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Development of Solar Photo-Fenton Process for the Removal of Color, COD, and Turbidity from Institutional Wastewater

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Abstract: In recent years, hard research has been aimed to develop effective technology for the treatment of wastewater and industrial effluent containing organic/inorganic contaminants. Amongst several technologies, the advanced oxidation processes (AOPs) recently have played a major role in the treatment of wastewater. In this study, the treatment of institutional wastewater by solar-photo-Fenton ($UV/Fe^{2+}/H_2O_2$) process based on AOPs was examined in terms of % color, % turbidity, and % COD removal. The solar-$UV/Fe^{2+}/H_2O_2$ process has revealed a higher removal of color (91%), turbidity (90%), and COD (86%) than the other processes. The effect of various experimental parameters such as hydrogen peroxide (H_2O_2) concentration (0.25 to 1.25 g/L) and Ferrous ion (Fe^{2+}) concentration (0.005 to 0.12 g/L), initial pH (2 to 10), reaction time (30 to 180 min) on the color, turbidity and COD removal has been studied to find out the optimum conditions leading to maximum removal efficiency of the solar $UV/Fe^{2+}/H_2O_2$ process. The best results of the solar $UV/Fe^{2+}/H_2O_2$ process of institutional wastewater treatment have been found using 0.75 g/L of H_2O_2, 0.045 g/L of Fe^{2+}, pH of 4, after 120 min of reaction time. The present study revealed that the solar-$UV/Fe^{2+}/H_2O_2$ process in an AOPs was well efficient in the institutional wastewater treatment, accomplishing a higher pollutant removal rate. The solar-$UV/Fe^{2+}/H_2O_2$ process is an effective treatment technique for the removal of pollutants from institutional wastewater.

Keywords: Institution Wastewater, Solar Photo-Fenton Process, Color, Turbidity, COD Removal, Operating Parameters, Optimization

1. Introduction

Water is vital to survival on earth; it is one of the important requirements in human life. Increasing water demand from the world population has become a major global concern [1]. It is essential that wastewater management be considered as part of an integrated, full life cycle, an ecosystem-based management system that operates across all three dimensions of sustainable development (social, economic, and environmental), geographical borders, and includes both freshwater and marine waters [2]. Despite wastewater reuse being able to provide important economic benefits by reducing wastewater disposal and irrigation costs, some public health and environmental concerns must be addressed for the safe and rational implementation of wastewater reuse in an agricultural setting [3].

The advanced oxidation processes (AOPs) could be promising for the degradation of persistent contaminants which are not removed during a biological process. The high efficiency of AOPs is associated with the generation of highly reactive •OH radicals, which can oxidize many organic compounds. Eventually, complete mineralization to CO_2, H_2O, and mineral compounds can be obtained. The most extensively studied AOPs are the solar photo-Fenton ($UV/Fe^{2+}/H_2O_2$) process [4–6], photocatalysis [7, 8], UV/H_2O_2 [9, 10], and others [11].

In recent years, the Fenton reaction has been widely applied in the field of environmental remediation, especially

for the decontamination of wastewaters containing chemically and biologically recalcitrant organic pollutants [12]. Various organic pollutants could be attacked non-selectively by •OH and decomposed to CO_2, H_2O, and inorganic ions [13, 14]. The UV/Fe^{2+}/H_2O_2 process involves sunlight or artificial light irradiation, which increases the rate of contaminants degradation by increasing the production of •OH from hydrogen peroxide (H_2O_2) decomposition and synchronously accelerating the reduction of Fe^{3+} to Fe^{2+}. Moreover, the photolysis of H_2O_2 also plays an important role in the photo-Fenton process to break the O–O bond for the production of •OH [15, 16].

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + \bullet OH + OH^- \quad (1)$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2\bullet + H^+ \quad (2)$$

