Feedback Control of Particle Positions using Electric Fields and Chemical Gradients

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Abstract

Programmable self-assembly is a promising route to new biomaterial and metamaterial synthesis methods. An important step towards programmable self-assembly is moving component parts to desired locations. We do this by designing and simulating a controller that modifies an external field to move particles on desired paths. We first design a controller using electric fields by modifying established methods, then we apply the insights gained from the electric field controller to a chemical controller that uses model predictive control.

Introduction

Programmable self-assembly is a long-standing goal of nanotechnology. Current technology for creating materials using self-assembly is limited, but biological systems use it regularly for tasks such as spontaneous healing of wounds and tissue growth and development. The potential applications of programmable self-assembly systems are numerous and include tissue engineering, synthetic biomaterials, and metamaterials applications. Of particular interest would be the ability to use programmable self-assembly to generate tissue or other organic material that would be essential for the health and well-being of astronauts on long-term space missions.

Current methods to assemble structures from their components include top-down and bottom-up approaches. Top-down approaches use external fields or forces to place building blocks one by one. These approaches are versatile but difficult to scale up. Bottom-up (self-assembly) approaches instead allow the natural interactions between particles to create a stable structure, which is usually an equilibrium structure. This approach is more scalable, but it is difficult to design systems with useful equilibrium structures. Recent efforts have also been able to combine the two approaches, using for example a top-down method to pattern a surface onto which particles assemble in a bottom-up fashion. Our approach is to create a new combination of top-down and bottom-up approaches inspired by biological systems.

For many systems, self-assembly ends when the system reaches equilibrium. However, biological systems are an example of dynamic, or non-equilibrium, self-assembly. Biological agents use a complex network of chemical signals and feedback control that is not completely understood. Our objective is to devise and simulate techniques for feedback control of position using simpler chemical gradients.

We combine methods currently used in both macro-scale systems and micro-scale systems. On the macro-scale, unmanned aerial vehicles (UAVs) use an equation called a Guidance Vector Field (GVF) to lead them on stable paths around a target. A GVF takes in the vehicle's current position and outputs the velocity vector needed to guide the vehicle to the desired path. The same equation can be used for small particles to find a target velocity at any position.

Small (nanometer to micrometer diameter) particles move in electric and chemical fields.In 2006, Chaudhary and Shapiro[1] published a method of steering multiple particles using electric fields. By modifying their method, we took

the velocity outputted by a GVF and calculated the strengths of electrical actuators needed to produce the necessary velocity at any time. Once we were able to simulate particle control using electrical actuators, we used the similarity between electric fields and chemical gradients to apply these methods to chemical actuators. The result was a simulation of particle feedback control using chemical gradients. While this is a top-down result, it is an important step towards future programmable non-equilibrium self-assembly.

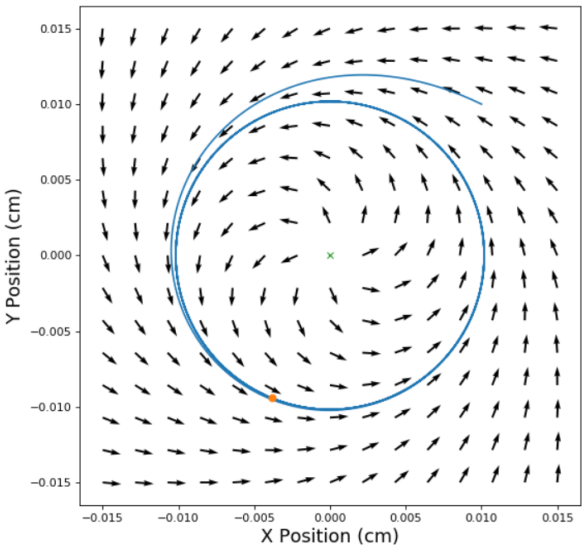

Figure 1: A guidance vector field that guides a particle into a circular trajectory. The target velocity vector for each position is represented by a black arrow. The blue line is an example path.

