

REPORT DOCUMENTATION PAGE			Form Approved OMB NO. 0704-0188		
<p>The public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA, 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to any penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number. PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ADDRESS.</p>					
1. REPORT DATE (DD-MM-YYYY) 12-02-2020		2. REPORT TYPE Final Report		3. DATES COVERED (From - To) 2-May-2016 - 1-Nov-2019	
4. TITLE AND SUBTITLE Final Report: A Fundamental Study of Electrokinetic Instabilities to Manipulate and Self-Assemble Nano- and Microparticles			5a. CONTRACT NUMBER W911NF-16-1-0278		
			5b. GRANT NUMBER		
			5c. PROGRAM ELEMENT NUMBER 611102		
6. AUTHORS			5d. PROJECT NUMBER		
			5e. TASK NUMBER		
			5f. WORK UNIT NUMBER		
7. PERFORMING ORGANIZATION NAMES AND ADDRESSES Georgia Tech Research Corporation 505 Tenth Street NW Atlanta, GA 30332 -0420			8. PERFORMING ORGANIZATION REPORT NUMBER		
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS (ES) U.S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211			10. SPONSOR/MONITOR'S ACRONYM(S) ARO		
			11. SPONSOR/MONITOR'S REPORT NUMBER(S) 68079-EG.6		
12. DISTRIBUTION AVAILABILITY STATEMENT Approved for public release; distribution is unlimited.					
13. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.					
14. ABSTRACT					
15. SUBJECT TERMS					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT UU	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON Minami Yoda
a. REPORT UU	b. ABSTRACT UU	c. THIS PAGE UU			19b. TELEPHONE NUMBER 404-894-6838

RPPR Final Report
as of 06-Mar-2020

Agency Code:

Proposal Number: 68079EG

Agreement Number: W911NF-16-1-0278

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EIN: 580603146

Report Date: 01-Feb-2020

Date Received: 12-Feb-2020

Final Report for Period Beginning 02-May-2016 and Ending 01-Nov-2019

Title: A Fundamental Study of Electrokinetic Instabilities to Manipulate and Self-Assemble Nano- and Microparticles

Begin Performance Period: 02-May-2016

End Performance Period: 01-Nov-2019

Report Term: 0-Other

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Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees:

STEM Participants: 2

Major Goals: Please see pdf under "Upload" section

Accomplishments: Please see pdf under "Upload" section

Training Opportunities: This grant supported two graduate students during their doctoral research:

1) Andrew Yee, a Ph.D. student in Mechanical Engineering at GT, has been trained in optical diagnostics, advanced microscopy and image processing methods, as well as colloid science and microfluidics. He should complete his doctoral dissertation in May this year, and has co-authored three (two accepted, plus one submitted) journal papers to date from this research. We expect to submit another journal paper on his work this year.

2) Varun Lochab, a Ph.D. student in Mechanical Engineering at OSU, has been trained in microfabrication and nanofabrication methods and fundamentals of optical and electrical characterization for microfluidic devices. He has been working on the confocal imaging for particle migration and developing the instrumentation and preliminary results on the printing of structures from microchannels. Mr. Lochab will complete his doctoral dissertation in Summer 2020, and has co-authored one journal paper to date from this research, along with an additional journal paper on a project he is assisting other members of the Prakash group in bacterial disinfection [D. H. Dusane, V. Lochab, T. H. Jones, C. W. Peters, S. Roy, C. K. Sen, V. V. Subramanian, D. J. Wozniak, S. Prakash and P. Stoodley (2019) In vitro evaluation of eradication of Pseudomonas aeruginosa biofilms by electrical stimulation. Scientific Reports 9(1), 1–3].

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Results Dissemination: To date, the results from this project have been published in two archival papers:

- 1) A. Yee and M. Yoda (2018) "Experimental observations of bands of suspended colloidal particles subject to shear flow and steady electric field," *Microfluidics and Nanofluidics* 22, 113/1-12
- 2) V. Lochab, V., A. Yee, M. Yoda, A. T. Conlisk and S. Prakash, S. (2019) "Dynamics of colloidal particles under combined pressure and electric potential gradients," *Microfluidics and Nanofluidics* 23, 134/1-13

An additional paper on banding (Rossi et al. 2019; cf. Ref. 5 in Accomplished Under Goals) from previous support by NSF (so ARO support is not acknowledged) was also published during this period.

A third archival paper is currently under review:

- 3) A. Yee, H. Onuki, Y. Tagawa and M. Yoda, M. "Determining time scales for directed assembly of particles into bands by shear flow and electric fields," submitted to *Experiments in Fluids* (2019)

We have also presented our results as invited talks at conferences and workshops:

- 1) M. Yoda (2017) "Near-wall dynamics of suspended colloidal particles in microscale shear and electrokinetic flows," Gordon Research Conference on Physics and Chemistry of Microfluidics, Lucca (Barga), Italy (2017)
- 2) M. Yoda (2018) "Some observations of the near-wall dynamics and assembly of colloidal particles suspended in steady shear and electroosmotic flows," Lorentz Center Workshop: Micro- and Nano-Fluidics: Fundamentals and Applications, Leiden, the Netherlands
- 3) M. Yoda (2020) "Near-wall dynamics and assembly of suspended colloidal particles driven by shear flow and electric field," Okinawa Institute of Science and Technology Mini-Symposium: Fluid-Structure Interactions: From Engineering to Biomimetic Systems, Okinawa, Japan

as seminars at several American and international universities and the Army Research Laboratory in Aberdeen, MD:

