

GREEN SYNTHESIS OF NANOPARTICLES & ITS APPLICATIONS: A REVIEW

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Abstract:

In recent years, area of material science is focused towards the green synthesis of nanoparticles in order to find out reliable, sustainable as well as eco-friendly technique for synthesis of metal and metal oxide nanoparticles. Living organisms, especially plants are widely utilized for green synthesis of nanoparticle due to the presence of various phytochemicals and biological components (e.g., flavonoids, alkaloids, terpenoids, amides, and aldehydes) that act as reducing agents rendering synthesis of nanoparticles possible. Nanoparticles synthesized through green routes are faster, easy as compared to traditional methods of synthesis. Moreover, these nanoparticles demonstrate more stability. Thus plants are widely utilized as the source of reducing agents for large scale biosynthesis of nanoparticles. In this review, we have summarized the various fundamental methods for "green synthesis" of metal and metal oxide [e.g., gold (Au), silver (Ag), copper oxide (CuO), and zinc oxide (ZnO)] nanoparticles utilizing natural plant products, trying to explore the role of biological components, essential phytochemicals as reducing agents and solvent systems. Lastly, we have covered applications of biosynthesized nanoparticles in terms of their antimicrobial activity, nanoparticles in biomedical, catalysis and biosensors fields.

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Keywords: Nanoparticles, green synthesis; reducing agent; antimicrobial property; biosensors.

1. INTRODUCTION

Nanotechnology has emerged as the most important area of research in science. Nanotechnology is referred to, "Synthesis, manipulation, and application of the structures of nanometer size" [1]. A Japanese researcher first introduced nanotechnology, Norio Taniguchi [2]. The science of nanotechnology is occupying vast area of research in material science nowadays, creating new nanostructures like nanoparticle, nanotubes and various other nanostructures with totally novel properties and varied [3]. One of the most

important area of research in nanotechnology deals with synthesis, characterization and applications of nanoparticles [4]. In fact the origin and knowledge of nanotechnology dates back to thousand years from use of golden ornaments to use of gold nanoparticles for staining drinking glasses to use of nanoparticles to cure variety of diseases. In short, nanotechnology encompasses knowledge of material science along with physics, chemistry and biology aiming at its biomedical use [5]. Green chemistry (GC) and nanotechnology (NT) together are influencing and evolving scientific research along with environmental safety. Together both these fields are emerging as „Green nanotechnology“, which is mainly focused towards the development of nanoscale, sized entities for enormous applications [6-9]. Nanoparticles are of different types including inorganic NPs as well as organic NPs. Inorganic nanoparticles include non-metallic, metallic and magnetic nanoparticles. Some of the examples of inorganic nanoparticles are ZnO, ZnS, CdS, and metallic NPs like Au, Ag, Cu, Al. Magnetic nanoparticles include Co, Fe, and Ni, while organic nanoparticles mainly incorporate carbon NP nanoparticles like quantum dots, carbon nanotubes and fullerenes. The more elaborated classification of nanoparticles is as following [11]-

1. Metals and metal alloys (Au, Ag, Pt, Pd, Cu, Fe, Ni, Co, Al, Mn, Mo)
2. Non-metallic inorganic nanoparticles (TiO₂, SiO₂, ZnO, Al (OH)₃, Fe₂O₃, Fe₃O₄, CeO₂, ZrO₂, CaO, ITO, ATO)
3. Nanomaterials based on carbon (fullerenes, carbon nanotubes, carbon nanofibers, graphene)
4. Nanopolymers and dendrimers (polymeric nanoparticles, polymer nanotubes, nanowires and nanorods, nanocellulose, nanostructured polymer films)
5. Quantum dots (Cadmium telluride, cadmium selenide and cadmium free quantum dots)

The gold (Au) and silver (Ag) nanoparticles are area of interest to researchers from broader application point of view. Increased emergence of antibiotic resistance amongst pathogens to antibiotics increased its applications as these metallic nanoparticles demonstrate promising antibacterial activity due to its high surface-area-to-volume ratio. Silver nanoparticles impair cell division as well as respiratory chain in bacteria. These nanoparticles mostly adhere to bacterial cell membrane and destroy sulphur containing proteins as well as bacterial DNA making its phosphorous containing component as the main target [12]. The present review article focuses on various green synthesis methods used for the synthesis of Silver (Ag), gold (Au), Zinc oxide (ZnO) and Copper oxide nanoparticles (CuO) using plant extracts. Various biomedical, environmental and pharmacological applications are also reviewed.

2. METHODS FOR SYNTHESIS OF NANOPARTICLES

The field of nanotechnology is most emerging area of research in material science nowadays. Nanotechnology is defined as „the manipulation of the matter at the atomic or molecular level of 1-100 nm size range.“ Due to this small size these substances with superior properties and applications can be produced, also these nanoparticles play bridging role between bulk structures and molecular structures. [13] Due to vast applications in biomedicine, and materials with superior properties like good tensile strength, conductivity and superior rigidity they are on demand in aircraft spares

and satellite parts manufacturing industries also. [14-17] The development of novel methods for the synthesis of stable dispersed nanoparticles of nanometer sizes and chemical composition is recent challenge in front of science of nanotechnology. Physical and Chemical techniques are the most common methods for synthesis of nanoparticles. Physical methods of synthesis include severe plastic deformation, ultrasonic shot peeling methods, and inert gas condensation methods. Grinding and pyrolysis processes are mainly employed for synthesis of metallic nanoparticles. Grinding mainly include reducing the size of macromolecules or micromolecules by size reduction mechanism. In the method of pyrolysis liquid or gas precursor is forced through small inlets under high pressure and oxidized nanoparticles are recovered from it. However physical method for synthesis of nanoparticles bears some disadvantages as the nanoparticles, as it has been reported that this technique results in production of the particles larger than 100 nm size, which is less considered as nano size. Also physical methods of synthesis of nanoparticles are expensive and its production results in losses to environment quality. [18]. The chemical methods employed for synthesis of nanoparticles involves a method in which nanoparticles are synthesized in presence of reducing agents like potassium bitartrate or sodium borohydride [19]. In these techniques polyvinyl pyrrolidone or sodium dodecyl benzyl sulfate are used as stabilizing agents to prevent the clumping or agglomeration of nanoparticles. methoxy polyethylene glycol or hydrazine are employed for same purpose [20-21]. The chemical methods used for synthesis of nanoparticles are expensive as well as not eco friendly, as it employs use of toxic hazardous chemicals during synthesis posing risk to ecosystems. Thus, there is tremendous need of development of cost effective, economical as well as eco friendly techniques for synthesis of nanoparticles.

