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Evaluation of Photo-Fenton and Photo-Fenton-Like Methods for Removing Direct Orange 118 Dye from Wastewater

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Keywords:

Decolorization; Direct Orange 118 dye; Kinetics; Photo-Fenton; Photo-Fenton-like.

Highlights:

- Using a photo-Fenton process (UV/Fe²⁺/H₂O₂) for treating wastewater from direct orange 118 dye.
- Using a photo-Fenton-like process (UV/Fe³⁺/H₂O₂) for treating wastewater from direct orange 118 dye.
- Using the factors: the time of radiation, solution pH, H₂O₂ concentration, Fe ions concentration, solution temperature, and Direct Orange 118 dye concentration.

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Abstract: Wastewater treatment has used efficient technology methods that can remove any pollutant. Advanced oxidation is one of the alternative methods to reduce wastewater loads from any water pollution source. The present study was conducted to evaluate the possibility of applying photo-Fenton (UV/Fe²⁺/H₂O₂) compared to photo-Fenton-like (UV/Fe³⁺/H₂O₂) using Fe²⁺ or Fe³⁺ as a catalyst for removing Direct Orange 118 (DO 118) dye from wastewater. The experiments were conducted in a batch system under different parameters, such as irradiation time, pH, H₂O₂ concentration, concentration of Fe²⁺ or Fe³⁺, temperature, and the dye's initial concentration. The highest decolorization of DO 118 was 97.817% and 91.244% for photo-Fenton and photo-Fenton-like processes, respectively, at optimum conditions of pH 3, temperature= 30 °C, and dye concentration= 10 mg/l within irradiation time= 105 min. On the other hand, for the photo-Fenton process, Fe²⁺ concentration was 50 mg/l, H₂O₂ concentration was 400 mg/l, and for the photo-Fenton-like process, Fe³⁺ concentration was 80 mg/l, and H₂O₂ concentration was 600 mg/l. The discoloration kinetics study followed pseudo-first-order kinetics for photo-Fenton and photo-Fenton-like processes. The present study results indicated that the photo-Fenton and photo-Fenton-like processes can be utilized for treating DO 118 dye from wastewater.

تقييم تقنيات فوتوفنتون الضوئية وفوتوفنتون الضوئية المشابهة لإزالة لون الصبغة البرتقالية ١١٨ (Direct Orange 118) من مياه الصرف الصحي

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الخلاصة

لإدارة معالجة مياه الصرف الصحي تستخدم طرق تقنية ذات كفاءة والتي بإمكانها إزالة أي ملوث. تعتبر الأكسدة المتقدمة إحدى الطرق البديلة لتقليل أحماض مياه الصرف الصحي والاثنية من أي مصدر تلوث، الدراسة الحالية تم إجراؤها لغرض تقييم إمكانية استخدام طريقة فوتوفنتون الضوئية (UV/Fe²⁺/H₂O₂) وبالمقارنة مع فوتوفنتون الضوئية المشابهة (UV/Fe³⁺/H₂O₂) باستخدام Fe³⁺ or Fe²⁺ كعامل محفز لإزالة لون الصبغة البرتقالية ١١٨ (Direct Orange 118) من مياه الصرف الصحي. التجارب أجريت بنظام الدفعات تحت تأثير عوامل مختلفة مثل زمن التعرض للإشعاع، الدالة الحامضية، تركيز البيروكسيد H₂O₂، تركيز أكسيد الحديد الثنائي Fe²⁺ أو أكسيد الحديد الثلاثي Fe³⁺، درجة الحرارة، وتركيز الأولي للصبغة. أعلى نسبة إزالة للصبغة البرتقالية ١١٨ كانت ٩٧,٨١٧ ٪ و ٩١,٢٤٤ ٪ بطريقة فوتوفنتون الضوئية وفوتوفنتون الضوئية المشابهة على التوالي في الظروف المثلى للدالة الهيدروجينية (٣)، درجة الحرارة (٣٠) درجة مئوية، وتركيز الأولي للصبغة (١٠) ملغم /لتر خلال زمن التعرض للأشعاع (١٠٥) دقيقة لكلا الطريقتين، من ناحية أخرى بالنسبة لطريقة فوتوفنتون الضوئية كان تركيز أكسيد الحديد الثنائي Fe²⁺ (٥٠) ملغم /لتر، تركيز البيروكسيد H₂O₂ (٤٠٠) ملغم /لتر، وبالنسبة لطريقة فوتوفنتون الضوئية المشابهة كان تركيز أكسيد الحديد الثلاثي Fe³⁺ (٨٠) ملغم /لتر، تركيز البيروكسيد H₂O₂ (٦٠٠) ملغم /لتر فتم إجراء دراسة الموديلات الحركية لإزالة اللون والتي اتبعت نموذج الدرجة الأولى الزائفة (pseudo-first-order kinetics) لطريقة فوتوفنتون الضوئية وفوتوفنتون الضوئية المشابهة. الدراسة الحالية أظهرت إمكانية استخدام طريقة فوتوفنتون الضوئية وفوتوفنتون الضوئية المشابهة لإزالة لون الصبغة البرتقالية ١١٨ (Direct Orange 118) من مياه الصرف الصحي.

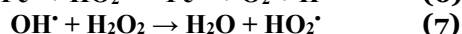
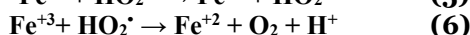
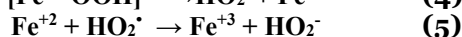
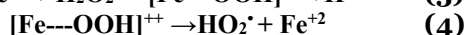
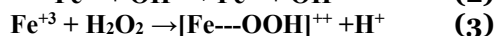
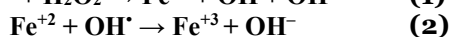
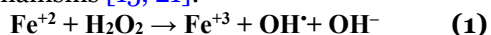
الكلمات الدالة: إزالة اللون، الصبغة البرتقالية ١١٨، موديلات حركية، فوتوفنتون الضوئية، فوتوفنتون الضوئية المشابهة.

1. INTRODUCTION

Dyes are an essential component in the textile industry, but the presence of residues in industrial effluent water leads to their presence in water resources. Colored water makes it difficult for the sun's rays to reach their depths, hindering photosynthesis [1, 2]. The most important thing is the toxicity of these dyes, their danger to aquatic animals and plants, and their threat to wildlife and humans [3]. The chemical structure of the dyes prevents them from being easily degraded in water or under sunlight. The nitrogenous azo dye is at the forefront of difficult-to-disintegrate dyes and contains many types of dyes, including the orange dye [4]. The advanced oxidation process is efficient for the decolorized dye discharged from textile industries. Furthermore, other industries, such as cosmetics, paper, and pharmaceuticals, also produce colored wastewater [5, 6]. Whereas dyes are very harmful substances to aquatic life, the advanced oxidation process can break down dye elements into a non-harmful substance. The advanced oxidation method is easy to use and handle and has high efficiency, low cost, few sediment residuals, and no need for complex reagents and devices [7]. In general, during the advanced oxidation method, hydroxyl radicals (OH•) are responsible for the oxidation process. Hydroxyl radicals (OH•) can destroy organic compounds; almost all the resultant components are water and carbon dioxide [8]. There are many kinds of advanced oxidation methods, such as photo-Fenton, sono-Fenton [9], ozone/Fenton [10], photocatalytic degradation, photo-Fenton oxidation [11], and biodegradation [12]. Among these previous methods, the photo-Fenton type is favorable [13]. Many types of research

