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PHOTOVOLTAIC CELL ELECTRO-FENTON OXIDATION PROCESS FOR TREATMENT OF ORGANIC CONTENT IN METHYL ORANGE WASTEWATER

AHMED SAMIR NAJE*[1], ISRA'A SADI SAMAKA[2], HAIDER M. ZWAIN[3] AND MOHAMMED A. AJEEL[4]

[1,3]College of Water Resources Engineering, AL-Qasim Green University, Babylon, Iraq. [2]Department of Environmental Engineering, College of Engineering, University of Babylon, Babylon, Iraq [4]Department of Environment, Faculty of Environment and Energy, Al-Karkh University of Science, Iraq.

**Corresponding author: ahmednamesamir@yahoo.com*
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Abstract: This paper explores the performance of Photovoltaic cell Electro-Fenton oxidation during the treatment of methyl orange wastewater. The treatment setup comprises a photovoltaic cell, a battery, DC to AC inverter and an electrooxidation reactor. Throughout the Photovoltaic cell Electro-Fenton oxidation experiment, the effect of operating parameters like current, H_2O_2, rotation speed and electrolysis time on the total organic carbon and energy consumption were examined. The optimization study was performed by varying the current (0.5-2 mA), hydrogen peroxide (25-100 mg/L), rpm (100-200), and the electrolysis time (5-15 min) using response surface methodology (RSM) and Minitab-17. The pH and the amount of NaCl in the solution were maintained as 3, 0.2g respectively. The optimization study shows that 98% removal efficiency is achievable, consuming 39.4 kWh/m^3 energy.

Keywords: Dye wastewater, organic content, wastewater treatment, electro-Fenton.

Introduction

Among the numerous forms of worldwide pollution found nowadays, water contamination is the most prevalent. According to the World Bank, the industrial water effluents account for about 20% of world water contamination (Haider T. Naeem & Ali A. Hassan, 2018). Dye wastewater, if not professionally treated, could constitute an ecological issue because of its poisonous nature and potential carcinogenic consequence, which could cause permanent injury to human health and the environment (X. Li *et al.*, 2017; Ali Saleh Jafer *et al.*, 2019). Due to the ecological rules, which demand the reuse and treatment of dye wastewater, many researchers have focused on the removal of organic content in wastewater (Ali A. Hassan *et al.*, 2018). Conventional treatments of dye wastewater have proven to be nonviable because of the emergence of their toxic, non-biodegradable and intractable effluence. The development of suitable alternative techniques is urgently needed because potable water is a precious commodity in numerous regions of the world (Alvarez-Corena *et al.*, 2016; Ali A. Hassan & Khalid M. Mousa Al-zobai, 2019; Fathy *et al.*, 2017 ; Galvão *et al.*, 2006; Hassan & Haider T. Naeem, 2019). Various conservative methods have been used to eliminate organic matter from wastewater: biological treatment (Q. Li *et al.*, 2005), adsorption (El-Din *et al.*, 2017; Jafer & Hassan, 2019), and membrane separation (Zoubeik *et al.*, 2017; Zsirai *et al.*, 2018). Several efforts have been made to explore the strong oxidation performance of the Fenton process. The photovoltaic cell Electro-Fenton process is an emerging Fenton process and has attracted considerable attention for removing organic substances from dye wastewater. Fenton oxidation process consists of the reaction between H_2O_2 and Fe^{2+} in acidic solution to produce hydroxyl radicals, a highly oxidant OH species responsible for pollutant degradation (Gutierrez-Mata *et al.*, 2017). As it is technically simple, low-cost and non-toxic because H_2O_2 and iron are its main reactants; the Fenton's process is one of the most viable advanced oxidation processes (AOP). The traditional Fenton process uses homogeneous catalysts, and is confronted with problems like slower ferrous ion renewal rate and extreme iron clay manufacture. Nonetheless, these disadvantages have to be overcome if this method is to be adopted on a commercial scale. This was trapped by the heterogeneous electrochemical advanced oxidation processes (EAOPs) after the electric fenton procedure (Baiju et al., 2018). EAOP has unique electro-oxidation advantages aimed at the comprehensive oxidation of tenacious organic content in wastewater (Divyapriya *et al*., 2018). The electro-Fenton eco-friendly methods has some exciting features, for instance high adaptability, excellent efficiency, high acquiescence and ecological compatibility, which distinguishes them from other AOPs (Ganiyu *et al*., 2018; X. Li *et al*., 2017). In a Photovoltaic cell Electro-Fenton process, pollutants are detached by Fenton's reagents composed of anodic oxidation on the anode surface. Ordinarily, anodic oxidation is not suitable for mineralization of most aromatic contaminants due to the formation of strong carboxylic acids (He & Zhou, 2017). These biodegradable techniques possess some exciting features, for example, high flexibility, excellent competence, long acquiescence, and ecological compatibility (Forat Y. AlJaberi et al., 2020). This process variance includes the in-situ generation of highly reactive oxidizing reagents, which are free radicals obtained from the reaction between H_2O_2 and Fe^{2+}(Buftia *et al*., 2018), (Atiyah *et al*., 2020). The crucial limits of the process are narrow pH range (2.0-3.0) and iron residue in the treated wastewater. The significant drawbacks of chemical treatment techniques are low removal efficiency and long separation time. The chemical methods also require a large amount of oxidant, low pH, and conductivity adjustment, making the process not cost-effective. Electrochemical methods have received immense attention recently because of the smooth process and small amount of chemicals required (Davarnejad *et al*., 2014; Brillas *et al*., 2003).

