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## TRANSPORT OF ELECTROLYTE SOLUTIONS ALONG A PLANE BY DIFFUSION-OSMOSIS

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### ABSTRACT

Diffusion-osmosis is an important electrokinetic flow mechanism in microchannels and nanochannels. In an electrolyte system, diffusion-osmosis transport results from the electrostatic interaction between a tangential concentration gradient of the electrolyte and a charged wall. The diffusion-osmotic flow of an electrolyte solution along a plane (in porous media) which is induced by the presence of a charged surface induced by the imposed electrolyte concentration gradient has been reviewed and investigated theoretically. It was assumed that there will be a constant concentration gradient imposed along the axial direction at the steady state. Electrostatic and induced potential distribution and velocity distribution of an electrolyte solution near a charged plate wall were determined from using Poisson, linearized Poisson-Boltzmann, and modified Navier-Stokes equations, respectively. It was found that the normalized induced electric field approaches unity as we go far away from the EDL and normalized velocity increases with an increase in the electro-kinetic distance and approaches a constant. The effect of the induced electric field in the EDL was found to be of a dominant significance on the diffusion-osmotic flow.

**Keywords:** diffusion-osmosis, transport, electrolyte, porous media, charged wall.

### 1. INTRODUCTION

In many areas of science and engineering, electrokinetic flows of electrolyte solutions in charged microchannels and nanochannels has gained much interest to the scientific community. In electrokinetics, electro-osmosis has been the most known transport mechanism used to manipulate the fluid flow in microfluidic and nanofluidic systems. Electro-osmosis transport occurs due to the interaction between an external, tangential, electric field with the electric double layer (EDL) adjacent to a charged wall [1].

Generally, transport of fluids in micropore systems (porous media) involves three different driving forces: (1) pressure difference between the two ends of a capillary pore (convection), (2) concentration gradient of the impermeable solute between the two bulk solutions surrounding the pore on both ends (osmosis), and (3) tangential electrical fields affecting the electrical double layer (EDL) near a charged pore wall (electro-osmosis). Transport problems related to the three mentioned driving forces have been studied extensively in the past [2, 3, 4].

However, it has been found that there is another driving force for electrolyte solutions in a capillary pore which is known as diffusion osmosis and is associated with the presence of a concentration gradient of a permeable solute along the capillary pore. Diffusion-osmosis is an important electrokinetic flow mechanism in microchannels and nanochannels for understanding transport of fluids in porous media [2, 4]. In an electrolyte system, diffusion-osmosis transport results from the electrostatic interaction between a tangential concentration gradient of the electrolyte and a charged wall. Dissociation of electrolytes along the charged solid surface would result in a fluid flow by two mechanisms: (1) chemi-osmotic effect due to stresses developed by tangential gradient of excess pressure within the EDL (2) electro-osmotic effect due to the inequality of the tangential diffusive and

convective fluxes of the two electrolyte ions which is related to the macroscopic electric field [1, 2].

Understanding the mechanism of diffusion-osmotic flow of electrolyte solutions in porous media would make it possible to improve and develop much more advanced applications related to separation technologies. Permeable membranes for molecular separation, nanofluidic devices for biological and chemical analysis, and polymer electrolyte fuel cells for energy storage are some important applications of diffusion-osmotic flow [1].

The diffusion-osmotic flow of an electrolyte solution along a plane (in porous media) which is induced by the presence of a charged surface induced by the imposed electrolyte concentration gradient has been discussed and investigated theoretically by Ma and Keh (2006) [2]. In other words, there will be a constant concentration gradient imposed along the axial direction at the steady state.

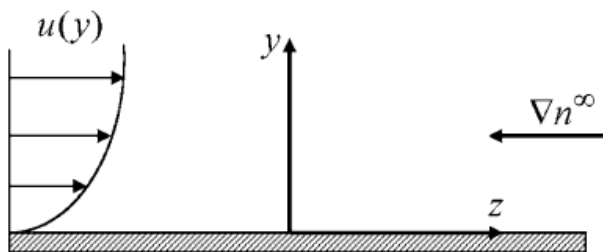
Analytical solutions of a diffusion-osmotic transport of a permeable solute (electrolyte solution) along a charged plane has been derived and identified. The use of the well-known Poisson and linearized Poisson-Boltzmann equations, which applies to the case of low potentials or low fixed-charge densities, with the basics of transport phenomena in electrolyte systems allowed us to obtain the electrostatic potential distribution of an electrolyte solution near a charged plate wall. Electrostatic potential distribution would help us in calculating thickness of EDL adjacent to the charged walls which is an arbitrary thickness relative to the capillary pore size. Moreover, fluid velocity profile along the tangential direction (the horizontal position) was obtained semi-analytically from solving the modified Navier-Stokes equation with neglecting electric current arising from the cocurrent diffusion, electric migration, and diffusion-osmotic convection of the electrolyte ions. It should be



