

Review Article

Honey Mediated Green Synthesis of Nanoparticles: New Era of Safe Nanotechnology

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With the advent of nanotechnology, many related industries rapidly developed over the recent past. Generally, top-down and bottom-up approaches are the two major processes used to synthesize nanoparticles; most of these require high temperatures, vacuum conditions, and harsh/toxic chemicals. As a consequence, adverse effects impacted organisms including humans. Some synthesis methods are expensive and time-consuming. As a corollary, the concept of "green nanotechnology" emerged with the green synthesis of nanoparticles commencing a new epoch in nanotechnology. This involves the synthesis of nanomaterial from microorganisms, macroorganisms, and other biological materials. Honey is documented as the world's oldest food source with exceptional medical, chemical, physical, and pharmaceutical values. Honey mediated green synthesis is a relatively novel concept used during the past few years to synthesize gold, silver, carbon, platinum, and palladium nanoparticles. Honey acts as both a stabilizing and a reducing agent and importantly functions as a precursor in nanoparticle synthesis of nanoparticles provides a simple, cost effective, biocompatible, reproducible, rapid, and safe method. The special activity of honey functionalized nanoparticles may provide valuable end products with numerous applications in diverse fields.

1. Introduction

Over the past decade, nanomaterials emerged as promising commodities in many fields including cosmetics, healthcare, biomedical, food and feed, drug-gene delivery, environment, health, mechanics, optics, chemical industries, electronics, space industries, energy science, catalysis, light emitters, single electron transistors, nonlinear optical devices, and photoelectrochemical applications [1–10].

The theoretical concept of nanotechnology was first described in 1959 by physicist Richard Feynman [11]. Nanotechnology is the ability to understand, control, and manipulate matter at the level of individual atoms and molecules [12]. The prefix nano is derived from the Greek word *nannos*. Nanotechnology refers to objects that are sized on a nanometer scale where at least one of the dimensions of a particle must be less than 100 nm. Recent investigations revealed that properties and potential applications of nanoparticles varied with the phases, sizes, and morphologies of these particles [13, 14]. Thus, controlled synthesis of nanomaterial with novel morphologies has gained much attention.

Synthesis of nanoparticles can be categorized into topdown or bottom-up approaches [15]. Top-down approach involves a process of breaking down of large structures to create small structures. Physical techniques such as lithography [16], laser ablation [17], sputtering deposition [18],



FIGURE 1: Schematic diagram of currently used chemical and physical methods of nanoparticle synthesis.

pulsed electrochemical etching [19], and vapor deposition [20] are among the most commonly used top-down methods. Bottom-up approaches such as sol-gel processing [21], chemical vapor deposition [22], plasma or flame spraying synthesis [23], laser pyrolysis [24], and microemulsion [25] involve the synthesis of material, atom by atom, molecule by molecule, or cluster by cluster. Figure 1 illustrates different chemical and physical methods of nanoparticle synthesis. Most of these usually require toxic and harsh chemical additives, for example, dimethyl formamide, hydrazine, and sodium borohydride, and physical conditions such as high temperatures, vacuum conditions, and expensive equipment. These methods may result in toxic and harsh products which may pose biological risks to the environment; due to high surface charge and high surface area of nanoparticles, harsh chemicals may remain adsorbed onto nanoparticles. Releasing these chemicals into the environment may cause adverse effects on organisms including microorganisms, plants, invertebrates, and vertebrates including humans at various trophic levels [26]. Therefore, it is essential to optimize green methods for nanoparticle synthesis.

In this review, we initially focus on various methods of green synthesis of nanoparticles. Furthermore, we describe physical and chemical characteristics of natural honey. Thereafter, honey mediated green synthesis of various types of nanoparticles is emphasized. Finally, we highlight the key challenges of green synthesis of nanoparticles.

2. Green Synthesis

Recent developments in nanotechnology focus on environmentally friendly, cost effective synthesizing methods. Green synthesis of nanoparticles is an ecofriendly and safe mode of synthesis of nanomaterial using biological resources. This green approach has opened up a new era of safe nanotechnology. Figure 2 compares conventional synthesis methods such as physical and chemical methods with those of green synthesis of nanoparticles. Furthermore, it illustrates the comparison between microorganisms mediated green synthesis and more productive honey mediated green synthesis.

