Optimal Design for Active Self-assembly System

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Abstract— Two key issues in stochastic self-assembly are whether the system will converge to the desirable global equilibrium and how quickly it converges. In this paper an optimal self assembly design approach, which guarantees the unique desirable convergence and provides the fastest convergence rate, is proposed for active self-assembly systems. We adopt a Markov chain to model the self-assembly system. Based on the convergence theory of a Markov chain, we solve an optimization problem in which minimizing a certain function involved in the Markov chain results in both maximum yield of the target assemblies at the equilibrium and optimal convergence rate to the desired equilibrium. Several examples are carried out to further illustrate the importance and the effectiveness of the proposed approach.

I. INTRODUCTION

Self-assembly is the spontaneous organization of particles into specific patterns or structures without outside intervention. Self-assembly has vast applications in science and engineering, e.g., in biology [14], [15], electrical engineering [9], computer science, etc.

One category of self-assembly is active self-assembly [1], [9], compared to passive self-assembly. In active self-assembly systems, particles play active roles in that they can somehow decide what reactions to take. Active self-assembly has been developed extensively in robotics, see [3], [6], [9], [12], [16], with the particles' decision to form and sever certain connections programmable. Moreover, according to [9], one possible natural active self-assembly system might be proteins undergoing conformational switching that changes the outcomes of their subsequent interactions.

Other than deterministic self-assembly, which uniquely assembles one supertile (assembly) through deterministic assembly rules, stochastic self-assembly provides probabilistic assembly rules that lead to different *macrostates* with different probabilities. The stochastic self-assembly system can be modeled as a Markov process. The assembly objective is to bring the system to the target macrostate and stay there with high probability. That is, starting from any initial macrostate, given an evolution time longer than the convergence time, the system would reach an output macrostate. Repeating this process for a large number of times, we expect that a high proportion of output macrostates are the objective macrostate. Equivalently, the desirable Markov system should converge to a distribution where the target macrostate is of high probability. This distribution is the desired global equilibrium of the system. Many algorithms have been proposed for this. For example, in [9], [12],

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optimal assembly rules have been developed so that the yield of a desired assembly type at equilibrium is maximized. In [5], [6], assembly rules that require reduced communication burden have been proposed.

Given a system with desirable equilibrium, the most important factor is the equilibration rate, i.e. convergence rate. This has caught much attention recently and been analyzed in different papers [7], [10], [16]. However, only limited work has been pursued, e.g. [2], [11], for designing the optimal convergence rate. In [2], [11], approaches have been proposed to optimize the reaction rates to achieve fast convergence to the specified target state.

In this paper, we propose a novel design of the stochastic active self-assembly system through specifying the optimal parameters in the corresponding Markov system. A Markov decision process (MDP) transformation is applied to come up with an equivalent MDP, thus enabling the convergence to the desirable global equilibrium and the optimization of the convergence rate. We first describe the active self-assembly system, resorting to a Markov chain model. Second, based on the convergence theory of Markov chains, we demonstrate a general optimization problem in which minimizing a certain function involved in the Markov chain would result in the optimal self-assembly rules. Then, we propose an illustrative self-assembly system design. Several examples are followed to further show the importance and the effectiveness of the proposed approach. Different from [2], [11], our approach is completely derived from Markov theories and properties, direct and concise.

II. PROBLEM FORMULATION

A. System Description

We denote the smallest unit in a self-assembly system as a *particle* and assume that the system consists of m identical *particle*s. Let *component* C_i denote the ith type of assembly composed of n_i connected particles and $\mathbf{C} = \{C_1, \cdots, C_I\}$ be the set of all possible I types of components (assemblies), assume that $n_1 \leq n_2 \leq \cdots \leq n_I$. Note it is possible that $C_i \neq C_j$ even if $n_i = n_j$. *Macrostate* $x_k \in X$ describes the number of each type of *component* in the system at time k, where X denotes the set of all the reachable macrostates. Let $N = \dim(X)$, then N is a known number when m is finite. Note here $x_k = [x_k^{(1)}, \cdots, x_k^{(I)}]$ with $\Sigma_{i=1}^I x_k^{(i)} n_i = m$ for any k. As an example, if at time k the system contains only components of type C_1, the macrostate is $x_k = [\ m\ \ 0\ \ \cdots\ \ 0\ ] \in \mathbf{R}^{1 \times I}$. Assume the objective assembly is $C_d \in \mathbf{C}$, the target macrostates thus belong to set $\mathfrak{x} = \{x \in X : \Sigma_{i \neq d} x^{(i)} n_i < n_d\}$, where the *particle*s are assembled to the maximum amount of C_d.

From time k to $k+1$, system transitions from state x_k to x_{k+1} via reactions, i.e. combination/break/decay/null action, among the existing components at time k. Actions taken at time k depend on the current state x_k only. The definition of each action will be given later in this paper.

