Mikko Haapalainen

DIELECTROPHORETIC MOBILITY OF A SPHERICAL PARTICLE IN 2D HYPERBOLIC QUADRUPOLE ELECTRODE GEOMETRY
MIKKO HAAPALAINEN

DIELECTROPHORETIC MOBILITY OF A SPHERICAL PARTICLE IN 2D HYPERBOLIC QUADRUPOLE ELECTRODE GEOMETRY

Academic dissertation to be presented with the assent of the Doctoral Training Committee of Technology and Natural Sciences of the University of Oulu for public defence in Kuusamonsali (Auditorium YB210), Linnanmaa, on 22 November 2013, at 12 noon

UNIVERSITY OF OULU, OULU 2013
Haapalainen, Mikko, Dielectrophoretic mobility of a spherical particle in 2D hyperbolic quadrupole electrode geometry.
University of Oulu Graduate School; University of Oulu, Faculty of Technology, Department of Electrical Engineering, Optoelectronics and Measurement Techniques Laboratory
University of Oulu, P.O. Box 8000, FI-90014 University of Oulu, Finland

Abstract

The dielectric properties of material are of importance in various industrial and scientific applications of measurement technology. These properties are usually measured by microwave sensors, methods that provide an average result of the entire sample volume. However, the need to know the dielectric properties of single particles has increased in various segments. Dielectrophoresis (DEP) as a method provides a way to determine dielectric properties, such as permittivity and conductivity, of single particles with good precision by using a sufficiently simple system.

In this thesis the DEP platform consists of four electrodes with hyperbolic edge geometry. Geometry produces a linear electric field gradient which leads to a constant DEP force in the active region and ensures fairly straightforward determination of particle properties by means of their DEP mobility. Particle mobility is dependent on particle size, the slope of the electric field gradient, the conductivity of the carrier fluid and the frequency of the electric field.

For this thesis has been studied particle characterization on a 2D platform, particle 3D behavior, simultaneous multiphenomena observation and the applicability of transparent indium-tin-oxide (ITO) electrode material to DEP platforms. Experiments were made with polystyrene particles, using carrier fluids of varying conductivity. Electrode transparency enabled holographic 3D imaging and thus also the observation of particle behavior in the depth direction. This 3D imaging revealed the restricted working distance from the electrode plane, as well as the mobility of particles in the depth dimension under the DEP force. It was also observed that there was only a marginal difference between 3D mobility and 2D mobility inside the active region. The dielectric properties of the polystyrene particles were determined and found to differ from those of solid polystyrene, and thus the properties of the carrier fluid have a distinctive effect on particle properties. The measurement achieved good precision in a single particle measurement and thus the total particle consistency could be very low.

The main restrictions of the DEP method are the limited working distance and restricted range of particle sizes: based on the results of this thesis, with a fixed size platform the particle size may not vary by more than tenfold.

Keywords: conductivity, dielectrophoresis, measurement technology, microparticles, permittivity
Materiaalin dielektriset ominaisuudet ovat merkittäviä monissa teollisissa sekä tieteellisissä mitтаusteknisissä sovelluksissa. Yleensä nämä ominaisuudet mitataan mikroaaltosensoreilla, joilla mitataan näytetilavuuden keskiarvo. Tarve tietää yksittäisen partikkelin dielektriset ominaisuudet on lisääntynyt useilla alueilla. Dielektroforeesi (DEP) on menetelmä, jolla voidaan toistettavasti mitata yksittäisen partikkelin permittivisyyys ja johtavuus suhteellisen yksinkertaisella mittausysteemillä.


DEP-menetelmän merkittävimpiä rajoitteita ovat rajallinen toimintaajatus sekä rajallinen partikkelikoon variaatio, mikä voi olla kriittinen pieni.

**Asiasanat:** dielektroforeesi, johtavuus, mikropartikkelit, mitatauksetekniikka, permittivisyyys
Acknowledgements

The experimental work of this thesis was carried out during the years 2009–2011 in CEMIS-Oulu unit of measurement technology of the University of Oulu. I would like to express my deepest gratitude to Professor Anssi Mäkynen for his enthusiastic supervision of the thesis, for offering me a great environment for research, and for his unselfish and tireless encouragement throughout the work. The meritorious and constructive review of the thesis by Dr. Artashes Karmenyan and Dr. Risto Oikari is gratefully acknowledged.

Thanks are owed to my co-authors Ville Kaikkonen and Ismo Kinnunen for their valuable contribution to the studies of this thesis. Great appreciation is due to Hannu Moilanen for his support in the construction of the platform this thesis is based on. I also wish to thank Sari Nieminen, Joni Hattuniemi and Matti Sarén who gave me the spark for research and with whom I had many enlightening and supportive conversations during the work as well as in our free time. I am grateful to my colleagues at the CEMIS-Oulu unit of measurement technology for creating a pleasant atmosphere for research. I'd also like to present my humblest thanks to Päivi Tikkakoski for providing me the possibility to finalize my thesis alongside my present professional responsibilities, and to Marjo Nygård for revising the English of the thesis.

Financial support from the Walter Ahlstrom foundation and the Kainuu Regional fund of Finnish cultural foundation is gratefully acknowledged.

I thank my mother, sister and brother-in-law for their understanding and support for all my decisions in life, and for not being too curious about the dissertation. I'm indebted to my parents-in-law Seija and Pertti Korhonen for their everyday support of our home and family.

Dear thanks go to my daughters Vilma and Verna for being the counterbalance for my thesis work and for being the lovely persons you are. In addition, great appreciation is owed to my beloved wife Elina, who has given me the opportunity and her support and encouragement throughout the time I've been working on this thesis.

Kajaani, September 2013

Mikko Haapalainen
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>2D</td>
<td>two-dimensional</td>
</tr>
<tr>
<td>3D</td>
<td>three-dimensional</td>
</tr>
<tr>
<td>AC</td>
<td>alternating current</td>
</tr>
<tr>
<td>DC</td>
<td>direct current</td>
</tr>
<tr>
<td>DEP</td>
<td>dielectrophoresis</td>
</tr>
<tr>
<td>DIHM</td>
<td>digital In-line holographic microscopy</td>
</tr>
<tr>
<td>$F_{st}$</td>
<td>friction force of particles, Stokes force</td>
</tr>
<tr>
<td>$F_{dep}$</td>
<td>dielectrophoretic force</td>
</tr>
<tr>
<td>ICP RIE</td>
<td>inductively-coupled-plasma reactive-ion-etching</td>
</tr>
<tr>
<td>ITO</td>
<td>indium-tin-oxide, transparent electrode material</td>
</tr>
<tr>
<td>KCl</td>
<td>potassium chloride</td>
</tr>
<tr>
<td>LCD</td>
<td>liquid crystal display</td>
</tr>
<tr>
<td>nDEP</td>
<td>negative - dielectrophoresis</td>
</tr>
<tr>
<td>pDEP</td>
<td>positive - dielectrophoresis</td>
</tr>
<tr>
<td>RIE</td>
<td>reactive ion etching</td>
</tr>
<tr>
<td>rms</td>
<td>root-mean-square</td>
</tr>
<tr>
<td>sccm</td>
<td>standard cubic centimeter per minute, unit of pressure</td>
</tr>
<tr>
<td>d</td>
<td>radius of the active region of the dielectrophoretic platform</td>
</tr>
<tr>
<td>Hz</td>
<td>Hertz, 1/s</td>
</tr>
<tr>
<td>Im</td>
<td>imaginary part of a complex equation</td>
</tr>
<tr>
<td>$K_s$</td>
<td>particle surface conductance</td>
</tr>
<tr>
<td>$K_{s,s}$</td>
<td>Stern layer</td>
</tr>
<tr>
<td>$K_{s,d}$</td>
<td>diffuse layer</td>
</tr>
<tr>
<td>$K(\omega)$</td>
<td>Clausius-Mossotti factor</td>
</tr>
<tr>
<td>M</td>
<td>mole, amount of substance</td>
</tr>
<tr>
<td>mM</td>
<td>milli-mole</td>
</tr>
<tr>
<td>M</td>
<td>mega, $10^6$</td>
</tr>
<tr>
<td>m</td>
<td>milli, $10^{-3}$</td>
</tr>
<tr>
<td>m</td>
<td>meter</td>
</tr>
<tr>
<td>n</td>
<td>nano, $10^{-9}$</td>
</tr>
<tr>
<td>R</td>
<td>particle distance</td>
</tr>
<tr>
<td>Re</td>
<td>real part of a complex equation</td>
</tr>
<tr>
<td>r</td>
<td>particle radius</td>
</tr>
<tr>
<td>$r^2$</td>
<td>correlation coefficient</td>
</tr>
<tr>
<td>Symbol</td>
<td>Description</td>
</tr>
<tr>
<td>--------</td>
<td>-------------</td>
</tr>
<tr>
<td>S</td>
<td>Siemens, unit of electric conductance</td>
</tr>
<tr>
<td>s</td>
<td>second, time unit</td>
</tr>
<tr>
<td>$U_{\text{rms}}$</td>
<td>voltage potential difference, rms-value</td>
</tr>
<tr>
<td>$\mu$</td>
<td>micro, $10^{-6}$</td>
</tr>
<tr>
<td>V</td>
<td>volt</td>
</tr>
<tr>
<td>$V_{p-p}$</td>
<td>voltage peak-to-peak</td>
</tr>
<tr>
<td>W</td>
<td>watt</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>dielectrophoretic mobility coefficient</td>
</tr>
<tr>
<td>$\beta$</td>
<td>dielectrophoretic efficiency factor</td>
</tr>
<tr>
<td>$\varepsilon_0$</td>
<td>vacuum permittivity, $8.854187 \times 10^{-12} \text{ F/m}$</td>
</tr>
<tr>
<td>$\varepsilon_m$</td>
<td>medium permittivity</td>
</tr>
<tr>
<td>$\varepsilon_p$</td>
<td>particle permittivity</td>
</tr>
<tr>
<td>$\eta$</td>
<td>viscosity</td>
</tr>
<tr>
<td>$\Theta$</td>
<td>particle diameter</td>
</tr>
<tr>
<td>$\omega$</td>
<td>angular frequency, radiance</td>
</tr>
<tr>
<td>$\pi$</td>
<td>pi, ~3.142</td>
</tr>
<tr>
<td>$\sigma_m$</td>
<td>medium conductivity</td>
</tr>
<tr>
<td>$\sigma_p$</td>
<td>particle conductivity</td>
</tr>
<tr>
<td>$\sigma_{p,\text{bulk}}$</td>
<td>particle bulk conductivity</td>
</tr>
<tr>
<td>$\sigma_{p,\text{surface}}$</td>
<td>particle surface conductivity</td>
</tr>
<tr>
<td>$\tau_{\text{MW}}$</td>
<td>Maxwell-Wagner relaxation time</td>
</tr>
</tbody>
</table>
List of original articles

This thesis is based on five original publications, referred to by Roman numerals:


The author’s contribution to the publications

Paper I is a study of the feasibility of ITO electrodes to the dielectrophoretic platform and their overall feasibility to particle manipulation by means of electrokinetic phenomenon; this paper presents experiments of particle characterization by DEP using a platform made with ITO electrodes. Paper II describes experiments with DEP observed with digital inline holographic microscope. These experiments verify that DEP has a limited working distance, and these aspects are briefly discussed in the paper. Paper III contains a study of particle behavior in a linear electric field gradient with a wider particle population on the same platform as Paper I and II. Paper IV is a more detailed study of the three dimensional behavior of particles under the influence of a dielectrophoretic force. Paper V is a broader study of the performance of ITO electrode based platform with regard to particle size and to the variation of carrier liquid conductivity. In this paper the relation of particle mobility factor to particle size is studied empirically.

