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Source AIChE Journal, v. 65, (6), June 2019, article number e16582

Version Accepted Version

DOI 10.1002/aic.16582

Publisher Wiley; American Institute of Chemical Engineers (AIChE)

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Gain Scheduling PID Control for Directed Self-assembly of Colloidal Particles in Microfluidic Devices

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Abstract

Directed self-assembly provides a promising route to fabricate small-scale structures for various engineering applications. The use of external fields of variable strength and frequency allows for active controls to be implemented. For assembling specific structural features, precise control over a local particle density is often needed, which can be achieved using feedback control. However, the strong nonlinear behavior of directed self-assembly complicates such control. In this work, a gain-scheduling feedback control strategy for directed self-assembly of colloidal particles is presented. The process gain is described by an empirical steady-state model, which is used for scheduling a proportional-integral feedback controller. The automated controller is implemented experimentally in a microfluidic device for directed self-assembly of colloidal particles. The gain-scheduled controller shows a significantly improved dynamic performance compared to a conventional proportional-integral controller over a broad range of operating parameters.

Topical Heading: Process System Engineering

Keywords: Self-assembly, Process Control, Gain Scheduling, Colloids, Microfluidics

Introduction

Self-assembly is the spontaneous association of molecules or particles into structures, which provides the possibility of "engineering up" approaches for the manufacture of advanced materials, functional devices or chemical processing systems at the micro/nano-scale.¹⁻⁴ Furthermore, as a novel synthesis method for chemical systems,^{5,6} self-assembly may play a critical role in the construction of "nanoscale factories" in the next frontier of processing scales.⁷ Extra degrees of freedom, such as external fields, can be introduced to guide the self-assembly process in a desired direction, which gives rise to the idea of directed self-assembly.^{8,9} Electric fields are commonly used in systems of directed self-assembly due to their tunable strength and direction, which can be used to induce a diverse set of electrokinetic phenomena to manipulate particles flexibly.¹⁰⁻¹⁷ Active controls have been proposed for directed self-assembly to account for design uncertainty, randomness, and disturbances.¹⁸⁻²⁰ In particular, feedback control of directed self-assembly has been demonstrated experimentally with optical video microscopy and image processing techniques.²¹⁻²⁴ The robustness of feedback control enables the effective assembly of colloidal structures with desired properties, such as colloidal crystals with targeted sizes²⁴ or defect-free lines made out of colloidal particles.²¹ For both types of structures, a crucial step is to control the local particle density such that defect-free assembly of desired structural features becomes practically attainable. The associated control strategies have been based on binary switching between operating conditions^{21,24} or on the assumption that the directed self-assembly system behaves linearly,²³ which leads to constant controller parameters. Closed-loop systems of directed

self-assembly of colloidal particles are generally speaking nonlinear (e.g., dielectrophoretic forces acting on particles depend in a nonlinear way on the electric-field properties^{21,25,26}), thus controllers with constant tuning parameters may exhibit satisfactory performance only for a limited range of operating conditions. Model-based controllers are well capable to deal with nonlinearities. However, the development of predictive dynamic models for directed self-assembly of colloidal particles that interact with an alternating-current (AC) electric field and are suitable for control purposes is challenging. As an alternative approach, gain scheduling has the potential to address the nonlinearities of directed self-assembly while preserving a simple controller design. Gain scheduling is commonly used to control a variety of engineering systems with nonlinear behavior.^{27–31} However, to the best of our knowledge, gain scheduling has not been applied for directed self-assembly systems yet, despite its potential.

