Applied Water Science (2024) 14:164
https://doi.org/10.1007/s13201-024-02223-1

REVIEW ARTICLE

# Penicillin removal from the aqueous environment based on AOPs/challenges and outlook. A review

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Received: 7 March 2024 / Accepted: 5 June 2024 / Published online: 27 June 2024


**Abstract**
Today, the deterioration of water quality is still a big concern among researchers, whose essential strategy is to develop water purification processes. The presence of contamination of emerging concerns has become a challenging issue, their toxicity, persistence in the aquatic environment, and their mass accumulation at low concentrations have had adverse effects on human health and aquatic organisms. Biological disposal or improper disposal, these pollutants cause serious damage to the population of non-target groups. Penicillin is a broad-spectrum group of beta-lactam antibiotics that has caused concerns for human health and the environment due to its slow decomposition in water sources. This review article focuses on the application of advanced oxidation processes to eliminate these antibiotics. Several studies have investigated the effects of different parameters on species with both ultraviolet and non-ultraviolet light. The results have been promising, with an average efficiency above 80% for these processes. Despite the limitations of various methods, the knowledge gap in future studies has been addressed by proposing the use of Fenton, ultrasound method and Integrated processes like Synergistic Remediation-Advanced Oxidation Processes.

**Keywords** Penicillin · Advanced oxidation processes (AOPs) · Antibiotic · Fenton · Ultrasound · Human health

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## Introduction

The existence of antibiotics in the aqueous Environment is regarded as an emerging problem, generally due to the pharmaceutical industry and the application of veterinary and human medicines (Le-Minh et al. 2010; Fuoco 2012; Klavarioti et al. 2009). More than 60% of these compounds remain undegraded in the human or animal body and are mostly eliminated as active substances (Choi et al. 2008; Chee-Sanford et al. 2001; Xiong et al. 2022).

Among the more than 200 antibiotics applied to treating infections, penicillin (PEN) is one of the most extensively utilized beta lactams (Abdipour and Hemati 2023) Penicillin's are among the first drugs developed to treat diseases caused by pathogenic bacteria and are still widely prescribed. 95 and 81 years have passed since the discovery of penicillin by Fleming and its widespread clinical use, but it is still one of the most commonly prescribed antibiotics in human society (Ligon 2004; Gaynes 2017; Liu et al. 2022). Ecological disorders due to the appearance of antibiotics in the water resources, including toxicity to algae, and disruption of the stability of the ecosystem through the proliferation of resistant bacteria, have led to the consideration of strict standards for the entry of these organic pollutants into influent waters (Schmitt et al. 2006; Kümmerer 2009; Young et al. 2013; Kamani et al. 2024; Lan et al. 2022). With the expansion of infectious diseases, the generation of different antibiotics has also risen (Kord Mostafapour et al. 2016). Thus, inefficient treatment of wastewater containing these compounds can have disastrous consequences for the environment, including unfavorable water quality (Wang et al. 2015; Norzaee et al. 2018; Asgari et al. 2023; Ganji et al. 2024; Jahangiri et al. 2023). Several factors, including the high efficiency of this antibiotic in the treatment of bacterial infections and the reasonable cost, have caused the annual consumption of penicillin to be more than 20,000 tons (Ma et al. 2018; Zhou et al. 2018; Hosseini and Faghihian 2019).

Conventional purification methods cannot very efficiently remove antibiotics, including penicillin (Joss et al. 2006) In order to remove penicillin in the direction of environmental sustainability, researchers use different methods, including adsorption (Ahmadi et al. 2017, 2018), membrane separation (Wang and Lim 2012), biological processes (Yazdi et al. 2018), dissolved air flotation (Ahmadi and Mostafapour 2017; Kord Mostafapoor et al. 2017; Zhou et al. 2024), electrolysis (Peterson et al. 2012; Homem and Santos 2011), oxidation (Martinez 2009), and biodegradation (Watkinson et al. 2007).

