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Variation of average coordination number with liquid fraction for two-dimensional foams with finite contact angle

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Abstract

We present Surface Evolver computer simulations for two-dimensional (2d) foams in which the contact angle at which liquid films and Plateau borders meet is non-zero. We determine the average coordination number Z of bubbles over a wide range of finite contact angles θ and liquid fractions ϕ. We find a linear scaling of Z for values of ϕ between about 0.07 and 0.15, corroborating previous data obtained for the case of zero contact angle. The results highlight the importance of attractive bubble interactions for 2d foams.

1 Introduction

Aqueous foam, a two-phase fluid consisting of gas bubbles surrounded by liquid, features both in our daily lives (e.g. shaving foam, carbonated drinks) and in various fields of engineering, such as oil-gas field development [1], foam flotation [2], and soil remediation [3]. An important quantity for the description of foam structure is its liquid volume fraction ϕ,

which is generally a function of both position and time. For values of ϕ around 30%, a three-dimensional (3d) wet foam resembles a dense packing of near-spherical bubbles. In dry foams, where the liquid fraction is less than about 3%, the bubbles take on near-polyhedral shapes [4].

This variation in character of the bubble packing, together with the fact that foams are generally disordered with a wide variation of bubble sizes (polydispersity), makes simulating them a difficult task. The Surface Evolver software developed by Ken Brakke proved an essential tool for this [5], but 3d simulations of foams over a wide range of liquid fraction are still very limited. However, it has long been recognized that the study of two-dimensional (2d) foams, which is computationally much less demanding, offers insight into many statistical properties of foam structure, such as the variation of the average number of contacts of a bubble (coordination number) as a function of liquid fraction. Such a 2d foam is not just a toy for a theorist, but may be realized experimentally, for example by placing a 3d foam between two glass plates with a spacing that is much smaller than the average bubble diameter [6, 7].

In the standard model of a dry 2d foam ($\phi = 0$), the infinitesimally thin liquid films separating two bubbles are represented by arcs of circles, as a consequence of the Laplace-Young law, and three such arcs meet at a vertex at 120 degrees (Plateau's law) [8, 9]. For a small non-zero value of liquid fraction the vertices swell and form what are called Plateau borders, which contain almost all the liquid. These three-sided Plateau borders grow in size with increasing liquid fraction, and eventually merge to become four- or more-sided. The 2d equivalent of a wet 3d foam is a random packing of circular bubbles (disks) at a critical liquid fraction ϕ_c of about 0.16 [8].

A key quantity in the statistical description of dense random packings of particles is the average number of contacts of a particle (average coordination number Z), which is generally

a function of the *packing fraction* 1- ϕ [10]. The main question that we address here is the functional form of its variation in a 2d foam over a wide range of liquid fractions below the wet limit [11].

In the dry limit $Z = 6$ as a consequence of Euler's theorem, which relates the number of faces, edges, and vertices of a cellular structure to a topological invariant [8]. Z remains equal to 6 up to about $\phi \approx 0.03$; the Decoration Theorem [8] states that such a foam can be obtained by simply "decorating" a dry foam with three-sided Plateau borders. Upon a further increase of liquid fraction, Z begins to decrease, as neighbouring bubbles lose contacts and Plateau borders with more than three sides start to appear.

At the wet limit [12] ϕ_c, also called the jamming point in the context of granular packings, the critical coordination number Z_c is four. This can be shown by matching constraints due to bubble-bubble contacts with the number of degrees of freedom, i.e. two in two dimensions; since each contact provides a constraint for two bubbles, this leads to an average value of $Z_c = 4$ in an infinitely large 2D foam [10, 13].

Using the PLAT software [14] , which accurately represents gas-liquid interfaces as arcs of circles and then directly implements Plateau's laws, Winkelmann et al. [15] carried out 600,000 independent simulations of foams comprised of 60 bubbles each. Their data showed that Z varies linearly over a wide range of ϕ. A fit of the data in the range $0.12 < \phi < \phi_c$ to the form

$$Z\text{-}Z_c \propto (\phi_c\text{-}\ \phi)^{\beta} \qquad (1)$$

gives an exponent β =1.000±0.004 (for details of the simulations see [16]). Note, however, that a linear variation gives an excellent description of the data for the even larger range of ϕ exceeding about 0.08.

Here we present simulations which are also based on the standard foam model, as introduced above. However, we have performed these with the Surface Evolver software, which allows for the setting of a finite contact angle, and thus the addition of an attractive force between bubbles, as detailed below.

