Advanced Oxidation Processes: An Effective Solution for Treating Emerging Pollutants from Pharmaceutical Industries.

Mamilla Vijaya Kumar

Assistant Professor, Department of Civil Engineering, KSRM College of Engineering, Kadapa, Andhra Pradesh, India

Abstract. The rapid increase in population poses a grave threat to environmental sustainability and water availability. India's pharma industry holds the 3rd position globally in pharmaceutical production by volume. However, the industry's wastewater contains harmful substances such as high concentrations of organic matter, microbial toxicity, salts, and non-biodegradable compounds, posing a significant danger to human health and the ecosystem. Regularly assessing wastewater discharge against regulatory standards is vital to ensure water safety. The presence of emerging contaminants in natural and drinking water directly jeopardises water quality and the survival of aquatic life. Advanced treatment methods are imperative, as trace amounts of solids and organic matter can persist even after secondary treatment. Treating pharmaceutical wastewater is particularly challenging due to the wide variety of products produced in the industry. The Advanced Oxidation Process is a highly effective technology for removing pollutants from wastewater. It is crucial to implement safe and effective treatment methods to mitigate the impact of pharmaceutical wastewater. This paper critically evaluates AOPs and their ability to treat pharmaceutical wastewater, providing valuable insights into pharmaceutical industry wastewater, the significance and characteristics of pharmaceutical wastewater, their impact on human health and the ecosystem, various treatment methods, and different AOPs employed for treating pharmaceutical wastewater in detail.

1. Introduction

Environmental pollution is a major challenge for modern civilization [1,2]. Industrial wastewater disposal, MSW, and hazardous waste constitute 33% of water pollution in India [3-6]. The toxic substances in wastewater pose a significant threat to the ecosystem's life forms and harm rivers and human health [2]. The impacts of industrialization intertwine pollution, public health, and the environment, highlighting the urgent need for action. Rapid industrialization causes 3.4 million waterborne disease deaths annually [7,8]. The extent of pollution varies based on industry size and processes [9]. Industrial wastewater is a mixture of chemicals and additives used in manufacturing finished goods. It can contain pollutants, colour, odour, suspended solids, etc. [10]. Controlling toxic waste disposal into water sources is crucial to minimize environmental pollution and ensure a healthy environment for future generations.

The Pharma industry is highly advanced, requiring significant investments and operating with high efficiency, making it a promising international industry [11]. India's pharmaceutical industry is a global leader, supplying low-cost vaccines and generic medicines. It contributes 8% to the global API Industry, producing 60% of the world's vaccine supply. Indian medicines are preferred worldwide for their affordability and high quality, earning India the title of "pharmacy of the world [12]. Over the past century, medicine has made great strides in extending human life and reducing illness. Over 3000 chemical compounds have been approved as ingredients in pharmaceutical products [13].

The pharmaceutical industry produces wastewater containing a range of toxic organic chemicals, posing significant risks to human health, the ecosystem, and microbial communities [14,15]. Pharmaceutical residues in water environments are ecologically toxic and can lead to the spread of antimicrobial resistance among bacteria and fungi [16,17]. Antibiotics, antifungals, and personal care products accelerate this process, particularly in hospitals and WWTPs [17-21]. The development of AMR undermines the efficacy of antimicrobials, prolongs morbidity, and increases mortality. It's crucial to address the spread of AMR in WWTPs to protect public health [22]. The pharmaceutical industry generates wastewater containing active pharmaceutical ingredients (API), organic, and solid content. Effective wastewater treatment is critical to reduce environmental impact and ensure safe disposal [23,24]. However, treating pharmaceutical

*Corresponding author : civilvijay1@gmail.com

wastewater is challenging due to different drug production types [25]. Disposing of such effluent without proper treatment can harm the environment and human health [26]. Despite various treatment methods employed, the challenge remains unresolved in many developing countries. It is essential to take necessary measures and ensure effective wastewater treatment processes reduce the industry's environmental impact, protect individual health, and safeguard community water supply [27]. Pharmaceutical contaminants, referred to as Emerging Contaminants (EC), pose a threat to public health, biota, and the ecosystem. However, pharmaceutical contaminants are not effectively removed during conventional wastewater treatment processes [28]. Rachel Carson's "Silent Spring" book revealed the dangers of pesticides, particularly DDT, on bird populations. It raised awareness about emerging pollutants and synthetic or naturally occurring chemicals that are not regularly monitored in the environment [29]. These pollutants pose a significant risk to ecological systems and human health as they can pass through wastewater treatment plants undetected [30].

2. Sources of pharmaceutical wastewater

The detection of pharmaceutical residues in watersheds dates back to 1976 in Kansas City, US, with Clofibric acids found in the range of 0.8 to 2.2 µg/L. In 1981, pharmaceutical residue was also found in river waters in the United Kingdom [31,32]. Pharmaceuticals and other xenobiotics in the aquatic environment can undergo three main fates: (a) mineralization to CO_2 and H_2O, (b) resistance to degradation due to lipophilicity and retention in sedimentation sludge, and (c) transformation into more hydrophilic molecules, passing through wastewater treatment and entering surface waters like rivers. These compounds are highly persistent in the environment [33]. Some pharmaceuticals, like antibiotics, can induce lasting genetic changes in microorganisms, resulting in resistance even at low quantities [34].

