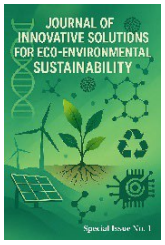
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Review Article

Recent Advancement in Bioremediation of Pollutants Using Green Synthesized Nanoparticles

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ARTICLE INFO

Keywords:
nanoparticles;
heavy metals;
bioremediation;
wastewater;
green synthesis

ABSTRACT

Advancements in agricultural approaches, increased population and industrial development have created the problem of environmental pollution. This pollution causes loss of biodiversity and several serious health diseases. Therefore, several approaches have been utilized to remove environmental pollution. However, most of these methods have drawbacks. Nanotechnology is an extensively growing field that has various applications in decreasing the adverse effects of toxic pollutants. Nanotechnology is a cost-effective and environmentally friendly method to solve the problem of environmental pollution. Nanoparticles (NPs) have been synthesized by traditional chemical and physical techniques. However, these traditional methods have several drawbacks, such as being expensive and producing toxic substances. The natural bioremediation method removes pollutants at a slow rate, and thus the removal process takes more time. Thus, it is important to introduce alternate techniques, which is nanobioremediation. Nanobioremediation includes the role of both microorganisms and NPs; as a result, the removal of pollutants by this method occurs within a shorter time. Biogenic resources such as bacteria, fungi, viruses, algae, yeast and plants are used to synthesize NPs. Biogenic method for the synthesis of NPs is a clean, safe, environmentally gentle, and sustainable method. The objective of the present review is to highlight the application of green synthesized NPs for the removal of pollutants such as dyes and heavy metals from wastewater. However, more research is required to study the toxic effects of green synthesized NPs, reusability and their commercialization on a larger scale. It is expected that the use of green-synthesized NPs for the removal of pollutants will expand in the future.

1. Introduction

Environmental pollution is becoming one of the major problems due to increasing population, urbanization, agricultural advancement and growth of industries. The main environmental contaminants include heavy metals, chemical insecticides, fertilizers, effluents released from industries and sewage. These contaminants are released into rivers and oceans, causing contamination and harmful effects on humans and aquatic animals [1]. The extreme use of synthetic dyes in various industrial activities and the utilization of chemical pesticides in agricultural fields is causing water and soil pollution. The removal of heavy metals and harmful substances from wastewater has become a major worldwide challenge [2, 3, 4]. Thus, conventional methods such as landfill techniques and capping, etc., are used for removing the pollutants from water and soil. These techniques are temporary and they only work for a short time. A more effective method for degrading pollutants than traditional approaches is dechlorination, which can be catalyzed by a base, by UV oxidation, or achieved through high-temperature incineration. These techniques efficiently decrease the amount of several contaminants. However, they have several disadvantages such as being expensive, highly complex and having enhanced exposure to workers. Therefore, to combat this problem, bioremediation was used as an alternative method [5].

Bioremediation is a technique that uses living micro-organisms to remove or neutralize different types of contaminants. These micro-organisms and plants convert hazardous and toxic pollutants into less toxic forms.

