DEVELOPMENT AND VALIDATION OF A MULTI-CONFIGURABLE MBR FOULING MODEL

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ABSTRACT

Membrane bioreactors (MBRs) are a recent innovation in wastewater treatment that combines a membrane filtration unit with a biological process (Tchobanoglous et al. 2003).The focus of this study is only on membrane filtration. It concentrates on the development of a mathematical model describing filtration and fouling on micro-filtration membranes and this work is based on the earlier published model by Liang et al. (2006). Initial modelling experiments with the Liang model showed its deficiencies at predicting trans-membrane pressures over a wide range of membrane fluxes. A very basic structure of this model and its behavioural character limit its applicability to only limited number of operational regimes. In an endeavour to develop a more universal yet simple model, the Liang model has been extended and modified to include: backwash mechanism, cake and SMP deposit compressibility effects, various cake removal models (air scouring and cross-flow velocity) and flux dependent SMP deposition rates. The model was developed in MATLAB/Simulink environment and validated on experimental data from a flux stepping experiment performed on a pilot scale with a tubular crossflow 0.45μm membrane. A second validation exercise was performed on long-term filtration data from a membrane pilot plant equipped with a hollow fibre 0.05 mm membrane. The model proved to be in good agreement with the measurements in both validation studies.

KEY WORDS

Cake, Filtration, Fouling, MBR, Modelling, Simulation, SMP, Wastewater

INTRODUCTION

Background

The first reported application of MBR technology in wastewater treatment was in 1969, when an ultrafiltration membrane was used to separate activated sludge from the final effluent of a biological wastewater treatment system and the sludge was recycled back into the aeration tank (Smith et al. 1969). Since then the MBR system has evolved greatly and now many full scale industrial plants of many different designs, membrane types and operational regimes are being successfully installed on industrial and communal wastewater treatment plants. Membrane bioreactors (MBR) for wastewater treatment have been receiving a lot of attention lately due to their advantages over other treatment processes like conventional Activated Sludge Process (CASP). These advantages have been reviewed by Stephenson et al. (2000) and include: very high effluent quality, greater (independent) control over the Solids Retention Time (SRT) and Hydraulic Retention Time (HRT), smaller footprint due to elimination of large final settlement tanks (FST), reduction of reactor volumes due to increased Mixed Liquor Suspended Solids (MLSS) and reduced sludge production due to maintaining higher than usual SRTs in the system. The main deficiencies of MBRs are higher operational expenses (OPEX) due to higher energy consumption, which is used for pumping, membrane back-washing and preventing cake build-up (air scouring, high cross-flow velocity) and chemical usage for regenerating the fouled membranes. The current research on MBRs is focussed at reducing these costs and therefore making membrane systems even more competitive with other wastewater treatment solutions. These research areas include: development of new membrane materials less susceptible to fouling, creation of chemicals and cleaning procedures for cheaper and most effective recovery of irreversible fouling, devising new and more efficient MBR system designs and the development of new more efficient MBR operational procedures.

Our research focuses on the last two areas, where design and control strategy optimisation can be done using accurate, validated mathematical models describing membrane filtration and membrane fouling.

Membrane fouling mechanisms

Membrane fouling in MBRs is attributed to the physicochemical interactions between the biofluid and the membrane (Chang et al. 2002). Fouling is often subdivided into two subcategories: *reversible* fouling and *irreversible* fouling. Reversible fouling is caused by the deposition of suspended solids and formation of cake on the membrane surface. This cake layer can be removed from the membrane by backwashing and its formation can be controlled by operation at low fluxes, air sparging or provision of high cross-flow velocities. Irreversible fouling is caused by the adsorption of dissolved matter into the membrane pores (pore constriction and pore blocking). This type of fouling can be removed (often only partially) by chemical cleaning. Restricting categorisation of membrane fouling to reversible and irreversible is however somewhat simplistic. As discussed in Wisniewski et al. (1998), gel layer formation over a membrane surface is most often irreversible although it is notionally reversible since it forms a cake layer. At the same time, some kind of irreversible fouling may be partially reversible depending on the strength of adhesion and the vigour of the physical wash.

Factors affecting membrane fouling

Membrane fouling in MBRs is a very complicated process as it is induced by various types of foulants (solids, colloidal matter, and soluble substances) and caused by different mechanisms (cake formation, adsorption, scaling, etc.). Findings of the last several years of research show that fouling is influenced by the following factors: biomass characteristics (floc size distribution, (Wisniewski et al. 1998), floc structure, (Meng et al. 2007), EPS concentrations, SMP production), operating conditions (flux, (Choi et al. 2005), periodical air or permeate backwash sequence (Psoh et al. 2006, Mugnier et al. 2000), intermittent suction operation, air sparging intensity (Psoh et al. 2006), cross-flow velocity, (Choi et al. 2005, Tardieu et al. 1998), SRT, MLSS concentration, addition of PAC etc.) and membrane characteristics (pore size distribution, membrane type and material). For given membrane type and operating conditions, fouling is a function of SMP and EPS concentration and floc size distribution. The relationships between these three factors and degree of fouling however are still unknown. As a floc size distribution of a polydisperse activated sludge is very difficult to obtain, most of research has focussed on establishing correlations between fouling and EPS and SMP concentrations in the bulk liquid and the permeate. As reported in (Nielsen et al. 1999) the bound EPS (or simply EPS) is composed of sheaths, capsular polymers, condensed gel, loosely bound polymers and attached organic material. Soluble EPS (or SMP) is composed of soluble macromolecules, colloids and slimes. The bound and soluble EPS include bacterially produced polymers, lysis products and hydrolysis products, (Nuenghamnong et al. 2005). Soluble EPS or SMP are biodegradable and additionally can be a product of dissolution of bound EPS (Hsieh et al. 1994, Nielsen et al. 1997). It is generally accepted that SMP contributes to irreversible fouling while suspended solids and EPS cause reversible fouling. EPS changes some properties of activated sludge flocs and is reported to decrease the cake permeability, (Nuenghamnong et al. 2005, Broeckmann et al. 2006).