Table 1 lists various methods of green synthesis of nanoparticles and special features manifested by the resultant products. These presently include different approaches such as use of microbial systems, plant systems, and biological methods. Bacteria were used to synthesize several nanoparticles including gold, silver, silver oxide, titanium dioxide, and cadmium sulfide [24-30] while fungi were used to synthesize silver [31-36], titanium dioxide [29], and cadmium sulfide [30]. Furthermore, actinomycetes such as Rhodococcus sp. were used to synthesize gold nanoparticles [25]. Recent experiments revealed the immense potential of algae, particularly in synthesizing silver, gold, zinc oxide, and iron oxide nanoparticles [37-43]. Au-Co₃O₄ nanoparticles were synthesized using a virus mediated method [44]. Moreover, leaf extracts, seed extracts, root extracts, bulbs, and latex of plants were used to synthesize gold, silver, and palladium nanoparticles [45-56]. Biological materials such as honey, starch, and ascorbic acid were used to synthesize gold, silver, palladium, carbon, and platinum nanoparticles [57-63].

3. Natural Honey

Natural honey, documented as the world's oldest food source, is an excellent food with high energy and nutritious value [61]. It is produced by *Apis mellifera* (*A. mellifera*; honey bee) from plant nectar, secretions, and excretions [67].

Natural honey was applied for medicinal purposes since ancient times. The first evidence of the use of honey as a medication dates back to 2100–2000 BC where a Sumerian tablet contained honey as a drug and an ointment [68]. Furthermore, many animal based studies evidenced the application of honey in cardiovascular diseases, where it affects cardiovascular risk factors such as hyperlipidemia and production of free radicals [69].

4. Physical Characteristics of Honey

Natural honey is a sticky and viscous solution, depending on its water content [70]. It also has the ability to absorb and hold moisture from the environment. Normal honey with a water content of 18.8% or less will absorb moisture from air of a relative humidity of above 60% [71].

The color of liquid honey varies from clear and colorless (alike water) to dark amber or black. The color of honey



FIGURE 2: Comparison of conventional methods and green synthesis methods of nanoparticle synthesis.

appears in all shades of yellow and amber; color varies with botanical origin, age, and storage conditions, but transparency or clarity depends upon the amount of suspended particles such as pollens [72].

5. Chemical Composition of Honey

Honey is one of the healthiest food sources since immemorial time. It consists of 80–85% carbohydrate (mainly glucose and fructose), 15–17% water, 0.1–0.4% protein, 0.2% ash, and minor quantities of amino acids, enzymes, and vitamins as well as other substances such as phenolic antioxidants. However, the precise chemical composition and physical properties of natural honey differ according to the plant species on which the bees foraged, differences in climatic conditions, and vegetation [70].

Fructose (32.56 to 38.2%) and glucose (28.54 to 31.3%) as the major carbohydrates present in honey represent 85–95% of total sugars and are readily absorbed in the gastrointestinal tract. Other sugars include disaccharides such as maltose, sucrose, isomaltose, turanose, nigerose, melibiose, panose, maltotriose, and melezitose. A few oligosaccharides are also present. Honey contains 4 to 5% fructooligosaccharides, which serve as probiotic agents [73].

Honey contains proteins in 0.1–0.5% quantities, which differ greatly with types and origin of honey [70]. Natural honey is also rich in vitamins such as C and B₁ (thiamine) and B₂ complex vitamins including riboflavin, nicotinic acid, B₆, and pantothenic acid [74].

Almost all natural honey varieties contain flavonoids (apigenin, pinocembrin, kaempferol, quercetin, galangin, chrysin, and hesperetin), phenolic compounds (ellagic, caffeic, and p-coumaric and ferulic acids), and bioactive compounds such as ascorbic acid, tocopherols, catalase (CAT), superoxide dismutase (SOD), and reduced glutathione (GSH) as antioxidant enzymes [75].

The concentration of mineral compounds ranges from 0.1% to 1.0%. Potassium is the major metal, followed by calcium, magnesium, sodium, sulphur, and phosphorus. Trace elements include iron, copper, zinc, and manganese [76].

A variety of enzymes, for example, oxidase, invertase, amylase, and catalase, are constituents of honey; the main enzymes are invertase (saccharase), diastase (amylase), and glucose oxidase. Dextrin and maltose are produced from long starch chains by the activity of amylase enzyme [70]. Each of these minor constituents is known to have distinctive nutritional or medicinal properties and the unique blend accounts for the varied and different applications of natural honey.

6. Honey Mediated Green Synthesis of Nanoparticles

Special chemical properties of honey render its usage in green synthesis of nanoparticles. As a consequence, honey mediated synthesis offers several advantages over the microorganism mediated methods; it is relatively a rapid process compared to the microbial method. Also, microorganisms must be cultured with extreme care and there is a time lag for the conversion of nanoparticles by microorganisms. Furthermore, separation of nanoparticles from microorganisms can be a difficult task.

