

The Performance of Electrocoagulation Process in Removing Organic and Nitrogenous Compounds from Landfill Leachate in a Three-Compartment Reactor

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ABSTRACT

In this study, the effectiveness of the electrocoagulation (EC) process was evaluated based on the reduction of organic and nitrogenous contaminants in landfill leachate. A three-compartment electrochemical reactor as pre-treatment of stabilized landfill leachate was carried out ahead of biological treatment. The removal efficiencies of COD, BOD, ammonia, and nitrate were analyzed at pH 4, 6, and 8 with the current densities of 20.83 and 29.17 mA·cm⁻². At pH 4, the highest removal of COD and NH₄⁺ was obtained, i.e., in the range of 72–81% and 43–59%, respectively. The ratio of BOD₅/COD was increased after EC, from initially 0.11 to 0.32 at pH 4. In addition, EC effectively removed humic substances in the leachate by targeting a large amount of high molecular weight humic substances, with around 10³ kDa. However, the higher removal efficiency observed at higher current density leads to higher specific energy consumption. At a current density of 29.17 mA·cm⁻², the specific energy consumption obtained in EC was around 10–17 Wh·g⁻¹ COD and 99–148 Wh·g⁻¹ NH₄⁺. This could be decreased up to 50% at an applied current density of 20.83 mA·cm⁻² with slightly lower efficiencies.

Keywords: current densities, electrocoagulation, humic substances, stabilized leachate, wastewater treatment

INTRODUCTION

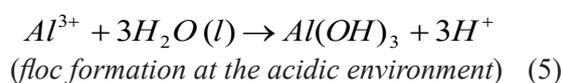
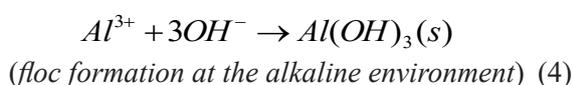
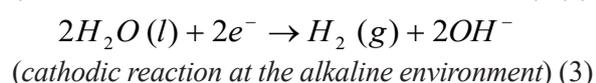
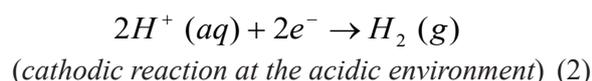
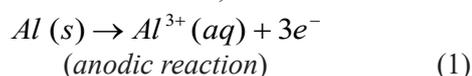
At prolonged operational time, the leachate wastewater generated from municipal solid waste landfill becomes more stabilized (mature) that poses more challenging treatment scenario to meet the stringent effluent standard. The complex properties in the stabilized leachate, such as smaller BOD₅/COD ratio and higher concentration of ammoniacal nitrogen and other refractory organic compounds, are difficult to degrade by biological processes alone (Deng and Englehardt 2007). Thus, it requires physicochemical

processes that can be applied as a pre-treatment or post-treatment to achieve more effective and efficient treatment train.

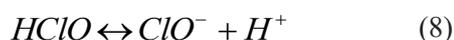
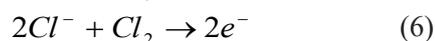
Electrocoagulation (EC) is one of the physicochemical processes that has proven to be effective in the removal of colloids, suspended solids, and high molecular weight organic compounds contained in wastewater, including landfill leachate (Dia et al. 2017). Moreover, it also has low sludge production, low carbon footprint, low salinity of effluent, and can enhance biodegradability index (Fernandes et al. 2014; Dia et al. 2017). Several factors, including

the material and shape of sacrificial anodes, pH, current density, electrode spacing, conductivity, current density, electrolysis time, and power consumption, may influence the EC performance (Galvão et al. 2020; Guo et al. 2022). Aluminum and iron electrodes are commonly used due to their low cost and availability. However, in the landfill leachate treatment, the Al electrode showed a higher removal of COD, color, turbidity, and total nitrogen compared to iron (Bouhezila et al. 2011). Previous studies reported the removal of approximately 80% of color and turbidity, 90% BOD, and between 50–65% COD using Al electrode (Ricordel and Djelal 2014; Pirsaneh et al. 2016; Galvão et al. 2020). Nevertheless, in some cases, the removal of nitrogenous compounds in the leachate was relatively difficult in the EC. Other studies reported only 40% of nitrate removal by EC with Al anode (Ricordel and Djelal 2014) and 38.6% of ammonia nitrogen removal using Fe anode (Li et al. 2011).

In the EC process, coagulants are generated in situ by an electrolytic process of sacrificial anodes, allowing destabilization of suspended particles, dissolved or emulsified pollutants to form flocs (Fernandes et al. 2015). The overall mechanism of pollutant removal through the floc formation can be explained as follows (Ilhan et al. 2008; Fernandes et al. 2015).



In the presence of high salinity, the removal of pollutant was significantly enhanced through the formation of strong oxidants (Cotillas et al. 2013; Galvão et al. 2020).