$$[Fe^{3+}(OH)]^{2+} + h\nu \rightarrow Fe^{2+} + \bullet OH \quad (3)$$

$$Fe^{3+} + H_2O + h\nu \rightarrow Fe^{2+} + \bullet OH + H^+ \quad (4)$$

$$Fe^{3+}(RCO_2) + h\nu \rightarrow Fe^{2+} + CO_2 + {}^{\cdot}R \quad (5)$$

$$H_2O_2 + h\nu \rightarrow {}^{\cdot}OH \quad (6)$$

$$\bullet OH + \text{organic pollutants} \rightarrow CO_2 + H_2O \quad (7)$$

$$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2 \quad (8)$$

In the last few decades, using the Fenton oxidation process for the treatment of institutional wastewater has been widely reported. However, the Fenton reaction has some limitations in practical application, such as the requirement for a large number of chemical reagents, large production of ferric hydroxide sludge, and very slow catalysis of the ferrous ions generation [17]. Direct photolysis of H_2O_2 produces •OH which can be used for the degradation of organic compounds, and in turn, increases the rate of degradation of organic pollutants [18]. Conventional biological processes are usually the most cost-effective alternatives for treating these effluents, but it is widely known that high concentrations of toxic or non-biodegradable compounds prevent mineralization [19]. Moreover, 12 h are required to achieve chemical oxygen demand (COD) removal efficiencies higher than 90% [20].

Advanced oxidation processes (AOPs) are an efficient alternative that could be used to solve this problem. Among AOPs, the ferrioxalate-induced solar-UV/Fe^{2+}/H_2O_2 process is a clean technology that is considered environmentally friendly and competitive at the industrial scale [21]. The AOPs have been demonstrated to be innovative suitable technologies to reduce organic load or toxicity and enhance the treatability of wastewater because they can oxidize refractory organic compounds into harmless substances and even lead to mineralization end-products [22].

The solar-UV/Fe^{2+}/H_2O_2 process is a clean technology that is considered environmentally friendly and competitive at any wastewater treatment [21]. Over the last years, the solar-UV/Fe^{2+}/H_2O_2 process became one of the most interesting alternative processes for wastewater treatment, given its simplicity and feasible application [23]. Besides the enhancement of the photo-Fenton process for wastewater treatment using natural products and solar radiation, another important factor worthy of consideration is the iron source. Up to date, different iron materials have been tested as precursors of the photo-Fenton process, including natural iron oxide [24], residue-based iron [24], zero-valent metallic iron [25], magnetic composites [26], or modified clays and carbon-based materials [27] to promote organic pollutant degradation. However, among the iron species, special attention is associated with the ferric ion, given that in natural conditions, Fe^{2+} is oxidized to Fe^{3+} by oxygen or H_2O_2, increasing the investigative interest for ferric sources.

The physical, chemical, and biological technologies have been widely used to treat wastewater. However, most of the real wastewater contains many organic pollutants with high toxicity and low biodegradability [28]. Over the last years, the solar-UV/Fe^{2+}/H_2O_2 process became one of the most interesting alternative processes for wastewater treatment, given its simplicity and feasible application [23, 29–31]. Since solar energy is a widely accessible energy source its application in processes beyond the generation of current or process heat will give a benefit the sustainability of a wide range of industrial techniques. The detoxification of wastewater by different solar photocatalytic systems is extensive work in the research field [32]. So, this research study was focused on the removal of pollutants in terms of color, turbidity, and COD removal and also studied various operating parameters based on the photo AOPs for the institutional wastewater.

The goal of this research work is to investigate the treatment performance of the solar-UV/Fe^{2+}/H_2O_2 process using different operating parameters in removing pollutants from institutional wastewater. Evaluating the operating variables such as H_2O_2 and Fe^{2+} dosages, pH, and reaction time on the removal efficiencies of color, turbidity, and COD by using the solar photo Fenton method.

2. Material and Methods

2.1. Characterization

The wastewater sample for this study was taken from Jimma Institute of Technology (JIT), Jimma University (JU), Jimma, Ethiopia. The sampling method had been a random sampling technique. Samples were collected in plastic bottles having 20 L volume. The wastewater sample was taken by inserting the plastic bottle to a certain depth of the wastewater at different positions. Then the sample collected was merged to form a composite sample represented in Figure 1. The experimental tests of the sample were conducted in JIT, JU, environmental engineering laboratory to analyze its characteristics wastewater and it was given in Table 1. Chemicals used in this study were Potassium dichromate, concentrated sulfuric acid, ferrous ammonium sulfate (FAS), Mercuric sulfate, Silver sulfate, Ferron indicator, and organic free distilled water, Sodium hydroxide, Diluted sulfuric acid, ferrous iron, and hydrogen peroxide.