In this work, a gain-scheduling feedback control strategy for directed self-assembly of colloidal particles is developed and validated experimentally in a microfluidic device. The control objective is to maintain a local particle density close to a desired set point. A special focus is on favourable applicability over a broad range of operating conditions in view of the system's nonlinearity. An empirical steady-state model is developed from experimental data, which describes the local particle density as function of the frequency and voltage of the applied electric field. The model has been used for controller design in two different ways. First, the attainable particle densities as function of electric field frequency and voltage have been identified when considering the

constraints on the inputs of the experimentally tested system. The resulting contour map visualizes all admissible set points of the system, irrespective of the controller design, which is used to select a single manipulated variable. Second, an automated feedback control strategy based on scheduled PID control is derived from the model by using the process gain for controller scheduling with frequency as the scheduling signal. The process time constant and dead time are assumed constant. Finally, the novel gain-scheduling controller is tested experimentally in a microfluidic device to assess the controller performance.

The article is organized as follows. In the Approach Section, the microfluidic device and the physical mechanisms that drive the directed self-assembly of the colloidal particles in the device are illustrated. Furthermore, the design of the scheduled PID controller is described including the input-output model. In the Results and Discussion Section, the performance of the scheduled PID controller implemented in the microfluidic device is presented and compared to a conventional PID controller. The article ends with conclusions and outlook for future research. A short account of our work has been published in conference proceedings.³²

Approach

Microfluidic device and directed self-assembly mechanisms

The design of the microfluidic device is based on a parallel-plate configuration (see Figure 1A). The bottom electrode (2cm×2cm) is fabricated by standard lithography techniques. In particular,

micro-patterned photoresist AZ7908 (thickness of $\sim 400\text{nm}$, non-conductive) is deposited on a uniformly ITO-coated (thickness of $\sim 35\text{nm}$, conductive) glass slide. A cell is fabricated by combining the bottom electrode with a counter electrode. The patterned bottom electrode is separated vertically by a double-sided adhesive silicone rubber spacer (thickness of $\sim 0.6\text{mm}$, Grace Bio-Laboratories) from a uniformly ITO-coated glass slide ($2\text{cm} \times 2\text{cm}$). A hole is punched into the rubber spacer to hold around $23\mu\text{L}$ of a suspension with colloidal particles. Monodisperse $2.0\mu\text{m}$ SiO_2 colloids (Bangs Laboratories) are used as received after dilution with deionized water (suspension with solids content of 0.005% - 0.05%). The two electrodes are connected with electric leads to a function generator (33500B, Keysight Technologies), which provides a potential difference over the microfluidic cell with an AC signal in the range of 0.1V to 10V (peak-to-peak) and a frequency in the range of 100Hz to 20.0MHz . A multimeter is also connected in the circuit to validate that no significant current from any potential electrochemical reactions in the cell occur. By applying electric fields with different properties over the cell, various electrokinetic phenomena can be induced to direct the self-assembly of the colloidal particles. The positions of the particles are continuously monitored using an optical microscope (Nikon Ni-U) equipped with a digital camera (Nikon Digital Sight Qi2 cooled camera system). The camera captures 1608×1608 pixel ($200\mu\text{m} \times 200\mu\text{m}$ under $60\times$ objective) digital images at a rate of 2 frames/s.

The dominating electrokinetic phenomena are illustrated in Figure 1B, C. The particles assemble near the bottom electrode due to gravity. At low field frequency, the particles are expected to self-

assemble at the electric field maxima driven by positive dielectrophoresis (pDEP).^{14,21,25} The field maxima exist around the edges of the photoresist stripe (see Figure 1B and Figure 1D).²¹ At a higher field frequency, the particles are expected to self-assemble at the electric field minima driven by negative dielectrophoresis (nDEP).^{14,21,25} The field minima exist at the center of the photoresist stripe (see Figure 1C and Figure 1D).²¹ The controlled variable is the particle density in the control area, which is indicated by the yellow dashed lines in Figure 1B, C.

