A Review on Green Synthesized ZnO Nanoparticles for Sustainable Energy Harvesting

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Abstract

The green synthesis of ZnO material, especially in nano formulations such as nanoparticles (NPs), quantum dots, and nano-rods are widely used in research and industry. ZnO is a favourable material for sustainable energy harvesting among metal oxide materials. Out of these formulations, ZnO NPs are the most demanded material of modern day's research due to their multiple applications in different fields of technical, medical and environmental fields. In this study, ZnO NPs are considered as an area of interest due to their special optical, electrical, optoelectronics, photocatalytic, antibacterial, electrochemical and semiconductor properties. So green synthesised ZnO NPs are used in making supercapacitors, bioimaging, biosensing, antimicrobial, anticancer and wastewater treatment due to their low toxicity and radical scavenging property. It is observed as a direct band gap semiconductor. The green synthesis method of preparing ZnO NPs is an easy, eco-friendly, non-toxic, simple and cost-effective method compared to the other chemical methods. The ZnO NPs produced from green synthesis routes are ecologically sustainable products used in the biomedical range, having anticancer, antibacterial, antioxidant and wound healing properties. So these are the favourable products of the biomedical field and can serve as a solution to environmental pollution created by synthetic

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Publisher: Anu Books
Book Name : Chemical Sciences at the Nexus of Sustainability: Bridging Disciplines

dyes, chemicals, paints and textiles. The size range of ZnO NPs can vary from 10 nm-86 nm depending on the type of plant extract and synthesis method used at different temperature conditions and concentration of dopant used. The main advantage of the green synthesis method is the huge availability of different types of plants and their parts having phytochemicals like polyphenols, ketones, flavonoids, and aldehydes in plants. These phytochemicals act as reducing agents as well as capping agents. This review highlights the latest developments in ZnO research, concentrating on green synthesis approaches, property control, and progressive applications in sustainable technologies, both medical, technical and environmental.

Keywords: *ZnO NPs, Green synthesis, Antibacterial, Sustainable energy, Biomedical*

1. Introduction

The metal oxide materials have great interest in recent research due to their multiple applications in different fields of technical and medical, out of which, ZnO formulations such as NPs, quantum dots, and nano-rods are widely used in research and industry. In this study, ZnO NPs are considered as an area of interest due to their special optical, electrical, optoelectronics, photocatalytic, antibacterial, electrochemical supercapacitors and semiconductor properties. The ZnO NPs exhibit a wide band gap (3.37 eV) and a large exciton binding energy (60 meV).

In most cases, the ZnO NPs are prepared by various techniques such as chemical precipitation, sol-gel, hydrothermal, and combustion methods. These methods require large experimental time, a tedious process, expensive instrumental setups, toxic chemicals, high temperatures, and specific protocols. It is observed as a direct band gap semiconductor [1]. On the other hand, the green synthesis method of preparing ZnO NPs is an easy, eco-friendly, non-toxic, simple and cost-effective method compared to the above methods. The ZnO NPs produced from green synthesis routes are ecologically sustainable products used in the biomedical range, having anticancer, antibacterial, antioxidant, and wound healing properties. So these are the favourable products of the biomedical field [2].

The main advantage of this method is the huge availability of different types of plants and their parts having phytochemicals like phenols, ketones, flavonoids, and aldehydes in plants. These phytochemicals act as reducing agents as well as capping agents. This review highlights the latest developments in ZnO research, concentrating on green synthesis approaches, property control, and progressive applications in sustainable technologies, both medical and technical [3].

2. Synthesis Methods for ZnO

ZnO QDs/NPs are synthesized using various chemical and physical methods that allow precise control over particle size, morphology, and surface characteristics:

2.1 Chemical precipitation method

In this method, 0.1 mol Zn $(NO_3)2\cdot6H_2O$ solution prepared in 100ml distilled water was stirred at 60°C for one hour. 100 ml of 0.2 mol NaOH solution was added dropwise into the solution of Zn $(NO_3)_2\cdot6H_2O$ under constant stirring, until the pH value of 8.5 was obtained for the solution. The solution slowly turned white and was further stirred continuously at 60 °C for one hour. It was then kept overnight for stabilization. The white precipitate so obtained was washed several times with distilled water and then dried at 90 °C for five hours to get a fine powder. The obtained powder was divided into six parts- one part was kept unannealed, while the other parts were subjected to two-hour annealing at different temperatures in a furnace. Finally, the prepared samples were used for further characterization to study the effect of annealing temperatures on their structural and optical properties [4].

