



Optimizing Sustainable Wastewater Treatment: Kinetic and Parametric Study of Methylene Blue Removal Using Electrocoagulation

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Abstract: This study investigates the removal of Methylene Blue (MB) from synthetic wastewater using a batch electrocoagulation (EC) reactor with two pairs of parallel monopolar iron electrodes, directly supporting Sustainable Development Goal 6 (Clean Water and Sanitation). Operational parameters, including initial pH, inter-electrode distance, applied voltage, and electrolysis time, were systematically evaluated. Within 15 minutes of electrolysis, a color removal efficiency of over 80% was achieved at an applied voltage of 10 V and an inter-electrode distance of 5 mm. Treatment efficiency decreased to 60.13% when the inter-electrode distance was widened to 10 mm due to increased cell resistance. Conversely, increasing the voltage from 10 V to 20 V and 30 V significantly enhanced the process efficiency. The MB removal rate was validated using a mass transfer-based kinetic model solved via the 4th-order Runge-Kutta method. The model aligned well with experimental data, displaying a Sum of Squared Errors (SSE) range of 0.09 to 193.57. At an extended electrolysis time of 75 minutes, the average efficiency reached 92.04% across all voltage variations, proving that coagulant saturation overcomes mass transfer limitations.

Keywords: Color removal efficiency; Electrocoagulation; Mass transfer kinetics; Methylene blue; Synthetic wastewater

Introduction

Methylene blue (MB) is a basic aromatic cationic dye that is highly soluble in water, producing a deep blue solution. Due to its unique physicochemical properties, this pigment is widely utilized across various commercial sectors, including textile manufacturing, paper processing, cosmetics, and the food industry as food additives (Balarak et al., 2020; Derakhshan et al., 2013; Rahimian & Zarinabadi, 2020).

Consequently, liquid discharges from production facilities utilizing this compound serve as a primary contributor to hazardous pollution in industrial waste streams (Alizadeh et al., 2015). Approximately 20–50% of reactive dyes are lost during industrial application and subsequently discharged directly into natural water

bodies (Mahmoud et al., 2013). Because of its easy nature dissolves, MB forms a highly colored solution that can obstruct transmission ray sun to in water. This matter result solubility oxygen become low, and in turn lower diversity and aesthetics waters (Zhou et al. 2019). Beside it was also reported, that material MB dye is toxic, carcinogenic, and difficult biodegradable (Khan et al., 2022). Therefore, mitigating its presence in water media is critical to safeguarding public health and ensuring environmental sustainability.

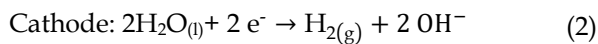
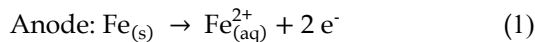
Various conventional engineering methods for removing MB from contaminated aqueous solutions have been documented, including physical adsorption (Derakhshan et al., 2013), coagulation (Jorge et al., 2022), biodegradation (Contreras et al., 2019), ultrafiltration (Fradj et al., 2014), photocatalytic (Abdrabou et al. 2023).

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However, each treatment technique possesses specific functional limitations, particularly regarding prohibitive operating costs, operational complexity, slow kinetic pathways, or the secondary generation of toxic chemical sludge (Khan et al., 2022). To address these drawbacks, electrocoagulation (EC) has been suggested as an especially effective, economically viable, and sustainable alternative treatment method for removing pollutants in raw water and wastewater (Mahmoud et al., 2013). Electrocoagulation is highly favored due to its high process efficiency, automated potential, environmental feasibility.

Electrocoagulation (EC) has been suggested as an alternative method for removing pollutants in raw water and wastewater. EC consists of three processes that occur sequentially: electrolysis, coagulation, and flotation. Under a parallel monopolar electrode configuration, the application of an electric potential induces specific electrochemical oxidation and reduction pathways. At the sacrificial iron (Fe) anode, electrochemical oxidation drives the continuous dissolution of metal into reactive iron (II) ions ($Fe^{2+}_{(aq)}$). Concurrently, a reduction process occurs at the cathode surfaces, converting H_2O into volatile hydrogen gas ($H_{2(g)}$) and hydroxide ions ($OH^{-}_{(aq)}$). The fundamental electrochemical mechanisms governing the anode and cathode are explicitly expressed in Equations 1 and 2 (Şengil & Özacar, 2006; Sen et al., 2019):