compared between Fenton and photo-Fenton efficiency to treat colors from wastewater; however, the results indicate that the photo-Fenton efficiency is better than the Fenton method because the Fenton process in the dark gave less efficient results [14, 15]. Better results were obtained from the UV-Fenton process, correlated with the Fenton process for removing Reactive Red 2 (RR 2) dye from wastewater. The RR2 achieved 88.10% decolorization for the photo-Fenton process when ferrous sulfate was used as the main salt in acidic conditions. For the Fenton process, the maximum decolorization efficiency was 82.18% [16]. In treating the Reaktoset Brilliant Orange P-2R (AO 33) and Telon Turquoise M-GGL (AB 279) dyes in an aqueous solution, the maximum removal efficiency of AO 33 was 95.81% by the Fenton process; for the Photo-Fenton process, the dye removal efficiency was 98.32%, while the maximum removal efficiency of AB 279 was 91.86% by the Fenton process; for the photo-Fenton process, the AB 279 dye removal efficiency was 94.28% [17]. In the photo-Fenton reaction, ferric or ferrous ions are used as catalysts together with hydrogen peroxide to increase the efficiency of the OH• radicals. This reaction works at low pH values because the iron ions strongly tend to precipitate as ferric hydroxide [18]. Dye degradation becomes a more efficient initiator through oxidation at higher Fe²⁺ and Fe³⁺ ions concentrations for a high reaction output [19]. Whereas pH values range between 3 and 4, usually involved in the photo-Fenton reaction [20], it is possible to generate the radicals of OH• at the photo reaction of H₂O₂ with Fe³⁺ too (photo-Fenton-like). Therefore, it decolorizes dyes with various molecular structures [21]. In a photo-Fenton-

like reaction, ferric ions react with H_2O_2 to produce ferrous ions at a very slow rate. A few ferrous ions can result and thus induce the photo-Fenton reaction [22]. The presence of UV light uplifts the oxidation of Fe^{+2} to Fe^{+3} , producing OH^\bullet radicals and increasing the solution acidity [23]. The chemical reactions of photo-Fenton and photo-Fenton-like are somewhat complex [24]. The photo-Fenton process includes the reaction in acidic conditions with H_2O_2 and Fe^{+2} in the presence of light (photo-Fenton) [20]. OH^\bullet radicals are produced during the reaction, and they are responsible for the oxidation process; therefore, they can decompose organic dyes. Fe^{+3} reacts with H_2O_2 , resulting in Fe^{+2} ; the reaction mechanism has catalytic properties. To enhance H_2O_2 and Fe^{+2} reactions, it is better to use UV because a high removal efficiency could be obtained because the oxidation reaction is due to the presence of ferrous/ferric ions coupled with coagulation, so ferrous/ferric ions play a double role as catalysts and coagulants [25]. The following equations explain the mechanisms [15, 21]:



Many previous researches used photo-Fenton and photo-Fenton-like processes to remove dyes from aqueous solutions. Dehghani et al. [20] studied the removal of Direct Red 81 (DR81) dye from an aqueous solution using the photo-Fenton process. Different parameters were investigated: pH, Fe^{+2} ions concentration, H_2O_2 concentration, initial dye concentration, and reaction time. The maximum removal efficiency was 98.1% at the optimum parameters: pH 3, Fe^{+2} concentration of 10 mg/l, H_2O_2 concentration of 50 mg/l, initial DR81 dye concentration of 100 mg/l, reaction time of 120 min. The increase in parameter concentration decreases the optimum removal efficiency. The results exhibited the photo-Fenton process effectiveness in removing DR81 dye from an aqueous solution. Simion et al. [18] investigated using a photo-Fenton-like oxidation process in removing Rhodamine 6G dye from textile wastewater using Fe^{+3} ions as a catalyst. H_2O_2 acts as an oxidizer for a large variety of organic pollutants. Therefore, the parameters of H_2O_2 concentration, Fe^{+3} concentration in addition to pH, and Rhodamine 6G dye concentration were considered in their study. It was concluded that the maximum Rhodamine 6G dye removal was 98% at pH 3.5, Fe^{+3} concentration 16 mg/L, H_2O_2 concentration 100 mg/L, and Rhodamine 6G dye concentration 27 mg/L. These results

revealed the effectiveness of the photo-Fenton process in removing Rhodamine 6G dye from textile wastewater. Ayed et al. [26] studied using the photo-Fenton process to remove Crystal Violet (CV) dye from an aqueous solution using local iron oxide as a natural catalyst. The considered parameters were the effect of catalyst dose, initial CV concentration, initial concentration of H_2O_2 , and pH. The results showed that the optimum conditions were 1 g/L catalyst dose, 5 mg/L initial CV concentration, pH 4, and an initial H_2O_2 concentration of 30 mg/L within 3 hrs. It was concluded that the photo-Fenton process was the most effective for CV dye removal, i.e., 98%. This paper investigates the effectiveness of two processes decolorizing Direct Orange 118 (DO 118) dye from wastewater: photo-Fenton and photo-Fenton-like. The optimum reaction time conditions, pH, initial DO118 concentration, concentration of H_2O_2 , temperature, and concentration of Fe^{+2} (photo-Fenton process) or Fe^{+3} (photo-Fenton-like process) are investigated. Additionally, kinetic models (pseudo-first-order and pseudo-second-order) for each reaction are studied. The percentage removal for the DO 118 dye of the two reactions is compared with other studies, and a comparison between photo-Fenton and photo-Fenton-like processes is performed.

2.METHODOLOGY

2.1.Chemicals

Chemicals used in this study were sulfuric acid (H_2SO_4) with a purity of 98%, sodium hydroxide (NaOH) with a purity of 98% provided by Merck Company (Germany), and hydrogen peroxide (Scharlau 30% w/w, molecular weight 34.01 g/mol) purchased from Chem-Supply (Australia). Ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) as a source of Fe^{+2} and ferric chloride (FeCl_3) as a source of Fe^{+3} were obtained from Merck Company (Germany). All chemicals were used directly without any further purification. Distilled water was used to dissolve all chemical samples in the requisite quantity and clean all the experimental system parts.

2.2.Dye Preparation

Direct Orange 118 (DO 118) (C.I. Orange E-2R) with molecular formula $\text{C}_{35}\text{H}_{24}\text{N}_6\text{Na}_4\text{O}_{15}\text{S}_4$ (molecular weight 988.82 g/mol) was supplied from the 14 Ramadan Factory of Textile Industries/Ministry of Industry in Baghdad, Iraq. The chemical structure of DO 118 dye is shown in Fig. 1. To prepare 1000 mg/l of stock solutions, 1 g of DO 118 dye was dissolved in one liter of distilled water and then diluted to the desired solution concentrations (10, 20, 30, 50, 80, and 100) mg/l. The maximum wavelength of DO 118 dye (420 nm) was calculated using a UV-VI spectrophotometer (model GENESYS™ 10 Series spectrophotometers; Thermo, USA).

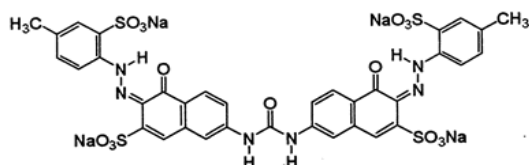


Fig. 1 Chemical Structure of DO 118 Dye.

2.3. Experimental Work

A batch mode laboratory-scale reactor was used in reaction tests consisting of 2 L volume of a Pyrex glass cylinder mounted at a magnetic stirrer with a hot plate (LMS-1003, Daihan Lab Tech, Korea), as illustrated in Fig. 2. A UV lamp (type: TUV 11W 4P-SE, Philips, England) with radiation (245 nm) was completely submerged in the solution and positioned vertically at the top of the reactor. The UV lamp (2.5 cm diameter and 22 cm length) was encased in a quartz sleeve for protection. The lamp was fixed 5 cm from the reactor's wall to ensure maximum radiation. The reactor was coated with a thin layer of aluminum foil to prevent UV radiation emission. A dye solution with different concentrations was prepared, and the pH of the solution was adjusted by adding H_2SO_4 and/or NaOH to get the desired pH. After that, a reagent ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ or FeCl_3) was added to the solution, and H_2O_2 was added. A magnetic stirrer was used for mixing the solution and operated for 30 min. at 200 rpm to get a homogeneous solution. To initiate the reaction, the UV lamp was switched on, and then the solution was stirred using a magnetic stirrer at 200 rpm for different time intervals. The heater was adjusted to the required temperature. Samples were withdrawn at selected intervals for dye concentration analysis

using a UV-VI spectrophotometer. Different factors were studied: irradiation time (10, 15, 30, 45, 60, 75, 90, 105, 120, and 135 min), pH (2, 3, 5, 8, and 10), dye concentrations (10, 20, 30, 50, 80, and 100 mg/l), H_2O_2 concentrations (100, 200, 400, 600, and 800 mg/l), $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ or FeCl_3 concentrations (10, 30, 50, 80, and 100 mg/l), and temperature (10, 20, 30, and 40 °C). The removal percentage (R%) was determined using Eq. (8) [27, 28]:

$$R(\%) = \left(\frac{C_o - C}{C_o} \right) \times 100 \quad (8)$$

where C_o is the dye concentration before treatment (mg/l), and C is the dye concentration at a specified time after treatment (mg/l).