The objectives of this study are to investigate the organic content removal of methyl orange wastewater by using photovoltaic cell Electro-Fenton Oxidation and to find the optimum concentration of H_2O_2, current and electrolysis time for best removal efficiency.

Materials and methods

Materials

All the reagents used in this investigation are of an analytical grade and distilled water was used to prepare the synthetic dye solution. The stock solution of 20 mg/L of organic concentre was prepared from methyl orange (MO) using double distilled water. Figure 1 gives the chemical structure of MO. NaOH (98% purity) and HCl (37% purity) were purchased from India.

Electrodes and photovoltaic cell

The electrodes used in this study are aluminium and iron as anode and cathode, respectively. The dimension of the iron electrode is 10cm x 4cm x 0.3cm, that of the Aluminum electrode is 10cm x 4cm x 0.15cm. The active area of the electrodes was maintained as 30cm^2, and the inner electrode spacing was maintained as 6cm. The electrochemical cell (Figure 2a) is composed of four blocks of acrylic, and sheets of

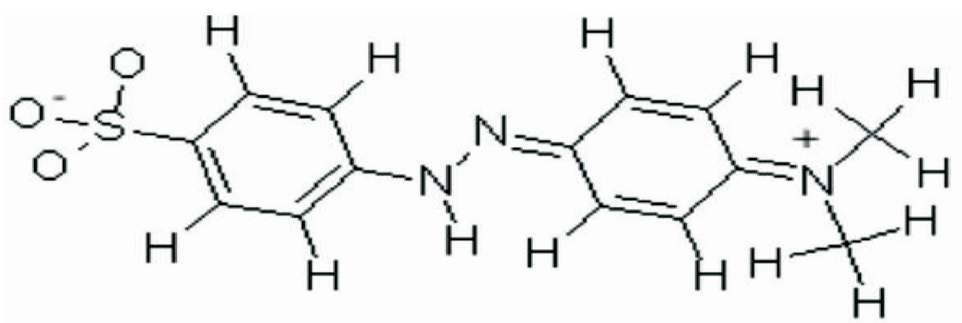

Figure 1: Description of the structure of methyl orange

silicone gaskets placed between them to prevent leaks. This cell shape permits two possible anode locations, at the anolyte section at the end of the plate (between the blocks of acrylic, the distance between anode and cathode was 1cm) is presented in Figure 2b.

Analytical Measurements

Methyl orange concentrate in aqueous solution was determined at maximum absorption wavelength (464.5 nm) using a UV-spectra meter (UV-1800 Shimadzu, Japan) spectrophotometer connected to a PC. Turbid meter (Lovibond, SN 10/1471, and Germany) and pH capacities were made through a pH meter (Model 2906, Jenway L_{td}, UK), correspondingly.

