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REVIEW

Recent advances and future outlook for treatment of pharmaceutical from water: an overview

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Abstract

Water pollution and wastewater treatment problems are considered one of the largest challenges in the world today, and it is very crucial to pick the best way to remove these kinds of pollutants. This systematic review's objective is to categorize and investigate the strategies for eliminating pharmaceutical contaminants from aquatic environments, focusing on advanced oxidation techniques. For this purpose, Google Scholar, PubMed, Nature, Scopus, Web of Science, American Chemical Society, Elsevier, Springer, and Royal Society of Chemistry databases with the keywords Pharmaceuticals, Water Treatment, Advanced Oxidation Process, Photolysis, Fenton, Ozonation, Sonolysis, and Electrochemical were used. Physical, chemical, thermal, and biological methods are employed to eliminate these contaminants. However, even after purification, the drugs and resulting metabolites might still be present in the environment and cause serious damage. The analysis of studies revealed that advanced oxidation processes, which deserve more attention, can eliminate pharmaceutical contaminants because of their quick reaction times and strong oxidation capacities. This study demonstrated how environmental factors like pH, temperature, the content of the water, sunlight, etc., affect drug removal. Also, methods based on ozonation, ultrasonic radiation, electrochemical, ultraviolet radiation, and Fenton were investigated. Multiple hybrid systems with characteristics like high degradation efficiency, no secondary pollution production, and compatibility with the environment, high mineralization, and low operating costs were recommended as potential systems for further research.

Keywords Advanced oxidation process · Degradation · Reactive oxygen species · Organic pollutants · Systematic review

Introduction

The discovery of medicines marked a turning point in human scientific progress since they increase the quality of life, prolong life, and save millions of people from fatal diseases (Arghavan et al. 2021). Drugs have become environmental pollutants, which are also linked to rising growth as a result of their success in production and excessive use. After use, a significant portion of drugs for humans or animals as well as their metabolites reach aquatic environments (Khalatbary et al. 2022). Drug residues have been found in practically every area of the world in the last few decades. Groundwater, sewage treatment plants, effluents, and sludge, as well as surface water (lakes, rivers, streams, estuaries, and oceans) are all included (Patel et al. 2019). Even in the world's purest environment, the arctic regions, pharmaceutical contaminants have reportedly been found. Most drugs do not last in the environment for very long. However, many of them become "pseudo-persistent" medications as a result of their continued, small addition to the environment (Yazdi et al. 2018). The 1970s through the 1990s, the first times drug contamination of water systems was reported. For example, in some regions of Germany and the UK, concentrations of tetracycline, theophylline, and estrogen were found to be between mg/L and ng/L (Kollarahithlu et al. 2021). The removal efficiency for medications including carbamazepine, atenolol, acetylsalicylic acid, diclofenac, mefenamic acid, propranolol, and enolic acid is less than 10% because wastewater treatment plants have not been developed for pharmaceutical compounds (Lee et al. 2019).

In order to evaluate the environmental risks of drugs, their consumption volumes, physicochemical properties,

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and environmental toxicities should be considered (Nasseh et al. 2022). High solubility in water, persistence, bioaccumulation, toxic and possible carcinogenic effects on organisms can help in the risk assessment analysis of drugs. Due to their complicated behaviors and varied drug interactions, several pharmaceuticals have the potential to cause significant damage (de Garcia et al. 2017). Some drugs have similar effects on humans and other organisms. Drugs must have a very high solubility in water in order to be absorbed by different types of target cells in the body of a living organism (human, animal, or plant). As a result, drugs have very strong biological activity that affects living organisms (Sayadi et al. 2010). Drugs are made to bind to particular receptors in both humans and animals. As a result, these compounds can interact with receptors that are similar in other organisms. Toxic effects on microorganisms and animals have also been reported. There have also been reports of antibiotic resistance in bacteria and microbial communities, gene expression alterations, abnormal protein and enzyme activity, and growth anomalies in mice, fish, and frogs (Beijer et al. 2013). The drug Diclofenac has been related to toxic effects on Daphnia magna, the green algae Pseudokirchneriella subcapitata, and the Cyanobacterium Anabaena Flos-aques, as well as a reported reduction in vulture populations in Southeast Asia. Ivermectin has also been documented to have other major environmental harmful effects on populations of insects and aquatic invertebrates (Rivera-Utrilla et al. 2013).

Nowadays, many methods, including physical, chemical, thermal, and biological methods, are used to remove pharmaceutical pollutants, among which the use of advanced oxidation processes (AOPs) has received much attention (Kargar et al. 2021). AOPs are environmentally friendly methods that use free radicals to oxidize organic substances. As powerful oxidizers, these radicals have the ability to completely oxidize most organic compounds into carbon dioxide, water, and mineral acids (Li et al. 2020). However, they may produce harmful byproducts and are ineffective in treating large volumes of contaminants. As a result, combining them with other techniques can increase their effectiveness (Ganiyu et al. 2015). With the help of this study, we could better understand the different types of pharmaceutical pollutants and how to remove them from aquatic environments. This study focused on determining and describing the nature of pharmaceutical pollutants. Additionally, by introducing AOP methods and comparing their performance, it is possible to gain a novel viewpoint on pollution removal techniques. The ideal alternative with the highest level of destruction efficiency was finally introduced for further studies by comparing different systems. This article is done by reviewing the literature of the last ten years (2013–2023).

Materials and methods

To conduct this systematic review research, Google scholar, PubMed, Nature, Scopus, Web of Science, American Chemical Society, Elsevier, Springer, and Royal Society of Chemistry databases were used, and the most relevant and up-to-date articles were studied. Keywords such as Pharmaceuticals, Water Treatment, Advanced Oxidation Process, Photolysis, Fenton, Ozonation, Sonolysis, and Electrochemical were used (Fig. 1). To achieve the research's objective, the articles from the collection of 1337 articles that discussed advanced oxidation methods for removing pharmaceutical contaminants were included in the study. 1238 articles including Review articles (653), Book chapters (231), Encyclopedia (9), Case reports (21), Conference articles (187), and Persian articles (131) were excluded from the study after screening. Finally, 105 articles were examined while keeping in mind the goal of the study and the entrance requirements. Additionally, the assistance of the technical professors was sought out for a deeper understanding of the problems. Finally, pharmaceutical pollutants, their consumption, presence and fate in the environment, influence of environmental parameters, and drug removal techniques, especially advanced oxidation processes, were investigated.

Results and discussion

The difference between pharmaceutical pollutants and other organic pollutants

Pharmaceutical pollutants differ from other pollutants in having a molecular mass of less than 500 dalton (although it is larger for some compounds), having complex chemical molecules with different structures, and having polar compounds with over one ionizable group. Additionally, depending on the pH of the environment, drugs may show different characteristics and degrees of ionization. They have intermediate water solubility and lipophilic characteristics (Sallam et al. 2021). On the other hand, drugs have the capacity to last in the natural environment, accumulate in the environment, and remain biologically active. For example, naproxen, sulfamethoxazole, and erythromycin can persist in the environment for approximately

