Effects of non-Newtonian power law rheology on mass transport of a neutral solute for electro-osmotic flow in a porous microtube

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Mass transport of a neutral solute for a power law fluid in a porous microtube under electro-osmotic flow regime is characterized in this study. Combined electro-osmotic and pressure driven flow is conducted herein. An analytical solution of concentration profile within mass transfer boundary layer is derived from the first principle. The solute transport through the porous wall is also coupled with the electro-osmotic flow to predict the solute concentration in the permeate stream. The effects of non-Newtonian rheology and the operating conditions on the permeation rate and permeate solute concentration are analyzed in detail. Both cases of assisting (electro-osmotic and poiseuille flow are in same direction) and opposing flow (the individual flows are in opposite direction) cases are taken care of. Enhancement of Sherwood due to electro-osmotic flow for a non-porous conduit is also quantified. Effects of non-Newtonian rheology on Sherwood number enhancement are observed. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4817770>]

I. INTRODUCTION

Mass transport in microfluidic systems has gained considerable interest, in the past decade, among various researchers from multidisciplinary fields. The art of miniaturization in the dimensions of ten to hundreds of micrometer has potential for technological innovations in microscale molecular transport.¹ Applications of microfluidic devices in bio-analysis,^{2,3} microelectronics,⁴ development of biosensors,⁵ controlled drug delivery⁶ and drug discovery,⁷ micro-separation systems separating biomolecules,⁸ ions,⁹ neutral solutes,¹⁰ etc., are very promising owing to their high surface to volume ratio and complex fluid flow associated with electrokinetic effects.^{11,12}

Microfluidic flows occur due to the effect of pressure gradient or electroosmotic effect or combination of both. Heat and Mass transfer analysis in microfluidic flows is significant for operation of all the microfluidic devices and separation systems.^{13,14} The velocity field integrated with heat transfer for Newtonian systems has been studied by various researchers in micro-chip cooling,^{15,16} microscale heat exchanger,¹⁷ micro-electronic-mechanical-systems (MEMS),¹⁸ microreactors,¹⁹ etc. The corresponding mass transfer problems have applications in transdermal drug delivery,^{20,21} electrolyte transport in fuel cells,²² transport in hydrogel,²³ electrokinetic separation,²⁴ etc. These applications include study of transport processes involving both fluid flow and mass transfer in a microchannel with porous wall or membrane. Permeation of desired solutes through the porous barrier (skin) is controlled by polarization of larger neutral solutes over the membrane wall. Relevant transport of species is entirely dictated by the mass transfer boundary layer developed by the rejected neutral solutes. Estimation of mass transfer coefficient and understanding of the mass transport mechanism are of paramount importance in design and operation of such systems. There are several instances where electrokinetic transport in microchannels is preferred for transport of fluids which does not follow Newtonian rheology, e.g., polymer

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solutions^{25,26} or biofluids.²⁷ Various studies involving power law and viscoelastic fluid models are available to characterize the flow behavior^{28,29} and heat transfer to quantify the Nusselt number^{30–33} in impervious conduits.

An excellent example of microfluidic technique for molecular sensing has been described by Chang and Yossifon.³⁴ Their experimental results provide deeper understanding of the solvent flux through the nanoporous membrane and the effect of electrolyte concentration on the nanopore conductance. Their analyses highlight the importance of electrokinetics in development of nanobiosensing diagnostic device. The effect of capillary flow in nanochannels is important for design of nanofluidic device. Hambling *et al.* analyzed the speed of capillary flow in nanochannels using sacrificial etching technique together with sacrificial core of different metals.³⁵ It shows that the flow characterization is considerably different from the prediction of classical continuum theory. It is well known that the electroosmotic flow in micro/nanochannels is significantly affected by the electrical properties of the channel walls. Altering the conductance of channels by surface functionalization is performed by Martins *et al.*³⁶ A variable charge model was developed which predicted the surface conductance due to change in electrolyte concentration at dilute ranges, change in wall properties due to surface reactions and change in pH of the solution. These findings are important for label-free biosensing, where detailed knowhow of the interaction of surface properties with fluid composition is necessary. Effect of wall permittivity on low Reynolds number electroviscous flow has profound influence on the distribution of charges (both co-ion and counterions) within the channel constrictions.³⁷ All these studies show that knowledge of the various physical parameters on the microfluidic device applications is very essential.

The mass transport analysis for Newtonian fluid rheology in porous microchannel and microtubes has been analyzed by Vennela *et al.*^{38,39} They have developed an analytical solution of Sherwood number in such geometry for Newtonian rheology, considering the combined effects of pressure and electroosmotic gradients. Mass transfer in a microchannel bioreactor partially filled with porous medium has been studied numerically by Chen *et al.*^{40,41} However, they have dealt with Newtonian fluid and pressure driven flow only. Mass transport in a micro-electrophoresis device was numerically computed by Barz *et al.*,⁴² which deals with Newtonian rheology and only electro-osmotic effects were taken into account in the calculations. Theoretical analysis of the velocity profile in a cylindrical microtube of fluid having non-Newtonian rheology is reported by Zhao and Yang.⁴³ Theoretical quantification of mass transfer coefficient (or Sherwood number) in a porous microtube for non-Newtonian rheology is more relevant in physiological system. The transport of fluid in the tissues and capillaries, transport of enzymes through blood brain barrier for Alzheimer's patient involves knowledge of the mass transfer of such physiological fluids.^{44–46}

Mass transport analysis in a porous rectangular microchannel has been analyzed by Mondal and De⁴³ for power law fluid. In the present work, an analytical expression for Sherwood number is obtained for a neutral solute in a porous microtube with a non-Newtonian fluid, having generalized power law behavior under combined electroosmotic and pressure driven flow. The domain of validity of Sherwood number has also been identified. Profiles of concentration of solute permeation under various operating conditions are also computed and their implications are analyzed. Since, the cylindrical geometry is the most common fabricated flow paths in microfluidic systems, the present work can be extensively applied for prediction of mass transfer for both electroosmotic and pressure driven flows in most of the customized microfluidic devices for bio-analysis, lab on a chip components, micro-separation process, and electrophoretic extraction.

