

Treatment of municipal solid waste landfill leachate by aeration assisted electrochemical peroxidation process using aluminium and iron electrodes

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ABSTRACT

The present work aims to assess the degradation and mineralization of organic and inorganic components of solid waste landfill leachate. The main consequential problem of generation of leachate from municipal solid waste landfills is in form of strong colour, their highly dissolved contaminant nature and multifaceted composition of pollutants which must be treated for the safe disposal into the natural streams and other resources. So as to achieve this intention, this lab research study was carried out to observe the fulfillment and effectiveness of batch aerated electrochemical peroxidation process by applying the voltage between iron and aluminium electrodes in electrochemical cell. In addition, the various combinations and arrangement of iron and aluminium electrodes have been chosen and implemented in order to achieve chemical oxygen demand (COD) degradation while simultaneously improving colour removal in solid waste landfill leachate. The effects of pH (2–6), H₂O₂ dosage(0–6 ml/L), Fe²⁺ dosage(0–2.5 mg/L), voltage(0–6 V), various groups of iron and aluminium electrodes pairs on COD and colour removal capability were investigated in order to determine the best experimental settings and design. The findings of the treatment results indicated that the aerated electrochemical peroxidation processes (ECP) was very resourceful in achieving 91% COD and 95% colour from solid waste landfill leachate under optimal circumstances of treatment period of 120 min, pH = 3, voltage = 6 V, Fe²⁺ = 1.5 g/L, H₂O₂ = 3 ml/L and Fe-Al electrode pair respectively. Additionally, the acidic pH state of solid waste landfill leachate and the provision of external supply of aeration were vital throughout the treatment to enrich the efficiency of the aerated ECP process as well as to enhance the action of electrocatalytic generation of reactive hydroxyl radicals (*OH) with a low energy consumption value of 3.6 kWh/m³. Besides, the outputs of the aerated electrochemical peroxidation process demonstrated as a well-organized and specific practice for diminishing the concentration of COD and colour by implementing the combined action of electro-Fenton and electro-coagulation unit process for conversion of non-bio-degradable solids into H₂O and CO₂ with some simple inorganic end substances. Furthermore, the application of RSM software has been adopted to do the optimization of aerated ECP treatment process, for minimization of experimental runs and trials by calculating the number of experiments with an empirical model approach for the outputs.

Keywords: Aerated electrochemical-peroxidation; Iron and aluminium electrodes; Optimization; RSM; Solid waste landfill leachate

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1. Introduction

A landfill is an engineered system for land disposal of municipal solid waste. Among all the methods of disposal of solid waste, landfilling is the one method of disposal of municipal solid waste that has received the most nominations in developing countries as both land disposal and treatment of municipal solid waste. Municipal solid waste landfill leachate is defined as a highly polluted wastewater discharge that is very hard to treat with simple treatment practices. This is due to the large unpredictability of its hardly dissolved organic, macro inorganic, heavy metal components, powerfully reliant on the stage of development, class of disposed solid wastes and materials of a municipal landfill disposal yard [1]. Although the different combinations of biological treatment systems (aerobic and anaerobic) are generally implemented for quality improvement of municipal landfill leachate, owing to its intricate composition and solidity, conventional biological treatment systems are regarded as time-consuming and typically infrequent for decomposing macromolecular recalcitrant compounds. Hence, to achieve the safe disposal of leachate in the environment as per the effluent regulatory disposal standard requirement, further physico-chemical alteration in the characteristics of leachate is needed for the qualitative upgradation and its discharge into natural sources [2]. The formation of municipal solid waste landfill leachate causes natural quality deprivation in the environmental components. In the past decades, intensive research has been carried out on treating municipal solid waste landfill leachate by chemical and biological methods such as coagulation and adsorption [3], air stripping [4], reverse osmosis [5], membrane bioreactor [6], phytoremediation [7], granular activated carbon adsorption [8], electrochemical oxidation [9], SBR [10], Fenton-ozone based oxidation [11] respectively.

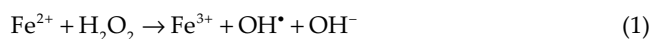
Nowadays, the improvement in the biodegradability index of municipal solid waste landfill leachate has been achieved by making significant treatment modifications in advanced electrochemical oxidation systems for oxidation of recalcitrant organic pollutants in raw landfill leachate into simple biodegradable substrates by the hydroxyl radicals. In a literature study, E-Fenton treatment was adopted as a former biological process to improve the biodegradability of leachate, which might be advantageous for the consequent biological process with 82% COD removal at pH 2, catalyst dosage of 50 mg/L, voltage of 5 V, electrode spacing of 3 cm, and electrode area of 25 cm² [12].

In the AOPs, the establishment of hydroxyl radicals could be stimulated by adding some suitable metal catalysts such as reactive chemical salts and the application of natural resources such as solar radiation and UV-visible light radiation [13]. The fundamental mechanism involved in the working principle of an electrochemical cell during electrolytic treatment or voltaic cell is to create an electric current field among connected cathodes and anodes, which will simultaneously aid the movement of positive and negative ions towards electrodes by the process oxidation and reduction [14]. The treatment analysis and practice of using electrochemical and photo electrochemical methods are focused in a larger way to yield a useful amount of hydroxyl radicals as the main oxidizing representative

in EAOPs [15]. Thirugnanasambandham and Sivakumar [16] investigated grey wastewater treatment using electro-Fenton with COD removal of 90% and TSS removal of 85% under optimized conditions of current density of 10 mA/cm², H₂O₂/Fe²⁺ molar ratio of 0.70, treatment time of 14 min, and pH 4. Furthermore, it could be used to treat persistent organic substances from various sources by utilizing the natural environment and simple equipment [17].

In the group of electrochemical advanced oxidation processes, electrochemical peroxidation processes (ECP) were explored as a cost-effective substitute to conventional wastewater treatment systems, and limited research studies were only investigated in the past decades. The working principle of this process is based on the destabilisation of soluble organic particles such as micro-colloidal, suspended, dissolved, and floatable substances in waste water in such a way of generating a greater number of hydroxyl ions through the onsite electro-desiccation of the sacrificial metal anode with the simultaneous reduction reaction at the cathode. The combined effect of Fenton's and electrocoagulation (EC) processes in the electrochemical peroxidation (ECP) process will enrich the quality of the effluent, after complete treatment. Pekey et al. [18] carried out electrochemical peroxidation to take away colour and total organic carbon (TOC) from co-complex (acid yellow 194) dye by means of iron electrodes in a batch-mode operation with colour and TOC removals were determined to be 86% and 33.5%, respectively, at optimum conditions. In addition to this, the ECP process involves the use of self-deleterious iron electrodes and stereochemistry which is a well-adjusted assistance of hydrogen peroxide to competently abolish solid toxic substances present in the municipal solid waste landfill leachate. Likewise, it galvanises the establishment of metallic coagulants from the self-disintegration of iron and aluminium electrodes by releasing electrons through an oxidative chemical reduction reaction on the electrode surface, and it will lead to the weakening of colloidal impurities and aggregation of insoluble flocs [19]. Moreover, the ECP process is initiated by injecting H₂O₂ into an outwardly similar process, and Fe²⁺ particles are generated by anodic dissociation of a sacrificial metal iron anode in an undivided electrochemical reactor. The produced hydroxyl radicals degrade the organic substrates present in the waste until the final complete mineralization is achieved [20]. The salient profit of the ECP process is that the ferric ions can be second-handed into ferrous ions. Subsequently, it executes like a catalytic agent which kindles the decomposition of hydrogen peroxide [21].

The aerated electrochemical peroxidation process in the batch reactor was started by adding a pre-planned amount of Fe²⁺ and H₂O₂ in order to trigger the unit processes such as direct Fenton according to Eqs. (1) and (2) and coagulation. The following chemical oxidation and reduction reactions on iron and aluminium electrodes were identified during the electrolytic peroxidation treatment of solid waste landfill leachate, where the self-diminishable iron and aluminium electrodes produce Fe²⁺ and Al³⁺ positive particles in the electrolytic batch reactor. Furthermore, there were two different electrochemical mechanisms found under the acidic and alkaline states of solid waste landfill leachate as per the following equations from Eqs. (3)–(12).

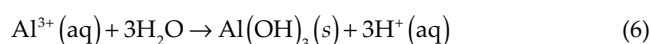
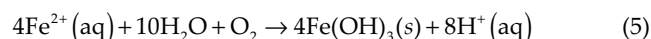


Mechanism 1 (acidic medium)

Anode:



In solution:



Cathode:

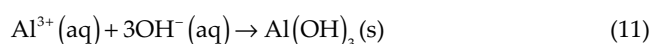
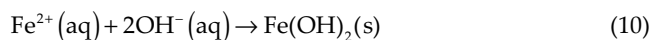


Mechanism 2 (alkaline medium)

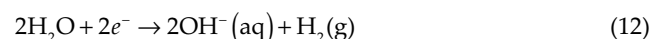
Anode:



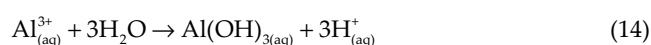
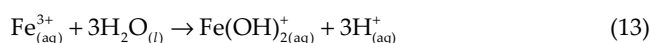
In solution:



Cathode:

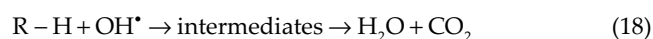


According to the developed electrochemical oxidation and reduction reactions at the anodes and cathodes, the diminishing of soluble colloidal solids and dissolved particles occurred in the leachate through the flotation mechanism by evolution of H_2 gases. Similarly, the generated Fe^{3+} and Al^{3+} particles will react with hydroxyl ions at various pH levels of the leachate, yielding stable final end products such as $\text{Fe}(\text{OH})_3$, $\text{Fe}(\text{OH})_2$ and $\text{Fe}(\text{OH})_3$, $\text{Al}(\text{OH})_2$, $\text{Al}(\text{OH})_2$, $\text{Al}(\text{OH})_3$. The chemical products developed during electro precipitation process involved were given in the following chemical reactions from Eqs. (13)–(16).



