

## ORIGINAL ARTICLE

## Color and Total Organic Carbon (TOC) Removal from Peat Water Using The Electrocoagulation Process: Central Composite Design for Optimization

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**ABSTRACT** – The electrocoagulation process is simple and environmentally friendly. It removes pollutants in peat water such as color and total organic carbon (TOC). In this study, the electrocoagulation process was designed, optimized, and investigated using central composite design (CCD) type response surface methodology (RSM). The effects of current density and reaction time in the range of 6–14 mA/cm<sup>2</sup> and 30–90 minutes on the efficiency of color and TOC treatment were evaluated. The best results for removing output efficiency were 10 mA/cm<sup>2</sup> for 30 minutes (98.1% color) and 10 mA/cm<sup>2</sup> for 30 minutes (91% TOC). By comparing actual and predicted data, the optimum condition in this process occurs when the current density is 6.140 mA/cm<sup>2</sup> and the reaction time is 76.042 minutes. The experimental data can be well described using the central composite design.

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### INTRODUCTION

Peat water is surface water that contains a high proportion of natural organic matter (NOM) in the form of humic acid, the direct consumption of which, particularly over a long period of time, is harmful [1], [2]. The water in peat becomes brown and acidic due to the high concentration of NOM, especially hydrophobic NOM, such as fulvic acid and humic acid. In addition to high NOM concentrations, peat water contains other pollutants, including heavy metals such as iron (Fe) and manganese (Mn), total coliform bacteria [3], total suspended solids (TSS), and salt ions [4], contribute to high conductivity [5]. Due to these difficulties, local communities, particularly those in rural areas, now depend primarily on precipitation, which is getting harder to come by due to climate change [6].

Some of the research work that has been conducted and contributed to the treatment of peat water as a source of clean water suitable for consumption includes conventional coagulation [7], filtration [3], oxidation, and adsorption [8]. In the treatment of water, electrocoagulation is an alternative to chemical coagulation. It combines the principles of coagulation, foaming, and electrochemistry in one system [9]. The main advantages of electrocoagulation are its fast reaction time, minimal sludge formation, and non-toxicity, which reduces environmental pollution as no chemical additives are required. Furthermore, the process is facilitated by simple devices that allow complete automation [10]–[12]. The cost of the electrocoagulation method is 82.93% lower than that of the chemical coagulation method [13]. Electrocoagulation is widely used in various types of water treatment, including peat water [1], [14], [15], lake water [16], [17] and wastewater [18], [19].

A statistical technique for developing mathematical models for process development, optimization, and improvement is called response surface methodology (RSM). Its application includes the conception, development, and planning of new studies as well as the improvement of existing studies. RSM offers several advantages over time-consuming conservative approaches, including increased speed and systematic accuracy [20]. Another advantage of RSM is that it can minimize the number of experiments performed and develop mathematical models to predict the response [21]. Although fewer tests are performed, RSM can also provide more information. RSM has two types of test plans: Box-Behnken design (BBD) and central composite design (CCD). Models built with BBD and CCD are subjected to analysis of variance (ANOVA) to test the effects of the various linear ( $X_i$ ), quadratic ( $X_i^2$ ), or by interaction ( $X_iX_j$ ) components [22].

This study aims to evaluate and find the optimal conditions for the electrocoagulation process parameters in order to mitigate and solve problems related to peat water quality. The effect of various operating parameters (current density and reaction time) on color and total organic carbon (TOC) removal was evaluated in a central composite design (CCD).

## EXPERIMENTAL METHOD

### Materials and Instruments

The peat water was collected from Pematang Panjang Village, South Kalimantan, Indonesia. The peat water was allowed to settle to minimize sediment and particles introduced during the sampling process. According to the parameter to be examined based on accepted techniques for examining water and wastewater, Table 1 displays the initial characteristics of peat water (APHA 2012).

