Chapter 13
Green Synthesized Nanoparticle-Mediated Wastewater Treatment

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Abstract Freshwater in the environment is getting contaminated day by day due to constant pollution of freshwater bodies through different anthropogenic sources which leads to scarcity of freshwater for the people usage. Green synthesis of nanoparticle is a very efficient method of nanoparticle synthesis. Nowadays these nanoparticles are used to treat wastewater system. The overview of recent advancement in nanotechnology for water and wastewater treatment mechanisms is provided, including nanobased materials, such as nanoadsorbents, nanometals, nanomembranes, and photocatalysts. Various advantageous roles governed of these materials as well as technical barriers when compared with conservative processes are being demonstrated. The commercial value of these materials is presented and viewpoint on further research value is given for each type of nanobased material and process. This chapter includes recent trends in green synthesis of nanoparticles using different plants and their parts like leaves, stem, etc., and their application in treatment of wastewater.

Keywords Wastewaters · Industries · Nanoparticles · Green synthesized · Treatment

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13.1 Introduction

The freshwater is essentially significant for humans, animals, and plants, not just for drinking, but also for irrigation, domestic use, and other vital activities. It is expected to become increasingly inadequate in the near future, and this is partly because of environmental change (Pretty 2007). Due to the rapid increase in world population as well as global warming, water reservoirs are decreasing at a very fast rate. In this approach, discreet use of water resources and reuse of treated wastewater for various purposes have been perceived as the best method for monitoring the restricted resource of freshwater (Shittu and Ihebunna 2017). The occurrence of microorganisms and trace metals is the principle indication of water contamination. The trace metal contamination of water is a public health concern, with several health risks associated with it (Bharagava et al. 2018; Kishor et al. 2019; Rahman and Singh 2018). The prokaryotic organisms, for example, *Escherichia coli*, *Shigella* spp., *Salmonella* spp., *Vibrio* spp., and *Cryptosporidium*, are known to be transmitted by water and cause ill-health when this water defiled by microorganisms is consumed (Ashbolt 2004).

Nowadays, metal nanoparticles especially gold and silver NPs are of particular interest to researchers due to their specific properties and wide applications in the field of medical sciences. Biosynthesis approach of NPs is considered as a better alternative than physical and chemical method (Ahmad et al. 2003; Song et al. 2010; Gautam et al. 2018) as it is eco-friendly, cost effective, and less time requiring. Different materials are categorized as biological for nanoparticle synthesis; these include plant extracts, microscopic organisms, growth chemicals, and actinomycetes (Bharagava and Chandra 2010; Chandra et al. 2012; Agarwal et al. 2017). But plant extracts have been the major focus due to their abundant nature and the phytochemical composition (Bharagava et al. 2008).

There is constrained chance of an expansion in the supply of freshwater due to ever-increasing demand by growing population throughout the world; additionally, water-related issues are anticipated to increase further because of atmospheric changes and because of population growth throughout the following two decades (Vörösmarty et al. 2000). It is evaluated that overall population will increment by about 2.9 billion as of now till 2050 (as indicated by UN's normal projections) (Rockström 2003). Nanotechnology has been considered as a useful method for identifying and solving problems related to the water quality (Bottero et al. 2006). Nanomaterials are adding to the improvement of progressively effective treatment forms among the new water system (Obare and Meyer 2004).

There are numerous parts of nanotechnology that address the different issues of water quality so as to make sure the ecological stability is maintained. This information gives a unique point of view on fundamental research of nanotechnology for water/wastewater treatment and reuse by concentrating on difficulties of future research. Advancement in the field of nanoscience technology and designing propose that huge numbers of the flow issues including water quality could be resolved or greatly diminished by utilizing nanoadsorbents, nanocatalysts, bioactive nanoparticles, nanostructured synergist layers, nanotubes, attractive nanoparticles, granules, chip, high surface region metal molecule supra subatomic assemblies with trademark length sizes of 9–10 nm including clusters, small-scale particles, nanoparticles, and colloids which significantly affect water quality in natural environment (Diallo and Savage 2005).

