REMOVAL OF REFRACTORY POLLUTANTS FROM WASTEWATER TREATMENT PLANTS

EDITED BY

Maulin P. Shah

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Preface

This book describes the state-of-the-art and emerging technologies in environmental bioremediation and reviews their various possible uses together with their related issues and implications. Considering the number of problems that define and concretize the field of environmental microbiology or bioremediation, the role of several bioprocesses and biosystems for environmental protection, control, and health based on the utilization of living organisms are analyzed.

The book aims to provide a comprehensive review of advanced emerging technologies with environmental approaches for wastewater treatment, heavy metal removal, pesticide degradation, dye removal, waste management, microbial transformation of environmental contaminants, and more. With advancements in the area of environmental bioremediation, researchers are looking for new opportunities to improve environmental quality standards. Recent technologies have given impetus to the possibility of using renewable raw materials as potential sources of energy. Cost-effectivet and eco-friendly technologies for producing high quality products and efficient ways to recycle waste to minimize environmental pollution is the need of hour. The use of bioremediation technologies through microbial communities presents another viable option to remediate environmental pollutants, such as heavy metals, pesticides, and dyes.

Since physico-chemical technologies employed in the past have many potential drawbacks, including higher costs and lower sustainability, there is a need to develop efficient biotechnological alternatives to overcome the increasing levels of environmental pollution. Hence, there is a need for environmentally-friendly technologies that can reduce pollutants which have hazardous effects on humans and the surrounding environment.

Environmental remediation, and pollution prevention, detection, and monitoring are evaluated considering the results achieved, as well as the perspectives in the development of biotechnology. Various relevant topics have been chosen to illustrate each of the main areas of environmental biotechnology – wastewater treatment, soil treatment, solid waste treatment, and waste gas treatment – dealing with both the microbiological and process engineering aspects. The distinct role of emerging technologies in environmental bioremediation in the future is emphasized, considering the opportunities to present new solutions and directions in the remediation of contaminated environments, minimizing future waste release, and creating pollution prevention alternatives. To take advantage of these opportunities, innovative strategies, which advance the use of molecular biological methods and genetic engineering technology, are examined. These methods would improve our understanding of existing biological processes in order to increase their efficiency, productivity, and flexibility. Examples of the development and implementation of such strategies are included. Also, the contribution of environmental biotechnology in creating a more sustainable society is revealed.

Editor

Dr. Maulin P. Shah has been an active researcher and scientific writer in the field of microbiology for over 20 years. He received a BSc degree (1999) in Microbiology from Gujarat University, Godhra (Gujarat), India. He also earned his PhD (2005) in Environmental Microbiology from Sardar Patel University, Vallabh Vidyanagar (Gujarat), India. His research interests include biological wastewater treatment; environmental microbiology; and the biodegradation, bioremediation, and phytoremediation of environmental pollutants from industrial wastewaters. He has published more than 240 research papers in several reputable national and international journals on various aspects of the microbial biodegradation and bioremediation of environmental pollutants. He is the editor of 65 books of international repute.

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1 Removal of Pharmaceuticals From Wastewater Using Nanomaterials

Bhumika Roy and Arpita Roy

CONTENTS

1.1 INTRODUCTION

Environmental pollution is one of the biggest challenges our planet has faced in the present century. The increasing world population along with the development of socio-economic, scientific, and technological advancements have directly or inadvertently resulted in ecological deterioration. The presence of pharmaceutical waste in wastewater is a growing threat to the aquatic ecosystem as well as land animals, including human beings. The presence of these contaminants in drinking water is a growing concern; however, proper research about the future impact on the environment is lacking. As a result, these pollutants enter our food chain. Pharmaceuticals are introduced into the environment through various sources like pharmaceutical manufacturing industries, wastewater treatment plants, veterinary use, and leakage in underground sewage systems. Pharmaceutical waste from pharmaceutical manufacturing industries enters the aquatic system due to a lack of properly designed wastewater treatment plants which leads to improper removal of these wastes and finally causes pollution in the aquatic ecosystem (Murray *et al.*, 2010). Animal excreta and its use as manure in agricultural fields introduce these pollutants into the environment. According to available literature, the highest concentration of pharmaceutical waste was found in industrial effluent

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followed by hospital and wastewater treatment effluents with surface water, groundwater, and drinking water showing least concentrations (Patel *et al.*, 2019). Figure 1.1 represents how pharmaceutical waste is introduced into the environment.