2.1 TOP DOWN AND BOTTOM UP PROCESS FOR THE SYNTHESIS OF NANOPARTICLES :

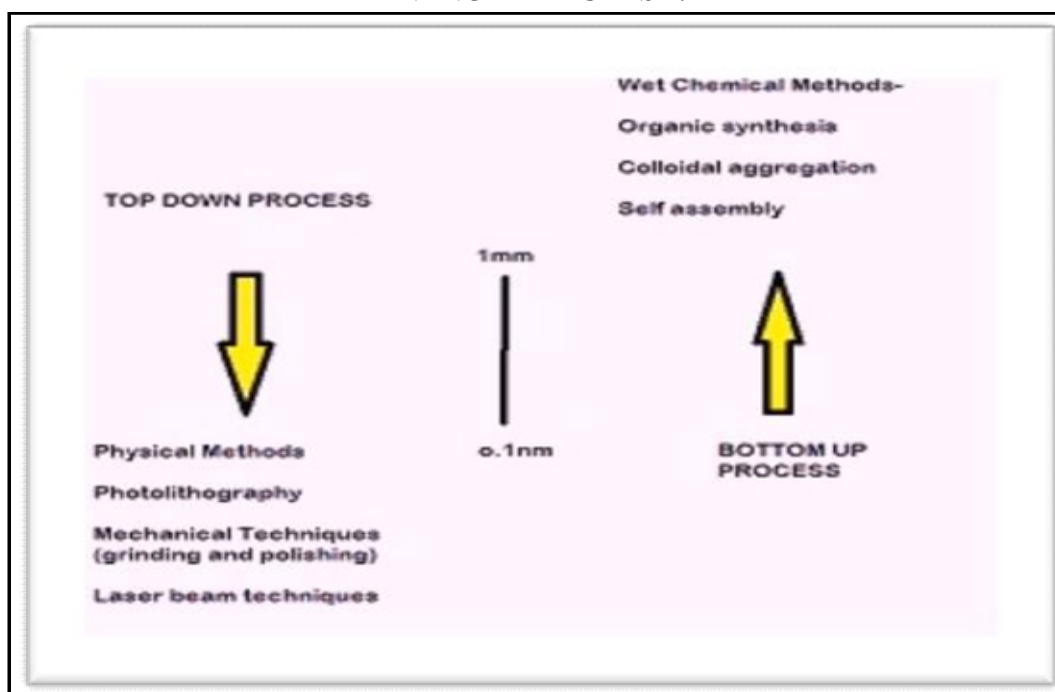


Figure 1: Top-down and Bottom-up Methods of Nanoparticles Synthesis [22].

The top-down process of synthesis of nanoparticles is amongst the physical techniques synonymously referred as the micro fabrication technique as it employs tools to cut and size materials in desired shape and sizes from suitable initial material. Due to larger sizes of resultant nanoparticles, addition of acid is needed in this process to reduce the size of NPs. Some of the techniques in this category are- Photolithography, grinding in ball mill as well as etching and sputtering. (Figure 1)

2.2 GREEN SYNTHESIS OF NANOPARTICLES:

However, “Bottom up” process is one of the reliable and effective techniques as compared to “Top down Process”. Here, the process initiates from smaller entities at atomic level or molecular level with self-assembly. One of the significance of this technique is that- it is possible to keep check the size of nanoparticles by varying various factors like concentration of precursors and reaction parameters [22]. Both of the above methods of synthesis of nanoparticles involve use of toxic chemicals as well as resulting into formation of byproducts, which are not safe from environment safety point of view. Therefore, to avoid these drawbacks, it is important to develop advancement of techniques into the newer, cheaper and eco-friendly methods of nanoparticle synthesis [23]. Hence, this review article is focused on green inspired synthesis of nanoparticles. The un-eco-friendly as well as toxic methods of synthesis of NPs leads to an urge to develop safer as well as cost effective methods for synthesis of nanoparticles. Exploiting green routes for synthesis of nanoparticles is one of the options to solve this issue. Researchers have found that, naturally occurring materials can be employed well for the synthesis of stable NPs. Advancement green of material science together with nanotechnology has enriched the newer dimensions of biosynthesis of nanoparticles. The term „Green Synthesis“ distinguishes natural or green methods from traditional physical and chemical methods. In green synthesis methods naturally occurring substances are used as reducing agents for synthesis of NPs. „Green synthesis“ is one of the approach for reaching green elements of the nature for example ethanobotanical, phytochemicals, materials derived from animals and biomolecules from microbial origin. The basic advantages of green synthesis methods over synthetic methods can be listed as below-

- a. Since natural components are used, it's an environment friendly approach since no toxic ingredients are involved during synthesis.
 - b. The natural biological ingredients derived from living things act as reducing as well as capping agents, thus it's a cost effective technique.
 - c. Maintenance of external experimental conditions of high energy and high pressure is not required, thus again its cost effective approach.
 - d. Large-scale production of nanoparticles is possible.
 - The various naturally occurring sources which can be employed for synthesis of metal and metal oxide includes-
 - Use of Plant and Plant Extracts.
 - Use of microorganisms (bacteria, fungi, actinomycetes etc.)
 - Use of templates like membranes, viruses DNA, and diatoms.
- Although use of microorganisms as templates are effective green methods of nanoparticle synthesis, our review describes the synthesis of NPs using plant and plant extracts in further sections.

3. PLANT EXTRACT MEDIATED GREEN SYNTHESIS OF NANOPARTICLES:

3.1 Plant Extract Mediated Green Synthesis of Silver nanoparticles (AgNps):

Amongst the biological methods available, use of plant extracts for synthesis of nanoparticles are most suitable as plants are huge reservoir of easily available metabolites which can act as reducing agents with faster reaction time, as compared to fungi and several bacteria which need long time of incubation during nanoparticle synthesis process for reduction of metal ions [24-26]. In addition, use of plant extracts for synthesis of nanoparticles can occur at lower temperatures and requires less controlled experimental conditions. The other advantages of plant extract mediated synthesis of nanoparticles require safe reagents, and can be easily applied for medicinal, surgical and pharmaceutical purposes [27].

