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"Comparative Study of Landfill Leachate Treatment by Electrocoagulation and Electro Fenton Processes"

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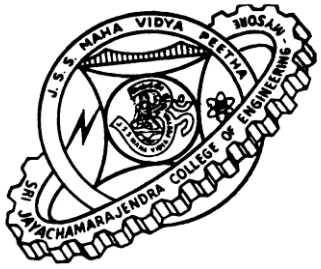

DEPARTMENT OF ENVIRONMENTAL ENGINEERING

SRI JAYACHAMARAJENDRA COLLEGE OF ENGINEERING

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SYNOPSIS

Landfill leachates consist of a complex mixture of organic and inorganic components which can be contain toxic and hazardous contaminants. Hence, landfill leachates have to be collected and treated. Since the variations in volume and composition, the conventional treatment methods are inadequate and its treatment is quite hard to supply the discharge standards. Therefore more effective treatment methods have been proven to treat leachate Leachate generation is an inevitable consequence of the deposition of solid wastes in sanitary landfills. It is the result of rainwater percolation through wastes, that extracts and brings is very complex and depends mainly on the type of solid wastes that are deposited with it several pollutant materials dissolved and in suspension. Sanitary landfill leachate composition depends on climatic conditions and the age of the sanitary landfill. Inadequate leachate management involves considerable risks, particularly contamination of water resources at the surface and groundwater and soils.

An effective treatment process is always in demand to destroy the recalcitrant substances. In the light of this problem, related to the leachate treatment, there is a need to find out a techno-economically feasible solution for the treatment. Hence, novel electrochemical oxidation technique is proposed for treatment of high strength leachate. Electrochemical technology has shown to be a promising method for the destruction of organic pollutants in wastewater and there is no need for adding extra chemicals. In addition, the high selectivity of the electrochemical process prevents the production of unwanted by-products.

It is also evident that, the earlier research works on Electrochemical treatment have been conducted using expensive electrodes such as titanium and platinum and haven't focused on further treatability studies of the treated effluent in terms of BOD to COD ratio. Hence, it was found necessary to employ certain low cost electrodes i.e., Iron, Stainless Steel and Aluminum electrodes to study their efficiency/applicability in the Electrochemical oxidation process to obtain maximum BOD to COD ratio and COD removal efficiency with optimized energy consumption. Fenton reagent is also proposed in electrochemical treatment to improve the COD, BOD and color removal. Electrochemical oxidation of leachate is one such area wherein it requires immense research work. Also, aforementioned literature clearly reveals that, not much work has been carried out, for the treatment of landfill leachate effluent using low cost electrodes in a bench scale electro-oxidation reactor. Electro Fenton is also one of the powerful and environmentally friendly

emerging technologies for the remediation of wastewaters containing organic, especially aromatic compounds In this context this research is taken for the treatability of leachate using different low cost electrodes under varying operational conditions also with Electro Fenton process.

The expected outcome of this present study is evolving technology for treatment of landfill leachate. The sludge obtained from treatment can be utilized in making fuel briquettes which could be fired in the boilers/incinerators/furnaces to recover its energy value. The bottom ash obtained after incineration of sludge may be used for blending with organic manure for use in agricultural/horticultural fields or may be blended with the cementitious material (clay/coal fly ash) to make bricks ceramic tiles for the building industry for secondary purposes.

Significance of the Research

In cities, pollution activities are high and are affecting human health as well as the environment. Mysore is the second largest city in Karnataka and commonly known as city of palaces and gardens. In recent years the city has grown extensively and the growth rate is considerably high over the past decades. The population has been increasing at a compounded annual rate in the last two decades, which is higher in comparison to the population growth of the state of Karnataka. The authorities have made adequate efforts to improve the city's infrastructure in terms of providing adequate potable water supply, solid waste management, sanitation, roads, and improving overall hygiene of the city. It is necessary to understand the earth surface activities such as sewage water flow, municipal solid waste management, landfill leachate and sludge Characteristics.

Present scenario, landfill leachates are very harmful to environments and it is necessary to treat while discharging to the ground. In an unlined landfill, the pollutants in soil remain in direct contact with the soil for relatively longer periods and hence the nature of soil pollution differs from that of air or water pollution. The unscientific management of municipal solid waste will lead to contamination of the soil. The presence of pollutants in the soil can change the desirable engineering properties of the soil. Reliable detection of leachate migration from the landfill is necessary to control widespread pollution of soil. If the impact of landfill leachate from municipal solid waste on engineering properties of soils can be assessed. An attempt has been made to assess the treatment of landfill leachate by using Electrocoagulation process.

The present research work indicates that the landfill leachate was treated by Electrocoagulation techniques to improve the removal efficiency of color and COD etc. The sample of landfill leachate was collected from dumpsite which is located in Vidyaranyapuram, Mysuru city, Karnataka state, India. In this research work a comparative study has been done to know the performance efficiency of different electrodes like Iron (Fe), Stainless Steel (SS) and Aluminum (Al). In addition to that optimization technique was employed to optimize the operating conditions, and also which individual parameters are effects on the output responses.

The main objective of the present research work is to carry out comparative experimental studies for the treatment of landfill leachate using different types of electrodes by Electrocoagulation (EC) and ElectroFenton (EF) processes.

Objectives of the present research work

1. Comparative studies have been performed, to maximize removal efficiency of Color and COD with different electrodes such as stainless steels. Aluminium and Iron.
2. To optimize the process parameters such as pH, current density, electrolysis duration and electrode distance in EC batch reactor on landfill leachate by Electrocoagulation process.
3. To carryout Electro-Fenton process with better performed electrode for achieving maximum removal efficiency of COD and Color.
4. To carryout kinetic studies and statistical analysis for parameters involved in the EC and EF studies.
5. Design of experiment (DOE) techniques are incorporated in order to optimize the operating parameters.

Research Gap

The earlier research works on electrochemical treatment have been conducted using expensive electrodes such as titanium and platinum and haven’t focused on further treatability studies of the treated effluent in terms of BOD to COD ratio. Hence, it was found

necessary to employ certain low-cost electrodes i.e., Iron, Stainless Steel and Aluminium electrodes to study their efficiency/applicability in the Electrocoagulation process to obtain maximum BOD to COD ratio and COD removal efficiency with optimized energy consumption. Fenton reagent is also proposed in Electrocoagulation treatment to improve the COD, BOD and color removal. Design of experiment (DOE) techniques is incorporated in order to optimize the operating parameters. Sparse research in open literature focusing of 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 BOD/COD ratio.

Outline of the research work

Chapter-1 This chapter explains the background of the landfill leachate. Source of solid waste, generation of leachate and its characteristics, environment impacts from solid waste and landfill leachate, Treatment employed for landfill leachate, significance the present study, Objectives of the research work, Research gap and outline of the research work.

Chapter-2 This chapter deals with the literature review from the experts, which involves research background of EC, principles and mechanism of EC process, advantages and disadvantages of EC process and highlights the novel electrocoagulation technology used by the previous researchers for different industrial wastewater as both treatment and pre-treatment method. It also includes the critical review of EC reaction mechanism involved in degradation of pollutants using different electrode assembly and effects of various operating parameters on EC process. The typical findings observed in the previous research work related to electrochemical and electro Fenton reaction mechanisms involved in wastewater treatment has been discussed. Recent advancements and future scope and design of experiment (DOE) also incorporated on this chapter.

Chapter 3 includes the material and methodology adopted in the present research work. Detailed laboratory scale EC batch reactor setup which is include from collection of samples to electrocoagulation process of landfill leachate wastewater with and without Fenton reagent. In addition to that detailed information about equipment's used in research work.

Chapter 4 In this chapter explains result and discussion of experimental work. Comparative study has been performed with different electrode like Iron (Fe), Stainless Steel (SS) and Aluminium (Al) with EC process. Most of the data have been represented in graphical form. In addition to that Main effect plots are drawn by using optimization technique, to know the significant terms for each individual output response.

Chapter 5 lists the summary of the work done and conclusions drawn from the present study. It also includes major recommendations for further study.

LIST OF RESEARCH PUBLICATIONS

INTERNATIONAL JOURNALS

- **Bharath M**., B. M. Krishna, and B. Manoj Kumar. "A review of electrocoagulation process for wastewater treatment." *International Journal of ChemTech Research* (2018).

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1. INTRODUCTION

General

Landfill leachates consist of a complex mixture of organic and inorganic components which can be contain toxic and hazardous contaminants. Hence, landfill leachates have to be collected and treated. Since the variations in volume and composition, the conventional treatment methods are inadequate and its treatment is quite hard to supply the discharge standards. Therefore more effective treatment methods have been proven to treat leachate (Selin Top et al., 2010).

Leachate generation is an inevitable consequence of the deposition of solid wastes in sanitary landfills. It is the result of rainwater percolation through wastes, that extracts and brings with it several pollutant materials dissolved and in suspension. Sanitary landfill leachate composition is very complex and depends mainly on the type of solid wastes that are deposited, the climatic conditions and the age of the sanitary landfill. Inadequate leachate management involves considerable risks, particularly contamination of water resources, at the surface and groundwater, and soils.

A common treatment for sanitary landfill leachates comprises biological reactors with nitrification/denitrification steps, followed by membrane technologies. However, due to variability in the quality and quantity of leachate throughout the life span of the treatment plant, these conventional treatments become ineffective. Thus, it is necessary to implement technologies that can be adjusted to the in situ needs. Electrochemical technologies have shown high efficiency in the elimination of persistent pollutants and several studies have described the application of electrochemical methods in wastewater treatment

A promising electrochemical method that can be used in wastewater treatment is the anodic oxidation. Despite several different materials are being used as anodes in the oxidation of persistent pollutants, the best results are obtained with boron-doped diamond (BDD) anodes, due to their unique chemical, electrochemical and structural stabilities that allow their use at high potentials, where most organic pollutants can be oxidized. There

are already several reports describing the application of electrochemical oxidation with BDD anodes for the treatment of landfill leachates (Annabel Fernandes et al., 2013).

Landfill leachate was formed by the complex physicochemical and biochemical transformations of the solid waste deposited at the sanitary landfill as well as by percolation of atmospheric water thorough the waste body of the sanitary landfill. It is characterized by dark color, unpleasant odor, high conductivity, high concentration of organic bio-refractory compounds and ammonia as well as increased to high concentration of heavy metals. Due to the complex nature of the effluent a combined treatment approach should be applied. summarized the methods used for the treatment of landfill leachate. Among them biological methods based on either aerobic or anaerobic processes, membrane technologies (reverse osmosis, ultrafiltration, nanofiltration), physicochemical methods (flotation, chemical precipitation, coagulation/flocculation, adsorption, stripping, chemical oxidation), advanced oxidation processes and various electrochemical treatment approaches were used most commonly (Visnja Orescanin et al., 2012).

Landfill effluents (leachate) need to be pre-treated on site to meet the standards for its discharge into the sewer or its direct disposal into surface water. In the world the problem of leachate treatment has been existed for sometime now, but a universal solution has not been found. The aim of this study is to make a review on the state of art in landfill leachate treatment and provides a comparative evaluation of various treatment processes (A. Abdulhussain Abbas et al., 2009)

2. LITERATURE SURVEY

Leachate production and characteristics:

Rainfall is the main contributor to generation of leachate. The precipitation percolates through the waste and gains dissolved and suspended components from the biodegrading waste through several physical and chemical reactions. Other contributors to leachate generation include groundwater inflow, surface water runoff and biological decomposition. Liquid fractions in the waste will also add to the leachate as well as moisture in the cover material. Moisture can be removed from the landfill by water consumed in the formation of landfill gas, water vapor removed in the landfill gas and leachate leaking through the liner.