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https://doi.org/10.46991/JISEES.2026.02.2.13684
Received 21 September 2025; Received in revised form 17 February 2026; Accepted 31 March 2026; Available online 12 July 2026
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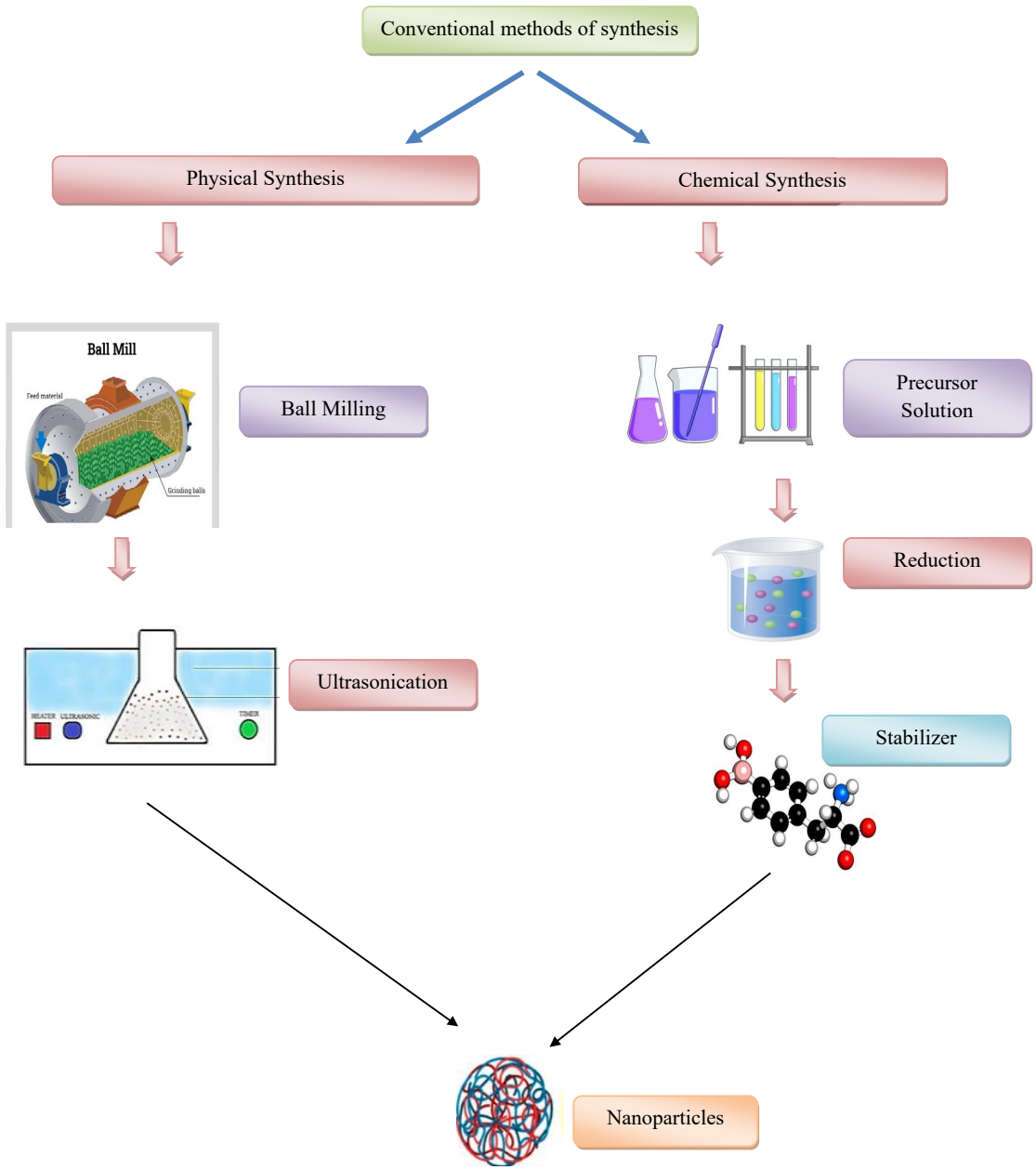


Figure 1. Conventional method for the synthesis of NPs [85]

Traditionally, various in situ and ex situ approaches for bioremediation of hazardous pollutants have been reported. These methods include biocomposting, biopiles, bioaugmentation, biostimulation, bioventing, landfarming and biosparging. However, these techniques have various drawbacks, such as the rate of growth and other technical factors. As a result, scientists have developed the field of nanobiotechnology to remove and degrade environmental pollutants [6].

Nanoparticles have a unique advantage as they contain more properties than their large counterparts [7] extensively. NPs are exceptional absorbents and catalysts, because of their larger specific areas, lower change in temperature, a greater number of related adsorption sites, and unique surface chemistry when compared to other options [8]. NPs that have a higher fraction of surface area, increased surface energy, precise dimensions, and spatial confinements are showing potential benefits in fields such as catalysis, magnetism, electronics, and optics [7]. There are various physical and chemical methods utilized for the synthesis of nanomaterials (Figure 1). Chemical methods involve the use of reduction and stabilizers, whereas physical methods utilize ball milling and ultrasonication for the synthesis of NPs. Although most methods consume high amounts of energy, produce hazardous substances and are less sustainable and costly [9].

