



## Treatment of mixed domestic–industrial wastewater by combined coagulation–sedimentation, biological processes and advanced oxidation

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Received 28 April 2022; Accepted 28 November 2022

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### ABSTRACT

Nowadays, the treatment of mixed domestic–industrial wastewater has gained more attention, so, the main target of the study is to degrade the wastewater through a combination of primary, biological, and advanced oxidation processes. To enhance the treatment of the wastewater, the processes of coagulation, flocculation and sedimentation were applied before the biological treatment. The effect of the type and dose coagulants and effect of using polyacrylamide (PAM) as a coagulant aid were studied. The optimized conditions selected for the experiments were ferric chloride and alum. They showed a close treatment effectiveness since 400 mg/L was the best coagulant dose, and 10 mg/L was the best PAM dose. The chemical oxygen demand (COD), biological oxygen demand (BOD<sub>5</sub>), and total suspended solids (TSS) removal efficiencies were 58.8%, 55.9% and 67.5%, respectively at 10 mg/L PAM and 400 mg/L alum doses. When doses of ferric chloride and PAM were at 400 and 10 mg/L sequentially, the removal efficiency of COD, BOD<sub>5</sub> and TSS were 61.8%, 59.7% and 71.1%, respectively. After the primary treatment, extended aeration was used for the biological wastewater treatment. The study aimed to evaluate the effect of hydraulic retention time (HRT) on the performance of the biological treatment via extended aeration. The removal rates at HRT of 24 h for COD, BOD<sub>5</sub>, TSS and total Kjeldahl nitrogen (TKN) were 70.6%, 86.2%, 58% and 70.7%, respectively. Increasing the HRT more than 24 h exerted a slight improvement in the pollutant removal efficiency. After the biological treatment, COD, BOD<sub>5</sub>, TSS and TKN in the effluent decreased, but the values did not meet the Egyptian standards. A Fenton process was provided in the treatment to ensure that the COD removal as stipulated by the regulations would be obtained in the final effluent. Different reaction conditions of Fenton treatment were examined, including H<sub>2</sub>O<sub>2</sub> concentration and H<sub>2</sub>O<sub>2</sub>/Fe<sup>2+</sup> ratios. The maximum removal was achieved at H<sub>2</sub>O<sub>2</sub>/COD = 4.7. In addition, five different H<sub>2</sub>O<sub>2</sub>/Fe<sup>2+</sup> molar ratios (20–160) were tested. COD removal percentage was 68.4% at the molar ratio of 160 and increased to 84.9% at the molar ratio of 40.

*Keywords:* Industrial effluents; Combined wastewater treatment; Water quality; Chemical oxidation; Biological treatment; Hydrogen peroxide; Fenton

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### 1. Introduction

To counter the continued population growth of major cities, the Egyptian government has implemented new cities to redistribute the country's population away from the banks of the Nile River into new desert cities [1]. Most of these cities are planned as industrial ones with a myriad

of factories. High-quality water is fundamental for most industries. Many of these industries produce huge effluents of contaminated wastewater and the safe disposal of it into receiving water bodies is a major environmental and health concern [2], since contamination of surface and groundwater by industrial effluents causes water shortage in industrialized cities [3]. Although it is mandatory for

factories to treat their wastewater before dumping it into the sewerage system, this wastewater is often discharged without meeting environmental requirements and usually mixes with municipal wastewater. Most of these cities' treatment plants are technically supposed to deal with municipal wastewater and pre-treated industrial wastewater, so the direct discharge of industrial effluents to the treatment plants results in treated wastewater rarely with dissatisfying regulatory standards and requirements. This increases effluent concentrations of chemical oxygen demand (COD), biological oxygen demand (BOD<sub>5</sub>), ammonia (NH<sub>3</sub>), and total suspended solids (TSS), indicating the poor level of treatment efficiency due to the high influent organic load resulting from industrial wastewater. The Egyptian Ministry of Water Resources and Irrigation (MWRI) has issued Law 48 (1982) in order to protect the waterways from pollution. The standards imposed by Law 48 are given in Table 1.

To overcome these problems, the idea of this research is to apply a combination of physical, chemical and biological processes for the treatment of mixed domestic-industrial wastewater. The mixture contains a considerable amount of non-biodegradable compounds, so physical and biological treatment only is not a feasible option.

The conceptual design for the liquid treatment process consists of influent equalization, coarse and fine screening, grit removal, neutralization, coagulation and flocculation and clarification, biological treatment and clarification, advanced oxidation process, filtration, disinfection, effluent storage and discharge.

The coagulation/flocculation process is considered one of the most important and widely used wastewater treatment techniques [4,5]. This is because it has been considered an effective and low-cost pre-treatment method [6]. The present study aimed to determine the physical and chemical parameters in flocculation and coagulation processes, using different coagulants and doses along with a polymer as a coagulant aid. In addition, the study assesses the feasibility of these processes in industrial wastewater treatment by making a performance comparison.

Although the moving bed biofilm reactor (MBBR) or the activated sludge process (BOD, nitrification, denitrification) can be used in secondary treatment, the activated sludge process is used in the current study because the MBBR effluent has high NO<sub>3</sub>-N [7].

It is anticipated that the mixed wastewater can contain high levels of COD from industries. These incoming loads of COD may not be fully removed by the primary and biological treatment and may require an advanced oxidation process (AOP) to remove the excess COD and to ensure that the limits of discharge are met. The various advanced oxidation process options of wastewater application include

Fenton's reaction, ozone, ultraviolet (UV), UV and hydrogen peroxide as well as UV and ozone.

