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LETTER TO THE EDITOR

The osmotic pressure of a two-dimensional disordered foam

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Abstract. The osmotic pressure for a disordered two-dimensional foam or emulsion is evaluated by a realistic computer simulation over the whole range of the liquid fraction. The data are consistent with a quadratic approach to zero as the liquid fraction nears the limit of stability of the froth.

The concept of osmotic pressure in a foam can be explained by a simple *gedanken* experiment. In the extreme case of a wet foam, the bubbles are spheres (in 3D) or circles (in 2D) that are touching each other. Extracting the liquid causes the bubbles to deform, increasing their surface area and hence the energy of the system. Thus there is a force necessary to counter the increasing bubble-bubble interaction when the foam is drained; the force per unit area (or unit length in 2D) is called the osmotic pressure Π . It is zero in the extreme limit of a wet foam and goes to infinity for a dry foam for which the bubbles have polyhedral shapes.

The osmotic pressure (in 3D) may be defined as

$$\Pi = - \left(\frac{\partial F}{\partial V} \right)_{V_g} \quad (1)$$

where F is the free energy of the foam (that is its surface energy), V is the total volume of the sample and V_g the volume of the gas in the sample. Note that V_g is kept fixed when the derivative is taken, assuming an incompressible gas. With small changes of terminology the same definition applies to an emulsion in which droplets of one liquid are dispersed in another liquid.

In this letter we shall restrict ourselves to the analysis of two-dimensional foams. These can be approximated experimentally by squeezing a foam between two glass plates. In equation (1) F , V and V_g then have to be replaced by the line energy E , the total area A of the sample and the area of gas in the sample A_g respectively.

Princen [1] calculated the osmotic pressure for an ordered (monodisperse) two-dimensional system analytically

$$\Pi = \frac{\sigma}{R} \left(\frac{\Phi}{\Phi_c} \right)^{1/2} \left[\left(\frac{1 - \Phi_c}{1 - \Phi} \right)^{1/2} - 1 \right] \quad (2)$$

in terms of the line tension σ , the radius of an undeformed bubble R and the gas fraction of the foam Φ . The critical packing fraction Φ_c denotes the value of the gas fraction at which the foam disintegrates into isolated bubbles. For the ordered hexagonal 2D foam, it is given by

$$\Phi_c = \frac{\pi}{2\sqrt{3}} \simeq 0.907.$$

Integration of equation (2) according to equation (1) then gives the work ΔE per unit area of foam (this area being evaluated at $\Phi = \Phi_c$), necessary to compress the foam from Φ_c to Φ . This leads to

$$\Delta E(\Phi, \sigma, R) = 2\Phi_c \frac{\sigma}{R} \left[\frac{1}{(\Phi_c \Phi)^{1/2}} - \left(\frac{1 - \Phi_c}{\Phi_c} \right)^{1/2} \left(\frac{1 - \Phi}{\Phi} \right)^{1/2} - 1 \right]. \quad (3)$$

The osmotic pressure as a function of the liquid fraction $\Phi_l = 1 - \Phi$ according to equation (2) is shown in figure 1 (solid line), where we used the following normalization: $\Pi^* = \Pi A^{1/2}$. Here, A is the area of a bubble, $A = \pi R^2$.

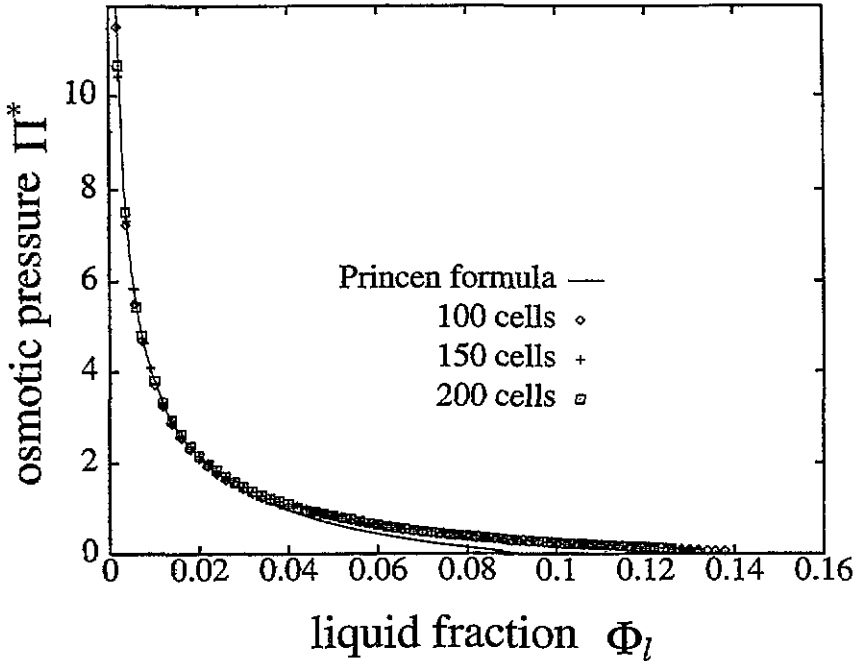


Figure 1. The osmotic pressure Π^* for ordered and disordered two-dimensional foams. The solid line represents Princen's normalized analytical result for ordered foam and the symbols represent simulation data for a disordered foam.