The above system is a discrete state Markov system on state space X, as pointed out in [9], [12]. When time intervals between reactions are not of concern, the system can be interpreted as a discrete time discrete state Markov chain, see in [5], [6], [7], [10].

Next we give the Markov chain model for the self-assembly system.

B. Markov model for self-assembly

In the following we give the generalized description of the self-assembly system.

Assume at time k the system is at state $x_k = x$. A random variable $F_1(x)$ is sampled so that two assemblies are selected from set $S_x = \{C_i \in \mathbf{C} : x^{(i)} \neq 0\} \times \{C_j \in \mathbf{C} : x^{(j)} > \chi_i(j)\}$, where $\chi_i(j) = 1$ when $i = j$ and 0 otherwise. One of the actions from set $\mathfrak{A} = \{null, decay, act\}$ is taken according to system attributes $F_2(\varepsilon)$, where ε is a parameter set. By $null$ it means that the current state x_k is copied to time $k+1$, by $decay$ it means that the selected components C_i and C_j are decayed to $n_i + n_j$ C_1 components. That is,

$null$:
$$x_{k+1} = x_k$$

$decay$:
$$
\begin{aligned}
x_{k+1}^{(i)} &= x_k^{(i)} - 1 \\
x_{k+1}^{(j)} &= x_k^{(j)} - 1 \\
x_{k+1}^{(1)} &= x_k^{(1)} + n_i + n_j \\
x_{k+1}^{(r)} &= x_k^{(r)}, \text{ for } i \notin \{1, i, j\}
\end{aligned}
\tag{1}
$$

The act includes three possible actions, i.e. $act.comb$, $act.break$, $act.null$ and its outcome is determined by the law $F_3(g, u_1, u_2, C_i, C_j, C_d)$

$act.comb$:
$$
\begin{aligned}
x_{k+1}^{(i)} &= x_k^{(i)} - 1 \\
x_{k+1}^{(j)} &= x_k^{(j)} - 1 \\
C_l &= \mathrm{rand}(u_1(C_i, C_j)), \ n_l = n_i + n_j \\
x_{k+1}^{(l)} &= x_k^{(l)} + 1 \\
x_{k+1}^{(r)} &= x_k^{(r)} \qquad \text{for } r \notin \{i, j, l\}
\end{aligned}
$$

$act.break$:
$$
\begin{aligned}
C_l &= g(C_i, C_j), \ l \in \{i, j\} \\
x_{k+1}^{(l)} &= x_k^{(l)} - 1 \\
\{\mathbf{C}_{lb}, \mathbf{S}_{lb}\} &= \mathrm{rand}(u_2(C_l)) \\
x_{k+1}^{(r)} &= x_k^{(r)} + S_{b_i}, \text{ if } r = b_i, C_{b_i} \in \mathbf{C}_{lb} \\
x_{k+1}^{(r)} &= x_k^{(r)}, \text{ for } C_r \notin \mathbf{C}_{lb}
\end{aligned}
\tag{2}
$$

$act.null$:
$$x_{k+1} = x_k$$

Where $\mathrm{rand}(\bullet)$ is the result of sampling a random variable with the specified distribution, u_1 and u_2 are the random variables with inputs C_i, C_j and C_l respectively, g denotes

the rule for further selecting one component out of $\{C_i, C_j\}$, $\mathbf{C}_{lb}$ denotes the set of all types of smaller size assemblies that the selected component C_l could break into and $\mathbf{S}_{lb}$ describes the number of each resulting assembly. Define $\mathfrak{F}(x) = \hat{\mathfrak{F}}(F_1, F_2, F_3, x, C_d)$ as the output given state x, it is easy to see that when $x_k = x$, the probability of $x_{k+1} = y$ is $p(x, y) \triangleq \Pr(x_{k+1} = y \mid x_k = x) = p(y = \mathfrak{F}(x))$, which relies only on the current state $x_k = x$ and is irrelevant to time k. Thus the system is a stationary Markov chain. As $\dim(X) = N$, the Markov transition matrix P is of $N \times N$ dimension, with entries being $p(x, y)$, $x, y \in X$.

Different forms of policies have been proposed to design $\mathfrak{F}(x)$ [2], [5], [6], [12]. As F_1, F_2 are in general determined by system attributes, the design is in fact focused on F_3. These policies, with or without specifying it, are basically aimed to design the system Markov transition probability so that the Markov system converges to a stationary distribution (equilibrium) where the probability of the desired microstate is maximized. This is the primary goal of self-assembly.

