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Seeing is believing: Insights from synchrotron infrared mapping for membrane fouling in osmotic membrane bioreactors

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ABSTRACT

We employed synchrotron infrared (IR) mapping to resolve forward osmosis (FO) membrane fouling in osmotic membrane bioreactor (OMBR). Synchrotron IR mapping offers a unique perspective to elucidate the fouling mechanisms and associated consequences in OMBR operation. We demonstrated the spatial distribution and relative intensity of carbohydrate and protein longitudinally along of the fouled FO membrane at the conclusion of OMBR operation. Both transmission and attenuated total reflection (ATR) modes were used to map the cross-section and surface of the fouled FO membrane. Micro X-ray computed tomography revealed patchy, "sand-dune" features on the membrane surface at the conclusion of OMBR operation. Synchrotron IR-ATR mapping demonstrated that the development of membrane fouling layer in OMBR operation was initiated by polysaccharide-like carbohydrate, followed by layering with protein-like substance, resulting in a characteristic "sand-dune" three dimensional feature. Synchrotron FTIR mapping shed light on foulant occurrence and accumulation in the draw solution. Strong penetration of protein-like substance into membrane matrix was visualised, resulting the detection of protein adsorption in the region of membrane supporting layer.

1. Introduction

Membrane-based process is a technology triumph in delivering gigalitres of purified water daily from seawater and wastewater (Elimelech and Phillip, 2011; Shaffer et al., 2015; Shannon et al., 2008). By integrating membrane filtration with activated sludge treatment, membrane bioreactor (MBR) has been globally deployed as a state-of-the-art technology for wastewater treatment and reuse (Hai et al., 2014; Judd, 2008). Recent development in MBR has led to the emergence of a new variation, namely osmotic membrane bioreactor (OMBR) (Achilli et al., 2009; Chen et al., 2014; Cornelissen et al., 2011; Nguyen et al., 2016). OMBR utilises forward osmosis (FO), an osmotically driven membrane process, to extract biologically treated water from the mixed liquor into a highly concentrated draw solution. Compared to conventional MBR that employs hydraulically driven membrane processes, such as microfiltration and ultrafiltration, OMBR has been well reported to show a lower fouling propensity, higher fouling reversibility, and better product water quality (Luo et al., 2017a; Wang et al., 2016a).

Despite the low fouling propensity of the FO process (Luo et al., 2015; Mi and Elimelech, 2010), membrane fouling, an accumulation of organic, inorganic, and biological foulants on the membrane surface and/or within the membrane matrix, occurs in OMBR operation (Yuan et al., 2015; Zhang et al., 2012). Membrane fouling of OMBR may be further exacerbated by the high membrane permeability and selectivity, which results in the detrimental accumulation of unfavorable foulants, such as inorganic salts, soluble microbial products, and extracellular polymeric substances, in the bioreactor (Luo et al., 2017a). Membrane fouling hinders the efficiency and productivity of OMBR by decreasing permeate flux, membrane selectivity and service lifetime. As a result, the ability to visualize and characterize the spatial distribution and transport of key membrane foulants in OMBR may play a pivotal role in the design of pre-treatment process, the formulation of operational procedure, and the optimization of cleaning approaches for fouled membranes in practical OMBR operation.

Dedicated efforts were made to characterize membrane fouling,

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such as surface-enhanced Raman spectroscopy (SERS), confocal laser scanning microscopy (CLSM) and transmission electron microscopy (TEM) 3D visualization. For instance, Cui et al. (2011) employed SERS technique to investigate protein fouling on polyvinylidene fluoride (PVDF) membranes as well as to distinguish fouling propensity of three different proteins. A similar vibrational spectroscopic imaging technique was also developed to probe the chemical structure of foulants and their 3-dimensional spatial distribution during microfiltration of protein and polysaccharide solutions (Chen et al., 2017). In addition, CLSM is a non-invasive approach to examine membrane structure (Wang et al., 2012) as well as biofilm characteristics in biofouling (Mukherjee et al., 2016; Xie et al., 2015). Furthermore, 3D visualization by TEM tomography has gained attention to visualize internal structures of polyamide membrane active layer (Kłosowski et al., 2016; Li et al., 2017; Lin et al., 2016; Pacheco et al., 2016). Such ultra-high resolution provides unique perspective in internal nanostructure as well as nano-sized pore distribution of polyamide thin films.