Design of a gain-scheduled feedback controller

Both the voltage over the cell and the field frequency can in principle be used as manipulated variables. Steady-state data are used to determine the most effective manipulated variable to design a single-input/single-output controller. In particular, the particle density is mapped as function of electric field properties with voltages in the range of 2-10 V and frequencies in the range of 60-300 kHz. Representative microscopy images for different voltages and frequencies are shown in **Figure 2A**. Note that only a part of the density control area is shown in these microscopy images for illustrative purposes only. For each combination of voltage and frequency, five sequential images are used to determine the steady-state particle density with automated image analysis (Matlab 2015b, The MathWorks). The particle density increases with increasing voltage in case nDEP is the dominating electrokinetic phenomenon (160, 300kHz) and decreases with voltage when pDEP is the dominating electrokinetic phenomenon (60, 90kHz) (see Figure 2B). For all tested conditions, the density depends approximately linearly on the voltage when the frequency

is fixed (see **Figure 3A-D**). Furthermore, a logarithmic function can describe the density change with respect to frequency at constant voltage well (see **Figure 3E, F**). Consequently, an empirical steady-state model describing the particle density ρ (μm^{-2}) as function of frequency f (kHz) and voltage v (V) based on a linear interpolation between two logarithmic functions is proposed as follows:

$$\rho(f, v) = a_1 \cdot \log_{10} \left(\frac{f}{f_0} \right) + b_1 + \frac{a_2 \cdot \log_{10} \left(\frac{f}{f_0} \right) + b_2 - a_1 \cdot \log_{10} \left(\frac{f}{f_0} \right) - b_1}{v_{\max} - v_{\min}} (v - v_{\min}), \quad \forall v = [v_{\min}, v_{\max}], \quad (1)$$

where a_1, a_2, b_1, b_2 are fitting parameters obtained from the experimental data and $v_{\max}$ and $v_{\min}$ are the maximum and minimum voltages tested, respectively. Numerical values for the model parameters are given in

Table 1. Figure 4 shows a plot of the density as function of frequency and voltage as described by Equation (1) to indicate the dominating electrokinetic phenomena as function of electric field properties.

The field frequency is used as the manipulated variable of the PID control loop, which allows for the largest attainable range of particle densities at a fixed voltage of 10V. Derivative action is not included in the controller design to avoid poor controller performance from measurement noise due to Brownian motion, which is common for the self-assembly system studied.²¹ Consequently, the control law is as follows:

$$f(t) = f_i + K_p (s - \rho(t)) + \frac{K_p}{\tau_i} \int_0^t (s - \rho(t')) dt' . \quad (2)$$

f_i represents the initial value of the frequency, s represents the set point, and $\rho(t)$ is the measured particle density. K_p and τ_i are controller tuning parameters, which have been determined by assuming that the system can be well represented by a First-Order-Plus-Dead-Time system and by using the following Internal Model Control tuning method:³³

$$K_p = \frac{2\tau + \theta}{2K\lambda}, \quad \tau_i = \tau + \frac{\theta}{2}, \quad \lambda \geq 1.7\theta . \quad (3)$$

The system dead time θ is assumed constant and determined by the time needed for image analysis, which is approximately 0.8s, and the desired closed-loop time constant λ has been set equal to 25s.

The latter is a tuning parameter, which has been set conservatively to account for model uncertainty.

In our earlier work,²¹ we developed a feedback control loop for particle alignment of the same system within the same type of device. In the context of that work, dynamic step-change experiments showed that the process time constant was fairly constant within a broad tested frequency range for control of particle alignment. In particular, the process time constant would gradually increase from 2.7s to 4.9s when decreasing the frequency from 20 MHz to 0.020 MHz, whereas the process gain would change several orders of magnitude and change sign as well within that range. Although the control objective of the current work is different, we assume that this system characteristic still holds qualitatively, i.e., we assume a constant process time constant and only use the process gain for scheduling. Therefore, a single dynamic experiment has been conducted involving a step change at an intermediate frequency to determine the process time

constant τ . From the dynamic response (see Figure 5), the process time constant is estimated to be

3.7s. Finally, the process gain K is scheduled using Equation (1) as follows:

$$K = \left. \frac{\partial \rho(f, v)}{\partial f} \right|_{v=10} = \frac{a_2}{\ln 10} \cdot \frac{1}{f} \quad (4)$$

A non-scheduled PI controller is also tested for comparison, which uses a constant process gain K_i equal to the initial process gain:

$$K_i = \frac{a_2}{\ln 10} \cdot \frac{1}{f_i} \quad (5)$$

Both the scheduled controller (with process gain K) and the conventional controller (with process gain K_i) are implemented in the experimental system for performance assessment and comparison.