The quantity of Leachates are depend on rainwater percolation through wastes, biochemical processes in waste's cells, the inherent water content of wastes and its degree of compaction into the landfill tip. The production is generally greater whenever the waste is less compacted, since compaction reduces the filtration rate. There are many factors affecting the quality of leachates that is age, precipitation, seasonal weather variation, waste type and composition. In particular, the composition of landfill leachates varies greatly depending on the age of the landfill. There are three types of leachates have been defined according to landfill age. As landfill age increased, organics concentration (COD) in leachate decreased and increase of ammonia nitrogen concentration. Landfill leachates from old sites are usually highly contaminated with ammonia resulting from the hydrolysis and fermentation of nitrogen containing fractions of biodegradable refuse substrates. The existing relation between the age of the landfill and the organic matter composition may provide a useful criterion to choose a suited treatment process. In general, leachates may contain large amounts of organic matter (biodegradable, but also refractory to biodegradation), where humic-type constituents consist an important group, as well as ammonia-nitrogen, heavy metals, chlorinated organic and inorganic salts.

The characteristics of the landfill leachate can usually be represented by the basic parameters COD, BOD, the ratio BOD/COD, pH, Suspended Solids, Ammonium nitrogen,

Total Kjeldahl Nitrogen (TKN) and heavy metals. Recirculation of leachate will produce stabilized leachates containing relatively low concentrations of degradable carbon compounds but high concentrations of ammonia therefore, COD and BOD will be removed, but ammonia concentrations will climb (A Abdulhussain Abbas et al., 2009)

Landfill Leachate Treatments:

Leachate Channeling:

Combined Treatment With Domestic Sewage:

One common means of leachate disposal is piping into the sewer system for discharge into the sea or, preferably, for combined treatment with domestic sewage at conventional sewage plant. It was preferred for its easy maintenance and low operating costs. However, this option has been increasingly questioned due to the presence in the leachate of organic inhibitory compounds with low biodegradability and heavy metals that may reduce treatment efficiency and increase the effluent concentrations. An argument in favor of this alternative treatment is that nitrogen (brought by leachate) and phosphorus (brought by sewage) don't need to be added at the plant. Among the few studies published, authors tried to optimize the volumetric ratio of leachate in the total wastewater. Combined treatment is investigated by using a Sequencing Batch Reactor (SBR) consisting of filling, anoxic, toxic and settling phases. When the ratio of sewage to leachate was 9/1, nearly 95% BOD and 50% nitrogen removals were obtained at the end of the daily cycles. COD and NH_3-N reduction decreased with increasing landfill leachate/domestic wastewater ratio. Moreover, the effluent quality may be improved with Powdered Activated Carbon (PAC) addition, particularly if the leachate input exceeds 10% (A. Abdulhussain Abbas et al., 2009)

Electrochemical Treatment.

Electrochemical treatment such as membrane electrodialysis has also contributed to environmental protection, the electrodegradation of stabilized land fill leachate was investigated by employing a flow electrochemical reactor. Using a constant flow rate of 2000 L/h for 180 min and at a current density of 1160 A/m^2, the maximum removal of COD and NH_3–N with initial concentrations of 1855 and 1060 mg/L was found to be 73

and 49%, respectively, The results suggest that electrodegradation was an alternative means to breakdown recalcitrant organic compounds in landfill leachate. Due to high energy consumption, however, this technology is more expensive than other treatment methods. As a result, this treatment technique has been investigated less extensively for the treatment of stabilized leachate (Tonni Agustiono Kurniawan et al., 2005).

Electrochemical technology became an inevitable treatment step for the resistant contaminants in wastewater. Especially, it is used efficiently in the treatment of textile, food, metal and galvanization, and petrochemistry industry effluents. Electrochemical processes are supposed to have a wide perspective. It is considered that this technology will have a widespread usage in water and wastewater treatment due to such its characteristics as less equipment requirement, shorter treatment period, no chemical matter need and less sludge formation as a result of all this. Besides all these advantages, method has some disadvantages such as exchange of anodes as electrodes consume and high operation costs where electricity is expensive. First of all, the general characteristics of the leachate collected from a younger domestic solid waste lot were analyzed, and the treatment efficiency of a pilot electro-coagulation reactor with respect to TOC, COD and color parameters was searched. The results, which were obtained from chemical-coagulation method have been compared with the results, which were obtained by the electro-coagulation method. By comparing the results obtained and the results of chemical-coagulation process, the place of the new process that is electro-coagulation process, was determined (Sevil Veli et al., 2007).

Electrochemical Methods electrochemical oxidation is one of the most widely used for the treatment of landfill leachate. An important advantage of this method is the possibility of direct oxidation of the heavily degradable organic matter into CO_2 and water. Electrooxidation of the pollutants present in the wastewater is carried out through two different mechanisms:

(i) Indirect anodic oxidation,

where oxidation of organic matter and ammonia takes place by means of electrochemically generated reactive oxygen species such as chlorine and hypochlorite,

hydrogen peroxide, ozone. Electrooxidation could be demonstrated with the following set of the reactions:

Anode

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

$6HOCl + 3H_2O \rightarrow 2ClO_3^- + 4Cl^- + 1.5O_2 + 6e^-$ (2)

$2H_2O \rightarrow O_2 + 4H^+ + 4e^-$ (3)

Summary reaction:

$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^-$ (4)

$HOCl \rightarrow H+ + OCl^-$ (5)

Cathode

$2H_2O + 2e^- \rightarrow 2OH^- + H_2$ (6)

$OCl^- + H_2O + 2e^- \rightarrow Cl^- + 2OH^-$ (7)

In the case of using iron anode indirect anodic oxidation of organic constituents was also possible by hydroxyl radicals generated electrochemically in the Electro-Fenton reaction from Fe 2^+ and hydrogen peroxide.

(ii) Direct anodic oxidation, which involves two processes:

(a) electrochemical conversion and (b) electrochemical combustion. During the electrolysis, two kinds of electrochemically generated active oxygen can occur at the anode. One is chemically bound in the structure of the anode and is responsible for the chemical conversion of the organic matter while the other one is adsorbed on the surface of the anode in the form of hydroxyl radicals which promoting electrochemical combustion. The electrochemical conversion, leads to the partial oxidation of organic matter into more easily degradable compounds suitable for biological degradation while the electrochemical combustion leads to complete decomposition of the organic matter into CO_2 and water.

In the real conditions the removal of the organic matter and ammonia in the leachate was mostly achieved through the indirect anodic oxidation by free chlorine and sodium hypochlorite generated by the oxidation of chloride (initially present in the wastewater) on the anode. The percentage of the removal depends on the composition of the effluent (depending on the age of the landfill), anode material, current density, concentration of electrolytes (chlorides, sulfates) and the treatment time. In this way it is possible to remove over 90% of the COD, over 99% color and almost 100% ammonia. Better results are achieved by the combination of the electrooxidation and electrocoagulation (Visnja Orescanin et al., 2012).

Electrochemical Method

Electrochemical methods have attracted a great deal of attention basically due to the increased efficiencies that can be achieved using easy to operate and control, compact bipolar electrochemical reactors. Oxidizing agents such as hypochlorite, oxygen-based radicals, ozone and nitrogen oxides can be generated in situ in the electrochemical reactors. During these processes, organic pollutants typically found in industrial wastewaters can be destroyed by direct or indirect oxidation. However, the reactions that take place during the electrochemical treatment are rather complicated, and the electrochemical removal mechanisms of organic pollutants are not fully clarified (Elisabetta Turro et al., 2012).

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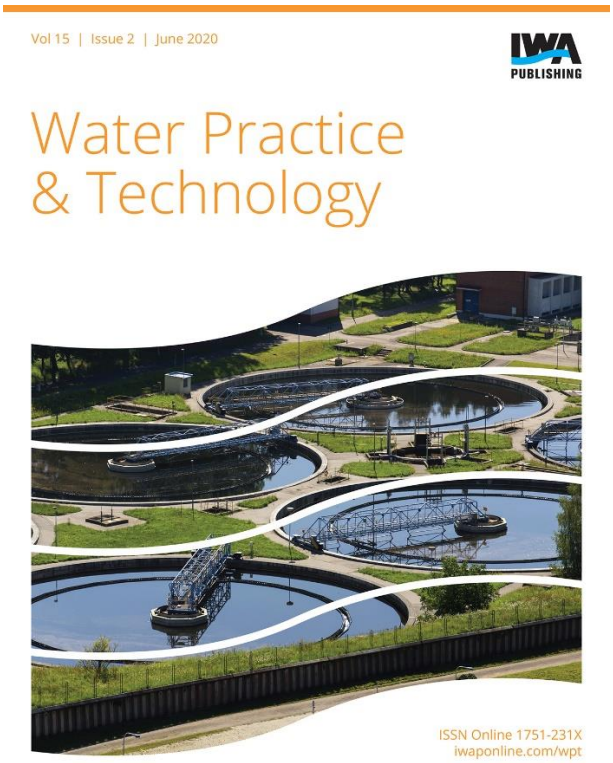

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Degradation and biodegradability improvement of the landfill leachate using electrocoagulation with iron and aluminum electrodes: A comparative study

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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

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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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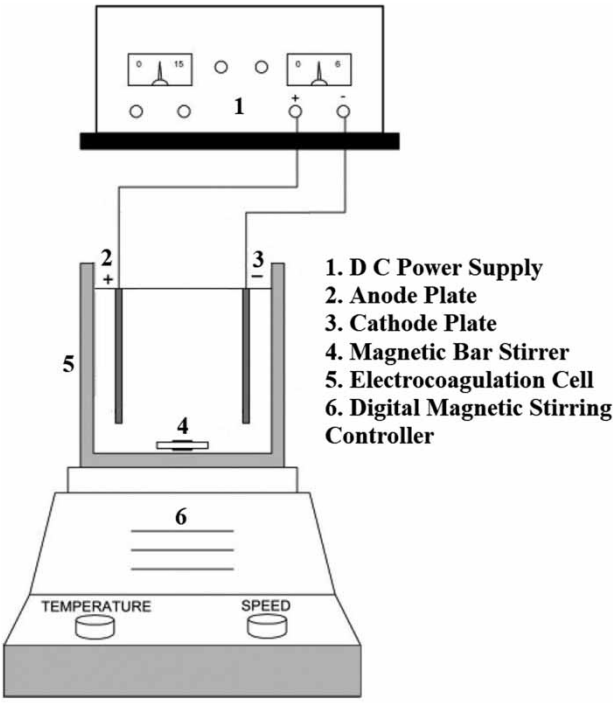

Figure 1 | Experimental set up of electrocoagulation treatment at a lab scale.

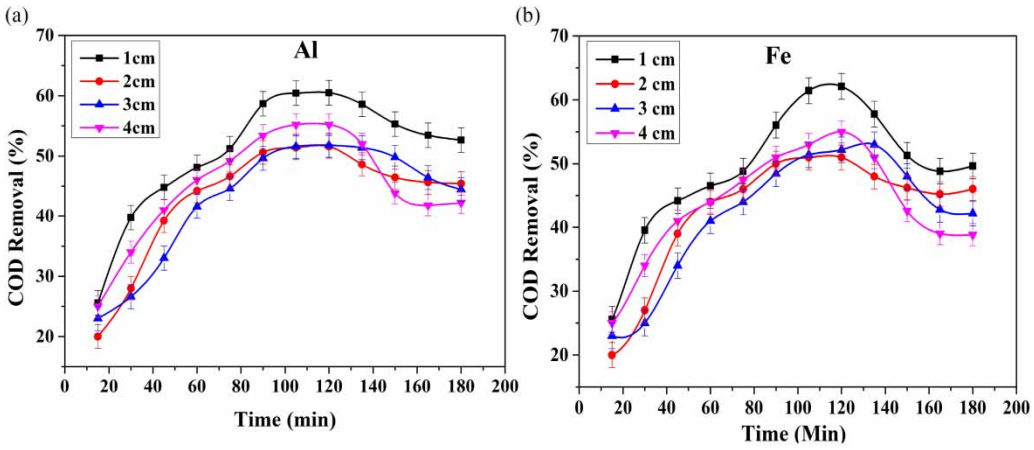

Figure 2 | (a) and (b) Effect of inter-electrode distance on percentage removal of COD in the leachate treatment by EC with Al and Fe electrodes.