In the subsequent coagulation process, the generated metal cations react spontaneously in the bulk solution phase to form active hydroxide flocs. This chemical path transitions from initial green/white iron (II) hydroxide, $Fe(OH)_{2(s)}$, which rapidly oxidizes in the presence of ambient air or dissolved oxygen to transform into the stable, insoluble brown iron (III) hydroxide matrix, $Fe(OH)_{3(s)}$, (Şengil & Özacar, 2006). The resulting $Fe(OH)_{3(s)}$ complex acts as a powerful coagulant and adsorbent that effectively removes pollutants from wastewater via adsorption or coprecipitation (Sen et al., 2019). Finally, during the flotation stage, the generated $H_{2(g)}$ microbubbles push the macro-flocs upward to the solution's surface, enabling them to be easily separated from the water medium.

Although macro-kinetic formulations (such as standard empirical first-order or second-order equations) are widely reported in the literature, they typically treat electrocoagulation as an isolated, static chemical reaction. This standard empirical approach completely ignores the actual boundary layer limitations

and liquid-to-solid mass transport phenomena taking place inside the active cell. Consequently, there remains a prominent knowledge gap in mathematically predicting how spatial configurations, such as inter-electrode gap sizes and potential fields, alter physical mass transport parameters.

The fundamental novelty of this research lies in the development and validation of a mathematically rigorous kinetic model explicitly derived from liquid-phase mass transfer boundary restrictions in a batch electrocoagulation reactor using iron (Fe) electrodes. Conducting this research is crucial because it bridges the gap between empirical observation and core transport phenomena, providing engineers with scalable mathematical tools rather than site-specific trial-and-error configurations. Furthermore, this study directly aligns with and advances the United Nations' Sustainable Development Goal 6 (Clean Water and Sanitation) by engineering a precise, energy-optimized, and quantifiable electrocoagulation protocol for hazardous dye mitigation.

Therefore, this study aims to evaluate the removal of Methylene Blue (MB) dye via electrocoagulation using iron (Fe) electrodes. The primary focus is to develop and validate a kinetic model based on mass transfer for MB separation in a batch system. The investigation covers the effects of electrode spacing (5, 10, 15, 20, and 30 mm) and applied voltage (10, 20, and 30 V) on optimizing the treatment efficiency.

Methods

Electrocoagulation Cell Up

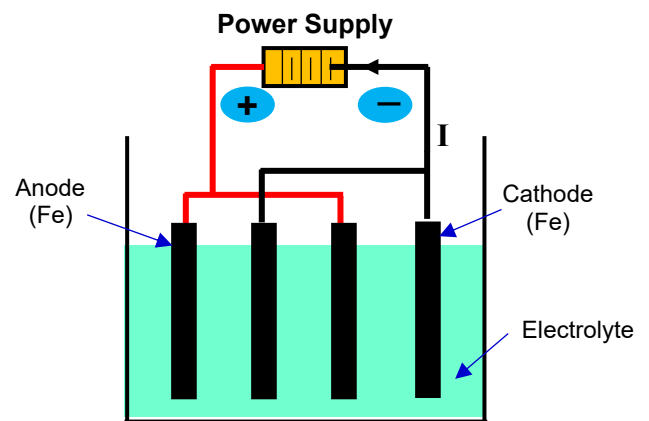


Figure 1. Electrocoagulation cell

The electrocoagulation unit with monopolar electrodes connected in parallel consists of an EC cell, a DC power supply, and electrodes. A direct current (DC) source is used to supply power to the EC system (0–30 V; 0–5 A). The Fe anode and Fe cathode are made of iron plates, each measuring 14 cm x 7 cm x 0.1 cm, which are

separated by a certain distance (Figure 1). Methylene blue stock solution (1000 mg/L) was prepared by dissolving a certain amount of methylene blue dye ($C_{16}H_{18}N_3ClS \cdot 3H_2O$, MW = 373.90 gmol⁻¹) in distilled water.

Experiment Method

The EC reactor is arranged as shown in Figure 1. The total number of Fe electrodes used is four, consisting of anode (2 units) and cathode (2 units). The experiment was carried out in batch mode. The experimental variations studied were electric voltage (10, 20, and 30 V) and electrode distance (5, 10, 15, 20, and 30 mm). The length of the experiment for each run was 75 minutes. A 2000 mL of 100 mg/L methylene blue dye solution was placed in the EC cell. NaCl solution is added to the solution, which functions as an electrolyte, and the solution's acidity is adjusted to neutral (pH = 7) by adding NaOH or HCl solution. The reaction begins when the DC power supply switches to the on position. All experiments were carried out at room temperature.

