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Treatment of leachate by electrocoagulation using aluminum and iron electrodes

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Abstract

In this paper, treatment of leachate by electrocoagulation (EC) has been investigated in a batch process. The sample of leachate was supplied from Odayeri Landfill Site in Istanbul. Firstly, EC was compared with classical chemical coagulation (CC) process via COD removal. The first comparison results with 348 A/m² current density showed that EC process has higher treatment performance than CC process. Secondly, effects of process variables such as electrode material, current density (from 348 to 631 A/m²), pH, treatment cost, and operating time for EC process are investigated on COD and NH₄-N removal efficiencies. The appropriate electrode type search for EC provided that aluminum supplies more COD removal (56%) than iron electrode (35%) at the end of the 30 min operating time. Finally, EC experiments were also continued to determine the efficiency of ammonia removal, and the effects of current density, mixing, and aeration. All the findings of the study revealed that treatment of leachate by EC can be used as a step of a joint treatment.

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Keywords: Electrocoagulation; Iron and aluminum electrodes; Leachate; Mixing and aeration; Unit treatment cost

1. Introduction

Leachate has a complex structure and high pollutant load, and its treatment is quite hard to supply the discharge standards. Leachate becomes ahead of wastewaters as being the most difficult to treat as it is a wastewater with a complex and widely variable content generated within a landfill. Therefore, many pretreatment and combined treatment methods have been proven to treat leachate. By today, many treatment methods for its treatment have been proven. Some treatment stories such as biological treatment methods [1], membrane processes [2–4], advanced oxidation techniques [5,6], coagulation–flocculation methods [7], lagoon and wetland applications [8] have been examined in the literature. Because its characteristics change with advancing years of the landfill, these test methods have troubles such as decreasing treatment efficiencies and increasing cost [5]. Therefore, the implementation of a joint treatment comprising of a few treatment steps has been used to solve the problem.

In recent years, electrochemical treatment having features like relatively more economic and higher treatment efficiency has been a promising method. EC is one of the simple and efficient electrochemical methods for the purification of many types of water and wastewaters [9]. In this technique, which is characterized by its simple equipment, easy operation, and decreased amount of sludge, the coagulant is generated by electrolytic oxidation of an appropriate anode material that leads, at an appropriate pH, to the insoluble metal hydroxide which is able to remove a large variety of pollutants [10]. These metal hydroxide species neutralize the electrostatic charges on suspended solids and oil droplets to facilitate agglomeration or coagulation and resultant separation from the aqueous phase [11,12]. A growing research interest is reported on the treatment of various wastewater types: metal processing wastewaters [13], semiconductor production wastewater [14], textile dyeing wastewaters [15–19], tannery wastewater pre-treatment [20-24], olive mill wastewater [9,10,25], urban wastewater [26], and organics removal from poultry slaughterhouse wastewaters [27]. EC has also been

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worked to treat the landfill leachate by some researchers [28–31]. There is no doubt that high COD and toxic matter in the landfill leachate are of the most important problems in leachate management. In particular, it is well known that the landfill leachate can reach very high COD levels.

The mechanism of EC is extremely dependent on the chemistry of the aqueous medium, especially its conductivity. The mechanism of generating ions by EC can be explained with the examples of iron and aluminum, which was used as both the anode and cathode in this study. In an electrolytic system, iron produces iron hydroxide. In the case of iron or steel and aluminum anodes, two mechanisms for the production of the metal hydroxide have been proposed [11,32,33]. The reactions are given in Eqs. (1)–(11).

Mechanism 1

Anode

$$4Fe_{(s)} \to 4Fe_{(aq)}^{2+} + 8e^{-}$$
(1)

$$AI \to AI_{(aq)}^{3+} + 3e^{-}$$
⁽²⁾

Chemical

$$4Fe_{(aq)}^{2+} + 10H_2O_{(1)} + O_2 \rightarrow 4Fe(OH)_3 + 8H_{(aq)}^{+}$$
(3)

$$Al_{(aq)}^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H_{(aq)}^{+}$$
(4)

Cathode

$$8\mathrm{H_{(aq)}}^{+} + 8\mathrm{e}^{-} \to 4\mathrm{H}_2 \tag{5}$$

Mechanism 2

Anode

$$Fe_{(s)} \to Fe_{(aq)}^{2+} + 2e^{-}$$
 (6)

