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Tertiary treatment of a mixture of composting and landfill leachates using electrochemical processes

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HIGHLIGHTS

- Treatment of landfill leachate was examined using electrochemical process.
- Coupling of electrocoagulation (EC) and electro-oxidation (EO) was investigated.
- The EC process was more efficient compared to couple EC/EO process.
- The treated effluent was not toxicity to Rainbow trout and Daphnia.

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ABSTRACT

The study investigated the treatment efficiency of coupled electrocoagulation (EC) and electrooxidation (EO) processes for landfill leachate treatment in batch and continuous mode. The EC process (iron anode and graphite cathode) at 18.2 mA/cm^2 for 2.5 min resulted in COD, turbidity, total phosphorus, total coliforms and fecal coliforms removal of 58.1, 72.9, 98.5, 97.9, and 97.2% respectively. Under the same operating conditions, the coupled EC/EO (Ti-Pt anode, bipolar iron electrode, and graphite cathode) processes showed that the COD, turbidity, total phosphorus, total coliforms, and fecal coliforms removal of 56.5%, 78.3%, 96.3%, 97.2% and fecal coliforms 72.7%, respectively. The energy costs associated with the EC and EC/EO were 0.11 and 0.25 $/m^3, respectively. Compared to the batch configuration, the continuous configuration of EC resulted in similar processing performance. However, the EC/EO process resulted in the production of chlorates, perchlorates, and trihalomethanes as by-products. Moreover, the continuous process slightly increases the pH and ammonia concentration of the leachate and also resulted in the metallic sludge production with an average dryness of 4.2%. The toxicity tests determined that the treated effluent was not toxic to *Rainbow trout* and *Daphnia*.

1. Introduction

In the context of waste management, the operating life of a composting site or landfill can range from 25 to 60 years. Some municipalities are planning post-closure monitoring for about 30 years until the site has no longer a negative impact on the surrounding environment. Such impacts could be manifested by percolations of leachate in the soil, to the groundwater or even drifting towards the river systems, which constitutes a high risk of disease of the inhabitants but also the contamination of the fauna and flora (Litvan, 1995; Kehila et al., 2009; Abiriga et al., 2021; Wijekoon et al., 2022). Indeed, the leachate pollution index calculated for many landfills such in Bangladesh, India and Malaysia were very high ~19.8 (Parvin and Tareq, 2021). During leachate infiltration, the soil structure is altered due to mineralogical transformations, which can reduce its specific surface area and its porosity (Onyelowe et al., 2021). On the one hand, the alterations of permeability coefficient and in the total pore volume of soil are linked to the high salinity of leachate that has infiltrated it (Khodary et al., 2021). Moreover, alteration of soil structure is also due to the cationic exchanges of the system with the formation of new minerals such as hydroxyapatite, pyromorphite, ferrihydrite, hydroxy-pyromorphite, and strengite (Frempong and Yanful, 2008). Specifically, in the event of a leachate leak and depending on the permeability of the soil, the contamination of groundwater is at high risk (Papadopoulou et al., 2007; Aderemi et al., 2011; Alghamdi et al., 2021). In fact, studies have reported high concentration of certain physicochemical parameters,

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including conductivity (4.2–21.5 mS/cm), total dissolved solids (3.3–16.3 g/L), chlorides (2.2–11.7 g/L), sulfates (0.6–1.1 g/L), Mn (0.2–0.6 mg/L), and Fe (0.04–5.9 mg/L) in well water near landfills (Abd El-Salam and I. Abu-Zuid, 2015; Alghamdi et al., 2021; Parvin and Tareq, 2021). In addition, the interaction of organic carbon with heavy metals increases the mobility of the metals which in turn contaminate the ground-water (Christensen et al., 1996; Wei et al., 2021). Apart from organic carbon, the pH of the leachate also influences the mobility of heavy metals (Cameron, 1980; Chen, 1996; Kulikowska and Klimiuk, 2008, Xie, Ma et al. 2015). The contamination of water by the leachate depends on the method of waste disposal (active or not, controlled or not), runoff, and precipitation. Research investigation reported high concentration of organic matter (up to 250 mg/L) and ammoniacal nitrogen (up to 200 mg/L) in a river near an uncontrolled active landfill site (Yusof et al., 2009). However, near controlled landfill sites (active and inactive), high concentrations of heavy metals and inorganic nitrogen were reported (Yusof et al., 2009). The continuous leaching of landfill leachate in water and in soil disturbed the native microbial community (Gu et al., 2022). In soil, studies reported that, following leachate contamination, aerobic chemohetrotrophic and cellulolysis communities were significantly reduced while denitrifying communities tended to be more abundant (Hou et al., 2021; Gu et al., 2022). In water, studies have shown that the contamination of ponds by leachate (BOD and nutrients increase) led to a proliferation of microorganisms such as *Escherichia coli, Pseudomonas flourescens, Streptococcus feacalis*, *Salmonella* species*, Staphylococcus aureus, Bacillus* species*, Flavobacterium* species *and Saprophytic* spores and also increases reproductive stress in fish (Röling et al., 2001; Nwabueze, 2011). Considering the soil as a vector of pollution and a growth substrate, its alteration by leachate can inhibit the growth of the roots of certain plants and germs due to its genotoxicity and sodicity (Wong and Leung, 1989; Devare and Bahadir, 1994; Pessarakli and Szabolcs, 1999; Sang et al., 2006; Li et al., 2008). Leachate phytotoxicity tests have effectively demonstrated their harmful impacts on model species such as Sinapis alba and *Triticum aestivum* (Palm et al., 2022). Various studies have focused on the lethal and sublethal effects of leachate on indicator organisms. The main conclusions relate to the fact that leachate induces endocrine disruption, birth and developmental anomalies in many organisms such as carps, mouse, daphnia magna and brook trout (Calleja et al., 1986; Li et al., 2004; Alkassasbeh et al., 2009).

