Science on Form: Proceedings of the First International Symposium for Science on Form, General Editor: S. Ishizaka, Editors: Y. Kato, R. Takaki, and J. Toriwaki, 95–101.

Lattice Models for Solidification and Aggregation

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Keywords: Cellular Automata, Solidification, Aggregation

Simple models with both discrete and continuous variables at every site of a lattice are used to investigate solidification and aggregation in two dimensions. These models display a rich variety of macroscopic forms growing from small seeds. Transitions between macroscopic forms are seen as parameters in the model are varied, and these transitions may be compared with those seen in experiments.

Crystal growth is an excellent example of a physical process that is microscopically very simple (attachment of molecules onto a solid), but that displays a beautiful variety of macroscopic forms. Many local features are predicted from continuum theory,[1] but global features may be analytically inaccessible. For this reason, computer simulation of idealized models for growth processes has become an indispensable tool in studying solidification.[2,3] Here we present a new class of models that represent solidification by sites on a lattice changing from zero to one according to a local deterministic rule. The strategy is to begin with very simple models that contain few physical elements, and then to add physical elements gradually, with the goal of finding those aspects responsible for particular features of growth. The models display both local and global features that may be compared to the results from solidification experiments.

The simplest deterministic lattice model for solidification is a two dimensional cellular automaton with two states per site to denote presence or absence of solid, and a nearest neighbor transition rule. Further, we will consider only rules which have the property that a site value of one remains one (solidification only; no melting or sublimation). An additional constraint for the rules considered here is that they depend on neighboring site values only through their sum:

$$a_i^{t+1} = f(\sigma_i^t) \qquad \text{with} \qquad \sigma_i^t = \sum_{\delta \,\varepsilon\, Nbrhd.} a_{i+\delta}^t \tag{1}$$

The domain of f ranges from zero to the number of neighbors; f takes on values of one or zero.

These rules display four types of behavior for growth from small seeds:* *(i)* No growth at all; This certainly happens for the rule that maps all values of σ to zero. *(ii)* growth into a plate structure with the shape of the plate reflecting the

lattice structure; an example is $f(\sigma) = 1$ when $\sigma > 0$. *(iii)* growth of dendritic structure, with sidebranches growing along lattice directions at unit velocity; this type of rule is obtained by adding growth inhibition to the previous rule, *e.g.* with $f(\sigma) = 1$ when $\sigma = 1$. Physically, growth inhibition occurs because of the combined effects of surface tension and radiation of heat of solidification. *(iv)* Growth of an amorphous, asymptotically circular form at less than unit velocity. This form is obtained by adding even more growth inhibition, *e.g.* with $f(\sigma) = 1$ when $\sigma = 2$.

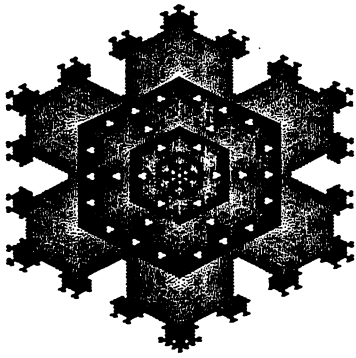
The dendritic forms produced by rules in class *(iii)* exhibit a striking self-similarity: every 2^n time steps, the growing seed forms a plate, then dendritic arms grow from the corners of the plate, sidebranches form, and finally all sidebranches grow into a plate and the process is repeated (Figure 1(a)). This self-similarity may be quantified with a growth dimension that can take on fractional values.[5,6,7] The growth dimension is measurable in experiments, but requires data consisting of the length of the boundary as a function of time, in contrast to the dimension that is often used to characterize other two dimensional patterns like diffusion limited aggregates.[8] It is possible, nevertheless, to see remnants of a snowflake's history embedded in its internal structure, and these sometimes indicate dendritic-plate alternation, with plate boundary length growing exponentially (Figure 1(b)).

The most crucial physical ingredient missing from the cellular automaton model is the flow of heat. This may be modeled with the addition of a continuous variable at each lattice site to represent temperature. The time evolution of the temperature field is given by a discrete approximation to the heat equation, $\dot{T}=c\nabla^2 T$. This amounts to changing a particular site's temperature T_i by taking the average over nearby sites σ_i, and moving T_i toward σ_i by an amount determined by the diffusion constant c. This relaxation method is numerically stable, so that an appropriate choice of diffusion constant assures an accurate simulation of a continuous temperature field in continuous space and time.

