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Original Article

Removal of Antibiotics From Hospital Wastewater Using Hybrid Chemical Purification and Batch Biological Reactor

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Abstract

Background: This study aimed to investigate the performance of tetracycline and amoxicillin antibiotics removal from hospital wastewater using a combination of chemical treatment and Batch biological reactor.

Methods: In this study, the effect of PS/Fe^{2+}/UV on tetracycline and amoxicillin removal was investigated. Different parameters including pH, different concentrations of iron, antibiotics and persulfate were investigated for removal efficiency. The remaining concentration of the solution was measured using a high performance liquid chromatography (HPLC) device. The degree of mineralization of the process was evaluated using the chemical oxygen demand (COD) parameter. Then, the removal of tetracycline, amoxicillin and COD from the pre-treated wastewater in the batch biological reactor was investigated.

Results: The results demonstrate that the PS/Fe^{2+}/UV process achieved complete removal of tetracycline and 97.3% removal of amoxicillin under laboratory conditions. In wastewater treatment, the process achieved a 69% reduction in tetracycline and a 67.2% reduction in amoxicillin within a 60-minute reaction time. Also, the amount of mineralization of tetracycline and amoxicillin antibiotics by PS/Fe^{2+}/UV process was evaluated using COD index, which resulted in removal of more than 60% of COD for both antibiotics. Also, the combination of this process and the batch biological reactor succeeded in removing 99% of BOD_5 from both antibiotics and 98% of COD from real wastewater, containing 10 mg of tetracycline and amoxicillin.

Conclusion: The combined PS/Fe^{2+}/UV process and batch biological reactor is an efficient method. It is effective for removing antibiotics and its mineralization for wastewater treatment containing tetracycline and amoxicillin antibiotics.

Keywords: Antibiotic, Advanced purification, Discontinuous biological reactor, Tetracycline, Amoxicillin

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Introduction

The rapid growth of human activities has led to the emergence of new toxic and dangerous chemical compounds called "emerging pollutants" that can cause unpredictable consequences for ecosystems.[1] Antibiotics, as one of the emerging pollutants, have received increasing attention in recent years.[2] The discharge of wastewater from pharmaceutical industries, hospitals, livestock farms, leachates from landfills and wastewater refineries are considered as the main sources of antibiotic release in the environment. The remarkable point about the use of medicinal substances is that only less than ten percent of medicinal substances are changed in the body and the rest is eliminated from the human body without any change.[3] Wastewater treatment facilities are typically designed without taking into account the removal of antibiotics, leading to the substantial release of these chemicals into the aquatic environment.[4] The unregulated release of antibiotics into the environment contributes to the proliferation of antibiotic-resistant bacteria and is associated with the presence of antibiotic-resistant genes in aquatic ecosystems. This trend may potentially render antibiotics ineffective in the treatment of various diseases in the near future[3]. Wastewater refineries act as reservoirs for antibiotic-resistant bacteria.[5] The concentrations of amoxicillin and tetracycline antibiotics have been reported as 6700 ng/L and 3743 ng/L in the environment.[6] Also, the conducted studies show that the concentration of antibiotics in hospital effluents is between 0.3 to 200 μg/L.[7]

There are different methods to remove antibiotics from water sources. These processes include advanced oxidation, ion exchange, activated carbon adsorption, ozonation and membrane processes, reverse osmosis and biological purification.[3] Each of these methods has its own set of advantages and disadvantages. For instance, techniques like adsorption with activated carbon and membrane processes do not eliminate pollutants but rather facilitate their transfer from one phase to another.[8] Advanced oxidation completely destroys the pollutant.[9] The batch biological reactor has provided the conditions for cell retention time and higher biomass concentration.[10] The integration of a continuous biological reactor process with chemical oxidation significantly enhances the purification efficiency for the removal of pollutants in hospital effluents, including chemical oxygen demand (COD), biochemical oxygen demand (BOD), nitrogen, and phosphorus. This approach is also employed to mitigate the presence of drugs, hormones, and antibiotics.[11]

Similar to some countries, improper design or management in Iran results in biological units being unable to effectively eliminate pollutants present in hospital wastewater, including COD, BOD, and even degradable organic substances like drugs, hormones, and antibiotics.

