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Treatment of landfill leachate by electrochemicals using aluminum electrodes

Soraya Mohajeri[1], Hamidi Abdul Aziz[1], Mohamed Hasnain Isa[2], Mohammad Ali Zahed[*,3]

[1]*School of Civil Engineering, Universiti Sains Malaysia, Nibong Tebal, Pinang, Malaysia.*
[2]*Civil Engineering Department, Universiti Teknologi PETRONAS, Perak, Malaysia.*
[3]*Faculty of biological sciences, Kharazmi University, Tehran, Iran.*

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ABSTRACT

Electrochemical oxidation process has been shown to be a favourable choice for Chemical oxygen demand (COD) and color removals from various types of wastewaters. The technique was employed for mineralization of semi-aerobic landfill leachate. Leachate sampling were carried out from Pulau Burung Landfill Site (PBLS), Penang, Malaysia. The main objective was to determine the effectiveness of electrochemical oxidation in leachate treatment using aluminum electrodes which are relatively nontoxic and cost-effective. The influence of pH, reaction time, current density, electrolyte concentration, agitation rate and dilution on COD and color removals was investigated. The highest COD and color removal were obtained as 57.1% and 72.0% respectively at pH 8, current density 60 mA/cm2, electrolyte concentration 2000 mg/L, agitation rate 400 rpm, dilution 50% and reaction time 4 h. The energy consumption was determined as 128 kWh/m^3 for this type of landfill leachate. The study shows that electrochemical oxidation can be used as a step of shared treatment.

1. Introduction

Municipal landfill leachate is complex wastewater that often comprises of organic substances, heavy metals, chloride and many other soluble compounds. Factors such as different type of waste and landfill age affect the composition and concentration of contaminants (Wang et al. 2018; Yusoff et al. 2018). Inadequate management of landfill, may develop it to a source of pollution due to leachate infiltration into soil and underlying water (Mohajeri et al., 2019; Vlyssides et al. 2003).

Given the biological refractory character of old landfill leachates, utilization of techniques rather than biodegradation necessitates to efficiently lessen the contaminant load of these sewage discharge (Rivas et al. 2005). Using of sole technology is often inadequate to achieve appropriate levels of pollution reduction and integration of biological, physical and chemical processes need to be employed (Fan et al. 2010).

In the event of landfill leachate, electrochemical oxidation process using various electrodes has been revealed to be capable of reducing high molecular weight organic compounds from leachates in comparison to other physiochemical technologies which only raise a question about phase transfer of the contaminants and do not affect chemical destruction (Fernandes et al. 2015; Moreira et al. 2015; Mohajeri et al. 2010a; Hermosilla et al. 2009; Deng and Englehardt 2007; Shao et al. 2006).

Hydroxyl radical (OH$^{\bullet}$) was known as a very influential, unselective and oxidizer agent due to its quick reaction with organic compounds through hydroxylation or dehydrogenation process of nonsaturated bond, succeeding a radical substitution till their comprehensive mineralization, i.e., the modification of inceptive pollutants into carbon dioxide, water and inorganic ions (Boye et al. 2003).

In addition to OH, active chlorine, formed by the oxidation of the existent or added chloride in the wastewater, also oxidizes organic compounds. These compounds are oxidised by assistance of present Cl^-, which is converted to Cl_2, hypochlorite, chlorate, and perchlorate during oxidation process because of electrolysis, and also another oxidizers e.g. O_3, H_2O_2 and peroxides which might be exist because of the electrolyte support oxidation (Chen 2004; Arevalo and Calmano 2007). Existence of chloride ions leads the following chemical reactions to occur (Eqs. 1-3):

$$2Cl^- \rightarrow Cl_2 + 2e^- \quad (1)$$

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \quad (2)$$

$$HOCl \rightarrow H+ + OCl^- \quad (3)$$

These anodic reactions take place concurrently with the following primary cathodic reactions (Eqs. 4-6):

$$2H_2O + 2e^- \rightarrow 2OH^- + H_2 \quad (4)$$

$$2H_2O + O_2 + 4e^- \rightarrow 4OH \quad (5)$$

$$OCl^- + H_2O + 2e^- \rightarrow Cl^- + 2OH^- \quad (6)$$

Hypochlorite (OCl^-) produced in solution is a powerful oxidizing agent that is able to oxidize aqueous organic compounds (Eq. 7):

$$R + OCl^- \rightarrow CO_2 + H_2O + Cl^- \quad (7)$$

*Corresponding author E-mail: zahed51@yahoo.com

Owning to the inexpensive chloride, its nearly high solubility and strong oxidizing attributes of the produced active chlorine, indirect electrochemical oxidation of pollutants in wastewater by electrochemically generated chlorine was utilized. Moreover, it is stated that among chloride, nitrate and sulfate, chloride was the best sustenance electrolyte to oxidize electrochemical of refractory organic pollutants (Chen 2004).

Once the current density and concentration of chloride is augmented; the COD deduction efficiency is enhanced (Szpyrkowicz et al. 2005; Chiang and Chang 2001; Szpyrkowicz et al. 2001). The applied current density and the chloride concentration was significant factors that influence the COD deletion from landfill leachates.

Electrochemical waste destruction reveals numerous profits in terms of cost and safety, operational ease and equipment simplicity. The process generally executes at high electrochemical efficiency and operates necessarily under the same conditions for several types of organic wastes (Oturan et al. 2015). The chief drawback of Electrochemical (EC) systems are due to their high electrical energy consumption and sludge production that depend on the applied operational conditions and electrode material (Bayramoglu et al. 2004). Iron (Fe) electrodes, for example, are particularly notorious for sludge generation. Aluminum electrodes are relatively cheap and are not associated with major sludge generation and disposal problems as with iron electrodes. The aim of this study is to investigate the electrochemical oxidation of landfill leachate using aluminum electrodes. The effect of different operating parameters comprising pH, current density, contact time, electrolyte (NaCl) concentration and agitation rate on the deletion of COD and color was determined.

2. Materials and methods

2.1. Sampling

Sanitary landfill leachate used for the experiments was sampled from Pulau Burung Landfill Site (PBLS), Penang, Malaysia. The site is a semi-aerobic municipal landfill and has been developed as semi-aerobic sanitary landfill Level II by establishing a controlled tipping technique in 1991. Later in 2001, it was upgraded to a sanitary landfill Level III using controlled tipping with leachate (Mohajeri et al. 2010b). The collected landfill leachate was stored at 4°C to keep the wastewater characteristics unaffected before laboratory analysis and electrochemical treatment. Table 1 presents the characteristics of leachate used in the study.

Table 1. Leachate characteristics sampled from PBLS, Malaysia.

Parameters	Range	Average± Standard deviation
pH	8.3-8.8	8.5±0.19
COD (mg/L)	2380-2480	2450±40.62
Color (Pt.Co.)	3020-3150	3100±48.48
Turbidity (FAU)	220-255	240±13.69
TSS (mg/L)	120-135	130±6.12
Conductivity (µs/cm)	22400-25100	23900±994
Chloride (mg/L)	990-1410	1240±161
Sulfate (mg/L)	140-240	184±38
Nitrate (mg/L)	20-95	63±30
Temperature (ºC)	27-29	28±0.84

2.2. Electrochemical oxidation

For every run, 500 mL of the leachate was put in the EC reactor. EC oxidation was conducted using prearranged current densities. The electric power needed for the electrolysis test was generated by a laboratory DC power supply 3 A-30 V (DAZHENG, PS-305D, China). Vertically positioned electrodes with a surface area of 15 cm^2 were placed side by side to each other in the electrolytic cell. They were separated by a distance of 3 cm. A magnetic stirrer maintained at the desired value was used to mix the electrolytic cell in the solution well. Once the electrode material was disappeared > 10 %, the electrodes were switched. Experiments were carried out at room temperature and atmospheric pressure. The solution was then settled for half an hour and the supernatant was extracted once the run was terminated (Mohajeri et al. 2010a).

2.3. Analytical procedure

Analytical grade sodium chloride (Merck, Darmstadt, Germany) was utilized for electrolyte and source of chloride reactant. Various quantities of NaCl (500–3000 mg/L Cl^-) were added to the leachate to test the influence of chloride concentration on the efficiency of COD and color removals (Mohajeri et al. 2010a).

The concentration of COD was quantified colorimetrically through a HACH DR/2010 (HACH Co., USA). The same spectrophotometer at 465 nm wavelength was also used to measure the color (APHA, 2017). Eq. (8) was used to discern removal percentage:

$$R = \frac{(C_0 - C_1)100}{C_0} \quad (8)$$

where C_0 presents the initial COD or color and C_1 indicates the residual COD or color after treatment. Experiments and analytical measurements were made in triplicate.

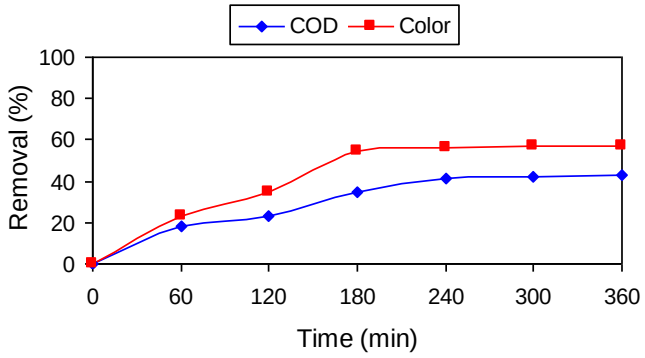

Fig. 1. Effect of reaction time on (a) the COD removal and (b) the color removal, (2000 mg/L NaCl, current density, 60 mA/cm^2, pH 8).

3. Results and discussion

3.1. Effect of reaction time on removal efficiencies

The initial pH, NaCl concentration and current density were kept constant at 8, 2000 mg/L, 60 mA/cm^2 respectively. Whereas the reaction time was differed between 30 and 360 min. Fig. 1. shows that color removal was more rapid than COD removal. The ideal operating time was ascertain to be 240 min; at which COD and color removal efficiencies were 42 % and 55 % respectively. Increases in removal efficiencies beyond 240 min were negligible. The choice of COD as the attribute to be degraded indicates that extended processing time will be needed.

3.2. Effect of pH on removal efficiencies

The initial pH of the solution was varied to test the effect of pH on the electrooxidation process. Fig. 2. shows the COD and color removals for samples containing 2000 mg/L NaCl, with pH values (adjusted with sulfuric acid or sodium hydroxide) of 4, 6, 8, 10 and 12, respectively. A current density of 60mA/cm2was employed. The results show that the organic compounds are oxidized preferably in the neutral and alkaline medium. The COD and color removal efficiencies did not change significantly at pH above 8. Alkaline conditions have been suggested to help enhance indirect oxidation (Moreira et al. 2016; Yi et al. 2008).

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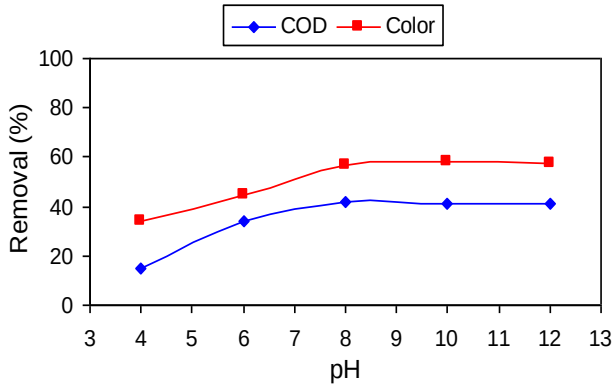

Fig. 2. Effect of initial pH on (a) the COD removal and (b) the color removal. (2000 mg/L NaCl, 240 min, current density, 60 mA/cm^2).

With the initial pH increasing from 4 to 8, the COD and color removal efficiency also increased after 240 min of electrolysis. Constant COD and color removal efficiencies of about 42% and 55% respectively were attained when initial pH was about 8.0; which indicate that leachate could be effectively degraded at its normal pH. Hence, the pH of leachate was not adjusted in subsequent experiments.

Some researchers reported that the pH effect on the electrooxidation process is not considerable. They emphasized that the initial pH does not affect significantly the degradation of organic pollutants by indirect electrochemical oxidation in the range of 3.0–10.0 (Gotsi et al. 2005). Conversely, other researchers discovered that COD reduced significantly by pH. Some of them proposed acidic and others recommended alkaline condition to increase indirect oxidation (Moreira et al. 2016; Vlyssides et al. 2003). These discrepancies were probably due to the complex compositions of leachates and type of waste.

3.3. Effect e of current density on removal efficiencies

Current density (the electric current per unit surface area of the electrode) plays an important role in the progress of electrochemical elimination of organics owning to its ability to control the reaction rate. Researchers have applied current densities from 5 to 540 mA/cm^2 for electrochemical oxidation of landfill leachate (Deng and Englehardt, 2007). As well as increasing the treatment (electricity) cost, operation at high current densities may cause darkening of leachate and formation of brown precipitates at the anode surface under weak oxidative conditions (Li et al. 2001).

The influence of applied current upon the degradative behavior of landfill leachate was examined by electrolyzing the solution at 20, 40, 60 and 80 mA/cm2. As shown in Fig. 3, COD and color removals of samples both correlated positively with the applied current density. With an increase in the current density from 20 mA/cm2 to 80 mA/cm2, the COD and color removal efficiencies raised from 22% to 45% and 38% to 60%, respectively.

High current density of the anodic oxidation of water accelerates the generation of hydroxyl radicals which could ease the indirect electrochemical oxidation of organic compounds (Fernandes et al. 2015; Un et al. 2008; Yi et al. 2008). It is also possible that an augmented current density throughout electrochemical oxidation increases active chlorine generation, which is liable for the deletion of the pollutants (Krishna et al. 2010). Additionally, some researchers have also reported color removal from leachate fully dependent on current density (Mohajeri et al. 2010c; Gotsi et al. 2005; Moraes and Bertazzoli 2005). Even though a high current density is more advantageous to organic compounds degradation, a current density of 60 mA/cm2 was adopted for the subsequent tests, because of lower energy consumption (increase in current density from 60 mA/cm2 to 80 mA/cm2 resulted in less than 5% increase in COD and color removal efficiencies).

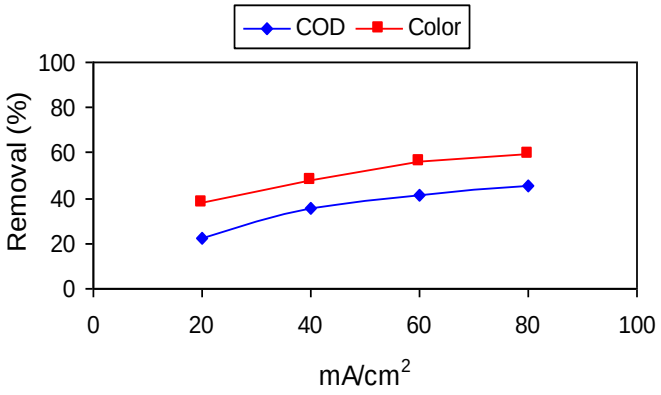

Fig. 3. Effect of current density on (a) the COD removal and (b) the color removal (2000 mg/L NaCl, 240 min)

Current efficiency has been utilized to show a total efficacy because of the activity of direct oxidation using hydroxyl radicals and indirect oxidation through electro-generated active chlorine. The COD values calculated using Eq. 9 was utilized to determine the average current efficiency (ACE) percentage of oxidation (Montanaroa and Petrucci, 2009):

$$ACE = \frac{(COD_0 - COD_t)FV}{8It} \times 100 \quad (9)$$

where COD_0 (g O_2 L^{-1}) indicates the initial chemical oxygen demand, CODt (g O_2 L^{-1}) stands for the chemical oxygen demands at a given time t (s), F for the Faraday constant (96,487 C mol^{-1}), V for the volume of the treated solution (L), I for the current applied (A) and 8 denotes the oxygen equivalent mass (g eq^{-1}). Generally, current density increases might cause a decline in the current efficiency (Mohajeri et al. 2010a). The maximal ACE of 34.7 % was detected for the current density of 20 mA/cm^2 in 240 min experimentation. The phenomenon may occur possibly owning to an enhance in the unwanted reaction of oxygen evolution (Hmani et al. 2009; Panizza and Cerisola, 2009) (Eq. 10):

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \quad (10)$$

These results approve active chlorine role in COD removal.

3.4. Effect of electrolyte concentration on removal efficiencies

A key matter in the application of electrochemical technique is supporting electrolyte usage, which can influence the conductivity of raw leachate because it simplifies the passage of current. Consequently, different concentrations of NaCl (electrolyte) was added to the system and variation in present COD and color removals was recorded.

To find the oxidation effect of active chlorine in electrochemical oxidation of leachate, chloride was added as the supporting electrolyte. Fig. 4. illustrates the electrolysis results regarding the COD and color removals at chloride ion concentrations ranging from 500 to 3000 mg/L. An increase of the Cl^- concentration from 500 to 3000 mg/L raised the COD and color removal efficiencies from 25 to 45 % and 49 to 60% respectively.

The trend might be ascribed to the indirect electro-oxidation of organics by active chlorine (chlorine/hypochlorite), produced from Cl^- at the anode (Un et al. 2008; Körbahti et al. 2007; Iniesta et al. 2001). Thus, the increased electrolyte concentration would improve removal efficiency. The additional NaCl will also decrease the power consumption owning to an elevation in conductivity as described (Mohajeri et al. 2010a). Albeit, addition of further Cl^- generally improves oxidation efficiency, toxic chlorinated organic generated over treatment

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may impede extensive use of electrochemical oxidation process (Fernandes et al. 2016; Moreira et al. 2015; Mohajeri et al. 2010a).

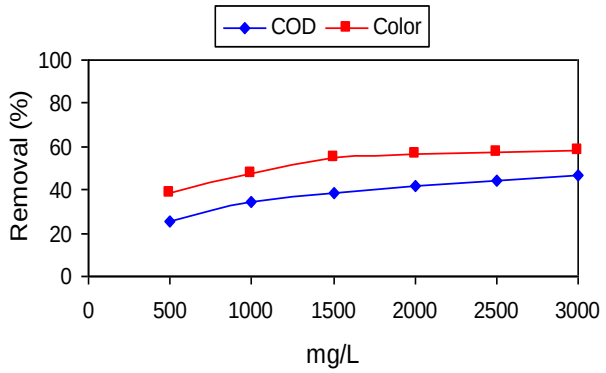

Fig. 4. Effect of electrolyte concentration on (a) the COD removal and (b) the color removal (240 min, current density, 60 mA/cm^2).

3.5. Effect of the agitation rate on removal efficiencies

To investigate the effect of mass transfer on the electrochemical treatment of leachate, the influence of agitation rate during electro-oxidation of leachate was studied by changing the magnetic stirrer rate. The rate of agitation was ranged between 0−600 rpm, and the results are shown in Fig. 5. The rest of the process parameters and media conditions remain unaltered.

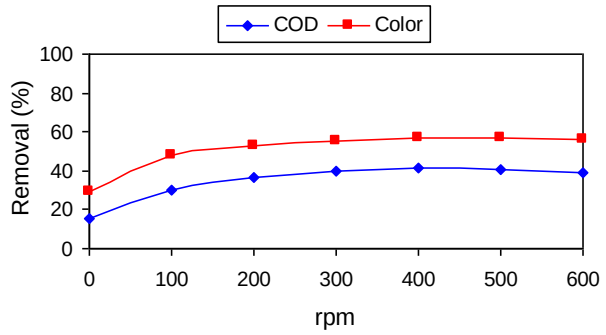

Fig. 5. Effect of agitation rate on (a) the COD removal and (b) the color removal (2000 mg/L NaCl, 240 min, current density, 60 mA/cm^2).

Varying the agitation speed affected markedly the COD and color removal efficiencies. High agitation rates led to a more rapid and high efficient electrochemical process. As shown in Fig. 5. via elevation of rotation speed, COD and color removals are increased up to 400 rpm beyond that, there is no substantial increase in COD and color removals by increasing rpm. A similar trend was also observed by Bensalah and co-authors (2009) during their study on the electrochemical treatment of synthetic wastewaters on BDD anodes. These results revealed that stirring the mixture mechanically necessitates to achieve the maximum effect of electro-oxidation. The 400 rpm was selected for all experiments because the set-up may be disrupted in higher speeds.

3.6. Effect of the dilution on removal efficiencies

The effectiveness of dilution on leachate mineralization and decolorization were examined by a series of experiments. When the effect of initial COD concentration was investigated, experimental conditions were as follows: a current density of 60 mA/cm2, electrolyte concentration of 2000 mg/L and the original pH of the leachate solution. The leachate samples were diluted 50, 100, 150, and 200% with distilled water. Fig. 6, shows the influence of dilution during electrolyzes of leachate. It was found that dilution affects significantly the removal efficiencies in the electrochemical oxidation process. At first, the degradation efficiency increased with decreasing the initial COD of leachate and then reached a maximum value of 57.1%, at a dilution of 50%. Further decreases of the initial COD lead to a sharp decrease of the removal efficiency from 57.1 % down to 36.6 % at dilution percentage of 200 %. It can be observed that high dilution provided low COD and color reduction. Oxidation of organic matters in leachate can be affected by dilution since the concentration of organics is a key factor. Like other chemical reactions, oxidation is limited in low concentrations.

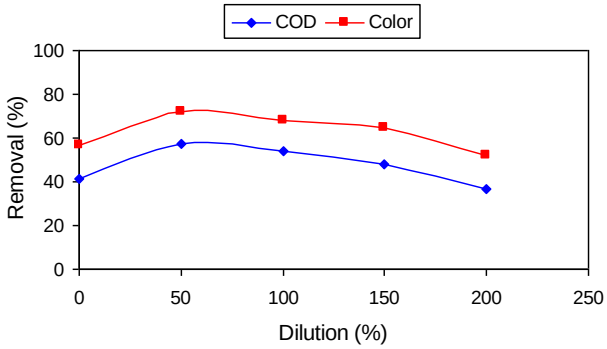

Fig. 6. Effect of dilution on (a) the COD removal and (b) the color removal (2000 mg/L NaCl, 240 min, current density, 60 mA/cm^2).

3.7. Economic evaluation

To evaluate the economy of the electrochemical process, the required electrical energy consumption (EEc) per volume of leachate is calculated as kWh/m3 according to (El-Ashtoukhya et al. 2009) using Eq. (11):

$$EE_c = \frac{VCt}{S_v} \qquad (11)$$

where V is voltage (volt), C is current (Ampere), t is treatment time (h) and Sv is sample volume (L). In this study the operational parameters of the experiment including electrolysis current, cell voltage, and the reaction time were determined to be 2 A, 8V and 240 min, respectively. The volume of the leachate treated was 0.5 L, therefore energy consumption was determined as 128 kWh/m^3 for Pulau Burung semi-aerobic landfill leachate.

3.8. Comparison with other techniques

Other researchers (e.g. Feki et al. 2009; Ilhan et al. 2008; Moraes and Bertazzoli 2005) also utilized electrochemical oxidation technique for the treatment of landfill leachate. Moraes and Bertazzoli (2005) applied electrochemical treatment for landfill leachate in a pilot scale flow reactor, using oxide-coated titanium anode. The maximum COD removal of 73 % was acquired at a current density of 116.0 mA/cm^2 and 180 min of processing. In a study performed by Cabeza et al. (2007), complete removal of both COD and ammonium were obtained after 6−8 h using a boron-doped diamond (BDD) anode, to treat biologically and physicochemically treated leachates from a municipal landfill site of Meruelo in Cantabria, Spain. Ilhan and coauthors (2008) reported that more than 59 % of COD and 14 % of ammonia removal were achieved within 30 min reaction time for a current density of 631 mA/cm^2 using aluminum electrode. Feki et al. (2009) also found that under the optimal operational conditions (t = 1 h, J = 4 A dm^{-2}, Ti/Pt electrode), COD, color and heavy metal concentrations of the final effluent meet the Tunisian discharge standards in the sewer. In addition, Bashir et al. (2009) used graphite carbon electrodes and sodium sulfate as the electrolyte for electrochemical treatment of landfill. The maximum COD removal of 68 % was obtained when the reaction time was 4 h and the

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current density was 79.9 mA/cm^2 whilst the original COD was 1414 mg/L. In comparison to the previous literatures, we obtained specific conditions for electrochemical treatment of highly polluted leachate, indicating that electrochemical degradation could be used as a step of joint treatment. Our results here call for the more treatment of leachate to reach higher COD and color removals compliance with the environmental regulations.

4. Conclusions

Electrochemical oxidation process, using aluminum electrodes, was applied on mineralization and decolorization of landfill leachate. The effects of operating parameters i.e., pH, reaction time, current density, electrolyte concentrations, agitation rate and dilution were tested. The results demonstrated that electrochemical oxidation could effectively eliminate COD and color from municipal landfill leachate under proper conditions. The highest deletion of COD and color were obtained as 41.6 % and 56.4 % respectively. By dilution of 50 % within this condition, COD and color removal raised to 57.1% and 72.0 % respectively. Electrochemical treatment may be a viable option for the degradation and remediation of landfill leachate. We therefore require additional treatments of leachate for increase of COD and color removals to comply with the environmental obligations.

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