A global critical evaluation stated that 203 different types of pharmaceutical waste were found in the environment in 41 countries. This data is, however, still incomplete as only a few countries were included in the study and, in some cases, contributors did not have enough resources to cover the true impact of pharmaceutical waste (Patel *et al.*, 2019). Factors such as demography, accessibility of health facilities, size of the manufacturing sector, sewage treatment systems, and effectiveness of regulation guidelines help in determining the ecological footprint of pharmaceuticals. The introduction of pharmaceutical waste into the environment is an important issue since these compounds are synthesized to bind with the receptors that are present in the body. In such a case, consumption by animals that they are not intended for can have grave effects and prove to be toxic to numerous organisms. Various types of pharmaceutical waste are listed in Table 1.1 with their intended functions, which could prove to be toxic to microorganisms. Apart from this, antibiotic resistance has also been observed in bacteria. Observations like alteration in gene expression, abnormal enzymatic activities, and growth deformities in fish, rats, and frogs have made it even more necessary to combat this source of waste in water.

Conventional wastewater treatment systems like activated sludge and biological trickling filters are not adequate enough to eliminate the wide variety of emerging contaminants and thus they remain soluble in the effluent. Various studies have reported the unsuccessful use of physicochemical treatments including coagulation, flocculation, or lime softening for removing pharmaceutical waste. Techniques such as ozonation and photolysis have some shortcomings as well in the form of being expensive, having a short lifetime, and being unable to remove micropollutants. Due to this, these methods need to be improved or replaced in order to successfully remove pollutants in a cost effective and reliable manner. Nanotechnology presents an efficient and advanced treatment process for the successful remediation of persistent water pollutants. Nanomaterials possess promising results in the adsorption of pollutants. Therefore, in this chapter, the application of nanotechnology over conventional methods for the remediation of pharmaceutical waste from wastewater will be discussed.

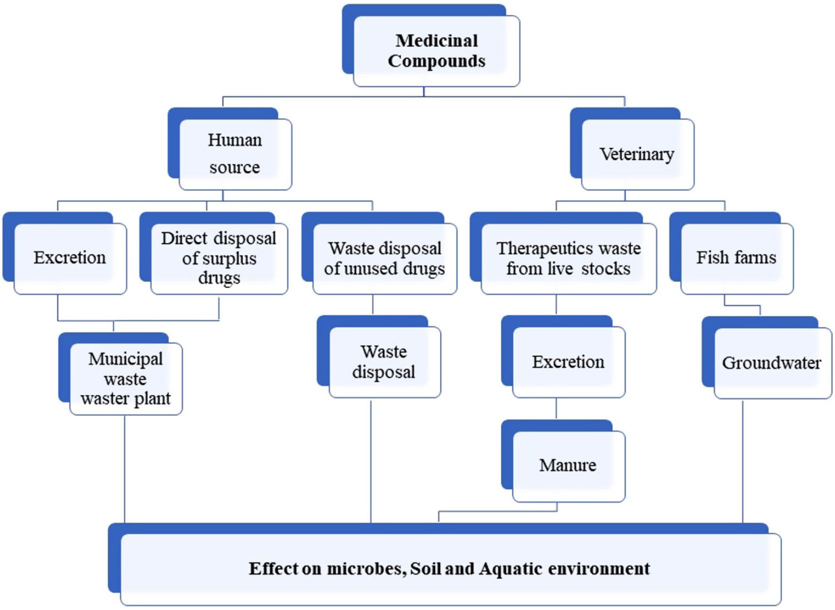

FIGURE 1.1 Entry of pharmaceutical waste into the environment.

