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Recent Technologies for The Elimination of Pharmaceutical Compounds from aqueous solutions: A review

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ABSTRACT

Water pollution is a major environmental issue that has a wide range of impacts on ecosystems, human health, and the economy. During recent years, pharmaceutical compounds have been considered an emerging water micro-pollutant due to their potential eco-toxicity. Pharmaceuticals, personal care items, steroid hormones, and agrochemicals are synthetic and indigenous products that makeup micropollutants, also known as emerging contaminants. Due to the Corona pandemic (Covid-19), excessive use of antibiotics in addition to the painkillers used in the treatment protocol established by the World Health Organization. Thus, these compounds have become in water sources with higher concentrations. New techniques have been used to remove these environmental pollutants to achieve sustainable development goals (SDGs). This article provides a critical review of various methods presenting the potential to be applied for removing pharmaceuticals from water. Several processes: adsorption, advanced oxidation processes (photodegradation, photocatalysis, ozonation, Fenton reaction, Wet Air Oxidation, ultraviolet radiation, hydrogen peroxide oxidation), and membrane-based techniques (ultrafiltration, nanofiltration, reverse osmosis, forward osmosis, membrane distillation) are analyzed, and their performance during removal of pharmaceuticals from water is compared. Moreover, summaries presenting the efficiency of various materials applied during each process are provided. Finally, the advantages and disadvantages of each proposed method are summarised and comprehensively discussed.

1. Introduction

1.1 Water Pollution

The most important natural resource in life worldwide is water, which is essential for human survival and the survival of all other living organisms, food production, and economic growth. Nowadays, water is scarce in many cities across the world. Nearly 40% of the world's food supply is produced using irrigation systems, and various industrial operations rely on water (Halder and Islam 2015). Currently, water shortages affect more than four billion people. By 2025, it is expected that most nations will be dealing with water scarcity and shortage issues (Elgarahy et al. 2023a). The water supply has been and will continue to be one of humanity's main issues in the twenty-first century due to the unsustainable and rising worldwide demand for water. Water resources will come under increasing strain locally and worldwide due to water withdrawals in areas already suffering scarcity, posing a severe threat to accomplishing long-term sustainable development goals (Dalstein and Naqvi 2022).

Another problem that developed countries may face is water contamination. Heavy metals, persistent organic pollutants, and waste materials are often found in groundwater and other water sources, negatively impacting human health and the environment (Dharwal et al. 2020). Most studies have shown that these harmful chemicals are found in the environment in higher quantities due to the proportionate increase in industrial, agricultural, and residential activities brought on by the expanding human population (Elgarahy et al. 2023b). Such pollutants have reportedly been linked to poisonous accumulation and human carcinogenic effects (Hiew et al. 2018).

1.2.Contamination of water by pharmaceutical compounds

Pharmaceutical compounds are crucial in treating and preventing diseases in humans and animals and are considered necessary globally. They are increasingly present in the environment due to their widespread consumption. These compounds have been found in drinking water, surface water, wastewater, and sediments in amounts between g/L and mg/L (aus der Beek et al. 2016). More than 3000 regularly used pharmaceutical chemicals have been identified and evaluated in the

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European Union market water samples (Chandel et al. 2022). The utilized pharmaceutical compounds for human and veterinary pruposes are mostly discharged into the environment. It is crucial to understand that all municipal sewage, regardless of location, will contain pharmaceuticals due to metabolic excretion, improper disposal, or industrial use, and that each geographical area will differ only in terms of the types, amounts, and relative abundances of specific pharmaceuticals (de Jesus Gaffney et al. 2015).

According to studies, more than 80 different types of pharmaceutical compounds, including nonsteroidal anti-inflammatory drugs, painkillers, tranquilizers, antidepressants, lipid-lowering medications, hormones, chemotherapy drugs, and antibiotics, among others, were found in water streams in many developed countries, as result of their use in hospitals and health institutions (Sharma et al. 2021). Such pharmaceutical substances are manufactured or natural chemicals that have been demonstrated to act in human and animal bodies via a particular mechanism (González-González et al. 2022). These pharmaceuticals may undergo several degradation mechanisms when they enter the aquatic environment before being entirely metabolized, as presented in Fig. 1. These compounds could also be bioaccumulated in the aquatic environment, where they frequently harm all living organisms (Liu et al. 2018).

Most of the published studies concluded that psychiatric medications, anti-hypertensives, anti-inflammatory drugs, and antibiotics are the therapeutic classes that are most frequently found in aquatic environments (Maged et al. 2020b, c; Abu El-Magd et al. 2022; El-Borhamy et al. 2022). Additionally, pharmaceutical compounds such as carbamazepine, ibuprofen, diclofenac, sulfamethoxazole, ciprofloxacin, and norfloxacin were found in the aquatic ecosystem, as shown in Table 1. Antibiotics and anti-inflammatories are of concern due to their growing global consumption (Phoon et al., 2020). It is well-established that there is a correlation between consumption rates and pharmaceutically active compound concentrations in ecosystems (Nannou et al., 2020). These substances are not entirely eliminated during water and wastewater treatment and are still detected in treated wastewater and drinking water in different locations (Dos Santos et al., 2021).

