



Ozonation pretreatment for stabilized landfill leachate high-pressure membrane treatment



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HIGHLIGHTS

- Ozone pretreated stabilized leachate was treated using high pressure membranes.
- Ozone reduced 78% UV-254 and 23% dissolved organic matter from stabilized leachate.
- Ozone pretreatment did not improve permeate flux as compared to raw leachate.
- The flux decline analysis did not show a distinct development of fouling stages.

ARTICLE INFO

Article history:

Received 19 March 2013

Received in revised form 4 March 2014

Accepted 7 March 2014

Available online 14 April 2014

Keywords:

Landfill

Leachate

Leachate treatment

Ozone

Membrane treatment

ABSTRACT

Laboratory-scale experiments were conducted to determine the effectiveness of ozonation as a pretreatment option for treating stabilized landfill leachate using reverse osmosis and nanofiltration membranes. Leachate from three different landfills was collected and characterized as stabilized leachate with a ratio of biochemical oxygen demand to chemical oxygen demand in the range of 0.02 to 0.12. Batch ozonation experiments were conducted by using an ozonation dose of 66.7 g m^{-3} for duration of 5 to 30 min. A maximum reduction of 78% UV-254 absorbing compounds and 23% dissolved organic carbon was observed for all three leachates. An effective ozonation time of 10 min at an ozone dose of 66.7 g m^{-3} was selected for leachate pretreatment before membrane operation. Even though the ozone-treated leachate contained approximately 50% fewer UV-254 absorbing compounds than raw leachate, ozonation of leachate prior to membrane treatment resulted in a greater reduction in permeate flux as compared to raw leachate. The cause of increased permeate flux reduction with the ozonated leachate was assessed by using a flux decline analysis alongside Hermia's crossflow filtration model. Hermia's crossflow filtration model indicated that each fouling mechanism occurred. The flux decline analysis suggested that distinct stages of fouling did not develop. While the cause of the increased fouling is suspected to be the coagulation of calcium and natural organic matter, further investigation is needed to determine the primary source of fouling.

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1. Introduction

Landfill leachate's complex chemical makeup, changing characteristics over time, and variable production rate make its management one of the fundamental challenges for landfill operators [1]. Multiple factors influence leachate characteristics (e.g., landfill age, waste type, rainfall volume, operation practices); however, no factor impacts leachate quality from a treatment perspective as much as the state of landfill stabilization. Leachate generated from waste in the preliminary stages of biological decomposition contains mostly organic matter (OM) biodegradable in nature and is thus amenable to standard biological wastewater treatment processes. However, once waste decomposition reaches the stable methanogenic phase, landfill leachate OM is dominated by biologically refractory OM, reducing the effectiveness of

biological treatment techniques and necessitating other physico-chemical processes. Leachate treatability is often evaluated as a function of landfill age [2], or by the ratio of BOD_5 (biochemical oxygen demand) to COD (chemical oxygen demand), with values close to 1 representative of young leachate and values of 0.1 or less described as stabilized leachate [3].

Physico-chemical treatment methods commonly used for stabilized leachate treatment include coagulation, oxidation, adsorption, and membrane systems [4,5]. Many investigations report stabilized leachate treatment using single or multiple treatment methods. However, meeting stringent discharge standards required by many jurisdictions remains a challenge as most treatment techniques are selective for particular chemical classes [6–9]. Leachate treatment is typically focused upon the removal of COD, BOD_5 , and ammonium [4]. For stabilized leachate to meet effluent discharge standards, many landfills utilize high-pressure membrane technologies, such as nanofiltration (NF) and reverse osmosis (RO), to remove a broad array of leachate

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constituents [10]. Membrane systems by themselves may not be economically competitive as the fouling of membrane surfaces during leachate treatment requires more frequent membrane cleaning and replacement, as well as higher operational pressures. Membrane fouling is influenced by membrane surface properties, turbulence in the membrane vessel, and the feed solution chemistry, including pH, calcium content, and the presence of hydrophobic humic- and fulvic-like OM [11–13]. Since stabilized leachate contains high molecular weight organic compounds, pretreatment of leachate to reduce humic- and fulvic-like OM is one strategy to reduce fouling and increase membrane life [9,14,15].

Ozone (O_3) has a high reactivity with humic- and fulvic-like OM and has been effectively applied to reduce membrane fouling during surface and groundwater treatment [14–17]. When O_3 comes in contact with this primarily aromatic OM, it ruptures the unsaturated bonds or aromatic rings and produces alcohols, carbonyl, and carboxyl compounds [18,19]. At high pH conditions ($pH > 8$), O_3 produces hydroxyl radicals, which accelerate the oxidation of OM from the complex wastewater matrix. Although O_3 has the potential to completely oxidize most OM, its utility as a complete leachate OM treatment step is limited due to the high doses required and the necessary reaction time [7].

Table S1 in the supplementary material summarizes previously reported leachate treatment research using ozonation. COD reductions of 10 to 75% from stabilized leachate have been reported for O_3 doses in the range of 6.3 g m^{-3} to $12.5 \times 10^3 \text{ g m}^{-3}$ at various ozonation durations [6,20–25]. While several researchers have examined the use of O_3 as a pretreatment step for stabilized leachate treatment using methods such as biological treatment, adsorption, and coagulation [20, 21,24], O_3 as a pretreatment for high-pressure membrane treatment of stabilized leachate has not been reported.

