Investigations of induced charge electrokinetic phenomena

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**NANYANG
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UNIVERSITY**

**INVESTIGATIONS OF INDUCED CHARGE
ELECTROKINETIC PHENOMENA**

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SCHOOL OF MECHANICAL AND AEROSPACE
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INVESTIGATIONS OF INDUCED CHARGE ELECTROKINETIC PHENOMENA

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Abstract

Induced charge electrokinetic phenomena are receiving increasing attention recently due to their promising potential applications for particle and fluid manipulations in micro/nanofluidics. Unlike linear electrokinetics, induced charge electrokinetic phenomena are caused by interactions between applied electric field and the self-induced polarisation surface charges on conducting or polarisable surfaces. The corresponding electric double layer formed on the surfaces possesses a zeta potential linearly proportional to the applied electric field strength. Therefore, the resulting flow velocity is a quadratic function of the applied electric field strength. This characteristic not only enables induced charge electrokinetic phenomena to provide stronger fluid flows, but also generate net fluid flows in AC electric fields.

Pair interactions in induced charge electrophoresis of conducting cylinders are analytically characterised based on the limits of thin electric double layer and weak applied electric field assumptions. A uniform electric field is applied normal to the axis of cylinders in two basic ways: perpendicular and parallel to the connecting line of the two cylinder centres. The study reveals that microvortices are generated on the cylinder surfaces due to the nonlinearity of induced zeta potentials, which is useful in fluid mixing in micro/nanofluidics. Furthermore, due to the nonuniformity of the surrounding electric field and the asymmetry of the surrounding fluid flow, the cylinders are driven into motion: translating away from each other under the perpendicular applied electric field and migrating towards each other under the parallel applied electric field. The electrostatic force generated by the nonuniform surrounding electric field is much smaller than the induced charge electrophoretic force due to asymmetric surrounding fluid flow, and diminishes to zero significantly as the distance between the two cylinders increases. Thus, the cylinder motion can be accurately described by the induced charge electrophoretic velocities.

Moreover, pair interactions between a conducting and a non-conducting cylinder in uniform electric fields are also analytically investigated with electric fields imposed perpendicular and parallel to the connecting line of the two cylinder

centres. The results show that electroosmosis and induced charge electroosmosis are generated on the non-conducting and the conducting cylinders, respectively. The nonuniform local electric field and the corresponding asymmetric fluid flow drive the cylinders into motion: both translating and rotating under the perpendicular applied electric field, while solely migrating along the x -axis under the parallel applied electric field. The velocity component due to electrostatic force is negligible compared to that due to electrophoresis or induced charge electrophoresis. Since the pair interactions reduce as distance increases, the cylinder velocities decrease accordingly.

Furthermore, efficient mixing is of significant importance in numerous chemical and biomedical applications but difficult to realise rapidly in microgeometries due to the lack of turbulence. Hence, mixing enhancement by introducing Lagrangian chaos through electroosmosis or induced charge electroosmosis in an eccentric annulus is proposed. The analysis reveals that the created Lagrangian chaos can achieve homogeneous mixing much more rapidly than either pure electroosmosis or pure induced charge electroosmosis. The systematic investigations on the key parameters, ranging from the eccentricity, the alternating time period, the number of flow patterns in one time period, to the specific flow patterns utilised for the Lagrangian chaos creation, show that the Lagrangian chaos is considerably robust. The system can obtain good mixing effect with wide ranges of the eccentricity, the alternating time period, and the specific flow patterns utilised for the Lagrangian chaos creation, so long as there are two flow patterns in one time period. When the applied electric field is large, the Lagrangian chaos created by induced charge electroosmosis can achieve homogenous mixing much more rapidly than that of electroosmosis.

Lastly, induced charge electroosmosis around a conducting cylinder in AC electric fields is experimentally investigated through micro particle image velocimetry (μ PIV). The captured velocity vector fields show that four vortices are generated around the cylinder. The fluid velocity is linearly proportional to

the square of the electric field strength, obtains a peak value as the electric field frequency increases, and increases as the NaCl concentration increases.

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Nomenclature

Symbols

A	Surface area [m ²]
a	A positive constant in the bipolar coordinates
c	Half distance between the two cylinders [m]
c_0	Electrolyte concentration [mol/m ³]
D	Mass diffusivity [m ² /s]
d	Distance between the centers of the inner and outer cylinders [m]
E	Electric field strength [V/m]
$\mathbf{e}$	Unit vector
F	Force [N]
F_a	Faraday constant [9.65×10^4 C/mol]
$\mathbf{I}$	Unit tensor
k_B	Boltzmann constant [1.38×10^{-23} J/K]
M	Moment [N·m]
$\min()$	minimum value of the parameters in parenthesis
N_A	Avogadro constant [6.02×10^{23} mol ⁻¹]
N_{fp}	Number of flow patterns in one time period
N_m	Number of mesh bins in the eccentric annulus meshing
N_p	Number of tracer particles in particle tracking
N_{piw}	Number of particles in the interrogation window of PIV processing
N_{ip}	Number of image pairs in the PIV processing
Pe	Peclet number
p	Pressure [Pa]
R	Radius [m]
R_r	Radius ratio of the eccentric annulus, $R_r = R_i/R_o$
$\mathbf{s}$	Distance vector [m]
T	Time period [s]

T_a	Absolute temperature [K]
t	Time [s]
U	Velocity component of cylinder [m/s]
U_0	Velocity scale [m/s]
$\mathbf{u}$	Velocity vector of fluid flow [m/s]
u	Velocity component of fluid flow [m/s]
V	Electric field function [V]
V_{p-p}	Peak-to-peak value of AC voltage [V]

Greek symbols

Δt	Time step [s]
δ	Error
ε_f	Dielectric permittivity of electrolyte solution [F/m]
ε	Eccentricity
ζ	Zeta potential [V]
θ_0	Electric field phase angle [°]
Π	Stress tensor [N/m ²]
ρ	Mass density of electrolyte solution [kg/m ³]
μ	Viscosity of electrolyte solution [Pa·s]
Λ	A constant in the Debye length definition [mol ^{1/2} /m ^{1/2}]
λ_D	Debye length [m]
ϕ	Electrical potential [V]
χ^2	Goodness of fitting
ψ	Stream function
Ω	Rotational velocity of cylinder [s ⁻¹]
(τ, σ)	Coordinates of the bipolar coordinate system

Overscripts

$\sim$ Dimensionless Parameters

Superscripts

$\perp$ Parameters in perpendicular applied electric field

$\parallel$ Parameters in parallel applied electric field

Subscripts

c Constant parameters

d Parameters related to viscous drag

e Parameters related to electric field

f Parameters related to fluid

H Parameters related to hydrodynamics

i Parameters related to induced quantities

s Parameters related to Helmholtz-Smoluchowski velocity

T Parameters related to thermal voltage

n Normal component of parameters

t Tangential component of parameters

x Parameter component along the x -axis in the Cartesian coordinate system

y Parameter component along the y -axis in the Cartesian coordinate system

τ Parameter component along the τ -axis in the bipolar coordinate system

σ Parameter component along the σ -axis in the bipolar coordinate system

∞ Parameters related to far-field

Abbreviations

AC	Alternating current
ACEO	Alternating current electroosmosis
DC	Direct current
EDL	Electric double layer
EO	Electroosmosis
EP	Electrophoresis
ES	Electrostatic
ICEK	Induced charge electrokinetics
ICEO	Induced charge electroosmosis
ICEP	Induced charge electrophoresis
NaCl	Sodium chloride
PDMS	Poly(dimethylsiloxane)
μ PIV	Micro particle image velocimetry

Chapter 1

Introduction

1.1 Background and Motivation

Fluid and particle manipulations in micro/nanofluidics are receiving increasing attention recently due to their great potential applications in various areas, ranging from chemical analysis [14], biomedical diagnosis [15], drug delivery [16], to environment [17] and food monitoring [18]. However, the commonly small Reynolds number in micro/nanofluidics poses a challenge for fluid manipulation. To crack this hard nut, numerous novel methods have been proposed, which generally utilise external fields, ranging from thermal, acoustic, magnetic, to electric fields [19]. Among all these methods, electrokinetics, which serves as an electrically-motivated and non-mechanical technique for fluid and particle manipulations, is attracting more and more attention recently [2].

In conventional electrokinetics, the solid surface is non-conducting (non-polarisable). When it contacts electrolyte solution, counterions in the solution are attracted to the surface, which form an electric double layer (EDL). After an external electric field is applied, the ions within the EDL are driven into motion, leading to a bulk fluid flow, referred to as electroosmosis (EO). When the surface is movable, e.g., a particle, it moves by electrophoresis (EP). The effective slip velocity on the surface is linearly proportional to the applied electric field. Thus, neither a net fluid flow nor a particle motion can be produced in AC electric fields, which is undesirable since many harmful issues, e.g., electrochemical reactions, would arise in DC electric fields. Moreover, this linear relationship leads to a small velocity, which results to a laminar flow as the characteristic length is small in micro/nanofluidics. Hence, mixing becomes extremely difficult since it solely relies on molecular diffusion. Furthermore, the uniform EO flow also causes difficulty in

flow manipulation. Additional mechanical parts, such as actuators, pumps, valves, are required for different applications.

Therefore, there is a need to explore other electrokinetic phenomena. Induced charge electrokinetics (ICEK), which describes fluid flow on conducting (polarizable) surface and the corresponding particle motion in electric fields, is attracting increasing attention as it is capable of flow and particle motion generation in either DC or AC electric fields. When the surface submerged in electrolyte solution is conducting (polarisable) and subjected to an electric field, it becomes polarised immediately: one part of the surface is positively charged, while the other part is negatively charged. Counterions in the solution are attracted to each part of the surface, establishing an induced EDL with a nonlinear zeta potential. The applied electric field interplays with its self-induced EDL, leading to a fluid flow, which is referred to as induced charge electroosmosis (ICEO). When the ICEO around a particle is asymmetric, the particle moves through induced charge electrophoresis (ICEP). As the induced zeta potential is a linear function of the applied electric field, $\zeta_i \propto E$, the resulted slip velocity on the surface is quadratically proportional to the applied electric field, $u_s \propto E^2$. Therefore, the ICEO velocity can be much stronger than that of the conventional EO and capable of vortex generation. Researchers have conducted many studies on ICEK [20]. However, as a relatively new field, some underlying physics of ICEK are still not well understood and require deeper studies. Moreover, potential applications of ICEK also need further exploration.

1.2 Objectives and Scope

This thesis aims to improve the understanding of basic physics and further to explore potential applications of ICEK in micro/nanofluidics. It is well-known that various types of particles, conducting or non-conducting, are provided for micro/nanofluidics due to recent advancement in material science

and nanotechnology. Thus, it is critical to understand how particles behave in uniform electric fields, especially the motion of conducting particles, i.e., the ICEP. In practical application, a lot of particles are suspended together in electrolyte solution. Hence, it is important to understand the interactions between particles so as to improve the development of particle manipulations. Studies on the interactions between particles of different properties are rare, if not none. Such interactions may lead to some promising results, which could be useful in micro/nanofluidics e.g., deposition pattern control in nanofluid droplet drying, therefore, need systematic studies.

Efficient mixing is of critical importance for many lab-on-a-chip systems for chemical reactions [14], biological analysis [21], particle synthesis [22], medical analysis [23], colloid science [24], etc. Therefore, developing effective techniques for a rapid and homogenous mixing remains a hot topic of extensive scientific and technological interest. However, this has long been a challenge in micro/nanofluidics due to the commonly small Reynolds number and laminar flow. So far various mixing methods have been proposed and studied in the effort to enhance mixing [25]. Electrically induced mixing has been confirmed to be simple and effective in prompt mixing [26], among which ICEO presents a great potential in mixing enhancement thanks to its inborn nature of vortex generation. However, as ICEO is intrinsically laminar, diffusion is required for transportation across streamlines. This badly deteriorates the mixing effect of ICEO when diffusion is weak. To overcome this limitation, introducing Lagrangian chaos into ICEO micromixers is a good choice. Thus, the mixing effect of Lagrangian chaos created by ICEO needs to be investigated.

Using ICEO around cylinders in microchannel for mixing and pumping [1] has been proposed in the past. Detailed theoretical studies on mixing effect through ICEO around cylinders have also been conducted [27]. While experimental investigations on ICEO around cylinders are not extensively reported yet. Levitan et al. [28] captured ICEO on a lying metal wire. Canpolat et al. experimentally

studied ICEO of Newtonian [29] and non-Newtonian [30] fluids around a conducting cylinder. They also captured ICEO around touching cylinders [31]. However, the influence of electrolyte concentration on ICEO has not been investigated. Thus, a detailed experimental study is required to explore this effect.

Consequently, the objectives and scope of this thesis include,

(1) To analytically evaluate pair interactions between conducting cylinders, and between conducting and non-conducting cylinders in uniform electric fields; capture the attraction and repulsion effects between the cylinders; analyse the translational and rotational velocities of the cylinders in detail. The study on pair interactions between a conducting and a non-conducting cylinders offers the first tip on the interactions between particles of different prosperities. Such analysis may improve the understanding of particle behaviours in uniform electric fields and facilitate the development of particle manipulations in micro/nanofluidics.

(2) To analytically investigate flow dynamics of EO and ICEO in an eccentric annulus; and numerically evaluate mixing behaviours of Lagrangian chaos created by periodically alternating two or more EO or ICEO flows with wide ranges of parameters. The study could contribute to the understanding of the flow dynamics of the EO and ICEO in eccentric annulus, and the mixing effect of the corresponding Lagrangian chaos so as to offer guidelines for chaotic micromixer development.

(3) To experimentally investigate ICEO around a cylinder in AC electric fields through micro particle image velocimetry (μ PIV); and characterise the influence of electrolyte concentration, applied electric field strength, applied electric field frequency, etc., on the ICEO. The study aims to contribute to the physical insights of ICEO and facilitate the future development of ICEK applications.

These studies aim to provide a better understanding of the fundamental physics of ICEK phenomena and their applications in micro/nanofluidics.

1.3 Outline of the Thesis

The thesis consists of seven chapters and includes both fundamental studies and practical applications of ICEK phenomena. The organisation of the thesis is shown as follows,

Chapter 1 gives a basic idea of ICEK phenomena in micro/nanofluidics, and illustrates the research motivation and objectives.

Chapter 2 is the literature review. The theoretical basics, the up-to-date research situations, and the promising applications of ICEK are introduced.

Chapter 3 shows the investigations on pair interactions in induced charge electrophoresis of conducting cylinders. The attraction and repulsion effects between the cylinders are captured and analysed.

Chapter 4 presents the evaluations of pair interactions between conducting and non-conducting cylinders in uniform electric fields. The results reveal that the cylinders not only translate but also rotate. A detailed analysis is carried out on the cylinder velocities.

Chapter 5 demonstrates the flow dynamics of EO and ICEO in eccentric annulus, and numerically analyses the mixing behaviors of Lagrangian chaos created by EO or ICEO. It is presented that two and four microvortices are generated within the annulus by the EO and ICEO, respectively. Moreover, the created Lagrangian chaos works robustly with wide ranges of parameters for a rapid and homogenous mixing.

Chapter 6 presents the experimental studies on ICEO flow around a conducting cylinder in AC electric fields using μ PIV. The results clearly show four vortices around the cylinder. The influences of the electric field strength, the electric field frequency, and the electrolyte concentration are characterised in detail.

Chapter 7 provides conclusions of this thesis including proposed work for future studies.

Chapter 2

Literature Review

2.1 Induced Charge Electrokinetic Phenomena

Electrokinetics deals with the motion of fluids or particles due to interactions between applied electric fields and EDLs adjacent to solid surfaces. It is connected with EDL on solid surface, which offers it great advantages in miniaturization for larger surface-volume ratio. Moreover, with benefit of other special advantages, such as no moving part, electrical actuation and sensing, easy integration with microelectronics, etc., electrokinetics plays an important role in many lab-on-a-chip systems for biomedical diagnosis, electrochemical analysis, etc. However, conventional electrokinetics has assumed a linear relationship between the velocity and the applied electric field based on the assumption of constant zeta potential, which is reasonable for insulating surfaces, but not for conducting (polarisable) surfaces. Furthermore, conventional electrokinetics has other disadvantages, such as failing to generate net flow or particle motion in AC electric fields; requiring a large voltage across microchannel so as to sustain a sufficiently strong electric field, which will easily result in electrochemical reaction and Joule heating; unable to manipulate flow locally; incapable of particle separation by size or shape through electrophoresis [2, 20]. To overcome these disadvantages, various kinds of nonlinear electrokinetics have been proposed and studied, among which ICEK remains a research hotspot recently due to its great potential applications in micro/nanofluidics [2]. The recent research achievements of ICEP and ICEO will be presented in Sections 2.2 and 2.3, respectively. In this section, the theoretical fundamentals of ICEK phenomena are illustrated.

When a conducting (polarisable) surface submerged in electrolyte solution is subjected an external electric field, it becomes polarised immediately: positive

Figure 2.1: Charging process of a conducting cylinder in a uniform electric field. (a) Initial electric field; (b) Steady state electric field [1].

charges accumulates at one part of the surface, while negative charges at the other part. Correspondingly, negative and positive ions in the solution are attracted to each part of the surface as shown in Figure 2.1(a). This process continues till an induced EDL is formed on the solid surface and all the electric field lines are expelled as shown in Figure 2.1(b). The characteristic time of the induced EDL formation is

$$t_c = \frac{\lambda_D R}{D}, \quad (2.1)$$

where λ_D is the Debye length; R is the characteristic length; and D is the mass diffusivity of electrolyte solution. In microfluidics, the typical value of t_c ($\sim$ ms) is much larger than the Debye relaxation time λ_D^2/D ($\sim \mu$ s) for bulk ionic screening and much smaller than the diffusion time L^2/D ($\sim$ s) for the relaxation of bulk concentration gradients [2].

The zeta potential of the induced EDL is a linear function of the applied electric field [32]

$$\zeta_i = -\phi + \phi_c, \quad (2.2)$$

where ϕ is the applied electrical potential on solid-fluid interface; and ϕ_c is an

integral constant given as

$$\phi_c = \frac{\int_A \phi dA}{A}, \quad (2.3)$$

where A is the surface area.

The tangential component of the applied electric field acts on the ions within the induced EDL and drives the ions into motion. In the limit of thin EDL and small zeta potential, the effective slip velocity on the surface is defined by the Helmholtz-Smoluchowski formula [33]

$$\mathbf{u}_s = -\frac{\varepsilon_f \zeta_i}{\mu} E_t \mathbf{e}_t, \quad (2.4)$$

where ε_f and μ are dielectric permittivity and viscosity of electrolyte solution, respectively; ζ_i is the induced zeta potential on the surface; E_t is the electric field component that acts on and is tangential to the surface; and $\mathbf{e}_t$ is the unit vector tangent to the surface.

This slip velocity leads to ICEO, which is a Stokes flow in micro/nanofluidics

$$\rho \frac{\partial \mathbf{u}}{\partial t} = -\nabla p + \mu \nabla^2 \mathbf{u}, \quad \nabla \cdot \mathbf{u} = 0, \quad (2.5)$$

where ρ and μ are the mass density and viscosity of electrolyte solution, respectively; $\mathbf{u}$ and p are the velocity and pressure fields, respectively.

When the ICEO flow around a particle is asymmetric, which can be resulted from asymmetric geometry and/or boundary, nonuniform applied electric field, or heterogeneous surface property, the particle moves through ICEP. The total force and moment acting on the particle can be obtained from the surrounding electric and flow fields through the following equations

$$\mathbf{F} = \int_A \boldsymbol{\Pi} \cdot \mathbf{e}_n dA, \quad (2.6)$$

$$\mathbf{M} = R \int_A \mathbf{e}_n \times (\boldsymbol{\Pi} \cdot \mathbf{e}_n) dA, \quad (2.7)$$

where $\mathbf{e}_n$ is the unit vector normal to the surface; and $\mathbf{\Pi}$ is the stress tensor

$$\mathbf{\Pi} = -p\mathbf{I} + \mu \left[\nabla \mathbf{u} + (\nabla \mathbf{u})^T \right] + \varepsilon_f (\mathbf{E}\mathbf{E} - \frac{1}{2}E^2\mathbf{I}), \quad (2.8)$$

where $\mathbf{I}$ is a unit tensor. The first term is the isotropic pressure tensor; the second is the viscous stress tensor of Newtonian fluid; and the last term is the Maxwell stress tensor of linear dielectric medium with constant permittivity. In a quasisteady Stokes flow, the translational and rotational velocities of freely suspended particles are determined by the constraints that net force and moment exerted on the particles are zero [2]

$$\sum \mathbf{F} = 0, \quad \sum \mathbf{M} = 0. \quad (2.9)$$

2.2 Recent Advancements of ICEP

Electrophoresis is referred to as the movement of particles or macromolecules in electrolyte solution with an electric field applied. The movement velocity depends on applied electric field strength, surface zeta potential, particle size and shape, ionic concentration, viscosity and temperature of the surrounding electrolyte solution, etc. As a most widely used technique in the particle/biocell separations, electrophoresis has been applied in various disciplinary, including biochemistry, biomedical science, food analysis, and environment science, etc. Two most common electrophoretic separation methods are gel electrophoresis and capillary electrophoresis. Although gel electrophoresis could provide high-resolution analysis, its application is constrained by the slow separation speed because of Joule heating. Compared to gel electrophoresis, capillary electrophoresis could dissipate Joule heating efficiently benefited from its high area-volume ratio, thus, provides a large separation speed. Thanks to the rapid advancement of lab-on-a-chip technology, alternative separation techniques have been provided. However, most of the studies are focused on the non-conducting particles. Recently, various

kinds of electrically conducting and polarisable particles (e.g., carbon, gold, silver, nickel) are involved in micro/nanofluidic systems due to the rapid development of microfluidics and nanotechnology. Therefore, understanding the fundamentals of ICEP of polarisable particles becomes critically important. This section presents detailed research status of the ICEP.

2.2.1 ICEP of particles due to asymmetric geometry

As early as 1980s, studies on ICEP have been conducted in colloid science [34, 35, 36]. Murtsokin [37] for the first time experimentally observed the ICEP motion of irregular quartz particles in a uniform AC electric field, but no theoretical explanation was proposed since the motion is only observed near a wall, not in the bulk solution. Recently, Bazant and Squires predicted that polarisable particles can undergo arbitrary translation and/or rotation by ICEP in a uniform electric field as long as they hold certain broken symmetries [5], such as asymmetric surface property, boundary condition, and geometry shape. The former two cases will be introduced in Sections 2.2.2 and 2.2.3. The latter case just poses a start to explain Murtsokin's observation and needs further experimental and theoretical studies, which is introduced in this section.

The general expressions of ICEP translational and rotational velocities of non-spherical particles were derived by Yariv [38]

$$\mathbf{U} = \frac{\varepsilon_f R}{\mu} \mathbf{C} : \mathbf{E}_\infty \mathbf{E}_\infty, \quad (2.10)$$

$$\boldsymbol{\Omega}_i = \frac{\varepsilon_f}{\mu} \mathbf{D} : \mathbf{E}_\infty \mathbf{E}_\infty, \quad (2.11)$$

where R is the characteristic dimension of particles; $\mathbf{C}$ and $\mathbf{D}$ are dimensionless third-order vector and pseudovector, respectively, which are integrals of the electric potential over the particle boundary. Therefore, Equations (2.10) and (2.11) enable to predict particle motions without solving the flow field. Squires and Bazant

[5] solved a number of nearly symmetric objects through boundary perturbation methods and developed simple principles for motion prediction of certain shapes.

Figure 2.2: (a) ICEP rotation of a rod-like polarizable particle in a uniform electric field; (b) ICEP translation of particles when they are aligned with electric field [2].

Equations. (2.10) and (2.11) show that particles can both rotate and translate through ICEP as shown in Figure 2.2. Saintillan et al. [39] utilised ICEP to control the stability of ideally polarisable rods in a uniform AC electric field. Besides the rod-like particles, studies on spheroids and elliptical particles have also been reported [4, 40, 41]. Potential applications, such as microvalves, have been proposed based on the ICEP rotation of elliptical particles [41].

2.2.2 ICEP of particles due to boundary effect

Since particles in micro/nanofluidics are neither single nor unbounded, it is important to investigate the boundary effect on ICEP motion of particles. Wall effect on a cylinder or a sphere has been reported in Refs. [3, 42, 43, 44]. All these studies show that particles, be it cylindrical, spherical or elliptical, are repelled away from the non-conducting wall due to the ICEO flow on the particle as shown in Figure 2.3(a). When the particles are near a conducting wall, they approach the wall as shown in Figure 2.3(b). Zhao and Bau [42] found that a hydrodynamic force and an electrostatic force are generated on the cylinder near a non-conducting wall. The hydrodynamic force repels the cylinder away from the wall; while the

Figure 2.3: Wall effect on ICEP of a conducting cylinder near (a) a non-conducting and (b) an unscreened conducting wall [3].

direction of electrostatic force depends on the relative value of permittivities of the cylinder and the solution as it is due to the difference of permittivities. The electrostatic force dominates when the permittivity of the cylinder is small, while the hydrodynamic force dominates when the permittivity of the cylinder is large. Kilic et al. [3] found that an ideally polarisable particle, cylindrical or spherical, experiences a repulsion force when it is located near a non-conducting wall in AC electric fields. Both hydrodynamic force and electrostatic force are repulsive, while electrostatic force is much smaller than hydrodynamic force. These results are consistent with the results of Zhao and Bau [42] under DC electric field condition.

The studies about wall effect on ICEP improve the understanding of the behaviours of conducting particles in electric fields. For further explorations, researchers focus their efforts on the case that particles are suspended in microchannels. Wu and Li [45] numerically studied the ICEP motion of an ideally polarisable spherical particle in a microchannel with a complete three-dimensional multiphysics model. Sugioka [4] numerically analysed the attitude and positioning control of elliptical conducting particles in a converging microchannel through ICEP. These two studies show that surrounding channel walls repel particles to the microchannel center. The elliptical particles not only experience repulsion forces but also rotation torques. The ICEP torque rotates the elliptical particles and aligns them with the electric field as indicated in Figure 2.4. This may be

helpful for particle manipulation in micro/nanofluidics.

Figure 2.4: Attitudes and positions of the elliptical particles in a converging microchannel at (a) $t/T_0 = 0$ and (b) $t/T_0 = 20$ [4].

Particle-particle interactions in ICEP are also of great interests. Saintillan [46], Wu et al. [47], and Sugioka [48] characterised pair interactions in ICEP of spherical [46, 47] and elliptical particles [48], respectively. The studies on spherical particles show that the particles attract and repel each other when they are aligned parallel and perpendicular to the applied electric field, respectively [46, 47]. The elliptical particles behave similar to the spherical particles [48].

2.2.3 ICEP of particles due to heterogeneous surface property

As aforementioned, ICEP motion could also happen due to heterogeneous surface property. A canonical example is Janus particle, whose two hemispheres have different properties. It rotates to align the interface between its two hemispheres along the applied electric field [5]. Figure 2.5 shows the ICEP motion of metal-dielectric Janus particles. For any orientation, Janus particle moves towards its non-conducting hemisphere due to ICEO flows on the conducting hemisphere. As Janus particles always move towards its non-conducting part, an ever-rotating pinwheel structure is proposed based on ICEP of Janus particles as shown in Figure 2.5(b). This pinwheel rotates in any DC or AC electric field (of sufficient low frequency) till the electric field is turned off, which would find applications in electric field sensing, motion generation, etc.

Figure 2.5: ICEP motion of metal-dielectric Janus particles. (a) Perpendicular and (b) parallel to the applied electric field towards its non-conducting side. (c) Ever-rotating pinwheel composed of Janus particles [5].

