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Bioresource Technology 113 (2012) 201–206

Contents lists available at SciVerse ScienceDirect

Bioresource Technology

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

Performance of a novel osmotic membrane bioreactor (OMBR) system: Flux stability and removal of trace organics

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ARTICLE INFO

Article history:
Available online 25 January 2012

Keywords:
Membrane bioreactor
Forward osmosis
Trace organic contaminants
Wastewater treatment

ABSTRACT

Results reported here highlight the potential and several challenges in the development of a novel osmotic membrane bioreactor (OMBR) process for the treatment of municipal wastewater. Following the initial gradual decline, a stable permeate flux value was obtained after approximately four days of continuous operation. There was evidence of continuous deterioration of biological activity of the OMBR system, possibly due to the build-up of salinity in the reactor. The removal of 25 out of 27 trace organic compounds with molecular weight higher than 266 g/mol was above 80% and was possibly governed by the interplay between physical separation of the FO membrane and biodegradation. In contrast, the removal efficiency values of the other 23 trace organic compounds with molecular weight less than 266 g/mol were very scattered. The removal efficiency of these low molecular weight compounds by OMBR treatment appears to depend mostly on biological degradation.

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1. Introduction

The occurrence of a wide range of trace organic contaminants in treated and untreated domestic wastewater has been identified as a significant environmental health concern (Ternes et al., 2004). Although most of these contaminants remain unregulated, there is a growing consensus globally among the scientific community and water authorities that their optimised removal during wastewater treatment is a justifiable and prudent approach to environmental protection. Recent trends towards indirect potable water reuse in many metropolitan and arid areas around the world provide further impetus for the effective treatment of trace chemical contaminants (Farré et al., 2011). It is well established that conventional water and wastewater treatment processes are not adequate for the effective removal of many trace organic contaminants (Ternes et al., 2004). The need for a more robust treatment process has triggered many dedicated scientific investigations to develop new methods to obtain high quality treated effluent from domestic wastewater, particularly with a focus on the removal trace organic contaminants (Cartinella et al., 2006; Shannon et al., 2008; Alturki et al., 2010; Cath et al., 2010; Bernabeu et al., 2011; Li et al., 2011; Patterson et al., 2011).

Recent developments in water science and technology have established membrane bioreactor (MBR) systems as a potential alternative to conventional activated sludge (CAS) treatment processes (Guo et al., 2008; Sahar et al., 2011). In comparison to CAS, MBR is more robust with a much smaller physical footprint and improved effluent quality (Judd, 2008). Evidence has emerged that MBR technology can offer an enhanced removal efficiency for moderately biodegradable and hydrophobic trace organics compared to CAS treatment (De Wever et al., 2007). MBR systems usually operate at a much higher mixed liquor suspended solids (MLSS) concentration than that of CAS reactors (Judd, 2008). It has been systematically demonstrated that during MBR treatment, hydrophobic trace organic contaminants can adsorb to the MLSS, resulting in a longer retention time in the bioreactor, hence leading to enhanced removal efficiency (Tadkaew et al., 2011). However, current MBR systems are not effective for the removal of some persistent and hydrophilic trace organic contaminants (Tadkaew et al., 2011). Existing MBR systems use large pore size membranes (either microfiltration or ultrafiltration) (Visvanathan et al., 2000). While these membranes can effectively retain both particulate matter and some pathogenic agents, they are not able to retain low molecular weight organic compounds. For these compounds, the retention time is the same as the hydraulic retention time, which is usually very short in a typical MBR process. As a result, conventional MBR is only effective for the removal of most but not all trace organic contaminants.

Forward osmosis (FO) is another emerging water treatment technology that has been the subject of numerous investigations in recent years (Cath et al., 2006; Cath et al., 2010; Wei et al., 2011; Chung et al., in press). In FO, water is extracted from a feed solution using the high osmotic pressure of a concentrated 'draw

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doi:10.1016/j.biortech.2012.01.082