Figure 1. *Institutional wastewater collection.*

2.2. Methods

The experimental setup of the solar-UV/Fe^{2+}/H_2O_2 process is shown in Figure 2. The solar-UV/Fe^{2+}/H_2O_2 process degradation of experiments was carried out in a 500 mL beaker under natural solar irradiation, outside the laboratory of environmental engineering, JiT, JU by the mixture of H_2O_2, Fe^{2+} and adjusting to different pH with 500 mL sample. The experiment was repeated for thirty runs. The working volume of the beaker was 500 mL. The Fe concentration used in all solar-UV/Fe^{2+}/H_2O_2 experiments was 0.005 to 0.12 g/L. The H_2O_2 concentration used in all solar-UV/Fe^{2+}/H_2O_2 experiments was 0.25 to 1.25 g/L. The reaction time interval used in all solar-UV/Fe^{2+}/H_2O_2 experiments was 30 to 80 min. The pH was adjusted at the beginning of each run to the range from 2 to 10. All pH adjustments were done with analytical grade NaOH and H_2SO_4. The pH range used in all experiments was 2 to 10. During the solar-photo-Fenton treatment, the wastewater was exposed to solar light for different duration of time.

The COD concentration in the wastewater sample was measured by the closed reflux method. The color absorbance in wastewater was measured with UV/Vis-spectrophotometer 6700 (Bibby scientific Ltd). The turbidity of the wastewater sample and treated water was measured by a microprocessor turbidity meter (HI 93703). Dissolved oxygen was measured dissolved oxygen meter (9500 DO_2 meter). The pH was measured by a pH meter (pH 3310). Oakton waterproof portable meter kit also measured pH, ORP, ION, conductivity, TDS, salinity, resistivity, DO, and temperature.

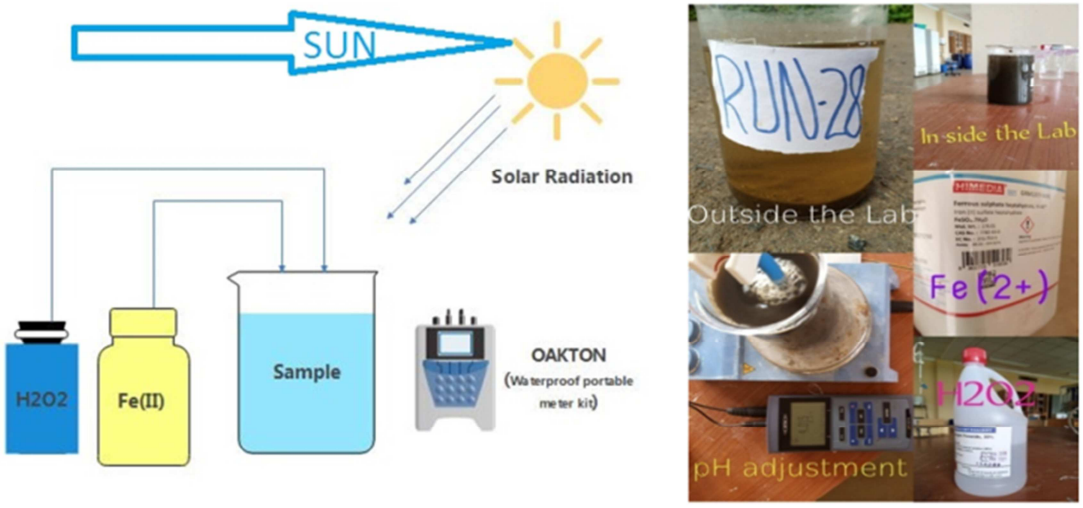

Figure 2. *Experimental setup of solar photo Fenton process.*

2.3. Analysis

The efficiency of color, turbidity and COD removal was calculated using the following equation is given below.

2.3.1. Color Removal, (%)

$$Color\ removal, (\%) = \frac{Abs_i - Abs_t}{Abs_i} * 100 \qquad (9)$$

Where,

Abs_i and Abs_t are the absorbance of initial and at any time t samples for the corresponding wavelength $\lambda_{max.}$

2.3.2. COD Removal, (%)

$$COD\ removal, (\%) = \frac{COD_i - COD_t}{COD_i} * 100 \qquad (10)$$

Where,

COD_i and COD_t (in ppm) are the chemical oxygen demand of initial and at reaction time t, respectively.