- 4) M. Yoda (2016) "The dynamics of near-wall colloidal particles suspended in Poiseuille and electroosmotic flows," Department of Mechanical Systems Engineering, Tokyo University of Agriculture and Technology, Japan
- 5) M. Yoda (2017) "Unexpected dynamics of near-wall colloidal particles suspended in Poiseuille and electroosmotic flows," Department of Mechanical Engineering, Texas Tech University, Lubbock, TX
- 6) M. Yoda (2018) "Near-wall dynamics of suspended colloidal particles in shear and electroosmotic flow," Physics of Fluids Group, University of Twente, Enschede, the Netherlands
- 7) M. Yoda (2018) "Near-wall dynamics of suspended colloidal particles subject to shear flow and dc electric field," Department of Mechanical Engineering, Clemson University, Clemson, SC
- 8) M. Yoda and S. Prakash (2018) "Heterogeneous assembly and printing of colloidal particle bands," Weapons and Materials Directorate, US Army Research Laboratory, Aberdeen, MD

and as talks at several conferences:

- 9) A. Yee and M. Yoda (2016) "Effect of flow parameters on assembly of colloidal particle bands in Poiseuille and electroosmotic flow," 69th Annual Meeting of the American Physical Society Division of Fluid Dynamics (APS DFD), Portland, OR
- 10) M. Yoda and A. Yee (2016) "How particle properties affect the assembly and characteristics of colloidal particle bands," Ibid.
- 11) A. Yee and M. Yoda (2017) "Colloidal band assembly from different suspended particles," 70th Annual Meeting of the APS DFD, Denver, CO
- 12) M. Yoda, A. Yee, V. Lochab and S. Prakash (2017) "Colloidal band assembly in different microchannels," Ibid.
- 13) Y. Li, Y. Tagawa, A. Yee, A., and M. Yoda (2017) "Observations of the initial stages of colloidal band formation," Ibid.
- 14) V. Lochab, A. Yee, Y. Li, M. Yoda, A. T. Conlisk and S. Prakash (2018) "Directed self-assembly of colloidal particles for high aspect ratio bands," Hilton Head: A Solid-State Sensors, Actuators and Microsystems Workshop, Hilton Head Island, SC [poster presentation]
- 15) A. Yee and M. Yoda (2018) "Heterogeneous colloidal assembly: Band formation in a mixture of suspended particles," 71st Annual Meeting of the American Physical Society Division of Fluid Dynamics, Atlanta, GA
- 16) S. Prakash and V. Lochab (2018) "Electrohydrodynamic printing of pre-assembled colloidal structures," Ibid.
- 17) A. Yee and M. Yoda (2018) "Colloidal band assembly: Effect of streamwise position," Ibid.
- 18) A. Yee and M. Yoda (2019) "Tracking particle assembly into bands," 93rd American Chemical Society Colloid & Surface Science Symposium, Atlanta, GA
- 19) S. Prakash and V. Lochab (2019) "Colloidal particle transfer from microchannel nozzle to porous substrates,"

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Ibid.

20) S. Prakash and V. Lochab (2019) "Electrohydrodynamic printing of pre-assembled colloidal particle structures," 72nd Annual Meeting of the APS DFD, Seattle, WA

21) A. Yee and M. Yoda (2019) "Colloidal particle dynamics during band assembly," Ibid.

22) M. Yoda, A. Yee, H. Onuki and Y. Tagawa (2019) "Determining time scales for directed assembly of particles by shear flow and electric field," 72nd Annual Meeting of the APS DFD, Seattle, WA (2019) [flash/poster presentation]

Finally, we expect to publish at least one more journal paper from Mr. Yee's thesis, and one more journal paper from Mr. Lochab's thesis. The organizers of the OIST workshop (cf. talk #3) will be editing a special issue in Physics of Fluids on the invited talks.

Honors and Awards: Nothing to Report

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI

Participant: Minami Yoda

Person Months Worked: 2.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Participant Type: Co PD/PI

Participant: Shaurya Prakash

Person Months Worked: 2.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Participant Type: Graduate Student (research assistant)

Participant: Andrew Yee

Person Months Worked: 12.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Participant Type: Graduate Student (research assistant)

Participant: Varun Lochab

Person Months Worked: 12.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

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Participant Type: Graduate Student (research assistant)

Participant: Hajime Onuki

Person Months Worked: 2.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

ARTICLES:

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Microfluidics and Nanofluidics

Publication Identifier Type: DOI

Publication Identifier: <https://doi.org/10.1007/s10404-018-2136-3>

Volume: 22

Issue:

First Page #: 113

Date Submitted: 10/17/18 12:00AM

Date Published:

Publication Location:

Article Title: Experimental observations of bands of suspended colloidal particles subject to shear flow and steady electric field

Authors: Andrew Yee, Minami Yoda

Keywords: colloidal particle assembly, colloidal suspensions, microfluidics, electroosmotic flow, Poiseuille flow

Abstract: Manipulating suspended colloidal particles flowing through a microchannel is of interest in microfluidics and nanotechnology. However, the flow itself can affect the dynamics of these suspended particles via wall-normal "lift" forces. The near-wall dynamics of particles suspended in shear flow and subject to a dc electric field was quantified using total internal reflection fluorescence in combined Poiseuille and EO flow through a ~30 um deep channel. When the two flows are in opposite directions, the particles are first attracted to the wall, then assemble into very high aspect ratio structures, or concentrated streamwise "bands," above a minimum electric field magnitude, and, it appears, a minimum near-wall shear rate. These bands, which only exist over the few um next to the wall, are roughly periodic in the cross-stream direction, although there are no external forces along this direction. Experimental observations and dimensional analysis of the time for the first band to form

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report
as of 06-Mar-2020

Major Goals

This is the final report for the US Army Research Office grant W911NF-16-1-0278, summarizing our results from May 2, 2016 through November 1, 2019. This research is based on our discovery that suspended 0.5 μm diameter fluorescent polystyrene (PS) particles are attracted to, and accumulate near, the walls of a microchannel in combined Poiseuille and electroosmotic (EO) “counterflow.” This serendipitous observation is unexpected given that both the particle and channel surfaces are negatively charged. More surprising, these particles, after accumulating near the wall, assemble into periodic bands aligned with the flow (and electric field) direction above a minimum electric field magnitude $|E_{\text{min}}|$ and, in most cases, a minimum shear rate $\dot{\gamma}_{\text{min}}$ (Fig. 1). These bands, which we now know only exist within a few μm of the (upper and lower) wall(s), are very elongated microstructures with cross sectional dimensions of micrometers and lengths of centimeters. Inspired by these discoveries, the objective of this research is to develop a fundamental understanding of how the dynamics and assembly of suspended colloidal particles with diameters of $O(100\text{ nm}–1\ \mu\text{m})$ are impacted by the external electric fields and shear flow characteristic of Poiseuille and EO flows.

