Cell growth in a pinned soap froth

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Experimental kinetic data on soap froth cell growth in a random array of fixed pinning centers is reported. The scaling of our two-dimensional data is analyzed and compared with pinning-free growth. The obtained data agree fairly well with a proposed cell growth law. We also discuss the relationship between the pin concentration and the final cell size.

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INTRODUCTION

Soap froth studies for ''free'' grain boundaries (i.e., with no pinning centers), are well documented in the scientific literature [1–3]. Krichevsky and Stavans' results for the foam growth kinetics and the corresponding topological reactions in a squared array of pinning centers was the first study on pinned foams [4]. Also, soap froth behavior in a matrix of quenched impurities has been used to study the kinetics and the area fraction dependence on the pinning effect in disordered arrays [5] and, furthermore, mimic computer simulations and experimental results, including the case of elongated pinning centers, have recently appeared [6]. An excellent introduction to the physics of foams appeared recently in the literature [7]. Soap froth behavior has a close analogy with other phenomena, among them, cell growth (grain growth) in polycrystalline materials [8]. Both phenomena are characterized by possessing topological constraints (to fill the space) and a high surface/volume ratio in a metastable state. It is usual to consider both cell size escalations as driven by capillary (surface-tension) forces.

Discussion of this pinning phenomenon appears to be mainly linked to the materials' metallurgical behavior, due to the importance of grain size in the properties of the finished products. Pinning of grain boundaries during grain growth by second-phase particles is a well-known phenomenon in metallic and ceramic polycrystalline samples. In this context, it is called Zener drag [9]. Apart from its theoretical value, its practical utility ranges from the obtaining of very large grain sizes in electrical steel sheets to the engineering of nanostructured materials [10]. Since Zener's work, a large amount of literature has considered this phenomenon, improving its treatment in both two and three dimensions. Recent papers show the contemporary interest in this matter [11–14].

In this paper, we will treat the cell evolution of two-dimensional foams in a random array of fixed pinning centers, giving experimental results and discussing them in a phenomenological framework. As noted, in previous works the kinetics has been described [4–6] but no mention has been made of the scaling involved in this phenomenon. Furthermore, there are no theoretical clues involving both the kinetics and the dependence of final cell size on the fraction of precipitates. These issues are covered in the present paper.

We will present our results in terms of $N(t)$, the number of cells at time t, but it is easy to switch them to (mean) grain size diameter D, which is usual in the materials' literature. Simply note that $N \propto 1/D^2$.

EXPERIMENT SETUP AND RESULTS

As described elsewhere [5,6], a two-dimensional soap froth is allowed to grow between two glass plates 1.5 mm apart. The plates are $20 \times 20\ \mathrm{cm}^2$ each. Within the plates we insert fixed circular pinning centers, covering different area fractions and pin radii as described in each case. Here we report our results for six experimental runs (see Table I). Stochastic allotment is a general characteristic of all these pin distributions, unlike the regular net used by Krichevsky and Stavans [4] in their pinning experiments. Into these frames we blow a soap mixture of detergent and glycerin forming a primary pattern with an initial cell size smaller than the interpinning distance. We follow the foam growth kinetics by taking snapshots at predetermined times. These images are digitized and the time evolution of the pattern is studied through a quantitative analysis of these photographs. The typical time for completion of one series of experiments is about a couple of months.

Figure 1 shows the experimental kinetics for one of our samples. The foam behavior during the development of the structure shows capillary-driven growth and topological reactions as described in the literature [2]. Also, our results show the rapid decrement in the number of cells at the initial

TABLE I. Experimental settings for the six reported experiments. f, area fraction of pins; r, pin radius.

Sample	$10^3 f$	r (mm)
A	10.5	1.5
B	6.3	1.1
C	22.0	1.8
D	6.6	1.1
E	9.2	1.1
F	35.0	2.2