Ozone is a powerful disinfectant, but some considerations have to be taken: (1) unless combined with UV or a catalyst, ozone is ineffective, (2) ozone generation devices are complicated and will necessitate oxygen supply for large capacities [8], (3) ozone is hazardous, and (4) the plant is capital intensive as well as has significant operation and maintenance (O&M) costs due to power and oxygen consumption. UV is easy to operate with the necessary level of automation, but (1) UV radiation alone does not have any applicability in the treatment of effluents [9] and needs addition of oxidizing agents or a catalyst like titanium dioxide etc., (2) UV increases the O&M cost due to power, bulb replacements and cleaning, (3) UV effectiveness is not fully established in large scale treatment facilities, and (4) efficiency of UV is affected by the presence of particles in the waste stream [10]. Photolysis oxidation by UV can be achieved in the presence of hydrogen peroxide, but some points have to be noticed: (1) a large quantity of hydrogen peroxide is needed, (2) it increases the O&M cost due to power, bulb replacements and cleaning, and (3) efficiency is affected by the presence of particles in the waste stream. Ozone-based AOPs (like ozonation in combination with UV) are not being considered for the aforementioned reasons.

Although Fenton requires a high chemical consumption and generates additional sludge, hydrogen peroxide releases hydroxyl ions, in the presence of ferrous salt, which act as a powerful oxidizing agent.

The Fenton process has been widely used to treat industrial wastewaters [11–15], therefore, to ensure proper removal of the COD, Fenton was considered the best option to be used in the current study.

On the basis of the foregoing, the treatment processes that have been considered in the current study include a preliminary treatment (screening, grit removal), primary treatment (coagulation, flocculation, settling), a biological treatment (the activated sludge process, settling) then (Fenton's section, settling). This research is a laboratory study aiming to investigate the potential of the integration of the coagulation/flocculation/settling, biological treatment (extended aeration), and AOP (Fenton section), with the purpose of treating mixed domestic industrial wastewater.

## 2. Material and methods

### 2.1. Description of the study site

Badr City is planned as an industrial region with a myriad of factories. The total area of the city is 18,500 acres, including 7,708 acres of housing areas and 2,200 acres of industrial areas in addition to service and green areas [16]. The city has an oxidation pond treatment plant and it is supposed to treat the municipal wastes as well as the pre-treated industrial wastewater. The existing treatment plant receives approximately 47,000 m<sup>3</sup>/d and 15,000 m<sup>3</sup>/d average daily industrial and domestic wastewater flow, respectively. The industrial area in Badr city includes chemical, pharmaceutical, wood, electrical, plastics, building materials, paper, and textiles industries. Since industrial wastewater is not pre-treated well, the effluent discharge concentrations do not meet regulatory requirements.

Table 1  
Effluent standards according to Law 48/1982

Parameter	Value
BOD <sub>5</sub>	≤60 mg/L
COD	≤80 mg/L
TSS	≤50 mg/L

Mixed domestic–industrial wastewater was collected from the inlet of the treatment plant. Composite wastewater samples were aerated for 4 min to remove oil and grease from wastewater. Then, the samples were stored in the refrigerator at 4°C to avoid further biodegradation. Conventional analyses such as pH, temperature, COD, BOD<sub>5</sub>, TSS and total Kjeldahl nitrogen (TKN) were measured according to the Standard Methods [17]. A summary of the characteristics of the wastewater obtained from this study is given in Table 2.

## 2.2. Physico-chemical experiments

The experiments were carried out using three stages. The first stage of treatment was the particle separation. This stage consists of coagulation followed by flocculation and sedimentation. The purpose of this stage was to remove the suspended solids. The removal of suspended solids would also result in the reduction of COD and BOD<sub>5</sub>. Chemical coagulation was carried out, using the Jar Test procedure to determine the optimum conditions, that is, the type and dose of the coagulant. The experiments were performed, using aluminum sulfate (alum), ferric chloride (FeCl<sub>3</sub>·6H<sub>2</sub>O), and zinc sulfate (ZnSO<sub>4</sub>·7H<sub>2</sub>O). All chemical dosages were expressed as mg/L of the coagulant (as alum, ferric chloride or zinc sulfate).

Sriwiriyarat and Jangkorn [18] have suggested the utilization of the waste activated sludge (WAS) as a coagulation aid in the coagulation–flocculation process for wastewater treatment. One of the objectives of the current study was to evaluate the feasibility of using WAS as a coagulant. The WAS was selected because of its adsorption capability and its availability from the wastage stream of biological wastewater treatment process [18]. The WAS used in this study was collected from the aeration tank of a pilot-scale activated sludge system, which is a biological wastewater treatment process, running in the Al-Qenayat Wastewater Treatment Plant (WWTP) in Zagazig, Sharkia, Egypt. The used activated sludge was aerated for 24 h to remove the residual organic compounds at approximately room temperature (25°C–30°C). Then, the air supply was shut off and the sludge allowed to settle for approximately 30 min, the supernatant was then siphoned off. The quantified volume of the thickened sludge was added to the tested wastewater to achieve the desired concentration. The average MLSS concentration for the thickened sludge was approximately 4,900 mg/L.

Table 2  
Wastewater characteristics

Parameter	COD (mg/L)	BOD <sub>5</sub> (mg/L)	TKN (mg/L)	TSS (mg/L)
Influent wastewater	1,950–2,200	790–875	48–53	894–940
Primary treated wastewater	942–1,035	468–540	42–48	360–412
Biologically treated wastewater	340–388	91–113	13–17	162–183

The recommended pH range for efficient coagulation varies depending on the coagulants used, but is usually between 6.0 to 8.5 [19]. Most chemical-based coagulants absorb a high percentage of alkalinity. Thus, adding a coagulant to wastewater with low alkalinity produces poor flocs [20]. Since the pH range for raw wastewater was 7.3–8.9, so the Jar Test experiments were conducted with a pH of 7.5 to avoid the pH correction of wastewater.

In the Jar Test, the wastewater sample was transferred to each Jar. Different coagulants with various doses were added to each Jar containing a 800 mL wastewater sample to determine both the best coagulant and dose. The samples were rapidly mixed at 160 rpm for 2 min and coagulants were added, followed by a slow stirring period at 60 rpm for 20 min. Finally, the mixture was allowed to settle for 60 min. The supernatant was withdrawn from the beakers and was taken to be analyzed (COD, BOD<sub>5</sub>, and TSS). All experiments were repeated three times, and the average value was taken.

