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STUDY ON SOME PARAMETERS AFFECTING DEGRADATION OF METHYLENE BLUE IN WATER BY ELECTRO-FENTON USING Ti/PbO$_2$ ANODE

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Abstract. In this work, a system combining two advanced oxidation processes, namely electro-Fenton (EF) in which the hydroxyl radical $^{\bullet}OH$ is generated by reactions on the cathode and anodic oxidation (AO) in which $^{\bullet}OH$ is produced directly on the anode, was studied to evaluate the treatment of methylene blue (MB) dye in aqueous solution. This electrochemical system was equipped with a commercial carbon felt cathode and lead dioxide-coated titanium (Ti/PbO$_2$) anode. The effects of operating parameters such as pH, applied current (I), electrolysis time (t), catalyst concentration ([Fe^{2+}]) and initial MB concentration (C$_0$) on MB removal efficiency were investigated through monitoring MB concentration. The optimal process was achieved at the condition of [Fe^{2+}] = 0.1 mM; pH 3.0; [Na$_2$SO$_4$] = 0.05 M; i = 2.5 mA.cm^{-2} and after 60 min of electrolysis, where 92.19 % of MB was removed. This performance was much higher than that of single EF system using carbon felt cathode and Pt anode (73.77 %) or single AO system using Ti cathode and Ti/PbO$_2$ anode (58.04 %), which were also tested under optimal conditions. These experimental results have demonstrated that the combination of EF and AO is a prospective method for the destruction of persistent dyes.

Keywords: Combination, anodic oxidation, electro-Fenton, methylene blue removal, Ti/PbO$_2$, color removal, textile wastewater.

Classification numbers: 3.3.3, 3.4.2, 3.7.3

1. INTRODUCTION

Organic dyes, widely used in the textile industry, are currently of great concern because of their environmental sustainability and negative impacts on ecosystems and human health. In aqueous media, even at small concentrations (below 1 ppm), they are highly visible and prevent sunlight - an essential component in maintaining normal biological activities - from penetrating into the water, which poses harm to virtually all aquatic organisms [1]. It is estimated that more than 7 x 10^5 tons of dye-stuffs are produced annually on a global scale, particularly 12 % of that volume is discharged into receiving waters during manufacturing and coloration processes [2]. Methylene blue (MB) is among the cationic dyes, its first synthesis was as early as the late 19th century [3]. Like other persistent dyes, MB is very complex in structure and designed to possess high values of fastness under any exposure conditions, so its removal is a difficult challenge [2]. Although MB is not considered an extremely hazardous substance, its exposure to humans can cause eyes burn, vomiting, heart rate increase, diarrhea, shock, cyanosis, jaundice, quadriplegia, and tissue necrosis [4]. Moreover, its degradation mediators can also be fatal as they are carcinogenic and mutagenic agents [5]. Thus, it is very important to treat dyeing waste before discharging into water bodies. So far, many methods have been studied for removing colourants such as photolytic degradation [6], adsorption [5], biodegradation [7], oxidation [8], electro-coagulation [9], membrane filtration [10], and electrolysis, etc. However, these methods still have common limitations, which are the generation of secondary pollution or incomplete elimination of pollutants [11]. Therefore, it is necessary to study a more efficient approach to organic dyes treatment.

Among the organic dye processing technologies that have been investigated recently, electro-advanced oxidation processes (EAOPs) have emerged as an efficient, non-selective and environmentally friendly way to mineralize persistent organic pollutants such as dyes. The principle of EAOPs is to generate electrochemically in situ hydroxyl radical $^{\bullet}OH$ with very high standard redox potential ($E^\circ (OH^{\bullet}/H_2O) = 2.80 V/SHE$), which can promote the complete degradation of targeted contaminants into CO_2, H_2O and inorganic ions or acids [12]. One of the most popular EAOPs is electro-Fenton (e-Fenton), where a sufficient $^{\bullet}OH$ concentration is obtained based on the reaction between Fenton's reagent and iron salt (Eq. 1).

