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Article

Enhanced Degradation of Sulfonamide Antibiotics by UV Irradiation Combined with Persulfate

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Abstract: In this study, the degradation of sulfonamide antibiotics was investigated through persulfate-enhanced UV advanced oxidation process. Factors that may affect the degradation efficiency were analyzed. Results showed that the persulfate imposed a significant enhancement on the UV oxidation process during the sulfathiazole degradation. The combined process of UV/persulfate can effectively remove about 96% of sulfathiazole within 60 min. With the increase in the dosage of persulfate, the removal efficiency increased as well. Different water matrix almost had no effect on the removal efficiency. Two intermediates were found during the sulfathiazole degradation. It can be predicted that the combined process of UV/persulfate has a broad application prospect for removing sulfonamide antibiotics in water treatment.

Keywords: sulfonamide antibiotics; UV irradiation; persulfate; advanced oxidation process; removal efficiency



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1. Introduction

In actual water, the environmental behavior of pharmaceuticals and personal care products (PPCPs), especially the existing level and source of antibiotics from them, are the hot spots and frontiers in ecotoxicology research around the world [1]. Among these PPCPs, sulfanilamide antibiotics (SAs) have attracted much attention due to their widespread usage in human medical treatment, animal husbandry, and aquaculture, and have entered the environment through industrial and domestic sewage discharge [2,3]. SAs mainly include sulfathiazole (STZ), sulfadiazine (SDZ), sulfamethoxazole (SMX), sulfamerazine (SMZ), and so on. Antibiotics have been detected in soil, lakes, rivers, groundwater, river sediments, and even in the urine of school-age children in countries all over the world [4,5]. Antibiotic residues of different concentration levels have been detected in pharmaceutical wastewater, livestock breeding wastewater, surface water, and even drinking water [6–8]. SAs are prone to show “false” persistence in the environment. Their long-term exposure can not only inhibit some environmental microorganisms but also induce bacterial resistance, which is harmful to the ecosystem and human health [9]. However, the structure and properties of SAs are stable and hard to be degraded, and the traditional wastewater treatment process cannot remove them efficiently [10]. Therefore, it is a hot topic of current concern and research to realize the degradation of these emerging pollutants quickly and effectively.

At present, the main methods to treat the water polluted by SAs include physical adsorption method, microbial method, and chemical oxidation method. Akhtar et al. [11] used powdered activated carbon as adsorption material to remove sulfamethoxazole. The results showed that the adsorption rate reached 73% after 5 h when 2 g/L of powdered activated carbon was added. Degradation is the main way to remove sulfonamides in the

environment, and ammonia-oxidizing bacteria are the important bacteria to degrade sulfonamides. However, little is known about the degradation mechanism of sulfa antibiotics by ammonia-oxidizing bacteria.

Advanced oxidation process (AOPs) provides an effective way to solve the environmental problem because of its strong oxidizing characteristics. The AOPs technology with hydroxyl radical ($\bullet\text{OH}$) as the oxidation active species can finally oxidize these persistent organic pollutants into CO_2 and H_2O efficiently and non-selectively realized the elimination of these pollutants completely [12,13]. According to the literature review, the AOPs methods for the degradation of sulfanilamide antibiotics mainly contain photocatalysis [14], ozone [15], Fenton-like [16], free chlorine [17], and ferrate [18]. Photocatalysis is a hot spot in the research of water treatment at present, but when it comes to the actual wastewater treatment, the efficiency of photocatalysis still needs to be improved. Although the ozone oxidation technology has high oxidation efficiency and only needs simple operation, the treatment cost is hard to be called cost-effective. Therefore, a breakthrough in efficient and practical wastewater treatment technology has been the focus in the field of advanced oxidation technology. A new oxidation removal technology of persistent organic pollutants by the AOPs technology is studied on the basis of the sulfate radical ($\text{SO}_4^{\bullet-}$) theory, which is of great development potential that has been developed recently in China [12,19–21]. With the advantages of simplicity, no secondary pollution, and high efficiency, sulfate radical based-AOPs technology shows a wide potential applied in organic wastewater treatment. Compared with $\bullet\text{OH}$, the life span of $\text{SO}_4^{\bullet-}$ is longer (half-life is 4s), and the standard redox potential is 2.5V, which is similar to that of $\bullet\text{OH}$. It is found that under neutral conditions, the redox potential of $\text{SO}_4^{\bullet-}$ is even higher than that of $\bullet\text{OH}$, which can completely degrade and remove most of the organic pollutants that have been applied more and more in water pollution control research [22–24].

