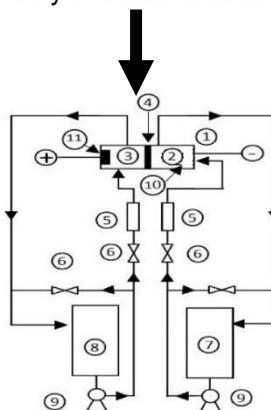
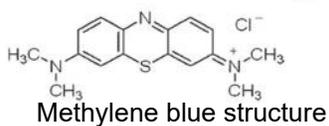
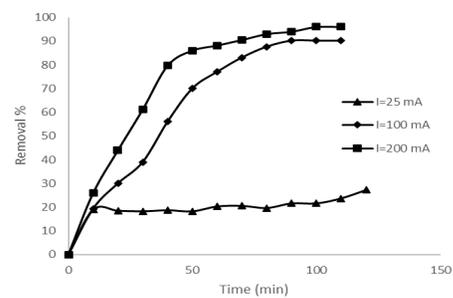
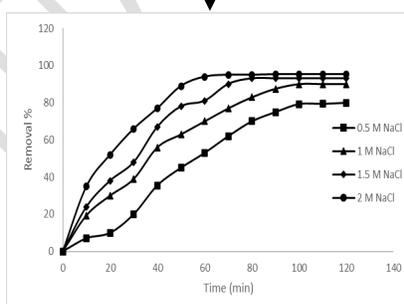
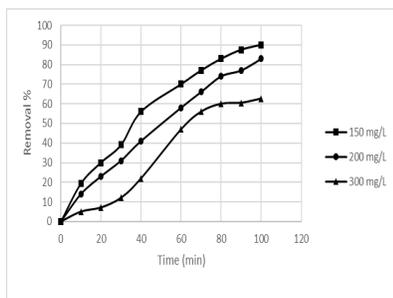


Graphical Abstract



Electro-oxidation treatment system



The effect of dye concentrations, molarity, and treatment current, on the methylene blue removal

Anodic Oxidation of Methylene Blue Dye by Aluminum Tubes Bundle Anode

Ghazi Faisal Naser^{1,2*}, Ali Saleh Jafer¹, Abbas Abdulameer Al-Raad³
Ihsan H. Dakhil¹

¹Chemical Engineering Department, College of Engineering, Al-Muthanna University, Iraq

²College of Engineering, Al-Ayen University, Thi-Qar, Iraq

³Directorate of Education Al-Muthanna, Ministry of Education, Iraq

Email: ghazi_faisal@mu.edu.iq^{1,2*}, ali.salih@mu.edu.iq¹, Alraadabbas89@gmail.com³,
em2ihsan@mu.edu.iq¹

* Corresponding author: Ghazi Faisal Naser

Email: ghazi_faisal@mu.edu.iq

ABSTRACT

The objective of this study was to evaluate the efficiency of the electro-oxidation technique for the degradation of methylene blue from simulated wastewater. A flow-through electrochemical batch-circulation reactor was utilized to remove methylene blue from wastewater using a concentric tubes anode constructed of aluminum. The parameters studied in this work were dye concentration, electrolyte molarity, applied current, pH, and flow rate. The response indicators were the dye removal percentage and chemical oxygen demand (COD).

The results demonstrated that the removal efficiency decreases as the dye concentration increases and also decreases with increasing pH. In contrast, the removal efficiency increases with increasing electrolyte molarity, flow rate, and applied current. The results clearly indicate that the applied process is effective for dye removal. The reactor operated in batch-circulation mode proved to be highly effective in removing the dye within a shorter time and with lower energy consumption.

Keywords: Pollutants; Electro-oxidation; Electrolyte; COD removal; Textile wastewater;

Degradation; Methylene Blue

1. Introduction

At present, water pollution represents a serious impact on environment especially with the rapid growth in water pollution sources, based on World Bank investigations, textile manufacturing including dyeing and finishing may be responsible for 17–20% of industrial wastewater (Christian *et al.* 2023). Synthetic dyes are the main source of waste in the textile sector and can further contaminate a large amount of water if improperly processed (Repon *et al.* 2024). Metals, dyes, and other contaminants can be found in textile wastewater. The characteristics of textile effluent differ depending on the industry, machinery, method of production, type of material, chemicals, season, and fashion trends (Kumari *et al.* 2025). Approximately 7×10^7 metric tonnes of synthetic dyes are produced worldwide each year. The textile industry uses around 10,000 metric tons of these dyes (Chandanshive *et al.* 2020). Since the colors applied to textile wastewater have a complicated and harmful element composition, the wastewater effluents are categorized as hazardous wastes (Kallawar *et al.* 2023). One of the main compounds that contaminate water is industrial dyes. Methylene blue (MB), one of these dyes, is poisonous, carcinogenic, and non-biodegradable, offering a serious risk to both environmental safety and human health (Khan *et al.* 2022). The textile, pharmaceutical, paper, dyeing, printing, paint, medical, and food industries are just a few of the numerous possible uses for MB dye (Koyuncu *et al.* 2020 ; Mijinyawa *et al.* 2019; Parakala *et al.* 2019; Balarak *et al.* 2020). In the textile sector, it is the most often used dye (Arias *et al.* 2020), and it is regarded as one of the most widely used colorants for textiles (Siddeeg *et al.* 2019). MB and other textile dyes can be extracted from industrial wastewater via a variety of techniques such as : coagulation (Lau *et al.* 2015; Liu *et al.* 2015), electrocoagulation (Mahmoud *et al.* 2013; Tir *et al.* 2015; Ghazi *et al.* 2023), electro-fenton (Ghazi *et al.* 2024;) ultrafiltration (Zheng *et al.* 2009; Khosa *et al.* 2011; Kim *et al.* 2020), biodegradation (Eslami *et al.* 2017; Kilany *et al.* 2017; Van Der *et al.* 2018), microwave treatment (García *et al.* 2017), nanofiltration (Kong *et al.* 2019; Cheng *et al.* 2012; Zhong *et al.* 2019), vacuum membrane distillation (Banat *et al.* 2005) , liquid-liquid extraction (El-Ashtoukhy *et al.* 2015), phytoremediation (Imron *et al.* 2019; Tan *et al.* 2016), adsorption/biosorption (Wang *et al.* 2020; Andrade *et al.* 2020; Regunton *et al.* 2018; Gopalakrishnan *et al.* 2020; Hameed 2020; Li *et al.* 2020 ; Ihsan *et al.* 2021) and hybrid systems (Naresh *et al.* 2020; Nguyet *et al.* 2019; Lee *et al.* 2012; Sun *et al.* 2020).

