Brief review on synthesis and applications of plant mediated ZnO nanoparticles-A green approach

P.S.R. Vidya Sagar[a,b], Dharmasoth Ramadevi[c], Keloth Basavaiah[a,*], Sathish Mohan Botsa[d,*], K Bharath Kumar Naik[e]

[a]Department of Chemistry, Andhra University, Visakhapatnam-530003, Andhra Pradesh, India
[b]Department of Chemistry, Adikavi Nannaya University, Rajahmahendravaram, Andhra Pradesh, India
[c]Samuel George Institute of Pharmaceutical sciences, Markapur, Andhra Pradesh 523316, India.
[d]Bio Enviro Chemical Solutions, Visakhapatnam-530017, India
[e]Govt. Degree college, Araku valley, Andhra Pradesh 531149, India.

*Correspondence email: klbasu@gmail.com; bsathish401@gmail.com

Abstract
Nanotechnology is a rapidly growing research topic in all areas like engineering, medicine, pharmacy and science which involves materials with a size of 1–100 nm. Many researchers are now interested in obtaining metal and metal oxide nanoparticles *via* biological methods as eco-friendly and less toxic. Compared to physical and chemical synthesis, the green approach is found to be safer and more eco-friendly. Many sectors rely on zinc oxide for its varied qualities which have been strengthened through nanoscale manufacturing. Green production of zinc oxide nanoparticles utilizing several biological substrates has been proven and achieved. This study summarized recent breakthroughs in biosynthesized ZnO NPS synthesis, mechanism, characterization, and applications in agriculture, medicine, and other sectors.

Keywords ZnO Nps; Plant extracts; Green synthesis; Bio-medical applications.

1. Introduction
The subject of Nanotechnology has seen remarkable advancements in science and technology in the last decade in particular. It is possible to create new systems, structures, devices, and nanoplatforms by incorporating nanoparticles with varied physicochemical properties [1-2]. Nanomaterials are tiny particles with increased thermal conductivity and catalytic activity that are in the range of 1-100 nanometers in diameter. The huge surface area to volume ratio makes them chemically inert [3]. Chemical toxicity in the environment can be reduced through the careful preparation of diverse metal and metal oxide nanoparticles using plant extracts, which is a plan of action for controlling toxic chemical production [4]. Numerous plants can be used to synthesise NPs, including *Physalis alkekengi L, Trifolium, Justicia adhoda*, *Cassia Auriculata*, *Aloe barbadenis*, *Pongamia pinna*, *Limonia acidissima*, *Aspidoterys cordata*, and *Bauhinia racemosa* and *Plectranthus amboinicus* [5–7]. ZnO NP synthesis using biological techniques has seen a significant rise in interest over the last few years. Covalent or ionic semiconductor ZnO is an amphoteric oxide. Hexagonal wurtzite, cubic rock salt, and cubic zinc blend are all forms of ZnO. The hexagonal wurtzite structure is the most frequent because it is the most stable at ambient temperatures.

2. Nanoparticle synthesis methods

Nanoparticles in general synthesized by two approaches, Top-Down and Bottom-Up synthesis. In the Top-Down approach, gradual reduction of large particles takes place. In this method, we synthesize large-scale patterns first and then reduce them to nanoscale levels through plastic deformation. This method cannot be used for large scale production of nanoparticles because it is a slow and costly process [8]. The bottom-up method is the green biosynthesis of nanoparticles in which the metal atoms form clusters, and then finally nanoparticles. The size and form of the nanoparticles can be managed, which can be used for different applications. The simple method of synthesizing nanoparticles requires only materials like metal salt (precursor) and green substrates. The nanoparticle synthesis process allows various parameters, such as metal salt concentration, green substrate concentration, time and temperature to be modified and the solution's pH to achieve properties necessary for the respective applications.

