



Filtration of protein colloids by fibrous membranes: A mechanistic investigation using packed bed filtration approach



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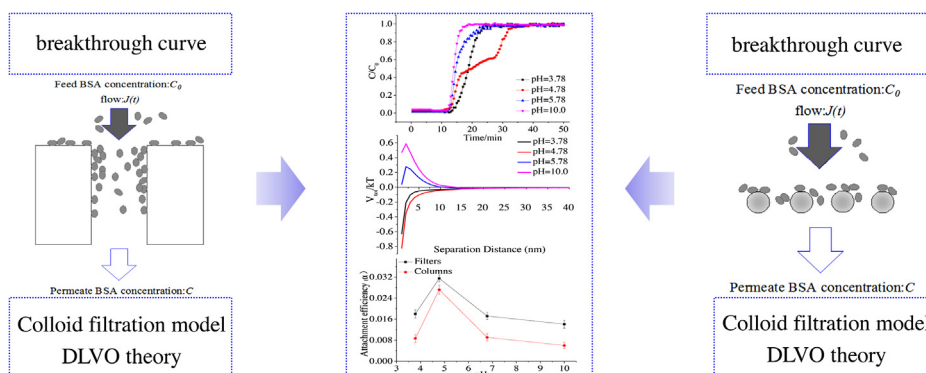
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HIGHLIGHTS

- BSA filtration by the fibrous membranes was simulated with column filtration.
- Microfiltration and column filtration yielded similar attachment efficiencies.
- Membrane fouling was the most severe at pH close to the isoelectric point of BSA.
- BSA–BSA interactions strongly affected its removal by microfiltration.

GRAPHICAL ABSTRACT



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ABSTRACT

This research aimed to use packed bed filtration as a novel approach to investigate fibrous membrane filtration of aqueous particles, a novel environmental application of fibrous materials and a new frontier for engineering colloidal science. For this purpose, bovine serum albumin (BSA) was filtered through a pre-characterized glass bead column to determine attachment efficiencies (α) of BSA onto glass beads. Subsequently, BSA was filtered through a glass-fiber membrane possessing similar surface elemental compositions as the glass beads. By simulating the glass fiber membrane as a packing of spherical glass particles with a diameter equivalent to fiber diameter, α values pertinent to BSA–glass fiber attachment were calculated using the clean bed efficiency model. A side-by-side comparison of the α values show that membrane filtration exhibited greater α values than column filtration, the α values calculated for membrane filtration were always about 1.5 times of those determined under similar conditions in column filtration. However, α values for the two systems varied with solution chemistry in similar trends and the maximal α values for both systems were both found at the isoelectric point of BSA ($\text{pH}_{\text{IEP}} = 4.78$). This finding implies that fibrous membrane filtration may be studied using classical packed bed filtration theories.

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1. Introduction

Recently, fibrous membranes have drawn widespread attention as novel materials for the separation of aqueous contaminants from water and wastewater, because of their superb energy efficiency, cost-effectiveness, and easy fabrication [1,2]. Like other depth-filtration membranes, particulate contaminants are removed by fibrous membranes due to their attachment/adsorption onto the fiber matrix, which also results in a significant reduction in the permeate flux and consequently increases the operating costs during long-term filtration [3]. The terms, attachment and adsorption, are used inter-changeably here for convenience.

Previous studies have revealed that membrane fouling during water treatment is attributed to the organic colloid (OC) fraction of natural organic matter or effluent organic matter, including humic substances and polysaccharide-like and protein-like substances [4]. Some researchers have reported that polysaccharide-like and protein-like substances are important contributors to membrane fouling [5–7]. In the case of protein fouling, many studies have focused on the illumination of the adsorption mechanisms and control strategies [8–10]. Protein adsorption onto membranes has been found to depend upon many inter-correlated factors, such as solution chemistry [11], operating conditions [12], pore morphology [13], surface physicochemical conditions [14], etc.

A large number of studies have used different analytical techniques to understand the mechanisms of colloid-membrane interactions and their impacts on membrane filtration. W. Richard Bowen et al. [15,16,18] used atomic force microscopy (AFM) in conjunction with the colloid probe technique to directly measure the repulsive electrostatic force experienced when a single colloidal particle approaches a microfiltration membrane. Scanning of such a membrane with the colloid probe provides a direct visualization of the membrane surface as would be experienced by a colloidal particle during filtration. Ding et al. [17] studied the protein-membrane and protein-protein interactions through the DLVO theory and the XDLVO approach. The measured results confirmed the fouling trends of a MF membrane and consistently showed that solution chemistries that induced higher fouling rates of MF membrane were associated with greater attractive and lower repulsive interaction energies among protein molecules and between protein molecules and clean MF membrane. Miao et al. [18] used a quartz crystal microbalance with dissipation monitoring combined with a PVDF-coated sensor crystal to investigate the effects of ionic strength on the deposition and adsorption behavior of BSA on the PVDF surface and on the structure of the BSA adsorption layers. The results showed that BSA passed more easily through the membrane and into the permeate. There was less accumulation of BSA on the membrane surface. A less rigid and more open-structured BSA layer was formed on the membrane surface.

