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Optimization of Turbidity Removal from Domestic Wastewater by Electrocoagulation Using Aluminum Electrodes: A Design of Experiments Approach

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Abstract

A significant quantity of pollutants are contained within domestic wastewater which creates a substantial environmental issue with a large quantity of effluent that contains high amounts of contaminants. Turbidity is a major indicator of water quality and a measure of suspended solids. The purpose of this investigation was to study the use of electrocoagulation (EC) as a method of removing turbidity from municipal wastewater using aluminum electrodes. Using a Design of Experiments (DOE) approach, specifically Response Surface Methodology (RSM), the effect of three important operating variables was studied. These were: the initial pH of the wastewater in the range from 3 to 9; the current (or amperage, ranged from 0.1 A to 1.1 A); and the time for which the wastewater was treated by the EC process (ranged from 10 minutes to 20 minutes). The initial turbidity of each of the municipal wastewaters used in the testing remained constant at 336 NTU (nephelometric turbidity units) throughout the entire investigation. The effect of a number of different experiments was made in order to evaluate the effectiveness of the EC process for removing turbidity from the municipal wastewaters, and in addition take a measure of a predictive model of turbidity removal efficiency. The main conclusion drawn from the investigation was that the EC process will be very effective for removing turbidity from municipal wastewaters, which can vary from 5% removal to total removal (as high as 97%). There appeared to be a statistical correlation between the removal efficiency and the three experimental variables: pH (r = 0.4316); amperage (r = 0.3714); and time of treatment (r = 0.3965). The removal efficiency was highest using the variables of Run 8 whereby the pH was equal to 9, the current was held constant at 0.6 A and the treatment time was 10 minutes, resulting in a turbidity removal efficiency of 97%. The various data showed that both slightly acid (pH = 6) and alkaline (pH = 9) gave a markedly superior removal than acid (pH = 3) for obtaining constant, high removal efficiencies (average of 90.00% and 90.33%, respectively). Also, it was determined that a current of 0.6 A provided the most optimum amperage, giving an average removal efficiency of 95.33%. In addition, it was shown that long treatment times resulted in high removal efficiency, with the most averages of removal efficiencies recorded when the time of treatment was set.

1. Introduction

In addition to a growing population and an increasing need for more land due to urbanization and industrialization, there is a growing demand on the earth's limited amount of fresh water; as a result, water is becoming increasingly scarce. At one time, it was thought that the lack of water was confined to arid regions of the globe, however, today, it is now considered a global issue; with billions of people being impacted, the future of sustainable development is at risk [1]. As a result, the management and treatment of wastewater will be extremely important. A large portion of the worldwide amount of wastewater produced is created domestically. If discharged into the environment without proper treatment, it will greatly pollute the waters that we rely upon [2] This type of wastewater is a mixture of many substances including suspended solids, organic and inorganic compounds, nutrients, and pathogens. The presence of these types of substances can cause the degradation of receiving water bodies, create adverse effects on aquatic ecosystems through eutrophication, and create serious health concerns for humans [3]. Turbidity is one of the primary physical properties of wastewater. Turbidity refers to the cloudiness or haziness of water caused by suspended and colloidal particles. Turbidity does not only affect the aesthetic appeal of water, but it also shields microorganisms from disinfectants; thus, reduces the disinfectant process's ability to kill them and presents a clear danger to the safety of water [4]. Therefore, in order to reclaim and utilize domestic wastewater for non-potable purposes such as irrigation of crops, industrial processes, or flushing toilets, etc. that will significantly reduce the pressures on our supply of fresh water, the efficient removal of turbidity will be crucial. Physical treatments such as sedimentation and filtration, and biological treatments such as activated sludge have been used for decades. However, each of these has serious disadvantages. For example, chemical coagulation is usually done by adding chemicals, such as alum or ferric chloride, to the raw water. The added chemicals produce a very large amount of sludge that must be treated and disposed of and may introduce secondary pollutants into the water [5]. Like biological treatments, electrochemical treatments tend to be very sensitive to the quantity of pollutants and the temperature under which the treatment occurs, require substantial land space, and require extended periods of time to retain pollutants [6]. Due to deficiencies in conventional wastewater treatment systems, there has been a growing interest in developing new and alternative methods of wastewater treatment that are more environmentally friendly, economic and efficient. One class of alternative technologies for the treatment of water and wastewater includes electrochemical technologies that may be versatile and suitable for various treatment scenarios [7, 8]. Electrocoagulation (EC), a technology that uses electrochemical reactions to remove contaminants from wastewater by forming coagulant in-situ through the sacrificial oxidation of a sacrificial anode which can be aluminum (Al) or iron (Fe), represents an area of growing popularity as an inexpensive, efficient and cost-effective approach to treat many different types of wastewaters including domestic, industrial and agricultural effluent [9, 10, 11]. When a direct current is applied across two electrodes, the anodic reaction causes the sacrificial anode to oxidize and release metal ions (e.g. Al^{3+} or Fe^{2+}/Fe^{3+}) into the wastewater [12, 8]. At the same time, the cathodic reaction produces hydrogen gas and hydroxyl ions (OH^{-}) [13, 14]. The metal ions produced through the anodic reaction undergo rapid hydrolysis forming a variety of mono- and polymetallic hydroxy complexes [15]. These metal-hydroxy species have coagulant properties acting to destabilize and aggregate suspended and colloidal particles causing turbidity in the water by mechanisms of charge neutralization and sweep flocculation [16, 14]. Additionally, the hydrogen gas bubbles produced at the cathode can aid in the removal of the formed aggregates through a process called electro flotation, lifting the aggregates to the surface of the water where they can be removed [16, 13].

