Science of the Total Environment 642 (2018) 77–89

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Integrated interrogation of causes of membrane fouling in a pilot-scale anoxic-oxic membrane bioreactor treating oil refinery wastewater

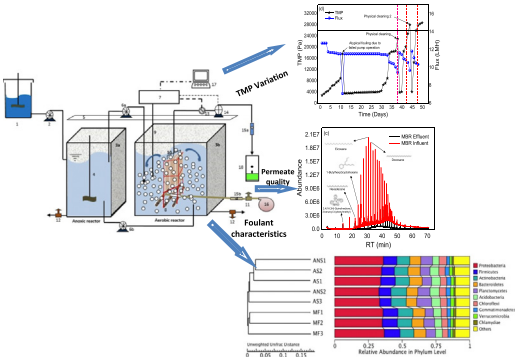
Olusegun K. Abass [a,b], Fang Fang [a], Maoshui Zhuo [a,b], Kaisong Zhang [a,*]

[a] CAS Key Laboratory of Urban Pollutant Conversion, Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, China
[b] University of Chinese Academy of Sciences, Beijing 100049, PR China

HIGHLIGHTS

- Advanced chemical and Illumina sequencing unveils key MBR foulants in ORW treatment.
- Less biodegradable hydrocarbon and organophosphonate groups are key MBR foulants.
- Emulsified oil with mean sizes >0.5 μm are potential membrane foulants in the MBR.
- Bio-colonization of membrane surface in the MBR is a result of species sorting.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 8 April 2018
Received in revised form 3 June 2018
Accepted 4 June 2018
Available online xxxx

Editor: Zhen (Jason He)

Keywords:
Emulsified oil
Illumina sequencing
Membrane bioreactor
Membrane fouling
Microbial community
Oil refinery wastewater

ABSTRACT

Studies on membrane fouling during treatment of oil refinery wastewater (ORW) via membrane bioreactor (MBR) are currently lacking, and associated fouling challenges are largely undocumented. Using advanced chemical and Illumina sequencing approach, we investigated the complex bio-physiochemical interactions responsible for foulant-membrane interactions during treatment of ORW. After nearly 2 months of the MBR operation, COD removal reached maximal of 97.15 ± 1.85%, while oil and grease removal was maintained at 96.6 ± 2.6%, during the treatment duration. Most of the less or non-biodegradable oil moieties (>0.5 μm) progressively accumulated on the membrane as the influent oil concentration increased. Presence of relatively higher unsaturated extracellular polymers (100.6 mg/g VSS) like fulvic acid and aromatic-like compounds at high volumetric loading (~18.7 kg COD/m^3/d), enhanced the adsorption of chemical elements (Fe = 88.9, Al = 63.4, and Ce = 0.56 mg/g dry-sludge, respectively). Moreover, shift in microbial community structure to hydrocarbon-utilizing and metals-tolerating genera, as *Comamonas* and *Rhodanobacter*, respectively, uncovers major membrane colonizers in ORW treatment via MBR.

1. Introduction

The global demand of crude oil and natural gas in modern day industrialization is ever increasing. However, like most production activities, natural gas and oil production processes generate large streams of wastewaters. These wastewaters are derived from a number of crude oil and gas drilling and refining activities, which results in streams of varied organic and inorganic compositions and concentrations with potent capabilities to migrate downstream to pollute groundwater, or spill into surface waters causing large-scale environmental disturbances (Fakhru'l-Razi et al., 2009). In particular, oil refinery wastewaters

* Corresponding author.
E-mail address: kszhang@iue.ac.cn (K. Zhang).

https://doi.org/10.1016/j.scitotenv.2018.06.049
0048-9697/

(ORW) contains varied class of pollutants such as oil, dissolved solids, phenols, sulfides, and toxic metals, which encumbers available treatment facilities and thus, makes this class of wastewaters very challenging to treat via conventional methods (Santos et al., 2016).

The vast prospect in membrane bioreactor (MBR) applications for remediation of different class of industrial wastewater matrices including ORW has been identified (Rahman and Al-Malack, 2006; Razavi and Miri, 2015; Yu et al., 2018). However, a major drawback associated with MBR treatment of ORW is membrane fouling (Abass et al., 2015; Munirasu et al., 2016; Padaki et al., 2015). These drawbacks are intricately tied to different factors including the complex biology (activated sludge and biofilms), chemicals (influent characteristics and inorganic elements), materials (membrane type and characteristics), and processes (operating conditions and reactor properties) affecting the sustained long-term treatment of ORW in MBR systems (Lin et al., 2011; Padaki et al., 2015; Qin et al., 2015; Shariati et al., 2011; Viero et al., 2008).

For instance, Viero et al. (2008), investigated the effect of high organic load in a submerged MBR (SMBR) treating oil refinery wastewater during long-term operation and observed that modification of the feed characteristics resulted in increased production of polysaccharide fractions, which aggravated the membrane permeability. However, the interaction leading to the membrane performance degradation was less explored. In another report by Rahman and Al-Malack (2006), a laboratory scale cross-flow MBR was utilized for the treatment of petroleum refinery wastewater, which showed good COD removal efficiency of over 93%. However, information regarding membrane fouling were scantly discussed. More recently, Razavi and Miri (2015) explored the treatment of real petroleum refinery wastewater (PRW) in a hollow fiber MBR. Details on the MBR treatment performance and sludge characteristics were reported, but the membrane fouling characteristics of the real PRW were less examined.

