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Designing asymmetric multilayered membrane filters with improved performance



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ABSTRACT

Asymmetric multilayered filters, comprising a series of membranes with varying pore sizes stacked on top of one another, allow filtration to be tailored in a variety of novel ways. We develop a network model that systematically captures the complex filtration behaviour in such multilayer filters. The model allows us to understand the response of the system when challenged with a particular feed composition, characterized through the particle size and adhesivity to the membrane. We show how the model enables comprehensive and time-efficient sweeps in parameter space to be conducted that determine the optimal multilayered filter configuration for a given filtration challenge, classified by the number of membrane layers, the change in pore size between each layer (filter taper angle), and the level of transpore interconnectivity between each layer. The model allows us to isolate and analyse the effect of each of the specific filter characteristics and identify the practical merits and disadvantages. In particular, we predict that the optimal arrangement for maximizing throughput through the filter is to have pore radius gradually decreasing with depth, and a slight level of pore interconnectivity, with the precise set-up a function of the particle size, adhesivity and number of filter layers. The results of the analysis are used to draw conclusions on the design of membrane filters for optimal filter performance.

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1. Introduction

Membrane filtration can be tailored in a variety of ways by using a multilayered structure, composed of an array of membranes with different pore sizes stacked on one another. Such filters can, for example, offer a simple way of sequentially separating cells or particles [1,2], or combined to form the filtration support layers required in ultrafiltration, gas separation and catalysis [1]. In other cases, by using a membrane impregnated with bacteriadestroying medication in parallel with another that sieves particles, a greater spectrum of contaminants may be removed in one filtration process [3].

Filters whose porosity decreases with depth, or *porosity-graded asymmetric membranes* have been observed to improve efficiency [4–8]. Their increased efficiency can be qualitatively attributed to a decrease in porosity compensating for a reduction of contaminant concentration with depth, owing to prior filtering. This effect is particularly desirable, since often only a small portion of the filter media near the surface is actually involved in the active removal of

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http://dx.doi.org/10.1016/j.memsci.2016.02.028 0376-7388/© 2016 Published by Elsevier B.V. contaminants, with much of the deeper filter media left unused when the filter clogs. This leads to premature clogging and thus inefficient use of the filter [9].

Despite the significant merits of multilayer filters being wellknown in industry, a systematic study of the underlying mechanisms responsible for the superior performance that characterizes the internal behaviour of such multilayer filters during operation has yet to be explored. Currently, experimentally it is difficult to observe the internal particle trapping within a filter during the filtration process directly, with limited techniques only now beginning to emerge, such as positron annihilation spectroscopy [10,11]. Instead, deductions are made only after dissecting the porous medium once filtration has ceased. For these reasons, predicting and designing the optimal multilayer filter structure via a systematic series of experiments is impractical, and a theoretical study is highly desired.

Mathematical and computational methods allow for investigation of filtration challenges. Computational fluid dynamics (CFD) for modelling filtration scenarios are described in [12]. In the same paper, the authors use scanning electron microscopy to obtain a full description of a given membrane microstructure, and implement a full CFD model of particles moving within the

membrane. While a full CFD simulation provides excellent insight into how an individual particle is trapped, computational costs associated with keeping track of all the particles within a complicated pore structure makes it impractical on a large scale.

Dalwadi et al. [13] use homogenization theory to explore the improved filtration observed in a continuous porosity-graded filter. The model presented describes the motion of contaminants within a continuous media as they are transported via advection and diffusion. The results corroborate experimental observations, and are able to predict the performance of a filter through a reduced and computationally efficient mathematical model. The model enables large-scale parameter sweeps to explore filtration behaviour, and assist in the selection of porosity-graded filters for a given challenge feed solution. The model, however, is limited in that it does not include filter blocking mechanisms.

In our previous study [14], a network model was laid out that captures blocking in a filter composed of discrete pores through the adhesion of particles to the internal pore structures (standard blocking), the complete blocking of pores, and formation of a cake layer on the surface of the membrane [15]. In particular, an emphasis was placed on a systematic method of coupling the interplay between each of these fouling routes. The model is able to demonstrate how an understanding of this coupling was essential to explain a series of recent experimental observations. As a consequence, the model is able to predict the type of fouling behaviour that is occurring at a given time by simply studying measurements of the volumetric flux and total throughput through the membrane, without the need for dissecting the filter.

In this paper we analyse the efficiency of a multilayer filter, composed of a series of membranes with varying pore sizes. We derive a network model, which develops the model laid out in [14], to simulate the transport of particles through a multilayer filter and the trapping within. The model allows for adhesion of particles that are able to pass within the membrane to the walls of the pores, and for complete blocking of pores for which the particles are too large to enter. Pore interconnectivity is also included, allowing particles that are not trapped within one layer to pass into a choice of pores in the next filter layer. The network model provides a predictive tool for choosing the appropriate membrane to use, characterized by taper angle, affinity of the membrane material to the particles, number of filter layers and the pore connectivity, for a given feed solution.

2. Filter characterization

We consider a filter of depth \hat{h} composed of a series of N membranes of equal thickness (\hat{h}/N) stacked on top of one other. We label each membrane layer in succession, with layer k corresponding to the kth membrane that the challenge feed will encounter. We consider each membrane layer to be composed of a two-dimensional $m \times n$ array of regularly spaced uniform pores, of initial radii \hat{R}_k within the kth layer, as illustrated in Fig. 1. Here we study regimes in which the difference in pore radius between any two successive membranes is constant, but may reduce (a constricting filter) or increase (a dilating filter). We characterize this variation through the taper angle, α , defined by

$$\tan(\alpha) = \frac{\hat{R}_1 - \hat{R}_N}{\hat{h}}.$$
(1)

To enable comparison between different filters we consider setups in which the initial mean pore radius across the entire filter, $\langle \hat{R} \rangle$, is a constant, where



Fig. 1. (a) Schematic diagram of a multilayer filter. Each layer, k, is initially composed of an array of $n \times m$ equal-sized pores, but whose radius \hat{R}_k may vary for each layer. (b) Front schematic view illustrating the different pore radii at each layer and the taper angle. At each level the particle may pass into the pore directly beneath, or may traverse to one of four neighbouring pores. (Here two neighbouring pores are shown; the additional two pores are located into and out of the page.) The total filter thickness is \hat{h} .

$$\langle \hat{R} \rangle = \frac{1}{N} \sum_{k=1}^{N} \hat{R}_k.$$
 (2)

We also classify a membrane through the interconnectivity of pores between layers. Here we consider filters for which if a particle passes through a pore then it has the opportunity either to pass into the pore in the layer directly beneath or to traverse to one of the four neighbouring pores on the square grid (Fig. 1b). We denote the hydraulic conductivity of the connection to an adjacent pore by $\hat{\gamma}$: when $\hat{\gamma} = 0$ the particle passes directly to the next pore in the layer beneath and we call this a *non-connected filter*; when

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