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Disinfection process intensification of treated municipal wastewater employing peroxymonosulfate-ultraviolet advanced oxidation process and simultaneous amoxicillin micropollutant removal

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ABSTRACT

Over the last years, novel disinfection processes for municipal wastewater treatment plant effluents have attracted attention from researchers. This issue was because of the global demand for reusable water. There are critical concerns about conventional disinfection processes, which cause more serious problems, that is, the effluent quality fluctuations and existing contaminants of emerging concern in the effluent that spread into the environment. In the present study, a primary analysis has been accomplished for the degradation of amoxicillin using the removal of emerging pharmaceutical contaminants. In this regard, sulfate radical-based advanced oxidation processes (SR-AOPs) are very efficient for disinfection purposes and generate highly reactive free radicals. The present study describes the advances in disinfection methods based on SR-AOPs activation by ultraviolet (UV) radiation. Besides, the results of the most relevant papers are analyzed to show the widespread use of UV-AOPs as a novel disinfection technology for a broad class of contaminants. Also, sulfate radicals ($SO_4^{\bullet-}$) are generated from UV-peroxymonosulfate (UV/PMS) and UV-persulfate (UV/PS). The achieved results have been compared with other processes. The findings demonstrated that the UV/PMS process was more effective than the other mentioned processes. Also, fecal coliforms reduction and amoxicillin removal efficiencies were 99.99 (e.g., the total coliforms reached less than 400 MPN consistently in 100 mL) and 94% ± 3%, respectively. Finally, other important effluent quality parameters were considered in the present study.

Keywords: Novel municipal wastewater disinfection processes; Amoxicillin; Peroxymonosulfate (PMS); Persulfate (PS); Ultraviolet (UV)

1. Introduction

Water management is a necessary process because of the crucial role of water resources in human life [1]. In recent years, wastewater has been discharged in large quantities without safe disinfection due to changing lifestyles, life quality, and economic growth [2]. It is estimated that about 2.1 billion people have no access to healthy and safe drinking water in the world. Besides, diseases transmitted by contaminated water are responsible for numerous deaths [3]. These diseases arise from the excessive release of pathogenic bacteria and microorganisms into natural water resources. Also, the European Environment Agency (EEA) reported that the chemical contamination of surface water resources was generally caused by urban wastewater discharge. In addition, treatment plants receive many ever-changing contaminants from various sources (e.g., urban, industrial, and hospitals). These contaminants can be transmitted to surface water resources if not removed in treatment by effective disinfection. Therefore, it is essential to disinfect the wastewater of treatment plants as a major source of pathogenic bacteria. Although previous studies demonstrated that wastewater

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could be an efficient alternative to control water and reduce clean water unavailability [4,5], reusing the treated wastewater for urban and agricultural purposes is a part of integrated extractable water resource management. Hence, the necessity of efficient and ensured disinfection of urban wastewater treatment plant effluent is obvious [6,7].

Contaminants of emerging concern (CECs) have attracted attention from researchers in recent years. This issue was because of the large CECs quantities within urban wastewater, leading to serious environmental and health threats [8,9]. This situation proves the necessity of enhancing disinfection knowledge and technology to disinfect and reuse wastewater optimally and safely so that CECs would no longer be a threat to the environment and water resources [10,11]. In this regard, medications and antibiotics account for a portion of CECs. Since humans use large quantities of medications and antibiotics, these CECs rapidly raised in urban treatment plans. They may be transmitted to the environment due to the insufficient disinfection abilities of conventional treatment plants. Also, CECs can be substantial health and environmental threat if they are not effectively and efficiently managed [12].

Overall, CECs must be removed before discharging into the environment. Hence, it is necessary to exploit advanced oxidation processes (AOPs) as an efficient wastewater disinfection technique [13]. An AOP involves generating strong radicals that react unselectively and attack and destroy contaminants [14–17]. The previous studies have widely investigated AOP technologies. These technologies have recently been of great environmental and economic interest to researchers for disinfection efficiency improvement and CECs removal. However, the optimal selection of AOP is a challenging issue in disinfection [18]. Since AOPs involve various processes, they need further studies to augment wide applications [19].