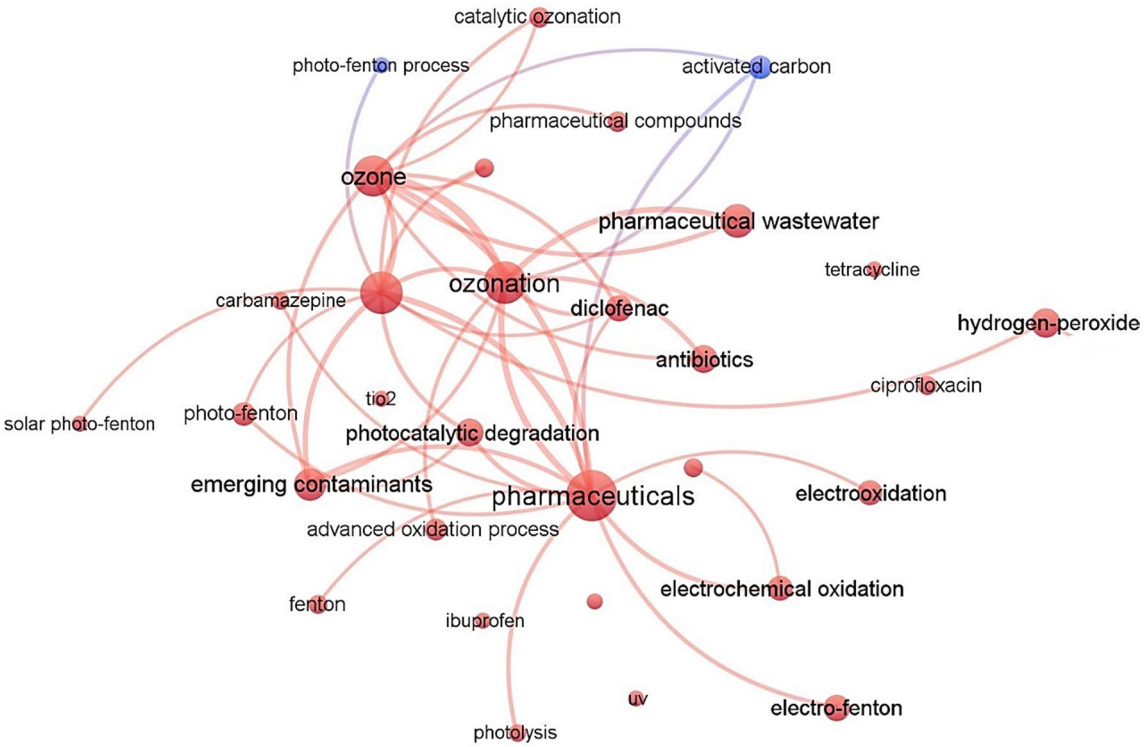

Fig. 1 Keyword and overlay visualization co-occurrence analysis of AOPs of drugs using VOS Viewer

one year, while clofibric acid can remain unchanged for several years. The body of the organism tends to absorb and distribute these molecules, which metabolically modify their chemical structure (Rivera-Utrilla et al. 2013).

Drugs consumption patterns

Drugs are frequently offered for sale as "over-the-counter" illegal medications. For assessing consumption volumes and patterns, this increases uncertainty. Over 5000 over-the-counter medications and about 3000 registered medications are available in the UK (Shaheen et al. 2019). High-income countries have a much higher average per capita drug consumption in non-hospital departments than middle-to-high, middle-to-low, and low-income countries. This significant difference in medicine use patterns has a substantial impact on the presence of pollutants in the environment (Mehta et al. 2016). Large-scale drug usage is prevalent in animal husbandry and agriculture. For the prevention and treatment of various infectious and non-infectious diseases, they are administered to animals orally (via water or food) and locally (by injection). There is widespread usage of food supplements, steroidal and non-steroidal anti-inflammatories, and antibiotics. Animal reproductive systems are regulated by hormones and estrogens like oxytocin, steroids, ergonovine, HCG, GnRH, progesterone, FNRH, and prostaglandin (Rogers and Aronoff 2016). Antibiotics, bovine somatotropin, ionophores, and growth hormone implants are some methods used to increase milk and meat production. Insecticides and vermicides have been used to control parasites. As a result, animal antibiotic use is significantly greater than human use. Animal husbandry can contribute more drugs to the environment because human waste is usually treated, whereas animal waste is not (Hu et al. 2017).

The presence and fate of drugs in the environment

Medicines are grouped as 1. Analgesic and anti-inflammatory (ibuprofen, paracetamol, diclofenac), 2. Antibiotics (sulfonamides, tetracyclines, penicillins, beta-lactams, macrolides, fluoroquinolones, imidazole). 3. Antiepileptic drug

(carbamazepine). 4. Antidepressants (benzodiazepines), 5. Fat-lowering agents (fiberrates), 6. antihistamines (famotidine and ranitidine), and 7. beta-blockers (metoprolol, atenolol, and propranolol) (Aus der Beek et al. 2016).

Pharmaceutical pollutants have been found in drinking water in concentrations as low as ng/L, significantly lower than their therapeutic doses. Daily doses at these levels are therefore regarded as insignificant, while it is yet unclear what their long-term effects would be. The relative frequency of finding different drugs varies by location. For instance, whereas antibiotics are more prevalent in Asia, painkillers are the most popular pharmacological class worldwide. The key factor influencing seasonal variations in drug use is consumption habits. In the spring and during the pollen release seasons, allergies are at their worst. This causes more people to use antihistamines, which increases the amount of this drug that ends up in wastewater (Patel et al. 2019). In order to treat illnesses, the use of antibiotics rises in the winter and autumn. Ephedrine, pseudoephedrine, and pholcodine are commonly used to treat coughs throughout the winter, increasing their quantities in water and wastewater. Sludge is used to remove large volumes of non-polar and less polar medicinal drugs that have been absorbed into the sediments and solids of sewage treatment plants. This sludge can introduce pollutants into the food cycle as well as surface and underground water systems when used as fertilizer in agricultural fields (Vinayagam et al. 2022).

The primary source of pharmaceutical pollution is municipal sewage. Due to their high concentrations, drug production factories are also very concerning. The severity of this problem is increased in underdeveloped nations when effective industrial wastewater treatment is lacking. For instance, maximum concentrations of Ciprofloxacin (31 mg/L) and Trimethoprim (28 mg/L) have been found in wastewater in India and Croatia, respectively (Balakrishna et al. 2017; Larsson et al. 2007). High concentrations of Metaxalone (3.8 mg/L) and Oxycodone (1.7 mg/L) have also been found in wastewater treatment in the United States (Phillips et al. 2010). Drugs are designed to be chemically stable, but undergo physicochemical and biological transformations. To forecast the environmental fate of pharmaceuticals, knowledge of their metabolic pathways, degradability, persistence, and absorption is required (Sayadi et al. 2019). The following section will look at the factors that have an impact on how drugs react in the environment.

The influence of environmental parameters

Pharmaceutical changes are influenced by a variety of environmental factors, including pH, temperature, sunlight, salinity, and others. Dilution and absorption in soil, sediments, natural organic matter, and suspended particles also have an effect on the degradation and removal of drugs from the environment (Hajiani et al. 2022). Their distribution in the environment and natural degradation are both influenced by water salinity. Where freshwater and saltwater mix, the importance of salinity increases (Noppe et al. 2007). Pharmaceutical compounds are removed from surface waters by a number of key processes, including biodegradation, photodegradation, and absorption in soil, sludge, etc. Medicines are made to be microbially resistant (especially antibiotics). This slows down the rate of microbial decomposition removal. Since many drugs contain photosensitive functional groups, photodegradation serves as a significant surface water removal mechanism (Ahmadpour et al. 2020).

Drug-removal techniques

Since stability is frequently regarded as a desirable quality for human use, many medications are environmentally stable. Improved removal techniques are therefore required in order to remove pharmaceutical compounds from water and wastewater. Pharmaceutical contaminants are eliminated via physical, chemical, thermal, and biological techniques. Surface absorption, electrodialysis, evaporation, dialysis, filtration, flocculation, reverse osmosis, and sedimentation are all types of physical purification (Ahmed et al. 2015). Ion exchange, neutralization, and advanced oxidation processes are examples of chemical procedures, while burning and thermal decomposition are examples of thermal techniques (Quintel et al. 2019). The most popular biological techniques include activated sludge, aeration ponds, anaerobic digestion, trickling filters, and waste stabilization ponds (Yahya et al. 2020). Drugs are typically converted from an aqueous phase to a solid phase via physical techniques. Drugs undergo chemical reactions during biological and chemical treatment to create new metabolites or degradable products (Kamaly et al. 2016).