**Table 1.** Characteristics of peat water

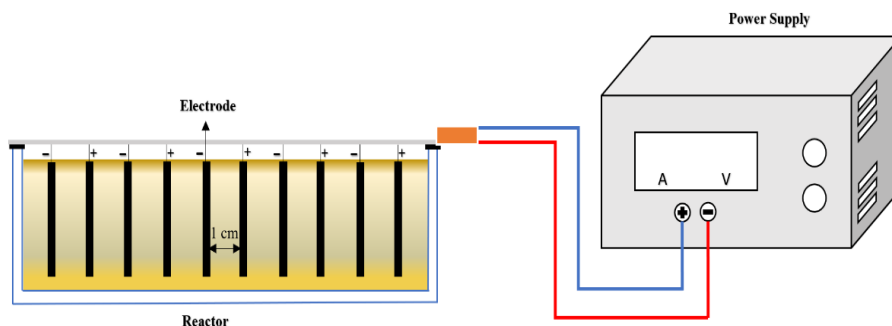
| Parameter                    | Value |
|------------------------------|-------|
| Turbidity (NTU)              | 24    |
| Color (PtCo)                 | 66    |
| pH                           | 6,8   |
| Total dissolved solid (mg/L) | 30    |
| Total organic carbon (mg/L)  | 23.86 |

The removal efficiencies of color and total organic carbon (TOC) were calculated from Equation (1):

$$\eta = \left( \frac{C_i - C_f}{C_i} \right) \times 100 \quad (1)$$

where  $C_i$  and  $C_f$  refer to the initial concentration and effluent concentration.

The electrocoagulation reactor in the electrocoagulation unit is composed of Perspex and measures 31.5 cm × 18.5 cm × 24.4 cm. The power was supplied via a DC power supply (RIDEN® RD6012W) and the aluminum electrode. There are ten monopolar electrodes with the same dimensions (12.5 cm × 9 cm × 0.1 cm) as anode and cathode, spaced 1 cm apart. The electrode, the central component of the electrocoagulation process, is made of aluminum because it is the most widely used material, affordable, easily obtainable, and an efficient way to treat wastewater and water. The electrocoagulation unit's schematic diagram is shown in Figure 1.



**Figure 1.** An electrocoagulation reactor schematic diagram

### Method and Procedure

This investigation set out to assess the effects of reaction time and current density on batch electrocoagulation experiment performance. Statistical response surface methodology (RSM) based on central composite design (CCD) and experimental data was used in the optimization process and analysis of the results using Design Expert version 13 software. The influences of current density (6–14 mA/cm<sup>2</sup>) and the reaction time (30–90 minutes) were selected for this experimental setup. In this study, the removal efficiency of color (%) and TOC was the main focus of the system response. The color in peat water was measured using an ultraviolet-visible (UV-vis) spectrophotometer. The color intensity in peat water samples can be measured by spectrophotometer according to SNI 6989.80:2011 with a wavelength of 400 nm. The color intensity value obtained is expressed in units of Pt-Co. This test method is used for true color measurement. The TOC test method refers to SNI 06-6989.28-2005. The principle of this method is that a test example has homogeneously aspirated into a combustion tube wrapped in an oxidative catalyst and heated at 680°C. The water evaporates, and the organic matter oxidizes into CO<sub>2</sub> and H<sub>2</sub>O. The CO<sub>2</sub> produced is flowed with the carrier gas, and the detector

response is measured with a nondispersive infrared analyzer (NDIR). From the measurement results, the total carbon and inorganic carbon values are obtained separately, while the TOC value is obtained from the difference. The variables and their values are also listed in Table 2 and Table 3, which can be calculated from Equation (2). These decisions were made after a preliminary review of the literature, a comprehensive analysis of results, and theoretical insights gained from simple experiments with 5 and 12 center and non-center points.

$$N = 2^n + 2n + n_c = 2^2 + (2 \times 4) + 5 = 17 \quad (2)$$

Where N is the total number of experimental variables, n and  $n_c$  refer to the number of independent variables and centered point variables.

**Table 2.** Experimental design ranges of CCD's levels.

| Factor      | Variables       | Unit               | Low | High | -alpha | +alpha |
|-------------|-----------------|--------------------|-----|------|--------|--------|
| A (Numeric) | Current density | mA/cm <sup>2</sup> | 6   | 14   | 6      | 14     |
| B (Numeric) | Reaction time   | Minutes            | 30  | 90   | 30     | 90     |

