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A Review on Treatment of Pharmaceutical Wastewater

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Author Note

The treatment of pharmaceutical wastewater is a crucial and current topic in the field of environmental engineering and sustainable practices, and I'm happy to give this in-depth review on it. This study is the result of in-depth investigation, consultation with industry leaders, and a serious desire to shed light on the issues and developments surrounding the treatment of this particular type of industrial wastewater.

Abstract: *Pharmaceutical wastewater has a complex composition that includes a high concentration of organic matter, microbial toxicity, a lot of salt, and difficulty in biodegrading. Trace amounts of dissolved organic materials and suspended particles remain after further treatment. Advanced treatment is necessary to raise the effluent quality of pharmaceutical wastewater. This study introduced the classification of pharmaceutical technology and provided a summary of the qualities of pharmaceutical wastewater discharge. Then, a review of advanced treatment techniques for pharmaceutical wastewater was done, including membrane separation, flotation, activated carbon adsorption, coagulation, some of the promising, practical, and sustainable methods for the degradation of pharmaceutical waste water include advanced oxidation processes (AOPs), such as photocatalysis, Fenton oxidation, ozonation, etc.*

I. INTRODUCTION

Water is a vital resource for both life and the environment, but because it is used so frequently in industrial operations, wastewater is produced, which creates serious environmental problems[3]. Novel pharmaceuticals and antibiotics have caused widespread contamination of surface water, which has eventually affected sources of drinking water. In underdeveloped countries like India, where a high disease frequency is exacerbated by poor sanitation practices, leading to greater medicine consumption, this issue is particularly serious[2]. These contaminants can be dangerous to both human health and the environment even at low concentrations (less than 1 g/L). Due to its complicated chemical makeup Pharmaceutical wastewater has high COD, and presence of non-biodegradable substances, poses a serious environmental risk. This wastewater comes from hospitals, pharmacies, pharmaceutical companies, and homes, and it frequently contains salts, used solvents, and leftover medications. Traditional therapeutic approaches are inadequate because of the complicated structural makeup and biological resistance of these pharmacological substances. Advanced oxidation methods are therefore necessary[1]. As of 2015, the global pharmaceutical market had reached USD 1.06 trillion in sales, with a steady annual growth rate of 5.2%. North America led with USD 363.2 billion in sales, followed by Europe at USD 315.1 billion and Asia at USD 281.3 billion. South Korea's pharmaceutical market showed significant growth due to rising chronic illnesses in its aging population, with a 3.4% increase in gross pharmaceutical production in 2015, totaling KRW 16,969.6 billion. The pharmaceutical market is expected to continue growing rapidly in the coming years[10].

Due to the large range of products generated in a medicine manufacturing plant, variable wastewater composition, and oscillations in pollutant concentrations, it has always been challenging to treat pharmaceutical wastewater in order to meet the acceptable effluent standards[4]. The organic chemicals that are manufactured in the pharmaceutical business have complicated structural properties and are resistant to biological deterioration. Because of this, it is frequently necessary to use advanced oxidation technologies to treat pharmaceutical wastewaters instead of using standard treatment methods[5].

II. CHARACTERISTICS OF PHARMACEUTICAL WASTEWATER

Pharmaceutical wastewater includes active pharmaceutical components that have not yet completely broken down in the body. Acetaminophen, diclofenac, antibiotics (such as sulfamethoxazole and trimethoprim), antiepileptic medications (such as carbamazepine), and painkillers are a few examples of these active ingredients. These materials can linger in the environment[11]. Pharmaceutical chemicals are often present in wastewater from pharmaceutical manufacturing facilities in relatively low amounts, frequently measured in nanograms per liter (ng/L) to micrograms per liter (g/L).

Pharmaceutical wastewater has a complicated composition that is often high in organic matter, microbially toxic, salty, and difficult to biodegrade. Additionally, the majority of pharmaceutical manufacturing facilities use batch processes, and because different raw materials and production methods are used, the wastewater produced varies greatly [1].

General characteristics of pharmaceutical waste water: [6]

Total COD mg/l	2429
BOD5 mg/l	358
BOD5/COD	0.147
TOC mg/l	702
Turbidity	16
TDS mg/l	1721
TSS mg/l	240
pH	7.2

A. Methods Of Advanced Treatment Of Pharmaceutical Wastewater And Other Effluent Drugs

Recently, enhanced treatment of pharmaceutical wastewater has become the focus of scientific study and technical applications, with physicochemical technology serving as the primary method. It denotes that wastewater is handled using physical or chemical techniques, such as membrane separation, activated carbon adsorption, flotation, coagulation and sedimentation, and advanced oxidation processes.