It is difficult to destroy MB dye into tiny molecules using standard techniques because its stable under thermal and light and non-biodegradability (Liu *et al.* 2019; Liu *et al.* 2020).

In order to address dangerous organic pollutants like MB, advanced oxidation processes (AOPs) were created using effective redox mechanisms that produce certain radicals without producing any more hazardous compounds (Fosso *et al.* 2020; Khan *et al.* 2020; Zhang *et al.* 2019). Therefore, advanced oxidation processes are usually applied to strip organic contaminants in the wastewater (Zou *et al.* 2023; Zheng *et al.* 2025; Xu *et al.* 2024) .

The capacity of electrochemical treatment methods for operation without the need of chemical additives lowers the amount of sludge produced. However, it is essential to recognize the disadvantages of these approaches, which include higher electrical energy expenditures and substantially lesser efficacy compared to alternative treatment options (Zhang *et al.* 2021).

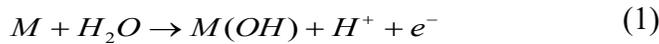
One of the more effective electro-oxidation methods for handling wastewater containing dyes is electro-oxidation (EO) (Ewuzie *et al.* 2022; Rodríguez-Narváez *et al.* 2021).

Because of its many uses: low environmental impact, easy design and operation, simple of adjusting pressure, voltage utilized, and temperature, automation abilities, breakdown of refractory organics, limited sludge manufacture, and total mineralization, electrochemical

oxidation has drawn the attention of researchers during the last 20 years (Deng *et al.* 2019; Mandal *et al.* 2017; Narenkumar *et al.* 2023).

Electro-oxidation (EOX) effectively removes a variety of organic contaminants using small equipment and few chemicals (Abou-Taleb *et al.* 2021). On electro catalytic surfaces, it generates hydroxyl and superoxide radicals that eliminate bacteria and transform organic materials into carbon dioxide and water (Sandoval *et al.* 2021; Hellal *et al.* 2022). There are two types of electro-oxidation of organic pollutants: direct and indirect. Hypochlorite and chlorine are produced on the anode for indirect degradation, and these groups will act as organic degradation. In addition to a possibility of chemisorbed oxygen creation at the anode, hydroxyl groups are created there for direct oxidation, and these groups will act as organics (Lee *et al.* 2022; Comminellis *et al.* 2010).

Two reactions will be involved in the oxidation mechanism; building of hydroxyl group at anode (reaction1), and the oxidation of organics with this hydroxyl (reaction2)(Louafi *et al.* 2016):



When NaCl presents in the supporting electrolyte, chlorine group will act a strong role in the oxidation (Nidheesh *et al.* 2018; Mario *et al.* 2012). When NaCl presents, Cl⁻ will be oxidized on the anode to form Cl₂ (reaction 3), the Cl₂ will be hydrolyzed to form HClO, then the equilibrium dissociation reaction of HClO (Garcia-segura *et al.* 2018):



In order to form chlorine by oxidation of chloride and prevent chlorine oxidation non oxidizing groups, active anodes like Pt, IrO₂ or TiO₂ may be used (Garcia-segura *et al.* 2018). Hydroxyl group may also form chlorine according to reaction 6(Bonfatti *et al.* 2000; Rosestolato *et al.* 2014; Neodo *et al.* 2012):



High oxygen evolution potential is the main characteristic that should be taken in consideration in choosing the electrode material in order to ensure complete degradation of organic contaminants with significant current effectiveness (Labiadh *et al.* 2016; Sopaj *et al.* 2015; Yang *et al.* 2016). Several materials have been successfully used for organic pollutants mineralization such as; PbO₂ (Chen *et al.* 2021), SnO₂ (Giannakopoulos *et al.* 2022), and BDD anodes (De Luna *et al.* 2022), carbon(Shestakova *et al.* 2017), titanium coated with RuO₂/IrO₂/TaO₂(Pattabhi *et al.* 2021), platinum(Kamyab *et al.* 2022), PbO₂/Ti(Qiu *et al.* 2024), Al(Yang *et al.* 2024), stainless steel(Abou-Taleb *et al.* 2021), IrO₂- SnO₂-Sb₂O₅ Coated Ti(Bravo-Yumi *et al.* 2022),

RuO₂/Ti(Mardani *et al.* 2023), porous graphite(Ghazi *et al.* 2023 ; Ihsan *et al.* 2025) graphite-felt(Pi *et al.* 2022), exfoliated graphite(Yu *et al.* 2022).

