Incremental Self Assembly in the Fluid Limit

Yuliy Baryshnikov[1] Ed Coffman[2] Petar Momčilović[3]

[1] Bell Labs, Lucent Technologies, Murray Hill, NJ 07974, ymb@research.bell-labs.com
[2] Department of Electrical Engineering, Columbia University, NY 10027, egc@ee.columbia.edu
[3] IBM T.J. Watson Research Center, Yorktown Heights, NY 10598, petar@us.ibm.com

November 24, 2003

Abstract

Self assembly (SA) processes are fundamental to a number of computational and fabrication technologies in nanoscience. Indeed, their efficiency and robustness are critical to the success of the underlying concepts. SA typically involves random phenomena at some level, yet the development of reference theories for the study of random SA processes is scarcely begun. This paper studies a model of SA processes in which assembled objects are built one element (tiles, molecules, ...) at a time. The contributions here are threefold: First, the hydrodynamic limit is introduced as a powerful tool for the study of SA, essentially reducing the study of complex stochastic structures to the solution of differential equations. A second contribution consists of explicit results on SA *yield*, the fraction of the absorbing state that contains the desired self-assembled objects. It is shown that, asymptotically, yields are factorially small in the sizes of the assembled objects. Low yields motivate our third contribution: the analysis of a control mechanism (modeling temperature variation for example) that implements *reversible* SA, where disassembly of objects can be exploited to improve the overall assembly process. An optimal control is exhibited for obtaining 100% yields.

1 Introduction

Self assembly (SA) refers to the basic bottom-up paradigm of numerous constructive and computational processes that have applications spanning nanoscale electronics, biotechnology, medicine, and sensor networks. Nanofabrication includes molecular SA [8, 6, 9] (e.g., polymerization) by which novel functional materials are produced, and nanowire SA for producing elementary nanodevices such as switches, gates, and memory elements [7] which can be further assembled into nanocomputers. On the computational side, SA plays a fundamental role in amorphous computing [1] and DNA computing [3, 5].

The dramatic advances in nanoscience belong primarily to the experimental scientists; as in other applications, mathematical modeling and the development of reference theories tend to follow experimental proofs of concept. There have been a number of combinatorial studies [3, 4], but SA is also typically a stochastic phenomenon; and although insightful stochastic models have been virtually ubiquitous in the physical sciences, they remain a fertile ground for SA research. The early work of Adleman and colleagues sets the stage, and serves as our point of departure. In this paper, we broaden their approach to SA modeling in a way that facilitates yield calculations for irreversible SA and the study of an intriguing problem of minimizing self-assembly times under a simple control on reversibility. To explain these issues in detail, we first describe the probability model of Adleman et al [2].

1.1 Stochastic polymerization.

In the DNA-computing terms of [2], which we use here, elementary particles called *tiles* combine to form progressively larger geometric shapes, or complexes, called *supertiles*. In the general tile-system model, there is a set of tile types and a collection of bonding (sticking or glueing) rules governing how tiles and supertiles combine into larger supertiles. Stochastic analysis of such systems has so far been limited to a process called *n-linear polymerization* [2]. In this problem, there are n tile types $T_1, \ldots, T_n$; a tile of each type has a left and right face at which bonding can take place. The right face of a tile of type T_i can bond to the left face of a tile of type T_{i+1}, $i = 1, \ldots, n-1$, and these are the only bonds possible. Thus, supertile types are restricted to subsequences of $T_1, T_2, \ldots, T_n$. In this setting, SA can be viewed as taking place on the integers, with a unit-width tile between every pair of adjacent integers. The initial ordering is uniformly random with equal concentrations of the n tile types. SA proceeds in time steps called "tosses". At the beginning of each step all allowable bondings occur independently with probability σ; then, at the end of each time step, the entire sequence is re-randomized (after being "tossed into the air" the supertiles return to random positions) with each existing bond being broken independently with probability σ'. Questions resolved in [2] include the existence of equilibria, a thermodynamic rationalization of the SA process, and, in the irreversible case with $\sigma' = 0$, an estimate of the time to reach the absorbing state in which only the supertiles with type sequences $T_1, \ldots, T_n$ exist.