TABLE 1.1

Types of Pharmaceutical Wastes, Their Function as Drugs, and Their Use in Specific Treatments

Type of Waste	Function	Use	Example
β-blockers	Block epinephrine and norepinephrine from binding on β receptors on nerves	Treatment of cardiovascular diseases	Atenolol, celiprolol, metoprolol, and propranolol
Hormones	Regulate hormone levels	Treatment of many diseases affected by hormone levels like cancer, growth hormone deficiency, and menopausal symptoms	Estrone, 17-β-estradiol, and 17-α-ethinylestradiol
β-lactam antibiotics	Antimicrobial activity against gram-negative and gram-positive bacteria generally by inhibiting cell wall synthesis	Treating bacterial infections of skin, ear, respiratory and urinary tract	Amoxicillin, cefradine, ceftriaxone, and sultamicillin
Cytostatic drugs	Block mitosis by inhibiting DNA synthesis	Treatment of various types of cancer, autoimmune diseases, and suppress transplant rejections	Cyclophosphamide and ifosfamide
Sulfonamide antibiotics	Inhibit growth and multiplication of bacteria by serving as competitive inhibitors of DHPS in folate synthesis	Treatment of allergies, cough, and antifungal and antimalarial functions	Sulphachlopyridazine, sulfadiazine, sulfadimethoxine, and sulfamerazine
Anticonvulsants and anti-anxiety agents	May block sodium channels or block GABA function	Treatment of mood disorders, depression, and anxiety, as well as to control seizures	Carbamazepine, diazepam, oxazepam, and primidone
Macrolide antibiotics	Inhibit growth of bacteria by blocking 50S ribosomal unit and hindering protein synthesis	Treating common bacterial infections	Azithromycin, clarithromycin, erythromycin, and roxythromycin
Quinolone antibiotics	Hinder the DNA synthesis process	Treating genitourinary infections	Ciprofloxacin, flumequine, norfloxacin and ofloxacin
Analgesic drugs	Affect peripheral and central nervous system in many ways	Treatment of pain and inflammation	Acetaminophen, diclofenac, ibuprofen, and naproxen

1.2 QUANTIFICATION OF PHARMACEUTICAL WASTE

Quantification can be done both before and after the treatment of wastewater to determine remediation effectiveness; however, procuring an accurate estimate is difficult due to low analytic concentration and complex matrix effects. Therefore, performing steps such as sample collection, sample preparation, chromatographic separation, detection, and data analysis is essential.

1.2.1 Sample Collection

A large number of samples are processed to procure reliable results that are as accurate as possible. To further modify the process, the volumes of different samples taken must be in accordance with the concentration of analyte, i.e., samples containing lesser concentrations must be collected in larger volumes. The proper storage of these samples is of utmost importance since errors can lead to the contamination or decomposition of sample analytes. The key players in contamination include microbial activity, unwanted chemical reactions, and exposure to UV radiation. Microbial activity and analyte decomposition can be controlled by adding chemical preservatives or storing in brown amber glass bottles with temperature control (Śliwka-Kaszyńska *et al.*, 2003).

1.2.2 Sample Preparation

Samples are prepared by purifying and concentrating them before analysis because of the confounding matrix effect; some pharmaceuticals can show effects in concentrations as low as 1 ng/L. The chemical and physical properties of analyte make this optimization. It must be performed every time the matrix chemicals change. The steps leading up to chromatographic separation include the adjustment of pH, extraction, and elution.

Solid-phase extraction (known for its multiple analyte extraction from complex mixtures) is employed to remove dissolved pharmaceuticals. The mixture is passed through a column where the pharmaceutical separates from the mobile phase to bind to a solid stationary phase. Conditioning of the column is done with a suitable solvent to make the stationary-phase surface wet, followed by loading of the sample onto the column wherein retention of the target analyte is observed. Further washing of the column is done to remove any persistent impurities and later the target pharmaceuticals are eluted and buffered by pH. The solvent is then prepared for analysis by adjusting the volume.

Chromatographic separation is done to further isolate target pharmaceuticals from matrix chemicals through mass spectrometry detection. MS, MS/MS, electrospray ionization, and atmospheric pressure chemical ionization are some techniques used for detection. Because matrix effects can largely reduce analyte signal intensity in environmental pharmaceutical analysis, detection may be hindered. Raman spectroscopy has also been used for pharmaceutical analysis in drug development, drug production, quality control, studies on stability, and drug metabolites analysis. Other analytical techniques include electrophoresis, electrochemical, flow injection analysis, and titrimetric-related processes (Siddiqui *et al.*, 2017).