Nevertheless, conventional two-dimensional (2D) planar electrodes have drawbacks, such as inadequate active sites, poor efficiency of mass transfer, and poor current distribution, which lead to treatment inefficiencies and excessive energy consumption that fall short of industrial standards (Zhang *et al.* 2018). Three-dimensional (3D) electrode technology has been created by combining conductive particles or porous media to create a three-dimensional reaction contact in order to overcome the intrinsic limits of 2D electrodes (Li *et al.* 2023; Wang *et al.* 2021; Thanigaivel *et al.* 2024). This research employs the electrochemical oxidation for removing methylene blue dye (MBD) from simulated wastewater and the best value was found from multiple variables such as dye concentration, supporting electrolyte molarity, treatment current, pH, and flow rate. The Methylene blue chemical structure is presented in **Figure 1**.



Figure 1. Methylene blue structure (Zhang *et al.* 2011)

Future tests in actual wastewater matrices offer an extensive overview of the system's performance and useful possibilities for wastewater treatment.

2. Materials and methods

2-1. Materials

In this study, the chemicals used were sodium chloride (99% purity), methylene blue dye, sodium hydroxide, and hydrochloric acid. All chemicals were of analytical grade. All electrolyte solutions were prepared using distilled water.

2-2. Experimental rig

Electrochemical oxidation was carried out in a flow-through electrochemical reactor operating in batch-circulation mode. The main body of the reactor consists of a cubic box (15 × 15 × 15 cm) made of Teflon, with a horizontal cylindrical hollow serving as the flow channel through the reactor.

The anode consists of a bundle of seventeen aluminum tubes. The specifications of these tubes are shown in Table 1. The cathode is a graphite disk with a diameter of 7 cm and a thickness of 3 mm. A DC power supply equipped with a multiturn high-precision potentiometer was used to accurately adjust the applied current. The applied current and voltage were measured using a digital

multimeter. The dye concentration was measured using a visible spectrophotometer, and the COD was determined using a Lovibond COD system. The experimental rig is illustrated in **Figure 2**.

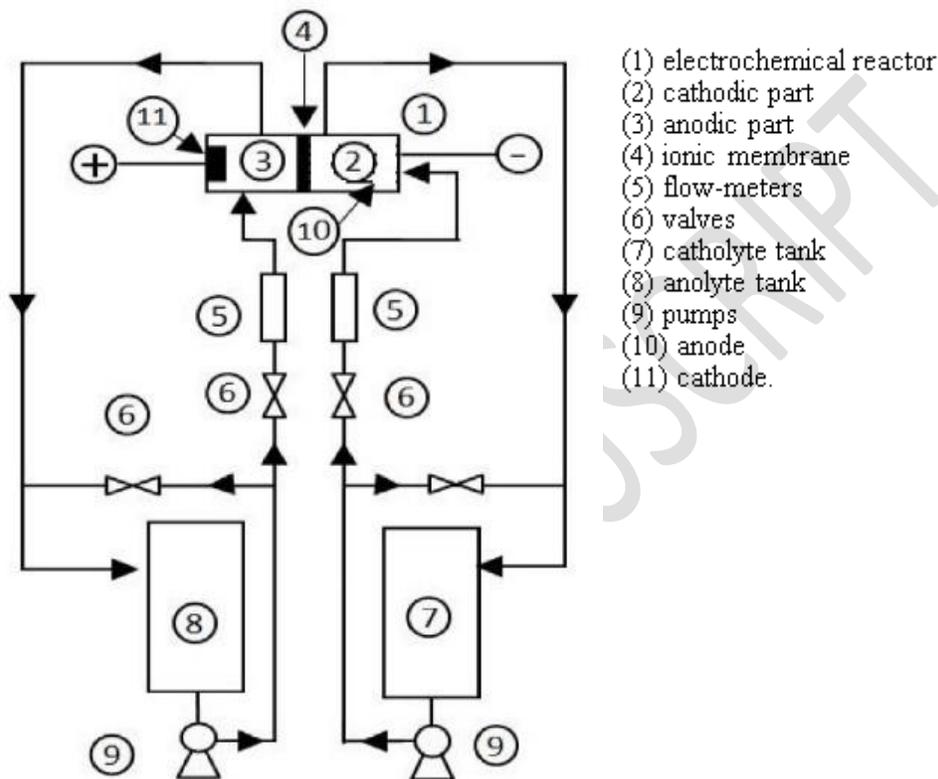


Figure 2. Electrochemical treatment system

Table 1. The specifications of Al tubes bundle

Property	Value
Length	11 cm
Outer diameter	0.6 cm
Inner diameter	0.4 cm

3. Results and discussions

3-1. Impact of concentration

Three solutions with different dye initial concentration (150, 200 and 300 mg/L) have been prepared with 0.5 molarity supporting electrolyte concentration. Experiments have been conducted

at pH 7, 2 L/min volumetric flow rate. The concentration of remained methyl and COD have been determined and plotted versus time.