C. Assembly Tasks

With *component* $C_d \in \mathbf{C}$ being the goal assembly and set $\mathfrak{r} = \{x \in X : \Sigma_{i \neq d} x^{(i)} n_i < n_d\}$ in turn the desirable macrostate set, resorting to Markov modeling language, one of our assembly tasks is to design an ergodic Markov system with transition matrix P, whose stationary distribution contains the maximized probability $\Pr(\mathfrak{r})$. Moreover, the convergence rate to the stationary distribution should be the fastest. We interpret our task as "optimal self-assembly system design". Aware that the set $\mathfrak{r}$ can be integrated into one Markov state of the system without loss of generality, we hereby equate $\mathfrak{r}$ to one Markov state.

III. THEORETICAL DEVELOPMENT

A. Convergence Rate for Nonreversible Markov Chain

In order to facilitate the introduction of the optimal self-assembly system design we begin with a brief introduction to the relevant background.

Let X be a finite set with $\dim(X) = N$ and consider a Markov chain on X with transition probabilities $p(x, y) = \Pr(x_{k+1} = y | x_k = x)$, $x, y \in X$. Let P be the $N \times N$ transition matrix with entries $p(x, y)$.

Definition 1: Markov Chain that is irreducible (i.e. the only absorbing set is the whole state set), aperiodic and positive recurrent is an ergodic Markov Chain.

Proposition 1: An ergodic Markov chain P has unique stationary probability distribution π and has only one eigenvalue equals to 1. Moreover, there exists

$$\lim_{k \to \infty} p^k(x, y) = \pi^{(y)} \tag{3}$$

where k is the number of time steps, $p^k(x, y)$ is the (x, y) entry of P^k, $\pi^{(y)}$ is the yth entry of π.

We only discuss ergodic Markov chains here.

The convergence rate is an index on how fast the system converges from the initial distribution to the stationary distribution π. For reversible Markov chains, i.e. chains that

satisfy the detailed balance condition

$$\pi^{(x)}p\left(x,y\right)=\pi^{(y)}p\left(y,x\right)$$

it can be shown that the convergence rate in (3) is bounded by β_1^k, where $\beta_1 = \max\left(\lambda_1, |\lambda_{N-1}|\right)$, λ_1 is the second largest eigenvalue of P and λ_{N-1} the smallest one (recall that an ergodic reversible chain P has all real eigenvalues belonging to the interval $(-1, 1]$). However, for nonreversible chains, the eigenvalues of P are not real. In order to derive the convergence rate in this case, *reversibilization* approaches have been proposed and broadly used to construct a reversible transition matrix $M\left(P\right)$ from P, see, in [4], [8], [13].

Let $\tilde{P} = (\tilde{p}\left(x,y\right))$ be the time reversal of P, i.e.

$$\tilde{p}\left(x,y\right)=p\left(y,x\right)\frac{\pi^{(y)}}{\pi^{(x)}}$$

Then $\tilde{P}$ is an ergodic Markov transition matrix that has the same unique stationary distribution π as P. Define the *multiplicative reversibilization* $M\left(P\right)$ of P by

$$M\left(P\right)=P\tilde{P} \qquad (4)$$

It is easy to see that $M\left(P\right)$ is a reversible Markov transition matrix that also has the same stationary distribution π, see in [4]. Furthermore, the eigenvalues of $M\left(P\right)$ are real and nonnegative, i.e. they belong to the interval $[0, 1]$. Let $\beta_1\left(M\right)$ be the second largest eigenvalue of $M\left(P\right)$. For distributions μ, π define the variation distance

$$\|\mu - \pi\|_{var} = \frac{1}{2}\sum_{x\in X}\left|\mu^{(x)}-\pi^{(x)}\right|$$

where $\mu^{(x)}$, $\pi^{(x)}$ denote one entry that corresponds to state x, i.e. the probability of x, in μ and π respectively. We have the following theorem [4]:

Theorem 2: Let P be an ergodic transition matrix on a finite state space X and let π be its stationary distribution. Then for any initial distribution μ

$$4\left\|\pi_k[\mu]-\pi\right\|_{var}^2 \le \left(\beta_1\left(M\right)\right)^k \sum_{x\in X}\frac{\left(\mu^{(x)}-\pi^{(x)}\right)^2}{\pi^{(x)}} \qquad (5)$$

where $\pi_k[\mu]$ is the distribution of the chain P at time k with initial distribution μ.

Theorem 2 tells us that for a nonreversible Markov chain P, from any initial distribution, the convergence rate of the system to stationary distribution is bounded by $\left(\beta_1\left(M\right)\right)^k$. The smaller $\beta_1\left(M\right)$ is, the faster the system converges to its stationary distribution.

