Modeling of Crossflow Ultrafiltration:

Finite element analysis of concentration-polarization layer for cross-flow ultrafiltration in a cylindrical membrane pipe with impermeable segments

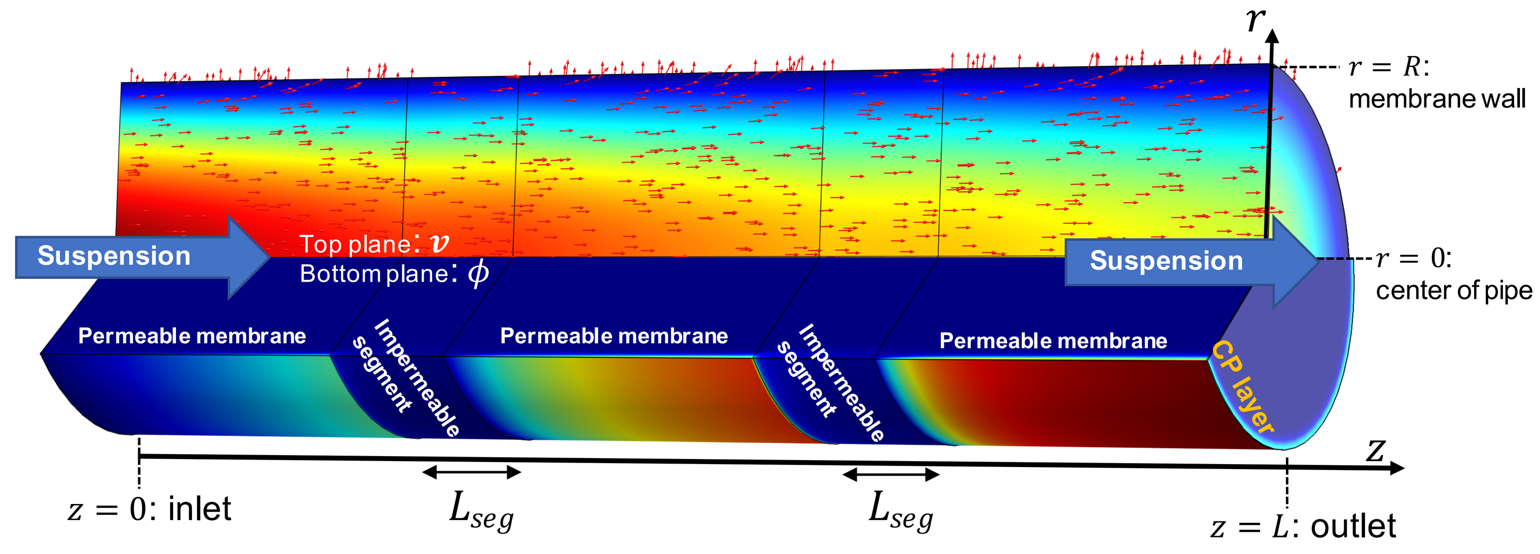
G. W. Park[1], **M. Brito**[1], **E. Zholkovskiy**[2], **and G. Nägele**[1] (e-mail: g.park@fz-juelich.de)

[1] *Institute of Complex Systems, ICS-3, Forschungszentrum Jülich*

[2] *Institute of Bio-Colloid Chemistry, Ukrainian Academy of Sciences, Kiev, Ukraine*

Abstract

Crossflow filtration is a pressure-driven separation and enrichment process of colloidal suspensions where the feed suspension is continuously pumped tangentially through a membrane pipe. The transmembrane pressure (TMP) causes permeate (solvent) to flow out through the membrane, while particles are retained inside the pipe. Consequently, a particle-enriched diffuse layer is formed near the membrane wall which reduces the filtration efficiency. This so-called concentration-polarization (CP) layer is due to the balance of flow advection of particles towards and gradient diffusion away from the membrane. The CP layer with its osmotic pressure operates against the TMP and counteracts the permeate flow. In this study, the suspension flow is described by the effective Stokes equation, and the permeate flux by Darcy's law. We use a hard-sphere model for the suspension properties such as gradient diffusion coefficient, suspension viscosity, and osmotic pressure. The coupled advection-diffusion and Stokes flow equations are solved by a finite element method (FEM) using Comsol. As a reference, we obtained analytic solutions of pure solvent flow and suspension flow where the transport properties are taken as concentration independent [1]. Furthermore, we generalize the FEM analysis to a segmented membrane with impermeable rings which we observe a weakening of the CP layer [2].