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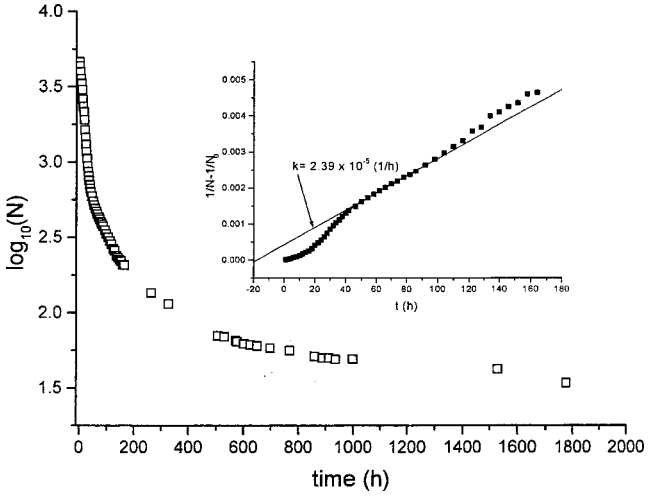

FIG. 1. Growth kinetics. N, number of cells; N_0, initial number of cells. Sample A.

stages of the process and at the final approach to stagnation [5].

In order to compare our results with the theoretical predictions, we must scale the experimental measurements. At first sight, when $t \to 0$, a pinned system must behave as a free foam and, therefore, it must follow the well-known free grain growth equation, $dD/dt \propto 1/D$. Its solution is commonly named the parabolic law. In terms of the number of cells, it can be written as

$$\frac{dN}{dt} = -kN^2. \tag{1}$$

In this equation, k is a constant including the boundary energy (surface tension), the mobility, and a geometrical factor. Within this hypothesis, it would be possible to use Eq. (1) to calculate the constant k. Experimentally, however, even free foams do not follow the parabolic law in the first stages of its development [2,7]. Instead, they remain a finite period of time in a quasiordered state, and after that they reach the scaled state. This last stage is characterized by a structural disorder. These two regimes (and a final stagnated regime) also appear in pinned grain growth. The initial order appears evident in the snapshot showing the pattern after 1 h of development in Fig. 2(a). Later on, the order disappears [Fig. 2(b)]. In their work on vertex simulated grain growth, Weygand *et al.* also note this effect [11,15].

In a straightforward manner, the solution of Eq. (1) is

$$\frac{1}{N} - \frac{1}{N_0} = kt, \tag{2}$$

with N_0 the initial number of cells.

As suggested by Eq. (2), the inset in Fig. 1 shows the linearized data obtained from the initial points in this experiment. As shown, our results show a strong deviation from linearity. After some time, however, a stage appears showing a constant slope, as indicated in the figure. This stage corresponds to the scaled state where the parabolic law is valid. Using this slope, we obtain a k value of 2.38×10^{-5}(1/h).

