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**INTERACTION AND AGGREGATION OF COLLOIDAL BIOLOGICAL
PARTICLES AND DROPLETS IN ELECTRICALLY-DRIVEN FLOWS**

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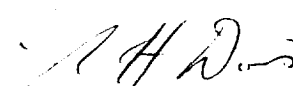
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Objectives

The primary objective of this research was to develop a fundamental understanding of aggregation and coalescence processes during electrically-driven migration of cells, particles and droplets. The process by which charged cells, particles, molecules, or drops migrate in a weak electric field is known as electrophoresis. If the migrating species have different charges or surface potentials, they will migrate at different speeds and thus may collide and aggregate or coalesce. Aggregation and coalescence are undesirable, if the goal is to separate the different species on the basis of their different electrophoretic mobilities.

Accomplishments

1. Development of a theory for electrically-driven particle aggregation

- (a) Calculations to determine the electrically-driven, near-contact motion of two particles were completed (Loewenberg and Davis, 1994). Electrically-driven, relative motion between two spherical particles depends only on their size ratio, relative position (center-to-center separation and orientation with respect to the applied electric field), and ζ -potential difference (see Figure 1). Near-contact particle motion is important because it has a large effect on electrically-driven, pairwise collision rates, and because it reveals the essential physics of the electrically-driven aggregation process. The results that we have obtained have a particularly simple, and therefore useful, form: the relative, near-contact velocity depends only on size ratio; it is proportional to the ζ -potential difference and the gapwidth separating the particle surfaces. A closed-form, analytical solution has been obtained for equisized particles and for the opposite, disparate-sized extreme. Between these complementary regimes, a numerically determined function of size ratio was determined. The results obtained show that electrically-driven relative motion is much more efficient than its gravity-driven analog (see Figure 2).
- (b) Calculations for determining electrically-driven, pairwise aggregation rates were also completed (Nichols, Loewenberg, and Davis, 1994); the foregoing results for near-contact, particle motion were incorporated. Stability criteria and pairwise aggregation rates were calculated for electrophoretic motion of nonBrownian, colloidal spheres with differing zeta potentials and thin, unpolarized electric double-layers. Aggregation rates were obtained by a trajectory analysis, and stability criteria were determined by analyzing the near-contact motion between two particles. The results predict that a given suspension is considerably less stable against aggregation during electrophoresis than during sedimentation (Figure 3), and that the aggregation efficiencies for electrophoresis exceed those for buoyancy-driven motion by an order of magnitude (Figure 4). These results are explained in terms of the relatively weak interaction between well-separated particles during electrophoresis and the electrically-driven withdrawal of fluid between closely spaced particles in an electric field. Electrophoretic and buoyancy-driven aggregation rates depend primarily on a dimensionless driving force and the size ratio of the particles; the stability of a colloidal suspension depends on four additional parameters that describe the colloidal forces that act between the particles. Further modeling of combined gravitational and electrophoretic aggregation revealed a "collision-forbidden" region of parameter space for antiparallel alignment of gravitational and electrophoretic motion, for which aggregation is prevented (Wang *et al.*, 1997).

2. Theoretical description of electrically-driven droplet motion

- (a) Electrophoretic motion has been analyzed for a spherical drop covered with a nondiffusing, insoluble ionic surfactant. A moderate ζ -potential and a thin unpolarized double layer are assumed. The charge distribution is obtained by a balance between tangential electric and hydrodynamic forces. The surface charge distribution is characterized by a field parameter which is the ratio of the external electric field to the field within the double layer. The migration velocity of the drop is a nonlinear function of the field strength parameter.

Solutions of the coupled hydrodynamic and electrostatic problems were obtained using expansions in scalar and vector spherical harmonics. The resulting set of quadratic equations for the multipolar coefficients was solved by a perturbation in the field parameter.

- (b) Electrophoretic migration and rotation has been analyzed for a spherical particle with an arbitrary surface charge distribution in a nonuniform electrostatic field. The Debye layer is described by the linearized Poisson-Boltzmann equation and is undistorted by the flow. A set of fundamental solutions of the Stokes equation has been constructed to describe the fluid flow induced by the applied electrostatic field. The solutions correspond to the flow induced by a concentric shell of force around the particle.

For a double layer of arbitrary thickness, angular and translational velocities of the particle, and the hydrodynamic traction on its surface have been calculated. In a uniform electrostatic field, the migration velocity depends on the charge monopole and quadrupole, in agreement with the result obtained earlier for thin double layers. The results may be used to analyze electrophoretic motion of drops covered with ionic surfactant and interacting particles with non-overlapping double layers.