For example, by ferric chloride, hydrophilic drugs including acetaminophen, sulfamethoxazole, and dehydro nifidipine had about a 25% reduction in concentration, and coagulation with ferric chloride leads to acid or alkaline hydrolysis of these compounds. With aluminum coagulants (alum) and iron salt, the average elimination of caffeine, ethinylestradiol, estrone, estradiol, progesterone, and androstenedione were only approximately 6%. Less than 20% of the following drugs were typically removed from the body: sulfamethoxazole, pentoxifylline, meprobamate, dilantin, carbamazepine,

estriol, iopromide, naproxen, diclofenac, triclosan, gemfibrozil, acetaminophen, testosterone, trimethoprim, and hydrocodone (Stackelberg et al. 2007). On the other hand, water chlorination reduced the levels of acetaminophen, sulfamethoxazole, and erythromycin by about 75% (Vieno et al. 2006).

The treatment plants' last step is disinfection. Through disinfection, a significant number of bacteria and contaminants can be removed, including pharmaceuticals. Some chemicals are destroyed by the chlorine present in disinfected drinking water sources. Conventional water treatment facilities use chlorination to disinfect water all around the world. It is still the most widely used treatment device because of how affordable it is (Mohapatra 2017). Many drugs cannot be entirely digested by the bacteria used in secondary treatment, and they cannot be efficiently adsorbed on activated sludge. As a result, the treatment facility's ability to extract pharmaceuticals from regular wastewater is somewhat limited. Additionally, some pharmaceutical contaminants in wastewater prevent microbial activity, which reduces the amount of wastewater treatment required to remove those pollutants (Patel et al. 2019).

Advanced oxidation processes

Nowadays, advanced oxidation processes are proposed to eliminate pharmaceutical compounds in aqueous environments. Reactive radical species or active oxygen (superoxide anion radical, hydroxyl radical, and hydroperoxyl radical) are strong oxidants that are produced in this process (Chamanehpour et al. 2023; Poorsajadi et al. 2020). These radicals are created by hydrogen peroxide (H_2O_2) or ozone (O_3) with catalysts, UV light, or gamma rays. Numerous types of organic and inorganic contaminants are oxidized by these free radicals with a high reaction rate constant due to their strong reactivity (Chamanehpour et al. 2022; Sayadi et al. 2022). Due to its strong reactivity, non-selective nature, and high oxidizing properties, the hydroxyl radical is more effective at removing pollutants than the others. AOPs have been used for ibuprofen, diclofenac, ciprofloxacin, tetracycline, carbacycline, carbamazepine, diclofenac, sulfamethoxazole, 17α-ethinyl estradiol, moxifloxacin, paracetamol, naproxen, clofibric acid, and many other drugs (Yazdani et al. 2019; Sayadi et al. 2019; Jiménez-Salcedo et al. 2022). The performance of several advanced oxidation techniques, such as photolysis, fenton, ozonation, sonolysis, and electrochemical, will be compared in the following section (Fig. 2).

Without irradiation

Ozone based Ozone is a strong oxidizing agent with a standard oxidation potential of 2.07 V that reacts with nucleophilic and electron-rich compounds. This powerful electrophile has two different mechanisms in which it might interact with contaminants. In a direct way, ozone attacks acidic sites, and in an indirect way, it uses strong oxida-

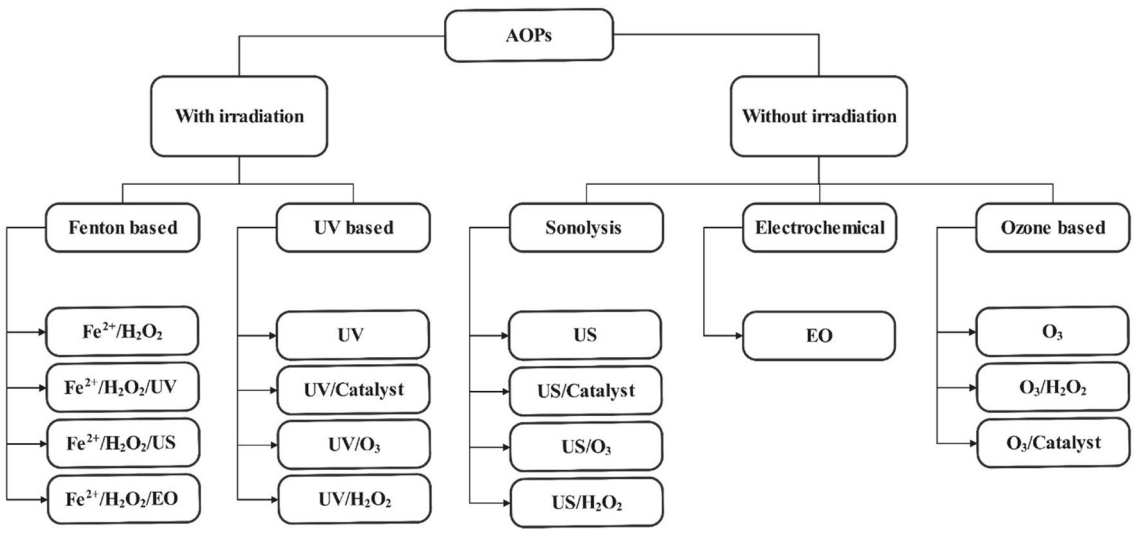

Fig. 2 Flowchart of under review AOPs methods

tive radicals as a secondary oxidation agent (Almomani et al. 2016). Compounds containing double or triple carbon–carbon bonds and also aromatic groups are employed as electrophiles in direct ozonation. However, the second route results in the formation of hydroxyl radicals from the reaction between ozone and water hydroxide anions, which leads to extremely quick reactions with organic molecules (Rasheed et al. 2021).

Direct path mechanism: Ozone quickly splits the unsaturated bond with pollutants in reaction 1 because of its high oxidative potential. This results in the destruction of the pollutants (Malik et al. 2020).

$$O_3 + OH^- + H^+ \rightarrow 2OH^\bullet + 4O_2 \quad (1)$$

Indirect path mechanism: This pathway begins with the reaction of ozone with hydroxide ions, which creates the superoxide anion and the hydroproxyl radical (reactions 2 and 3). Then superoxide anion enters the next reactions with ozone to finally produce hydroxyl radicals (reactions 4–6). The generated hydroxyl radical can react with ozone (reactions 7 and 8) and organic matter (Ikehata and Li 2018).

$$O_3 + OH^- \rightarrow HO_2^\bullet + O_2^- \quad (2)$$

$$HO_2^\bullet \leftrightarrow O_2^{\bullet-} + H^+ \quad (3)$$

$$O_3 + O_2^{\bullet-} \rightarrow O_3^{\bullet-} + O_2 \quad (4)$$

$$HO_3^\bullet \rightarrow H^+ + O_3^{\bullet-} \quad (5)$$

$$HO_3^\bullet \rightarrow OH^\bullet + O_2 \quad (6)$$

$$OH^\bullet + O_3 \rightarrow OH_4^\bullet \quad (7)$$

$$OH_4^\bullet \rightarrow O_2 + HO_2^\bullet \quad (8)$$

The reaction of the hydroxyl radical with the R organic molecule can act as a catalyst and form the $R^\bullet$ organic radical, which can restart the chain reaction in the presence of oxygen (reaction 9–12). Finally, some molecules interact with $OH^\bullet$ to create secondary radicals, where carbonate can serve as a scavenger and bicarbonate as an absorber of hydroxyl radicals (reactions 13 and 14), or it can break the chain (15th Reaction) (Malik et al. 2020).

$$H_2R + OH^\bullet \rightarrow HR^\bullet + H_2O \quad (9)$$

$$HR^\bullet + O_2 \rightarrow HRO_2^\bullet \quad (10)$$

$$HRO_2^\bullet \rightarrow R + HO_2^\bullet \quad (11)$$

$$HRO_2^\bullet \rightarrow RO + OH^\bullet \quad (12)$$

$$OH^\bullet + CO_3^{2-} \rightarrow CO_3^{\bullet-} + OH \quad (13)$$

$$OH^\bullet + HCO_3^- \rightarrow HCO_3^\bullet + OH \quad (14)$$

$$OH^\bullet + OH_2^\bullet \rightarrow O_2 + H_2O \quad (15)$$

It should be noted that a variety of factors, including hydroxyl dose, drug concentration, mode of operation, and water quality parameters such as pH, total dissolved solids, total suspended solids, and salinity, are effective on ozone oxidation rate (Kanakaraju et al. 2018).