The rest of this paper details the research accomplishments in particle assembly. First we will provide some background on the current state of particle assembly and the thoery and physics that are needed to understand our methods. Then we will explain the methods and results for our electric field controller and chemical controller.

Background

The term "particle assembly" refers to any method that uses small particles to create useful structures. Particles can be colloids, cells, polymers, or many other materials, and structures can be any desirable arrangement of the particles. This section begins by giving justification for developing particle assembly techniques. It then explains the assembly methods that are currently used. After that it describes two mechanisms of particle motion, electrophoresis and diffusiophoresis, that can be used in particle assembly. Finally, it describes control theory approaches that may apply to particle assembly.

Applications of particle assembly

Potential applications of particle assembly techniques include tissue engineering and metamaterials synthesis methods. These applications are still distant and require much foundational work before they can be developed and implemented.

Tissue engineering involves using cells to repair, maintain, or replace biological tissue. In nanoscale tissue engineering, one of the large challenges is the creation of a synthetic extracellular matrix.[2] The extracellular matrix is a scaffolding outside the cell membrane that helps with physical support and cell signaling. The main backbone of the extracellular matrix is made of fibers assembled from biomolecules.[2] Creating fibers of a reliable and controllable size would allow larger and sturdier cellular structures to be built. Controlling the size and shape of the fibers is especially important because different tissues require specialized extracellular matrix components to effectively carry out their roles.[3] One possible application of tissue engineering is growing tissues for astronauts that are sick or injured on long-term space missions.

Another potential application of particle assembly is in metamaterials synthesis. Metamaterials are artificially engineered materials with properties that arise from the structure rather than the component parts.[4] One example of a desirable metamaterial is a material with a negative refractive index which could be used for cloaking a vehicle or person to avoid detection. Negative refractive index materials require repeating patterns, and many repeating patterns can be created using self-assembly.

Another type of desirable metamaterial is a self-regulating or self-repairing material. Self-repairing materials would improve safety and save costs in any industry, but especially on long space missions where bringing replacement parts is very costly. Materials created using dynamic self-assembly have the possibility of being self-repairing, as well as self-regulating. In nature, swarms of bees change size to regulate the swarm's temperature, so a dynamic metamaterial could potentially act in a similar way to regulate its own temperature.[5]

Particle assembly techniques

Current techniques for assembling particles into structures include top-down assembly, equilibrium self-assembly, and dynamic (non-equilibrium) self-assembly. Top-down techniques involve placing particles or groups of particles one at a time. These techniques are versatile but difficult to scale to macroscopic levels. By contrast, self-assembly is where parts of a system arrange themselves into an ordered structure, and it is much easier to scale. However, currently existing self-assembly methods can only create a limited set of structures. Self-assembly can be divided into equilibrium self-assembly, where the final assembled structure is the equilibrium state of the system, and dynamic (non-equilibrium) self-assembly, where a continuous supply of energy allows a stable structure to exist far from equilibrium.[6] Most existing artificial self-assembly methods use equilibrium self-assembly. However, since dynamic self-assembly is how biological systems grow and differentiate, dynamic self-assembly has the potential to eventually be as versatile as top-down approaches.

Top-down

The most famous top-down assembly approach is optical tweezers. The history and impact of optical tweezers are described in a review paper by Ashkin.[7] Optical tweezers use a highly focused laser to trap and move a particle. The laser exerts a force on the particle through radiation pressure in such a way that moving the laser also moves the particle. The only design restriction is that the refractive index of the particle must be higher than that of the surrounding solution. In Figure 1, researchers used optical tweezers to place silica beads into the shapes of English letters.[8] In 3 seconds, they were able to transform twenty-five one-micron silica particles from the letter "Y" into the letters "LUX". This demonstrates how versatile optical tweezers are in creating arbitrary shapes.