The EC process was mostly performed in a single compartment reactor. The application

of membrane-divided electrochemical reactor offers enhanced removal of ionic compounds (Bagastyo et al. 2020). However, there is a lack of knowledge regarding the increasing performance of the EC process particularly in a three-compartment reactor. Thus, this study aims to evaluate the effectiveness of the EC process in a three-compartment reactor combined with simultaneous ion removal through electrodialysis process in removing organic and nitrogenous compounds from the landfill leachate. The removal of dissolved organic matter (DOM) was also investigated through spectrophotometric analysis and molecular weight distribution.

MATERIALS AND METHODS

Landfill leachate sample

The stabilized leachate was taken from a local municipal solid waste (MSW) landfill that has been operated since 2002. Samples were collected in plastic containers and immediately stored in the refrigerators. Prior to the experiment, discrete solid particles were removed from the sample. Table 1 depicts the characteristics of the leachate.

Experimental setup

The experimental reactor was made of acrylic and consists of three compartments. The anode and cathode compartments have a dimension of 18×8×2 cm (288 mL) each, while the central compartment was 18×8×1 cm. The ion exchange membranes, i.e., anion exchange membrane (AEM) AMI-7001 type and cation exchange membrane (CEM) CMI-7000 type supplied by Membrane International Inc., USA were inserted in the reactor. Al and stainless steel plate (SS 404) electrodes were applied as the anode and cathode, respectively. The electrodes have a rectangular shape with 3×8 cm dimension and an inter-electrode distance of 2 cm. Figure 1 illustrates the schematic experimental setup.

The experiments were performed in a batch system. A total volume of 5 L of leachate sample and 5 L of buffer solution were continuously recirculated through the EC reactor for 6 hours using peristaltic pumps. The leachate was pumped at a flow rate of 5 mL s⁻¹ from the feed tank to the cathodic compartment and flowed to the anodic compartment. Before circulating back to the feed

Table 1. Initial leachate characteristics

Parameter	Unit	Concentration
pH	–	8.42 ± 0.07
TDS	mg L ⁻¹	3160 ± 164
COD	mgO ₂ L ⁻¹	1552 ± 65
BOD ₅	mgO ₂ L ⁻¹	157 ± 12
BOD ₅ /COD	–	0.11 ± 0.02
Total N	mg L ⁻¹	1280 ± 73
NH ₄ ⁺	mg L ⁻¹	202 ± 17
NO ₃ ⁻	mg L ⁻¹	12.5 ± 1.4

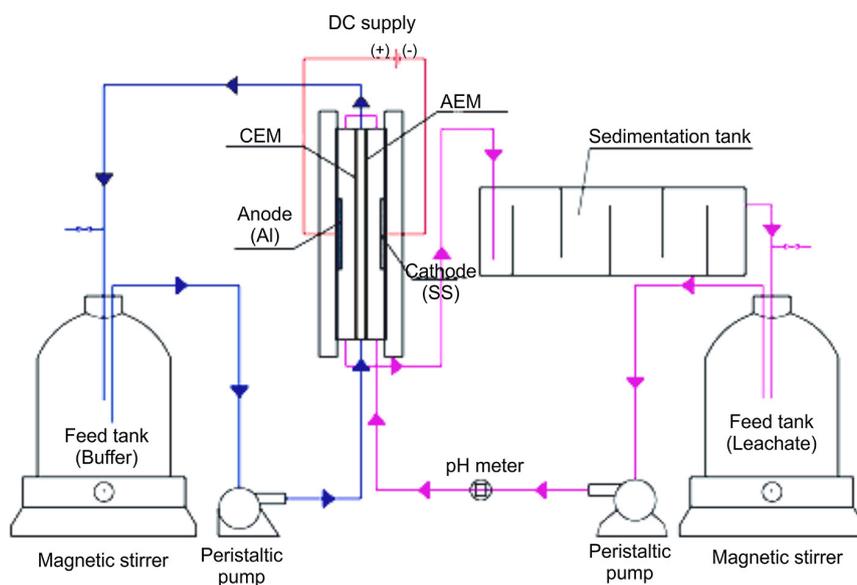
tank, it passed through the baffled sedimentation tank. The applied current of 0.5 and 0.7 A was constantly supplied by an adjustable DC power supply (3005DT, Cody, China). The initial solution pH was adjusted to pH 4, 6, and 8 using 4 N of NaOH and H₂SO₄. Leachate was magnetically stirred at a speed of 200–400 rpm during the experiments.

