Desalination and Water Treatment
www.deswater.com
doi: 10.1080/19443994.2015.1015174

57 (2016) 7149–7154
April

Taylor & Francis
Taylor & Francis Group

Macropollutants removal from compost leachate using membrane separation process

Hassan Hashemi[a], Yaghoub Hajizadeh[b], Mohammad Mehdi Amin[b], Bijan Bina[b], Asghar Ebrahimi[c], Abbas Khodabakhshi[d,*], Afshin Ebrahimi[b], Hamid Reza Pourzamani[b]

[a]*Health Faculty, Shiraz University of Medical Sciences, Shiraz, Iran, Tel. +98 3137922685; email: hashemi@hlth.mui.ac.ir*
[b]*Environment Research Center, Isfahan University of Medical Sciences, Isfahan, Iran, Tel. +98 3137922686; email: envrc@mui.ac.ir (H.R. Pourzamani)*
[c]*Health Faculty, Yazd University of Medical Sciences, Yazd, Iran, Tel. +98 3137922685; email: ebrahimi20007@gmail.com*
[d]*Health Faculty, Shahrekord University of Medical Sciences, Shahrekord, Iran, Tel. +98 3117922677; Fax: +98 3137922509; email: khodabakhshi16@gmail.com*

Received 9 June 2014; Accepted 29 January 2015

ABSTRACT

Advanced treatment of a biologically pre-treated compost leachate was carried out in this study. The leachate was fed to a membrane-based bioreactor and then its supernatant effluent was purified by a hollow fiber membrane (HFM) and a flat sheet membrane (FSM) separately. Macropollutants including biological oxygen demand (BOD_5), chemical oxygen demand (COD), and total suspended solids (TSS) were monitored in the reactor feed and permeate after the system reaches to a steady-state condition which lasted 280 d. The values of pH and total dissolved solids (TDS) were also measured routinely. Both the membranes were thoroughly rinsed with permeate and then immersed individually in hypochlorite solution for 30 min occasionally. Among the experiments, total COD values ranging from 50 to 2,200 mg/l in the feed were decreased to 32.5 and 4 mg/l in the HFM and FSM permeate, respectively. Average TSS content in the effluent of the HFM was 2.5 mg/l and that in the FSM effluent was 0.26 mg/l. Also, produced BOD_5 in the HFM and in the FSM effluents were 1.3 and 0.2 mg/l, respectively. TDS concentration in the membranes' feed were varied from 4,567 to 14,811 mg/l, whereas, there were no significant decreases in the membranes' permeate. The results demonstrated that the FSM was more suitable module than the HFM in terms of providing high quality permeates.

Keywords: Macropollutants; Compost leachate; Membrane; HFM; FSM

1. Introduction

Complete treatment of composting leachate due to the complexity of its composition is today's challenge. In the last 20 years, more effective treatments based on membrane technology have emerged as a viable treatment alternative to comply with water quality regulations in most countries [1]. The membrane is a barrier that retains all particles, colloids, bacteria, and viruses, providing a complete disinfection of treated water. High quality and less fluctuation of final

*Corresponding author.

effluent are the important specifications of membrane processes. This is due to the capability of membrane bioreactors to better biomass maintaining inside the bioreactor [2].

According to the literatures cited [3,4], membrane bioreactor gives high-quality treated water with reduced sludge production. However, membrane fouling and its mitigation has become a major challenge. The nature and degree of membrane fouling is influenced by membrane properties, operation conditions, and solution characteristics, which include the physicochemical properties of the biomass. A widely used method of fouling control in membrane bioreactors involves the use of additives such as inorganic coagulants or powdered activated carbon [4]. The main limitation for their widespread application is their high energy demand (between 0.45 and 0.65 kWh/m^3) for the highest optimum operation from a demonstration plant [5]. Few research works are related to the compost leachate purification by membranes. In comparison with side stream (sMBR) configuration, submerged or immersed (iMBR) one is the most widely used due to lower associated costs of operation [3]. Due to the high fouling potential and low filterability of leachates from composting plant, majority of membrane processes are utilized based on external systems. However, studies on submerged systems are currently on the rise. Feeding a submerged membrane bioreactor with a solution containing 68,000 mg/l chemical oxygen demand (COD) reduced its level to 1,733 mg/l in the effluent [6]. Refractory matters such as fulvic acid and compounds with carboxylic and aromatic hydroxyl groups, not only are difficult to be biodegraded but also can pass through the membrane, thus causing high soluble COD in the effluent. Several researchers have investigated the potential of using MBRs for leachate treatment in various applications. This study aims to investigate the removal of macropollutants from pre-treated compost leachate using hollow fiber membrane (HFM) and flat sheet membrane (FSM) separation process.