O_3/H_2O_2 In the ozonation process, side products with low molecular weight may be produced, which have more acute toxicity than the primary pollutant, and H_2O_2 can improve the ozonation process as an initiator. In the peroxone reaction (O_3/H_2O_2), hydroperoxide ions (HO_2^-), which are more potent than the hydroxyl radical, are created (Feng et al. 2021). These ions can interact with ozone in addition to interacting with contaminants. Ozone oxidation starts a series of chain events that generate the hydroxyl radical. The created hydroxyl radical can also interact with ozone once more to create additional hydroperoxide ions. The cycle just keeps going. The initial ozone and hydrogen peroxide concentrations depend on how effectively these reactions proceed. As a result, there are two ways that pollutant oxidation happens (Cuerda-Correa et al. 2019): 1. direct reaction with ozone (reaction 16–21) and 2. indirect reaction with hydroxyl radical (reaction 22).

$$H_2O_2 \leftrightarrow HO_2^- + H^+ \quad (16)$$

$$HO_2^- + O_3 \rightarrow HO_2^\bullet + O_3^{\bullet-} \quad (17)$$

$$HO_2^\bullet \rightarrow H^+ + O_2^{\bullet-} \quad (18)$$

$$O_2^{\bullet-} + O_3 \rightarrow O_2 + O_3^{\bullet-} \quad (19)$$

$$O_3^{\bullet-} + H^+ \rightarrow HO_3^\bullet \quad (20)$$

$$HO_3^{\bullet} \rightarrow OH^{\bullet} + O_2 \quad (21)$$

$$OH^{\bullet} + O_3 \rightarrow HO_2^{\bullet} + O_2 \quad (22)$$

An important factor in the peroxone process is that for the system to operate at its best, the pH and O_3/H_2O_2 ratio must be carefully controlled. Two moles of ozone to one mole of hydrogen peroxide are the ideal molar ratio for the peroxone process (Lee et al. 2019).

O_3/Catalyst A catalyst can speed up the ozone's decomposition and the generation of free radicals during the ozonation process. Pollutants are effectively removed with this method. Additionally, when compared to ozonation, this procedure results in greater mineralization of organic pollutants (Wang and Chen 2020). There are two types of catalysts used in the catalytic oxidation of ozone: homogeneous and heterogeneous. Due to their economic nature and the absence of side pollutant production, heterogeneous catalysts are currently used more frequently than homogeneous catalysts (Wang et al. 2019).

Homogeneous catalytic oxidation: This method is used to improve ozonation by using transition metal cations including Mn, Fe, Co, Cu, Ni, Cd, Zn, and Cr. Metal ions have two possible actions: producing free radicals by breaking down ozone; and facilitating oxidation by oxidizing organic materials (Rekhate and Srivastava 2020). In fact, metal ions increase the selectivity of ozone oxidation, reaction speed, and ozonation efficiency (Khuntia et al. 2016).

Heterogeneous catalytic oxidation: In the heterogeneous catalytic oxidation of ozone, metal oxides (such as MnO_2, TiO_2, MgO, Al_2O_3, and Fe_2O_3) (Wang et al. 2018) and porous materials (such as activated carbon and zeolite) are used to improve ozonization (Ikhlaq et al. 2017). In this process, the absorption mechanism is dominant and it is done in three ways: 1. ozone absorption on the catalyst surface, 2. organic molecule absorption on the catalyst surface, and 3. ozone and organic molecule absorption on the catalyst surface simultaneously. In general, the mechanism of the ozone catalytic oxidation process is as follows (Wang and Chen 2020):

$$M^{n+} + O_3 + H^+ \rightarrow M^{(n+1)} + OH^{\bullet} + O_2 \quad (23)$$

$$O_3 + OH^{\bullet} \rightarrow O_2 + HO_2^{-\bullet} \quad (24)$$

$$M^{(n+1)} + HO_2^{-\bullet} + OH^- \rightarrow M^{n+} + H_2O + O_2 \quad (25)$$

$$M^{n+} + OH^{\bullet} \rightarrow M^{(n+1)} + OH^- \quad (26)$$

Figure 3 compares the removal rates of ciprofloxacin, metronidazole, ofloxacin, and amoxicillin antibiotics in different ozonation methods (Chen and Wang 2021; Xu et al. 2021;

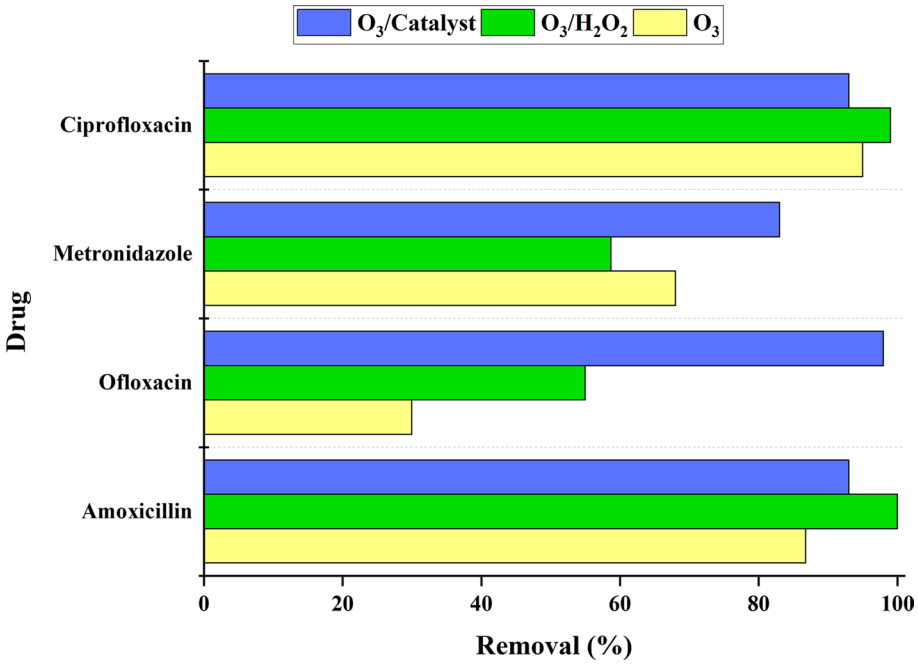

Fig. 3 Comparison of antibiotic removal rate in different ozonation methods

Wulansarie et al. 2020; Nasseh et al. 2020; Zazouli et al. 2019; Cuerda-Correa et al. 2019; Sani et al. 2019; Bahrami Asl et al. 2015). As can be seen, combined methods have higher efficiency and the highest removal is related to O_3/H_2O_2 in the amoxicillin antibiotic.

Sonolysis (Ultrasonic) Sonolysis, often known as ultrasound (US), is a process that destroys contaminants by causing chemical and mechanical changes using waves that have a frequency greater than the range of human hearing (20–10,000 kHz). A sound cavity is created when ultrasonic waves react with dissolved gases (Torres-Palma and Serna-Galvis 2018). The formation, growth and intense collapse of bubbles occur during two main cycles of successive expansion and compression. In the nucleation sites during these cycles, cavitation bubbles occur. They expand by absorbing energy until they reach a critical level, at which point they collapse. Within a few microseconds, this collapse creates shock waves with a pressure of 500 atmospheres and a temperature of about 5000 °C. Free radicals are created when water is split, which causes the contaminant to oxidatively degrade (Adityosulindro et al. 2017).

$$H_2O+))) \rightarrow OH^{\bullet} + H^{\bullet} \tag{27}$$

$$\text{Pollutant} + OH^{\bullet} \rightarrow \text{Degradation} \tag{28}$$

The key benefits of this process include the absence of chemicals, high destruction efficiency, high penetration of generated waves and absence of side pollution and sludge production. However, because the ultrasound method uses a lot of energy, it has a very high operating cost and is not practical on a large scale. Additionally, both mineralization and electrical efficiency are very low (Merabet and Kerboua 2022). Therefore, adding other technologies to this process can boost energy efficiency.

