TUNABLE ON-CHIP ASSEMBLY OF COLLOIDAL PARTICLES USING AC ELECTROKINETICS

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APRIL 2019

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by

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Submitted

In fulfilment of the requirement of the degree of Doctor of Philosophy

to the

Indian Institute of Technology Delhi

April 2019

Dedicated to my family

(Parents, Sisters, Akshaya and Arin)

CERTIFICATE

This is to certify that the thesis entitled "***Tunable on-chip assembly of colloidal particles using AC electrokinetics***" being submitted by **Meenal Goel** for the award of the degree of **Doctor of Philosophy** in Chemical Engineering is a record of bonafied work carried out by her under my guidance and supervision at the Indian Institute of Technology Delhi.

The results contained in the thesis have not been submitted elsewhere, either in part or in full, to any other University or Institute for the award of any degree or diploma.

I certify that she has pursued the prescribed course of research.

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ACKNOWLEDGEMENTS

I am highly fortunate to pursue my Ph.D. under the supervision of Dr. Shalini Gupta. I thank her from bottom of my heart for her tremendous patience, encouragement, motivational talks and most importantly her support during my entire Ph.D. tenure. I express my gratitude for her help and long scientific discussions that always lead me in the right direction. I admire her for the kind of relationship she maintains with all of her students and with me. Whenever I was in a bad phase of the life (professionally or personally) she always listened and gave her advice. I will always admire her advices, sincerity and dedication towards science. For all her efforts on my behalf, I thank her and wish her continued success in future endeavours.

I would especially like to express my kind regards to Profs. Anurag Rathore and Sudip Pattanayek from the Department of Chemical Engineering and Dr. Amartya Sengupta from the Department of Physics, IIT Delhi for their constant evaluation of my work, insightful comments and logical suggestions that boosted my confidence for successful completion of all my projects. I would also like to thank Profs. Ravi Elangovan and Atul Narang from the Department of Biochemical Engineering and Biotechnology, IIT Delhi for providing the bacterial strains and Dr. Sameer Sapra from the Department of Chemistry, IIT Delhi for giving the QDs. I am also highly indebted to Dr. Gaurav Goel from the Department Chemical Engineering, IIT Delhi for the helpful technical discussions.

I would like to acknowledge Centre for Excellence in Nanotechnology (CEN) at IIT Bombay for providing the facility for fabrication of microelectrodes. My sincere thanks go to GE team, specifically, Dr. Anette Persson, Dr. Sailesh Bajpai and Dr. Likhesh Sharma for being a part of my third project based on SPR spectroscopy. Although due to technical limitations I could not bring that project to a conclusive end, but all of them gave their constant support and active participation in numerous discussions till the end.

I am thankful to Abhishek Verma, Akshay Singh, Ashwin Bhola and Pragya Swami for their scientific contribution as a team in various projects during the course. I would like to extend my sincere thanks to all my wonderful lab members, particularly to former students Prasanta Kalita and Namrata Pal for exposing me to the realities of the research world and sharing their experiences with me. I am also thankful to Rohini Singh, Pramod Jagtap, Vikas Pandey, Sarul Malik, Gaurav Jhakda, Dr. Puja Prasad and Dr. Shruti Khanna, who unconditionally supported me and never let me feel alone in life. No matter where I go, they will always remain close to my heart.

No words can express my gratitude and appreciation for my parents and for everything they have done for me, especially my mother. My PhD would not have been possible without her continuous motivation and support. Thanking her would not be sufficient for whatever she has done for me- from waking up till midnight, preparing numerous breakfasts and lunches for me in the early cold winter mornings, and most importantly, taking care of my six month old son Arin when I was working till late hours in the lab during the last phase of my PhD. I extend my special thanks to my sisters Ruchi Goyal and Vini Goel and Dheeraj jiju for providing unconditional love and courage to cope up with adverse situations.

Finally but most importantly, my highest gratitude is for my most loving and supporting husband Akshaya who has been my pillar of strength throughout my PhD journey. Akshaya, I thank you for always believing in me, motivating me and for patiently dealing with my countless complaints and dreadful frustrations and most importantly for taking care our loving son Arin when I was not there for him.