There are many studies in the field of contamination caused by antibiotics in effluent and water sources. Ghaffari et al used the Fenton oxidation process to remove tetracycline from synthetic wastewater. In the study, the optimal conditions of the Fenton process for the removal of tetracycline are initial concentration of 57 mg/L, H_2O_2 / Fe_2 molar ratio of 11.27 and contact time of 30 minutes, which resulted in the removal of 90.15% of tetracycline and 74.6% of COD.[12] Furthermore, a study conducted in Singapore in 2016 investigated the removal of antimicrobial substances in biological membrane systems as compared to conventional activated sludge systems. The results showed that biological membrane systems exhibited higher efficiency in removing these substances, with the beta-lactam, glycopeptide, and fluoroquinolone classes predominantly eliminated by the biological wastewater purification processes. However, trimethoprim and lincosamides appeared to remain stable.[13]

Given the escalating significance of water source pollution, employing high-efficiency processes in industries becomes an imperative concern. Hence, this study was conducted to assess the effectiveness of the advanced persulfate oxidation process coupled with the process of suspended biological aerobic growth for the reduction of antibiotics, specifically tetracycline and amoxicillin, extracted from hospital wastewater.

Materials and Methods

The present study is an experimental study. which aims to evaluate the efficiency of the combined method - advanced oxidation (persulfate, iron and UV) and batch biological reactor in removing tetracycline and amoxicillin from aqueous solutions on a pilot scale and a real sample of hospital wastewater.

Materials

In this research, all chemicals such as mercury sulfate ($HgSO_4$), silver sulfate (Ag_2SO_4), concentrated sulfuric acid (H_2SO_4), Hydrochloric acid (HCl), sodium thiosulfate (Na_2S_2O3), iron sulfate (VII) ($FeSO_4.7H_2O$), sodium persulfate ($Na_2S_2O_8$) (99%), Nitric acid (HNO_3) with laboratory purity were obtained from Merck and standard powders of antibiotics tetracycline and amoxicillin with a purity of more than 98%. To prepare laboratory solutions, stock solutions with 1000 mg/L of tetracycline and amoxicillin were prepared by adding a certain amount of them to one liter of distilled water and diluting with double distilled water. To investigate the effect of pH, 0.1 normal NaOH and HCl solutions were used. In this study, a reactor with a volume of 500 mL of quartz and several cylindrical aerobic biological reactors with an approximate volume of 2 L of Plexiglass or glass were also used. To measure the pH, the Haft Asan pH meter of Metler Toledo, Switzerland was used. A mechanical mixer was applied to mix the contaminants and reactants. Also, to measure the residual concentration of antibiotics in the reaction solution from high performance liquid chromatography (HPLC), Agilent 1200 USA with C18 ODS 250 column×4×6×5 was used.[13,14] The HPLC system equipped with a UV detector set to the maximum absorption wavelengths for amoxicillin at 254 nm.[15] and for tetracycline at 360 nm,[13] as described in the design and usage details.[16] A type C UV lamp with an intensity of 8 watts, made in Poland, PHILIPS model, was used. All the devices used were calibrated according to the relevant catalogs before conducting the experiments.

In order to optimize the factors affecting the process, the effect of each important operating parameters including initial pH (2, 4, 6, 8, and 10), iron concentration (1.5-5 g/L), initial concentration of persulfate (5 1-), the initial concentration of tetracycline and amoxicillin (5, 10, 15, 20.30), reaction time (5, 10, 15, 20, 30 and 60 minutes), the radiating power of the UV lamp 8 W and the cell retention time (16-24-36 hours) were studied. To measure the parameters effect in each step, only one of the parameters was selected as the variable and the others were fixed. The study determined the optimal dosage and the impact of each variable on the removal of antibiotics. The range of these variables was selected based on a comprehensive review of the literature. It is important to note that throughout all stages of the research, each experiment was conducted three times, and the average of these repetitions was taken as the final results.[9]

In this research, data normality assessment and the comparison of means between different treatments were conducted using SPSS software version 26. Graphs were generated, and equations were applied using Microsoft Excel 2013. All data are presented as mean ± standard deviation with a precision of ± 0.05.

Methods

The tests were performed in two steps as follows:

Step 1: Chemical Purification Process

In this study, which was conducted both in a laboratory and at a pilot scale, all chemical oxidation experiments were conducted within a cylindrical quartz reactor designed to transmit UV waves. The reactor had a total volume of 500 mL, with 250 mL of solution used for each reaction. The experimental procedure began by preparing synthetic effluent solutions containing varying initial concentrations of tetracycline and amoxicillin (5, 10, 15, 20, 30). These solutions were then transferred to the reactor. Following this transfer, the pH and temperature were adjusted as needed, and specific concentrations of iron ions and sodium persulfate were added to the solution inside the reactor. Following these steps, a UV lamp with an intensity of 8 watts was employed, and the contents within the reactor were mixed at a rate of 300 revolutions per minute.

Subsequently, at specified time intervals (5, 10, 15, 20, 30, and 60 minutes), wastewater samples were collected. To measure the concentrations of tetracycline and amoxicillin in these samples, HPLC)equipped with an ultraviolet detector was utilized. In this step, the degree of mineralization of the processes was investigated using the COD parameter.[17-19] In an optimal condition, the efficiency of the simultaneous removal process of both antibiotics and their removal from the actual sample of hospital wastewater were evaluated.

