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A comparison of open-loop and closed-loop strategies in colloidal self-assembly☆

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ABSTRACT

Ordered colloidal crystals possess unique photonic properties for a wide range of applications in engineering, material science, communications, and medicine. However, ordered structures are hard to achieve even in small systems, due to the formation of defects during the process. In this paper, we focus on the control of a micron-sized SiO_2 colloidal self-assembly process for defect-free crystal in an externally applied electric field batch system. Five control strategies including time-constant and time-varying, heuristic and model-based open-loop and closed-loop policies are investigated, to understand their control mechanisms. The model-based policies are designed using a reduced-order Markov state model, built with samples from Brownian dynamics simulations, and are calculated with dynamic programming. The performance of all the five strategies are evaluated on both the Brownian dynamics simulation and the experiments. Time-varying control strategies can improve the yield of grain-boundary-free crystals over that of their time-constant counterparts. Moreover, using feedback as "endpoint detection" to terminate the process can also shorten the process time, compared to open-loop strategies.

1. Introduction

Self-assembly is a term used to describe the formation of organized structures from many discrete components due to direct or indirect interactions with each other and with their environment [1,2]. Self-assembly of nanometer and/or micrometer objects, such as particles, colloids, and folded proteins, into ordered structures holds the promise for new materials with new properties for applications in photonics, biomaterials, energy harvesting, and communications [3–7]. For example, a recent study by Ni reported the feasibility of making an ultrathin skin cloak out of nanoantennas, which exploits a negative index of refraction to render objects invisible [8]. However, ordered structures are still challenging to achieve at nano- or micro-scales due to the formation of defects during the process. Therefore, the ability to understand the system dynamics and furthermore, to control the formation of defects could significantly advance the ability to manufacture materials with low defect densities.

Micron-sized colloidal particles, by virtue of their size, can be monitored optically in real time and real space to study the system dynamics, including significant random Brownian motion. Understanding the behavior and the control of micron-sized colloidal self-assembly processes could also shed light on systems with molecular and nano-scaled components, since micron-sized colloidal particles also serve as model systems to study the phase transition behavior and crystallization kinetics for atomic and molecular crystals [9,10]. The specific system considered here is the directed self-assembly of micron-sized colloidal particles in a fluid via an externally applied electric field to form a perfect crystal. Assemblies formed via such a field compressing force are generally not perfect, and this is the problem we want to address with control.

Directed self-assembly can be used to describe a process in which directing agents, external fields, or templates are intentionally manipulated to influence the assembly process [2]. Here we use an electric field to drive the assembly; other actuators in colloidal assembly include flow field [11], magnetic field [12–14], and light [15,16]. Controlled assembly provides the opportunity to circumvent what appears to be an inherent tradeoff in self-assembly processes having high free energy barriers—the perfect crystal is the thermodynamic ground state, and theoretically could take an infinite time to reach, while rapid assembly can lead to metastable defected structures that are stuck in local minima on the free energy landscape. Time-varying directed assembly has been

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used to address this challenge, including slow voltage ramps to anneal to the perfect crystal [14], and toggling of the magnetic field to periodically relax out defects [13]. However, these open-loop strategies do not address the stochastic nature of defect formation. If the formation of a defect could be detected early and healed locally and quickly before getting locked in, it might be possible to achieve rapid assembly without the inefficiency of unneeded relaxations that slow down the assembly. There has been limited application of closed-loop feedback control in colloidal assembly, including heuristic switching rules applied in simulation [17], and proportional control applied in experiments [18], as well as the previous simulation [19,20] and experimental [21] approaches for model-based control underpinning the results presented here.

Model predictive control (MPC) was previously investigated for an electric field mediated colloidal self-assembly, using a one-dimensional Langevin equation to simulate the system dynamics [22]. In that study, 174 SiO_2 colloidal particles were simulated and order parameter C_6 was used to describe the crystallinity of the assembly. The optimal control action was solved with simulated annealing using the ten-realization averaged simulation trajectory to account for the system stochasticity. The results demonstrate that MPC is capable of accelerating the assembly process; however, the online computational time makes it challenging for practical application in such a stochastic system [22]. Alternatively, dynamic programming can be used to avoid the online computation as demonstrated in our previous study [21]. The policy was calculated by dynamic programming using a Markov decision process framework, and was able to achieve grain-boundary-free colloidal assembly in 98% yield. The closed-loop assembly was compared to a constant voltage input, which resulted in undesired grain boundary formation in almost half of the runs. In this manuscript we widen our scope to compare closed-loop optimal policies with the commonly used open-loop "toggling" approach, in which the input is periodically switched between high and low levels. We also expand beyond our previously published infinite-horizon closed-loop policy, where the policy structure is time-independent, to include a finite-horizon closed-loop policy, where the policy structure is time-dependent. The time-dependent policy enables a more direct comparison to toggling and more directly incorporates our objective of achieving crystalline assembly at the end of the process.

