



Enhanced methylene blue removal from aqueous solution by electrocoagulation technique using combined iron and copper electrode

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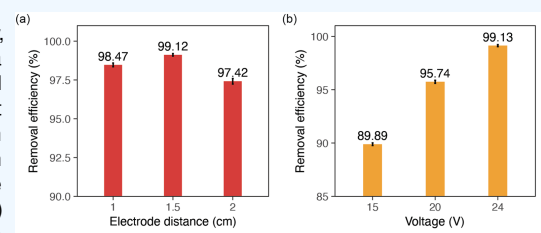
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ABSTRACT

The rapid advancement in the industrial sector, particularly within the textile industry, necessitates the use of colorants for fabric processing, with methylene blue being a predominant choice. However, the improper disposal of methylene blue dyes can lead to significant environmental hazards, underscoring the need for effective treatment methods before release into ecosystems. This study explores electrocoagulation, an innovative treatment technique powered by electrical energy, as a potential solution for mitigating the environmental impact of methylene blue in textile wastewater. The research investigates the efficiency of the electrocoagulation process using iron (Fe) and copper (Cu) electrodes under various operational conditions, including electrode spacing (1, 1.5, and 2 cm), applied voltage (15, 20, and 24 volts), and electrode contact time (5, 10, 15, 20, 30, and 60 minutes). The findings reveal that the electrocoagulation method achieves an optimal methylene blue removal efficiency of 99% under specific conditions: an electrode distance of 1.5 cm, an applied voltage of 24 volts, and a contact duration of 30 minutes. This research underscores the potential of electrocoagulation as an efficient and environmentally friendly approach for the treatment of textile wastewater, contributing to the sustainable management of industrial effluents.

Keywords: *Electrocoagulation, Iron, Copper, Electrode spacing, Voltage effect, Time effect*



1. INTRODUCTION

The burgeoning textile industry is a significant contributor to escalating environmental degradation, with water pollution being a particularly grave concern. Textile effluents are notoriously difficult to manage due to their high organic load, intense coloration, elevated salinity, and recalcitrant nature [1]. A prime example of these challenging pollutants is methylene blue, a dye ubiquitously employed in textile production for its vibrant blue hue [2]. The pervasive use of methylene blue is attributed to the fundamental role that blue dyes play in textile color schemes. This deep-seated reliance on methylene blue in the textile sector underscores the pressing need for innovative treatment methodologies to address the environmental toll of textile wastewater.

The presence of methylene blue in aquatic ecosystems poses a significant threat to biodiversity due to its inherent toxicity, which can inflict harm across a spectrum of living organisms [3]. Not only does it bear a high level of toxicity, but methylene blue also impedes photosynthetic processes in aquatic plants, disrupting the delicate balance of underwater ecosystems [4]. The dangers of methylene blue extend to numerous toxicological impacts, including its propensity to cause poisoning in aquatic life. In humans, exposure to methylene blue can result in a range of serious health issues, from hypothermia and heightened blood pressure to corneal damage and detrimental effects on the respiratory system, as documented by Hatem A. Al-Aoh (2013) [5]. These findings serve as a stark reminder of the critical need for stringent con-

trols on the release of methylene blue into the environment.

The detrimental effects of methylene blue on both the environment and human health necessitate the exploration and development of more effective and sustainable waste treatment methods. While biological processes present a potential avenue, their effectiveness is often compromised by the inhibitory effects dyes have on bacterial growth, as discussed by Paz et al. (2017). Alternative technologies, including adsorption, photocatalytic degradation, and membrane separation, have been advanced to address the challenges posed by textile effluents, according to research by Melo et al. (2017), Farzana (2014), and Wang et al. (2018) [6, 7, 8]. However, the adoption of these methods is hindered by significant limitations, notably the high operational costs involved, as identified by Aravind et al. (2016) [9]. Consequently, there is a compelling need for innovative, cost-effective, and environmentally benign solutions to mitigate the impact of textile dyes on ecosystems and public health.

