Investigation the effect of intensity and direction of light on the removal of reactive blue dye from simulated wastewater using photo-Fenton oxidation under UV irradiation: Batch and continuous methods

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Abstract: This study aims to remove reactive blue dye (R.B) from simulated wastewater by utilizing advanced oxidation processes (AOPs) using a photo-Fenton (UV/H$_2$O$_2$/Fe$^{2+}$) system. A photo-reactor containing four fixed UV lamps had been employed in a batch and continuous mode under the effect of several operating variables (dosage of H$_2$O$_2$; dosage FeSO$_4$; pH; temperature; irradiation time). Response surface methodology (Box–Behnken design) and Portable Statgraphics Centurion statistical software were used to design the experiments and conducting the mathematical correlation of the required responses as well as the interaction effects among variables. The optimal conditions of the operating variables; dosage hydrogen peroxide, ferrous sulphate, pH, temperature and irradiation time were 78 ppm, 20 ppm, 3, 40\degree C and 90 min, respectively which give 84.82\% of removal efficiency. Then, the effect of light intensity and distance from a UV source were studied at optimum conditions in the batch photo-reactor where the highest dye removal efficacy was obtained at a light intensity of 24 w and a distance of 15cm. For continuous system, two operating conditions were studied, the direction of the light radiation and the flow rate which proofed that the dye removal efficiency decreased with an increase of flow rate and the top direction compared to the side direction of the light where the removal percentage of (R.B) was 100\% for the flow rate of 10 ml/min while it was 84.16\% for a flow rate of 50 ml/min. The results show that the photo-Fenton method is an effective treatment method of Wastewater containing dyes.

Keywords: Wastewater treatment; Reactive blue dye; Advanced oxidation; Photo-Fenton process; RSM; Optimization.

1. Introduction

Industrial and domestic wastewaters are characterized as the main reason of the aquatic pollution which are discharged annually into the ecosystem without using effective treatment methods especially in many of the developing countries [1, 2]. The essential environmental crisis of these wastewaters is that they containing a huge amount of organic compounds, such
as textile dyes, aromatic compounds, chlorinated hydrocarbons and phenolic compounds [3-5]. Dye contaminants from the dyestuff industrial printing, coloring and textile industries are important sources of environmental contamination. The wastes discharged from these productions are frequently dyed extensively, and the direct announcement of the wastewater into getting water body will reason injury to together human and aquatic life existences because of their high solubility, artificial dyes are common water pollutants that are likewise toxic and oncogenic [6,7]. Typical methods used to overcome this problem are biological, physical, and chemical [2,8,9]. Regrettably, most of traditional wastewater treatment systems are inefficient in treating such emerging and priority contaminants as this needs the pollutants to be mineralized [10]. Advanced oxidation processes (AOPs) had established to provide better alternatives to protect public health and the environment as a result of total water pollutant degradation and mineralization [11]. The AOPs are focused on the generation of hydroxyl radical (OH•) which leads the organic pollutants converting to carbon dioxide, water and inorganic materials [12]. A variety of AOPs involve chlorination, ozonation, Fenton, photo-Fenton, photocatalytic and wet-air oxidation [13]. Photo-Fenton technique is the blending of Fenton reagents (hydrogen peroxide and ferrous sulphate) and UV–via energy that extends to free radical [14]. Hydrogen peroxide (H₂O₂) is extensively secondhand by a way of an initiator in AOPs, anywhere it can upsurge the rate of free-radical creation because of the reaction between ozone and its conjugate base. Consequently, the usefulness of advanced oxidation processes involving hydrogen peroxide has lately augmented aimed at the active oxidation of dangerous contaminants in wastewater [15]. Numerous researchers studied about advanced oxidation processes to remove organic pollutants, Ghaly et. al. [16]. In lab-scale studies on p-chlorophenol degradation, advanced oxidation processes (AOPs) utilizing combinations of UV / H₂O₂ and photo-Fenton reaction were studied. The study showed that the photo-Fenton procedure was the most effective therapy under acidic conditions and provided a higher rate of degradation of p-chlorophenol at a very short radiation time. Hsueh et.al. [17] studied Fenton and Fenton-like reactions to oxidize three common azo dyes at low iron concentrations, namely Red MX-5B, Reactive Black 5 and Orange G. The study reveals that both of these processes will completely remove the color of these dyes. Sun et. al.[18] studied the azo degradation Amido black in aqueous solution by the Fenton oxidation method under the influence of different reaction parameters such as initial pH, initial hydrogen peroxide concentration, initial ferrous concentration. They found that after 60 min of reaction, 99.25 % removal of the pollutant from the aqueous solution was obtained under the optimal conditions. Ebrahiem et. al.[19] investigated the feasibility of using advanced photo-oxidation methods to eliminate the residual organic compounds present in cosmetic wastewater. The optimum conditions were found to be: 40min of irradiation time, pH 3, the dose of 1 ml/l H₂O₂ and 0.75g/l for Fe(II) and Fe(III). In such circumstances, the removal of 95.5 % of chemical oxygen demands. Aljubour et. al. [20] studied the quality of utilizing Fenton's reagent in the solar photocatalysis of TiO₂ (H₂O₂/Fe²⁺/TiO₂/sunlight) for the degradation of TOC and COD from petroleum wastewater. The results showed that the removal rates of TOC and COD were 62% and 50% respectively. The photo-Fenton solar
technique is very efficient for treating oil wastewater with acidic pH, i.e. less than 7, and more expense-effective free energy. The aim of the present work is to assess the treatability of dye removal from simulated wastewater containing reactive blue utilizing photo-Fenton reactor, batch and continuous modes. For the batch mode, the effect of the operating variables; irradiation time, pH, hydrogen peroxide concentration, ferrous sulphate concentration and effect of temperature; had been studied. Moreover, the influence of light intensity, direction and distance on the decolorization efficiency and flow rate in continuous mode was investigated.