As in the PLAT calculations, we start with a dry foam (small ϕ) and progressively increase the liquid fraction. We again find a linear variation of the average coordination number for an extensive range of liquid fractions below $\phi_c \approx 0.16$. We also note that bubble attraction allows foam structures to be found for higher values of ϕ [17], which we explore below.

Many other results for the variation of average coordination number with packing fraction are concerned with jamming; starting from a loose random assembly of particles, isotropic compression of the system leads to the onset of rigidity at some critical value of packing fraction.

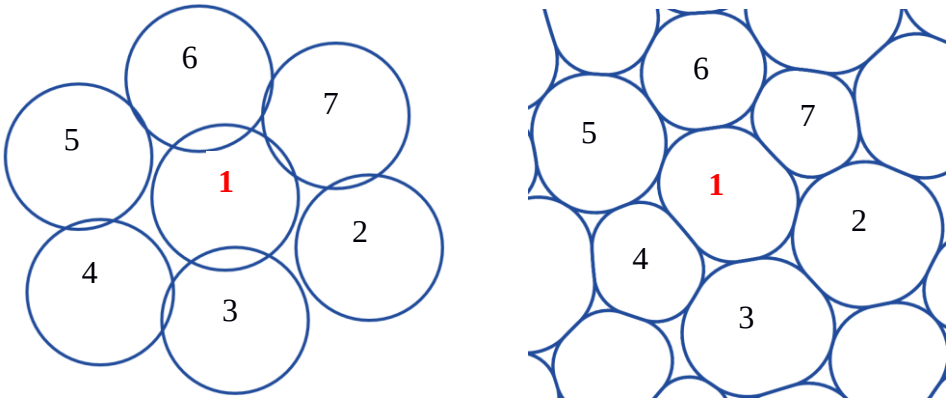
To explore the relevance of these results to our own work, we distinguish between packings of soft particles, which in 2D are represented by overlapping disks, and deformable particles, which conserve their areas, and may correspond to bubbles in a wet foam. The particles may interact purely repulsively, or may exhibit some attraction.

The earliest soft particle model appears to be that of Durian, also known as the soft disk model [18]. It has often been used as a toy model for 2d wet foams as it illustrates both quasi-static and rheological properties of foams [19]. At liquid fractions below the wet limit the disks overlap but repel each other with a harmonic potential. A 2d bubble model foam then corresponds to a minimal-overlap packing of disks for a given value of liquid fraction [18]. Figure 1 illustrates the difference between an overlapping disk packing and a packing of deformable bubbles. In the soft disk model, the exponent in equation (1) is $\beta = 0.5$ [10].

Koeze et al. [20,21] extended the soft disk model by adding an attractive shell around the (harmonically) repulsive core of the particles. Strong attraction leads to the formation of rigid clusters of particles, even at very loose packings, which grow continuously as the packing is compacted above the critical packing fraction (a second order continuous phase transition). For weak attraction the onset of rigidity is sudden (first order transition).

Dunne *et al.* [22] implemented the Morse-Witten theory to model a foam consisting of deformable and repulsive bubbles. The theory accounts for the non-local and anharmonic character of bubble-bubble interactions and is exact in the wet limit [23, 24]; as for the soft disk model, the simulations [22] also find a square root scaling (i.e. $\beta = 0.5$).

Otherwise, packings of deformable particles, as we generate here, currently appear to be largely restricted to those exhibiting repulsive interactions. Boromand *et al.* [11] developed a model for deformable bubbles and emulsion droplets which is exact in the wet limit. Away from this limit, the excess contact number variation may be expressed as an expansion, $\Delta Z \sim (\phi_c - \phi)^{1/2} + (\phi_c - \phi)$. Note that the linear variation which features in the PLAT data and that we find below for attractive bubbles only shows the second of these two terms over a large range of ϕ.



(a) (b)

Figure 1. Computer simulations of wet 2d foams. (a) In the bubble model, the bubbles are disks that can overlap. In the configuration shown the bubble labelled 1 contacts three other bubbles. (b) In a packing of deformable bubbles, here simulated with Surface Evolver, the non-circular shape of bubbles will generally lead to a different local foam structure. In the configuration shown, which is similar to that of (a), the bubble labelled 1 contacts six other bubbles. Thus a similar local neighbourhood might be associated with different coordination numbers in different models.