At each 15-minute interval, 25 mL of fluid samples were taken and analyzed. According to standard methods, the MB concentration in the solution was determined using a UV/VIS spectrophotometer at a wavelength of 665 nm. Dye removal efficiency (E(%)) is calculated using Equation 3.

$$E(\%) = \frac{(C_0 - C_t)}{C_0} \times 100\% \tag{3}$$

C_0 = dye concentration at the beginning (mg/L), C_t = dye concentration after t minutes (mg/L). The concentration of methylene blue dye was determined by plotting the absorbance value of the sample solution on a calibration curve using a standard methylene blue solution ($C_{16}H_{18}N_3ClS \cdot 3H_2O$, MW = 373.90 gmol⁻¹).

Kinetic Model Development

As previously explained, the process of removing pollutants (MB dye) from water/liquid waste in a batch process takes place through three stages as follows:

- a. Formation of Fe(OH)₃ coagulant particles through electrolysis.
- b. Mass pollutant (MB dye) transfer from liquid to Fe(OH)₃ coagulant particles
- c. Separation of flocs from a mixture by flotation.

The formation of Fe(OH)₃ coagulant particles generally takes place relatively quickly, so the rate is not controlled (process a). In EC, a certain amount of H₂ gas is generated due to the H₂ reduction process at the cathode. H₂ gas bubbles rush from the liquid phase to the surface and escape into the atmosphere. The movement of H₂ gas bubbles towards the surface of the liquid helps the flocs formed in the liquid rise. Therefore, separation of the flocs from the mixture by flotation

(process c) is assumed not to control the rate. Thus, in the electrocoagulation process that controls the rate of pollutant removal, only the mass transfer of pollutant (MB dye) from the liquid to the Fe(OH)₃ coagulant particles is used (process b).

In this proposed kinetic model, the rate of mass transfer from the liquid to the surface of the coagulant particles (as an adsorbent) can be approximated by Equation 4:

$$N_A \left[\frac{\text{mg A}}{(\text{volume adsorben}) \times (\text{time})} \right] = k_{ca} (C_A - C_A^*) t \tag{4}$$

C_A is the concentration of A (MB dye) in the liquid (mg A/L), C_A^* = concentration of A in the liquid which is in equilibrium with the concentration of A on the surface of the adsorbent or coagulant particles (mg A/L), t is the time (min), and k_{ca} is the volumetric mass transfer coefficient (men⁻²).

Based on the proposed model, the mass balance of substance A (MB pollutant) in the liquid phase can be written as:

$$[\text{rate of input}] - [\text{rate of output}] = [\text{rate of accumulation}] \tag{5}$$

$$0 - \{N_A V_B\} = V_L \frac{dC_A}{dt} \tag{6}$$

$$\frac{dC_A}{dt} = - \left(\frac{V_B}{V_L} \right) k_{ca} (C_A - C_A^*) t \tag{7}$$

$$\frac{dC_A}{dt} = - K_{OG} (C_A - C_A^*) t \tag{8}$$

$$K_{OG} = \left(\frac{V_B}{V_L} \right) k_{ca} \tag{9}$$

Equation (8) describes the change in concentration of pollutant A (MB dye) in the liquid (mg A/L/time) as a function of C_A and t. K_{OG} is the overall mass transfer coefficient (men⁻²), V_L is the liquid volume (L), and V_B is the adsorbent particle volume (L).

Results and Discussion

MB Dye Removal Efficiency

Processing efficiency is the percentage of pollutants (MB dye) that can be removed from the mixture at a particular time. In this study, the removal efficiency of MB dye was evaluated in two conditions, namely during the first 15 minutes of EC operation and at the end of the experiment (75 minutes). MB dye removal efficiency (E(%)) was calculated based on equation 3.

Influence of Applied Voltage on MB Removal Efficiency

The effect of varying electrical voltage (10 V, 20 V, and 30 V) on the efficiency of MB dye removal in EC was

evaluated under batch operating conditions with an electrode distance of 15 mm and an initial MB concentration of 100 mg/L. Figure 2 shows the plot of MB dye removal efficiency (E(%)) as a function of electrical voltage at 15 minutes and 75 minutes.

During the initial stage of the process at 15.00 minutes, the color removal efficiencies were recorded at 18.79%, 25.66%, and 88.12% for the applied potentials of 10.00 V, 20.00 V, and 30.00 V, respectively (Figure 2).

