On the Purification of Waste Waters Using Multi-bore Filters: Simulation of a Long-term Filtration Stage

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Abstract. We present the progress of the simulation activity we are carrying out within the *PURIFAST LIFE+* project (started in January 2009). In particular, we first present the model we formulated to describe the macroscopic effects of the filtration process taking place in a multi-bore filter, focusing on the fouling phenomenon (namely the aging of the membrane due to the attachment of the pollutant species on its surface). In membrane-based filters the fouling phenomenon is the major reason of a decreasing filtration efficiency. Therefore, it is useful to simulate the long-term behavior of a filtration process in order to foresee the performance of the purification task.
The model was firstly implemented in COMSOL considering a single cycle of filtration (corresponding to few minutes) and a single cycle of back washing (few seconds). The latter is the step of inverted flux imposed in the plant in order to clean the membrane. Then we converted these two COMSOL models in MATLAB functions, so that we implemented a MATLAB program calling these functions in an iterative way: this procedure describes a filtration of some days (typically 4-9 days).
Our results are in good agreement with experiments and they are promising for the incoming application in the framework of the PURIFAST project.

Keywords: flow in porous media, waste water filtration, membrane fouling, driving COMSOL from MATLAB.

1. Introduction

The filters based on polymeric membranes are a technology widely used by industry and municipal companies (devoted to the control of the water quality) in order to obtain a purification of waste water which is efficient and quite cheap. Indeed, the cost of materials and the management of the plants is very competitive compared to other filtration methods.

In our context, we deal with a filtration module consisting in a cylindrical pressure vessel housing seven multi-bore polymeric fibers, each one hollowed by seven bores. The pore diameter of the polymeric membrane is 0.1 μm, which means that we are considering the so-called *ultrafiltration* process. The water to be filtered enters the module from the inlet (placed either at the top or at the bottom of the cylinder, depending on the operation mode) and it is channeled into the bores of the fibers. A pressure gradient is applied between the inner and the outer part of the fiber, so that the water flows through the membrane going out towards the external part. Such a procedure is known as *inside/out* filtration process. Consequently, all the pollutant particles in water larger than the pore diameter are cut of by the membrane and they remain inside the fiber capillaries. The clean water (the so--called *permeate*) is then collected and it flows away by the outlet, placed on the lateral surface of the module, quite close to the top. The filtration operates in a *dead-end* configuration, i.e. the polluted water (*feed)* entering the module can flow away just going through the membranes. In Fig. 1 a magnified picture of the fibers is reported, along with a scheme of the inside/out filtration. In Fig. 2 we show an example of a module.
The main problem in these filtering systems is the membrane *fouling*. Indeed, a part of the filtered particles can attach on the inner surface of the membrane, forming a thin layer (the so--called *cake*) which eventually soils the medium. Moreover, adsorption of some particle on the membrane structure (also clogging some pore) can occurs, reducing porosity and permeability of the medium. These phenomena reduce the filtration efficiency. While the initial porosity and permeability value cannot be never recovered, the cake may be partially removed. To do that, a periodic *back wash* process is imposed to the system, inverting the flux and let the clean water flow through the membrane, eventually remove part of the cake. A single

filtration stage takes 10-60 *min.*, while the backwash duration is in the range 20-60 *sec.*
In this paper we report the model we defined to describe the cycle of filtration/backwash (Section 2) and the use of COMSOL to simulate a single cycle (Section 3). Then, we show the MATLAB program we wrote in order to manage a long-term simulation (Section 4), made by several cycles and corresponding to an activity of 4-9 *days* of a filtration plant. The model results are then compared with the experimental data coming from the pilot plant set up within the project *PURIFAST* (Section 5).

(A) (B)

Figure 1. (A): a magnified picture of the fibers considered through the paper – courtesy of *Inge GmbH, Greifenberg, Germany*. (B) A scheme of the inside/out process taking place in the module.

Figure 2. An example of the module considered in the study– courtesy of *Inge GmbH, Greifenberg, Germany*.

2. Model description

In order to model the process from a macroscopic scale, we use the approach of multi-porosity, multi-permeability porous medium. In other words, the module and its content are idealized as the mixture of three continua:
(a) the shell region, in which the void space is made by the space outside the fibers.
(b) the membrane region, made by the sum of the solid matrix of the seven polymeric fibers and in which the void space is due to the membrane pores.
(c) the capillary region, in which the void space is the total space occupied by the lumina of the 49 capillaries.

