REVIEW ARTICLE

Remediation of Dyes in Water using Green Synthesized Nanoparticles.

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Abstract

With the rapid growth of population and increasing urbanization and industrialization, environmental pollution is becoming a serious concern worldwide. Different type of pollutants is released into the water bodies enormously from the expansive range of industries. Among all the pollutants, dyes are the major used noxious waste discharged by these productions. Even at minute content (< 1ppm), dyes are posing a detrimental threat to the ecosystem and human health risks. Recently, nanotechnology has emerged as an efficient technology for the remediation of environmental pollutants from water. Green synthesis of nanoparticles (NPs) can be done to degrade molecules of dyes in wastewater. Various nanoparticles such as iron, palladium, and cerium dioxide using *Camellia sinensis*, *Boswellta serrata*, and *Azardirachta indica* extracts have been reported successfully for the remediation of various dyes like rhodamine B, methylene blue, etc. Removal of dyes from the wastewater using green synthesized nanoparticles with the help of microbes or plant extracts is a sustainable technique, i.e., by the use of this technique, our environment will not get polluted, and its quality will also be maintained. The present review discusses the classification of dyes, nanoparticle formation by using microbes and plant extract, and, finally, the remediation of dye using such nanoparticles.

Keywords: Dye pollution, Green synthesis, Nanoparticles, Nanotechnology, Remediation.

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Introduction

Along with growing urbanization, industrialization, and rapid advancement of population, environmental pollution, and other crucial difficulties are becoming critical globally. Various types of pollutants discharged from the diverse industries are the major source of pollution (Yang *et al*., 2010; Tang *et al*., 2014; Chen *et al*., 2015; Wu *et al*., 2017; He *et al*., 2018). These industrial pollutants modify the quality of water bodies. Wastewater discharges from chemical industries contain toxic substances such as heavy metal ions, dyes, and organic pollutants. One of the harshest water pollution sources is dye effluent discharge from various industries (Mua and Wang, 2016). A large amount of dyes is produced annually from the different industries such as textile, paint, cosmetic, paper, plastic, leather, agricultural research, hair coloring, photo-electrochemical cell, light-harvesting array, pharmaceutical and nutrition industries. However, the exact estimation of the amount of dyes that are discharged from these industries is a tough task (Khataeea and Kasiri, 2010). Approximately 70,000 tons is the worldwide annual production of dyes, whose commercial types are around 100,000 (Robinson *et al*., 2001). Discharge of dyes into water bodies without proper treatment has caused serious problems to aquatic life and human health (McKay, 1982; Shimada *et al*., 2010; Khan *et al*., 2013).

Dyes are commonly observed to be carcinogenic and mutagenic, as well as pernicious in nature. In the natural environment, dyes retain their color and structural stability as well as show strong resistance to microbial degradation under the exposure to sweat, sunlight, soil and bacteria (Banat *et al*., 1996; Robinson *et al*., 2001; Baban *et al*., 2010; Pathakoti *et al*., 2018). Rate of photosynthesis reduces because these dyes prevent the penetration of light across the water bodies, therefore the level of dissolved oxygen of entire aquatic ecosystem getting affected which cause aesthetic damage to the water bodies (Imran *et al*., 2015; Hassan and Carr, 2018; Lellis *et al*., 2019). When dyes enter into the drinking water system, they impart serious health hazards. Dye effluent contains chemicals that are carcinogenic, mutagenic, or teratogenic, which can cause damage to genetic material in various organisms (Weisburger, 2002; Umbuzeiro *et al*., 2005). In humans, some dyes have been linked to bladder cancer; dysfunction of the kidneys, reproductive system, liver, brain and central nervous system (Medvedev *et al*., 1988; Kadirvelu *et al*., 2003; Dinçer *et al*., 2007; Shen *et al*., 2009). Only 1.0 mg L^{-1} of dye concentration in drinking water could impart a significant color, making it unfit for human consumption (Malik *et al*., 2007). So the remediation of dyes is essential even though at a very minute concentration of theirs is present in the environment.

Dyes have complex structures, stable, non-biodegradable, and remaining ecosystems for a longer time. For their complete degradation, there is a constant need to have an effective

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method that can efficiently remove these dyes from wastewater. However, such a method for dye removal has remained a challenge for scientists because conventional methods for dye removal were not very efficient and economical. Many conventional treatment methods applied for the remediation of dyes from wastewater include physical decolorization (sedimentation, filtration adsorption, and reverse osmosis), chemical decolorization (neutralization, recovery, chemical oxidation, ion exchange methods), and biological decolorization by using bacteria, fungi, and actinomyces (Morshedi *et al.*, 2013). The drawback of physical methods is high operational cost, while drawback for the chemical method is the production of a concentrated sludge (Robinson *et al.*, 2002). The biological method takes a long time for degradation, and for some dyes, this method has a low degradation efficiency. Moreover, the management and start-up of biological techniques is a tricky process (Lin *et al.*, 2008; Shan *et al.*, 2008; Yadav *et al.*, 2012). Consequently, there is an urgently needed technique for dye remediation, which should be cost-effective, eco-friendly, and have high removal efficiency for decolorization of wastewater discharges from the various industries.

Nanotechnology is an emerging field that is being used in several applications to improve the quality of the environment. It is a field of applied science that is concerned with materials and systems whose structure and components exhibit novel and significantly improved biological, chemical, and physical properties, owing to their nanosized structure. Generally, nanotechnology refers to materials of size 100 nanometer (nm) or smaller in at least one dimension, and it involves the development of materials or devices in this size range (Rawtani *et al.*, 2018). Various nanomaterials, such as a nanoparticle, nanofiber, nanotubes, nanowire, nanorods, nanoribbon, etc. are used for remedial techniques. But out of these, nanoparticles are of great scientific interest for the remediation because of their small size and relatively large reactive surface area. Different methods such as physical, chemical, biological, and hybrid processes are used to synthesize myriad nanoparticles (Pandey *et al.*, 2016; Rawtani *et al.*, 2017; Tharmavaram *et al.*, 2017, 2018; Rawtani *et al.*, 2019). However, physical methods are too expensive, while chemical methods create some adverse effects on our environment and are very harmful to living organisms as well as human health (Panigrahi *et al.*, 2004; Narayanan and Sakthivel, 2010; Thakkar *et al.*, 2010). These methods are not much suitable for wide-scale production because of demerits such as high preparation costs, consumption of extraordinary energy, use of hazardous organic solvents, production of hazardous intermediates, and harmful waste products, which leads to environmental pollution and several biological risks. During the formation of nanoparticles, aggregation of particles occurs due to attractive forces between the nanoparticles. Consequently, there is a requirement to add some capping agents to prevent aggregation and attain the uniformity of the product. All these incidents are responsible for the necessity of improved or appropriate alternative technology, which is consistently good for the development of nanoparticles and also is environmentally friendly. For the remediation of different pollutants from the environment, green technology is recommended as the superior-most technique because in this technique biogenic substances or natural substances such as plants or microbes are used as a reducer and capping agents. Nanoparticles derived from green synthesis using microbe or plant extract have no toxic substances, and their by-products are also eco-friendly. So as far as remedial processes are concerned, it is evident that green synthesis of nanoparticles is the best technique.

