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Aluminum–Electrocoagulation Reactor for Removal of Brilliant Green Dye from Aqueous Solutions

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ABSTRACT

The elimination of brilliant green dye (BGD) from aqueous solutions was investigated by electrocoagulation (EC) using aluminum electrodes (Al-EC) in a batch reactor with a monopolar parallel electrode configuration. The most influential factors on BGD percentage removal include initial solution pH, electrolysis time, supporting electrolyte concentration (NaCl), current density (CD), initial concentration of BGD, and inter-electrode distance (IED). These factors were discussed to define the optimal removal conditions. The experimental results presented that the highest elimination of BGD was observed at pH values between 4 and 8, with an optimum of 93.88% at pH 4 for a CD of 47.1 A/m^2 after 60 minutes of electrolysis time with a BGD solution of 30 mg/L and 1.5 cm of IED. Additionally, the rise in CD enhanced the rate of dye removal, and the highest BGD removal of 97.85% was attained for a CD of 94.3 A/m^2 after 60 minutes of electrolysis time with a BGD solution of 30 mg/L and 1.5 cm of IED at pH 4. The BGD removal dropped as the initial BGD concentration and IED increased. It was also observed that electrical energy consumption (EEC) decreased, while solution conductivity increased as the added NaCl dose in the cell wastewater was increased from 0.1 to 0.6 g/L. The 0.5 g/L NaCl solution proved to be an ideal concentration for dye removal. After all experiments, results demonstrated that EC was effectively used, with an elimination efficacy of 93.88% under the best operational parameters: 1.5 cm of IED, 47.1 A/m^2 of CD, 60 min of electrolysis time, pH 4, and a 0.5 g/L dose of salt. After statistical analysis, it was shown that all the effects of parameters on the BGD removal efficiency were statistically significant during the EC process. Based on the experimental outcomes, it was revealed that the Al-EC can be used for effective BGD removal from aqueous solutions with high efficiency and low EEC. So, it can offer a better opportunity to apply this technique to decolourise coloured wastewater.

1. INTRODUCTION

The great concentrations of different dyes contained in coloured wastewaters are generated by various industrial applications, such as textile, food, plastic, paper, leather, and mineral processing. They are considered environmental contaminants that pollute ecosystems when discharged without treatment due to their carcinogenic effects.

There are various chemical, physical, biological, and combined methods for dye removal from wastewater. Physical methods involve adsorption, ultrasonic waves, and membrane separation; chemical methods include conventional and advanced oxidation, electrolysis, ion exchange, and coagulation; and biological techniques utilising fungi, algae, and bacteria [1]. All techniques have advantages and disadvantages. Electrocoagulation (EC) is an established and favorable technique for the elimination of various pollutants, including dyes, from polluted solutions.

The EC process is preferred as it is a simple, dependable, cost-effective system to remediate wastewater without adding chemicals that become secondary pollutants.

The EC method is based on using the electrodes dipped in contaminated solution and connected to a DC power source. The dissolution of the metal electrodes, which are generally aluminum or iron, occurs as the electrical current passes through the EC cell. The oxidation reaction occurs at the anode and generates the metal ions, but the hydrogen gas is created from the cathode during the reduction reaction [2].

At a suitable pH, different types and hydroxides of metal destabilise and agglomerate suspended solids, then remove pollutants by adsorbing dissolved forms and precipitating others. Then, the flocculated particles were removed from the solution through electroflocculation with the assistance of the H_2 gas.

EC has been successfully decolourising wastewater using EC based on iron and aluminum hydrated or hydroxides generated from the anode electrode [3]. Different studies investigated the removal of brilliant green dye (BGD) using batch reactor-EC with different modes of Al-electrode connections. This study depended on 3-pairs of Al-electrodes (3 anodes and 3 cathodes) connected in a monopolar parallel mode.

A study of the influences of a number of operational factors on BGD removal in the EC process, including NaCl concentration used as supporting electrolyte, initial pH of dye solution, CD, electrolysis time, initial BGD concentration, and inter-electrode distance (IED), has been carried out. The measurement of electrical energy consumption (EEC) was also carried out. All factors studied should be considered in large-scale plant design for EC in industrial uses.

2. ELECTROCOAGULATION PRINCIPLE AND THEORY

The produced coagulants through EC allow various mechanisms for the elimination of suspended solids and solutes in solution, using an electrical current as the basis of the EC technique. There are three major stages involved in this technique: (i) electroproduction of aluminum- or iron-dependent coagulants in situ from anodes of Fe/Al, (ii) coagulants yield to destabilise contaminants, (iii) floc formation, which are separated easily from the cell solution [4]. At repulsive forces neutralisation, the suspended contaminants arrange into larger particles that can down-precipitate or up-float, then are raised by hydrogen gas bubbles [5].

Redox reactions that convert contaminants to less toxic substances may be achieved through an EC unit [6]. Aluminum and iron are 2 characteristic types of electrodes that have been extensively used in the EC method. Through an EC approach, the flow of electric current into the dipped electrodes led to anode dissolution, producing metal ions (Al^{3+} or Fe^{2+}/Fe^{3+}) by oxidation. At the same time, the water reduction reaction occurred at the cathode, producing hydroxide ions and H_2 gas. With Al electrodes, the following reactions are presented briefly in Eq. (1), which refers to the reaction at the anode, but Eq. (3) represents the reaction occurring at the cathode, and the outcomes in the bulk solution [4].

$$Al_{(s)} \longrightarrow Al^{3+}_{(aq.)} + 3e^- \quad (1)$$

$$3H_2O + 3e^- \longrightarrow 1.5H_2(g) + 3OH_{(aq.)} \quad (2)$$

$$Al^{3+}_{(aq.)} + 3OH^-_{(aq.)} \longrightarrow Al(OH)_{3(s)} \quad (3)$$

Al^{3+} ions in aqueous solution exhibit complex equilibria with various monomeric species, depending on the solution pH [7, 8]. Numerous researchers have stated the polymerisation of the monomeric species [8-10]. Then, the key responsible for the floccules and aggregates creation in amorphous $Al(OH)_3(s)$, which is formed by complex precipitation mechanisms from the soluble monomeric and polymeric cations, is the total reaction in the bulk (Eq. (3)) [11, 12].

