

## Rigidity Loss Transition in a Disordered 2D Froth

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Disordered two-dimensional soap froth has been simulated, with a gas fraction  $\phi$  less than unity. Upon decreasing  $\phi$ , it is found that the system loses its rigidity at  $\phi_c = 0.84$ . This value is identified with the dense random packing of hard disks. Results are presented for the variation of yield stress and shear modulus, the latter being tentatively related to elastic-network theory.

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In this Letter we present the first results of simulations of the two-dimensional soap froth with gas fraction  $\phi$  less than unity. We explore the nature of the transition by which this system loses its rigidity as  $\phi$  is decreased. The question is simply: How does foam (which acts as an elastic solid under low stress) fall apart, as it must, if we steadily increase the liquid fraction?

A remarkably simple scenario emerges for this transition, linking it with two classic computational problems—random hard-disk packings and the rigidity of random elastic networks.

Until now, theories of disordered 2D froth structure and properties had the limited objective of understanding the case defined by  $\phi = 1$ , in which the cell walls (curved lines) meet at point vertices, rather than joining liquid Plateau borders, which are the consequence of  $\phi < 1$ . Very recently, various discrepancies<sup>1,2</sup> in the comparison of experimental results with theoretical expectations have forced these Plateau borders upon our attention, and it has been realized that even borders of quite modest size can be significant in their effects.<sup>3,4</sup> The theorems<sup>4</sup> and procedures which we used to demonstrate this are of limited applicability, being confined to small, three-sided Plateau borders. If  $\phi$  is decreased to values considerably less than unity, we encounter borders with ever greater numbers of sides. These can only be analyzed by recourse to a fresh approach to the simulation problem, which takes account of them. We have developed the necessary program which can equilibrate large samples in a manner broadly similar to that of the previous work.<sup>5</sup>

With Plateau borders incorporated, the equilibrium conditions are as follows. The cell walls (which are still not given any finite width) join the Plateau borders smoothly; the two border edges have a common tangent with the adjoining cell wall. The border edges themselves have radii of curvature  $r$  which satisfy  $\Delta p = \sigma r^{-1}$ , where  $\sigma$  is surface tension, while the cell walls satisfy  $\Delta p = 2\sigma r^{-1}$  as before.<sup>5</sup> The cell areas are fixed and hence the cell pressures are variables, but the pressure is put equal to a common value throughout the Plateau borders, which is adjusted to achieve a specified  $\phi$ . Both liquid and gas are treated as incompressible.

The technical details of the program cannot be detailed here: Its success is self-evident from Fig. 1. This sample of 100 cells was created by the Voronoi procedure,<sup>5</sup> with periodic boundary conditions.

A sequence of structures was generated by reducing  $\phi$  by intervals of 0.01, equilibrating the structure for each successive value. The starting structure at  $\phi = 1$  was characterized by  $\mu_2 = 1.44$  and  $\mu_2^4 = 0.15$ . These are respectively the second moment of the distribution of numbers of cell sides and that of cell area normalized by division by the square root of mean cell area. The degree of disorder represented by this choice of structure is roughly typical of a mature 2D froth.<sup>6</sup>

Such a simulation allows us to address for the first time the loss of rigidity which must occur as  $\phi$  is decreased, since the system must eventually consist of isolated bubbles.

How and when does this happen? Only an *ordered* hexagonal array of cells has previously been studied.<sup>7</sup> In this case, there is a sudden collapse at  $\phi = 0.9069$ , with no change in the shear elastic modulus up to that point. This has very limited bearing on the behavior of the typical disordered froth.

In various preliminary runs, we found it impossible to equilibrate structures below  $\phi = 0.84$  and percolation of the Plateau borders was evident as the cause. Hence we estimate this to be the critical value  $\phi_c$  for the disordered system. This critical density is indeed distinct from the value for the ordered froth, but it has a simple, and related, significance: it is the packing density for random hard disks. As Bideau and Troadec<sup>8</sup> have shown, there is a wide range of random mixtures of hard disks for which the packing fraction  $0.84 \pm 0.1$  is obtained.

Such random hard-disk packings have coordination number close to  $Z = 4$ . Bideau and Troadec observed a slightly lower figure but Weaire<sup>9</sup> has argued that this is largely attributable to the effects of rigid wall boundary conditions. The identification of the limiting 2D froth with the dense random hard-disk packing is reinforced by the observation that the value  $Z = 4$  is indeed approached as  $\phi \rightarrow \phi_c$ , as shown in Fig. 2. Note that  $Z$  is defined as the average number of neighbors with which a cell makes contact. As triangular Plateau borders

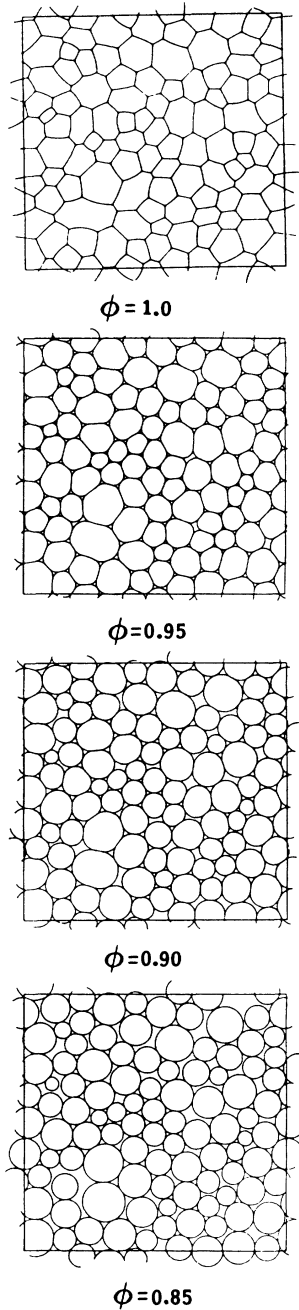


FIG. 1. A sequence of disordered simulated froth structures, with decreasing gas fraction  $\phi$ .

coalesce to form many-sided borders,  $Z$  drops from its initial value  $Z=6$ , which is fixed by Euler's theorem.

