EFFECTIVE PERMEABILITY OF MULTI-CHANNEL CROSS-FLOW FILTRATION MEMBRANES FROM PERMEATE FLUX MEASUREMENTS USING THE BOUNDARY INTEGRAL METHOD

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The problem of fluid transport inside ceramic, porous, multi-channel microfiltration membranes has been studied experimentally and computationally. The transport of permeate in a seven (star-shaped) channel tubular membrane is modeled according to Darcy's equation for flow in porous media; the resultant Laplace equation is solved numerically on the complex, multi-connected geometry of interest using the Boundary Integral Method (BIM). Based on this model, we propose an efficient and novel procedure for determining the effective permeability (K_m) of the membrane module by comparing computational results with on-line experimental measurements of permeate flux at various levels of Trans-Membrane Pressure (TMP). Besides determining K_m , the proposed approach can be used to identify alternative designs and operating conditions in membrane modules of arbitrary geometrical complexity.

Introduction

Cross-flow filtration is a widely used method for the separation of phases in many types of suspensions, for example in water and sewage treatment, harvesting of plant and animal cells, food processing and dewatering of mineral slurries (Fordham & Ladva, 1989; Kroner *et al.*, 1984; Lu & Ju, 1989; Pritchard *et al.*, 1992). Cross-flow filtration has important applications in the filtration of both sub-millimeter and sub-micron particles. Numerous experimental and theoretical studies on cross-flow filtration have been reported and a comprehensive review is beyond the purpose of this communication (Hunt *et al.*, 1987; Belfort *et al.*, 1994). Such experimental studies are typical in that they have concentrated on cake formation, its interaction with the flow on the side of the retentate and its effect on permeate flux. This approach may be valuable in giving a qualitative insight into the mechanisms governing membrane fouling, but neglects fluid transport inside the membrane material itself as well as the effect geometrical parameters of the membrane may have on filtrate flux. The total hydraulic resistance of a membrane (R_t) is usually expressed in the form

$$R_t = R_c + R_m \quad (1)$$

where R_m is the resistance of the membrane itself and R_c is the cake resistance. The former is usually determined

from dead-end filtration of particle-free liquid in single-tube arrangements (Nagata, *et al.*, 1989) and is considered a material property rather than a process characteristic. The shape of the membrane channels and, in particular, their spatial arrangement within a module are ignored in such analyses, even though they can influence profoundly the efficiency of multiple-channel membrane modules, in which the overall effective filtration area may be only a small fraction of the apparent area, as a result of competing flows within the membrane material itself. The objective of this communication is to develop and apply a model suitable for filtrate transport into a general multi-channel membrane, whose flow channels are of arbitrary cross-sectional shape. In particular, we consider channels of circular as well as of star-shaped cross-section in a seven-channel membrane of circular total cross-section. The Boundary Integral Method (BIM) employed for this purpose allows for consideration of channels of arbitrary geometrical complexity with no (or very little) extra effort and, for linear problems such as those considered in this study, is faster and more accurate compared to domain methods (such as finite differences or finite elements). Having developed and tested such a model, we propose a procedure for determining the effective permeability of a multi-channel tubular ceramic membrane by combining on-line measurements of filtrate flux (using particle-free water) with BIM computations for filtrate transport in the membrane. Section 2 is an outline of the experimental procedure. An overview of the BIM, as applied to the solution of linear flow in porous media is presented in Section 3. Finally, the numerical and experimental results are presented and discussed in Section 4.

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Fig. 1 Schematic representation of the ceramic crossflow filtration module

1. Experimental

1.1 The membrane module

The ceramic element at the heart of the membrane module is shown schematically in **Fig.1**. It consists of a tubular, multichannel α -alumina substrate whose (seven) channels have a star-shaped cross-section. It is claimed by the manufacturer (Faurey Industrial Ceramics Ltd., UK) that the star shape of the flow channels increases the filtration area of the membrane and has the effect of inducing turbulence at lower crossflow velocities, thus controlling the thickness of the cell layer deposited on the membrane surface during operation. The housing of the module is made of stainless steel. Each membrane module had a filtration area (S) of 0.06 m^2 .

1.2 Experimental Setup and Procedure

A schematic of the experimental setup is illustrated in **Fig.2**. The cell suspension reservoir was a 5 L, temperature-controlled agitated vessel. A gear pump (Watson Marlow Micropump, type 500 3U), operated at its maximum speed (500 RPM), was used to circulate de-ionized water from the reservoir, whose temperature was maintained at 25°C . A gear pump was chosen instead of a peristaltic one because it yields a constant flow rate to the filtration unit. The transmembrane pressure was varied from 0.05 to 0.3 MPa by changing the settings of a valve located at the exit of the filtration module and was measured via a pressure gauge. The flowrate into the filtration unit was maintained at the maximum value of $50 \times 10^{-6} \text{ m}^3\text{sec}^{-1}$ during all experiments. In all experimental runs, the permeate flux (J) was determined by measuring the volume of permeate, ΔV , collected in a time period Δt , and dividing by the surface area of the membrane.