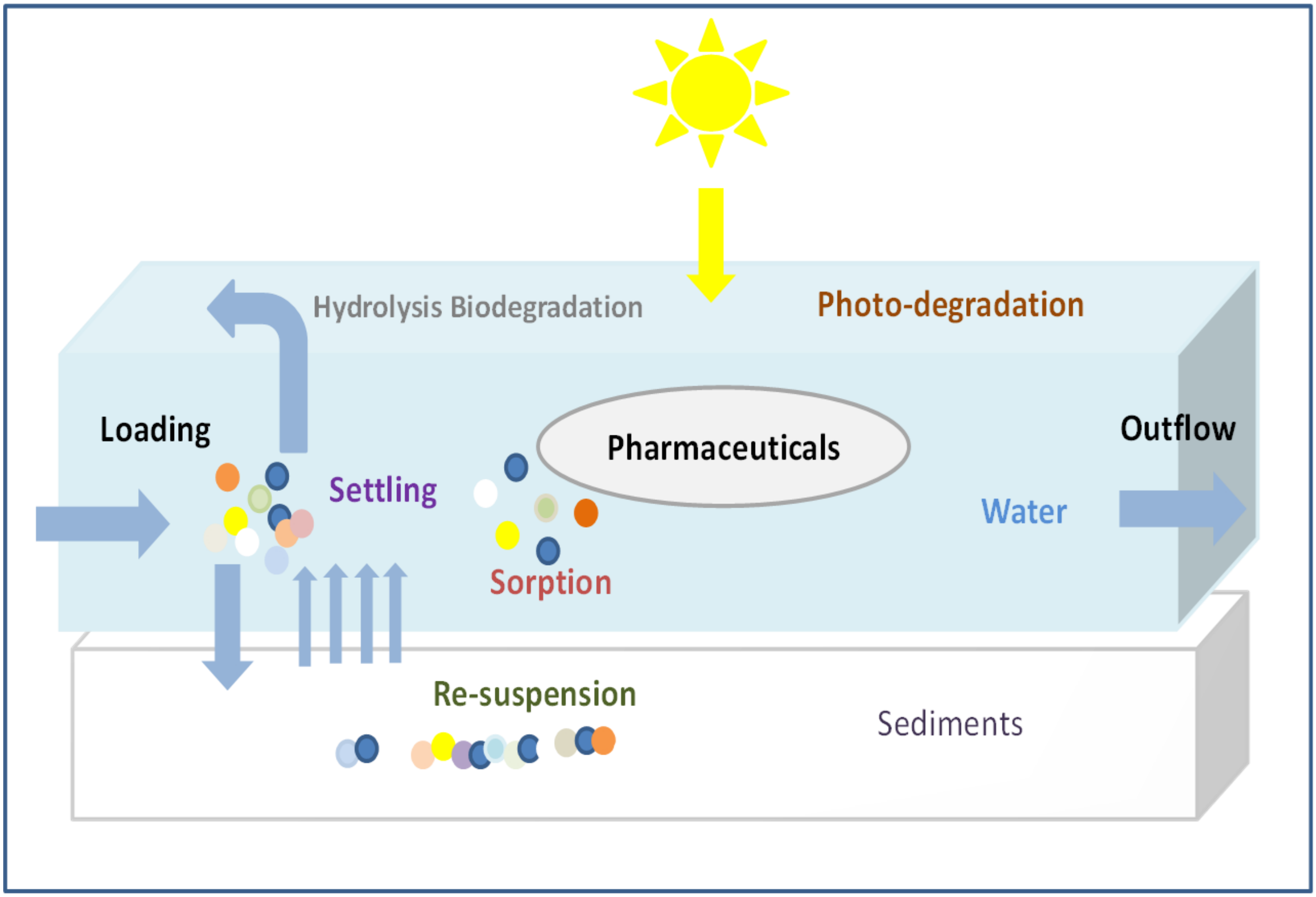

Figure 1. The mechanisms of degradation of pharmaceutical compounds in the aquatic environment.

Table 1: Sources and concentration of pharmaceuticals in the aquatic ecosystem.

Class	Pharmaceutical compound	Country	Source	Concentration µg/L	Reference
Anti-inflammatories	Acetaminophen	India	River water	11.55	(Archana et al., 2016)
		California	Groundwater	0.18	(Fram & Belitz, 2011)
		Singapore	Wastewater treatment plants	0.09	(Tran & Gin, 2017)
Anti-inflammatories	Diclofenac	China	River	67.00	(Dai et al., 2015)
		Spain	Drinking water	0.001	(Carmona et al., 2014)
Anti-inflammatories	Ibuprofen	Pakistan	Wastewater	703	(Ashfaq et al., 2017)
		India	River	0.20	(Shanmugam et al., 2014)
		Spain	Groundwater	0.20	(López-Serna et al., 2011)
Anti-hypertensives	Atenolol	USA	Wastewater treatment plants	0.20	(Lara-Martín et al., 2014)
		USA	Drinking water	0.03	(Padhye et al., 2014)
	Gemfibrozil	Singapore	Wastewater treatment plants	0.337	(Tran & Gin, 2017)
		USA	River	7.80	(Sengupta et al., 2014)
Psychiatric drugs	Caffeine	Singapore	Wastewater treatment plants	4.123	(Tran & Gin, 2017)
		USA	Drinking water	11.60	(Padhye et al., 2014)
		S. Africa	Wastewater	1.041	(Matongo et al., 2015)
	Carbamazepine	India	Wastewater treatment plants	2100	(Fick et al., 2009)
		Singapore		0.323	(Tran & Gin, 2017)
Antibiotics	Ciprofloxacin	India	River water	41.84	(Archana et al. 2016)
		Spain	Groundwater	0.012	(López-Serna et al., 2011)
	Norfloxacin	India	Wastewater treatment plant effluent	55	(Fick et al., 2009.)
		China	Wastewater treatment plant effluent	0.010	(Zhang et al., 2015)
	Ofloxacin	Spain	Fresh water	5.617	(Rodríguez-Navas et al., 2013)
		Spain	Groundwater	0.024	(López-Serna et al., 2013)
		Canada	Wastewater treatment plant effluent	0.120	(Guerra et al., 2014)
	Sulfamethoxazole	S. Africa	Wastewater	1.240	(Matongo et al., 2015)
	Tetracycline	Canada	Wastewater treatment plant effluent	0.053	(Guerra et al., 2014)
		China	Wastewater treatment plant effluent	0.002	(Sengupta et al., 2014)

Because of the potential for these drugs to be hazardous, even in low quantities, the assessment of wastewater treatment methods to achieve effective micropollutants removal is the focus of modern research, which is thus primarily concerned with risk elimination or reduction (Dos Santos et al., 2022). Most of these micropollutants are partially eliminated by wastewater treatment plants because they lack specialized equipment to overcome their low concentration. In this review, we will survey the most important traditional and advanced methods that have been recently monitored in many studies and may be effective in removing pharmaceutical compounds from the aqueous medium, especially those found in low concentrations in drinking water.

2. Methods of Pharmaceutical compounds removal from wastewater

2.1 Adsorption technology

The adsorption process is a surface activity where a soluble chemical (adsorbate) is extracted from a fluid by encountering a solid surface (adsorbent). This method has a great chance of getting rid of remaining organic and inorganic substances that may be found in wastewater (Titchou et al., 2021). The adsorbent and adsorbate molecules interact through interactive forces; this may take place via chemisorption (attributed to the formation of a strong covalent bond because it involves the transfer or sharing of electrons) or physisorption (due to poor electrostatic interaction between the adsorbent and adsorbate) (Tan & Hameed, 2017)**.** This interaction occurs through changes in functional groups, hydrogen bonds, electrostatic interaction, and the donor-acceptor electron mechanism instead of altering the structure of the adsorbent during the interaction, as shown in Figure 2 (Duarte et al., 2022).

Adsorption is thought to be a powerful method for treating pharmaceutical effluent. The advantages of adsorption over other technologies are its low cost, the reusability of its adsorbents, ease of handling, etc. Table 2 shows the drugs that were removed from the water using different adsorbents (Zhang et al. 2019). Several organic and inorganic substances, including zeolites, clay, polymers, graphene, carbon nanotubes, and agricultural waste, have been used as adsorbents to extract drugs from water (Eniola et al., 2022).

