

Stabilized Old Landfill Leachate Treatment Using Electrocoagulation

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Abstract

Electrocoagulation of leachate samples was performed by using aluminium and iron electrodes. The objective of study was to optimize the parameters such as current density, electrolysis time and pH for maximum removal of chemical oxygen demand by electrocoagulation process from landfill leachate collected from Deonar landfill in Mumbai, India. Landfill leachate was found to have Turbidity (177 NTU), Chemical oxygen demand (2300 mg/l), low BOD/COD ratio (0.10) and Total solids (10910) mg/l respectively. The Chemical Oxygen Demand (COD) of leachate sample was found to be 2300 mg/l and Biochemical Oxygen Demand (BOD) confirming that it was old or stabilized leachate. 47% COD was removed at current density of 466A/m² with optimum electrolysis time of 60 mins and pH 8 with aluminium electrode while for iron electrodes, the COD removal efficiency came out to be 56% at current density of 466A/m², optimum electrolysis time 90 mins and pH 6.

Keywords: electrocoagulation; iron electrode; aluminium electrode; chemical oxygen demand

1. Introduction

Presently the population of world has reached 7 billion. As the population is increasing, solid waste generation is also following similar trend. Urbanization is a major trend all over the world mainly due to rapid growth in population. More than half the population of the world now lives in urban areas. This may be attributed to the continuous increase in global population (Kumar *et al.*, 2016). Municipal landfills are used worldwide for the decomposition of the municipal solid waste. Before 1975 nobody thought about the contamination of the groundwater because of landfill leachate. So the design of landfills was not having any system to control or check the flow of landfill leachate (Yadav and Dikshit, 2016). Today the landfill leachate has taken the attention of research. When refuse decomposed in landfills gets moisture through rainwater or by other mean, then contaminants (like organic matter, heavy metals) get dissolved and flows out of the landfill as called leachate (Wu *et al.*, 2004). The composition of refuse has changed over a years due to change of life styles which is now producing more strength landfill leachate. This high strength leachate contaminates the nearby groundwater and surface water. Generation of landfill leachate depends on design

and operation of landfills. Characteristics of landfill leachate vary from one to another, and it also changes with space and time on same landfill with fluctuations. It depends mainly in variations in climate, hydrogeology and waste composition (Keenan *et al.*, 1984). Landfill engineering focuses on reducing leachate production, collection and treatment prior to discharge (Farquhar, 1989). So there are requirements of effective treatment technologies to treat landfill leachate for stopping the contamination of groundwater and to save the people from diseases living nearby landfills.

Landfill leachate contains both larger fraction of higher molecular weight organic material and heavy metals (Wang *et al.*, 2002; Knox *et al.*, 1979). Separate treatment either by biological or physico-chemical cannot give high removal efficiencies of pollutants like BOD, COD, heavy metals etc. So the combination of physicochemical and biological treatment is required to remove both organic matter and heavy metals (Ehrig, 1984; Crawford *et al.*, 1985). Landfill leachate is classified in young, medium and old depending on its age. Young leachate contains very high COD and BOD around more than 10000 mg/l and 2000 mg/l respectively while old leachate contains very low BOD/COD ratio around 0.1 (Lema *et al.*, 1988).

Effective pre-treatment like adsorption, coagulation of leachate is required which can reduce the load on further treatment process with economic feasibility. As coagulation is the pre-treatment process which is applied on many types of wastewater from decades but now, there is new emerging pre-treatment technology of electrocoagulation also which uses electric current which functions as a trigger for the anode electrolysis to generate ions to enhance the formation of metal hydroxide for removing pollutants (Veli *et al.*, 2008).

Electrocoagulation is an efficient technique because hydroxides when adsorbed on mineral surfaces form large flocs which are stable and can be easily removed by filtration (Khandegar *et al.*, 2013). This method is simple and requires simple instruments which can be easily designed. It also does not involve chemical treatment and hence there is no production of secondary pollutants with minimum sludge generation (Mollah *et al.*, 2001). However, in this method, sacrificial anodes need periodic replacement and there can be formation of oxide film on cathode which can provide resistance to current (Ilhan *et al.*, 2008; Labanowski *et al.*, 2010). The objective of present study was to optimize the parameters such as current density, electrolysis time and pH for maximum removal of chemical oxygen demand by electrocoagulation process from landfill leachate.

