On Creating Shapes in 2D Tile Self-Assembly

Abstract

We study the times to grow structures within the tile self-assembly model proposed by Winfree, and the possible shapes that can be achieved. Our earlier work was confined to the growth of rectangular structures, in which the rates of attachment of border tiles and rule tiles were the same. By varying the relative rates one can engineer interesting new shapes, which have been observed in the laboratory. We show that the results from an extension of our earlier stochastic models agree remarkably closely with experimental results.

1 The Tile Self-Assembly Model

The general focus of the work here is on mathematical foundations of self assembly based on Winfree's DNA tile model [9] to be described shortly. More precisely, the emphasis is on the analysis of stochastic models. Although insightful such models and reference theories are ubiquitous in the physical sciences, they remain a fertile ground for self-assembly research in DNA-Based Computing, where stochastic analysis has only recently begun. The early work of Adleman [1] and colleagues and that of the authors [3, 2, 4, 5] sets the stage in this area, and serves as the point of departure for the analysis here.

The seminal mathematical *tile* model of DNA self-assembly, as developed by Winfree [9] and pursued by many others, has led to a much improved understanding of DNA self-assembly in two dimensions. At the physical layer being modeled, single-strand DNA molecules are manipulated to form DNA molecules (e.g., *double-crossover* molecules [6, 7]) which are designed to assemble (bond) with other such molecules in a two dimensional crystal-growth process obeying bonding rules determined by the molecular motifs [9]. These building-block molecules are modeled as tiles.

There are three types of tiles involved in a self-assembly process: rule tiles, border tiles, and seed tiles. They participate in a growth process beginning at the origin of the positive lattice; the unit squares of the lattice are the potential sites occupied by tiles.

- The seed tile occupies the lower-left corner of the positive lattice and is responsible for initiating the tile self-assembly process. Only border tiles, as described next, can stick to the two free (upper and right-hand) sides of the seed tiles.
- A border tile can join the structure only by attaching to the seed tile or another border tile along the horizontal and vertical boundaries of the positive lattice, each such attachment extending one of the borders of the structure assembled so far.
- A rule tile can attach to the growing structure at any available site which is adjacent to occupied sites both to the left and below the available site, where either site may contain a border tile.

In the probability model governing growth (cf. [2, 5]) , as soon as a site becomes available to a rule tile (for attachment to the left and below) its waiting time for such a tile is exponentially distributed with mean 1.

All such waiting times are independent. The attachment of new border tiles is also subject to independent exponential waiting times, but with a different rate parameter α. In the analysis of the first such model [2], the border (input) tiles were assumed to be prefabricated, occupying sites, say, from (0,0) to (M,0) and to (0,N), the so-called *L input*; the problem was to estimate the time $C_{M,N}$ until a rule tile became attached to the site at (M,N).

One of the principal tools used in solving this type of problem has been the analysis of the TASEP (Totally Asymmetric Simple Exclusion Process) [8]; the main result is given below in terms of fluid limits where the discrete position variables i and j are replaced by continuous variables x and y. We have, with the obvious change in notation,

$$\lim_{(x,y)\to(\infty,\infty)} C_{(x,y)} = \sup_{\gamma} \left(\sqrt{x}+\sqrt{y}\right)^2 \tag{1}$$

where γ represents all possible paths from the origin (the seed tile) to the point (x, y).

In the more general setting, where border tiles are not prefabricated, but take part in the self-assembly process, the problem becomes much more interesting and entails the assembly of a much larger class of shapes, particularly when $\alpha < 1$, i.e., the border-tile attachment rate is less than that of the rule tiles.[1]

The more general extremal problem is illustrated in Figure 1. Since $\alpha < 1$, a path along which the sum of expected attachment times is maximum must first move along one of the axes; the slow growth rate of the borders is less than the rate of tiling the rectangles bounded by the borders. At a point where these rates balance, the trajectory is determined by the rectangle tiling result of (1). Thus, in the figure, to compute a path to (x, y) along which expected total attachment time is maximum, one must find this maximum among paths like $\overline{ba}$, if $x \le y$, and $\overline{dc}$, if $x \ge y$.

