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Fractal Growth of Sodium Tartrate Crystals

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Abstract. Fractal pattern formation was investigated in the "tree-like" crystals of sodium tartrate $Na_2C_4H_4O_6 \cdot 2H_2O$ grown from aqueous solution by slow evaporation. The tree-like whiskers evolved by creep-growth inside the test-tube without any influence of surface tension and growing crystals between the solution and the wall surface of the vessel. The patterns are not like crystalline one but fractals. The fractal dimension increases from 1.5 to 1.9 as its growth evolves.It suggests that the fractal patterns of the morphology in the growth of sodium tartrate crystals may be effected not by the crystalline habits but by the growth environment.

1. Introduction

Most of the recent papers about fractal pattern formation have been concerned with the two-dimensional DLA (diffusion limited aggregation) model proposed by Witten and Sander. This model of pattern formation by aggregation of a cluster in the diffusion field treats simple physical conditions without any interaction between the wall surface of a vessel containing the solution and the growing and the crystalline nature of grown crystals. We mentioned the connections of DLA with pattern formation phenomena like dendritic crystal growth. On the other hand, the pattern formation of crystal growth depends on both the crystalline habits and environment of crystal growth. Crystal growth of snow, especially, have attracted great interest. Relations of theoretical studies and experimental studies of patterns formed by growth processes have attracted more attentions in recent years. There are few experimental studies of such crystal growth. In the present paper we report an experimental study of the DLA-type growth pattern of sodium tartrare crystals.

2. Experiments

A saturated solution of sodium tartrate of guarantee regent about 3 to 10 cc was poured into test-tubes (2.5 cm diameter and 15 cm long). These vessels were put on a table vertically, and the solution was left to dry at room temperature. The vessels were covered with gauze in order to avoid the contamination by dust etc., and at the same time to regulate the rate of evaporation of aqueous solution. For the control of humidity, the growth vessels were placed in an apparatus of crystal growth with silica gel. The relative humidity in our experimental room was 60–80%, and the one in the crystal growth apparatus (with silica gel) 5–20%. The fluctuation of temperature (15–20°C in our experiment) did not influence the growth rate, but it influenced the growth patterns. The humidity exerted a strong influence both on the growth patterns and on the growth rate. Usually, whiskers began to grow up from seed at the interface between solution and air in the vessel. The tree-like whiskers continued to grow in a few days, Fig. 1. Needle-like crystals were grown in mother solution by slow evaporation. The whiskers having hollows were raised or budded

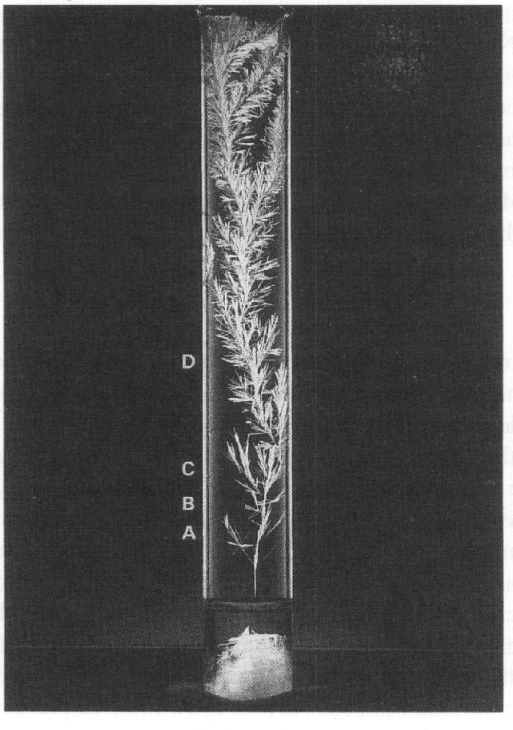

Fig. 1. Tree-like growth of sodium tartrate whiskers.

at the interface between the solution and air. It was found by a microscopic observation that the tree-like whiskers have hollows.