NPs are synthesized by either a top-down or a bottom-up technique.

In the top-down method, large structures are converted into small ones with the utilization of physical methods. The bottom-up method uses small atoms or molecules to synthesize NPs through self-assembly. Biological methods have gained more attention due to being environmentally friendly, consuming less energy and not producing any hazardous substances [10–13]. Micro-organisms in a polluted environment adjust and change themselves for the degradation of hazardous compounds having enough catabolic activity toward the contaminated environment [14]. Green synthesis of metal NPs has various benefits because of their physico-chemical characteristics and broad applications in the biotechnology field [15]. The main objective of the present review is to provide knowledge on recent developments in the bioremediation of pollutants using green synthesized NPs.

2. Green Synthesis of NPs

Green synthesis is an environmentally friendly method and alternative technique to the chemicalmethod for the synthesis of NPs. The term "green synthesis" means the processes that prepared NPs with the less amount of energy consumption while preventing hazadrous byproducts via cleanup, control, remediation, regulation and cleanup [16]. Bacteria are ubiquitous in the environment, occurring in soil, water and air. They can be easily grown under laboratory conditions. These characteristics make bacteria potential candidates for the synthesis of NPs. Bacteria is capable of manufacturing NPs both extracellularly or intracellularly [17]. Several NPs were synthesized from bacteria. Gold (Au) NPs were prepared from the marine bacteria *Marinobacter algicola* and *Paracoccus haeundaensis* [18,19]. Similarly, fungi are also they easily cultured in high amount and rapidly synthesized NPs [20]. Various fungi were utilized for the synthesis of NPs. *Aspergillus niger* was utilized for the synthesis of CuNPs [21]. Algae were also used for the synthesis of various NPs. *Sargassum plagiophyllum* mediated the synthesis of AgCl NPs [22]. Several NPs were prepared from different forms of yeast. Ag and SeNPs were prepared by utilizing the extract of *Saccharomyces cerevisiae* [23–26]. Actinomycetes contain various secondary metabolites that can facilitate the synthesis of various NPs [27,28]. Various NPs were prepared from distinct types of actinomycetes. *Gordonia amarae* mediated the synthesis of AuNPs [29].

3. Methods for Green Synthesis of NPs

The synthesis of NPs using microbes and plants is fast, feasible, less expensive and eco-friendly [30]. The working principle of green synthesis of NPs includes bio-precipitation and reduction methods. These methods are carried out by amino acids, peptides, other bioactive compounds and polyphenols, which are released from living organisms [31]. These compounds work as a stabilizing agent. These compounds also prevent the mass formation of synthesized NPs [32]. Biosynthetic methods for NPs can be classified into three types intracellular, extracellular type of biosynthesis (according to the location where NPs are synthesized) and utilization of specific biochemicals [33]. Out of both, the extracellular synthesis of NPs is more preferred due to simple down-stream processing and fast scale-up processing.The basic method for the synthesis of NPs involves the utilization of the intracellular and an extracellular extract of microorganisms and plants. Then the extract is mixed with metal salt solution at room temperature. The process is completed within few minutes. Figure 2 shows a picture of various microorganisms and plants used forthe biosynthesis of NPs. The nature of the living extract, concentration of extract, the amount of the metal salt, the pH and temperature affect the speed of production of the NPs, their yield and other features [34].

4. Mechanism of Bioremediation

4.1 Adsorption Technique

Adsorption techniques are a commonly used method for treating polluted liquids. In this technique, metal ions are deposited on adsorbent surfaces to form a layer that consists of the ions and other contaminants. Compared to traditional methods for the removal of metal, adsorption has various benefits such as being cheaper, capacity for regeneration, less consumption of energy and chemicals, no production of sludge, and the ability for recovery of metal [35]. The performance of adsorption is affected by various factors involving as the quantity of adsorbent utilized, pH, the relations between the adsorbate and adsorbent, surface area of adsorbent, time required for contact, particle size, temperature and primary concentration of pollutant [36].