The persistent recalcitrant organic pollutants from the solution were removed through the formation of suspended iron and aluminium hydroxides through electro catalytic-absorption, electro-precipitation or electrolytic degradation and magnetism, trailed by means of electro-coagulation and sedimentation [22].

However, some of hydroxyl radicals might respond with Fe^{2+} and H_2O_2 , resulting in HO_2^\bullet ions, which are not viable in the oxidation process. This necessitates quantifying H_2O_2 and Fe^{2+} as per Eq. (17). Moreover, in Eq. (18), the following stable inorganic products of H_2O and CO_2 were observed at the end of electrochemical destruction of organic solids in municipal landfill leachate by oxidative reactive radicals species.



Even though advanced electrochemical systems have these successful benefits, the consequential influence on this process is energy consumption and its additional requirements, which will cause huge expenditure and maintenance prices. Hence, to invent a cost-beneficial treatment process, the single variable approach of the batch experimental optimization procedure has been executed, and keen observations have been made in varying the concentration of significant treatment factors, for example, pH, dosage of H_2O_2 , Fe^{2+} , applied voltage, and reaction time, respectively, for accomplishing the qualitative improvement in effluent characteristics.

Still, in advanced electrochemical systems, there is a research shortage and investigation into improving the quality of municipal solid waste landfill leachate by the collective action of iron and aluminium electrodes in electrolytic chemical cell. It will be considered as the utmost significant predominant feature of the aerated electro-chemical peroxidation process. With this in mind, we inspected the usefulness of this technology for enhancing the biodegradability of landfill leachate at a laboratory scale. The purpose of this batch experimental study is to modify the characteristics of municipal solid waste landfill leachate as per standard regulatory requirements and guidance by selecting an aerated electro-chemical peroxidation process with iron and aluminium electrodes as anode and cathode, and to qualitatively measure the drop in concentration of COD and colour, varying the experimental batch combinations, and the condition of different features like pH, H_2O_2 , Fe^{2+} dosage, voltage and the treatment time to reach the fulfilment of treatment effectiveness.

2. Materials and methods

2.1. Materials

All the reagents and chemicals such as sodium hydroxide, sulphuric acid, hydrogen peroxide (30%, w/w), ferrous sulphate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), H_2SO_4 , NaOH were received from Merck and analytical grade. The iron and aluminium sheets were purchased from the commercial metal shop and cut into required shape and size. The DC

power supply instrument with 30 V and 5A capacity was used for developing electrical voltage between the electrodes in the aerated electrochemical cell. To speed up the oxidation process, the external supply of sufficient oxygen through diffused aquarium aerator was used.

2.2. Collection and characterization of landfill leachate

Leachate samples were collected from Madurai municipal solid waste corporation compost and landfill yard, Avaniyapuram Municipality, Madurai, Tamilnadu, India. In Madurai city, garbage is produced at the rate of 406 g/d/head, gathering to a huge amount of about 548 ton/d. This is to some extent higher when compared to cities with similar levels of waste generation, where the per capita generation of waste is around 400 g/d, and the cause of the excess rate of formation of waste is due to the heavy influx of floating population, which is valued at around 3 lakhs. Out of the accumulated garbage released by the houses, shops, daily and weekly markets, commercial establishments, hotels, hospitals, and industries, the garbage generated by households accounts for nearly 64% of it. The current system of municipal solid waste collection, transportation, disposal is executed by the Corporation of Madurai under the influence of the Health and Engineering Departments.

For the efficient administration and for day-to-day operational purposes, the town is divided into 4 zones covering all the 72 municipal wards. Solid waste collected from various locations in the city is disposed off by open dumping at the Vellakkal site, located south of Madurai in the Avaniyapuram Municipality over an extent of approximately 110 acres, including a sewage farm that receives sewage and partially treated effluent from the predominantly lacking sewage treatment plant. The geographical coordinates of this location are 9°52'57.9"N latitude and 78°07'09.9"E longitude. The existing dumping ground has been reportedly used by Madurai Corporation for over 15 y. The lining system protects the surrounding environment, including soil, groundwater, and surface water, by containing leachate generated within the landfill, controlling the ingress of ground water and assisting in the control of the migration of landfill gas. It can be observed that approximately 68.42% of waste comprises organic waste, while the rest is non-degradable (silt, paper, plastic, glass, etc.). Silt accounts for approximately 30% of total waste. As per the CPHEEO manual, the quantity of leachate generated is 23 m³/d. The leachate from Vellakkal landfill site is stored in a collection tank and the details are given in the following, leachate collection quantity of collection tank is 161 m³, and assuming that the leachate was collected for 7 d, the depth of the collection tank is 1.5 m. So the plan area of the leachate tank is 107 m².

Samples were taken in plastic cans and kept in a refrigerator at 4°C. The physical and chemical characterization analysis and batch treatment systems were carried out at room temperature in Environmental Engineering Laboratory. Standard methods were followed to characteristics the leachate. The characterized leachate was considered as the stabilized leachate based on the examination of its characteristics and while compared with previous studies. The pH of leachate was alkaline and the BOD₅ to COD ratio was

0.03 which indicates that the physico-chemical treatment is appropriate for treatment of leachate. The physical and chemical characteristics of leachate were pH = 9.5, COD = 5120 mg/L, BOD = 135 mg/L, colour = dark brown colour, turbidity = 385 NTU, total solids = 19,000 mg/L.

2.3. Aerated ECP reactor and procedure

The aerated electrochemical peroxidation reactor consists of a cylindrical beaker of borosil glass with a working volume of 2 L. The anodes and cathodes were coupled to the DC power supply instrument. The DC power supply (0–30 V, 0–5 A) has been preferred to apply the voltage between the anode and cathode. The electrodes were made of iron (Anode) and aluminium (cathode) sheets with dimensions of 2.5 cm × 6 cm × 1 mm and positioned vertically which is kept at a distance of 3 cm apart. The electrodes were cleansed with diluted sulphuric acid and washed with distilled water before each run. A magnetic stirrer was attached for proper mixing of the solution in the reactor. Aeration was provided near the cathode using commercial aquarium aerator for the in-situ generation of hydrogen peroxide. The experimental setup is shown in Fig. 1.

In order to initiate the optimization process, a 1 L volume of leachate sample was filled into the batch electrolytic reactor. The aerated electrochemical peroxidation treatment process involves the initial pH measurement of the leachate sample and adjustment of the raw leachate to a prefixed value 2–6 by adding sulphuric acid reagent (H₂SO₄) or sodium hydroxide (NaOH). Then, to begin the batch electrochemical peroxidation, the essential amounts of ferrous sulphate heptahydrate (0–2 g/L) and H₂O₂ (0–6 ml/L) were injected into the reactor and thoroughly mixed together using an electrical magnetic stirrer to create uniformity and even distribution of the added chemicals in the reactor. The voltage differences between the electrodes

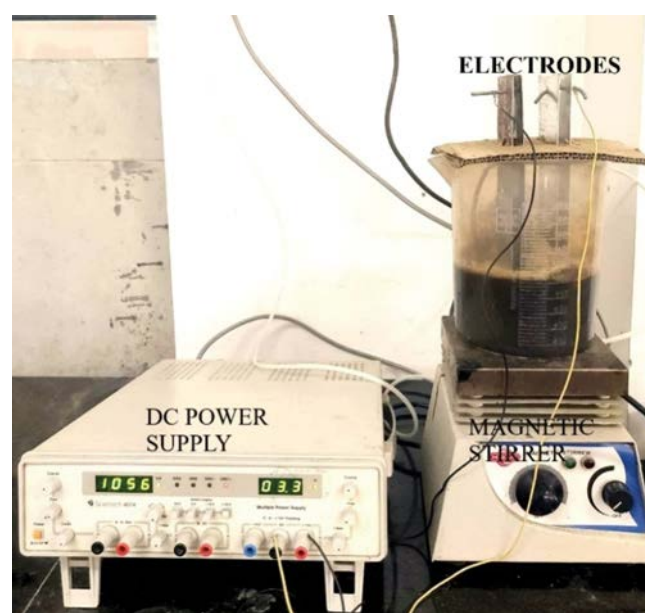


Fig. 1. Schematic diagram of the aerated ECP batch reactor setup.

were fixed within the range of 0 to 10 V. Moreover, aeration was provided to enhance the electrochemical oxidation and reduction processes. The time intervals for treatment were assigned between 0 to 160 min. At every 20 min break, samples were collected for characterization of the treated leachate and instantaneously the pH of the sample was adjusted by adding NaOH and sodium sulphite, thereby avoiding the formation of hydroxyl radicals. After treatment, the leachate was allowed to settle for about 30 min, leading to electro-coagulation and flocculation of end particles. Finally, the supernatant was analysed for the final quality measurements. The COD and colour reduction were measured using the COD closed reflux method and a UV Spectrophotometer.

2.4. RSM model development using face centred central composite design

In order to study the rectilinear, collaborative and quadratic polynomial impacts on the treatment process of aerated electrochemical per oxidation of stabilized landfill leachate, the RSM model with Face centred central composite design has been assisted in order to design, and run the optimization process variable quantity on COD and colour removal efficiency by using Design Expert 7.0.0., Stat-ease software. The ultimate aim of this analysis was to diminish the experimental trials and runs. The regression model work has been developed on parameters such as pH, H₂O₂ Dosage, Fe²⁺ concentration, and applied voltage with data analysis using RSM. By studying the outcome of influence of every parameter on COD and colour reduction, the optimal state of the aerated ECP method could be evaluated. Principally, RSM optimization system comprises three key phases: (1) running the statistically derived experimental trials; (2) assessing the factors and coefficients; and (3) foreseeing and analysing the results and examining the competence of the model.

The response was expressed as removal (%) which could be calculated by using the following equation.

$$\text{Removal (\%)} = \frac{C_i - C_0}{C_i} \times 100 \quad (19)$$

where C_i and C_0 are initial and final COD concentrations.