Chemical

$$\operatorname{Fe}_{(\mathrm{aq})}^{2+} + 2\operatorname{OH}_{(\mathrm{aq})}^{-} \to \operatorname{Fe}(\operatorname{OH})_2 \tag{7}$$

$$AI^{3+} + 3OH^{-} \rightarrow Al(OH)_{3}$$
(8)

Cathode

 $2H_2O_{(l)} + 2e^- \rightarrow H_2 + 2OH_{(aq)}^-$ (9)

Overall

$$Fe_{(s)} + 2H_2O_{(l)} \rightarrow Fe(OH)_2 + H_2$$
 (10)

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+$$
(11)

In the EC application with Fe or Al anodes, it is hoped that the reactions presented in Eqs. (1)-(11) occur.

The main aim of the work has been to investigate the treatment performance of a landfill leachate by EC technique. The experimental work covers the comparison of CC and EC, COD and ammonia removal performances under different electricity currents and durations for different anode types, formation of sludge and sulfate ion, variations in pH, temperature and ionic conductivity, and affects of mixing and aeration.

Table 1	
The properties of leachate from Odaveri Sanitary Landfill	

Analysis	Value
pH	8.2
Flow (m ³ /d)	2,200
COD (mg/L)	12,860
BOD ₅ (mg/L)	5,270
BOD ₅ /COD	0.41
TKN-N (mg/L)	2,580
Ammonia-N (mg/L)	2,240
Suspended solids (mg/L)	345
Turbidity (NTU)	1,340
Sulfate (mg/L)	32
Chloride (mg/L)	3,100
Alkalinity (mg CaCO ₃ /L)	8,700
Conductivity (mS/cm)	19.62 (at 23 °C)

2. Materials and methods

In the experimental study, leachate from the Odayeri Landfill in Istanbul was used. The Odayeri Landfill was founded in 1995, and has served 75 ha area for 8000 tonnes municipal solid waste (MSW) a day since that date. The properties of the leachate were analyzed and average values were given in Table 1.

All the experimental analysis was made according to Standard Methods [34]. COD tests were performed as recommended in the open reflux method, and ammonia nitrogen tests using the titration method. Merck analytical quality chemicals were used in the preparation of reagents. All the runs were performed at room temperature.

In the study, a digital dc power supply (GW Instek, GPS 3030 DD, 0–30.0 V, 0.0–3.0 A) was used in order to give a regulated electricity current to the electrochemical cell. The pH was measured by a Jenway 3040 brand a pH-meter.

Experiments were run in a batch reactor consisting of a 0.6 L glass beaker equipped with a cathode and an anode, both made of iron or aluminum and installed in parallel. The distance between the electrodes was 6.5 cm. The dimensions of electrodes



Fig. 1. Experimental set-up.

Table 2 The variation of electrical parameters applied during the runs

	Voltage (V)	Current density (A/m ²)	Power consumption ^a (kW-h/kgCOD _{removed})
Run 1	8	348	0.46
Run 2	10	435	0.67
Run 3	12	524	0.89
Run 4	14	631	1.13

^a These values are for later than 30 min contact time.

in 2.0 mm thickness were $5.0 \text{ cm} \times 15.0 \text{ cm}$. The total effective electrode area is calculated to be $(9 \text{ cm} \times 5 \text{ cm}) 45.0 \text{ cm}^2$. The electrodes were dipped into the beaker containing leachate with a 0.5 L working volume. The experimental set-up used in the present study is presented in Fig. 1.

CC process was compared with EC process for using different coagulant species and dose. The coagulant $(Al_2(SO_4)_3 \cdot 18H_2O vs. Fe_2(SO_4)_3 \cdot 7H_2O)$ doses which were used in CC process are equivalent to dissolved metal ions in 1, 5, 15 and 30 min for EC process according to Faraday's Law.