The hollows supply crystals with the solution to creep inside the capillaries. Moreover, the tree-like crystals grown on the wall surface inside the vessel were aggregated as single or twin crystals. This was confirmed by the crossed polarized microscope and X-ray diffraction. The growing tree-like whiskers were observed with crossed polarizers at magnification of 1×. When the solution was dropped on a horizontal glass plate and dried at room temperature and humidity, the patterns grew into both DLA like crystals and homogeneously oriented crystals. Further, three-dimensional fractal crystals are grown from an edge of the vessel. It is noted that the crystals are grown by creeping on the vertical wall surface inside the vessel.

3. Analysis

We analyzed the DLA like and the homogeneous patterns quantitatively as follow. We enclosed the pattern in a square of side length L and divide the square into $(L/l)^2$ squares of side l. We counted the number N of small squares which hits the pattern and plotted log S against log l, where S is the total mass $S = l^2 \times N$. The fractal dimension D_f is obtained from the relation

$$S \propto l^{D_f}$$

The value of the fractal dimension D_f was thus obtained for some tree-like crystals, as shown in Fig. 2. We found the D_f increases from 1.5 to 1.9 as the length of growing crystal increase. When the height of grown crystal is less than a half height of the vessel, the value of D_f gives 1.5–1.7 as expected for DLA-like crystal growth. The

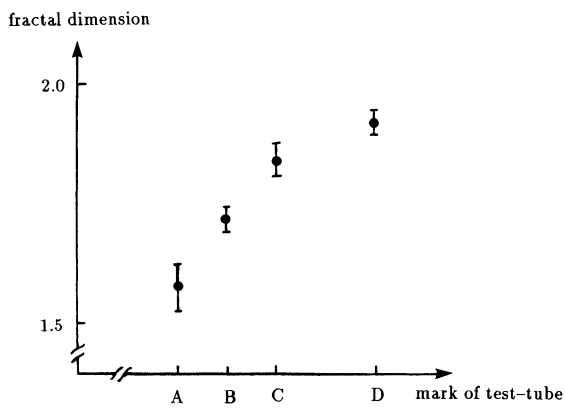

Fig. 2. Plot of the estimated value for fractal dimension versus growth evolves.

value of D_f then increases when the height increases. When the growth height is over the half of the vessel height, the value of D_f gives 1.9 as expected for a homogeneous pattern like a plane ($D_f = 2$).

4. Results

The force by which the solution is driven to the tip passing through inside the whisker tube may be caused by the extruding pressure by the capillary effect rather than by the surface tension between the wall surface of a vessel and grown crystals. This was confirmed by observation of the movements of dyed solution into the whisker tube, and by observation of the growth of the three-dimensional crystals. The growth orientation of branched whiskers did neither depend on the sodium tartrate crystalline habits nor was influenced by the gravitation, because the patterns grew up with vapor pressure decrease in vessel. The tip of the whiskers thins down due to increase of branched whiskers splitted from the whisker and spread over the surface of the vessel. The branched whiskers grown by the extruding pressure is DLA like formation. The DLA like whiskers continued to grow for a few days but homogeneous whiskers continued to grow for few hours or few minutes when the solution in the tip of the whisker tube was depleted. The homogeneous whiskers may be caused by the surface tension because the growth rate increases the depletion of solution. The whiskers growing in the glass box, under no influence of curvature of wall, was DLA like as crystals grew in test-tube.

5. Conclusion

In summary, we have shown two kinds of sodium tartrate whiskers. One is fractal pattern ($D_f = 1.5$–1.7) which grew up under the extruding pressure inside the whisker tube and the other is homogeneous pattern ($D_f = 1.9$) grew up by the surface tension on the glass wall in the saturation environment. These results suggest that the fractal patterns of the morphology of sodium tartrate crystals growth may depend not on the crystalline habits but on the growth environment.

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