To reduce pharmaceutical residue in water, reliable data on contributing sources is crucial [35]. Sources can be natural or human-made [36]. According to the OECD, pharmaceutical residue comes from various sources including pharmaceutical industries, wastewater treatment plants (WWTPs), agriculture, aquaculture, septic tanks, and waste management services. The OECD categorizes released pathways into point sources like WWTP discharge and diffuse sources like agricultural runoff [36]. Pharmaceuticals in catchment surface water come from hospitals, livestock, and fish farms, houses, industrial drug waste, and STPs [37]. Human and veterinary body secretions release pharmaceutical compounds into watersheds [36].

Animal pharmaceuticals enter the environment through sources like animal husbandry, horticulture, sewage effluent, waste disposal, and aquaculture. Veterinary antibiotics are essential for treating animal infections and improving their health. However, the use of animal feces as fertilizer has raised concerns about the environmental impact of antibiotic residues [38]. Manure used as fertilizer can contaminate soil and water through runoff or leaching [39]. Global pharmaceutical consumption has surpassed 30 million tons in 2023 due to population growth and economic development [40]. A significant portion of pharmaceuticals consumed by patients is not metabolized and ends up in the sewage system. These compounds ultimately enter water systems through wastewater treatment plants [41]. Human antibiotics also enter the environment through wastewater, reaching surface waters and groundwater [42]. Similarly, veterinary antibiotics from aquaculture, improper drug disposal, and manufacturing effluents are significant sources of contamination [43].

Pharmaceutical residues in the environment decrease from wastewater to freshwater due to natural breakdowns influenced by processes like biotransformation, photolysis, sorption, volatilization, and dispersion [44]. The fate of these residues is determined by their natural breakdown and their physio-chemical properties, such as solubility [45]. Pharmaceutical pollutants are consistently released into our water as compounds or decomposition products. Various sources contribute to this pollution [46], as depicted in Fig. 1

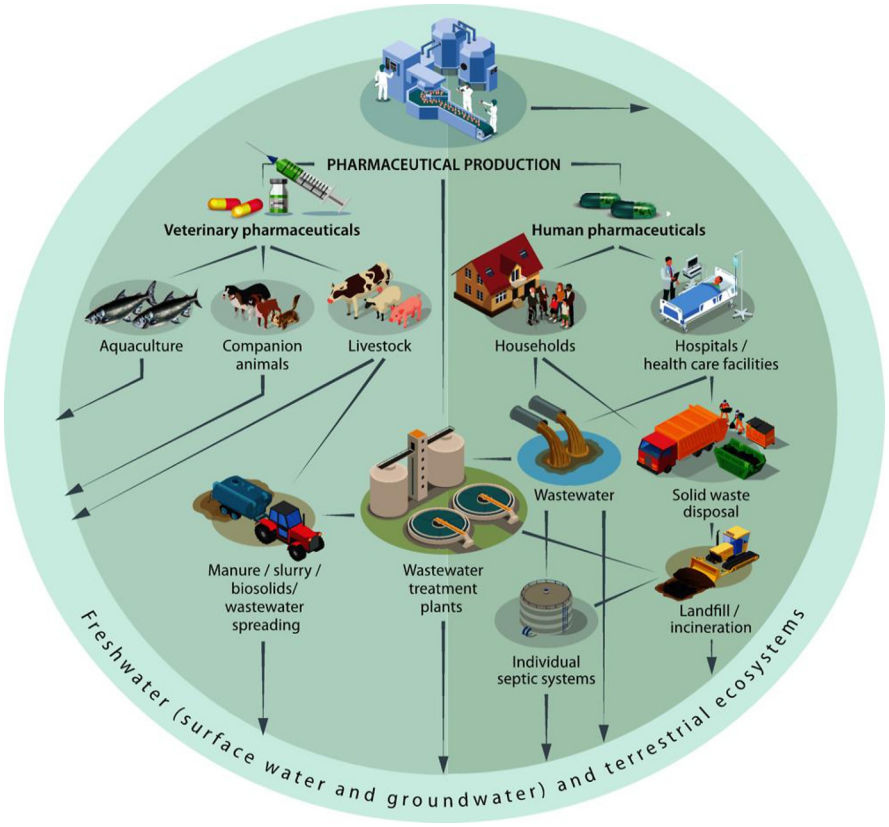

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Fig. 1: Antibiotics pathway into the ecosystem [47]

3. Classification & characteristics of pharmaceutical effluents

Effluents from the pharmaceutical industry contain harmful chemicals that can be carcinogenic, mutagenic, teratogenic, and toxic to aquatic life [48].

To maintain safety and quality standards, the components and their nature must be categorized before treatment [49-51]. A proper study of effluents' physicochemical properties is required before selecting the appropriate treatment process. Types of possible pollutants in pharmaceutical wastewaters are shown in Fig.2.

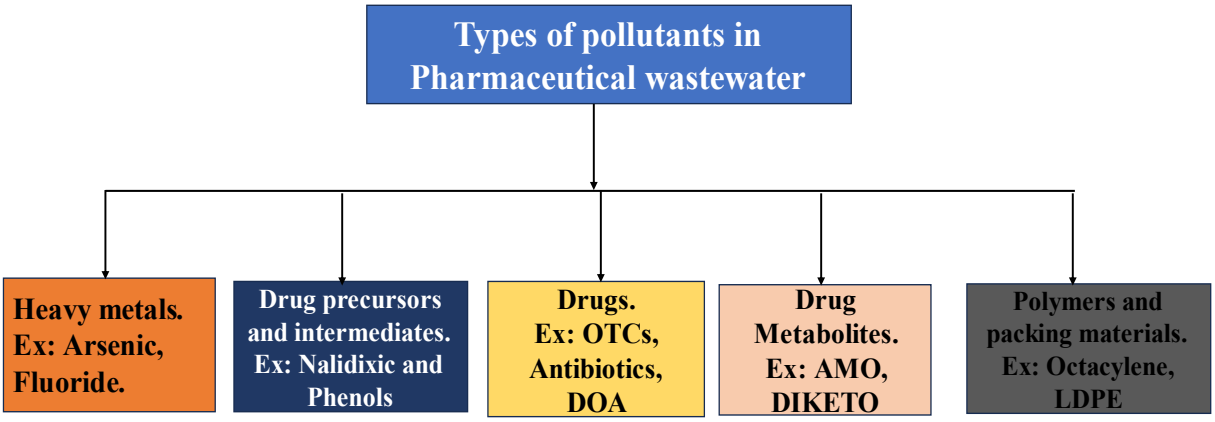

Fig. 2: Classification of various types of pollutants in pharmaceutical wastewater [52]