1.2 Incremental SA

In the title process, all elementary, unit tiles are subject to the same bonding rules, or equivalently, the system is homogeneous with just a single tile type. Bonding rules are allowed to vary only among supertiles containing at least 2 tiles. Hereafter, we say *i-tile*, $i \geq 1$, when referring to a supertile containing i unit tiles. Initially, consider a system of n 1-tiles, with n large, and assume that assembly events occur in a rate-1 Poisson stream. Let $\eta_i(t)$ denote the number of i-tiles at time t of the process, so the initial condition of the

assembly process is $\eta_1(0) = n,\ \eta_i(0) = 0,\ i > 1$. Introduce the time-scaled concentrations

$$x_i^{(n)}(t) = \frac{\eta_i(nt)}{n}$$

so that

$$\sum_{i \geq 1} x_i^{(n)}(t) = 1 \tag{1}$$

for all n, t. We focus on the fluid limit, $n \to \infty$, of the assembly process, which we denote by $\{x_i(t)\}$. Informally, by a mean-field argument, the rate of formation/decomposition of i-tiles is proportional to the concentrations of its constituent tiles. At assembly times in a discrete model, this corresponds to choosing the assembling tiles independently at random with probabilities given by their concentrations. Throughout the paper we assume that each possible *pair* of tiles is chosen uniformly at random. Alternatively, one could examine the model in which two *tiles* are chosen at random and then glued if possible. This approach leads in the usual way to systems of differential equations, as illustrated below. Note that there is no intrinsic limitation to spatial linearity in this model. Also, since we are exploring behavior in the fluid limit, our results also apply to situations in which there are different tile types, as long as they have the same initial concentrations and symmetric behavior. For example, suppose the final supertiles are 3-tiles, there are 3 tile types, no two tiles of the same type can appear in the same supertile, and the concentrations of the 3 tile types are equal in the initial state.

The next two sections present the analysis of a baseline model for calculating yields and for studying the problem of optimizing concentrations in reversible systems. The irreversible model is defined by a parameter $N > 2$, an initial state consisting only of 1-tiles, and the following (incremental) bonding rule:

> *Two i-tiles can bond together only if one of them is a 1-tile and the other is an i-tile for some $i < N$.*

Thus, the N-tiles are the objective of the assembly process. The yield of the irreversible process is the fraction β of 1-tiles that have assembled into N-tiles in an absorbing state containing only i-tiles with $1 < i \leq N$.

2 Assembly of triangles

We begin with an analysis of the case $N = 3$, which we call the case of *triangles*, for both irreversible SA and a reversible version in which the bonding rule is augmented by a "breaking" rule:

> *The bonds of 2-tiles are broken independently, each with a given probability.*

2.1 The irreversible process

For 1-tiles, we express the dynamics over a small time interval $[t, t+\Delta t]$ as

$$x_1^{(n)}(t+\Delta t) = x_1^{(n)}(t) - 2[x_1^{(n)}]^2(t)\Delta t - x_1^{(n)}(t)x_2^{(n)}(t)\Delta t + o(\Delta t) \tag{2}$$

from which one obtains

$$\frac{dx_1^{(n)}}{dt} = -2[x_1^{(n)}(t)]^2 - x_1^{(n)}(t)x_2^{(n)}(t) \tag{3}$$

and in the fluid limit, $n \to \infty$,

$$\frac{dx_1}{dt} = -2x_1^2 - x_1x_2 \tag{4}$$

with $x_1(0) = 1$. The factor of two reflects the fact that *two* 1-tiles are removed from the population for each 2-tile formed. A similar argument leads to

$$\frac{dx_2}{dt} = x_1^2 - x_1x_2 \tag{5}$$

$$\frac{dx_3}{dt} = x_1x_2 \tag{6}$$

with $x_2(0) = x_3(0) = 0$. There are no 1-tiles in the absorbing state, so by (1), the yield is

$$\beta = 3x_3(\infty) = 1 - 2x_2(\infty)$$

To find $x_2(\infty)$, we could treat x_2 as a function $y(\cdot)$ of $x \equiv x_1$, in which case (4)-(5) would reduce to the ODE

$$\frac{dy}{dx} = \frac{y-x}{y+2x} \tag{7}$$

with the boundary condition $y(1) = 0$ (there are no 2-tiles in the initial state where $x = x_1 = 1$). The change of variable $\hat{y} = y/x$ would then convert this to a separable ODE which could be integrated to give y as an implicit function of x. However, it is more instructive to apply the following approach, as it easily generalizes to arbitrary N. The *trajectories* of (x_1, x_2) described by (7) are also described by the *linear* system

