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Current developments in green synthesis of metallic nanoparticles using plant extracts: a review

Morteza Yadi[a], Ebrahim Mostafavi[b], Bahram Saleh[b], Soodabeh Davaran[a,c], Immi Aliyeva[c,d], Rovshan Khalilov[c,e], Mohammad Nikzamir[f], Nasrin Nikzamir[g], Abolfazl Akbarzadeh[a,b,c,h,i], Yunes Panahi[j] and Morteza Milani[a]

[a]Department of Medical Nanotechnology, Faculty of Advanced Medical Science, Tabriz University of Medical Science, Tabriz, Iran; [b]Department of Chemical Engineering, Northeastern University, Boston, MA, USA; [c]Joint Ukrain-Azerbaijan International Research and Education Center of Nanobiotechnology and Functional Nanosystems, Drohobych Ukraine and Baku, Azerbaijan; [d]Department of Environmental Engineering, Azerbaijan Technological University, Ganja, Azerbaijan; [e]Institute of Radiation Problems, National Academy of Sciences of Azerbaijan, Baku, Azerbaijan; [f]Department of industrial Engineering, Islamic Azad University, Tehran, Iran; [g]Department of Chemistry, Islamic Azad University, Science and Research Branch, Tehran, Iran; [h]Biotechnology Research Center, Tabriz University of Medical Sciences, Tabriz, Iran; [i]Universal Scientific Education and Research Network (USERN), Tabriz, Iran; [j]Pharmacotherapy Department, School of Pharmacy, Baqiyatallah University of Medical Sciences, Tehran, Iran

ABSTRACT

Metal nanoparticles (MNPs) produced by green approaches have received global attention because of their physicochemical characteristics and their applications in the field of biotechnology. In recent years, the development of synthesizing NPs by plant extracts has become a major focus of researchers because of these NPs have low hazardous effect in the environment and low toxicity for the human body. Synthesized NPs from plants are not only more stable in terms of size and shape, also the yield of this method is higher than the other methods. Moreover, some of these MNPs have shown antimicrobial activity which is consistently confirmed in past few years. Plant extracts have been used as reducing agent and stabilizer of NPs in which we can reduce the toxicity in the environment as well as the human body only by not using chemical agents. Furthermore, the presence of some specific materials in plant extracts could be extremely helpful and effective for the human body; for instance, polyphenol, which may have antioxidant effects has the capability for capturing free radicals before they can react with other biomolecules and cause serious damages. In this article, we focused on of the most common plants which are regularly used to synthesize MNPs along with various methods for synthesizing MNPs from plant extracts.

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Introduction

Nanotechnology is a wide-range area of science which opens a new world of diagnostic and treatment for many autoimmune diseases such as inflammation and cancer. Nanoparticles (NPs) have very specific physicochemical and biological properties due to their size (1–100 nm). Subject to their morphology, size, and distribution, NPs display better properties than the bulk materials [1,2]. Additionally, the higher surface area to volume ratio of NPs is important and plays a vital role for more efficiency in biological applications. In particular, silver and gold NPs have a lot of applications such as catalysis [3], biosensors [4], antimicrobial [5], photo thermal therapy [6], and pharmaceutical production [7]. As an example, Figure 1 shows an overall schematic of general applications of metal nanoparticles (MNPs) in biological field.

Nanotechnology

Nanotechnology has become one of the more important technologies applied in all areas of science. Nanotechnology can be defined as the science and engineering behind the design, synthesis, characterization, manipulation, and application of functional materials and devices in which at least one dimension is in the range of nanometer, 1–100 nm. Nanoscale materials contain specific properties including melting point, charge capacity, tensile strength, and so on. During the past few decades nanotechnology has grown enormously, not only in terms of the number of the applications (such as biomedical, healthcare, drug delivery, environment, electronic, magnetic, space science, sensors, energy storage and conversion, and so on), also the number of the industrial R&D companies whose qualitatively applying the nanotechnology in their field are extremely increasing.