Persulfates contains peroxymonosulfate (PMS, HSO_5) and peroxodisulfates (PDS, $\text{S}_2\text{O}_8^{2-}$). They have a limited oxidation ability but can be activated to produce $\text{SO}_4^{\bullet-}$ under the conditions of light, heat, sound, and transition metal ions. For example, Zhang et al. used UV/PDS to degrade sulfonamides in wastewater and urine [25]; Yin et al. used thermal activated PDS to study the oxidation process of sulfonamides [26]. The transition metal activation is widely studied due to the moderate reaction condition of room temperature and normal pressure without additional energy. It has been reported that there are homogeneous catalysts based on $\text{SO}_4^{\bullet-}$, such as cobalt(II), manganese(II), iron(II), etc. [27,28]. However, there are some problems in this activation method such as the toxicity of metal catalysts, reuse, and secondary pollution to the environment, and so on. Heterogeneous catalysts such as Fe_3O_4 have become a research hotspot, but various research results show that the heterogeneous catalysts such as metals, metal oxides, or supported metal oxides developed by researchers cannot avoid complete metal leaching [29–31].

In this study, the ultraviolet process, peroxodisulfates process, and the integrated ultraviolet and peroxodisulfates process (UV/PDS) were used to remove the sulfonamide antibiotics for comparison. The influencing factors in these degradation processes were investigated. This study may provide an effective way to degrade the SAs in water.

2. Experiment

2.1. Reagents

Sulfadiazine (SDZ), sulfathiazole (STZ), sulfamethoxazole (SMX), sulfamerazine (SMZ), persulfate (PDS), NaCl, NaNO_3 , Na_2SO_4 , NaHCO_3 , KH_2PO_4 , and NaOH were purchased from Aladdin (Shanghai, China). Humic acid (HA) was from Maclin Biochemical Co. (Shanghai, China). All chemicals in this work are AR grade. Ultrapure water (18.2 m Ω cm) was obtained from Millipore (Burlington, MA, USA).

Instruments: UV lamp (88-1) from Changzhou Guohua Instrument Co., Ltd. (Changzhou, China), UV spectrophotometer purchased from Shanghai Mapada Instrument Co., Ltd.

(Shanghai, China), and SHZ-A water bath constant temperature oscillator (GTCS-2016) from Shanghai Bosun Medical Biological Instrument Co., Ltd. (Shanghai, China).

2.2. Methods

The glass dish is used as the reaction vessel, and the ultraviolet light source is provided by the ultraviolet lamp with the power of 8 W, and the light intensity is adjusted to be 2 mW/cm^2 . One hundred milliliters solution of SAs (10 mg/L) was prepared with ultrapure water, then added 300 mg/L of PDS. The mixed solution was stirred with a magnetic stir bar at 800 rpm. One milliliter of the reaction solution was sampled with a pipette at different reaction times, and 1 mL of methanol was added for terminating the reaction. During the experiment, the initial concentrations of PDS were adjusted from 150 mg/L to 600 mg/L. The absorbance of SDZ, SMX, SMZ, and STZ was measured at 270, 269, 270, and 285 nm, respectively. The degradation intermediates were detected by a Shimadzu LC-30A HPLC (Kyoto, Japan), which connected with an AB Sciex Triple TOF 5600 MS (Framingham, MA, USA).

3. Results and Discussion

3.1. The Effects of Oxidation Processes on STZ Removal

STZ was degraded by ultraviolet (UV), peroxodisulfates (PDS), and UV/PDS combined process, respectively. The experimental results are shown in Figure 1. From Figure 1, the removal efficiency of STZ within 60 min was about zero by a single PDS process. PDS is relatively stable, thus the SAs cannot degraded by PDS oxidation process only [32]. UV process was used as oxidant for free radical production, but only about 72.6% of STZ could be removed by UV irradiation after 60 min. These results indicated that the removal efficiency of STZ by a single UV process or single PDS process was limited within 60 min. On the contrary, about 96% of STZ can be removed by UV/PDS combined process after 60 min, indicating the combined process of UV/PDS is effective. This could be due to that PDS can be activated by UV and produce $\text{SO}_4^{\bullet-}$ as following Equation (1), which has a high oxidation activity and longer half-life than $\bullet\text{OH}$, thus the combined process improved the removal efficiency of SAs.

