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Water Practice & Technology Vol 15 No 2
doi: 10.2166/wpt.2020.041

Degradation and biodegradability improvement of the landfill leachate using electrocoagulation with iron and aluminum electrodes: A comparative study

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Abstract

This present study investigates the comparative study of iron and aluminum electrodes for the treatment of landfill leachate by the Batch Electrocoagulation (EC) technique. The performance of EC was used to determine the removal efficiency of COD and Color. The effects of operating conditions such as electrode material, stirring speed, inter-electrode distance, electrolysis time, initial pH, and applied voltage were studied to evaluate the performance of the electrode. The electrodes were arranged in a monopolar mode by applying different cell voltages of 4, 6, 8, 10 and 12 V for 180 min of electrolysis time (ET) with a varying inter-electrode distance between 1 and 4 cm. The iron and aluminum electrodes can be successfully used as anodes and cathodes for the treatment process, which makes the process more efficient and easier to maintain. Based on the obtained results, it was observed that there was an increase in BOD/COD ratio from 0.11 to 0.79. The maximum removal of COD and Color was found to be 76.5% and 67.2% respectively, accomplished with 105 min optimum electrolysis time with a pH of 9.25 using an iron electrode. In the case of the aluminum electrode, the BOD/COD ratio was increased from 0.11 to 0.66. Over 78.4% of COD and 77.0% of Color removal was obtained with 90 min optimum electrolysis duration and pH 9.3 with an optimum 10 V and an optimum inter-electrode distance of 1 cm. However, the aluminum electrode is superior to iron as a sacrificial electrode material in terms of Color and COD removal efficiency. The aluminum electrode significantly treated landfill leachate by the electrocoagulation method under optimum experimental conditions.

Key words: aluminum electrode, electrocoagulation, iron electrode, landfill leachate

INTRODUCTION

Leachate can be defined as water (rainwater or groundwater) that has percolated through solid waste. Rainfall is the main contributor for the generation of leachate (Abbas *et al.* 2009). There are many factors that affect the quality of leachates such as age, seasonal weather variation, precipitate, waste type and waste composition, landfill leachate composition mainly depending on the age of the landfill (Silva *et al.* 2004). The main characteristic of leachate is BOD, COD, BOD/COD ratio, suspended solids, pH, ammonia-nitrogen, and heavy metals. Leachate may contain a large quantity of organic matter, biodegradable, humic-type constituents, and chlorinated organic and inorganic salts (Renou *et al.* 2008). Pre-treatment is required for the landfill leachate to meet the standards for its discharge for direct disposal into surface waters or sewers. There are many wastewater treatment technologies that have been used to treat landfill leachate, such as the membrane process (Amokrane *et al.* 1997; Alizadeh *et al.* 2015), Sequencing Batch Reactor (SBR) (Neczaj *et al.* 2005; Laitinen *et al.* 2006; Bashir *et al.* 2010; Khosravi *et al.* 2017), coagulation-flocculation (Amokrane

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et al. 1997), constructed wetland (Ogata *et al.* 2015) and Thermophilic Membrane Bioreactor (Visvanathan *et al.* 2007), which have been used in the literature.

The electrocoagulation process has proven to be more economic, highly efficient in the removal of pollutants, and has been considered as a promising treatment technology. Hence, the EC process has been applied for a variety of wastewater treatments such as dairy wastewater (Kushwaha *et al.* 2010), potato chip manufacturing wastewater (Kobya *et al.* 2006), distillery wastewater (Krishna *et al.* 2010; Farshi *et al.* 2013), dye wastewater (Riadi *et al.* 2017), restaurant wastewater (Chen *et al.* 2000), health care wastewater (Singh *et al.* 2018). The mechanism of EC reactions are as follows from Equations (1) through (12).