Basically, each of the three region is a different porous medium, having a specified porosity and permeability. Such an approach may look quite artificial, but it allows to treat the complex interaction among the flow through the membrane, the lumina and the shell. The basic concept is the one used to represent the fractured media (and the flow through them), see [1], and it has been already applied to model hollow fiber filters, see [2] and [3].

Initially, the model was defined in a 3D setting. The simulations (made by COMSOL using a similar technique described later on, see Sec. 3) showed that the dependence of the solution upon the horizontal variable is negligible, except that very close to the outlet. Therefore, the equations were averaged w.r.t. the horizontal coordinates, getting a 1D version of the system. It is important to note that in this new version the boundary condition at the outlet turns into a sink term for the hydrodynamic problem in the shell region (see Eq. 3 later on).
In the sequel we describe directly the 1D model and the following notation will be used: subscript $()_c$ is referred to the capillary region; subscript $()_m$ to the membrane region; no subscript to the shell region. We denote by z the vertical coordinate, with $0 \le z \le L$, where L is the module length.
The model consists in a system of three equations describing the hydrodynamics in each region, coupled with a system of two equations accounting for the advection/reaction and adsorption process of the pollutant, taking place in the capillary region.

2.1 The hydrodynamic problem

In order to write the mass balance equation in the three regions, we recall that we have mass exchange from the capillaries to the membrane and from the membrane to the shell region.

Therefore, assuming a constant fluid density, we have

$$\frac{\partial}{\partial z} q_c = -\Gamma_c \tag{1}$$

$$\frac{\partial}{\partial} q_m = \Gamma_c - \Gamma \tag{2}$$

$$\frac{\partial}{\partial z} q = \Gamma - \frac{1}{\pi R^2} \frac{Q}{A_{out}} \chi(z)(2\pi R_{out}) \tag{3}$$

where:

- q is the superficial velocity (specific discharge).
- Γ_c is the source/sink term between capillaries and membrane
- Γ is the source/sink term between membrane and shell.
- The second term in the r.h.s. of equation (3) is the sink term accounting from the water flowing out from the outlet and it is the result of the average procedure applied to the outlet boundary condition of the 3D model. Indeed, A_{out}, R_{out} are the area and the radius of the outlet, respectively, and Q is the volumetric flux of water. Moreover, $\chi(z)$ is the characteristic function of the outlet position; namely, it vanishes everywhere except that for $(h - R_{out}) \leqslant z \leqslant (h + R_{out})$, being h the quote of the outlet center.

The porous regions are characterized as follows.

In the shell and capillary region, porosity and permeability are constant. Conversely, in the membrane they depend on the pollutant concentration adsorbed on the medium (see later on).

In each capillary and shell region the porosity is calculated as usual, see [4]

$$\varepsilon_c = 7N\left(\frac{r_i}{R}\right)^2, \tag{4}$$

$$\varepsilon = 1 - N\left(\frac{r_o}{R}\right)^2. \tag{5}$$

where N is the number of fibers (in our case, $N=7$), and r_i, r_o are the inner and outer radius of the fiber, respectively, and R is the module radius.

In the capillary region we have a longitudinal permeability, k_c, defined as

$$k_c = 7\frac{Nr_i^4}{8R^2}. \tag{6}$$

Porosity and permeability of the membrane, k_m and ε_m, respectively, depend on the pollutant concentration adsorbed on the membrane, see later on.

In the shell region the permeability is defined as (see [5]),

$$k = \frac{r_o^2}{4\phi}\left(-\log\phi - \frac{3}{2} + 2\phi - \frac{1}{2}\phi^2\right), \tag{7}$$

where $\phi = 1 - \varepsilon$.

Moreover, estimating the Reynolds number in each region we get $Re \leq O(10)$, so that we are allowed to use Darcy's law to link pressure and velocity field, which in our case reads as

$$q_c = -\left(\frac{k_c}{\mu}\right)\left(\frac{\partial P_c}{\partial z} - \rho g\right) \tag{8}$$

$$q_m = -\left(\frac{k_m}{\mu}\right)\left(\frac{\partial P_m}{\partial z} - \rho g\right) \tag{9}$$

$$q = -\left(\frac{k}{\mu}\right)\left(\frac{\partial P}{\partial z} - \rho g\right) \tag{10}$$

where P is the pressure, μ, ρ are the water viscosity and density, respectively (assumed constant) and g is the gravity acceleration.

