

Green Synthesis of Zinc Oxide (ZnO) Nanoparticles: A Review

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Abstract: The agglomeration of nanoparticles (NPs) could be stabilised by the addition of polyvinylpyrrolidone (PVP) and polyethylene glycol (PEG) during synthesis of nanoparticles. However, there are some limitation including producing risky by-products and need extensively treated before discharged to the environment. Therefore the development of a greener approach have received a lot of attention from the scientific researchers. The green synthesis of Zinc oxide (ZnO) nanoparticles using plant extracted are still lacking. This review article summarize the information on the application of green capping agent for the synthesis of ZnO nanoparticles. Some researchers are investigating the synthesis of the nanoparticles using various methods, such as physical, chemical and biological. It entails a high cost of equipment, a high temperature and pressure for the physical procedure and requires enormous space for installing the machine. The chemical methods for synthesizing nanoparticles mostly involve the usage of toxic chemicals which is very harmful to the society and environment.

Keywords: Capping Agents, Nanoparticles, Precipitation Methods, Zinc Oxide

1. Introduction

Nanoparticles define as a particles with a nanoscale dimension containing tiny size particles from 1 to 100 nanometres which able to increase the catalytic reactivity, thermal conductivity and chemical stability such as silver [1], zinc oxide [2], and gold nanoparticles [3]. Zinc Oxide with wurtzite is a semiconductor that has a wide bandgap of 3.3 eV and large excitation binding energy (60 meV), which is larger than the thermal energy at room temperature (meV) [4]. ZnO is used in various applications including as photocatalyst, solar cells, piezoelectric transducers, electroluminescent devices, transparent electrodes, ultraviolet laser diodes, electronic and optoelectronic devices, chemical sensor, laser technology and antibacterial activity [5]. ZnO nanoparticles have well-known as an effective photocatalyst due to several of advantages such as strong excitation binding energy, higher reactivity, surface area, photosensitivity, non-toxic nature and chemical stability [6].

There are several methods that are used in synthesizing nanoparticles such as physical, chemical and biological methods. The physical methods like the ball milling method [7] and Pulse laser ablation [8] are available for synthesizing nanoparticles. The chemical methods also available for the synthesis of metallic nanoparticles such as chemical reduction [9], hydrothermal [10], sonochemical [11], and thermal decomposition [12]. Most of these methods have been employed hazardous reagents during the synthesis of nanoparticles. In view of environmental pollution control and awareness, there is an urgent need to develop an eco-friendly method for the synthesis of nanoparticles.

Recently, the biological method has emerged as an attractive alternative to the traditional methods for synthesizing nanoparticles. Biosynthesis of nanoparticles involves using an environment-friendly chemistry-based such as actinomycetes [13], bacteria [14], fungus [15], plants [16], viruses [17], and yeast [18]. The synthesis of nanoparticles through biological factories provides a safe, non-toxic and environmentally friendly method of synthesizing nanoparticles with a broad range of sizes, shapes, compositions and physicochemical properties [19].

During the synthesizing of nanoparticles, agglomeration might be occurred due to the Van der Waals and Ostwald ripening interactions which leads to the aggregation of the particles. In recent years, the precipitation method has been recognized as the most recommended method as they can provide the simpler route, inexpensive and occur moderately at the most energy-saving condition which is room temperature. Some of the studies that have been carried out by the other researcher are using PVP and PEG as a capping agent in synthesizing nanoparticles but due to hazardous by-product that has been generated and requires extensive treatment before being discharged to the environment, the development of a greener approach is desired to eliminate and minimize environmental problems [20]. In this regard, the synthesis of ZnO nanoparticles from plant extraction as a capping agent is the ultimate solution that approach the significant size of nanoparticles and contributes to their stability. This review article focuses on a summary of information relating to the utilization of green capping agents for synthesising the nanoparticles.

2.0 Body of Literature

2.1 Zinc Oxide nanoparticles

Zinc oxide nanoparticles (ZnO NPs) are environment friendly, offer easy fabrication, nontoxic, bio-safe and biocompatible. Due to these advantages, ZnO NPs have become an ideal candidate for biological applications [21]. Various methods have been explored for the synthesising of ZnO NPs such as the reaction of zinc with alcohol, vapour transport, hydrothermal synthesis and precipitation method [22]. However, these methods suffer from various disadvantages due to the involvement of high temperature, pressure conditions and the use of other chemicals [23]. To reduce the high costs, the usage

of toxic chemicals and harsh conditions for reduction and stabilization of nanoparticles, green synthesis like *Parthenium hysterophorus* [24], *Nephelium lappaceum* [6], and Aloe vera [25] had been explored. A Wide range of the usage of ZnO NPs has been reported including as food packaging [26], application in the cosmetic product (such as sunscreen, lotion and deodorant) [27] and as an ingredient in antibacterial and antifungal creams [28].