Aluminum electrode usage is extremely common in EC systems because of some very valuable properties. Aluminum is inexpensive and available, nontoxic, and has a high valency as well, making it a good coagulant [17, 18]. When aluminum is dissolved from the electrode to form aluminum hydroxide flocs $Al(OH)_3$, they are quite amorphous and have large surface areas, so they are excellent at trapping and removing pollutants [19]. Electrocoagulation provides many advantages over traditional chemical coagulation, such as: removal efficiencies for a wide range of contaminants, less sludge, no chemicals to store or handle, easy automation, and smaller equipment footprints for easier installation of decentralized treatment systems [20, 21]

The interaction between the operating conditions in electrocoagulation systems greatly affect how efficiently the electrocoagulation system will remove contaminants. The three main parameters that control the effectiveness and efficiency of electrocoagulation include: the initial pH of the contaminant containing water being treated, the current density (total current / area) applied to the electrodes, and the length of time the electrodes are energized (the treatment or electrolysis time) [22]. The pH of the water being treated is considered the master variable in determining the extent of contamination removal since it dictates the type of aluminum hydroxide that forms based on the pH level of the solution and the charge on the surface of the contaminants [8]. In terms of aluminum electrodes, there are multiple aluminum hydroxide compounds that can exist depending upon the pH of the solution being treated with aluminum electrodes, therefore impacting the ability of aluminum to effectively remove contaminants. The amount of current density applied to the electrodes controls the rate of formation of aluminum hydroxide (through anodic dissolution) and the rate of hydrogen bubble formation, therefore controlling the rate of contaminant removal and the energy required to treat the contaminated water [23]. Typically increasing the current density will increase the rate of contaminant removal however it will also increase the cost of treating the contaminated water [24]. Finally, the treatment time determines the amount of coagulant that is added to the contaminated water, the time that the coagulant is mixed and allowed to form floc with the contaminants and therefore ultimately determines the efficiency of contaminant removal.

Effective removal of turbidity from domestic wastewater is essential for reuse and thereby, reduces pressure on the existing fresh water sources. Treated wastewater of relatively low turbidity could be reused efficiently for certain non-potable purposes. Such uses can include, but are not limited to, irrigation of farmland or landscaping, industrial processes requiring water of a particular quality level, flushing toilets in buildings and dust control on construction or mining sites. Facilitating such reuse, efficient turbidity removal helps to reduce the amount of potable water used as coolant, sparing valuable domestic drinking water reserves for key household applications.

Experimental design, an alternative to the traditional experimental approach using one factor at A time (OFAT) is beneficial for assessing interaction and non-linear relationships between multiple variables. Many have utilized experimental design to identify optimal operating conditions. The design of experiments (DOE) methodology was created as an improvement to OFAT by allowing for better use of the data collected from experiments. The DOE methodology includes response surface methodology (RSM), which contains the tools and methods to characterize and analyze systems where a variable of interest is dependent on multiple independent variables [25, 26, 27, 28]. RSM allows researchers to assess the individual effect of each independent variable and how these variables interact with one another. This information can then be utilized to identify the best possible operating conditions to Achieve the highest level of desired performance (i.e., Turbidity Removal Efficiency) while minimizing the number of experimental runs required to find those conditions [5]. Typical designs used to create a second order polynomial model to relate the independent variables to the response of interest include Box-Behnken Design (BBD) and Central Composite Design (CCD) [29].

Electrocoagulation (EC) has been developed as a promising and clean technology in the treatment of different types of wastewaters such as domestic effluent. Many studies investigated the removal performance of pollutants, for instance, turbidity and heavy metals, organic matters etc., with aluminum or iron electrode because they are cheap effective electrodes. Such as [8] has evaluated the efficiency of EC in treating pollutants and its correlation with operational parameters such as pH, current density, treatment time that always play a significant role in removal rate. Other studies, including [27]. are reported to be using RSM to optimize EC processes for different contaminants and a few researchers [30], urban wastewater. Although the overall applicability of EC has been demonstrated in such studies, many are limited to individual pollutants or use classical 'one factor at a time' (OFAT) approaches that do not efficiently capture interactions among variables.