3. Results and discussion

3.1. Degradation of landfill leachate COD by empirical design and multivariate analysis

The RSM model was developed by using face centred central composite to do the planning, design of the experimental batch work on the treatment of leachate in electrochemical cells, and to study the impacts of treatment process parameters on COD removal efficiency. According to the outcomes of the FCCC based RSM model, the obtained results were acceptable in the form of a second-order polynomial equation and the pragmatic relationship (coded factors) among the reliant and self-reliant factors in the removal of COD from solid waste landfill leachate recognized by Design Expert 7.0.0., was obtained as follows:

$$\begin{aligned} \text{COD removal (\%)} = & +76.911 + 3.76A - 6.44B + 6.29C + \\ & 7.50D + 3.38E + 2.03AB - 2.22AD + 2.22B - \\ & 2.28DE - 6.99A^2 - 9.49B^2 - 27.99C^2 \end{aligned} \quad (20)$$

Generally, RSM was adopted for evaluating the relationship among the parameters to be varied during treatment (Time, pH, H₂O₂, Fe²⁺ and Voltage) and the process response was COD removal for aerated electrochemical peroxidation process. Significant model features were chosen in order to attain the most suitable fit in this model analysis. Moreover, CCD permitted the evolution of mathematical calculations and equations where foreseen consequences (Y) were assessed with respect to the functions of time (A), pH (B), H₂O₂ (C), Fe²⁺ (D) and voltage (E). In addition, the outcomes were calculated and analysed as per the amount of a constant, four first order impacts (terms in B , C , D and E), four interface effects (AB , AD , BC , DE), and three second-order effects (A^2 , B^2 and C^2). In total, 50 tests were plotted using $2k$, 26 replicates at the Centre point, 16 factorial points, and 8 axial (star) points, where " k " is the variable number. In order to know the precision of fit and its model validity, the outcomes were examined by ANOVA. All models were important in 5% confidence level, for the reason that probability findings were not more than 0.05.

The potency of this model has been proved by using four features viz, the Prob. > F of the model feature, the lack of fit test, adequate precision and the regression coefficient (R^2). Moreover, the Prob. > F pointed to know the model significance or its insignificance, lack of fit indicates the fitness tendency of the model, adequate precision indicates the proportion of signal to noise whereas R^2 designates the changeability among the real and foreseen data. Furthermore, for the model quality to be effective, the model indicators Prob. > F should be significant (no more than 0.0500), the lack of fit should be significant (more than 0.0500), adequate precision should be at least 4 and the R^2 should be no less than 0.8. According to Table 1, the model analysis for COD fulfilled these norms and conditions; hence it was chosen to visualize the process accomplishment. Correspondingly, the BOX-COX plot, normal probability, and the residual plots which interconnects the experimental and predicted values are shown from Figs. 2–6.

The attained correlation coefficient ($R^2 = 0.9477$) of the RSM model verified the acceptable relation among the foreseen and the gained experimental outcomes. Likewise, model relationships were estimated via the P -value (probability) by means of 95% confidence level. Consequently, three-dimensional analysis of graphical representation and their corresponding contour plot analysis were established according to the impacts of the four aspects (pH, H₂O₂, Fe²⁺ and voltage) on three different stages.

The statistical analysis showed that all the variables had significant effects on the models. The quadratic model statistical results for COD removals are summarized in Table 2. They show a high reliability in the estimation of COD removal efficiency ($R^2 = 94.77$). A high R^2 coefficient ensures a satisfactory adjustment of the quadratic model to the experimental data. In optimizing a response surface, an adequate fit of the model should be achieved to

Table 1
Experimental design factors for aerated ECP process

Factors	Units	Low actual	High actual	Mean	Std. Dev.
A = Time	min	0	160	70	41.231
B = pH	–	2	6	4	1.649
C = H ₂ O ₂ dosage	mL	0.00	6	3	2.474
D = Fe ²⁺	g/L	0.00	2.5 g/L	1.5	0.825
E = Voltage	V	0.00	10	5	4.123
Response	Units	Model	–	Mean	Ratio
Y ₁ = COD	mg/L	Quadratic	–	46.672	8.200

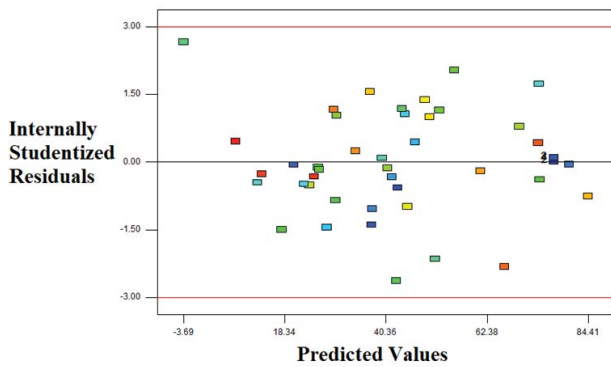


Fig. 2. BOX–COX plot.

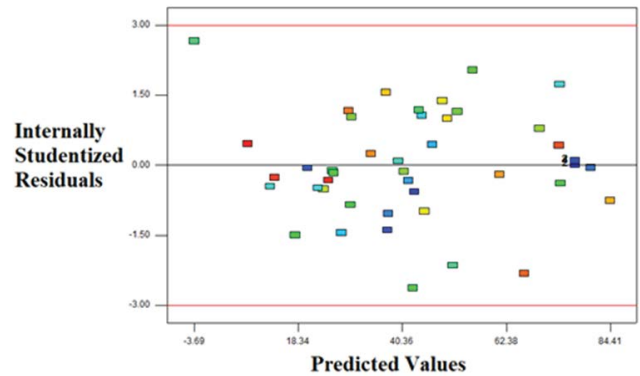


Fig. 4. The studentized residuals and predicted values.

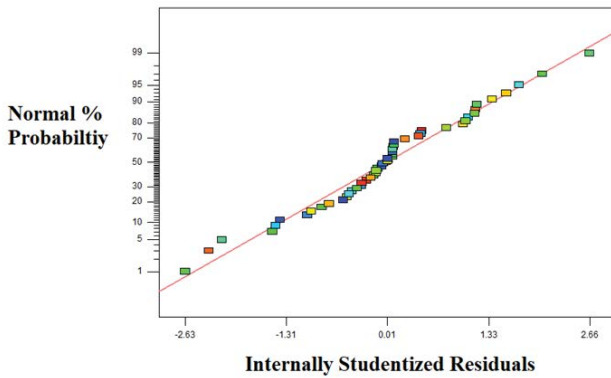


Fig. 3. The studentized residuals and normal % probability.

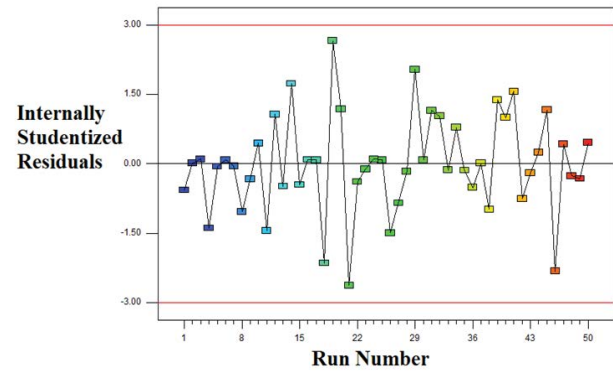


Fig. 5. The studentized residuals and run number.

keep away from a poor outcome. The value of the adjusted determination coefficient (adjusted $R^2 = 93.07$) also proved to be the high significance of both the models. In addition, the low value of the coefficient of variation (C.V. = 13.23) suggested the high precision and reliability of the experiments. The 3-D plots of the aerated ECP reaction with variables giving significant linear and interaction effects on the process efficiency.

3.2. Effects of pH

The most active vital parameter of electrochemical advanced oxidation system is the pH state of the leachate. The variation in pH governs the yield of hydroxy radicals

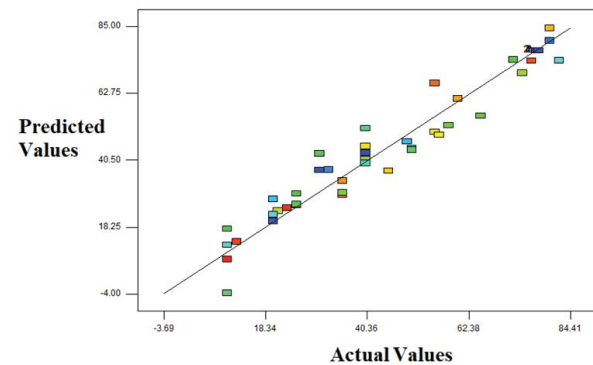


Fig. 6. Predicted vs. actual value plot for overall COD removal.

Table 2
Evaluated regression coefficients conforming to ANOVA outcomes

Analysis of variance (ANOVA) for the degradation of landfill leachate COD by ECP Process					
Source	Sum of squares	df	Mean square	F-value	p-value Prob. > F
Model	25,557.90	12	2,129.82	55.85	<0.0001
A – Time	481.88	1	481.88	12.64	0.0011
B – pH	1410	1	1,410.62	36.99	<0.0001
C – H ₂ O ₂ dosage	1,346.94	1	1,346.94	35.32	<0.0001
D – Fe ²⁺ dosage	1,912.50	1	1,912.50	50.16	0.0029
Voltage	388.97	1	388.97	10.20	0.0707
AB	132.03	1	132.03	3.46	0.0493
AD	157.53	1	157.53	4.13	0.0493
BC	157.53	1	166.53	4.13	0.0617
DE	166.53	1	141.60	4.37	0.0128
A ²	141.60	1	261.02	3.71	<0.0001
B ²	261.02	1	2,270.88	6.85	
C ²	2,270.88	1	38.13	59.55	
Residual	1,410.86	37	47.02		
Lack of fit	1,410.57	30	0.042	1,115.70	<0.0001
Pure error	0.30	7			
Cor. total	26,968.76	49			

R²: 0.9477; Adj. R²: 0.9307; Adeq. precision: 27.980; C.V.%: 13.23

and controls the process of liberation of ferrous ions in the electrochemical reaction cell. Hence, in the aerated electro-chemical peroxidation process, pH extremely affects the performance of the overall treatment effectiveness. To examine the outcome of pH on aerated electro-chemical peroxidation process, the removal of COD and colour was calculated at diverse pH values from 2 to 6 for the time interval of 0–160 min. The attained result signifies that the removal percentage of COD was amplified while raising pH up to 3. In this aerated ECP process an extreme of 91% COD and 95% colour reduction was attained at pH 3 for a time intermission of 120 min under the condition of 6 V between the iron anode and aluminium cathode. Moreover, the increment in the alkaline state of leachate pH from an acidic state led to a reduction in COD and the colour removal ability of the treatment.

