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Research Article

Investigation of the usability of the electrocoagulation method in malachite green removal from water solution

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ABSTRACT

In this study, the removal of Malachite Green dye in synthetically prepared aqueous solution by electrocoagulation process was investigated. In the study, initial dye concentration, electrolyte amount, mixing speed, current density, electrolysis time, pH value, and distance between electrodes parameters that affect the removal efficiency of the electrocoagulation method were investigated. As a result of the study, optimum parameters were found as initial dye concentration of 200 mg/L, electrolyte amount of 150 mg/L, stirring speed of 100 rpm, current density of 8 mA/cm^2, pH 4.5 value, the distance between electrodes 1 cm, and electrolysis time 20 min. 93.6% color removal efficiency and 37.5% COD removal were obtained under optimum conditions.

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INTRODUCTION

Negative changes in the chemical, physical, bacteriological, radioactive and ecological characteristics of the water source are defined as water pollution. Water pollution occurs as a result of the discharge of substances or energy wastes that will directly or indirectly cause preventive deterioration in biological resources, human health, fisheries, water quality and the use of water for other purposes [1].

The textile industry is one of the longest and most complex industries in the manufacturing industry. The textile industry consumes a large amount of water in its production processes. The wastewater generated as a result of the textile industry is in the toxic class. These wastewaters, which can be in different colors according to the raw material used, spoil the natural appearance of the water environment and reduce the light transmittance. The decrease in light transmittance and dissolved oxygen causes damage to aquatic organisms and prevents the use of water resources [2].

The substances used to color the materials are called colorants. These substances are mainly divided into two as dyestuff and pigment. Dyestuffs can be dissolved in the dyeing process. On the other hand, do not dissolve in the substance to which they are transported.

Malachite green (MG) is one of the basic dyes and is widely used in the dye and textile industry. MG's mixing with water and its high concentration in water cause serious problems for all living organisms, including humans, due to its toxic, carcinogenic, and mutagenic factors. In particular, it shows biological toxicities to humans and living things by consuming fish produced in Malachite green dyed or

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polluted water and by causing irritating the gastrointestinal tract. Thus, the removal of these organic dyes will directly benefit the environment and living things [3].

Many different methods are used in the treatment of dyestuffs. The toxic properties of dyestuffs and the high content of organic substances resistant to biodegradation in textile wastewater limit the applicability of biological methods [4]. In the treatment with the chemical coagulation/flocculation technique, this method cannot be preferred as a treatment alternative because the sludge formation is higher compared to other methods and because the dyestuffs are dissolved in the wastewater in the enterprises where the reactive dyeing technique is used [5]. Chemical oxidation methods are widely used in the treatment of textile wastewater and their color removal efficiency is high [5]. However, the potential for the formation of substances known to be toxic as a result of oxidation reactions is high [6].

Towards the end of the 20th century, a process called electrocoagulation was developed. This process consists of anode material, cathode material, a conductive liquid power supply components in a reactor [7].

This study, it is aimed to investigate an alternative method for the treatment of wastewater containing dyestuffs. In this context, the removal of Malachite Green dyestuff by electrocoagulation method was investigated and optimum values were determined for initial dyestuff concentration, mixing speed, pH optimization, electrolyte concentration, electrolysis time, current density, and distance between electrodes.

MATERIALS AND METHODS

Materials

Malachite green dyestuff, whose properties are given in Table 1, was obtained from Sigma Aldrich. In the study, AA TECH ADC-3303D brand power supply, Edmünd Bühler GmbH brand mixer and aluminum electrodes with 50 x 70 x 2 mm dimensions and 30 cm^2 active surface area were used. Hack DR-3900 brand spectrophotometer was used to find the dyestuff removal.

Methods

In the studies, color removal from the aqueous solution prepared in the laboratory with the dyestuff was investigated by the electrocoagulation process. The prepared 2500 mg/L stock solution was stored under cold and dark conditions and diluted with distilled water to bring it to the desired concentration.

Table 1. Properties of Malachite Green Dye

Molecule Formula	$C_{23}H_{25}ClN_2$
Molecular Weight (g/mol)	364.9
Wavelength (λ)	617 nm

The dyestuff prepared at the desired concentrations in the experiments was added to the 600 ml beakers against the risk of overflow by using 250 ml volume and put into the system. Then, the variables of dyestuff concentration, electrolyte amount, current density, pH, mixing speed, the distance between electrodes, and electrolysis time were investigated to find the optimum conditions.