Orkun and Kuleyin (2012) investigated that electrocoagulation with the iron anode gives the COD removal efficiency of 65.85%. The optimum values of current density, operation time, conductivity and pH are 30 mA/cm², 180 min and pH 6.54 respectively. They also evaluate the performance of electro fenton and observed that COD removal efficiency is 10% more

than that of electrocoagulation with optimum dose of 5000 ppm hydrogen per oxide.

Bouhezila *et al.* (2011) studied that after 30 min of treatment of landfill leachate by aluminium electrode with current density 500 A/m², the removal efficiency of COD, turbidity, color and total nitrogen becomes more than 70%, 60%, 56% and 24% respectively. But with Fe electrodes the removal efficiency of COD, total nitrogen got reduced upto 68% and 15% respectively. The final pH increased from 8.69 to 9.2.

Li *et al.* (2011) examined the factors affecting the removal efficiencies of COD and BOD₅. The results showed that at optimum condition of 4.96 mA/cm² current density, 2319 mg/l Cl concentration, 90 min electrolysis time the removal efficiency of COD, BOD₅ were 49.8% and 69.7% respectively with the Fe electrodes. It was concluded that electrocoagulation can be applied as pretreatment for leachate.

Top *et al.* (2011) studied electrocoagulation treatment of the membrane concentrate with aluminium plate electrodes. 15.9 mA/cm² and 30 min came most favourable current density and time for removing COD, color and phosphorus and their removal efficiencies were 45%, 60% and 94.8% respectively. So these results signifies that electrocoagulation can further treat the nanofiltration concentrate of leachate.

Chiang *et al.* (1995) investigated that electrochemical process is a promising technology for the treatment of low BOD/COD ratio (0.2) leachate. 92% of COD get removed by the operating condition of 15 A/dm² and 240 mins with SPR anodes. 7500 mg/l additional chloride also added due to indirect effect of chlorine and hypochlorite on the removal of ammonium. Addition of sulphate also tried to get better results but it failed.

Table 1. Relationship among landfill age, leachate characteristics and treatment technologies

| Parameters | Leachate type | | |
|--------------------------|---------------|---------------|-----------|
| | <5 (young) | 5-10 (medium) | >10 (old) |
| Landfill age (years) | <5 (young) | 5-10 (medium) | >10 (old) |
| pH | <6.5 | 6.5-7.5 | >7.5 |
| COD (mg/l) | >10,000 | <10,000 | <5,000 |
| COD/TOC | <2.0 | 2.0-2.7 | >2.0 |
| BOD ₅ /COD | >0.5 | 0.1-0.5 | <0.1 |
| VFA (% TOC) | >70 | 5-30 | <5 |
| Biological treatment | Good | Fair | Poor |
| Chemical oxidation | Fair-Poor | Fair | Fair |
| Chemical precipitation | Fair-Poor | Fair | Poor |
| Activated carbon | Fair-Poor | Fair-Poor | Good |
| Reverse osmosis | Fair | Good | Good |
| Coagulation-flocculation | Fair-Poor | Good-fair | Good |

Source: Amokrane *et al.* (1997)

2. Materials and Methods

2.1 Chemicals

All the chemicals used in this study were of analytical grade. Chemicals such as concentrated sulphuric acid (98%), ferrous ammonium sulphate, potassium dichromate, nitric acid, ferric chloride, glycerol, sodium chloride, ammonium chloride, ammonium hydroxide, calcium chloride, sodium bicarbonate were purchased from Merck chemicals, Mumbai.

2.2 Analytical methods

pH and turbidity of the leachate were measured using a digital pH meter (APX 175 E/C, Control Dynamics, India) and turbidity meter (2100P, Hach, USA) respectively. COD was determined by the standard closed reflux method using a COD reactor (DRB200, Hach, USA). Total organic carbon was measured using a TOC analyzer (TOC-VCSH, Shimadzu, Japan). The BOD₅ of leachate were determined by the modified Winkler's method. Fe, Na, Cd, Ag, Al, Mn and other metals were analysed by the ICP-AES method. Sample was digested with concentrated HNO₃ in a Mars6 microwave digester (CEM, USA). Easy plus vessels were used to keep samples in the microwave. Time was set to 12 minutes to reach 180°C and allowed to hold for 15 minutes. And then samples were allowed to cool down. Then solution was filtered and then analysed by the ARCOS ICP-AES (Spectro, Germany) at sophisticated analytical instrument facility, IIT Bombay.