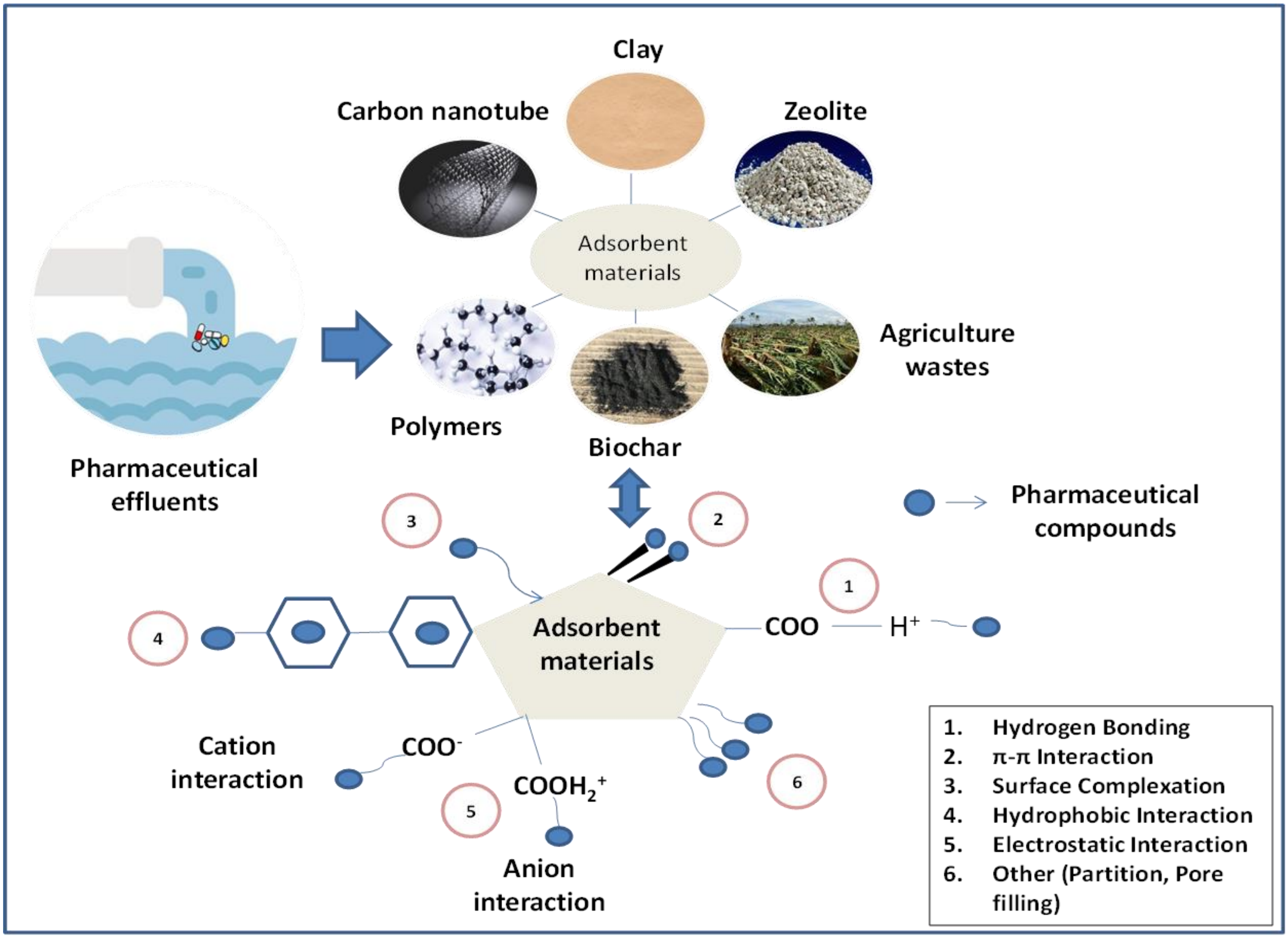

Figure 2. Adsorption mechanisms for removal of pharmaceutical compounds from water.

Table 2: Summary of Adsorption capacity of different adsorbents for removing pharmaceutical compounds from water.

Pharmaceutical compounds	Adsorbent	Adsorption capacity (mg/g)	Reference
Diclofenac	Polyamidoamine silica	134	(Lotfi et al. 2019)
	Carbon nanosphere	27.3	(Feng et al. 2018)
Ibuprofen	Polyamidoamine silica	124	(Lotfi et al. 2019)
Ketoprofen	Polyamidoamine silica	112	(Lotfi et al. 2019)
Tetracycline	Agricultural adsorbent Hazelnut shell–derived activated carbon	321.5	(Fan et al. 2016)
	Multi-layered graphene phase Biochar	388.33	(Li et al. 2017)
Oxytetracycline	Agricultural adsorbent Hazelnut shell–derived activated carbon	313.5	(Fan et al. 2016)
Chlortetracycline	Agricultural adsorbent Hazelnut shell–derived activated carbon	302.9	(Fan et al. 2016)
Amoxicillin	Activated carbon	234.6	(Balarak et al. 2017)
Norfloxacin	Powdered activated carbon	124	(Fu et al. 2017)
Ampicillin	Clay-based adsorbents Montmorillonite nanoparticle	134.48	(A.O 2012)
Sulfamethoxazole	Clay nanosorbent	11.2 0.12	(Martínez-Costa et al. 2018)
Acetylsalicylic acid	Functionalized magnetic Al-MOF Fe_3O_4@C Matrix	234.02	(Li et al. 2020)

2.1.1 Clays

Clays are aluminosilicate natural resources that are found in nature. Its natural abundance makes it a reasonably affordable adsorption material. Kaolinite, montmorillonite, and bentonite are common clay minerals (Eniola et al., 2022). Montmorillonite consists of a 2:1 layered phyllosilicate with one dominant alumina octahedral layer and two tetrahedral silicate layers. It is the most prevalent type of clay mineral tested for adsorptive antibiotic removal from aqueous solutions (Hacıosmanoğlu et al., 2022). It has a high surface area, specific adsorption capacity, swelling characteristics, and cation exchange capacity (Y. Yang et al., 2019). Whereas kaolinite, with a structure of $Al_2Si_2O_5(OH)_4$ is a 1:1 layered phyllosilicate mineral with one silica tetrahedral sheet and one alumina octahedral sheet in each layer. It is the second most typical clay mineral utilized in studies on the adsorption of antibiotics. Furthermore, bentonite is a composite material mostly made of montmorillonite (i.e., quartz, feldspar, and calcite), in addition to other minerals (Hacıosmanoğlu et al., 2022). It has been used to eliminate cationic and anionic contaminants (Maged et al., 2020). Gulen and Demircivi (2020) studied montmorillonite clay to eliminate the drug ciprofloxacin from water. Under the experimental circumstances, the system showed good stability after 60 min, with a 113.64 mg/g adsorption capacity being obtained. The maximum amount of ciprofloxacin that the montmorillonite clay could absorb was 98%, and the maximum amount of adsorption achieved at pH 7 was 128 mg/g.

Ibuprofen, paracetamol, aspirin, diclofenac, carbamazepine, and diazepam-anti-epileptic medicines were studied for their ability to be adsorbed to kaolinite material by (Hounfodji et al., 2021). Paracetamol is the most strongly adsorbed molecule on kaolinite, with an adsorption energy of -159.4 kJ/mol, whereas diazepam is the least adsorbed, with an adsorption energy of -96.8 kJ/mol, even though interactions with dispersion only adsorb this. The authors also demonstrate that there is no danger of harmful by-products being produced when these molecules are adsorbed on kaolinite. All these compounds can be removed by heating the clay to 360 K, proving the material's reusability. Consequently, kaolinite is a promising and affordable possibility for removing pharmaceutical chemicals from wastewater. Moreover, Maged et al. (2020) used modified natural bentonite clay by thermal activation to remove the tetracycline drug from water through the adsorption technique. Thermally activated bentonite (TB), according to the Brunauer–Emmett–Teller (BET) study, has superior characteristics over bentonite clay. Thermally activated bentonite had a surface area that was much larger than that of bentonite clay. Al-OH, Si-O, and Si-O-Si functional groups were seen in the sample's FTIR spectra, which supported the theory that the clay contains hydrated aluminosilicate. Through optimization experiments, the effects of different operating factors were examined. For bentonite clay and thermally activated bentonite, the highest monolayer adsorption capacities were reported to be 156.7 and 388.1 mg/g, respectively.

2.1.2 Biochar

Biochar is a stable material that is manufactured from the pyrolysis/ carbonization of biomass as a carbon-rich by-product. Pharmaceuticals and other organic pollutants have recently been removed using biochar as an alternative, affordable adsorbent because of its unique qualities, such as its wide surface area, porous structure, and surface functional groups (Ihsanullah et al., 2022). Bimová et al., (2021) reported the use of biochar in removing pharmaceutical compounds from solutions by adsorption mechanism. The sorption efficiency of a few specific medicines was examined due to their frequent occurrence in aquatic systems. The findings demonstrate that the best biochar sorption efficiency (>90%) was obtained for the following drugs: tramadol, venlafaxine, o-desmethylvenlafaxine, cetirizine, diclofenac, fexofenadine, irbesartan, metoprolol, and venlafaxine.