The three most ingredients required for green synthesis of nanoparticles using plant extracts include - metal salt, reducing agent and capping or stabilizing agent for preventing agglomeration of nanoparticles. Plants are the sources of enormous biomolecules including carbohydrates, proteins, amino acids, saponins, phenolic compounds, terpenoids, alkaloids, tannins, vitamins and reducing sugars which act as reducing agents during stable nanoparticle synthesis mechanism. The concentration of all these components in plant describes the reducing capacity of the plant [28]. The rate of NPs synthesis is dependent on factors like amount of plant extract, concentration of metal ions, its pH and temperature [29]. Gardea-Torresdey et al. (2003) using Alfalfa sprouts, reported the first description of synthesis of silver nanoparticles. They reported that Alfalfa roots absorb silver ions from agar medium and direct them towards shoots of this plant in oxidized state. Thus eventually synthesis of silver nanoparticles occurs (Ag-Np) in shoots [30]. Sadeghi et al. (2015) reported the synthesis of Ag-Np using *Z. tenuior* plant extract. 0.2 grams of dried extract of *Z. tenuior* leaf was added to 50 ml of deionized water, stirred for around 1 hr. and centrifuged. When clear *Z. tenuior* extract was mixed with 0.1 mM AgNO_3 . The silver nanoparticles were formed. TEM studies of these synthesized silver nanoparticles revealed that AgNps formed were monodispersed, spherical in shape with average size of 20 nm. Stability studies using zeta potentiometer revealed that synthesized AgNps were stable at pH range 6-12. Also FTIR studies revealed that proteins from plant extracts form a coat over synthesized AgNp to prevent its agglomeration, moreover XRD and UV-VIS studies revealed that the leaf extract of *Z. tenuior* can synthesize AgNps [31]. Philip et al. (2011) reported the synthesis of AgNp using *M. uniflorum* leaf extract. Appearance of signatory brown color in reaction mixture revealed the synthesis of Silver nanoparticles in reaction mixture. Characterization studies carried out using XRD, TEM, UV-Visible and FTIR studies revealed that silver nanoparticles synthesized using *M. uniflorum* were well dispersed, and demonstrated face centered cubic structure. Additionally FT-IR studies revealed the presence of functional groups associated with capping agents in nanoparticle solution [32].

Sulaiman et al. (2013) in a study conducted to synthesize silver nanoparticles reported that, silver nanopart

icles can be synthesized from *E.*

Chapmanian leaves extract when 10 ml of leaves extract from *E. Chapmanian* was mixed with 0.01 mM/ml to 0.02 mM/ml of aqueous AgNO_3 and exposed to sunlight. Characterization studies carried out using XRD studies revealed the presence of face centered cubic AgNP, whereas UV-Visible spectra demonstrated silver Plasmon resonance at 413 nm. Thus,

E. Chapmanian leaves are capable of synthesizing silver nanoparticles [33]. In another study, Kumar et al. (2013) reported the green synthesis of silver nanoparticles using *Alternanthera dentata* leaf extract and their antimicrobial activity. For this, 20 ml of 1 mM of aqueous solution of AgNO_3 were added to 2 ml of leaf extract at room temperature. Change in color of solution from yellow to brown indicated the synthesis of silver nanoparticles, which were confirmed by surface plasmon resonance band at 430 nm. XRD studies revealed that size of nanoparticles was found to be 10-80 nm. Whereas TEM studies demonstrated size of silver nanoparticles synthesized using this technique were 50-100 nm. FTIR studies indicated that water soluble fraction of plant extract *A. dentata* plays crucial role in reduction of precursors involved in synthesis process [34].

In another experiment on green synthesis of silver nanoparticles, Velmurugan et al. (2015) reported that silver nanoparticles could be synthesized using extract derived from peanut shells. TEM, XRD and FTIR studies carried out concluded that AgNPs synthesized by this method are analogous to that of particles synthesized by commercial methods. AgNPs recovered by this method were 10-50 nm in size with oval shape [35]. In another study, Bar et al. (2009) reported that, green synthesis of silver nanoparticles is possible by reducing aqueous solution of AgNO_3 by *Jatropha curcas* extract. This extract also acts as capping agent during synthesis [36]. Gavhane et al. (2012) illustrated the synthesis of AgNPs by reducing aqueous salt solution of AgNO_3 using the extract of Neem and Triphala leaves. AgNP characterization studies like TEM, and nanoparticle tracking analysis revealed that the silver nanoparticles formed by this technique are spherical with diameter in range of 43 nm - 59 nm [37]. Brahma Chari et al. (2014) in a study on green synthesis of AgNPs using *O. sanctum* leaf extract illustrated that, when varying concentration of this leaf extract was added to different conical flasks containing 45 mL of 10-30 M silver nitrate solution each, and incubated under direct sunlight, silver nanoparticle formation was revealed by gradual change in color of the solution.

TEM studies demonstrated that, AgNPs formed were in nanometer size range [38]. Dulen Saikia (2014) (also illustrated the green synthesis of AgNPs using plant extracts of two medicinal plants namely, *Ocimum tenuiflorum* and *Catharanthus roseus*. During the experiment, 5 gm of dried leaves of these plants were added to 100 ml of deionized water and boiled for 30 min. When extract was filtered and added to desired quantity of aqueous AgNO_3 and incubated. After incubation appearance of AgNPs were revealed by change in color of the solution from brown to dark reddish brown [39]. In another study, Vilchis -Nestoretal. (2008) reported the biosynthesis of silver as well as gold nanoparticles using aqueous solution of green tea (*Camellia sinensis*). This study revealed that amount of plant extract has a significant role in determining and controlling the size of silver nanoparticles. Increase in concentration of plant extract in reaction medium results in synthesis of nanoparticles with reduced diameter [40]. One study on *C. zeylanicum* bark extract mediated synthesis of AgNPs revealed that the size of biosynthesized AgNPs varied with concentration of *C. zeylanicum* bark extract [41]. In another study, Awwad and Salem