Besides, ultraviolet (UV) light has been widely utilized to disinfect and reuse wastewater. It destroys DNA and subsequently impedes cell proliferation. The effectiveness of a UV disinfection system depends on wastewater, reactor, and exposure time and intensity. This technology is broadly employed to disinfect water and wastewater worldwide [20,21]. UV-based AOPs could effectively remove many contaminants, such as bacteria, viruses, fungi, algae, and protozoa [22,23]. Also, it has been reported to be a promising and effective technique for resolving CECs. Recently, various studies have investigated the use of UV-based AOPs to cope with CECs [24–26]. In this regard, many studies have developed the UV-activated peroxymonosulfate (PMS) technology as a highly efficient method for the degradation of CECs, including sulfadimethoxine (SDM) [27], tris(2-chloroethyl) phosphate (TCEP) [28], and flumequine (FLU) [29]. These researches demonstrated that UV-activated PMS could efficiently remove these contaminants in water and wastewater.

The present study utilizes AOPs disinfection to control pathogenic bacteria and CECs in wastewater. The UV-based AOPs performance has been evaluated by comparing the proposed processes with other techniques. According to the list of Decision 2018 published in June 2018, amoxicillin (AMX) had been known as a CECs [30]. Amoxicillin is a broad-spectrum β-lactam antibiotic (with a chemical formula of $C_{16}H_{19}N_3O_5S$ and a molecular weight of 365.4 g mol^{-1}), which belongs to the penicillin group. It is systematically employed for treating gastrointestinal bacterial infections in medicine and veterinary medicine [31]. Nowadays, the use of antibiotics is increasing, and many of them are discharged into the environment, especially by wastewater treatment plants [32]. Hence, the authors were motivated to measure the AMX elimination from the effluent. In this regard, sulfate radical-based advanced oxidation processes (SR-AOPs) are environmental-friendly and play a vital role in protecting the environment. These processes have a higher oxidation potential and longer half-life than hydroxyl radical-based advanced oxidation processes (HR-AOPs) [33,34]. The most common way of generating sulfate radical ($SO_4^{-\bullet}$) is to activate PMS and persulfate (PS). Since activating these substances through UV irradiation needs high UV energy, it will destroy a considerable range of contaminants [35–38]. In the present study, several technologies were employed, including UV-peroxymonosulfate (UV/PMS), UV-persulfate (UV/PS), UV/H_2O_2, and UV alone. Additionally, the removal efficiencies of parameters related to municipal wastewater treatment plant (WWTP) effluent quality (i.e., chemical oxygen demand (COD) and biochemical oxygen demand (BOD_5) have been investigated for the processes under the optimized reactor operational conditions.

2. Materials and methods

This section and its subsections provide the required information about the WWTP effluent features, experimental procedures, chemicals, and the proposed step-by-step approach for the analysis process.

2.1. Materials

2.1.1. WWTP effluent

The wastewater samples used in this study were taken from the effluent of a secondary settling tank in a municipal WWTP using an activated sludge system (Tehran, Iran). Table 1 gives some information about the secondary effluent quality.

2.1.2. Experimental setup

Disinfection experiments were performed on four processes, including UV/PMS, UV/PS, UV/H_2O_2, and UV. In the UV disinfection process, a 250-cc cylindrical reactor has been employed to conduct the studies. Accordingly, UV-C lamp (Hitachi Corporation, Japan) Model UV-6W with a peak wavelength of 254 nm and lamp length of 20 cm was used (Fig. 1).

2.1.3. Chemicals

Several types of chemicals were employed in the present study, encompassing potassium peroxymonosulfate (99%), potassium persulfate (99%), hydrogen peroxide (35%) made by Merck company, and amoxicillin by Sigma-Aldrich company.

