Computation of Equilibrium Foam Structures Using the Surface Evolver

Robert Phelan, Denis Weaire and Kenneth Brakke

CONTENTS

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The Surface Evolver has been used to minimise the surface area of various ordered structures for monodisperse foam. Additional features have enabled its application to foams of arbitrary liquid fraction. Early results for the case of dry foam (negligible liquid fraction) produced a structure having lower surface area, or energy, than Kelvin's 1887 minimal tetrakaidecahedron. The calculations reported here show that this remains the case when the liquid fraction is finite, up to about 11%, at which point an f.c.c. arrangement of cells becomes preferable.

INTRODUCTION

A foam consists of gas cells (bubbles) in a liquid medium, separated by thin films wherever they impinge on one another. Depending on their size and the pressure of the liquid in their interstices, the bubbles may remain approximately spherical with little contact (Kugelschaum or wet foam) or be pressed together to form polyhedra with curved faces (Polyhederschaum or dry foam). Several idealisations are commonly applied to describe such a system in simple mathematical terms:

(a) The thickness of the thin films is neglected.
(b) The bubble surfaces are associated with a constant surface energy per unit area. No other energy terms are considered.
(c) Gas and liquid are both incompressible.

The problem of finding an equilibrium structure under given conditions then becomes one of area minimisation under constraints, of the general kind for which the Surface Evolver [Brakke 1992] has been developed (see also [Brakke 1991; Hsu et al. 1992]). In this paper we describe such an application of the Evolver, and present some results. The

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first result has already been reported [Weaire and Phelan 1994a], and has excited widespread interest [Rivier 1994]. It provides a structure for monodisperse dry foam having lower energy than that of Kelvin [Thomson 1887]. We will therefore begin by sketching the historical background and the implications of this result. Further calculations will be presented for other dry foam structures.

The Evolver has also been adapted to calculate energies of wet foam structures. We will present the first such results, and discuss the topological changes that occur as the liquid fraction is varied. The relevant details of the Evolver, its application to dry foams, its adaptation to wet foams and other technical aspects of the work are described in the sidebar on page 184.

As the liquid fraction is varied, the mathematics dictating the structure of foam ranges from the minimal spatial partitioning problem (space-filling cells) in the dry limit to the optimal sphere packing problem in the wet limit touching (spherical bubbles). Wet foam may give a valuable insight into the connection between these and other familiar extremisation problems, such as that of the mimimal sphere covering (filling space with equal overlapping spheres of minimum total volume).

1. SOME HISTORY

In 1887 Lord Kelvin addressed the question of the minimal partitioning of space into equal volumes [Thomson 1887]. Why he did so is not immediately apparent. Although he remarked that the problem 'is solved in foam', his motivation lay in the theories of George Green, and had to do with models of the ether. The British school of physical scientists of that period sought concrete realisations of their ideas in mechanical models. In this case, it seems that foam provided a model of the 'labile ether'.

In terms of the above definitions, Kelvin was concerned only with dry foam made up of cells of equal volume (monodisperse foam). He proceeded to describe a minimal structure for this case. Although he did not directly assert that it was the global minimum, that is, the structure of lowest energy among all possible, this idea has been imputed to him, and the question of its validity has been debated ever since.

Certainly Kelvin's is the best, indeed essentially the only, simple solution in which all cells are identical in shape and orientation, so that their centers correspond to the points of a lattice. His minimal tetrakaidecahedron (Figure 1) is associated with the body-centred cubic lattice. Its four-sided faces are flat, since they correspond to mirror planes of the lattice, while the hexagonal faces are not.

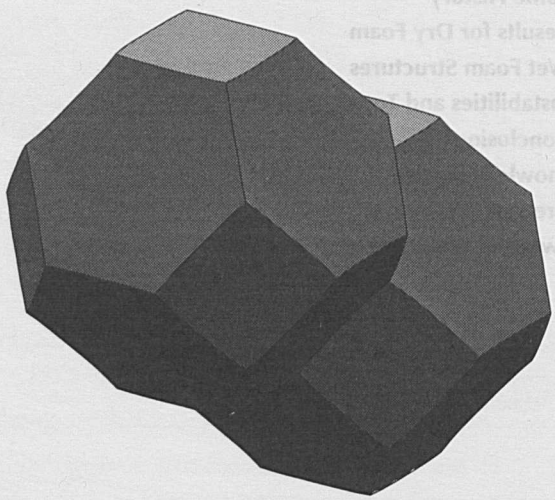

FIGURE 1. The cubic unit cell of Kelvin's structure, consisting of two of his tetrakaidecahedra.