2. Materials and methods

2.1. Pilot-scale bioreactor configuration

The designed and applied pilot-scale membrane bioreactor was composed of sequencing batch reactor (SBR) and a submerged membrane. Two different types of membranes, a HFM and a FSM were used in the experiments. The specifications of the membranes are given in Table 1.

Also, configuration of the two types membranes used in the bioreactor is presented in Fig. 1.

Leachate samples were obtained from an Isfahan (Iran) municipal composting plant. After treatment by anaerobic migrating baffled reactor, anaerobic sequencing batch reactor, and SBR processes in series, the supernatant was sucked through the membrane wall with a pump. Air was diffused at the bioreactor bottom in order to provide aerobic condition. FSM fouling was controlled by coarse bubbling of air flow and by intermittent filtration of permeate. While, during backwashing of HFM, permeate was pumped in the opposite direction through the membrane. Backwashing effectively removed most of the reversible fouling, which was due to pore blocking.

2.2. Analytical methods

The values of pH (Metrohm Herisau-E520) and total dissolved solids (TDS) (HACH Sension5) were monitored routinely. However, analyses of total COD (spectrophotometer DR-5000, Model 8452A, Hatch-Lange), biological oxygen demand (BOD_5) (Oxitop bottles, WTW IS 6, Germany), and total suspended solids (TSS) (Gravimetery) were done twice a week according to the Standard Methods for the Examination of Water and Wastewater [7].

3. Results and discussion

3.1. Feed characteristics

Table 2 shows the bioreactor effluent quality used as membranes feed. It is clear that the parameters in the feeding leachate have very wide ranges.

3.2. pH variations

Variation of feed and permeate pH is shown in Fig. 2.

As illustrated, the pH values were approximately stable during operation time and measured between 7 and 8.

3.3. COD removal

Fig. 3 shows the total COD in feed and permeate vs. operating time. Total COD values in the feed were ranged 50–2,200 mg/l in different loading rate and decreased to 32.5 and 4 mg/l in HFM and FSM permeate, respectively. Researches revealed that, leachate contains non-biodegradable matters with COD range of 400–1,500 mg/l even after the biological treatment, and

Table 1
Specification of membranes used in this study

Membrane type	Material	Pore size (μm)	Effective area	Operating pressure
Hollow fiber	Polypropylene	0.01–0.2	0.1 m^2/module	−0.01 to −0.03 MPa
Flat sheet	Polyethersulfone	0.2	1 m^2/ea	−0.6 to 0 Kgf/cm^2