Both methods were tested for metal type, COD removal, and sludge and sulfate formations. Following this comparison, during EC treatment, the variations of some parameters such as pH, temperature, and conductivity were searched. By taking into consideration COD and ammonia removals, more efficient electrode material in the EC experiments was selected. Later, the effects of working parameters such as mixing and current density were also studied to achieve an optimum COD and ammonia treatment of the leachate sample. Applied current densities have been shown in Table 2. EC tests with mixing were realized by a magnetic stirrer in a speed of 200 rpm approximately which was optimum stirring rate according to literature [35]. All the contaminant removal performance tests were realized for 1, 5, 15, and 30 min electricity application durations. A separate batch study for each contact duration was made. At the end of each EC treatment study, a solution with flocks was allowed to settle for 60 min in the container before chemical analysis. The samples for chemical analysis were taken from limpid phase. Neither centrifuging nor filtration was performed in this study.

To understand the effect of aeration on ammonia removal after EC treatment, 0.25 L/min aeration by an aquarium type porous material (in 5 μ m pore size) producing coarse bubbles was made. Sludge volume was determined after 1 h settling period following the treatment and the amount of sludge produced was expressed as the ratio to whole solution. Experiments were repeated three times and the experimental error was around 3%.

3. Experimental results and discussion

In this section, results obtained during the study are given and discussed. Leachate treatment performance was determined by both CC and EC processes. This section deals with some considerations about the processes. Moreover, an evaluation of EC process was made via a lot of distinct operational conditions such as pH, current density, etc.

3.1. The comparison with CC and EC processes

Chemical coagulation is a treatment process that has been successfully applied for years. It is most commonly used for wastewaters rich in suspended and settleable material. As a result of fundamental processes occurring in EC, coagulation is succeeded via electrode and electrical current by releasing a coagulant. Ignoring power requirement, the most significant advantages of EC are higher removal rate, less sludge production and prevention of unnecessary ion transfer into treated wastewater. On the other hand, EC is not a single treatment step and is used in combination with electro-flotation and/or electro-oxidation (EOx). Besides, EC and EOx are not used separately those process occur spontaneously during the EC process. This is why high treatment efficiencies are obtained. In use of chemical coagulants, some anion concentrations in the solution treated can reach 1000–2000 mg/L, for instance SO_4^{2-} ion from $Al_2(SO_4)_3 \cdot 18H_2O$. This is another aspect that EC has an advantage over CC.

In the section, COD removal, sludge production and anion increment were evaluated in order to fully understand the difference between EC (current density: 348 A/m^2) and CC. COD removal efficiencies obtained under similar operational conditions are presented in Fig. 2.

It is seen from Fig. 2 that EC is a more reliable process than CC process for COD removal. Because of this, it can be said that an oxidation process also occurred due to electrical current in addition to chemical coagulation in EC process. COD removal efficiency was obtained as 33% in 30 min contact time with Feelectrode in EC process. On the other hand, the efficiency in CC process was obtained as 22% at the same situations. And it is noticed that COD removal efficiency was obtained as 45% in 30 min contact time with Al-electrode in EC process. On the other hand, the efficiency in the other hand, the efficiency in CC process was obtained as 45% in 30 min contact time with Al-electrode in EC process. On the other hand, the efficiency in CC process was obtained as 31% under the same conditions.

Since the results are related to the dissolved metal ions, they may be expressed in terms of COD removal efficiency (%)/dissolved metal ion (mg). In this way of representation, at the end



Fig. 2. Comparison of EC (with Al- and Fe-electrode) and CC.



Fig. 3. Comparison of EC and CC processes for sulfate change.

of 30 min, the results of EC-Fe, EC-Al, CC-Fe and CC-Al were obtained as 0.037, 0.136, 0.021 and 0.095 COD_{removed}/mg metal, respectively.

In the EC process, pure Fe-ions are introduced to water by means of electrodes greatly reduces the anion transfer to water with respect to chemical coagulation. In this context, a change of sulfate concentration in leachate with respect to time is observed and given in Fig. 3. According to Fig. 3, in the EC process sulfate concentration was decreased gradually from 32 to 1 mg/L for both electrodes. In other words, EC process takes into account higher efficiency in sulfate removal to be over 95% about in 1 min. However in this study removal was approximately 30 mg/L for lower initial sulfate concentrations (32 mg/L SO_4) . Due to the fact that initial sulfate concentration is very low, it is of importance that no sulfate formation occurs instead of removal. In contrast, chemical coagulation involved sulfate addition to the water instead of sulfate removal. In a study performed recently [36], sulfate removal efficiency was obtained for Al-anode and Fe-anode as 67 and 65%, respectively (contact time 5 min, initial sulfate concentration: 100 mg/L and current density: 620 A/m^2). On the other hand, in the CC process, sulfate concentration increased up to 1300 mg/L due to sulfate coming from the coagulants.