A minimal surface between equivalent cells must have zero total curvature everywhere. Lord Kelvin showed how the hexagonal faces could be distorted in a way consistent with this requirement, in order to satisfy the conditions that the cell faces must meet at 120° and cell edges meet at an angle of $\arccos(-\frac{1}{3})$, the tetrahedral angle. These equilibrium conditions were familiar to him from the empirical work of Plateau [1873]. Kelvin's description of the subtle curvature of the hexagonal faces was based on an approximation that reduced the problem to one of solving Laplace's equation for the local displacement of the surface.

The surface energy of Kelvin's structure was calculated by Princen and Levinson [1987], who also

compared it with other simple structures, most of which are mechanically unstable or non-space-filling. In the meantime it had become common opinion that Kelvin's analysis was irrelevant to real foams. This was not just because it was concerned with bubbles of equal size, but also because it was thought that in practice such a macroscopic system could not be expected to find its global minimum. We shall not pursue the rather convoluted origins of this thinking, or the experiments upon which it was based [Matzke 1946]. Certainly Kelvin's (attributed) proposition remained unconfirmed either by experiment or by proof for more than a century.

The present study has overthrown the proposition, but that was not its original motivation. It was primarily intended to throw light on the dependence of foam structure (and related propertes such as elastic moduli) on the liquid fraction [Weaire 1994]. It was natural to begin at the extreme of dry foam, and compare Kelvin's structure with some others, not previously evaluated, on account of their complexity, and likely to be favoured by increasing liquid fraction. The resulting comparison is presented in the next section, adding some new results for dry foams to those already published [Weaire and Phelan 1994a; 1994b].

2. RESULTS FOR DRY FOAM

The structures proposed in [Weaire 1994] for comparison with that of Kelvin were two clathrates occuring in the chemistry of tetrahedrally bonded materials. Note the close analogy between tetrahedral structures and our foam problem, provided that the bonds form closed cages. ('Clathrate' refers precisely to this kind of structure, where molecules of one kind are included in cavities of the crystal lattice of another.)

The simpler of these two structures has cubic symmetry and consists of eight cells of two types: irregular pentagonal dodecahedra, and cells with fourteen sides, twelve pentagonal and two hexagonal. The fourteen-sided cells are arranged as three mutually perpendicular, interlocking columns with the pentagonal dodecahedra lying between them on a b.c.c. (body-centered cubic) lattice. The second clathrate structure is also cubic with a unit cell of 24 cells. There are sixteen pentagonal dodecahedra and eight sixteen-sided polyhedra of twelve pentagonal and four hexagonal faces. The latter cells are arranged on the bonds of a tetrahedral diamond cubic network. Both structures have long been recognised as particularly neat solutions to the problem of space-filling with plane polyhedra [Williams 1968].

An alternative viewpoint concentrates on the cell centres, in which case the first clathrate structure is identified with the so-called A15 or β-tungsten structure, while the second is identified with the C15 structure [Rivier 1994]. See Figure 2 for the coordinates and arrangement of the A15 cell centres, and Figure 3 for the corresponding foam structure. Figure 4 (left) shows the minimised foam based on the C15 centers.

The choice of these structures was based on some vague structural principles, suggesting that the optimum number of sides for a cell is around fourteen, and that four-sided faces are expensive in terms of energy.

Recently Rivier [1994] has suggested that the ideal dry foam structure may yet be another of the many Frank–Kasper phases (which include both the A15 and C15 structures). In particular he proposed the structure of β-uranium. Figure 4 (right) shows a foam with the β-U topology.

A convenient means of comparing the relative merit of different structures is to use (a version of) the isoperimetric quotient, $36\pi V^2/A^3$, where A is the average cell surface area. Table 1 lists this quotient for several structures, including the Weaire–Phelan (WP), the C15 and Kelvin's structure. In terms of surface energy, the WP structure uses approximately 0.3% less than Kelvin's and C15 needs approximately 0.4% more. The β-uranium structure suggested by Rivier is a more efficient space filler than Kelvin but uses some 0.1% more surface area than WP.

x	y	z	x	y	z
0.00	0.00	0.00	0.50	0.50	0.50
0.25	0.50	0.00	0.75	0.50	0.00
0.00	0.25	0.50	0.00	0.75	0.50
0.50	0.00	0.25	0.50	0.00	0.75