ICEP motion of Janus particles in uniform AC electric fields was recently observed by Gangwal et al. [49]. Consistent with theoretical predictions of Ref. [5], the Janus particles move perpendicular to the electric field towards its non-conducting side. The ICEP motion persists even after the Janus particles are attracted to a glass wall. The influence of key parameters, ranging from electric field strength, concentration of NaCl solution, radius of particles, to frequency of applied electric field, on the velocities of Janus particles are studied in detail. The ICEP velocity increases as the electric field strength increases, while decays as NaCl solution concentration increases. It approaches zero when NaCl solution concentration reaches 1×10^{-2} mol/L. Similar concentration dependence has also been observed in AC electroosmosis (ACEO) and other nonlinear electrokinetic phenomena. The results show that the ICEP velocity peaks at an intermediate frequency. An interesting feature captured in the experiments is that Janus particles are always attracted to the nearby non-conducting wall, which is different from the ICEP motion of homogenous particles near a non-conducting wall as introduced in Section 2.2.2. A possible explanation is that this attraction might be a result of ICEP moment, which turns Janus particles towards a nearby wall and makes them tilt during translating perpendicular to the applied electric field [3].

Daghighi et al. [50] numerically simulated ICEP motion of Janus particles in microchannels with a complete three-dimensional multiphysics model. Their results agree well with the experimental results of Gangwal et al. [49]. Through the comparison among ICEP motion of fully non-conducting, fully conducting, and Janus particles in a microchannel, Daghighi et al. [50] found that ICEO microvortices on the conducting part of the Janus particle acts like an engine and pushes the Janus particle to move with a larger velocity than that of the fully conducting and non-conducting particles. Such numerical conclusions are experimentally verified in their latter studies [51]. Boymelgreen et al. [52, 53] modelled the ICEP motion of leaky dielectric Janus particles. The key parameters including the dielectric permittivities of two hemispheres are analysed. Later on they reported experimental observation of ICEP rotations of Janus doublets in uniform AC electric fields [54], which validated the proposed idea of Figure 2.5(c) in a different configuration. Peng et al. [55] captured and analysed the breaking ICEO flow around Janus spheres.

Besides the fundamental studies, promising potential applications based on ICEP motion of Janus particles have also been proposed. Daghighi et al. [6] designed a microvalve utilising ICEP motion of Janus particles. As illustrated in Figure 2.6, a Janus particle is placed in the junction of three microchannels. When the electric fields are applied, ICEO flows push the Janus particle to one side of the junction and block the entrance of that microchannel. By adjusting the two applied electric fields, different microchannels could be blocked. This three-dimensional transient numerical study shows that the Janus particle-based microvalve is feasible for flow switching and flow rate control. The comparison studies on fully conducting and non-conducting particles reveal a non-suitability.

Advancement of microfluidics and nanotechnology opens the possibility of new applications of heterogeneous particles. Theoretically, heterogeneous particles can be designed and fabricated into certain irregular shapes with heterogeneous surface properties for specific applications. Their complex ICEP motions offer

Figure 2.6: Switching process of Janus particle-based microvalve [6].

them application potentials ranging from separation, concentrating, directional transport, drug delivery, electric field sensing, to force and/or moment supply.

2.3 Recent Advancements of ICEO

2.3.1 Fundamentals

Although pioneering studies on ICEO around polarisable particles has been reported decades ago [34, 35, 36], the research enthusiasm on this subject is triggered till the discovery of ACEO [56], which is a branch of ICEO, and fluid pumping through ACEO [11]. Besides pumping, ICEO has many other potential applications, such as mixing, concentrating, particle transport, electric field sensing, etc., benefited from its essential nonlinear characteristics. Compared

to conventional EO, ICEO offers several advantages: 1) the nonlinearity of ICEO enables net flow generation by a small AC electric field, which in turn eliminates electrochemical reactions; 2) zeta potential is tunable by changing applied electric field since it is induced by the field, thus, providing tunable velocities; 3) by breaking the symmetry of the particles or electrodes, a net ICEP motion or ICEO pumping can be achieved and greatly enhanced [57].

However, many underlying issues of ICEO are still not well understood although many studies have been carried out on ICEO recently. Thus, further studies are needed for a better understanding. Soni et al. [58] found that surface conduction creates a large normal electric field and greatly reduces tangential electric field as well as the slip velocities when the applied electric field is large. Gregersen et al. [59] found that Debye length exerts a significant influence on ICEO. Pascall and Squires [7, 60] presented an automated experimental system, which is capable of rapid ICEK data collection under a variety of conditions. Microelectrodes controllably “contaminated” by dielectric layers were experimentally studied in this experiment rig. A theory accounting the effect of dielectric coating was developed and a quantitative agreement between the experiment results and the theoretical predictions was found over nearly one thousand conditions (Figure 2.7). Through this experimental system, the physicochemical effects on ICEO can be studied and optimisation of these flows for lab-on-a-chip devices can be done. As microelectrode is used frequently in ACEO flows, this experiment rig is valuable for ICEK studies.

ICEO can be induced on any polarisable surfaces, not only the conducting (ideally polarisable) objects with or without “contaminated” dielectric layers, but also the dielectric objects with finite polarisability. To understand this phenomenon, Zhao and Yang theoretically studied ICEO on dielectric surfaces [61, 62, 63]. They analytically analysed ICEO in a microchannel embedded with two symmetric polarisable dielectric blocks, and found that ICEO microvortices become stronger when the polarisability of dielectric blocks increases [61]. In Refs.

Figure 2.7: The scaled slip velocity versus dimensionless frequency of the applied electric field. The equivalent circuit models used in (a,d), (b,e) and (c,f) are presented in (g), (h) and (i), respectively [7].

[62, 63], they developed effective electrokinetic boundary conditions for ICEO on a leaky dielectric surface in uniform AC electric fields, and discovered that counter-rotating microvortices on the embedded leaky dielectric surfaces can be deformed, relocated, eliminated and even direction-reversed, which will be useful in mixing since Lagrangian chaos may be induced.

Squires and Bazant [1] analytically studied ICEO around a conducting cylinder. Levitan et al. [28] first reported experimental observation of ICEO around a metal wire lying in a microchannel. Zhao and Yang analytically investigated ICEO around a leaky dielectric cylinder [62]. Canpolat et al. captured ICEO around conducting cylinders surrounded with Newtonian [29, 31] and non-Newtonian [30] fluids in AC electric fields using μ PIV. Davidson et al. [8] conducted a direct numerical simulation on ICEO around a conducting cylinder in DC electric fields. They found that chaotic sub-vortices appear in ICEO when the applied electric field is large as shown in Figure 2.8. This is because an extended space charge region, which is hydrodynamically unstable, is formed between EDL and bulk solution in

Figure 2.8: Instantaneous snapshots of (a) the dimensionless salt concentration and (b) the dimensionless free charge density with flow streamlines superposed for $E_\infty R/\phi_T=50$. The arrows track a single void [8].

large electric fields due to the concentration polarisation. Together with chaotic sub-vortices, concentration and free charge density fields also become unstable (Figure 2.8). This direct numerical simulation presents a better agreement with experimental results compared to the asymptotic model.

2.3.2 Applications

2.3.2.1 Mixing

ICEO presents potential for various applications in micro/nanofluidics. A key application is mixing due to its inborn nature of vortex generation. Two possible mixing designs are proposed by Squires and Bazant as shown in Figure 2.9, which utilise metal cylinders and metal coatings to generate ICEO microvortices for mixing. The detailed theoretical studies on the configurations shown in Figures 2.9(a) and (b) are carried out by Sugioka [27], and Zhao and Yang [63], respectively.

Systematic evaluations on mixing behaviours of ICEO around triangular cylinders were carried out by Wu and Li [9, 32] and Harnett et al. [64]. Both numerical and experimental investigations are conducted. The results show that

Figure 2.9: ICEO mixing by (a) an array of metal cylinders in a vertical electric field, and (b) metal objects (or coatings) embedded in microchannel walls in an electric fields along flow direction [1].

Figure 2.10: Mixing effect using ICEO flow around conducting triangular cylinders [9].

ICEO microvortices around triangular cylinders can greatly enhance mixing as indicated in Figure 2.10. Through comparison of mixing effect among several shapes of cylinder, Jain et al. [65] identified that the right triangle cylinder is the optimal one in ICEO mixing. The numerical algorithm developed by Shuttleworth et al. [66] and the topology optimisation method developed by Gregersen et al. [67] can be helpful for the design and optimisation of ICEO micromixers. Daghighi and Li [10] proposed a novel micromixer using ICEP motion of a conducting particle in a channel junction as shown in Figure 2.11. Their results show that ICEO microvortices around the particle tremendously enhance the mixing of different solutions. As ICEO is intrinsically laminar, the mixing effect of these ICEO micromixers is badly limited when diffusion is weak since diffusion is the only

mechanism for transportation across streamlines. To overcome this limitation, Zhao and Bau [68] proposed to introduce Lagrangian chaos into ICEO micromixers through periodically alternating two intersecting ICEO flows, which show a great advancement in mixing.

Figure 2.11: Mixing effect (a) without particle, (b) with a non-conducting particle, and (c) with a conducting particle [10].

2.3.2.2 Pumping

Besides mixing, ICEO could also be used for other applications. Squires and Bazant proposed to achieve pumping by ICEO [57]. As shown in Figure 2.12(a), fluid is pumped in from two microchannels and supplied to the other two microchannels using ICEO around a circular cylinder; while in Figure 2.12(b), fluid is pumped along the direct microchannel through ICEO on triangular cylinders. As a branch of ICEO, ACEO also presents a great potential in fluid pumping. Fluid pumping through asymmetric ACEO flows was first proposed by Ajdari [11] as shown in Figure 2.13. After that, many studies were carried out on this topic. Brown et al. [69] experimentally studied ACEO micropumps with coplanar microelectrodes of different widths and gaps. Green et al. [70] numerically and experimentally studied ACEO flow on coplanar microelectrodes. Through the comparison between numerical and experimental results, they found that constant phase angle impedance model for the EDL polarisation impedance could correctly predict the size and shape of the vortices rolls. This model could also be beneficial to the asymmetric coplanar ACEO micropump designs. Ramos et al. [71] conducted numerical studies on the asymmetric coplanar ACEO micropumps proposed by

Brown et al. [69], and a good agreement is achieved between their numerical results and the experimental results of Brown et al. [69].

Figure 2.12: ICEO pumping in (a) microchannel junction and (b) direction microchannel [1].

Figure 2.13: Schematic diagram of ACEO pumps with asymmetric electrode arrays. (a) Nonuniform surface coatings; (b) Nonuniform surface height [11].

Studer et al. [72] designed and fabricated an asymmetric coplanar ACEO micropump, and conducted in depth experimental studies. A velocity up to $500 \mu\text{m/s}$ is achieved. Unexpectedly, a flow reversal was observed at high frequencies around $50 \sim 100 \text{ kHz}$. The pumping velocity increases as the voltage increases, but at high frequencies, the velocity changes into the opposite direction, i.e., flow reversal appears. The pumping velocity significantly decreases as KCl solution concentration increases from 10^{-3} mol/L to 10^{-2} mol/L , and eventually approaches

zero. The unexpected flow reversal is also experimentally observed at high frequencies in the planar ACEO micropumps by Wu et al. [73].

Olesen et al. [74] modified the existing theory of planar ACEO micropumps by taking vertical confinement of the pumping channels, nonlinear surface capacitance of the EDL, and current injection from Faradaic reactions into account, and numerically studied the asymmetric coplanar ACEO micropumps with this model. Their numerical results agree well with experiments in many aspects, however, the flow reversal at high frequencies is still not predictable by the model. Besides the flow reversal at high frequencies, a flow reversal at high voltages in asymmetric coplanar ACEO micropumps was also observed by Garcia-Sanchez et al. [75].

To understand these unexpected flow reversal at high frequencies and high voltages, and velocity saturation at high solution concentrations, Bazant and coworkers theoretically and experimentally studied the electrolyte dependence of the ACEO flow [76] and developed new theoretical models for coplanar ACEO micropumps [77, 78]. Bazant et al. [76] presented the first investigation of the electrolyte dependence in ACEO. It is stated that the classic dilute solution theory breaks down in ACEO as ion crowding occurs on the electrodes because of the large applied electric field. By taking steric effects and a charge-dependent viscoelectric effect into consideration, a modified theory was proposed, which predicts that ACEO are influenced by salt concentration, ion size and valence. Experiments show significant variations in ACEO pumping as salt concentration ($10^{-6} \sim 10^{-3}$ mol/L KCl) and/or ionic species (10^{-3} mol/L NaCl, CaCl_2 , MgCl_2 , KI, KOH) change. The concentration dependence is roughly logarithmic at all frequencies. And the ACEO velocity approaches zero around 10^{-2} mol/L. For frequency dependence, ACEO velocity peaks around 1 kHz, and reverses around 10 kHz. Moreover, a strong dependence on ionic species is observed.

Storey et al. [77] established two theoretical models for planar ACEO pumps by adopting the finite-sized ions theory, i.e., taking the steric effects into consideration. The numerical results show the first demonstration of flow reversal of planar

ACEO pumps at high frequencies. The Bikerman model and Carnahan-Starling model show similar tendency, and both agree well with experimental results in a qualitative manner. However, to fit the experiment results well quantitatively, both of these two models result in an ion radius with an order of magnitude larger than the hydrated radius of ions. Furthermore, they found that flow reversal can be predicted by any model as long as steric effect is considered. Bazant et al. [78] took a further step. They not only took steric effect into consideration, but also accounted increased viscosity in the condensed layer at large voltages. Through this model, they found that EDL capacitance decreases and electroosmotic mobility saturates at large voltages. Moreover, flow reversal at high frequencies and salt concentration dependence of ICEO become predicable. To further understand and explain the flow reversal at high voltages observed by Garcia-Sanchez et al. [75], more detailed and in-depth studies are needed.

Besides the valuable works on planar ACEO micropumps, Bazant and coworkers also made another contribution in ACEO micropumps development. Bazant and Ben [12] proposed a brand new three-dimensional ACEO micropump as shown in Figure 2.14, which could provide a flow rate with an order of magnitude larger than the state-of-the-art coplanar micropumps under the same applied electric field and characteristic length. At the same time, Urbanski and Bazant et al. [79] fabricated a three-dimensional symmetric ACEO micropump and experimentally studied its performance against the state-of-the-art asymmetric planar ACEO micropumps. Their results validated the former theoretical predictions of Bazant and Ben [12].

The reason why three-dimensional ACEO micropumps work much more efficiently than planar ACEO micropumps is that they turn the microvortices on one half of the microelectrode into “fluid conveyor belt” by stepping up another half of electrode. The “fluid conveyor belt” speeds up fluid transport in three-dimensional ACEO micropumps but hinder that in planar ACEO micropumps as shown in Figure 2.14. According to the theoretical predication of Bazant and Ben [12], by stepping up half surface of each microelectrode, the flow rate of asymmetric

Figure 2.14: Schematic diagram of “fluid conveyor belt”. (a) Planar ACEO micropump; (b) Non-planar ACEO micropump [12].

coplanar ACEO micropumps can be increased more than threefold. Moreover, the symmetric three-dimensional ACEO micropump works better than the asymmetric one, providing a flow rate up to five to six times of that of the asymmetric one. The experimental study on the symmetric three-dimensional ACEO micropumps conducted by Urbanski and Bazant et al. [79] offers a practical proof for these theoretical predictions.

As a faster flow (mm/s) is obtainable at a small voltage (< 10 V, at the order of battery voltage), the symmetric three-dimensional ACEO micropumps open a gate to portable lab-on-a-chip devices with fast tunable flow rate. Further studies have been conducted by Bazant and coworkers. Urbanski and Bazant et al. [80] investigated the effect of step height on the performance of symmetric three-dimensional ACEO micropumps. Numerical simulations show that the three-dimensional micropumps perform better as the step height increases till an optimal step height is reached, which is in a qualitative agreement with experiments. Within a broad range around the optimal step height, the observed flow rates are much larger than that of planar pumps with the same voltage and feature size. A significant flow reversal appears at the optimal frequency when step height is

small, which has not been explained theoretically. Burch and Bazant [81] improved the three-dimensional ACEO micropumps by changing the boundary conditions of fluid flow rather than the geometries of microelectrode. Their numerical simulation shows that flow rate of the symmetric three-dimensional ACEO micropump could be further doubled by fabricating a non-polarisable vertical surface in electrode of micropumps. This is because the “fluid conveyor belt” is amplified and slip velocities are increased as the opposing flows on vertical surfaces are removed. Huang et al. [82] fabricated a symmetric three-dimensional ACEO micropump for portable biomedical microfluidics. The experimental investigation shows that this micropump could provide a 1.3 kPa head pressure and a 1.3 mm/s effective slip velocity with no flow reversal under a low AC voltage. Moreover, by utilising low ionic strength solutions as working solution, this three-dimensional ACEO micropump can be applied to pump biological solutions in separation devices, offering a potential for application in portable biomedical devices.

Besides the valuable studies of Bazant and coworkers, some other researchers have also conducted useful studies on ACEO micropumps. Kim et al. [83] designed diagonal/herringbone shaped asymmetric coplanar electrode in ACEO to achieve mixing and pumping simultaneously. Their three-dimensional numerical simulation shows that the helical flow plays a key role in mixing, while the slip velocity at the centre region is more important to pumping than that near the side wall. When two microelectrodes are located closely, EDL overlap may happen, which affects the ICEO flows. To investigate this situation, Talapatra et al. [84] developed a mathematical model for the EDL overlap between the gap of two planar microelectrodes in parallel plate microelectrode ACEO flows. The numerical results satisfactorily explain the ACEO flow under EDL overlap conditions, and show that the overlap effect has a larger impact at lower frequencies. Although Ref. [84] investigate the parallel plate configuration, coplanar and three-dimensional ACEO micropumps could also draw valuable references from it.

Symmetric ACEO flows offer no net pumping flows. Thus, to achieve pumping

effect, all the aforementioned ACEO micropumps are geometrically asymmetric. And these geometrically asymmetric ACEO micropumps provide a good pumping performance. However, there are also some other methods to offer net pumping flows. Wu [85] proposed a technique by adding a DC bias to the applied AC electric field since an asymmetric electrode polarisation can be induced by this DC bias, which results in a breaking flow symmetry, thus, produces a net pumping flow. The experimental results show that a good pumping effect can be realised with several voltages applied, which makes it suitable for portable microfluidic devices. Furthermore, this technique could also concentrate and sweep particles into certain location for detection.

Islam and Reyna [86] achieved a bidirectional flow by adding a DC bias in ACEO. Their numerical simulations show that by changing the polarity of electrical signal, bidirectional flow can be created on symmetric electrode array. Moreover, the experimental results on several types of electrode arrays reveal that the asymmetric electrode polarisation induced by DC bias could produce a much larger flow rate than the asymmetric coplanar electrode array with unbiased AC electric field. Islam and Askari [87] improved the bidirectional ACEO micropump through hydrophobic surface modification. The experimental studies on several symmetric ACEO micropumps with different surface coatings show that Si-NP/PDMS nanocomposites hydrophobic monolayer could eliminate electrode reaction, thus, a higher voltage can be applied and a larger pumping velocity is achievable. Mansuripur et al. [88] reported another symmetry broken method in ACEO. They found that the capacitive coupling between the gate electrode and the microscopic stage produce an asymmetric component in the ACEO flows. This novel capacitive coupling offers an additional way to manipulate ICEO flows over polarisable surfaces and provides a new way to produce net ACEO pumping.

2.3.2.3 Particle and biocell manipulation

Particle and biocell manipulation is another important application of ICEO. Positioning colloid particles or biocells for detection through ACEO has been reported [89, 90, 91, 92]. As ACEO flow can be reversed by adding a DC bias, the particles or biocells can be removed from the stagnation lines, which offers a possibility to write and erase the suspended particles, bacteria, or microalgae on microelectrodes [2]. Furthermore, particle trapping using ICEO on a metal strip has also been reported [85, 93, 13]. Ren et al. [13] realised precise position-controllable particle trapping. As indicated in Figure 2.15(a), a metal strip is subjected to an AC electric field and connected to another AC electric source. ICEO flow on the metal strip traps particles on its stagnation region as illustrated in Figure 2.15(c~e). By adjusting the AC electric source connected to the metal strip, the particle trapping line can be manipulated as shown in Figure 2.15(b). Therefore, a position-controllable particle trapping is realised through this chip.

Figure 2.15: (a) Schematic of experiment chip. (b) Distance of particle trapping line to the metal strip centerline versus potential deviation at 600 Hz. (c~e) Particle trapping process by ICEO under an AC voltage of 62.5 V at 600 Hz: (c) $t = 0$, initial distribution; (d) $t = 12$ s, particles move to the central region; (e) $t = 108$ s, steady trapping is realised [13].

ICEO has also been explored for other applications. Leinweber et al. reported ICEO in porous media [94] and realised continuous flow demixing by ICEO [95]. Saintillan et al. [39] used ICEO for stability control of freely suspended conducting cylinders sedimenting under gravity in a vertically applied electric field. Zhang et al. [96] achieved flow rate and concentration control through ICEO mixing in microchannel networks. Jain et al. [97] generated user-defined concentration gradient profile through ICEO. Yazdi [98] reported localized flow control through ICEO around conducting cylinders.

2.4 Summary

The basic concepts and latest research achievements of ICEK phenomena are presented in this chapter. The literature review shows that ICEK phenomena have been receiving increasing attention. A large number of studies have been conducted to offer a better understanding on the basic physics and explore potential applications in micro/nanofluidics. However, due to the complex nature of the subject, there are still many aspects that are not fully understood. ICEO around a single cylinder has been reported [1]. There is a lack of knowledge regarding ICEO around two cylinders and the corresponding ICEP motion of cylinders. Nevertheless, such multi-particles effect are important due to suspension and thus should be investigated. ICEO and chaotic mixing through ICEO in a concentric annulus has been presented [68]. However, a more practical situation, ICEO in an eccentric annulus has not been studied. Moreover, no systematic investigations have been conducted on chaotic mixing through ICEO in either a concentric or an eccentric annulus.

Chapter 3

Pair Interactions in Induced Charge Electrophoresis of Conducting Cylinders

3.1 Introduction

Electrophoresis (EP) has been widely used for particle and biocell manipulation in various areas, ranging from biochemistry [99, 100], biomedical science [101, 102], to pharmaceutical science [103]. Numerous studies have been conducted on it, such as particle-particle interactions of spherical particles [104, 105, 106], and particle-wall interactions of spherical particles [107] and circular cylinders [108]. Many theoretical models investigating the phenomena can be referred from Hunter [33] and Lyklema [109]. While induced charge electrophoresis (ICEP) received far fewer attentions compared to EP. Pioneering studies on ICEP were carried out in colloid science by Gamayunov et al. [34, 110], Dukhin and Murtsovkin [36], and Dukhin [35] decades ago. After the work on ACEO [56] and fluid pumping [71] achieved through ACEO by Ramos et al. [56, 71], the research enthusiasm on induced charge electrokinetics (ICEK), including ICEO and ICEP, was triggered. Many studies have been carried out and can be referred from Bazant and Squires [20].

Unlike the conventional electrokinetics, ICEK is due to the interactions between polarisation charges and the applied electric field [2], which results in a nonlinear zeta potential. Thus, microvortices are generated on the conducting (polarisable) surface, which makes ICEO an effective technique for fluid mixing in micro/nanofluidics. Analytical studies on the ICEO around a spherical particle [34] and a circular cylinder [1, 62] predict a quadrupolar flow on the particle/cylinder.

Experimental observation of ICEO microvortices around a platinum wire lying in microchannel was reported by Levitan et al. [28]. Due to the inborn nature of vortex generation, plenty of studies have been conducted on mixing performance of ICEO [9, 64, 68]. Harnett et al. [64] and Wu and Li [9] reported that ICEO can achieve an efficient mixing by positioning conducting triangle rods in a microchannel. Furthermore, fluid pumping [67, 82] and particle manipulation [5, 49, 52] are also realisable when ICEO flow is asymmetric.

As the advancement of material science and nanotechnology in recent years provides various kinds of conducting (ideally polarisable) particles (like nickel, gold, silver, etc.) for micro/nanofluidics [111, 112, 113], ICEP of polarisable particles has attracted the attentions of researchers again. Besides the pioneering study of Gamayunov et al. [34] on pair interaction of spherical particles, many studies were conducted on particle-wall [3, 45, 48] and particle-particle [47, 39, 114] interactions with particles of different shapes. Saintillan et al. [47] reported that ICEP can be utilised to control the suspension stability of cylindrical particles. The attraction effect of glass wall in ICEP of Janus particles in AC electric fields has been experimentally observed by Gangwal et al. [49]. Squires and Bazant [5] proposed an ever-rotating pinwheel composed of three Janus particles, which shows potential in applications of motion generation, electric field detection, etc. Experimental observation of ICEP rotation was reported by Boymelgreen et al. [54], although the structure geometry is different (a doublet composed of two Janus particles). Besides these complex structures, Yossifon et al. [40] found that spheroid particles can rotate (although not continually) when an external electric field is applied. Daghighi and Li simulated ICEP motion of a Janus particle suspended in a chamber and proposed to use this structure as a microvalve [6].

The actual situation is more complicated due to many particles in suspension. The surrounding electric field of one particle is distorted by the presence of other particles. Thus, it is important to understand the interactions between particles. This chapter characterises pair interactions of two infinitely long conducting

cylinders in a uniform applied electric field. The two-dimensional governing equations of electric and flow fields are solved in the bipolar coordinates with appropriate boundary conditions. The typical electric, velocity and pressure fields around the cylinders are presented. The repulsion and attraction effects of two cylinders are captured and discussed.

3.2 Mathematical Formulation

This section considers two infinitely long conducting (ideally polarisable) cylinders with radius R , freely suspended in the unbounded electrolyte solution. The Cartesian coordinate system is defined in such a way that the two cylinder centres are located on the x -axis. The weak external electric field of strength E_∞ is applied perpendicular and parallel to the connecting line of the two cylinder centres, i.e., along the y -axis and the x -axis, respectively (Figure 3.1). The EDLs on the cylinder surfaces are assumed to be much thinner than the radius of the cylinders and the distance between the cylinders. By introducing a bipolar coordinate system, the complex geometry can be conveniently represented. The relationship between the Cartesian coordinate and the bipolar coordinate systems is given by [115]

$$x = a \frac{\sinh \tau}{\cosh \tau - \cos \sigma}, \quad y = a \frac{\sin \sigma}{\cosh \tau - \cos \sigma}, \quad (3.1)$$

where $-\infty < \tau < \infty$, $0 < \sigma \leq 2\pi$; (τ, σ) indicates the coordinates of the bipolar coordinate system; a is a positive constant in the bipolar coordinates. The surfaces of cylinder-1 and cylinder-2 are represented by $\tau = -\tau_0$ and $\tau = \tau_0$, respectively.

3.2.1 Electric field

After the external electric field is imposed to the electrolyte solution, the two cylinders polarise simultaneously and the counterions in the electrolyte solution are attracted to the cylinder surfaces. Within an instant moment ($\sim 10^{-4}$) s [1],

Figure 3.1: The relationships among Cartesian coordinates, polar coordinates and bipolar coordinates. In the bipolar coordinates, the surfaces of the two cylinders are represented by $\tau = \pm\tau_0$; the x -axis and y -axis are indicated by $\sigma = 0$ and $\tau = 0$, respectively; $(-a, 0)$ and $(a, 0)$ are two foci of the bipolar coordinates; c is the half distance between the centres of the two cylinders; $R = a/\sinh \tau_0$ is the radius of the cylinders.

the cylinders are fully screened by the ions. Induced EDLs are formed enclosing the cylinder surfaces. Outside the EDLs, the bulk solution remains electrically neutral. Thus, the electrical potential ϕ satisfies the Laplace equation

$$\nabla^2 \phi = 0, \quad (3.2)$$

where ϕ is the electrical potential of the electrolyte solution.

Since the two cylinders are fully screened by the ions, the cylinders plus the EDLs act as ideal insulators. Therefore, no-flux boundary condition is applied at the outer edge of EDLs

$$\mathbf{e}_\tau \cdot \nabla \phi = 0 \quad \text{at} \quad \tau = \pm\tau_0, \quad (3.3)$$

where $\mathbf{e}_\tau$ is the unit vector normal to the cylinder surfaces in the bipolar coordinates.

The far-field boundary conditions are determined by the applied electric field,

given as

$$\phi \rightarrow -E_\infty y \quad \text{as} \quad r \rightarrow \infty, \quad (3.4)$$

and

$$\phi \rightarrow -E_\infty x \quad \text{as} \quad r \rightarrow \infty, \quad (3.5)$$

for the perpendicular and parallel applied electric fields, respectively.