3. EXPERIMENTAL WORK

3.1 Chemicals

All compounds utilised in this investigation are of analytical grade and were supplied by Merck, India. Basic BGD is classified as a cationic type and categorised as ammonium, 4(diethylamino)–alpha (phenylbenzylidene), and classified as basic green 1 with C.I. of 42040; molecular formula as $C_{27}H_{34}N_2O_4S$; Molecular mass of 482.63 g/mol. After scanning, the maximum absorption (λ_{max}) is equal to 625 nm. The BGD chemical structure is displayed in Figure 1.

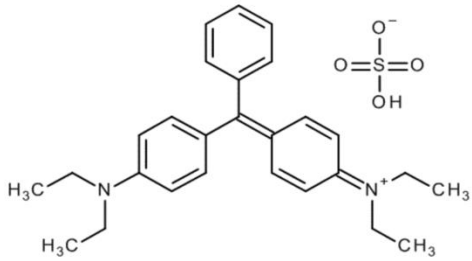

Figure 1. Brilliant green dye (BGD) molecular structure [13]

The preparation of desired concentrations of dye solutions can be done by dissolving a solid BGD in distilled water. The solution's conductivity improved with increasing NaCl concentration. The BGD solution pH was controlled using 1 M hydrochloric acid or 1 M sodium hydroxide.

3.2 Methods

The schematic of the EC reactor utilised for the EC process is shown in Figure 2. An effective wastewater volume of 2 litres was used for batch mode experiments in the EC reactor at ambient temperature (25 ± 2 °C) and made of glass (10 mm thickness) with dimensions of 20 cm × 20 cm × 15 cm. Plates of aluminum were collected from the local market and utilised to make six electrodes (3 anodes and 3 cathodes). Each plate has dimensions of 10 × 10 × 0.3 cm and is vertically immersed in a reactor, providing a 0.0318 m^2 as an effective surface area. Al-electrodes were linked in a monopolar parallel way to a supply of direct power (0-5 A and 0-30 V). The reactor content was magnetically stirred at 100 rpm during experiments using a magnetic stirrer (Daihan-Labtech Co., Ltd.). The electrodes were first washed for 15 min using dilute HCl (5% v/v) and then rinsed using distilled water before and after each experiment to remove remaining acid, and finally drying them is necessary.

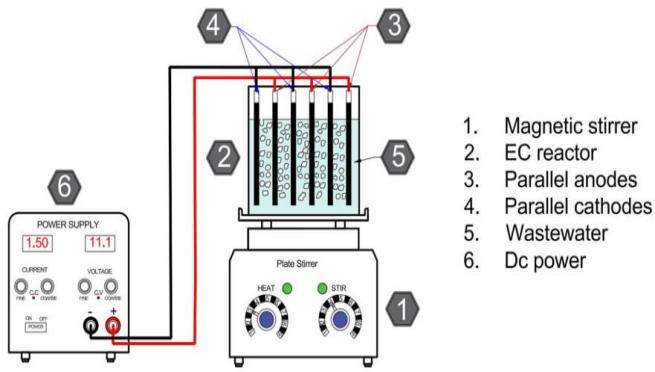

Figure 2. The schematic of the batch electrocoagulation (EC) reactor

An investigation of a good concentration of NaCl as supporting electrolyte, different concentrations with values of 0.1 - 0.6 g/L were studied, then, various factors included influence of pH (2 - 10), influence of current density (CD) ranging from 15.7 to 94.3 A/m^2, effect of variation of BGD concentration varied as 30 - 250 mg/L, and effect of inter-

electrode distance (IED) in the range of 1 - 2 cm.

All the EC experiments were conducted in triplicate, and the average values were used in data analysis.

Table 1 lists the details of the experimental parameters investigated in this study.

Table 1. Different experimental parameters studied during the Al-electrocoagulation (Al-EC) of brilliant green dye (BGD)

Parameter Studied	Parameter Range	Parameter Kept Constant During Experiment
Effect of NaCl concentration (C_S)	0.1, 0.2, 0.3, 0.4, 0.5, 0.6 g/L	C_o: 30 mg/L, CD: 47.1 A/m^2, IED: 1.5 cm, pH: 4
Effect of initial BGD solution pH	2, 3, 4, 5, 6, 7, 8, 9, 10	C_S: 0.5 g/L, C_o: 30 mg/L, CD: 47.1 A/m^2, IED: 1.5 cm
Effect of BGD initial concentration (C_o)	30, 70, 100, 125, 150, 200, 250 mg/L	C_S: 0.5 g/L, pH: 4, CD: 47.1 A/m^2, IED: 1.5 cm
Effect of current density (CD)	15.7, 31.4, 47.1, 62.8, 78.6, 94.3 A/m^2	C_S: 0.5 g/L, pH: 4, C_o: 30 mg/L, IED: 1.5 cm
Effect of the inter-electrode distance (IED)	1, 1.5, 2 cm for every CD of 47.1, 62.8, 94.3 A/m^2	C_S: 0.5 g/L, pH: 4, C_o: 30 mg/L, CD: 47.1 A/m^2
Electrolysis time (T)	10 - 60 minutes for all the experiments	

The samples are pipetted from the middle of the supernatant portion every 5 minutes during each experiment with a detention time of 20 min and then filtered using a 0.42 µm pore-sized filter paper (Whatman) before test using a UV Spectrophotometer (UV-VIS-6800 JENWAY) with λ_{max} = 625 nm using distilled water as blank solution and based on dilution factor for residual dye concentration greater than the upper value of calibration range.

The BGD removal efficiency (R%) was determined according to Eq. (4) [4]:

$$R\ (\%) = \frac{C_o - C_e}{C_o} * 100\% \quad (4)$$

The C_o and C_e represent the BGD concentrations at the primary and final stages of each experiment (mg/L).

The electrical energy consumption (EEC, kWh/m^3) was determined depending on Eq. (5) [14]:

$$EEC = \frac{IVt}{Vol} \quad (5)$$

where, I refers to the current (A), V represents the voltage (V), t denotes the time of EC operation (h), and Vol represents the volume of BGD solution (m^3).

Descriptive statistics were used to describe all the experimental data obtained from different experiments.