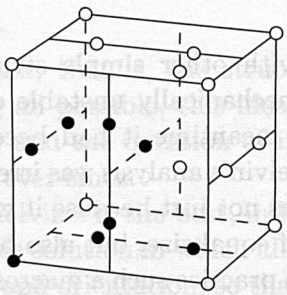

FIGURE 2. The cell centres in a cubic unit cell of the A15 structure are indicated by filled circles in the diagram; their coordinates are given in the table. For clarity, some neighbouring centres also have been shown as open circles. The body-centred positions correspond to dodecahedral sites, all other centres correspond to 14-hedral sites. A Voronoi calculation, followed by volume equalisation and surface minimisation based on these centres, using the Surface Evolver, generates the WP dry foam structure of Figure 3 [Weaire and Phelan 1994a].

USING THE EVOLVER TO MODEL FOAMS

The Surface Evolver is an interactive computer program for the study of liquid surfaces shaped by various forces and constraints. Surfaces are described as sets of triangles, permitting representation of the complicated topologies found in foams. The energy of a surface may be minimized by gradient descent and related methods.

Here we give some particular details of the Evolver's application to dry foams and of its adaption for wet foams. For a fuller description of the program, see [Brakke 1992].

The first step, common to both calculations, is to take a unit cell of the relevant periodic space-filling structure of plane-faced polyhedra as an initial input. As mentioned previously, it is most convenient to start from a unit cell of bubble centres. A Voronoi calculation based on these cell centers and using Sullivan's Voronoi Cell Solver [Almgren et al. 1991], yields the initial plane-polyhedral structure and generates an input suitable for the Evolver. For the more complex structures (A15, C15, etc.), the cell volumes need to be equalised before the surface minimisation. The final step involves repeatedly refining the surface tesselation and minimising the surface energy (with volume constraints) at each refinement level, yielding an accurate representation of an idealised dry foam.

A wet foam has a network of liquid Plateau borders in place of the triple lines of a dry foam. One may represented it in the Evolver by having one 'body' for each cell and one body for the entire Plateau border, each body being of fixed volume. Because the Evolver cannot detect intersections of surfaces, double-sided films between adjacent cells must be explicitly included as single surfaces, along with surfaces between the cells and the Plateau border. The surfaces between cells are given twice the surface tension of the surfaces shared by cells and the Plateau border.

The initial Evolver datafile for a wet foam is considerably more complicated than that for a dry foam. To ease the work, a program was written in the Evolver command language to construct a wet foam datafile from an existing dry foam whose only singular points are triple lines and tetrahedral points and whose domain is a flat torus. The program inserts a triangular tube in place of each triple edge, and inserts an octahedron in place of each tetrahedral point. Half of the octahedron's faces are omitted, since they are attachment faces for the triangular tubes. The tension of the new surfaces is set as indicated above, and the body volumes recomputed to account for the volume in the Plateau border. The resulting datafile can be run without manual editing, but it is necessary to adjust the volumes to get the desired liquid fraction.

The topology of the wet foam structures is more complex than that of the corresponding dry foam, so that the minimisation procedure must be closely controlled. In particular, since a range of liquid fractions needs to be examined, there is the possibility of topological chages. For values of φ_{liq} bracketing such a change, regions of the tesselation shrink to zero area, and care must be taken to 'repair' the surface by weeding out small triangles and at the approrate point popping the non-minimal vertex that occurs as two contacting surface pinch apart.

