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Review Article

Degradation of Antibiotics by Ultrasound-Assisted Heterogeneous Activation of Persulfate and Peroxymonosulfate: A Review

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ABSTRACT

Background: The presence of antibiotic residues in water is a matter of concern for both public health and the environment, owing to the phenomenon of antibiotic resistance. One of the Advanced Oxidation Processes (AOPs) employed for the removal of refractory pollutants in the aqueous matrix is sonication. The sonochemical processes have been found to have a multitude of advantages, including the mitigation or complete elimination of secondary pollutants, energy conservation, enhanced safety, and improved cleanliness. **Methodology:** Antibiotic degradation mechanisms are thoroughly reviewed in this article, with a particular focus on activating Persulfate (PS) and Peroxymonosulfate (PMS) using sonication from 2000 to 2023. The analysis of articles retrieved from Google Scholar, Web of Science, and Scopus was conducted using the suggested reporting items for systematic review and meta-analysis methodology. A total of 195 published publications were found after the initial search. Following a comprehensive evaluation, a total of 58 publications were chosen for the investigation of the utilization of ultrasound-assisted heterogeneous activation in PS and PMS operations. **Results:** Our findings were indicative of a discernible increase in publications in recent years, especially between 2017 and 2023. In addition, the elements that contribute to the effective degradation of antibiotics, such as pH, initial antibiotics concentration, PMS and PS dosage, contact time, and Ultrasonic frequency intensity, were analyzed and reviewed in this review study. **Conclusion:** This method has successfully removed antibiotics from aqueous solutions and hospital wastewater

Keywords: Persulfate, Peroxymonosulfate, Antibiotics, Sonication, Wastewater, Degradation.

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INTRODUCTION

The widespread use of antibiotics to safeguard human health and enhance animal growth has been found to be an undeniable necessity.[1,2] It is a well-established fact that a considerable proportion of antibiotics remain incompletely metabolized inside the human and animal physiological systems. Consequently, these antibiotics are inevitably discharged into sewage treatment facilities through the excretion of urine and feces, ultimately finding their way into the surrounding environment.[3,4] The world's daily antibiotic consumption has surged from 21.1 billion tones in 2000 to 34.8 billion tones in 2015.[5] According to estimates, the worldwide usage of antibiotics is predicted to rise by 67% by 2030, with countries such as China, the United States, India, Brazil, and Germany being the major consumers.[6,7] One possible consequence of overusing antibiotics is the growth of antibiotic-resistant microorganisms and environmental damage.[8]

At present, there exist several treatment methods for eliminating antibiotics from wastewater, such as biological treatment, chemical treatment, adsorption, and so on.[9] Nonetheless, adsorption can only isolate antibiotics from wastewater without fully degrading them.[10] In the case of biological treatment methods, the inhibition of bacterial activity by antibiotics has led to diminished efficiencies.[11,12] AOPs are highly effective in

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generating extremely reactive $OH^{\bullet}$, $SO_4^{\bullet-}$, and $O_2^{\bullet-}$ radicals; they are capable of efficiently decomposing antibiotics into significantly less toxic byproducts.[13,14]

Catalytic, physical, ozone-based, ultraviolet-based, and electrochemical-based AOPs are the five main categories into which AOPs are typically separated. Nevertheless, the acidic conditions required for conventional Fenton-AOPs severely limit their practical application across different industries.[15-17] Additionally, the disposal of considerable amounts of iron-containing sludge poses significant obstacles.[18] Surprisingly, persulfate-based AOPs represent a potential solution for addressing the aforementioned issues.[19] In recent times, the heightened redox potential (E_0=2.5-3.1 V) and robust oxidation capacity exhibited by the sulfate radical have contributed to its growing prominence.[20] Activating PS (e.g., PMS and Peroxydisulfate (PDS)) in the presence of electricity, heat, ozone, ultraviolet, ultrasound, carbon materials, and transition metals oxides can produce reactive radicals.[21-23]