Using the decoration theorem of Bolton and Weaire [2] it may be shown that equation (2) is also valid for a disordered froth to within a very good approximation, so long as the plateau borders remain three sided. This will generally hold over some range close to the dry foam limit $\Phi_l = 0$. Our aim was to extend Princen's results to disordered wet foams where numerical methods have to be applied. There are experimental measurements of osmotic pressure as a function of liquid fraction in foams [3] and emulsions [4, 5]. Inasmuch as these are made on three-dimensional systems, there can be no immediate comparison with our results. Nevertheless, they may be useful in developing a fuller theoretical understanding of the problem posed by these measurements [5].

We used a computer program developed by Bolton and Weaire [2, 6] to simulate two-dimensional liquid foams. Figure 2 shows examples of 150-cell systems with liquid fractions $\Phi_l = 0.12$ and $\Phi_l = 0.02$ respectively. A straightforward approach to calculating the osmotic pressure would be to take the numerical derivative of the line energy of the cell

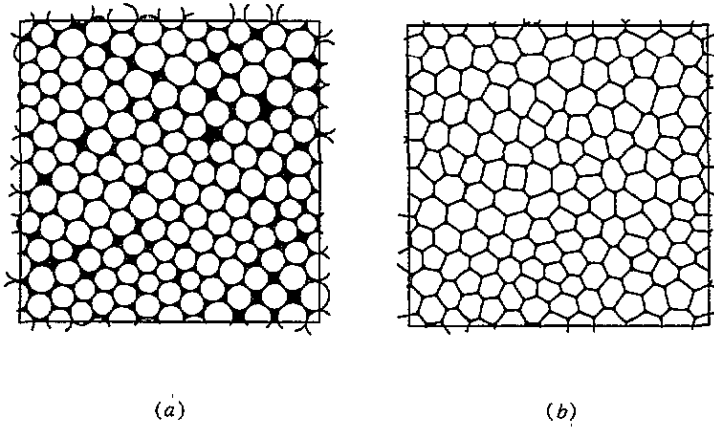


Figure 2. Examples of two-dimensional disordered foams with (a) liquid fraction $\Phi_l = 0.12$, 150 cells, (b) liquid fraction $\Phi_l = 0.02$, 150 cells.

system as a function of the gas or liquid fraction; see equation (1). The line energy is proportional to the sum of all the arc-lengths, a quantity that is readily evaluated by our program. This method is not very satisfactory in practice because the topological changes that inevitably occur as we change the liquid fraction are associated with discontinuities in the total energy. The correct interpretation of equation (1) is in terms of the derivative taken between such discontinuities.

We therefore calculated the stress components τ_{xx} and τ_{yy} directly by summing up the force components (that is pressure and surface tensions) acting on the boundaries perpendicular to the x and y directions respectively. The osmotic pressure is then given by

$$\Pi = \frac{1}{2}(\tau_{xx} + \tau_{yy}) - p_{pb} \quad (4)$$

where p_{pb} is the average plateau border pressure. We then normalize our results by $\Pi^* = \Pi \bar{A}^{-1/2}$, where \bar{A} denotes the *mean* cell area.

Similar methods have been used by us to calculate the shear modulus as a function of the gas fraction in a disordered two-dimensional foam [7]. We found it to fall linearly towards zero at the point at which the bubbles come apart. This result still lacks any complete or rigorous theoretical derivation. However, if a loose analogy with rigidity percolation [8] is accepted, it leads to the expectation of a similar linear variation of all the elastic constants and the bulk modulus [9]. As the latter corresponds to the first derivative of the osmotic pressure with respect to Φ , one would expect a *quadratic* variation of the osmotic pressure, as a function of $\Phi - \Phi_c$. For a disordered foam, the gas fraction at which the foam separates into isolated bubbles (the rigidity loss transition) is given as $\Phi_c \simeq 0.84$ by Bolton and Weaire [8].