2. Experimental Work

2.1. Materials

The reactive blue dye with 585 nm wavelength that shown its chemical structure in Fig. 1 was supplied by Al-Hilla textile factory located in the south of Baghdad-Iraq. A stock solution of 1000ppm was prepared in 250 ml of distilled water with dissolved 0.25 g of reactive blue. Ferrous sulphate Fe$^{2+}$ (India 99 % purity), hydrogen peroxide H$_2$O$_2$ (Germany 45% wt./wt.), hydrochloric acid (98 % purity) and NaOH (Thomas baker) were used in this work.

![Chemical structure of Reactive Blue][21].

2.2. Apparatus

2.2.1. Photo-Fenton batch reactor:

The photo-Fenton process was conducted in a 150 ml glass reactor and it supplied with two UV tubes each of 6 W overwhelming a wavelength of 365 nm (Fig. 2). A magnetic stirrer was used and fixed its speed at 200 rpm to homogenize the solution and a digital WTW pH-720 for measuring the pH of the treated solutions.
2.2.2. Continuous photo-Fenton process:

The schematic of the continuous mode of the photo-Fenton process is clearly explained in Fig. 3. Where a feed tank (1) of (2.5 liter) was employed to provide the dye solution, then it pumped using a water pump (2) to the photo-Fenton reactor (6) through the rotameter device (5). The hollow containing the photo-Fenton reactor (6) was made totally of wood painted by black color with an active volume of 60x60x60 cm$^3$ where the photo-Fenton glass reactor volume had the dimension of 50x50x20 cm$^3$. The photo-Fenton oxidation procedure of dye contaminant is attained through the usage of a postponement of the required amount of Fenton reagent into 2.5 liter of solution. Hydrogen peroxide of 78 mg/l and Fe$^{2+}$ of 20 mg/l were used in the processes. In the reactor, the source of UV light was turned on and the treated wastewater was discharged to the storage tank (7) that samples were taken. The flow rates of 10, 20, 30, 40, 50 ml /min were used in the experimental work. UV spectrometer study was conducted to detect the removal of reactive blue from samples using Eq. (1):

$$R = \frac{A_0 - A_t}{A_0} \times 100\%$$ (1)

Where R is the decolonization efficiency; $A_0$ and $A_t$ are the initial and final concentration of reactive blue dye (mg/l), respectively.
2.3. Statistical analysis:

Statistical design of experiments decreases the number of tests to be carried out, takes into account the correlations between the studied variables and can be used for the optimization of the operating parameters in systems [22]. Response surface methodology (RSM) is a valuable device for conducting factor design and regression analysis [23]. It helps to determine the efficient factors and construct models to test interaction, and to choose optimum conditions of the operating variables for the required response [24]. There are different types of RSM designs such as the central composite design (CCD), Box-Behnken design and D-optimal design. The Box–Behnken design is an independent, rotatable quadratic design with no embedded factorial or fractional factorial points where the variable combinations are at the midpoints of the edges of the variable space and at the center[25]. The experimental response is normally represented by a second order polynomial model (Eq. (2)) with interaction of all the variables [9]:

\[
Y = B_0 + \sum_{i=1}^{q} B_i X_i + \sum_{i=1}^{q} B_{ii} X_i^2 + \sum_{i<j}^{q} B_{ij} X_i X_j + \varepsilon
\]  

(2)

where \(Y\) is the predicted response, \(B_0\) the constant terms, \(B_i\) the linear effect, \(B_{ii}\) the squared effect, \(B_{ij}\) represents the interaction effect and \(\varepsilon\) is a random error. The number of runs \(N\) is measured using Eq. (3) as follows:

\[
N = 2^K + 2K + n
\]  

(3)

where \(K\) represents the number of independent variables (or factors) and \(n\) is the number of replicates of the central experiment [26].

In the present work, the initial reactive blue dye concentration of 20 ppm was used. The experimental ranges of the operating variables are explained in Table 1. These parameters were chosen in this study to investigate their impact on the removal of reactive blue dye using \(\text{H}_2\text{O}_2/\text{Fe}^{2+}/\text{UV}\). The removal efficiency of (R.B) dye was regarded as the required response.

Table 1
Experimental design for degradation of R.B.

| Parameters | Ranges |
|------------|--------|
| \(X_1\): \(\text{H}_2\text{O}_2\) concentration (ppm) | 25-100 |
| \(X_2\): \(\text{Fe}^{2+}\) concentration (ppm) | 5-20 |
| \(X_3\): Irradiation time (min) | 20-90 |
| \(X_4\): Temperature (°C) | 25-60 |
| \(X_5\): pH | 3-10 |
3. Results and discussion:

Forty-six experiments were conducted according to the statistical design of the experiments using RSM-Box–Behnken method performing ranges listed in Table 1 to obtain the studied response as shown its results in Table 2 as follows:

Table 2
Results of the Box–Behnken experiments

| Run no. | $\text{H}_2\text{O}_2$ (ppm) | $\text{Fe}^{2+}$ (ppm) | Irradiation time (min) | Temperature ($^\circ\text{C}$) | PH | Dye removal (%) |
|---------|-----------------------------|------------------------|------------------------|-----------------------------|----|-----------------|
| 1       | 25                          | 12.5                   | 20                     | 42.5                        | 6  | 5.44            |
| 2       | 25                          | 12.5                   | 90                     | 42.5                        | 6  | 6.69            |
| 3       | 100                         | 12.5                   | 20                     | 42.5                        | 6  | 9.81            |
| 4       | 100                         | 12.5                   | 90                     | 42.5                        | 6  | 22.31           |
| 5       | 62.5                        | 5.0                    | 55                     | 42.5                        | 3  | 94.81           |
| 6       | 62.5                        | 5.0                    | 55                     | 42.5                        | 3  | 98.44           |
| 7       | 62.5                        | 5.0                    | 55                     | 42.5                        | 10 | 10.44           |
| 8       | 62.5                        | 20.0                   | 55                     | 42.5                        | 10 | 16.06           |
| 9       | 25                          | 12.5                   | 55                     | 25                          | 6  | 15.44           |
| 10      | 100                         | 12.5                   | 55                     | 25                          | 6  | 34.19           |
| 11      | 25                          | 12.5                   | 55                     | 60                          | 6  | 5.44            |
| 12      | 100                         | 12.5                   | 55                     | 60                          | 6  | 12.31           |
| 13      | 62.5                        | 5.0                    | 20                     | 42.5                        | 6  | 16.69           |
| 14      | 62.5                        | 5.0                    | 90                     | 42.5                        | 6  | 22.94           |
| 15      | 62.5                        | 20.0                   | 20                     | 42.5                        | 6  | 21.69           |
| 16      | 62.5                        | 20.0                   | 90                     | 42.5                        | 6  | 58.56           |
| 17      | 62.5                        | 12.5                   | 55                     | 25                          | 3  | 94.19           |
| 18      | 62.5                        | 12.5                   | 55                     | 25                          | 10 | 10.44           |
| 19      | 62.5                        | 12.5                   | 55                     | 60                          | 3  | 98.56           |
| 20      | 62.5                        | 12.5                   | 55                     | 60                          | 10 | 15.44           |
| 21      | 25                          | 5.0                    | 55                     | 42.5                        | 6  | 17.31           |
| 22      | 100                         | 5.0                    | 55                     | 42.5                        | 6  | 5.44            |
| 23      | 25                          | 20.0                   | 55                     | 42.5                        | 6  | 45.44           |
| 24      | 100                         | 20.0                   | 55                     | 42.5                        | 6  | 53.56           |
| 25      | 62.5                        | 12.5                   | 20                     | 42.5                        | 3  | 91.69           |
| 26      | 62.5                        | 12.5                   | 90                     | 42.5                        | 3  | 97.94           |
| 27      | 62.5                        | 12.5                   | 20                     | 42.5                        | 10 | 10.44           |
| 28      | 62.5                        | 12.5                   | 90                     | 42.5                        | 10 | 22.94           |
| 29      | 62.5                        | 5.0                    | 55                     | 25                          | 6  | 35.44           |
| 30      | 62.5                        | 20                     | 55                     | 25                          | 6  | 49.81           |
| 31      | 62.5                        | 5.0                    | 55                     | 60                          | 6  | 10.44           |
| 32      | 62.5                        | 20.0                   | 55                     | 60                          | 6  | 56.06           |
| 33      | 62.5                        | 12.5                   | 20                     | 25                          | 6  | 9.19            |
3.1 Regression model:

From the experimental results presented in Table 2, the mathematical correlations of dye removal (Y) response which is related to the quadratic and interaction effects of the operational parameters, is revealed in Eq. (4) as follows:

Dye removal (%): \[ Y = 192 + 0.616X_1 - 6.66X_2^2 + 0.109X_3 + 0.120X_4 - 36.880X_5 - 0.006X_1X_2 + 0.099X_2X_2 + 0.006X_3^2 - 0.009X_4^2 + 1.811X_5^2 + 0.018X_1X_2 + 0.002X_1X_1 - 0.005X_1X_4 + 0.014X_1X_1 + 0.029X_2X_3 + 0.059X_2X_4 + 0.069X_2X_5 + 0.002X_3X_1 + 0.014X_3X_3 + 0.006X_4X_5 \] \( R^2 = 0.9294 \) \hspace{1cm} (4)

Experimental results had been analyzed using Portable Statgraphics Centurion 15.2.11.0 statistical software to approximate the response of dependent variables and to obtain the optimum conditions of the operating variables. Moreover, the statistical significance was analyzed using analysis of variance (ANOVA)[27] as shown in Table 3. The ANOVA table decomposes the variability of dye Removal (%) into contributions due to various factors. The contribution of each factor is measured having removed the effects of all other factors. The P-values test the statistical significance of each of the factors. Since four of P-values are less than 0.05, these factors have a statistically significant effect on dye Removal (%) at the 95.0% confidence level. The correlation coefficient \( R^2 \) of 0.9294 demonstrates how well the model fits the experimental data.
Table 3
ANOVA test results for the photo-Fenton process

| Source                      | Sum of Squares | Df | Mean Square | F-Ratio | P-Value |
|-----------------------------|----------------|----|-------------|---------|---------|
| MAIN EFFECTS                |                |    |             |         |         |
| Irradiation Time (min)      | 1040.66        | 2  | 520.331     | 6.30    | 0.0046  |
| Hydrogen Peroxide (ppm)     | 905.631        | 2  | 452.815     | 5.48    | 0.0085  |
| Ferrous Sulphate (ppm)      | 2439.0         | 2  | 1219.5      | 14.76   | 0.0000  |
| PH                          | 30504.5        | 2  | 15252.3     | 184.54  | 0.0000  |
| Temperature                 | 88.0016        | 2  | 44.0008     | 0.53    | 0.5919  |
| RESIDUAL                    | 2892.72        | 35 | 82.6491     |         |         |
| TOTAL (CORRECTED)           | 40459.8        | 45 |             |         |         |