Solving the Laplace equation together with the boundary conditions, the electrical potentials for the perpendicular and parallel applied electric fields are obtained as

$$\phi^\perp = -E_\infty a \frac{\sin \sigma}{\cosh \tau - \cos \sigma} - 2E_\infty a \sum_{n=1}^{\infty} \frac{e^{-n\tau_0}}{\sinh n\tau_0} \cosh n\tau \sin n\sigma, \quad (3.6)$$

and

$$\phi^\parallel = -E_\infty a \frac{\sinh \tau}{\cosh \tau - \cos \sigma} - 2E_\infty a \sum_{n=1}^{\infty} \frac{e^{-n\tau_0}}{\cosh n\tau_0} \sinh n\tau \cos n\sigma, \quad (3.7)$$

respectively, which turn out to be of same expressions as given by Keh et al. [108] on the electrophoresis of a non-conducting cylinder near a wall.

In Equations (3.6) and (3.7), the second terms on the right-hand side are the far-field electrical potentials. The superscripts $\perp$ and $\parallel$ indicate the perpendicular and parallel applied electric field, respectively, for all the equations in the thesis.

Electric field function V is related to electrical potential ϕ by the following relations [108]

$$\frac{\partial \phi}{\partial \tau} = -\frac{\partial V}{\partial \sigma}, \quad \frac{\partial \phi}{\partial \sigma} = \frac{\partial V}{\partial \tau}, \quad (3.8)$$

Substituting Equations (3.6) and (3.7) into Equation (3.8), the electric field function for the perpendicular and parallel applied electric fields are obtained as

$$V^\perp = E_\infty a \frac{\sinh \tau}{\cosh \tau - \cos \sigma} - 2E_\infty a \sum_{n=1}^{\infty} \frac{e^{-n\tau_0}}{\sinh n\tau_0} \sinh n\tau \cos n\sigma, \quad (3.9)$$

and

$$V^{\parallel} = -E_{\infty}a \frac{\sin \sigma}{\cosh \tau - \cos \sigma} + 2E_{\infty}a \sum_{n=1}^{\infty} \frac{e^{-n\tau_0}}{\cosh n\tau_0} \cosh n\tau \sin n\sigma. \quad (3.10)$$

Similarly, these two electric field functions are also of the same form as that given by Keh et al [108]. The second terms at the right-hand side of Equations (3.9) and (3.10) are the far-field electric fields.

The induced zeta potential ζ_i at cylinder-fluid interface depends on the external electric field by the following equation [32]

$$\zeta_i = -\phi + \phi_c, \quad (3.11)$$

where ϕ is the external electrical potential at cylinder-fluid interface; ϕ_c is an integral constant given as $\phi_c = \int_A \phi dA/A$, where A is the surface of the conducting cylinders [32].

Substituting Equations (3.6) and (3.7) into Equation (3.11), the induced zeta potential ζ_i on the two cylinder surfaces for the perpendicular and parallel applied electric fields are obtained as

$$\zeta_i^{\perp} = E_{\infty}a \frac{\sin \sigma}{\cosh \tau_0 - \cos \sigma} + 2E_{\infty}a \sum_{n=1}^{\infty} e^{-n\tau_0} \coth n\tau_0 \sin n\sigma, \quad (3.12)$$

and

$$\zeta_i^{\parallel} = \pm E_{\infty}a \left[\frac{\sinh \tau_0}{\cosh \tau_0 - \cos \sigma} + 2 \sum_{n=1}^{\infty} e^{-n\tau_0} \tanh n\tau_0 \cos n\sigma - 1 \right], \quad (3.13)$$

where the lower and upper signs of $\pm$ on the right-hand side of Equation (3.13) are for cylinder-1 ($\tau = -\tau_0$) and cylinder-2 ($\tau = \tau_0$), respectively.

According to the relationship between electric field and electrical potential ($\mathbf{E} = -\nabla\phi$), the tangential electric field on the cylinders surfaces (i.e., the local electric field along σ -direction on cylinders surfaces) can be obtained from Equations (3.6)

and (3.7), given as

$$E_{\sigma}^{\perp} = E_{\infty} \frac{\cosh \tau_0 \cos \sigma - 1}{\cosh \tau_0 - \cos \sigma} + E_{\infty} (\cosh \tau_0 - \cos \sigma) \sum_{n=1}^{\infty} 2ne^{-n\tau_0} \coth n\tau_0 \cos n\sigma, \quad (3.14)$$

and

$$E_{\sigma}^{\parallel} = \mp E_{\infty} \left[\frac{\sinh \tau_0 \sin \sigma}{\cosh \tau_0 - \cos \sigma} + (\cosh \tau_0 - \cos \sigma) \sum_{n=1}^{\infty} 2ne^{-n\tau_0} \tanh n\tau_0 \sin n\sigma \right], \quad (3.15)$$

where the lower and upper signs of $\mp$ on the right-hand side of Equation (3.15) are for cylinder-1 ($\tau = -\tau_0$) and cylinder-2 ($\tau = \tau_0$), respectively.

The local electric field around one cylinder is distorted by the presence of the other cylinder, which may exert electrostatic (ES) force and/or moment on the cylinders [46]

$$\mathbf{F}_e = \int_A \mathbf{\Pi}_e \cdot \mathbf{e}_n dA, \quad (3.16)$$

$$\mathbf{M}_e = R \int_A \mathbf{e}_n \times (\mathbf{\Pi}_e \cdot \mathbf{e}_n) dA, \quad (3.17)$$

where $\mathbf{e}_n$ is the unit vector normal to the cylinder surface, $\mathbf{\Pi}_e$ is the Maxwell stress tensor

$$\mathbf{\Pi}_e = \varepsilon_f (\mathbf{E}\mathbf{E} - \frac{1}{2} E^2 \mathbf{I}). \quad (3.18)$$

where ε_f is the dielectric permittivity of the fluid; and $\mathbf{I}$ is a unit tensor.

The electric field is expelled by the EDLs on the cylinders, thus, only tangential component of the electric field remains. The Maxwell stress tensor on the cylinder surfaces are normal, $\mathbf{\Pi}_e \cdot \mathbf{e}_n = -\varepsilon_f E_t^2 \mathbf{e}_n / 2$. Therefore, it can be concluded that $\mathbf{M}_e = 0$. Substituting Equations (3.6) and (3.7) into Equation (3.16), the ES force per unit length on cylinder-2 under the perpendicular and parallel applied electric

fields are obtained as

$$\begin{aligned}
 \mathbf{F}_e^\perp = & \pi\epsilon_f E_\infty^2 R \left\{ e^{-3\tau_0} \coth \tau_0 (2 \cosh 2\tau_0 + \sinh 2\tau_0 - 1) \right. \\
 & - \sum_{n=2}^{\infty} n e^{-(2n+1)\tau_0} \coth n\tau_0 \sinh \tau_0 [e^{2\tau_0} (n-1) \cosh \tau_0 \coth(n-1)\tau_0 \\
 & + (n+1) \coth(n+1)\tau_0 \cosh \tau_0 \\
 & \left. - 2e^{\tau_0} (n + n \coth n\tau_0 + \sinh 2\tau_0) + 2ne^{\tau_0} \cosh 2\tau_0] \right\} \mathbf{e}_x, \quad (3.19)
 \end{aligned}$$

and

$$\begin{aligned}
 \mathbf{F}_e^\parallel = & -\pi\epsilon_f E_\infty^2 R \left\{ \frac{e^{-3\tau_0} \sinh \tau_0 \tanh^2 \tau_0}{\cosh 2\tau_0} (\sinh \tau_0 - 2 \cosh 3\tau_0 - \sinh 3\tau_0) \right. \\
 & + \sum_{n=2}^{\infty} n e^{-(2n+1)\tau_0} \sinh \tau_0 \tanh n\tau_0 [2ne^{\tau_0} (2 \sinh^2 \tau_0 - \tanh n\tau_0) \\
 & + \cosh \tau_0 \{ e^{2\tau_0} (n-1) \tanh(n-1)\tau_0 \\
 & \left. - 4e^{\tau_0} \sinh \tau_0 + (n+1) \tanh(n+1)\tau_0 \} \} \right\} \mathbf{e}_x, \quad (3.20)
 \end{aligned}$$

respectively. The ES force on cylinder-1 has the same magnitude but opposite direction.

3.2.2 Flow field

Since the Reynolds number is small in ICEO flow [1] and the fluid is incompressible, the Stokes and continuity equations can be applied outside the thin EDLs. As the problem is two-dimensional (infinite cylinders), the stream function method can be adopted

$$\nabla^4 \psi = 0, \quad (3.21)$$

with the velocities in the bipolar coordinates defined as

$$u_\sigma = h \frac{\partial \psi}{\partial \tau}, \quad u_\tau = -h \frac{\partial \psi}{\partial \sigma}, \quad (3.22)$$

where $h = (\cosh \tau - \cos \sigma)/a$.

Following Jeffery's procedure [115], a general solution of the stream function for the present case is obtained as

$$\begin{aligned}
 h\psi = & h_1\tau \sin \sigma + \sum_{n=1}^{\infty} [e_n \cosh(n+1)\tau + f_n \cosh(n-1)\tau \\
 & + g_n \sinh(n+1)\tau + h_n \sinh(n-1)\tau] \sin n\sigma.
 \end{aligned} \tag{3.23}$$

The applied electric field exerts a force on the ions within the EDLs, driving the ions as well as the surrounding bulk solution into motion. Since the EDLs are thin, a tangential slip velocity is generated at the outer edge of the EDLs, which is defined by Helmholtz-Smoluchowski formula

$$\mathbf{u}_s = -\frac{\varepsilon_f \zeta_i}{\mu} E_\sigma \mathbf{e}_\sigma \quad \text{at} \quad \tau = \pm\tau_0, \tag{3.24}$$

where $\mathbf{e}_\sigma$ is the unit vector tangent to the cylinder surfaces in the bipolar coordinates; μ is the viscosity of the solution.

At far-field, the fluid remains quiescent

$$\mathbf{u} \rightarrow 0 \quad \text{as} \quad r \rightarrow \infty. \tag{3.25}$$

Substituting Equations (3.12) and (3.14) as well as Equations (3.13) and (3.15) into Equation (3.24), the slip velocities on cylinder surfaces under the perpendicular and parallel applied electric fields are obtained. Through Fourier transformation

and mathematical manipulation, these two slip velocities are presented as

$$\begin{aligned}
 \mathbf{u}_s^\perp = & U_0 \left\{ -\frac{e^{-2\tau_0}}{2} \left[\frac{1}{\sinh \tau_0} - 4 \sinh \tau_0 \right. \right. \\
 & - 2 \sum_{k=2}^{\infty} (c_{k,k+1}^\perp - c_{k,k-1}^\perp) + \frac{\tanh \tau_0}{\cosh \tau_0} \left. \right] \sin \sigma \\
 & - \sum_{n=2}^{\infty} \left[2e^{-n\tau_0} \coth n\tau_0 - e^{-(n+1)\tau_0} \frac{\cosh 2\tau_0}{\cosh \tau_0} \right. \\
 & \times (e^{2\tau_0} \coth(n-1)\tau_0 + \coth(n+1)\tau_0) \\
 & - 2 \left(\sum_{\substack{k=2, \\ n-k \geq 1}}^{\infty} \Gamma_{k,n-k}^\perp + \sum_{\substack{k=2, \\ k-n \geq 1}}^{\infty} (\Gamma_{k,n+k}^\perp - \Gamma_{k,k-n}^\perp) \right) \\
 & \left. + \left(\frac{n-1}{\sinh(n-1)\tau_0} - \frac{n+1}{\sinh(n+1)\tau_0} \right) \sinh \tau_0 \right] \sin n\sigma \Big\} \mathbf{e}_\sigma, \quad (3.26)
 \end{aligned}$$

where

$$\Gamma_{k,l}^\perp = \frac{\sinh^2 \tau_0 (\sinh 2k\tau_0 - k \sinh 2\tau_0)}{\sinh k\tau_0 (\cosh 2k\tau_0 - \cosh 2\tau_0)} e^{-l\tau_0} \coth l\tau_0, \quad (k \geq 2, l \geq 1), \quad (3.27)$$

and

$$\begin{aligned}
 \mathbf{u}_s^\parallel = & U_0 \left\{ \sinh \tau_0 \left[2e^{-2\tau_0} \left(\frac{\tanh 2\tau_0}{\cosh 2\tau_0} - \tanh \tau_0 \right) \right. \right. \\
 & + \sum_{k=2}^{\infty} (c_{k,k-1}^\parallel - c_{k,k+1}^\parallel) \left. \right] \sin \sigma \\
 & + \sum_{n=2}^{\infty} \left[\frac{n-1}{\cosh(n-1)\tau_0} - \frac{2ne^{-\tau_0}}{\cosh n\tau_0} + \frac{n+1}{\cosh(n+1)\tau_0} \right. \\
 & + \sum_{\substack{k=2, \\ n-k \geq 1}}^{\infty} \Gamma_{k,n-k}^\parallel + \sum_{\substack{k=2, \\ k-n \geq 1}}^{\infty} (\Gamma_{k,k-n}^\parallel - \Gamma_{k,k+n}^\parallel) \\
 & \left. + \frac{4e^{-n\tau_0} \sinh^3 \tau_0 (1 + \cosh 2\tau_0 - \sinh 2n\tau_0)}{\cosh 2\tau_0 \cosh(n+1)\tau_0 \cosh(n-1)\tau_0} \right] \sin n\sigma \Big\} \mathbf{e}_\sigma, \quad (3.28)
 \end{aligned}$$

where

$$\Gamma_{k,l}^{\parallel} = \left[\frac{2k \cosh \tau_0}{\cosh k \tau_0} - \frac{k-1}{\cosh(k-1)\tau_0} - \frac{k+1}{\cosh(k+1)\tau_0} \right] e^{-l\tau_0} \tanh l\tau_0. \quad (k \geq 2, l \geq 1). \quad (3.29)$$

In both Equations (3.26) and (3.28), U_0 is the natural velocity scale of the ICEO, defined as

$$U_0 = \frac{\varepsilon_f E_{\infty}^2 R}{\mu}. \quad (3.30)$$

Substituting Equation (3.23) into Equations (3.26)/(3.28) and (3.25) through Equation (3.22), the unknown coefficients are determined

$$e_n^{\perp} = f_n^{\perp} = 0, \quad (n \geq 1), \quad (3.31)$$

$$g_1^{\perp} = -U_0 \tau_0 \frac{4e^{2\tau_0} \sum_{k=2}^{\infty} (\Gamma_{k,k+1} - \Gamma_{k,k-1}) + \frac{1}{\sinh \tau_0} - 4 \sinh \tau_0 + \frac{\tanh \tau_0}{\cosh \tau_0}}{1 + 2\tau_0 + e^{4\tau_0}(2\tau_0 - 1)}, \quad (3.32)$$

$$h_1^{\perp} = -g_1^{\perp} \frac{\sinh 2\tau_0}{\tau_0}, \quad (3.33)$$

$$\begin{aligned} g_n^{\perp} = & U_0 \frac{\sinh(n-1)\tau_0}{\sinh 2n\tau_0 - n \sinh 2\tau_0} \left\{ 2 \sum_{\substack{k=2, \\ n-k \geq 1}}^{\infty} \Gamma_{k,n-k} + \sum_{\substack{k=2, \\ k-n \geq 1}}^{\infty} (\Gamma_{k,k+n} - \Gamma_{k,k-n}) \right. \\ & + \frac{\cosh 2\tau_0}{\cosh \tau_0} e^{2(n-1)\tau_0} \coth(n-1)\tau_0 + \frac{\cosh 2\tau_0}{\cosh \tau_0} e^{2(n+1)\tau_0} \coth(n+1)\tau_0 \\ & \left. - 2e^{-n\tau_0} \coth n\tau_0 - \left[\frac{n-1}{\sinh(n-1)\tau_0} - \frac{n+1}{\sinh(n+1)\tau_0} \right] \sinh \tau_0 \right\}, \\ & (n \geq 2), \end{aligned} \quad (3.34)$$

$$h_n^\perp = -g_n^\perp \frac{\sinh(n+1)\tau_0}{\sinh(n-1)\tau_0}, \quad (n \geq 2), \quad (3.35)$$

and

$$e_n^\parallel = f_n^\parallel = 0, \quad (n \geq 1), \quad (3.36)$$

$$g_1^\parallel = U_0 \frac{4\tau_0 \sinh \tau_0}{\sinh 2\tau_0 - 2\tau_0 \cosh 2\tau_0} \times \left[2e^{-2\tau_0} \left(\frac{\tanh 2\tau_0}{\cosh 2\tau_0} - \tanh \tau_0 \right) + \sum_{k=2}^{\infty} (\Gamma_{k,k-1} - \Gamma_{k,k+1}) \right], \quad (3.37)$$

$$h_1^\parallel = -g_1^\parallel \frac{\sinh 2\tau_0}{\tau_0}, \quad (3.38)$$

$$g_n^\parallel = U_0 \frac{\sinh(n-1)\tau_0 \sinh \tau_0}{n \sinh 2\tau_0 - \sinh 2n\tau_0} \left\{ \frac{n-1}{\cosh(n-1)\tau_0} - \frac{2ne^{-\tau_0}}{\cosh n\tau_0} + \frac{n+1}{\cosh(n+1)\tau_0} - \frac{-4e^{-n\tau_0} \sinh^3 \tau_0 (1 + \cosh 2\tau_0 - \sinh 2n\tau_0)}{\cosh 2\tau_0 \cosh(n+1)\tau_0 \cosh(n-1)\tau_0} + \sum_{\substack{k=2, \\ n-k \geq 1}}^{\infty} \Gamma_{k,n-k} + \sum_{\substack{k=2, \\ k-n \geq 1}}^{\infty} (\Gamma_{k,k-n} - \Gamma_{k,k+n}) \right\}, \quad (n \geq 2), \quad (3.39)$$

$$h_n^\parallel = -g_n^\parallel \frac{\sinh(n+1)\tau_0}{\sinh(n-1)\tau_0}, \quad (n \geq 2), \quad (3.40)$$

for the perpendicular and parallel applied electric fields, respectively. The obtained series solutions converge rapidly as the coefficients decay in power-law. The pressure field can be readily obtained since p and $\mu \nabla^2 \psi$ are conjugate functions [116].

3.2.3 Cylinder ICEP velocities

The hydrodynamic force and moment per unit length exerted by the surrounding ICEO flow on the stationary cylinders can be obtained from Equations (3.16) and (3.17), respectively, by replacing the Maxwell stress tensor $\mathbf{\Pi}_e$ with the viscous stress tensor $\mathbf{\Pi}_H$

$$\mathbf{\Pi}_H = -p\mathbf{I} + \mu \left[\nabla \mathbf{u} + (\nabla \mathbf{u})^T \right], \quad (3.41)$$

where p and $\mathbf{u}$ are pressure and velocity of the solution, respectively.

Substituting Equation (3.23) into Equations (3.16) and (3.17) through Equation (3.22) with the obtained coefficients, it is found that the hydrodynamic moment is zero due to the flow symmetry, while the hydrodynamic force per unit length on cylinder-2 for the perpendicular and parallel applied electric fields, respectively, are listed as follows:

$$\begin{aligned} \mathbf{F}_H^\perp &= -4\pi\mu U_0 \frac{4\tau_0 \cosh 2\tau_0 - \sinh 2\tau_0}{2\tau_0 + 1 + e^{4\tau_0}(2\tau_0 - 1)} \\ &\times \left[\frac{1}{\sinh \tau_0} + 4e^{2\tau_0} \sum_{k=2}^{\infty} (\Gamma_{k,k+1}^\perp - \Gamma_{k,k-1}^\perp) - 4 \sinh \tau_0 + \frac{\tanh \tau_0}{\cosh \tau_0} \right] \mathbf{e}_x, \end{aligned} \quad (3.42)$$

and

$$\begin{aligned} \mathbf{F}_H^\parallel &= -16\pi\mu U_0 \frac{e^{-2\tau_0} \sinh \tau_0 (4 \cosh 2\tau_0 - \sinh 2\tau_0)}{2\tau_0 \cosh 2\tau_0 - \sinh 2\tau_0} \\ &\times \left[-2 \tanh \tau_0 + e^{2\tau_0} \sum_{k=2}^{\infty} (\Gamma_{k,k-1}^\parallel - \Gamma_{k,k+1}^\parallel) + \frac{2 \tanh 2\tau_0}{\cosh 2\tau_0} \right] \mathbf{e}_x. \end{aligned} \quad (3.43)$$

When the cylinders solely translate with velocity $\pm U_x$ (cylinder-1 with $-U_x$, cylinder-2 with U_x), the drag force exerted on the cylinders can be determined by applying this condition to Equation (3.23), Equations (3.16) and (3.17). The drag force per unit length exerted on cylinder-2 caused by the translation velocity $\pm U_x$ is obtained as

$$\mathbf{F}_d = -\frac{8\pi\mu U_x}{2\tau_0 - \tanh 2\tau_0} \mathbf{e}_x. \quad (3.44)$$

The cylinder motion is force-free and the electrostatic force is negligible [42]. Thus, the cylinder velocities can be obtained by counteracting the ICEP force (Equation (3.42)/(3.43)) with the drag force (Equation (3.44))

$$\mathbf{F}_H + \mathbf{F}_d = 0. \quad (3.45)$$

Substituting Equations (3.42)/(3.43) and (3.44) into Equation (3.45), the ICEP velocities of cylinder-2 for the perpendicular and parallel applied electric fields are obtained as

$$U_x^\perp = -\frac{1}{2}U_0 \frac{(2\tau_0 - \tanh 2\tau_0)(4\tau_0 \cosh 2\tau_0 - \sinh 2\tau_0)}{2\tau_0 + 1 + e^{4\tau_0}(2\tau_0 - 1)} \times \left[\frac{1}{\sinh \tau_0} + 4e^{2\tau_0} \sum_{k=2}^{\infty} (\Gamma_{k,k-1} - \Gamma_{k,k+1}) - 4 \sinh \tau_0 + \frac{\tanh \tau_0}{\cosh \tau_0} \right], \quad (3.46)$$

and

$$U_x^\parallel = -2U_0 e^{-2\tau_0} \sinh \tau_0 (4\tau_0 - \tanh 2\tau_0) \times \left[\frac{2 \tanh \tau_0}{\cosh 2\tau_0} - 2 \tanh \tau_0 + e^{2\tau_0} \sum_{k=2}^{\infty} (\Gamma_{k,k-1} - \Gamma_{k,k+1}) \right]. \quad (3.47)$$

3.3 Results and Discussion

In the following discussion, the electric, velocity and pressure fields are calculated with the dimensionless half distance between the two cylinders $c/R = 2.0$, where c is the half distance between the cylinder centres and R is the radius of the cylinders as illustrated in Figure 3.1. To facilitate the discussion, two kinds of surfaces are defined: facing surfaces indicate the surfaces of the cylinders facing each other; non-facing surfaces indicate the surfaces of the cylinders facing away from each other. The same meanings apply to facing/non-facing regions.

3.3.1 Electric field

Figure 3.2: Electric field lines (solid curves) and equipotential lines (dashed curves) around the two fully screened stationary cylinders under the (a) perpendicular and (b) parallel applied electric field, respectively.

The electric field lines and the equipotential lines around the two fully screened stationary cylinders under the perpendicular and parallel applied electric fields, respectively, are examined. The electric field lines and equipotential lines are presented in Figure 3.2 by solid and dashed curves, respectively. As mentioned earlier, after the application of the external electric field, the cylinders become polarised immediately and attract counterions in the electrolyte solution to their surfaces. Within an instant moment, the cylinders are fully screened by the ions, forming induced EDLs on the cylinder surfaces with nonlinear zeta potentials. The conducting cylinders with the induced EDLs act as ideal insulators. Different from the case of a single cylinder reported by Squires and Bazant [1], here the symmetry of the electric field around one cylinder is broken due to the presence of the other cylinder. Under the perpendicular applied electric field, as illustrated in Figure 3.2(a), the local electric field in the facing region is enhanced compared to that in the non-facing regions. Hence, the induced zeta potentials on the facing surfaces are relatively stronger than that on the non-facing surfaces. With the decrease

in distance between the two cylinders, the local electric field in the facing region becomes even stronger and gives rise to a more intensified fluid flow in this region.

When the imposed external field is in parallel (Figure 3.2(b)), the electric field is distorted in a different way. The local electric field in the facing region is attenuated; hence, the induced zeta potentials on the facing surfaces are weakened. As c/R decreases, the local electric field in the facing region reduces so that the fluid flow in this region is further weakened. The distortion of electric field due to the presence of the cylinders affects the slip velocity on the cylinder surfaces as well as the fluid flow around the cylinders.

3.3.2 ICEO flow and pressure fields

Figures 3.3 and 3.4 show the ICEO flow and pressure fields around the two stationary cylinders under the perpendicular and parallel applied electric fields, respectively. Resembling to the electric field, the velocity and pressure fields around each cylinder also lose symmetry due to the presence of the other cylinder.

Figure 3.3: (a) Velocity field (streamlines) and (b) pressure field around two conducting stationary cylinders under the perpendicular applied electric field. The blue arrows indicate the flow directions.

The ICEO flow field expressed by streamlines under the perpendicular applied electric field is presented in Figure 3.3(a). The solid and dashed curves indicate

the positive and negative streamlines, representing counterclockwise and clockwise rotations, respectively. We can see that there are four bounded microvortices between the two cylinders. The microvortices are outward rotating, which generate a high pressure zone in the facing region as illustrated in Figure 3.3(b), driving the cylinders to translate away from each other. When the external electric field is applied in parallel, the four microvortices become inward rotating (Figure 3.4(a)), which gives rise to a low pressure zone in this region (Figure 3.4(b)) and causes the two cylinders to attract each other. The four microvortices and the corresponding pressure zone in the facing region are much stronger when the external electric field is imposed in perpendicular direction. That is because the local electric field in this region is stronger under the perpendicular applied electric field (Figure 3.2(a)) but weaker under the parallel applied electric field (Figure 3.2(b)). Therefore, the induced fluid flow in the facing region is also stronger under the perpendicular applied electric field but weaker under the parallel applied electric field, so as the corresponding pressure zone in this region. This leads to a more intensive interaction between the two cylinders under the perpendicular applied electric field while a weaker interaction under the parallel applied electric field. The local electric field at the upper and lower sides of the cylinders is stronger under the parallel applied electric field (Figure 3.2(b)); the velocity and pressure zones in these regions are more intensified as well. The pressure zones at the upper and lower sides of the cylinders are symmetric to the x -axis, while the pressure zones at the facing and non-facing sides of each cylinder are totally different. Thus, the cylinders experience horizontal force but not vertical force. A qualitative agreement is shown between the present results and the numerical simulation on two spherical particles in Ref. [47].

3.3.3 Electrostatic and ICEP forces

Due to the nonuniformity of local electric field around each cylinder caused by the appearance of the other cylinder, the cylinders experience electrostatic (ES) force.

Figure 3.4: (a) Velocity field (streamlines) and (b) pressure field around two conducting stationary cylinders under the parallel applied electric field. The blue arrows indicate the flow direction.

As discussed previously, the ES force is negligible compared to the ICEP force in the present model. To visualise the relative magnitude of ES and ICEP forces, we hereby present the ICEP+ES, ICEP, and ES forces on cylinder-2 under the perpendicular and parallel applied electric fields in Figures 3.5 and 3.6, respectively.

Figure 3.5: (a) Variation of the dimensionless ICEP+ES, ICEP and ES forces $F_x/(\epsilon_w E_\infty^2 R)$ with the dimensionless half distance c/R on cylinder-2 under the perpendicular applied electric field.

Figure 3.5 shows that the ES force reduces significantly as c/R increases and becomes negligible when c/R reaches 2.0. While the ICEP force increases at small c/R and decreases when $c/R > 1.13$. As the ICEP force is much larger than the ES

force, the total force varies with the similar trend of the ICEP force. The ES force under the parallel applied electric field is not only smaller than the ICEP force under this field (Figure 3.6), but also smaller than that under the perpendicular applied electric field (Figure 3.5). Both the ES and ICEP forces diminishes to zero as c/R increases under the parallel applied electric field. The total force are almost zero once c/R reaches 5.0. The ICEP force under the perpendicular applied electric field (Figure 3.5) reduces much slower at larger c/R compared with that under the parallel applied electric field (Figure 3.6).