2.2. Hydrothermal method

The hydrothermal method is an effective method to produce ZnO NPs as used by Hossein Safardoust-Hojaghan et al. [5]. In this method, 0.3 g Zn $(CH_3COO)_2.2H_2O$ was added to 30 cc deionized water under stirring until the formation of a transparent solution, and 0.1 g SDS (Sodium dodecyl sulphate) was added to the solution and stirred vigorously for 30 min. Ammonia, diethanolamine, and ethylenediamine were added for pH adjustment. The obtained solution was moved to a Teflon-lined stainless autoclave and heated at 180 °C for 5 h. The final whitish product was centrifuged and washed several times with deionized water and ethanol. Finally, the product was dried at 60 °C overnight.

2.3. Sol–gel synthesis

The sol-gel method is the typical method to produce ZnO QDs as used by Pushpendra Singh et al. [6]. Zinc acetate (ZnAc) is used as a precursor material in this method, and water-ethanol (2:1) is the solvent used to prepare the zinc acetate solution. The LiOH (lithium hydroxide), NaOH (sodium hydroxide), and KOH (potassium hydroxide) are also dissolved in ethanol and used as a precipitating agent. A white precipitate is formed after dissolving these two solutions, which is washed with ethanol and collected by centrifugation. If the reaction time and temperature increased, the particle size increased. These white precipitates are again suspended in ethanol or water as a dispersing medium, and colloidal ZnO QDs are formed.

3. Green Synthesis Methods for ZnO NPs

3.1. Ultrasonication and magnetic stirring method

This method is the green synthesis method to form ZnO NPs using Punica granatum peels, as used by Komal Bhandari et al. [7]. In this method, Zinc nitrate $[Zn (NO_3)_2.6H_2O]$ is used as a source/ precursor of Zn^{++}, NaOH and ethanol with pomegranate peel powder. Punica granatum (pomegranate) fruit contains high concentrations of phenolic compounds, such as punicalagin, Gallic, and the presence of phenolic compounds makes the pomegranate fruit peels a potential reducing and stabilizing agent for the synthesis of ZnO NPs.

The ultrasonication and magnetic stirring produce NPs with a more uniform size distribution and larger crystalline size, potentially leading to improved material properties. The zinc ions interact with the polyphenols in the plant extract to form a complex, which undergoes hydrolysis to form zinc hydroxide ($Zn(OH)_2$), eventually leading to the synthesis of ZnO NPs.

3.2. Precipitation method for ZnO NPs

In this method, ZnO NPs are produced with the help of plant extract Tilia Tomentosa (Ihlamur), a yellow color solution made in deionised water, which acts as both a reducing agent as well as capping agent. This plant extract is added to a solution of Zinc nitrate hexahydrate [$Zn(NO_3)_2.6H_2O$] in double-distilled water by constant stirring. Then NaOH was added dropwise until the yellow-colored solution turned into a pale yellow-colored precipitate. These precipitates were centrifuged and washed with water, and again centrifuged, resulting in the pale yellow/white ZnO NPs. The dried ZnO NPs were calcined in a furnace at 400 °C with a holding time of 15 min to remove any evaporable impurities. The calcined samples were cooled to room temperature and stored for further characterization. This method is used by R. Shashanka et al. [3].

3.3. Biosynthesis Approach

In this method, a sustainable and environmentally friendly process known as green synthesis of ZnO NPs are formed by using microorganisms and plant extracts prepared from plant parts like roots, leaves, stems, seeds, and fruits have also been utilized for the NPs synthesis as their extract is rich in phytochemicals that act as both reducing and stabilizing agents [8, 9]. The presence of alkaloids, amino acids, enzymes, proteins, and polysaccharides in plant extracts contributes to their role as reducing agents and capping agents, simplifying the synthesis process and enhancing its safety, sustainability, and environmental friendliness compared to conventional physical and chemical methods [10].

