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Evaluate the performance of the electrocoagulation method to remove the reactive green dye using aluminum electrodes

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ABSTRACT

The electrocoagulation technology is one of the effective methods used for wastewater treatment. Numerous environmental and sustainability advantages make electrocoagulation an increasingly attractive alternative for treating the water. In this study, the electrocoagulation method was used for removing reactive green dye from water by evaluating some of the operating parameters. Several experiments were conducted using aluminium electrodes. Factors considered include pH level (4.5, 7.5 and 10.5), distances between electrodes (0.5, 1, 2, and 3 cm), electrolysis (5, 10, 15 and 20 minutes), and applied electric current (5, 7.5, 10, and 12.5 volts). The optimal removal efficiency of reactive green dye was achieved under conditions of 12.5 volts electric current, pH of 7.5, for 20 minutes, and an interelectrode distance of 1 cm. The maximum removal efficiency was 98.82%. Electrochemical treatments have garnered considerable attention based on their simplicity and effectiveness.

Keywords: Electrocoagulation, water, treatment, reactive green dye.

INTRODUCTION

Lack of water is also another serious problem many countries face, mostly because of the over-extraction of fresh water resources (Huang et al., 2021). The problem is also caused by the contamination of water sources by domestic, agricultural, and industrial wastages. In areas surrounding industrial setups, surface waters and groundwater sources are also highly contaminated by dangerous substances from inadequately treated industrial wastewater, heavy metals, and reactive green dye (Gemeda et al., 2021; Yeh et al., 2022). Among these, the problem of synthetic dyes is highly serious because these are widely used in various industrial setups, including the manufacture of textiles, pigments, ink, paints, pharmaceutical products, polymers, paper production, and cosmetics, thus contribute largely to the problem of environmental contamination (Islam et al., 2023). The problem caused by synthetic dyes is highly toxic, with serious health impacts due to their harmful or toxic properties, which are generally categorized on the basis of the period of exposure acute or chronic toxicity (Ruel-Bergeron et al., 2015). In addition to the problem of toxicity, the health impacts caused by the consumption or exposure to these also include teratogenic, mutagenic, genotoxic, damage to central nervous system, gastrointestinal system disorders, and nerve impairments (Ramamurthy, et al. 2024). A major part of the dye is also generally fugitive or unbound materials in the products processed from them, meaning their eventual release into the water resources is due to the inevitable breakdown of these products in the waters (Jorge, et al 2024). With serious impacts on the health of humans, animals, and plants, because their entrances generally create hazardous conditions in the water resources, seriously threatening the survival or well-being of all beings thriving in or linked with these water resources (Khatri, 2015).

Electrocoagulation is an innovative technology applied to water treatment and wastewater treatment as well (Nidheesh et al., 2021; Titchou et al., 2021; Boinpally, et al., 2023). The treatment process integrates three major processes, which are electrochemistry, coagulation, and flotation, in one step only (Wang, et al., 2021). The treatment process is conducted with the help of two electrodes, the anode and the cathode, connected with a direct electric current supply, hence facilitating the course of electrolysis (Bharath et al., 2019). In the process of electrocoagulation, the current passes between the two metal electrodes, most likely made from iron or aluminum, hence releasing metal ions, which work as coagulants inside the process. The coagulants remove contaminants, heavy metals, suspended particulates, oils, or microorganisms, which then sink or float due to the process of sedimentation or flotation, respectively, depending on the material properties (Iwuozor, 2019). The advantages of the electrocoagulation process over the conventional coagulation process are numerous. The process uses fewer chemicals, hence creating less sludge with the possibility of causing secondary pollution, often caused by the conventional process, which uses alum or ferric chloride, another chemical, hence contaminating the entire water body with harmful residues left by the chemicals in the process (Canizares et al., 2009).

