Self-correcting Self-assembly: Growth Models and the Hammersley Process

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Abstract. This paper extends the stochastic analysis of self assembly in DNA-based computation. The new analysis models an error-correcting technique called *pulsing* which is analogous to checkpointing in computer operation. The model is couched in terms of the well-known tiling models of DNA-based computation and focuses on the calculation of computation times, in particular the times to self assemble rectangular structures. Explicit asymptotic results are found for small error rates q, and exploit the connection between these times and the classical Hammersley process. Specifically, it is found that the expected number of pulsing stages needed to complete the self assembly of an $N \times N$ square lattice is asymptotically $2N\sqrt{q}$ as $N \to \infty$ within a suitable scaling. Simulation studies are presented which yield performance under more general assumptions.

1 Introduction

In many respects, the current state of DNA-based computing resembles the state of standard, electronic computing a half century ago: a fascinating prospect is slow to develop owing to inflexible interfaces and unacceptably low reliability of the computational processes. We concentrate in this paper on the latter aspect, specifically addressing the interplay between the reliability and speed of DNA computing.

While DNA-based computational devices are known to be extremely energy efficient, their reliability is seen as a major obstacle to becoming a viable computing environment. As DNA based computing becomes more fully developed, the speed of self assembly will become a crucial factor; but as of now, little is known concerning the fundamental question of computation times. We emphasize the intrinsic connection between the two problems of reliability and speed, because of the unavoidable trade-off that exists between them. A clear understanding of the limitations of self-assembly reliability and speed, specifically that of DNA-based computing, and the interplay between these properties, will be paramount in determining the full potential of the paradigm. Our past work,

A. Carbone and N.A. Pierce (Eds.): DNA11, LNCS 3892, pp. 1–11, 2006.

briefly reviewed later, analyzed for a given function the time required to determine its value on given inputs, and therefore established theoretical limits on the performance of DNA-based computers. In the simplest instance, the analysis of computation times has surprising connections with interacting particle systems and variational problems, as shown in [1], and as further developed here. The critical new dimension of this paper is that of error correction; the new contributions lie in (a) a novel approach to dramatic improvements in the reliability of computations and (b) in the analysis of the inevitable performance losses of reliable computations.

The early theoretical work on DNA-based computation focused chiefly on various measures of complexity, in particular, program-size and time complexity [2,3,4]. However, Adleman et al [2,5] also investigated interesting combinatorial questions such as the minimum number of tile types needed for universality, and stochastic optimization questions such as the choice of concentrations that leads to minimum expected assembly times. Apart from these works, the mathematical foundations of computational speed in a stochastic context appear to be restricted to the work of Adleman et al [6] and Baryshnikov et al [1,7,8,9]. The former work studies random self assembly in one dimension. In a problem called *n−linear polymerization*, elementary particles or monomers combine to form progressively larger polymers. The research of Baryshnikov et al [8] on linear self assembly has resulted in exact results for dimer self assembly, which reduces to an interesting maximal matching problem.

Any implementation of DNA computing is constrained fundamentally by the fact that all basic interactions have known and fixed energy thresholds, and these thresholds are much lower than those in electronic devices. This means that any realistic computational device based on organic structures like DNA is forced to operate at signal-to-noise ratios several orders of magnitude lower than those in electronic computing. Therefore, error correction at the computation stage becomes a necessity. Not surprisingly, recent research on error correction has concentrated on approaches that are analogous, at some level, to the repetition coding of information theory, or the concurrent execution of the same computational algorithm with subsequent comparison of results. Note also that biological computations achieve redundancy at little or no extra cost, as by the inherent virtues of the process, many copies of it are run independently. The work within this circle of ideas will be reviewed shortly. However, in our view, the notion of *pulsing* should be introduced into this paradigm, a concept analogous to checkpointing in computer operation[1]. In our context, described in more detail below, the temperature (or some other parameter) of a self assembly process is pulsed periodically, a method in wide use to grow crystals; the pulse effectively rescues the most recent fault-free state (the current state if there have been no errors since the previous pulse). Clearly, the cost of washing out defective subassemblies must be balanced by a higher speed of these checkpointed computations.