The electrocoagulation method stands out for its high efficacy in extracting dyes from textile wastewater, operating on the principle of using electrical currents to destabilize and aggregate dissolved contaminants for easier removal. This process is particularly sensitive to operational parameters such as the duration of exposure (contact time), the intensity of the electric current (voltage), and the spatial arrangement of the electrodes (electrode distance). A significant advantage of electrocoagulation lies in its proficiency in targeting and removing even the most minute colloidal particles, which

traditional methods might leave behind. Moreover, it offers a reduced treatment time, enhancing efficiency and making it a more attractive option for industrial applications, as detailed by V & AK (2013)[1]. This method presents a promising direction for improving the sustainability of wastewater management in the textile industry.

An electrocoagulation reactor fundamentally comprises a pair of electrodes, namely an anode and a cathode, which are essential for the process. Copper is recognized for its potential as an effective electrode material in electrocoagulation, while iron electrodes have been identified as more efficient than aluminum in mitigating methylene blue concentrations, a finding supported by Li et al. (2011) [10]. Building on this knowledge, the present study is designed to evaluate the efficacy of electrocoagulation for the removal of methylene blue using a synergistic approach that combines iron (Fe) and copper (Cu) electrodes. The investigation will methodically vary the distance between electrodes, the applied voltage, and the duration of contact time to determine the optimal set of conditions for maximizing dye removal efficiency. This research aims to refine the electrocoagulation process parameters to enhance its application in treating textile industry effluents.

The exploration of electrocoagulation using a hybrid approach of iron and copper electrodes for the reduction of methylene blue concentrations is a relatively uncharted area of study. This research intends to fill a gap in the current scientific literature, providing a comprehensive analysis of the effectiveness of this novel electrode combination. The outcomes are anticipated to contribute significantly to existing knowledge on electrocoagulation efficacy and to potentially establish a new standard for electrode materials suitable for application within the textile industry. By advancing this method, the study aims to offer a viable and efficient solution for wastewater treatment, aligning with the industry's growing commitment to environmental stewardship and sustainable practices.

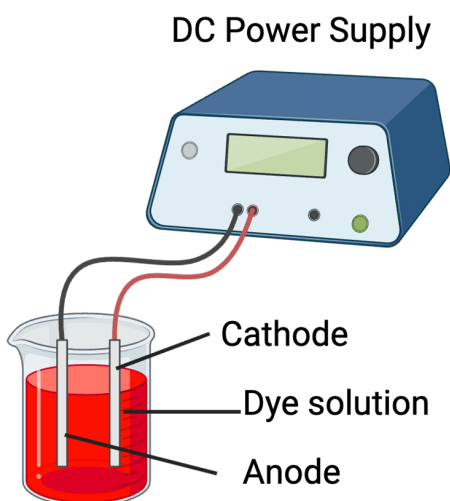


Figure 1. Electrocoagulation reactor

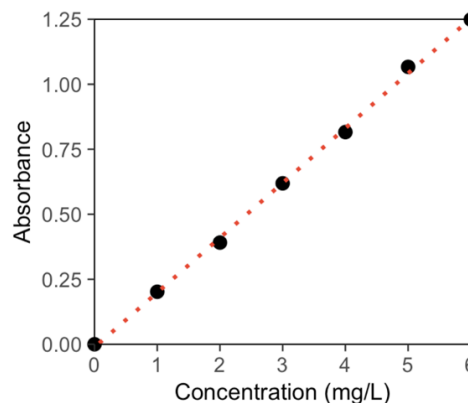


Figure 2. Methylene blue calibration curve

2. EXPERIMENTAL SECTION

2.1 Material

The methylene blue (MB) solution was prepared and used as a model of dyes containing wastewater. MB was purchased from Merck, Germany, in analytic grade and used as received without further purification. The water used in this study was deionized water and tap water. The Fe and Cu electrode was purchased from a local store in the form of metal plates. Before use, the electrodes were prepared to be 2 cm wide.

2.2 Electrocoagulation experiment

A 1000 ppm methylene blue (MB) stock solution was prepared by dissolving 1 gram of MB powder in 1000 ml of distilled water. This stock solution was then utilized to prepare both a series of standard solutions for the calibration curve and a 500 ml batch of 50 ppm MB artificial wastewater. For the electrocoagulation process, a 500 ml glass beaker was set up as the reactor, with iron (Fe) and copper (Cu) electrodes positioned adjacently, designating Fe as the anode and Cu as the cathode, and connected to a power supply. The schematic illustration of the electrocoagulation set-up is presented in Figure 1. The electrocoagulation process was conducted by adjusting the electrode spacing to 1, 1.5, and 2 cm and varying the applied voltage at 15, 20, and 24 volts. Following the determination of the optimal electrode distance and voltage, the experiment will explore contact time effects, with samples collected at intervals of 5, 10, 15, 20, 30, and 60 minutes.