Treatments such as Fenton and the electrochemical Fenton's reactions process were well recognised as the best in the treatment of organic pollutants at the low pH state of waste water in the advanced electrochemical oxidation process. Ozyonar et al. [23] investigated pH = 3 as optimum with a reduction of 92% of COD from coke waste by process electrochemical peroxidation. Ahmadzadeh et al. [24] identified that pH 3 was optimum for the 100% removal of bisphenol during electrochemical peroxidation in ground water treatment. According to the obtained outputs and findings in the literature, it was observed that the Fe³⁺ ion particles were converted into compounds like Fe(OH)₃ while raising the pH of the leachate above 5, which led to the lessening of hydroxyl radical formation along with the self-breakdown of hydrogen peroxide into oxygen and water [25]. Furthermore, beyond pH 6, the observed COD degradation was approximately 64%, owing to iron particles being

transformed into ferric hydroxides at higher pH values, whereas at pH 2, iron particles generate firm complexes such as [Fe(H₂O)₆]²⁺ with H₂O₂ and cause deactivation of metal catalysts, reducing the colour and COD removal efficiency.

According to the obtained results in Fig. 7, COD removal was attained at 80% at pH 2 and 91% at pH 3. Therefore, the increment in the alkaline state of leachate will bring a substantial reduction in COD degradation beyond pH 4 and with only 64% efficiency attained at pH 6. As a result, the acidic range of leachate provides greater elimination in COD and colour proficiency, and pH 3 is chosen as the optimal in electrolytic destruction of organic and inorganic complexes in leachate and all types of waste water. In addition, the alkaline state of pH enhances the Fe³⁺ precipitation process and, therefore, stops the revitalization of ferrous ions. Consequently, it leads to a drop in Fenton's oxidation reaction. Correspondingly, self-decomposition of hydrogen peroxide is developed in alkaline solution at pH upstairs 5. Besides, the steady hydroxyl radicals are exceedingly yielded during the pH range between 2 and 4 with high oxidising capability.

The 3D plot represents the role of two variables, upholding all other variables at their static levels, which is shown in Figs. 8 and 9. According to the literature, 3D surface and equivalent contour plots provide a strong and direct estimation of the effects of the tested test variable quantity. In this plot, the impact of pH state and treatment time on the COD removal of leachate by the aerated ECP method was displayed. In accordance with the profile of the 3D surface, greater reduction of COD concentration was gained by increasing the treatment time than by changing the pH from an acidic state to an alkaline state of leachate. Therefore, it could be decided that the treatment time is a more dominant

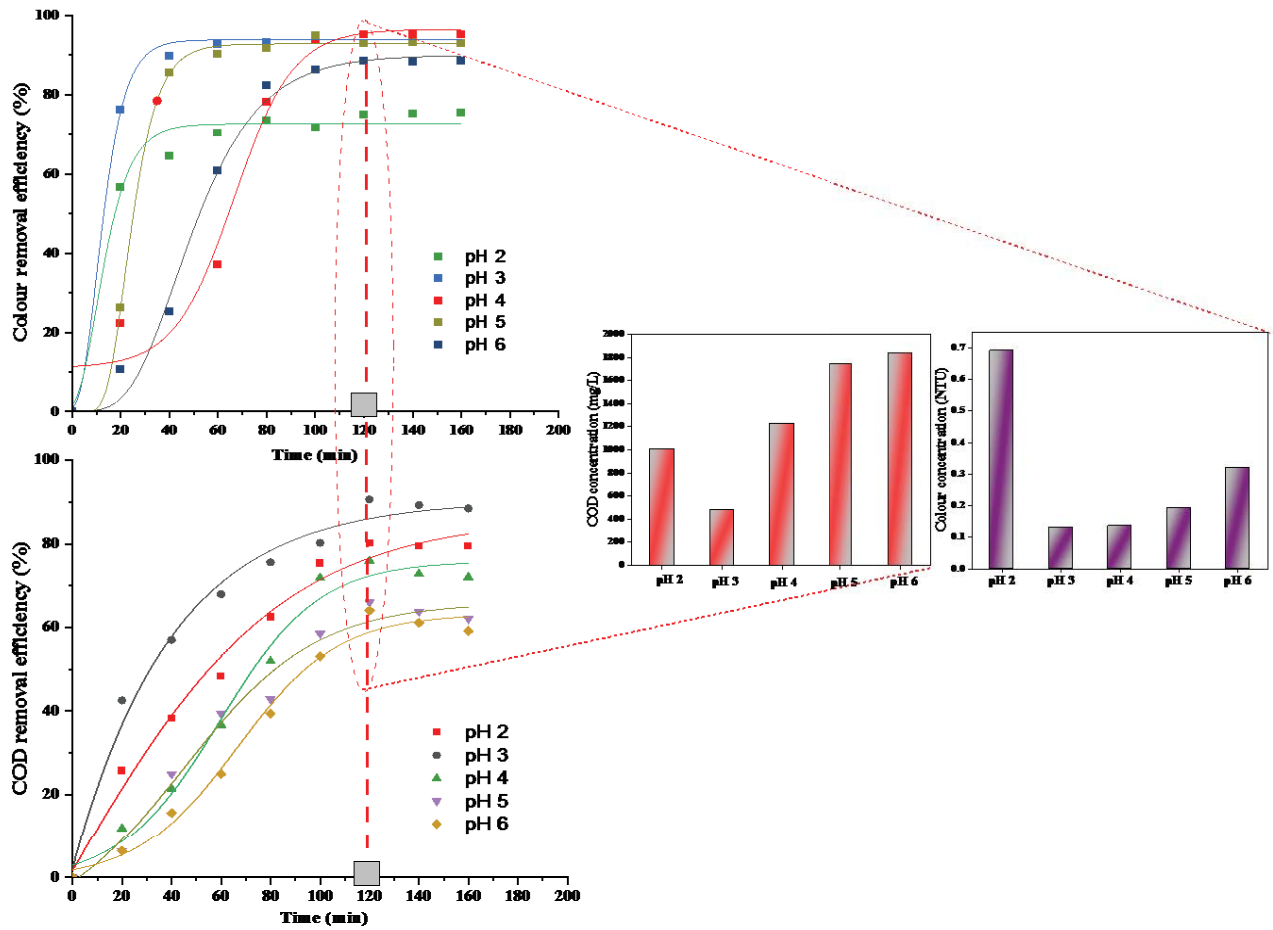


Fig. 7. Effect of pH on COD and colour removal from solid waste landfill leachate.

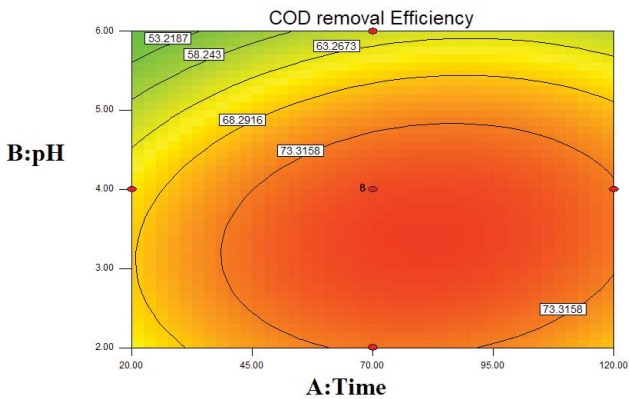


Fig. 8. Contour illustration display the effects of the correlative relations between pH and time.

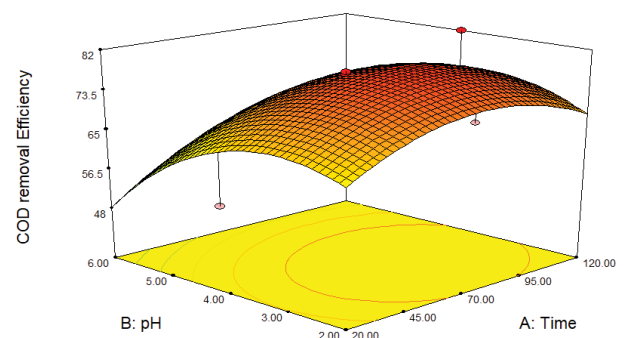


Fig. 9. 3D response surface shows the effects of the correlative relations between pH and time [$H_2O_2 = 3 \text{ ml/L}$, voltage = 5 V, $Fe^{2+} = 1.5 \text{ g/L}$].

parameter than the pH state. Likewise, it was apparent that the COD removal increased as the initial pH and reaction time increased to their central levels, that is, pH 3 and 120 min, and thereafter the removal decreased slightly as the pH and reaction time further increased to their high levels.