Aluminum (Al) is also often used as the electrode material for the process of electrocoagulation because of the material's lower cost of operation and higher efficiency (Afriani and Tiandho, 2020; Gafoor et al., 2021). During the process of electrocoagulation, the formation of coagulants occurs due to the electrochemical dissolution of the aluminum ion (Al^{3+}) from the sacrificial anodes, which leads to the formation of flocs of the pollutants. The process of formation of flocs in the process of electrocoagulation is similar to those that are generated by the process of chemical coagulation. Simultaneous electrolysis occurs on the cathode, from which the formation of the hydroxyl ion (OH^-) or the H^+ ion occurs, depending on the mechanism involved in the reaction, leading to the pH of the solution turning to be mostly neutral, ranging between 6.8 and 7.2, throughout the process. Additionally, the formation of H^2 gas, which is important for the formation of the buoyancy effect on the flocs, acting as the main force or mechanism involved in the flotation of the flocs to the surface of the container or the reactor. The sizes of the H^2 gas bubble generally falls between 20 and 70 µm, depending on the current used or the pH level of the solution in the process (Barrera-Díaz et al., 2018). Also, according to Cavaliere, (2023), near the neutral pH the bubble sizes be the smallest in size as shown in Figure 1.

The current study aims to evaluate and explore the impact of different operational variables in the electrocoagulation process, namely electrolysis time, pH, electric current, electrode distance, electrode pair, and concentration, on the efficiency

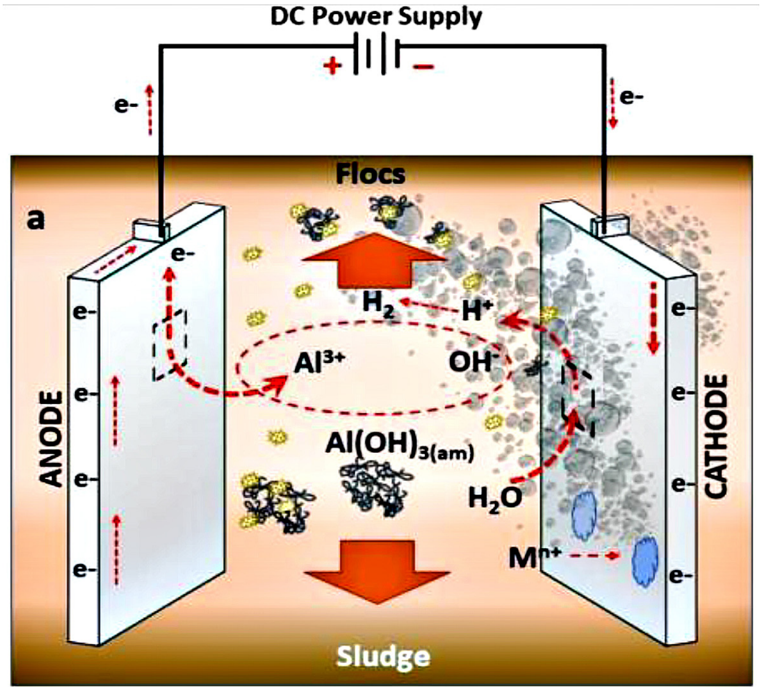

Figure 1. Mechanism of electrocoagulation (Cavaliere, 2023)

of removal of reactive green dye from water. The sustainability of the electrocoagulation process is in its efficiency in cutting down the requirement for the use of chemical additives, in turn reducing the production of chemical waste, hence lowering the levels of environmental pollution.

MATERIALS AND METHODS

Materials

Reactive green dye is a synthetic anionic dye commonly employed in textile industry applications for coloring cellulosic fibers including cotton, wool, and silk. This dye is also known to fall into the category of reactive azo dyes, which possess functional groups to form covalent bonds with hydroxyl or amino groups of fibers, which results in strong colors as well as stability of dyes. The dye is also known to be highly water-soluble, having an intense green shade of coloring, absorbing in a wavelength range of approximately 620–640 nm. For wastewater treatment research, reactive green dye is commonly used as a model pollutant in consideration of stability, biodegradability, as well as possible toxicity in untreated wastewater discharge to water bodies as environmental hazards. Typically, reactive green dye has a molecular structure consisting of sulfonate functional groups ($-SO_3^-$) responsible for increasing water solubility but hindering removal from aqueous solution. For this work, reactive green dye was employed to examine the effectiveness of electrocoagulation as a function of varying conditions, namely pH, conductivity, reaction temperature, electric voltage, and the interspace of electrodes, as well as time of electrocoagulation. The reactive green dye used in the study has a molecular weight of 571.46 g/mol, with the formula C32H16CoN8. All other chemicals used in the experiments are of analytical grade, meaning the highest purity, and are used without purification (Aljeboree, 2023). The molecular structure of the reactive green dye is shown below in Figure 2.