Experimental Procedure

In the Photovoltaic cell Electro-Fenton procedure, the experiment was performed in a batch reactor containing an 800 mL glass reactor with 300 mL of methyl orange. The solution was prepared by diluting the stock with distilled water, and the initial pH was tuned to 3 by 1N sulfuric acid and sodium hydroxide. The electrodes were connected to a DC power source (RXN-305D) from the photovoltaic cell (solar energy) and supplementary 0.2 gm of NaCl. DC voltage was conserved as 29.6 V. The reactor was operated for 10 minutes to sweep the Fe^{2+} ions, and subsequently, H_2O_2 was added with continuous stirring (Figure 3). The samples were allowed to rest at a regular time and centrifuged at 2000 rpm (to separate the sludge) for 10 min, and the supernatant was allowed to settle to estimate the amount of organics left in the

a

b

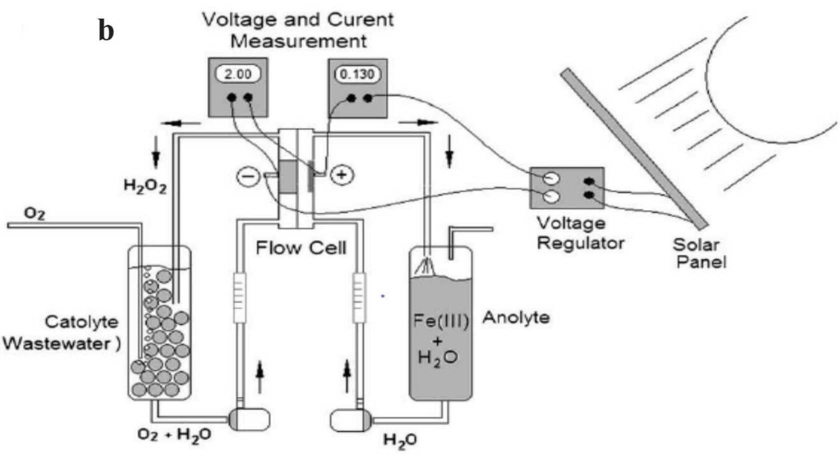

Figure 2: a) Main parts of the flow-cell. b) Hydraulic circuit, flow-cell, and solar panel

solution. Methyl orange wastewater reduction was expressed as the ratio of remaining organic matter at time t (C_t) to the initial concentration of the organic matter (C_0). Furthermore, the effect of H_2O_2 amount, current and time were studied. Before the commencement of the photovoltaic cell Electro-Fenton procedure, all the electrodes were cleaned with water to prevent further reaction. The electrodes were previously saturated in 1N hydrochloric acid for 1 hour, and subsequently in 1M sodium hydroxide for 1 hour. The electrodes were put in distilled water when not in use. After each use, the electrodes were eroded in 1N HCl and 1M NaOH to eliminate any likely contamination. The initial conductivity of the solution was 126 ms/cm. The significant bounds of the process is the narrow working pH (2.0–3.0) and iron residue in the conserved water (*Davarnejad et al*, 2014).

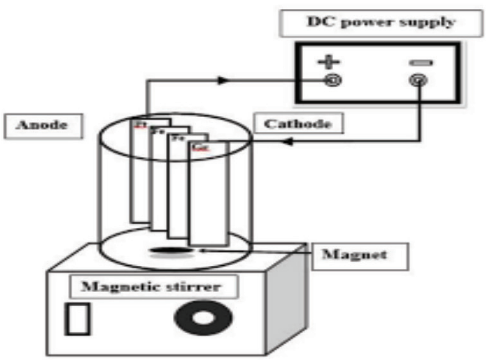

Figure 3: Schematic of the electrooxidation reactor

Energy consumption (kWh/m^3) is a significant aspect of such treatment approaches. Consequently, it can be envisioned using the following equation (Forat Yasir AlJaberi, 2018):

$$E = (U. I. t) / (1000. V) \tag{1}$$

Where U is the applied voltage (V), I is the current (A), t is the electrolysis time (h), and V is the volume of the methyl orange (m^3).

The variation of organic content in methyl orange concentrate through the electro-Fenton procedure was measured using a UV spectrophotometer (UV-1800 Shimadzu, Japan) at 464.5nm. The organic removal efficiency was determined using the following Equation (2):

$$\eta = \frac{C_0 - C_t}{C_0} x100 \tag{2}$$

Where η, percentage of organic matter removed (removal efficiency); Co, measured concentration before treatment (mg/L); Ct concentration value after treatment (mg/L).