II. DESCRIPTION OF THE VELOCITY PROFILE IN THE CYLINDRICAL MICROTUBE

The description of a 2D electroosmotic flow of a neutral solute, in a porous microtube, having a 1:1 symmetric electrolytic solution is illustrated in Fig. 1. The tubular inner layers are charged at the wall potential, ζ . Variation of permittivity is mainly affected by ordering and orientation of water molecules within electric double layer which is more valid for finite sized ions.^{47,48} A modified Poisson-Boltzmann theory was developed by Outhwaite considering the

FIG. 1. Schematic of the flow profile and boundary layer within the microtube.

finite sized ions and point dipoles using lattice statistical method.⁴⁹ However, it is identified that variation of permittivity is prominent for high surface density, in the order of 0.1 C/m^2 and very high electrical field strength, in the order of 10^8 V/m .⁴⁸ High electric field strength is practically realizable only in case of nanochannels. Therefore, within Debye-Huckel approximation, these phenomena are insignificant because the surface charge density varies in the order of 10^{-3} to 10^{-4} C/m^2 for moderate to dilute electrolyte concentration and electric field strength in a typical microchannel is in the order of 10^3 V/m . Hence, the spatial variation of relative permittivity is assumed to be invariant in the present study. Considering the Debye-Huckel approximation, the Poisson-Boltzmann equation in cylindrical co-ordinates, for 1:1 electrolytes, is expressed as,⁵⁰

$$r \frac{d}{dr} \left(r \frac{d\psi}{dr} \right) - \kappa^2 r^2 \psi = 0, \quad (1)$$

where ψ is the surface potential. The relevant boundary conditions are,

$$\text{at } r = 0; \quad \frac{d\psi}{dr} = 0, \quad (2a)$$

and

$$\text{at } r = R; \quad \psi = \zeta, \quad (2b)$$

where R is the radius of the microtube and ζ is the wall potential. The expression of surface potential is obtained by solution of the above equation,³⁸

$$\psi = \zeta \frac{I_0(\kappa r)}{I_0(\kappa R)}. \quad (3)$$

The rheology of a power law fluid flowing in a circular cross section is defined as,

$$\tau_{zr} = m \left(-\frac{dv_z}{dr} \right)^n \quad \text{for } 0 \leq r \leq R, \quad (4)$$

where τ is the shear stress, m is the consistency index and n is the power law exponent. The radial velocity profile in the tubular cross section, for a power law fluid due to pressure gradient ($p_z = -\frac{dp}{dz}$) exclusively, is given as,⁵¹

$$v_z(r) = \left(\frac{p_z}{2m} \right)^{\frac{1}{n}} \left(\frac{n}{n+1} \right) R^{\frac{n+1}{n}} \left[1 - \left(\frac{r}{R} \right)^{\frac{n+1}{n}} \right]. \quad (5)$$

The fluid flow in the tube is driven by the combined effects of an external electric field of uniform strength E_z , and a pressure gradient (p_z). Considering the combined effects, the governing equation of fully developed, one-dimensional radial velocity profile (for $0 \leq r \leq R$) under the Debye-Huckel approximation in a microtube, can be expressed as,

$$\frac{1}{r} \frac{d}{dr} \left[r \left(-\frac{dv_z}{dr} \right)^n \right] = \frac{p_z}{m} \pm \left(-\frac{\varepsilon E_z \zeta}{m} \right) \frac{\kappa^2}{I_0(\kappa R)} I_0(\kappa r), \quad (6)$$

where κ is the inverse of Debye layer thickness, $\kappa = \left(\frac{2n_\infty \phi^2 e^2}{\varepsilon k_B T} \right)^{\frac{1}{2}}$, ε is the dielectric constant of the medium, E_z is the electric field strength and ζ is the zeta potential. The $\pm$ sign represents the relative direction of the individual effect in comparison of the resultant flow direction. More precisely, the negative sign corresponds to the electric field and pressure gradient in opposite direction to each other, whereas the positive sign represents both the effects in similar direction. Integration of Eq. (6), using the boundary condition, $\frac{dv_z}{dr} = 0$ at $r = 0$, results in the following expression (presented in non-dimensional form):

$$\left(-\frac{dv_z^*}{dr^*} \right)^n = \frac{1}{n^n} \left[G^n (n+1)^n r^* \pm \frac{(\kappa R)^n}{I_0(\kappa R)} I_1(\kappa R r^*) \right], \quad (7)$$

where $r^* = \frac{r}{R}$; $v_z^* = \frac{v_z}{v_H}$; $G = \frac{v_{p_{\max}}}{v_H}$; $v_{p_{\max}}$ is the maximum average cross sectional velocity in the channel due to pressure driven flow, which is $v_{p_{\max}} = \left(\frac{n}{n+1} \right) \left(\frac{p_z}{2m} \right)^{\frac{1}{n}} R^{\frac{n+1}{n}}$. The expression of Helmholtz-Smolchowski velocity is $v_H = n \kappa^{\frac{1-n}{n}} \left(-\frac{\varepsilon E_z \zeta}{m} \right)^{\frac{1}{n}}$. This expression reduces to Helmholtz-Smolchowski velocity for Newtonian fluid.