The research employs descriptive analysis to elucidate the gathered data, which will initially be presented in tables before being graphically depicted. This approach facilitates an in-depth examination of methylene blue reduction under varying conditions namely electrode spacing, applied voltage, and contact time within the electrocoagulation process. The percentage reduction in color of the textile wastewater is quantified using the UV-Vis spectroscopy (Thermo scientific Genesys 20) technique, and the removal efficiency of the experiment was calculated by Equation 1,

$$E = \frac{C_0 - C_x}{C_0} \times 100\% \quad (1)$$

where is the removal efficiency (%), C_0 is the initial concentration of the dye (mg/l), and C_1 is the final concentration of the dye (mg/l)

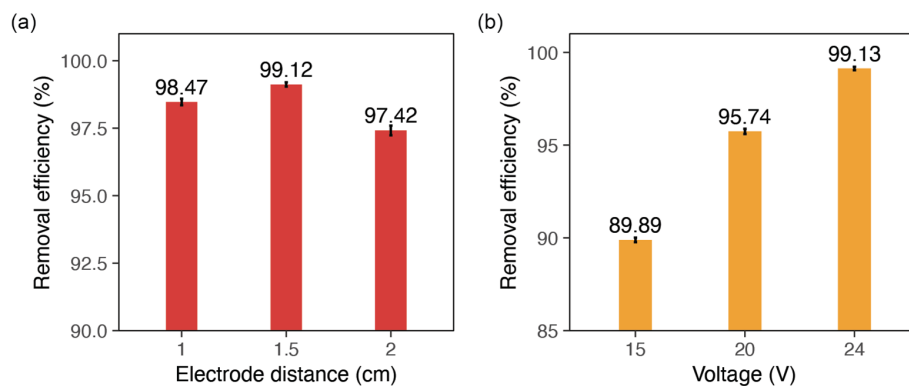


Figure 3. Distance efficiency between electrodes (a) and Efficiency of voltage variation (b)

3. RESULT AND DISCUSSION

3.1 Standard Solution and Calibration Curve

The creation of a standard solution was a critical step in evaluating its adsorption characteristics via UV-Vis spectrophotometry. An initial scan was conducted to identify the optimal wavelength for methylene blue, which is crucial for constructing the standard calibration curve. The scanning process on the UV-Vis spectrophotometer yielded an optimal wavelength of 664 nm for methylene blue. This wavelength aligns with those reported in previous studies on methylene blue electrocoagulation (D, et al., 2018), confirming the consistency and reliability of our findings in the context of established research [11].

The coefficient of determination, R^2 , quantifies the strength of the correlation between the concentration and absorbance of the methylene blue standard solution. A value of R^2 nearing 1 indicates a strong correlation, suggesting that the calibration curve for the standard solution possesses high accuracy, as supported by findings from Utomo et al. (2018) [12]. This close correlation underscores the reliability of the calibration curve in reflecting the true relationship between methylene blue concentration and its absorbance.

As can be found in 2, The calibration curve for methylene blue in this research yielded an R^2 value of 0.9987, indicating an excellent fit and suggesting that the curve is highly reliable for assessing methylene blue concentrations. The derived linear regression equation, $y = 0.2082x - 0.01168$, where y represents the absorbance and x the concentration, will be instrumental in calculating the concentration levels of methylene blue in subsequent analyses. This high degree of correlation reinforces the validity of the calibration curve for precise concentration determinations.

3.2 Effect of Distance Between Electrodes

The electrode spacing plays a pivotal role in the electrocoagulation process, influencing the electron transfer rate between the electron-releasing anode and the reduction-occurring cathode, as highlighted by Trisnaawati & Purnama (2021)[13]. In this investigation, electrode distances were systematically varied at 1 cm, 1.5 cm, and 2 cm to explore their impact on the efficiency of the electrocoagulation process in removing methylene blue from the solution.