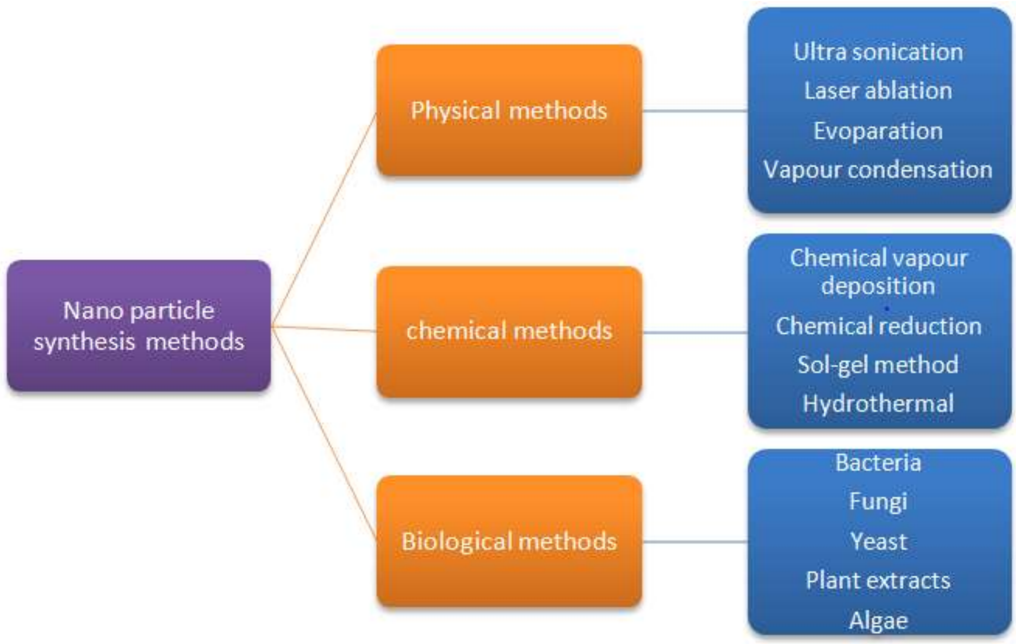

Fig.1: Various methods for the preparation of nanoparticles

2.1 Physical Method

To create stable and well-defined nanoparticles, physical forces are exploited as an attraction between microscopic particles. Physical approaches include such things as vapour condensation, amorphous crystallisation, colloidal dispersion, and physical fragmentation, to name just a few [9]. Plasma technique, laser ablation, and thermal evaporation are the most extensively utilised physical methods for the synthesis of ZnO NPs [10]. Physical methods have several drawbacks, including high pressure and temperature, a large machine setup area, and the need for expensive equipment [11].

2.2 Chemical Methods

Precipitation, hydrothermal procedures, and sol-gel synthesis are the most commonly utilised chemical processes for producing ZnO NPs. By using zinc precursor and chemical reagent in the sol-gel synthesis, the pH of the solution can be controlled and precipitation of Zn $(OH)_2$ is prevented. The ZnO NPs are then formed by heating the solution to high temperatures [12-13]. Because of their high energy consumption, hazardous chemicals, and expensive equipment requirements, chemical methods are not used extensively.

2.3 Biological Methods

Bio-synthesis involves the green route for the production of ZnO nanoparticles and this method utilizes Yeast, Algae, Fungi, Bacteria, and plant extracts as the reducing agents. Because plant extracts include such high amounts of phytochemicals or secondary metabolites, they are thought to have a high ZnO NP synthesis potency [15]. It is safe, cost-effective, environmentally friendly and nonhazardous; it is biocompatible; and it can be produced in big quantities. [16–19]. There have also been attempts to synthesise NPs using plant parts such as roots, leaves, stems, seeds, and fruits since the phytochemicals found in these components serve to reduce and stabilise formed nanoparticles [20–26].

3. ZnO nanoparticles

Zinc oxide (ZnO) is an n-type metal oxide. Zinc oxide nanoparticles (NPs) have gained attention in recent years because of their wide range of applications in electronics, optical, and biological systems [27–33]. The US Food and Drug Administration (FDA) has designated ZnO as a GRAS metal oxide [34]. For example, ZnO NPs have strong catalytic activity, optical characteristics, UV filtering properties, anti-inflammatory properties, and wound healing properties (35–39) because of their high exciton binding energy (60meV) and huge band gap (3.37eV). Drug delivery, anticancer, antidiabetic, antibacterial/antifungal and agronomic qualities are only a few of the many medicinal applications of this substance [42-44]. Many distinct ZnO NPs have been reported, including nanoflake, nanoflower, nanorod and nanowire [45-47].