2.3.3. Turbidity Removal, (%)

$$\text{Turbidity removal, } \% = \frac{\text{Turbidity (initial)} - \text{Turbidity (final)}}{\text{Turbidity (initial)}} * 100 \qquad (11)$$

3. Results and Discussion

3.1. Various AOPs Comparison

The operational parameters such as Fe^{2+} – 0.045 g/L, H_2O_2 – 0.75 g/L, COD – 296 mg/L, pH – 4, and reaction time – 120 min were applied in of H_2O_2, Fe^{2+}, Fe^{2+}/H_2O_2, and solar-UV/Fe^{2+}/H_2O_2 processes to compare their efficiency in terms of % color, % turbidity, % COD removal from the institution wastewater and the results are given in Figure 3. It is evident from Figure 3, the single H_2O_2 and Fe^{2+} processes were not effective for the removal of color (30% & 22%), turbidity (29% & 21%) and COD (25% & 18%). Moderately low color (60%), turbidity (55%), and COD (45%) removal were observed for the Fe^{2+}/H_2O_2 processes. The combination of solar UV with Fe^{2+}/H_2O_2 such as solar-UV/Fe^{2+}/H_2O_2 processes for the removal of color, turbidity, and COD were about 91%, 90%, and 86%, respectively. As expected, the hybrid solar- UV/Fe^{2+}/H_2O_2 process has the best % color, % turbidity, and % COD removal efficiency within the investigated solar UV with AOPs. The above results indicated that introducing H_2O_2, Fe^{2+} and into the sloar UV system led to a significant increase in the color, turbidity, and COD removals. This could be attributed to the available parallel pathways to generate abundant •OH radicals for efficient color, turbidity, and COD removals from institution wastewater within a shorter reaction time [33, 34]. Further studies were focused on the solar-UV/Fe^{2+}/H_2O_2 process for removal of % color, % turbidity, and % COD from institution wastewater.

3.2. Effect Operating Parameters

The solar UV/Fe^{2+}/H_2O_2 processes are affected by several operating parameters such as H_2O_2 (0.25 to 1.25 g/L) and Fe^{2+} (0.005 to 0.12 g/L) concentration, solution pH (2 to 10), and reaction time (30 to 180 min) was studied in terms of % COD, % color and % turbidity removal and the results and discussion was given below.

Table 1. *Characteristics of institutional wastewater.*

№	Parameters	Range of concentrations in institutional wastewater
1	COD	1296 mg/L
2	pH	6.54
3	Conductivity	2.508 µS/cm
4	TDS	1358 mg/L
5	DO	46 mg/L
6	Turbidity	89.65 NTU
7	Temperature	20.9°C
8	Odor	Objectionable
9	Color	Slightly brown

Table 2. *Overview of work done in the area of solar photo-Fenton application in recent years.*

No	Process	Type of wastewater	Parametsr	Maximun removal Efficiency (%)	References
1	UV/H_2O_2/Fe^{2+}	Textile	Color	91.92%	[37]
2	UV/H_2O_2/Fe^{2+}	Winery	COD Color	70±3.3% 75±2.2%	[38]
3	UV/H_2O_2/Fe^{2+}	Citrus	COD Color	77% 53%	[39]
4	Fe^{2+}/H_2O_2	Colored	Color	95%	[40]
5	UV/H_2O_2/Fe^{2+}	Municipal	COD	50%	[41]
6	UV/Fe^{2+}/H_2O_2	Industrial	COD Color	84.40% 96.50%	[42]
7	UV/Fe^{2+}/H_2O_2	Wastewater	COD	90%	[43]
8	UV/Fe^{2+}/H_2O_2	Colored soft drink	COD	99.3%	[44]
9	UV/Fe^{2+}/H_2O_2	Petroleum	COD	74.7%	[45]
10	Soloar UV/Fe^{2+}/H_2O_2	Institution	Color Turbidity COD	91% 90% 86%	This study

3.2.1. The Effect of Reaction Time

Solar radiation is the main factor that influences degradation during the different time intervals of exposed solution due to the generation of •OH radicals. The influence of reaction time on the removal of % color, % turbidity, and % COD from institutional wastewater using solar-UV/Fe^{2+}/H_2O_2 process with H_2O_2 – 0.75 g/L, Fe^{2+} - 0.045 g/L, solution pH-4, and reaction time-120 min the results are depicted in the Figure 4. The results indicate that the % color, % turbidity, and % COD removal have increased with an increase in treatment time. The color, turbidity, and COD removed are proportional to the generation of •OH radicals by the solar UV/Fe^{2+}/H_2O_2. When the treatment time increases, a constant amount of •OH radicals are produced. Consequently, an increase in the treatment time increases the color, turbidity, and COD removal efficiency.