The values of solute diffusivity considered here are in the order of 10^{-11} – 10^{-14} m²/s, which is typical for high molecular weight proteins and DNA.⁵² This results in the Schmidt number in the order of 10^5 – 10^8 . Since, mass transfer boundary layer thickness is inversely proportional to Schmidt number, it may be considered that thickness of mass transfer boundary layer would not more than 0.1–0.2% of the tube radius (typical microtube diameter are in the range of 50–500 μ m). This assumption is more consolidated in the subsequent sections. The velocity profile in the region of our interest lies within the mass transfer boundary layer, which is inherently very close to the wall.

Considering extremely thin mass transfer boundary layer, the curvature effect of the wall can be neglected. Thus, the cylindrical co-ordinate system can be reduced to Cartesian system by considering the co-ordinate system fixed at the tube wall. In order to apply the transformation, the following shift of axis is performed,

$$r = R - y, \quad (8)$$

and, in non-dimensional form, $r^* = 1 - y^*$, where, $y^* = y/R$. Hence, Eq. (7) can be modified in terms of y^* ,

$$\left(\frac{dv_z^*}{dy^*} \right)^n = \frac{1}{n^n} \left[G^n (n+1)^n (1 - y^*) \pm \frac{(\kappa R)^n}{I_0(\kappa R)} I_1[\kappa R (1 - y^*)] \right]. \quad (9)$$

It may be noted that as $G \rightarrow 0$, the combined flow becomes purely electroosmotic flow and as $\frac{1}{G} \rightarrow 0$, the flow is purely pressure driven.

A. Approximated velocity profile

As described above, for $y^* \ll 1$, Eq. (9) can be modified as,

$$\frac{dy_z^*}{dy^*} = \frac{1}{n} \left[G^n (n+1)^n \pm (\kappa R)^n \frac{I_1(\kappa R)}{I_0(\kappa R)} \right]^{\frac{1}{n}}. \quad (10)$$

Now, the above equation can be integrated, with the no-slip boundary condition (i.e., $v_z^* = 0$ at $y^* = 0$), to obtain the velocity profile.

$$v_z^* = \frac{1}{n} \left[G^n (n+1)^n \pm (\kappa R)^n \frac{I_1(\kappa R)}{I_0(\kappa R)} \right]^{\frac{1}{n}} y^*. \quad (11)$$

Equation (9) is solved numerically to get the exact velocity profile and compared with approximated velocity profile (Eq. (11)), for the range of parameters, within 0.1% of the tube radius is depicted in Fig. 2. This figure clearly shows that the actual velocity profile is linear and the approximate profile does not deviate more than 1% of the exact profile. Eq. (11) represents a linear form of the axial velocity with y^* , within the mass transfer boundary layer, which is consistent with the exact solution.

Fig. 3 represents the case of opposing effects of pressure driven and electroosmotic flow. The negative sign in Eq. (11) represents opposing flow. The region above or left of the curve indicates the electroosmotic dominated flow. For regions towards right or bottom of the curve signify the dominance of pressure driven flow. The pressure driven flow becomes dominant with G and beyond a critical limit of G (which lies on the curve in Fig. 3, corresponding to a particular rheology and κR), the flow direction is always along the pressure gradient. Similarly, electroosmotic flow becomes dominant at low G . It is evident from the figure that for shear thinning ($n=0.5$), on the left of the curve, overall bulk flow is in the direction of electric field even with larger values of G . For example, at higher electrolyte concentration, i.e., $\kappa R = 10$,

FIG. 2. Comparison of the exact and approximate velocity profiles within 0.1% of the tube diameter covering the entire range of the parameter variation (a) $n = 2.5$ and (b) $n = 0.5$.

FIG. 3. Phase space of the velocity direction in case of opposing electro-osmotic effect or vice-versa.

bulk flow occurs in the direction of electric field, even if pressure gradient is seven times stronger than the electric field ($G=7$). This behavior is diminished as the fluid rheology changes from shear thinning to thickening.

B. Solute transport in the microtube

In this work, transport of a neutral solute is analyzed in a semi-permeable microtube. The non-Newtonian fluid rheology is represented by power law model.⁵³ It is assumed that the electrical and colloidal interaction of the solutes with the channel or electrolyte is negligible. Therefore, the species balance equation within the mass transfer boundary layer at steady state is,

$$\vec{v} \cdot \vec{\nabla} c = \vec{\nabla} \cdot (D \vec{\nabla} c). \quad (12)$$

Since, the mass transfer boundary layer is thin, y -component velocity (v_y) is approximately equal to permeation velocity at the wall (v_w), i.e., $v_y \approx -v_w(x)$.⁵⁴ In Cartesian co-ordinate system, the above equation is expanded assuming constant diffusivity,

$$v_z \frac{\partial c}{\partial z} - v_w \frac{\partial c}{\partial y} = D \frac{\partial^2 c}{\partial y^2}. \quad (13)$$