1 Modeling of crossflow ultrafiltration

1.1 Membrane geometry

- Axisymmetric cylindrical membrane pipe with length $L = 0.5m$ and radius $R = 0.5mm$ ($\epsilon = R/L = 10^{-3}$)
- In the case of a segmented membrane, two impermeable rings are tethered of the length L_{seg}

1.2 Effective Navier-Stokes equation

We use the effective Navier-Stokes equation for constant suspension mass density ρ:

$$\rho\left(\frac{\partial}{\partial t} + \mathbf{v}\cdot\nabla\right)\mathbf{v} = -P\,\mathbf{I} + \eta(\phi)\left(\nabla\mathbf{v} + (\nabla\mathbf{v})^T\right)$$

where $\eta(\phi)$ is the suspension viscosity using the hard-sphere model in [3] which depends on particle volume fraction ϕ and P is the pressure. The solvent viscosity η_s is that of water at room temperature.

1.3 Advection-diffusion equation

In the ultrafiltration regime, the diffusivity of particles is dominated by Brownian motion with the particle flux given by

$$\mathbf{j}_\phi = \phi\mathbf{v} - D(\phi)\nabla\phi,$$

where D is the long-time collective diffusion coefficient of particles. For a hard-sphere suspension, we use the short-time form [3]

$$D(\phi)/D_0 = 1 + 1.454\phi - 0.45\phi^2 + \mathscr{O}(\phi^3),$$

where $D_0 = 2.14\times10^{-11}m^2/sec$ is the single-particle self-diffusion coefficient described by the Stokes-Einstein relation. In the above particle flux, the advection-diffusion equation is

$$\frac{\partial\phi}{\partial t} + \nabla\cdot(\phi\mathbf{v}) = \nabla\cdot(D(\phi)\nabla\phi)\,.$$

1.4 Darcy's law

We describe the solvent permeable flux by an integrated version of Darcy's law,

$$v_w(z) = L_p\left(P(z) - P_{perm} - \Pi(\phi_w; z)\right),$$

where $L_p = 6.7\times10^{-10}m/(Pa\,sec)$ is the solvent permeability of the membrane, $P_{perm} = 1\,atm$ is the applied constant pressure outside the membrane wall, and $\Pi(\phi)$ is the osmotic pressure described by Carnahan-Starling equation:

$$\Pi(\phi) = k_BT\,c(\phi)\frac{1+\phi+\phi^2-\phi^3}{(1-\phi)^3},$$

where c is a number concentration of particles.

1.5 Boundary conditions

- (center) Axisymmetry: $v(r=0,z)=0$
- (wall) Darcy's law: $v(r=R,z)=v_w(z)$
- (wall) No slip along z: $u(r=R,z)=0$
- (wall) No particle flux: $-\hat{\mathbf{r}}\cdot\mathbf{j}_\phi(r=R,z)=0$
- (inlet) Particle: $\phi(r,z=0)=\phi_b$
- (inlet and outlet) Pressure: $P(r,z=0)=P_{in}$, $P(r,z=L)=P_{out}$

1.6 Operating conditions

For given geometrical parameters (L and R), membrane property (L_p), and suspension properties (D, η, and Π), the operating conditions are characterized by the longitudinal pressure difference ($\Delta_L P = P_{in} - P_{out}$) and the transmembrane pressure (TMP: $\Delta_T P = (1/L)\int_0^L (P(z) - P_{perm})dz$). The reference operating condition is $\Delta_T P$=5 kPa and $\Delta_L P$ = 130 Pa.

