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Influence of backwashing on the pore size of hollow fiber ultrafiltration membranes

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ABSTRACT

Backwashing is a common method for fouling mitigation. However, its impact on the pore-size distribution (PSD) of hollow fiber (HF) membranes has not been studied to date. This study quantitatively assessed the effects of filtration and backwashing cycles on the PSDs of polyacrylonitrile (PAN) and polyvinylidene difluoride (PVDF) HF membranes by evapoporometry (EP) characterization. The membranes were characterized before and after repeated cycles of filtration and backwashing in the absence of any foulants, and for a feed solution of bentonite and humic acid that caused fouling both on and within the membrane pores. Firstly, in the absence of any foulants, backwashing caused the appearance of larger pores, the effect of which was greater for the rubbery PVDF membrane than for the glassy PAN membrane. Secondly, backwashing was more effective in removing the fouling within the larger pores, but could not remove all the deposits within the smaller pores, which provides a mechanistic explanation for the progressive increase in the transmembrane pressure (TMP) with each backwashing cycle. Thirdly, the membranes that did not undergo the 10th backwashing at the end of 10 cycles of filtration and backwashing displayed a marked shift of the PSD towards smaller pores due to the deposition of foulants on and within the largest pores, whereas those that underwent the 10 complete backwashing cycles achieved nearly complete recovery of the larger pores accompanied by an irreversible increase in the diameter of the largest pores. Fourthly, a higher backwashing flux led to similar average pore diameters of the fouled and virgin membranes due to the increased effectiveness in restoring the smallest pores, but the corresponding higher filtration flux negated the benefits due to a greater fouling extent particularly for the larger pores. Finally, in order to achieve the desired permeation and rejection properties, possible enlargement of the pores needs to be taken into consideration when choosing a ultrafiltration (UF) membrane and when specifying the backwashing intensity and protocol.

1. Introduction

Backwashing is commonly used to remove fouling deposits in ultrafiltration (UF) and microfiltration (MF) [1–4] and is particularly convenient as a part of a dead-end filtration cycle. However, backwashing has been found to be less effective than chemical cleaning and sonication [5], although it is acknowledged to improve the permeate flux by decreasing internal pore fouling [6] or cake fouling [1,7] or both [8]. Unfortunately, frequent backwashing has been noted to promote the entry of macromolecules into the membrane pores [9], while lengthy backwashing durations and high backwashing fluxes have been observed to promote severe membrane fouling [7,10–12], possibly due to deposition of permeate-side contaminants [12]. Also, depending on the backwashing protocol [10,13,14] and nature of the feed or backwashing solvent [9,15,16], the effectiveness of backwashing was found to decrease as the applied transmembrane pressure (TMP) increased [17]. Clearly, backwashing influences the effective pore-size distribution (PSD) by removing fouling deposits within the pores and possibly by increasing the pore size due to the applied backpressure. Hence, the focus of this study was to characterize

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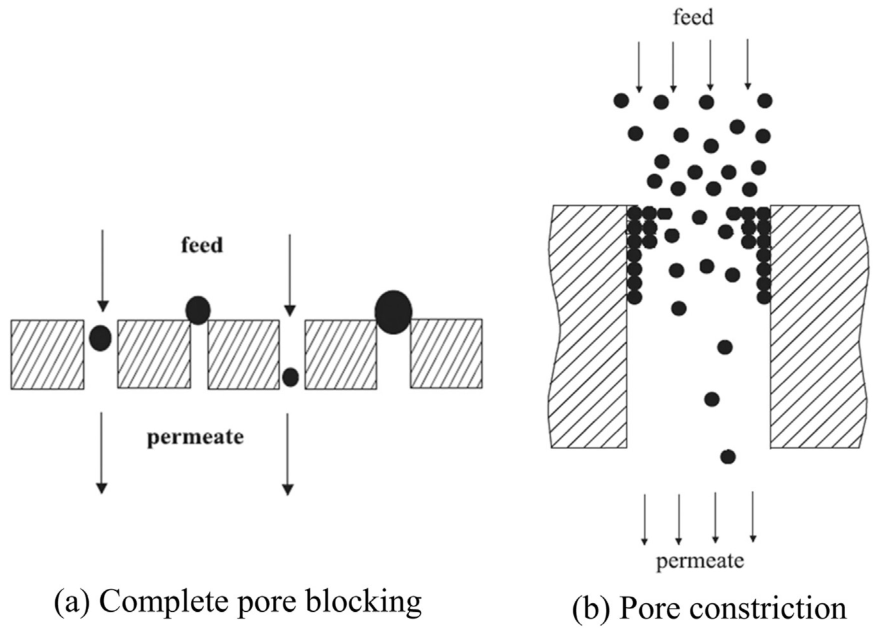

Fig. 1. Schematic of (a) complete pore blocking; and (b) pore constriction [18].

quantitatively the impact of backwashing on the PSD in order to develop an optimal backwashing protocol.

