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Effect of Dye and NaCl Concentrations on Methylene Blue Dye Removal by Electrocoagulation

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Abstract

This study investigated the influence of two significant operating factors of dye and NaCl concentrations on dye removal efficiency by electrocoagulation (EC) process. The methylene blue (MB) was represented dye molecule containing in the wastewater, whereas the NaCl was playing a role as electrical conductivity for promoting chemical reactions. The initial concentrations of MB and NaCl were varied from 3 to 60 mg/L and 0.10 to 1.00 g/L respectively to find the best condition for dye removal. The results revealed that the maximal dye removal of 91% was observed at MB concentration of 45 mg/L and NaCl of 0.50 g/L. The higher dye concentration (i.e., 60 mg/L) and lower dye concentration (i.e., 3, 15 and 30 mg/L) were affected on decreasing the dye removal. Similarly, the higher NaCl concentration (i.e, 0.75 and 1.00 g/L) and lower NaCl concentration (i.e. 0.10 and 0.25 g/L) had negative impacts on dye removal. Furthermore, the actual batik wastewater was preliminary treated by EC process; the COD was decreased from 3720 mg/L in the initial concentration to 320 mg/L after 60 min. In addition, the dye removal by EC process can be enhanced by controlling other parameters such as submerged surface area, applied current density and pH which will be further studied prior to imply as treatment process for textile industry.

Keywords: Electrocoagulation process, dye wastewater, NaCl concentration.

1. INTRODUCTION

At the present, the removal of dye molecule from textile wastewater has been considered as one of the major issues which received the increasing attention from global viewpoints (de Carvalho et al., 2005). The wastewater containing high dye concentration is widely produced from many industries such as textile, leather, paper, printing and photographs. Among those industries, the textile industry consumed a considerable quantity of dyes and generated the wastewater containing high color, suspended particles, high chemical oxygen demand (COD) and pH (Mahmoud et al., 2013; Mollah et al., 2004). In addition, approximately 15% of dye was lost during dyeing process and discharged as contaminated water into aqueous environment (Daneshvar et al., 2006). The existence of dye in the aqueous environment can cause aesthetical trouble (even at low concentration of 1 mg/L) (Mohammed et al., 2014), reduction of photosynthesis and gas solubility. Furthermore, a natural biochemical process can change dye molecule to other dangerous by-products which were toxic, carcinogenic or even destroy aquatic species (Mohammed et al., 2014; Daneshvar et al., 2003).

Several technologies have been used for dye wastewater treatment including biological process (e.g., activated sludge, sequencing batch reactor; SBR, bio-filter), chemical oxidation (e.g., fenton/like-fenton, hypochlorite), electrochemical oxidation (e.g., O_3/UV, O_3/H_2O_2, O_3/UV/H_2O_2, H_2O_2/UV), photodegradation (e.g., TiO_2/UV, photo-fenton), adsorption (e.g., activated carbon), membrane filtration (e.g., microfiltration, ultrafiltration, nanofiltration) and chemical coagulation/flocculation (e.g., aluminium, iron or calcium salts) (Zidane et al., 2008). Although the biological process was considered as a low cost technique, most of dye molecules are toxic and resistant to bacterial growth at high concentration (Daneshvar et al., 2006; Raghu & Basha, 2007). Meanwhile, the chemical and photo oxidation process was required a lot of chemical addition, which may cause further environmental problems (Yari et al., 2013). The other advanced technologies were limited on high operating cost (Mountassir et al., 2015), which was the most concerning parameter for actual wastewater treatment application.

The electrocoagulation (EC) method has been developed as a promising technology in removing very polluted wastewater in recent years. The spent coolant with initial COD of 122 g/L was pre-treated by modified EC reactor using Al electrode; the 65% of COD removal was achieved (Pantorlawn et al., 2018). The tannery wastewater was also treated by a combined system of EC and biological fungal; the efficient COD and Cr^{6+} removals were 96% and 97% respectively (Deveci et al., 2019). The principle of this technology is based on the electrochemical dissolution of metal electrode into wastewater that improves the efficiency of coagulation, adsorption and precipitation of pollutants by using a direct current source (Bani-Melhem & Smith, 2012). Iron and aluminium are two typical types of electrodes which have been widely used in EC systems. The aluminium electrode has been demonstrated to be cheap and effective material for the EC process (Thirugnanasambandham et al., 2015). The main reactions at aluminium electrode occurs as follows: Dalvand et al. (2011).