3.2 Effect of H₂O₂ dosage:

In the photo-Fenton process, the initial concentration of hydrogen peroxide plays an important part in oxidizing organic compounds as an oxidizing agent. From a practical perspective, the selection of an optimal concentration of hydrogen peroxide to degrade R.B by Fenton oxidation is important due to the cost of hydrogen peroxide. The efficiency of degradation may be constrained with either small or excess dose H₂O₂ [28]. In this study, concentrations of H₂O₂ in the range of 25-100 ppm have also been evaluated for dye degradation to determine the optimal H₂O₂ dosage for R.B removal as shown the results in Fig. 4. It was found that the efficiency of dye removal was around 17.84% when the dosage of H₂O₂ was 25 ppm, while the best performance of dye removal was 31.06% at 78 ppm of H₂O₂. With the further rise of hydrogen peroxide concentration from 78 to 100 ppm, the efficacy of dye removal is decreasing. The consequence of this can be explained by the so-called radical scavenger of hydrogen peroxide towards OH• radicals as revealed in Eq. (5) and Eq. (6) when it presented in a higher concentration [29]. Excess of H₂O₂ will react with OH' competing with organic pollutants which leads to the formation of perhydroxyl radicals (OH₂•) which are significantly less reactive species than hydroxyl radicals and thus directly influence the efficiency of dye degradation.

\[
H₂O₂ + OH• \rightarrow HO₂• + H₂O \tag{5}
\]

\[
HO₂• + OH• \rightarrow H₂O + O₂ \tag{6}
\]

So, the bests dosage of H₂O₂ for R.B removal was 78 ppm. These findings are the same that have been reached by Muruganandham et al., 2003[30].
3.3 Effect of Ferrous Sulphate:

The dosage of Fe$^{2+}$ is also a significant operational parameter in the degradation of the Photo-Fenton process. The efficiency of the removal of dye is affected by the concentration of Fe$^{2+}$ ions that catalyze the hydrogen peroxide decomposition resulting in the creation of hydroxyl radicals with a strong oxidation potential according to Eq. (7) to Eq. (10) [31].

\begin{align*}
Fe^{2+} + H_2O_2 & \rightarrow Fe^{3+} + HO^* + HO^- \\
Fe^{3+} + H_2O_2 & \rightarrow Fe^{2+} + O_2^*H + H^+ \\
Fe^{3+} + O_2^*H & \rightarrow Fe^{2+} + O_2 + H^+ \\
H_2O_2 + hv & \rightarrow 2HO^* 
\end{align*}

Fig. 5 explains the effect of Fe$^{2+}$ concentration on the experimental results of dye removal efficiency through the process of the photo-Fenton reactor, in which the iron concentration varied from 5 to 20 ppm. The increase of the treatability is due to the fact that the higher the ferrous concentration, the more photo-Fenton reactions will affect the OH*, which results in a higher rate of dye removal. The high removal efficiencies can be clarified by that higher ferrous doses not only make the redox reaction complete but also the fact that oxidation reaction is coupled to coagulation occurring due to the presence of ferrous, hence these metallic ions perform a double role in the process as a catalyst and coagulant [32,33]. The residual dye fraction was found to decrease significantly with an increase in dye removal from 22.73% to 50.27% respectively by rising ferrous dose from 5 to 20 ppm. This can be seen that
R.B degradation efficacy improves with a higher initial concentration of Fe\(^{2+}\). These explanations are agreed with that reached by Kang et al. [34].