The study's findings (3(a)) indicate that among the tested electrode spacings—1 cm, 1.5 cm, and 2 cm—the 1.5 cm distance proved most effective for methylene blue removal. At this spacing, the electrocoagulation process achieved an im-

pressive removal efficiency of $99.12\% \pm 0.08$, reducing the methylene blue concentration from an initial 50 ppm in the artificial wastewater to just 0.44 ppm. This highlights the critical role of electrode spacing in optimizing the electrocoagulation process for dye removal.

The outcomes of this research suggest that a narrower electrode gap generally enhances dye removal efficiency, yet an overly close spacing is counterproductive due to the tendency of formed flocs to obstruct the electrode surfaces. This observation aligns with the findings of Purwati, Alimuddin, and Erwin (2018), emphasizing the delicate balance required in electrode spacing for optimal electrocoagulation performance. Specifically, in this study, a 1 cm electrode spacing presented practical challenges, including the risk of electrodes coming into contact, which could lead to a surge in current and potentially damage the equipment. This highlights the importance of selecting an electrode distance that maximizes removal efficiency while avoiding operational pitfalls.

3.3 Effect of Voltage

Prayitno, Ridantami, and Prayogo (2016) highlight that the efficacy of electrodes in generating coagulants during electrocoagulation is directly influenced by the applied electric voltage [14]. The production of coagulants is directly proportional to the electric charge, indicating that an increase in voltage enhances the removal efficiency. Consequently, this process leads to the formation of flocs, which can either settle at the bottom or float to the water's surface. In this particular study, it was observed that the majority of the flocs formed were floating above the water surface, suggesting a specific behavior of the flocs under the applied conditions and possibly influencing the overall effectiveness and the subsequent removal process.

The applied voltage significantly influences the contact time required for effective dye removal in the electrocoagulation process. The findings from this study illustrate this relationship: at the 30-minute mark, a 24-volt application achieved a $99.13\% \pm 0.09$ removal efficiency of methylene blue. Conversely, at the same time interval, a 20-volt setting resulted in a slightly lower efficiency of $95.74\% \pm 0.14$, and the 15-volt application exhibited the least effectiveness, with an $89.89\% \pm 0.12$ removal rate. These results indicate that lower voltages necessitate longer contact times to achieve comparable levels of dye removal, underscoring the importance of optimizing both voltage and contact time to enhance the electrocoagulation process's efficiency.

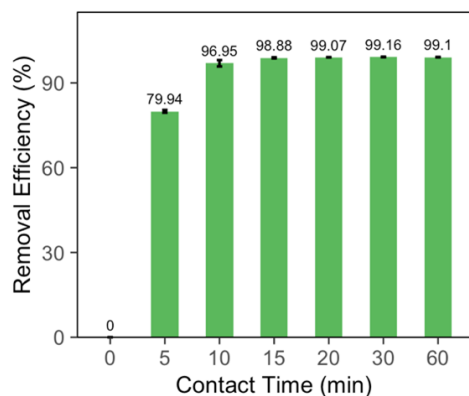


Figure 4. Effect of contact time on the methylene blue removal efficiency

3.4 Effect of Contact Time

Contact time is a crucial factor in the electrocoagulation process, significantly influencing the efficiency of pollutant removal. As contact time extends, more pollutant particles undergo coagulation, leading to enhanced removal effectiveness. This trend is evident in the observed data, where the removal efficiency of methylene blue consistently rises with the extension of contact time from 5 minutes to 60 minutes. This pattern underscores the importance of optimizing contact time to maximize the electrocoagulation method's ability to purify water from contaminants such as dyes.

The initial phase of the electrocoagulation process, particularly within the first 10 minutes, showcases a remarkable increase in removal efficiency. Starting with a $79.94\% \pm 0.48$ removal rate at 5 minutes, the efficiency surges to $96.95\% \pm 1.13$ by the 10-minute mark. This rapid improvement can be attributed to the proliferation of flocs or deposits that begin to accumulate at the water's surface. According to Hidayanti, Afifa, Ismuyanto, and Juliananda (2021) [15], these deposits are formed through the interactions between the coagulants and pollutants, highlighting the effectiveness of the electrocoagulation process in aggregating and separating pollutants from water within a relatively short timeframe.

