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Feedback Control for Shaping Density Distributions of Colloidal Particles in Microfluidic Devices

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Abstract

Directed self-assembly has great potential for the precise manufacture of structured materials at the micro/nano-scale. A local particle density often has to be controlled to make the assembly of complicated structures with no defects attainable. However, the control of spatial particle density distributions is challenged by the need for multiple actuators, kinetic trapping and the stochastic nature of self-assembly systems. In this paper, a novel feedback control approach for shaping spatial density distributions of colloidal particles is presented. The control objective is to maintain the ratio of the particle densities of two adjacent regions close to a desired value. A microfluidic device with a triple-parallel microelectrode is fabricated to provide multiple actuators for particle manipulation. The multiple-electrode actuators can be operated flexibly to either direct particles between two adjacent regions or to maintain particles within regions by preventing undesired particle movements. A feedback control scheme is implemented to control the density ratio over a broad range of tested set points. The method is generic and can be extended to include additional parallel electrodes for the control of density distributions at higher resolutions due to a modular design.

Introduction

Self-assembly promises a practical route for the manufacture of materials with unique functionalities enabled by specific structural features at the micro/nano-scale.^[1,2] Such materials can find important applications in areas such as electronics,^[3,4] photonics,^[5-7] chemical and biochemical sensors,^[8-10] integrated circuits,^[11] drug delivery,^[12] synthetic biomaterials^[13,14] and small-scale chemical processing systems.^[15] Among the various building blocks that can be used in self-assembly processes, colloidal particles are widely used.^[16] The local particle density plays a critical role when assembling a structure made out of colloidal particles with high quality. In particular, an insufficient or overabundant number of particles can lead to defective structural features.^[17-20] The particle density can only be controlled macroscopically when no local controls are available by adjusting the average density of the particles in suspension. However, for more complicated structures, the density distribution over multiple adjacent regions should be shaped in such a way that defect-free assembly of a specific structure becomes attainable. In general, a specific non-uniform density distribution is required for self-assembly of non-periodic structures, which requires local actuation to manipulate the particles at the micro/nano-scale.

Electric fields are particularly useful for local actuation due to their tunable strength and direction, which can be used to induce various electrokinetic phenomena to manipulate particles flexibly.^[18,19,21,22] In case the particles can be observed in real-

time, feedback control can be applied to account for randomness and disturbances.

Feedback control has been implemented experimentally for particle assembly using electric fields in combination with optical video microscopy, image processing techniques, and specific actuator designs,^[23,24] which allows for control of a particle density in a single region.^[25–27] However, to our best knowledge, feedback control for shaping particle density distributions over multiple local regions has never been developed. Such control is challenging due to several reasons. First, existing methods for density control are based on one or two independent actuators,^[25–27] which limits the capability to shape density distributions flexibly. Therefore, to control the particle density in multiple regions, more actuators are needed, which introduces more degrees of freedom and thus complicates the controller design and operation. Second, when a desired density distribution is obtained, the particles should either be immobilized or some controls should remain active, since entropy drives a system of particles towards uniformity. However, further control steps may still be needed to shape a density distribution at a finer resolution, which calls for a systematic decomposition of the system's ergodicity by restricting particle movements between certain regions only. Such systematic control over the system's ergodicity by shaping spatial particle density distributions at various resolutions has been approached via dynamic simulation,^[28,29] but not yet experimentally.

In this work, an automated feedback control strategy for manipulating a particle density

distribution is developed and validated experimentally. The approach is illustrated in a microfluidic device with patterned electrodes in a parallel-plate configuration. AC electric fields are applied for local electrokinetic actuation. The control objective is to maintain the density ratio of two neighboring regions close to a certain value. Three parallel microelectrodes are fabricated to provide three independent actuators for particle manipulation. Four types of system states are characterized, which can be enabled using different combinations of the electric field parameters (i.e., voltage and frequency). Sequencing of the four types of states with control steps leads to a systematic way of directing particles from one region to the other such that the density distribution can be shaped flexibly. Furthermore, particle movements are restricted to local regions to maintain the density distribution, which effectively decomposes the system's ergodicity. All control steps are embedded within an automated feedback control scheme to control the density distribution over the two regions reliably. A variety of targeted particle density distributions are tested to demonstrate the approach.