Table 1 : Characterization of pharma effluents

Parameters	References						
	[27]	[53]	[54]	[55]	[56]	[57]	[58]
pH	6.81	7.5	5.4–5.6	1-6	3.9-9.2	6.9	3.7–8.5
TSS (mg/lit)	577	---	--	200-500	--	680	48–1113
TDS (mg/lit)	1084	---	695–1170	--	675-9320	1760	600–1770
TS (mg/lit)	---	---	--	--	--	2440	880–4934
BOD_5 (mg/lit)	2480	260	5460–9000	500-2500	200−6000	140	20–1800
COD (mg/lit)	6400	2600	11,980–15,840	1000-10000	375−32500	540	128–3500
$\frac{BOD_5}{COD}$	--	---	--	--	0.1−0.6	0.259	0.15–0.51
TOC (mg/l)	--	---	--	--	860-4940	--	--
TKN (mg/l)	--	---	--	500-1500	165-770	--	80–164
NH_3−N, mg/l	--	25	---	--	148-636	--	--
NH_4^+ - N	--	---	---	--	--	--	74–116
Cl^-, mg/l	--	1250	--	--	760−4200	--	205–261
SO_4^{2-}, mg/L	--	11000	--	5000-25000	890−1500	---	---
EC, µS/cm	--	---	1230–1830	--	--	--	157–1673
Alkalinity	--	2500	--	--	--	--	90–564
Total phosphate	--	---	--	--	--	--	18–47
Turbidity, NTU	--	---	--		--	--	2.2–138
Temperature (°C)	--	---	--	25-80	--	--	32–46

Characterizing wastewaters is crucial for determining the water's quality. The CPCB sets standards for environnemental pollutant discharge from the pharmaceutical industry, summarized in Table 2.

Table 2: Effluent standards of the pharmaceutical industry [59].

S. No	**Effluent parameter**	**Standard**
Compulsory parameters		
1	p H	6.0-8.5
2	BOD (5-Days @ 27°C)	30
3	COD	250
4	TSS	100
5	Oil & Grease	10
6	Ammonical Nitrogen	100
7	Bio - Assay Test	90% Survival of Fish after first 96 hours in 100% effluent
Additional Parameters		
8	PO_4^{3-} as P	5
9	Sulphides as S	2
10	C_6H_5O Compounds	1

11	Zn	5
12	Cu	2
13	Cr	3
14	Hexavalent Chromium (Cr^{6+})	0.1
15	Cyanide (as HCN)	0.1
16	As	0.2
17	Hg	0.01
18	Pb	0.1

4. Advanced treatment methods for pharmaceutical effluents

India's MoEFCC has classified pharmaceutical manufacturers as "red category" due to toxic discharge. Traditional wastewater treatment is ineffective in removing pharmaceutical discharges [60-62], reducing overall efficiency [63]. The pharmaceutical industry produces diverse wastewater, which contains high levels of pollutants [64,65], posing a risk to flora and fauna. Hence, treating wastewater is crucial to reduce toxic substances before discharge into natural systems [66]. Designing an appropriate treatment system requires analyzing wastewater characteristics. Treating wastewater is essential to prevent environmental damage and promote sustainable growth of the pharma industry [67,68].

The pharma industry uses advanced methods to treat and dispose of wastewater, which varies based on raw materials and manufacturing processes [69].Various treatment methods include physicochemical treatments such as screening, chlorination, coagulation, sedimentation, flocculation, adsorption with AC, ASP, and membrane separation processes like microfiltration, ultrafiltration, reverse osmosis, and electrodialysis [69], additionally, biological treatments such as aerobic and anaerobic processes, as well as advanced oxidation processes, are available [70]. These methods are employed to effectively reduce the organic load and eliminate toxic pharmaceutical compounds [68]. The use of AOP has shown remarkable performance in degrading pharmaceuticals in aqueous solutions, making them an attractive option for wastewater treatment. These innovative methods are crucial in tackling the problems caused by pharmaceutical effluent. wastewater and are essential for environmental protection and public health [71].

Various treatment processes employed for pharmaceutical wastewater with their contaminate removal efficiencies are detailed in Table 3.

AOPs for pharmaceutical wastewater treatment involve various processes, as shown in Fig. 3

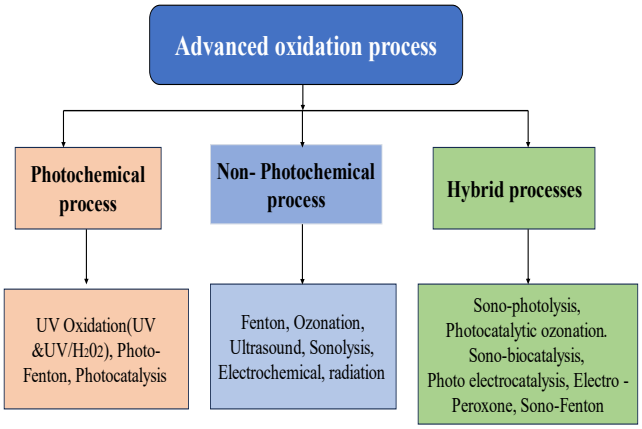

Fig.3: Different types of AOPs [82].