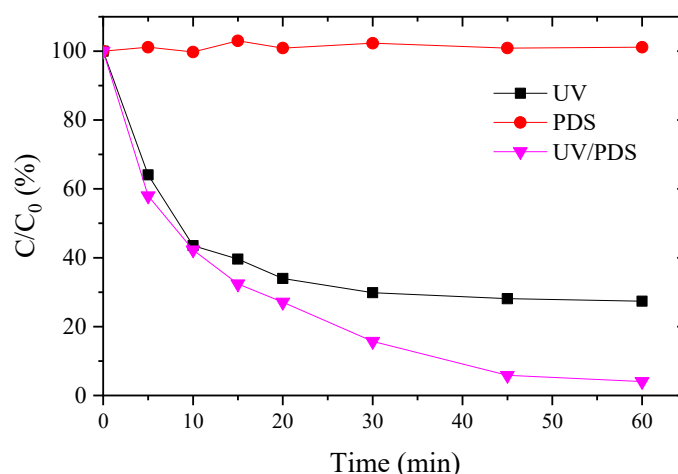


Figure 1. The effects of different oxidation processes on sulfathiazole (STZ) removal. Conditions: [STZ] = 10 mg/L, [peroxodisulfates (PDS)] = 300 mg/L and UV (ultraviolet) irradiation: 2 mW/cm^2 .

3.2. The Effects of PDS Amount on STZ Removal

The influences of PDS amount on STZ removal were investigated and the results are shown in Figure 2. With the increase in the amount of PDS, the degradation efficiency of STZ increased within 60 min, which disclosed that STZ removal obviously increased

when the amount of oxidant increases. This is probably because the increase in the PDS concentration increases the concentration of $\text{SO}_4^{\bullet-}$ and $\bullet\text{OH}$, enhancing STZ removal. When the amount of PDS was 300 mg/L, the removal efficiency of STZ reached 95.9%, which was basically the same as 98.6% when the amount increased to 600 mg/L. Therefore, 300 mg/L of PDS was selected for further experiments.

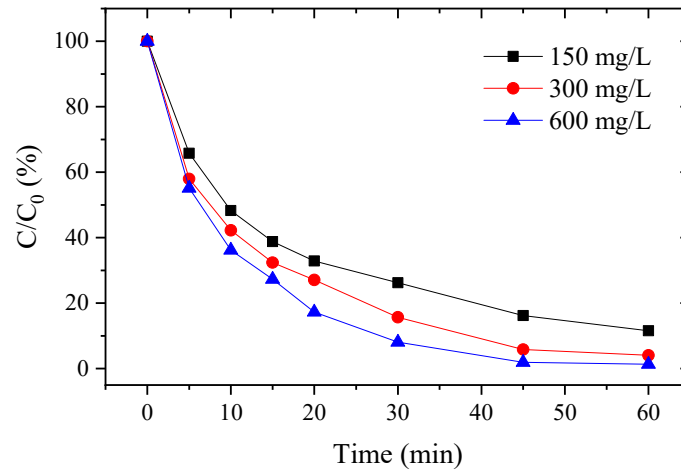


Figure 2. Effects of PDS amount on STZ removal. Conditions: [STZ] = 10 mg/L, [PDS] = 150–600 mg/L and UV irradiation: 2 mW/cm².

3.3. The Effects of Initial Concentrations of STZ on its Removal

Figure 3 shows the UV/PDS performance for the degradation at different initial concentrations of STZ. The residue ratio decreased with the time passes, and the residue ratio fell in the range from 4.1% to 28.6% at the concentration ranged between 10 and 40 mg/L. Thus, the higher the concentration of STZ, the harder the removal. This can be attributed to the higher the pollutant concentration is, the more free radicals are needed for pollutant removal during the reaction time.

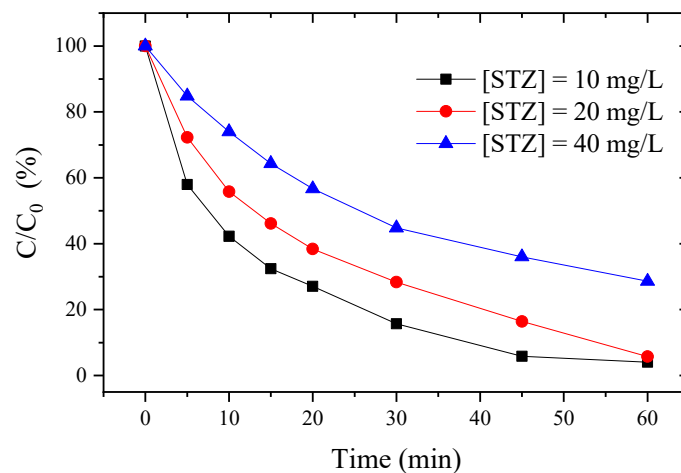


Figure 3. Effects of STZ concentration on STZ removal. Conditions: [STZ] = 10–40 mg/L, [PDS] = 300 mg/L and UV irradiation: 2 mW/cm².