Sludge production is also important in these processes. So, the ratio of sludge produced in EC and CC processes were examined and presented in Fig. 4.

According to Fig. 5, the least volumetric sludge production was determined to be in the EC process with Al-electrode. Also, this figure clearly shows the change of sludge volume produced with respect to the increase in pH in EC process.

The results of analyses for leachate treatment have revealed that EC is more efficient in COD removal and sludge production as well as sulfate production. Also, it is seen that Al is better than Fe in the aspects of COD removal and sludge production. Although this result proves the advantage of EC process over CC process, this topic should be re-studied for other types of wastewaters. Because, higher conductivity of leachate leads required metal ions to get dissolved easily while a wastewa-



Fig. 4. Comparison of EC and CC processes for sludge ratio.

ter with a lower conductivity should be treated under longer retention times or by adding higher amounts of electrolyte.

It is seen that from the results presented above, the EC method has many pollutant removal advantages. In the following sections, experimental findings obtained for further operational parameters during EC process have been discussed.

3.2. Operational conditions of EC process

It was seen in previous sections that COD and sulfate removal efficiencies were higher in the EC process than the CC process. As a result, in the current section, the operational conditions such as pH, temperature and conductivity, etc., of EC process are discussed.



Fig. 5. Change of pH with different electrodes (Al and Fe) and with different current density $(348 \text{ A/m}^2 \text{ and } 641 \text{ A/m}^2)$.



Fig. 6. Change of temperature depending on electrode type and applied current densities with respect to time.

3.2.1. pH

The pH is continuously observed during the study. The results showed that, depending on the activities of the anode and cathode, pH gradually increased due to dominant activities of the cathode. The results are presented in Fig. 5.

From Fig. 5, when current density was 631 A/m^2 , pH changed from 8.1 to 9.5 with Al-electrode and from 8.1 to 9.2 with Feelectrode. When current density was 348 A/m^2 , pH changed from 8.3 to 8.9 with Al-electrode and from 8.3 to 8.7 with Feelectrode.

As seen in the figures, an important change in pH was experienced in a short period of time. With the consideration that EC will be followed by an ammonia stripping process, the pH of water is expected to be high after EC. The pH adjustment brings about an extra operation cost for leachate treatment plants. Therefore, it is said that the increase in pH is a desired result. Al electrodes showed a better efficiency than Fe electrodes did. The results of 435 and 524 A/m² studies showed similar pollutant removal efficiencies. According to the results of the study performed by Can et al. [19], pH was increased from 6.9 to 7.8 with 100 A/m² and in 10 min contact time.

3.2.2. Temperature and conductivity

The temperature in the reactor tends to increase during the study as a result of reactions. This increase in temperature as a result of electrolytic reactions depending contact time, electrode type and applied electrical power is shown in Fig. 6. In addition, depending on contact time and applied electrical power, electrical conductivity also changes. The change in the conductivity is shown in Fig. 7.

From Fig. 6, when current density was 348 A/m^2 , temperature changed from 23.8 to 25.5 °C with Al-electrode and from 23.8 to 25.3 °C with Fe-electrode. The temperature tends to increase as a result of electrolytic reactions.



Fig. 7. Change of conductivity depending on electrode type and applied current densities with respect to time.

When Fig. 7 is examined, it is concluded that conductivity decreases as a result of electrochemical treatment. When current density was 348 A/m^2 , conductivity with Al-electrode was decreased from 19.6 to 18.6 mS, and was decreased from 19.6 to 18.6 mS with Fe-electrode. When current density was 631 A/m^2 , conductivity was decreased from 19.6 to 18.2 mSwith Al-electrode, and was from 19.6 to 18.3 mS with Feelectrode. According to the experimental results, increasing electrical power has supplied an unremarkable drop in the conductivity.

3.2.3. Electrode type

Fe and Al electrodes were compared under similar operational conditions for COD and NH₃-N removals. The results obtained from the experiments for both electrode types are presented Fig. 8.