The variation of $Z(\phi)$ has been studied experimentally for planar elastic disks [25] and quasi-two-dimensional foams [26]. The quasi-2d foams consisted of disordered rafts of bidisperse bubbles between a liquid pool and a glass plate. Varying the distance between the plate and the liquid surface compresses the bubbles against each other, resulting in a change of nominal 2d liquid fraction. In the wet limit, where the gap between the covering plate and the liquid interface is similar to the bubble diameter parallel to the plate, the description of the foam as 2d becomes more and more approximate. The determination of average contact number from image analysis is not an easy task, and the oft-used assumption in the analysis that the bubbles can be treated as circles might lead to bias towards the bubble model. Katgert and van Hecke [26] report an exponent β of 0.5 or 0.7 (depending on how liquid fraction is measured), with Z_c close to 4 and ϕ_c close to 0.16, which is somewhat inconclusive.

In the following we describe computer simulations using the Surface Evolver software [5] to investigate the variation of $Z(\phi)$. As mentioned above, this is the standard software for the simulation of (dry) foams, particularly in 3d, although it has so far only found limited application to random wet foams [17]. Here we apply the Surface Evolver to 2d foams with varying liquid fraction. Foam structures are computed via the minimisation of energy (total surface energy (3d), total line energy (2d)) under the constraint of constant bubble size (volume, area) [27].

Of particular interest to the problem considered here is that the Surface Evolver allows the setting of a finite contact angle θ [17], as illustrated in Figure 2. A finite contact angle is a consequence of having different values for surface tension in bulk and in liquid films, as is often encountered in experiments. This results in increased bubble-bubble attraction, or stickiness, which may inhibit the reorganisation of foam structure upon a change of liquid fraction. The smallest contact angle that we can simulate in Surface Evolver is 0.81°.

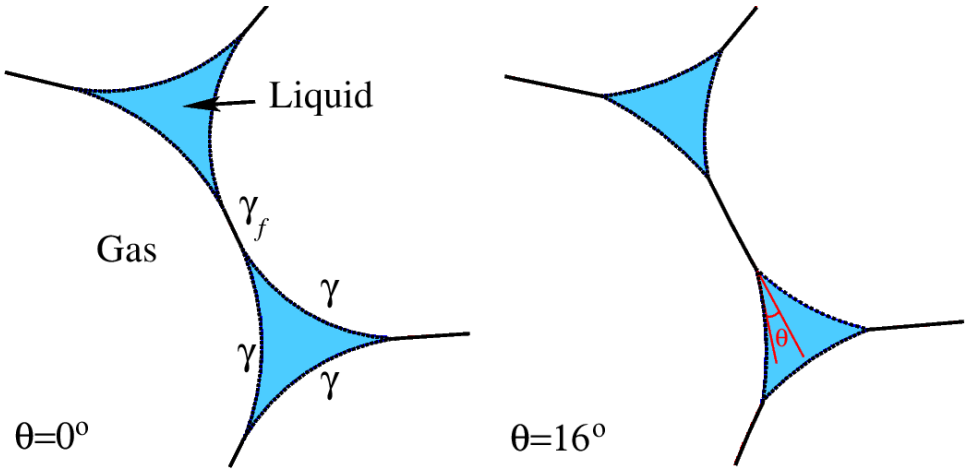


Figure 2. Two Plateau borders with contact angle $\theta = 0°$ and $\theta > 0°$ between the gas-liquid interface and the gas-liquid-gas interface. Here γ_f is the surface tension of the gas-liquid-gas interface (i.e. a liquid film) and γ is the tension of the gas-liquid interface. In an equilibrium state, $\gamma_f = 2\gamma \cos\theta$.

The detailed simulation procedure will be described in Section 2, followed by a presentation of the results for $Z(\phi)$ for foams of a range of different contact angles in Section 3. Finally, Section 4 presents the conclusions.

2 Simulation procedure

In our simulations using the Surface Evolver [5], the equilibrium structure of a wet 2d foam at fixed liquid fraction is obtained by minimizing the total energy of the interfaces under the constraint of fixed bubble areas**.** The energy is a sum of two contributions: from the films separating neighbouring bubbles and from the interfaces of the Plateau borders, weighted by their respective interfacial tensions.

The tension γ_f of a film (gas-liquid-gas interface) is set to 1, without loss of generality. The tension γ of a single gas-liquid interface is chosen just above 0.5 to set the desired contact angle θ [17] from

$$\theta = \cos^{-1}\frac{\gamma_f}{2\gamma}. \tag{2}$$

In this way we simulate foams with contact angles from less than 1° to almost 12°.

