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Striping phenomenon during cross-flow microfiltration of oil-in-water emulsions

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ABSTRACT

The striping phenomenon in membrane filtration, whereby the foulants deposit as regular streaks rather than a more uniform layer, was first observed more than 30 years ago (Jonsson, 1987), but the understanding has remained limited to a few subsequent studies (Henriksen and Hassager, 1993; Larsen, 1991; Li et al., 2016; Tanudjaja et al., 2017; Tarabara et al., 2002). In view of the potential practical implications of the stripes in terms of membrane fouling and fouling mitigation, this study was targeted at an in-depth characterization of the stripes. The direct observation through the membrane (DOTM) technique was employed to observe the conditions when the stripes formed by oil emulsions stabilized by three Tween surfactants at a lower cross-flow velocity, and higher oil concentrations and permeate fluxes. The results indicate that (i) other than hydrodynamic factors (e.g., permeate drag, tangential shear), foulant-membrane and foulant-foulant interactions played a role in stripe formation or disappearance, as evident in the formation of stripes only by the oil emulsion stabilized by the Tween surfactants and the effect of pH; (ii) stripes made up of oil droplets appeared to be easier to remove than the more uniform layer of oil droplets, as evident in the shorter time taken for the former to detach; and (iii) among the three Tween surfactants, the striping characteristics investigated were largely similar, except for the time taken for the deposits to detach. These are expected to have implications for fouling control and mitigation.

1. Introduction

Because membrane fouling remains the Achilles' heel of membrane filtration, studies related to obtaining mechanistic understandings of the fouling behavior and correspondingly advancing means of fouling mitigation are widespread. Specifically for microfiltration, which is a pressure-driven membrane process in which suspended colloids and particles larger than approximately 0.1 μm are retained by microporous membranes, the deposition of such particulates/macrosolutes on the membrane is well-acknowledged to be a key limitation [7]. The deposition of foulants on the membrane hinges on a balance between the attractive drag towards the membrane (typically attributed predominantly to permeate drag and foulant-membrane affinity) and repulsive drag away from the membrane (typically attributed predominantly to hydrodynamic shear and foulant-membrane repulsion). A particularly interesting phenomenon of such fouling is that of the stripes, in which the foulants self-arrange into regular streaks of several microns wide that are parallel to the flow direction, reported in just a few membrane-filtration studies thus far [1–6].

The deposition of the foulants as stripes is particularly intriguing, because such particulate/macrosolute foulants are generally known to either deposit as a uniform layer or as clusters depending on the interaction energies at the foulant-membrane and foulant-foulant interfaces [8]. The stripes were first observed by Jonsson [1] during the study of boundary layer phenomena in ultrafiltration of macrosolutes, namely, dextran and whey protein. Subsequently, Larsen [3] observed striping by the blue dextran solution and proposed a stability model to describe the phenomenon. Larsen [3] found that the appearance of stripes was a slow process; in contrast, the removal of stripes, which can be achieved by increasing the cross-flow velocity (CFV) or relaxing the

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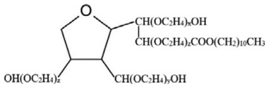
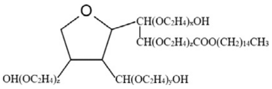
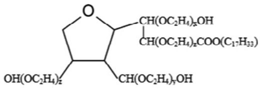
Table 1
Properties of Tween (Polyoxyethylene Sorbitan) 20, 40 and 80 (provided by Sigma-Aldrich).

	Tween 20	Tween 40	Tween 80
Structure			
Acid[a]	Lauric Acid ($C_{12}H_{24}O_2$)	Palmitic Acid ($C_{16}H_{32}O_2$)	Oleic Acid ($C_{18}H_{34}O_2$)
MW (g/gmol)	1228	1277	1310
ρ (g/mL)	1.095	1.08	1.064
CMC (mM)	0.06	0.027	0.012
HLB	16.7	15.6	15
γ (mN/m)	40.0	41.3	41.5

[a] The added acid during the ethoxylation of sorbitan.

permeate flux, was rapid. Stripes were also observed by Henriksen and Hassager [2] in the simulation of the macrosolute deposit layer on the membrane surface. Tarabara et al. [6] found via a two-dimensional on-lattice deposition model that stripes formed for cohesive particles under conditions of high Peclet number, and concluded that chemical heterogeneity of the suspensions can lead to fouling layers with different substructures. Another report on stripes was by Li et al. [4] in the study of colloidal fouling using silica and bentonite via Optical Coherence Tomography (OCT), which, consistent with Henriksen and Hassager [2], found the stripes on the fouled layer atop the membrane. Li et al. [4] further used the model developed by Larsen [3] to calculate the spacing between the stripes. In our recent study [5], we also observed the striping pattern formed by deposition of oil emulsions on the membrane via the Direct Observation Through the Membrane (DOTM) technique.