Figure 3.6: Variation of the dimensionless ICEP+ES, ICEP and ES forces $F_x/(\epsilon_w E_\infty^2 R)$ with the dimensionless half distance c/R on cylinder-2 under the parallel applied electric field.

Figures 3.5 and 3.6 show that the ES force is small compared to the ICEP force and decreases rapidly with the increase of c/R . Thus, the cylinder motion can be accurately described by the ICEP velocities. Both the ES and ICEP forces make the cylinders repel each other under the perpendicular electric field, while attract each other under the parallel electric field. Similar conclusions are obtained for the ICEP of two spherical particles [46] and a conducting cylinder near a wall [42]. However, one should note that when the applied electric field is nonuniform or the particles are not highly polarisable, such as dielectric particles, the ES force could be of same magnitude or larger than the ICEP force [5].

3.3.4 ICEP velocities

The variations of the ICEP velocity U_x/U_0 of cylinder-2 with c/R under the perpendicular and parallel applied electric fields are presented in Figures 3.7 and 3.8, respectively. Not surprisingly, the cylinders translate due to the asymmetry of the electric, velocity and pressure fields. This behaviour is understandable as the local electric field around one cylinder is disturbed by the presence of the other cylinder. The local electric fields on the right-hand and left-hand sides of each cylinder are no longer identical. As a result, the corresponding velocity and pressure fields become asymmetric. This unbalanced pressure field drives the cylinders into motion. At small c/R , the magnitude of U_x/U_0 increases rapidly when c/R increases; while at large c/R , U_x/U_0 decreases to zero as c/R increases. The turning points occur at $c/R = 2.30$ and 1.36 with the maximum magnitudes of the velocities ± 3.08 and ∓ 0.42 for the perpendicular and parallel applied electric fields, respectively. After crossing these peak values, the velocities show power-law decays in general. As c/R increases, the interactions between the two cylinders diminish. When c/R becomes very large ($c/R = 10$ and 500 for the perpendicular and parallel applied electric fields, respectively), the disturbance of the local electric field around one cylinder caused by the presence of the other cylinder becomes negligible. The electric, velocity and pressure fields around each cylinder regain their symmetries. Thus, the two cylinders no longer interact with each other and remain stationary as reported by Squires and Bazant [1].

The reason why U_x/U_0 varies with such trend is that there are two factors influencing U_x/U_0 as c/R changes: the electric field asymmetry around each cylinder and the viscous effect between the two cylinders. The competition of these two effects determines the variation trend of the ICEP velocities U_x/U_0 . When c/R is smaller than the critical values ($c/R = 2.30$ and 1.36 for perpendicular and parallel applied electric fields, respectively), U_x/U_0 increases as c/R increases. That is because the viscous effect reduces more significantly than the asymmetry

Figure 3.7: Variation of dimensionless ICEP velocity U_x/U_0 of cylinder-2 with the dimensionless half distance c/R under the perpendicular applied electric field.

of electric field in these ranges of c/R . Once c/R crosses the critical values, the attenuation of electric field asymmetry shows more significant effect on U_x/U_0 and causes it to decrease as c/R increases. However, one should note that when c/R is very small, the EDLs on the two cylinders may interact with each other. In this situation, other factors have to be taken into consideration, like van der Waals force, EDL interaction or overlapping, etc. This is beyond the scope of the present model.

Figure 3.8: Variation of dimensionless ICEP velocity U_x/U_0 of cylinder-2 with the dimensionless half distance c/R under the parallel applied electric field.

The direction of the applied electric field also influences the hydrodynamic interactions between the two cylinders. As discussed previously, when the external electric field is imposed in the perpendicular direction (Figure 3.7), the two cylinders repel each other; when the external electric field is applied in parallel, the two cylinders attract each other (Figure 3.8). The possible reason for these different behaviours of the cylinders lies in the specific ways that the external electric field gets distorted by the cylinders. When the external electric field is applied in the perpendicular direction, the local electric field in the facing region is stronger compared to that in the non-facing regions (Figure 3.2(a)). Thus, the four outward rotating microvortices in the facing region is stronger (Figure 3.3(a)), which leads to larger ICEP velocities of the cylinders. While under the parallel applied electric field, the local electric field in the facing region is weaker (Figure 3.2(b)). Hence, the four inward rotating microvortices are weak as well (Figure 3.4(a)), so as the attraction effect between the two cylinders. Therefore, the ICEP velocities of the cylinders are much smaller and approach zero more rapidly under the parallel applied electric field.

The numerical simulation on two spherical particles in Ref. [47] reveals a similar trend: the particles repel and attract each other under the perpendicular and parallel electric fields, respectively; and the particle interactions under the parallel electric field are weaker than that under the perpendicular electric field.

The previous discussion is carried out in dimensionless form. However, the parameters are dimensional in reality. To ensure the thin EDL approximation holds, the EDL thickness λ_D should be much smaller than the cylinder radius R and the distance between the cylinders $2c$, $\lambda_D \ll \min(R, 2c)$. $\lambda_D = \Lambda/\sqrt{c_0}$, where

$$\Lambda = \sqrt{\frac{\varepsilon_f k_B N_A T_a}{2F_a^2}} \quad (3.48)$$

where ε_f is the dielectric permittivity of the electrolyte solution; k_B is the Boltzmann constant; N_A is the Avogadro constant; T_a is the absolute temperature

of the electrolyte solution; F_a is the Faraday constant; and c_0 is the electrolyte concentration. $\Lambda \approx 10 \text{ mol}^{1/2}/\text{m}^{1/2}$ at room temperature [33]. The weak electric field approximation requires that the induced zeta potential of the cylinders ζ_i is smaller than the thermal voltage ϕ_T , $\zeta_i < \phi_T$, where ϕ_T is around 25 mV at room temperature. ζ_i is of order $2E_0R$ (Equations (3.12) and (3.13)). Thus, $2E_0R < \phi_T$. The ranges of c_0 , E_0 and R are interrelated due to the criteria of the thin EDL and weak electric field approximations. In practical applications, c_0 ranges from $1 \times 10^{-6} \text{ mol/L}$ to $1 \times 10^{-3} \text{ mol/L}$, which leads λ_D ranging from 300 nm to 10 nm. It therefore requires that $\min(R, 2c)$ should be larger than $3 \mu\text{m}$ to 100 nm corresponding to the relevant c_0 . Assuming $\min(R, 2c)=R$, E_0 should be smaller than $4.3 \times 10^3 \text{ V/m}$ to $1.3 \times 10^5 \text{ V/m}$ corresponding to the relevant c_0 . When $\min(R, 2c)=2c$, the upper limit of E_0 reduces.

3.4 Summary

The present analytical model characterises the electrostatic and hydrodynamic interactions of two infinite long conducting cylinders under the uniform external electric field with thin EDL and weak external electric field approximations. The results reveal that microvortices are generated on the cylinders surfaces: four bounded microvortices in the facing region and four unbounded microvortices in the non-facing regions. Furthermore, the two cylinders are driven into motion due to the nonuniformity of the electric field and the asymmetry of fluid flow around each cylinder. The electrostatic force is much smaller than the ICEP force for both the perpendicular and parallel applied electric fields. The cylinder motion can be accurately described by the ICEP velocities: repelling and attracting each other under the perpendicular and parallel applied electric fields, respectively. Moreover, the ICEP velocities are much smaller and approach zero more rapidly under the parallel applied electric field compared to those under the perpendicular applied electric field.

Chapter 4

Pair Interactions between Conducting and Non-conducting Cylinders in Uniform Electric Fields

4.1 Introduction

As introduced in Section 3.1, electrophoresis (EP) has been extensively studied due to its great potential in particle manipulations. Many theoretical models have been developed [33]. More recently, various types of conducting particles are studied due to the advancement in nanotechnology [111, 112, 117, 118, 119, 120]. Thus, it becomes critically important to understand the behaviour of conducting particles in electric fields, i.e., ICEP motion of particles. A number of studies have been carried out on ICEP [2] and many promising application ideas based on ICEP of different particles have been discussed, such as micromixers [10], microvalves [6, 41], and micromotors [5, 54]. ICEP interactions between particles [39, 47, 121], and between wall and particles [3, 42, 45, 48] with particles of different shapes have been characterised. Gangwal et al. [49] experimentally observed the ICEP of Janus particles in microchannels. Boymelgreen et al. [54] captured the ICEP rotation of Janus doublets lately, which shows the potential application to motion controller and electric field detection.

However, all these studies focus on the interactions between particles of same property, i.e. both conducting or both non-conducting. How the particles of different properties behave in electric fields remains unclear. Therefore, pair

interactions between a conducting cylinder and a non-conducting cylinder are investigated in this chapter. The governing equations with appropriate boundary conditions are analytically solved in the bipolar coordinates. The study captures the microvortices and net fluid flow around the two stationary cylinders, which can be utilised for mixing and pumping in micro/nanofluidics. The cylinder velocities are also obtained and analysed, which contributes to a better understanding of particle behaviour in colloid science and micro/nanofluidics.

4.2 Mathematical Formulation

4.2.1 Problem statement

Two infinitely long (two-dimensional) cylinders, which are freely suspended in the unbounded electrolyte solution, are subjected to a uniform electric field. A Cartesian coordinate system with the cylinder centres located on the x -axis is introduced. The left and the right cylinders are non-conducting (non-polarisable) and conducting (ideally polarisable), respectively. For the convenience of the following discussion, the two cylinders are labelled as cylinder-1 and cylinder-2, respectively, as illustrated in Figure 4.1. A bipolar coordinate system (τ, σ) is used to describe the two cylinders. The surfaces of cylinder-1 and cylinder-2 are represented by $\tau = -\tau_0$ and $\tau = \tau_0$, respectively.

4.2.2 Electric field

The electric field is applied along two directions, perpendicular and parallel to the connection line of the two cylinder centres, i.e., along the y -axis ($E_\infty \mathbf{e}_y$) and the x -axis ($E_\infty \mathbf{e}_x$), respectively. The thickness of the EDLs formed on the two cylinders is commonly small ($\sim \text{nm}$) [1], thus, the thin EDL approximation is adopted. The bulk solution outside the EDLs remains electrical neutrality. Hence,

Figure 4.1: Schematic diagram of the two cylinders suspended in the unbounded electrolyte solution. The surfaces of the two cylinders are represented by $\tau = \pm\tau_0$; $R = a/\sinh \tau_0$ is the radius of the cylinders. Cylinder-1 and cylinder-2 are non-conducting and conducting, respectively.

the Laplace equation is applied

$$\nabla^2 \phi = 0, \quad (4.1)$$

The electric field lines are expelled by the EDLs on the cylinders, hence, the no-flux condition is applied

$$\mathbf{e}_\tau \cdot \nabla \phi = 0 \quad \text{at} \quad \tau = \pm\tau_0. \quad (4.2)$$

The distortion of the electric field due to the presence of the two cylinders disappears at far-field, thus

$$\phi \rightarrow -E_\infty y \quad \text{as} \quad r \rightarrow \infty, \quad (4.3)$$

and

$$\phi \rightarrow -E_\infty x \quad \text{as} \quad r \rightarrow \infty, \quad (4.4)$$

for the perpendicular and the parallel applied electric fields, respectively.

Solving Equation (4.1) together with Equations (4.2) and (4.3)/ (4.4) in the bipolar coordinates, the electrical potentials under the perpendicular and parallel

applied electric fields are obtained

$$\phi^\perp = -E_\infty a \frac{\sin \sigma}{\cosh \tau - \cos \sigma} - 2E_\infty a \sum_{n=1}^{\infty} \frac{e^{-n\tau_0}}{\sinh n\tau_0} \cosh n\tau \sin n\sigma, \quad (4.5)$$

and

$$\phi^\parallel = -E_\infty a \frac{\sinh \tau}{\cosh \tau - \cos \sigma} - 2E_\infty a \sum_{n=1}^{\infty} \frac{e^{-n\tau_0}}{\cosh n\tau_0} \sinh n\tau \cos n\sigma. \quad (4.6)$$

The corresponding electric field functions under the perpendicular and the parallel applied electric fields are obtained through the following equations [108]

$$\frac{\partial \phi}{\partial \tau} = -\frac{\partial V}{\partial \sigma}, \quad \frac{\partial \phi}{\partial \sigma} = \frac{\partial V}{\partial \tau}, \quad (4.7)$$

given as

$$\begin{aligned} V^\perp = & E_\infty a \frac{\sinh \tau}{\cosh \tau - \cos \sigma} \\ & - 2E_\infty a \sum_{n=1}^{\infty} \frac{e^{-n\tau_0}}{\sinh n\tau_0} \sinh n\tau \cos n\sigma, \end{aligned} \quad (4.8)$$

and

$$\begin{aligned} V^\parallel = & -E_\infty a \frac{\sin \sigma}{\cosh \tau - \cos \sigma} \\ & + 2E_\infty a \sum_{n=1}^{\infty} \frac{e^{-n\tau_0}}{\cosh n\tau_0} \cosh n\tau \sin n\sigma, \end{aligned} \quad (4.9)$$

respectively.

The zeta potential ζ_c of cylinder-1 (non-conducting) is a constant value, while the zeta potential of cylinder-2 (conducting) is induced by the applied electric field and can be obtained through

$$\zeta_i = -\phi + \frac{\int_A \phi dA}{A}, \quad (4.10)$$

where ϕ is the external electrical potential at cylinder-fluid interface [32].

The induced zeta potential of cylinder-2 under the perpendicular and parallel applied electric fields are obtained by substituting Equation (4.5)/(4.6) into Equation (4.10), given as

$$\begin{aligned}\zeta_i^\perp &= E_\infty a \frac{\sin \sigma}{\cosh \tau_0 - \cos \sigma} \\ &\quad + 2E_\infty a \sum_{n=1}^{\infty} e^{-n\tau_0} \coth n\tau_0 \sin n\sigma,\end{aligned}\tag{4.11}$$

and

$$\begin{aligned}\zeta_i^\parallel &= E_\infty a \frac{\sinh \tau_0}{\cosh \tau_0 - \cos \sigma} \\ &\quad + 2E_\infty a \sum_{n=1}^{\infty} e^{-n\tau_0} \tanh n\tau_0 \cos n\sigma - E_\infty a,\end{aligned}\tag{4.12}$$

respectively.

4.2.3 Flow field

Generally, the Reynolds number is very small in EO and ICEO [1] and the fluid is incompressible. Such a flow satisfies the form of Stokes equations in terms of stream function ψ

$$\nabla^4 \psi = 0,\tag{4.13}$$

with the velocities in the bipolar coordinates defined as

$$u_\sigma = h \frac{\partial \psi}{\partial \tau}, \quad u_\tau = -h \frac{\partial \psi}{\partial \sigma},\tag{4.14}$$

where $h = (\cosh \tau - \cos \sigma)/a$.

The general solution of the stream function ψ in the bipolar coordinates was

given by Jeffery [116]

$$\begin{aligned}
h\psi = & A_0 \cosh \tau + B_0 \tau (\cosh \tau - \cos \sigma) + C_0 \sinh \tau + D_0 \tau \sinh \tau \\
& + \sum_{n=1}^{\infty} [a_n \cosh(n+1)\tau + b_n \cosh(n-1)\tau + c_n \sinh(n+1)\tau \\
& + d_n \sinh(n-1)\tau] \cos n\sigma + h_1 \tau \sin \sigma + \sum_{n=1}^{\infty} [e_n \cosh(n+1)\tau \\
& + f_n \cosh(n-1)\tau + g_n \sinh(n+1)\tau + h_n \sinh(n-1)\tau] \sin n\sigma. \quad (4.15)
\end{aligned}$$

The electric field exerts an electrical force on the ions within the EDLs, which drives the ions into motion. Hence, a flow is aroused around the two cylinders correspondingly. Under the approximations of thin EDL and weak electric field, the slip velocities on the cylinders can be described by the Helmholtz-Smoluchowski formula

$$\mathbf{u}_s = -\frac{\varepsilon_f \zeta}{\mu} E_\sigma \mathbf{e}_\sigma \quad \text{at} \quad \tau = \pm \tau_0, \quad (4.16)$$

Substituting the tangential electric field (obtained from Equation (4.5)/(4.6) through $\mathbf{E} = -\nabla\phi$) and the zeta potentials (ζ_c and ζ_i for cylinder-1 and cylinder-2, respectively) on the cylinders into Equation (4.16), the slip velocities on the cylinders are obtained. The fluid stays quiescent at far-field

$$\mathbf{u} \rightarrow 0 \quad \text{as} \quad r \rightarrow \infty. \quad (4.17)$$

Substituting Equation (4.15) into Equation (4.16) and (4.17) through Equation (4.14), all the unknown coefficients are determined.

Under the perpendicular applied electric field

$$\begin{aligned}
A_0^\perp = & \frac{1}{64} \tilde{\zeta}_c U_0 [128 \tau_0 \sinh^3 \tau_0 \cosh \tau_0 + \cosh 6\tau_0 + 4 + (16\tau_0^2 - 1) \cosh 2\tau_0 \\
& - 4 \cosh 4\tau_0] / [\sinh^3 \tau_0 \cosh \tau_0 (2\tau_0 + \sinh 2\tau_0)], \quad (4.18)
\end{aligned}$$

$$B_0^\perp = \frac{1}{8} \frac{\tilde{\zeta}_c U_0 \cosh 2\tau_0}{\sinh^3 \tau_0 \cosh \tau_0}, \quad (4.19)$$

$$C_0^\perp = -\frac{1}{8} \frac{\tilde{\zeta}_c U_0 (\cosh 2\tau_0 - 2) \coth \tau_0}{\sinh^2 \tau_0}, \quad (4.20)$$

$$D_0^\perp = -\frac{\tilde{\zeta}_c U_0 \coth \tau_0}{2\tau_0 + \sinh 2\tau_0}, \quad (4.21)$$

$$a_1^\perp = \frac{1}{16} \frac{\tilde{\zeta}_c U_0 \coth 2\tau_0}{\sinh 2\tau_0}, \quad (4.22)$$

$$b_1^\perp = -\frac{1}{32} \tilde{\zeta}_c U_0 \frac{2\tau_0 (\cosh \tau_0 + \cosh 3\tau_0) - \sinh \tau_0 - 2 \sinh 3\tau_0 + \sinh 5\tau_0}{\sinh^3 \tau_0 \cosh^2 \tau_0}, \quad (4.23)$$

$$c_1^\perp = -\frac{1}{16} \frac{\tilde{\zeta}_c U_0 (\cosh 2\tau_0 - 2)}{\sinh^3 \tau_0 \cosh \tau_0}, \quad (4.24)$$

$$a_n^\perp = -\frac{1}{2} \frac{K_n^\perp \cosh(n-1)\tau_0}{n \sinh 2\tau_0 + \sinh 2n\tau_0}, \quad (4.25)$$

$$b_n^\perp = \frac{1}{2} \frac{K_n^\perp \cosh(n+1)\tau_0}{n \sinh 2\tau_0 + \sinh 2n\tau_0}, \quad (4.26)$$

$$c_n^\perp = -\frac{1}{2} \frac{K_n^\perp \sinh(n-1)\tau_0}{n \sinh 2\tau_0 - \sinh 2n\tau_0}, \quad (4.27)$$

$$d_n^\perp = \frac{1}{2} \frac{K_n^\perp \sinh(n+1)\tau_0}{n \sinh 2\tau_0 - \sinh 2n\tau_0}, \quad (4.28)$$

where $2 \leq n \leq N - 1$.

$$a_N^\perp = -\frac{1}{4} \frac{K_n^\perp + 2(n-1)[A_0^\perp + \sum_{n=1}^{N-1}(a_n^\perp + b_n^\perp)] \sinh(n-1)\tau_0}{n \cosh n\tau_0 \sinh \tau_0 + \cosh \tau_0 \sinh n\tau_0}, \quad (4.29)$$

$$b_N^\perp = \frac{1}{4} \frac{K_n^\perp + 2(n+1)[A_0^\perp + \sum_{n=1}^{N-1}(a_n^\perp + b_n^\perp)] \sinh(n+1)\tau_0}{n \cosh n\tau_0 \sinh \tau_0 + \cosh \tau_0 \sinh n\tau_0}, \quad (4.30)$$

$$\begin{aligned} c_N^\perp = & -\frac{1}{4} \left\{ K_n^\perp (\cosh 2n\tau_0 - n \cosh 2\tau_0 + n - 1) \right. \\ & + (n-1)[A_0^\perp + \sum_{n=1}^{N-1}(a_n^\perp + b_n^\perp)] \\ & \times [(n+1) \sinh(n+1)\tau_0 - n \sinh(n-3)\tau_0 \\ & \left. + \sinh(3n-1)\tau_0] \right\} \\ & / [(n \cosh n\tau_0 \sinh \tau_0 + \cosh \tau_0 \sinh n\tau_0) \\ & \times (n \sinh 2\tau_0 - \sinh 2n\tau_0)], \end{aligned} \quad (4.31)$$

$$\begin{aligned} d_N^\perp = & \frac{1}{4} \left\{ K_n^\perp (\cosh 2n\tau_0 + n \cosh 2\tau_0 - n - 1) \right. \\ & + (n+1)[A_0^\perp + \sum_{n=1}^{N-1}(a_n^\perp + b_n^\perp)] \\ & \times [(n-1) \sinh(n-1)\tau_0 - n \sinh(n+3)\tau_0 \\ & \left. + \sinh(3n+1)\tau_0] \right\} \\ & / [(n \cosh n\tau_0 \sinh \tau_0 + \cosh \tau_0 \sinh n\tau_0) \\ & \times (n \sinh 2\tau_0 - \sinh 2n\tau_0)], \end{aligned} \quad (4.32)$$

where

$$K_n^\perp = \tilde{\zeta}_c U_0 \left\{ \frac{n-1}{\sinh(n-1)\tau_0} - \frac{2n \cosh \tau_0}{\sinh 2\tau_0} + \frac{n+1}{\sinh(n+1)\tau_0} \right\}, \quad (4.33)$$

and

$$e_1^\perp = \frac{1}{16} \frac{\Lambda_1^\perp}{\sinh 2\tau_0}, \quad (4.34)$$

$$f_1^\perp = -\frac{1}{4} \Lambda_1^\perp \coth 2\tau_0, \quad (4.35)$$

$$g_1^\perp = \frac{1}{2} \frac{\Lambda_1^\perp \tau_0}{2\tau_0 \cosh 2\tau_0 - \sinh 2\tau_0}, \quad (4.36)$$

$$h_1^\perp = \frac{1}{2} \frac{\Lambda_1^\perp}{1 - 2\tau_0 \coth 2\tau_0}, \quad (4.37)$$

$$e_n^\perp = \frac{1}{2} \frac{\Lambda_n^\perp \cosh(n-1)\tau_0}{n \sinh 2\tau_0 + \sinh 2n\tau_0}, \quad (4.38)$$

$$f_n^\perp = -\frac{1}{2} \frac{\Lambda_n^\perp \cosh(n+1)\tau_0}{n \sinh 2\tau_0 + \sinh 2n\tau_0}, \quad (4.39)$$

$$g_n^\perp = -\frac{1}{2} \frac{\Lambda_n^\perp \sinh(n-1)\tau_0}{n \sinh 2\tau_0 - \sinh 2n\tau_0}, \quad (4.40)$$

$$h_n^\perp = \frac{1}{2} \frac{\Lambda_n^\perp \sinh(n+1)\tau_0}{n \sinh 2\tau_0 - \sinh 2n\tau_0}, \quad (4.41)$$

where $2 \leq n \leq N$ and

$$\begin{aligned} \Lambda_1^\perp = & -\frac{1}{2} U_0 \left(6 \cosh \tau_0 - 2 \cosh 3\tau_0 - \frac{5}{\sinh \tau_0} + 8 \sinh^3 \tau_0 + \frac{\tanh \tau_0 - 2}{\cosh \tau_0} \right) \\ & - 2U_0 \sum_{k=2}^{\infty} (\Gamma_{k,k+1}^\perp - \Gamma_{k,k-1}^\perp), \end{aligned} \quad (4.42)$$

$$\begin{aligned}
 \Lambda_n^\perp = & -U_0 \left\{ \frac{(n-1) \sinh \tau_0}{\sinh(n-1)\tau_0} - \frac{(n+1) \sinh \tau_0}{\sinh(n+1)\tau_0} \right. \\
 & - 2e^{-n\tau_0} \coth n\tau_0 + \frac{\cosh 2\tau_0}{\cosh \tau_0} e^{-(n-1)\tau_0} \coth(n-1)\tau_0 \\
 & \left. + \frac{\cosh 2\tau_0}{\cosh \tau_0} e^{-(n+1)\tau_0} \coth(n+1)\tau_0 \right\} \\
 & - 2U_0 \left[\sum_{\substack{k=2, \\ n-k \geq 1}}^{\infty} \Gamma_{k,n-k}^\perp + \sum_{\substack{k=2, \\ k-n \geq 1}}^{\infty} (\Gamma_{k,k+n}^\perp - \Gamma_{k,k-n}^\perp) \right], \quad (4.43)
 \end{aligned}$$

where

$$\Gamma_{k,l}^\perp = \frac{\sinh^2 \tau_0 (\sinh 2k\tau_0 - k \sinh 2\tau_0)}{\sinh k\tau_0 (\cosh 2k\tau_0 - k \cosh 2\tau_0)} e^{-l\tau_0} \coth l\tau_0, \quad (k \geq 2, l \geq 1), \quad (4.44)$$

and $\tilde{\zeta}_c$ and U_0 are the dimensionless zeta potential of cylinder-1 and the natural velocity scale of the ICEO, respectively. Given as

$$\tilde{\zeta}_c = \frac{\zeta_c}{E_\infty R}, \quad (4.45)$$

$$U_0 = \frac{\varepsilon_f E_\infty^2 R}{\mu}. \quad (4.46)$$

Under the parallel applied electric field

$$e_1^\parallel = \frac{1}{8} \frac{\Lambda_1^\parallel \cosh 2\tau_0 + 2\tilde{\zeta}_c U_0 (\cosh 2\tau_0 - 1)}{\sinh 4\tau_0}, \quad (4.47)$$

$$f_1^\parallel = \frac{1}{4} \left[\frac{2\tilde{\zeta}_c U_0 (1 - \cosh 2\tau_0)}{\sinh 2\tau_0} - \Lambda_1^\parallel \coth 2\tau_0 \right], \quad (4.48)$$

$$g_1^\parallel = \frac{1}{2} \frac{\tau_0 [\Lambda_1^\parallel \cosh 2\tau_0 + 2\tilde{\zeta}_c U_0 (1 - \cosh 2\tau_0)]}{\cosh 2\tau_0 (2\tau_0 \cosh 2\tau_0 - \sinh 2\tau_0)}, \quad (4.49)$$

$$h_1^{\parallel} = \frac{1}{2} \frac{\Lambda_1^{\parallel} + 2\tilde{\zeta}_c U_0 (1 - \cosh 2\tau_0)}{\cosh 2\tau_0 (1 - 2\tau_0 \coth 2\tau_0)}, \quad (4.50)$$

$$e_n^{\parallel} = \frac{1}{2} \frac{(\Lambda_n^{\parallel} - K_n^{\parallel}) \cosh(n-1)\tau_0}{n \sinh 2\tau_0 + \sinh 2n\tau_0}, \quad (4.51)$$

$$f_n^{\parallel} = -\frac{1}{2} \frac{(\Lambda_n^{\parallel} - K_n^{\parallel}) \cosh(n+1)\tau_0}{n \sinh 2\tau_0 + \sinh 2n\tau_0}, \quad (4.52)$$

$$g_n^{\parallel} = -\frac{1}{2} \frac{(\Lambda_n^{\parallel} + K_n^{\parallel}) \sinh(n-1)\tau_0}{n \sinh 2\tau_0 - \sinh 2n\tau_0}, \quad (4.53)$$

$$h_n^{\parallel} = \frac{1}{2} \frac{(\Lambda_n^{\parallel} + K_n^{\parallel}) \sinh(n+1)\tau_0}{n \sinh 2\tau_0 - \sinh 2n\tau_0}, \quad (4.54)$$

where $2 \leq n \leq N$ and

$$\Lambda_1^{\parallel} = U_0 \sinh \tau_0 \left[2e^{-2\tau_0} \left(\frac{\tanh 2\tau_0}{\cosh 2\tau_0} - \tanh 2\tau_0 \right) + \sum_{k=2}^{\infty} \left(\Gamma_{k,k-1}^{\parallel} - \Gamma_{k,k+1}^{\parallel} \right) \right], \quad (4.55)$$

$$\begin{aligned} \Lambda_n^{\parallel} = & U_0 \left[\frac{n-1}{\cosh(n-1)\tau_0} + \frac{n+1}{\cosh(n+1)\tau_0} - \frac{2ne^{-\tau_0}}{\cosh n\tau_0} \right. \\ & \left. - \frac{4e^{-n\tau_0} \sinh^3 \tau_0 (1 + \cosh 2\tau_0 - \sinh 2n\tau_0)}{\cosh 2\tau_0 \cosh(n+1)\tau_0 \cosh(n-1)\tau_0} \right] \\ & + U_0 \left[\sum_{\substack{k=2, \\ k-n \geq 1}}^{\infty} (\Gamma_{k,k-n}^{\parallel} - \Gamma_{k,k+n}^{\parallel}) + \sum_{\substack{k=2, \\ n-k \geq 1}}^{\infty} \Gamma_{k,n-k}^{\parallel} \right], \end{aligned} \quad (4.56)$$

$$K_n^{\parallel} = \tilde{\zeta}_c U_0 \left[\frac{2n \cosh \tau_0}{\cosh n\tau_0} - \frac{n-1}{\cosh(n-1)\tau_0} - \frac{n+1}{\cosh(n+1)\tau_0} \right], \quad (4.57)$$

where

$$\Gamma_{k,l}^{\parallel} = \left[\frac{2k \cosh \tau_0}{\cosh k \tau_0} - \frac{k-1}{\cosh(k-1)\tau_0} - \frac{k+1}{\cosh(k+1)\tau_0} \right] e^{-l\tau_0} \tanh l\tau_0, \quad (k \geq 2, l \geq 1). \quad (4.58)$$

4.2.4 Cylinder motion

The following analysis solely considers the forces and moment on cylinder-2. The corresponding forces and moment on cylinder-1 have the same magnitudes but opposite directions.

