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ORIGINAL CONTRIBUTION

On the maximum of the magnitude of the electrophoretic mobility of a spherical colloidal particle in an electrolyte solution

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Abstract The magnitude of the electrophoretic mobility μ of a spherical colloidal particle in an electrolyte solution with $\kappa a>3$ (κ=the Debye-Hückel parameter of the electrolyte solution and a=particle radius), when plotted as a function of the particle zeta potential ζ, exhibits a maximum $\mu_{\max}$ at $\zeta=\zeta_{\max}$. Analytic expressions applicable for large κa ($\kappa a\geq 30$) are derived for $\mu_{\max}$ and $\zeta_{\max}$ for a spherical particle in a symmetrical electrolyte solution. Analytic expressions for $\mu_{\max}$ and $\zeta_{\max}$ are also derived for a spherical particle in a 2:1 or 1:2 electrolyte solution. Finally, it is to be noted that $\mu_{\max}$ and $\zeta_{\max}$ for a cylindrical particle of radius a when the particle is oriented perpendicular to the applied electric field are the same as those for a spherical particle of radius a for large κa ($\kappa a\geq 30$).

Keywords Electrophoretic mobility · Zeta potential · Relaxation effect · Mobility maximum · Spherical particle · Cylindrical particle

Introduction

A number of theoretical studies have been made on the electrophoretic mobility of a charged colloidal particle in an electrolyte solution [1–25]. It is known that the magnitude of the electrophoretic mobility μ of a spherical colloidal particle with $\kappa a>3$ (κ=the Debye-Hückel parameter of the electrolyte solution and a=particle radius), when plotted as a function of the particle zeta potential ζ, exhibits a maximum $\mu_{\max}$ at a certain value of ζ, which we denote to be $\zeta_{\max}$ [6–11] (Fig. 1). The appearance of the mobility maximum is due to the relaxation effect. With further increase in zeta potential, the electrophoretic mobility tends to a non-zero limiting electrophoretic mobility, which was treated in detail in refs. [18] and [19]. The purpose of the present paper is to derive analytic expressions for $\mu_{\max}$ and $\zeta_{\max}$. We employ approximate mobility expressions for a charged sphere derived in ref. [20], which are applicable for all values of zeta potentials and large particle radii a such that $\kappa a>30$ and have been applied to analyze the mobility of spherical particles with high zeta potentials [26–28].

Theory

Consider a spherical particle of radius a and zeta potential ζ moving with a velocity $\boldsymbol{U}$ under an external electric field $\boldsymbol{E}$ in a symmetrical electrolyte solution of valence z. The electrophoretic mobility μ of the particle is defined by $\mu=U/E$, where $U=|\boldsymbol{U}|$ and $E=|\boldsymbol{E}|$. In a previous paper [20], we have derived the following general mobility expression, which is correct to the order of $\exp(ze|\zeta|/2kT)/\kappa a$, where z is the valence of counter ions, e is the elementary electric charge, k is Boltzmann's constant, T is the absolute temperature, and κ is the Debye-Hückel parameter, so that it is applicable for all values of ζ at large κa ($\kappa a\geq 30$):

$$\mu=\frac{\varepsilon_r\varepsilon_o}{\eta}\left\{\zeta\mp\frac{2F}{1+F}\left(\frac{kT}{ze}\right)\ln\left(\frac{1+\exp(ze|\zeta|/2kT)}{2}\right)\right\} \tag{1}$$

or its magnitude is given by

$$|\mu|=\frac{\varepsilon_r\varepsilon_o}{\eta}\left\{|\zeta|-\frac{2F}{1+F}\left(\frac{kT}{ze}\right)\ln\left(\frac{1+\exp(ze|\zeta|/2kT)}{2}\right)\right\} \tag{2}$$