To the best of authors' knowledge this paper is a novel contribution by application of an efficient Design of Experiments (DOE) method for selection, namely Response Surface Methodology (RSM), to find out the optimum operating conditions in electrocoagulation process in treatment of turbidity from domestic wastewater with aluminum electrode. Our work which does not appear to rely on less efficient experimental design (compared to other authors) or based more specifically on industrial effluent typology, has allowed for determining the independent and interaction effects of initial pH, applied current and treatment time towards the decolorization of turbidity. This well-designed and systematic study with a constant initial turbidity of 336 NTU not only determines optimal operating conditions, generates robust statistical model but also provides fundamental understanding of the process performance under different parameters. The detailed examination of the interaction of two different parameters, in particular, pH and current when both varying together (heatmap), gives a refined insight that has not been proposed up to date; thus, it becomes a feasible and optimized tool for design and operation of EC system for domestic wastewater treatment.

Therefore, this research is aimed at investigating how to optimize turbidity removal from household wastewater using an electrocoagulation process with aluminum electrodes. To analyze the impact of the initial pH, the current (amperage), and the treatment time as parameters on the electrocoagulation process, the design of experiments methodology (response surface methodology), has been used to study systematically the three variables. The main goals of this study are: (1) to assess individually and in interaction the influence of each parameter and the combination of two or all of them on the efficiency of turbidity removal; (2) to develop a mathematical model to represent statistically the process; and (3) to identify the operational conditions for which the electrocoagulation process can be optimized to remove the maximum amount of turbidity. This research will help to improve our knowledge of the EC process for the treatment of household wastewater [31, 32], and will provide a useful tool for designing and optimizing the operation of treatment processes for domestic wastewater.

2. Materials and methods

2.1. Wastewater characterization

This study utilized domestic wastewater that was initially characterized before the experiments began. It is primarily turbidity that is of interest with respect to the experimental results; however, the initial turbidity (Turb.1) of each experimental run was identical at 336 NTU. Thus, there was an identical starting point from which to compare the effectiveness of the process under various operating conditions. Although other physicochemical characteristics of the wastewater were present and representative of those typically present in domestic effluent, they were not the primary subject of investigation for this study.

2.2. Experimental setup

All electrocoagulation experiments were performed in a laboratory batch reactor that was made up of a 1-liter glass beaker. The Electrode Assembly consists of two sacrificial aluminum (Al) plates for both the anode and cathode. The electrodes are located vertically and parallel to each other with a set inter-electrode distance to provide a consistent electric field. Both electrodes were connected to a regulated DC power supply (sugon3005pm) which is able to provide a stable current. Mechanical agitator was used to provide a low intensity agitation approximately 100-150 revolutions per minute to enhance dispersal of generated coagulant and to stimulate formation of flocs, while preventing breakage of newly formed flocs. All experiments were conducted under ambient temperature approximately 25°C.

2.3. Experimental design and procedure

To study the effect of the control variables and to optimize the process, a design of experiments approach was used. The experimental runs were made using the response surface methodology framework. This study had three independent variables, these were initial pH (X_1), current in amperes (X_2), and treatment time in minutes (X_3). The experimental design consisted of 9 runs varying in levels of these 3 factors, the actual values for each run are given in the results section as in Table 1.

For each experimental run, the following procedure was followed:

Each experimental run consisted of the following steps:

1. Domestic wastewater was placed into the EC reactor.
2. The pH of the wastewater was adjusted to one of three levels pH 3, 6, and 9 with diluted hydrochloric acid (HCl) or sodium hydroxide (NaOH) to achieve the appropriate PH.
3. Aluminum electrodes were placed within the reactor; the current 0.1 A, 0.6 A, or 1.1 A was selected from the power supply for the experiment and activated at the same time as the magnetic stirrer.
4. Once the electrodes were energized, the DC power supply and mechanical stirrer began the electrocoagulation (EC) process and this process ran for the predetermined treatment time 10, 15, or 20 minutes.
5. Upon expiration of the treatment time, the power supply was shut down, the magnetic stirrer was stopped and the supernatant solution was allowed to sit undisturbed for 30 minutes to allow for floc sedimentation.
6. Once the flocs had settled, a sample of the clear supernatant was carefully collected from the upper portion of the supernatant for analysis.

2.4. Analytical methods

Turbidity in the pre-treatment wastewater (Turb.1), and post treatment wastewater (Turb.2) samples were determined by a turbidimeter commonly used for this purpose. Removal efficiency of the electro-coagulation process was determined by determining percent reduction in turbidity as illustrated by Eq. (1).

$$\text{Percent Reduction in Turbidity} = \left(\frac{\text{Turb.1} - \text{Turb.2}}{\text{Turb.1}}\right) \times 100 \quad (1)$$

Where, Turb.1 is the turbidity prior to treatment (336 NTU) and Turb.2 is the turbidity of the treated water. All data analysis (statistical evaluation and graphical illustration) was done based on the 9 experimental runs.