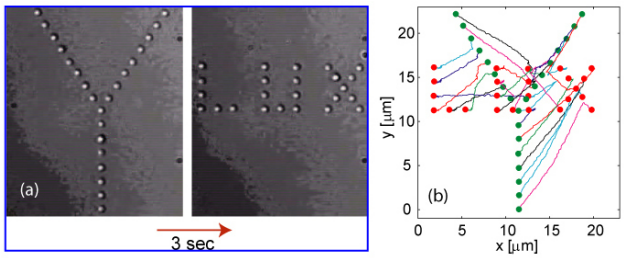

Figure 2: Structures made of silica particles assembled using optical tweezers. a) Partciles transformed from "Y" to "LUX". b) Particle trajectories. Taken from "Automated trapping, assembly, and sorting with holographic optical tweezers," Chapin (2006).[8]

Another example of top-down assembly is magnetic tweezers,[9] where a pair of magnets is used to trap a particle. The magnets can be used to place the particle in the desired location, then removed and used to trap a different particle. The main design restriction for magnetic tweezers is that they can only manipulate magnetic particles. Other top-down assembly techniques exist, including atomic force microscopy,[9] acoustic traps[10] and dielectrophoretic traps.[11] Each of these approaches can manipulate one or more objects at a time very precisely, but they do not scale well.

Equilibrium self-assembly

Self-assembly describes any process where parts of a system arrange themselves into an ordered structure. Examples of self-assembly in nature include the formation of lipid bilayers, polymer molecules, and colloid crystals. Self-assembly is normally an autonomous process, meaning the component parts will assemble into a struc-

ture without any control or direction from outside forces. This is especially true of equilibrium self-assembly, where the final state is the equilibrium state of the system. However, some researchers have found that feedback control using external variables such as temperature or electric field strength can either allow the system to reach equilibrium more quickly or reach equilibrium with fewer defects in the final structure.[12] Juarez et al.[13] created one of the first feedback-controlled equilibrium self-assembly systems. They found that the degree of crystallinity of a colloid crystal depended on the voltage of an applied electric field, and they used a proportional controller for the electric field strength to guide the degree of crystallinity to a setpoint.

Equilibrium self-assembly can also be used to create arbitrary 2-dimensional shapes by using lithography techniques. Xie et al.[14] used lithography to create patterns of insulated and exposed surface on an electrode. When the electrode was turned on, polystyrene particles moved to the exposed surface. In this way, the researchers were able to create self-assembled structures in arbitrary patterns. Figure 3 shows how the charged polystyrene particles go from an initial random state to a cross shape when the electrode is turned on.

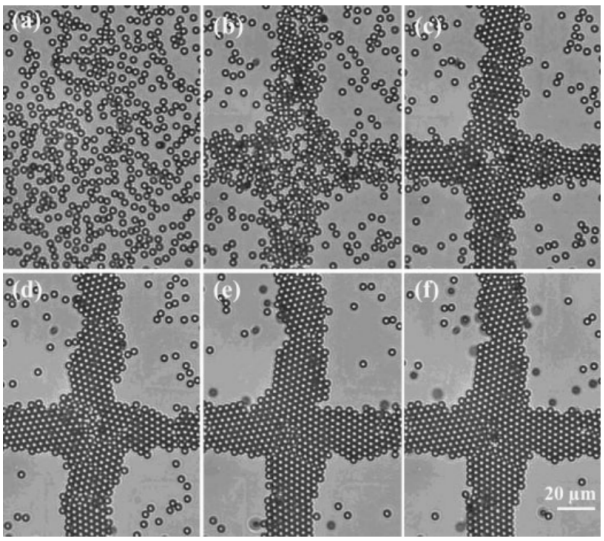

Figure 3: Cross shape assembled from polystyrene particles using a lithographic template. Taken from "Electrically Directed On-Chip Reversible Patterning of Two-Dimensional Tunable Colloidal Structures," Xie (2008).[14]