3.3. Effects of H_2O_2

In the electrochemical advanced peroxidation process, H_2O_2 is a key component because it is used for the production of OH radicals and to carry out the deterioration of recalcitrant organics present in waste water by Fenton's

chemistry. With the intention of analysing the consequence of the initial concentration of H_2O_2 in the electrochemical cell reaction and to determine the impact of COD and colour removal efficiency, H_2O_2 was varied between 1 and 6 ml/L under the conditions of voltage = 6 V, Fe^{2+} = 1.5 g/L and pH = 3, space among electrodes = 3 cm, and with the arrangement of Fe–Al electrodes. According to the results shown in Fig. 10, the removal efficiency of COD and colour increased linearly from 63% to 91% and 75% to 95% when the H_2O_2 dosage was increased from 1 to 3 ml/L. However, when the H_2O_2 dosage was increased to 6 ml/L in the electrochemical peroxidation reaction, the removal efficiency of biodegradable and non-biodegradable organic load decreased. Therefore, the optimum concentration essential for H_2O_2 dosage was fixed as 3 ml/L for further studies. According to literature studies, the excess addition of hydrogen peroxide beyond the optimum dosage will lead to the consumption of free hydroxyl radicals and a reduction in COD elimination from leachate due to the scavenging impact of H_2O_2 and re-joining of hydroxyl radicals. Herein, iron and aluminium were used as anodic electrodes and cathodic electrodes to harvest Fe^{2+} and Al^{3+} particles and H_2O_2 was taken as a corroding reagent to liberate hydroxyl radicals in the aerated electrochemical peroxidation process. Moreover, aeration is supplied to enhance the availability of oxygen for the system.

Daiana Seibert et al. [26] investigated sanitary landfill leachate treatment using a two-stage integrated system photo-electro-Fenton and biological oxidation processes under optimum conditions of current intensity = 2.3 A, H_2O_2 = 9,000 mg/L, Fe^{2+} = 60 mg/L, electrolysis time = 45 min, pH = 3.5–4.5, and an increase in BOD5/COD = 0.4. The interconnection between the pH and the H_2O_2 on the removal of COD rate is evidently clear. As exposed in this plot, the maximum COD removal is attainable at a very high concentration of H_2O_2 . This circumstance may be understood by reason of the high absorption of recalcitrant and non-biodegradable organic solids in solid waste landfill leachate. Still, the surplus of H_2O_2 dosage could negatively distress the COD removal efficiency. Furthermore, increased H_2O_2 concentration may have an inhibitory effect due to hydroxyl radical scavenging and the formation of an added radical (HO_2^*), allowing for significantly less oxidation than HO^* . Furthermore, a three-dimensional (3D) response surface and contour plot are presented in Figs. 11 and 12. This plot denotes the roles of two variables, keeping all other variables at static levels. As supposed by the literature, 3D surface and equivalent contour plots afford a stout and straight estimation of the effects of the studied test variable quantities. In accordance with the profile of the 3D surface, greater removal of COD degradation was attained by varying the dosage of H_2O_2 from 0 to 6 ml/L than by altering pH the

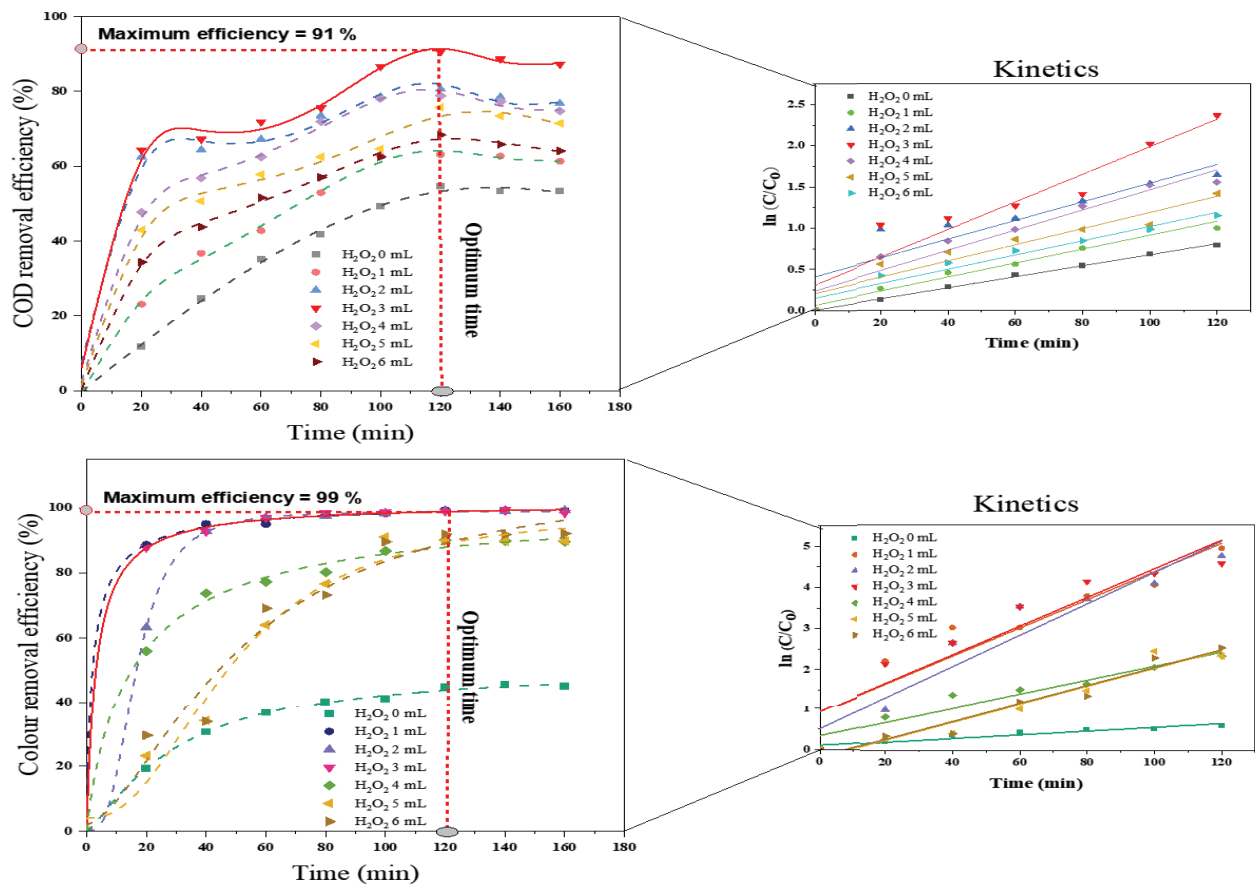


Fig. 10. Effect of H_2O_2 on COD and colour removal from solid waste landfill leachate.

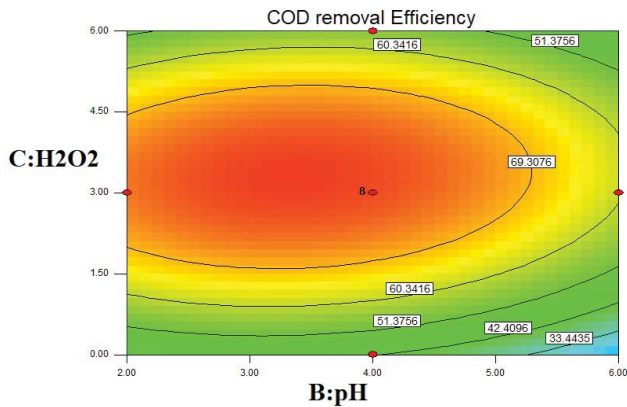


Fig. 11. Contour illustration display the effects of the correlative relations between pH and H_2O_2 .

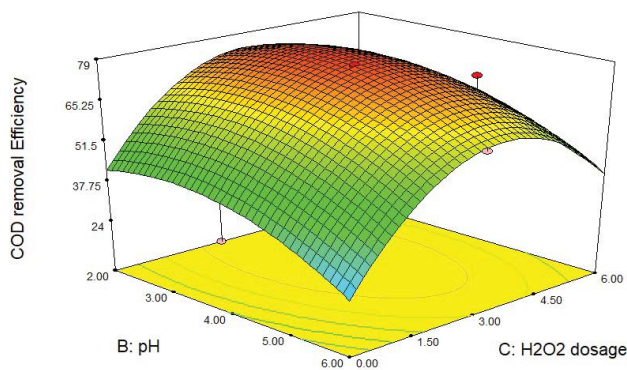


Fig. 12. 3D response surface shows the impacts of the correlative relations between pH and H_2O_2 [Time = 70 min, voltage = 5 V, Fe^{2+} = 1.5 g/L].

state of the leachate. Hence, it can be concluded that H_2O_2 is a more leading parameter as compared with pH in this ECP treatment.

3.4. Effects of Fe^{2+}

The upshot of Fe^{2+} absorption on the COD and colour removal was estimated by fluctuating the dosage of Fe^{2+} in the range of 0–2.5 g/L with H_2O_2 addition of 3 ml/L, voltage of 6 V and at acidic pH = 3. During electrolytic conversion of solid waste landfill leachate, 90% of colour was removed within 40 min and a maximum of around 95% of COD reduction occurred at Fe^{2+} measure was about 1.5 g/L and hence, it was reserved as the optimum quantity for the elimination of organic substances from leachate. Moreover, without the addition of Fe^{2+} in this electrolytic process, the removal of COD was very low. According to previous work, it was necessary to add appropriate Fe^{2+} for the commencement of Fenton's action to destroy the recalcitrant organics present in solid waste landfill leachate. The degradation of COD rises as Fe^{2+} dosage increases and it contracted after the top dosage of Fe^{2+} particles in accordance with Fig. 13. Moreover, while increasing the volume of Fe^{2+} concentration from 0 to 1.5 g/L, COD removal reached 54% to 91%

due to the catalytic agent ferrous sulfate, which quickly disintegrates H_2O_2 . Thereafter, the use of added iron turned out to be unproductive in electrolysis. Moreover, in this electrolytic peroxidation process, ferric ions were renewed into ferrous ions at the iron cathode electrode in the way of cyclic Fenton's chain chemical reactions. In addition, the gap between the two electrodes was too high, which will lead to the reduced mass transport of ferric ions to the cathode electrode and its effects on lessened ferrous ion rejuvenation. Consequently, it affects the yield of hydroxyl radicals produced competently from ferrous ion. Similarly, adopting a larger gap between electrodes will lead to heavy energy consumption of this system. In contrast, the space between electrodes was very low, which would cause the oxidative conversion of ferrous ions into ferric ions on the anode electrode and prevent the usefulness of Fenton progression. Additionally, Fe^{2+} dosage is outside the optimum 1.5 g/L range to scavenge the upshot of OH radicals produced.