In this regard, this review focuses on the recent developments for the remediation of dyes from industrial wastewater by using nanoparticles, which are synthesized by greener routes involving extract of plants, microbes, and other natural products. Previous studies majorly focused on the remediation of various dyes using nanoparticles synthesized by physical and chemical methods. The novelty of present review summarizes the classification of dyes of various aspects such as on their sources, attached chromophore and their applications in various sectors, synthesis of nanoparticles with the help of green technology, and these nanoparticles applied for the remediation of various dyes which was not focused by the past reviews.

Categorization of Dyes

Organic compounds which are have colored substances called as dyes. When these colored substances or dyes applied to some substrates, these colored substances get tied up or bind to these substrates chemically and impart permanent color to these substrates. Any type of substrate such as fur, hair, paper, plastics, cosmetics, textiles is used for the coloration from dyes. The imparted color is not removed by washing with water, detergents, or exposure to light. Dyes are used by diverse industries such as plastic, paint, textiles, food, etc. (Chincholi *et al.*, 2014; Yagub *et al.*, 2014). Various types of manner occur for the categorization of dyes such as categorized by the material source, i.e., dyes obtained from which kind of source, naturally or synthetically; categorization by the attached chromophores; and categorization by application (Fig. 1).

Categorization by the Material Source

This type of categorization is very familiar. According to this categorization, dyes are of two types: one is a natural dye, and the other is a synthetic dye.

Natural dyes

These dyes or colorants are derived from plants, animals, and minerals. As the name suggests, natural dyes are derived naturally from diverse plants or plant parts (wood, root, bark,

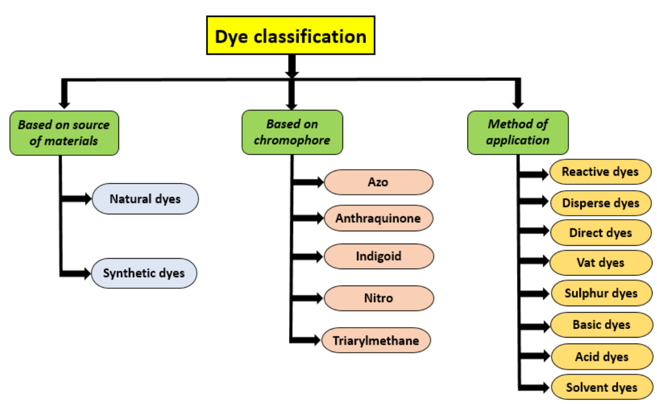

Fig. 1: Dye classification

and stem), fungi, and lichens. Some minerals (*sangraj*, *lajerd*, *gem*, *sindur*, and *sajeda*) are also used as natural dyes.

Synthetic dyes

Now a day, synthetic dyes are used extensively in almost all places. The production of synthetic dyes is economical, as well as they are very bright in color, and their application to textile is uncomplicated. These reasons make synthetic dye's usage to be ubiquitous. Classes of synthetic dyes are acid dyes, azo dyes, basic dyes, and mordant dyes.

Categorization by the Attached Chromophore

Dyes are categorized by chromophores, which are found in the structure of dyes.

Azo chromophore

Azo dyes have gained the most significant among all the dyes available. They are extensively useful in industries such as textiles, food, and leather. In the structure of these dyes, the azo group is present, which is normally named as azo. Sometimes, the dye may also contain two, three, four, and more azo groups, which are named as disazo, trisazo, tetrakisazo, and polyazo, respectively. Azo dyes can supply yellow/red and blue/brown dyes mostly. Various azo dyes are solvent yellow 14, disperse red 13, disperse blue, reactive brown 1, acid black 1, direct green 26, and direct black 19.

Anthraquinone chromophore

This class of dyes is the second most significant of all the dyes. Some dyes of anthraquinone are very oldest, and they were used in the process of mummification. Almost all the major natural red dyes are anthraquinones (Gordon and Gregor, 1983). Anthraquinone group, which is generally colorless, is present in the ring structure, and their position defines the anthraquinone dyes. When these dyes are used commercially, some amino or hydroxyl groups are added in the ring structure at α position. Anthraquinone dye imparts the combined properties, and generally, these dyes are used for the shades of red and blue.

Indigoid chromophore

For the coloration of different textiles such as cotton and wool indigoid, dyes are very utilitarian. Many indigoid dyes have been synthesized using only indigo. These dyes are used mainly for denim jeans and also jackets for the coloration. Various expensive, branded, and luxurious clothes were dyed-through these dyes. Some dyes of this class were so costly that poor community was not capable of affording garments that were dyed with these dyes.

Nitroso and nitro dyes

Nitroso dyes are those compounds which carry chromophore named as nitroso and -OH as auxochrome. This nitroso group involved in a carbon or nitrogen atom. Sometimes, this nitroso group in some substances gets involved with an oxygen atom, and then named as nitrites, while sometimes called nitrosyls when involved with a metal ion. The molecules of these dyes were perfect for the penetration of polyester fibers, for example, disperse dyes. These dyes are also very useful for the coloration of papers, for example: Acid Green 1.

Triarylmethane dyes

Triarylmethane dyes were produced synthetically and derived from the triphenylmethane. Auxochromes (amino, hydroxyl) present in these dye are responsible for their deep color. Some examples of these are methyl violet, malachite green, and phenol dyes.

Categorization by the Application

Reactive dyes

The reactive group is found in the structure of reactive dyes; that is why these dyes are named as reactive dyes. These are the only dyes that carry the reactive groups in their structure, and this reactive group is responsible for the establishment of a covalent bond between the dye molecule and respective fiber such as cotton, wool, and nylon.

Disperse dyes

The solubility of disperse dyes in water is very low, and are sometimes called insoluble or non-ionic dyes. Polyesters, nylon, acrylic are the target fibers for dyeing with these dyes. The requirements at the process of dying are that these dyes need a dispersing agent, high temperature, and acidic condition. Disperse dyes are derived from nitro, anthraquinone, and azo groups.

Direct dyes

Generally, direct dyes are also termed as substantive dyes, i.e., these dyes are attracted to the textiles by some physical forces. Substantivity is quantified by the proportion or the degree of attraction that occurs between the molecules of dyes and the textile. These dyes are soluble in water, and during the process of dying, these dyes need an alkaline medium.