Second Stage: Biological Purification Process

In the second stage, several cylindrical aerobic biological reactors with an approximate volume of 2 L made of plexiglass or glass were used (Figure 1). Each cycle of the reactor was fed with 2 L of fresh wastewater, and 2 L of purified wastewater was removed at the end. The age of the sludge was about 20 days, which was kept constant by the amount of excess sludge removed from the reactor in every 24-hour period and the amount of biomass in the effluent.[15] A deep membrane diffuser was employed to aerate the reactor, and the selection of working cycles for the reactors was based on the specific type and characteristics of the wastewater. The working cycles for all reactors consisted of aeration periods lasting 16, 24, and 36 hours, followed by a settling period of one hour and 30 minutes. The sludge age was maintained at 10, 20, and 30 days.

To initiate the reactors, 1 L of activated sludge was extracted from the return line of the hospital wastewater treatment plant and introduced into each reactor. The remaining volume of the reactors, up to 2 L, was filled with the effluent from the PS/Fe^{2+}/UV process. For the initial series of experiments, the operational regimen of the reactors was set in accordance with a predetermined schedule. Subsequently, the reactors operated based on the regulatory program for a duration of two weeks. At the end of each week, samples were collected from both the inlet and outlet of the reactors, and analyses were conducted to measure pH and oxygen concentration (COD). A minimum of one week was allocated for each alteration in reaction or aeration time to establish the cell retention time, including periods of 10 days, 20 days, and 30 days. Likewise, at least one week was allowed for adjusting the sludge age to adapt the system. The measurements were conducted under the new conditions once the system had reached a state of stability. Some parameters such as pH, concentration of suspended solids within the reactor, and outlet COD were used as indicators to confirm the stabilization of the conditions.[19]

All samplings and tests were done according to the guidelines of the 2012 *Standard of Water and Wastewater Testing Methods* book, which has been presented in Table 1.[18]

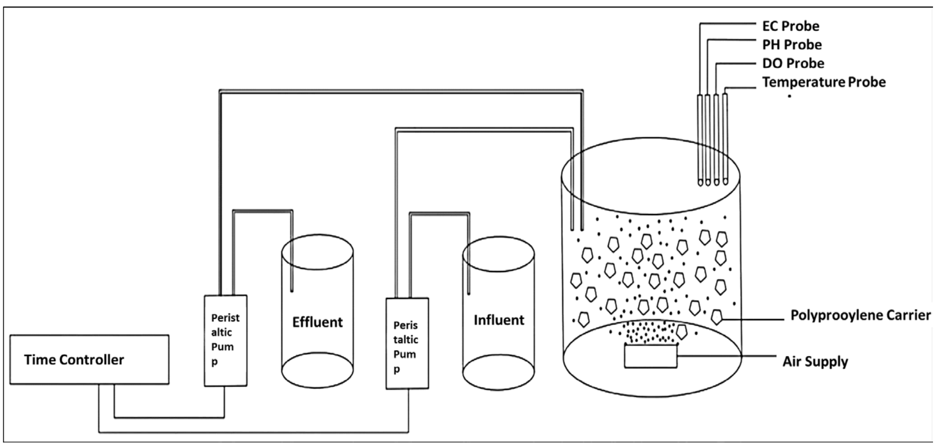

Figure 1. Schematic Diagram of the Hybrid Sequential Discontinuous Reactor System.[19]

Table 1. The Instructions of the Book Related to Standard Methods for Water and Wastewater Testing in 2012

Type of Test	Measurement Method
Tetracycline	HPLC or GC-mas method
Amoxicillin	HPLC or GC-mas method
pH	Electrode method (Standard printing method 22 pages 4 - 94)
MLSS	Gravimetric method (Standard printing method 22 pages 2 -81) or EPA 1983 Method 4/160)
SVI and settling capability	Gravimetric method (standard printing method 22 pages 2 - 84 or EPA 1983 Method 5/160)
DO	Electrode method (standard printing method 22 pages 4 - 152 or EPA 1983 method 2/360)
BOD_5	Manometric method (Printing method standard 22 pages 4 - 5 or EPA 1983 Method 1/405)
COD	Chemical method (standard printing method 22 pages 14 - 5 or method Calorimeter with DR2000)
Sludge age	Computational method with relation
F/M	Computational method with relation
Temperatures	Thermometer
Kinetic coefficient Kd	Computational method with relation
Kinetic coefficient Y	Computational method with relation

Abbreviations: MLSS, mixed liquor suspended solids; COD, chemical oxygen demand; BOD, biochemical oxygen demand; SVI, sludge volume index; DO, dissolved oxygen.