Based on Faraday's law, in the electrolysis process, the amount of anode metal that undergoes dissolution is directly proportional to the amount of electric current and the length of electrolysis time (t). Besides that, in the electrolysis process, the amount of electric current flowing through the EC system circuit is directly proportional to the amount of electric voltage used. Therefore, in the first 15 minutes, the amount or concentration of Fe(OH)₃ coagulant formed in the solution increases as the electrical voltage increases.

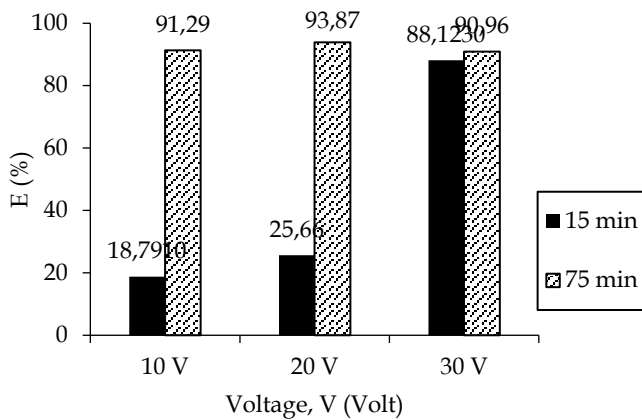


Figure 2. MB removal efficiency profile as a function of electrical voltage (15 mm electrode distance)

Thus, the magnitude of the applied potential difference will affect the availability of coagulant dosage in the reactor, the bubble production rate, floc size, and growth rate, and in turn, will affect the efficiency of phosphate removal from the EC process (Mollah et al., 2004; Mithra et al., 2017). The results obtained in this study are in line with the results of the study reported by Janpoor et al. (2011), Nguyen et al. (2017), and Sibanda et al. (2017).

In contrast, the removal efficiency of MB dye at 75 minutes was not significantly different at an electrical voltage of 10 V, 20 V, or 30 V (Figure 2). As time goes by, the amount of Fe(OH)₃ coagulant formed in the solution continues to increase. Therefore, the adsorption process occurs continuously until the remaining MB dye concentration becomes small. In this study, the initial concentration of MB dye in the solution was the same (100 mg/L), so at the end of the adsorption process (75 minutes), the MB dye removal efficiency (E (%))

achieved was also relatively no different. The average E (%) value for the three electrical voltage variations is 92.04%.

Effect of Inter-Electrode Distance (Effect of Inter-Electrode Distance)

The distance between electrodes (inter-electrode distance) is an essential parameter in the EC process because the electrostatic field depends on the distance between the anode and cathode (Jovanović et al., 2021; Naje et al., 2017). In this study, the influence of the distance between electrodes is studied at five different levels: 5, 10, 15, 20, and 30 mm. Experiments were carried out in batches at an initial MB dye concentration of 100 mg/L and a constant electrical voltage of 10 V. A plot of the MB dye removal efficiency as a function of the distance between the electrodes is shown In Figure 3.

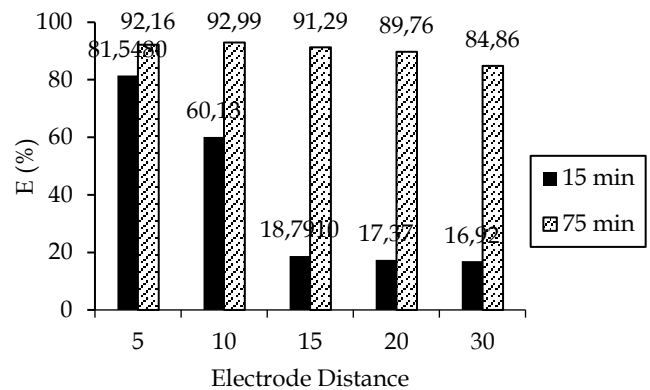


Figure 3. MB removal efficiency profile as a function of electrode distance

In the first 15 min, a significant decrease in processing efficiency was observed when the inter-electrode distance was increased by greater than 10 mm (Figure 3). Based on the data, the best processing efficiency is an electrode distance of 5 mm with an E value of 81.55%, followed by an electrode distance of 10 mm with an E value of 60.13%. Increasing the distance between electrodes to be greater than 10 mm, the processing efficiency is observed to be lower than 20%.

According to Ohm's law, the amount of electric current that passes through a metal conductor in a circuit is inversely proportional to the material's resistance (Maghanga et al., 2017). Furthermore, increasing the distance between electrodes (cathode and anode) will also increase cell resistance (Jovanović et al., 2021). Therefore, at a constant electric voltage, the number of Fe(OH)₃ molecules produced per unit time will decrease as the distance between the electrodes increases. This will ultimately lead to a decrease in the efficiency of MB dye processing.