3. Results and discussion

The experimental evaluation studied how to optimize an electrocoagulation process for removing turbidity from a domestic wastewater. An experimental study was conducted to evaluate three parameters that could affect the efficiency of the electrocoagulation process; these parameters included pH levels, electrical current (or amperage) and duration of treatment time. The same initial turbidity value of 336 NTU was used as the starting point for all experimental treatments. A summary of the final turbidity values and the percent turbidity removal for each of the nine experimental runs is contained in (Table 1).

Table 1. Experimental design and results for turbidity removal by electrocoagulation.

Run	pH (X_1)	Amperage (A) (X_2)	Time (min) (X_3)	Initial Turbidity (NTU)	Final Turbidity (NTU)	Removal Percentage (%)
1	3	0.1	10	336	321	5
2	3	0.6	15	336	17.6	95
3	3	1.1	20	336	55.9	85
4	6	0.1	15	336	49	87
5	6	0.6	20	336	20	94
6	6	1.1	10	336	41	89
7	9	0.1	20	336	31.3	91
8	9	0.6	10	336	9.4	97
9	9	1.1	15	336	58	83

3.1. Statistical analysis and key findings

Statistical analysis was conducted on the results of the experiments to assess the influence of various operational parameters on the removal efficiency of turbidity. The mean of all 9 experiment runs was 80.67% for removal efficiency, and the standard deviation for removal efficiency was 28.76%. The wide range of removal efficiencies of the nine runs is evident in the relatively large standard deviation. Removal efficiencies for the nine runs varied greatly; ranging from 5% (Run 1) to 97% (Run 8). Moderate positive correlations between removal efficiencies and the three independent variables evaluated were found through correlation analysis; pH (Pearson's r = 0.4316), Amperage (r = 0.3714), and treatment time (r = 0.3965). It can therefore be inferred that an increase in each of the independent variables tested will lead to enhanced removal of turbidity, although interactions or non-linear effects of each independent variable cannot be ruled out.

3.2. Visual analysis of parameter effects

A set of plots were developed from the experimental data to provide further visualization of how each parameter affects process performance.

3.2.1. Effect of pH on turbidity removal

Figure 1 displays the impact of the original wastewater pH on the turbidity removal efficiency by utilizing a scatter plot. Results from tests conducted at pH 3 had significantly higher variability as compared to tests conducted at pH 6 and pH 9; removal efficiencies ranged from 5% to 95%. Removal efficiencies for all of the pH 6 and pH 9 test results exceeded 80%. Average removal efficiencies were 61.67%, 90.00%, and 90.33% for pH 3, pH 6, and pH 9, respectively. These results indicate that removal of turbidity by aluminum electrodes will be both efficient and reliable when the original wastewater is at a neutral to alkaline condition.

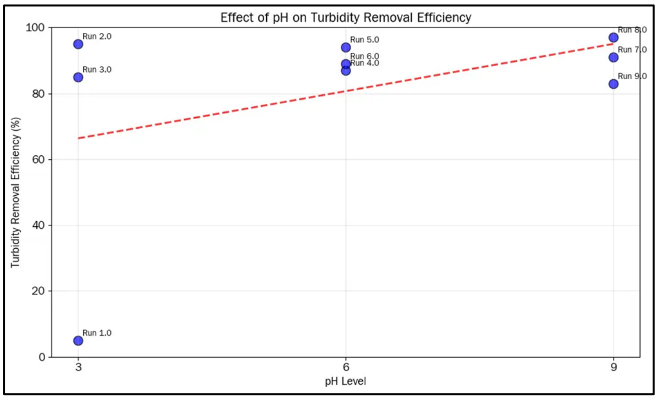


Fig. 1 Effect of pH on turbidity removal efficiency.

3.2.2. Effect of current (amperage) on turbidity removal

The relationship between turbidity removal and the applied amperage is shown in Fig. 2. Clearly the amperage has a large influence on the performance of the treatment process. At 0.1 A (the lowest amperage), the average removal achieved was only 61.00% Turbidity removal efficiencies were much larger and much more consistent as amperage increased to 0.6 A, which also yielded the largest and most consistent removal efficiencies, with an average efficiency of 95.33%. However, increasing the amperage to 1.1 A resulted in a slight decrease in average efficiency to 85.67%, which suggests that an optimal current in this range for generating sufficient coagulants exists, and that too high and amperage could result in passivation of the electrodes or excessive gas production, both of which are averse to performance.