The practical implementation is realized by an interface between Matlab (using the Instrument Control Toolbox) and the function generator (using Keysight Command Expert) in combination with automated image analysis (Matlab 2015b, The MathWorks).

Results and Discussion

The closed-loop system behaviour when using the gain-scheduled controller or the conventional controller during a set-point tracking experiment is illustrated in Figure 6. The density is well controlled around each set point when using the gain-scheduled controller despite the large range over which the set points are varied (see Figure 6A). Furthermore, the system with the gain-scheduled controller reaches the set point faster compared to the conventional PI controller, especially for higher set points, with essential no overshoot for both cases. The process gain

decreases at higher frequencies. Therefore, the proportional gain can be increased for a faster response, which is automatically implemented with the gain-scheduled controller during the set-point tracking experiment (see Figure 6B, C). In contrast, the proportional gain of the conventional controller remains constant, which results in more sluggish behavior. The noise in the measured density is mainly due to the stochastic nature of Brownian motion, which causes those particles that are near the boundary of the control area to move in and out randomly. Despite the large frequency range utilized by the controller (see Figure 6C), which leads to different dominating physical phenomena (nDEP and pDEP), the gain-scheduled controller maintains a similar performance.

To compare the controller performance quantitatively, the time required to reach the set point for the first time (T) and the accumulated error (ψ) are calculated for each tested set point (see Figure 7 for illustrations and Table 2 for numerical values). For all tested cases, the set point is reached faster and the accumulated density error is smaller when using the gain-scheduled controller. Furthermore, the performance of the gain-scheduled controller is much more consistent over the range of tested conditions compared to the conventional case. The performance of the conventional controller clearly deteriorates at set points further away from the initial point, whereas the gain-scheduled controller retains a similar performance. In particular, the conventional controller behaves very inefficient at the highest set point ($0.15 \mu\text{m}^{-2}$), as demonstrated by the outliers in the

values of the corresponding T and ψ (see Table 2).

Conclusion

A gain-scheduling feedback control strategy for directed self-assembly of colloidal particles has been developed and experimentally validated in a microfluidic device. An empirical model based on experimental data can be used to estimate the process gain online for different operating conditions such that a local particle density can be controlled effectively over a broad range of set points with electric field frequency as the manipulated variable, despite the strong nonlinear behaviour of such system. Experimental implementation of the scheduled controller shows the advantage over a conventional controller. The scheduled controller outperformed the conventional controller for all tested cases, at least in terms of the time needed to reach the set point and the accumulated error. Furthermore, the performance of the gain-scheduled controller is much more consistent over a broad operating range. The method can potentially be combined with self-assembly systems that require control over local particle density to assemble final structures with reduced defects. The current method can also be extended to include several hierarchical density control steps to self-assemble complicated non-periodic structures, which has been proposed in the framework of a multi-resolution control approach.^{34–38}

The current work is based on scheduling of the controller gain only by using an empirical steady-state model and by assuming a constant process time constant. Although the developed controller

already demonstrates a significantly improved performance over a conventional PID controller, further model development is expected to improve the controller performance in several possible ways. First, the predictive capability of the model is expected to be limited outside the tested operating conditions with potentially a significant error due to its empirical nature. Incorporating first principles into the model could improve the predictive capability of the steady-state model. Second, the dynamic response of the system could be further characterized such that the process time constant can also be used for scheduling, which would extend the use of gain-scheduling control for this type of system. Third, development and validation of a dynamic model combined with first principles would allow for more advanced model-based controllers such as model-predictive control (MPC) to be explored. MPC can handle multiple inputs and allows for process constraints to be included, which are examples of advantages of MPC compared to the controller presented in this work.