Experimental set-up

The electrocoagulation system treats the contaminated water, as illustrated in Figure 3. The system consisted of an electrochemical cell with a total volume of 1000 mL, wherein the treatment by both electrocoagulation and mixing took place in one basin only. The design employed a turbine impeller for the homogenous distribution of coagulants throughout the solution, with the inclusion of two aluminum (Al) electrodes acting as baffles to reduce the formation of vortices inside the reactor in which the reaction took place. The other two electrodes are mounted inside the basin to act as the working electrode pair, with distances between the electrodes changed alternatively to 0.5, 1, 2, and 3 cm. Following number of experiments, the electrodes are thoroughly cleaned with water for consistency in their efficiency. The level of removal of reactive green dye was evaluated with the help of different operational variables, which include pH value, electrode distances, current, and the time for the process of electrolysis, with sampling carried out with the help of water according to the different specified conditions of the experiment. The effluent from the electrocoagulation process was then filtered with the help of qualitative filter paper with a pore size of 0.9 cm. The filtered samples were then transferred to clean glass vials, and the efficiency of the dye removal was evaluated with the help of the UV-Vis Spectrophotometer Model PD-303UV.

Different parameters of water quality have been considered to determine the factors affecting the electrocoagulation process. The factors include pH, conductivity, reaction temperature, and electric voltage, as well as the interspace of electrodes because they directly affect the electrochemical reaction process in an electrocoagulation system. The efficiency of reactive green dye removal has been calculated using Equation 1 (Kzar et al, 2019).

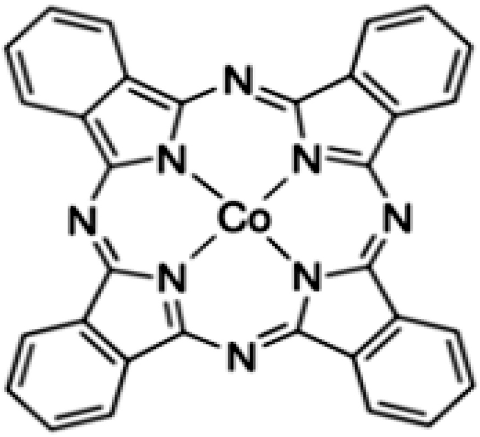

Figure 2. Structures for reactive green (RG) (Karam et al. 2020)

$$RDD\ efficiency\ \% = (Co - Ct)/Co \times 100 \quad (1)$$

where: Ct and Co are the reactive green dye value before and after electrocoagulation process.

RESULTS AND DISCUSSIONS

In order for effective water treatment to be achieved using the electrocoagulation process, a number of operational factors have to be considered. For purposes of research, the operational factors examined in this study include pH values, electrode distances, electrolysis time, as well as the electric current.

Effect of pH on removal efficiency

The pH value of the solution plays a very significant role in determining the species, solubility, and coagulation of the aluminum ions in the electrocoagulation process. In current study, the pH values of the water samples were set to 4.5, 7.5, and 10.5, which were achieved using sodium hydroxide (NaOH) solution and sulfuric acid (H_2SO_4), as confirmed using a calibrated pH meter. As explained in Abd Al-Hussein, 2021. Figure 4 depicts the impact of pH on the removal efficiency of reactive green dye. The account depicts findings pointing towards the impact of pH on the efficiency of the electrocoagulation process, where a maximum removal efficiency of 98.82% would be achieved at a pH value of 7.5.

