A Study of Yield Predictions for a Model of Homogeneous Self-Assembling Components

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Abstract: Self-assembly of homogeneous components has the advantage of being a decentralised and highly parallel method for assembling multiple target structures, and is ideal for effective large-scale manufacturing. Yet assembly yield may be negatively affected by the formation of incompatible substructures that prevent the formation of complete target structures. In this work we present physical and theoretical analysis of a simple magnetomechanical self-assembling systems exhibiting the problem of incompatible substructures in the formation of closed circular target structures out of eight homogeneous components. The assembly yield of physical experiments from 8 to 40 components is compared with the predictions of a computational model, and the model is found to accurately predict both the mean and standard deviation of the experimental yield.

Keywords: incompatible substructures, reaction networks, self-assembly, yield problem.

1. INTRODUCTION

Self-assembly is the unsupervised formation of ordered structures from disordered components [11]. Self-assembly not only underlies the formation of many natural systems, foremostly crystals and proteins; recently, it has been increasingly utilised for the bottom-up construction of artificial systems [12]. Although self-assembling system are autonomous and decentralised, their emergent behaviours can be engineered through the design of both components and of their interactions. Of specific interest here is the class of self-assembling systems of discrete, passive components that form multiple *target structures* in parallel using geometry to control the growth process.

If the components of such a self-assembling system belong to a homogeneous set, parallelism is high and time-to-completion short, as any available component can in principle be used for the assembly of target structures. However, a drawback of such systems is the formation of *incompatible substructures*: partially formed target structures that cannot completely assemble [3]. If the assembly process is irreversible, incompatible substructures prevent the formation of target structures, thus reducing the system's assembly yield. An example comes from the division of a hexagonal target structure along its axes of symmetry into 6 triangular and homogeneous components, as first proposed by Hosokawa et al. [3]. When exactly 6 triangles are available, the self-assembly yield is theoretically 100% since the components can assemble correctly in only one way. However, 2 times 6 triangles may form e.g. 3 substructures of 4 components each. As geometry of these substructures prevents their assembly into target structures, the resulting yield is 0%.

In this work we present an empirical and theoretical study of the impact of incompatible structures on the self-assembly of target structures. We conducted exhaustive self-assembly experiments on a magnetomechanical system closely reproducing the system introduced in [3]. From each set of experiments conducted with a fixed number of components, we recorded the frequency of formation of target structures. We thus present the trend of probabilities of the formation of target structures out of 33 sets of self-assembly experiments (with 8 up to 40 components). We then compare the data of the physical experiments with corresponding predictions drawn from a theoretical model that abstracts physical systems as a multiset of integers. We finally discuss our results and outline future developments.

2. BACKGROUND

Self-assembly was originally studied in the context of molecular chemistry, biology and material sciences [12]. Seminal work on a centimetre-scale artificial self-replicating system was carried out by Penrose and Penrose in the 1950s, whereby a *seed* configuration was replicated by initially disordered components [9]. It was not until the late 1990s that macroscopic self-assembling systems emerged as a research field [2]. Whitesides et al. studied self-assembly in many artificial systems and across different scales [11].

Hosokawa et al. designed a self-assembling system where six triangular components self-assembled into a hexagon [3]. In their experiments, Hosokawa et al. encountered the incompatible substructure problem, and first described the steady-state distribution of structures assembled in their system through the formalism of chemical reaction kinetics. The authors also suggested the use of a seed, echoing Penrose and Penrose. The seed could impose a predetermined pathway for the self-assembly of the components that avoided the formation of incompatible substructures. To this end, they conceived, but never realised, a seeding component that could switch conformation upon assembly. Saitou later studied the 1D

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self-assembly of copies of conformational switches, and showed through rate equations that the use of conformational switches can increase self-assembly yield [10].

To eliminate the incompatible substructure problem, Bhalla et al. modified the morphology of the target structures and structured the self-assembly process by introducing geometrical seeding components [1]. The authors used a heterogeneous set of components, so that the target structures self-assembled by ray components filling shape-complementary slots in templating disc components. The assembly was thus directed from the inside-out to exclude incompatible substructures.