3. RESULTS AND DISCUSSION

3.1. Impact of irradiation time on dye removal rate

The effect of irradiation time was studied to have the best removal efficiency of DO118 dye. Figures 3 and 4 show the effect of irradiation time on the removal efficiency of dye using pH 3, initial H_2O_2 concentration 200 mg/L, initial concentration of iron ions (Fe^{+2} or Fe^{+3}) 50 mg/L, initial DO 118 dye concentration 20 mg/L, and temperature 20 °C. The removal efficiency increased rapidly in the first minutes due to the presence of the oxidizing substance in abundance; hence, the highest removal efficiency of 64.347% and 54.325% using Fe^{+2} and Fe^{+3} , respectively, at 105 min, was obtained. With time, the removal efficiency slowly increased until it became almost constant after 120 min due to consuming most of the oxidant substance. These results conform with many studies [5, 29, 30].

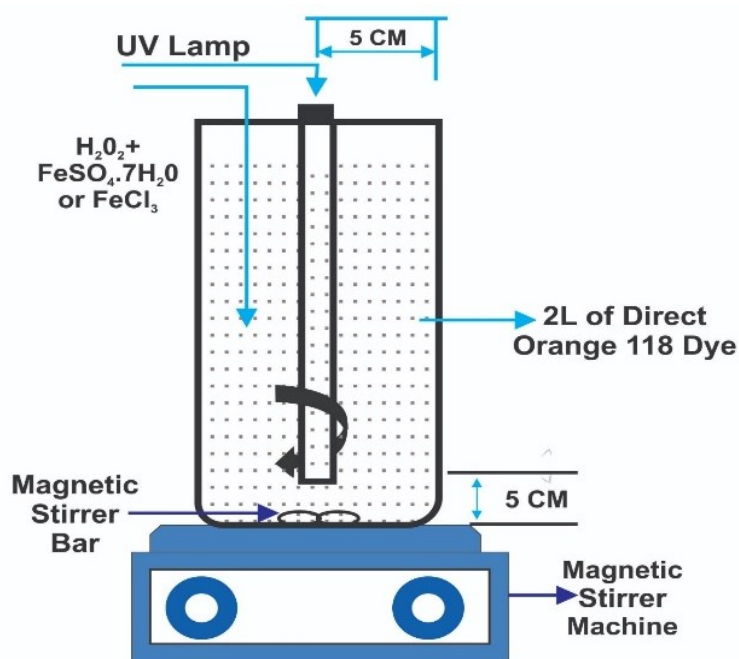


Fig. 2 Schematic Diagram of Batch Reactor.

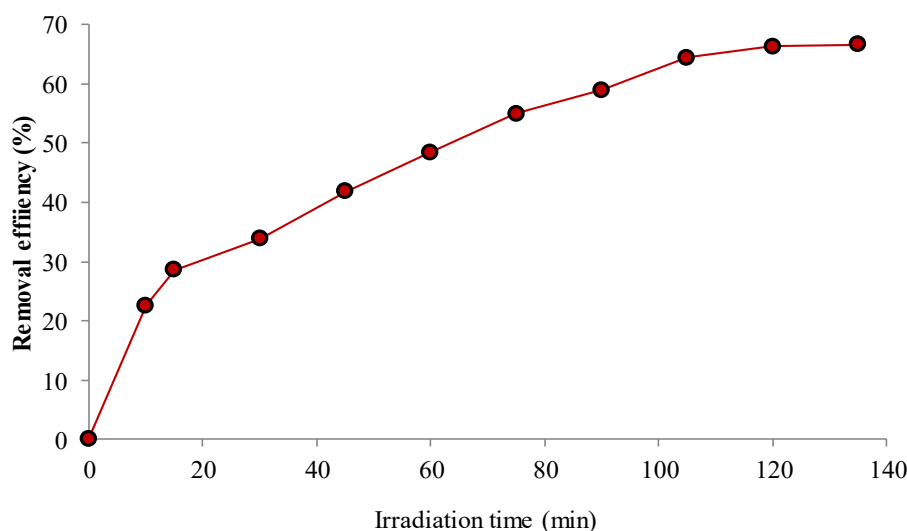


Fig. 3 Removal Efficiency of DO 118 Dye by the Photo-Fenton Process at Various Irradiation Times.

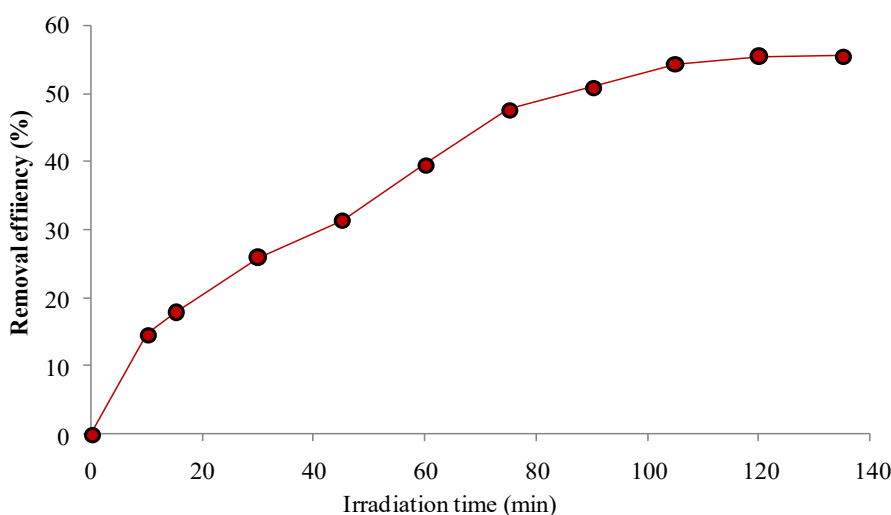


Fig. 4 Removal Efficiency of DO 118 Dye by the Photo-Fenton-Like Process at Various Irradiation Times.

3.2. Impact of pH on the Dye Removal Rate

pH value significantly affects the photo-Fenton and photo-Fenton-like treatment processes. Several tests were conducted under an initial dye concentration of 20 mg/l, initial iron concentrations (Fe^{+2} or Fe^{+3}) of 50 mg/l, initial H_2O_2 concentrations of 200 mg/l, temperature of 20 °C, and a variable pH value. Figures 5 and 6 illustrate that the higher dye removal efficiency for photo-Fenton and photo-Fenton-like reactions was 63.572% and 55.344%, respectively, within an irradiation time of 105 min at pH 3. Photo-Fenton and photo-Fenton-like reactions have maximum catalytic activity at pH 3 and would generate the maximum amount of hydroxyl radical (OH^\bullet); hence, color removal was observed. Several studies have indicated that the optimum pH for maximum dye removal occurs at an acidic pH of about 3

for photo-Fenton and photo-Fenton-like reactions [21, 25, 31]. At lower pH values, color removal decreased because hydrogen ion (H^+) acts as an OH^\bullet radical-scavenger and also because $[\text{Fe}(\text{H}_2\text{O})_6]^{+2}$ complex formation occurred rather than the $[\text{Fe}(\text{OH})(\text{H}_2\text{O})_5]^+$ complex, which slowly reacts with hydrogen peroxide (H_2O_2) and forms less amount of OH^\bullet radical [32]. On the other hand, when $\text{pH} > 3$, the color removal was strongly decreased to reach 31.678% and 26.775% at pH 10 for photo-Fenton and photo-Fenton-like reactions, respectively, because Fe^{+2} or Fe^{+3} react with OH^\bullet to precipitate the iron hydroxide ($\text{Fe}(\text{OH})_2$ or $\text{Fe}(\text{OH})_3$) which is unresponsive with H_2O_2 and prevents the transmission of radiation [20]. In addition, the reason for the incompetent removal of dye at $\text{pH} > 3$ was the dissociation and auto-decomposition of H_2O_2 [33].