Since cross-flow filtration is a pressure-driven process, the effect of pressure on permeate flux is paramount (Caridis & Papathanasiou, 1997). In the absence of fouling, the resultant permeate fluxes are only affected by the permeability of the membrane

Fig. 2 Schematic diagram of the apparatus used in the experimental program

material and, in multi-channel modules, by the geometrical arrangement of the channels. In our experiments, the transmembrane pressure was varied from 0.05 to 0.3 MPa by partially closing the back pressure valve; this range is typical of such operations (Kroner *et al.*, 1984). The permeability of the membrane (K_m) was then determined by correlating the experimental results on retentate flow at various pressures with predictions based on a solution of Darcy's flow in the actual geometry of the seven-channel, star-shaped module of **Fig.1** using the Boundary Integral Method. The permeate was collected at atmospheric pressure and thus the transmembrane pressure was effectively the back-pressure of the system.

2. The Mathematical Model

The starting point for the development that follows is Darcy's law for flow in porous media (Darcy, 1856). This is an empirical equation which states that fluid velocity in a porous medium is linearly proportional to the local pressure gradient. The constant of proportionality is termed the permeability (K) and its magnitude is a function of the porosity and internal geometry of the medium. For two-dimensional flow in an isotropic medium, Darcy's law can be written as:

$$u(x, y) = \frac{K}{\mu} \left(\frac{\partial P}{\partial x} \right) \quad (2)$$

$$v(x, y) = \frac{K}{\mu} \left(\frac{\partial P}{\partial y} \right) \quad (3)$$

where (u) and (v) are the superficial velocities (the velocities one observes macroscopically) in the (x) and (y) directions respectively, K is the permeability of the medium and (μ) is the fluid viscosity. The pressure

gradients in Eqs.(2)-(3) are the local gradients at the location (x,y) . Differentiating Eqs.(2) and (3) with respect to (x) and (y) respectively and after addition and consideration of the incompressibility condition for the permeating fluid, one obtains the following potential equation for pressure:

$$\nabla^2 P = 0 \quad (4)$$

where (∇^2) is the Laplacian operator and where it is implied that the permeability of the membrane medium is spatially uniform. In membranes with a gradient or multi-layer structure, this assumption implies that the permeability obtained by comparing numerical predictions to experimental data is an effective average.

The solution of Eq.(4) using the Boundary Integral Method has been well documented in the literature (Brebbia & Dominguez, 1992). The major advantage of the boundary integral technique over domain methods is that for linear applications only discretization of the boundaries of the domain under consideration is necessary. Hence mesh generation in the BIM is fairly simple, which is especially advantageous when moving and/or complex-shaped boundaries are considered. A further advantage is the high accuracy of the method; the derivation of the Boundary Integral Equation (BIE) is exact until the boundary integrals are numerically evaluated with the use of appropriate shape functions. The boundary integral method is based on the solution of the Boundary Integral Equation:

$$c_i P_i = \int (-P \frac{\partial P^*}{\partial n} + P^* \frac{\partial P}{\partial n}) d\Gamma \quad (5)$$

in which the parameter c_i equals $1/2$ if the point under consideration is on a smooth boundary, c_i equals 1 if the point is placed inside the domain and c_i equals zero, if the point is located outside the domain. P^* is the solution of the singularly-forced Laplacian:

$$\nabla^2 P^* = -\Delta_i \quad (6)$$

where Δ_i represents the Dirac delta function. The solution of Eq.(6), termed the fundamental solution of the Laplace equation, is obtained by applying the method of Fourier transform and for two-dimensional applications is:

$$P^* = \frac{1}{2\pi} \ln\left(\frac{1}{r}\right) \quad (6.a)$$

Here (r) represents the distance from point (i) to the point under consideration. The BIE is further discretised by use of shape functions and the resulting system of

Fig. 3 Schematic diagram of the coaxial-cylinder geometry used in the validation of the numerical results. The boundary conditions on the external and internal cylinder surfaces ($r=R_2$ and $r=R_1$) are given by Eq.(7) or Eq.(10)

linear equations is solved numerically for the unknown values of the potential (P) and/or of its normal derivative $(\frac{\partial P}{\partial n})$ on all boundary segments.