2.1.3 Modified and synthetic porous adsorbents

Adsorbent materials are modified to improve their physicochemical properties, which enhance the efficacy of medicines' adsorption onto the materials. Nanomaterials are extraordinary types of adsorbents due to their exceptional qualities, such as high specific area and porosity, high surface area, possibilities for fabrication into various morphological forms, possibilities for customized functionalization, and higher adsorption efficiencies with a fast adsorption rate. Nanoadsorbents can be categorized according to their composition. Nanometal oxide supports, and carbon nanomaterials are two of the main nano adsorbents that have been employed to remove antibiotics from an aqueous medium (Eniola et al., 2019). Ravikumar et al., (2019) investigated the green synthesis method for producing NiFe nanoparticles utilizing an ethanolic extract of punica granatum (pomegranate) peels to eliminate tetracycline drug from aqueous solutions. Using the response surface methodology technique, the clearance of tetracycline was optimized for the amount of nanoparticles present. 93 ±1.55% of the tetracycline could be removed under the ideal conditions (starting tetracycline concentration: 20 mg/L; NiFe nanoparticles concentration: 300 mg/L; interaction time: 90 min). The elimination was determined to be 77 ±1.12 and 70 ±2.02 percent, respectively, when the application potential of the NiFe nanoparticles was evaluated in groundwater and lake water samples spiked with tetracycline.

Furthermore, two different types of novel phosphate-based porous organic polymers (P-POP-1 and P-POP-2) were created using a Friedel-crafts reaction and a simple one-pot synthesis. P-POP-1 was created using diphenyl phosphate and 1,1,2,2-tetraphenylethylene as a precursor, and P-POP-2 was created using diphenyl phosphate and 1,3,5-triphenylbenzene. Thermal gravimetric examination revealed that P-POP-1 and P-POP-2 have intact phosphates and porous structures with specific surface areas of 714 and 581 m^2/g, respectively, and mesoporosity of 19.6 and 32.5 percent. The maximal adsorption capacity of P-POP-2 was outstanding (caffeine 301 mg/g, diclofenac 217 mg/g, and carbamazepine 248 mg/g). A shorter adsorption equilibrium time of 50 min was also demonstrated by P-POP-2 than by other materials. In the presence of additional competing cations and humic acid, P-POP-2 also showed substantial pharmaceutical drug removal (>95% for caffeine and >82 % for carbamazepine for diclofenac). Using P-POPs repeatedly for five cycles revealed a 10% decline in adsorption capabilities. The findings of this study suggest that a porous organic polymer material based on phosphate may be a suitable adsorbent for extracting medicinal substances from water (Ravi et al., 2020).

2.2 Biological treatment methods

Worldwide wastewater treatment plants currently use biological treatments, which are well-established water treatment techniques. Biological systems offer several microenvironments where processes for physical, chemical, and biological waste disposal can occur. Additionally, it has been demonstrated that biological systems can effectively clean wastewater, particularly effluent that contains emerging compounds (Li et al., 2014). It is possible to simulate how effectively and sustainably natural ecosystems may reduce surface water contamination by treating wastewater with microorganisms (bacteria, algae, and fungi) (Taoufik et al., 2021a). These treatment methods can include anaerobic or aerobic biological reactors. Aerobic reactors were done by infusing air or pure oxygen (or by mechanically stirring). In contrast, enough dissolved oxygen (DO) is delivered (typically >2.0 mg/L) to create an environment for aerobic bacteria to flourish. On the other side, anaerobic reactors provide no oxygen to the system. Anaerobic bacteria are primarily responsible for removing nutrients and organic materials from the influent water. Despite anaerobic reactors being free from oxygen molecules and only containing bound oxygen, such as nitrates (NO_3^-), anoxic reactors have DO ≤ 0.2 mg/L (Alfonso-Muniozguren et al., 2021).

Martins et al. (2018) studied the ability of anaerobic microorganisms to remove pharmaceutical products such as ciprofloxacin and 17 β-estradiol from wastewater. The results demonstrate that 17-estradiol was only biodegraded under nitrate-reducing settings, reaching a removal of 84%, but ciprofloxacin was efficiently biodegraded under nitrate and sulfate-reducing conditions, reaching a pharmaceutical products removal superior to 80%. The studied pharmaceutical products still degraded without adding a carbon source, with ciprofloxacin degrading at a rate of 85% under sulfate-reducing settings and 62 and 83%, respectively, under nitrate-reducing conditions.

The utilization of the common freshwater diatom navicula sp microalgae for the removal of several drugs was examined. After 21 days of exposure, the results indicated that navicula sp. could remove atenolol, carbamazepine, ibuprofen, and naproxen with efficiencies greater than 90%. The degradation of sulfamethoxazole, bezafibrate, and naproxen was improved in the combination treatment compared to the removal efficiency of each pharmaceutical in the individual pharmaceutical treatments, but the removal efficiencies of carbamazepine and atenolol declined. Additionally, in the mixture with the combination of six pharmaceuticals, the presence of hydrophobic pharmaceuticals (i.e., ibuprofen and naproxen) accelerated the degradation of carbamazepine and sulfamethoxazole and inhibited the removal of atenolol, while the addition of other pharmaceuticals had no discernible effect on the removal of ibuprofen and naproxen (Ding et al., 2020).

Urban wastewater can be treated with microalgae-based systems like high-rate algae ponds to remove emerging organic pollutants. Removal effectiveness varied from 0% to 99%. According to the average removal efficiency of each group in high-rate algal ponds, they were divided into four categories: high removal (>90%: caffeine, acetaminophen, ibuprofen, methyl di-hydrojasmonate, and hydrocinnamic acid), moderate-high removal (from 60% to 90%: oxybenzone, ketoprofen, 5-methyl/benzotriazole, naproxen, galaxolide, tonalide, tributyl, tributyl phosphate, triclosan, bisphenol A and octylphenol), whereas diclofenac, benzotriazole, OH-benzothiazole, triphenyl phosphate, cashmeran, diazinon, celestolide, and atrazine all have moderate-low removal rates (from 40 to 60%), while carbamazepine, benzothiazole, methyl paraben, tris (2-chloroethyl) phosphate, and 2,4-D have poor or no removal rates (30%). The hydraulic retention period only impacted the elimination of emerging pollutants in high-rate algal ponds during the cold season; no variations were seen during the warm season (Matamoros et al., 2015).

2.3 Membrane technology

Membrane technology has become increasingly popular in the world of wastewater treatment due to its high separation efficiency, affordability, environmental friendliness, simplicity of use and maintenance, and relatively minimal environmental footprint (Esfahani et al., 2019).

A membrane is a selective layer that permits only certain constituents to flow through and prevents unwanted constituents based on size exclusion, as shown in Figure 3 (González-González et al., 2022). Inorganic and organic contaminants can be removed more effectively using these technologies, which also have advantages over conventional techniques for eliminating pharmaceutically active compounds. Membranes can remove micropollutant through adsorption, electrostatic repulsion, or size exclusion (Taheran et al., 2016). The two streams that result from membrane filtration of liquid effluent feed are retentate and permeate. The permeate stream is the water that passes through the membrane, and the retentate stream is a concentrated stream containing substances excluded by the membrane. The water permeate flow controls the filtration process's effectiveness or efficiency, and the membrane's effectiveness in rejecting the desired component determines the quality of the water permeate stream (Alfonso-Muniozguren et al., 2021). Membranes are commonly split into numerous basic categories and subcategories: microfiltration, ultrafiltration, nanofiltration, and reverse osmosis (Nasir et al., 2022a).