Several research projects have been carried out in recent decades in the field of removing antibiotics such as penicillin from wastewater. Some of these studies have been effective in eliminating penicillin. In addition, modern wastewater treatment methods that use advanced oxidation techniques are preferred due to the operational difficulties and high cost of conventional methods (Kamani et al. 2024; Lee and Gunten 2010; Robinson et al. 2001; Forgacs et al. 2004) Advanced oxidation processes (AOPs) are physicochemical elimination procedures that have been utilized in recent decades for the treatment of organic pollutants like antibiotics in drinking water and wastewater (Rubio-Clemente et al. 2014; Cruz et al. 2013; Wang et al. 2024). The oxidation power of the hydroxyl radical is much greater than the usual oxidizing agents (Tekin et al. 2006; Song et al. 2023). This reaction continues until the materials are fully mineralized. The reaction occurs at average temperature and pressure and does not harm the chosen materials. In addition, it can oxidize nearly all kinds of organic substances without limitation to their composition or group (Comninellis et al. 2008). The degradation of organic compounds is hastened by the radicals produced in the AOP process (Velegraki and Mantzavinos 2015; Samadi et al. 2024a). Ultrasound is a promising non-chemical process for degrading antibiotics. It uses high-frequency sound waves to create OH and pyrolysis without catalysts or oxidants. The degradation of antibiotics in this method is based on the interaction of high-frequency ultrasound with the water medium to produce OH or through pyrolysis (Kıdak and Doğan 2018; Serna-Galvis et al. 2016) Since no literature review have been published on the removal of penicillin based on advanced oxidation, this review is the first to cover the gap caused by the lack of detailed investigation of this absolutely essential topic with the aim of identifying advanced oxidation methods for the removal of this antibiotic. Determining the influential factors in the removal of penicillin, the advantages and disadvantages of each method, the challenges facing each process, the degradation mechanism of this antibiotic will have kinetic studies and a critical approach focusing on the future perspective.

## Toxicology of PEN

Penicillin is a group of beta-lactam antibiotics whose features are indicated in Table 1. This group consists of amoxicillin, ampicillin, dicloxacillin, cephalosporin & oxacillin as well as penicillin types V and G. and which have a variable R group in their composition. Studies have demonstrated that it has high toxicity compared to other antibiotics (Guo et al. 2016). The unit toxicity is an $EC_{50}$ value of 0.13 mg/L (Tong et al. 2014). For Synechococcus leopolensis bacteria, significant toxicity at nano concentrations has been demonstrated (Andreozzi et al. 2004). Acute toxicity tests with Daphnia magna illustrate that penicillin at a dilution of about 50% can cause mortality of almost 90% in untreated effluents (Arslan-Alaton and Caglayan 2006).

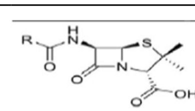
## Sources of penicillin

Penicillin is an antibiotic that can be derived from two sources: natural and synthetic.

Natural penicillin

- Natural penicillin is obtained from the Penicillium mold, specifically Penicillium chrysogenum.
- It is produced through the fermentation of the mold in a controlled environment.

**Table 1** Features of PEN antibiotic

| | |
|---|---|
| Structure of PEN |  |
| Molar mass | $\frac{g}{mol}$ 4/334 |
| boiling point | 212–209 °C |
| Density | $\frac{g}{CM3}$ 41/1 |
| Solubility in water | 15–10 mol/L in 25 °C |

Synthetic penicillin

- Synthetic penicillin is chemically synthesized in a laboratory setting, without the need for mold fermentation.
- It is created through a series of chemical reactions using different starting materials.

## Production aspects

### Natural penicillin

- Requires the cultivation and fermentation of the Penicillium mold.
- The process can be time-consuming and is subject to variations in yield due to factors like environmental conditions and the quality of the mold.

### Synthetic penicillin

- Involves chemical synthesis under controlled laboratory conditions, allowing for greater control over the process and yield.
- The production of synthetic penicillin is generally more predictable and less susceptible to external factors.

## Differences and characteristics of wastewater

### Wastewater from natural penicillin production

Contains organic compounds from the fermentation process.

May contain residual nutrients used to cultivate the Penicillium mold.

### Wastewater from synthetic penicillin production

May contain chemical by-products and solvents used in the synthesis process.

Generally, requires more stringent treatment due to the presence of synthetic chemicals.

### Other differences

- Purity and consistency

  Synthetic penicillin is generally purer and more consistent in its composition compared to natural penicillin.

- Cost and scalability

  Synthetic penicillin production can be more cost-effective and scalable due to the controlled nature of the synthesis process.

### Data comparison

When comparing the two sources of penicillin, factors such as production yield, environmental impact, and the composition of wastewater should be considered to provide a comprehensive analysis of their differences.

## AOPs method

These processes are a new chemical treatment technique that is extensively applied in water and wastewater treatment (Samadi et al. 2024b). The inefficiency of conventional methods on the one hand and the high potential of AOP to eliminate antibiotics on the other, have caused researchers to focus on these methods. The basis of advanced oxidative processes, the generation of (–OH), which is mainly caused by $H_2O_2$, $O_3$, photocatalysis, or oxidants by (UV) light or sunlight. Hydroxyl radicals, as powerful and non-selective chemical oxidants, start attacking organic compounds after their formation and to the full degradation of these organic compounds (Kurt et al. 2017)