with

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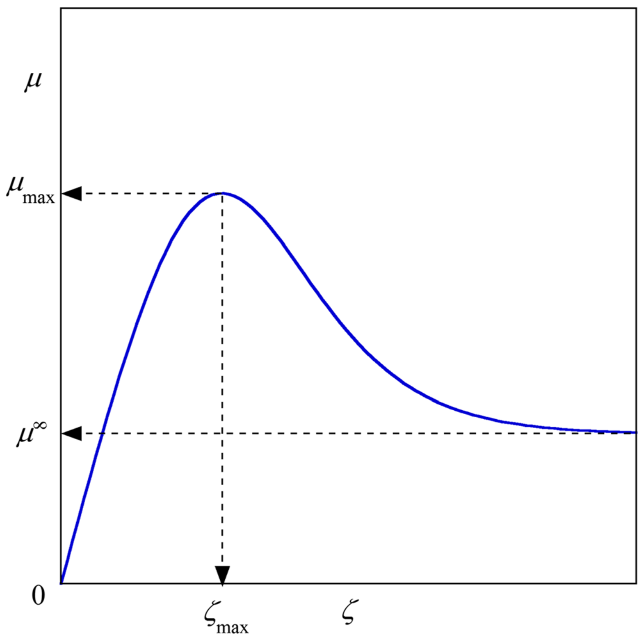

Fig. 1 Electrophoretic mobility μ of a spherical particle plotted as a function of the particle zeta potential ζ for the case where there is a maximum μ_{max} of the magnitude of μ at $\zeta=\zeta_{max}$ and a non-zero limiting value μ^{∞} at $\zeta\rightarrow\infty$

$$F=\frac{2(1+3m)}{\kappa a}\left\{\exp\left(\frac{ze|\zeta|}{2kT}\right)-1\right\} \tag{3}$$

$$m=\frac{2\varepsilon_r\varepsilon_o kT}{3\eta z^2e^2}\lambda=\frac{2N_A\varepsilon_r\varepsilon_o kT}{3\eta z\Lambda^o} \tag{4}$$

where the minus and plus signs in Eq. (1) are used for positive and negative zeta potentials, respectively; λ is the drag coefficient of counterions; m is the scaled drag coefficient, which is further related to the limiting equivalent conductance Λ^0 of counterions; N_A is Avogadro's number; ε_r and η are, respectively, the relative permittivity and the viscosity of the electrolyte solution; and ε_o is the permittivity of a vacuum. Note that F given by Eq. (3) corresponds to Dukhin's number.

For high ζ, where the magnitude of μ reaches a maximum, Eq. (2) is approximated by

$$|\mu|=\frac{\varepsilon_r\varepsilon_o}{\eta}\left\{\frac{|\zeta|-\left(2kT/ze\right)\ln 2}{1+2(1+3m)\exp\left(ez|\zeta|/2kT\right)/\kappa a}+\left(\frac{2kT}{ze}\right)\ln(2)\right\} \tag{5}$$

The value of ζ at which μ reaches a maximum, which we denote by ζ_{max}, is derived from the condition $d\mu/d\zeta=0$ with the result that

$$\left\{\frac{ze|\zeta_{max}|}{2kT}-1-\ln(2)\right\}\exp\left(\frac{ze|\zeta_{max}|}{2kT}\right)=\frac{\kappa a}{2(1+3m)} \tag{6}$$

which gives

$$\zeta_{max}=\pm\left(\frac{2kT}{ze}\right)\left\{W\left(\frac{\kappa a}{4(1+3m)\exp(1)}\right)+1+\ln(2)\right\} \tag{7}$$

where $W(x)$ is the Lambert W function (or the product logarithm) and satisfies $x=W(x)e^{W(x)}$. By substituting Eq. (7) into Eq, (5), we have the following approximate expression for the maximum value μ_{max} of the electrophoretic mobility μ:

$$\mu_{max}=\pm\frac{\varepsilon_r\varepsilon_o}{\eta}\left(\frac{2kT}{ze}\right)\left\{W\left(\frac{\kappa a}{4(1+3m)\exp(1)}\right)+\ln(2)\right\} \tag{8)}$$

In Eqs. (7) and (8), the plus and minus signs are used for positive and negative zeta potentials, respectively. Equations (7) and (8) are the required expressions for μ_{max} and ζ_{max}.