The design of the device and attainable potential difference over the cell presented in this work did not allow for such high densities to be reached that colloidal crystallization could be induced. However, such crystalline structures have important applications when made defect-free, which requires tight control over particle density as well.²² Therefore, extending the approach towards higher particle densities is also an interesting direction for future research. Finally, other applications such as the self-assembly of biological cells are of interest to be investigated, as the exploited electrokinetic phenomena utilized by the controller presented in this work are generic.

Acknowledgment

The authors acknowledge Nanosystem Fabrication Facility (NFF) of HKUST for the device fabrication. This work was financially supported by the Research Grant Council of Hong Kong under Grant No. 16214617.

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Table 1. Numerical values of the model parameters

$a_1/\mu\text{m}^{-2}$	$b_1/\mu\text{m}^{-2}$	$a_2/\mu\text{m}^{-2}$	$b_2/\mu\text{m}^{-2}$	v_{min}/V	v_{max}/V	f_0/kHz
0.024	0.013	0.155	0.267	2	10	1

Table 2. Controller performance indicators for the gain-scheduled controller and the conventional controller during the set-point tracking experiment

Set point	0.09		0.11		0.13		0.15	
/μm ⁻²								
Performance	<i>T</i>	<i>ψ</i>	<i>T</i>	<i>Ψ</i>	<i>T</i>	<i>Ψ</i>	<i>T</i>	<i>Ψ</i>

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indicator	/s	/s· μm^{-2}	/s	/s· μm^{-2}	/s	/s· μm^{-2}	/s	/s· μm^{-2}
With gain	27.0	0.5385	28.5	0.5693	20.5	0.5025	27.5	0.6945
scheduling								
Without gain	47.5	0.7381	82.5	1.0191	98.5	0.9753	188	1.8827
scheduling

Figure 1. (A) Design of the microfluidic device. (B)-(C) Illustration of the directed self-assembly behavior when positive dielectrophoresis and negative dielectrophoresis are dominating phenomenon, respectively. The upper figures show the bottom electrode of the microfluidic device and the bottom figures show illustrative microscopy images. The yellow dashed lines indicate the density control area. (D) 2D map of the electric field strength around the electrode interface.

Figure 2. (A) Microscopy images of part of the density control area. (B) Measured particle densities as function of voltage and frequency. Five sequential images were used to calculate the density for each combination of frequency and voltage.

Figure 3. (A)-(D) Linear regression of measured particle density as function of voltage for selected frequencies. (E)-(F) Logarithmic regression of measured particle density as function of frequency for selected voltages. Each dot represents the average density value calculated from five sequential images, which has a neglectable variation.

Figure 4. Contour plot of empirical model describing particle density as function of field frequency and voltage. The regions where either nDEP or pDEP are the dominating electrokinetic phenomenon are approximately separated by the dashed line.

Figure 5. Dynamic development of particle density during a step change in frequency from 90kHz to 110kHz at a constant voltage of 10V. The horizontal blue dashed lines represent the assumed steady-state values and the vertical solid black line indicates the time at which the step change was implemented.

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Figure 6. Closed-loop system behavior with gain-scheduled PI controller or conventional PI

controller (A) Density (controlled variable), (B) Proportional gain and (C) Frequency (manipulated variable) as function of time during a set-point tracking experiment. The dashed step-wise line in (A) represents the set point. Data is collected approximately every 0.7s, which is determined by the speed of the automated image analysis.

Figure 7. Illustration of two controller performance indicators: time to reach the set point (T) and accumulated error (ψ).

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