The aluminium oxidation at the anode:

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

The water reduction at the cathode:

$$3H_2O + 3e^- \rightarrow \frac{3}{2}H_2 + 3OH^- \quad (2)$$

Overall:

$$Al + 3H_2O \rightarrow Al(OH)_{3(s)} + \frac{3}{2}H_2 \quad (3)$$

These reactions produce monomeric and polymeric species of aluminium hydroxide (i.e., $Al(OH)_3$) that remove pollutants by complexation and electrostatic attraction. In complexation, pollutants such as dye act as ligand to bind hydrous aluminium by the following reaction:

$$\text{Dye-H} + (OH)Al_{(s)} \rightarrow \text{Dye-OAl}_{(s)} + H_2O \quad (4)$$

Meanwhile, the positive and negative charge iron/alminium oxide particles attract the opposite charge species in electrostatic attraction (Golder et al., 2005). Besides the efficiency, this technique was also known well by its easy operation, simple equipment and small amount of sludge generation (Bayramoglu et al., 2007). However, an electrical conductivity in the wastewater was a significantly factor for effective dye removal. Similarly, dye molecule structure and its concentration were concerning factors. The aim of this study was that a methylene blue (MB) which is positive charge blue dye was removed by EC process using aluminium electrode. The experiments were set to investigate the effect of initial dye and NaCl (related to electrical conductivity) concentrations. Later, the actual dye wastewater from a batik company was preliminary tested.

2. MATERIAL AND METHODS

2.1 Electrocoagulation (EC) reactor

The experimental set up used in this study consists of a 200 ml acrylic reactor. The four electrodes of aluminium were connected in a bipolar mode with dimensions of 10 cm (length) x 4.5 cm (width) x 0.01 cm (thick), and were placed at a distance of 1.5 cm from the bottom of reactor. The submerged surface area was approximately 160 cm^2. The electrodes were connected to direct current power supply (DeltaPSU, PMT-5V350W1AM) with 5 V as maximal tension and 60 A as maximal intensity. The schematic of EC reactor is presented in Figure 1

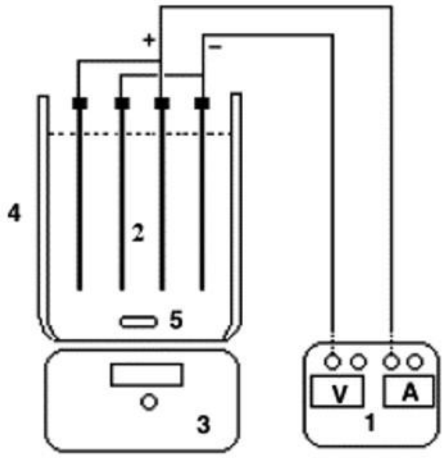

Figure 1 Schematic of electrocoagulation reactor; (1) power supply, (2) aluminium electrodes (1 cm of interelectrode distance), (3) magnetic stirrer, (4) acrylic reactor and (5) magnetic bar

2.2 Preparation of dye wastewater

Synthetic wastewater was prepared by mixing an appropriate amount of methylene blue (MB) into deionized water. The MB was purchased from Ajax Finechem Pty Ltd as purified agent. The chemical structure of the hydrolyzed from of this dye is present in Figure 2.

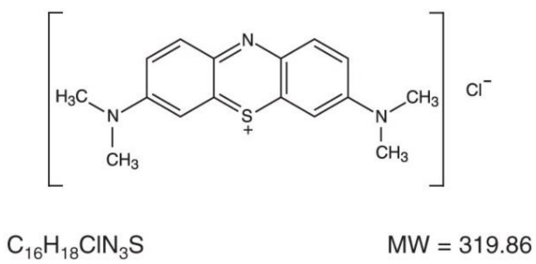

Figure 2 Chemical structure of methylene blue

2.3 Experiments

The experimental EC reactor was conducted with 200 mL of MB solution and stirred at 200 rpm. All experiments were carried out at room temperature (~25 °C). The sample was collected every 10 minutes and then centrifuged at 5,000 rpm for 10 min. Subsequently, the collected sample was measured the absorbance spectra (Abs) at a wavelength of 664 nm by UV-6100 Double Beam spectrophotometer (Shanghai Mapada Instruments Co., Ltd). The percentage of MB removal was determined according to the following equation:

$$\text{Dye removal (\%)} = \frac{\text{Abs}_0 - \text{Abs}_t}{\text{Abs}_0} \times 100 \qquad (5)$$

Where Abs_0 = Initial absorbance intensity.
Abs_t = Absorbance intensity at time t.