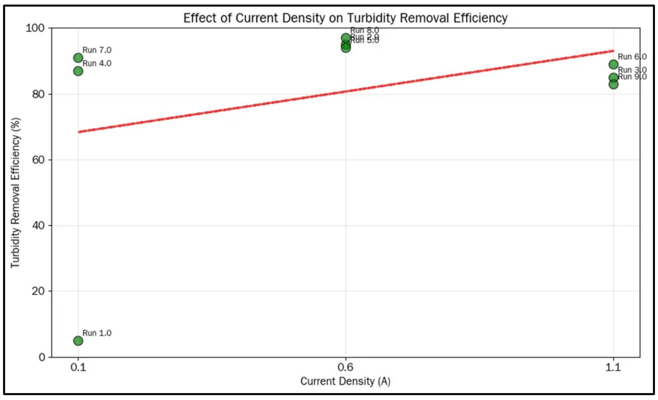


Fig. 2 Effect of current (amperage) on turbidity removal efficiency.

3.2.3. Effect of treatment time on turbidity removal

Figure 3 shows the relationship between electrolysis time and percent removal of turbidity. There appears to be a direct correlation between increased electrolysis time and better percent removal of turbidity; however, this may have been due to the fact that more time was available for aluminum coagulant production, along with more time for flocculation and particle aggregation. Turbidity removal efficiencies were 63.67%, 88.33%, and 90.00% after 10, 15, and 20 minutes respectively.

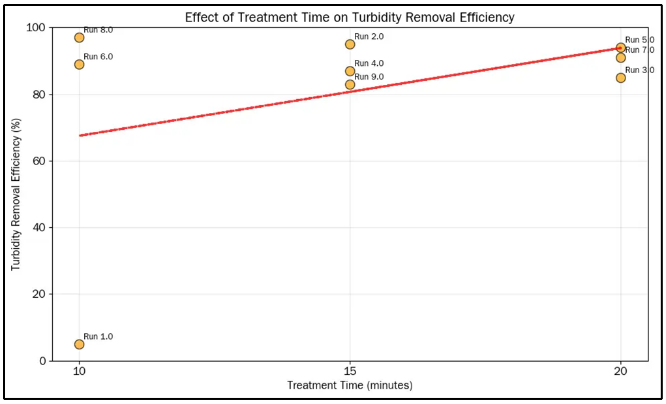


Fig. 3 Effect of treatment time on turbidity removal efficiency.

3.3. Overall performance and parameter interactions

Figures 4 and 5 show a general summary of performance for each run, as shown below. Figure 4 displays the turbidity levels at the end of each experimental run. The best turbidity level was found with Run 8, which had the lowest final turbidity of 9.4 NTU. At the other end of the spectrum, run 1 that was conducted under the least favorable conditions (pH was lower than the others, current was less than the others, duration was less than the others) produced a final turbidity of 321 NTU, or nearly the same as the initial turbidity.

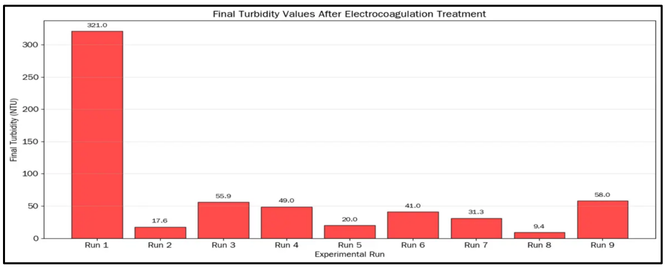


Fig. 4 Comparison of final turbidity values across experimental runs.

The color-coded bar chart (Fig. 5), is an effective representation of removal efficiency of all runs using color to determine category of performance. It is clear that the majority of experimental conditions (i.e., 7 of the 9 runs) produced high efficiencies (>80%). Only one run (Run 1) produced very low efficiency (<50%) which demonstrates that there are important parameters that need to be considered when setting up experimental conditions.

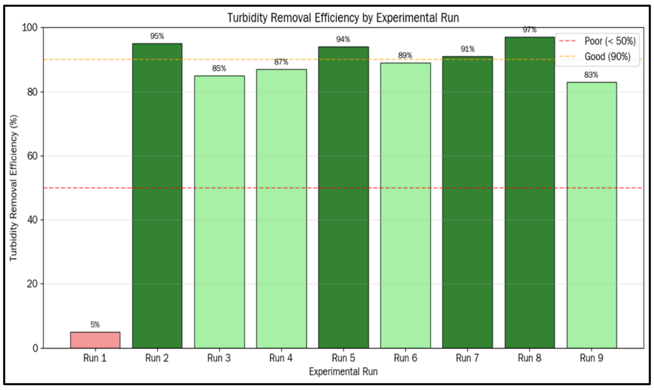


Fig. 5 Comparison of removal efficiency by experimental run.

An interaction heatmap and box plots were produced in order to investigate the combined effects of the parameters identified. Figure 6 presents a heat map displaying the interaction between the pH and current density on removal efficiency. Here it can easily be seen that there is an optimum zone of performance at a pH of 6 and 9 at the current of 0.6 A. This again indicates that the optimum levels of these two parameters when taken separately will also promote optimum results when taken in conjunction.