In acidic mediums (pH 4.5), the removal efficiency showed a steady rise with time, reaching a value of about 78% in 20 minutes. In such a medium, aluminium species are mainly in the form of soluble ions like Al^{3+} and $Al(OH)^{2+}$. Since these types can destabilize some of the dye molecules, their affinity to remain in solution greatly restricts the formation of polymer hydroxide flocs, which are essential for pollutant aggregation. In addition, the intensity of hydrogen gas evolution is low at a lower pH, thereby reducing the formation of flotation bubbles. Thus, the pollutant-floc interaction becomes poor. In a neutral medium of pH 7.5, the removal efficiency significantly increases, reaching a highest value of about 98.82% in 15 minutes. The result can be attributed to the formation of the most optimal aluminium hydroxide species, which are mainly amorphous $Al(OH)_3$. The species have high adsorption forces, which facilitate strong neutralization of charges on the dye molecules.

Additionally, hydrogen gas evolution contributes to increasing the concentration of hydroxyl ions (OH^-) around the cathode, thus increasing the pH. This favors a process where both dissolution and precipitation occur, resulting in efficient coagulation. The quick attainment of nearly complete removal under slightly neutral conditions demonstrates that this process is most efficient between slightly and neutral pH.

Contrary to this effect, with a much higher alkalinity and a pH value of 10.5, the clearance efficiency reduced greatly, reaching only about 38% after 20 minutes. This can be attributed to the dominance of strongly negative aluminate ions ($Al(OH)_4^-$) in alkalotic medium. These compounds have a high degree of solubility and lower efficiency in dismantling negative dye molecules because of electrostatic repulsions. Furthermore, high levels of OH^- ion hinder the formation of Al^{3+} ion centers to produce adequate amounts of flocs, thus inhibiting coagulation. Thus, it can be noted that not many flocs will be formed, and there will be a great decrease in the clearance

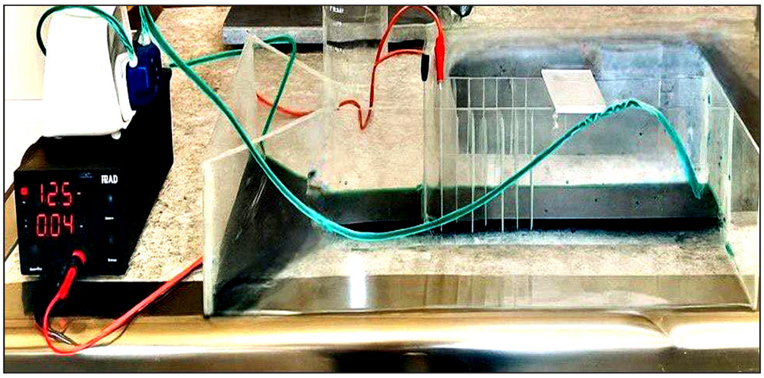

Figure 3. Experiment of the electrocoagulation process

efficiency. From this performance value, it can be noted that a fundamental environment inhibits the formation of precipitates for effective coagulant species. This is in agreement with Sharma et al. (2021) and Leng et al. (2022), which indicated similar levels in decaying contamination cleared with strongly alkalotic environments. From Figure 4, for all electrolysis times, it can be noted that the highest efficiency is attained in the first 10 minutes for both electrolyte concentrations, especially for pH 7.5. This is due to the highest rate of formation for aluminum hydroxides and bubbles. After this point, it levels off to reach a plateau because all dye molecules have been greatly destabilized or adsorbed on pre-formed flocs. The plateau for each efficiency can be taken to mean that any process past these 15 minutes will have minimal actual benefits.

These findings confirm that the efficiency of electrocoagulation is largely dependent on the pH value of the solution because it determines the species formed by aluminum and their activity. From these findings, it can be confirmed that it is imperative to control the pH value to 7.5, which is slightly alkaline. At these levels, efficient removal can be expected. At lower levels, it might not result in efficient removal due to ineffective formation or non-functional species due to the formation of aluminate complex. Thus, for efficient treatment of reactive green dye, it is necessary to aim for slightly alkaline levels.

Electrolysis time

Electrolysis time refers to the duration over which the electrochemical reaction occurs and plays a critical role in the performance of the electrocoagulation process. In this study, electrolysis was carried out for a total of 20 minutes, with removal efficiency evaluated at 5-minute intervals using the initial concentration as a baseline. Experimental results demonstrated that a 20-minute electrolysis time was generally sufficient for effective pollutant removal with maximal removal efficiency of reactive green dye (98.82%). Although longer durations led to increased removal efficiency, they also resulted in higher energy consumption (Figure 5). Extending electrolysis time enhances reactive green dye removal from water, as reported by Hassoune et al. (2024). This improvement is attributed to the increased generation of hydroxyl radicals and the formation of additional metal hydroxide and polymeric species, which improve coagulation efficiency (Asaithambi et al., 2016; Cui et al., 2020).