$$\frac{dx_1}{ds} = -2x_1 - x_2 \tag{8}$$

$$\frac{dx_2}{ds} = x_1 - x_2 \tag{9}$$

where $ds = x_1dt$, and where, as functions of s, $x_1(0) = 1,\ x_2(0) = 0$. This system is readily solved explicitly. Indeed, as is easily checked, the solution is given by

$$x_1(s) = e^{-3s/2}\left(\cos\tfrac{\sqrt{3}}{2}s - \tfrac{\sqrt{3}}{3}\sin\tfrac{\sqrt{3}}{2}s\right)$$

$$x_2(s) = \tfrac{2\sqrt{3}}{3}e^{-3s/2}\sin\tfrac{\sqrt{3}}{2}s$$

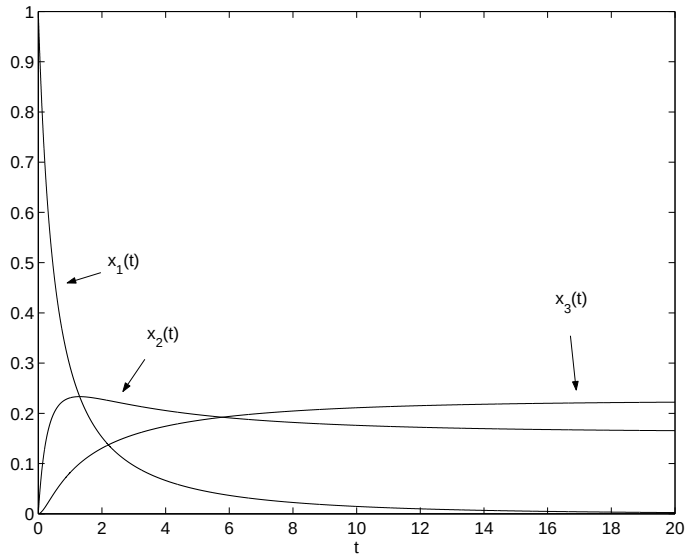

Figure 1: Concentrations of 1, 2 and 3-tiles as functions of time for the irreversible process. The assembly starts at time $t = 0$.

The value of $x_1(s)$ is 0 at

$$s_* = \frac{2\sqrt{3}\pi}{9}$$

which corresponds to the state at which there are no more 1-tiles. At that point

$$x_2(s_*) = \exp\left(-\sqrt{3}\pi/3\right) \approx 0.163033535^-$$

This gives a yield of

$$\beta = 1 - 2x_2(s_*) = 0.67393293^+$$

so that roughly a third of the 1-tiles are wasted in pairs.

The numerical solutions for concentrations of 1, 2 and 3-tiles as functions of time is shown in Figure 1.

2.2 The reversible process

Higher, indeed maximum, yields are achieved by the reversible process in which 2-tiles can decompose, returning two 1-tiles to the system. Reversibility is controlled by a time-varying

parameter $u(t)$ and is reflected in "control terms" appended to (4) and (5) as shown below:

$$\frac{dx_1}{dt} = -2x_1^2 - x_1x_2 + 2ux_2$$
$$\frac{dx_2}{dt} = x_1^2 - x_1x_2 - ux_2$$

It might be appropriate to think of $u(t)$ as time-varying temperature.

The goal now is to set up the control so that, for a given time horizon T, the concentrations converge to a state at time T in which the yield is maximized. First, to maximize the growth rate $\frac{dx_3}{dt} = x_1x_2$ of 3-tiles for a given number, say $x_1 + 2x_2 = c$, of all 1-tiles as yet unassembled into 3-tiles, we must take $x_1 = c/2$ and $x_2 = c/4$, i.e., we must have twice as many 1-tiles as 2-tiles. Then, to find the desired control $u(t)$, we solve a related problem

$$\frac{dz_1}{dt} = -2z_1^2 - z_1z_2 + 2wz_2$$
$$\frac{dz_2}{dt} = z_1^2 - z_1z_2 - wz_2$$
$$\frac{dz_3}{dt} = z_1z_2$$
$$z_1 = 2z_2$$

with $z_1(0) = 2K$, $z_2(0) = K$, and $z_3(0) = 0$, for some $K > 0$. Solving this system gives $w = 7z_2/2$, which results in