The objective of the research presented here was to examine the effectiveness of ozonation as a pretreatment step for stabilized landfill leachate treatment using RO and NF membranes. In the first phase of the experiment, an effective O_3 dose for leachate pretreatment was assessed by using leachate from three different landfills. In the second phase, ozonated leachate was used as a feed solution for NF and RO membrane treatment, and time-dependent permeate flux and permeate quality were measured (and compared to treatment using raw leachate). The type of fouling that occurs during the treatment of both raw and ozonated leachate by NF and RO membranes was assessed by using Hermia's constant pressure, crossflow filtration model [26] along with a flux decline analysis [27].

2. Experimental material methods and analysis

2.1. Materials

Leachate samples were collected from three municipal solid waste landfills in Florida, USA: the Alachua County Southwest Landfill (ACL), the Polk County North Central Landfill (NCL), and the New River Regional Landfill (NRL). All three landfills have been fully or partially operated as bioreactor landfills, where leachate was added into the waste to accelerate the waste degradation rate. Leachate samples were collected in Nalgene containers and kept at 4°C in the dark until used in each experiment.

Ozonation experiments were carried out in a laboratory-scale, semi-batch bubble reactor (Fig. S1 in the supplementary material) consisting of a Plexiglas column with a height of 45 cm and an internal diameter of 10 cm, having a total volume of 3500 mL. O_3 was produced by using high purity oxygen (O_2) as a feed gas to the laboratory-scale O_3 generator (Pacific lab series O_3 generator, Model: L20) and supplied to the ozonation column through a ceramic porous diffuser of porosity 10 to 15 μm from the bottom of the reactor. O_3 concentration in the column inlet and outlet gas stream was measured by using a gas phase digital O_3 analyzer (T-API Model 452). The residual gas stream was passed through an O_3 destruction unit.

Flat-sheet RO (BW-30) and NF (NF-90) membranes were obtained from DOW FILMTEC (Minneapolis, MN) to conduct the membrane performance studies. The physical and operational characteristics of these membranes are presented in Table S2 in the supplementary material. The flat-sheet membranes were cut into $14.6 \text{ cm} \times 9.5 \text{ cm}$ coupons, stored dry in the dark, and soaked in deionized water for 24 h before use.

Membrane filtration experiments were conducted by using a high-pressure crossflow Osmonics SEPA CF Membrane Cell (Fig. S2 in the supplementary material). The membrane coupons were placed into the membrane cell sandwiched between a feed spacer of thickness 0.86 mm and a permeate carrier of thickness 0.2 mm. The feed solution was pumped by using a variable speed Hydra-Cell industrial pump from a 15 L Nalgene feed tank. A recirculation water bath (RTE-5B, Neslab Instruments Inc.) was used to control the feed water temperature. The crossflow velocity and the operating pressure were controlled by the valves connected to the concentrate return line and the bypass line and were monitored by a flow meter and a pressure gauge, respectively. A hydraulic hand pump (Enerpac, P142) was used to pressurize membrane cells and the pressure was maintained constant ($2.06 \times 10^6 \text{ N m}^{-2}$) throughout the experiment.

2.2. Landfill leachate pretreatment using ozonation

The O_3 generator was turned on approximately 20 min prior to the start of each experiment to stabilize the rate of O_3 generation by the instrument. The concentration of O_3 in the produced O_3/O_2 gas mixture was controlled by the feed gas pressure and the applied voltage. Before starting the experiments, leachate was brought to room temperature, approximately 23°C . Leachate samples were shaken to resuspend settled solids to avoid changing leachate characteristics (removing settled solids or prefiltration of leachate could possibly reduce the organic loading of leachate). To determine the optimum ozonation time for the selected O_3 dose, 1000 mL leachate samples were added to the ozonation column and ozonated for 5, 10, 15, and 30 min at a constant O_3 dose of $66.7 \pm 0.3 \text{ g m}^{-3}$ ($13.8 \times 10^{-4} \text{ mol L}^{-1}$) and a feed gas flow rate of 3.5 L min^{-1} . At the end of each experiment, samples were withdrawn from the column and analyzed for pH, dissolved organic carbon (DOC), ultraviolet-254 (UV-254) absorbance, and dissolved organic matter (DOM) characterization. All experiments were conducted in duplicate. The amount of O_3 consumed during leachate ozonation was calculated by subtracting the off-gas O_3 concentration from the O_3 concentration in the feed gas.

2.3. Landfill leachate treatment using membranes

Raw and ozonated leachate were treated by RO and NF membranes to study the membranes' performance. The experiments were conducted in two steps as described by Tang et al. [28]. In the first step, membranes were pre-compacted with deionized water for 48 h at a constant pressure ($1.4 \times 10^6 \text{ N m}^{-2}$), crossflow velocity (0.2 m s^{-1}), and temperature ($23 \pm 1^\circ\text{C}$), until a constant permeate flux was achieved. The second step involved pre-compaction of the membranes by using a 10 mMol NaCl solution. After the pre-compaction run, the membrane system was stopped and deionized water was drained from the feed tank. Approximately 8 to 10 L of feed leachate was filtered by using a $0.7 \mu\text{m}$ glass-fiber filter and added to the feed tank. Raw and ozonated leachate were used separately as feed leachate. The membrane system was then restarted and operated at the same operating conditions as in the pre-compaction run for 24 h [28]. Feed pressure, crossflow velocity, and temperature were maintained constant throughout the experiment. The permeate flux was determined intermittently by measuring the time taken to reach a predetermined volume of permeate. Concentrate and permeate were recycled to the feed tank, except for the samples collected intermittently to measure pH, conductivity, DOC, and UV-254 absorbance.

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