2.2 Capping agents

The synthesis of controlled-size metallic and well-defined forms according to the synthesis of metal nanoparticles plays an important role as an influencer in the conformation of the ligand [29]. It had a high potential for better return production and unforeseen selectivity checks. It had a high potential for improved yields and unforeseen selectivity controls as well. Several surfactants and the small ligands, polymers, dendrimers, cyclodextrins and polysaccharides, are used in the various forms of nanoparticles synthesis [30]. Organic ligands changed the metal surface to improve the compatibility with other steps or to incorporate additional functionalities to prevent nanoparticle agglomeration [31].

Capping products in the colloidal synthesis are typically used for avoiding the overgrowth and aggregation of nanoparticles and for precisely regulating the structural features of the resulting nanoparticles. Therefore, capping agents act as a physical obstacle in restricting free access to catalytic nanoparticles by reactants while at the same time promoting the catalytic efficacy of nanocrystals [32]. Referred to Dutta et al., [33] electrostatic force or by using capping agents such as polymers and other macromolecules can prevent the metallic nanoparticle from agglomeration. Therefore capping agent can be used to control the shapes of semiconductor crystals in surface engineering.

2.3 Polymers as capping agents

The synthesis of any product that affects the size, structure, morphology, and optical properties of the product may be significant with capturing agents. Guo et al. [34] declare that the capping agents of highly monodispersed nanoparticles of zinc PVP can act as the capping agents. Many of the properties of the zinc nanoparticles can also be optimized with the molar ratio of Zn (II) and PVP simply changing. Some researchers have tested additional capping agents such as PEG, dodecyl benzene sulfonate (DBS) and ethylene diamine tetra acetic acid (EDTA) [35]. The PVP ZnO NPs are usually well balanced as uncapped nanoparticles concerning morphology and structure. According to Singh et al., [36] the study state that synthesis of ZnO NPs by using different capping agents has shown different size and other ZnO properties.

Guo et al. [35] reported that the size distribution and structure of ZnO NPs can be stabilised using PVP. Where PVP play as capping molecules that affected the nucleation and accretion kinetics. Then the large particle growth slows down while the small particles grow the same during synthesis of ZnO nanoparticles. Therefore, it is essential to modify the surface of the nanoparticles using polymer surface molecules or other materials which can enhance the dispersion stability of nanoparticles in aqueous media or polymer matrices due to a strong repulsion between nanoparticles which is generated by modifiers [37].

2.4 Microorganism as capping agents

Hulkoti and Taranath [38] have been reported some microorganisms in the biosynthesis by intracellular, and extracellular, metallic compounds, such as silver, gold, platinum, zirconium, palladium, iron cadmium and metal oxides. In the transformation of metallic ions into nanoparticles, the microorganisms play an important role. Besides, Nicotinamide adenine dinucleotide hydrogen (NADH) relies on the enzyme nitrate reductase. They claimed, however, that the microorganisms are acceptable capsules and able to give particle stability by preventing agglomeration and growth. Afterward, the microorganism has shown that the biosynthesis of nanoparticles is broad, as its

implementation is healthy and ecosystem friendly. Various species used to use bio-synthesis including bacteria, yeast, actinomycetes, and fungi [39].

The microorganism's cell wall functions as an essential part of the intracellular synthesis of the nanoparticles. The mechanism, therefore, involves electro-static interaction between the positive metal ion charge and the negative cell wall charge. They were used for intracellular synthesis of nanoparticles to explain that the trapping, bio-reduction, and coping mechanisms are involved. The fungal cell surface interacts electrostatically with the metal ions and traps the ions by secreting an enzyme to reduce the metal nanoparticles' metal ions. To help the bioreduction of metal ions and nanoparticle synthesis, fungi secreted the nitrate reductase enzyme [40]. Finally, the ion reduction and silver nanoparticles formation are caused by the reductase enzyme.