noted that any charged surface or plane wall may have either a constant surface potential or a constant surface charge density with an arbitrary quantity, which would play a key role in simplifying our transport problem analysis [2, 4, 5]

## 2. PROBLEM FORMULATION AND MATHEMATICAL MODELING

Under steady state conditions, the diffusion-osmotic flow of a symmetrically charged electrolyte solution, of valence  $Z$  (a positive integer), that is tangential (in  $z$ -axis) to a uniformly charged plane wall of length  $L$  is analyzed analytically, Figure-1. To ease our calculations, several assumptions were used, including that the applied electrolyte concentration gradient  $\nabla n^\infty$  is constant along the tangential direction, where  $n^\infty$  is the linear electrolyte concentration in the bulk phase and far away from the surface at  $y \rightarrow \infty$ , and setting  $z = 0$  at the midpoint along the plane wall allows us to neglect any variations in the electrostatic potential and ionic concentrations of the EDL near to the wall in the tangential direction [1, 2, 5].



**Figure-1.** Diffusion-osmosis flow tangential to a plane wall due to the presence of an applied concentration gradient of an electrolyte.

### 2.1 Electrostatic potential distribution

Considering the previous assumption and with the utilization of the well-known Poisson equation in microscopic models as shown in Eq. (1) and Eq. (2); substitution of Eq. (2) into Eq. (1) gives us Eq. (3) [1-3, 6].

$$\nabla^2 \psi = -\frac{\rho_e}{\epsilon} \quad (1)$$

$$\rho_e = F \sum_i z_i n_i \quad (2)$$

where  $\psi$  is the electric potential,  $\rho_e$  is the charge density,  $\epsilon = 4\pi\epsilon_0\epsilon_r\epsilon_r$  is the relative permittivity of the electrolyte solution and  $\epsilon_0$  is the permittivity of a vacuum,  $F$  is Faraday's constant,  $z_i$  is a valence (a positive or negative integer), and  $n_i$  represents local concentrations of both cations,  $n_+(y, 0)$ , and anions,  $n_-(y, 0)$  in the electrolyte solution [1,2].

$$\frac{d^2 \psi}{dy^2} = -\frac{4\pi Ze}{\epsilon} [n_+(y, 0) - n_-(y, 0)] \quad (3)$$

the Boltzmann distribution equation, Eq. (4), relates between the local ionic concentrations to the electrostatic potential as shown in Eq. (5) [1, 2].

$$n_i = n^\infty e^{\pm \bar{\psi}} \quad (4)$$

$$n_{\pm} = n^\infty e^{-\bar{\psi}} ; \quad n_- = n^\infty e^{+\bar{\psi}} \quad (5)$$

where  $\bar{\psi} = (z_i F \psi / RT) = (Ze \psi / kT)$  is the dimensionless electrostatic potential at a position  $y$  from the wall relative to that in the bulk solution,  $R$  is universal gas constant,  $k$  is Boltzmann constant,  $T$  is the absolute temperature. Substituting Eq. (5) into Eq. (3) results in the well-known Poisson-Boltzmann equation, Eq. (6) [1-3, 6];

$$\frac{d^2 \bar{\psi}}{dy^2} = \mathcal{K}^2 \sinh \bar{\psi} \quad (6)$$

where  $\mathcal{K} = [4\pi(Ze)^2 n^\infty(z=0) / \epsilon kT]^{1/2}$  is the Debye screening parameter. Table-1 shows the boundary conditions and the dimensionless potential distributions of the two popular case scenarios at the charged plate wall; which were solved by using Eq. (6) and the given boundary conditions. For the constant surface charge density case, the dimensionless potential equation shows that  $\rho_e$  is proportional to  $\mathcal{K}$ , where  $\zeta$  is inversely proportional to  $\mathcal{K}$  [1, 2].