Statistical analysis was performed using SPSS spatial analysis of variance (ANOVA) through regression analysis to show the significance of all parameters and their effects on BGD removal efficiency.

3.3 Statistical data analysis

The descriptive statistics of experimental removal efficiencies for all parameters studied are listed in Table 2 as mean ± SD for different times.

Table 2. The experimental data of brilliant green dye (BGD) removal efficiency (mean ± SD%) for different experimental parameters studied during the Al-electrocoagulation (Al-EC) at different times

Parameter	Time (min.)		
	10	30	60
Initial BGD solution pH			
pH2	56.23 ± 16	62.58 ± 17	68.55 ± 17
pH3	66.32 ± 16	72.45 ± 17	78.45 ± 17
pH4	86.08 ± 3	89.86 ± 59	93.89 ± 1
pH5	83.64 ± 17	87.52 ± 18	92.12 ± 19
pH6	80.22 ± 15	84.92 ± 17	90.14 ± 17
pH7	78.50 ± 16	82.86 ± 17	87.92 ± 15
pH8	76.60 ± 16	80.30 ± 17	85.40 ± 17
pH9	72.13 ± 16	77.52 ± 16	82.52 ± 17
pH10	69.42 ± 16	75.12 ± 16	80.45 ± 17
NaCl concentration (C_S, g/L)			
0.1	65.69 ± 17	70.32 ± 18	78.58 ± 18
0.2	70.24 ± 18	74.67 ± 17	83.66 ± 18
0.3	78.97 ± 15	83.68 ± 18	88.45 ± 17
0.4	81.44 ± 17	85.78 ± 19	91.26 ± 16
0.5	84.64 ± 18	88.91 ± 17	93.89 ± 18
0.6	82.34 ± 16	87.16 ± 17	92.23 ± 19
Initial BGD concentration (C_o, mg/L)			
30	80.44 ± 18	86.26 ± 18	93.88 ± 18
70	75.23 ± 18	83.57 ± 17	91.33 ± 18
100	72.45 ± 15	80.53 ± 18	89.22 ±17
125	65.33 ± 17	76.56 ± 18	86.12 ± 16
150	55.23 ± 18	68.33 ± 17	80.23 ± 18
200	50.55 ± 16	58.67 ± 17	72.14 ± 19
250	45.16 ± 17	50.23 ±17	60.49 ± 16
Current density (CD, A/m^2)			
15.7	65.33 ± 15	70.69 ± 14	78.54 ± 16
31.4	78.24 ± 18	84.24 ± 17	89.96 ± 19
47.1	81.65 ± 15	86.69 ± 18	93.20 ± 17
62.8	82.15 ± 17	88.87 ± 18	94.96 ± 16
78.6	84.25 ± 18	91.12 ± 17	96.11 ± 18
94.3	86.54 ± 16	93.22 ± 17	97.85 ± 19
Inter-distance between electrodes (IED, cm)			
1	84.13 ± 16	90.95 ± 18	96.33 ± 17
1.5	81.67 ± 17	86.67 ± 17	93.89 ± 18
2	74.53 ± 17	79.46 ± 18	85.34 ± 17

The statistical analysis depended on all the experimental removal efficiencies at all times from 10 to 60 min to show the significance tests using ANOVA through multiple linear regression analysis to determine whether the effects of parameters are statistically significant by fitting them to a mathematical model.

Table 3. The analysis of variance (ANOVA)

Model	Sum of Squares	df	Mean Square	F	Sig. (p)
Regression	26236.227	6	4372.704	125.570	0.000
Residual	11630.817	334	34.823		
Total	37867.004	340			

Through Table 3, a high F-value and a low p-value (typically $p < 0.05$) indicate that the regression model is statistically significant to represent the effects of parameters studied during the EC process. Also, the contribution (B) and the effect of each parameter are listed in Table 4. The t-test was also used to test the significance of regression coefficients. A significant p-value (e.g., $p < 0.05$) suggests that the effect of each parameter is significant and contributes to

predicting the BGD removal efficiency. The multiple linear regression model obtained with $R^2 = 0.70$ is:

$$\text{BGD removal efficiency} = 74.388 - 0.126\ C_o + 0.266\ T + 0.234\ CD + 23.44\ C_s - 10.902\ IED - 0.433\ pH$$

Table 4. The regression coefficients entered the analysis

Model	Unstandardized Coefficients		Standardized Coefficients	t	Sig
	B	Std. Error	Beta		
Constant	74.388	4.416		16.846	0.000
C_o	-0.126	0.006	-0.648	-20.854	0.000
T	0.266	0.020	0.399	13.173	0.000
CD	0.234	0.026	0.271	8.890	0.000
C_s	23.440	3.400	0.213	6.894	0.000
IED	-10.902	2.516	-0.131	-4.332	0.000
pH	-0.433	0.196	-0.068	-2.202	0.028

Notes: C_o: Initial BGD concentration, T: Electrolysis time, CD: Current density, C_s: NaCl concentration, IED: Inter-electrode distance.

4. RESULTS AND DISCUSSION

4.1 Influence of NaCl concentration

Under specific conditions of the EC process, contaminant removal will be based on the coagulant generation rate, which depends on the conductivity of the cell solution. So, the conductivity, which is important for current efficiency, EEC in the electric cell, and cell voltage drop, is determined by the system resistance and its influence on the electrodes and bulk wastewater. So, the addition of NaCl reduces energy consumption by increasing solution conductivity, thereby reducing ohmic resistance [15, 16]. Table 5 displays the obtained results of using different concentrations of NaCl to optimise the dose in an electrical cell.

From the table, it is clear that the rise in salt dose value (0.1 - 0.6 g/L) at constant CD of 47.1 A/m^2, IED of 1.5 cm, 30 mg/L as initial concentration of BGD, pH 4 with 60 min as an electrolysis time, enhances the media conductivity (0.33 - 4.38 mS/cm). This enhancement is due to the decline of voltage through the cell from 18.1 to 6.2 V, which then drops the EEC value (13.57 – 4.65 kWh/m^3) as shown in Figure 3.

After 60 min, the BGD percentage removal increased from 78.58% to a maximum of 93.88% with dropping of EEC from 13.57 to 5.32 kWh/m^3 as the NaCl dose varied from 0.1 to 0.5 g/L.

