

*Original Article*

## Influence of quadrupole-needle direct corona discharge on degradation of methylene blue in water

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### Abstract

This study focused on the performance of a novel quadrupole-needle direct corona discharge on degradation of methylene blue (MB) in water. MB was used as a model pollutant due to its stability, strong color, and common presence in textile wastewater. The system generated a stable plasma at 25°C, with an electrode gap of 1 cm, producing hydroxyl radicals, which played a key role in MB degradation through oxidation and fragmentation. The plasma device was driven at 15.8 kV (peak to peak) with 10 kHz DC pulses. The results indicated that plasma treatment effectively reduce MB concentration from 50 ppm to 0 ppm within 8.8 min. The experiments further demonstrated that an 8.8-min plasma treatment significantly impacted MB degradation, having 78.5% efficiency and 7.29 g/kWh energy yield. These preliminary results suggest a possible pathway for wastewater treatment.

**Keywords:** multi-needle direct corona discharge plasma, advanced oxidation process, methylene blue, energy yield, wastewater, degradation efficiency

### 1. Introduction

In the present day, the industrial and agricultural sectors have undergone a revolution, resulting in a substantial amount of wastewater. The release of this wastewater into the environment is the most critical issue affecting the ecosystem. Raising animals for food processing has led to various pollution issues, including wastewater, unpleasant odors, and flies, which serve as breeding grounds for various pathogens and parasites. For instance, when wastewater enters the ground or surface water, it depletes the oxygen, suffocating and killing fish and other aquatic life. The pollutants accumulating in land and water environments ultimately cycle back into the food chain and affect humans. This problem has a significant impact on the quality of water sources used for making potable water, thus being a crucial issue that must be

addressed (Akpor & Muchie, 2011; R, Thirumoorthy, Banu, & Yeom, 2010; Shahbuddin, 2018).

To prevent environmental degradation, it is necessary to employ appropriate methods for treating wastewater before discharging it. Slaughterhouse wastewater (SWW) is scandalously polluted, with high levels of biological oxygen demand, chemical oxygen demand, total suspended solids, blood, and nutrients such as nitrogen and phosphorus, from slaughtering and cleaning activities (Shahbuddin, 2018; Bustillo-Lecompte, Mehrvar, & Quiñones-Bolaños, 2016). The presence of blood and a complex mixture of fats, proteins, and fibers in SWW contributes to the content of organic matter, which can induce algal blooms (Paulista, Presumido, Theodoro, & Pinheiro, 2018), as noted by Rajakumar *et al.* (2010). Recently, the demand for poultry meat has risen significantly in Thailand, with poultry processing consuming a considerable amount of water, about 26.5 liters per bird. According to statistics from the Office of Agricultural Economics, Thailand (2020), meat production reached 2,126,947 tons in 2019, increasing by

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about 15% to 2,503,961 tons in 2020. Therefore, to safeguard nature against catastrophic pathogenic bacterial outbreaks and prevent further environmental damage, it is essential to treat slaughterhouse wastewater (SWW) before discharging it into the environment (Sobsey, Khatib, Hill, Alocilja, & Pillai, 2006).

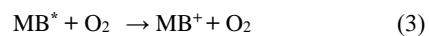
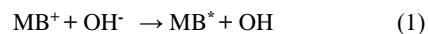
Methylene blue (MB) is a dye with characteristics that make it a hazardous wastewater contaminant, posing risks to human health and environmental safety due to its toxic, carcinogenic, and non-biodegradable nature. Theoretically, MB exists as a solid, odorless, dark green powder at room temperature and forms a blue solution upon dissolution in water (Chakraborty, Ray, & Basu, 2016; Wijaya, Andersan, Permatasari Santoso, & Irawaty, 2020). The molecule's width is approximately 9.5 angstroms while its length ranges from 13.82 to 14.47 angstroms (Jia, Tan, Liu, & Gao, 2018). It exhibits a solubility of 43.6 g/L in water at 25°C, and its melting point falls within the range of 100-110°C (Nasrullah *et al.*, 2015).