The PSD of a membrane is key to determining critical membrane performance metrics such as the permeability and selectivity. Membrane fouling is known to reduce the active pore sizes, thereby reducing the membrane permeability while possibly improving its selectivity depending on the membrane and foulant properties [17]. Conversely, although an increase in the membrane pore sizes increases the membrane permeability, the selectivity and rejection are reduced [18,19]. Fig. 1 shows a schematic of two of the proposed fouling models by Hermia [18] that have been used to better understand and assess the fouling mechanisms in membrane applications. Fig. 1 illustrates two of the mechanisms relevant to this study. Complete pore blocking, which occurs when particles larger than the membrane pores seal the membrane pore openings as shown in Fig. 1a, decreases the number of active pores per unit area of the membrane. Pore constriction, which occurs when particles smaller than the membrane pores deposit on the pore walls as shown in Fig. 1b, decreases the effective pore diameter, which in turn reduces the permeate flux but enhances the selectivity. For a fixed permeate flow, i.e., constant flux operation, both pore blocking and pore constriction change the flow distribution among the pores. When the number of active pores and effective diameter of the pores are reduced due to fouling, the active pores encounter a higher permeate flow due to increased local flux, which has been reported to significantly increase the fouling rate and cake layer density in dead-end filtration [18].

The effects of fouling and the efficacy of backwashing for mitigation are usually assessed via measurements of the change in the TMP. The TMP after backwashing and at the start of a new cycle may gradually rise due to residual fouling [20]. However, TMP measurements do not provide any information on what is happening on or within the pores due to the fouling or backwashing. This information could be obtained by observing changes in the PSD. However, this is non-trivial due to the limitations of conventional methods such as liquid-displacement porometry (LDP) in measuring the PSD of fouled membranes [21]. Characterization of the PSD of a fouled membrane by LDP is problematic since the deposits can be displaced by the flow through the pores that is required in LDP; that is, this measurement technique changes the nature of the sample. A related concern is possible interaction of the wetting liquid used in LDP characterization with the fouling deposits. Fortunately, the newly developed evapoporometry (EP) technique for characterizing the PSD [22–24] can be applied to fouled membranes [21,23]. EP is a non-destructive, accurate, and low cost technique that has a small laboratory footprint. Since EP uses a microbalance to obtain the gravimetric data required to determine the PSD, it can have higher accuracy than methods that require measuring quantities such as volumetric flow rates, partial pressures, or heat input. EP allows the use of different wetting liquids such as water that do not interfere with the foulant on the membrane or inside the pores. Compared to other techniques such as scanning electron microscopy (SEM), field-emission electron microscopy (FESEM) and atomic force microscopy (AFM), EP characterizes a relatively large membrane area that is more representative of the membrane.

The focus of this paper was to evaluate the effects of fouling and backwashing on external pore blockage as well as internal pore constriction (Fig. 1) through characterizing the PSD of UF membranes. The specific objectives of this study were the following: (1) to determine the effect of backwashing on the PSD of unfouled membranes (i.e., filtration and backwashing in the absence of foulants); (2) to assess the effect of repeated cycles of fouling and backwashing on the PSD of the membranes; (3) to relate changes in the TMP recovery to those in the PSD for repeated cycles of fouling and backwashing; (4) to assess the effects of backwashing on restoring the PSD of membranes subjected to repeated fouling and backwashing; and (5) to suggest a strategy for selecting UF membranes to accommodate the effects of backwashing.

2. Evapoporometry

A method for determining the PSD of both clean and fouled membranes is the recently developed EP technique [22–24] that has been applied to both flat sheet [22] and hollow fiber (HF) membranes [21,25]. EP is based on the Kelvin equation that relates the vapor pressure to the curvature of the interface between the liquid in the pores and the vapor given by the following [26]: