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Transient electrophoresis of colloidal particles in a salt-free medium

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A general theory is developed for the time dependent transient electrophoretic mobility of spherical colloidal particles in a salt-free liquid medium containing only counterions when a step external electric field is suddenly applied to the colloidal suspension. It is found that as in the case of the steady electrophoretic mobility in a salt-free medium, there is a certain critical value of the particle surface charge separating two cases, that is, the low-surface-charge case and the high-surface-charge case. In the latter case the counterion condensation takes place near the particle surface. For the low-surface charge case, the transient electrophoretic mobility agrees with that of a sphere in an electrolyte solution in the limit of very low electrolyte concentrations. For the high-surface-charge case, however, the transient mobility becomes independent of the particle surface charge because of the counterion condensation effects. A simple expression is derived for the ratio of the transient electrophoretic mobility to the steady electrophoretic mobility, which is found to take the same form irrespective of the magnitude of the particle surface charge. Using this equation, it is now possible to predict how the system will approach its final steady state.

KEYWORDS

transient electrophoresis, transient electrophoretic mobility, salt-free medium, laplace transform, step electric field

1 Introduction

The fundamental theories of electrophoresis developed in colloid and interface science have recently attracted significant attention in micro- and nano-fluidic lab-on-a-chip systems for the manipulation of liquids and colloidal particles. According to the standard theories of electrophoresis (von Smoluchowski, 1921; Hückel, 1924; Henry, 1931; Overbeek, 1943; Booth, 1950; Wiersema et al., 1966; O'Brien and White, 1978; Ohshima et al., 1983; Delgado, 2001; Dukhin and Goetz, 2001; Spasic and Hsu, 2005; Masliyah and Bhattacharjee, 2006; Ohshima, 2006; Lee, 2018), the zeta potential of colloidal particles is crucial in determining the stability of colloidal suspensions (Derjaguin and Landau, 1941; Verwey and Overbeek, 1948). Usually, the zeta potential is calculated from the electrophoretic mobility of the particles under a constant electric field, known as steady electrophoresis. In contrast, transient electrophoresis involves the non-steady motion of colloidal particles in a liquid medium when subjected to a sudden step electric field. Over time, the transient electrophoretic mobility tends to its static value. Understanding this relaxation time is vital for designing effective electrophoresis measurement systems. The theory of transient electrophoresis originated from Morison (Morrison, 1969; Morrison, 1971) and Ivory (Ivory, 1983; Ivory, 1984), was expanded by Keh and others (Keh and

Tseng, 2001; Huang and Keh, 2005; Keh and Huang, 2005; Chiang and Keh, 2015a; Lai and Keh, 2020; Li and Keh, 2020; Lai and Keh, 2021), and has been further developed by various researchers (Khair, 2012; Ohshima, 2022a; Ohshima, 2022b; Ohshima, 2022c; Ohshima, 2023b). Many studies cover different particle types and extend to transient dynamic electrophoresis and transient gel electrophoresis. A lot of theoretical studies exist on transient electrophoresis, covering various particle types such as spherical rigid particles (Morrison, 1971; Ivory, 1984; Huang and Keh, 2005; Keh and Huang, 2005; Chiang and Keh, 2015a; Lai and Keh, 2021; Ohshima, 2022a), cylindrical rigid particles (Morrison, 1971; Li and Keh, 2020; Ohshima, 2022b), porous particle (Chiang and Keh, 2015b; Lai and Keh, 2020; Yu and Keh, 2024), soft particles (Ohshima, 2022c) and particles with a slip surface (Ohshima, 2023b). Beyond the aforementioned transient free-solution electrophoresis theories, there are also theories on transient gel electrophoresis, which deal with the transient electrophoretic behavior of colloidal particles (Ivory, 1984) in a polymer gel medium (Saad, 2018; Saad and Faltas, 2018; Saad, 2019; Sherief et al., 2021; Ohshima, 2023c; Ragab, 2023; Ayman et al., 2024). In addition, there are theories on the transient dynamic electrophoresis, which refers to the transient dynamic electrophoresis of colloidal particles when a dynamic electric field is suddenly applied (Ohshima, 2023a; Ohshima, 2024a). Further, there are theories on transient electrophoresis of colloidal particles in a multi-particle suspension (Chiang and Keh, 2015a; Ohshima, 2024b).

The above-mentioned theories of transient electrophoresis deal with colloidal particles in an electrolyte solution, or, in a salt-containing medium. When colloidal particles are suspending in a liquid medium containing only counterions, that is, in a salt-free liquid medium, a remarkable phenomenon called the counterion condensation effect occurs, especially in the case of highly charged colloidal particles (Ohshima, 2002a; Ohshima, 2002b; Ohshima, 2003a; Ohshima, 2003b; Carrique et al., 2010; Delgado et al., 2016; Luo and Keh, 2021). Chang (Chang, 2009; Chang, 2010; Chang, 2012) presented theoretical studies on the transient electroosmotic flow of a salt-free solution in a cylindrical microcapillary and found that the counterion condensation effect occurs when the microcapillary wall is highly charged.

In the present paper we focus on the transient electrophoresis in a salt-free medium. In micro- and nano-fluidic lab-on-a-chip systems, the importance of understanding the electrophoresis theory of charged colloidal particles in a salt-free medium, compared to salt-containing systems, lies in the precision and control it offers. Salt-free conditions eliminate interference from electrolyte ions, enabling more accurate manipulation of particle behavior within confined channels. This advanced control is crucial for applications such as separation, sorting, and sensing within lab-on-a-chip platforms, where even small variations in mobility of colloidal particles can have a significantly impact on their performance. Moreover, insights gained from studying salt-free electrophoresis contribute to the development of novel techniques for precise particle manipulation and analysis in micro- and nano-fluidic devices, advancing their capabilities in various fields such as biotechnology, medicine, and environmental monitoring. We derive here a simple closed form expression for the time dependent transient electrophoretic mobility

of spherical colloidal particles suspended in a salt-free medium when a step external electric field is suddenly applied to the colloidal suspension.