The ZnO synthesis procedure starts by placing 2 g of zinc nitrate in the extracts of orange peel (1, 2 and 4% by weight of peels of C. sinensis), creating three different samples (M1, M2, and M3, respectively). The samples were stirred for 1 h and then placed in a water bath at 60 °C until each mixture presented a thick consistency due to water evaporation. The samples were then heat-treated at 400 °C for 1 h, then pulverized in a mortar, obtaining a white to cream or beige coloured powder [11].

Table 1: Summary of green synthesis of ZnO NPs through various plants parts.

Botanical source	Plant part used	Synthesis method used	Particle size (nm)	Shape of NPs	Applications	Ref.
Tilia Tomentosa (Ihlamur)	Plant leaves	Precipitation method	22	Hexagonal	Dye-sensitized solar cells (DSSC)	[3]
Punica granatum (Pomegranate)	Fruit peels	Ultra sonication magnetic stirring	57-72 65-81	Spherical	Engineering, pharmaceutical medical	[7]
C. sinensis	Orange peel	green chemistry method	12.7-24.3	Hexagonal wurtzite	Photo catalytic applications	[11-14]
Deverra tortuosa	Aerial parts plant	Precipitation method	9.26 - 31.18	Hexagonal wurtzite	Antimicrobial and anticancer agent	[15]
Aloe barbadensis miller	Aloe vera leafs	chemical and biological method	25-45	Spherical	Biomedical & cosmetics	[16]
Mangifera indica	Mango seeds	chemical and biological method	40-70	Cylindrical	Antibacterial & antioxidant activity	[17]
Ruellia tuberosa	Leaves	chemical and biological method	40-50	Rod shape	Antibacterial agent and photocatalytic degradation of MB dye & water pollutants.	[18]
Ziziphus mauritiana Lam	LeavesMg/ Cu doped	green chemistry method	85.1-86.6	Hexagonal disk-like shaped	Antibacterial and radical scavenging activities	[19]

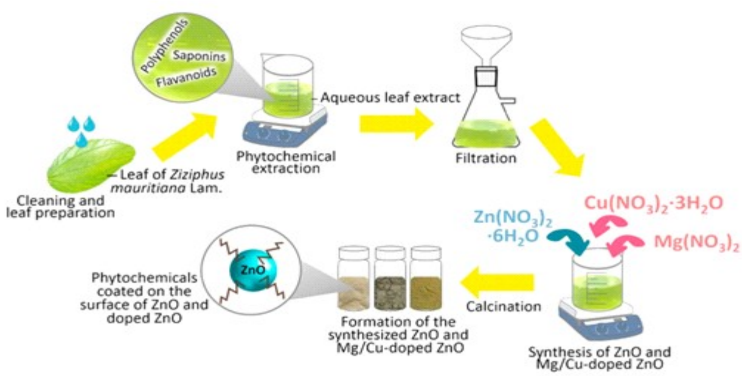

Figure1: Synthesis method of ZnO and Mg/Cu-dual doped ZnO using green approach [19]

4. Characterization and Properties of ZnO NPs

4.1 Crystal structure

ZnO structure is hexagonal Wurtzite structure with space group $P6_3mc$ in which Zn^{2+} and O^{2-} are both tetrahedrally coordinated. The typical lattice parameters: a ~3.25Åc~5.20Åc/a~1.60Å

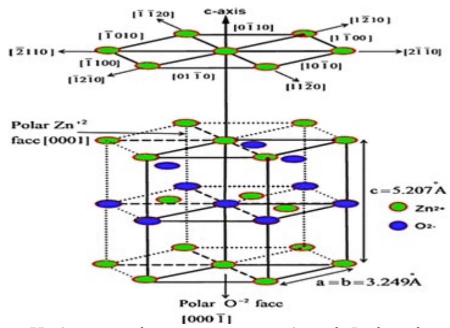

Figure 2: ZnO unit cell (wurtzite structure) with ionic positions of Zn^{2+} and O^{2-} ions, with various crystal planes of ZnO wurtzite structure (Figure2 adapted from research gate) [12]

Table 2: The basic physical parameters of ZnO structure [13]

