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Research article

Landfill leachate treatment by sequential membrane bioreactor and electro-oxidation processes

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Combination of high performance membrane bioreactor (MBR) equipped with ultrafiltration and electro-oxidation process (EOP) by boron-doped diamond electrode (BDD) was used to effectively treat highly contaminated old landfill leachate. MBR and EOP were optimized for raw and pretreated landfill leachate. Seasonal changes dramatically affected the both processes' performance, as the landfill leachate was ¾ more concentrated in winter. For MBR, organic load rate of 1.2 gCOD/L/day and sludge retention time of 80 days was considered as the optimum operating condition in which COD, TOC, NH_4^+ and phosphorous removal efficiencies reached the average of 63, 35, 98 and 52%, respectively. The best performance of EOP was in current intensity of 3 A with treatment of time of 120 min. Effluent of electro-oxidation was more toxic due to the presence of radicals and organochlorinated compounds. These compounds were removed by stripping or assimilation of sludge if EOP was used as a pretreatment method. Furthermore, the energy consumption of EOP was decreased from 22 to 16 KWh/m^3 for biologically treated and raw landfill leachate, respectively.

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1. Introduction

Among prevalent high quantity wastewater, landfill leachate (LFL) is undoubtedly one of the most challenging wastewaters in terms of treatment (Gotvajn et al., 2009; Urtiaga et al., 2009; Zhou et al., 2016), due to several reasons. Firstly, flow rate of LFL largely varied by season and age of landfill. Secondly, its characteristic widely depended on years of operation, amount of precipitation, and type of landfilling. Finally, it has high concentration of ammonia and toxicity, low (biochemical oxygen demand/chemical oxygen demand (BOD/COD) ratio, and presence of heavy metals and emerging contaminants (Kjeldsen et al., 2002). As LFL gets older, the more complex dissolved organic matter (DOM) are produced out of simple ones, dramatically decreasing the COD removal efficiency of biological treatment; hence, physio-chemical processes installation seems to be inevitable for proper elimination of recalcitrant DOM (Aloui et al., 2009; Bashir et al., 2013; Gotvajn et al., 2009). Intense brown coloration of old landfill leachate indicates the presence of these DOM with high molecular weight, known as humic substances (HS), which act as the best media for adsorption of metal and emerging contaminants (Panizza and Martinez-Huitle, 2013). Furthermore, LFL has the highest detection rate and concentration of variety of emerging contaminants among wastewaters (Oturan et al., 2015).

Conventional biological processes are expectedly inefficient for treatment of LFL because of low BOD, and phosphorous concentration, while high concentration of ammonia and emerging contaminant. Membrane bioreactors (MBR), therefore, are widely used in LFL treatment plant, due to high performance in ammonia removal by biological nitrification or air stripping (Ahmed and Lan, 2012; Gotvajn et al., 2009), metal and emerging contaminant by sludge adsorption, and finally turbidity and suspended solid by the membrane (Ahmed and Lan, 2012). The concentrated effluent of MBR contains refractory compounds, mainly HS, with low amount of ammonia and metal which is ideal for advanced oxidation process (AOP) (Schwarzenbeck et al., 2004), especially electro-oxidation process (EOP) (Urtiaga et al., 2009; Zhou et al., 2016). The main objective of EOP utilization is either removal or transformation of humic substances into biodegradable organic matter (Panizza and Martinez-Huitle, 2013), or maximum elimination of

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List of symbols

AOP	Advanced Oxidation Processes
BDD	Boron-doped diamond
BOD	Biochemical Oxygen Demand
CCD	Central Composite Design
COD	Chemical Oxygen Demand
DEHP	Di 2-ethyl hexyl phthalate
DOM	Dissolved Organic Matter
EC	Electrical Conductivity
EOP	Electro-oxidation Process
FD	Factorial Design
F/M ratio	Food/Microorganism Ratio
HA	Humic Acid
HRT	Hydraulic Retention Time
HS	Humic Substances
LFL	Landfill Leachate
MBR	Membrane Bioreactor
MLSS	Mixed Liquor Suspended Solid
MLVSS	Mixed Liquor Volatile Suspended Solid
SRT	Sludge Retention Time
TA	Total Alkalinity
TCU	True Color Unit
TS	Total Solid
VS	Volatile Solid

COD when it is used as a post treatment. COD in EOP was mainly removed by direct oxidation (oxidation on the surface of electrode or by hydroxyl radical), while ammonia was removed by means of indirect oxidation (reaction with chlorine oxidant) (reaction in supplementary files: appendix A) (Bashir et al., 2009; Chu et al., 2015; Deng and Englehardt, 2007; Panizza and Martinez-Huitle, 2013).