2 Theory

Let us consider a suspension of spherical colloidal particles of radius a , mass density ρ_p , and surface charge density σ or total surface charge $Q = 4\pi a^2 \sigma$ in a salt-free liquid medium of relative permittivity ϵ_r , mass density ρ_0 , and viscosity η containing only counterions. We employ Kuwabara's cell model for a concentrated suspension of colloidal particles (Kuwabara, 1959). In addition to Kuwabara's cell model, Happel's cell model (Happel, 1958) is also known. The reason for adopting Kuwabara's model is that in the steady electrophoresis problem, Kuwabara's cell model correctly yields the Smoluchowski mobility equation in the limit of the dilute case. We thus assume that each spherical particle is surrounded by a spherical free volume of radius R , within which counterions are distributed so that electroneutrality is satisfied. The particle volume fraction ϕ is given by

$$\phi = \left(\frac{a}{R}\right)^3 \quad (1)$$

We treat the dilute case, $\phi \ll 1$. Let the valence of the counterions and the average concentration (number density) in the absence of the applied electric field be $-z$ and n_0 , respectively. Then we have

$$Q = \frac{4}{3}\pi(R^3 - a^3)zen_0 \quad (2)$$

where e is the elementary electric charge. Eq. 2 is the electroneutrality condition. The right-hand side of Eq. 2 is the total amount of charge resulting from the counterions in the spherical free volume with a radius R .

We suppose that at time $t = 0$, a step electric field $E(t)$ is suddenly applied to the particle suspension, viz.,

$$E(t) = \begin{cases} 0, & t = 0 \\ E_0, & t > 0 \end{cases} \quad (3)$$

where E_0 is constant. Then the particles start to move with a time-dependent transient electrophoretic velocity $U(t)$ in the direction parallel to E_0 . We define the transient electrophoretic mobility $\mu(t)$ of the particle as $U(t) = \mu(t)E(t) = \mu(t)E_0$. The origin of the spherical polar coordinate system (r, θ, ϕ) is held fixed at the center of the representative particle and the polar axis ($\theta = 0$) is set parallel to E_0 . Let λ_c be the drag coefficient of the counterions. The main assumptions in our analysis are as follows. (i) The applied field $E(t)$ is weak so that $U(t)$ is proportional to $E(t)$ and terms of higher order in $E(t)$ may be neglected. (ii) The applied electric field $E(t)$ is weak so that $U(t)$ is proportional to $E(t)$ and terms of higher order in $E(t)$ may be neglected so that the convection term $\rho_0(\mathbf{u} \cdot \nabla)\mathbf{u}$ involving the square of the liquid velocity $\mathbf{u}(r, t)$ in the Navier-Stokes equation can be neglected. (iii) The slipping plane (at which the liquid velocity relative to the particle becomes zero) is located on the particle surface (at $r = a$). (iv) Counterions cannot penetrate the particle surface. (v) The fluid vorticity ω is zero at the outer surface of the free volume, following Kuwabara's model (Kuwabara, 1959).

The fundamental electrokinetic equations for the flow velocity $\mathbf{u}(\mathbf{r}, t)$ of the liquid at position \mathbf{r} and time t and that of the counterions $\mathbf{v}(\mathbf{r}, t)$ are similar to those for the dynamic electrophoresis of spherical colloidal particles in a salt free medium (Ohshima, 2003b), viz.,

$$\rho_0 \frac{\partial}{\partial t} \{\mathbf{u}(\mathbf{r}, t) + \mathbf{U}(t)\} - \eta \nabla^2 \mathbf{u}(\mathbf{r}, t) + \nabla p(\mathbf{r}, t) + \rho_c(\mathbf{r}, t) \nabla \psi(\mathbf{r}, t) = \mathbf{0} \quad (4)$$

$$\nabla \cdot \mathbf{u}(\mathbf{r}, t) = 0 \quad (5)$$

$$\mathbf{v}(\mathbf{r}, t) = \mathbf{u}(\mathbf{r}, t) - \frac{1}{\lambda_c} \nabla \mu_c(\mathbf{r}, t) \quad (6)$$

$$\frac{\partial n(\mathbf{r}, t)}{\partial t} + \nabla \cdot \{n(\mathbf{r}, t) \mathbf{v}(\mathbf{r}, t)\} = 0 \quad (7)$$

$$\rho_c(\mathbf{r}, t) = -zen(\mathbf{r}, t) \quad (8)$$

$$\mu_c(\mathbf{r}, t) = \mu_c^0 - ze\psi(\mathbf{r}, t) + kT \ln n(\mathbf{r}, t) \quad (9)$$

$$\Delta \psi(\mathbf{r}, t) = -\frac{\rho_c(\mathbf{r}, t)}{\epsilon_r \epsilon_0} \quad (10)$$

$$\frac{4\pi a^3}{3} \rho_p \frac{d\mathbf{U}(t)}{dt} = \mathbf{F}_H(t) + \mathbf{F}_E(t) \quad (11)$$