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solution' that flows along the permeate side of an FO membrane. A reverse osmosis or distillation process can be used to reconcentrate the draw solution for reuse in the FO process and to produce purified water. A low energy technique to recover the draw solutes has also been proposed (McCutcheon et al., 2005; McCutcheon et al., 2006). McCutcheon et al., 2005 successfully demonstrated a novel FO process using ammonium bicarbonate as the draw solution. Upon moderate heating (approximately 60 °C), ammonium bicarbonate decomposes into ammonia and carbon dioxide gases that can be separated from the product water and recycled as draw solutes. FO operates at virtually zero (or very low) hydraulic pressure. As a result, it has a lower membrane fouling propensity and the fouling is much more controllable than pressure-driven membrane processes such as nanofiltration and reverse osmosis (Lee et al., 2010). The configuration of an FO system can be very simple because only minimal hydraulic pressure is required in the FO process to overcome flow resistance in the membrane module. The use of FO for water and wastewater treatment has been evaluated in numerous bench scale and pilot scale studies (Cath et al., 2006). Full scale application of FO for landfill leachate treatment has also been reported (Cath et al., 2006). In addition, FO has the potential to effectively remove a wide range of contaminants of concern in typical water and wastewater treatment applications, although this particular application is yet to be fully substantiated (Cartinella et al., 2006; Cath et al., 2010).

A novel concept of combining activated sludge treatment and FO membrane separation for wastewater treatment has been recently explored by a few research groups (Cornelissen et al., 2008; Achilli et al., 2009; Qin et al., 2009). This process is commonly referred to as an osmotic membrane bioreactor (OMBR). OMBR retains the inherent advantages of both MBR and FO. The high rejection capacity of the FO separation process can effectively retain small and persistent trace organic contaminants in the biological reactor, thus significantly prolonging their retention time in the reactor and subsequently facilitating their biodegradation. Limited evidence from three recent short term bench scale studies (Cornelissen et al., 2008; Achilli et al., 2009; Qin et al., 2009) available in the open literature indicates that OMBR may offer a simple and elegant technological solution for the production of high quality effluent for water reuse or for effluent discharge in environmentally sensitive areas. Nevertheless, the OMBR concept is still in its infancy. In particular, the removal efficiency of trace organic contaminants by OMBR has, until now, not been evaluated. This paper provides and assessment of the technical feasibility of OMBR for wastewater treatment applications. Permeate flux and basic biological performance of a laboratory scale OMBR system were systematically investigated. The removal of 50 trace organic contaminants over an extended operation period is reported and discussed in detail.

2. Methods

2.1. Forward osmosis membrane

An FO membrane supplied by Hydration Technologies Inc (Albany, Oregon, USA) was used in this study. The physical characteristics of this FO membrane are unique compared to other thin film composite membranes commonly used for nanofiltration or reverse osmosis applications. Although the information regarding actual membrane composition has not been disclosed, it is well known that the active layer of the membrane is made mainly of cellulose acetate. According to McCutcheon et al., 2006 this is an asymmetric membrane consisting of a thin polyester mesh embedded within the porous support portion of the membrane. The polyester layer provides mechanical strength to the membrane while the cellulose acetate layer is wholly responsible for solute separation.

2.2. Trace organic contaminants

In this study, 50 organic compounds (Supplementary data Table S1) were selected to represent four major trace organic groups of concern in water reuse applications (pharmaceutically active compounds, pesticides, steroid hormones, and other endocrine disrupting chemicals). The occurrence of many of these trace organic compounds in domestic sewage has been widely reported in the literature. Their molecular weights are in the range from 138 to 458 g/mol. Thus these selected trace organic compounds are not expected to be retained by microfiltration or ultrafiltration membranes, which are usually employed in conventional MBR systems. All selected trace organic compounds were of analytical grade. A combined stock solution was prepared in pure acetonitrile. The trace organic stock solution was kept in a freezer and was used within less than a month.

2.3. Forward osmosis system

FO experiments were conducted using a closed-loop laboratory scale membrane system consisting of a membrane cell, circulation pumps, a conductivity control device, and a temperature control unit (Supplementary data Figures S1 and S2). The membrane cell was designed to hold a flat-sheet membrane under moderate pressure gradients without any physical support. The flow channels were engraved in each of two Plexiglass blocks that made up the feed and draw solution semi-cells. Each channel was 0.2 cm deep, 10.5 cm wide and 15.5 cm long. The total active membrane area for mass transfer was 162 cm^2.