The author has carried out all the experiments and data analysis and has also written the main part of papers I, III and V. The author was responsible for the DEP related sections in papers II and IV, and has participated in the experiments and data analysis and written the DEP related parts in both papers. The author has been a corresponding author for all the papers.
Contents

Abstract
Tiivistelmä
Acknowledgements 7
List of symbols and abbreviations 9
List of original articles 11
Contents 13
1 Introduction 15
  1.1 DEP applications ........................................................................ 16
  1.2 The potential of single particle characterization in industrial
      applications .............................................................................. 18
  1.3 The goal and structure of the thesis ............................................. 19
2 Dielectrophoresis and particle characterization through
dielectrophoretic mobility 21
  2.1 Dielectrophoretic force .................................................................. 22
  2.2 Other external and electrokinetic forces during DEP experiments .... 23
      2.2.1 Brownian motion and diffusion ........................................... 23
      2.2.2 AC electro-osmosis ............................................................. 24
      2.2.3 Buoyancy, gravitation and sedimentation ......................... 24
  2.3 The effect of carrier fluid .............................................................. 24
  2.4 Particle characterization through DEP mobility in a hyperbolic
      quadrupole geometry .................................................................. 25
      2.4.1 Particle conductivity and permittivity ................................ 27
      2.4.2 The effect of particle surface conductivity ......................... 28
3 DEP platforms 31
  3.1 Electrode geometries ..................................................................... 31
      3.1.1 Quadrupoles ........................................................................ 32
      3.1.2 Castellated electrodes ........................................................... 33
      3.1.3 Interdigitated and parallel electrodes ................................... 33
  3.2 Electrode materials .......................................................................... 34
  3.3 ITO electrode .............................................................................. 35
4 Experimental results 37
  4.1 DEP platform ................................................................................ 38
      4.1.1 Preparation of the ITO based DEP platform ....................... 38
      4.1.2 The simulated electric field of the platform ....................... 40
  4.2 Measurement system and experimental protocol ......................... 41
1 Introduction

The dielectric properties of materials play a key role in many scientific and industrial processes. It is therefore hardly surprising that numerous methods have been developed to measure the electrical properties of solid and liquid materials in various production and laboratory applications. One of the most common methods is the consistency measurement used in pulp and paper production: the transmission time of microwaves is measured to determine the dielectric constant of a liquid bulk material (suspension) containing wood fibers and fillers whose consistency affects the measured dielectric value (Metso Automation 2003, Nyfors et al. 1989). Industrial applications also include the measurement of various other compositional or physical changes of liquids based on their dielectric properties. Microwave measurement is the most commonly used technique in these applications (Canos et al. 2007, Friiso & Tjomsland 1997). These so-called bulk methods measure the average dielectric properties within a certain material volume but they are only hypothetically capable of measuring the dielectric properties of the individual particles suspended in the material, because this would assume knowledge of particle concentration and the properties of the medium surrounding the particles. Thus bulk methods cannot be used to measure the dielectric properties of individual particles. Such a measurement would, however, be useful and beneficial in many industrial processes, such as bioleaching where the efficiency of the process is dependent on the amount of living bacteria in a population, detectable by their dielectric properties (Nurmi 2009).

Dielectrophoresis (DEP) is a phenomenon that provides the possibility to characterize the important dielectric properties even from a single particle. This method has been used for trapping, manipulating, fractioning and characterizing particle populations based on their dielectric properties (Aldaeus et al. 2005, Morgan 2003, Pethig 2010, Kostner et al. 2010, Menachery et al. 2010, Imasato & Yamakawa 2010). The phenomenon was first observed and described by Pohl (1951) in the 1950s, but it took a while before it excited scientific recognition or industrial interest. DEP has been widely demonstrated, but so far its use is confined to the laboratory while the industrial segment is still looking for real applications. The functionality and usefulness of the DEP method have been demonstrated and reported in numerous laboratory experiments. Also the knowledge of DEP theory has reached such a point that simulations of DEP have become reliable (Salonen et al. 2005, Cummings 2003, Kua et al. 2008, Chang &
Today most applications employing dielectrophoresis are connected to medical research. The use of this method is justified when inspecting for example the effects of drugs or the life state of cells, both of which have an effect on various electrical properties of particles: surface charge (Cen et al. 2004, Khoshmanesh et al. 2011), membrane capacitance (Archer et al. 1999) or the conductivity of cytoplasm (Pethig et al. 2010). This makes the DEP method applicable for monitoring physiological changes in cells (Broche et al. 2007, Pethig et al. 2003). A review paper concludes that the DEP phenomenon is well mastered today and therefore it is time to step towards “real” applications (Pethig 2010). As opposed to laboratory analysis, the environment and matrixes of industrial online measurements are typically uncontrollable and more complicated, thus requiring a high level of robustness from the measurement. In this thesis the applicability of the DEP method for determining the permittivity and conductivity of individual particles is studied. In addition to factors such as the applicable size range and the fundamental mobility behavior of particles, the robustness of the method against carrier fluid conductivity variations and other electrokinetic effects is studied and discussed.

1.1 DEP applications

Even though many experiments with DEP have been made in order to study its possible uses for various purposes and applications, its utilization is not widespread. A review paper by Pethig (2010) discusses in detail its industrial and biomedical applications and primary uses, and lists the most relevant patents from 2005 to 2010 employing DEP.

Dielectrophoresis in its usual system scale is best suited for biomedical research where the target particles and the observation environment are on the same scale and fully controllable (Kua et al. 2005). The most common particle size for DEP is from one to ten micrometers, although some studies with submicrometer particles have also been done (Morgan et al. 1999); basically it is a question of the ratio between particle size and electrode geometry. DEP has been actively employed in the areas of medical and biomedical research. A wide range of bioparticles have been researched by using DEP characterization and manipulation: blood cells (Yang et al. 1999, Chang et al. 2003), stem cells (Pethig et al. 2010), human malignant cells (Yang et al. 2010, Cen et al. 2004, Huang et al. 2002), DNA (Hölzel 2009, Tuukkanen et al. 2005), bio-polymers (Washizu et al. 1994), and viruses (Archer et al. 1999), just to name a few. Cheng et al. (2007)
and Wakizaka \textit{et al.} (2004) have reported of a continuously flowing filter employing DEP, the advantage being that the filter can be cleaned by turning off the filtering electric field. A common feature in all these biomedical applications is that the DEP method does not require any biochemical labels; the target particles remain in their native state and thus the biosystem can be tested in a more authentic environment (Jesús-Perez & Lapizco-Encinas 2011).

In addition, DEP has been utilized to manipulate particles for various purposes. In one application the deposition of substrate with nanoparticles (Abe \textit{et al.} 2004) was controlled by DEP, and other researchers have characterized and fractionated (Kang \textit{et al.} 2006) particles from each other by applying the knowledge obtained from characterization experiments (Li & Kaler 2003, Choi \textit{et al.} 2008, Braschler \textit{et al.} 2007, Watarai \textit{et al.} 1997, Pamme 2007, Zhu \textit{et al.} 2010, Krajl \textit{et al.} 2006).

Since the early 20th century, phenomena similar to DEP have been pushed towards industrial uses: for example a US patent from 1924 reveals that minerals have been separated by using their dielectric capacitance (Pethig 2010). More recent studies have employed DEP to nano- and micro-scale deposition and, with a more sophisticated approach, to produce patterned deposition. DEP has also been employed in a micro-droplet dispenser (Schwartz \textit{et al.} 2004, Teh \textit{et al.} 2008, Ahn \textit{et al.} 2006) or mixer (Cooney \textit{et al.} 2006), in a micro-polisher with abrasive particles (Kim \textit{et al.} 2004) and for the determination of organic pollutants (Jesús-Perez & Lapizco-Encinas 2011).

While some industrial laboratory applications can be found, applications using DEP in industrial processes still face some partly unsolved challenges common to all microfluidic online applications. The most significant of these are the contamination of platforms and the high fluid volumes needed for reliable measurement. For example flow-through DEP traps or fractionators (Li \textit{et al.} 2005, Tsukahara \textit{et al.} 2001, Krajl \textit{et al.} 2006) could be utilized in industrial processes by using multiple parallel devices in the functional part of the system. However, a number of challenges can be expected along the road to a ready product, for example device contamination and its effect on overall performance. In addition, the uncontrolled environment with factors such as varying carrier fluid conductivity and the complicated matrixes of substances will influence the performance of DEP platforms.
1.2 The potential of single particle characterization in industrial applications

The dielectric properties of materials and microparticles play a significant role in industrial applications. The effect of bioparticles on an industrial process may vary depending on whether the particle is dead or alive, or whether or not it is influenced by some drug. Due to the fact that the dielectric properties of biological materials change during the state of life, bioparticle populations may be fractionated or manipulated by identifying their dielectrical properties, for example by separating live bioparticles from dead ones or, more generally, by fractionating them based on some other characteristics that affect their dielectric properties (Jesús-Perez & Lapizco-Encinas 2011). One potential example of such industrial application is the monitoring of bioleaching solutions (Nurmi 2009) to find out whether or not the bacteria population is prospering as it should. Another type of potential industrial applications of DEP could include the monitoring of decontamination processes, i.e. the removal of undesired particles from bioparticle populations, one example being the removal of fatal bacteria such as E. coli from drinking water. Thus DEP could be utilized in tasks that are highly relevant for the society. A significant aspect is that nearly all DEP applications can be implemented without using additional chemicals or dyeing solutions, a remarkable advantage in industrial processes where the consumption of such chemicals could otherwise be extremely high (Jesús-Perez & Lapizco-Encinas 2011). However, bioparticle populations could suffer from the strong dispersion which may on occasion make the application rather challenging (Markx & Davey 1999).