Wang et al. [27] investigated an intensification in the rate of DMP degradation using the E-Fenton process, while increasing the initial Fe^{2+} concentrations from 0.5 to 1 mmol/L. Beyond that, the $\cdot OH$ concentration was weakened and, consequently, the DMP degradation kinetics was diminished due to the formed Fe^{3+} responding with H_2O_2 to harvest hydroperoxyl radicals ($HO_2\cdot$) but not hydroxyl radicals. Henceforth, the oxidation ability of hydroperoxyl is considerably lower than that of hydroxyl radicals. As well, the build-up of surplus ferrous ions brings about the establishment of a yellow precipitate of $Fe(OH)_3$ throughout electrolysis. These flocs come to be dropped at the lowest point of the electrochemical cell and on the electrode exterior surface. As soon as it has been dumped, this Fe^{3+} will not change into Fe^{2+} , hence the readiness of Fe^{2+} is diminished to respond with available H_2O_2 . This is the reason for the lowest productivity detected with the gradual alteration in ferrous ion dosage. In Figs. 14 and 15 the influence of Fe^{2+} and time on the COD removal of leachate by the aerated ECP process is presented in the form of 3D and contour plot. Consistent with the figure of the 3D surface, more removal of COD was reached by fluctuating the Fe^{2+} than by changing time and it can be determined that Fe^{2+} is a more important parameter than time.

3.5. Effects of voltage

In aerated ECP processes, applied voltage is very significant among all the parameters for creating an electrolytic field for directing the oxidation and reduction reactions at a very high frequency in the electrolytic treatment cell. Besides, the voltage range to be applied among electrodes is very imperative in estimating the energy consumption cost and treatment proficiency. It also interferes with the production of H_2O_2 , the rejuvenation of Fe^{2+} , and the formation of $OH\cdot$. The applied voltage between electrodes will perform in such a way as to yield more oxidised reactive iron (Fe) particles at the anode surface, while on the other hand, it will form sufficient HO at the cathode surface with simultaneous oxygen reduction reactions. In addition, the formed Fe^{2+} at the anode combines with H_2O_2 and harvests more active hydroxyl radicals for degradation of high-strength organic components of landfill stabilised leachate.

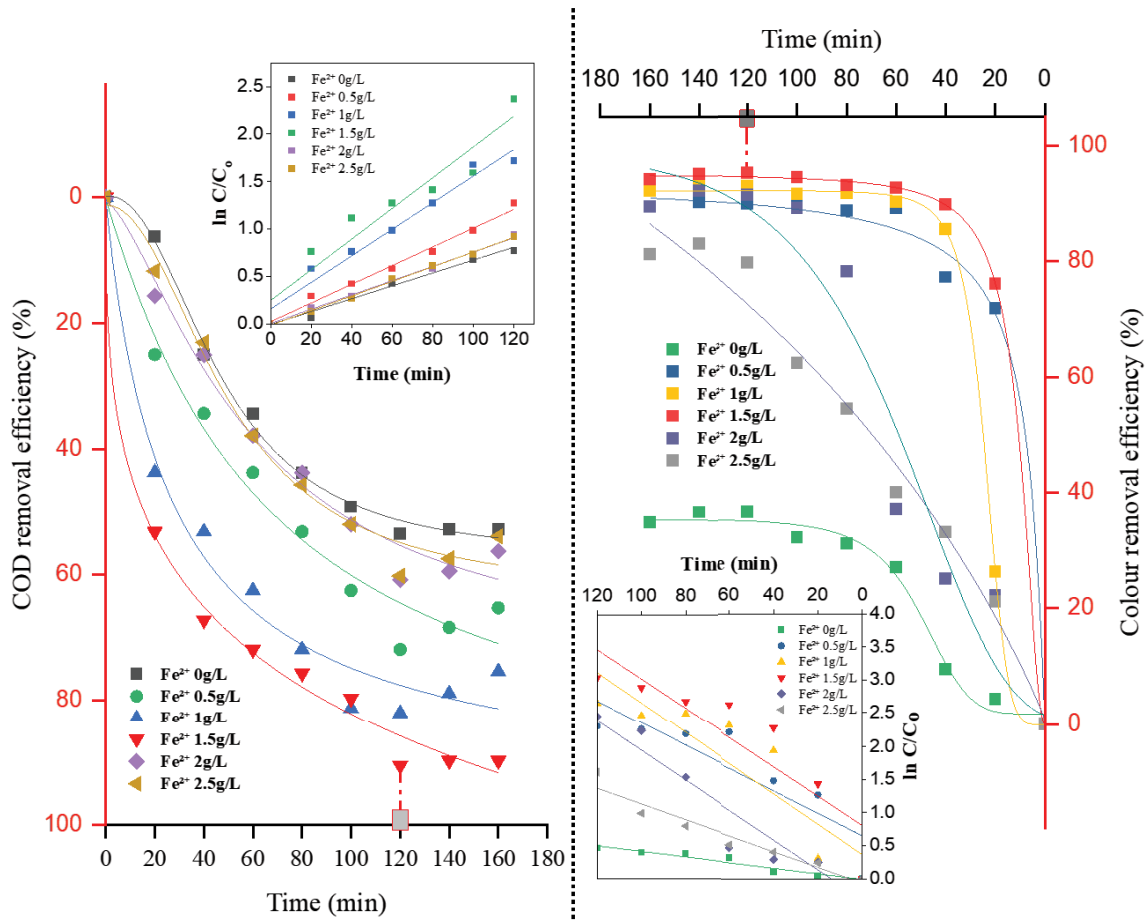


Fig. 13. Effect of Fe^{2+} on COD and colour removal from solid waste landfill leachate.

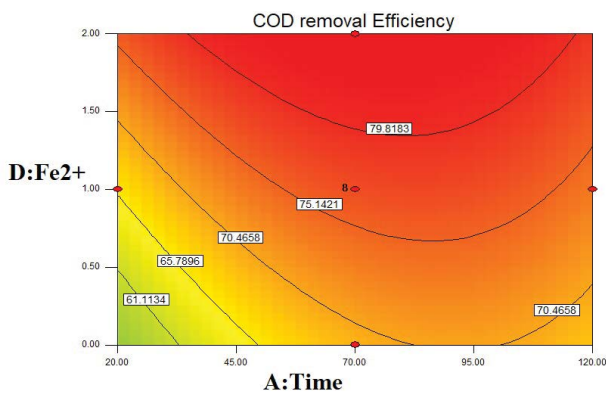


Fig. 14. Contour illustration display the effects of the correlative relations between time and Fe^{2+} .

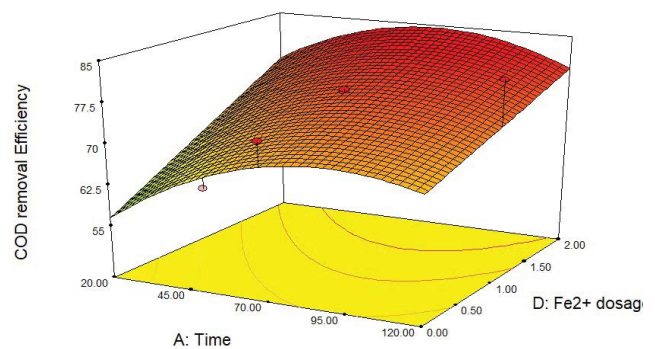


Fig. 15. 3D response surface displays the impacts of the correlative relations between time and Fe^{2+} [pH = 3, voltage = 5 V, $H_2O_2 = 3$ ml/L].

Direct and linear proportionality has been observed while elevating the voltage with respect to a straight step up in the Fe^{2+} dosage. Henceforth, a keen observation has been made on the response of iron electrodes under different voltage applications according to Fig. 16.

In a previous research study on the EF process, Sruthi et al. [28] found the COD elimination was 87.5% in stabilised

landfill leachate functioning at pH 3 and 4 V current, with a ferric ion dosage of 25 mg/L. The process of electro-regeneration of Fe^{2+} on the iron anode was enhanced by the increment in the significant amount of voltage potential. Batch experiments were carried out on five different voltages, such as 2, 4, 6, 8, and 10 V, under different treatment time breaks ranging from 0 to 160 min at optimised conditions of pH 3, H_2O_2 3 ml/L, Fe^{2+} 1.5 g/L, and a 3 cm gap

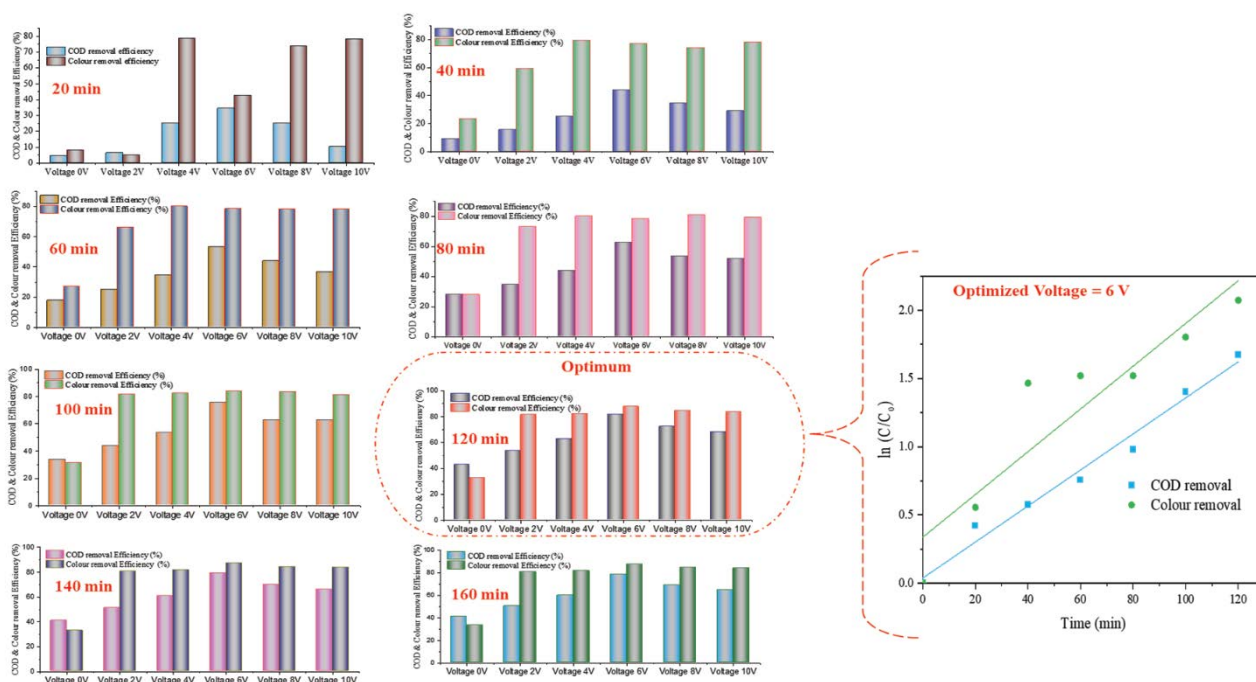


Fig. 16. Effect of voltage on COD and colour removal from solid waste landfill leachate.

