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On-line monitoring of cake layer structure during fouling on porous membranes by in situ electrical impedance analysis

S. Bannwarth [a,b], T. Trieu [a], C. Oberschelp [a,c], M. Wessling [a,b,*]

[a] Chemical Process Engineering, RWTH Aachen University, Turmstrasse 46, D-52064 Aachen, Germany
[b] DWI-Leibniz-Institute for Interactive Materials, Forckenbeckstrasse 50, D-52056 Aachen, Germany
[c] Institute of Environmental Engineering, ETH Zurich, John-von-Neumann-Weg 9, CH-8093 Zurich, Switzerland

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ABSTRACT

The structure of a fouling cake layer building up during membrane filtration can have a decisive influence on the performance of the membrane process. Hence a comprehensive description of the cake layers structure is mandatory. In this work we report a new methodology of simultaneously recording hydraulic resistance and electrical impedance during a membrane filtration experiment for flat sheet as well as hollow fiber membranes. An equivalent circuit model is derived to describe the development of the measured impedance during filtration. In situ e.g filter cake height and filter cake porosity as a function of time can be extracted from the experimental data for flat sheet as well as for hollow fiber membranes. The extracted data support traditional filtration theories for dead-end filtration on flat sheet membranes predicting an increase of cake layer porosity with proceeding filtration. However, the data obtained from inside-out filtration using a hollow fiber membrane reveal an apparent decrease of cake layer porosity during filtration. So the decreasing deposition area for the particles in inside-out filtration apparently results in a higher packing density of the particles and thus lower cake layer porosity. The electrical impedance data give an additional correlation allowing us to characterize the fouling cake layer structure or composition.

1. Introduction

Fouling on membranes poses one major challenge during pressure driven filtration because of a resulting decrease in permeate flux and hence process efficiency. Various fouling mechanism models have been proposed to describe the flux decline behavior due to fouling. Comprehensive models exist in the literature for the four most common fouling mechanisms: cake filtration, complete blocking, intermediate blocking and standard blocking [1–4]. One of the most recognized models for cake filtration developed by Zydney and co-workers involves a combination of pore blockage and cake filtration [5,6] that was further developed to also include pore constriction [7].

A series of methods is reported in the literature for the monitoring of fouling and the analysis of cake layer build up. The most common monitoring techniques are based on (a) direct observation through the membrane [8], (b) direct visualization above the membrane [9], (c) laser triangulometry [10], (d) optical laser sensor [11,12], (e) ultrasonic time-domain reflectometry [13–19], (f) NMR imaging [20,21] and (g) electrical impedance spectroscopy (EIS) [22]. Comparing all these methods in detail is beyond the scope of this paper. But it can be concluded that all these methods enable the analysis of the fouling cake layer thickness during built-up and porosity is usually subsequently derived from a cake layer's hydraulic resistance model. EIS additionally can also give information on the porosity of a material [23]. For a detailed comparison between the different monitoring methods the reader is referred to [24–26].

The fouling layer causes an additional hydraulic resistance on the membrane surface that is generally assumed to be in series with the membrane resistance in Darcy's law as presented in the following equation [27]:

$$J = \frac{1}{\eta_L\left(R_f + R_m\right)}\Delta p \tag{1}$$

wherein J is the area specific volume flux, Δp the transmembrane pressure, η_L the viscosity of the permeating liquid, R_f the hydraulic resistance caused by the fouling and R_m the hydraulic resistance of the membrane itself. If only cake layer build-up takes place, the additional hydraulic resistance is exclusively attributed to the hydraulic resistance R_c of the cake layer:

* Corresponding author at: Chemical Process Engineering, RWTH Aachen University, Turmstrasse 46, D-52064 Aachen, Germany
E-mail address: manuscripts.cvt@avt.rwth-aachen.de (M. Wessling).

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$$R_f = R_c = f(\delta_c, \epsilon_c) \quad (2)$$

This hydraulic resistance is highly dependent on the cake layer structure. Typically the parameters porosity ϵ_c and cake layer thickness δ_c are used to describe this structure. In this work, a novel way of obtaining thickness and porosity of a fouling cake layer during filtration is reported.

2. Background

2.1. Hydraulic resistance

Hydraulic resistances of porous media such as fouling cake layers are usually modelled by a Carman–Kozeny approach. For fouling on a planar surface, e.g. a flat sheet membrane, proportionality between the cake layer thickness and the increase in hydraulic resistance is assumed. This in turn requires the assumption that the cake layer porosity remains constant over the cake layer thickness as shown in the following equation [28,29]:

$$R_{c_{flat}} = \underbrace{\frac{K'(1-\epsilon_c)^2 S_s^2}{\epsilon_c^3}}_{\alpha'} \delta_c = \alpha' \cdot \delta_c \quad (3)$$

wherein $R_{c_{flat}}$ is the hydraulic resistance of the fouling layer on top of the flat-sheet membrane, α' the volume specific intrinsic hydraulic resistance given by Carman and Kozeny, ϵ_c the porosity, δ_c the cake layer thickness, S_s the specific surface of the foulant particles and K' the Kozeny-constant. The specific surface of the foulant S_s gives the ratio of surface area A_p of a foulant particle to its volume V_p. The specific surface is dependent on the particle size and it is decisive for the radius of the pores inside the filter cake. For spherical particles it is $S_s = \frac{A_p}{V_p} = \frac{\pi d^2}{\frac{1}{6}\pi d^3} = \frac{6}{d}$, wherein d is the diameter of the particle.