Sludge management of LFL treatment plant was also a concern, due to high concentration of pollutants; hence, low quantity or zero production of sludge will be favorable that could be achieved by simultaneous utilization of MBR and EOP (Zhou et al., 2016). Both processes are well-known in the formation of low quantity of solid residue and removal of emerging contaminants (Fernandes et al., 2012).

Till this date, all the studies have investigated EOP as post-treatment of biological treated landfill leachate (Aloui et al., 2009; Bashir et al., 2013; Chu et al., 2015; Fernandes et al., 2012; Urtiaga et al., 2009), though production of organochlorine compounds and remaining radicals are the main problems. In this study, combination of MBR and EOP was used in the optimal sequence to effectively treat old landfill leachate. Firstly in MBR, sludge was developed and optimized according to various operating conditions and seasonal changes. Secondly, factorial and central composite design models were used to estimate the optimal conditions for oxidation performance. Finally, the optimized sequence of two processes was determined based on best removal efficiency, lowest energy consumption and toxicity. The fate of Di 2-ethyl hexyl phthalate (DEHP) was also investigated as the model of hydrophobic emerging contaminants in landfill leachate.

2. Material and methods

2.1. Landfill leachate sampling

Municipal landfill leachate employed in this study, was collected from Frampton's landfill, (Québec, Canada) with capacity of 180 tons/day and annual average production of 100 m^3/day landfill leachate. The landfill was located around 60 km to south east of Quebec City, received waste mainly from Levis household agglomeration, Québec, Canada. Samples were monthly taken from 2750 m^3 storage tank and before aeration pretreatment. All samples were instantly stored at 4 °C before applying to MBR or EOP. The flow rate and characteristic of LFL varied from season to season, despite the fact that the high precipitation was evenly distributed throughout the year. In spring (April and May), precipitation and melting of snow results in higher production of diluted landfill leachate. The precipitation in winter (between December to March) is in the form of snow. Furthermore, freezing temperature (Fig. 1-S), dramatically decreases LFL production. As a consequence, the characteristic of raw landfill leachate fluctuated from being heavily loaded in winter to highly diluted throughout spring and summer. It is worth mentioning that Frampton landfill leachate treatment processes contains aeration tank, as pretreatment, followed by biological aerated lagoon, and peat adsorption for the post treatment.

2.2. Experimental pilots

Membrane bioreactor used in this study comprised 5 L aeration tank, a submerged hollow fiber ultrafiltration (ZW-1, Zenon environmental Inc.) with nominal pore size of 0.04 μm and total filtration surface area of 0.047 m^2. Feed and filtration peristaltic pumps were controlled for cycle of 110 s of filtration and 10 s of backwash by four times flow rate of filtration. The automatization and membrane wash was fully described in previous study (Zolfaghari et al., 2016).

Electro-oxidation lab scale cell comprised 1-L Plexiglas storage tank, 1-L reaction tank (dimension of 10 × 5 × 12 cm) equipped with an anode and a cathode with an inter-electrode gap of 2 cm. The anode had a solid surface area of 65 cm^2, thickness of 0.1 cm and the void surface area of 45 cm^2. The rectangular anode electrode was made of niobium coated with boron doped diamond (Nb/BDD), while the cathode was made of titanium with same physical characteristic of anode. The current was provided by a DC power supplier xantrex XFR 40–70 VA (Aca Tmetrex, Mississauga, Ontario, Canada). A peristaltic pump with flow rate of 170 mL/min, was used for recirculation of liquid between storage and reaction tank. A magnet stirrer was also used in reaction tank to homogenize the liquid. The schematic of both reactors are shown in supplementary files (Fig. 2-S).

2.3. MBR startup

The MBR pilot was initially inoculated by activated sludge collected from east municipal wastewater treatment plant of Quebec City, Quebec, Canada. Development of proper microorganisms was performed under continuous mode by setting the hydraulic retention time (HRT) at 48 h, and sludge retention time (SRT) of 200 days by raw LFL. During 28 days of startup stage, sludge concentration (represented by volatile solids (VS) concentration) was increased from 2.5 g/L to 7.1 g/L. Later on, HRT was gradually decreased to 18 h, to develop nitrification and assess the performance of MBR under different operating conditions. During all sets of experiments, dissolved oxygen and temperature was kept

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