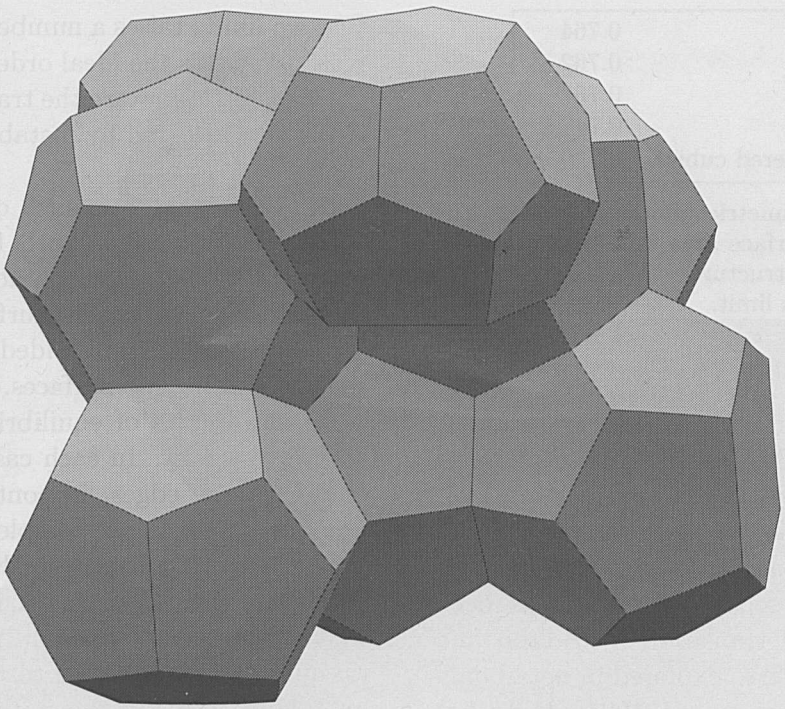

FIGURE 3. The cubic unit cell of the WP structure, consisting of six fourteen-sided polyhedra and two irregular pentagonal dodecahedra. This structure requires 0.3% less surface area than the equivalent arrangement of Kelvin cells.

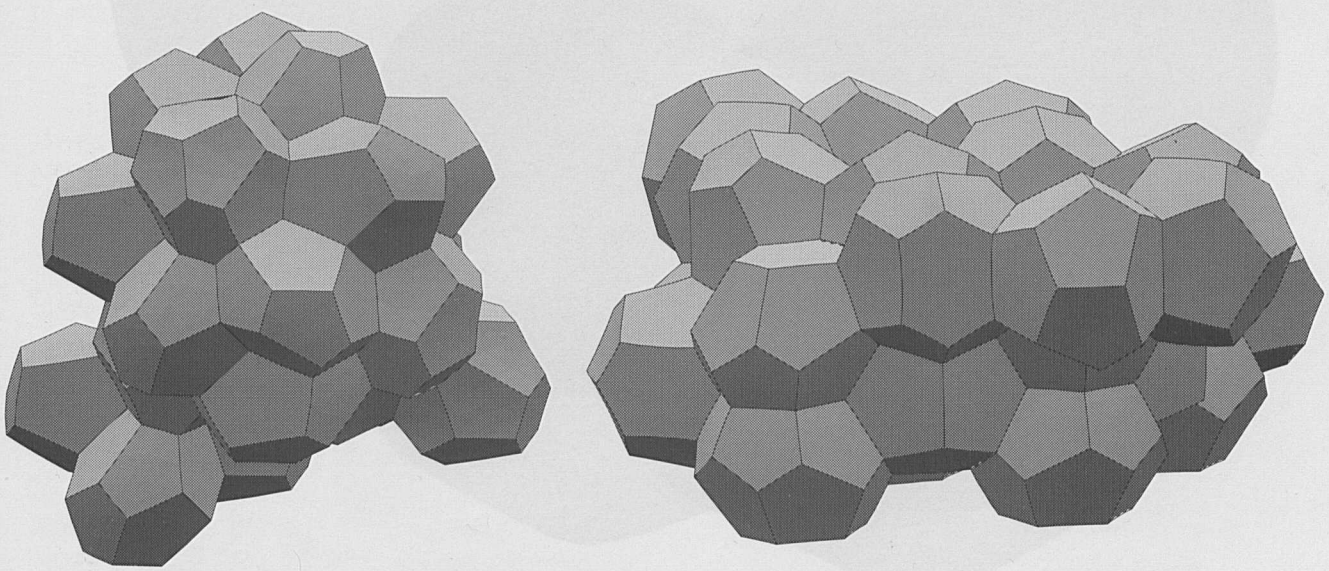

FIGURE 4. Left: The cubic unit cell of the second clathrate or C15 structure. It is built from sixteen irregular pentagonal dodecahedra and eight 16-sided polyhedra. Right: The (tetragonal) unit cell of the β-uranium structure.

Weaire–Phelan	0.764
β-uranium	0.762
Kelvin	0.757
C15	0.749
f.c.c. (face-centered cubic)	0.7405

TABLE 1. The isoperimetric quotient $36\pi V^2/A^3$ (figure of merit for surface area minimisation) of some rival dry foam structures. The f.c.c. structure is unstable in this limit.

