

54 (2015) 2986–2993 June



Combination of Electro-Coagulation and biological treatment by bioaugmentation for landfill leachate

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Received 4 November 2013; Accepted 18 March 2014

ABSTRACT

This study examines the use of the coupling of electro-coagulation (EC) and biological treatment of mature landfill leachate from a non-hazardous waste storage characterized by a relatively low Chemical oxygen demand (COD) and a low BOD₅/COD with a low biodegradability and a high level of inorganic compound such as NH₄⁴. During electrocoagulation treatment, a decrease in the chemical oxygen demand (COD) was shown with an increase in the current density. A toal of 33 and 56% COD removal were respectively obtained with 23 and 95 A m⁻². However, nothing was observed concerning the removal of ammonium. Nevertheless, biological treatment with or without addition of activated sludge (AS) has allowed the decrease in ammonium with a yield of 62% after 166 h. However, with the addition of activated sludge, the efficiency of nitrification is increased by 10%. As the bioaugmentation with AS has not improved the removal of organic matters and that the electrochemical treatment is not appropriate for removing NH₄⁺, coupling treatment such as EC-biotreatment was envisaged. An integrated process involving electro-coagulation as pretreatment of biological treatment to treat mature leachate appeared therefore relevant in the tested conditions at 23 A m^{-2} with a 15 and 98% global removal, respectively, of COD and NH_4^+ .

Keywords: Landfill leachate; Electro-coagulation; Biological treatment; COD removal; NH₄⁺ removal

1. Introduction

Landfills represent a potential source of wastewater called leachate, resulting from percolation of water contained in waste and rain through solid wastes. Leachates may contain large amounts of organic matter (OM), ammonia-nitrogen, heavy metals, chlorinated organic and inorganic salts [1]. Leachate consists of many different organic and inorganic compounds that may be either dissolved or suspended. They can either be biodegradable or non-biodegradable [2]. Organic contaminants in leachate are described using global parameters such as mainly chemical oxygen demand (COD), 5-day biochemical oxygen demand (BOD₅), and Total organic carbon (TOC) [3]. Landfill leachate is characterized by its generation rate and composition, both of which are affected by the age of the landfill site [4]. In young landfills (typically <1–2 yrs old), leachate is characterized by a high COD and a high BOD₅/COD. In contrast, leachate in old landfills (typically >5–10 yrs old)

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is characterized by a relatively low COD and a low BOD_5/COD [3,4].

Toxicity analyses using various tests organisms have confirmed the potential dangers of landfill leachates [5] and can cause significant environmental impacts if emissions are not controlled. However, leachates, discharged directly in a municipal wastewater treatment plant, may cause corrosion of the pump station and obstacles in maintaining constant effluent chloride residual, sludge bulking, and settling problems [6]. So, landfill leachates, characterized by a high variable composition and proportion of refractory materials, must be treated before discharge into the natural environment.

These effluents can be treated by different methods including physicochemical processes such as coagulation, flocculation and settling, electro-flocculation, adsorption, ultra filtration, reverse osmosis [7,8]. More recently, electrochemical processes had been proposed such as electo-coagulation (EC), which seems an interesting electrochemical process for removing OM. EC is an electrolytic process consisting of the dissolution of aluminum anodes upon application of a current between two aluminum electrodes for treatment of liquid wastewater containing inorganic or OM. As a matter of fact, Al³⁺ ions generated by the dissolution form a neutral hydroxide complex which acts as a coagulating agent for the suspended pollutants [9-12]. It's possible to use electro-coagulation process powered directly by photovoltaic solar modules for low cost treatment [13].

Conventional biological processes are universally used in leachate treatment [4,14], but due to its complex composition and stability, refractory OM is not treated. It should be pointed out also that the high levels of ammonium ions inhibit the biotreatment of landfill leachate [15]. Revolution in bioresearch has been conducted to develop some advanced biological processes: Among bioaugmentation is a cost-effective environmental technology which involves inoculation of cultivated microorganisms into a target environment, such as a wastewater treatment system, to enhance overall microbiological activity. It is often used to degrade toxic recalcitrant compounds such as organochlorines into harmless compounds, but also has been applied to improve activated sludge performance, enhancing the tolerance of the microbial community against various stresses, or protecting an indigenous microbial population from a shock load of a toxic recalcitrant substrate [16-18].