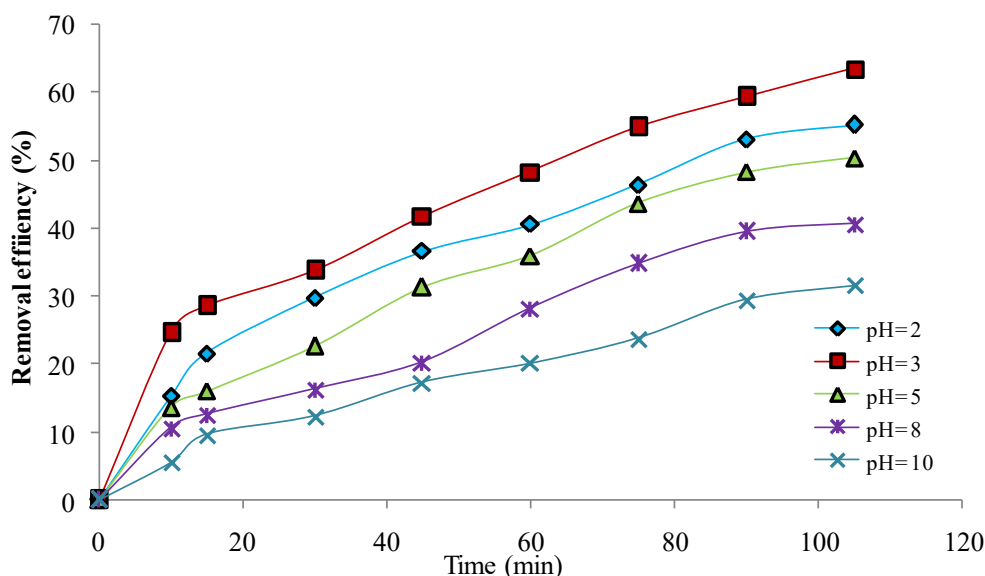


Fig. 5 Removal Efficiency of DO 118 Dye by the Photo-Fenton Process at Various pH.

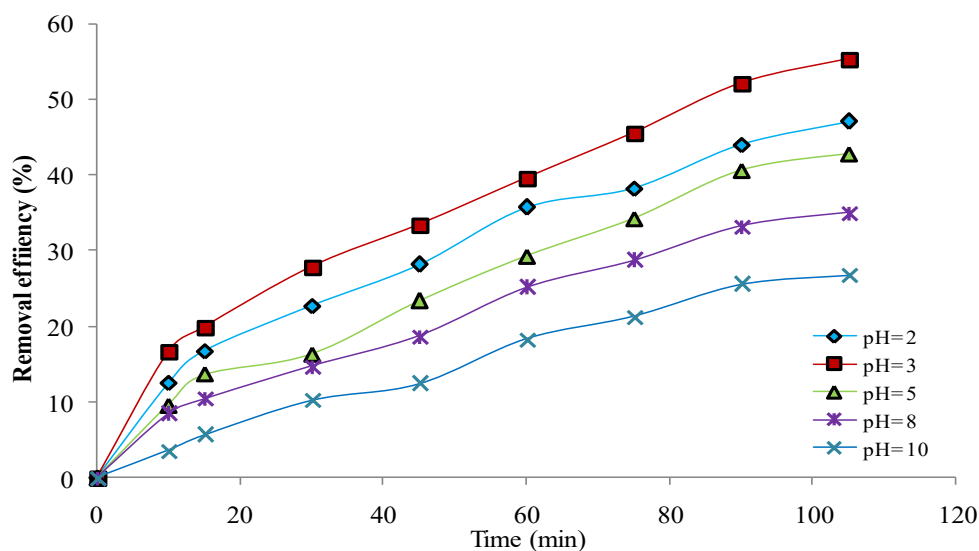


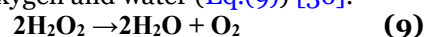
Fig. 6 Removal efficiency of DO 118 dye by the photo-Fenton-like process at various pH.

3.3. Impact of Initial H_2O_2

Concentration on Dye Removal Rate

To study the effect of initial H_2O_2 concentrations on the removal of DO 118 dye by photo-Fenton and photo-Fenton-like processes, various initial H_2O_2 concentrations were tested at Fe^{+2} and Fe^{+3} concentration of 50 mg/L for the photo-Fenton and photo-Fenton-like reactions, initial DO 118 dye concentration 20 mg/L, pH 3, and temperature 20 °C within irradiation time of 105 min. Figure 7 shows that the removal efficiency increased from 49.945% to 81.655% by increasing H_2O_2 concentrations from 100 to 400 mg/L for the photo-Fenton reaction. Figure 8 reveals that the removal efficiency increased from 41.145% to 64.422% by increasing H_2O_2 concentrations from 100 to 600 mg/L for a photo-Fenton-like reaction. Thus, providing the right amount of H_2O_2 may encourage producing OH^\bullet radical and increase

the ability to remove the DO 118 dye; these findings agree with the results of Ebrahiem et al. [34]. When the concentration of H_2O_2 was low, it generated insufficient OH^\bullet radical; therefore, the removal efficiency of DO 118 dye decreased [35]. Further increases in H_2O_2 concentrations more than the optimum concentration induced a drop in the removal rate of dye due that when H_2O_2 concentrations increased, excess H_2O_2 reacted with OH^\bullet radicals to produce hydroperoxyl radicals (HO_2^\bullet), which are significantly less reactive species than OH^\bullet radicals. This reaction reduced dye removal efficiency (auto-scavenging of OH^\bullet radicals by H_2O_2) (Eq.(7)) [16] as well as induced auto-decomposition of H_2O_2 to oxygen and water (Eq.(9)) [36].



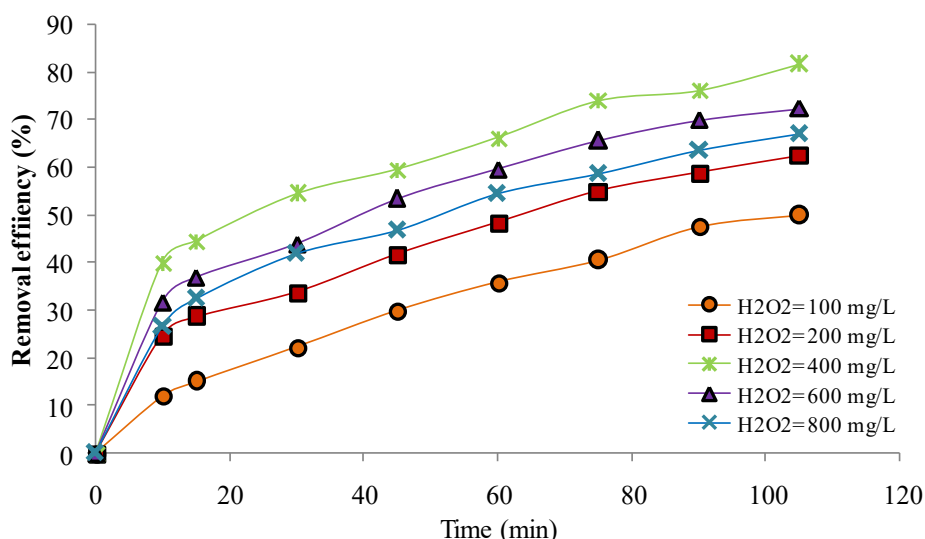


Fig. 7 Removal Efficiency of DO 118 Dye by the Photo-Fenton Process at Various Initial H₂O₂ Concentrations.