Electric current applied

Among the most influential parameters governing the performance of the electrocoagulation process is the applied electric current or voltage. In this study, voltages of 5, 7.5, 10, and 12.5 V were applied to investigate their effect on the removal efficiency of the reactive green dye at different electrolysis times (5, 10, 15, and 20 minutes), under controlled conditions of pH 7.5 and 1 cm electrode spacing using aluminium electrodes. The results presented in Figure 6 clearly show that both the applied voltage and the treatment duration significantly influence pollutant removal efficiency.

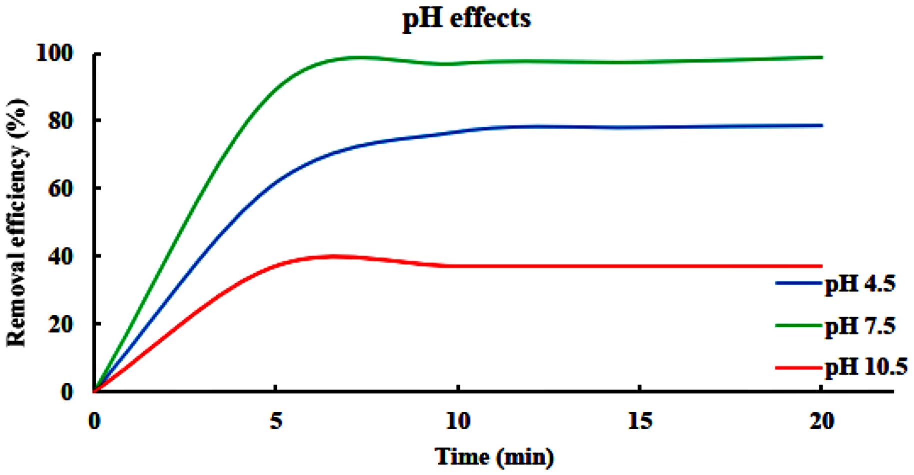

Figure 4. Removal efficiency of reactive green dye as a function of pH levels at electric current applied of 12.5 volts, electrode spacing of 1 cm, electrolysis time of (5, 10, 15, 20 min)

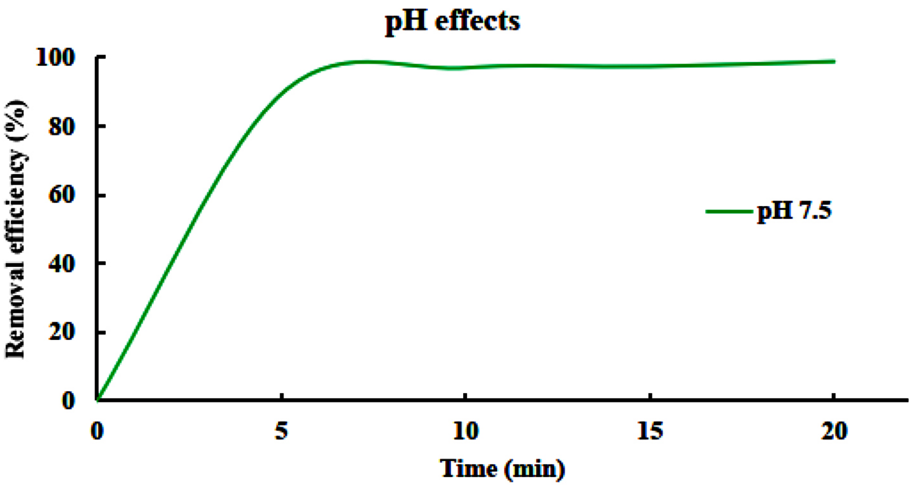

Figure 5. Removal efficiency of reactive green dye as a function of electrolysis time at electric current applied of 12.5 volts, electrode spacing of 1 cm, pH of 7.5