2.2 Pollutant transport, adsorption and attachment

The pollutant transport takes place in the capillaries: therefore, the eq.s are coupled with the hydrodynamics of the capillaries region. We define:

c, the pollutant concentration in the volume of water flowing through the capillary region;

c_m, the concentration of matter attached on the inner part of the membrane capillaries;

c_p, the mass fraction of adsorbed pollutant;

and we write a standard transport equation, adding a term accounting for the attachment process and proportional to the rate of water flowing from the shell to the membrane region, namely Γ_c. Thus:

$$\varepsilon_c \frac{\partial c}{\partial t} + \frac{\partial}{\partial z}(cq_c) = \varepsilon_c D\left(\frac{\partial^2 c}{\partial z^2}\right) - \gamma\Gamma_c\left(\varepsilon_c c\right) - \rho_b \frac{\partial c_p}{\partial t} \tag{11}$$

$$\frac{\partial c_m}{\partial t} = +\gamma\Gamma\left(\varepsilon_s c\right) \tag{12}$$

where γ is the attachment coefficient, ρ_b is the bulk density of the membrane and *D* is the diffusion coefficient (assumed constant).
Concerning the adsorption process, we assume there is an equilibrium relationship between *c* and the mass fraction of adsorbed pollutant:

$$c_p = \frac{\bar{s} K_L c}{1 + K_L c} \tag{13}$$

which is the well-known *Langmuir* adsorption isotherm, see [4], (K_L and $\bar{s}$ are constant). Therefore,

$$\frac{\partial c_p}{\partial t} = \frac{\partial c_p}{\partial c}\frac{\partial c}{\partial t}. \tag{14}$$

2.3 Mass exchange and effect of attachment and adsorption

The sink/source terms are defined as follows

$$\Gamma_c = \alpha_c(c_m)\frac{k_m}{\mu l}\left(P_c - P_m\right), \tag{15}$$

$$\Gamma = \alpha \frac{k_m}{\mu l}\left(P_m - P\right), \tag{16}$$

where α and α_c are the filtering coefficient (dims. = 1/L), and *l* is the membrane thickness, i.e. the distance between two adjacent capillaries. We have:

$$\alpha = \frac{External\ surface\ of\ the\ membrane}{Volume\ of\ the\ shell},$$

while the filtering efficiency of the inner part decreases due to the pollutant soiling the membrane:

$$\alpha_c = \alpha_c\left(c_m\right) = A_v \frac{1}{1 + c_m / c_{ref}}$$

where A_V is the specific filtering area and c_{ref} is a reference value.
The adsorbed matter affects porosity and permeability of the membrane in the following way: the mass fraction c_p is proportional to the volume occupied into the pores. Assuming a capillary tubes structure,

$$c_p(z,t) = \eta\left(d_0^2 - d(x,t)\right), \tag{17}$$

where *d* is the pore diameter, with initial value d_0 , and η is a constant.
We assume a Poiseuille-type law (see [4]), so that:

$$\varepsilon_m = \frac{N_p \pi}{4A_{filt}} d^2 , \tag{18}$$

$$k_m = \varepsilon_m d^2 , \tag{19}$$

where N_p is the number of pores and A_{filt} is the filtering area.
Exploiting (17) in (18) and (19), we get the variation of porosity and permeability due to the adsorption.
Notice that the above argument is valid only in the stage of filtration, since the back wash has no effect on this process: thus, the value of ε_m and k_m before and after backwashing are the same (this is the concept of *irreversible fouling*).

2.4 The complete system and initial and boundary conditions

The system of PDEs summarizes as follows

$$-k_c \frac{\partial^2 P_c}{\partial z^2} = -\alpha_c(c_m)\frac{k_m(c_p)}{l}\left(P_c - P_m\right)$$

$$-\frac{\partial}{\partial z}\left(k_m \frac{\partial P_m}{\partial z}\right) = \frac{\alpha_c(c_m) k_m(c_p)}{l}\left(P_c - P_m\right) - \alpha \frac{k_m(c_p)}{l}\left(P_m - P\right)$$

$$-k\frac{\partial^2 P}{\partial z^2} = \alpha \frac{k_m(c_p)}{l}\left(P_m - P\right) - \frac{1}{\pi R^2}\frac{Qin}{A_{out}}\chi(z)(2\pi R_{out})$$