$$z_1 = 2z_2 = \frac{4w}{7} = \frac{2}{3t/2 + 1/K}$$

and

$$z_3 = \frac{4K}{3} - \frac{4/3}{3t/2 + 1/K}$$

Then going back to the original problem, the optimal strategy is defined by

$$u(t) = \frac{7/2 \cdot 1\{t > \tau\}}{3(t-\tau)/2 + 1/\alpha}$$

where $\tau := \inf\{t > 0 : x_1 = 2x_2\}$ and $x_2(\tau) = \alpha$ must be determined numerically. In words, the system is allowed to evolve (absent control) from the initial state $x_1 = 1$ until such time as the concentration of 1-tiles first becomes exactly twice that of 2-tiles (at time τ). At this point, control is exerted as above so as to preserve the relative concentrations of 1- and 2-tiles.

The numerical solutions for x_1, x_2 and x_3 as functions of time for the reversible process in the optimal-control case are shown in Figure 2. As can be seen in the figure, the reversible setup results in a higher yield than the irreversible one.

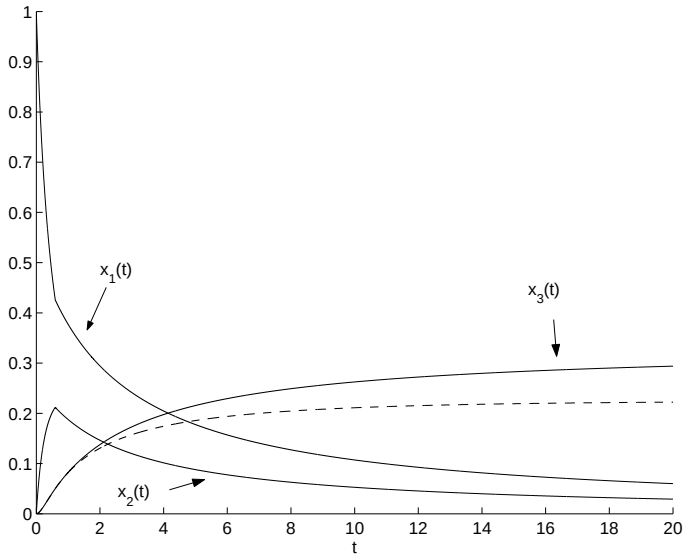

Figure 2: Concentrations of 1, 2 and 3-tiles as functions of time for the reversible process under the optimal control. The assembly starts at time $t = 0$. After time $t = \tau$ the ratio of x_1/x_2 is fixed at 2. The dashed line indicates the concentration of 3-tiles in the corresponding irreversible process.

3 The irreversible process for arbitrary size objects

We return to the irreversible model and study the yield for large N. First, replace the system (8)-(9) by the easy generalization

$$\frac{dx_1}{ds} = -2x_1^2 - x_1 \sum_{k=2}^{N-1} x_k$$

$$\frac{dx_k}{ds} = x_1 x_{k-1} - x_1 x_k, \qquad k = 2, \ldots, N-1$$

with initial conditions $x_1(0) = 1$ and $x_k(0) = 0$, $k = 2, \ldots, N-1$. Introducing a new variable $ds = x_1\, dt$ then yields the linear system

$$\begin{aligned} \frac{dx_1}{ds} &= -2x_1 - \sum_{k=2}^{N-2} x_k \\ \frac{dx_k}{ds} &= x_{k-1} - x_k, \qquad k = 2, \ldots, N-1. \end{aligned} \tag{10}$$

Next, let A be the coefficient matrix of this system, and let E be the identity matrix of dimension $N-1$. In order to obtain the characteristic roots of the system we compute

$$\det(A - \lambda E) = \det(\tilde{A} - (\lambda + 1)E),$$

where

$$\tilde{A} = \begin{bmatrix} -1 & -1 & -1 & \ldots & -1 \\ 1 & 0 & & & \\ & 1 & 0 & & \\ & \vdots & & & \end{bmatrix}$$