Synthesis of nanoparticles

There are different physical and chemical methods for successfully synthesizing NPs (Figure 2(a)). One can categorize all these methods into two main approaches that can apply to any research in the field of nanoscale science: (1) the

CONTACT Abolfazl Akbarzadeh akbarzadehab@tbzmed.ac.ir Biotechnology Research Center, Tabriz University of Medical Sciences, 5154853431 Tabriz, Iran; Mortaza Milani mohammadmilano@gmail.com Department of Medical Nanotechnology, Faculty of Advanced Medical Science, Tabriz University of Medical Science, 5154853431 Tabriz, Iran; Yunes Panahi yunespanahi@yahoo.com Pharmacotherapy Department, School of Pharmacy, Baqiyatallah University of Medical Sciences, Tehran, Iran

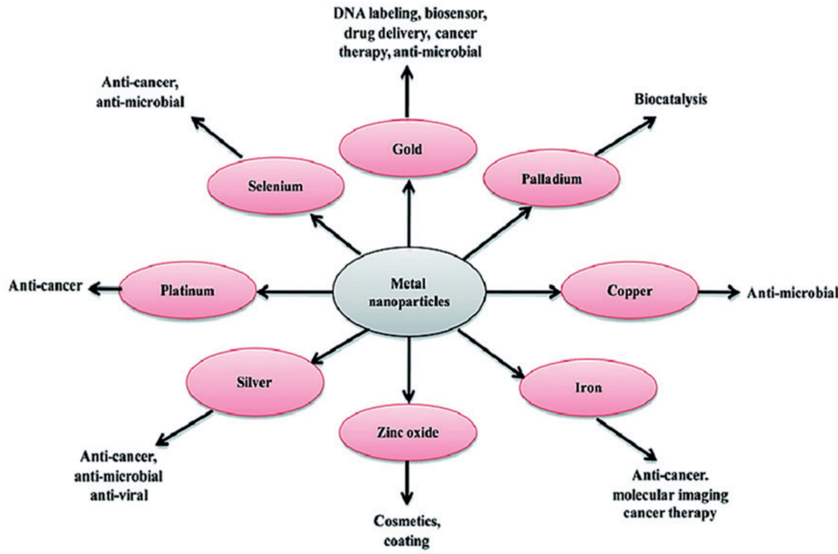

Figure 1. Different types of metal nanoparticles (MNPs) and their applications in biotechnology [8].

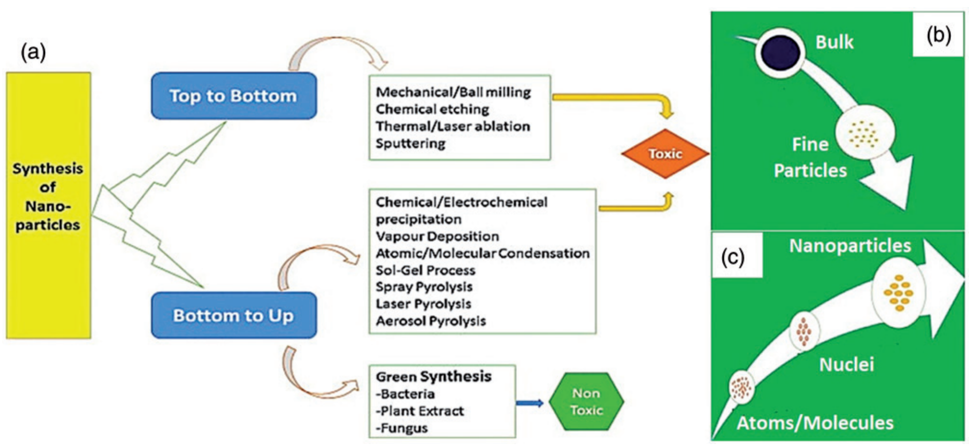

Figure 2. (a) Different approaches for synthesis of nanoparticles. Protocols employed for synthesis of NPs (b) top to bottom approach and (c) bottom to top approach [9,15].

top-bottom and (2) the bottom-top approach (Figure 2(b,c)). Each of which has specific characterization and application.