Table 3: summary of the AOPs and their relative treatment efficacy w.r.t different pharmaceutical pollutants

Treatment method	**Applications**	**Efficiency**	**References**
Anaerobic Packed Bed Reactor	Pharmaceutical wastewater	COD removal efficiency is up to 18%.	[27]
Multi-effect evaporator (MEE) process	Pharmaceutical wastewater	Removal efficiencies are 85% for TDS and TSS, 93% for BOD, and 81% for COD.	[72]
wet Peroxide oxidation	Pharmaceutical wastewater	The highest TOC removal was 57.96%.	[73]
Ozone-based AOP& adsorption by activated char	anti-cancer drugs, antipsychotic drugs, & painkillers	The AOP & adsorption process has COD removal efficiencies of 75−88.5% & 85.4−92.7% respectively.	[53]
Fenton's oxidation	Pharmaceutical industry	62% of Total Organic Carbon.	[10]
cyanobacterium Phormidium fragile	Pharmaceutical wastewater	NH_3, P_4, COD, and BOD removal efficiencies are 58.89%, 62.23%, 54.55%, and 59.85% respectively.	[74]
ASP + UV-free surface reactor process	Pharmaceutical wastewater	The treatment achieved impressive removal rates: 98.57% for COD, 98.99% for BOD, and 98.31% for TSS.	[75]
Rotating Biological Contactor	Antibiotic pharmaceutical wastewater	45% COD, NH^+_4-N, and BOD_5 removal efficiencies. are 45, 40%, and 85% respectively.	[76]
Coagulation and Fenton-like AOP	wastewater containing Azithromycin	COD removal up to 96.89%	[77]
UASB	Cosmetic wastewater treatment	COD is 95% and TSS is 85% removed.	[78]
ASP followed by Fenton's oxidation process	herbal pharmaceutical industrial wastewater	COD, BOD, SS, and TOC removal efficiencies are 85.19%, 88.10%, 70.00%, and 78.22% respectively.	[60]
solar photo-Fenton + biological treatment	Pharmaceutical wastewater	DOC degradation efficiency was over 95%.	[79]
Coagulation, flocculation, sedimentation, & sand filtration followed by AC adsorption	Pharmaceutical wastewater	AC adsorption delivers remarkable reductions: 99.9% for phenol, 99.1% for TDS, 21.4% for TSS, 81.3% for BOD, and 71.1% for COD.	[80]
Sequencing batch biofilter.	pharmaceutical wastewater effluent	COD removal efficiency up to 95–97%.	[81]

The most commonly used advanced oxidation process is explained as:

Pharmaceuticals pose a significant challenge in wastewater treatment due to their resistance and toxicity. AOPs offer an eco-friendly and innovative approach to enhancing wastewater treatment, generating free radicals that effectively oxidize pharmaceuticals in aqueous solutions and on catalyst surfaces [83]. AOPs use reactive substances like ozone, hydrogen peroxide, and hydroxyl radicals to break down pollutants at ambient conditions [84]. The hydroxyl radical is especially effective due to its non-selective nature and high reactivity. When it reacts with organic compounds, it can produce carbon dioxide, water, and inorganic acids [85]. Different AOPs will be discussed in more detail later.

4.1 Ozonation:

Ozone is highly effective in breaking down pharmaceuticals [86]. Ozone is used for odour and taste control in drinking water treatment and as a disinfectant also. Studies have been conducted in drinking water plants and WWTPs, with a focus on treating pharmaceuticals in subsequent stages [87-90]. Ozonation breaks down target molecules in wastewater and produces easily biodegradable byproducts [91]. It can significantly improve biodegradability, especially in wastewater containing antibiotics [92]. Ozonation, used as a pre-treatment process, significantly improves the biodegradability of pharmaceutical wastewater, especially those containing antibiotics. It can be used alone or in combination with biological treatment to enhance the breakdown of pharmaceuticals in wastewater [93].

To improve the degradation of pharmaceutical pollutants in water, utilize ozone-based processes

with H_2O_2 or UV light to enhance hydroxyl radical production [94]. Factors like pH, ozone dosage, and temperature play a crucial role in the conversion and mineralization of pharmaceuticals during these treatments. However, it's important to note that the short lifespan of ozone makes it a relatively expensive method, and its high energy demand may be a limitation for real-world applications [95]. Ozonation effectively removes APIs like carbamazepine, sulfamethoxazole, and trimethoprim, but leads to low mineralization rates due to persistent byproducts. Low ozone doses can remove target APIs but often result in incomplete mineralization rates [96- 98]. Studies show that the effectiveness of ozonation on pharmaceuticals varies based on their chemical structure [99].