4.2 Photocatalytic Degradation

Photocatalytic degradation is a method that utilizes photocatalysts such as semiconductor materials, titanium dioxide (TiO_2), or zinc oxide (ZnO) to facilitate the degradation of pollutants under the influence of light. Upon irradiation with ultraviolet (UV) or visible light, photocatalysts generate electron–hole pairs, which can initiate oxidation and reduction reactions with surrounding molecules, such as water and dissolved oxygen. The use of photocatalysis, in particular, has gained attention because of its environmentally friendly nature, more efficient, cost-effective and ability to remove hazardous pollutants [37].

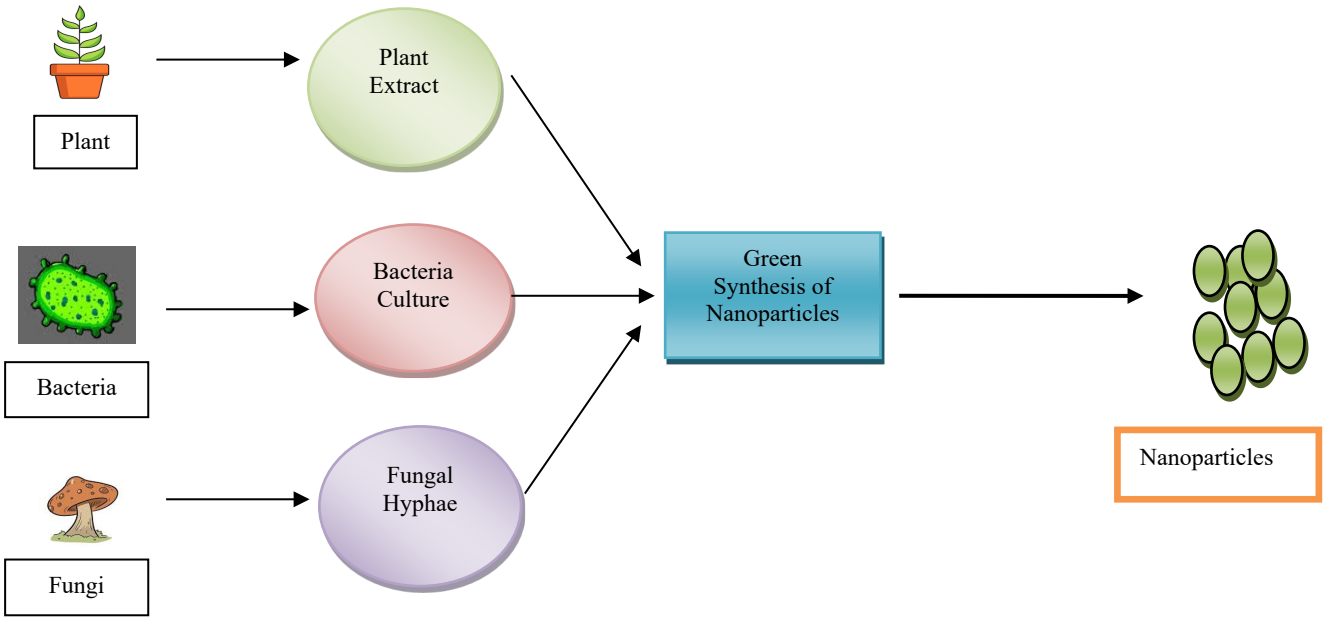


Figure 2. Mechanisms of synthesis of NPs from micro-organisms and plants [9]

5. Application of Algae-Based NPs for Bioremediation

Algae-based NPs have the potential of removing, degrading, or converting contaminants into less toxic or non-toxic forms, thus promoting nanoremediation [38, 39]. In a study, green silver (AgNPs) was synthesized using *Chlorella ellipsoidea* effectively removed the dyes such as methyl orange and methylene blue [40]. Fe_3O_4-NPs were produced using the species of brown macroalgae *Padina pavonica, Colpomenia sinuosa,* and *Petalonia fascia* reduced nitrogen and phosphorus-containing compounds from wastewater [41]. AgNPs and gold (AuNPs) were prepared from algae *Nannochloropsis* sp and *Chlorella vulgaris*. It has been observed that these algal NPs decreased the concentration of lead and zinc in wastewater [42]. *Sargassum latifolium* based AgNPs removed malachite green dye by bioadsorption process [43]. CdS NPs synthesized from blue green algae removed pollutants from wastewater [44]. Algae biofilm based FeONPs removed methylene blue (MB), malachite green (MG), and phenol red (PR) dyes [45]. In another study, *Pterocladia capillacea* based NPs increased bioremediation of toxic dyes [46].