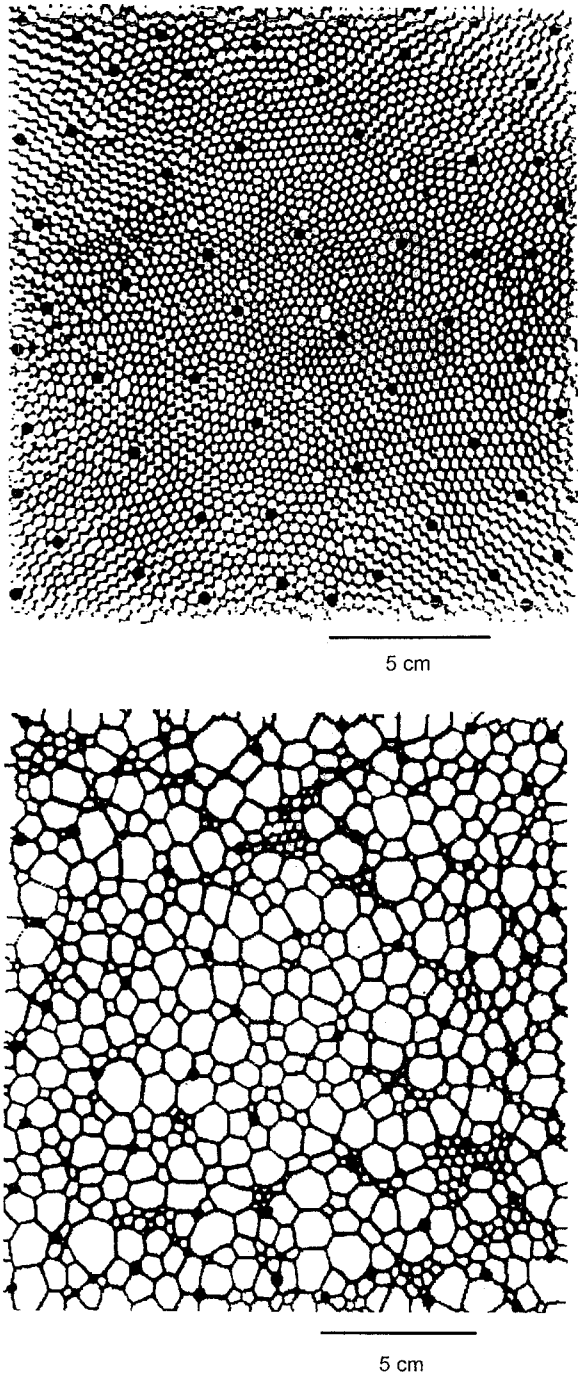

FIG. 2. Typical snapshots in pattern development: (a) 1 h and (b) 60 h after start. Marks correspond to 5 cm.

This value was used to adjust our experimental results to the nondimensional time, $\tau = N_{\min} kt$, and the scaled number of cells $n = N/N_{\min}$. We call $N_{\min}$ the minimum (stagnated) cell number. These scaled variables are plotted in Fig. 3 for the six experimental runs we are reporting in this paper.

We include our estimated error bars. Errors appear from different sources; we use a finite system and, therefore, there are some inevitable boundary effects (theoretical or simulation models use either infinite or periodic boundaries). On the other hand, film breaking could contribute to the uncertainty in the measured area and, finally, it is impossible to

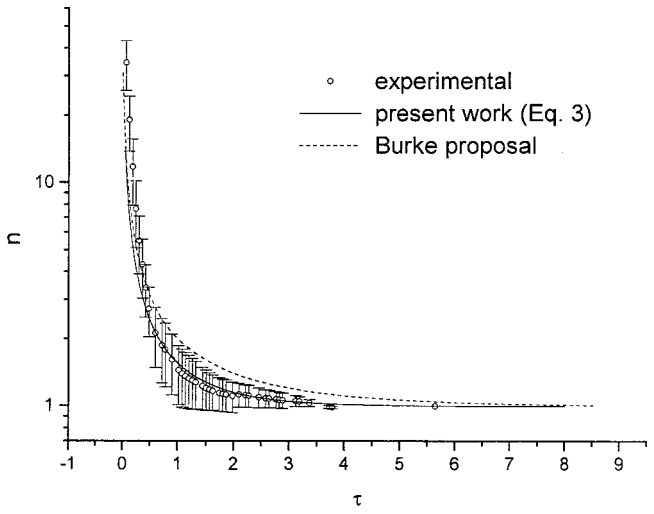

FIG. 3. Experimental results and theoretical estimates. n, scaled number of cells vs τ, scaled time.

ensure that the initial state of our different experimental runs is identical. As is easily recognized, the experimental uncertainty is bigger in the zone of rapid decline of the curve.

DISCUSSION

The product of the right-hand side of Eq. (1) can be interpreted as the ''source'' of the kinetic phenomenon for free growth. However, in the case of a Zener pinned grain structure, it is possible to argue that there are two types of grains; some of them grow freely (far away from the obstacles) and others, say $(N-N^*)$, are affected by the pinning effect of the anchor centers. Therefore, in the basic equation for the pinned growth, we could replace the N^2 factor on the right-hand side of Eq. (1) with the product of the quantity of these two types of grains, N and $(N-N^*)$ in number. Within this ansatz, in a natural manner, we further assume that the correction number N^* is proportional to N_p, the number of pinning centers in the system. In this way, we propose that the rate of decrease for the number of grains in a Zener pinned structure is given by

$$\frac{dN}{dt} = -kN(N-\alpha N_p), \tag{3}$$

with α a numerical constant. This equation appeared, in the study of nanostructured materials, in treating grain-boundary restrained growth due to solute drag [16]. Also, it is applied to the study of population-restricted growth (the so-called ''logistic growth'') [17]. The solution to this equation is plotted (with scaled variables) in Fig. 3, showing a fair agreement with the experimental results. In a different way, Burke [18] considered the pin braking effect by introducing in the parabolic law a constant, negative term $\propto A/D_{\max}$, $D_{\max}$ being the maximum (final) grain size. The results of this last approach are also displayed in Fig. 3, showing a poorer fit to the experimental data.

A careful inspection of the reported data for the initial times, however, reveals a difference between the predictions of the theoretical models and the experimental results. This is not surprising; neither model includes the initial quasiordered stage.

