Development of a predictive mathematical model for crossflow filtration of colloidal suspensions

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Abstract

The step-by-step development of a Galerkin finite element model for crossflow filtration of colloidal particles is explained. Important features of the process from both the fluid dynamical and rheological points of view are discussed and the solution methods that were adopted to provide realistic computer simulations for crossflow filtration of colloids are presented. The developed algorithm takes into account the process dependant nature of the flow regime, i.e. combined free and Darcy flows in a crossflow filter, by updating the viscosity of the fluid as a function of particle volume fraction distribution. Hence any change in the volume fraction due to filtration and any knock on effects on the rheological behaviour can be continually taken into account. The algorithm has been developed with generalised Newtonian suspensions in mind. To simulate the filtration of a suspension the equation used to relate fluid viscosity to particle volume fraction should be defined separately. In this way a realistic model of the process can be obtained. Similarly the model has the potential to include the commonly adopted back transport models such as shear-induced diffusion. The paper includes samples of simulation results and discussions concerning their validity.

1 Introduction

Crossflow filtration has been researched since the 1950’s when Sourirajan developed reverse osmosis to prove his theory that salt could be removed from seawater without a phase change [1]. Over 50 years later, novel developments and refinements in membrane technology continue to be active themes of research. These advances are rapidly enlarging the capabilities to restructure production processes, protect the environment and public health, and provide new technologies for sustainable growth. Membrane technologies now play an
increasingly important role as unit operations for resource recovery, pollution prevention and energy production. They are also used in environmental monitoring, quality control, fuel cells and bio-separation applications.

Fundamentally as shown in Figure 1, crossflow membrane filtration involves a suspension flowing tangential to the filter surface. This crossflow initially provides enough shear to encourage particles away from the membrane surface. Although with time, continued particle migration leads to a build up on the filter surface causing a decline in the permeate flux. This process is generally known as ‘fouling’. It is observed that fouling by colloidal and larger particulate materials in crossflow filtration is considerably less than that predicted for porous media by the convective diffusion equation, known as the ‘flux paradox’. This paradox has not been fully explained by anyone although various people have described backtransport mechanisms such as inertial lift on the particles, shear-induced diffusivity originating from particle-particle interactions, and effects of particle-membrane surface chemistries as the likely causes [2–6] However the generated models have so far tended to be for suspensions with uniformly shaped particles and known particle size distributions.

An integral part of separation processes is the fluid transport phenomena that occur while the various separation mechanisms are at work. The performance of a crossflow filter is inherently linked to the fluid motion through its volume, and thus the geometrical configuration is an important design consideration. In this respect laws governing the flow of liquids through uniform, incompressible beds serve as a basis for the development of formulae for the more complex, non-uniform, compressible cakes, which occur in many filtration operations.

Fluid dynamical description of free flows usually does not cause a problem and in a great majority of instances the well known Navier-Stokes equations can be used to model these sections. The fluid dynamics underlying filtration processes are complex, however, models to interpret the processes all stem from Darcy’s law. When Darcy’s law is combined with the appropriate continuity equations a fluid dynamic model for such processes is formed.

In the present paper a computer simulation methodology for crossflow filtration is outlined. This methodology is based on the de-coupled solution of the Navier-Stokes equation with a porous wall boundary condition (i.e. Darcy equation) and the convective-diffusion equation while updating for viscosity variations due to filtration. This procedure is developed using rational approximations and depends on mathematical techniques based on the finite element method.
2 Mathematical model

The main purpose of mathematical modelling is to represent the physics of a given problem in terms of a set of equations in conjunction with boundary and initial conditions. Due to the complex nature of the crossflow filtration process, the development of a universal mathematical model which can take into account all of the physical features of this process is a prohibitively difficult task. Consequently, efforts have been concentrated on the development of a crossflow filtration model that is based on rational approximations and therefore the starting point for the development of a complete filtration model.

Consider an incompressible, homogeneous suspension under steady state, laminar, and isothermal flow in a two dimensional domain with a permeable wall. The flow regime is described by the following set of equations.

2.1 Mass conservation

The continuity equation is the expression of the conservation of mass for incompressible fluids and in a Cartesian co-ordinate system \((x, y)\) is written as:

\[
\frac{\partial v_x}{\partial x} + \frac{\partial v_y}{\partial y} = 0
\]  

(1)

where \(v_x\) and \(v_y\) are the components of the velocity field.