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^{\bullet} + OH^- \quad (k = 63\ L.mol^{-1}.s^{-1}) \tag{1}$$

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

$$Fe^{3+} + e^- \rightarrow Fe^{2+} \tag{3}$$

This process is an upgraded version of the classic Fenton because H_2O_2 is automatically produced by 2 electron reduction of O_2 from compressed air (Eq. 2) at the cathode and Fe^{2+} is continuously regenerated by the cathodic reduction of Fe^{3+} (Eq. 3). Hence, it consumes less amount of reagent, generates less amount of ferric sludge but has higher efficiency compared to the classic version [13]. The contaminant degradation efficiency relies on the amount of generated hydroxyl radicals, so highly depends on the nature of cathode materials. In general, the high stability, conductivity and overpotential for hydrogen evolution reaction (HER), low catalytic activity for H_2O_2 decomposition are required for cathode materials [14]. Therefore, the non-toxic, and inexpensive carbonaceous materials such as activated carbon fiber, carbon felt (CF), carbon sponge, reticulated vitreous carbon, etc. were widely used as the cathode for EF process [15]. Furthermore, along with the role of such a cathode, if the EF system uses an anode capable of catalyzing in situ $^{\bullet}OH$ generation, then the amount of $^{\bullet}OH$ will be significantly increased, thus improve the rate of contaminant degradation, thereby reducing treatment time and energy consumption [16]. Indeed, non-active anodes with high overvoltage for oxygen evolution reaction (OER) such as PbO_2, SnO_2, boron-doped diamond (BDD), Ti/Pt, Ti/PbO_2, etc. can electrocatalyze the generation of $^{\bullet}OH$ (Eq. 4) and physisorb it on the surface [17]. This phenomenon occurring at the anode is called anodic oxidation (AO) and recent studies have shown that AO alone can effectively destroy different dyes, such as acid green 50 [18], alphazurine A [19], Alizarin Red S [20] and MB [21].

$$H_2O \rightarrow (OH^{\bullet})_{ads} + H^+ + e^- \tag{4}$$

The combination of EF-AO processes in an electrolyser has been studied by some research groups, such as: Vasconcelos *et al*. [22] used reticulated vitreous carbon as cathode and BDD as anode of a filter-press flow cell for degradation of Reactive Black 5 dye; Wang *et al*. [23] studied the degradation of perfluorooctanoic acid by the combination of EF-AO in an system using FeMn-doped carbon cathode and BDD anode; Tian *et al*. [24] synthesized Ti-PbO_2 material to fabricate the anode and used the graphite felt-polytetrafluoroethylene/carbon black gas diffusion cathode in the EF-AO system to decompose Rhodamine B. However, the combination of AO using Ti/PbO_2 anode and EF using CF cathode to degrade MB dye has not been reported.

The objective of this paper is to study the degradation of MB by the combination of AO using commercial Ti/PbO_2 anode and EF using CF cathode (these materials are inexpensive and easy to apply to larger electrochemical systems), specifically investigating the influence of several factors, such as pH, current intensity, catalyst concentration, MB concentration on the MB removal efficiency. The performance of 3 processes: EF, AO and the combination of EF-AO was also evaluated to find the best method to remove MB.