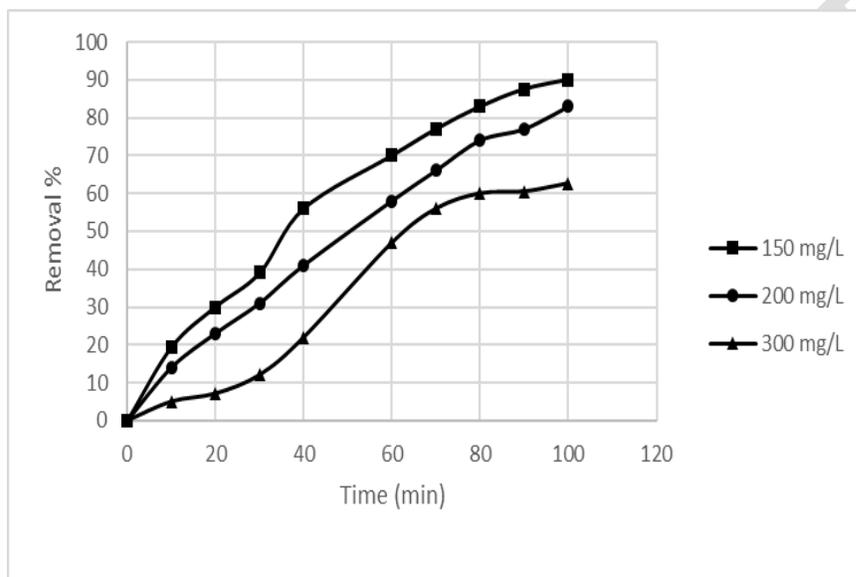


Figure 3. Impact of methyl blue concentrations on removal percentage

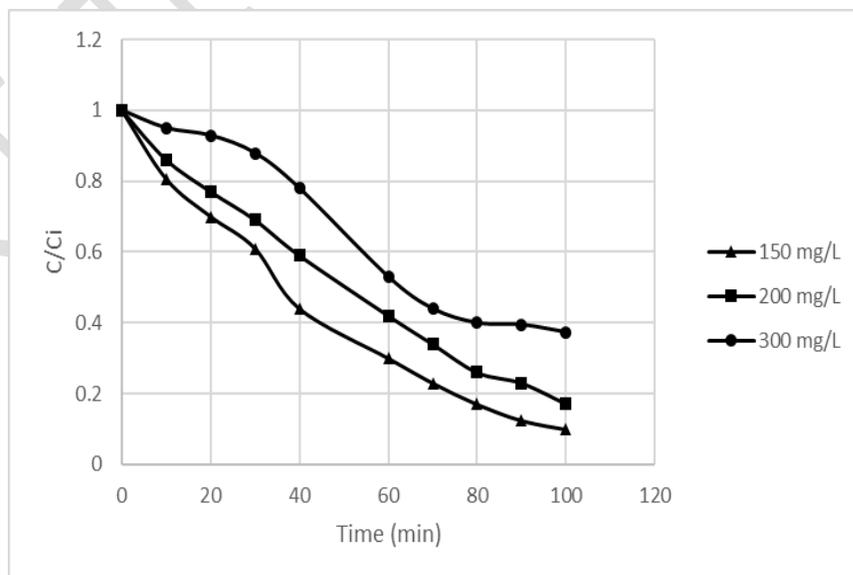


Figure 4. Impact of methylene blue concentrations on concentration decay

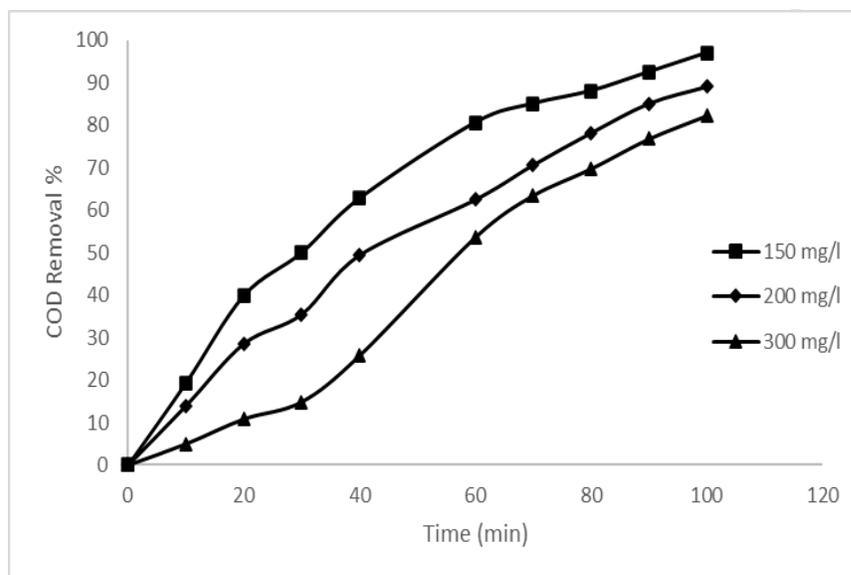


Figure 5. Impact of methylene blue concentrations on COD removal

Figures 3, 4 and 5 shows the impact of initial dye's concentration on removal efficiency. It is clear that the increase in dye concentration will reduce the removal efficiency since the amount of oxidizing agents formed depends on the supporting electrolyte concentration, so the increase of dye concentration will need more oxidizing agent and hence reduce removal efficiency.

3-2. Impact of molarity of the electrolyte

In this set of experiments, different sodium chloride electrolyte concentrations have been adopted (0.5, 1, 1.5 and 2 M). All the experiments have been conducted at 150 mg/L dye concentration, 2 L/min flow rate, 100 mA treatment current.