![Figure 5. Effect of Ferrous Sulphate on dye removal, hydrogen peroxide = 78 ppm, irradiation time = 55min, temperature=42.5 °C, PH=7.](image)

3.4 Irradiation time effect:

The performance of the photo-Fenton process depends on the formulation and recovery rates of OH• differ depending on the type of organic substrate and the time of irradiation[35]. Therefore, the effect of the irradiation time required for photo Fenton treatment was studied at different time periods (20- 90 min) to obtain the best dye removal. Fig. 6 clarify the effect of irradiation time on the photo-Fenton degradation of Reactive Blue dye. At 90 minutes, the best time irradiation was about 58.62% dye removal efficiency for R.B. It is apparent that the percentage of dye removal rises with that irradiation time. If the dye solution is exposed to UV for a longer period of time, this will enable further free radicals to be produced as a result of several reactions. These explanations are agreed with that achieved through Hassan et al.,2019[36].
3.5 Effect of temperature:

The photo-Fenton process is usually carried out at room temperature. However, considering that water coming from the textile industry has a temperature of 60 to 90 °C, we found it extremely important to evaluate the effect of temperature on the dye removal rate[37]. Therefore, it was taken into consideration in this study with the range of (25-60) °C to test the effect of temperature on R.B decomposition. A raise in the R.B dye degradation rate was noted with an increase in temperature from 25 to 40 °C that reaches 70.13% at 40 °C, which may be due to an increase in the rate of interaction of organic material with hydroxyl radicals, while the R.B dye removal efficiency decreases to 59.68% due to the increase temperatures up to 60 °C as shown in Fig. 7. It can be possible that hydrogen peroxide may undergo self decomposition at high temperature as shown in Eq. (11).

$$H_2O_2 \rightarrow H_2O + 0.5 O_2 \quad (11)$$

This are the similar results founded by Panizza and Cerisola [38]. Therefore, the best temperature was chosen as 40 °C to remove R.B by photo-Fenton.
Figure 7. Effect of temperature on dye removal, hydrogen peroxide = 78 ppm, ferrous sulphate = 20 ppm, irradiation time = 90 min, pH = 7.

3.6 Effect of pH:

Another significant factor that must be taken into consideration in the oxidation via the photo-Fenton process is the pH. The acidic factor of the solution determines the rate of hydroxyl radical formation and the nature of iron species in solution [39]. The hydroxyl radicals can form acidic environments effectively [40]. The value of pH was adjusted by using amounts of HCl and/or NaOH to obtain the designed range of 3–10. The effect of the pH parameter on dye removal is shown in Fig. 8. It is apparent from the findings that an increase in pH from 3 to 10 will decrease the dye removal from 84.82% to 54.97%. The reduction in the elimination rate within this pH range is due to the lowering of radical hydroxyl concentration. At pH larger than 3, dye removal values decrease due to the formation of Fe$^{2+}$ complexes and sedimentation of ferric oxyhydroxides (Fe(OH)$_3$). Furthermore, the oxidation potential of hydroxyl radicals decreased with an increase in the pH. For pH more than 6, the dye removal efficiency decreased to 54.97% for pH equals 10. This behavior could be explained by the formation of ferric hydroxide complexes during the reaction, which prevents hydrolysis of hydrogen peroxide catalyzed by ferric iron. The best value of pH for R.B removal was observed to be 3. Lucas and Peres found similar results for pH effect [41].
Figure 8. Effect of pH on reactive blue dye removal, hydrogen peroxide = 78 ppm, Ferrous Sulphate = 20 ppm, irradiation time = 90 min, temperature = 40 °C.

3.7 Optimization of the operating variables:

The photo-Fenton degradation capability of the system was tested under a set of conditions. The best system parameters were chosen based on the capacity to reduce the concentration of dye in the shortest amount of time. The optimum values of the operating variables and the authenticated values of the studied response aimed at the treatment of reactive blue dye that are the $[\text{H}_2\text{O}_2]$, $[\text{Fe}^{2+}]$, irradiation time, temperature and pH are that 78 ppm, 20 ppm, 90 min, 40 °C and 3, respectively. The obtained value of the removal efficiency according to these optimum conditions was about 84.82%.