Upon thorough analysis of the provided information, two noteworthy conclusions have been drawn. Firstly, it has been found that antibiotic have been recognized in surface water, groundwater, and wastewater, posing significant hazards to both human and aquatic life.[24-26] (II) Persulfate-based Secondly, persulfate-based AOPs offer an auspicious solution for removing these pollutants.[27,28] However, a comprehensive review of the various types of sonication-activated persulfate AOPs used for degrading different antibiotics is yet to be conducted. Hence, the objective of this work is to provide an overview of the degradation mechanisms involved in using sonication-activated persulfate AOPs to break down antibiotics.

MATERIALS AND METHODS

The purpose of this systematic review was to analyze the antibiotic degradation in aqueous solutions through the use of sonication with PS and PMS. The study examined articles published in electronic form from 2000 until the end of 2023 in various international databases, including Google Scholar, Scopus, PubMed, Web of Science, and Science Direct. A search was conducted using keywords such as antibiotic, degradation, Peroxymonosulfate (PMS), Peroxydisulfate (PDS), sonication-activated persulfate AOPs, and removal, with the use of AND/OR operators in the title and abstract.

RESULTS

The study aimed to investigate the degradation of antibiotics through the sonolysis process with the assistance of PS or PMS. According to the report by Safari *et al.*, sonication with PS in a 100 mL solution of TC for 120 min at 500 W and 35 kHz under a pH of 10 resulted in the removal of 94.4% of 30 mg/L TC. The removal efficiencies for TOC and COD were 59.7% and 72.8%, respectively.[29] According to Yin *et al.*, they achieved impressive results in their experiment on the removal of 50 mg/L SMZ. Using sonication alone, PMS alone, and Sono/PMS, they were able to achieve removal efficiencies of 8.6%, 54.3%, and 99.6%, respectively; their result was obtained at pH 7.5 under 600 W and 20 kHz for 30 min.[30] The process of destroying antibiotics through sonolysis with PS or PMS is illustrated in Figure 1. Studies have been conducted on the sonochemical degradation of antibiotics in aqueous solutions with PS. Table 1 provides a summary of the utilization of sonication in breaking down antibiotics utilizing PS or Oxone.[25-33] Under specified conditions represented in Table 1, Combining sonication with PS results in greater antibiotic degradation compared to sonication or oxidation alone, while achieving partial mineralization.[31,32] Using sono/PS, the removal efficiency of TC was 96.5%, while 61.2% of TOC and 74% of COD were removed.[33] Nevertheless, many antibiotics, like TC, exhibit resistance to degradation when subjected to a hybrid system, even after 240 min.[34] Furthermore, the TC degradation pathway by the sono/$S_2O_8^{2-}$ process (as depicted in Figure 2) illustrates the presence of the protonated TC molecular ion and four primary by-products.[33]

By incorporating PMS and sono/PS, significant enhancement in the antibiotic removal efficiency from aqueous solutions could be achieved.[20,35] Based on observations, neither the presence of Fe^0 nor PS alone is sufficient to offer substantial SDZ degradation. Similarly, single use of sonication could only lead to marginal degradation of SDZ with a removal efficiency of 9.7% after 1 hr. In addition, the removal efficiency of SDZ was only 9.8% and 13.7% in 1 hr when two components, e.g., sono/PS and sono/Fe^0, were combined. The further degradation of SDZ was impeded by the presence of surface passivation, which effectively prohibited the dissolution of Fe^0 and the release of Fe^{2+}. Thus, in order to increase mass transfer and remove the passivation coating, sonication was employed. This resulted in removing 95.7% SDZ after 1 h in the sono/Fe^0/PS process.[36] Eliminating TC using only PS or sonication is also challenging. Moreover, the Fe_3O_4 catalyst alone or in combination with sonication could not remove much TC because of the insufficient TC adsorption by Fe_3O_4 and the incomplete generation of active species. Nonetheless, after 90 min, the Fe_3O_4/PS and sono/PS exhibited the capability of enhancing the TC removal efficiency to 50.5% and 51.5%, respectively, by activating the catalyst through sonication and producing more $SO_4^{\bullet-}$ and $^{\bullet}OH$ on the catalyst surface. Moreover, the implementation of sono/Fe_3O_4/PS was found to be highly effective, with a TC removal efficiency of 89%. This was achieved through the activating PS by Fe_3O_4 and sonication, which effectively produced a greater $SO_4^{\bullet-}$.[35] Also, The parameters that are effective for removing antibiotics are presented in Table 2.