Figure 1 shows the results of our simulation for three different samples consisting of 100, 150 and 200 cells respectively. The osmotic pressure was calculated by use of equation (4). Figure 3 shows that the osmotic pressure of a disordered system Π_d can be fitted very well to a quadratic in the wet limit

$$\Pi_d^* = a(b - \Phi_l)^2. \quad (5)$$

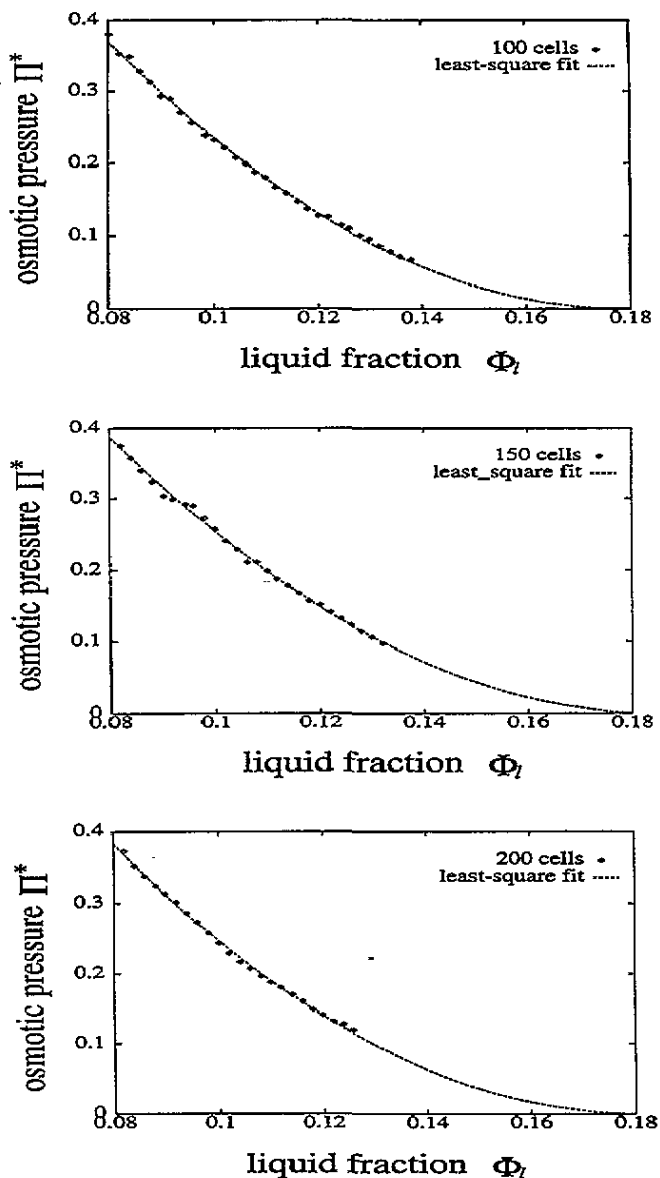


Figure 3. Close to the rigidity loss transition, the osmotic pressure for disordered systems can be fitted to a quadratic (see text). Data shown are from 100-, 150- and 200-cell systems.

The parameters a and b were determined to be $a \simeq 37$, $b \simeq 0.18$ in obtaining the fitted curve shown. Note that the parameter b represents the value of the critical liquid fraction which was previously given by $\Phi_{c,l} = 1 - \Phi_c \simeq 0.16$. We associate this discrepancy with the uncertainty of our extrapolation, as follows.

The shear modulus G as a function of Φ can be described over the whole range of gas fractions by the functional form $G = c - d * \Phi_l^2$ where c and d are fit parameters [7]. This gives $\Phi_{c,l} \simeq 0.16$. For low liquid fractions however, G should strictly speaking be