The future target could be to design optimized microfluidic DEP platforms for the characterization of neutral, in-process particles that are presumed to have a critical effect on the operation of an industrial or natural process. Another application would be in the characterization of some artificial, biochemically activated particles purposely added to a process in order to monitor process status. The general idea is that by measuring the polarizability spectrum of individual particles, determined according to the electrical properties of the particle and the surrounding medium, and by collecting the distributions of these individual spectra indicating the state of the process, could ultimately be applied on-line DEP sensors for real-time process control and monitoring.
1.3 The goal and structure of the thesis

Two-dimensional (2D) hyperbolic quadrupole electrode geometry is used to measure the permittivity and conductivity of single micro-scale particles in an aqueous conductive medium based on their dielectrophoretic mobility. The factors affecting the applicable particle size are established experimentally and also the sensitivity of the measurement results to variations in the carrier fluid conductivity is studied; both of these are critical factors with regard to online measurements of industrial particles. The main research questions of this thesis are:

- What is the quantitative difference between the acquired mobility values in the direction of electrode plane and in the direction of a particle’s true 3D motion in case of 2D (thin electrode) platforms?
- What is the mobility versus particle size dependence in practical experiments, i.e. does the first order theory describe the mobility of a particle accurately enough in case of 2D platforms so that it can be used to calculate the permittivity and conductivity of the particle?

Based on the answers to these questions, the suitability of the method and the 2D platform for determining the electrical properties of particles is discussed.

One of the main technical challenges in this study was to enable 3D imaging of the particle motion caused by dielectrophoresis and other electrokinetic or hydrodynamic effects within the whole platform. Such imaging requires a fully transparent platform – also including the electrodes – and an imaging method capable of determining the 3D position of particles. During this study a fully functional dielectrophoretic platform with transparent ITO electrodes was prepared, and a digital in-line holographic imaging method was used to determine the particle positions in 3D space above the transparent platform.

The thesis consists of six chapters. Following the introduction, chapter 2 deals with the theory of dielectrophoresis and how the measured particle mobility can be used to determine its electrical properties. Chapter 3 describes the common electrode geometries for DEP, and also discusses the relevant properties of ITO as an electrode material and its characteristics for the geometries used. Chapter 4 summarizes the measurement methods and presents the results and main findings of the experimental work. Chapters 5 and 6 discuss in detail the results and the present and future state of research, and it also contains the conclusions of this thesis.
2 Dielectrophoresis and particle characterization through dielectrophoretic mobility

A non-uniform electric field polarizes a particle and the conductive medium surrounding it. When the two differ from each other with regard to their polarizability, a dielectrophoretic force will appear. This polarizability is described through the dielectric properties of the material: permittivity and conductivity. To mobilize a particle unequal polarizability is required, and when the polarizabilities are unequal the force effective upon the particle can be either repulsive or attractive. Fig. 1 illustrates in principle the effect of uniform and non-uniform electric fields on a particle. In alternating current dielectrophoresis, an inflection frequency will appear where the polarizabilities of these materials are equal. Thus the forces on both sides of the particle are equal and do not mobilize the particle. If the polarizability of the particle is smaller than that of the surrounding medium, the particle undergoes a repulsion force of the high electric field: negative dielectrophoresis (nDEP). When particle polarizability is higher than that of the surrounding medium, it undergoes an attraction force of the high electric field: positive dielectrophoresis (pDEP). The dielectrophoretic force is directed in the direction of the electric field gradient. The magnitude of the force depends on particle size, the strength of the electric field gradient and the relation between the polarizabilities of the particle and the surrounding medium.

Fig. 1. Comparison between a particle in a uniform (left) and non-uniform (right) electric field. The green bars and triangle stand for the electrodes, the light blue arrows illustrate the forces affecting on the particle (yellow). The dielectric particle is in uniform (equal forces) and in non-uniform (unequal forces) electric fields, and the pDEP particle (right) is attracted by the denser electric field.
2.1 Dielectrophoretic force

The time averaged dielectrophoretic force for spherical particles can be expressed in the form

\[ F_{\text{dep}} = 2\pi r^3 \varepsilon_m \text{Re}[K(\omega)] \nabla \left| E_{\text{rms}} \right|^2, \quad (1) \]

where \( \varepsilon_m \) is the absolute permittivity of the carrier fluid, \( \nabla \left| E_{\text{rms}} \right|^2 \) is the gradient of the squared electric field, \( r \) the radius of the particle and \( \text{Re}[K(\omega)] \) the real part of the Clausius-Mossotti factor, where \( \omega \) is the angular frequency of the applied field (Morgan et al. 2003, Pethig 2010, Watarai et al. 1997, Kuokkanen 2009, Castellarnau 2006, Markx & Davey 1999). All factors in Equation 1 except \( \text{Re}[K(\omega)] \) are positive, hence the sign of the DEP force (direction of particle movement) is defined by the real part of the Clausius-Mossotti factor. The equation involves some simplifications: the particle is suspended in a homogenous medium that does not exhibit conductive losses or carry a net charge; there are no boundary effects; and the surrounding medium is assumed to be infinite. Usually the polarized particle is a dipole, but in cases where the effective wavelength (i.e. the non-uniformities of the electric field) is smaller than particle size, also multipoles may form: quadrupole, octopole, etc. Equation 1 reveals the strong effect of the particle radius and the magnitude of the squared field gradient on the DEP force. Therefore, the smaller the particle, the weaker its movement by DEP, unless extremely high electric field gradients are available. The latter is best achieved by minimizing the distance of electrodes. In practice the low DEP force of small particles makes it difficult to determine their mobility value due to the dominating Brownian random motion whose magnitude increases as the size of the particle decreases, while at the same time the DEP force decreases.

The direction of the DEP force experienced by particles depends on the relative polarizability of the particle and the surrounding medium, which again is determined by their electrical properties. This is described by the complex Clausius-Mossotti factor:

\[ K(\omega) = \frac{\varepsilon_p - \varepsilon_m}{\varepsilon_p + 2\varepsilon_m}, \quad (2) \]

where \( \varepsilon_p \) and \( \varepsilon_m \) define the complex permittivity (\( \varepsilon = \varepsilon' - j\varepsilon''/\omega \)) of the particle and the carrier fluid, respectively (Morgan & Green 2003). The DEP cross-over
frequency, the inflection point where nDEP turns into pDEP and vice versa, is referred to as the Maxwell-Wagner relaxation frequency and it is defined by

$$f_{MW} = \frac{1}{2\pi \tau_{MW}} = \frac{1}{2\pi} \frac{\sigma_p + 2\sigma_m}{\varepsilon_p + 2\varepsilon_m},$$

where $\tau_{MW}$ is the relaxation time, $\sigma_p$ is the conductivity of the particle and $\sigma_m$ is the conductivity of the carrier fluid (Morgan & Green 2003, Pethig 2010, Watarai et al. 1997). The Maxwell-Wagner relaxation frequency is the exact frequency point where Re[$K(\omega)$] is zero. Equations 2 and 3 define the spectral behavior of the DEP force and its dependence on the electrical properties of the measured particle and the carrier fluid. In a standardized environment each particle has its own characteristic DEP force spectrum, which can offer multiple possibilities for developing the sensing principles.

### 2.2 Other external and electrokinetic forces during DEP experiments

In addition to DEP, there are also other phenomena at play that affect the behavior of particles in a suspension: for example AC electro-osmosis, particle-particle interaction, the Brownian force and gravitation (Pethig & Markx 1997). These forces, including DEP, can be categorized into two types: deterministic forces and random, non-deterministic forces. Generally the Brownian force represents non-deterministic forces, while the other mentioned forces are deterministic.

#### 2.2.1 Brownian motion and diffusion

The Brownian force is the result of thermal vibration or movement in a suspension, as the molecules in a suspension are continuously bumping into suspended particles and into each other (Morgan & Green 2003). Once this force overcomes the gravitation and inertia of a particle, it gives the particle a random mobility. Even though the Brownian force is random, its amplitude could be restricted by controlling the temperature and viscosity of the carrier suspension.

Diffusion begins when the system consists of a large number of particles. It is generally described as the transport of particles or molecules from high to low concentration, to the state where particles are randomly and uniformly distributed, where the driving force is the Brownian force of single particles. The external
force drives the system towards a non-uniformly distributed population while the diffusion driven by Brownian motion leads the particle population and system to a state where they are uniformly distributed.

2.2.2 AC electro-osmosis

AC electro-osmosis is another one of the most significant causes of fluid flows in experiments with dielectrophoresis. Electro-osmotic flow is an essential technique in chemical separation methods, such as capillary electrophoresis. AC electro-osmosis does not work within the active region but it has a significant effect outside this area, for example on top of the electrodes. Electro-osmosis is the fluid flow caused by the Coulomb force induced by an applied potential, parallel to a charged surface (Hughes 2003). Earlier observation of electro-osmosis in DEP experiments has been challenging, but the ITO as a transparent material was found to effectively decrease the effort needed.

2.2.3 Buoyancy, gravitation and sedimentation

Buoyancy and gravitation are forces leading to sedimentation when a particle is suspended in a fluid: the particle displaces a fluid volume that is equal to its volume. Gravity is a downward force if the particle is denser than the carrier fluid, whereas buoyancy is an opposite force that correlates with the volume of the particle. Thus the direction – downward or upward – and the sign of sedimentation are determined by the densities of the materials in question.