DISCUSSION

Advanced oxidation processes that use sulfate radicals are an emerging technology for treating wastewater.[36,37] In this method, PS, SO_5^{2-} or $S_2O_8^{2-}$ or PMS, HSO_5^- can be activated to form sulfate radicals ($SO_4^{\bullet-}$) by various means, such as UV radiation, sonication, heat, alkaline pH, or transition metal ions.[38-40] Oxone, which is the triple salt $KHSO_5$·0.5 $KHSO_4$·0.5 K_2SO_4, is a more stable.[41] The sulfate radical-based AOPs are more effective and potent than ·OH-based AOPs; this is derived from more stability of $SO_4^{\bullet-}$, compared to ·OH, during degradation reactions so that it has exhibited an outstanding oxidation ability in an extensive range of pH (2-8).[42,43]

·OH exhibits a redox potential range of 1.89-2.8 V, which classifies it as a highly effective oxidant. $SO_4^{\bullet-}$ is produced from PS, which has a standard redox potential of 2.01 V, exceeding that of PMS (1.81 V).[44,45]

When compared, it can be observed that $SO_4^{\bullet-}$ possesses a redox potential that is either similar or greater (ranging from 1.81-3.1 V) than that of other substances, depending on its method of activation.[46,47] $SO_4^{\bullet-}$ is produced from PS, which has a standard redox potential of 2.01 V, exceeding that of PMS (1.81 V).[48] The removal efficiency of Acid Orange 7 dye by heat activation follows the order of PS > PMS > H_2O_2. However, it is PS > H_2O_2 > PMS after employing UV for activation.[48] Achieving a removal efficiency of 95.3% and 58.4% for 25 mg/L furfural was possible under sonication activation using PS and PMS, respectively.[49] The order of SMX removal efficiency by UV activation was found to be different from that of other factors, with PMS showing the highest efficiency followed by PS and H_2O_2.[42] This suggests that the contribution of the entire oxidation system on changing oxidation potential of PS and PMS.[50]

Sonication is an effective method for eliminating resistant pollutants. It also triggers the production of $SO_4^{\bullet-}$ through the reactions of PS and PMS with ·OH, which are generated in situ during sonication. These reactions can be represented by Eqs (1) to (6).[51-53]

$$S_2O_8^{2-} + {}^{\bullet}OH\ HSO_4^- + SO_4^{\bullet-} + 0.5\ O_2 \quad (1)$$

$$HSO_5^- + US\ SO_4^{\bullet-} + {}^{\bullet}OH \quad (2)$$

$$HSO_5^- + SO_4^{\bullet-} + SO_4^{2-} + H^+ \quad (3)$$

$$S_2O_8^{2-} + US/thermal\ 2\ SO_4^{\bullet-} \quad (4)$$

$$SO_4^{\bullet-} + {}^{\bullet}OH\ HSO_4^- + 0.5\ O_2 \quad (5)$$

$$S_2O_8^{2-} + SO_4^{\bullet-}\ SO_4^{2-} + {}^{\bullet}S_2O_8^{2-} \quad (6)$$

Thus, the degradation of antibiotics has been ascribed to producing $SO_4^{\bullet-}$ and ·OH resulting from the PS or PMS activation during sonication.[54]

Table 1: Summary of sonication systems for degrading antibiotics.