2.5 Green Capping agents

Biosynthesis of nanoparticles provides new avenues for synthesizing nanoparticles using plants and microorganisms. This is considered an eco-friendly, simple, cost-effective, biocompatible and safe method. Many researchers have embarked on the subject of synthesising metal nanoparticles from various plant extracts to facilitate the size of the nanoparticles.

Table 1: Plant mediated synthesis of ZnO

Type of plant	Size (nm)	Shape	Common name	Part taken for extraction	Ref
<i>Parthenium hysterophorus</i>	16-45 (TEM)	Spherical, cylindrical	Weed	Leaf	[24]
<i>Nephelium lappaceum</i>	25.67(XRD)	Spherical	Rambutan	Fruit peel	[6]
<i>P. Caerulea</i>	30-40 (TEM)	Spherical	Bluecrown passionflower	Leaf	[41]
<i>Sesbania grandiflora</i>	2–200 (SEM)	Spherical	Hummingbird tree	Leaf	[42]
<i>Trifolium pratense</i>	43.09 (TEM)	Spherical	Red clover	Flower	[43]
<i>Aloe vera</i>	100-900 (SEM)	Rod	-	Leaf	[25]
<i>Plectranthus Amboinicus</i>	18000-618000 (SEM)	Spherical	Cuban oregano	Leaves	[44]
<i>Corymbia citriodora</i>	38.18 (SEM)	Polyhedron	Lemon-scented gum	Leaf	[45]
<i>Punica granatum</i>	20-120 (TEM)	Hexagonal	Pomegranate	Fruit peel	[46]
<i>Euphorbia prolifera</i>	32.98-81.94 (TEM)	Spherical	Pine spurge	Leaf	[47]
<i>Pisonia grandis</i>	5-17 (TEM)	Hexagonal, spherical	Cabbage tree	Leaf	[48]
<i>Albizia lebbeck</i>	50.95 (XRD)	Spherical	Flea tree	Stem bark	[49]

Table 1 indicated various types of the plant used for synthesized ZnO nanoparticles from the previous study. From Table 1, the smallest size for plant-mediated synthesis of ZnO is *Euphorbia prolifera* also known as pine spurge. The size of this plant is 5-17 nm using TEM. When the size is smaller, this causes the total area for the reaction to increase. The shape of *Euphorbia prolifera* is spherical. The part taken for extraction for the Pine spurge is the leaf.

Euphorbia prolifera can act as a reduction and stabilization agent without the use of any harmful reduction or surfactant due to the bio-constituent such as phenolic and flavonoid compound in the extracted leaf. Eco-friendliness and compatibility for biomedical and pharmaceutical applications offer enormous benefits from environmentally benign plant material. The flavonoid and other extract phenolic were not only responsible for the reduced size of nanoparticles but also functioned as capping ligands to the surfaces of the nanoparticles.

Table 2: Different parameters used of synthesis ZnO nanoparticles

Plant	Variables/ parameter	Results	Ref
<i>Catharanthus roseus</i>	pH(8-14) Temperature(30-90 °C) Reaction Time (0.5-4 h) Concentration metal ion (0.0025-0.01 M)	It is suggested at pH 12, T= 30°C _o , conc metal=0.01 M and reaction time =2 h zinc dehydrate convert to ZnO NPs.	[50]
<i>Sesbania grandiflora</i>	Temperature (60-400 °C) Concentration metal ion Reaction time (0.25- 3 h)	UV/VIS: The bulk ZnO appeared at 385 nm. Temperature: 400 °C (for calcination to remove completely water and high crystallinity.	[42]
<i>Trifolium pretense</i>	Reaction time (1 h) Temperature(30 °C-90 °C) Concentration metal ion (0.5 M)	UV/VIS: The absorption edge shifts to the lower wavelength. Size of ZnO NPs is decrease / SEM: use EDS techniques to gain more insight of nanoparticles.	[43]
<i>Punica granatum</i>	Reaction Time (0.25-2 h) Concentration metal ion (0.001 M) Temperature (60 °C- 80 °C)	UV/VIS: Peak at range of 420 nm raised because of oscillation of more electrons after 5 hr./ SEM: Show the presence of carbon that involve in plant phytochemical groups in reduction and capping of the synthesized ZnO NPs	[41]
<i>Nephelium lappaceum</i>	Concentration metal ion (0.1 M) Reaction time(0 h) Temperature(80 °C-450 °C)	SEM: Homogenous particle size of the photocatalyst (essential parameter to improve efficiency in photocatalytic reaction)./ UV/VIS : positioned around 373 nm	[6]
<i>Pisonia grandis</i>	Concentration metal ion(0.1 M) Reaction time(0.1-8 h) Temperature(80 °C-400 °C)	SEM: surface morphology of granules and monodisperse clusters. XRD pattern: crystal planes of Hexagonal phase of ZNO.	[48]