13.2 Green Synthesis of Nanoparticles

Nanoparticles can be prepared by various methods which are usually categorized in to two major synthetic routes which are the top-down and the bottom-up approaches (Fig. 13.1) (Rahman and Padavettan 2012). In the top-down (physical) approach, nanoparticles are obtained from their bulk materials (macroscopic) using different methods and techniques like mask, ball milling thermal decomposition, irradiation, laser ablation, arc discharge, etc. (Mijatovic et al. 2005). In bottom-up (chemical and biological) approach, nanoparticles are obtained from their atomic level building blocks which react to generate nanoparticles of the desired shape and size. These methodologies utilize larger of nanostructures (Barth et al. 2010). During self-gathering the physical forces working at the nanoscale are utilized to join fundamental units into bigger stable structures. Physical and chemical methods are being utilized broadly for generation of metal and metal oxide nanoparticles as shown in Fig. 13.1 (Ma et al. 2017). However, these methods of production require the use of highly reactive and toxic reducing agents such as sodium borohydride and hydrazine hydrate, which account for undesired detrimental impacts on the environ-

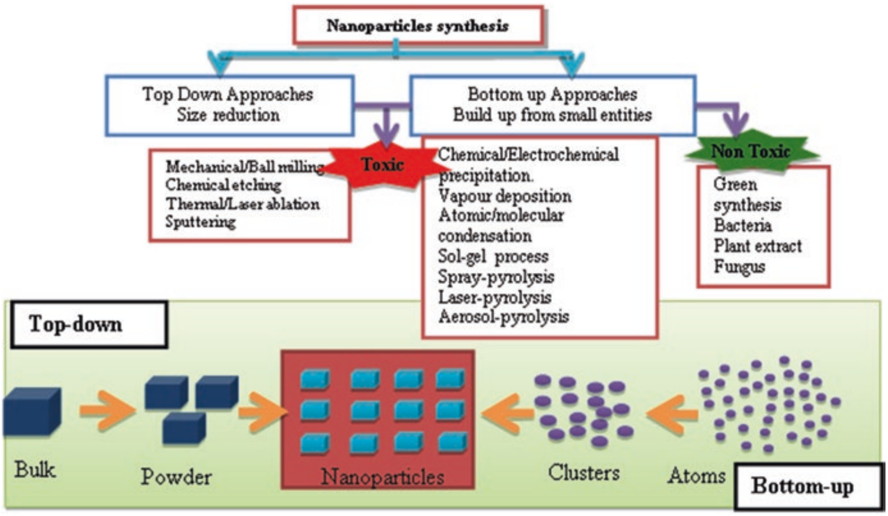

Fig. 13.1 Top-down and bottom up approaches of nanoparticles synthesis involving their toxic and nontoxic effects

ment, plant, and animal life (Saif et al. 2016). Researchers are putting continue efforts to develop facile, effective, and reliable green chemistry processes for the production of nanomaterials.

Different organisms behave as clean, eco-friendly, and sustainable precursors in order to produce stable and well-functionalized nanoparticles. These may incorporate bacteria, actinomycetes, fungi, yeast, viruses, etc. Therefore, it is critically important to develop a more reliable and sustainable process for the synthesis of nanomaterials. The idea of economic viability, ecological sustainability, and social adaptability as well as the availability of local resources should be kept in mind during the production of nanomaterials (Saif et al. 2016). In order to keep the cost of the final completed nanotechnology-based products affordable to consumers, industries must maintain a delicate balance between environmentally sound green processes and their sustainability. The green nanotechnology-based production processes operate under such conditions where the involvement of toxic chemicals is negligible. Metal NPs have been extensively studied because of their specific characteristics such as catalytical, optical, electronic, antimicrobial, and magnetic properties.