(2012) illustrated that Mulberry leaves extract can be successfully utilized for green synthesis of monodispersed silver nanoparticles with average diameter in range of 20 nm. Moreover, these, AgNPs demonstrated potential antibacterial activity against *S. aureus* and *Shigella* sp. [42]. Roy et al. (2017) demonstrated biosynthesis of AgNPs using *Azadirachta indica* leaf extract. In this experiment, 1 mM solution of silver nitrate is prepared by adding 0.17 gram of AgNO_3 to 1000 ml of distilled water in volumetric flask. From this, 100 ml of solution was used for preparation of Silver nanoparticles. Fresh neem leaves were collected from neem tree (*Azadirachta indica*). Fresh and healthy leaves were plucked and then thoroughly washed under running tap water to remove the dirt over leaves and then dried over sterile muslin cloth until water gets removed from surface of leaves. Using sterile knife, these leaves were then finely chopped. 20 grams of these leaves were then added to 100 ml of sterilized distilled water. The water containing leaves was then boiled for around 10 min. The preparation was then cooled and extract is then filtered using sterile muslin cloth and stored for preparation of silver nanoparticles. To prepare silver nanoparticles with neem leaves extract, 5 ml of silver nitrate (1 mM) solution is added in each of the separate test tube. Then to each of the test tube 5 ml neem extract is added. All the tubes were then incubated in dark chamber to minimize the photo activation of silver nitrate. Nanoparticle and its color formation is then confirmed by change in color from colorless to brown. The method successfully illustrates the formation of silver nanoparticles [43].

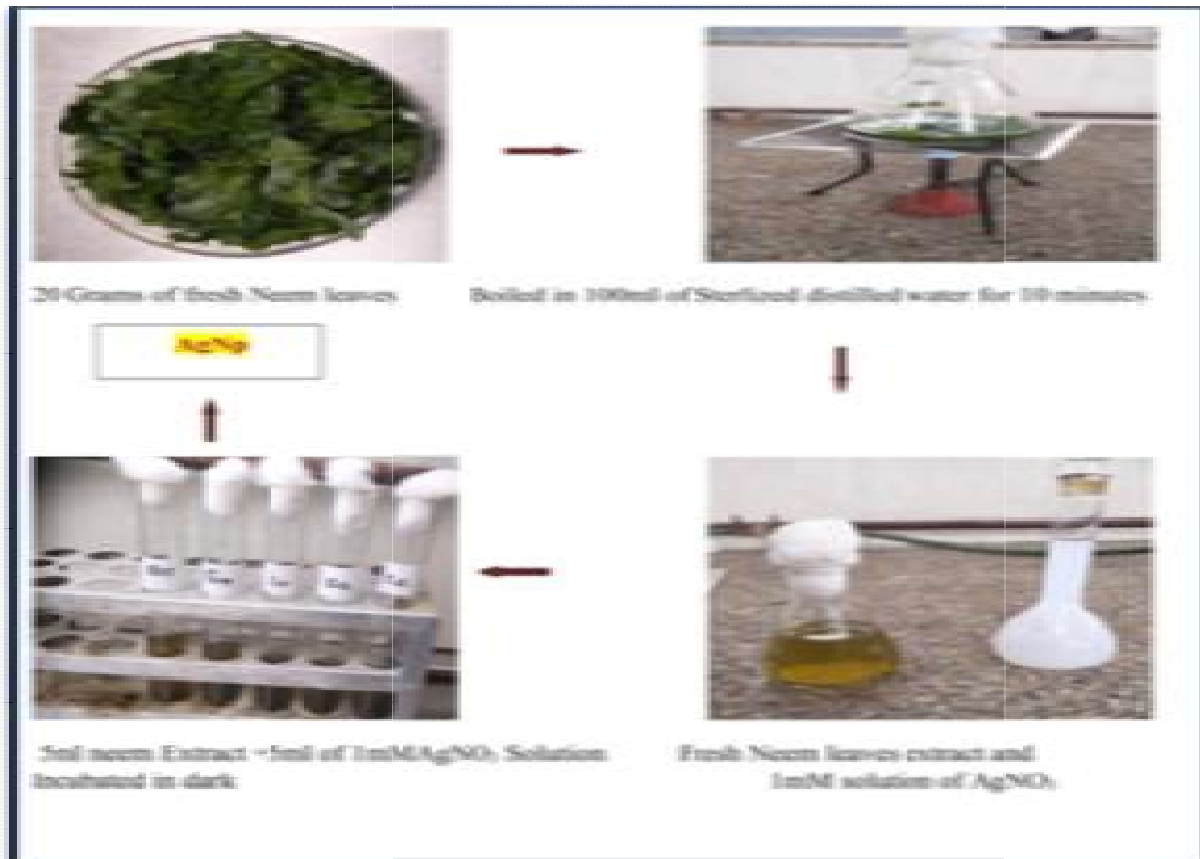


Figure: 2 Green Synthesis of Silver Nanoparticles
using *Azadirachta indica* leaf extract using the method of Roy et al. (2017)

In an attempt to synthesize, silver nanoparticles using aqueous extract of turmeric powder as reducing as well as capping agent. After addition of desired amount of AgNO_3 to turmeric extract the appearance of dark reddish brown color due to reduction of AgNO_3 after 24 hrs. Characterization studies carried out using XRD analysis, UV-Visible spectroscopy, FTIR analysis and Transmission electron microscopy (TEM). UV-Visible spectra demonstrated maximum absorbance spectra at 432 nm. TEM studies revealed the average size of particle size was $18 \pm 0.5 \text{ nm}$ [44]. It has been reported that Silver nanoparticles could also be synthesized using *Magnifera indica*, *Eucalyptus tereticornis*, *Carica papaya*, and *Musa paradisiaca* under desired conditions. Then nanoparticles of different size and shapes in nanometric range were obtained. Thus green synthesis of silver nanoparticles utilizing plant extract as reducing and capping agents is vast and therefore work using *Magnifera indica*, *Eucalyptus tereticornis*, *Carica papaya*, and *Musa paradisiaca* under desired conditions. The nanoparticles of different size and shapes in nanometric range were obtained [45]. Thus green synthesis of silver nanoparticles utilizing plant extract as reducing and capping agents is vast and therefore work done by various researchers in same line is summarized as under in Table 1.