Results and Discussion

Figure 2 shows the removal efficiency of different processes for the studied antibiotics under the same operating conditions. The maximum and minimum removal efficiency of both antibiotics were related to PS/Fe^{2+}/UV and UV processes, respectively. Therefore, among the studied processes, the PS/Fe^{2+}/UV process was chosen due to its higher efficiency in the removal of tetracycline and amoxicillin. Its optimization steps and the effect of different operating parameters on its performance were evaluated.

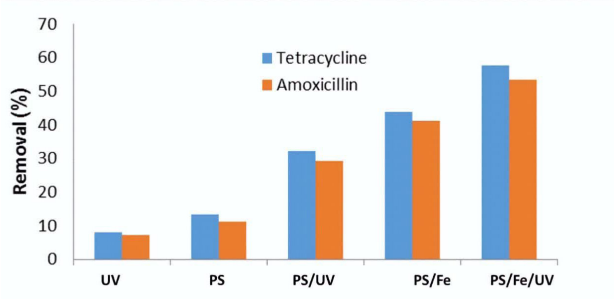

Figure 2. Comparison of Different Processes for the Removal of Tetracycline and Amoxicillin Under the Same Operating Conditions

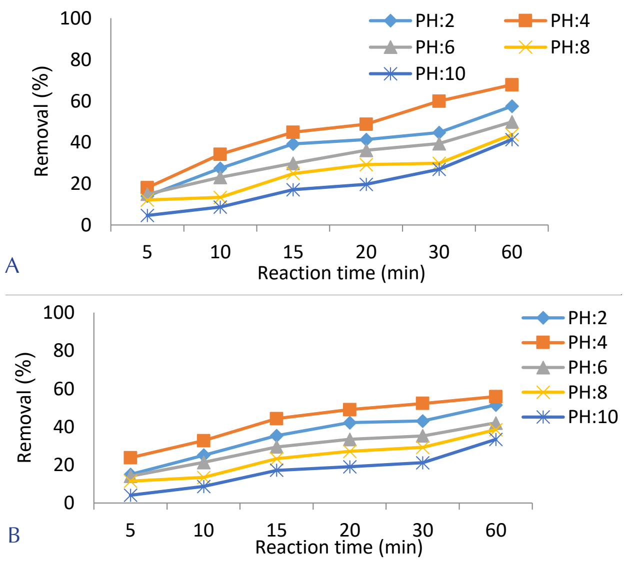

Figure 3. Effect of Different pH on the Removal Efficiency of Tetracycline (A) and Amoxicillin (B) by PS/Fe^{2+}/UV

The Effect of Experimental Factors

Effect of pH Solution

The pH is considered as one of the most important parameters for the removal process, which affects the chemical and biological reactions of wastewater[20] (Figure 3). The results show that the effect of pH is between 2% and 10% at standard ambient temperature and in the presence of UV lamp with an intensity of 8 watts. As seen in Figure 3, antibiotic removal efficiency increases with decreasing pH. Reduction of antibiotic removal in alkaline conditions can be caused by the simultaneous precipitation of iron and formation of hydroxide complex, conversion of sulfate radicals to hydroxide radicals, which produces species with lower oxidation potential.[21] The maximum removal efficiency of tetracycline (67.8%) and amoxicillin (55.9%) were related to pH 4. The reason for the high efficiency of this process in acidic pH can be attributed to the reactions in acidic environment as presented in the following reactions (Equations 1-4).[22] However, at high and low values of pH, the removal performance of both antibiotics decreased significantly. In order to continue the other optimization steps of the PS/Fe^{2+}/UV process, pH 4 was selected and used as the optimal value for both antibiotics. In the researches by Yazdani et al and Kamani et al, the same results were found. The studies revealed that the process exhibited higher overall efficiency under acidic pH conditions compared to alkaline pH. Increasing pH was observed to diminish the effectiveness of organic substance removal from synthetic wastewater.[22-24]

$$2HO_2^{\circ} \rightarrow O_2 + H_2O_2 \quad (1)$$

$$H_2O_2 + O_2^{\circ-} \rightarrow OH^{\circ} + OH^{-} + O_2 \quad (2)$$

$$e^{-} + O_2 \rightarrow O_2^{\circ-} \quad (3)$$

$$O_2^{\circ-} + H^{+} \rightarrow HO_2^{\circ} \quad (4)$$

Effect of Iron Content

Figure 4 represents the effect of different concentrations of iron (Fe^{2+}) (i.e., 0.1 to 0.5 g/L) on t removal efficiency of tetracycline and amoxicillin by the PS / Fe^{2+}/ UV process at optimal pH of 4 and in the presence of 3 mM persulfate over time which was performed for 60 minutes in the presence of a UV lamp with an intensity of 8 watts and at ambient