between electrodes in order to discover the applied voltage variation required for the ideal condition of the aerated ECP process. The aerated ECP process was best at working with 81% COD and 87% colour elimination when raising the voltage from 2 to 6 V, and beyond this, there was no noteworthy removal of COD and colour up to 10 V due to parasitic adversative conversion and chemical reactions, and simultaneously steady state was achieved with oxygen production and hydrogen development. Additionally, H_2O_2 is concentrated in water during high voltage, which further decreases the effectiveness of aerated ECP treatment. Moreover, the excess quantity of generated Fe^{2+} ion particles scavenges the OH radicals [29]. According to the 3D and contour plots in Figs. 17 and 18, the effect of Fe^{2+} concentration is much stronger than the effect of voltage applied between electrodes. According to the obtained results, when the voltage increased from 2.0 to 6 V, the COD removal efficiency increased gradually to 81%. Compared to the initial pH and the reaction time, the voltage affected the COD removal most for the electrochemical peroxidation process. It seemed that higher voltage, according to Faraday's law, sped up anodic scarification and the production of Fe^{2+} ions, which is a main element required to run the Fenton reaction.

3.6. Effects of electrode material and its combinations

The overall treatment efficiency of the electrochemical advanced oxidation process will depend on the assortment of the best quality materials for electrodes, and their price is an important factor for aiding cost-effective treatment applications. Because the inappropriate adoption of unstable materials as electrodes will affect the working

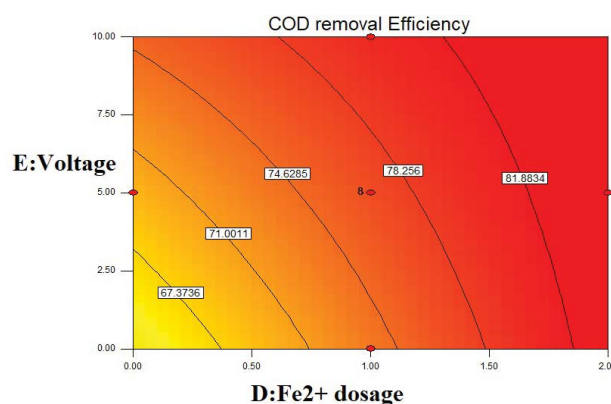


Fig. 17. Contour illustration displays the effects of the correlative relations between Fe^{2+} and voltage.

mechanism and usefulness of the electrodes during the electrolytic process, the electrodes will disintegrate into useless form and finally collapse. In the EAOP process, the choice of electrode material is based on obtainability in the market and best in handling and practice, as well as inexpensive in price. Such materials are iron, stainless steel, aluminium, carbon, copper etc. In this batch work, two different types of electrodes such as Al and Fe, were used to determine the removal of COD and colour from solid waste landfill leachate by aerated ECP process, under the optimised condition of pH 3, H_2O_2 3 mL and Fe^{2+} 1.5 g/L concentration and voltage 6 V were obtained from the earlier steps of the treatment process. Likewise, the batch study was conducted on different combinations of electrodes as

anodes and cathodes. There were five combinations that have been elected and tested, such as Al–Al, Al–Fe, Fe–Fe, Fe–Al, and Fe–Fe–Al–Fe. According to earlier studies, iron and aluminium materials are not only cheap but also act as very good coagulants in flocculating organic pollutants in all types of waste water treatment. Thus, the reduction of COD and colour was achieved not only by Fenton’s oxidation and reduction reactions but also by the combined action of electro-coagulation and flocculation of particles in solid waste landfill leachate. Additionally, rejuvenation and generation of iron particles have been enriched while using iron as a self-destructive anode. Consequently, the oxidation of hydrogen peroxide has been enhanced with Fenton’s reaction through iron as a vital catalytic agent in the electrochemical translation of recalcitrant organic substances into simple end products [30].

The effect of different combinations of aluminium and iron electrodes in the ECP reactor on the COD and colour removal with an electrode arrangement of 3cm gap is

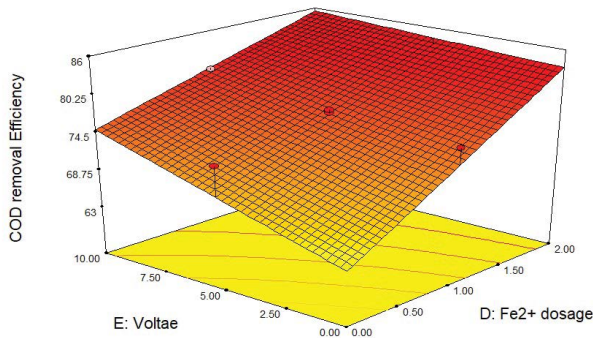


Fig. 18. 3D response surface shows the effects of the correlative relations between Fe^{2+} and voltage [pH = 4, time = 70 min, $H_2O_2 = 3$ ml/L].

shown in Fig. 19. The lowest COD and colour removal were 41% and 31% with the Al–Al electrode pair within 120min of operation among the tests. Further, with modification in the grouping of electrode pairs, the COD elimination efficiency was amplified in the order of 73% (Al–Fe), 81% (Fe–Fe), 91% (Fe–Al) and 90% (Fe–Fe–Al–Al) respectively.

3.7. Effects of reaction time

Zhang et al. [31] conducted research studies at 20–60 min in the electrochemical peroxidation process, and the optimum condition was found to be 40 min for mineralization of solids present in the landfill leachate. Similarly, in this study, the aerated electrochemical peroxidation process was subjected to a treatment time range of 0–160 min (Fig. 20) displays the consequence of reaction time on the removal of COD in aerated electrochemical peroxidation processes.

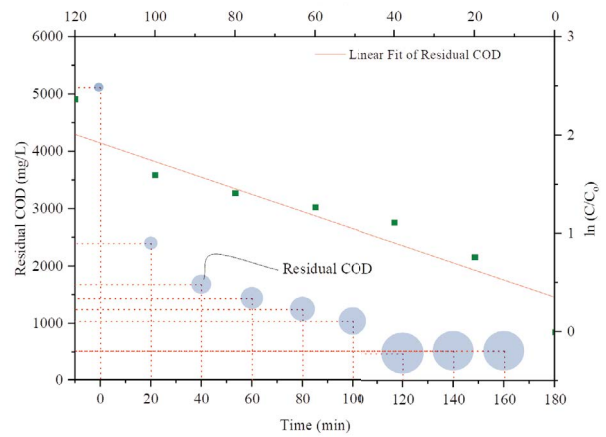


Fig. 20. Effect’s reaction time on aerated ECP process.

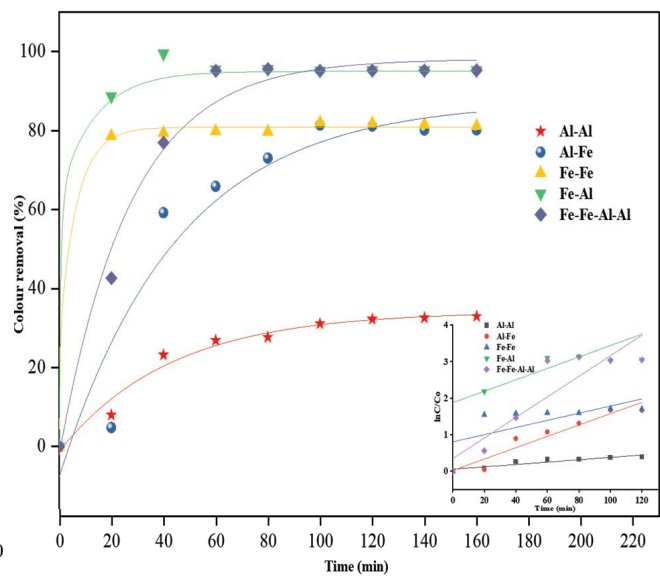
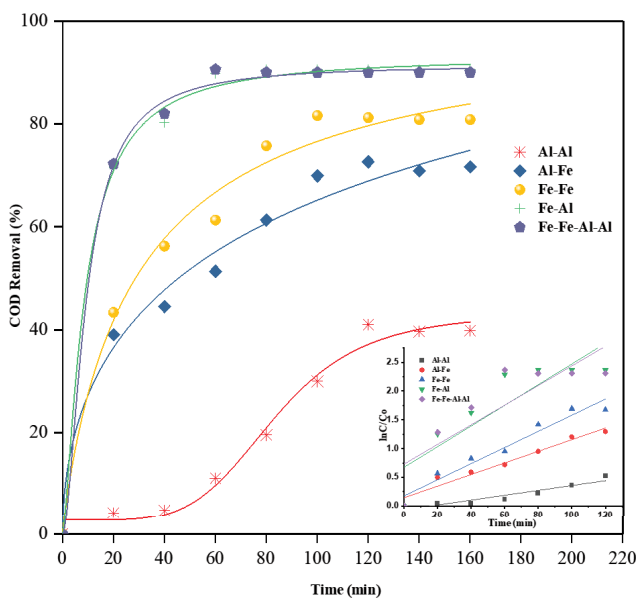


Fig. 19. Effect of electrode on COD and colour removal from landfill leachate.