3. Results and Discussion

3.1 Model validation

3.1.1 Comparison with analytical solutions The BIM code for the numerical solution of Eq.(4) was benchmarked by comparing its predictions with existing analytical solutions in an annulus with inner radius $R_1=1$ and outer radius $R_2=10$. The relevant geometry is shown in **Fig. 3**. In particular, the numerical solution is compared to analytical solutions of the Laplace equation subject to the boundary conditions:

$$P(R_1) = 2x \quad \text{and} \quad P(R_2) = 1.01x \quad (7)$$

It can be shown that such a solution is:

$$P(x,y) = x \cdot \left[1 + \frac{1}{x^2 + y^2}\right] \quad (8)$$

Numerical predictions were compared to this solution along the line $(\theta=0)$ and along the arc $r=(R_1+R_2)/2$. **Table 1** contains the results of the comparison, in terms of the maximum relative percent error (RPE):

$$R.P.E \equiv \left(\frac{P_{\text{num}} - P_{\text{an}}}{P_{\text{an}}}\right) \cdot 100 \quad (9)$$

where the subscripts (num) and (an) indicate numerical and analytical solutions respectively.

When the boundary conditions are constant values of the potential (P) on the surfaces of the bounding cylinders

$$P(R_1) = A \quad \text{and} \quad P(R_2) = B \quad (10)$$

an analytical solution to Eq.(4) subject to conditions (10) is:

Table 1 Comparison of the results between numerical and analytical solution of the Laplace equation subject to Dirichlet conditions given by Eq.(7)

Number of elements	Maximum R.P.E
50	1.7
110	0.92
230	0.54
440	0.13
620	0.06

Table 2 Comparison of the results between numerical and analytical solution of the Laplace equation subject to boundary conditions of Eq.(10)

Number of elements	Maximum R.P.E
50	0.91
110	0.42
230	0.10
440	0.013
620	0.006

$$P(r) = m \cdot \ln(r) + k \quad (11)$$

where

$$m = \frac{A - B}{\ln(R_1/R_2)} \quad \text{and} \quad k = A - \frac{(A - B)}{\ln(R_1/R_2)} \cdot \ln(R_1) \quad (12)$$

The numerical code was also tested by comparing its predictions corresponding to the boundary conditions Eq.(10) to the analytical solution given by Eqs.(11) and (12) along the line ($\theta=0$). **Table 2** contains the results of the comparison, in terms of the maximum relative percent error observed.

3.1.2 Mesh refinement In the absence of analytical solutions for permeate flow in the star-shaped ceramic membrane used in this study, one quarter of which along with a 170-element mesh is shown in **Fig.4**, the developed BIM code was validated by examining the convergence of the numerical solution with mesh refinement. The boundary conditions used were constant pressure ($P=1.5$) along the inner surfaces, symmetry conditions along the lines BC, DE and FA (Fig.4) and ambient (zero) pressure along the external surface (line AB). Calculated was a normalized flux (Q^n), defined as the integral of the outward-normal pressure derivative along the boundary (AB):

$$Q^n = \sum_{j=1}^N \left(\frac{\partial P}{\partial n} \right)_j \delta l_j \quad (13)$$

where the subscript (j) indicates the j^{th} element (of length δl_j) along the boundary (AB). **Figure 5** demonstrates that the numerical solution to the problem

Fig. 4 One quarter of the star-shaped ceramic membrane add a spare with a representative 170-Element Boundary Element Mesh

Fig. 5 Results of a mesh refinement study on the star-shaped membrane of Fig. 4. Plotted is the normalized flux (Q^n) given by Eq.(13)

converges rapidly with mesh refinement. Based on this, a mesh with 340 elements was used in all subsequent simulations.

3.2 Experimental results

A number of experiments was carried out in the filtration system presented in section (2) using particle-free distilled water. The retentate flow generated by the gear pump was around $50 \cdot 10^{-6} \text{ m}^3/\text{s}$. This resulted in a pressure drop in the membrane module of around 400 Pa (calculated using Poiseuille equation for laminar flow in a tube) while the back-pressure in the system was varied between 0.2 Bar (0.02 MPa) to 3 Bar (0.3 MPa). Under these conditions, the pressure inside the membrane module is practically equal to the back-pressure of the system. Considering that the permeate exits at ambient pressure, the system back-pressure also gives the transmembrane pressure which drives the filtration process. The results of a number of experiments with deionized water are shown in **Fig.6** as graphs of permeate flux per unit area of the membrane (J) vs. transmembrane pressure (ΔP_{tm}). It is customary in the analysis of crossflow micro-filtration using resistance models to obtain a "permeability coefficient - P.C" as the slope of such a graph (Nagata *et al.*, 1989), based on the equation:

$$J = (P.C) \cdot \Delta P_{tm} \quad (14)$$

Comparing this with the original Darcy model (Eqs.(2)-(3)), it is evident that the thus obtained permeability coefficient is not the ratio (K/μ) , but includes a length scale, which is the characteristic length over which the trans-membrane pressure is applied. This may be deduced from the geometry in simple filtration systems (such as the coaxial cylinder one) but it is not known a-priori in more complex membranes such as the multi-channel one used in our study. This is so not only because the geometry of the flow channels is not circular but also because a multi-channel arrangement does not allow for an a-priori determination of the regions through which permeate flow occurs. Therefore, the permeability (K_m) of the membrane material cannot be deduced unambiguously from crossflow filtration experiments in actual commercial membrane modules by use of a permeability coefficient as in Eq.(14). The BIM model developed in this study will be used subsequently for this purpose. In the context of Eq. (14), linear regression based on the data of Fig. 6 yielded a value of $1.18 \cdot 10^{-9}$ m/s/Pa for the permeability coefficient (P.C) of our seven-star-shaped-channel membrane. This is comparable to the results of Nagata *et al.*, (1989), for certain ceramic membranes. The intercept in Fig. 6 is calculated to be $-4.96 \cdot 10^{-2}$; this gives a value of 4.2 kPa as the pressure required to initiate permeate flow. If, based on Fig. 4, we take as the length over which the trans-membrane pressure is applied the average distance of the outer ring of channels from the outside surface of the filter (measured to be 0.002 m) and take (μ) as the viscosity of water (0.0009 Pa.s at 25 °C), we obtain for the membrane permeability a value $K_m \sim 2 \cdot 10^{-15}$ m². This is similar to the value obtained by fitting experimental results for permeate flux to the boundary element predictions (following section) as well as to previously reported resistance values for crossflow filtration membranes (Prichard *et al.*, 1992).

3.3 Numerical predictions

A number of simulations in the geometry of Fig. 4 were carried out for various values of the retentate pressure. The predicted relationship between the normalized permeate flux, as defined by Eq. (13), and the pressure in the retentate side of the membrane (which is equal to the TMP) is shown in Table 3. The computational results show a linear relationship, in accordance to the experimental data of the previous section. The variation of the outward-normal pressure gradient, which is a measure of the local permeate flux, along the arc (AB) is shown in Fig. 7. Evidently, the permeate flux is not constant along the circumference of the membrane module but is highest in the vicinity of the flow channels.

Fig. 6 Experimental results showing the dependence of permeate flux on retentate pressure using deionised water and a seven-channel membrane with star-shaped channels (Fig.1)

Fig. 7 Variation of the normal pressure gradient (a measure of the permeate flux) along the circumference of the tubular membrane (Line AB in Fig. 4)

The numerical predictions for Q^n , along with the experimental flux measurements at the corresponding retentate pressures, allow for the determination of the ratio (K/μ) in the membrane. The local normal permeate superficial velocity (u_n) along the line (AB) is given by:

$$u_n = \left(\frac{K}{\mu}\right) \left(\frac{\partial P}{\partial n}\right) \quad (15)$$

The corresponding permeate flux therefore is:

$$J = \int_L u_n dL = \left(\frac{K}{\mu}\right) \sum_{j=1}^N \left(\frac{\partial P}{\partial n}\right)_j \delta l_j = \left(\frac{K}{\mu}\right) Q^n \quad (16)$$

where Q^n is defined by Eq. (13) and is calculated from the BIM model. The effective permeability of the membrane material is therefore calculated as:

$$K = \frac{J_{exp} \cdot \mu}{Q^n} \quad (17)$$

Table 3 Calculation of membrane permeability from experimental and numerical results

Retentate pressure (MPa)	Experimental Flux ($\times 10^6 \text{ m}^2/\text{s}$)	Computed Q'' ($\times 10^{-6} \text{ Pa}$)	Permeability (K) ($\times 10^{15} \text{ m}^2$)
0.1	7.45	2.13	3.15
0.15	10.67	3.19	3.01
0.2	14.53	4.26	3.07
0.25	18.42	5.32	3.12
0.3	21.92	6.38	3.09
Average(K): $3.09 \times 10^{-15} \text{ m}^2$			

where (J_{exp}) is the experimental permeate flux per unit length of the membrane module. Based on Eq.(17), K is determined from experiments and BIM computations at a number of retentate pressures between 0.1 and 0.3 MPa. Table 3 summarizes the results. (μ) is taken as the viscosity of water at 25 °C (0.0009 Pa.s).