Top-bottom

In top-bottom approaches, the desired bulk of materials break down into the particles in the nanosized range (Figure 2(a)) [9] e.g. mechanical milling/alloying and sputtering [10,11] are examples of methods for reducing the size of the particles. This method can cause the surface imperfections in the product which causes serious restriction since the surface structure of a material plays a vital role in surface chemistry and physical properties of a material [8].

Bottom-top

In bottom-top approaches, the synthesis process start with the self-assembly of atoms/molecules into nuclei and afterwards the formation of particles in the nanoscale range (Figure 2(c)) [9] i.e. co-precipitation, sol–gel, and atomic condensation [12–14] are examples of this approach. The bottom-top synthesis mostly relies on chemical and biological methods of production.

In both top-bottom and bottom-top approaches, nanoparticles preparation is relying on utilizing chemical and physical methods which are quite expensive and more likely hazardous to the environment which involve use of toxic and dangerous chemicals that are responsible for various biological risks [8,9]. Since, plant-mediated NPs synthesis approach not only does not involve physical and chemical methods, also this method is environment-friendly, biocompatible, highly stable, therefore this method has attracted a substantial attention among researchers worldwide.

Plant extract

Many chemical methods are chosen for synthesizing NPs because of their quick reaction time [16] and their capability to produce monodispersed NPs [17]. Methods such as chemical reduction [18], electrochemical reduction [19], photochemical reduction [20], and heat evaporation [21] have all been employed for synthesis of NPs. Although all these methods are able to successfully produce the MNPs, they have few disadvantages such as the high price of the process and not being environment friendly since they make lots of pollution in the environment because of using toxic solvents and reducing agent [22]. Although chemical stabilizers are used more than plant extract, that materials are not safe for the environment and for a human health condition. To avoid these drawbacks, green chemistry approaches have been employed for production of NPs [23–25] which is simple, convenient, less energy-intensive, eco-friendly and minimize the usage of unsafe materials, and maximize the efficiency of the process.

Plant-based polyphenols are considered to be the largest groups of natural antioxidants with extraordinary potential as drugs, nutraceuticals, and food additives [26]. The underlying principle in the green synthesis approaches is that the phytochemicals present in the plant parts serve the twin role of a natural reductant besides being a nanoparticle stabilizer [27]. According to other reports [28], highly stabilized NPs may be quickly synthesized from plant extracts rather than microbe-based synthesis. Therefore, the plant extract could be an efficient approach for reducing NPs early material plus stabilized that.

Green synthesis of metallic nanoparticles (MNPs)

Copper oxide nanoparticles (CuO NPs)

K. Rayapa Reddy reported the green approach of CuO NPs synthesis using *Calotropis procera* which belongs to the asclepiadaceous family [29]. These NPs are widely used in many applications such as catalysis because of their narrow band gap, used in photo catalytic properties [30]. *Calotropis procera* used in the many treatments like diseases of spleen, piles, tumors, etc.

In this article, fresh leaves of Calotropis were washed with distilled water and after sun-dried, 25 g of the plant, which cut into fine peaces, added to deionized water for boil until the color of the solution changes into yellow. After that, cupric acetate was added to the solution and boiled again to reach a blue-green paste. This powder calcinated at 700 °C until the color of material changed into the black. In another study, Alaa Y. Ghidan *et al*. [31] reported the green synthesis of copper NPs using *Punica granatum* peels extract. Punica granatum fresh peels are obtained and washed for several times. After peels dried, they turned into powder and mixed with sterile distilled water and boiled until the color of solution change to yellow.

To synthesize CuO NPs, copper acetate powder was dissolved in the water and stirred with magnetic stirrer. After that, *P. granatum* extract was added to the solution; at the first step, the color of solution turned to to green and then it turned to brown which shows the formation of monodispersed Cu NPs.