As seen in Fig. 8, the performances of electrodes changed under similar conditions. Al electrodes showed a higher treatment efficiency than Fe ones on the basis of COD removal. The rates of COD removals for Fe and Al electrodes obtained were 35% and 56% in 30 min contact time, respectively. Fe electrodes transfer higher numbers of Fe ions into solution and they produce a higher amount of sludge. Due to the fact that the costs of both types of electrodes are almost same, it will be a good choice for higher treatment efficiencies to select Al electrodes. According to a study performed on diluted leachate (%10) by Inan et al. [25], COD removal efficiencies with Al-electrode and Fe-electrode were obtained 52% and 42%, respectively (pH: 6.2, current density: 200 A/m^2 , and contact time: 30 min). Al-electrode has a higher treatment efficiency than Fe one for NH₃-N removal. The removals of NH₃-N were determined to be almost 11% and 14% in 30 min contact time for Fe and Al electrodes, respectively.



Fig. 8. Effects of electrodes on COD and NH_3 -N removal efficiency (current density: 631 A/m²).

3.2.4. Mixing

EC process being an electrochemical treatment method is realized by mobilization of ions by means of electricity applied. During this process a stable solution medium will be much more efficient for their mobilization. Mixed medium will cause an upset of ion mobilization and some uselessly electricity consumption. By this consideration, the effects of mixing on the EC process were examined. By this examination, findings shown in Fig. 9 for COD and NH₃-N removal were obtained.

It is seen from Fig. 9 that the decrease in the efficiency of COD removal by mixing operation was 34% to 20% in the first min,



Fig. 9. Effects of mixing on COD and NH₃-N removal efficiency.

and from 59% to 44% at the end of 30 min contact time. These experimental data supported the above consideration. That is, mixing operation decreases the efficiency of COD removal.

It is seen from Fig. 9 that the increase in the efficiency of NH_3 -N removal by mixing operation was only 2% at the end of 30 min contact time.

The objective was to reveal the effects of mixing on treatment efficiency. Mixing process negatively affected COD removal efficiency although it increases NH₃-N treatment performance. The main reason for this result is the fact that COD removal is accomplished only via EC treatment while NH₃-N is removed via both EC treatment and stripping action. Mixing operation influences the movement of ions in water (negatively charged ones towards anode and positively charged ones towards cathode) negatively. Considering the fact that mixing may cause the break up of flocks, mixing is not recommended to be a good operation for EC treatment. The treatment efficiency for NH₃-N is slightly increased by 2% because ammonia is easily stripped by mixing. Taking into account these considerations it was concluded that no mixing should be included in EC treatment process to obtain higher removal efficiencies and to keep the system operating. Mixing reduces removal efficiency in the experiments in which it is applied together with electrodes. EC and mixing processes were applied by Alinsafi et al. [17]. The results of experiments performed in this manner showed higher COD removal efficiencies.

3.2.5. Current density

Another experimental study was performed to determine the effects of operational condition on COD removal efficiency by changing current density. For this purpose, 8, 10, 12 and 14 V potentials were applied corresponding to 1.5, 2, 2.5 and 3 A. The treatment efficiency seemed to be slightly increasing with increasing current density. The results are shown in Figs. 10 and 11.

Fig. 10, current density was changed from 348 to 631 A/m^2 . When current density was increased from 348 to 631 A/m^2 , the efficiency of COD removal was also increased from 18.3% to 27.3% in the first minute and from 45.5% to 59.1% at the end of 30 min contact time.

According to Fig. 11, current density was also changed from 348 to 631 A/m^2 . When current density was increased from 348 to 631 A/m^2 , the efficiency of NH₃-N removal was also increased from 2.8% to 3% in the first min and from 8.8% to 14.3% at the end of 30 min contact time. As a matter of fact, after the 15 min with the help of the increase in pH and temperature much ammonium is converted to ammonia nitrogen is stripped with gases formed around the cathode.

3.2.6. Cost analysis

Operational costs of electrical current are based on contact time. So short contact time is preferred and current density with optimum pollutant removal [37]. Therefore, the best operational condition is estimated to be work with low current density. The energy requirements per cubic meter of leachate and per kilogram COD are shown in Figs. 12 and 13, respectively. It is concluded from the figures that the most economical treatment



Fig. 10. Effect of current density on COD removal.

is accomplished under low current density with an ignorable disadvantage (for this case) of longer required contact time.