After more than three years of work, we have made significant progress on particle accumulation and band assembly, although there remain major gaps in our understanding of these phenomena. To summarize, the research objectives from the original proposal were to:

- I) Determine how the characteristics (*e.g.* time scales and spatial period) of the bands depend upon particle properties such as average radius a , bulk volume fraction ϕ_{∞} , zeta-potential ζ_p ; flow properties such as near-wall shear rate $\dot{\gamma}$ and electric field magnitude $|E|$; and channel properties such as channel dimensions, material, and wall zeta-potential ζ_w .
- II) Model a single particle suspended in an infinite conducting fluid subject to flow shear and a steady electric field along the flow direction to gain insight on why the particle experiences a “lift” force whose direction changes with the electric field polarity.
- III) Determine if the bands only exist near the wall (and the particles in the bulk flow are then depleted by Continuity), and if formation occurs only when the concentration of near-wall particles exceeds some minimum, or “threshold,” value.
- IV) Develop methods for isolating and extracting bands to form assembled structures.

In the absence of any relevant theory (to our knowledge) that explains these observations, our approach over the period of this grant focused on experimental characterization. We have achieved objectives I, IV, most of III (with completion scheduled for summer 2020), and have partially achieved objective II, as detailed in the next section.

Our current understanding of band assembly, based on extensive experimental observations, divides this process into three stages,¹ namely:

- 1) **Accumulation**, where the particles are attracted to and concentrated near the wall, depleting particles from the bulk of the flow;
- 2) **Band formation**, where the particles assemble into a relatively large number of unstable bands which shift in their cross-stream (y) position, with significant band merging and splitting; and
- 3) **Steady state**, where the bands have a fairly consistent y -position and the total number of bands (over a given y -extent) decreases to a nearly constant value.

Given that we have concluded that band formation is not associated with an instability, the **working hypothesis** guiding this research, updated based on our results obtained during this period, is that:

- the suspended particles are attracted to the wall during the accumulation stage due to inertial lift-type forces associated with electrophoretic “slip” between the particle and the suspending fluid;
- the mechanism(s) underlying band assembly is distinct from that for the lift force;
- band assembly is driven instead by interparticle interactions.

Accomplished under Goals

Objective I

Dr. Yoda’s group has performed extensive experimental studies to characterize how the time scales for the accumulation stage, and hence the time required for the start of the band formation stage, and the number of bands observed in the steady-state stage, depend upon particle solution properties (a , φ_∞ , ζ_p); flow properties ($\dot{\gamma}$, $|E|$); and channel properties such as material and ζ_w ,² as summarized in Table I. All of the flow experiments started with “pure” Poiseuille flow, and EO flow (in the opposite direction) was started by applying a dc electric field at time $t = 0$. Grayscale peaks above a threshold value were identified as “bands” using row-averaged (*i.e.*, averages along the x -direction) grayscale profiles, and the number of these bands N was then plotted as a function of t , and a sigmoid function was curve-fit to $N(t)$. A time scale for band formation T_0 was defined to be the time when $N = 1$, *i.e.*, when the first band formed, based upon this curve-fit. The minimum electric field magnitude $|E_{\min}|$ when bands, defined again by spatially averaged grayscale peaks, form increases with a ; conversely, $|E_{\min}|$ increases as both φ_∞ and ζ_p decrease. Band characteristics scale with the electric field offset $\Delta E \equiv |E| - |E_{\min}|$, vs. $|E|$ itself.

The time for the first band to form T_0 decreases as φ_∞ , ζ_p , ΔE and $\dot{\gamma}$ increase; T_0 increases with a , however. Most interestingly, T_0 scales with the inverse of the shear rate $\dot{\gamma}$, and $T_0\dot{\gamma}$ decays exponentially with ΔE (Fig. 2), at least for $\varphi_\infty \geq 1.7 \times 10^{-3}$. The average number of bands in the

¹ A. Yee, H. Onuki, Y. Tagawa and M. Yoda (2019) “Determining time scales for directed assembly of particles into bands by shear flow and electric fields,” submitted to *Experiments in Fluids*

² A. Yee and M. Yoda (2018) Experimental observations of bands of suspended colloidal particles subject to shear flow and steady electric field. *Microfluidics and Nanofluidics* **22**, 113

steady-state stage N_s appears to increase roughly linearly with $\dot{\gamma}$; N_s also appears to decrease as ΔE and a increase.

Dr. Prakash's group has shown, using confocal scanning microscopy (CSM) visualizations of the entire channel cross-section, that colloidal particle migration occurs either towards or away from the microchannel walls depending on the relative directions for the applied potential and pressure gradients. When the pressure gradient driving the fluid flow and the electric potential gradient (*i.e.*, electric field) are applied in the same direction, the colloidal particles migrate away from the microchannel walls. In the case where the pressure and potential gradients are in the opposite direction, the particles migrate towards the microchannel walls and subsequently assemble into distinct bands next to both the bottom glass and top PDMS walls. Their results demonstrate that the particle dynamics due to electrophoresis in Poiseuille flow within a microchannel results in nonuniform spatial distributions of colloidal particles *via* cross-stream migration, with the ability to assemble particles into distinct band structures at channel walls.