where $p(\mathbf{r}, t)$ is the pressure, $\rho_c(\mathbf{r}, t)$ is the charge density resulting from the counterions of concentration (number density) $n(\mathbf{r}, t)$, $\psi(\mathbf{r}, t)$ is the electric potential outside the sphere, $\mu_c(\mathbf{r}, t)$ is the electrochemical potential of the counterions, μ_c^0 is a constant term in $\mu_c(\mathbf{r}, t)$, which corresponds to the standard electrochemical potential of counterions, $\mathbf{F}_H(t)$ and $\mathbf{F}_E(t)$ are, respectively, the hydrodynamic and electric forces acting on the particle, ϵ_0 is the permittivity of a vacuum, k is the Boltzmann constant, and T is the absolute temperature. Eq. 5 are the Navier-Stokes equation and the equations of continuity for an incompressible flow. Eq. 6 expresses that the flow $\mathbf{v}(\mathbf{r}, t)$ of the counterions is caused by the liquid flow $\mathbf{u}(\mathbf{r}, t)$ and the gradient of the electrochemical potential $\mu_c(\mathbf{r}, t)$ given by Eq. 9 of the counterions. Eq. 7 is the continuity equation for the counterions. Eqs 10, 11 and are, respectively, the Poisson equation and the equation of motion of the particle.

The initial condition for $\mathbf{u}(\mathbf{r}, t)$ is given by

$$\mathbf{u}(\mathbf{r}, t) = \mathbf{0} \quad \text{at } t = 0 \quad (12)$$

The slipping plane (at which the liquid velocity $\mathbf{u}(\mathbf{r}, t)$ relative to the particle is zero) is assumed to be located on the particle surface (at $r = a$), viz.,

$$\mathbf{u}(\mathbf{r}, t) = \mathbf{0} \quad \text{at } r = a \quad (13)$$

According to Kuwabara's cell model (Kuwabara, 1959), we assume that at the outer surface of the unit cell ($r = R$) the liquid velocity is parallel to the electrophoretic velocity $\mathbf{U}(t)$ of the particle, viz.,

$$\mathbf{u}(\mathbf{r}, t) \cdot \hat{\mathbf{n}} = -U(t) \cos \theta \quad \text{at } r = R \quad (14)$$

where $\hat{\mathbf{n}}$ is the unit normal outward from the particle surface, $U(t)$ is the magnitude of $\mathbf{U}(t)$, and that the vorticity $\boldsymbol{\omega}(\mathbf{r}, t)$ is zero at $r = R$,

$$\boldsymbol{\omega}(\mathbf{r}, t) = \nabla \times \mathbf{u}(\mathbf{r}, t) = \mathbf{0} \quad \text{at } r = R \quad (15)$$

Also, we assume that on the outer surface of the unit cell ($r = R$) the gradient of the electric potential is parallel to the applied field $\mathbf{E}(t)$, viz.,

$$\nabla \delta \psi(\mathbf{r}, t) \cdot \hat{\mathbf{n}} = -E(t) \cos \theta \quad \text{at } r = R \quad (16)$$

where $E(t)$ is the magnitude of $\mathbf{E}(t)$. Finally, we adopt the boundary condition that no electrolyte ions can penetrate the particle surface, viz.,

$$\mathbf{v}_i(\mathbf{r}, t) \cdot \hat{\mathbf{n}} = 0 \quad \text{at } r = a \quad (17)$$

For a weak field $E(t)$ [assumption (i)], we may write

$$n(\mathbf{r}, t) = n^{(0)}(r) + \delta n(\mathbf{r}, t) \quad (18)$$

$$\psi(\mathbf{r}, t) = \psi^{(0)}(r) + \delta \psi(\mathbf{r}, t) \quad (19)$$

$$\mu_c(\mathbf{r}, t) = \mu_c^{(0)} + \delta \mu_c(\mathbf{r}, t) \quad (20)$$

$$\rho_c(\mathbf{r}, t) = \rho_c^{(0)}(r) + \delta \rho_c(\mathbf{r}, t) \quad (21)$$

where the quantities with superscript (0) refer to those at equilibrium, $\delta n(\mathbf{r}, t)$, $\delta \psi(\mathbf{r}, t)$, $\delta \mu_c(\mathbf{r}, t)$, $\delta \rho_c(\mathbf{r}, t)$ are small quantities, and $\mu_c^{(0)}$ is a constant independent of r . We assume that the equilibrium concentration $n^{(0)}(r)$ of the counterions obeys the Boltzmann distribution and the equilibrium electric potential $\psi^{(0)}(r)$ around the particle with a zeta potential ζ

$$n^{(0)}(r) = n_0 \exp\left(\frac{-ze\psi^{(0)}(r)}{kT}\right) = n_0 \exp\left(\frac{ze\psi^{(0)}(r)}{kT}\right) \quad (22)$$

and

$$\frac{d^2 \psi^{(0)}(r)}{dr^2} + \frac{2}{r} \frac{d\psi^{(0)}(r)}{dr} = -\frac{\rho_c^{(0)}(r)}{\epsilon_r \epsilon_0} \quad (23)$$

with

$$\rho_c^{(0)}(r) = -zen^{(0)}(r) = -zen_0 \exp\left(\frac{ze\psi^{(0)}(r)}{kT}\right) \quad (24)$$