Antibiotics	PS (mM)	Frequency (kHz)	% Removal PS	% Removal sono	% Removal sono/PS	References
SMX	2	20	4.8	6.7	91.3	21
TC	200	20	20.1	4.5	55.5	22
TC	5	35	57.3	26.9	88.5	23
SFZ	4	20	4.9	37.3	62.4	24
TC	4.4	40	7.5	2.5	18.5	25
CMH	4.8	20	8.8	9.7	45.2	26
SMX	1	40	11.1	1.6	47.2	27
TC	2	20	54.3	8.6	99.6	28

Table 2: Results of various reported studies on the effect of different parameters.

Antibiotics	pH	C_0 (mg/L)	Time (min)	Reaction rate	% Removal	References
AMX	2	10	60	0.221	95.4	7
LFX	4	25	60	0.172	76.3	8
AMX	3	25	75	0.144	77.9	9
TC	3	25	60	0.085	79.3	13
SMX	5	50	60	0.074	84.3	16
TC	3	20	75	0.096	91.2	17
TC	2	25	75	0.141	62.4	18
CIP	3	50	45	0.116	99.6	19

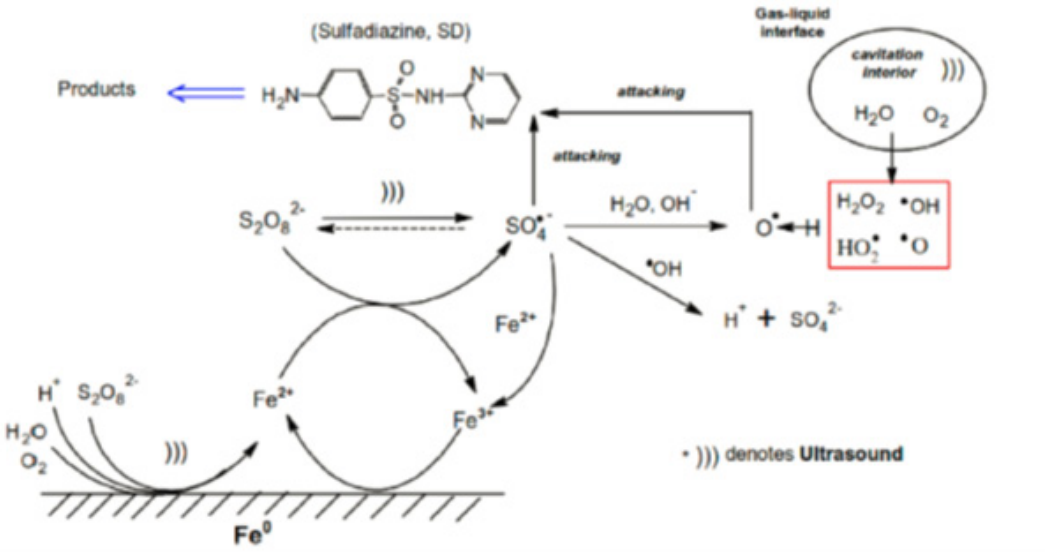

Figure 1: The SDZ degradation mechanism in sono/Fe0/PS system.