Feed solution was circulated from a 5 L glass reservoir through the feed membrane semi-cell and back to the reservoir. Analytical grade sodium chloride was used as the draw solute. Draw solution was circulated from a 1.2 L glass reservoir through the other semi-cell and back to the reservoir. The overflow from the draw solution reservoir as a result of the permeation of water through the FO membrane was continuously weighed on an electronic balance (PB32002-S, Mettler Toledo Inc., Hightstown, NJ). The draw solution reservoir was also equipped with a conductivity device which would activate a peristaltic pump to transfer concentrated draw solution of 6 M NaCl to the reservoir once the conductivity fell below the set point. The concentrated draw solution reservoir was also placed on the balance to account for the transfer of concentrated draw solution to the draw solution reservoir. The temperatures of the feed and draw solutions were kept constant throughout the experiment using a temperature control unit (Neslab RTE 7, Thermo Fisher Scientific, Waltham, MA, USA) equipped with a stainless steel heat-exchanging coil, which was submerged in the feed and draw solution reservoirs. Two gear pumps (Model 120/IEC71-B14, Micropump Inc., Vancouver, WA, USA) were used to circulate feed and draw solution from their respective reservoirs through the membrane cell and back to the reservoirs. Flow rates of the feed and draw solution were monitored using two rotameters and were 1.5 L/min (equivalent to a cross flow velocity of 4 cm/s).

During the OMBR experiment, the feed solution reservoir was modified into a biological reactor. An air pump (Heilea, model ACO 012) was used to supply oxygen to the mixed liquor solution via a diffuser located at the bottom of the reactor. The dissolved oxygen concentration within the reactor was maintained at 2 mg/L. A peristaltic pump was used to circulate the feed solution instead of the gear pump mentioned above to avoid any grinding effect on the activated sludge.

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2.4. Experimental protocol

Synthetic wastewater was used in this study to simulate medium strength municipal sewage. The synthetic wastewater was prepared daily and consisted of glucose (100 mg/L), peptone (100 mg/L), KH_2PO_4 (17.5 mg/L), $MgSO_4$ (17.5 mg/L), $FeSO_4$ (10 mg/L), urea (35 mg/L) and sodium acetate (225 mg/L). This composition was modified from a high strength wastewater used in a previous study by Tadkaew et al., 2010. Activated sludge was obtained from another MBR which had been continuously used to treat wastewater containing a set of trace organic contaminants (Tadkaew et al., 2011) similar to that used in this study for over three years. Thus, this activated sludge had been acclimatised for the treatment of trace organic contaminants. The activated sludge was centrifuged, reconstituted with Milli-Q water, centrifuged again, and finally reconstituted with the synthetic wastewater to make up a mixed liquor solution (*MLSS* concentration = 3.4 g/L) for the OMBR experiment. The OMBR was operated continuously and the reactor was refilled with the synthetic wastewater on a daily basis.

The feed solution contained each trace organic contaminant at a concentration of approximately 750 ng/L. Feed and permeate samples of 500 mL each were collected at the end of the experiment after seven days of continuous operation and were subjected immediately to solid phase extraction. Since the permeate sample was diluted with 1.2 L of the initial draw solution, the total permeate volume was measured to calculate the dilution factor, which was then used to back calculate the concentration of trace organic contaminant in the actual permeate sample. Temperature of the mixed liquor was maintained at 22.5 ± 0.1 °C. The initial pH and conductivity of the mixed liquor were 7.4 and 268.3 μS/cm (corresponding to approximately 2.5 mM of NaCl), respectively.

2.5. Analytical methods

The analysis of the model trace organics was based on a previously reported method (Vanderford and Snyder, 2006; Tadkaew et al., 2010). Analytes were extracted using solid phase extraction hydrophilic/lipophilic balance cartridges (Waters, Millford, MA, USA). All samples were spiked with a solution containing 50 ng of an isotopically labelled version of each analyte. The sample was then loaded onto the cartridges at 15 mL/min, after which the cartridges were rinsed with 5 mL of MilliQ water and dried with a stream of nitrogen for 30 min. Loaded cartridges were stored at 4 °C in sealed bags until elution and analysis.