III. ADVANCE OXIDATION PROCESS

Advanced oxidation processes (AOPs), which adhere to the principles of green chemistry, are efficient ways to treat wastewater that are also environmentally beneficial. They reduce the production of toxic byproducts while effectively removing contaminants. The production of hydroxyl radicals ($\bullet\text{OH}$), which are essential for decontaminating and dissolving dangerous contaminants, is a major characteristic of AOPs. AOPs can be divided into several categories, each with a different mechanism for hydroxyl radical production. These categories include ozone-based processes, photocatalysis, Fenton and photo-Fenton reactions, UV-hydrogen peroxide processes, and Fenton and photo-Fenton reactions[12].

A. Photocatalysis

In photocatalysis, a semiconductor material, such as titanium dioxide (TiO_2), is exposed to light energy which helps for the AOP process[16]. Semiconductors are vital for photocatalysis because they can absorb photons (light particles) and produce the electron-hole pairs required to catalyze processes[8] [9]. When photons (particles of light) strike the photocatalyst's surface, they generate pairs of electrons (e^-) and holes (h^+) within the material[14]. These electron-hole pairs are highly reactive and can participate in various chemical reactions. Electrons can reduce certain compounds, while holes can oxidize others. In the presence of water and oxygen, the generated holes (h^+) can react with water molecules to produce hydroxyl radicals ($\bullet\text{OH}$), which are powerful oxidative species capable of breaking down organic contaminants into harmless byproducts like water and carbon dioxide[13]. A crucial characteristic of semiconductors is the band gap. The energy difference between the conduction band (LUMO - Lowest Unoccupied

Molecular Orbital) and the valence band (HOMO - Highest Occupied Molecular Orbital) is referred to. The semiconductor's capacity to absorb light of various energies depends on its band gap[7]. Operational costs might be decreased by making use of several photocatalysts over and again. By targeting particular pollutants, photocatalysts can reduce the influence on unintended chemicals[15]. Photocatalysts can be created using a variety of approaches, including as hydrothermally assisted sol-gel processes, sol-gel dip coating, electrochemical deposition, precipitation, combustion procedures, and more[9] [13] [15].

Bandgaps of semiconductors

Semiconductor	Bandgap (eV @300K)
ZnS (Wurtzite) 3.91	3.91
ZnS (Zinc blende) 3.54	3.54
SnO ₂ 3.60	3.60
TiO ₂ 3.20	3.20
ZnO 3.03	3.03
WO ₃ 2.60	2.60
CdS 2.42	2.42
Fe ₂ O ₃ 2.20	2.20
CdO 2.10	2.10
Cu ₂ O 2.10	2.10

5[17]

The advantages of using hydrogen peroxide as a treatment, increases the efficiency or helps the catalyst to break down the waste provided. The target chemical is only 52% removed by TiO₂ and H₂O₂, ferric ions increase the removal efficiency of some substances (such as 2,4,6-trichlorophenol) in the UV/H₂O₂ treatment[15].

WO₃/CNT catalyst concentrations ranging from 0.1 to 1 g/L was used to treat Tetracycline and pharma wastewater ,however beyond 0.7g/L catalyst aggregation and diminished ultrasonic efficacy caused the removal efficiency to degrade[6].

Another issue that arises is that in increased opacity of the solution at high TiO₂ concentrations can prevent UV light from passing through, decreasing the efficiency of the process.

Then too 100% IBU removal was obtained with catalyst loads of 2.0 g L⁻¹[9]. The MWCNT/TiO₂ nanocomposite significantly increased TC removal when compared to plain TiO₂, most likely because of improved electron-hole separation and the function of MWCNT as a photosensitizer. Here COD and TOC contents of pharmaceutical wastewater were reduced to 342 mg/L and 228 mg/L when TiO₂ nanocomposite was used[8].

Photocatalysis is a widely employed method in wastewater treatment to efficiently remove pollutants. It is considered the most cost-effective Advanced Oxidation Process (AOP) for breaking down water contaminants. This approach is extensively utilized across various industries due to its effectiveness and environmentally friendly nature.

B. Ozonation (O₃ Processes)

Ozonation is a wastewater treatment method that relies on ozone gas (O₃) to eradicate pollutants[20]. The ozone dosage is determined by producing ozone, guiding it through a solution, neutralizing it with sulfuric acid, and measuring it using sodium thiosulfate. This approach is highly efficient in eliminating various contaminants, such as pharmaceuticals, as well as organic and inorganic pollutants from wastewater[18][19]. The main objective of ozonation is to treat or eradicate excessive amount of CODs and BODs present in the wastewater[18]. Ozone is very effective at disinfecting water by eliminating bacteria, viruses, and other pathogens.