If the fouling takes place on a hollow fiber membrane, the radial geometry must be taken into account. In the case of inside-out filtration, this results in the following expression for the hydraulic resistance $R_{c_{rad}}$ in a radial geometry [30]:

$$R_{c_{rad}} = \alpha' \cdot r_0 \ln\left(\frac{r_0}{r_0 - \delta_c}\right), \quad (4)$$

and, respectively, for outside-in filtration in [29]:

$$R_{c_{rad}} = \alpha' \cdot r_0 \ln\left(\frac{r_0 + \delta_c}{r_0}\right), \quad (5)$$

with r_0 being the radius of the surface on which the fouling takes place.

Eqs. (3)–(5) include two unknown geometrical parameters to describe the cake structure: cake layer porosity and height. At this point it needs to be clarified that in these equations a homogeneous cake layer of equal thickness and porosity on the membrane is assumed. But especially in hollow fiber membranes these values can vary over the length of the membrane due to different axial hydrodynamic conditions [31]. By combining Eqs. (3)–(5) with Eq. (1) and measuring the liquid flow permeating through the membrane, one correlation to determine these two unknown structural parameters is available. More information is required to calculate both parameters.

Therefore a mass balance is used to gain a second correlation in order to obtain the two unknown parameters height and porosity. The mass balance can be simplified to a volumetric balance in the case of nearly constant densities. It correlates the total amount of filtered slurry V_{slur} with the total cake layer volume V_c by including cake layer porosity and volumetric concentration of foulant ϕ_{foul} in the feed slurry under the assumption of total foulant particle retention [32,33]:

$$V_c = \frac{V_{slur} \cdot \phi_{foul}}{(1 - \epsilon_c)}. \quad (6)$$

The slurry is divided into two parts during the separation process: the cake layer with its volume V_c and the purified permeate with the volume V_{perm}. Hence the slurry volume $V_{slur} = V_{perm} + V_c$ may be substituted in Eq. (6) providing a correlation of cake layer volume and the volume of purified permeate directly measured during dead-end filtration:

$$V_c = \frac{\phi_{foul}}{(1 - \epsilon_c - \phi_{foul})} V_{perm} \quad (7)$$

In the case of fouling on flat sheet membranes, the cake layer volume can be substituted using the cake layer height and the constant fouling area which is the membrane surface A_{mem}. This finally results in the required expression including the height and porosity of the cake layer:

$$\delta_c = \frac{V_c}{A_{mem}} = \frac{V_{perm}}{A_{mem}} \frac{\phi_{foul}}{(1 - \epsilon_c - \phi_{foul})} \quad (8)$$

In the case of fouling on hollow fiber membranes, the radial geometry must be considered when substituting the cake volume in Eq. (7). For inside-out filtration the cake layer volume is:

$$V_c = A_{mem}\left(\delta_c - \frac{\delta_c^2}{2r_0}\right) \quad (9)$$

wherein A_{mem} is the hollow fiber membrane outer surface area and r_0 the corresponding outer diameter, while for outside-in filtration it is:

$$V_c = A_{mem}\left(\delta_c + \frac{\delta_c^2}{2r_0}\right) \quad (10)$$

wherein A_{mem} is the inner surface area of the membrane and r_0 the corresponding inner diameter.

This approach of combining the mass balance with a hydraulic resistance model to obtain the two unknown parameters thickness and porosity is based on the assumption of a permeating liquid with a constant known concentration of foulant which completely deposits on the membrane surface. In many cases, the simplification of using the bulk concentration of the foulant in the feed slurry is made but for example effects like foulant sedimentation, back-diffusion or concentration polarization may cause deviations from this concentration. Particularly in cross-flow filtration mode it is challenging to determine the filter cake volume by an appropriate mass balance. Furthermore, additional information on the foulant geometry is needed to assess the specific particle surface mandatory for the Carman–Kozeny approach. Thus an additional correlation linking the structural cake parameters with a different independent parameter would be desired as it allows one to determine a further unknown or assumed parameter such as the specific surface of the foulant or the actual concentration of the permeating liquid.

3. Development of an impedance model

The depositions on the membrane due to fouling do not only cause a hydraulic resistance but also an electric resistance that can be measured by electrical impedance spectroscopy (EIS).