An alternative to constraining the kinetics of self-assembly through seeds is the use of reversible inter-component bonds, avoiding permanent incompatible substructures. Miyashita et al. studied the effect of stochasticity and morphology on the yield of a reversible self-assembling system [8]. The authors used self-propelled components similar in shape to those of Hosokawa et al. but with reversible bonds, achieving improved yield and validating their findings using kinetic rate calculations.

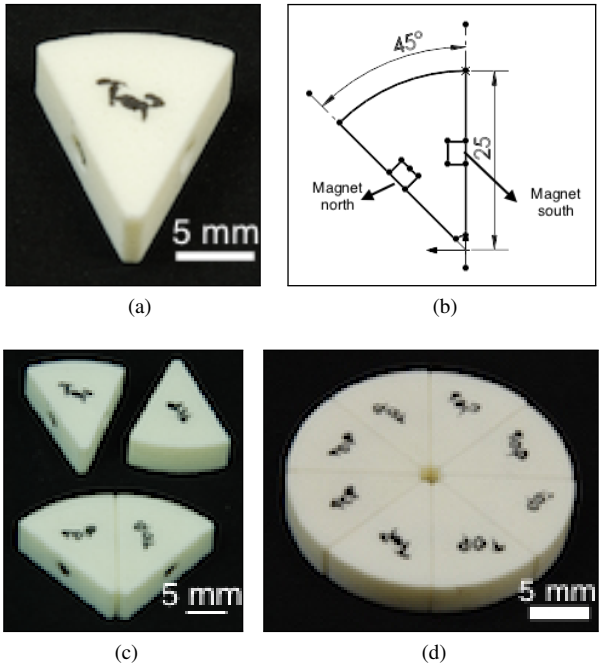
In addition to possible solutions, Hosokawa et al. [3] and Miyashita et al. [8] presented analyses of the yield problem in parallel self-assembly as affected by incompatible substructures. They compared yield data from physical experiments with data predicted by chemical-reaction-based models. However, the analyses in both studies considered only a fixed number of components and, in the case of Miyashita et al., only mean assembly yield. We provide the missing comparison of both mean and variance of yield across system sizes in this work.

Klavins et al. built an upscaled, mechatronic version of the triangular components of Hosokawa et al. called *programmable parts* [4]. They also developed target-specific and automatically-derivable sets of software-encoded rules based on graph grammars. The rule sets, stored in each part's onboard memory, distributedly enforced the parallel self-assembly of the hexagonal target structures without incurring in the formation of incompatible substructures. After bonding with neighbouring parts, the parts themselves could judge the consistency of the bond with the formation of a target structure, and acted consequently, possibly reversing the bond.

Several other works have used changes in the environmental conditions to stir the self-assembly process away from the appearance of incompatible substructures. In the system of Mao et al. [5], mutually exclusive sets of capillary bonds were enabled for differing densities of the liquid over which the components floated. Variations in liquid density consequently induced changes in the morphology of the assembled structures. In the fluidic systems studied by Mermoud et al. [7] at centimetric scale and by Mastrangeli et al. [6] at micrometric scale, the turbulence of the liquid hosting the components was exploited to induce stochastic reconfigurations of the self-assembled structures. In these systems, substructures incompatible with predefined target structures were automatically detected and dissolved under high turbulence conditions, ultimately resulting in the exclusive formation of target structures.

3. PHYSICAL MODEL

To study the trend of the probability of target structure formation, we designed a simple two-dimensional self-assembling system, conceptually similar to that used by Hosokawa et al. [3]. In our system (Figure 1), passive components are orbitally stirred inside a container and mutually latch by means of embedded magnets.



(a) (b)

(c) (d)

Fig. 1: (a) A 3D printed component. (b) Dimensions of the components and magnet orientation (c) Incorrect (top) and correct (bottom) pairwise combinations of components. (d) A target structure.