The local electric field around one cylinder is disturbed by the presence of the other cylinder. Thus, the cylinders may experience electrostatic force and moment, which can be obtained through the following equations [46]

$$\mathbf{F}_e = \int_A \boldsymbol{\Pi}_e \cdot \mathbf{e}_n dA, \quad (4.59)$$

$$\mathbf{M}_e = R \int_A \mathbf{e}_n \times (\boldsymbol{\Pi}_e \cdot \mathbf{e}_n) dA, \quad (4.60)$$

where $\mathbf{e}_n$ is the unit vector normal to the surface; $\boldsymbol{\Pi}_e$ is the Maxwell stress tensor

$$\boldsymbol{\Pi}_e = \varepsilon_f (\mathbf{E}\mathbf{E} - \frac{1}{2} E^2 \mathbf{I}). \quad (4.61)$$

which is normal on the cylinders, $\boldsymbol{\Pi}_e \cdot \mathbf{e}_n = -\varepsilon_f E_t^2 \mathbf{e}_n / 2$. Thus, the electrostatic moment $\mathbf{M}_e = 0$. As the cylinders are fully screened by the EDLs, only this tangential component exists. Substituting Equation (4.5)/(4.6) into Equation (4.59) through $\mathbf{E} = -\nabla\phi$, the electrostatic forces per unit length on cylinder-2

under the perpendicular and parallel applied electric fields are obtained

$$\begin{aligned}
 F_{e,x}^{\perp} = & \pi \varepsilon_f E_{\infty}^2 R \left\{ e^{-3\tau_0} \coth \tau_0 (2 \cosh 2\tau_0 + \sinh 2\tau_0 - 1) \right. \\
 & - \sum_{n=2}^{\infty} n e^{-(2n+1)\tau_0} \coth n\tau_0 \sinh \tau_0 [e^{2\tau_0} (n-1) \cosh \tau_0 \coth (n-1)\tau_0 \\
 & + (n+1) \coth (n+1)\tau_0 \cosh \tau_0 - 2e^{\tau_0} (n + n \coth n\tau_0 + \sinh 2\tau_0) \\
 & \left. + 2ne^{\tau_0} \cosh 2\tau_0] \right\}, \quad (4.62)
 \end{aligned}$$

and

$$\begin{aligned}
 F_{e,x}^{\parallel} = & -\pi \varepsilon_f E_{\infty}^2 R \left\{ \frac{e^{-3\tau_0} \sinh \tau_0}{\cosh 2\tau_0} (\sinh \tau_0 - 2 \cosh 3\tau_0 - \sinh 3\tau_0) \tanh^2 \tau_0 \right. \\
 & + \sum_{n=2}^{\infty} n e^{-(2n+1)\tau_0} \sinh \tau_0 \tanh n\tau_0 [2ne^{\tau_0} (2 \sinh^2 \tau_0 - \tanh n\tau_0) \\
 & - 4e^{\tau_0} \sinh \tau_0 + \cosh \tau_0 (e^{2\tau_0} (n-1) \tanh (n-1)\tau_0 \\
 & \left. + (n+1) \tanh (n+1)\tau_0)] \right\}. \quad (4.63)
 \end{aligned}$$

The electrostatic forces are along the x -axis, which is easy to understand since the electric fields are symmetric to the x -axis as shown in Figure 4.2.

The induced flow around the cylinders may exert hydrodynamic force and/or moment on the cylinders, which can be obtained by substituting Equation (4.15) into Equations (4.59) and (4.60) by replacing Equation (4.61) with the viscous stress tensor

$$\mathbf{\Pi}_H = -p\mathbf{I} + \mu \left[\nabla \mathbf{u} + (\nabla \mathbf{u})^T \right]. \quad (4.64)$$

The obtained hydrodynamic forces and moment per unite length on cylinder-2 under the perpendicular and parallel applied electric fields are given as

$$F_{H,x}^{\perp} = 2\pi\mu U_0 K_1^{\perp} \left(4 - \frac{1}{1 - 2\tau_0 \coth 2\tau_0} \right), \quad (4.65)$$

$$F_{H,y}^{\perp} = -4\pi\mu \tilde{\zeta}_c U_0 \frac{\coth \tau_0}{2\tau_0 + \sinh 2\tau_0}, \quad (4.66)$$

$$M_H^\perp = -4\pi\mu\tilde{\zeta}_c U_0 R \frac{\coth \tau_0 \cosh \tau_0}{2\tau_0 + \sinh 2\tau_0}, \quad (4.67)$$

and

$$F_{H,x}^\parallel = \pi\mu U_0 \frac{K_1^\parallel [8\tau_0(1 + \cosh 4\tau_0) - 3 \sinh 4\tau_0] - 16\tilde{\zeta}_c \cosh \tau_0 \sinh^3 \tau_0}{\cosh^2 2\tau_0 (2\tau_0 - \tanh 2\tau_0)}, \quad (4.68)$$

$$F_{H,y}^\parallel = 0, \quad (4.69)$$

$$M_H^\parallel = 0, \quad (4.70)$$

respectively. $K_1^\perp$ in Equation (4.65) and $K_1^\parallel$ in Equation (4.68) are the coefficients of $\sin \sigma$ to the slip boundary conditions on cylinder-2 under the perpendicular and parallel applied electric fields, respectively.

When the two cylinders conduct creeping motion with velocities of same magnitudes but opposite directions, they experience drag forces and moment. Substituting Equation (4.15) into the boundary conditions, and Equations (4.59) and (4.60), the drag forces and moment per unit length exerted on cylinder-2 due to their motions are obtained

$$F_{d,x} = -\frac{8\pi\mu U_x}{2\tau_0 - \tanh 2\tau_0}, \quad (4.71)$$

$$F_{d,y} = -\frac{8\pi\mu U_y}{2\tau_0 + \sinh 2\tau_0}, \quad (4.72)$$

$$M_d = 8\pi\mu \left(\frac{U_y \cosh \tau_0}{2\tau_0 + \sinh 2\tau_0} + \frac{\Omega R \sinh^2 \tau_0}{2\tau_0 - \sinh 2\tau_0} \right). \quad (4.73)$$

The drag forces and moment on cylinder-1 are of same magnitudes but opposite directions.

As the two cylinders are in free suspension, the net forces and moment exerted

on them should vanish

$$F_{e,x} + F_{H,x} + F_{d,x} = 0, \quad (4.74)$$

$$F_{e,y} + F_{H,y} + F_{d,y} = 0, \quad (4.75)$$

$$M_e + M_H + M_d = 0. \quad (4.76)$$

Substituting Equations (4.62)/(4.63), (4.65)/(4.68), (4.71), Equations (4.66)/(4.69), (4.72) and Equations (4.67)/(4.70), (4.73) into Equations (4.74), (4.75) and (4.76), respectively, the velocities of cylinder-2 under the perpendicular and parallel applied electric fields are obtained

$$\begin{aligned} U_x^\perp &= \frac{1}{4}U_0K_1(8\tau_0 - 3\tanh 2\tau_0) + \frac{1}{8}U_0(2\tau_0 - \tanh 2\tau_0) \\ &\times \left\{ e^{-3\tau_0} \coth \tau_0 (2 \cosh 2\tau_0 + \sinh 2\tau_0 - 1) \right. \\ &- \sum_{n=2}^{\infty} n e^{-(2n+1)\tau_0} \coth n\tau_0 \sinh \tau_0 [2n e^{\tau_0} \cosh 2\tau_0 \\ &+ e^{2\tau_0} (n-1) \coth (n-1)\tau_0 \cosh \tau_0 + (n+1) \cosh \tau_0 \coth (n+1)\tau_0 \\ &\left. - 2e^{\tau_0} (n + n \coth n\tau_0 + \sinh 2\tau_0) \right\}, \end{aligned} \quad (4.77)$$

$$U_y^\perp = -\frac{1}{2}\tilde{\zeta}_c U_0 \coth \tau_0, \quad (4.78)$$

$$\Omega^\perp = -2\frac{\tilde{\zeta}_c U_0}{R} \frac{\coth^2 \tau_0 (\cosh \tau_0 - \tau_0 / \sinh \tau_0)}{2\tau_0 + \sinh 2\tau_0}, \quad (4.79)$$

and

$$\begin{aligned} U_x^\parallel &= \frac{1}{4}U_0[K_1(8\tau_0 - 3\tanh 2\tau_0) - \tilde{\zeta}_c \frac{(e^{2\tau_0} - 1)^2}{1 + e^{4\tau_0}} \tanh 2\tau_0] \\ &+ \frac{1}{8}U_0(2\tau_0 - \tanh 2\tau_0) \left\{ e^{-3\tau_0} \frac{\sinh \tau_0}{\cosh 2\tau_0} (\sinh \tau_0 - 2 \cosh 3\tau_0 - \sinh 3\tau_0) \right. \\ &\times \tanh^2 \tau_0 + \sum_{n=2}^{\infty} n e^{-(2n+1)\tau_0} \tanh n\tau_0 \sinh \tau_0 [2n e^{\tau_0} (2 \sinh^2 \tau_0 - \tanh n\tau_0) \\ &- 2e^{\tau_0} \sinh 2\tau_0 + e^{2\tau_0} (n-1) \cosh \tau_0 \tanh (n-1)\tau_0 \\ &\left. + (n+1) \cosh \tau_0 \tanh (n+1)\tau_0 \right\}, \end{aligned} \quad (4.80)$$

$$U_y^{\parallel} = 0, \quad (4.81)$$

$$\Omega^{\parallel} = 0. \quad (4.82)$$

Equations (4.77) $\sim$ (4.82) show that the cylinders not only translate along the x -axis and the y -axis but also rotate under the perpendicular applied electric field, while solely translate along the x -axis under the parallel applied electric field. $U_x^{\perp}$ is due to the nonuniform electric field and the ICEO induced on cylinder-2; $U_y^{\perp}$ and $\Omega^{\perp}$ are solely due to the electroosmosis (EO) induced on cylinder-1. $U_x^{\parallel}$ is resulted from the nonuniform electric field as well as the electroosmosis (EO) and the ICEO induced on the cylinders.

4.3 Results and Discussion

In the following discussion, the presented electric and flow fields are calculated at the dimensionless half distance between the two cylinders $c/R = 2.0$, where c is the half distance between the cylinder centres and R is the radius of the cylinders as illustrated in Figure 4.1.

4.3.1 Electric field

Figure 4.2 presents the electric fields around the two stationary cylinders under the perpendicular and the parallel applied electric fields. The electric field lines and the equipotential lines are indicated by the solid and the dashed curves, respectively. The increment between the neighbouring electric field lines (equipotential lines) is $V/(E_{\infty}R) = \phi/(E_{\infty}R) = 0.4$. The two cylinders (conducting or non-conducting) are fully screened by the EDLs. The electric field lines do not enter the cylinders. Due to the appearance of the other cylinder, the local electric field around one cylinder becomes asymmetric to the y -axis. The symmetry to the x -axis still remains. Therefore, the two cylinders experience electrostatic force along the x -axis as shown by Equations (4.62) and (4.63).

Figure 4.2: Electric field around the two stationary cylinders under (a) the perpendicular and (b) the parallel applied electric fields. The electric field lines and the equipotential lines are indicated by the solid and the dashed lines, respectively. The increment between the neighbouring electric field lines (equipotential lines) is $V/(E_\infty R) = \phi/(E_\infty R) = 0.4$.

Under the perpendicular applied electric field, as shown in Figure 4.2(a), the local electric field between the two cylinders are strengthened compared to that on the other side of the cylinders. Moreover, the induced zeta potential ζ_i on the left-hand side of the conducting cylinder (cylinder-2) is stronger than that on the right-hand side. Thus, a relatively intensified flow is induced between the two cylinders. As the distance between the two cylinders further reduces, the corresponding local electric field becomes even stronger and leads to a more intensified flow in this region. When the electric field is applied in parallel, see Figure 4.2(b), the local electric field between the two cylinders is weakened rather than strengthened. The induced zeta potential ζ_i of cylinder-2 reduces on the left-hand side accordingly, which leads to a relative weaker flow between the two cylinders. As the distance decreases, the local electric field between the two cylinders is further attenuated. Corresponding, the slip velocities on the cylinders as well as the aroused flow is reduced.

4.3.2 Flow field

Figures 4.3 and 4.4 present the streamlines around the two stationary cylinders under the perpendicular and the parallel applied electric fields, respectively. The flow directions are indicated by the arrows. The increment between the neighbouring streamlines is $\varphi/(U_0R) = 0.2$ except the specifically labelled streamlines $\varphi/(U_0R) = \pm 0.1$ in Figures 4.4(a) and (b), and $\varphi/(U_0R) = \pm 0.5$ in Figure 4.4(c).

Figure 4.3: Flow field (streamlines) around the two stationary cylinders under the perpendicular applied electric field. In (a) and (b), the dimensionless zeta potential of cylinder-1, $\tilde{\zeta}_c = 0$ and 0.5 , respectively. The increment between the neighbouring streamlines is $\varphi/(U_0R) = 0.2$. The flow directions are indicated by the arrows.

Figures 4.3(a) and (b) show the flow fields around the two stationary cylinders under the perpendicular applied electric field when the dimensionless zeta potential of cylinder-1 (non-conducting), $\tilde{\zeta}_c = 0$ and 0.5 , respectively. It has been reported that the ICEO around a conducting cylinder is quadrupolar and symmetric to both the x -axis and the y -axis [1]. While in the present model, the ICEO around cylinder-2 is disturbed by the presence of cylinder-1. Thus, as shown in Figure 4.3(a), the local flow field around cylinder-2 becomes asymmetric to the y -axis but remains symmetric to the x -axis when cylinder-1 is uncharged. Two bounded

microvortices are formed between the two cylinders. Under such condition, the two cylinders experience hydrodynamic force along the x -axis but not the y -axis, and experience no hydrodynamic moment. When the dimensionless zeta potential of cylinder-1, $\tilde{\zeta}_c = 0.5$, the flow around the two cylinders loses symmetry to both the x -axis and the y -axis as illustrated in Figure 4.3(b). The EO on cylinder-1 merges with the ICEO on the cylinder-2, forming a large microvortex encircling the two cylinders as well as the two microvortices around cylinder-2. Therefore, the cylinders experience hydrodynamic forces along both the x -axis and the y -axis, and also the hydrodynamic moment.

When the electric field is applied in parallel, as shown in Figure 4.4, the flow field around the two stationary cylinders is always symmetric to the x -axis but not the y -axis. Therefore, the cylinders experience hydrodynamic force along the x -axis solely, and translate along the x -axis when they are free to move. The ICEO on the right-hand side of cylinder-2 is much stronger than that on the left-hand side as shown in Figure 4.4(a). That is because the local electric field on the left-hand side of cylinder-2 is weaker (Figure 4.2(b)). Moreover, the ICEO is also attenuated by the viscous effect due to the presence of cylinder-1. Thus, as shown in Figure 4.4(a), a net flow along the negative x -axis is generated due to the asymmetric ICEO on cylinder-2. When cylinder-1 carries positive net charges, e.g., the dimensionless zeta potential of cylinder-1, $\tilde{\zeta}_c = 0.5$, the microvortices on the left-hand side of cylinder-2 is further reduced as presented in Figure 4.4(b). The generated net flow along the negative x -axis is strengthened correspondingly due to the EO aroused on cylinder-1. When the dimensionless zeta potential of cylinder-1 is negative, e.g., $\tilde{\zeta}_c = -0.5$, the EO generated on cylinder-1 is along the positive x -axis, however, the ICEO on the right-hand side of cylinder-2 is still stronger and encircles the EO with two microvortices formed on cylinder-1. As illustrated in Figure 4.4(c), the net flow along the negative x -axis remains. However, this net flow direction can be reversed if the negative net charges on cylinder-1 keep increasing.

Figure 4.4: Flow field (streamlines) around the two stationary cylinders under the parallel applied electric field. In (a), (b) and (c) the dimensionless zeta potential of cylinder-1, $\tilde{\zeta}_c = 0, 0.5$ and -0.5 , respectively. The increment between the neighbouring streamlines is $\varphi/(U_0 R) = 0.2$ except the specifically labelled streamlines $\varphi/(U_0 R) = \pm 0.1$ in (a) and (b), and $\varphi/(U_0 R) = \pm 0.5$ in (c). The flow directions are indicated by the arrows.

4.3.3 Cylinder velocities

Not surprisingly, the cylinders move when they are not fixed. This is understandable since the local electric field around one cylinder is disturbed by

the presence of the other cylinder. The nonuniform local electric field exerts electrostatic force on the cylinders which contributes to the cylinder velocities. Furthermore, as illustrated in Figures 4.3 and 4.4, the corresponding flow field is asymmetric even when cylinder-1 carries no net charge. The flow asymmetry is further enhanced by the EO on cylinder-1 when it is charged. This asymmetric flow field drives the cylinders into motion. In the following analysis, we consider the velocities of cylinder-2. The velocities of cylinder-1 are of the same magnitudes but opposite directions.

The variation of the velocities of cylinder-2 under the perpendicular and the parallel applied electric fields is presented in Figures 4.5 and 4.6, respectively. Clearly, the cylinder velocities due to the nonuniform local electric field are small and negligible compared to that due to the asymmetric flow. Similar conclusions have been reported on the spherical particles [46] and wall effect to cylinders [42]. However, one should note that when the applied electric field is nonuniform the cylinder velocities due to the electrostatic force could be significant [5].

Figure 4.5 shows that cylinder-2 not only translates but also rotates when the dimensionless zeta potential of cylinder-1, $\tilde{\zeta}_c$, is not zero. All the three velocities of cylinder-2 decrease as the distance becomes large. However, the decline rate of U_x is much smaller than that of U_y and Ω . Figure 4.5(a) shows that U_x is due to two factors, namely the ICEP and the electrostatic force. The velocity component due to the electrostatic force is positive and very small compared to that due to the ICEP. Thus, the total U_x of cylinder-2 can be accurately described by the ICEP component. As the distance increases, U_x enlarges first and then decreases to zero. That is because the local electric field and the corresponding induced zeta potential of cylinder-2 increase as the distance reduces, which enlarges U_x . While the viscous effect also becomes stronger as the distance reduces, which reduces U_x . The competition of these two factors produces the final variation curve of U_x . As introduced previously, U_y and Ω are solely due to the electrophoresis (EP) as presented by Equations (4.78) and (4.79). Figures 4.5(b) and (c) present that U_y

Figure 4.5: Variation of the velocities of cylinder-2 (a) U_x/U_0 , (b) $U_y/(\zeta_c U_0)$ and (c) $\Omega R/(\zeta_c U_0)$ with the dimensionless half distance c/R under the perpendicular applied electric field. ICEP and ES in (a) indicate the velocity components due to the ICEP and the electrostatic force, respectively. The component due to the electrostatic force is amplified 10 times for a better observation.

and Ω are of opposite sign of $\tilde{\zeta}_c$. As the distance increases, both U_y and Ω decrease. The difference is that Ω reduces to zero while U_y approaches -0.5. This is reasonable since the cylinders rotate due to the hydrodynamic interactions between them. When the distance is very large, the interactions between the two cylinders become very weak or even vanish. Thus, the cylinder rotation disappears correspondingly. At the large distance, U_y of cylinder-2 equals to -0.5. Thus, cylinder-1 moves along the y -axis with velocity 0.5. Therefore, cylinder-1 moves along the y -axis

with velocity $U_y = 1$ relative to cylinder-2, which is the electrophoresis velocity of cylinder-1 when it is isolated [108].

Figure 4.6: Variation of the velocity of cylinder-2 U_x/U_0 with the dimensionless half distance c/R under the parallel applied electric field. In (a) and (b), the dimensionless zeta potential of cylinder-1 $\tilde{\zeta}_c = 1$ and -1 , respectively. ES, EP and ICEP indicate the velocity components due to the electrostatic force (ES), EP and ICEP, respectively. “Total” means the total velocity of cylinder-2. Insets: the flow fields indicated by streamlines with increment $\varphi/(U_0 R) = 0.5$ except the specifically labeled streamlines $\varphi/(U_0 R) = \pm 0.1$.

Figure 4.6 presents the variation of the velocity components of cylinder-2 under the parallel applied electric field. There are three factors contributing to U_x/U_0 , namely the electrostatic force (ES), the electrophoresis (EP) and the ICEP (Equation (4.80)). The component due to electrostatic force is negative and small. It decreases from zero and approaches zero rapidly as c/R increases. This is due to the fact that the local electric field between the two cylinders is weaker than that on the other side of the cylinders. Thus, the electrostatic force drives the two cylinders move towards each other. The ICEP component is positive and increases from zero and diminishes to zero as the distance enlarges. This means that the ICEO induced on cylinder-2 drives the cylinder apart from each other. The electrophoresis (EP) component is negatively large at $\tilde{\zeta}_c = 1$ (Figure 4.6(a)) but positively large at $\tilde{\zeta}_c = -1$ (Figure 4.6(b)). As c/R increases, its magnitude increases and approaches -0.5 and 0.5 as shown in Figures 4.6(a) and

(b), respectively. Similar to the perpendicular case, cylinder-1 move with velocity $U_x = 1$, the electrophoretic velocity [108], relative to cylinder-2 when the distance is large. These three components form the total velocity of cylinder-2, which generally varies with a similar trend of the electrophoresis (EP) component. However, the attraction and repulsion effects between the two cylinders vary as $\tilde{\zeta}_c$ changes. When $\tilde{\zeta}_c$ is positively large, i.e. the EP component plus the ES component is larger than the ICEP component, the two cylinders attract each other (Figure 4.6(a)). While when $\tilde{\zeta}_c$ is positively small or negative, the two cylinders repel each other (Figure 4.6(b)). When c/R approaches 1.0, the velocity of cylinder-2 approaches zero. However, the EDL overlap may occur under such extreme situation, which is beyond the scope of the present model.

As discussed in Section 3.3.4, to validate the thin EDL and weak electric field approximations, the EDL thickness λ_D should be much smaller than the cylinder radius R and the distance between the cylinders $2c$, $\lambda_D \ll \min(R, 2c)$, and the zeta potentials of the conducting (ζ_i) and non-conducting (ζ_c) cylinders smaller than the thermal voltage ϕ_T , $\max(\zeta_i, \zeta_c) < \phi_T$, where ϕ_T is around 25 mV at room temperature. $\lambda_D = \Lambda/\sqrt{c_0}$, where $\Lambda \approx 10 \text{ mol}^{1/2}/\text{m}^{1/2}$ at room temperature [33]; and c_0 is the electrolyte concentration. ζ_i is of order $2E_0R$ (Equations (4.11) and (4.12)). Thus, $\max(2E_0R, \zeta_c) < \phi_T$. The ranges of c_0 , E_0 and R are coupled due to the criteria of the thin EDL and weak electric field approximations. c_0 ranges from $1 \times 10^{-6} \text{ mol/L}$ to $1 \times 10^{-3} \text{ mol/L}$ in practical applications. Hence, λ_D ranges from 300 nm to 10 nm, which requires $\min(R, 2c)$ be larger than 3 μm to 100 nm corresponding to c_0 . When $\min(R, 2c)=R$ and $\max(2E_0R, \zeta_c) = 2E_0R$, the maximum E_0 should be smaller than $4.3 \times 10^3 \text{ V/m}$ to $1.3 \times 10^5 \text{ V/m}$ corresponding to c_0 . When $\min(R, 2c)=2c$ and/or $\max(2E_0R, \zeta_c) = \zeta_c$, the range of E_0 changes. As the ranges of c_0 , E_0 and R are interrelated, the parameter ranges can be determined using the presented criteria of the thin EDL and weak electric field approximations.

4.4 Summary

In this chapter, the pair interactions of two infinite cylinders under the uniform external electric field are analytically studied with one cylinder conducting and one non-conducting. The thin EDL and weak external electric field approximations are adopted. The results reveal that the external electric field generates microvortices around the two cylinders. Moreover, the two cylinders are driven into motion due to the nonuniform local electric field and the asymmetric flow. Three factors contribute to the cylinder velocities, namely the electrostatic force, the electrophoresis (EP) and the induced charge electrophoresis (ICEP). The component due to the electrostatic force is much smaller and negligible compared to the other two components. The cylinders both translate and rotate under the perpendicular applied electric field, but solely translate along the x -axis under the parallel applied electric field. As the microvortices and net fluid flow are generated around the cylinders, the present study shows potential applications in fluid mixing and pumping. Additionally, the study also helps the understanding of particle manipulations in colloid science and micro/nanofluidics.

Chapter 5

Chaotic Mixing Utilising Electroosmosis and Induced Charge Electroosmosis in Eccentric Annulus

5.1 Introduction

Efficient mixing is of critical importance for many lab-on-a-chip systems for chemical reactions [14, 122, 123], biological analysis [15, 21, 124, 125, 126], particle synthesis [22, 127], medical analysis [23], colloid science [24], etc. Therefore, developing effective techniques for a rapid and homogenous mixing remains hot topics of extensive scientific and technological interests. However, this has long been a challenge in micro/nanofluidics due to the small Reynolds number and laminar flow. So far various mixing methods, either passive or active, have been proposed and studied in the effort to enhance mixing [25, 128, 129, 130]. Passive micromixers have advantages as free of moving part and no additional energy consumptions, while their commonly employed geometries are complicated and pose fabrication challenges [131, 132, 133]. Compared to the passive mixing method, active micromixers utilize external fields, such as pressure [134, 135], electric [136, 137], magnetic [138], acoustic [139], and thermal fields [140], to create secondary flows for mixing enhancement. The flexibility in tunable mixing control and the resulted better mixing effect often enable the active mixing to be preferable [141].

Electrically induced mixing, as a type of active mixing method, has been

confirmed to be simple and effective in prompt mixing [26, 32, 129, 142], among which the induced charge electroosmosis (ICEO) presents a great potential in mixing. ICEO microvortices around the polarizable spherical particle and circular cylinder have been both theoretically predicted [1, 34, 62] and experimentally observed [29, 55]. Some studies have been performed on the mixing behaviour of the ICEO [9, 10, 64, 65]. Harnett et al. [64], and Wu and Li [9] developed ICEO micromixers by positioning many triangular posts in microchannels. However, the ICEO microvortices are intrinsically laminar, therefore, molecular diffusion is required for the transportation across streamlines. Hence, the mixing enhancement is still limited by molecular diffusion. In order to conquer such limitations, this chapter proposes to realise an efficient mixing by introducing Lagrangian chaos into the EO and the ICEO micromixers.