Vat dyes

Cellulose fibers are the target fibers for dyeing by using vat dyes. Vat dyes are poorly dissolved in water, but for the dyeing process, it needs to be in its soluble form, which is attained by the aid of reducing agents (e.g., sodium dithionite). After attaining its soluble form, they attached to the respective fiber and imparted the strong color to the fabric.

Sulfur dyes

Like vat dyes, sulfur dyes are also water-insoluble, and these dyes also require reducing agents (sodium sulfide, sodium hydrosulfide) for attaining their soluble form. During the process of dying, the reducing agent aid in the dissolution of dye particles and facilitate the absorption of dyes into the fabric.

Cationic (basic) dyes

These dyes are also named as cationic dyes. The reason behind the name of cationic dye is the color production. At the time of ionization of the salts of organic bases, the positively charged cation and negatively charged anion are produced, and the produced cation is responsible for the production of color. The uses of basic dyes are mainly in the textile industry, where they are employed to color the fabrics. The target fibers of these dyes are wool, silk, cells/tissues of humans (e.g., safranin and crystal violet).

Acid dyes

The solubility of acid dyes is very high in water. The target fibers are silk, wool, and protein. Various types of bonds (van der Waals, ionic, and hydrogen) are established by the molecules of dyes with the respective fibers.

Solvent dyes

Solvent dyes are those dyes which do not solubilize in water and are much soluble in organic solvents. The uses of solvent dyes are that they utilize the coloration of organic solvent, waxes, fuels, plastics, and glass. Most of the solvent dyes are azo dyes, e.g., red and yellow dyes, and anthraquinone dyes, e.g., green and blue.

Green Synthesis of Nanoparticles

Nanoparticles can be synthesized by various methods that are categorized into two approaches: top to down and bottom to up (Figs 2 and 3). By the employment of the top-down approach, the nanoparticle is synthesized by the reduction of size from minute particles, and these minute particles are formed from the bulk material. Different techniques utilized in the nanoparticle formation through this approach include arc discharge, pulsed laser ablation, spray pyrolysis, evaporation-condensation, and lithography (Rafique *et al*., 2017). These are the physical methods, where physical forces are involved in the attraction of nanoscale particles and the formation of a large, stable, well-defined nanostructure. Limitations of the physical method involve the use of expensive equipment for the synthesis, high temperature, and pressure (Chandrasekaran *et al*., 2016), large space area for setting up of instruments, defective surface formation, and low production rate.

By the employment of the bottom-up approach, nanoparticles are obtained by the use of chemical and biological methods. Under this approach, atoms clump and form clusters or new nuclei, which at last develop into a nanoparticle. Different techniques occur in the nanoparticle synthesis by chemical methods such as solvothermal, pyrolysis, co-precipitation, sonochemical, and electrochemical (Ealias and Saravanakumar, 2017). Chemical synthesis methods are not eco-friendly and involve the usage of toxic chemicals, formation of hazardous by-products, which create biological risk and contamination from precursor chemicals (Thakkar *et al*., 2010; Vijayan *et al*., 2016). On the whole, the conclusion is that these conventional methods for nanoparticle synthesis have certain drawbacks at the time of the fabrication process of nanoparticles. In addition, the major limitation is when these synthesized nanoparticles are applied to certain fields like medical, agriculture, where they create toxicity and alter the quality of our ecosystem (Ahmed *et al*., 2016). Consequently, there is a growing exigency to establish clean, non-toxic, and environment-friendly procedures for nanoparticle synthesis.

Nanoparticle synthesis by green route is fascinating all investigators because the use of the green route for the generation of nanoparticles has overcome all the downsides or limits which come when the chemical or physical method is adopted for the development of nanoparticles. In the chemical methods, chemicals which were used in the process of nanoparticle synthesis were too expensive, and also, they are toxic as well as their by-products are also very hazardous, but if we select the green route for nanoparticle genesis, this problem is controlled because, in the development of nanoparticles through green route, the process is completed by the use of non-toxic natural products. Physical methods of nanoparticle synthesis consume more energy, which was also not good, and that can also be overcome by green synthesis. The generation of nanoparticles by the green route method or biological method adopts the bottom-up approach. In the process of synthesizing nanoparticles from the biological method, three most important selections are required: choice of the solvent medium which is for the development of nanoparticles; choice of environment-friendly reducer; and choice of stabilizer agent which acts as capping agent (Narayanan and Sakthivel, 2011; Singh *et al*., 2011).

For the production of advanced nanoparticles, nature has provided many ways, and sometimes these biogenic or natural products which are helpful in the generation of nanoparticles are termed as the laboratory of the natural products especially in the fabrication of metallic and metal oxide nanoparticles (Sharma *et al*., 2019). The biological approach includes different types of microorganisms such as bacteria (Shivaji *et al*., 2010), fungi (Chan and Mat Don, 2013), yeast (Kumar *et al*., 2011), and plant extract (Akhtar *et al*., 2013) (Fig. 4).

Nanoparticle Synthesis using Microorganisms

In the generation or synthesis of diverse type of nanoparticles, different microbes have acted as utilitarian agents in the development of desirably sized nanoparticles. Different microbes are used in the process of nanoparticle genesis due to their simplicity to work, medium for the evolution of microbes is cheap, and they are maintainable. In previous years, the synthesis of nanoparticles using microbes has enlarged comprehensively due to its immense application. The generation of metallic nanoparticles through microbes is an appropriate approach. Gold, silver, and cadmium sulfide nanoparticles are extensively

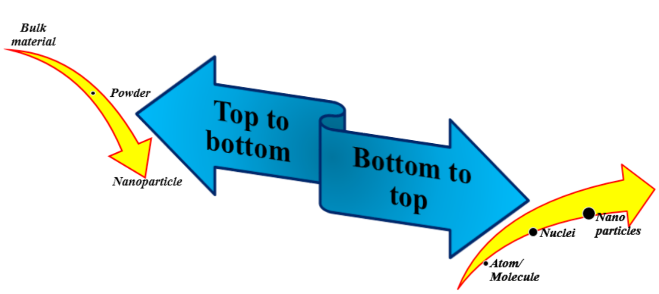

Fig. 2: Approaches for nanoparticle synthesis

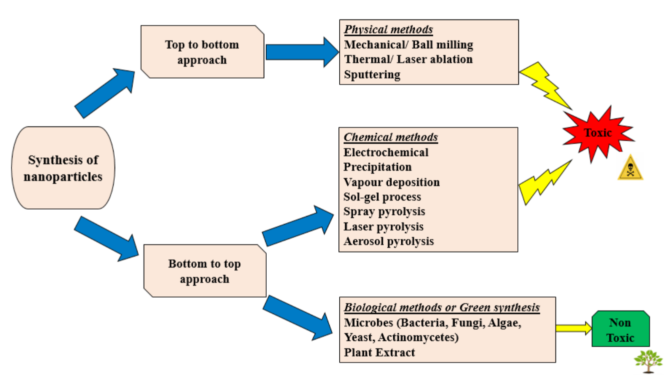

Fig. 3: Synthesis of nanoparticles by various methods

Fig. 4: Advantages of green synthesized nanoparticles

synthesized by microbial cells as these cells are considered a potential bio-factories for these nanoparticles fabrication. Various biomolecules (enzymes, vitamins, polysaccharides, amino acids) perform as a capping agent and reducing agent for the generation of nanoparticles. These biomolecules can be obtained from the extracts of microbes. It seems that microbes play a role in providing the nucleation centers and establish conditions for obtaining highly disperse nanoparticle systems. Microbes prevent aggregation and have the potential to immobilize nanoparticles by providing a viscous medium.