In the system, circular *target structures* as seen in Figure 1d of radius $25\ mm$ and thickness $7\ mm$ are composed of 8 homogeneous 3D-printed components having the shape of circular sectors spanning an angle of $45°$ (Figure 1a, 1b). Each component has a NdFeB magnet (N48 magnetisation, strength of approximately 210 g) embedded in the middle of each of its main straight edges. The weight of a single component with the embedded magnets is $1.77\ g$. The magnets in each component are arranged such that the left and right edges of all components have opposite polarities (Figure 1b). This magnetic configuration leads to the pairwise latching of components oriented along edges of opposite magnetic polarity and with sufficient proximity. The permanent assembly of components in wrong mutual orientations (Figure 1c) is excluded because the weak magnetic bonds formed in such configurations are easily released by the kinetic energy of impact, either with other components or with the borders of the container (seen in Figure 2).

Together, the magnetic constraints and the geometry of the components make the assembly of the components into a circular geometry the most energetically favourable

pathway. Target structures correspond to global minima in the energy landscape. However, other local minima correspond to incompatible substructures (Figure 2), whose assembly is sterically hindered.

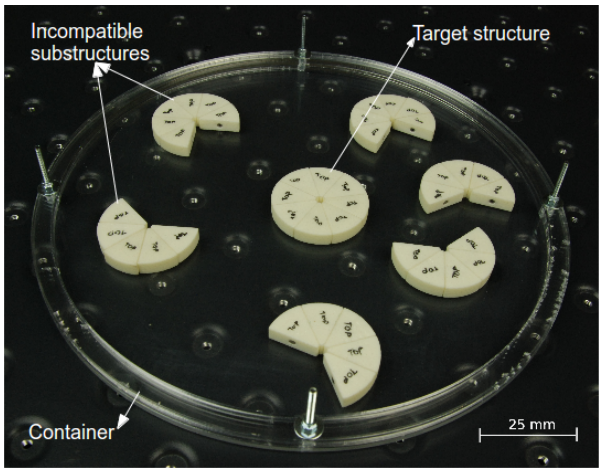


Fig. 2: A trial of a 5 target structures experiment, wherein only one target structure was formed and all remaining substructures are incompatible.

8 components per target structure, compared to 6 as in the original work of Hosokawa et al., were chosen to increase the number of possible substructures that can be formed, thus increasing the complexity of the system dynamics and the probability of assembly of incompatible substructures. We were unable to use target structures with higher number of components due to fabrication issues as well as practical difficulties in correctly handling the components during experiments.

The experiments were carried out in a circular container with an inner diameter of 250 mm as shown in Figure 2. The ratio between the diameter of the container and that of the target structures, as well as the maximum number of components used in a single experiment, were chosen to allow free motion of the components within the boundaries of the container while keeping assembly time reasonable (on the order of 10 minutes).

Kinetic energy was imparted to the components by fastening the container to an orbital shaker (Excella E5). We chose to use circles as target structures because the curved edge of each sector allowed the components to rotate after impacting the border of the container, thereby improving component mixing. The rotation frequency of the shaker (300 rpm in all experiments) was chosen to maximise component and substructure interaction rates while avoiding structure breakage due to impacts.

A set of 11 trials were conducted for each given number of components in the system. The number of components was incremented from 8 to 40, for a total of 363 trials. Each trial begins with random manual positioning of approximately equidistant single components in the container. The components are shook until the system reaches an *absorbing state* consisting exclusively of full target structures and incompatible substructures. At the end of each trial, the distribution of assembled structures is recorded by photograph.

4. MODEL

The model we developed is based on the one of Hosokawa et al. [3]. This model abstracts physical systems of components (n in total) and substructures as a multiset of integers, each integer representing a substructure consisting of a corresponding number of components. The state of a theoretical system is represented as a vector $\vec{x} = \langle x_1, x_2, \ldots, x_m \rangle$, where $x_i \geq 0$ is the number of substructures of i components and m is the number of components in a target structure (in this work, $m = 8$).

A system evolves by combinations of compatible substructure pairs, equivalent to changing $\vec{x}$ by decrementing x_i, x_j and incrementing x_{i+j} for some i, j with $i + j \leq m$. The probability of a specific combination occurring next is the product of the probability that such a pair of structures meets and the probability that they are at relative orientations that allow them to combine. The first probability is the product of their concentrations in the system. The second probability is determined by the likelihood that two such substructures oriented randomly in the plane have two faces that can attach and completely see each other (for more details, see Appendix A of [3]).