As is seen in Figs. 12 and 13 that when current density was applied as 348 A/m^2 , unit energy consumptions were obtained as 12.5 kWh/m^3 treated leachate and 0.46 kWh/kg COD removal. When current density was applied as 435 A/m^2 , unit energy consumptions were obtained 19.6 kWh/m^3 treated leachate and 0.67 kWh/kg COD removal. When current density was applied as 524 A/m^2 , unit energy consumptions were obtained as 28.3 kWh/m^3 leachate and 0.89 kWh/kg COD removal. When current density was applied as 631 A/m^2 , unit energy consumptions were obtained as 11 kWh/kg COD removal.



Fig. 11. Effect of current density on NH₃-N removal.



Fig. 12. Energy requirements per m³ of leachate.

3.2.7. Ammonia removal via aeration

One of the most important problems in leachate is nitrogenous compounds. The treatment of this type of wastewater is difficult due to very high ammonia content. The problems are especially related to fluctuations in nitrogenous compounds depending on landfill age. When a leachate treatment plant is designed and constructed, these fluctuations in nitrogenous content of leachate should also be considered. The removal of nitrogenous compounds in leachate treatment plants is generally accomplished via increasing pH along with diffused aeration, which is called ammonia stripping. Therefore, during the direct treat-



Fig. 13. Energy requirement per kg COD removed.



Fig. 14. The effect of aeration and pH on ammonia removal efficiency.

ment of leachate using EC, ammonia concentration in leachate was observed. 10–15% of ammonia removal was accomplished depending on current density and contact time. Although this seems to be low, 250–350 mg/L of ammonia was removed to accomplish this removal efficiency. To increase ammonia removal efficiency, a diffused aeration system with a 0.5 μ m pore size at 1 L/min air flow rate was applied in this study. Ammonia removal efficiency was increased in this way. In the study, to improve the pH increase, 6N NaOH was used and pH was increased to 9.6. Only then was the ammonia removal efficiency of 24% was accomplished in the first 30 min in this way. The data obtained from the experiment is presented in Fig. 14.

4. Conclusion

COD removal efficiency was obtained as 32% in 30 min contact time with Fe-electrode in EC process for a current density of 348 A/m^2 . On the other hand, the efficiency in CC process was obtained as 22% at the same as situations. COD removal efficiency was obtained as 45% in 30 min contact time with Alelectrode in EC process. On the other hand, the efficiency in CC process was obtained as 31% in the same situations. In other words, EC process takes into account higher efficiency in sulfate removal to be over 95% about in 1 min. In the CC process, sulfate concentration increased up to 1300 mg/L due to sulfate coming from the coagulants. The results of analyses for leachate treatment have revealed that EC is more efficient in COD removal and sludge production as well as sulfate production.

When current density was applied as 348 A/m^2 , unit energy consumptions were obtained as 12.5 kWh/m^3 treated leachate and 0.46 kWh/kg COD removal. When current density was applied as 435 A/m^2 , the unit energy consumptions obtained were 19.6 kWh/m^3 treated leachate and 0.67 kWh/kg COD removal. When current density was applied as 524 A/m^2 , the

unit energy consumptions obtained were 28.3 kWh/m^3 leachate and 0.89 kWh/kg COD removal. When current density was applied as 631 A/m^2 , unit energy consumptions were obtained as 39.7 kWh/m^3 treated leachate and 1.1 kWh/kg COD removal.

The results showed that over 59% of COD and 14% of ammonia removal were accomplished with 30 min contact time for a current density of 631 A/m² with aluminum electrode. When desired, ammonia removal efficiency can be increased up to 24% by means of various modifications (aeration, and alkalinity addition). Considering these results, it is recommended to use EC for leachate pre-treatment. The increase in pH will dramatically decrease the cost of alkalinity addition for ammonia stripping. When the effect of temperature on ammonia stripping is considered, the temperature increase in this process is one of its advantages. In conclusion, the pre-treatment of leachate is accomplished at a low cost in a short period of contact time. It also provides effluents with alkaline characteristics and at a high temperature. The potential conversion of TKN into ammonia during EC was also evaluated. In conclusion, ammonia removal was considered to be satisfactory.

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