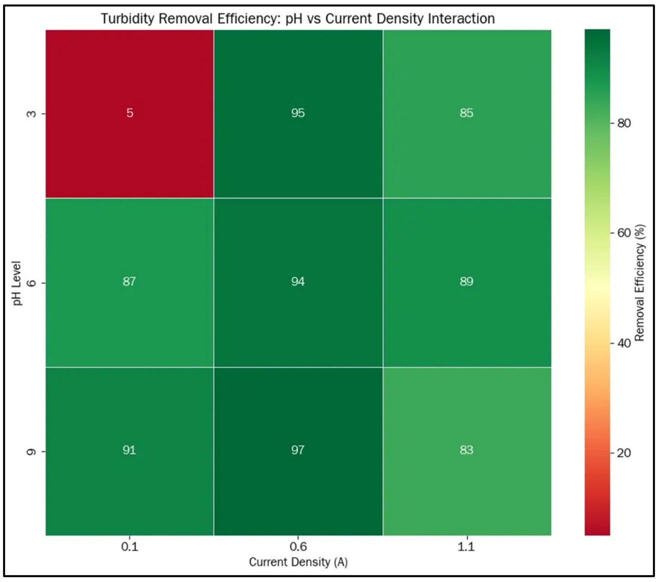
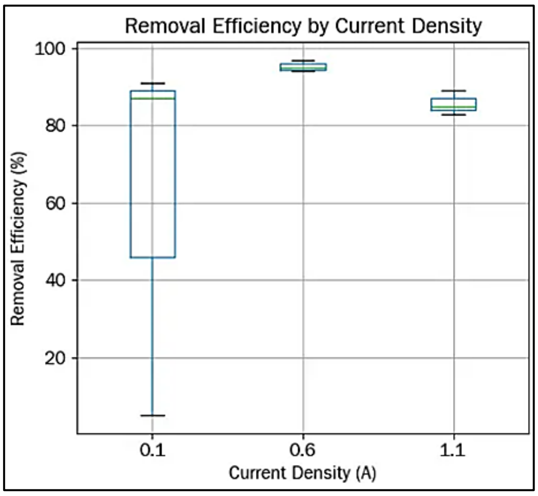


Fig. 6 Parameter interaction heatmap for pH vs. current density.

The box plots depicted in Fig. 7 graphically illustrate the variability of removal efficiencies of all levels of each of the three parameters. These graphs confirm the results of the previous analysis; i.e., pH levels of 6 and 9 have nearly equal distributions centered on the highest efficiency levels, which indicates that the systems operate consistently. The 0.6A current has the highest median and smallest variance. With respect to time, the median efficiency is seen to increase as the treatment duration goes from 10 min to 20 min.



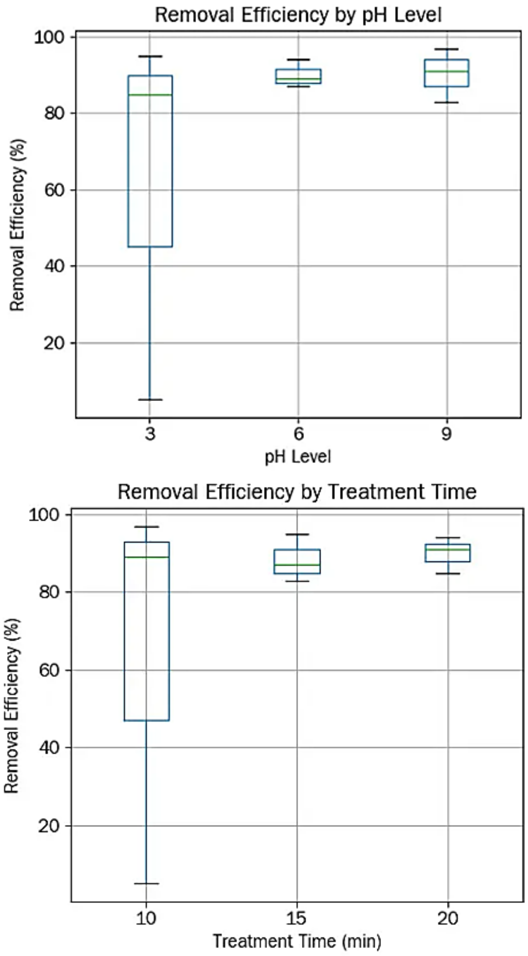


Fig. 7 Distribution of removal efficiency across parameter levels.

3.4. Optimal operating conditions

The combination of experimental data and analysis revealed the maximum turbidity reduction using an electrocoagulation process with aluminum electrodes from domestic wastewater under optimum operational conditions. The highest turbidity reduction in all experiments occurred in Run 8 with a turbidity reduction of 97% with pH 9, 0.6 A, and a contact time of 10 min. In addition to defining the single best run (Run 8), it has been possible to define an optimal operational window as follows:

pH: 6 to 9

Current (Amperage): 0.6 A

Treatment Time: 15 to 20 min.