3.4. The Effects of Anions on STZ Removal

There are many impurities in natural water, and it is necessary to simulate the effects of different water quality for STZ removal [33]. Therefore, the influences of different anions on STZ removal were also studied. Common anions, such as Cl^- , NO_3^- , SO_4^{2-} , HCO_3^- , and H_2PO_4^- , were selected to study their effects on the degradation of STZ. NaCl, NaNO₃,

Na_2SO_4 , NaHCO_3 , and NaH_2PO_4 were added into the solution to make the concentration of each anion at 5 mmol/L.

The results in Figure 4 show that various anions had a different extent of influence on the removal efficiency. Anions like NO_3^- and HCO_3^- had certain inhibition effect. NO_3^- is easily photolyzed to NO_2^- and O^{2-} , which in turn combine with H^+ to form $\bullet\text{OH}$, but NO_2^- may also be the scavenger of $\bullet\text{OH}$ [34,35]. The results show that nitrate may also play a role in scavenging hydroxyl radicals in the system that inhibited the degradation rate most. HCO_3^- reacting with sulfate radical can produce bicarbonate radical in solution, and $\bullet\text{OH}$ reacting with HCO_3^- can also produce $\text{CO}_3^{\bullet-}$. Although the free radical of $\text{CO}_3^{\bullet-}$ is also an oxidant, its activity is not as good as $\bullet\text{OH}$ [36], thus the removal rate was decreased a little. Radicals like $\text{Cl}\bullet$, $\text{Cl}_2^{\bullet-}$ and $\text{ClHO}\bullet$ can be produced by the reaction between sulfate radicals and chloride ions that cause the increase in STZ removal rate [37]. Sulfate itself is the ion produced by the catalyst, so it will accelerate the reaction rate slightly. It is well known that more SO_4^{2-} means more reactive oxygen species (ROS), resulting in an increase in the degradation rate. Generally, common ions in water inhibit the STZ removal. The removal efficiencies of STZ by the UV/PDS process were still as high as 91.1–99.3% after a 60 min-reaction time, which indicates that the UV/PDS process has a potential application in the water treatment field.

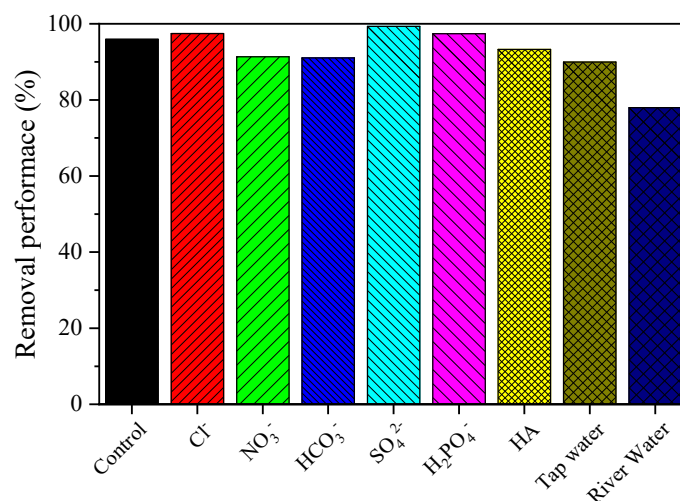


Figure 4. Influence of anions, humic acid (HA), and natural water on STZ removal. Conditions: [STZ] = 10 mg/L, [PDS] = 300 mg/L, UV irradiation: 2 mW/cm², [anions] = 5 mM and [HA] = 10 mg/L.

HA can promote the reaction at low concentration. Previous publications have reported that HA can not only promote the photodegradation of pollutants as photosensitizer but also inhibit the photodegradation of pollutants as a photomasking agent or free radical trapping agent [38], and its effect on the photolysis of antibiotics shows a dual effect. In Figure 4, the STZ removal efficiency was 93.3% after a 60 min-reaction time, indicating that 10 mg/L of HA has no inhibitory influence on STZ degradation.

The actual water condition is complex, so it is particularly important to study whether the degradation of SAs will be affected. To determine the removal efficiency of the combined process for the removal of sulfanilamide antibiotics in actual water, 10 mg/L of STZ was also prepared using tap water and surface water, and similar removal experiments were conducted. In Figure 4, STZ removal in tap water reduced to about 90.0% after a 60 min reaction time, which is basically consistent with that in ultrapure water. The removal efficiency of STZ in surface water was slightly lower, but removal efficiency could reach around 76.1% after a 60 min-reaction time. The removal efficiency was lower than that of ultrapure water, which indicated that some ions reacted with free radicals in the two kinds of water, reducing the number of free radicals. The above results reveal that the

UV/PDS process has good performances for STZ degradation not only in model water but also in actual water; therefore, it has a wide potential to be applied in wastewater treatment practice.

