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The study of organic removal efficiency and halophilic bacterial mixed liquor characteristics in a membrane bioreactor treating hypersaline produced water at varying organic loading rates

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HIGHLIGHTS

- >95% O&G removal from synthetic PW in MBR with halophilic consortium at high OLR.
- O&G and SMP transient accumulation occurred but did not affect removal performance.
- Bioflocculation and compressibility of flocs affected by EPS and surface charge.
- At high OLR non-Newtonain rheology and EPS bulking in the mixed liquor was observed.
- Membrane fouling mainly due to cake formation at highest OLR of 2.6 kg COD m^{-3} d^{-1}.

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ABSTRACT

In this study the organic pollutant removal performance and the mixed liquor characteristics of a membrane bioreactor (MBR), employing a halophilic bacterial consortium, for the treatment of hypersaline synthetic produced water – at varying organic loading rates (OLR) from 0.3 to 2.6 kg COD m^{-3} d^{-1} – were considered. The oil and grease (O&G) and COD removal efficiency were 95–99% and 83–93%, respectively with only transient O&G (mainly polycyclic aromatic hydrocarbons) and soluble microbial products accumulation being observed. With increasing OLR, in the range 0.9–2.6 kg COD m^{-3} d^{-1}, as a result of change in both extracellular polymeric substances (EPS) and zeta potential, bioflocculating ability improved but the compressibility of the flocs decreased resulting in the occurrence of EPS bulking at the highest OLR studied. The latter resulted in a change in the rheology of the mixed liquor from Newtonian to non-Newtonian and the occurrence of significant membrane fouling.

1. Introduction

Mixed halophilic bacterial populations have been recently considered for the industrial treatment of hypersaline wastewaters (usually considered as those having salt concentration higher than 30 g L^{-1}) (Fakhru'l-Razi et al., 2010; Lay et al., 2010; Lefebvre and Moletta, 2006; Pendashteh et al., 2012). Such wastewaters are generated by industries such as the oil and gas, leather and food-processing. According to previous research, halophilic bacterial consortia (or activated sludge bioaugmented with halophilic bacterial cultures) should be employed in order to achieve satisfactory biological removal of organic pollutants from such wastewaters (Lay et al., 2010; Lefebvre and Moletta, 2006). The presence of a diverse range of bacterial species in natural halophilic mixed bacterial populations capable of biodegrading a range of industrially important organic pollutants has been shown in previous work (Diaz et al., 2000; Fakhru'l-Razi et al., 2010; Lefebvre and Moletta, 2006; Pendashteh et al., 2012).

Produced water (PW) is considered as the most important wastewater of the oil industry and contains a variety of oily compounds such as n-alkanes as well as aromatic and polycyclic aromatic hydrocarbons. The salt concentration of produced water, depending on the location of the oil well, may range from a few mg L^{-1} to 300,000 mg L^{-1} (Fakhru'l-Razi et al., 2010). Therefore, high salinity PW is an important example of a hypersaline wastewater.

Membrane bioreactor (MBR) technology has been considered for treatment of saline wastewater as the presence of membrane can potentially counteract the negative impact of salinity on the treatment of biological treatment processes containing activated sludge (Jang et al., 2013). However, there is scarcity of information

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for the application of MBR for treatment of hypersaline PW. This is because halophilic bacterial consortium – as opposed to activated sludge – should be employed for this application but – unlike activated sludge – only limited information is available on the removal performance and halophilic bacterial mixed liquor characteristics inside MBRs. The research group of Professor Fakhru'l-Razi in Malaysia (Fakhru'l-Razi et al., 2010; Pendashteh et al., 2012) have considered the use of an external membrane sequencing bioreactors (MSBR) inoculated with a halophilic bacterial consortium for the treatment of both synthetic and real PWs. However, there is only one previous report of the use of submerged MBR (which is more industrially relevant compared to external MSBR) for the treatment of a hypersaline PW (Abdollahzadeh Sharghi et al., 2013). In that study the organic removal performance for organic loading rates (OLRs) up to 0.9 kg COD m^{-3} d^{-1} was studied and biomass characteristics were only investigated to a limited extent. However, a proper assessment of MBR technology for industrial treatment of hypersaline PW necessitates the considerations of much higher OLRs and should include a detailed study of halophilic bacterial mixed liquor characteristics such as mixed liquor suspended solids (MLSS)/mixed liquor volatile suspended solids (MLVSS), extracellular polymeric substances (EPS) and soluble microbial products (SMP) concentration, the size, surface charge and structure of bacterial flocs and mixed liquor rheology. Also, in the study by Abdollahzadeh Sharghi et al. (2013) – due to the use of fairly low OLR – no membrane fouling was observed. However, at relatively high OLR membrane fouling is likely to occur and information regarding such fouling (i.e., what OLR initiates it and what is its nature and cause) is essential for proper assessment and design of MBRs for treatment of hypersaline wastewaters. Although information on membrane fouling in MBRs for saline wastewater (up to 30 g L^{-1} NaCl) is available to some extent (Jang et al., 2013), to the knowledge of the authors such information for hypersaline wastewaters has not been previously reported for submerged MBRs.