The particle migration and accumulation preceding band formation is driven by an 'electrophoretic lift' force. Past work on particle sedimentation has attributed particle migration to an 'inertial lift' force. Therefore, the evaluation of inertial terms in the governing equations for fluid motion remain a vital question. We expect to continue our investigation by tuning fluid properties, such as viscosity μ and density ρ , for a parametric study to evaluate how this lift force depends upon the fluid properties.

Objective II

Dr. Prakash's modeling efforts on this objective (II) have shown that it is unlikely that band formation is caused by a hydrodynamic flow instability because the flow Reynolds numbers are very small ($Re < 1$); it also appears that dielectrophoretic (DEP) forces between particles are, based on order-of-magnitude estimates, also far too weak to drive assembly. Regarding the lift force, two new models from other groups describing 'inertial migration' or 'phoretic lift' due to the electrophoretic 'slip' between the particles and the flow were published in 2019.^{3,4} Although these initial models assume that viscous effects are dominant (and electrokinetic effects are negligible), which is not the case for our experiments, they show that the particles are attracted to (or repelled from) the walls when they lead (or lag) the Poiseuille flow, in qualitative agreement with our observations.

Objective III

Our studies have shown that bands only exist near the wall, and that particles are depleted from the bulk. Results using anamorphic (astigmatic) imaging show that the maximum in particle concentration occurs about 3 μm from the wall (for $a \approx 250 \text{ nm}$).⁵ The CSM visualizations by Dr.

³ A. S. Khair and B. Balu (2019) The lift force on a charged sphere that translates and rotates in an electrolyte. *Electrophoresis* **40**, 2407

⁴ A. Choudhary, T. Renganathan and S. Pushpavanam (2019) Inertial migration of an electrophoretic rigid sphere in a two-dimensional Poiseuille flow. *Journal of Fluid Mechanics* **874**, 856

⁵ M. Rossi, A. Marin, N. Cevheri, M. Yoda and C. J. Kähler (2019) Particle distribution and velocity in electrokinetically-induced banding" *Microfluidics and Nanofluidics* **23**, 67

Prakash's group show that bands are observed near the top and bottom walls in monolithic fused-silica channels, and next to both the bottom silica and top polydimethylsiloxane (PDMS) walls (in the same microchannel) of silica-PDMS microchannels (*cf.* Fig. 1).⁶ Interestingly, the number of, and distance between, bands appears to be essentially independent of the nearly two-fold change in ζ_w between silica and PDMS.

Most recently, Dr. Yoda, in collaboration with Dr. Y. Tagawa and his PhD student H. Onuki at the Tokyo University of Agriculture and Technology, has examined an alternative time scale for band formation T_1 based directly upon the image grayscales (*vs.* a more indirect measure where bands are taken to be grayscale "peaks" that exceed a "threshold" by a somewhat arbitrarily determined margin).¹ The spatially averaged normalized grayscale \bar{G} was used as an estimate of overall brightness of the image and the normalized standard deviation $\bar{\sigma}_G$ was used to estimate the overall contrast. The time scale T_1 was determined from the inflection point in $\bar{\sigma}_G$, and appears to be comparable to T_0 ; moreover, T_1 (like T_0) decreases as $\dot{\gamma}$, φ_∞ , and ΔE increase, and decreases with streamwise position x (measured from the channel inlet). Interestingly, \bar{G} appears in most cases to have self-similar exponential growth in terms of physical time scaled by T_1 (Fig. 3), a time scale determined from $\bar{\sigma}_G$ (*vs.* \bar{G}).

Unfortunately, \bar{G} is not a direct measure of the particle concentration. Dr. Yoda's group is therefore performing "two-color" experiments where a 1% of the suspended particles are labeled with a different fluorophore, and used as tracers. Initial results (Fig. 4) suggest that the growth in the number of near-wall tracer particles (over a given area of 203 μm square), like the average grayscale, has an exponential growth during the accumulation stage. These results confirm that the accumulation of particles during this stage is an avalanche-type process.

The initial estimates of tracer particle velocities from these two-color experiments also clearly show that the particle velocities are less than what would be expected from theory (*i.e.*, the sum of the flow and electrophoretic velocities) during both the accumulation and steady-stage stages (Fig. 5). Interestingly, the initial results also suggest that the particles in the bands during the steady-state stage have a speed that is independent of their distance from the wall (Fig. 5*b*), at least for the particles with centers within 500 nm of the wall visualized in these experiments (astigmatic imaging results suggest that the bands extend at least 5 μm from the wall). The two-color experiments, which will be completed by May this year, will obtain more robust estimates of the actual number of near-wall particles at the end of the accumulation stage and particle velocities during the accumulation stage, as well as between and within the bands during the steady-state stage.

Objective IV

Dr. Prakash's group has developed a process to extract bands from, and print bands outside, the microchannel. Their approach is a template-free process, which is facilitated by continuous

⁶ V. Lochab, A. Yee, M. Yoda, A. T. Conlisk and S. Prakash (2019) Dynamics of colloidal particles under combined pressure and electric potential gradients. *Microfluidics and Nanofluidics* **23**, 134

solvent drainage through porous substrates and works in real-time for particle transfer of the colloidal particles into high-aspect ratio (length / height $>10^3$), structures, such as linear arrays or grid structures.

An Arduino-controlled stage (built in-house) translates a microchannel print head (Fig. 6) along (x, y, z) relative to the porous substrate with the external substrate placed below. Poiseuille flow was generated with a syringe pump (PicoPlus, Harvard Apparatus) for flow rates $Q \leq 5 \mu\text{L}/\text{min}$; an electric field was applied with a Bertan High Voltage Power Supply (Series 230) between a ~ 0.6 mm diameter stainless steel wire electrode and a chromium-gold (Cr/Au) (4 nm/25 nm) layer on a glass microscope slide under the substrate.