Leachate is a refractory waste effluent heavily loaded with polluting fractions. It is produced when water infiltrates the landfill or compost pile and is combined with the water produced during the aerobic decomposition of organic matter. Given its complexity, the regulations require adequate treatment of this effluent before it is released into the natural environment. Nevertheless, the concentration and the physicochemical characteristics of pollutants varies from one leachate to another because of the type of buried waste, the structure of the site, the climatic conditions, and their variability. According to several studies, COD concentrations can vary from 199 to 12,000 mg/L and BOD_5 from less than 1–3000 mg/L (García-López et al., 2014; Tahiri et al., 2016; Naveen et al., 2017). Ammonia and phosphorus were range from 1.3 to 21,800 mg N-NH_4^+/L and 0.52–485 mg/L, respectively (Krogmann and Woyczechowski, 2000; Gagnaire et al., 2011; Rajabi and Vafajoo, 2012; Brown et al., 2013). The pH of the leachate is another important factor which depends on the age of the site. In case of open site, the physicochemical parameters of leachates varied significantly with time (Cameron, 1980; Kulikowska and Klimiuk, 2008; Xie et al., 2015).

The presence of emerging and refractory pollutants such as chlorinated alkyl-phosphates, diethyl toluamide, perfluorinated compounds, atrazine, and morphine in leachates has been detected with a concentration ranging from ng/L to μg/L, (Chian and DeWalle, 1976; Chian, 1977; Christensen et al., 2001; Wiszniowski et al., 2006; Öman and Junestedt, 2008; Renou et al., 2008; Eggen et al., 2010; Masoner et al., 2014). Due to the potential impact of landfill leachate on the environment, various research investigations were carried out at laboratory and pilot scales for its treatment. Described as innovative and promising, those works are based on robust purification processes including membrane bioreactors, sequencing batch reactor and advanced oxidation processes such as process based on Fenton reaction (Lin and Chang, 2000; Laitinen et al., 2006; Bohdziewicz et al., 2008; Hu et al., 2016; Jagaba et al., 2021; Wu et al., 2021). Indeed, compared to conventional treatment processes, advanced oxidation processes have demonstrated their high efficiency in the degradation of refractory pollutants present in leachate following the production of species with high oxidation potential such as hydroxyl radicals and persulfates (Al-Qodah and Al-Shannag, 2019; Ushani et al., 2020; Bandala et al., 2021). The combination of advanced oxidation processes also showed even greater treatment performance with a clear reduction in energy costs (Al-Qodah et al., 2020a, 2020b; Sanni et al., 2022).

The treatment of leachates by electrolytic processes, in particular electrocoagulation and electrooxidation, is a substitution of physicochemical processes. The main advantage of EC, EO, and the coupled EC/EO process are: decrease in the production of metallic sludge compared to chemical coagulation, in situ coagulant production, stability of the salinity of the effluent by not adding anions associated to the metal salts (chemical coagulants), possibility of application of EC at native pH, direct oxidation at the anode and indirect oxidation by the species generated as a function of the ionic content of the effluent. Studies have highlighted the performance of these electrochemical processes compared to physicochemical processes for the treatment of leachate in terms of COD reduction, color removal, and heavy metals removal (Meunier et al., 2006; Ilhan et al., 2008; Veli et al., 2008). This work aims to develop the treatment process for the operational and closed composting site of landfill leachates located in Quebec, Canada. Moreover, The study attempted to develop a single-step treatment process to achieve maximum clarification, phosphorus removal, and disinfection for leachate mixture (from a closed sanitary landfill and an operating composting site). Further, the study analyzed the toxic effect of the treated leachate on the indicator organisms.

2. Material and methods

2.1. Leachate sampling

The leachates (comprised of composting and landfill leachates) utilized in the study were provided from an aerated lagoon process installed in a sanitary site located in the municipality of Bury in the province of Quebec. The aerated lagoon process was followed by chemical coagulation/flocculation and disinfection using hydrogen peroxide. The composition of mixture leachates (ML) recovered at the outlet of the biological process is presented in Table 1.

In order to assess the performance of electrolytic treatment (EC and EC/EO) leachate samples were taken after the lagoon step. For the purpose of testing disinfection, raw (untreated) leachate was sampled to seed the post-lagoon leachate in terms of bacteria, especially during the shutdown of the treatment station (winter period) and when coliform concentrations are very low. The 1% v/v spiking allowed to increase the total coliform and fecal coliform concentrations and therefore to properly assess the disinfection rates. Table 1 presents the average physicochemical and microbiological characteristics of the two types of effluent.

2.2. Experimental devices

2.2.1. EC and EC/EO operated in static batch

The EC static batch reactor consists of an iron anode (Fe) and a graphite cathode (Gr) connected in a monopolar configuration (Fig. 1. a.). The electrodes were completely immersed in the reactor. The electrodes were 1 cm apart and was chosen on the basis of the previous work. The electrodes were fixed at this distance since it influences the electrical resistance of the electrolyte which is proportional to the ohmic drop and therefore influences the effectiveness of the treatment

Table 1
Physico-chemical and microbiological characteristics of leachate.

Physico-chemical and microbiological parameters	Unit	Leachate after lagoon treatment	Raw leachate
COD	mg/L	997.2 ± 89.3	6243,6 ± 1763.7
Turbidity	UTN	77.4 ± 23.4	1873.7 ± 413.3
pH	–	7,65 ± 0,01	7.8 ± 1.1
Suspended solid	mg/L	235 ± 71,8	–
Total solid	mg/L	2496,25 ± 50,2	–
Total phosphorus	mg/L	2.9 ± 1.1	9.6
Residual iron	mg/L	8.6 ± 2.1	10.3
Ammonia	mg N/L	0.4 ± 0.2	–
Nitrate	mg/L	310 ± 7,1	–
Nitrite	mg/L	<0,4	–
Chloride	mg/L	282.6 ± 127.9	670
Chlorate	µg/L	<10	–
Perchlorate	µg/L	<0,5	–
Trihalomethane	µg/L	<4	–
Total coliform	UFC/ 100 mL	$2.7*10^4 \pm 2.9* 10^3$	$1.8*10^6 \pm 2.4* 10^6$
Fecal coliform	UFC/ 100 mL	$4.6 *10^2 \pm 3.1*10^2$	$3.4*10^4 \pm 2.3* 10^4$

(Hakizimana et al., 2016; Ding et al., 2021). The two electrodes had an active surface area of 110 cm^2. The electric current was applied using current generator type EXTECH R1.8. The varying current density from 4.5 to 18.2 mA/cm^2 was examined. The working volume of the reactor was 0.5 L and the continuous mixing of the leachate was carried out inside the reactor using a magnetic stirrer.