1.7 Finite element methods (FEM)

- We use Comsol Multiphysics with *laminar flow* and *transport of dilute species* packages using a time-dependent solver for the advection-diffusion equation
- P1/P1 elements for $\mathbf{v}$ and P, and P2 element for ϕ
- Meshes are stretched along z (with mesh-independent check)

2 Reference suspension

2.1 Assumptions

We make the following assumptions for the reference system and the operating conditions, which are similar to those in [4]:

- *Effective permeability*: $Q_{surf} \sim Q_{in}$
  (Flow rate through the membrane wall (Q_{surf}) is comparable with the flow rate through the inlet (Q_{in}))
- *Stokes flow assumption*: $\epsilon Re \ll 1$
- *Regular perturbation expansion* with respect to ϵ^0

Based on these assumptions, we derive an exact solution for the flow profile of pure solvent using a separability ansatz.

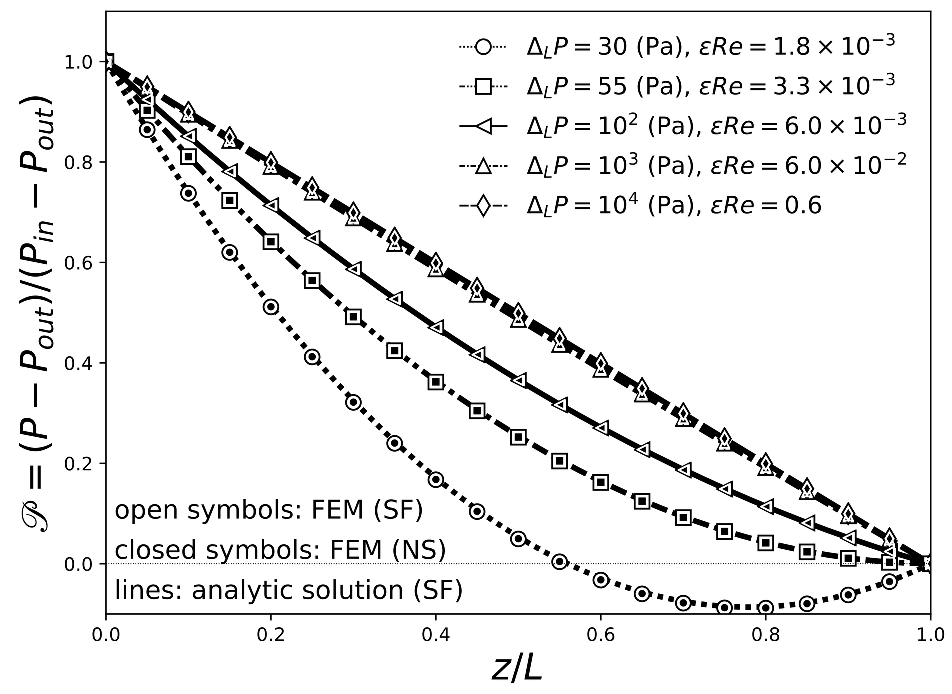


Figure 1: Pressure distribution function of the analytic solution of the Stokes equation (lines), the corresponding FEM results (open symbols) and the Navier-Stokes equation for steady-state (closed symbols) for pure solvent flow with different $\Delta_L P$ = 30, 55, 100, 1000, and 10000 (Pa) as indicated.

2.2 Boundary layer analysis (BLA)

We propose a new BLA employing the **dominant balance** between *the convective contribution from the solvent permeate flux* and *the diffusive contribution* in the advection-diffusion equation. Based on the stretching variable $\epsilon_\delta = \delta/R$ where δ is the expected thickness of the boundary layer due to this dominant balance, we obtained a solution using a singular perturbation expansion in ϵ_δ^0 [1].

