Towards simulation of gas separation modules based on hollow fiber membranes

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Abstract. Processes of C\textsubscript{1}-C\textsubscript{4} hydrocarbons gas separation with membranes yielding in methane and C\textsubscript{3+} fractions are highly attractive. The significant breakthrough on this issue can be achieved by the development of novel gas separation modules based on C\textsubscript{3+} hydrocarbons-selective hollow fiber membranes. The efficiency of this technology significantly depends on the module design, which should take into account the mass and heat transfer processes within the module with the simultaneous external constrained flow in the fiber system, the internal flow in hollow fibers, and molecular transport in the selective membrane layer. The developed simulation model includes the theory of convective diffusion transport in the shell-side constrained flow in hollow fiber membrane modules with an ordered arrangement of fibers. The simulated dependencies of the Sherwood number for circular fiber in a fiber row arranged in a transverse laminar flow on the Reynolds, Peclet and Schmidt numbers, and membrane mass transfer coefficient will be presented.

1. Introduction
Processes of C\textsubscript{1}-C\textsubscript{4} hydrocarbons gas separation with membranes yielding in methane and C\textsubscript{3+} fractions are highly attractive [1]. The novel polyalkylmethylsiloxanes are promising membrane materials for this task [2,3]. However, the significant breakthrough on this issue can be achieved by the development of novel gas separation modules based on C\textsubscript{3+} hydrocarbons-selective hollow fiber membranes. The efficiency of this technology significantly depends on the module design, which should take into account the mass transfer processes within the module with the simultaneous external flow in the fiber system, the internal flow in hollow fibers, and molecular transport in the selective membrane layer. In this work, we consider the convective transport and retention of a substance presented in a fluid (gas or liquid) flow past a fibrous medium with a given regular arrangement of fibers. We simulate the flow and concentration fields and calculate the deposited fraction of molecules (point particles) defined as a fiber retention efficiency. This quantity depends on dimensionless Reynolds, Re = \textit{a}2\textit{U}/\nu, Peclet Pe = ReSc and Schmidt numbers Sc = \textit{v}/D, where D is the particle diffusion coefficient, a is the fiber radius, \nu is the kinematic viscosity, U is the inlet flow velocity.
Moreover, the retention efficiency strongly depends on the porosity of the fiber media. When dealing with hollow-fiber membranes, one has to include inter-membrane transport into consideration.

2. Governing equations
Hydrodynamics and convective mass transfer in fibrous media are intensively studied since fibrous media are used in different technologies (hollow-fiber gas separation modules [4,5], membrane contactors [6,7], sorption filters and fibrous filters for removing suspended particles in gases [8, 9] and liquids). Models with a well-defined flow field are used in studying the hydrodynamics of real porous media. The use of models eliminates the effect of uncertainty on the structure of the porous medium. Arrays with regular fiber arrangement provide an opportunity to quantitatively study the constrained flow field and thus the efficiency of absorption of a solute. As models in which the neighboring fibers significantly affect the flow field, systems of parallel fibers perpendicular to the laminar flow, with square or hexagonal packing, or the simplest model - a single row of parallel fibers - are commonly used.

![Figure 1. Laminar transverse 2D flow past a row of parallel fibers in the simulation cell: the values of the streamlines are 10–4 (curve a), 0.2 (b), 0.5 (c), 1 (d), 1.5 (e); Re = 20, a/h = 0.5; the arrow indicates the flow direction.](image)

In this work, the 2D laminar transverse (cross) flow and concentration fields in model fibrous media are simulated for a single row of parallel fibers, perpendicular to the direction of viscous incompressible fluid (Figure 1). The dimensionless stationary Navier-Stokes equations were solved numerically.