Among noble metals, silver is the metal of preference in the field of biological systems, living organisms, and medicine (Bootharaju et al. 2017). The biosynthesis of nanoparticles involves use of biogenic matter including plant extracts, biopolymers, and microbial residue like bacteria, fungi, algae, and yeast for nanomaterial fabrication (Sharma et al. 2015). Development of biocompatible, nontoxic, and eco-friendly methods for the synthesis of nanoparticles is mainly concerned in green chemistry (Narayanan and Sakthivel 2010). Biosynthesis of metal nanoparticles using plants is currently under exploitation. The biological synthesis of metal nanoparticles (especially gold and silver nanoparticles) using plants (inactivated plant tissue, plant extracts, and living plant) has gained more attention as a suitable alternative to chemical procedures and physical methods (Iravani 2011). Synthesis of metal nanoparticles using plant extracts is quite cost effective and therefore can be used as an economic and useful alternative for the large-scale production of metal nanoparticles. Extracts from plants may act as both reducing and capping agents in nanoparticle synthesis. The bioreduction of metal nanoparticles by combinations of biomolecules found in plant extracts (e.g., enzymes, proteins, amino acids, vitamins, polysaccharides, and organic acids such as citrates) is environmentally beneficial, yet chemically complex (Sharma et al. 2009).

Because of the important and critical role of plants in biosynthesis of metal nanoparticle production, the green synthesis of metal nanoparticles using plants has been discussed in this part. Current research in biosynthesis of nanometals using plant extracts has provided a new opportunity in fast and nontoxic methods for production of nanoparticles. Many researchers have reported the biosynthesis of metal nanoparticles by plant leaf extracts and their subsequent potential applications (Ghosh et al. 2012). MubarakAli et al. (2011) have studied the bioreduction of gold and silver ions by leaf broth of *Pelargonium graveolens* and *Azadirachta indica* (Thakkar et al. 2010). Moreover, they have explored the formation mechanism of triangular gold nanoprisms by *Cymbopogon flexuosus* (lemongrass) extracts where the nanotriangles seemed to grow by a process involving rapid bioreduction, assembly, and room temperature sintering of "liquid-like" spherical gold nanoparticles (Brumbaugh et al. 2014).

Also rapid synthesis of stable gold nanotriangles can be achieved which involves the use of *Tamarindus indica* (tamarind) leaf extract as a reducing agent (Kumar and Yadav 2009). The shape of metal nanoparticles subsequently changed their optical and electronic properties (Storhoff et al. 2000). They have also demonstrated the synthesis of gold and silver nanoparticles having variety of shapes (spherical and triangular) and sizes using *Aloe vera* plant extracts (Bar et al. 2009). It was explained that only biomolecules having molecular weight less than 3 kDa caused reduction of chloroaurate ions, leading to the formation of gold nanotriangles. Nevertheless, the bioreduction of silver ions proceeded barely in the presence of ammonia. The aqueous solution of gold ions when exposed to *Coriandrum sativum* leaf extract was reduced and resulted in the extracellular biosynthesis of gold nanoparticles with spherical, triangle, truncated triangle and decahedral morphologies ranging from 6.75 to 57.91 nm. These nanoparticles were stable in solution over a period of 1 month at room temperature (Korbekandi et al. 2009).

Njagi et al. (2010) used aqueous *Sorghum* sp. (hybrid sorghum) bran extract for nZVI synthesis; eucalyptus globulus leaf extract was used as a bioreducing agent to synthesize nZVI. P. Wang synthesized iron nanoparticles using *Eucalyptus* leaf extract by adding 0.1 M $FeCl_3$ solution in a ratio of 1:2; Wang et al. synthesized polydispersed iron nanoparticles employing *Eucalyptus* leaf extract obtained from its leaf litter. nZVI, Fe_3O_4, and Fe_2O_3 were the different forms of nanoparticles synthesized during the process; FeO/Fe_3O_4 nanoparticles were successfully synthesized using pomegranate leaf extract by Rao et al. (2013); these nanoparticles were coated on two strains (NCIM 3589 and NCIM 3590) of heat-killed yeast cells *Yarrowia lipolytica*, which is considered a good biosorbent itself; Venkateswarlu et al. (2014) used plantain peel extract as a low-cost bioreducing agent for synthesizing magnetite nanoparticles.