Substituting the expression of v_z from Eq. (12) and non-dimensionalizing the above equation, the following equation is obtained:

$$\frac{1}{4} \left(\text{ReSc} \frac{d}{L} \right) \left(\frac{3n+1}{n+1} \right) \frac{1}{G} A(\kappa R, G, n) y^* \frac{\partial c^*}{\partial z^*} - \frac{Pe_w}{2} \frac{\partial c^*}{\partial y^*} = \frac{\partial^2 c^*}{\partial y^{*2}}, \quad (14)$$

where the dimensionless quantities are defined as, solute concentration, $c^* = \frac{c}{c_0}$; $A(\kappa R, n, G) = \frac{1}{n} |G^n(n+1)^n \pm (\kappa R)^n \frac{I_1(\kappa R)}{I_0(\kappa R)}|^{\frac{1}{n}}$; Reynolds number, $\text{Re} = \frac{\rho v_{avg} d}{\mu_{eff}}$; $v_{avg} = \left(\frac{n}{3n+1} \right) \left(\frac{p_z}{2m} \right)^{\frac{1}{n}} R^{\frac{n+1}{n}}$; Schmidt number, $\text{Sc} = \frac{\mu_{eff}}{\rho D}$ and non-dimensional permeation velocity, $Pe_w = \frac{v_w d}{D}$. The relevant boundary conditions for the above equation are,

$$\text{at } z = 0; \quad c = c_0, \quad (15a)$$

$$\text{at } y = 0; \quad v_w(c - c_p) + D \frac{\partial c}{\partial y} = 0, \quad (15b)$$

$$\text{at } y \rightarrow \infty; \quad c = c_0. \quad (15c)$$

The wall concentration ($c|_{y=0} = c_m$) is related to permeate concentration through the definition of real retention (R_r) as,

$$R_r = 1 - \frac{c_p}{c_m}. \quad (16)$$

For a solute-solvent-porous wall system, R_r is constant.⁵⁵ The boundary conditions (Eqs. (15a)–(15c)) in terms of non-dimensional quantities are

$$\text{at } z^* = 0; \quad c^* = 1, \quad (17a)$$

$$\text{at } y^* = 0; \quad Pe_w c_m^* R_r + 2 \frac{\partial c^*}{\partial y^*} \bigg|_{y^*=0} = 0, \quad (17b)$$

$$\text{at } y^* \rightarrow \infty; \quad c^* = 1. \quad (17c)$$

Considering an order of magnitude analysis, at the edge of the mass transfer boundary layer, Eq. (14) leads to an expression of thickness of non-dimensional mass transfer boundary layer, $\delta^* = \left(\frac{z^*}{A_1}\right)^{\frac{1}{3}}$, where $A_1 = \frac{1}{4} (\text{ReSc}_L^{\frac{d}{2}}) \left(\frac{3n+1}{n+1}\right) \frac{1}{G} A(\kappa R, G, n)$. Equation (14) can be solved with the similarity parameter, equals to $\eta = \frac{y^*}{\delta^*} = y^* \left(\frac{A_1}{z^*}\right)^{\frac{1}{3}}$. In terms of the similarity parameter (η), the concentration profile can be expressed as,⁵⁶

$$c^* = \frac{1 - BR_r \int_0^\eta \exp\left(-\frac{\eta^3}{9} - B\eta\right) d\eta}{1 - BR_r \int_0^\infty \exp\left(-\frac{\eta^3}{9} - B\eta\right) d\eta}, \quad (18)$$

where the constant B is represented as, $B = \frac{Pe_w}{2} \left(\frac{z^*}{A_1}\right)^{\frac{1}{3}}$. The product of $Pe_w z^{\frac{1}{3}}$ is constant, since $v_w \propto \delta^{\frac{1}{3}}$. The value of c_m^* is determined using Eq. (18), ($c^* = c_m^*$ at $y = 0$, i.e., $\eta = 0$)

$$\text{Thus, } c_m^* = \frac{1}{1 - BR_r \int_0^\infty \exp\left(-\frac{\eta^3}{9} - B\eta\right) d\eta}. \quad (19)$$

The solvent flux through the membrane is presented by Darcy's law⁵⁷

$$v_w = L_p (\Delta P_w - \Delta \pi). \quad (20)$$

The non-dimensional form of the above equation is determined by the osmotic pressure model,

$$Pe_w = \beta \left(1 - \frac{\Delta \pi}{\Delta P_w}\right), \quad (21)$$

where $\beta = L_p \Delta P_w^{\frac{d}{2}}$ and L_p is the permeability of the porous microtube. The osmotic pressure difference across the feed and permeate side is represented by $\Delta \pi$. The osmotic pressure contribution is solely due to concentration difference of the neutral solute which is transported across the porous wall, as the wall is completely permeable to the electrolytes. It may be noted that osmotic pressure due to electric-double layer interactions by charged solute is prominent.⁵⁸ Typically, the variation of osmotic pressure with concentration is either polynomial or power law form.⁵⁹ In the present analysis, osmotic pressure is expressed in power form as,

$$\pi = \alpha_1 c^{\alpha_2}, \quad (22)$$

Thus, $\Delta\pi$ is defined as,

$$\Delta\pi = \pi_m - \pi_p = \alpha_1 c_0^{\alpha_2} c_m^{*\alpha_2} [1 - (1 - R_r)^{\alpha_2}]. \quad (23)$$

The determination of c_p^* is done by the following sequence of calculations provided the following set of parameters are known.

Operating parameters: ReSc_L^d , G and ΔP ; fluid rheology: n ; solution chemistry: kR ; membrane performance parameter: R_r and β . At any particular z^* :

- (i) The value of c_m^* is guessed.
- (ii) Pe_w is calculated from Eq. (21), using Eq. (23).
- (iii) The parameter A_1 is evaluated from its definition, $A_1 = \frac{1}{4} (\text{ReSc}_L^d)^{\frac{3n+1}{n+1}} \frac{1}{G} A(\kappa R, G, n)$.
- (iv) c_m^* is calculated from Eq. (19).
- (v) Convergence between calculated c_m^* with guessed value at step (i) is checked.
- (vi) If the relative error is more than tolerance, step (ii)-(v) is repeated with the new guess value of c_m^* as calculated in step (iv).
- (vii) This process is repeated for all the discrete z^* locations from 0 to 1.