Two electrodes, 1 anode and 1 cathode, were used in the monopolar connected state inside the reactor. The distance between the electrodes was adjusted to 1 cm until the optimum value was found.

In the experiments, 0.1 M HCl and 0.1 M NaOH chemicals were used to adjust the pH value. The electrodes were kept in the cleaning solution for 10 minutes before the experiments and after being washed with distilled water, they were dried and made ready for weighing and the next experiment.

During the experiment, 5 ml samples were taken from the system and the samples were subjected to centrifugation at 6000 rpm for 5 minutes and analyzed in a spectrophotometer at 617 nm.

Equations Used in Calculations

In Table 2, the equations of the calculations made as a result of this study and the explanations of the parameters applied when using these equations are given.

RESULTS AND DISCUSSION

Optimization Parameters

Initial Dye Concentration

In the electrocoagulation process working with aluminum electrodes, solutions with concentrations of 50, 100, 150, 200 and 250 mg/L were prepared and their removal efficiencies were investigated in order to determine the optimum concentration in the aqueous solution containing the dyestuff. Other parameters in the process; The amount of electrolyte was 100 mg/L, stirring speed was 100 rpm, current density was 6 mA/cm^2, the distance between the electrodes was 1 cm, the electrolysis time was 30 minutes and the original pH was used. As the concentration value increased, the removal efficiency decreased from 99.88% to 76.88%, and it was observed that the required time for removal was prolonged. At the beginning of the study, it was observed that the removal efficiency of wastewater with high concentrations was lower compared to the studies with low dye concentrations. When Figure 1 is examined, since electricity consumption is taken into account in terms of treatment cost, it is determined as 200 mg/L with less energy. Figure 2 shows the effect of initial concentration on color removal efficiency and energy consumption.

Electrolyte Amount

The pollutant removal efficiency and operating cost are related to the conductivity of the solution. The conductivity

Table 2. Equations Used in Calculations

Calculated Parameter	Equation	Parameters to use
Current Density	$J = \frac{I}{2 \times S}$	• J : Current density (mA/cm^2) • I : Current intensity (mA) • S : Electrode area
Dyestuff Removal	$Dyestuff\ removal\ (\%) = \frac{(C0 - Ct)}{C0} X100$	• C_0 : Initial dyestuff concentration • C_T : Dye concentration at time T
Mole Amount of Substance to Precipitate	$m = \frac{I\ x\ t}{n\ x\ F}$	• I : Current Intensity (Amps) • t : Time (sec) • n : Ion charge (+3 will be taken for aluminum) • F : Faraday constant (96485 $^0C\ mol^{-1}$)
Theoretical Amount of Dissolution at the Anode Electrode	$M_{ATe} = m \times (M_W)$	• M_{ATE}: Theoretical Amount of Dissolution at the Anode Electrode • M_W : Molecular weight
Current Efficiency	$E = \frac{M_A}{M_{ATe}}$	• E : Current Efficiency • M_A : The amount of dissolution in the anode electrode material in the experiment (g)
Total amount of dissolved aluminum (M_T)	$M_T = M_A + M_K$	• M_K : The amount of dissolution in the cathode electrode in practice (g)
Energy Consumption Value	$W = \frac{V \times I \times t}{v}$	• W : Energy Consumption Value ($kW.hour/m^3$) • V : Potential Difference Occurring in the System • I : Applied Current Intensity • T : time (hour) • v : Total Solution Volume in the Reactor
Energy Consumption	$Ec = V \times I \times t$	• Energy Consumption (Ec) • V: potential difference (volts) • I: Current flowing through the circuit (ampere) • t: Process time (hours)
Chemical Oxygen Demand	$COD\ (mg\ L) = \frac{(A-B)xNx8000}{Sample\ (ml)}$	• A: Iron Ammonium Sulphate consumption of the blind (mL) • B: Iron ammonium sulfate consumption of the sample (mL) • N: Normality of Iron Ammonium Sulphate Solution (N)

