

TUNABLE ON-CHIP ASSEMBLY OF COLLOIDAL PARTICLES USING AC ELECTROKINETICS

MEENAL GOEL



**DEPARTMENT OF CHEMICAL ENGINEERING
INDIAN INSTITUTE OF TECHNOLOGY DELHI**

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TUNABLE ON-CHIP ASSEMBLY OF COLLOIDAL PARTICLES USING AC ELECTROKINETICS

by

Meenal Goel

Department of Chemical Engineering

Submitted

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

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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.

Dr. Shalini Gupta

Associate Professor

Department of Chemical Engineering

Indian Institute of Technology Delhi

Hauz Khas, New Delhi -110016

India

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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, नैनोट्रियेंगल्स, 2 डी क्रिस्टल और 3 डी माइक्रोस्ट्रक्चर। विशेष रूप से, एसी डाइलेक्ट्रोफोरेसिस (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 कैपचर 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।

TABLE OF CONTENTS

CERTIFICATE	i
ACKNOWLEDGEMENTS	iii
ABSTRACT	vi
TABLE OF CONTENTS	xi
LIST OF FIGURES	xiv
LIST OF TABLES	xix
DEFINITIONS OF TERMS AND SYMBOLS USED IN THE TEXT	xx
ABBREVIATIONS	xxi
Chapter 1: INTRODUCTION	1
1.1 INSPIRATION FROM NATURE’S GALLERY	2
1.2 WHAT ARE COLLOIDS?	5
1.3 COLLOIDAL ASSEMBLY	6
1.4 TECHNIQUES FOR COLLOIDAL SELF-ASSEMBLY	7
1.4.1 Evaporation	7
1.4.2 Layer-by-layer (LbL)	8
1.4.3 Spin coating	9
1.4.4 Langmuir-Blodgett (LB)	9
1.4.5 Template-assisted	10
1.4.6 Emulsion droplets	11
1.5 FIELD-ASSISTED DIRECTED ASSEMBLY OF COLLOIDAL PARTICLES	11
1.5.1 Optical field	12
1.5.2 Magnetic field	13
1.5.3 Electric field (EF)	14
1.6 ADVANTAGES OF ELECTRIC FIELDS OVER OTHER TECHNIQUES FOR COLLOIDAL ASSEMBLY	16
1.7 THESIS OBJECTIVES	17
1.8 THESIS LAYOUT	19
1.9 REFERENCES	20
Chapter 2: LITERATURE REVIEW AND THEORY OF AC ELECTROKINETICS	40
2.1 BACKGROUND	41
2.2 THEORY OF ELECTRICAL DOUBLE LAYER (EDL)	42
2.3 ELECTROKINETIC PHENOMENA IN AN AC FIELD	47

2.3.1 Dielectrophoresis (DEP).....	47
2.3.2 Electrorotation (ROT) and travelling wave DEP (tw-DEP)	50
2.3.3 Manipulation of particles using DEP	50
2.3.4 Manipulation of particles using AC electrohydrodynamics (EHD).....	52
2.4 REFERENCES	56
Chapter 3: MICROWELL ELECTRODE FABRICATION	66
3.1 INTRODUCTION	67
3.2 EXPERIMENTAL SECTION	74
3.2.1 Materials	74
3.2.2 Mask fabrication.....	74
3.2.3 Electrode fabrication	75
3.3 RESULTS AND DISCUSSION.....	76
3.3.1 Effect of spin speed and time	76
3.3.2 Effect of prebaking.....	78
3.3.3 Effect of UV dosage	79
3.4 CONCLUSIONS	83
3.5 REFERENCES	84
Chapter 4: SIZE-TUNABLE ASSEMBLY OF GOLD NANOPARTICLES USING COMPETITIVE AC ELECTROKINETICS*	89
4.1 INTRODUCTION	90
4.2 EXPERIMENTAL SECTION	92
4.2.1 Materials	92
4.2.2 Methods	92
4.2.2.1 AuNP suspensions	92
4.2.2.2 Experimental setup.....	94
4.2.2.3 Electrostatic simulations	94
4.3 RESULTS	95
4.3.1 Manifestation of particle size on colloidal structure morphology	98
4.3.2 Effect of frequency and voltage	101
4.3.3 Control experiments	104
4.3.3.1 Effect of salt	104
4.3.3.2 Effect of electrode size.....	106
4.3.3.3 Effect of electrode geometry	107
4.3.3.4 Effect of particle concentration	108

