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Eco-Friendly Synthesis of Zinc Oxide: Green Chemistry Approaches and Innovations

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Abstract

Techniques of green production of zinc oxide nanoparticles, also called ZnO-NPs utilizing extracts of plants have drawn study interest in sustainable innovation due to its low cost, ease of use, and benefits for the natural world. This review presents a sustainable method for synthesizing zinc oxide nanoparticles, also known as ZnO-NP from plant extracts, and the features of zinc oxide nanoparticles are examined. These physical features attest to zinc oxide form, structure, crystal formation, and proportions. The dimensions form and dimensions significantly influence the zinc oxide nanoparticle's interface reactivity. A further benefit of capping compounds as stabilizing agents that limit nanoparticle growth, and avert the accumulation and disintegration in colloid synthesizing, is presented in the article review. The capping ligands maintain the connection between nanoparticles and their formation medium. Ultimately, zinc oxide has become one of the most favored options for the use of sunlight because of its spectral properties. The mechanism and its application in different fields have been summarized.

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1. Introduction

At the moment, a wide range of industries, including the graphic arts business, rubber-based products, writing materials, leather products, materials for plastics, and fabrics, use dyes made from organic materials. These companies always produce effluent that contains colorants, which are hazardous to the surroundings and seafood. Therefore, the removal of these organic dyes from wastewater before they enter the environment is substantial [1]. For a long time, most physicochemical treatment processes were used in wastewater treatment technology. These traditional technologies are classified into three major categories: a) chemical, b) physical, and c) biological treatment processes [2]. Inevitably, because of the intricate chemical makeup of the pollutants, many organic dyes are not soluble in conventional methods of elimination. Accordingly, new modern wastewater treatment processes need to be flexible, cost-effective and efficient for marketing purposes [3]. The development of nanotechnology processes is now one of the most active study areas in modern materials through the synthesis of different types of nanoparticles i.e. organic and inorganic materials that attracted great interest due to their diverse beneficial properties and applications in various fields. The importance of nanomaterials is ascribed to tunable physical and chemical properties, good biocompatibility, bioavailability and biodegradability [4]. Although physical and chemical methods are more common in nanoparticle synthesis, however, biogenic production is a better option for environmental reasons [5]. Green synthesis often uses secure, chemical-free, and renewable materials. The fact that no extra substances are required and the method being tested is easy enough to conserve energy also lowers the total cost of the synthesis process. As such green technology improves environmental safety and drives developments in the technological and scientific fields of electronics, cosmetics, coatings, and biotechnology. Different bio-sources have been used in the manufacturing of nanoparticles such as plant parts, algae, fungi, bacteria, and viruses. Plants are broadly dispersed and simply available, and they are sources of various metabolites [6] Plant extracts may generate safe and non-toxic nanoparticles, making them a new supply [7]. During the last two decades, a lot of research has focused on the synthesis of different metal oxide nanoparticles as promising materials in different fields of applications. Examples of these nanoparticles include cerium oxide NPs (CeO_2 NPs), iron oxide NPs (Fe_2O_3 NPs), silver oxide NPs (AgO NPs), magnesium oxide NPs (MgO NPs), titanium oxide NPs (TiO_2 NPs), nickel oxide NPs (NiO NPs), zirconium oxide NPs (ZrO NPs), cadmium oxide NPs (CdO NPs) and zinc oxide NPs (ZnO NPs). The specific properties of zinc oxide nanoparticles (NPs) might differ based on their dimensions, forms, shape, and ratio of sides. Nanoparticles are a harmless, biodegradable biological material [8]. It should be highlighted that although the nanoparticles can be synthesized safely however, different toxic effects can occur depending on particle size and type of individual particles and/or mixtures[9] In this review we explore the green synthesis of ZnO NP using various types of plant extract applied as capping and reduced agent. We discuss the specification of ZnO NP, its mechanism and photocatalytic Activity. By providing a comprehensive overview of the state-of-the-art in green ZnO synthesis, this article aims to foster the development of sustainable nanotechnology and promote environmentally responsible practices in nanoparticle production.

2. Zinc oxide

ZnO is the chemical description for zinc oxide, an inorganic material. Water hardly dissolves this white powder at all. Glass,cement-based materials, and ceramics are just a few of the products and components that frequently use zinc oxide powder as an addition. Zinc oxide nanoparticles are the second most abundant metal oxide after iron. They are cheap, safe, and easy to manufacture [10]. Wurtzite is the normal crystalline form of zinc oxide, a mineral which is usually found during the mineralogical phase known as zincite in an environment [11]. Zinc oxide is a suitable host for doping because of its electromagnetic characteristics, which include a wide bandwidth(3.37 eV)and an ambient temperature activation with an electrical binding energy of 60 meV [12]. At room temperature (about 25 degrees Celsius), unprocessed zinc oxide displays varying magnetised reactions, such as electromagnetic qualities, depending on the chemical reaction technique used [13]. The diverse features of ZnO, which are increased by producing this material at the nanoscale level, make it extremely important for multiple sectors. However, as worries about the effects on our surroundings have grown, eco-friendly manufacturing techniques have been developed. A growing body of research reports on the interest in utilizing natural processes to produce metal and metal oxide nanomaterials. The term "green synthesis route" was used for this process. Inspired by the idea of safety-by-design, a number of straightforward, affordable, repeatable, and sustainable environmentally friendly production methods for nanoparticles have been developed. Figure 1 illustrates the benefits of using the environmentally friendly synthesis process for nanoparticles made of zinc oxide. This has led to the widespread use of several natural systems, including yeast, bacteria, fungi, and extracts from plants, in sustainable synthesis methods to create zinc oxide nanoparticles [14].