Anode:

$$Fe(s) \rightarrow Fe^{2+}(aq) + 2e^- \quad (1)$$

$$Fe^{2+}(aq) + 2OH^-(aq) \rightarrow Fe(OH)_2(s) \quad (2)$$

Cathode:

$$2H_2O + 2e^- \rightarrow H_2(g) + 2OH^- \quad (3)$$

Overall:

$$Fe(s) + 2H_2O \rightarrow Fe(OH)_2(s) + H_2(g) \quad (4)$$

Oxidation:

$$2Cl^- \rightarrow Cl_2 + 2e^- \quad (5)$$

$$Cl_2(g) + H_2O \rightarrow HOCl + H^+ + Cl^- \quad (6)$$

$$Fe(OH)_2 + HOCl \rightarrow Fe(OH)_3(s) + Cl^- \quad (7)$$

$$Fe^{2+} \rightarrow Fe^{3+} + e^- \quad (8)$$

$$Fe^{3+} + 3H_2O \rightarrow Fe(OH)_3 + 3H^+ \quad (9)$$

Depending on the pH range, the ferric ions generated from the electrocoagulation process may result in the formation of monomeric ions, ferric hydroxo complexes with hydroxide ions and polymeric species such as $Fe(OH)^{2+}$, $Fe(OH)_2^+$, $Fe_2(OH)_2^{4+}$, $Fe(OH)_4^-$, $Fe(H_2O)_2^+$, $Fe(H_2O)_5(OH)^{2+}$, $Fe(H_2O)_4(OH)_2^+$, $Fe(H_2O)_8(OH)_2^{4+}$, $Fe_2(H_2O)_6(OH)_4^{2+}$, which finally converts into $Fe(OH)_3$. The larger surface area resulting from freshly formed $Fe(OH)_3$ is advantageous for the adsorption of soluble organic compounds and trapping of colloidal particles (Kobya *et al.* 2003; Feng *et al.* 2007). The reaction when the iron is used as an electrode is given below:

$$nFe(OH)_3 \rightarrow Fe_n(OH)_{3n} \quad (10)$$

The amount and variety of hydrolysis products formed by anodic dissolution significantly depend on electrolysis time when iron is used as an electrode. The formed $Fe(OH)_n(s)$ complexes are in the form of gelatinous suspension. These gelatinous complexes may play a very good role in the effective removal of pollutants. Various processes may be involved in the pollutant removal process such as neutralization of charge, adsorption, electrostatic attraction and complexation.

The variety of monomeric and polymeric species formed due to $Al^{3+}$and OH^- ions generated by electrode reactions include $Al(OH)_{2+}$, $Al(OH)_2^+$, $Al(OH)_2^{4+}$, $Al(OH)_4^-$, and $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$, and $Al_{13}(OH)_{34}^{5+}$, respectively. All these monomeric and polymeric species finally lead to the formation of $Al(OH)_3$. The reaction when aluminum is used as an electrode is given below:

$$\text{At anode: } Al \rightarrow Al^{3+} + 3e^- \tag{11}$$

$$\text{At cathode: } 3H_2O + 3e \rightarrow 3/2H_2 + 3OH^- \tag{12}$$

Sparse research in open literature focusing on the BOD/COD ratio during and after electrocoagulation treatment of landfill leachate promotes this research work. The main significance of this research work is to study the degradation and biodegradability of landfill leachate aiming at the BOD/COD ratio and to optimize the process parameters such as inter-electrode distance, electrolysis time and voltage (current density) with a main focus on COD and Color removal, and to compare the investigations between iron and aluminum electrodes.

MATERIALS AND METHODOLOGY

Study area

The area selected for the study is in Mysuru city, Karnataka, India. It is located at 12.30°N 76.65°E with an average altitude of 770 meters. The dumpsite was situated at Vidyaranyapuram, Mysuru, Karnataka. The dumping of waste in this area has taken place for the past 6–7 years. The area consists of accumulated waste of about 2, 50,000 m^3 and the area used for dumping of waste is about 41.47 acres. The present study attempts to treat landfill leachate using the electrocoagulation process. The sample landfill leachate was collected in a tank, wherein the leachate is coming from the pipes which are shown in Figure 2. The various physical and chemical parameters were analyzed in this study. The physical and chemical parameters in the initial characterization of the sample are shown in Table 1.