B. Optimal Design for Self-assembly

From Section II-B, we know that the Markov transition probability from macrostate x to macrostate y is $p(x,y) = p(y = \mathfrak{F}(x))$, which is the (x,y) entry of Markov transition matrix $P = P(\mathfrak{F})$. In general, the Markov chain corresponding to a self-assembly system is non-reversible. We thus resort to (4) for the reversibilization and result in $M(\mathfrak{F})$.

Let μ_0 be the initial distribution of the system's macrostate and $\mu_0^{(x)}$ be the probability that the system is in macrostate $x \in X$ at time 0. For example, if system always starts from macrostate $\mathring{x} = [\ m\ \ \ 0\ \ \cdots\ \ 0\ ]$, the system's initial distribution is an impulse distribution with $\mu_0^{(\mathring{x})} = \Pr(\mathring{x}) = 1$, $\mu_0^{(x\neq\mathring{x})} = \Pr(x \neq \mathring{x}) = 0$. We know $\mu_k = \mu_0 P^k$ in turn describes the distribution system's macrostate and $\mu_k^{(y)}$ the probability that the system is in macrostate $y \in X$ at time k. We want to design $\mathfrak{F}$ to fulfill three tasks. First, the system has to be ergodic. Second, the yield of a desired assembly type at equilibrium is maximized. That is, we want to have a stationary distribution π with the component $\pi^{(\mathfrak{r})}$ being maximized. Here $\pi^{(\mathfrak{r})}$ is the probability of the desirable macrostate $\mathfrak{r}$ corresponding to C_d. Third, we want to optimize the system so that it converges to this π with the highest rate. That is, the system evolves to the stationary distribution very fast.

It is not hard to see the *decay* action in (1) guarantees the system to be ergodic. We have the following optimal design rule,

$$\mathfrak{F}_s \quad \in \quad \left\{\mathfrak{F}:\pi^{(\mathfrak{r})} > 0.95 \times \pi^{(\mathfrak{r})}_{\max}\right\} \qquad (6)$$
$$\mathfrak{F}_{opt} \quad = \quad \arg\min_{\mathfrak{F}_s}(\beta_1\left(M(\mathfrak{F}_s)\right))$$

Thus, the system has the fastest convergence rate to the desirable stationary distribution. Note that $\mathfrak{F}_s$ could be a set.

IV. ILLUSTRATIVE SYSTEM DESIGN AND OPTIMIZATION

In this section we design an illustrative active self-assembly system. We remark that the optimal design proposed can be extended to general systems with various self-assembly regulations and properties. We choose the following illustrative system, similar to that in [6], [2], in order to better illustrate the detailed design ideas and logic.

A. Self-assembly Rule

Recall the desirable assembly is C_d. We give the assembly rule here.

When system is at state x, random variable $F_1(x)$ is sampled to select the two components C_i, C_j out of the existing assemblies. Assume without loss of generality throughout that $n_i \le n_j$. Random variable F_2 is sampled to select an action from set $\mathfrak{A} = \{null, decay, act\}$ at every time step. F_1 and F_2 are usually determined by the system attributes. When $F_2 = \{act\}$, the self-assembly policy F_3 is proposed as:

$$\begin{array}{lll} a)\ act.comb & \text{if } n_i + n_j \le n_I \text{ and } d \notin \{i,j\} & \\ b)\ act.break\ C_j & \text{if } n_i + n_j > n_I \text{ and } j \neq d & \\ c)\ act.break\ C_i & \text{if } n_i + n_j > n_I, j = d, i \neq d & \\ d)\ act.null & \text{if } i = j = d & \end{array} \qquad (7)$$

We give more information on *act.comb* and *act.break* rules here. When *act.comb* is taken, the outcome assembly is according to the sampling on random variable $u_1(C_i, C_j)$. u_1 is usually predefined according to the system attributes. We assume here, similar to that in [9], that we can interfere with the *act.break* action through the preprogramming on

every component. Here we interpret C_1, C_2, C_3 as the basic tiles for all assemblies. We assume all the components are appropriately preprogrammed so that when it is subject to $act.break$, it would break according to $\mathbf{C}_{lb} = \{\{$case 1: all $C_1\}$, $\{$case 2: maximum number of C_2 with the remainder being $C_1\}$, $\{$case 3: maximum number of C_3 with the reminder being $C_1\}\}$ with probabilities b_1, b_2, b_3 subject to $b_1+b_2+b_3 = 1$.

As we have mentioned before, F_1, F_2 are predefined by the system, so clearly the only parameters left for us to design are b_1, b_2, b_3 in function u_2 such that sampling u_2 gives the outcome of different combination of C_1, C_2, C_3 assemblies according to $\mathbf{b}$ for each input C_l in (2). Let $\mathbf{b} = [b_1, b_2, b_3]$, the parameterized Markov matrix $P(\mathfrak{F})$ thus turns into $P(\mathbf{b})$.