The physicochemical and morphological characteristics of mixed liquor in MBRs – which can be affected by OLR – have important bearing on both the removal performance and membrane fouling rates (Meng et al., 2009). For example, EPS and SMP have been implicated in membrane fouling in MBR (Gao et al., 2013; Zuthi et al., 2013). On the other hand, according to previous work, both the bioflocculating ability of the bacterial population in activated sludge as well as the compressibility of the flocs are affected by the value of EPS and zeta potential (ZP) (Li et al., 2013; Liao et al., 2001; Peng et al., 2012; Sheng et al., 2010; Wilen et al., 2008). The effect, however, is not very clear cut (Peng et al., 2012), and to the knowledge of the authors has not been studied for halophilic bacterial consortia.

Another important characteristic of the mixed liquor inside MBR is its rheology. Whether the mixed liquor is Newtonain or non-Newtonian and the actual value of viscosity or apparent viscosity can have important bearing on the rate of oxygen transfer, extent of mixing, and the rate of membrane fouling. For example, increase in (apparent) viscosity can lead to reduced oxygen and mixing rates which means that higher air rates (and hence higher operating costs) are needed to achieve acceptable pollutant removal performance. Also, according to Meng et al. (2007) increase in apparent viscosity of the sludge suspension over 2 mPa s will cause a sharp decrease in membrane cross flow velocity, which means that the use of air sparging near to the sparger for retarding membrane fouling will not be very efficient. Knowledge regarding the rheological characteristics of halophilic mixed liquor at various OLR is therefore essential for the design and assessment of MBRs to be used for treatment of hypersaline PW.

The aim of the present study was the assessment of the organic pollutant removal performance and a comprehensive characterization of the halophilic bacterial mixed liquor properties (MLSS/MLVSS/bacterial population, SMP and EPS and their components, particle size distribution (PSD), supernatant turbidity, sludge volume index (SVI), floc surface charge and rheology) including elucidation of their interrelationships in a MBR treating hypersaline synthetic PW at varying OLR in the range 0.3–2.6 kg COD m^{-3} d^{-1}. Organic compounds accumulation in the mixed liquor – which has been previously reported for MBRs operating at high SRT (Masse et al., 2006; Shin and Kang, 2003) and/or treating oily wastewater (Pendashteh et al., 2012; Scholz and Fuchs, 2000) – was also studied in detail.

2. Methods

2.1. The mixed halophilic bacterial inoculum

The halophilic bacterial consortium used to inoculate the MBR was originally enriched from a saline soil sample contaminated through an oil spill and adapted to the synthetic PW by repeated subculturing in fresh PW for a period of 6 months. Before addition to the MBR, the halophilic consortium was cultured in an aerated vessel using a synthetic PW with the same composition as described in Section 2.2, except that it lacked Tween 80 and the concentration of $CaCl_2$ was set at 1.7 g L^{-1}. Because of the absence of Tween 80, the synthetic PW was homogenized at 3000 min^{-1} for 17 h (Abdollahzadeh Sharghi et al., 2013).

Identification of the halophilic bacterial species in the consortium present in a sample from the MBR during its operation using the 16S rDNA (16S ribosomal RNA) analysis revealed the presence of the following species: *Marinobacter hydrocarbonoclasticus*, *Marinobacter oulmenensis*, *Halomonas neptunia*, *Halomonas shengliensis*, *Halomonas alimentaria*, *Psychroflexus halocasei*, *Idiomarina loihiensis* (Abdollahzadeh Sharghi et al., 2013).

2.2. Synthetic produced water preparation

The synthetic PW was prepared by adding appropriate volumes of crude oil (Table 1), as the only carbon source, to a mineral salt medium containing (in g L^{-1}) $CaCl_2{\cdot}2H_2O$: 0.06; KCl: 2.0; $MgCl_2{\cdot}6H_2O$: 0.05; $NaHCO_3$: 0.8, supplemented with 50.0 g L^{-1} NaCl. The composition of the mineral salt medium was chosen according to the requirements of halophilic bacteria (Pendashteh et al., 2012). The pH of the synthetic PW was adjusted to around 7 with 0.8 g L^{-1} $NaHCO_3$. The crude oil was obtained from an Iranian oilfield (Hengam oilfield, Khark). The C/N/P ratio of the medium was adjusted to approximately 100/10/1 by incorporating appropriate concentrations of NH_4Cl and KH_2PO_4 in the synthetic PW. In order to obtain a fairly stable synthetic PW, Tween 80 was added at a ratio of 1:8 (V/V) to crude oil and the synthetic PW prepared by mixing the constituents in a homogeniser operated at 3000 min^{-1} for 2 h. The TDS, salinity and conductivity of the PW was measured as 64.4 g L^{-1}, 7.12% and 96.8 mS cm^{-1}, respectively.

2.3. Experimental setup

A schematic diagram of the laboratory scale MBR setup used in the present study is presented in Fig. 1. Details of the setup and mode of operation have been described elsewhere (Abdollahzadeh Sharghi et al., 2013). The MBR system was inoculated with the mixed halophilic bacterial consortium (initial bacteria concentration of 1.6 ± 0.4 CFU mL^{-1} ($\times 10^7$), MLSS of 1143 ± 91 and MLVSS of 685 ± 72) operated over a period of 242 days with SRT of 80 days. During this period OLR was increased through increase in the chemical oxygen demand (COD) of the synthetic PW and