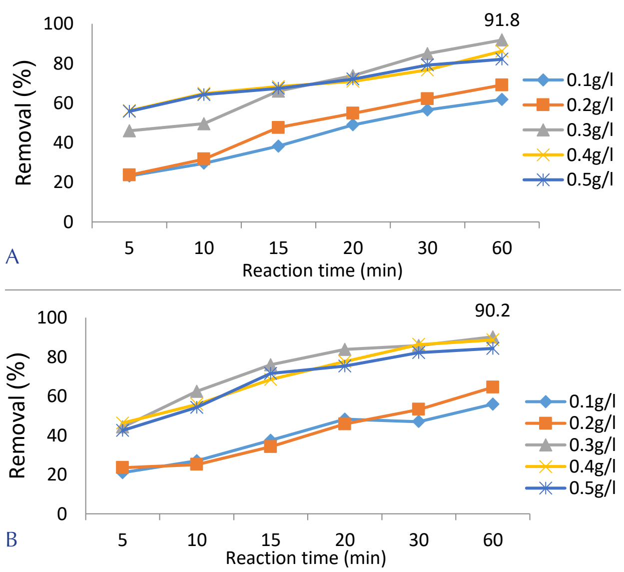

Figure 4. Effect of Different Concentrations of Ferrous Ions on the Performance of the Removal Process of Tetracycline (A) and Amoxicillin (B) by PS/Fe^{2+}/UV

temperature. According to Figure 4, the results show that by increasing the amount up to 0.3 g/L, the removal efficiency of tetracycline and amoxicillin antibiotics was 91.8 and 90.2, respectively, which had an increasing mode. These findings can be explained by the fact that the increase in PS decomposition is high in the presence of additional redox active centers and the removal efficiency decreases as the concentration increases from 0.4 to 0.5 g/L.[25] The results showed that the increasing Fe^{2+}ions up to a certain level can improve the efficiency of the process in antibiotic removal, while increasing Fe^{2+}ions does not have a significant effect on the efficiency of the process. This phenomenon can be attributed to the effect of $SO^{4\bullet-}$ with the presence of additional iron ions and the self-association of iron particles, which reduces the number of reactive sites of the catalyst at higher concentrations.[9] In the case of both studied antibiotics, the optimal concentration of iron ions was determined to maximize the removal efficiency during the Fe^{2+}/PS process. A concentration of 0.3 g/L Fe^{2+}/UV was selected and subsequently employed in other oxidation experiments. In the studies conducted by Ahmadi et al and Mehrdadi et al, two major reasons for this phenomenon were reported: (1) the reaction of excess ferrous ions with free radicals and preventing their reaction with organic compounds, (2) The increased production of free radicals in the reaction solution leads to their subsequent re-generation of $(S_2O_8)2$ through mutual reactions.[9,26]

Effect of Persulfate Amount

Figure 5 shows the effect of persulfate content on the removal efficiency of tetracycline and amoxicillin by the PS/Fe^{2+}/UV process. At low concentrations of PS, there is not enough concentration of sulfate free radicals to remove antibiotics. Indeed, as the molar ratio of PS to pollutant increases, more oxidant molecules are available

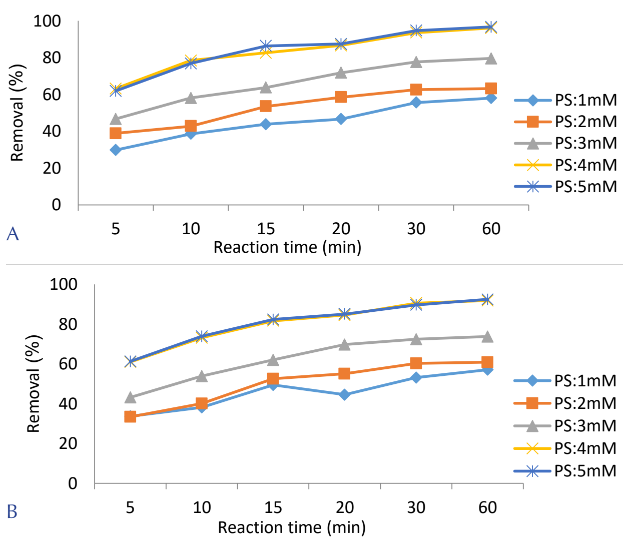

Figure 5. Effect of Different Amounts of Persulfate on the Removal Efficiency of Tetracycline (A) and Amoxicillin (B) by the PS/Fe^{2+}/UV Process

to attack the organic matter, which improves the removal efficiency.[25] This is consistent with our findings. Since the highest removal efficiency of antibiotics was achieved at a 5 mM persulfate concentration, this value was chosen as the optimal amount for further optimization experiments in the PS/Fe^{2+}/UV process.