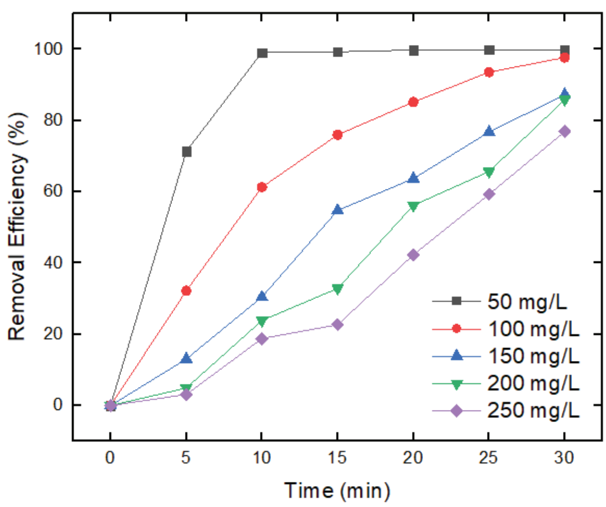

Figure 1. Results of color removal efficiency versus time with Al electrode at different initial concentrations (C_{salt}=100 mg/L, 100 rpm, J=6 mA/cm^2, original pH, Electrodes Distance=1 cm, t=30 min).

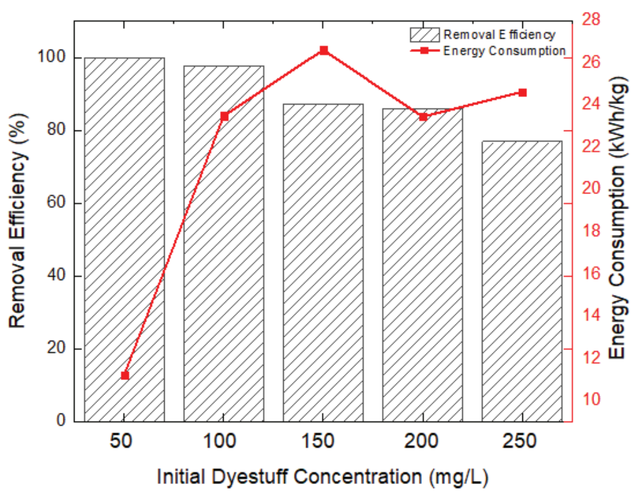

Figure 2. Effect of initial concentration on color removal efficiency and energy consumption (C_{salt}=100 mg/L, 100 rpm, J=6 mA/cm^2, original pH, Electrodes Distance=1 cm, t=30 min).

of the solution is adjusted by adding salts such as sodium chloride and sodium sulfate to the wastewater with low conductivity [8]. After the initial dyestuff concentration was determined as 200 mg/L, the amount of electrolyte was determined. In order to determine the optimum amount of electrolyte, NaCl concentrations of 50, 100, 150, 200 and 250 mg/L were added to the solution and their removal efficiencies were investigated. It was observed that as the amount of NaCl increased, the removal efficiencies increased from 69.5% to 93.5%. As the amount of added NaCl increased, the solution conductivity also increased, and accordingly, the voltage value decreased. As the voltage value decreased, the required energy amount also decreased. It was observed that the energy consumption value for kilogram pollutant amount decreased from 35.9 to 3.69 kWh/kg as the amount of electrode increased. When the kilogram pollutant amount and removal graph were examined, it was seen that the optimum NaCl amount was 150 mg/L and the removal efficiency was 88.05%. In the study, the amount of electrolyte was determined as 0.15 g/L. Higher removal was achieved with less amount of NaCl. Figure 3 shows the effect of electrolyte concentration on color removal efficiency and energy consumption.

Mixing Speed

Stirring prevents the formation of a concentration gradient from the electrocoagulation cell and increases the velocity of the ions in the cell. As the mixing speed increases, the pollutant removal efficiency increases. After the amount of electrolyte was determined, the mixing speed was determined. In order to determine the optimum mixing speed, the mixer 100, 150, 200, 250 and 300 rpm values were set on the magnetic stirrer. It was observed that the removal efficiency decreased as the mixing speed increased. Increasing

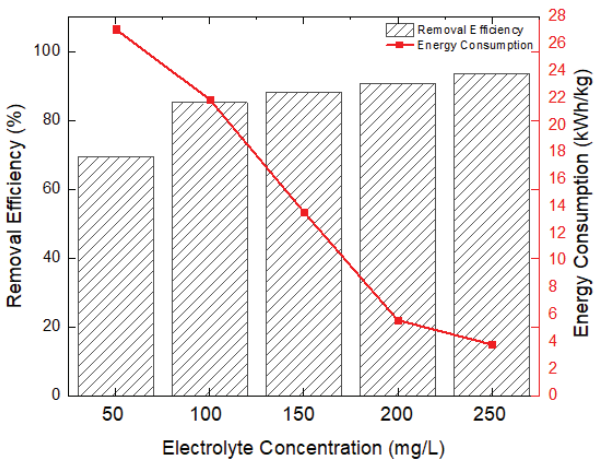

Figure 3. Effect of electrolyte concentration on color removal efficiency and energy consumption (C_0=200 mg/L, 100 rpm, J=6 mA/cm^2, original pH, Electrodes Distance =1 cm, t=30 min).