The identification and quantification of analytes is done by using calibration curves. In this process, the matrix effect plays a vital role. Therefore, a range of matrix chemicals should be included in calibration standards, which helps in the reduction of any errors related to matrix effects. Interference-free quantification in environmental analysis can be ensured by second-order multivariate calibration algorithms, internal standard (IS)-based calibration, standard addition calibration, and matrix-matched calibration.

1.3 ENVIRONMENTAL AND HEALTH ISSUES OF PHARMACEUTICAL WASTE

The environmental exposure of pharmaceuticals generally occurs through, amongst others, manufacturing units, hospital effluents, and land applications. Wastewater treatment plants are not reliably effective at eliminating these pollutants. Therefore, these pollutants enter the aquatic environment where they directly affect aquatic organisms and can be incorporated into food chains. High concentrations of pharmaceuticals can change the structure of microbial communities and ultimately affects food chains. Long-term exposure to even low concentrations of complex pharmaceuticals can result in an acute and chronic damage, behavioral variations, reproductive damage, inhibition of cell proliferation, and more. Several reports suggested that wastewater effluents exhibit reproductive abnormalities in fishes. Furthermore, fish exposed to trace levels of birth control pharmaceuticals showed dramatic reductions in reproductive success, which suggests impacts on population levels.

1.4 CONVENTIONAL TREATMENT METHODS AND THEIR SHORTCOMINGS

Conventional treatment methods include activated carbon adsorption, membrane filtration, biological processes, UV photocatalysis, and ozonation. These methods have their shortcomings in the form of low efficiency or high cost and thus needed to be replaced. Biological processes, for instance, cannot fully degrade recalcitrant pharmaceutically active compounds (PhACs) (like bezafibrate) and carbamazepine. The treatment of wastewater discharged from pharmaceutical industries poses additional difficulties due to the presence of active pharmaceutical ingredients (APIs), organic solvents, reactants, intermediates, raw materials, and catalysts (Chelliapan *et al.*, 2006). Commonly-used adsorbent-like activated charcoal removes hydrophobic substances and charged PhACs from the aqueous phase with the help of non-specific dispersive interactions for removing non-polar PhACs, and interactions of electrostatically activated charcoal and ionic PhACs to remove polar PhACs. This method is mostly used after biological treatment as a post-treatment measure. The drawbacks of this method are in terms of its effectiveness which is governed by the natural dissolved organic matters in the wastewater matrix and its high total energy demand.

Ozonation is used as a secondary treatment method for removing PhACs from wastewater (Dodd *et al.*, 2009). Shortcomings, such as the consumption of high and uncertain effects of oxidation products, necessitate further development of the method. Catalytic ozonation acts as an alternative to the ozonation process and is one of the most advanced oxidation processes. This method's efficiency for removing organic pollutants makes it better than ozonation; however, it has its shortcomings, including a decline in rate due to the limited adsorption and diffusion of PhACs on catalysts steps.

Membrane filtration removes low molecular weight PhACs (Le-Minh *et al.*, 2010) via an adsorption process. In the steady state, the efficiency of removal depends on membrane characteristics (material, surface morphology, and pore size), solution parameters (such as pH and ionic strength), and physicochemical properties of PhACs (such as pK_a, molecular weight, hydrophobicity, or hydrophilicity) (Le-Minh *et al.*, 2010). Membrane fouling causes restrictions in the engineering application of membrane processes and serves as a challenge for effective operation.

UV radiation in association with photocatalysts shows relatively high efficiency in degrading PhACs in wastewater (Murgolo *et al.*, 2015) and has a low energy cost. Conventional photocatalysis methods come with their own drawbacks, such as low solar light activity, high energy consumption, and low quantum yield efficiency. These drawbacks of conventional methods call for a need for new photocatalysts which can improve the removal efficiency of pharmaceutical wastes.