An increase in dye removal due to conductivity improvement when NaCl quantity increased from 0.1 to 0.5 g per liter, and provided anti-passive ions of (Cl^-) that can destroy the passive oxide region produced on the anode surface, which leads to enhancing the dissolution rate of Al-anodes. The Cl^- ions could decrease the negative influence of other anions like SO_4^{2-} and HCO^{3-} present in the solution when these ions precipitate the Ca^{2+} that is naturally found in all the wastewater types [9]. Then, the electrode surface would be insulated by the precipitate layer, increasing the ohmic resistance across the cell [17-19]. But after 0.6 g/L of NaCl was added, the dye removal dropped to 92.23%. This means that an increase in cell wastewater conductivity did not significantly affect dye elimination. However, the excessive Cl^- ions cause a reduction in BGD removal, which may be due to irregular aluminum dissolution, which is due to overconsumption of the Al electrode through the oxidation process, acceleration of the anode, and augmentation of the corrosion pitting rate, and excessive Al electrode consumption [20]. The balance between removal rate and energy consumption is a criterion for selecting 0.5 g/L. Therefore, the amount of supporting electrolyte in the solution should be controlled. Hence, in all the following experiments, the NaCl dose is 0.5 g/L for effective dye elimination.

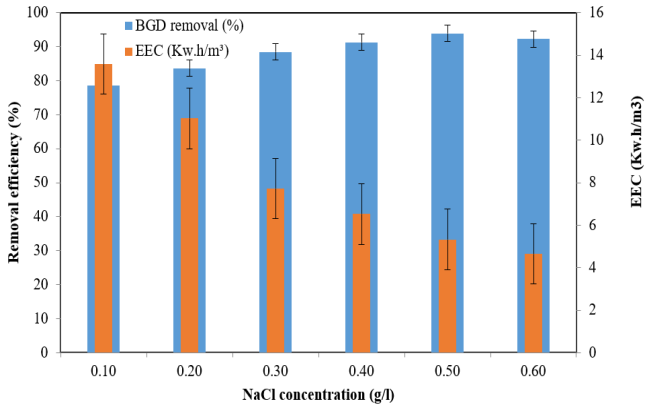

Figure 3. Influence of NaCl concentration on EEC and BGD removal

Notes: BGD: Brilliant green dye, EEC: Electrical energy consumption.

Table 5. Alteration of wastewater conductivity, BGD reduction, drop of voltage, and EEC under different concentrations of NaCl

NaCl Concentration (g/L)	Solution Conductivity (mS/cm)	BGD Elimination (%) at 60 min ± SD	Voltage Drop (V)	EEC (kWh/m^3) ± SD
0.1	0.33	78.58 ± 0.18	18.1	13.57 ± 0.11
0.2	0.82	83.65 ± 0.17	14.7	11.02 ± 0.11
0.3	1.55	88.45 ± 0.18	10.3	7.72 ± 0.12
0.4	2.40	91.26 ± 0.17	8.7	6.52 ± 0.12
0.5	3.12	93.88 ± 0.16	7.1	5.32 ± 0.14
0.6	4.38	92.23 ± 0.17	6.2	4.65 ± 0.13

Notes: BGD: Brilliant green dye, EEC: Electrical energy consumption.

Although the maximum removal efficiency (93.88%) was attained at 0.5 g/L NaCl, a higher concentration of NaCl (0.6 g/L) decreased the removal efficiency by only 1.65%, but reduced the EEC by about 12.6%. So, from an engineering perspective, the 0.6 g/L may also be considered good-looking when reducing operational energy cost is prioritized over attaining the maximum possible dye elimination efficiency. The choice of 0.5 g/L in this work was established to maximize pollutant removal, whereas the choice of 0.6 g/L could be economically better for large-scale industrial applications where EEC represents a main operating cost. So, the optimal operating state depends on the design objective. If maximum removal of dye is required, 0.5 g/L NaCl is suggested. However, if reducing operating cost is the priority, 0.6 g/L NaCl may represent a more economically favorable operating state.

Figure 4 displays the dye removal as a function of electrolysis time at various NaCl concentrations.

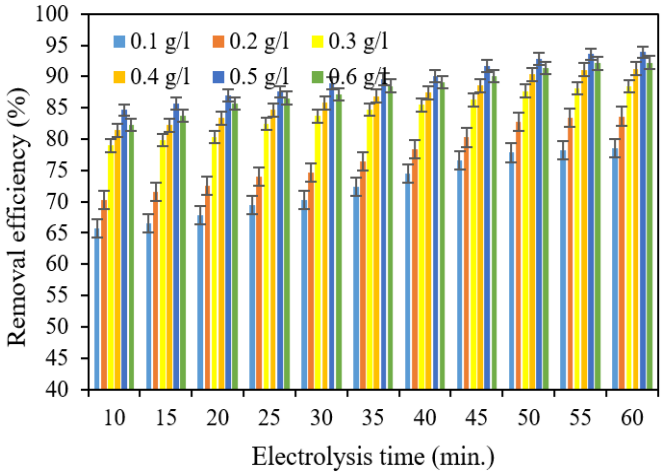

Figure 4. Effect of NaCl concentration on BGD removal as a function of electrolysis time (pH 4, CD = 47.1 A/m², C_o = 30 mg/L, IED = 1.5 cm)

Notes: BGD: Brilliant green dye, CD: Current density, C_o: Initial BGD concentration, IED: Inter-electrode distance.

4.2 Effect of initial brilliant green dye solution pH

Solution pH is considered a key operational factor that influences the degradation reaction, as it determines the chemistry of both the coagulants (solubility of the electrode and speciation of metal), dye molecules, and EC process efficiency in the aqueous media [8, 21, 22]. When investigations using aluminum electrodes, monomeric and polymeric aluminum hydroxide complexes are formed, and the coagulation mechanism depends on the solution pH. So, the main chemical species are also different according to the initial solution pH [23, 24]. For that reason, several experiments with BGD solutions were adjusted to the desired values of initial pH with a range of 2-10 to explain the effect of the BGD solution pH on the EC performance. Figure 5 displays the variation in the adjusted initial and final pH during the experiments, along with the percentage removal of BGD after 60 min of reactive time.