Fouling modelling approaches

Capturing membrane fouling phenomena in the form of a mathematical model has been a task of many different research groups around the world for more than a decade. Over that time many approaches of various complexities have been adopted. Steady state mass transport models are used for an approximate MBR design and are usually based on empirical relationships. In this paper we will deal with dynamic models which are capable of predicting the dynamics, not just steady state behaviour of an MBR reactor. These models are based on different mathematical approaches and are of very different levels of detail starting from simple empirical models, behavioural models to fully mechanistic ones. So far the backbone of fouling models was a resistance in series model which is given in Equation 1. In this approach the total resistance of the membrane is equal to the sum of unit resistances such as pore constriction, cake, biofilm, polarisation layer, scaling, etc. Each model may have these separate phenomena modelled in a different way. Apart from modelling only a membrane resistance other aspects of MBR operation such as cake accumulation control with air sparging or cross-flow velocity and backwashing will also need to be included in the MBR fouling model. The following sections of this paper will summarise the recent advancements in MBR modelling.

Broeckmann et al. (2006) consider pore size and floc size distributions. Irreversible fouling is modelled as pore blocking where porosity of the membrane decreases over time. The resistance is represented with the Karman-Kozeny equation used for flow modelling in porous media. Cake formation calculated using mechanistic equations of forces acting on a spherical particle in contact with a flat membrane. The cake resistance is then calculated using the Blake-Kozeny equation. Busch et al. (2007) developed a model for hollow fibre MF/UF filtration units which takes into consideration the geometry of the system, the hydrodynamics of the feed and of the permeate flow and the filtration resistance. The filtration resistance considered membrane resistance, pore blocking, cake layer formation, polydispersed particles, biofilm formation and concentration polarisation on high levels of detail. Cake layer formation considers polydispersibility of flocs and irreversible fouling is based on the Karman-Kozeny equation. Ognier et al. (2004) introduced a concept of local critical flux. Under constant flux operation membrane with pores which are modelled as round openings of the same diameter are gradually closed due to the deposition of soluble organic substances (SMP). This progressive decrease of open pore number occurs as long as the flux through remaining open pores does not reach a so-called critical flux. At that moment the cake formation starts to occur. Ye et al. (2006) developed a model for predicting TMP evolution under long-term subcritical filtration of EPS solutions using combined standard pore blocking (gradual pore closure) and cake filtration model as well as combined complete pore blocking (particles block individual pores) and cake filtration model. Their model was based on standard pore blocking and was capable of predicting a sudden transition between a slow rate of TMP increase to a rapid rise of the resistance. The lower the subcritical flux applied, the longer the first fouling stage lasts. This behaviour has been experimentally observed by Li and Yuan, (2002). Ye et al. (2006) took into account the pore size distribution and modelled the membrane pores as capillary pipes to which they applied the Hagen-Poiseuille law for the calculation of pressure losses during filtration. The pore sizes gradually reduce as SMP is deposited inside the membrane. When the pore sizes reach a critical value, the fouling mechanisms changes from pore constriction to cake build-up and a sudden increase in TMP occurs. Ye et al. (2006) have also developed shown that the rate of SMP deposition is related to the local permeate flux and formulated a mathematical model of this relation. Ho and Zydney (2000) developed a combined fouling model accounting for both pore blockage and cake filtration which was based on classical fouling model equations: complete pore blockage equation, intermediate pore blockage equation and cake filtration equation. The model was validated and was in good agreement with flux decline data obtained from bovine serum albumin filtration experiments. The model was extended by Duclos-Orsello et al. (2006) who added an internal fouling mechanism which is modelled with a pore constriction equation (reduction of pore diameters) and a Hagen-Poiseuille law. In addition to the models based on either Hagen-Poiseuille law or Karman-Kozeny equation for water flow in porous media other, often completely different approaches have also been developed by some researchers. Hermanowicz (2004) proposed a model based on a percolation theory. Initially, the filtration or adsorption of small, colloidal or subcolloidal material slowly changes the porosity and the permeability of the membrane and cake. Then at a critical porosity, further porosity loss provides a dramatic decrease of permeability. This model shows the two stage TMP profile, but predicts that TMP increases to ‘infinity’ in minutes in the second stage which is more rapid than observed in practice. Seminaro et al. (2002) took a stochastic approach to modelling of fouling in microfiltration membranes. The membrane pore space was represented by a bundle of nonintersecting tubes. Pore segments in the model membranes had circular cross-sections, random locations and sizes distributed according to distributions determined with Scanning Electron Microscopy. Permeability reduction in time during cross-flow microfiltration of a well-characterised particle suspension on a well-defined model membrane was calculated using a Monte-Carlo technique. The dominant mechanisms of membrane fouling were found to be particle capture and size exclusion at pore segments. In the simulations a matching size criterion between pore size and particle size was used to define pore blocking. Simulation results accorded with those obtained experimentally. Meng et al. (2005) developed a permeation model based on fractal theory and Darcy's law to evaluate the permeability of cake formed in microfiltration of activated sludge. As the microstructure of a cake layer is usually disordered and complicated and therefore cannot be describe by traditional geometry, the fractal theory was applied there to characterise the irregular objects in terms of its average, self-similar properties. This fractal permeation model provided a method for determining the permeability of cake build up on a membrane surface but will need more validation studies in order to determine it's applicability in real-life problems.