## 2.3. Effect of polymer dosage

Polyelectrolytes can be utilized as coagulant aids in the treatment of industrial wastewater and they also may be used as a primary coagulant for the same purpose [21,22]. Polyelectrolytes, such as polyacrylamides, chitosan, polyvinyl and others, are usually used as coagulant aids to enhance the formation of larger floc and, therefore, the rate of sedimentation [23]. In the present research, anionic polyacrylamide (PAM) was used as a coagulant aid. The dose of PAM was changed from 5 to 50 mg/L.

## 2.4. Biological study

After primary treatment, the clarified wastewater was subjected to the secondary treatment which mainly consists of a biological treatment followed by settling. The biological treatment that has been considered is the extended aeration. In the current study, removal efficiencies were investigated for different hydraulic retention times (HRTs). According to the literature studies on industrial wastewater that were examined and basis for the appropriate design of extended aeration systems, a 36-h HRT was selected as the beginning [24–26]. Then, this HRT value was decreased to 24, 12 h, 6 h, and 3 h.

For acclimatization, the seed sludge was brought from a pilot scale sewage treatment plant located in Al-Qenayat WWTP, Zagazig, Sharkia, Egypt. The activated sludge was aerated for 24 h to remove the residual organic compounds prior to the acclimation process. The quantified volume of the tested wastewater was added to the bioreactor to achieve the desired initial concentration. Acclimatization of the sludge was done in a batch mode slowly by taking out the percentage of the medium and adding the tested wastewater to the reactor.

The experiments performed in reactors have a working volume of 2.5 L. The oxygen was supplied through an air pump from the bottom of bioreactor and the oxygen concentration was kept at 3–5 mg/L. The reactor was operated at various HRTs. The reactor was operated at a batch mode condition at approximately room temperature

(25°C–30°C) and MLSS concentrations of approximately 2,500 mg/L.

The overall process involved the clarified wastewater after the primary treatment has been mixed with the sludge with intermittent aeration (simulating oxidation ditch). Several studies have shown this strategy leads to high treatment performance [27–29]. In the present study, the intermittent aeration schedule was 150/30 min aeration on/ off time. In aeration periods, COD removal and nitrification take place. The nitrification process involves the conversion of ammonia into nitrites and then to nitrates. During periods of aeration interruption, denitrification becomes predominant and denitrifies a major portion of the nitrates that are produced in the aerobic zone along with consuming some of COD in this process. Samples of MLSS were collected at different times to study the effect of HRT on the biological treatment. The samples were left for 60 min to settle and clear supernatants were subjected to analysis.

### 2.5. Tertiary treatment

After biological treatment, Fenton's reaction was used for the tertiary treatment of the clarified wastewater. The Fenton process is a viable alternative for the removal of organic pollutants from wastewater [8]. In the Fenton process, hydrogen peroxide ( $H_2O_2$ ) reacts with iron salts ( $Fe^{2+}$ ) to generate strong reactive species [30]. Ferrous sulfate heptahydrate ( $FeSO_4 \cdot 7H_2O$ ) was used, as a source of  $Fe^{2+}$  catalyst in the Fenton process and hydrogen peroxide solution was 30%, w/w.

In the current study, batch experiments were carried out at room temperature (25°C–30°C) to determine the optimum operating conditions of the reaction. The bench-scale batch vessel was 1-L cylindrical glass. A predetermined ferrous sulfate dose was added to a 250 mL wastewater sample. The mixture was acidified with the sulfuric acid (to pH = 3) to enhance the oxidation [8,31]. This was followed by the addition of hydrogen peroxide under vigorous stirring to start the Fenton reaction [32]. The reaction time lasted for 4 h, during which the treated samples were collected each hour for analysis. Once the process was completed, NaOH was used to raise the pH of the wastewater samples to nine in order to precipitate the dissolved  $Fe^{3+}$  and degrade the remaining  $H_2O_2$  [33]. To ensure complete removal of the residual  $H_2O_2$ , the samples were heated to 60°C for 30 min. The samples were left until they reached the ambient temperature to sediment the residual iron. Then, they were filtered to remove the precipitated ferrous hydroxide [34]. These experiments were repeated using different ratios of  $H_2O_2$ /COD and  $H_2O_2$ / $Fe^{2+}$ .

## 3. Results and discussion

In the current study, the wastewater was firstly subjected to the coagulation/ flocculation/ sedimentation process to determine the optimum conditions. Chemical coagulation was carried out using the Jar Test procedure. After sedimentation, biological treatment was investigated using an extended aeration system to remove organic materials. Then, advanced oxidation methods were used to remove non-degradable organic materials.

### 3.1. Coagulation, flocculation, sedimentation

The purpose of this stage is to remove the TSS. The removal of TSS will also result in the reduction of COD and  $BOD_5$ .

Chemical coagulation was carried out using the Jar Test procedure to determine the optimum conditions, that is, the type of coagulant and the best coagulant dose. The dose was 200, 300, 400, 500 and 1,000 mg/L of all coagulants. The mixture was rapidly mixed, followed by a slow stirring period. Then, the mixture was allowed to settle.

#### 3.1.1. Effect of type and dose of coagulant

Figs. 1–3 show the effect of the type and dose of coagulant on the removal of COD,  $BOD_5$  and TSS from wastewater. The examined coagulants were sludge, alum, zinc sulfate, and ferric chloride. Using the Jar-test, variable doses of coagulants ranging from 200 to 1,000 mg/L at a pH value of 7.5 were carried out to determine the optimum dose to serve for the treatment.

