Electrochemical treatment of solid waste Leachate using combined electrocoagulation and electrochemical oxidation treatment

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The combined electrocoagulation and electrochemical oxidation (EC-EO) technique was used to electrochemically treat solid waste leachate by efficiently removing COD, nitrogen-based products, and chromium using bipolar and monopolar electrodes arranged in one reactor with BDD as an anode, stainless steel as a cathode, and Al as bipolar electrode. After 12 hours of treatment, the removal efficiency of the EC-EO reactor was 22% higher than that of the EO reactor, and after 24 hours of treatment, the COD removal efficiency of the EC-EO and EO reactors was 98.5% and 97.8%, respectively. After 24 hours of EC-EO treatment at 0.7 A current intensity, NH_4^+-N, Cr (VI), and TN removal efficiency was 85.8%, 85.3%, and 62.2% for NH_4^+-N, Cr (VI), and TN, respectively. The integration of EO with the EC process enhanced overall performance, as indicated by the results in terms of energy usage. The EC-EO procedure uses 50kWh/kg to remove 98% of COD and 847kWh/kg to remove 85% of NH_4^+-N, respectively. The combined EC-EO method removed more than 97%, 63%, 62%, and 40% of COD, NH_4^+-N, Cr (VI), and TN from solution after 16 hours. The pH effect in the study also revealed that neutral or acidic conditions are beneficial for great treatment during the EC-EO process, and removal efficiencies are increased by decreasing the initial pH value to 4 and 6.5, and more than 90% treatment of COD, NH_4^+-N, Cr (VI), and TN was obtained in less than 16 hours in the combined EC-EO process, and pH=6.5 was chosen as the optimal pH value for the combined EC-EO process. A comparison of the developed treatment method with other reported reactors in the literature revealed that the developed reactor performed similarly to some reported reactors, if not better, which can be attributed to the well-organized and efficient activity of electrodes in the presented EC-EO reactor.

1. INTRODUCTION

Rainwater filters through wastes in a landfill and comes into contact with buried wastes, forming leachate, which leaches (or pulls out) chemicals or elements from those wastes [1, 2]. This contaminated liquid might collect impurities and leak into the ground [3, 4]. This highly poisonous liquid has the ability to damage land, groundwater, and waterways, as well as pollute the environment and threaten human health [5-7]. The leachate contains a wide range of hazardous compounds, many of which have been linked to cancer or other major health problems in humans [8, 9].

Many studies on determining the chemical content of leachate have revealed that it contains a significant amount of organic contaminants, including the 5-day biochemical oxygen demand (BOD_5), chemical oxygen demand (COD), ammonia (NH_4^+-N), hydrocarbon suspended solids, heavy metal concentrations, and inorganic salt [10, 11].

Traditional landfill leachate treatment solutions can be divided into three categories to tackle leachate problems: I leachate transfer, (ii) biological treatment, and (iii) chemical-physical processes [12-14]. The first phase of treating landfill leachate is usually leachate transfer, followed by recycling and mixed treatment with household sewage [15-17]. Biological treatment involves removing nitrogen and other biological chemicals from wastewater using a variety of filters [18, 19]. An aerobic zone would be followed by an anoxic zone in a standard biological therapy system [20, 21]. In a process known as nitrification, microbes in the aerobic zone breakdown organic substances into carbon dioxide and water, and oxidize ammonia and ammonium into nitrite and nitrate [22, 23]. Chemical-physical procedures are wet oxidation methods that can be employed if organic chemicals can be oxidized while metals, ammonia, and dissolved solids can be removed [24, 25]. Chemical oxidation, adsorption, chemical precipitation, Fenton and electro-Fenton oxidative treatments, coagulation/flocculation, sedimentation/flotation, electrochemical oxidation, electrocoagulation, and air stripping are some of the processes used [26-31].