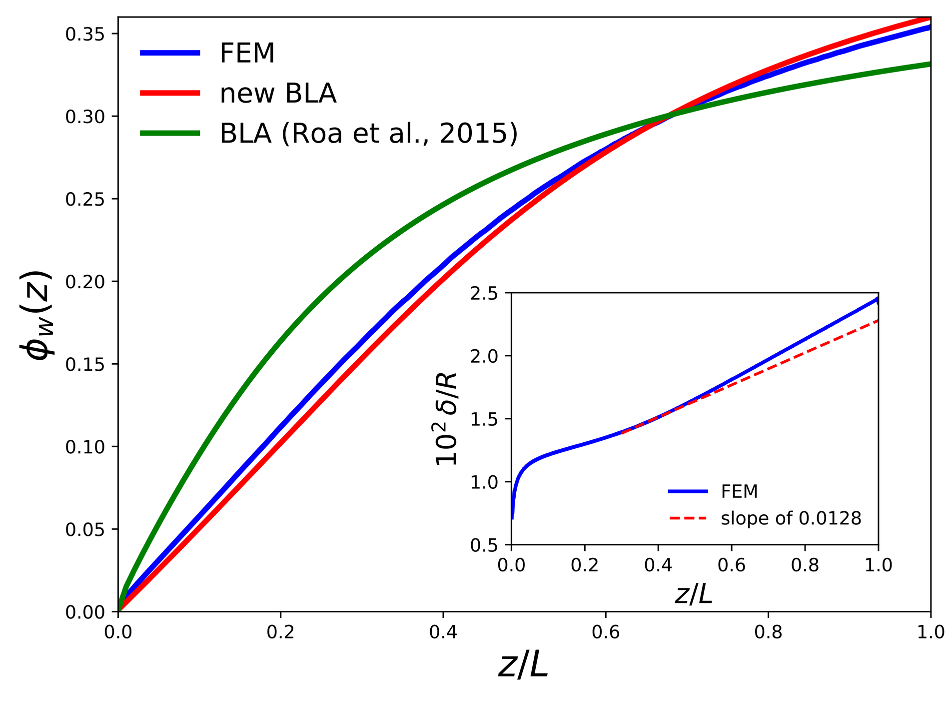


Figure 2: Particle concentration at membrane wall for constant viscosity $\eta = \eta_s$ and constant diffusion coefficient $D = D_0$. Inset shows $\epsilon_\delta = \delta/R$ obtained from FEM using $\delta(z) = \int_0^R (R-r)(\phi-\phi_b)rdr / \int_0^R (\phi-\phi_b)rdr$ (blue), and its expected slope on the new BLA (red).

2.3 Concentration-dependent transport properties

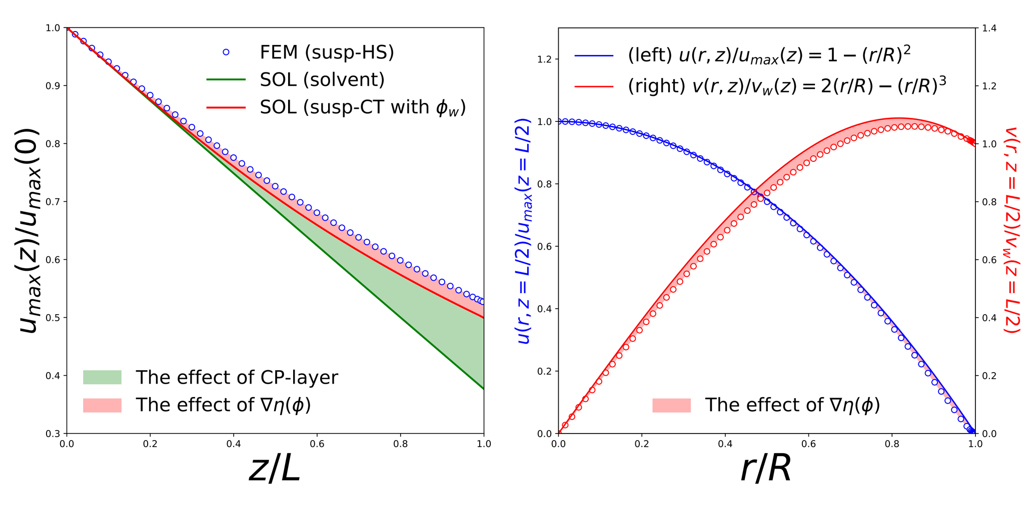


Figure 3: FEM results of a hard-sphere suspension (open symbol) and associated solutions for the suspension flow for constant transport properties (lines) shows at the normalized longitudinal velocity along the centerline $u_{max}(z) = u(r=0,z)$ (left), and normalized longitudinal and transversal velocities along the radial direction (right).