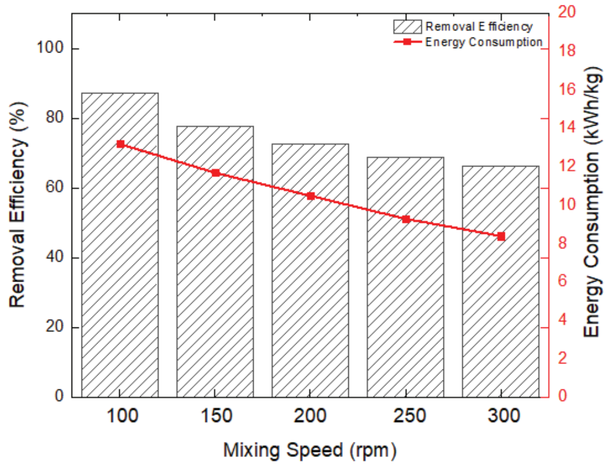

Figure 4. The effect of change in mixing speed on color removal efficiency and energy consumption (C_0=200 mg/L, C_{salt}=150 mg/L, J=6 mA/cm^2, original pH, Electrodes Distance =1 cm, t=30 min).

the mixing speed requires extra energy. In Figure 4, the effect of the change in mixing speed on the color removal efficiency and energy consumption is given. When Figure 4 is examined, it is seen that the removal efficiency decreases when the mixing speed is increased. The optimum mixing speed was determined as 100 rpm.

Current Density

Current density affects electrocoagulation efficiency, coagulation rate, bubble generation rate and size. The anodic dissolution rate increases with the increase in current density. In this way, the number of metal hydroxide clumps increases, increasing the pollutant removal efficiency [8]. It is one of the most important parameters that directly affect the removal efficiency in the electrocoagulation process. After the mixing speed was determined, the current density was determined. 2, 4, 6, 8 and 10 mA/cm^2 are set on a digital power supply to determine the optimum current density. It was observed that the removal efficiency increased as the current density increased. As the current density increases, the amount of energy required also increases. As seen in Figure 5, it has been observed that the difference between the applied current density and the removal efficiency does not increase much as it approaches the highest value. Since the removal values between 10 mA/cm^2 and 8 mA/cm^2 are close to each other, 8 mA/cm^2 was determined as the optimum current density.

pH Effect

pH is an important factor as it directly affects the reactions taking place in the electrocoagulation process. It is important both because of the formation of hydroxyl radicals at the cathode and the presence of metal hydroxides at the anode. After the current density was determined, the pH value was determined. In order to find the optimum pH

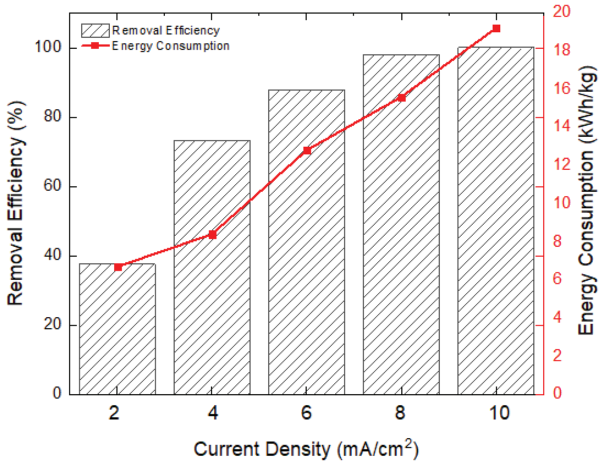

Figure 5. The effect of current density on color removal and energy consumption(C_0=200 mg/L, C_{salt}=150 mg/L, 100 rpm, original pH, Electrodes Distance =1 cm, t=30 min).

value, solutions of 2.5 -4.5 -6.5-8.5 were prepared. It was observed that the difference between pH value and removal efficiency did not increase much as it approached the highest value. At the end of 20 minutes, when the pH value was 4.5 and 6.5, the removal efficiency was found to be 98.28% and 96.95%. The optimum pH value was determined as 4.5. It has been observed that pH changes do not directly affect the processing time. The effect of pH change on color removal and energy consumption is given in Figure 6.