1.5 NANOPARTICLES IN WASTEWATER TREATMENT

Nanoadsorbents like carbon nanotubes and metal oxides have wide applications in wastewater treatment. Their key features such as high surface area, high dispersion area, microporous structure, and being economically viable make their application stronger (Gupta *et al.*, 2015). Nanoparticles used for adsorption include carbon nanotubes, graphene, magnesium oxide, ferric oxides, manganese oxide, zinc oxide, and titanium oxide (Gupta *et al.*, 2015). The factors affecting the process of adsorption include temperature, dose of adsorbent, pH, and contact time. Nanocatalysts are gaining attention for their properties especially the ones obtained from inorganic materials. The different types of catalysts involved in wastewater treatment include photocatalysts, electrocatalysts, and fenton-based catalysts. These catalysts are used for the chemical oxidation of organic pollutants, especially the ones made from noble metals such as gold, platinum, and palladium (Liu *et al.*, 2013). They are advantageous over conventional methods due to lessened treatment times, target recalcitrant compounds, and the transformation of wastes into valuable by-products. Nano-membranes use nanoparticles for membrane filtration technology and are the most effective wastewater treatment method. It is advantageous to use because it provides effective disinfection, quality water treatment, and is efficient, economical, and simple. One-dimensional nanomaterials consisting of organic and inorganic materials such as nanotubes, nanofibers, and nanoribbons are used for constructing these membranes (Liu *et al.*, 2014).

Nanoadsorbents sometimes lead to secondary pollution due to difficulties in the separation of small particles from the aqueous solution, which affect the bioavailability and mobility of pollutants and cause environmental toxicity. Their reuse and regeneration also pose a challenge. Nanocatalysts act as an advancement to catalytic water treatment techniques such as electrocatalysis, photocatalysis, and fenton catalysis, though there are many drawbacks. Zinc oxide and titanium dioxide used in photocatalysis require UV radiation for their activity which raises serious health risks like skin cancer for workers. Nanocatalysts, like AgBr, when introduced in the solution cannot be recycled for reuse. Electrocatalysts widely require Pt, however, its limited availability, poisoning of intermediates, and high cost act as limiting factors (Zhou *et al.*, 2003). Fenton catalysis requires the maintenance of acidic conditions throughout the process and catalyst material is continuously lost. Nanomembrane manufacturing has a large ecological footprint. A study by Khanna *et al.* (2008) showed that carbon nanofiber contributes 100 times more to toxicity, ozone depletion, and global warming than conventional methods. Another disadvantage is membrane fouling, which occurs when organic compounds interact with hydrophobic membranes. This reduces the quality of water treated, and the life and reliability of the membrane equipment (Gu *et al.*, 2013). This problem can be overcome by using biogenic nanomaterials.

1.6 BIOGENIC NANOMATERIAL

Employing the use of biogenic nanomaterials is advantageous over conventional methods. Conventional wastewater treatment methods are, to a certain extent, detrimental to the environment due to their inability to degrade the pollutants into environmental-friendly end products and passing them off from one phase to another. Apart from this, they require large areas to function, high capital, and maintenance costs. Nanotechnology is therefore used as an alternative technique in the form of nano-based adsorbents, catalysts, and membranes (Diallo and Brinker, 2011). They are mass produced using physicochemical methods; however, toxic waste gets introduced into the environment which affects human health and the environment. For instance, sodium borohydride generates hydrogen diborane which is a highly toxic by-product (Li *et al.*, 2006). Biogenic nanoparticles synthesized from bacteria, algae, fungus, and plants are a new alternative. Their attributes makes them suitable for pharmaceutical waste removal from wastewater, including low cost of production, no toxic waste production, greater surface area, stability due to lipid bilayer structure in some, better physiological solubility and manipulation of size and shape by altering pH, contact time, and substrate availability (Li *et al.*, 2011) (see Figure 1.2).

1.7 SYNTHESIS OF BIOGENIC NANOPARTICLES

The synthesis of nanoparticles via microorganisms can take place either intracellularly or extracellularly. The positively charged metal ions diffuse via electrostatic interactions into the negatively charged cell wall in intracellular processes. The uptake is further facilitated by endocytosis, ion channels, and carrier channels. The enzymes convert the toxic metals to non-toxic nanoparticles. Extracellular processes are achieved via enzymes, including nitrate reductase (from fungus), that convert metal ions into metal nanoparticles. Plants are also an alternative source for biogenic synthesis of nanoparticles (see Figure 1.3).

