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## Treatment of landfill leachate with high proportion of refractory materials by electrocoagulation: System performances and sludge settling characteristics

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#### A R T I C I E I N E O

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#### A B S T R A C T

The purpose of this study is to examine the feasibility of electrocoagulation (EC) in treating landfill leachate which has low biodegradability and a high level of inorganic compounds. Landfill leachate was taken from a French landfill which wants to cut down the organic matter present in the leachate especially the concentration of chemical oxygen demand (COD) which must be less than 120 mg O<sub>2</sub>/L. For this purpose, the effects of the operating parameters of EC, such as the current density  $j$  (23–95 A/m<sup>2</sup>). electrode gap (2–4 cm), and cathode natures were evaluated to find optimum operating conditions. These parameters were evaluated via COD, turbidity, absorbance at 254 nm, color,  $NH_4^+$ ,  $NO_3^-$ , and electrical energy consumption. The COD objective of the French landfill has been obtained after 135 min of electrocoagulation with the following optimum conditions:  $j = 95 \text{ A/m}^2$ , 2 cm gap between aluminum electrodes. In this experimental condition, residual turbidity was around 5 NTU and 80% discoloration ratio was acquired. The decrease absorbance at 254 nm showed a removal of humic substances. Nitrate removal was 40% but EC treatment was not appropriate for removing  $NH_4^+$ . The sludge aptitude to settling was studied in terms of sludge volume index.

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#### Introduction

Optimizing landfill leachate treatment, in order to reduce the negative impact on the environment, is today's challenge. Leachates are defined as the aqueous effluent generated as a consequence of rainwater percolation through waste, biochemical processes in waste cells and the inherent water content of waste itself. The composition of landfill leachates varies greatly depending on the age of the landfill  $[1]$ : leachates may contain large amounts of organic matter (biodegradable, but also refractory to biodegradation), where humic-type constituents make up an important group, as well as ammonia–nitrogen, heavy metals, chlorinated organic and inorganic salts [\[2\]](#page--1-0). These compounds are the main contributors of high chemical oxygen demand (COD), and nitrogen and color pollution [\[3,4\]](#page--1-0). High COD and toxic matter in the landfill leachate are the most important problems in leachate management  $[1,5]$ . COD is a measure of the oxygen used in the chemical oxidation of inorganic and organic

matter present in wastewater. It is also an indicator of the degree of pollution in the effluent and of the potential environmental impact of the discharge of wastewater in bodies of water. Hence, Governments strictly control these oxygen-demanding pollutants by setting standards for maximum levels of oxygen demand for all discharged wastewaters. However, leachate is difficult to treat, in order to satisfy the discharge standards for its variable composition and high proportion of refractory materials [\[1\]](#page--1-0).

Many treatment methods have been used to treat landfill leachate, such as advanced oxidation techniques [\[3\],](#page--1-0) membrane processes [\[6\]](#page--1-0), biological processes [\[7\]](#page--1-0), coagulation–flocculation methods [\[8,9\],](#page--1-0) and flotation process [\[10\]](#page--1-0). Some studies have been reported in the literature on the use of electrocoagulation (EC) alone for the treatment of landfill leachate [\[5,8,11,12\].](#page--1-0)

Electrocoagulation, a process that generates coagulant electrochemically, has been reported to be efficient in removing a wide range of pollutants from water and wastewater. The burgeoning interest of EC can be attributed to its many potential advantages such as simple equipment, easy operation, and a brief reactive retention period, no need to add chemicals, and a decreased amount of sludge. EC is a process consisting of creating flocs of metallic hydroxides within the effluent to be cleaned, by electro

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dissolution of soluble anodes, usually aluminum or iron. The formed flocs can be skimmed from the surface when the bubbles of hydrogen produced at the cathode allow flotation, or they settle. Compared with traditional flocculation and coagulation, electrocoagulation has, in theory, the advantage of removing the smallest colloidal particles; the smallest charged particles have a greater probability of being coagulated because of the electric field that sets them in motion [13–[16\].](#page--1-0)

The aim of this article is to propose electrocoagulation as a landfill leachate treatment process to treat the problem of a high organic load, such as persistent organic compounds, to meet the COD target (120 mg  $O_2/L$ ) of the French landfill a non-hazardous waste storage. To evaluate EC treatment performances, current density, electrode gap, and the nature of cathode are studied. COD,  $\rm NH_4^+$ ,  $\rm NO_3^-$ , turbidity, absorbance at 254 nm (ABS $_{254}$ ), color, and electrical energy consumption are also monitored. In an EC process, a significant amount of sludge is generated which has to be separated. Thus, in this work, parameter optimization was completed by investigation as sludge velocity settling and by identification of a sludge settling model. Our knowledge of the model will facilitate the sludge separation by using the regular processes for the waste water.