**Table-1.** Dimensionless potential profile along a charged plane wall [1, 2, 5-7].

Case scenario	Boundary conditions	Dimensionless potential
Constant surface zeta potential ( $\zeta$ )	$y = 0; \bar{\psi} = \zeta$ $y \rightarrow \infty; \bar{\psi} = 0$	$\bar{\psi} = 2 \ln \left[ \frac{1 + \gamma e^{-\mathcal{K}y}}{1 - \gamma e^{-\mathcal{K}y}} \right]$ Where $\gamma = \tanh(Ze \zeta / 4kT)$
Constant surface charge density ( $\rho_e$ )	$y = 0; \frac{d\bar{\psi}}{dy} = -\frac{4\pi\rho_e}{\epsilon}$ $y \rightarrow \infty; \bar{\psi} = 0$	$\zeta = \frac{2kT}{Ze} \sinh^{-1} \left( \frac{2\pi Ze \rho_e}{\epsilon \mathcal{K} kT} \right)$ Obtained as the above solution still holds.

### 2.2 Induced electric field distribution

The total flux can be expressed by using Nernst-Einstein equation as in Eq. (7) [1-3, 6];

$$J_{\pm} = -D_{\pm} \left( \nabla n_{\pm} + \frac{z_i F}{RT} n_{\pm} \nabla \psi \right) + n_{\pm} u \quad (7)$$

where  $u = u(y)$  is the fluid velocity relative to the plane wall in the direction of decreasing electrolyte concentration,  $D_+$  and  $D_-$  are the diffusion coefficients of the cations and anions, respectively,  $E = E(y)$  is the macroscopic electric field induced by the concentration gradient of the electrolyte. Substitution of



$(z_i F \psi / RT) = (Ze \psi / kT)$  into Eq. (7) and applying the principle of superposition for the electric potential gives [1-3, 6];

$$J_{\pm} = -D_{\pm} \left[ \nabla n_{\pm} + \frac{Ze}{kT} n_{\pm} (\nabla \psi - E) \right] + n_{\pm} u \quad (8)$$

assuming that  $J_+ = J_- = J$  is possible if we have no net electric current arising from the transport of cations and anions by cocurrent diffusion, electric migration, and diffusion-osmotic convection; which will yield to have an analytical expression for the induced electric field distribution as obtained in Eq. (9) [1, 2, 7];

$$E = \frac{kT \nabla n^{\infty}}{Ze n^{\infty}(z=0)} \left[ \frac{(1+\beta)e^{-\bar{\psi}} - (1-\beta)e^{\bar{\psi}}}{(1+\beta)e^{-\bar{\psi}} + (1-\beta)e^{\bar{\psi}}} + \frac{Pe \sinh \bar{\psi}}{(1+\beta)e^{-\bar{\psi}} + (1-\beta)e^{\bar{\psi}}} \frac{u}{U^*} \right] \quad (9)$$

where  $U^* = 2kT|\nabla n^{\infty}|/\eta \mathcal{K}^2$  is a characteristic value of the diffusio-osmotic velocity,  $\beta = (D_+ - D_-)/(D_+ + D_-)$  is the dimensionless effective diffusivity,  $Pe = [4n^{\infty}(z=0)U^*/(D_+ + D_-)|\nabla n^{\infty}|] = [8n^{\infty}(z=0)kT/(D_+ + D_-)\eta \mathcal{K}^2]$ , and  $\eta$  is the fluid viscosity. Eq. (9) shows that  $E(y)$  is strongly dependent on the local electrostatic potential  $\psi$  and fluid velocity  $u$ . Considering a situation where we have a point that is far away from the charged plate,  $\mathcal{K}y \rightarrow \infty$ , then  $\psi \rightarrow 0$  and Eq. (9) becomes Eq. (10). Note that  $\mathcal{K}y$  is the dimensionless position relative to Debye screening parameter and  $\beta$  is between, or equal to,  $-1$  and  $1$  with the upper and lower bounds occurring as  $D_-/D_+ \rightarrow 0$  and  $\infty$ , respectively [1, 2, 5].

$$E^{\infty} = \frac{kT \beta \nabla n^{\infty}}{Ze n^{\infty}(z=0)} \quad (10)$$