Despite the aforementioned progress, colloid filtration by fibrous membranes is a fundamental process involved in membrane filtration of water and wastewater, with an emerging interest from the engineering society due to the potential use of novel, high-flux fibrous membranes. These fibrous membranes often possess open pore space with dimensions much greater than those of many aquatic colloids. As such, depth filtration should become an important mechanism for the removal of colloidal contaminants. Therefore, an in-depth understanding on the interactions between aquatic colloids and fibrous membrane materials is prerequisite for membrane design and material fabrication. However, it is currently difficult to directly apply any delicate instrumental techniques to the study of complex membrane filtration processes encountered in full-scale water or wastewater treatment. Most importantly, there is currently no viable approach to extract or separate aquatic protein-like colloids or other OC without destructing their native structures or status of agglomeration. This makes it par-

ticularly difficult for membrane scientist and engineers to simulate and ultimately, predict the filtration behaviors of OC during fibrous membrane filtration of water and wastewater. Consequently, we propose herein that packed-bed filtration using collectors with surface properties similar to those of fibrous membranes may be used as a viable approach to simulate the behaviors of protein, or generally, OC during membrane water treatment.

Previous works on packed-bed filtration have set a solid foundation for a translational study of fibrous membrane filtration by using packed-bed filtration technique. For example, Ryde et al. [19] studied the deposition and detachment of fine particles by the packed column technique. The results were presented in terms of breakthrough curves and interpreted by means of three phenomenological parameters, which are related to interaction forces between colloidal particles and the collector. This approach makes it possible to distinguish between monolayer and multilayer depositions. Elimelech et al. [20] investigated the deposition and detachment of particles in packed-bed filtration and quantitatively interpreted particle adhesion effects in terms of forces acting between two approaching surfaces.

Considering the complex nature of organic colloids in natural water or wastewater effluent, it is necessary to test this technique using well-characterized surrogate colloids or proteins and membrane materials. This serves as the primary goal of this study. The main objectives of this paper were three-fold: (1) to systematically investigate colloid breakthrough and the resulting pressure increase during fibrous membrane filtration and packed-bed filtration under similar conditions, (2) to quantify the related surface interactions by calculating corresponding attachment efficiencies for both modes of filtrations, and (3) to determine the possibility of using packed-bed filtration to simulate or study surface interactions involved in fibrous membrane filtration. For this purpose, pairwise membrane filtration (Fig. 1(a) and (b)) and packed-bed filtration (Fig. 1(c) and (d)) experiments were conducted by using glass fiber membranes and spherical glass beads with similar surface properties, at controlled solution pH and calcium concentrations. Classical colloid filtration model was used to quantitatively interpret the breakthrough results by calculating the attachment efficiencies of BSA on glass surfaces. Subsequently, the validity of packed-bed filtration for studying colloid-membrane interactions was determined.

2. Theory

2.1. Packed bed filtration model

Steady-state removal of BSA removal in a packed bed, assuming that dispersive transport and BSA desorption are negligible, can be described using the one-dimensional filtration equation [21].

$$\frac{C}{C_0} = \exp \left[-\frac{3}{2} \alpha \eta (1 - \varepsilon) \frac{L}{d_c} \right] \quad (1)$$

where C/C_0 is the fractional breakthrough, or the ratio of effluent BSA concentration (C) divided by the influent concentration (C_0). Since this is a steady-state equation, C/C_0 represents a balance between BSA adsorption and desorption. Also, L is the thickness of the filter bed, ε the porosity, α the attachment efficiency, η the collector efficiency, and d_c the collector diameter. For the glass beads used in this study, d_c is the average diameter of the spherical beads. In the case of the fibrous membrane, d_c is the average diameter of the fibers, assuming that each fiber is a string of spherical beads with diameter equal to the fiber diameter.

Physical factors that account for particle collisions with the porous matrix (collectors) are incorporated into the collector's attachment efficiency, η . The attachment efficiency is the ratio of the number of BSA that approaches a collector to the number that

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