [11]. What this shows is that redistribution of surfactant is critical

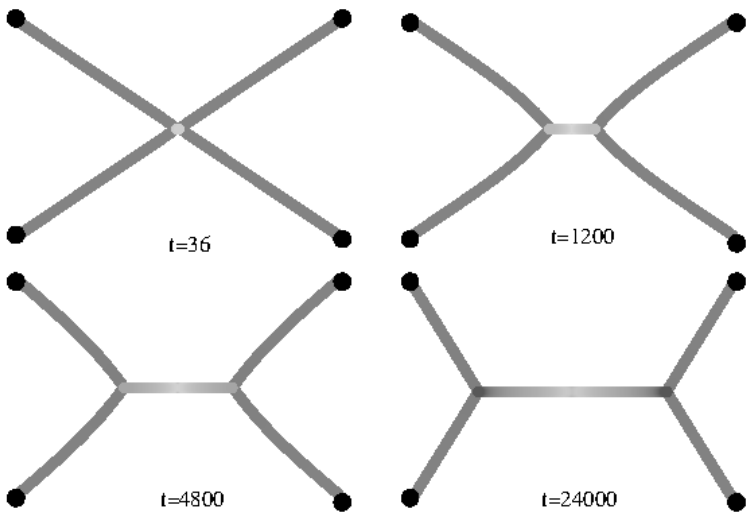

Figure 4: A 2D system of soap films between four fixed pins at four different times (in units of λ/γ, where λ is the drag coefficient). The initial configuration is an unstable one with four films meeting at a point. A short film is formed at the centre of the system with low surfactant concentration (lighter shading) compared to the four "arms". In this simulation with the Viscous Froth Model [47], the drag with the bounding plates induces curvature in the arms and there is a flow of surfactant from the arms to the central film which balances the concentration throughout the system when it reaches equilibrium, some time after the last image shown. Films are thickened for clarity.

to the stability of the foam structure: above a certain velocity, disparities in the surface tensions of different films (or more precisely, different parts of the different films) lead to a situation where the flow of the foam along the channel cannot be sustained. Then bubbles are left behind and the foam structure is gradually lost.

Bubble-scale simulations of bulk foam flow that incorporate the local effects of surfactant flow are rather rare. We highlight Cantat's work [41] which uses the vertex model (so the films are straight) to simulate 500 bubbles at a range of shear rates, with a supplementary time-scale associated with the adsorption of surfactant at the surface of each film and a linearised version of eq. (2)) to determine variations in surface tension due to surfactant flow:

$$\gamma = \gamma_0 - G\left(\frac{\Gamma - \Gamma_0}{\Gamma_0}\right), \tag{3}$$

appropriate for small variations in concentration. (This model could be taken to include some effects of surfactant diffusion, provided they are not dominant in determining surfactant flow.) There is no surfactant transfer across vertices and the surface tension is constant on each film. This work shows how the foam stress increases with strain-rate due to the effects of film stretching on film tension, albeit without the inclusion of film curvatures.

5. Outlook

There is a variety of methods now available for predicting the effects of surfactant concentration on foam dynamics. Each has advantages and disadvantages, and the choice of method will depend on factors such as the level of geometric detail required (for example the extent to which interface curvature drives the dynamics), the chemistry of the surfactants (is the interface rigid or mobile?) and of course the available computing power.

Our own work now concerns the scaling up of VFM calculations which incorporate curved films and surfactant transfer throughout the foam [49] to predict the complex moduli of bulk foams in the dry limit. There is also much scope to extend vertex model calculations [41] with additional film-level surfactant dynamics. For wet foams, the DySMaL model [38] appears a promising base on which to build.

To the best of our knowledge, there has been no research on how surfactant transfer might be included in a soft disk model; this is surprising given

its popularity. While this would necessarily neglect some aspects of the interface geometry, such a large-scale calculation is likely to lead to further insight.

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