**Table 3.** Experimental design from a design expert program using CCD

| Run | Factor A                              | Factor B             |
|-----|---------------------------------------|----------------------|
|     | Current density (mA/cm <sup>2</sup> ) | Reaction time (min.) |
| 1   | 10                                    | 60                   |
| 2   | 6                                     | 60                   |
| 3   | 14                                    | 30                   |
| 4   | 14                                    | 60                   |
| 5   | 10                                    | 60                   |
| 6   | 10                                    | 60                   |
| 7   | 10                                    | 90                   |
| 8   | 6                                     | 60                   |
| 9   | 6                                     | 90                   |
| 10  | 10                                    | 60                   |
| 11  | 14                                    | 60                   |
| 12  | 6                                     | 30                   |
| 13  | 10                                    | 30                   |
| 14  | 10                                    | 60                   |
| 15  | 10                                    | 30                   |
| 16  | 10                                    | 90                   |
| 17  | 14                                    | 90                   |

Regression analysis was performed using the coefficient of determination ( $R^2$ ), the adjusted  $R^2$  and predicted  $R^2$  as performance indicators. The model terms are also tested with p and F values.

## RESULT AND DISCUSSION

The second-order (quadratic) polynomial response surface model was applied to fit the experimental results obtained by central composite design (CCD). Based on the experimental design results, the regression equations with the coded variables obtained for describing the color and total organic carbon (TOC) removal from peat water by electrocoagulation using aluminum (Al) electrodes are presented in Equation (3).

$$\text{Color removal (\%)} = 89.43 - 0.1866X_1 + 3.9X_2 + 0.1797X_1X_2 - 0.6306X_1^2 - 2.26X_2^2 \quad (3)$$

$$\text{TOC removal (\%)} = 95.39 + 1.75X_1 - 6.22X_2 + 0.0702X_1X_2 - 4.72X_1^2 - 2.80X_2^2 \quad (2)$$

The equation in terms of coded factors can be used to make predictions about the response for each factor. CCD shows the effects of varying current density ( $X_1$ ) and reaction time ( $X_2$ ). CCD evaluated 17 experimental runs with five center points. The best color removal performance (98.1%) was achieved in run 15 with an applied current density of 10 mA/cm<sup>2</sup> within 30 minutes of the reaction time. The best TOC removal performance (91%) was achieved in run 7 with an applied current density of 10 mA/cm<sup>2</sup> within 90 minutes of reaction time. Table 4 shows the response results of color removal efficiency and TOC removal efficiency. Table 5 shows that the predicted  $R^2$  and adjusted  $R^2$  for color and TOC removal are in reasonable agreement, with the difference being less than 0.2.

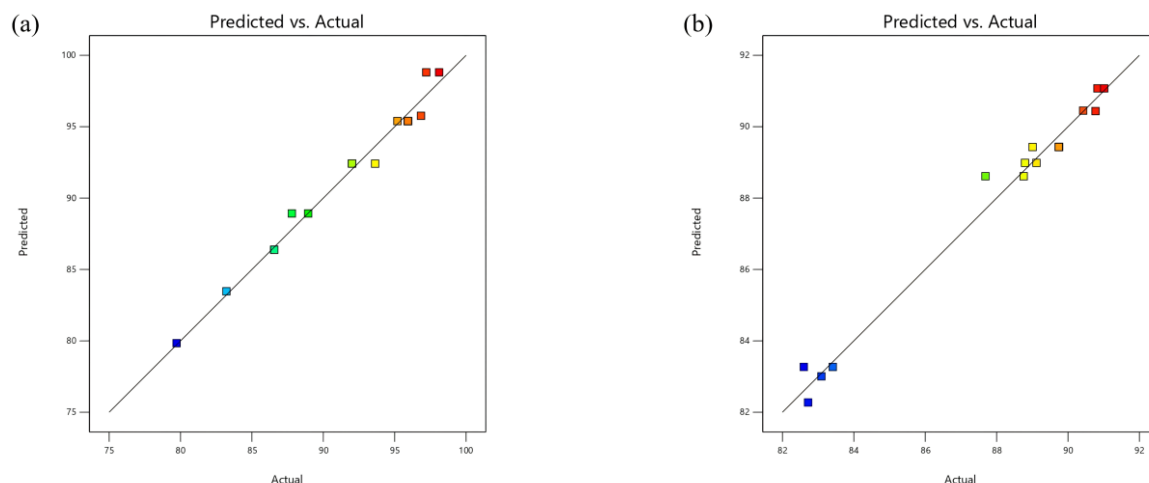
**Table 4.** Response results of color and TOC removal efficiency