Analytical methods

Samples were collected from the cathode and central compartments and then immediately filtered using a 0.45 μm PTFE membrane syringe filter (ANPEL Technology, Shanghai, Inc). The pH measurement and the analyses of BOD₅, COD, ammonium, nitrate, and TDS were conducted each hour. Meanwhile, the analysis of TOC was conducted every two hours. Each analysis was carried out based on the Standard Method (American Public Health Association 2005). A handheld pH meter unit (YK-2005 WA, Lutron, Taiwan)

was used to measure pH, DO, and total dissolved solid (TDS). The concentration of BOD₅ was determined using the titrimetric method (Winkler analysis), whereas the concentration of COD was carried out using the colorimetric method (closed reflux, Merck and Spectroquant NOVA). The concentration of ammonium and nitrate was analyzed according to Nessler Method using a UV-Vis spectrophotometer (Genesys 20, Thermo Fisher Scientific, USA), respectively. The argentometric method was applied to measure the concentration of chloride ions in the solution. At the same time, a Shimadzu TOC-N analyzer (TOC-N 5000A, Shimadzu Scientific Instrument Inc., Japan) was used to determine the concentration of TOC and TN.

The TOC concentration of the samples was kept at 5 mg·L⁻¹ for DOM analyses. The measurement of DOM was conducted only for the final sample using a fluorescence excitation-emission matrix (F-EEM) (RF-6000 fluorescence spectroscopy, Shimadzu, Japan) according to the previous study (Lin et al. 2021). The average fluorescence intensity (AFI) measurement of DOM represents the summation of intensity in 4 fluorophore regions, i.e., soluble microbial products-like (SMPL), humic acid-like (HAL), fulvic acid-like (FAL), and aromatic protein-like (APL) substances. The concentration of dissolved organic species in the solution was performed using high-performance liquid chromatography-size exclusion chromatography (LC-20 ATV HPLC-SEC, Shimadzu, Japan) equipped with organic carbon detector (OCD, modified Sievers Total Organic

**Figure 1.** Schematic reactor configuration of the EC process

Carbon Analyzer 900 Turbo, GE Water and Process Technologies, USA) and ultraviolet detector (UVD, SPD-20A UV-Vis detector, Shimadzu, Japan). The setting measurement was following the earlier study (Lin and Ika 2020).

RESULTS AND DISCUSSIONS

Removal of COD and BOD₅

The removal of COD and BOD₅ during the EC process was evaluated based on initial pH and current density variations. As shown in Figure 2, COD removal efficiencies at pH 8 and under current densities of 20.83 and 29.17 mA cm⁻² were 56 and 71%, respectively. At pH 6 and 4, when the current density was adjusted to 29.17 mA cm⁻², the removal percentages rose gradually to 75 and 81%, respectively. Interestingly, COD removal (74%) was significantly increased at 20.83 mA cm⁻² when pH was decreased to 6 but slightly dropped to 72% at pH 4. Within the first 60 min of the EC process, COD removal was sharply increased between 32–40% under pH 4. By contrast, the removal of COD was only 5–15% at pH 6 and between 7–21% when pH was increased to 8. At this initial state, under all pH, a higher rate of removals was observed in higher current density. These results clarify that pH and current density strongly affect the EC process. A study performed by Apaydin and Özkan (2020) stated that an acidic environment is favorable for COD removal than in neutral or alkaline conditions. At pH 5, the removal of COD was 65% higher than at pH 7. Additionally, the removal efficiency of soluble COD was

increased by 31% compared to pH 7. Furthermore, another study also reported a similar result of immediate removal of COD at the beginning of the experiment, particularly when the current density was increased (Lu et al. 1999).

The effects of current density and pH on the BOD₅/COD ratio are presented in Figure 3. The highest BOD₅ reduction rate, i.e., approximately 65%, was obtained at pH 6 in both applied current densities (20.83 and 29.17 mA cm⁻²) after 0.98 Ah L⁻¹. The effect of increasing current densities on BOD removal was clearly observed at pH 4–6. In addition, EC process seems to be more effective to remove highly concentrated biodegradable compounds. For instance, higher BOD removal of around 90% was reported in the applied current density of 12.89–16.11 mA cm⁻² for 90 mins to remove initial BOD concentration of 923 mg L⁻¹ (Galvão et al. 2020).

Overall, the increase in BOD₅/COD ratio was observed at all pHs and current densities for the EC process. The increase was identified at pH 4, i.e., after 0.62 Ah L⁻¹, from initially 0.11 to approximately 0.24–0.32. However, at the end of EC (0.98 Ah L⁻¹), a smaller biodegradability index was obtained, i.e., 0.20–0.26. Likewise, a similar BOD₅/COD ratio profile was observed at pH 6 (with a lower index, 0.14–0.22), which might be due to the generated chlorine species involved in the COD removal during treatment. Meanwhile, at pH 8, the increase in BOD₅/COD ratio seemed to be in line with the prolonged time.