Results and discussion

We have derived approximate analytic Eqs. (7) and (8) for the maximum μ_{max} and its position ζ_{max} of the magnitude of the electrophoretic mobility μ of a spherical particle of radius a and zeta potential ζ in a symmetrical electrolyte solution of valence z. These expressions are derived on the basis of Eq. (5) (or Eqs. (1) and (2)). Some examples of the calculation of the electrophoretic mobility μ for a negatively charged spherical particle for κa=50 in an aqueous electrolyte solution with z=1, 2, and 3 at 25 °C obtained from Eq. (1) are shown in Fig. 1. Figure 1 shows the scaled electrophoretic mobility $E_m=(3\eta e/2\varepsilon_r\varepsilon_o kT)\mu$ plotted as a function of the scaled zeta potential $e\zeta/kT$. The results obtained from Eq. (5) are also plotted as dotted lines in Fig. 1, which agree with the results of Eq. (1) with negligible errors. As counterions, we have chosen K^+, Mg^{2+}, and La^{3+} ions for the cases of z=1, 2, and 3, respectively. We use the following values of Λ^0 and m: $\Lambda^0=73.5\times10^{-4}$ m^2 Ω^{-1} equiv^{-1} and m=0.176 for K^+ ions, $\Lambda^0=53.1\times10^{-4}$ m^2 Ω^{-1} equiv^{-1} and m=0.122 for Mg^{2+} ions, and $\Lambda^0=69.6\times10^{-4}$ m^2 Ω^{-1} equiv^{-1} and m=0.0618 for La^{3+} ions (Note that the values of Λ^0 and m for Mg^{2+} ions given in ref. [25] are incorrect and should be replaced by the above values). The numerical values of the scaled mobility maximum defined by $E_m^{max}=(3\eta e/2\varepsilon_r\varepsilon_o kT)\mu_{max}$ and the scaled zeta potential $ze\zeta_{max}/kT$ (at which $\mu=\mu_{max}$) are $(ze\zeta_{max}/kT, E_m^{max})$ = (5.470, 5.249) for z=1, (2.793, 2.714) for z=2, and (1.913, 1.886) for z=3, while the values obtained from Eqs. (7) and (8) are $(ze\zeta_{max}/kT, E_m^{max})$ = (5.490, 5.235) for z=1, (2.803, 2.705) for z=2, and (1.919, 1.879) for z=3, showing that Eqs. (7) and (8) are excellent approximations with negligible errors (less than 0.4 %).

It follows from Eqs. (7) and (8) that ζ_{max} and E_m^{max} are both inversely proportional to the valence z of counterions, that is,

$$|\zeta_{max}|\propto\frac{1}{z}\text{ and }\left|E_m^{max}\right|\propto\frac{1}{z} \tag{9}$$

as is seen in Fig. 2. It also follows from Eqs. (7) and (8) that $\zeta_{\max}$ and $E_m^{\max}$ are both proportional to $\ln(\kappa a)$ for large κa, that is

$$|\zeta_{\max}| \propto \ln(\kappa a) \text{ and } |E_m^{\max}| \propto \ln(\kappa a) \tag{10}$$

as is actually seen in Figs. 3, 4, and 5. Figure 3 shows the dependence of the electrophoretic mobility μ upon κa, and Figs. 4 and 5 give the κa dependence of $\zeta_{\max}$ and $E_m^{\max}$, respectively.

Similar expressions for $\zeta_{\max}$ and $E_m^{\max}$ for the case of a spherical particle in 2:1 or 1:2 electrolyte solutions can be derived on the basis of the approximate mobility expressions given in ref. [20]. The results are given below.