The fouling models discussed above are all called "lumped models" which means that they do not represent spatial variability of fouling and filtration conditions on the membrane. For accurate modelling of fouling where spatial differences and geometry is taken into account, so called distributed models such as in Computational Fluid Dynamics (CFD) need to be used.

MODEL FORMULATION & DEVELOPMENT

This study was done as a part of a DTI funded project number TP/3/DSM/6/I/15123 entitled Improving the *Design and Efficiency of MBR Plant by Using Modelling and Simulation* which intends to develop a complete integrated biological (activated sludge) and filtration model of an MBR reactor. For the simulation of the filtration module we have initially chosen the Liang's model (Liang et al. 2006) as it had a simple mathematical structure but still it proved to be quite accurate in a validation study on real activated sludge filtration unit. In our calibration study, we have found that the model was loosing its validity when a wider range of permeate fluxes were applied and therefore the model needed to be modified. The structure of the Liang's model, our proposed modifications and the results of the calibration and validation studies are explained in the next paragraphs.

Liang's model equations

The Liang's model calculates trans membrane pressure (TMP) for a given flux using a resistance in series approach as described by the Darcy equation:

$$J = \frac{\Delta P}{\mu \cdot \sum R_i} = \frac{\Delta P}{\mu \cdot R_t} \tag{1}$$

where

J	=	permeate flux, m/s
ΔP	=	transmembrane pressure (TMP), Pa
μ	=	dynamic viscosity of the permeate, Pa·s
R_t	=	total resistance, 1/m

Total resistance R_t is divided into three parts accordingly to the classic resistance in series model:

$$R_t = R_m + R_r + R_i \tag{2}$$

where

R_m	=	intrinsic membrane resistance, 1/m
R_r	=	reversible (cake) resistance, 1/m
R_i	=	irreversible resistance caused by SMP/EPS deposits, 1/m

The model represents two time-dependent resistances: resistance due to cake build-up (reversible fouling) and resistance due to SMP deposition on the membrane and inside the membrane pores (irreversible fouling). These resistances are proportional to the mass of cake and SMP deposited on the membrane and are given in the equations 3 and 4.

$$R_r = \alpha \cdot m_r \tag{3}$$

$$R_i = k_i \cdot m_i \tag{4}$$

where

m_r	=	mass of cake deposited on the membrane, kg/m^2
m_i	=	mass of SMP/EPS deposited on the membrane, kg/m^2
α	=	specific cake resistance, m/kg
ki	=	fouling strength factor, m/kg

Accumulation of m_r and m_i on the membrane is modelled with two ordinary different equations given below:

$$\frac{dm_r}{dt} = J \cdot X_T - k_r \cdot m_r \tag{5}$$

$$\frac{dm_i}{dt} = f_{SMP} \cdot J \cdot S_T \tag{6}$$

where

X_T	=	concentration of Mixed Liquor Suspended Solids in the feed, g/m^3
k_r	=	cake detachment rate, 1/s
S_T	=	concentration of Soluble Microbial Products (SMP) in the feed, g/m^3

The model assumes that all SMP in the feed ($f_{SMP} = 1$) is deposited inside the membrane pores or on top of the membrane and add to the mass m_i. The sludge cake is deposited on the membrane surface by the work of advection (mass flow of water through the membrane) but is also being detached by shear stresses caused by air scouring and cross-flow velocity. The rate of cake removal is proportional to the mass of cake on the membrane. k_r is a function of many different operational parameters such as: membrane type, membrane chamber geometry, air sparging intensity, cross-flow velocity, properties of the cake, etc. It is however modelled here as a single constant parameter which needs to be first calibrated. Further information about the Liang's model, its calibration and accuracy of predictions can be found in the original paper, (Liang et al. 2006).

Liang's model extensions

As part of the project, a computer representation of the Liang's model was created in the MATLAB/Simulink® environment. The following extensions were added to the original model equations to make the model usable for our simulation purposes.

Backwashing

In order apply the Liang's model to our measurements it needed some representation of backwashing. Due to the lack of information about backwash rates in our experimental data, the backwash model was limited to resetting initial conditions of each of the two differential equations. This means that after each backwash the mass of cake and deposited SMP was reset to some value which was then used as an initial condition for the next filtration cycle. The backwash equations are given below:

$$m_r^{i+1}(t_F = 0) = fr_r \cdot m_r^i(t_F = t_{end}) \tag{7}$$

$$m_i^{i+1}(t_F = 0) = fr_i \cdot m_i^i(t_F = t_{end}) \tag{8}$$

where

i	=	filtration cycle number, -
t_F	=	filtration cycle time, s
t_{end}	=	filtration cycle duration time, s
fr_f	=	fraction of m_r remaining after the backwash, (0-1)
fr_i	=	fraction of m_i remaining after the backwash, (0-1)

Mass of cake and mass of SMP deposits at the beginning of the next filtration cycle is equal to a fraction of the mass of cake and SMP accumulated by the end of the previous filtration cycle. Filtration and backwash cycles are controlled in the model by a binary backwash logic signal where 0 stands for a filtration cycle and 1 for a backwash cycle.