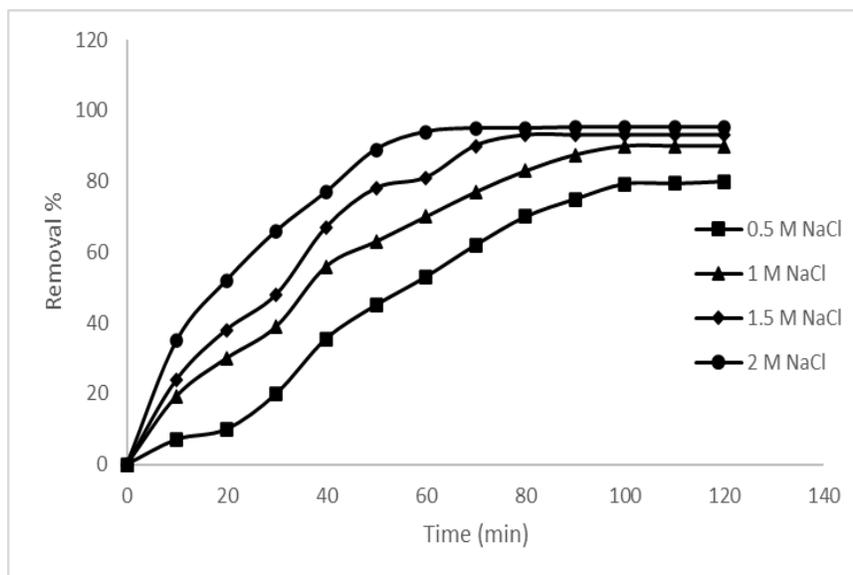


Figure 6. Effect of molarity on removal percentage

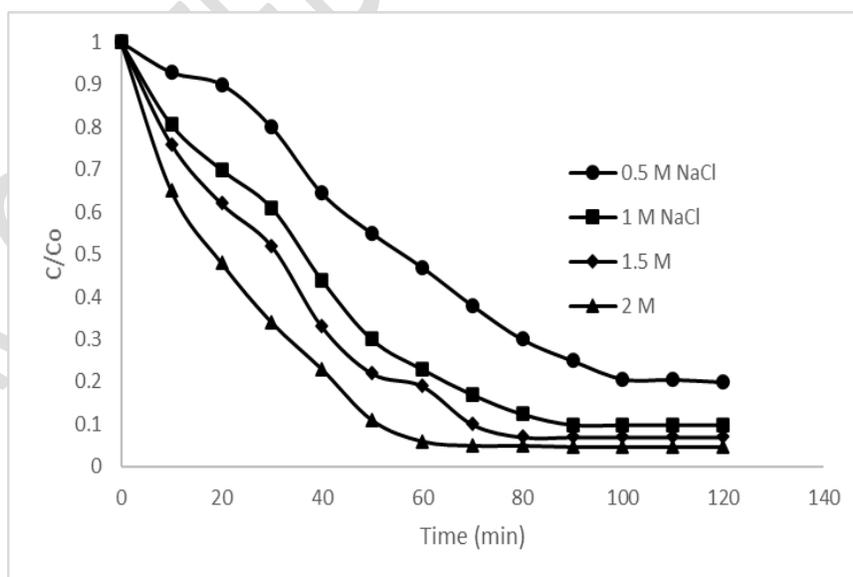


Figure 7. Effect of molarity on concentration decay

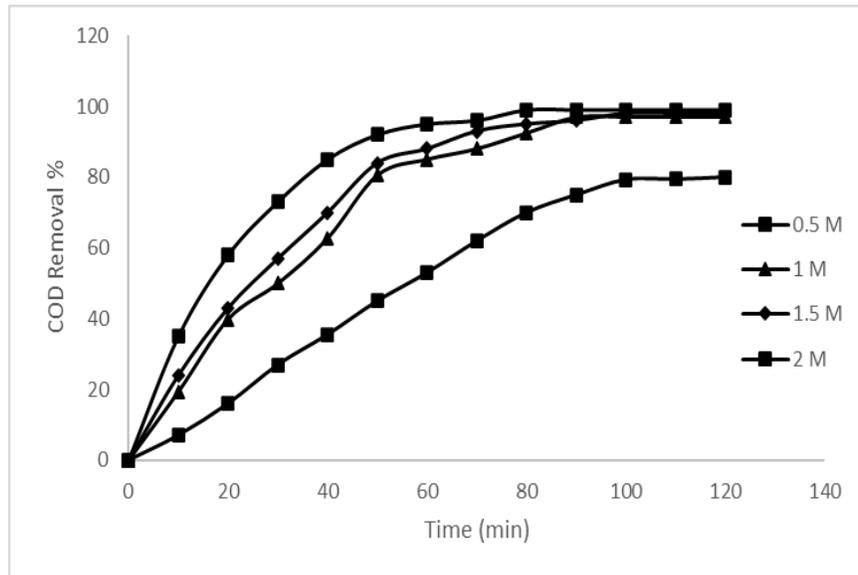


Figure 8. Effect of molarity on COD removal percentage

Effects of supporting electrolyte solution are shown in **Figures 6, 7, and 8** respectively. Because there are more powerful oxidizing agents (chlorine and HClO), which are thought to be the primary dye degradation agents, it is evident that the removal efficiency rises as the NaCl concentration does. In addition, solution conductivity increases as a result of molarity's increasing and hence decreases the resistance to current pass through the solution.

3-3. Impact of treatment current

Treatment current's impact has been studied by applying different values of treatment current (25, 100, 200 mA) at pH 7, 2 l/min flow, 0.5 M NaCl, rate and 150 mg/l dye concentration.