Similarly, Shariati et al. (2011) focused mainly on the effect of hydraulic retention time (HRT) in relation to its impact on extracellular polymeric substances (EPS) and soluble microbial products (SMP) production, and the accompanying fouling implications. Increased contributions on fouling characteristics and control were made by Pajoumshariati et al. (2017) and Qin et al. (2015), where they both utilize physical and chemical method to investigate fouling development and control during treatment of PWR/oily wastewater in membrane sequencing batch reactors (MSBR) and SMBR, respectively. However, information regarding chemical and biological interactions in relation to the fouling propensity of the membrane were lacking in the study. Thus, the evident shortage of research on the complex causes of fouling in MBR systems treating ORW can potentially hamper the promising prospect of MBR for ORW remediation and reuse.

Therefore, studies that seek to understand membrane fouling development in MBR applications treating ORW are urgently needed. Accordingly, this paper aims to interrogate via an integrated approach the complex physical, chemical and biological interactions responsible for membrane fouling during treatment of ORW in MBR. Findings on the influent compositional characteristics and its influence on membrane fouling were made. The inextricable links between chemical foulants and bio-products were discussed, and finally, we showed via high-throughput sequencing the microbial distribution and structure of membrane colonizers during treatment of oil refinery wastewaters in MBR systems.

2. Materials and methods

2.1. Pilot scale anoxic-oxic MBR design and operation

The pilot scale anoxic-oxic MBR (A/O-MBR) plant was built at the Urban Pollution Conversion Centre of the Institute of Urban Environment, Xiamen as shown in the Fig. 1. Other design characteristics of the MBR setup, membrane type and operating parameters are presented in Table 1. The model wastewater samples based on oil-refinery effluent (Alexandre et al., 2016) were prepared by emulsifying a mixture of motor oil (Shengao, China) and Phenol (Sinopharm Chemical Reagent Co. Ltd., China) with Pluronic F-127 (Sigma Aldrich, USA), which serves two purposes; emulsification of the oil molecules, and to reduce coalescence in the MBR sludge, thereby enabling higher cells oxygen contact (Alexandre et al., 2016). Mean sizes and zeta-potential of the oil-in-water emulsions were characterized using Zetasizer (Nano ZS, Malvern). The synthetic wastewater was augmented with essential nutrients (including NH_4Cl, KH_2PO_4, $CaCl_2.2H_2O$, $NaHCO_3$, $MgSO_4.7H_2O$, $CoCl_2.6H_2O$, $FeCl_3$, and $MnSO_4$ in proper amounts) to aid microbial activity and the A/O-MBR was operated at volumetric loading of ~1.6 kg COD/m^3/d to ~18.7 kg COD/m^3/d for about 2 months to mimic periods of low and high volumetric loading rates. The reactors were operated under low volumetric loading for 20 days at 1.6 to 2.5 kg COD/m^3/d and under high volumetric loading for 30 days at 5.9 to 18.7 kg COD/m^3/d. Physical cleaning only (to ensure that natural surface environment of the membrane is preserved to meet the experimental aim) were performed, as the transmembrane pressure increased to 23 kPa and 28 kPa, respectively.

2.2. Sampling and analysis

Influent and effluent water samples were collected from the A/O-MBR set-up daily and weekly for COD, oil and grease (O&G) analyses, respectively. Concentrations and compositions of O&G were quantified using standard gravimetric method (USEPA, 2009) and gas chromatography–mass spectrometry (GC–MS) investigation, respectively (Abass et al., 2017). Procedure for the O&G sample extraction is described in the Supporting information (SI). Other physicochemical indices such as COD, TOC, MLSS, MLVSS and TDS were measured in accordance with Standard Methods for the Examination of Water and Wastewater (APHA, 2005).

Triplicate samples of 10 g each, mixed liquor (anoxic and oxic) and membrane foulant were retrieved during physical cleaning (PC) episodes corresponding to TMP jumps (on Day 11, 37 and 50) for chemical, spectroscopic and microbial community analysis. Foulant samples were collected by scrapping off the sludge cake along with the thin gel layer using plastic collectors at different times corresponding to the cleaning periods. The membrane surface was subsequently flushed with tap water and re-inserted in the MBR module for further use. All samples collected for DNA extraction were immediately frozen under sub-zero temperature (−20 °C).

2.3. Spectroscopic and chemical analysis of mixed liquor and foulant components

SMP and EPS were extracted from the A/O-MBR mixed liquor and foulant layer using heat treatment method as described by Zhang et al. (2011). Briefly, harvested samples were centrifuged at 7000 rpm (at 4 °C) for 10 min, followed by filtration of 3 mL of the supernatant through a 0.22 μm PTFE membrane. The resulting filtrate represents the SMPs. Subsequently, the dewatered pellet was washed and re-suspended to its initial volume using a buffer solution (165 mM Phosphate-buffered saline – 1.094 g Na_2HPO_4, 0.277 g NaH_2PO_4 and 8.476 g NaCl, at pH 7.2), and mixed for 10 min. The resulting mixed liquor was heat treated at 80 °C for 10 min, and centrifuged at 10,000 rpm at 4 °C for 10 min. The recovered filtrate after centrifugation was regarded as the EPS. The Anthrone sulfuric method (Koehler, 1952) and the modified Bradford method (López et al., 1993) were used for determination of the polysaccharides and proteins fractions, respectively. Three-dimensional fluorescence excitation-emission matrix (EEM) spectral fingerprints of the foulant and bulk sludge supernatant samples (filtered through a 0.45 μm cellulose membrane) were collected using luminescence spectrometry (F-4600 FL spectrophotometer, Hitachi, Japan) as described by Wang et al. (2010). Origin Pro 9.0 software