2.3 Experimental study

Electrocoagulation of landfill leachate samples was performed by using aluminium and iron electrodes. Electro coagulator was fabricated from plexiglas having total volume of 735 cm³ with dimensions of 70 mm × 70 mm × 150 mm stirred at constant stirring speed of 200 rpm by magnetic stirrer. Two electrodes were used one acting as cathode and other as anode with dimensions of 50 mm width, 160 mm height with 2 mm thickness. The electrodes used were of aluminium and iron with purity of 99.2% each. Total effective area of electrodes was 90 cm² and electrodes were dipped upto height of 90 mm. The spacing between electrodes was maintained 50 mm. Electrodes were connected to the DC supply of capacity 6 Ampere. Fig.1 shows the diagram of experimental set up. For removing surface grease after every run from electrode, acetone was used. Landfill leachate was collected from Deonar landfill in Mumbai, India.

2.4 Characteristics of landfill leachate

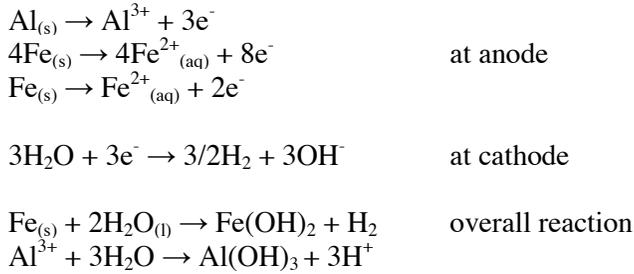
Landfill leachate samples were characterized for various parameters (like pH, COD, BOD, TS, TSS, TDS, alkalinity and turbidity). The COD of leachate sample was found to be 2300 mg/l which it tells that it was old/stabilized leachate. The BOD₅/COD ratio also found very less which signifies that it has very less biodegradable organics and may have high concentration of heavy metals (Table 1). So, ICP-AES of leachate sample was performed to estimate the concentrations of heavy metals in leachate. Total solids (TS) concentration was found to be around 10910 mg/l out of which almost 70% were in suspended form while rest (3400 mg/l) were in form of total dissolved solids.



Figure 1. Experimental setup for electrocoagulation

3. Results and Discussion

The main process which happens in electrochemical process is electrolytic reactions which produce coagulants in aqueous phase. Pollutants get adsorbed on these coagulants and then get removed in sedimentation. The reactions involved in this process are (Ilhan *et al.*, 2008)



Species such as “Al(OH)²⁺, Al(OH)₂⁺, Al₂(OH)₂⁴⁺, Al(OH)₄⁻, Al₆(OH)₁₅³⁺, Al₇(OH)₁₇⁴⁺, Al₈(OH)₂₀⁴⁺ forms initially due to reaction of Al³⁺ and OH⁻ and finally

comes in form of Al(OH)₃” (Gürses *et al.*, 2002). These are the species forms by Fe ions: “FeOH²⁺, Fe(OH)₂⁺, Fe₂(OH)₂⁴⁺, Fe(OH)₄⁻, Fe(H₂O)₂⁺, Fe(H₂O)₅OH²⁺, Fe(H₂O)₄(OH)₂⁺, Fe(H₂O)₈(OH)₂⁴⁺, Fe₂(H₂O)₆(OH)₄²⁺, which transform finally into Fe(OH)₃” (Mollah *et al.*, 2001).

3.1 Optimization of current density

Electrode materials used were aluminium and iron to compare their COD removal efficiency with Al and Fe chemical coagulants. They are cheap and readily available also. Current density was varied keeping electrolysis time as 60 mins with initial pH of 8. When the current density increased from 166 A/m² to 466 A/m² (Figs. 2 and 3) the removal efficiency of COD also increases from 27% to 47% in case of aluminium electrodes and 38% to 48% with iron electrodes. As the current density increases there is generation of more bubbles to remove the pollutants because of high current flow.