A study found that catalytic ozonation with Mn-anchored zeolite removed 97% of cephalexin in 2 minutes, compared to non-catalytic ozonation, which showed a 79.2-fold increase in cephalexin degradation with the catalyst [100]. Ozone-based water treatment has drawbacks, including low solubility in water, more energy consumption, and harmful by-product production [101]. However, using a catalyst in a process called catalytic ozonation can address these issues. Numerous catalysts, including metals and carbon materials, play a crucial role in enabling the full breakdown of organic compounds while minimizing the generation of harmful byproducts [102]. Incorporating catalysts can favour indirect oxidation over direct electrophilic ozone reactions, leading to the comprehensive removal of pollutants [71]. Ozonation effectively removes micropollutants, microbial count, odour, color, and foam. However, it may not fully mineralize target compounds, potentially leading to the production of more harmful substances [103]. Additional treatments like sand filtration are needed to break down reactive oxidation products. It's important to note that while ozonation provides significant benefits, it also entails higher energy costs [104]. The process of the ozonation AOP is shown in Fig. 4.

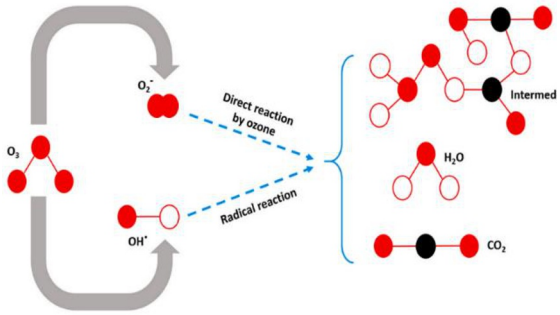

Fig. 4: Process of the ozonation AOP [105].

4.2 Photo- Fenton Advanced Oxidation Process

In the late 19th century, Henry John Horstman Fenton pioneered Fenton's reagent, a potent solution of H_2O_2 and ferrous ions with robust oxidizing properties [106]. This oxidation can take place in homogeneous or heterogeneous systems, with homogeneous oxidation being the most widely employed method [47]. The Fenton process stands out as a highly effective technique for addressing a broad spectrum of wastewater contaminants, encompassing C_6H_5O, CH_2O, BTEX, and intricate waste from pharmaceuticals [107].

The Fenton reaction uses iron salts (Fe^{2+}) and H_2O_2 to produce hydroxyl ($HO^{\cdot}$) radicals in mildly acidic conditions. The Fe^{2+} catalyst is regenerated through reactions with Fe^{3+} and other intermediates [108].

$$Fe^{2+} + H_2O_2 \longrightarrow Fe^{3+} + OH^- + HO^{\cdot} \quad \text{----} \quad (1)$$

$$Fe^{3+} + H_2O_2 \longrightarrow Fe^{2+} + H^+ + HO_2^{\cdot} \quad \text{------} \quad (2)$$

The schematic diagram of the Photo-Fenton AOP is shown in Fig.5.

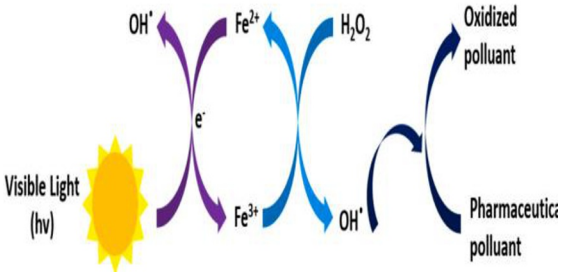

Fig.5: The schematic illustration of photo-Fenton [105].

The Fenton and photo-Fenton processes effectively degrade pharmaceuticals and can be enhanced by UV radiation [109,85]. Solar photo-Fenton effectively treats pharmaceuticals. Using a solar simulator, amoxicillin is completely removed within 5 minutes with the ferrioxalate complex or within 15 minutes with $FeSO_4$ and 120 mg/l H_2O_2 [110]. Increasing the temperature has a positive effect on the Fenton and photo-Fenton processes by boosting reaction rates. However, it can also accelerate hydrogen peroxide decomposition, reducing the available amount for the reaction and decreasing process efficiency when an excess of hydrogen peroxide is used.

$$2H_2O_2 \longrightarrow 2H_2O + O_2 \text{ ------ } (3)$$

Homogeneous oxidation efficiency is optimized at a solution pH of 2-4 and with the right COD: H_2O_2: Catalyst ratio. Harness the power of UV irradiation to maximize the efficiency of the photo-Fenton reaction, yielding an increased production of hydroxyl radicals [111-113]. Adjusting catalyst and oxidant concentrations based on the effluent's

pollution levels makes Fenton oxidation ideal for treating heavily contaminated hospital or pharmaceutical effluents, resulting in less toxic effluents [114 - 116,91]. Research has shown that Fenton oxidation can effectively reduce refractory effluents, making them more amenable to biological posttreatment [116,91]. Additionally, penicillin was eliminated after just 40 minutes of advanced Fenton/UV treatment [117]. The Fenton process has drawbacks including high operating costs, the risk of using reagents, and the accumulation of iron sludge in treated wastewater [118]. Despite the drawbacks of pH dependency and iron sludge generation, the Fenton process is an ideal pretreatment technology for converting non-biodegradable pharmaceutical effluent into a biodegradable form, enhancing the efficiency of subsequent biological treatment [79,138].

Fenton's process offers cost-effective and straightforward reactor arrangements, making it advantageous for construction and operation. The process can utilize both homogeneous and heterogeneous catalysts. However, the expensive nature of Hydrogen Peroxide and increasing water regulations pose challenges. Maintaining the required acidic conditions is crucial for stabilizing the radicals [119,120]. The efficiency of homogeneous Fenton processes is significant under optimal conditions, but limitations arise, such as the formation of Fe $(OH)_3$ sludge at pH values above 4 [121]. Heterogeneous catalysts can overcome some of these limitations and maintain effectiveness over a wider pH range. However, heterogeneous catalysis has a slower oxidation rate [122].