Based on literature [77], Figure 3 illustrates the proposed mechanism and Table 2 lists the reducing and stabilizing agents and specific temperatures used in honey mediated biosynthesis of different nanoparticles. While the generalized procedure was used for gold, silver, and palladium nanoparticles, carbon and platinum nanoparticles had utilized specific conditions.

7. Gold Nanoparticles

Philip synthesized gold nanoparticles by reducing HAuCl₄ with different volumes of diluted honey where the reduction speed of gold particles increased with the increasing volume of honey [55]. Accordingly, fructose as the primary ingredient of honey may have acted as the prime reducing agent collectively with vitamin C, that is, a mild reducing agent. Furthermore, he postulated that this reduction may be facilitated by the presence of H_2O_2 and gluconic acid, produced when diluting honey with distilled water. Fructose is claimed as the possible reducing agent for the reaction while proteins present in honey were responsible for the stabilization of the nanoparticles [61].

Olaitan and colleagues reported green synthesis of gold nanoparticles at room temperature using honey as both a

Biological resource	Types of nanoparticle synthesized	Organism/s	Size and special features	
Microbial systems	·			
	Au	Acinetobacter sp. SW 30	20 ± 10 nm [27]	
		Rhodococcus sp.	5–15 nm [28]	
	Aσ	Bacillus subtilis	5–60 nm [29]	
Bacteria	119	Pseudomonas stutzeri AG259	Up to 200 nm [30]	
	Ag ₂ O	Lactobacillus mindensis	2–20 nm [31]	
	TiO_2	Lactobacillus sp.	15–35 nm [32]	
	CdS	Lactobacillus sp.	4.93 ± 0.23 nm [33]	
		Aspergillus terreus	1–20 nm [34]	
Fungi	Ag	Saccharomyces spp.MKY3 (yeast strain MKY3)	2–5 nm [35]	
0		Fusarium oxysporum	25–50 nm [36], ~50 nm [37], 10–20 nm [38]	
		Aspergillus niger	1–20 nm [39]	
	TiO ₂	Saccharomyces cerevisiae	8–20 nm [32]	
	CdS	Saccharomyces cerevisiae	$3.57 \pm 0.21 \mathrm{nm} [33]$	
		Chlorella vulgaris	15–47 nm. [40]	
		Polysaccharide isolated from <i>Porphyra vietnamensis</i>	$13 \pm 3 \text{ nm}$ [41]	
		Polysaccharide extracted from <i>Pterocladia capillacea</i>	7–18 nm [42]	
	Ag	Polysaccharide extracted from <i>Jania rubens</i>	5–20 nm [42]	
Algae		Polysaccharide extracted from <i>Ulva fasciata</i>	7–24 nm [42]	
		Polysaccharide extracted from <i>Colpomenia sinuosa</i>	15–35 nm [42]	
		Turbinaria conoides	96 nm [43]	
	Au	Sargassum wightii	8–12 nm [44]	
	ZnO	Sargassum muticum	35–57 nm (hexagonal wurtzite structure) [45]	
	Fe ₃ O ₄	Sargassum muticum	$18 \pm 4 \text{ nm} [46]$	
Virus	Au-Co ₃ O ₄	M13 virus	Nanowires [47]	
Plant systems				
	Au	Cassia auriculata	15–25 nm (triangular and spherical shape) [48]	
	Ag, Au	Rosa rugosa	12 nm and 11 nm, respectively [49]	
Leaf extracts	Au, Ag	Aloe vera	50–350 nm and 15.2 \pm 4.2 nm, respectively [50]	
		Vitex negundo L. extract	10–30 nm [51]	
	Ag	Argemone mexicana	30 nm [52]	
		Acalypha indica	20–30 nm [53]	
		Pelargonium graveolens	16–40 nm [54]	
	Pd	Cinnamomum camphora	3.2–6 nm [55]	
Seed extracts	Ag	Jatropha curcas	15–50 nm [56]	
Latex	Ag	Jatropha curcas	10–20 nm [57]	
Bulbs	Ag	Allium sativum	7.3±4.4 nm [58]	
Root extracts	Au	Zingiber officinale	5–20 nm [59]	
noor extracto	Ag	Zingiber officinale	10–20 nm [59]	

TABLE 1: Different methods of green synthesis of nanoparticles, relevant organisms, and special characteristics of synthesized nanoparticles.