Table 2. Physical and chemical characteristics of landfill leachate

| Parameter | Methods | Concentrations |
|------------------------|-----------------------------------|----------------|
| pH | Digital pH meter | 8.0-8.5 |
| Color | - | Dark brown |
| Turbidity | Turbidity meter | 177 NTU |
| BOD | Modified winkler’s method | 238±30 mg/l |
| COD | Standard closed reflux method | 2304±152 mg/l |
| Conductivity | Conductivity metre | 17mS/cm |
| TOC | TOC analyser | 650±45 mg/l |
| BOD ₅ /COD | - | 0.10 |
| Total solids | Filtration and evaporation method | 10910±152 mg/l |
| Total dissolved solids | Filtration and evaporation method | 3400±61 mg/l |
| Total suspended solids | Filtration and evaporation method | 7510± 105 mg/l |
| Chlorides | Argentometric method | 349.53 mg/l |
| Total hardness | EDTA titrimetric method | 1051.52 mg/l |
| Total alkalinity | Titration method | 12450 mg/l |
| Fe | ICP-AES | 1.06 mg/l |
| Na | ICP-AES | 1412.43 mg/l |
| Cd | ICP-AES | 0.09 mg/l |
| Ag | ICP-AES | 0.346 mg/l |
| Al | ICP-AES | 6.64 mg/l |
| Mn | ICP-AES | 0.035 mg/l |
| Ba | ICP-AES | 0.480 mg/l |
| B | ICP-AES | 7.555mg/l |
| K | ICP-AES | 1107.2 mg/l |
| Ca | ICP-AES | 167.8 mg/l |
| Cr | ICP-AES | 0.024 mg/l |
| Sr | ICP-AES | 2.313 mg/l |
| Mg | ICP-AES | 118.52 mg/l |
| Zn | ICP-AES | Not determined |

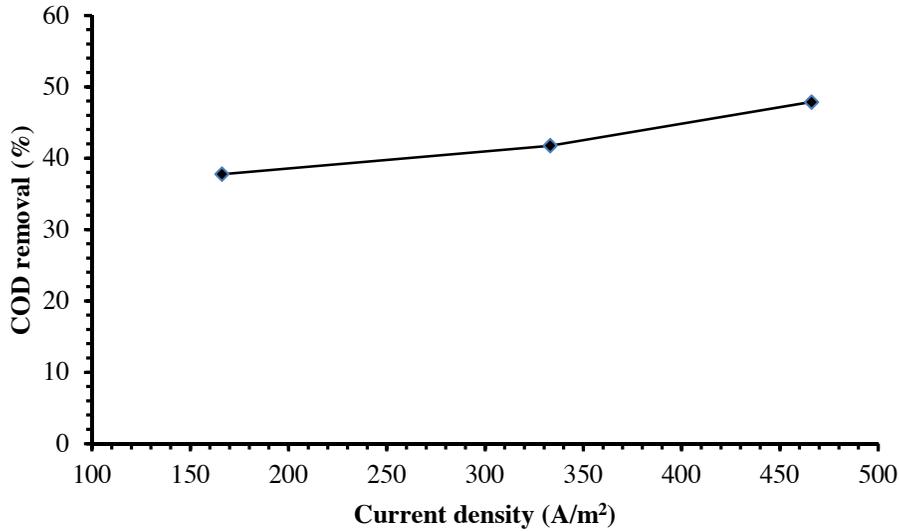


Figure 2. COD removal with current density using Fe electrode (Time = 60 mins, Initial pH = 8)

3.2 Optimization of electrolysis time

The electrolysis time affecting on COD removal was seen by varying time from 15 mins to 120 mins. The current density of 466 A/m² was maintained with initial pH of 8 while optimizing the time. When the time was varied from 15 mins to 120 mins (Fig. 5), the removal efficiency of COD increased up to 47% till 60 mins and then it became constant in case of aluminium

electrode. Iron electrode gave (Fig. 4) 53% removal efficiency of COD at optimized time of 90 mins. Unit energy consumption increases from 25.2 KWh/m³ to 202 KWh/m³ with the time from 15mins to 120mins for Fe electrode. Increment of electrolysis time increases concentration of Al and Fe ions and their hydroxide. Final pH is seen to increase with the time due to formation of OH⁻ ions shown in Figs. 8 and 9.