Materials and methods

Microfluidic device and particles

A microfluidic device (Figure 1) with electrodes in a parallel-plate configuration was fabricated. Two ITO electrodes (2cm×2cm) were separated vertically by a double-sided adhesive silicone rubber spacer (thickness of ~0.6mm, Grace Bio-Laboratories). The rubber spacer with a punched hole was attached to the bottom electrode. Subsequently,

a colloidal suspension ($\sim 22\mu\text{L}$) was placed carefully inside the cell to avoid air inclusion. Finally, the cell was closed by pressing the top slide on the adhesive rubber spacer. A schematic drawing of the microfluidic cell is shown in Figure 1b. The top electrode was a uniformly ITO-coated glass slide and the bottom electrode was micro-patterned using standard microfabrication techniques. In particular, glass coated with ITO with a thickness of 25-45nm was purchased (KINTEC) and cleaned by sonicating in acetone, isopropyl alcohol and deionized (DI) water for 15 minutes each. The ITO-coated glass was primed with HMDS, spin-coated with positive photoresist (HPR504, thickness of $\sim 1.3\mu\text{m}$), and exposed to UV light under a mask. The exposed photoresist was dissolved in FHD-5 developer uncovering the underlying ITO layer, which was, subsequently, etched away with aqua regia. The remaining photoresist was stripped in MS-2001 and the unetched ITO formed the pattern. Three isolated ITO stripes in a parallel pattern were fabricated to provide multiple actuators for particle manipulation, which also marked two regions for particle density control as illustrated in Figure 1c, d. The width of each stripe is $8\mu\text{m}$ and the distance between the neighboring stripes was $60\mu\text{m}$. Monodisperse $2.0\mu\text{m}$ SiO_2 colloids (Bangs Laboratories) were used as received after dilution with DI water (suspension with solids content of around 0.01%).

Experimental setup

The bottom and top electrodes were connected to a function generator (33500B, Keysight Technologies) using copper wires providing an AC signal in the range of 0.1V

to 10V (peak-to-peak) and a frequency in the range of 1kHz to 1MHz to apply AC electric fields inside the microfluidic cell. An independent AC signal could be applied to each ITO stripe on the bottom electrode. In total, the cell was connected to three channels via four copper wires (three from the stripes at the bottom electrode and one from the top electrode). An optical microscope (Nikon Ni-U) equipped with a digital camera (Nikon Digital Sight Qi2 cooled camera system) was used to continuously monitor the particles in the cell. The 1608×1608 pixel digital images ($200\mu\text{m} \times 200\mu\text{m}$) were captured at a rate of 2 frames/s. Each image was analyzed using MATLAB 2015b (The Mathworks, function *imfindcircles*) to locate the positions of the particle centers automatically, which were used to calculate the particle density distribution. During closed-loop feedback control, the most recent image was stored and used for image analysis. A feedback control scheme (implemented in MATLAB) determined the applied voltages and frequencies for each of the three channels of the function generator based on the position of the particles in the cell in real-time. The controller output values for voltage and frequency were sent to the function generator automatically via an interface between MATLAB and the function generator (Keysight Command Expert) such that they could be applied as inputs to the microfluidic cell. The input voltage and frequency were determined approximately every 0.7s based on the time needed for image analysis. At the start of each experiment, the particles were allowed to settle for 30 minutes in the cell to effectively create a two-dimensional system.

Figure 1. Illustrations of the microfluidic cell. **a**, Photograph of the microfluidic cell. **b**, Schematic design of the microfluidic cell. **c-d**, bottom electrode design.