The pH influence in the EC process is mainly caused by controlling the Al speciation and its solubility via hydrolysis and polymerization

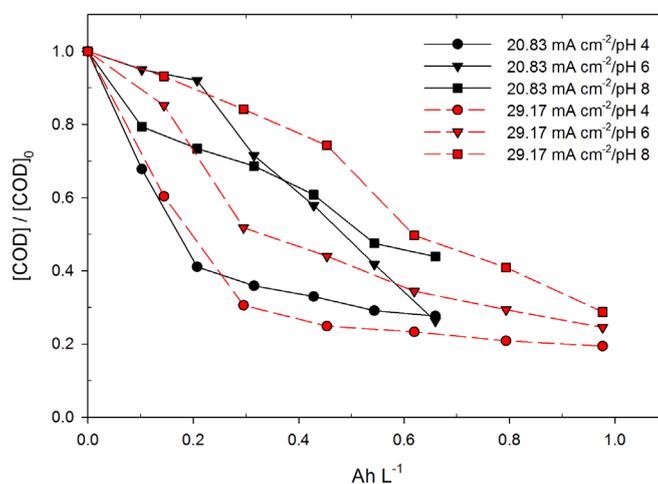


Figure 2. Removal of COD during Al-based EC process at 20.83 and 29.17 mA cm⁻² from pH 4 to 8

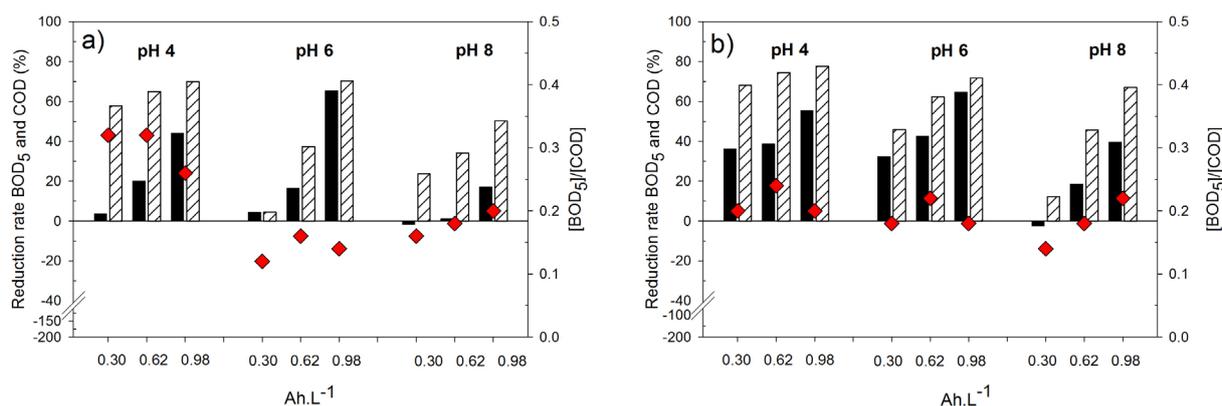


Figure 3. The ratio of BOD_5/COD and reduction rate of BOD_5 and COD from pH 4 to 8 at (a) 20.83 mA cm^{-2} ; (b) 29.17 mA cm^{-2} .

(Note: dark bar, BOD_5 ; shaded bar, COD ; red diamond, BOD_5/COD)

reaction of Al^{3+} . Theoretically, $Al_3(OH)_4^{5+}$, $Al_{13}(OH)_{32}^{7+}$, $Al(OH)_2^+$, $Al(OH)_2^+$ are existed at pH 3–5, whereas $Al(OH)_3$, which is preferred for the coagulation process, is dominantly formed in the pH 5–7 (Lu et al. 1999; Lin et al. 2014). As the pH increased (>7), the concentration of $Al(OH)_3$ decreased and turned into $Al(OH)_4^-$ (Lu et al. 1999). Nevertheless, in this study, the highest removal of COD and BOD were achieved at pH 4. According to the literature, electrocoagulation has two primary mechanisms for removing dissolved organic matter (Cañizares et al. 2006). The first is through the binding of metal species to anionic sites of the organic molecules, neutralizing their charge and resulting in reduced solubility; the second is the adsorption of organic matter on the amorphous metal hydroxide precipitate. These mechanisms are pH dependent. Therefore, the removal of BOD and COD did not solely depend on $Al(OH)_3$ formation.

Noteworthy, current density also plays an important role in EC. The increasing of current density will amplify reaction rate, hydroxide concentration, the generation rate of H_2 gas, mass transfer at the electrodes, and growth of flocs, resulting in the rapid coagulation and flotation of coagulated pollutants (Liu et al. 2010; Fernandes et al. 2015; Apaydin and Özkan 2020). Nonetheless, the increasing current density may raise pH level by enhancing hydroxyl ions formation in the cathode (Ricordel and Djelal 2014; Fernandes et al. 2015), thus reducing its ability to remove the pollutants.