The regeneration of Fe^{2+} from photoreduction of Fe^{3+} by solar radiation increases the decolorization/degradation of pollutants from wastewater. In the presence of sufficient H_2O_2 and Fe concentrations, the exposure to solar radiation increases the amount of degradation in wastewater due to the continuous regeneration of Fe^{2+} from the photo-reduction of

Fe^{3+} by solar light (according to equations 12 and 13) and the generation of extra free radicals (mainly •OH) due to ferrioxalate ($Fe^{3+}(OH)^{2+}$) photochemistry as commented in the following section.

$$Fe^{3+}+H_2O\ Fe^{3+}\rightarrow(OH)^{2+}+H^{+} \quad (12)$$

$$Fe^{3+}(OH)^{2+}+hv\rightarrow Fe^{2+}+\bullet OH \quad (13)$$

3.2.2. The Effect of H_2O_2

The dosing rate of H_2O_2 is considered to be one of the most important factors which should be considered in the solar UV/Fe^{2+}/H_2O_2 process. It was found that color, turbidity, and COD removal efficiency increase with increasing the dosing rate of H_2O_2 from 0.25 to 1.25 g/L. The result was shown in Figure 5. The highest percentage removal of COD was attained at a time of 120 min, Fe^{2+} concentration of 0.045 g/L, and initial effluent pH of 4 when using an H_2O_2 dosing rate of 0.750 g/L, so further addition of H_2O_2 is not necessary. Excessive H_2O_2 reacts with •OH (equation (14)) competing with organic pollutants and consequently reducing treatment efficiency. Excessive H_2O_2 may cause an unutilized •OH radical.

$$H_2O_2+\bullet OH\rightarrow HO_2\bullet+H_2O \quad (14)$$

The H_2O_2 is the main source of •OH in the solar UV/Fe^{2+}/H_2O_2 process. H_2O_2 is an oxidant widely used for water treatment due to its oxidation potential of around 1.4 V at near-neutral pH [35]. Moreover, it is cheap, safe, easy to handle, and does not generate residues as it easily decomposes to H_2O and O_2. H_2O_2 alone (even at non-toxic microbial concentration, lower than 0.050 g/L) in combination with natural sunlight can promote very high microbial inactivation rates. To find out the optimum condition in solar photo Fe^{2+}/H_2O_2 reaction different dosages of H_2O_2 were added to the solution. All experiments were exposed to different duration of solar radiation.

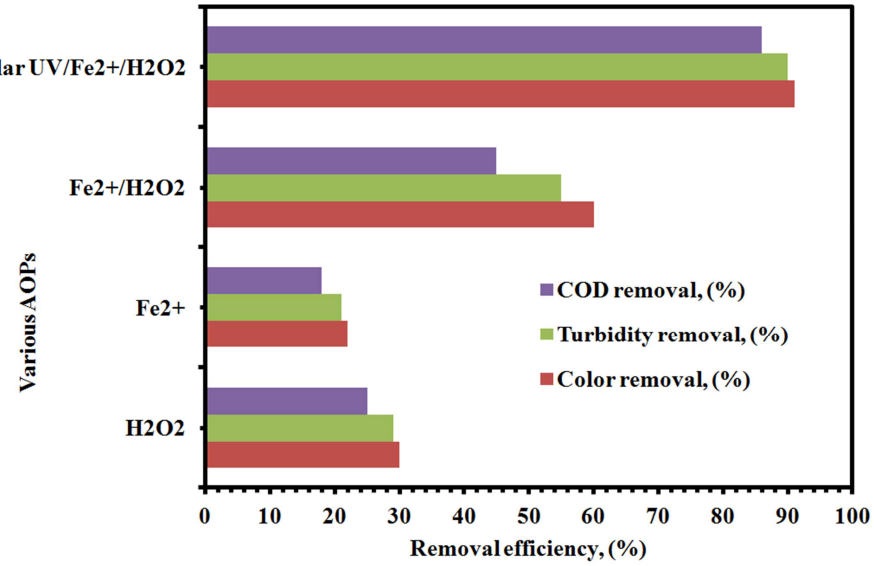

Figure 3. *Comparisons of various AOPs on removal efficiency of % color, turbidity, and COD.*