Statistical Model

A statistical technique, response surface methodology (RSM) as well as the statistical program Minitab-17, were used to design the experiments and predict the influence of the operational factors individually and in interaction with each other. The main impacts of these factors: electrolysis time (X_1), rpm(X_2), Current(X_3) and H_2O_2 concentration (X_4) were studied according to their ranges shown in Table 1. The natural and coded values of the operational variables are listed in Table 2. These variables were chosen on the basis of preliminary experiments before starting the research methodology. The time chosen was because the electrochemical processes occur at a high speed. Similar situation with respect to the rest of the variables.

Table 1: Operational parameters

Parameters	Ranges
X_1: electrolysis time (min)	5-15
X_2: rpm	100-200
X_3: Current (A)	05-2
X_4: H_2O_2 concentration (ppm)	25-100

Results and discussion

The Effect of Current

Figure 4 presents the significance of current on the performance of the Photovoltaic cell Electro-Fenton system. The experiments were performed by varying the current from 0.5 to 2 Amps. The MO removal efficiency increases with increasing current, and the maximum removal efficiency of 98.46 % was achieved at 1.69 mA, 200 rpm for 15 minutes using 87.87 ppm H_2O_2, and the solution pH of 3. A higher current would lead to higher production rate of H_2O_2 on the surface of the cathode. As a result, free radicals will accumulate in the bulk solution, leading to a greater degradation of the oil in the wastewater. Similar consequences have been observed in the electro-Fenton treatment of ibuprofen (Liu *et al.*, 2018). The removal efficiency obtained at 0.5 mA is 91% in the heterogeneous electrooxidation process at the same condition. As the applied current to the electrode increases, the rate of activity of H_2O_2 is amplified proportionately, thereby increasing the rate of generation of $HO^{\bullet}$ radical to enhance the removal efficiency at 1.69 mA at 0.5 V and 1.0 V. Increasing current to 2 mA, reduces the recovery efficiency to 86% due to the occurrence of subsequent ide reactions: (i) growth of hydrogen at cathode (Equation (8)), (ii) oxidation of H_2O_2 at anode (Equation (4))