The function of hydroxyl radicals in the elimination of penicillin is described as follows. These radicals attack penicillin and take up a hydrogen atom (R–H), which causes the generation of an organic radical (−R). The reaction is illustrated in Eq. 1. In order to theoretically form many products, AOPs must totally mineralize organic compounds (MOCs) to form $CO_2$ and $H_2O$ (Samadi et al. 2024b; Cuerda-Correa et al. 2019; Delavari et al. 2024)

$$R - H + \bullet OH \rightarrow H_2O + \bullet R \tag{1}$$

In recent decade, advanced oxidation processes (AOPs) have been extensively applied in the field as guaranteed Techniques for the elimination of various antibiotics, including PEN G (Azari et al. 2022). because of their high oxidation potential and exceptional efficiency in the oxidative degradation of organic calcium compounds (Ahmadi et al. 2018), as well as their flexibility and wide range of applications. AOPs encompass a number of very successful ways of approaching wastewater treatment and management (Klavarioti et al. 2009; Cuerda-Correa et al. 2019). Although AOPs have attractive advantages and produce a small amount of sludge, they have the drawback of being an expensive method. They are highly selective for high removal efficiency of various pollutants. It should be noted that this method alone is not very effective, but the use of persulfate (PS), nanoparticles and catalytic particles has the ability to improve its efficiency. AOPs are divided into non-photochemical and photochemical groups. The non-photochemical subgroup can be divided into ozone, Fenton, wet oxidation, etc. Photochemical oxidation processes are both homogeneous and heterogeneous (Litter 2005).

## Non-photochemical processes

Among all the methods that are not based on UV, studies have focused on ozone (Kurt et al. 2017). Ozone as a strong oxidant has been considered for many years in modern wastewater treatment approaches (Gottschalk et al. 2010). In the ozone process, there are two manners of perfecting the oxidation mechanism: the direct way, which comes from the interaction of ozone with dissolved substances, and the other method, the radical approach, which appears as a consequence of the interaction between free-radical reactions. It is created from the degradation of ozone (hydroxyl radicals) by means of soluble compounds. These two mechanisms relate to the degradation of the compound to various variables such as the type of contaminant, ozone concentration, or pH of the solution. The breakdown of ozone leads to the Radical formation ($HO_2$• and •OH) reacting with organic material, for example micro-organisms, hydrogen ions, and hydroxide. This technique is applicable to the purification of wastewater from containing penicillin. Several factors like temperature, pH, ozone dose, and impurity concentration affect the performance of the ozone process. To attain a high level of elimination efficiency in ozone treatment, a pH value of above 8 is preferable, as this result in a rapid breakdown of the ozone into free hydroxyl radicals. To be the optimal pH for the oxidation of all organic components, including antibiotics, is 8–10 (Konsowa 2003). Below are the Facilitated reaction mechanisms for ozone at elevated pH values, and Table 2 shows the studies based on this for the removal of penicillin.

$$3O_3 + H_2O \rightarrow 2 \bullet OH + 4\, O_2 \quad (2)$$

The challenges and disadvantages of ozonation in the removal of penicillin are:

1. The possibility of producing dangerous chemicals such as formaldehyde and nitrous amine
2. The need for careful monitoring in the process
3. High cost of the process
4. Delay in elimination of penicillin
5. Change in water quality such as change in pH

Cheapness, low dependence on organic materials, high solubility, and extreme stability of radical production have led to the application of persulfate in antibiotic removal processes.

In order to check the excess of oxidant, the molar ratio of oxidant to pollutant (i.e., rox) is taken into account. Acceptable performance for rox between 8 and 10 has been obtained in the removal of antibiotics with persulfate.

In the process of penicillin antibiotic degradation, persulfate increases the degradation efficiency. And the destruction of penicillin due to the UV/PS process is far more effective because.

The possibility of additional $SO^{4-\bullet}$ formation increases.

Persulfates, such as sodium persulfate and potassium persulfate, offer several advantages compared to other Advanced Oxidation Processes (AOPs) in the removal of penicillin from wastewater. Some of the key advantages include:

### Strong oxidizing power

Persulfates exhibit strong oxidative capabilities, which enables them to effectively degrade a wide range of organic contaminants, including penicillin, in wastewater.

### Activation flexibility

Persulfates can be activated by various means, such as heat, UV radiation, or the addition of catalysts like iron or manganese compounds. This flexibility allows for tailored activation methods based on the specific requirements of the wastewater treatment process.

#### Rapid reaction rates

When properly activated, persulfates can rapidly oxidize organic compounds, leading to faster degradation of penicillin compared to some other AOPs.

#### Stability

Persulfates are relatively stable in storage and transportation, making them convenient for use in wastewater treatment processes.

#### Compatibility with various wastewater conditions

Persulfates are effective across a wide range of pH levels and can be utilized in both acidic and alkaline wastewater treatment systems, providing versatility in application.