Plant	Precursor	Reducing and capping agent	Size (nm)	Reference
<i>Quercus brant</i>	AgNO_3	Leaves hydro alcoholic extract	6	[46]
<i>Garlic</i>	AgNO_3	Extract	4-6	[47]
<i>Hibiscus rosasinensis</i>	AgNO_3	Extract	14	[48]
<i>Cuminum cyminum</i>	AgNO_3	Seed powder	12	(49)
<i>Lantana camara</i>	AgNO_3	Fruit extract	5-12.99	[50]
<i>Peach gum</i>	AgNO_3	Peach gum powder	56 ± 7.87	[51]
<i>Citrus limon</i>	AgNO_3	Limon extract	than 50	[52]
<i>Tea</i>	AgNO_3	Leaves extract	20	[53]
<i>Terminalia arjuna</i>	AgNO_3	Bark extract	2-100	[54]

<i>na</i>				
<i>Brassicarapa</i>	AgNO ₃	Leavesextract	16.4	[55]
<i>Solanumtuberosum</i>	AgNO ₃	Potatoinfusion	1.-12	[56]
<i>Ficuscarica</i>	AgNO ₃	Leavesextract	13	[57]
<i>Syzygiumcumini</i>	AgNO ₃	Barkextract	20–60	[58]
<i>wieteniamahogany</i>	AgNO ₃	Leavesextract	50	[59]
<i>Menthapiperita</i>	AgNO ₃	Leavesextract	5-30	[60]
<i>Alliumcepa</i>	AgNO ₃	Extract	33.6	[61]
<i>Magnoliakobus</i>	AgNO ₃	Leavesextract	16	[62]
<i>Cycas</i>	AgNO ₃	Leavesextract	2-6	[63]
<i>Cocousnucifera</i>	AgNO ₃	Flower	22	[64]
<i>Calotropisprocera</i>	AgNO ₃	Leavesextract	10	[65]
<i>Abutilonindicum</i>	AgNO ₃	Leavesextract	7-17	[66]
<i>Belgianendive</i>	AgNO ₃	Leavesextract	75-80	[67]
<i>Ananascomosus</i>	AgNO ₃	Fruitextract	5-30	[68]
<i>opanaxseptemlobus</i>	AgNO ₃	Leavesextract	30.8	[69]
<i>Azadirachtaindica</i>	AgNO ₃	Kernelextract	50-100	[70]
<i>Avenasativa</i>	AgNO ₃	Stemextract	5-85	[71]
<i>Geranium</i>	AgNO ₃	Leavesextract	16-40	[72]

Table 1 : Methods for the synthesis of Silver Nanoparticles

3.2 PLANT EXTRACT MEDIAD SYNTHESIS OF GOLD NANPARTICLES:

Nanotechnology is advancing like no other science and is making revolution in every aspect of life. Along with silver nanoparticles, researchers are nowadays focusing towards synthesis of gold nanoparticles (AuNps) due to its superior properties and applications. Use of phytochemicals for

synthesis of these gold nanoparticles is of significant importance due to reduced environmental safety issues as well as its biomedical importance. Plants are reservoirs of number of phytochemicals, which can be exploited as reducing as well as stabilizing agents during gold nanoparticle synthesis. Therefore this issue is choice of many researchers across the globe from last decade. In following Para, we have reviewed various methods for green synthesis of gold nanoparticles [73]. Chandran et al. (2014) illustrated in their studies that gold nanoparticles can be synthesized from extracts of two known medicinal plants namely *Cucurbita pepo*, and *Malvacrispas* successfully; they also reported that the AuNps synthesized were potential antibacterial agents inhibiting the growth of many bacteria associated with food spoilage [74]. A study conducted by Wan et al. (2009) illustrated that, monodispersed gold nanoparticles with diameter in the range of 5-30 nm can be synthesized using extracts of plant *Scutellariabarbata* as reducing and stabilizing agent. The study also concluded that in plant extract of *Scutellariabarbata* flavonoids, diterpenoids, alkaloids, steroids and polysaccharides were remaining reducing and stabilizing agents [75]. In another study, Raghunandan et al. (2010) reported that gold nanoparticles (AuNps) in diameter range of 5-100 nm can be synthesized from extracts of buds of *Syzygium aromaticum*. In this study they reported that the reduction of salt solution during synthesis of AuNps is carried out by flavonoids from buds of *Syzygium aromaticum*. [76]. Shankar et al (2003) illustrated the synthesis of gold nanoparticles with size 2.5 to 27.5 nm from plant extract of *Pelargonium roseum* (rose geranium). [77]. In an experiment carried out on Pear fruit extract for biosynthesis of gold nanoparticles, Ghodke et al. (2009) illustrated successful synthesis of stable, triangular shaped AuNps with diameter in the range of 200-500 nm, the study conducted also proposed that these AuNps can be used in hexagonal catalysis and biosensing processes [78]. Shankar et al. (2005) also reported that, gold nanoparticles can be synthesized using green synthesis method in which plant extract of *Cymbopogon flexuosus* (lemongrass) acts as both reducing as well as capping agent. The size of triangular shaped AuNps synthesized by this method is in the range of 200-500 nm and can find application as infrared-absorbing material in optical coatings [81]. In different experiments, Gardea-Torresdey et al. (2003) illustrated the biosynthesis of gold nanoparticles from plant extract of *Medicago sativa* (alfalfa), the AuNps produced by this method were irregular, tetrahedral, hexagonal platelet, decahedral, icosahedral which find application in labeling in structural biology, paints [82]. Thus, the plant extracts are efficient in synthesizing gold nanoparticles from plant based materials. Table 2 enlists the variety of plant based green synthesis of gold nanoparticles.