The electrolytic cell combining EC and EO (EC/EO) contained three electrodes (Fig. 1.b.). A Ti-Pt electrode was used as anode with a surface area of 65 cm^2 and a Gr electrode was used at the cathode with an area of 110 cm^2. A bipolar Fe electrode, not connected to the current generator, was installed between the cathode and the anode with a surface area of 110 cm^2. This configuration was chosen to promote both the anodic oxidation reactions at the Ti-Pt electrode and the half surface of the Fe electrode (bipolar) while ensuring its dissolution which subsequently will form an iron coagulant. The bipolar Fe electrode was installed at a distance of 1 cm on either side of the anode and cathode. The working volume of the reactor was 0.5 L.

2.2.2. EC operated in continuous mode

The continuous EC experimental unit contained three compartments for (a) EC process, (b) flocculation, and (c) settling (Fig. 1.c.). The EC and flocculation compartments have a working volume of 0.5 L while the settling compartment had a working volume of 4.7 L. The configuration of the EC electrodes and the connection to the current supply were similar to those adopted in the static batch. The settling tank contained five inclined slats having a total area of 1126 cm^2. The leachate inlet feed was set at 186 mL/min and that of the flocculant preparation at 7.4 mL/min (0.5 g/L of anionic polyacrylamide polymer). These parameters have been set so as to have an electric charge of 0.16 Ah/L.

2.3. Analytical methods

The concentration of the chemical oxygen demand was analyzed according to the MA protocol. 315-COD 1.0 from the Center of Expertise in Environmental Analysis of Quebec (CEAEQ). The COD values were read using a spectrophotometer at 600 nm using a spectrophotometer of the UV 0811 M136 type from the Varian Canada Inc. brand. The pH measurements were carried out with Fisher Scientific Accument brand pH meter (model XL25). The turbidity measurement was evaluated with Hach 2100 turbidimeter.

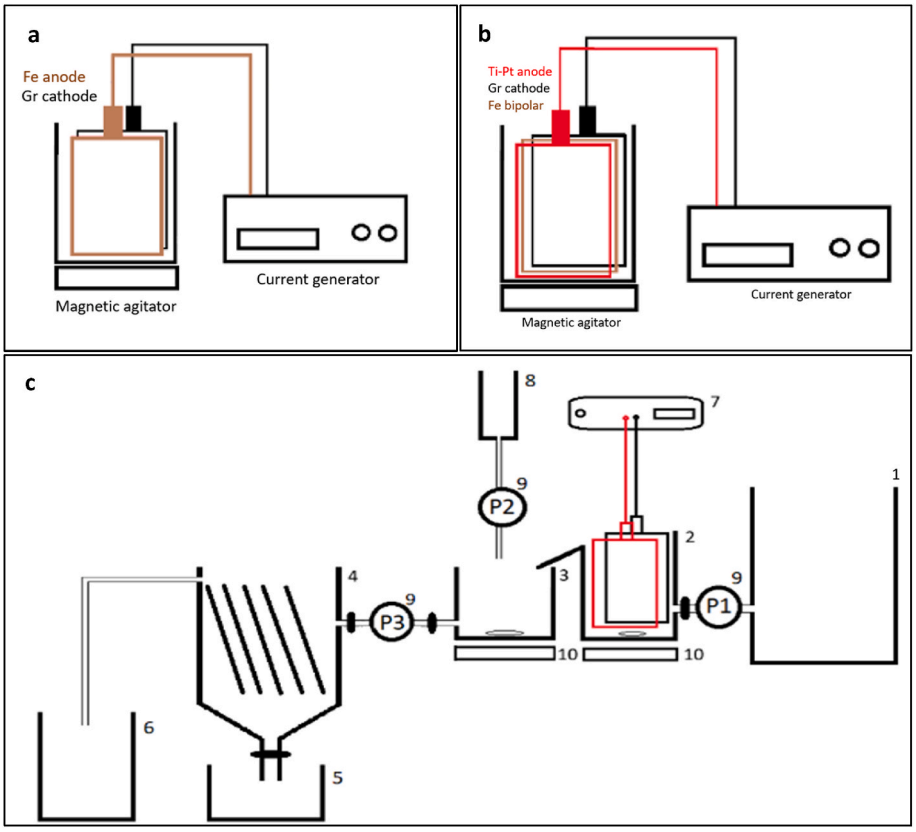

Fig. 1. Schematic description of the electrolytic units: a. EC operated in batch mode, b. EC/EO operated in batch mode and c. EC operated in continuous mode
1: Supply tank; 2: EC cell; 3: Flocculation cell; 4: Settling tank; 5: Sludge recovery tank; 6: Recovery tank for treated leachate; 7: Current generator; 8: Preparation of anionic polymer.

The analysis of residual iron and total phosphorus was carried out using ICP-AES of the Varian brand, model vista AX, Australia. Leachate samples were acidified with 5% nitric acid and stored at 4 °C until analyzed. Chloride ions were analyzed by the ion chromatography method using an ion exchange resin column of Ion brand PAC AS11-HC 4 μm and using the Integrion HPIC device from Thermo. The chlorites, chlorates, and perchlorates were analyzed by LC-MS-MS using the Thermo TSQ Quantum analyzer. The nitrites and nitrates were analyzed by the Lachat autoanalyzer following the 4500-NO3 E standards. The ammoniacal nitrogen was also analyzed by the Lachat auto-analyzer, QuickChem, according to the colorimetric method 10-107-06-2-B. The analysis of trihalomethanes was carried out by GC-MS/headspace of the Clarus 500 Perkin brand.

The concentration of suspended solids (SS), total solids (TS), and the measurement of dryness of metallic sludge were carried out by using MA method. 115-S.S. 1.2 of CEAEQ.

