

A Review on Zinc Nanoparticles: Synthesis and Application

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Abstract:

Green synthesis of nanoparticles gained huge attention due to ecofriendly protocol to reduce toxic effects of chemical synthesis mostly used in the industry. Among a variety of nanoparticles (ZnONPs) have advantages because of its relevant physio-chemical properties. It is one of the cheap material in cosmetic industry, nanofertilizers, an excellent component for detecting ecological pollutants, a semiconductor photocatalyst for dye decolorization and environmental remediation. Ecotoxicity studies on ZnONPs and ZnO based composites are most abundant in bacteria than other species tested. It suggests the generation of Reactive oxygen species (ROS) and photo-induced toxicity related with photocatalytic property of ZnONPs may be another mechanism of toxicity. For this reasons, this review aims to carefully document the role of Plant-mediated green synthesized, photocatalyst ZnONPs and ZnO based composites in dye decolorization and its ecotoxicity towards an aquatic vertebrate like fish under safety assessments. We also look at how photocatalytic dye decolorization is affected by ZnO based composites. Finally, current knowledge gaps within this areas are briefly discussed.

Keywords: ZnONPs, ZnO based composites, ROS, photo-induced toxicity.

I. INTRODUCTION

One of the most important environmental challenges, particularly emerging countries, has been identified as irregular industrial development. The textile, plastic, and paper sectors all produce large amount of wastewater containing synthetic dyes (Mansour et al.). Several of those (artificial dyes) were found to be toxic to living organisms (Azharuddin et al.). This growing trend raises crucial concerns about ecological balance and human health implications. Particularly, due to advancement of science and technology, the uses and applications of nanoparticles are increasing in our daily lives and on industrial scale (Aliko; Chukwuka ; Vajargah). Green nano technology is a methodology for creating nanoparticles that is devoid of or employs fewer toxic substances throughout generation process Metal nanoparticles have several applications in environmental pollution treatment, including heavy metals, degradation of toxic dyes, and antimicrobial substances (Mansour et al.). Zinc nanoparticles (ZnONPs) have a significant surge in usage across various applications including industrial catalyst, gas sensors, biomedicines, and environmental remediation (Alprol et al.). Chemically, in ZnONPs the Zinc atom has an atomic number of 30 and belong to group 12, while the Oxygen atom has an atomic number of 8 and belong to group 16 of periodic table (Bedi et al.). ZnO Exhibits three crystalline structures namely, wurtzite, zinc blende and an occasionally noticed rock salt (Ozgun; Moezzi). The hexagonal wurtzite structure posses lattic spacing $a = 0.325\text{nm}$ and $c = 0.521\text{nm}$, the ratio $c/a \sim 1.6333$. Each tetrahedral Zn atom is surrounded by four Oxygen atoms and vice versa (George et al.). The structure is thermodynamically stable in an ambient environment (Moezzi et al.). This tetrahedral symmentry is responsible for polar property of the Wurtzite crystal of ZnO (Coleman and Jagadish). These crystal structure are illustrated in (Fig 1). ZnO has a slew of unique chemical and physical properties, viz., high chemical stability, high electrochemical coupling coefficient, broad range of radiation absorption and high photostability, which make it among all metal oxides a key technological material and confer upon its wide applications in

varied fields (Raha and Ahmaruzzaman). (Table 1) shows some of the physiochemical properties of ZnONPs.

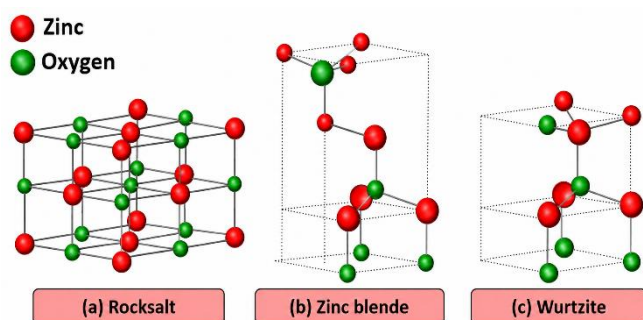


Figure 1 : Structure of Rocksalt, Zinc blende, Wurtzite.