Physical Parameters	Values
Stablephaseat300K	Wurtzite
LatticeConstants	a=b=0.32495nmandc=0.52069nm
MeltingPoint	1975 ^{0}C
Density	5.66g/cm^3
RefractiveIndex	2.01
BandGap	3.4eV,Direct
Electroneffectivemass	0.24
Holeeffectivemass	0.59
Excitonbindingenergy	60meV
Static dielectric constant	8.656

4.2. Optical properties

ZnO has tightly bound 2p electrons of oxygen and 3d electrons of Zn, and it has Zn interstitials and oxygen vacancies. It has a well-known visible luminescence is mainly due to defects that are related to deep-level emissions such as Zn interstitials and oxygen vacancies [14]. The UV–Vis spectra of biosynthesized zinc oxide NPs using Solanum torvum showed absorption peak maxima at 359 nm, and this is a characteristic signature for ZnO NPs [20], **[Figure 3].**

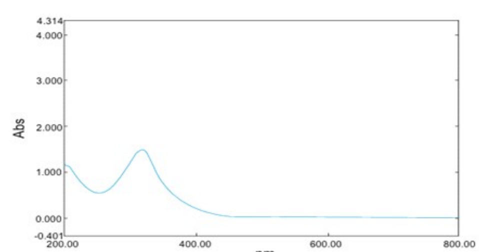

Figure 3. UV–Vis spectroscopic analysis of synthesized zinc oxide NPs, a single peak corresponding to zinc oxide NPs (359 nm)[20].

4.3. Photocatalytic properties

The Photocatalytic degradation is a process that begins with the generation of the electron–hole pair in the ZnO samples when they are irradiated with ultraviolet light, as a smaller band gap allows a faster electron–hole pair generation. The electron and hole interact and react in different areas of the catalyst surface with the O_2 and the OH^- within the MB (Methylene Blue) solution, forming hydroxyl and hydroperoxide radicals, and superoxide radical anions known as reactive oxygen species (ROS) [11], **[Figure 4].**

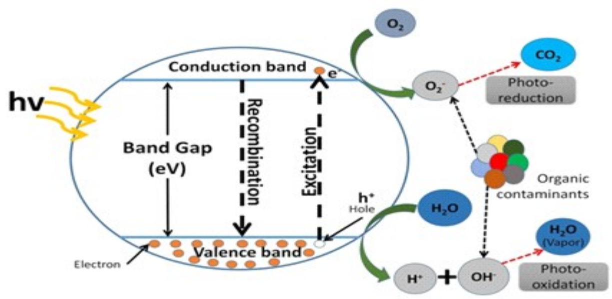

Figure 4: Photocatalysis mechanism [11]

Further SEM images (as shown in **figure 5**) of green synthesised samples confirms rod like and small flower form of ZnO NPs, which make them permeable and good adsorbents for synthetic organic dye molecules like IV2R (Ismate Violet 2R). So ZnO NPs could be a promising material for wastewater treatment for removing organic dyes from water [21].

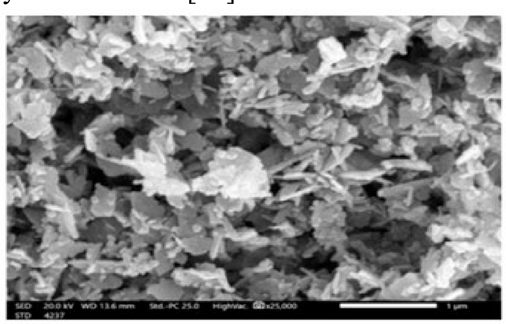

Figure 5. SEM image of green Synthesised NPs [21]

4.4. Antibacterial properties

The given material ZnO produced by green synthesis methods is found to have antibacterial capabilities. ZnO nanostructures have been more harmful to bacteria and less reactive towards human cells [22]. This happens due to the reason that ZnO NPs has an excellent antibacterial mechanism based on the production of reactive oxygen species (ROS) that kill bacterial cells [23].These ROS, such as free radicals, have high penetration power in small organism cells and destroy their molecular structure making these materials (ZnO NP's) as resistance against microorganism infections and treating cancer cells [3], **[Figure 6].**

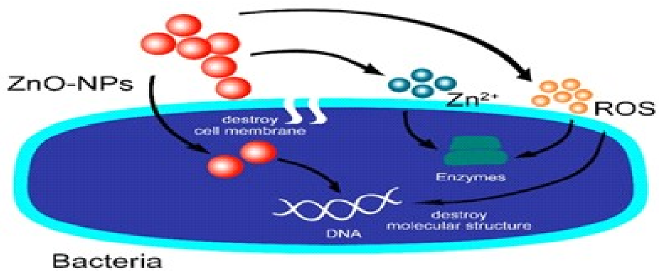

Figure 6. Antibacterial mechanism of ZnO NPs. (ROS formation, Zn^{2+} release, internalized ZnO NPs, and electrostatic interactions) [24].