The removal efficiency was comparatively low at the initial stages (5 minutes) for all the applied voltages due to the fact that coagulant generation and floc formation are just starting. However, with the rise in electrolysis time, the removal efficiency increased gradually for all voltage values. Nonetheless, the rise of the graph became much sharper at higher voltages. A maximum of about 98.82% removal efficiency was achieved at 12.5 V within 20 minutes, which justifies the strong dependence of the electrocoagulation process on the applied electrical potential. This can be explained by the increased current density with an increase in voltage and, consequently, the rate of anodic dissolution of aluminium. When the voltage is increased, the anodic reaction precedes faster, releasing more Al^{3+} ions into solution. These ions undergo hydrolysis to form different aluminium hydroxide species such as $Al(OH)_2^+$, $Al(OH)_2^+$, and $Al(OH)_3$, responsible for the destabilisation of negatively charged dye molecules by adsorption and charge neutralisation. At the same time, increasing cathodic production of hydrogen gas bubbles enhances flotation and mixing, and thereby the contact between flocs and pollutants is greatly improved. The synergistic action accelerates the removal of the reactive green dye from the solution.

At lower voltages such as 5 and 7.5 V, the kinetics was slower, and the maximum removal efficiencies stayed below 70%. Lower potential translates into smaller current density and thus slower alum dissolution and coagulant formation rates. Moreover, reduced bubble generation causes less efficient mass transport and weaker floc-pollutant collisions, hence reducing overall efficiency. These conditions are good from the point of view of energy consumption because operation at low voltages reduces both power consumption and wear of electrodes. Increased removal efficiency with time and voltage was in agreement with the theory of electrochemical processes and the results obtained in several other studies. According to Hassoune et al. (2024), the value of an applied current (or voltage) regulates the rates of coagulant generation and gas evolution and, thus, directly influences the efficiency of pollutant removal. The linear growth of efficiency in the early stage and its gradual saturation after 15–20 minutes attest that the system is approaching a state of balance due to thorough destabilisation and removal of the available dye molecules. It is worth mentioning that too high voltages, while allowing for higher removal efficiency, may result in some side effects that are undesirable and thus limit their applicability: overheating, electrode passivation, and unjustified energy losses. High voltages also accelerate the corrosion of electrodes, reduce the service life of electrodes, and increase maintenance costs. Thus, although the highest removal efficiency was achieved in this work at 12.5 V, this value might not be the most reasonable option for use in continuous or large-scale processes.

Considering practical applications, operating voltage selection must be an optimisation of treatment efficiency, energy consumption, and electrode stability. Therefore the present findings reflect the importance of environmental engineers, researchers, and students interested in wastewater

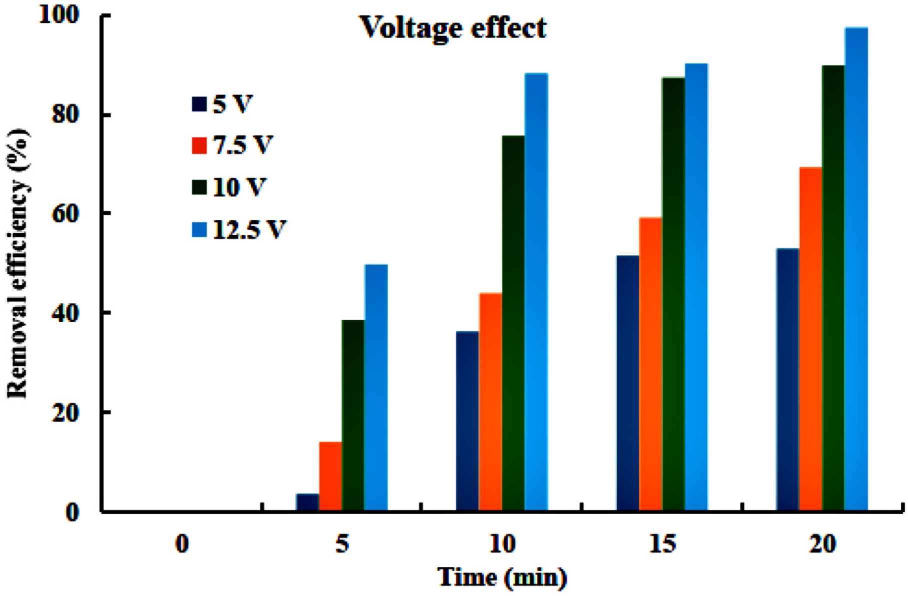

Figure 6. Removal efficiency of reactive green dye as a function of electric current applied with pH (7.5), electrode spacing of 1 cm, electrolysis time (5, 10, 15, 20 min)