2.2 Equations of motion

In the absence of body forces the components of the equation of motion in a Cartesian co-ordinate system representing the transient flow of viscous fluids are given by:

\[
\begin{align*}
\rho \frac{\partial v_x}{\partial t} &= - \frac{\partial p}{\partial x} + \frac{\partial \tau_{xx}}{\partial x} + \frac{\partial \tau_{yx}}{\partial y} \\
\rho \frac{\partial v_y}{\partial t} &= - \frac{\partial p}{\partial y} + \frac{\partial \tau_{xy}}{\partial x} + \frac{\partial \tau_{yy}}{\partial y}
\end{align*}
\]  

(2)

where \(\rho\) is the fluid density, \(v_x\) and \(v_y\) are the components of the velocity, \(p\) is the pressure and \(\tau_{xx}, \tau_{xy}\), etc. are the components of the stress tensor.

2.3 Rheological equations

It's reasonable to assume that the suspension fed into the filter system behaves as a generalised Newtonian fluid. Then the components of the stress tensor in the equation of motion are given in terms of the rate of deformation tensor as:

\[
\tau_{xx} = 2\eta \frac{\partial v_x}{\partial x}
\]  

(3)

\[
\tau_{yy} = 2\eta \frac{\partial v_y}{\partial y}
\]  

(4)
The shear dependant viscosity of the suspension is found by the Power-Law equation:

$$\eta = \eta_0 \dot{\gamma}^{n-1}$$

Where $\eta$ is the shear dependant viscosity of the fluid. In the present simulations the shear dependant viscosity of the suspension is found by the Power-Law equation:

$$\tau_{xy} = \tau_{yx} = \eta \left( \frac{\partial v_x}{\partial y} + \frac{\partial v_y}{\partial y} \right)$$

Where $\eta$ is the shear dependant viscosity of the fluid. In the present simulations the shear dependant viscosity of the suspension is found by the Power-Law equation:

$$\eta = \eta_0 \dot{\gamma}^{n-1}$$

Where $\eta_0$ is the zero shear viscosity (consistency coefficient), $n$ is the power law index, and $\dot{\gamma}$ is the shear rate.

Eqns (1) – (6) can be used to obtain the velocity and pressure fields in the feed stream (free flow) section of the filter domain. However within the filter there is also particle migration to consider. This migration can be predicted using the well-known convective-diffusion equation.

### 2.4 Convective-diffusion equation

In filtration the migration of particles is of relevant importance, as it is this mechanism that affects the hydrodynamical behaviour of the fluid system and causes the problems with fouling and alters the filtration rates.

Assuming constant diffusivity the convective-diffusion equation is represented by:

$$\frac{\partial c}{\partial t} + v_x \frac{\partial c}{\partial x} + v_y \frac{\partial c}{\partial y} = D \left( \frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} \right)$$

Where $c$ is the concentration, $D$ is the diffusion coefficient and $v_x$ and $v_y$ are the components of the velocity field.

### 2.5 Particle volume fraction distribution (PVFD)

This distribution is found via the following conservation equation for PVFD. A source term is included on the right hand side of the equation to take into account changes in PVFD during filtration.

$$\frac{\partial \Phi}{\partial t} + v_x \frac{\partial \Phi}{\partial x} + v_y \frac{\partial \Phi}{\partial y} = S(\Phi, c)$$

Assuming that the concentration in the filter increases at a constant rate as the filtration progresses then a reliable source term in eqn (8) could be given by a first order kinematic equation as:

$$S(\Phi, c) = k \Phi (\Phi + c)$$

The relationship between the relative viscosity and the PVFD is expressed by Einstein’s equation. The multiple of the PVFD is an experimentally determined parameter, $\Lambda$. 
In a filtration domain the above model combining of eqns (1) – (10) should be solved subject to the following boundary conditions.

3 Boundary conditions

Boundary conditions are essential to all mathematical problems. Without them the working equations of the problem cannot be solved. It is therefore important that the correct boundary conditions are set. In this problem it is necessary to know the velocity field and the concentration field at the boundaries of the filter or the problem domain. Relatively speaking it is fairly easy to set the velocity field boundary conditions, however for the concentration field it is has proved to be the main difficulty of the problem. The problem being that finding the concentration at the porous wall is in essence the aim of the problem. With the porous wall being a boundary it causes complications with the working equations. Figure 2 shows a schematic of the filter and indicates using letters (a) – (f) where the boundary conditions are.

3.1 Velocity boundary condition

With velocity, it is made simple by assuming that there is no slip at the solid wall and it is stress free at the exit of the filter. Also by imposing the Darcy equation as the boundary condition for the Navier-Stokes equation along the porous wall the combined nature of free/porous flow regimes in this system is taken into account:

**Boundary (a)** A known inlet velocity.

**Boundary (b)** Along this wall it is assumed that there is no slip and hence the velocity components are set to zero.

**Boundary (c)** The same conditions are given as for boundary (b).
Boundary (d) The Darcy equation is imposed (for details see ref. [7]).
Boundary (e) The same conditions are given as for boundary (b).
Boundary (f) Stress free conditions are imposed. It is assumed that section (e) is long enough to justify this condition.