Lagrangian chaos has been demonstrated to enhance mixing significantly in various flow conditions, such as secondary flow induced by herringbone structure on the bottom of channel wall [143], and droplet flow in meandering microchannels [144, 133]. Zhao and Bau [68] reported the Lagrangian chaos created by periodically alternating two ICEO flows in the concentric annulus. Such ICEO flows are produced by changing the applied electric field directions. Compared to the concentric annulus, the eccentric annulus, however, is a more general case (with a concentric annulus as a special case when the eccentricity becomes zero), which has been more frequently encountered in reality, thus, the examination of which could have more practical implications. Questions are raised in terms of the effect brought by the eccentricity: Could Lagrangian chaos be generated by periodically alternating EO or ICEO flows as well in the eccentric annulus? If so, does the presence of eccentricity harm or benefit the mixing? Besides the eccentricity, many other factors, including the alternating time period, the number of flow patterns in one time period, and the specific flow patterns utilised for Lagrangian chaos creation, deserve systematic investigations and optimisations for either concentric or eccentric annuli.

In this chapter, the aforementioned questions are addressed by carrying out a comprehensive analysis on the mixing behaviour of the Lagrangian chaos created by either the EO or the ICEO. The results show that using the proposed micromixing method, a homogenous mixing can be achieved rapidly, e.g., within 1 s under a moderate electric field, which is much faster than many classic micromixers, e.g., herringbone micromixers [143]. Furthermore, due to its simple geometry, the proposed micromixer is easy to fabricate and integrate to other lab-on-a-chip devices. This study could facilitate the understanding of the flow dynamics and the chaotic mixing mechanism for both the EO and the ICEO in the eccentric annulus, and provide an important insight for the design and optimisation of the microchannels aiming for an effective mixing.

5.2 Mathematical Formulation

5.2.1 Problem statement

An eccentric annulus is formed with two two-dimensional cylinders and filled with electrolyte solution. A Cartesian coordinate system is defined in which the centres of the inner and the outer cylinders are located on the positive x -axis. A bipolar coordinate system (τ, σ) is used to describe the eccentric geometry as shown in Figure 5.1. $\tau = \tau_i$ and $\tau = \tau_o$ represent the surfaces of the inner and the outer cylinders, respectively.

Two parameters are introduced to quantitatively describe the eccentric geometry, namely the radius ratio R_r and the eccentricity ε ,

$$R_r = \frac{R_i}{R_o}, \quad \varepsilon = \frac{d}{R_o - R_i}, \quad (5.1)$$

where R_i and R_o are the radii of the inner and the outer cylinders, respectively; d is the distance between the centres of the inner and the outer cylinders (Figure 5.1). The annulus becomes concentric when the eccentricity becomes zero.

Figure 5.1: (a) Schematic diagram of the eccentric annulus micromixer; (b) Definition of the bipolar coordinates in the Cartesian coordinates. The surfaces of the inner and the outer cylinders are represented by $\tau = \tau_i$ and $\tau = \tau_o$, respectively; $R_i = a/\sinh \tau_i$ and $R_o = a/\sinh \tau_o$ are the radii of the inner and the outer cylinders, respectively; $(a \coth \tau_i, 0)$ and $(-a \coth \tau_o, 0)$ are the centres of the inner and the outer cylinders, respectively; d is the distance between the centres of the inner and the outer cylinders.

The study is carried out in the limits of the thin EDL and weak electric field approximations. The EDL thickness λ_D should be much smaller than the cylinder radius R_i and the distance between the cylinder and cylindrical pore $R_o - R_i - d$, $\lambda_D \ll \min(R_i, R_o - R_i - d)$. The zeta potentials of the conducting (ζ_i) and non-conducting (ζ_c) cylinders should be smaller than the thermal voltage ϕ_T , i.e., $\zeta_c < \phi_T$ and $\zeta_i < \phi_T$ for the EO and ICEO, respectively. $\lambda_D = \Lambda/\sqrt{c_0}$, where $\Lambda \approx 10 \text{ mol}^{1/2}/\text{m}^{1/2}$ at room temperature [33]; and c_0 is the electrolyte concentration. ζ_i is of order $2E_0R_i$ (Equation (5.7)). Thus, $\zeta_c < \phi_T$ and $2E_0R_i < \phi_T$ should hold for the EO and ICEO, respectively. The ranges of c_0 , E_0 and $\min(R_i, R_o - R_i - d)$ are coupled due to the criteria of the thin EDL and weak electric field approximations. λ_D ranges from 300 nm to 10 nm corresponding to c_0 ranging from $1 \times 10^{-6} \text{ mol/L}$ to $1 \times 10^{-3} \text{ mol/L}$ in practical applications. This requires $\min(R_i, R_o - R_i - d)$ be larger than 3 μm to 100 nm according to c_0 . The range of $\min(R_i, R_o - R_i - d)$ limits the range of E_0 . The present analysis is carried out in the dimensionless form as it is more informative compared to the dimensional form. For practical applications, one may choose parameters according to the presented criteria of the

thin EDL and weak electric field approximations.

5.2.2 Electric field

The fluid outside the EDLs is electrically neutral, whose electrical potential is governed by the Laplace equation

$$\nabla^2 \phi = 0, \quad (5.2)$$

Assuming that the established electric field is weak so that the Faradaic reaction on the inner cylinder can be neglected [68], no electric lines enter the EDL on the inner cylinder

$$\mathbf{e}_\tau \cdot \nabla \phi = 0 \quad \text{at} \quad \tau = \tau_i, \quad (5.3)$$

where $\mathbf{e}_\tau$ is the unit vector in the bipolar coordinates normal to the cylinder surface as shown in Figure 5.1.

Given the uniformly applied electrical potential $E_0 r \sin(\theta + \theta_0)$, the boundary condition on the outer cylinder is [145]

$$\phi = E_0 a \frac{\sin \sigma \cos \theta_0 + \sinh \tau_o \sin \theta_0}{\cosh \tau_o - \cos \sigma} \quad \text{at} \quad \tau = \tau_o, \quad (5.4)$$

where θ_0 is the phase angle of the applied electric field, which defines the direction of the applied electric field (Figure 5.1).

Solving Equation (5.2) together with Equations (5.3) and (5.4) in the bipolar coordinates, the electrical potential is obtained

$$\phi = E_0 a \sin \theta_0 + E_0 a \sum_{n=1}^{\infty} \frac{2e^{-n\tau_o} \cosh n(\tau_i - \tau)}{\cosh n(\tau_i - \tau_o)} (\sin \theta_0 \cos n\sigma + \cos \theta_0 \sin n\sigma). \quad (5.5)$$

When the inner cylinder is non-conducting (non-polarisable), the zeta potential is a constant, ζ_c ; while when the inner cylinder is conducting (ideally polarisable),

the zeta potential is induced by the applied electric field [32]

$$\zeta_i = -\phi + \frac{\int_A \phi dA}{A}. \quad (5.6)$$

Substituting Equation (5.5) into Equation (5.6), the induced zeta potential of the conducting inner cylinder is obtained

$$\zeta_i = -E_0 a \sum_{n=1}^{\infty} \frac{2e^{-n\tau_o}}{\cosh n(\tau_i - \tau_o)} (\sin \theta_0 \cos n\sigma + \cos \theta_0 \sin n\sigma) \quad \text{at} \quad \tau = \tau_i. \quad (5.7)$$

5.2.3 Flow field

Generally, the Reynolds number is very small in the EO and the ICEO [1] and the fluid is incompressible. Thus, the flow field satisfies the Stokes equations, which can be written in terms of stream function ψ

$$\nabla^4 \psi = 0, \quad (5.8)$$

with the velocities in the bipolar coordinates defined as

$$u_\sigma = h \frac{\partial \psi}{\partial \tau}, \quad u_\tau = -h \frac{\partial \psi}{\partial \sigma}, \quad (5.9)$$

where $h = (\cosh \tau - \cos \sigma) / a$.

The general solution of the stream function ψ in the bipolar coordinates suitable for the present case was given by Jeffery [116]

$$\begin{aligned} h\psi = & A_0 \cosh \tau + B_0 \tau (\cosh \tau - \cos \sigma) + C_0 \sinh \tau + D_0 \tau \sinh \tau \\ & + \sum_{n=1}^{\infty} [a_n \cosh(n+1)\tau + b_n \cosh(n-1)\tau + c_n \sinh(n+1)\tau \\ & + d_n \sinh(n-1)\tau] \cos n\sigma + h_1 \tau \sin \sigma + \sum_{n=1}^{\infty} [e_n \cosh(n+1)\tau \\ & + f_n \cosh(n-1)\tau + g_n \sinh(n+1)\tau + h_n \sinh(n-1)\tau] \sin n\sigma. \end{aligned} \quad (5.10)$$

The established electric field exerts an electrical force on the ions within the EDL, which drives the ions into motion. Consequently, a flow is aroused in the bulk fluid. In the limit of the thin EDL ($\sim \text{nm}$) [1] and the weak electric field (ζ is smaller than the thermal voltage, $\sim 25 \text{ mV}$) [146] approximations, the slip velocity on the inner cylinder can be described by the Helmholtz-Smoluchowski formula [109]

$$\mathbf{u}_s = -\frac{\varepsilon_f \zeta}{\mu} E_\sigma \mathbf{e}_\sigma \quad \text{at} \quad \tau = \tau_i, \quad (5.11)$$

where E_σ is the electric field component that acts on and is tangential to the inner cylinder surface; $\mathbf{e}_\sigma$ is the unit vector tangent to the cylinder surface in bipolar coordinates. Substituting the tangential electric field (obtained from Equation (5.5) through $\mathbf{E} = -\nabla\phi$) and the zeta potentials (ζ_c and ζ_i for the non-conducting and conducting inner cylinders, respectively) on the inner cylinder into Equation (5.11), the slip velocities on the non-conducting and conducting inner cylinders are obtained.

This study aims to investigate the flow dynamics of the EO and the ICEO generated on the non-conducting and conducting inner cylinder, respectively; and further examine the mixing behavior of the corresponding Lagrangian chaos within the eccentric annulus. It is assumed that the possible electroosmotic effect on the inner surface of the outer cylinder is negligible. Hence, the no-slip boundary condition is applied on the outer cylinder

$$\mathbf{u} = 0 \quad \text{at} \quad \tau = \tau_o. \quad (5.12)$$

Substituting Equation (5.10) into Equations (5.11) and (5.12) through Equation (5.9), all the unknown coefficients are determined. To facilitate the following analysis, the translational velocity scale of the inner cylinder U_i and dimensionless zeta potential $\tilde{\zeta}$ are defined as

$$U_i = \tilde{\zeta} \frac{\varepsilon_w E_0^2 R_i}{\mu}, \quad (5.13)$$

and

$$\tilde{\zeta} = \begin{cases} \frac{\zeta_c}{E_0 R_i}, & \text{for the non-conducting cylinder,} \\ 1, & \text{for the conducting cylinder.} \end{cases} \quad (5.14)$$

respectively.

The coefficients for the EO on the non-conducting inner cylinder are obtained as

$$\begin{aligned} A_0^{nc} = & \left\{ K_0^{nc} \left[\frac{(\tau_i - \tau_o) \sinh \tau_o (\sinh 2\tau_o - 2\tau_i)}{\sinh(\tau_i - \tau_o)} \right. \right. \\ & - \sinh \tau_i (2\tau_o - 3\tau_i + \tau_i \cosh 2\tau_o + \sinh 2\tau_o) \\ & + \cosh \tau_i ((2\tau_i - \tau_o) \sinh 2\tau_o - 2\tau_i (\tau_i - \tau_o)) \Big] \\ & + \frac{1}{4} K_1^{nc} [\cosh 2\tau_i + \cosh 2(\tau_i - \tau_o) - 1 - 4\tau_i (\tau_i - \tau_o) \\ & - \cosh 2\tau_o - 2\tau_o \sinh 2\tau_i + 2\tau_i \sinh 2\tau_o] \Big\} \\ & / [(\tau_i - \tau_o) (\cosh 2\tau_i + \cosh 2\tau_o - 2) - \sinh 2\tau_i \\ & + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o], \end{aligned} \quad (5.15)$$

$$\begin{aligned} B_0^{nc} = & \left\{ K_0^{nc} \coth(\tau_i - \tau_o) [\cosh(\tau_i - 2\tau_o) - \cosh \tau_i + 2(\tau_i - \tau_o) \sinh \tau_i] \right. \\ & + \frac{1}{2} K_1^{nc} [2(\tau_i - \tau_o) + \sinh 2\tau_i - \sinh 2\tau_o] \Big\} \\ & / [(\tau_i - \tau_o) (\cosh 2\tau_i + \cosh 2\tau_o - 2) - \sinh 2\tau_i \\ & + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o], \end{aligned} \quad (5.16)$$

$$\begin{aligned} C_0^{nc} = & \left\{ K_0^{nc} \left[-\frac{2(\tau_i - \tau_o) \cosh \tau_i \sinh \tau_o}{\sinh(\tau_i - \tau_o)} \right. \right. \\ & + \cosh \tau_i (3\tau_o - 2\tau_i - \tau_o \cosh 2\tau_o) + \sinh 2\tau_o (\cosh \tau_i + \tau_i \sinh \tau_i) \Big] \\ & - \frac{1}{2} K_1^{nc} [(\tau_i - \tau_o) - \tau_o \cosh 2\tau_i + \tau_i \cosh 2\tau_o + \cosh^2 \tau_o \sinh 2\tau_i \\ & - \cosh^2 \tau_i \sinh 2\tau_o] \Big\} / [(\tau_i - \tau_o) (\cosh 2\tau_i + \cosh 2\tau_o - 2) \\ & - \sinh 2\tau_i + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o], \end{aligned} \quad (5.17)$$

$$\begin{aligned}
 D_0^{nc} = & -\sinh(\tau_i - \tau_o) [2K_0^{nc} \sinh \tau_o + K_1^{nc} \sinh(\tau_i + \tau_o)] \\
 & / [(\tau_i - \tau_o)(\cosh 2\tau_i + \cosh 2\tau_o - 2) - \sinh 2\tau_i \\
 & + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o], \tag{5.18}
 \end{aligned}$$

$$\begin{aligned}
 A_1^{nc} = & \left\{ \frac{1}{2} K_0^{nc} \sinh(\tau_i + \tau_o) [\cosh \tau_i - \cosh(\tau_i - 2\tau_o) - 2(\tau_i - \tau_o) \sinh \tau_i] \right. \\
 & - \frac{1}{8} K_1^{nc} [\cosh \tau_o \sinh 3\tau_i - 3 \cosh 3\tau_o \sinh \tau_i + \cosh \tau_i \sinh 3\tau_o \\
 & + \sinh \tau_o (\cosh 3\tau_i - 4 \cosh \tau_i - 8(\tau_i - \tau_o) \cosh(\tau_i + \tau_o) \sinh \tau_o)] \left. \right\} \\
 & / \{ \sinh(\tau_i - \tau_o) [(\tau_i - \tau_o)(\cosh 2\tau_i + \cosh 2\tau_o - 2) \\
 & - \sinh 2\tau_i + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o] \}, \tag{5.19}
 \end{aligned}$$

$$\begin{aligned}
 B_1^{nc} = & \left\{ \frac{1}{2} K_0^{nc} [(2\tau_i \coth(\tau_i - \tau_o) - 1) (\cosh(\tau_i - 2\tau_o) \right. \\
 & - \cosh \tau_i + 2(\tau_i - \tau_o) \sinh \tau_i)] \\
 & + \frac{1}{4} K_1^{nc} [\cosh 2\tau_i (1 - 2 \cosh 2\tau_o) + \cosh 2\tau_o \\
 & + 2 \left(2(\tau_i - \tau_o) \left(\tau_i - \frac{\sinh \tau_i \sinh \tau_o}{\sinh(\tau_i - \tau_o)} \right) \right. \\
 & \left. \left. - \tau_o \sinh 2\tau_o + \sinh 2\tau_i (\tau_o + \sinh 2\tau_o) \right) \right] \left. \right\} \\
 & / [(\tau_i - \tau_o)(\cosh 2\tau_i + \cosh 2\tau_o - 2) - \sinh 2\tau_i \\
 & + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o], \tag{5.20}
 \end{aligned}$$

$$\begin{aligned}
 C_1^{nc} = & \left\{ \frac{1}{2} K_0^{nc} \cosh(\tau_i + \tau_o) [\cosh(\tau_i - 2\tau_o) - \cosh \tau_i + 2(\tau_i - \tau_o) \sinh \tau_i] \right. \\
 & - \frac{1}{4} K_1^{nc} [\cosh(\tau_i + \tau_o) - \cosh(\tau_i - 3\tau_o) \\
 & - \sinh(\tau_i - \tau_o) (\tau_i - \tau_o + \sinh 2(\tau_i + \tau_o)) \\
 & \left. + (\tau_i - \tau_o) (\sinh(\tau_i + 3\tau_o) - 2 \sinh(\tau_i + \tau_o)) \right] \left. \right\} \\
 & / \{ \sinh(\tau_i - \tau_o) [(\tau_i - \tau_o)(\cosh 2\tau_i + \cosh 2\tau_o - 2) \\
 & - \sinh 2\tau_i + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o] \}, \tag{5.21}
 \end{aligned}$$

$$\begin{aligned}
 a_n^{nc} = & -\frac{1}{2}K_n^{nc} [(n-1) \sinh(n+1)\tau_i - n \sinh((n-1)\tau_i + 2\tau_o) \\
 & - \sinh((n-1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\
 & + \cosh 2n(\tau_i - \tau_o)],
 \end{aligned} \tag{5.22}$$

$$\begin{aligned}
 b_n^{nc} = & -\frac{1}{2}K_n^{nc} [(n+1) \sinh(n-1)\tau_i - n \sinh((n+1)\tau_i - 2\tau_o) \\
 & + \sinh((n+1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\
 & + \cosh 2n(\tau_i - \tau_o)],
 \end{aligned} \tag{5.23}$$

$$\begin{aligned}
 c_n^{nc} = & \frac{1}{2}K_n^{nc} [(n-1) \cosh(n+1)\tau_i - n \cosh((n-1)\tau_i + 2\tau_o) \\
 & + \cosh((n-1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\
 & + \cosh 2n(\tau_i - \tau_o)],
 \end{aligned} \tag{5.24}$$

$$\begin{aligned}
 d_n^{nc} = & \frac{1}{2}K_n^{nc} [(n+1) \cosh(n-1)\tau_i - n \cosh((n+1)\tau_i - 2\tau_o) \\
 & - \cosh((n+1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\
 & + \cosh 2n(\tau_i - \tau_o)],
 \end{aligned} \tag{5.25}$$

where $n \geq 2$ and

$$K_0^{nc} = -\tilde{\zeta}U_i \frac{e^{-\tau_o} \cos \theta_0}{\cosh(\tau_i - \tau_o)}, \tag{5.26}$$

$$\begin{aligned}
 K_n^{nc} = & \tilde{\zeta}U_i \left[\frac{2ne^{-n\tau_o} \cosh \tau_i}{\cosh n(\tau_i - \tau_o)} - \frac{(n-1)e^{-(n-1)\tau_o}}{\cosh(n-1)(\tau_i - \tau_o)} \right. \\
 & \left. - \frac{(n+1)e^{-(n+1)\tau_o}}{\cosh(n+1)(\tau_i - \tau_o)} \right] \cos \theta_0, \quad (n \geq 1),
 \end{aligned} \tag{5.27}$$

and

$$e_1^{nc} = \frac{1}{4}\Lambda_1^{nc} \frac{2(\tau_i - \tau_o) \cosh 2\tau_o - \sinh 2\tau_i + \sinh 2\tau_o}{(\tau_i - \tau_o) \sinh 2(\tau_i - \tau_o) - 2 \sinh^2(\tau_i - \tau_o)}, \tag{5.28}$$

$$f_1^{nc} = -\frac{1}{4}\Lambda_1^{nc} \frac{2\tau_i - 2\tau_o \cosh 2(\tau_i - \tau_o) - \sinh 2(\tau_i - \tau_o)}{(\tau_i - \tau_o) \sinh 2(\tau_i - \tau_o) - 2 \sinh^2(\tau_i - \tau_o)}, \quad (5.29)$$

$$g_1^{nc} = -\frac{1}{4}\Lambda_1^{nc} \frac{2(\tau_i - \tau_o) \sinh 2\tau_o - \cosh 2\tau_i + \cosh 2\tau_o}{(\tau_i - \tau_o) \sinh 2(\tau_i - \tau_o) - 2 \sinh^2(\tau_i - \tau_o)}, \quad (5.30)$$

$$h_1^{nc} = -\frac{1}{2} \frac{\Lambda_1^{nc}}{(\tau_i - \tau_o) \coth(\tau_i - \tau_o) - 1}, \quad (5.31)$$

$$\begin{aligned} e_n^{nc} = & -\frac{1}{2}\Lambda_n^{nc} [(n-1) \sinh(n+1)\tau_i - n \sinh((n-1)\tau_i + 2\tau_o) \\ & - \sinh((n-1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\ & + \cosh 2n(\tau_i - \tau_o)], \end{aligned} \quad (5.32)$$

$$\begin{aligned} f_n^{nc} = & -\frac{1}{2}\Lambda_n^{nc} [(n+1) \sinh(n-1)\tau_i - n \sinh((n+1)\tau_i - 2\tau_o) \\ & + \sinh((n+1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\ & + \cosh 2n(\tau_i - \tau_o)], \end{aligned} \quad (5.33)$$

$$\begin{aligned} g_n^{nc} = & \frac{1}{2}\Lambda_n^{nc} [(n-1) \cosh(n+1)\tau_i - n \cosh((n-1)\tau_i + 2\tau_o) \\ & + \cosh((n-1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\ & + \cosh 2n(\tau_i - \tau_o)], \end{aligned} \quad (5.34)$$

$$\begin{aligned} h_n^{nc} = & \frac{1}{2}\Lambda_n^{nc} [(n+1) \cosh(n-1)\tau_i - n \cosh((n+1)\tau_i - 2\tau_o) \\ & - \cosh((n+1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\ & + \cosh 2n(\tau_i - \tau_o)], \end{aligned} \quad (5.35)$$

where $n \geq 2$ and

$$\begin{aligned} \Lambda_n^{nc} = & -\tilde{\zeta} U_i \left[\frac{2ne^{-n\tau_o} \cosh \tau_i}{\cosh n(\tau_i - \tau_o)} - \frac{(n-1)e^{-(n-1)\tau_o}}{\cosh(n-1)(\tau_i - \tau_o)} \right. \\ & \left. - \frac{(n+1)e^{-(n+1)\tau_o}}{\cosh(n+1)(\tau_i - \tau_o)} \right] \sin \theta_0, \quad (n \geq 1). \end{aligned} \quad (5.36)$$

The coefficients for the ICEO on the conducting inner cylinder are obtained as

$$\begin{aligned} A_0^c = & \frac{1}{4} [K_1^c \cosh 2\tau_i - \cosh 2\tau_o + \cosh 2(\tau_i - \tau_o) - 4\tau_i(\tau_i - \tau_o) \\ & - 2\tau_o \sinh 2\tau_i + 2\tau_i \sinh 2\tau_o - 1] / [(\tau_i - \tau_o)(\cosh 2\tau_i + \cosh 2\tau_o - 2) \\ & - \sinh 2\tau_i + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o], \end{aligned} \quad (5.37)$$

$$\begin{aligned} B_0^c = & \frac{1}{2} K_1^c \left(2(\tau_i - \tau_o) + \sinh 2\tau_i - \sinh 2\tau_o \right) \\ & / [(\tau_i - \tau_o)(\cosh 2\tau_i + \cosh 2\tau_o - 2) - \sinh 2\tau_i \\ & + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o], \end{aligned} \quad (5.38)$$

$$\begin{aligned} C_0^c = & -\frac{1}{4} K_1^c [2(\tau_i - \tau_o) - 2\tau_o \cosh 2\tau_i + 2\tau_i \cosh 2\tau_o \\ & + \sinh 2\tau_i + \sinh 2(\tau_i - \tau_o) - \sinh 2\tau_o] \\ & / [(\tau_i - \tau_o)(\cosh 2\tau_i + \cosh 2\tau_o - 2) \\ & - \sinh 2\tau_i + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o], \end{aligned} \quad (5.39)$$

$$\begin{aligned} D_0^c = & -K_1^c \sinh(\tau_i - \tau_o) \sinh(\tau_i + \tau_o) / [(\tau_i - \tau_o)(\cosh 2\tau_i + \cosh 2\tau_o - 2) \\ & - \sinh 2\tau_i + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o], \end{aligned} \quad (5.40)$$

$$\begin{aligned}
 A_1^c &= \frac{1}{8} K_1^c [3 \cosh 3\tau_o \sinh \tau_i - \cosh \tau_o \sinh 3\tau_i + 8(\tau_i - \tau_o) \cosh(\tau_i + \tau_o) \sinh^2 \tau_o \\
 &\quad - 2 \cosh \tau_i (\cosh 2\tau_i - 2 + \cosh 2\tau_o) \sinh \tau_o] / \{ \sinh(\tau_i - \tau_o) [(\tau_i - \tau_o) \\
 &\quad \times (\cosh 2\tau_i + \cosh 2\tau_o - 2) - \sinh 2\tau_i + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o] \}, \quad (5.41)
 \end{aligned}$$

$$\begin{aligned}
 B_1^c &= \frac{1}{4} K_1^c [\cosh 2\tau_i (1 - 2 \cosh 2\tau_o) + \cosh 2\tau_o + 2(2(\tau_i - \tau_o) \\
 &\quad \times \left(\tau_i - \frac{\sinh \tau_i \sinh \tau_o}{\sinh(\tau_i - \tau_o)} \right) - \tau_o \sinh 2\tau_o + \sinh 2\tau_i (\tau_o + \sinh 2\tau_o) \Big] / [(\tau_i - \tau_o) \\
 &\quad \times (\cosh 2\tau_i + \cosh 2\tau_o - 2) - \sinh 2\tau_i + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o], \quad (5.42)
 \end{aligned}$$

$$\begin{aligned}
 C_1^c &= \frac{1}{8} K_1^c [2 \cosh(\tau_i + \tau_o) - 2 \cosh(\tau_i - 3\tau_o) - \cosh(3\tau_i + \tau_o) \\
 &\quad + \cosh(\tau_i + 3\tau_o) + 2(\tau_i - \tau_o) (\sinh(\tau_i + 3\tau_o) - \sinh(\tau_i - \tau_o) \\
 &\quad - 2 \sinh(\tau_i + \tau_o))] / \{ \sinh(\tau_i - \tau_o) [(\tau_i - \tau_o) (\cosh 2\tau_i + \cosh 2\tau_o - 2) \\
 &\quad - \sinh 2\tau_i + \sinh 2(\tau_i - \tau_o) + \sinh 2\tau_o] \}, \quad (5.43)
 \end{aligned}$$

$$\begin{aligned}
 a_n^c &= -\frac{1}{2} K_n^c [(n-1) \sinh(n+1)\tau_i - n \sinh((n-1)\tau_i + 2\tau_o) \\
 &\quad - \sinh((n-1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\
 &\quad + \cosh 2n(\tau_i - \tau_o)], \quad (5.44)
 \end{aligned}$$

$$\begin{aligned}
 b_n^c &= -\frac{1}{2} K_n^c [(n+1) \sinh(n-1)\tau_i - n \sinh((n+1)\tau_i - 2\tau_o) \\
 &\quad + \sinh((n+1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\
 &\quad + \cosh 2n(\tau_i - \tau_o)], \quad (5.45)
 \end{aligned}$$

$$\begin{aligned}
 c_n^c &= \frac{1}{2} K_n^c [(n-1) \cosh(n+1)\tau_i - n \cosh((n-1)\tau_i + 2\tau_o) \\
 &\quad + \cosh((n-1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\
 &\quad + \cosh 2n(\tau_i - \tau_o)], \quad (5.46)
 \end{aligned}$$