3.8 Effects of Intensity of Light and Distance:

Ultraviolet light irradiates degrades dye quickly because of the $\text{OH}^\bullet$ radical content dose. The sample dye loss may be correlated with the generation of rising hydroxyl radicals, which serves as a strong oxidizing agent [42]. So, the effect of the UV radiation intensity on the dye removal efficiency was studied using the optimum conditions, by treating the samples in (6, 12, 18 and 24W) UV chamber. The influence of various ultraviolet light intensity on dye degradation rate was observed, and dye degradation was greatly maximized by an increase in ultraviolet intensity, suggesting that ultraviolet light had a significant impact on dye degradation. Similar results were reported for the intensity effect by [43]. In addition, the impact of UV lamps distance on the efficiency of decolorization was analyzed. The distance of UV lamps was adjusted from the dye solution as 15, 30, and 50 cm. The results of the intensity light effects relating to the distance are shown in Fig. 9. The dye removal by photo-Fenton was a straight-line decreasing with the rising of UV lamps from the reactor. The optimum distance, at which decolorization efficiency occurs most rapidly is 15 cm from the dye solution surface. The results agree with the findings of [44].
Figure 9. Effects of intensity of light and distance on dye removal efficiency, $H_2O_2=78$ ppm, $Fe^{+2}=20$ ppm, pH=3, irradiation time =90 min , temperature=40 °C.

3.9 Continuous photo Fenton process:

The photo-Fenton scheme was deliberate by the continuous oxidation procedure. The UV radiation, light radiation direction and flow rate were deliberate. The photo Fenton and squalor competence of the organization were verified below individually set of circumstances.

3.9.1 Light direction effect:

To study the effect of direction on R.B removal efficiency, experiments were carried out in different light directions (top and side) by changing the place of the UV lamp in the photo-Fenton chamber and keeping the initial dye concentration at 40 mg/l, with the optimum conditions of the concentrations of iron and hydrogen peroxide. The results are shown in Fig. 10 revealed that the efficiency of dye removal increases when the direction of the light from the top in comparison to the side direction of the light. The efficiency of dye reduction in the top direction was 99.16% compared to 86.22% in the case of the side direction where both of them had maintained at 24W. This behavior could be explained by the fact that the surface area of the sidelight exposure was smaller than that of the top light so that the light direction had a significant impact on the degradation rate. When the top light direction increases, the generation rate of free radicals was more than the side direction of the light as states by Shaker et al.,2011[45].
Figure 10. Effect of light direction on dye removal, $H_2O_2=78$ ppm, $Fe^{+2}=20$ ppm, pH=7, flow rate=20 ml/min.

3.9.2 Effect flow rate:

The dye removal efficiency had been analyzed by changing the flow rate of the dye solution from 10 to 50 ml/min. This range of treatment flow rate is surely a pilot-scale process. The effect of the flow rate on the decolorization of R.B at an intensity of 24 watts, pH 7, and 40ppm as the initial concentration of dye is shown in Fig. 11. The highest color removal efficiency was completely obtained, i.e. 100 %, at 10ml/min of the flow rate. Fig. 11 revealed that an increase of the flow rate from 10 ml/min to 50 ml/min the dye removal efficiency was decreased which means that the flow rate and the color removal efficiency are inversely proportional. Increasing the flow rate of the polluted solution, the UV lamp will exposure in a shorter time; therefore, the amount of $H_2O_2$ should be decreased to form more of hydroxyl radical ($OH^•$) where these results agree with[46].

Figure 11. Effect of flow rate on the dye removal, pH=7, $H_2O_2=78$ ppm, $Fe^{+2}=20$ ppm, Intensity= 4W, Top direction.
4. Conclusions:

Reactive blue dye (R.B) was removed from simulated wastewater by utilizing a photo-reactor containing four fixed UV lamps in a batch and continuous mode under the effect of several operating variables (dosage of H$_2$O$_2$; dosage FeSO$_4$; pH; temperature; irradiation time) using Box–Behnken design and Portable Statgraphics Centurion statistical software to design the experiments and conducting the mathematical correlation of the studied parameters. The optimal conditions of the operating variables; dosage hydrogen peroxide, ferrous sulphate, pH, temperature and irradiation time were 78 ppm, 20 ppm, 3, 40°C and 90 min, respectively which give 84.82% of removal efficiency. Moreover, the highest dye removal efficacy was obtained at a light intensity of 24 w and a distance of 15cm. For continuous system, the dye removal efficiency decreased with an increase of flow rate and the top direction compared to the side direction of the light where the removal percentage of (R.B) was 100% for the flow rate of 10 ml/min while it was 84.16% for a flow rate of 50 ml/min. The results show that the photo-Fenton method is an effective treatment method of Wastewater containing dyes and could be useful to maintain large-scale photo-Fenton oxidation reactors.

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