Figure 5 shows the possible degradation pathways of STZ, mainly including the oxidation and fracture of thiazole substituted ring to form TP216 and TP218 products. Finally, these intermediates can be further degraded into small molecular products.

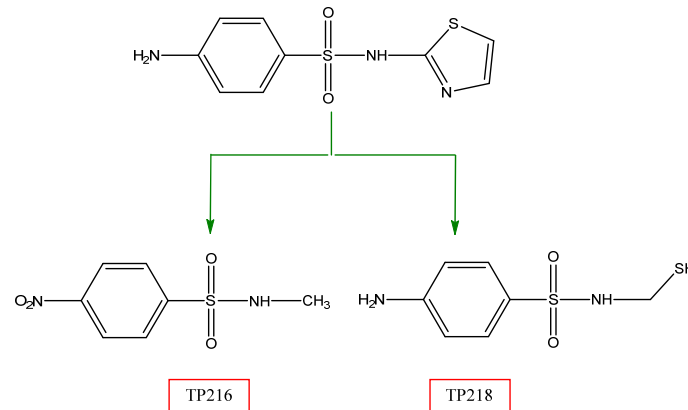


Figure 5. Degradation pathways of STZ by UV/PDS process.

3.5. The Oxidative Process for Other SAs Removal

In the previous experiments, it was found that UV/PDS is effective in degrading STZ, so we will focus on alternative sulfonamides, such as SDZ, SMX, and SMZ. The contents of these sulfonamides were all 10 mg/L. Other conditions were the same as the degradation experiment of STZ. Figure 6 shows that the removal soared up to about 50% in the first 10 min rapidly, and reached more than 90% in 60 min. The removal efficiency of SDZ, SMX, and SMZ was 90.1%, 96.2%, and 93.2%, respectively. These results disclosed that UV/PDS process had advantages in sulfonamides degradation and has a broad application prospect in practical wastewater treatment.

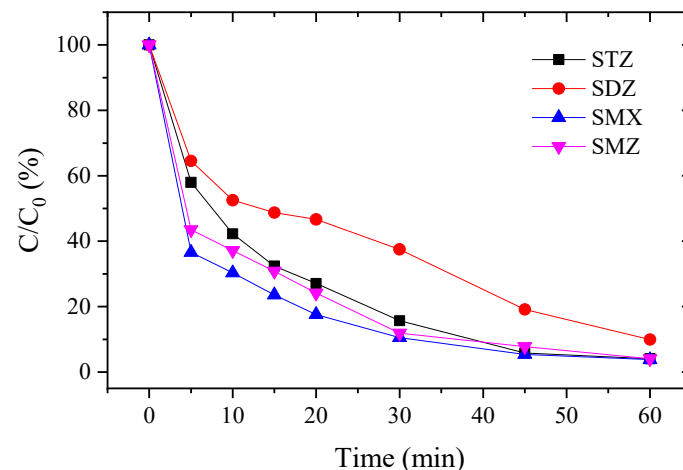


Figure 6. The removal efficiency of different SAs. Conditions: [SAs] = 10 mg/L, [PDS] = 300 mg/L and UV irradiation: 2 mW/cm².

4. Conclusions

The removal efficiency, influencing factors, and the mechanism of STZ degradation through an integrated UV/PDS process in water environment were studied and discussed in this work. The results revealed that the integrated process enhanced STZ removal rate from 72.6% to 95.9% within 60 min, suggesting excellent adaptability under different

conditions for different types and different concentrations of sulfanilamide antibiotics, common anions, and HA in water. As the amount of oxidant increased, the STZ removal rate increased significantly. The integrated UV/PDS process has a potential application in water treatment.

Author Contributions: Conceptualization, H.W. and P.S.; methodology, P.S.; validation, H.W. and P.S.; formal analysis, Z.L., H.Z., and W.H.; investigation, Z.L., H.Z., W.H., and P.S.; data curation, Z.L., H.Z., W.H., and P.S.; writing—original draft preparation, Z.L. and P.S.; writing—review and editing, Z.L., W.H., H.Z., H.W., and P.S.; visualization, H.W. and P.S.; supervision, H.W. and P.S.; project administration, H.W. and P.S.; funding acquisition, H.W. and P.S. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

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