Many of these solutions, however, necessitate a large reactor area, expert labor, regular cleaning and removal of membrane fouling, as well as expensive capital and operating costs. Electro-coagulation (EC) and electrochemical oxidation (EO) are two treatment techniques that can be used to degrade organic and inorganic contaminants in solid waste leachate without the requirement for trained labor or a lengthy start-up process. As a result, this study was carried out to electrochemically treat solid waste leachate by removing COD, nitrogen-based products, and chromium using a combined EC-EO technique with BDD as anode, SS as cathode, and Al as bipolar electrode using BDD as anode, SS as cathode, and Al as bipolar electrode.

2. EXPERIMENT

2.1. Bio-treated landfill leachate

The real leachate was collected from a municipal sanitary landfill site in northeast China's secondary sedimentation tank. For leachate, post-biological treatment was performed, followed by membrane filtration. Table 1 shows the composition of the original leachate effluent. A DDS-307A

conductivity meter was used to measure the conductivity (Rex Shanghai, China). A pH meter was used to determine the pH levels. The BOD_5 was determined using Standard Method 4500-O C and Standard Method 5210 B of the standardized respirometric OxiTop method (WTW, Xylem, Weilheim, Germany). The total solids in the sample were assessed by drying it at 103–105°C (APHA, Method 2540 B). A Hach COD reactor 45600-00/Hach DR 2010 spectrophotometer was used to do the COD measurements (method 5220-D). The spectrophotometer SM3500-Cr B detected chromium metal in wastewater. The APHA mercuric nitrate method 4500-C1C was used to determine the chloride ions. NH4+ ions and total nitrogen (TN; sum of all nitrogen-based products such as NH_4^+-N, NO_3–N, NO_2–N, and organic nitrogen) were measured using the Hach method with a spectrometer. The samples were collected and stored in 25 L plastic containers at a temperature of 4 °C after collecting the leachate. Before the measurements, 1 M sulfuric acid (99%, Hebei Jinmengmiao Chemical Products Co., Ltd., China) and 1 M sodium hydroxide were used to correct the pH of the samples (99%, Xiamen Huaxuan Gelatin Co., Ltd., China).

Table 1. Composition and properties of original effluent of leachate

Properties	Value
5-day biochemical oxygen demand (BOD_5)	700 mg/l
Chemical oxygen demand (COD)	1150 mg/l
Suspended solids	300 mg/l
Total Nitrogen (TN)	1010 mg/l
NH_4^+-N	750 mg/l
Cr (VI)	300 mg/l
Chloride ion	700 mg/l
pH	6.5
conductivity	28.3 mS/cm

2.2. Reactor set-up and operation steps

The EC-EO process was carried out at room temperature in a monopolar-bipolar EC-EO reactor made of a cylindrical glass cell with a monopolar configuration and anode electrodes made of Boron-doped diamond (BDD; Adamant Technologies, Switzerland) and cathode electrodes made of stainless steel (SS; AISI 304, Mervilab, Spain).The electrodes' effective surface area was 20 cm2. An Al plate was put in a bipolar configuration between the anode and the cathode. This plate is used as a bipolar electrode [32]. The electrodes were separated by 1 cm. The sample was introduced to the cell at a volume of 500 mL. A DC power supply (BK-Precision, 0–30 V, 0–5 A, Yorba Linda, California) working in galvanostatic mode was used to manage the current and voltage input. The first step of batch studies was carried out for 24 hours at a current flow of 0.4 to 1.0 A with no changes in the pH of the samples. The batch experiments were completed in the second step with a current intensity of 0.7 A and pH constants of 4, 6, and 8. Experiments were carried out in the absence of a bipolar

aluminum electrode to assess the bipolar electrode effect and the individual role of EO in the combined EC-EO process. Electrochemical experiments were used to treat the samples at various intervals (0 to 24 hours), and the concentrations of composition effluent and leachate were evaluated following the treatment process. The electrodes were cleaned with 1 M H_2SO_4 for 1 hour and rinsed with deionized water multiple times after each experimental run. Removal efficiency (η) was determined by measuring concentration target parameters before (C_i) and after (C_f) treatment as per the following equation [33, 34]:

$$\eta\ (\%) = = \frac{C_i - C_f}{C_i} \times 100 \quad (1)$$

3. RESULTS AND DISCUSSION

3.1. Removal of pollutants using the EO and combined EC-EO process

Figure 1 demonstrates the removal effectiveness of COD, NH4+-N, Cr (VI), and TN after 24 hours of EC-EO (presence of bipolar electrode) and EO (absence of bipolar electrode) therapy. Figure 1a shows that after 12 hours of treatment, removal efficiency in the presence of a bipolar electrode is 22% higher than in the absence of a bipolar electrode, and after 24 hours of EC-EO and EO therapy, COD removal efficiency are 98.5% and 97.8%, respectively. Figures 1b and 1c demonstrate that removal efficiency for NH_4^+-N, Cr (VI), and TN is 85.8%, 85.3%, and 62.2% after 24 hours of EC-EO treatment, and 52.5%, 58.5%, and 37.0% after 24 hours of EO treatment, respectively. Furthermore, due to the bipolar configuration of the aluminum electrode in the combined EC-EO process, the removal rate is improved. The BDD electrode as anode has been shown to have a very high oxygen evolution over-potential in the combined EC-EO process, and it has shown great efficiency for mineralization of various organic pollutants because it can oxidize organic pollutants present in the water medium until carbon dioxide, water, and minerals are formed by forming surface adsorbed hydroxyl radicals [35, 36]. Researches have been indicated that BDD electrode as anode can oxidize the chloride ions in wastewater to form active and powerful chlorine oxidants (Cl_2, HOCl and OCl^-) which can act as a mediator for oxidation of organic compounds [37, 38]. Furthermore, when an electric current is passed through the cathode and anode electrodes, the bipolar configuration of the Al electrode causes it to carry both positive and negative charges on opposing faces, as well as become polarized and induce positive and negative charges on opposing faces [39, 40]. As a result, the intake on the side of the bipolar electrode facing the SS electrode acted as an anode, causing negative surface charges, while the inlet on the side facing the BBD electrode served as a cathode, causing positive surface charges [41, 42]. At the beginning of the treatment process, Al ions were produced by anodic dissolution from the anodic side of the bipolar electrode as the following reactions [43].
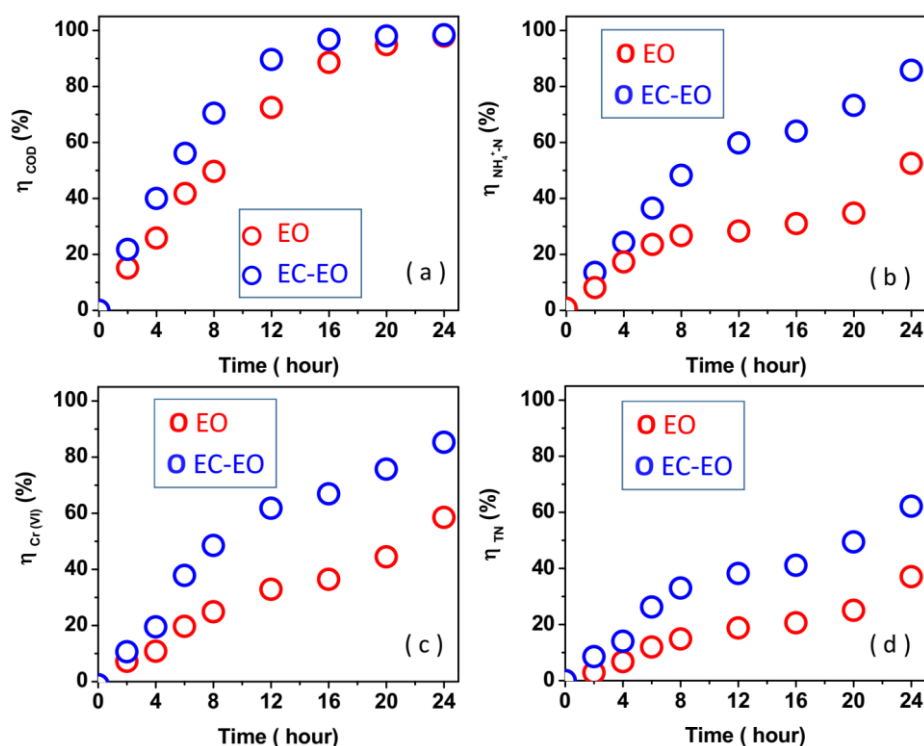
$$Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3e^- \qquad (2)$$