4.5. Electrochemical properties

The electrochemical properties of ZnO NPs analysed by Cyclic voltammetry (CV) analysis, which shows that these materials have higher charge storage properties and a fast charge transfer process, which confirm by the higher area under the CV curve. The quasi-rectangular shape of the CV curve of the sample (as shown in **Figure 7**) confirms these materials as ideal capacitance [25,35].

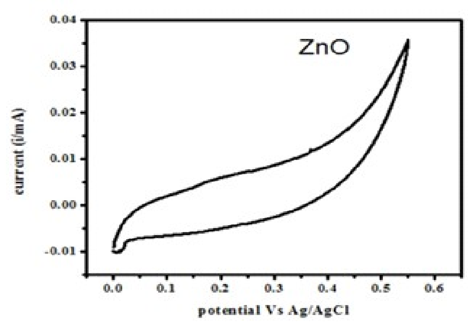

Figure 7: Cyclic Voltammetry analysis of ZnO NPs [25].

4.6. XRD/SEM Analysis

The Hexagonal wurtzite structure is confirmed by the XRD analysis such that (1 0 1) planes peak intensity is observed in Carica papaya mediated ZnO NPs, also with Sn and Mn Co-doped ZnO (as shown in **Figure 8**) and granule like spherical ZnO NPs are formed with size range 20-30 nm (as shown in SEM micrograph Figure 9 (a-c) and Histogram 9(d-f))[26].

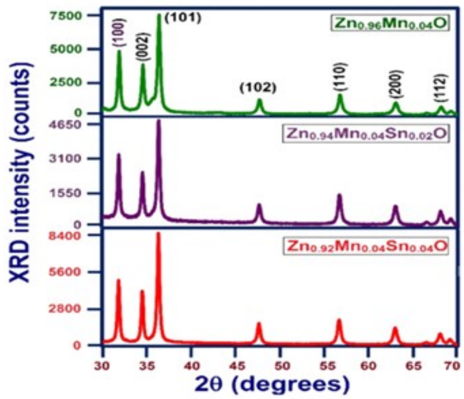

Figure 8: XRD analysis of Carica Papaya mediated ZnO-NPs. [26]

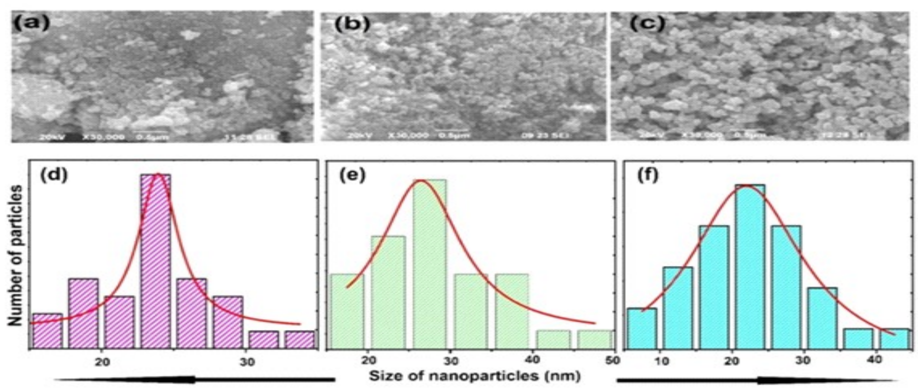

Figure 9. (a-c) SEM micrograph and (d-f) Histogram of C. papaya mediated ZnO NPs [26]