5.1 Application of Bacteria-Based NPs for Bioremediation

AgNPs synthesized from *Macrococcus caseolyticu s*and eliminated Total Petroleum Hydrocarbons in water samples [47]. Magnetite Fe_3O_4 and hematite Fe_2O_3 prepared from *Lyptolyngbyafoveolarum* extract and *Azospirillumbrasilense* and removed ammonia, nitrate and phosphate from wastewater [48]. Platinum (Pt) and palladium (Bio-Pd) NPs synthesized using bacteria *Desulfovibrio vulgaris* removed various antibiotics from wastewater [49].

5.2 Application of Fungi-Based NPs for Bioremediation

It had been observed that fungi *Aspergillus niger* based AgNPs increased the bioremediation of diesel oil contaminated soil and water [50]. *Fusarium oxysporum* based CuO-NPs removed heavy metals and dyes from wastewater of textile industry [51]. *Fusarium oxysporum* based FeNPs had effectively removed various heavy metals (Zn, Cd, Pb, Cr and Ni) from wastewater [52]. Fe_3O_4 synthesized using *Aspergillus elegans* extract degraded dyes in wastewater with high efficiency [53]. In another study it was observed that TiO_2 NPs synthesized from *Aspergillus penicillioides* degraded crystal violet dye by the process of photocatalysis [54]. Similarly, ZnONPs synthesized using *Fusarium proliferatum* degraded anthracene [55]. Fungi based CuONPs and ZnONPs removed pollutants from groundwater [56]. Magnetite NPs (Fe_3O_4) synthesized from the endophyte *Aspergillus flavus* reduced the toxicity of Reactive Black 5 (RB5) dye [57]. Iron oxide NPs synthesized from *Aspergillus niger* AUMC 16028 reduced heavy metals from industrial wastewater [58]. Fe_3O_4 NPs synthesized using *Aspergillus elegans* showed higher dye efficiency removal from wastewater [59].

5.3 Application of Plant-Based NPs for Bioremediation

AuNPs prepared from *Avicennia marina* 4-nitrophenol (4-NP reduced into 4-aminophenol [60]. *Adenophora triphylla* extract based AuNPs degraded MB within 5 minutes of treatment and congo red in 10 minutes [61]. Bismuthoxide (BiO-NPs) prepared from almond gum reduced 4-nitrophenol to 4-aminophenol and degraded Congo Red and Brilliant Green dyes in wastewater [62]. CuONPs prepared from *Solanum tuberosum* degraded MB dye [63]. *Vaccinium floribundum* based FeNPs removed of 88.24% and 81.90% of petroleum hydrocarbons from contaminated water and soil [64]. FeNPs prepared from *Lawsonia inerm* is degraded hydrocarbon in crude oil [65]. FeONPs synthesized from *Ruellia tuberosa* degraded dyes from industrial and domestic wastewater [66]. Apple peel extract based FeNPs removed Cr and Pb [67]. Fe_2O_3 NPs synthesized from coconut pulp degraded Rhodamine B dye by the process of photocatalysis [68]. Spinach (*Amarnthus blitum*) based Fe_3O_4 NPs effectively removed Cu (98%) from wastewater [69]. Magnetite Fe_3O_4 and hematite Fe_2O_3 NPs were prepared from *Lyptolyngbya foveolarum* and *Azospirillum brasilense* and efficiently removed nitrate, phosphate and ammonia from wastewater [70]. rGO/MgO nanocomposite was synthesized from *Hylocereus polyrhizus* and degraded dyes in wastewater [71]. Tin oxide (SnO_2 NPs) synthesized from ginger extract degraded MB, methyl orange (MO), Eriochrome black-T (EB) and rhodamine B (RB) dyes in wastewater by photocatalytic process [72]. Titanium dioxide (TDO) NPs was synthesized from *durva* herb. TDO NPs degraded both Rh-B and MB dyes [73]. Zirconium (ZrO_2NPs) synthesized from *Phyllanthus niruri* degraded various dyes such asmethyl red, methyl orange, and MB [74]. ZnO NPs synthesized using *Ruellia tuberose* degraded MB and malachite green dyes from textile industry wastewater [75]. ZnO and CuO NPs synthesized from *Serratula coronata* L. removed Pb(II) ion [76]. ZnO NPs synthesized from *Elettaria cardamomum* degraded MG dye in wastewater [77]. ZnO NPs prepared from extractof *Eucalyptus* leaf and effectively removed calcium ions (Ca^{2+}) from groundwater [78]. ZnONPs synthesized using *Sesbania grandiflora* degraded crystal violet dye and decreased total solid content in glass industry wastewater [79].