2. MATERIALS AND METHODS

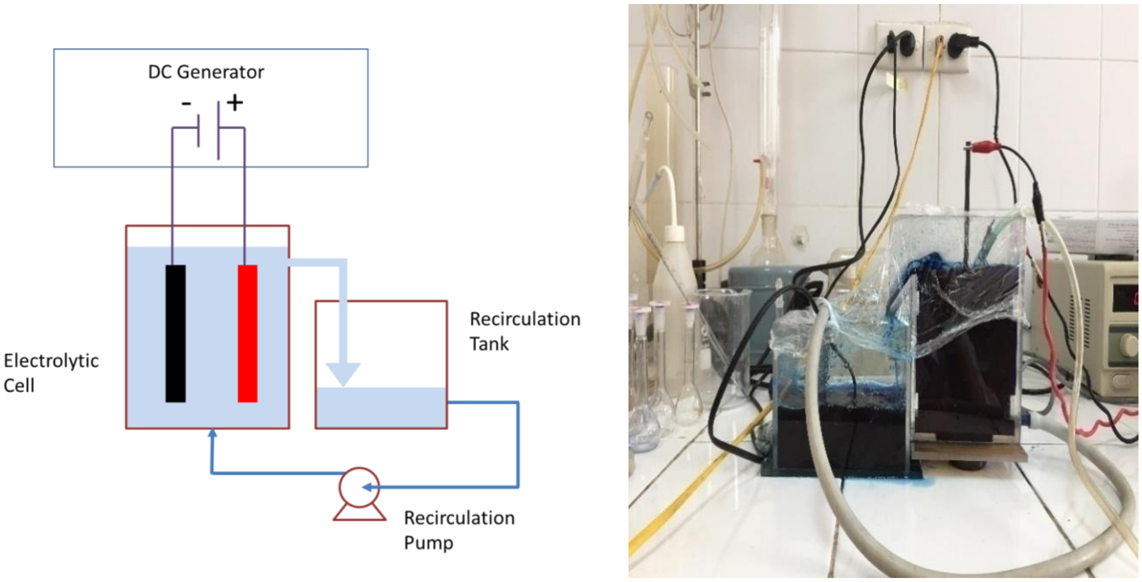
2.1. Materials

Carbon felt was supplied by A Johnson Matthey Co., Germany; Ti/PbO_2 was purchased from Baoji Qixin Titanium Co., Ltd., China and Pt mesh was provided by Shaanxi Elade New Marerial Technology Co. Ltd., China. MB of analytical grade ($C_{16}H_{18}ClN_3S$, Sigma Aldrich NY, USA) was used without further purification. Iron (II) sulphateheptahydrate (99.5 %, Merck) acted as a catalyst, while sodium sulphate (99 %, Merck) was used as a supporting electrolyte. To change the pH of the solution, we chose sulfuric acid (98 %, Merck). The ultrapure water obtained from a Millipore Milli-Q system with resistivity >18 MΩ.cm was used to prepare all solutions.

2.2. Electrochemical systems

The electrochemical degradation experiments of MB were carried out in a batch mode with a rectangular Plexiglass reactor (21 mm (width) × 150 mm (length) × 180 mm (height)). Rectangular electrodes having a dimension of 100 mm × 150 mm were vertically fixed on a perforated Plexiglas plate placed 20 mm from the bottom of the cell. The distance between the electrodes was 1 cm. Mixing in the reactor was accomplished by circulating water through the cell in a continuous mode by means of a pump operating at a constant speed of 1000 $mL.min^{-1}$ (Figure 1). The Plexiglass circulation tank had a dimension of 120 mm (width) × 150 mm (length) × 70 mm (height). In all experiments, a total volume of 1.0 L of contaminated water was used. The working volume of the electrolytic cell was 540 mL, while 460 mL was required for the recirculation tank.

The anode and cathode materials used in the case of AO-EF combination were Ti/PbO_2 and CF, in the case of AO alone were Ti/PbO_2 and Pt mesh, in the case of EF alone were Pt mesh and CF, respectively. The solution was continuously aerated 30 min before and during the electrolysis (at about 1 $L.min^{-1}$) to ensure a constant supply of oxygen diffusing into the solution throughout the experiment for producing H_2O_2 from reaction (2). Before the electrolysis initiation, an amount of ferric ion catalyst was introduced into the solution. Sulfuric acid was required for pH adjustment (around pH 3.0). The anode and cathode were connected to the positive and negative outlets of a DC power supply, respectively (model VSP4030, B&K Precision, CA, US). The current was kept constant during the tests.



(a)

(b)

Figure 1. Scheme (a) and real image (b) of e-Fenton combined with anodic oxidation Ti/PbO_2 system on lab scale.

2.3. Experimental procedure

The influence of the main factors (pH, current applied, Fe^{2+} catalyst concentration, MB concentration) on the MB removal efficiency was investigated to find out suitable conditions for the AO-EF process. During these tests, the MB concentrations were monitored to examine the performance of the AO-EF system. Because the dye concentration in textile wastewater is usually in the range of 50 - 250 $mg.L^{-1}$ [25], synthetic MB solutions of 50 $mg.L^{-1}$ were used in most of the experiments (except for the study on the effect of MB concentration on MB removal efficiency) to minimize external factors affecting the results. The effect of pH value was investigated in the range of 2 - 6. The current intensities ranged from 0.1 to 0.5 A (current densities from 0.67 $mA.cm^{-2}$ to 3.33 $mA.cm^{-2}$). The treatment time up to 60 min was tested. Likewise, the effects of MB concentration (20, 30, 40, 50, 60 mg.L^{-1}) and Fe^{2+} concentration (0.05, 0.1, 0.2, 0.5 mM) were investigated.

The performance of MB removal using the AO-EF process was then compared with that of the AO and EF processes alone at the same applied current density. The optimal pH value and Fe^{2+} concentration found above were used for the AO-EF process. For single AO and EF processes, the optimal conditions were determined in previous works by the same authors, namely [Fe^{2+}] = 0.1 mM, pH 3.0 for the EF process [26] and pH 3.0 for the AO process [27]. In these tests, MB concentrations were determined at different time points, ranging from 0 to 60 min. All experiments were repeated 3 times and reported values are the mean of experimental data.