The gained outcomes exhibited that COD removal abilities improved by time. The electrolytic precipitation and flocculated sedimentation of dissolved and colloidal organics during the settling process enhanced the COD and colour removal efficiency. Additionally, the quantity of iron and aluminium hydroxide flocs formed will impact the COD degradation efficiency. Furthermore, COD removal efficiency was elevated throughout the first half of 60mins of treatment time, and afterward, the maximum removal of 91% was attained gradually at 120 min of treatment time, and beyond this treatment time, the COD degradation stabilized. Since the amount of precipitate formed was low during the first shorter time periods of treatment and since the portion of COD is solvable, diminished elimination effectiveness was detected at the initial stage of electrolytic treatment.

3.8. Effects of aeration

The consequence of aeration in ECP treatment was observed with various time interval gaps under the acidic range of pH 3. The removal of COD with and without aeration is shown in Fig. 21. Moreover, the application of

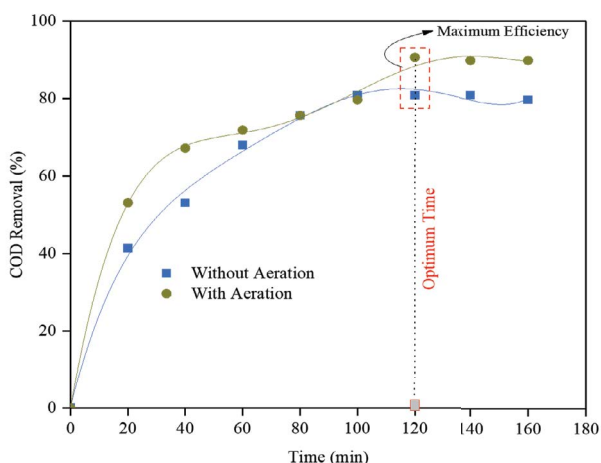


Fig. 21. Effects of aeration.

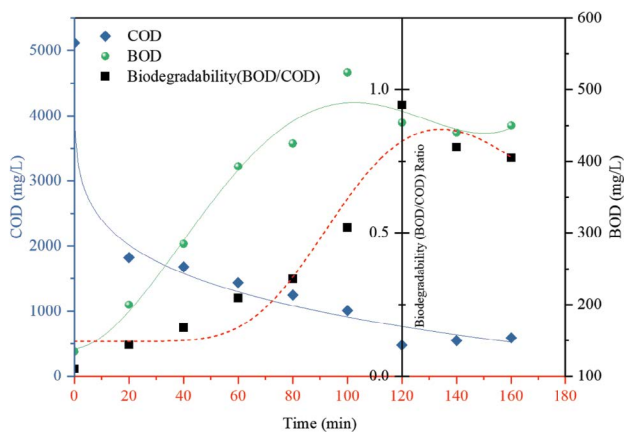


Fig. 22. Biodegradability of leachate.

supplying aeration during electrolytic treatment enhanced the degradation of COD removal by 10%. Thus, the treatment efficiency of the ECP process was elevated as a result of aeration. Furthermore, aeration in electrochemical peroxidation accelerates the chemical oxidation reaction on the anode surface, increasing the production of Fe²⁺ and Al³⁺ ions particles, which are triggered to aid in the formation of Al(OH)₃ and Fe(OH)₂ flocs. Likewise, in the former phase of the electrochemical reaction, the development of (OH) ions particles was at a significant percentage. But the intensification in treatment time strengthens the production of hydroxide ions at the cathode during electrolytic treatment. Consequently, the COD reduction was taking place constantly by means of increasing treatment time owing to the occurrence of enhanced collaboration among the metal hydroxide flocs formation and pollutants [32].

3.9. Electrical energy consumption

Electrical energy usage was one of the most important concerns for the successful adoption of electrochemical wastewater treatment. Therefore, the energy consumption of the aerated electrochemical peroxidation process was observed and designed with respect to the treatment time and quantity of leachate to be treated by the following equation:

$$\text{Energy Consumption} = \frac{VIt}{60Q} \text{ in kWh/m}^3 \quad (21)$$

where V is the applied voltage (V); I is the current development (A); t is the duration of treatment (min); Q is the capacity of leachate (L). Energy consumption for removal of COD from landfill leachate under optimum condition was 3.6 kWh/m³. Moreover, the operation and maintenance costs of electrochemical treatment processes were dependent upon the energy consumption during treatment. The applied electrical potential, current and dissolution of the electrodes into their corresponding ionic forms directly distress the features of treatment cost and effectiveness of the process. In addition to this, the quantity of electrode liquified was designed theoretically by using the Faraday's law:

$$C_E = \frac{I \times T \times M_w}{Z \times F \times V} \quad (22)$$

where C_E (kg/m³) is the iron or aluminum electrode consumption in the electrolytic cell, I is the current intensity (A), T is the treatment time (s), M_w is the molecular mass of the electrode (g/mol), Z is the number of electrons transferred ($Z_{\text{Fe\&Al}} = 3$), F is the Faraday constant (96,487 C/mol), and V is the volume of the treated leachate (m³).

4. Kinetics

With the intention of diagnosing the precise mechanism of the aerated ECP process for COD degradation in solid waste landfill leachate, the first order kinetics was developed. A first-order reaction is defined as a reaction that performs at a rate that is subjected directly to only one reactant's attentiveness.

$$\ln\left(\frac{C_0}{C_t}\right) = kt \quad (23)$$

$$\log\left(\frac{C_0}{C_t}\right) = \frac{kt}{2.303} \quad (24)$$

where C_0 , C_t , t , and k are the initial and final COD concentrations, response time, and rate constant (min^{-1}), respectively. The experimental statistics seems to fit the linear kinetic equation, and consequently the decomposition effectiveness tracks the first-order kinetic model. The value of the kinetic rate constant was 0.02 min^{-1} , and the first-order model was appropriate for analysing this treatment study. Hence, the first-order kinetic model outcomes displayed that the reduction of COD by means of aerated ECP treatment is very prospective to come to pass on the exterior of the electrode.

4.1. Effect of biodegradability of landfill leachate

The biodegradability index (fraction of BOD/COD) is the ability of persistent organic compound present in the leachate to be fragmented into simpler elements like CO_2 and H_2O by means of microbes. The presence of a large number of rigidly biodegradable COD like recalcitrant organic fragments, primarily humic and fulvic particles, in landfill leachate is revealed by a BOD/COD fraction less than 0.1. The biodegradability enrichment of landfill leachate was accomplished by means of an aerated electrochemical peroxidation process. The enhancement in biodegradability of the leachate was assessed with the determination of BOD, COD parameters after treatment as well as its proportion. The attained outcomes are revealed in Fig. 22. Primarily, the BOD/COD proportion of the leachate before the treatment process was 0.03. Then, subsequently treating the leachate through an aerated electrochemical peroxidation procedure, the biodegradability tendency of recalcitrant organic components was boosted and it was detected that at 60 min of reaction time, the BOD/COD proportion touched 0.27, which shows the readiness of organic components of leachate to be biodegraded in a quicker way. Further, increase in the treatment duration of the aerated ECP process, the biodegradability index elevated to 0.95 during 120 min of reaction. Shao et al. [33] carried out research on electrolytic degradation of bio-refractory organics and ammonia in leachate from bioreactor landfills under optimised conditions of current density of 100 mA/cm^2 , electrolysis time of 1.5 h, and electrode gap of 1 cm with 56% of COD with an initial concentration of 1.2 g/L. Therefore, the BOD/COD ratio could be improved to 0.3.

5. Conclusion

Solid waste landfill leachate was treated by the aerated electrochemical peroxidation method by changing the combination of aluminium and iron electrodes in the electrolytic cell on COD and colour removal. The successful application of face-centred central composite design and response surface methodology (RSM) in the construction of quadratic models with high implication and high R^2

coefficients. Thus, the response surface models were precise for forecasting experimental analysis and results, and could replicate not only the linear impacts but also the collaboration effects of crucial variables concerning the COD degradation of the solid waste landfill leachate. Moreover, response surface methodology was implemented in this work to find the optimal trial situations for the aerated ECP process. It has been exposed to produce statistically consistent outcomes for treatment of leachate. Therefore, the optimal circumstances of treatment were explored to be pH 3, H_2O_2 3 ml/L, Fe^{2+} 1.5 g/L with best combination of electrode Fe-AL pair enhanced the performance of this aerated electrochemical peroxidation system in such a way of producing high yield of hydroxyl radical's production and rapid regeneration of Fe^{2+} under treatment time of 120 min. Also, 91% of COD degradation has been achieved under experienced functioning environments with a reaction rate constant of 0.02 min^{-1} . The maximum removal of COD and colour is attained with the removal mechanism of organic substances by the combined application of electro-coagulation and electro-Fenton. Also, it is clearly concluded that organic substances can be efficiently removed from the solid waste landfill leachate by adding hydrolyzing electro-coagulants such as Fe^{2+} and Al^{3+} . In addition, there were two main process mechanisms concerning the efficiency of electrochemical peroxidation. The first process is bonding of cationic metal species to anionic sites then it leads to the neutralisation of organic substances, later the lessening of their solubility. The second treatment mechanism is based on the absorption of humic constituents onto the formed amorphous metal hydroxide precipitates. Likewise, acceptance of the quadratic model analysis with the experimental statistics was adequate. Similarly, analysis of variance disclosed virtuous coefficient of determination values and standards ($R^2 > 0.99$). Hence, aerated electrochemical peroxidation is suggested as an influential method for the mineralization of organic and inorganic components and decolorization of landfill leachate in the best combination of iron and aluminium electrodes.

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