3.2 Concentration boundary condition
Unlike with the velocity field the only boundary where one can be sure about the concentration is at the inlet. Particles are constantly moving, the movement is mainly due to hydrodynamical convection, although when the particles get close to the porous wall other forces become significant. The particles may then attach themselves to the porous surface. Hence a build up of particles may form on the surface of the wall. With time this build up is likely to increase, however, as mentioned in the introduction it is possible for particles to escape from the wall and back into the bulk flow (backtransport). Due to this complexity we need to calculate concentration variations along the porous wall and a predetermined profile cannot be imposed. It may however, be possible, as shown later to impose a method, i.e. Von Neumann boundary condition along the porous wall. However at the porous wall boundary we wish to calculate the concentration profile not impose one. The potential solution to this problem is discussed later.

In the present solution scheme the following boundary conditions were used:

Boundary (a) At the inlet the concentration is known and the concentration field is given.
Boundary (b) A constant concentration value is imposed.
Boundary (c) A constant concentration value is imposed.
Boundary (d) No boundary condition is imposed and the concentration value is free to be determined by the solution to the working equation.
Boundary (e) The same conditions are given as for boundary (d).
Boundary (f) The same conditions are given as for boundary (d).

4 Development of the working equations of the finite element schemes
The development of the working equations of the fluid flow model has been reported previously and will not be discussed here [7].

The novel aspect of this work is to include filtration dependant viscosity in this model. This is done through the relationship between the particle volume fraction and the relative viscosity of the fluid being filtered in an algorithm described later in this paper.

Using the weighted residuals Galerkin finite element method in a two-dimensional domain, \( \Omega_c \), subject to boundary limits of \( \Gamma_c \), the weighted residual statement of the convective-diffusion equation is written as:

\[
\int_{\Omega} W_f R_n \, d\Omega = 0
\]  

(11)

where
\[ R_{\Omega} = \frac{\partial \bar{c}}{\partial t} + v_x \frac{\partial \bar{c}}{\partial x} + v_y \frac{\partial \bar{c}}{\partial y} - D \frac{\partial^2 \bar{c}}{\partial x^2} - D \frac{\partial^2 \bar{c}}{\partial y^2} \]  

(12)

Where

\[ \bar{c} = \sum_{i=1}^{r} c_i(t) N_i(x, y) \]  

(13)

In the Galerkin method the weight function \( N_i \) and the interpolation function \( N_j \) are identical. The discretised domain can be written as:

\[ \sum_{e=1}^{E} \Omega_e = \Omega, \quad \text{and}, \quad \sum_{e=1}^{E} \Gamma_e = \Gamma \]  

(14)

Where \( E \) is the total number of elements and \( e \) represents an individual element.

At this stage the formulated Galerkin weighted residual statement contains second order derivatives. Thus using \( C^0 \) elements derivative of the interpolation function will be discontinuous across element boundaries and the integral of their second derivative will tend to infinity. To resolve this difficulty the second order terms are integrated by parts to obtain:

\[ \int_{\Omega_e} N_j N_i \left( \frac{\partial c_i}{\partial t} \right) d\Omega_e + \int_{\Omega_e} N_j N_i \left( v_x \frac{\partial c_i}{\partial x} + v_y \frac{\partial c_i}{\partial y} \right) d\Omega_e + \int_{\Gamma_e} N_j D \left( \frac{\partial N_i}{\partial x} \frac{\partial c_i}{\partial y} - \frac{\partial N_i}{\partial y} \frac{\partial c_i}{\partial x} \right) d\Gamma_e = \]  

(15)

Where \( n_x, n_y \) are components of the unit vector normal to the boundary [8] and \( c^e \) is the known concentration from the previous time step.

The elemental working equations of the convective-diffusion equation is hence given as:

\[ [M] \{ \dot{c} \} + [K_c] \{ c \} = \{ F_c \} \]  

(16)

Where \([M]\) is the mass matrix, \([K_c]\) is the stiffness matrix, \( \{ c \} \) and \( \{ \dot{c} \} \) are the solution vector of the concentration field and its first order time derivative, respectively, and \( \{ F_c \} \) is the load vector. The members of these matrices are given as:

\[ (M)_{ij} = \int_{\Omega_e} N_j N_i dxdy \]  

(17)
\[ (K_c)_{ij} = \iint_{\Omega_x} \left\{ N_j \left( v_x \frac{\partial N_i}{\partial x} + v_y \frac{\partial N_i}{\partial y} \right) + \right. \]
\[ D \left( \frac{\partial N_j}{\partial x} \frac{\partial N_i}{\partial x} + \frac{\partial N_j}{\partial y} \frac{\partial N_i}{\partial y} \right) \right\} dx dy \]  
\[ (F_c)_{i} = \int_{e} N_j \left[ k \left( \frac{\partial c^e}{\partial x} n_x + \frac{\partial c^e}{\partial y} n_y \right) \right] d\Gamma_e \]  
\[ (18) \]
\[ (19) \]

Eqn (19) is provides a ‘natural’ boundary condition for the convective-diffusion equation and can be used to impose a concentration condition along the porous wall.