1.1 Analytical methods and apparatus

The pH of the solution was measured with a Hanna HI 991001 pH-meter. According to Wang *et al*. [28], the products of MB degradation by AOPs are: Cl^-, NO_3^-, SO_4^{2-}, HSO_3^-, non-toxic lower molecular weight intermediates and very small amounts of benzothiazole, phenol, so in this work, MB removal was evaluated instead of mineralization. MB removal efficiency was then determined from Eq. (4)

$$H(\%) = \frac{C_0 - C}{Co} x100\% = \frac{A_0 - A}{A_0} x100\%$$

where: C and C_0 are MB concentration at time t and initial time, respectively (mg.L^{-1}); A andA_o are the absorbance value of solutions at time t and initial time, respectively.

MB concentration was analyzed by absorbance measurement at λ = 664 nm using UV-VIS spectrophotometer (Labomed UVS-2700, USA).

3. RESULTS AND DISCUSSION

3.1. The effect of some factors on the treatment efficiency of the AO-EF process

3.1.1. Effect of pH

In the EF process, the H^+ concentration influences the amount of H_2O_2 formed (by reaction (2)), which then controls the generation of $OH^\bullet$ radicals. The removal of MB by the AO process is favored at acidic pH (~ 3.0) [21]. Hence, pH is one of the most important factors affecting the AO-EF process. High pH will reduce the concentration of Fe^{2+} catalyst due to the formation of $Fe(OH)_3$ precipitate, while too low pH will lead to H_2O_2 decomposition [23] and in both cases, the efficiency of the EF process is drastically reduced. Therefore, to clarify the influence of pH on MB decay efficiency, we only changed the initial pH of the solution in the range of 2 - 6 and other parameters were kept constant: I = 0.3 A, [Fe^{2+}] = 0.1 mM, t = 60 min, MB concentration C_0 = 50 mg.L^{-1}.

As can be seen from Figure 2, pH greatly affected the MB removal efficiency, and the peak of the treatment was obtained at pH 3.0 with a maximum efficiency of 84.21 %. These results can be explained as follows: the MB removal efficiency will depend on the amount of $OH^\bullet$ produced by the

Fenton reaction (Eq. 1) and by the AO process (Eq. 4). When the pH was decreased from 6 to 3, the concentration of H^+ ions increased, leading to an increase in the amount of H_2O_2 produced by the reduction of O_2 on the cathode (Eq. 2), and thus, a growth in the quantity of $OH^\bullet$ radicals generated by Eq (1). A low-pH medium also avoided the formation of amorphous $Fe(OH)_3$ precipitate which reduced MB removal efficiency because it was less reactive than Fe^{2+} and could partially cover the cathode surface, inhibiting there generation of the catalyst at this electrode (Eq. (3)) [29]. Moreover, when the pH value was above 5, oxidants that were weaker and more selective than the $OH^\bullet$ radical such as ferryl ions (e.g., FeO^{2+}) could also be formed according to Eq. (5) [30]. In addition, H^+ in the acid solution could inhibit the reaction of oxygen evolution at the anode and the decomposition of $OH^\bullet$ radical, which improved the MB removal efficiency by the AO process [21]. So, the lower the pH value, the higher the MB removal efficiency.

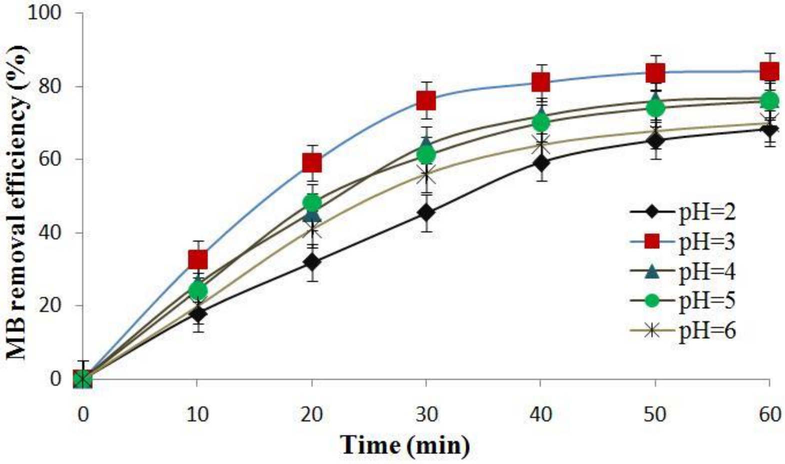


Figure 2. Effect of pH on the MB removal under experimental conditions: Na_2SO_4= 0.05 mol L^{-1}; T = 25 C°; $[Fe^{2+}]$ = 0.1 mmol L^{-1}; I = 0.3 A, C_0 = 50 $mg.L^{-1}$