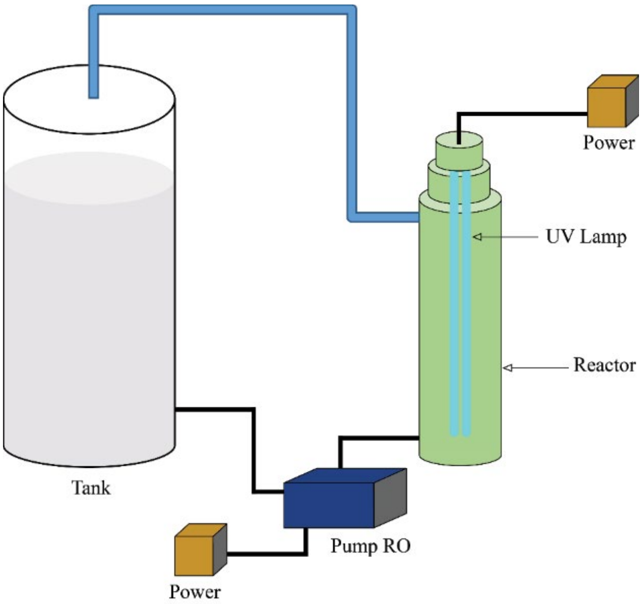

Fig. 1. Schematics of the test setup.

Table 1
Wastewater effluent quality parameters

Parameter	Average
pH	7.1 ± 0.2
	–
Total coliform, MPN/100 mL	(2.1 ± 0.3) × 10^6
Turbidity, NTU	10 ± 3
Chemical oxygen demand (COD), mg L^{-1}	34 ± 5
Biological oxygen demand (BOD_5), mg L^{-1}	20 ± 5
Total nitrogen (TN), mg L^{-1}	40 ± 5
Total phosphorus (TP), mg L^{-1}	10 ± 2
Temperature, °C	18 ± 1
Electrical conductivity (EC), µS cm^{-1}	715 ± 20
Total dissolved solids (TDS), mg L^{-1}	453 ± 50
Total suspended solids (TSS), mg L^{-1}	18 ± 5

2.2. Methods

The 5220D, 5210B, and 9221B standard methods have been used to measure the COD, biological oxygen demand (BOD_5), and total coliform, respectively [39]. Also, HANNA pH meter-211 device was utilized to measure pH.

According to the objective of the present paper, the authors have chosen several related studies and summarized some of the UV-based processes to show their efficiency in removing a wide range of micro-pollutants. Since the removal evaluation of total coliform bacteria as a bacteria log removal is beyond the scope of this work, we have reviewed and compared the two principal methods (i.e., UV/PMS and UV/PS) with one of the latest developments in wastewater. Although a primary investigation has been performed on the AMX removal, it is not discussed in this paper and needs more investigations in future studies. In addition, the JENWAY 6315 UV-spectrophotometer at a wavelength of 228.3 nm was used to measure the amoxicillin concentration [40]. The initial concentration of amoxicillin was 36.5 mg L^{-1} because the concentration of more than 36.5 mg L^{-1} causes inhibitory for the test organisms [41]. It is essential to note that this study is based on amoxicillin dosage. The best performance of AMX occurs at the wavelength of 228.3 nm, and amoxicillin's transformation products must be considered in future studies. Also, amoxicillin was added synthetically to the samples.

3. Results and discussion

In this study, the treated wastewater effluent disinfection and amoxicillin residues removal were investigated using UV/PMS, UV/PS, UV/H_2O_2, and UV alone to secure availability for reuse purposes. In particular, the principal processes were UV/PMS and UV/PS methods that are SR-AOPs.