2.3 The effect of carrier fluid

As the results section of this thesis indicates, the properties of the carrier fluid have a major effect on the properties determined from microscale particles. According to the electrical properties of the carrier fluid, it affects the $K(\omega)$ and thereby also $F_{\text{DEP}}$. In the experiments the viscosity of the carrier fluid affects particle mobility, and it also has an effect on the minimum particle size as it restricts the amplitude of the Brownian motion. Carrier fluid conductivity has an especially notable effect on the conductivity of the studied particles, and the attained high particle conductivities can be explained by the inclusion of their surface conductivity components (Morgan & Green 2003).
2.4 Particle characterization through DEP mobility in a hyperbolic quadrupole geometry

A straightforward way to determine the electrical properties of particles by means of their dielectrophoretic mobility is to create a linear electric field gradient and use a carrier fluid with known properties. This chapter describes the method used to determine particle properties by measuring their mobility: particle location is detected as a function of time from microscopic images. The hyperbolic quadrupole electrode geometry used in this thesis produces the desired non-uniform electric field with a linear gradient. Due to the linearly changing electric field, the DEP force affecting a particle remains constant as the particle moves within the active region of the platform (between the electrode tips). The net effect of a constant DEP force and the opposing frictional force of viscous fluid results in predetermined mobility behavior of particles. Based on this behavior, the permittivity and conductivity of a particle can be calculated from its mobility in the set magnitude of the electric field gradient, the known viscosity, and the permittivity and conductivity of the carrier fluid.

The definition of particle mobility begins with the Newton equation of motion. In case the DEP is positive, Newton’s equation is

\[ m \frac{d^2 R}{dt^2} = F_{\text{DEP}} - F_{\text{ST}}, \]

where \( F_{\text{DEP}} \) represents the dielectrophoretic force that mobilizes the particles, in this case the counter force is the friction force of a particle in viscous fluid \( F_{\text{ST}} \), \( R \) is the distance of the particle from the center point of the active area of electrodes, and \( t \) is time (Morgan & Green 2003, Pethig 2010, Watarai et al. 1997, Kuokkanen 2009). \( F_{\text{DEP}} \) and \( F_{\text{ST}} \) are counter forces to each other, and proportional to the distance \( R \) and particle speed \( \frac{dR}{dt} \), respectively. Thereby Equation 4 could be written using proportionality coefficients \( a \) and \( b \) (Pethig 2010, Watarai et al. 1997)

\[ m \frac{d^2 R}{dt^2} = aR - b \frac{dR}{dt}. \]

In case the target particle moves at a constant speed, the acceleration term becomes zero. The Equation 5 could then be written in the form

\[ aR - b \frac{dR}{dt} = 0. \]
From the differential Equation 6 further is got
\[ R = Re^{\frac{\omega}{b}} \Rightarrow \ln R = \frac{a}{b} t + \ln R_0, \]  
(7)

where \( R_0 \) represents the initial distance of the particle from the center of the electrode tips (Pethig 2010, Watarai et al. 1997).

The coefficient of proportionality \( a \) can be deduced as follows: The squared electric field gradient in the second order polynomial shape electrode geometry used is
\[ \nabla |E_{rms}| = 8 \left( V_2 - V_1 \right)^2 \sqrt{x^2 + y^2} \]  
(8)

where the particle location in the active region within the electrode plane is defined by coordinates \( x \) and \( y \), and \( d \) defines the radius of the active region (Morgan & Green 2003, Watarai et al. 1997, Hughes 2003). The potential difference of the opposite electrodes \( V_1 \) and \( V_2 \) is equal to the \( rms \) voltage applied between the electrodes:
\[ (V_2 - V_1)^2 = U_{rms}^2. \]  
(9)

When Equation 9 and particle distance \( R \) are added to Equation 8, the squared electric field gradient is written as follows:
\[ \nabla |E_{rms}| = \frac{2RU_{rms}^2}{d^4}. \]  
(10)

When this description is applied into Equation 1 and \( F_{DEP} = aR \) is obtained from Equations 4 and 5, the proportionality coefficient \( a \) becomes
\[ a = 4\pi r^2 e_\infty \text{Re}[K(\omega)] U_{rms}^2, \]  
(11)

Proportionality coefficient \( b \) can be derived from the Stokes law of friction force; for a spherical particle in an aqueous medium it is described as
\[ F_{ST} = 6\pi \eta r \frac{dR}{dt}, \]  
(12)

From Equations 4 and 5 is known that \( F_{ST} \) equals \( b \cdot dR/dt \), and thus the \( b \) coefficient of proportionality is
\[ b = 6\pi \eta r, \]  
(13)
where $\eta$ represents the viscosity of the surrounding suspension and $r$ the radius of the particle, respectively (Morgan & Green 2003). In the next phase, substituting both proportionality coefficients $a$ and $b$ into Equation 7 yields

$$
\ln R = \frac{2r^2 \mathcal{E}_\omega U^2_{\omega}}{3\eta d^4} \text{Re}[K(\omega)] t + \ln R_0.
$$

(14)

According to Equation 14 $\ln R$ is linearly proportional to time, as all factors except $R$ and $t$ remain constant during particle mobilization. The slope factor of $\ln R$ represents the dielectrophoretic mobility coefficient $\alpha$

$$
\alpha = \frac{2r^2 \mathcal{E}_\omega U^2_{\omega \text{re}}}{3\eta d^4} \beta \text{Re}[K(\omega)]
$$

(15)

where $\beta$ describes the efficiency factor of the DEP force (Watarai et al. 1997, Tsukahara et al. 2000). Efficiency is a factor of electrode thickness and particle size. As an example, the factor is 1 when electrode thickness is larger than particle diameter, according to the behavior of electric field lines (Watarai et al. 1997, Tsukahara et al. 2000). In 2D DEP platforms electrode thickness is usually smaller than particle diameter, and thus the efficiency factor is less than one. In Equation 1 $\text{Re}[K(\omega)]$ defines the direction of mobility. We can see that the dielectrophoretic mobility of a spherical particle is dependent on the squared radius of the particle and the real part of the Clausius-Mossotti factor (Morgan & Green 2003). Since all other factors except for $\text{Re}[K(\omega)]$ are known, the equation could be written in the form

$$
\text{Re}[K(\omega)] = \frac{\alpha \eta d^3}{\beta 2r^2 \mathcal{E}_\omega U^2_{\omega \text{re}}}
$$

(16)

2.4.1 Particle conductivity and permittivity

The determination of particle permittivity is based on the Equation 15 of dielectrophoretic mobility coefficient $\alpha$. As already mentioned, all the other factors except the real part of the Clausius-Mossotti factor are known, and once the properties of the carrier liquid are known, particle properties can be derived from the equation of this factor. When the applied frequency is far below the inflection point, $\text{Re}[K(\omega)]$ could be described by the conductivities of the fluid and particle (Watarai et al. 1997, Morgan & Green 2003):
When the applied frequency is higher than inflection frequency, \( \text{Re}(K(\omega)) \) can be described by the permittivities of the materials used (Watarai et al. 1997, Morgan & Green 2003, Hughes 2003).

\[
\text{Re}[K(\omega)] = \frac{\epsilon_p - \epsilon_m}{\epsilon_p + 2\epsilon_m}
\]  

(18)

The real part of the Clausius-Mossotti factor could be described for the entire frequency range.

\[
\text{Re}[K(\omega)] = \frac{(\epsilon_p - \epsilon_m)\omega^2}{(\epsilon_p + 2\epsilon_m)(1 + \omega^2\tau^2)} + \frac{\sigma_p - \sigma_m}{(\sigma_p + 2\sigma_m)(1 + \omega^2\tau^2)}
\]  

(19)

However, using the whole range description basically complicates the numerical treatment. Reasonable accuracy can be achieved even by approximating with truncated Equations 17 and 18, in case the electric field frequency is clearly below (pDEP) or above (nDEP) the inflection frequency.

### 2.4.2 The effect of particle surface conductivity

Particle conductivity is a combination of bulk conductivity \((\sigma_{p,\text{bulk}})\) and surface conductivity \((\sigma_{p,\text{surface}})\). The total conductivity of the particle is described as follows:

\[
\sigma_p = \sigma_{p,\text{bulk}} + \sigma_{p,\text{surface}} = \sigma_{p,\text{bulk}} + \frac{2K_S}{r},
\]  

(20)

where \(K_S\) is the surface conductance of the particle, and \(r\) is its radius (Hughes et al. 1999, Morgan & Green 2003, Pethig 2010, Hughes 2003). Surface conductivity is strongly dependent on the properties of the electrical double layer that forms on the particle surface when it is immersed in the carrier fluid. The thickness of the double layer is inversely proportional to carrier fluid conductivity; thus it has a direct effect on the obtained electrical property values of the particle (Watarai et al. 1997). The electrical double layer consists of the Stern layer and the diffuse layer. The total surface conductance is described as follows.
\[ K_s = K_{s,s} + K_{s,d} \]  

where \( K_{s,s} \) is the Stern layer and \( K_{s,d} \) is the diffuse layer (Hughes et al. 1999). The Stern layer is outside the particle surface and it is assumed to be stagnant, meaning that the carrier fluid is immobile in this layer. The diffuse layer is located between the bulk fluid and Stern layer and is assumed to be more mobile. Both of these layers affect the true and thereby measured surface conductance of the particle. Note that for very small particles with low intrinsic bulk conductivity, the effect of surface conductance and therefore the carrier fluid conductivity can be very high.

The permittivity values of particles are also dependent on the conductivity of the carrier fluid. The electrical double layer affects polarizability in a contrary manner compared to natural permittivity. The applied electric field also tends to mobilize the ions of the double layer, and therefore it weakens the overall effect of the DEP force. This matter is one of the factors that lead to overestimation of the determined particles properties.

Hughes (2003) has compared the effect of the Stern layer with different electrolytes. This study has revealed that the Stern layer effect not only correlates with the conductivity but is also dependent of the electrolyte used in a solution. In addition, particle conductivity might be highly dispersive under DEP experiments (Nishimura et al. 2007). The conductivity of the carrier liquid affects the overall DEP force: increased conductivity increases the DEP force, whereas it has no significant effect on the determined particle properties.
3 DEP platforms

3.1 Electrode geometries

This chapter deals with the most common types of DEP electrode geometries and their potential in different applications. Common electrode designs with regard to their intended uses are discussed. The electrode platform could be designed specifically to meet the needs of the intended DEP application.