$$\left(\varepsilon_c + \rho_b \frac{\partial c_p}{\partial c}\frac{\partial c}{\partial t}\right) + \frac{\partial (c q_c)}{\partial z} = \varepsilon_c D \frac{\partial^2 c}{\partial z^2} - \gamma\left[\alpha_c(c_m)\frac{k_m(c_p)}{\mu l}\left(P_c - P_m\right)\right]\left(\varepsilon_c c\right)$$

$$\frac{\partial c_m}{\partial t} = \gamma\left[\alpha_c(c_m)\frac{k_m(c_p)}{\mu l}\left(P_c - P_m\right)\right]\left(\varepsilon_c c\right)$$

$$\alpha_c(c_m) = A_v \frac{1}{1 + c_m / c_{ref}},$$

The equations are endowed with *no flux* boundary conditions with the following exception: for the capillary region at the inlet we impose a inward flux and a prescribed concentration, i.e.

$$q_c = Q/A\ ;\ c = c_{inlet}$$

Moreover, what is known by the experimental apparatus is the measure of the pressure at inlet and outlet. Therefore, the initial datum for the hydrodynamic problem is a pressure in the hydrostatic equilibrium with the inlet (outlet) pressure for the capillary (shell) region. The initial pressure in the membrane region is taken as an average of the two. The initial concentrations are set to zero.

Finally, a similar model has been defined for the backwash stage, only varying the transport/attachment equation, for which in that case Γ_c drives a source term for *c*. Moreover, the boundary condition are changed according to the fact that the flux is inverted.

3. Use of COMSOL Multiphysics

The system of PDEs has been solved in COMSOL by separating the two stages of the process (i.e the filtration and the back wash step, respectively). For each stage we applied the following modes (see [6]):

- The *Darcy's law - Pressure* for a saturated medium (*Earth Science Module)*, to solve equations (1)--(3).
- The *Solute Transport* mode (*Earth Science Module*) for equation (11), where the model of *Langmuir* adsorption is a pre-built option in the *Subdomain setting*. The attachment term is included in the *source* option.
- The *Diffusion* mode for equation (12). In particular, we notice that this mode was applied with a vanishing diffusion coefficient: even if this approach seems far to the mathematical character of equation (actually the unknown c_m does not depend directly on the spatial coordinate *z*), it allows to couple this equation with the system of PDEs better than considering that as an evolution equation.

The terms Γ_c, Γ drive the coupling between hydrodynamics and transport process, and there were defined as *Global Expressions*, as well as ε_m, k_m and α_c .

Finally, we remark that (1)-(3) are stationary equations. Nevertheless, they are treated in a *transient* mode with a zero *storage term* (see [6] for more details): this fictitious numerical approach ensures a better coupling between the hydrodynamic problem and the transport--reaction one, which were solved all together using the *time dependent segregated* solver.

After the filtration stage, the solution was stored and the final evaluation of *c* and c_m were used as initial condition for the backwash step, which was solved in a similar way.

This first part of the simulation task was useful to calibrate the order of magnitude of the unknown parameters, such as $\gamma, c_{ref}, K_L, \bar{s}$. To provide that, the simulations was compared to experimental data concerning a single cycle of filtration. An example is reported in Fig. 3.

4. Modelling long-term filtration by coupling COMSOL and MATLAB

As pointed out in the Introduction, a real filtration process consists in several steps of filtration and backwash stages. Actually, we are referring to a pilot plant working in the following way:

1. Filtration 1. The waste water enters from the bottom inlet.
2. Back wash 1. The clean water enters from the outlet and exits from the bottom inlet.
3. Filtration 2. The waste water enters from the inlet at the top.
4. Back wash 2. The clean water enters from the outlet and exits from the inlet at the top.

Therefore, starting from the basic COMSOL model, defined using the COMSOL User Interface, we created a MATLAB script and four specialized MATLAB functions, each one corresponding to a different step. The script is the main part of the program, in which we saved the *fem* geometry, the *Constants* and the *Global Expressions.*

The input for the functions are the fem structure and the initial value for c, c_m, c_p. The output is the solution object *sol*.

From the main file, the four functions (in the right order) are called iteratively, launching at each time the COMSOL solver algorithm.

Finally, during the cycle and at the end of computation, the post-processing functions of COMSOL are used to concatenate the solution and evaluate it at inlet and outlet, which are the points of the real plant where the pressure is measured.

5. Results and conclusions

The complete version of the model has been calibrated, using experimental data.

