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Analyzing external and internal membrane fouling by oil emulsions via 3D optical coherence tomography

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ABSTRACT

Membrane-based filtration is an emerging technique for processing oily wastewater. Improving the efficiency of oil-water separation via membranes entails novel techniques for studying the complex interactions between the oil droplets and membrane underlying fouling. This paper presents the first study that applies optical coherence tomography (OCT) to the characterization of membrane fouling by oil emulsions. A series of dead-end filtration experiments was performed to characterize the rejection of oil droplets (~ 10 µm; hexadecane) by the membrane structure (0.45 µm PVDF) via three-dimensional (3D) OCT scanning in real time. The experimental results were compared with the control experiments with ~ 10 µm glass beads to identify the optical artifacts. Both the external and internal fouling by the oil droplets were successfully revealed by analyzing the variation in OCT intensity at various layers that were mathematically defined in terms of the coordinate surfaces parallel to, and above and below, the feed-membrane interface. The evolution of membrane fouling was quantified by evaluating the fraction of fouling voxels as a function of time at varied depths. This study demonstrates that the OCT-based characterization has the potential to shed light on the complex interactions occurring in oil-water separations via membrane filtration, particularly by providing real-time non-invasive monitoring of internal fouling, the understanding of which is valuable for both fundamental research and practical applications.

1. Introduction

Oily water, a by-product from many industries such as petroleum refining, food and metal processing [1], can cause severe health and environmental impacts in the absence of adequate treatment. In comparison with conventional treatment methods such as flotation [2,3], coagulation [4] and biotreatment [5], membrane-based separation is an emerging technology in this field due to its ease of operation, no requirement for additional chemicals, high separation efficiency and relatively low cost [6–8]. However, the performance of such membrane-based filtrations is inevitably compromised by fouling – a complex phenomenon whereby deposition of retained compounds such as oil droplets onto the feed-membrane interface occurs, thereby reducing the throughput and selectivity of the separation [9]. Therefore, an in-depth understanding of the fouling mechanisms is crucially important to mitigate the negative effects resulting from the fouling.

The investigation of membrane fouling by oil emulsions necessitates characterization methods which can non-invasively monitor the filtration process in real time. Several techniques have been reported for a range of foulants, such as direct observation through the membrane (DOTM) [10–14], ultrasonic reflectometry [15], and electrical impedance spectroscopy (EIS) [16–20]. However, some limitations are associated with each of these methods. DOTM, which relies on an optical microscope to observe the feed-membrane interface, requires transparent membranes which are not as practical and is limited to two-dimensional (2D) imaging which means the loss of depth information. As for ultrasonic reflectometry and EIS, they detect fouling via acoustic waves and alternating currents, respectively, where the fouling is inferred from signal changes but direct high-resolution images are not possible.

Optical coherence tomography (OCT) has been used to characterize membrane fouling in recent studies [21–27]. Taking advantage of low coherence interference, OCT is able to obtain depth profiles of a semi-transparent sample by employing near-infrared light (wavelength from ~ 900 to ~ 1300 nm). The depth profiles can be synthesized to yield a three-dimensional (3D) image with a depth resolution of ~ 2 µm. Quantitative analysis of the growth of the fouling layer during membrane-based filtration has been achieved by analyzing a series of real-time OCT scans with image processing techniques based on background subtraction [21,23,28,29]. Despite the success in detecting the fouling

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layers formed by solid particles (e.g., bentonite and silica particles [21,23,24]) or soft matter (e.g., a biofilm [25–28,30–32]) using OCT, no study applying OCT to the characterization of the membrane fouling by oil emulsions is available to date. Compared with solid particles, oil droplets are unique in that they can coalesce and are deformable. Therefore, the inherent convolution effect of OCT imaging could prevent the detection of the initial deposition of oil droplets by background subtraction-based image analysis, especially when the oil droplets spread out on the feed-membrane interface (i.e., wetting) [33] or flow into the polymer network (i.e., internal fouling) [12,34]. Moreover, the visualization of a transparent particle depends on the discontinuity of the refractive index at the liquid-liquid interface, which could result in complex artifacts in the OCT images.

The current study was aimed at developing novel methods for quantitatively analyzing membrane fouling by oil emulsions via 3D OCT imaging. The proposed method was based on statistical analysis (e.g., averaging, standard deviation, 95% confidence interval) of the OCT data in a series of layers, above and below, parallel to the feed-membrane interface. Fouling experiments with hexadecane-in-water emulsions were compared with the control experiments with glass beads to validate the proposed method and assess the associated optical artifacts. It is shown that the mechanisms accounting for both the external and internal fouling by oil droplets can be revealed by statistically analyzing the variations in the OCT intensity as a function of time and space.