Table 1. Recent examples of green synthesized NPs for bioremediation

Nanoparticles	Source	Application in Bioremediation	Ref.
Algae			
AgNPs	*Chlorella ellipsoidea*	Removed the dyes such as methyl orange and methylene blue	[40]
Fe_3O_4-NPs	*Padina pavonica*, *Colpomenia sinuosa*, and *Petalonia fascia*	Reduced nitrogen and phosphorus-containing compounds from wastewater	[41]
AgNPs	*Nannochloropsis* sp	Decreased the concentration of lead in wastewater	[42]
AuNPs	*Chlorella vulgaris*	Decreased the concentration of lead in wastewater	[42]
AgNPs	*Sargassum latifolium*	Removed malachite green (MG) dye	[43]
CdS NPs	Blue green algae	Removed pollutants from wastewater	[44]
FeONPs	Algae biofilm	Removed methylene blue (MB), MG and phenol red (PR) dyes	[45]
NPs	*Pterocladia capillacea*	Increased bioremediation of toxic dyes	[46]

Bacteria			
AgNPs	*Macrococcus caseolyticus*	EliminatedTotal Petroleum Hydrocarbons in water samples	[47]
Magnetite Fe_3O_4 and hematite Fe_2O_3	*Lyptolyngbya foveolarum* extract and *Azospirillum brasilense*	Removed ammonia, nitrate and phosphate from wastewater	[48]
Platinum (Pt) and palladium (Bio-Pd) NPs	*Desulfovibrio vulgaris*	Removed various antibiotics from wastewater	[49]
Fungi			
AgNPs	*Aspergillus niger*	Increased the bioremediation of diesel oil contaminated soil and water	[50]
CuO-NPs	*Fusarium oxysporum*	Removed heavy metals and dyes from wastewater of textile industry	[51]
FeNPs	*Fusarium oxysporum*	Removed heavy metals from municipal wastewater	[52]
Fe_3O_4	*Aspergillus elegans*	Degraded dyes in wastewater	[53]
TiO_2 NPs	*Aspergillus penicillioides*	Degraded crystal violet dye	[54]
ZnO NPs	*Fusarium proliferatum*	Degraded anthracene	[55]
CuONPs and ZnONPs	Fungi	Removed pollutants from groundwater	[56]
Fe_3O_4 NPs	*Aspergillus flavus*	Reduced the toxicity of Reactive Black 5 (RB5) dye	[57]
Iron oxide NPs	*Aspergillus niger*	Reduced heavy metals from industrial wastewater	[58]
Fe_3O_4 NPs	*Aspergillus elegans*	Showed higher dye efficiency removal from wastewater	[59]
Plants			
AuNPs	*Avicennia marina*	Reduced 4-nitrophenol (4-NP) into 4-aminophenol	[60]
AuNPs	*Adenophoratriphylla*	Degraded MB and congo red (CR) dyes	[61]
Bismuthoxide (BiO-NPs)	Almond gum	Reduced 4-nitrophenol to 4-aminophenol and degraded CR and brilliant green dyes in wastewater	[62]
CuONPs	*Solanum tuberosum*	Degraded MB dye	[63]
FeNPs	*Vaccinium floribundum*	Removed of 88.24% and 81.90% of petroleum hydrocarbons from contaminated water and soil	[64]
FeNPs	*Lawsonia inermis*	Degraded hydrocarbon in crude oil	[65]
FeONPs	*Ruellia tuberosa*	Degraded dyes from industrial and domestic wastewater	[66]
FeNPs	Apple peel extract	Removed Cr and Pb	[67]
Fe_2O_3 NPs	Coconut pulp	Degraded Rhodamine B dye	[68]
Fe_3O_4 NPs	*Amarnthus blitum*	Removed Cu (98%) from wastewater	[69]
Magnetite Fe_3O_4 and hematite Fe_2O_3 NPs	*Lyptolyngbya foveolarum* and *Azospirillum brasilense*	Efficienly removednitrate, phosphate and ammonia from wastewater	[70]
rGO/MgO nanocomposite	*Hylocereus polyrhizus*	Degraded dyes in wastewater	[71]
SnO_2 NPs	Ginger extract	Degraded MB, methyl orange (MO), Eriochrome black-T (EB), and rhodamine B (RB) dyes in wastewater	[72]
TDO NPs	*durva* herb	Degraded both Rh-B and MB dyes	[73]
ZrO_2NPs	*Phyllanthus niruri*	Degraded various dyes such as methyl red, methyl orange, and MB	[74]
ZnO NPs	*Ruellia tuberose*	Degraded MB and malachite green dyes from textile industry wastewater	[75]
ZnO and CuO NPs	*Serratula coronata* L.	Removed Pb(II) ion	[76]
ZnO NPs	*Elettaria cardamomum*	Degraded MG dye in wastewater	[77]
ZnO NPs	Extract *of Eucalyptus* leaf	Effectively removed calcium ions (Ca^{2+}) from groundwater	[78]
ZnONPs	*Sesbania grandiflora*	Degraded crystal violet dye and decreased total solid content in glass industry wastewater	[79]