This algorithm finally results into profiles of c_m^* , c_p^* , and Pe_w as a function of tube length. Averaging over the length of the microtube using a numerical technique (for example, trapezoidal rule in this case), the length averaged quantities are determined.

The mass transfer coefficient (k) is defined as,

$$k = - \frac{D \left(\frac{\partial c}{\partial y} \right)_{y=0}}{(c|_{y=0} - c_0)}. \quad (24)$$

In terms of non-dimensional Sh , it is expressed as,

$$Sh(z^*) = \frac{kd}{D} = -2 \frac{(\partial c^* / \partial y^*)|_{y^*=0}}{c_m^* - 1}. \quad (25)$$

Using Eq. (17b), the above equation is reduced to,

$$Sh(z^*) = \frac{Pe_w c_m^* R_r}{c_m^* - 1}. \quad (26)$$

Using expression of c_m^* from Eq. (19), the following equation is obtained:

$$Sh(z^*) = \frac{Pe_w}{B \int_0^\infty \exp\left(-\frac{\eta^3}{9} - B\eta\right) d\eta}. \quad (27)$$

Substituting the value of $B = \frac{Pe_w}{2} \left(\frac{z^*}{A_1} \right)^{\frac{1}{3}}$ in above equation, we get the variation of local Sherwood number,

$$Sh(z^*) = \frac{2}{\int_0^\infty \exp\left(-\frac{\eta^3}{9} - B\eta\right) d\eta} \left(\frac{A_1}{z^*} \right)^{\frac{1}{3}}. \quad (28)$$

Hence, the length averaged Sherwood number, $\overline{Sh}_L$ is determined using Eq. (28) and substituting B from the relation $\overline{Pe}_w = 2BA_1^{\frac{1}{3}} \int_0^1 \frac{dz^*}{z^{*\frac{4}{3}}} = 3BA_1^{\frac{1}{3}}$, in terms of length averaged non-dimensional permeation rate ($\overline{Pe}_w$),

$$\overline{Sh}_L = \int_0^1 Sh(z^*) dz^* = \frac{3 \left(\frac{1}{4} \text{ReSc} \frac{d}{L} \left(\frac{3n+1}{n+1} \right) \frac{1}{G} A(\kappa h, G, n) \right)^{\frac{1}{3}}}{\int_0^\infty \exp \left(-\frac{\eta^3}{9} - \frac{\overline{Pe}_w}{3 \left\{ \frac{1}{G} \left(\frac{3n+1}{n+1} \right) \text{ReSc} \frac{d}{L} A(\kappa h, R, n) \right\}^{\frac{1}{3}}} \eta \right) d\eta}. \quad (29)$$

In case of no permeation, $\overline{Pe}_w = 0$ and purely pressure driven flow $\frac{1}{G} = 0$, the expression of average Sherwood number, simplifies to the expression of purely pressure driven flow of power law fluid in a circular tube,⁶⁰ as presented in the following equation:

$$\overline{Sh}_L = 1.018 \left(3 + \frac{1}{n} \right)^{\frac{1}{3}} \left(\text{ReSc} \frac{d}{L} \right)^{\frac{1}{3}}. \quad (30)$$

C. Enhancement of Sherwood number (for no suction $\overline{Pe}_w = 0$)

The relative improvement of mass transport with electroosmotic effect, in terms of Sherwood number can be demonstrated by considering the ratio of Sherwood number to Sherwood number for purely pressure driven flow, which defined as the enhancement factor E , in the following equation:

$$E = \frac{\overline{Sh}_L}{\overline{Sh}_L|_{G \rightarrow \infty}} = \left[1 \pm \frac{(\kappa R)^n I_1(\kappa R)}{G^n (n+1)^n I_0(\kappa R)} \right]^{\frac{1}{3n}}. \quad (31)$$

The above expression is for electric field assisting (+) as well as opposing (−) the flow. The variation of E with κR is presented in Fig. 4. The opposing flow effects clearly show a stagnation point, represented by zero E , depending on the values of G and n . It is to be understood here that the stagnation zone is the plane of zero shear or stationary plane. Considering the rheology of the fluid, Sherwood number increases as the shear thinning behavior is dominant. Sherwood number enhancement can be as large as 1.5 to 3 times for equally dominating effects ($G = 1$) corresponding to power law index $n = 0.5$ and $\kappa R = 40$. This property typically enhances the mass transport significantly. Elaborate analysis of the mass transfer with solution rheology and operating conditions is presented in the subsequent figures.

FIG. 4. Enhancement of Sherwood number with scaled Debye length for different operating conditions and flow directions corresponding to (a) $n = 2.5$ and (b) $G = 1$.