1.7.1 Bacterial-Facilitated Synthesis

Many bacterial strains employ processes like bioleaching, biomineralization, and bioaccumulation to solubilize metal ions and are used as potential biofactories. Their protective and defensive strategy against metal ions aids metallic nanoparticle synthesis, and the ease of manipulation makes

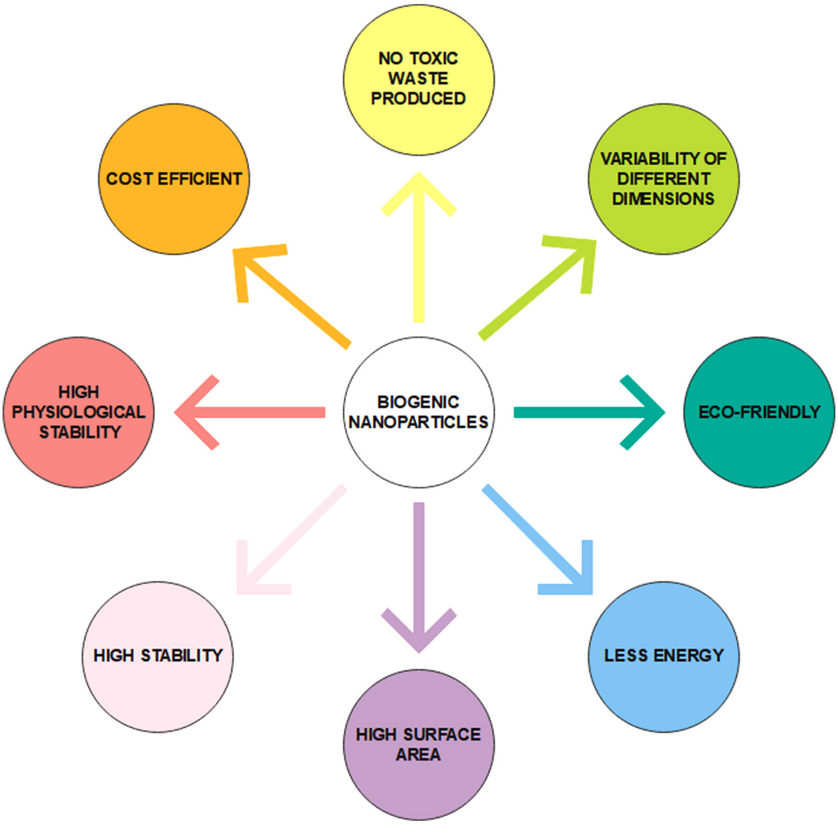

FIGURE 1.2 Advantages of biogenic nanoparticles.

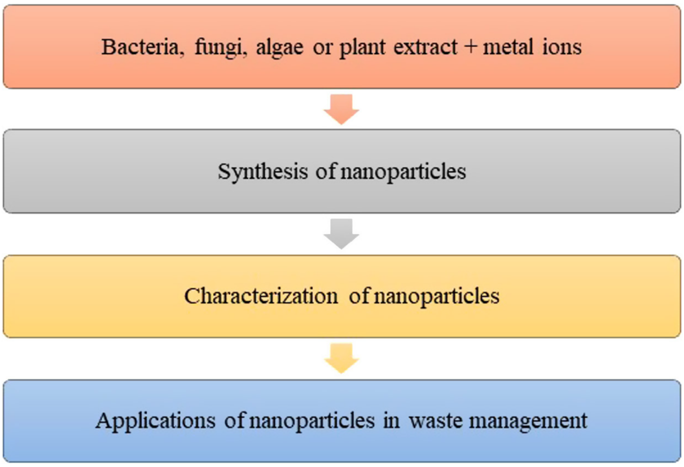

FIGURE 1.3 Biosynthesis of nanoparticles.

them appropriate for both intracellular and extracellular synthesis (Prasad *et al.*, 2016). Examples of bacterial species being investigated for nanoparticle synthesis include gold: *Klebsiella pneumonia* (Malarkodi *et al.*, 2013); iron: *Bacillus subtilis* (Sundaram *et al.*, 2012) and *Klebsiella oxytoca* (Anghel *et al.*, 2012); copper: *Escherichia coli* (Singh *et al.*, 2010) and *Pseudomonas fluorescens* (Shantkriti and Rani, 2014); and silver: *Bacillus cereus* (Sunkar and Nachiyaar, 2012).