| Run | Factor A                                 | Factor B                | Response                        |                               |
|-----|------------------------------------------|-------------------------|---------------------------------|-------------------------------|
|     | Current density<br>(mA/cm <sup>2</sup> ) | Reaction time<br>(min.) | Color Removal Efficiency<br>(%) | TOC Removal Efficiency<br>(%) |
| 1   | 10                                       | 60                      | 95.925                          | 89.741                        |
| 2   | 6                                        | 60                      | 87.8                            | 88.796                        |
| 3   | 14                                       | 30                      | 96.841                          | 82.719                        |
| 4   | 14                                       | 60                      | 92.011                          | 88.759                        |
| 5   | 10                                       | 60                      | 95.19                           | 89.009                        |
| 6   | 10                                       | 60                      | 95.924                          | 89.743                        |
| 7   | 10                                       | 90                      | 86.565                          | 91.016                        |
| 8   | 6                                        | 60                      | 88.94                           | 89.117                        |
| 9   | 6                                        | 90                      | 79.724                          | 90.425                        |
| 10  | 10                                       | 60                      | 95.926                          | 89.748                        |
| 11  | 14                                       | 60                      | 92.012                          | 87.688                        |
| 12  | 6                                        | 30                      | 93.632                          | 83.092                        |
| 13  | 10                                       | 30                      | 97.211                          | 82.599                        |
| 14  | 10                                       | 60                      | 95.927                          | 89.75                         |
| 15  | 10                                       | 30                      | 98.112                          | 83.411                        |
| 16  | 10                                       | 90                      | 86.564                          | 90.825                        |
| 17  | 14                                       | 90                      | 83.214                          | 90.771                        |

**Table 5.** Model statistics

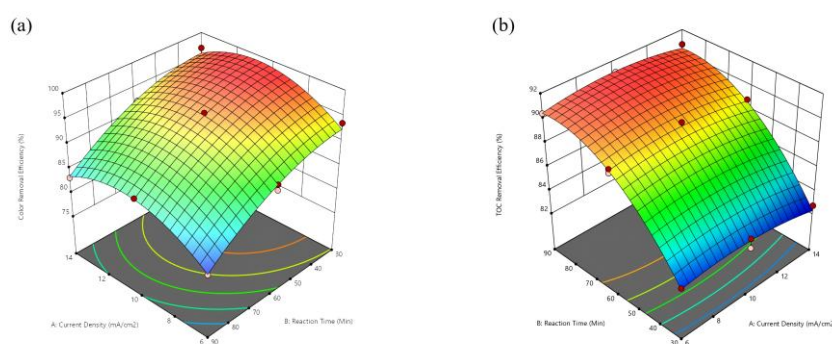
|                 | Color removed | TOC removed |
|-----------------|---------------|-------------|
| $R^2$           | 0.9819        | 0.9832      |
| Adjusted $R^2$  | 0.9758        | 0.9777      |
| Predicted $R^2$ | 0.9578        | 0.9644      |

The mere presence of a high  $R^2$  value does not guarantee the quality of the model, as it is unable to assess the appropriateness of the regression model and whether the coefficient estimates are misleading [14].  $R^2$ , adjusted  $R^2$ , and predicted  $R^2$  is the main correlation coefficient of the statistical model [23]. Figure 2 shows the integration of the actual and predicted removal efficiencies and negligible variation between the removal efficiencies of all runs.



**Figure 2.** Predicted vs. actual for (a) color removal and (b) TOC removal

CCD can be used to illustrate the effects of process variables and responses. Figure 2 and Figure 3 illustrate the effect of operational factors (current density and reaction time) on the color and TOC removal efficiency of peat water. From Figure 3(a), the best color removal performance was achieved at a current density of 10 mA/cm<sup>2</sup> within 30 minutes of the reaction time. The color removal tends to minimize the reaction time at a current density of 6 mA/cm<sup>2</sup> within 90 minutes. In Figure 3(b), the TOC removal efficiency was increased as the operating conditions approached the highest time range at a current density of 10 mA/cm<sup>2</sup>. Within 90 minutes of reaction time, color removal tends to be minimized at a current density of 10 mA/cm<sup>2</sup> within 30 minutes of reaction time.