4.3 Photocatalysis

Photocatalysis uses light to drive chemical reactions with metal oxides or metal sulfide nanoparticles [123,124]. Photocatalysis is an eco-friendly method for removing organic waste from water using light-induced reactions with catalysts [125-127]. Heterogeneous photocatalysis is the process of using a catalyst to assist in photoreactions. It has been applied to purify air and water contaminated with organic and inorganic compounds since the 1970s when it was discovered by Fujishima and Honda [128]. Heterogeneous photocatalysis separates catalysts after the process, unlike homogeneous photocatalysis [129]. It efficiently destroys organic pollutants in wastewater and initiates oxidation reactions, breaking down contaminants. This method offers a sustainable solution for environmental purification [130]. Intensive research has been dedicated to finding photocatalysts with specific properties such as strong photoactivity, low bandgap, and potent catalytic activity [131]. Several metal oxides including TiO_2, ZnO, CeO_2, Mn_3O_4, $SrTiO_3$, $BiVO_4$, and Bi_2WO_6 nanoparticles, meet these criteria. Metal oxides like TiO_2 and ZnO_2 nanoparticles demonstrate excellent photoactivity in removing pharmaceuticals [132,133].

Heterogeneous semiconductor photocatalysis uses TiO_2 as the catalyst with UV radiation to accelerate photochemical transformations [134,135]. The process is cost-effective, non-toxic, and stable under ambient conditions [136]. Semiconductor photocatalysis shows promise for treating effluents with low organic matter loads but has not been practically applied due to its low energy consumption [47]. Rutile TiO_2 is the primary catalyst in pharmaceutical studies. It effectively treats high COD effluents and transforms refractory organic contaminants for biological treatment [33]. TiO_2, particularly Degussa P25 with 80% anatase and 20% rutile, is the primary catalyst for pharmaceutical photocatalytic treatment studies. Its exceptional activity is due to the unique crystallite morphology, enabling efficient charge separation. ZnO and CdS are also effective photocatalysts for water treatment [138]. ZnO is a strong alternative to TiO_2 due to its similar photodegradation mechanism and higher solar spectrum absorption, making it more suitable for photocatalytic degradation under sunlight [137]. It also shows greater catalytic activity than TiO_2 for degrading sulfamethazine and chloramphenicol [l33]. UV/ TiO_2 with H_2O2 enhances phenol and chemical oxygen demand removal from fermentation effluent [139]. Photocatalysis with ozonation improves chemical oxygen demand removal in penicillin formulation effluent [140].

Photocatalytic reactions depend on catalyst concentration, light intensity, solution pH, and H_2O_2 addition [33]. Renewable energy sources power both heterogeneous and homogeneous photocatalysis, with solar photocatalysis gaining attention for pharmaceutical treatment [141–143]. Life cycle analysis shows a lower environmental impact for homogeneous photocatalysis compared to the heterogeneous process [142]. The process of photocatalysis is shown in Fig.6

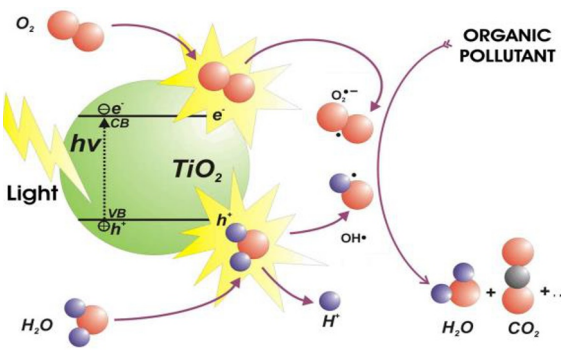

Fig.6: Schematic illustration of Photocatalysis [144].

The combination of antibiotics with UV achieves a 98% removal rate, while carbamazepine removal rates are under 10% [136,145]. TiO_2 is affordable and recyclable in industrial applications, and solar studies have shown effectiveness in treating pharmaceuticals without the need for expensive UV light. The implementation of this technology in commercial settings faces challenges due to various operating parameters [135]. Photocatalysts must overcome limitations to increase effectiveness, including the need for energy to surpass bandgap energy for electron excitation, limited light penetration, and rising costs [146,147]. Recombination rate and charge carrier transfer can also limit efficiency [148]. Photocatalytic materials can have a significant environmental impact due to the potentially harmful transformation products they produce. Pharmaceuticals containing aromatic rings can also produce harmful compounds if not properly degraded [149]. Additionally, the instability of photocatalysts in water raises concerns about the potentially toxic effects of the ions released during their dissolution. Understanding these degradation mechanisms and improving the stability of photocatalysts is crucial to mitigate their adverse environmental impacts [150].

The study developed a low-cost Fe−TiO_2 composite to remove Escherichia coli from water. The composite showed better performance than traditional methods and achieved a 93% reduction of E. coli under optimized conditions. It also demonstrated high stability under continuous flow, making it a promising option for wastewater treatment [151]. A study found that waste-derived Fe-TiO_2 composite effectively inactivated E. coli in water within 45 minutes. The process showed 23% synergy over single processes and displayed exceptional durability and recyclability even after 100 cycles. This suggests its potential for wastewater treatment [152]. A recent study used a pilot-scale reactor with a durable iron-mediated TiO_2 catalyst made from waste materials. It achieved impressive reductions in bacteria and organic matter in just 45 minutes, showing promise for water disinfection applications [153]. A recent study introduces a novel concept of a fixed-bed in-situ once-through reactor designed for the disinfection of E. coli. The reactor employs a low-cost composite material made of Foundry Sand (FS) and fly ash (FA) to achieve hybrid effects. In a cascade reactor with three reactors in series, the reactor approached PFR with a retention time of 45 minutes and achieved 100% inactivation of E. coli cell count [154].