Plant	Precursor	Reducing and capping agent	Size (nm)	Reference
<i>Emblica officinalis</i>	Leaf extract	Gold and silver	10–20	85
<i>Coriandrum sativum</i>	Leaf extract	Gold	75–57.91	86
<i>Avena sativa</i>	Leaf extract	Gold	5–20	87

<i>Nnamomumcamphora</i>	Sundriedleaf	Gold	55-80	88
<i>Garcinia combogia</i>	Fruitextract	Gold	20-100	89
<i>Cocosnucifera</i>	Greencoconut shellexttract	Gold	15	90
<i>Magnoliakobus</i>	Leafextract	Gold	5-300	91
<i>Volvariellavolvacea</i>	Extract	Gold	-150nm	92
<i>nacardiumoccidentale</i>	Leafextract	Gold	50-100	93
<i>Ocimumsanctuml</i>	Leafextract	Gold	30nm	94
<i>Chenopodiumalbum</i>	Leafextract	Gold	0-30nm	95
<i>Lantanacamara</i>	Leafextract	Gold	100nm	96
<i>Mangiferaindica</i>	Leafextract	Gold	17-20	97
<i>Sorbusaucuparia</i>	Leafextract	Gold	18	98
<i>Stevia rebaudiana</i>	Leafextract	Gold	17	99
<i>Terminaliacatappa (almond)</i>	Leafextract	Gold	21.9	100
<i>Solanumnigrum</i>	Leafextract	Gold	50	101
<i>Terminaliaarjuna</i>	Barkextract	gold	60	102
<i>Pistaciaintegerrima</i>	Leafextract	Gold	50	103
<i>Polysciasscutellaria</i>	Leafextract	Gold	5-20	104
<i>Zizyphusmauritiana</i>	Leafextract	Gold	27	105
<i>Euphorbiahirta</i>	Leafextract	Gold	6-71	106
<i>Pistaciaintegerrima</i>	Leafextract	Gold	20-200	107

Table 2. Methods of Synthesis of Gold Nanoparticles

3.3 PLANT EXTRACT MEDIATED GREEN SYNTHESIS OF ZINCOXIDE(ZnO)NANOPARTICLES:

Zinc oxide may be a useful material as a result of its distinctive physical and chemical properties like medicament, deodorizing property, high piezoelectric effect, semiconductor, giant energy, UV filtering

capability etc. ZnO nanoparticles have several applications in varied industries like pharmaceutical, chemical, textile, paint, rubber, packaging industries etc. apart from these, they are currently employed in agricultural formulations, ecotoxicological studies, environmental fate. They are used for disposal of aquatic weed that is immune to all kinds of destruction techniques like physical, chemical and mechanical [108]. Therefore, day by day ZnO NPs demand is increasing. Therefore, manufacturing should be done by eco-friendly methods and it needs to be cost-effective. Hence, during this review, synthesis of ZnO NPs by an inexperienced synthesis technique and its applications are reviewed.

Zinc oxide nanoparticles may be synthesized by various methods like chemical vapor condensation, arc discharge, chemical element plasma-metal reaction method, and optical maser transmutation within the vapor section, microemulsion techniques, hydrothermal methods, sol-gel methods, sonochemical methods, plant extract mediated synthesis technique, microorganisms mediated synthesis. Out of all the techniques of synthesis of nanoparticles, exploiting plant extracts for ZnO synthesis is straightforward, not pricy, and eco-friendly as compared to ancient synthesis. The traditional chemical synthesis method possesses drawbacks like need of high pressure-temperature and toxic solvents.

Plant extract mediated green synthesis of Zinc

oxide nanoparticles (ZnO) eliminated a lot of chemical process and also limits the utilization of costly

and cyanogenic chemicals. Hence, researchers are specializing in inexperienced synthesis of nanoparticles. Another benefit of plant extract mediated synthesis is that they are a lot of stable and also the rate of synthesis is quicker than within the case of microorganisms. Moreover, the nanoparticles obtained by plant extract mediated synthesis are variable in forms of size and shape as compared with those synthesized by using other different approaches.

Thus, these numerous benefits of plant and plant-derived materials for synthesis of zinc oxide nanoparticles have interested researchers to analyze mechanisms of metal ions uptake and bio-reduction by plants, and to know the mechanism behind its synthesis. [109]. Plants are the foremost common biological substrate used for the eco-friendly green synthesis of nanoparticles with metal like ions [110-111]. Plants are abundant sources of cost-effective, non-toxic natural reducing agents. During green synthesis processes, these natural reducing biochemicals from plants can be easy mechanisms of extraction like treating the plant material to easily available solvents like H_2O or alcohols. [112] Different components of the plants are utilized for green synthesis of ZnO NPs most of which comprised of leaves, roots, seeds and fruits. The extracts prepared from these plant sources act as reducing agents during biosynthesis processes. [113-116] Nath et al. (2014) reported that Margosa plant can be utilized for biosynthesis of zinc oxide nanoparticles (ZnO) as it is the rich source of flavanones, terpenoids and reducing sugars, the constituents of the tree leaf broth. They illustrated that, the organic components in leaf broth of Margosa plant are responsible for reduction of zinc oxide to ZnO NPs as well as its stabilization [117]. Zinc oxide is also known as Philosopher's wool synonymously. Noorjahan et al. (2015) reported the green synthesis of philosopher's wool nanoparticles (ZnO NPs) from leaf extract of Margosa plant. When these ZnO NPs were characterized by SEM and FTIR techniques. In FTIR analysis was revealed that ZnO NPs