The precise mechanism behind the construction of nanoparticles via microbes is not known up until now because, for the duration of the fabrication of nanoparticles, diverse microbes respond in a different way. Inorganic materials are produced by microbes either intracellularly or extracellularly, but the mechanisms are different with different microbial cells. For the intercellular genesis of nanoparticles, the cell wall of microorganisms performs an extensive character. Microbes possess enzymes in the cell wall, which help in the metal ion reduction, and these cations get fixed with the negatively charged cell wall. This is the intercellular manner of development of nanoparticle. Fungi also secrete the enzyme named as nitrate reductase, which promotes the metal ion reduction and helps the formation of nanoparticles extracellularly (Joerger *et al.*, 2001; Nair and Pradeep 2002; Durán *et al.*, 2005; He *et al.*, 2007; Kumar *et al.*, 2007a,b; Ingle *et al.*, 2008).

First of all, the nucleation of clusters in the bacterial cell stake place for the generation of nanoparticles (Shivaji *et al.*, 2010). In the second step of the development of nanoparticles, the bacterial cell and the clusters of metal bind electrostatically to each other. After binding, the formed nanoparticles travel into the bacterial cell wall. Several bacterial species are reported for the fabrication of silver nanoparticles, such as *Aeromonas* sp. (Fu *et al.*, 2006), *Enterobacter cloacae* (Shahverdi *et al.*, 2007), *Bacillus subtilis* (Saifuddin *et al.*, 2009), *Pseudomonas stutzeri* AG259 (Klaus *et al.*, 1999), *Proteus mirabilis* (Nasrin *et al.*, 2009), *Cornebacterium* sp. (Huang *et al.*, 2007), *Plectonema boryanum* (Lengke *et al.*, 2007), and *Lactobacillus* sp. (Armendariz *et al.*, 2004). Regarding the fabrication of gold nanoparticles, some reported bacterial species are *Rhodo pseudomonas capsulate* (Bai *et al.*, 2009), *Marinobacter Pelagius* sp. (Joerger *et al.*, 2000), *Lactobacillus* sp. (Tom *et al.*, 2003), *Pseudomonas aeruginosa* (Nayantara and Kaur, 2018), *Stenotrophomonas malophilia* (Sharma *et al.*, 2012), *E. coli* K12 (Srivastava *et al.*, 2013), *Geobacillus* sp. strain ID17 (Narayanan and Sakthivel, 2008), *Thermomonospora* sp. (Kasthuri *et al.*, 2008), *Rhodococcus* sp. (Park *et al.*, 2011), and *Delftia acidovorans* (Johnston *et al.*, 2013).

Some other nanoparticles; synthesis has also been reported in previous literature by using bacterial strains such as cadmium sulphide (CdS NPs) nanoparticles using *Klebsiella aerogenes* (Holmes *et al.*, 1995), *Escherichia coli* (Sweeney *et al.*, 2004), *Rhodobacter sphaeroides* (Bai *et al.*, 2009); Palladium (Pd) nanoparticles by *Clostridium butyricum*, *Citrobacter braakii*, *Klebsiella pneumoniae*, *Enterococcus faecium*, *Escherichia coli*, *Bacteroides vulgatus* (Hennebel *et al.*, 2011), and *Desulfovibrio desulfuricans* (Yong *et al.*, 2002). *Serratia* sp. for copper oxide (CuO, Cu_2O, Cu_4O_3) nanoparticles, *Actinobacters* spp. for iron oxide (Fe_3O_4) nanoparticles, *Lactobacilli* for Titanium dioxide (TiO_2) nanoparticles, *Actinobacters* spp. for silicon/silica nanoparticles, *Shewanella putrefaciens* for uranium dioxide (UO_2) nanoparticles, *Lactobacillus sporoge* for zinc oxide (ZnO) nanoparticles (Durán and Seabra, 2012). Selenium and tellurium nanoparticles have been synthesized by *Stenotrophomonas maltophilia* and *Ochrobactrum* sp. (Zonaro *et al.*, 2015).

Several species of fungi have been reported successful in the previous studies, and it has been observed that fungal cells assist in fabricating monodispersed nanoparticles. Fungal cells are rich sources of enzymes, which make them a tremendous contender for the production of nanoparticles. As compared to bacteria, fungi synthesize a great amount of nanoparticles because fungi secrete more amount of proteins, which leads to higher productivity of nanoparticles. Various fungal species were used in the production of silver (Ag) nanoparticles, such as *Verticillium* spp. (Mukherjee *et al.*, 2001), *Aspergillus fumigatus* (Bhainsa and D'souza, 2006), *Aspergillus flavus* (Vigneshwaran *et al.*, 2007), white-rot fungi (Gudikandula *et al.*, 2017), *Aspergillus terreus* (Li *et al.*, 2012), *Fusarium oxysporum* (Ahmad *et al.*, 2003c), *Humicola* sp. (Syed *et al.*, 2013), *Macrophomina phaseolina* (Chowdhury *et al.*, 2014). Gold nanoparticles (AuNPs) have been synthesized by *Fusarium oxysporum* (Mukherjee *et al.* 2002), *Collitotrichum* sp. (Shankar *et al.*, 2003), *Trichothecium* sp. (Ahmad *et al.*, 2005), *Verticillium luteoalbum* (Erasmus *et al.*, 2014), and *Aspergillusoryzae* var. *viridis* (Binupriya *et al.*, 2010). *Fusarium oxysporum* fungal species are also used in some other nanoparticles' synthesis such as cadmium sulfide (Ahmad *et al.*, 2002), silica, titanium (Bansal *et al.*, 2005), cadmium selenide (CdSe) (Kumar *et al.*, 2007a), zinc oxide (Baskar *et al.*, 2017), zirconia (Bansal *et al.*, 2004), bismuth oxide (Bi_2O_3). Apart from this, nanoparticles of iron oxide (Fe_3O_4) from *Fusarium oxysporum* and *Verticillium* sp., antimony trioxide (Sb_2O_3) from *Saccharomyces cerevisiae* (Durán and Seabra, 2012), magnetite from the species of *Fusarium oxysporum* and *Verticillium* sp. (Bharde *et al.*, 2006).