A study investigated the use of Fe-TiO_2 composite beads for inactivating E. coli using photocatalysis and the photo-Fenton process. The beads showed effective E. coli reduction and durability for over 50 recycles. Analysis confirmed the presence of TiO_2 and the activity of the catalyst [155]. A low-cost iron-titanium oxide composite efficiently treats municipal wastewater with 100% bacteria inactivation and substantial reductions in organic matter within 60 minutes. The composite remains stable after 35 recycling cycles, demonstrating its long-lasting durability and potential for widespread use [156].

4.4 Ultrasound Irradiation/Ultrasonic Irradiation /Sonolysis

Ultrasound irradiation/Ultrasonic Irradiation, also known as sonolysis is an innovative and sustainable advanced oxidation process (AOP) that has shown potential for degrading pharmaceuticals in wastewater [157]. Sonochemistry harnesses the power of micro-bubbles that form, grow, and then violently collapse through acoustic cavitation induced by ultrasonic waves. This process generates unique conditions of pressure (~1000 atm) and temperature (~5000 K), leading to the production of hydroxyl radicals through the dissociation of water molecules and oxygen [158].

$$H_2O +))) \longrightarrow HO + H^{\cdot} \quad \text{------ (4)}$$
$$2HO^{\cdot} \longrightarrow H_2O_2 \quad \text{------ (5)}$$
$$2HO^{\cdot} \longrightarrow H_2O + O \quad \text{------ (6)}$$

where))) refers to the ultrasound irradiation.

Sonochemical reactions occur in three potential sites: the cavitation bubble, the interfacial region, and the solution bulk[153].This process is suitable for degrading pharmaceutical micro-pollutants with low solubility and high volatility. Ultrasound frequency and intensity, reactor geometry, contaminant type, bulk temperature, and water matrix affect ultrasound irradiation process [159]. Lower frequencies and higher power densities can enhance the breakdown of pharmaceutical compounds. Cavitational activity is also critical in sonochemical treatment efficiency, as ultrasonic power increases the degradation rate linearly by generating more HO radicals [160,161].

Sonolysis has been found effective in removing various chemical compounds from dilute solutions [162,163]. Sonolysis is effective but energy-intensive and limited in scale. Combining it with other AOPs, such as sono-photolysis can reduce costs and improve mineralization. Understanding pharmaceutical degradation mechanisms is crucial for AOP integration [82]. For instance, combining sonolysis and photolysis (UV/H_2O_2) in sono-photolysis has been shown to effectively treat pharmaceutical wastewater, resulting in a remarkable 91% removal of TOC. This is a significant improvement over the 3% and 8% removal rates achieved by single sonolysis and photolysis (UV) methods, respectively [164]. Sonolysis has the drawback of high energy consumption and low electrical efficiency [71]. In a

recent study, magnetic Fe_3O_4–bentonite nanocomposite (Fe_3O_4-Bt) with a 17-minute ultrasonication process can effectively remove 98.06% of sulfamethoxazole, 95.49% of sulfadimethoxine, and 96.52% of sulfamerazine [165]. Ultrasonic irradiation effectively treats two-phase wastewater with low-solubility organics. A recent method combines biological treatment with hydrodynamic cavitation to remove pharmaceutical compounds, achieving high removal efficiencies for clofibric acid, carbamazepine, and diclofenac [166]. The schematic illustration of Sono-photocatalysis is shown in Fig. 7.

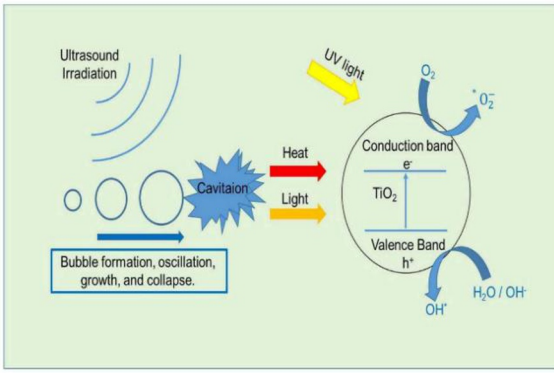

Fig.7: Sono-photocatalysis [167].

4.5 Electrochemical oxidation process

The electrochemical oxidation process uses electricity to create reactive species, eliminating the need for chemicals and reducing waste in pharmaceutical removal [168]. They effectively remove toxic organic compounds [169,170]. The electrochemical method uses hydroxyl radicals (•OH) to react with organic contaminants until complete mineralization [171]. Electrochemical oxidation can occur through direct and indirect mechanisms. Direct oxidation occurs at the anode, while indirect oxidation occurs through the creation of reactive oxygen species by oxidants at the electrode surface [169,172,173]. Electrochemical oxidation using various anodes including BDD electrodes with NaCl electrolyte effectively decontaminates pharmaceuticals. BDD is preferred for its efficiency, corrosion resistance, and inert surfaces, displaying high thermal conductivity and chemical inertness [174,175]. To avoid electrode fouling, the process chosen relies on the electrode material and experimental setup [176]. The process of electrochemical oxidation over BDD is shown in the below equation [177].

$$BDD + H2O \longrightarrow BDD + (\cdot OH)_{ads} + H^+ + e^- \quad \text{--- (7)}$$

The schematic illustration of Sono-photocatalysis is shown in Fig.8.