Furthermore, the combined electrochemical systems with biological treatment and AS showed interesting results on synthetic and industrial wastewater [19–21].

Precisely, this study examines the use of the coupling of electro-coagulation (EC) and biological treatment of mature landfill leachate characterized by a relatively low COD and a low BOD₅/COD with a low biodegradability and a high level of inorganic compound as NH_4^+ . This manuscript shows combination of these two methods for the treatment of landfill leachate, which will hardly remove recalcitrant organic constituents.

COD and ammonium are the main concern of leachate treatment and thus selected as the main observed parameters. To increase COD and NH_4^+ removal, studies were oriented toward an EC-biological coupling. These two treatments were operated sequentially to find the optimal operating conditions for the combined process and in combined process to remove COD, and NH_4^+ while EC was used in pretreatment to improve the removal efficiencies.

2. Experimental

2.1. Landfill leachate samples

Leachate samples were collected from a French landfill Company in operation for 2 years. Since the beginning of the exploitation, leachates have been stored in a retention pond which contains approximately 2,500 m³. The official authorization states that the discharge of treated leachate into the natural environment can be performed during only the months of December to May, the remainder of the year they will be stored on site in tanks waterproof. The targets for rejection are total nitrogen < 10 mg L⁻¹ and COD < 120 mg L⁻¹. Leachate samples were collected and stored in the dark at 4°C until the time of analysis. No electrolyte was added to the solution, all experiments were conducted with initial conductivity and pH.

2.2. Electro-Coagulation

EC tests were conducted using equipment which was composed of two aluminum electrodes; they have the same dimensions and are plunged in 4 L solution in a 5 L cylindrical reactor. The electrodes were made of commercial aluminum plates. For each electrode, the immerged (active) surface was 148 cm² and the distance between them was set at 2 cm. For this study, aluminum electrodes were preferred to iron because the current efficiency for aluminum electrode can be 120–140% [11]. The over 100% current efficiency for aluminum is attributed to the pitting corrosion effect especially when there are chlorine ions present [22]. Before electrolysis, the aluminum electrodes were scraped to remove the alumina layer formed during

electrolysis. Electrodes were connected to direct current power supply (Micronix MX300) with 300 V as maximal tension and 1 A as maximal intensity. All the runs were performed at room temperature, under magnetical agitation. The experiments were conducted at the natural pH of the leachate. Current density was optimized between 23, 46, 68, and 95 A m⁻². Current intensity was chosen in order to avoid any heating of the solution, a phenomenon which would influence the action of electrolysis on bacteria. For EC experimentation, COD, NH₄⁺, and NO₃⁻ were measured on supernatant after 30 min of settling.

The amount of aluminum dissolved in an EC experiment was calculated by Faraday's law (Eq. (1)).

$$w = (\text{ItM}/\text{nF}) \tag{1}$$

where *w* is the theoretical weight of the oxidized aluminum (g), *I* is the electrolysis current (A), *t* is the duration of the electrolysis (s), *M* is the molecular weight of Al (27 g mol⁻¹), *n* is the number of electrons (3) involved in the oxidation of Al, and F is the Faraday constant (96,500 C mol⁻¹).

2.3. Study of the biodegradation on landfill leachate

Cultures were carried out in triplicate in 1 L Erlenmeyer flasks with a net volume of 500 mL. All culture media were shaken by an Innova 40 shaker at 170 rpm and incubated at 20 °C. To analyze the impact of the endogenous flora and that of activated sludge (AS), experiments were carried out on the leachate inoculated with or without (raw sample) AS.