4.3.4 Microstructural characterization	111
4.4 DISCUSSION	112
4.5 CONCLUSIONS	117
4.6 REFERENCES	119
Chapter 5: EF-DRIVEN ASSEMBLY OF LIVE BACTERIAL CELL MICROARRAYS	123
5.1 INTRODUCTION	124
5.2 DEP AS A TOOL FOR BACTERIAL COLLECTION	125
5.3 EXPERIMENTAL SECTION	127
5.3.1 Materials	127
5.3.2 Methods	127
5.3.2.1 Bacterial cell culture and sample preparation	127
5.3.2.2 Experimental setup.....	128
5.3.2.3 Cell viability testing	129
5.3.2.4 Impedance measurements	129
5.4 RESULTS AND DISCUSSION.....	129
5.4.1 Calibration-free estimation of cell numbers in two-component mixtures.....	134
5.4.2 Assessment of cell viability in the presence of antibiotic via impedance spectroscopy	136
5.5 CONCLUSIONS	140
5.6 REFERENCES	142
Chapter 6: SUMMARY AND FUTURE OUTLOOK.....	147
6.1 REFERENCES	151
Appendix A: Mathematical calculations used in the thesis.....	152
A.1 Calculation of EHD, DEP and Brownian forces	153
A.2 Calculation of temperature rise of the fluid ¹	154
A.3 Calculation of polydispersity index (PDI) ²	154
A.4 Calculation of the critical interparticle distance between colloids	155
A.5 Calculation of chip collection efficiency	156
A.6 REFERENCES	157
BIODATA	158

LIST OF FIGURES

Figure 1.1 Examples of patterns found in nature that have inspired scientists to develop new products.(A) Sand dunes,³³ (B) cracked earth,³⁴ (C) cloud formation,³⁵(D) patterns formed on the skin of giraffes,³⁶(E) antireflective moth eyes,⁶ (F) ordering of petals in sunflowers³⁷, fractal patterns formed in (G) snowflakes¹⁰ and (H) bacteria colonies¹¹ (I) lamellar pattern formed by polystyrene-polyisoprene block copolymer³⁸ and (J) quasi-crystal formed in AlCuFe alloy.³⁹ 3

Figure 1.2 Schematic of the organization of nanoparticles in 1D chains, 2D sheets and 3D structures..... 6

Figure 1.3 Non-external field mediated assemblies of colloids. Evaporation induced 2D assembly of colloids in (A) vertical and (B) horizontal deposition mode.¹⁰² (C) LB technique for the assembly of 1D colloidal crystal arrays. (D) SEM micrograph of a 1D colloidal string.¹⁰⁰ (E) Template assisted assembly of 2D CdSe nanorods.¹⁵⁰ (F) Emulsion assisted assembly of particles collected around oil-in-water or water-in-oil droplets.¹⁵⁷ (G) Semipermeable¹⁵⁸ and (H) hairy colloidosomes.¹⁵⁹ 12

Figure 1.4 Field assisted assembly of colloids. (A)¹⁷⁵and (B)¹⁷⁶ Optically controlled assembly. (C) Distribution of magnetic field and dipolar interaction between two magnetic particles.¹⁸³ SEM image of (D) 1D magnetic chains¹⁸⁵ and (E) 2D magnetic walls.¹⁸⁴ (F, G) Flower structures formed from the mixture of diamagnetic and paramagnetic spherical particles within a magnetized fluid.¹⁸⁶ (H) EF driven branched microwires of metallic nanoparticles²⁰⁵ and (I) microwires of CdSe QDs.²⁰⁶ (J) Collection of *E.coli* near castellated electrode edges.²⁰⁸ (K) Bridge formed by CNTs²⁰⁷ and (L) 2D assembly of latex particles.²¹⁰ 15

Figure 2.1 Schematic illustration of formation of an electrical double layer (EDL) on a negatively charged interface based on Gouy-Chapman-Stern model and variation of potential with distance from the surface. 44

Figure 2.2 DEP assisted colloidal assemblies. (A-B) 2D and 3D assembly of polystyrene particles,⁵¹ (C) AuNPs microwires,³² (D) chain formation in latex particles,⁵³ (E) collection of latex microspheres near the electrode edges,⁵² (F) chain formation in yeast cells,⁶⁴ (G) collection of protein particles⁶⁶ and (H) electropatterning of HeLa cells on microwell array electrodes.⁷¹ 51

Figure 2.3 Origin of AC EHD effect. The tangential EF present on the electrodes near to the edges results in EHD flow. The sign of the counterions in the double layer formed on the electrodes and the EF change simultaneously in each half cycle resulting in net fluid flow directed towards the electrodes (adapted from¹³). 53