Pressure dependency

The original Liang's model assumed that the cake and SMP deposits are incompressible, while it has been reported (Psoh and Schewer, 2006) that biological slurries are highly compressible. We have therefore upgraded the model with compressibility equations for both the cake and the SMP. According to Flemming, (1995) as well as Lee and Wang, (2000) these are considered the most accurate equations for modelling cake compression and outperform several other simpler equations such as in Parameshwaran et al. (2001).

$$\alpha = \left(\frac{\Delta P}{\Delta P_{crit}^{\alpha}} + 1 \right)^{n_\alpha} \cdot \alpha_0 \tag{9}$$

$$k_i = \left(\frac{\Delta P}{\Delta P_{crit}^{k_i}} + 1 \right)^{n_{k_i}} \cdot k_{i0} \tag{10}$$

where

ΔP_{crit}^{α}	=	threshold pressure below which no cake compression occurs, Pa
ΔP_{crit}^{ki}	=	threshold pressure below which no compression of SMP deposits occurs, Pa
n_α	=	cake compressibility factor, -
n_{ki}	=	SMP deposit compressibility factor, -
α_0	=	specific cake resistance at atmospheric pressure, m/kg
k_{i0}	=	fouling strength factor at atmospheric pressure, m/kg

The cake resistance according to the equation 9 increases with TMP. For activated sludge the threshold pressure should be around 30 kPa, (Psoh and Schiewer, 2006). The cake compressibility has reported values of n = 0.8-1.5 and we have chosen n_α and n_{ki} of 1.0 for this work. The Liang's model with a backwash add-on and pressure dependency was calibrated to experimental data and the results of this calibration are presented in the section "Model Calibration and Validation". During model calibration the Liang's model was unable to predict the TMP in the low flux region and so it was upgraded with several add-on modules to improve the model's accuracy in the entire operating domain.

Flux dependent SMP deposition constant

The original Liang model assumes that all SMP is deposited on or inside the membrane which leads to null effluent SMP concentrations. The experimental observations however show that effluent SMP concentrations are comparable to the influent SMP concentrations. This presumes that only a small fraction of SMP is deposited during its passage through the membrane. Ye et al. (2006) also state that as the applied permeate flux decreases, the fraction of alginate deposited on the membrane pores gets smaller. The experiments they performed showed an exponential relation between the permeate flux and deposition rate which suggests a link between the deposition and the surface concentrate as defined by the film model. Following Ye et al. (2006) we have developed our own equation which calculates a fraction of SMP deposited on the membrane as a function of flux. The proposed exponential relationship is given in equation 11 below.

$$J < J_{\min} \rightarrow f_{SMP} = 0$$
$$J \geq J_{\min} \rightarrow f_{SMP} = f_{SMP,\max} \cdot \left(1 - \beta \cdot \exp - \left(J - J_{\min}\right)\right) \tag{11}$$

where

J_{min} = minimum flux below which no SMP deposition occurs, lmh
f_{SMP} = fraction of SMP deposited on the membrane pores, -
$f_{SMP,max}$ = maximum fraction of SMP deposited on the membrane pores, -
β = a constant determining how quickly f_{SMP} reaches $f_{SMP,max}$ with increasing flux, -

Detachment rate with critical cake thickness

Detachment of cake is most likely to be dependent on the cake thickness as thicker cakes are less bound to the membrane layer and therefore are easier to get rid of. The original Liang's model assumes that the detachment rate is proportional to the mass of cake deposited on the membrane. This entails close to zero cake detachment rates at lower fluxes where cake deposition is low. This leads to formation of thin cake layers at low fluxes. These then create slight TMP gradients in filtration cycles but which are not observed experimentally as will be shown in the Results and Discussion. Our proposed model assumes a threshold cake thickness d_{crit}. For cake layers of a thickness less than d_{crit}, detachment rate is constant and equal to k_{r1}. For thicker cake layers the detachment rate is a function of cake thickness as shown in the equations below:

$$d < d_{crit} \rightarrow k_r = k_{r1}$$
$$d \geq d_{crit} \rightarrow k_r = k_{r1} + k_{r2} \cdot \left(d - d_{crit}\right) \tag{12}$$

where

d = cake thickness, mm
d_{crit} = critical cake thickness, mm
k_{r1} = detachment parameter no. 1, $kg/(m^2 \cdot d)$
k_{r2} = detachment parameters no. 2, $kg/(m^3 \cdot s)$

In order to avoid negative cake thicknesses, the Simulink model was equipped with a saturation block with a zero lower limit. The cake thickness is calculated from the mass of cake according to the equation listed below.

$$d = f \cdot \frac{m_c}{\rho_c} \tag{13}$$

where

m_c = mass of cake per membrane area, kg/m^2
ρ_c = wet cake density, kg/m^3
f = ratio of the wet weight to the dry weight (SS) of the sludge cake

For activated sludge, the values of ρ_c = 1.06 x 10^3 kg/m^3 and f = 3.45 have been previously determined (Li and Yuan, 2002).

Shear stress model

Nagaoka et al. (1998) presented a mathematical relationship between the cake detachment rate and shear stresses caused by cross-flow velocity or air bubbles (τ_m). The detaching force due to τ_m is diminished by a pressure dependent static friction term which determines how strongly the cake adheres to the membrane. The expression for k_r is presented below.

$$k_r = \gamma_m \cdot \left(\tau_m - \lambda_m \cdot \Delta P\right) \tag{14}$$

where

k_r = cake detachment rate, 1/s

γ_m = constant, 1/Pa·s

τ_m = shear stress, Pa

λ_m = static friction coefficient, -

The value of τ_m can be correlated with cross-flow velocity or aeration intensity, so then a detachment rate parameter k_r can be directly linked to operational conditions in a MBR system.