Research on the UV interactions with methylene blue (MB) is crucial for understanding its photodegradation and adsorption processes, as nearly all computations rely on its UV-visible spectra. The absorption spectra of MB show a highly pronounced monomer peak at approximately 664 nm and a shoulder peak at 612 nm (Mondal, De Anda Reyes, & Pal, 2017). Moreover, in the ultraviolet region, peaks around 292 nm and 245 nm correspond to substituted benzene rings (Yang *et al.*, 2017). To preserve the environment, effective and innovative methods for the degradation of MB solution should be developed.

Advanced oxidation processes (AOPs) based on hydroxyl radical ( $\cdot\text{OH}$ ) production effectively degrade and mineralize recalcitrant organic matter in wastewater (Khan *et al.*, 2020; Zhang, Jia, Lyu, Liang, & Lu, 2019). Traditional AOPs include cavitation, ozone, hydrogen peroxide, Fenton process, and plasma technology, with novel applications in industrial manufacturing, biomedicine, agriculture, and other industries (Khan *et al.*, 2020; Zhang *et al.*, 2019). These processes are crucial for converting organic compounds into smaller molecules.

The plasma-based oxidation method is widely recognized as one of the most efficient techniques for wastewater treatment, in comparison to other traditional AOPs (Sgroi, Anumol, Vagliasindi, Snyder, & Roccaro, 2021). Cold atmospheric plasma (CAP) is known as the fourth state of matter, consisting of ionized gas (Ishijima, Sugiura, Saito, Toyoda, & Sugai, 2009). CAP is highly effective in generating and controlling reactive oxygen and nitrogen species (RONS) (Kogoma & Okazaki, 1994; Luque A & Ebert, 2008). The physical effects such as high-intensity ultraviolet radiation and shockwaves create chemical RONS, which include hydroxyl ( $\text{OH}$ ), singlet oxygen ( ${}^1\text{O}_2$ ), superoxide ( $\text{O}_2^-$ ), hydrogen peroxide ( $\text{H}_2\text{O}_2$ ), nitric oxide ( $\text{NO}$ ), nitrogen dioxide ( $\text{NO}_2$ ), among others (Fan *et al.*, 2021). The traditional water treatment techniques such as ultrasonication, coagulation, adsorption, and chlorination have some drawbacks in eliminating all harmful contaminants from water. For example, chlorination of organic materials in water generates carcinogenic compounds, while UV-radiation has a low penetration efficiency, and filtration cannot remove viruses (Banach, Hoffmans, Appelman, van Bokhorst-van de Veen, & van Asselt, 2021; Shi *et al.*, 2021). Therefore, it is

essential to enhance wastewater treatment methods to effectively reduce harmful and toxic substances from effluents. Considering potential alternative techniques, multi-needle direct corona discharge is a promising source of reactive species capable of reducing wastewater contamination and eliminating pollutant molecules. In brief,  $\text{OH}^-$  from plasma can react with  $\text{MB}^+$  by reduction to form  $\text{OH}$  radical. Then,  $\text{OH}$  reacts with  $\text{OH}$  to produce  $\text{H}_2\text{O}_2$ , which is a crucial active species in the degradation process. In the same way,  $\text{O}_2$  reacts with excited  $\text{MB}^*$  radicals and forms  $\text{O}_2^-$ . These processes are expressed in Equations (1)-(3), according to (Soltani & Entezari, 2013).



Most of the mechanisms in degradation of MB proceed by the breakage of the aromatic ring of the MB dye. The fragmented components of dyes undergo degradation into additional reaction intermediates, which include amines, phenols, carboxylic species, and aldehydes (Din, Khalid, Najeeb, & Hussain, 2021). Typically, the important radicals from plasma such  $\text{O}_2^-$  and  $\text{OH}$  initiate redox reactions and degrade MB dye into organic compounds or inorganic ions. As a result, a colorless solution forms due to the fragmentation of aromatic rings (Dagher *et al.*, 2018; Kuila *et al.*, 2020; Yang *et al.*, 2019).

In this study, the efficiency of the plasma reactor in treating wastewater was evaluated by measuring the degradation of MB dye within the plasma treated solution. The impacts of time duration, oxidation-reduction potential, and electric conductivity on the MB degradation efficiency were investigated.