#### Minimal by-products

In some cases, persulfate oxidation results in fewer harmful by-products compared to alternative AOPs, contributing to a more environmentally friendly treatment process.

In conclusion, persulfates offer advantages in terms of their strong oxidizing power, activation flexibility, rapid reaction rates, stability, compatibility with various

**Table 2** Studies based on non-photochemical processes with the aim of eliminating penicillin derivatives

| Antibiotic name | Type of AOP | Concentration (mg/L) | Operation condition | Removal efficiency (%) | References |
|---|---|---|---|---|---|
| AMX | O3/Fe2+ | 20 | Time = 5 min<br>Rate = 60 L/h<br>pH = 3<br>$Fe^{2+}$ = 10 mg/L | 33 | Souza et al. (2018) |
| AMX | O3/$H_2O_2$ | 105 | [H2O2] = 0−20mM; pH = 3−11<br>Time = 30−180min; ozone stream = 0.27g O3/h | 100 | Elmolla and Chaudhuri (2010) |
| PEN G | Fenton-like ($Fe^{3+/}H_2O_2$ | 40 | pH = 3<br>Time = 5−30 min<br>[H2O2] = 5−40 mM<br>$Fe^{3+}$ = 5 mg/L | 44 | Arslan-Alaton and Gurses (2004) |
| PEN G | Persulfate/Heat activation process | 20 | Time = 15−90 min; temperature = 313−353 K<br>Persulfate = 0.05−0.5 mM;<br>pH = 3−11 | 98 | Norzaee et al. (2018) |
| PEN G | O3 | 50 | pH = 3.5; Time = 0−5 min<br>O3feed rates = 600–2600 mgh; | COD = 82 | Arslan-Alaton and Caglayan (2005) |
| AMX | Combined system with Fenton activated sludge procedure | 5 – 650mg/L | H2O2/Fe2 + molar ratio = 1−5; Reaction time = 1−100h; Temperature = 30−80 °C; pH = 1−9;<br>Temperature = 298 K | 88.79 | Guo et al. (2015) |
| Sulfamethoxazole | O3 | 30 | pH = 2 and 8,<br>flow = 3.0 ml min−1, T = 25 °C,<br>gas flow = 8.5 gNm − 3 | 80 | Aguinaco et al. (2014) |
| Amoxicillin | Fenton | 150 | pH = 3 $Fe^{2+}$ = 10 mg/L | 85 | Chaudhuri et al. (2013) |
| Sulfamethoxazole (SMT) | Fenton | 100 | conc: 5 mg/ L<br>pH = 5 | 88 | Miralles-Cuevas et al. (2013) |
| Sulfamethoxazole (SMT) | O3 | 50 | time = 60 min<br>($1.3 \times 10^4$ M) O3 dosag | 70–90% TOC | Farkas et al. (2024) |

wastewater conditions, and minimal generation of harmful by-products, making them a favorable choice for the removal of penicillin and other organic contaminants in wastewater treatment applications (Lin et al. 2022).

## Photochemical processes

AOPs have a great potential to generate OH radicals, which are essential for UV radiation (Jahantiqa et al. 2020). They are based on the absorption of oxidants. Photochemical reactions are divided into two groups: isotropic processes (ozone radiation/ultraviolet radiation, hydrogen peroxide/ultraviolet radiation, and photo-Fenton) or Complicated and heterogeneous procedures, which encompass.

Photocatalytic (Parsons 2004; Kamani et al. 2023a, 2023b). Fenton filtration is extensively applied in wastewater treatment and is effective in eliminating several organic compounds. In the photo-Fenton reaction, the Fenton reagent is mixed with ultraviolet (UV) light (Yonar 2011). The object of Add UV light to the Fenton process is to generate more hydroxyl radicals so that they can exhibit their extraordinary ability to degrade penicillin and its derivatives. This is done by targeting them (Buthiyappan et al. 2015). The aim of adding persulphate as a catalyst for the process is to activate and actualize the power of oxidation of some pollutant particles, generating hydroxyl radicals from $H_2O_2$. The reaction is illustrated in Eq. 3.

$$2\,O_3 + H_2O_2 \rightarrow 2\,OH + 3O_2 \tag{3}$$

The oxidation of organic substrates by Fe (II) and hydrogen peroxide is the Fenton process. This method is the most effective way to oxidize pollutant, for example penicillin (Barbusiński 2009). The mechanism of the process is described by Eqs. (4–8) also Fig. 1 illustrates a graphical representation of the elimination of penicillin by the Fenton process.

$$Fe^{2+}H_2O_2 \rightarrow Fe^3 + HO\bullet \ + HO- \quad (4)$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^2 + HO_2 \bullet + H \quad (5)$$

$$RH + \ HO\bullet \rightarrow R \bullet + H_2O \quad (6)$$

$$Fe^3 + R\bullet \rightarrow Fe^2 + R^+ \quad (7)$$

$$R^+ + HO- \rightarrow ROH \quad (8)$$

The main challenges of the photochemical process of penicillin removal are:

1. Choosing the right process: This is achieved by evaluating the processes and comparing their different effects.
2. The effect of environmental conditions
3. The effect of light emitting sources such as sunlight
4. Effect of additives in the process
5. Executive costs that economic indicators in the removal of antibiotics from wastewater in him.