\[
\Delta \mathbf{u} - 0.5 \text{Re} (\mathbf{u} \cdot \nabla) \mathbf{u} = \nabla p, \quad \nabla \cdot \mathbf{u} = 0
\]

where \(\Delta\) is the Laplace operator, \(\nabla\) the Nabla operator, \(\mathbf{u}\) the flow velocity vector, \(p = p^* a / U \mu\) the pressure, \(\mu\) is the dynamic viscosity. Asterisk * denotes dimensional values. Hereinafter, all values were reduced to a dimensionless form by normalization to the fiber radius \(a\) and the inlet velocity of the flow \(U\). The no-slip condition was set as the boundary condition on the fiber surface, \(u = 0\). The condition of an unperturbed flow was set at the input boundary of the cell, \(x = -X\), while the condition of the absence of viscous stresses was set at the outlet, \(x = X\). On the side faces of the cell, symmetry conditions were set for the velocity components. To determine the distribution of the concentration of a substance in a stream, together with Eq. 1, the stationary elliptic equation of convection-diffusion was solved

\[
2\text{Pe}^{-1} \Delta C - \mathbf{u} \cdot \nabla C = 0
\]

where \(C = C^* / C_0\) is the dimensionless concentration in the stream, \(C_0\) the inlet concentration, \(\mathbf{u} \cdot \nabla C = u \frac{\partial C}{\partial x} + v \frac{\partial C}{\partial y}\). On the fiber surface at \(r = 1\) two types of boundary conditions were set, the full absorption condition:
and the flux condition
\[ \frac{\partial C}{\partial r} = kC \]  

(4)

Here \( r \) and \( \theta \) are dimensionless polar coordinates, the angle \( \theta \) is measured counterclockwise, the \( k \) parameter is the “reaction rate constant”, describing the trans-membrane transport in the hollow-fiber membrane. The first condition (3) can be also obtained for the case of constant wall concentration \( C_w \) after the following change of the variable \( C = (C^* - C_w)/(C_0 - C_w) \). At the cell inlet the condition of uniform inlet concentration was set, \( C = 1 \), while the condition \( \partial C/\partial x = 0 \) was set at the cell outlet. On the side faces of the cell, symmetry conditions were set. The equations (2) and (3) were solved with the help of the monotone stable finite-difference scheme derived by Berkovski and Polevikov in [10]. This scheme was employed for aerosol filtration modeling in a wide range of Peclet numbers for creeping flow conditions in [11].

The overall retention (collection) efficiency of a fibrous layer with a thickness \( H \) is related with the penetration coefficient and the dimensionless fiber retention (collection) efficiency \( \eta \) by the formula [9]:

\[ E = 1 - C/C_0 = 1 - \exp(-2alH\eta) \]

where \( C/C_0 \) is the penetration coefficient, i.e. the ratio between the outlet to inlet solute concentrations, \( l = \alpha/\pi a^2 \) is the length of fibers per unit surface, \( L = lH \) the total fiber length per unit surface for the layer of fibers, \( \alpha \) the packing density of fibers. The fiber collection efficiency is found by integrating the local density of the diffusion flux of particles (per unit fiber length) over the fiber surface:

\[ \eta = 2\pi \frac{Sh}{Pe} = 2\pi \left. \frac{\partial C(r, \theta)}{\partial r} \right|_{r=1} \; d\theta \]  

(5)

where \( Sh \) is the mean Sherwood number [12]. Here, Sherwood number and fiber collection efficiency possess different dependences on Peclet number: \( Sh \sim Pe^{\nu} \), \( \eta \sim Pe^{-2/3} \).

3. Results of simulations and discussions
Figure 2 gives an example of calculated \( \eta \) as functions of Re at fixed Sc for the case of a relatively loose system of fibers. It is seen from Figure 2 that the slope of the \( \eta(\text{Pe}) \) curves varies with Re since the curves become nonlinear. This is explained by mixing in the wake region. Hydrodynamic wake modifies the diffusion wake and reduces the effect of concentration polarization, pressing the concentration lines towards the fiber.
Figure 2. Fiber retention efficiencies vs. the Reynolds number at different Schmidt numbers $Sc = Pe/Re = 10$ (1), $100$ (1′), $1000$ (1″): $a/h = 0.2$; solid lines (1, 1′, 1″) – direct simulations with account for Re-number effects, dotted lines (2–2″) – by the asymptotic formula at high Peclet numbers, $Re \ll 1$ [13], curve 3 – by the asymptotic formula at low Peclet numbers, $Re \ll 1$ [14].