The Effect of Initial Antibiotics Concentration

Figure 6 shows the effect of initial concentration of tetracycline and amoxicillin (5, 10, 15, 20.30 mg/L) on the performance of PS/Fe^{2+}/UV in the presence of an 8 W UV lamp and standard ambient temperature. The results indicate that increasing the antibiotic concentration from 5 to 30 mg/L under optimal conditions leads to a decrease in antibiotic removal efficiency. The increase in antibiotic concentration reduces the ratio of hydroxyl radicals produced in relation to the antibiotic and elevates the formation of intermediate compounds that tend to consume free radicals. Furthermore, an increase in antibiotic concentration diminishes the penetration of UV light into the solution, potentially due to increased turbidity. This, in turn, leads to a reduction in the rate of pollutant decomposition by persulfate and a decrease in the percentage of decomposition.[27] In studies conducted by Tavassoli et al on the removal of the antibiotic ofloxacin using sodium persulfate activated by ultraviolet light, a similar trend was observed as in the research by Hoseini et al, where the percentage of removal decreased with an increase in antibiotic concentration.[22,27]

Comparison Between Single and Simultaneous Systems

Figure 7 shows the performance of PS/Fe^{2+}/UV process on the simultaneous removal of both antibiotics under optimal conditions. It was observed that the efficiency of the PS/Fe^{2+}/UV process in removing both antibiotics in a separate system was higher than the simultaneous system.

According to Figure 7, the removal percentage of tetracycline and amoxicillin antibiotics was 99.6 and

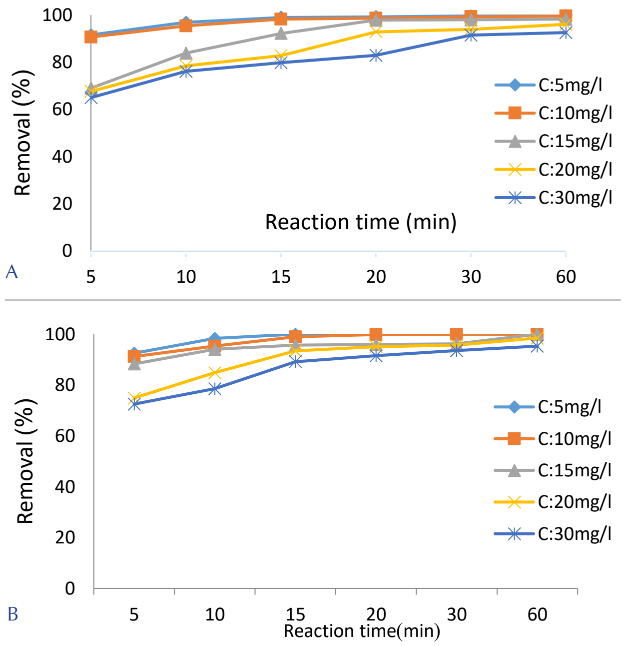

Figure 6. Effect of Initial Concentrations of Tetracycline (A) and Amoxicillin (B) Antibiotics on PS/Fe^{2+}/UV Process Performance

98.1 in the individual system and 92.5 and 88.4 in the simultaneous system, respectively. The removal efficiency of antibiotics in the simultaneous system was lower than the individual system.

It can be concluded that a sufficient quantity of radicals is generated to enable the simultaneous removal of antibiotics. The results suggest that the PS/Fe^{2+}/UV system exhibits high baseline efficiency for the simultaneous removal of antibiotics, making it a promising option for treating polluted water containing multiple pollutants.

The Degree of Mineralization

Figure 8 shows the amount of mineralization of tetracycline and amoxicillin antibiotics using the PS/Fe^{2+}/UV process by checking the COD index.

According to the results presented in Figure 8, COD removal of more than 60% was obtained for tetracycline and amoxicillin antibiotics. The mineralization trend in tetracycline antibiotic was more than that of amoxicillin antibiotic. Here, the lack of complete degradation and COD is due to various intermediates that are not easily oxidized compared to the main compounds.[28]

In this study, the experiment was evaluated using carbon oxidation state index (COS). (Equation 5). Their values ranged from -4 to +4 for carbon dioxide as the most oxidized C states.[25]

$$COS = 4 - 1.5\left[\frac{COD}{TOC_i}\right] \quad (5)$$

Performance of PS/Fe2+/UV System for a Real Wastewater Sample

Figure 9 shows the efficiency of the PS/Fe^{2+}/UV process in removing antibiotics from real hospital wastewater samples. At this stage, after preparing a

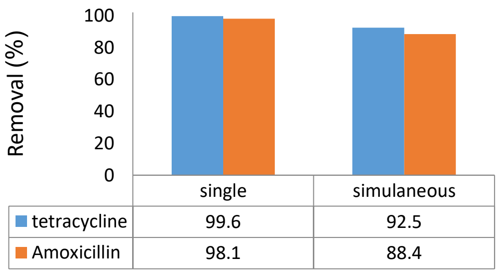

Figure 7. Efficiency of the PS/Fe^{2+}/UV Process in Simultaneous Removal of Tetracycline and Amoxicillin Antibiotics Under Optimal Conditions During 60 Minutes