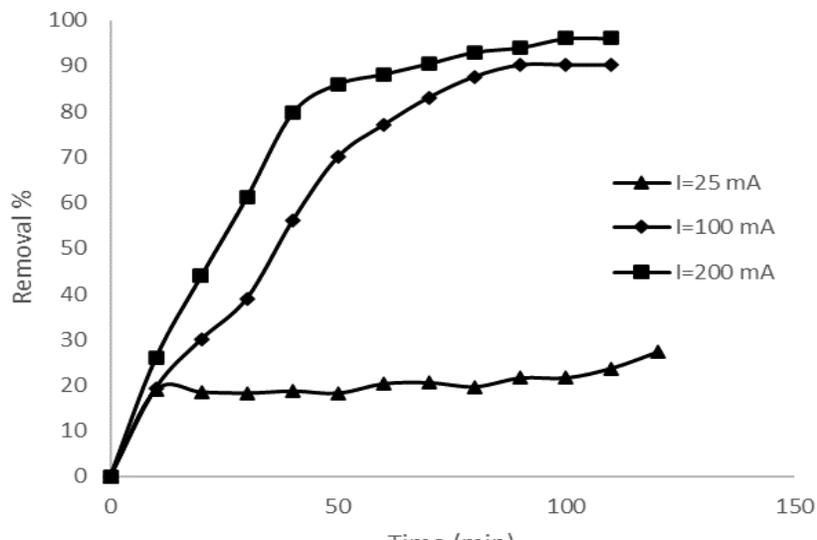


Figure 9. Impact of treatment current on removal percentage

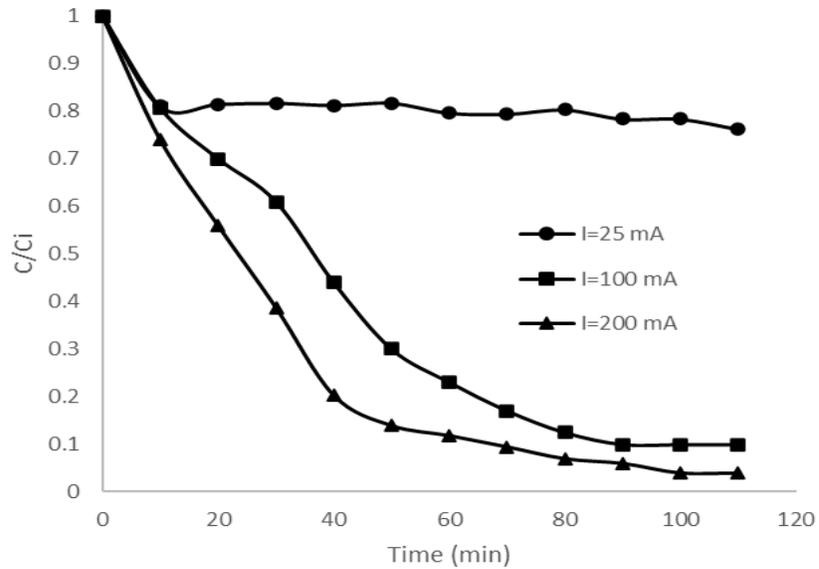


Figure 10. Impact of treatment current on concentration decay

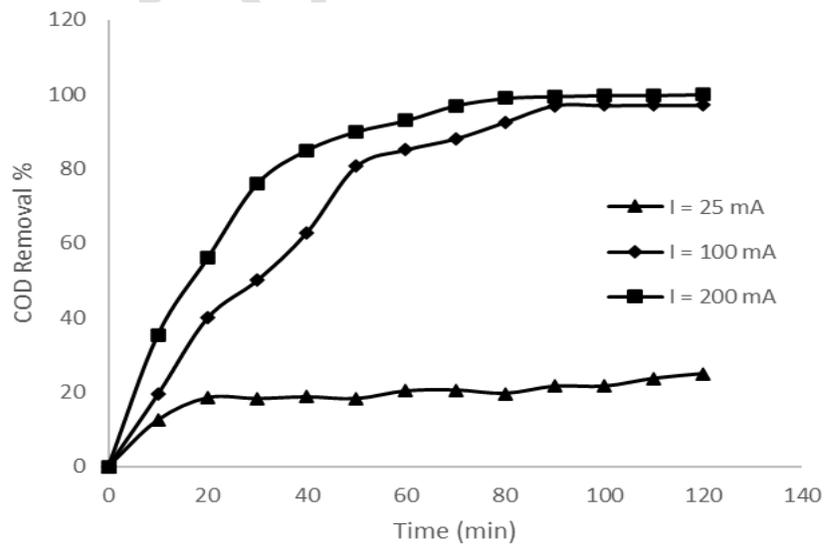


Figure 11. Impact of treatment current on COD removal-percentage

From the **Figures 9, 10 and 11**, It is clear that increasing the current greatly improves removal effectiveness, particularly between 25 and 100 mA.

Amount of hydroxyl and chlorine ions increases by increasing the applied current, and these compounds are considered as the main incineration agent for dye degradation, increasing the treatment current to a value higher than 100 mA, the oxygen evolution reaction will be initialized leading to reduction in the removal efficiency.

3-4. Impact of pH

The effects of pH have been investigated for 0.1 M NaCl, 100 mA applied current, and 2 l/min flow rate at various pH values (2, 7, and 12).