3.1. Green synthesis of ZnO nanoparticles

Toxic chemicals aren't used, and a large amount of energy is put into the biological synthesis, thus this new strategy is both cheaper and better for the environment [48-51]. The biosynthesis of metal oxide and metal nanoparticles is more environmentally friendly than the standard chemical or physical processes according to the literature [49-50]. That's why they're more commonly known as "green synthesis" now. NP production has been widely researched by plants belonging to the Lamiaceae family, such as Plectranthus amboinicus [52], which exhibited various shapes and sizes of NPs, including spherical and quasi-spherical, hexagonal, and rod-shaped NPs. The size of generated NPs decreased as the concentration of a plant extract increased [52].

3.2. Mechanism for the green synthesis of ZnO NPs

The chemical elements of the plant extract utilised to obtain the Zinc oxide nanoparticles have been the theme of the mechanism pathway proposed by certain writers. Instead of forming a coordinated complex, they claim that plant extract reduces zinc (II) ions to metallic zinc. ZnO nuclei were formed after the zinc precursor had been reduced to metallic zinc and dissolved oxygen in the solution. Phtyo-constituents were also proposed to act as a stabilizing agent to avoid particle accumulation [53-55].

Biological sources such as plants are used to synthesize nanoparticles made from metal ions [56, 49]. Plant-based substrates are more economical, easier to process, and safer to use than microorganisms because of these factors. Plant-based substrates eliminate risks to human health. There are many ways to obtain plant extracts, including simple exposure to a solvent such as distilled water or alcohol [57]. Methylxanthines, phenolic acids, flavonoids, and saponins are only a few examples of the plants' potent active constituents that have been studied extensively and known as antioxidants [58-61]. These chemicals can neutralise ROS and free radicals as well as chelate metals [62]. Due to their ability to reduce or chelate metal ions, they stabilize the generated nanoparticles, it may be identified that antioxidants which are present in plants are accountable for the green production of metal oxide and metal Nps [63-64].

3.3. Factors influencing biosynthesis of nanoparticles

When plants are used in biosynthesis, several aspects influence the synthesis, characterization, and application of nanoparticles. The following is a list of some of the most critical considerations. Zinc salt concentrations, pH changes, reaction times, and temperatures all affect particle size and shape significantly. ZnONPs with varied morphologies and sizes were formed when the technique adopted was heated or cooled. When the particles were heated to 30°C, the morphology became erratic and their crystallinity decreased significantly. Agglomerated nanoparticles in the shapes of cauliflower and dumbbell were found in the nanoparticles produced by heat treatment at 60°C and 100°C, respectively [65]. As the temperature rises, they found that crystal growth and nucleation rates increase at a quicker pace, which leads to greater particle sizes [66]. The period between heat treatments can also affect the formation of clusters, which is related to agglomeration. The thermal treatment at 50 °C produced more agglomerates and particle growth when the treatment time was extended from 30 to 90 minutes.

Metals and metal oxides' particle size and morphology can be dramatically altered during synthesis by changes in pH conditions [67-71]. ZnO Nps was synthesized at pH-6 or 7, and bigger agglomerates were detected when the pH was increased to 11.0 [72]. The ZnO NP biosynthesis proposed in their paper would be best carried out at a neutral pH. It is possible that the creation of $Zn(OH)_2$ may occur in alkaline pH solutions, which could affect the generation of ZnO NPs [73]. Study the effect of zinc salt and biological extract concentration on particle size and yield, operating time and temperature on the green synthesis of zinc oxide nanoparticles. According to the results of this study, a higher yield was obtained by utilizing a low zinc precursor concentration (65 g L^{-1}) and the reaction time (2 h) and the highest temperature (200 °C). Finally, we concluded that zinc nitrate concentration was the most significant influence on particle size [74].

Table 1: Summary of ZnO nanoparticles synthesized via a green approach

S.No.	Plant Extract	Shape	Size Distribution (nm)	Reference
1	Azadirachta indica	spherical	40nm	[75]
2	Leschenaultiana.L	hexagonal, Spherical	42.5nm	[76]
3	Vitex negundo	spherical	60nm	[77]
4	Plectranthus amboinicus	Spherical ,hexagonal	35nm	[78]
5	Solanum nigrum	hexagonal	29.79nm	[79]
6	Tamarindus indica	spherical	28nm	[80]