Property	Value
Appearance	White Solid
Odour	Odourless
Molecular Weight	81.38 g/mol
Density	5.6 g/cm ³
Coordination geometry	Tetrahedral
Crystal Structure	Hexagonal wurtzite
Melting Point	1975 °C(decomposes)
Flash Point	1436 °C
Toxicity Hazards	None

Refractive Index	2.0041
Bandgap	Direct and 3.37 eV at RT
Electrical Conductivity	Semiconductor
Hole Effectivity Mass	0.59
Electron Effective Mass	0.24
Exciton Binding Energy	60 meV
Bohr Radius	2.34 nm
Luminescence	Luminescent in UV and visible light

Table 1 Property and their Values

Chemical method of synthesis is advantageous as it takes short period of time for synthesis of large quantity of nanoparticles. However, in this capping agents are required for size stabilization of the nanoparticles. Moreover, chemical reagents used generally for NPs synthesis and stabilization are toxic and lead to by products that are not environment benign (Vithiya and Sen). Green synthesis of ZnONPs using plant is a viable synthesis method that has the potential to replace chemical and physical synthesis methods as it offers a cheaper and environment friendly option. Generally, plant mediated ZnONPs exhibited better photoactivity against organic dyes and antibacterial activity than chemical mediated ZnONPs (Mutukwa et al.).

Photodegradation of dyes using semiconductor metal oxides such as ZnONPs offers an alternative remedy that is consistent, secure, and capable of degrading dyes even at low concentrations (Tanwar et al.).

Merits	Demerits
Environment friendly	Relatively less stable NPs
Cost effective	Reproductivity less stable issues
Less toxic	Difficult to control morphology
Low energy consumption	
No expensive equipment requirement	

Table 2 The Merits and Demerits of Green synthesis of ZnO-NPs.

In light of these, this review focuses on the green synthesis of ZnONPs using plant materials and ZnO based nanocomposites synthesis their role in photocatalytic degradation of dyes and Ecotoxicological effects on an aquatic vertebrate fish.

2.Plant mediated synthesis of ZnONPs:

Plants are abundant and easily accessible natural resources that are "chemical factories" and have a vast majority of applications. For instance, they are employed in environmental remediation to eliminate heavy metals and other pollutants from the environment through processes like detoxification, accumulation, or phytoremediation (Jadoun et al.). Plant parts such

as leaves, stems, flowers, fruits produce large quantities of biomolecules via various metabolic pathways that can be used to fabricate NPs (Mohamad et al.). The synthesis can be divided into three steps : Plant collection and treatment, biomolecule extraction and synthesis. The process begins with the collection of fresh plant material which is followed by wash of the plant material to remove dirt and debris. The washed plant material is dried or used in its fresh form. After drying, the plant material is ground to powder to increase the interaction with the solvent during at low temperatures (RT to 80°C) using conventional extraction methods such as maceration, decoction, and Soxhlet extraction. The qualitative analysis of the extract yielded saponins, polyphenols, tannins, flavonoids, terpenoids, carbohydrates and proteins. After obtaining a plant extract, it can be mixed with Zn salt precursor and heated at low temperatures usually from RT to 100°C), which is one of the appeals of plant mediated synthesis (Mutukwa et al.). A summary of plant mediated synthesis of ZnONPs is shown in (Figure 2).

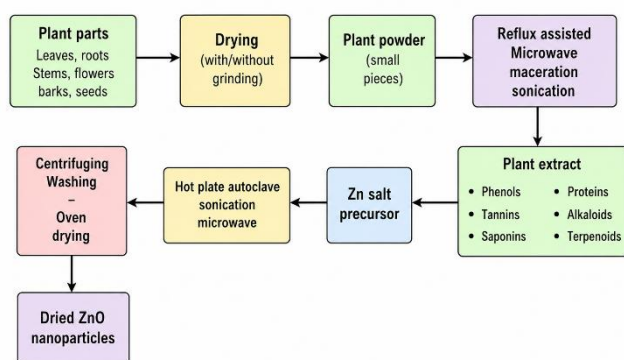


Figure 2. Summary of Plant Mediated synthesis ZnONPs.