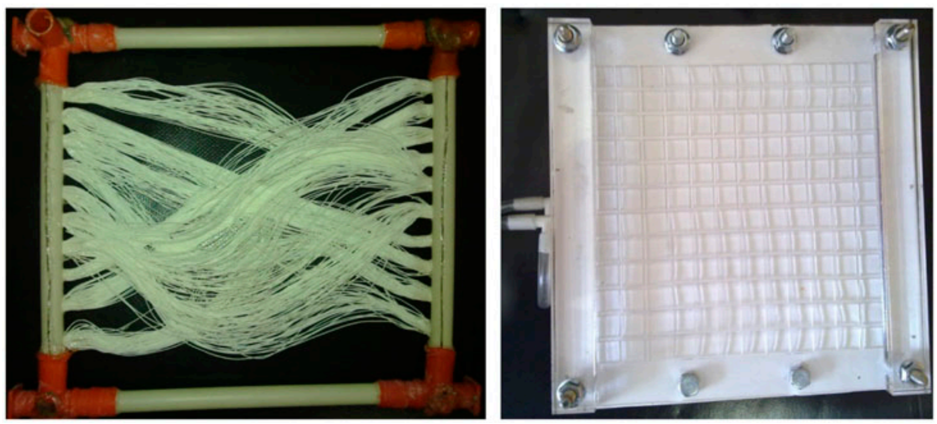

Fig. 1. View of membranes used in MSBR (left: HFM, right: Flat sheet).

Table 2
Results of the main membranes feed parameters analyzed

Parameter	Range	Mean ± SD
Total COD (mg/l)	50–2,200	654 ± 639
BOD_5	5–163	54 ± 43
BOD_5/COD	0.04–0.2	0.12 ± 0.02
TSS (mg/l)	140–18,500	84.5 ± 57
pH	7.4–8.1	7.6 ± 0.5

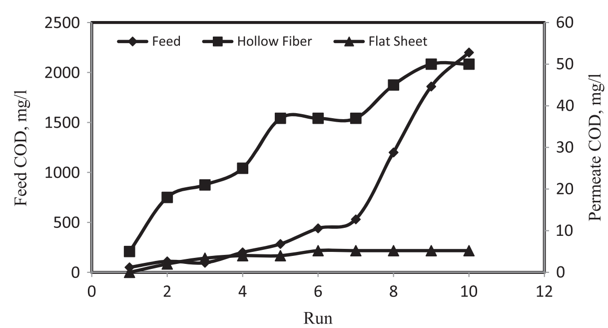

Fig. 3. Total COD concentration variations vs. time.

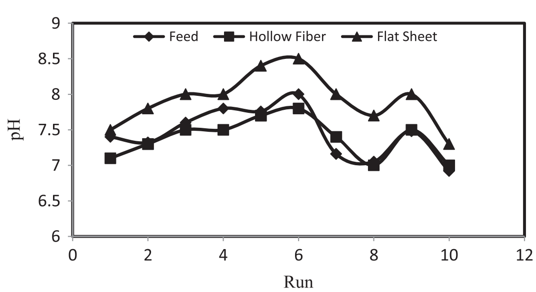

Fig. 2. pH variations in feed and membranes permeate in time.

should be removed further to attain the discharge standards [8]. Evaporation techniques are useful for separation of humic substances from leachate. The high permeate recovery rate of RO is achieved due to the significantly low salinity of the inlet from evaporation [9]. Semi-permeable membranes have lower fouling propensity than pressure driven systems and therefore, require less frequent backwashing. Microporous membranes do not remove soluble matter due to their porous nature. However, because of formation of a cake layer on their surface, it can typically hold 28–87% of soluble organic matter. The semi-permeable membrane has been shown to greater than 99% of TOC removal due to its nonporous composition [10].

With increasing loading rate, COD values were increased. However it was relatively steady in FSM permeate. According to the Independent Samples *t*-test, there was significant difference between COD values of two type membranes permeate ($p < 0.05$).

Mahmoudkhani et al. noted that the MBR process efficiencies were in the range of 95–98% in terms of TOC reduction, and reached to 97% for specific organic pollutants [6]. Overall COD in the membrane permeate was considered soluble non-biodegradable COD (snbCOD). Measured COD values were less than Iranian environmental protection organization standards (<200 mg/l), whereas other studies reported greater than this limit (1,733 mg/l) [4]. In feed, CODs of more than 350 mg/l may lead to membrane fouling and permeate was colored. Aziz et al. announced that the presence of high concentration of organic matters make yellow color permeates. Membrane could not remove color completely [11]. Significant amount of COD in the stabilized leachate indicates that the leachate contains natural organic matters such as recalcitrant matter (e.g. humic and fulvic acids), which are not easily degradable. In a similar study, Aslan et al. showed that difference between COD and TOC removals was negligible in a membrane modules designed at different forms [12]. It seems that chemical processes like advanced oxidation processes could be effective in complete removal of residual refractory organic matters for reuse purposes.

