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Numerical modelling of concentration polarisation and cake formation in membrane filtration processes

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HIGHLIGHTS

- A unifying model for predicting concentration polarisation and cake formation
- A novel coupling of Lattice Boltzmann - Finite volume schemes is developed
- Model is validated with theoretical and experimental results from literature

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ABSTRACT

A model capable of predicting concentration polarisation and cake formation in cross-flow membrane filtration is proposed. The cross-flow hydrodynamics is resolved by solving the Navier–Stokes equations and convection–diffusion equation is used to resolve the solute distribution. A Lattice-Boltzmann (LB) scheme is used for the hydrodynamics and this is coupled with an LB scheme or a finite volume (FV) scheme for modelling the cake formation. The equations are coupled through the velocity and the viscosity, which is assumed to vary with the solute concentration. Concentration polarisation is modelled for interacting nano-particles by using concentration dependence on diffusion coefficient and osmotic pressure gradient as a function of solute concentration. Cake formation phenomenon is predicted for interacting nano-particles and non-interacting solute particles using the same approach. The proposed model is validated by comparing the results obtained for a number of problems with the predictions produced by other computational models and experimental results available in literature.

1. Introduction

The importance of membrane filtration in industries has increased enormously during the past three decades. The first use of membrane technology can be dated back to the middle of 18th century when Nollet discovered the phenomenon of osmosis. Most of the early studies on membrane filtration were carried out using natural materials like animal's bladders. The commencement of practical applications using membrane technology started only during the middle of 20th century. Depending on the size of the particles segregated and the operating pressures, membrane filtration can be classified into micro-filtration (MF), ultra-filtration (UF), nano-filtration (NF) and reverse osmosis (RO) processes. There is an extensive amount of research related to the membrane filtration process being carried out. This includes testing of new membrane materials, use of different pore sizes, use of different trans-membrane pressures (TMP), and methods to minimize fouling. Typically, the design of a new membrane is a time consuming process that requires considerable resources to consider all the aspects that affect the performance of the membrane. Some of these aspects are related to the decline in permeate flux due to cake formation, concentration polarisation, pore blocking, etc. Furthermore, the experimental testing that is usually needed to find the optimum operating conditions for the filtration process will have to be repeated for each different type of solutes. The development of simulation techniques and advancement of computer power provides a very good alternative to the time consuming and costly experimental studies. Therefore, modelling can be used in the optimization of membrane filtration process and to reduce the number of experimental testing required.

There are several models available in the literature for modelling cross flow and concentration polarisation in the membrane filtration. In the present paper, only membrane fouling aspects like cake formation and concentration polarisation models are considered for nano-particles. Chudaeck and Fane [1] used the resistance model of the filtration theory to describe the flux decline in cross-flow UF. Their numerical solution was based on experimentally determined steady state flux which is used as fitting parameter. Porter [2] used simplified convection–diffusion equation that takes into account the back diffusion phenomenon during cross-flow filtration process. Even this model uses mass transfer coefficient that has to be determined experimentally. Zydeny and Colton [3] proposed that the flux is reduced due to the

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formation of concentration polarisation and their model is based on shear induced diffusion which is a major back diffusion mechanism in MF [4]. These models are analytical and one dimensional in nature where often experimental data is needed.

Lee and Clark [5] developed a two dimensional model by solving 2-D convection diffusion equation using appropriate numerical schemes and assuming a cross-flow velocity field in UF. Their model is based on particle mass transfer which increases the thickness of the cake layer. Their model was successful in predicting the flux decline for different operating conditions but it fell short in accounting for solute–solute interactions. Bhattacharjee et al. [6] proposed a model that takes solute–solute interactions into consideration in NF. They embedded the interactions in the diffusion coefficient and osmotic pressure and solved 2-D convection diffusion equation using finite difference scheme. But this model is limited to TMP where no cake formation is observed and hence, flux decline with cake formation cannot be predicted. Kim et al. [7] proposed a model that takes the Derjaguin, Verway, Landau and Overbeek (DLVO) interactions between solute particles into consideration in NF. Their model is based on the equilibrium distance between the solute particles in the cake that effects the porosity of cake layer. The flux decline in time is predicted by a simplified mass balance equation which results in a closed form proportional to inverse of the square root of time.

Most of these models can be used only when the filtration cells have simple rectangular or circular geometries. When the design is complex, for instance if the filtration cell has spacers for promoting turbulence, these models will not be sufficient to predict the process. Hence, robust and rigorous techniques are needed to develop more generic models. Huang and Morrissey [8] and Lee and Clark [5] used finite elements and finite difference schemes to solve 2-D convection diffusion equation respectively. But they assumed a velocity profile in the cross-flow direction in their works. Geraldes et al. [9] used finite volume methods to solve for both cross-flow velocity and concentration distribution for a simple channel flow. But their model needed very fine mesh and is applicable to relatively thick concentration boundary layers. Kromkamp et al. [10] developed a model that solves cross-flow hydrodynamics and particle distribution in the filtration cell in a coupled manner for MF. Their model is based on Lattice Boltzmann methods (LBM) and can be applied to complex geometries effectively.