However it should be noted that the mechanism of formation of ZnONPs using plant extracts is not fully understood due to presence of a large variety of biomolecules in the plant extract. As more than one type of biomolecule may be involved in the reduction and stabilization of the

ZnONPs. Hence, research efforts should also focus on to investigate mechanism of formation of ZnONPs using plant extracts

3.ZnONPs as a Photocatalyst

The release of untreated or inadequately treated industrial wastewater into water bodies is detrimental to the environment, aquatic life, and human health because dyes are highly carcinogenic, mutagenic, toxic, and highly resistant to biodegradation. The removal of dyes from industrial wastewater is challenging since organic dyes are resistant to heat, light, oxidation and biodegradation (Mutukwa et al.). The increased public concern with these environmental pollutants has prompted the need to develop novel treatment methods with photocatalysis gaining a lot of attention in field of pollutant degradation (Zeltner et al.). The photocatalytic detoxification of wastewater is the process that combines heterogeneous catalysis with solar technologies (Zhang et al.). Photocatalysis has garnered more interest, and this is owing to its nontoxic nature, relative cost effectiveness, and non-energy intensiveness since operations can take place in ambient conditions. Moreover, there are no secondary pollution concerns because the technique is capable of decomposing organic dyes into harmless compounds (Pirhashemi ; Zhu). ZnONPs have been utilized extensively for the photocatalytic degradation of dyes due to their properties such as having low cost, high photostability, high chemical stability, high photoactivity in the UV region and environmental friendliness (Lanjwani et al.).

When ZnONPs are irradiated with energy greater or equal to their bandgap energy, the free electrons in the valence band (VB) absorb the energy and become excited to the free conduction band (CB). This leaves holes in the VB, and the electrons and holes (Charge carriers) migrate to the surface of the ZnONPs. At the surface, the holes react with the water and hydroxyl ion to form hydroxyl ($\cdot\text{OH}$) radicals, whereas the electrons react with adsorbed Oxygen to produce

superoxide anion radicals, which will result in the formation of Hydrogen peroxide. The hydrogen peroxide (H_2O_2) reacts with superoxide radicals to produce more ($\cdot\text{OH}$) radicals. The hydroxyl radicals are potent ROS and react with the adsorbed organic products: Carbon dioxide (CO_2) and Water (H_2O). (Figure 3) shows a schematic diagram of the photocatalytic degradation mechanism of dyes using ZnONPs (Fagier ; Ong).

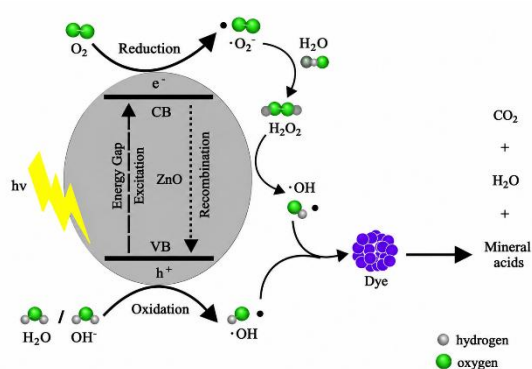


Figure 3. Schematic diagram of the photocatalytic degradation mechanism of dyes using ZnONPs.

The particle size of nano-photocatalysts is one of the major factors that influence the photocatalytic activity of NPs. When the particle size of a nano-photocatalyst is small, there is a high surface area to volume ratio, which means increased active sites for the adsorption of surface oxygen and dye molecules, thus leading to better photoactivity (Gangwar et al.). When the band gap energy is too narrow, recombination of created charge carrier occurs which reduces the generation of the hydroxyl radicals, resulting in decreased photodegradation (Kebede et al.). The particle size and bandgap energy of nano-photocatalysts plays an essential in photocatalytic degradation of dyes. The Shape of Nano-photocatalyst also influence the

process of photocatalytic degradation of dyes. Waseem et al. reported that degradation efficiency was higher with a smaller bandgap compared to larger bandgap. The authors studied, *Citrus jambhiri lushi* mediated synthesis of ZnONPs and their role in photocatalytic degradation of MB dye under UV irradiation for 120 min (Waseem et al.).