3. Experiments to observe electrically-driven aggregation

- (a) We obtained and installed a microelectrophoresis device that has 800 $\times$ magnification capability, and built the requisite microelectrophoresis chamber. We possess a computer program that interprets raw migration velocity data, corrects for the electrosmotic background velocity, and yields electrophoretic mobilities. Fixed red blood cells from humans and rabbits which form approximately spherical particles have been obtained, and their electrophoretic mobilities have been measured as functions of ionic strength. Interestingly, the mobilities of these two cells types cross at an intermediate ionic strength.

We purchased the image processing equipment described in our proposal, including a 486 personal computer, a basic image-processing software package, and a high-quality frame grabber capable of capturing paused images; the equipment has been installed and calibrated. Microvideo experiments to observe aggregation of human and rabbit red blood cells in an electric field using this equipment were attempted, but quantification proved difficult since even moderately low concentrations (1% cells by volume) are optically opaque, whereas very dilute suspensions have very low aggregation rates. A more promising approach which we have initiated is zone electrophoresis, in which a band of higher mobility particles passes through a band of lower mobility particles, yielding heteroaggregates.

Significance

Our fundamental study of particle aggregation in electric fields will have practical application to electrically-controlled cell flocculation for cell separation and recycle in space-based bioreactors, where gravity cannot be employed as done previously. Similarly, the study on drop interactions and coalescence is expected to provide an understanding of electrically-driven demixing of two liquid phases, such as those encountered in biphasic aqueous extraction of biological cells and molecules, under reduced gravity when buoyancy-driven demixing is weak. The theoretical descriptions of two charged, migrating particles or drops, will also have broader scientific and engineering value.

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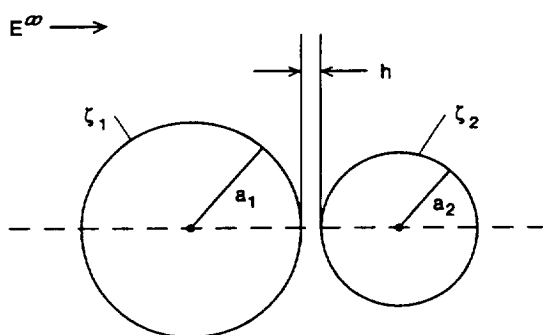


Figure 1. Schematic of two charged particles in near-contact electrophoretic motion.

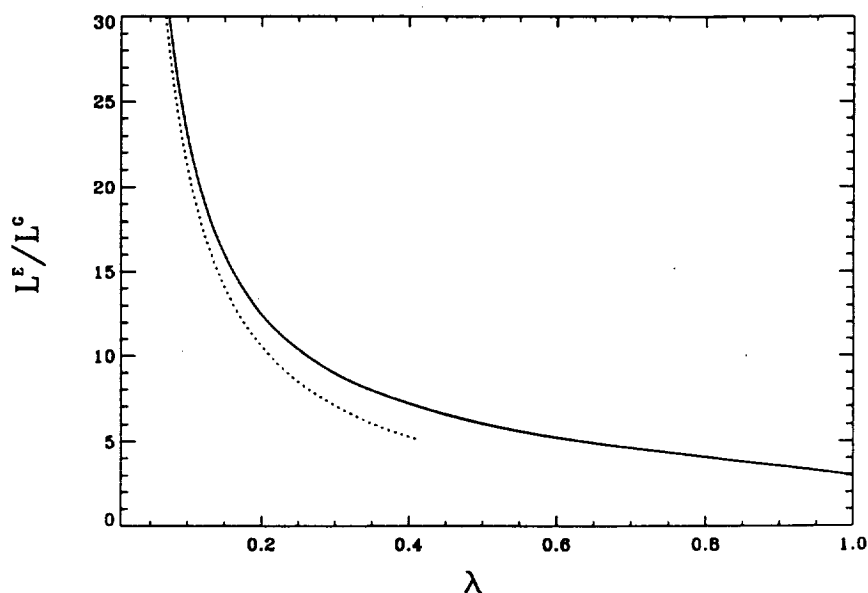


Figure 2. Ratio of electrophoretic and gravitational relative velocities of two nearly-touching particles toward one another, as a function of the particle radius ratio. The relative velocities are made dimensionless with the relative velocities of widely-separated particles; the dotted line is from an asymptotic analysis for small size ratios.