[1] Checkpointing techniques have been in use since the early days of computing; see, e.g., [10].

Fig. 1. The set of four tiles on the left implements the operation $\oplus$. A tile glues to other tiles only if their corner labels match. On the right, operation $0 \oplus 1 = 1$ is performed. The input tiles are preassembled and the correct output tile simply attaches itself to the pattern, effectively obtaining the value of the output bit.

Formally, the now standard *tiling system* introduced and validated as a universal computational framework in [11,12] will be our abstract model, and follows the adaptation to elementary logic units of DNA computing described by Winfree and Rothemund [13,3]. The tile is modeled as shown in Figure 1 as a marked or labeled square. Briefly, in the simplest version, the label values are 0 or 1, and they break down into two input labels on one edge and two output labels on the opposite edge. As illustrated in Figure 1, a computational step consists of one tile bonding to others according to given rules that match input labels of one tile to the output labels of one or two adjacent tiles. Successive bonding of tiles in a self assembly process performs a computation.

Currently, in a typical implementation of this scheme, the tiles are DNA-based molecular structures moving randomly, in solution, and capable of functioning independently and in parallel in a self assembly process. This process results in a crystal-like construct modeled as a two dimensional array whose allowable structural variations correspond to the possible results of a given computation. We emphasize the contrast with classical computing paradigms: the random phenomena of self assembly create a randomness in the time required to perform a given computation.

2 Growth Models

The tiling self assemblies of the last section are growth processes. Through the abstraction described next, the times to grow constructs or patterns can be related to classical theories of particle processes; growth in the latter processes is again subject to rules analogous to those governing the self assembly process of the previous section. An initial set of tiles (the input) is placed along the coordinate axes, and growth proceeds outward in the positive quadrant; the placement of a new tile is allowed only if there are already tiles to the left and below the new tile's position which match labels as before. The left-and-below constraint is equivalent to requiring a newly added tile to bond at both of its input labels. The completion of the computation occurs at the attachment of the upper-right corner tile which can happen only after all other tiles are in place.

The fundamental quantity of interest is the computation time, or equivalently, the time until the final square (represented by the upper right corner square in position (M, N)) is in place. Let $T_{i,j}$ be the time it takes for a tile to land at position (i, j) once the conditions are favorable; that is, once both positions $(i, j-1)$ and $(i-1, j)$ are tiled. In a reference theory for self assembly, it is natural to take the $T_{i,j}$'s as independent exponential random variables with unit means. Let $C_{i,j}$ be the time until the square (i, j) becomes occupied, so that the random completion time of interest is given by $C_{M,N}$.

On discovering the isomorphic relationship between the self assembly process and the totally asymmetric simple exclusion process (TASEP), Baryshnikov et al [1] exploited the results on TASEP behavior in the hydrodynamic limit to show that, as N, M grow to infinity such that M/N tends to a positive constant, one has [14, p. 412]

$$C_{M,N}/(\sqrt{M}+\sqrt{N})^2 \sim 1, \tag{1}$$

a formula quantifying the degree of parallelism in the computation. One can generalize this formula to schemes where the tiles can depart as well (like the schemes described in [15]) with the rates of departures $\rho < 1$, and also to more general shapes D than mere squares. In this model, growth is clearly not monotonic, but still can be mapped to a generalization of TASEP for which similar results are known. Baryshnikov et al [1] proved the following:

Theorem 1. *The time $E_{\lambda\mathcal{D},\rho}$ required to complete computation on a DNA computer of shape $\lambda\mathcal{D}$ with tiles arriving at rate 1 and departing at rate ρ is given by*

$$\lim_{\lambda\to\infty} \lambda^{-1} E_{\lambda\mathcal{D}} = \frac{1}{1-\rho} \sup_{\gamma} \int \left(\sqrt{\frac{d\xi}{dz}} + \sqrt{\frac{d\eta}{dz}} \right)^2 dz. \tag{2}$$

3 Error Correction

Since the early days of computing, various methods have been used to deal with error prone computers, parity checking being a standard one. Checkpointing was a popular method implemented in operating systems, and it is still being used today in high-performance systems. This method creates milestones, or checkpoints, at different locations in the process. All the required information is stored at a checkpoint so that the process can restart from that location without having to perform the work done before that checkpoint. Typically a control mechanism periodically creates checkpoints. When the controlled process fails, it is rolled back to the most recent checkpoint and resumes from that location.