The objective of this chapter is to reveal the relationship between electrode geometry and field gradient, thus allowing also the behavior of particles to be understood. The topic, however, is restricted only to the most common types of electrode geometries: quadrupole electrodes, castellated electrodes and interdigitated electrodes. The article by Khoshmanesh et al. (2011) introduces in detail the different dielectrophoretic platforms, their basic characteristics and main areas of application. The study of Khoshmanesh et al. (2011) and the dissertation of Limin (2007) also discuss the advantages of three-dimensional platforms for the manipulation of cells using DEP. The 3D electrode design is based on a planar 2D design, but the electrodes are thicker. Fig. 2 illustrates such a structure of circular electrodes which are extruded to produce poles.

![Fig. 2. One example of 3D DEP structure with oppositely phased pole structure. The applied signal is fed for example to the dark green poles, ground to the light green poles. This could be used for example as a flow-through filter controlled with DEP force.](image)

A three-dimensional DEP platform has a deep flow channel and therefore a high carrier fluid throughput (Ma et al. 2011, Iliescu et al. 2008). While two-dimensional and planar DEP platforms suffer from their limited working depth,
the three-dimensional platform works with the same performance through the entire channel depth. The thesis of Limin (2007) introduces a broad variation of electrode shapes for three-dimensional DEP applications. The fluid channels can also be parallelized in order to increase the obtained fluid flow. The advantages of three-dimensional electrodes over two-dimensional ones include higher DEP force efficiency and the lack of dead zones, i.e. zones where DEP has no effect on the particles.

### 3.1.1 Quadrupoles

Quadrupole geometry, illustrated in Fig. 3, consists of four electrodes positioned symmetrically on the opposite sides. The phase of the applied signal is fed to one opposite pair (dark green) of electrodes, the ground to the other pair (light green). Electrode shape usually varies from circular to hyperbolic, according to the intended use. The electric field gradient changes depending on the shape of the electrode edge: a hyperbolic electrode produces a linear field gradient (Watarai et al. 2000, Masahiko et al. 2004), which is straightforward to use for particle characterization based on their DEP related mobility, because the DEP force is constant along the active region of the platform.

The quadrupole electrode geometry has several advantages: it is easily manufactured, it does not require very strong AC feed signals and is applicable to single particle experiments.

Fig. 3. Principle of quadrupole geometry (left), where positive electrodes are colored dark green, negative electrodes light green. This geometry produces a circularly symmetrical electric field gradient to the circle placed in between the electrode edges. The picture on the right illustrates an example of particle experiment in a steady state situation (right), where the red particles undergo a negative DEP, the blue particles a positive DEP. The red particles are moved by nDEP from the electrode edge towards
the center, the blue particles are moved by pDEP from the center towards the high field strength area at the electrode edges.

### 3.1.2 Castellated electrodes

A castellated electrode is shown in Fig. 4, with a set of square features on parallel lines. This geometry is capable to simultaneously perform nDEP and pDEP, and it is commonly used in flow-through systems such as DEP fractionators. Particles affected by pDEP flow between the electrode tips, while those experiencing nDEP are trapped between the square electrode shapes. Two variations of this particular electrode geometry are commonly used: the offset and non-offset versions (Khoshmanesh K. et al. 2011), based on whichever is better suited for the intended application.

![Fig. 4. Castellated electrodes (left), offset and non-offset versions. Functional illustration of a particle experiment (right), where the red particles undergo nDEP and the blue particles pDEP.](image)

The castellated electrode geometry is easy to manufacture, it is applicable to flow-through systems with a high flow rate, and the DEP force is distributed to a large part of the microchannel, thus giving a larger effective volume of DEP (Khoshmanesh et al. 2011). On the other hand, it suffers from the usual drawback of 2D DEP structures: a limited working depth, which obviously means that the channel on top of the electrodes must be shallow in order to achieve reasonable performance.

### 3.1.3 Interdigitated and parallel electrodes

The simplest and most common dielectrophoretic platform is the interdigitated parallel electrode geometry, where the electrodes are fed with AC voltage that is phased by 180° in consequential electrodes (Khoshmanesh et al. 2011). Fig. 5
shows the principle of such a system. The platform is applicable to basic DEP fractionation (Park & Beskok 2008). Particles experiencing pDEP are trapped to the electrode edges while those experiencing nDEP are repulsed from the electrode plane.

The parallel electrode geometry is largely similar to the castellated geometry in that it is simple to make, applicable to flow-through systems with a high flow rate, and the DEP force is distributed to a large part of the microchannel (Lee & Voldman 2007, Khoshmanesh et al. 2011).

### 3.2 Electrode materials

The electrode material is in direct contact with the conductive carrier fluid and therefore exposed to the carrier fluid electrolyte with a high voltage potential on the electrode edge; in some cases corrosion has been observed to affect the electrode edges during experiments. This corrosion issue is one of the reasons why such a wide range of different materials have been tested and utilized in the field of dielectrophoresis.

Typically DEP platforms are manufactured using borosilicate, quartz glass as a substrate for metal electrodes or by patterning silicon wafers (Urdaneta & Smela 2008, Liu et al. 2008). Common to all metal electrode DEP platforms is that an adhesion layer is required, for example chromium (Tsukahara et al. 2000, Abe et al. 2004). Depending on the solution on top of the adhesion layer there is a layer of gold (Liu et al. 2008, Lin & Yeow 2007, Lee & Voldman 2007, Watarai et al. 2011).
1997), ruthenium (Tsukahara et al. 2000), tungsten (Kuokkanen 2009), titanium (Zhang et al. 2005) or even carbon nanotubes (Tuukkanen et al. 2006). The platforms utilized with patterned silicon wafer (Bhatt et al. 2005) are usually multiplexed platforms where the electrodes could be individually controlled. Jen et al. (2008) and Demierre et al. (2006) have utilized electrode-less dielectrophoresis, where the obtained electric field is produced by placing insulating obstacles or dielectric constrictions.

Most of the metal electrodes are easily corroded due to high amplitudes and carrier fluid conductivities. In addition, if the carrier liquid is acidic the metal electrodes need a buffering layer. One solution for acidic environments is the electrode-less system where the insulating obstacles could be made of acid resistant material. A common feature of all metal electrodes is that they are easy to make and pattern, as the necessary processes are common and well known. The resolution requirement in DEP platforms, however, might lead to challenges.

### 3.3 ITO electrode

Indium-tin-oxide (ITO) is a transparent, electrically conductive material. ITO is mainly used when transparent conductive layers are needed on top of LCD panels, touch screens being just one example. The conductivity of the ITO layer is controlled by its thickness: a thicker layer has a higher conductance, and vice versa. However, when the layer gets thicker its transparency decreases, and thus the use of ITO electrodes always involves a compromise between transparency and conductivity. In DEP experiments the transparency enables basic microscoping but also more sophisticated imaging methods, for example holographic microscopy, whereby information also in the depth direction may be obtained. Thus ITO electrodes make it possible to observe not only DEP but also various other electrokinetic or hydrodynamic phenomena taking place during the experiments, since particle mobility also above the electrodes can be seen. In the experiments the ITO electrode proved to be adequately resistive chemically, and unlike most metal electrodes it could also withstand quite high voltages. The desired electrode geometry was reactive-ion-etched to the ITO layer which was sputtered on a glass substrate.

ITO as an electrode material has not been used a lot in DEP applications. This may be because of the challenging etching process, or because the availability of the material and knowledge about it have not been sufficient; or perhaps the need for a feature such as transparency of electrodes has not been recognized before.
In most articles discussing both ITO and dielectrophoresis the ITO electrode has been used as a blank ground plane (Lin et al. 2009) for interdigitated or light induced dielectrophoretic platforms (Yang et al. 2010, Williams et al. 2008, Bhatt et al. 2005, Fan et al. 2009). Patterned ITO electrodes have been seen in a few of the previous experiments of dielectrophoresis, for example interdigitated ITO electrodes used by Kumar et al. (2008).
4 Experimental results

To estimate the applicability of the DEP method for determining the mobility and thereby properties of single particle, a series of measurements was carried out using a hyperbolic quadrupole electrode geometry with transparent electrodes. The transparency of the electrodes also enabled the observations related to other electrokinetic phenomena, such as AC electro-osmosis or gravitation; these phenomena partially affect the particle mobility, as their effects are typically present on top of the electrodes where DEP has no intended effect.

The very first experiments were made using polystyrene particles with a diameter of 2 μm; these experiments were documented in paper I. These experiments proved that the platform was functional and thus enabled DEP imaging by means of a digital inline holographic microscope. The subsequent DIHM experiments showed that, as expected, the effective depth range of the DEP force in 2D electrode geometry is fairly limited, and therefore very challenging to observe with a traditional video microscope due to the limited depth of its focus. By contrast, in experiments was possible to image the entire sample volume and detect particle motion using just a few holograms. These experiments and conclusions are documented in paper II.

The next phase was to evaluate the performance of the platform by using a particle population with a larger size range. At the lower end the limit for particle size is set by comparing the Brownian force to dielectrophoretic force in order to determine which one is the prevailing force. The upper limit, on the other hand, is set by the geometry: what is the largest particle that fits in the measurement area so that there is still enough room to measure its mobility. These experiments and the conclusions from them are presented in paper III.

Paper IV is a more detailed study delving into the topics dealt with in paper II. In the earlier phase the analysis was restricted to a few pre-chosen depths, whereas now the data analysis was done for both nDEP and pDEP, single particle behavior was represented with 3D images and particle speeds were determined both with 2D projection and in the direction of particle motion. These experiments verified the strengths of the ITO-based DEP platform, as also other phenomena could be observed in connection with the DEP experiments.

The experiments of paper III were elaborated on in paper V: particle population was still wider, carrier fluid conductivity was varied and particle concentration decreased. This enabled us to determine the particle size limits for
the platform used, and to evaluate how carrier fluid conductivity affects the
determined particle properties.