The absorption at low Peclet numbers, which characterizes the transport in gases at low Reynolds and Schmidt numbers, is also considered. In Figure 2, the transition from a convection-diffusion transport to a full diffusion transport at low Peclet numbers is illustrated. In this case, the curvature of streamlines has a negligible effect on the transport, and the fiber retention efficiency curves reach the plateau at $Pe \to 0$ [14]. Here, the fiber retention efficiency is equal to the inlet flux, which, in dimensionless form, is equal to the distance between the axes of the neighbor fibers.

In Figure 3 the dependencies of the fiber retention efficiency on the “reaction rate constant” $k$ from the Neumann (membrane flux) boundary condition (4) are plotted. This figure illustrates the role of the trans-membrane selective transport of the substance deposited on the streamlined fiber from the external “shell-side” flow. The calculations were performed for different Re and Sc at a fixed $a/h$. It can be seen from the figure that the dependence of $\eta$ on $k$ is rather sharp in the convectively dominant region at large $Pe$ (in the region of small and intermediate $k$). With the growth of $k$ the solution tends to the limit corresponding to the solution found with the boundary condition for complete absorption (3). For small Peclet numbers (small Re and Sc), the considered dependence changes only slightly, since diffusion in the external region dominates, and the contributions of convection and trans-membrane transport (described by the parameter $k$) are small.
Figure 3. Fiber retention efficiency vs. “reaction rate constant” \( k \) at \( Re = 20 \) (1–3) and 0.01 (1’–3’); \( Sc = 1000 \) (1, 1’), 10 (2, 2’), 1 (3, 3’), \( a/h = 0.5 \).

Conclusions

Numerical simulation of the flow field and convection-diffusion mass transfer in a row of parallel fibers, arranged normally to the direction of the flow, was performed by solving the Navier-Stokes and convection-diffusion equations in a wide range of the fiber packing density \( a \), and the fiber Reynolds number up to \( Re = 100 \). Two types of boundary conditions for concentration at the outer fiber surface were employed – the constant concentration condition (Dirichlet), \( C = 0 \), and the flux boundary condition (Neumann) \( \partial C/\partial r = kC \), which depends on the solute transport characteristics both in the shell-side flow and in the hollow-fiber membrane. It was shown that the use of the complete absorption condition \( C = 0 \) gave an upper estimate for the absorption (retention) efficiency. At the same time, the calculations with the two boundary conditions of Dirichlet and Neumann types gave significantly different results for high and intermediate Peclet numbers, i.e., they revealed different power dependences of the mean Sherwood number on \( Pe \). At low Peclet numbers, in the diffusion dominant region, the fiber retention efficiency calculated with both boundary conditions tends to a constant value inversely proportional to the row blockage ratio, \( h/a \). It should be noted, that the calculation of specific hollow fiber membranes requires the knowledge of membrane parameters and their transport properties since the account for the trans-membrane transport of a substance noticeably modifies the results of simulations for the retention efficiency. The simulations are compared with existing analytical asymptotic solutions for low and high Peclet numbers, \( Pe = ReSc \), in the range of Schmidt numbers covering the cases of gas and liquid flows, \( Sc = 1 – 1000 \). The simulations reveal complex multi-parametric dependencies of the fiber absorption (retention) efficiency in conditions of the confined flow, which is strongly influenced by the neighbour fibers. In our future studies, multi-fiber configurations with inner (lumen) flows will be considered. The results of the work will be used to design and optimize hollow fiber membrane modules.

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