Back transport phenomenon

Ho & Zydney, (2006) described a back transport equation which determines the rate of cake removal due to hydrodynamic forces (inertial lift and shear diffused diffusion) acting on the cake layer. Back transport rate (M_{det}) is equivalent to the term $k_r \cdot m_r$ in equation 5. The model is shown below:

$$\dot{M}_{det} = k \cdot \gamma^n \cdot MLSS \tag{15}$$

where

M_{det} = back transport rate, kg/m^2/s

k = proportionality constant

γ = shear rate (cross flow velocity)

n = constant, - ; n = 1 for shear-induced diffusion, n=2 for inertial lift

The term $k \cdot \gamma^n$ (steady-state flux) increases with increasing particle radius (a) with a dependence on a^3 for inertial lift and $a^{1.33}$ for shear-induced diffusion. Thus large cells and flocculated material tend to be kept away from the membrane with the steady-state flux dominated by the smaller colloidal matter, (Ho & Zydney, 2006). A number of investigators have developed empirical correlations for the steady-state flux in terms of the wastewater properties and operating conditions (Kraut and Staab, 1993, Shimizu et al. 2001), however the functional form and parameters are likely to be unique to the membrane, module design, wastewater, and biological condition of the activated sludge.

MODEL CALIBRATION AND VALIDATION

Experimental procedure

Our developed multi-configurable fouling model has been calibrated on the data obtained from a flux stepping experiment performed on the ITT Sanitaire's MBR pilot plant with a tubular crossflow 0.45µm membrane. In this flux stepping experiment the membrane was subjected to a range of fluxes from 30 lmh to 55 lmh as shown in Figure 1. This allowed for testing the irreversible and reversible fouling under different operating conditions both below and above the critical flux.

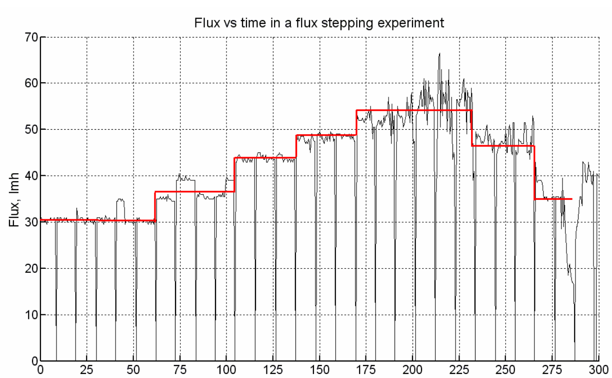

Figure 1 Telemetry data showing permeate flux vs. time in the flux stepping experiment

Parameters of different submodels constituting the fouling model were estimated using a combination of manual procedures and automatic optimisation based model fitting methods such as the Nelder-Mead downhill simplex method implemented in Matlab in the 'fminsearch' function. The objective function for the optimisation-based calibration was chosen to be a sum of absolute differences between the measurements and the model output.

Model calibration and simulations

In order to calibrate the unknown parameters in each and every submodel, the calibration procedure was split into 5 separate tasks in which different parts of the model were switched on and off to give a unique complete fouling model. In the course of the following 5 calibration procedures all required unknown parameters have been estimated. Then the calibrated models have been simulated and results of these simulations are shown in Figures 2-6. The following model configurations have been used:

Calibration 1	Basic model with backwashing and compressibility effects (A).
Calibration 2	(A) with critical cake thickness detachment rate.
Calibration 3	(A) with critical cake thickness detachment rate and flux dependent SMP deposition.
Calibration 4	(A) with shear stress model and flux dependent SMP deposition rate.
Calibration 5	(A) with back transport model and flux dependent SMP deposition rate.

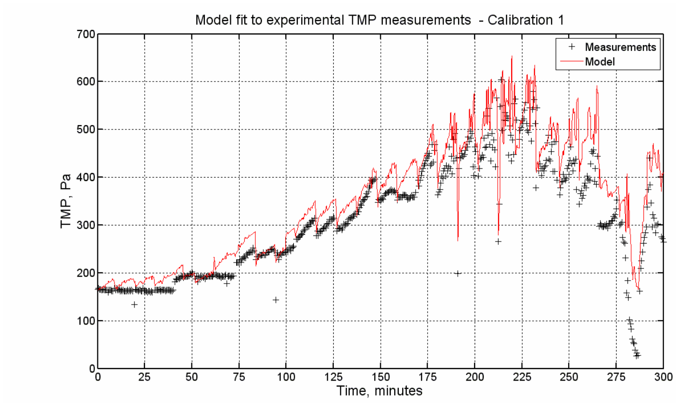

Figure 2 Model calibration with flux stepping experimental data – simulation option 1

The Liang's model with compressibility effects and a backwash module showed some inabilities to predict the reversible and irreversible fouling in a wider span of applied permeate fluxes. TMP measurements in the initial 50 minutes of operation (black) form a horizontal flat line whereas the model predictions (red) show a seesaw curve with a slight upward gradient (Figure 2). These slight TMP increases during the filtration cycle are caused by the model as it predicts cake deposition even at the sub critical fluxes whereas the experimental data show no cake accumulation in this region. This is caused by the cake removal term $k_r \cdot m_r$ in the equation (5) which tends to zero at low m_r values and this then creates an opportunity for thin layers of cake to build up on the membrane surface. The longer term TMP build-up which can be manually calculated as the difference in the TMP between the start of each consecutive filtration cycle after a full backwash and under a constant flux is due to SMP accumulation (irreversible fouling). The experimental data show no long-term TMP gradient in the first 35 minutes of operation where the flux was kept at a constant value of about 30 lmh. This leads to a conclusion that SMP deposition at low fluxes is minimal. The long term TMP gradient gets larger as the experiment progresses and as the applied flux gets higher. This may lead to the conclusion that SMP deposition rate is a function of the applied flux. The Liang's model assumes a constant SMP deposition rate regardless of the applied flux, so we must calibrate a single SMP deposition constant for an entire range of data. This is the reason why the model predicts a non-zero irreversible fouling at all even very small applied fluxes.