III. VARIATION OF AVERAGE WALL CONCENTRATION WITH VARIOUS OPERATING CONDITIONS

The length averaged wall surface concentration plays a major role in the dynamics of the process. The factors that affect the surface concentration are transmembrane pressure ΔP , Debye layer thickness determined by κR , solution osmotic pressure difference $\Delta\pi$, rheology n , solute diffusivity D , real retention of the porous wall R_r and $ReSc \frac{d}{L}$. However, the effect of the parameters D , ΔP and $\Delta\pi$ are known to affect mass transport, and their influence is well documented in standard textbooks. Moreover, these parameters do not affect the electrokinetic behavior of the system. Hence, these parameters are kept fixed ($\Delta P = 50$ kPa, $D = 5 \times 10^{-12}$ m²/s, $d = 10$ μ m, $\alpha_1 = 1000$ m^{3.5}.kg^{-0.5}.s⁻², $\alpha_2 = 1.5$), and effect of change of the remaining variables on the surface concentration is presented in Fig. 5. Curves 1 and 2 show the effect of κR . As expected the effect of κR is more prominent in case of electro-osmotic dominant flow regime (low G), where, the $\bar{c}_m$ grows significantly. This effect is more in case of diffused electric double layer (small κR) compared to compact EDL (curve 2). As the solution κR increases, it results to compact double layer that lies within mass transfer boundary layer, thereby offering less resistance against the solute transport. This reduces the solute concentration at wall. At lower κR , the Debye layer thickness is large which hinders the diffusion of solute over the membrane surface leading enhanced thickness of mass transfer boundary layer. To maintain a fixed $ReSc \frac{d}{L}$ while increasing G , the electroosmotic velocity must decrease. This leads to decrease in overall fluid velocity, which in turn decreases permeation. However, at higher G , the electroosmotic effect becomes negligible leading to dominance of purely pressure driven flow, and hence, permeation rate and permeate concentration become independent of κR . In case of pressure and electric field in opposing direction, (refer to Fig. 5(b)) there exists a region of stationary plane, when the axial motion of the solute ceases. When the net solute velocity approaches to zero, the convection decreases rapidly leading to high concentration polarization and membrane surface concentration. The sharp peaks ($\bar{c}_m^*$ limited to 10) are due to the stationary zone. Since, κR determines the thickness of the electric double layer, it affects the position of the stationary zone. At higher κR (lower electroosmotic effect), the stagnation point is attained at higher value of G . The locus of the stationary zone can be obtained from the phase space plot, as shown in Fig. 3.

The effect of solution rheology n , is observed by comparing curves 2 and 3. In case of shear thinning fluid, the surface concentration is less than the shear thickening case. Moreover, the surface concentration always increases with G at fixed $ReSc \frac{d}{L}$. This is because, on increasing G , the overall fluid velocity decreases ($ReSc \frac{d}{L}$ is fixed, so G can be increased only by reducing the electric field strength), leading to reduced forced convection. However, the explicit reason for decreased surface concentration in case of shear thinning fluid, cannot be stated. There are several issues responsible for such behavior, as pointed out by many researchers in

FIG. 5. Variation of the average wall surface concentration $\bar{c}_m$ for different flow configuration (a) pressure gradient and electric field assisting, (b) pressure gradient and electric field in opposing effect.

last couple of decades.⁶¹ These explanations suggest that there exists strong particle layering and molecular ordered behavior in case of shear thinning fluid flow. As observed from Fig. 5(b), in case of opposing effect, when the system approaches to stagnation, the shear thinning fluid becomes more dispersive in nature. However, in case of strong electro-osmotic effect, both the fluid rheology exhibit similar behavior, indicated by overlapping of curves 2 and 3 at lower G .

The effect of $ReSc \frac{d}{L}$ is expressed by curves 3 and 4. As the magnitude of $ReSc \frac{d}{L}$ increases, the wall concentration decreases considerably, primarily due to reason that forced convection arrests the development of concentration boundary layer over the membrane surface. The magnitude of $ReSc \frac{d}{L}$ also increases by decreasing the length of the channel. The development of the boundary layer is also affected by the length of the channel. As the channel length increases, the boundary layer develops, leading enhanced average surface concentration. The thickness of mass transfer boundary layer is inversely proportional to the $ReSc \frac{d}{L}$, and hence surface concentration increases with length. In case of opposing effect, the position of the stagnation layer remains unchanged with respect to the value of G .

The consequence of wall selectivity (R_r) on the surface concentration is represented by curves 2 and 5. By the definition of real retention, the surface which has high value of R_r has higher capacity of solute retention, which is clearly evident from the curves. In case of $R_r = 0.8$, the limiting value of $\bar{c}_m^*$ is 5, unlike the case of $R_r = 0.9$ (where the limiting value is 10).

IV. EFFECT OF OPERATING CONDITIONS ON THE SYSTEM PERFORMANCE

The variation of operating conditions and physical parameters of the system have a strong influence on the rate of permeation and permeate concentration. All the computations are performed for fixed transmembrane pressure drop, $\Delta P = 50$ kPa and diffusivity, $D = 5 \times 10^{-12}$ m²/s. The effect of κR on the length averaged permeate flux and permeate concentration is represented in Figs. 6(a) and 6(b). As the magnitude of electric field increases (decrease in G), the overall fluid velocity increases (for the assisting case) leading to enhanced forced convection and higher permeation. The effect of κR on surface concentration is illustrated in Fig. 5, which shows that at low G , the wall surface concentration is higher for lower κR , leading to more permeate concentration. At higher G , the variation of surface concentration with κR is small, which is manifested in the marginal variation of permeate concentration. However, when electric effect becomes negligibly small ($G > 10$) all the curves tend to merge and become constant. This phenomenon is attributed due to forced convection that masks the electric forces. In case of reversing flow, similar behaviour is observed for larger values of G . For pressure and electric field in opposing effects, there exists a region of stationary plane, when the axial motion of the solute ceases. When the net solute velocity approaches to zero, the convection decreases very rapidly leading very high concentration polarization and membrane surface concentration. This fact is well represented in the trend of the opposing effect curves (dotted lines). At this point, permeate concentration also becomes very high.