Here we have set the equilibrium electric potential $\psi^{(0)}(r)$ to be zero at points where the volume charge density $\rho_c(r)$ resulting from counterions equals its average value ($-zen_0$). We define the equilibrium surface potential as $\psi^{(0)}(a) - \psi^{(0)}(R)$. The boundary conditions for $\psi^{(0)}(r)$ at the particle surface $r = a$ and at the outer surface of the free volume $r = R$ are

$$\left. \frac{d\psi^{(0)}(r)}{dr} \right|_{r=a} = -\frac{\sigma}{\epsilon_r \epsilon_0} \quad (25)$$

$$\left. \frac{d\psi^{(0)}(r)}{dr} \right|_{r=R} = 0 \quad (26)$$

Further, symmetry considerations permit us to write

$$\mathbf{u}(\mathbf{r}, t) = \left(-\frac{2}{r} h(r, t) E(t) \cos \theta, \frac{1}{r} \frac{\partial}{\partial r} (r h(r, t)) E(t) \sin \theta, 0 \right) \quad (27)$$

$$\delta \mu_c(\mathbf{r}, t) = ze\Phi(r, t) E(t) \cos \theta \quad (28)$$

$$\delta \psi(\mathbf{r}, t) = -Y(r, t) E(t) \cos \theta \quad (29)$$

Here $\Phi(r, t)$, $Y(r, t)$, and $h(r, t)$ are functions of r and t .

By substituting Eq. 27–29 into Eqs 4–7, and neglecting the products of the small quantities, we obtain the following equation for $h(r, t)$, $\phi_i(r, t)$, and $Y(r, t)$:

$$L \left[Lh(r, t) - \frac{1}{\nu} \frac{\partial h(r, t)}{\partial t} \right] = G(r, t) \quad (30)$$

$$L\Phi(r, t) - \frac{\lambda_c}{kT} \frac{\partial}{\partial t} \{\Phi(r, t) - Y(r, t)\} = \frac{dy(r)}{dr} \left\{ -z \frac{\partial \Phi(r, t)}{\partial r} - \frac{2\lambda_c}{e} \frac{h(r, t)}{r} \right\} \quad (31)$$

$$LY(r, t) = \frac{1}{\epsilon_r \epsilon_0 kT} z^2 e^2 n^{(0)}(r) \{Y(r, t) - \Phi(r, t)\} \quad (32)$$

with

$$L = \frac{\partial}{\partial r} \frac{1}{r^2} \frac{\partial}{\partial r} r^2 = \frac{\partial^2}{\partial r^2} + \frac{2}{r} \frac{\partial}{\partial r} - \frac{2}{r^2} \quad (33)$$

$$G(r, t) = -\frac{zen_0}{\eta r} \frac{dy}{dr} e^{y(r)} \Phi(r, t) \quad (34)$$

$$y(r) = \frac{ze}{kT} \psi^{(0)}(r) \quad (35)$$

$$\nu = \frac{\eta}{\rho_0} \quad (36)$$

where $y(r)$ is the scaled equilibrium electric potential, ν is the kinematic viscosity of the liquid.

The initial and boundary conditions (Eqs 12–16) and the equation of the motion of the particle (Eq. 11) can be rewritten as those for $h(r, t)$ and $Y(r, t)$ as follows.

$$h(r, 0) = \frac{\partial}{\partial r} h(r, t) \Big|_{r=0} = 0 \quad (37)$$

$$h(a, t) = \frac{\partial}{\partial r} h(r, t) \Big|_{r=a} = 0 \quad (38)$$

$$h(R, t) = \frac{RU(t)}{2E(t)} = \frac{R\mu(t)}{2} \quad (39)$$

$$Lh(r, t) \Big|_{r=R} = 0 \quad (40)$$

$$\eta \frac{\partial}{\partial r} \left[r \left(Lh(r, t) - \frac{1}{\nu} \frac{\partial h(r, t)}{\partial t} \right) \right] \Big|_{r=R} - \rho_{el}^{(0)}(R) Y(R, t) + \rho_0 R \left(1 - \phi \frac{\rho_p - \rho_0}{\rho_0} \right) \frac{d\mu(t)}{dt} = 0 \quad (41)$$

Eqs 16, 17 and give

$$\frac{\partial}{\partial r} Y(r, t) \Big|_{r=R} = 1 \quad (42)$$

$$\frac{\partial}{\partial r} \Phi(r, t) \Big|_{r=a} = 0 \quad (43)$$

The transient electrophoretic mobility $\mu(t)$ can be obtained from Eq. 39, viz.,

$$\mu(t) = \frac{2h(R, t)}{R} \quad (44)$$

Eq. 31 for $h(r, t)$ is most easily solved by using the Laplace transformation with respect to time t . We introduce the Laplace transforms $\hat{h}(r, s)$, $\hat{G}(r, s)$, $Y(r, s)$, and $\hat{\mu}(s)$ of $h(r, t)$, $G(r, t)$, $Y(r, t)$, and $\mu(t)$, respectively, which are given by

$$\hat{h}(r, s) = \int_0^\infty h(r, t) e^{-st} dt \quad (45)$$

$$\hat{G}(r, s) = \int_0^\infty G(r, t) e^{-st} dt \quad (46)$$

$$\hat{Y}(r, s) = \int_0^\infty Y(r, t) e^{-st} dt \quad (47)$$

$$\hat{\mu}(s) = \int_0^\infty \mu(t) e^{-st} dt \quad (48)$$

and the Laplace transform of Eq. 42 is

$$\hat{\mu}(s) = \frac{2\hat{h}(b, s)}{R} \quad (49)$$

The Laplace transform of Eq. 29 thus gives

$$L \left[L\hat{h}(r, s) - \frac{s}{\nu} \hat{h}(r, s) \right] = \hat{G}(r, s) \quad (50)$$

