



Assessment of a Fenton Reaction in treating Greywater for reuse in Irrigation

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Abstract

Greywater is considered a new water resource because of water scarcity all over the world. Thus collecting, treating, and reusing it safely is critical for protecting the environment from any hazard. As a common application of greywater reuse, irrigation includes specific characteristics of the water that need to adapt a grey wastewater treatment technology.

In this paper, an advanced oxidation process based on Fenton's reaction was used to evaluate the greywater treatment efficiency by testing specific parameters before and after oxidization and determining the optimum chemical ratio between iron salts (ferrous sulfate) and Hydrogen peroxide (Fe^{2+}/H_2O_2) and the optimum retention time to reach the permissible limits according to the ECP, 501-2005 (Egyptian Code No. 501 of the year 2005).

An experimental model including three glass tanks with dimensions of 30×30×50 cm was established. The model consisted of a primary sedimentation tank to allow total suspended solids to settle, followed by a tank for chemical treatment with Fenton's reagent where iron salts of 0.3 gm per liter and hydrogen peroxide with four different doses were added representing chemical ratios between Fe^{2+}/H_2O_2 of 1:3, 1:4, 1:5, and 1:6 to determine the highest efficiency of the reaction, then followed by the collecting tank.

From the obtained results, it was concluded that the optimum chemical ratio of Fe^{2+}/H_2O_2 is 1:5 for a retention time of 30 minutes to achieve the permissible limits of the treated greywater to be reused in irrigation according to the ECP.

Keywords : Chemical oxidation, Fenton's reagent, Hydrogen peroxide, COD, BOD, TSS, TDS>

1. Introduction

Greywater treatment and reuse are emerging as sustainable concepts to reduce overall water demand. Irrigation is a common application of greywater reuse after treatment. Greywater represents 50–75%

of household water consumption [1], and agriculture accounts for more than 85% of the total water consumption.[2] As the population grows and agriculture expands, water resources have been depleted and wastewater has increased. In this context, greywater is defined as any domestic wastewater generated in households and offices, excluding sewage and kitchen wastewater, which are

classified as black water due to their high concentrations of organic matter that encourage the growth of bacteria. Organic loading is the primary distinction between greywater and sewage.

Common sources of greywater in the home include showers, baths, sinks, and clothes washers [3,4,5,6] If greywater is not properly treated, it can harm the environment and human health. Reusing the treated effluent for agriculture or industrial recycling protects public health and the environment.[7] Treatment methods to remove contaminants from wastewater are classified as physical, chemical, and biological methods.

Chemical treatment methods, particularly oxidation, are used to achieve high levels of contaminant removal and treatment of high concentrations of organic compounds. Chemical oxidation wastewater treatment methods are referred to as Advanced Oxidation Processes (AOPs), which involve the production of some highly reactive and powerful oxidants. AOPs generally means the application of either advanced oxidation technologies using UV (Ultraviolet light)/O₃ (Ozone), O₃/H₂O₂ (Hydrogen peroxide), UV/H₂O₂, photo-Fenton reaction [UV or solar irradiation/H₂O₂/Fe²⁺ (Ferrous ion) or Fe³⁺ (Ferric ion)], Sono-Fenton [US (ultrasound)/ H₂O₂/ Fe²⁺ or Fe³⁺] or the Electro-Fenton reaction [8,9] AOPs were applied by [10] as an effective method consisting of highly reactive species used in the oxidative destruction of target pollutants to the preferred end products of carbon dioxide and water.

Among AOPs, Fenton's Reaction was discovered by.[11] The COD (Chemical Oxygen Demand) is a reference parameter when using chemical oxidation as a treatment process. Only waters with low COD levels (0.5 g/L) can be treated effectively using this method. The oxidation of organic substrates by iron (II) and hydrogen peroxide (H₂O₂) is known as "Fenton chemistry" after H.J.H. Fenton, who first observed tartaric acid oxidation by H₂O₂ in the presence of ferrous ions. [12] Alternatively, the terms "Fenton reaction" or "Fenton reagent" are frequently used. It is well known that the Fenton reagent is a solution of hydrogen peroxide (H₂O₂) with ferrous ions (typically Fe²⁺ sulphate, FeSO₄) that catalyzes the decomposition of H₂O₂ leading to the generation of free hydroxyl radicals that are used to oxidize contaminants and destroy organic compounds in wastes [13].