4.1 DEP platform

The advantage of the hyperbolic quadrupole electrode geometry is that it
produces a linear electric field gradient (Abe et al. 2004, Watarai et al. 1997).
This creates a constant force to the measured particle as it moves and thus
provides a simple way to determine its mobility coefficient (Watarai et al. 1997),
needed for calculations of the electrical properties.

Fig. 6 shows a photograph of the used DEP platform; this photo also shows
the actual transparency of the electrode material.

![Fig. 6. Photograph of the used platform, visualizing the transparency of the ITO
electrodes. The cross section in the middle of the electrodes represents the active
measurement area of the platform (V, published by permission of © 2012 IEEE).](image)

Fig. 7 is a schematic drawing of the hyperbolic quadrupole geometry of the
platform. A circle with the radius \( d \) is drawn between the electrode edges (yellow)
to show the effective area where the electric field gradient is linear.

4.1.1 Preparation of the ITO based DEP platform

The processing of ITO layers was done in the Center of Microscopy and
Nanotechnology, a separate department of the University of Oulu. The reason for
this decision was that the author was not familiar with the process of ITO etching,
and the required resolution could not be easily achieved by wet etching methods.
Reactive-ion-etching was chosen as the processing method of the electrode geometries, but some experiments with the RIE method had to be done before platforms acceptable for measurement use could be produced. After some unsuccessful efforts the correct parameters for creating a suitable layer thickness were found. Our substrate was a borosilicate glass covered with a 300nm ITO layer, and in the process were used AZ1512 positive-photoresist and the ICP RIE procedure with duration of 30 minutes. Argon and chlorine were used as etching gases with flows of 30sccm and 20sccm, respectively. Both ICP power and Forward power were set to 100W. During the 30-minute etching session the photoresist was fully worn out and the remaining ITO layer of the electrode geometry was at least 160nm thick.

![Diagram of hyperbolic quadrupole electrode geometry](image)

**Fig. 7.** Principle of the hyperbolic quadrupole electrode geometry; circle indicates the active region of the platform with radius $d$ (V, published by permission of © 2012 IEEE).

The electrical connections to the ITO electrodes were made with colloidal silver. Because high frequencies were used, the signal line at the platform end of the transmission line was terminated to 50ohms. As both traditional microscoping and a digital inline holographic microscope were used, the droplet shape of the carrier fluid had to be flattened. To standardize the measurement volume and to prevent optical errors caused by droplet shape, a 150μm thick glass rod was attached on both sides of the electrode geometry to support the cover glass. Once the sample droplet was on top of the active region, the cover glass was laid on the support rods.
4.1.2 The simulated electric field of the platform

The electric field produced by the electrode geometry was simulated with CST Microwave Studio. Field frequency was 15MHz, amplitude 10V_{p-p}. The same carrier fluid conductivities (1.47, 7.63 and 13.86mS/m) as in the experiments made for paper V were also used in the simulations. To simplify the model and to save processing time, the electrodes were modeled with zero thickness, and the substrate and electrolyte thicknesses were reduced to 50μm and 30μm, respectively. In principle, the simplification reduces the physical size of the model and the number of effective calculation voxels of the simulated volume, and thereby it directly shortens the calculation time.

Fig. 8. Simulation results with various medium conductivities; from this can be concluded that there is no significant difference between the produced electric field gradients, and also the field strengths remain on a feasible level even when conductivity is varied (V, published by permission of © 2012 IEEE).

Fig. 8 illustrates the electric field simulation results in planar form, with three different electrolyte conductivities. From these simulations could be deduced that
electrolyte conductivity only had a negligible effect on the average field strength or gradient behavior in the same plane with the electrodes. In principle the field strength should remain constant when the conductivity of electrolyte varies, since the potential difference between the electrodes is kept constant. However, conductivity affects the strength of the electric field in the depth direction. The behavior of the electric field gradient was verified with these simulations, in order to execute comparable measurements in different electrolyte concentrations. In addition, these simulations revealed that the hyperbolic quadrupole electrode geometry produces a rotationally symmetric electric field where the field strength is almost zero at the center of the active region.

### 4.2 Measurement system and experimental protocol

The measurement system is illustrated in Fig. 9. It consists of an inverted compound microscope (Zeiss Axiovert 40CFL) equipped with a high-resolution digital camera, a function generator (Hewlett-Packard 33120A) for generating the sinusoidal signals for the DEP platform, and the functional DEP platform for creating the desired electric field for particle manipulation. 2D particle tracking experiments were carried out using this system. For 3D experiments the microscope was equipped with a commercial DIHM device from Resolution Optics Inc. and specialized data acquisition software with hologram reconstruction calculations. The DIHM set-up used is a lens-less system with a 405nm diode laser and a round pinhole of 500nm in diameter.

![Fig. 9. Schematic figure of the experimental set-up which consists of the following components: Function generator for feeding the desired sinusoidal signal to DEP platform, PC and digital camera with microscope for image capturing, and the functional DEP Platform with a cover glass and glass support rods.](image)
The experimental protocol is common for all of the 2D experiments. Once the platform was aligned to the microscope and focused to the electrode plane, the dilution of target particles was dropped onto the DEP platform. Image capturing was started simultaneously with switching on the sinusoidal signal. A detailed list of the experimental parameters is collected into Table 1.

Table 1. Parameters used in the experiments, divided into columns corresponding to the parameters used in studies.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Experiment type</td>
<td>2D</td>
<td>3D</td>
<td>2D</td>
<td>3D</td>
<td>2D</td>
</tr>
<tr>
<td>nDEP/pDEP</td>
<td>nDEP/pDEP</td>
<td>nDEP</td>
<td>nDEP</td>
<td>nDEP/pDEP</td>
<td>nDEP</td>
</tr>
<tr>
<td>Particle Ø [µm]</td>
<td>2</td>
<td>4.08</td>
<td>2.07, 4.08, 5.98</td>
<td>9.65</td>
<td>5.98, 0.98, 2.07, 4.08, 9.65</td>
</tr>
<tr>
<td>KCl dilution [mM]</td>
<td>0.1</td>
<td>5</td>
<td>0.1</td>
<td>0.1</td>
<td>0.1, 0.5, 1.0</td>
</tr>
<tr>
<td>Conductivity [mS/m]</td>
<td>1.42</td>
<td>69.5</td>
<td>1.44</td>
<td>1.44</td>
<td>1.47, 7.63, 13.9</td>
</tr>
<tr>
<td>Voltage [Vp-p]</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td>5</td>
<td>10</td>
</tr>
<tr>
<td>Frequency</td>
<td>15MHz / 100kHz</td>
<td>15MHz</td>
<td>15MHz</td>
<td>15MHz / 10kHz</td>
<td>15MHz</td>
</tr>
</tbody>
</table>

Once the images were captured, particle coordinates were determined with a Matlab script where the centre point of the active region was set as the point of origin. Using these coordinates the distance of particles from the active region centre could be analyzed as a function of time. From this distance data was determined the natural logarithm and calculated the slope of the linear fitting which corresponds to the DEP mobility coefficient \( \alpha \).

Experimental protocols in the 3D experiments are similar to the 2D experiments, with the exception of the image capturing equipment and image analysis. The dilution containing the particles is dropped to the platform, the cover glass is set on top of the support rods and the DIHM device is aligned to image the active region of the platform. Once the DIHM device was ready, image capturing was started and the sinusoidal voltage was turned on; real time observation from these experiments was not feasible. Once the authors decided that a reasonable amount of tracking data had been acquired, holograms (Fig 16) were reconstructed with the software from Resolution Optics Inc. Using the reconstructed holograms at different depths of the sample volume, particle locations were determined and the obtained location coordinates were used to plot the particle routes in 3D perspective; Fig. 17 and 18.
4.3 Measurement results

4.3.1 Platform characterization experiments

The basic characterization of the functionality of ITO based DEP platform is described in Paper I. The dielectric properties of particles were determined according to their DEP mobility using both nDEP and pDEP. The measured DEP mobility results were then used to determine the dielectric properties of the diameter 2µm polystyrene particles used in the experiments. The conductivity and permittivity of the particles were found to be 3.3mS/m and 53.7ε₀, respectively. The results show that the ITO electrode is suited for DEP experiments, and a comparison of the obtained particle properties with the results of Watarai et al. (1997) proves that the same results can be obtained with both metal and ITO electrodes.

Fig. 10. Experiment of pDEP (left) and nDEP (right) with ITO electrodes. The upper figures illustrate particle distance as a function of time from the center of the active region. The lower figures show the natural logarithm of the particle position, in order to determine the mobility coefficient from the fitted linear curve (I, published by permission of © 2010 SPIE).

During the experiments the inflection frequency was determined empirically, by observing particle behavior when the applied frequency was changed to see whether the particles moved towards the electrodes or away from them; inflection frequency was found to be in the range 700–800 kHz. This observation was confirmed also by calculating the DEP spectrum from the measured values of the particles (3.3mS/m, 53.7ε₀) and the known properties of the carrier. The calculated spectrum in Fig. 11 indicates that the frequency point (~850kHz) where the real part of the Clausius-Mossotti factor turns from positive to negative is close to the observed inflection frequency.
4.3.2 Particle characterization experiments

Once the basic functionality of the platform was confirmed and the determined electrical properties of the particles were ascertained to be comparable with other studies, variations in both particle size and carrier liquid conductance were introduced in the experiments. The purpose was to search out the functional particle size range of the platform. Towards the smaller particle sizes the Brownian force gradually becomes a more dominating force than DEP (Pethig & Markx 1997). It is worth mentioning that all of these measurements were done once for single particles, since comparable results were obtained by averaging the mobility of multiple particles.

Fig. 12 illustrates the effect of the Brownian force during the nDEP experiment. Due to the slow mobility of the particle (diameter of 0.98µm) and the multiplicity of random movement, the frame rate was set to 0.2 frames/second. A lower frame rate averages the movement to emphasize the DEP effect, but the noise caused by the Brownian motion is still evident. A comparison to a "well behaving" particle (Fig. 13) shows that the linear fitting of particle distance from the center (ln(R[m])) has a clear and clean correlation, whereas the correlation for the particle suffering from Brownian motion is poorer.
Fig. 12. Effect of Brownian force, where Brownian motion effectively adds noise to the measured particle mobility (upper figure). The noise due to Brownian motion is still evident when the natural logarithm of particle distance (lower figure) has been determined (V, published by permission of © 2012 IEEE).