According to the results, it can be concluded that the COD removal rate for wastewater ranged between 36.1%–50.9% for  $FeCl_3$ , and 34.8%–48.1% for alum, while  $ZnSO_4$  and sludge showed a low COD removal capacity with 24.5%–35.3% for  $ZnSO_4$  and 20.2%–34.8% for sludge. The removal rate for  $BOD_5$  ranged between 30.4%–48.5% for  $FeCl_3$ , 28.6%–46.6% for alum, and 20.4%–41.6% for  $ZnSO_4$ , while sludge showed a low  $BOD_5$  removal capacity with 20.8%–35.6%. For TSS, the same trend was observed as the TSS removal rate ranged between 58.8%–66.4% for  $FeCl_3$ , 57.7%–65.6% for alum, and 54.7%–65.0% for  $ZnSO_4$ , while the TSS removal capacity for sludge was 41.7%–53.9%.

The removal percentage increased from 34.8% to 47.0% for COD, 28.6% to 46.5% for  $BOD_5$ , 57.3% to 63.3% for TSS when the dose of the alum increased from 200 to 400 mg/L, respectively. The percentage removal increased from 36.1% to 47.7% for COD, 30.4% to 46.8% for  $BOD_5$ , 58.8% to 64.9% for TSS when the dose of the ferric chloride was increased from 200 to 400 mg/L, respectively.

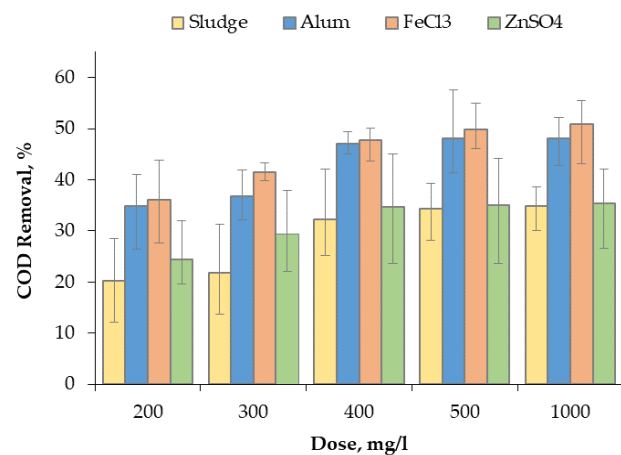


Fig. 1. Effect of type and dose of coagulant on removal of COD in physico-chemical process: initial pH = 7.5, rapid mixing speed = 160 rpm, rapid mixing time = 2 min, slow stirring speed = 60 rpm, slow stirring time = 20 min.

The optimum dose of a coagulant is defined as the value above which there is no significant difference in the magnitude of removal efficiency with a further addition of coagulant or flocculant [35]. Figs. 1–3 show that there is no significant difference in the removal efficiency when increasing the ferric chloride or alum dose up to 1,000 mg/L. Thus, the optimum dose of ferric chloride and alum that enhanced the removal percentage would not exceed. In this regard, El-Gohary et al. [36] indicated that increasing the dose of the ferric chloride enhances the super saturation of the  $Fe(OH)_3$ , which increases the nucleation rate and thus the floc growth rate. As a result, the suspension of a greater number of flocs was improved, resulting in the removal of larger amounts of COD due to the availability of a larger surface area for organic matter adsorption. On the other hand, low doses of ferric chloride led to the formation of larger but fewer flocs as

a result of the faster growth rate compared to the nucleation one, resulting in a smaller surface area on which the adsorption of organic matter occurred [21]. At low doses of coagulant, the predominant removal mechanism is adsorption and charge neutralization [36]. However, the sweep floc coagulation by enmeshment at high coagulant doses is the most common precipitates removal mechanism [37,38].

Accordingly, it is obvious that  $FeCl_3$  and alum showed the best treatment effectiveness with slight preference for  $FeCl_3$ . This is in agreement with the findings of Gautam and Saini [39] who mentioned that alum and ferric chloride are the most efficient coagulants to treat wastewater generated by different industries.

3.1.2. Effect of polymer dosage

Since the best removal rates were obtained from both alum and ferric chloride, sludge and zinc sulfate were excluded from being used as coagulants in this run. It is possible to use either of the two types depending on the availability and costs.

The dose of PAM was changed from 5 to 50 mg/L while the coagulant dose was kept constant at 400 mg/L. In order to determine the optimal dose of PAM, the removal efficiency of COD,  $BOD_5$ , and TSS was taken into consideration (Table 3). From Table 3, it can be seen that the COD and  $BOD_5$  removal efficiency reached 54.9% and 48.8%, respectively at a 400 mg/L alum dose and a 5 mg/L PAM dose, while the COD and  $BOD_5$  removal efficiency increased to 58.8% and 55.9%, respectively when the dose of the PAM increased to 10 mg/L. The COD and  $BOD_5$  removal efficiencies did not increase with a further increase in the PAM dose. For TSS removal, the maximum TSS removal efficiency (67.5%) was achieved at 10 mg/L PAM and a 400 mg/L alum dose. According to Table 3, a 400 mg/L dose of ferric chloride and a 5 mg/L dose of PAM improved the removal efficiency of COD and  $BOD_5$  to 55.7% and 50.6%, respectively. The COD and  $BOD_5$  removal efficiencies increased to 61.8% and 59.7%, respectively when the dose of PAM increased from 5 to 10 mg/L. For the TSS removal, the maximum TSS removal efficiency (71.1%) was achieved at 10 mg/L PAM and a 400 mg/L ferric chloride.

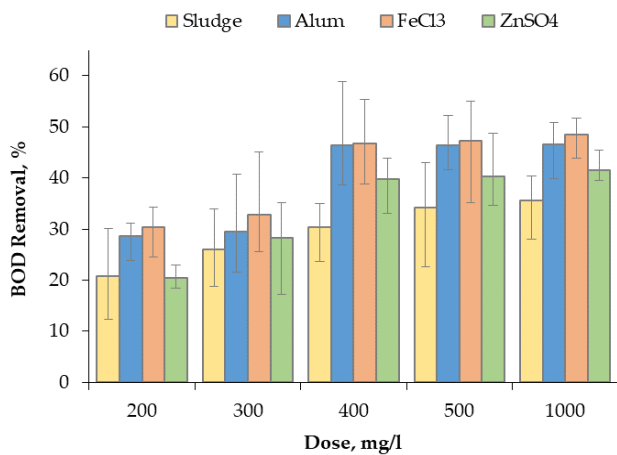


Fig. 2. Effect of type and dose of coagulant on removal of  $BOD_5$  in physico-chemical process: initial pH = 7.5, rapid mixing speed = 160 rpm, rapid mixing time = 2 min, slow stirring speed = 60 rpm, slow stirring time = 20 min.