Hydrogen peroxide is one of the most powerful and effective oxidizers and is even more strong than chlorine and chlorine dioxide. Through catalysis, H₂O₂ can be converted into hydroxyl radicals (.OH)

which attack and degrade strongly the organic matter in grey wastewater.

The Fenton reagent was not used as an oxidizing process for destroying hazardous organics until the late 1960s.[14,15] Comprehensive studies showed that the Fenton reagent is effective in treating a wide range of industrial wastewater components including aromatic amines[16], dyes[13], pesticides [17,18], and many others. Also, the Fenton reagent has been used to treat a wide range of wastes including those from the textile industry and chemical substances manufacturing.[19]

The application of the Fenton reaction in the decomposition of phenol and formaldehyde from their aqueous solutions and industrial wastewater was studied achieving a very good removal efficiency of over 90% [20] The removal of color and COD from a mixture of four reactive azo dyes using the Fenton oxidation process was investigated which achieved high efficiency of more than 90%.[21]

The application of Fenton's reaction in paper industrial wastewater treatment was investigated achieving Total Suspended Solids (TSS), Biological Oxygen Demand (BOD), COD, Total Nitrogen (TKN), and Total Phosphorous (TP) removal efficiency up to 95.4%, 92.7%, 93.7%, 90.5%, and 91.7%, respectively. It was proved that the Fenton treatment was effective leading to reusing paper industrial wastewater for irrigation purposes according to the Egyptian code.[22] .[23] applied Fenton's reagent and adsorption techniques using Starbon (mesoporous material derived from polysaccharides such as starch) as an alternative to already conventional studied treatments of greywater such as filtration, sedimentation, physical separation, and/or biological processes. The main objective was to treat the waste laundry water from contaminants and to provide an alternative water supply for many applications except for drinking water in York, United Kingdom. It was concluded that experiments with Fenton's reagent at optimum conditions of Fe³ = 40 mg/L, H₂O₂ = 400 mg/L, and Ph = 3 achieved a 95% COD removal after 15 minutes. Starbon adsorption and the combined treatment with Fenton's reagent and Starbon were also effective in removing up to 81% and 93% of COD, respectively.

[24] investigated the use of the Fenton process to treat greywater of toilet flushings in Dhaka, Bangladesh. Four approaches with different doses of Fenton's Reagent and times of chemical treatment were conducted. From the obtained results, the optimum doses were 1.13 g/L for Fenton's reagent (0.5 g/L for H₂O₂ and 0.63 g/L for FeSO₄). The time of chemical treatment was 30 minutes. In Egypt,

research has been set by [25] to treat and reuse greywater through the use of chemical oxidation of Fenton's reaction. TSS, COD, BOD₅, and oil & grease were the parameters measured using standard methods. It was concluded that the Fenton technology was effective as it achieved TSS, COD, BOD, and oil & grease removal efficiency up to 81%, 90%, 91%, and 75%, respectively. [26] applied different Fenton technologies to greywater to evaluate the possibility of using three AOPs for the removal of organic pollutants present in grey wastewater.

This current study aims to evaluate the efficiency and increase the effectiveness of the greywater treatment by employing a chemical treatment. The chemical treatment includes Fenton's reaction as advanced oxidation to reduce the physicochemical characteristics to acceptable limits for irrigation set by. [27]

2. MATERIALS AND METHODS

Domestic raw greywater samples were collected and transported to the Egyptian Foundation for Scientific Services and Analysis of Water in Cairo for immediate analyses to prevent spontaneous chemical reactions and microbial activity. The measured parameters were COD, BOD, TSS, Total Dissolved Solids (TDS), TKN, TP, and pH. These parameters were measured according to standard methods and the physical and chemical characteristics of raw greywater are listed in Table 1.

The materials used in this study for the Fenton reaction were Ferrous sulfates ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), Hydrogen peroxide solution (H_2O_2 , 50% w/w), and Hydrochloric Acid (HCL), and Sodium Hydroxide (NaOH). Both HCL and NaOH were used to adjust the pH to the desired values.

In this experimental study, a miniature model was used, which consisted of three glass rectangular tanks, as illustrated in Table 2 and Fig. 1.