During printing, the stage was translated with velocity $\bar{\mathbf{u}}_s = (-0.63, 0, 0)$ mm/s along (x, y, z) (Fig. 6), while the channel was fixed at $z \approx 190 \mu\text{m} \pm 60 \mu\text{m}$ from the top surface of the Cr/Au coated slide. The run-time for each printing operation was ~ 20 s, which was adequate for printing structures over substrate lengths (x -dimensions) >1 cm (Fig. 6).

Figure 7 shows the band assembly at a $\sim 45^\circ$ angle obtained by the changing the orientation of the substrate with respect to x (Fig. 7a) prior to the start of the assembly process at a volume flowrate $Q = 1 \mu\text{L}/\text{min}$ and $|E| = 35$ V/cm. Figure 7b shows an scanning electron microscopy (SEM) image of the self-assembled particles in the bands, where a few particles can also be observed in between bands, which could be due to the Brownian motion of the colloidal particles. Moreover, grid patterns were also assembled (Fig. 7c, d) by first printing along one direction on the substrate, then rotating the substrate by $\sim 90^\circ$ and printing over (and perpendicular to) the previous pattern.

One challenge with the printing process is identifying particles of different ζ_p in, for example, heterogeneous assembly of materials from particle mixtures. Dr. Prakash's group received a Defense University Research Instrumentation Program (DURIP) grant to purchase a Fourier transform infrared (FTIR) system coupled to an IR microscope for chemical imaging of the particle distributions. Figure 8 shows their first attempt to print and identify particle distributions for a homogenous set of particles on a porous silver substrate.

Figures

Table I Summary of experimental parameters where band formation is observed.

Mean particle radius a [nm]	245, 355, 500
Particle ζ -potentials ζ_p [mV]	-80 to -40
Bulk particle volume fraction ϕ_∞	3.3×10^{-5} to 5×10^{-3}
Wall ζ -potentials ζ_w [mV]	-110 (fused silica) to -50 (PDMS-silica)
Aqueous electrolyte solutions / pH (anion concentration = 4 mM)	$\text{Na}_2\text{B}_4\text{O}_7$ (pH9); H_3BO_3 - $\text{Na}_2\text{B}_4\text{O}_7$ (pH6); KNO_3 - KOH (pH7)
Electric field magnitude $ E $ [V/cm]	70-500
Near-wall shear rate $\dot{\gamma}$ [s^{-1}]	700-1800

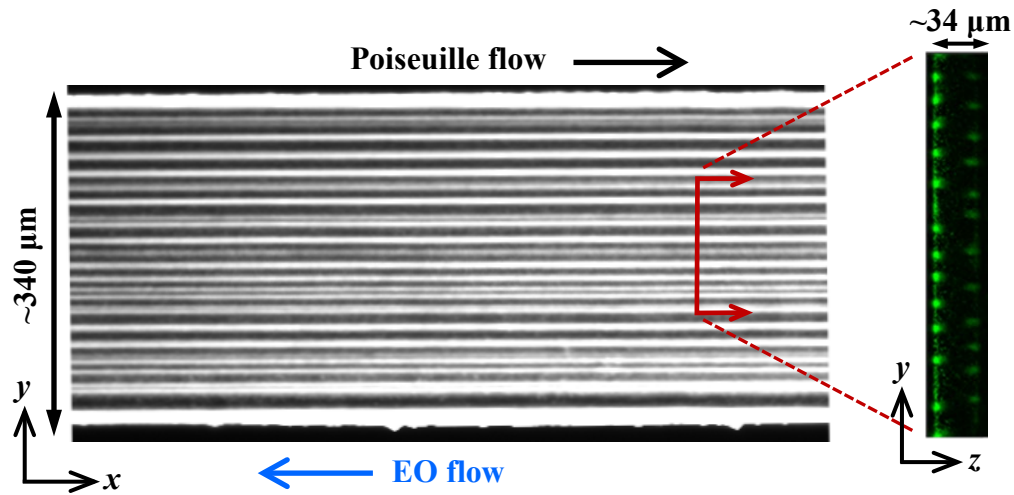


Figure 1 Evanescent-wave visualization of a x - y plane next to the wall showing very elongated structures, or “bands,” in a fused-silica channel with a roughly consistent cross-stream (y) spacing [*left*], and a confocal microscopy image of a y - z cross-section of a different silica-PDMS channel [*right*] (with the flow direction normal to the page) showing bands only next to the left silica and right PDMS walls. Note that there are almost no particles in the bulk away from the walls.

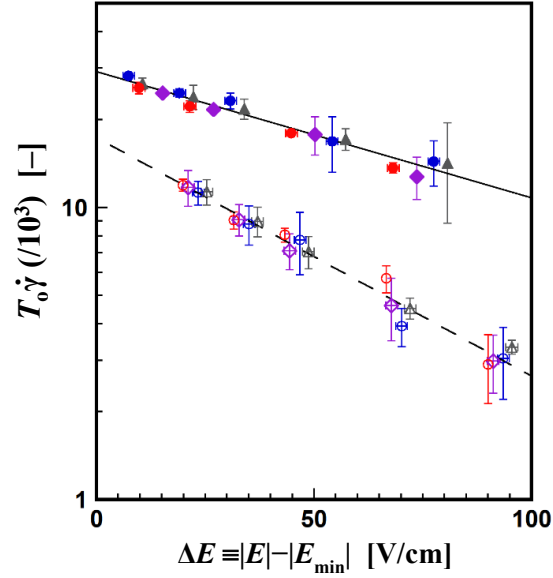


Figure 2 Semi-log plot of dimensionless band formation time $T_0 \dot{\gamma}$ (note the vertical axis is scaled by 10^3) vs. electric field offset ΔE for $a = 245$ nm, $\zeta_p = -44$ mV particles at $\dot{\gamma} = 730$ s $^{-1}$ (\blacktriangle), 1070 s $^{-1}$ (\bullet), 1390 s $^{-1}$ (\blacklozenge) and 1760 s $^{-1}$ (\blacksquare) and $\varphi_\infty = 1.7 \times 10^{-3}$ (filled) and 3.3×10^{-3} (open symbols). The exponential curve-fits (lines) have $R^2 > 0.96$. The error bars represent the uncertainty in these data.