In Table 3, several studies by researchers to remove penicillin and its derivatives based on photochemical processes in the twenty-first century are mentioned. The scheme of the photodegradation mechanism of penicillin is also illustrated in Fig. 2.

## Sonochemical processes

Ultrasound energy is an especially significant factor in penicillin decomposition. This is due to the fact that this power correlates with the amount of ORP and the level of cavitation bubbles present at the electrode interface (Kamani et al. 2018, 2023c; Norabadi et al. 2022). Its range is between 20 and 120 w. In this method, ultrasound waves from the energy generator at high voltages (between 20 and 1000 kHz) create acoustic cavities and result in the emission of reactive OH· and H· (hydrogen radicals) which is the Outcome of the thermal dissociation of water-oxygen radicals (Klavarioti et al. 2009; Ahmadi et al. 2020; Kamani et al. 2023d). Some studies for the demotion of penicillin G and Amoxicillin using ultrasonically assisted AOPs are reported in Table 4.

### Ultrasound-based AOPs in penicillin removal

Ultrasound-based Advanced Oxidation Processes (AOPs) offer a promising approach for the removal of penicillin from wastewater. These processes typically involve the application of ultrasound in combination with powerful oxidants to generate reactive species, such as hydroxyl radicals, for the degradation of penicillin molecules.

#### Advantages

*Enhanced oxidant dispersion* Ultrasound promotes the effective dispersion of oxidants in the wastewater, leading to improved contact between the oxidants and penicillin molecules.

*Rapid degradation kinetics* Ultrasound enhances the mass transfer and collision frequency of oxidant molecules, accelerating the degradation kinetics of penicillin.

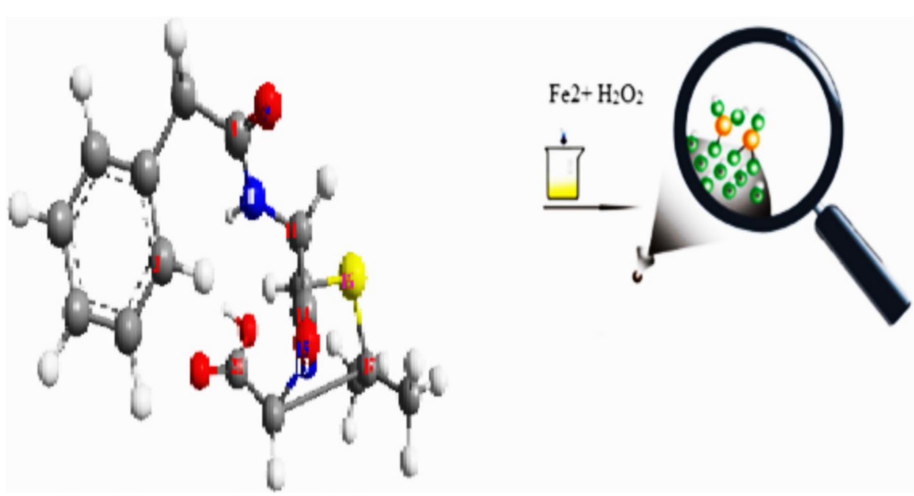



**Fig. 1** Graphical illustration of the Fenton-Photo procedure for the elimination of penicillin

**Table 3** Overview of research on the elimination of penicillin with photochemical processes and their conditions and efficiency