By extending the duration of the EC process until the end of the experiment (75 minutes), the MB dye

removal efficiency was found not to differ significantly when the distance between the electrodes was increased. The average processing efficiency of MB dye at the end of the process is 90.21%. The logical reason for this phenomenon is that the number of Fe(OH)₃ coagulant molecules in the solution continues to increase as the duration of the EC process increases. Therefore, the MB dye adsorption process in solution can occur well within the available period.

MB Dye Removal Kinetics

The kinetics study of the processing process is essential in determining the design parameters of a reactor and separation systems. One frequently reported design parameter is the rate constant. This study modeled the kinetics of MB dye removal by electrocoagulation as shown in Equation 7. In this study, experiments were carried out in batches under

isothermal conditions. Data on changes in MB dye concentration at various times (t) were recorded and used to verify the adsorption kinetic model and evaluate the model parameter values, namely KOG. In this model, the C_A* value is determined based on C_A value data when no significant changes occur. Solving Equation 9 numerically uses the fourth-order Runge Kutta method (Chapra & Canale, 2010). The KOG value is estimated by minimizing the Sum Square Error (SSE) between experimental data and model data (equation 10) numerically using the Golden Section Search method (Bazaraa et al., 2006; Chapra & Canale, 2010) using Microsoft Excel software.

$$SSE = \sum (C_{A,data} - C_{A,model})^2 \tag{10}$$

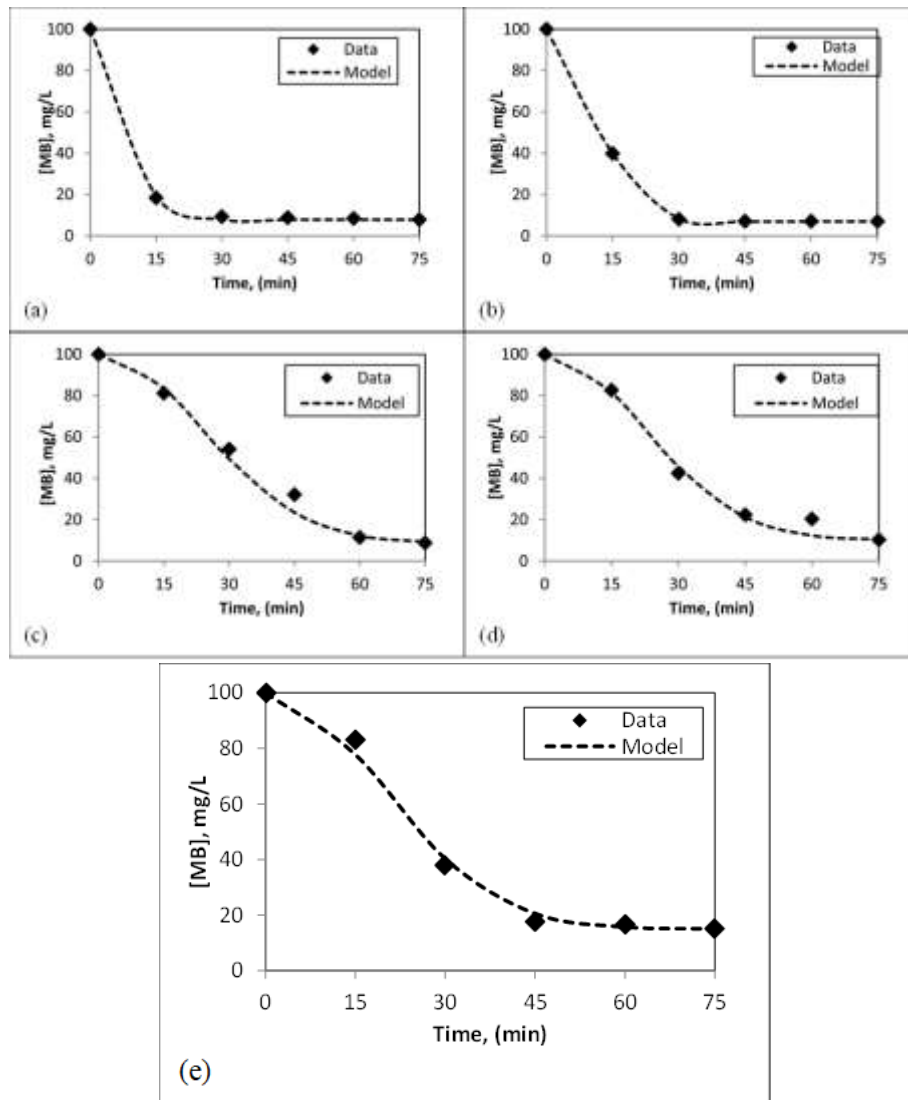


Figure 4. Plot of methylene blue (MB) concentration as a function of time at an electrical voltage of 10 V. (a) 5 mm, (b) 10 mm, (c) 15 mm. (d) 20 mm and (e) 30 mm