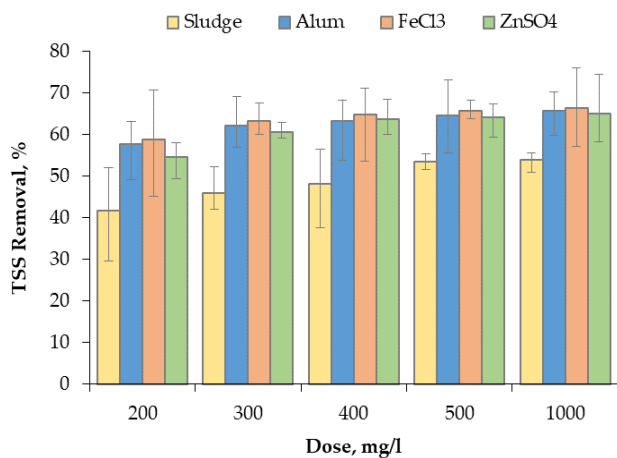


Fig. 3. Effect of type and dose of coagulant on removal of TSS in physico-chemical process: initial pH = 7.5, rapid mixing speed = 160 rpm, rapid mixing time = 2 min, slow stirring speed = 60 rpm, slow stirring time = 20 min.

Table 3 Removal efficiency of COD, BOD, and TSS using 400 mg/L coagulant dose and different doses of PAM as the coagulant aid

Polyelectrolyte dose (mg/L)	0	5	10	15	20	50
<b>Alum</b>						
Removal rate (%)						
COD	46.97	54.94	58.78	57.27	58.13	57.96
$BOD_5$	46.49	48.82	55.89	55.48	55.63	55.78
TSS	63.29	65.01	67.48	67.22	66.96	65.38
<b>Ferric chloride</b>						
Removal rate (%)						
COD	47.72	55.73	61.84	62.41	61.67	61.84
$BOD_5$	46.80	50.65	59.67	59.45	59.07	60.08
TSS	64.90	67.07	71.09	69.06	67.73	66.96

There was a decrease in the removal efficiency when the dose of the PAM increased above 10 mg/L for both alum and ferric chloride.

Based on the previous results, the cooperative effect of the alum or ferric chloride-polymer combination showed a contaminant removal efficiency outperforming the use of alum or ferric chloride alone.

The optimal coagulant dose is the value above which there is no considerable difference in the increase of the removal efficiency with a further addition of coagulant [40]. Thus, the optimum doses of coagulant and PAM that enhanced the removal efficiency were 400 and 10 mg/L, respectively. The use of further doses of PAM caused a further decrease in the TSS removal efficiency. This can be attributed to particle resuspension at higher polyelectrolyte dosages and a high coagulant. It can also be associated with polymer concentrations that may confer positive charges on the particle surface (a positive zeta potential), causing the particles to redisperse [21].

For physico-chemical step, the results show that the pollutant removal rates were close for both ferric chloride and alum. The dose of PAM was changed from 5 to 50 mg/L and the best removal rate was achieved 10 mg/L. Because of the local availability of alum, the closed removal rates and the low cost of alum compared to ferric chloride, pretreated wastewater using alum and PAM was used ahead of biological treatment with doses of 400 and 10 mg/L for alum and PAM respectively.

3.2. Biological treatment

After the primary treatment with alum and PAM, the clarified wastewater was subjected to a biological treatment. Fig. 4 and Table 4 show the effect of changing the HRTs on the removal efficiency of COD, BOD<sub>5</sub>, TSS and TKN. The results show that increasing the HRT for more than 24 h exerted a slight improvement in the pollutant removal efficiency.

The average COD removal rates were 61.2%, 68.9%, 70.6% and 73.1% at HRT of 12, 18, 24 and 36 h, respectively. The average BOD removal rates at HRT of 12, 18, 24 and

36 h were 80.9%, 84.9%, 86.2% and 87.5%, respectively. For the same HRTs, the average TSS removal rates were 54.5%, 57.1%, 58.0% and 61%, respectively. The observed results are similar to those of Krishnan et al. [41] (2008) who stated that an increase in the organic removal is associated with a corresponding increase in the HRT. Also, Pirsahab et al. [42] demonstrated that high HRT (about 18–36 h) increases the ability of the process to withstand the shock organic loading.

Although the biological treatment was satisfactory to some extent at HRT of 24 h, pollution in terms of COD (286 mg/L on average), BOD<sub>5</sub> (70 mg/L on average), TSS (161 mg/L on average) and TKN (13.3 mg/L on average) in the effluent does not meet the Egyptian standards for influents into water bodies.

It is important to note that the biodegradability index (i.e., BOD<sub>5</sub>/COD) of the wastewater decreased after the biological treatment as a result, which means that the effluent cannot be treated by biological degradation.

For biological treatment, the previous results show that increasing the HRT more than 24 h exerted a slight improvement in the pollutant removal efficiency, so biologically treated wastewater at 24 h was used for Fenton process.

3.3. Fenton treatment

Biological wastewater treatment in general is the most economic system if the majority of pollutants contained in the wastewater are biodegradable. This is not always the case due to the presence of a large proportion of pollutants, like those of non-biodegradable compound of industrial wastewater. Although some compounds are biodegradable, the required residence times are excessively long, resulting in significant investment costs [43]. In such cases, additional treatment techniques have to be applied with a significant increase in investment and operating costs.