$$2H_2O_2 + 2e^- \rightarrow H_2 + HO^- \tag{4}$$

$$H_2O_2 \rightarrow O_2 + 2H^+ + 2e^- \tag{5}$$

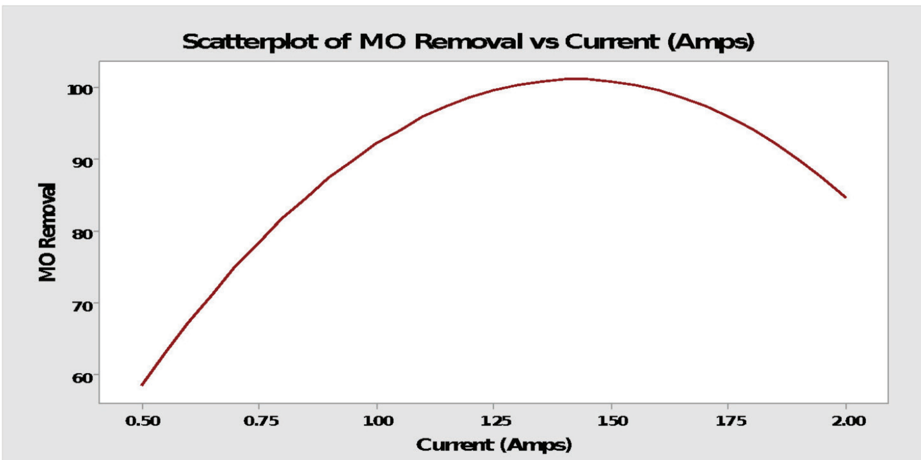

Figure 4: The effect of current on the recovery efficiencies of MO wastewater

The of Hydrogen Peroxide

In the direction of the best illustration, the role of H2O2 concentration on a photovoltaic fog Electro-Fenton of methyl orange wastewater in this study was occupied in electrical oxidation. Hydrogen peroxide, as a dominant source of OH•, is always considered as a critical parameter in the Fenton and Fenton-like processes. But, an excessive H_2O_2 dosage can scavenge hydroxyl radicals and reduce efficiency of the process; hence raising the cost of treatment (Jaafarzadeh et al., 2018). The concentration of H_2O_2 was varied during the electro-Fenton treatment within electrolysis time from 5 to 15 min). Figure 5 presents the removal efficiency represented by percentage MO removal when H_2O_2 concentration is increased from 25 to 87.87 mg/L. This is attributed to the generation of more OH radicals. The removal efficiency is increased gradually with increasing H_2O_2 concentration until it reaches the best value, then the value decreases, where the free root starts to react with hydrogen peroxide. Nevertheless, a further increase in the level of H_2O_2 declines the removal efficiency because the reaction rate plateaus. This trend could be attributed to the recombination of OH radicals in addition to the decay of H_2O_2 into water and oxygen. Thus, excess H_2O_2 may lead to the ingestion of active oxidized free radicals, which leads to further termination of the organolysis reaction. Because of that, a balance is reached between low levels of H_2O_2 and extra quantities of it. These observations are similar to the one reported by Haider *et al*. (2018).

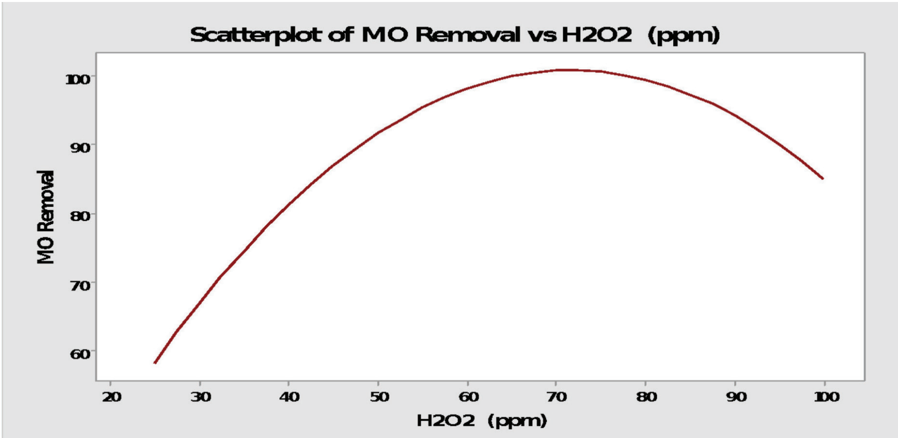

Figure 5: The H_2O_2 effect on the recovery efficiencies of MO wastewater

The Effect of Agitation Speed

Electro-Fenton oxidation of organic content in MO wastewater was undertaken employing varying degrees of agitation. It was observed that the MO removal increased linearly, with faster stirring speed. The maximum removal efficiency attained is 94.3% using 170 rpm for 15 min with 87.87 ppm H_2O_2 (Figure 6). Reduced organic eliminations were observed at speeds ≤ 100 rpm. This trend is mainly because an increase in the agitation speed increases the mass transfer (Sundarapandiyan *et al.*, 2009). Treatment of synthetic waste and wastewater from the dye and tanning industry was deliberate, and it was decided that stirring was one of the major factors in the electrochemical process (Szpyrkowicz, 2006). Therefore, the development in oxidation might be credited to higher mass transfer through agitation.