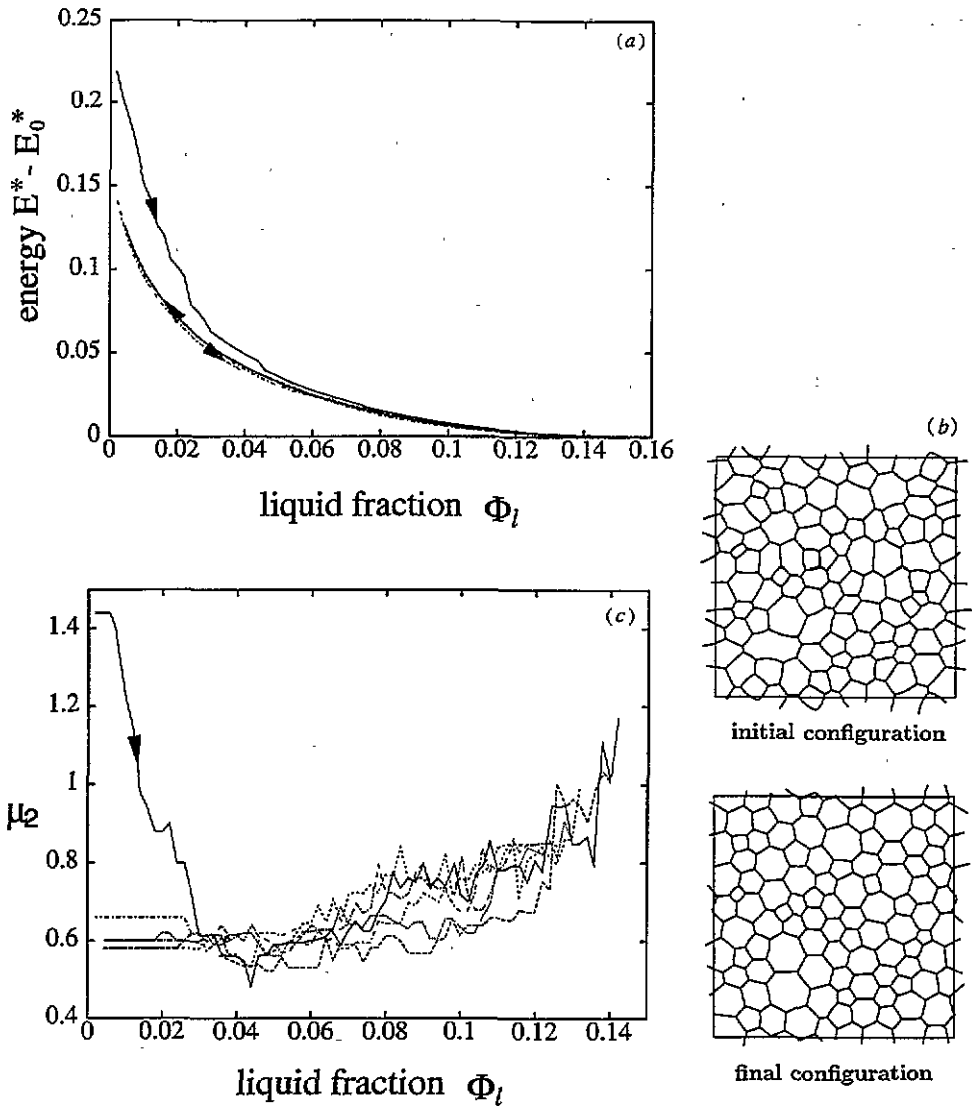


Figure 4. An increase followed by a decrease of the liquid fraction leads to a more ordered and energetically favoured system. Data are for a 150-cell system. (a) The normalized free energy $E^* = E\bar{A}^{-1/2}/A_{foam}$ is plotted relative to its value E_0^* for separated bubbles. (b) Initial and final configuration of the foam. (c) The second moment of the distribution of number of cell sides μ_2 .

constant (decoration lemma) and hence it might seem better to fit G to a linear decrease for $\Phi_l > 0.04$, that is, when the first four-sided plateau borders occur. The critical liquid fraction is then determined as $\Phi_{c,l} \approx 0.18$, in agreement with our results gained from the osmotic pressure calculations.

The limited range of the calculations makes the claimed critical behaviour quite tentative and we cannot exclude power laws of the form of equation (5), with indices β close to two. Similarly, the recent experiments in three dimensions [5] cannot be used to identify the

critical index with much precision, although it was claimed that $\beta \simeq 1$. Another recent simulation [10] in two dimensions with a more approximate model has been interpreted in terms of $\beta = 3/2$. It is not possible to reconcile or relate these various claims at this stage.

An interesting extension of our simulations is the following. After having increased the liquid fraction Φ_l in order to get from a dry to a wet foam, we decreased Φ_l to zero again and then increased it again to values close to $\Phi_{l,c}$. (We encountered computational difficulties in equilibrating the structure in the extremely wet regime.)

In figure 4(a) we plotted the normalized energy of the sample $E^* = EA^{-1/2}/A_{foam}$ during this cycle, relative to its value E_0^* for separated bubbles; A_{foam} is the total area of the *compressed* foam. (Note that Princen computes the energy per area of *uncompressed* foam.) It can be seen that the system finds a state of lower energy as a consequence of this cycle. This is due to the increased order in the final state; see figure 4(b). The osmotic pressure, however, is not significantly affected by the cycling. In figure 4(c) we display the variation of the second moment μ_2 of the distribution of the number of sides of a cell about its mean. This is a reminder, should this be necessary, that the structure, and therefore the properties, of a disordered foam can be strongly dependent upon its previous history.

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