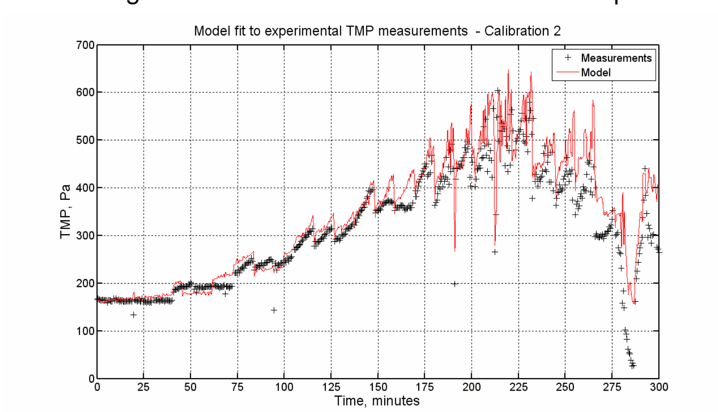

Figure 3 Model calibration with flux stepping experimental data – simulation option 2

In order to improve the model with regards to reversible fouling (cake formation) in low flux regions, the cake detachment rate was modelled with equations 12 and 13. Introduction of this new sub-model added two new unknown parameters but improved the model performance under low flux conditions as well as at higher applied fluxes, i.e. between the 75th and the 150th minute of operation (Figure 3). The long-term TMP gradient due to irreversible fouling is still present at low applied fluxes. This leads us to the simulation option 3 where a flux dependent SMP deposition model was applied.

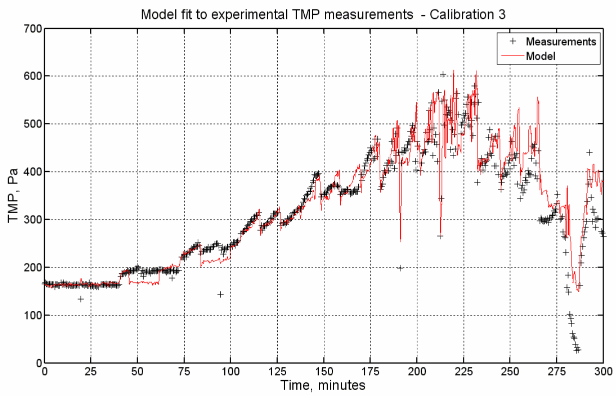

Figure 4 Model calibration with flux stepping experimental data – simulation option 3

Outputs from the Liang's model with thickness dependent cake removal and flux dependent SMP deposition are shown in Figure 4. The model is able to represent the long-term and short-term TMP gradients quite accurately in an entire range of applied fluxes and throughout the duration of the experiment. The model begins to diverge from the experimental data at higher applied fluxes where TMP rises above 450 Pa. This is due to the onset of cavitation forming in the suction pump, which created uneven pumping conditions (flux and pressure) and might contribute to erroneous pressure difference readings due to vapour bubbles present in the permeate. The model diverges from the measurements in the down-stepping part of the flux stepping experiment. The reason for that is unknown and would need to be further investigated.

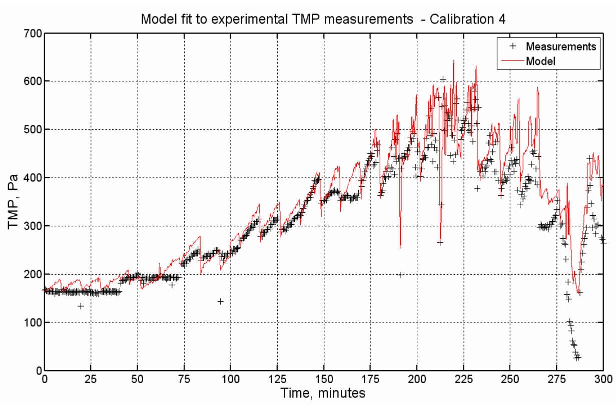

Figure 5 Model calibration with flux stepping experimental data – simulation option 4

Figure 5 presents the simulation results with the shear stress model and flux dependent SMP deposition. The shear stress model allows for relating the cake detachment coefficient to air sparging intensity and cross-flow velocity. There is however no relation to the cake thickness and therefore if a constant air sparging intensity and cross-flow velocity are assumed, the model will compute a constant detachment rate regardless of the cake thickness and applied flux. On top of this the detachment rate is a product of the detachment coefficient and mass of cake and leads to very small detachment rates at low fluxes. Thus the model suffers from a similar problem as the original Liang's model where cake develops even at very low subcritical fluxes.

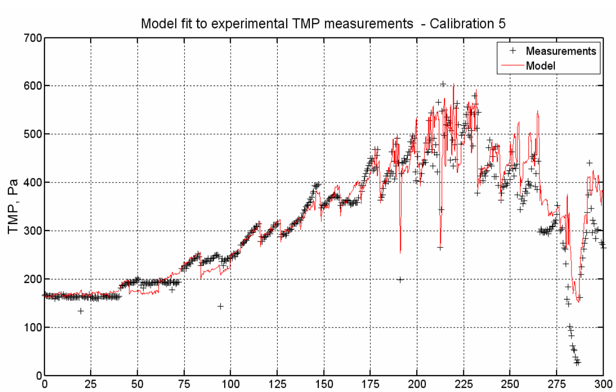

Figure 6 Model calibration with flux stepping experimental data – simulation option 5

An alternative cake detachment model called a back transport model (Ho & Zydney, 2006) which relates the detachment rate to shear stresses and solids concentration in the bulk liquid.