Microfiltration (MF) is a low-pressure membrane technology that retains suspended colloids and particles of about 0.1 and 20 micrometers. Based on physical separation, microfiltration may eliminate micrometer-sized particles such as suspended particles, significant pathogens, giant bacteria, proteins, and yeast cells. The pore sizes of microfiltration membranes range from 0.1 to 5 µm. A somewhat open membrane structure often separates particles with a diameter greater than 0.1 µm. Such membranes require low hydrostatic pressures for high contaminant rejection and solvent flux since the hydrodynamic resistance is low (Anis et al., 2019)**.**

Yu et al. (2020) produced a microfiltration membrane that uses electro-oxidation to remove refractory medicines. The electro-oxidation performance was enhanced by adding graphene/tin dioxide (G/SnO_2) to the carbon nanofibers of the membrane filter. The microfiltration membrane successfully broke down sulfamethoxazole with an efficiency of 85%. When the peristaltic pump's power was increased from 10 to 30 rpm, the permeate flux of the membrane filter increased linearly from 74 to 216.4 L/m^2 h, indicating that the membrane's pores would permit water to pass through during a rapid reaction. According to the study's findings, the membrane could break down a high concentration of sulfamethoxazole in water. Adding G/SnO_2 to carbon nanofibers increased the membrane's stability and the stability rate at which sulfamethoxazole could be broken down electrochemically. Ultrafiltration is a cutting-edge separation method that was first developed as a separation and purification method in the late 1960s. Since then, ultrafiltration membranes have undergone constant improvement, and their uses have expanded to a wide range of industries, including water purification, medical use, wastewater reclamation, and chemical recovery (Swayamsiddha & Mohanty, 2020). Other frequent benefits for ultrafiltration include removing germs, viruses, and fouling in the wastewater industry (Ahmed et al., 2020). Ultrafiltration uses a membrane with low pressure. On average, this membrane has pores ranging from 1 to 100 nm(Nasir et al., 2022b).

Kim et al., (2020) used hybrid systems of metal-organic frameworks (MOFs) and ultrafiltration (UF) to handle several pharmaceutically active substances, such as ibuprofen and 17-ethinyl estradiol, as well as organic debris from the environment (humic acid and tannic acid; ratios of 10:0, 5:5, and 0:10).

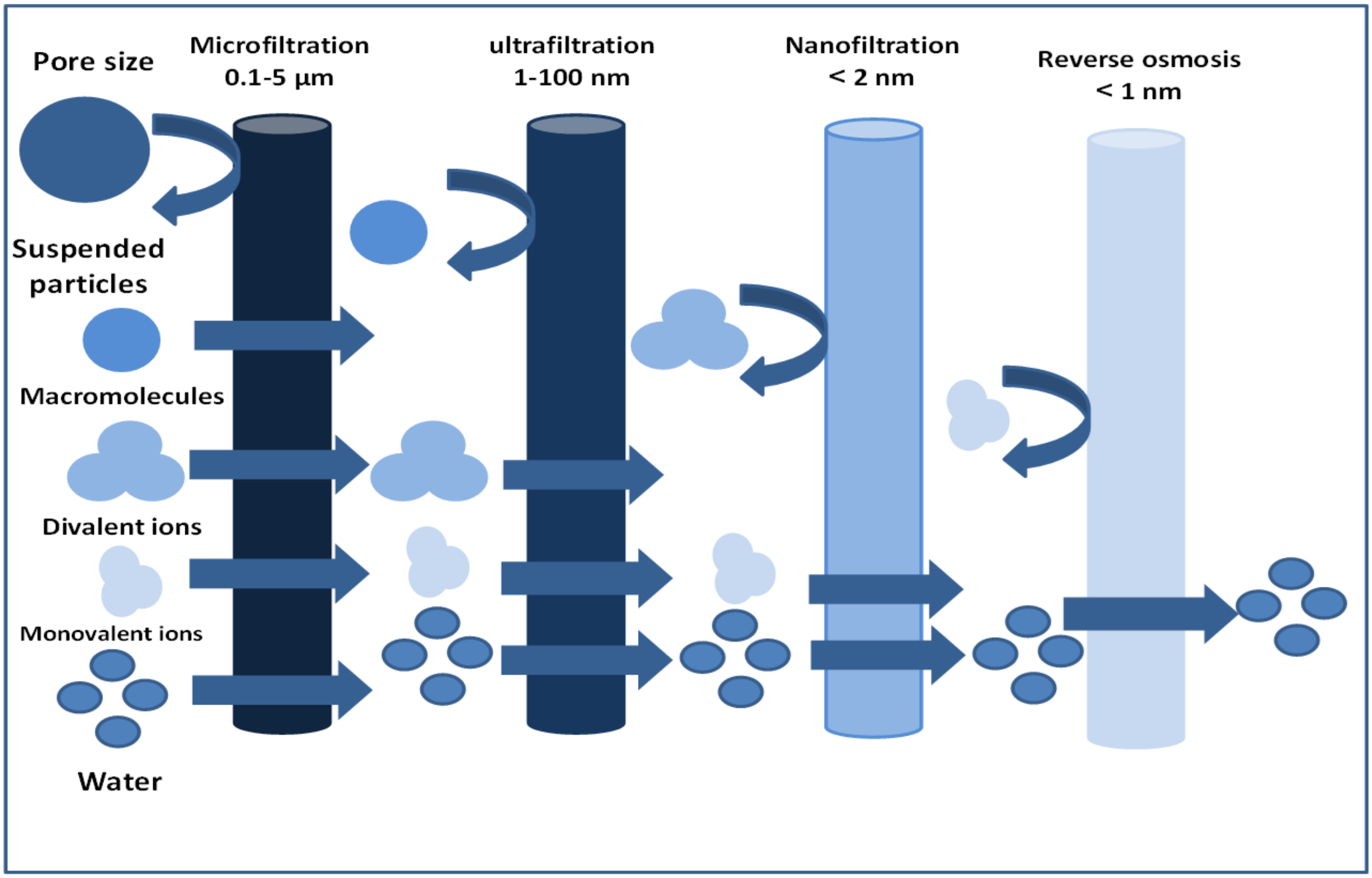

Figure 3. Representation of membrane technology mechanism for water purification.

MOFs have a significant potential for eliminating pollutants and minimizing fouling in adsorbent-UF hybrid systems because of their high adjustable porosity. Compared to ultrafiltration alone (36.7%), the average retention rate of pharmaceutically active molecules in MOF-UF (53.2%) was improved. While ultrafiltration alone had a 75.7% average retention rate for natural organic materials, MOF-UF had an average retention rate of 86.1%. Additionally, the MOF-UF flow of natural organic matter (0.79) performed better than UF alone (0.74). This is because the MOF's substantial porosity allowed the pharmaceutically active substances to be successfully adsorbed there.

Nanofiltration is a membrane separation technique utilizing membranes with pores ranging from 0.2 to 2.0 nm. Operating pressures for nanofiltration membranes typically range from 3 to 30 bars. Since most nanofiltration membranes include a little charge, the separation process of these membranes uses both size sieving and electrostatic effects (Nasir et al., 2022a). Maryam et al. (2020) investigated the filtering of specific small molecular-weight pharmaceuticals from synthetic wastewater using two loose nanofiltration membranes, typically used for high molecular-weight organics. The impact of pH on membrane function revealed that medicines' behaviors changed as pH changed. Diclofenac (99.7%) was treated more effectively than ibuprofen (81.2%) and paracetamol (49%), while also removing 95.3% of total organic carbon (TOC) and 84% of chemical oxygen demand (COD), according to the results. Unexpectedly, the nanofiltration of wastewater with ibuprofen drug rose to 90.2% compared to pure medication (80.5%). Diclofenac removal from the medication mixture was reduced (23%); however, clearance rates for ibuprofen and paracetamol rose to 17.1 and 67%, respectively.