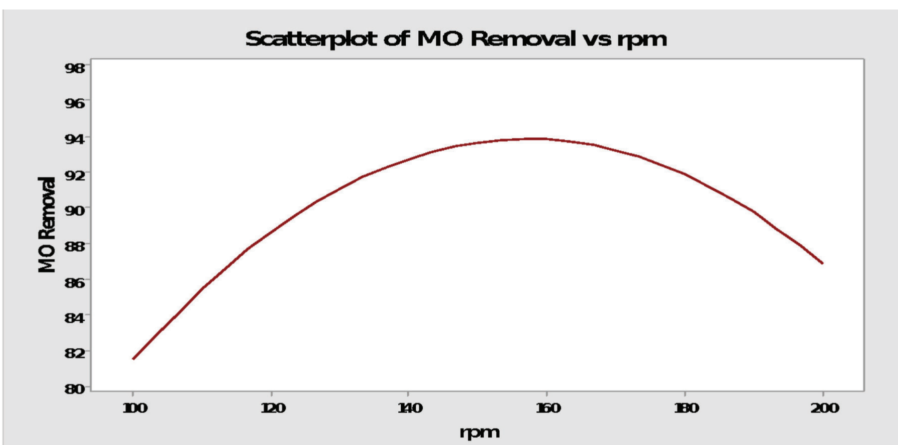

Figure 6: The rpm effect on the removal efficiencies of MO wastewater

The Effect of Electrolysis Time

The effect of electrolysis time was studied in a wastewater solution, which contains 20 ppm organic content (MO) at a pH of 3. The highest removal efficiency was achieved using 175 rpm ppm and 1.69 Am for 15 min. The removal efficiency of organic in methyl orange increased with longer electrolysis time (Fig .7). This result is in agreement with that of Gaber *et al.* (2012). This trend is because of an increase in the activity of the adsorption process, which occurred throughout the electro–Fenton reactor with increasing electrolysis time (Apaydin, 2014). Increasing the oxidation time leads to a step-by-step detoxification until the formation of mineral tin and CO_2 (Yong *et al.*, 2017). Therefore, complete mineralization was obtained.

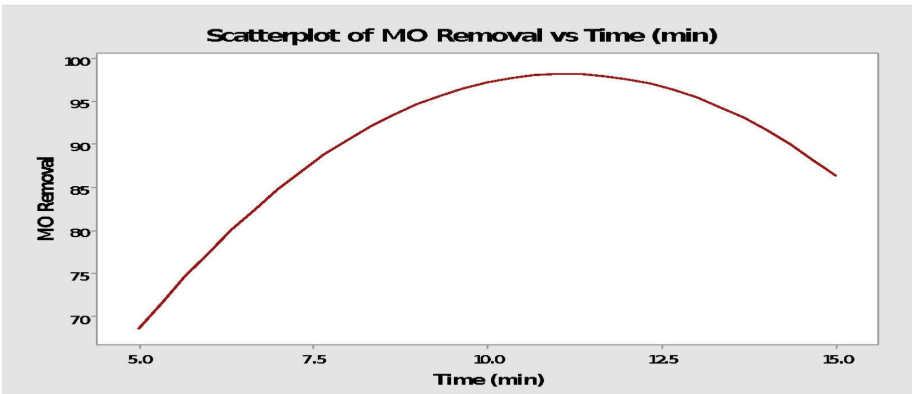

Figure 7: The electrolysis time effect on the removal efficiencies of MO wastewater

Optimization of Operational Variables

The best values of electrolysis time, rpm, and current and hydrogen peroxide were obtained using a statistical software program (Minitab-17). Figure 8 presents the results of the D-optimization study. The best value of the removal efficiency of the organic content is 98.46 %.

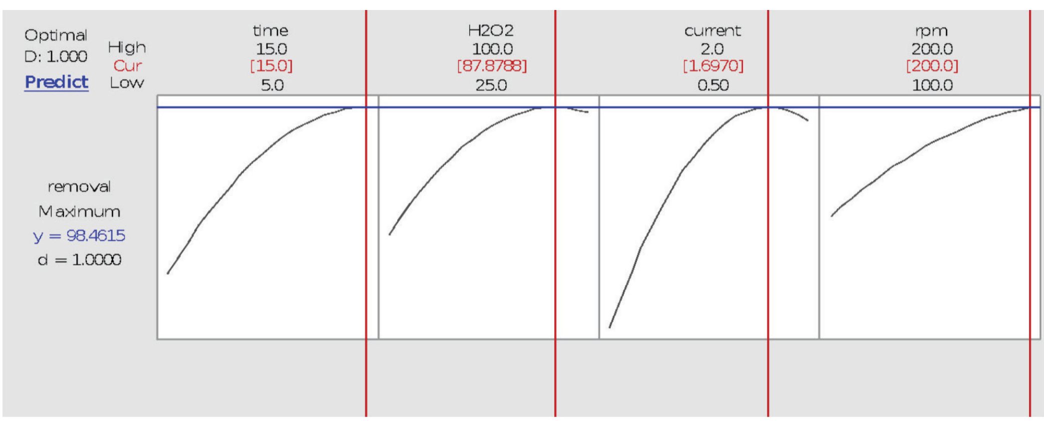

Figure 8: The optimum values of the operational variables and the validated values of the studied responses for the treatment of MO wastewater

Estimation of Energy Consumption

The acceptance of electro-Fenton technology in wastewater treatment is influenced by anode material, energy consumption and operating cost. Figure 9 presents the energy consumption, as a function of current and electrolysis time, during Photovoltaic cell Electro-Fenton removal of organic in methyl orange wastewater. It is observed, for all types of wastewater, that the values are relative to the applied time throughout Photovoltaic cell Electro-Fenton treatment (da Silva *et al.*, 2013).