Current developments in tile self-assembly resemble developments in the early computing era, after a Hegelian development cycle. We expect the checkpointing method, being a simple but elegant error-correction technique to become a viable tool in the area, at least until a dramatic change in the underlying chemical technology takes place. The narrow question we address below is how to apply it to DNA tile self-assembly. We briefly review the literature before turning to our new approach.

Alternative approaches. The two most frequent errors in DNA self-assembly are growth errors and nucleation errors. Growth errors occur when a wrong type of tile, an *error* tile, attaches to the lattice; a sublattice that forms with the error tile at its origin will then be corrupt. A nucleation error occurs when only one side of a tile attaches to the lattice, and hence at a wrong position. Thermodynamic controls that slow down growth can be introduced to help ensure the relatively early separation of error tiles.

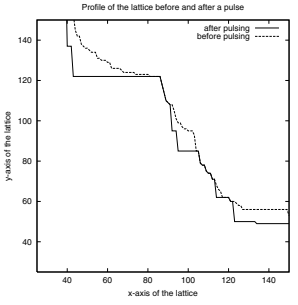
A tile can also be designed to have its own error-correction capability, or a new type of tile that assists the self-assembly process in lowering error rate can be introduced. Several methods for this have been proposed. For example, Winfree and Bekbolatov's Proofreading Tile Set [15] shows that the error rate can be reduced significantly by creating an original Wang Tile using four or nine smaller tiles (2×2 or 3×3) in order to ensure that the small incorrect tiles will fall off before they are assembled to form an incorrect Wang tile. Chen and Goel's Snake Tile Set [16] improves the Proofreading Tile Set by ensuring that the smaller tiles can be assembled only in certain directions.

Reif et al [17] use pads to perform error checking when a new tile is attached to the lattice. Each pad acts as a kind of adhesive, connecting two Wang tiles together, whereas in the original approach the Wang tiles attach to each other. This method allows for redundancy: a single pad mismatch between a tile and its immediate neighbor forces at least one further pad mismatch between a pair of adjacent tiles. This padding method can be extended further to increase the level of redundancy.

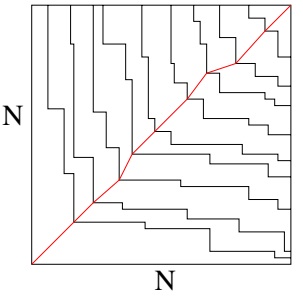
Chen et al's Invadable Tile Set [18] applies the invading capability of the DNA strand to emulate the invasion of a tile. In this model, the tiles are designed so that the correct tile can invade any incorrect tile during the lattice growth process. Fujibayashi and Murata's Layered Tile Model [19] significantly reduces the error rate by using two layers of tiles: the Wang tile layer and the protective tile layer. The protective layer does not allow tiles to attach to the lattice incorrectly. When the attachment is correct, the protective tile releases the rule tile, to which the next tile attaches itself. As one must expect, all methods have one or more shortcomings or costs associated with them, such as prolonged self-assembly times, enlarged lattices, potential instabilities, and failure to deal effectively with both error types.

Modeling checkpointing in DNA self-assembly. We now return to periodic temperature pulsing, whereby pulses remove the defective parts of a crystal; in particular, the hydrogen bonds between improperly attached DNA tiles are broken so that defective substructures can separate from the lattice, thus restarting growth at an earlier fault-free structure. Parameters other than temperature can also be considered in the pulsing approach. Pulsing applied to the DNA tile self-assembly model removes the incorrectly attached tiles from the assembly at a higher rate than the correct ones. More targeted pulsing systems can employ enzymatic or conformational ways to shift the binding energy.