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).

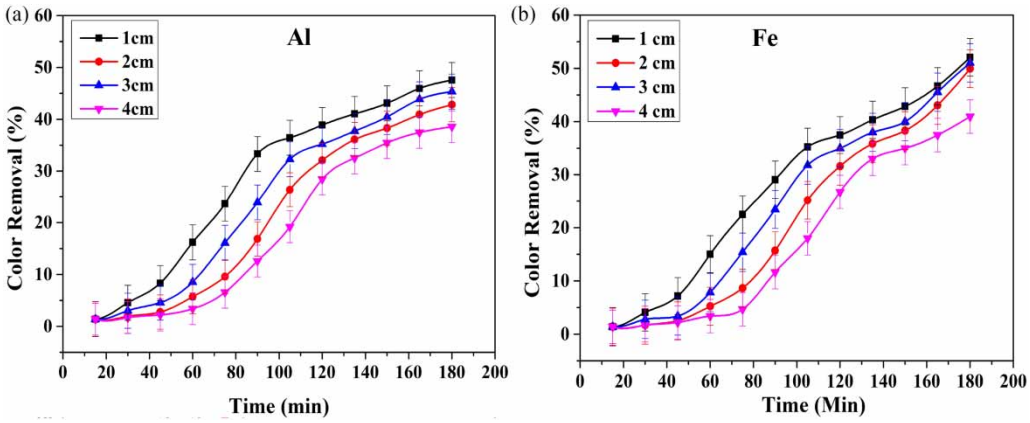

Figure 3 | (a) and (b) Effect of inter-electrode distance on percentage removal of Color in leachate treatment by EC with Al and Fe electrodes.

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

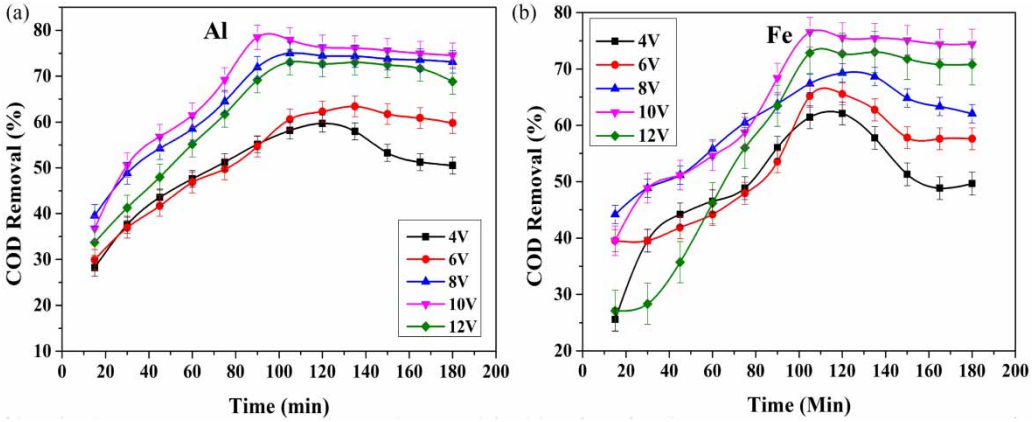

Figure 4 | (a) and (b) Effect of applied voltage on leachate treatment by EC (COD removal) for Al and Fe electrodes.

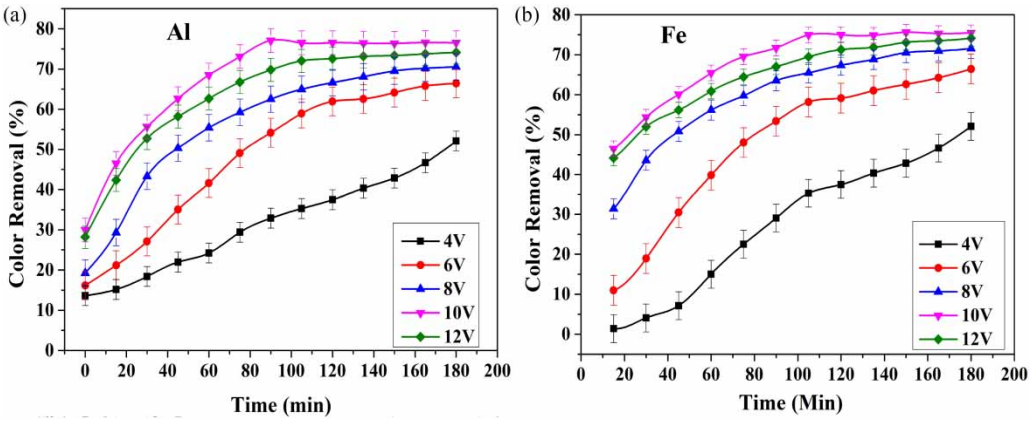

Figure 5 | (a) and (b) Effect of applied voltage on leachate treatment by EC (Color removal) for Al and Fe electrodes.

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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.

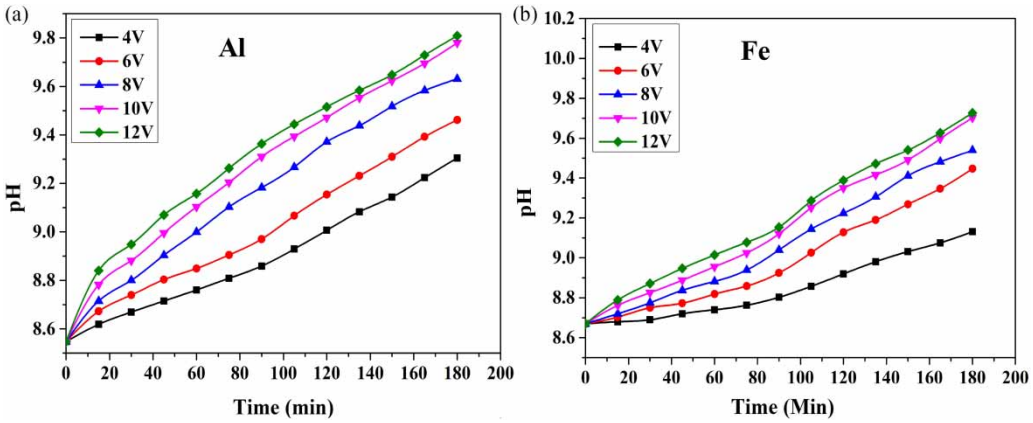

Figure 6 | (a) and (b) Effect of applied voltage on leachate treatment by EC (pH) for Al and Fe electrodes.

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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).

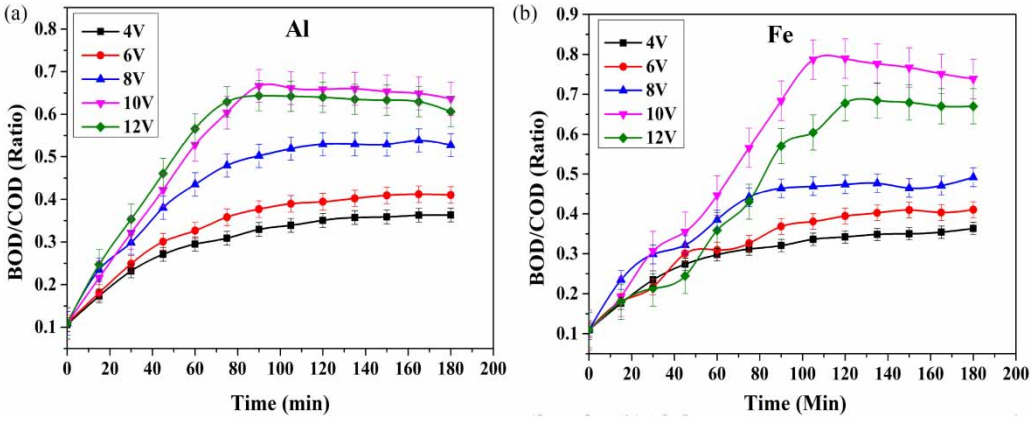

Figure 7 | (a) and (b) Effect of BOD/COD ratio on leachate treatment by EC for Al and Fe electrodes.

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

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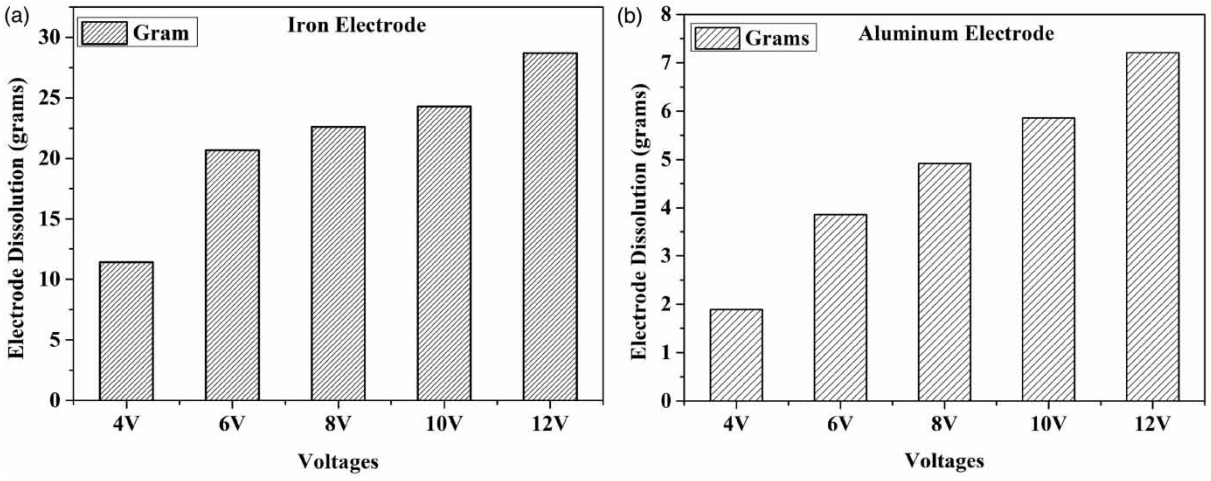

Figure 8 | (a) and (b) Effect of electrode dissolution pattern for Fe and Al electrodes.

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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Electrocoagulation Treatment for Removal of Color and Chemical Oxygen Demand in Landfill Leachate using Aluminum Electrode

Bharath M, Krishna B M, Shiva Kumar B P

Abstract*: The present research work mainly deals with the removal percentage of Color and Chemical Oxygen Demand (COD) on landfill leachate by using electrocoagulation (EC) process. An EC process was carried out with an aluminium electrode and it act as both anode and cathode. The study mainly targets the factors affecting on electrode material, electrolysis time, initial pH, applied voltage, inter-electrode distance. The experimental result reveals that there was raise in BOD/COD ratio from 0.11 to 0.66 and the maximum percentage removal achieved were COD and Color 78.4% and 77.0% respectively. The optimum inter-electrode distance 1cm with electrode surface area 35 cm2 and optimum electrolysis time of 90 min at optimum applied voltage 10V, stirring speed 250 rpm and pH is 9.3. These results showed that the EC process is appropriate and well-organized approach for the landfill leachate treatment.

Keywords* : Landfill leachate; Electrocoagulation; Aluminium electrode and Process parameters.

I. INTRODUCTION

Leachate is produced from rain water percolation through waste and decomposition of waste. A serious environmental problem can occur from landfill leachate discharge and it includes heavy metals, biodegradable/non-biodegradable carbon, organic/inorganic salt and recalcitrant. Many factors that influence the leachate quality, i.e., landfill age, seasonal weather, precipitation, type of waste, and composition.

The leachate composition mainly depends on the age of landfill. As the landfill age increases, COD and other organic concentrations are decreased, and ammonia nitrogen concentrations increased. Naturally, the landfill leachates are in the form of liquid and strongly odour, the physical appearance of leachates are orange or yellow cloudy liquid and offensive smell due to the presence of nitrogen, hydrogen, and sulphur rich organic species.