5. Applications

The doped ZnO has many applications, such as UV photodetectors and sensors (gas and pH sensing), viz., the Fe-doped ZnO nano rods prepared by CBD (Chemical bath deposition method) with photolithography procedure serve as a UV photodetector as mentioned by Yen-Lin Chu et al. [27]. These materials show the good detection for the presence of gases like O_2 and NO_2 [28,29]. Further, ZnO and doped ZnO can be used in the medical field in a limited way against cancer, such as 2%Fe-doped ZnO NPs can reduce tumor growth in animal cells. These materials also show antibacterial properties and can be used against bacterial infection, for both gram-positive and gram-negative bacteria [30]. The green synthesis prepared ZnO materials are the best suitable material for supercapacitor applications, as confirmed by the CV (Cyclic voltammetry) curve, which indicated an excellent capacitance behaviour, low equivalent series resistance (ESR) and hence fast diffusion of electrolyte ions into the composite [31]. The ZnO NPs exhibited efficient photocatalytic activity in degrading MB (methylene blue) and MO (methyl orange) dyes under UV light exposure. It shows the degradation of organic dyes MB and MO by using ZnO NPs as a catalyst. The Synthesized ZnO NPs also exhibited high antioxidant activity against DPPH (1,1-Diphenyl-2-picrylhydrazyl) molecule compared to ascorbic acid as a standard reference material [32]. The green synthesized ZnO NPs proved to be an effective antibacterial agent and photocatalyst, which can be efficiently utilized for the manufacturing of antibacterial coated cotton fabrics and the degradation of harmful toxic dye pollutants persisting in the environment [33]. The ZnO NPs synthesized using the extracts of H. officinalis L. have anti-inflammatory activities, so they can be used as an anti-cancer and anti-inflammatory drug candidate [34]. The carbon-coated ZnO NPs (ZnO/C) prepared from green synthesis sol-gel route using sucrose, which acts as a carbon source and capping agent, is a sustainable method for environment safety point of view. The electrochemical study reveals that ZnO/C behaves as an aqueous symmetric supercapacitor with two electrodes, and it shows a maximum specific capacitance of 92 F/g at a specific current of 2.5 A/g, having an energy density of 32.78 W h /Kg in cyclic voltammetry analysis [35].

Table 3: Summary of energy harvesting from ZnO NPs in different technical fields.

Botanical source	Synthesis method used	Characterisation	Particle size (nm)	Shape of NP's	Applications	Ref.
Sucrose ($C_{12}H_{22}O_{11}$)	Sol-gel method	XRD, FTIR, SEM, CV	27	Hexagonal	Supercapacitor electrode	[35]
Citrus Reticulata (Mandarin peels)	green chemistry method	XRD, UV-Vis,TEM, Wave guide simulation	22.6	Wurtzite	Optoelectronic Sensors as cladding material for waveguide	[36]
Dandelion leaves	Green synthesis	XRD, FTIR, SEM, UV-Vis	65-100	Spherical	PVA thin film in solar cell	[37]
Rhododendron aerboreum flowers	Co-precipitation method	XRD, EDM,XPS,FESEM,HRTEM	4.5-15.4	Hexagonal wurtzite	Electrochemical biosensors, Green photocatalyst	[38]

6. Conclusion

From the above discussion, it is concluded that green synthesis of ZnO NP's through various routes is eco-friendly, commercially low-cost production, which are used in various medical applications. The toxicity in these ZnO NPs is extremely low due to the plant extract made up from different parts of plant. The diverse range of plant extracts, including phytochemicals like flavonoids, terpenoids, polyphenols, tannins, and alkaloids, is a promising resource for the environmentally friendly production of ZnO NPs. The ZnO and its doped samples have technical applications such as UV photodetectors and sensors (gas and pH sensing) and symmetric supercapacitors.

The presence of phytochemicals that were responsible for surface stabilization and capping in the synthesis and the formation of ZnO was confirmed by FT-IR analysis. Furthermore, it was found that both antibacterial and radical scavenging activities of the S–ZnO and Mg/Cu-doped were better under irradiation of visible light. Although these materials have Antibacterial, anticancer and radical scavenging applications and toxicity is reduced in ZnO NP's by green synthesis methods. According to Kenneth Maduabuchi Ezealisiji et al. [20], these materials still show some toxic effect to kidney and liver cells and could cause damage to the organs following a long period of application. So extra care should be taken for prolonged application to human tissues.

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