System characterization and control steps

Four types of system states with different particle-field interactions were characterized, which we refer to as “stabilization”, “shifting”, “collection”, and “transfer”. In particular, three different electrokinetic phenomena were involved: negative dielectrophoresis (nDEP), positive dielectrophoresis (pDEP), and electroosmotic flow (EOF). DEP refers to the electrically induced movement of dielectric particles in a non-uniform electric field. The particles will move towards the electric field minimum (nDEP) if they are less polarizable than the medium. Otherwise, the particles will move towards the electric field maximum (pDEP). Besides the properties of particles and medium, this relative polarizability is also determined by the frequency of the applied

electric field. Finally, EOF is the result of electro-osmosis due to the movement of ions that form an electrical double layer caused by a transverse electric field. The effect of EOF in our system was the strongest at a low field frequency. A sequential implementation of these four types of states led to a systematic way to control the particle density difference between two adjacent regions. Furthermore, undesirable particle movements could be avoided such that the system ultimately converged to a desired set point. Figure 2 shows an illustration of the four-step approach to direct particles from the left region to the right region. First, in the stabilization step, nDEP at a high field frequency ($\sim 100\text{kHz}$) was induced using all actuators (i.e., ITO stripes) to stabilize the system (Figure 2a). In particular, the particles were restricted in the regions where the field minima existed as demonstrated with a multi-physics simulation (COMSOL), so that the particles could not cross the middle ITO stripe, which acted as the boundary between the two regions. Therefore, the initial density distribution was kept constant in the stabilization step. The particles formed chains due to their induced dipoles. Second, in the shifting step, the voltage on the left actuator was increased and the voltage on the right actuator was decreased (Figure 2b), which caused particles in the left region to move closer to the middle actuator while the particles in the right region moved further away due to a shift of the local electric field minima towards the right. The particle chains disassembled in the right region, as the local field strength was apparently not sufficiently strong anymore to polarize the particles for chaining. Third, in the collection step, EOF at a low field frequency ($\sim 5\text{kHz}$) was generated using

only the middle actuator. The flow carried the particles mainly from the left region towards the center of the cell for collection on the middle stripe (Figure 2c). Although the particle movement could in principle also be consistent with pDEP, as the particles at least initially also moved toward the edges of the middle stripe where the field maximum existed (Figure 2c), the microscopy image in Figure 2c shows that the particles eventually accumulated at the center of the middle stripe, which was consistent with EOF. Therefore, EOF was likely the main phenomenon despite the fact that pDEP might also have played a role, which was consistent with observations from similar systems in literature.^[19,25] When sufficient particles were collected from the left region, a subsequent transfer step was executed to move (only) the collected particles from the middle stripe to the right region (Figure 2d). The input parameters of the electric field at this step were the same as those applied in the shifting step (Figure 2b), yet the particles in the system behaved differently, which can be understood from the differences in particle positions. In the shifting step, despite the lower field strength in the right region, the particles in the left region were not able to cross the middle stripe due to a local field maximum at the left boundary of the middle stripe, which created an energy barrier. In contrast, at the start of the transfer step, some particles were present on the middle stripe and, therefore, could move to the right region towards the global field minimum driven by nDEP and were unhindered by the local field maximum, as shown by the corresponding simulation of the field strength. Finally, the system was stabilized again to maintain the newly formed density difference between the two

regions (Figure 2e). Note that the illustrations in Figure 2 illustrate how particles can be directed from the left region to right region, but that the approach can also be applied reversibly to direct particles from the right region to the left region. This reversibility allowed for an outer feedback control loop to be implemented for precise control over the density difference between both regions.

Figure 2. Four-step approach for shaping the particle density distribution between two adjacent regions. The top figures show for each step a schematic illustration of the dominating electrokinetic phenomena and the resulting particle behavior. The arrows refer to the active ITO stripes and the thickness of the arrows indicates the strength of the applied voltages. The second figures show for each step a

representative microscopy image. The third figures show for each step the electric field strength distribution from simulation in both the horizontal and vertical directions. The fourth figures show for each step the electric field strength distribution in the horizontal direction at a height of 1 μ m (particle radius) from simulation. The red dashed lines represent the area of the middle ITO stripe for reference. a, Stabilization (start). b, Shifting. c, Collection. d, Transfer. e, Stabilization (end).