Exploiting the structure of $\tilde{A}$, we find that

$$\det(A - \lambda E) = (-1)^{N-1} \frac{(\lambda + 1)^N - 1}{(\lambda + 1) - 1}$$

which implies that the characteristic roots are given by

$$\lambda_\ell = e^{i2\pi\ell/N} - 1, \;\; \ell = 1, \ldots, N-1 \tag{11}$$

with $i = \sqrt{-1}$. Therefore, the x_k as functions of s have the following form

$$x_k(s) = \sum_{\ell=1}^{N-1} c_{k\,\ell} e^{\lambda_\ell s} \tag{12}$$

for coefficients $c_{k\ell}$ to be determined. Substitution of (12) into (10) results in

$$\sum_{\ell=1}^{N-1} \lambda_\ell c_{k\,\ell} e^{\lambda_\ell s} = \sum_{\ell=1}^{N-1} (c_{\{k-1\}\,\ell} - c_{k\,\ell}) e^{\lambda_\ell s}, \quad k = 2, \ldots N-1$$

from which one obtains the recurrence $c_{k\,\ell}(\lambda_\ell + 1) = c_{\{k-1\}\,\ell}$ with the solution

$$c_{k\,\ell} = (\lambda_\ell + 1)^{-k+1} c_{1\,\ell} \tag{13}$$

For notational convenience let $c_\ell \equiv c_{1\,\ell}$, $\ell = 1, \ldots, N-1$. Combining (13) with (12) produces

$$x_k(s) = \sum_{\ell=1}^{N-1} c_\ell e^{-i\frac{2\pi\ell(k-1)}{N}} e^{\lambda_\ell s} \tag{14}$$

At this point all x_k are defined in terms of the unknown c_ℓ. In what follows we use the initial conditions $x_1(0) = 1$ and $x_k(0) = 0$, $k = 2, \ldots, N-1$ to determine the unknown coefficients. With $\varepsilon := e^{-i\frac{2\pi}{N}}$, the initial conditions and (14) result in

$$\begin{aligned} &\sum_{\ell=1}^{N-1} c_\ell = 1 \\ &\sum_{\ell=1}^{N-1} c_\ell \varepsilon^{\ell(k-1)} = 0, \qquad k = 2, \ldots, N-1 \end{aligned}$$

This system can be rewritten in the matrix form $BC = U$,

$$\begin{bmatrix} 1 & 1 & 1 & 1 & 1 & \ldots & 1 \\ \varepsilon & \varepsilon^2 & \varepsilon^3 & \varepsilon^4 & & & \varepsilon^{N-1} \\ \varepsilon^2 & \varepsilon^4 & \varepsilon^6 & & & & \varepsilon^{2N-2} \\ \vdots & & & & & & \vdots \\ \varepsilon^{N-2} & \ldots & & & & & \varepsilon^{N^2-2N+2} \end{bmatrix} \begin{bmatrix} c_1 \\ c_2 \\ c_3 \\ \vdots \\ c_{N-1} \end{bmatrix} = \begin{bmatrix} 1 \\ 0 \\ 0 \\ \vdots \\ 0 \end{bmatrix}$$

By Cramer's rule the solution is given by

$$c_\ell = \frac{\det B_\ell}{\det B} \tag{15}$$

where B_ℓ is obtained from B by replacing the ℓth column with U. It can be shown that, because of the form of B, one has

$$\det B = \prod_{1 \le j < k \le N-1} (\varepsilon^k - \varepsilon^j) \tag{16}$$

whereupon, after tedious algebra,

$$\begin{aligned}
\det B_\ell &= (-1)^{\ell+1} \begin{vmatrix} \varepsilon & \varepsilon^2 & \dots & \varepsilon^{\ell-1} & \varepsilon^{\ell+1} & \dots & \varepsilon^{N-1} \\ \varepsilon^2 & \varepsilon^4 & \dots & & & & \\ \vdots & & & & & & \\ \varepsilon^{N-2} & & \dots & & & & \end{vmatrix} \\
&= (-1)^{\ell+1} \varepsilon^{\frac{N(N-1)}{2}-\ell} \begin{vmatrix} 1 & 1 & \dots & 1 & 1 & \dots & 1 \\ \varepsilon & \varepsilon^2 & \dots & \varepsilon^{\ell-1} & \varepsilon^{\ell+1} & \dots & \varepsilon^{N-1} \\ \vdots & & & & & & \\ \varepsilon^{N-3} & \varepsilon^{2N-6} & \dots & & & & \end{vmatrix} \\
&= (-1)^{\ell+1} \varepsilon^{\frac{N(N-1)}{2}-\ell} \prod_{\substack{1\le j<k\le N-1 \\ j,k\neq\ell}} (\varepsilon^k - \varepsilon^j)
\end{aligned}$$