In Fig. 4 we show an example of a comparison referred to a short period of filtration (1 hour) corresponding to the run of four cycles. The simulation time is in the order of 2 *min.*

We see that the simulation of the filtration steps is in good agreement with the data, except that for the first instants of each step: this is due to

the fact that in the real plant the initial pressure at every start-up is always the same guaranteeing a constant feed flux: it needs some seconds to recover the value compatible with the resistance of the membrane at that time. Conversely, the backwash is not fitted very well, but we have to notice that for any cycle we have only two data points for comparison: it means that the calibration for the backwash step cannot be carried out in a confident way.
Afterwards, the calibrated model was used to run long time simulations (4-9 *day*). The typical elapsed time for the simulation is about 2 hours. In Fig. 5 we show an example of the evolution of TMP (Trans Membrane Pressure) which basically is the difference between inlet and outlet pressure. The simulation is in good agreement with the data provided by the industrial partners of the project, from both the qualitative and the quantitative point of view. In particular, it is interesting to note that after some days (∼5) the pressure difference rapidly increases. This behaviour is due to the irreversible fouling: basically after some period the backwash is no longer sufficient to control the increasing resistance of the membrane, whose permeability is deeply reduced. Such a behaviour corresponds very well to what happens in the real plants.

7. References

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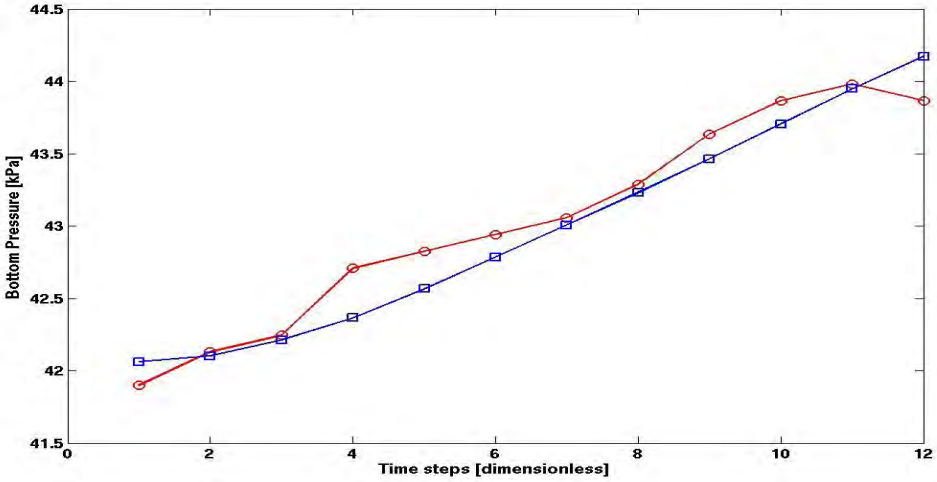

Figure 3. An example of comparison of simulation (blue) with experimental data (red) referred to the bottom pressure during a cycle of filtration (data provided by *Inge A.G.*)

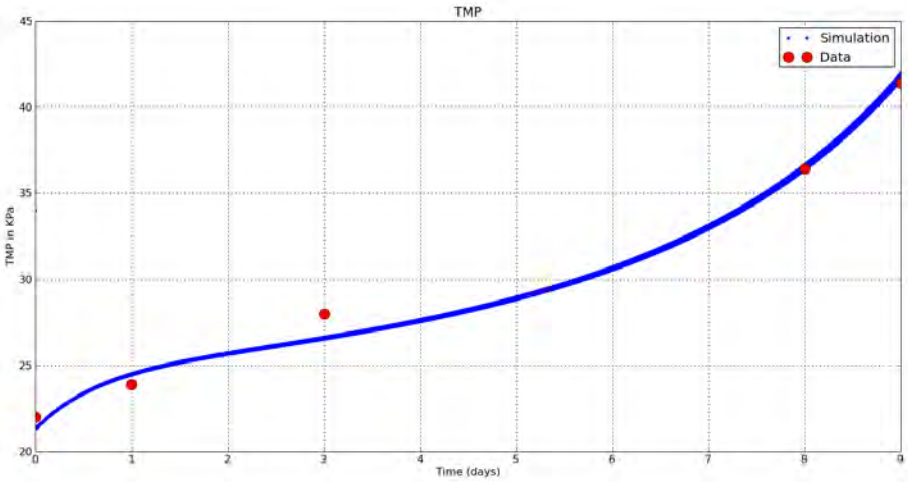

Figure 4. An example of comparison of simulation (blue) with experimental data (red) referred to the TMP during a 9-day process of filtration (data provided by *Inge A.G.*)