Saif et al. studied *Withania coagulans* fruit mediated synthesis and chemical mediated synthesis of ZnONPs against MB dye under sunlight irradiation for 120 min. The authors reported that, the dye degradation efficiency was higher (90%) due to smaller crystalline size (25nm) and reduced bandgap (3.34eV) of *W. coagulans* fruit mediated ZnONPs compared to chemical mediated ZnONPs (78%) having larger crystalline size (30nm) and increased bandgap (3.44eV) respectively (Saif et al. 9). (Wijesinghe et al.) studied different morphologies of ZnONPs synthesis via *Heliotropium indicum* against MB dye. The authors concluded that, the spherical ZnONPs due to its higher surface area show highest degradation efficiency than flower-shaped ZnONPs. All this studies concluded that, Lower particle size, Lower bandgap energy and morphologies of nano-catalyst are major factors that influence the photocatalytic degradation efficiency of dyes. In addition to this properties other factors like initial pH, catalyst dosage and initial dye concentration also influence efficiency. Therefore, some research efforts can be taken on exploring this factors in photoactivity of nanocatalysts.

4. ZnO based composites and dye degradation

When ZnONPs combined with other materials such as polymers, metals, metal oxides, and carbon based nanostructures lead to the formation of ZnO based composites exhibits improved stability and enhanced functionally suitable for advanced technological applications. A Semiconductor ZnO with unique exciton binding energy (60meV) make it suitable for nanocomposite formation. (Sadasivuni ; Kafy ; Al-Gaashani ; Parangusan). ZnO based

composites has wide applications including wastewater treatment, air purification, and degradation of Organic pollutants. They play significant role in energy conversion hence used in supercapacitors, and solar cells to enhance energy efficiency and durability (Purushothaman ; Chen). The support material which have been used in photocatalytic degradation of pollutants (Ramadhani and Helmiyati) investigated that the hydrogel - ZnO composites shows higher degradation efficiency (90.12%) against CR dye in 110 min under visible light irradiation. In this studies, the authors embedded the NPs in a CMC-alginate hydrogel that was crosslinked using Ba (II) ions. (Bel Hadjltaief et al.) synthesize ZnO-TNC composite in which authors immobilized ZnONPs on Tunisian natural clay (TNC) used against MG and CR dyes under solar and UV irradiation. The authors reported that, ZnO-TNC composite shows higher degradation efficiencies under solar than UV irradiation for both dyes. Thus, ZnO based composites can increase photocatalytic activity, improve their photostability but more studies are required to explore this avenue. There is very less literature about plant mediated ZnO based composites. Therefore, there is a need to investigate the plant mediated ZnO based composites on supports.

5. Ecotoxicity of ZnONPs in fish

A photocatalyst ZnONPs promotes generation of ROS during photocatalytic degradation of dye under irradiation with energy at or above its band gap energy (3.37 eV) and this shows to produce significant toxicity in higher organisms (Ma et al., “Toxicity of Manufactured Zinc Oxide Nanoparticles”). (Brunner; Heinlaan) reported that, In aquatic environment ZnONPs partially dissolve in water release Zn metal ions. The solubilized Zn ions from ZnONPs has proven to cause cytotoxicity in aquatic flora and fauna. (Brunner et al.) investigated that the toxic action of metal and metal oxide NPs involve three distinct mechanisms First, release of toxic substances into media e.g. free Ag ion from Ag nanoparticles. Second the surface