2. Experimental procedures

2.1. Membrane fouling experiments

In the current study, dead-end filtration was employed to validate the OCT-based characterization of membrane fouling by oil. The filtration setup is schematically shown in Fig. 1(a). The membrane cell consisted of two chambers separated by a flat-sheet hydrophilic PVDF microfiltration membrane (Merck, product No. HVLP04700) with a nominal pore size of 0.45 μm and effective filtration area of 962 mm^2. The feed entered the upper chamber (35 mm by 35 mm by 15 mm height) of the cell via four inlets located at the four side walls of the chamber and 14.5 mm above the feed-membrane interface. This configuration was to minimize the horizontal flows and the corresponding effects on the dead-end filtration process. The upper wall of the upper chamber had an optical window (35 mm by 35 mm quartz plate) for the OCT light to pass through. In the lower chamber (35 mm diameter by 15 mm height), a porous stainless steel plate (35 mm diameter by 2 mm height) was positioned flush with the membrane to provide mechanical support and stabilize the membrane during the filtration process.

The particulate foulants investigated were oil droplets and glass beads. The oil-in-water emulsion was prepared by blending (Waring, model No. 8010S) hexadecane (Merck, product No. 8.20633.1000) and DI water with a volume ratio of 1:2000 for 5 s at the speed of 18,000 rpm. The oil emulsion droplet size distribution (Fig. S1, Supporting information) was measured by the focused beam reflectance measurement (FBRM) technique (Lasentec, model No. PI-14/206), with the mean determined to be approximately 10 μm. The emulsions used as the filtration feed were prepared by dilution with DI water to 5 ppm by mass. Freshly prepared oil emulsions which were kept well-stirred throughout the experiment, were used for each experiment, and the distributions before and after each experiment were similar. Silica glass beads (Sigma Aldrich, product No. 440345–500G) with a particle size range of 9–13 μm and density of 1.1 g/mL were also used as foulants for comparison with oil droplets. Because the oil droplet size distribution (Fig. S1, Supporting information) indicated that the droplets in approximately the same size range of the glass beads was about 10% by mass of the total oil droplets, the feed concentration of glass beads was

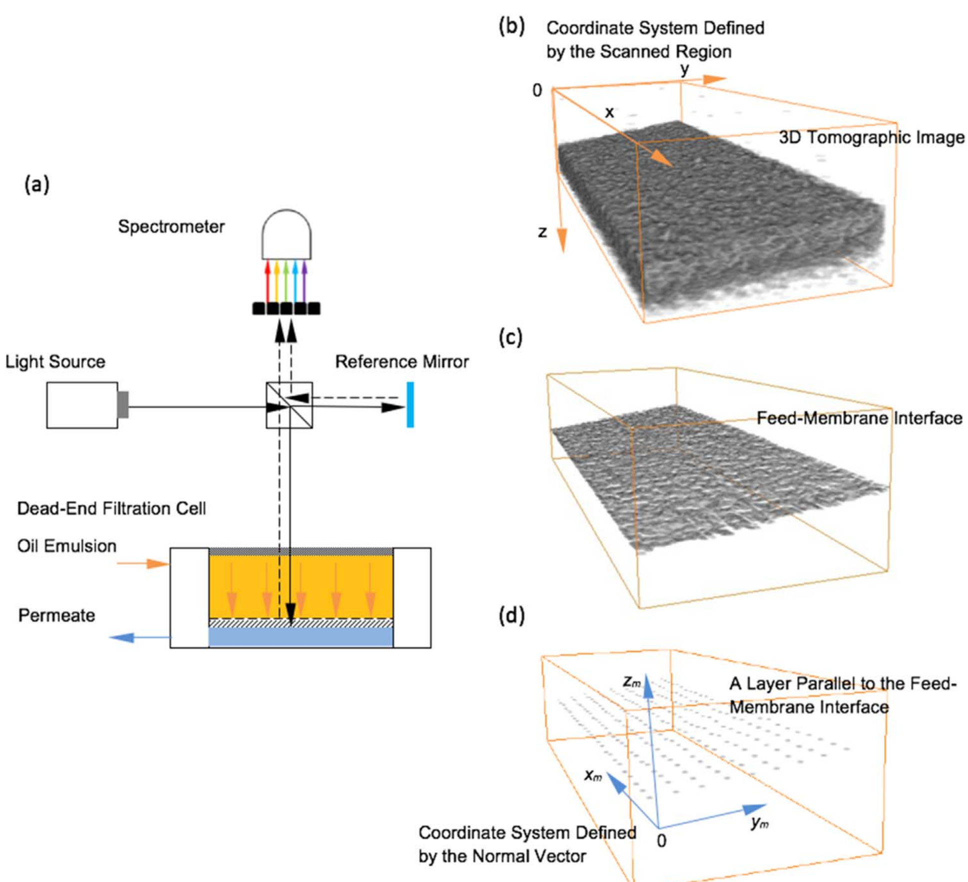

Fig. 1. (a) Schematic showing the principle of a Fourier-domain OCT for characterizing dead-end filtration. (b) A 3D tomographic image with the coordinate system defined based on the scanned region. (c) The feed-membrane interface determined by analyzing the intensity gradient on the 3D tomographic image. (d) A layer constructed parallel to the determined feed-membrane interface.