By the 15-minute mark, the removal efficiency of the electrocoagulation process had further escalated to $98.88\% \pm 0.18$, bringing the average methylene blue concentration from experiments 1 and 2 down to 0.56 ppm. Beyond this point, while the efficiency continued to rise through the 20th and 30th minutes, the increments became less pronounced. Specifically, at 20 minutes, methylene blue removal reached $99.07\% \pm 0.04$, with the combined average concentration from both experiments dropping to 0.46 ppm. The peak efficiency was observed at the 30-minute interval, achieving a $99.16\% \pm 0.07$ removal rate and reducing the average concentration to 0.42 ppm. This pattern underscores a diminishing rate of efficiency improvement over time, suggesting an approaching equilibrium in the electrocoagulation process's ability to remove contaminants.

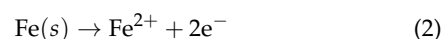
The concentration of coagulants generated in the electrocoagulation process is directly proportional to the electric charge applied, implying that an increase in voltage enhances the efficiency of contaminant removal. This relationship is supported by findings from Setianingrum, Prasetya, & Sarto (2017) and Hamid, Purwono, & Oktiawan (2017), which also

note that the formed flocs have a tendency to either settle at the bottom or float to the water's surface [16, 17]. The distribution of flocs between settling and floating is an important aspect of the electrocoagulation process, affecting the ease of subsequent floc removal and overall water treatment efficacy.

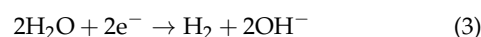
3.5 Electrocoagulation Effectiveness of Iron (Fe) and Copper (Cu) Electrodes

In the electrocoagulation process involving an Fe anode, several key chemical reactions take place. At the Fe anode, the primary reaction involves the oxidation of iron to form iron ions, which then interact with water to produce coagulants. The generalized reactions at the Fe anode can be described as follows:

On the Fe anode:



At the Cu cathode:



The electrocoagulation process, as applied in this study with iron anodes and copper cathodes, has demonstrated significant efficacy in reducing methylene blue concentrations in wastewater. The mechanism involves the release of electrons from the iron anode, which subsequently interact with water molecules to produce hydroxide ions. These hydroxide ions are crucial for the formation of insoluble hydroxide flocs through reactions with cationic species present in the wastewater, facilitating the aggregation and removal of pollutants.

Moreover, the oxidation reaction occurring at the iron anode plays a pivotal role in the process, enabling the binding and removal of pollutants from the wastewater. This dual action, encompassing both the formation of coagulating agents and the direct oxidation of pollutants, underscores the effectiveness of electrocoagulation in treating dye-laden effluents, as supported by the work of Andesgur, Hakim, & Julianto (2014)[18]. This process not only reduces the levels of methylene blue but also contributes to the overall improvement of water quality through the removal of a broad spectrum of contaminants.

The production of hydroxide ions in the electrocoagulation process is a key step in the treatment of wastewater, particularly for the removal of contaminants like methylene blue. These hydroxide ions readily react with the iron ions (Fe^{2+}) which are generated from the dissolution of the iron anode, leading to the formation of iron hydroxide precipitates, specifically $\text{Fe}(\text{OH})_2$. The formed $\text{Fe}(\text{OH})_2$ precipitates play a crucial role in the purification process. They act as adsorbents, binding dissolved compounds, and also as flocculants, capturing and aggregating suspended particles. This dual functionality enhances the removal of methylene blue from the water, as the dye molecules are adsorbed onto the $\text{Fe}(\text{OH})_2$ precipitates and subsequently removed from the solution through sedimentation or filtration. This mechanism of action, as detailed by Mahmoud, Farah, & Farrag (2013)[19], highlights the effectiveness of iron-based electrocoagulation in treating dye-polluted waters.

4. CONCLUSION

The research focused on methylene blue removal via electrocoagulation with iron and copper electrodes leads to the

following conclusions. Electrode spacing impacts the process efficiency, with 1.5 cm identified as the optimal distance. Moreover, a direct correlation between voltage and removal efficiency was observed, with 24 volts yielding the best results. Additionally, the data indicate that a contact time of 30 minutes is most effective for maximum dye removal, underscoring a general trend where longer electrocoagulation durations enhance removal efficiency. The electrocoagulation technique, employing iron (Fe) and copper (Cu) electrodes, proves to be highly effective in reducing methylene blue concentrations, achieving up to 99% removal efficiency. This high level of effectiveness highlights the potential of electrocoagulation as a viable method for treating dye-contaminated waters.

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