Recall that $\varepsilon = e^{-i\frac{2\pi}{N}}$, which in conjunction with the preceding formula, implies

$$\det B_\ell = (-1)^{N+\ell} \varepsilon^{-\ell} \prod_{\substack{1\le j<k\le N-1 \\ j,k\neq\ell}} (\varepsilon^k - \varepsilon^j) \tag{17}$$

Substituting (16) and (17) into (15) one obtains

$$\begin{aligned}
c_\ell &= (-1)^{N-1} \varepsilon^{-\ell} \left[\prod_{\substack{1\le j\le N-1 \\ j\neq\ell}} (\varepsilon^j - \varepsilon^\ell) \right]^{-1} \\
&= (-1)^{N-1} (1-\varepsilon^\ell) \left[\prod_{1\le j\le N-1} (\varepsilon^j - 1) \right]^{-1}
\end{aligned}$$

where, to get the last expression, we used the properties of $\varepsilon = e^{-i\frac{2\pi}{N}}$. Hence, in view of (14)

$$x_k(s) = \left[\prod_{1\le j\le N-1} (1-\varepsilon^j) \right]^{-1} \sum_{\ell=1}^{N-1} (\varepsilon^{-\ell} - 1) \varepsilon^{\ell k} e^{\lambda_\ell s}$$

Since $x_1(0) = 1$, the continued product must evaluate to N, so

$$x_k(s) = \frac{1}{N} \sum_{\ell=1}^{N-1} (\varepsilon^{-\ell} - 1) \varepsilon^{\ell k} e^{\lambda_\ell s} \tag{18}$$

For the yield calculation, let s_* be the first time that $x_1(s) = 0$ (no singles are left), i.e., $s_* = \inf\{s : \ x_1(s) = 0\}$ is the time the system enters an absorbing state in the s-domain. Then by (18) a compact formula for the yield develops as follows:

$$\begin{aligned}\beta = 1 - \sum_{k=1}^{N-1} k\, x_k(s_*) &= 1 - \sum_{\ell=1}^{N-1} \frac{\varepsilon^{-\ell} - 1}{\varepsilon^{\ell} - 1} e^{\lambda_\ell s_*} \\ &= 1 + \sum_{\ell=1}^{N-1} \varepsilon^{-\ell} e^{\lambda_\ell s_*} \\ &= \sum_{\ell=0}^{N-1} \varepsilon^{-\ell} e^{\lambda_\ell s_*}\end{aligned}$$

where $\lambda_0 = 0$. Introducing the formula for the λ_ℓ given by (11), we find that

$$\begin{aligned}\beta &= e^{-s_*} \sum_{\ell=0}^{N-1} \varepsilon^{-\ell} \sum_{k=0}^{\infty} \frac{\varepsilon^{-k\ell} s_*^k}{k!} \\ &= e^{-s_*} \sum_{k=0}^{\infty} \frac{s_*^k}{k!} \sum_{\ell=0}^{N-1} \varepsilon^{-\ell(k+1)}\end{aligned}$$

Next, we observe that

$$\sum_{\ell=0}^{N-1} \varepsilon^{-\ell k} = \sum_{\ell=0}^{N-1} \varepsilon^{\ell k} = \begin{cases} N & k = mN, \ m \in \mathbb{Z} \\ 0 & \text{otherwise} \end{cases} \tag{19}$$

and hence

$$\beta = e^{-s_*} \sum_{k=1}^{\infty} \frac{N s_*^{kN-1}}{(kN-1)!} \tag{20}$$

Returning to (18), we set $k = 1$ and use $x_1(0) = 1$ to obtain the value of s_*

$$\begin{aligned}0 &= N^{-1} e^{-s_*} \sum_{\ell=0}^{N-1} (1 - \varepsilon^{\ell}) e^{\varepsilon^{-\ell} s_*} \\ &= N^{-1} e^{-s_*} \sum_{k=0}^{\infty} \frac{s_*^k}{k!} \sum_{\ell=1}^{N-1} \left[\varepsilon^{-\ell k} - \varepsilon^{-\ell(k-1)} \right]\end{aligned}$$

Thus, in view of (19), s_* satisfies

$$1 - s_* + \sum_{k=1}^{\infty} \frac{s_*^k}{(kN)!} \left(1 - \frac{s_*}{kN+1} \right) = 0 \tag{21}$$

Computing s_* numerically from this equation and substituting into (20) then gives the yield.