$$Fe^{2+} + H_2O_2 \rightarrow Fe(IV)\ (e.g., FeO^{2+}) + H_2O \quad (5)$$

However, when the pH was decreased from 3 to 2, the MB removal efficiency did not increase but decreased, possibly because at very acidic pH, below pH 3.0, a reaction can occur between H^+ and electrogenerated H_2O_2 to form an oxonium ion ($H_3O_2^+$) (Eq. 6) that impeded the reactivity with Fe^{2+}, making less $OH^\bullet$ to be produced [31]. Also, the low pH could lead to in situ decomposition of H_2O_2 (Eq. (7)) and thus, a significant decrease in the concentration of H_2O_2 in the medium [31].

$$H_2O_2 + H^+ \longrightarrow H_3O_2^+ \quad (6)$$

$$H_2O_2 + 2H^+ + 2e^- \longrightarrow 2H_2O \quad (7)$$

Therefore, the optimal pH for this process is 3.0 and this pH will be used for all subsequent experiments. This result is similar to the case of MB decomposition by EF alone using stainless steel mesh electrodes [32] and by AO alone using SnO_2 electrode [21].

3.1.2. Effect of applied current and electrolysis time

In electrochemical processes in general, EF and AO in particular, the applied current intensity plays a significant role in the oxidation efficiency of the degradation process. The experiment was conducted with different currents: 0.1 A; 0.2 A; 0.3 A; 0.4 A; 0.5 A, while other parameters were kept constant: pH = 3.0, T = 25 °C (room temperature), $C_0 = 50$ mg.L^{-1}, $[Fe^{2+}] = 0.1$ mM.

Figure 3 demonstrated that the MB decomposition rate was accelerated as the value of applied current and electrolysis time increased. Specifically, after 60 min of electrolysis, when the applied current was changed from 0.1 A to 0.5 A, the MB removal efficiency increased from 77.77 % to 97.09 %. At I = 0.1 A, when the electrolysis time was increased from 10 min to 60 min, the MB decomposition efficiency increased from 14.10 % to 77.77 %. A similar trend was observed for other currents (0.2 A - 0.5 A). This result can be explained by Faraday's law: the volume of substance released/deposited at the electrodes is directly proportional to the current and electrolysis time, thus the longer the electrolysis time or the higher the current, the more H_2O_2 was produced at the cathode (Eq. 2) and the faster the Fe^{2+} catalyst was regenerated (Eq. 3), as a result, the more $OH^\bullet$ radicals was created by the Fenton reaction (Eq. 1), at the same time, the more $(OH^\bullet)_{ads}$ is generated at the anode (Eq. 4) [31], leading to an increase in the MB decomposition efficiency.

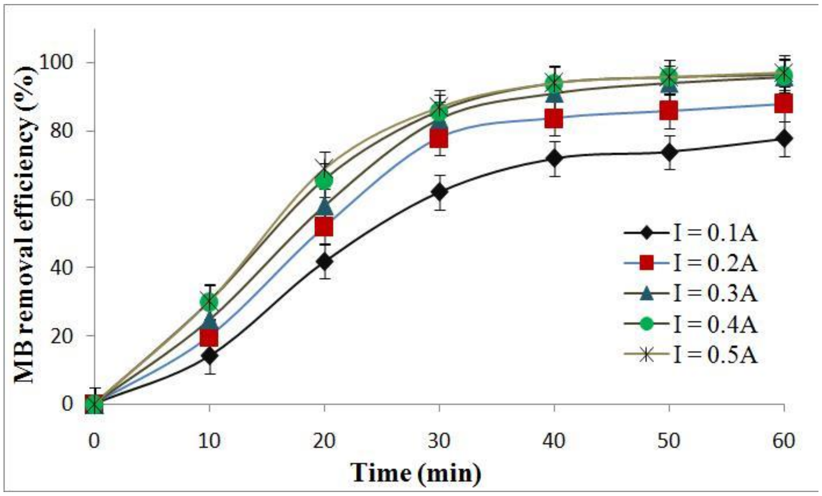


Figure 3. The influence of applied current on MB removal under experimental conditions: Na_2SO_4 0.05 mol L^{-1}; T = 25 °C; $[Fe^{2+}] = 0.1$ mmol L^{-1}; pH 3.0, $C_0 = 50$ mg.L^{-1}.