Experimental setup for electrocoagulation

Electrochemical experiments were conducted in a plexi-glass laboratory scale batch electrochemical reactor 11 cm × 14 cm × 13 cm of 2 L capacity with a working volume of 1.75 L at room temperature, which was used in the setup. The reactor was kept under the process of continuous agitation using a magnetic stirrer at 250 rpm to avoid the formation of concentration gradients. The T-shaped electrodes were made from iron and aluminum plates with a size of 5 cm × 7 cm, which were used as both anode and cathode electrodes (35 cm^2 effective surface area). At the bottom of the electrodes, a gap of 2 cm was maintained to facilitate continuous and easy stirring. Before each treatment process, the electrodes were cleaned and degreased. The power supply used to run all experimental conditions was DC power. The distance between the anode and cathode electrodes was varied from 1 cm to 4 cm, wherein the voltage used in the electrolysis process was 4 V. The duration of the electrolytic process was 180 mins, with 15 mins time intervals. Every 15 mins, a sample was collected for further processing. The collected samples after electrolysis were used to analyze the parameters, such as voltage (current density), electrolysis duration, COD, Color, and pH. Among these analyzed parameters, pH, electrolysis duration, as well as the distance between the electrode and the voltage (current density) were optimized in this study. The experimental setup for electrocoagulation to a lab-scale process is shown in Figure 1.

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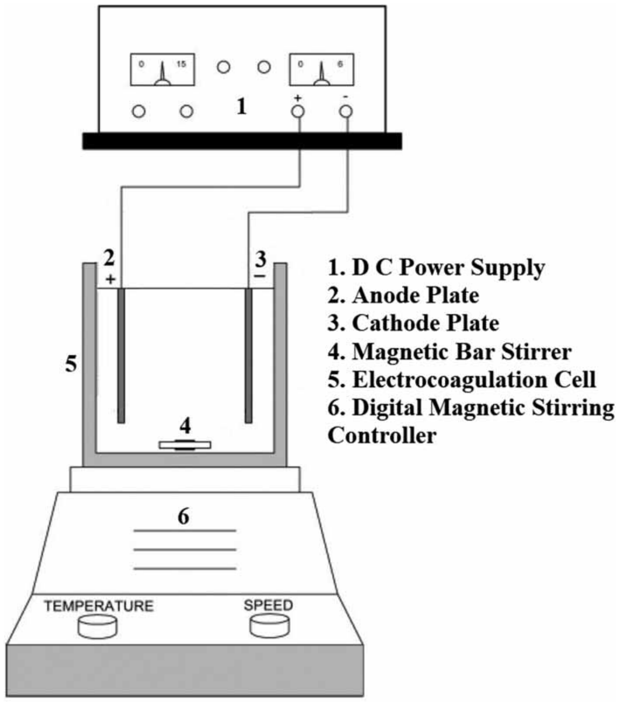

Figure 1 | Experimental set up of electrocoagulation treatment at a lab scale.

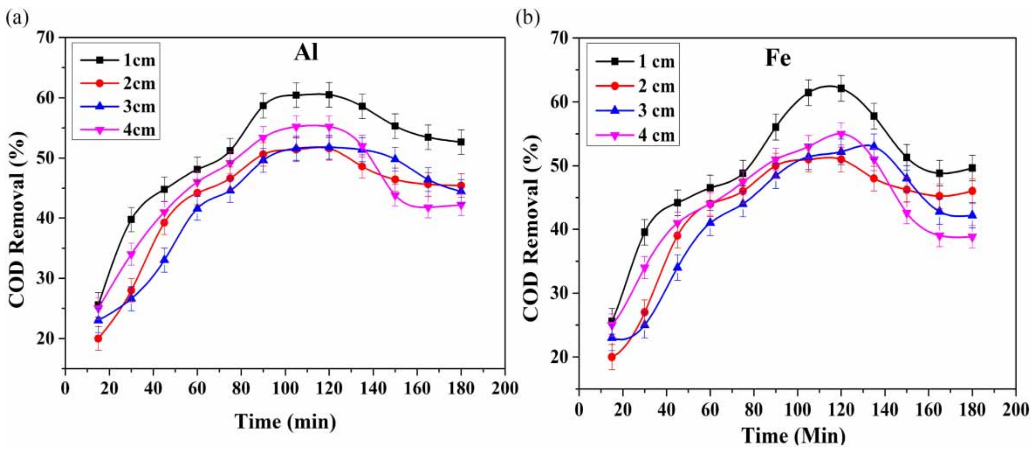

Figure 2 | (a) and (b) Effect of inter-electrode distance on percentage removal of COD in the leachate treatment by EC with Al and Fe electrodes.