3. WET FOAM STRUCTURES

The (volume) liquid fraction of a foam may be designated by φ_{liq}. The calculations of the previous section are for dry foam, that is, $\varphi_{\text{liq}}=0$. For any given structure there is a maximum value of φ_{liq}, defining the wet foam limit, beyond which the foam bubbles become separated. This has been called the rigidity loss transition by Bolton and Weaire [1990], and hitherto explored in detail only for two dimensions [Weaire et al. 1994]. For b.c.c. and f.c.c., the critical liquid fractions are 0.32 and 0.26, while for the WP structure it is 0.47. Consideration of the range of φ_{liq} intermediate between the two limits raises a number of questions [Weaire 1994]. What is the ideal ordered structure for each value of φ_{liq}? How are the transitions between rival structures dictated by instabilities and topological changes?

In order to address such questions, we adapted the Evolver to allow both for thin film surfaces where bubbles are in contact and for the rest of the bubble surfaces. The surface energy to be associated with the double-sided films is clearly twice that of the bubble surfaces. Figures 7–5 present typical examples of equilibrium structures generated in this way. In each case the individual cells have rounded edges, in contrast to those of Figures 1, 3 and 4. A complementary view of the structure is that based on the liquid-filled regions (the Plateau borders), as shown in Figure 8.

We can extend the definition of the isoperimetric quotient given above to the case of wet foams by taking V to be the initial cell volume of the dry foam structure. Generalised isoperimetric quotients calculated using the program are presented in Figure 9. In obtaining these data, we fixed the

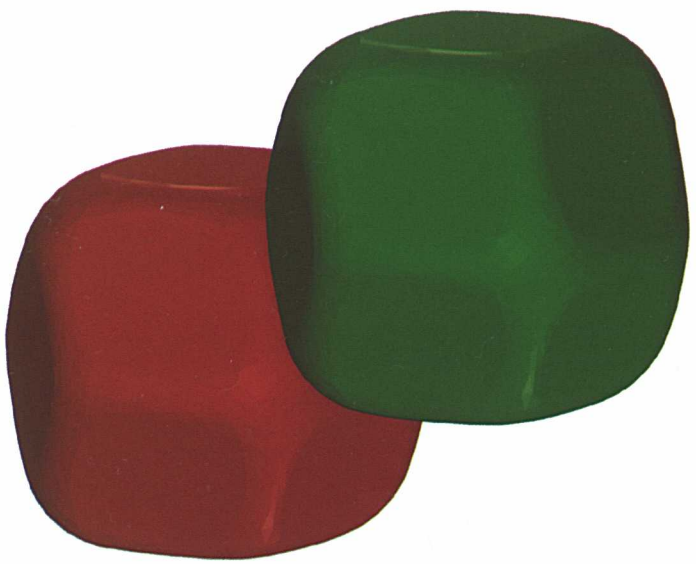

FIGURE 5. The f.c.c. structure for a liquid fraction of approximately 1%. Note the eightfold vertices, which are unstable in the limit $\varphi_{\text{liq}} \to 0$. These and the following images were generated using Geomview [Phillips et al. 1993].

FIGURE 6. The WP structure for a liquid fraction of approximately 1%.