$$3H_2O_{(l)} + 3\ e^- \rightarrow \frac{3}{2}H_2 + 3H^+ \qquad (3)$$
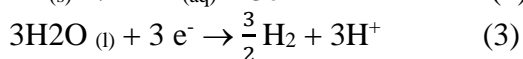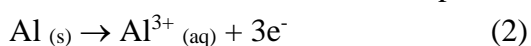
Figure 1. (a) COD, (b) NH_4^+-N, (c) Cr (VI) and (d) TN removal efficiency under 24 hours EC-EO (presence of bipolar electrode) and EO (absence of bipolar electrode) treatment processes at current intensity of 0.7 A and without any adjustment in pH.

It is suggested that the optimal removal of pollutant through adsorption and charge neutralization is associated with the effect of hydrolysis and polymerization of Al ions by forming gelatinous charged hydroxo-cationic complexes such as $Al(H_2O)_6^{3+}$, $Al(H_2O)_5(OH)_2^{+}$, $Al(H_2O)(OH)^{2+}$, $Al_2(OH)_2^{4+}$, $Al(OH)^{4-}$, $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$, and $Al_{13}(OH)_{34}^{5+}$, etc [43-45]. To cause coagulation, these hydroxides/polyhydroxides/polyhydroxymetallic compounds have a high attraction for scattered particles as well as counter ions [46]. The reduction of H^+ and NO^{3-} produces hydrogen and nitrogen gas bubbles at the cathodic poles of bipolar electrodes [47, 48]. As a result, oxygen and hydrogen gases are produced at the anode and cathode in the form of gas bubble nucleates [46], causing the absorbents to float [49]. For the simultaneous removal of residual organic and nitrogenous pollutants from bio-treated landfill leachate, an integrated system with different electrode arrangements, including bipolar and monopolar electrodes in one reactor, and higher simultaneous removal efficiency through EO and EC was achieved [41, 50], which is essential for the further industrial application of electrochemical processes.

As a result, it's possible that the bipolar Al electrode, the formation of aluminum hydroxide flocs, and the structure of aluminum fluoride hydroxide complexes, which can trap organic and inorganic compounds in leachate through floatation and adsorption mechanisms, are linked to improved pollutant removal efficiency in the EC-EO process [51, 52]. During EO treatment, no visible flocs are noticed. However, visible flocs are generated during EC-EO treatment. The NO_3–N content in the reactor is continuously increased during the EO process due to continued oxidation of NH_4^+-N, but during the EC-EO process, the presence of a bipolar Al electrode leads to further anodic oxidation of

NH_4^+-N and, as a result, more NO_3–N production, which is adsorbed by aluminum hydroxide flocks [53, 54].