(i) For a positively charged particle ($\zeta>0$) in a 2:1 electrolyte solution, the electrophoretic mobility μ is given by

$$\mu = \frac{\varepsilon_r\varepsilon_o}{\eta}\left\{\zeta - \frac{2F}{1+F}\left(\frac{kT}{e}\right)\ln\left[\frac{1}{2}+\frac{1}{2}\sqrt{\frac{2}{3}\exp\left(\frac{e\zeta}{kT}\right)+\frac{1}{3}}\right]\right\} \tag{11}$$

with

$$F = \frac{\sqrt{6}}{\kappa a}(1+3m_-)\left\{\exp\left(\frac{e\zeta}{2kT}\right)-1\right\}, \tag{12}$$

$$m_- = \frac{2\varepsilon_r\varepsilon_o kT}{3\eta e^2}\lambda_- = \frac{2N_A\varepsilon_r\varepsilon_o kT}{3\eta\Lambda_-^0}$$

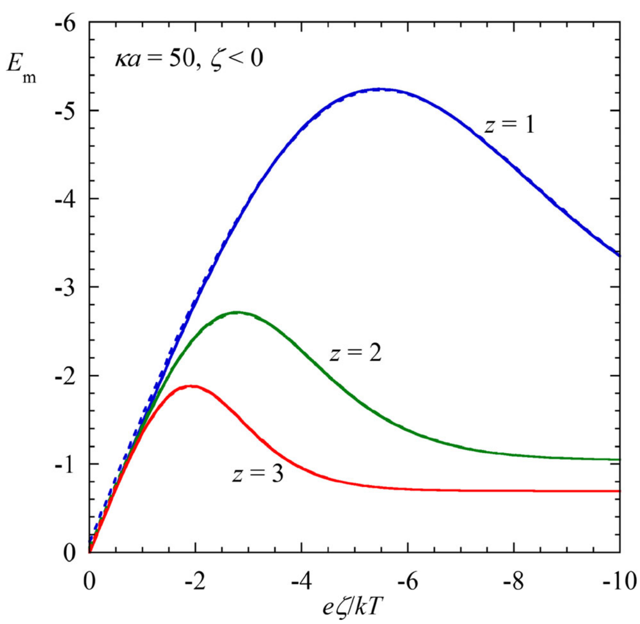

Fig. 2 Scaled electrophoretic mobility $E_m=(3\eta e/2\varepsilon_r\varepsilon_o kT)\mu$ of a negatively charged spherical particle of radius a and zeta potential $\zeta(\zeta<0)$ in an aqueous symmetrical electrolyte solution for three values of the valence of counterions z=1, 2, and 3 as functions of scaled zeta potential $e\zeta/kT$. The values of the reduced ionic drag coefficients for Na^+, Mg^{2+}, and La^{3+} are used as those of counterions for z=1, 2, and 3, respectively. Calculated with Eq. (1) at κa=50 and 25 °C. The *dotted lines* are approximate results obtained from Eq. (5)