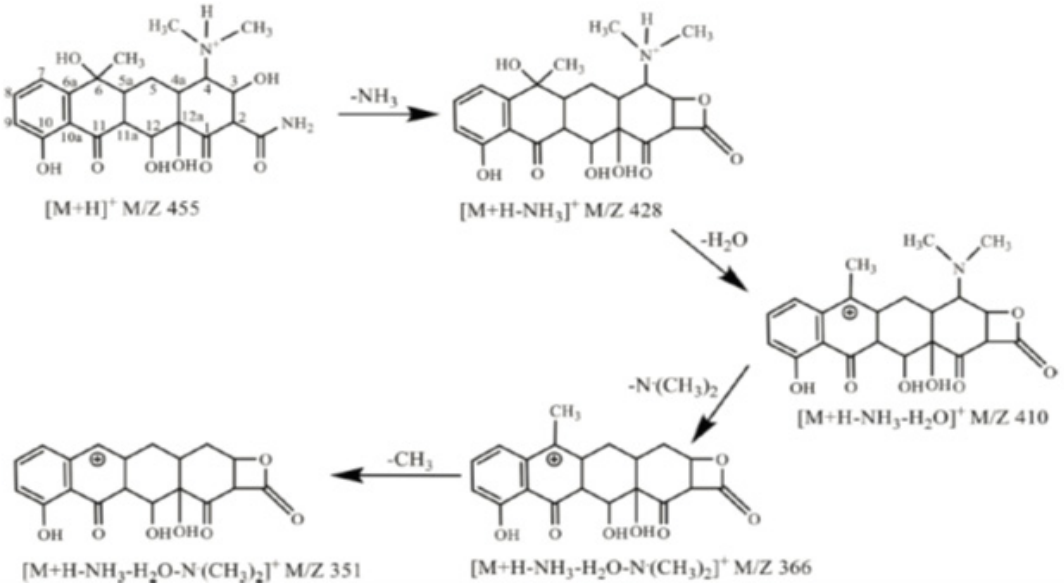

Figure 2: Suggested TC degradation pathway in a sono/$S_2O_8^{2-}$ system.

After that, chemical bonds of antibiotics, e.g., S-N, S-C, and N-C in SMZ, or N-methyl, hydroxyl, and amino groups in TC, are cleaved through oxidation with $^{\bullet}OH$ and $SO_4^{\bullet-}$.[30]

The TC sonocatalytic degradation in the Fe_3O_4/PS system has been observed to exhibit an increase in the removal efficiency of TC with a rise in PS concentration (20 to 200 mM). Nevertheless, the removal efficiency of TC begins to decrease when the PS concentration exceeds 200 mM. This is attributed to excessive production of sulfate anions by PS instead of active $SO_4^{\bullet-}$, hence diminishing the effectiveness of the process.[52] Speculations suggest that the formation of $SO_4^{\bullet-}$ could be potentially mitigated by excess PS that may act as a scavenger, thereby impeding the generation of $^{\bullet}OH$. It is noteworthy that the supply of 200 mM PS is deemed sufficient to facilitate generation.[53] The solution pH level has a significant impact on several factors, including the dissociation of antibiotics, the adsorption of antibiotics onto catalysts, and the leaching of metals and their oxides. These factors ultimately affect the antibiotic sonocatalytic degradation in the presence of PS or PMS.[35]

Achieving efficient degradation of SDZ (95.7%-98.4%) in the sono/PS system is possible at pH values from 3.0 to 7.0. However, at pH=10, the system's efficiency was significantly diminished by 35.7%. In lower pH ranges, Fe^0 is more prone to corrosion and results in the formation of soluble Fe^{2+}. Conversely, in alkaline

conditions, the soluble iron ions precipitate and passivate the Fe^0 surface, leading to low production of •OH and $SO_4^{\bullet-}$.[36] Moreover, for neutral or alkaline pH, the generated $SO_4^{\bullet-}$ not only undergoes a reaction with H_2O and OH^- but also causes a reduction in •OH reactivity.[36] In addition, during the antibiotic degradation at pH ranges of 3.0-7.0, a gradual diminution in the pH of the solution could be detectable, which is attributed to generating carboxyl acid products and the decomposing PS. At the end of a degradation process, a decrease in pH value from 10 to 6.5.[29]

The research of Pan *et al.* has revealed that the sono/premagnetized-Fe^0/PS system experiences a more rapid decrease in pH as reaction time increases when compared to other systems. This swift decrease in pH facilitates the faster generation of Fe^{2+} and consequently, the more generation of $SO_4^{\bullet-}$, which results in a highly effective degradation of SMZ. Additionally, it also leads to remarkable synergistic effects in the mentioned system, further aiding in the degradation of SMZ.[55]