US/Catalyst The degradation efficiency of the sonocatalyst process is greater than that of the sonolysis process alone. This improvement in efficiency may be the result of a synergistic interaction between the increased production of hydroxyl radicals, the higher catalyst dispersion, and the absence of particle accumulation. Due to the increased likelihood of pollutant contact with the generated radicals, the destruction efficiency is increased (Ryu et al. 2021). Increasing the degradation efficiency occurs through several processes: 1. The catalyst facilitates the cavitation process by acting as a nucleation point. 2. The catalyst may be stimulated as a result of the high heat produced during the sonolysis process. Facilitate the generation of electron–hole pairs as a result. 3. Energy absorption activates the catalyst, causing the electron to move from the catalyst's valence band to the conduction band. After then, the recently created electron–hole pairs react with the oxidants already existing to produce reactive radicals.

Additionally, in the sonocatalyst process, intermediary components are made to provide more active sites for cavitation occurrences, which boosts the generation of more reactive radicals. Typically, these radicals are used to create H_2O, $HO_2^{\bullet}$, H_2O_2, and O_2^{-} (Abdurahman et al. 2021).

$$H^{\bullet} + O_2 \rightarrow HO_2^{\bullet} \tag{29}$$

$$2^{\bullet}OH \rightarrow H_2O_2 \tag{30}$$

$$2HO_2^{\bullet} \rightarrow H_2O_2 + O_2 \tag{31}$$

$$H_2O + HO^{\bullet} \rightarrow H_2O_2 + H^{\bullet} \tag{32}$$

US/O₃ The conversion of O_3 into hydroxyl radicals is facilitated by the combination of ultrasound and ozonation. Numerous micro-bubbles are created as a result of ultrasound cavitation, allowing O_3 to enter the liquid phase reaction or the gas–liquid interface. The formation of $OH^{\bullet}$ free radicals for further oxidation of organic pollutants rises due to an increase in mass transfer and O_3 decomposition processes, local high temperature, and ultrahigh pressure during cavity collapse. This results in a higher reaction rate (Xiong et al. 2016). However, the intensity of ultrasonic cavitation is increased by small ozone-derived bubbles (Cui et al. 2011).

$$O_3 + H_2O \rightarrow 2^{\bullet}HOO \tag{33}$$

$$O_3 +^{\bullet} HOO \rightarrow HO^{\bullet} + 2O_2 \tag{34}$$

$$H_2O \rightarrow OH^{\bullet} + H^{\bullet} \tag{35}$$

$$O_3 \rightarrow O_2(g) + O\left(^3p\right)(g) \tag{36}$$

$$O\left(^3p\right)(g) + H_2O \rightarrow 2OH^{\bullet} \tag{37}$$

US/H₂O₂ Because it has access to oxygen, hydrogen peroxide (H_2O_2), one of the potent oxidizing agents, can be a good combination with ultrasonication. In the US/H_2O_2 process, the energy of ultrasonic waves is used to divide H_2O_2, which produces high hydroperoxyl radicals (HOO) and strong hydroxyl radicals ($OH^{\bullet}$). These free radicals have

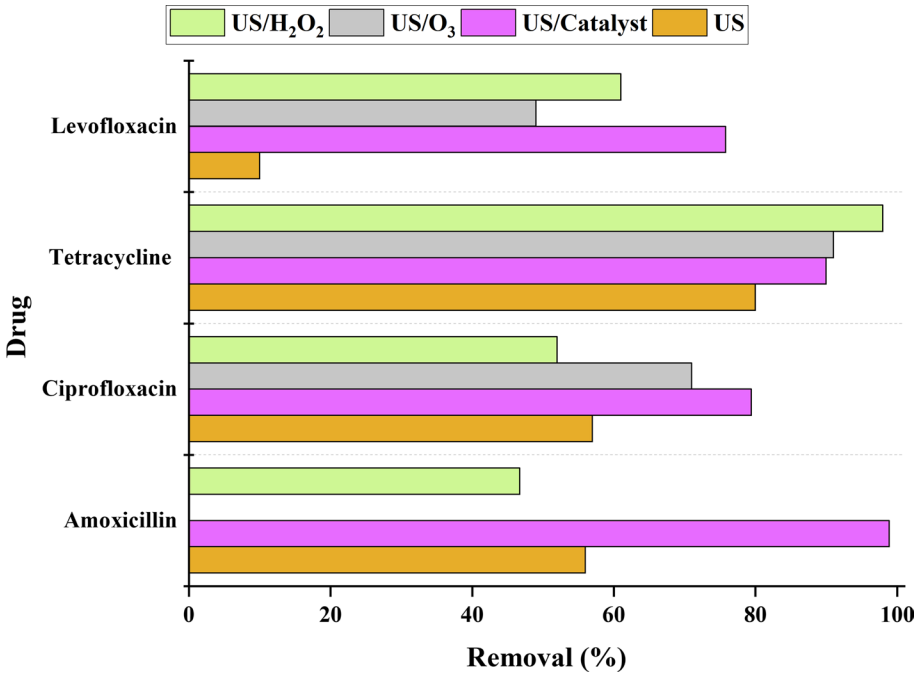

Fig. 4 Comparison of antibiotic removal rate in different sonolysis methods

the power to destroy the pollutant, and the $OH^{\bullet}$ radical can use heat to break down hydrophobic components (Truc and Lee 2019).

$$H_2O_2 \rightarrow 2OH^{\bullet} \quad (38)$$

$$OH^{\bullet} + H_2O_2 \rightarrow H_2O + HO_2^{\bullet} \quad (39)$$

$$HO_2^{\bullet} + HO_2^{\bullet} \rightarrow H_2O_2 + O_2 \quad (40)$$

$$HO_2^{\bullet} + OH^{\bullet} \rightarrow H_2O + O_2 \quad (41)$$

Figure 4 compares the removal amounts of levofloxacin, tetracycline, ciprofloxacin, and amoxicillin antibiotics in different sonolysis methods. The highest removal is related to the sonocatalyst combination method because more

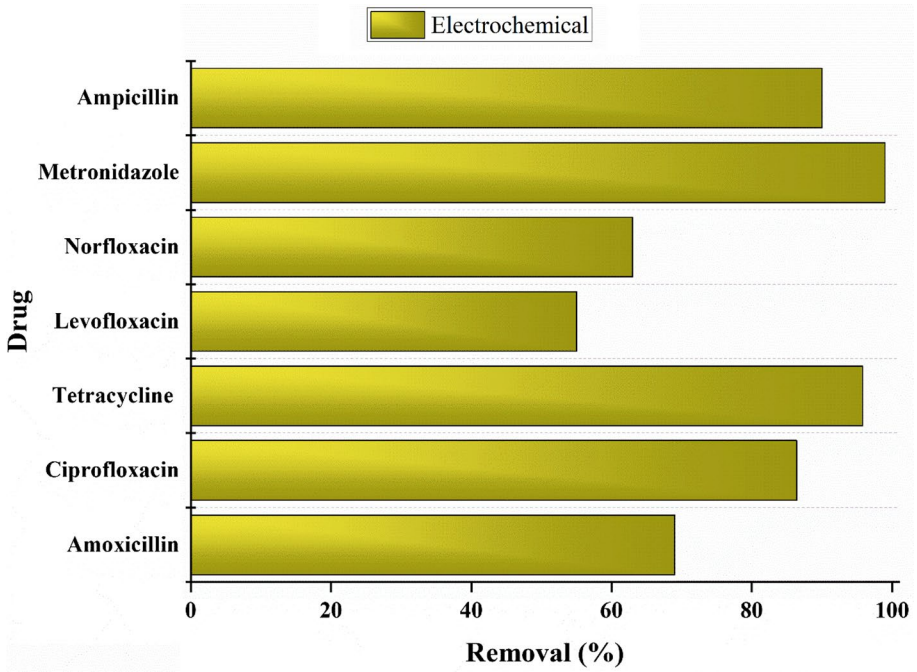

Fig. 5 Comparison of antibiotic removal rate in different electrochemical method

hydroxyl radicals are produced (Kyzas et al. 2022; Ayanda et al. 2021; Stucchi et al. 2020; Reheman et al. 2019; Wei et al. 2015; Wang et al. 2012; De Bel et al. 2009).