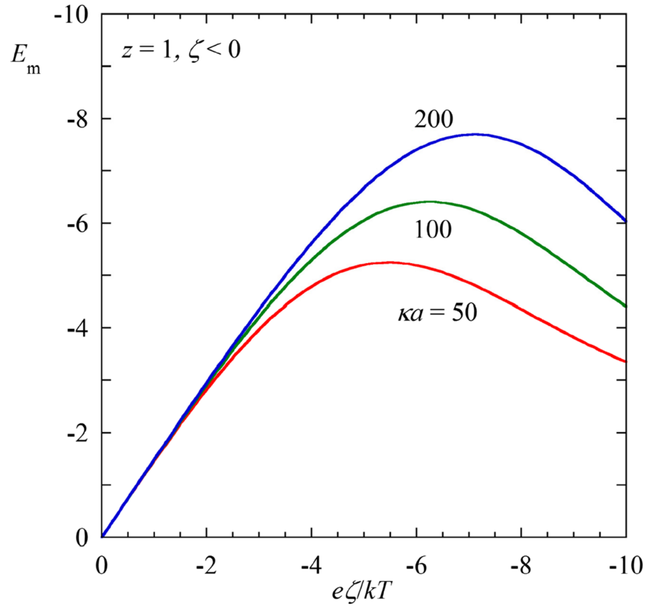

Fig. 3 Scaled electrophoretic mobility $E_m=(3\eta e/2\varepsilon_r\varepsilon_o kT)\mu$ of a negatively charged spherical particle of radius a and zeta potential $\zeta(\zeta<0)$ in an aqueous 1:1 symmetrical electrolyte solution for three values κa plotted as a function of scaled zeta potential $e\zeta/kT$. The value of the reduced ionic drag coefficient m (=0.176) for Na^+ is used as that of counterions. Calculated with Eq. (1) at κa=50, 100, and 200 at 25 °C

where λ_- and Λ_-^0 are, respectively, the drag coefficient and limiting equivalent conductance of counterions (anions of valence −1) and m_- is the scaled drag coefficient. From Eq. (11), we obtain

$$\zeta_m = \left(\frac{2kT}{e}\right)\left\{W\left(\frac{\kappa a}{6(1+3m_-)\exp(1)}\right)+1+\frac{1}{2}\ln(6)\right\} \tag{13}$$

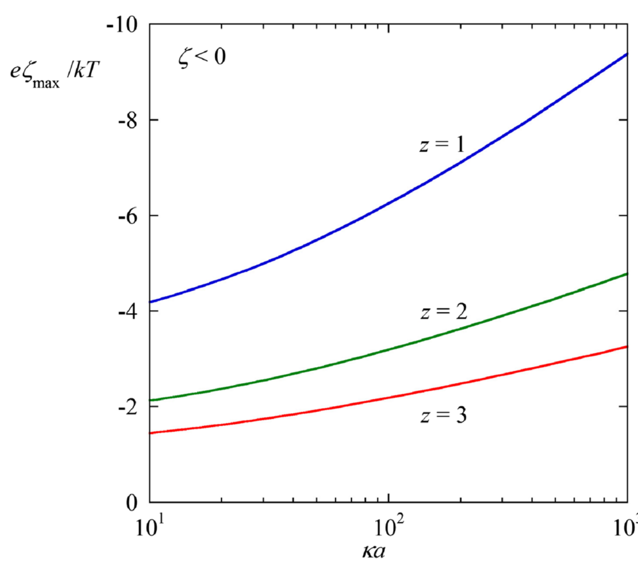

Fig. 4 $e\zeta_{\max}/kT$ as a function of κa calculated with Eq. (7) for z=1, 2, and 3 at 25 °C

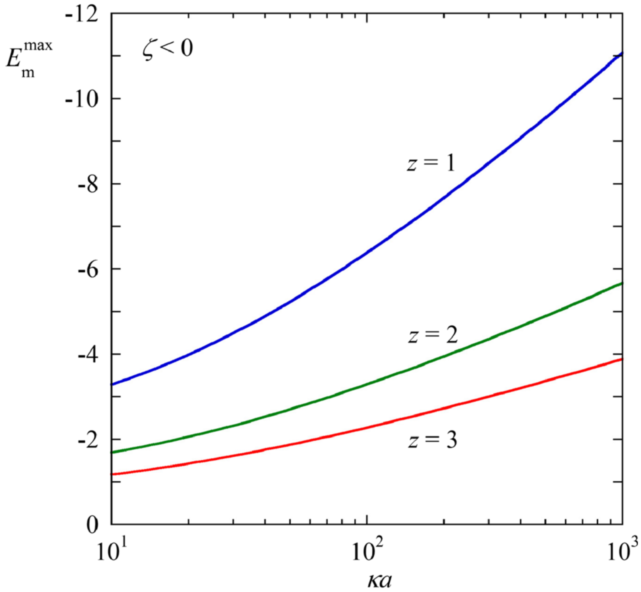

Fig. 5 E_m^{max} as a function of κa calculated with Eq. (8) for z=1, 2, and 3 at 25 °C

$$\mu_{max} = \frac{\varepsilon_r \varepsilon_o}{\eta}\left(\frac{2kT}{e}\right)\left\{W\left(\frac{\kappa a}{6(1+3m_-)\exp(1)}\right)+\frac{1}{2}\ln(6)\right\} \quad (14)$$