TABLE 1. Physico-chemical characteristics of raw greywater

Parameters	Unit	Raw 1	Raw 2	Raw 3	Raw 4
Chemical oxygen demand, COD	mg/L	510	501	498	453
Biological oxygen demand, BOD	mg/L	180	175	193	167
Total suspended solids, TSS	mg/L	165	169	170	146
Total dissolved solids, TDS	mg/L	985	981	996	946
Total nitrogen, TKN	mg/L	10.5	15.6	16.8	18.6
Total phosphorous, TP	mg/L	8.0	7.8	8.7	7.0
pH	---	7.9	7.8	8.0	7.8

TABLE 2. The dimensions and the operational parameters of the model

Treatment unit	Parameters	Values
Primary sedimentation tank	Length × width × depth	50 × 30 × 30 cm
	Volume of greywater	40 liters
	Hydraulic retention time	1 hr.
Fenton tank	Length × width × depth	50 × 30 × 30 cm
	Volume of greywater	40 liters
	Dose of iron salt (Fe^{+2})	0.3 g/L
	Doses of Hydrogen peroxide (H_2O_2)	0.9, 1.2, 1.5, and 1.8 g/L
	Hydraulic retention time	10, 15, and 30 min.
Collecting tank	Length × width × depth	50 × 30 × 30 cm

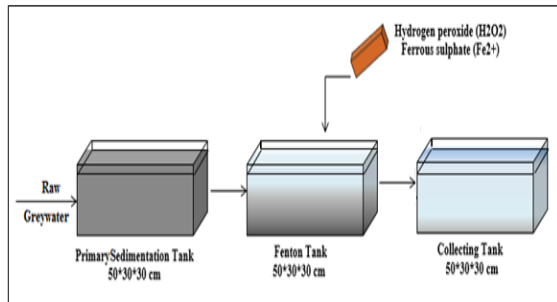


FIGURE 1. Schematic diagram of the treatment system

The first tank was a primary sedimentation tank to allow total suspended solids to settle, followed by a Fenton tank to perform the chemical oxidation process and followed finally by a collecting tank. The study was based on four scenarios including a constant dose of iron sulfates, different H₂O₂ doses, and various reaction times, as shown in Fig. 2.

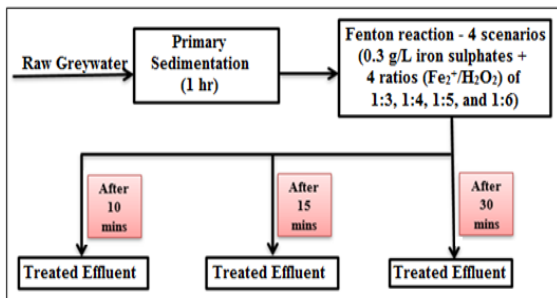


FIGURE 2. A diagram of the applied scenarios

First, grey wastewater was collected in the primary sedimentation tank, and samples were taken from it to investigate the characteristics of untreated greywater.

Then the greywater was left in the primary sedimentation tank for one hour to allow the total suspended solids to settle under gravity without the addition of any chemical additives and samples were collected.

Following that, the greywater was transferred to the Fenton tank, where iron salts (ferrous sulphate) of 0.3 g/L and hydrogen peroxide with different doses were added according to four scenarios to determine the highest efficiency of the reaction. The employed doses of hydrogen peroxide were 0.9, 1.2, 1.5, and 1.8 g/L representing chemical ratios of 1:3, 1:4, 1:5, and 1:6. Samples were collected at 10, 15, and 30 minutes.

3. RESULTS AND DISCUSSION

To use treated greywater instead of fresh water for irrigation purposes, a series of experiments were carried out with a constant dose of iron salts, various hydrogen peroxide concentrations, and different retention times. That is to investigate the optimum chemical ratio ($\text{Fe}^{2+}/\text{H}_2\text{O}_2$) and the optimum retention time for COD, BOD, TSS, TDS, TKN, and TP removal and to achieve the required specifications for irrigation purposes according to the ECP, 501-2005.

Table 3 shows the concentrations of all experimental scenarios of greywater treatment by Fenton reaction. All the results were compared with the Egyptian code No. 501, 2005 for reusing the treated wastewater in irrigation.

3.1 EFFECT OF FENTON REACTION ON COD

Figure 3 shows the relation between the COD concentration for different chemical ratios and retention times of 10, 15, and 30 minutes. The results showed a significant enhancement of the degradation process for each chemical ratio when the treatment time was increased. According to the permissible concentration of COD, it was concluded that this concentration was achieved only for the chemical ratio of 1:5 after a retention time of less than 30 min (starting from 18 min). Regression analyses were applied and polynomial equations were obtained to predict the concentration of COD at any treatment time.