3.2.3. Effect of Fe^{2+}

The amount of Fe^{2+} ion is one of the main parameters which influence the solar-UV/Fe^{2+}/H_2O_2 processes. In this study, to obtain the optimal initial Fe^{2+} concentration, the investigation was carried out in the range of 0.005-0.12 g/L Fe^{2+}, at different pH, solar radiation, and H_2O_2 concentration. The result was shown in Figure 6. The Ferrous Sulphate Heptahydrate ($FeSO_4.7H_2O$) was introduced into the solution at start-up. The solutions were continuously stirred using a magnetic stirrer. This is because increasing Fe^{2+} produces more quantity of •OH radical with solar radiation. This •OH radical enhances the oxidation of pollutants and the removal of color, turbidity, and COD. It can be seen that the removal rate of color, turbidity, and COD increased with the increasing amount of Fe^{2+} in the range of 0.005 to 0.045 g/L. It was known that Fe^{2+} had a catalytic decomposition effect on H_2O_2. When the Fe^{2+} concentration increased, the catalytic effect increased accordingly. However, for Fe^{2+} doses higher than 0.045g/L, the color, turbidity, and COD percent removal decreased slightly. This decrease was essentially due to the competitive consumption of •OH radicals. The amounts of Fe^{2+} ions should be as low as possible for economic and environmental reasons; high amounts of Fe^{2+} ions might produce a larger quantity of Fe^{3+} sludge. The removal/treatment of the sludge containing Fe^{3+} at the end of the wastewater treatment is expensive and requires a large number of chemicals and manpower. As shown in the inset of Figure 6, 0.045 g/L Fe^{2+} can be used as an optimum dosage in this study. Solar radiation was used as an alternative to tackle this problem in the degradation of pollutants and molecules. Fe^{2+} has relatively high solubility under acidic or neutral conditions and can be oxidized easily into Fe^{3+} by dissolved oxygen in water [36].

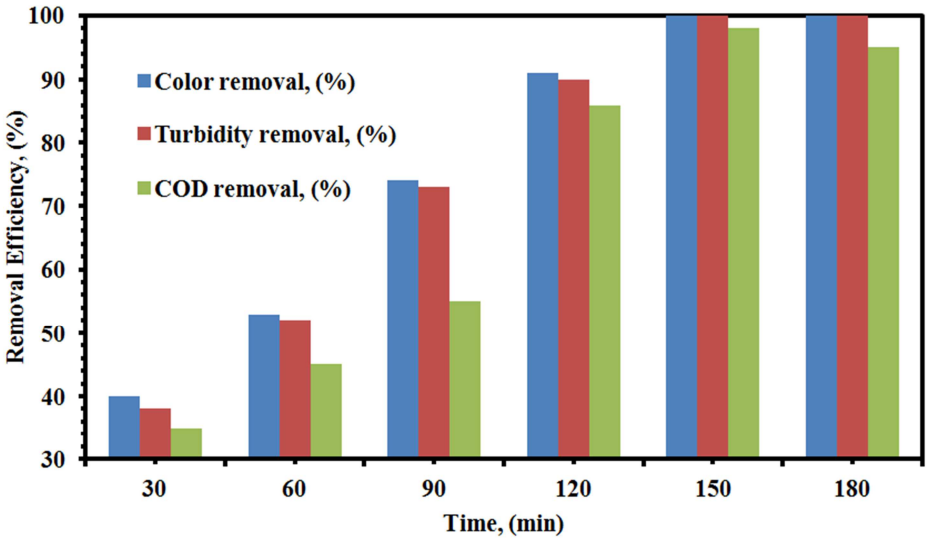

Figure 4. *Effect of reaction time on removal efficiency of % color, turbidity, and COD.*

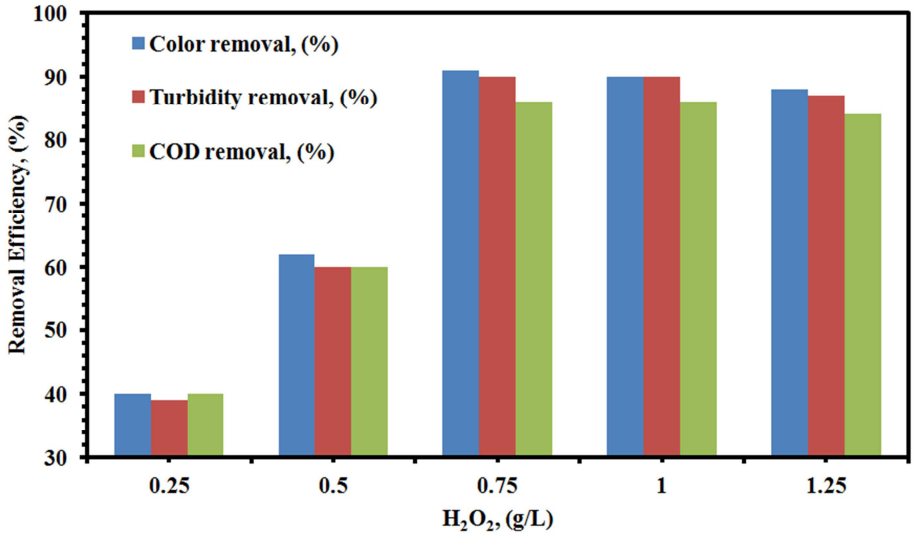

Figure 5. *Effect of H_2O_2 on removal efficiency of % color, turbidity, and COD.*