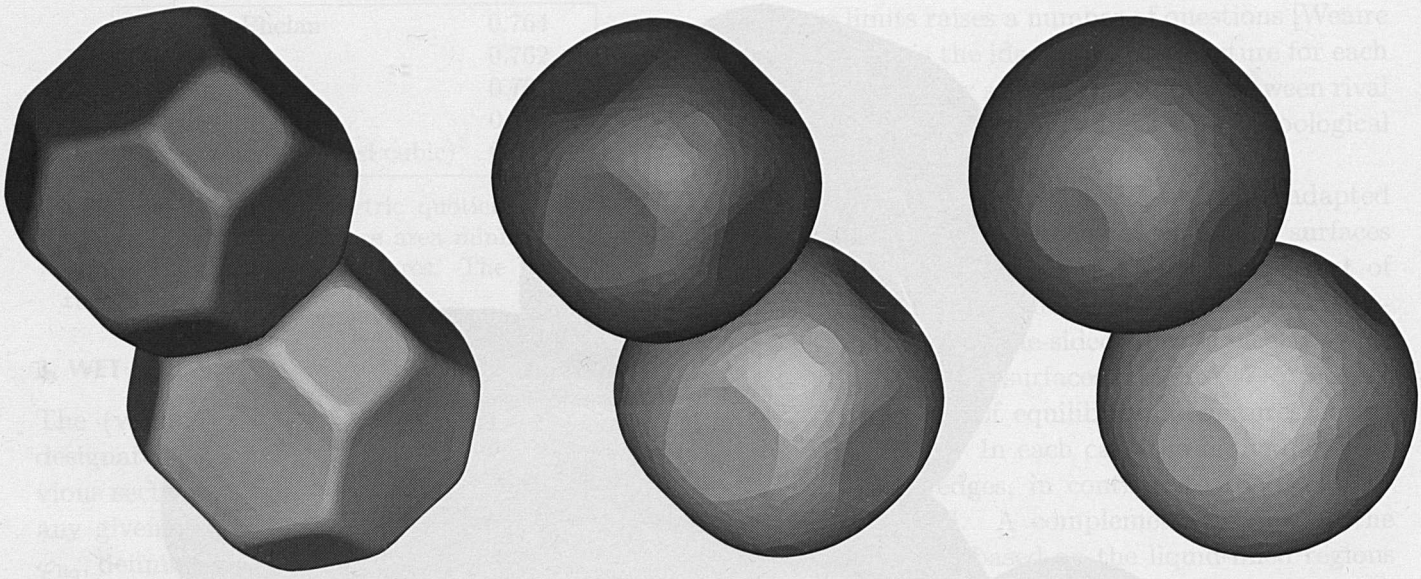

FIGURE 7. Kelvin's structure for varying liquid fractions (left, about 1%; middle, about 10%; right, about 12%), showing the disappearance of the contacts corresponding to the quadrilateral faces. The Plateau border network has been removed for clarity.

FIGURE 8. Left: A single vertex from the Plateau border network. The four edges, approximately triangular in cross section, meet at the tetrahedral angle. Right: A more extended sample of the Plateau border network, showing two vertices.

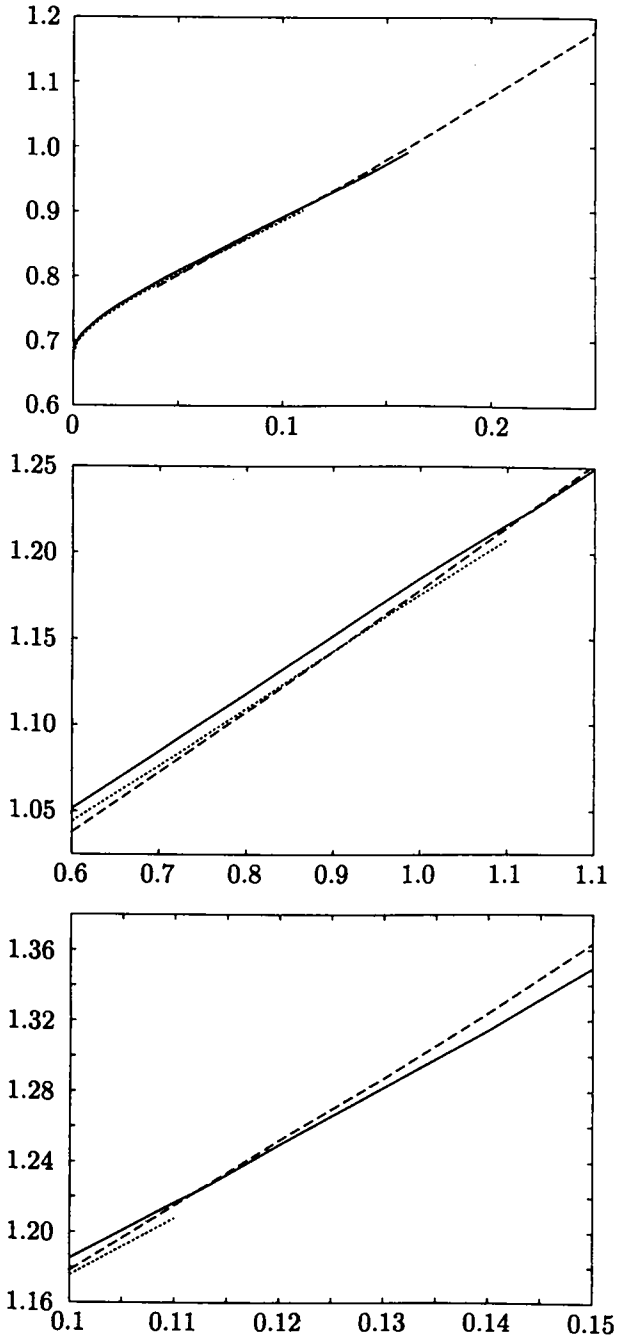

FIGURE 9. Generalised isoperimetric quotient as a function of liquid fraction for Kelvin's structure (dotted lines), the WP structure (solid lines) and f.c.c. (dashed lines). Errors have been omitted for clarity. The bottom two diagrams focus on the b.c.c. to f.c.c. crossover at $\varphi_{\text{liq}} = 9 \pm 1\%$, and on the WP to f.c.c. crossover at about $\varphi_{\text{liq}} = 11 \pm 1\%$.

lattice parameters, suppressing instability. Therefore, not all the structures plotted are necessarily stable with respect to elastic distortion. In particular, Kelvin's structure will be unstable beyond the point at which the quadrilateral contacting faces are lost (at $\varphi_{\text{liq}} = 11 \pm 0.5\%$), as discussed in the following section.