6. Challenges of Green NPs in the Bioremediation Field

The green synthesis method for production of NPs as discussed earlier is cheaper, environmentally friendly, laborious, easy to use and shows high efficiency. Due to these properties, green synthesized NPs are preferred in various fields of environmental science [80]. Microbe-based NPs provide a better down-streaming process, also the growth of microbes is controllable and highly achievable in less time therefore green synthesis method is a better approach to get a clean, safe and biodegradable environment in no time [81]. Besides the various benefits of nanotechnology, many disadvantages are also observed. One of the main disadvantages of nanotechnology is the commercialization of NPs on a larger scale. Large scale-up production of green synthesized nanotechnology requires the long-term stability of capping and reducing agents [82] and reusability of NPs which after a few trials show decreased efficacy [83]. Synthesis of NPs also needed chemical substances and green synthesis technology needed high temperature, lack of knowledge in the NPs production, different extracts generate variable size of NPs which make them unfit for large scale-up production. The low yield of green synthesized NPs cause hinders in field of nanobiotechnology [84].

7. Conclusions and Future Prospects

Bioremediation applications are increased due to overpopulation and environment pollution. However, traditional methods of bioremediation have some technical disadvantages that can be overcome by using nanotechnology. Nanotechnology combined with microorganisms has provided a green method toward the bioremediation of hazardous pollutants. In future, the green synthesized NPs can simply separate and reuse due to their efficiency and more stability. Furthermore, the green synthesis of NPs for water and soil treatment is an environment-friendly and potential technology to accomplish the zero effluent release in wastewater treatment and soil effluents. Thus, green nanotechnology focuses on enhancing treatment techniques by raising the process efficiency and reusability can save the operation cost in soil treatment as well as water. Therefore, the green synthesized NPs are important components utilized to remediate the water and soil pollution in the environment. More research is needed to investigate the toxic effects of NPs when introduced into the environment. Additional research will also be required to study the effectiveness of green synthesized NPs in scale up remediation of pollutants from wastewater. By addressing these problems, biogenic NPs can provide a great solution for pollutant remediation.

Author Contributions

All the authors actively contributed to manuscript drafting.

Data Availability Statement

Not applicable.

Conflicts of Interest

The authors declare no conflict of interest.

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