Despite the existence of a variety of models in literature, a more generic model that can form the bases for the simulation of all aspects of membrane filtration is missing. Most of the abovementioned models used different approaches for predicting the flux decline depending on the governing phenomenon of flux decline. In this paper a generic model is proposed which can be used as blackbox model for membrane fouling. The proposed model is based on Kromkamp model which is used in MF for predicting concentration polarisation. The model is extended to predict the concentration polarisation in NF and cake formation and associated flux decline. The present model is validated with the experimental results from the different experiments available in the literature. Although the present model can be easily extended to complex 3D geometries, as the first step, only 2D geometries are considered.

2. Mathematical model

Membrane filtration is a pressure driven process where the suspended particles in the fluid medium are separated by semi-permeable membrane. The system can be described by modelling fluid and suspended solute particles. The hydrodynamics of the fluid can be resolved by solving the Navier–Stokes (NS) equations, which may be expressed as

$$\frac{\partial \rho}{\partial t} + \operatorname{div}(\rho u) = 0 \tag{1}$$

$$\frac{\partial(\rho \boldsymbol{u})}{\partial t} + (\mathbf{u} \cdot \operatorname{grad})(\rho \boldsymbol{u}) = -\operatorname{grad} p + \nu(\phi)\nabla^2(\rho \boldsymbol{u}) + \boldsymbol{b}. \tag{2}$$

Here, ρ is the density, $\boldsymbol{u}$ is the velocity, $\nu(\phi)$ is kinematic viscosity, p is the pressure and $\boldsymbol{b}$ is the body force per unit mass. To couple the solute model to the fluid, kinematic viscosity is assumed to be dependent on the local solute concentration. This coupling is accomplished here using the model

$$\nu(\phi) = \nu_0 \left[1 + 1.5 \frac{\phi}{(1-\phi/0.6)}\right]^2 \tag{3}$$

that was proposed by Romero and Davis [11]. In this model, ϕ denotes the volume fraction of the solute and ν_0 is the viscosity of the fluid in the absence of any solute. The value 0.6 in the denominator is derived from the maximum possible packing fraction of spherical particles.

The distribution of the solute particles is resolved by solving the convection diffusion (CD) equation

$$\frac{\partial \phi}{\partial t} + (\boldsymbol{u} \cdot \operatorname{grad})\phi = D(\phi)\nabla^2\phi \tag{4}$$

where $D(\phi)$ denotes the diffusion coefficient for the solute particles.

The appropriate choice of the diffusion coefficient is the key to an effective implementation of the model. Cho et al. [12] determined that, for particle sizes less than 0.1 μm, Brownian motion dominates, while shear induced diffusion dominates when the size is more than 0.5 μm. The shear induced diffusion coefficient in the filtration device can be obtained as

$$D = 0.33\ \dot{\gamma}\, a^2 \phi^2 \left(1 + 0.5 e^{8.8\phi}\right) \tag{5}$$

where a denotes the particle radius, and $\dot{\gamma}$ represents the shear rate. The shear rate is obtained from the computed velocity field. For particle sizes less then 0.1 μm, we employ the Brownian diffusivity

$$D(\phi) = D_\infty K(\phi) \frac{d}{d\phi}(\phi Z) \tag{6}$$

where D_∞ is the diffusion coefficient for a single solute particle, $K(\phi)$ is the sedimentation coefficient and $Z(\phi)$ is the osmotic compressibility. The diffusion coefficient for a single particle is given by Stokes–Einstein equation

$$D_\infty = \frac{kT}{6\pi\eta a} \tag{7}$$

where, k is the Boltzmann constant, T is the absolute temperature, η is the dynamic viscosity of fluid and a is the radius of the solute particles. The sedimentation coefficient, $K(\phi)$, can be expressed, in terms of the solute volume fraction, as

$$K(\phi) = (1-\phi)^\alpha \tag{8}$$

where the coefficient α is determined from the properties of the interacting solute [6]. The osmotic compressibility, $Z(\phi)$, is given by

$$Z(\phi) = \frac{\Pi}{nkT} \tag{9}$$

The osmotic pressure, Π, is determined using the virial equation of state [13], which requires the evaluation of a radial distribution function where n is the number density of the solute particles. the number density of particles is defined as the number of solute particles in a unit volume. The radial distribution function is evaluated using the Ornstein–Zernike (OZ) integral equation, obtained from the van der Waal and electrostatic double layer interaction forces. Bhattacharjee [6] provides full details on computing the osmotic pressure and the diffusion coefficient and solving the OZ integral equation.

Starting from an assumed initial concentration distribution, the equations are advanced in time. At each time step, the NS equations are solved first to obtain the velocity, $\boldsymbol{u}$. This velocity is then used in the CD equation to obtain the concentration distribution and, hence, the local viscosity using Eq. (3). A brief description of the Lattice