(ii) For a negatively charged particle (ζ<0) in a 2:1 electrolyte solution, the electrophoretic mobility μ is given by

$$\mu = \frac{\varepsilon_r \varepsilon_o}{\eta}\left\{\zeta + \frac{2F}{1+F}\left(\frac{kT}{e}\right)\ln\left[\frac{1}{2}\exp\left(\frac{e|\zeta|}{2kT}\right)+\frac{1}{2}\sqrt{\frac{1}{3}\exp\left(\frac{e|\zeta|}{kT}\right)+\frac{2}{3}}\right]\right\} \quad (15)$$

with

$$F = \frac{\sqrt{3}}{\kappa a}(1+3m_+)\left\{\exp\left(\frac{e|\zeta|}{kT}\right)-1\right\}, \quad (16)$$

$$m_+ = \frac{\varepsilon_r \varepsilon_o kT}{6\eta e^2}\lambda_+ = \frac{N_A \varepsilon_r \varepsilon_o kT}{3\eta \Lambda_+^0}$$

where λ_+ and Λ_+^0 are, respectively, the drag coefficient and limiting equivalent conductance of counterions (cations of valence +2) and m_+ is the scaled quantity. From Eq. (15), we obtain

$$\zeta_{max} = -\left(\frac{kT}{e}\right)\left\{W\left(\frac{\kappa a(3+2\sqrt{3})}{18(1+3m_+)\exp(1)}\right)+1+2\ln\left(\frac{2}{1+1/\sqrt{3}}\right)\right\} \quad (17)$$

$$\mu_{max} = -\frac{\varepsilon_r \varepsilon_o}{\eta}\left(\frac{kT}{e}\right)\left\{W\left(\frac{\kappa a(3+2\sqrt{3})}{18(1+3m_+)\exp(1)}\right)+2\ln\left(\frac{2}{1+1/\sqrt{3}}\right)\right\} \quad (18)$$

(iii) For a positively charged sphere (ζ>0) in a 1:2 electrolyte solution, the electrophoretic mobility μ is given by

$$\mu = \frac{\varepsilon_r \varepsilon_o}{\eta}\left\{\zeta - \frac{2F}{1+F}\left(\frac{kT}{e}\right)\ln\left[\frac{1}{2}\exp\left(\frac{e\zeta}{2kT}\right)+\frac{1}{2}\sqrt{\frac{1}{3}\exp\left(\frac{e\zeta}{kT}\right)+\frac{2}{3}}\right]\right\} \quad (19)$$

with

$$F = \frac{\sqrt{3}}{\kappa a}(1+3m_-)\left\{\exp\left(\frac{e\zeta}{kT}\right)-1\right\}, \quad m_- = \frac{\varepsilon_r \varepsilon_o kT}{6\eta e^2}\lambda_- = \frac{N_A \varepsilon_r \varepsilon_o kT}{3\eta \Lambda_-^0} \quad (20)$$

where λ_- and Λ_-^0 are, respectively, the drag coefficient and limiting equivalent conductance of counterions (anions of valence −2) and m_- is the scaled quantity. From Eq. (19), we obtain

$$\zeta_{max} = \left(\frac{kT}{e}\right)\left\{W\left(\frac{\kappa a(3+2\sqrt{3})}{18(1+3m_-)\exp(1)}\right)+1+2\ln\left(\frac{2}{1+1/\sqrt{3}}\right)\right\} \quad (21)$$

$$\mu_{max} = \frac{\varepsilon_r \varepsilon_o}{\eta}\left(\frac{kT}{e}\right)\left\{W\left(\frac{\kappa a(3+2\sqrt{3})}{18(1+3m_-)\exp(1)}\right)+2\ln\left(\frac{2}{1+1/\sqrt{3}}\right)\right\} \quad (22)$$