Last but not the least, I would like to acknowledge IIT Delhi for giving me an opportunity to continue my research and the funding resources. I would also like to thank

Asian Office of Aerospace Research and Development (AOARD, Japan) for financially supporting (Grant # FA2386-16-1-4072) the work presented in Chapter 5 of this dissertation.

Meenal Goel

ABSTRACT

Controlled patterning of colloidal particles to form one dimensional (1D), 2D and 3D structures is of global interest for many applications like photonic crystals, chemical sensing and bio-electronic devices. In particular, the use of electric fields (EFs) to guide colloidal assembly is a facile way to exploit the nanoscale functionalities of materials in microscopic constructs. In the past, colloids have been organised into various patterns including microwires, nanotriangles, 2D crystals and 3D microstructures using rapid on-chip alternating current (AC) electrokinetics. In particular, AC dielectrophoresis (AC-DEP) and AC electrohydrodynamics (AC-EHD) have been exploited to manipulate particles with high throughput and control. My Ph.D. research work entails understanding the basic mechanisms involved in the colloidal assembly process using these two field-induced forces and applying them to form tunable assemblies from synthetic particles and biological cells.

The first part of my thesis focuses on investigating the parameters involved in AC electrokinetic driven flows and their role in controlling the formation of different gold nanoparticle (AuNP) architectures. Since AC-DEP varies with both particle size and frequency of the field whereas, AC-EHD depends only on the frequency, we hypothesise that there must be a critical particle size at which transition occurs from one type of force to the other. To test this hypothesis, we have carried out both theoretical and experimental analyses to determine the value of this critical size. The effect of field frequency, applied voltage, particle number, electrolyte concentration and electrode geometry and size on the AuNP microstructures have been investigated in detail. Our results indicate that there does exist a size threshold below which AC-EHD dominates, while AC-DEP governs the particle assembly process above it. The EHD force leads to film formation and the DEP force results into microwires. To initiate the assembly process a minimum particle number, a low electrolyte concentration and an optimum frequency range is required. The microwires grow

in two different modes: surface or bulk depending on the frequency of the field, and never short circuit being on the same potential.

The second part of my thesis involves AC-DEP driven fabrication of “0D” cellular microarrays of live bacterial cells with tunable densities at a single cell level. We have performed detailed optimization of electrical parameters of the microarray chip to achieve maximum cell capture efficiency. The microarrays have also been used to carry out phenotypic assessment, cell counting in mixtures and real-time viability studies in the presence of antibiotics using optical microscopy and impedance spectroscopy. We believe that our research paves ways for developing new platforms for multiplexed sensing, heterogeneous cell population studies and ultrafast drug susceptibility testing to reduce the use of broad-spectrum antibiotics and hence, the emergence of new antibiotic resistant strains.

सार

एक आयामी (1D), 2 डी और 3 डी संरचनाओं को बनाने के लिए कोलाइडल कणों की नियंत्रित पैटर्निंग कई अनुप्रयोगों जैसे फोटोनिक क्रिस्टल, रासायनिक संवेदन और जैव-इलेक्ट्रॉनिक उपकरणों के लिए वैश्विक हित की है। विशेष रूप से, कोलाइडल असेंबली का मार्गदर्शन करने के लिए विद्युत क्षेत्रों (ईएफ) का उपयोग सूक्ष्म निर्माणों में सामग्री के नैनोस्केल कार्यात्मकताओं का फायदा उठाने का एक शानदार तरीका है। अतीत में, रैपिड ऑन-चिप अल्टरनेटिंग करंट (एसी) इलेक्ट्रोकेनेटिक्स का उपयोग करते हुए कोलोइड्स को विभिन्न पैटर्न में व्यवस्थित किया गया है, जैसे कि, माइक्रोवेयर्स, नैनोट्रियैंगल्स, 2 डी क्रिस्टल और 3 डी माइक्रोस्ट्रक्चर। विशेष रूप से, एसी डाइलेरोप्रोफोरेसिस (एसी-डीईपी) और एसी इलेक्ट्रो-हाइड्रोडायनामिक्स (एसी-ईएचडी) का उपयोग उच्च थ्रूपुट और नियंत्रण के साथ कणों में हेरफेर करने के लिए किया गया है। मेरी पीएच.डी. अनुसंधान कार्य इन दो क्षेत्र-प्रेरित बलों का उपयोग करके कोलाइडल असेंबली प्रक्रिया में शामिल बुनियादी तंत्रों को समझने और सिंथेटिक कणों और जैविक कोशिकाओं से ट्यून करने योग्य विधानसभाओं को बनाने के लिए उन्हें लागू करने पर जोर देता है।