### 2.3 Fluid velocity distribution

The momentum balance from the modified Navier-Stokes equations can be applied for the steady diffusion-osmotic flow along a plane wall to determine pressure and velocity distributions. Assuming the fluid is Newtonian and incompressible;  $y$  and  $z$  direction equations, respectively, gives [1-3, 6];

$$\frac{\partial p}{\partial y} + Ze(n_+ - n_-) \frac{d\psi}{dy} = 0 \quad (11)$$

$$\eta \frac{\partial^2 u}{\partial y^2} = \frac{\partial p}{\partial z} - Ze(n_+ - n_-)E = 0 \quad (12)$$

boundary conditions for the velocity profile at no-slip wall and at infinity are [1-3,6];

$$y = 0 \quad ; \quad u = 0 \quad (13)$$

$$y \rightarrow \infty \quad ; \quad \frac{du}{dy} = 0 \quad (14)$$

substitution of Eq. (5) into Eq. (11) and integration gives [1-3,6];

$$\int_p^{p_{\infty}} \frac{\partial p}{\partial y} dy + Ze(n^{\infty} e^{-\bar{\psi}} - n^{\infty} e^{+\bar{\psi}}) \int_0^{\bar{\psi}} \frac{d\psi}{dy} dy = 0 \quad (15)$$

$$p_{\infty} - p + n^{\infty} Ze(e^{-\bar{\psi}} - e^{+\bar{\psi}}) \bar{\psi} = 0 \quad (16)$$

$$p = p_{\infty} + n^{\infty} kT(e^{-\bar{\psi}} - e^{+\bar{\psi}}) \bar{\psi} \quad (17)$$

$$p = p_{\infty} + 2n^{\infty} kT(-\sinh \bar{\psi}) \quad (18)$$

$$\sinh \bar{\psi} + \cosh \bar{\psi} = ? \quad (19)$$

evaluating Eq. (19)

$$\frac{1}{2} e^{\bar{\psi}} + \frac{1}{2} e^{-\bar{\psi}} + \frac{1}{2} e^{\bar{\psi}} - \frac{1}{2} e^{-\bar{\psi}} = e^{\bar{\psi}} \quad (20)$$

for  $\mathcal{K} \rightarrow \infty$ ; ( $Ze \rightarrow \infty$  or  $kT \rightarrow 0$ ), thus  $\bar{\psi} \rightarrow 0$ ;  $e^0 = 1$ , and then Eq. (19) becomes

$$\sinh \bar{\psi} + \cosh \bar{\psi} = 1 \quad (21)$$

substitution of Eq. (21) into Eq. (18) results in

$$p = p_{\infty} + 2n^{\infty} kT(\cosh \bar{\psi} - 1) \quad (22)$$

where  $p_{\infty}$  is the pressure far away from the wall which is constant. Substituting Eq. (5) and Eq. (22) into Eq. (12) and integrating with respect to  $y$ -direction twice subject to the given boundary condition in Eq. (13) and Eq. (14) gives [1, 2, 7];

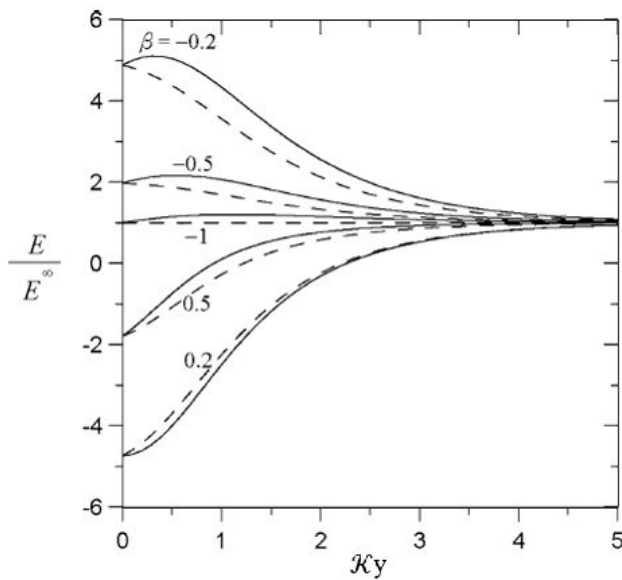
$$\frac{u}{U^*} = \int_0^{\mathcal{K}y} \int_{\infty}^{\mathcal{K}y} \left[ \cosh \bar{\psi} - 1 + \frac{Ze n^{\infty}(z=0)}{kT |\nabla n^{\infty}|} E \sinh \bar{\psi} \right] d(\mathcal{K}y) d(\mathcal{K}y) \quad (23)$$