| Antibiotic name | Type of AOP | Concentration (mg/L) | Operation condition | Removal efficiency | References |
|---|---|---|---|---|---|
| PEN G | $Fe^{+3}-TiO_2$/UV-A | 10–45 | Time = 90 min<br>$Fe^{+3}$– TiO2 = 60 mg/L<br>pH = 3 | 84% | Dehghani et al. (2014) |
| PEN G | Photodegradation using FeNi3@SiO2@ZnO | 10 | $H_2O_2$ concentration = 150 mg/L<br>pH = 5<br>nanoparticle concentration from 0.005–1 g/L | 100% | Kamranifar et al. (2021) |
| Amoxicillin, ampicillin, and cloxacilli | PhotoFenton | AMX, AMP, CLX: 104, 105, – 1.103 | UV lamp, 230 V, 0.17 A, 6 W, 365 nm | AMX = 80<br>AMP = 85<br>CLX = 97 | Elmolla and Chaudhuri (2009) |
| Sulfamethoxazole | $UV/TiO_2$ | 100 | T = 25 °C, xenon lamp (1000 W),<br>wavelength = below 290 nm | 95 | Abellán et al. (2007) |
| PEN G | UV/leaves shaped-BiVO4/O3 | 150–50 | $O_3$ = 90–45<br>Flow rate = 100<br>light intensity = 400 mW/cm2 | 71.4 | Liu et al. (2019) |
| PEN G | UV/CoFe2O@CuS4 | 10–100mg/L | CoFe2O@CuS4 = 0.1–0.8g/L;<br>time = 10–120 min;<br>radiation intensity = 294–282W/m<br>pH = 3–11 | 70.7 | Kamranifar et al. (2019) |
| AMX | UV/H2O2/PMFeOx | 20–60 | Time = 0–60 min;<br>[PMFeOx] = 0–2.5g;<br>Temperature = 30–50 °C;<br>UV = 200–500 nm | 99.65 | Ayodele et al. (2012) |
| PEN G | $US/UV/WO_3$ | 150–50 | $WO_3$ = 100–200<br>Time = 60–120 min; | 91.3 | Almasi et al. (2016) |
| PEN G | $Fe^{3+}$/ $H_2O_2$/UV | 10–100 | [H2O2] = 5 and 40mM;<br>Treatment time = 5–30 min; irradiation = 125w | 56 | Arslan-Alaton and Gurses (2004) |
| AMX | Photocatalytic ozonation (O3 + UV/Vis + TiO2) | 90 | $O_3$ = 50–70<br>time = 0–80 min | 90 | Moreira et al. (2015) |

*Minimal by-products* Ultrasound-based AOPs can result in the generation of fewer harmful by-products, contributing to a more environmentally friendly treatment process.

## Disadvantages

*Energy consumption* Ultrasound-based AOPs may require significant energy input, especially for the generation of cavitation, which can impact operational costs.

*Equipment complexity* Implementing ultrasound-based AOPs may require specialized equipment and operational expertise, potentially increasing the complexity of the treatment system.

## Challenges

*Optimization of parameters* Achieving the optimal combination of ultrasound intensity, frequency, and oxidant dosage for efficient penicillin degradation is essential but can be challenging.

*Scale-up considerations* Scaling up ultrasound-based AOPs for industrial applications while maintaining efficiency and cost-effectiveness poses challenges that need to be addressed.

*Wastewater matrix interference* The presence of other organic and inorganic compounds in wastewater can interfere with the oxidation of penicillin, necessitating careful process design.

Ultrasound-based AOPs offer advantages such as enhanced oxidant dispersion, rapid degradation kinetics,

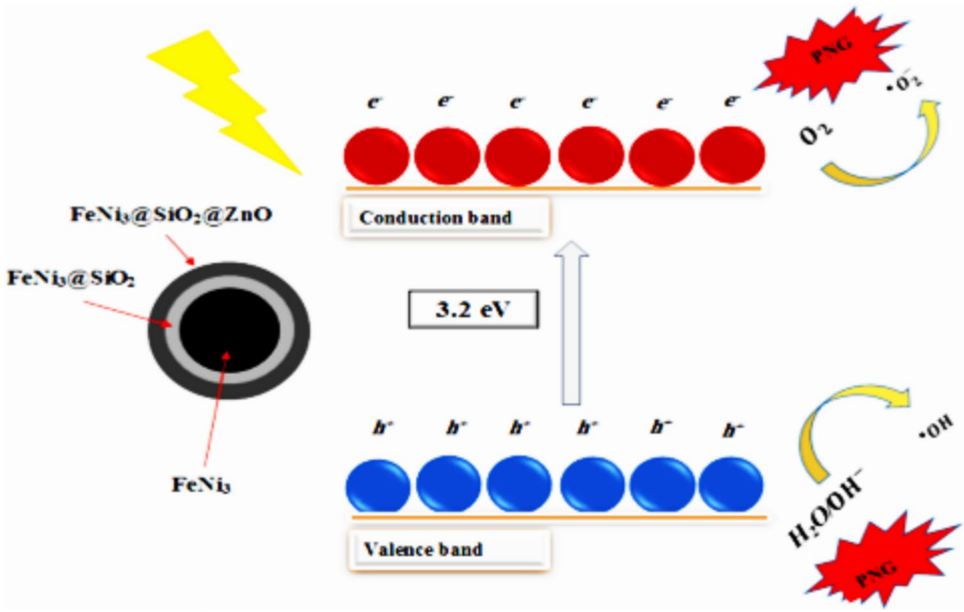



**Fig. 2** Schematic illustration of the photodegradation pathway of penicillin over FeNi3@SiO2@ZnO semiconductor nanoparticles (Kamranifar et al. 2021)

