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Enhancing forward osmosis water recovery from landfill leachate by desalinating brine and recovering ammonia in a microbial desalination cell

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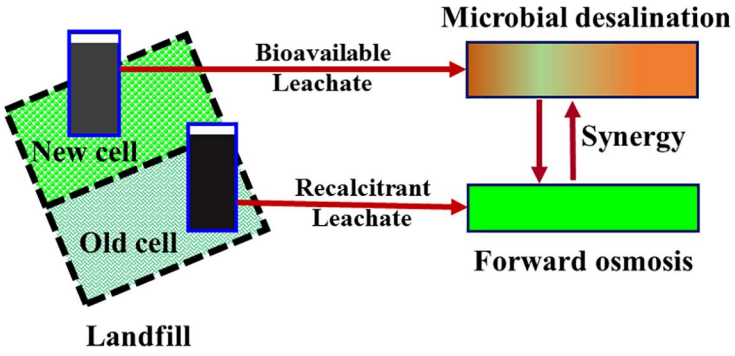
GRAPHICAL ABSTRACT



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ABSTRACT

In this work, a microbial desalination cell (MDC) was employed to desalinate the FO treated leachate for reduction of both salinity and chemical oxygen demand (COD). The FO recovered 51.5% water from a raw leachate and the recovery increased to 83.5% from the concentrated leachate after desalination in the MDC fed with either acetate or another leachate as an electron source and at a different hydraulic retention time (HRT). Easily-degraded substrate like acetate and a long HRT resulted in a low conductivity desalinated effluent. Ammonia was also recovered in the MDC cathode with a recovery efficiency varying from 11 to 64%, affected by current generation and HRT. Significant COD reduction, as high as 65.4%, was observed in the desalination chamber and attributed to the decrease of both organic and inorganic compounds via diffusion and electricity-driven movement.

1. Introduction

Landfill leachate is a complex wastewater that contains dissolved organic substances, inorganic macro components (i.e. Na^+, NH_4^+, K^+, Mg^{2+}, Cl^-, PO_4^{3-}, SO_4^{2-}), heavy metals (i.e. Fe, Hg, Zn, Cu, Cd, Pb), xenobiotics (i.e. aromatics), and a substantial amount of water (Christensen et al., 1998; Jensen & Christensen, 1999; Kjeldsen et al., 2002). Thus, it must be properly treated to reduce its impact on the environment, and various treatment methods have been employed or investigated, including biological, physicochemical, and electrochemical treatments (Renou et al., 2008). These processes can be effective in reducing the concentrations of contaminants to a certain degree, but in general, leachate management is very challenging (Bohdziewicz et al., 2001; Welander et al., 1998). In addition to contaminant removal, reducing the volume of leachate could also help with its treatment and management, for example, decreasing the use of

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external chemicals or the size of the treatment systems. Volume reduction could also recover high quality water that may be directly discharged or used for other purposes. Membrane technologies, especially pressure driven filtrations (e.g. ultra, micro, nano, reverse osmosis), have been employed for leachate treatment to separate contaminants from the final effluent and because of the need for external pressure, those membrane treatments are usually energy intensive (Chianese et al., 1999; Di Palma et al., 2002; Renou et al., 2008).

Forward osmosis (FO) is an emerging separation technology that uses an osmotic gradient between a draw solution and a feed solution to drive water molecules through the semipermeable membrane (Cath et al., 2006). FO has been investigated for water recovery from sea water, municipal wastewater, and landfill leachate with simultaneous volume reduction, and can reject contaminants such as heavy metals, organics, ammonia nitrogen, etc. (Elimelech & Phillip, 2011; Iskander et al., 2017; Linares et al., 2014). In the absence of an external pressure, FO treatment is potentially energy efficient. For example, a recent study reported an energy consumption of 0.005 kWh m^{-3} for treating landfill leachate with 30 mL min^{-1} recirculation and a 3-M NaCl draw (Iskander et al., 2017). In a submerged FO system, 621.5 mL water was recovered from 1700 mL leachate in 59 h by using the 4-M NaCl draw (Wu et al., 2018). The use of a 2-M NH_4HCO_3 draw solution achieved a higher water recovery of 51% with pretreatment of leachate by a microbial electrolysis cell that decreased the conductivity of leachate and thus benefited FO treatment (Qin et al., 2016). The FO treatment could effectively separate polycyclic aromatic hydrocarbons in the leachate from the final effluent, which may be used for direct fertigation when using NH_4HCO_3 as a draw (Li et al., 2017). FO has also been incorporated with a membrane distillation (MD) unit for recovering water and removing contaminants (i.e. TOC, NH_4^+-N) from landfill leachate (Zhou et al., 2017).

During FO treatment, the feed (leachate) becomes highly concentrated and this brine creates more resistance for the water flux that is controlled by the salinity gradient across an FO membrane. For example, it was found that the water flux decreased from 4.5 LMH to 1.5 LMH after 12 h treatment or from 2.9 LMH to 0.1 LMH after 59 h treatment, which was related to the conductivity increase of the feed side with time (Iskander et al., 2017; Wu et al., 2018). In addition, because brine has a high concentration of dissolved solids, inorganic fouling may be promoted and subsequently affect the FO membrane (Afrasiabi & Shahbazali, 2011). To minimize the volume of the treated leachate using FO technology, the brine should be treated to facilitate further water extraction. Proper brine management is both environmentally and economically important (Pramanik et al., 2017). Conventional processes of brine management include deep well injection, land application, evaporation ponds, conventional crystallizers, and landfilling, and the advanced processes such as electrodialysis, membrane distillation, and capacitive deionization are still under development (Afrasiabi & Shahbazali, 2011; Pramanik et al., 2017).