The landfill leachate has some basic parameters such as pH, suspended solids, biochemical oxygen demand (BOD), chemical oxygen demand (COD), ratio of BOD/COD and ammonia nitrogen etc. Stabilized leachate will be formed due to recirculation of leachate, which results degradable of carbon compounds but a higher concentration of ammonia, it improves COD and BOD and it will be eliminated. The classification and characterization of landfill leachate based on age are depicted in Table 1. Based on the literature survey, some of the treatment methods such as coagulation-flocculation [1], membrane processes [2-3] activated carbon adsorption [4]. combined physicochemical-nanofiltration [5]. biological treatment [6]. have been reported in the literature. Electrocoagulation (EC) process treating various types of wastewater, for example electroplating wastewater [7], Distillery wastewater [8] [9], Dairy wastewater [10]. The main importance of research work is to optimize the parameters like initial pH, electrolysis time, current density, inter-electrode distance on the landfill leachate treatment using EC process with aluminum electrodes.

Table 1 Landfill leachate classification of landfill age

Parameters	Leachate Type		
Landfill age (Years)	<5 Young	5-10 (Medium)	>10 (old)
BOD_5/COD	>0.5	0.1-0.5	<0.1
pH	<6.5	6.5-7.5	>7.5
COD (mg/l)	>10,000	<10,000	<5,000

II. EXPERIMENTATION

A. Study Area

Mysore city is located at 12.30°N 76.65°E by having an average altitude of 770 meters. Dumpsite situated vidyaranyapuram and 8 km away from the Mysore city.

Fig 1 Landfill leachate collection tank

Especially this site is used for dumping garbage from past 6-7 years, which has gathered waste of about 2,50,000 cubic meters, and dumping area is occupying 41.47 acres to accumulate wastages.

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The present study attempts to treat landfill leachate using Electrocoagulation process. The sample Landfill leachate was collected from the tank in which the leachate is coming from the pipes, as shown in Fig 1, and it was analyzed for various physical, chemical parameters. The initial characterization of the sample has been given in Table 2.

Table 2 Initial characterization of the landfill leachate Parameters

Sl. No.	Parameters	Concentration
1	pH	8.67
2	Conductivity	38.5 mS/cm
3	Turbidity	140NTU
4	Total solids	15800(mgL^{-1})
5	Total Dissolved Solids	14240(mgL^{-1})
6	COD	13760(mgL^{-1})
7	Phosphate	198.5(mgL^{-1})
8	Total suspended solids	1560(mgL^{-1})
9	Nitrates	95.5(mgL^{-1})
10	BOD	1503(mgL^{-1})
11	Chloride	6098(mgL^{-1})
12	BOD/COD	0.109 (mgL^{-1})
13	Color	8750 PCU

B. Experimental Setup for Electrocoagulation

Experiments are performed in a plexi-glass laboratory scale, Batch electrochemical reactor (11cm x14cm x13cm) of 2L capacity with the working volume 1.75L at room temperature. The reactor content was kept under complete mixed condition facilitated using a magnetic stirrer speed 250rpm to avoid concentration gradients. The T shaped electrode materials such as aluminum plates of size 5cm x 7cm are used in a monopolar arrangement as anode and cathode having 35 cm^2 effective surface area. For easy stirring, 2cm gap was kept between the bottoms of the electrodes. The analytical details were depicted in Table 3.

Table 3 Analytical Details

Parameters	Analytical technique/Method	Instruments/Equipment's Used, Make
pH	Digital pH meter	-------
BOD	27^o C, 3 days incubation/ Titrimetric/ Modified Winkler's method	
COD	COD digester (Open reflux system)/Titrimetric	Hach 389, USA
Color	Platinum-cobalt method	------
Solids	Gravimetry	Hot air oven
Chlorides	Argentometric method	Standard method
Conductivity	Conductivity meter	-----
Sulphate	Spectrophotometric method	UV spectrophotometer
Nitrate	Phenoloic disulphonic Acid Method	UV spectrophotometer
Phosphate	Ammonium Vandate/Molybdate	UV Spectrophotometer
DC Power Supply Unit	0-10 A, 0-15 V, DC power supply unit	APLAB, Regulated dual DC power supply LD3210.

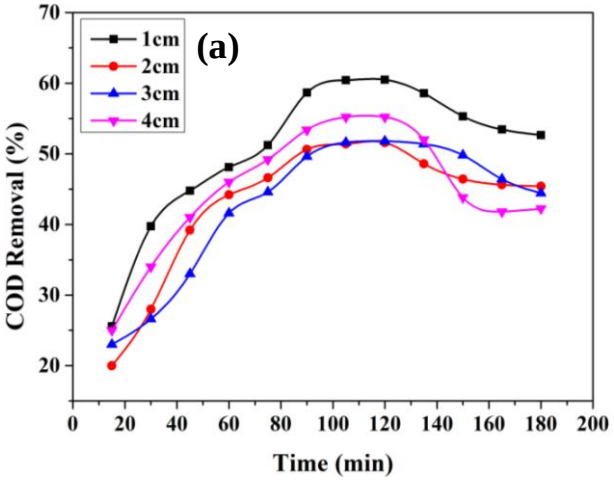
Before each treatment, the electrodes were cleaned and degreased. Experimental run conditions were maintained using DC power supply; the distance was varied from 1cm - 4cm by maintaining 4V. The total electrolysis duration was 180 min during electrolysis; samples were drawn at regular intervals of 15 min. Experiments were conducted by varying the effect of operating parameters like inter-electrode distance, electrolysis duration, voltage (current density). Electrocoagulation experimental setup is shown in Fig 2.

Fig 2 Experimental set up of electrocoagulation treatment in a lab scale

III. RESULT AND DISCUSSION

A. Effect of inter electrode distance on landfill leachate treatment by EC:

From Fig 3 (a-b) it is observed that the electrolysis time increases with an increase in the removal of COD and Color, respectively. Experiments are conducted with different inter-electrode distance, such as 1cm, 2cm, 3cm, and 4cm. The better performance obtained with 1cm inter-electrode distance. From that condition, the percentage removal of COD and Color 60.50% and 47.50% respectively is achieved.



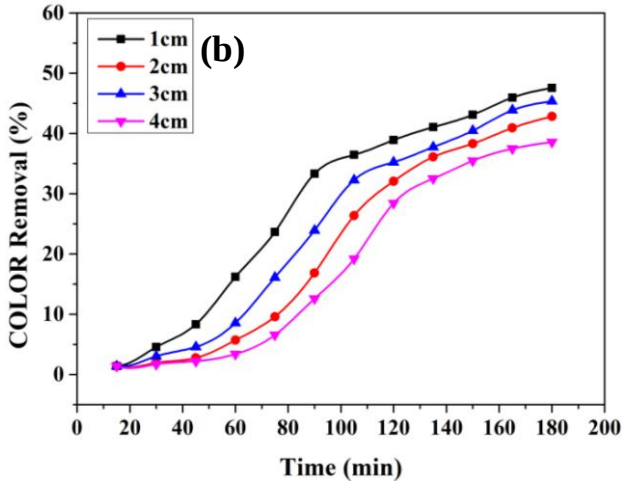
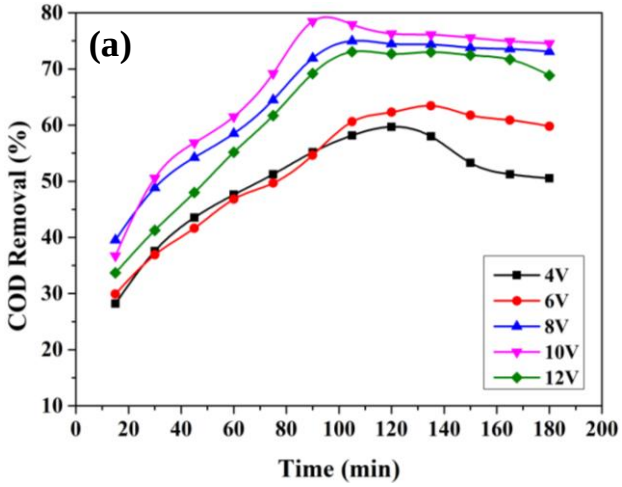


Fig 3 (a-b) Percentage removal of COD and Color with different distance

This is due to the shortest distance facilitates the higher removal efficiency; the similar results obtained by [11]. Some of the researchers reported as increased inter-electrode distance leads to higher power consumption and increased operating cost. The proper placement of electrode distance, which affects the current density. If the electrode distance is too close, the flow of fluid and solids are clogged that cause higher electrical resistance.

B. Effect of applied voltage on leachate treatment by EC Applied:

From Fig 4 (a-b) is noticed that the electrolysis time increases with an increase in the removal of COD and Color. EC experiments have been carried out at 4V, 6V, 8V, 10V, and 12V. From the experimental analysis, 10V exhibits higher removal efficiency and the percentage removal of COD obtained 78.48%. Color removal was achieved 77.09%. For 90min, indicating that increase the destabilization of a colloidal particle with an increase in electrolysis duration. The reaction between generated chlorine/hypochlorite that results in decolorization.



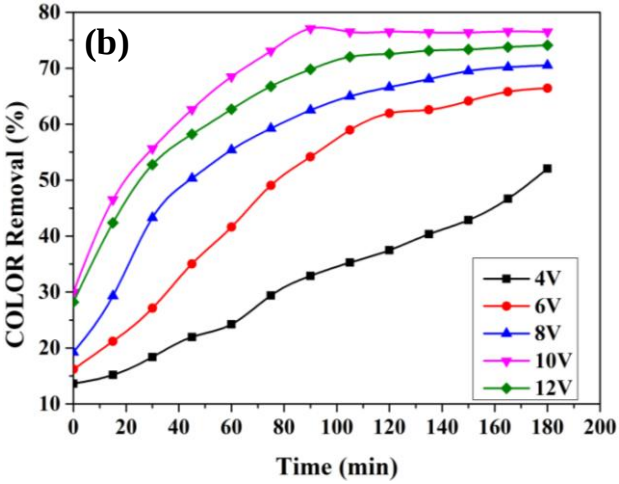


Fig 4 (a-b) Percentage removal of COD and Color with different voltage

Treatment efficiency also increased by increasing the current density and also which influence the higher rate of bubble generation and dosage of coagulant [12]. Operating voltage was not raised beyond 12V due to faster dissolution.

C. Effect of pH changes during an electrocoagulation process:

The pH value is strongly dependent on the electrocoagulation technique and initially, the pH value of leachate 8.67. The electrolysis time increases, pH also increases.

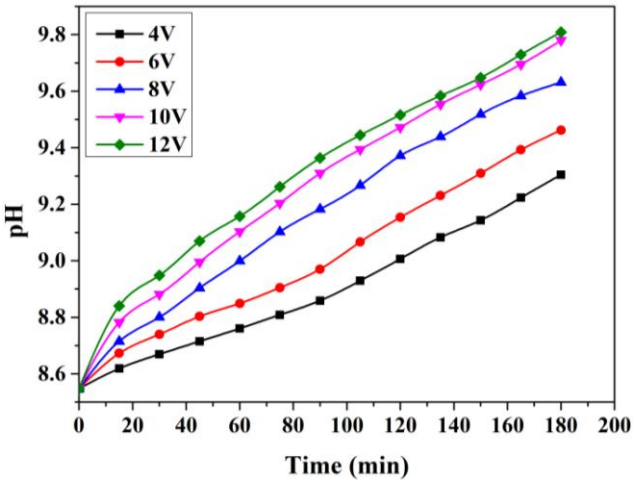


Fig 5 Effect of pH versus electrolysis time

From the experimental work, the better performance is observed at 90min of electrolysis time, and pH value shows 9.3 at 10V, as shown in Fig 5. pH of the leachate solution is evident in the alkaline range a slimy layer of floating contaminants is removed easily at the top, which leads to maximum removal of Color and COD. The pH progressively increased due to foremost activities of the cathode and pH of leachate solution is expected to be high after electrocoagulation treatment [13]. Some researchers found that COD removal is not much impact on the variation of pH. The alkaline condition is more effective for the treatment of landfill leachate wastewater [14]. Decolorization of leachate is very low at acidic pH condition and very high at neutral and alkaline pH condition.