3 Segmented membrane pipe

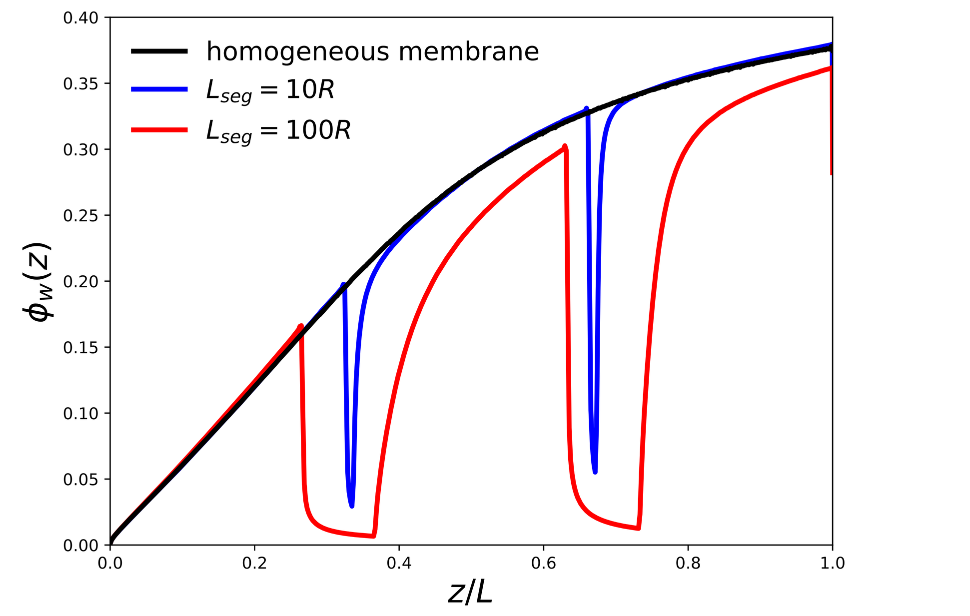


Figure 4: Membrane concentration of particles in a homogeneous membrane pipe (black), and a membrane pipe with impermeable rings of length $L_{seg} = 10R$ (blue), and $L_{seg} = 100R$ (red) using a hard-sphere model.

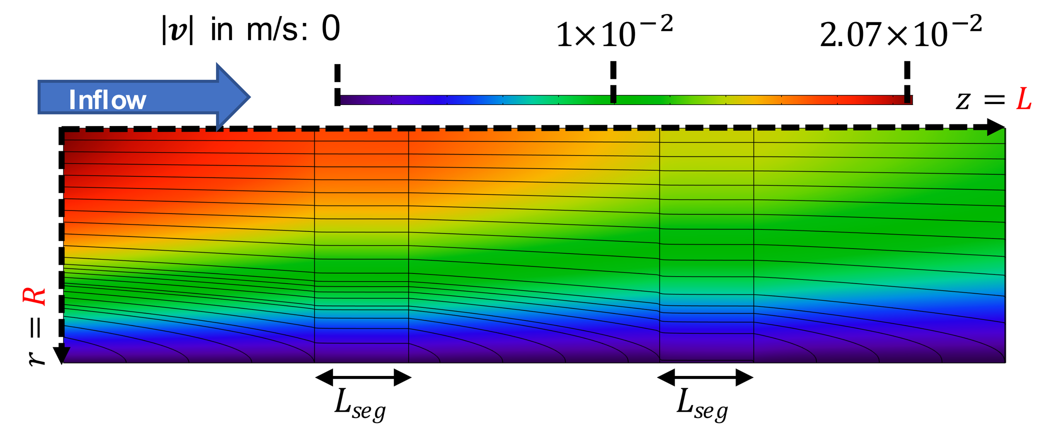


Figure 5: Suspension flow streamlines of a hard-sphere suspension pumped through the segmented membrane of length $L_{seg} = 100R$. Upper bar is $|\mathbf{v}|$.