Also in Figure 3 it can be seen that, during the first 40 min of electrolysis, the MB removal rate increased very rapidly then slowed down for all applied intensities. This effect was due to the fact that the operating parameters remained constant throughout the entire test period; the production of $OH^\bullet$ was constant [33, 34]. However, most of the MB molecules were mineralized in the first 40 min, so the ratio between organics and radicals got lower after 40 min, meaning that $OH^\bullet$ started to participate in the wasting reactions (8 -10) [35], so the MB removal rate slowed down.

$$OH^\bullet + OH^\bullet \rightarrow H_2O_2 \quad (8)$$

$$OH^\bullet + H_2O_2 \rightarrow H_2O + HO_2^\bullet \quad (9)$$

$$OH^\bullet + HO_2^\bullet \rightarrow H_2O + O_2 \quad (10)$$

Thus, for high MB removal performance, the current between the two electrodes must be 0.3 A or the current density should be 2.5 mA.cm^{-2}.

3.1.3. Effect of ferrous ion concentration

The amount of Fe^{2+} catalyst is also a main factor affecting the oxidation efficiency of the AO-EF process. The effect of Fe^{2+} concentration on the oxidation of 50 mg.L^{-1} MB was investigated under the following conditions: pH 3.0, applied current intensity of 0.3 A and Fe^{2+} concentration from 0.05 to 1 mmol.L^{-1}.

It is clear from Figure 4 that the best MB removal efficiency (92.19 %) was achieved when the Fe^{2+} concentration was 0.1 mmol.L^{-1}. A slight decrease in pollutants was followed by an increase in the amount of catalyst, from 0.05 mmol.L^{-1} to 0.1 mmol.L^{-1}. This is reasonable because according to the law of mass action, the more Fe^{2+} content, the more $^{\bullet}$OH radicals were produced, which increased the rate of reaction in Eq. (1). However, when the Fe^{2+} concentration exceeded 0.1mmol.L^{-1}, it led to a decrease in MB removal performance. This could be due to the reaction between the excess ferrous ion and the $^{\bullet}$OH radicals (Eq. (11) and (12)) that consumed the Fenton reagent, leading to a decrease in MB removal efficiency. In addition, the produced Fe^{3+} could also react with H_2O_2 (Eq. (13) and (14)) resulting in reduced MB removal [36].

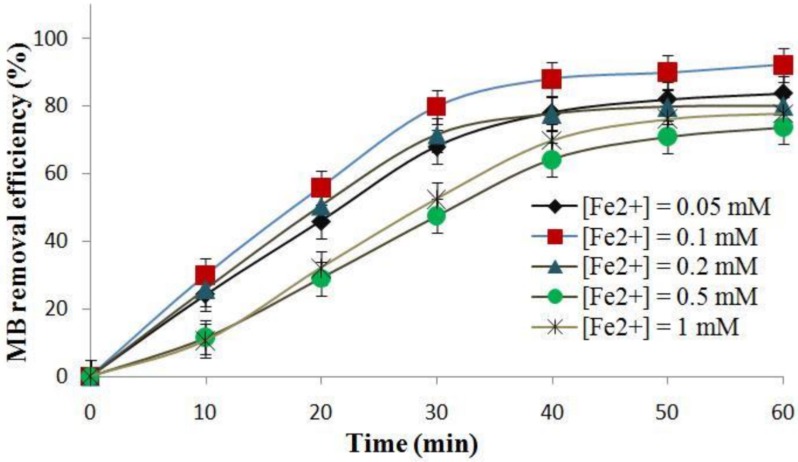


Figure 4. The effect of Fe^{2+} concentration on MB removal under experimental conditions: [Na_2SO_4] = 0.05 mol.L^{-1}; T = 25 °C; pH = 3.0; I = 0.3 A, C_0 = 50 mg.L^{-1}

$$Fe^{2+} + HO^{\bullet} \rightarrow Fe^{3+} + OH^{-} \quad (11)$$

$$Fe^{2+} + {}^{\bullet}OH \rightarrow Fe^{3+} + HO^{-} \quad (12)$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe{-}OOH^{2+} \quad (13)$$

$$Fe{-}OOH^{2+} \rightarrow Fe^{2+} + HO_2^{\bullet} \quad (14)$$

Furthermore, using a high amount of catalyst is not economically viable because it produces a large amount of ferric oxide sludge, resulting in additional costs for another sludge separation and disposal process [37]. Therefore, the optimal catalyst concentration for this experiment is 0.1 mmol.L^{-1}.