Ijaz *et al*. [32] reported the green synthesis of copper NPs from fresh leaves of *Abutilon indicum*. The fresh leaves of *Abutilon indicum* were collected and washed gently to remove dust particles as well as dried and shaded parts. Next, the leaves were pulverized and seived using a 200-nm mesh sieve to be used as fuel.

To synthesize CuO NPs, Copper (II) nitrate trihydrate was mixed with *Abutilon indicum* extract in double-distilled water. The solution was homogenized for 2–5 min with constant stirring using a magnetic stirrer. Next, combustion reaction was performed on the mixture using a pre-heated muffle furnace at the temperature $400 \pm 5\,^{\circ}C$ to produce CuO NPs. The resultant mixture was filtered to remove the ash contents of the plant extracts.The solution was washed with distilled water, followed by methanol to remove impurities.

Palladium nanoparticles (Pd NPs)

Owing to various applications of palladium NPs especially in the field of medicine, the approaches for synthesizing these NPs have been extensively expanded recently. One of this approaches is a green synthesis which uses a different plant to obtain NPs. In a report, Pd NPs synthesized using *Filicium decipiens* extract to the specified antibacterial efficacy of these NPs [33]. Fresh leaves of *F. decipiens* were collected and washed with water, next they were dried and cut to small pieces and mixed with distilled water to obtain the extract. The leave extract mixed with 1 mM $PdCl_2$ solution and isolated in a dark place at room temperature for a few days.

Another study has shown an approach to manufacture Pd NPs with *Hippophae rhamnoides* Linn extract which obtained by Nasrollahzadeh *et al.* [34]. In this study leaves of the plant were collected, turned into the powder and mixed with the hydroalcoholic solution. The extract was added dropwise to palladium chloride and after few minutes, the yellow color of the solution change to dark brown that shows the synthesized of NPs was complete.

Zinc oxide nanoparticles (ZnO NPs)

ZnO NPs have attracted a lot of research interest because of their significant and important roles as catalysts, ceramic resistors, super-conducting materials, gas sensors, as well as their roles in biological fields and in the energy sector [15,35]. Furtherore, ZnO NPs are inexpensive and safe; US FDA has enlisted ZnO as GRAS (generally recognized as safe) metal oxide [36]. ZnO NPs have been used in cancer therapy, drug delivery and they posses antibacterial properties [37,38].

The green approach of ZnO NPs synthesis is reported by Dhanemozhi *et al.* [39]. In this article, powder of the green tea leaf was dried and added to distilled water and mixed in constant speed. After that, zinc acetate was dissolved in water to obtained zinc acetate solution. The extract was added to the solution and dried for 12 h to yield white ZnO NPs and then desiccated at 100 °C to obtained NPs.

In the other study, ZnO NPs was manufactured using fruit peel extract [40]. In this method, fruits being peeled and dried for 12 h, then turned into the fine powder. Afterwards, the powder was mixed with de-ionized water stirring well. The filtered solution used for NPs synthesis. This solution was added to the zinc nitrate and mixed with a stirrer and then placed in a water bath to complete the process. The mixture was heated at 400 °C to produce white-colored powder.

Orange peel, grapefruit peel, and lemon peel were used in this experiment.

Cerium oxide (CeO_2) nanoparticle

CeO_2 is a semiconductor metal oxide with 3.19 eV band gap energy and wide range of applications such as bio-imaging, catalyst, sun screen cosmetics, and antibacterial activity. This metal oxide has been synthesized by various chemical and physical approaches [41]. For instance, egg withe, plant extract, and honey have been used to synthesize CeO_2 NPs.

Gloriosa superba leaves used for cerium NPs synthesis in which 3.72 g of Cerium salt ($CeCl_3$) was added to the extraction product of 10 g boiled leaves in 100 ml of distilled water and followed with stirring for 4–6 h in 80 °C until the color of solution turned to yellowish brown (Figure 3).