The AS was collected from the aeration tank of the municipal wastewater treatment plant of Bruz, near Rennes (France). This wastewater treatment plant (WWTP) has a total treatment capacity of 20,000 equivalent inhabitants and a low organic loading rate. The collected activated sludge was aerated and agitated for 24 h at ambient temperature. After settling for 30 min, the sludges have a concentration of 5 g L⁻¹. Inoculation was then carried out with 10 mL of pretreated AS, after oxygen saturation of the medium, namely after 2 h of agitation. For study of biodegradability, samples of 5 mL were periodically taken, which were centrifuged at 4°C (4,000 rpm, 15 min).

2.4. Description of the combined leachate treatment processes

The combination of biological treatment and EC is applied in this study. EC was used in pre-treatment: Leachate underwent an EC treatment for one hour, as described previously, and the supernatant after 30 min of settling was recovered for biological treatment with or without 10 mL L^{-1} activated sludge (AS) under the conditions previously defined. The methodologies for treatment process are described hereafter:

- electro-coagulation of leachate at various current densities followed by 30 min of settling (operation no. 1);
- (2) biological treatment of leachate with and without addition of activated sludge and without pretreatment (operation no. 2);
- (3) electro-coagulation of leachate at various current densities followed by biological treatment after 30 min of settling (operation no. 3). Operation no. 3 was schematized in Fig. 1.

Duplicate runs were conducted for each testing condition to ensure the reliability of the experimentation.

2.5. Analytical methods

Samples were not filtered before analysis. COD on supernatant after centrifugation at 4°C (4,000 rpm, 15 min) was measured according to the HACH method. Five-day biochemical oxygen demand (BOD₅) measurements were carried out in Oxitop IS6 WTW after 5 days of incubation at 20 °C in the dark. $NO_3^$ and NH_4^+ analyses were made on the soluble portion of samples for biological treatment settled for electrocoagulation. Analysis of different forms of nitrogen (ammonium (NH_4^+ -N), nitrites (NO_2^- -N), and nitrates (NO₃⁻-N)) was done by using Merck Method Photometric. Chloride analyses were carried out by following standard method (NFT 90-014). Total suspended solids (TSS) were dried at 105 °C for 48 h. All analysis was performed in duplicate and measurement errors are estimated at 5%. A turbidity meter AQUALYTIC PC Compact was used for turbidity measurement; conductivity was measured using conductimeter WTW 315i and pH using pH meter WTW 315.



Fig. 1. Schematic diagram of the experimental apparatus for the coupling process (operation 3).

The removal efficiency of COD, NH_4^+ , and NO_3^- after treatment was determined using the following Eq. (2):

$$R = (C_0 - C) \times 100/C_0 \tag{2}$$

In which, C_0 and C represent the measured parameters, respectively, in initial and final solution.

3. Results and discussion

3.1. Leachate characterization

Experiments were carried out on landfill leachate and their typical characteristics were presented in Table 1.

 BOD_5/COD was less than 0.2. It's typical for mature leachate and the proportion of refractory materials is high. Characterization of effluent indicates high levels of nitrogen and essentially nitrites, and the ratio BOD_5/NO_3^- -N value shows an inappropriate ratio for rapid heterotrophic denitrification. The results of Bouhezila et al. and Liu et al. were consistent with those obtained in this study [14].

3.2. EC treatment

In a first set of experiments, the efficiency of EC treatment is tested and Fig. 2 showed COD removal, which increases with current density. A total of 33 and 56% COD removal were respectively obtained with 23 and 95 A m⁻². A current density of 95 A m⁻² and 150 min treatment was necessary to obtain a COD value below requirements by the considered French landfill Company (120 mg L⁻¹). With 23 A m⁻², a value of 179 mg L⁻¹ was obtained (Fig. 3). Results about COD reported here are comparable with a previous

Table 1Composition of the tested landfill leachate

Parameters	Data ranges
pН	7.7 ± 0.38
Turbidity (NTU)	25 ± 1.25
Conductivity (mS cm^{-1})	4 ± 0.20
TSS (mg L^{-1})	1.4 ± 0.07
$COD \ (mg O_2 L^{-1})$	200 ± 10.00
BOD ₅ /COD	0.2 ± 0.01
$NO_3^N (mg L^{-1})$	25 ± 1.25
HN_4^+ -N (mg L ⁻¹)	170 ± 8.50
BOD_5/NO_3^N	1.5 ± 0.07
$\operatorname{Cl}^{-}(\operatorname{mg} \operatorname{L}^{-1})$	290 ± 14.5



Fig. 2. COD removal at various current densities in relation to time (O 23 A m⁻², \blacklozenge 46 A m⁻², △ 68 A m⁻², \bullet 95 A m⁻²).