**Figure 3.** 3D Surface plots for (a) color removal and (b) TOC removal

Reaction time is an important factor in determining electrode suitability and sludge formation in the electrocoagulation process. At the same time, several electrochemical reactions take place on the electrode, making pollutants unstable [23]. In Figure 3(a), the current density increases dramatically from 6 mA/cm<sup>2</sup> after 30 minutes. This is due to the release of excess aluminum hydroxide (Al(OH)<sub>3</sub>) compounds during the electrocoagulation process. The release of excess Al(OH)<sub>3</sub> compounds is due to the fact that the pollutants contained in the peat water have been completely bound, so it can be assumed that the water only contains unbound coagulant. Higher current densities and longer reaction times produce more hydrogen gas in solution, resulting in greater flotation and bringing suspended particles to the surface [14], [24]. In Figure 3(b), the TOC removal efficiency increases proportionally with the increase in reaction time, while too high current density leads to faster electrode passivation [2].

Electrocoagulation can cause the removal of color and TOC in peat water due to the release of  $\text{Al}(\text{OH})_3$  compounds in the form of coagulants, which then bind to pollutants and settle as sludge; at the same time, the  $\text{H}_2$  compounds that are released then also bind to pollutants to produce flocs that form to the surface of the water as foam, the greater the current density and the longer the contact time, the better the clarity of peat water. However, in this study, the best conditions were obtained in the efficiency of color removal at a current meeting of  $10 \text{ mA/cm}^2$  with a reaction time of 30 minutes and the efficiency of TOC removal at a current meeting of  $10 \text{ mA/cm}^2$  with a reaction time of 90 minutes. This can be caused by the release of excess  $\text{Al}(\text{OH})_3$  compounds because the pollutants contained in peat water have all been bonded, so it can be mentioned that the water only contains non-bonded coagulants. High current density and longer reaction times will produce more hydrogen gas in the solution, resulting in increased flotation and stirring of the solution [25]. In addition, there is an increase in  $\text{Al}^{3+}$  ions (more floc) in the solution, thus increasing the release of coagulants. More  $\text{Al}^{3+}$  ions will stimulate the formation of coagulants after aggregation [26]. During this electrocoagulation process, there was a change in the pH of the sample, which was initially 6.79 to 9.62 after 90 minutes.

Optimal conditions for peat water treatment concerning pollutant removal consider the energy consumption during the process. The desirability function has the role of an objective function, which indicates a value that ranges from zero to one at the targeted goal. The optimization program aims to maximize this desirability function. By these experiments, it is found that the optimum condition in this process occurs when the current density is  $6.140 \text{ mA/cm}^2$  and the reaction time is 76.042 minutes. Under this condition, the efficiency removal for color and TOC attained 85.149% and 90.371%, respectively, with a desirability score of 1. These optimal values were then experimentally validated, resulting in a color removal of 85.185% and a TOC removal of 90.464%.

The specific objectives of the controlled approach of each factor and response variable are useful to maximize, minimize, and target within a set range. The objective of this color and TOC removal method using the electrocoagulation process is to identify suitable and cost-effective solutions. Compared to previous studies, the results of this study show competitive efficiency in color removal and TOC in peat water using electrocoagulation process. Rahman et al. [1] reported a TOC removal of 90% at higher current densities ( $\geq 10 \text{ mA/cm}^2$ ), and Sari et al. [13] also reported an optimal density current ranging from  $8\text{--}12 \text{ mA/cm}^2$  with a TOC removal efficiency of 85–92%. The results of this study show that electrocoagulation can effectively remove color and TOC under more economical conditions than previous studies.

## CONCLUSION

The optimization process aimed at maximizing color and total organic carbon (TOC) removal efficiencies involved the evaluation of several starting points, with optimal conditions identified at a current density of  $6.140 \text{ mA/cm}^2$  and reaction time of 76.042 minutes. In this study, reaction time and voltage contributed to removing color and TOC in peat water. Increased treatment efficiency up to a threshold at which no additional increase in TOC or color removal was indicated. For appropriate design conditions with comparable characteristics ( $R^2 > 0.9$ ), the found mathematical equations can accurately predict the treatment efficiency values. This study highlights the need for targeted strategies to address specific contaminants (color and TOC) and emphasizes the critical role of understanding and controlling operational variables to improve treatment efficiency and ensure environmental safety.

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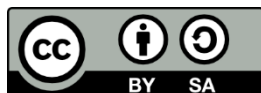
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