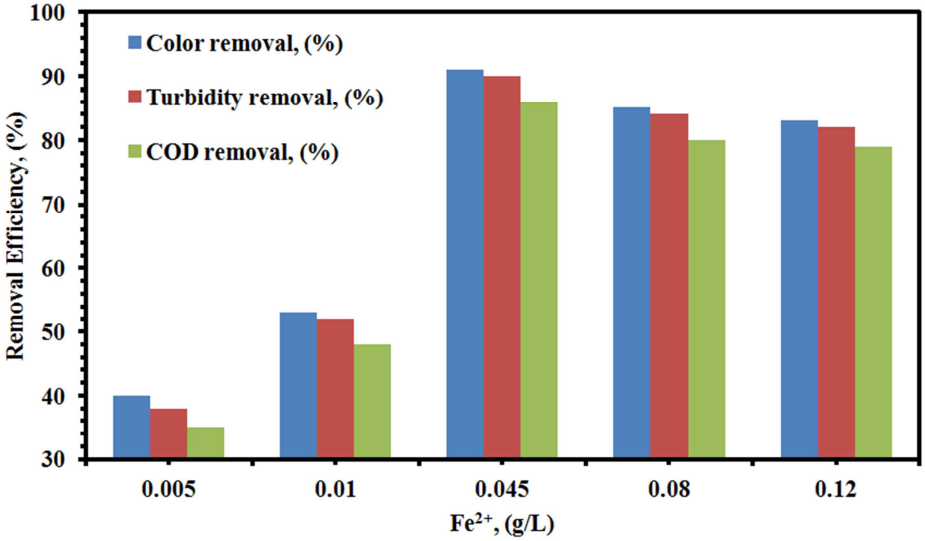

Figure 6. *Effect of Fe^{2+} on removal efficiency of % color, turbidity, and COD.*

3.2.4. Effect of pH

Solution pH is an important operating parameter that can influence the solar-UV/Fe^{2+}/H_2O_2 process. The solar UV/Fe^{2+}/H_2O_2 reaction was strongly pH-dependent. The pH value influences the generation of •OH radicals and the removal efficiency. To study the effects of effluent pH on the treatment efficiency, the initial pH of the wastewater was adjusted to between 2 and 10. The removal efficiency color, turbidity, and COD as a function of initial pH were presented in Figure 7. When the initial pH was changed from 2 to 4, the % color, % turbidity, and % COD removal efficiencies increased. However, when the initial pH was changed from 4 to 10, the removal efficiencies decreased.