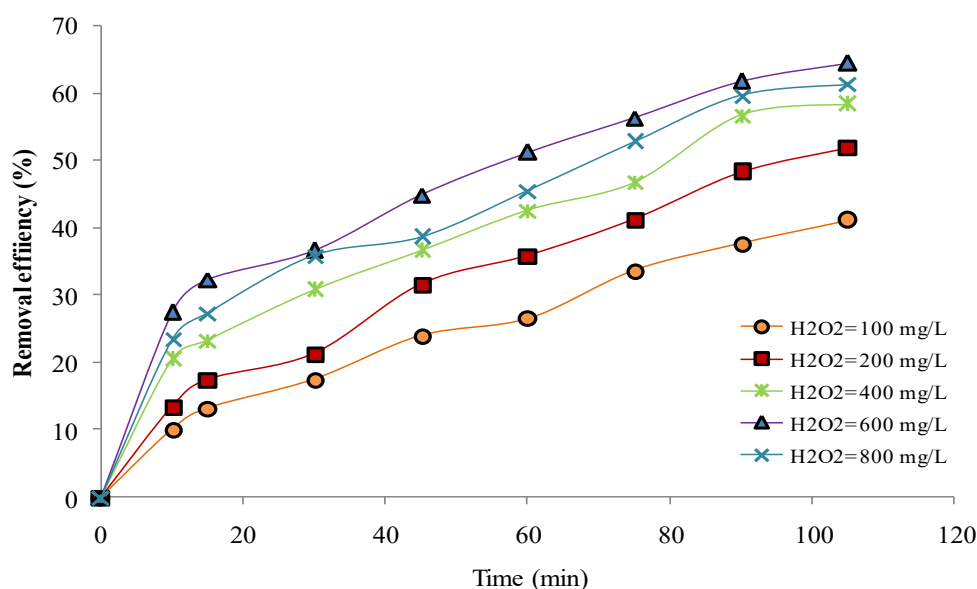


Fig. 8 Removal Efficiency of DO 118 Dye by the Photo-Fenton-Like Process at Various Initial H₂O₂ Concentrations.

3.4. Impact of Initial Fe Ions Concentration on the Dye Removal Rate

To explain the role of the initial concentration of Fe²⁺ and Fe³⁺ on the degradation of DO 118 dye, a series of experiments were conducted with different initial Fe²⁺ and Fe³⁺ concentrations. These experiments were conducted at an initial dye concentration of 20 mg/l, pH 3, a temperature of 20 °C, and an irradiation time of 105 min for the photo-Fenton and photo-Fenton-like processes. The H₂O₂ concentration from previous experiments for the photo-Fenton process was 400 mg/l, and for the photo-Fenton-like process was 600 mg/l. Figure 9 reveals that the removal DO 118 dye efficiency reached 81.731% at a Fe²⁺ concentration of 50 mg/l; however, it dropped

to 69.108% at 100 mg/l for the photo-Fenton process within 105 min. However, Fig. 10 shows that the maximum result for the photo-Fenton-like process was 73.173% at Fe³⁺ concentrations of 80 mg/l; however, it dropped to 64.122% at 100 mg/l within 105 min. The optimum concentration of Fe²⁺ was 50 mg/l for the photo-Fenton process, and the optimum concentration of Fe³⁺ was 80 mg/l for the photo-Fenton-like process because increasing Fe²⁺ and Fe³⁺ to these values increased producing OH• radicals for the oxidation process [21]. At low Fe²⁺ or Fe³⁺ concentrations, the removal efficiency decreased due to the lowest production of OH• radicals [37]. Any additional amount of Fe²⁺ or Fe³⁺ above the optimum value causes brown turbidity and prohibits the adsorption of the light required

for the process [34]. In this case, excess Fe^{+2} reacted with the OH^\bullet radical as a scavenger by converting it to (OH^-) ions during the oxidation

of Fe^{+2} , according to Eq. (2) [24]. The results are consistent with Tariq and Khan [38].

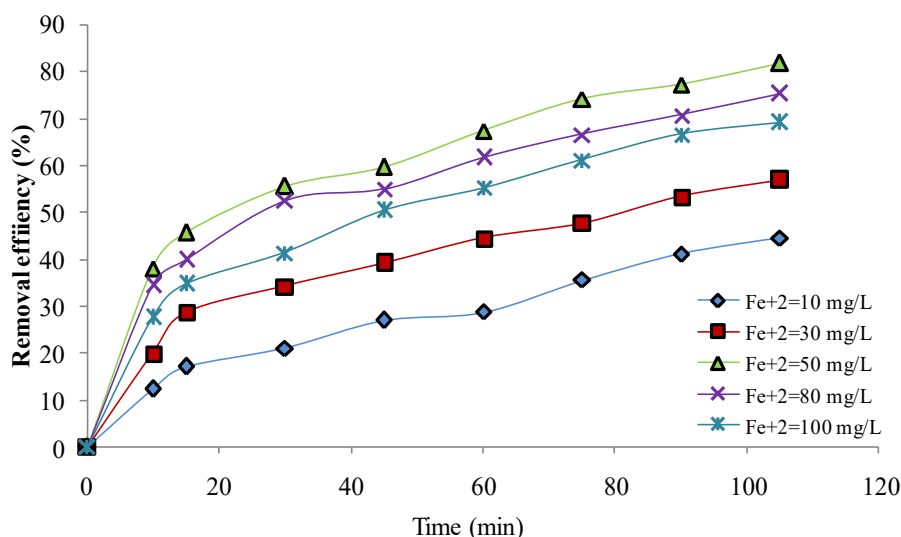


Fig. 9 Removal Efficiency of DO 118 Dye by the Photo-Fenton Process at Various Initial Fe^{+2} Concentrations.

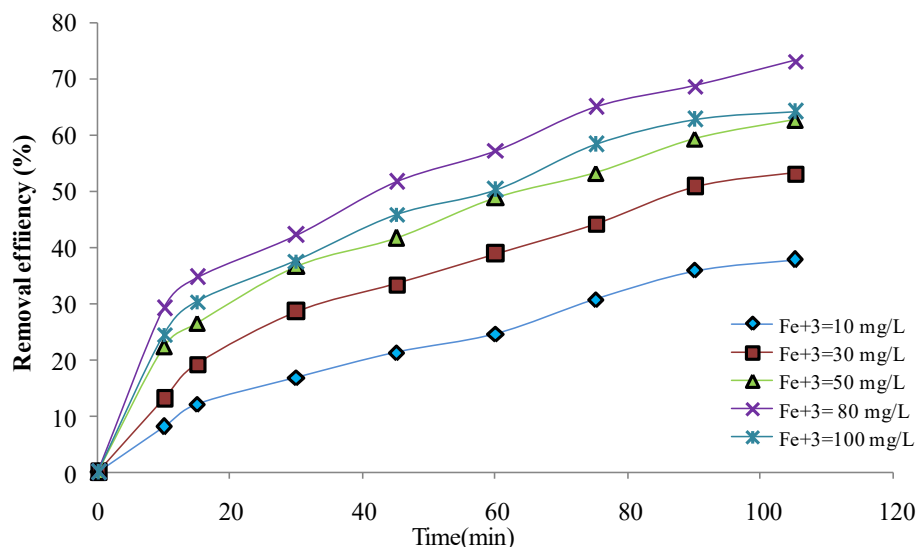


Fig. 10 Removal Efficiency of DO 118 Dye by the Photo-Fenton-Like Process at Various Initial Fe^{+3} Concentrations.

3.5. Impact of Solution Temperature on the Dye Removal Rate

The removal efficiency of DO 118 dye by photo-Fenton and photo-Fenton-like processes was studied at variable temperatures versus constant parameters as follows: Fe^{+2} concentration=50 mg/l, H_2O_2 concentration=400 mg/l, irradiation time= 105 min, DO 118 dye concentration= 20 mg/l, and pH 3 for the photo-Fenton process; Fe^{+3} concentration= 80 mg/l, H_2O_2 concentration= 600 mg/l, irradiation time= 105 min, DO 118 dye concentration= 20 mg/l, and pH 3 using a photo-Fenton-like process. Figures 11 and 12 show that the removal efficiency of DO 118 dye increased as temperature increased; therefore,

temperature positively affected the dye removal efficiency, in agreement with [19, 39]. The results indicated that after the temperature was increased from 10 to 30 °C, the removal efficiency increased from 76.101% to 88.126% with adding Fe^{+2} . On the other hand, for the same increasing range of temperature, the percentage of removal increased from 69.181% to 83.922% with adding Fe^{+3} . Therefore, the increase in temperature increased the reaction between H_2O_2 and $\text{Fe}^{+2}/\text{Fe}^{+3}$ ions, which improved generating OH^\bullet radicals, which enhanced the DO 118 dye removal [1]. However, the decline in removal efficiency was beyond 30 °C because H_2O_2 disintegrated into water molecules and O_2 [37, 40].