It was reported that the solution pH increases during the reaction time, depending on the adjusted initial pH. Similar behavior has been reported in a previous study [25], and these changes also depend on the electrode type. The initial pH value increases during the EC process, which is due to water electrolysis, then releases the hydrogen and generates hydroxide ions at the cathode electrode. After that, the relative stability of pH probability is a result of the form of the flocs of insoluble $Al(OH)_3$ and the rest of the metal hydroxides. This fact is consistent with the previous study [26]. The influence of the initial value of pH from 2 to 10 on the removal efficacy of BGD with reactive time is shown in Figure 6. According to the figure, the initial pH value has a significant influence on BGD reduction.

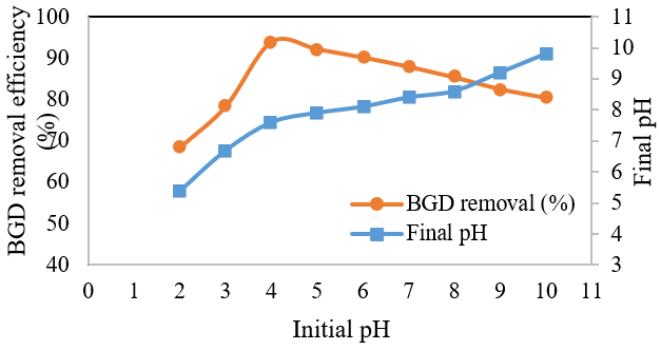

Figure 5. Effect of initial pH on BGD removal (%) (CD = 47.1 A/m², C_o = 30 mg/L, IED = 1.5 cm, electrolysis time = 60 min)

Notes: BGD: Brilliant green dye, CD: Current density, C_o: Initial BGD concentration, IED: Inter-electrode distance.

The BGD removal after 60 min was low at low pH, due to the amphoteric behavior of metal hydroxides, which tend to produce monomeric, soluble cations [27].

According to Figure 5, the experimental outcomes explained that there was maximum elimination of colour as the adjusted initial dye solution pH was in the range of 4 to 8. The optimal elimination was 93.88% at pH 4. An additional increase in pH to 9 and 10 resulted in a drop of BGD removal efficiency to 82.52% and 80.45%, respectively. It should be noted that the optimal value of pH = 4 described in this work refers to the initial dye solution pH before EC started. Throughout electrolysis, OH- ions generated at the cathode with time increased the dye solution pH to about 8, as shown in Figure 5. Consequently, although the process was started under acidic conditions, the final contaminant removal likely happened under near-neutral to mildly alkaline conditions, where amorphous $Al(OH)_3$ flocs stayed abundant and effective for contaminant adsorption. So, the superior act observed at an initial pH of 4 must be interpreted as the result of both the favorable initial Al dissolution and the following increasing of dye solution pH during EC [28]. An initial dye solution pH of 4 provided the most favorable operating state for starting the EC process. During electrolysis, the dye solution pH augmented toward alkaline values, where aluminum hydroxide flocs continued to serve dye removal. Aluminum species and floc characteristics were not measured in this study, so the results may be associated with the formation of the main Al species ($Al(OH)_3$) (s) precipitates (with polymerised classes) and changes in aluminum speciation, as reported in previous studies, which have a long surface area utilised for fast adsorption of soluble organic pollutants. So that, at solution pH greater than 9, the predominant species is $Al(OH)_4^-$, which is not useful for coagulating contaminants [29-31].

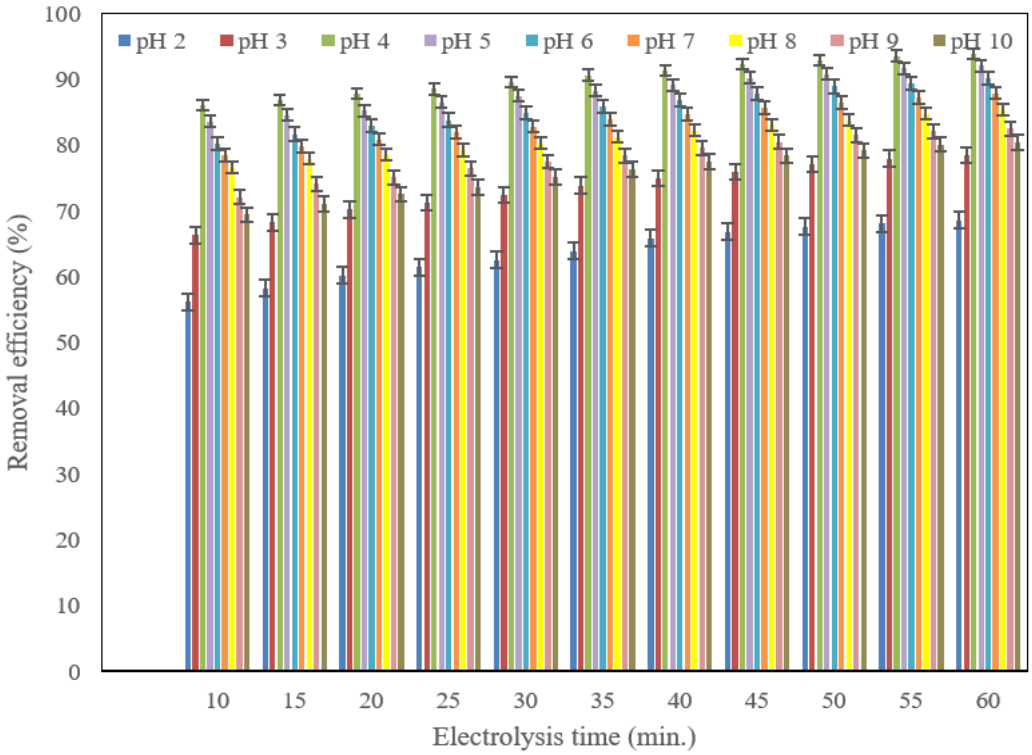

Figure 6. Effect of initial pH on BGD removal after 60 min of electrolysis (CD = 47.1 A/m², C_o = 30 mg/L, IED = 1.5 cm)
Notes: BGD: Brilliant green dye, CD: Current density, C_o: Initial BGD concentration, IED: Inter-electrode distance.