Another study has shown that CeO_2 nanoparticle could be synthesized using honey [42]. In this article, 12 g of dehydrated cerium salt was dissolved in 30 ml of deionized water and specified amount of honey was dissolved in specified amount of distilled water. After that, these two solutions were added to each other and stirred in 60 °C for 6 h to obtain a light yellow color resin (Figure 4).

Silver nanoparticle (Ag NPs)

Noble metal NPs have provoked sufficient attention because their structures display extensively unique and better physical, chemical, and biological properties, and functionalities due to their ultra-small sizes [43]. Among the various NPs, silver NPs (Ag NPs) have fascinated considerable researcher's attention because of their attractive properties, such as a high electrical and thermal conductivity, surface-enhanced Raman scattering, chemical stability, high catalytic activity, and antimicrobial activities [44]. Amongst other noble metals, the synthesis of silver NPs has established great significance as an antimicrobial agent against the ever-increasing hazard postured by antibiotic-resistant microbes [45,46]. Notably, physical and chemical methods for synthesizing of Ag NPs with desired size and shape as well as antimicrobial activities have been reported previously [47]. The unique properties of Ag NPs are mainly advantageous for cancer therapeutics since they led to an enhanced chemotherapeutic efficacy along with the minimal systemic toxicity [48,49]. Nowadays, nanoparticle-based combinatorial therapies, utilizing NPs with anticancer activity in combination with a chemotherapeutic agent have been employed by several researchers.

Chemical synthesise

For synthesize of Ag NPs from the chemical approach, you need $AgNO_3$ and Trisodium citrate ($Na_3C_6H_5O_7$). In this method, 5 ml $AgNO_3$ 10–3 M is prepared and heat that until received to the boiling temperature, then 5 ml $Na_3C_6H_5O_7$ was added dropwisely to the $AgNO_3$ while stirring vigorously. After that solution was heated until changing color to brown. Then the heat was removed and stirring was continued at the solution temperature receive to room temperature and the color of the particles is turned to the yellow [50].

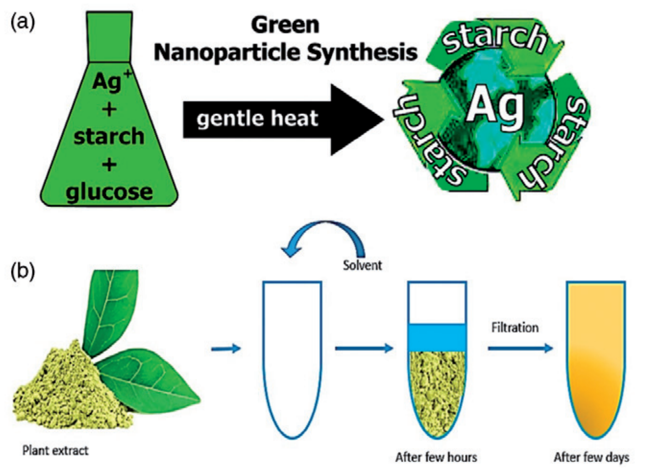

Figure 4. (a) a completely "green" synthetic method for producing silver nanoparticles [25]. (b) Plant extract isolation method. In this approach at first step chopping the plant parts then dissolve in the solvent and mixed with stable speed; then put the container in the dark place for few days.