Operational conditions within this window resulted in removal efficiencies greater than 88% for all runs, indicating the robustness and high potential of the optimized electro-coagulation process.

3.5. Effect of design parameters on removal

The results of this research demonstrate that using an Aluminum Electrode for Electrocoagulation (EC) is a highly effective method for removing Turbidity from Domestic Wastewater. The high levels of Turbidity removals observed in this study, ranging from zero percent to ninety-seven percent, suggest that EC may represent a viable alternative to traditional treatment technologies. In this study, a Design of Experiments approach was employed to identify the main variables influencing the performance of EC treatment (pH, Current, Treatment Time), to understand their interactions and to determine the Optimal Operating Conditions necessary for successful implementation of the EC Process. The results of this study are consistent with previous studies related to EC treatment systems but contain new and valuable information about the use of EC to treat domestic wastewater.

3.5.1. Effect of initial pH

Research has shown that the pH level of the wastewater is one of the most important factors to determine the effectiveness of turbidity removal for this process. In comparison to other EC studies [32, 8], the process was much more effective and stable when it was run under conditions of both neutral pH (pH = 6) and high pH (pH = 9) than under acidic pH (pH = 3). At the neutral pH 6 an average turbidity removal of 90.0% occurred while at the alkaline pH 9 an average turbidity removal of 90.3% occurred. These two averages were clearly higher than the average of 61.7% removal at the acidic pH 3. The reason for the difference in performance between these three pH levels can be explained by the different types of aluminum species present in the solution during the electrolysis process. When operating at low pH (around 3), the primary aluminum species in solution is the positively charged aluminum cation, Al^{3+}, which is less effective as a coagulant than its hydroxide forms [19]. As the pH level increased toward the neutral pH 6-8, Al^{3+} formed several polymerized species and the amorphous aluminum hydroxide solid, $Al(OH)_3(s)$. Aluminum hydroxide is a good coagulant due to its large surface area; thus, it promotes the removal of pollutants from the water via the sweep flocculation process. Sweep flocculation occurs when particulate matter is enmeshed in the growing precipitate. When the pH level increased further to around 9, the negatively charged aluminum hydroxyl ion, $Al(OH)_4^-$ began to form. Some researchers have reported that when the pH level is too high, the effectiveness of the removal decreases due to the formation of this soluble species. However, in this study the process continued to remove turbidity very effectively at pH 9. One possible explanation for this observation is that the localized pH near the cathode became highly alkaline due to the generation of hydroxide ion, even though the bulk solution pH may not accurately represent the complex electrochemical environment in which the process is taking place. The high effectiveness of the process at pH 9 in this study, especially in Run 8, where 97% of the turbidity was removed, supports findings reported by [21], who also found the greatest percentage of turbidity removal occurred at pH 9 when treating domestic wastewater using Al-Fe electrodes. The high degree of variability exhibited at pH 3 may have been caused by the opposing effects of neutralizing charges by the Al^{3+} cations and the incomplete development of hydroxide flocs.

3.5.2. Effect of current (Amperage)

The applied current, which is directly proportional to the current density, controls the rate of production of the coagulant, and the production of bubbles, which is an important factor in terms of the operating cost and efficiency of the process [23]. Our results showed that there was a clear optimum applied current of 0.6 A which had the maximum average removal efficiency of 95.33%. At the lower applied current of 0.1 A the removal was markedly lower (average of 61.00%). This can be expected since at lesser currents the rate of dissolution of Al3+ ions from the anode will be slower thus leading to an insufficient dose of coagulant being produced to destabilize and remove the particles which cause turbidity, within the time for which treatment is applied [22]. The applied current increases from 0.1 A to 0.6 A gave a marked improvement in efficiency since a sufficient amount of the flocs of aluminum hydroxide was produced for good coagulation to take place.

Interestingly when the applied current is increased further to 1.1 A the average removal efficiency, decreased slightly (85.67%). Such a phenomenon has been observed for EC studies and can be done to a number of factors. Firstly, too high operating current density can cause electrode passivation, whereby insulating oxide layer forms on the anode surface which prevents any further dissolution taking place [33]. Secondly when too high currents are applied there is violent production of hydrogen and oxygen gas bubbles, which produces violent turbulence in the reactor. This turbulence can lead to the breaking up of the newly formed flocs, which are still fragile, into smaller settleable particles thus decreasing the overall removal efficiency [10]. Therefore, an optimum current density exists which enables a sufficient dose of coagulant to be produced yet overcomes the deleterious effects of excessive gas evolution as well as excessive energy consumption. The other important aspect that we find is that the optimum at 0.6 A gives the important information that energy costs can be minimized since energy consumed is directly proportional to the current applied.