$$\begin{aligned}
 d_n^c &= \frac{1}{2} K_n^c \left[(n+1) \cosh(n-1)\tau_i - n \cosh((n+1)\tau_i - 2\tau_o) \right. \\
 &\quad \left. - \cosh((n+1)\tau_i - 2n\tau_o) \right] / \left[n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \right. \\
 &\quad \left. + \cosh 2n(\tau_i - \tau_o) \right], \quad (5.47)
 \end{aligned}$$

where $n \geq 2$ and

$$\begin{aligned}
 K_n^c &= U_i \sinh \tau_i \left\{ \sum_{\substack{k=1, \\ k-n \geq 1}}^{\infty} \frac{2ke^{-(2k-n)\tau_o} \cosh \tau_i}{\cosh k(\tau_i - \tau_o) \cosh(k-n)(\tau_i - \tau_o)} \right. \\
 &\quad - \sum_{\substack{k=1, \\ k+n \geq 1}}^{\infty} \frac{2ke^{-(2k+n)\tau_o} \cosh \tau_i}{\cosh k(\tau_i - \tau_o) \cosh(k+n)(\tau_i - \tau_o)} \\
 &\quad - \sum_{\substack{k=1, \\ n-k \geq 1}}^{\infty} \frac{2ke^{-n\tau_o} \cosh \tau_i \cos 2\theta_0}{\cosh k(\tau_i - \tau_o) \cosh(n-k)(\tau_i - \tau_o)} \\
 &\quad - \sum_{\substack{k=1, \\ k-n \geq 2}}^{\infty} \frac{ke^{-(2k-n-1)\tau_o}}{\cosh k(\tau_i - \tau_o) \cosh(k-n-1)(\tau_i - \tau_o)} \\
 &\quad + \sum_{\substack{k=1, \\ k+n \geq 2}}^{\infty} \frac{ke^{-(2k+n-1)\tau_o}}{\cosh k(\tau_i - \tau_o) \cosh(k+n-1)(\tau_i - \tau_o)} \\
 &\quad - \sum_{\substack{k=1, \\ k-n \geq 0}}^{\infty} \frac{ke^{-(2k-n+1)\tau_o}}{\cosh k(\tau_i - \tau_o) \cosh(k-n+1)(\tau_i - \tau_o)} \\
 &\quad + \sum_{\substack{k=1, \\ k+n \geq 0}}^{\infty} \frac{ke^{-(2k+n+1)\tau_o}}{\cosh k(\tau_i - \tau_o) \cosh(k+n+1)(\tau_i - \tau_o)} \\
 &\quad + \sum_{\substack{k=1, \\ n-k \geq 0}}^{\infty} \frac{ke^{-(n+1)\tau_o} \cos 2\theta_0}{\cosh k(\tau_i - \tau_o) \cosh(n-k+1)(\tau_i - \tau_o)} \\
 &\quad \left. + \sum_{\substack{k=1, \\ n-k \geq 2}}^{\infty} \frac{ke^{-(n-1)\tau_o} \cos 2\theta_0}{\cosh k(\tau_i - \tau_o) \cosh(n-k-1)(\tau_i - \tau_o)} \right\}, \quad (n \geq 1), \quad (5.48)
 \end{aligned}$$

and

$$e_1^c = \frac{1}{4} \Lambda_1^c \frac{2(\tau_i - \tau_o) \cosh 2\tau_o - \sinh 2\tau_i + \sinh 2\tau_o}{(\tau_i - \tau_o) \sinh 2(\tau_i - \tau_o) - 2 \sinh^2(\tau_i - \tau_o)}, \quad (5.49)$$

$$f_1^c = -\frac{1}{4}\Lambda_1^c \frac{2\tau_i - 2\tau_o \cosh 2(\tau_i - \tau_o) - \sinh 2(\tau_i - \tau_o)}{(\tau_i - \tau_o) \sinh 2(\tau_i - \tau_o) - 2 \sinh^2(\tau_i - \tau_o)}, \quad (5.50)$$

$$g_1^c = -\frac{1}{4}\Lambda_1^c \frac{2(\tau_i - \tau_o) \sinh 2\tau_o - \cosh 2\tau_i + \cosh 2\tau_o}{(\tau_i - \tau_o) \sinh 2(\tau_i - \tau_o) - 2 \sinh^2(\tau_i - \tau_o)}, \quad (5.51)$$

$$h_1^c = -\frac{1}{2} \frac{\Lambda_1^c}{(\tau_i - \tau_o) \coth(\tau_i - \tau_o) - 1}, \quad (5.52)$$

$$\begin{aligned} e_n^c = & -\frac{1}{2}\Lambda_n^c [(n-1) \sinh(n+1)\tau_i - n \sinh((n-1)\tau_i + 2\tau_o) \\ & - \sinh((n-1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\ & + \cosh 2n(\tau_i - \tau_o)], \end{aligned} \quad (5.53)$$

$$\begin{aligned} f_n^c = & -\frac{1}{2}\Lambda_n^c [(n+1) \sinh(n-1)\tau_i - n \sinh((n+1)\tau_i - 2\tau_o) \\ & + \sinh((n+1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\ & + \cosh 2n(\tau_i - \tau_o)], \end{aligned} \quad (5.54)$$

$$\begin{aligned} g_n^c = & \frac{1}{2}\Lambda_n^c [(n-1) \cosh(n+1)\tau_i - n \cosh((n-1)\tau_i + 2\tau_o) \\ & + \cosh((n-1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\ & + \cosh 2n(\tau_i - \tau_o)], \end{aligned} \quad (5.55)$$

$$\begin{aligned} h_n^c = & \frac{1}{2}\Lambda_n^c [(n+1) \cosh(n-1)\tau_i - n \cosh((n+1)\tau_i - 2\tau_o) \\ & - \cosh((n+1)\tau_i - 2n\tau_o)] / [n^2 - 1 - n^2 \cosh 2(\tau_i - \tau_o) \\ & + \cosh 2n(\tau_i - \tau_o)], \end{aligned} \quad (5.56)$$

where $n \geq 2$ and

$$\Lambda_n^c = U_i \sinh \tau_i \left\{ - \sum_{\substack{k=1, \\ n-k \geq 1}}^{\infty} \frac{2ke^{-n\tau_o} \cosh \tau_i}{\cosh k(\tau_i - \tau_o) \cosh(n-k)(\tau_i - \tau_o)} \right. \\ + \sum_{\substack{k=1, \\ n-k \geq 0}}^{\infty} \frac{ke^{-(n+1)\tau_o}}{\cosh k(\tau_i - \tau_o) \cosh(n-k+1)(\tau_i - \tau_o)} \\ \left. + \sum_{\substack{k=1, \\ n-k \geq 2}}^{\infty} \frac{ke^{-(n-1)\tau_o}}{\cosh k(\tau_i - \tau_o) \cosh(n-k-1)(\tau_i - \tau_o)} \right\} \sin 2\theta_0, \quad (n \geq 1). \quad (5.57)$$

Figure 5.2: Flow fields (streamlines) of (a) the EO and (b) the ICEO within the annulus. Here the radius ratio $R_r = 0.1$, the eccentricity $\varepsilon = 0.5$, and the dimensionless zeta potential of the non-conducting inner cylinder $\zeta_c/(E_0 R_i) = 1$. The electric field phase angle $\theta_0 = 0, \pi/2$ in (a); $\theta_0 = 0, \pi/4$ in (b). The increment between the neighbouring streamlines is $\psi/(U_i R_i) = 0.2$. The flow directions are indicated by the arrows.

The obtained flow fields of the electroosmosis (EO) and the ICEO are periodic functions of the electric field phase angle θ_0 with periods 2π and π , respectively. Figure 5.2 presents the flow patterns of the EO and the ICEO in the annulus with electric field applied in different directions, which is defined by the electric field

phase angle θ_0 . Clearly, the EO and the ICEO form two and four counter-rotating microvortices in the annulus, respectively. By changing the electric field direction, i.e. the electric phase angle θ_0 , different flow patterns are formed in the annulus. From the flow fields of the EO at $\theta_0 = 0$, as shown in Figure 5.2(a), we can see that the microvortex on the right-hand side is much larger than that on the left-hand side and encircles the inner cylinder. When the electric phase angle $\theta_0 = \pi/2$, two microvortices symmetric to the x -axis are formed at the upper and the lower sides of the inner cylinder. The microvortices of the ICEO, as shown in Figure 5.2(b), are smaller on the left-hand side of the inner cylinder than those on the right-hand side. They are symmetric to the x -axis at $\theta_0 = 0$, while asymmetric to both the x -axis and the y -axis at $\theta_0 = \pi/4$ as illustrated by the flow directions indicated by the arrows in Figure 5.2(b).

5.2.4 Mixing evaluation

Aref [147] demonstrated that Lagrangian chaos could be created by periodically alternating two or more closed-orbit flows. As shown in Figure 5.2, both the EO and the ICEO can form different closed-orbit flows in the annulus by choosing different electric field phase angles θ_0 . Hence, the Lagrangian chaos could be created by periodically alternating two or more different θ_0 .

The particle tracking method [133] is adopted to quantify and visualise the mixing effect. The non-interacting, passive tracer particles are initially located in the flow field (x_k, y_k) and advected by the EO or the ICEO

$$\frac{d\mathbf{s}}{dt} = \mathbf{u}(t, \theta_0), \quad (5.58)$$

where θ_0 is varied periodically with time

$$\theta_0 = \begin{cases} \theta_1, & 0 \leq t < T/N_{fp}, \\ \dots, & \dots, \\ \theta_{N_{fp}}, & (N_{fp} - 1)T/N_{fp} \leq t < T, \end{cases} \quad (5.59)$$

which corresponds to periodically activate a series of electric fields, and consequently produces different flow patterns. Hence, the chaotic movement of fluid elements in the annulus is created. Here N_{fp} is the number of flow patterns in one time period; $\theta_1 \sim \theta_{N_{fp}}$ are the electric field phase angles of the flow patterns $1 \sim N_{fp}$, respectively; T is the time period for flow pattern alternating.

Integrating Equation (5.58) with the fourth order Runge-Kutta method, the positions (x_k^*, y_k^*) of the tracer particles after advection at each time step are determined. Different time steps are tested until the tracer particles faithfully follow the streamlines of the given flow pattern. The dimensionless time step adopted in the calculation is $\Delta\tilde{t} = \Delta t/t_0 = 10^{-3}$ with time scale $t_0 = R_i/U_i = \mu R_i/(\varepsilon_f \zeta_c E_0)$ and $\mu/(\varepsilon_f E_0^2)$ for the EO and the ICEO, respectively. The velocities $(u_{\tau,k}, u_{\sigma,k})$ of the tracer particles at any spatial location within the annulus can be obtained from Equation (5.10) through Equation (5.9) with the given θ_0 and the obtained coefficients. The dimensionless zeta potentials $\tilde{\zeta} = 1$ are used for both the non-conducting and conducting inner cylinders in the following discussion.

The diffusion of the tracer particles is simulated through random walk

$$x_k = x_k^* + \alpha_{x,k} \sqrt{2D\Delta t}, \quad y_k = y_k^* + \alpha_{y,k} \sqrt{2D\Delta t}, \quad (5.60)$$

where D is the diffusivity of the species in the electrolyte solution; $\alpha_{x,k}$ and $\alpha_{y,k}$ are two independent random numbers with the standard normal distribution for particle k at current time step; (x_k^*, y_k^*) and (x_k, y_k) are the particle positions before and after diffusion. The tracer particles are ensured to be within the annulus by imposing a mirror-reflection if any of them goes out of the annulus.

Due to the eccentric geometry, the annulus is meshed with unequal-sized bins in the bipolar coordinates (τ, σ) with $\Delta\tau = (\tau_i - \tau_o)/N_\tau$ and $\Delta\sigma = 2\pi/N_\sigma$. The particle concentration (particle number divided by bin area) is used to calculate the mixing index η [148]. The standard deviation of the particle concentration at time t is defined as

$$SD(t) = \sqrt{\frac{1}{N_m} \sum_{j=1}^{N_m} (c_j(t) - \bar{c})^2}, \quad (5.61)$$

where N_m is the number of bins ($N_m = N_\tau \times N_\sigma = 40 \times 50$ in the calculation); $c_j(t)$ is the particle concentration in bin j at time t , which is obtained from particles' positions determined through Equations (5.58) and (5.60); and $\bar{c}$ is the particle concentration at ideal mixing

$$\bar{c} = \frac{N_p}{S}, \quad (5.62)$$

where N_p is the total number of particles in the particle tracking ($N_p = 30,000$ in the calculation); S is the area of the annulus.

Initially, all the tracer particles are located in a single bin. Thus, the initial standard deviation SD_0 is large. As time elapses, $SD(t)$ decreases gradually and approaches 0. The mixing index η is defined as the ratio of $SD(t)$ and SD_0

$$\eta = \frac{SD(t)}{SD_0}. \quad (5.63)$$

Ideally, η decreases from 1 and eventually diminishes to 0 when the tracer particles are homogeneously mixed in the whole annulus. In the simulation with equal-sized mesh, η approaches an asymptotic limit $\eta_{\text{asympt}} = 1/\sqrt{N_p}$ when the particles achieve complete spatial randomness, i.e., the homogenous mixing [148]. As the annulus is unequally meshed, the analytical η_{asympt} is unavailable. However, since the tracer particles are uniformly distributed at the complete spatial randomness, η_{asympt} can be numerically obtained by imposing a uniform distribution to the tracer particles within the annulus, which is around 2.7×10^{-3} . Hence, the calculation of η is completed when it reaches this value.

5.3 Results and Discussion

To facilitate the following discussion, the Peclet number Pe is defined as

$$Pe = \frac{U_i R_i}{D}, \quad (5.64)$$

which indicates the relative effect of fluid transport due to the advection and diffusion. The area of the annulus is defined by the radius ratio R_r . Mixing is faster at a larger radius ratio R_r , which is reasonable since a larger R_r represents a smaller annulus space. In the following study, the Peclet number $Pe = 100$ and the radius ratio $R_r = 0.1$.

5.3.1 Typical mixing process

To analyse the mixing behaviour of the Lagrangian chaos, the variation of the mixing index η with the dimensionless time t/t_0 , the particle distributions at different times, and the Poincaré sections are shown in Figure 5.3.

The Poincaré section is a good tool to visualise the chaotic nature of mixing, which is created by recording the particle positions at the end of every time period and presenting them in one figure [149]. Generally, a random distribution of dots in the Poincaré section indicates a chaotic state, whereas the well-defined curves, i.e., the Kolmogorov-Arnold-Moser (KAM) curves [149], represent a regular state. In the random regions, the tracer particles can arrive at any position within the same region regardless of their initial locations. Contrarily, the KAM curves indicate a periodic motion of the tracer particles. The regions encircled by the KAM curves indicate a periodic motion or contain both regular and chaotic regions. However, the tracer particles initially released at any point of a region (regular or chaotic) can never cross the KAM curve to enter another region because the KAM curves act as walls preventing the fluid mixing among the separated regions when diffusion is not considered.

Figure 5.3: Variation of the mixing index η with the dimensionless time t/t_0 for mixing realised by the Lagrangian chaos of (a) the EO and (b) the ICEO. The dashed horizontal line $\eta_{\text{asympt}} = 2.7 \times 10^{-3}$ indicates the homogenous mixing. Here the eccentricity $\varepsilon = 0.5$; the dimensionless time period $T/t_0 = 20, 10$ for the Lagrangian chaos of the EO and the ICEO, respectively; the number of flow patterns $N_{fp} = 2$; and the electric field phase angle $\theta_0 = 0, \pi/2$ and $\theta_0 = 0, \pi/4$ for the Lagrangian chaos of the EO and the ICEO, respectively. The Poincaré sections are created by releasing 10 tracer particles into the flow with 10,000 time periods recorded. Note: the values of parameters are same in other figures unless otherwise noted.

The Poincaré sections in Figure 5.3 are created by releasing 10 tracer particles into the flow with 10,000 time periods recorded. Clearly, both the random regions and the KAM curves appear in the Poincaré sections. The random regions demonstrate that the chaotic behavior of the tracer particles can be induced by periodically alternating either the EO or the ICEO. As the major area of the annulus is occupied by the random regions, a good mixing can be expected for the Lagrangian chaos created by either the EO or the ICEO .

Figure 5.3 shows that the mixing index η of both the EO and the ICEO reduces gradually from 1 and approaches the constant value $\eta_{\text{asympt}} = 2.7 \times 10^{-3}$, where the homogenous mixing is obtained. Clearly, a homogeneous mixing is achievable by the Lagrangian chaos created through either the EO or the ICEO. The particle distributions at different times in Figure 5.3 demonstrate that the tracer particles are initially located in a single bin ($t/t_0 = 0$), then dispersed by the Lagrangian chaos ($t/t_0 = 20, 100$) and eventually distributed homogeneously ($t/t_0 = 500$).

As introduced previously, the time scales of the Lagrangian chaos created by

the EO and the ICEO are $\mu R_i/(\varepsilon_f \zeta_c E_0)$ and $\mu/(\varepsilon_f E_0^2)$, respectively. The electric field of order 10^4 V/m is commonly used in the EO [150] and the ICEO [29]. Given the inner cylinder radius $R_i = 10 \mu\text{m}$, the zeta potential of the non-conducting inner cylinder $\zeta_c = 20$ mV, the fluid viscosity $\mu = 1 \times 10^{-3}$ kg/(m·s), the dielectric permittivity of fluid $\varepsilon_f = 7 \times 10^{-10}$ kg·m/(V²·s²), and the electric field strength $E_0 = 1 \times 10^4$ V/m, the time scales of the Lagrangian chaos created by the EO and the ICEO are 50 ms and 10 ms, respectively. Thus, the homogenous mixing can be achieved by the Lagrangian chaos of the EO and the ICEO within 15 s and 3 s (at $t/t_0 = 300$ in Figure 5.3), respectively. While when E_0 is increased to 2×10^4 V/m, the time scales are reduced to 25 ms and 2.5 ms, and the homogenous mixing are realized within 7.5 s and 0.75 s, respectively. As the applied electric field increases, the time needed for a homogenous mixing through the Lagrangian chaos of either the EO or the ICEO can be significantly reduced. The time consumption of the ICEO will be much shorter than that of the EO under a large electric field.

5.3.2 Mixing effectiveness validation

To illustrate the effectiveness of the Lagrangian chaos for mixing, the variation of the mixing index η with time t/t_0 for the mixing achieved by several ways is shown in Figure 5.4. The EO and the ICEO are simulated by considering both the advection and the diffusion with the electric field phase angle $\theta_0 = 0$. The flow fields are shown in Figure 5.2. The curves of the mixing index η clearly show that the Lagrangian chaos, created by either the EO or the ICEO, produces the best and fastest mixing (homogeneous mixing realized around $t/t_0 = 300$). The mixing realized by diffusion is the slowest, and the EO achieves homogeneous mixing ($t/t_0 = 440$) earlier than the ICEO. In the EO and the ICEO, the tracer particles are transported by two mechanics, advection and diffusion. Advection moves the particles along the streamlines, while diffusion disperses them in different directions including across the streamlines. Thus, the tracer particles can enter one microvortex from the other microvortex through diffusion. The microvortices in the

ICEO are confined at the four sides of the annulus (Figure 5.2(b)). Hence, it is not easy to disperse the tracer particles from one side of the cylinder to the other side. While a microvortex in the EO is large and encircles the inner non-conducting cylinder at $\theta_0 = 0$ (Figure 5.2(a)). The tracer particles can be advected from one side of the cylinder to the other side. With the help of diffusion, the tracer particles can be dispersed to the other microvortex and eventually homogenously distributed. Therefore, the EO achieves a homogenous mixing within a shorter time than the ICEO at $\theta_0 = 0$. However, one should note that when the electric field phase angle θ_0 changes, a longer time may be needed for the EO flow, such as $\theta_0 = \pi/2$ (Figure 5.2(a)). At $\theta_0 = \pi/2$, the microvortices no longer encircle the cylinder, hence cannot advect the tracer particles effectively. The mixing performance of the EO presents a large variation as θ_0 changes, while that of the ICEO is more stable and insensitive to θ_0 . As the four microvortices are always located around the conducting inner cylinder (Figure 5.2(b)) and the transportation of the tracer particles from one microvortex to the other microvortex is solely due to diffusion.

Figure 5.4: Variation of the mixing index η with the dimensionless time t/t_0 , where the mixing is realised through the diffusion, the ICEO, the EO, the Lagrangian chaos created by the ICEO, and the Lagrangian chaos created by the EO. Here the electric field phase angle $\theta_0 = 0$ for the EO and the ICEO, which are simulated by considering both the advection and the diffusion.

5.3.3 Effect of the eccentricity ε

How the eccentricity ε influences the behaviour of the Lagrangian chaos is of great concern since the present eccentric annulus is more practical in reality. To evaluate the influence of ε on the mixing behaviour, the variation of mixing index at different eccentricities is presented in Figure 5.5. It shows that the eccentricity ε exerts an important impact on the mixing behaviour. As the eccentricity ε increases, the mixing effect reduces. Longer time is needed for homogenous mixing when ε is larger than 0.5. Especially when ε increases to 0.9, the mixing is greatly worsened. However, in the range $0 \sim 0.5$, homogenous mixing can be achieved by the Lagrangian chaos created with either the EO or the ICEO before t/t_0 reaches 500. The difference of the mixing effects at these eccentricities ε is negligible. Clearly, the created Lagrangian chaos works robustly and effectively within a broad scope of eccentricity, ranging from 0 to 0.5.

Figure 5.5: Variation of the mixing index η with the dimensionless time t/t_0 at different eccentricities ε , where the mixing is realised through the Lagrangian chaos created by (a) the EO and (b) the ICEO.

5.3.4 Effect of the time period T

The Lagrangian chaos is created by periodically alternating the EO or the ICEO, thus, the alternating time period T is a key factor influencing the mixing behaviour. To analyse the influence of T on the Lagrangian chaos created by the EO or the

ICEO in the present model, Figure 5.6 shows the variation of mixing index for the EO and the ICEO.

Figure 5.6: Variation of the mixing index η with the dimensionless time t/t_0 at different time periods T/t_0 , where the mixing is realised through the Lagrangian chaos created by (a) the EO and (b) the ICEO.

Clearly, as time t/t_0 elapses, the mixing indices η at different T/t_0 decrease significantly for both the EO and the ICEO. Figure 5.6(a) shows that the Lagrangian chaos of the EO presents the poorest mixing at $T/t_0 = 5$. The mixing index η approaches η_{asympt} at $t/t_0 = 300$ when $T/t_0 = 10, 20$ and 100 (Figure 5.6(a)). For the Lagrangian chaos of the ICEO, the poorest mixing is shown at $T/t_0 = 100$ (Figure 5.6(b)). In general, the mixing performance of the Lagrangian chaos of either the EO or the ICEO is better when the time period T/t_0 is in the range of $10 \sim 20$. Moreover, the Lagrangian chaos of the ICEO is relatively less sensitive to the time period T/t_0 than that of the EO.

5.3.5 Effect of the number of flow patterns N_{fp}

The Lagrangian chaos could be created by periodically alternating either two or more closed-orbit flows. Thus, the influence of the number of flow patterns N_{fp} in one time period on the Lagrangian chaos and the corresponding mixing behavior remains great concern. The electric field phase angles θ_0 of the EO and the ICEO utilized for the Lagrangian chaos creation are listed in Table 5.1.

To quantitatively illustrate the influence of N_{fp} on the mixing, the variation of

Table 5.1: The electric field phase angles θ_0 of the flow patterns utilised for Lagrangian chaos creation at different numbers of flow patterns N_{fp} .

N_{fp}	θ_0 of the EO	θ_0 of the ICEO
2	$0, \pi/2$	$0, \pi/4$
3	$0, \pi/2, \pi$	$0, \pi/4, \pi/2$
4	$0, \pi/2, \pi, 3\pi/2$	$0, \pi/4, \pi/2, 3\pi/4$
5	$0, 2\pi/5, 4\pi/5, 6\pi/5, 8\pi/5$	$0, \pi/5, 2\pi/5, 3\pi/5, 4\pi/5$

the mixing index η with the dimensionless time t/t_0 at different N_{fp} is presented in Figure 5.7. It is clearly illustrated that the mixing index η gradually decreases as time t/t_0 increases at all N_{fp} . However, the decline rates vary significantly as N_{fp} increases. η reaches η_{asympt} much faster at a smaller N_{fp} . $N_{fp} = 2$ shows good mixing performance, while $N_{fp} = 4$ and 5 show that an increase in N_{fp} reduces the mixing performance. The Lagrangian chaos of the EO shows good mixing at $N_{fp} = 2$ and 3 (Figure 5.7(a)), while that of the ICEO only performs well at $N_{fp} = 2$ (Figure 5.7(b)).

Figure 5.7: Variation of the mixing index η with the dimensionless time t/t_0 at different numbers of flow patterns N_{fp} , where the mixing is realized through the Lagrangian chaos created by (a) the EO and (b) the ICEO.

5.3.6 Effect of the electric field phase angle θ_0

The flow fields of the EO and the ICEO are periodic functions of the electric field phase angle θ_0 with periods 2π and π , respectively. Hence, the flow patterns of the EO or the ICEO are determined by the values of electric field phase angles θ_0 .

Section 5.3.5 shows that $N_{fp} = 2$ performs good mixing. Thus, two flow patterns are chosen to evaluate the influence of the electric field phase angle on the mixing behavior. Different electric field phase angles (θ_2) of flow pattern 2 are chosen within $0 \sim 2\pi$ and $0 \sim \pi$ for the EO and the ICEO, respectively. Here the electric field phase angle (θ_1) of flow pattern 1 is set to be 0.

Figure 5.8: Flow fields (streamlines) utilized for the Lagrangian chaos creation and Poincaré sections of the Lagrangian chaos created by (a) the EO and (b) the ICEO. The solid black lines indicate the flow pattern 1 (corresponding to $\theta_1 = 0$); the dashed blue lines indicate the flow pattern 2 (corresponding to θ_2). The flow directions are indicated by the arrows.

Figure 5.8 presents the flow fields utilized for the Lagrangian chaos creation and the Poincaré sections of the Lagrangian chaos at different electric field phase angles (θ_2) of flow pattern 2. In the flow fields, the solid black lines indicate the flow pattern 1, while the dashed blue lines indicate the flow pattern 2. The flow directions are represented by the arrows.

Figure 5.8(a) shows that the streamlines in one flow pattern intersect with the

other flow pattern. The angles of intersection at $\theta_2 = \pi/2$ and $3\pi/2$ are larger than those at $\theta_2 = \pi/4, 3\pi/4, 5\pi/4$ and $7\pi/4$. A better mixing can be expected at $\theta_2 = \pi/2$ and $3\pi/2$ compared to the others since the Poincaré sections are more random at these two values (Figure 5.8(a)). Besides the angle of intersection, the flow direction is also important. The streamlines of flow pattern 2 at $\theta_2 = \pi/4, \pi/2$ and $3\pi/4$ are identical to those at $\theta_2 = 5\pi/4, 3\pi/2$ and $7\pi/4$, respectively. But the corresponding flow directions are opposite. The Poincaré sections at $\theta_2 = \pi/4$ and $7\pi/4$ are more random compared to those at $\theta_2 = 5\pi/4$ and $3\pi/4$, respectively. This is due to the fact that the flow directions at $\theta_2 = \pi/4$ and $7\pi/4$ are along the directions of flow pattern 1, while the flow directions at $\theta_2 = 5\pi/4$ and $3\pi/4$ are along the opposite direction of flow pattern 1 (Figure 5.8(a)). Thus, the flow is strengthened at $\theta_2 = \pi/4$ and $7\pi/4$ rather than reduced at $\theta_2 = 5\pi/4$ and $3\pi/4$. Generally, the Poincaré section at $\theta_2 = \pi/2$ is more random than those at other θ_2 .