Analytes were separated using an Agilent (Palo Alto, CA, USA) 1200 series high performance liquid chromatography (HPLC) system equipped with a 150 × 4.6 mm, 5 μm particle size, Luna C18 (2) column (Phenomenex, Torrence CA, USA). Mass spectrometry was performed using an API 4000 triple quadrupole mass spectrometer (Applied Biosystems, Foster City, CA, USA) equipped with a turbo-V ion source employed in both positive and negative electro-spray modes. Steroid hormones were analysed using an atmospheric pressure chemical ionisation method and all other compounds were analysed using an electro-spray ionisation method. For each analyte and internal standard a precursor ion and two product ions were monitored for reliable confirmation. Relative retention times of the analyte and isotopically labelled internal standard were also monitored to ensure correct identification (Vanderford and Snyder, 2006). A relative response ratio of analyte/internal standard over a 1–1000 ng concentration range was generated enabling quantification with correction for losses due to ion suppression and incomplete SPE recovery. All calibration curves had a correlation coefficient of 0.99 or better. The limit of reporting was determined using an s/n ratio of greater than 10 (Supplementary data Table S1).

Conductivity and pH were measured using an Orion 4-Star Plus pH/conductivity meter. MLSS and mixed liquor volatile suspended solid (MLVSS) contents were measured in accordance to the Standard Methods for the Examination of Water and Wastewater (Eaton et al., 2005). Total organic carbon (TOC) was analysed using a Shimadzu TOC-V_{CSH} analyzer. Mixed liquor samples taken from aerated biological reactor were centrifuged (Allegra X-12R, Beckman Coulter, USA) at 3270 g and the TOC concentration in the supernatant was measured as an indication of bioreactor performance.

3. Results and discussion

3.1. Pure water and reverse salt flux

The asymmetric membrane used in this study can be orientated in two different configurations. When the active layer (AL) is in contact with the draw solution (DS), it is called the pressure retarded osmosis (PRO) mode. When the membrane is orientated in the opposite direction and the active layer is in contact with the feed solution, it is called the forward osmosis (FO) mode. Both orientations were evaluated using Milli-Q water as the feed solution and a NaCl solution as the draw solution. As expected, the permeate flux in the PRO mode was significantly higher than that in the FO mode with the same draw solution concentration (Supplementary data Figure S3a). In addition, changes in the reverse salt flux closely resembled those of the water flux (Supplementary Data Figure S3b). The difference in permeate flux between the PRO and FO mode can be attributed to the internal concentration polarisation (ICP) phenomenon which has been previously described in detail by Elimelech and co-workers (Gray et al., 2006; McCutcheon et al., 2006). In an ideal situation, under an osmotic driving force, pure water is transported across the membrane from the feed solution to the draw solution. Therefore, this spontaneous flow of water across the membrane is directly proportional to the osmotic pressure difference across the active layer of the membrane. In the FO mode, the solute in the draw solution must penetrate the porous support layer to the interior surface of the active layer to facilitate an osmotic gradient across the active layer. However, the flow of water across the active layer into the support layer will dilute the draw solution concentration within the porous support layer. As a result, the actual osmotic pressure at the interior surface of the active layer is lower than in the bulk draw solution. This phenomenon is commonly referred to as "dilutive ICP". ICP can also arise in the PRO mode but in a different form. In the PRO mode, pure water is transported from the feed solution through the porous support layer, then the active membrane layer, and finally to the draw solution. Solutes transported into the porous supporting layer due to convection can be retained by the active layer, leading to a build up of osmotic pressure at the interior surface of the active layer. This "concentrative ICP" phenomenon is insignificant when pure (or Milli-Q) water is used as the feed solution.

The results regarding water and reverse salt flux reported above are indeed consistent with the theory of internal concentration polarisation. Because Milli-Q water was used as the feed solution in this set of experiments, ICP did not occur in the PRO mode. As a result, water flux increased linearly as the draw solution concentration increased. In the FO mode, a linear correlation between pure water flux and the draw solution concentration was also observed but only in the range between 0.5 and 1.5 M of NaCl. It is notable that the water flux did not increase as the concentration of NaCl in the draw solution increased beyond 1.5 M. It is also noteworthy that, in the FO mode, the effect of ICP on the water flux was more severe as the draw solution concentration increased. In previous investigations (Cartinella et al., 2006; Cornelissen et al.,

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2008; Achilli et al., 2009), the draw solution concentration was allowed to decrease as water diffused through the membrane from the feed solution, resulting in a gradual flux decline overtime. In this study, a conductivity control device was used to maintain a constant draw solution concentration throughout the experiment (see Section 2.4). Because Milli-Q water was used as the feed solution in these experiments, the permeate flux was always stable and independent of the operating time. The reported results indicate that the ICP could instantaneously reach a steady state condition (Fig. 1).