In our model of self-assembly with checkpointing, we consider a lattice of size $N \times N$. (While our results are valid for general shapes, the square lattice



(a) T_A



(b) L_p

Fig. 2. (a) - the profile of DNA tile self-assembly process before and after a pulse. (b) the relationship between the number of layers and the longest increasing subsequence for a Poisson Point Process on the plane. Crystal size: 500×500.

helps focus discussion.) We study the standard growth process described earlier, with the modification that there are two competing populations of tiles to be attached, correct tiles and erroneous tiles. With an appropriate rescaling, the waiting time until a tile attaches at a vacant position is taken to be exponential with mean 1 (all attachment times are independent). Attached tiles are erroneous with probability q.

We call an error tile that attaches to a valid structure a *seed error tile*. Any tile attached to the northeast of the seed error tile is automatically an error tile as it participates in an incorrect computation. (See Figure 2(a)). In our initial analysis we assume that a pulse succeeds in washing out all defective regions of the structure. (See Figure 2(a).)

A growth *stage* consists of a growth period of duration P between consecutive pulses. At the end of one such stage, the locations of the seed error tiles define the boundary of the lattice for the next growth period, on which more tiles will be attached. A growth *layer* is the region of tiles that attach to the lattice during one growth step. The number of stages required to complete the $N \times N$ lattice is the number of pulses or layers required to complete the lattice.

The profiles at the beginning of each growth stage (which we will call the *rectified profiles*, as they are the result of the removal of all erroneous tiles, including the seed tiles) form a Markov process which is clearly significantly more complicated than the growth process without pulsing. Moreover, it is easily seen that these processes cannot be mapped onto 1-dimensional particle processes with local interaction. Hence to evaluate performance, we are forced to resort to asymptotic analysis and simulation studies.

There are several remarks we can make before starting our analysis. Denote the pulsing times by $t_1 < t_2 < \ldots$, and the corresponding rectified profiles, that is the profiles after pulses, by $\mathcal{S}_i$. Clearly, these profiles, from one to the next,

are nondecreasing. In fact, one can describe the evolutions of these profiles using the growth models as in Section 2: the only modification is that if the tile (k, l) is an error seed, then the completion time for this tile becomes

$$C_{k,l} = \min\{t_i > \max(C_{k-1,l}, C_{k,l-1})\}.$$

Using this nondecreasing property, and the standard (in the analysis of algorithms) "principle of deferred decisions", it is not difficult to verify that the collection of the *seed error tiles* over all growth process, form an independent q-Bernoulli sample from the $N \times N$ lattice points.

Small q asymptotics. When q is small, the sample from the planar domain $N \times N$, each dimension rescaled by $\sqrt{q}$ (so that the expected number of seed error tiles in any part of the domain of unit area is 1), approaches, in distribution, the Poisson sample.

The number of pulses required to complete a crystal can be approximated using Hammersley's Process. In our version of this process, we have an underlying Poisson process in two dimensions with samples Ω taken from the square $S = [0, a] \times [0, a]$, with $a = \sqrt{q}N$.

For each $z = (x, y) \in S$ let $n(z)$ be the length of the longest *monotone subsequence* in Ω between $(0, 0)$ and z, that is the maximal length ℓ of a chain of points $(x_1, y_1) < (x_2, y_2) < \ldots < (x_\ell, y_\ell) < (x, y)$, where $(u, v) < (u', v')$ iff $u < u'$ and $v < v'$. See Figure 2(b): The right-hand picture shows layers of points in Ω which can be reached via monotone paths in Ω of length 0 (these are the points adjoining a coordinate axis), 1 (next layer) and so on. A longest path connecting the origin and the point (a, a) is shown as well.

The problem of finding this length is closely related to the famous problem of finding the longest increasing subsequence in a random permutation (Ulam's problem). It turns out that the expected value $\mathbb{E}\ell \sim 2a$, as $a \to \infty$, see, e.g., [20].