D. Effect of BOD/COD ratio changes during the EC process:

From Fig 6, it is observed that there was a development in biodegradability of landfill leachate with an increase in the ratio of BOD/COD from 0.11 to 0.66. This is due to increasing voltage. Under different voltage, the other operating condition was altered, and the performance of the reactor also affected. The low BOD/COD ratio (0.11) in the effluent indicates that it contains recalcitrant substances which were not easily biodegradable or non- biodegradable material present in leachate.

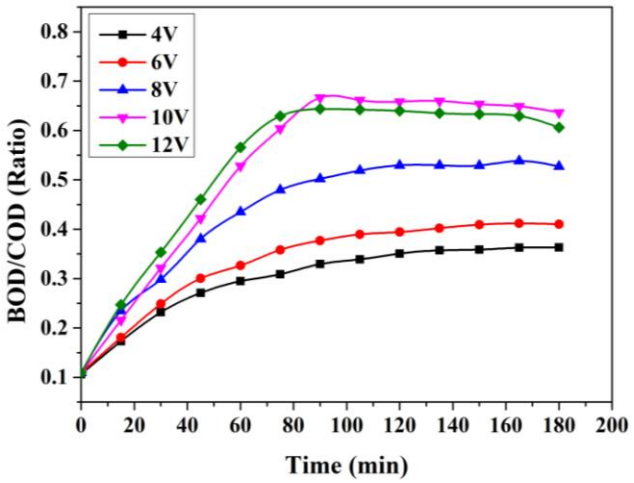


Fig 6 Effect of BOD/COD ratio vs. electrolysis duration

IV. CONCLUSIONS

The present study shows that the performance of electrocoagulation is an efficient process for treating landfill leachate. Aluminum electrodes were used for performing electrocoagulation. To optimize the process parameters such as electrolysis time, inter-electrode distance, applied voltage, and effect of initial pH. From that experimental work, some of the conclusions are drawn.

The maximum removal achieved was COD and Color, 60.50% and 47.50% respectively, at the shortest inter-electrode distance of 1cm.

The higher removal efficiency was obtained COD and Color, 78.48% and 77.09% respectively, at the optimum inter-electrode distance 1cm, optimum electrolysis time of 90 min. At optimum applied voltage 10V, stirring speed 250 rpm and pH 9.30. .

The electrocoagulation process is more effective for removal of Color and COD, BOD/COD ratio raises from 0.11 to 0.66.

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Electrocoagulation Treatment for Landfill Leachate using Stainless Steel Electrode

Bharath M, B M Krishna

Abstract*: Electrocoagulation (EC) process uses direct electric current source between metal electrode submerged in the effluent that results in electrode dissolution, with a suitable pH, metal ion can form a wide range of metal hydroxide and coagulated species that destabilized and dissolved contaminants absorbed. Electrocoagulation (EC) has been working for the percentage removal of BOD (Biochemical oxygen demand)/ chemical oxygen demand (COD) ratio, Color and COD on leachate in a batch Electrocoagulation reactor using stainless steel (SS) electrode. EC technology depends on so many factors such as electrode material, initial pH, applied voltage, inter-electrode distance, and electrolysis time. From the experimental work, results reveal that the maximum percentage of removal achieved were COD and Color 73.5% and 65.0% respectively and increasing BOD/COD ratio 0.11 to 0.62. The optimum inter-electrode distance 1cm with electrode surface area 35 cm2 and optimum electrolysis time of 120 min at optimum applied voltage 12V, stirring speed 250 rpm and pH 9.8. These results proved that the EC process is an appropriate and proficient approach for treating the landfill leachate.

Keywords: Landfill leachate; Electrocoagulation; Stainless Steel electrode and Process parameters.

I. INTRODUCTION

Leachate can be defined as the rainwater percolation through waste which generates effluent. Leachate may contain high dissolved solids, a large amount of organic matter, suspended solids, chlorinated organic/inorganic salts, ammonia-nitrogen, and heavy metals. The elimination of organic material, such as ammonium, BOD, color, and COD from leachate. There are so many factors that affect the leachate quality, such as seasonal weather, age, composition, precipitation, and waste type. In general, landfill leachate composition mainly depending on the landfill age. The treatment technology for leachate mainly includes physicochemical–nanofiltration processes [1], flocculation/precipitation [2], sequencing batch reactor (SBR) process [3].

Electrocoagulation (EC) has been involved for treating different types of process wastewater, for example, restaurant wastewater [4], electroplating wastewater [5], Distillery wastewater [6]EC process has been effectively used for the treatment and pollutants removal from industrial wastewater, municipal wastewater, and inorganic ion and heavy metal removed. The main importance of research work is to optimize the process parameters such as electrolysis time, inter-electrode distance, voltage (current density) and biochemical oxygen demand (BOD)/COD ratio, color and COD removal efficiencies. EC technique is an easy operation, simple equipment and produces less amount of sludge with an appropriate anode material, and the coagulants are generated by electrolysis oxidation that leads to the insoluble components and metal hydroxide which is capable of eliminating a huge range of contaminants from wastewater [7]. The mechanism of an EC cell can be defined by the following equations 1 to 4.

At the anode, metal is oxidized into cations:

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

$$Fe_{(s)} + 2OH^{-} \rightarrow Fe(OH)_{2(s)} \quad (2)$$

At the cathode, water is reduced into hydrogen gas and hydroxyl anions:

$$2H_2O + 2e^{-} \rightarrow H_{2(g)} + 2OH^{-}{}_{(aq)} \quad (3)$$

In the bulk solution

$$Fe^{2+}_{(s)} + 2OH^{-}_{(aq)} \rightarrow Fe(OH)_{2_{(s)}} + H_{2_{(g)}} \quad (4)$$

The objective of this research work is to remove the COD and Color from landfill leachate samples and to understand the effect of various operating parameters such as electrode material, initial pH, applied voltage (current density), electrolysis time and inter-electrode distance using EC process.

II. EXPERIMENTATION

A. Study Area for the Treatment of Landfill Leachate:

The area of study was selected for the study is in Mysuru city, Karnataka. 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 is being used from past 6- 7 years. The area consists of accumulated waste of about 2,50,000 cum and the area used for dumping of waste is about 41.47 acres.

The present study attempts to treat landfill leachate using Electrocoagulation process. The sample landfill leachate was collected in the tank wherein, and the leachate is coming from the pipes which have shown in Fig 1. The various physical and chemical parameters were analyzed in this study. The physical and chemical parameters in the initial characterization of the sample were shown in Table 1.

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Figure 1 Landfill leachate collection tank

Table 1 Initial characterization of the landfill leachate Pa rameters

Sl. No.	Parameters	Concentration
1	pH	8.64
2	Conductivity	36.5 mS/cm
3	Turbidity	160NTU
4	Total solids	14600(mgL-1)
5	Total Dissolved Solids	13552-14240(mgL-1)
6	COD	13760(mgL-1)
7	Phosphate	200.5(mgL-1)
8	Total suspended solids	1840(mgL-1)
9	Nitrates	90.5(mgL-1)
10	BOD	1519(mgL-1)
11	Chloride	7053(mgL-1)
12	BOD/COD	0.1104 (mgL-1)
13	Color	8750 PCU

B. EC Reactor Setup for Electrocoagulation

EC experiments were carried out in a plexi-glass laboratory scale. Batch electrochemical reactor (11cm x14cm x 13cm) of 2 L capacity with the working volume of 1.75L at room temperature was used in the setup.

The reactor was kept under the process of continuous agitation using magnetic stirrer with 250 rpm to avoid the formation of concentration gradients. The T shaped electrodes with the material of stainless steel (SS) plates with a size of 5 cm X 7 cm was used as both anode and cathode electrode having 35 cm2 effective surface area. At the bottom of the electrodes, the gap of 2 cm was maintained to facilitate continuous and easy stirring. Before each treatment process, the SS electrodes were cleaned and degreased. The power supply used to run all experimental conditions was DC power. The inter-electrode distance between anode and cathode electrode was varied from 1cm to 4cm, wherein the voltage used in the electrolysis process was 4V. The duration of the electrolytic process was done for 180 mins with a 15mins time interval. At every 15 min, the sample was collected for further process. The collected samples after electrolysis were used to analyze the process parameters such as current density, electrolysis duration, Color, COD, BOD/COD ratio, and pH. Among these analyzed parameters, pH, Electrolysis duration, the distance between electrode and voltage (current density) were optimized in this study. The experimental setup for electrocoagulation to the lab scale process has shown in Fig 2. The analytical details as shown in Table 2.

Figure 2 Experimental set up of electrocoagulation treat ment in a lab-scale

Table 2 **Analytical** details

Parameters	Analytical technique/Method	Instruments/Equipment's Used, Make
pH	Digital pH meter	-------
Chlorides	Argentometric method	Standard method
Nitrate	Phenoloic disulphonic Acid Method	UV spectrophotometer
COD	COD digester (Open reflux system)/Titrimetric	Hach 389, USA
Solids	Gravimetry	Hot air oven
Phosphate	Ammonium Vandate/Molybdate	UV Spectrophotometer
BOD	27o C, 3 days incubation/ Titrimetric/ Modified Winkler's method	
Conductivity	Conductivity meter	-----
Color	Platinum cobalt method	------
Sulphate	Spectrophotometric method	UV spectrophotometer
DC Power Supply Unit	0-10 A, 0-15 V, DC power supply unit	APLAB, Regulated dual DC power supply LD3210.

III. RESULT AND DISCUSSION

The several factors that regulate the process of electrolysis in the removal of biological and chemical contaminations in the wastewater technique include electrode material, the distance between the two electrodes, duration of the electrolysis process, applied voltage (current densities), pH and the presence of other coagulants in the technique. The initial study was done to figure out the removal efficiency of landfill leachate treatment. The detailed study was conducted for the changes in voltage and distance between the electrodes.

A. Effect of inter-electrode distance on landfill leachate treatment by EC:

The effect of inter-electrode distance has been considered as a process parameter to reduce current consumption for the landfill leachate treatment. The distance of the electrode varied at 1, 2, 3, and 4cm. The COD and Color removal have been observed to increase with decrease in inter-electrode distance from 4 to 1cm. Percentage COD removal and Color removal as shown in Fig 3 and Fig 4.

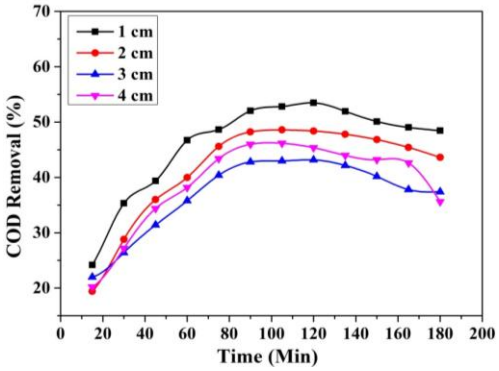

Figure 3 Percentage removal of COD with different distance

The maximum percentage removal achieved was COD and Color 53.0% and 35.3% respectively, at the shortest distance 1 cm between the electrodes with surface area 35 cm2. Because less than the 1cm spacing between electrodes prevent the flow of liquid adsorbate in the interstitial spaces of the electrodes thus, hindering percentage removal efficiency. Similar observations have been reported by [8]. At the anode, the faster anion discharge takes place, that results in increasing removal efficiency and enhance oxidation, it also minimizes the electrical current consumption, resistance, and price of the leachate wastewater treatment[9].