Iron salt solution was hydrolyzed which led to the formation of ferric hydroxide, and then it was subsequently reduced by various biomolecules to form Fe_3O_4 nanoparticles, banana peel ash extract was used to synthesize iron oxide nanoparticles, and aqueous extract of *Colocasia esculenta* leaves was used to reduce graphene oxide by Thakur and Karak (2014). Daniel et al. (2013) used leaf extract of the evergreen shrub *Dodonaea viscosa* to synthesize iron nanoparticles where the effect of leaf extract concentration on nanoparticle synthesis was studied. Senthil and Ramesh (2012) reported the green synthesis of Fe_3O_4 nanoparticles at room temperature using leaf extract of *Tridax procumbens*, and Narayanan et al. (2011) synthesized superparamagnetic magnetite/gold (Fe_3O_4/Au) hybrid nanoparticles at room temperature using grape seed proanthocyanidin (GSP) for the first time. Machado et al. (2014) screened leaf extracts of 26 plants for the production of nZVI. During synthesis variables like leaf extraction, temperature, time, and leaf mass to solvent volume ratio were checked. Eighty degree Celsius was identified as the optimum temperature, whereas the other variables like extraction, time, and leaf mass to solvent volume ratio varied as per leaf type.

In another study, Machado et al. (2014) synthesized nZVI using grape marc, black tea, and vine leaf extract. Kumar et al. (2013) synthesized stable iron oxide (Wustite) using aqueous extract of *Terminalia chebula* dry fruit pericarp. Leaves of three plants native to Australia, namely, *Eucalyptus tereticornis* (A), *Melaleuca nesophila* (B), and *Rosmarinus officinalis* (C), were explored by Wang et al. (2014). Phumying et al. (2013) synthesized Fe_3O_4 nanoparticles by the hydrothermal method using *Aloe vera* plant extract. Ahmmad et al. (2013) successfully synthesized highly pure hematite (α-Fe_2O_3) nanoparticles by the hydrothermal method using green tea (*Camellia sinensis*) leaf extract. Nanoparticle synthesis was carried out by exposing pretreated and milled powder of *Medicago sativa* (Alfalfa) to the salt solution of ferrous ammonium sulfate.

13.3 Use of Various Green Synthesized Nanoparticles in Wastewater Treatments

Treating polluted water has become a major challenge as various heavy metals and pathogenic organisms cause human health problems and can present a major threat to the environment (Ayangbenro and Babalola 2017). The use of magnetite nanoparticles as a sorbent has shown promise as an emerging treatment of polluted water. In this study, a common weed plant (*Lantana camara*) was used for synthesis of magnetite nanoparticles (MNPs). Synthesized MNPs were characterized for physical properties and batch absorption studies were undertaken to assess the potential for removal of heavy metals, namely, lead, under varying conditions (Sun et al. 2011). Nanotechnology can enable a distributed water reuse and treatment paradigm and offer leapfrogging opportunities to obviate concerns of water quality degradation within distribution networks, alleviate dependence on major system infrastructure, exploit alternative water sources (e.g., recycled new water) for potable and agricultural use, and abate energy consumption.

This vision may be particularly appropriate for developing countries that face rapid degradation of water quality with increasing pressure for cleaner water to meet more stringent environmental, public health, and food safety standards. This scenario underscores the need for a new class of “high-performance” water treatment technology. Future urban systems in developing countries will likely increasingly rely on nanotechnology-enabled water monitoring, treatment, and reuse systems that target a wide variety of water pollutants and are affordable and easy to operate. This will also contribute toward a zero discharge paradigm, which is an ultimate goal of sustainable urban water management. Examples of engineered nanomaterials (ENMs) that can enable this vision are summarized in this chapter. Such novel technologies for water treatment at both point of use and community scale are of great value for increasing the effectiveness and robustness of water distribution networks, allowing access to clean water to users that are not connected to a central network, and for emergency response following catastrophic events.