were encircled by plant derived terpenoids, ketones, aldehydes and acids which suggest their role as stabilizing as well as capping agents. SEM studies showed stable philosopher's wool Nanoflakes and spindle formed nanoparticles. The dimensions of the ZnO nanoparticles synthesized were found to 50 nm in size [118]. Mariametal. (2014) reported a unique synthesis for In₂O₃ and ZnO Nanoparticles with particle sizes within the vary of ten to thirty nm exploitation metal nitrate and Zn nitrate solutions. They used Aloe Vera extract as a solvent rather than organic solvents. [119] In another study, Singhet al. (2018) illustrated the effect of variation in concentration of plant extract and zinc precursor, incubation time and temperature of reaction in a study of the biosynthesis of ZnO NPs using *Eclipta alba* leaves extract. In this study they reported that, ZnO NPs formation turned more uniform and smaller after increasing zinc acetate concentration (1.0–5.0 mM). [120] These findings were found to be in agreement with studies of Ain Samat et al. (2013) in this study, for synthesis of ZnO NP s from *Citrus aurantifolia* extract the increase of zinc acetate concentration resulted in enhanced size uniformity and reduced size of newly formed nanoparticles [121]. Matinise et al. (2017) reported the study in which *Moringa oleifera* leaves extract components react with the zinc (II) ions which formed zinc oxide after calcination process. These researchers studied the effect of plant extract mediated synthesis on ZnO nanoparticles synthesis at different temperatures. The FTIR studies revealed that ZnO NPs synthesized at 100°C demonstrated hydroxyl (–OH) stretching bands which was indicative of formation of zinc complexes with antioxidants in synthesis process. [122]. Nath et al. (2013) reported the green synthesis of ZnO nanoparticles from leaf extract of *Azadirachta indica*. In this study they suggested that, neem plant leaves are potential source of flavones, terpenoids and reducing sugars. And aldehyde groups are responsible for reduction as well as stabilization of zinc oxide to zinc oxide nanoparticles [123]. Raut et al. (2013) reported the green synthesis of Zinc oxide nanoparticles mediated by leaves extract of *Ocimum Tenuiflorum* plant. The constituents present in plant extract act as reducing agents. They synthesized Zinc oxide nanoparticles were characterized by X-Ray diffraction (XRD), scanning electron microscope (SEM) technique, Fourier Transform Infrared Spectroscopy (FTIR). The average size of ZnO NPs obtained by this method were 13.86 nm [124]. In following table 3 we have summarized the various plant based green synthesis methods for ZnO NP biosynthesis.

Plant	Precursor	Reducing and capping agent	Size (nm)	References
Aloevera leaves	Zinc Nitrate (NS)	Leaves extract	25	125
<i>Lycopersicon esculentum</i>	Zinc Nitrate (NS)	Fruit extract	40-100	126

<i>Peganum harmala</i>	ZincNitrate (NS)	Seedextract	40	127
<i>Stevia rebaudiana</i>	ZincAcetate (ZA)	Leavesextract	50	128
<i>Hibiscus subdariffa</i>	ZincAcetate (ZA)	Leavesextract	190-400	129
<i>Costus woodsonii</i>	ZincNitrate (NS)	Seedextract	20-25	130
<i>Citrus sinensis</i>	ZincNitrate (NS)	Peelextract	12	131
<i>Couroupita guianensis</i>	ZincAcetate (ZA)	Leavesextract	15-25	132
<i>Mentapulegium</i>	ZincNitrate(NS)	Leavesextract	38-49	133
<i>Simarauba glauca</i>	ZincNitrate (NS)	Leavesextract	NS	134
<i>Vitexnegundo</i>	ZincNitrate (NS)	Leavesextract	NS	135
<i>Solanum nigrum</i>	ZincNitrate (NS)	Leavesextract	29	136
<i>Jacaranda mimosifolia</i>	ZincNitrate (NS)	Flowerextract	2-4	137
<i>Parthenium hysterophorus</i>	ZincNitrate (NS)	Leavesextract	27-84	138
<i>Anisochilus carnosus</i>	ZincNitrate (NS)	Leavesextract	20-40	139
<i>Petroselinum</i>	ZincNitrate (NS)	Leavesextract	70	140

Table3:PlantExtractMediatedGreenSynthesisofZincOxideNanoparticlesfromSome

3.4

PLANTEXTRACTMEDIATEDGREENSYNTHESISOF COPPER OXIDE NANOPARTICLES:

In recent past years synthesis of nano particles utilizing greensynthesis methodshas attained enormous importance due to numerous advantages over conventional chemical and physical methods. Plants are widely distributed across the world, are rich source of uncountable number of phytochemicals as well as easily available and processible [141]. Copper oxide nanoparticles are important due to numerous applications as antimicrobial agents, and also used in batteries, super conductors, and gas sensors.

Copper oxide nanoparticles are also used as antifouling agent, antibacterial, purifiers, algacides, fungicide [142-144]. The synthesis of copper oxide nanoparticles using inactivated plant tissue, plant extracts, exudates, and other parts has attracted numerous researchers [145]. It is a need to develop ecofriendly and economic methods to synthesize metal oxide nanoparticles as there is rapid urbanization, industrialization and population growth, hence demand of nanoparticles in different industries is also increasing tremendously. To fulfill this demand green synthesis of metal oxide nanoparticles is best method. Single step green synthesis process can be used to prepare nanoparticles from metal ions. This can be happened as plant extracts contain biomolecules which reduce metal ions to nanoparticles. This is a very rapid process, can be conducted at room temperature and pressure, easy to perform. It is ecofriendly and economic also. Reducing agents are the biomolecules present in plants as their metabolites like alkaloids, phenolic compounds, terpenoids etc. and coenzymes [146].

K. Rayapa Reddy studied the green approach of CuO NP synthesis using *Calotropis procera* (easily found in India) which belongs to the *Asclepiadaceae* family. *Calotropis procera* used in many treatments like diseases of spleen, piles, tumors, etc. These NPs are widely used in many applications such as catalysis because of their narrow band gap, used in photocatalytic properties [147-148]. Alaa Y. Ghidan et al. (2017) reported the green synthesis of copper NPs using *Punica granatum* peels extract. *Punica granatum* fresh peels were obtained and washed for several times. Powder was prepared from dried peel of *P. granatum* and added to sterile distilled water and this mixture was boiled until appearance of yellow color. Then CuO NPs were synthesized, for this copper acetate powder was dissolved in the water and stirred with magnetic stirrer. Then *P. granatum* extract was added to the stirred solution; it gives green color to the solution and eventually it turned to brown which shows the formation of monodispersed Cu NPs. [149]

Synthesis of CuO NPs from *Abutilon indicum* was illustrated by Ijaz et al. (2018) They collected fresh leaves of *Abutilon indicum* and washed gently to remove dust particles as well as dried and shaded parts. Then the leaves were pulverized and sieved using a 200-nm mesh sieve to be used as fuel. They prepared nanoparticles by green combustion method with leaves extract of *A. indicum* as a fuel. To synthesize CuO NPs, Copper (II) nitrate trihydrate was mixed with *Abutilon indicum* extract in double-distilled water. Homogenized solution was prepared by constant stirring for 2–5 min using a magnetic stirrer. Combustion reaction was performed on the mixture using a pre-heated muffle furnace at the temperature 400 ± 5 °C to produce CuONPs. The ash content of the plant extract was removed by filtration. The solution was first washed with distilled water, then by methanol to remove impurities. Then CuONPs were calcinated for 2 hrs to get pure form. This pure fine black CuONPs can be stored in an airtight container for further use. [150] Few of the plant extract mediated synthesis processes for CuO nanoparticles are listed in following table 4-