Feedback control scheme

The automated feedback control scheme to maintain the particle density distribution between two regions close to a desired value is shown in Figure 3. The controlled variable r is the ratio of the particle density in the left region over the particle density in the right region. The control objective is to maintain this density ratio close to a set point s within a tolerance δ (set as 10%). After the system has stabilized, the density distribution is compared to the set point and, if needed, a target number of particles to be directed, N_D , is calculated by

$$N_D = (N_L + N_R) \cdot \left| \frac{r}{r+1} - \frac{s}{s+1} \right|, \quad (1)$$

where N_L and N_R represent the number of particles in the left and right region, respectively, which provides the input for the subsequent control steps. Transitions between sequential states are activated by three criteria (referred to as transition criteria),

which are designed to enable convergence of the feedback loop towards the desired density ratio. In particular, in the shifting step, the average distance between the particles in the region from which the particles have to be directed and the middle stripe, ξ , is calculated, from which a dimensionless parameter, λ , is calculated as follows

$$\lambda = \left| \frac{\xi_t - \text{Median}(\xi_{t-1}, \xi_{t-2}, \xi_{t-3})}{\xi_t} \right| \quad (2)$$

where ξ_t is the average distance obtained from the current image and ξ_{t-1} , ξ_{t-2} , ξ_{t-3} are the average distances from the three most recent images. The median value is taken to make the method more robust for stochastic noise. The transition criterion1 is defined as follows

$$\text{if } \lambda \leq 0.01 \Rightarrow \text{activate Collection and deactivate Shifting,} \quad (\text{P1})$$

which assures that the particles in the active region are as close as possible to the middle stripe under the applied electric-field conditions. The transition from the collection step to the transfer step can be triggered by several factors. First, the number of collected particles from the active side should not have exceeded N_D . Second, the applied field frequency in this step is the lowest and thus the Joule's heating effect is most severe, which might lead to fusion between particles or between particles and the ITO electrodes.^[18,30,31] Therefore, to reduce the Joule's heating effect, the collection time should not exceed a specified value. Finally, the maximum number of particles that can be collected is constrained by the surface area of the middle ITO stripe. In summary, transition criterion2 is defined as follows:

If $N_T \geq 0.95N_D \vee N_M \geq 0.95N_P \vee T_{EOF} \geq 30s \Rightarrow$ activate Transfer and deactivate Collection (P2)

N_T is the number of particles directed from the active side to the middle ITO stripe and N_M is the number of particles on the middle stripe. In practice, N_M is not necessarily equal to N_T as the middle stripe might also collect particles from the other side or adhesive particles might be present. N_P represents the maximum number of particles that can be collected on the middle ITO stripe, which has been set equal to the number of particles that can form a single line of connected colloidal particles ($N_P = 57$). Considering the delay in image analysis, which typically needs 0.7s and postpones the control actuation slightly, the collection is terminated when N_T reaches 95% of N_D or when N_M reaches 95% of N_P . T_{EOF} represents the maximum time for active EOF, starting from the moment when the middle stripe collects the first particle. Finally, the transfer step is deactivated and the stabilization step is activated when N_M does not change for 4 seconds (transition criterion3). The sharp gradient of the field strength (see Figure 2d) will drive the collected particles away from the center ITO stripe rapidly. Therefore, the value of 4 seconds has been chosen conservatively.

The control objective of the feedback control scheme can be changed to direct the self-assembly in different ways. For example, the control objective could also be defined in terms of a desired number of particles in specified regions instead of using a density ratio. For such formulation, Equation (1) would take a different form such that N_D is calculated from the difference between the actual number of particles and the desired

number of particles in a region. The remaining part of the control methodology would not change, as the variable N_D is already used in transition criterion2 and transition criterion3 only depends on time. Finally, the termination criterion of the whole algorithm would have a similar form, but would be based on a specified number of particles instead of on a ratio.

Figure 3. Sequential function chart of the control scheme for shaping the particle density distribution between two adjacent regions. Four states (stabilization, shifting, collection and transfer) are connected by transitions that can activate the next state and deactivate the previous state when the corresponding criterion is satisfied.