## 2. Materials and Methods

### 2.1 A quadrupole-needle direct corona discharge device

A quadrupole-needle direct corona discharge (QDCD) device was used (Figure 1). The experimental setup comprises a discharge chamber and measurement systems. As the wastewater model, the QCDC was designed to activate water for degrading methylene blue (MB). Its apparatus consisted of 4 needles of tungsten, 150-mm long with a 3.7 mm diameter, and was sharply tapered at the nozzle and covered with an outer case of nylon material. A high-voltage flyback transformer providing 15.8 kV (peak to peak) at 10 kHz-DC pulses was applied to quadruple metallic external electrodes. The HV probe (Tektronix, P6015A, 75 MHz) and PEARSON® current probe were used to determine the voltage ( $U$ ) and current ( $I$ ) applied to the QCDC reactor, respectively. To determine the dissipated power ( $P$ ), we integrated the average  $U(t) \cdot I(t)$  over the period ( $T$ ) as follows.

$$P = \frac{1}{T} \int_0^T U(t) \cdot I(t) dt \quad (4)$$

## 2.2 Effect of QCDC plasma on MB concentrate degradation

The study investigated the impacts of various parameters, including treatment duration, and MB solution concentration, on the efficacy of the QCDC system. Regarding the materials, MB dye ( $C_{16}H_{18}ClN_3H_2O(x=2-3)$ , wt%) was purchased from the QRëCTM company, with deionized water used as solvent. To elaborate, a magnetic stirrer bar was placed in a beaker containing 30 ml of 10 ppm MB solution, and the temperature was set at room temperature (25 °C) on a magnetic hotplate, as illustrated in Figure 1. The treatment duration varied between 0 and 125 sec in increments of 25 sec. The pH of the solution was measured using a pH meter (HI1131, HANNA instrument), and the conductivity of the solution was measured using a conductivity meter (DIGICON, CD-439SD), respectively. Next, 3 ml of the MB solution was transferred to cuvettes, and the absorbance was measured at 664 nm using a UV-Vis spectrophotometer (U-3900H HITACHI) to determine the MB degradation efficiency ( $\eta$ ) as follows (Wu *et al.*, 2019):

$$\eta = \frac{C_0 - C_t}{C_0} \times 100 \quad (5)$$

To calculate the MB degradation efficiency, two concentrations are required: the initial concentration of MB ( $C_0$ ) and the concentration of MB at time  $t$  ( $C_t$ ). The energy yield of the MB degradation can be determined using the following equation (Balu, Uma, Pan, Yang, & Ramaraj, 2018; Saeed, Khan, & Park, 2015):

$$Y(\text{g/kWh}) = \frac{c(\text{g/l}) \times V(\text{l}) \times \frac{1}{100} \times \eta(\%)}{P(\text{kW}) \times t(\text{h})} \times 100 \quad (6)$$

Here  $c$  is the initial concentration,  $V$  represents the volume of MB solution (0.1 liter) solution,  $\eta$  is the degradation efficiency at  $t$  time, and  $P$  is input power.

## 3. Results and Discussion

### 3.1 Effect of treatment duration on MB concentration

The innovative QCDC plasma device was developed for use in wastewater treatment in a small scale model. This device is composed of a quadrupole needle to expand the treatment surface for maximum efficiency. The effect of time duration on both absorbance and color visualization in a 10 ppm MB solution was investigated using QCDC at 137.8 mW as the activation tool. Figure 2a displays the absorbance versus wavelength plot for 2 min. The findings indicate that the absorbance at 668 nm gradually decreased with treatment duration, showing values of 0.64, 0.23, 0.091, 0.045, 0.023, and 0.022 a.u. for 0, 25, 50, 75, 100, and 125 sec, respectively. Notably, the treatment duration of 125 sec resulted in the oxidation of the solution, changing it from blue to colorless, as observed in the small picture in the left corner. To examine the impact of QCDC plasma on MB