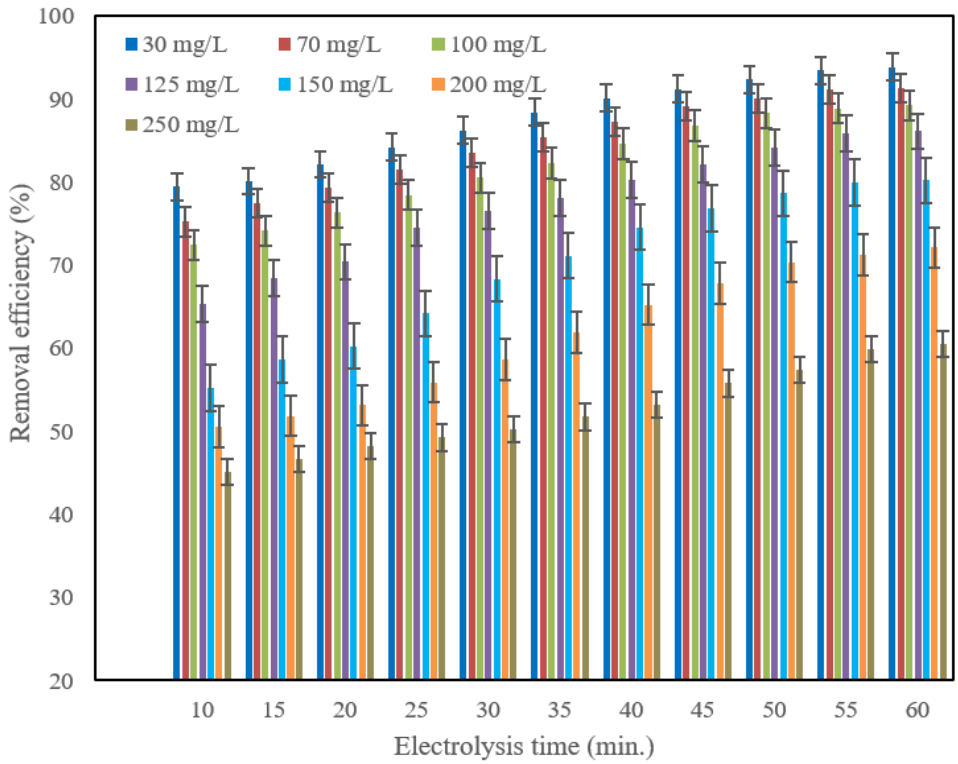

Figure 7. Effect of initial BGD concentration on removal efficiency after 60 min of electrolysis (CD = 47.1 A/m², pH = 4, IED = 1.5 cm)
Notes: BGD: Brilliant green dye, CD: Current density, IED: Inter-electrode distance.

4.3 Brilliant green dye initial concentration effect

To investigate the impact of BGD concentration on EC elimination, six dye solutions with different initial concentrations (30-250 mg/L) were treated at a constant CD and an optimum pH for 60 min. Figure 7 displays the percentage elimination of BGD for a variant initial BGD concentration. Subsequently, results showed that the

efficiency of dye elimination after 60 min of reaction decreased from 93.88% to 60.48% as the initial BGD concentration increased from 30 to 250 mg/L. This is attributed to the fact that at a fixed time and CD, the same quantity of aluminum hydroxide and consequently the same flocs were produced in all of the BGD solutions, agreeing with Faraday's law for passing the amount of aluminum ions to the solution. So, the generated flocs at high BGD concentration were insufficient for the adsorption of all BGD molecules in the solution. Similar observations were listed by the previous investigations [32-34].

4.4 Influence of applied current density and time of electrolysis

The current density and electrolysis time are the two important operational factors that should be considered for an electrochemical process to control the reaction rate in the reactor. So, these factors affect the lifetime of the performance electrode and the EC process by determining the rate and total amount of coagulant production, the size of gas bubbles, the formation rate, and the growth of flocs.

The variation in BGD percentage elimination with electrolysis time of 10 to 60 min at various CDs of 15.7 to 94.3 A/m^2, while keeping other factors constant, is shown in Figure 8. From the figure, it can be shown that the BGD reduction rate improved from 65.34% to 86.55% with a rise in CD from 15.7 to 94.3 A/m^2 at 10 min, but the BGD reduction rate improved from 78.55% to 97.85% at 60 min.

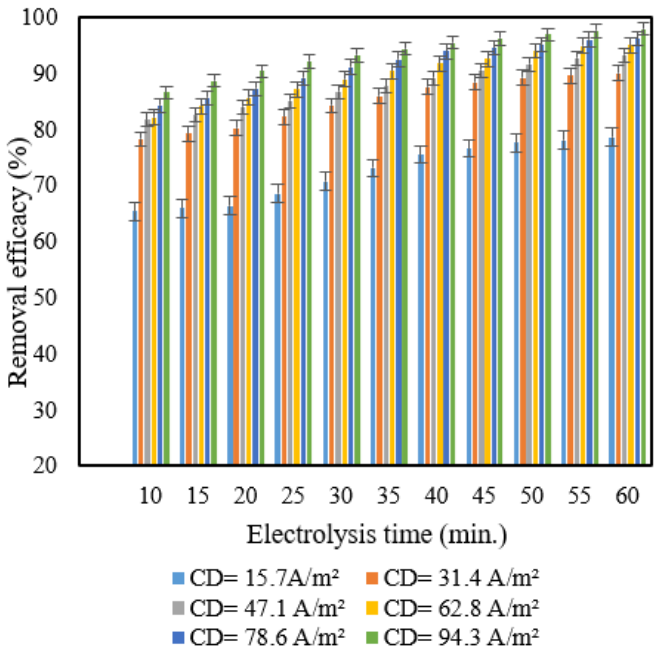

Figure 8. Time profiles of BGD removal at different CDs (pH = 4, C_o = 30 mg/L, IED = 1.5 cm)
Notes: BGD: Brilliant green dye, CD: Current density, C_o: Initial BGD concentration, IED: Inter-electrode distance.