Ignoring this for the moment, we see that f.c.c. has lower surface energy than b.c.c. beyond $\varphi_{\text{liq}} = 9 \pm 1\%$. The fractional error of the data in these graphs is approximately 0.1%, as estimated from a comparison of the final reported energy of the minimisation procedure and a power-law extrapolation of the energy based on previous refinement levels. These errors have been omitted for clarity, but yield an uncertainty in the estimation of the crossover points of $\pm 1\%$ liquid fraction. Figure 10 shows the surface energy of the WP structure fitted to a functional approximation, valid for low liquid fractions, suggested by Weaire [1994].

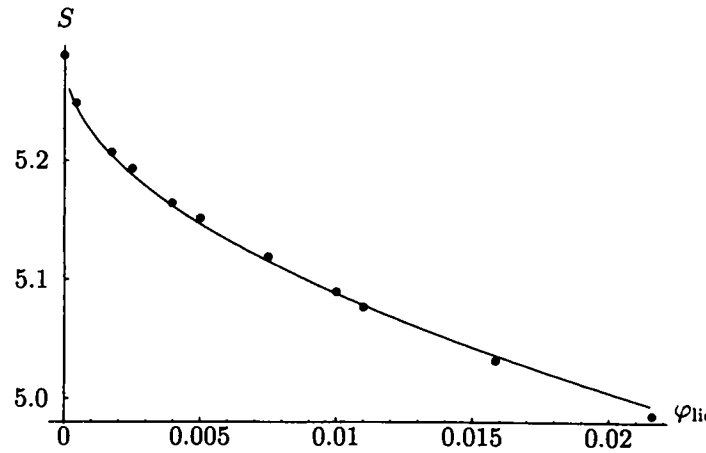

FIGURE 10. Surface energy per unit volume S for the WP structure consisting of unit volume cells, fitted for low liquid fraction φ_{liq} to the function $S = S_0 + S_1(\varphi_{\text{liq}})^{1/2}$ proposed by Weaire [1993], where S_0 is the dry foam energy. The second term is based on a consideration of the spreading of the Plateau borders along the cell edges. S_1 is approximately -2.0.

The f.c.c.–b.c.c. comparison is rendered less important by the intervention of the WP structure, which, as Figure 9 shows, remains lower in energy

up to at least $\varphi_{\text{liq}} = 11 \pm 1\%$. We have not pushed the calculation of its energy and detailed structure far beyond this point, because topological changes take place that are poorly described at the level of tesselation used.

4. INSTABILITIES AND TOPOLOGICAL CHANGES

As φ_{liq} is varied, an instability may develop that precipitates a structural change. Such a point will define the limit of stability of that structure, and is important in estimating the hysteretic effect to be expected in practice in transitions between foam structures.

For b.c.c. it has been argued [Weaire 1994] that the limit of stability, as φ_{liq} is increased, is the point at which the contact areas of what were originally quadrilateral faces are lost (see Figure 7). The argument is as follows. Consider the interaction between two bubbles in a foam structure to be crudely represented by Hooke's law springs. There are springs corresponding to first and second nearest neighbours, the hexagonal and quadrilateral contacts, respectively. All are in compression so long as there is bubble-bubble contact at the corresponding face.

The shear modulus $\frac{1}{2}(c_{11} - c_{12})$ has a negative contribution from the nearest neighbours, because their distances increase quadratically with the corresponding extensional shear along a cubic axis. Within such a model, the structure is stabilised by the second-nearest neighbour springs, whose contribution is positive, but vanishes discontinuously as contact is lost.

This is a classic argument in the metallurgical literature, often used by Zener [1948] and others in rationalising relative stability of metallic structures. The present context is probably a more reasonable one for its application, but it should nonetheless be noted that the model is not strictly correct. The energy is not an additive function of the distances between bubble centres. Moreover, Morse and Witten [1992] have shown that it does not vary quadratically as contacts are made. There is a logarithmic factor that implies that such springs become softer as they are switched off. We have, nevertheless, some faith in the heuristic value of the model.