Dynamic self-assembly

Biological agents such as cells perform dynamic (non-equilibrium) self-assembly using feedback loops. Understanding these feedback loops involves understanding two key features: chemical reaction networks to signal other cells using electric potential/chemical gradients and autonomous motion in response to the gradients. As a simplified example, two cells that are too close together signal to each other, and upon receiving this signal manipulate their chemical or electrical environment so that they move apart. When a particle such as a cell moves due to an electric potential gradient it is called electrophoresis, and when it moves due to a chemical gradient it is called diffusiophoresis. Biological agents use promoters and inhibitors in a complex network of chemical reactions and feedback loops to perform feats such as tissue development and growth.

Some researchers have been able to create patterns using dynamic self-assembly. Tagliazucchi et al.[5] found in simulation that oscillating the pH created patterns that were not seen at any static value of the pH. This and other examples demonstrate that dynamic self-assembly can create novel patterns, but it is still far from the versatility of biological systems.

Mechanisms of particle motion

Electrophoresis

When small (micrometer to nanometer diameter) particles are placed in a constant electric field, they move along the field. This phenomenon is called electrophoresis. This section explains the phenomenon and gives a model for electrophoretic motion.

A particle suspended in fluid will generally have an electric surface charge due to ions that adsorb to the surface. This charge is screened by ions of the opposite charge in the fluid, which are attracted by the surface charge. For example, a particle with a positive surface charge

will attract negative ions. In an electric field, two main effects will occur: the positive surface charge will be pushed towards the negative electrode, while the negatively charged fluid in the screening ion layer will be pushed towards the positive electrode. Each of these effects will impart motion to the particle. Experimentally, the velocity of the particle is directly proportional to the negative of the electric field:[1]

$$\hat{v} = -\mu_e \hat{E} \tag{1}$$

where $\hat{v}$ is the particle's velocity vector, μ_e is the proportionality constant known as the mobility, and $\hat{E}$ is the electric field. An electric field can be produced by applying charge to an electrode. The electric field due to a single point charge on an infinite domain in three dimensions is given by Coulomb's Law,

$$\hat{E} = \frac{q}{4\pi\epsilon_0 r^2}\hat{r} \tag{2}$$

where $\hat{r}$ is the unit vector pointing from the point charge to the measured location, r is the distance between the point charge and measured location, q is the strength of the point charge, and ϵ_0 is the permittivity of free space. For multiple point charges (or multiple electrodes), the electric field is the sum of the contribution from each point charge.

Another way to calculate the electric field is, instead of using Coulomb's Law, to numerically solve for the electrical potential at discrete points. This approach assumes that the point sources are all on the edge of the domain so that they can then be treated as boundary conditions. This is the approach used by Chaudhary and Shapiro.[1]

Chaudhary and Shapiro created a method for guiding multiple particles using electric fields. They placed 20 electrodes around a square 2-dimensional domain and used a minimization algorithm to calculate the voltages of the electrodes needed to guide colloid particles on specified trajectories. They found that the algorithm worked for up to 9 particles, but for more than that the minimization problem became ill-conditioned. Any combination of trajectories worked except for moving particles that were close together in opposite directions. In another article[15] they extended this method to a 3-dimensional domain. Later, a different group wrote an algorithm to plan the optimal path for each particle.[16] This algorithm is used for path planning in general, and is not limited to electric fields.

Diffusiophoresis

Diffusiophoresis is when particles located in a fluid that contains a solute concentration gradient move in response to that gradient. Diffusiophoresis occurs because the solute concentration gradient acts like an osmotic stress, and a large enough stress can induce motion. This motion can be either up or down the concentration gradient, depending on the interactions between the particle and the solute and solvent.

