



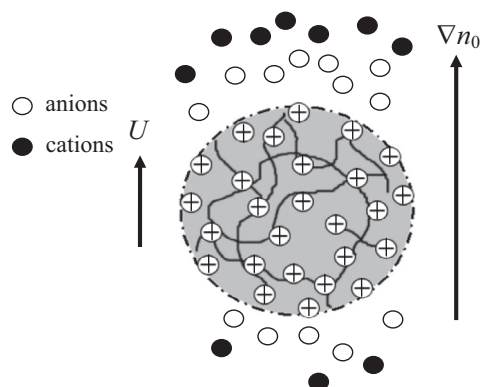
Diffusiophoretic motion of an isolated charged porous sphere



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GRAPHICAL ABSTRACT



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ABSTRACT

Diffusiophoretic motion, the migration of a colloidal particle in response to an externally applied solute concentration gradient, is investigated theoretically in this study for an isolated charged porous sphere suspended in an unbounded medium of electrolyte solution. The porous sphere is treated as a Brinkman medium with a uniformly distributed fixed charge density. The resulted general electrokinetic equations adopting the full nonlinear Poisson equation are solved numerically with a pseudo-spectral method based on Chebyshev polynomials. In particular, the convection contribution of the ion flux is taken into account properly as well.

Key parameters of electrokinetic interest are examined for their respective effect on the particle motion. The particle mobility is much smaller in general than the analytical prediction neglecting the convection-induced double layer polarization effect, which is by far the most important factor in determining the porous particle motion. A less charged particle may actually move faster than a highly charged one due to this effect. Visual demonstration of the polarization is provided. Formation of a separate axisymmetric vortex flow is responsible for the observation that a particle may reverse its direction of motion across a threshold permeability. This implies that a porous polyelectrolyte (like a protein or a DNA) assuming a random coil conformation may tango back and forth as it makes gyrations in diffusiophoretic motion.

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1. Introduction

The migration of a colloidal particle in response to an externally applied concentration gradient of the solute in solution (electrolyte or non-electrolyte), known as diffusiophoresis [1], is one of the

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fundamental electrokinetic phenomena with promising potential in various practical applications nowadays, especially in the microfluidic operations [2–7]. Compared with the conventional driving forces like electroosmosis and electrophoresis, diffusiophoresis has several obvious advantages. For instance, it can avoid undesired Joule heating effect [2], which tends to set up an unwanted convection flow. More importantly, it does not require an active external energy input. The size/weight of the corresponding arrangement may prevent its use in real portable complicated lab-on-a-chip devices [4,7]. As demonstrated by Abecassis et al. [4,5] recently, significant focusing effect leading to desirable high resolution can be easily obtained with a simple set-up utilizing diffusiophoresis. In addition, utilizing calcium carbonate particles of roughly $\sim 10 \mu\text{m}$ in size as a micropump as they dissolve in water to produce ions that migrate into the bulk solution [7], up to $40 \mu\text{m/s}$ particle speed is observed originating from the diffusiophoresis mechanism. Moreover, it can be used in connection with DNA translocation and entrapment [8], colloidal transport [4,6,9], and the enhancement of microporous membrane performance [10].

Diffusiophoresis was first discovered by Aitken [11] in 1883 for gases systems, and the corresponding observation in liquid systems was reported by Deryagin in 1947 [1]. Since then, many research efforts has been devoted to this interesting but lesser known phoretic motion of particles, either experimentally or theoretically [12–34]. For a colloidal particle suspended in a gas system with a concentration gradient, the kinetic theory of gases suffices to explain the particle motion as the result of the molecule-particle collision. In liquid systems with non-electrolyte solute, the unbalance of the osmotic pressure around the particle is easily understood to be responsible for the resulting particle motion. In a liquid electrolyte solution, however, the situation is much more complicated, and a general electrokinetic consideration is necessary to fully explain the particle behavior. In a binary electrolyte system, for instance, if the diffusion rate of cations and anions are different, an induced electric field is generated spontaneously as both species of ions migrating through the solution to ensure the electroneutrality of the bulk solution everywhere. This is called “diffusion potential” by some research groups as an analog to the well-known “sedimentation potential” with essentially the same argument [35]. This results in a particle motion similar to the well-known electrophoretic motion, except that the electric driving force now is of a diffusion origin. “Induced electrophoresis” may be a more appropriate name to this component pertinent to solutions with non-identical diffusion rates of ions. The other component comes from the migration of ions across the border of the electrical double layer from one side of the bulk stream to the other. This generates an uneven distribution, or deformation, of the ions within the double layer which in turn alters the electric field there spontaneously. The ultimate force balance is re-established again as the particle is set to motion to induce a hydrodynamic drag force. It was referred to as “chemiphoresis” by some researchers [14,15]. No chemical reaction, or anything of chemical nature are involved though. It is a purely physical phenomenon indeed. A possible origin of this calling may be from the thermodynamic terminology “chemical potential” ($\mu_j = \mu_{j\text{ref}} + z_j e \phi + k_B T \ln n_j$), which neglects the convection contribution in the ion flux expression when the particle is in motion. Dukhin and Deryagin [1,13] called it double layer polarization to emphasize the origin of the concentration gradient, which is appropriate but care should be taken to avoid the confusion with the convection-induced double layer polarization which has profound impact upon particle motion for highly charged particles conducting phoretic motion [24–29]. When the ions diffuse at the same rate, the former disappears (diffusion potential) and the particle motion is solely determined by the latter (double layer polarization).