3.1. Reduction of total coliforms

The effects of using UV/PMS, UV/PS, UV/H_2O_2, and UV alone have been evaluated on the wastewater effluent disinfection. Among all microbe and bacteria, the total coliforms have been identified as an effective indicator of wastewater effluent disinfection. An allowable limit for the total coliforms (e.g., 400 MPN in 100 mL) was specified as the criterion for successful disinfection. Consequently, the allowable limit criterion for the fecal coliform is met according to the wastewater reuse standards for agricultural purposes. The experiments were performed for all methods in the same conditions at a temperature of 17.3°C, pH = 6.9 ± 0.1, constant UV dosage rate of 1.7 × 10^4 µW cm^{-2}, and the maximum time of 30 min. If only UV participated in the reaction, the number of total coliforms did not meet the allowable limit, and after the reaction time of 30 min, it just reached from 2,100,000 to 3,965 MPN/100 mL. In UV/PMS method, several peroxymonosulfate dosages were considered, including 0.03, 0.06, and 0.09 mmol L^{-1}. In this method, the productions of hydroxyl and sulfate radicals by UV radiation are carried out based on Eq. (1) [42]. It is necessary to note that an increase in the PMS dose enhances the total coliform removal. Fig. 2 depicts the allowable limit for the reactor to reach a value less than the maximum value of 400 coliforms in 100 mL. In UV/PS method, the PS dosages of 1, 2, and 3 mmol L^{-1} were used. As shown in Fig. 2, only 3 mmol L^{-1} reached the allowable limit. In addition, the UV/H_2O_2 method was performed in the dosages of 0.35, 0.70, and 1.05 mmol L^{-1}. Furthermore, the efficient amounts of removal are shown in Fig. 2.

$$HSO_5^- + h\nu \rightarrow SO_4^{-\bullet} + {}^{\bullet}OH \quad (1)$$

Table 2 gives the resulting inactivation of total coliform in log-inactivation. According to this table, the optimum operating conditions of each reactor to reach the maximum allowable coliform number of 400 in 100 mL are expressed. In UV/PMS process, the optimum operating condition is obtained in log (MPN) of 2.45 (e.g., 295 MPN/100 mL < 400) at 20 min and 0.06 mmol L^{-1} PMS dosage. These situations

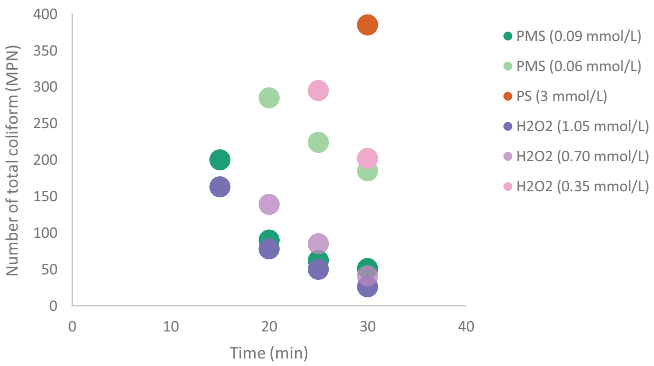

Fig. 2. The effect of different dosages of PMS, PS and, H_2O_2 under UV irradiation on the removal of total coliform.

provided the best result. Inactivation states with UV/PS and UV/H_2O_2 processes have been achieved in 2.59 (385) and 2.47 (300) at PS and H_2O_2 dosages of 3 and 0.35 mmol L^{-1} and reaction times of 30 and 25 min, respectively. In addition, this table presents the results of one of the latest studies on removing total coliform in wastewater. In [43], the authors reported that all the UV-based processes were the most effective case for removing total coliform. Indeed, they result in a complete inactivation.

The energy efficiencies of the AOPs have been evaluated by estimating electrical energy per order (EEO) in all processes. EEO is the required electrical energy to reduce the concentration of contaminants in wastewater. EEO has been calculated using the following equation:

$$\frac{EE}{O} = \frac{P \cdot t \cdot 1{,}000}{V \cdot \log\left(C_0 / C_f\right)} \quad (2)$$

where P is the power (kW) of UV, V is the volume (L) of wastewater, and t (min) is reaction time. Also, C_0 and C_f are initial and final concentrations of total coliform, respectively [44,45].

Among all processes, the EEO value in UV/PMS process had the lowest performance for removing total coliform (3.1 kWh m^{-3} $order^{-1}$). Overall, the energy efficiency of processes follows the order of UV/PMS > UV/H_2O_2 > UV/PS > UV alone.

3.2. Amoxicillin removal

This section briefly explains the UV-based advanced oxidation technologies to analyze the efficiency of UV-based processes. Table 3 summarizes some of the developments in UV-based advanced oxidation technologies, which have been used for removing amoxicillin.