All our foam samples were polydisperse, with N=1600 bubbles. Polydispersity is evaluated as the standard deviation of the distribution of bubble radii R, divided by the average bubble radius, defined in terms of the means $\langle R\rangle$ and $\langle R^2\rangle$ of the distribution of R as

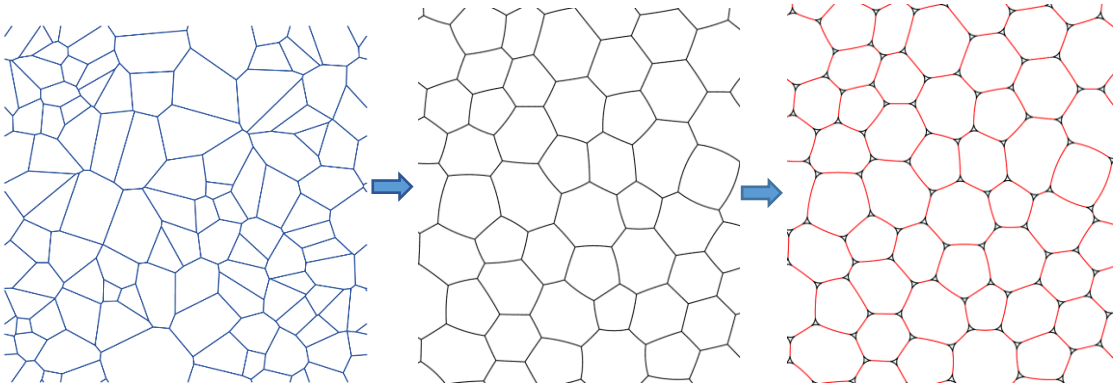
$$\sigma_R = \sqrt{\frac{\langle R^2\rangle}{\langle R\rangle^2} - 1}\,. \tag{3}$$

Therefore, for a monodisperse (but not necessarily hexagonal) foam, $\sigma_R = 0$. The data presented below is for σ_R = 0.0079.

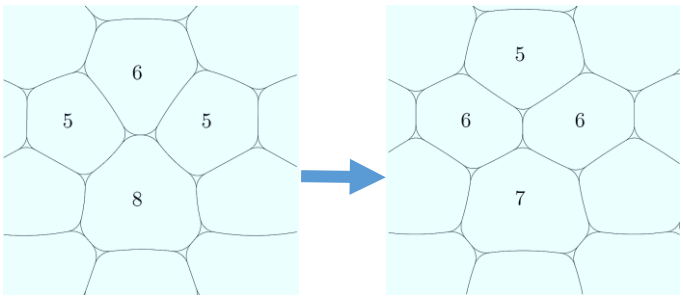
The simulation procedure is described in [17]. Briefly, we construct the initial foam structure from a Voronoi partition with periodic boundary conditions, as shown in Figure 3(a), resulting in a uniform random distribution of bubble areas. The films are represented as arcs of circles and Surface Evolver is used to find an equilibrium dry foam with these bubble areas (Figure 3(b)). Then three-sided Plateau borders are added at each threefold vertex to create a wet foam with low liquid fraction, typically between 0.01 and 0.02, as shown in

Figure 3(c). Conjugate gradient descent, interspersed with occasional Hessian iterations, is used to find a new equilibrium with energy accurate to eight decimal places.

The liquid fraction is increased in small steps of 0.002, with minimization at each step. During this equilibration, T1 topological changes [28] are triggered when the length of an interface decreases below a small cut-off. At lower liquid fraction, only T1 rearrangements that do not change Z are possible (see Figure 3(d)), but this is not the case above about ϕ = 0.03. We also account for bubbles coming into contact and two interfaces merging partway along their lengths. At the end of each step, the average coordination number is recorded.