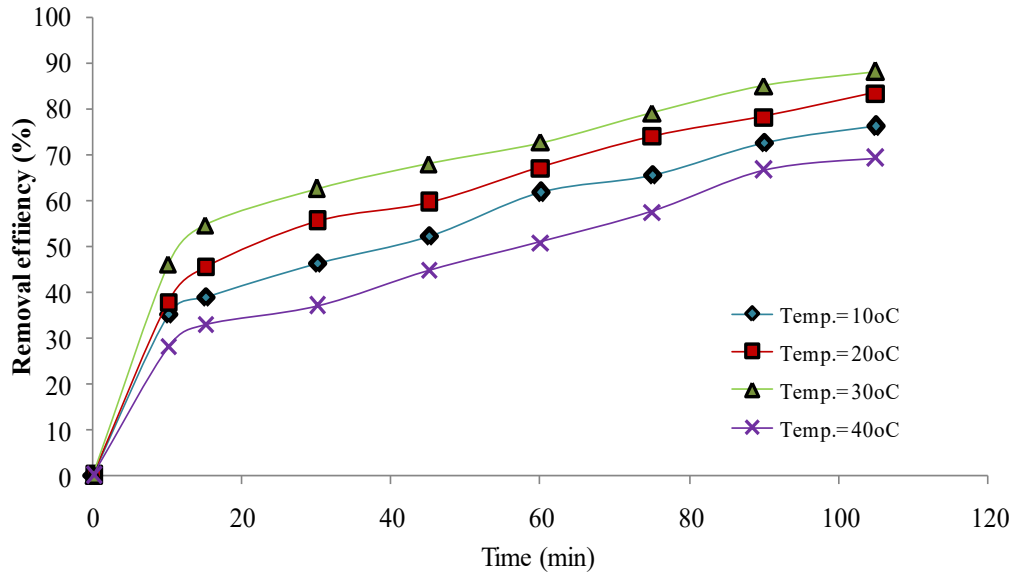


Fig. 11 Removal Efficiency of DO 118 Dye by the Photo-Fenton Process at Various Solution Temperatures.

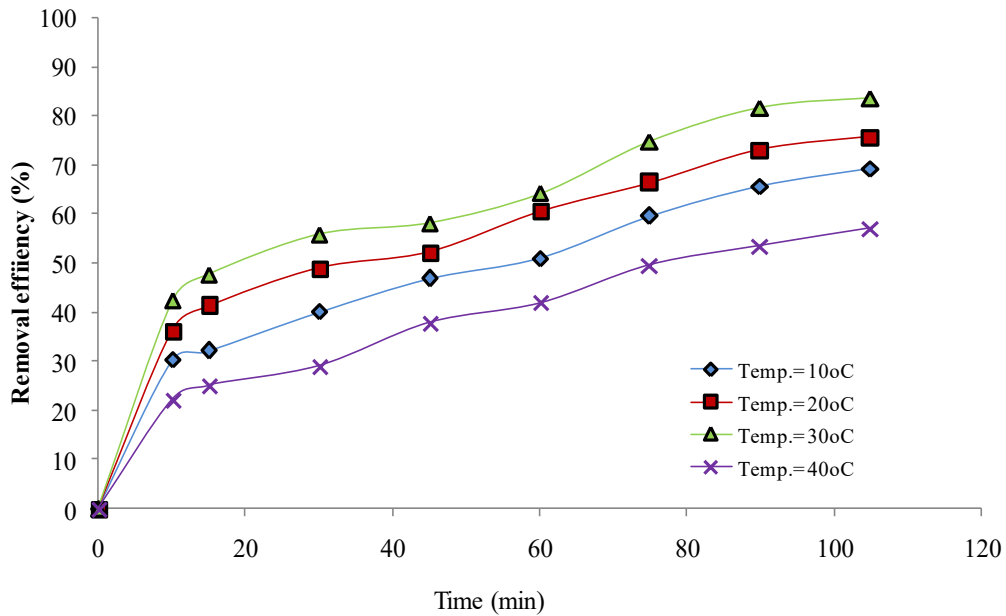


Fig.12 Removal Efficiency of DO 118 Dye by the Photo-Fenton-Like Process at Various Solution Temperatures.

3.6.Impact of Initial Dye Concentration on the Removal Rate

The DO 118 dye concentration should be included in the present study because it is essential in photo-Fenton and photo-Fenton-like processes. So, different initial concentrations of DO 118 dye were measured to test the optimum dye concentration required for the best removal efficiency. At the same time, other parameters from previous experiments were fixed at the optimum value. Figures 13 and 14 show that the maximum DO 118 dye removal efficiency was 97.817% and 91.244% at 10 mg/l of DO 118 dye concentration within 105 min for photo-Fenton and photo-Fenton-like processes, respectively, because at

low dye concentration, there are more active sites provided on the catalyst surface than the number of dye molecules, so the dye degradation increased [41]. However, the results become low when the dye concentration exceeds 10 mg/l, agreeing with [42, 24]. It can be noted that when the initial dye concentration increased, the generation of OH^\bullet radicals on the molecular surface of the catalyst decreased due to a few active sites available [43]. Moreover, an increase in dye concentration led to most UV light being absorbed by the surface of dye molecules instead of H_2O_2 . Thus, OH^\bullet radicals generation decreased, reducing the removal dye rate [44].

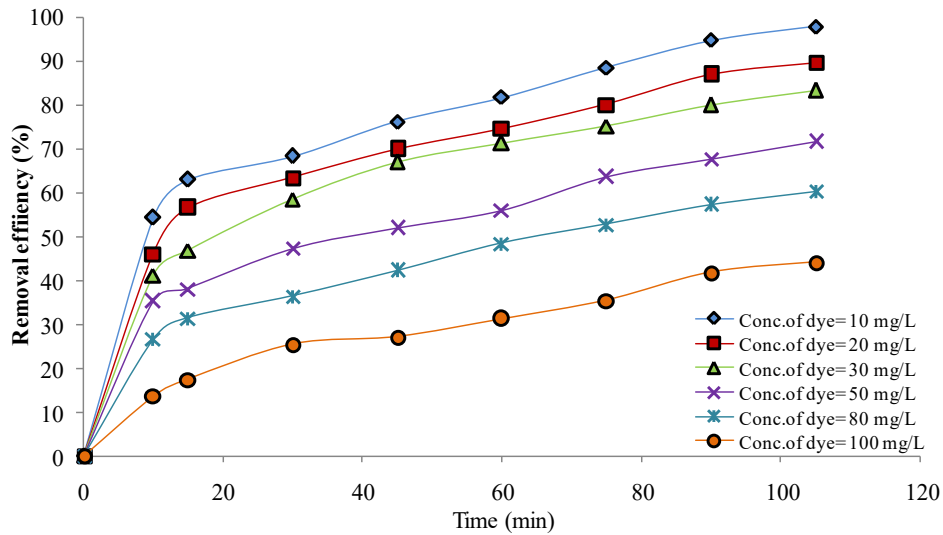


Fig. 13 Removal Efficiency of DO 118 Dye by the Photo-Fenton Process at Various Initial DO 118 Dye Concentrations.