treatment technologies in this balance. Voltages in the range of 10–12.5 V provide a favourable compromise to ensure high pollutant removal efficiency above 90%, with manageable energy requirements. These results confirm that the applied voltage directly and strongly influences the electrocoagulation process. With an increase in voltage, the rates of anodic aluminium dissolution, bubble generation, and pollutant destabilisation increase, leading to faster and almost complete dye removal. The removal efficiencies thus show an improving trend both with time and voltage, depicting the dynamic nature of the electrocoagulation process. Hence, there is a need for contributions to the optimisation of electrical parameters in wastewater treatment for efficiency and sustainability.

Electrode spacing

Electrode spacing is defined as the distance between the cathode and anode in an electrocoagulation cell. In this work, inter-electrode distances of 0.5, 1, 2, and 3 cm were used to analyze their effect on reactive green dye removal. Figure 7 shows that removal efficiencies were strongly affected by inter-electrode distances in an electrocoagulation cell. A stronger inverse relationship between electrode spacing and removal efficiencies means that electrode proximity has a greater influence on one of its most important operational factors in determining overall electrochemical and mass transfer efficiencies of an electrocoagulation cell. In a 0.5 to 1 cm inter-electrode distance, removal efficiencies of up to 98.85% were achieved, reaffirming that greater electrode proximity leads to increased rates of electrochemical reactions and coagulant formation, as a result of which ohmic resistances in an electrocoagulation process decrease, allowing higher current densities as well as higher rates of anodic material dissolution in electrodes. Consequently, increased reactive coagulant components such as Al^{3+} ion concentration in an aqueous solution become possible, thus allowing higher efficiencies of charge neutralization reactions with increased formation rates of flocs. An increased ionic path also enables higher magnitudes of a uniform electric field in an electrocoagulation cell, thus increasing opportunities for colliding interactions of micro-bubbles produced with suspended matter of pollutants such as reactive dyes in an aqueous medium.

When the inter-electrode distance was increased to 2 cm, removal efficiency was moderately affected, as a slight increase in the electric field strength was witnessed in the electrolyte. Even at that inter-electrode distance, removal efficiency was well acceptable, but floc agglomeration and the pollution removal rate were lower. Nonetheless, it is also considered economically favourable, as it allows a balance between electrolytic performance as well as economical aspects. Beyond 2 cm, a marked decline was seen in electrolytic performance. At a 3-cm inter-electrode

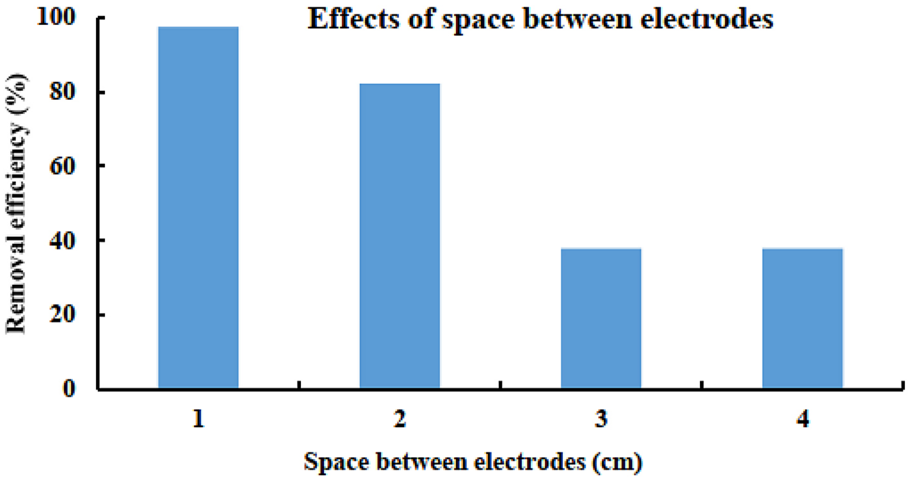

Figure 7. Removal efficiency of reactive green dye as a function of electrode spacing with pH (7.5), electrode spacing of 1 cm, electric current applied of 12.5 volts, electrolysis time (20 min)