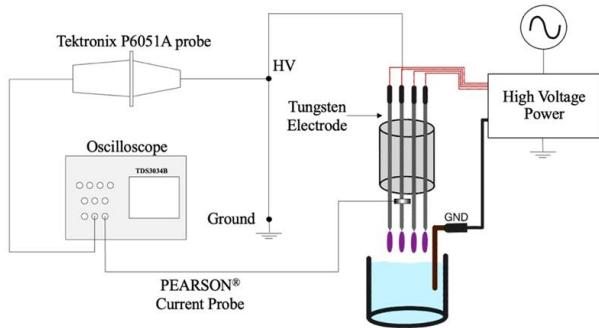
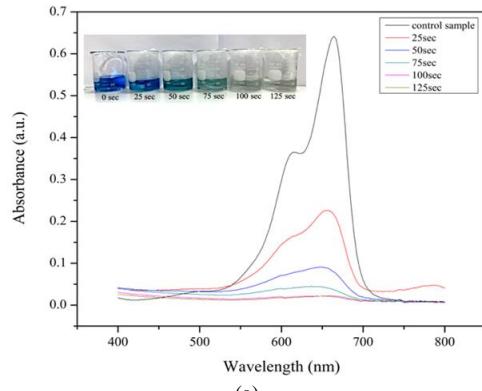


Figure 1. Illustration of the QCDC plasma configuration operated by DC pulse



(a)

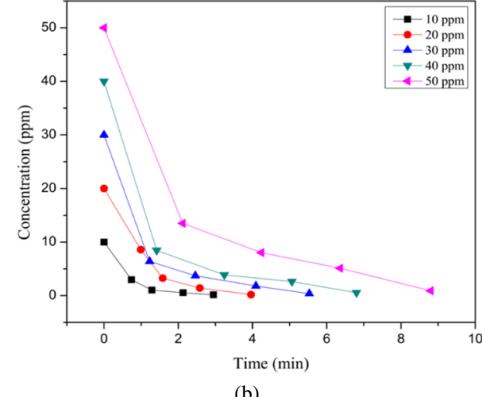


Figure 2. (a) The absorbance and the colors of MB dye solutions for the control sample and plasma treatment time of 25, 50, 75, 100, and 125 seconds, respectively, and (b) the effect of QCDC plasma on the initial concentration of MB dye solutions of 50, 40, 30, 20, and 10 ppm after treatment time for 10 min.

concentration, solutions with initial concentrations of 10, 20, 30, 40, and 50 ppm were subjected to the optimal plasma condition. Then, the samples were measured for MB concentration by determining absorbance at 668 nm using the UV-Vis spectrophotometer. The discoloration of the MB solutions was observed to decrease significantly from their initial levels, with values of 0.16, 0.19, 0.37, 0.56, and 0.92

ppm for treatment durations of 2.9-, 3.9-, 5.5-, 6.8-, and 8.8-min, respectively. It was discovered that the duration required to degrade the MB concentration was dependent on the initial concentration, with discoloration occurring after 2.9- and 8.8-min plasma treatment for 10 and 50 ppm, respectively. Previous studies have demonstrated that direct arc plasma has potential in wastewater treatment (Dzinun, Ichikawa, Mitsuhiro, & Zhang, 2020; Samuel & Yam, 2020). In brief, the direct effect resulting from electron collisions and other impacts generated ions and radicals ( $\text{OH}$ ,  $\text{O}_2^-$ , and  $\text{O}_2$ ) via photolysis and pyrolysis reactions (Lukes, Locke, & Brisset, 2012), and these were utilized for purification in wastewater treatment. However, the QCDC system has demonstrated a significant reduction in treatment time for MB degradation compared to traditional plasma systems. The previous studies using direct arc plasma and dielectric barrier discharge plasma have reported treatment durations ranging from 10 to 30 min for similar pollutant concentrations, often requiring higher energy inputs and facing challenges such as the secondary recombination of radicals (Dzinun *et al.*, 2020; Lukes, Locke, & Brisset, 2012; Samuel & Yam, 2020). Thus, the QCDC system not only achieves faster treatment times but also offers an energy-efficient alternative for wastewater treatment.

The effect of the QCDC plasma device on MB degradation depends on various parameters, with initial concentration and time duration being crucial factors to consider. According to Lihang Wu *et al.* (Wu *et al.*, 2019), plasma treatment with a longer duration can effectively degrade MB dye solutions. In addition, the chemical kinetics of degradation in MB solution also need to be considered. According to a study by Zuhuela *et al.* (2021), the 1<sup>st</sup> order and 2<sup>nd</sup> order reaction models and rate constants can be expressed as follows.

$$\text{1}^{\text{st}} \text{ order} \quad \ln\left(\frac{C_t}{C_0}\right) = -kt \quad (7)$$

$$\text{2}^{\text{nd}} \text{ order} \quad \frac{1}{C_t} - \frac{1}{C_0} = -kt \quad (8)$$