Distance Between Electrodes

Here, the effect of the distance between the electrodes on the removal efficiency was investigated. In order to find the most suitable distance between the electrodes, the effect on the removal efficiency was examined by opening 1-1.5-2 cm between the electrodes. It was observed that the removal efficiency decreased when the distance between the electrodes was increased. It was observed that the energy consumption increased when the distance between the electrodes was increased. When looking at the time-dependent removal graph, it was seen that the highest yield was obtained in the 1 cm range. In Figure 7, the effect of the distance between the electrodes on color removal and the energy consumption is given.

Electrolysis Time

If the electrolysis time changes, the amount of ions and hydroxide flocs produced change. In order to determine the optimum electrolysis time, the results of the previous

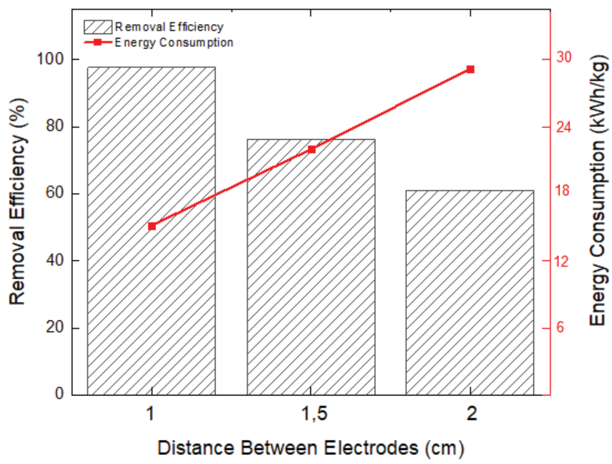

Figure 7. The effect of the distance difference between the electrodes on color removal and energy consumption (C_0=200 mg/L, C_{salt}=150 mg/L, 100 rpm, J=8 mA/cm², original pH, t=30 min).

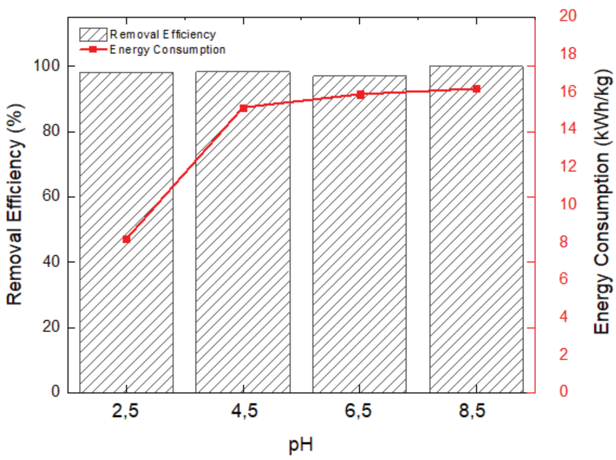

Figure 6. The effect of pH change on color removal and energy consumption (C_0=200 mg/L, C_{salt}=150 mg/L, 100 rpm, J=8 mA/cm², Electrodes Distance =1 cm, t=30 min).

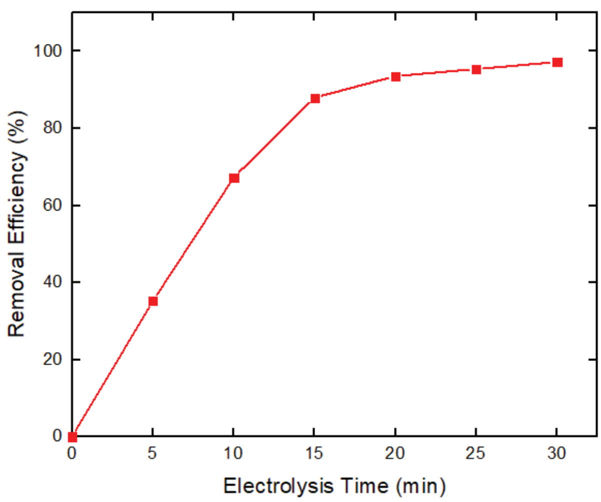

Figure 8. The effect of electrolysis time on color removal (C_0=200 mg/L, C_{salt}=150 mg/L, 100 rpm, J=8 mA/cm², original pH, Electrodes Distance =1 cm).