Consider next asymptotics in N. Recalling that $ds = x_1\,dt$ we see that s_* can be interpreted as the integral of x_1 in the time domain, and so $s_* = s_*(N)$ is a nondecreasing function in N. This coupled with (21) leads to

$$s_*(N) = 1 + O(1/N!)$$

which together with (20) gives

Proposition 1 *As $N \to \infty$ the yield satisfies*

$$\beta(N) = \frac{N^2}{N!}(1 + o(1))$$

4 Concluding remarks

The small yields of Proposition 1 lead to obvious, yet exciting challenges. We mention just two. First, can the analysis of the reversible process be generalized to larger object sizes? Second, what can be proved about yields in the more general SA process where any two objects can combine so long as the size of the newly created object does not exceed the size of the final object of the assembly? Simulations of such processes have been conducted for values of N up to 20. The yield results shown in Fig. 3 suggest that generalizing incremental assembly can lead to substantial increases in yield. The monotonicity shown in the figure is to be expected, but the dependence on parity is intriguing; yield for any even final-object size $N \geq 4$ is higher than that for the odd object size $N - 1$.

References

[1] H. Abelson, D. Allen, D. Coore, C. Hanson, G. Homsy, T. Knight, R. Nagpal, E. Rauch, G. Sussman, and R. Weiss. Amorphous computing. *Communications of the ACM*, 43(5):74–82, 2000.

[2] L. Adleman, Q. Cheng, A. Goel, M. Huang, and H. Wasserman. Linear self-assemblies: Equilibria, entropy, and convergence rates. In *New progress in difference equations.* Proceedings, 6th International Conf. on Difference Equations and Applications (to appear).

[3] L. Adleman, Q. Cheng, A. Goel, and M-D. Huang. Running time and program size for self-assembled squares. In *Proceedings, ACM Symposium on Theory of Computing (STOC'01)*, pages 740–748, 2001.

[4] L. Adleman, Q. Cheng, A. Goel, M-D. Huang, D. Kempe, P. Moisset de Espanés, and P. Rothemund. Combinatorial optimization problems in self-assembly. In *Proceedings, ACM Symposium on Theory of Computing (STOC'02)*, May 19–21 2002.

[5] A. Carbone and N. Seeman. Circuits and programmable self-assembling DNA structures. *PNAS*, 99, October 2002.

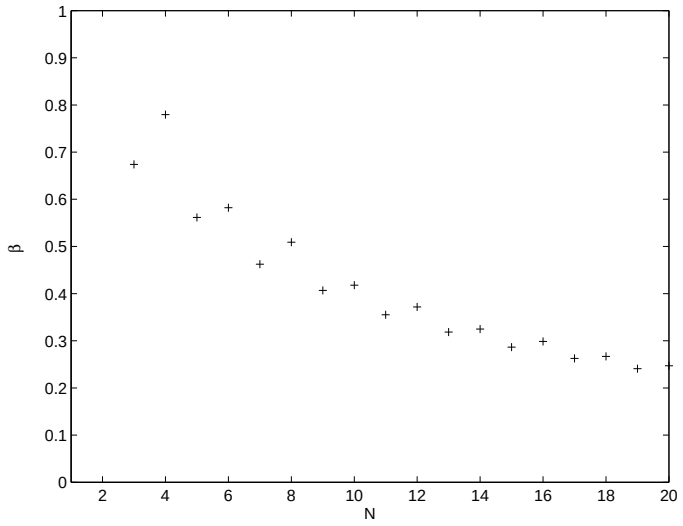

Figure 3: Yield as a function of the final object size for the case when objects of arbitrary size can combine (non-incremental self assembly).

[6] M. Gomez-Lopez, J. Preece, and J. Stoddart. The art and science of self-assembling molecular machines. *Nanotechnology*, 7:183–192, 1996.

[7] D. Gracias, J. Tien, T. Breen, C. Hsu, and G. Whitesides. Forming electrical networks in three dimensions by self-assembly. *Science*, 289:1170–1173, 2000.

[8] G. Lopinski, D. Wayner, and R. Wolkow. Self-directed growth of molecular nano-structures. *Nature*, 406(48), 2000.

[9] G. Whitesides, J. Mathias, and C. Seto. Molecular self-assembly and nanochemistry: A chemical strategy for the synthesis of nanostructures. *Science*, 254:1312–1319, 1991.