Here  $C_0$  is the initial MB concentration,  $C_t$  is the MB concentration at a certain time, and  $k$  is the reaction rate constant. To study the performance of QCDC plasma device in degradation of MB solution, the 1<sup>st</sup> order reaction equation was plotted to a linear graph with  $\ln(C_t/C_0)$  against time ( $t$ ) for MB dye solutions at concentration of 50, 40, 30, 20, and 10 ppm as shown in Figure 3a. The results showed coefficients of determination  $R^2$  as 0.9773, 0.9856, 0.9481, 0.7333, and 0.9388 for linear fits to data from 10, 20, 30, 40, and 50 ppm cases. Remarkably, the 20 ppm initial MB concentration gave the best linear fit. A higher value of  $R^2$  indicates that the independent variables in the regression model can explain more of the variance in the dependent variable. In addition, the slope of the linear fit ( $y=mx+b$ ) reveals the reaction rate constant. The reaction rate constants obtained from the fitting of 2<sup>nd</sup> order reaction models, as shown in Figure 3b, are 1.90, 1.24, 0.41, 0.22, and 0.11 for 10, 20, 30, 40, and 50 ppm, respectively.

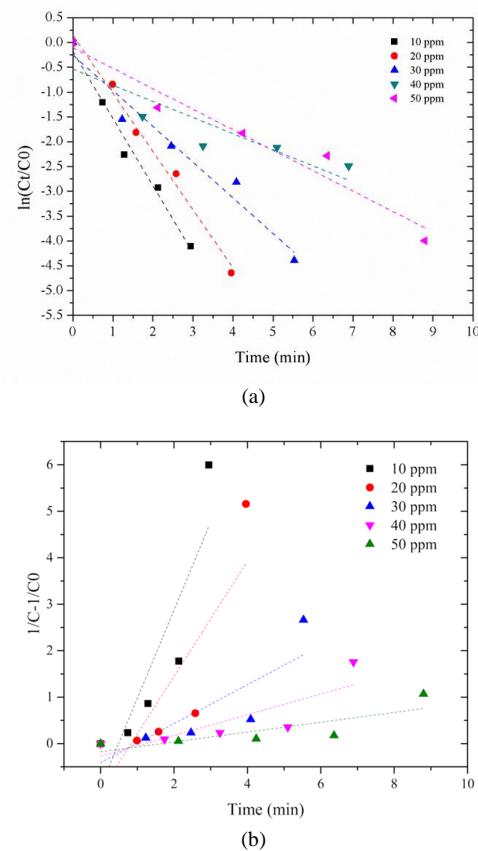


Figure 3. (a) Illustration of  $\ln C_t/C_0$  versus time in the 1<sup>st</sup> order reaction for MB dye solutions at concentration of 50, 40, 30, 20, and 10 ppm, and (b) a linear graph of 2<sup>nd</sup> order reaction with  $\ln(1/C_t/C_0)$  against time for MB dye solutions at concentration of 50, 40, 30, 20, and 10 ppm.

As in a prior study, the second-order reaction rate model is appropriate, as the rate is influenced by the  $\text{OH}$  concentration and  $\text{O}_3$  produced during the treatment with QCDC plasma. Consequently, the proposed first-order and second-order reaction rate laws for MB degradation using QCDC plasma are represented as follows (Reddy, Bendi, Karuppiah, Linga Reddy, & Challapalli, 2013; Zuhuela *et al.*, 2021).

$$\text{Reaction rate} = k[\text{OH}] \quad (9)$$

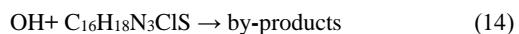
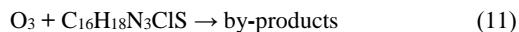
$$\text{Reaction rate} = k[\text{OH}][\text{O}_3] \quad (10)$$

### 3.2 Effect of QCDC plasma on the pH of the MB dye solution

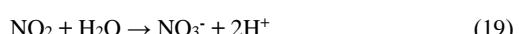
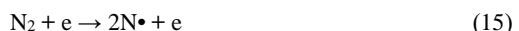
The study aimed to assess the impact of plasma treatment on the pH level of MB solution, considering the diverse pH levels of actual dying wastewater, which range from alkaline to acidic. The anticipated effect of plasma-dissipated power on treatment duration was also examined at 137.8 mW. Figure 3 displays the pH values obtained for initial MB solution concentrations of 10, 20, 30, 40, and 50 ppm,

following plasma treatments for 2.9-, 3.9-, 5.5-, 6.8-, and 8.8-min, respectively. The results revealed pH levels of 2.7, 2.6, 2.6, 2.5, and 1.8 for these cases.