Figure 2 also shows the variation of energy, shear modulus, and yield stress.<sup>10</sup> The dimensionless quantities shown are defined in terms of the mean gas cell area  $\bar{A}$ , gas fraction  $\phi$ , and surface-tension parameter (energy per unit-cell edge length)  $\sigma$  by

$$E^* = \bar{A}^{1/2} \phi^{-1} \sigma^{-1} E. \tag{1}$$

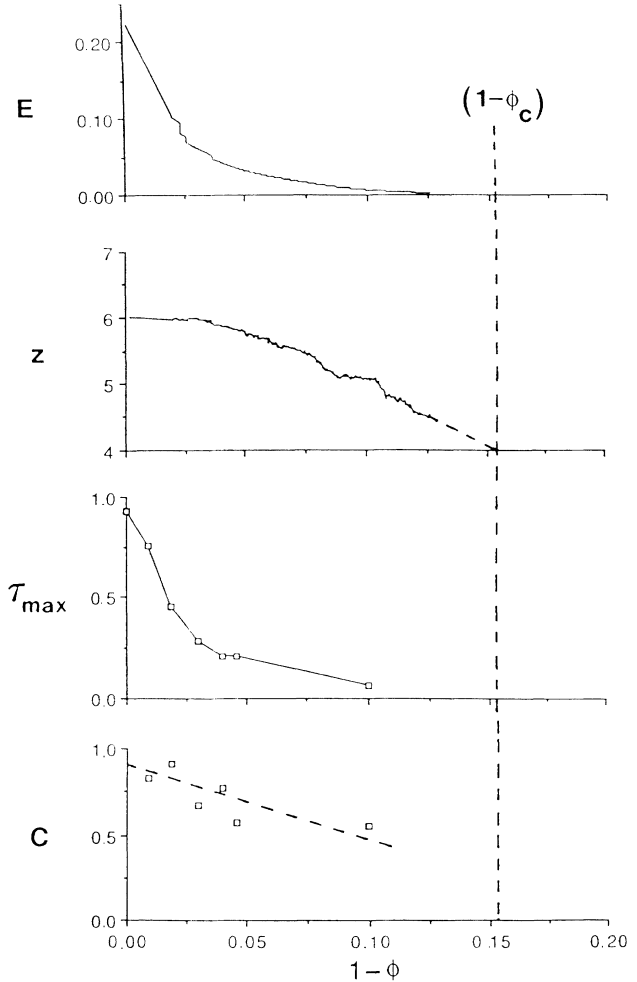


FIG. 2. Calculated variation of structural and mechanical parameters with gas fraction  $\phi$ , for the structure shown in Fig. 1. From the top: energy  $E^*$ , coordination number  $Z$ , yield stress  $\tau_{max}^*$ , and shear modulus  $C^*$ , as defined in the text.

The shear modulus and yield stress are derived from calculations in which an increasing extensional shear is imposed by a change of boundary conditions.<sup>10</sup> For these quantities there are some difficulties in following the variation into the critical region. In part this is due to the small size of our sample: We intend to undertake studies of much larger ones in the future.

Bearing in mind the variation of  $Z$ , this transition is highly reminiscent of that recently investigated for elastic networks, in which elastic springs are randomly severed until there is a loss of rigidity (rigidity percolation).<sup>11</sup> Loosely speaking, the contacts between the disks act as elastic springs, at least close to  $\phi_c$ . These springs act only under compression so that they are effectively severed as a contact is lost, upon decreasing  $\phi$ . Note that the critical value  $Z=4$  recurs in the elastic-network model, as the value at which the number of constraints defined by zero deformation of the springs equals the

number of degrees of freedom. This strongly suggests that the prediction of the effective-medium theory,<sup>11</sup> which is also successful for elastic networks, that the shear constant goes to zero *linearly*, should describe this system well in the critical region (although not necessarily at the transition itself).

The yield stress<sup>10</sup>  $\tau_{\max}$  shows a dramatic initial drop with decreasing  $\phi$ , reinforcing our assertion that Plateau border effects can be very large, even when the borders themselves are small. Indeed a simple analysis which treats the elastic modulus as constant and estimates the change in yield strain and hence stress, due to Plateau borders, suggests that the derivative of  $\tau_{\max}$  is infinite at  $\phi = 1$ .

Note that although we have used the word *percolation* from time to time, the rigidity loss transition is *not* provoked by the growth of a percolating Plateau border, as one might suspect. The analogy with elastic-network models is particularly valuable in understanding this. One does not have to cut such a network in two in order to cause it to lose its rigidity.

Such results present a challenge to further experimentation and perhaps an opportunity to review some existing data to determine, for example,  $Z$  as a function of  $\phi$ . Recent investigations include not only the prototypical 2D soap froth,<sup>2</sup> but also analogous structures in lipid monolayers.<sup>12,13</sup> Photographs of the latter system<sup>12,13</sup> provide a particularly interesting comparison with our Fig. 1. It may even be possible to perform two-dimensional rheological measurements in this case, an objective which seems unattainable for the soap froth itself.

In conclusion, we have established the broad features of this type of transition for the first time. Its intrinsic interest is reinforced by its relation to hard-disk packings and elastic networks. Ultimately it can offer us insights into the practical problem of three-dimensional foam rheology. Indeed, the entire scenario which we have established should carry over to the three-dimensional case, with minor modifications, such as critical coordination number  $Z = 6$ .

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