The reverse transport of solute from the draw solution to the feed solution is another important aspect in osmotically driven membrane processes. In good agreement with a previous study by Hancock and Cath (Hancock and Cath, 2009), the specific reverse salt flux (or the ratio of the reverse salt flux over water flux) was independent of the draw solution concentration. In the PRO and FO mode, the specific reverse flux of NaCl was 0.87 ± 0.5 and 0.79 ± 0.02 g/L, respectively, over the entire range of draw solution concentration investigated here (Supplementary Data Figure S3b). The reverse salt flux leads to an increase in salinity in the feed solution, and thus, can have a significant implication on long term operation of the OMBR process.

3.2. Osmotic membrane bioreactor operation

The OMBR experiment was performed in the PRO mode because this mode of operation was shown to result in a higher water flux than the FO mode. A mixed liquor solution (*MLSS* concentration = 3.4 g/L) was used as the feed solution and the dissolved oxygen level in the bioreactor was maintained at above 2 mg/L. Due to the osmotic pressure of the mixed liquor solution, as expected, the initial permeate flux obtained from the OMBR system was slightly lower than the values obtained when Milli-Q water was used as the feed solution (Section 3.1). There was a significant permeate flux decline within the first two days of the experiment. A stable permeate flux value of approximately 3 L/m^{2}h was observed following four days of continuous operation (Fig. 2). This low value of water flux could be attributed to the cake layer of biomass on the membrane surface which can hinder the transport of water and at the same time lead to an osmotic pressure build up on the feed side of the membrane.

Biological performance of the OMBR system was assessed by evaluating the production of biosolids and associated TOC removal by the reactor. The MLSS concentration in the reactor increased from 3.4 to 3.7 g/L over seven days of continuous operation. In addition, after seven days of operation, TOC concentration in the supernatant taken from the biological reactor was reduced from 205.0 to 27.3 mg/L, representing 87% removal solely due to biological degradation. Because membrane filtration would provide an additional removal of organic colloidal particles, the overall removal efficiency of TOC by the OMBR system can be expected to be even higher than 87%. Due to the very high NaCl concentration of the draw solution, it was not possible to accurately measure the TOC concentration of the permeate sample. Thus, the exact TOC removal efficiency of the OMBR system could not be directly determined. The small increase in MLSS content and the TOC removal efficiency of 87% by the reactor confirmed that that the system was biologically active throughout the experiment. However, it is necessary to note a significant salinity build up in the biological reactor. Conductivity of the mixed liquor solution after seven days of continuous operation was 8270 μS/cm, corresponding to approximately 4.13 g/L of NaCl (in comparison to the initial conductivity of 268 μS/cm). This build up of salinity is also consistent with a gradual decrease in the ratio of MLVSS over MLSS from 0.87 to 0.66 after seven days of operation. The decrease in the ratio of MLVSS/MLSS indicates that biological activity of the reactor may have deteriorated over the time.

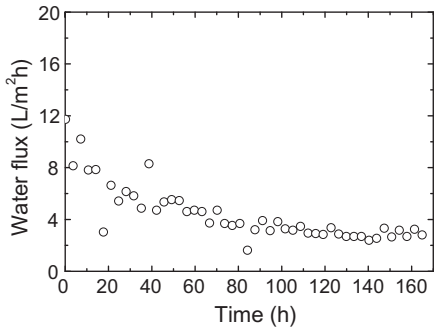

Fig. 2. Water flux as a function of operation time at different concentrations of NaCl in the draw solution. A mixed liquor containing 3.4 g/L of MLSS was used as the feed solution. The active layer of the FO membrane was placed against the draw solution (PRO mode). Cross flow velocity of both the feed and draw solution was 2.0 cm/s. Feed and draw solution was maintained at 22.5 ± 0.1 °C.