The previous study by Pavithra *et al.* (2019) emphasized the crucial role of pH as an important parameter in wastewater treatment. Specifically, the efficiency of MB dye solution degradation was closely linked to pH variations. Figure 4 illustrates a significant reduction in pH within the MB dye solution. Particularly, at an initial concentration of 50 ppm, the pH was the lowest after a 4-min treatment compared to other initial concentrations. Basically, MB dye solution is slightly alkaline, so the pH usually ranges from around 6 to 8. However, the reactive species in PAW can oxidize MB, potentially leading to decolorization and structural changes. Moreover, the pH level in the MB solution is affected by the oxidation pathways involving OH and O<sub>3</sub>. The first mechanism is the direct reaction of O<sub>3</sub> with the reactant, whereas the second involves the decomposition of O<sub>3</sub> with intermediate species. Subsequently, the product OH reacts with the reactant, promoting its degradation as shown in the following equations (Jaramillo-Sierra *et al.*, 2019).



According to Jaramillo-Sierra *et al.* (2019), the decomposition of O<sub>3</sub> is influenced by pH, with alkaline conditions accelerating its breakdown. Hence, a low pH also indicates low efficiency in the degradation of MB solution, which agrees with our results in Figures 4 and 6. In addition, the low pH levels originate from specific acidic compounds like nitric and nitrous acid, which are believed to form from nitrogen gas and water vapor in the air during the discharge process, as shown by the following equations (Huang, Chen, Wang, & Yan, 2010; Fahmy *et al.*, 2018; Manoj Kumar Reddy, Rama Raju, Karuppiah, Linga Reddy, & Subrahmanyam, 2013).



Compared to previous studies, the QCDC plasma system achieved a faster reduction in pH and higher degradation efficiency within a shorter treatment time. This suggests that non-thermal plasma can accelerate MB degradation more efficiently than conventional ozone or UV-based systems, making it a promising alternative for wastewater treatment applications as shown in Table 1.

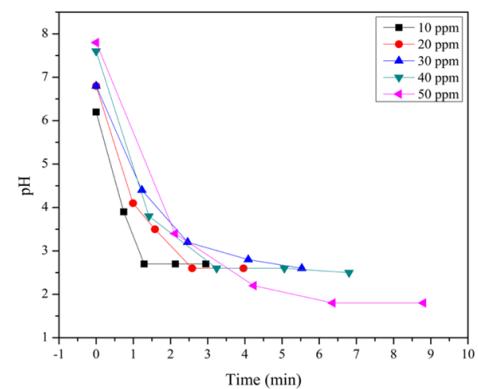


Figure 4. The effect of QCDC plasma on the pH value inside the initial concentration of MB dye solutions of 50, 40, 30, 20, and 10 ppm after treatment time with 10 min.

### 3.3 Effect of QCDC plasma on the electrical conductivity of MB dye solution

Electrical conductivity (EC) is a crucial parameter for investigating the formation of dissolved ions and other substances during the degradation of actual dying wastewater (Zhang, Chen, Wang, Wu, & Hu, 2020). It can be utilized to monitor the process. In Figure 4, the change in conductivity during the MB dye degradation process is displayed, along with the MB dye degradation efficiency under various initial solution conductivities. An increase in solution conductivity was continuously observed throughout the experiment, attributed to the formation of acidic substances and degradation of intermediates via plasma. The results revealed EC values of 0.57, 0.68, 0.81, 1.05, and 1.53 mS for treatment durations of 2.9-, 3.9-, 5.5-, 6.8-, and 8.8-min, respectively. In addition, EC also needs to be considered in the discharge plasma process. Typically, a high EC of the solution influences the generation of a large number of radicals (•OH, O<sub>2</sub><sup>-</sup>, O<sub>3</sub>), which are essential for the oxidative breakdown of organic pollutants (Tichonovas *et al.*, 2013). As shown in Figure 5, the EC increased both with the initial MB dye concentration and with the time duration. These findings are consistent with the conductivity variation observed in the MB degradation during DBD treatment, as reported by Liahang Wu *et al.* (2019). On the other hand, Jiang *et al.* (2012) found that the removal efficiency decreased as the medium conductivity increased for methyl orange. However, the concentration of radicals inside the solution, produced from plasma, is controlled by the optimal EC of the solution. Thus, the discharge efficiency and the production of radicals might decrease due to the rapid neutralization of the electric field charge at the plasma head (Sunka *et al.*, 1999).