7	Moringa oleifera	spherical	14nm	[81]
8	OcimumbasilicumL.var.purpurascens benth leaf	hexagonal	50nm	[82]
9	Mimosa pudica	Wurtzite	2.71nm	[83]
10	Vitex trifolia L.	spherical	30nm	[84]
11	Hibiscus rosa-sinensis	spherical	35.95nm	[85]
12	Carica papaya	Hexagonal ,wurtzite	10.2nm	[86]
13	Cassia fistula	Quasi spherical	15.8nm	[87]
14	L. aculeata	spherical	12nm	[88]
15	*Aloe Vera* (Liliaceae)	Spherical, hexagonal	15nm	[89]
16	*Phyllanthus niruri*	Hexagonal, wurtzite and spherical	25.61nm	[90]
17	*E. crassipes*	Spherical without aggregation	32nm	[91]
18	*Anisochilus carnosus*	Hexagonal ,wurtzite	56.14nm	[92]
19	*Parthenium hysterophorus L.*	Spherical, hexagonal	27±5nm	[93]
20	*Plectranthus amboinicus*	Rod-shaped	88nm	[94]
21	*S. album*	Nanorods	100nm	[95]
22	*Sphathodea campanulata*	spherical	20nm&50nm	[96]
23	Aloe barbadensis miller	Spherical	35nm	[97]
24	Corriandrum sativum	Wurtzite, hexagonal	66nm	[98]
25	Acalypa indica	Nano cubes	80nm	[99]
26	Calotropis gigantea	Spherical	32.5nm	[100]
27	Sargassum myriocystum	Spherical	36nm	[101]
28	Tabernaemontana Divaricate	Spherical	36nm	[102]
29	Murraya koeininggi	Hexagonal	32.5nm	[103]
30	Melia azedarach	spherical	64.5nm	[104]
31	Euphorbia hirta	spherical	22.5nm	[105]
32	Urtica dioica	spherical	22nm	[106]
33	Prosopis juliflora	hexagonal	65nm	[107]
34	Calliandra. Haematocephala	flower	19nm	[108]
35	Cinnamonum tamala	Spherical, hexagonal	26.57nm	[109]
36	Vaccinium erctostaphylos L	Spindle	12.4nm	[110]
37	Berberis aristata	needle	96nm	[111]

38	Juglans regia L	Spherical	55nm	[112]
39	Artocarpus heterophyllus	spherical	18nm	[113]
40	*Camelia sinensis*	spherical	8nm	[114]
41	*Couroupita guianensis*	Nano flakes	-	[115]
42	*Costus woodsonii*	Spherical	22.5nm	[116]
43	*Eclipta alba* leaves	spheres	6nm	[73]
44	*Menta pulegium* L.	spheres	40nm	[117]
45	*Stevia* leaves	rectangular	50nm	[118]
46	*Punica granatum leaves*	spheres	20nm	[119]
47	*Alliumsativum*, *Petroselinum Crispum* ,*Allium cepa*	wurtzite, Hexagonal	70nm	[120]
48	*Anisochilus carnosus*	Hexagonal wurtzite	30nm	[121]

3.4. Characterization of ZnO NPs

The characterization of the prepared ZnO nanoparticles is done by different techniques like X-ray diffraction (XRD), UV-Visible spectroscopy, FT-IR spectroscopy, Field emission scanning electron microscopy (FE-SEM), Energy dispersive X-ray analysis (EDAX), Transmission electron microscopy (TEM), Raman spectroscopy, X-ray photoelectron microscopy (XPS), Thermo gravimetric-Differential thermal analysis (TG-DTA), and Atomic force microscopy (AFM) [121-124].

3.5. Applications of green synthesized ZnO NPs

Fig.2: Nanoparticles have a wide range of applications

3.5.1. Antimicrobial activity

In comparison to synthetically generated zinc oxide nanoparticles, biologically green and environmentally friendly nanoparticles have numerous advantages. Aside from their antibacterial activity against bacteria and other microscopic organisms and their strong photodegradation properties, these nanoparticles have shown promise for use in medication delivery and anticancer treatment. Here, we've discussed some of the applications [63]. Years of research have established zinc oxide's medical benefits. Poison ivy, poison oak, poison sumac, sunburns and bug bites can all be treated with calamine lotion that contains a blend of iron oxide (Fe_2O_3) and zinc oxide. Zinc oxide and eugenol ligand, known as zinc oxide eugenol, is used in dentistry as a prosthetic and restorative [125-126].