Electrochemical One of the AOP processes used to remove organic contaminants from wastewater is electrochemical oxidation (EO). This process has good performance for destroying non-degradable pollutants. Redox reactions may take place in this process at both the anode (such as the oxidation of contaminants) and the cathode (for example, the reduction of heavy metals). Anodic oxidation does not require chemicals and does not produce any secondary pollutants (Donoso et al. 2021). EO oxidizes the pollutants directly and indirectly. In the first technique, there is no participation of other materials, and the oxidation of contaminants with hydroxyl radicals formed on the anode surface occurs directly. This technique uses a highly conductive bare metal as the anode, which can react with the pollutant. The characteristics of the anode metal and the type of pollutant affect this reaction's effectiveness (Brillas 2020).

$$R + M(^{\bullet}OH) \rightarrow M + CO_2 + H_2O \quad (42)$$

In indirect EO, electroactive species formed on the surface of the anode (such as chlorine, hypochlorite, and hypochlorous acid) are used to oxidize organic materials. These electroactive species act as electron mediators between the electrode and organic compounds. The metal ions in this condition have changed into a reactive state with a strong oxidizing capacity that can remove contaminants. The effectiveness of this method depends on the type of pollutant, the concentration of electricity, and the reactivity of the chlorine (Donoso et al. 2021).

$$2Cl^- \rightarrow Cl_2 + 2e^- \quad (43)$$

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \quad (44)$$

$$HOCl \rightarrow H^+ + OCl^- \quad (45)$$

We may list the EO method's benefits as not requiring auxiliary chemicals, generating no secondary pollutants, being able to easily combine with other existing technologies, having a compact operational area, and being environmentally friendly (Chaplin 2018). Figure 5 displays the electrochemical method's removal rates for several antibiotics Mora-Gómez et al. 2019; Xia et al. 2019; Xia and Dai 2018; Ganiyu et al. 2015).

With irradiation

Photolysis (UV based) In the presence of either artificial or natural light, photon absorption is used to carry out the process of photolysis. The energy required for the production of free radicals and electron excitation is provided by these quantized units. Indirect and direct photolysis processes are the two main categories of photolysis (Rahimi et al. 2022). Direct photolysis occurs when an organic substance directly absorbs ultraviolet (UV) light and decomposes. Indirect decomposition, on the other hand, requires the formulation of reactive excited states that result in the degradation of contaminants through sensitivity to light or the use of catalysts (Rezaei et al. 2021). The intensity of UV light, the structure of the pollutant, the quantum yield, and the rate of free radical production all affect the photolysis process' rate of degradation kinetics. The application of this process is constrained by the reduced light penetration caused by the appearance of organic molecules, the formation of side products, and the low range of UV in the electromagnetic spectrum (Kanakaraju et al. 2018).

As a result, complementary techniques are applied to boost the generation of hydroxyl radicals and accelerate the degradation of pollutants, such as the use of catalysts, ozonation, and hydrogen peroxide.

UV/Catalyst Due to unique characteristics, including complete oxidation of pollutants, high efficiency, high surface area, and the presence of an accessible surface for spatial separation in redox reactions, the use of catalysts in AOP processes has recently attracted a lot of interest. This process breaks down a variety of organic and mineral materials using the energy from ultraviolet radiation (Sayadi et al. 2021). UV radiation provides the necessary photon energy for the band gap of the catalyst, which causes the transfer of e^- from the valence band to the conduction band. As a result, an h^+ is created in the VB band. The generated e^- may combine with the oxygen present (in the solution or on the photocatalyst surface) to form superoxide radicals, which then combine with water to form a powerful hydroxyl radical oxidizing agent. Finally, the hydroxyl radical reacts with the pollutant and produces carbon dioxide and water.

$$Catalyst + h\vartheta \rightarrow h^+_{VB} + e^-_{CB} \quad (46)$$

$$e^-_{CB} + O_2 \rightarrow O_2^{\bullet -} \quad (47)$$

$$O_2^{\bullet -} + H_2O \rightarrow HO_2^{\bullet} + OH^{\bullet} \quad (48)$$

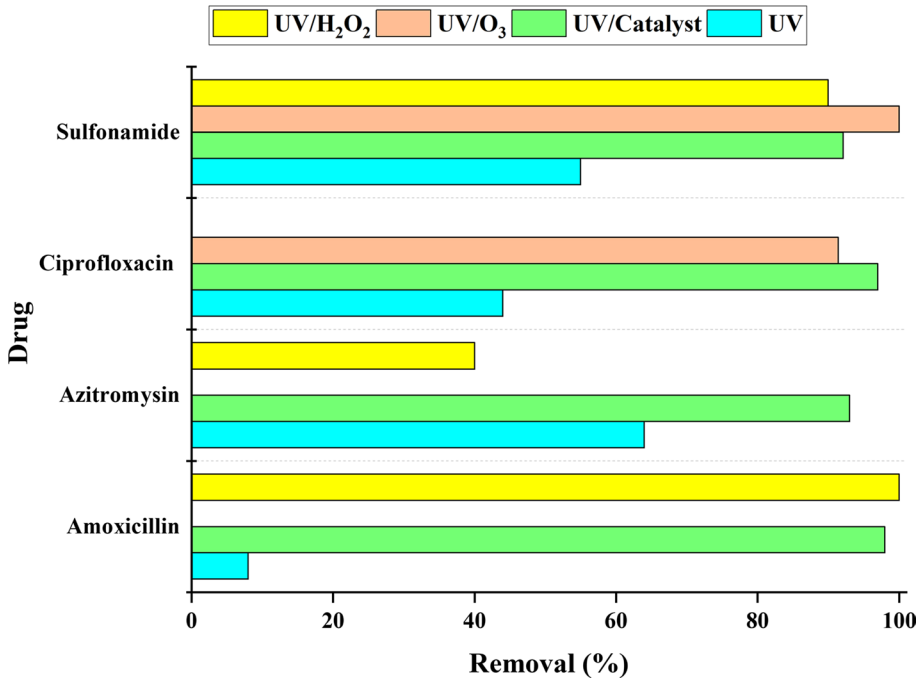

Fig. 6 Comparison of antibiotic removal rate in different photolysis methods

$$OH^{\bullet} + \text{Organic Pollution} \rightarrow H_2O + CO_2 \quad (49)$$

UV/O$_3$ The dissolved ozone is photolyzed by ultraviolet light in the UV/O_3 process, and the oxygen atom then combines with the water to generate H_2O_2. The excited peroxide then splits into two $OH^{\bullet}$ radicals. Finally, the system's generated $OH^{\bullet}$ oxidizes organic materials, causing them to break down into small molecules (Cuerda-Correa et al. 2019).

$$O_3 + H_2O + h\vartheta \rightarrow H_2O_2 \quad (50)$$

$$H_2O_2 + h\vartheta \rightarrow 2OH^{\bullet} \quad (51)$$

This technique can degrade organic compounds that cannot be removed by ozonation or UV light alone because it produces hydroxyl radicals with a very high oxidation power. Because the radicals produced in the UV/O_3 process have a high redox potential. Other features of this process are the high degree of mineralization, the 2.8 V redox potential, and the capacity to destroy bacteria and pathogens (Lu et al. 2022).