3.4. TSS removal

Considerable amounts of solids escaped from the upstream processes, especially in high loading rate that led to membrane fouling (140–18,500 mg/l). As shown in Fig. 4, average concentrations of the final effluent TSS were between 2.5 and 0.26 mg/l in the HFM and FSM, respectively. Up to 99.9% of solids were removed with micropore membrane, which mainly might be included colloidal solids.

Similarly, Arrojo et al. revealed that the removal of colloidal and suspended particles by membrane filtration is 100%, suggesting that the integrated process could produce an effluent suitable for agriculture [13]. Bae et al. achieved 5–10% additional solids removal using ultrafiltration membrane [14]. In subsequent polishing of leachate treatment, TSS removal was more than 99%. Membrane-coupled SBR results in purification of turbid SBR effluent [15]. According to these results, the membranes played a positive role in maintaining high biomass in the bioreactor, enabling a stable treatment efficiency of the reactor. This agreed with Bae et al. results [14]. A paired sample *t*-test analysis showed that TSS removal was significant in both membrane modules ($p < 0.05$). Based on one-sample *t*-test, permeate quality increased significantly down to national standard limit ($p < 0.05$).

3.5. BOD_5 removal

The standard five-day BOD value, a main indicator of the effluents strength in discharge to receiving waters, was analyzed during the examinations. As shown in Fig. 5, BOD_5 concentration in the feed was 5–163 mg/l. Significant amounts of organic matters were attributed to the suspended solids. So that, by separation of solids by the HFM and the FSMs, BOD_5 of the effluents were reduced to 1.3 and 0.2 mg/l, respectively, which are below the standard limit.

In general, based on paired *t*-test analysis, organic macropollutants residuals from biological treatment were removed significantly through membrane filtration ($p < 0.05$).

3.6. TDS removal

The salinity content of the compost leachate was high, which causes improper reflux utilization of leachate. This should be treated by a combined biochemical and physicochemical process before discharge [16]. The value of conductivity for the leachate sample was 0.87–3.98 mS/cm in 22°C which was decreased by FSM significantly ($p < 0.05$). Usually, submerged membranes used in bioreactor are micro

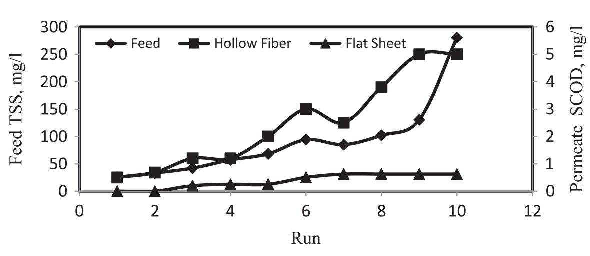

Fig. 4. Changes in concentrations of TSS in different run times.

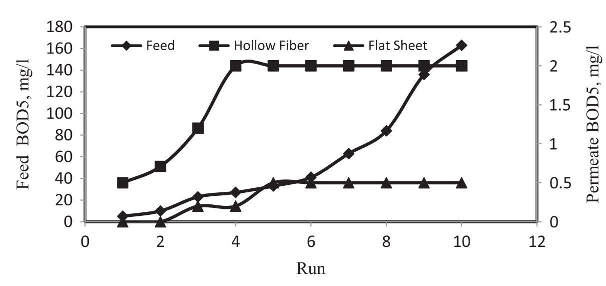

Fig. 5. Feed and permeate BOD_5 during membranes operation.