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Biological resource	Types of nanoparticle synthesized	Organism/s	Size and special features			
Biological material						
Ascorbic acid and starch	Ag	_	17–30 nm (truncated triangle nanoplates) [60]			
	Au	_	~15 nm [61]			
	Ag	_	18.98–26.05 nm [62]			
Honey	Pd	_	5–40 nm [63]			
	Pt	_	2.2 nm [64]			
	С	_	~7 nm [65]			
Starch	Ag	_	10–34 nm [66]			

TABLE 1. Continued



FIGURE 3: Generalized schematic diagram of honey mediated green synthesis of nanoparticles; proposed mechanism [77].

reducing and a stabilizing agent [71]. Consequently, a 9.9 \pm 3.7 nm particle size was observed by transmission electron microscopy (TEM) that manifested moderate antibacterial activity against both gram positive and gram negative bacterial strains. The lowest minimum inhibitory concentration (MIC) was reported against *Staphylococcus aureus* at 31.25 g/mL [78].

8. Silver Nanoparticles

Bar and colleagues synthesized silver nanoparticles using honey at room temperature, where honey acted as both a reducing and a stabilizing agent [56]. Thus, it replaced toxic and harsh reducing agents such as dimethyl formamide, hydrazine, and sodium borohydride used previously. Synthesized nanoparticles were stable for five months without any TABLE 2: Reducing and stabilizing agents and specific temperatures used in honey mediated biosynthesis of different nanoparticles.

Nanoparticle type	Reducing agent	Stabilization agent	Specific temperature
Gold	(i) Vitamin C (ii) H_2O_2 and gluconic acid, produced when diluting honey with distilled water [61]	Proteins present in honey	RT [61, 78]
Silver	Honey	Proteins in honey [79]	RT [62, 78–80]
Palladium	Honey	Honey	RT [63]
Platinum	Honey	Proteins in honey	100°C [64]

stabilizer. The size and morphology of silver nanoparticles were dependent upon the concentration of honey used and the pH, where particle size decreased with increasing pH. Moreover, scanning electron microscopy (SEM) studies revealed an inverse correlation between size of the silver nanoparticles and the honey concentration; size of the silver nanoparticles was in the range of 18.98–26.05 nm, when 10 g of honey was used, while the size further reduced to 15.63–17.86 nm range with increased honey concentration up to 40 g [62].

A sunlight mediated method to synthesize Ag nanoparticles using honey as both a reducing and a stabilization agent was reported [72]. These honey capped silver nanoparticles were effective corrosion inhibitors for mild steel and remained stable at room temperature for more than six months. Proteins in honey seemed to be the capping agent which stabilized the nanoparticles while fructose acted as the reducing agent [79].

Nearly spherical and monodispersed silver nanoparticles were synthesized with the size of ~4 nm at pH of 8.5 at room temperature [73]. The resultant silver nanoparticles were synthesized in various sizes by adjusting the pH of the solution by using honey as the reducing and stabilization agent. TEM studies and X-ray diffraction (XRD) studies showed a high crystalline and face-centered cubic (fcc) structure. XRD patterns corresponded to (1 1 1), (2 0 0), (2 2 0), (3 1 1),

and (2 2 2) planes. Furthermore, Fourier Transform Infrared Spectroscopy (FT-IR) spectrum indicated that the proteins were bound to silver nanoparticles through the carboxylate group [80]. Furthermore, silver nanoparticles with 11.9 \pm 5.25 nm particle size were synthesized at room temperature using honey as both reducing and stabilizing agents. These nanoparticles demonstrated significant antimicrobial activity against a wide range of bacterial and fungal strains. The lowest minimum inhibitory concentration (MIC) was demonstrated against *Staphylococcus aureus* at 2.81 g/mL [78].

9. Palladium Nanoparticles

Pd nanoparticles with the range of 5 to 40 nm were synthesized using honey as both reducing and stabilizing agents [63]. This nanocatalyst showed higher reusability; Pd nanoparticles were used as a catalyst for Suzuki crosscoupling and in the hydrogenation of conjugated olefins. Thus, honey coated Pd nanoparticles possess potential applications in different fields including nanobiotechnology, organic catalytic transformation, and sensors [63].

10. Carbon Nanoparticles

In the green synthesis of nanomaterial, honey is commonly used as both a stabilizer and a reducing agent. Wu and colleges, however, used honey as a precursor in synthesizing carbon nanoparticles for real time photoacoustic imaging [65]. These surface coated polysorbate and polyethyleneglycol C nanoparticles were comparatively smaller (\sim 7 nm) than the previously used particles such as silica coated gold nanoparticles (20 nm) [81], single wall nanotubes (SWNT), and Cu, used for sentinel lymph node (SLN) imaging. Authors claimed a rapid signal enhancement (\sim 2 min) with these carbon nanoparticles [65].