When the batik wastewater was tested by EC process, the COD value was measured in accordance with the standard method for the examination of water and wastewater (American Public Health Association, 1995). The COD removal efficiency was calculated in the below equation:

$$\text{COD removal (\%)} = \frac{\text{COD}_0 - \text{COD}_t}{\text{COD}_0} \times 100 \qquad (6)$$

Where COD_0 = Initial COD concentration.
COD_t = COD concentration at time t.

In the experiment 1, the MB concentration was varied from 3 to 60 mg/L, while the NaCl were controlled at 0.5 g/L. In the experiment 2, the NaCl concentration was varied from 0.10 to 1.00 g/L, whereas the MB concentration was kept at the best condition from the experiment 1. In the last experiment, the actual batik wastewater was tested for 60 min, as summarized in Table 1. The MB and batik wastewater were treated by EC process without pH adjustment; the initial pH was in the range of 8-9. The applied current density was constant at 20 mA/cm^2 for all experiments.

Table 1 Various experimental parameters in this study

Experiment	Wastewater source and concentration	NaCl concentration	Time
1	MB 3, 15, 30, 45 and 60 mg/L	0.50 g/L	10, 20, 30, 40, 50 and 60 min
2	MB 45 mg/L	0.10, 0.25, 0.50, 0.75 and 1.00 g/L	
3	Batik wastewater	0.50 g/L	60 min

3. Results and Discussion

3.1 Effect of initial MB concentration

The experiment was carried out for five different MB concentrations of 3, 15, 30, 45 and 60 mg/L for 60 min. The NaCl and current density were constant of 0.5 g/L (conductivity was ~ 1 mS/cm) and 20 mA/cm^2. Figure 3 shows that the dye removal by EC process was increased gradually by electrolysis times. At the lowest MB concentration of 3 mg/L, the dye removal achieved 31% at 10 min and approximately 75% at 60 min.

At increasing MB concentrations, the maximal dye removal was increased to 84%, 89% and 91% at 60 min for MB concentration of 15, 30 and 45 mg/L respectively. However, the maximal dye removal was dropped to 83% at the highest MB concentration of 60 mg/L. This is because the amounts of coagulant ($Al(OH)_3$) were constant at the fixed current density. The produced coagulant adsorbed the MB molecule until the maximal adsorption capacity was reached. Later, the produced coagulant was insufficient to adsorb MB molecule at the very high concentration, resulting in the decrease in dye removal finally (Mahmoud et al., 2013). According to this experiment, the effective dye removal by EC process could not achieve at the low MB concentration, therefore the EC process was an appropriate treatment process for high pollutants concentration. The best dye removal efficiency was observed at the MB concentration of 45 mg/L, therefore this concentration value was used as a controlled factor in the next experiment.

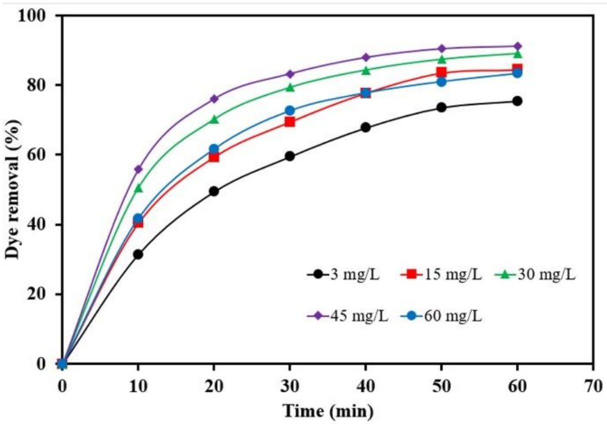

Figure 3 Effect of initial MB concentration on the removal efficiency (NaCl 0.5 g/L)

3.2 Effect of NaCl concentration

The experiment was carried out for five different NaCl concentrations of 0.10, 0.25, 0.50, 0.75 and 1.00 g/L, which referred to different electrical conductivity. The initial MB and current density were controlled at 45 mg/L and 20 mA/cm^2. As shown in Figure 4, the dye removal was increased by electrolysis times and NaCl concentrations. The maximal dye removal was observed at 60 min; the dye removal was 83%, 89% and 91% for NaCl 0.10, 0.25 and 0.50 g/L respectively. However, the excessive NaCl of 0.75 and 1.00 g/L caused a reduction of dye removal to 87% and 81%. The explanation was that the excessive NaCl and electrical conductivity caused the overconsumption of aluminium electrodes, and resulted in the irregular dissolution (Calvo et al., 2003). Furthermore, the high NaCl and electrical conductivity caused the increase in electron transportation rate in the MB solution, resulting in the decreasing reaction time and subsequently the decreasing dye removal. According to this experiment, the optimal NaCl was 0.5 g/L for effective MB dye removal.