Fig. 13 shows results from experiments with a diameter 5.98µm polystyrene carboxylate particle in three different dilutions of KCL electrolyte (0.1mM, 0.5mM and 1.0mM). This illustration shows that the higher the conductance of the carrier liquid is, the faster the negative DEP appears to mobilize particles; a similar observation has also been made and discussed by Ermolina & Morgan (2005). Basically this can become a restricting boundary for particle measurement, if the particles are able to move so fast that their mobility is no more measurable.

Fig. 13. The effect of carrier fluid conductivity variations on the negative dielectrophoretic mobility of particles. Carrier fluid conductivity increases from left to right. The figures are plotted in the same scale in order to amplify the effect or conductivity variation. When conductivity increases, mobility increases as well (V, published by permission of © 2012 IEEE).
The particle properties determined in the experiments described in this chapter are collected in Table 2, 3 and 4. The columns list particle diameter $\Omega$, DEP mobility coefficient $\alpha$, correlation coefficient $r^2$, and particle permittivity $\varepsilon_p$ and particle conductivity $\sigma_p$ respectively.

**Table 2. Determined parameters and particle properties in 0.1 mM potassium chloride (V, published by permission of © 2012 IEEE).**

<table>
<thead>
<tr>
<th>Ø [µm]</th>
<th>$\alpha$ (nDEP)</th>
<th>$r^2$</th>
<th>$\varepsilon_p$</th>
<th>$\sigma_p$ [mS/m]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.98</td>
<td>-0.016</td>
<td>0.96</td>
<td>27 $\varepsilon_0$</td>
<td>0.5</td>
</tr>
<tr>
<td>2.07</td>
<td>-0.051</td>
<td>0.99</td>
<td>43 $\varepsilon_0$</td>
<td>0.8</td>
</tr>
<tr>
<td>4.08</td>
<td>-0.129</td>
<td>0.96</td>
<td>54 $\varepsilon_0$</td>
<td>1.0</td>
</tr>
<tr>
<td>5.98</td>
<td>-0.219</td>
<td>0.99</td>
<td>59 $\varepsilon_0$</td>
<td>1.1</td>
</tr>
<tr>
<td>9.65</td>
<td>-0.366</td>
<td>0.95</td>
<td>65 $\varepsilon_0$</td>
<td>1.2</td>
</tr>
</tbody>
</table>

**Table 3. Determined parameters and particle properties in 0.5 mM potassium chloride (V, published by permission of © 2012 IEEE).**

<table>
<thead>
<tr>
<th>Ø [µm]</th>
<th>$\alpha$ (nDEP)</th>
<th>$r^2$</th>
<th>$\varepsilon_p$</th>
<th>$\sigma_p$ [mS/m]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.98</td>
<td>-0.079</td>
<td>0.88</td>
<td>50 $\varepsilon_0$</td>
<td>4.8</td>
</tr>
<tr>
<td>2.07</td>
<td>-0.046</td>
<td>0.97</td>
<td>46 $\varepsilon_0$</td>
<td>4.5</td>
</tr>
<tr>
<td>4.08</td>
<td>-0.170</td>
<td>0.99</td>
<td>47 $\varepsilon_0$</td>
<td>4.6</td>
</tr>
<tr>
<td>5.98</td>
<td>-0.354</td>
<td>0.97</td>
<td>48 $\varepsilon_0$</td>
<td>4.7</td>
</tr>
<tr>
<td>9.65</td>
<td>-0.806</td>
<td>0.98</td>
<td>51 $\varepsilon_0$</td>
<td>5.0</td>
</tr>
</tbody>
</table>

**Table 4. Determined parameters and particle properties in 1.0 mM potassium chloride (V, published by permission of © 2012 IEEE).**

<table>
<thead>
<tr>
<th>Ø [µm]</th>
<th>$\alpha$ (nDEP)</th>
<th>$r^2$</th>
<th>$\varepsilon_p$</th>
<th>$\sigma_p$ [mS/m]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.98</td>
<td>-0.015</td>
<td>0.95</td>
<td>30 $\varepsilon_0$</td>
<td>5.4</td>
</tr>
<tr>
<td>2.07</td>
<td>-0.040</td>
<td>0.99</td>
<td>50 $\varepsilon_0$</td>
<td>8.8</td>
</tr>
<tr>
<td>4.08</td>
<td>-0.138</td>
<td>0.99</td>
<td>53 $\varepsilon_0$</td>
<td>9.3</td>
</tr>
<tr>
<td>5.98</td>
<td>-0.351</td>
<td>0.98</td>
<td>48 $\varepsilon_0$</td>
<td>8.6</td>
</tr>
<tr>
<td>9.65</td>
<td>-0.807</td>
<td>0.99</td>
<td>51 $\varepsilon_0$</td>
<td>9.1</td>
</tr>
</tbody>
</table>

The conductivity of the carrier fluid has no notable effect on the determined permittivity values of the particles, whereas the conductivity of particles is strongly dependent of the properties of carrier fluid. This is eventually a noteworthy aspect, and it could be explained by the conductive double layer (Stern layer and diffuse layer) that is formed around the particle when it is exposed to a non-uniform electric field.
When comparing the permittivity values of the 0.98µm particles can be seen a wide variation range, from $27\varepsilon_0$ to $50\varepsilon_0$. This could be explained by the Brownian motion that adds noise to the particle position graphs and thus affects the determined particle properties. One reason for this could be the single particle measurement, since no averaged particle positions were used and thus the Brownian motion could have more effect on the obtained results than with averaging.

### 4.3.3 Particle mobility versus particle size

It has been discussed in various research papers that particle mobility is either linearly or quadratically dependent on particle size (Wang & Ou-Jang 2010). These results have been determined from the Equation 1 for DEP mobility and its opposite force, viscosity. Proving this experimentally is, however, a more complicated task because both alternatives seem to return satisfactory results. To establish the true situation a comparison should be made to find out which of these is correct.

![Fig. 14. Visualized comparison between linear and quadratic fittings. Mobility plotted as a function of particle size (blue dots), fittings for the data series are shown with red lines (V, published by permission of © 2012 IEEE).](image)

This figure shows the results at varying conductivities, with the mobility data fitted in both linear and quadratic fashion as a function of particle size. At first
glance both of them appear satisfactory, but when the conductivity of the carrier liquid increases the difference can be seen more clearly: the quadratic fitting gives correlations of 0.999, 0.973 and 0.966 respectively, while the correlations for the linear fitting are slightly lower. This finding is consistent with theory: particle mobility should behave quadratically (Morgan et al. 1999, Tsukahara et al. 2000), since the dielectrophoretic force (Equation 1) is proportional to the cube of the particle radius and the counter force (the Stokes force, Equation 12 friction force) is proportional to particle radius.

4.3.4 Particle behaviour in three dimensions

Experiments with a microscope showed that particles also tend to have mobility in the z-direction; this is illustrated in Fig. 15. The tracked particle is at first out of focus, but as time passes it dives into the plane of focus. This finding led to experiments with a digital-inline-holographic-microscope (DIHM) that is capable of simultaneously reconstructing the entire sample volume.

![Fig. 15. Microscope images of the depth directional mobility of a particle during a DEP experiment. The particle indicated with a red square dives from out of focus into the focus.](image-url)
Paper II reports the first 3D experiments with the ITO platform. These experiments clearly confirmed that DEP has a restricted functionality range in the depth direction, which is explained by the quadratic attenuation of field strength as a function of distance. In addition, the experiments confirmed that particles tend to have mobility also in the depth direction. Fig. 16 illustrates that while DEP controls particles tightly on the electrode plane, it has no effect whatsoever at a distance of 140µm from the electrode plane. The particle was also observed to dive, which excited curiosity concerning the particle routes experiencing nDEP and pDEP, and how particle speed changes between electrode plane projection and in the direction of particle mobility.

Fig. 16. Holographic reconstruction of six consequential frames, at 10 µm (left) and 140 µm (right) above the electrodes. In the figure at 10 µm depth nDEP has an effect on the particle, whereas the particles 140 µm above the electrode plane are not affected by the DEP force; however, some unidentified fluid flow seems to mobilize the entire particle population (II, published by permission of © 2010 Elsevier).

Fig. 17 is from an experiment with nDEP. While observing the phenomenon with DIHM the path of a single particle was traced, and the distance of this particle from the electrode plane is color-coded. When the nDEP control voltage was switched on, the particle tended to jump in the z-direction, up from its initial position; when the particle was pushed by the nDEP force towards the neutral electric field area, it was also pulled down towards the electrode plane of the platform by gravitation. This is partly a new finding, as most of the previous studies have only observed particle behavior in the electrode plane and thus this
three-dimensional route could not be seen. This aspect has been briefly discussed by Morgan & Green (2003), but no studies related this behavior could be found.

Fig. 17. Particle track in nDEP-experiment, reconstruction of DIHM (left) and particle in 3D (right). In both figures the particle moves from right to left (IV, published by permission of © 2011 Photonics Society of Poland).

Fig. 18 illustrates experiments with pDEP where the particles have mobility also outside the active area of the platform. When comparing the observations to the previous DIHM experiment (Fig. 16), at a distance of 80 µm from the electrode plane the mobility of particles could no more be caused by pDEP. As mentioned in literature by Jones (1995), AC electro-osmosis is at its most effective in low frequency experiments. Particle mobility between the electrode tips is caused by DEP, whereas when the particle has migrated to the electrode edge, the fluid flow induced by AC electro-osmosis causes particle mobility.

Fig. 18. Particle track in pDEP-experiment, reconstruction of DIHM data (left) and particle in 3D (right). In both figures the particle moves from right to left (IV, published by permission of © 2011 Photonics Society of Poland).

These single particle tracking results were used to determine the speed of particles in the direction of particle mobility, and also as an electrode plane projection which is similar to traditional microscope measurements. The speeds were determined from both nDEP and pDEP. Even though the speed of a particle in the direction of movement is different from its speed in projection, the difference is
so negligible that in practice there is no difference in particle properties calculated using either of these speed values.