Removal of TOC and TN

In the present study, TOC was evaluated to quantitatively measure organic carbon in the leachate after the EC process. To reach the optimum result, TOC and TN analyses were performed for 8 h at pH 4 and 29.17 mA cm^{-2} . Figure 4 depicts

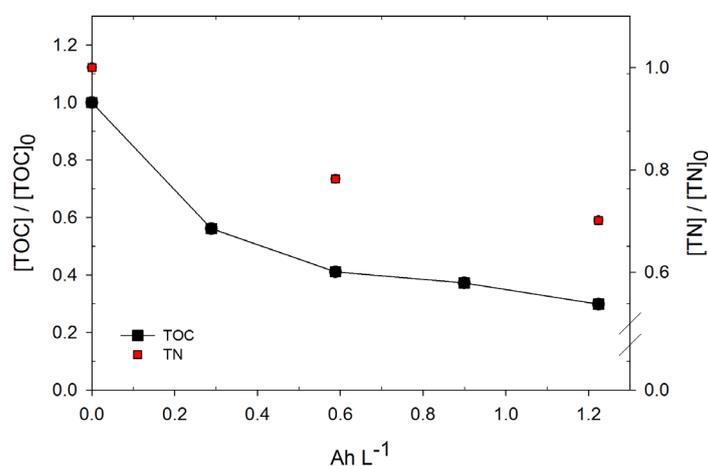
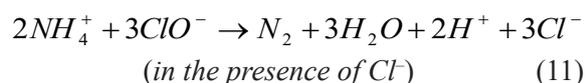
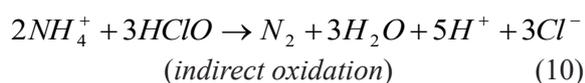
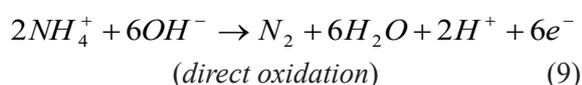


Figure 4. Removal of TOC and TN at pH 4 and current density of 29.17 mA cm^{-2}

TOC and TN removal in the solution. According to the graph, the removal of TOC through the EC process was quite high, i.e., 70%.

In contrast, the obtained TN removal was 29.8%, slightly lower than the previous study (Nanayakkara et al. 2018), which acquired 32.66% of TN removal. The TN removal is due to the electrochemical oxidation and reduction that possibly occurred simultaneously in the anode and cathode compartments, respectively. Apart from entrapment by aluminum floc formed in the anodic compartment, the nitrogenous compound can also be removed through various oxidation and reduction reactions (Amarine et al. 2020). Eqs. (9)–(11) present some important reactions that could occur (Pirsaheb et al. 2016). The N_2 gas generated from the reactions leaves the system contributing to TN removal:



Removal and recovery of ammonium and nitrate

The removal (66.6%) and recovery (32.81%) of nitrate were achieved at medium pH under low current density. While for ammonium, the removals were better achieved at low to medium pH, and the recovery was better achieved at higher pH, under both current densities. In the EC process, nitrogenous compounds were removed through coagulation and indirect oxidation method (Murphy 1991). The coagulation mechanism, including charge neutralization and sweep flocculation, favors most nitrogenous compounds turned into sludge, and only small fractions can be recovered

(i.e., transferred into the central compartment by electro dialysis process).

The increased mass of ammonium and nitrate in the buffer solution was in conjunction with the decreasing mass of ammonium and nitrate in the raw leachate (remained). In addition, the mobility of ammonium and nitrate ions was also monitored by measuring TDS in each compartment (Fig. 5). As can be seen in Figure 5, current density seems to have more significant effects on TDS mass transfer into the central compartment than pH. It is confirmed that the increase of current density led to the increase of TDS removal and recovery. At 29.17 mA cm^{-2} , the maximum removal and recovery of TDS reached 49 and 60%, respectively. Regardless of pH and current densities variations, the average TDS recovered after the EC process was 1207 mg L^{-1} . This finding is quite smaller than the previous study, which obtained 78% of TDS removal (Bakry et al. 2018), probably due to higher TDS concentration in the feed water.

Identification of degraded organic pollutants

Dissolved organic matter (DOM) has a certain composition of organic matter that can be classified into HAL, FAL, and a hydrophilic fraction (Hyl) through their fluorescent intensity (Keyikoglu et al. 2021). Since the characteristics of DOM in young and mature leachate were different, the identification of DOM can be used to determine the age of the landfill. In the mature landfill leachate, SMPL and HAL were dominant than other fractions. Figure 6 presents the fluorescent intensities of DOM before and after EC treatment.