It can be used to disinfect water instead of or in addition to chlorine. Ozone does not create toxic disinfection byproducts in contrast to chlorine. In recent years, catalytic ozonation with heterogeneous catalysts has received much attention in the field of water treatment, because of its potentially significant effectiveness and minimal detrimental effect on water quality, Activated carbon, graphene, carbon nanotubes, and other types of carbon materials, metal oxides, metal oxides bonded to supports, and natural minerals are the main types of catalysts used in ozonation. Metal-based catalysts (Ag, Al, Ti, Fe, and others) are one type of these catalysts[23].

Advantages for Ozonation processes:

- High reaction rate, leading to more OH generation as a result of O₃ splitting;
- Useful for pretreatment prior to water purification
- More stable byproducts

[\[22\]](#) [\[24\]](#)

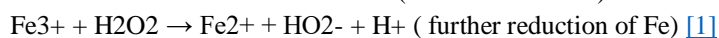
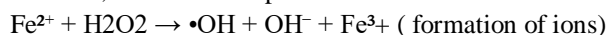
Ozone-based processes can be broadly categorized into two main types: ozone/hydrogen peroxide and ozone/UV[\[25\]](#). The ozone/hydrogen peroxide method is commonly preferred in wastewater treatment due to its ability to enhance the production of hydroxyl radicals through the dissociation of H₂O₂. On the other hand, in ozone/UV processes, UV radiation triggers the photolysis of O₃ to generate H₂O₂, which subsequently generates the required OH radicals for water treatment[\[26\]](#).

A UV/O₃/TiO₂ system was very useful to treat the groundwater including chlorinated ethene compounds (which are very difficult to remove)[\[22\]](#), where trichloromethane (TCM) was eradicated by 84%. For pharmaceutical waste removal, a catalyst Fe-Z was used with ozonation process which ensure us the removal of specific pharmaceutical compounds (90% removal rate for amoxicillin)[\[19\]](#). A custom made Fe foam was used with ozone which encouraged the production of hydroxyl radicals also improved utilization on ozone. It helped to remove excessive BOD and COD present [\[18\]](#).

Due to the O₃ splitting reaction, ozone-based processes generate the strongest oxidants which significantly remove the organic matter and helps us to treat the wastewater [20]-[24], but O₃ based are highly sensitive to pH levels. Various pH level hinders the procedure of the given processes[\[23\]](#). Generally this kind of processes are used in treatment of drinking water.

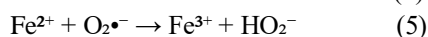
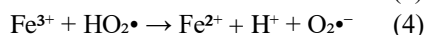
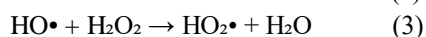
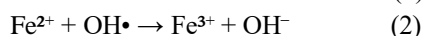
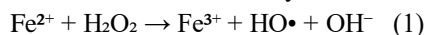
C. Fenton-Like Processes

Fenton-like procedures are a class of improved oxidation methods drawn from the traditional Fenton process. In the Fenton method, ferrous ions (Fe²⁺) and hydrogen peroxide (H₂O₂) are used to create very reactive hydroxyl radicals (OH•)[\[5\]](#). After being produced, hydroxyl radicals interact with different pollutants found in the wastewater, the organic molecules are attacked by the hydroxyl radicals, which break up chemical bonds and create a number of intermediate intermediates.



Although the reaction rate may change based on pH, it may function under a wide range of pH. Generally acidic pH helps in production •OH ions. When properly regulated, it does not produce any toxic byproducts[\[1\]](#). The Fenton method has the unusual capacity to precipitate pollutants as solid phases, this particularly is beneficial for COD & BOD treatment[\[27\]](#).

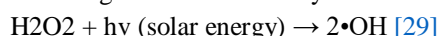
The main reactions of the Fenton system are shown below[\[27\]](#).



Because of how quickly and strongly these radical reactions occur, the Fenton process is very effective.

In recent years, the photo-Fenton process, a variation of Fenton reaction, has drawn a lot of interest for its efficiency in tackling challenging water quality issues. This sustainable and environmentally acceptable method uses solar radiation to create highly reactive hydroxyl radicals (HO•), which are essential for the breakdown of a variety of pollutants[\[12\]](#).