### 3.4 Effect of initial MB dye concentration on degradation efficiency and energy yield consumption

Figure 6a illustrates the impact of varying initial MB dye concentrations (10, 20, 30, 40, and 50 ppm) on degradation efficiency. Within the first 5 min, the degradation

Table 1. pH reduction and MB degradation efficiency of alternative plasma sources

Study	Plasma type	Initial pH	Treatment time	pH change	MB degradation efficiency
(Pavithra <i>et al.</i> , 2019) (Jaramillo-Sierra <i>et al.</i> , 2019) This Study (QCDC)	Conventional AOP	6.8-7.5	10-15 min	pH↓ (5.0-5.5)	75-85%
	O <sub>3</sub> +UV	7.0	10 min	pH↓ (5.2)	80%
	Non-Thermal Plasma	6.8-7.2	4-8 min	pH ↓ (4.8)	85-95%

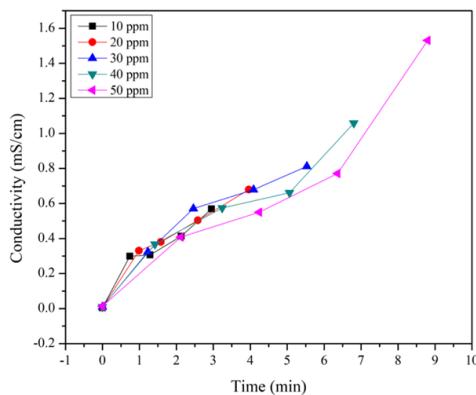
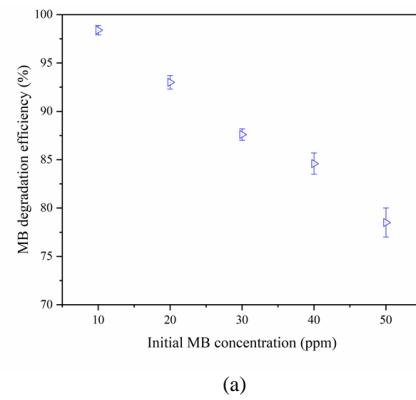


Figure 5. The effect of QCDC plasma on the EC value inside the initial concentration of MB dye solutions of 50, 40, 30, 20, and 10 ppm after treatment time within 10 min.

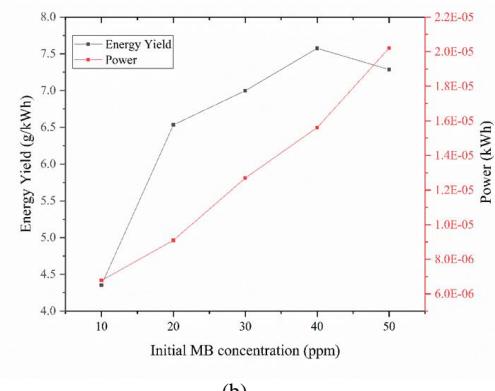
efficiency of MB dye decreased as the initial concentration increased. The results showed degradation efficiencies of 98.4%, 93.0%, 87.6%, 84.6%, and 78.5% for initial concentrations of 10, 20, 30, 40, and 50 ppm, respectively. Nevertheless, regarding the efficiency of the QCDC plasma device, Figure 6b illustrates the energy yield and power consumption versus initial MB dye concentrations (10, 20, 30, 40, and 50 ppm). It is noted that the energy yield and power consumption were 4.36, 6.53, 7, 7.58, and 7.29 g/kWh, and  $6.78 \times 10^{-6}$ ,  $9.10 \times 10^{-6}$ ,  $1.27 \times 10^{-5}$ ,  $1.56 \times 10^{-5}$ , and  $2.02 \times 10^{-5}$  kWh for the initial concentrations of 10, 20, 30, 40, and 50 ppm, respectively.