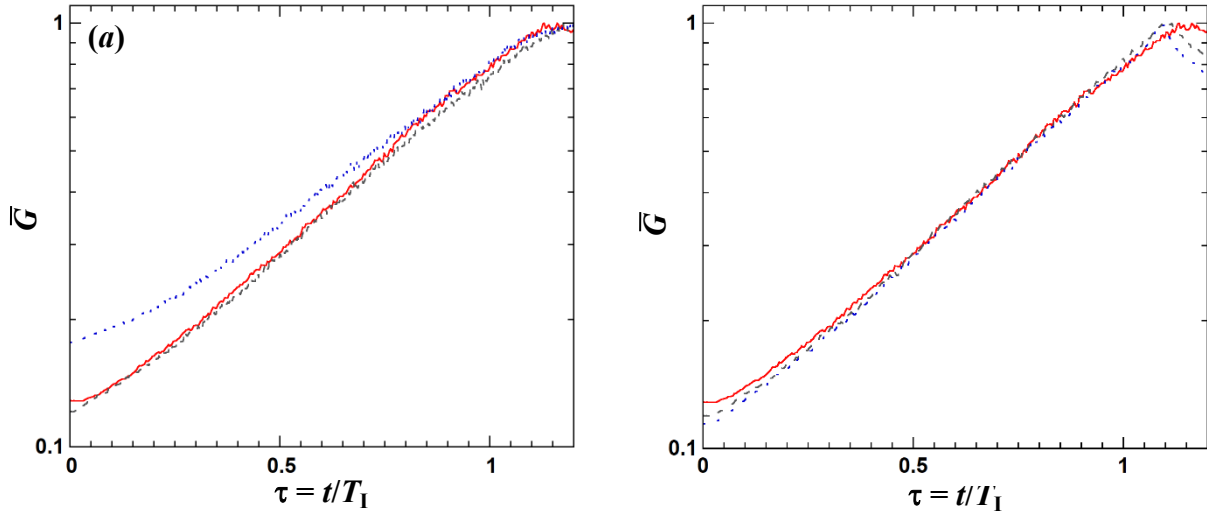


Figure 3 Graphs of \bar{G} (semilog plot) as a function of scaled time $\tau \equiv t/T_1$ at $\Delta E = 4.7$ V/cm for: (a) $(\dot{\gamma}, \varphi_\infty) = (730$ s $^{-1}, 8.5 \times 10^{-4})$ (dotted line), $(730$ s $^{-1}, 1.7 \times 10^{-3})$ (dashed line), and $(1390$ s $^{-1}, 1.7 \times 10^{-3})$ (solid line), all at $x = 6.1$ mm; (b) $(\Delta E, \dot{\gamma}, \varphi_\infty) = (4.7$ V/cm, 1390 s $^{-1}, 8.5 \times 10^{-4})$ at channel streamwise positions $x = 6.1$ mm (solid line), 14.1 mm (dashed line), and 22.1 mm (dotted line).

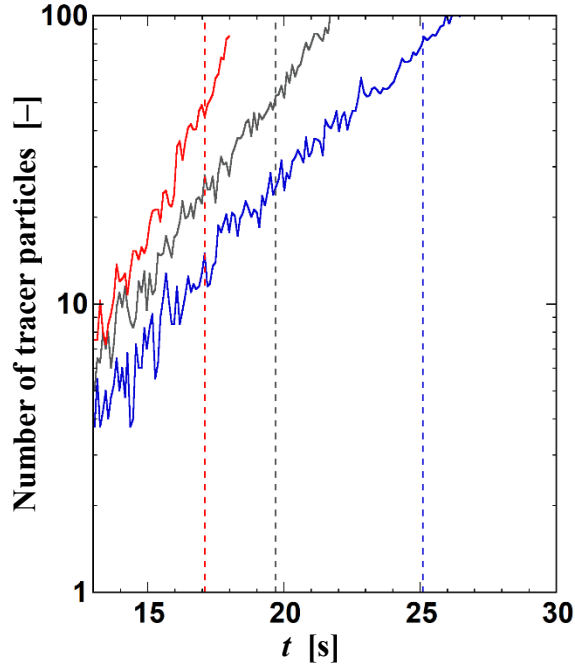


Figure 4 Graph of the number of tracer particles (semilog plot) as a function of (physical) time t for $\dot{\gamma} = 1760 \text{ s}^{-1}$, $x = 14.1 \text{ mm}$ and total particle volume fraction of 1.7×10^{-4} at $\Delta E = 16 \text{ V/cm}$ (**blue**), 40 V/cm (**grey**), and 63 V/cm (**red**). The dashed lines denote T_0 for each case.