1.7.2 ALGAE-FACILITATED SYNTHESIS

The properties of algae promoting its use in nanoparticle synthesis are vast and include high tolerance, metal bioaccumulation ability, easy handling, economic feasibility, and richness of bioactive molecules having amine, carboxyl, and hydroxyl functional groups serve as reducing and capping agents. Examples of algal species utilized for nanoparticle synthesis include gold: *Chlorella vulgaris* (Annamalai and Nallamuthu, 2015); iron: *Sargassum muticum* (Mahdavi *et al.*, 2013); copper: *Bifurcaria bifurcata* (Abboud *et al.*, 2014); and silver: *Cystophora moniliformis* (Prasad *et al.*, 2013).

1.7.3 FUNGUS-FACILITATED SYNTHESIS

Fungi – being diverse, highly metal tolerant, and a good source of extracellular enzymes – are able to accumulate metal ions, and are used for the synthesis of highly stable and economically-viable nanoparticles (Siddiqi and Husen, 2016). Mechanisms of biomineralization and biotransformation are employed to form mycogenic nanoparticles (Das *et al.*, 2012a). Some yeast strains have also been studied for nanoparticle synthesis. Examples of fungus and yeast species being investigated for nanoparticle synthesis include gold: *Cylindrocladium floridanum* (Narayanan *et al.*, 2013); iron: *Fusarium oxysporum* and *Pleurotus sp.* (Mazumdar and Haloi, 2017); and copper: *Aspergillus sp.* (Cuevas *et al.*, 2015).

1.7.4 PLANT-FACILITATED SYNTHESIS

This involves a single-step biosynthesis process and no toxic production, making it an effective alternative for nanoparticle synthesis (Roy and Bharadvaja, 2019). Plants are advantageous to use due to their availability, and easy and safe handling. They are better than bacteria and fungi since they require much less incubation time. Metallic and oxide nanoparticles can be synthesized on an industrial scale by employing plant tissue culture techniques and downstream processes. The effect of the nanoparticle synthesized varies from species to species and depends on their mode of application, size, and concentrations. Research for nanoparticle synthesis using plants is still in the initial stages and extensive work is required to fully understand the physiological, biochemical, and molecular mechanisms of plants. Also, the mode of action of the synthesized nanoparticles needs to be studied and explored. Examples of plant species being investigated for nanoparticle synthesis include gold: *Amaranthus spinosus* (Das *et al.*, 2012b); silver: *Centella asiatica* (Roy and Bharadvaja, 2017) and *Plumbago zeylanica* (Roy and Bharadvaja, 2019); palladium: *soybean* [*Glycine max* (L.)] (Petla *et al.*, 2012).

1.8 REMOVAL OF PHARMACEUTICAL WASTE USING BIOGENIC NANOPARTICLES

Biogenic manganese oxide (BioMnOx) and bio-palladium (Bio-Pd) nanoparticles have been synthesized using *Pseudomonas putida* MnB6 strains and *Shewanella oneidensis*, respectively, by Meerburg *et al.* (2012) to remove several recalcitrant pharmaceutical pollutants from sewage wastewater via oxidation or reduction techniques. BioMnOx is capable of removing organic micropollutants such as naproxen, diclofenac, clarithromycin, chlorophene, iohexol, ibuprofen, and steroid hormone estrone at ppb level concentrations (Furgal *et al.*, 2014). Bio-Pd can reportedly remove iomeprol, iopromide, and iohexol via catalytic reduction.

Two biocatalysts, bio-platinum (bio-Pt) and bio-palladium (bio-Pd), derived from *Desulfovibrio vulgaris* were found to be acting against ciprofloxacin, 17b-estradiol, and sulfamethoxazole (Martins *et al.*, 2017) with bio-Pt showing a higher catalytical role than bio-Pd. Ibuprofen was also included in this study; however, both bio-Pt and bio-Pd were unsuccessful in removing it. When whole cells of *D. vulgaris* were introduced, they transformed the ibuprofen, and it was deduced that sulfate-reducing bacteria can completely remove it in an anaerobic environment (Kumari *et al.*, 2019).