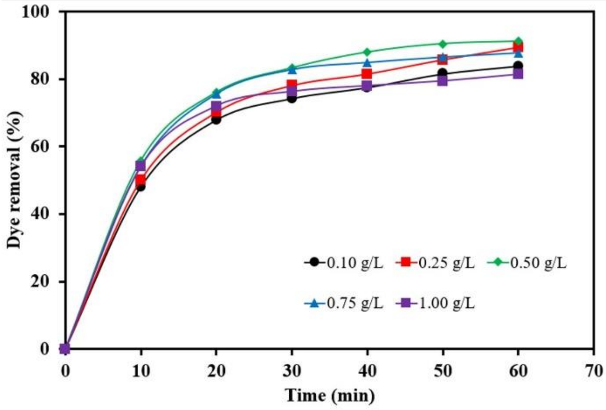

Figure 4 Effect of NaCl concentration on the removal efficiency (MB 45 mg/L)

The actual batik wastewater was preliminary treated by EC process. The appearance wastewater was dark blue and chemical smell. The wastewater with initial COD of 3,720 mg/L was tested under NaCl of 0.5 g/L, and the results are shown in Figure 5. The COD removal was 67% at 10 min and reached 91% at 60 min. The final COD was decreased to only 320 mg/L. The comparison of batik wastewater before and after treatment is shown in Figure 6. It can be seen that the excellent removal efficiency was also found in the actual wastewater, even though the pollutants concentration was higher rather than the synthetic MB solution. This is because the particular characteristic of batik wastewater such as conductivity, pH, ions and biodegradable compounds effected on enhancing the EC process. The further study on effect of other operating factors on enhancing the EC process is necessary and clarified before applying this technology to dye and textile industry.

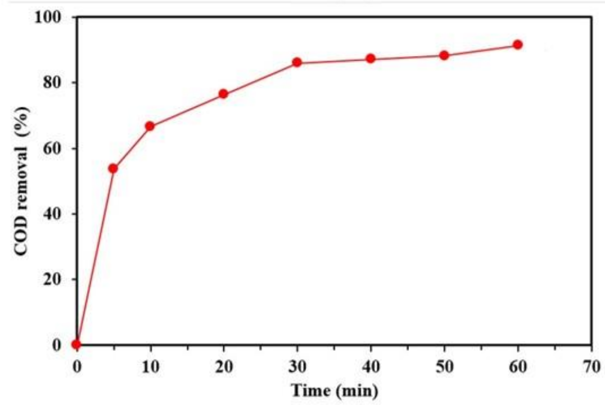

Figure 5 Actual batik wastewater treatment by EC process; (a) effluent COD and (b) COD removal efficiency (NaCl 0.5 g/L)

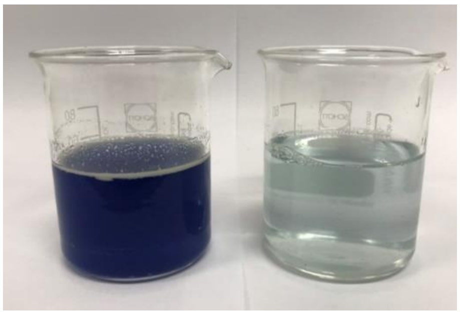

Figure 6 Actual batik wastewater treatment before and after treatment by EC process (NaCl 0.50 g/L, time 60 min)

4. CONCLUSION

The EC process was a potential treatment process for dye removal. The maximal dye removal of 91% was observed at the optimal concentration of MB and NaCl, which were 45 mg/L and 0.5 g/L respectively. The excessive and insufficient MB and NaCl could cause the decreasing dye removal. When the actual batik wastewater containing initial COD of 3,720 mg/L was tested, the COD removal reached 91% at 60 min and the final COD was only 320 mg/L.

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