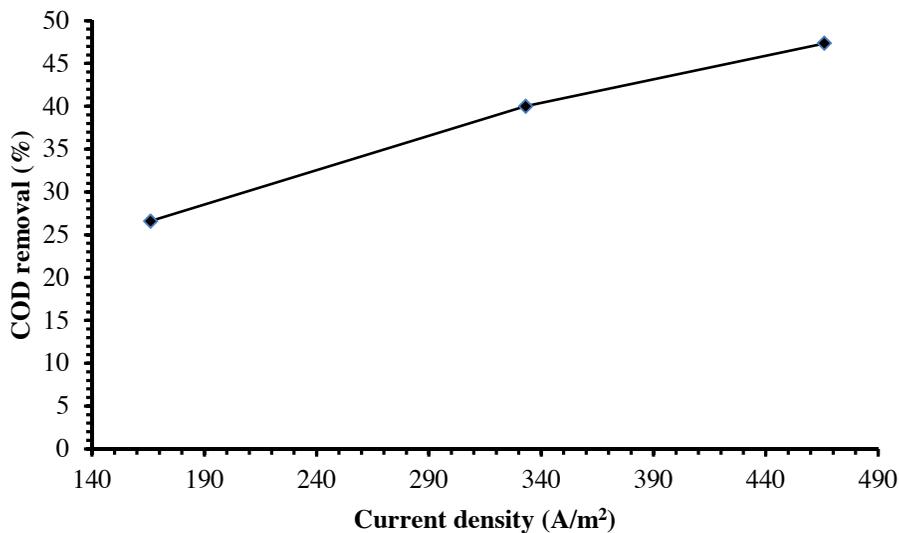


Figure 3. COD removal with current density using Al electrode (Time = 60 mins, Initial pH = 8)

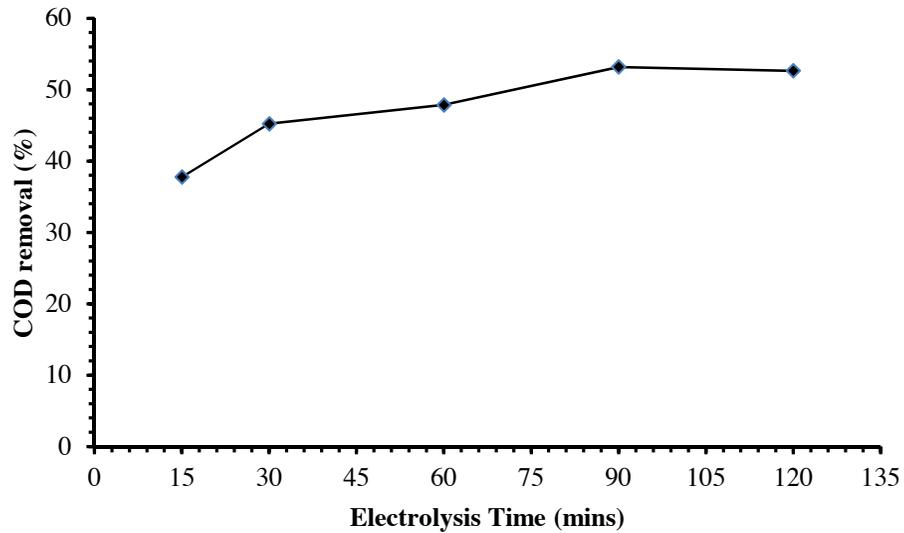


Figure 4. COD removal with electrolysis time using Fe electrode (Current density = 466 A/m², Initial pH = 8)

3.3 Optimization of initial pH

Initial pH has important role in the electrocoagulation process, so it needs to be optimized. pH optimization was done with optimized current density and optimized time of respective Al and Fe electrodes. Al electrode gave COD removal efficiency of 47% at optimum pH of 8 and 56% of COD removal at optimum pH of 6 with iron electrodes as shown in Figs. 6 and 7.

3.4 Energy consumption

Analysis of cost for performing electrocoagulation is done. The cost of electricity in Mumbai is INR 3.60 per KWh. The energy consumption is 151 KWh per m³ for removing 56% COD from the leachate by using iron electrodes. So the cost of electrocoagulation came

out to INR 542 per m³ volume of treatment of leachate (Feng *et al.*, 2007). The limitation of cost analysis is that the cost of electricity needed in electrocoagulation for stirring is not included and the cost of Iron electrodes is also not included.

4. Conclusions

The performance of electrocoagulation was evaluated as a pretreatment of landfill leachate. Iron and aluminium electrodes were used for performing electrocoagulation. Iron electrode with current density of 466A/m² gave 56% removal of COD at optimized time of 90 mins and pH of 6. Aluminium electrode on same current density gave 47% removal of COD at optimized time of 60 mins and pH of 8. Iron electrode gave higher removal efficiency of COD as compared to aluminium electrode.