A Fenton section is provided in the treatment in order to fulfill the environmental regulations for discharge into the aquatic environment (pH = 6–9; COD = 80 mg/L; BOD<sub>5</sub> = 60 mg/L). In general, this stage of treatment is not expected to be used on a regular basis. However, this stage is included to ensure adequate treatment during the sustained peak of COD in the influent from any of the discharging industries. The Fenton's section consists of dosing

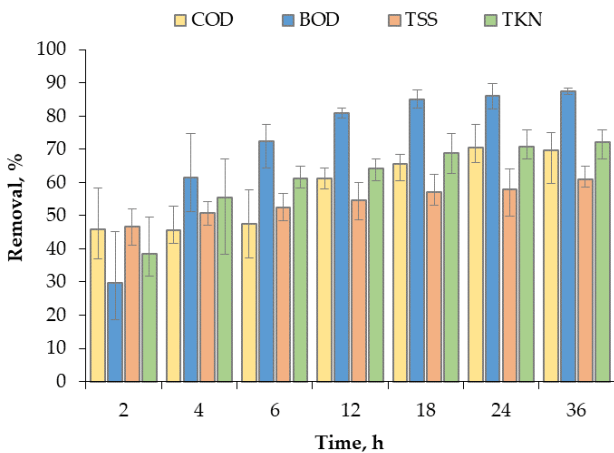


Fig. 4. Effect of HRT on the removal of (a) COD, (b) BOD<sub>5</sub>, and (c) TSS in biological treatment process.

Table 4 Effluent concentrations and removal efficiency of COD, BOD, TSS and TKN at different HRTs

HRT (h)		6	12	18	24	36
Effluent concentrations	COD (mg/L)	510	376	302	286	261
	BOD <sub>5</sub> (mg/L)	138	96	76	70	63
	TSS (mg/L)	183	175	165	161	150
	TKN (mg/L)	18	16	14.2	13.3	13
Removal rate (%)	COD	47.4	61.2	68.9	70.6	73.1
	BOD <sub>5</sub>	72.5	80.9	84.9	86.2	87.5
	TSS	52.3	54.5	57.1	58	61.0
	TKN	61.1	64.1	68.9	70.7	72.1



systems, for hydrogen peroxide, and ferrous sulfate. The influent is dosed with the chemical in an oxidation tank, followed by settling.

### 3.3.1. Effect of hydrogen peroxide concentration

The biologically treated wastewater at HRT of 24 h was subjected to the Fenton treatment. hydrogen peroxide is the most important component in the Fenton process. The concentration of hydrogen peroxide used in the Fenton process depends primarily on the initial COD value. According to the stoichiometric ratio between hydrogen peroxide ( $H_2O_2$ ) and COD, the typical  $H_2O_2$ /COD ratio is 2.125 [44], but this value does not apply to all wastewaters, especially those with inorganic components as well as organic compounds. In the present study, different  $H_2O_2$ /COD ratios, ranging from 0.9 to 4.7, were investigated in order to determine the optimum ratio for the treatment. The pH was set to 3 and the  $H_2O_2/Fe^{2+}$  molar ratio was to 50. Regular samples were obtained from the reaction in order to determine the residual COD,  $BOD_5$ , and TSS.

Figs. 5–7 show the effect of  $H_2O_2$  concentration on the removal rate of COD,  $BOD_5$  and TSS. Results indicated that the COD and  $BOD_5$  removal rates increased along with the increase of  $H_2O_2$  concentration. These results are similar to those of Afolabi et al. [45] who indicated that increasing concentrations of hydrogen peroxide and reaction time favored the BOD and COD removal for brewery wastewater. Regarding the Fenton's reaction, increasing the  $H_2O_2$  concentration led to an increase in the reaction rate, so more hydroxyl radicals were generated [46]. The maximum removal was achieved at  $H_2O_2$ /COD = 4.7; however, the difference in the removal percentage in the case of  $H_2O_2$ /COD = 3.7 and  $H_2O_2$ /COD = 4.7 was negligible. Thus,  $H_2O_2$ /COD = 3.7 was assumed to be the optimum ratio. Maximum percentage removal at  $H_2O_2$ /COD = 3.7 was found to be 83.61%, 55.12% and 61.49% for COD,  $BOD_5$  and TSS, respectively.

The results demonstrated that the Fenton treatment of the mixed domestic–industrial wastewater effluent improves the biodegradability index ( $BOD_5$ /COD increases

from 0.28 to 0.55). Similarly, Jagadevan et al. [47] showed that  $BOD_5$ /COD increases from 0.160 to 0.538 for metal-working fluids.

### 3.3.2. Effect of the $Fe^{2+}$ concentration

For the Fenton process, the ratio of hydrogen peroxide and ferrous iron ( $H_2O_2/Fe^{2+}$ ) is a critical operational parameter, since iron salts are used as a catalyst for the decomposition of hydrogen peroxide to hydroxyl radicals. For this reason, five different  $H_2O_2/Fe^{2+}$  molar ratios were tested to determine the optimum  $H_2O_2/Fe^{2+}$  molar ratio. To study the effect of the  $Fe^{2+}$  concentration, the hydrogen peroxide dosage was kept constant at 40 mM, while the  $Fe^{2+}$  dosage was variable to produce an  $H_2O_2/Fe^{2+}$  molar ratio ranging from 20 to 160. The pH was controlled to be around 3. An aliquot of the sample was obtained from the reaction at predetermined intervals in order to assess the residual COD.