(iv) For a negatively charged sphere (ζ<0) in a 1:2 electrolyte solution

$$\mu = \frac{\varepsilon_r \varepsilon_o}{\eta}\left\{\zeta + \frac{2F}{1+F}\left(\frac{kT}{e}\right)\ln\left[\frac{1}{2}+\frac{1}{2}\sqrt{\frac{2}{3}\exp\left(\frac{e|\zeta|}{kT}\right)+\frac{1}{3}}\right]\right\} \quad (23)$$

with

$$F = \frac{\sqrt{6}}{\kappa a}(1+3m_+)\left\{\exp\left(\frac{e|\zeta|}{2kT}\right)-1\right\}, \quad (24)$$

$$m_+ = \frac{2\varepsilon_r \varepsilon_o kT}{3\eta e^2}\lambda_+ = \frac{2N_A \varepsilon_r \varepsilon_o kT}{3\eta \Lambda_+^0}$$

where λ_+ and Λ_+^0 are, respectively, the drag coefficient and limiting equivalent conductance of counterions (cations of valence +1) and m_+ is the scaled quantity. From Eq. (23), we obtain

$$\zeta_{max} = -\left(\frac{2kT}{e}\right)\left\{W\left(\frac{\kappa a}{6(1+3m_+)\exp(1)}\right)+1+\frac{1}{2}\ln(6)\right\} \quad (25)$$

$$\mu_{max} = -\frac{\varepsilon_r \varepsilon_o}{\eta}\left(\frac{2kT}{e}\right)\left\{W\left(\frac{\kappa a}{6(1+3m_+)\exp(1)}\right)+\frac{1}{2}\ln(6)\right\} \quad (26)$$

Finally, it is to be noted that the electrophoretic mobility of an infinitely long cylindrical particle of radius a in an electric field when the particle axis is oriented perpendicular to the applied electric field coincides with that of a spherical particle of radius a provided that κa is large ($\kappa a \geq 30$) [29]. Thus, the expressions for $\zeta_{\max}$ and $E_m^{\max}$ obtained above hold good also for $\kappa a \geq 30$.

Conclusion

We have derived simple approximate analytic expressions for the maximum $\mu_{\max}$ of the magnitude of the electrophoretic mobility μ of a charged spherical colloidal particle of radius a and zeta potential ζ in an electrolyte solution and the zeta potential $\zeta_{\max}$ that gives $\mu = \mu_{\max}$. The obtained expressions are derived on the basis of approximate expressions of the electrophoretic mobility which take into account the relaxation effect. These expressions, which are obtained by neglecting terms of order $1/\kappa a$ in the general mobility expression and correct to the order of $\exp(ze|\zeta|/2kT)/\kappa a$ (where z is the valence of counterions), are applicable for all values of zeta potential at large κa ($\kappa a \geq 30$). It is shown that $\zeta_{\max} \propto 1/z$ and $\mu_{\max} \propto 1/z$, and it is also shown that $\zeta_{\max} \propto \ln(\kappa a)$ and $\mu_{\max} \propto \ln(\kappa a)$ for large κa. The corresponding expressions are also derived for a sphere in a 2:1 or 1:1 electrolyte solution. It is to be noted that the results for a cylindrical particle of radius a when the particle is oriented perpendicular to the applied electric field are the same as those for a spherical particle of radius a provided that κa is large ($\kappa a \geq 30$). Finally, it is to be mentioned that the existence of $\mu_{\max}$ at large κa becomes important also in micro-fluidic applications (see e.g., [30]).

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