Reverse osmosis (RO) is a water purification technique that separates ions, unwanted substances, and larger particles from water. It can remove a wide variety of dissolved and suspended chemical species and biological species (most notably bacteria) from water. As a result, only the permeate is allowed to pass through the membrane, leaving the solute confined on the pressured boundary. For this reason, most research utilizes this technique to remove pharmaceuticals from wastewater (Medhat Mohamed et al., 2022.). Couto et al. (2020) examined how membrane distillation, reverse osmosis, and nanofiltration can reject pharmaceutically active substances in the doce river in Minas Gerais, Braz is utilized to gather drinking water and dispose of raw sewage. Among the 28 pharmaceutically active substances evaluated, betamethasone and fluconazole were the two that came up most frequently during monitoring. The outcomes demonstrate that when the permeate recovery rate rises, the rejection of pharmaceutically active substances by nanofiltration and reverse osmosis membranes reduces. Membrane distillation excludes pharmaceutically active chemicals because of their poor volatility, whereas nanofiltration and reverse osmosis do so primarily through size exclusion and hydrophobic interactions. Hollman et al. (2020) investigated the removal of pharmaceutically active compounds by reverse osmosis from wastewater. The results showed that metformin, cotinine, trimethoprim, caffeine, venlafaxine, carbamazepine, erythromycin, and fluoxetine drugs were removed in the following percentages: 92.6, 99.0, 99.6, 97.8, 99.0, 99.6, and 99.2%.

2.4 Membrane bioreactor technology

Membrane bioreactors are one of the most efficient treatments that combine traditional biological degradation with membrane filtering technology, owing to its many benefits, including low sludge creation, high pathogen and virus elimination, and the production of high-quality effluent (Díaz et al., 2016). Due to recent technological advancements and the significantly lower cost of the used membranes, membrane bioreactors are the center of interest for municipal wastewater treatment (Monteoliva-García et al., 2020). The three elimination processes for emerging compounds in membrane bioreactor treatment are sorption, biological degradation, and membrane separation. The two main mechanisms were identified as adsorption and biodegradation. Additionally, the pH, dissolved oxygen (DO), solids retention time (SRT), hydraulic retention time (HRT), and other factors may affect how well microbial sludge removes drugs from a membrane bioreactor's system (Ma et al., 2018).

Membrane bioreactors have developed during the past few decades as an alternative to traditional activated sludge treatment. They separate the sludge retention period and hydraulic retention time by combining microfiltration or ultrafiltration membrane with traditional activated sludge. To provide superior effluent quality, a minor environmental impact, and less sludge generation, membrane bioreactors can be operated at higher biomass concentrations and longer sludge retention times. So it can improve the removal of micropollutants to promote wastewater treatment and reuse (W. Liu et al., 2022).

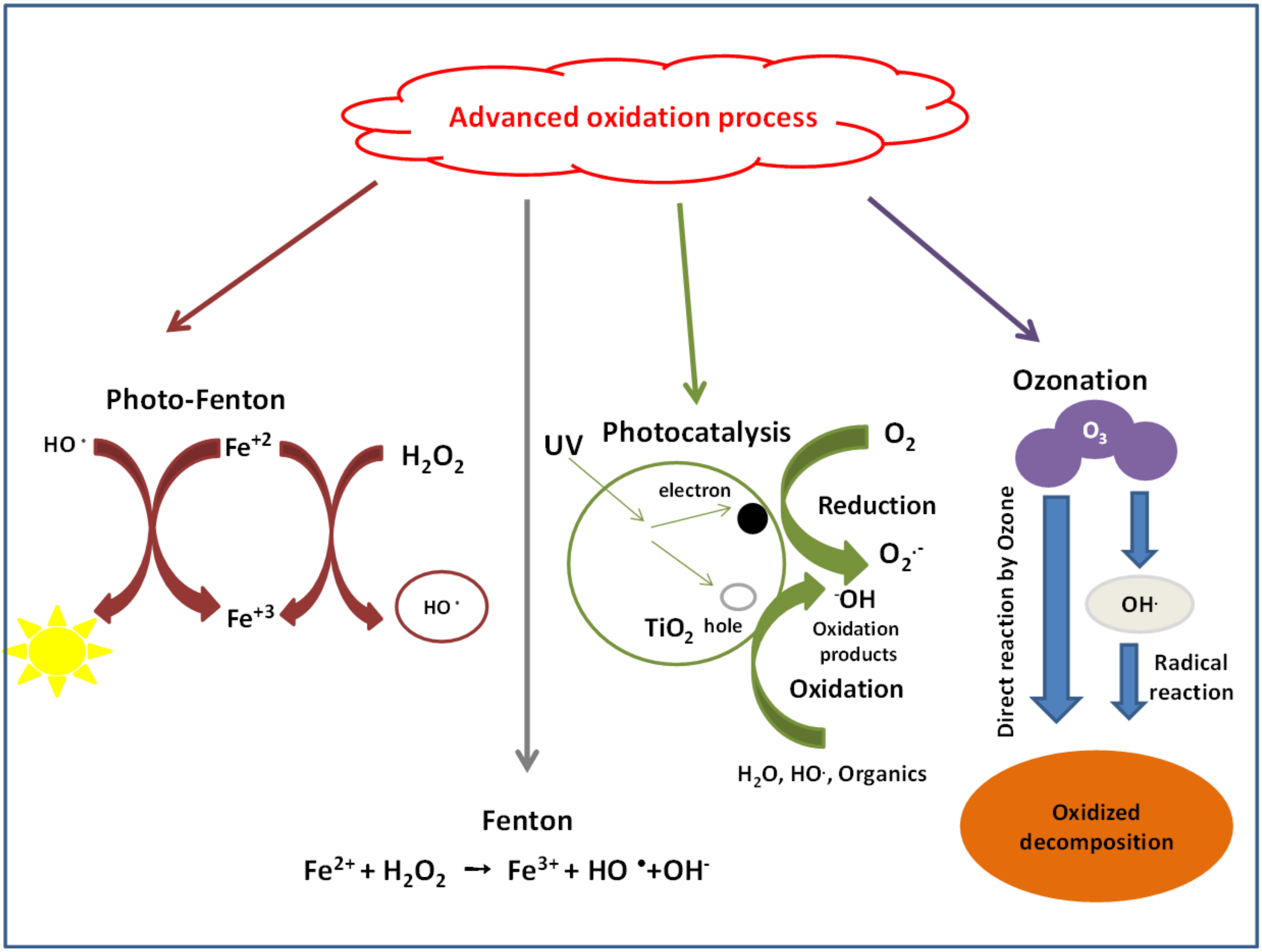

Figure 4. Illustrates the techniques employed in advanced oxidation process.

The removal effectiveness of seven drugs; ketoprofen, prednisone, fenofibrate, fluconazole, betamethasone, loratadine, and 17 α-ethinyl estradiol was assessed in a hybrid system (EGSB-MBR) that combined an anaerobic expanded granular sludge bed (EGSB) reactor with an ultrafiltration membrane. The elimination efficiencies of pharmaceuticals were increased by this integrated system (>84%). Anaerobic biodegradability tests and bioreactor results highlighted the pathways responsible for each drug's elimination (Faria et al., 2020).

2.5 Advanced oxidation process

Advanced oxidation processes have demonstrated considerable promise in removing resistant pharmaceuticals that are difficult for traditional water treatment techniques to break down from wastewater (Orimolade et al., 2021). A viable option for the full breakdown of these chemicals at trace levels has been made possible by the advent of advanced oxidation processes (Capodaglio et al., 2018). The effectiveness of this technique is typically based on the use of auxiliary energy sources, such as ultraviolet-visible (UV-Vis) radiation, electronic current, g-radiation, and ultrasound, as well as highly reactive oxygen or free radical species, such as ozone (O_3), hydrogen peroxide (H_2O_2), and $^{\bullet}OH$, in the mechanisms that result in the degradation of target pollutants into by-products and, eventually, nontoxic end products (Taoufik et al., 2021b). Highly reactive hydroxyl radicals are produced locally. The reactive species created in the process produce the strongest oxidation that is useful in treating water. They can oxidize practically all pollutants existing in an aqueous solution due to their robust nature (Massima Mouele et al. 2021). In spite of the fact that various chemicals oxidize at varying rates, pollutants in water are quickly and effectively broken down and converted into smaller molecules through a non-selective reaction with the hydroxyl radical ($^{\bullet}OH$) (Issaka et al., 2022). This technology has demonstrated efficacy in the decontamination of municipal wastewater, industrial wastewater, textile wastewater, and pharmaceutical wastewater because of its remarkable effectiveness, simplicity of use, safety, and environmental compatibility (Elkacmi & Bennajah, 2019). Various technologies can be used (Taoufik et al., 2021b), including the Fenton process, electrochemical oxidation, heterogeneous photocatalysis, ozonation, and a mixture of several of them (e.g., electro-Fenton and photo-Fenton processes), as illustrated in Figure 4.