Prieve and Anderson et al. [14,15] conducted a theoretical investigation of the diffusiophoresis of a spherical particle immersed in both the electrolyte and non-electrolyte solutions. They showed that the diffusiophoretic mobility in an electrolyte solution was determined essentially by three factors: the surface potential on the particle surface, the double layer thickness, and the relative diffusivity of ions, $\beta = (D_1 - D_2)/(D_1 + D_2)$, where D_1 and D_2 are respectively the diffusion coefficient of cations and anions in a binary electrolyte solution. Prieve and Roman [16] further calculated diffusiophoretic mobility with arbitrary double layer thickness, using the same numerical method as employed by O'Brien and White [36]. Lee and his co-workers recently studied the particular effect of convection-induced double layer polarization for suspensions of rigid spheres, liquid droplets, and so on [24–26]. Boundary effect of a nearby solid plane or an air–water interface were investigated as well [27,28]. They concluded that the presence of a boundary poses not only as a conventional hydrodynamic retarding force, but distorts the shape of double layer greatly hence alters the particle velocity. Meanwhile, Hsu et al. considered the boundary effect of an uncharged spherical cavity and an uncharged cylindrical pore upon a rigid sphere [31,32].

Previous studies focused on the rigid spherical particles only, in that no penetration of ions across the colloid surface is allowed. However, many biologically important macromolecules are porous in nature, such as proteins, nucleic acids, polysaccharides, DNAs and so on [37–39]. These macromolecules, or polyelectrolytes, are permeable to fluid and often treated as charged porous spheres [33,34,40–42]. Wei and Keh [33] first studied the diffusiophoretic motion of a lowly charged porous particle, where the Debye–Hückel approximation was adopted which is valid for low electric potential situations. Essentially all the polyelectrolytes of practical interest such as DNAs and proteins are highly charged, however.

The (convection-induced) polarization effect referred to here indicates the deformation of ion cloud surrounding the particle, or the double layer, due to the (convective) particle motion. As the particle moves through the electrolyte solution, the counterions within the double layer tend to be swept to the wake of the particle, leading to a deformed (polarized) double layer in shape, which generates an induced electric field opposite to the direction of the particle motion hence slows it down in general. This motion-detering double layer polarization effect for a porous sphere is especially profound, as a permeable particle moves much faster due to the great reduction of the hydrodynamic drag force. Without the consideration of this effect, the predictions of Wei and Keh [33] tend to overestimate the particle mobility severely for highly charged and highly permeable particles, as we will show later.

Recently, Liu et al. [34] studied the diffusiophoretic motion of a charged porous sphere within a cylindrical pore. They assumed that the boundary effect was negligible by setting the particle-pore separation distance at 12.5 particle radius. They then used this configuration to predict the motion of a single porous particle in an infinite medium. This assumption, however, is of approximation nature and reasonable only when the double layer is thin enough to avoid touching the wall of the pore. When the double layer is thick enough to touch the supposedly far away cylindrical pore, severe error in predicting the particle mobility is anticipated to take place. For instance, the famous Faxen's law [43] shows a 20% error in terms of the hydrodynamic drag force at this distance, if the presence of a cylindrical pore is neglected. For a porous sphere the error of neglecting boundary effect should be smaller though. As a result, a direct approach to a single isolated porous particle system is necessary to clarify situations. More importantly, the permeability of the porous sphere considered there is too large for a protein or DNA particle conducting free diffusiophoretic motion, as we will elaborate in the section of the Results and Discussion.

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