Name of plant	Part used	Precursor	References
Aloe Vera	Leaves	Copper Sulphate	151

<i>Azadirachta indica</i>	Leaves	Cupric acetate (monohydrate)	152
<i>Caloropis procera</i>	Leaves	Cupric acetate	153
<i>Euphorbia esula</i>	Leaves	Copper chloride dihydrate	154
<i>Punica granatum</i>	Peel of fruit	copper sulfate	155

Table 4: Plant Extract mediated n Green Synthesis of Copper oxide Nanoparticles

4. APPLICATIONS OF Ag, Au, ZnO, CuO NANOPARTICLES:

Due to inhibitory action on pathogens Silver nanoparticles (Ag-NPs) are broadly used as the antibacterial and antifungal agents in the form of protective coatings over biomedical instruments inclusive of wound dressings, catheters, dental composites, as well as in certain implants employed in cardiovascular treatments. Silver nanoparticles are also used as nano-bio sensing materials as well as in agricultural engineering products [156]. Prosthetic Silicone valve coated with Ag-NPs was the first cardiovascular device. The role of Ag-NP coating in this device was to reduce or avoid microbial contamination in order to reduce the chances of inflammatory reactions.

Ag-NPs are also used as protective surface coatings on stents as Ag-NPs are antibacterial as well as antithrombogenic in nature [157]. It has been also reported that use of Silver nanoparticles in Ag-NPs into orthodontic adhesive enhances device strength increasing resistance to bacteria [158]. Ag-NP coated dressings are used in wound dressings as potent antibacterial agent to treat burns, chronic ulcers, pemphigus, and toxic epidermal necrosis [159]. It has been also reported that combination of Chitosan Ag-NPs used in wound demonstrated potential antibacterial activity as compared to essentially 1% Ag sulfadiazine alongside the deposition of less Ag. [160] Roe et al. (2008); Hussain et al. (2006); and Chou et al. (2008) reported that, silver nanoparticles are used as antibacterial coating over catheters. Class of Polyurethane catheters is coated with

silver nanoparticles which are antibacterial catheters [160-162]. Silver nanoparticles are used in Electronic components and transportation as Sensors, electrodes and integrated circuit [163-165]. Other than this silver nanoparticles find enormous applications in environmental applications like air disinfection, wastewater treatment, drinking water purification, ground water disinfection. [166-169]. Also Ag-NPs find application in Nutraceuticals, food safety and food packaging [170-172]. It has been reported that, nanoparticles are the most accurate agents for drug delivery as well as cancer therapy. In cancer therapy these nanoparticles are successful in overcoming the drawbacks caused by traditional chemotherapeutic agents as these agents were not accurate enough in targeting the cancerous cells. Nowadays bio-based nanoparticles are filled with chemotherapy drugs and attached to RNA like structures. These nanoparticles with chemotherapeutic drugs accurately targets the cancerous cells and releases the drugs into cancer cells [173]. Das et al. (2013) & Alt et al.

(2004) reported that plant mediated nanoparticles are effective against various cancer cell lines such as Hep 2, HCT 116 and Hela cell lines. The enhanced cytotoxic effect is due to secondary metabolites. The plant material based nanoparticles play active role in regulating the cell cycle [174-175]. Russier-Antoine et al. 2008 reported that, gold nanoparticles could bind with negatively charged DNA molecules substituting citrate ions resulting into formation of novel DNA-nanoparticle. Such probes are used in diagnosis of pathogenic and genetic diseases. [176] Huang et al. 2008 illustrated that, gold nanoparticles coupled antigen can be used for diagnostic purposes. Antigen conjugated gold nanoparticles biconjugated with antibodies. Thus, Biosensors for immunoassays in human serum have been developed from gold nanoparticles. [177] Gold nanoparticles stabilized by Phthalocyanine demonstrated a potential delivery vehicle for photodynamic therapy. [178] Besides these Gold nanoparticles are also employed as biosensors by coupling with GDP molecules and also as immunosensors. Gold nanoparticles have been in active use in the identification of chemical and biological agents. The visualization methods with the use of GNP and optical microscopy [179]. Zinc oxide (ZnO) nanoparticles have numerous bio applications. It has been reported that drug metronidazole benzoate has much effect on biological membranes [180]. As ZnO nanoparticles have essential blue and near-emission, these are used in cellular imaging. Several studies have reported that, ZnO nanoparticles are used for cancer cell imaging, transferrin-conjugated green fluorescent ZnONPs were utilized with least cytotoxicity [181].

Zinc oxide nanoparticles have found to play significant role in efficient gene delivery. ZnO QDs were layered using positively charged poly(2-(dimethylamino)ethyl methacrylate) (PDMAEMA) polymers which are utilized for condensing pDNA for gene delivery [182].

ZnO nanomaterials various desirable characteristics like semiconducting properties such as biosensing, strong adsorption capability, high isoelectric point, and high catalytic efficiency (IEP; ~9.5) which are appropriate for adsorption of certain proteins such as antibodies and enzymes with less IEPs by electrostatic interaction [183]. Along with silver nanoparticles copper oxide nanoparticles (CuO) are most frequently used antimicrobial agents due to their killing effect on pathogens. It has been reported that CuO nanoparticles have capacity to kill more than 99.9% of Gram-positive and negative bacteria within 2 hrs. of exposure, if desired dose is administered or applied. In several studies it has been reported that, CuO nanoparticles reduce the chances of nosocomial infections. Nanoparticles coated bed sheets is considered to be one of the most recent innovation in health care practices in order to reduce the chance of acquiring nosocomial infections [184]. Copper oxide nanoparticles also show considerable antifungal properties as well as effective against biofilm development. Due to wound healing activity CuO nanoparticles are also incorporated in wound dressings [185-186].

Note: All figures used in this article are created by authors.

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