With z as defined in the figure, the extremal path of tile attachments will start from the origin, go to position $(0, y-z)$ in expected time $(y-z)/\alpha$ or $(x-z, 0)$ in expected time $(x-z)/\alpha$, and then go on to reach (x, y). We need only find the supremum over z to identify the extremal path. Thus,

$$C_{(x,y)} \sim \sup_{z} \left(\left(\sqrt{x}+\sqrt{z}\right)^2 + \frac{y-z}{\alpha} \right) \tag{2}$$

as $(x, y) \to (\infty, \infty)$. A calculation then shows that

$$\begin{aligned} C_{(x,y)} &\sim x\left(1+\frac{\alpha}{1-\alpha}\right)^2 + \frac{y}{\alpha} - \frac{x\alpha}{(1-\alpha)^2} \\ &= \frac{y}{\alpha} + \frac{x}{1-\alpha} \end{aligned} \tag{3}$$

as $(x, y) \to (\infty, \infty)$, where

$$z = x\left(\frac{\alpha}{\alpha-1}\right)^2 \text{ and } z < y$$

[1] K. Fujibayashi working in E. Winfree's lab at Cal Tech notes that $\alpha \approx 1/2$ was common in certain of his experiments, where α is a stoichiometric parameter giving the concentration of border tiles. In this case he found that the profile of the self-assembly structure is triangular, a fact that will also emerge in our purely mathematical framework.

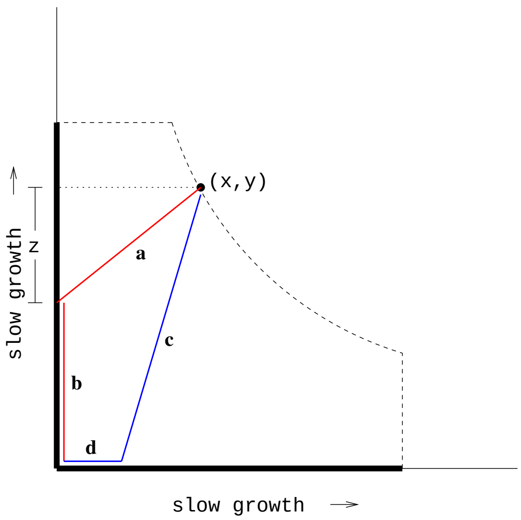


Figure 1: Expected shape of the structure

An analysis of the alternative path $\overline{dc}$ follows the same arguments and yields the same result with x and y interchanged.

By fixing the computing time, we can plot values of x and y for different values of α as shown in Figure 2. For $0 \leq \alpha \leq \frac{1}{2}$, the shape of the structure is covered by the yellow and light blue line. When $\alpha = \frac{1}{2}$,

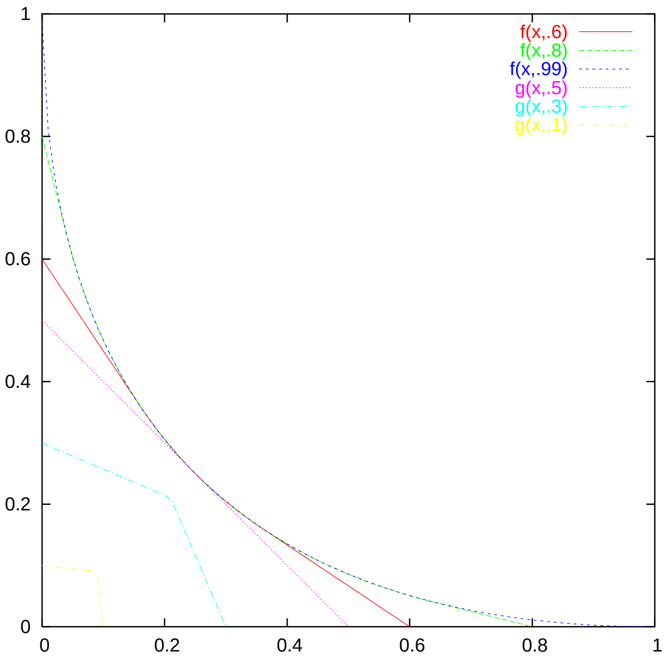
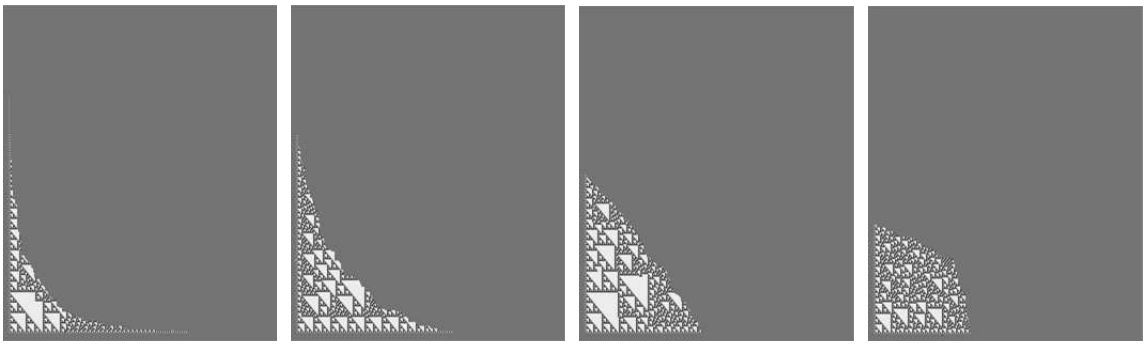