Simulation results in this paper were presented in a 2016 IFAC DYCOPS-CAB conference paper [23], with a focus on comparison between different "toggling" strategies and the model-based optimal open-loop and closed-loop strategies. The results in Ref. [23] demonstrate in simulation that the yield of defect free crystal can be improved with time-varying strategies, over that of a constant input strategy. Comparison between different toggling strategies further indicates an optimal switching frequency exists around 100 s, for high yield and short process time. Therefore, in this manuscript, we focus on 100 s switching "toggling" strategy and the model-based policies. We also extend the approach to experiments to better understand the similarities and differences between the simulation and the experiments, and the role of feedback to correct for unmodeled effects.

2. Methods

2.1. Experimental methods

The system we study in this work is an AC electric-field-mediated SiO_2 colloidal particle self-assembly batch process, as shown in Fig. 1, similar to the experiments in Refs. [21] and [24]. Colloidal particles with nominal diameter of 2.87 μm are suspended in a 1.0 mM NaOH solution, contained between two glass coverslips. Four electrodes are attached to 22 gauge copper wires

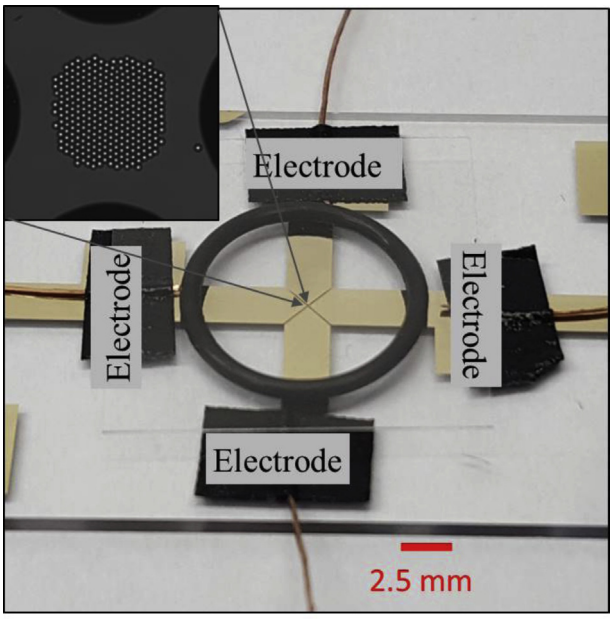

Fig. 1. Experimental batch container with four electrodes attached to generate the electric field in the system. Inset figure shows the top view of 300 SiO_2 colloidal particles suspended in 1.0 mM NaOH solution in the container under the microscope.

using conductive tape, which are connected in series to a function generator (Agilent 33220a). One lead is attached to the north-south poles and another to the east-west poles, creating an electric field across the particle suspension.

The dynamics of the system is monitored with microscopy—the inset figure in Fig. 1 shows a top view under the microscope. Microscopy is performed on an inverted optical microscope with a 63× Zeiss air objective lens (0.6 numerical aperture) at 1.25 magnification. Particle configurations are captured by a 12-bit CCD camera in the form of 336 pixel × 256 pixel (104 μm × 79 μm) digital images, at a rate of 10 frames/s. Individual particles in a two-dimensional plane are located and tracked with image analysis algorithms coded in MATLAB, which also compute the values of the two order parameters, ψ_6 and C_6, in real time. The definition of these two order parameters is given in Appendix A.

To prepare for the experiments, 100 μL of the colloidal particle dispersion was dispensed into the batch cell and allowed to sediment for 15 min prior to sealing with a coverslip, to obtain approximately 300 particles in the system. The particle location is identified as the brightness maxima within an image taken under the optical microscope, and this is recorded as the centroid of the particle. A detailed description of the optical microscopy and image analysis methods can be found in Ref. [25]. The accuracy of the particle location detection can be influenced by optical effects due to particles in close proximity [26]. To account for particle tracking errors, experimental values of the system metrics ψ_6 and C_6 were calibrated, normalized by constants of $\psi_{6\max} = 0.85$ and $C_{6\max} = 0.98$, before use in feedback in the closed-loop strategies. The closed-loop policies were computed beforehand, and were implemented as lookup tables—details are given in Section 2.4.

To make a direct comparison with a periodic toggling strategy [13], we consider two voltage levels: $0.1V$ and $0.95V$, where V is the size-dependent lowest voltage to completely crystallize the system at long time within a two-dimensional planar space, i.e. without buckling [27]. The corresponding dimensionless representations of the two input levels are defined as $\lambda = 0.2$ and $\lambda = 19.7$ respectively, to indicate the strength of the electric compression force in the system [21,24]. The mathematical relationship between λ and voltage is provided in Appendix B.

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