(a) Voronoi network (b) A dry foam (c) A wet foam



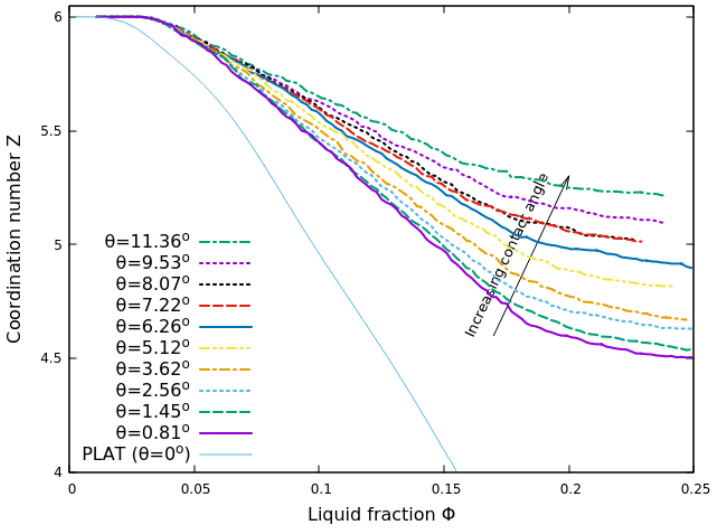
(d)

Figure 3. Surface Evolver simulation of 2d foams. (a-c): Preparation of a wet foam structure with the contact angle 5.12° and the liquid fraction 0.013 (note that a part of the 1600-bubble foam is shown). (d) Example of a T1 transition following an increase in liquid fraction by 0.002. Note that in this low liquid fraction foam (Φ < 0.03) the average coordination number, Z, stays constant, although the number of neighbours of each bubble

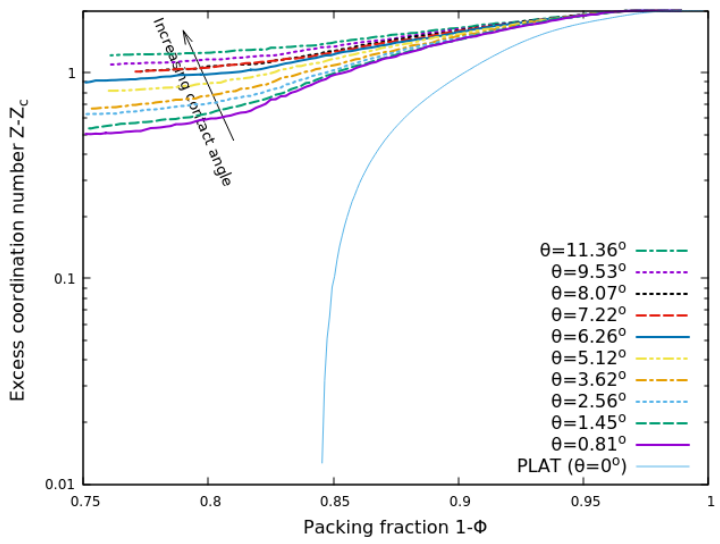
involved in the T1 changes. Increasing the liquid fraction to higher values leads to a decrease in Z as lost contacts are not necessarily regained following bubble rearrangements.

3 Results and analysis

Figure 4 shows the computed variation of the average coordination number Z with liquid fraction for different contact angles θ up to 12^o All data sets show the same qualitative behaviour: at values of ϕ less than about 3% the coordination number remains roughly constant and close to the value six (as a consequence of the Decoration Theorem) and then a further increase in liquid fraction leads to a regime in which Z decreases linearly with liquid fraction.



(a)



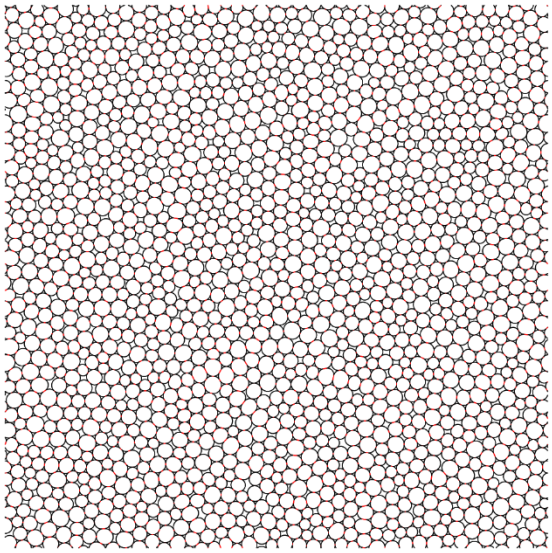
(b)

Figure 4. Surface Evolver results for the variation of average coordination number Z with the liquid fraction, for foam samples with N = 1600 bubbles and polydispersity 0.0079. (a) For values of liquid fraction greater than about 0.07 and less than about 0.15, $Z(\phi)$ decreases linearly with ϕ. Also shown is the data for zero contact angle obtained with PLAT [15] . (b) The same data is shown on log-linear scales (cf [20]) to illustrate the difference between the PLAT data and the finite contact angle data.