2.5.1 Electrochemical treatment

One intriguing solution to reducing pharmaceutical pollution has been thought of is electrochemical oxidation techniques that produce reactive species using electricity instead of chemicals (Taoufik et al., 2021b). The two main groups of this technology are those based on the electrochemical synthesis of oxidants and those based on the direct exchange of electrons between the electrodes and the pollutants. Due to the significant limitations caused by the transfer of contaminants to the electrodes' surface, the first is not appropriate for treating low-concentrated species. On the surface of the anode or cathode of the second kind, oxidants are created from precursors already present in the electrolyte, including hydroxyl radicals, sulfate or peroxide, and ozone radicals. Among many others, these oxidants either directly break down the pollutants or, alternatively, can be activated by interaction with ultrasound, UV radiation, or even other oxidants, enhancing the degradation attained during the therapy (Souza et al., 2022).

The electrochemical oxidation of the pharmaceutical chemicals Iopromide, sulfamethoxazole, 17-alpha-ethinylestradiol, and diclofenac in simulated wastewater and actual hospital effluent wastewater is evaluated. The effects of the electrochemical cell's flow rate, applied current and initial compound concentration were measured. Iopromide decomposition happened much more slowly. The flow rate in the electrochemical cell affected the breakdown rates of 17-alpha-ethinylestradiol and diclofenac. A biological hospital wastewater treatment plant's effluent was successfully cleaned of pharmaceutical components using the boron-doped diamond electrochemical oxidation method (Loos et al., 2018).

2.5.2 Photolysis

The principle behind photolytic methods for the destruction of pollutants dissolved in water is to give chemical compounds energy in the form of radiation, which is then absorbed by various molecules to achieve excited states for the duration required to undergo multiple chemical reactions. The energy needed to excite electrons and create free radicals, which undergo a series of chain reactions to produce the reaction products, is provided by photons, quantized units of radiant energy that molecules absorb. These free radicals can be produced through homolysis of weak bonds, electronic transfer from the excited state of the organic molecule to molecular oxygen, which produces the superoxide radical (O_2), or through the use of other chemical reagents like ozone or hydrogen peroxide, which results in the production of hydroxyl radicals. Due to their photons' higher energy, these photolytic techniques employ UV radiation (Cuerda-Correa et al., 2020).

Hora et al. (2019) confirmed that the degradation of specific pharmaceutical compounds of different therapeutic classes (atenolol, carbamazepine, fluoxetine, and trimethoprim) during medium-pressure UV irradiation (at $\lambda \geq 220$ or ≥ 280 nm) increased with the presence of nitrite in buffer and wastewater matrices due to the production of hydroxyl radicals ($^{\bullet}OH$). In a wastewater matrix that was irradiated at $\lambda \geq 280$ for 120 minutes, the overall pharmaceutical removals were 47% for trimethoprim, 50% for carbamazepine, 60% for atenolol, and 57% for fluoxetine at fluences of 58.6 mEi m^{-2} (2033.1 mJ cm^{-2}). Removals for trimethoprim, carbamazepine, atenolol, and fluoxetine at 634.7 mEi m^{-2} (23 969.2 mJ cm^{-2}) after 60 minutes of radiation were 52, 56, 69, and 90%, respectively. The reaction with $^{\bullet}OH$ was responsible for 78–90% of the drug elimination at 280 nm. Even though direct photolysis helps remove target compounds when exposed to light at $\lambda \geq 220$, the majority of the light was absorbed by the wastewater and buffer matrices, and interaction with

•OH was responsible for between 70 and 93% of the pharmaceutical removal.

2.5.3 Fenton and photo-Fenton

Ultrasound Fenton and photo-Fenton are two other advanced oxidation processes that have grown in interest. Both methods can potentially be effective ways to degrade various medications. The Fenton process (Fe^{2+}/H_2O_2/dark) relies on the oxidation of iron salts Fe^{2+} to Fe^{3+} and the production of hydroxyl radicals (HO) in an acidic media. The Fenton theory is based on the powerful oxidizing power of hydroxyl radicals. In this context, recent years have seen the invention and development of Fenton-type techniques like photo-Fenton oxidation and electro-Fenton. Free hydroxyl radicals can form spontaneously and without the need for light when the Fe^{2+} catalyst is renewed (Taoufik et al., 2021b).

The antineoplastic medication 5-fluorouracil's mineralization and degradation were examined using photo-Fenton and Fenton-like mechanisms (5-FU). The impacts of several operating factors, including the concentrations of 5-FU, Fe^{3+}, and oxidant, on the treatment efficiency, were examined to optimize photo-Fenton treatment under simulated solar light radiation. The technique of simulated solar light radiation/$[Fe(C_2O_4)_3]^3$ was shown to be the most effective based on the results, since 5-FU degraded more quickly and higher mineralization percentages were attained. Through the processes of defluorination, hydroxylation, and ring opening, which involved the attack of hydroxyl radicals on the C-F bond and aromatic ring, 5-FU was transformed. After treatment, all detected intermediates were quickly broken down within 20 to 60 minutes. The toxicity testing discovered that v. fischeri was only moderately harmful to the treated samples and 5-FU (Koltsakidou et al., 2017).

2.5.4 Ozonation techniques

Removing organic contaminants using ozonation techniques in wastewater treatment has attracted significant concern, particularly in surface samples taken from various water channels and water treatment facilities. Fifteen of the chosen 21 organic contaminants were found to be less than 20 mg/L, a comparatively low concentration. The addition of catalysts to catalytic ozonation is anticipated to enhance the breakdown of molecular ozone and produce highly active free radicals that promote the efficient mineralization of organic materials (W. Yang & Wu, 2019). To increase the effectiveness of pollution removal, various metals, metal oxides, activated carbon, and minerals are used as catalysts. Additionally, the addition of catalysts improves ozone usage efficiency (Issaka et al., 2022). Ibuprofen and Gemfibrozil were removed using ozonation and an improved oxidation procedure using ozone/hydrogen peroxide (O_3/H_2O_2) (AOP). With an ozone dosage of 1.5 mg/L and a ratio of 1:0.25 in the O_3/H_2O_2 process, it was possible to completely remove the gemfibrozil and a maximum of 80% of the drug ibuprofen. The elimination effectiveness of these two chemicals was insignificant for both processes from 25 to 40 ^{0}C. Gemfibrozil removal was likewise shown to be unaffected by pH values of 6 to 9, but ibuprofen removal by ozonation at pH 6 was less effective than at higher pH levels (Farzaneh et al., 2020).

2.6 Flocculation and coagulation

Coagulation-flocculation can be used as a beginning step in the elimination of pharmaceuticals. Coagulation-flocculation is a process in which surface charges of dispersed particles are reduced or neutralized by coagulants like metal salts (such as calcium chloride and aluminum sulphate), polymers (such as polymeric aluminum sulphate, polyaluminum chloride, and polyaluminum chloride sludge), or other naturally occurring materials, and flocculants like polymers aggregate to form large particles. However, because most pharmaceuticals are originally organic, coagulants cannot absorb them. From the overall contaminant load, only 3% of medicines may be removed through coagulation. Furthermore, this method is inefficient since it produces more sludge, necessitating more sludge storage and transportation, and requiring more workers to check the pH or other characteristics of the effluent (Yasmin, 2021).