Fig. 8 illustrates the effect of the  $Fe^{2+}$  concentration on the COD removal percentage. The COD removal efficiencies

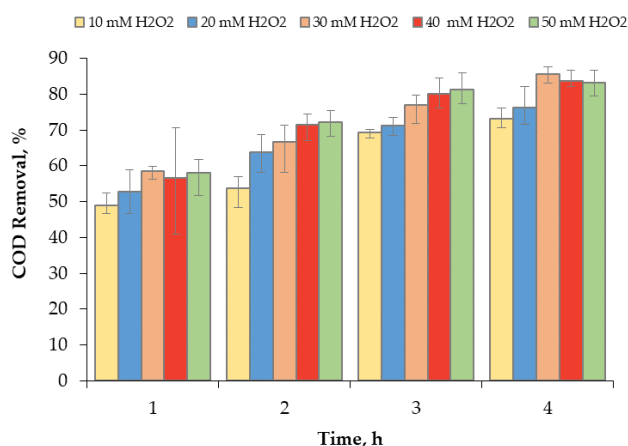


Fig. 5. Effect of  $H_2O_2$  concentration on the removal of COD in Fenton process: pH = 3,  $H_2O_2/Fe^{2+}$  molar ratio = 50.

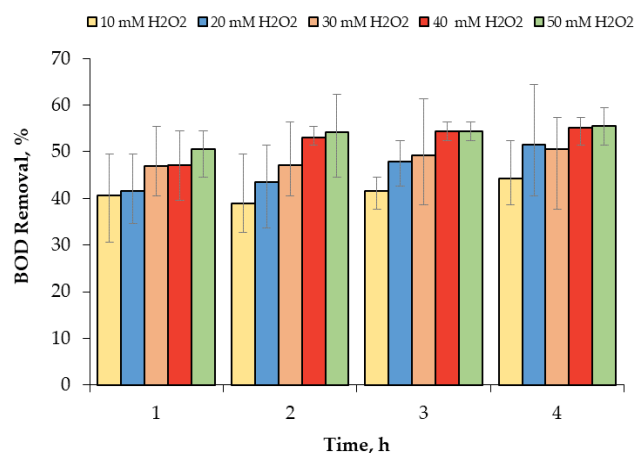


Fig. 6. Effect of  $H_2O_2$  concentration on the removal of  $BOD_5$  in Fenton process: pH = 3,  $H_2O_2/Fe^{2+}$  molar ratio = 50.

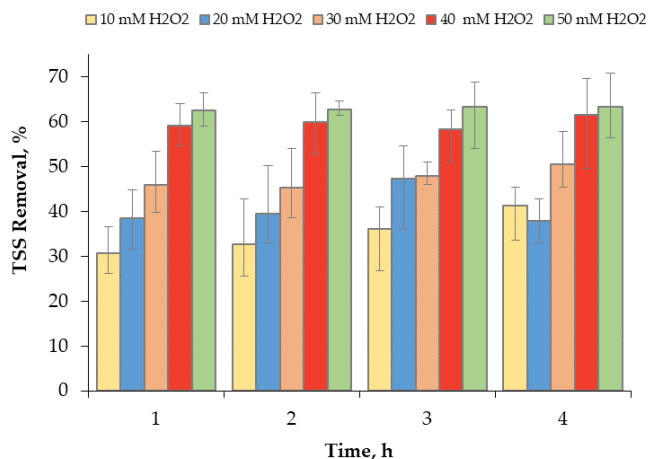


Fig. 7. Effect of  $H_2O_2$  concentration on the removal of TSS in Fenton process: pH = 3,  $H_2O_2/Fe^{2+}$  molar ratio = 50.

Table 5  
Operational costs analysis

Treatment step	Chemicals	Dose	Cost (\$/m <sup>3</sup> )	Cost per step (\$/m <sup>3</sup> )
Physico-chemical step	Alum	400 g/m <sup>3</sup>	0.5	0.8
	Polyelectrolyte	10 g/m <sup>3</sup>	0.3	
Fenton process	Hydrogen peroxide	40 mol/m <sup>3</sup>	10.6	11.9
	Ferrous sulfate	1 mol/m <sup>3</sup>	1.3	

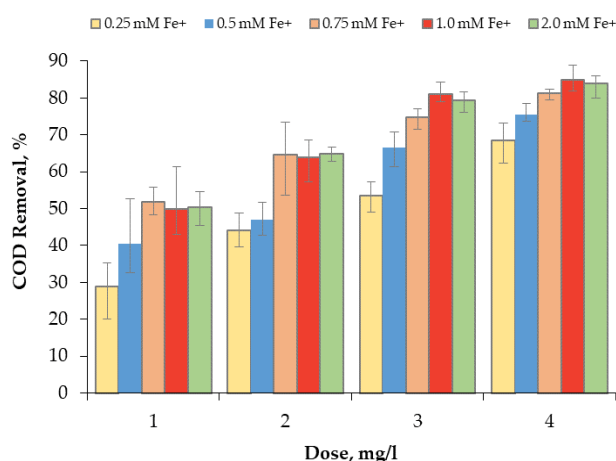


Fig. 8. Effect of Fe<sup>2+</sup> concentration on the removal of COD: pH = 3, H<sub>2</sub>O<sub>2</sub> = 40 mM.

increased with the reduction of the H<sub>2</sub>O<sub>2</sub>/Fe<sup>2+</sup> molar ratio. COD removal percentage was only 68.38% at the molar ratio of 160 but increased to 84.91% at the molar ratio of 40. A further decrease in H<sub>2</sub>O<sub>2</sub>/Fe<sup>2+</sup>, from 40 to 20, did not lead to any improvement in COD removal percentage. The observed COD removal percentage using the Fenton process was within the reported COD removal efficiency, which ranged from 45% to 85% [48–51].