RESULT AND DISCUSSION

Influence of inter-electrode distance:

The inter-electrode distance has been studied as one of the parameters to minimize the consumption of electricity in the treatment of landfill leachate. The distance between the electrodes was varied between 1, 2, 3 and 4 cm. An increased percentage removal of COD and Color was observed with decreased inter-electrode distance from 4 to 1 cm for both Fe and Al electrodes. The obtained results show the insignificant effect of inter-electrode distance on the COD and Color removal percentage. Maximum removal efficiency was observed at 1 cm for the shortest distance between the electrodes, which had an electrode area of 35 cm^2. Suppose the spacing was less than 1 cm, it would disallow the

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Table 1 | Characterization of the landfill leachate

No.	Parameters	Concentration
1	pH	8.67
2	Conductivity	38.5 mS/cm
3	Turbidity	140NTU
4	Total solids	16,760(mgL^{-1})
5	Total dissolved solids	14,580(mgL^{-1})
6	COD	13,760(mgL^{-1})
7	Phosphate	208.5(mgL^{-1})
8	Total suspended solids	1,648(mgL^{-1})
9	Nitrates	97.3(mgL^{-1})
10	BOD	1,519(mgL^{-1})
11	Chloride	7,034(mgL^{-1})
12	BOD/COD	0.11 (mgL^{-1})
13	Color	8,750 PCU

flow of liquid absorbate in the intermediate space between the electrodes, and hence impede the removal efficiency.

The results show that the electrochemical method has significant efficiency in the removal of COD and Color. Greater efficiency was observed; 62.1% and 60.5% of COD and 52.0% and 47.5% of Color was removed by iron and aluminum electrodes respectively, as shown in Figure 2(a) and 2(b). Figure 3(a) and 3(b) show the efficiency of COD and Color respectively using different inter-electrode distances at the same experimental conditions. The ohmic potential drop is proportional to the distance between the electrodes (Alizadeh *et al.* 2015).

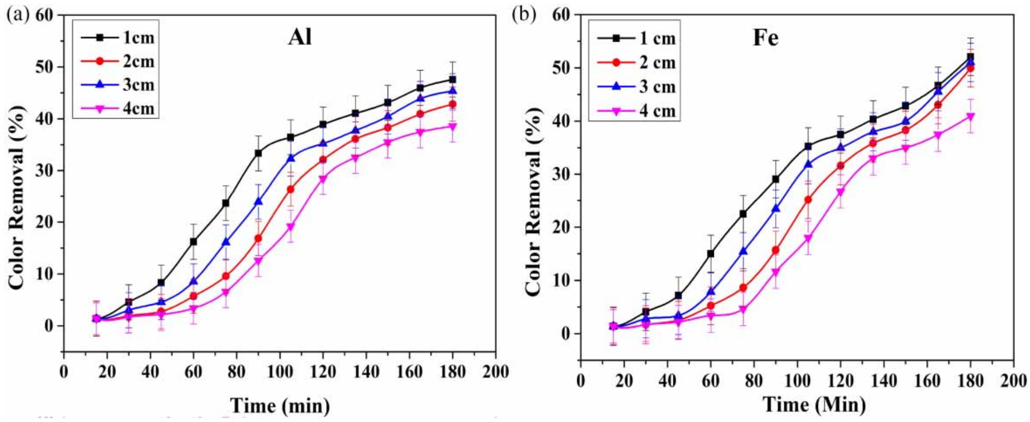

Figure 3 | (a) and (b) Effect of inter-electrode distance on percentage removal of Color in leachate treatment by EC with Al and Fe electrodes.

As the inter-electrode distance is increased, the energy consumption also increases due to the electrostatic effect of the distance between the electrodes. The electric field can be controlled by changing the applied current but once the distance between the electrodes changes, the electric current also changes (Bouhezila *et al.* 2011). Further experiments were carried out by keeping 1 cm spacing as the optimum inter-electrode distance.