3.1.4. Effect of MB concentration

The removal of MB from aqueous solutions at different concentrations is illustrated in Figure 5. The obtained results indicate an inverse trend between MB concentration and MB removal efficiency. Indeed, when the MB concentration was increased from 20 mg.L^{-1} to 60 mg.L^{-1}, the treatment rate decreased from the highest (99.50 %) to the lowest (75.80 %), in contrast to the upward trend of the absolute amount of contaminants eliminated, from 19.9 mg.L^{-1} to 45.48 mg.L^{-1}. This fact may be due to the presence of a large amount of organics (MB and its degradation by-products) in the electrolyte solution at high MB concentration, while the same amount of •OH was generated by reactions (1) and (4) in these tests, so according to the law of mass action, the rate of MB removal was reduced. Moreover, at high MB concentrations, dimer molecules could be formed in sequential reactions of dye molecules. These dimers were difficult to break down, so the MB removal efficiency was reduced as observed above [32]. This result is consistent with the research results by Loloei and Rezaee [32] on MB decolorization by EF process using stainless steel mesh electrodes.

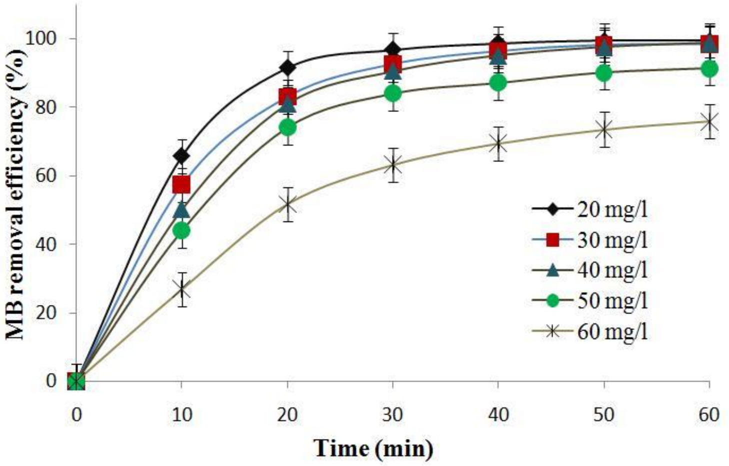


Figure 5. The effect of MB concentration on treatment efficiency under experimental conditions: [Na_2SO_4] = 0.05 mol.L^{-1}; T = 25 °C; [Fe^{2+}] = 0.1 mmol.L^{-1}; I = 0.3 A, pH 3.0.

Also in Figure 5, it can be seen that the decrease in MB removal efficiency was non-linear; its rate is gradually raised with the increase of the MB initial concentration. For example, after 20 min of electrolysis, the MB removal efficiency decreased to 91.53 %, 83.04 %, 80.89 %, 74.06 % and 51.63 % for the MB initial concentration of 20, 30, 40, 50 and 60 mg.L^{-1}, respectively. This comportment could be explained as follows: the higer the MB initial concentration, the more by-products were generated, leading to the consumption of •OH by these by-products and the reduction in MB removal efficiency. The adsorption of MB on the electrode surface also occured, but the effect of this phenomenon is weak (less than 12 % for 1 h) [38].

3.2. Comparison of 3 processes: e-Fenton, anodic oxidation and e-Fenton combined with anodic oxidation

To compare the performance of the AO-EF process with the AO and EF processes alone, 50 mg.L^{-1} MB aqueous solutions were used for all three cases, the same current density (2.5 mA.cm^{-2}) was applied between the two electrodes of the three systems. The other optimal conditions applied to the AO-EF and EF systems were [Fe^{2+}] = 0.1 mM, pH 3.0, for the AO system was pH 3.0. The results were depicted in Figure 6.