Table 3. Calculation results according to optimum values

Parameter	Results
Current Density (J)	8 mA/cm^2
Current Efficiency (E)	0,80
Total Dissolved Aluminum Amount (M_T)	0,0431 g
Energy Consumption (W)	10,58 kW.saat/m^3
Electricity Cost	5,22 TL/m^3
NaCI Cost	0,594 TL/m^3
Aluminum Cost	5,405 TL/m^3
Chemical Cost	0,594 TL/m^3

experiments were evaluated. When evaluated together with other optimum parameters in the process, the optimum time was found to be 20 minutes. It was observed that the removal efficiency increased as the electrolysis time increased (Figure 8). It has been observed that the removal is reduced if the electrolysis time is extended more than necessary.

Table 4. Analysis results at optimum conditions

	Initial	Final
Temperature ^{0}C	21	29
pH	4,40	6,17
Conductivity (μS)/cm	1728	614
COD (mg/L)	2561	1604
Color (Pt-Co)	3500	260

Calculations Based on Optimum Values

In Table 3, the results of the calculations made according to the optimum values of the study and in Table 4, the results of the analysis made under the optimum conditions are given.

COMPARE OTHER STUDIES

Table 5 shows what some other researchers have done before; optimum conditions of dye solution removal from aqueous solutions by electrocoagulation are given.

Tablo 5. Compare with other studies

DyeStuff	Parameters	Electrode	Dye Stuff Removal	References
Remazol Brillant Blue	Dyestuff Concentration: 100 mg/L, Current Density:1 A/m^2, Electrolysis time: 20 min	Al	%98.1	[9]
Remazol Brillant Blue	Dyestuff Concentration: 50-200 mg/L, Current Density: 0,5-5 A/m^2	Al	%99.6-98.2 %96.8-99.4	[10]
Procion Yellow H-EXL	Dyestuff Concentration: 500 mg/L, Elekcrolide concentration: 6,5 mg/L, Current Density: 7 mA/cm^2	Fe	%91.5-98.4	[8]
Metilene Blue	Dyestuff Concentration: 100 mg/L, Current Power: 20 A	Al	%94	[11]
Reaktif Yellow 160	Current Density: 100 A/m^2 Electrolysis time: 10 min Dyestuff Concentration: 100 mg/L,	Al	%96.4	[5]
Direct Red 23	Current Density: 0,1 mA/cm^2 Mixing Speed:150 rpm	Al	%98	[12]
Basic Blue	Current Density: 333 A/m^2 pH 8	Al	%97	[13]
Malachite Green	Dyestuff Concentration: 150 mg/L, Electrolysis time: 20 min pH 8	Steel	%99,5	[14]
Malachite Green	Dyestuff Concentration: 100 mg/L, Current Density: 76,5 A/m^2 pH 8 Electrolysis time: 30 min	Al-Fe	%99.9	[15]
Malachite Green	Dyestuff Concentration: 200 mg/L Current Density: 8 mA/cm^2 Electrolysis time: 20 min	Al	%94.5	This Study

CONCLUSIONS

In this study, the effects of electrocoagulation process operating parameters on color removal were investigated. Optimum conditions in the process; The initial dyestuff concentration was 200 mg/L, the electrolyte amount was 150 mg/L, the stirring speed was 100 rpm, the current density was 8 mA/cm^2, the pH was 4.5, the distance between the electrodes was 1 cm, and the electrolysis time was 20 minutes. During the studies, it was observed that color removal increased over time. It was observed that the color removal efficiency decreased when the initial dyestuff concentration was increased. It was observed that the amount of dissolution from the anode increased when the current density was increased. It was observed that the required power amount decreased when the amount of electrolyte added to increase the conductivity was increased. As a result of increasing the current too much, the water temperature increased after the paint was removed.

In this study, 93.6% removal efficiency and 37.5% COD removal efficiency were achieved after 20 minutes in the removal of malachite green dyestuff in an aqueous solution by electrocoagulation method under optimum conditions. Color measurement was made from the Pt-Co unit under optimum conditions and 92.5% removal efficiency was observed.

The necessary consumption calculated in the examination of the treatability of synthetic wastewater prepared with malachite green dyestuff was calculated according to the unit energy price of January 2021, and the energy cost to treat 1 m^3 of wastewater was found to be 11.22 TL/m^3. The electrocoagulation process has succeeded in providing a high rate of color removal in wastewater containing dyestuffs, but it has been found to be insufficient in COD removal. It should be used together with other treatment processes in wastewater with high COD value.

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CONFLICT OF INTEREST

The authors declare no potential conflicts of interest regarding the research, authorship and/or publication of this article.

DATA AVAILABILITY

The data used to support the findings of this study are included within the article.

AUTHOR'S CONTRIBUTIONS

All authors are contributed equally to bring out this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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