After 60 min, it is clear that there is a fast increase in BGD removal rate up to a CD of 94.3 A/m^2. The augmentation in the quantity of metal ions and their hydroxide flocs produced in the cell solution by oxidation at the anode electrode, based on the current and time, which is determined by means of Faraday's law (Eq. (6)):

$$m = \frac{ItM}{zF} \quad (6)$$

where, m is the amount of dissolved metal (g), I is the current (A), t is the electrolysis time (s), M is the molecular weight of Al, z is the number of electrons involved in the redox reaction (3), and F is Faraday's constant (96500 C/mol) [26, 35].

In addition, the generation rate of the bubbles (O_2 and H_2) at both electrodes increases with increasing CD, which is useful for enhancing the mixing of Al^{+3} hydroxide and dye molecules by separating the flocs via flotation [28, 36].

In this study, the theoretical and actual dissolution of aluminum, sludge generation, and residual aluminum concentration were not assessed.

There was no significant increase in the dye removal rate at CD greater than 62.8 A/m^2. Nevertheless, after 60 min of EC procedure, more than 89.95% BGD reduction was observed at all applied current densities, except 15.7 A/m^2. Figure 7 also illustrates that there is a direct relation between CD and reactive time as the BGD percentage reduction increased up to 78.55, 89.95, 93.88, 94.96, 96.1, and 97.85% as current density was 15.7, 31.4, 47.1, 62.8, 78.6, and 94.3 A/m^2, respectively, when electrolysis time increased from 10 to 60 min.

This can be attributed to the rise in floc generation rate, driven by increased anode dissolution over time, and hence an increase in dye removal efficiency [2, 26, 37]. Also, a rise in reactive time yielded an extended contact time between contaminants and coagulant agents, which then augmented the reduction efficiency [38].

At the same time, BGD removal increased with rising current density; the energy consumption of the EC process also increased. As the CD increased from 15.7 to 94.3 A/m^2, the potential difference between the electrodes augmented from 2.7 to 10.32 V, and the energy consumption augmented from 0.675 to 15.48 kWh/m^3, as shown in Figure 9. Using higher current leads to electrode passivation and accelerates polarisation, increasing EEC [39]. So, it is necessary to balance between the removal efficiency, electrode consumption, and EEC. Other previous studies agree with the results obtained [40].

According to the results reported, a CD of 47.1 A/m^2 and 60 min of electrolysis time are used as the best parameters for the following experiments.

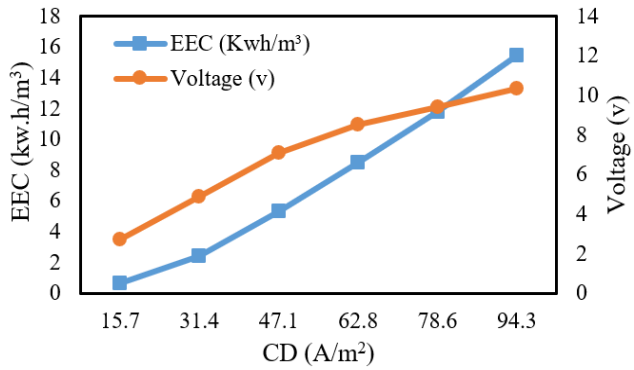

Figure 9. Effect of current density on BGD removal, cell voltage, and EEC during EC (pH = 4, electrolysis time = 60 min, C_o = 30 mg/L, IED = 1.5 cm)
Notes: BGD: Brilliant green dye, CD: Current density, EEC: Electrical energy consumption; EC: Electrocoagulation, C_o: Initial BGD concentration, IED: Inter-electrode distance.

4.5 Influence of the inter-electrode distance

The arrangement of the electrode assembly relative to the required electrode effective surface area and IED is very significant for the proper functioning of an EC cell. So, many studies evaluated the EC process as a function of IED and its effect on the percentage of pollutant removal, which depends on the nature of the pollutant, the electrode setup, the hydrodynamic conditions of the process, etc. [41]. Voltage drop increases with increasing IED at a fixed anodic surface area and cell solution conductivity. Between two electrodes, the resistance rises as the gap between them widens, so the electrical current declines [42]. Therefore, to achieve a specific CD, the voltage needs to be increased. As a result, the increase in IED leads to a drop in current, lower generation of aluminum and hydroxyl ions, reduced interaction between hydroxide ions and polymers, and finally reduced dye reduction efficiency [43].

To study the impact of this parameter on percentage dye removal and EEC, different IEDs ranging from 1 to 2 cm are studied at CD of 47.1, 62.8, and 94.3 A/m^2, with other parameters held constant, as displayed in Figure 10.

Figure 10 shows that, after 60 min, BGD removal at CD = 47.1, 62.8, and 94.3 A/m^3 was 96.33%, 98.25%, and 99.2% for an IED of 1.0 cm; 93.88%, 94.96%, and 97.85% for an IED of 1.5 cm; and 85.33%, 88.65%, and 90.45% for an IED of 2.0 cm. The highest removal was obtained at the shortest IED. A decrease in percentage BGD elimination with a rise in IED was attributed to the increase in the time of the ions' transportation, and then a weaker electrostatic attraction at a longer distance, and interaction between the produced flocs and BGD molecules was weak [44, 45]. On the other hand, at longer IED, the lower coagulant formation is attributed to a decrease in the generation rate of poly-hydroxyl complexes from the reaction between Al^{+3} formed at the anode and hydroxide produced at the cathode [4, 46].