A similar trend is presented in Figure 5.8(b). The angle of intersection between the streamlines of flow pattern 1 and 2 varies as θ_2 increases. The peak values appear at $\theta_2 = \pi/4$ and $3\pi/4$. The Poincaré sections at those two values are more random compared to those at other values. The streamlines at $\theta_2 = \pi/8, \pi/4$ and $3\pi/8$ are identical to those at $\theta_2 = 5\pi/8, 3\pi/4$ and $7\pi/8$, respectively. But the corresponding flow directions are opposite as shown in the flow fields of Figure 5.8(b). As the flow directions at $\theta_2 = \pi/8, 7\pi/4$ and $\theta_2 = 5\pi/8, 3\pi/8$ are along and opposite to the direction of flow pattern 1, the Poincaré sections at $\theta_2 = \pi/8, 7\pi/4$ are more random than those at $\theta_2 = 5\pi/8, 3\pi/8$. Generally, the Lagrangian chaos at $\theta_2 = \pi/4$ and $3\pi/4$ provide better potential in mixing than those at other θ_2 .

The variation of the mixing index η with the time t/t_0 at different field phase angle (θ_2) of flow pattern 2 is shown in Figure 5.9. It is evidently illustrated that η presents different decline rates as θ_2 changes. Large difference is shown when t/t_0 is small ($t/t_0 < 200$). The variation of η with θ_2 at $t/t_0 = 500$ as insets in Figure 5.9. Clearly, as θ_2 increases, η shows a *W*-shape variation in both the EO and the ICEO. The optimum θ_2 for the Lagrangian chaos created by the EO and the

Figure 5.9: Variation of the mixing index η with the dimensionless time t/t_0 at different electric field phase angles of flow pattern 2, θ_2 , where the Lagrangian chaos is created by (a) the EO and (b) the ICEO. Here the electric field phase angle of flow pattern 1, $\theta_1 = 0$. Inset: variation of the mixing index η with the electric field phase angle of flow pattern 2, θ_2 , at the dimensionless time $t/t_0 = 500$.

ICEO are $\pi/2$ and $\pi/4$, respectively, which show good agreement with the Poincaré sections in Figure 5.8. Moreover, the Lagrangian chaos of the ICEO is less sensitive to the values of θ_2 compared to that of the EO.

5.4 Summary

This chapter presents an analytical study on the electroosmosis (EO) and the induced charge electroosmosis (ICEO) in the eccentric annulus, and proposes to enhance mixing by introducing Lagrangian chaos into the EO and the ICEO micromixers. The flow dynamics of the EO and the ICEO, and the mixing behaviour of the corresponding Lagrangian chaos in the eccentric annulus are investigated in detail. The results show that the created Lagrangian chaos performs better and faster mixing than either the pure EO or the pure ICEO. When the applied electric field is large, the Lagrangian chaos of the ICEO can realise more rapid mixing than that of the EO.

Chapter 6

Experimental Investigations of Induced Charge Electroosmosis around Conducting Cylinders

6.1 Introduction

Although many studies have been conducted on the design of ICEO systems [2], experimental investigations of ICEO have relatively been rare to date. Levitan et al. [28] first captured ICEO velocity field around a horizontal metal wire. Pascall and Squires [7, 60] measured ICEO flows over gold electrodes “contaminated” with dielectric layers. Canpolat et al. measured ICEO flow pattern around vertical metal cylinders in 10^{-3} mol/L KCl solution with [29, 31] and without [30] polymer using μ PIV. However, the effect of electrolyte concentration on ICEO flow is unclear. In this chapter, a systematic investigation is conducted on the ICEO around a gold-coated stainless steel cylinder. The influence of key parameters, ranging from electric field strength, electric field frequency, to electrolyte concentration, on the ICEO around the cylinder is analysed in detail.

6.2 Experimental Methods

6.2.1 Microchannel fabrication

The experiments were conducted in a straight rectangular poly(dimethylsiloxane) (PDMS)/glass microchannel with dimensions of 0.7 mm (height) $\times$ 10 mm (width) $\times$ 50 mm (length) (Figure 6.1). The two channel ends are connected to circular fluid

wells with diameter of 10 mm. The possible pressure-driven flow are eliminated by maintaining the same hydrostatic level in the wells. The microchannel is fabricated with soft lithographic technique [151]. The PDMS (Sylgard 184, Dow Corning) mixture is prepared with the weight ratio of prepolymer and curing agent as 10 : 1. A thin layer of PDMS mixture with thickness of 0.7 mm is poured into a container and placed on a hot plate at 80 °C. After an hour of heating, the PDMS becomes solid. The straight microchannel is constructed by cutting through the PDMS layer. This PDMS channel and a clean glass slide are treated by oxygen plasma (Harrick Plasma PDC-32G) for 45 s, then bonded immediately. Another PDMS layer is prepared similarly but with a gold-coated stainless steel cylinder of radius 175 μm vertically positioned in it. The cylinder touches the bottom of PDMS container. After an hour of heating on the hot plate, the PDMS becomes solid and the cylinder is firmly secured. This PDMS layer is bonded to the PDMS channel on the glass slide following the same procedure. Two fluid wells of diameter 10 mm are punched through in the PDMS layer. The fabricated microchannel is shown in Figure 6.1. The cylinder is vertically fixed in the microchannel and touches the bottom of the microchannel.

Figure 6.1: Microchannel and its key dimensions.

6.2.2 μPIV measurement

Spherical fluorescent particles (Fluoro-Max, Thermo Scientific) of 3.2 μm in diameter was used to measure the velocity field. The NaCl (Sigma-Aldrich) solution

is seeded with such particles at a volume fraction of 0.01 %. Tween 20 (Sigma-Aldrich) with volume fraction of 0.005% is added into the solution to avoid particle adhesion on the cylinder surface. The particle solution is stirred before injecting into the microchannel for experiments.

As shown in Figure 6.2, a sinusoidal AC electric field is created using a function generator (Agilent 33500B), and amplified by a high-voltage amplifier (Trek 5/80). The AC electric field is imposed to the microchannel through two electric wires positioned in the two wells (Figure 6.1). The distance between the two electric wires is 50 mm. The applied AC electric field is monitored by an oscilloscope (Tektronix TDS210). Experiments are conducted under conditions with different voltages, frequencies, and NaCl concentrations. Particle motion is visualised and recorded through an inverted optical microscope (Leica DM ILM) with a high resolution CCD camera (Vision Research V611). Pressure-driven flow is carefully eliminated before experiment by balancing the fluid in the two wells until the fluid inside the microchannel becomes stationary.

Figure 6.2: Experimental setup.

μ PIV is a well-known optical technique for flow field study in various areas [152]. It records the displacement of particles from sequentially taken images with certain time interval through a high resolution CCD camera. The μ PIV adopted here includes the aforementioned inverted optical microscope with the

CCD camera, a laser source (Dantec Dynamics) with wavelength of 527 nm at frequency of 1 kHz, and a computer for data acquisition and processing. The particle motion is captured at rate of 24 Hz and the images are taken by the CCD camera with resolution of 1280×800 pixels. The $2.5\times$ objective lens ($N_A = 0.07$) is used to focus on the centre plane of the microchannel, which provides a field view of 3.64×2.27 mm. DaVis 8.0 (LaVision) software is utilised to process the recorded images for the velocity vector field. In the image processing, 32×32 pixels rectangular effective interrogation windows having 50 % overlap are employed. The instantaneous particle velocities are obtained using the cross-correlation technique. 200 instantaneous sequential images are processed and leads to 200 velocity vector fields. The velocity vector field presented in the following discussion is the average vector field of these 200 sequential vector fields. Due to the overlapping of interrogation windows with the stationary cylinder surface [153], the measured velocity near the cylinder surface is always smaller than its true value.

6.2.3 Experimental Error Analysis

This section analyses possible factors that may influence the ICEO measurement. The depth of field (DOF) of the $2.5\times$ objective lens is $120\ \mu\text{m}$. In the optical measurement, the original point becomes a blur spot due to the defocusing effect when the point does not precisely locate on the focus plane. The experiment is carried out when the object is within the DOF. As the cylinder is vertically positioned in the microchannel, the DOF does not affect the cylinder image. Although the fluorescent particles are blurred, the brightest point of the fluorescent particles is always at the centre. The μPIV measurement collects particle positions based on the brightest point of particles. The DOF of the lens does not influence the particle position determination. The viscous effect on the top and bottom surfaces of the microchannel affects the ICEO velocity. However, as the ICEO flow is in parallel plane of the top and bottom surfaces of the microchannel, and the microchannel depth is $700\ \mu\text{m}$, much larger than the DOF, the influence of the

viscous effect should be small and negligible.

Brownian motion of the fluorescent particles may not be negligible since the particle diffusivity rapidly increases as its diameter decreases. The average particle displacement by Brownian motion during an time interval is $x \approx \sqrt{2D\Delta t}$ and the Brownian velocity is estimated as

$$u_B = \sqrt{\frac{2D}{\Delta t}}, \quad (6.1)$$

where the diffusivity of particles with diameter of $3.2 \mu\text{m}$ suspended in water at 25°C is

$$D = \frac{k_B T_a}{6\pi\mu R} \approx 1.33 \times 10^{-13} \text{ m}^2/\text{s}, \quad (6.2)$$

Thus, the resulted Brownian velocity is $2.50 \mu\text{m/s}$ for imaging rate of 24 Hz .

The experimental error of PIV measurement due to Brownian motion is expressed as [154]

$$\delta_B = \frac{u_B}{u} \frac{1}{\sqrt{N_{piw} \times N_{ip}}}, \quad (6.3)$$

where u is the characteristic velocity; N_{piw} is the number of particles in the interrogation window; N_{ip} is the number of image pairs for PIV processing.

The characteristic velocity of the ICEO is of order $100 \mu\text{m/s}$. 200 pairs of images are processed with at least 10 particles in each interrogation window. Hence, the Brownian error is 0.05% .

The cylinder deforms the applied electric field, which may lead to the dielectrophoresis (DEP) of the fluorescent particles. Ref. [2] shows that DEP velocity scale is

$$u_{DEP} = \frac{\varepsilon_f}{6\mu} r^2 \nabla |E|^2, \quad (6.4)$$

where r is the radius of the fluorescent particles.

The velocity scale of ICEO flow is

$$u_{ICEO} = \frac{\varepsilon_f}{\mu} E^2 R, \quad (6.5)$$

Thus, the ratio of the two velocity scales can be estimated by $u_{DEP}/u_{ICEO} \sim r^2/R^2$ so long as the electric field gradient is of order E/R , which is reasonable in the absence of sharp corners [2]. As $r = 3.2 \mu\text{m}$ and $R = 175 \mu\text{m}$, this ratio is roughly $\sim 10^{-4}$. Therefore, the experimental error due to DEP effect can be safely neglected.

ACEO occurs when two electrodes are separated with a gap at the order of μm [155]. In the study, ACEO is negligible as the distance between the two electrodes is 50 mm. The conventional electroosmosis on the microchannel wall and electrophoresis of fluorescent particles are also negligible with the operating frequency ranging from 0.8 kHz to 2.0 kHz [28]. Electrothermal flow would arise in the microchannel with nonuniform permittivity and conductivity, which is proportional to the fourth order of the applied electric field, $u_{ET} \propto E^4$ [156]. In the experiment, the measured flow field is proportional to the second order of the applied electric field (Figure 6.5), which implies that the electrothermal flow is insignificant. Hence, the measured fluid velocity represents the ICEO arising on the cylinder due to the interactions between the applied electric field and the induced EDL.

6.3 Results and Discussion

6.3.1 Effect of electric field strength

The influence of the electric field strength on the ICEO around the cylinder is discussed at the electric field frequency of 1.5 kHz and the NaCl concentration of 10^{-3} mol/L .

Figure 6.3 shows the velocity vector fields at electric field strength of (a) 300 $\text{V}_{\text{p-p}}/\text{cm}$, (b) 400 $\text{V}_{\text{p-p}}/\text{cm}$, (c) 500 $\text{V}_{\text{p-p}}/\text{cm}$, and (d) 600 $\text{V}_{\text{p-p}}/\text{cm}$. The direction of the electric field and the reference vector are shown at the top of Figure 6.3. Clearly, fluid is driven towards the cylinder along the direction of the electric field, and driven away perpendicular to the direction of the electric field. Hence, four

Figure 6.3: Velocity vector fields at electric field strength of (a) $300 \text{ V}_{\text{p-p}}/\text{cm}$, (b) $400 \text{ V}_{\text{p-p}}/\text{cm}$, (c) $500 \text{ V}_{\text{p-p}}/\text{cm}$, and (d) $600 \text{ V}_{\text{p-p}}/\text{cm}$.

vortices are generated around the cylinder. As the electric field increases, the vortices grow stronger. The velocity magnitude near the cylinder is much larger than that faraway from the cylinder. The velocity fields around the cylinder are not ideally symmetric.

To obtain a clearer picture of the ICEO flow field, Figure 6.4 presents the variation of velocity magnitude along (a) the x -axis and (b) the y -axis at different

Figure 6.4: Variation of velocity magnitude along (a) the x -axis and (b) the y -axis at different electric field strengths. Note: the x -axis and the y -axis are defined as shown in Figure 6.3 for the following discussion unless otherwise mentioned.

electric field strengths. The peak value of velocity magnitude does not occur on the cylinder surface. This is due to the under-estimation resulted from the overlapping of interrogation windows with the cylinder surface, which has been reported in Refs. [29, 153]. The velocities are not symmetric with respect to the cylinder centre. The maximum magnitude of velocity on the left-hand side of the cylinder is larger than that on the right-hand side (Figure 6.4 (a)). Similar trend is shown in Figure 6.4 (b). The maximum magnitude of velocity along the x -axis (Figure 6.4 (a)) is larger than that along the y -axis (Figure 6.4 (b)) for electric field with strengths of 300 V_{p-p}/cm , 400 V_{p-p}/cm , 500 V_{p-p}/cm , while the trend is reversed at 600 V_{p-p}/cm . This asymmetric phenomenon in ICEO flow around a metal cylinder has also been reported in Refs. [28, 29, 30]. A possible reason of such asymmetry in ICEO flow is the unstable flow due to concentration polarisation in strong electric fields. The electrical potential drop across the cylinder is of orders $40\phi_T$ and $100\phi_T$ in Refs. [28] and [29, 30], respectively, where ϕ_T is the thermal voltage. The electrical potential drop in the present experiment is $50\phi_T \sim 100\phi_T$. Ref. [8] reported that when electrical potential drop across a cylinder is larger than $30\phi_T$, the ICEO flow becomes unstable. Sub-vortices are generated on the cylinder surface as shown in Figure 2.8. Therefore, the ICEO flow around the cylinder becomes asymmetric. To systematically clarify this phenomenon, further study needs to be conducted.

Figure 6.5: Variation of the maximum fluid velocity with the square of electric field strength E^2 .

Figure 6.5 shows the variation of the maximum fluid velocity with E^2 and the corresponding linear fitting. The goodness of fitting $\chi^2 > 0.99$ suggests an excellent linear fitting. Hence, the maximum magnitude of velocity is linearly proportional to E^2 . Comparison of the experimental results with existing theoretical prediction by Squires and Bazant [1] shows that the experimental results are of an order of magnitude smaller than the prediction. This is because the model [1] was derived in the limits of thin EDL and small zeta potential assumptions. The approximation holds for thin EDL as $\lambda_D \ll R$, however, the limitation of small zeta potential cannot be applied as $\zeta_i/\phi_T = 50 \sim 100$ in the experiments. Theoretical overestimation has also been reported in Refs. [28, 29]. Ref. [29] also reported that ICEO flow around a vertical cylinder linearly increases with E^2 , but the geometric parameters of microchannel and cylinder, and the range of E are different from the present experimental study. The geometric parameters of microchannel and cylinder influence ICEO flow through the charging times of the electrodes and cylinder, which will be discussed in Section 6.3.2. Moreover, Ref. [29] used 1×10^{-3} mol/L KCl solution. Although the concentration is the same to the present study, the different ion species affect the ICEO velocity [76].

6.3.2 Effect of electric field frequency

The influence of the electric field frequency on the ICEO around the cylinder is examined at the electric field strength of $500 \text{ V}_{\text{p-p}}/\text{cm}$ and the NaCl concentration of $5 \times 10^{-4} \text{ mol/L}$.

Figure 6.6: Velocity vector fields at electric field frequency of (a) 0.8 kHz, (b) 1.2 kHz, (c) 1.8 kHz, and (d) 2.0 kHz.

The velocity vector fields at electric field frequency of (a) 0.8 kHz, (b) 1.2 kHz, (c) 1.8 kHz, and (d) 2.0 kHz are shown in Figure 6.6. Clearly, four vortices are

generated around the cylinder. The velocity field increases as the electric field frequency increases from 0.8 kHz to 1.8 kHz, while decreases as the electric field frequency further increases to 2.0 kHz, which is clearly shown in Figures 6.7 and 6.8. The decrease of fluid velocity at high electric field frequency has also been reported in Refs. [29, 72].

Figure 6.7: Variation of velocity magnitude along (a) the x -axis and (b) the y -axis at different electric field frequencies.

Figure 6.8: Variation of the maximum fluid velocity with the electric field frequency.

Figure 6.7 shows the variation of velocity magnitude along (a) the x -axis and (b) the y -axis. It can be concluded that the velocity field is not ideally symmetric with respect to the cylinder centre. The velocity magnitude along the y -axis (Figure 6.7(b)) is larger than that along the x -axis (Figure 6.7(a)). The peak value of velocity magnitude occurs around $x/R = y/R = 3.0$. As the electric

field frequency increases, the fluid velocity obtains a peak value. Such trend is demonstrated more evidently in Figure 6.8, which shows the variation of the maximum fluid velocity with the electric field frequency. The peak value of the velocity field is obtained at 1.8 kHz. Squires and Bazant [1] reported that ICEO flow can only exist in AC electric fields when the electric field frequency f ranges from τ_e^{-1} to τ_c^{-1} , where $\tau_c^{-1} = \lambda_D R/D$ and $\tau_e^{-1} = \lambda_D L/D$ are the charging times of the cylinder and the electrodes, respectively. Here λ_D is the EDL thickness; R is the cylinder radius; L is the distance between the two electrodes; and D is the mass diffusivity of ions. When f is smaller than τ_e^{-1} , stable EDLs are formed on the electrodes, which prevent the establishment of electric field across the NaCl solution. Therefore, ICEO flow cannot be generated. When f is larger than τ_c^{-1} , the cylinder does not have sufficient time to form stable EDL on its surface. Hence, ICEO flow also cannot be generated. Thus, it is reasonable that a peak ICEO velocity appears at certain electric field frequency within this range. Ref. [29] also shown that ICEO obtains peak value as electric field frequency increases. The frequency range is related to the geometric parameters, R and L . Such parameters are different in the present experiment ($R = 175 \mu\text{m}$, $2L = 5 \text{ cm}$) and Ref. [29] ($R = 335 \mu\text{m}$, $2L = 2 \text{ cm}$). Therefore, the electric field frequencies of peak ICEO velocity are different.

6.3.3 Effect of NaCl concentration

The influence of NaCl concentration on the ICEO around the cylinder is characterised at electric field strength of $400 \text{ V}_{\text{p-p}}/\text{cm}$ and electric field frequency of 1.5 kHz.

Figure 6.9 shows the velocity vector fields at different NaCl concentrations. It presents that the velocity fields are asymmetric with respect to the cylinder centre. The variation of velocity magnitude along (a) the x -axis and (b) the y -axis at different NaCl concentrations is shown in Figure 6.10. The variation of maximum fluid velocity with the NaCl concentration and the exponential fitting

Figure 6.9: Velocity vector fields at NaCl concentration of (a) 5×10^{-4} mol/L, (b) 6×10^{-4} mol/L, (c) 8×10^{-4} mol/L, and (d) 1×10^{-3} mol/L.

are illustrated in Figure 6.11. The results (Figures 6.10 and 6.11) show that as the NaCl concentration increases, the velocity field increases. The slope of the u_{max} vs NaCl concentration decreases as the NaCl concentration increases. The goodness of fitting $\chi^2 \approx 1$ shows that the velocity presents an exponential decay (increasing form) as the NaCl concentration increases. Such flow enhancement in ICEO due to ion increase has also been reported by Canpolat et al. [30]. Non-ionic, cationic and anionic polymers are added into 1×10^{-3} mol/L KCl solution. The results showed

Figure 6.10: Variation of velocity magnitude along (a) the x -axis and (b) the y -axis at different NaCl concentrations.

that ICEO flow increases with increasing concentration of cationic and anionic polymers while decreases with increasing concentration of non-ionic polymer [30]. They claimed that the decrease of ICEO flow with increasing concentration of non-ionic polymer is due to the exponential increase of fluid viscosity, while the increase of ICEO flow with increasing concentration of cationic and anionic polymers is because of the increasing ion concentration [30]. However, the polymer-containing fluids are non-Newtonian and the ionic polymers may act as surfactant, therefore, change the EDL on the cylinder.

Electroosmotic flow reduces with increasing electrolyte concentration is due to the fact that zeta potential reduces as electrolyte concentration increases. In ICEO, zeta potential is induced by the externally applied electric field [32]

$$\zeta_i = -\phi + \frac{\int_A \phi dA}{A}, \quad (6.6)$$

which shows that the increasing electrolyte concentration does not decrease the induced zeta potential ζ_i . However, Equation (6.6) also cannot explain the observed increase of ICEO flow with increasing NaCl concentration.

The induced zeta potential ζ_i is of order $67\phi_T$. Ref. [8] reported that surface conduction becomes important and leads to unstable ICEO flow when electrical potential drop across the cylinder is larger than $30\phi_T$. The possible concentration

polarisation due to the surface conduction may influence the ICEO flow in the experiment. However, the numerical simulation in Ref. [8] was conducted in dilute solution assumption, which may not be appropriate in strong electric fields. Because the steric effect of ions and the influence of electric field on the fluid viscosity may not be negligible in such strong electric fields [77, 78]. Further theoretical investigations are required to clarify this phenomenon.

Figure 6.11: Variation of the maximum fluid velocity with the NaCl concentration.

6.4 Summary

In this chapter, the ICEO around a gold-coated stainless steel cylinder in AC electric fields is experimentally investigated using μPIV . The four vortices around the cylinder is captured and analysed. The influences of electric field strength, electric field frequency, and NaCl concentration, on the ICEO velocity field are examined in detail. The results show that the ICEO velocity is linearly proportional to the square of the electric field strength, obtains a peak value as the electric field frequency increases, and increases as the NaCl concentration increases.

Chapter 7

Conclusions and Future Work

7.1 Conclusions

Induced charge electrokinetic phenomena are capable of zeta potential manipulation and vortex generation, which are desirable in micro/nanofluidics. This thesis presents theoretical and experimental investigations on the basic physics of induced charge electrokinetic phenomena and their potential applications in micro/nanofluidics. The major contributions during my PhD study are listed as follows:

1. Pair interactions in induced charge electrophoresis

A two-dimensional analytical model is established for pair interactions in induced charge electrophoresis of conducting cylinders. The thin electric double layer and weak applied electric field approximations are adopted. The governing equations of electric and flow fields are solved in bipolar coordinates with appropriate boundary conditions. The electric, flow and pressure fields are obtained and analysed. The microvortices around cylinders present potential applications for mixing in micro/nanofluidics. Moreover, such fields become asymmetric around one cylinder due to the presence of the other cylinder, which drive the cylinders into motion. The obtained cylinder velocities demonstrate that the cylinders repel and attract each other in perpendicular and parallel applied electric fields, respectively. The cylinder velocity component due to the electrostatic force is negligible compared to that due to the induced charge electrophoresis. This study helps to improve the

understanding of particle behaviour in micro/nanofluidics.

2. Pair interactions of conducting and non-conducting cylinders in uniform electric fields

Interactions between particles of same property has been widely studied. In this thesis, pair interactions between a conducting and a non-conducting cylinders in uniform applied electric fields are analytically characterized in bipolar coordinates. The study reveals that electroosmosis and induced charge electroosmosis are generated around the non-conducting and conducting cylinders, respectively. The net fluid flow due to electroosmosis and microvortices due to induced charge electroosmosis demonstrate potential applications for pumping and mixing, respectively. Furthermore, the asymmetric electric and flow fields drive the cylinders into motion: both translating and rotating in the perpendicular applied electric field, while solely migrating along the x -axis in the parallel applied electric field. The cylinder velocity component due to the electrostatic force is also negligible compared to other factors. The study can be helpful to understand how particles behave when they are suspended with particles of different properties.

3. Chaotic mixing through electroosmosis and induced charge electroosmosis

The flow fields of electroosmosis and induced charge electroosmosis in an eccentric annulus are analytically obtained in bipolar coordinates. The results reveal that two and four counter-rotating microvortices are formed in the annulus by electroosmosis and induced charge electroosmosis, respectively. Various flow patterns can be generated by applying electric field along different directions. The Lagrangian chaos is created by periodically alternating two or more such electroosmotic or induced charge electroosmotic flows. The particle tracking method is adopted to numerically analyze the mixing behaviors of the created

Lagrangian chaos. The mixing index and Poincaré section are utilised to quantitatively and qualitatively evaluate the mixing effect. The results show that homogenous mixing can be achieved rapidly through Lagrangian chaos created by either electroosmosis or induced charge electroosmosis. The key parameters influencing mixing effect are studied in detail. It is found that the system can work efficiently with large ranges of eccentricity, dimensionless alternating time period, and specific flow patterns utilized for Lagrangian chaos creation, so long as there are two flow patterns in one time period. Moreover, when the applied electric field is large, Lagrangian chaos of induced charge electroosmosis can realize homogenous mixing much more rapidly than that of electroosmosis.

This study can help to deepen the understanding of flow dynamics of electroosmosis and induced charge electroosmosis in eccentric annulus, and also the mixing behaviour of corresponding Lagrangian chaos. The systematic investigations may also offer a guidance for design and optimisation of chaotic micromixers.

4. μ PIV measurement of induced charge electroosmosis around a metal cylinder

Induced charge electroosmosis around a gold-coated stainless steel cylinder is experimentally investigated using μ PIV. The ICEO velocity vector field around the cylinder is captured. A systematic study is then conducted where the key parameters, ranging from electric field strength, electric field frequency, to NaCl concentration are examined. The results show that the ICEO velocity is linearly proportional to the square of electric field strength; obtains a peak value as the electric field frequency increases; and increases as the NaCl concentration increases.

7.2 Future Work

Based on the presented study, some recommendations are made for future research to improve the understanding of basic physics of induced charge electrokinetic phenomena and explore further potential applications in micro/nanofluidics.

1. Particle manipulations by induced charge electrophoresis

The models presented in Chapters 3 and 4 can be extended to three-dimensional and/or AC electric fields. Moreover, experimental investigations can be conducted to verify the theoretical models. Besides the fundamental study, induced charge electrophoresis can also be utilised to manipulate conducting particles in suspension, e.g., controlling deposition patterns in nanofluid droplet drying.

2. Induced charge electrophoresis of cylinder in cylindrical pore

The model presented in Chapter 5 can be further extended to AC electric fields, and the induced charge electrophoresis of the inner conducting cylinder. Furthermore, the induced charge electrokinetic phenomena within the eccentric annulus would also be of great interest when the inner and the outer cylinders are leaky dielectric.

3. Chaotic mixing by induced charge electroosmosis

Further investigations on chaotic mixing using ICEO can be conducted experimentally utilising micro particle tracking velocimetry (μ PTV) to produce Poincaré sections so as to prove the Lagrangian chaos and analyse its mixing effect. Three-dimensional ICEO flow in the annulus could be studied numerically and experimentally.

4. Electrolyte dependence in induced charge electroosmosis

The literature survey shows that how the ICEO flow changes in different electrolyte solutions remains unclear, and requires further experimental investigations. Additionally, theoretical modelling on ICEO in AC electric fields requires further investigations, especially for situations of strong electric field, thick electric double layer, and/or high electrolyte concentration.

Publications Arising from The Thesis

- Huicheng Feng, Teck Neng Wong and Marcos, Pair interactions in induced charge electrophoresis of conducting cylinders, *International Journal of Heat and Mass Transfer*, 2015(88), 674-683.
- Huicheng Feng and Teck Neng Wong, Pair interactions between conducting and non-conducting cylinders under uniform electric field, *Chemical Engineering Science*, 2016(142), 12-22.
- Huicheng Feng, Teck Neng Wong, Zhizhao Che, and Marcos, Chaotic micromixer utilizing electroosmosis and induced charge electroosmosis in eccentric annulus, accepted by *Physics of Fluids*, 2016.

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