There is a reasonable agreement between the predicted values at different pressures as well as between the above average result and the value for (K) obtained in the previous section through the use of an arbitrary (intuitive) value of the characteristic length of the system ($K \sim 2 \times 10^{-15} \text{ m}^2$).

3.4 The Effect of Channel Shape and Size

The values of the effective permeability obtained previously are used to examine the effect of channel shape and size on the performance of the filtration module. Changes in the flow patterns (fluid velocity and transition to turbulence) in the side of the retentate caused by the change in the shape/size of the flow channel are not considered, since the focus is on the permeate flow inside the membrane and since, when pure water is considered, the formation of a cake and its interaction with the flow of the retentate is non existent. Two membranes with circular channels, whose diameters are those of the inside- ($R=0.001\text{m}$) and outside-circles ($R=0.002\text{m}$) corresponding to the star-geometry, are considered. The relevant geometries and the BIM meshes are shown in Figs.8 and 9. The case of circular channels with total filtration area equal to the original star-shaped membrane (0.06 m^2) is also considered. In this case the radius of the circular channels is 0.00227m . The corresponding geometry and BE mesh are shown in Fig.10.

The permeate flux across the membrane for each case is calculated through Eq.(16), where the ratio (K/μ) is taken from Table 3 and Q'' is calculated as outlined in previous section. Fig.11 shows the Flux(per-unit-length) vs. Pressure curves corresponding to the three cases. Plotted as filled circles are also the experimental data obtained from the actual membrane module (with star-shaped channels). Even though the filtration area in the case of circular channels with $R=2 \text{ mm}$ is slightly smaller than that in the star-shaped channel (0.0528 m^2 vs. 0.06 m^2), the predicted flux is higher. This is explained by the smaller thickness of porous material

Fig. 8 Geometry and boundary element mesh of a seven-channel membrane with channels of circular cross-section, when the diameter of the channel is equal to the inscribed circle corresponding to the star-shaped membrane ($D=2\text{mm}$)

Fig. 9 Geometry and boundary element mesh of a seven-channel membrane with channels of circular cross-section, when the diameter of the channel is equal to the circumscribed circle corresponding to the star-shaped membrane ($D=4\text{mm}$)

between retentate and permeate (and therefore the lower total resistance to flow) in the case of the (larger) circular channels. Similarly, the module with circular channels of $R=2.27 \text{ mm}$ is predicted to yield higher permeate flux compared to the actual module (star-shaped channels), even though the filtration area is the same in both cases. This result is a direct consequence of the assumption that the membrane module is

Fig. 10 Geometry and boundary element mesh of a seven-channel membrane with channels of circular cross-section, when the total filtration area is equal to that of the star-shaped membrane ($D=4.54\text{mm}$)

Fig. 11 Predicted flux-per-unit-length vs retentate pressure curves corresponding to Figs. 8-10. Experimental data are also plotted as full circles

homogeneous. When the flow resistance is concentrated around the perimeter of the flow channels, as is for example the case when a cake forms during filtration, the effect of channel shape and size will be largely determined by the manner in which they modify the flow inside the membrane channels and only in a secondary manner by the thickness of ceramic material between permeate and retentate. Apparently for this reason the module manufacturer suggests that the star-shaped membrane offers better performance compared to smooth-shaped channels.

Conclusion

The problem of fluid transport inside ceramic porous, multi-channel microfiltration membranes has been studied experimentally and computationally. Unlike previous studies, in which permeate flow inside the membrane material is neglected, we investigate this particular process based on Darcy's law for flow in porous media. A model for permeate flow in a seven (star-shaped) channel tubular membrane, based on the

Boundary Integral Method, is presented and validated. Based on this model, we propose a procedure for determining the effective permeability (K_m) of the membrane module by comparing computational results with on-line experimental measurements for the permeate flux at various levels of trans-membrane pressure. Besides determining K_m , this approach can be used to identify alternative designs and operating conditions in membrane modules of arbitrary geometrical complexity.

Nomenclature

c_i	= coefficient in the boundary integral equation
J	= permeate flowrate per unit length of the membrane
K, K_m	= permeability
P	= pressure
P^*	= fundamental solution (Eq.6.a)
Q^*	= normalised permeate flux (Eq.13)
R	= resistance of porous medium ($R=1/K$)
u, v	= permeate velocities in the (x) and (y) directions
x, y	= cartesian coordinates
Δ_i	= dirac delta function
$\Delta P_{tm}, \text{TMP}$	= transmembrane pressure
μ	= viscosity

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