B. Approach

Our optimal design approach is given as the following:

- Given a self-assembly system $\{m, \mathbf{C}, \mathfrak{F}, C_d\}$ with initial state of m number of C_1 assemblies, i.e. macrostate $\mathring{x}$, calculate Markov transition matrix P.
  - First, calculate only the "nonbreak" transition matrix P_1. That is, P_1 denotes the system that involves only $act.comb$, $null$ and $decay$ actions, with $act.break$ being substituted by $null$. P_1 can be easily calculated by Monte Carlo simulations according to action rules (7) and system attributes.
  - Second, write down the "break" probability matrix $P_2(\mathbf{b})$ analytically, with only $act.break$ involved.
  - Third, analytically calculate the whole transition matrix

$$P(\mathbf{b}) = P_1 + (\Lambda_{P_1} - p_{null}I)(P_2(\mathbf{b}) - I)$$

  where p_{null} is the null rate of the system, I is an identity matrix, Λ_{P_1} is a diagonal matrix with the diagonal entries of P_1.

- Vary the values of $\mathbf{b}$ under the constraint $b_1+b_2+b_3 = 1$ to get different $P(\mathbf{b})$ matrices. Using $\mathbf{b}$ notation, and let $\pi_{\max}^{(\mathfrak{r})} = \max_{\mathbf{b}}(\pi^{(\mathfrak{r})}(\mathbf{b}))$, equation (6) now becomes:

$$\begin{aligned} \mathbf{b}_s &= \{\mathbf{b} : \pi^{(\mathfrak{r})}(\mathbf{b}) \geq 0.95 \times \pi_{\max}^{(\mathfrak{r})}\} \\ \mathbf{b}_{opt} &= \arg\min_{\mathbf{b}_s}(\beta_1(M(P(\mathbf{b}_s)))), \ \mathbf{b}_{opt} < 1 \end{aligned} \quad (8)$$

V. SIMULATION

We adopt the self-assembly system used in [9], i.e., system with programmable triangular particles (parts), in order to illustrate our optimization procedure. Consider a system with $m = 8$ and $I = 11$, with set $\mathbf{C}$ as shown in Figure 1.

Note here the assembly pool is slightly different from that in [9] since we have removed several duplicated assemblies here. That is, some assemblies that are the same in shape but different in orientation have been removed. The combination chart in Table 1 lists all the possible combinations of two selected assemblies in this system. That is, all the possible output of $rand(u_1)$.
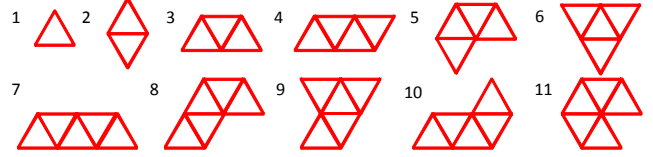

Figure 1. All possible assemblies

Component	Component	Products
C_1	C_1	$\{C_2\}$
C_1	C_2	$\{C_3\}$
C_1	C_3	$\{C_4, C_5, C_6\}$
C_1	C_4	$\{C_7, C_8, C_9, C_{10}\}$
C_1	C_5	$\{C_8, C_9, C_{10}, C_{11}\}$
C_1	C_6	$\{C_9\}$
C_2	C_2	$\{C_4, C_5\}$
C_2	C_3	$\{C_7, C_8, C_9, C_{10}, C_{11}\}$

Table 1. List of possible combinations

As for the function $\mathfrak{F}$, at any time step k, we take F_1 uniform over all the existing assemblies. F_2 is assumed to have an outcome with 5% $null$, 0.05% $decay$ and otherwise act. Take u_1 uniform over the appropriate combinations and determine g according to the $act.break$ rule in (7). Moreover, $u_2(\mathbf{b})$ is a random variable whose distribution is parameterized by $\mathbf{b}$.

As preparation, first we count all possible *macrostate*s in the given system; in this example there are $N = 43$ possible *macrostate*s. From each macrostate, we generate 10000 random samples and calculate the one-step forward transition probabilities p_1 according to $\hat{\mathfrak{F}}(F_1, F_2, F_3, x, C_d)$ with Monte Carlo method. We then analytically calculate the complete transition matrix $P(\mathbf{b})$ according to the proposed approach in Sec. IV-B, here $\mathbf{b}$ is subject to $b_1+b_2+b_3 = 1$ and $0 \leq b_1, b_2, b_3 \leq 1$. Then, by varying the value of $\mathbf{b}$ as well as applying the reversibilization approach, we obtain $\beta_1(M(P(\mathbf{b})))$ and stationary distribution $\pi(\mathbf{b})$. The set $\mathbf{b}$ that satisfies the optimal rule (8) is chosen as the optimal solution $\mathbf{b}_{opt}$.