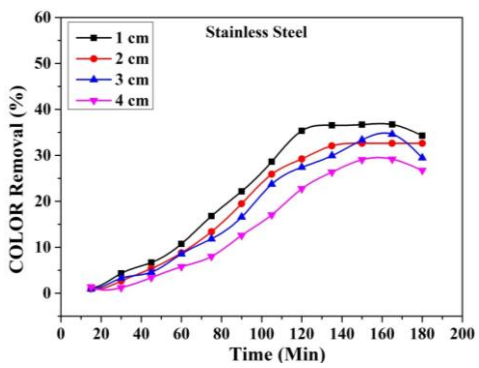

Figure 4 Percentage removal of Color with different distance

B. Effect of applied voltage on COD and color removal efficiency:

The applied voltage is a significant parameter that affects the treatment efficiency of the electrocoagulation degradation method. The batch study was carried out to find the effect of voltage on COD and color removal efficiency from leachate. EC experiments have been carried out at 4V, 6V, 8V, 10V, and 12V, as shown in Fig 5 and Fig 6. At 12V maximum COD and Color, removal had been found. It can be noticed that the percentage removal efficiency of COD and Color was relatively high whereas the voltage of 12V. If the voltage (current density) increases, charge loading also increases that leads to increase the pollutants removal [4].

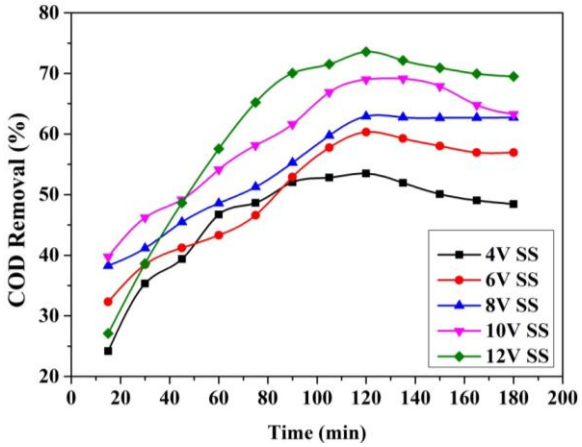

Figure 5 Percentage removal of COD with different voltage

At this voltage, over 73.5 % of COD and 65.0 % of Color was removed. Stainless steel electrodes can be successfully used as anode and cathodes, which make the process more efficient and easier to maintain. The sharp decrease of COD was because of the combined effects of destabilization of colloidal organic compounds and cathodic reduction. Certainly, after EC, a thin brownish deposit layer was noticed on the cathodic electrode surface, that results in the cathodic reduction phenomenon [10]. The voltage of 12V and 120 min was selected as the optimum conditions for the EC treatment.

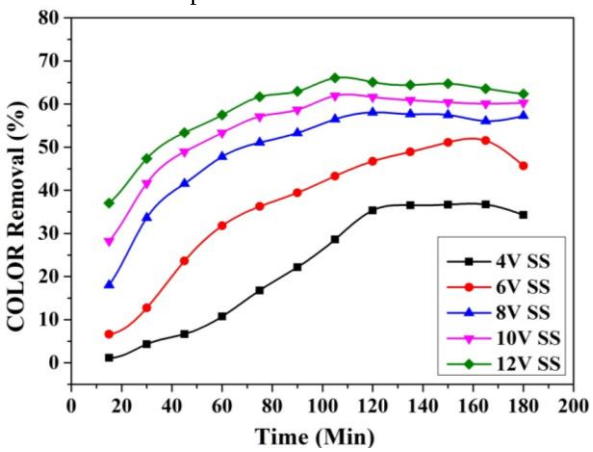

Figure 6 Percentage removal of Color with different voltage

C. Effect of pH changes during electrochemical treatment

In electrocoagulation, it has been proven that pH has a significant effect on COD and Color reduction. The initial pH of the landfill leachate was 8.64 after the end of 120 min electrocoagulation; it was noticed that the pH had been increased to 9.8 at 12 V, as shown in Fig 7. The results reveal that the highest COD and Color removal was 73.5 % and 65.0 % respectively at pH 9.8. Some of the researchers found that pH was the most important controlling operating parameter in electro-oxidation of landfill leachate correlate with chlorine concentration, applied voltage, temperature, and leachate input rate. Because of dominant activities at the cathode pH increases gradually [11]. In neutral and alkaline condition, decolorization of effluent is very high but very less in acidic condition. At the cathode, the production of hydroxide results in electrochemical reduction. pH variation did not significantly change COD removal in electro-oxidation of leachate. [12].

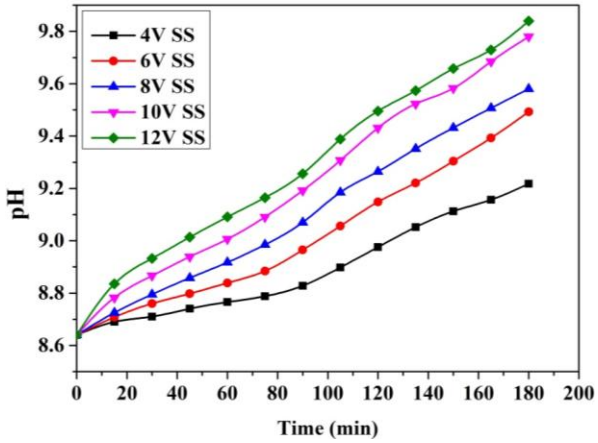

Figure 7 Effect of pH versus electrolysis time

D. Effect of BOD/COD ratio changes during the EC process

From Fig 8, it is observed that there was an improvement in biodegradability of landfill leachate evaluated through the evolution of the BOD/COD ratio. With a raise in BOD/COD ratio from 0.11 to 0.62 at optimum time 120 min, voltage 12, and distance 1 cm. It can be observed that when the voltage increases the degradation of COD also increases consecutively, the BOD/COD ratio of the effluent also increases.

This is due to increasing voltage, increase the overall potential essential for the production of chlorine and hypochlorite. Under different voltage, the other operating condition was altered, and the performance of the reactor also affected. An increasing BOD/COD ratio signifies a remarkable improvement of biodegradability [13]. The low BOD/COD ratio (0.11) in the effluent specifies that it contains recalcitrant substances which were not easily biodegradable or non biodegrabale material present in leachate [14].

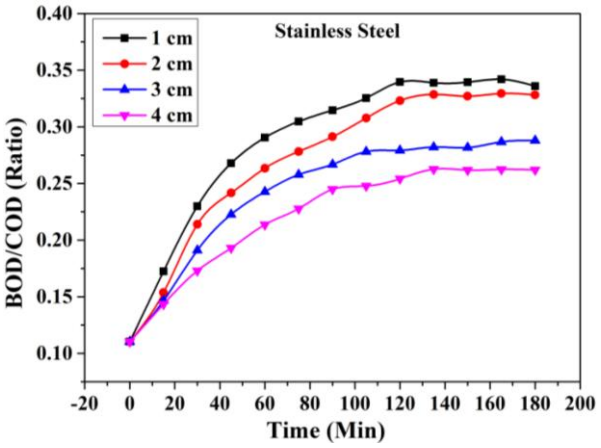

Figure 8 Effect of BOD/COD ratio vs Electrolysis duration

IV. CONCLUSIONS

Electrocoagulation technique is a promising process for the remediation and degradation of effluents of low bio-degradability. The research work shows that the performance of electrocoagulation is an efficient process for treating landfill leachate. Stainless steel electrodes were used for performing electrocoagulation. To optimize the process parameters such as inter-electrode distance, applied voltage, electrolysis time, and effect of initial pH. From that experimental work, some of the conclusions are drawn.

- The maximum removal achieved was COD and Color, 53.0 % and 35.3% respectively, at the shortest inter-electrode distance of 1cm.
- The result reveals that an increase in BOD/COD ratio from 0.11 to 0.62.
- The higher removal efficiency was obtained COD and Color, 73.5% and 65.0% respectively, at the optimum electrolysis time of 120 min. Optimum inter-electrode distance 1cm at optimum applied voltage 12V, pH 9.8, and stirring speed 250 rpm.
- The results reveal that EC is more efficient for removal of COD, Color, and BOD/COD ratio in leachate.

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A Review of Electrocoagulation Process for Wastewater Treatment

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Abstract : The control of environmental pollution and also the treatment of polluted water are of great concern. Within the past decade, electrochemical coagulation process has emerged as most effective wastewater treatment process as compared to conventional techniques of treating wastewater. Electrocoagulation is robust, cost effective, reliable, low sludge generating process, it has automation amenability and it has high pollutant removal efficiency. It has been proved effective in treating various types of wastewater but is seldom accepted. The aim of the review is to explain the basics and up to date advancement of electrocoagulation method for the improvements in the pollutant removal efficiency. In this review paper, an overview of electrocoagulation method with effect of key operational parameters on it is provided. Limitations of the method are also represented for the better understanding of the mechanism of pollutant removal and its optimization. The recent advancements and future scope of the electrocoagulation process are also reviewed.
Keywords : Electrocoagulation; wastewater; poly hydroxides; sacrificial electrode.

1. Background of Ec Treatment

Electrolysis is a method in which oxidation and reduction occur due to application of electric current to the electrolytic solution. Electrochemical technology has shown to be a hopeful technique for the destruction of organic pollutants in the wide collection of wastewater and there is no need for adding additional chemicals. In addition, the high property of the electrochemical process prevents the assembly of unwanted by-products. And it can also be used for the metal recovery from the different wastewater.

In the 19th century (1889) in London, the electrochemical method was proposed with a well-established plant for the sewage treatment. In this process, wastewater was electrolyzed by mixing with sea water. The prime interest of primary stage development of the EC process was to generate chlorine for the removal of odor and disinfection of sewage wastewater. Electrochemical processes include: electro-coagulation, electro flotation, electro oxidation, electro-flocculation, electro-disinfection, electro reduction, electro-deposition, etc. Electrocoagulation (EC) is the most established electrochemical process. EC process was developed and patented by A. E. Dietrich in 1906 for the treatment of blige water from ships. Later in 1909 in the US, wastewater treatment by the electrocoagulation using aluminum and iron electrodes was proprietary by J.T. Harries. In 1984 in the US for the first time, a large scale drinking water treatment by electrocoagulation method was implemented.

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Electrocoagulation (EC) finds its application in treating different types of process wastewater, for example electroplating wastewater[1], Heavy oil refinery [2], textile wastewater[3], [4], Dairy wastewater[5], Distillery wastewater[6], [7], Leachate wastewater[8], [9].In recent years EC process has successfully applied for the de-fluoridation of groundwater[10].

In the 20th century electrochemical process had narrow success and acceptance in spite of being a competitive and effective treatment technique for most of the wastewater. The initial improvements were in minimization of electrical power consumption and throughput rates of effluent. Therefore present study focuses on the mechanism of EC process and operational factors (voltage, current density, temperature, time of treatment, electrode arrangement, inter electrode distance and pH) effecting its efficiency for the improvement of the EC process.

1.1 Electrocoagulation

Electrocoagulation process involves oxidation and reduction reaction in which destabilization of contaminants (suspended, emulsified, or dissolved) happens because of application of electric current to the electrolytic solution. EC unit consists of an electrolytic cell and metal (Al or Fe) electrodes which are connected to an external power supply. The conductive metal plates are well known as 'sacrificial electrodes' which are made up of same or completely different materials as anode or cathode. In the EC process, anodic dissolution generates *in situ* coagulants along with hydroxyl ions and hydrogen gas at the cathode. These *in situ* coagulants cause the formation of flocs within the sort of metal (Al or Fe) hydroxides and/or poly hydroxides. The hydrogen gas generated at the cathode brings flocs at the water surface by providing further buoyancy. The benefits and drawbacks of EC process are given below.

1.1.1 Benefits of Electrocoagulation Process

1. EC involves artless equipment and is easy to work.
2. EC requires low investment, maintenance, energy, and treatment costs.
3. EC treated wastewater furnish pleasant, odorless, clear and colorless water.
4. EC is a low sludge producing process. EC generated Sludge is mainly composed of metallic oxides/hydroxides.
5. There are no additional chemicals required in EC process.
6. Flocs formed by EC are similar to chemical floc. EC flocs are much larger in size, enclose less bound water and are acid-resistant and more firm.
7. The reuse of EC produced effluent contributes to a lesser water recovery cost because it contains a lesser amount of total dissolved solids (TDS) as related with chemical treatments.
8. The gas bubbles generated at the time of electrolysis can proceeds the pollutants to the top of solution from where it can be separated without difficulty.
9. EC provides greater efficient pH range and pH neutralization result and can be suitably used with other renewable sources of energy.