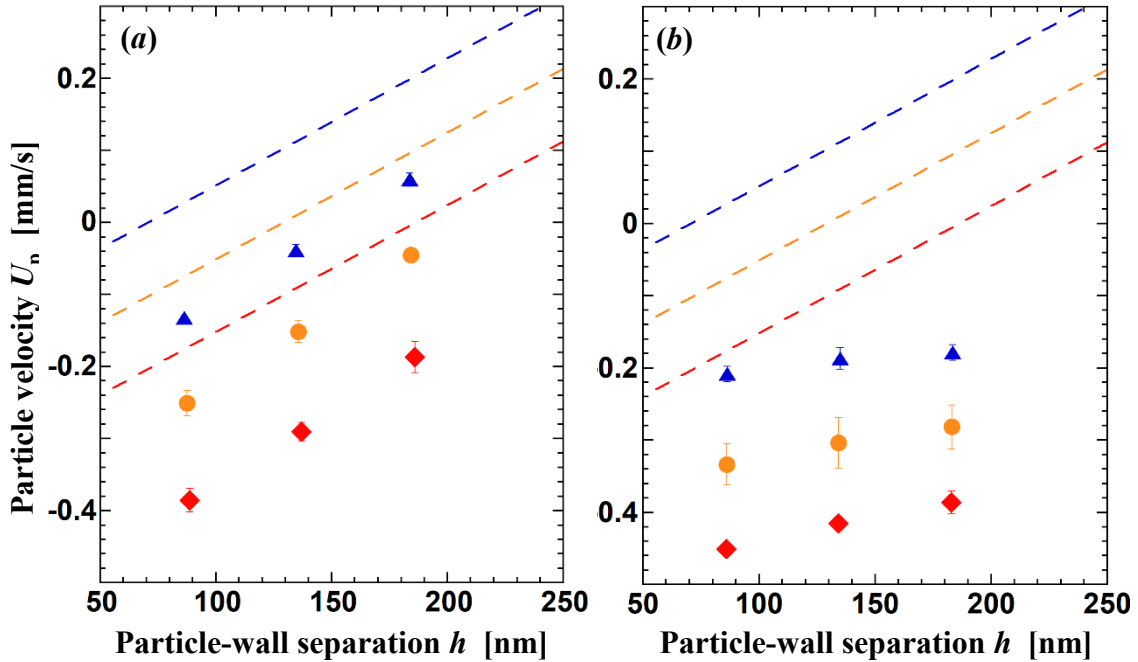


Figure 5 Average tracer particle velocity profiles U_p as a function of particle-wall separation h (where $h = 0$ for a particle touching the wall) for $\dot{\gamma} = 1760 \text{ s}^{-1}$, $x = 14.1 \text{ mm}$ and total particle volume fraction of 1.7×10^{-4} at $\Delta E = 16 \text{ V/cm}$ (**▲**), 40 V/cm (**●**), and 63 V/cm (**◆**) between (a) and within (b) the bands during the steady-state stage. The dashed lines denote the expected particle velocity profiles, corresponding to the superposition of the flow and particle electrophoretic velocities; the error bars denote standard deviations over four realizations.

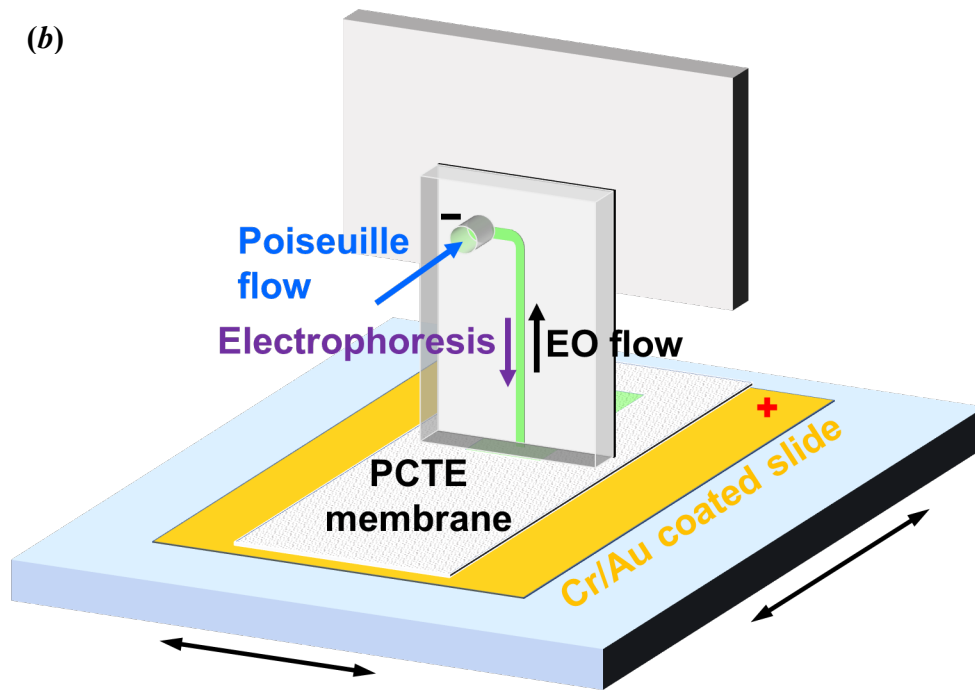
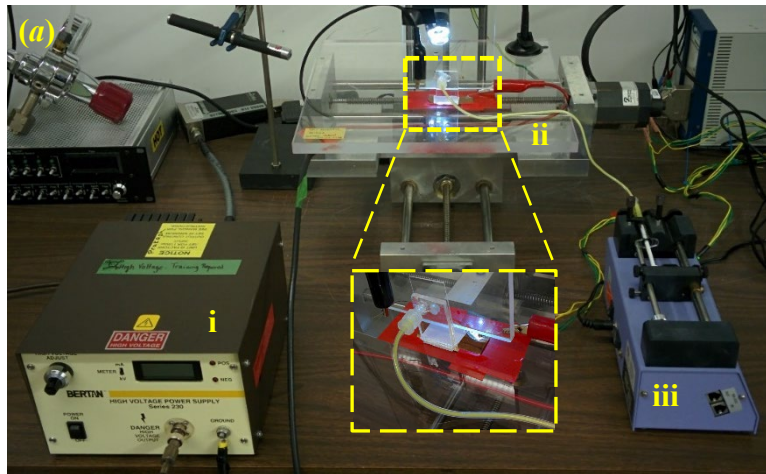


Figure 6 (a) Digital image of the set-up, where label (i) is a high voltage supply, (ii) is an Arduino-controlled device set-up, and (iii) is the syringe pump for introducing a fluid flow from the microchannel outlet to the polycarbonate track etched (PCTE) membrane. Inset shows zoomed image of the microchannel print head, whereas (b) shows the schematic of device arrangement on the stage and the possible flow conditions. Double sided arrows show degrees of freedom for different components.