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Influence of applied voltage on the process efficiency

In electrocoagulation, voltage and electrolysis time are the important operational parameters to be set effectively, for the crucial removal of leachate under defined electrical energy and consumption of power. The electrocoagulation experiment was carried out for different voltages, such as 4 V, 6 V, 8 V, 10 V, and 12 V.

The removal of COD and Color was found to be extreme at 10 V for both Fe and Al electrodes ascribed to the reaction between organic compounds and Fe and Al ions and the formation of insoluble products (Alimohammadi *et al.* 2017). The removal efficiencies for iron and aluminum was found to be 76.5% and 78.4% of COD respectively, as shown in Figure 4(a) and 4(b) and 67.21% and 77.0% of Color respectively, as shown in Figure 5(a) and 5(b), at an optimum 10 V and optimum time of 105 min for Fe and 90 min for Al electrodes. The results suggested that if the voltage in the electrocoagulation increases, the treatment efficiency also increases. The aluminum electrode was more efficient compared to the iron electrode. Another research group had shown that an increased voltage resulted in an increased treatment efficiency; this might lead to increased coagulant dose and bubble generation rate (Golder *et al.* 2007). Further increases in voltage would result in the faster dissolution of the anode material. Removal of COD by the electrochemical method is by oxidizing organic matter, and producing oxidant agents such as hydroxyl radicals (•OH) or hypochlorite (HOCl) (if Cl^- is present) (Pirsaheb *et al.* 2016). It was observed that the COD removal decreased

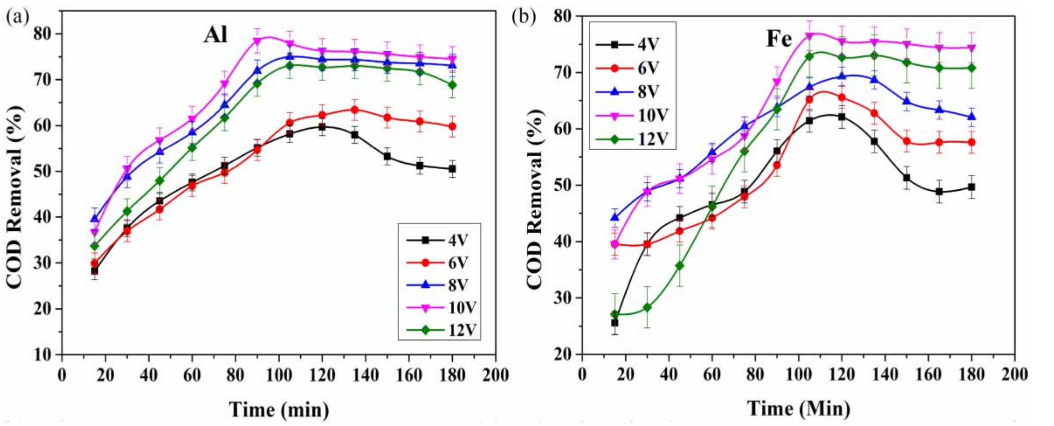

Figure 4 | (a) and (b) Effect of applied voltage on leachate treatment by EC (COD removal) for Al and Fe electrodes.