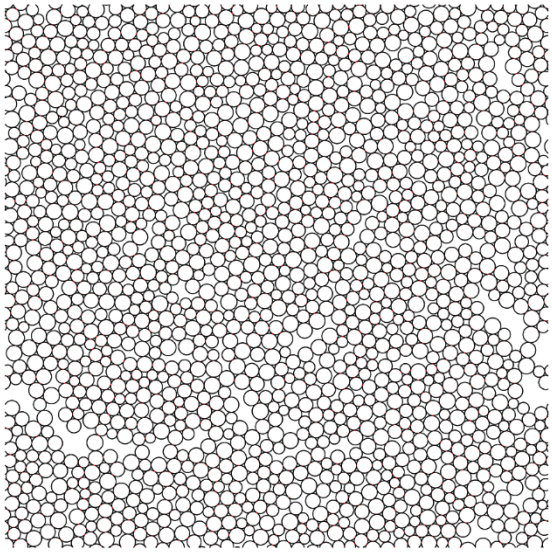
In the case of zero contact angle (i.e. the PLAT data) this linear regime extends all the way to ϕ_c where $Z \approx 4$. For our new data with finite contact angle, we find that $Z(\phi)$ begins to flatten off when the liquid fraction exceeds about 0.15, and then approaches a constant value, greater than 4. This is a result of "flocculation" [17]: bubbles cluster together, leading to inhomogeneities in the foam structure (Figure 5).

As the liquid fraction increases, large liquid regions develop from the four-sided Plateau borders, similarly to what is found in the simulations of sticky disks by Koeze *et al.* [20, fig. 1a]. Eventually one of these comes to dominate the sample, and the bubbles form a single large cluster surrounded by liquid. The bubbles in this cluster are close to circular, although there can be large liquid regions still present within the cluster. However, the surrounding

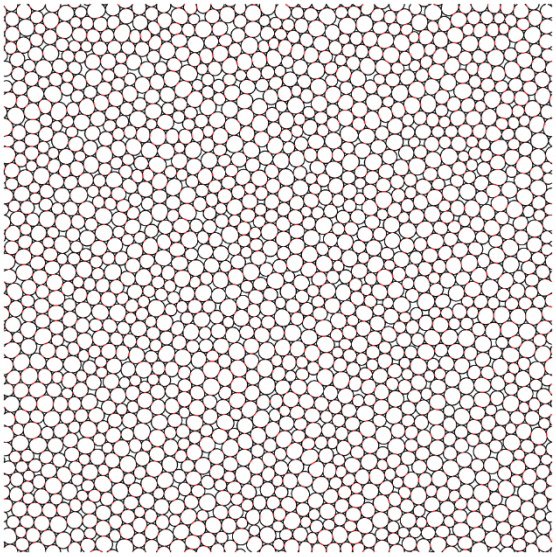
liquid can absorb any further increases in the liquid fraction without the bubble shapes changing, and hence the value of Z saturates. In addition, the fact that the cluster is surrounded by liquid ensures that the *average* coordination number saturates to a value above 4.