Compared to zinc oxide as a whole, zinc oxide nanoparticles have a more potent antibacterial effect [127] when it comes to nanoparticles' antibacterial properties, the high surface area-to-volume ratio and their small size is crucial. Zinc oxide nanoparticle antibacterial activity rises with decreasing particle size, According to Akbar et al. [128]. When used at low concentrations, nanoscale zinc oxide is a highly efficient antibacterial and antifungal agent for both Gram-positive and Gram-negative bacteria. [129-134] it has been shown that coating materials and textiles with zinc oxide nanoparticles provides antibacterial and antifungal properties [135]. These nanoparticles' antibacterial activities are attributed to their electrostatic interaction with microbial cell surfaces. Zinc oxide nanoparticles' cytotoxic properties cause pores to form on the surface of the microbial cell, allowing cytoplasmic contents to leak out and causing cell death [136], the integrity of the microbial cell membrane can be compromised by nanoparticles' direct interaction with it [137]. ZnO NPs are capable of destroying germs and have been shown to have a positive antibacterial effect by numerous researchers. ZnO nanoparticles were synthesized by Bhuyan et al. using a variety of concentrations of *Azadirachta indica* and tested against Gram-positive and Gram-negative bacteria. The results showed that as ZnO NP concentration increased, bacterial growth decreased and Gram-negative bacteria were more sensitive to ZnO NPs. This suggests that these nanoparticles may have antibacterial properties. Thus, ZnO NPs can easily be used in investigations where a bacteria-free environment is required [138].

3.5.2. Photocatalytic Activity

Photocatalytic degradation of dyes such as methylene blue, methyl orange, etc. may be achieved using ZnO nanoparticles with biological origin. The textile and cosmetics industries, as well as other sources, are major sources of water pollution caused by methylene blue dye. As a result, environmental concerns drew attention to these ZnO NPs [138]. Toxic effluents generated by the textile industry harm aquatic life and the ecology, resulting in serious environmental issues. ZnO, as a photocatalyst, is one of a variety of semiconductors that have been studied to reduce water pollution. With a 3.37 eV band gap. All Zinc oxide nanoparticles generated using *Allium sativum*, *Petroselinum crispum* and *Allium cepa*, are capable of dissolving methylene blue dye, according to a comparison of their degradation efficiency. It was shown that ZnO NPs degraded the dye more efficiently than ordinary zinc oxide due to the NPs' smaller size, which provided more reaction sites on the catalyst surface [139].

Factors like the ratio of catalyst loading to the dye concentration have been shown to have a significant impact on dye removal. The degrading efficiency of ZnO NPs was affected by this. The rise in dye concentration reduces the path of photons entering the solution, decreasing the number of holes and radicals needed to degrade ZnO NPs, which is demonstrated to be inversely related to dye degradation efficiency [140]. Among many synthesised ZnO NPs with various structures, an experiment on the photodegradation effectiveness of Zinc oxide nanoparticles against Alizarin Red-S dye discovered the best degradation performance of ZnO NPs with nano-flowers [141].

3.5.3. Drug Delivery and Anticancer Activity

It has been demonstrated that ZnO NPs can induce apoptosis in leukemic cells while having no harmful effects on healthy cells [142]. Furthermore, it has been shown that ZnO NPs can cause considerable tumour T cell specific toxicity while causing little harm to normal body cells 143]. In comparison to other nanoparticles, ZnO NPs are increasingly used in cancer treatment delivery due to their biodegradable and low hazardous qualities. When medications such as baicalin, curcumin, doxorubicin, and paclitaxel are placed onto Zinc oxide nanoparticles as delivery vehicles, the solubility and toxicity increase [144-146].
As a result of drug delivery, the condition is treated quickly, and subsequently, the curative impact can be maintained throughout time. They also showed outstanding observational results demonstrating better in vitro reduction of malignant cell reproduction at elevated concentrations and a minor effect on target cancer cells at minor concentrations in a dose dependent manner. As a result, nanoparticles of varied sizes are a good agent for improving the administration of leukaemia malignant cells' medication. *Borassus flabellifer* (palm) fruit extract ZnO NPs are another application of Zinc oxide nanoparticles in drug delivery and anti-cancer therapy, as demonstrated in this study [147].