Experimentally, diffusiophoresis is directly proportional to the concentration gradient,

$$\hat{v} = \mu_c \nabla C \tag{3}$$

where μ_c is the proportionality constant known as the mobility and C is the concentration of the solute. The mobility is related to the interactions between the particle and the solute and solvent. It is possible to use partial differential equations methods[17] to find the concentration gradient ∇C as a function of the distance from a point source (a chemical reaction occurring at a single point) and combine this function with Equation 3. On a 2-dimensional domain, this results in Equation 4:

$$\begin{pmatrix} v_x \\ v_y \end{pmatrix} = \begin{pmatrix} (x-x_0)\int_0^t \frac{-\mu_c}{8\pi D^2(t-\tau)^2} e^{\frac{(x-x_0)^2-(y-y_0)^2}{4D(t-\tau)}} g(\tau)d\tau \\ (y-y_0)\int_0^t \frac{-\mu_c}{8\pi D^2(t-\tau)^2} e^{\frac{(x-x_0)^2-(y-y_0)^2}{4D(t-\tau)}} g(\tau)d\tau \end{pmatrix} \tag{4}$$

where (x_0, y_0) is the location of the point source, D is the diffusion coefficient of the solute, μ_c is the mobility of the particle, and (x, y) is the position of the particle at time t. Equation 4 relates the velocity v of the particle to any time-varying point source strength $g(t)$ with units of concentration per time. Such a model is necessary for some of the control strategies explained in the next section.

Control theory

Many principles from control theory that apply on the macro-scale also apply on the micro- or nano-scale. These include feedback control, model predictive control, and guidance vector fields.

Control operations can be divided into feedback (closed-loop) and open-loop control. Feedback control is where a measurement of the current state (position, velocity, temperature, etc.) is fed to a controller to correct disturbances. This is contrasted with open-loop control, which uses a controller that does not rely on measurements. Open-loop control is desirable in situations where taking measurements is difficult or impossible. When this is not the case, feedback control is often preferred because it is more robust to disturbances or uncertainty in the system.

Two main types of controllers exist: PID controllers and controllers that use model-based control. PID controllers use the difference between a measurement and a setpoint to calculate the controller output, based on a proportional, integral, and derivative of that error. Model-based controllers rely on an empirical or theoretical model of the process to optimize for the best controller output. Model-based control is useful for dealing with multiple variables and multiple inputs. One common model-based control approach is called model predictive control. Model predictive control finds the best input strengths to minimize a cost function (the difference between the desired and projected trajectory) for a short time into the future. It then implements the first of the calculated steps, samples the state again, and repeats.

A guidance vector field (GVF) is an equation that is used to guide UAVs on stable paths around a target. The equation takes in the vehicle's current position and produces the velocity vector needed to guide it to or along the path. An example of a GVF that produces a circular trajectory around a target is given by Frew et al.[18] The same GVF with different parameters can be used for controlling particles. Using a GVF to produce target velocity vectors for particles lets us avoid the need to manually input a trajectory for each particle because the target velocity at any given time is produced from measurements of a particle's position. This also means that the trajectory will be robust to disturbances in the position.

Methods and Results

In this project we created a controller that modifies an external field to move particles on desired paths. We simulated stationary external probes that act as either charges or sources of a chemical reaction. This involved the following tasks: 1. Create a controller that uses an electric field produced by stationary point charges to steer multiple particles. 2. Create a controller that uses a chemical gradient produced by stationary point sources to steer a single particle on an arbitrary path.

Electric field controller

Electric fields have already been used successfully to control the motions of small particles. In 2006, Chaudhary and Shapiro[1] created an algorithm for controlling multiple particles by optimizing for the strengths of probes in a microfluidic device. Their intended application was to use the microfluidic device to move a particle to a sensor, but we believe it will also be useful as a starting point for self-assembly. Our first task was to recreate their methods in simulation to use as a baseline for our other results.