UV/H$_2$O$_2$ When H_2O_2 and UV are used together, the photolysis of H_2O_2 can generate a radical, which causes the oxidation of pollutants. It is a non-selective oxidant that has very high reactivity and can break down a variety of contaminants (Nguyen et al. 2019). In this reaction, a decrease in the UV wavelength and a rise in the H_2O_2 concentration cause an increase in the molar absorption coefficient of H_2O_2, which causes more H_2O_2 to decompose and more to be produced. Because of its electrophilic nature, it can attack and destroy organic contaminants (Sharma et al. 2019).

$$H_2O_2 + h\vartheta \rightarrow 2OH^{\bullet} \quad (52)$$

The amount of removal of various antibiotics using the photolysis process is shown in Fig. 6. The removal efficiency can be increased by up to 100% by combining the photolysis process with ozonation, H_2O_2, and a catalyst (Liu et al. 2021; Palmisano et al. 2015).

Fenton based In the Fenton process, the hydroxyl radical is formed from the reaction of iron ions (Fe^{2+}) and hydrogen peroxide (H_2O_2) (reaction 53). An acidic environment is suitable for this process and accelerates it. The Fenton process is also created by the redox catalytic reaction of Fe^{3+}/Fe^{2+} (reaction 54). By generating free radicals and Fe^{2+}, additional intermediates also keep the Fenton reaction cycle intact (reactions 55–62) (Fischbacher et al. 2017). Oxidative radicals $OH^{\bullet}$ can produce water and carbon dioxide by decomposing organic pollutants. The concentration of Fenton's reagent, the pH of the environment, and the amount of pollutants all affect how effectively this degradation occurs (Zhang et al. 2019).

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^{\bullet} + OH^{-} \quad (53)$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2^{\bullet} + H^{+} \quad (54)$$

$$Fe^{2+} + H_2O + H^{+} \rightarrow Fe^{3+} + H_2O + OH^{\bullet} \quad (55)$$

$$Fe^{3+} + HO_2^{\bullet} \rightarrow Fe^{2+} + O_2 + H^{+} \quad (56)$$

$$Fe^{3+} + O_2^{\bullet -} \rightarrow Fe^{2+} + O_2 \quad (57)$$

$$Fe^{2+} + H_2O_2 \leftrightarrow FeOOH^{+} + H^{+} \quad (58)$$

$$FeOOH^{+} + H^{+} \leftrightarrow Fe(H_2O_2)^{2+} \quad (59)$$

$$Fe^{2+} + H_2O_2 \leftrightarrow Fe(H_2O_2)^{2+} \quad (60)$$

$$Fe(H_2O_2)^{2+} \rightarrow Fe^{3+} + OH^{\bullet} + OH^{-} \quad (61)$$

$$Fe(H_2O_2)^{2+} \rightarrow FeO^{2+} + H_2O \quad (62)$$

It should be noted that the Fenton method has some limitations as well. For instance, the pH range should be acidic and between 2 and 4. However, hydrogen peroxide is a dangerous gas that can explode. Production iron sludge is also considered as secondary pollution. These restrictions are all expensive and risky (Huang et al. 2017). As a result, modifying the Fenton process can improve the efficiency of pollutant removal while decreasing costs and improving environmental compatibility.

Photo-Fenton The addition of UV rays to the Fenton process increases the catalyst reduction and the hydroxyl radical's production, which ultimately increases the oxidation efficiency. In the photo-fenton process, the hydroxyl radical is produced by three methods, which are (Ameta et al. 2018):

1. Photodecomposition of hydrogen peroxide
2. Increasing reduction of iron (III) by hydrogen peroxide
3. Photoreduction of ferric iron to iron (II)

Considering that the photo-fenton process has the potential to use a variety of UV areas as a source of light energy, in addition to Fenton cases, the oxidation efficiency is dependent on the UV light's intensity. The UV-B region, with a wavelength range of 285–315 nm, has the best efficiency due to the highest levels of $OH^{\bullet}$ generation and $Fe(OH)^{2+}$ radiation absorption.

The photo-fenton process, like the Fenton process, performs better at low pH levels, and at pH levels higher than 4, the oxidation efficiency decreases due to iron hydroxide precipitation and a reduction of UV rays' transmission. One of the advantages of the photo-fenton method is its higher efficiency compared to the Fenton process. Although there is not much difference in the pollutant removal efficiency compared to the Fenton process, the increase in mineralization in this process has been highly noticeable and, in some cases, up to double (Clarizia et al. 2017).

$$Fe^{3+} + h\nu + H_2O \rightarrow Fe^{2+} + OH^{\bullet} + H^{+} \quad (63)$$

$$FeOH^{2+} + h\nu \rightarrow Fe^{2+} + OH^{\bullet} \quad (64)$$

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^{\bullet} + H^{+} \quad (65)$$

Sono-Fenton The combination of Fenton and ultrasonic processes can accelerate the mineralization of pollutants and increase their removal efficiency. This rise is brought on by the production of more radicals, the increase in the contact between the pollutant and existing radicals, and the increase in the reproduction of iron ions. The water splits as a result of the high ultrasonic frequencies used in this process. Free radicals produced in this process include $OH^{\bullet}$, $HO_2^{\bullet}$ and $H^{\bullet}$, which are very reactive and decompose the pollutant (Ammar 2016).

$$Fe^{3+} + H_2O+))) \rightarrow Fe^{2+} + H^{+} + OH^{\bullet} \quad (66)$$

$$Fe^{3+} + H_2O_2+))) \rightarrow Fe^{2+} + HO_2^{\bullet} + H^{+} \quad (67)$$

Electro-Fenton In the electro-Fenton process, pollutants are decomposed by anodic oxidation and Fenton's reagents, and their mineralization increases significantly with the produced hydroxyl radicals. In this process, H_2O_2 is formed on the cathode and Fe^{2+} is formed on the anode. Which is finally associated with the production of $OH^{\bullet}$ as a strong oxidizer that decomposes the pollutant. Additionally, a trace amount of $OH^{\bullet}$ is produced when water on the anode oxidizes. Electrolyte, oxygen sparing rate, Fe^{2+}/Fe^{3+} concentration, H_2O_2 concentration, initial concentration of pollutants, pH, electrode materials, and electrode distance all affect how effective this process is (He and Zhou 2017).

$$\text{Cathode}: O_2 + 2H^{+} + 2e^{-} \rightarrow H_2O_2 \quad (68)$$

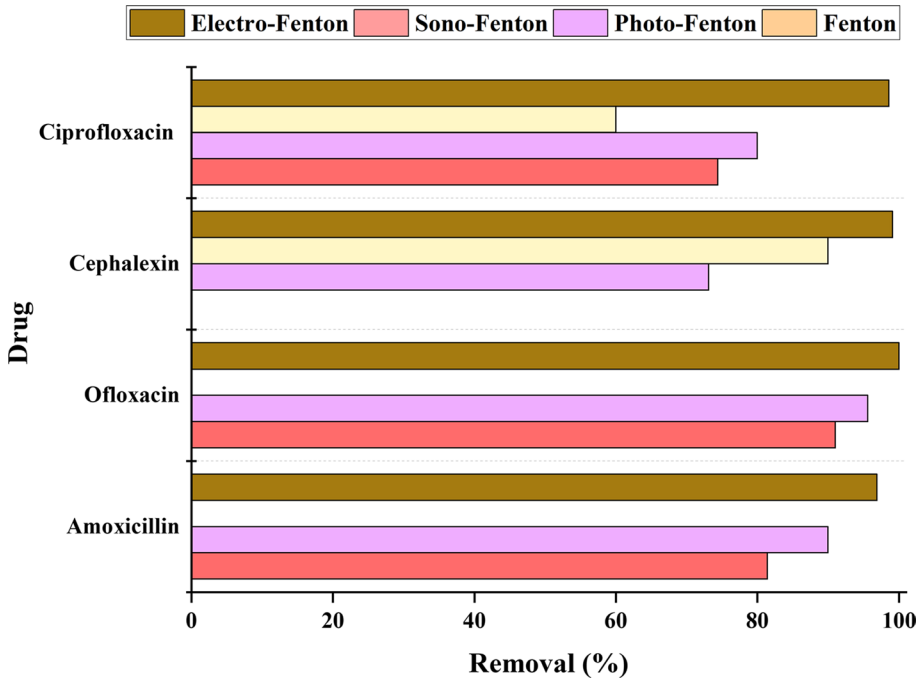

Fig. 7 Comparison of antibiotic removal rates in different Fenton method

$$\text{Anode}: Fe \rightarrow Fe^{2+} + 2e^{-} \quad (69)$$

$$Fe^{2+} + H_2O_2 + E \rightarrow Fe^{3+} + OH^{\bullet} + OH^{-} \quad (70)$$

$$\text{Anode}: H_2O \rightarrow OH^{\bullet} + H^{+} + e^{-} \quad (71)$$

Figure 6 compares the amount of removal of ciprofloxacin, cephalexin, ofloxacin and amoxicillin antibiotics in different ozonation methods (Cao et al. 2022; Basturk et al. 2021; Gou et al. 2021; Guerra et al. 2019; Al-Musawi et al. 2019; Perini et al. 2017). As can be seen, the most elimination is related to electro-fenton in all antibiotics.