As a result, the degradation efficiency of the MB dye solution at 10 ppm is significantly higher than that at 50 ppm. This difference may be attributed to the amount of plasma active species formed in the discharge process and the constant energy input, upon which the maintenance of specific concentration levels of MB dye solution depends. Consequently, the degradation efficiency would be reduced at higher pollutant concentrations.

To assess the performance of the QCDC plasma device, the energy yield and power consumption are important parameters that indicate the reactor's potential for treating dyeing wastewater. In Table 2, the effect of degradation induced by QCDC with different plasma systems is also compared. It is noteworthy that the energy efficiency was 7.58 g/kWh under the plasma treatment with a treatment duration of 6.8 min, which was significantly more effective than that reported by other researchers (Nasrullah *et al.*, 2015; Wang *et al.*, 2017; Zhang, Zheng, Qu, & Chen, 2008). For example, the degradation efficiency of MB dye using a double-chamber DBD under different carrier gases was only 0.14 g/kWh (Wang, Dong, Xu, Chi, & Wang, 2017). Next, pulsed corona discharge and ozonation degrade MB in water effectively at around 0.341 g/kWh (Muhammad Arif, Ubaid ur, Abdul, &



(a)



(b)

Figure 6. (a) The impact of QCDC plasma on the degradation efficiency of MB dye solution at initial concentrations of 10, 20, 30, 40, and 50 ppm within a treatment time of 5 minutes, and (b) Energy yield and power consumption of different concentrations of MB dye solution until discoloration process

Kurshid, 2002). Additionally, methyl orange degraded by non-equilibrium plasma has an energy yield of approximately 3.6 g/kWh in the results obtained by Zhang *et al.* (2008).

#### 4. Conclusions

This study highlights the effectiveness of the QCDC plasma device in wastewater treatment, particularly in methylene blue (MB) degradation. The results confirm that increasing electrical conductivity (EC) enhances radical generation ( $\cdot\text{OH}$ ,  $\text{O}_2^-$ ,  $\text{O}_3$ ), significantly influencing degradation efficiency. However, maintaining an optimal EC threshold is essential to prevent radical neutralization and plasma discharge inefficiency, as observed in previous studies on methyl orange degradation. Energy efficiency analysis revealed that the QCDC plasma device achieved a high energy yield of 7.58 g/kWh within 6.8 minutes, outperforming other

Table 2. Comparison of the different plasma techniques in dye degradation

Type of dye		Condition	Energy yield (g/kWh)
Quadrupole-needle direct corona discharge (This study)	Methylene Blue	V0 : 0.03L, C0: 50 ppm, t: 6.8 min, V: 8 kV, F: 9.7 kHz, Ambient air	7.58
Pulsed corona (Muhammad Arif <i>et al.</i> , 2002)	Methylene Blue	V0 : 0.02L, C0: 13.25 ppm, t:20 min, V: 40 kV, F: 60 kHz, O <sub>2</sub> :0.01 L/min	0.341
DBD (Wang <i>et al.</i> , 2017)	Methylene Blue	V0 : 0.05L, C0: 100 ppm, t:40 min, V: 6 kV, F: 50 kHz, O <sub>2</sub> :0.06L/min	0.14
Non-equilibrium plasma (Zhang <i>et al.</i> , 2008)	Methyl Orange	V0 : 0.1L, C0: 100 ppm, t:15 min, V: 46 kV, F: 100 kHz, Air:1.6 L/min	3.6

plasma-based treatments, such as double-chamber DBD (0.14 g/kWh), pulsed corona discharge with ozonation (0.341 g/kWh), and non-equilibrium plasma (3.6 g/kWh). This demonstrates its superior efficiency in dye wastewater treatment. Additionally, the study confirmed that treatment duration directly affects MB degradation, with higher initial concentrations requiring longer exposure times. A significant pH reduction during plasma treatment suggests oxidation and structural changes in MB molecules, aligning with previous research on plasma-activated water (PAW) chemistry. Overall, the QCDC plasma system proves to be a highly efficient and energy-effective method for wastewater treatment. However, whether the device can perform equivalently under pre-test conditions remain unclear for wastewater purification. Thus, further testing on wastewater is necessary before practical use.

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