In Eq. (23)  $E$  can be numerically solved as a function of the dimensionless parameters  $\mathcal{K}y$ ,  $Ze\zeta/kT$ ,  $\beta$ , and  $Pe$ . Then, velocity distribution of the electrolyte solution can be determined by the numerical integrations and with the known results of  $\bar{\psi}$  and  $E$ . Obviously,  $u/U^* = 0$  everywhere if  $\zeta = 0$ . One can obtain the bulk-phase diffusio-osmotic velocity  $u_{\infty}$  of the electrolyte solution at a large distance from the plane wall by taking  $\mathcal{K}y \rightarrow \infty$  and then Eq. (23) becomes [1, 2, 7];

$$\frac{u_{\infty}}{U^*} = \beta \frac{Ze\zeta}{kT} + 4 \ln \cosh \left( \frac{Ze\zeta}{4kT} \right) \quad (24)$$

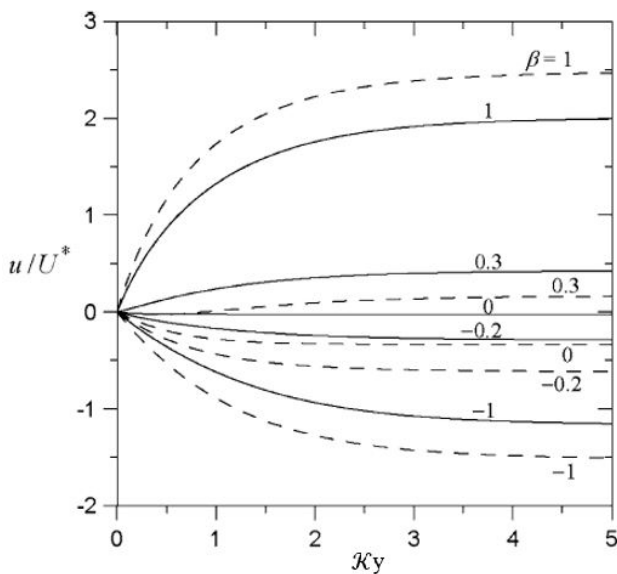
### 3. RESULTS AND DISCUSSIONS

The dimensionless and normalized induced electric field  $E/E^{\infty}$  was plotted against the electro-kinetic distance  $\mathcal{K}y$  from the wall for various values of the parameters  $Pe$  and  $\beta$  as shown in Figure-2. It was noted that there was a symmetry between the curves with a specified value of  $-Ze\zeta/kT$  and  $\beta$  versus  $Ze\zeta/kT$  and  $-\beta$ . In general, the normalized induced electric field approaches unity (the bulk-phase value) as we go far away from the EDL (Debye screening parameter) near to the wall and at  $\mathcal{K}y = 5$ . As  $\beta \rightarrow 0$ , the normalized induced electric field  $E/E^{\infty} \rightarrow \infty$  [1,2].



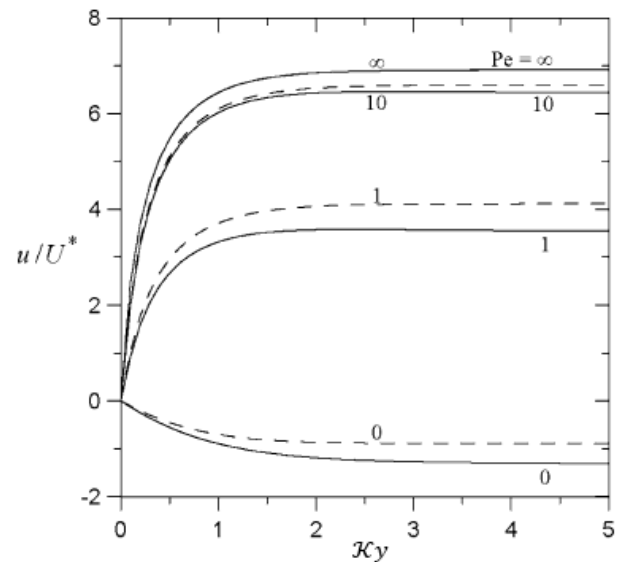
**Figure-2.** Normalized electric field induced by an electrolyte gradient along a plane wall versus the electro-kinetic coordinate  $Ky$  for various values of the parameter  $\beta$  with  $Ze\zeta/kT = 2$ ; solid curves:  $Pe = 1$ , dashed curves:  $Pe = 0$ .