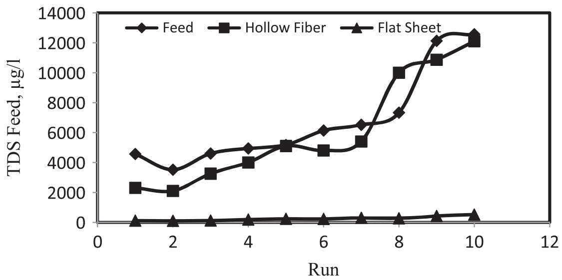

Fig. 6. Leachate TDS concentration during process operation.

or ultrafilters, which can rarely remove dissolved materials. In Clifford et al. study, TDS concentration in feed and permeate of MBR was 15,000 and 16,633 mg/l, respectively [17]. In similar study, the TDS of 37–42 g/l in influent of two-stage submerged combustion evaporation system was decreased to 0.02–0.56 g/l in condensate [18]. As shown in Fig. 6, TDS concentration in the feed was between 4,567 and 14,811 mg/l and there was no significant decrease in the HFM permeate (Efficiency of 27%). However 93% TDS reduction was achieved by the FSM.

3.7. Membrane fouling and cleaning study

The images of scanning electron microscopy showed that the FSM had a narrow pore size ranging between 0.008 and 0.009 µm. In spite of mechanical separation by membrane, dynamic layer of accumulated biomass and proteinaceous matter on the surface of the membrane enhances the separation efficiency by reducing the effective pore size. Membrane fouling occurs by the deposition of soluble and particulate materials on the membrane surface and inside its pores [19]. The feed quality plays a significant role in determining the membrane fouling. Proteinous and carbohydrate soluble microbial products ratio (EPS_P: EPS_C) analysis was performed to assay fouling in both membrane types. This ratio was increased from 2.1 to 6.4 when the SRT was increased from 5 to 15 d and then, decreased slightly to 3 when the sludge age was further increased to 20 d. In a similar study, with SRT range of 10–30 d, greatest EPS production was found at 20 d SRT. From Wei et al. research, the concentrations of soluble and bounded EPS were 288 and 146 mg/g VSS, respectively; the EPS_P: EPS_C ratio in both soluble and bounded forms were 0.81 [20]. Since, the membranes were fed by treated leachate, fouling was not occurred significantly. Consequently, permeate flux was relatively stable during operation. While, in Jung and Son study, permeates flux was rapidly declined due to simultaneous pore blocking and cake formation [21]. Membrane cleaning was an essential part of the system operation and maintenance, which significantly influenced the membranes performance. At the beginning of all the experiments, the HFM module was backwashed with permeate stream until permeate flux stabilized. In addition, before loading both the membranes into the bioreactor, they were rinsed thoroughly with permeate and then, immersed individually in 200 ppm sodium hypochlorite as a cleaning solution, for 30 min.

4. Conclusions

Nowadays, there are still some problems and challenges depending on the characteristics of compost leachate for biological treatment processes, which are hardly efficient for removal of pollutants. Hence, membrane processes have been widely used as post-treatment of biologically pre-treated leachate. The results of this study demonstrated that the FSM is more suitable module than the HFM in terms of providing high quality permeate. As we used membranes for post-treatment of leachate, fouling was not notable. Consequently, permeate flux was not declined during operation. Applying an advanced treatment process after membranes could be very effective in the removal of refractory organic matters.

Acknowledgments

We are gratefully acknowledging the Waste Management Organization of Isfahan for their help in the collection of leachate samples.

References

[1] S. Renou, J.G. Givaudan, S. Poulain, F. Dirassouyan, P. Moulin, Landfill leachate treatment: Review and opportunity, J. Hazard. Mater. 150 (2008) 468–493.
[2] F.N. Ahmed, C.Q. Lan, Treatment of landfill leachate using membrane bioreactors: A review, Desalination 287 (2012) 42–54.
[3] P. Le-Clech, B. Jefferson, S.J. Judd, A comparison of submerged and side stream tubular membrane bioreactor configurations, Desalination 173 (2005) 113–122.
[4] Y. Satyawali, M. Balakrishnan, Effect of PAC addition on sludge properties in an MBR treating high strength wastewater, Water Res. 43 (2009) 1577–1588.
[5] W. Garces, C. Wilde, G. Gueldre, Operational cost of MBR schilde, Proceedings of the 4th IWA International Membranes Conference, Membranes for Water and Wastewater Treatment, May 2007, Harrogate, UK, pp. 15–17.
[6] R. Mahmoudkhani, A.H. Hassani, A. Torabian, S.M. Borghei, Study on high-strength anaerobic landfill leachate treatability by membrane bioreactor coupled with reverse osmosis, Int. J. Environ. Res. 6 (2012) 129–138.