Chaudhary's method for steering particles using an electric field is as follows: Electrodes were placed around a square 2-dimensional domain in a microfluidic device with several colloid particles inside. The electric field inside the domain was calculated from the electrode voltages using a finite element method. A vision system measured the positions of particles in real time to enable feedback control. This position information was fed to a controller that used a least squares minimization algorithm to calculate the voltages needed to guide the particles on pre-specified trajectories. Finally, errors such as thermal noise and uncertainty in the ini-

tial positions were corrected using an additional feedback controller.

When we reproduced this method, we changed some aspects to make it easier to deal with and more applicable to our goals. First, instead of calculating the electric potential using a finite element method, we used Coulomb's law to find the electric field strength analytically. This is computationally simpler and gives increased insight into the relationship between actuator strength and particle motion. Note that by using Coulomb's law we are calculating charge strengths instead of voltages. Second, instead of inputting a desired path for each particle, we used a GVF to find the desired velocity of each particle given its position. This eliminates the need to manually input trajectories, and it also eliminates the need for an additional feedback controller to correct for disturbances since the GVF always leads particles back to the target path.

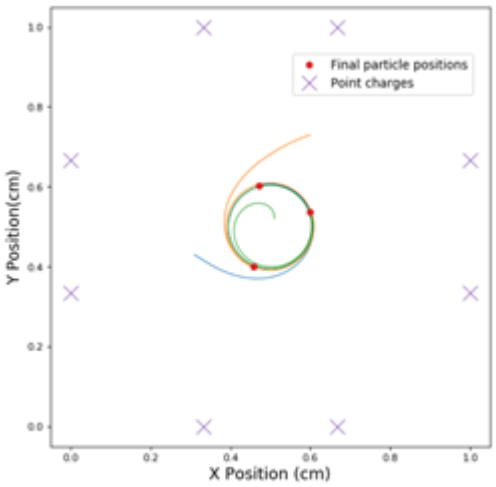

Figure 4: Motion in a circular pattern created using an electric field. The strengths of eight point charges are changed with time to guide three particles on the desired trajectory.

Implementing this task involved the following steps: First, creating a simulation with point charges arranged around a square domain to produce an electric field, and particles inside the domain that move in response to the electric field. Second, calculating the strengths of the point charges needed to move each particle along the path given by the GVF. Third, testing this simulation under different initial positions and numbers of particles.

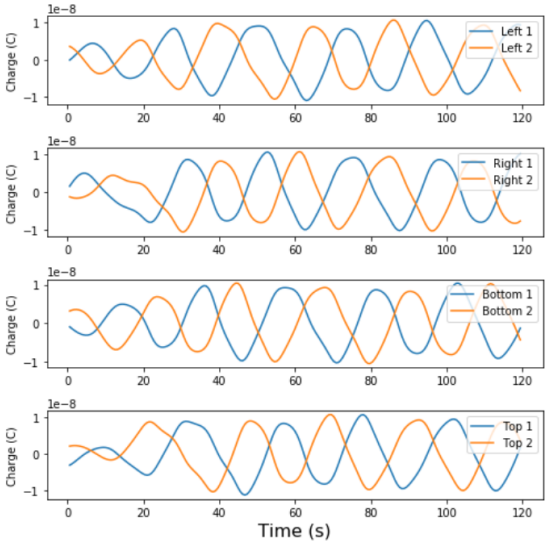

Figure 5: The charges of each of the electric probes are plotted with time.

The simulation was implemented in Python. For an example using eight probes to control three particles, the resulting path is shown in Figure 4 and the strengths of two of the probes with time are shown in Figure 5. By using more probes, up to seven particles can be easily controlled to move in a circular pattern using charge strengths up to 10^{-6} Coulombs.

Chemical controller using model predictive control

Biological systems use chemical gradients to cause motion through diffusiophoresis. To mimic biological dynamic self-assembly, we produced programmed motion using a simulation of particles experiencing diffusiophoresis.