This implies immediately information about the asymptotic scaling of the number L_p of pulses necessary to achieve a correct assembly when the interpulse time P is large compared to $q^{-1/2}$. In the limit of small q and large N, this yields a very precise description of this number. Indeed, we have

Theorem 2. *The following assertions hold:*

1. *For any given sample of seed error tiles, L_p is at least the length of the longest increasing subsequence in the sample.*
2. *If $qP^2 \to \infty$, then for samples of seed error tiles, L_p is asymptotically the length of a longest increasing subsequence.*
3. *If $N, qP^2 \to \infty$, and $q \to 0$, then L_p grows as*

$$L_p \sim 2N\sqrt{q}.$$

Sketch of the proof. The first assertion follows immediately from the fact that along any increasing sequence of seed error tiles, the higher error tile is corrected by a pulse coming later than the pulse correcting the previous error tile. Further, the second assertion follows from the fact that if the time P is

large enough, the rectified profiles $\mathcal{S}_i$ coincide with the layers of points reached by the increasing subsequences in Ω having constant length (this can be proved by induction). The last assertion is simply the limit corresponding to the Poisson approximation introduced earlier.

Remarks.

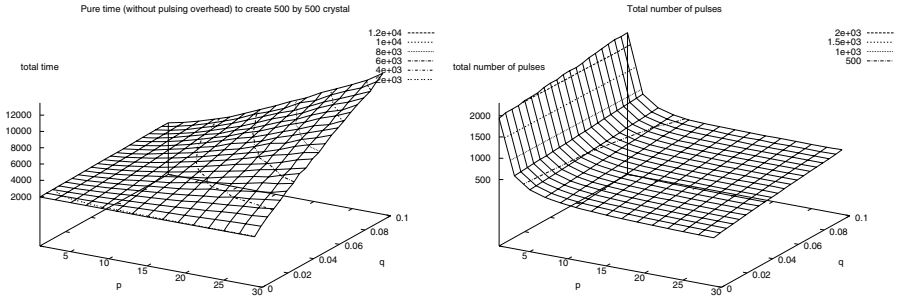
1. Using this result, we can adjust the value of P so as to obtain an estimate of the minimal time required to complete the lattice for any given N, q, and average time p_s taken by a pulse to remove error tiles. More details can be found in [21].
2. The Layered Tile Set [19] can be seen as a variant of our method where P is 1 time unit. When the value of P is very small, the total number of pulses becomes very large because the process pulses once per time unit. As a result, when the value of P is small, the completion time for the formation of the crystal is inversely large. Furthermore, in the case of a high p_s, a very low value of P will not be suitable for the process because of the length of time required during the checkpointing process. Therefore, if P is adjusted appropriately, our checkpointing method will be better than the Layered Tile Set technique.
3. The total growth time in the model is the sum of the interpulse time (plus a constant pulse setup time) times the number of pulses. In the regime described in part 3 of of Theorem 2, this gives the growth rate $\sqrt{q}PN$, which can be arbitrarily close to the obvious lower bound $\Omega(N)$.

Simulation analysis. The total crystal completion time, C, consists of the total time required by tile attachment, C_A, pulsing setup time and the pulsing overhead, C_p. Our simulations determine the effect of P and q on C_A and C_p. The simulation of a 500×500 lattice yielded C_A and C_p for various P and q. The total pulsing overhead time, C_p, is given by $C_p = p_s L_p$ (recall that p_s is the average time taken by a pulse to remove all erroneous tiles).

Our self assembly simulations created more than a million tiles. Developing the simulator was a challenge in itself, given current limits on computer memory. We designed our simulator so that it contains only the information of the crystal, which for our purposes will suffice without having to assign memory space for each tile. Implementation details can be found in [21].