The next set of figures, Figs. 6(c) and 6(d) describe the effect of solution rheology on the system performance. In case of dilatant fluid ($n = 1.5$), the fluid experiences more shear stress during flow, thereby offers strong viscous resistance to solute convection, leading to higher concentration polarization and decrease in permeation. For the case of electro-osmotic effect dominating ($G < 1$), the flow profile is unaffected by viscosity. With the increase in smolchowski velocity (as G decreases), the overall fluid velocity also increases, leading to higher Pe_w . In case of large $ReSc \frac{d}{L}$ (greater than 10^6), the permeation rate for the opposing and assisting case merges for the shear thickening fluid, when κR is more than 10. For shear thinning fluid, this effect is more gradual with κR . This may be due to the interference of the pressure gradient on the electroosmotic flow profile of non-Newtonian fluid leading to origin of some sort of secondary flows.⁶² Since, physiological fluids exhibit shear thinning behavior, the assisting flow should be preferred on account of enhanced mass transfer.

The influence of $ReSc \frac{d}{L}$ on the mass transfer is described in Figs. 6(e) and 6(f). On increasing the $ReSc \frac{d}{L}$ the permeation increases. This is due to the fact that the surface concentration decreases with $ReSc \frac{d}{L}$ as shown in Fig. 5(a). Decrease in surface concentration leads to

FIG. 6. Variation of permeation and permeate concentration for different operating conditions: (a) and (b) – $R_r=0.9$; $d=10\mu\text{m}$; $n=1.5$; $\text{Re.Sc.d/L}=10^2$; $\alpha_1=1000\text{m}^{3.5}.\text{kg}^{-0.5}.\text{s}^{-2}$; $\alpha_2=1.5$ (c) and (d) – $R_r=0.9$; $d=10\mu\text{m}$; $\kappa R=20$; $\text{Re.Sc.d/L}=10^2$; $\alpha_1=1000\text{m}^{3.5}.\text{kg}^{-0.5}.\text{s}^{-2}$; $\alpha_2=1.5$ (e) and (f) – $R_r=0.9$; $d=10\mu\text{m}$; $\kappa R=20$; $n=1.5$; $\alpha_1=1000\text{m}^{3.5}.\text{kg}^{-0.5}.\text{s}^{-2}$; $\alpha_2=1.5$ (g) and (h) – $d=10\mu\text{m}$; $\kappa R=20$; $\text{Re.Sc.d/L}=10^2$; $n=1.5$; $\alpha_1=1000\text{m}^{3.5}.\text{kg}^{-0.5}.\text{s}^{-2}$; $\alpha_2=1.5$ (i) and (j) – $R_r=0.9$; $d=10\mu\text{m}$; $\kappa R=20$; $n=1.5$; $\text{Re.Sc.d/L}=10^2$; $\alpha_2=1.5$

reduction in osmotic pressure at the wall and solution interface, thereby, increasing the driving force and hence throughput. However, increase in the permeation rate is marginal at high values of ReSc_L^d . For example, $\overline{Pe}_w$ changes by 1% when ReSc_L^d changes from 10^3 to 10^5 , for $G = 1$. As thickness of mass transfer boundary layer is inversely proportional to ReSc_L^d , decrease in thickness at higher value of ReSc_L^d is gradual. Therefore, increase in rate of permeation is insignificant at higher values of ReSc_L^d . Variation of permeate concentration with ReSc_L^d is presented in Fig. 6(f). As discussed in Fig. 5, wall concentration decreases with ReSc_L^d leading to less concentration polarization and hence improves the permeation of solute. Thus, at $G = 1$, c_p^* is 0.1 for $\text{ReSc}_L^d = 10^5$ and it is 0.2 for $\text{ReSc}_L^d = 10$.

The system performance is also affected by the real retention of the porous channel, as represented in Figs. 6(g) and 6(h). As the channel wall becomes more permeable, the permeate concentration and permeation rate increase. This fact is evident from the solid curves in both the figures. More porosity in the wall increases the mass transport due to decrease in thickness of mass transfer boundary layer.^{63,64} It can be noted that for transdermal drug delivery, a relatively less porous and highly selective membrane wall ($\overline{Pe}_w < 100$ and $R_r > 0.9$) is suitable for controlling the flow behavior and consequent transport of solute by electrokinetic means.

The effect of osmotic pressure of the solute on the permeation rate is presented in Fig. 6(i). As the solution osmotic pressure increases the permeation rate decreases, as explained in context of Fig. 6(d). As the transmembrane pressure drop is fixed, higher the osmotic pressure, lower is the net driving force for transport of solute across the porous wall. It must be mentioned here that the osmotic pressure difference between the feed and permeate side is crucial. It may be noted that the electrolyte constituents has very high osmotic pressure, however, these molecules are completely permeable, hence, do not have any contribution on the overall osmotic pressure difference. Increase in osmotic pressure leads to more concentration polarization and hence solute concentration at the wall. This increases diffusive transport of solute across the porous wall and solute concentration in permeate increases. However, in the range of values of α selected in Fig. 6(j), the change in permeate concentration is marginal.

V. CONCLUSION

Solute mass transport in a porous microtube under non-Newtonian rheology is important for physiological systems and microfluidic devices. This study accurately maps the phase space of velocity direction and the interplay of thickness of electric double layer, ratio of Poiseuille to Smolchowski velocity and power law exponent. For flow where Poiseuille and electro-osmotic effects are equally dominant ($G = 1$) and in the same direction, Sherwood number increases to 3 times compared to that for pure Poiseuille flow ($\kappa R = 40$ and $n = 0.5$). Solute concentration at the wall increases with decrease in κR leading to reduction in permeation rate and enhancement of permeate concentration. Interestingly, in the case of opposing flow, there exists a stationary plane where wall concentration becomes maximum. At this condition, permeate concentration is the highest and permeate rate tends to zero. In case of shear thinning fluid, the permeation rate enhances compared to shear thickening for the flow assisting case. The permeate concentration is marginally more in case of shear thickening fluid. The permeation rate increases and the permeate concentration decreases with ReSc_L^d . The selectivity of the porous wall affects the permeation rate and concentration. When R_r is increases by 15%, the permeate concentration decreases by 5 times. The solution osmotic pressure also affects the permeation rate considerably without much change in permeate concentration. More realistic rheological fluid models can be applied to improve the predictive features of the mass transport phenomena. Also, taking into account, the overlapping of electric double layer would further enhance the accuracy of the model and make it more realistic. Validation of the model developed against transport of real life solutes in physiological systems is a future scope of this study.