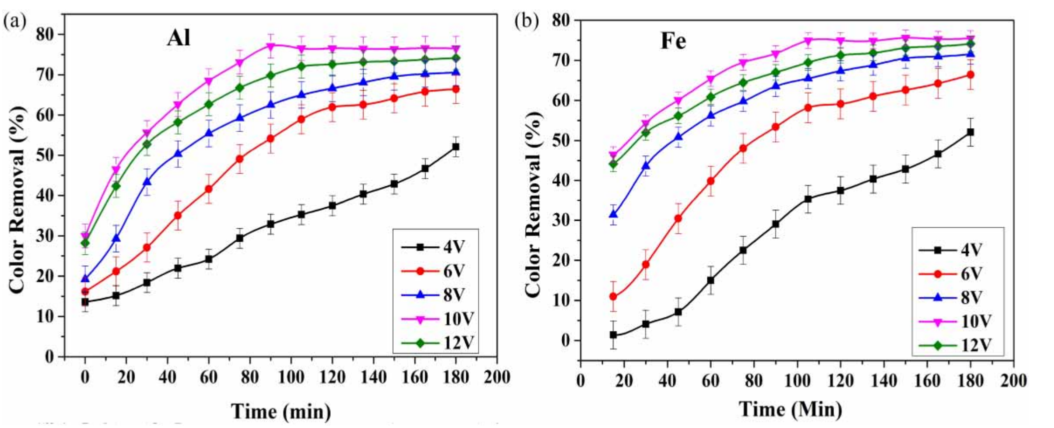

Figure 5 | (a) and (b) Effect of applied voltage on leachate treatment by EC (Color removal) for Al and Fe electrodes.

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after 100 min electrolysis time because the chloride ions in the wastewater are exhausted under the influence of high voltage (Singh *et al.* 2019). It was noticed that the rate of COD and Color removal was relatively high at 10 V compared to 12 V.

Hence, iron and aluminum electrodes can be successfully used as anodes and cathodes for electrocoagulation processes due to their increased efficiency and easier maintenance. Faraday's law explains that the applied current was directly proportional to the amount of ionized metal. Hence, the COD removal was high and an increased current density was noticed.

When the current density increases, it will result in the generation of more magnesium ions, which is favorable for co-precipitation and electrocoagulation methods. Asselin *et al.* (2008) have shown that the decrease in COD level was due to the destabilization of colloidal organic compounds and the combined effects of cathodic reduction. They have also observed a thin brownish layer deposited on the surface of the cathodic electrode after the process of electrocoagulation, which is an indication of the cathodic reduction phenomenon.

Influence of pH changes during electrochemical treatment:

It was observed that the pH of the landfill leachate in the electrocoagulation process had been raised from 8.67–9.25 for the iron electrode and 8.67–9.31 for the aluminum electrode at 10 V, as shown in Figure 6(a) and 6(b). The highest COD and Color removal were obtained at pH 9.25 for the iron electrode and pH 9.31 for the aluminum electrode. At the end of the processing time, the results specified that based on the level of activity of the anode and cathode, the pH in the process will increase. This is due to the foremost activities at the cathode (Ilhan *et al.* 2008), which resulted from the generation of hydroxide ions at the cathode through the electrochemical reduction of water (Oumar *et al.* 2016). De-colorization of effluent is very low at acidic pH of the medium, whereas it is very high under neutral or alkaline conditions (Huda *et al.* 2017). The formed iron hydroxides remain as a suspension, which induces the removal of pollutants through adsorption, coagulation, and co-precipitation under alkaline conditions (Gengec *et al.* 2012). This leads to the increased removal of Color under neutral and alkaline pH. In the electrocoagulation process, the pH of water was found to be high due to an ammonia stripping process (Ilhan *et al.* 2008). Some of the research groups have found that the variation in the pH of the medium did not significantly alter the removal of COD in the treatment process (Deng & Englehardt 2007). Another research group also reported that COD removal in the treatment process at alkaline conditions, that is, pH 8.9 and 10, was achieved, 4% higher compared to the neutral conditions, that is, pH 7.5 (Wang *et al.* 2001). Hence, it is proved that alkaline conditions are more favorable for the treatment of landfill leachate wastewater.