For this particular experiment the back transport model produced almost identical results to the cake detachment model with critical cake thickness.

All calibrated parameters are collated and presented in Table 1.

Parameter	Value	Unit	Description
R_m	1.68E+12	1/m	Clean membrane resistance
θ	0.005	--	Fraction of SMP deposited on the membrane
k_{i0}	1.1E+16	m/kg	SMP deposit resistance
α_0	4.0E+15	m/kg	cake resistance
ΔP_{crit}	30000	Pa	Critical TMP below which compression does not occur
k_r	150	1/d	cake removal rate constant
$d_{critical}$	0.005	mm	Cake thickness above which sloughing starts to occur
k_{r1}	0.012	$kg/m^2/d$	Cake detachment rate constant
k_{r2}	10.42	$kg/m^3/d$	Cake detachment rate constant
$flux_{min}$	30	lmh	Flux, below which no SMP deposition occurs
k_{SMP}	0.8	--	SMP deposition vs. flux dependency
γ_m	0.1	1(d·Pa)	Constant
τ_m	1000	Pa	Shear stress
λ_m	0.01	--	Static friction coefficient
γ_{flux}	0.002	1/s	Wall shear rate
n_{flux}	1.5	--	Exponent in back-transport model
k_{flux}	0.07	$m/(s^{1-n})$	Proportionality coefficient for back-transport

Table 1 Calibrated parameters of the multi-configurable fouling model

Long-term filtration experiment for model validation

After completing the calibration, the model was validated based on a long term filtration data from a pilot plant equipped with a submerged hollow-fibre PES (Sterlitech Polyethersulfone) membrane with 0.05mm average pore diameter. For this validation we have used a 162 hour long (6 days and 18 hours) subset of filtration data collected on a membrane filtration plant operating at a subcritical flux with 4 minute long filtration phases and 45 second backwashes. The model that was used for the validation was the Liang's model with flux dependent SMP deposition rate. Reversible and irreversible fouling processes in the Liang's model occur at very different time-scales. Reversible fouling can happen very quickly in a range of seconds to minutes and can be then reduced or completely eliminated after each backwash. In turn the irreversible fouling is a long-term process and it attributes to a slow but constant build-up of resistance in the membrane. The reason behind validating the model on long-term data is to test its flux dependent SMP submodel on long-term filtration data as the flux stepping experiment offered only a 5 hour set of data where the irreversible fouling had only a very limited contribution to the build-up of total membrane resistance.

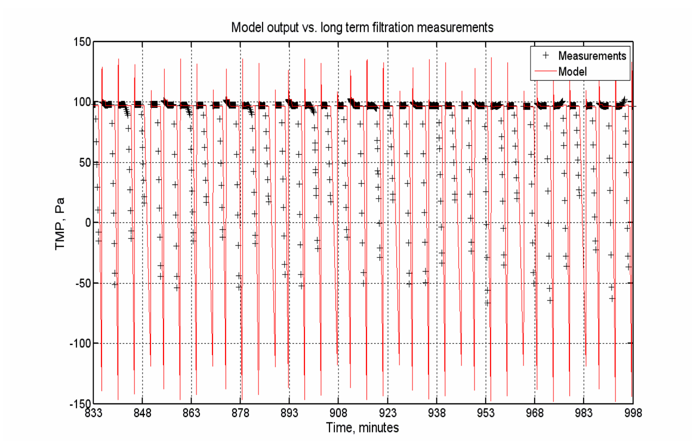

Figure 7 Long-term filtration experiment, 13hr:53min - 16hr:38min

The simulated filtration unit was operating under a very low subcritical flux and at very high cross-flow velocity, thus there was virtually no cake deposition on the membrane. As a result of this, TMP curves between each backwash were flat. The model performed very well at predicting trans-membrane pressure throughout this 162-hour-long simulation period. The quality of its predictions is shown on three separate figures below:

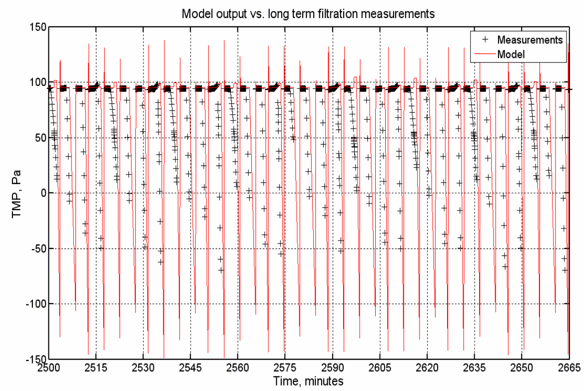

Figure 8 Long-term filtration experiment, 41hr:40min - 44hr:25min

The TMPs during filtration cycles are very well represented on all four graphs. However the model and experimental TMPs diverge in backwash cycles. This difference is due to a very simplified treatment of backwashing where the cake is removed instantly by resetting an initial condition in the Simulink integrator block and the pressure loss is calculated using the same specific resistance coefficient α as in the filtration phase. The model requires a more sophisticated backwashing module so that the TMP can be accurately predicted in both filtration and backwash cycles. The model parameters used in this modelling experiment were the same as for the flux stepping experiment.