The rate of pollutant breakdown is accelerated when UV light is combined with Fe²⁺ or Fe³⁺. Only the buildup of the process inhibitory Fe³⁺ ions cause this photo-Fenton AOP to be hampered. However, additional OH radicals are created during this AOP by photochemical regeneration caused by UV/solar radiation.



The key advantage of this process is utilization of solar as an ecofriendly source[\[28\]](#).

Yunus ahmed came up with EDDS modified photo-Fenton method for removal of ARGs (Antibiotic Resistance Genes), MPs (Microplastics), and ARBs (Antibiotic-Resistant Bacteria) from various water matrices at neutral pH. Process removed ARB within 30 mins and MPs within 10 mins[\[28\]](#). The solar-fenton process also showed excellent PCT (paracetamol) removal efficiency which was 99.1%, 6.0 pH was used as desiring derivative for proper COD removal[\[27\]](#).

Pyrite as a catalyst was used in the Fenton oxidation process to clean pharmaceutical effluent, citrate was used as an agent that is used to mobilize iron species which increases the fenton process[30]. Liangliang Ling proposed the degradation of organic contaminants in pharmaceutical wastewater which was achieved using a Fenton-like reaction and a CoMnAl-CMO-Al₂O₃ catalyst, which demonstrated significant catalytic activity. The catalyst provided more active sites increasing the production of hydroxyl radicals and the breakdown of hydrogen peroxide (H₂O₂), which improved TOC removal[1].

D. Electro-Fenton

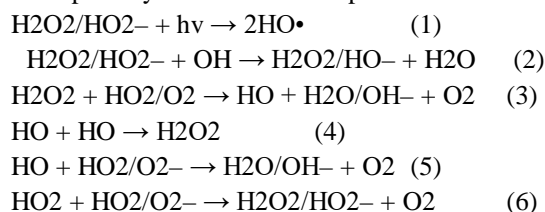
An effective advanced oxidation method is used for treating pharma wastewater is called electro-Fenton. It is very efficient at breaking down and removing a variety of organic pollutants in wastewater, including pharmaceuticals and industrial chemicals, because it depends on the electro-generation of hydroxyl radicals (\bullet OH) through the reaction between iron ions and hydrogen peroxide. The treatment of persistent and difficult-to-degrade organic contaminants makes use of this method particularly effective, which helps to clean up water supplies and lessen environmental pollution[5][31].

In contrast to conventional Fenton processes that rely on the external input of hydrogen peroxide, Electro-Fenton has the singular capacity to manufacture hydrogen peroxide on-site through the reduction of oxygen at the cathode. This makes it more cost efficient and reliable[31].

E. UV/H₂O₂ (Ultraviolet/Hydrogen Peroxide Processes):

The UV process is an advanced oxidation process (AOP) that treats and removes organic contaminants from wastewater using ultraviolet (UV) radiation. For the breakdown of different organic compounds in pharmaceutical and industrial effluent, it is a strong and efficient technique. The process uses ultraviolet (UV) radiation to treat wastewater that contains organic pollutants. In order to disinfect and deactivate microorganisms, this procedure involves exposing wastewater to UV light, specifically ultraviolet-C (UVC) radiation[32][33]. Major AOPS like UV/H₂O₂ and UV/Cl₂ were used, where H₂O₂-based process (UV/H₂O₂) was more effective at degrading some medications (PRM, CBZ, and GMF). While UV/Cl₂ process produced reactive chlorine species (RCS) and hydroxyl radicals (\bullet OH) which higher selectivity for certain pharmaceutical compounds[34]. In UV-based, H₂O₂ is essential because it produces highly reactive hydroxyl radicals when as UV light initiates it and these radicals then reacts with organic compounds[35].

The following are the primary reactions of OH \bullet production in the H₂O₂/UV process[32]:



Regarding cost-effectiveness and convenience of use, H₂O₂/UV advanced oxidation looks to be the approach with the most promise.

The UV process was regarded by Fayaz Ali as a useful instrument in the degradation of CBZ, either by itself or in conjunction with other agents. It was established that UV irradiation may produce reactive oxygen species, particularly hydroxyl radicals, which helped to eliminate CBZ and lessen its negative effects on the environment[36].

F. Coagulation Process

During the coagulation stage of water treatment, a coagulant is introduced to the water to destabilize and bring together suspended particles. This process promotes the formation of larger particles, facilitating their swift removal through filtration or sedimentation. The addition of a coagulant helps in the agglomeration of particles, making the subsequent elimination process more efficient[37][38].

