Pinned state in a two-dimensional soap froth

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Experimental results of the pinning state of a two-dimensional soap froth are herein reported. In the current literature there are some results concerning the pinning of grain boundaries by a periodic array of pinning centers. The present work details with a random array of pinning centers, a more realistic situation also related to the metallurgical problem of grain boundary pinning in polycrystalline materials (Zener pinning). Current trends in the theoretical comprehension of this phenomenon are analyzed. Our results show that the cell size scales as $f^{-0.5}$ in the stagnated state, the area fraction of pinning centers being *f*. [S1063-651X(97)11904-3]

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INTRODUCTION

The pinning of grain boundary displacements by secondphase particles is a phenomenon which has both theoretical and practical interest. The first theoretical model on this matter is owed to Zener, and is called Zener pinning or Zener drag. It helps develop materials of ultrafine grain size (nanomaterials) and also, through the mechanism of secondary recrystallization, to obtain materials with a large grain size, such as sheets of electrical steels (for a review of Zener pinning's structural effects on metals and ceramics, see Ref. [1]). Furthermore, there is a close analogy in the behavior of grain boundaries and soap films. Soap froth behavior has been reviewed by Weaire and Rivier [2] and recently by Stavans [3]. As an example, the recent works of Levitan [4] and Jiang, Monbach, and Glazier [5] consider the coarsening of a single cell in a two-dimensional soap froth, a phenomenon analogous to secondary recrystallization in polycrystalline materials. Also, in normal grain growth, the analogy is well established. Obviously, the time scales are different as they are controlled by different diffusion mechanisms, but both normal grain and soap froth growth show a similar dependence between cell size and time. In fact, for both phenomena, the cell size scales as $t^{1/2}$ [6]. If we consider materials with a dispersion of second-phase particles (impurities), these dispersion particles break the grain boundary movements, resulting in a pinned grain structure. There is a considerable number of papers dealing with this problem (see [7] and references therein). The analogous problem of the pinning of soap films is apparently far less frequent in the literature, with the sole exception of the results yielded by the Krichevsky and Stavans study [8]. In this, the effect of a square array of pinning centers forming a two-dimensional network was experimentally analyzed. In the present work we will report the results obtained with a two-dimensional array of randomly positioned pinning centers.

EXPERIMENTAL PROCEDURE

Two-dimensional soap froth systems were built bounded by two glass plates, 20×20 cm² each, separated by a 1.5-mm gap. Between these plates, cylindrical pins with a diameter varying between 0.20 and 0.44 cm were randomly arrayed. In these frames, a soap mixture of detergent and glycerine that formed a two-dimensional pattern with an initial cell size smaller than the interpinning distance was blown.

The kinetics of these patterns (with a typical time of some months) were followed by taking snapshots at various times. These photographic images were digitalized for the subsequent analysis of the relevant topological parameters. A total of ten runs are reported in the present paper. Typically, each one consists of a series of snapshots ranging between 9 and 20 photographs for each pin configuration, that is, a determined concentration of pinning centers.



FIG. 1. Four snapshots of a typical growing sequence. (a) Initial state, (b) 206 h, (c) 673 h, and (d) 2328 h. The area fraction of pins is 0.022.

<u>55</u> 4475



FIG. 2. Mean area vs time plots for different fractions of pinning centers, as indicated in the figure.

RESULTS AND DISCUSSION

The series of photographs in Fig. 1 display the development of the cell structure as it evolves towards the stagnated state. At early times, the cell structure behaves as free (that is, nonpinned) soap froth, and, as time elapses, it shows a gradual transition to a stagnated state. These facts can be fully appreciated in Fig. 2 which shows graphs of the mean area evolution against time for the different experimental settings. In this figure it appears evident that the mean area (and consequently the cell size) arrives at a stagnation state asymptotically. This result is present in all the experiments we ran, and conforms to the expected common belief. More interesting is the behavior of the limiting grain size. Current theories [1,7] disagree as to the dependence of the final grain size on the concentration of pinning centers. A recent discussion on this matter can be found in a paper by Liu and Patterson [9].

We will define D, the final stagnated mean cell diameter, as the diameter of the equivalent circle of the same area. Our results validate an expression of the type

$$D = k \frac{r}{f^m},\tag{1}$$

as shown in Fig. 3, in a logarithmic plot, r being the radius of the pinning centers, f the (area) fraction of pins, k a numerical constant, and m an adimensional numerical exponent. In this case, $k=1.6\pm0.2$ and $m=0.55\pm0.02$ were obtained.

The outcome of this experiment is very close to the predicted value of 0.5, obtained by Monte Carlo simulations and by an analytical model based on the Friedel-Labusch approach, both for two-dimensional systems. The small difference between our measured value and the predicted value can be attributed to the finite size of the sample. Also, film breaking contributes to the uncertainty of the measured area. This is particularly evident when cells appear which are abnormally larger than the mean cell. However, we estimate that this effect is within the error included in the symbols



FIG. 3. Logarithmic plot for the normalized stagnated cell size D/r against *f*, the area fraction of pinning centers.

of Fig. 2. Theoretical models assume an infinite array of pinning centers, and Monte Carlo simulations generally use periodic boundary conditions. The Friedel-Labusch approach, used by Hazzledine and Oldershaw [10], models the grain boundary-particle interaction in a way similar to the string model for dislocation pinning. Monte Carlo simulations of Srolovitz and co-workers [11] model the grain growth as a Potts model with some fixed lattice sites (pinning obstacles). In conjunction with their analytical approach, Hazzledine and Oldershaw [10] also tackle the problem with simulation techniques, finding the same 0.5 exponent. On the contrary, the original Zener model [12] and the dimple analytical model (that considers the particle as making a "dimple" of minimal surface area on a moving grain boundary), first proposed for three-dimensional pinning by Hellman and Hillert [13], and later applied to twodimensional pinning problems [7], show a 1/f dependence. The recently proposed stereological approach of Liu and Patterson [9] also predicts a 1/f dependence.

Our results are comparable with those obtained by Krichensky and Stavans [8] although they do not stress the f dependence of the final stagnated grain size. Their two experimental measurements fit a $f^{-0.55}$ dependence precisely.

The present experimental results seem to confirm, at least in two-dimensional soap froths, that the grains mold themselves according to the preexisting pattern of pinning particles instead of the grain boundaries sweeping the material "freely" until they become anchored by the pinning particles. This distinction is the basic difference in behavior assumed for the above mentioned analytical models. Furthermore, the coincidence between our results and the computermodeled results seems to corroborate that the physical assumptions lying behind the modeling are essentially correct.

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