3.2. Current density effect on pollutant removal efficiency in the combined EC-EO process

In the combined EC-EO process, Figure 2 depicts the current density influence on pollutant removal efficiency. As can be seen, raising the current intensity improves removal efficiency. After 16 hours in the combined EC-EO process, more than 97%, 63%, 62%, and 40% of COD, NH_4^+-N, Cr (VI), and TN were removed from solution for I > 0.5 A, respectively. The enhancement of nitrogenous chemical oxidation is linked to an increase in the formation rate of hydroxyl radicals with current on the surface of the BDD electrode [55]. Furthermore, as the current density rises, the charge density on the opposing surfaces of the bipolar electrodes rises, the anode dissolution rate rises, leading to a rise in the number of metal hydroxide flocs and a rise in pollutant removal efficiency [56]. The denitrification process is also shown to be regulated by an electric current [57].
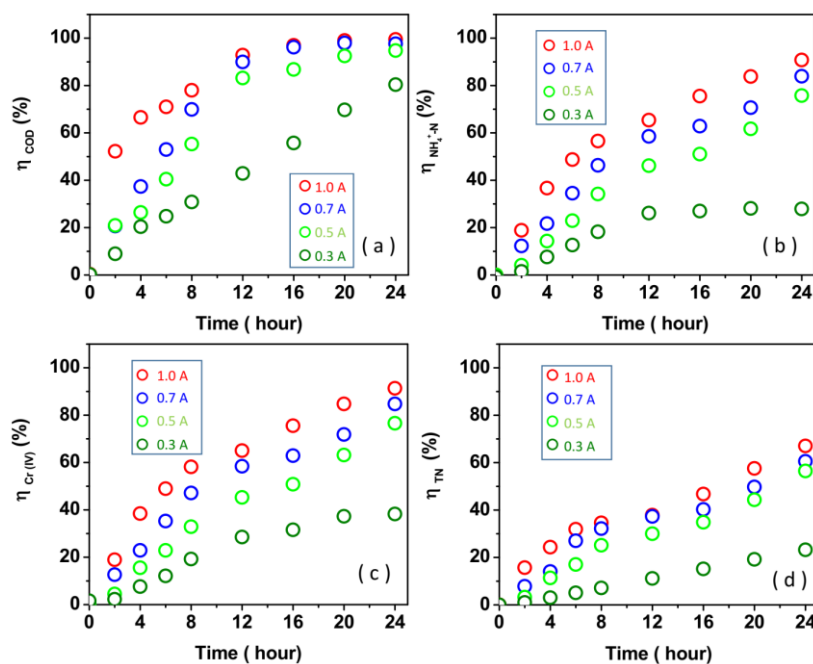

Figure 2. Current density effect (0.3, 0.5 , 0.7 and 1.0 A) on (a) COD, (b) NH_4^+-N, (c) Cr (VI) and (d) TN removal efficiency in the combined EC-EO process without any adjustment in pH.

3.3. pH effect on pollutant removal efficiency in the combined EC-EO process

Theoretically, acidic and alkaline mediums can be beneficial for the treatment of organic pollutants. At acidic pH, decrease of pH results in low concentrations of carbonate species HCO_3^- and CO_3^{2-}, which are scavengers of free radicals. $^\bullet OH$ generated on anode, as consequence enhances oxidation rate [58], while at alkaline pH, the $Cl^- \rightarrow Cl_2 \rightarrow ClO^- \rightarrow Cl^-$ redox cycle is promoted which enhances indirect oxidation [58, 59]. The highest COD removal efficiencies have been reported under

acidic conditions in the pH range of 4.0–6.0, whereas extremely poor removals have been reported at lower pH values (pH 3) due to the amphoteric feature of $Al(OH)_3$, which does not precipitate at very low pH [59, 60]. On the other hand, the increase $Al(OH)_3$ solubility and formation of the $Al(OH)_4^-$ and AlO_2^- compounds in high pH values (pH ≥ 9) is ineffective to treatment of the leachate [58, 59]. The effect of neutral or acidic conditions on reduced electrical energy usage during EC and EO wastewater treatment has been demonstrated in research [61]. Therefore, starting pH values were done in the range of 4.0–8.0 at a current intensity of 0.7 A in the current work to investigate the effects of pH effect on leachate treatment. The initial pH effect on pollutant removal efficiency in the combined EC-EO process is depicted in Figure 3. As observed, removal efficiencies are increased with decreasing the initial pH value to 4 and 6.5, and more than 90% treatment of COD, NH_4^+-N, Cr (VI) and TN is obtained in less than 16 hours in the combined EC-EO process. The removal efficiencies of the initial pH value to 4 and 6.5 are very close, and by considering the pH=6.5 of the original effluent of leachate, pH=6.5 was selected as the optimal pH value for the combined EC-EO process.