3.5.3. Effect of treatment time

The treatment time is the amount of time that electrolysis takes place for a given sample. Therefore, the treatment time has two major impacts on treatment outcomes: the amount of coagulant formed and the amount of time available for flocculation. This study demonstrated a strong positive correlation between treatment time and turbidity removal rates, with turbidity removal efficiencies increasing from an average of 63.67% at 10 minutes of treatment time to 90% at 20 minutes of treatment time. These findings demonstrate an expected trend supported by other research studies [30, 21].

As the treatment time increases, there will be more Al^{3+} ions released into the solution, therefore, more aluminum hydroxide flocs will form. More Al^{3+} ions in the solution will increase the chances of collisions between the coagulant species and colloids, therefore more destabilized and aggregated colloids will exist. Additionally, longer times allow for the formation of larger, denser and more stable flocs, which will be easier to remove via gravity separation (sedimentation).

It should also be noted that the relationship is not endless and that after a certain point most all pollutants will have been removed and additional time will provide little benefit but will continue to use energy and the electrodes [34]. The successful results from Run 8, which resulted in a 97% removal rate in only 10 minutes when both the pH and current were at the optimal levels, illustrates that the best removal rates can be obtained in a shorter treatment time, depending on the conditions of other parameters. The interaction between the various parameters studied in this study is illustrated in the heat map (Fig. 6) and it is essential to consider these interactions in addition to the individual effect of each parameter. The combination of pH 9 and 0.6A provided the most effective removal of turbidity in the shortest amount of time, nearly complete in 10 minutes.

3.5.4. Parameter interactions and process optimization

The strength of the DOE investigation lies in the determination of interaction of parameters. The heat map (Fig. 6) is a clear indication of the synergistic result of pH and current. The higher efficiencies lie in the region of pH 6-9 and a current of 0.6 A. This indicates that it is not the maximum of one parameter which is the determining factor in the optimization but rather the interrelationship of the parameters is the resultant factor giving good efficiencies. Thus, at the optimum current of 0.6 A the efficiencies of pH 6, and pH 9 are both good but at the non-optimum current of 0.1 A the increase of pH of the coagulants due to the presence of lime from 3-9 has a much more marked positive effect. This shows that at the low coagulant usage (low current) the chemical effect (pH) is even more important due to the limited coagulant present to get the maximum efficacy. The total analysis shows that there is an optimum range of coagulant usage for strong, high efficiency production of turbidity removal. The parameters indicated by the results (pH 6-9, 0.6 A, 15-20 min) are the practical guide to the operation of an EC plant for domestic waste water. The remarkable efficacy of Run 8 (pH 9, 0.6 A, 10 min) indicates that if it were a case of rapid treatment required then the higher range of the optimum pH gives advantages in lesser time of treatment and less treatment difficulty of the reaction vessel, and hence lesser capital cost. The study satisfactorily illustrates the instrument ability of the RSM which is needed to traverse through the complex parameter space of the EC process, and hence allow the inspiring of a highly efficacious treatment system and being able to minimize the amount of experimental work [28]. The conclusions give a good basic for the scale up of the process and a techno-economical study for the checking of the performance of the system against the traditional systems [35].

5. Conclusions

The electrocoagulation (EC) process was successfully investigated in this study to determine the best conditions for removing turbidity from municipal wastewater with aluminum anodes. The effects of initial pH, current, and treatment time on both the EC process and turbidity removal were fully investigated by employing a well-defined design of experiments strategy that examined all single and combined effects of the three experimental parameters. As a result, the findings of this research can be summarized as follows:

1. Electrocoagulation was shown to be a very effective technology achieving a maximum turbidity removal of 97% from an initial value of 336 NTU which indicates its potential use as a strong technique for the treatment of domestic waste waters.
2. All three parameters of pH, current and time studied were shown to have a significant effect on the efficiency of removal. Neutral to alkaline conditions of pH 6-9 were superior to those which were acidic. An optimum current of 0.6 A was established which gave a good performance with no detrimental effects witnessed with higher currents. Longer periods of treatment generally increased efficiency which was only to be expected.

3. The optimum performance results were obtained from Run 8 with a pH of 9, a current of 0.6 A and a 10-minute treatment period. In general, a dependable zone of operation producing over 88% removal efficiency was obtained at a pH of 6-9, a current of 0.6 A and a treatment period of 15-20 minutes.
4. The study emphasizes the merits of employing Response Surface Methodology (RSM) in process optimization. This statistical method yielded not only the conditions for optimum operation, but also the interaction between the parameters studied which would not have been found with traditional methods of experimentation.

The results from this study form a strong foundation for using electrocoagulation as a pre-treatment or post-treatment treatment method for treating household wastewater. The optimum operating parameters determined during this study can be utilized in designing and running cost effective, compact and efficient electrocoagulation-based treatment systems. Work should focus on scaling up the electrocoagulation process, detailed techno-economic evaluation (electrode and energy usage), and removal of additional contaminants (COD, BOD and nutrients) at the optimized conditions established in this study.

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