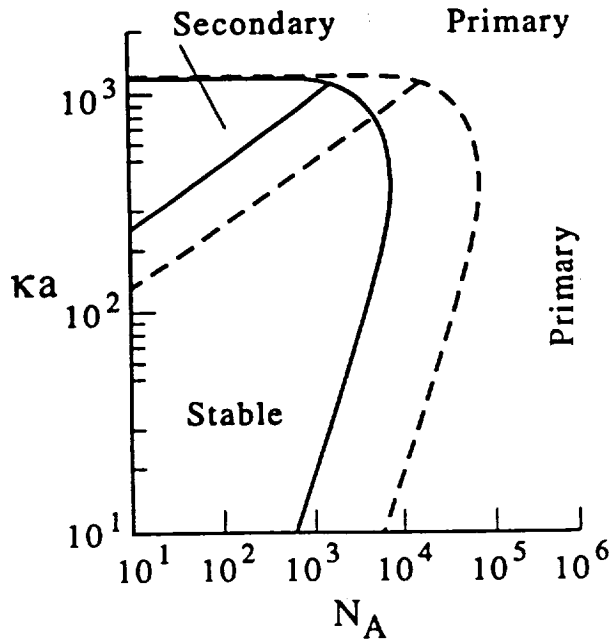


Figure 3. Stability diagram for particle suspensions subject to electrophoresis (solid curves) or gravity (dashed curves), where a is the average particle radius, κ is the inverse of the ionic double-layer thickness, and N_A is the ratio of the electrophoretic or gravitational driving force to the van der Waals attractive force; stable refers to no aggregation, and primary and secondary refer to aggregation in the primary and secondary minima, respectively.

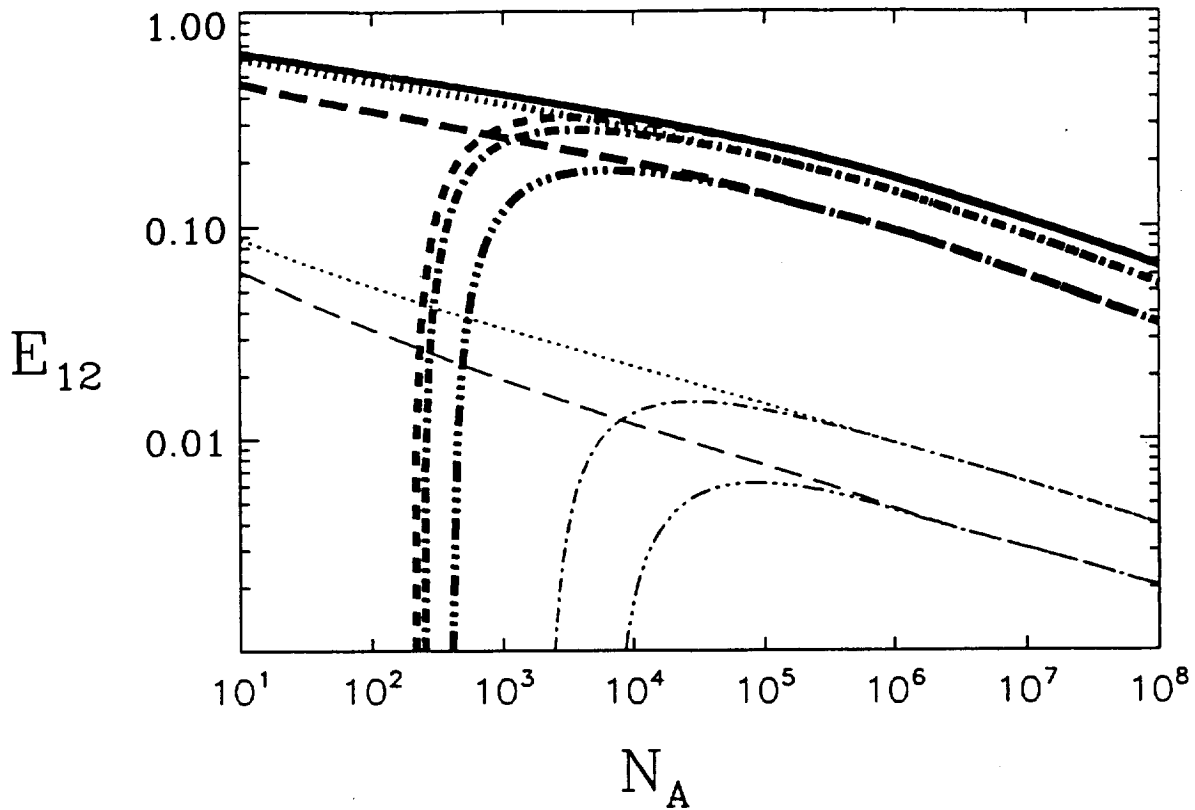


Figure 4. Collision efficiency versus dimensionless driving force for electrophoresis (thick curves) and gravitational motion (thin curves) for $\lambda = 1$ and $\kappa a = 10^2$ (short-dash curve) or 10^4 (solid curve), $\lambda = 0.5$ and $\kappa a = 10^2$ (dash-dot curves) or 10^4 (dotted curves), and $\lambda = 0.2$ and $\kappa a = 10^2$ (dash-dot-dot-dot curves) or $\kappa a = 10^4$ (long-dash curve), where λ is the particle radius ratio.