The addition of solid again uses a local rule that depends on σ_i, the sum of solid in neighboring sites. Now, however, the local rule yields a continuum temperature threshold value, $T_{thresh}=f(\sigma_i)$. If $T_i<T_{thresh}$ the site is filled with solid; otherwise the site remains empty. The amount of neighboring solid σ_i may be considered as a coarse approximation to the curvature: a boundary site near a convexly curved interface will have fewer neighbors filled with solid than a site near a flat interface. The Gibbs-Thompson effect implies that solidification is more difficult at a convex interface because extra energy must be invested against the force of surface tension. This may be modeled by choosing a function f that takes low values for small σ_i, and high values for larger σ_i. The form of f to be used henceforth will be a quadratic maximum:

$$f(\sigma_i) = \lambda\sigma_i(1-\sigma_i). \qquad (2)$$

*Note that these four classes are quite distinct from the four classes observed in a broader context by Wolfram.[4] Wolfram's classification is on the basis of asymptotic behavior of a cellular automaton rule acting on a random initial condition. Under such circumstances, all the rules discussed here would lead to fixed points, and so would be in Wolfram's class two.



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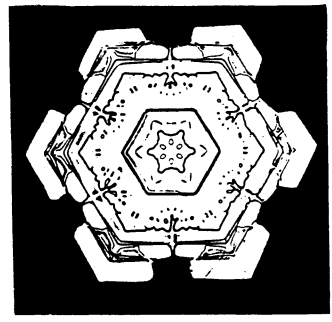

b

Figure 1. (a) Growth from a single site initial condition under the action of a solidification cellular automaton rule on a hexagonal lattice. The grey level changes with time, and repeats a cycle of light to dark every 2^n time steps to display the self-similarity of the growth process, as the growing seed alternates between dendrite and plate forms. (b) A picture of a snowflake, showing internal structure reminiscent of the dendrite-plate alternation.

The final ingredient in the dynamical rule is the effect of solidification on the temperature field. When solid is added to a growing seed, latent heat of solidification must be radiated away. This is modeled by causing an increment in the temperature field. The amount of increase corresponds crudely to the increase in the temperature gradient at the interface. In the following simulations, we simply set the temperature to a constant (high) value when new solid is added. This means that heat flows to nearby interface sites and inhibits their solidification.

This model is a hybrid of discrete and continuum elements. The addition of solid happens in a discrete way, which can be only a very coarse approximation to solid deposition on molecular length scales. There is about ten orders of magnitude between the capillary length and the size of a macroscopic crystal; there is only about 2.4 orders of magnitude between the lattice spacing and the size of the macroscopic crystal in the model. Thus, the micro-scales are brought comparatively quite close to the macro-scales, with the hope that many of the generic macroscopic features of the dynamics will remain. In this respect, this model is similar in spirit to recent molecular dynamics simulations of fluid flow using a cellular automaton rule.[9,10]

The use of a continuum variable at each lattice site to represent temperature gives the model unique features lacking in a purely discrete cellular automaton model. The dynamics may now become parameterized. One parameter is the diffusion rate. Another parameter is the amount of latent heat added upon

solidification. Other parameters may characterize the local temperature threshold function. These parameters may be varied to obtain transitions between macroscopic forms that may be compared with experiment. Figure 2 (a-c) shows a sequence of pictures as the parameter λ is varied from low to high values.

When λ is small, more heat must diffuse away before a boundary site will solidify, so the diffusion length is quite long compared to the lattice spacing. Figure 2(a) illustrates such a case. In the limit of infinite diffusion length, solidification has become known as *diffusion limited aggregation.*

The usual simulations of diffusion-limited aggregation model diffusion by random walking particles which can stick to a growing seed.[8,11] These simulations show the resulting macroscopic form to be a fractal with dimension of $\approx$1.7. Objects with other fractal dimension were also observed. Within the framework of the present model, diffusion limited growth is obtained by having the temperature threshold be small for all values of σ; *i.e.* by setting λ in Eq. (2) to be small. The resulting macroscopic form is displayed in Figure 2(a). The mechanisms for pattern formation in the Witten-Sander model and the present model are slightly different. In the former, voids in the growing structure form because long arms shield regions from subsequent particles. In the latter, voids form because heat is trapped between arms. Nevertheless, the fractal dimension of the two agree to the present accuracy, indicating that they may be in the same universality class. Similar agreement has been indicated in a deterministic simulation of diffusion limited growth using a continuum model.[12]

Figure 2(b) illustrates the effects of raising λ in Eq. (2). There are no longer arbitrarily large voids, but rather a chaotic network of tendrils that appears to have dimension two asymptotically. Though the tendrils are seen to grow in every direction, they show some tendency to grow along lattice directions. The tip splitting instability is apparent,[13] preventing the formation of long dendrites with regular sidebranching.