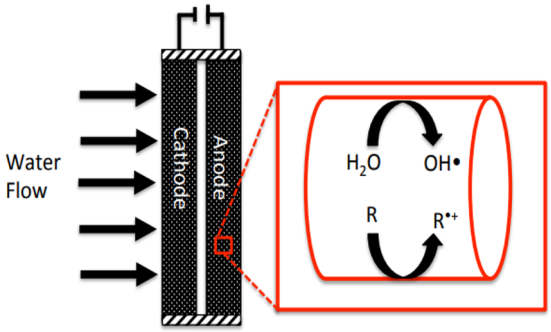

Fig.8. Electrochemical AOP of pharmaceutical wastewater [178].

The performance of pollutant oxidation depends on the working electrode, supporting electrolyte, and applied current. BDD anodes are gaining attention due to their stability and high efficiency in mineralization [179]. Sodium chloride (NaCl) is essential for increasing effluent conductivity and facilitating chlorine production for contaminant oxidation [180]. However, its utilization can result in the generation of hazardous byproducts [181]. The BDD electrochemical treatment removed over 97% of TOC in paracetamol and diclofenac-contaminated wastewater [182,183]. By increasing boron concentration and decreasing electrode thickness, antibiotic degradation was enhanced. Electrocoagulation combined with photocatalysis achieved an 86% removal efficiency [184]. Dissolved iron in the electro-Fenton process improves electrooxidation efficiency by catalyzing the degradation of H_2O_2 [184,185].

Choosing the correct anode material is crucial for improving the performance of electrochemical oxidation AOP, as it directly affects the production of hydroxyl ions. The efficiency of degradation is intricately linked to both current density and electrolyte concentration, directly impacting the system's conductivity [186]. Electrochemical-based AOP is efficient for post-RO disinfection, maximizing H_2O_2 utilization. It's versatile, safe, and environmentally friendly, but costly and requires high oxygen overpotential anodes to prevent electrode fouling. Halogenated byproducts are a concern for process efficiency [187].

4.6 UV advanced oxidation process

The UV AOP combines UV light with oxidants like H_2O_2 and O_3 to effectively break down contaminants in water, promoting the generation of free radicals for efficient wastewater treatment [187]. These radicals break down contaminants, reducing their concentration from several hundred ppm to less than 5 ppb, significantly lowering COD and TOC [188]. When using ozone as an oxidant, maintain the sample pH within the 7-8 range and start with a

lower initial pollutant concentration [189]. Ozone undergoes a crucial reaction in the presence of UV radiation as shown in below equation [181].

$2O_3 + H_2O_2 \longrightarrow 2 \cdot OH + 3\ O_2$ ------ (8)

Hydrogen peroxide is a natural metabolite that decomposes into oxygen and water. Industrially, H_2O_2 is manufactured by BASF [128]. Hydrogen peroxide undergoes a specific mechanism when it exposed to UV radiation as follows [190].

$H_2O_2 + hv \longrightarrow 2 \cdot OH$ ------ (9)

Pharmaceutical wastewater contains stubborn organic compounds that resist biodegradation. Conventional treatments are often ineffective, leading to high levels of pollutants in the treated effluent [191]. Ozone/hydrogen peroxide treatment has emerged as a cost-effective solution for addressing these challenges. Studies have shown promising results in removing high concentrations of penicillin and enhancing biodegradability [192]. Additionally, the combination of O_3 with H_2O_2 has demonstrated a remarkable 76% enhancement in COD removal efficiency. These findings underscore the potential of innovative treatment approaches in addressing the complex challenges posed by pharmaceutical wastewater [92]. It's crucial to use the optimal amount of peroxide to avoid hazardous effects. Low pH and temperature are preferred due to the reaction's kinetic nature [189].

UV-based AOPs efficiently remove contaminants from water and leave no chemical residues, making the water safe for consumption. However, they have drawbacks such as high energy consumption, expensive reagents, and limited UV light penetration in water, which require careful system design for optimal performance [187].UV photolysis of APIs depends on factors like light intensity, API response, and peroxide addition. Combining UV and H_2O_2 reduces required UV energy as degradation is not solely reliant on API absorption [193].

5. Conclusion

The pharmaceutical industry plays a crucial role in providing essential medicines, but its manufacturing process generates pollutants that can harm water quality and the environment. The increasing number of pharmaceutical industries has increased water pollution, emphasizing the importance of regularly assessing pharmaceutical industrial wastewater. New treatments, such as advanced oxidation processes, show the potential to enhance the removal of pharmaceuticals from wastewater. Advanced high-tech applications in pharmaceutical wastewater treatment have rapidly advanced the chemical and pharmaceutical industries, offering prospects for addressing high-concentration organic wastewater. The focus is on developing treatment technologies for high COD concentration, complex composition, and poor biodegradability. It is crucial to assess technical, economic, and environmental aspects, including effluent compatibility. The presence of antibiotics in the environment has become a concern due to their persistence and resistance to biodegradation. Conventional treatments have been ineffective, leading to the development of advanced oxidation processes.

AOPs significantly improve water and wastewater treatment by addressing limitations and increasing efficiency. Further research is needed to identify new pollutants, understand their effects, and develop cost-effective treatment processes. AOPs offer a promising solution to mitigate the impacts of emerging contaminants. Pharmaceutical manufacturers must adhere to strict water quality standards and regulations. It is important to consider economic factors and prioritize the recovery and reuse of pharmaceutical wastewater. Further research is vital to enhance treatment efficiencies and identify degradation compounds. AOPs show promise for improving removal rates, and a comprehensive analysis is essential for selecting the most suitable AOP based on specific characteristics of the wastewater, regulatory requirements, and economic considerations.

6. References

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