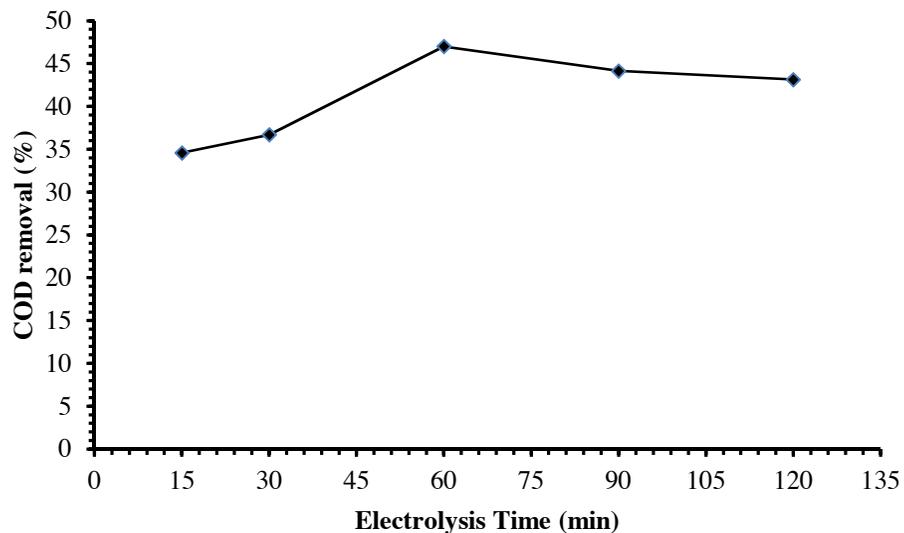


Figure 5. COD removal with electrolysis time using Al electrode (Current density = 466A/m², Initial pH = 8)

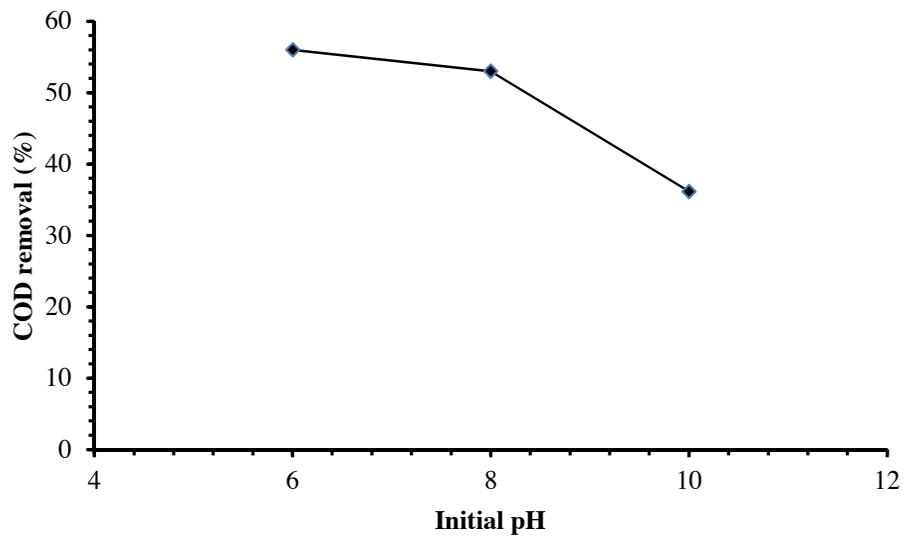


Figure 6. COD removal with pH using Fe electrode (Current density = 446A/m², Electrolysis time = 90 mins for Fe)