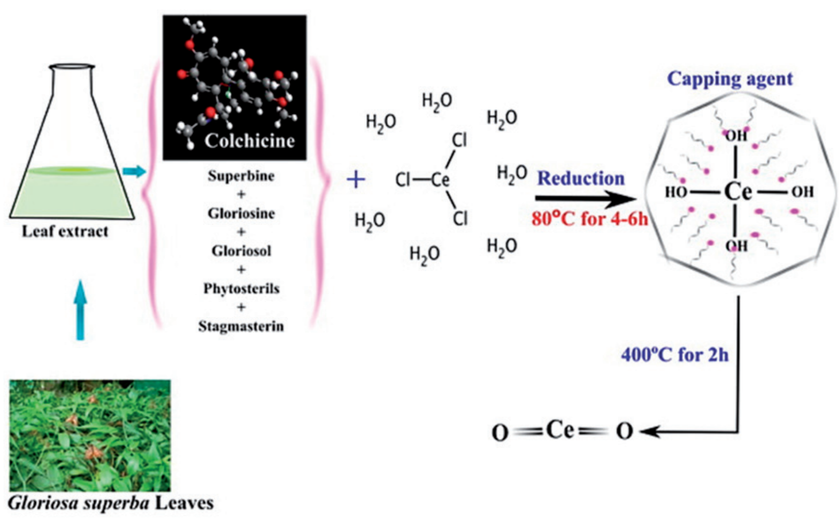

Figure 3. CeO_2 nanoparticle synthesized using *G. superba* leaf extract [41].

Green synthesise

In this approach, plant extracts have been used usually as reducing agent instead of chemical reducing agents. A group of researchers successfully stabilized and synthesized Ag NPs by a completely green method (Figure 4) which is a mild, renewable, inexpensive method and no need to use any toxic reducing agent [25]. For example, Velmurugan *et al*. used *Zingiber officinale* root extract; The *Z. officinale* root extract was used without any additional modifications. The *Z. officinale* root extract and metal ion reaction mixture slowly turned a brown-yellow shade for Ag NPs. This was the initial means of detection for the formation of Ag NPs. About 3.0 ml sample was withdrawn from the reaction mixture at different time breaks, and the maximum absorbance was measured using a UV-1800 UV–VIS spectrophotometer. Later, the reaction mixture was filtered through 0.22-μm Steritop Millipore filters, which attach to a vacuum pump, and then filtrates were centrifuged at 90,609×*g* for 15 min to separate Ag NPs [51].

In another work, Ahmed *et al*. [52] have worked on green synthesis of Ag NPs using *Azadirachta indica* extrancts. The plant extract acts both as reducing agent as well as capping agent. Fresh leaves were cleaned with running tap water, followed by double distilled water and air dried at room temperature. Finely cut leaves were boiled in double distilled water for 30 min and the extract was stored at 4 °C before use. They also proposed a new, simple, single step, rapid green method for synthesizing Ag NPs by using plant extracts of *Crotolaria retusa* as well as *Terminalia arjuna* as reducing and stabilizing agents [53,54]. The synthesized AG NPs demonstrated high catalytic activity as well as excellent antimicrobial properties against both gram-negative and gram-positive bacteria.

For synthesis of Ag NPs,1 mM solution of silver nitrate was prepared in an Erlenmeyer flask and 1–5 ml of plant extract was added separately to 10 ml of silver nitrate solution. This setup was incubated in a dark chamber to minimize photoactivation of silver nitrate at room temperature. Reduction of Agþ to Ag0 was confirmed by the color change of solution from colorless to brown. Its formation was also confirmed by using UV–Visible spectroscopy.

Gold nanoparticle (Au NPs)

One of the most important classes of metal NPs is that made of noble metals such as platinum (Pt), gold (Au), and silver (Ag) [55,56]. Among the noble metal, Au NPs appeal great and large attention due to their ability to interact with light by SPR [57,58]. The recent progresses in the field of nanotechnology have revealed that Au NPs have the potential to serve as the building blocks for plasmonic devices and future photonic [59,60]. In addition, Au NPs have received the most attention as bio-molecular conjugates [57,61].

Gold NPsNPs have been employed in numerous fields, such as catalysts for numerous environmental developments, antimicrobial agents against a wide range of microorganisms, and so on [62–64]. Usually, these applications are related to the size, shape, and surface morphology of particles, which can be adjusted for an specific application [65,66]. Hence, the controlled fabrication of gold NPs in terms of size and shape can supplement biological, optical, catalytic, and electronic of gold NPs that can have broader applications [67]. Hitherto, a number of methods have been developed to synthesize gold NPs, including physical, an electrochemical, photochemical and liquid chemical reduction methods NPs [68].