The setup is similar to the setup for electric fields: sources are arranged around a square domain, and the strengths of the sources are calculated so that the velocity of a particle matches the velocity given by the GVF. In this case, the sources are chemical reactions instead of electric charges. Despite the similarity, there are two challenges that make using chemical gradients more difficult than electric fields. First, the concentration felt by the particle is a time-variant function of the source strength. This

means that one must know not only the current source strength but also all previous source strengths to find the concentration. Second, unlike an electric charge, a chemical reaction cannot be negative. This is a significant restriction because, as we saw in the results for electric fields, a circular pattern requires the actuator strength to oscillate between positive and negative. We can solve both these problems using model predictive control.

In the background of this document, we found a model (Equation 4) that relates the strengths of chemical point sources to the velocity of a particle. We will use this model to perform model predictive control. To set up our system, we place one point source on each of the four edges of a square domain. We use four sources because the source strengths must be non-negative, and we need two to produce both positive and negative x-direction motion and two more to produce both positive and negative y-direction motion.

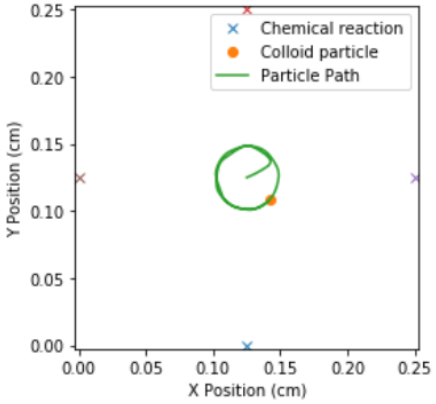

Figure 6: Motion in a circular pattern created using a chemical gradient. The strengths of the four sources of chemical reaction are varied with time to produce the desired path.

To implement model predictive control, we first set decision times with an even spacing of Δt_d at which we will calculate the strengths of the point sources. At each decision time, we optimize for the next n steps to fit the model trajectory to a target trajectory. (The quantity $n\Delta t_d$ is known as the decision horizon.) Then we implement the first of the calculated strengths, run the simulation to the next decision time, and redo the optimization. These steps are repeated until the simulation reaches its end.

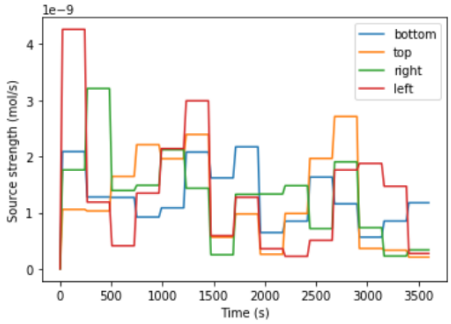

Figure 7: The strengths of the four sources of chemical reaction are plotted with time.

An example of a particle moving in a circle is shown in Figure 6. The strengths of the sources are shown in Figure 7. The sources were placed 0.25 cm apart, with the particle starting near the center of the domain. Using only 15 decision times spaced over one hour, the particle was able to approximate a circular path. One significant result was that the controller was best able to match the target velocity when the decision horizon was set equal to the characteristic diffusion time of the solute. This relationship will be explored in future work.

Summary

Existing strategies for assembling particles into structures include self-assembly approaches and top-down assembly approaches such as optical tweezers. Most existing self-assembly techniques go to equilibrium, but there is a large area to explore in non-equilibrium self-assembly. Particle assembly techniques require exerting forces on particles to induce motion. Motion can be induced through mechanisms such as electrophoresis and diffusiophoreis. Electrophoresis has been studied and applied to controlling particle motion by manipulating the electric field. Diffusiophoresis has not yet been studied deeply with respect to manipulating particle position, but the equations that govern it are similar to those that govern electrophoresis. By making use of control strategies that exist in macroscale systems, it

is possible to manipulate chemical gradients to control particle motion, which is a step towards chemical non-equilibrium self-assembly.

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