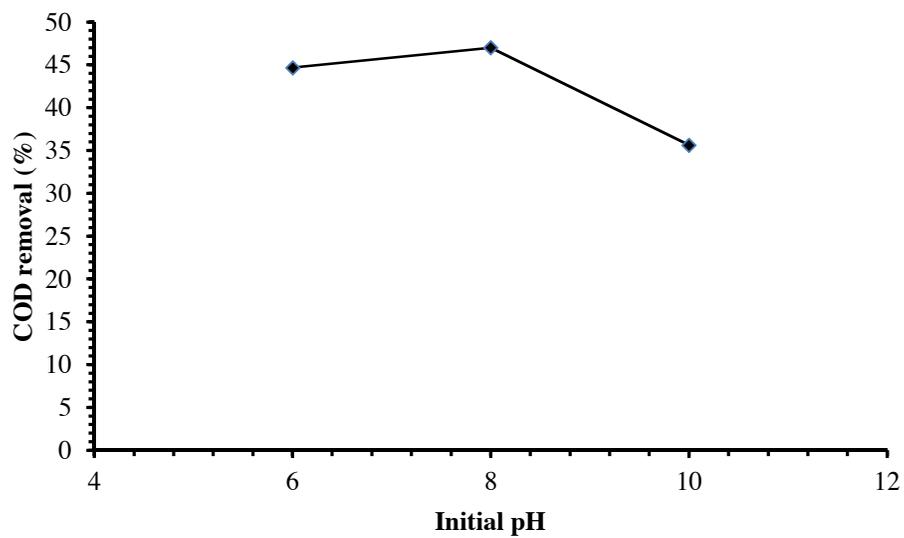


Figure 7. COD removal with pH using Al electrode (Current density = 446A/m², Electrolysis time = 60 mins for Al)

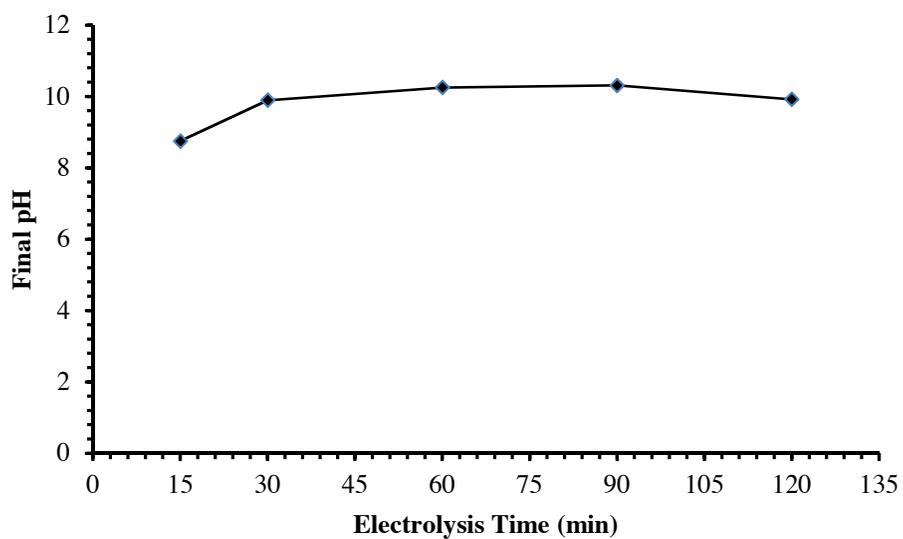


Figure 8. Final pH with electrolysis time using Fe electrode

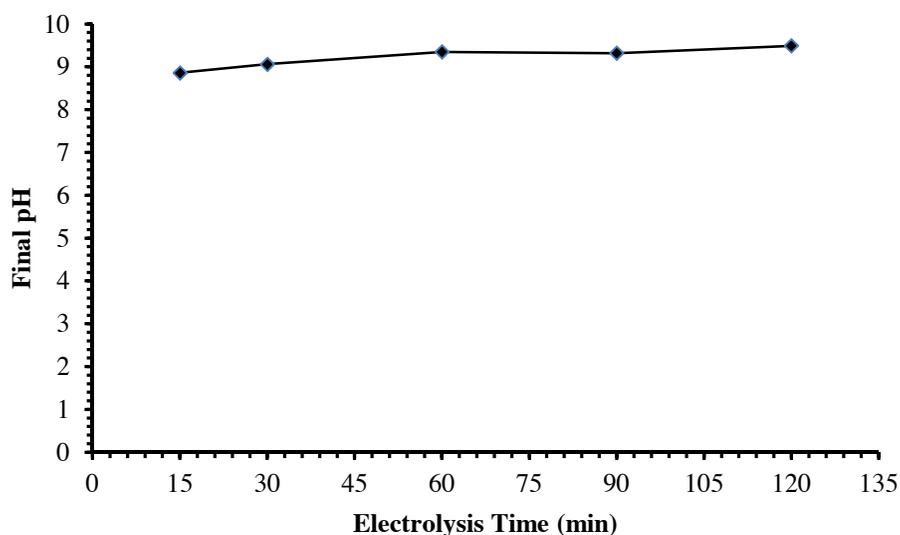


Figure 9. Final pH with electrolysis time using Al electrode

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