3.2. EFFECT OF FENTON REACTION ON BOD

The relation between the concentration of BOD for different chemical ratios between Fe^{2+} and H_2O_2 versus retention time is illustrated in Fig. 4. From the obtained results, it was found that the longer the retention time, the lower the BOD concentration. According to the permissible concentration of BOD, it was concluded that this concentration was achieved only for the chemical ratio of 1:5 after a retention time of less than 30 min (starting from 28 min). Regression analyses were applied and polynomial equations were obtained to predict the concentration of BOD at any treatment time.

TABLE 3. Physico-chemical characteristics of greywater and Egyptian Code

Scenario	Parameters	COD	BOD	TSS	TDS	TKN	TP	pH
	Units	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	---
1	Raw	453.0	167.0	146.0	946.0	18.6	7.0	7.8
	P.S (1 hr.)	384.0	107.0	85.9	922.0	15.5	5.6	7.5
	Fenton tank (10 min)	198.0	72.0	50.0	718.9	10.2	4.5	4.0
	Fenton tank (15 min)	119.8	44.3	41.2	510.0	9.2	3.3	3.5
	Fenton tank (30 min)	77.9	35.1	30.0	499.0	8.2	2.9	3.4
2	Raw	510.0	180.0	165.0	985.0	10.5	8.0	7.9
	P.S (1 hr)	176.0	113.0	50.0	720.9	8.1	4.6	7.6
	Fenton tank (10 min)	100.5	84.0	34.8	617.0	6.0	3.2	4.6
	Fenton tank (15 min)	86.5	64.3	22.9	560.0	4.1	2.0	3.5
	Fenton tank (30 min)	51.0	27.0	16.5	423.6	3.2	1.3	3.2
3	Raw	501.0	175.0	169.0	981.0	15.6	7.8	7.8
	P.S (1 hr)	446.0	120.0	141.0	965.0	11.5	6.5	7.6
	Fenton tank (10 min)	138.0	86.0	48.0	578.0	7.0	1.6	4.5
	Fenton tank (15 min)	89.9	43.0	17.1	450.9	4.2	1.2	3.8
	Fenton tank (30 min)	20.1	18.2	7.8	304.1	3.2	0.7	3.1
4	Raw	498.0	193.0	170.0	996.0	16.8	8.7	8.0
	P.S (1 hr)	415.0	151.0	141.0	973.0	14.0	6.5	7.5
	Fenton tank (10 min)	182.0	82.0	128.0	749.0	9.8	4.3	4.6
	Fenton tank (15 min)	90.7	53.1	41.9	685.0	7.1	2.1	4.0
	Fenton tank (30 min)	74.7	36.7	26.0	547.8	6.7	1.2	3.9
ECP, 501 - 2005		40.0	20.0	20.0	2000.0

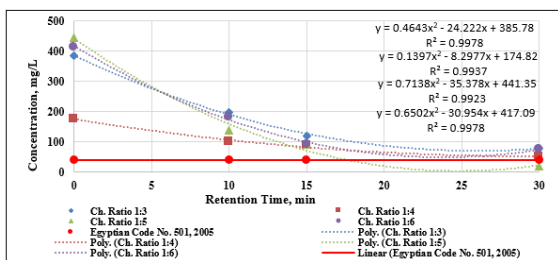


FIGURE 3. COD concentration for different chemical ratios and retention times

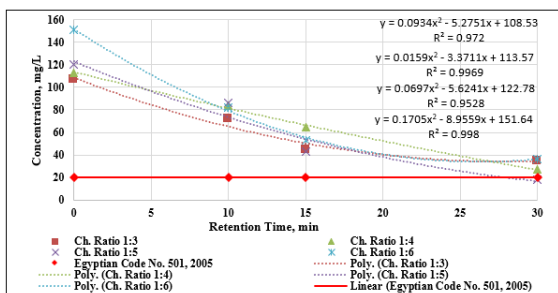


FIGURE 4. BOD concentration for different chemical ratios and retention times

3.3. EFFECT OF FENTON REACTION ON TSS

Figure 5 shows the relation between the concentration of TSS for different chemical ratios

(Fe²⁺/H₂O₂) and retention times. It was found that increasing treatment time decreased the TSS concentration. According to the permissible concentration of TSS, it was concluded that this concentration was achieved for the chemical ratios of 1:4 and 1:5 after retention times less than 30 min (starting from 21 and 15 min for chemical ratios of 1:4 and 1:5, respectively). Regression analyses were applied and polynomial equations were obtained to predict the concentration of TSS at any treatment time.