Figure 2.4 EHD mediated assembly of (A) latex particles¹³ and (B) yeast cells on conductive corrals,¹³ (C-D) polystyrene particles and (E) AuNPs microwires between coplanar electrodes.^{50,106,107} 55

Figure 3.1 Schematic for the fabrication of PDMS stamp and μ CP on a planar substrate with a (A) planar and (B) rolling PDMS stamp.^{20,21} 68

Figure 3.2 Components of laser micromachining tool. ³⁹	69
Figure 3.3 Chemical structure of PMMA. ⁴⁶	70
Figure 3.4 Reaction chemistries of (A) positive PR DNQ and (B) negative PR SU8 upon UV irradiation. ^{49,53}	72
Figure 3.5 Schematic showing the difference between negative and positive photolithography methods. ⁵⁰	73
Figure 3.6 The chemical structure of SU8 epoxy (glycidyl-ether-bisphenol-A novolac). ⁵² ..	73
Figure 3.7 (A) A mask design prepared using Clewin4 software and containing ~9600 microwell electrodes, each of diameter (D) 50 μm and electrode edge-to-edge spacing (S) of 50 μm . (B) A brightfield optical micrograph of an actual mask plate used for our photolithography process.	75
Figure 3.8 Thickness of the PR measured using profilometer at three different spinning conditions.	78
Figure 3.9 Optical micrographs of underdeveloped microwells with (A) D100 S75, (B) D75 S75 and (C) D50 S100 when the PR coated substrate was exposed to UV light at 150 mJ/cm^2	80
Figure 3.10 Optical micrographs of developed microwells with (A) D100 S75 and (B) D75 S75 and (C) D50 S50 after exposing the PR coated substrate to UV light at 120 mJ/cm^2	80
Figure 3.11 Optical micrographs of developed microwells with (A) D50 S100, (B) D50 S75 and (C) D50 S50 and under developed microwells with (D) D25 S75 and (E) D25 S50 when the PR coated substrate was exposed to UV light at 60 mJ/cm^2	82
Figure 3.12 Optical images of perfectly developed (A) D25 S75 and (B) D25 S50, poorly developed (C) D10 S50 and (D) D5 S5 and developed (E) D10 S50 and less developed (F) D5 S5 microwells fabricated on different substrates and exposed to UV doses of 50 mJ/cm^2 (A-D) and 40 mJ/cm^2 (E-F).	83
Figure 4.1 Geometrical layout of coplanar and microwell electrodes.	91
Figure 4.2 Schematic of the experimental setup used for our AuNP microwire and film assembly: (A) 2D and (B) 3D side view.	96
Figure 4.3 (A) Charge and EF distribution for positive half-cycle of the applied AC field. (B) The direction of particle collection due to EHD (FEHD) and DEP (FDEP) forces. (C) The effect of particle size on the DEP and EHD forces assuming no size dependence on FEHD. (D) Spatial variation in the x-components of DEP and EHD forces exerted on a 6 μm size particle kept 1 μm above the bottom surface.	97
Figure 4.4 Variation of DEP (x component) and EHD force with particle size for coordinates (A) (-2,1), (B) (3,1), (C) (10,1) and (D) (0,3) where (0,0) is the coordinate of the microwell edge and positive x-axis points towards the centre of the microwell.	98

Figure 4.5 TEM (A&B) and optical (C&D) micrographs of AuNPs used in our experimental study with their respective particle size distribution curves below. Average particle sizes were (A) 17 ± 4 nm, (B) 130 ± 170 nm, (C) 5 ± 3 μ m and (D) 10 ± 6 μ m. Images in (C) and (D) are hazy because some particles are out of focus in the liquid sample..... 99

Figure 4.6 Particle size distribution in AuNP suspensions before and after centrifugation using (A) TEM (14.28 ± 3.91 nm before; 17.25 ± 3.21 nm after), (B) DLS (37.8 ± 0.6 nm before; 42.1 ± 0.44 nm after) and (C) UV-visible spectroscopy (16.99 nm before; 16.44 nm after). These experiments were performed after extensive dilution of the suspensions in each case and hence, allowed estimation of the size reversibility aspect. For concentrations at which EF experiments were performed, the sizes were estimated without dilution using TEM or optical microscopy. For estimating the particle size using UV-vis data, the formula used was d (nm) = $\exp(3\lambda_{\max}521\lambda_{\max}450 - 2.2)$.²⁹ 100

Figure 4.7 Time-lapsed images of (A) AuNP films formed using 17 nm sized AuNPs and (B) microwires formed using 10 μ m sized AuNPs at 20 V_{pp} and 100 Hz. 101