We have carefully identified the point at which the second-nearest-neighbour contacts are lost in our calculations (see Figure 7), finding it to be $11\% \pm 0.5\%$. This is remarkably close to the previous rough estimate [Weaire 1994], and also close to the f.c.c.–b.c.c. crossover point in Figure 9. We have examined an extensional shear along a cubic axis and found a negative elastic constant (and hence an instability) for liquid fractions above 11%.

For f.c.c. the situation is reversed. The structure is stable for large φ_{liq} and becomes unstable as φ_{liq} is decreased. This must be so because there are eight-fold junctions of Plateau borders, and only fourfold junctions can be stable in the limit of dry foam. So far, this instability has completely eluded us, in various calculations of energy versus strain for supercells of f.c.c. Its location and mechanism remain an intriguing challenge to future work.

In the case of the WP structure, we believe that the first instability to be encountered is at $15 \pm 2\%$, but can only crudely estimate its position on the basis of present calculations.

5. IMPLICATIONS FOR EXPERIMENTS

Results for the relative energies of ordered structures such as those discussed above, have a bearing on current experimental investigations of monodisperse foams and emulsions.

Weaire and Phelan [1994c] have reported clear identification of a small fragment of the WP structure in a dry foam, but this isolated observation has not yet been pursued any further.

Monodisperse emulsions are the subject of active investigation by a number of groups; see [Weitz 1994], for example.

In either case, our present results tend towards the following predictions. Assuming that the sample is first prepared as a wet foam (or its equivalent for an emulsion), it should crystallise as a close-

packed phase, probably but not necessarily face-centered cubic. Upon extraction of the continuous phase to move towards the dry foam limit, there should be a sharp structural transition due to mechanical instability, resulting in the WP structure. Upon reversal of this procedure, the WP structure should remain stable up to a liquid fraction of approximately 15%.

6. CONCLUSIONS

Much remains to be done to complete the picture of structural transformations in wet foams that has been partially delineated here. In particular, accurate calculations of elastic constants will be most valuable. Semiquantitative models, which have already provided useful predictions, require further development and comparison with full computer calculations.

The foams and emulsions that are of commercial interest are polydisperse random structures. They present us with structural changes that are smoother when macroscopically averaged, and also more complex when identified at the local scale, than those considered here. They will require orders of magnitude more computing time and storage if we are to simulate them in detail, but this seems a reasonable prospect for the near future.

Previous work on the simulation of foams has relied on two-dimensional models (for a review in the context of rheology, see [Weaire and Fortes 1995]). This offered qualitative insights, but a fully realistic theory requires the full three-dimensional simulations described here.

In the short term, the goal should be accurate static structures of wet foams for a few hundred cells, and hence an accurate simulation of those properties that can be described in the quasistatic approximation. Two-dimensional simulations have taught us that the study of dry foams can be quite misleading, since quite small Plateau borders can have large effects. Beyond this goal, a variety of non-equilibrium time-dependent effects will remain to be addressed. These effects relate to the rheological response at finite strain rates and also to drainage, the process by which a freshly made wet foam comes into equilibirum under gravity. Finally the familiar collapse of a foam, due to the bursting of its bubbles, also remains to be understood.

These problems bring into play a nexus of questions of stability, surface chemistry and physics, and fluid dynamics: they will not be easily solved in any detail. The Surface Evolver, or some adaption of it, will provide only one of the necessary ingredients. Nevertheless it has already enabled us to make rapid progess on three-dimensional structures that seemed unattainable until recently, and should continue to throw up interesting and unexpected results.

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SOFTWARE AVAILABILITY

The Surface Evolver and Geomview are available via anonymous ftp from the server geom.umn.edu, in directory pub/software.

For information about the Voronoi Cell Solver, contact John Sullivan at sullivan@geom.umn.edu.

Robert Phelan, Physics Department, Trinity College, Dublin 2, Ireland (rphelan@alice.phy.tcd.ie)

Denis Weaire, Physics Department, Trinity College, Dublin 2, Ireland (dweaire@tcd.ie)

Kenneth Brakke, The Geometry Center, 1300 South Second Street, suite 500, Minneapolis, MN 55454, and Susquehanna University, Selinsgrove, PA 17870 (brakke@geom.umn.edu)

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