In Figure-3, the dimensionless diffusion-osmotic velocity distribution  $u(Ky)/U^*$  of an electrolyte solution along a plane wall was plotted for various values of the parameters  $Pe$ , and  $\beta$ . Similar to the induced potential analysis, the normalized velocity increases with an increase in the electro-kinetic distance (moving away from the EDL) and approaches a constant (the bulk-phase value) as  $Ky = 5$ .



**Figure-3.** Normalized diffusion-osmotic velocity along a plane wall versus the electro-kinetic coordinate  $Ky$  for various values of the parameter  $\beta$  with  $Ze\zeta/kT = 2$ ; solid curves:  $Pe = 1$ , dashed curves:  $Pe = 0$ .

Higher values of  $Pe$ , which is associated with the effect of the electrolyte convection, reduces the probability of having a negative flow toward the higher concentration region. In general, when  $\beta = 0$ , an increase in  $Pe$  results in a monotonic increase in  $u/U^*$  and as  $Pe \rightarrow \infty$ ,  $u/U^*$  becomes finite, Figure-4. When  $Pe = 0$  and  $\beta = 0$ ,  $E$  vanishes everywhere and  $u$  is negative due to the chemi-osmotic contribution only. When  $Pe = 0$  and  $\zeta * \beta$  is negative,  $u$  is also negative. However, when  $Pe = 0$  and  $\zeta * \beta$  is positive, the flow direction depends on the combination of parameters  $Ze\zeta/kT$ ,  $\beta$ , and  $Ky$ .



**Figure-4.** Normalized diffusion-osmotic velocity along a plane wall versus the electro-kinetic coordinate  $Ky$  for various values of the parameter  $Pe$  with  $Ze\zeta/kT = 6$ ; solid curves:  $\beta = -0.2$ , dashed curves:  $\beta = 0$ .

Analysis of the  $\beta$  factor effect showed that if the magnitude of  $\beta$  is sufficiently large,  $u$  is positive; but if the magnitude of  $\beta$  is sufficiently small,  $u$  is negative. When  $u$  is negative, it means the fluid flows toward the higher electrolyte concentration. However, having a positive value of  $u$  indicates that the fluid flows against the electrolyte concentration gradient and toward the lower electrolyte concentration (from high to low). In both cases, the normalized velocity increases monotonically with the increase in the electro-kinetic distance  $Ky$  from the wall and with the increase in  $Ze|\zeta|/kT$ , as concluded from Figure-3 and Figure-4.

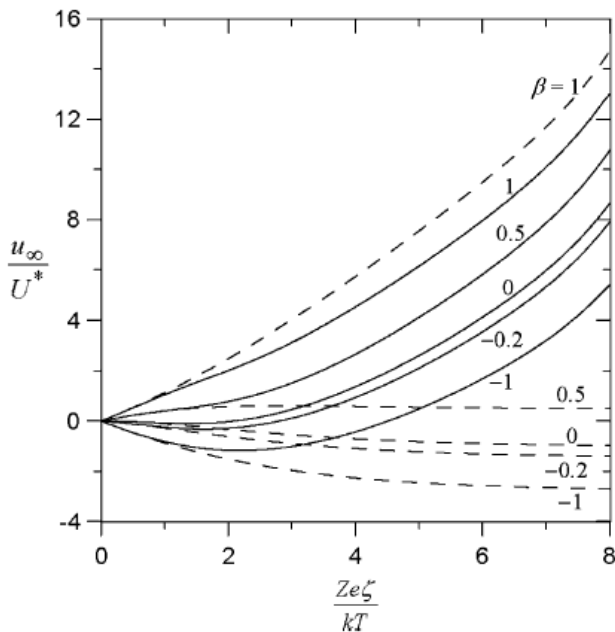
It was demonstrated that the analysis of  $u_\infty/U^*$  far away from the plane wall would be quite similar to that of  $u/U^*$  for a given value of  $Ky$ . Figure-5 shows that higher surface potential values have a noticed effect in increasing  $u/U^*$  values when  $Pe = 1$ .

Theoretical calculations determined that it was possible to estimate the direction of the diffusion-osmotic flow relative to the concentration gradient from the fundamental parameters such as zeta potential (or surface charge density) of the wall and the electrolyte solution properties. It was found that the effect of the lateral





distribution of the induced tangential electric field on the diffusion-osmotic flow was very significant [1, 2, 5].