By using internal micro-electrolysis-coagulation, an amazing removal rate of 98.9% for S²⁻ (sulphide ion) and a removal rate of 72.4% for COD were successfully achieved by Kangle Wang and his teammates[39].

IV. CHARACTERISATION METHODS OF PHARMACEUTICAL WASTEWATER

1) BET

A common procedure in material science, the BET (Brunauer-Emmett-Teller) process offers a way to calculate the specific surface area of porous materials. This method was used in the investigation to assess the PS-TiO₂ photocatalyst's active surface area, pore volume, and pore size[40]. The BET study provides information on the physical properties of the photocatalyst, which are essential to its ability to effectively degrade contaminants in pharmaceutical effluent, by evaluating gas adsorption onto the material's surface[1].

Christine M. El-Maraghy, Ola M. El-Borady & Omnia A. El-Naem conducted a process involving removing of levofloxacin, specific surface area of the nanoparticles (ZnONP and GONS) was ascertained using BET analysis. In order to do this analysis, the produced samples were exposed to nitrogen gas at different pressures and temperatures. Then, the gas adsorption isotherms were measured, and the specific surface area was determined using the BET equation. The findings demonstrated that GONS has a high surface area and promise for adsorption applications, with a specific surface area of roughly 53 m²/g[4].

The BET outcomes show that Pumice Stone (PS) has a surface area of 0.4 m²/g, which indicates that there are few pores and low porosity because of the hollow spaces[43].

2) TEM

TEM refers to Transmission Electron Microscopy, a characterization technique used to analyze the structure, morphology, size, and shape of ZnO (zinc oxide) nanoparticles synthesized for the degradation of norfloxacin, an antibiotic, in wastewater treatment. The TEM analysis helps determine the properties of the nanoparticles, such as their size distribution and crystallinity, which are crucial for understanding their catalytic efficacy in the degradation process[4][41].

S. Vijayalakshmi and his mates found out that TEM images reveal the morphology of ZrO₂ nanoparticles, showing spherical shapes. TEM images also depict the PANI/ZrO₂ nanocomposite, showing ZrO₂ nanoparticles attached to PANI structures[42].

3) XRD

The term "X-ray diffraction" (XRD) refers to a method for examining a material's crystal structure [44]. A sample is exposed to X-rays during an XRD study, and the resultant diffraction pattern tells us something about how the atoms are arranged in the material[43][42].

Authors saw a sharp diffraction peak at $2\theta = 25.65^\circ$ is associated with the anatase TiO₂ (101) plane. Other crystallographic planes of anatase TiO₂ include (004), (200), (211), (204), (116), (215), and (303) were also observed [45]. The crystalline phases found in the samples and the interactions between the various components in the composites are both well-understood by using the XRD measurements. The XRD results obtained from the analysis of the CoMnAl-CMO-Al₂O₃ catalyst after reaction were changes in peak intensities, peak positions, or the appearance of new peaks[1].

The hexagonal wurtzite phase of ZnO was identified by different diffraction peaks in the XRD pattern. This shows the synthetic material's crystallographic orientation[46]. Specific 2-Theta angles were used to examine the diffraction peaks, which usually ranged from 20 to 70 degrees- by Bhawana Jain. Pumice stone (PS) features an amorphous silica phase (SiO₂) and peaks that show the presence of Fe₂O₃, Na₂O, and Al₂O₃. The anatase and rutile phases of TiO₂ are represented by peaks in PS-TiO₂, suggesting that TiO₂ was successfully impregnated onto the PS material[43].

4) FTIR

Fourier Transform Infrared Spectroscopy is referred to as FTIR. This method measures the amount of infrared light absorbed by a sample in order to determine its chemical makeup[42] [46]. A wideband at around 3400 cm⁻¹ indicates the presence of hydroxyl groups (OH).

ZrO₂ is proven to be present by peaks at 510 cm⁻¹ (ZrO₂ stretching) and 743 cm⁻¹ (Zr-O-Zr symmetric stretching) [42].

Characteristic absorption bands corresponding to particular functional groups and chemical bonds were found in the FT-IR investigation of ZnONP and GONS[4].

ZnONP: Zn-O stretching at 1631 cm⁻¹, C-O stretching at 1473 cm⁻¹, and O-H vibration at 3430 cm⁻¹.

Stretching O-H at 3425 cm⁻¹ and C=O at 1735 cm⁻¹ are the GONS.

The presence of TiO₂, ZnO, chitosan (CS), and graphene (Gr) in the nanocomposites was verified by the FTIR spectra. Because each component had unique absorption bands, it was possible to identify and characterize them[45].

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