The total and faecal coliforms were measured according to the membrane filtration method (MA. 700-Col 1.0 for total coliforms and MA.700-Fec.EC 1.0 for faecal coliforms) proposed by CEAEQ and carried out by laboratories of Quebec City, water quality service. Before being sent to the laboratory, the samples were placed in 250 mL containers treated with sodium thiosulfate, in order to inhibit persistent oxidative reactions. The samples were then placed in a cold room (4 °C) away from light. Subsequently, the analysis were carried out within a period not exceeding 48 h.

The analysis of the toxicity of leachate was carried out by Bureau Veritas laboratories. The toxicity on rainbow trout (*Oncorhynchus mykiss*) was evaluated according to the method QUE SOP-00408 under reference SPE1/RM13 (2nd edition Environment Canada 2000). The test was carried out on a volume of 16 L of leachate under controlled temperature, lighting and density conditions. Secondly, the toxicity on Magna daphnia was evaluated according to the method QUE SOP-00406 under reference SPE1/RM14. The method consists in evaluating the LC_{50} of the sample (at six different concentrations % v/v: 0, 6.25, 12.5, 25, 50 and 100) over 48 h in containers containing 10 organisms and a volume of 150 mL of sample. As in the rainbow trout test, the temperature, lighting and loading density conditions were controlled. Finally, the toxicity to *Vibrio fischeri* (bioluminescent bacteria) was evaluated according to the AB SOP-00083 method under reference SPE1/RM24. The method consists in exposing *Vibrio fischeri* to different concentrations of the sample and then measuring their light inhibition (IC_{50}) from 0 to 15 min.

3. Results and discussion

3.1. Treatment of leachate by EC and EC/EO operated in static batch

3.1.1. Effect of electrical charge

The EC process is an electrochemical coagulation process which consist of formation of coagulating species from the anodic dissolution of metal electrodes (electrodes of iron, aluminum, zinc, and magnesium) (Kobya et al., 2003; Sahu et al., 2014; Nidheesh and Singh, 2017). These reactive species subsequently react with suspended matter and colloids to stabilize their negative charges (due to pH, isoelectric points, and zeta potential) to allow the aggregation, and thus reduce the turbidity of the effluent. The formation of aggregates was due to the van der Waals attraction forces and the electrostatic repulsion forces (DLVO theory: Derjaguin Landau Verwey Overbeek) (Lin et al., 2014). The repulsive forces are closely related to the thickness of the double layer of molecules which is compressed by the increase of ionic strength (additions of metallic ion).

The EC and EC/EO processes are mainly based on the dissolution of the anodic metal. In the case of iron, it leads to the formation of iron hydroxide acting as a coagulant. The following equations describe the coagulant production reactions, in particular ferrous and ferric hydroxides ($Fe(OH)_2$ and $Fe(OH)_3$) (Lakshmanan et al., 2009; Moussa et al., 2017).

$$Fe(s) \rightarrow Fe2+ + 2e- \; : E0 = 0.44\,V \quad (1)$$

$$Fe(s) \rightarrow Fe3+ + 3e- \; : E0 = 0.037 \;\; V \quad (2)$$

$$Fe2+ \rightarrow Fe3+ + e- \; . : E0 = -0.771 \;\; V \quad (3)$$

$$2H2O + 2e- \rightarrow 2OH- + H2(g) \quad (4)$$

$$Fe2+ + 2OH- \rightarrow Fe(OH)2(s) \quad (5)$$

$$4Fe2+ + 10H2O + O2 \rightarrow 4Fe(OH)3\,(s) + 8H+ \quad (6)$$

Dissolved iron concentrations can be estimated using Faraday's law, presented in Eq. (7), which relates the intensity of the applied current to the process start-up time. With the concentration of iron theoretically produced and that experimentally obtained, it is possible to describe the faradaic efficiency of the process which in the case of the study is an average of 89.9%:

$$FE = \frac{m(Fe)experimental}{m(Fe)theoritical} = \frac{mi(Fe) - mf(Fe)}{\frac{i \times t \times MW(Fe)}{z \times F}} \quad (7)$$

where FE: Faradaic efficiency [%]; mi (Fe): initial mass of the iron electrode [g]; mf (Fe): final mass of the iron electrode [g]; i: intensity of the applied current [A]; t: processing time [s]; MW (Fe): molecular weight of iron [g/mol]; z: number of valence of iron; F: Faraday constant.

In order to determine the optimal electric charge (product of current intensity and treatment time per unit of volume) for the clarification, disinfection and dephosphatation of the leachate pretreated by lagoon, EC and EC/EO tests have been carried out by varying the applied current density and the treatment time. Table 2 shows the percentages of COD, turbidity, and total phosphorus removal by varying current density from 4.5 to 9.1 mA/cm^2 and treatment time from 2.5 to 10 min which corresponds to quantities of electricity of 0.08, 0.16, and 0.32 Ah/L. The initial concentrations of COD, total phosphorus, and turbidity of leachate were 997.1 ± 89.2 mg/L, and 2.9 ± 1.1 mg/L, and 77.4 ± 23.5 NTU respectively.

The obtained results showed that the residual total phosphorus

Table 2
Effects of the amount of electricity and the combination of EC and EO on COD. turbidity and total phosphorus removal.