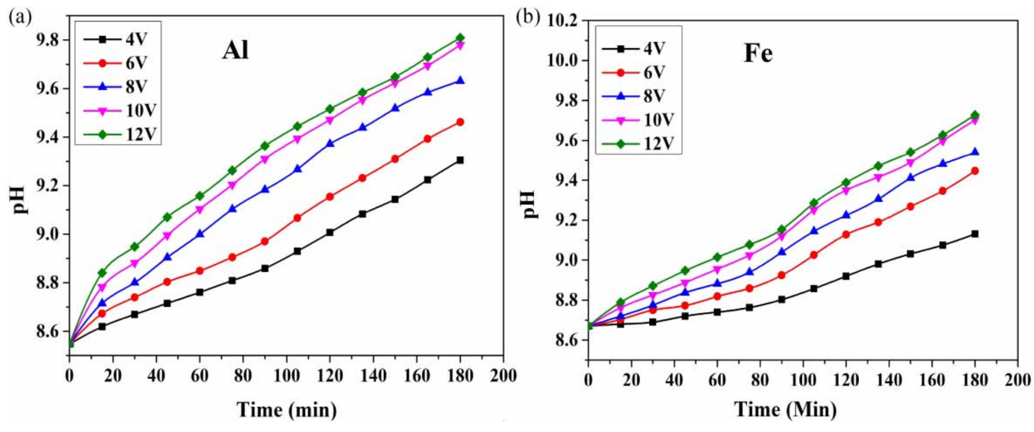

Figure 6 | (a) and (b) Effect of applied voltage on leachate treatment by EC (pH) for Al and Fe electrodes.

doi: 10.2166/wpt.2020.041

Effect of BOD/COD ratio in the electrocoagulation process

In Figure 7(a) and 7(b), it is observed that there was an improvement in biodegradability of landfill leachate evaluated through the evolution of the BOD/COD ratio. When iron was used as an electrode, it was observed that the ratio of BOD/COD increased from 0.11 to 0.79. It was found that the maximum removal percentage of COD and Color at the optimum experimental conditions was at 10 V with an inter-electrode distance of 1 cm for 180 min. Similarly, when aluminum was used as the electrode, the BOD/COD ratio was found to be increased from 0.11 to 0.66 under optimum experimental conditions. As time passes, COD degraded with time for the iron electrode. There was an improvement in the BOD/COD ratio. When the voltage increases, the degradation of COD also increases and that results in an increase in the effluent BOD/COD ratio. In the case of the aluminum electrode, the BOD/COD value gradually increases along with time and COD degradation is high compared to the iron electrode, and it is therefore confirmed that the BOD/COD ratio increases with the increase in voltage, and that can be observed with both the iron and aluminum electrodes. This is due to increasing voltage, which increases the overall potential essential for the generation of chlorine/hypochlorite. The low BOD/COD ratio in the effluent indicates that it contains recalcitrant substances which are not easily biodegradable or that non-biodegradable material is present in the leachate (Visvanathan *et al.* 2007).

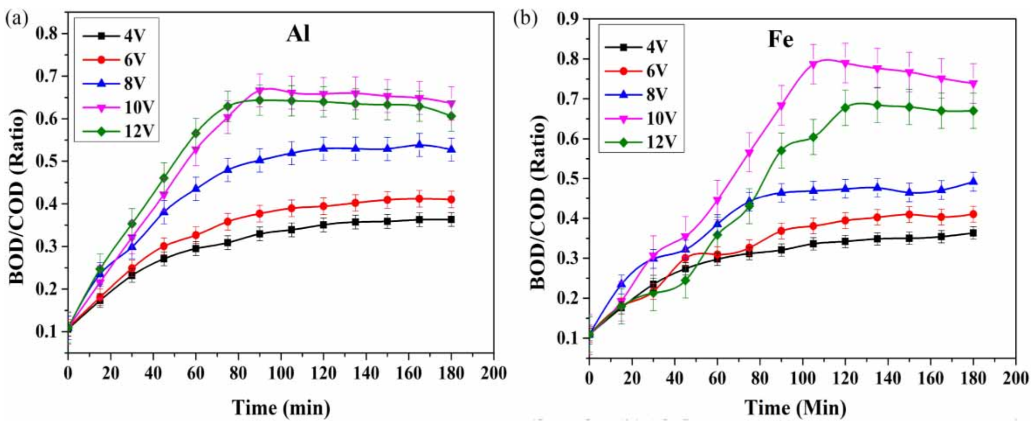

Figure 7 | (a) and (b) Effect of BOD/COD ratio on leachate treatment by EC for Al and Fe electrodes.

Electrode dissolution pattern

The electrode dissolution (ED) plays a vital role in the electrocoagulation process, where it offers information on the amount of consumption of electrode per kilogram of removed COD per cubic meter of wastewater to be treated. This helps to estimate the operational treatment cost. The removal of contaminants/pollutants from the wastewater will be assisted by electrode dissolution through the formation of electro-flocs, which is an essential part of any of the treatment methods. Figure 8(a) and 8(b) represent ED for different cell voltages such as 4,6,8,10, and 12 V with 1 cm of inter-electrode distance and Fe and Al electrodes arranged in monopolar mode in a batch reactor, 24.3 g and 5.86 g of electrode dissolution of Fe anode and Al anode electrodes for 105 min and 90 min electrolysis duration respectively, as shown in Figure 8. Aluminum electrodes are more effective than iron electrodes. In monopolar mode, ED will be strongly influenced by the position of the electrodes and the applied voltage with the corresponding current across the two electrodes.