The stripes are reminiscent of the clusters prevalently observed in gas-solid fluidization [9,10] and also granular flows [11,12], although the patterns are more disordered than the parallel ones observed here, presumably due to the highly turbulent flows. This indicates that such stripping or clustering phenomena exist regardless of whether the operating regime is laminar or turbulent. Although the exact underlying mechanisms are not fully known, hypotheses have been reported based on the experimental observations. In gas-solid flows, clusters are understood to result from inelastic collisions leading to decreased kinetic energy and thereby lower local pressure regions that attracts surrounding particles [9]. In granular flows, Royer et al. [11] found that clustering was enhanced by increased inter-particle cohesion. For membrane-filtration processes, the underlying mechanisms leading to the stripes have been hypothesized to be the hydrodynamic instability caused by the flow along the channel [1] and perturbation in the mass transfer boundary layer, in which the variation of the local permeate flux due to the buildup of the deposit (boundary layer resistance) on the membrane surface led to a wave-like hydrodynamic profile [3]. Regarding the boundary layer, nonlinear instability has been reported to lead to stripes or streaks [13,14]. Another study further stated that the hydrodynamic instability in the flow channel was linked to the viscosity variation, and a critical thickness in the polarization layer and Reynolds number must be attained before the stripes appeared [15]. The stripes therefore appear due to a delicate balance of forces (mainly understood to be permeate drag and hydrodynamic shear), and any disturbance to the balance would result in the disappearance of the stripes [1,3,15]. Accordingly, seeking a more in-depth understanding of the striping phenomenon, particularly the characteristics of the stripes, underlies one of the goals of the current study.

The stripes are not merely an interesting research topic, but also have broader practical implications for the membrane-filtration process, particularly with regards to their deposition mechanism and removal from the membrane. Therefore, this study was targeted at systematically characterizing the stripes and evaluating its effect on membrane fouling. The DOTM technique was used to enable non-intrusive, real-time observation of the striping phenomenon by oil emulsion stabilized by surfactants. The specific objectives were to: (i) determine if stripes appeared for oil emulsions regardless of surfactant types, since only the Tween 20 surfactant was used in our previous study that observed stripes [5]; (ii) characterize the stripes in terms of the flux at which they appeared vis-à-vis critical flux; (iii) quantify the stripe parameters (namely, width, lateral velocity and number density); and (iv) understand the influence of pH and ease of detachment. These are expected to have implications for fouling control and mitigation.

2. Experimental methods

2.1. Oil-in-water emulsion

The oil-in-water emulsions were prepared by dispersing hexadecane ($C_{16}H_{34}$) (≥99% purity, Sigma Aldrich) in deionized (DI) water (Millipore) with either Tween 20, Tween 40 or Tween 80 surfactants (Sigma Aldrich). The properties (namely, structure, molecular weight (MW), density (ρ), critical micelle concentration (CMC) at 20–25 °C, hydrophile-lyophile balance (HLB) and surface tension (γ)) of the three surfactants are listed in Table 1. All the properties were provided by Sigma Aldrich; only the surface tension was measured with 100 ppm of surfactant at ambient conditions using a tensiometer (Dataphysics DCAT 11). 500 mL of oil emulsion stock was prepared for each surfactant (each containing 25,000 mg/L of hexadecane, each surfactant at a concentration of 100 times of the CMC value in DI water) using a blender (Warring 8011 S) at 18,000 rpm for 10 s. Before each experiment, a fresh feed solution was made by diluting the stock solution to a desired oil concentration (250 ppm, 500 ppm, or 750 ppm). The Focused Beam Reflectance Measurement system (FBRM; Lasentec S400, PI-14/206) was used to monitor the droplet size distributions of the oil emulsions in the feed throughout each experiment to ascertain that the distributions did not undergo significant changes. Fig. 1 presents droplet size distributions of the three oil emulsion stocks with 25,000 ppm of oil. Regardless of which Tween surfactant was used, the number-

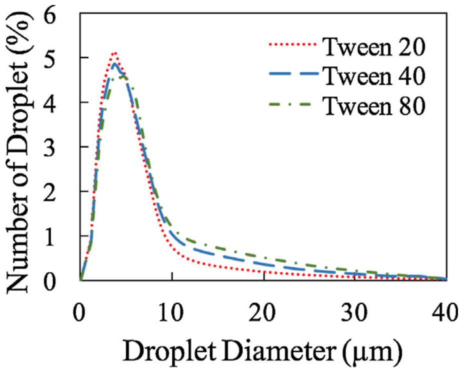


Fig. 1. Number-based droplet size distribution of 25,000 ppm of oil emulsion stabilized by each Tween surfactant.