Various industries such as paper and pulp, textile, tanneries, cosmetic, coffee pulping, pharmaceutical food processing, electroplating, and dye manufacturing units discharge colored and toxic effluents to water (Erdem et al. 2005; Hameed 2009; Chandra et al. 2011; Zainith et al. 2019; Kishor et al. 2019). The untreated wastewater containing colored compounds which have complex structures is difficult to biodegrade. Some dyes used in the textile industries are toxic and carcinogenic, which present eco-toxic hazard and introduce the potential danger of bioaccumulation and also affect humans through the food chain (Babu et al. 2007; Sujata and Bharagava 2016; Kishor et al. 2019). Green synthesized NPs are highly proficient for recycling and removal of heavy metal from wastewaters without loss of their stability and degradation of a variety of organic pollutants from wastewaters and, thus, purify the wastewaters for reuse and recycling and could solve various water quality issues worldwide.

Nickel oxide is considered as good adsorbent due to its chemical and magnetic properties (Nateghi et al. 2012). Rafique et al. (2012) synthesized nano NiO using coprecipitation method and the obtained particles were probed for their adsorption nature toward sulphate and nitrate. It was noted that the adsorbent was effective at pH 7 for the removal of both anions from waters. The synthesized silver nanoparticle also shows heavy metal removal activity in laboratory simulated wastewater. The safety toxicity studies show no significant difference between the orally administered silver nanoparticle-treated water group and control group, while the histopathological studies show well-preserved hepatic architecture for the orally administered silver nanoparticle-treated wastewater group when compared with the control group.

Therefore, it can be concluded that the biosynthesized silver nanoparticles have efficient ability in heavy metal removal without subchronic adverse effects in experimental rats (Shittu and Ihebunna 2017). As water becomes scarcer and more contaminated, the food industry stands to gain much from advanced technology that would provide cleaner and more plentiful water supplies. Nanotechnology has the potential to meet these water-related needs in an inexpensive, efficient, yet flexible way that is crucial for developing nations. Wang et al. (2014) utilized the leaf extracts of green tea and eucalyptus separately for the formation of iron nanoparticles (Fe NPs) and employed for the efficient removal of nitrate from wastewater. Synthesis of spheroidal iron nanoparticles (Fe NPs) was confirmed by employing characterization techniques. A comparison study was conducted between plant-synthesized and chemically synthesized iron materials. Green tea and eucalyptus-mediated Fe NPs were able to remove 59.7% and 41.4% of nitrate from wastewater, respectively, compared to 87.6% and 11.7% removal of nitrate by nZVI and Fe_3O_4 nanoparticles, respectively.

Despite the higher removal efficiency of nZVI, the green synthesized Fe NPs were found to be more stable in nature. Reactivity of aged nZVI, green tea, and eucalyptus synthesized Fe NPs was compared after being completely exposed to air for 2 months. Green tea and eucalyptus synthesized Fe NPs retained the same efficiency of 51.7% and 40.7%, respectively, whereas the efficacy of nZVI significantly dropped about 2.1-fold (45.4%). New study, a green one-pot synthesis of L-serine

(L-Ser) capped magnetite nanoparticles (Fe_3O_4 NPs) and its potential application for adsorption of RhB dye from aqueous solution (Belachew et al. 2017).

13.4 Conclusion and Future Prospective

This chapter highlights the recent advancement in deciphering iron NP synthesis by plant extracts and their applications in water and wastewater treatment. Use of physical and chemical methods for NP synthesis is common but recently several new eco-friendly and economically feasible synthesis techniques are being explored. Green synthesized NPs are being effectively exploited in the area of medical and environmental remediation. Green synthesized NPs and several characterization techniques are currently being utilized in water and wastewater treatment because of their high proficiency and biocompatible nature. Green synthesized NPs are greatly proficient in recycling and eradication of heavy metal contaminants from wastewaters without disrupting their stability, consequently, for purification of wastewaters for reuse and recycling could solve numerous water quality problems globally. Moreover, experiments and problems correlated with the use of green synthesized Fe NPs in water and wastewater treatment are also discussed. However, the enormity of future research scope in this field cannot be accentuated enough.

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