Algae is also used in the biosynthesis of nanoparticles because algae have the ability to accumulate heavy metals. Algae is very beneficial in multiple fields because a kind of polysaccharide named fucoidans, which is secreted from the cell wall of algae, is very advantageous since it has properties such as anti-cancerous, anti-viral agent, slow aging agent, and anti-inflammatory. Metabolites are present in the algal cells, which perform the role of stabilizing and reducing agents for attaining the size of NPs at the range of nanometer. Various algal species were reported successfully in the generation of diverse nanoparticles. Silver (Ag) NPs were produced by *Chaetomorpha linum* (Kannan *et al*., 2013), *Pterocladia capillacae, Jania rubins, Ulva faciata*, and *Colpmenia sinusa* (El-Rafie *et al*., 2013), *Hypnea musciformis* (Selvam and Sivakumar, 2015), and *Enteromorpha flexuosa* (Yousefzadi *et al*., 2014). *Sargassum muticum* was used in the production of gold (Au) NPs (Namvar *et al*., 2015), *Tetraselmis kochinensis* (Senapati *et al*., 2012), *Ecklonia cava* (Ghodake and Lee, 2011), *Chlorella vulgaris* (Annamalai and Nallamuthu, 2015), *Padina gymnospora* (Singh *et al*., 2013), and *Fucus vesiculosus* (Mata *et al*., 2009). *Sargassum myriocystum, Caulerpa peltata,* and *Hypnea valencia* were used in the production of zinc oxide (ZnO) NPs (Nagarajan *et al*., 2013), *Gracilaria gracilis* (Francavilla *et al*., 2014). Ferric oxide (Fe_3O_4) NPs have been synthesized by *Sargassum muticum* (Mahdavi *et al*., 2013), copper oxide NPs by *Bifurcaria bifurcate* (Abboud *et al*., 2014), cadmium sulfide NPs by *Phaeodactylum tricornutum* (Scarano and Morelli, 2003), and ferrihydrite NPs by *Euglena gracilis* (Brayner *et al*., 2012).

In the fabrication process of various types of nanoparticles, actinomycetes are also a very fabulous candidate because they have the characters of both fungi and bacteria, and also the genetic modification is very facile for the development of desirably sized nanoparticles. Metallic nanoparticles are produced in large numbers with the aid of actinomycetes. Gold nanoparticles were synthesized by *Thermomonospora* sp. (Ahmad *et al*., 2003b) and *Rhodococcus* sp. (Ahmad *et al*., 2003a). *Streptomyces hygroscopicus* (Husseiny *et al*., 2007), *Gordonia amarae* (Montes *et al*., 2011), *Gordonia amicalis* (Baker and Satish, 2015), *Streptomyces fulvissimus* (Balagurunathan *et al*., 2011), *Streptomyces* sp. (Meysam *et al*., 2015), and *Streptomyces viridogens* (Kumar *et al*., 2011). Actinomycetes such as *Streptomyces* sp., *Pilimeliacolu mellifera*, and *Rhodococcus* sp. have been used in the development of silver (Ag) nanoparticles (Patrycja *et al*., 2016). Various species of yeast are also used in nanoparticle synthesis. Some reported literature for gold nanoparticles is *Pichia jadinii* (Gericke and Pinches, 2006), *Yarrowia lipolytica* 3589 (Ganesh Babu and Gunasekaran, 2009), *Hansenula anomala* (Waghmare *et al*., 2014), and *Candida guilliermondii* (Tripathi *et al*., 2014), Magnusiomycesingens (Venkatesan *et al*., 2014). Cadmium sulfide nanoparticles synthesized by *Candida glabarata,* and *Schizosaccaromyces pombe* (Agnihotri *et al*., 2009). Amorphous iron phosphate NPs by *Saccharomyces cerevisiae* (He *et al*., 2009). *Saccharomyces cerevisiae* was also used in titanium dioxide nanoparticle (Jha *et al*., 2009a) and antimony trioxide (Sb_2O_3) nanoparticles'synthesis (Jha *et al*., 2009b).

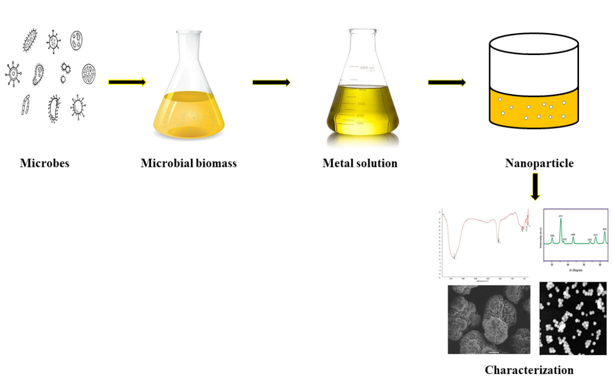

Fig. 5: Nanoparticle synthesis using microbes

Nanoparticle Synthesis using Plant Extract

The synthesis of plant-based nanoparticles is a simple one-step process. In the plant-based nanoparticle fabrication, a wide range of green reducing and capping agents are used, which can be cost-effective, biocompatible, non-hazardous, and eco-friendly. Biomolecules and the existence of functional groups in the extracts of plant aid in the generation of nanoparticles because during the synthesis process, they perform the role of capping and reducing agents.

As compare to microbes, plant extract could be an efficient approach according to previous reports (Iravani, 2011) because the plant-based synthesis of nanoparticles produces highly stabilized nanoparticles in a single step process and in a very short duration.

Silver nanoparticles were synthesized from the solution of silver nitrate with the help of the various type of plant extracts such as *Alternanthera dentate* leaf extract (Kumar *et al*., 2014), *Acorus calamus* rhizome (Nakkala *et al*., 2014), tea extract (Suna *et al*., 2014), *Vitis vinifera* fruit extract (Gnanajobitha *et al*., 2013), *Salvadora persica* stem extract (Tahir *et al*., 2015), Vasaka (*Justicia adhatoda* L.) leaf extract (Bose and Chatterjee, 2015), and beetroot extract (Bindhu and Umadevi, 2015). Gold nanoparticles have been synthesized by *Coleus amboinicus*, *Dillenia indica* fruit extract, tuber extract of *Dioscorea bulbifera*, and leaf extract of *Euphorbia hirta*, *Zingiber officinale*, *Mentha piperita*. Zinc oxide nanoparticles have been synthesized by various plant parts like leaf, stem, root, fruit, and seed. Bio-reduction involves reducing metal ions or metal oxides to 0-valence metal NPs. Other fabulous plant species also reported successfully such as *Calatropis gigantean, Plectranthus amboinicus*, *Agathosma betulina, Vitex negundo*, *Nephelium lappaceum, Azadirachta indica, Moringa oleifera, Plectranthus amboinicus*, and *Anisochilus carnosus* (Agarwal *et al*., 2017; Kumar *et al*., 2017; Nadeem *et al*., 2017).