By solving Eq. 50 subject to Eqs 37–41 and using Eq. 49, we obtain the following general expression for the Laplace transform $\hat{\mu}(s)$ of the transient electrophoretic mobility $\mu(t)$:

$$\hat{\mu}(s) = -\frac{2a^2}{3S\{M(a, S) + \Gamma S\}} \left[\int_a^R \{M(a, S) - M(r, S)\} \hat{G}(r, S) dr + \frac{S\rho_{el}^{(0)}(R)\hat{Y}(R, S)}{3\eta\phi R} \right] \quad (51)$$

where

$$M(r, S) = \frac{H(r, S) - H^*(r, S)\Omega(S)}{\{1 - 3\phi S^{-1}H(a, S)\} - \{1 - 3\phi S^{-1}H^*(a, S)\}\Omega(S)} \quad (52)$$

$$M(a, S) = \frac{H(a, S) - H^*(a, S)R(S)}{\{1 - 3\phi S^{-1}H(a, S)\} - \{1 - 3\phi S^{-1}H^*(a, S)\}R(S)} = \frac{H(a, S) + \frac{2\sqrt{S}\Omega(S)}{1-\Omega(S)}}{1 - \phi - 3\phi S^{-1} \left(1 + \sqrt{S} \frac{1+\Omega(S)}{1-\Omega(S)} \right)} \quad (53)$$

$$H(r, S) = \left(1 + \sqrt{S} \frac{r}{a} \right) \exp \left[-\sqrt{S} \left(\frac{r}{a} - 1 \right) \right] + S \frac{r^3}{3a^3} \quad (54)$$

$$H^*(r, S) = \left(1 - \sqrt{S} \frac{r}{a} \right) \exp \left[\sqrt{S} \left(\frac{r}{a} - 1 \right) \right] + S \frac{r^3}{3a^3} \quad (55)$$

$$H(a, S) = 1 + \sqrt{S} + \frac{S}{3} \quad (56)$$

$$H^*(a, S) = 1 - \sqrt{S} + \frac{S}{3} \quad (57)$$

$$\Omega(S) = \frac{1 + \sqrt{S}\phi^{-1/3}}{1 - \sqrt{S}\phi^{-1/3}} \exp \left[-2\sqrt{S} (\phi^{-1/3} - 1) \right] \quad (58)$$

$$S = \frac{s}{\nu} a^2 \quad (59)$$

$$\Gamma = \frac{2(\rho_p - \rho_0)}{9\rho_0} \quad (60)$$

3 Results and discussion

Eq. 51 is the general expression Laplace transform of the transient electrophoretic mobility $\mu(t)$ of a spherical colloidal particle in a salt-free liquid medium. The transient mobility $\mu(t)$ can be obtained from Eq. 51 by the numerical inverse Laplace transformation. It can be shown that as in the case of steady electrophoresis in salt-free media, the electrophoretic mobility is determined almost solely by the second term of the right-hand side of Eq. 51 (Ohshima, 2002a; Ohshima, 2003a; Ohshima, 2003b), viz.,

$$\hat{\mu}(s) = -\frac{2a^2}{9\{M(a, S) + \Gamma S\}} \frac{\rho_{el}^{(0)}(R)\hat{Y}(R, S)}{\eta\phi R} \quad (61)$$

This term comes from the contribution of the pressure due to the counterions at the outer surface of the free volume (at

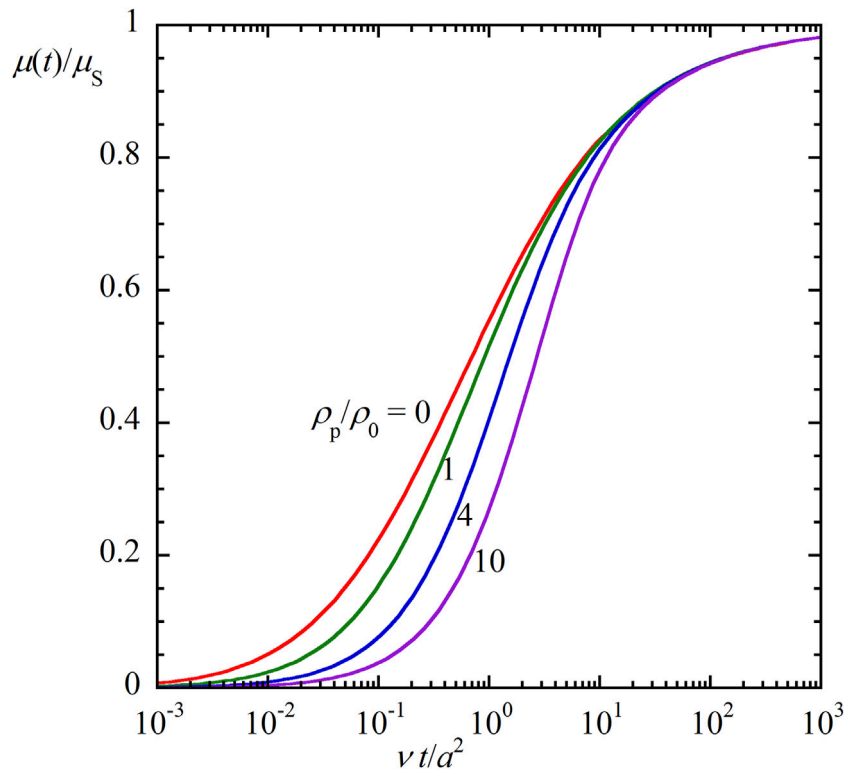


FIGURE 1
The ratio $\mu(t)/\mu_s$ of the transient electrophoretic mobility $\mu(t)$ to the steady electrophoretic mobility μ_s for spherical colloidal particles of radius a and mass density ρ_p in a salt-free liquid medium of mass density ρ_0 and kinematic viscosity ν as a function of the scaled time $\nu t/a^2$ for four values of the mass density ratio ρ_p/ρ_0 ($\rho_p/\rho_0 = 0, 1, 4,$ and 10).