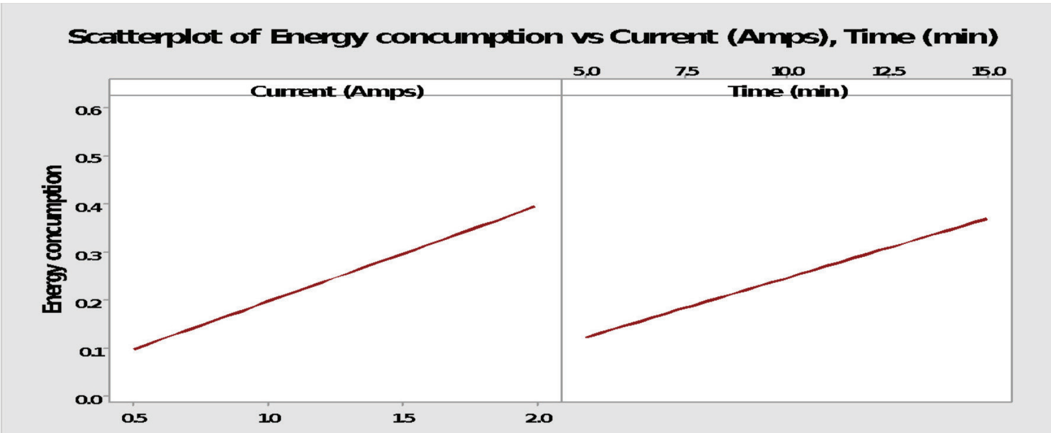

Figure 9: The effect of the operational variables on the energy consumption for the treatment of 20 mg/L methyl orange wastewater

Based on the optimum values of current and electrolysis time that was determined, the energy consumption is 39.4 kWh/m^3. Energy consumption is a function of the active variables during the electrooxidation to remove the organic substances. The increase in energy consumption is due to increased current and the electrolysis time. However, the current is more crucial for the determination of energy consumption, which agrees with the literature (Ganiyu *et al.*, 2018).

Regression Models

The relationship between the responses and the independent variables is obtained using the second-order model by a least-squares method as follows (Davarnejad *et al.*, 2014):

$$Y = B_0 + \sum_{i=1}^{q} B_i X_i + \sum_{i=1}^{q} B_{ii} X_i^2 + \sum_{i}\sum_{j} B_{ij} X_i X_j + \varepsilon \quad (3)$$

Where Y is the investigated response; X_1, X_2, to Xq are the working variables; B_o is the regression constant. B_i is the linear regression constant, B_{ii} is the squared regression constant, and B_{ij} is the relationship regression constant; ε is a random error. Table 3 presents the values of the operational variables, percentage elimination (MO removal efficiency), and energy consumption.

Conclusion

This paper investigates the possibility of using Photovoltaic cell electro-Fenton oxidation technology for the removal of organic content from methyl orange wastewater by directly connecting the electro-Fenton reactor to the photovoltaic generator. In this work, the Electro-Fenton technique is shown to be promising for the treatment of organic substances from wastewater. The operating parameters were changed to achieve the highest removal efficiency. The mathematical relationship obtained has a reasonably high regression constant for all the responses, showing the satisfactory adjustment of the second-order polynomial model. The maximum organic removal of 98% was achieved at a pH of 3, electrolysis time of 15 minutes, applied current of 1.69 mA, and 87.87 ppm of H_2O_2. It appears that Nano-porosity is essential to facilitate the decay of H_2O_2, which eventually favors the formation of hydroxyl radicals. Because of the good performance of Photovoltaic cell Electro-Fenton operation, this technique is promising for treatment of simulated wastewater**.**

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