Two nm high-fluorescent carbon dots were synthesized from honey for sensing and imaging with a quantum yield (QY) of approximately 19.8% [82]. Green synthesized carbon dots exhibited higher stability with other advantages including nontoxicity, high-fluorescent quantum yield, and photostability. Moreover, these carbon dots were used as a sensor for the detection of Fe^{3+} and were applied for fluorescent staining and cell imaging. Authors suggest many potential applications of carbon dots in biosensing and bioimaging [82].

11. Platinum Nanoparticles

Venu and colleges reported a honey mediated, novel, economically feasible method to synthesize platinum nanoparticles and nanowires [64]. They synthesized 2.2 nm sized Pt nanoparticles at 100° C in aqueous honey solution and, furthermore, 5–15 nm length platinum nanowires formed with longer thermal treatment by self-assembly. Characterization of the resultant nanoparticles by spectroscopic, morphological, and structural studies suggests that honey played an important role in the reduction of platinum nanoparticles. Furthermore, honey functionalized platinum nanoparticles were highly crystalline and the structure was a face-centered



FIGURE 4: Schematic diagram of various current applications of honey mediated green synthesis of nanoparticles [63, 64, 78, 79, 82].

cubic. FT-IR analysis suggested that the proteins were bound to platinum nanoparticles through the carboxylate group. These nanoparticles were stable in water for more than four months and showed catalytic properties for the formation of antipyrilquinoneimine dye from 4-aminoantipyrine and aniline in an acidic aqueous medium. Venu et al. posited the application of this catalytic property in the detection and removal of anilines from soil and water samples [64].

Figure 4 illustrates the numerous current applications of honey mediated green synthesis of gold, silver, palladium, carbon, and platinum nanoparticles. Honey mediated green synthesized gold and silver nanoparticles can be used as potent antibacterial agents [78]. Furthermore, silver nanoparticles show antifungal activity and anticorrosion activity [79]. Palladium nanoparticles show catalytic activity and can be used in sensor related applications [63]. Honey synthesized carbon nanoparticles are used in many areas including biosensing, bioimaging, fluorescent staining, and real time photoacoustic imaging [82]. Platinum nanoparticles show catalytic activity [64].

12. Challenges

Honey mediated green synthesis of nanoparticles is a novel concept and yet remains to be fully developed and established. To date, a limited number of studies were conducted, restricted to the synthesis of metallic and carbon nanoparticles. It is essential to synthesize monodispersed nanoparticles to obtain maximum usage. Methods must be optimized to synthesize other complex nanoparticles such as Fe_3O_2 , TiO_2 , ZnO, and CdS. Further studies are warranted to identify the actual ingredients that are responsible for the reduction of metal ions. In some studies, FT-IR spectrum revealed that proteins were responsible for the stabilization. However, further studies are crucial to identify the relevant proteins responsible for the functionalization of these nanoparticles.

13. Conclusion

Most common nanoparticle synthesis methods frequently use harsh chemicals such as hydrazine, dimethylformamide, and sodium borohydride and high temperatures which may pose biological hazards to the environment. On the contrary, honey mediated green synthesis requires relatively low temperatures (generally room temperature) and does not produce any toxic products. Green synthesis methods can be used as an alternative for the currently used chemical and physical synthesis methods. Furthermore, green synthesis of nanoparticles using honey mediated methods demonstrated ecofriendly, cost effective, time saving, and easy processing methods, where honey acted as both reducing and stabilizing agents. Honey functionalized nanoparticles express special activities such as catalytic properties, anticorrosive activities, antimicrobial activity, and biosensing and bioimaging ability. With further improvement, nanoparticles synthesized by honey mediated green methods may provide potential and valuable end products with numerous applications in many fields providing a superior, ecofriendly alternative for harsh and toxic procedures, practiced to date.

Abbreviations

- TEM: Transmission electron microscope
- MIC: Minimum inhibitory concentration
- SEM: Scanning electron microscope
- XRD: X-ray diffraction
- FCC: Face-centered cubic
- FT-IR: Fourier Transform Infrared Spectroscopy
- SLN: Sentinel lymph nodes
- QY: Quantum yield
- UV: Ultraviolet.

Competing Interests

The authors declare that they have no competing interests.

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