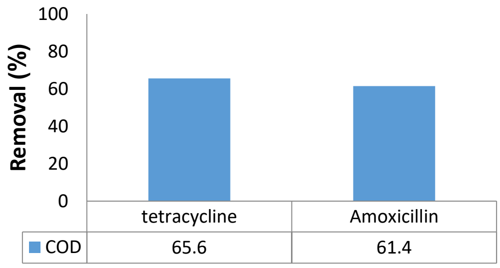

Figure 8. Mineralization Index Values of Tetracycline and Amoxicillin Removal by the PS/Fe^{2+}/UV Process Under Optimal Conditions During 60 Minutes

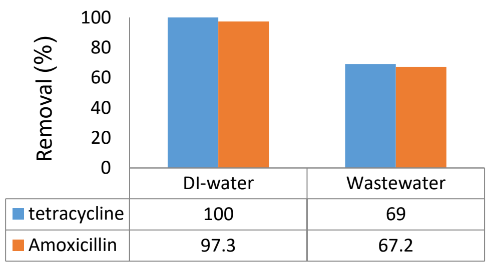

Figure 9. Comparison of the Removal Rate of Tetracycline and Amoxicillin Antibiotics by the PS/Fe^{2+}/UV Process in Synthetic and Real Wastewater Samples

real wastewater sample from the hospital's refinery, some relevant chemical characteristics were identified. It was observed that this wastewater sample contains very low concentrations of tetracycline and amoxicillin antibiotics. Therefore, in order to evaluate the removal of tetracycline and amoxicillin through the PS/Fe^{2+}/UV system in a real wastewater sample, the concentration of both antibiotics was set to 10 mg/L in real wastewater samples. All tests of artificial samples were performed under optimal conditions (Table 2). Based on this, the removal percentage of tetracycline and amoxicillin in DI water solution was measured as 100 and 97.3, respectively. The removal percentage of antibiotics tetracycline and amoxicillin in the real wastewater sample was 69% and 67.2%, respectively, which are less than the laboratory sample. It is the dissolved and solid substances that reduce the efficiency of the system. According to the obtained results, it is clear that the PS/Fe^{2+}/UV system shows complete efficiency in removing antibiotics in a real wastewater sample.

Figures 10-12 show the effect of cell retention time on

Table 2. Optimal Values of the Parameters Affecting the Performance of the PS/Fe^{2+}/UV Process in the Removal of Tetracycline and Amoxicillin

	pH	Fe^{2+}(mg/L)	PS (mM)	UV (W)	Antibiotic Removal Efficiency	COD Removal Efficiency
Tetracycline	4	0.3	5	8	100	65.6
Amoxicillin	4	0.3	5	8	99.7	61.4

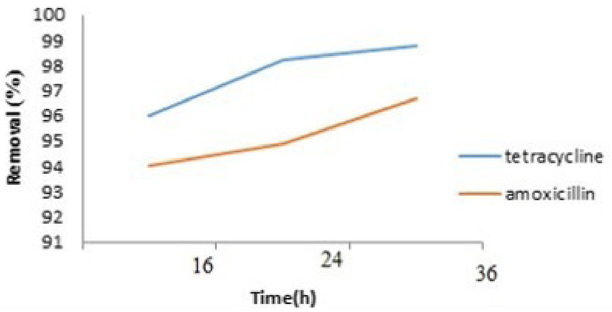

Figure 10. Removal Efficiency of Tetracycline and Amoxicillin Antibiotics with a Residence Time of 10 Days

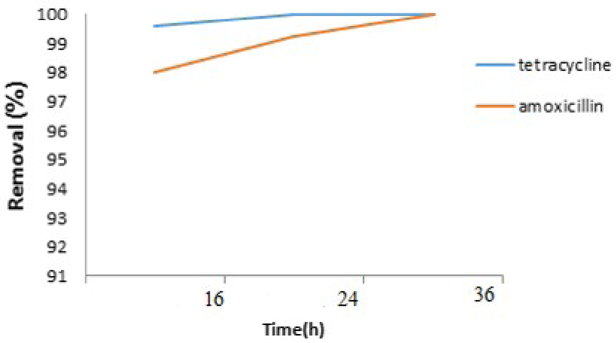

Figure 11. Removal Efficiency of Tetracycline and Amoxicillin Antibiotics with a Residence Time of 20 Days

Table 3. The Effect of Cell Retention Time on Various Parameters

	MLSS (mg/L)	MLVSS (mg/L)	SVI	F/M	BOD_5 Removal Percentage	COD Removal Percentage
Sludge age of 10 days	1490	830	86	1.1	97	95
Sludge age of 20 days	1840	995	110	0.32	99	98
Sludge age of 30 days	2700	1563	230	0.07	95	93

Abbreviations: COD, chemical oxygen demand; BOD, biochemical oxygen demand; MLSS, mixed liquor suspended solids; MLVSS, mixed liquor volatile suspended solids.