NOMENCLATURE

A non-dimensional parameter as a function of $\kappa R, G, n$ is defined as $A = \frac{1}{n} \left| G^n (n+1)^n \pm (\kappa R)^n \frac{I_1(\kappa R)}{I_0(\kappa R)} \right|^{\frac{1}{n}}$

A_1	non-dimensional parameter defined as $A_1 = \frac{1}{4} (\text{Re} S c_1 \frac{d}{L}) \left(\frac{3n+1}{n+1} \right) \frac{1}{G} A$
B	non-dimensional parameter defined as $B = \frac{P e_w}{2} \left(\frac{z^*}{A_1} \right)^{\frac{1}{3}}$
c	concentration, kg/m ³
c_0	feed concentration, kg/m ³
c^*	non-dimensional concentration defined as $c^* = c/c_0$
c_m	wall surface concentration, kg/m ³
c_m^*	non-dimensional surface concentration defined as $c_m^* = c_m/c_0$
c_p	permeate concentration, kg/m ³
c_p^*	non-dimensional permeate concentration defined as $c_p^* = c_p/c_0$
d	diameter of the microtube, m
D	solute diffusivity, m ² /s
e	elementary charge, equal to 1.6×10^{-19} C
E	Sherwood number enhancement factor, defined as $E = \frac{\overline{Sh}_L}{\overline{Sh}_L _{G \rightarrow \infty}}$
E_z	electric field strength within the channel, V/m
G	non-dimensional parameter, $G = \frac{v_{pmax}}{v_H}$
I_0	modified Bessel function of zeroth order
I_1	modified Bessel function of first order
k	mass transfer coefficient, m/s
k_B	Boltzmann constant, m ² . kg. s ⁻² . K ⁻¹
L	length of the microtube, m
L_p	permeability of the porous wall, m/ Pa.s
m	flow consistency index in case of power law fluid rheology, Pa. s ⁿ
n	flow behavior index in case of power fluid rheology
n_∞	molar number concentration, M/m ³
P	axial pressure in the channel, Pa
$P e_w$	non-dimensional permeation velocity defined as $P e_w = \frac{v_w d}{D}$
$\overline{P e_w}$	length averaged non-dimensional permeation rate ($P e_w$)
p_z	axial pressure drop per unit length ($p_z = -\frac{dP}{dz}$), Pa/m
r	local radial co-ordinate inside the microtube, m
R	radius of the microtube, m
r^*	non-dimensional radial co-ordinate defined as $r^* = \frac{r}{R}$
Re	Reynolds number, $Re = \frac{\rho v_{pavg} d}{\mu_{eff}}$
R_r	real retention of the porous wall of the microtube
Sc	Schmidt number, $Sc = \frac{\mu_{eff}}{\rho D}$
Sh	Sherwood number
$\overline{Sh}_L$	length averaged Sherwood number
T	temperature, K
v_H	Helmholtz-Smolchowski velocity defined as $v_H = n \kappa^{\frac{1-n}{n}} \left(-\frac{\varepsilon E_z r}{m} \right)^{1/n}$, m/s
v_{pavg}	average cross-sectional velocity due to pressure gradient, m/s
v_{pmax}	maximum average cross-sectional velocity in the microtube due to pressure gradient, m/s
v_w	permeation velocity across the porous wall, m/s
v_y	bulk fluid velocity in the vertical direction, m/s
v_z	axial velocity inside the microtube, m/s
v_z^*	non-dimensional axial velocity defined as $v_z^* = \frac{v_z}{v_H}$
y	local Cartesian co-ordinate as defined by $y = R - r$, m
y^*	non-dimensional local Cartesian co-ordinate defined as $y^* = y/R$
z^*	non-dimensional axial co-ordinate defined as $z^* = z/L$

Greek symbols

α_1, α_2	coefficients in the polynomial expression of variation of π with concentration in Eq. (22), unit of α_1 is Pa.m ^{3α_2} .kg ^{-3α_2}
β	non-dimensional factor in Eq. (21) defined as, $\beta = L_p \Delta P_w \frac{d}{D}$
$\Delta \pi$	difference in osmotic pressure defined as $\Delta \pi = \pi_m - \pi_p$, Pa

ΔP_w	pressure drop across the porous wall of the microtube, Pa
δ^*	non-dimensional thickness of the mass transfer boundary layer
ε	di-electric constant of the solution medium
ϕ	total charge valence of the electrolyte
η	similarity parameter used for similarity transformation, $\eta = \frac{y^*}{\delta^*} = y^* \left(\frac{A_1}{z^*} \right)^{\frac{1}{3}}$
κ	inverse of Debye layer thickness, m^{-1}
μ_{eff}	effective solution viscosity, Pa.s
π	solution osmotic pressure, Pa
π_m	osmotic pressure at the wall, Pa
π_p	osmotic pressure of permeate stream, Pa
ρ	density of the solution, kg/m^3
τ_{zr}	shear stress along the axial direction, Pa
ψ	surface potential, V
ζ	zeta potential of the porous wall, V

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