Mahdi et al. [46] particularly reviewed the photo-Fenton process for the degradation of antibiotics and amoxicillin. It included H_2O_2 and iron salts with ultraviolet radiation. In this system, UV has been used to accelerate Fenton reactions by generating $^{\bullet}OH$ radicals. This procedure needed an acidic pH to provide higher efficiency. According to the literature, it is an economical method so that its chemical compounds are available and cheap. Besides, the photo-Fenton process had high efficiency in removing amoxicillin. The amoxicillin degradation reached 67% and 85% in 1 and 2 mmol L^{-1} H_2O_2 concentrations, respectively. In [47], the efficiencies of three UV/AOP processes have been investigated for amoxicillin degradation. The findings demonstrated that UV alone system had an insignificant amoxicillin degradation. While UV/PS and UV/H_2O_2 systems significantly increased the amoxicillin degradation efficiency. This situation occurred due to the generation of two free radicals. Besides, the results showed that UV/PS, UV/H_2O_2, and UV methods had the highest efficiency, respectively. Also, H_2O_2 in amoxicillin degradation was more cost-effective than PS. In addition, the outcomes showed that these processes were efficient approaches for disinfecting contaminants. It is essential to note that photo-based processes such as heterogeneous photocatalysis have been known as a green technology that gained attention due to their potential applications in environmental remediation.

The importance of removing specific pollutants with a pharmaceutical origin has been investigated by analyzing amoxicillin removal, which is one of the commonly residual drugs in wastewater. However, this issue needs more evaluations in future studies. The amoxicillin concentration of 36.5 mg L^{-1} has synthetically been added to the samples. The abilities of the proposed methods to remove amoxicillin were evaluated under optimum operating conditions. UV/PMS, UV/PS, and UV/H_2O_2 methods reduced amoxicillin levels by 94%, 78.5%, and 74.5%, respectively. This reduction was in addition to the total coliform reduction to the standard limit. Also, UV alone reduced the amount of amoxicillin by 61.4%. It is necessary to note that these percentages are the average removal efficiency in the experiments. This issue is illustrated in Fig. 3.

The results showed that UV irradiation alone could negligibly destroy organic pollutants. The degradation efficiency of contaminants was improved by combining PMS with UV. Thus, this combined system worked more efficiently. Also, the generation of $SO_4^{-\bullet}$ and $^{\bullet}OH$ free radicals enhanced the micropollutants' degradation. In addition, previous studies demonstrated that UV/PMS-based processes were developed, and they were primary steps for applying on different scales [35].

3.3. Effluent characterization

The effects of using UV/PMS, UV/PS, UV/H_2O_2, and UV have been investigated on the wastewater effluent qualitative parameters (i.e., COD and BOD_5) under optimum disinfection conditions. The attained results are indicated in Fig. 4.

As shown in Fig. 4, the UV/PMS method had the highest performance. Also, it significantly disinfected total coliform and reduced amoxicillin as an emerging contaminant. In this process, radicals of hydroxyl and sulfate act as oxidation agents. Therefore, an increase in the production of these agents enhances the removal of contaminants.

The PMS consumption value was very low in this study. In the UV/PMS system, the PMS dosage was 0.06 mmol L^{-1} (9.132 g PMS in 1 m^3) under optimum conditions. Also, the

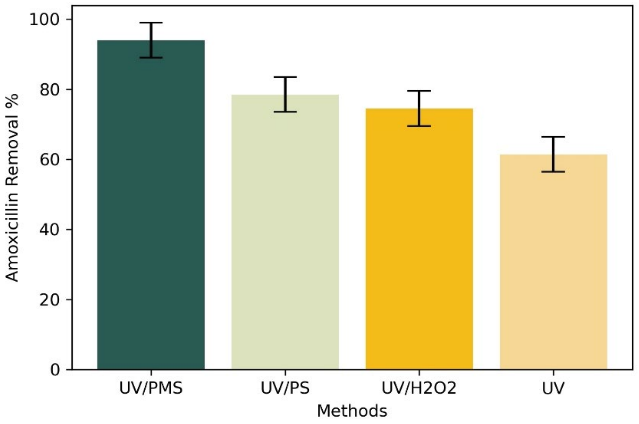

Fig. 3. The effect of UV/PMS, UV/PS, UV/H_2O_2 and UV processes on the removal of amoxicillin.