Figure 4.8 (A) Frequency-dependent assembly in 0.5 nM AuNP suspension. (B) A representative TEM micrograph and the corresponding particle size distribution. The average particle size was found to be 20 ± 7 nm which was quite similar to that for 2 nM suspension as shown in Fig. 4.5A..... 102

Figure 4.9 Effect of applied voltage on the rate of assembly of (A) films using 17 nm sized AuNPs and (B) microwires using 10 μ m sized Au suspension. All images were taken at 100 Hz, 5 min after the start of the experiment. 102

Figure 4.10 Size and frequency dependent phase chart of film to microwire transition. The EF applied was 20 V_{pp} for 5 min. 103

Figure 4.11 Film deposition in the presence of NaCl for 17 nm AuNPs: (A) 1 mM, (B) 10 mM and (C) 20 mM. The EF was 100 Hz and 20 V_{pp}..... 105

Figure 4.12 (A) Effect of salt concentration on the stability of 17 nm sized AuNPs. The 1 and 10 mM NaCl suspensions were stable for many days whereas, (B) Image of 20 mM aggregated just after one day. The particles were highly unstable in the presence of 50 mM NaCl and immediately aggregated upon addition of salt (data not shown). The average particle sizes estimated from these spectra were as follows: 17 nm (No NaCl), 10 nm (1 mM NaCl), 11 nm (10 mM NaCl) and 9 nm (20 mM NaCl).²⁹ 106

Figure 4.13 Effect of electrode dimension. (A) AuNP assemblies obtained for different particle sizes using electrodes with well diameter (D) equal to 22 μ m and edge-to-edge spacing (S) 45 μ m. The EF applied was 20 V_{pp} and 100 Hz. (B) The new curve showing upward shift in critical particle diameter to ~ 8 μ m..... 107

Figure 4.14 Effect of electrode geometry. (A) Assembly of 17 nm and 10 μ m sized AuNP suspensions using interdigitated electrodes (IDEs) and 5V_{pp}, 100 Hz AC field. Voltages higher than 5 V_{pp} led to bubble formation. (B) Schematic of the directions of the normal and tangential EFs along with the direction of the EHD and DEP forces on the IDEs. (C) Assembly of water-soluble CdTe QDs of size 3.5 ± 0.5 nm and thioglycolic acid (TGA) cap using 50 μ m gap IDEs and an EF of strength 10 V_{pp}, 10 kHz. The brightly lit regions represent the electrodes whereas, the gap between the electrodes contain microwires. 108

Figure 4.15 Particle size irreversibility in case of salt-aggregated Au NPs. (A) Size distribution of Au particles aggregated via salt addition at two different particle volume fractions, $\phi_1 = 3.1 \times 10^{-4} \%$ and $\phi_2 = 3.1 \times 10^{-3} \%$ (v/v). Their corresponding optical micrographs are shown in (B) and (C), respectively. The particle size distributions were estimated from ~ 1000 particles in case of ϕ_1 and ~ 5000 particles in ϕ_2 . The average particle size in both cases was $6 \pm 4 \mu\text{m}$.	109
Figure 4.16 Effect of particle size at constant volume fraction on EF-driven morphology. (A&B) $\phi_1 = 3.1 \times 10^{-4} \%$ and (C&D) $\phi_2 = 7.7 \times 10^{-3} \%$ (v/v). The average particle sizes were (A) $17 \pm 4 \text{ nm}$, (B) $6 \pm 4 \mu\text{m}$, (C) $< 1 \mu\text{m}$ and (D) $5 \pm 3 \mu\text{m}$. An EF of strength $20 V_{pp}$, 100 Hz was applied for 5 min in case of (A), (C) & (D) and 15 min in case of (B). Longer time was given to (B) to overcome any kinetic-limitations.	110
Figure 4.17 The effect of particle preparation method on the morphology of the assembled particles. The particles were prepared by (A) filter-assisted centrifugation and (B) salt-assisted aggregation of AuNPs. In both cases, no significant particle collection was seen (transition zone). Both (A) and (B) used similar sized particles (average $\sim 5 \mu\text{m}$) and volume fraction ($7.7 \times 10^{-3} \%$ v/v) and the EF applied was $20 V_{pp}$, 100 Hz for 5 min in (A) and 15 min in (B).	110
Figure 4.18 Thickness measurement of the AuNP film using (A) optical profiler and (B) SEM, deposited on the microwell surface after applying an EF at $20 V_{pp}$ and 500 Hz for 10 min.	111
Figure 4.19 Stability of the Au films after two consecutive washes with MQ, IPA and 70 % (v/v) ethanol.	112
Figure 4.20 Instability of Au microwires formed at 1 kHz, $20 V_{pp}$: (A) before wash, (B) after 1 st wash and (C) after 2 nd wash with DI water.	112
Figure 4.21 (A) Schematic for arrangement of particles in a box and (B) Variation of F_{dip}/F_B with interparticle distance, a .	114
Figure 4.22 The theoretical curve for real part of the CM factor $\text{Re}[K(\omega)]$ versus frequency for AuNPs in water. ³¹	116
Figure 5.1 (A) Variation of CM factor with frequency in case of bacteria based on a two-shell dielectric model, specifying the contribution of each subcellular component at different regions. ²⁷ EHD and DEP controlled collection of (B) AuNPs and (C) cells at different frequencies.	126
Figure 5.2 (A) Schematic of the experimental setup. Collection of <i>S.typhi</i> cells into the microwells (B) before and (C) after applying EF for 1 h ($20 V_{pp}$, 5 MHz). The cell concentration used was 10^6 cells/mL .	129
Figure 5.3 Settling of <i>S.typhi</i> cells (10^6 cells/mL) on the electrode surface (A) $t = 0 \text{ min}$ and (B) $t = 60 \text{ min}$ due to gravitational field.	130
Figure 5.4 Phase diagram for positive DEP cell response of <i>S.typhi</i> cells. The results were obtained after 15 min of EF exposure on 10^6 cells/mL at each frequency-voltage.	131