Table 2 indicate the different parameters used in synthesising ZnO NPs using the extracted plant. The plant with the best result is *Punica granatum* due to the properties of the plant has the fastest reaction time which is 0.25-2 h. The concentration metal ion is 0.001 M which is the fastest concentration. Based on the SEM results, it shows the presence of carbon that involve in plant phytochemical groups in the reduction and capping of the synthesized ZnO NPs.

P. granatum F. peel extract was successfully used during the synthesis process to produce ZnO NPs by acting as a reduction and stabilisation agent. *P. granatum F.* Peel extract contains an abundance of

phytochemical compounds that play a major role in reducing and stabilizing the ZnO NPs yield. *Punicalagin* and gallic acid make up about 73 percent of P among all these compounds *Punica granatum* fruit (*P. granatum F.*) or grenade pomegranate was described as a power fruit, well known for its therapeutic excellence [46]. Consumers benefit from the properties and health benefits. *P. F. granatum* Peel accounts for around one-third of the fruit and remains a by-product after consumption. Due to their availability and cost-effectiveness, the use of these fruit agro-wastes has gained considerable attention in recent years. *P. F. granatum* Peel was well-recognized for containing an extraordinarily high number of phenolic compounds as natural antioxidant sources. As reported earlier, some of the major phenolic compounds identified in grenade peels include punicalagine, gallic acid, ellagic acid, chlorogenic acid, caffeic acid, punicaline, apigenine, quercetin, pelargonidine, cyanidine, granatin A, and B. These compounds are concentrated primarily in the *P. granatum* peel portion and have been shown to assist in the green synthesis of a wide range of different NPs [46].

Table 3: Green synthesis metal nanoparticles by using *Cymbopogon*

Type of nanoparticles	Size (nm)	Shape	Type of <i>Cymbopogon</i>	Part of taken for extraction	Ref
Silver nanoparticle	10-33 (TEM)	Spherical	<i>Citratus</i>	Leaf	[51]
Gold nanoparticle	20-50 (TEM)	Spherical, triangular, hexagonal, rod shape	<i>Citratus</i>	Leaf	[3]
Silver nanoparticle	41 (XRD)	Spherical	<i>Citronella</i>	Leaf	[52]
Silver nanoparticle	16.72 (XRD) 10 (TEM)	Spherical	<i>Nardus</i>	Leaf	[53]

Table 3 shows the green synthesis of metal oxide nanoparticles from *Cymbopogon Nardus* and *Citratus* leaf extracted. The green synthesis of silver nanoparticles by using *Cymbopogon Nardus* shows that smaller size (10 nm) might provide larger surface area for the photocatalytic degradation application. The leaves extract was prepared using a direct electrochemical in situ method that operated under a mild condition. The average particle size, from diffraction patterns was calculated to be 16 and 29 nm for calcinated Ag / extract and uncalcinated Ag / extract, respectively. TEM image Proven that nanoparticles with less agglomeration are well dispersed with Ag / extract calcinates. The photocatalytic performance of the green synthesized Ag under visible light showed its increased activity against the organic dye methylene blue [53]. It was also observed that the calcination process has resulted in the highest percentage degradation of Blue methylene. This metal preparation method, directly in situ, is believed to contribute to the new path in controlling organic pollutants.

3. Conclusion

Nature has elegant and ingenious ways of creating the most efficient miniaturized functional materials. Increasing awareness towards green chemistry and the use of the green route for synthesis of metal nanoparticles lead to a desire to develop environment-friendly techniques. Synthesis of zinc oxide nanoparticles by using a green capping agent which is *Cymbopogon Nardus* extracted via precipitation method has been studied by other researchers for years. Green capping agents such as plants can act as both stabilizing and reducing agents for the synthesis of shape and size-controlled nanoparticles. In synthesis of zinc oxide nanoparticles, it has been indicated as a good green synthesis of metallic nanoparticles by using plants because of the practical way of experimental, eco-friendly, protect the environment and lead to lesser waste.

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