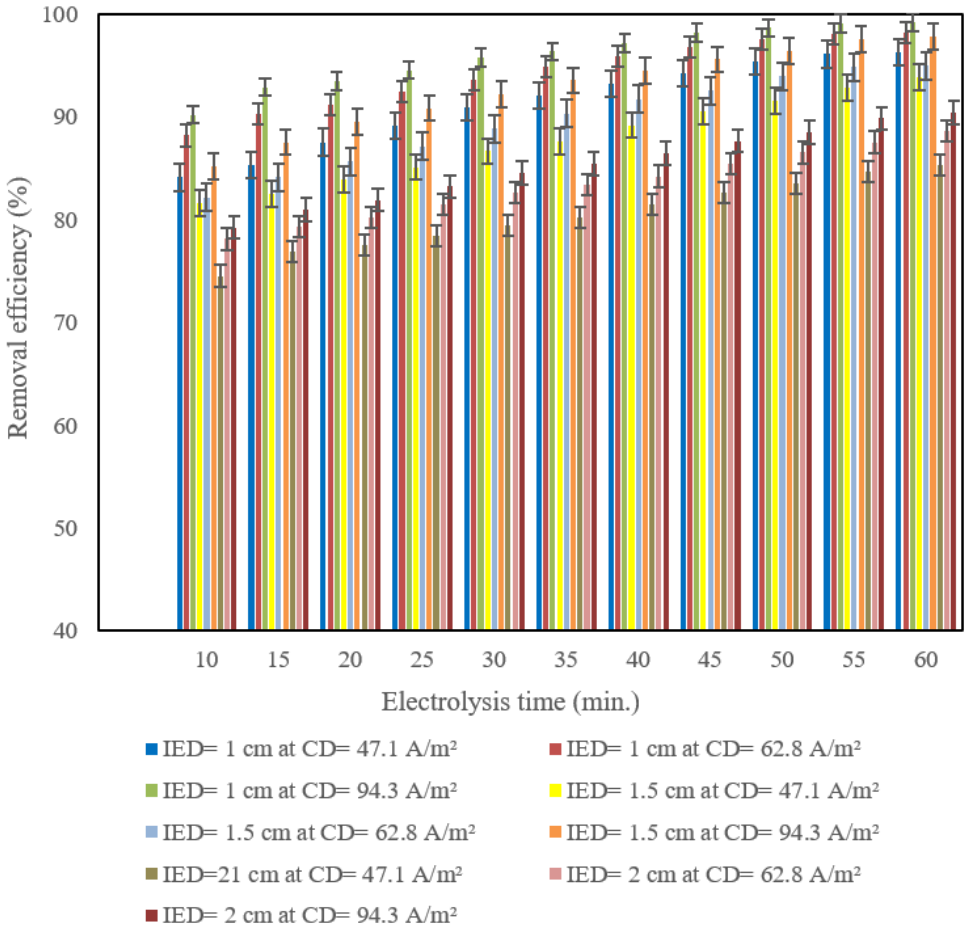

Figure 10. Effect of IED on BGD removal at different current densities (pH = 4, C_o = 30 mg/L, electrolysis time = 60 min)
Notes: BGD: Brilliant green dye, CD: Current density, C_o: Initial BGD concentration, IED: Inter-electrode distance.

A very short IED should not be used as it reduces the precipitation of sludge by increasing the rate of collision among the produced flocs, which leads to their degradation; short gaps also inhibit the extraction of air bubbles collected in the EC unit, which has a negative influence on the depletion of energy [1, 46]. So, the distance of 1.5 cm will be chosen for the other experiments to achieve a balance between removal rate and energy consumption.

Figure 11 explains the effect of IED on the EEC. It was noticed that EEC increased as IED increased because a higher cell voltage was required, and it also increased with CD. Therefore, more energy was consumed to sustain the imposed current density. A similar result was observed in another study [41].

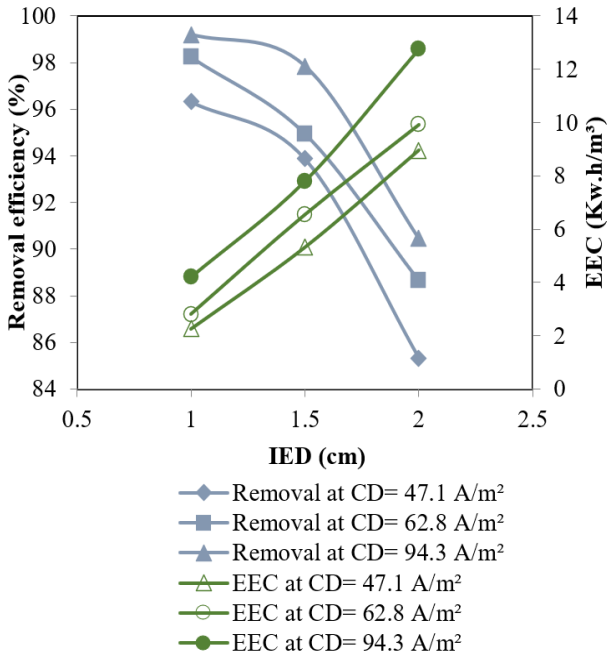

Figure 11. Effect of IED on BGD removal and EEC at different CDs (pH = 4, C_o = 30 mg/L, electrolysis time = 60 min)

Notes: IED: Inter-electrode distance, BGD: Brilliant green dye, EEC: electrical energy consumption, CD: Current density, C_o: Initial BGD concentration.

5. CONCLUSIONS

In the present investigation, the EC technique with 3 pairs of aluminum electrodes was used to remove the synthetic BGD solution in a batch reactor with a monopolar parallel connection. The influence of different operational factors on the EC process was investigated with electrolysis time and optimized, including NaCl concentration as the supporting electrolyte, initial pH of the dye solution, reactive time, CD, initial dye concentration, and IED. The results show that the percentage reduction in dye was increased as the NaCl concentration, electric current density, and electrolysis time increased. However, this decrease was less pronounced as the initial BGD concentration and the IED were increased. At the low and high values of solution pH, low dye removal efficiency was observed, but the maximum dye removal occurred in the pH range of 4-8. It also showed that the solution conductivity significantly affected EEC and dye removal. The solution's conductivity increased, and energy consumption decreased as the NaCl dose increased. The results showed that the EC has been effectively used to remove the dye from aqueous solutions under optimal operating conditions: concentration of NaCl = 0.5 g/L, pH 4, CD = 47.1 A/m^2, 60 min of electrolysis time, and IED = 1.5 cm. For future work, it is required to apply real wastewater and sludge handling with tests of chemical oxygen demand (COD), total organic carbon (TOC), and residual aluminum. The theoretical and actual dissolution of aluminum, sludge generation, and residual aluminum concentration need to be studied in future work.

For statistical analysis, required significance tests were conducted and showed that all the effects of parameters on the BGD removal efficiency are statistically significant.

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NOMENCLATURE

EC	Electrocoagulation
R	Removal efficiency (%)
BGD	Brilliant green dye
CD	Current density (A/m^2)
IED	Inter-electrode distance (cm)
EEC	Electrical energy consumption (kWh/m^3)

Greek symbols

λ_{max}	Dye maximum absorption (nm)