$r = R$). Further, for the dilute case ($\phi \ll 1$) we may approximate $\hat{Y}(R, S)$ by R/s and $M(a, S)$ by $H(a, S)$ so that Eq. 61 becomes

$$\hat{\mu}(s) = -\frac{2a^2}{9\{H(a, S) + \Gamma S\}} \frac{\rho_{el}^{(0)}(R)}{\eta\phi s} \quad (62)$$

By substituting Eq. 25 into Eq. 43 and using Eq. 8, we have for the dilute case

$$\hat{\mu}(s) = \frac{Q}{6\pi\eta a\{H(a, S) + \Gamma S\}s} \exp\left[\frac{ze\psi^{(0)}(R)}{kT}\right] \quad (63)$$

An accurate analytic expression for the equilibrium potential distribution around a spherical particle in a salt-free medium for the dilute case ($\phi \ll 1$) has already been derived (Ohshima, 2002a). The expressions for $\psi^{(0)}(a)$ and $\psi^{(0)}(R)$ are given below.

Case 1. (low-surface-charge case)

$$\frac{Q}{4\pi\epsilon_r\epsilon_0 a} \frac{ze}{kT} \leq \ln\left(\frac{1}{\phi}\right) \quad (64)$$

is satisfied, then

$$\psi^{(0)}(a) = \frac{Q}{4\pi\epsilon_r\epsilon_0 a} \quad (65)$$

$$\psi^{(0)}(R) = 0 \quad (66)$$

If the zeta potential ζ is identified as the surface potential defined by $\psi^{(0)}(a) - \psi^{(0)}(R)$, then we have

$$\zeta = \psi^{(0)}(a) - \psi^{(0)}(R) = \frac{Q}{4\pi\epsilon_r\epsilon_0 a} \quad (67)$$

which is the Coulomb potential. That is, in the low-surface-charge case, the expression for the zeta potential ζ of a particle in a salt-free medium agrees with the limiting form of the zeta potential of a particle in an electrolyte solution with a very low electrolyte concentration.

Case 2. (high-surface-charge case)

$$\frac{Q}{4\pi\epsilon_r\epsilon_0 a} \frac{ze}{kT} > \ln\left(\frac{1}{\phi}\right) \quad (68)$$

is satisfied, then

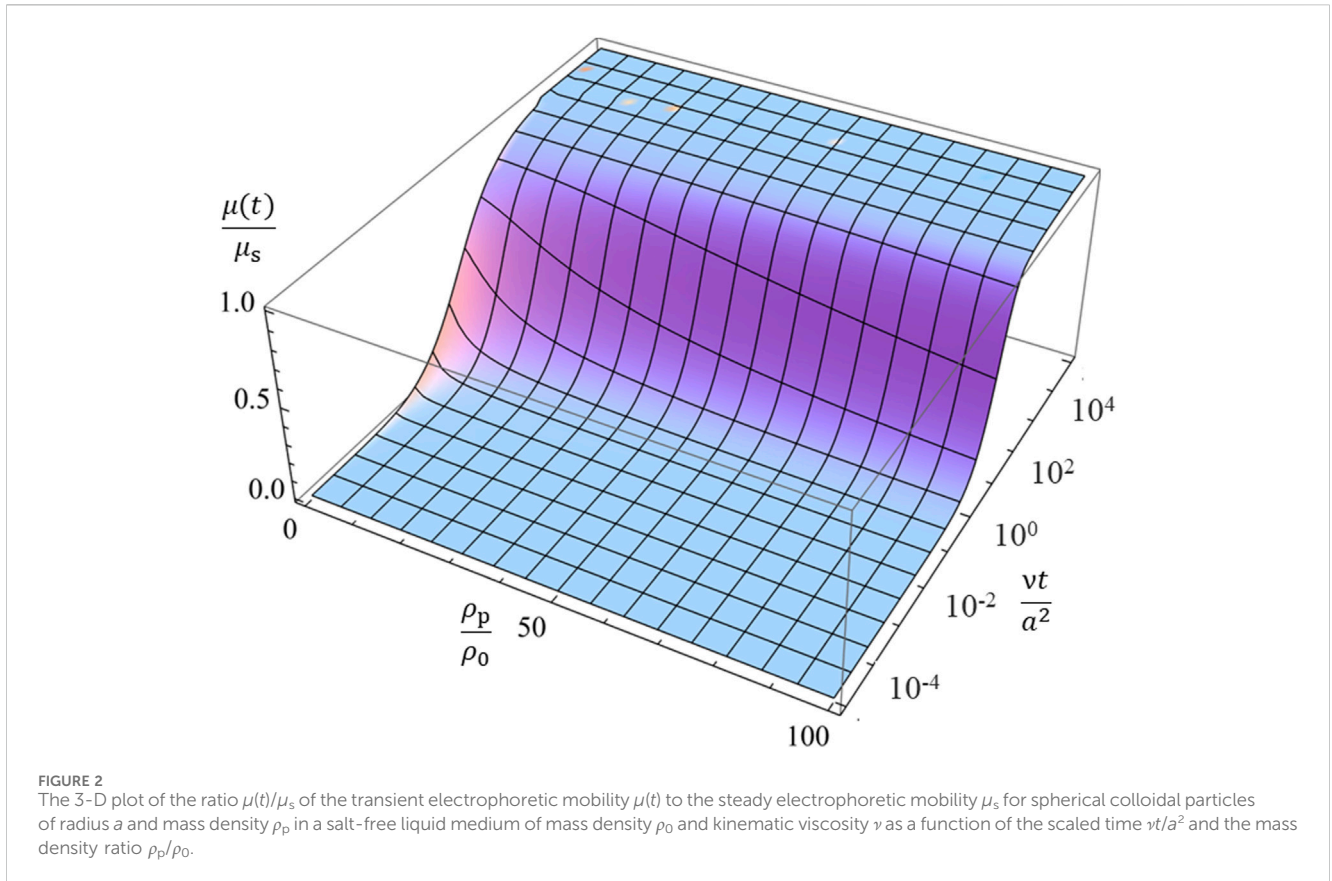
$$\psi^{(0)}(a) = \frac{kT}{ze} \ln\left[\frac{1}{6\phi} \left(\frac{ze}{kT}\right) \left(\frac{Q}{4\pi\epsilon_r\epsilon_0 a}\right)\right] \quad (69)$$