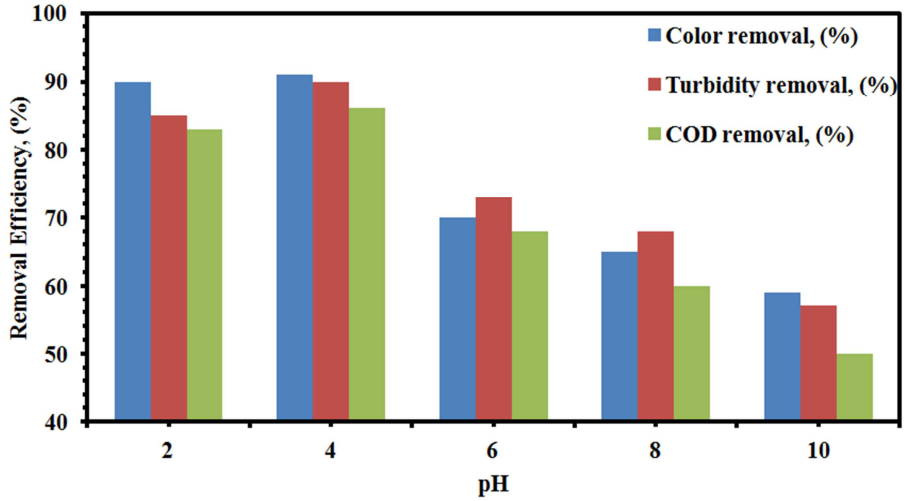

Figure 7. *Effect of pH on removal efficiency of % color, turbidity, and COD.*

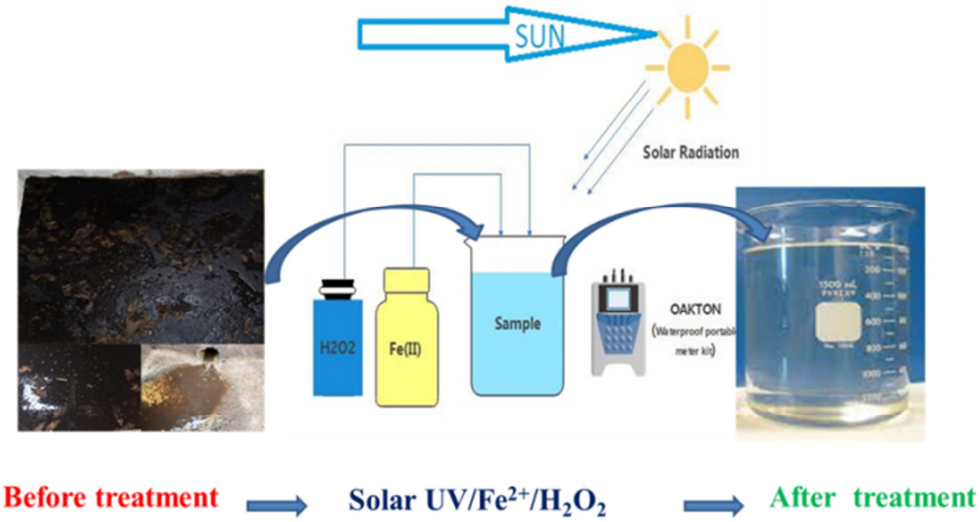

Figure 8. *Color removal before and after treatment of wastewater.*

The optimum pH was found to be about 4, it can be seen that the removal rate of color, turbidity, and COD was increased with the pH in the range of 2 to 4 and decreased from 4 to 10. The degradation decreased at pH values higher than 4 because iron precipitated as hydroxide. Additionally, the oxidation potential of the •OH radical was known to decrease with increasing pH. Another reason for the inefficient degradation at pH>4 is the dissociation and auto-decomposition of H_2O_2. For pH values below 2.5, the reaction of H_2O_2 with Fe^{2+} is seriously affected causing the reduction in •OH radical production, due to •OH radical scavenging by H^+ ions.

Figure 8, shows the color removal of wastewater before and after treatment using the solar-UV/Fe^{2+}/H_2O_2 process. From Figure 8, it is observed that complete color removal is achieved within the process of time.

4. Conclusions

From this research study, the following conclusions were drawn. The solar-UV/Fe^{2+}/H_2O_2 process for the treatment of institution wastewater was successfully carried out in terms of % color, % turbidity, and % COD removal. The results showed that the solar-UV/Fe^{2+}/H_2O_2 process was more

effective than the listed other AOPs. The effects of various operating parameters such as H_2O_2, Fe^{2+}, initial effluent pH, and reaction time by the solar-UV/Fe^{2+}/H_2O_2 process were studied using institution wastewater. The solar-UV/Fe^{2+}/H_2O_2 process was optimized to maximize pollutant removal of institutional wastewater. The maximum color, turbidity, and COD removal of 91%, 90%, and 86%, respectively were observed at the H_2O_2 of 0.75 g/L, initial effluent pH of 4, and Fe^{2+} of 0.045 g/L, and reaction time of 120 min. The results of this study indicated that the solar UV/Fe^{2+}/H_2O_2 was a practical method to treat institutional wastewaters and allowing achieved higher removal of color, turbidity, and COD. The solar UV/Fe^{2+}/H_2O_2 process described in this study was suitable to be applied for the treatment of institutional wastewater as it has higher treatment efficiency.

Declarations

Data Availability Statement

Data will be made available on request.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

Research Highlights

A combination of solar UV, Fe^{2+}, and H_2O_2 for wastewater treatment developed.

Solar-UV/Fe^{2+}/H_2O_2 highly successful for removal of pollutant than other processes.

Maximum pollutant efficiencies were obtained using solar UV/Fe^{2+}/H_2O_2.

Operating parameters studied to obtain the process performance efficiency.

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