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Fouling behavior of microstructured hollow fibers in cross-flow filtrations: Critical flux determination and direct visual observation of particle deposition

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ABSTRACT

The fouling behavior of microstructured hollow fiber membranes was investigated in cross-flow filtrations of colloidal silica and yeast. In addition to the as-fabricated microstructured fibers, twisted fibers made by twisting the microstructured fibers around their own axes were tested and compared to round fibers. In silica filtrations, the three different fibers showed similar behavior and increasing Reynolds number increased the critical fluxes significantly. In yeast filtrations, the twisted fiber performed similar to the round fiber and better than the structured fiber. Among the three fibers, during yeast filtrations the critical flux for irreversibility was highest for the twisted fiber. The Reynolds number had little effect on the critical fluxes for particle deposition, which was attributed to the strong adsorption of yeast particles on the membrane. On the other hand, the critical fluxes for irreversibility increased with increasing Reynolds number for all three fibers. Direct visual observation of yeast particles on the surface of the three different hollow fibers revealed that for the structured and twisted fibers, the initial deposition rate on the fins is much lower than that in the grooves. This is attributed to the shear-induced migration of the yeast particles from areas of high shear (fins) to those of low shear (grooves). Furthermore, on the fins of the twisted fiber the deposition rate was lower than that on the fins of the structured fiber. This observation, together with the observed high critical fluxes for the twisted fiber led to the conclusion that the twisting induces a secondary flow in the liquid. This secondary flow is effective in depolarizing the buildup of micron-sized yeast particles since the diffusion of these particles is strongly effected by gradients in shear rate. On the other hand, for the silica colloids which are much smaller, shear-induced diffusion is not significant and twisting does not have an improving effect on filtration.

1. Introduction

The performance of pressure-driven membrane processes for liquid-phase separations is adversely effected by concentration polarization and subsequent membrane fouling. Membrane fouling is one of the most important problems in a membrane process as it increases operational costs and reduces membrane lifetime. There are two main approaches to mitigate membrane fouling. By suitable choice of the membrane material or modification of the membrane surface, the adsorption of foulants on the membrane can be minimized [1,2]. However, this approach by itself is not enough to prevent fouling. Even though a membrane may be repulsive to potential foulants, retained material will still accumulate near the membrane and form a concentration polarization layer which can initiate the formation of a cake or gel layer [3]. The extent of concentration polarization and cake/gel formation ultimately depends on the balance between attractive and repulsive surface interactions of the filtered material, which is determined by the material's properties (e.g. size, surface charge), solution properties (e.g. pH, ionic strength, concentration) and the hydrodynamics (e.g. permeation rate, pressure, cross-flow velocity, shear rate) [1,4]. The latter can be modified such that concentration polarization is alleviated and cake or gel formation is prevented.

By applying cross-flow over the membrane, part of the foulant that would otherwise build up on the membrane can be swept away. Laminar flow decreases concentration polarization and compared to dead-end operation it becomes possible to carry out filtrations for extended periods without performance loss. Turbulent flow is more effective in diminishing the concentration polarization layer and preventing fouling and therefore enables the use of higher permeate fluxes. However the energy consumption is much higher than for laminar flow. To make use of the mixing effect of turbulence in disrupting concentration polarization layers without bringing about a high energy input, several approaches have

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been suggested, such as the use of turbulence promoters [5], two-phase flow [6], pulsatile flow [7], corrugated membranes [8–12] and curvature-induced fluid instabilities [13–16].

A number of studies in literature have shown that introducing corrugations that lie normal to the feed flow direction on flat sheet membranes can promote turbulence and reduce concentration polarization significantly [8–12]. Apart from corrugated surfaces and turbulence promoting structures, turbulence can be passively created by curvature as well. Dean vortices, which are centrifugal instabilities formed in curved ducts, have been shown to depolarize foulant buildup during microfiltration and ultrafiltration in spiral, coiled, meander-shaped and helically twisted tubes or channels [13–16]. Broussous et al. fabricated ceramic tubular membranes with helical grooves on the inner surface and observed significant flux improvement compared to tubular membranes with smooth walls. They attributed this improved fouling performance to the flow disturbance by the helical structure [17,18].

In a previous study, we reported the fabrication of hollow fiber ultrafiltration membranes with microstructured outer surfaces. We showed that these membranes can be fabricated with the same intrinsic properties as round fibers fabricated under the same conditions. Therefore, they enable the same separation, and due to the microstructured surface they can offer enhanced productivity [19]. In this study, we investigate the fouling performance of these microstructured fibers in cross-flow filtrations. Additionally, by twisting the microstructured fibers around their axis we investigate the effect of having helical grooves that lie at an angle to the feed flow on the particle deposition. We use a flux-cycling method to determine the critical flux and direct visual observation to observe the deposition of yeast particles on the surface of the membranes (Fig. 1). Both straight and twisted microstructured fibers are compared to round fibers with the same intrinsic properties.