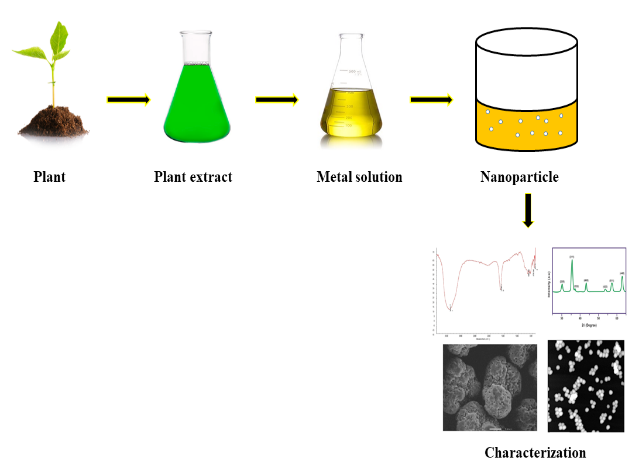

Fig. 6: Nanoparticle synthesis using plant extract

Remediation of Dyes by Green Synthesized Nanoparticles

According to Barizãoa *et al*. (2020), tartrazine and Bordeaux red dye can be removed by iron oxide nanoparticles using two agro-industry residues: *Cucurbita moschata* leaves and *Beta vulgaris* stalks. The synthesized nanoparticles with an estimated diameter of 2 and 20 nm, were observed by diffraction peaks in X-ray Diffraction (XRD). In the adsorption process for dye, equilibrium was obtained after 1,200 minutes for tartrazine and 240 minutes for Bordeaux red when *Cucurbita moschata* leaves were applied in the dye solution. When *Beta vulgaris* stalks were used, equilibrium reached after 180 minutes for tartrazine and 120 minutes for Bordeaux red. Kouhbanani *et al*. (2019) also reported a successful generation of iron oxide nanoparticles. These nanoparticles were synthesized using aqueous leaf extract of *Teucrium polium* in size range of 5.68 to 30.29 nm, which was characterized using particle size analysis (PSA). The green synthesized iron oxide nanoparticles IONPs were able to decolorize methyl orange dye with 73.6% efficiency in a 6-hour reaction. The maximum rate of methyl orange degradation occurred with IONPs catalyzed H_2O_2 after 6-hour with 73.6% efficiency. Decolorization of methyl dye became possible due to the combination of IONPs and H_2O_2. This combination caused the release of free hydroxyl radical (OH·) that attacked and cleaved the azo bond (-N=N-) found in the methyl orange, which led to the decolorization of dye solution (Kouhbanani *et al*., 2019). Another study of dye decolorization using iron oxide nanoparticles (Fe_2O_3NPs) was reported by Bibi *et al*., in which the nanoparticles were successfully fabricated via green route using pomegranate (*Punica granatum*) seeds extract, which were confirmed by UV-Vis., XRD, EDX, SEM, and AFM techniques. The shape (semi-spherical) and size (25–55 nm) of produced nanoparticles was confirmed with the aid of scanning electron microscopy (SEM). Fe_2O_3 NPs showed excellent photocatalytic activity against reactive blue under UV light irradiation, and maximum degradation of 95.08% was achieved with 56 minutes of reaction time. It was observed that the absorption peak of dye decreased rapidly as a function of UV irradiation time, which was due to the breakdown of the chromophore group in the dye. The dye was degraded up to 95.08% in 56 minutes of reaction time (Bibia *et al*., 2019). According to Ismail *et al*. (2019), *Duranta erecta*, flowering shrub's fruit extract, was used in the synthesis of copper nanoparticles. In the existence of $NaBH_4$, these nanoparticles played a great role in discoloration of methyl orange and congo red dye. The reduction of both dyes followed the pseudo-first-order reaction. In the presence of $NaBH_4$ and CuNPs, the reduction of methyl orange achieved 96% in 4 minutes and 90.35% reduction of congo red in 5 minutes. This CuNPs reusability was checked four times for methyl orange. For the first time, the degradation rate achieved was 96% in 4 minutes, then for the second time, it took 6 minutes for the 95% degradation, and 10 and 15 minutes for 95% in the third and fourth time (Ismail *et al*., 2019).

Fatimah *et al*. (2020) fabricated iron oxide (Fe_3O_4 and Fe_2O_3) nanoparticle (10–80 nm) by using *Parkia speciosa* Hassk pod extract. These nanoparticles contained magnetic property as well as showed a great reduction rate (98%) of bromophenol blue dye under both UV and visible light exposure. The photocatalytic reduction rate of bromophenol blue was affected by the formation of hydroxyl radical, which formed after the addition of H_2O_2. The addition of H_2O_2 significantly accelerated the degradation rate, and faster degradation occurred under UV light as compared with that under visible light. In the research work of Kolya *et al*. (2015), silver (Ag) nanoparticles (11–15 nm) were produced by the use of leaf extract of *Amaranthus gangeticus* Linn through the solution of silver nitrate ($AgNO_3$). The produced silver (Ag) nanoparticles exhibited great degradation towards congo red dye. Silver nanoparticles were also synthesized using an extract of *Clitoria ternatea* pods with an average size of 62.51 nm. Methylene blue dye degraded in the presence of $NaBH_4$ with the aid of green synthesized silver nanoparticles. The decolorization of dye occurred from blue color to completely vanish within 18 minutes. For the degradation process, silver nanoparticles help in transferring the electron from BH_4^- to methylene blue dye (Varadavenkatesan *et al*., 2019).

As per Wang *et al*. (2018), *Klebsiella oxytoca* GS-4-08 is an anaerobic bacteria that was used in the generation of palladium nanoparticles (5–20 nm) in the existence of glucose. These bio-Pd nanoparticles were efficient for the removal of azo dyes (methyl orange, acid blue 113, reactive black 5, and acid red 1). The reduction efficiency was about 96.54 ± 0.23% in 24 hours. The enhancement of the reduction rate of azo dyes obtained by the use of anthraquinone-2-disulfonate (AQS). The reduction rate in the presence of AQS achieved at 68.55 ± 0.21% only in 2 hours, while in the absence of AQS, the reduction rate was only 58.35 ± 0.45%. According to Kora and Rastogi (2016), Palladium nanoparticles are also produced from palladium chloride ($PdCl_2$) via gum olibanum (*Boswellia serrata*) with an average size of about 6.6 nm. By the use of these nanoparticles, the reduction in the concentration of the synthetic dyes such as rhodamine B, coomassie brilliant blue G-250, and methylene blue with $NaBH_4$ was observed. The whole reduction process completed within 2 minutes, with a color change from yellow to colorless.