Before treatment, the maximum fluorescent intensity of SMP, HA, FA, and AP consists in the raw leachate were measured at Ex/Em of 320–330/320–330 nm (4059–19211 a.u.); 330/415–425 nm (899–940 a.u.); 250/450 nm (250–257 a.u.); and 240/380 nm (108–119 a.u.),

Table 2. Removal and recovery of ammonium and nitrate

Current density (mA cm^{-2})	pH	NH_4^+			NO_3^-		
		Removed (%)	Recovered (%)	Remained (%)	Removed (%)	Recovered (%)	Remained (%)
20.83	4	58.60	13.85	27.55	65.74	31.96	2.30
	6	56.16	8.57	35.27	66.60	32.81	0.59
	8	43.38	20.68	35.94	36.99	32.53	30.48
29.17	4	43.12	12.28	44.60	68.90	30.88	0.22
	6	50.42	12.61	36.97	31.61	27.29	41.10
	8	47.63	20.23	32.14	44.93	44.36	10.71

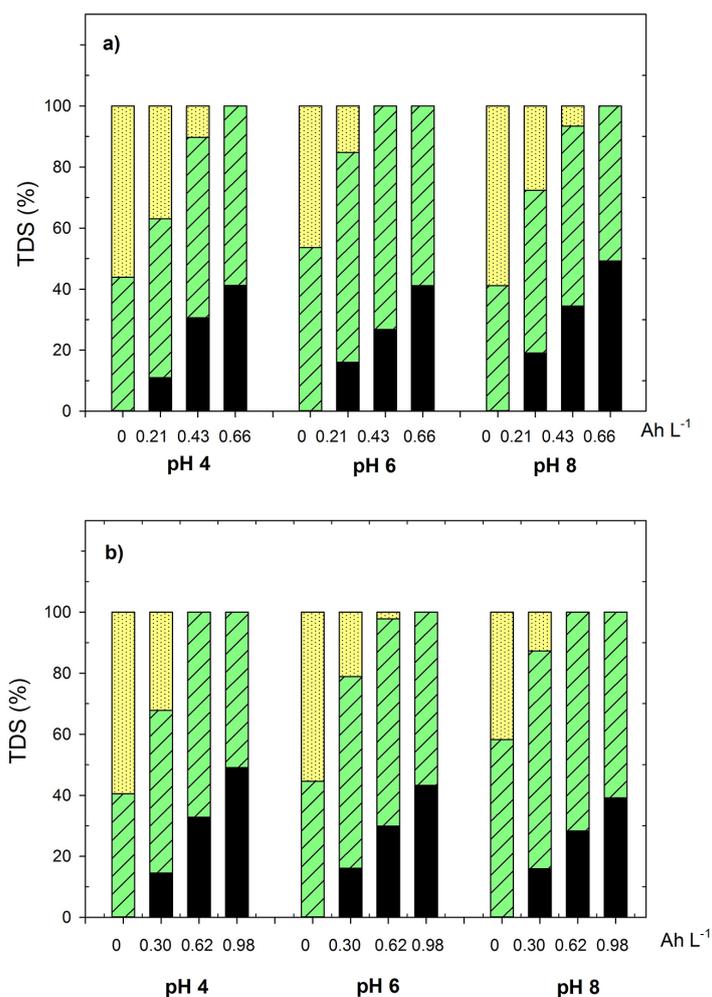


Figure 5. TDS removal after EC treatment at a) 20.83 mA cm⁻² and b) 29.17 mA cm⁻² (Note: dark bar, removed; green shaded bar, recovered; yellow dotted bar, remained)

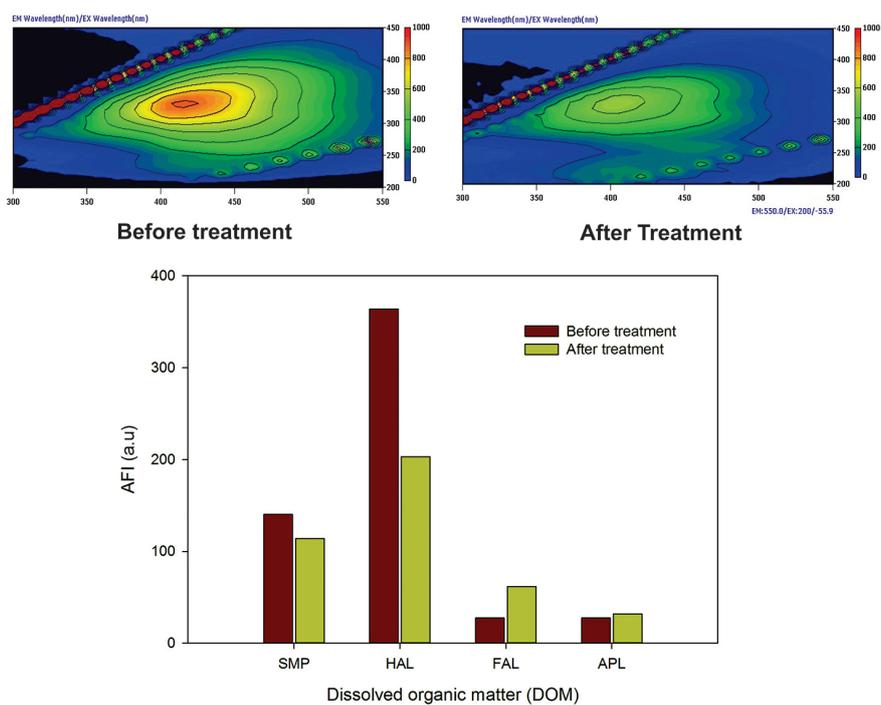


Figure 6. Fluorophore intensities of DOM in leachate before and after treatment of 8 h

respectively. The high intensity of SMP and humic substances are consistent with stabilized and mature leachate. After the EC process, the fluorophore intensity decreased to 1859 a.u. (at 330/330 nm for SMP), 566 a.u. (at 330/415 nm for HA), 215 a.u. (at 250/455 nm for FA), and 180 a.u. (at 220/380 nm for FA). The most significant AFI reduction occurred in the HAL fraction of 44.14%, SMP decreased by 18.8%, followed by a slight increase in the APL and FAL fractions of 15.29 and 18.45%, respectively. This result indicates that EC was effective for fluorescent DOM removal from the leachate. The effective removal of organic matter by the EC process was also confirmed in the several studies (Vepsäläinen et al. 2012; Särkkä et al. 2015; Dia et al. 2017). With the application of the Al electrode, the removal of humic acid, fulvic acid, and hydrophilic compounds in the landfill leachate could reach 100, 60, and 46%, respectively (Dia et al. 2017).

Molecular weight distributions

In addition, the profiles of MW distribution in terms of DOM intensity before and after EC processes are provided in Figure 7. According to the graph, the MW distribution of DOM in the raw leachate can be divided into three colloidal fractions (i.e., peak 1 = 776 kDa; peak 2 = 207 kDa, and peak 3 = 112 kDa). As observed after EC treatment, a pollutant with MW > 300 kDa was completely disappeared, whereas a removal rate > 98% was detected for DOM > 250 kDa. Among the remained organic pollutants, fractions of 50–250 kDa were dominant. The highest fraction was monitored within the size of 113 kDa, with the $SUVA_{254}$ intensity increased by 34%, i.e., from initially 6577 a.u. to 8831 a.u. The decrease of DOM in high MW fraction and the increase of DOM in low MW fraction was in line with those obtained in the previous study (Palacios et al. 2016). It is reported that Al anode effectively