A. Case Study

1) Case 1 $C_d = C_4$: Assume that the system always starts from the initial macrostate with 8 C_1 components and evolves. That is, the system has an initial distribution μ_0, where $\mu_0^{(\mathring{x})} = \Pr(\mathring{x}) = 1$, $\mu_0^{(x \neq \mathring{x})} = \Pr(x \neq \mathring{x}) = 0$, $\mathring{x} = [\ 8 \quad 0 \quad \cdots \quad 0\ ]$. The desirable macrostate set $\mathfrak{r} = \{$macrostate with two C_4, which is macrostate No. 39$\}$. The $\mathbf{b}_{opt}$ is calculated to be $[0, 1, 0]$ where $\beta_1 = 0.9758$ is of the optimal value according to (8). That is, $[0, 1, 0]$ provides the best combination of small β_1 and large $\pi^{(39)}$ over all possible $\mathbf{b}$. The stationary distribution π and the distribution evolution of the system are given in Figure 2a. Note in Figure 2a-right and similar figures later, we can tell that the system reaches the stationary distribution when the composition of all probabilities, corresponding to different macrostates in the distribution, does not change in time any more.

In order to clarify the difference between different **b** values, we list here also some nonoptimal **b** and their corresponding eigenvalues of $M(P)$.

$$\mathbf{b} = [1, \ 0, \ 0] \quad \rightarrow \quad \beta_1 = 0.9864$$
$$\mathbf{b} = [0, \ 0, \ 1] \quad \rightarrow \quad \beta_1 = 0.9995$$

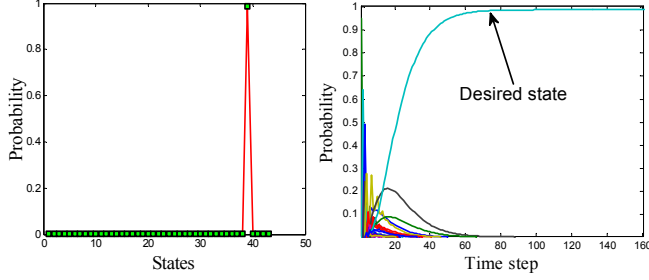We show in Figures 2b-2c the system performance given different values of **b**.
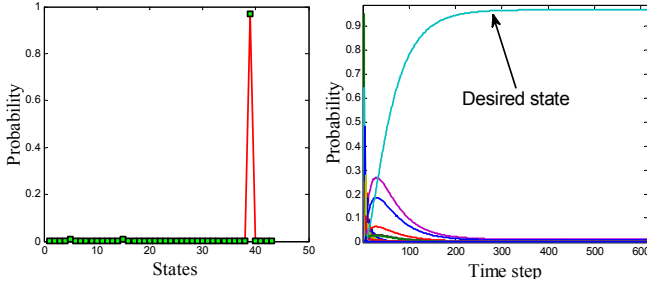

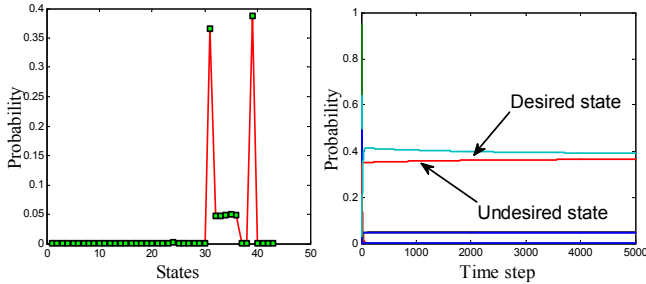Figure 2a. $\mathbf{b}_{opt}$=[0,1,0]

Figure 2b. **b**=[1,0,0]

Figure 2c. **b**=[0,0,1]

left - stationary distribution, right - distribution evolution

With the optimal set $\mathbf{b}_{opt} = [0, 1, 0]$, we see the system has a desirable stationary distribution, as shown in Figure 2a-left, where the probability for the desirable macrostate is 0.9880, close to 1. That means after reaching the stationary distribution, the system stays at the desirable macrostate almost for sure. In Figure 2a-right, we show the probability composition from all macrostates at different time steps, i.e. the evolution of distribution. We can see that the probability of the desirable macrostate reaches its stationary value quickly, within 150 time steps. While when $\mathbf{b} = [1 \ 0 \ 0]$ the system has a suboptimal stationary distribution with probability corresponding to the 39th macrostate being 0.9676 and it converges at around 500 time steps, as shown in Figure 2b. Even worse, if $\mathbf{b} = [0, 0, 1]$, the system converges