In this study, a microbial desalination cell (MDC) was proposed to act as a brine control unit to help with FO water extraction from landfill leachate. MDCs are bioelectrochemical systems that use bioelectricity to accomplish desalination (Cao et al., 2009; Kim & Logan, 2013; Sevda et al., 2015). In the proposed system, the brine from the FO treatment was desalinated in the MDC, driven by electricity generation from organic oxidation (e.g., the waste organic matter), and thus the desalinated brine could be further treated by FO for water extraction. Meanwhile, cation movement in the MDC would help to recover valuable compounds such as ammonia in its cathode. This is different from a prior study of FO-MDC system, in which the MDC was used to treat the diluted draw solution from the FO (Yuan et al., 2015). The specific objectives of this study were to: (1) demonstrate the feasibility of the proposed system for enhancing water recovery in the FO; (2) examine the effects of organic loading rates in the anode and salt loading rates in the desalination chamber on the performance of the FO – MDC system; and (3) investigate the recovery of ammonia in the MDC.

Table 1
Chemical properties of the two leachates. Leachate A was used for water recovery and desalination. Leachate B was used as an anode substrate in the MDC.

Parameters	Leachate A	Leachate B
pH	8.05 ± 0.02	7.81 ± 0.03
Conductivity, mS cm^{-1}	31.11 ± 0.01	27.1 ± 0.1
COD, mg L^{-1}	11,650 ± 109	4740 ± 30
TOC, mg L^{-1}	3353 ± 89	1771 ± 44
BOD_5/COD	0.07	0.19
NH_3-N, mg L^{-1}	2000 ± 80	1897 ± 6
Na^+, mg L^{-1}	5691 ± 81	3837 ± 45
Cl^-, mg L^{-1}	4923 ± 78	5273 ± 67
K^+, mg L^{-1}	1771 ± 98	993 ± 54
Mg^{2+}, mg L^{-1}	141 ± 12	219 ± 29
Ca^{2+}, mg L^{-1}	54 ± 11	59 ± 11

2. Materials and methods

2.1. Leachate

Two types of leachates were studied, Leachate A and Leachate B. Leachate A was low in biodegradability (BOD_5/COD = 0.07), while Leachate B (BOD_5/COD = 0.19) was higher. Hence, physicochemical treatment would be more effective for Leachate A, while biological treatment was chosen for Leachate B. For this reason, Leachate A was used in the FO treatment followed by the MDC desalination, while leachate B was used in the later stage of the study for electricity generation in the MDC anode. The leachates were collected from two different cells of a landfill in Virginia, USA. After collection, leachates were stored at a 4 °C temperature until use. The chemical properties of the leachates are given in Table 1.

2.2. Forward osmosis – Microbial desalination cell system setup and operation

The FO-MDC system is shown in Fig. 1. Leachate A was used for the main experiment in the FO for water recovery and in the MDC for brine desalination. The FO unit was operated in a batch mode for 10 h with the initial feed (leachate) volume of 500 mL and a draw solution volume of 200 mL containing 3-M NaCl. Then, the concentrated leachate feed was fed into the desalination chamber of the MDC for desalination. A SEPA CF Cell (Sterlitech Corporation, Kent, WA, USA) was used as the FO unit with an Aquaporin embedded flat-sheet FO membrane (Aquaporin A/S, Lyngby, Denmark) that had a surface area of 139 cm^2. The active layer of FO membrane was facing the feed (leachate), while the support layer was in contact with the draw solution. Both draw and feed solutions were recirculated at 40 mL min^{-1}, respectively. A tubular MDC was constructed according to a previous study (Jacobson et al., 2011). Ion exchange membranes (Membranes International, Inc., Ringwood, NJ, USA) were used to create an anode chamber of 330 mL and a desalination chamber of 110 mL. The membrane tubes were installed in a plastic tube which provided a cathode chamber of 750 mL with continuous aeration of 60 mL min^{-1}. A 50-cm carbon brush (Gordon Brush Mfg. Co., Inc.) was used as the anode electrode, while a piece of 450-cm^2 carbon cloth (coated with 5 mg cm^{-2} activated carbon) was used as the cathode electrode. The anode and cathode electrodes were connected across a 1-Ω external resistor for high current generation. To start the MDC, its anode was inoculated with anaerobic sludge from a local wastewater treatment plant (Christiansburg, VA). Ammonia that migrated from the desalination chamber into the cathode chamber was stripped out of the catholyte by aeration and collected in a 1-M sulfuric acid solution.

Four operating conditions were studied for treating Leachate A in both FO and MDC (Table 2), with C1 and C2 using 5 g L^{-1} sodium acetate in the MDC anode, and C3 and C4 using Leachate B as the MDC anode substrate. The use of synthetic organic compounds in C1 and C2