Process	Amount of electricity [Ah/L]	Current density [mA/cm^2]; Time of treatment [min]	COD removal [%]	Turbidity removal [%]	Total phosphorus removal [%]
EC/EO	0.32	9.1 mA/cm^2; 10 min	53.72	90.62	99.26
	0.16	9.1 mA/cm^2; 5 min	53.45	90.64	97.55
		4.5 mA/cm^2; 10 min	58.72	92.88	98.77
	0.08	9.1 mA/cm^2; 2.5 min	30.34	51.20	88.02
		4.5 mA/cm^2; 5 min	17.77	43.17	86.06
EC	0.32	9.1 mA/cm^2; 10 min	43.72	72.74	98.04
	0.16	9.1 mA/cm^2; 5 min	42.91	71.15	96.82
		4.5 mA/cm^2; 10 min	47.17	49.52	97.55
	0.08	9.1 mA/cm^2; 2.5 min	27.26	68.88	91.44
		4.5 mA/cm^2; 5 min	10.74	53.94	96.57

concentration was higher than the discharge guideline value (<0.3 mg/L) while using EC/EO process at a current density of 4.5 and 9.1 mA/cm^2 (for an electric charge of 0.08 Ah/L). In fact, phosphorus abatement rates between 86.06 and 88.02% were obtained while 89.6% abatement was required to meet the rejection criteria for residual concentrations of total phosphorus. A significant reduction in COD, turbidity, and total phosphorus were achieved at current density of 4.5 and 9.1 mA/cm^2 (for an electric charge of 0.16 Ah/L) by using EC and EC/EO processes respectively. It is worth noting that a relatively high concentration of coagulant agent (around 173.6 mg/L) was produced during electrochemical treatment. The treatment efficiency of EC/EO process when applying electric charge of 0.16 and 0.32 Ah/L were similar. This might be due to the enhanced production of coagulants at higher current density which decreases the turbidity removal (Agoungbome et al., 2016). By comparison, studies have reported 93% total phosphorus removal from leachate, with an initial concentration of 4.5 mg/L of total phosphorus and by applying EC (iron electrodes) with an electric charge of 0.03 Ah/L and a current density of 3 mA/cm^2 (Devlin et al., 2019). For higher total phosphorus concentrations (52.13 mg/L), an investigation obtained a reduction of more than 99.9% by applying an electric charge of 0.41 Ah/L and a current density of the order of 2 mA/cm^2 and that is 0.12 A for 100 min of treatment using a hybrid EC (Omwene et al., 2018). Compared to the present work, the two studies show a tendency for the reduction of total phosphorus apart from the imposed operating conditions. The physico-chemical characteristics are different from one leachate to another. Among the characteristics influencing their treatability by coagulation processes are the pH, redox potential, and the concentration of suspended matter (Sansalone and Kim, 2008). Since the concentrations of suspended solids are different and can compete with particulate phosphorus and therefore promote or limit its reduction rates. The EC/EO process showed to have high removal efficiency compared to EC alone. This is probably due to the ionic charge of the produced cogulant, where during the EC/EO, the oxidation of iron to ferric iron is favored then its reaction with the hydroxide ions leads to the production of $Fe(OH)_3$. $Fe(OH)_3$ has a greater coagulant power than $Fe(OH)_2$ generally produced during EC alone (Lakshmanan et al., 2009).

3.1.2. Effect of reaction time and current density

The effect of varying reaction time and current density to a given electrical charge has been experimentally identified as optimal at 0.16 Ah/L. By applying 0.16 Ah/L, the COD and turbidity reductions by EC process were around 45.1 and 60.3% and were 56.1 and 91.7% by EC/EO. Knowing that these tests were carried out at the same electrical charge of 0.16 Ah/L, and according to Faraday's law, the amount of coagulant (iron hydroxides) would be the same (Chen et al., 2018; Garcia-Segura et al., 2018). For example, by operating EC during 10 min of treatment by applying 4.5 mA/cm^2 of current density would lead to similar COD abatements obtained during the application of EC during 2.5 min at a current density of 18.2 mA/cm^2. This proposition remains valid as long as the quantity of electricity has been tested under a relatively low current density. Indeed, the reaction kinetics and tendency are less predictable by increasing the current density and reducing the treatment time below a limit threshold specific to the electrode used, to the cell, etc. Above this limit, other secondary reactions occur, such as significant development of H_2 at the cathode instead of OH^-, which may reduce the treatment efficiency (Dubrawski et al., 2014; Barışçı and Turkay, 2016; Guo et al., 2022). Also, the reduction of treatment time by increasing the current density cannot be done systematically since a minimum contact time is required for the reaction between coagulants and suspended matter/colloids. A study using EC process in batch mode, reported that the reductions of COD and turbidity did not exceed 50 and 40% respectively at an electrical charge of 0.2 Ah/L for 50 min of treatment (Li et al., 2011). Moreover, several studies have obtained COD reduction between 21 and 48% by applying current densities between 2.98 and 75 mA/cm^2 with varying treatment times of 30–100 min (Zailani and Zin, 2018). Compared to these studies, the present results are very promising in terms of COD reduction. This can be due to the initial characteristics of the treated leachate but also to the passivation of the electrodes during the experiment which lasts relatively longer (Ingelsson et al., 2020; Al-Raad and Hanafiah, 2021). Since, the aim of the present work is to scale up the processes for leachate treatment, the long processing times are limiting, and therefore the present work opted to increase the current density (from 9.1 mA/cm^2 to 18.2 mA/cm^2) against short processing times while keeping the electrical charge constant. In fact, at pilot and pre-industrialization visions of the process, it is advisable to operate at relatively low current densities to facilitate scale-up (Den, 2006; Santiago et al., 2014).

The treatment efficiency of EC and EC/EO are comparable to those obtained by conventional physicochemical treatment comprising of chemical coagulation (CC), chemical flocculation followed by settling. Fig. 2 shows COD values, turbidity, and total phosphorus concentrations after EC, EC/EO, and CC treatment. In terms of removal of COD and total phosphorus, and compared to EC/EO and CC, EC resulted in maximum removal of 56.9 and 97.5% of COD and total phosphorus, respectively. However, CC achieved the highest turbidity reduction rate of 88.1%. Along with these results, the residual iron concentrations in all the supernatant samples, after EC or EC/EO followed by flocculation and settling, were between 1.3 and 4.6 mg Fe/L. This iron concentration gives an orange-yellow color and may be the source of residual turbidity in leachate treated with EC and EC/EO. The CC process adopted by the station is supported by a pH adjustment which helps to define iron speciation and its solubility. Under optimal conditions, all iron used as a coagulant will be insoluble and will end up in the metallic sludge with minimal iron ion concentration in the supernatant.