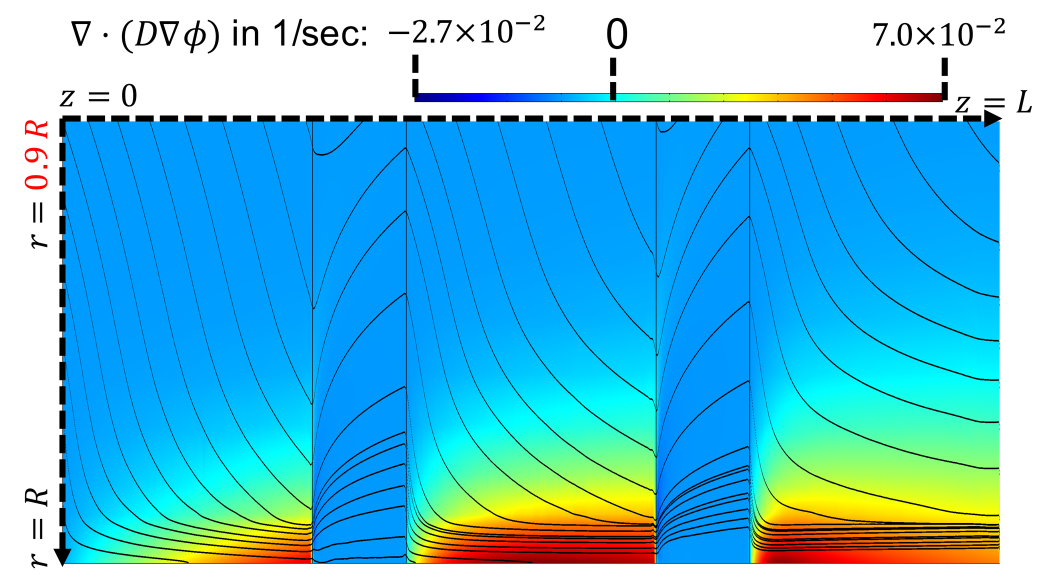


Figure 6: Diffusive flux contribution $\nabla\cdot(D\nabla\phi)$ to the advection-diffusion equation (colors), and streamlines of the particle flux $\mathbf{j}_\phi$ (lines) near the membrane wall for $0.9R \le r \le R$.

Concluding Remarks

1. We have obtained an exact solution for the solvent flow using a separability ansatz with the assumptions (i) of effective permeability, (ii) Stokes flow, and (iii) regular perturbation expansion with respect to ϵ^0.
2. Based on assumptions of (i-iii), we developed a new boundary layer analysis making use of the dominant balance between the transversal (solvent permeate) convection contribution and the diffusion contribution in the advection-diffusion equation.
3. The quantitative agreement between FEM results and reference solutions supports the validity on FEM in this model. We have extended FEM case for a membrane with segmented by impermeable rings.
4. In the impermeable segmentation, a relaxing CP layer is observed where the particles are diffusing away from the membrane wall. The flow profile in the impermeable ring region is similar to that of the Hagen-Poiseuille flow.

Acknowledgments

The authors gratefully acknowledge financial support from the Deutsche Forschungsgemeinschaft (SFB-985, Project B6).

References

[1] G. W. Park and G. Nägele, work in progress.

[2] G. W. Park, M. Brito, E. K. Zholkovskiy, and G. Nägele, work in progress.

[3] R. Roa, E. K. Zholkovskiy, and G. Nägele, "Ultrafiltration modeling of non-ionic microgels", Soft Matter **11**, 4106–4122 (2015).

[4] N. Tilton, D. Martinand, E. Serre, and R. M. Lueptow, "Incorporating Darcy's law for pure solvent flow through porous tubes: Asymptotic solution and numerical simulations", AIChE Journal **58**, 2030–2044 (2012).