Results and discussion

The dynamic development of the density ratio r when the proposed feedback control

scheme is implemented starting from an initial random distribution is illustrated in Figure 4. The set point is the so-called golden ratio (i.e., $s = 1.61$), which can be achieved after one iteration of the four steps is completed. Some noise is observed in the measured ratio due to the stochastic nature of Brownian motion. The dynamic change of ζ and λ during the shifting step is presented in the insets of Figure 4. It can be seen that the transition from the shifting step to the collection step occurs somewhat later than the moment when λ decreases below 0.01 due to the delay from image analysis. Furthermore, there is no significant decrease of ζ when λ is smaller than 0.01, which means that the transition criterion1 (see P1) guarantees full completion of the shifting step at the active side with little idle time. The dynamic development of the number of particles in different regions of the triple-parallel electrode is illustrated in Figure 5, which reveals more details of the performance of the control approach. Furthermore, some characteristic numbers related to the controller performance are listed in Table 1. It can be seen that during the collection step, the middle stripe collects a small number ($N_4 = 4$) of particles from the left, which is unintentional due to randomness. Furthermore, there are four particles adhered to the middle stripe. These eight particles in total do not contribute to the number of directed particles from the right region to the left region. Therefore, according to transition criterion2 (see P2), N_T , instead of N_M , is used to assess whether sufficient particles have been collected. Since the maximum number of particles collected at the middle strip ($N_0 = 38$) does not exceed the maximum number of particles ($N_p = 57$) and the effective EOF time ($T_l = 26$ s) does

not exceed the maximum, the collection step terminates when $N_T > 0.95N_P$, which can also be seen from Figure 5b. The actual particles directed from the right region to the left region ($N_3=30$) is slightly more than required ($N_2=27$) possibly due to the delay from the image analysis.

Figure 4. Density ratio as function of time when the feedback control scheme is implemented. The dashed line indicates the set point. The two insets show the corresponding dynamic changes of the system parameters ξ and λ during the shifting step.

Figure 5. Dynamic development of the number of particles in different region of the microfluidic cell during the golden ratio control experiment (see also Table 1).

a, The number of particles in the middle stripe area (N_M). N_1 represents the total number of particles collected during the collection step and N_0 represents the value of N_M at the end of the collection step. The inset illustrates when EOF effectively starts. **b**, The number of particles in the right region (N_R). N_2 represents the number of particles needed for transfer, N_D , and N_3 represents the number of particles that are actually

directed from the active side (right side in the golden ratio experiment) to the middle stripe at the end of the collection step. T_l represents the total time at which EOF is active. c , The number of particles in the left region (N_L). N_4 represents the number of particles that are directed from the non-active side (left side in the golden ratio experiment) to the middle stripe at the end of the collection step.

Table 1. Characteristics of the control performance (see Figure 5)

N_0	N_1	N_2	N_3	N_4	T_l/s
38	34	27	30	4	26

Additional experiments with different set points (i.e., $s = 2, 3, 1/2, 1/3$) have been conducted from an initial random state. The system reaches all set points effectively (see Figure 6), which demonstrates the flexibility of the proposed feedback control scheme. Representative microscopy images confirm the anticipated changes in density distributions (see Figure 6). Note that it seems that the density ratio is controlled tighter for lower set points (i.e. $1/2, 1/3$), which is a consequence of controlling the density ratio r instead of controlling the absolute number of particles. Since the maximum number of directed particles is limited during each collection step (see P2), more than one iteration is needed when the set point is far from the initial value. All of these four experiments undergo multiple iterations, as can be seen in Figure 6. In general, the number of required iterations is also determined by the initial condition and the

stochastic nature of the system. For example, the particles on the active side may not be at the same distance to the middle stripe during the shifting or stabilization step of a certain iteration due to Brownian motion, which will eventually affect the number of collected particles on the middle stripe. The microscopy images (see Figure 6) also show that the particles are not exactly at the same distance from the middle stripe during the stabilization step in each iteration. Illustrative movies for the four experiments are available in Supplementary Information.

Figure 6. Dynamic development of the density ratio when the feedback control scheme is implemented with different set points (indicated by the dashed lines).

The microscopy images are taken at the start and at the end of the experiments (at the stabilization steps).