$$\psi^{(0)}(R) = -\frac{kT}{ze} \ln\left[\frac{1}{6\phi \ln(1/\phi)} \left(\frac{ze}{kT}\right) \left(\frac{Q}{4\pi\epsilon_r\epsilon_0 a}\right)\right] \quad (70)$$

from which

$$\zeta = \psi^{(0)}(a) - \psi^{(0)}(R) = \frac{kT}{ze} \ln\left[\frac{1}{6\phi \ln(1/\phi)} \left(\frac{ze}{kT}\right)^2 \left(\frac{Q}{4\pi\epsilon_r\epsilon_0 a}\right)^2\right] \quad (71)$$

Eq. 71 demonstrates that in the high-surface-charge case, unlike in the low-surface-charge case, the zeta potential ζ is not



longer given by the Coulomb potential (Eq. 67) but by a more complicated equation (Eq. 71). Eq. 71 depends less on Q than Eq. 67 and becomes a function of the particle volume fraction ϕ , as the counter ion condensation occurs in the vicinity of the particle surface.

By substituting the above results into Eq. 63, we obtain the following results:

Case 1. (low-surface- charge case)

$$\hat{\mu}(s) = \frac{Q}{6\pi\eta a \{H(a, S) + \Gamma S\} s} = \frac{2\varepsilon_r \varepsilon_0 \zeta}{3\eta} \frac{1}{\{H(a, S) + \Gamma S\} s} \quad (72)$$

Case 2. (high-surface- charge case)

$$\begin{aligned} \hat{\mu}(s) &= \frac{Q}{6\pi\eta a \{H(a, S) + \Gamma S\} s} \left(\frac{kT}{ze} \right) \ln\left(\frac{1}{\phi}\right) \\ &= \frac{2\varepsilon_r \varepsilon_0}{3\eta} \frac{1}{\{H(a, S) + \Gamma S\} s} \left(\frac{kT}{ze} \right) \ln\left(\frac{1}{\phi}\right) \end{aligned} \quad (73)$$

In the limit of $s \rightarrow 0$, Eqs 72, 73 become the steady electrophoretic mobility $\mu_s = \mu(\infty)$ at $t = \infty$ of spherical colloidal particles in a salt free medium (Ohshima, 2002a), that is,

Case 1. (low-surface- charge case)

$$\mu_s = \mu(\infty) = \lim_{s \rightarrow 0} s \hat{\mu}(S) = \frac{2\varepsilon_r \varepsilon_0 \zeta}{3\eta} \quad (74)$$

Case 2. (high-surface- charge case)

$$\mu_s = \mu(\infty) = \lim_{s \rightarrow 0} s \hat{\mu}(S) = \frac{2\varepsilon_r \varepsilon_0}{3\eta} \left(\frac{kT}{ze} \right) \ln\left(\frac{1}{\phi}\right) \quad (75)$$

The transient electrophoretic mobility $\mu(t)$ can be obtained analytically from Eqs 72, 73 by the inverse Laplace transformation. The result is

Case 1. (low-surface- charge case)

$$\mu(t) = \frac{2\varepsilon_r \varepsilon_0 \zeta}{3\eta} \left\{ \frac{q_2}{q_2 - q_1} M\left(\frac{q_1 \sqrt{\nu t}}{a}\right) - \frac{q_1}{q_2 - q_1} M\left(\frac{q_2 \sqrt{\nu t}}{a}\right) \right\} \quad (76)$$

Case 2. (high-surface- charge case)

$$\mu(t) = \frac{2\varepsilon_r \varepsilon_0}{3\eta} \left(\frac{kT}{ze} \right) \ln\left(\frac{1}{\phi}\right) \left\{ \frac{q_2}{q_2 - q_1} M\left(\frac{q_1 \sqrt{\nu t}}{a}\right) - \frac{q_1}{q_2 - q_1} M\left(\frac{q_2 \sqrt{\nu t}}{a}\right) \right\} \quad (77)$$

with

$$q_1 = \frac{9}{2\left(1 + \frac{2\rho_p}{\rho_0}\right)} \left(1 + \frac{1}{3} \sqrt{5 - \frac{8\rho_p}{\rho_0}} \right) \quad (78)$$

$$q_2 = \frac{9}{2\left(1 + \frac{2\rho_p}{\rho_0}\right)} \left(1 - \frac{1}{3} \sqrt{5 - \frac{8\rho_p}{\rho_0}} \right) \quad (79)$$

$$M(z) = 1 - e^{-z^2} \operatorname{erfc}(z) \quad (80)$$

where $\operatorname{erfc}(z)$ is the complementary error function, defined by

$$\operatorname{erfc}(z) = \frac{2}{\sqrt{\pi}} \int_z^{\infty} e^{-x^2} dx \quad (81)$$