Fig. 19. Left: Particle speed in nDEP. Blue bars indicate speed in the direction of mobility, red bars the speed on projection to the electrode plane. Right: Particle speed in pDEP. Blue bars indicate speed in the direction of mobility, red bars indicate speed on projection to the electrode plane (IV, published by permission of © 2011 Photonics Society of Poland).

In Fig. 19 the results of pDEP experiments also include particle speed outside the active region of DEP. The nDEP experiment observations began from the electrode edge. The overall difference between the speeds is not significant, but when a particle is simultaneously under the influence of gravitation and DEP, the sum vector of its mobility is faster than the mobility vector parallel with the electrode plane. The pDEP experiment results including speed comparison figures show that when a particle is inside the active DEP region these speeds are similar and slow, but when the AC electro-osmosis becomes dominant the particle tends to move very fast in comparison to DEP mobility. However, inside the active region could not be found any large overall difference between particle speeds in the direction of movement and in projection.
5 Discussion

The ITO electrode platform used in the experiments was found to be well suited for particle mobility observations and for the measurement of their dielectric properties, even though it had the same restrictions that apply to all 2D geometries. The most effective working distance from the electrode plane is limited to approximately 10–20µm due to quadratic attenuating electric field strength and the thin electrodes used in the 2D platforms. Above this distance the efficiency factor $\beta$ decreases, thus also decreasing the effect of the DEP force responsible for the mobility of the particle. By observing particles at a distance of 140µm could be concluded that the DEP force is totally overwhelmed by the thermal fluid flow. It would be beneficial, however, if the dielectric properties of particles could be determined without the need to control the particle depth position by restricting the depth of the measuring chamber, as this would allow for example a higher throughput of particles. Even though the 3D structure could provide a higher throughput and more efficient DEP force in the entire depth scale of the flow channel, clogging of the device is in all probability due to the relatively narrow gap between the electrodes, which is required in order to produce sufficiently high electric field strengths. In 2D platforms the DEP efficiency close to the electrode plane is high, and therefore the disturbing effect of other electrokinetic forces to particle mobility is relatively small. On the other hand, various friction effects may affect particle mobility when it is in close contact with the platform. For the best usability, the observed 2D mobility of the particle together with the calculations using the first order equations to describe the interdependence of mobility and the dielectric properties of the particle should give accurate enough results, irrespective of the 3D motion and the exact depth position of the particle, the only allowable adjustment being the value of the efficiency factor $\beta$ in Equation 15.

From the comparison between the results of this thesis and those from previous studies by Kuokkanen (2010) and Watarai et al. (1997), done on metal electrodes of chromium and gold, it can be stated that the dielectric properties measured using an ITO electrode platform were similar to those measured with the metal electrodes used in the previous studies. The various boundary effects (e.g. friction) are not taken into account in the first order equations, but they are quite likely to affect the mobility of particles on the platform; the inconsistent results reported concerning the behavior of mobility as a function of particle size might be a manifestation of this. Here it was observed that particle mobility as a
function of particle size is closer to quadratic behavior, as predicted by the theory suggesting that simple first order equations could be used to calculate the dielectric properties of particles.

It was something of a surprise to observe that within the active region of the platform, there was only a small difference between particle mobility when observing it in the direction of platform and in the direction of mobility. This suggests that 2D observations together with some adjustments in the efficiency factor $\beta$ would be enough to calculate the dielectric properties of particles, even though some 3D motion is always present in deep channels due to various electrokinetic and hydrodynamic effects. Studies using electric field simulations have suggested that an electric field gradient tends to have only a planar effect on particles, and that depending on particle position any z-directional movement is more likely caused by gravity or AC electro-osmosis. An exception to this was found in the experiments of nDEP, where the repulsion force of the high electric field at first pushes particles away from the electrode plane also in the z-direction, and thus in that sense a small speed difference seems realistic. Morgan & Green (2003) have mentioned that when frequency increases, the magnitude and thereby the effect of AC electro-osmosis decreases. According to the results of this thesis, AC electro-osmosis was evident only in the experiments of pDEP: AC electro-osmosis drags the particles within the areas where DEP did not have a net effect, and the particle routes during the experiments revealed that the observed phenomenon was an AC electro-osmotic fluid flow.

Experimental observations of the effect of carrier fluid conductivity variations on the conductivity of the measured particle suggests that in order to get reliable results of particle properties, carrier fluid conductivity should be either known exactly or kept at a reasonable level ($\gg 1\text{mS/m}$ and allowing some variation), just as predicted by theory. Controlling carrier fluid conductivity, however, may not be straightforward in all cases, especially in industrial process environment. Another obvious restriction of the DEP method when using a fixed size 2D electrode geometry is that the size range of measurable particles is limited and in fact simply determined by the dimensions of the electrode geometry. This might become a limiting factor in environments consisting of a wide and/or unknown range of particle sizes.
6 Concluding remarks

The dielectric properties of materials are as important for industry and science as they are complex to measure. Some applications have been developed for decades, while others have only just reached the level of being recognized scientifically. The full potential of DEP as a measurement method has not been fully understood and no major applications have as yet been found, even though the phenomenon could be employed for material characterization, to the fractionation process (Kang et al. 2008, Kang et al. 2006, Choi & Park 2005) or even to transport (Jones 2002) and dispense liquid in small volumes (Fan et al. 2008, Schwartz et al. 2004). At the moment the phenomenon is well mastered and interest in it is increasing rapidly.

In this study the electric field of the DEP hyperbolic quadrupole geometry has been modeled, the theory of DEP discussed and the main results and findings described in detail. A DEP platform with transparent ITO electrodes is utilized for a simple task: to mobilize various sets of particles in a carrier fluid of various concentrations of potassium-chloride electrolyte. The dielectric properties of the particles have been determined several times, observing both their planar and three-dimensional mobility. According to the theory of DEP particle mobility is proportional to particle size, and this was also demonstrated empirically. The attained dielectric properties of particles were compared to other studies of particle characterization based on DEP mobility, and the results were found to be comparable. Particle mobility in projection to the electrode plane was compared to mobility in the direction of particle mobility. The main findings of this study are:

- The DEP platform with patterned ITO electrodes was found to be fully functional, and additionally the particle characterization results obtained from the experiments were comparable to previous studies where metal electrodes were used.
- DEP mobility is proportional to the square of particle size. Even though both linear and quadratic fittings yield satisfying results, the DEP theory, supports quadratic behavior as well.
- In the active region of the DEP platform no significant difference was found between particle speeds in planar direction and in the direction of 3D mobility. Mobility in the z-direction is often a result of gravity or buoyancy.
During the DEP experiments was also found z-directional mobility due to the DEP force. Both theory and literature assume that the DEP effect on this particular platform is planar only; however, at the beginning of nDEP experiments the repulsion force was so strong that particles were pushed away from the electrode plane.

With a fixed size DEP platform the particle size could vary by a tenfold. The smallest particle size is determined by the relation between Brownian motion and DEP force (DEP should dominate the Brownian motion in order to execute reliable measurements), while the largest possible particle size is restricted by the feasible physical size.

The transparency of ITO electrodes enabled simultaneous multiphenomena observation with a 3D imaging device (Digital in-line holographic microscopy), for example the observation of AC electro-osmosis during the pDEP experiments. Previously such observations have been challenging due to opaque electrodes.

In view of the obtained results, particle characterization and fractionation using a 2D platform could be possible even when the particles are not in immediate proximity (closer than 20µm) to the electrode plane. The height of the flow channel could be increased up to 100µm to decrease the risk of channel clogging while still retaining a feasible functionality of the DEP effect; this improves the potential of the platform implementation for industrial solutions. Moreover, the use of a 3D structured DEP platform could in principle provide benefits in industrial uses, but it still does not completely eliminate the clogging risk due to the narrow electrode gap required to produce sufficiently strong electric fields. The 2D structure could be made wide enough for particle fractioning to decrease the clogging risk, and long enough to retain the overall efficiency of the flow-through channel.

This study shows that by using an ITO based DEP platform, particles can be characterized with the same accuracy as with platforms using metal electrodes. The measurement performance was good even in single particle measurements, so that averaging of the mobility of multiple particles is not necessarily required. The main target was not to fully and correctly characterize particle properties but instead to obtain knowledge of individual particles in a larger population. The ITO based DEP platform enables simultaneous multi-phenomena observations, and thus may be concluded that the full capabilities of transparent electrodes have not yet been thoroughly researched.
The hyperbolic quadrupole DEP platform with ITO electrodes offers a useful concept for particle characterization, for example for flow-through DEP platforms in target processes. The true applications of DEP as a method are still being searched; it does not require a large particle population, when even a single particle could be mobilized in the active region of the platform. In addition, the platform created using transparent and patterned ITO electrodes has potential that has not yet been fully exploited. The future of the combination of DEP and ITO looks promising.
References


Original publications


Reprinted with permission from SPIE (I), Elsevier ltd. (II), IEEE (III, V) and Photonics Society of Poland (IV).

Original publications are not included in the electronic version of the dissertation.


458. Ferreira, Denzil (2013) AWARE: A mobile context instrumentation middleware to collaboratively understand human behavior


460. Koscela, Adrian (2013) Theory of rational decision-making and its applications to adaptive transmission

461. Lauri, Janne (2013) Doppler optical coherence tomography in determination of suspension viscosity

462. Kukko, Jarmo (2013) Gas sensors based on nanostructured tungsten oxides

463. Reiman, Arto (2013) Holistic work system design and management: — a participatory development approach to delivery truck drivers’ work outside the cab

464. Tammela, Simo (2013) Enhancing migration and reproduction of salmonid fishes: method development and research using physical and numerical modelling

465. Yadav, Animesh (2013) Space-time constellation and precoder design under channel estimation errors


468. Remes, Jukka (2013) Method evaluations in spatial exploratory analyses of resting-state functional magnetic resonance imaging data

469. Oravisjärvi, Kati (2013) Industry and traffic related particles and their role in human health

470. Czajkowski, Jakub (2013) Optical coherence tomography as a characterization method in printed electronics

Book orders:
Granum: Virtual book store
http://granum.uta.fi/granum/
Mikko Haapalainen

DIELECTROPHORETIC MOBILITY OF A SPHERICAL PARTICLE IN 2D HYPERBOLIC QUADRUPOLE ELECTRODE GEOMETRY