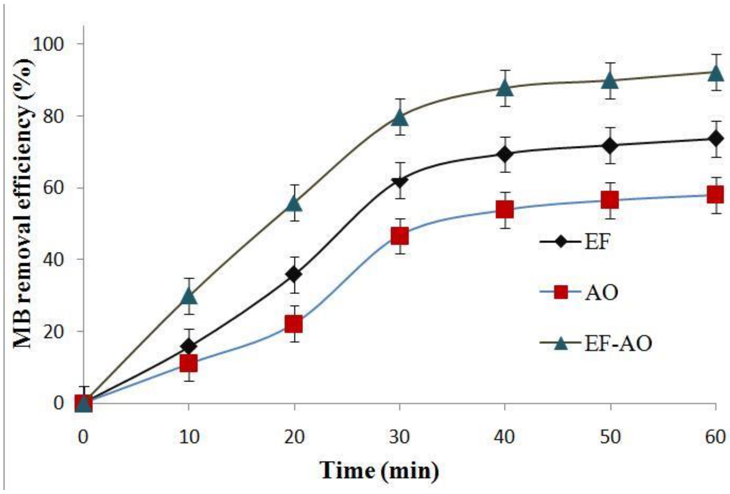


Figure 6. The MB removal performance of 3 processes: AO-EF, EF and AO under experimental conditions: pH 3.0, [Na_2SO_4] = 0.05 mol.L^{-1}; T = 25 °C; Fe^{2+} = 0.1 mmol.L^{-1}; i = 2.5 mA.cm^{-2}, C_0 = 50 mg.L^{-1}

The obtained results show that under the same conditions, the AO-EF process had the highest efficiency of 92.19 %, while the single EF and AO processes had lower treatment efficiency, 73.77 % and 58.04 %, respectively. This result is reasonable because MB concentration and current density were the same in all three cases, so the case that produced more •OH radicals had higher MB removal efficiency. For the EF process, the •OH radicals were produced only by the Fenton reaction (Eq. (1)) between Fe^{2+} and H_2O_2 generated at the cathode (Eq. (2)). For the AO process, the •OH radicals were generated only by the oxidation of water at the anode (Eq. (4)). For the AO-EF process, the •OH radicals were generated by both the Fenton reaction and the water oxidation at the anode, then the amount of •OH radicals created is the most, so the MB removal efficiency is the highest.

The MB removal efficiency by the AO-EF process in this study is also higher than that of the AO and EF processes in other studies. Indeed, Loloei and Rezaee removed MB from an aqueous solution of 60 mg.L^{-1} concentration by the EF process using stainless steel mesh electrodes, and the MB removal efficiency was achieved at about 85 % after 60 min at a current density of 4 mA.cm^{-2} [32]. Baddouh *et al*. degraded MB in an aqueous solution of 50 mg.L^{-1} concentration by the AO process using SnO_2 as anode, Pt as cathode and MB remove efficiency was achieved at about 55 % after 60 min when applying a current density of 30 mA.cm^{-2} [21].

This result shows that the combination of EF and AO processes in the same reactor gives a much higher MB removal efficiency from aqueous solutions than either process alone.

4. CONCLUSION

This study is a proof that the combination of EF and AO processes is an effective method in removing MB from aqueous solution. The results obtained from experiments under various conditions demonstrate that pH, applied current, Fe^{2+} concentration and MB concentration have a great influence on MB removal efficiency. A single EF process or AO process could only remove 73.77 % and 58.04 % of 50 $mg.L^{-1}$ MB from the simulated solutions, which is a requirement for the set-up of 2 processes in 1 reactor. That could not only improve the treatment efficiency, but also reduce the operating cost. Indeed, it was found that under optimal conditions: pH 3.0, $[Na_2SO_4]$ = 0.05 M, $[Fe^{2+}]$ = 0.1 mM, i = 2.5 $mA.cm^{-2}$, the AO-EF combination could remove 92.19 % MB after 60 min of electrolysis. Therefore, this model is very prospectively effective if it can be applied to actual wastewater treatment of some textile plants in practice.

Further studies are needed to understand the mechanism of MB degradation and identify the by-products.

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CRediT authorship contribution statement: Le Thanh Son: methodology, investigation, conceptualization, writing - review & editing; Nguyen Tran Dung: data curation; Tran Thu Huong: formal analysis; Nguyen Tran Dien: supervision; Dao Phuong Uyen: investigation and writing original draft.

Declaration of competing interest: The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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