interactions of this metal ions with media produce toxic products, e.g. chemical radicals or ROS. Third, the toxic substances or surface interactions directly targets and disrupt biological targets e.g. intercalation in DNA (DNA mutation). (Hao and Chen) studied, toxicity of ZnONPs to carp (*Cyprinus carpio*) the authors reported that ZnONPs at up to 50 mg/l was not fatal to the fish, but causes increase in lipid peroxidation, oxidative stress due to decrease in activity of several antioxidative enzymes. The oxidative stress found to be more with organs like gill, liver, brain than intestine. (Johnston et al.) reported that the limited uptake of the NPs (up to 5mg/l) may lead to particle aggregation reduces their bioavailability in Zebrafish tissues. (Deepa et al.) studied the treatment of ZnONPs in the testis of *Cyprinus carpio* and observed, slow rate of spermatogenesis, defective lumen, decreased expression of several transcription factors and few steroidogenic enzyme genes. ZnONPs inhibited oocyte ovulation and follicular development in female Zebrafish (Mawed et al.).

Thus, ZnONPs exhibits negative impacts within aquatic ecosystems influencing physiology and biochemistry of fish. An important aspect is to investigate, photo-induced toxicity of ZnONPs to aquatic ecological species under environmentally relevant UV irradiation. Thus more studies are required on photo-induced toxicity study of ZnONPs. (Figure 4) shows a toxic effects of ZnONPs on fish physiology and biochemistry (Mandal et al.).

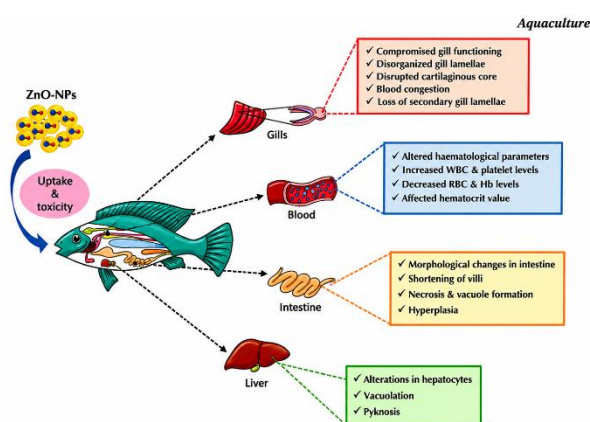


Figure 4. Toxic effects of ZnONPs on fish physiology and biochemistry.

6. Conclusions and recommendations for future research

The literature reviewed herein suggests several conclusions and recommendations for future research on plant mediated ZnONPs synthesis, ZnO based composites, and their role in dye degradation and ecotoxicity against an aquatic vertebrate like fish. Green synthesis of ZnONPs using plant extract is a cheaper, ecofriendly, cost effective and less toxic approach as compared to physical and chemical synthesis of ZnONPs leads to toxic by products. Biomolecules present in plant materials can stabilize ZnONPs but exactly which biomolecules are involved in the stabilization process need to be investigated. The green synthesized NPs are less stable and shows reproducibility issues as compared to chemical synthesized NPs. Furthermore, research efforts are required to understand stability, reproducibility of plant mediated ZnONPs. The photocatalytic degradation efficiency of dyes by using plant mediated ZnONPs mainly depends upon three major factors : Particle size (Smaller particle size shows more photoactivity), Bandgap energy (lesser band gap energy shows more photoactivity), and morphology of nanocatalyst. Other factors such as pH, initial dye concentration and catalyst dosage also influence the photoactivity. Thus, more studies are required to understand this parameters. Moreover, ZnO based composites plays an important role in photocatalytic degradation of dyes and shows higher degradation efficiencies. There are very less reports in the review about plant mediated ZnO based nanocomposites synthesis and their role in photoactivity. Therefore, there is need to be investigated. Ecotoxicological studies of ZnONPs shows harmful effects on physiology and biochemistry of fish due to generation of ROS. While, photo-induced toxicity is another level of toxicity which associated with both terrestrial and aquatic environment. There is no sufficient data related to long term exposure of fish to ZnONPs and photoinduced toxicity of ZnO based composites on fish. Based on this reasons, there are several knowledge gaps need to be filled to understand photoinduced toxicity in ecotoxicological studies.

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