3.3. Removal of trace organics

Feed and permeate concentrations as well as the corresponding removal efficiency of 50 trace organic compounds after seven days of continuous operation are presented in Fig. 3 in order of increasing molecular weight. Data presented here clearly highlight the

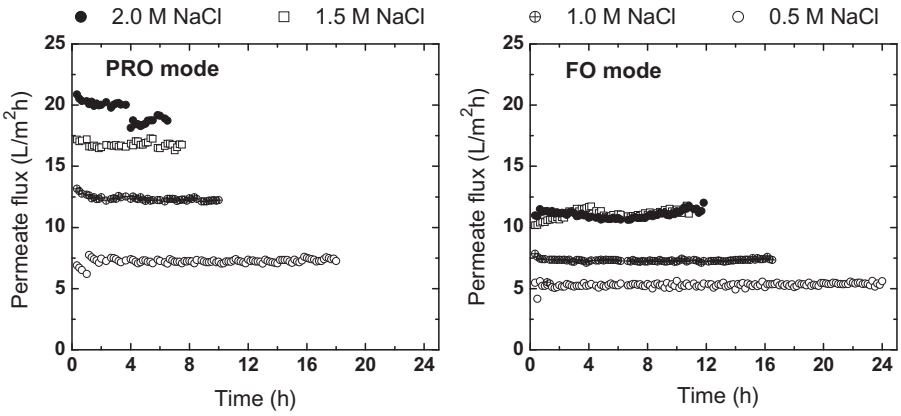

Fig. 1. Water flux as a function of operation time at different concentrations of NaCl in the draw solution. Milli-Q water was used as the feed solution. Cross flow velocity of the feed and draw solution circulation flow was 4.0 cm/s. Feed and draw solution was maintained at 22.5 ± 0.1 °C.

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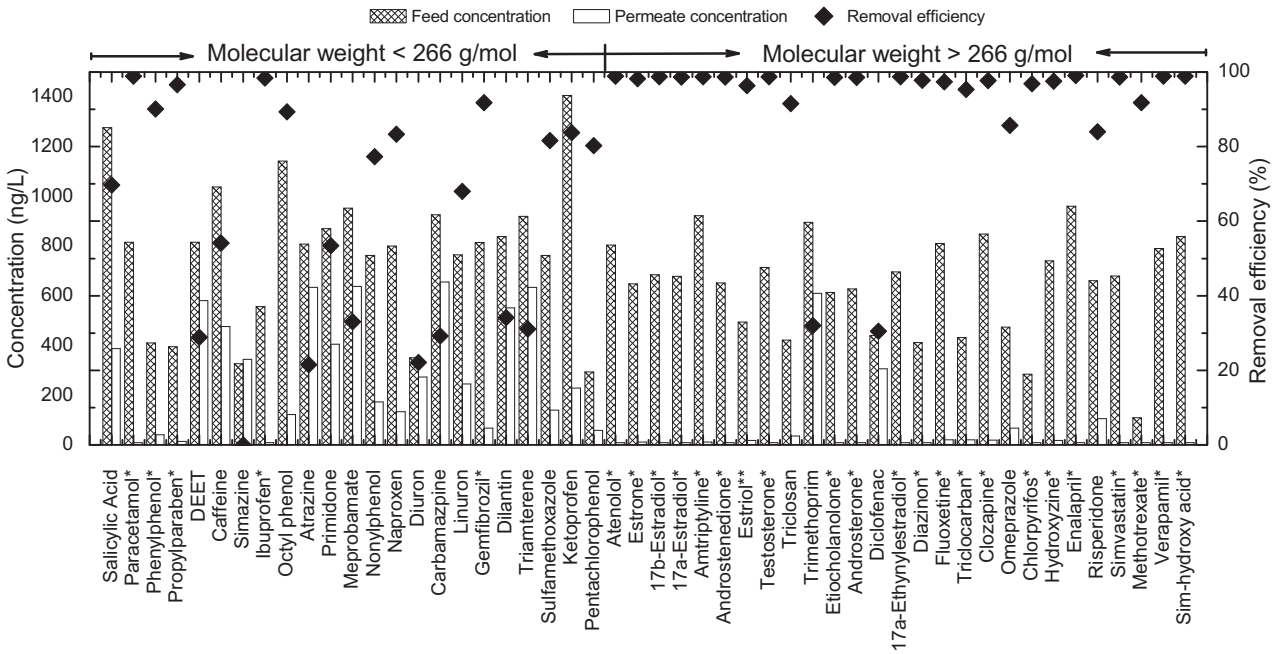