1.1.2.Disadvantages of Electrocoagulation Process

1. The sacrificial anodes are dissolved into solution due to oxidation, and need to be replaced at regular interval.
2. Conductivity of the wastewater suspension must be high.
3. Viscous hydroxide may be likely to solubilize in some cases.
4. The electricity may be not easily available and expensive in some area.
5. The efficiency of the electro coagulation unit decreases due to an impervious oxide film shaped on the cathode.

1.2. Mechanism of Electrocoagulation

The EC reactor configuration varies with the number of electrodes and arrangement of electrodes (monopolar or bipolar).

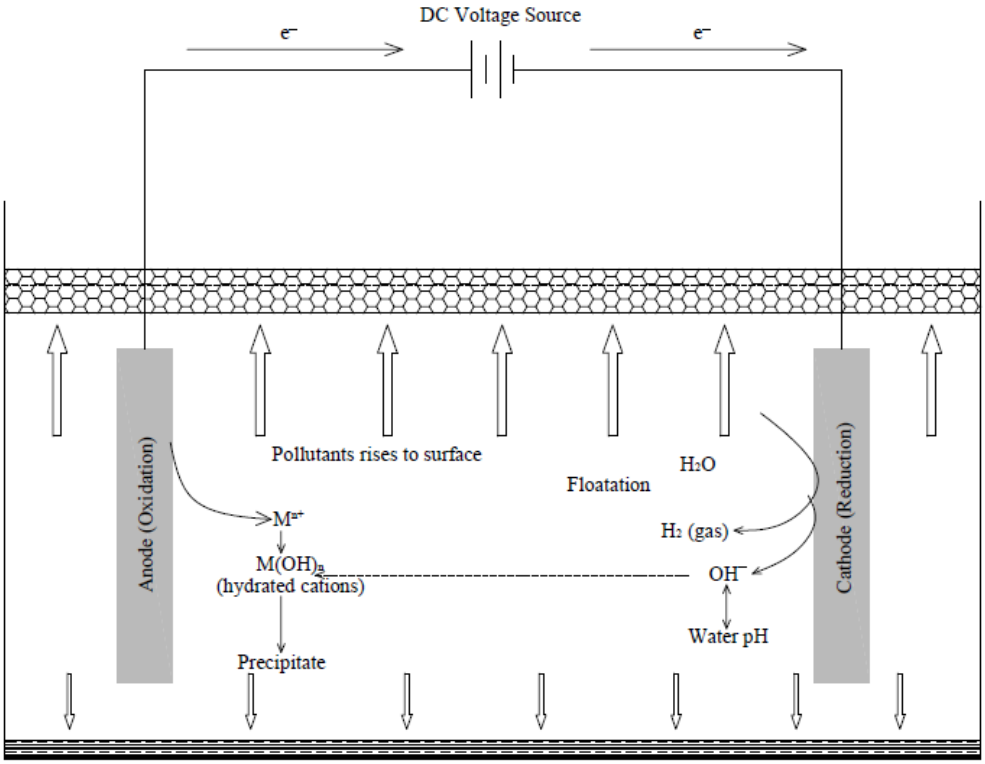

Figure 1. Diagram of a bench-scale EC reactor

The reaction mechanism of the electrochemical method using aluminum and iron electrodes is shown in Figure 1. On an applied electric current, oxidation of anodic material and reduction of cathodic material takes place.

Anodic reactions:

$Al(s) \longrightarrow Al^{3+} + 3e^-$
$Fe(s) \longrightarrow Fe^{2+} + 2e^-$
$2H_2O\ (l) \longrightarrow O_2\ (g) + 4H^+ + 4e^-$
Anode oxidation or atmospheric oxygen oxidizes ferrous ion to Fe^{3+}.
$Fe^{2+} \longrightarrow Fe^{3+} + e^-$
$2Fe^{2+} + 1/2\ O_2\ (g) + H_2O\ (l) \longrightarrow 2\ Fe^{3+}$
Cathodic reactions :
$2H_2O + 2e^- \longrightarrow H_2\ (g) + 2OH^-$
Moreover, in the presence of chloride and high anode potential, the subsequent reactions may occur in the EC cell:
$2Cl^- + Cl_2 + \longrightarrow 2e^-$
$Cl_2 + H_2O \longrightarrow HOCI + CI^- + H^+$
$HOCI \longrightarrow OCI^- + H^+$

Electrochemically produced metal cations (Fe^{3+} or Al^{3+}) will react spontaneously and produce corresponding hydroxide and /or poly hydroxides according to complex precipitation kinetics. Many monomeric species of Al^{3+}such as $Al(OH)^{2+}$, $Al(OH)^{2+}$, and $Al(OH)^{4-}$ and polymeric species such as $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}\ O_4(OH)_{24}^{4+}$, and $Al_{13}(OH)_{34}^{4+}$ transform into $Al(OH)_3$.Similarly Ferric ions species such as $FeOH^{2+}$, $Fe(OH)_2^+$, $Fe_2(OH)_2^{4+}$,$Fe(OH)_4^-$,$Fe(H_2O)_5\ OH^{2+}$, $Fe(H_2O)_4(OH)_2^+$,$Fe(H_2O)_8(OH)_2^{4+}$and $Fe(H_2O)_6(OH)_4^{2+}$ transform in to $Fe(OH)_3$. These insoluble $Fe(OH)_3$ and $Al(OH)_3$ remain in the aqueous medium and destabilize contaminants by completion or electrostatic attraction followed by coagulation. The formation of these complexes is highly pH dependent. Destabilization of contaminants mainly occurs through two distinct mechanisms, i.e.

(i) Cationic hydrolysis products neutralize negatively charged colloids.

(ii) Sweep flocculation: Entrapment and removal of contaminants in the form amorphous hydroxide precipitate.

Gas bubbles H_2 and O_2 adhere to agglomerates which are released at the electrode surfaces during electrolysis and carry them to the water surface [11].

2. Effects of Operational Parameters on EC Process

To achieve the maximum removal efficiency using electrocoagulation process in minimum electrolysis time with minimum operational cost, it is essential to understand the effect of various operational parameters on EC process.

2.1. Electrode Arrangement

In the EC process, electrode material and type of electrode connection play a major role in the cost analysis.

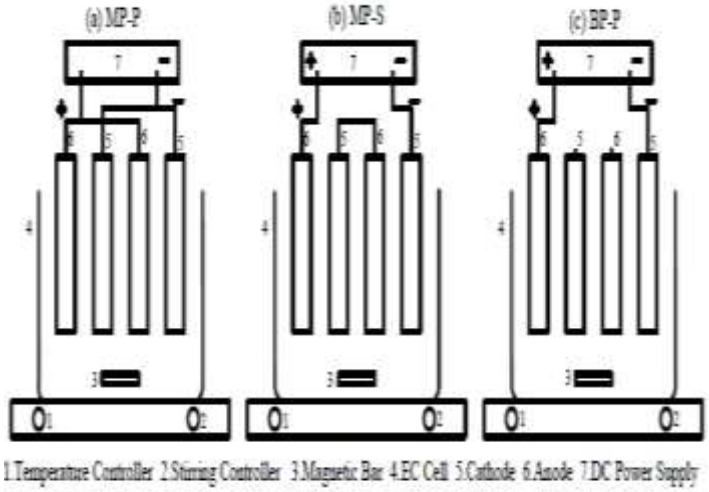

Figure 2. Diagrammatic representation of different mode of electrode connection (a) monopolar parallel (b) monopolar series and (c) bipolar parallel

The diagrammatic representation of different types of electrode connection is shown in Figure 2 (a), (b) and (c).

In the EC process, electrode materials define the type of electrochemical reaction that will occur in the EC processes. Al or Fe plate can be used as the anode and inert material such as steel, stainless steel, platinum coated titanium etc. can be used as cathode [12]. In few cases, similar material is used for the anode and cathode. An EC system can include either one or multiple anode-cathode pairs and may be connected in either a monopolar or a bipolar mode.

Monopolar mode of parallel connection (MP-P) is shown in Figure 2 (a). During this connection, current is split between all the electrodes in regards to the resistance of individual electrodes. A low potential difference is needed as compared with serial connections. In the monopolar mode of series connections (MP-S), each pair of sacrificial electrode is internally connected with one another as shown in Figure 2(b).

In the bipolar mode of parallel connection (BP-P) as shown in Figure 2(c), sacrificial electrodes are placed between the two parallel electrodes without any electrical connection. Bipolar electrode arrangement has simple set-up and hence the maintenance is low. Once an electric current is passed through the outer electrodes, the uncharged sites of conductive plates get charged with the opposite charge compared to the parallel side beside it. In the EC process, positive sides of electrodes undergo anodic reactions and negative side undergoes cathodic reactions throughout electrolysis [13].

2.2 Effect of Electrolysis Time

Electrolysis time also has significant effect on pollutant removal efficiency of electrochemical coagulation method. It defines the amount of coagulant formed and cost of the process. An increase in electrolysis time up to the optimum level increases the pollutant removal efficiency but it does not increase beyond optimum level. The actual fact is that at constant current density coagulant formation increases with an increase in electrolysis time which leads to increased removal efficiency. Whereas the above optimum electrolysis time and increase in coagulant dose does not increase the pollutant removal due to the presence of sufficient number of flocs[14]. Electrolysis time has a negative impact on cost of treatment due to increase in energy and electrode consumption at longer electrolysis time.

2.3 Inter Electrode Distance

In the EC process, inter-electrode distance plays an important role on EC potency because the electrostatic field depends on the distance between the anode and the cathode. An optimum distance between electrodes provides maximum pollutant removal efficiency. Minimum inter-electrode distance provides low pollutant removal efficiency. The more the inter electrode distance the slower the movement of the generated ions. Due to the slower movement ions gets extra time to form floc required for the coagulation of pollutants[14].Whereas an additional increase in inter electrode distance above optimal value decreases anodic dissolution and will increase the distance that ions essential to travel for floc formation, which results in the decrease in the electrocoagulation efficiency [15].

Electrocoagulation efficiency depends upon conductivity of the solution. As shown in the equation below the electrical conductivity is directly proportional to the inter electrode distance. An increase in distance between the anode and the cathode (g), increases resistance (R) offered by the cell [16].

$$R = \frac{g}{KA}$$

Where K is the cell specific conductance and A is electrode surface area. According to Ohm's law current increases with the decrease in resistance. An increase in current leads to an increase in anodic dissolution thus increasing the electrocoagulation efficiency.

2.4 Effect of Current Density

One of the most significant operational parameters in electrocoagulation process is current density i.e., current per area of the electrode. According to the literature a wide range of current densities applied between 1-100 mA/cm^{2}depending on the case study. The separation processes which involves flotation cells or large settling tanks requires high current density, whereas sand and coal filter integrated EC process needs low current density.

The amount of electrode dissolution is directly proportional to the amount of current passed through the electrolytic solution. Faraday's law explains the relationship between current density (mA/cm^2) and the amount of metal (m) dissolved (g of M/cm^2).

$$w = J \times t \times \frac{m}{n} \times F (4)$$

Where the quantity of electrode material dissolved (w) in g /cm^2; applied current density (J) in mA/cm^2;electrolysis time (t) in s; m the relative molar mass of the electrode material under study; n the number of electrons in oxidation/reduction reaction; and F is Faraday's constant (96,500 C/mol).

At high current density, there is wastage of electrical energy in heating up of water which results in decrease in current efficiency. To ensure a high current efficiency, the current density must be selected in accordance with the other operating parameters such as pH, temperature and flow rate. In the EC process, current density and the types of the anions define the current efficiency[17].