**Table 4** Review of studies on the elimination of penicillin with ultrasonic processes and their conditions and efficiency

| Antibiotic | Type of AOP | Concentration (mg/L) | Operation condition | Removal efficiency | References |
|---|---|---|---|---|---|
| PEN G | US –$Fe_2O_3$NPs | 10–100 | NPs = 0.05–0.3<br>pH = 3–11<br>Time = 10–90<br>ultrasonic light = 35–130 | 92.5% | Abdipour and Hemati (2023) |
| PEN G | US –MgO-NPs | 50–200 | Time = 20–150<br>pH = 3–11<br>Input power = 500<br>NPs = 0.3–2.3 | 81.14% | Rahdar et al. (2018) |
| PEN G | US/UV/$WO_3$ | 50–150 | $WO_3$ = 100–200<br>Time = 60 – 120min | 91.3 | Almasi et al. (2016) |
| AMX | US-$H_2O_2$/ZnO | 150–250 | [H2O2] = 0.1–5 Mol/L<br>Time = 10 – 100min<br>ZnO = 0.01–0.08g/L | 92.47 | Rahdar and Ahmadi (2019) |
| AMX | PS/US | 40–200 | US power = 40 – 400W; Sonication time = 60min<br>PS = 20 – 200mmol/L | 98 | Eslami et al. (2016) |
| AMX | US/O3 | 25–100 | pH = 4–10; frequencies = 575, 861 and 1141kHz; US power = 75W; diffused power = 14.6W/L; | 45 | Kıdak and Doğan (2018) |

and minimal generation of harmful by-products in the removal of penicillin from wastewater. However, challenges related to energy consumption, equipment complexity, process optimization, scale-up considerations, and wastewater matrix interference need to be carefully addressed to ensure the practical and efficient implementation of ultrasound-based AOPs for penicillin removal.

## Integrated processes

The synergistic effect of cavitation with oxidants, particularly in the context of hybrid Advanced Oxidation Processes (AOPs) based on the hydrodynamic cavitation phenomenon, can significantly enhance the removal of penicillin from wastewater. The combination of cavitation and oxidants offers several synergistic effects that improve the efficiency of penicillin degradation compared to conventional AOPs. Some of these effects include:

## Enhanced oxidant dispersion

Cavitation promotes the effective dispersion of oxidants, such as hydrogen peroxide or persulfates, in the wastewater. The formation and collapse of cavitation bubbles create microstreaming and turbulence, facilitating the uniform distribution of oxidants throughout the solution.

## Increased reactive surface area

Cavitation-induced microjets and shock waves lead to the formation of numerous reactive surfaces within the wastewater. When combined with oxidants, these reactive surfaces promote the generation of highly reactive radicals, such as hydroxyl radicals, which are instrumental in the degradation of penicillin molecules.

## Accelerated degradation kinetics

The presence of cavitation enhances the mass transfer and collision frequency of oxidant molecules, accelerating the degradation kinetics of penicillin. This acceleration is attributed to the intensified generation of hydroxyl radicals and other reactive species, leading to more rapid and efficient penicillin removal.

## Reduction of activation energy

Cavitation reduces the activation energy required for the oxidation reactions, thereby promoting the efficient utilization of oxidants in the degradation of penicillin. This reduction in activation energy enhances the overall efficiency of the AOPs, resulting in improved penicillin removal rates (Fedorov et al. 2022).

## Minimization of bypass pathways

The synergistic effect of cavitation with oxidants helps minimize bypass pathways and promotes the penetration of oxidants into hard-to-reach areas within the wastewater, ensuring more comprehensive and thorough degradation of penicillin molecules.

In summary, the synergistic effects of hybrid AOPs based on hydrodynamic cavitation and oxidants offer a promising approach for enhancing the removal of penicillin from wastewater. By leveraging the combined benefits of cavitation and oxidants, these processes can achieve improved oxidant dispersion, increased reactive surface area, accelerated degradation kinetics, reduced activation energy, and minimized bypass pathways, ultimately leading to more efficient and thorough penicillin removal.

## Process of SR-AOPs for penicillin elimination

Synergistic remediation-advanced oxidation processes (SR-AOPs) for eliminating penicillin from wastewater involve the integration of remediation techniques with advanced oxidation methods. The process typically includes the following steps:

1. *Generation of reactive species* Highly reactive species like hydroxyl radicals (·OH) or sulfate radicals ($SO4\cdot-$) are produced by activating powerful oxidants such as hydrogen peroxide or persulfates. This activation can be achieved through methods like hydrodynamic cavitation, ultrasound, or ozone.
2. *Reaction with penicillin* The generated reactive species initiate oxidation reactions with penicillin molecules in the wastewater, breaking them down into less harmful by-products.
3. *Complete degradation* The oxidation reactions continue until the penicillin molecules are completely degraded, effectively eliminating their detrimental effects on the environment.