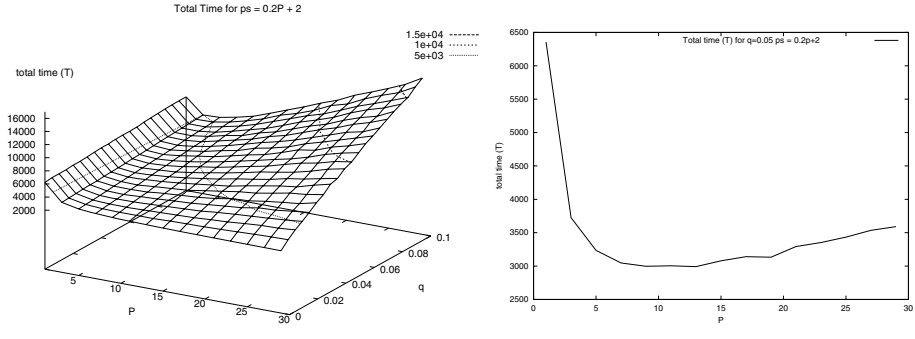
Figure 3 shows the effects of P and q on the performance of self-assembly with pulsing. Since the total time C required to complete the crystal is $C_p + C_A$, we see that C in general has an optimal point for given p_s.

For example, Figure 4(a) shows the total-time surface plot as a function of (P, q). For simplicity, we assume that the time required for each pulse, p_s, is linearly proportional to the growth time, $p_s = 0.2P + 2$, to show how p_s can affect the total time, C. For a given value of q, there is an optimal P that minimizes the total time to complete the self-assembly. Figure 4(b) shows the total time for different values of P with the error probability $q = 0.05$. The figure shows that one obtains the highest over-all lattice growth rate when P is approximately 9 time units.



(a) C_A (b) L_p

Fig. 3. The performance of pulsing for various P and q. Crystal size: 500×500.



(a) Total completion time (b) Total time when $q = 0.05$

Fig. 4. (a) total-time pulsing performance, various (P,q). (b) cross section of (a), q=0.05.

4 Future Directions

We have introduced and analyzed the performance of an error-correcting self assembly pulsing (checkpointing) technique. This comprises the modeling and analysis component of an over-all project that will include the essential experimental component. To fully verify the validity of the method, we propose an experimental setup that produces a simple periodic system capable of errors, as a testbed for examining our proposed error-correcting techniques. It is possible to 'tag' particular elements in 2D crystals; the addition of a DNA hairpin that sticks up from the plane of the array is the simplest tag. Thus, rather than using a single motif to act as a 'tile' in forming the crystal, we can use two or more different tiles, say, an A-tile and a B-tile. An example of this approach is shown for DX molecules in Figure 5.

The experiments we propose here are based on this notion of two differently marked tiles. The idea is to make a 2-D array with two tiles whose sticky ends are distinct, as they are in the DX example above. It is unlikely that individual

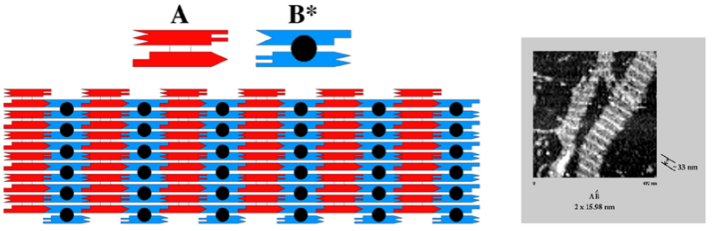


Fig. 5. A Schematic DX Array with 2 Components, A and B*; B* contains a hairpin (the black circle) that protrudes from the plane. Sticky ends are shown geometrically. Note that the A and B* components tile the plane. Their dimensions in this projection are 4×16 nm, so a 32 nm stripe is seen on the AFM image to the right.

4 nm x 16 nm DX tiles can be recognized, but many motifs with large dimensions in both directions exist (see e.g., [22]). The uniqueness of the sticky ends is central to the robust formation of the pattern shown above. Were the sticky ends of the two molecules identical, a random array of protruding features would result, rather than the well-formed stripes shown. It is clear that there must exist some middle ground between unique sticky ends and identical sticky ends. Each tile contains four sticky ends, each of six or so nucleotides. We propose to explore steps on the way from uniqueness to identity (starting from unique) so that we can get a set of tiles that produce an array with a well-defined error rate, low but detectable.

Once we have such a system, it will then be possible to do prototype rescue operations. The basis of these operations will be thermodynamic pulsing, but the study of techniques based on recognition of structural differences in the 2D array, or some combination of the two basic approaches, will be pursued as well.

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