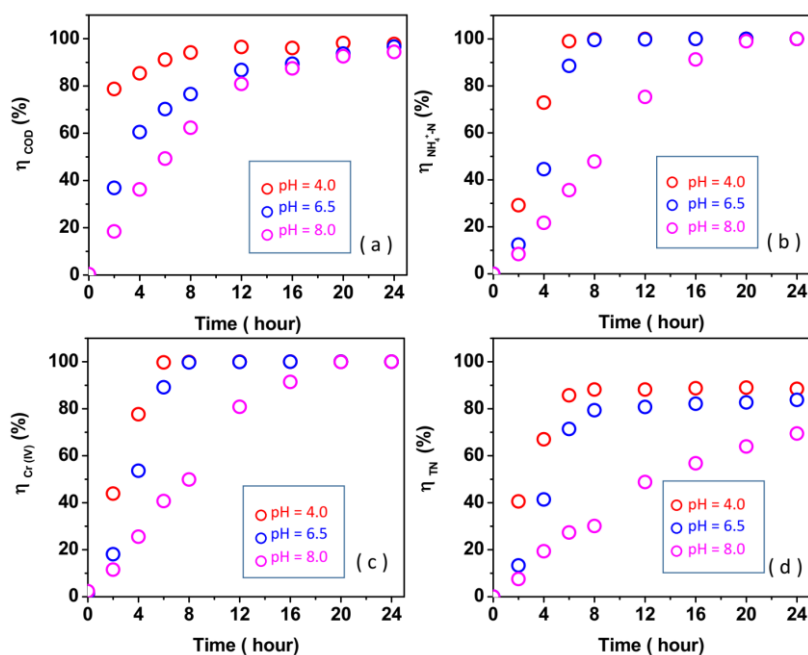**Figure 3.** Initial pH effect (0.4, 6.5 and 8.0) on (a) COD, (b) NH_4^+-N, (c) Cr (VI) and (d) TN removal efficiency in the combined EC-EO process at a current intensity of 0.7 A.

3.4. Energy consumption

The specific energy consumption (SEC) of removed pollutant is defined as the amount of electrical energy consumed per unit mass of pollutant according following equation [62, 63]:

$$SEC \ (kWh/kg) = \frac{U \, I t}{V \, \Delta p} \quad (4)$$

Where U(V) is the applied voltage, I (A) is the current intensity, t (hour) is the experimental time, and V (L) is the volume of the treated wastewater and Δp (kg/L) is the differential pollutant.

Figures 4a and 4b show the SEC in the EO and combined EC-EO processes for COD and NH_4^+-N removal, respectively, at 0.7 A current intensity and no pH adjustment for COD and NH_4^+-N removal, indicating that the integration of EO with the EC process improves overall performance but results in aluminum electrode dissolution in the system. For COD in Figure 4a, the EC-EO process consumed 20kWh/kg and 50kWh/kg for 55% and 98% removal of COD, respectively, and the EO process consumed 52kWh/kg for 98% COD removal. It is observed from Figure 4b that the EC-EO process consumed 420 kWh/kg and 847kWh/kg for 50% and 85% removal of NH_4^+-N, respectively, and the EO process consumed 1250 kWh/kg for 50% NH_4^+-N removal. The EO process alone did not allow for 52% treatment of NH_4^+-N after 24 hours.
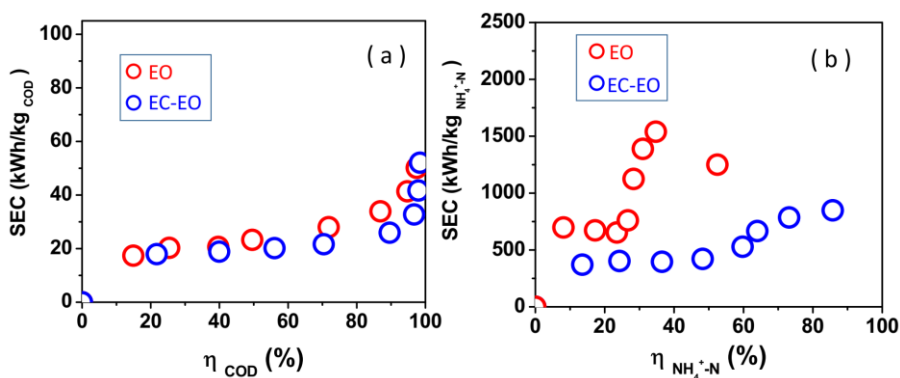