### Challenges and advantages

**Challenges** *Optimization of conditions* Achieving the optimal combination of parameters, including oxidant dosage, cavitation intensity, and reaction time, can be challenging.

*Energy consumption* Some SR-AOPs may require significant energy input, especially in the case of ultrasound or ozone activation.

*Wastewater matrix interference* The presence of other organic and inorganic compounds in wastewater can interfere with the oxidation of penicillin, requiring careful process design.

**Advantages** *Enhanced degradation efficiency* SR-AOPs offer improved degradation efficiency compared to conventional AOPs, leading to more effective penicillin elimination.

*Synergistic effects* The combined action of remediation techniques and advanced oxidation methods results in synergistic effects that enhance the overall degradation process.

*Broad applicability* SR-AOPs can be applied to a wide range of wastewater matrices and are effective in the removal of various organic pollutants beyond penicillin (Honarmandrad et al. 2023).

**Table 5** Summary of kinetics study various research on the elimination of penicillin

| Type of AOP | Degree | References |
|---|---|---|
| US –$Fe_2O_3$NPs | First order | Abdipour and Hemati (2023) |
| UV/ZnO | Pseudo-first-order | Chavoshan et al. (2020) |
| UV-A/ZnO/PPS | First order | Berkani et al. (2022) |
| UV/peroxydisulfate | Pseudo-first-order | Zhou et al. (2018) |

### Mechanism

The mechanism of SR-AOPs involves the formation and utilization of highly reactive species to oxidize and degrade penicillin molecules in the wastewater. This mechanism is characterized by the synergistic combination of remediation techniques, such as cavitation or ultrasound, with advanced oxidation processes, leading to the enhanced generation and utilization of potent oxidizing agents for the efficient degradation of penicillin.

SR-AOPs present a promising approach for the elimination of penicillin from wastewater, offering enhanced degradation efficiency, synergistic effects, and broad applicability. However, challenges related to process optimization, energy consumption, and wastewater matrix interference need to be carefully addressed to ensure the practical implementation of SR-AOPs for penicillin removal.

## Kinetics study

The kinetic modeling study was conducted to assess the extent of the control Elimination step of penicillin antibiotics from the solution. The pseudo-first order, pseudo-second order, and intraparticle diffusion equations were examined, which are utilized to explain the kinetic rate given in Table 5 of some studies.

## Conclusion and outlook

Due to the spread of infectious diseases, penicillin derivatives are one of the most widely used antibiotics in the world, and their release in aquatic environments has endangered human health and the environment. The limitations of different water and wastewater treatment methods including high cost, phase transfer, more sludge production and non-compliance with established standards have turned researchers' attention to advanced oxidation-based methods. Various factors such as penicillin concentration, temperature, pH, wastewater quality and type of treatment affect the quality of wastewater treatment in the face of this antibiotic. This review discusses a variety of advanced oxidation processes that are effective in penicillin degradation. The combination of ozone, hydrogen peroxide and UV can enhance the removal efficiency, and the photo-Fenton process can be economical.

The compound used by Fenton is readily available to researchers and minimizes the use of UV light. Regarding the use of hydrogen peroxide, it should be noted that it increases the production of hydroxyl radicals, but a high dose of the catalyst reduces its efficiency. The method of using UV-TiO2 despite the high efficiency percentage is difficult due to the challenge of recycling titanium dioxide and the high cost of this photocatalyst. It seems that the use of ultrasound in the elimination of antibiotics has not yet been institutionalized and received less attention. Since the removal of antibiotics by the sono-Fenton process has not been investigated, while the removal of organic contaminants has been performed with high efficiency by this method, sono-Fenton may be promising for the removal of penicillin.

Considering the optimal efficiency of using ultrasound waves and nanoparticles in the removal of penicillin and the lack of research on the removal of penicillin with ultrasound and persulfate complex, it is suggested that researchers use this approach to research to increase the efficiency of penicillin removal.

**Acknowledgements** The authors would like to acknowledge the financial and technical cooperation of Zahedan University of Medical Sciences for this study with ethical code: IR.ZAUMS.REC.1403.080

**Author contributions** All persons who meet authorship criteria are listed below. Hossein Abdipour: Drafting the manuscript, revising the manuscript and improve English; Tayebe zomorrodi jangaee, Shima goodarzi, Gholamreza Parsaseresht and Masoomeh Torabideh: acquisition of data, conception and design of study; Hossein kamani: Supervision.

## Declarations

**Conflict of interest** The authors confirm that they don't have any conflict of interest.

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