Synchrotron infrared (IR) spectroscopy, utilising the ultra-bright synchrotron IR beamline, offers a novel approach to precisely identify specific foulant molecules with satisfactory spatial resolution. Synchrotron IR spectroscopy has been developed as a rapid, direct, non-destructive, and non-invasive analytical technique for micron-sized samples (Bertrand et al., 2012; Ellis and Martin, 2016; Lehmann and Solomon, 2010). This advanced technique has been applied to a range of environmental applications, such as living cellular exposure and early molecular responses to environmental pollutants (Holman et al., 2000), TiO_2 nanoparticle accumulation in cucumber fruit (Servin et al., 2013), and chemical and structural changes of enzyme on catalysts (Ash et al., 2016). However, the application of synchrotron IR technique in spatially and chemically resolving membrane fouling layer was rather scarce. We previously employed synchrotron IR mapping to resolve combined membrane fouling (organic foulants with colloidal silica) in membrane distillation (MD) (Xie et al., 2017), elucidating the fouling mechanism in MD filtration.

In this study, we employed the synchrotron IR technique to examine membrane fouling in OMBR. Two mapping modes – transmission and attenuated total reflection (ATR) – were utilised to comprehensively investigate membrane fouling development and foulant distribution in OMBR, where membrane fouling develops in a highly heterogeneous environment. We mapped the surface and cross-section of the fouled membrane in OMBR, demonstrating the location and spatial distribution of foulants and extracting chemical information of foulant-membrane interaction. These findings also highlight the efficiency of synchrotron IR technique in characterizing foulant-membrane interfaces.

2. Materials and methods

2.1. OMBR system

A flat-sheet aquaporin FO membrane (Aquaporin Asia, Singapore) was used in OMBR. This aquaporin FO membrane was structurally similar to that of the conventional thin film composite (TFC) FO membrane, but consisted of a polyamide selective layer embedded with aquaporin proteins vesicles and a porous polysulfone supporting layer (Luo et al., 2017b; Madsen et al., 2015). Key mass transport parameters are tabulated in Table S1, Supplementary Data. The combination of these transport parameters resulted in an average FO water flux of approximately 15 $Lm^{-2}h^{-1}$ for 0.5 M NaCl draw solution and deionized water feed solution.

A lab-scale OMBR comprising a bioreactor, a submerged, plate-and-frame FO membrane module and a set of draw solution delivery and control system, was used (Figure S1, Supplementary Data). The membrane module was made of acrylic plastic with a draw solution flow channel of 15 cm long, 8 cm wide, and 0.3 high. The FO membrane was sealed on the module with the active layer facing the mixed liquor and an effective area of 120 cm^2. Details of this OMBR configuration can be found in our previous publications (Luo et al. 2017a, 2018) and Supplementary Data.

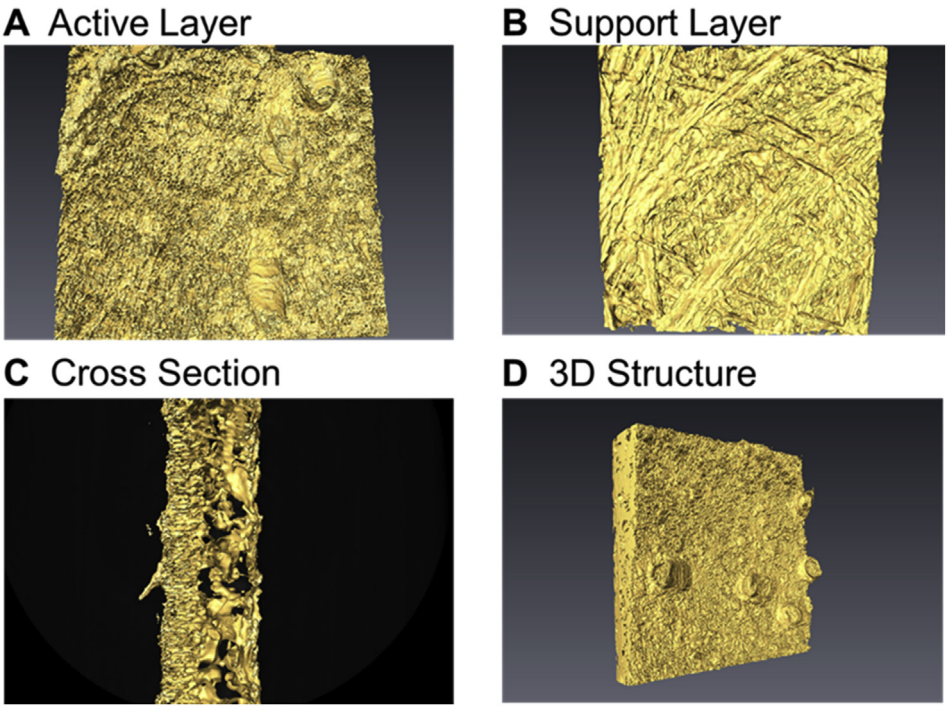

Fig. 1. Three-dimensional (3D) structure of the fouled forward osmosis (FO) membrane in osmotic membrane bioreactor (OMBR) acquired by micro X-ray computed tomography: (A) active layer, (B) supporting layer, (C) cross-section and (D) 3D structure.