2. Experimental

2.1. Membranes and modules

The structured and round fibers used were made by the dry–wet phase inversion of the polymer dope 16.68% PES, 4.91% PVP K30, 4.91% PVP K90, 7.18% H_2O, 66.32% NMP, with water as the external coagulant. Details of the fabrication can be found elsewhere [19]. The structured fiber has 60% higher surface area per length compared to the round fiber (Fig. 2(a) and (b)). The pure water permeability of the fibers are 235 ± 11 L/h m^2 bar and 233 ± 12 L/h m^2 bar for structured and round fibers, respectively. The mean pore diameter of both fibers was found to be 12 nm by permporometry measurements. In addition to the structured fibers, twisted fibers were made by twisting each structured fiber around its own axis with about one full turn in 5 cm. The twisting was done after fabrication, when the fibers were dry. The pure water permeability and the pore size distribution remain unchanged after twisting. For flux-cycling experiments single-fiber modules of 40 cm length were prepared in 3 mm inner-diameter tubes. For direct visual observation (DVO), a flowcell made of three transparent PMMA plates was used. The plates were held together by a steel jacket and O-rings were placed between them to prevent leakage. The top plate was 2 mm thick, while the bottom and middle plates were 1 cm thick. The middle plate had an opening of 3 cm × 18 cm in the middle, and three 6-mm diameter holes on the two ends for potting the fibers. 6 mm outer-diameter tubes were glued into these holes in the middle plate to pot the fibers and extract the permeate. Three fibers were placed with equal spacing in the flowcell. A schematic of the middle plate of the flowcell is given in Fig. 2(c).

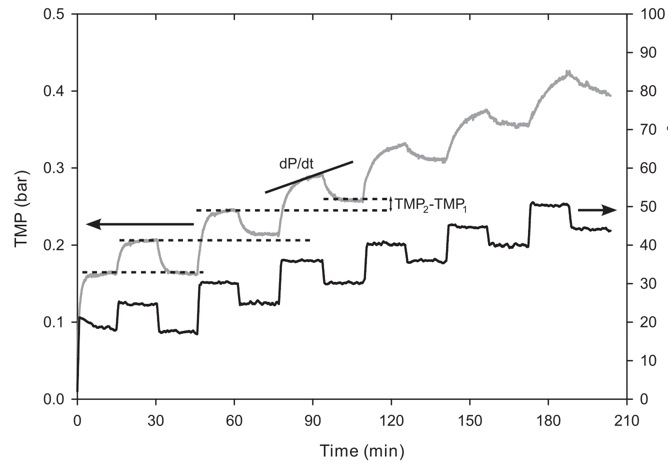

Fig. 1. Example of TMP-flux behavior and the calculation of the fouling rate and degree of irreversibility.

2.2. Flux-cycling experiments

0.25 wt% Ludox-TMA colloidal silica (Sigma–Aldrich) and 0.025 wt% Baker's yeast, *Saccharomyces cerevisiae*, (dry active yeast of Dr. Oetker) was used as feed suspensions. The as-purchased yeast was washed, filtered and dried overnight under air flow before preparing the feed suspensions. A fresh feed suspension was prepared for each experiment.

Before each experiment the pure water permeability of the membranes was measured as a check of membrane integrity.

To determine the critical flux, a flux-cycling method was used (Fig. 1) [20–22]. Flux steps of 5 L/h m^2 were applied, each step lasting 15 min. Two critical fluxes were defined, one with respect to the onset of particle deposition and the other one regarding the reversibility of this deposition [23]. To determine the critical flux for particle deposition, the evolution of TMP at constant flux during upward flux steps was monitored. The slope of the TMP during the last five minutes of the flux step, where the increase of TMP was linear, was calculated and plotted as dP/dt versus permeate flux. The flux where dP/dt becomes nonzero was taken as the critical flux for particle deposition. The degree of fouling reversibility was assessed by comparing the TMP's at the same flux on the way up and down (Fig. 1). For example, if the TMP on the way down is higher than that on the way up at 40 L/h m^2, then the critical flux for irreversibility was taken as 45 L/h m^2, as this is the flux step that caused irreversible deposition.

Each experiment was conducted twice for the determination of the critical flux. The first of these experiments was stopped after reaching the critical flux, while the second was continued further in order to determine the rate of particle deposition (dP/dt) and the degree of fouling reversibility (TMP_2–TMP_1) after the critical flux. The experiments were done under laminar conditions with Reynolds numbers of 120 and 400. $Re = 120$ corresponds to average cross flow velocities of 0.09 m/s for the structured and twisted fibers and 0.08 m/s for the round fiber. At $Re = 400$, the cross flow velocity was 0.31 m/s for the structured and twisted fibers and 0.27 m/s for the round fiber.

The flux-cycling experiments were carried out in the setup shown in Fig. 2(d). In this setup, the flowrates of feed and permeate as well as the pressures on the feed and permeate sides were logged every 5 s on a computer. The pressure on the retentate side was measured at the end of each experiment to calculate the TMP taking into account the pressure drop along the module length. When the yeast suspension was used as the feed, the suspension was stirred mildly during the course of the experiment to prevent