The higher electrode dissolution rate is obtained at the positive terminal (anode) connected electrode compared to the negative terminal (cathode) connected electrode. In the cathode, there was

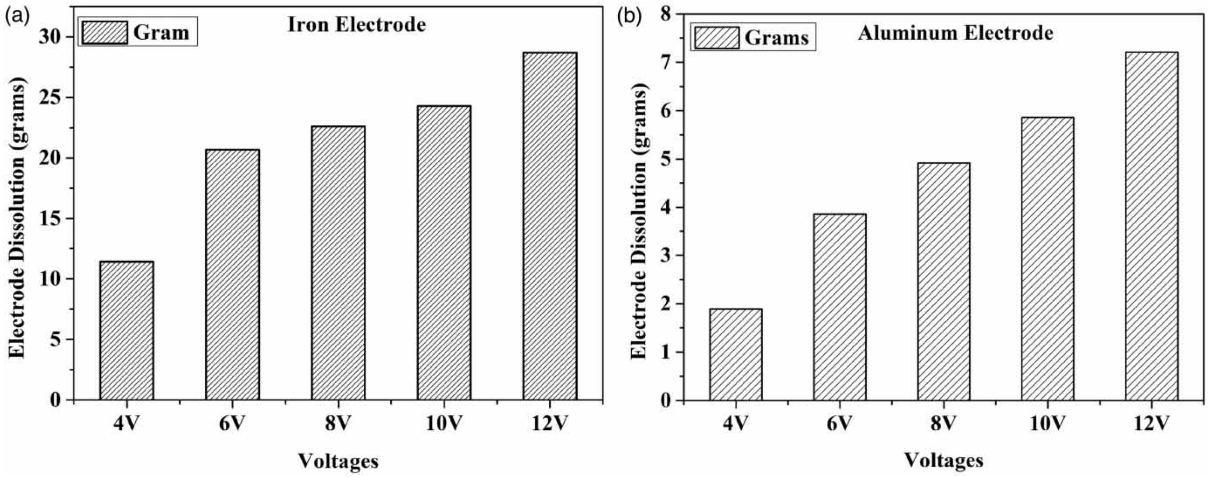

Figure 8 | (a) and (b) Effect of electrode dissolution pattern for Fe and Al electrodes.

low electrical resistance and hence ED is least. When the anode and cathode electrodes are close to each other, an increased oxidation rate is shown, producing more coagulant generation because of the higher current between the electrodes. If the inter-electrode distance is too close, it can cause short-circuiting during the treatment process (Singh *et al.* 2018).

CONCLUSIONS

The present paper shows the performance of electrocoagulation using Al and Fe electrodes for the treatment of landfill leachate. The effect of initial pH, inter-electrode distance, electrolysis time and applied voltage were studied for COD and Color removal. Experiments were carried out for different applied voltages of 4, 6, 8, 10 and 12 V using Al and Fe electrodes for an electrolysis time of 180 min. The electrodes were placed at an inter-electrode distance varying from 1 to 4 cm and connected in a monopolar mode. Maximum removal of COD and Color using the Fe electrode was found to be 76.5% and 67.2% respectively for an applied voltage of 10 V, pH 9.25 and 1 cm electrode separation distance for 105 min electrolysis time. At that time, the BOD/COD ratio increased from 0.11 to 0.79. Removal of COD and Color using Al electrodes was found to be 78.4% and 77.0% respectively for an applied voltage of 10 V, pH 9.3 and 1 cm electrode separation distance for 90 min electrolysis time. At that time, BOD/COD ratio increased from 0.11 to 0.66. The overall data thus showed that the aluminum electrode was more efficient than the iron electrode material in treating landfill leachate, in terms of COD and Color removal.

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