3.1.3. Disinfection by-products formation by EC and EC/EO

Concerning leachate disinfection, and as shown in Fig. 3, EC and EC/EO allowed 97.9% of total coliforms removal after 2.5 min of treatment at 18.2 mA/cm^2. As for fecal coliforms, and under the same operating conditions, EC allowed a reduction of 97.2%, whereas EC/EO resulted in 72.7% of fecal coliforms removal. This removal would allow the leachate treatment plant to meet the discharge criteria set respectively at 2400 and 200 CFU/100 mL for total coliforms and fecal coliforms. In fact, the total coliforms of leachate supernatant treated either by EC or EC/EO are 300 CFU/100 mL and fecal coliforms 20 and 200 CFU/100 mL, respectively. The disinfection mechanism by these processes consists on the removal of bacteria attached to colloids and suspended solids, hence the relevance of clarification. Furthermore, it may also be linked to direct oxidations at the anode or via electrogenerated oxidants such as hypochlorite and hydrogen peroxide (Finch and Smith, 1986; Hakizimana et al., 2016; Elazzouzi et al., 2017). Contrary to what was expected, EC allowed more disinfection than the EC/EO pair although

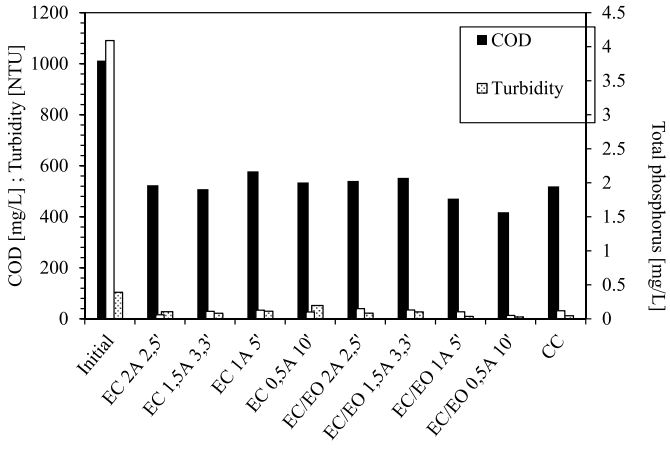

Fig. 2. Effect of current intensity and treatment time on COD, turbidity and total phosphorus reduction by EC and EC/EO.

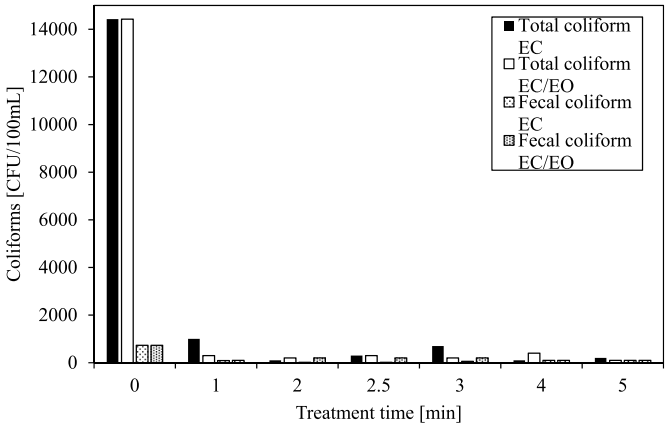

Fig. 3. Abatement of total coliforms and fecal coliforms by EC vs EC/EO.

the objective behind this coupling was to increase the generation of oxidants on the Ti-Pt anode.

In addition to evaluating the performance of leachate treatment by the electrolytic process, this work also analyzed the formation of by-products. From the point of view of producing disinfection chlorinated by-products, the leachates studied contain an average chloride ion concentration of 282.6 mg/L. The oxidation of these ions can lead to the formation of chlorates and perchlorates which constitute a risk for human health and the environment (Bergmann et al., 2009; Azizi et al., 2011; Ghernaout et al., 2011; Pérez et al., 2012). The following equations describe their formation from the chloride ions successively forming hypochlorite ions, chlorite, chlorates, and perchlorates where several reaction pathways are possible:

$$Cl- + 2OH- \rightarrow ClO- + H2O + 2e- \quad (8)$$

$$ClO- + 2OH- \rightarrow ClO2- + H2O + 2e- \quad (9)$$

$$ClO2- + 2OH- \rightarrow ClO3- + H2O + 2e- \quad (10)$$

$$ClO3- + 2OH- \rightarrow ClO4- + H2O + 2e- \quad (11)$$

$$ClO3- + H2O \rightarrow ClO4- + 2H+ + 2e- \quad (12)$$

$$ClO3- + OH0 \rightarrow ClO4- + H+ + e- \quad (13)$$

Therefore, residual concentrations of 600 μg/L of chlorates and 230 μg/L of perchlorates are generated after 2.5 min of electrolysis following the application of EC/EO process operated at a current density of 18.2 mA/cm^2 (Fig. 4.a.). Studies have also reported the formation of chlorate following EC/EO on leachate initially containing 5000 mg/L of chlorides. Chlorate concentrations were in the order of 230 mg/L when applying an electrical charge less than 1 Ah/L with a BDD anode. This study also showed that the use of DSA anode (Ir-Ru and Ir-Ta-Sn) led to significantly less chlorate formation (Ding et al., 2018). Otherwise, chlorate and perchlorate concentrations of less than 25 μg/L are recorded when the EC process is applied. This is probably related to the type of anode used in the two processes (EC: Fe vs EC/EO: Ti-Pt and $Fe_{Bipolar}$) and the density of the current applied (Czarnetzki and Janssen, 1992; Chen, 2004; Sánchez-Carretero et al., 2011; Lacasa et al., 2012). Indeed, this is mainly due to the anode material which influences the reactions of anodic oxidation and oxygen evolution. Electrodes with high oxygen evolution potential (non-active anode) such as BDD and PbO_2 tend to generate a lot of chlorinated by-products while electrodes with low oxygen evolution potential (activate anode) such as IrO_2 tend to convert chlorides into residuals chlorine useful for disinfection (Ghernaout et al., 2011).

On the other hand, the electrolytic oxidation of chloride ions can lead to the formation of hypochlorous acid (HClO) and hypochlorite (ClO^-).