Finally, a set-point tracking experiment in closed-loop mode has been conducted (see Figure 7). The set point is updated to a new arbitrary value each time when the density ratio approaches the set point. The dynamic development of the density ratios shows that these set points are tracked well (see Figure 7), which further demonstrates the effectiveness and flexibility of the proposed feedback control scheme. Each time, only one iteration is needed to reach the next set point, as the neighboring set points are relatively close. In summary, the data demonstrate how arbitrary density distributions of colloidal particles between two adjacent regions can be controlled in an automated fashion using a novel feedback control scheme.

Figure 7. Dynamic development of the density ratio during a set-point tracking

experiment with feedback control.

Conclusion and Outlook

A novel feedback control approach for shaping the density distribution of colloidal particles with AC electric fields is developed. The approach is illustrated by controlling the particle densities between two adjacent regions in a microfluidic device with a triple-parallel electrode to a specified ratio effectively. Since the exploited electrokinetic phenomena are generic, it is expected that the proposed approach can be extended to other applications involving micro/nano-scale objects, such as other types of colloidal particles or biological cells to synthesize advanced metamaterials, crystals, or biological tissue. Nanoscale objects cannot be observed directly using an optical microscope, as was used in the presented work. However, indirect observation via fluorescence analysis or direct observation using liquid cell electron microscopy are feasible methods for observing smaller particles. Furthermore, methods based on X-ray scattering have been developed for real-time observation of self-assembly of nanocrystals.^[32] Besides the challenges in observation, the design of the device and execution of the control approach likely have to be adjusted in case smaller particles are approached due to the scale-dependency of the exploited electrokinetic phenomena. In particular, the random particle displacement for Brownian motion is inversely proportional to the square root of particle size and independent of voltage and

dimensions of the device. The particle displacement per unit of time as a result of DEP is proportional to the square of particle size and to the square of the voltage over the cell and inversely proportional to the cube of a characteristic dimension of the cell.^[33] Therefore, the (random) particle displacement of Brownian motion becomes more important compared to the (directed) particle displacement from DEP when the particle size decreases. For example, a detailed analysis of a comparable system has demonstrated that the displacement as a result of Brownian motion can be comparable when the particle size is smaller than 0.5 μm at low voltage.^[33] The stabilization, shifting and transfer steps are all based on DEP. In order to achieve directed displacement of particles based on DEP at similar rates as observed in this work, either the voltage would have to be increased or the characteristic dimensions of the device would have to be reduced. The former is easy to implement when the voltage is within the range of a specific function generator. Reducing a characteristic dimension of the device has potentially a strong effect and could practically be achieved by reducing the distance between the 3-stripe electrodes and the height of the cell. The collection step is based on EOF, which is not affected by particle size. However, to achieve a similar collection rate as observed in this work, a higher field strength is also desired, which once more can be achieved by increasing the voltage or reducing the characteristic dimensions of the device. A higher field strength induces more Joule heating, which may impose a practical limitation. In case of the proposed control methodology, strong Joule heating would limit the maximum execution time for the collection step and

would increase the whole processing time as more iterations would be needed for directing a certain number of particles. Finally, in general, Brownian motion inevitably leads to reduced performance of conventional feedback control schemes. Therefore, it is of interest for future research to explore, for example, stochastic control methods for the application presented in this work to take such noise into account in a robust fashion. Those methods could all potentially support an extension of the presented approach towards smaller particles. In case a system would not allow for real-time and non-invasive observation, the presented approach may also be implemented as an open-loop control strategy.^[34] A predictive mathematical model^[35–40] of the self-assembly system is needed for such open-loop control implementation. It is expected that the 3-stripe configuration can be extended into a multi-stripe configuration in a modular fashion, which would allow for the movement of particles over larger distances and consequently would enable the assembly of more complicated density distributions. Furthermore, regions between stripes could be patterned with additional electrodes or functional groups for self-assembling and immobilizing non-periodic structures at the micro/nano-scale.

Author Contribution

Y.G. designed the system, performed the experiments, analyzed the data, and wrote the paper. R.L. supervised the study, analyzed the data, and wrote the paper.

Conflicts of interest

There are no conflicts to declare

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