Fig. 3. Comparison between COD value of objectives considered French landfill Company (---) and obtain after 210 min electro-coagulation with variable amount of dissolved Al (2 cm gap, aluminum electrodes, 210 min run).

study by Li et al. [9] who investigated the treatment of landfill leachate using EC: With 29.8 Am^{-2} , Li obtained 21.3% of COD removal after 50 min EC treatment. NH₄⁺ was not removed by EC treatment even with greater current density. A total of 23 and 40% NO₃⁻ removal were obtained respectively with current density of 23 and 95 A m⁻². As in other studies [24,25], nitrate removal was improved with an increase in current density and was explained by Eq. (3) [26]:

$$3NO_{3}^{-} + 6Al + 12H_{2}O \rightarrow NH_{3} + N_{2(g)} + 6Al(OH)_{3(s)} + 3OH^{-}$$
(3)

Moreover, it was reported that the removal of the nitrate with aluminum (chemical reduction) may first happen by adsorption onto the particles [26].

Typical chemical reactions at both the aluminum anode (Eq. (4)) and cathode (Eq. (5)) were:

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$$Al \to Al^3 + 3e^- \tag{4}$$

$$H_2O + e^- \rightarrow 1/2H_2 + OH^-$$
 (5)

According to Faraday's law (Eq. (1)), the electrochemical dissolution of the aluminum anode (Eq. (4)) produces Al^{3+} ions which further react with OH⁻ ions formed during reaction (Eq. (5)), transforming Al^{3+} ion initially into $Al(OH)_3$ and then into the gelatinous hydroxyl precipitate ($Al_n(OH)_{3n}$) [27]. They can effectively destabilize contaminant particles by adsorption and charge neutralization, resulting in an agglomeration due to the attractive Van der Wall forces and formation of stable precipitates that could be separated by a conventional separation technique [28,29]. It could be formation of particles with reduced solubility that entraps the pollutants [30].

On the cathode, the electrochemical reduction in water produces the formation of hydrogen bubbles that promotes a soft turbulence in the system and bonds with the pollutants, decreasing their relative specific weight. NH_4^+ was not removed by EC despite a greater current density. Concerning the influence of current density of the pH, an average increase in half unit is observed at the end of the treatment.

3.3. Biological treatment

In a first set of experiments, the effect of the addition of activated sludge on leachate treatment was examined. Fig. 4 showed an absence of COD variation during the course of the experiment after biological treatment. The value was above the standard discharge limit.

As can be seen in Fig. 5, the decrease in the ammonium was observed since the beginning of the culture.



Fig. 4. COD variation during the treatment of French land-fill Company.



Fig. 5. Ammonium variation during the treatment of French landfill.

In the presence of only indigenous microorganisms, ammonium decreases with a yield of 62% after 166 h of treatment. However, with addition of AS, the efficiency of nitrification increases by 10%. Indigenous flora and AS are able to grow on leachate and addition of AS increases the nitrification, but the lack of decrease of the COD proves the presence of refractory OM. This is in agreement with different studies [6,20].

As a consequence microbial inocula, namely activated sludge (AS) did not seem to have a significant effect on degradation of OM in leachate landfill. This suggests that since the leachate samples used for tests were from a two-year-old landfill, the indigenous bacterial community in the effluent was naturally adapted to degrade it components.

As the bioaugmentation with AS has not improved the process of removing OMs and that electrochemical treatment is not appropriated for removing NH_4^+ , coupling treatment was envisaged as EC-biotreatment.