The efficiency of the Sono/PS system for TC degradation is strongly influenced by the initial value of pH. The degradation rates of TC (in the absence of any buffer) at pH 4, 7, and 10 were found to be 77.4%, 62.5%, and 88.5%, respectively, after 120 min.[35] At different pH levels, TC (pKa of 3.3, 7.7, and 9.7) behaves differently due to its amphoteric nature. At pH = 4, TC molecules are mostly neutral or have positive charges while at pH=9, they have negative charges. TC molecules with a negative charge are highly reactive and can attract species such as •OH, due to the increased electric density on the ring system. This leads to speeding up in TC degradation. At pH≥10, alkaline-activated PS is the main source of $SO_4^{\bullet-}$, $O_2^{\bullet-}$ and •OH. Moreover, the reaction between $SO_4^{\bullet-}$ and OH^- under alkaline conditions is an effective way to generate •OH.[35] Thus, an increase in pH leads to improvement in decomposing PS to produce •OH and $SO_4^{\bullet-}$, as described in Equations no. 7-9.[55-57]

$$S_2O_2^{-8} + 2H_2O\ HO_2^- + 2SO_4^{2-} + 3H^+ \quad (7)$$

$$HO_2^- + S_2O_2^{-8}\ SO_4^{\bullet-} + SO_4^{2-} + H^+ + O_2^{-\bullet} \quad (8)$$

$$SO_4^{\bullet-} + OH^-\ SO_4^{2-} + {}^{\bullet}OH \quad (9)$$

The effectiveness of Sono/$S_2O_8^{2-}$ or Sono/Oxone processes in degrading antibiotics was improved significantly due to the rise in temperature, which increased the cavitational activity and chemical reactions. As depicted in equation no.10, $SO_4^{\bullet-}$ can be produced by activating PS through heating.[58]

$$S_2O_8^{2-}\ 2SO_4^{\bullet-} \quad (10)$$

At pH of 4, $SO_4^{\bullet-}$ is the primary agent that leads to the degradation of TC, whereas at pH of 7, the degradation of TC is caused by both $SO_4^{\bullet-}$ and •OH. Consequently, the degradation rate of TC at a pH of 7 is reduced, which is ascribed to the competition between $SO_4^{\bullet-}$ and •OH and TC.[31]

CONCLUSION

After thoroughly reviewing published literature, it has become evident that the degradation mechanism and kinetics of antibiotics via ultrasound-assisted heterogeneous activation of PS and PMS are subject to various factors. These factors include the chemical composition of the materials utilized, and physico-chemical experimental conditions such as pollutant concentration, PS and PMS dosages, and pH levels within the system. It is crucial to take into account that most of the research regarding the removal of antibiotics in the US is conducted on a small scale. Therefore, additional investigations on a larger scale are recommended. When using pilot-scale systems, several factors such as energy consumption, mass transfer, pH regulation, temperature control, and application in actual wastewater samples should be considered.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest.

ABBREVIATIONS

PS: Persulfate; **PMS:** Peroxymonosulfate; **OH•:** Hydroxyl radical; **$SO_4^{\bullet-}$:** Sulfate radical, **$O_2^{\bullet-}$:** Superoxide radicals; **PDS:** Peroxydisulfate; **AMX:** Amoxicillin; **LFX:** Levofloxacin; **TC:** Tetracycline; **SMX:** Sulfamethoxazole; **CIP:** Ciprofloxacin; **SFZ:** Sulfadiazine; **CMH:** Chloramphenicols.

SUMMARY

One of the advanced oxidation processes employed for the removal of refractory pollutants in the aqueous matrix is sonication. Antibiotic degradation mechanisms are thoroughly reviewed in this article, with a particular focus on activating PS and PMS using sonication from 2000 to 2023. The elements that contribute to the effective degradation of antibiotics, such as pH, initial antibiotics concentration, PMS and PS dosage, contact time, and Ultrasonic frequency intensity, were analyzed and reviewed in this review study. This method has successfully removed antibiotics from aqueous solutions and hospital wastewater.

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