distance, a substantial decrease in removal efficiency was seen because of higher resistance and irregular current distribution within the electrolytic cell. This also led to a decrease in metal hydroxide formation at a greater inter-electrode distance, as well as a decrease in bubble density because of hydrodynamic mixing. This eventually caused a decrease in the removal of dye molecules. Higher cell voltages were also needed because of higher cell resistance to achieve a definite current flow rate as proposed by Mousset et al. (2024).

In general, these results have reaffirmed that smaller spacing is beneficial for improving electrochemical performance as well as for enhancing removal of pollutants, whereas too large spacing is detrimental to the process because of electric and mass-related issues. In terms of practical considerations, it is important to maintain a smaller spacing of 1–2 cm in order to attain a higher removal of dye using electrocoagulation.

CONCLUSIONS

Electrocoagulation is a sustainable and innovative water treatment technology that uses electrochemical methods to remove a broad spectrum of reactive green dye from contaminated water. From the experimental results, it is clear that electrocoagulation is a highly effective reactive green dye removal technique, with several operational variables such as pH, applied electric current, electrode distance, and electrolysis time playing an important role in determining its removal efficacy. In this study, it is validated that the pH of the system is a highly important factor that determines the removal efficacy of electrocoagulation of reactive green dye. This study clearly shows that near-neutral pH conditions around 7.5 offer a highly favorable region for the formation of aluminum hydroxide, flocculation, and overall removal of pollutants. The removal efficacy of EC at near 7.5 pH conditions is around 98.82% within a time frame of 15 minutes. Under highly acidic pH conditions of 4.5, removal efficacy is moderate as a result of the dominance of aluminum ion in its aqueous form and low formation of bubbles of Hydrogen gas, thereby creating an ineffective region for floc formation. On the contrary, at highly basic pH conditions of 10.5, the formation of negatively charged aluminate anions leads to low removal efficacy of around 38%.

The electric potential applied to the system was also discovered to greatly influence electrocoagulation process performance. Higher applied voltages result in an increased rate of anodic dissolution of aluminum, enhanced formation of the active form of aluminum hydroxide, and a greater rate of hydrogen bubble generation, thereby promoting the destabilization and removal of organic dye molecules. From the experiment, it was seen that a higher removal of organic pollutants was possible at an applied electric potential of 12.5 V, although applied electric potentials ranging from 10 V to 12.5 V have been considered suitable for applications in systems for electrically assisted removal of pollutants, considering higher removal capabilities as well as higher usage of electric power.

Another important operational factor that was found to influence electrocoagulation performance is the electrode spacing. From the findings of this study, it was determined that smaller electrode spacing of 0.5 to 1 cm is most beneficial in terms of removal efficiency, while increasing the electrode spacing to 2 to 3 cm will significantly decrease removal efficiency. This is because smaller electrode spacing will lead to low ohmic current resistance with intensified current distribution, and an enhanced electric field distribution, thus contributing to easy formation of a coagulant or flocs. In conclusion, this study highlights the need for EC process variables such as pH, applied voltage, and electrode distance to be optimized to attain a high- efficiency electrocoagulation process. Using an electrocoagulation process that operates under near-neutral pH, employing a suitable voltage, and with a small electrode distance, it is possible to remove reactive green dye from aqueous solutions in wastewater treatment efficiently while remaining economical in terms of energy.

Overall, the study underscores the importance of optimizing key operational parameters pH, applied voltage, and electrode spacing to achieve high-efficiency electrocoagulation. By maintaining near-neutral pH, selecting an appropriate voltage, and ensuring narrow electrode spacing, the EC process can effectively remove reactive green dye from aqueous solutions while maintaining energy efficiency, electrode stability, and overall process reliability. These findings provide a practical framework for the design and operation of EC systems for wastewater treatment applications.

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