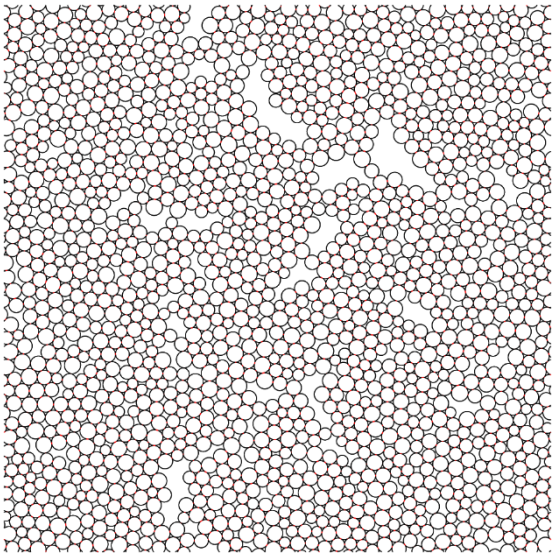
(a) θ =0.81° and ϕ =0.099



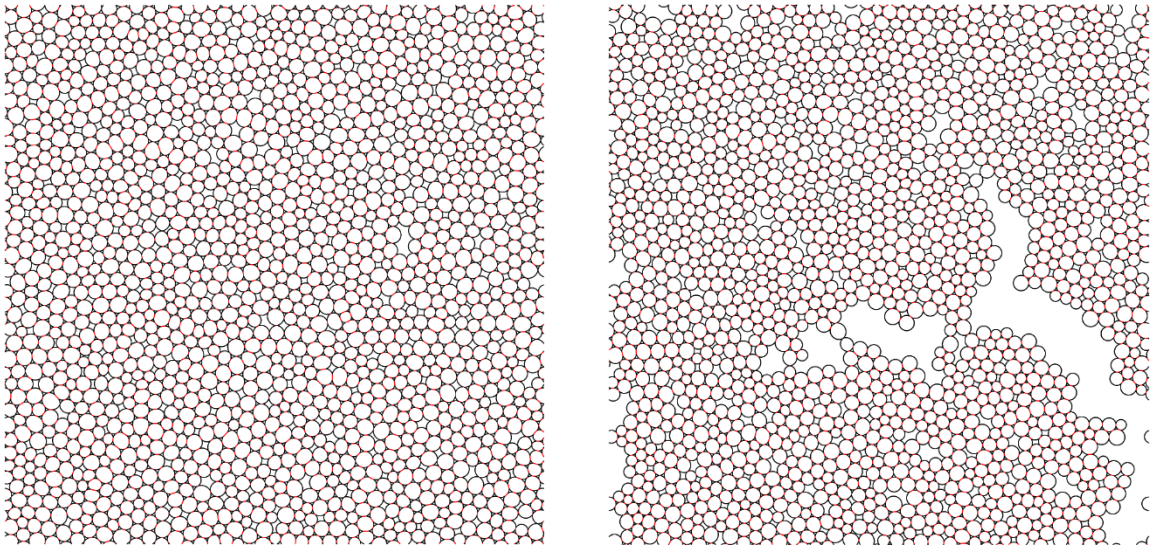
(b) θ =0.81° and ϕ =0.199



(c) θ =5.12° and ϕ =0.099



(d) θ =5.12° and ϕ =0.199



(e) θ =11.36° and ϕ =0.099 (f) θ =11.36° and ϕ =0.199

Figure 5. Foam structures with different contact angles and the liquid fractions ϕ = 0.099 and 0.199. At higher contact angles, the liquid is less evenly distributed, as bubbles begin to cluster together and flocculate, and large liquid regions form.

The slope of Z(ϕ) in the linear regime depends strongly on the contact angle. As the contact angle approaches zero, the bubbles "share" less interface (they are less "sticky") and the average coordination number decreases more quickly as the liquid fraction increases. In addition, varying the polydispersity at fixed contact angle (data not shown) does not change the slope of $Z(\phi)$ in the linear regime. Only beyond this regime do the curves separate.

We have determined the slope dZ/dϕ in the linear $Z(\phi)$ regime using linear regression. This is shown in Figure 6. The slopes are consistent with the data of Boromand *et al.* [11] who find, for soft deformable particles, slopes of around 4 in their linear regime.

A fit to the empirical expression

$$\frac{dZ}{d\phi} = c_1 - c_2\theta \tag{4}$$

accurately describes our Surface Evolver simulations with fitted constants c_1 =9.90 and c_2 = 0.45.

