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Characterization of biofoulants illustrates different membrane fouling mechanisms for aerobic and anaerobic membrane bioreactors

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ABSTRACT

This study compares the membrane fouling mechanisms of aerobic (AeMBR) and anaerobic membrane bioreactors (AnMBR) of the same reactor configuration at similar operating conditions. Although both the AeMBR and AnMBR achieved more than 90% COD removal efficiency, the fouling mechanisms were different. Molecular weight (MW) fingerprint profiles showed that a majority of fragments in anaerobic soluble microbial products (SMP) were retained by the membrane and some fragments were present in both SMP and in soluble extracellular polymeric substances (EPS), suggesting that the physical retention of SMP components contributed to the AnMBR membrane fouling. One of the dominant fragments was comprised of glycolipoprotein (size 630–640 kDa) and correlated in abundance in AnMBR–EPS with the extent of anaerobic membrane fouling. In contrast, all detected AeMBR–SMP fragments permeated through the membrane. Aerobic SMP and soluble EPS also showed very different fingerprinting profiles. A large amount of adenosine triphosphate was present in the AeMBR–EPS, suggesting that microbial activity arising from certain bacterial populations, such as unclassified Comamonadaceae and unclassified Chitinophagaceae, may play a role in aerobic membrane fouling. This study underlines the differences in fouling mechanisms between AeMBR and AnMBR systems and can be applied to facilitate the development of appropriate fouling control strategies.

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1. Introduction

The membrane bioreactor (MBR) is a treatment process that couples membrane separation to the biological process for solid–liquid separation of the mixed liquor [1]. The integration of a membrane filtration unit achieves better effluent quality and de-couples the sludge retention time (SRT) from the hydraulic retention time (HRT) of the reactor, enabling higher biological oxygen demand (BOD) and chemical oxygen demand (COD) removal efficiency as compared to conventional activated sludge systems [2]. The MBR system can be applied to both aerobic (AeMBR) and anaerobic (AnMBR) treatment processes. Unlike the AeMBR, which has already found widespread application in low to medium strength municipal wastewater treatment [3], AnMBRs have been considered impractical for similar applications due to the perception that comparable transmembrane flux rates are not achievable. Nonetheless, the AnMBR has continued to gain consideration as an alternative treatment technology for municipal wastewater due to its potential advantages in reduced energy input, energy generation by methane production, and low sludge production [4–7].

Despite each MBR type's potential advantages, membrane fouling remains the major obstacle hindering their extensive application. It has been reported that the primary contributor to membrane pore blockage in MBRs is the deposition of the dissolved fraction of activated sludge [8,9]. This form of less-reversible membrane fouling can lead to more rigorous forms of membrane maintenance (e.g. chemical cleaning, backwashing and high cross-flow) being necessary, diminishing the economic viability of operating MBRs for municipal wastewater treatment.

The MBR, being a biological treatment process, contains microbial cells as part of the activated sludge that produces soluble microbial products (SMP) and extracellular polymeric substances (EPS). SMP was defined by Namkung and Rittmann as being comprised of utilization-associated SMP (i.e., UAP) and biomass-associated SMP (i.e., BAP) [10]. On the other hand, EPS is comprised of a matrix of polysaccharides, proteins and other macromolecules, which collectively provide adhesion, aggregation and stabilization functions for microorganisms on a membrane surface [11]. A unified theory put forward by Laspidou and Rittmann further stated that the soluble fraction of EPS is actually SMP [12]. Based on these

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technical definitions of SMP and EPS, much of the existing research has focused on quantification of SMP and EPS components (e.g. proteins, polysaccharides, nucleic acids and so on) and abundance ratios between those individual components in their soluble phase [13–17]. Along with the organic matter from influent wastewater, these non-settleable organic components are one of the primary culprits inhibiting MBR performance based on their role as biofoulants [15,18,19].

Most of the studies that focus on elucidating the role of the soluble components of EPS and SMP on membrane biofouling were carried out in AeMBRs [15,18–20]. Little is known about the differences between AeMBR and AnMBR fouling mechanisms as there have not been any studies that have specifically examined soluble foulants in both systems under the same operating conditions. Instead, it has conventionally being presumed that the fouling mechanisms between aerobic and anaerobic systems are similar and that most of the knowledge related to the fouling mechanisms in AeMBR systems can be extrapolated to the AnMBR [21]. Furthermore, no significant attempts have been made in previous studies to investigate the specific bacterial populations present on MBR membrane foulant layers and their correlation with the occurrence of specific soluble biofoulants contributing to membrane fouling.