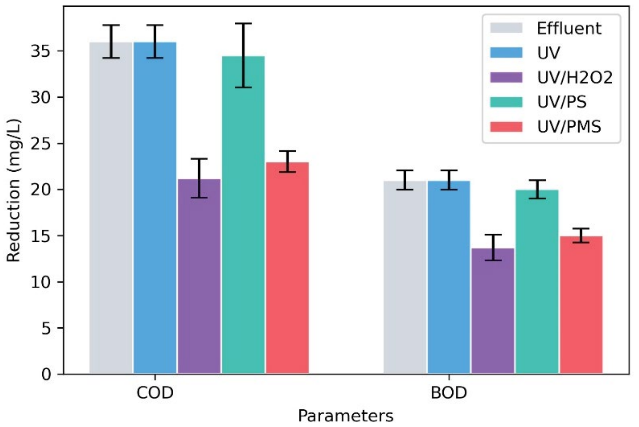

Fig. 4. The effect of methods on characteristics of the treated wastewater sample.

PMS consumption cost was 27.4 (unit cost of cents) in this system. It is essential to note that the disinfection process efficiency improvement depends fundamentally on the WWTP operational conditions and effluent characteristics. Moreover, UV/PMS method provided considerable efficiency in COD and BOD_5 removal compared to other investigated methods.

Table 2
Log-inactivation of total coliform by UV-AOPs

Process	Total coliform
UV/Cl_2	3.8
UV/H_2O_2	
UV/O_3	
UV/Cl_2/O_3	
UV/H_2O_2/O_3	
UV/PMS	2.45
UV/PS	2.59
UV/H_2O_2	2.47

3.8 = log-inactivation related to the complete elimination under different operating conditions [41].

Table 3
UV-based AOPs for removing amoxicillin

Process	Operation conditions	Reference
Photo-Fenton	pH = 2.5 UV (15 W) = 365 nm H_2O_2 = 1–2 mmol L^{-1} AMX = 3.21 mg L^{-1}	[46]
UV/PS UV/H_2O_2 UV	pH = 7 T = 40°C UV (5 W) = 254 nm H_2O_2 = PS = 0.5 mmol L^{-1} AMX = 0.64 mg L^{-1}	[47]
UV/PMS UV/PS UV/H_2O_2 UV	pH = 6.9 ± 0.1 T = 17.3°C UV (6 W) = 254 nm H_2O_2 = 0.35 mmol L^{-1} PS = 3 mmol L^{-1} PMS = 0.06 mmol L^{-1} AMX = 36.5 mg L^{-1}	

4. Conclusions

Recently, researchers have paid special attention to the reliable usage of municipal WWTP effluents in agriculture and greenspace irrigation in arid and semi-arid areas. In this regard, optimum wastewater discharge disinfection is one of the principal reuse prerequisites. In other words, there are critical concerns about CECs and their presence in the wastewater effluent. Indeed, the incapacity of WWTP disinfection technologies is a crucial issue and needs to be considered in novel technologies. UV is one of the most environmentally friendly technologies used for disinfection. This technology has significant advantages over the other methods. In the present study, the treated wastewater disinfection and residual amoxicillin micro-pollutant removal were simultaneously investigated using the UV/PMS method. This method was compared with the conventional UV-radiation technique and other advanced oxidation processes. Also, UV/PS and UV/H_2O_2 methods have been considered to evaluate their capability. The results demonstrated that the UV/PMS method was a proper approach to achieving a more reliable solution in disinfection process improvement and elimination of emerging contaminants. Therefore, the UV/PMS method is proposed as a new solution for emerging pharmaceutical contaminants that need further research. Moreover, a complementary study is performed on the possible by-products, including sulfur compounds in the process on a semi-industrial scale. This procedure is carried out through the UV/PMS method in wastewater treatment plants.

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