2.5 Effect of pH

The pH of the solution determines the conductivity of the solution, the electrode dissolution, and formation of hydroxides in the electrocoagulation process. Various empirical studies show that at high current efficiency using aluminum electrodes occurs at either acidic or alkaline condition than at neutral conditions. The nature of the pollutants determines the EC efficiency, however near pH 7 pollutant removal was found best. And, at neutral pH, the power consumption was high due to conductivity variations. pH effect is not significant at high conductivity. EC efficiency using aluminum electrodes decreases at pH 10 [18]. The optimal pH for wastewater treatment by electrochemical coagulation ranges from 6.5 to 7.5.

The pH of solution varies during electrocoagulation method and the final pH of the effluent significantly influences the pollutant removal efficiency.

2.6 Effect of Temperature

Temperature significantly influences the pollutant removal efficiency by using EC process. Temperature can have a positive or negative effect on electrochemical coagulation process, therefore in an EC process which is carried out at ambient temperature, it is necessary to know the factors that cause variations in temperature during the process. The increase in temperature decreases the pollutant removal efficiency due to the decrease in metal hydroxide formation [18]. At low temperature pollutant removal efficiency is also low due to the low anodic dissolution rate[19].

Temperature influences the EC process by altering the rate of reactions, solubility of metal hydroxides, liquid conductivity, and kinetics of gas bubbles, or small colloidal particles. Effect of temperature (20-50°C) on phosphate removal from wastewater using aluminum electrodes showed that increase in temperature increases the removal efficiency due to the increased mass transfer of aluminum ions from anode surface to bulk solution and increase in the rate of aluminum hydroxides formation. Increase in temperature above 50°C is not preferred as it is uneconomic and alters the rate of reaction and removal efficiency.

2.7 Effect of Initial Pollutant Concentration

The initial pollutant concentration is also considered as one of the effective parameters in pollutant removal by electrochemical coagulation. According to literature an increase in initial pollutant concentration (by keeping other parameters constant) reduces the pollutant removal efficiency of EC process. This is due to the circumstance that at fixed operating parameter the amount of coagulant generated will be fixed which is insufficient to form floc with high pollutant concentration. Therefore a longer electrolysis time is required to reduce the residual pollutant concentration up to the desired removal yield. Pollutant concentration not only decreases the EC efficiency it also increases the energy consumption by increasing the resistance and conductivity of solution.

3. Kinetics of EC Process

The kinetics of pollutant removal by electrochemical coagulation can be exhibited by the estimation of coagulant dose using an adsorption phenomenon. Kinetic studies are necessary to determine the dependency of pollutant removal rate on the operation parameters. The amount of pollutant (Qt) removed at time t is calculated by using the Equation $Qt = (C_0 - C_t)\ v\ w$

Where C_0 (mg/L) is the initial pollutant concentration and C_t (mg/L) is the pollutant concentration at time *t* in the aqueous phase, v is the solution volume (L), and w is the mass of metal hydroxide calculated. The mechanism of the adsorption of the pollutant on in situ-generated metal hydroxide process can be described by applying different kinetic models such as pseudo-first order, pseudo-second-order, Avrami and Elovich models.

3.1 Operating Cost Analysis

In the Electrocoagulation process, it is very necessary to achieve highest removal efficiency with minimum operating cost. The operating cost of electrochemical coagulation process depends on the cost of the electrode, electrical energy cost, sludge disposal, and fixed cost. The operating cost can be calculated by the following equation

Operating cost = a C_{Energy} +b $C_{Electrode}$

Where C_{Energy} and $C_{electrode}$ is the amount of electrical energy and electrode consumed per liter of wastewater treated, which is experimentally achieved. The unit price of electrical energy "a" and electrode material "b" can be obtained by the market.

The amount of electrical energy consumed per liter wastewater treated can be calculated by the given equation.

$E_{EC} = UIt_{EC}$

Where $E_{EC}$is the electrical energy in kWh/L, U the cell voltage in volt (V), *I* the current in ampere (A) and $t_{EC}$is electrolysis time.

The amount of electrode consumed (E_{EL}) per liter wastewater treated can be calculated by the given equation

E_{EL} = Initial Weight of Electrode - Final weight of Electrode

Table 1. Recent Application of EC in the Treatment of Wastewater

Sl. No	Author and year	Waste water type	Anode/ cathode	Reactor type	Electrode arrangement:	No of electrodes:	Sample volume in (ml)	Optimum electrode gap in (cm)	Optimum current density	Optimum detention time in (min)	Temperature	pH	Initial pollutant Level (mg/L)	Optimum removal efficiency in (%)
1	Tyagi et al. 2014[20]	Textile wastewater	Fe	Batch	-	2	1500	4	14-17 mA/cm^2	20	-	8.5	COD=600-650	COD=76 Colour=95
2	Farshi et al 2013.[7]	Distillery wastewater	Al SS	Batch	-		200	1	2	180	-	4	-	COD=70-72 Colour= 97-98
3	Akbal et al 2011.[21]	Metal plating wastewater	Al/Al Al/Fe Fe/Fe Fe/Al	Batch	Monopolar	6	650	1	10 mA/cm^2	20	-	9	Ni=394 Cu=45 Cr=44.5	Ni=100 Cu=100 Cr=100
4	Katal et al 2011.[22]	Paper mill wastewater	Al Fe	Batch	-	-	1500	1	70 mA/cm^2	30	20^0C-60^0C	5-7	COD=1700 BOD=850 TOC=910 TSS=1060 TS=9801 Phenol=34 Colour=NM	COD=86(Fe) Colour=92(Al) Phenol=96 (Al) BOD,TOC,TS,TSS=NM
5	Kobya et al. 2011.[23]	Potable water	Al Fe	Batch	-	-	650	1.3	2.5 A/m^2	4(Al) 2.5(Fe)	-	7(Al) 6.5(Fe)	As=150mg/l	As =93.5 (Al) As = 94.1(Fe)
6	Kobya et al. 2011.[24]	Drinking water	Al Fe	Batch	MP-P	4	560	1.3	2.5 A/m^2	12.5(Fe) 15 (Al)	-	6.5(Fe) 7 (Al)	As =75-500mg/l	As=93.5(Fe) As= 95.7(Al)
7	Linares Hernandi et al 2009.[12]	Industrial wastewater	Al,Fe (Al+Fe)	Batch	MP	2	4000	2	45.45 A/m^2	60	-	8	COD =1700-2500, BOD_5=900-930 Colour(pt-C_0)=2500-3700 Turbidity(NTU) =1400-1800	COD=>99 BOD_5=>99 Colour=100 Turbidity=100
8	Kobya.et al 2010.[25]	Electroplating rinse wastewater	Fe	Batch	MP-P	2	650	1	30-60 A/m^2	30-80	25^0C	8-10	Cd=102 Ni=1175 Cy=120-261 COD=180-220	Cd=99.4 Ni=99.1 Cy=100 COD=NM

9	Zodi et al 2010[3].	Industrial textile wastewater	Al	Batch	-	2	2200	2	60-140 A/m^2	30-90	20^0C	7	COD=1260 Turbidity=1310[NTU] TS=1750	COD=70 Turbidity=90 TS=50
10	Kushwaha et al 2010.[5]	Dairy wastewater	Fe	Batch (synthetic)	BP	-	1.5	1	270	50	-	6-8	COD=3900 Turbidity=1744 NTU TS=3090 TN=113 Chloride=31	COD=70 Turbidity=100 TS=48.2 TN=92.75
11	B M Krishna et al 2010.[6]	Distillery wastewater	Al	Batch	-	2	1500	2	0.03-0.01A/cm^2	120	-	3	COD=42240-46440 BOD_5=6757-8600 BOD_5/COD=0.15-0.19	COD=72.3 BOD_5/COD=0.15-0.68
12	Vasudevan et al 2010.[19]	Drinking water containing boron	Mg SS	Batch	-	2	900	0.5	0.2Ad/m^2	30	±2K	7	Boron=3-7	Boron =86.32
13	Maghanga et al 2009.[26]	Tea factory wastewater	Steel	Batch	-	2	400	0.5	24V	NM	Above 20^0C	6	COD=293-607 BOD_5=42-193 Colour=2004-9210[Pt/Co]	COD=96.6 BOD_5=84 Colour=100 Electric conductivity=31.5
14	Kalyani,balasubramanian et al 2009.[27]	Pulp and paper industrial effluent	Steel Al	Batch	-	-	-	1.5	NM	NM	-	NM	COD=32000 BOD=8225 BOD/COD=0.26	Colour=92-84 COD=95-89
15	InoussaZongo et al 2009. [4]	textile wastwater	MS Al Fe	Batch	-	2	$1cm^3$	NM	50-200A/m^2	60	20^0C	7.5	COD=1787 Turbidity=115 Conductivity=28	COD=74-88
16	Kobya et al 2008.[28]	Baker's Yeast wastewater	Al Fe	Batch	MP-P	4	800	2	70 A/m^2	50	20^0C	7	COD=2485 TOC=1061 Turbidity=2075 NTU TSS=503	COD=71(Al)-69(Fe) TOC=53(Al)-52(Fe) Turbidity=90(Al)-56(Fe)

17	Tir&Moulai-Mostefa et al 2008. [29]	Industrial oil-in-water emulsion	Al/SS	Batch	MP-P	3	400	1	25 mA/cm^2	22	20^0C-22^0C	7	COD=62300 Turbidity=29700 [NTU]	COD=90 Turbidity=99
18	Kongjao. et al.2008[30]	Tannary wastewater containing organic and inorganic pollutants	Fe	Batch	MP-P MP-S BP-P	6	3000	5	15.7-24.6 A/m^2	20	30^0C	7-9	COD=4100-6700 BOD=630-975 Cr=11.5-14.3 TSS=600-955 Oil &Grease638-780	COD=95 BOD=96 Cr=100 TSS=96 TDS=50 TKN=62 Oil &Grease=99
19	Ilhan,kurt et al 2008.[31]	Leachate wastewater	Al Fe	Batch	Parallel	2	0.5	6.5	348-631 A/m^2	30	-	NM	COD=12860 BOD_5=5270 BOD_5/COD=0.41 Ammonia=2240 Turbidity=1340 Chloride=3100	COD,NH_4-H=56 (Al) COD,NH4-H=35 (Fe)
20	Sevilveli et al 2008.[32]	municipale solid waste leachate	Fe-Al	Batch (laboratary scale)	-	-	2000	0.3	2-15mA/cm^2 (Al) 1-10 mA/cm^2 (Fe)	15	21^0C-22^0C	9	COD=4022 TOC=1295 Conductivity=25.11 Colour(Hz)=2650	COD=56-73(Al^{3+}), 87-90(Fe^{2+}) TOC=46-49(Al^{3+}), 58-68(Fe^{2+}) Colour=69-76(Al^{3+}),86-99(Fe^{2+})
21	Roa-Morales et al 2007.[33]	Pasta and cookie proccessing wastewater	Al	Batch(pilot scale)	-	8	1.5	-	18.2 mA/m^2	60	-	4	COD=7500 BOD_5=3445 Colour=35Pt-Co Turbidity=1153	COD=90 BOD_5=96 Colour=57 Turbidity=97 TS=95
22	Alaadin A et al 2008. [34]	Muncipal wastewater	stainless steel	Batch	-	2	1200	3	0.8A	5	-	7	TSS=126-160 Turbidity=49-53 Total BOD=84-112 Soluble BOD=26-32 Particulate BOD=51-84 Chloride=1238 Conductivity=4000	TSS=95.4 BOD=99

23	Kobya et.al 2006.[35]	Potato chips manufacturing wastewater	Al Fe	Batch	MP	4	250	1.1	20-300 A/m^2	5-40	22^0C	4-6	COD=2200-2800 Turbidity:260-610 NTU BOD=1650-2150	COD=60 Turbidity=98 BOD=NM
24	NN Rao et al 2001.[36]	tannery wastewater	Ti/mno2 - titanium	Batch	-	2	$60cm^3$	4	0.1-0.6A	20	-	8	Colour=1100-1150 Pt-C_o COD=480-550 Chloride=4500-4600 Ammonia=100-110	Colou=75-95 COD=50-75 Ammonia=96

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