The optimum H<sub>2</sub>O<sub>2</sub>/Fe<sup>2+</sup> molar ratio was considered to be 40, since there is no substantial removal improvement when the ratio of H<sub>2</sub>O<sub>2</sub>/Fe<sup>2+</sup> was 20. According to Cuerda-Correa et al. [8], as the iron concentration increases, the rate of oxidation of organic compounds increases to the point where a further increase in iron concentration is ineffective. This result, attributed to the traditional Fenton reaction, implies the oxidation of ferrous (Fe<sup>2+</sup>) to ferric ions (Fe<sup>3+</sup>) which decomposes hydrogen peroxide into hydroxyl radicals (HO<sup>•</sup>) and OH<sup>-</sup> [52,53]. However, the generated Fe<sup>3+</sup> can be reduced and form more radicals and ferrous iron in the presence of excess hydrogen peroxide [54]. The formation of hydroperoxyl radicals, which are less reactive in attacking the recalcitrant compounds in the liquid phase, can be considered another explanation [55]. The higher molar ratio is not favorable because large amounts of these dissolved iron salts are necessary. In addition this makes the process more expensive [56]. Based upon these results, the optimum reaction conditions with different initial concentrations are H<sub>2</sub>O<sub>2</sub>: COD = 3.7 and H<sub>2</sub>O<sub>2</sub>:Fe<sup>2+</sup> = 40.

After the oxidation treatment at a hydrogen peroxide dosage of 40 mM, the effluent parameters in terms of COD,

BOD<sub>5</sub> and TSS can reach the values of 56, 43, and 49 mg/L, respectively. Accordingly, these values meet the standards for influents into water bodies according to the Egyptian standards.

From the obtained results, it can be concluded that complete oxidation was not possible. This may be due to the inhibitors found in industrial wastewater, such as high alkalinity, chlorides, sulfates, etc. Another factor is the stable carboxylic acid-Fe complexes produced by the Fenton process. As such, complete mineralization cannot generally be achieved using Fenton.

#### 3.4. Chemical cost

The costs of treatment vary a lot, depending on the local costs of power, chemicals, financing, labor and sludge handling and disposal options [57]. In this study, operational costs in terms of chemicals (coagulation/flocculant aid, hydrogen peroxide and ferrous sulfate) are shown in Table 5.

## 4. Conclusions

The chemical coagulation process using ferric chloride and alum (200–1,000 mg/L) achieved the COD removal efficiencies of 36.1%–50.9% and 34.8%–48.1%, respectively. The removal rate of BOD<sub>5</sub> ranged between 30.4%–46.8% for FeCl<sub>3</sub>, 28.6%–46.5% for alum, and 20.4%–41.6% for ZnSO<sub>4</sub>, while sludge showed an insufficient BOD<sub>5</sub> removal capacity. The TSS removal rate ranged between 58.8%–66.4% for FeCl<sub>3</sub>, 57.7%–65.65% for alum, and 54.7%–65.05% for ZnSO<sub>4</sub>, while the insufficient TSS removal capacity was shown for sludge. Generally, results pointed out that FeCl<sub>3</sub> and alum showed the best treatment effectiveness with a slight preference for FeCl<sub>3</sub>. The best coagulant dose was considered 400 mg/L, since increasing the ferric chloride or alum dose up to 1,000 mg/L exerted a slight improvement in the coagulation efficiency. Using 10 mg/L PAM as a coagulant aid could improve the pollutant removal efficiency. The COD, BOD<sub>5</sub> and TSS removal efficiencies at 10 mg/L polyelectrolyte and 400 mg/L alum doses were 58.8%, 55.9% and 67.5%, respectively. On the other hand, the removal efficiencies of COD, BOD<sub>5</sub> and TSS were 61.8%, 59.7% and 71.1%, respectively at a 400 mg/L dose of ferric chloride and a 10 mg/L dose of polyelectrolyte. Due to the low cost of alum compared to ferric chloride and the closed removal rates for them, alum was considered the most suitable coagulant, so pretreated wastewater with 400 mg/L alum and 10 mg/L PAM was used ahead of biological treatment. The clarified wastewater after



primary treatment was subjected to a secondary treatment, which mainly consisted of a biological treatment followed by settling. The removal rates at HRT of 24 h for COD, BOD<sub>5</sub> and TSS were 70.6%, 86.2% and 58%, respectively. Results showed that increasing the HRT more than 24 h exerted a slight improvement in the pollutant removal efficiency. Although the biological treatment was satisfactory to some extent at HRT of 24 h, but pollution in terms of COD, BOD<sub>5</sub>, TSS and TKN in the effluent does not meet the standards for influents into water bodies or for the reuse according to the Egyptian standards. It is important to note that the biodegradability index (i.e., BOD<sub>5</sub>/COD) of the wastewater decreased after the biological treatment, which means that the effluent cannot be further treated by biological degradation. Therefore, a Fenton section was provided in the treatment in order to fulfill the environmental regulations for discharge into the aquatic environment. Fenton was effectively used to reduce concentrations of organic contaminants of industrial wastewater. The effect of hydrogen peroxide and Fe<sup>2+</sup> concentrations on the removal percentage of pollutants was highly studied. The maximum removal was achieved at H<sub>2</sub>O<sub>2</sub>/COD = 4.7; however, the difference in the removal percentage in case of H<sub>2</sub>O<sub>2</sub>/COD = 3.7 and H<sub>2</sub>O<sub>2</sub>/COD = 4.7 was very small. So the ratio H<sub>2</sub>O<sub>2</sub>/COD = 3.7 was assumed to be the optimum one. In order to investigate the optimum H<sub>2</sub>O<sub>2</sub>/Fe<sup>2+</sup> molar ratio, five different H<sub>2</sub>O<sub>2</sub>/Fe<sup>2+</sup> molar ratios were tested. The H<sub>2</sub>O<sub>2</sub>/Fe<sup>2+</sup> molar ratio ranged from 20 to 160. The COD removal percentage was only 68.38% at the molar ratio of 160 but increased to 84.9% at the molar ratio of 40, a further decrease in H<sub>2</sub>O<sub>2</sub>/Fe<sup>2+</sup> from 40 to 20 did not lead to any improvement. From the obtained results, it can be concluded that complete oxidation was not possible. This can be due to the presence of inhibitors in the industrial wastewater, such as high alkalinity chlorides and sulfates.

### Acknowledgments

The author expresses her gratitude to the staff of the Environmental Engineering Laboratory, Faculty of Engineering, Zagazig University.

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