We see that Eq. 76 for the low-surface-charge case agrees with the low κa limiting form of the transient electrophoretic mobility $\mu(t)$ of a spherical colloidal particle of radius a in an electrolyte solution of the Debye-Hückel parameter κ (Ohshima, 2022a). For the high-surface-charge case, the transient electrophoretic mobility $\mu(t)$ deviates from Eq. 76 and given by Eq. 77, which is independent of Q . In other words, in the high-surface-charge case, as Q increases, the zeta potential ζ does not increase linearly with Q (Eq. 71). This is because the number of counterions condensed near the particle surface increases, which tends to suppress the rise in the zeta potential. Consequently, the particle zeta potential depends less on Q .

It follows from Eqs 74–77 that the ratio of the transient electrophoretic mobility $\mu(t)$ to the steady mobility $\mu_s = \mu(\infty)$ in a salt-free medium is always simply given by

$$\frac{\mu(t)}{\mu_s} = \frac{q_2}{q_2 - q_1} M\left(\frac{q_1 \sqrt{\nu t}}{a}\right) - \frac{q_1}{q_2 - q_1} M\left(\frac{q_2 \sqrt{\nu t}}{a}\right) \quad (82)$$

for both cases 1 (low-surface-charge case) and 2 (high-surface-charge case), irrespective of the magnitude of the particle surface charge Q .

Figure 1 illustrates the ratio $\mu(t)/\mu_s$ calculated with Eq. 82 as a function of scaled time $\nu t/a^2$ for four values of the mass density ratio ρ_p/ρ_0 , showing how $\mu(t)$ reaches the corresponding steady value μ_s . It is observed that the transient mobility $\mu(t)$ of heavier particles with larger mass density ρ_p requires a longer time to reach its steady value μ_s . Figure 2 gives a 3-D plot of the ratio $\mu(t)/\mu_s$ calculated with Eq. 82 as a function of scaled time $\nu t/a^2$ and the mass density ratio ρ_p/ρ_0 , which covers wider ranges of $\nu t/a^2$ and the mass density ratio ρ_p/ρ_0 .

Figures 1, 2 show that the relaxation time required for $\mu(t)$ to reach its steady value μ_s becomes longer as the mass density ratio ρ_p/ρ_0 increases. An approximate expression for the relaxation time T can be derived as follows. As a crude approximation, Eq. 82 reduces to

$$\frac{\mu(t)}{\mu_s} = 1 - e^{-t/T} \quad (83)$$

with

$$T = \frac{1}{9} \left(1 + \frac{2\rho_p}{\rho_0} \right) \frac{a^2}{\nu} \quad (84)$$

where T can be regarded as the relaxation time.

4 Concluding remarks

We have developed a theory of the transient electrophoresis of colloidal particles in a salt-free medium, which provides vital information for designing effective electrophoresis measurement systems related to micro- and nano-fluidic lab-on-a-chip technologies. We first derived a general expression (Eq. 51 for the Laplace transform $\hat{\mu}(s)$) of the time dependent transient

electrophoretic mobility $\mu(t)$ of spherical colloidal particles in a salt-free liquid medium containing only counterions when a step external electric field is suddenly applied to the colloidal suspension. On the bases of this general expression for $\hat{\mu}(s)$, we derived an accurate analytic expression for $\mu(t)$, which takes the different forms for the low-surface-charge case (Eq. 76) and the high-surface-charge case (Eq. 77). Only for the low-surface-charge case, the transient electrophoretic mobility agrees with that of a sphere in an electrolyte solution in the limit of very low electrolyte concentrations. For the high-surface-charge case, however, the transient mobility becomes independent of the particle surface charge because of the counterion condensation effects. Finally, we derived a simple expression (Eq. 82) for the ratio of the transient electrophoretic mobility to the steady electrophoretic mobility, which is found to take the same form irrespective of the magnitude of the particle surface charge. Finally, it is to be noted that since we have considered the transient motion of colloidal particles under a weak electric field, an overshoot phenomenon, which is sometimes observed in transient processes, is not observed.

Data availability statement

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding author.

Author contributions

HO: Conceptualization, Data curation, Formal Analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing—original draft, Writing—review and editing.

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Conflict of interest

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Glossary

a	Particle radius
e	Elementary electric charge
$E(t)$	Applied step electric field
k	Boltzmann constant
$n(r, t)$	Number density of counterions
n_0	Average number density of counterions
$p(t)$	Pressure
Q	Total surface charge
R	Radius of a spherical free volume containing a colloidal particle
T	Absolute temperature
$u(r, t)$	Liquid velocity
$U(t)$	Transient electrophoretic velocity
$v(r, t)$	Velocity of counterions
γ	Scaled electric potential
$-z$	valence of counterions
ϵ_0	Permittivity of a vacuum
ϵ_p	Relative permittivity of a particle
ϵ_r	Relative permittivity of a medium
η	Viscosity of a medium
λ_c	Drag coefficient of counterions
μ	Transient electrophoretic mobility
μ_c	Electrochemical potential of counterions
$\hat{\mu}(s)$	Laplace transform of $\mu(t)$
$\rho_c(r, t)$	Charge density resulting from counterions
ρ_0	Mass density of a medium
ρ_p	Mass density of a particle
ϕ	Particle volume fraction
Σ	Surface charge density
$\psi(r, t)$	Electric potential
$\psi^{(0)}(r)$	Equilibrium electric potential
ζ	Zeta potential