Outlook

The investigation done in this study suggests that using AOP methods could be very beneficial and efficient for removing organic pollutants, especially pharmaceuticals. In general, the type of produced oxidative radical, the concentration and rate of radicals' formation, the concentration and structure of the pollutant, and the pH of the environment are the most important factors that affect the efficiency of pollutant degradation in these processes. The design of a system that has characteristics such as high destruction efficiency, no secondary pollutant production, compatibility with the environment, high mineralization, low cost, and ability to be combined with other technologies should thus be given serious consideration. According to a comparison between different systems (Figs. 3, 4, 5 and 6), hybrid systems have the ability to produce more oxidizers and improve the effectiveness of the degradation process. The more powerful oxidative radicals are created in these mixed systems, such as $OH^{\bullet}$ (oxidative power of 2.8 V), O_3 (oxidative power of 2.07 V), and H_2O_2 (oxidative power of 1.75 V), the more efficient the process will be. Additionally, by generating a synergistic effect, a system that uses many degradation components, such as UV light, ultrasonic radiation, and electric radiation, can be more effective (Fig. 7).

Farhadi et al. (2021) were able to degrade ibuprofen up to 99.5% by designing a fourfold system US/UV/H_2O_2/zeolite-titanate photocatalyst (at a concentration of 0.05 mM H_2O_2, pH of 5, UV power of 6 W, photocatalyst dose of 1 g/L, and contact time of 100 min). In another study, the antibiotic berberine hydrochloride (BH) was removed by 94.1% using the O_3/UV/H_2O_2 process under the optimal conditions of initial pH 7, a dose of 3 mM H_2O_2, a dose of 10.3 mg/min of O_3 and a reaction time of 45 min (Qin et al. 2015). Also, the Sono-Electro-Fenton process was able to remove 97.5% of cefixime antibiotic under optimal conditions of pH 3.07, H_2O_2 concentration of 0.85 ml/liter, voltage of 15 V, the initial antibiotic concentration of 10.4 mg/liter, and electrolysis time of 81.5 min (Hasani et al. 2020). The results of these

studies also show that the destruction efficiency is higher in triple or quadruple systems.

However, given that the majority of studies have been conducted in laboratories, future studies should focus on methods that can lower the cost of operation when using hospital, urban, and industrial wastewater and increase the effectiveness of destruction. Additionally, most of the systems in the research we studied are closed, but continuous systems have to be designed. The by-products and toxicity of the sludge produced by the wastewater treatment process are another critical factor. Therefore, the system must be environmentally friendly and not create any hazardous secondary pollutants. The ozonation process is currently applied on a large scale in industry. Evaluating the potential of other technologies is also necessary. For instance, catalysts can be added to the purification system to remove some unique, hazardous, and resistant pollutants while also making it more cost-effective. TiO_2, a photocatalyst that is highly reactive, stable, non-toxic, safe for the environment, and widely accessible, can be a good choice for this use. In addition, by altering the structure of photocatalysis, the use of noble metals, intermediates, lanthanides, and heterogeneous connections can considerably improve the capacity to target pollutants.

Conclusion

Antibiotics, which are widely spread in various amounts in aquatic environments, are now recognized as emerging contaminants and have attracted a lot of attention in recent years. Drugs are able to enter the environment even in small concentrations and harm living species since current wastewater treatment systems are not designed to remove pharmaceutical pollutants. They could threaten both the ecosystem and human health by moving up the food chain. Additionally, they lead to the growth of bacteria and genes resistant to antibiotics over time. Despite the world's rapid technological and scientific advancement, the problem of pharmaceutical pollutants remains unresolved, and the knowledge currently accessible for pharmaceutical risk assessment and management is insufficient. Recent trends in the development of advanced oxidation technologies offer a great chance to quickly and effectively remove pharmaceutical contaminants. The efficient production of ROS $\left(O_2^{\bullet-},\ OH^{\bullet}\text{ and }HO_2^{\bullet}\right)$ and the harmless degradation of organic pollutants are the foundation of AOPs' activities. As an emerging and efficient wastewater treatment method, AOPs have the advantages of no secondary pollution, high reaction speed, high mineralization efficiency, and high oxidation capability. The effects of drug exposure and their interactions, however, require more study. Consideration should also be given to the risk assessment of pharmaceuticals for human health and the related ecotoxicological risk assessment.

In this study, methods based on ozonation (O_3, O_3/H_2O_2, O_3/Catalyst), ultrasonic radiation (Sonolysis, US/Catalyst, US/O_3, US/H_2O_2), electrochemical, ultraviolet radiation (Photolysis, UV/Catalyst, UV/O_3, UV/H_2O_2), and Fenton (Fenton, Photo-Fenton, Sono-Fenton, Electro-Fenton) were investigated. The findings demonstrated that numerous factors, including the dose of hydroxyl, the concentration of medicinal substances, the mode of operation, and water quality parameters, had an effect on the amount of ozone oxidation in the ozonation method. But highly toxic by-products may be produced. Therefore, modification of ozonation with H_2O_2 as a reaction initiator or addition of a catalyst as a facilitator of free radical production will lead to better removal of pollutants and more mineralization. Sonolysis is also a very fascinating method for researchers because it doesn't require the addition of chemicals, has high generated wave penetration, high destruction efficiency, and generates no side pollutants or sludge during the process. But due to high energy consumption and high operating costs, it is not economical on a large scale. Therefore, combining this process with other technologies such as catalyst, O_3 and H_2O_2 can increase energy efficiency by producing more hydroxyl radicals. On the other hand, the electrochemical method has many advantages, such as no need for auxiliary chemicals, no production of secondary pollutants, the possibility of easy combination with other existing technologies, small operating space, and compatibility with the environment, but it is expensive and requires special equipment. Photolysis, which depends on UV light intensity, pollutant structure, quantum yield, and the rate of free radical generation, is another approach being researched to remove pharmaceutical contaminants. The reduction of light penetration due to the appearance of organic molecules, the production of by-products, and the low range of UV in the electromagnetic spectrum, limits the application of this process. As a result, complementary methods such as the use of catalysts, ozonation and hydrogen peroxide are used to increase the production of hydroxyl radicals and increase the level of pollutant degradation. The last method investigated is Fenton, and in this method, the degradation efficiency depends on the concentration of Fenton's reagent, the pH of the environment, and the pollutant concentration. Modifying the Fenton process with UV, ultrasonic and electro-Fenton can improve the

efficiency of pollutant destruction while reducing the cost and increasing compatibility with the environment.

The findings of this research indicate that, as a result of the synergistic effect, the combined oxidation capacity of various processes is typically higher than that of a single process. In general, the combined approaches of various AOPs degraded pollutants more effectively than those of individual AOPs. Future studies should therefore concentrate on improving AOPs, hybrid systems, understanding the processes that lead to oxidative radical formation and pollutant degradation, continuous and complex systems, and reducing operational costs.

Declarations

Conflict of interest The authors have no conflicts of interest to declare that are relevant to the content of this article.

Human or animal rights This article does not contain any studies with human participants or animals performed by any of the authors.

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