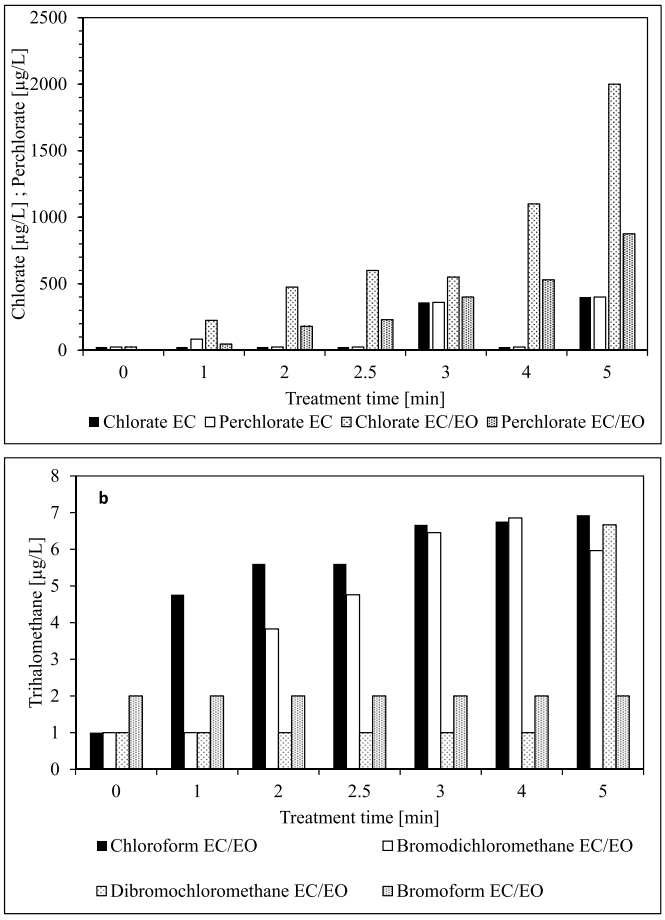

Fig. 4. Formation of by-products by applying EC vs EC/EO: a. Formation of chlorate and perchlorate and b. Formation of trihalomethane.

HClO and ClO^- can react with large numbers of organic molecules present in the leachate. These reactions on organic compounds are often the source of chlorinated organic compounds such as trihalomethanes (THMs), as well as haloacetic acids (Amy et al., 1987; Hong et al., 2007). Fig. 4.b. shows that the application of the EC, for 2.5 min of treatment time at the current density of 18.2 mA/cm^2, does not generate THMs while the EC/EO process leads to the formation of chloroform and dichlorobromomethane with concentrations of 5.6 and 4.7 μg/L. However, under these operating conditions, the concentrations of THMs are relatively low or even lower than the drinking water limits set at 80 μg/L (Regulation on the quality of drinking water in Quebec). This could be due to the type of organic and ammoniacal dissolved substances which are precursors to the formation of trihalomethanes (Díaz et al., 2011; Ben-Asher and Lahav, 2016). In support, studies have shown that the formation of THMs, mainly chloroform, depends on the current density applied, the organic and ionic content of the leachate (Anglada et al., 2011; Xu et al., 2020).

3.1.4. Estimation of energy costs of EC and EC/EO

Given that the objective of this work is to evaluate a potential substitution of the physico-chemical treatment (chemical coagulation followed by disinfection with hydrogen peroxide) of leachate by a one step electrochemical treatment, it is necessary to consider the treatment cost along with the performance efficiency (Ebba et al., 2021a). The operating costs are a determining factor in the implementation of the treatment process at pilot scale. This includes its processing performance, its autonomy, the life of its components and the energy costs linked to its continuous operation (Demirbas and Kobya, 2017; Hashim et al., 2017; Ebba et al., 2021b). Starting from the fact that the EC and

EC/EO applications allow a significant improvement in the treatment of leachate, subject of this study, the estimation of their associated energy costs is based on the values of current intensity applied, the required processing time, and the current voltage. The energy cost was calculated according to the formula presented below:

$$Energy\ costs\ [\$ / m3] = (i * U * t * PKw * 10 - 3) / V \quad (14)$$

With i: current intensity [A]; U: Voltage [V]; t: processing time [h]; P_{Kw}: Price per kilowatt hour [$ 0.09]; V: Volume treated [m^3].

The energy costs related to the operation of EC were estimated to 0.11 $ CAD for the treatment of one cubic meter of leachate, which is approximately two times less than the EC/EO process (0.25 $/$m^3$). This would be due to the voltage imposed during the application of the potential difference which is greater during EC/EO. In other words, since the inter electrode distance of EC/EO is 2 cm while that of EC is 1 cm, it would lead to greater resistance of the electrolyte (leachate) and therefore, higher voltage imposition under galvanostatic conditions (Béjar and Gutiérrez, 1993; Caspersen and Kirkegaard, 2012). Compared with the literature, these results are in line with most of the work that has been interested in the treatment of leachate electrochemically. The energy costs vary between 1.4 and 10.1 $/$m^3$ and are closely linked to the operating conditions adopted but also to the characteristics of the effluent, notably by its conductivity/resistivity (Ilhan et al., 2008; Ding et al., 2018; Sediqi et al., 2021).

3.2. Treatment of leachate by EC operated in continuous mode

The EC and EC/EO tests carried out in static batch have shown that the rates of clarification and phosphorus removal are quite similar by applying an electric charge of 0.16 Ah/L and a current density of 18.1 mA/cm^2. However, the EC/EO process has led to a relatively large production of by-products mainly chlorates and perchlorates. Based on these results and taking into account the energy consumption of each process (0.11 $/$m^3$ for EC and 0.25 $/$m^3$ for EC/EO), the EC operation was investigated in continuous mode for 300 min to analyze the toxicity of treated leachate by EC with a working volume of 34 L. The aim of the study was to provide an overview of the treatment of leachate by electrochemical means, notably by EC as a substitution for the physicochemical processes adopted by the leachate treatment station (chemical coagulation followed by disinfection with hydrogen peroxide). To do this, the amount of electricity and the current density were kept constant. The input flow rate of leachate pretreated by lagoon was set so as to have the residence time of 2.5 min in EC compartment.