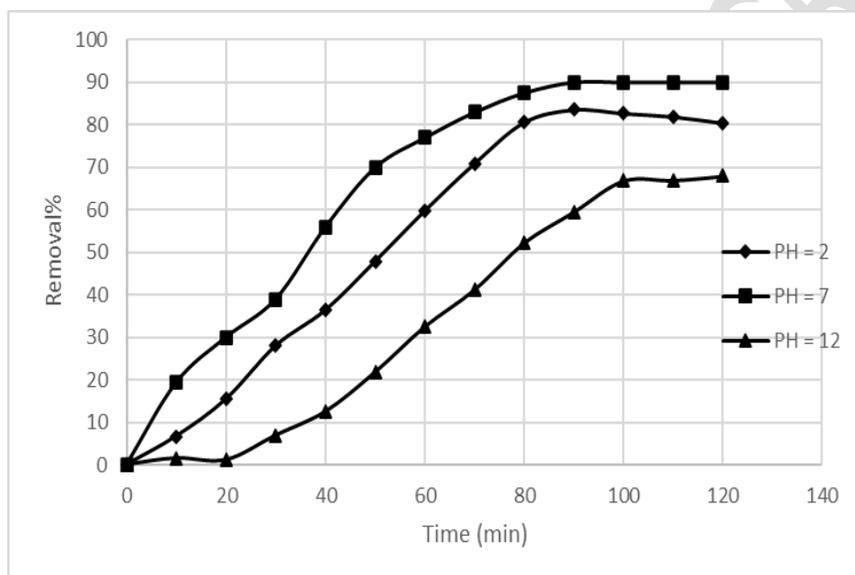


Figure 12. Impact of pH on removal percentage

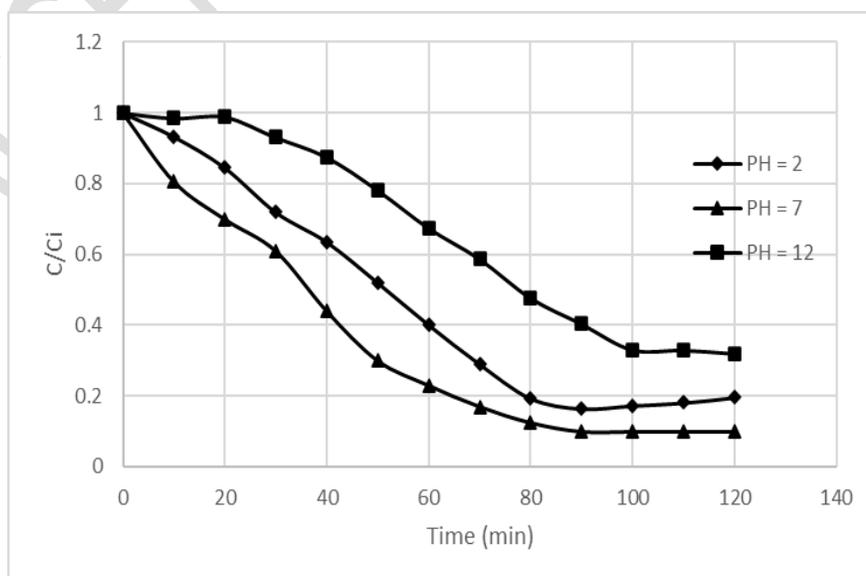


Figure 13. Impact of pH on concentration decay

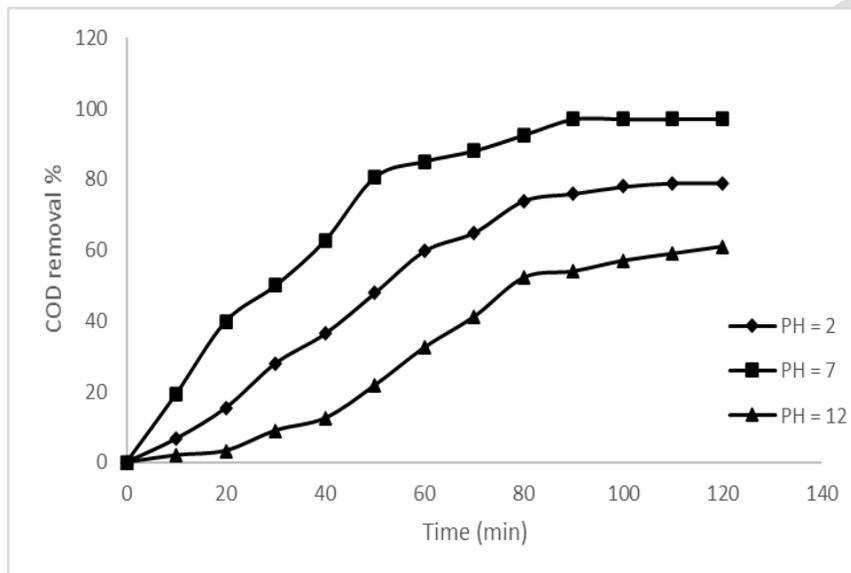


Figure 14. Impact of pH on COD removal

As shown in the **Figures 12, 13 and 14**, it is clear that the efficiency is higher at pH range from 2 to 7, and lower for pH values higher than 7. This is due to the nature of methylene blue dye which can exist as quinoid or azo structure. Quinoid structure exists at low pH which is characterized with hydrogen atom attached to the dye molecule, while at high pH (higher than 7), the hydrogen atom will be detached from the dye structure to convert it into azo form.

When pH is near 7, the predominant structure will be azo which has a cleavage that makes the dye degradation easier, at the same time the degradation reaction will be predominant on the oxygen evolution reaction. When pH increases more than 7, oxygen formation reaction will be the main leading to lower the efficiency. At the same time, increasing the alkalinity will decrease the solution conductivity leading to spoil in the applied current and hence decreases the removal efficiency.

3-5. Impact of flow rate

Impact of flow rate has been studied at different flow rates (2, 4, 6 l/min) when the initial dye concentration affixed at 100 mg/l, supporting electrolyte molarity is 0.1 M and pH 7.

It is clear in **Figures 15, 16 and 17** that the increase in solution flow rate is significantly increasing the removal efficiency due to reduction in the diffusion layer thickness and hence increasing the rate of molecules transfer towards the anode. At the same time, the increase in the solution flow rate will enhance the amount of the active oxidizing agents and increases the dispersion of these agents via the act of turbulence leading to higher contact area among the dye molecules and the oxidizing agents.