In the stagnated state, the cellular system has the minimum number of domains, $N_{\min}=\alpha N_p$. Let us call f the fraction of pinning centers (measured as total pin area/total area). In the present two-dimensional approach the f dependence of the final grain size is easily obtained. By using the already written relationship for the minimum number of domains in the stagnated condition, it is easily shown that the final grain diameter D, scaled by the precipitate radius r, is given by

$$\frac{D}{r} = \frac{2}{\sqrt{\alpha f}}, \tag{4}$$

This last equation has been proposed on experimental and simulation grounds [5,6,19]. Also, it has been shown that this last result stems from an analytical model considering Friedel-type statistics [20]. It represents an answer (in two dimensions) to the question suggested by the original Zener insight relative to the dependence of the final grain size on the fraction of precipitates. Let us stress that Eq. (4) follows from Eq. (3) and therefore, its validity may justify the used ansatz. On the other hand, Burke's equation does not consider in its formulation the fraction of precipitates and, therefore, it is not possible to extract any information about this parameter.

Furthermore, our results show that it is possible to estimate the α value in the final pinned condition. By using the experimental result (see Ref. [5]),

$$\frac{D}{r} = \frac{1.6}{f^{0.55}}, \tag{5}$$

we obtain

$$\alpha = \frac{f^{0.1}}{0.64}. \tag{6}$$

This weak f dependence for the α value may or may not be introduced by an artifact in the former experimental data [5], as demonstrated by the fact that Eqs. (5) and (6) show a small disagreement with Eq. (4). Whatever the cause, this effect is shown in Fig. 4. Also, we directly measure the α parameter from the final structures. These experimental results are also shown in the figure. The basic fact is that, according to our results, $\alpha = 1.0 \pm 0.1$ within the concentration range we employ. In simple terms, there is one pin per final cell or grain.

An opposite view of this problem is to assume, *ab initio*, that there is one pin per cell in the final stagnated state of the foam. This assumption can be justified by arguing that the pin lattice induces a scaling on the foam behavior. With this hypothesis the $f^{-1/2}$ dependence of the final cell size follows in a trivial manner. Obviously, the kinetic behavior does not appear in this view.

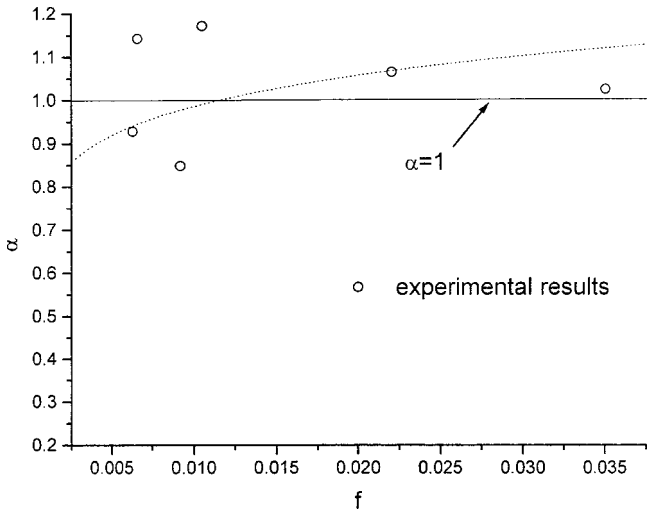

FIG. 4. α parameter vs f, area fraction of pins. Dotted line, Eq. (6). Circles, experimental results. Also, the estimate $\alpha=1$ is shown.

The present results extend the findings of previous works [4–6]. Although in these papers the kinetics have already been described, they concentrate on the study of the topological reactions in ordered arrays [4], the final stagnated state [5], and the comparison of mimic simulations and experimental settings extending the study to nonrounded pinning centers [6]. In the last two cases, use is made of a random array of pinning centers. Here, we present results of several experimental runs, concentrating on the evolving kinetics of the phenomenon, establishing the scaling, and proposing a descriptive equation.

In summary, we report results for the time evolution of the cell sizes in two-dimensional soap froths constrained by fixed pins. They show three growth regimes: the quasiordered state, the quasifree (parabolic) regime, and the final braking state. We develop a model that gives fair account of the experimental results for the last two stages, and we discuss the implications of this assumption on the f dependence of the final cell size. In addition, we show that in these systems, there is one pin per cell in the stagnated state.

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