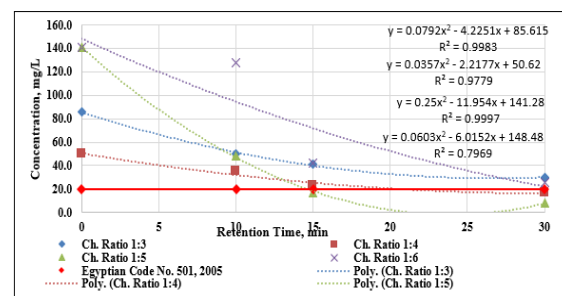


FIGURE 5. TSS concentration for different chemical ratios and retention times

3.4. Effect of Fenton reaction on TDS

The effect of the Fenton process on TDS concentration in greywater is shown in Fig. 6 for

different chemical ratios and retention times. Because the permissible concentration of TDS is much higher than the concentrations found in greywater, it was concluded that all chemical ratios of 1:3, 1:4, 1:5, and 1:6 were accepted overall treatment retention times. Regression analyses were applied and polynomial equations were obtained to predict the concentration of TDS at any treatment time.

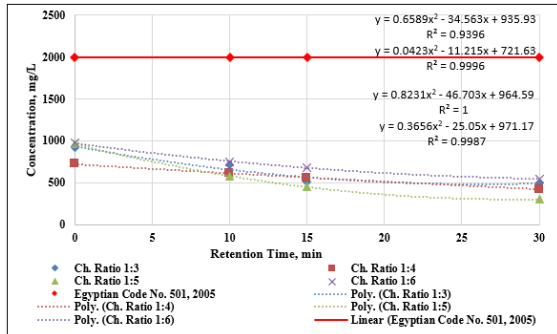


FIGURE 6. TDS concentration for different chemical ratios and retention times

3.5. Effect of Fenton reaction on TKN

The effect of the Fenton process on TKN concentration in greywater is shown in Fig. 7 for different chemical ratios and retention times. The concentrations of TKN decreased with increasing the treatment time. Because there is no permissible concentration of TKN, it was concluded that all chemical ratios of 1:3, 1:4, 1:5, and 1:6 were accepted for overall treatment retention times.

Regression analyses were applied and polynomial equations were obtained to predict the concentration of TKN at any treatment time.

Figure 8 indicates percentages of removal efficiency of TKN for all chemical ratios and retention times, where the removal efficiency of TKN increased with increasing the treatment time.

The chemical ratio of 1:5 achieved the best removal efficiency at a retention time of 30 minutes.

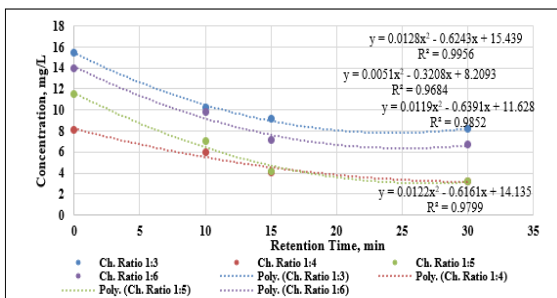


FIGURE 7. TKN concentration for different chemical ratios and retention times

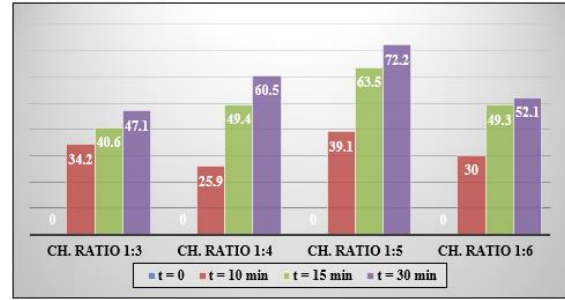


FIGURE 8. TKN removal efficiency for different chemical ratios and retention times

3.6. Effect of Fenton reaction on TP

The relations between the concentration of TP were studied versus retention time at different chemical ratios, as shown in Fig. 9. The concentrations of TP decreased with increasing the treatment time. Because there is no permissible concentration of TP, it was concluded that all chemical ratios of 1:3, 1:4, 1:5, and 1:6 were accepted for overall treatment retention times.

Regression analyses were applied and polynomial equations were obtained to predict the concentration of TP at any treatment time.

Figure 10 illustrates percentages of removal efficiency of TP for all chemical ratios and retention times, where the removal efficiency of TP increased with increasing the treatment time.