3.5.4. Agricultural Applications

When it comes to treating fungus and other microbial illnesses in agricultural animals and plants, ZnO NPs are considered a better option than conventional antibiotics. *Artemia salina* larvae have been shown to be highly resistant to ZnO NPs [148]. The inorganic properties of ZnO NPs are more effective than several commercially available antibiotics in inhibiting microbe growth at higher temperatures and other extreme circumstances [149]. In bacteria, ZnO NPs' small size, composition, and morphology increase their interaction and penetration through the cell's core, resulting in cell damage and death [150]. ZnO NPs have been found to have the ability to boost food crop yields in several studies [151-155].

3.5.5. Anti-fungal efficacy

More than a dozen studies on ZnO NPs' antibacterial and antifungal properties have been published [156-157] and significant antimicrobial efficacy reported. ZnO NPs were found to have strong antifungal action against *Alternaria alternative*, *Botrytis cinerea*, and *Fusarium oxysporum* development, with inhibition zones of 64, 128, and 64 mm, respectively, in previous research [158].

3.5.6. Anti-viral activity

For the Herpes simplex virus 1, zinc oxide nanoparticles have demonstrated antiviral activity (HSV- 1). HSV-1 pathogenesis may be affected by zinc oxide, which has been reported to interact with proteins [159]. Herpes simplex virus 2 (HSV-2) can also bind to zinc oxide nanoparticles, preventing the virus from attaching to the host cell. [160]

Table 2.Summary of reported activities of zinc oxide nanoparticles:

S.No	Plant name	Activity	Ref
1	Aloe vera	Antibacerial	[161]
2	*Limonia acidissima* (leaf)	Antibacterial	[162]
3	Vitex negundo	Antibacterial	[163]
4	Camellia sinensis	Antibacterial	[164]

5	Catharanthus roseus	Antibacterial	[165]
6	*Euphorbia Jatropha* (stem)	Antibacterial	[166]
7	Cassia fistula	Antibacterial	[167]
8	Trifolium pratense	Antibacterial	[168]
9	Emblica Officinalis	Antibacterial	[169]
10	Hibiscus subdariffa	Antibacterial	[65]
11	Punica granatum	Antibacterial	[170]
12	Prunus sp. [plum]	photocatalytic	[171]
13	Cinnamomum Tamala	Antibacterial	[172]
14	Prosopis farcta	Antifungal and cytotoxic	[158]
15	Mentha pulegium	Antimicrobial	[173]
16	Manginefa. indica	Antioxidant	[174]
17	Ceropegia candelabrum L	Antibacterial	[175]
18	Suaeda aegyptiaca	Antioxidant	[168]
19	Lavandula vera	Cytotoxic and antioxidant	[176]
20	Averrhoa carambola L	Agricultural	[177]
21	Suaeda japonica	Photodegradation	[178]
22	Marsdenia tenacissima	anticancer	[179]
23	Costus igneus	Antidiabetic, antibiofilm, and antioxidant	[180]
24	Glycosmis pentaphylla	antimicrobial	[181]
25	Echinochloa frumentacea	cytotoxicity	[182]
26	Cuminum cyminum	Antibacterial	[183]
27	Calotropis gigantean	cytotoxicity	[184]
28	*Ceropegia candelabrum* (leaf)	Antibacterial	[185]
29	*Celosia argentea* (leaves)	drug delivery, antibacterial	[186]
30	*Couroupita guianensis* (leaves)	Antibacterial	[187]
31	*Solanum nigrum* (leaves)	Antibacterial	[188]
32	*Jacaranda mimosifolia* (flower)	Antibacterial	[189]

Conclusion and future perspective

Plats-based biogenic synthesis of ZnO Nps has been thoroughly examined in this review. It also explains how ZnO nanoparticles are formed. Synthesizing with plant extracts has been shown to be less harmful to the environment and more cost-effective. Nanoparticles' final properties can be predicted using parameters like concentration, temperature, pH, and reaction time, which are closely linked to these parameters. The biological source is critical in this regard. Despite the complexity of biological substracts, the evaluation of green synthesis of Nps remains a difficulty. To have an insight into the chemical process and reactions that take place throughout the synthesis of ZnO Nps through biological synthesis, additional research is needed. In addition, the food, pharmaceutical, and biological industries were well-represented in this review. Plants' generic alteration with improved metal accumulation capacity and metal liberality is the next strategy for increasing productivity in nanoparticle synthesis.

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