According to Ismail *et al*. (2018), silver nanoparticles were synthesized by taro (*Colocasi aesculenta*) plant rhizome powder with a mean size of about 68 ± 12 nm. These nanoparticles showed high degradation efficiency towards the organic azo dyes such as methyl orange (MO), congo red (CR), methyl red (MR), and rhodamine B (RhB) by $NaBH_4$. 100% degradation of methyl orange was achieved in the presence of $NaBH_4$ with green synthesized silver nanoparticles only in 7 minutes. The discoloration of congo red (96.9%)was achieved in 12 minutes by $NaBH_4$ in the presence of active catalysts silver (Ag) nanoparticles. Sodium borohydride in the presence of catalyst reduced congo red molecule at the azo sites (-N=N-) by producing hydrazine derivative compounds. Similarly, 96.29% of methyl red reduction took 14 minutes, and 97.78% reduction of rhodamine B took just 6 minutes. The prepared silver nanoparticles also showed a great reduction of the mixtures of dyes. Complete reduction obtained of the mixture of methyl orange and methyl red only in 9 and 10 minutes for the mixture of methyl orange, methyl red, and congo red. The general hypothesis behind nanoparticle fabrication and dye removal is shown in Fig. 7. Some other literature works have been shown in Table 1.

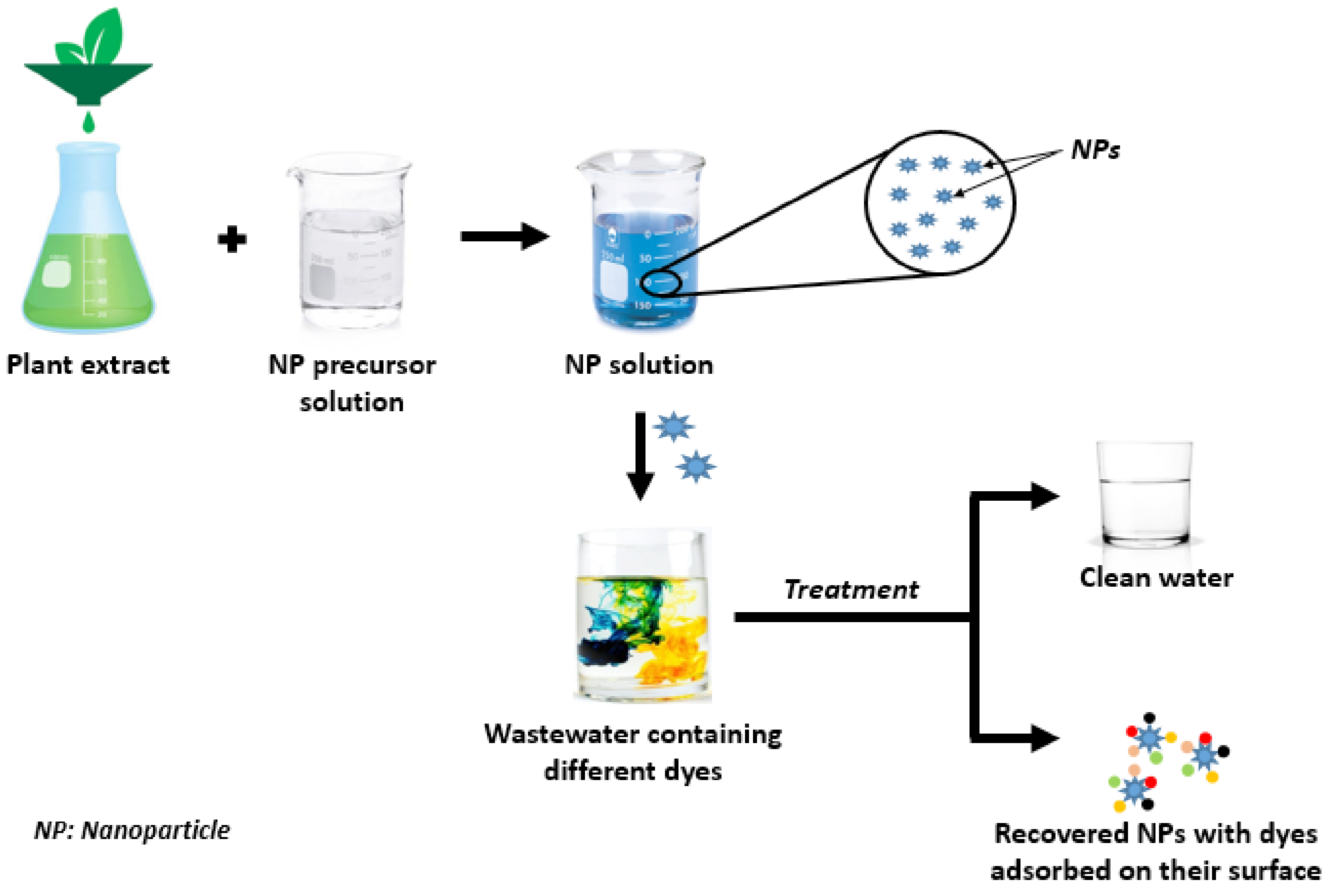

Fig. 7: Hypothesis behind synthesis of nanoparticle and dye remediation

Table 1: Green synthesized nanoparticles for dye remediation

S. No.	*Plant extract/ microorganism*	*Nanoparticle (NPs)*	*Characterization techniques*	*Size/ diameter of NPs (nm)*	*Targeted dye*	*References*
1	*Cucurbita moschata* leaves	Iron oxide	UV-VIS	2–20	Tartrazine	Barizãoa *et al.* (2020)
	Beta vulgaris stalks		FTIR		Bordeaux red	
			TEM			
			XRD			
2	Leaf extract of *Teucrium polium*	Iron oxide	TEM	5.68–30.29	Methyl orange	Kouhbanani *et al.* (2019)
			PSA			
			XRD			
			FTIR			
			VSM			
			TGA			
3	*Punica granatum* seeds extract	Iron oxide	UV-VIS	25–55	Reactive blue	Bibia *et al.* (2019)
			XRD			
			EDX			
			SEM			
			AFM			
4	Fruit extract of *Duranta erecta*	Copper	UV-VIS	70	Methyl orange	Ismail *et al.* (2019)
			XRD		Congo red	
			EDX			