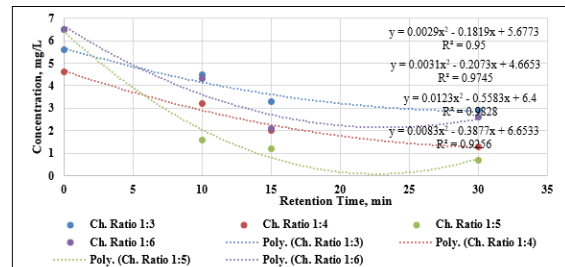


FIGURE 9. TP concentration for different chemical ratios and retention times

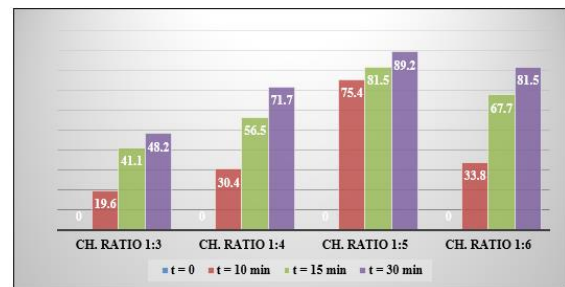


FIGURE 10. TP removal efficiency for different chemical ratios and retention times

The chemical ratio of 1:5 achieved the best removal efficiency at a retention time of 30 minutes.

From the obtained results, it was found that chemical oxidation (Fenton reaction) successfully achieved the goals of the proposed treatment process. The required permissible concentration of COD was achieved only for the chemical ratio of 1:5 after a retention time of less than 30 min (starting from 18 min). The permissible concentration of BOD was achieved only for the chemical ratio of 1:5 after a retention time of less than 30 min (starting from 28 min). The permissible concentration of TSS was achieved for the chemical ratios of 1:4 and 1:5 after retention times less than 30 min (starting from 21 and 15 min for chemical ratios of 1:4 and 1:5, respectively). The permissible concentration of TDS is higher than the concentrations found in raw greywater, so all chemical ratios of 1:3, 1:4, 1:5, and 1:6 were accepted overall treatment retention times. There is no permissible concentrations of TKN and TP, so all chemical ratios of 1:3, 1:4, 1:5, and 1:6 were accepted for overall treatment retention times.

Coping to the Egyptian code No. 501, 2005 for reusing the treated wastewater in irrigation, the optimum chemical ratio of iron salt / Hydrogen peroxide is 1:5 for a retention time of 30 minutes.

4. CONCLUSIONS

Greywater reuse is a potential option for water demand management that can help to reduce freshwater consumption for irrigation, especially in remote areas.

It has been concluded that chemical oxidation (Fenton reaction) could successfully achieve the goals of the proposed treatment process.

The parameters values determined in this study revealed that greywater characteristics were varied due to the different uses of family members and product types used.

From the obtained results concerning using the chemical ratio of 1:3, it was concluded that it did not achieve the permissible concentrations of COD, BOD, and TSS. While it was applicable for TDS (very high permissible concentration) and both TKN and TP (no required permissible concentrations). The best TKN and TP removal efficiency was achieved at a retention time of 30 minutes.

From the obtained results concerning using the chemical ratio of 1:4, it was concluded that it did not achieve the permissible concentrations of COD and BOD. It achieved the permissible concentrations of TSS after retention times of less than 30 min (starting from 21 min). While it was applicable for TDS (very

high permissible concentration) and both TKN and TP (no required permissible concentrations). The best TKN and TP removal efficiency was achieved at a retention time of 30 minutes.

From the obtained results concerning using the chemical ratio of 1:5, it was concluded that it achieved the permissible concentrations of COD, BOD, and TSS after retention times of less than 30 min (starting from 18 min for COD, 28 min for BOD, and 15 min for TSS). While it was applicable for TDS (very high permissible concentration) and both TKN and TP (no required permissible concentrations). The best TKN and TP removal efficiency was achieved at a retention time of 30 minutes.

From the obtained results concerning using the chemical ratio of 1:6, it was concluded that it did not achieve the permissible concentrations of COD, BOD, and TSS. While it was applicable for TDS (very high permissible concentration) and both TKN and TP (no required permissible concentrations). The best TKN and TP removal efficiency was achieved at a retention time of 30 minutes.

It was concluded that the optimum chemical ratio of iron salt / Hydrogen peroxide is 1:5 for a retention time of 30 minutes, which cope with the Egyptian code No. 501 - 2005 for reusing the treated wastewater in irrigation.

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