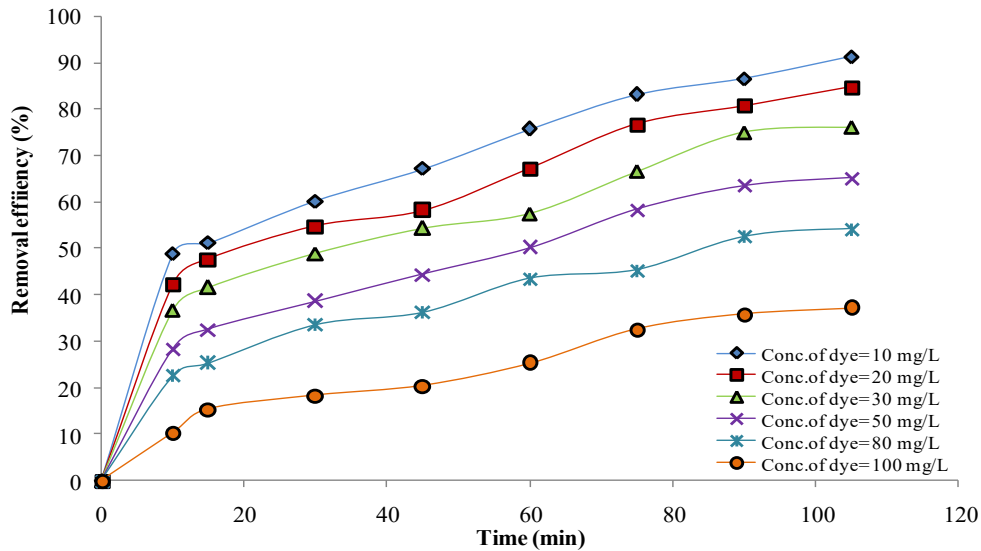


Fig. 14 Removal Efficiency of DO 118 Dye by the Photo-Fenton-Like Process at Various Initial DO 118 Dye Concentrations.

3.7. Models of Kinetics

The experimental data of DO 113 dye degradation can be fitted according to the kinetic models of pseudo-first-order and pseudo-second-order at different DO 113 dye concentrations of 10, 20, 30, 50, 80, and 100 mg/l and optimum parameters (the concentration of $\text{H}_2\text{O}_2 = 400$ mg/l, concentration of $\text{Fe}^{+2} = 50$ mg/l, $\text{pH} = 3$, temperature = 30°C , and irradiation time = 105 min for the photo-Fenton process, and $\text{pH} = 3$, Fe^{+3} concentration = 80 mg/l, H_2O_2 concentration = 600 mg/l, temperature = 30°C , and irradiation time = 105 min for the photo-Fenton-like process). The kinetic models were determined using the following equations:

For a pseudo-first-order kinetic model [44]:

$$\ln\left(\frac{C_0}{C}\right) = K_1 t \quad (10)$$

For a pseudo-second-order kinetic model [17]:

$$\frac{1}{C} - \frac{1}{C_0} = K_2 t \quad (11)$$

where C_0 is the initial dye concentration (mg/l), C is the concentration of dye at the time of reaction (mg/l), t is the reaction time (min), K_1 is the pseudo-first-order rate constant (1/min), and K_2 is the pseudo-second-order rate constant (1/mg/min). For each experiment, a plot of $\ln\left(\frac{C_0}{C}\right)$ and $\frac{1}{C} - \frac{1}{C_0}$ versus time yields a straight line whose slopes are K_1 and K_2 , respectively. Tables 1 and 2 indicate that the pseudo-first-order kinetic model fits the experimental data better than the pseudo-second-order kinetic model for photo-Fenton and photo-Fenton-like reactions. For the photo-Fenton reaction, the pseudo-first-order rate constant (K_1) decreased from (0.034005 to 0.005546) 1/min at the regression coefficient (R^2) range of (0.990 to 0.98388). For the

photo-Fenton-like reaction, the pseudo-first-order rate constant (K_1) decreased from (0.023195 to 0.004416) l/min at the regression coefficient (R^2) range of (0.9847 to 0.9686) as DO 118 dye concentration increased from 10 to 100 mg/L, which is compatible with the results of [45, 46]. The results showed that K_1 values decreased when the initial dye concentration increased and demonstrated high R^2 values of the DO 118 dye degradation at different DO 118 dye concentrations, suggesting that the DO118

dye degradation efficiency decreased with increasing initial dye concentration, which is consistent with the experimental findings detailed in Section (3.6). Table 3 compares different dye removal efficiencies under optimum conditions between the present study and previous studies using photo-Fenton and photo-Fenton-like techniques. The present study results were excellent compared to other studies.

Table 1 Pseudo-First-Order and Pseudo-Second-Order Kinetic Models' Parameters for a Photo-Fenton Reaction at Different Initial DO 118 dye Concentrations.

Concentration of DO 118 dye (mg/l)	Removal efficiency (R%) at 105 min	Pseudo-first-order		Pseudo-second-order	
		K_1 , l/min	R^2	K_2 , l/mg/min	R^2
10	97.817	0.034005	0.990	0.032892	0.8263
20	89.635	0.021588	0.984	0.004118	0.9035
30	83.235	0.017008	0.9916	0.001576	0.9809
50	71.583	0.011983	0.9944	0.00048	0.9778
80	60.352	0.008811	0.9969	0.000181	0.9950
100	44.138	0.005546	0.98388	7.53E-05	0.9816

Table 2 Pseudo-First-Order and Pseudo-Second-Order Kinetic Models' Parameters for a Photo-Fenton-Like Reaction at Different Initial DO 118 Dye Concentrations.

Concentration of DO 118 dye (mg/l)	Removal efficiency (R%) at 105 min	Pseudo-first-order		Pseudo-second-order	
		K_1 , l/min	R^2	K_2 , l/mg/min	R^2
10	91.244	0.023195	0.9847	0.009925	0.8784
20	84.682	0.017868	0.9795	0.002633	0.9275
30	76.018	0.013599	0.9722	0.001006	0.9317
50	65.180	0.010047	0.9896	0.000357	0.9773
80	54.145	0.007426	0.9856	0.000141	0.9831
100	37.102	0.004416	0.9686	5.62E-05	0.9648

Table 3 Results of the Present Study Compared with Previous Studies Results.

Type of dye	Type of treatment methods	Catalyst dosage	Optimum reaction time	Optimum conditions	Kinetic models	Removal efficiency	Ref.
Orange G azo dye	photo-Fenton-like-process	Ferric sulfate ($\text{Fe}_2\text{SO}_4)_3$ as a source of Fe^{+3}	180 min	[pH=2.9±0.1, Fe^{+2} =0.435 mM, H_2O_2 =6 mM, dye = 45.2 g /L, and Temp=40 °C]	Pseudo-first-order	3.41%	46
Disperse Red1 (DR 1) dye	photo-Fenton-like-process	Ferric nitrate nonahydrate ($\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$) as a source of Fe^{+3}	45 min	[pH= 2.5±0.1, Fe^{+3} =0.2 mM, H_2O_2 =5 mM, dye =23 mg/L, and Temp=25 °C]	-	98%	47
Acid Orange 24 (AO 24)	photo-Fenton process	Ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}_2$) as a sources of Fe^{+2}	65 min	[pH=5.5, Fe^{+2} = 1.43×10^{-4} M, H_2O_2 = 5.2×10^{-3} M, dye =200 mg/L, and Temp=25 °C]	Pseudo-first-order	95%	48
Acid Red 33 (AR 33)	photo-Fenton process	Ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}_2$) as a source of Fe^{+2}	10 min	[pH=3, Fe^{+2} =0.25 mM, H_2O_2 =20 mM, dye =250 mg/L, and Temp= 25 °C]	Pseudo-first-order	98. 5%	43
Acid Blue 161 (AB 161)	photo-Fenton process	Ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}_2$) as a source of Fe^{+2}	10 min	[pH=3.5, Fe^{+2} = 1.0×10^{-4} M, H_2O_2 = 1.1×10^{-3} M, dye =1.0 $\times 10^{-4}$ M, and Temp=18 to 22 °C]	Pseudo-second-order	40 %	14
Reactive Red dye (RR dye)	photo-Fenton process	Ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}_2$) as a source of Fe^{+2}	90 min	[pH=3, Fe^{+2} = 20 mg/L, H_2O_2 = 78 mg/L, dye= 50 mg/L, and Temp=60 °C]	-	48%	33
Crystal Violet (CV) dye	photo-Fenton process	Ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}_2$) as a source of Fe^{+2}	60 min	[pH=6.5, Fe^{+2} = 0.05×10^{-3} M, H_2O_2 = 2×10^{-3} M, dye = 0.01×10^{-3} M, and Temp= 25 °C]	Pseudo-first-order	58%	49
Reactive Red 2 (RR 2) dye	photo-Fenton process	Ferric sulphate hydrate ($\text{Fe}_2(\text{SO}_4)_3 \cdot \text{H}_2\text{O}$) as a sources of Fe^{+2}	45 min	[pH=3, Fe^{+2} =0.125 mM, H_2O_2 =1 mM, dye =0.08 mM, and Temp= 25 °C]	-	88.10%	16
Reactive Blue dye (R B)	photo-Fenton process	Ferrous sulfate (FeSO_4) as a source of Fe^{+2}	90 min	[pH=3, Fe^{+2} = 20 mg/L, H_2O_2 =78 mg/L, dye =20 mg/L, and Temp=40 °C]	-	84.82 %	50