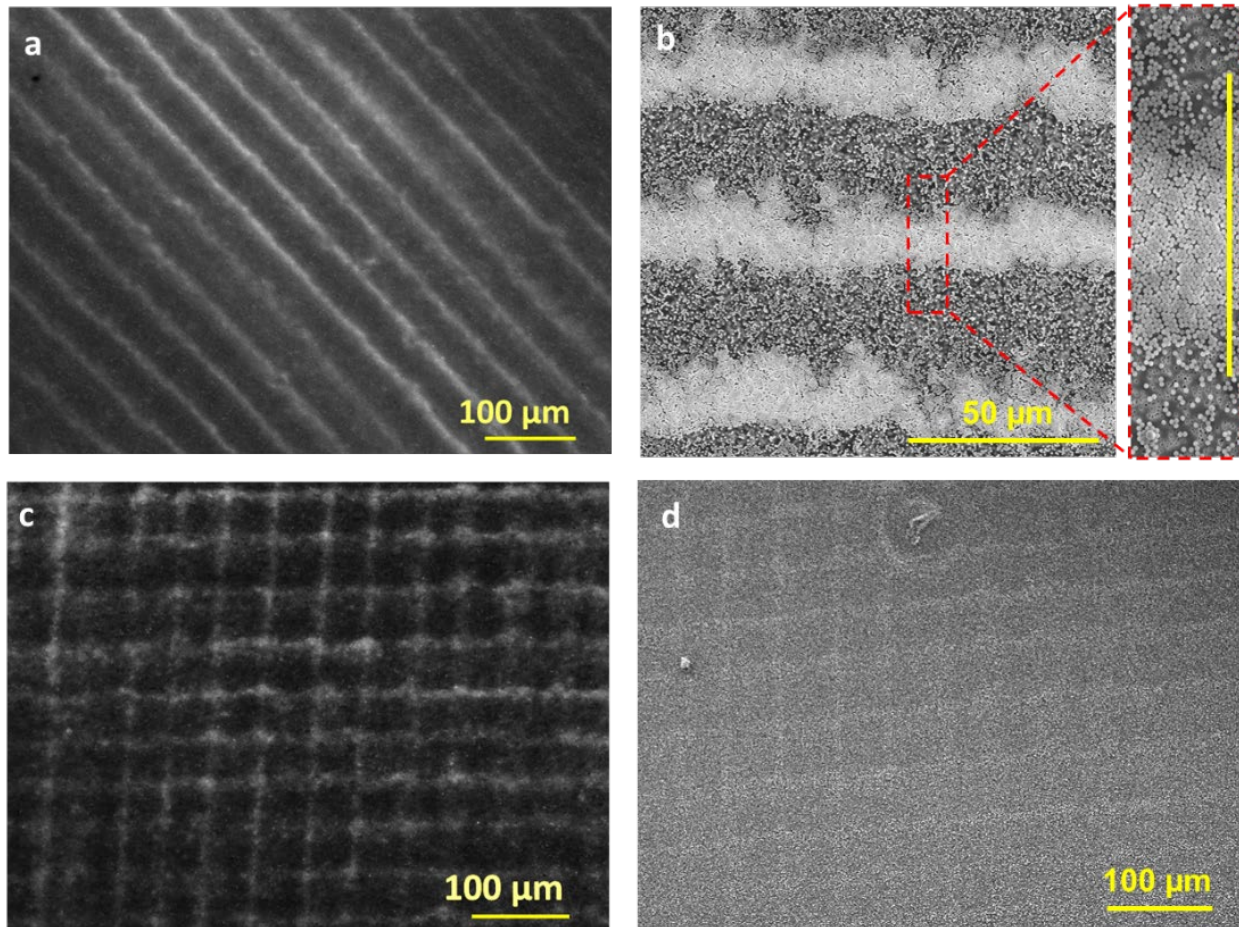


Figure 7 Band structures at a volume flowrate $Q = 1 \mu\text{L}/\text{min}$ and $|E| = 35 \text{ V}/\text{cm}$. (a) Linear array of bands printed at 45° angle to the horizontal. (b) SEM images of the assembled bands, where a small number of colloidal particles are visible between the bands. The inset shows self-assembly of particles within a band, where scale bar is $30 \mu\text{m}$. (c) A grid pattern formed by printing two linear arrays orthogonally. (d) SEM micrograph of the check pattern.

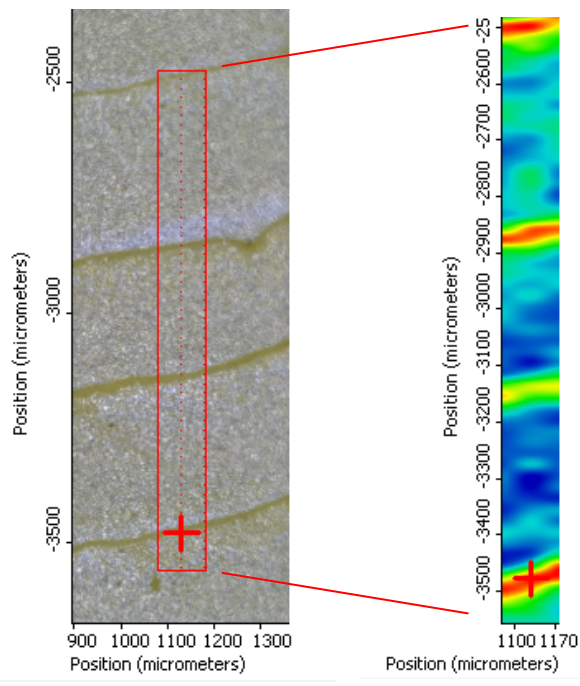


Figure 8 Image of printed bands (yellow lines) [*left*] and FTIR spectrum [*right*] of the region within the red box on the image. The axis labels are the relative positions (in μm) along the physical substrate.