[7] American Public Health Association, American Water Works Association, Water Environment Federation, Standard Methods for the Examination of Water and Wastewater, twenty-first ed., Port City Press, Baltimore, MD, 2005.
[8] L. Ziyang, S. Yu, C. Xiaoliang, Z. Youcai, Z. Nanwen, Application of hydration reaction on the removal of recalcitrant contaminants in leachate after biological treatment, Waste Manage. 34 (2014) 791–797.
[9] D. Yue, Y. Xu, R.B. Mahar, F. Liu, Y. Nie, Laboratory-scale experiments applied to the design of a two-stage submerged combustion evaporation system, Waste Manage. 27 (2007) 704–710.
[10] A. Achilli, T.Y. Cath, A.E. March, A.E. Childress, The forward osmosis membrane bioreactor: A low fouling alternative to MBR processes, Desalination 239 (2009) 10–21.
[11] S.Q. Aziz, H.A. Aziz, M.S. Yusoff, M.J.K. Bashir, M. Umar, Leachate characterization in semi-aerobic and anaerobic sanitary landfills: A comparative study, J. Environ. Manage. 91 (2010) 2608–2614.
[12] M. Aslan, Y. Saatci, O. Hanay, H. Hasar, Membrane fouling control in anaerobic submerged membrane bioreactor, Desalin. Water Treat. (2013) 7520–7530.
[13] B. Arrojo, A. Mosquera-Corra, J.M. Garrid, R. Mdndez, E. Ficara, F. Malpei, A membrane coupled to a sequencing batch reactor for water reuse and removal of coliform bacteria, Desalination 179 (2005) 109–116.
[14] J.H. Bae, E.Y. Lee, A.H. Heo, H.K. Kim, J.H. Kim, S.K. Park, Treatment of garbage leachate with a pilot scale two-phase anaerobic digestion with ultra filtration, Proceedings Venice, Third International Symposium on Energy from Biomass and Waste, Venice, Italy, 2010, pp. 8–11.
[15] J. Tsilogeorgis, A. Zouboulis, P. Samaras, D. Zamboulis, Application of a membrane sequencing batch reactor for landfill leachate treatment, Desalination 221 (2008) 483–493.
[16] G. Li, W. Wang, Q. Du, Applicability of nanofiltration for the advanced treatment of landfill leachate, J. Appl. Polym. Sci. 116 (2010) 2343–2347.
[17] S. Clifford, W.J. Patoczka, J. Williams, T.G. Grau, MBR pretreatment of landfill leachate for the removal of ammonia and potential future removal of total dissolved solids, in: WEFTEC, Water Environment Federation, Alexandria, VA, 2010, pp. 4064–4075.
[18] K. Farahbakhsh, D.W. Smith, Removal of coliphages in secondary effluent by microfiltration—Mechanisms of removal and impact of operating parameters, Water Res. 38 (2004) 585–592.
[19] A. Vargas, I. Moreno-Andrade, G. Buitron, Controlled backwashing in a membrane sequencing batch reactor used for toxic wastewater treatment, J. Membr. Sci. 320 (2008) 185–190.
[20] X. Wei, Z. Wang, F. Fan, J. Wang, S. Wang, Advanced treatment of a complex pharmaceutical wastewater by nanofiltration: Membrane foulant identification and cleaning, Desalination 251 (2010) 167–175.
[21] C.W. Jung, H.J. Son, Evaluation of membrane fouling mechanism in various membrane pretreatment processes, Desalin. Water Treat. 2 (2009) 195–202.