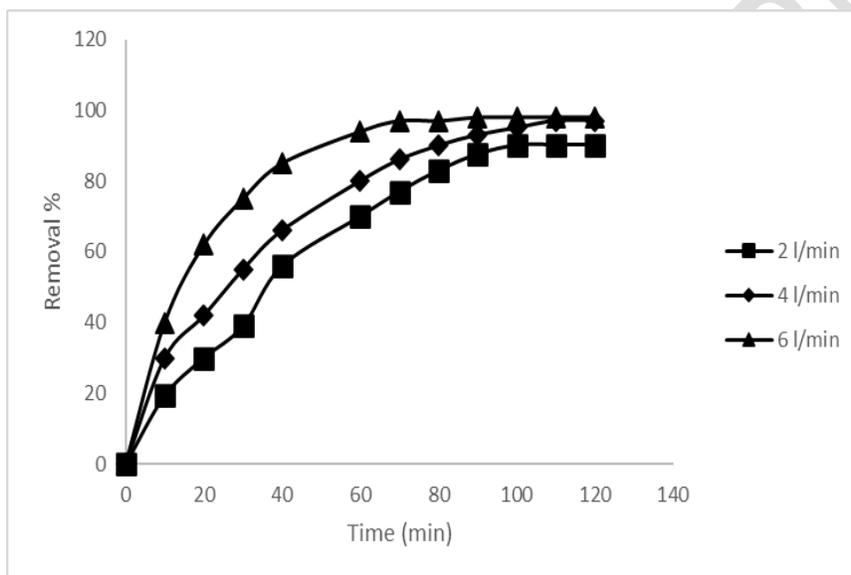


Figure 15. Impact of flow rate on removal percentage

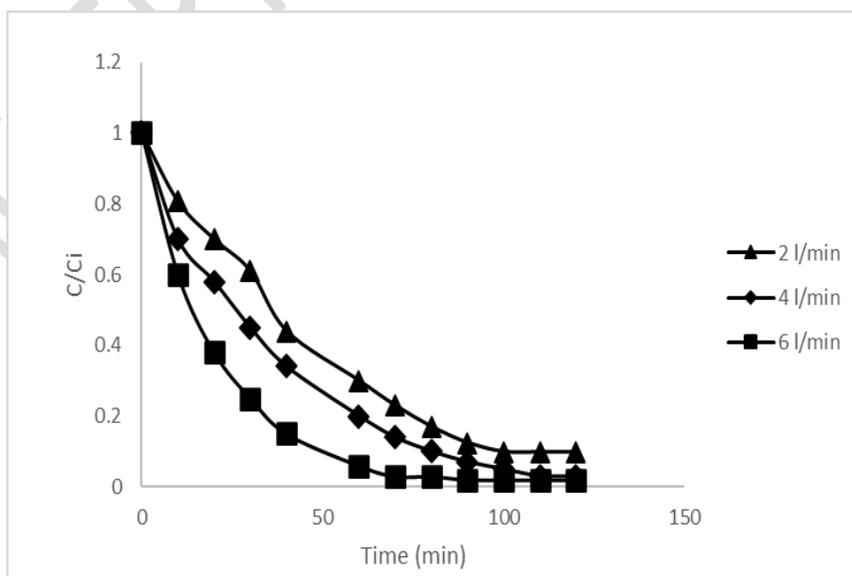


Figure 16. Impact of flow rate on concentration decay

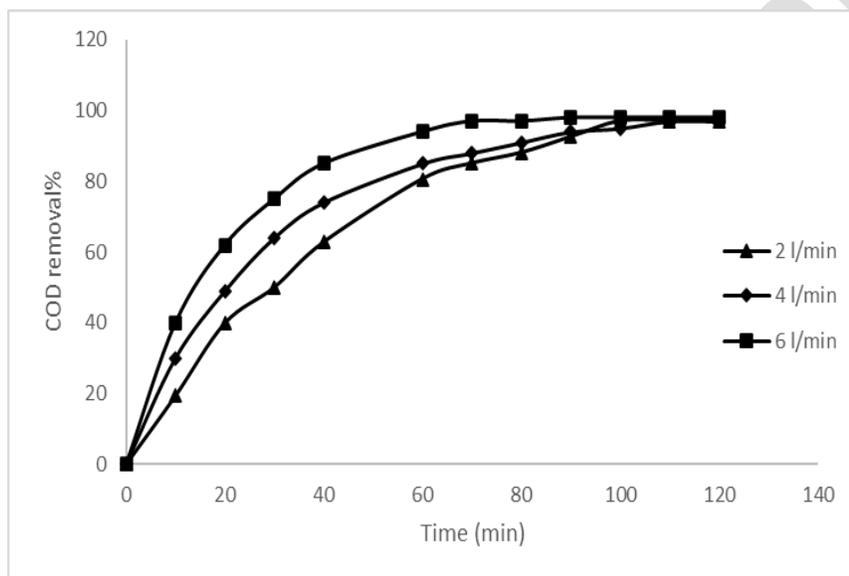


Figure 17. Impact of flow rate on COD removal percentage

4. Conclusion

This work emphasizes the role of electrochemical oxidation, which has been recognized as a flexible and effective technique for dye removal in wastewater treatment. An electrochemical reactor operating in batch-circulation flow-through mode was utilized for the degradation of methylene blue. The results indicate that increasing the dye concentration decreases the removal efficiency, reaching 90% at 150 mg/L, 83% at 200 mg/L, and 63% at 300 mg/L. Increasing the molarity of the supporting electrolyte enhances the removal efficiency from 80% to 95% when the molarity increases from 0.5 to 2. In addition, the applied current is a significant factor affecting the removal efficiency. As the treatment current increases 25 - 200 mA, the efficiency of removal

increases from 27% to 96%. Furthermore, the elimination efficiency rises from 90% to 98% when the electrolyte flow rate ranges from 2 to 6 L/min. The pH value also influences the removal efficiency, showing a positive effect within the range of pH 2 to pH 7, where the removal efficiency increases from 79% to 97%. However, the removal efficiency decreases when the pH exceeds 7. Overall, the findings support the potential of electrochemical oxidation as an effective method for improving the quality of textile wastewater and contributing to sustainable water management.

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