As shown in Table 3, in continuous EC process, the average COD and turbidity removal were 73.3 and 72.2%, respectively. Compared to static batch tests, which showed the reduction of 56.9 and 75.4% respectively, COD reduction is more effective in continuous configuration while it is relatively similar in terms of turbidity removal. This could be related to the variable contents of the same leachate affected by temporal variations of the sample. Also, an increase in pH from an average of 7.5–8.5 was observed. This increase was also noticed during the static batch tests. It is mainly related to the cathodic reduction of water and the release of hydroxide ions (Ilhan et al., 2008; Aoudjehane et al., 2010). An average reduction of 67.4% of TSS was observed. For higher current densities and longer treatment times, previous studies were reported to achieve removal from 84 to 99% (Ahsan et al., 2014; Amani et al., 2014; Kabuk et al., 2014). As with suspended particles and colloids (negatively charged due to pH and isoelectric point), total phosphorus, mainly particulate, and total coliforms were reduced by 86.6 and 86.7% respectively. Following the application of EC, there is the cathodic reduction of nitrates to ammonia (Koparal and Öğütveren, 2002; Dia et al., 2017) according to the following equations:

$$NO3- + H2O + 2e- \rightarrow NO2- + 2OH- \quad (15)$$

$$NO2- + 5H2O + 6e- \rightarrow NH3 + 7OH- \quad (16)$$

$$NH3 + H2O \rightarrow NH4+ + OH- \quad (17)$$

A slight decrease in nitrate concentration were observed along with an increase in the concentration of ammonia between the inlet and the outlet at a rate of 14.1%. Several studies have also reported the reductions of nitrates to ammonium by different cathodes such as Cu/Zn, Fe, SS, and Al assisted by the effect of chloride ions and by anodes with weak oxygen evolution potential, such as Ti-Pt and Ti-IrO_2-Pt, which do not favor the production of amino by-products (Chen, 2004; Li et al., 2009; Dia et al., 2017). Also, the analysis of the percentage of dry matter in the metallic sludge showed an average dryness of 4.2%. The literature indicates that the percentage of dry matter in metallic sludge is closely linked to the characteristics of the effluent and to the doses of coagulant and flocculant used (Pouet and Grasmick, 1995).

Table 3
Leachate characteristics before and after continuous EC operation.

Physicochemical characteristics	Unit	Inlet	Outlet
DCO	mg/L	705.8 ± 33.8	188.5 ± 40.6
TSS	mg/L	260 ± 25.7	84.8 ± 17.3
Turbidity	NTU	197.1 ± 25.2	52.7 ± 4.6
pH	–	7.5 ± 0.1	8.5 ± 0.1
Total phosphorus	mg/L	2.6 ± 0.1	0.3 ± 0.1
NO3-	mg/L	347.1 ± 8.1	283.2 ± 38.7
N-NH4+	mg/L	9.2 ± 0.5	11.1 ± 0.1
Total coliform	CFU/100 mL	$3.5*10^4$	$4.6*10^3$

3.3. Toxicity assessment of leachate treated by EC

Since these leachates are intended to be released into the natural environment, their potential effect on aquatic organisms must be assessed (LégisQuébec, 2014). On cumulative leachate treated by continuous EC, bioassays were carried out to determine their toxicity. Leachate treated by lagoon followed by EC, with an adjustment of the final pH to values reaching neutrality (addition of 0.12 mL of H_2SO_4 $_{(99.9\%)}$/L of leachate), leads to 10% of mortality in *Rainbow trout* and *Daphnia*. While the leachate toxicity test without pH adjustment does not induce mortality in trout. Since the exposure of these two different species to the resulting leachate does not induce the mortality of 50% of their population, the effluent is qualified as non-toxic (CEAEQ, 2018). The literature reports that *Rainbow trout* and *Daphnia magma* are sensitive, to different degrees, to nutrients including ammoniacal nitrogen and to ionic content which can be described by total dissolved solids and heavy metals (Blaise and Férard, 2005). Previous investigations demonstrated that the toxicity of leachate treated by different processes, and on different species, present zero to moderate mortality (Wong, 1989; Rutherford et al., 2000). However, the exposure of *Vibrio fischeri* to the treated leachate effluent has shown a disturbance in its energy metabolism, thus being manifested by the inhibition of its bioluminescence. This type of test is very sensitive, rapid, and inexpensive compared to two other bioassays. Besides, interference can have an impact on the quality of the test, in particular by the turbidity of the effluent but also by its duration (15 min) which have certain limitations of reliability (Froehner et al., 2000; Parvez et al., 2006).

4. Conclusion

The EC and EC/EO processes tested in batch for the tertiary treatment of leachate gave a fairly global view on the post-treatment performance. They resulted in quite similar clarification, phosphorus removal and disinfection rates, at electric charge of 0.16 Ah/L, current density of 18.2 mA/cm^2, for 2.5 min of treatment time. In addition, both EC and EC/EO were as effective as the physicochemical process adopted by the leachate treatment plant and have achieved discharge goals in one step. The EC process was more efficient compared to EC/EO process due to its lower energy consumption and minimum by-products

production especially chlorates, perchlorates.

Regarding the post-treatment of the leachate by continuous EC, the reduction rates of COD, turbidity, total phosphorus, and total coliforms were respectively 73, 72, 86 and 87% and the dryness of the metal sludge was an average of 4.2%. The effluent at the end of the process showed no toxicity to *Rainbow trout* and *Daphnia*, but disrupted the energy metabolism of *Vibrio fischeri* probably due to the residual turbidity of the treated leachate. The study on the temporal variability of the characteristics of the leachate should be investigated to control and optimize the treatment process, before scaling up EC as a replacement for the chemical coagulation.

Author contributions statement

Alae Benguit: Conceptualization, methodology, formal analysis, investigation, writing original draft. Patrick Drogui: Supervision and review. Bhagyashree Tiwari: Conceptualization, methodology and review and editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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