to the stationary distribution shown in Figure 2c-left, where the undesirable 31th macrostate $x = [0, 2, 0, 0, 0, 0, 1, 0]$, i.e. macrostate with 2 C_2 and 1 C_7 assemblies, has comparable probability to that of the desired 39th macrostate. From Figure 2c-right we can tell the convergence time is more than 5000 time steps. From the above scenarios, observing the relationship between convergence rate and β_1, we verify Theorem 2 in that the smaller β_1 is, the faster the convergence rate of the system is.
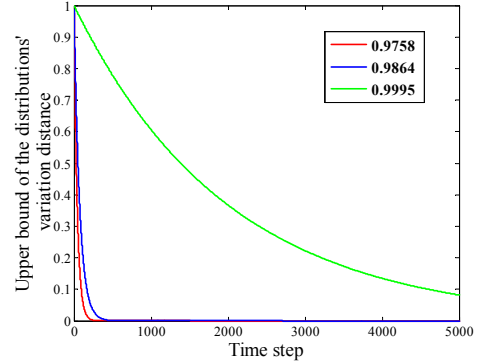

Figure 3. Upper bound of the variation distance between π_k and stationary distribution, over different value of β_1.

We show in Figure 3 the sensitivity of the upper bound of the variation distance between π_k, the system distribution at time step k, and the stationary distribution π towards β_1, i.e. the upper bound of the left side in the equation in Theorem 2. It can be seen that when $\beta_1 = 0.9758$, the upper bound of the variation distance decreases to a negligible value at time step 150, while when $\beta_1 = 0.9864$, the upper bound decreases to a trivial value at time step 500. In contrast, when $\beta_1 = 0.9995$, even at time step 5000, the upper bound still has a considerable value. This explains the convergence results we had earlier in Figures 2a-2c.

Considering the relationship between the conformation of the objective assembly C_d and optimal set $\mathbf{b}_{opt}$, it is easy to see that $C_2 + C_2 \rightarrow C_4$, while neither of $C_1 + C_2$, $C_3 + C_3$, $C_1 + C_1$, $C_3 + C_2$ would give C_4 directly. Thus, it makes sense that maximizing the number of C_2 results in the fastest convergence to the maximum yield of C_4.

2) Case 2 $C_d = C_6$: Similar to Case 1, assume the system always starts from the initial macrostate with 8 C_1 components and evolves. Now the desirable macrostate set $\mathfrak{x} = \{$macrostate with two C_6, i.e. macrostate No. 42$\}$. According to (8), $\mathbf{b}_{opt}$ is calculated to be

$$\mathbf{b}_{opt} = [0.415, \ 0.005, \ 0.580] \quad \rightarrow \quad \beta_1 = 0.9970$$

The corresponding stationary distribution π and the distribution evolution of the system are shown in Figure 4.

We also list here a nonoptimal **b** value with corresponding eigenvalue of $M(P)$ and stationary distribution π, shown in Figure 5 for comparison purposes.

$$\mathbf{b} = [0, \ 0, \ 1] \quad \rightarrow \beta_1 = 0.9991$$

We can see that in this case the optimal set $\mathbf{b}_{opt}$ is calculated to be $\mathbf{b}_{opt}=[0.415,\ 0.005,\ 0.580]$, where the probability for the desirable macrostate in the stationary distribution is the highest, 0.825, as shown in Figure 4-left. In Figure 4-right, we see the system converges to the stationary distribution at around 3000 time steps. While when $\mathbf{b} = [0, 0, 1]$ the system converges to a stationary distribution where the undesirable macrostate, No. 31, has higher probability than that of the 42th macrostate, as shown in Figure 5.
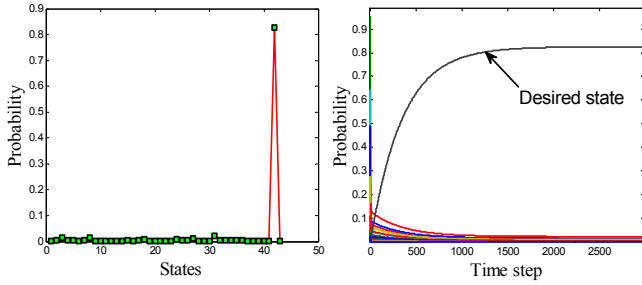

Figure 4. $\mathbf{b}_{opt}=[0.415, 0.005, 0.580]$

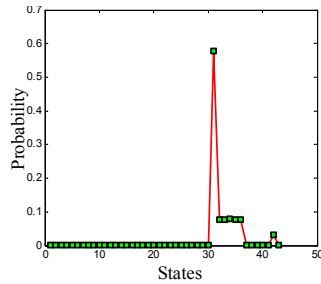left - stationary distribution, right - convergence time

Figure 5. Stationary distribution for $\mathbf{b}= [0, 0, 1]$

Similar to the analysis in Case 1 and observing the conformation of $C_d = C_6$, we know that $C_1 + C_3 \rightarrow C_6$, while neither of $C_1 + C_2$, $C_2 + C_3$, $C_2 + C_2$ would give C_6 directly. Thus, designing $\mathbf{b}$ to balance the number of C_1 and C_3 should result in the fastest convergence to the maximum yield of C_6.