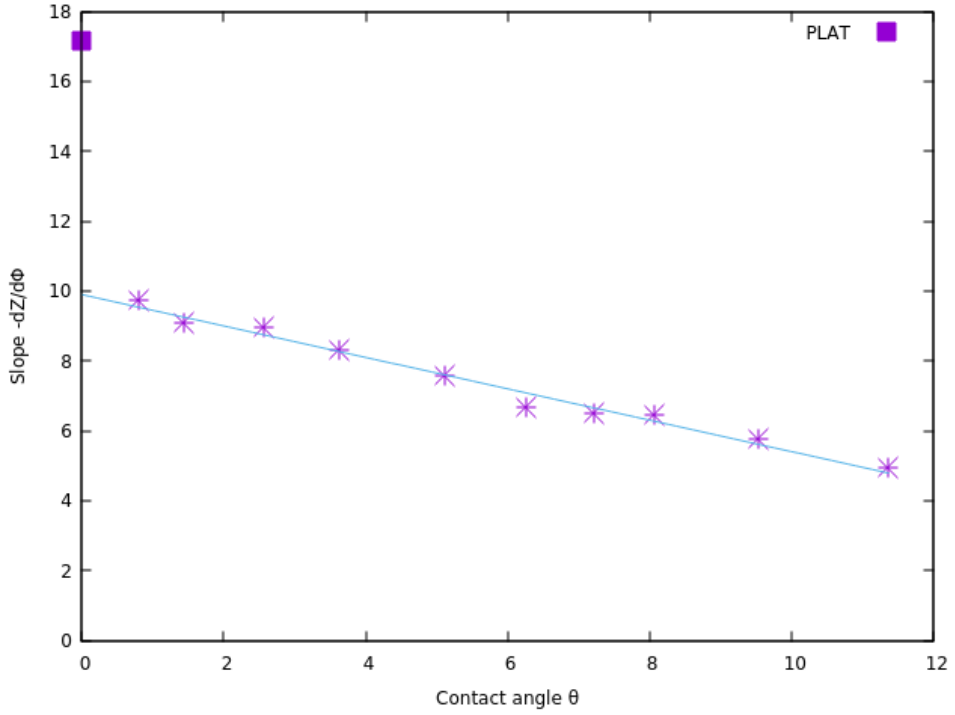


Figure 6. The slope of the linear region in the variation of the coordination number with liquid fraction as a function of contact angle θ. It may be described by linear relationship Equation (4). The PLAT simulation, for contact angle zero, indicates that the limit in which the contact angle tends to zero is discontinuous.

The Surface Evolver results for the finite contact angle are broadly consistent with the previous PLAT results in that they too reveal a linear variation of $Z(\phi)$ for moderate values of ϕ. We stress here again that the methods of calculation used by Surface Evolver and PLAT differ: the former uses energy minimisation, the latter equilibrates a foam structure via balancing the forces due to surface tension and pressure differences. The linearity of $Z(\phi)$ is thus not an artefact of a particular numerical procedure.

However, the value of the slope for the finite contact angle does not extrapolate to that of zero contact angle, because even a small non-zero contact angle induces flocculation.

In their study of soft (non-deformable) "sticky disk" packings, Koeze *et al.* [20] vary the strength of the attraction between disks and compute the variation of the excess coordination number ΔZ as a function of packing fraction 1- ϕ [20] (the quantity n_r in their fig. 4 corresponds to $2\Delta Z$). While in the limit of weak attraction there is a sharp drop in ΔZ *as* $1\text{-}\phi_c$ is approached, with any finite attraction present the decrease in ΔZ is more gradual. In foams, *i.e.* in packings of deformable bubbles, it appears that the contact angle plays a similar role to Koeze et al's attraction parameter. In Figure 4(b) we have replotted our data from Figure 4(a) to match the presentation of Koeze *et al.* [20, fig. 4] and find broadly similar features. This presentation also emphasises the distinction between the PLAT data, with zero contact angle, and our finite contact angle data, which we already noted when discussing the variation of slope $dZ/d\phi$ in the linear $Z(\phi)$ regime in Figure 6. The excess coordination number for our PLAT data drops more quickly than for any sticky disk system, while with a finite contact angle the data saturate at a relatively high value of Z (and thus ΔZ). Our computations are very time-consuming and we do not have sufficient data to compute what Koeze *et al*. [20] call the "sticky jamming point" $\phi_c(\theta)$, or indeed examine any other scaling relations related to the shear modulus for example.

4 Conclusion

We have simulated two-dimensional wet foams with deformable bubbles and finite contact angle to analyse how the coordination number of these structures is affected by the liquid fraction. We identify a linear regime for all foam structures, covering a wide range of liquid fractions, in which the coordination number decreases linearly with liquid fraction. This linearity contrasts with data from the bubble model [29], which show a square-root dependence. In the linear regime, the slope itself depends linearly on the contact angle, and the effect of polydispersity is negligible.

Beyond the linear regime, at even higher liquid fractions, there remain unanswered questions. In particular, it is unclear how to define the usual wet limit since the coordination number never reaches 4. Instead, the contact angle, and to a lesser extent the polydispersity in bubble size, controls the value of Z to which the curves saturate.

At these high liquid fractions, our simulations indicate that the foam becomes a large cluster of bubbles, surrounded by an extensive body of liquid. In this respect, it is difficult to distinguish the response of the flocculated cluster from the response of the system as a whole.

Similar effects are encountered in what was recently called “sticky jamming” [20], i.e. the jamming of particles with attractive forces, and we have explored the similarities between these systems and foams.

Acknowledgments

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