3.4. Combined leachate treatment processes

After biological treatment, we have seen an absence of COD variation (Fig. 4), while COD decreases with EC treatment (Fig. 2). Sixty min EC run may be retained as the best compromise between good performance and limited power consumption. With EC as pre-treatment, we observed a COD removal increase after biological treatment, in comparison with only biological treatment whether in the presence of AS or not (Fig. 6). This is explained by the hydrolysis of some macromolecules. A COD increase has been previously reported during the biodegradation of synthetic dairy effluents [31]. This biodegradation can be attributed to the release of soluble compounds contained in the suspended solids or to the breaking down of the microbial floc [31]. In combined process, the reduction in COD increases compared to individ-



Fig. 6. COD removal after 166 h of biological treatment at various currents densities with EC in pre-treatment (EC treatment: 2 cm gap, aluminum electrodes, 60 min run).

ual process (Fig. 6). The same result was observed by Senthilkumar et al. [21] who studied combined treatment of dye wastewater.

The reduction in ammonium is not affected by electro-coagulation pre-treatment at $J = 23 \text{ Am}^{-2}$. EC does not inhibit nitrifying bacteria of the indigenous or exogenous flora. But for the value $J = 95 \text{ Am}^{-2}$, a decrease in removal of NH₄⁺ was observed (Fig. 7).

It means that the applications of various current densities don't disturb the biological degradation of ammonium until a sufficient value of current density is reached. Wei et al. [32] found that the bacterial viability in the presence of direct current depends on duration of the current application and current density. Lower yield at 95 A m⁻² may result in a reduction in phosphate. Ricordel et al. [33,34] show that the EC applied to various surface waters provokes a significant reduction in nitrate and phosphate ions, which



Fig. 7. Ammonium removal after 166 h of biological treatment at various currents densities with EC in pre-treatment during 2 h. (EC treatment: 2 cm gap, aluminum electrodes, 60 min run).

may constitute nutrients for bacteria. Applied current creates a potential difference from one extremity to the other of the cellular membrane on account of its electrical resistance. This potential difference modifies consequently the transmembrane potential, producing destruction of the cellular membrane. But total destruction requires an oxidant able to pass through the membrane and to reach vital centers [35,36]. During EC, the removal of oxygen and the variations of the potential oxidation reduction could be unfavorable conditions for living aerobic bacteria. The electro-coagulation supplies a robust packaging of nanoparticles susceptible to present some biocide or inhibition properties [33].

Djelal et al. just done a work on toxicity of leachate of a biogas plant before and after various treatment. It would be interesting to make the same study on the landfill leachate which studied in this paper [37].

4. Conclusion

After 150 min of EC treatment at a current density of 95 Am^{-2} , COD obtained is lower than 120 mg $O_2 L^{-1}$, value required by the considered landfill Company. EC was an effective method for removing recalcitrant OM (56% as COD). But with this treatment, the ammonium wasn't removed.

Concerning COD during biological treatment, augmented indigenous flora by conventional activated sludge, didn't enhance the biodegradation of the OM composed essentially by recalcitrant compounds (amines, amides, alcohols and aliphatic compounds, carboxylic acids).

For ammonium, augmented indigenous flora by conventional activated sludge enhance ammonium decreases after 166 h of biotreatment of 10% and results were in agreement with legislation. Successful bioaugmentation relies on various factors: The survival of AS inoculated into the landfill leachate was the most significant factor.

In the tested condition, the efficiency of an combined process involving Electro-Coagulation such as pre-treatment followed by biological treatment to treat landfill leachate from a two year-old appeared a technique suitable for mature leachate treatment: at 23 A m⁻² with a 15 and 98% global removal respectively of COD and NH₄⁺. But at current density 95 A m⁻², only around 80% of ammonium degradation was achieved in last of total treatment.

Possible strategies, such as EC pre-treatment and aeration of landfill leachate *in situ*, should be considered to create the optimum operational conditions for the growth and activity of the indigenous flora.

To explore a new solution to treat landfill leachate by coupling electro-coagulation and biological treatment at a low cost, it's possible to use electrocoagulation process powered directly by photovoltaic solar modules.

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