Given this, the probabilistic evolution of the system is captured by a system of differential equations with a variable for each possible state of a system, whose rate of change is determined by the probabilities of possible prior states (before the last combination of substructures) multiplied by the probability of the corresponding combinations minus the probabilities of possible subsequent states multiplied by the probability of the state.

For fixed n, the distribution of final states is found by computing the evolution of this system of equations until the probabilities of system states with remaining combinations of substructures is less than 0.001 in total.

5. RESULTS

Figure 3 shows both average and standard deviation of the assembly yield for both the physical system and the model of Hosokawa et al. The data are displayed for n ranging from 8 to 40. While the experimental data have significant dispersion due to the relatively small sample size, overall the model can be seen to qualitatively predict both the average and standard deviation of the yield for systems across the entire range of values of n.

6. DISCUSSION AND FUTURE WORK

The work presented here is a preliminary study of the ability of the model of Hosokawa et al. to describe self-assembly yield for physical systems richer in possible substructures, dynamics and number of components than the original system realized by the authors. The results qualitatively demonstrate the model's predictive power over self-assembly outcomes for such extended systems.

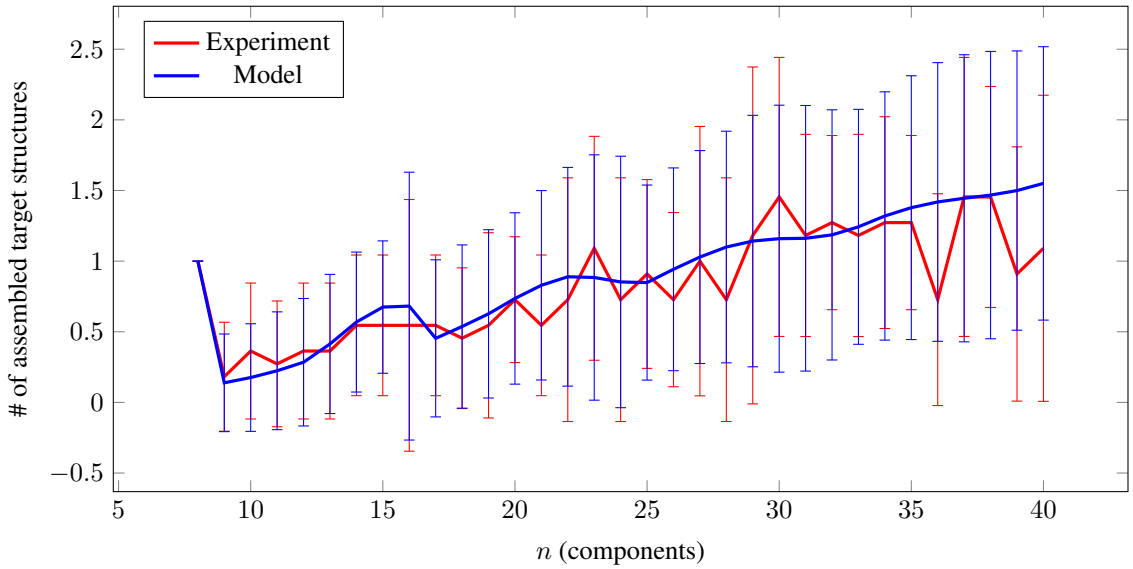


Fig. 3: The mean (lines) and standard deviation (bars) of target structure yield observed in the physical experiments and predicted by the model of Hosokawa et al.

While these initial results are very encouraging, several aspects of the model and of its predictions remain to be further studied. We plan to perform additional trials to increase the confidence in the observed results. A more robust comparison metric may also improve data analysis. Moreover, it remains unclear whether not only the outcomes, but also the dynamics of the physical systems are captured by the model. We are currently constructing an optical tracking setup to address this question in our self-assembling system.

Finally, a central obstacle in understanding and even computing the predictions of the model lies in the analysis of the system of differential equations it contains. The number of variables in these systems grows proportional to n^7. The consequent computational cost may have prevented Hosokawa et al. from performing the analysis across a large range of values of n, as done hereby, and currently prevents us from further extending this range. Finding a closed form description for the evolution of these systems could make such analysis feasible, and possibly give insights into the mechanisms governing the behaviour of the physical systems.

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