**Figure-5.** Normalized bulk-phase diffusion-osmotic velocity along a plane wall plotted versus the dimensionless surface potential  $Ze\zeta/kT$  for various values of the parameter  $\beta$ ; solid curves:  $Pe = 1$ , dashed curves:  $Pe = 0$ .

#### 4. SHORTCOMINGS/LIMITATIONS AND FUTURE PROGRESS

Diffusion-osmotic flow along a charged parallel plane was derived with various assumptions such that the fluid is Newtonian, steady state, incompressible, y-direction dependent only, and slightly nonuniform in the electrolyte concentration with only a small external gradient of the electrolyte concentration along the plane wall [1, 5]. Since there is only a small external gradient, it was assumed that the applied electrolyte concentration gradient  $\nabla n^\infty$  is constant along the tangential direction, where  $n^\infty$  is the linear electrolyte concentration in the bulk phase and far away from the surface at  $y \rightarrow \infty$  [1, 2]. Yet, considering a high external concentration gradient where  $|\nabla n^\infty|$  is relatively large would lead to a varying electrostatic potential  $\bar{\psi}$  in the EDL adjacent to the wall and therefore potential variations along the tangential position may not be neglected.

The consideration of many assumptions in solving the distribution profile of both induced electric potential and fluid velocity may result in some kind of errors and deviation from a real case scenario. However, a better judgment can be inferred when comparing experimental results with numerical analysis.

The theoretical and numerical investigations were carried out for electrolyte systems with bare walls. In contrast to this, the existence of a surface charge layer can result in a quite different diffusion-osmotic flow relative to a charged plane wall. Earlier studies found that the

structure of the surface charge layer have a tremendous effect on the flow velocity depending on the plane (or capillary wall) characteristics and the electrolyte solution [9]. For example, a slit with a surface charge layer can lead to an augmented or a diminished electro-kinetic flow and the flow may also be reversed relative to that in a capillary with bare walls [4].

Linearized Poisson-Boltzmann equation have only a few terms analyzed (after applying Taylor's series expansion to the non-linear form) and this may result in a huge error in calculations if and only if the further terms have large magnitude values. Non-linear form of Poisson-Boltzmann will obviously result in having a much better prediction, but the problem will be more difficult to solve analytically.

Nevertheless, X. Xing (2011) applied the non-linear Poisson-Boltzmann equation for two parallel uniformly charged plates and obtained new exact asymptotic results in various regime, but it turned out that the theory has some limitations and assumptions that might affect exact solutions. There are three different important issues which had not been considered in the Poisson-Boltzmann equation, and they are as the following [10, 11]:

- Crystallization:** High surface charge density may crystallize the counter-ion density on the plate surface if they become so high near the surface.
- Neutral bound pairs:** The response of ions forms neutral bound pairs and ions with multiple valences to the external charged objects is a serious understood issue.
- Chemical interactions:** There must be short range chemical interactions between ions and charged surfaces in real situations; which may not be neglected in a formalized theory.

Taking above problems into consideration is critical in analyzing transport of electrolytes in porous media. Even though crystallization possibility has been extensively explored by many authors, the problem has not been completely understood. Hence, the latter two issues must be investigated thoroughly and should be the major mission of future works [10].

#### 5. CONCLUSIONS

Electrostatic and induced potential distribution and velocity distribution of an electrolyte solution near a charged plate wall were obtained theoretically. In general, the normalized induced electric field approaches unity as we go far away from the EDL and normalized velocity increases with an increase in the electro-kinetic distance and approaches constant. When  $u$  is negative, it means the fluid flows toward the higher electrolyte concentration. However, having a positive value of  $u$  indicates that the fluid flows against the electrolyte concentration gradient.

The effect of the induced electric field in the EDL was found to be of dominant significance on the diffusion-osmotic flow. The direction of the diffusion osmotic flow



relative to the concentration gradient is determined by the combination of the zeta potential (or surface charge density) of the wall and the properties of the electrolyte solution. Understanding the mechanism of diffusion-osmotic flow of electrolyte solutions in porous media would make it possible to improve and develop much more advanced applications related to separation technologies.

## ACKNOWLEDGEMENTS

The author would like to acknowledge the Saudi Arabian Cultural Mission (SACM) for their support, fund, and encouragement to accomplish this work.

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