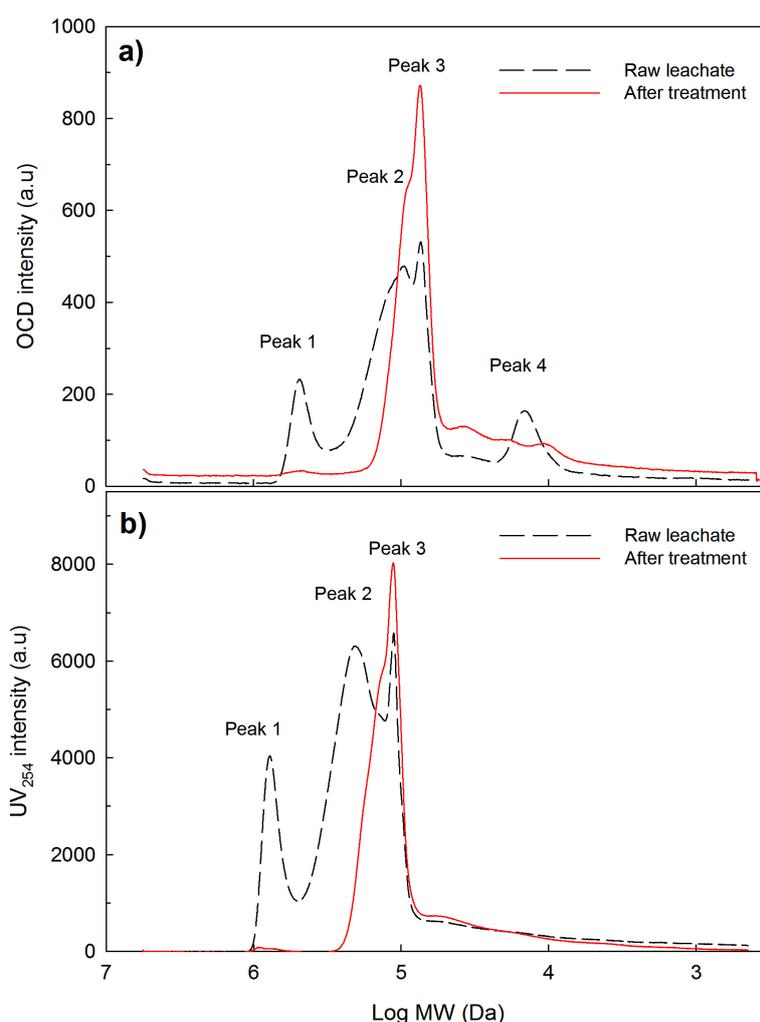


Figure 7. (a) OCD and (b) UV_{254} MW distribution of DOM during EC process

Table 3. Specific energy consumption for COD and NH₄ removal

I (A)	pH	Voltage	COD removed		NH ₄ ⁺ removed		Energy consumption	
		(V)	(%)	(g)	(%)	(g)	COD (Wh·g ⁻¹)	NH ₄ ⁺ (Wh·g ⁻¹)
0.7	4	14.77	81	5.93	59	0.59	10.47	105.33
	6	14.30	75	5.02	56	0.60	11.97	99.30
	8	18.84	71	4.57	43	0.53	17.31	148.23
0.5	4	11.59	72	4.70	43	0.46	7.40	75.88
	6	9.68	74	5.25	50	0.44	5.53	65.91
	8	11.41	56	3.79	48	0.55	9.02	62.05

removed DOM with large MW, and the oxidation of organic compounds was enhanced by the presence of active chlorine species in the leachate.

Energy requirements

The treatment efficiency was estimated according to the watt hour energy consumption per COD or NH₄ mass removed as described in the previous study (Bagastyo et al. 2021).

$$W(\text{Wh g}^{-1}) = \frac{I \times V \times t}{\Delta m} \quad (12)$$

where: W is the energy consumed, I is the average applied current (A), V is the average cell voltage (V), t is electrolysis time (h), and Δm is the mass removed (g).

The specific energy consumption for COD and NH₄ removal during the EC process is presented in Table 3.

Overall, energy consumption was increased with the rise of current density. To achieve maximum removal of COD in the EC process, the minimum energy required was 10.47 Wh g⁻¹ and accomplished at acidic pH and higher current density ($j = 29.17 \text{ mA cm}^{-2}$, $I = 0.7 \text{ A}$). Comparatively, a current density of 20.83 mA cm⁻² could remove a quite high COD (74%) with 50% less energy in a slightly neutral environment (pH 6). The increase of specific energy consumption with the rise of current density was also reported by Ilhan et al. (2008). They estimated that the energy consumption was 0.46 Wh g⁻¹ COD at a current density of 34.8 mA cm⁻². When the current density rose to 43.5 and 52.4 mA cm⁻², the energy consumption substantially increased by 45.6 and 93.5%, respectively.

In terms of ammonium removal, the maximum removal of NH₄⁺ in the EC process was also accomplished at acidic pH and higher current density. The energy of 105.33 Wh g⁻¹ was at least

required to remove 59% of NH₄⁺. While the minimum energy of 62.05 Wh g⁻¹ can remove 48% of NH₄⁺ in leachate. These results imply that EC was an efficient treatment for ammonia removal, performing the lowest energy consumption in all experiments tested (in the range of 65.91–75.88 Wh g⁻¹, at pH 4–6, 0.5 A).

CONCLUSIONS

EC process shows satisfactory removal of COD, BOD₅, TOC, NH₄⁺, TN, and humic substances with removal percentages in the range of 56–81, 17–65, 70, 43–59, 29.8, and 14–37%, respectively. In general, the effect of pH adjustment on the treatment performance is more pronounced than applied current density, where pH 4 was more favorable than pH 6 and 8. The increase in BOD₅/COD ratio during EC was observed at all current densities tested, from 0.11 to 0.32 at pH 4. In the EC process, more selective removal of DOM was observed, i.e., targeting high molecular weight DOM of around 10³ kDa, primarily HAL and FAL substances. At higher current density, although the removal of COD and NH₄⁺ was increased, the removed mass was not significant, and thus the current density of 20.83 mA cm⁻² (0.5 A) was more suggested.

The specific energy consumption for COD removal in the EC process was between 9.02–7.40 and 10.47–17.31 Wh g⁻¹ at a current density of 20.83 and 29.17 mA cm⁻², respectively. At the same time, the specific energy requirement for NH₄ removal ranges from 62.05–148.23 Wh g⁻¹. Overall, EC in a three-compartment reactor was an effective treatment for COD and ammonia removal. In addition, the recovery of NH₄⁺ and NO₃⁻ transferred into the central compartment also contributes to the overall removal of nitrogenous compounds. These results imply that the

EC process is applicable as a pre-treatment of mature landfill leachate since it could remove larger colloids, suspended particles, and some complex substance from landfill leachate. Therefore, it is effective in reducing the organic and inorganic loads towards biological processes.

Acknowledgement

The authors would like to acknowledge the Institut Teknologi Sepuluh Nopember for their financial support of this study, under the project scheme of the Publication Writing and IPR Incentive Program (PPHKI).

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