Figure 4. SEC in the EO and combined EC-EO processes for removal of (a) COD and (b) NH_4^+-N at current intensity of 0.7 A and without any adjustment in pH.

Figure 6 also illustrates the SEC in a combined EC-EO process for COD and NH_4^+-N removal at current intensities of 0.3, 0.5, 0.7, and 1.0 A, with no pH modification. Figure 5a shows that after 24 hours, COD removal efficiencies of 99% and 98% are achieved at current intensities of 1.0 A and 0.7 A, respectively. SEC per kg COD removed at a current intensity of 0.7 A is significantly lower than SEC per kg COD removed at a current intensity of 1.0 A for removal efficiencies greater than 70%. After 24 hours, the lower removal efficiency (81%) is obtained at a lower current intensity (0.3 A).

Figure 5b also shows that after 24 hours of operation at current intensities of 0.7 A and 0.5 A, high NH^{4+}-N removal efficiencies of 85 percent and 76 percent, respectively, are possible. While SEC per kg of NH_4^+-N removed at current intensities of 0.7 A and 0.5 A is lower than that at 1.0 A, SEC per kg of NH_4^+-N removed at current intensities of 1.0 A is higher. At a current intensity of 1.0 A, good removal efficiencies for NH_4^+-N (93%) were attained, but the SEC per kg NH_4^+-N removed was roughly 1.45-fold higher than at a current intensity of 0.7 A. Because it can only provide gentle mixing for the EC-EO process, the lower elimination efficiency ($\leq$ 28%) is obtained at a lower current intensity (0.3 A) after 24 hours, showing that the NH_4^+-N was not the primary target of the oxidation mechanism. According to studies, both activities use ions, and as a result, as time passes and active ion removal increases, electrolyte activity declines. As a result, for the same fixed current density, a greater voltage is required, implying increased energy consumption [64, 65]. Table 2 shows a comparison of the developed treatment method with other reported reactors in the literature, implying

that the developed reactor performs similarly to some reported reactors, if not better. This can be attributed to the well-organized and efficient activity of electrodes in the presented EC-EO reactor, as well as optimal oxidation in the presence of oxidant species in solution.
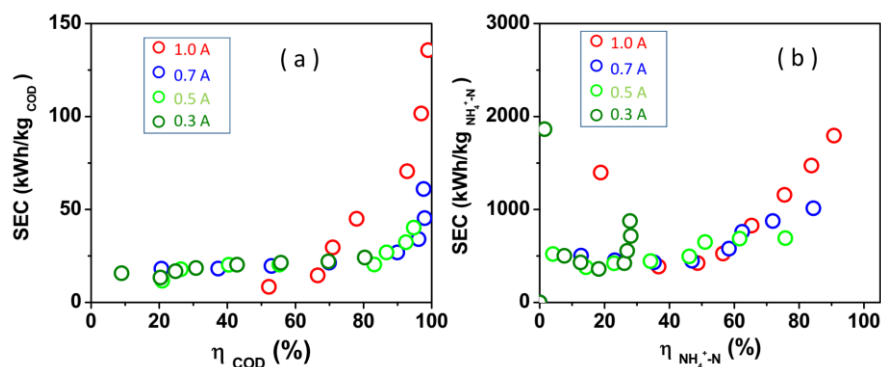

Figure 5. SEC in combined EC-EO processes for removal of (a) NH_4^+-N and (b) COD at current intensity of 0.3, 0.5, 0.7 and 1.0 A and pH= 6.5.