Acid Yellow 17 (AY 17) dye	photo-Fenton process	Ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) as a source of Fe^{+2}	25 min	[pH=3, Fe^{+2} =0.09 mM, H_2O_2 =1mM, dye =0.14 mM, and Temp= 25 °C]	Pseudo-first-order	88%	38
Acid Yellow 17 (AY 17) dye	photo-Fenton-like process	Ferric sulfate ($\text{Fe}_2\text{SO}_4)_3$ as a source of Fe^{+3}	60min	[pH=3, Fe^{+3} =0.09 mM, H_2O_2 =10 mM, dye =0.14 mM, and Temp= 25 °C]	Pseudo-first-order	93%	38
Direct Orange 118 (DO 118) dye	photo-Fenton process	Ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) as a source of Fe^{+2}	105 min	[pH=3, Fe^{+2} = 50 mg/L, H_2O_2 = 400 mg/L, dye =10 mg/L, and Temp=30 °C]	Pseudo-first-order	97.817%	Present study
Direct Orange 118 (DO 118) dye	photo-Fenton-like process	Ferric chloride (FeCl_3) as a source of Fe^{+3}	105 min	[pH=3, Fe^{+3} = 80 mg/L, H_2O_2 =600 mg/L, dye =10 mg/L, and Temp=30 °C]	Pseudo-first-order	91.244%	Present study

3.7.Comparison of Photo-Fenton and Photo-Fenton-Like Processes

Figure 15 compares between photo-Fenton and photo-Fenton-like reactions for DO118 dye decolorization at optimum parameters: pH= 3, temperature= 30 °C, irradiation time= 105 min, and an initial DO118 dye concentration= 10 mg/l for photo-Fenton and photo-Fenton-like reactions. H_2O_2 concentration= 400 mg/l, Fe^{+2} concentration= 50 mg/l for the photo-Fenton process, and Fe^{+3} concentration= 80 mg/l, H_2O_2 concentration= 600 mg/L for the photo-Fenton-like process. From Fig. 16, it is clear that decolorization with UV/ Fe^{+2} / H_2O_2 and

UV/ Fe^{+3} / H_2O_2 reactions was effective at a various removal percentage. The photo-Fenton reaction had a higher removal percentage (97.817%) than the photo-Fenton-like reaction (91.244%) because, in the photo-Fenton reaction, Fe^{+2} ions react quickly with H_2O_2 to produce hydroxyl radicals, while in the photo-Fenton-like reaction, Fe^{+3} reacts with H_2O_2 to produce hydroperoxide radicals which have a lower oxidation capability than the hydroxyl radicals [21]. Many studies have shown that the decolorization percentage is faster with the photo-Fenton reaction [1, 51].

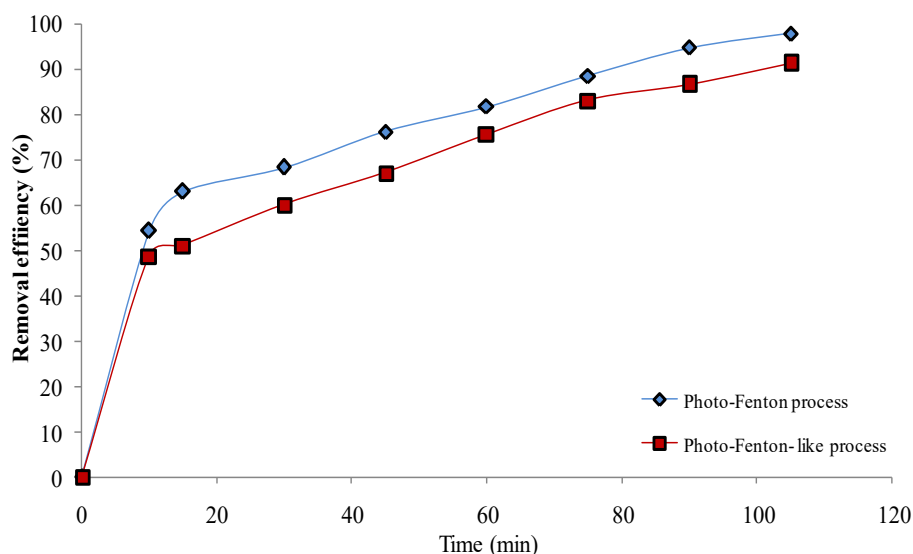


Fig. 15 Comparison of the Photo-Fenton and Photo-Fenton-Like Reactions for DO 118 Dye Decolorization.

4.CONCLUSIONS

The findings of this research have shown that photo-Fenton and photo-Fenton-like processes are potentially effective for decolorizing DO 118 dye from wastewater. The decolorization degree of DO 118 dye depends on different parameters, such as irradiation time, pH, Fe^{+2} or Fe^{+3} concentration, H_2O_2 concentration, temperature, and initial dye concentrations. At optimum parameters, the maximum removal percentages when using the photo-Fenton-like process and the photo-Fenton process are illustrated in Table 4. The decolorization efficiency of DO 118 dye was more effective

when increasing pH, H_2O_2 concentration, Fe^{+2} or Fe^{+3} concentrations, and temperature, while it was negatively affected by the increase in initial DO 118 dye concentration. At an acidic pH (3), all the applicable methods were effective. Furthermore, the presence of Fe^{+2} or Fe^{+3} as a catalyst made H_2O_2 act as a higher oxidant. Two kinetic models for the degradation of DO 118 dye were investigated. It was found that the pseudo-first-order kinetic model was more fitted than the pseudo-second-order kinetic model due to the higher regression coefficients. The photo-Fenton

process was more efficient than the photo-Fenton-like process.

Table 4 Comparison between Photo-Fenton and Photo-Fenton-Like Processes at Optimum Parameters.

Parameter	Photo-Fenton process	Photo-Fenton-like process
Irradiation time (min)	105	105
pH	3	3
H ₂ O ₂ concentration (mg/L)	400	600
Fe ions concentration (mg/L)	50 (Fe ⁺²)	80 (Fe ⁺³)
Solution temperature (°C)	30	30
Dye concentration (mg/L)	10	10
Kinetics models	Pseudo-first order	Pseudo-first order
Removal percentages (%)	97.817%	91.244%

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ABBREVIATION

AB 161	Acid Blue 161
AB 279	Telon Turquoise M-GGL
AO 24	Acid Orange 24
AO 33	Reaktoset Brilliant Orange P-2R
AR 33	Acid Red 33
AY 17	Acid Yellow 17
CV	Crystal Violet
DO 118	Direct Orange 118
DR 1	Disperse Red 1
DR 81	Direct red 81
FeCl ₃	Ferric chloride
FeSO ₄ ·7H ₂ O	Ferrous sulfate heptahydrate
H ₂ O ₂	Hydrogen peroxide
H ₂ SO ₄	Sulfuric acid
NaOH	Sodium hydroxide
OH•	Hydroxyl radical
RB	Reactive Blue
RR	Reactive Red
RR 2	Reactive Red 2

NOMENCLATURE

C ₀	Initial dye concentration, mg/l
C	Concentration of dye at the time of reaction, mg/l
K ₁	Pseudo-first-order rate constant, l/min
K ₂	Pseudo-second-order rate constant, l/min
R ₂	Regression coefficient
t	Reaction time, min

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