Figure 5.5 Assembly of tunable density microarrays (20 V _{pp} ; 60 min; 3x10 ⁶ cells/mL). (A) Frequency-dependent non-monotonic collection behaviour of <i>S.typhi</i> cells. (B) Spatio-temporal distribution of cells across different wells at 20 MHz.....	132
Figure 5.6 Cell viability of 10 ⁶ <i>S.typhi</i> cells/mL with and without applying AC EF (20 V _{pp} , 5 MHz) for 60 min.	132
Figure 5.7 Time-lapsed collection of <i>S.typhi</i> cells on the microelectrode array at 20 MHz and 20 V _{pp} for two different bulk concentrations.	133
Figure 5.8 Calibration-free cell identification in binary mixtures. (A) Brightfield image of both cell types entrapped inside the microwells (inset shows a zoomed in view of a particular section). (B) Complementary fluorescence image depicting only YFP-expressed <i>E.coli</i> cells (highlighted in circles). (C) Data showing good correlation (R ² = 0.99) between initial and final ratios obtained after collection (20 V _{pp} , 5 MHz; 1 h; 10 ⁶ cells/mL).....	136
Figure 5.9 Time-lapsed response of <i>S.typhi</i> cells in the presence of different concentrations of antimicrobial drug (A) with and (B) without EF. The ΔI values on the Y-axis represent the relative impedance change since the start of the incubation process. The line break on the X-axis in (A) includes 20 min of incubation followed by 15 min of AC field (20 V _{pp} , 5 MHz) and in (B) includes 35 min of incubation without any EF.	138
Figure 5.10 Cell viability testing using impedance spectroscopy in the presence and absence of DEP (20 V _{pp} , 5 MHz). The data are plotted for 45 min after drug incubation.....	139

LIST OF TABLES

Table 3.1 Variation of PR thickness with spinning conditions. Each experiment was repeated twice and the thickness of the film was reported as an average from these two sets ± 1 SD..	77
Table 3.2 Dependence of feature size of microwells on UV exposure dose.	82
Table 3.3 Variation of diameter (D) and edge-to-edge spacing (S) in different microwell electrodes.	82
Table A.1 Values of the experimental parameters used in our calculations. It may be noted that $\kappa - 1 = 0.96 \mu\text{m}$ for water at pH 7 but can reduce to $0.39 \mu\text{m}$ if the pH drops to 5.93 due to dissolved CO_2	154
Table A.2 Polydispersity index values of AuNPs at different sizes.....	155

DEFINITIONS OF TERMS AND SYMBOLS USED IN THE TEXT

Symbol	Notes	Units
e	Elementary charge with a value equal to $1.602176634 \times 10^{-19}$ C	Coulomb, C (As)
z_i	Valence of ions, where, $z = \frac{q}{e}$	
k_B	Boltzmann constant equal to $1.38064852 \times 10^{-23}$ 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 its 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*

μ TAS - Micro total analysis systems

HP - Helmholtz plane

iHP – Inner Helmholtz plane

oHP – Outer Helmholtz 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