Fig. 3. Feed and permeate concentration as well as the removal efficiencies of trace organic contaminants by the OMBR system. The hydraulic retention time was approximately 80 h. The permeate sample was collected after seven days of continuous operation. Permeate concentration has been corrected for dilution due to the initial volume of draw solution. Compounds being removed to below the analytical limit were denoted with '*' and the detection limit was used to calculate the removal efficiency. Experimental conditions are as in the caption of Fig. 2.

complementarity between a high rejection membrane process and a biological treatment system. The OMBR system demonstrated excellent removal capacity for almost all compounds with molecular weight more than 266 g/mol (Fig. 3). Indeed, 22 out 27 compounds with molecular weight above 266 g/mol selected in this study were removed to below the analytical detection limit. Three other compounds (triclosan, omeprazole, and risperidone) were detected in the permeate at concentrations marginally above the analytical detection limit. Because large molecular weight compounds can be effectively retained by the FO membrane, their actual retention time in the biological reactor can be much higher than the hydraulic retention time of the system. An increase in their retention time in the reactor facilitates further biological degradation, thereby leading to high removal efficiency. Of the 27 compounds with molecular weight above 266 g/mol selected in this study, only two compounds were not effectively removed by OMBR treatment. The removal efficiencies by the OMBR system of these compounds, trimethoprim and diclofenac were 32% and 30%, respectively. These two compounds have been previously reported to be resistant to biological treatment (Hai et al., 2011; Tadkaew et al., 2011). Since these two compounds are not amenable to biological treatment, their concentration would continuously build up in the reactor over time. This would eventually lead to their permeation across the FO membrane as can be seen in Fig. 3. This hypothesis is supported by a short term experiment by Cartinella et al., 2006 who investigated the rejection of the steroid hormone estrone by FO membranes. Using a feed volume of 900 mL and a draw solution volume of 300 mL, Cartinella et al., 2006 reported a gradual decrease in estrone rejection from almost 100% to approximately 75% as the draw solution volume increased from 300 to 900 mL (corresponding to a system recovery of 70%). The authors attributed this decrease in estrone rejection to the gradual diffusion of the compound through the membrane matrix under an increasing chemical concentration gradient as the feed solution became more concentrated (Cartinella et al., 2006). The low removal efficiency of trimethoprim and diclofenac clearly indicates that physical rejection by the FO membrane alone cannot explain the high removal efficiency of most compounds with molecular weight higher than 266 g/mol as can be seen in Fig. 3. Indeed, the removal of trace organic compounds with molecular weight higher than 266 g/mol by OMBR treatment appears to be governed by the interplay between physical separation and biological degradation (Fig. 3).

The removal efficiency of trace organic compounds with molecular weight less than 266 g/mol varied from negligible removal (i.e. simazine, atrazine, and diuron) to removal to below the analytical detection limit (i.e. paracetamol, phenyl-phenol, and ibuprofen) (Fig. 3). Given their small molecular weights, physical rejection of these compounds by the FO membrane is expected to be minimal. Therefore, their removal efficiency by OMBR treatment depends mostly on biological degradability. In fact, compounds with less than 50% removal efficiency have been shown to be very resistant to biological treatment in several previous studies. These include DEET, simazine, atrazine, meprobamate, diuron, carbamazepine, dilantin, and triamterene (Clara et al., 2005; Bernhard et al., 2006; Radjenovic et al., 2007; Hai et al., 2011; Tadkaew et al., 2011). On the other hand, compounds previously reported to be very amendable to biological treatment (Clara et al., 2005; Radjenovic et al., 2007; Tadkaew et al., 2011) were removed to below the analytical detection limit. These include paracetamol, phenyl-phenol, propylparaben, ibuprofen, and gemfibrozil (Fig. 3).

4. Conclusion

The removal of 25 out of 27 trace organic compounds with molecular weight higher than 266 g/mol was high and was possibly governed by the interplay between physical separation of the FO membrane and biological degradation. On the other hand, the removal efficiency values of the other 23 trace organic compounds with molecular weight less than 266 g/mol were very scattered. The removal efficiency of these low molecular weight compounds by OMBR treatment appears to depend mostly on biological degradability. There was evidence of continuous deterioration of

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biological activity of the OMBR, possibly due to salinity build-up in the reactor.

Acknowledgement

The Saudi Arabian Cultural Mission in Australia is acknowledged for providing a PhD scholarship to Abdulhakeem Ali Alturki. Hydration Technology Innovations is thanked for the provision of membrane samples.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.biortech.2012.01.082.

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