मेरी थीसिस का पहला भाग एसी इलेक्ट्रोकेनेटिक संचालित प्रवाह में शामिल मापदंडों की जांच करने और विभिन्न स्वर्ण नैनोपार्टिकल (एयूएनपी) आर्किटेक्चर के गठन को नियंत्रित करने में उनकी भूमिका पर केंद्रित है। चूंकि एसी-डीईपी कण आकार और विद्युत

क्षेत्र की आवृत्ति दोनों के साथ भिन्न होता है, जबकि एसी-ईएचडी केवल आवृत्ति पर निर्भर करता है, हम परिकल्पना करते हैं कि एक महत्वपूर्ण कण आकार होना चाहिए, जिस पर संक्रमण एक प्रकार के बल से दूसरे में होता है। इस परिकल्पना का परीक्षण करने के लिए और इस महत्वपूर्ण आकार के मूल्य को निर्धारित करने के लिए हमने सैद्धांतिक और प्रयोगात्मक दोनों विश्लेषण किए हैं। एयूएनपी माइक्रोस्ट्रक्चर पर विद्युत क्षेत्र की आवृत्ति, लागू वोल्टेज, कण संख्या, इलेक्ट्रोलाइट एकाग्रता और इलेक्ट्रोड ज्यामिति और आकार के प्रभाव की विस्तार से जांच की गई है। हमारे परिणाम दर्शाते हैं कि एक आकार सीमा मौजूद है जिसके नीचे एसी-ईएचडी हावी है, जबकि एसी डीईपी इसके ऊपर कण विधानसभा प्रक्रिया को नियंत्रित करता है। ईएचडी बल फिल्म का निर्माण करता है और डीईपी बल सूक्ष्मतारों का। छलरचना प्रक्रिया को आरंभ करने के लिए एक न्यूनतम कण संख्या, एक कम इलेक्ट्रोलाइट सांद्रता और एक इष्टतम आवृत्ति रेंज की आवश्यकता होती है। विद्युत क्षेत्र की आवृत्ति के आधार पर माइक्रोवेर्स दो अलग-अलग मोड में बढ़ते हैं: सतह या बल्क, और एक ही वोल्टेज पर होने के कारण वे कभी भी शॉर्ट सर्किट नहीं होते हैं।

मेरी थीसिस के दूसरे भाग में एसी डीईपी का उपयोग कर जीवित जीवाणुओं के “0डी” माइक्रोएरे बनाना शामिल है। हम इसका उपयोग एकल कोशिका स्तर के अध्ययन के लिए कर सकते हैं। हमने अधिकतम सेल कैप्चर दक्षता प्राप्त करने के लिए माइक्रोएरे चिप के विद्युत मापदंडों का विस्तृत अनुकूलन किया है। माइक्रोएरे का उपयोग फेनोटाइपिक

मूल्यांकन, मिश्रणों में कोशिका गणना और ऑप्टिकल माइक्रोस्कोपी और प्रतिबाधा स्पेक्ट्रोस्कोपी का उपयोग करके एंटीबायोटिक दवाओं की उपस्थिति में वास्तविक समय व्यवहार्यता अध्ययन के लिए किया गया है। हमारा मानना है कि हमारे शोध में बहुसंकेतन संवेदन, विषम सेल जनसंख्या अध्ययन और अल्ट्राफास्ट दवा संवेदनशीलता परीक्षण के लिए नए प्लेटफार्मों को विकसित करने के तरीके हैं जिससे व्यापक स्पेक्ट्रम एंटीबायोटिक दवाओं के उपयोग को और नए एंटीबायोटिक प्रतिरोधी उपभेदों के उद्भव को कम किया जा सकता है।