#### Materials and methods

#### Landfill site description and landfill characteristics

The leachate used in this study was collected from a French landfill which has been in operation for 2 years. This landfill, a nonhazardous waste storage, has collected no dangerous and no fermentable solid waste. Since the beginning of the operation, leachates have been stored in a retention pond which contains approximately  $2800 \,\mathrm{m}^3$ . The targets for discharge are total nitrogen  $(TN) < 10$  mg/L and  $COD < 120$  mg/L. Samples were always collected at the same location and were stored at  $4^{\circ}$ C and brought to room temperature before experimentation. No electrolyte was added to the solution, all experiments were conducted with initial conductivity and pH.

Average physicochemical characteristics of landfill leachate are shown in Table 1.

#### Electrocoagulation

All electrochemical experiments were carried out with 4 L solution in a 5L single-compartment electrolytic cell equipped with two parallel electrode plates. The anode was made of commercial aluminum plates. Two electrode gaps (2 cm and 4 cm) and two types of cathodes (aluminum and graphite) were studied. The active surface of the electrodes was around  $148 \text{ cm}^2$ . Before electrolysis, the aluminum electrodes were scraped to remove alumina layer formed during electrolysis. The electrodes

Table 1

Average physicochemical characteristics of leachate from French landfill.

Parameter	Men value
рH	8
Turbidity (NTU)	29
Conductivity (mS/cm)	31
TS $(mgL^{-1})$	1.4
$COD$ (mg $O2/L$ )	260
$NO_3$ <sup>-</sup> -N (mg/L)	170
$NH_4^+$ –N (mg/L)	187
$BOD_5$ (mg $O_2/L$ )	47
$Cl^{-}$ (mg/L)	293
ABS <sub>254</sub>	2.8

were installed vertically in the middle of the reactor. The electrodes were connected to a DC power supply (Micronix MX300) providing a controlled voltage up to 300V. All the runs were performed at room temperature and magnetically agitated. The electrical conductivity of the solution was sufficient for electrolysis, no salt addition was used as supporting electrolyte. Current density J was calculated as the applied current divided by the submerged surface area of the studied electrode. The current passing through the solution is accompanied by a Joule effect which can change the solution temperature [\[13\]](#page--1-0). In order to control the temperature influence during EC treatments, a preliminary study was carried out about the choice of the current density. This four current densities, 23, 46, 68, and 95 A/m<sup>2</sup>, were chosen in order to avoid any heating of the solution.

#### Batch settling test

After an EC run, liquids pre-treated were further submitted to settling tests to identify sedimentation sludge settling phases [\[17\].](#page--1-0) The batch test described by Kynch analyzed the settling of a homogeneous sludge concentration in a tube without a stirrer [\[17\]:](#page--1-0) liquids pre-treated by EC  $(1 L)$  was poured into a Pyrex glass column, 40 cm high and 6 cm in diameter for settling. The height of the liquid/sludge interface was registered at regular intervals and sludge settling was monitored for 1 h. This time has been selected to reach a complete settling at the end of the run: turbidity of the supernatant was constant.

#### Chemical analysis

Landfill leachate was taken to the top of the solution and used for analysis after 30 min. Samples were not filtered before analysis.

Chemical oxygen demand (COD) was measured according to the HACH method (Merck). Biochemical oxygen demand (BOD<sub>5</sub>) measurements were carried out in Oxitop IS6 WTW. The oxygen demand was determined after a 5 day incubation period at 20 $\degree$ C in the dark. Chloride analyses were carried out by a standard method (NFT 90-014). Total solids (TS) were dried at  $105^{\circ}$ C for 48 h. Turbidity was monitored with an Aqualytic PC compact. The pH and the conductivity of the samples were measured with a WTW 315i apparatus. Ammonium ( $NH_4^+$ –N) and Nitrates ( $NO_3^-$ –N) were measured using a Merck Method Photometric. Measurement errors are estimated at 5%.

Absorbances were monitored at  $600 \text{ nm}$  (ABS<sub>600</sub>) to study the evolution of the color and at  $254 \text{ nm}$  (ABS<sub>254</sub>), representing the presence of aromatics in the wastewater samples with a Merck spectroquant pharo 100 model UV–visible spectrophotometer, equipped with a 1 cm quartz cuvette.

To determine the influence of some water parameters on the EC process, we investigated the efficiency of the treatment applied to landfill leachate: it is the difference between  $C_0$  and C concentrations of the constituents before and after electrocoagulation divided by  $C_0$ .

#### Electrical energy consumption

The electrical energy consumption  $(E)$  was determined as Wh/kg COD removal using Eq.  $(1)$  where E is the electrical energy consumption (Wh/kg COD), U is the potential  $(V)$ , I is the current (A), t is the time (h), V is the volume of the solution treated (L),  $C_0$ concentration (mg/L) before EC and C concentrations (mg/L) after EC:

$$
E = \frac{Ult}{(C_0 - C)V} \tag{1}
$$

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