Diclofenac is an anti-inflammatory drug that is modestly biodegradable and present in very low concentrations in water. When treated via ozonation, it gave rise to mutagenic by-products. Therefore, bio-Pd nanoparticles were improved to test their activity on it. Bio-Pd nanoparticles synthesized from metal-reducing bacteria when doped with zero-valent Au NPs degrade diclofenac from water. The study concluded that the catalytic activity is determined by the mass ratio between Pd and Au with a 50/L ratio resulting in maximum degradation of the drug in 24 hours (De Corte *et al.*, 2012).

HCl was employed for the regeneration of green composite iron nanoparticles by Ali *et al.* (2016). The regenerated NPs showed a consistent removal rate of 85–92% for ibuprofen.

BioMnOx produced from *Desmodesmus sp.* WR1 algae was similar to bacteria derived BioMnOx in degrading organic pollutants via oxidative degradation and was successful against Bisphenol A (BPA), an endocrine disrupter (Wang *et al.*, 2017).

Magnetic chitosan nanoparticles were used for the removal of tetracycline from wastewater. They showed a maximum adsorption capacity of 78.11 mg/g at pH 5 and a temperature of 25°C. Incorporating magnetic material is advantageous as it promotes easy separation of adsorbent (Raeiatbin and Açıkel, 2017).

Camiré *et al.* (2019) in their study showed the use of novel alkali lignin and poly (vinyl alcohol) (AL: PVA) nanofibrous membrane for the adsorption of pharmaceutical waste. The study showed 90% adsorption of fluoxetine which was in parallel with costly ion-exchange resins (75–80 mg/g). The nanofibers follow the pseudo-first order kinetic model and the Sips isotherm model. This helps in deducing that the nature of the adsorbent was physical and adsorption was taking place at multiple sites (Camiré *et al.*, 2019).

Green-synthesized copper nanoadsorbents were also evaluated for their activity against pharmaceutical waste present in water by Husein *et al.* (2019). Their activity was checked against three pharmaceutical compounds: diclofenac, ibuprofen, and naproxen, and the adsorption capacities were 36, 33.9, 33.9 mg/g, respectively. The nature of the sorption process was stated to be spontaneous, endothermic, and physical.

Silica-based nanoparticles obtained from rice husk were used by Nassar *et al.* (2019) for studying the adsorptive removal of ciprofloxacin from polluted water. The adsorption occurred at pH 7 and the adsorptive capacity of the nanoparticles was 24.1 mg/g. These nanoparticles are cheap to synthesize, are non-toxic to the environment, and are biocompatible. Their advantages such as high surface area, high porosity, and mechanical resistance make their candidature as green nanoparticles strong (Nassar *et al.,* 2019).

Green-synthesized zinc oxide nanoparticles were coated on a ceramic ultrafiltration membrane and were used to remove atenolol and ibuprofen drugs from a synthetic solution, as evaluated by Bhattacharya *et al.* (2020). The membrane's hydrophilicity was enhanced by the coating of zinc oxide nanoparticles and the study concluded that effective removal of the pharmaceutical compounds was observed and can be tested in pharmaceutical wastewater.

1.9 CONCLUSION

A nanotechnology-based approach for the degradation of pharmaceutical waste has gained popularity due to its various advantages. However, toxic and volatile substances being used for the same pose a major concern to manufacturing units. To overcome this challenge, greener ways of producing nanomaterials were explored by various research groups and the best alternative deduced was production with the help of biomolecules from organisms. Nanoparticles from bacteria, algae, fungi, and plants are non-toxic, sustainable, and have a low cost, thus they are gaining popularity. They successfully convert toxic material into non-toxic forms and hence their activity on all types of waste material in water is being explored. Some challenges of biogenic nanoparticles, such as stability and size control, still exist and need to be studied. Research focusing on species that have not been studied yet is underway.

LIST OF ABBREVIATIONS

Ng	Nanogram
L	Liter
MS	Mass spectrometry
MS/MS	Tandem mass spectrometry
PhAC	Pharmaceutically active compound
API	Active pharmaceutical ingredient
Ppb	Parts per billion
Pd	Palladium
Au	Gold
HCl	Hydrochloric acid
NP	Nanoparticle
Mg	Milligram
g	Gram
°C	Celsius

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