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DEFINITIONS OF TERMS AND SYMBOLS USED IN THE TEXT

Symbol	Notes	Units
e	Elementary charge with a value equal to $1.602176634\times10^{-19}$ C	Coulomb, C (As)
z_i	Valence of ions, where, $z = \frac{q}{e}$	
k_B	Boltzmann constant equal to 1.38064852×10^{-23} J/K	Joules/Kelvin, JK^{-1}
T	Absolute temperature	Kelvin, K
n_i	Number concentration of ions	L^{-1}
φ	Surface potential	Volts, V (A^{-1} kg m^2 s^{-3})
ζ	Zeta potential	Volts, V (A^{-1} kg m^2 s^{-3})
κ^{-1}	Debye length	Meter, m
ε	Dielectric permittivity	Farad/meter, Fm^{-1} (A^2 kg^{-1} m^{-3} s^4)
η	Viscosity	Pascal.s, Pa.s (kg m^{-1} s^{-1})
E_0	Electric field	Volt/meter, Vm^{-1}
V	Electric potential	Volt, V
v	Velocity	Meter/s, ms^{-1}
μ	Electrophoretic mobility	m^2 V^{-1} s^{-1}
K	Clausius–Mossotti (CM) factor	unit less
σ	Electrical conductivity	Siemens/meter, Sm^{-1}
τ_{MW}	Maxwell-Wagner charge relaxation time	Second, s
ω	Frequency	Hertz, Hz
ω_C	Crossover frequency is the frequency at which DEP force changes it sign from positive to negative	Hertz, Hz
π	A dimensionless quantity written as pi with value equal to 3.14	
r	Particle radius	Meter, m

ABBREVIATIONS

AC – Alternating current

DC- Direct current

NPs – Nanoparticles

AuNPs – Gold nanoparticles

CC – Colloidal crystals

EF – Electric field

EHD – Electrohydrodynamics

DEP – Dielectrophoresis

EO – Electroosmosis

EP – Electrophoresis

F_{EHD}– Electrohydrodynamic force

F_{DEP}– Dielectrophoretic force

F_B– Brownian force

F_{dip} – Dipole-dipole attractive force

d_c – critical distance

a_c - Critical interparticle distance

FCC – Face centered cubic

PS - Polystyrene

PMMA - Polymethyl methacrylate

SPR - Surface plasmon resonance

LbL – Layer-by-layer

wrt – with respect to

LB – Langmuir Blodgett

CNTs – Carbon nano tubes

ITO – Indium tin oxide

PDMS - Polydimethylsiloxane

DNQ – Diazonaphthoquinone

OSTE - Off-stoichiometry thiolene

IPA – Isopropyl alcohol

IPTG - Isopropyl β-D-1-thiogalactopyranoside

HEPES - 4-(2-hydroxyethyl)-1-piperazine ethanesulfonic acid

EDL - Electrical double layer

H – Magnetic field

m – Magnetic dipole

QDs – Quantum dots

FEM - Finite element method

S.typhi - *Salmonella typhi*

E.coli - *Escherichia coli*

μTAS - Micro total analysis systems

HP - Helmoltz plane

iHP – Inner Helmoltz plane

oHP – Outer Helmoltz plane

SP – Slip plane

U_{eo} - Electroosmotic velocity

U_{ep}- Electrophoretic velocity

CM - Clausius–Mossotti function

ROT – Electrorotation

tw-DEP - Travelling wave dielectrophoresis

nDEP – Negative dielectrophoresis

pDEP – Positive dielectrophoresis

ICEO - Induced charge electroosmosis

IDE – Interdigitated electrode

μCP - Micro contact printing

EBL – Electron beam lithography

SAM - Self-assembled monolayer

PR – photoresist

UV – Ultraviolet

rpm – Revolutions per minute

rcf – Relative centrifugal force

SERS - Surface-enhanced Raman scattering

OD – Optical density

YFP – Yellow fluorescent protein

FoV – Field of view

ELISA – Enzyme linked immunosorbant assay

PCR – Polymerase chain reaction

LPS – Lipopolysaccharide