When λ is raised even further, the tip begins to stabilize, and the tip splitting instability gives way to the sidebranch instability. [1] Anisotropic macroscopic forms have also been seen in stochastic models for diffusion limited aggregation (using an integrated version of random walking particles), if the rule governing the sticking of the particles is made anisotropic.[14] As an anisotropy parameter is varied in the sticking rules, transitions similar to those seen in figure 2 are observed.

Depending on the value of λ and the diffusion constant, the sidebranches can show a variety of structure. In addition to simple, regular sidebranching, the sidebranches can "period double" to display long and short sidebranches alternately. This is evidence for pattern selection mechanism that involves a simple causal relationship between sidebranches rather than filtering of noise at the tip.[15]

The macroscopic forms yielded by this model show remarkable similarity to experiments in pattern formation. The most recent experiments are in two rather different systems: the Hele-Shaw cell, evolution of an interface between two liquids of different viscosity trapped between two plates;[16] and electro-deposition of zinc from an electrolyte solution confined between two plates.[17,18] Both these experiments show transitions between anisotropic forms such as diffusion limited

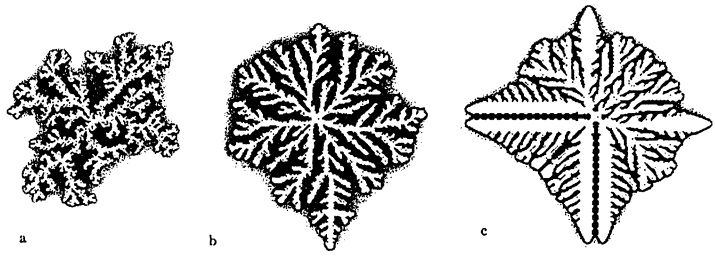

Figure 2. As parameters are varied in the deterministic growth rule (*e.g.* λ in Eq. 2) transitions occur between different macroscopic forms: (a) Amorphous, isotropic fractal growth. The form displays fractal scaling over 2.4 orders of magnitude in length, with a fractal dimension of $1.7 \pm .1$. (b) Tendril growth, dominated by tip splitting, but no apparent fractal structure. Some anisotropy is evident. (c) A macroscopic form showing strong anisotropy, stable parabolic tip with side branching. The temperature field is denoted by a grey scale.

aggregates and forms that show strong anisotropy. In the case of electrodeposition, the anisotropy comes from the underlying crystal structure; in the Hele-Shaw experiment, the anisotropy is imposed by the boundary conditions (scratches on the two dimensional surfaces containing the fluids). A careful comparison of this model with experiment will require new data analysis techniques based on the processing of spatial images.

I am grateful to many people for discussions and comments; most recently these include J. Crutchfield, N. Goldenfeld, Y. Kuramoto, H. Levine, L. Sander, Y. Sawada, and R. Shaw. I am especially grateful to D. Farmer and S. Wolfram for discussions and extensive comments on earlier versions of the manuscript. This work was supported in part by an RCA fellowship, and in part by the U.S. Office of Naval Research under contract number N00014-85-K-0045.

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Q: I understood that your model is two-dimensional. What would be reached by adding one dimension? The snow flakes, though flat, are three-dimensional structures. (Y. Collan)

A: I would expect to see three dimensional structures like those seen in real crystal growth. Current limitations on computer memory prohibit three dimensional simulation.

Q: There is some parallel work performed in Ann Arbor which has had similar results. Garik, Hautman, Hautman, and Richter (Physical Review A) have looked at deterministic growth for a mixed lattice-continuum model. Sander, Ramanlal, and Ben-Jakob (Physical review A) have given a purely continuum theory which is deterministic and seems to produce fractals.
Is the transition from random fractal to dendrite sharp in your system? (L. Sander)

A: I am glad to hear that a simulation of the fully continuous system seems to match the results of my semi-discrete model. This positive comparison for one parameter value increases confidence that my similations of other parameter values are accurate. I cannot yet tell how sharp the transiton is from random fractal to dendrite. Simulations must be done on a larger lattice to answer this question.

Q: Have you tried to simulate the evaporation forms using your deterministic model? You'll obtain a quite different change in forms with time. During evaporation, a hexagonal plate is expected to apear. However, the orientation of the hexagonal plate may be rotated by 30 degrees from that of the initial larger plate. (T. Kuroda)

A: I have not tried evaporation or melting, but it should be easy with the computational machinery I have already developed. Thank you for the suggestion.