Chemical synthesise

Cetyl Tri-methyl Ammonium Bromide (CTAB) is a commonly used surfactant for the structuration of nonspherical Au NPs, and the role of its purity and apparatus of action have been debated [69]. For example, the addition of ascorbic acid to a mixture of CTAB, $HAuCl_4$ reduced orange Au^{3+} to almost colorless Au^{+}. The rapid addition of NaOH then encouraged the formation of anisotropic Au NPs, whose color changed from

Table 1. Green synthesis of Au and Ag nanoparticles by different researchers using plant extracts.

Plant	Size (nm)	Plant's part	Type of NPs Au	Type of NPs Ag	References
Alternanthera dentate	50–100	Leaves		*	[74]
Coleus forskohlii	silver (5–15) gold (5–18)	Root	*	*	[75]
Curculigo orchioides	15–18	Rhizome		*	[76]
Digitaria radicosa	90	Leaves		*	[77]
Dioscorea alata	10–20	Tuber		*	[78]
Diospyros paniculata	14–28	Root		*	[79]
Elephantopus scaber	11–100	Leaves		*	[80]
Emblica officinalis	10–70	Fruit		*	[81]
Abutilon indicum	1–20	Leafs	*		[82]
Butea monosperma	gold (10–100) silver (20–80)	Leafs	*	*	[83]
Carica papaya	15–28	Leaves	*		[84]
Gymnema sylvestre	72.8	Leaves	*		[85]
Hibiscus sabdariffa	10–60	Leaves	*		[86]
Hygrophila spinosa	50–80	Leaves	*		[87]
Ficus benghalensis	40	Bark		*	[88]
Ocimum sanctum	1–50, 10–300, 50–300, > 200	Leaves	*		[89]
Parkia roxburghii	5–25	leaves	*	*	[90]
Piper longum	20–200	Fruits	*		[91]
Podophyllum hexandrum L.	5–35	Leaves	*		[92]
Rhus chinensis	20–40	Galls	*		[93]
Stachys lavandulifolia Vahl	34–80	Aerial part	*		[94]

pale blue to dark red within one day. NaOH plays a role in the splitting of the NPs, as the use of $NaBH_4$ instead only yields spherical and rod-shaped Au NPs [70,71].

Green synthesise

Due to the chemical compound hazardous and their effect on the environment, researchers have used an eco-friendly structure such as the plant extracts. For instance, 10 g of *Sphaeranthus indicus* fresh leaves were transferred into 100 ml of boiling double purified water and kept for 10 min. After that, the hot leaf extract was filtered through Whatman filter paper no. 1, and then used for further process. The reaction was carried out in 250 ml conical flask containing 100 ml of 1 mM $AuCl_4$ solution plus 10 ml of *S. indicus* leaf extract and stirred well for 30 min. After 30 min, the light yellow-colored mixture changed to wine red color, representative the synthesis of Au NPs (pH 5.4) [72].

In another work by Ahmed *et al.* [73], different plant parts (leaf, bark, stem, root, etc.) were washed with distilled water, chopped into small pieces and boiled in distilled water to obtained extract. Next, the purified extract is mixed with the auric salts solution at room temperature to attain Au NPs in a one pot reaction. There is a collection of plants used in the synthesis of gold and SNPs in Table 1.

Conclusions

The use of plant extracts for making metallic nanoparticles is simple, convenient, inexpensive, easily scaled up, less energy-intensive, eco-friendly, and minimize the usage of unsafe materials and maximize the efficiency of the process. It is specifically suitable for making NPs that must be free of toxic contaminant as required in biomedical and therapeutic applications. The plant extract-based synthesis of NPs can provide not only more stable in terms of size and shape, also the yield of this method is higher than the other physical and chemical methods.

Disclosure statement

No potential conflict of interest was reported by the authors.

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