$C_{A,data}$ is the concentration of MB in liquid (mg/L) experimental results; $C_{A,model}$ is the concentration of MB in the liquid (mg/L) calculated using the kinetic model. A plot of the relationship between MB dye concentration values versus contact time (t) is shown in Figure 4 (a - e).

Based on the plot shown in Figure 4 (a-e), changes in the concentration of MB dye in the solution can be modeled well-using equation 8. The smallest SSE value (0.0897) was obtained in experiments with an electrode distance of 5 mm and a voltage of 10 V. Meanwhile, the most considerable SSE value (193.57) was found in experiments with a distance of 15 mm and a voltage of 20V (Graph not shown).

The kinetic constant value of MB dye removal based on the model built is shown in Table 1. At a voltage of 10 V, the mass transfer coefficient (K_{OG}) value decreases from 0.01895 min^{-2} to 0.00939 min^{-2} when the distance between electrodes is increased from 5 mm to 10 mm. The same thing was found when the distance between electrodes was increased again to 15 mm. At an electrode distance of 15 mm, the K_{OG} value is significantly reduced to 0.00153 min^{-2} . However, the K_{OG} value increased again when the electrode distance was increased to 20 mm or 30 mm.

The influence of variations in electric voltage on the kinetic constants of MB dye removal was also observed from the K_{OG} values in Table 1. At the same electrode distance (15 mm), the K_{OG} values were found to be more significant when the electric voltage was increased from 10 V to 20 and 30 V. 10 V, the K_{OG} value obtained is 0.00153 min^{-2} , while at voltages of 20 V and 30 V, the K_{OG} value obtained is 0.00425 min^{-2} and 0.03115 min^{-2} respectively.

Table 1. Kinetic parameters for EC removal of MB using Iron (Fe) electrodes

Voltage (V)	Electrode Distance (mm)	$K_{OG}(\text{min}^{-2})$	SSE
10	5	0.01895	4.2892
	10	0.00939	0.0897
	15	0.00153	46.6143
	20	0.00207	76.6913
	30	0.00271	42.6759
20	15	0.00425	193.5701
30	15	0.03115	4.4688

Conclusion

This study demonstrates that applied voltage, electrode spacing, and reaction time are key factors governing the efficiency of methylene blue (MB) dye removal via electrocoagulation. At an applied voltage of 10 V, a reduction in electrode spacing from 15 mm to 5 mm markedly increased the mass transfer coefficient K_{OG} from 0.00153 min^{-2} to 0.01895 min^{-2} , achieving a peak removal efficiency of 81.55% within 15 minutes.

Conversely, a non-linear trend was observed at extended distances of 20 mm and 30 mm, where the K_{OG} values exhibited a minor upward shift 0.00207 min^{-2} and 0.00271 min^{-2} , indicating complex fluid dynamics or altered passivation behaviors at wider gaps. Furthermore, increasing the applied voltage from 10 V to 30 V at a constant 15 mm spacing significantly enhanced the K_{OG} value from 0.00153 min^{-2} to 0.03115 min^{-2} due to accelerated anodic dissolution. Crucially, the overall removal kinetics were successfully quantified by the proposed mass transfer-based kinetic model, demonstrating robust alignment with experimental data as validated by a low and concise Sum of Squared Errors (SSE) range of 0.0897 to 193.57.

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Author Contributions

Conceptualization, A.H., D.T.A., and L.S.; methodology, A.H. and D.T.A.; software, investigation, data curation, D.T.A.; validation, A.H., L.S., and D.S.; formal analysis, visualization, D.T.A. and F.F.; resources, supervision, A.H. and L.S.; writing—original draft preparation, D.T.A., F.F., and A.H.; writing—review and editing, D.T.A., F.F., A.H., and L.S.; project administration, funding acquisition, A.H. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest

The authors declare no conflict of interest.

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