Table 2. A comparison of the as developed treatment method with the other reported reactors in the literatures

Reactor	Electrodes arrangement	Target	Energy consumption (kWh/ kg)	Applied voltage or current	η(%)	Ref.
EC-EO	BDD anode, SS cathode, and Al bipolar	1150 mg/L COD	61	0.7 A	98	This study
		750 mg/L NH_4^+-N	691	0.5 A	75	
EC-EO	RuO_2-IrO_2-Ti anode graphite cathode, and Iron bipolar	200 mg/L NH_4^+-N	622	10 V	98.84	[66]
EC-EO	RuO_2-IrO_2-Ti anode, graphite felt/titanium cathode, and Iron bipolar	372 mg/L COD	420	---	64	[47]
EO	DSA® anode and DSA® cathode	3937 mg/L COD	9034	83 mA/cm^2	87	[67]
EC-EO	IrO2-Ti anode, Al cathode, and Al- Al bipolar	25 mg/L NH_4^+-N	428	30 mA/cm^2	100	[68]
		75 mg/L NH_4^+-N	1006			

4. CONCLUSION

This work used a combination EC-EO technique with bipolar and monopolar electrodes arranged in one reactor, with BDD as anode, SS as cathode, and Al as a bipolar electrode to electrochemically treat solid waste leachate by efficiently removing COD, nitrogen-based compounds, and chromium. The following are some of the most important findings:

(i)   The removal efficiency in the EC-EO reactor is 22% higher than that of the EO reactor after 12 hours of treatment, and 98.5% and 97.8% COD removal efficiencies were obtained after 24 hours of EC-EO and EO treatment, respectively. NH_4^+-N, Cr (VI) and TN removal efficiency showed 85.8%, 85.3% and 62.2% removal efficiency for NH_4^+-N, Cr (VI) and TN after 24 hours of EC-EO treatment at 0.7 A current intensity. The results indicated that in terms of energy consumption, the integration of EO with the EC process improved the overall performance. The EC-EO process consumed 20 kWh/kg and 50 kWh/kg for 55% and 98% removal of COD, respectively, and consumed 420 kWh/kg and 847 kWh/kg for 50% and 85% removal of NH_4^+-N, respectively.

(ii)   The study of the current density effect on pollutant removal efficiency in the combined EC-EO process showed that removal efficiencies were increased with increasing the current intensity. After 16 hours of combined EC-EO treatment for I > 0.5 A, more than 97%, 63%, 62%, and 40% of COD, NH^{4+}-N, Cr (VI), and TN were removed from solution, respectively. It was associated with the increase in generation rate of hydroxyl radicals with current on the surface of the BDD electrode that hydroxyl radicals enhance the oxidation of nitrogenous compounds.

(iii)   The pH effect of the study also revealed the beneficial effect of neutral or acidic conditions for great treatment during the EC-EO process, and removal efficiencies are increased by lowering the initial pH value to 4 and 6.5, and more than 90% treatment of COD, NH_4^+-N, Cr (VI), and TN was obtained in less than 16 hours in the combined EC-EO process, and pH=6.5 was chosen as the optimal pH value for the combined EC-EO process..

(iv)   A comparison of the developed treatment method with other reported reactors in the literature revealed that the developed reactor performed similarly to some reported reactors, if not better. This can be attributed to the well-organized and efficient activity of electrodes in the presented EC-EO reactor, as well as optimal oxidation in the presence of oxidant species in solution, which is necessary for further industrial development.

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