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			FTIR			
			SEM			
5	*Parkia speciosa Hassk* pod extract	Iron	XRD	10–80	Bromophenol blue	Fatimah *et al*. (2020)
			SEM			
			TEM			
6	Leaf extract of *Amarranthus gangeticus*	Silver	HR-TEM	11–15	Congo red	Kolya *et al*. (2015)
			SAED			
			UV-VIS			
			FTIR			
7	Extract of *Clitoria ternatea* pods	Silver	UV-VIS	62.51	Methylene blue	Varadavenkatesan *et al*. (2019)
			SEM			
			XRD			
			FTIR			
			EDX			
8	*Klebsiella oxytoca* GS-4-08	Palladium	TEM	5.20	Azo dyes	Wang *et al*. (2018)
			XRD			
9	*Boswellia serrata*	Palladium	UV-VIS	6.6 ± 1.5	Coomassie brilliant blue G-250	Kora and Rastogi (2016)
			DLS		Rhodamine B	
			TEM		Methylene blue	
			XRD			
			FTIR			
10	Taro (*Colocasia esculenta*) plant rhizome powder	Silver	SEM	68 ± 12	Methyl orange	Ismail *et al*. (2018)
			EDX		Congo red	
			XPS		Methyl red	
			XRD		Rhodamine B	
11	*Camellia sinensis* tea extract	Iron	XRF	20–100	Methylene blue	Carvalho and Carvalho (2017)
			TGA		Methyl orange	
			TEM		Bromothymol blue	
12	Palm dates fruit	Silver-iron bimetallic NPs	UV-VIS	5–40	Bromothymol blue	Al-Asfar *et al*. (2018)
			TEM			
			EDX			
13	Extracts of green tea leaves	Iron	TEM	40–60	Methylene blue	Shahwana *et al*. (2011)
			SEM		Methyl orange	
			EDX			
			XRD			
			FTIR			
14	extract of *Cupressus sempervirens*	Iron	TEM	19	Methyl orange	Ebrahiminezhad *et al*. (2017)
			FTIR			
			UV-VIS			
			XRD			

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15	Green tea	Iron	TEM		Remazol brilliant Blue R	Truskewycza *et al.* (2016)
			SEM		Direct red 80	
			EDX			
			FTIR			
			Zeta potential			
16	Green tea	Iron	UV-VIS	50–60	Malachite green	Abbassi *et al.* (2013)
17	Leaf extract of *Azadirachta indica*	Cerium dioxide	XPS	10–15	Rhodamine B	Sharma *et al.* (2017)
			DSC			
			TGA			
			UV-VIS			
			XRD			
			SEM			
			TEM			
18	*Angelica gigas* ribbed stem extracts	Silver and gold	FTIR	20–50	Eosin Y	Chokkalingam *et al.* (2019)
			EDX		Malachite green	
			XRD			
			FTIR			
			UV-VIS			
			PSA			
20	Leaf extract of *Camellia sinensis*	Zinc Oxide	UV-VIS	60	Malachite green	Batool *et al.* (2018)
			FTIR			
			XRD			
			SEM			
21	Mulberry leaves	Iron	DLS	47.70	Methylene blue	Lim *et al.* (2018)
			SEM		Methyl orange	
			FTIR			
			UV-VIS			
			Zetasizer			
22	Datura leaf extract	Iron	SEM	326–327	Solo chromo black (SCB)	Raju *et al.* (2017)
			UV-VIS			
23	*Zanthoxylum armatum* leaves	Silver	UV-VIS	15–50	Safranine O	Jyoti and Singh (2016)
			FTIR		Methyl red	
			SEM		Methyl orange	
			TEM		Methylene blue	
			SAED			
			XRD			
			EDX			
24	*Gymnema sylvestre* extract	Silver	UV-VIS	95.2	Methylene blue	Kumar *et al.* (2019)
			FTIR			
			XRD			

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			TEM			
			SEM			
25	*Brassica oleracea* L. var. *italica*	Zinc oxide	XRD	14–17	Methylene blue	Osuntokun *et al*. (2019)
			EDX		Phenol red	
			TEM			
			SEM			
			UV-VIS			
			FTIR			
			Photoluminescence			
26	Ampicillin	Silver	EDX	14.1	Methylene green	Junejo *et al*. (2014)
			TEM			
			SEM			
			UV-VIS			
			FTIR			
27	*Catharanthus roseus* leaf extract	Palladium	TEM	38	Phenol red	Kalaiselvi *et al*. (2015)
			SEM			
			UV-VIS			
			FTIR			
			XRD			
30	*Kashayam*, *Guggulutiktha*, an *ayurvedic* medicine	Silver and gold	UV-VIS	15–50	Methylene blue	Suvith and Philip (2014)
			FTIR			
			XRD			
			TEM			
31	Arabic gum	Zinc oxide	UV-VIS	10	Direct blue 129	Fardood *et al*. (2017)
			FTIR			
			XRD			
			TEM			
32	*Persia americana* seed	Tin oxide	UV-VIS	4	Phenol red	Elango *et al*. (2015)
			FTIR			
			XRD			
			SEM			
			EDX			
33	*Helicteres isora* extracts	Silver	UV-VIS	25–45	Methyl violet	Bhakya *et al*. (2015)
			PSA		Safranin	
			FTIR		Eosin methylene blue	
			TEM		Methyl orange	
			SEM			
34	Lemon juice	Zinc oxide	SEM	21.5	Methyl orange	Davar *et al*. (2015)
			EDX		Methylene blue	
			XRD		Methyl red	
			TG-DTA		Reactive blue 21	
			FTIR			

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			UV-VIS			
			Photoluminescence			
35	*Trigonella foenum-graecum* seeds	Silver	UV-VIS	17	Methyl orange	Vidhu and Philip (2014)
			XRD		Methylene blue	
			HRTEM		Eosin Y	
			FTIR			
36	*Morinda morindoides* leaf extract	Cu/Fe3O4	UV-VIS	16–21	Congo red	Nasrollahzadeha *et al*. (2016)
			XRD		Rhodamine B	
			FTIR			
			TGA			
			DTA			
			TEM			
			SEM			
37	*Eucalyptus tereticornis*	Iron–polyphenol NPs	SEM	50–80	Acid black 194	Wang *et al*. (2014)
	Melaleuca nesophila		EDX			
	Rosemarinus officinalis		XRD			
			FTIR			
38	*Carica papaya* leaf extracts	Zinc oxide	SEM	50	Methylene blue	Rathnasamy *et al*. (2017)
			TEM			
			XRD			
			UV-VIS			

Conclusion

In the fabrication of nanoparticles, naturally occurring substances are the aptest candidate for playing the role of reducing and capping agent. Green synthesis of nanoparticles has several advantages such as cost-effective, biocompatible, rapid synthesis, high stability, and easily available. One major advantage of green synthesized nanoparticles is that they are environment friendly. The remediation of environmental pollutants using green synthesized nanoparticles has witnessed as an emerging trend in the last decade. The effluent of the dyeing industry poses a threat to the environment because of long term disposal. Conventional and advanced treatment techniques for the remediation of dyes failed because these were not degrading the dye completely, effectively, and economically. This review specially focused on the remediation of dyes by using different sizes and shape nanoparticles, which were synthesized by green nanotechnology because of the protection of the environment. Therefore, for our safe environment, green nanotechnology needs to be explored by further future studies in various fields such as water purification, air purification, agriculture, food science, etc.

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