In this study, it is hypothesized that AeMBRs and AnMBRs may be subject to different fouling mechanisms arising from the differences in the soluble foulant components generated by the microbial communities of both systems. To address this hypothesis, an AeMBR and AnMBR were operated using a similar reactor configuration that combined an upflow attached-growth (UA) reactor with three-successive PVDF membrane filtration units, and were operated for varying time periods of up to 9 weeks to reflect different extents of membrane fouling. Quantification of the protein and carbohydrate concentrations was coupled with high performance size exclusion chromatography to determine the molecular weight distributions of proteins and polysaccharides in SMP in the permeate and retentate streams of both the AeMBR and AnMBR. The same quantification procedure was also performed on the soluble EPS from biomass attached on both aerobic and anaerobic membranes. Specific microbial populations that correlated with the extent of membrane fouling and the biofoulants were further examined using high-throughput sequencing, and their bioactivity levels evaluated by measuring adenosine triphosphate (ATP) and quorum-sensing signal molecule concentrations. Ultimately, this study aims to provide a direct multifaceted comparison of the differences in biofouling mechanisms between AeMBR and AnMBR systems.

2. Materials and methods

2.1. Reactor configuration and operating conditions

To facilitate the comparison of aerobic and anaerobic systems, the same upflow-attached (UA) reactor configuration was applied to both AeMBR and AnMBR systems (Fig. 1). This reactor configuration was evaluated in this study as it was previously found to have a positive role in controlling membrane fouling in MBRs [22]. Both UA reactors were filled with ceramic ring media with an average 1.5 cm diameter and length. The seed sludge in both systems originated from the same source, and comprised of camel manure and anaerobic sludge from a wastewater treatment plant in Riyadh, Saudi Arabia [23]. No oxygen or air supply was provided in the AnMBR. Aeration was supplied to the AeMBR by two air diffusion stones at the bottom of the reactor to achieve more than 2 mg/L O_2 throughout the AeMBR system. Both reactors were fed with a synthetic wastewater of 750 mg/L carbon oxygen demand

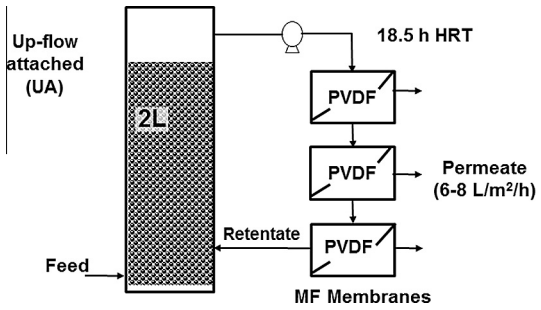

Fig. 1. Operational setup for both aerobic MBR and anaerobic MBR. An attached-growth reactor configuration was used and the MBR systems were operated at 18.5 h hydraulic retention time (HRT) and with flux of 6–8 L/m^2/h through the microfiltration (MF) membrane.

(COD) [24], and operated at 35 °C and pH of 7. Hydraulic retention times (HRTs) of both reactors were 18.5 h.

Prior to connection with membrane separation units, both UA reactors were at steady-state operation and stable performance conditions. The UA reactors were individually connected to three PVDF microfiltration (MF) membrane modules in external cross-flow mode. Membrane modules were connected in series along the recirculation line with a recirculation to effluent flow ratio of 500:1. The MF membranes were JX model MF PVDF (GE Osmonics, Minnetonka, MN, USA) and had a nominal pore size of 0.3 μm. Constant flux was maintained at 6–8 L/m^2/h (LMH) while changes in transmembrane pressure (TMP) were recorded by a pressure gauge connected to each membrane unit. There was no sludge wasted in the anaerobic system, while 150 mL of sludge suspension was taken from the aerobic reactor per day, resulting in a sludge retention time (SRT) of 13 days. This was done to maintain a MLSS level comparable to that in the anaerobic MBR.

2.2. Soluble microbial products (SMP) and soluble extracellular polymeric substances (EPS) sampling procedure

Permeate from each membrane cassette as well as the retentate in both systems were sampled weekly throughout the course of the experiment, and were collectively referred to as soluble microbial products (SMP) in this study. All SMP samples were centrifuged at 9400 *g* for 10–30 min. The supernatant was filtered with a 0.2 μm cellulose acetate membrane and stored at −20 °C prior to analyses. Analyses for SMP included determination of COD, protein and carbohydrate concentrations and the corresponding fingerprinting profiles of proteins and carbohydrates based on molecular weight (MW) fragments. Membranes were harvested from the AeMBR at time intervals of 3, 4 and 5 weeks. Membranes were harvested from the AnMBR at time intervals of 3, 6 and 9 weeks. The sampling intervals were decided based on the measured TMP and chosen to represent different extents of fouling on the membranes (Fig. 2). To harvest the membrane at each time point, one of membrane cassettes was removed from the successive filtration unit, and replaced with a new membrane module to ensure constant operating conditions throughout the system. Each harvested membrane with an area of 20 cm by 2.5 cm was sectioned into three equal parts, namely inlet, mid and outlet, based on the flow direction of the wastewater stream. Soluble EPS was extracted from each section as follows: the membrane with an area of 4 cm by 2.5 cm was cut into small strips and dispersed into two 2 mL microcentrifuge tubes. 2 mL 1X PBS was added into each tube. The tubes were ultrasonicated by a QSonica Q500 Sonicator (QSonica LLC, Newton, CT, USA) for 5 min at 25% amplitude and with 2 s pulsating intervals. The membrane strips were then removed and the remaining suspension centrifuged at 9400 *g* for 30 min. The

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