Figure 2: Shape of the structure for various α

the shape of the structure is a triangle (covered by a pink line). When $\alpha > \frac{1}{2}$, the shape of the structure is



(a) $\alpha = 1.2$ (b) $\alpha = 0.8$ (c) $\alpha = 0.5$ (d) $\alpha = 0.3$

Figure 3: Simulation of 2D structures with various values of α

covered by the red, green and blue lines. The shape will look quite linear near the border of the growth, then the profile will be hyperbolic in the middle.

It is clear from (3) that the time a tile takes to attach to position (x, y) depends linearly on the value of x and y, given that α is a constant. This explains why the asymptotic shape of the structure is triangular. In the case of $\alpha = 0.5$ (used in Fujibayashi's experiment), both paths are exactly the same which yields a triangular structure. The results of Fujibayashi's simulations of self assembly structures, as shown in Figure 3 for various α, are in excellent agreement with our predictions for all values of $\alpha \leq \frac{1}{2}$.

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References

[1] L. Adleman, Q. Cheng, A. Goel, M.-D. Huang, and H. Wasserman. Linear self-assemblies: Equilibria, entropy, and convergence rates. In Elaydi, Ladas, and Aulbach, editors, *New progress in difference equations*. Taylor and Francis, London, 2004.

[2] Y. Baryshnikov, E. Coffman, and P. Momčilović. DNA-based computation times. In *Proc. of the Tenth International Meeting on DNA Computing*, Milan, Italy, June 2004.

[3] Y. Baryshnikov, E. Coffman, and P. Momčilović. Incremental self-assembly in the fluid limit. In *Proc. 38th Ann. Conf. Inf. Sys. Sci.*, Princeton, NJ, 2004.

[4] Y. Baryshnikov, E. Coffman, and P. Momčilović. Phase transitions and control in self assembly. In *Proc. Foundations of Nanoscience: Self-Assembled Architectures and Devices*, Snowbird, UT, April 2004.

[5] Y. Baryshnikov, E. Coffman, N. Seeman, and B. Yimwadsana. Self correcting self assembly: Growth models and the hammersley process. In *Proc. of the Eleventh International Meeting on DNA Computing*, London, Ontario, 2005.

[6] B. Ding, R. Sha, and N. Seeman. Pseudohexagonal 2D DNA crystals from double crossover cohesion. *J. Am. Chem. Soc.*, 126:10230–10231, 2004.

[7] T.-J. Fu and N. Seeman. DNA double crossover structures. *Biochemistry*, 32:3211–3220, 1993.

[8] T. M. Liggett. *Interacting Particle Systems*. Springer-Verlag, New York, 1985.

[9] E. Winfree. *Algorithmic Self-Assembly of DNA*. PhD thesis, California Institute of Technology, Pasadena, CA, 1998.