The removal of trace pharmaceuticals from surface water was studied using a potential flocculant like hydrophobically modified chitosan (HC). When alum and HC (alum+HC) were administered sequentially for the treatment of five different water types (three synthetic water and samples of two actual waters from the Yangtze river) and the Thames river had five representative pharmaceuticals (initial concentration: 100 ng/L), the flocculation performance was evaluated. Comparing alum+HC to a typical coagulant or flocculant, all five medicines were removed from all fluids with astoundingly better removal efficiency (highest removal efficiency: 73%-95%) (alum or polyacrylamide, respectively). Compared to utilizing HC alone, alum+HC also produced a greater clearance efficiency of pharmaceuticals with almost half the HC dosage, increasing the alum+HC dosing system's cost (Wang et al., 2022).

2.7 Hybrid technologies

The simultaneous employment of two or more treatment methods in a single process for pharmaceutical wastewater treatment has resulted from the necessity to develop new treatment technology to compensate for the conventional treatment methods and the flaws observed in a few recent technologies. For example, a hybrid treatment method that combines photocatalysis and biodegradation has demonstrated the removal of pharmaceuticals with bioresistance, such as antibiotics, using photocatalysis and vice versa. After treatment, a hybrid approach can provide pharmaceutical wastewater effluent of higher quality (Yu et al., 2020).

Flutamide, a commonly used anti-cancer medication, raises serious environmental concerns. This type of micropollutant can be effectively removed using the sun photo Fenton (SPF) technique. 20% primary degradation was accomplished with a single addition of 5 mg/L of Fe^{2+} and 50 mg/L of H_2O_2, however, only 3.05% mineralization was achieved. With three injections of 5 mg/L Fe^{2+} and a starting H_2O_2 concentration of 150 mg/L, 58% primary degradation and 12.07% mineralization were accomplished. In order to remove residual flutamide, the solar photo Fenton method was further integrated with adsorption onto avocado seed-activated carbon. They may be removed most effectively under the ideal circumstances of 14 mg of avocado seed-activated carbon and a 40-minute contact period. The adsorption procedure completely eliminated any remaining flutamide as well as the transformation products (Della-Flora et al., 2020).

Guo et al. (2015) compared the effectiveness of removing amoxicillin using the Fenton process, a single-activated sludge method, and a combination Fenton-activated sludge system. The ideal Fenton process parameters for the antibiotic at 1 g/L were 40 °C, 6 mL H_2O_2 solution, and 4 mL $FeSO_4.7H_2O$ solution (20.43 g/L). The clearance rate of amoxicillin reached up to 80% in 70 minutes under ideal circumstances. Additionally, the ability of the activated sludge method to remove contaminants was constrained by the effect of amoxicillin on microorganisms. 69.04–88.79% of the amoxicillin was eliminated when the concentration was less than 350 mg/L. When the concentration rose to 650 mg/L, the activated sludge could not eliminate the antibiotic. However, amoxicillin was partially processed by the Fenton process before being destroyed by the same activated sludge. In light of this, the combined system had two steps: step (i) saw an 80% degradation of amoxicillin, and the less expensive biological treatment entirely eliminated step (II). The result demonstrated that the Fenton process enhanced the combined system's removal capacity for the succeeding activated sludge process compared to the individual activated sludge process.

3. Comparison of existing techniques for removal of pharmaceutical compounds

Each technique for treating wastewater has its benefits and drawbacks. Adsorption is quite effective at removing numerous pharmaceuticals from pharmaceutical effluent. Although it still has certain difficulties, such as its low selectivity as an adsorbent. On the other hand, biological treatments are preferable, as they can cause complete destruction. However, it considers a slow process, and most of its techniques have been expensive (Saravanan et al., 2021). Membrane technology has good chemical and mechanical stabilities, although it may encounter membrane fouling issues. Advanced oxidation processes could break down most organic substances into harmless or biodegradable chemicals, but this process may not be economical (Khoo et al., 2022). Furthermore, Table 3 lists the benefits and drawbacks of several methods for removing pharmaceutical substances.

Table 3. the advantages and disadvantages of several methods utilized to remove pharmaceuticals from water/wastewater.

Treatment method	Advantages	Disadvantages
Adsorption	Simple design and operation, there is little to no by-product generation. (Moran 2015), inexpensive, superior effectiveness, non-toxic, and regenerative (Saravanan et al. 2021)	Its efficacy is influenced by a variety of variables. (Moran 2015), low selectivity of the adsorbent and issues with disposal (Saravanan et al. 2021)
Biological treatment methods	Pharmaceuticals can be removed more effectively when used in conjunction with other treatment methods (Pajares et al. 2019), simple process, restriction of odor production, reduces pathogens and lipids, it is possible to use a larger variety of microorganism types for processing, generation of renewable energy and low toxicity (Saravanan et al. 2021)	poor pharmaceutical elimination when compared to alternative treatment methods (Pajares et al. 2019), high cost and issues with maintenance (Saravanan et al. 2021)
Membrane separation process	Excellent mechanical and chemical stability (Khoo et al. 2021b), suitable for treating saline water and wastewater treatment plant influents (Khoo et al. 2020), it applies to various medications and has low energy requirements compared to other methods, and uses nontoxic chemicals. (Moreira et al. 2021)	difficulty with membrane disposal following use (Khoo et al. 2021a) the durability and toughness of the chosen membrane affect the removal efficiency. Membranes are prone to clogging, have a wide pH range, and can be damaged by pressure changes. (Moreira et al. 2021)
Membrane bioreactor (MBR)	More excellent loading rates per volume, less sludge generation (Iorhemen et al. 2016),and a high degree of bacterial and viral removal efficiency (Kowalik et al. 2021)	Increased energy costs and concerns with fouling (Iorhemen et al. 2016)
Advanced oxidation process	Widespread (Segura et al. 2021), might convert the vast majority of organic molecules into safe or biodegradable products. (Zhou et al. 2018), rapid and highly reactive process (Cuerda-Correa et al. 2020)	semiconductors' wide bandgap (Segura et al. 2021), It is not economical to mineralize dangerous and resistant substances (Vincenzo Naddeo 2013). Furthermore, the generation of by-products (Vieira et al. 2020) and demand for further treatment for any remaining oxidant (Wang et al. 2019)
Flocculation and Coagulation	Used to remove tiny particles and get rid of metals, color, and turbidity (Saravanan et al. 2021)	Multiple steps in the process; toxic if misused, a lot of sludge is produced, and a high operational cost(Saravanan et al. 2021)
Hybrid technology	Hybrid technology has been shown to be more effective than single methods. (Gupta et al. 2021).	For various types of wastewater samples, pretreatment can be necessary, and integrated processes necessitate additional treatment units, which increases energy usage. (Gupta et al. 2021).

Conclusions

This review explains many techniques used recently in treating water sources that may contain significant quantities of medicines used daily, whether by humans or animals. Some researchers used the adsorption method, a powerful technique for eliminating pharmaceutical wastewater, while others resorted to biological processes that provide various microenvironments to remove such pollutants. On the other hand, a large group of studies headed to membrane technology based on a selective layer's size exclusion, which only allows specific components to get through while blocking undesirable constituents. An advanced oxidation process is also used, which is an effective method for completely removing these compounds at trace amounts. The idea of hybrid techniques, which depend on integrating more than one of the techniques mentioned above to increase the efficiency of the removal process, was also clarified.

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