VI. CONCLUSION AND FUTURE WORK

In this paper an optimal design approach for active self-assembly is proposed, based on the convergence theory of Markov chains. The proposed approach serves two purposes. First, it guarantees that the system converges to the global equilibrium with desirable assembly compositions. Second, it provides the fastest convergence rate. An illustrative self-assembly system design is proposed to verify the developed scheme. By the simulation examples we learned the impact of parameter set $\mathbf{b}$ on the convergence result. If $\mathbf{b}$ was selected randomly, the system might converge to an undesirable global equilibrium, i.e. the system is highly inclined to end up in undesired assemblies. With our optimized selection of the $\mathbf{b}$ set, the fastest convergence rate to the desirable equilibrium is attained.

As it is known, the Markov chain modeling in self-assembly is subject to the curse of dimensionality of the system states. Thus, although the proposed scheme is quite useful for designing small systems or understanding fragments of larger systems, its complexity is an issue. For example, for a system with m particles and $O(m)$ component types, there are $N \sim O(2^m)$ macrostates, which introduces into the analysis an $N \times N$ Markov matrix. In the future, we plan to adjust our design to larger systems by reducing the system dimension. In this case, instead of sticking to the original optimal design, we might find a state reduction approach or an approximation of the optimization that offers the best trade-off between calculation complexity and accuracy.

REFERENCES

[1] D. Arbuckle and A. Requicha, "Active self-assembly," in *Proceedings of the IEEE International Conference on Robotics and Automation, New Orleans*, 2004, pp. 896–901.
[2] S. Berman, A. H. M. A. Hsieh, and V. Kumar, "Optimized stochastic policies for task allocation in swarms of robots," *IEEE Trans. on Robotics*, vol. 25(4), pp. 927–937, August, 2008.
[3] Y. Brun and D. Reishus, "Path finding in the tile assembly model," *Theoretical Computer Science*, vol. 410, pp. 1461–1472, 2009.
[4] J. Fill, "Eigenvalue bounds on convergence to stationarity for nonreversible Markov chains, with an application to the exclusion process," *The Annals of Applied Probability*, vol. 1, pp. 62–87, 1991.
[5] M. Fox, "Stochastic self-assembly," *Master thesis, Georgia Institute of Technology*, 2010.
[6] M. Fox and J. Shamma, "Communication, convergence, and stochastic stability in self-assembly," in *Proceedings of the 49th IEEE CDC*, 2010, pp. 7245–7250.
[7] S. Greenberg and D. Randall, "Convergence rates of Markov chains for some self-assembly and non-saturated Ising models," *Theoretical Computer Science*, vol. 410, pp. 1417–1427, 2008.
[8] O. Junge, J. Marsden, and I. Mezic, "Uncertainty in the dynamics of conservative maps," in *Proceedings of the 43rd IEEE CDC*, 2004, pp. 2225–2230.
[9] E. Klavins, S. Burden, and N. Napp, "Optimal rules for programmed stochastic self-assembly," in *Proceedings of Robotics: Science and Systems II*, 2006, pp. 9–16.
[10] U. Majumder, J. Reif, and S. Sahu, "Stochastic analysis of reversible self-assembly," *Journal of Theoretical and Computational Nanoscience*, vol. 5(7), pp. 1289–1305, July 2008.
[11] L. Matthey, S. Berman, and V. Kumar, "Strategies for a swarm robotic assembly system," in *Proceedings of the IEEE International Conference on Robotics and Automation*, May, 2009, pp. 1953–1958.
[12] N. Napp, S. Burden, and E. Klavins, "The statistical dynamics of programmed self-assembly," in *Proceedings of the 2006 IEEE International Conference on Robotics and Automation*, 2006, pp. 1469–1476.
[13] T. Runolfsson and Y. Ma, "Model reduction of nonreversible Markov Chains," in *Proceedings of the 46th IEEE CDC*, 2007, pp. 3739–3744.
[14] S.C.Harvey, A.S.Petrov, B. Devkota, and M.B.Boz, "Viral assembly: a molecular modeling perspective," *Physical Chemistry Chemical Physics*, vol. 11, pp. 10 553–10 564, 2009.
[15] E. Winfree, "Algorithmic self-assembly of DNA," *PhD thesis, California Institute of Technology*, 1998.
[16] C.-H. Yu and R. Nagpal, "Sensing-based shape formation on modular multi-robot systems: A theoretical study," in *Proceedings of the International Conference on Autonomous Agents and Multi-Agent Systems*, 2008, pp. 71–78.