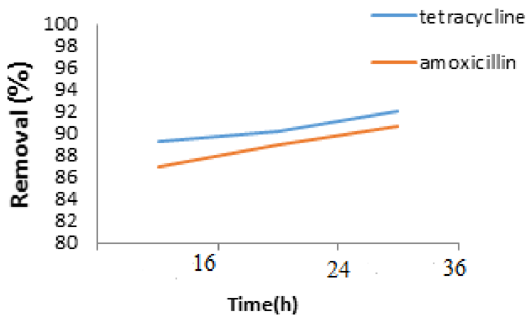

Figure 12. Removal Efficiency of Tetracycline and Amoxicillin Antibiotics with a Residence Time of 30 Days

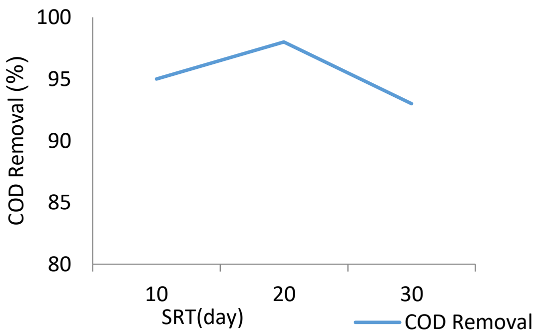

Figure 13. COD Removal Efficiencies During Different Cell Retention Times. Abbreviations: SRT, sludge retention time; COD, chemical oxygen demand

the removal efficiency of tetracycline and amoxicillin antibiotics. During the 20-day cell retention period, the removal efficiency is high for both antibiotics. The removal efficiency of tetracycline and amoxicillin antibiotics also increased while the reaction time was increasing as well.

The results show that the removal efficiency of antibiotics has increased and the removal efficiency of both tetracycline and amoxicillin antibiotics has decreased within 20 days. However, the cell retention time has increased from 10 to 20 days.

Figure 13 shows the effect of cell retention time (10, 20 and 30 days) on COD removal efficiency. In the initial stages of the process, there was an increasing trend in COD removal. With a cell retention period of 20 days, COD removal efficiency reached 98%. However, following this period, a decreasing trend in COD removal was observed. The COD removal efficiency reached 98% due to the microorganisms being in a reduced or stable growth phase.[14] During this stage, the sludge volume index (SVI) was calculated as 110 mg/L (Table 3), and the sludge settled effectively. During the 30-day cell retention period, a balance between filamentous and floc-forming microorganisms resulted in clear effluent. However, as the F/M ratio became very low, filamentous bacteria experienced high growth, leading to an SVI of 230 mg/L, and less effective sludge settling. Takdastan et al also observed that COD, BOD, and nutrient removal efficiencies increase significantly as sludge age increases to a certain level.[29]

As indicated in Table 3, an increase in sludge age (10, 20, and 30 days) leads to an increase in the SVI and the concentration of mixed liquor volatile suspended solids (MLVSS) (mg/L). This increase can be attributed to the growth of filamentous microorganisms. However, as the cell retention time increases, the F/M ratio decreases as the microorganisms enter their feeding phase. This decrease in concentration can be attributed to microbial activity and the utilization of carbon and nitrogen as energy sources. It indicates that the decomposition of organic matter is effectively taking place. With an increase in the sludge age, the percentage of COD and BOD_5 removal reaches approximately 92%.

More than 90% of the remaining microorganisms are organic matter, and when they increase, the percentage of sludge water increases. In other words, the dry weight percentage of sludge decreases because the cell retention time increases.[30]

Conclusion

Based on the findings, the applied process was highly dependent on the pH of the solution. To use 0.3 mg/L of iron caused a higher removal percentage of antibiotics. In addition, with the increase in the concentration of primary antibiotics, the percentage decreased significantly. When the amount of PS in the solution was increased, the system demonstrated an enhanced ability to remove antibiotics from the aqueous solution. Also, a good performance was observed for the PS/Fe^{2+}/UV system to remove antibiotics in a real wastewater sample, which resulted in an acceptable performance. According to the results, the removal efficiency by the combined biochemical process was 100% for the antibiotic tetracycline and 99.6% for the antibiotic amoxicillin after 20 days of cell retention and 36 hours of aeration. The combined process of PS / Fe^{2+}/ UV along the biological reactor was an effective method of removing antibiotics and its mineralization for the purification of the wastewater containing tetracycline and amoxicillin. Reuse of the method is also cost-effective for a short time before chemical oxidation treatment which is affordable.

Authors' Contribution

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Competing Interests

None.

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