5 Outline of solution strategy

The solution strategy used in the present work can be summarised by the following steps.

1. The entire domain of interest is first discretised into a mesh of finite elements.
2. The sets of initial values for the nodal velocity, pressure and concentration fields in the solution domain are assumed and stored as input arrays. An array containing the boundary conditions along the external boundaries of the solution domain is prepared and stored. (For the first step values of \( c^e \) in eqn (19) are identical to the initial.
3. The time variable is updated, incrementing by \( \Delta t \).
4. The flow equations are solved and the velocity and pressure fields are obtained.
5. The convective-diffusion equation is solved using the velocity field found in step 4 and then the concentration field is updated.
6. The particle volume fraction equation is solved, and the viscosity field is updated accordingly.
7. The convergence of the scheme is checked and steps 4 to 6 are repeated if necessary.
8. The time variable is incremented by \( \Delta t \) and the value of the field variables are updated to be used as the new initial conditions.
9. If the current time becomes greater than the pre-selected final process time, the calculations are terminated otherwise steps 3 to 9 are repeated.

6 Sample results and discussion

In the solution of the concentration equation new ways of imposing a necessary boundary condition at the porous wall are investigated. There are three possible ways that is being discussed at present. (a) To treat the line integrals, eqn (19) as unknowns and re-integrate into the left hand side of eqn (15).
However this is a laborious technique and will only be used as a last resort, (b) To make use of virtual elements [8], which allow the possibility of avoiding the calculation of line integrals. However this may result in unacceptable approximations in the present study, or (c) To make the solution scheme transient and modify the boundary condition so that the algorithm uses the previous value. The initial value in this case could be zero. This is the simplest case.

Colloidal suspensions have a low diffusion coefficient and therefore the convective-diffusion equation in the present model is mainly dominated by the convection terms. Therefore, to obtain stable solutions it may be necessary to use streamline upwinding.

A FORTRAN-90 computer program has been developed to implement the aforementioned algorithm. Figure 3 shows a graphic example of one set of results. The program was run with the following conditions set: (i) Consistency coefficient in the power law model = 0.84 and the Power Law index = 0.64, (ii) Density = 1055 m$^3$kg$^{-1}$, (iii) Kinetics characteristic time = $106.2 \text{s}$, (iv) Diffusion coefficient set to a constant value of 63.0E-13, (v) The boundary conditions were set as (according to Figure 2); Velocity: (a) $v_x = 0.1 \text{ m/s}$ and $v_y = 0 \text{ m/s}$, (b) $v_x = 0 \text{ m/s}$ and $v_y = 0 \text{ m/s}$, (c) $v_x = v_y = 0 \text{ m/s}$, (d) $v_x = 0 \text{ m/s}$ and $v_y = -0.061 \text{ m/s}$, (vi) The boundary conditions were set as (according to Figure 2); Concentration: (a) $c=5$, (b) $c=1$, (c) $c=5$, (d, and e) $c$ set as a ‘free’ boundary condition, (vii) Dimensions: Width = 9mm and Length =18mm. Note the dimensions that were used are not usual filter dimensions but and were only set to demonstrate the model.

The x-axis in Figure 3 represents the width of the filter, where the width is measured outward from the solid wall. Each curve shown (A-E) indicates the concentration profile along the width at a known length of the domain. Curve A shows the concentration profile at the entrance to the filter; Curve B shows the
concentration profile at the start of the porous wall; Curves C and D show the concentration profile within the porous section; Curve E shows the concentration profile at the end of the porous wall and Curve F shows the concentration profile after the porous section.

The model has been verified without a porous wall boundary condition. The algorithm gave expected results for flow in a pipe. On the application of a porous wall the results so far have been promising. Results to be published soon.

7 Concluding remarks

A macroscopic continuum approach for the simulation of crossflow filtration is developed. This approach is shown to give concentration profiles within the filter domain and hence provide a fast engineering technique for the analysis and design of crossflow filters.

Future work is to further explore new techniques of imposing appropriate boundary conditions along the porous wall. The effects of geometry on the filtration rate can be studied using this work as a basis.

It is envisaged that this work will result in the completion of another step towards the creation of a complete mathematical model for crossflow filtration.

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References