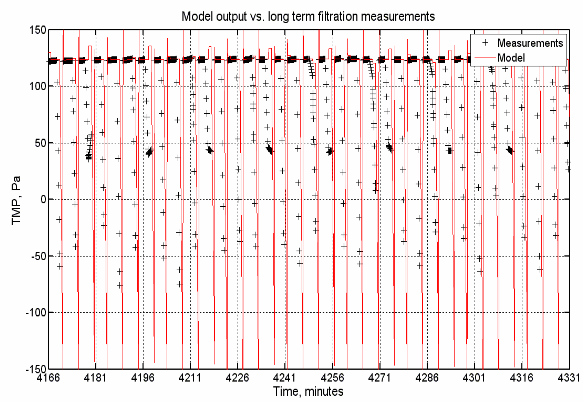

Figure 9 Long-term filtration experiment, 69hr:26min - 72hr:11min

DISCUSSION OF RESULTS

The Liang's model in its original form was able to pick up some general trends in the filtration process, but was lacking generality as was shown in the calibration and validation exercise. The model was therefore modified with an addition of several add-on modules and this improved its predictability in terms of both reversible and irreversible fouling. Major add-ons were: flux dependent SMP deposition, cake thickness dependent cake removal and Ho & Zydney's (2006) back transport model. Computed trans-membrane pressures from the modified model were in a good agreement with experimental data in both the flux stepping experiment and the long term subcritical filtration experiment. This provided a reasonable proof that the model is able to predict membrane fouling effects in at least some cases of microfiltration. The model however still lacks some generality as is shown in the next simulation experiment where the membrane was subjected to a constant subcritical flux filtration with pure SMP solution. According to Ye et al. (2006) at such filtration conditions a two stage TMP profile can be observed where an initial slow and gradual TMP rise is followed by a sudden transition to a rapid TMP rise. The long-term subcritical flux filtration simulation with the Liang's model showed no infliction point in the TMP curve (Figure 10 - solid lines) as it models the irreversible fouling with only a simple 1st order ordinary differential equation. Therefore the irreversible fouling equation in the Liang model (Equation 6) was replaced with a Hagen-Poiseuille equation for filtration through capillaries.

$$\Delta P = \frac{8 \cdot \mu \cdot L \cdot Q}{3.6 \cdot N \cdot \pi \cdot r^4} \cdot 10^{15} \qquad (16)$$

where

ΔP = trans-membrane pressure (TMP), Pa

L = membrane thickness, µm
Q = permeate flow, m^3/h
µ = permeate dynamic viscosity, Pa·s
N = total number of pores, -
r = pore radius, µm

In this model SMP is adsorbed inside the membrane and contributes to a gradual constriction of pores. As the filtration goes on, the pores gradually get smaller which in turn causes higher pressure losses accordingly to Equation 16. Simulation of the long-term subcritical filtration with this modification showed that the Hagen-Poiseuille equation is able to at least qualitatively generate two-stage TMP profiles at subcritical flux filtration as shown in Figure 10 (dashed lines).

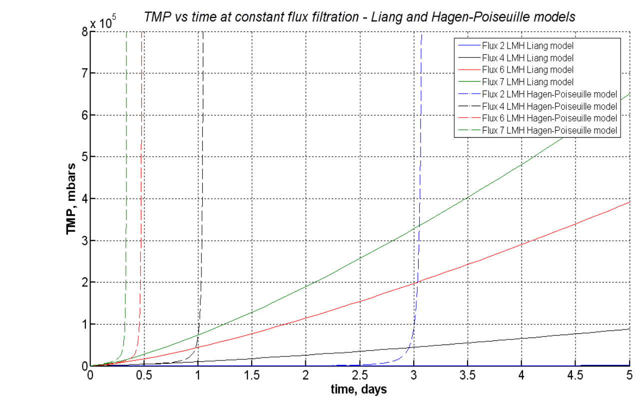

Figure 10 Predictions of TMP increase in time during a constant flux filtration – Liang's and Hagen-Poiseuille models

To summarise this experiment, by using the Liang's approach to modelling irreversible fouling and implementing our flux dependent SMP deposition constant, we were able to represent the actual TMP profiles in two real filtration experiments. The model however lacks generality as it is unable to generate the two stage TMP profiles which have been experimentally observed by various researchers. The model will therefore need to be modified in order to being able to represent this effect and for this purpose an approach similar to Equation 16 could be used.

CONCLUSIONS & FURTHER WORK

As we have seen our fouling model in the current form is not yet complete and will require some more work. Once all required add-ons are added it will be then combined with an activated sludge model and form an integrated MBR model. The proposed add-ons include:

1. Connection between bound EPS concentration in the mixed liquor and the specific resistance of sludge cake (α). Cho et al. (2004) have already proposed a sigmoidal relationship between these two variables.
2. Correlate cake detachment rate constant, k_r with cross-flow velocity and air flow rates in our pilot plant setup.
3. Investigate the model at predicting transmembrane pressures at large negative steps in permeate flux (as in the last stage of the flux stepping experiment).
4. Test and investigate the performance of the model with irreversible fouling modelled as SMP deposition inside the membrane pores and the pressure difference calculated from Hagen-Poiseuille law.
5. Model an increase of cake adhesion to membrane surface with irreversible fouling due to higher velocity fields around smaller (constricted) diameter pores.
6. Create a mechanistic backwash model which will be a function of backwash intensity, cake type, shear stress, or similar parameters affecting the rate of cake removal. One such model has been present in the work of Gehlert et al. (2005) where cake removal is modelled by equation 17 shown below:

$$\left.\frac{dm_c}{dt}\right|_{back} = J \cdot X_{T,back} - \gamma_{back} \cdot (\tau - \lambda \cdot TMP) \cdot m_c \tag{17}$$

Pressure loss during backwash is calculated in a similar was as for standard filtration but a different specific cake resistance α is used.

$$\alpha = \alpha_{back} \tag{18}$$

$$R_{c,back} = \alpha_{back} \cdot m_c \tag{19}$$

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