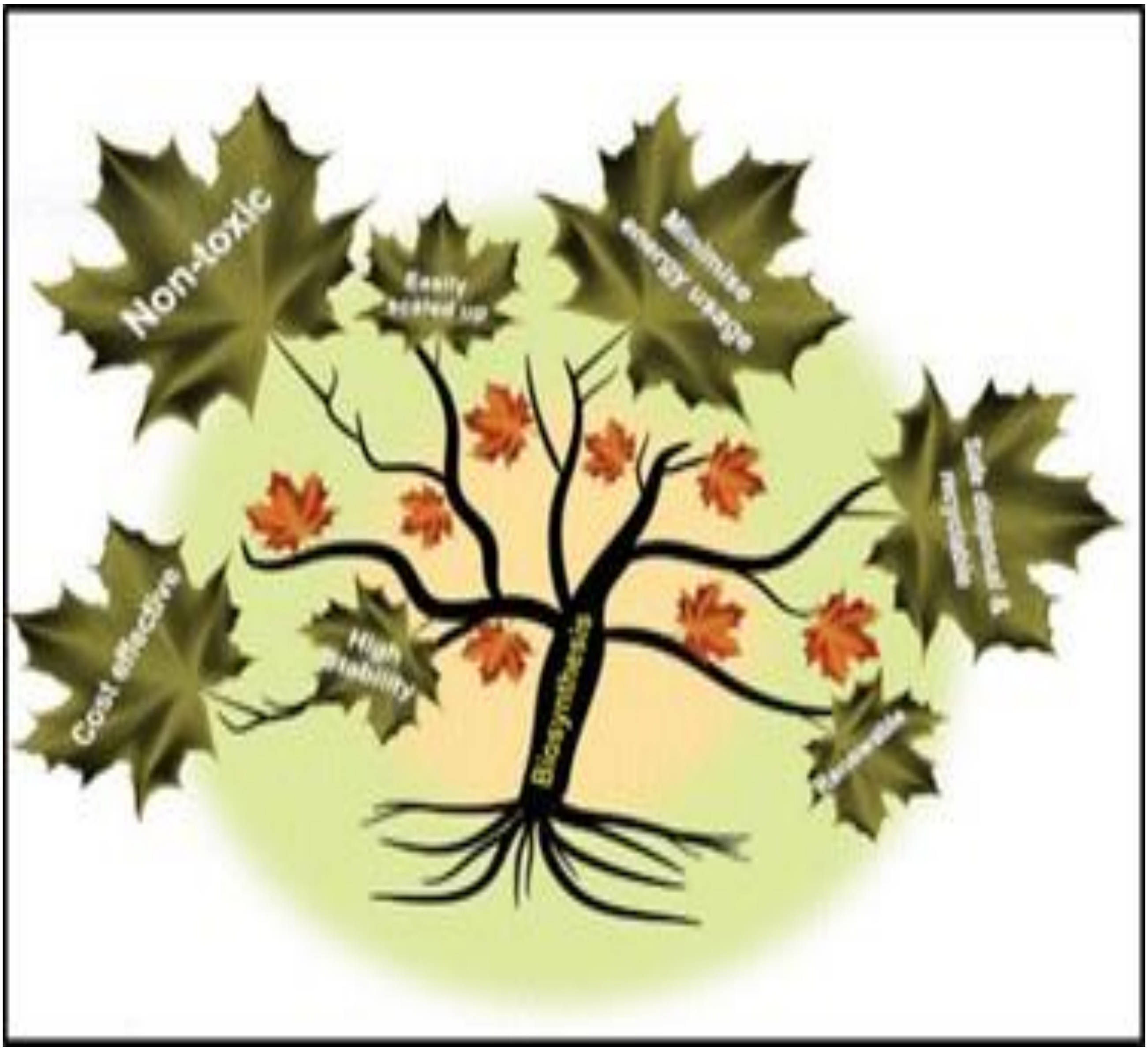

Figure 1. Advantages of biosynthesis using plants or plant extracts as resources for ZnO nanomaterials production [14].

Figure 2 illustrates the standard sustainable route technique for ZnO nanoparticle production using plant extracts that are naturally obtained. The components of the plant were cleaned in the first step by being washed with water that had been distilled. This step's goal is to make sure every single epiphytes and related particle is eliminated. The fresh, damp plant material is then dried before being chopped, ground, or utilized straight to make the extract from it. According to published research, cutting or crushing the plant portion into tiny pieces can improve extraction process efficiency without destroying the compounds found in the plant [15]. Table 1 presents the effect of Zinc source and plant extract on the production of nanoparticle size of ZnO.

Table 1.The effect of Zinc source and plant extract on the nanoparticle size

ZnO Precursor	Extract	NP size	Applications	Ref.
Zinc nitrate	Ailanthus altissima	5-18 nm	Antibacterial	[16]
Zinc nitrate	C.sinesis	Was influenced by water amount.	Photocatalytic	[17]
Zinc nitrate hexahydrate	Tomato peels	9.7 nm	Photocatalytic reaction	[18]
Zinc acetate	Pomegranate	28.4 nm	Photocatalytic	[19]
Zinc nitrate hexahydrate	Orange peel	10-20 nm	Antibacterial	[20]
Zinc nitrate hexahydrate	pomegranate	18-30 nm	Antibacterial	[21]
Zinc acetate hexahydrate	Orange peel	8 nm	Photocatalytic	[22]
Zinc nitrate	Orange peel	7.106 nm	Biological	[23]
Zinc nitrate	Cherry extract	20.18 nm	Biological	[24]

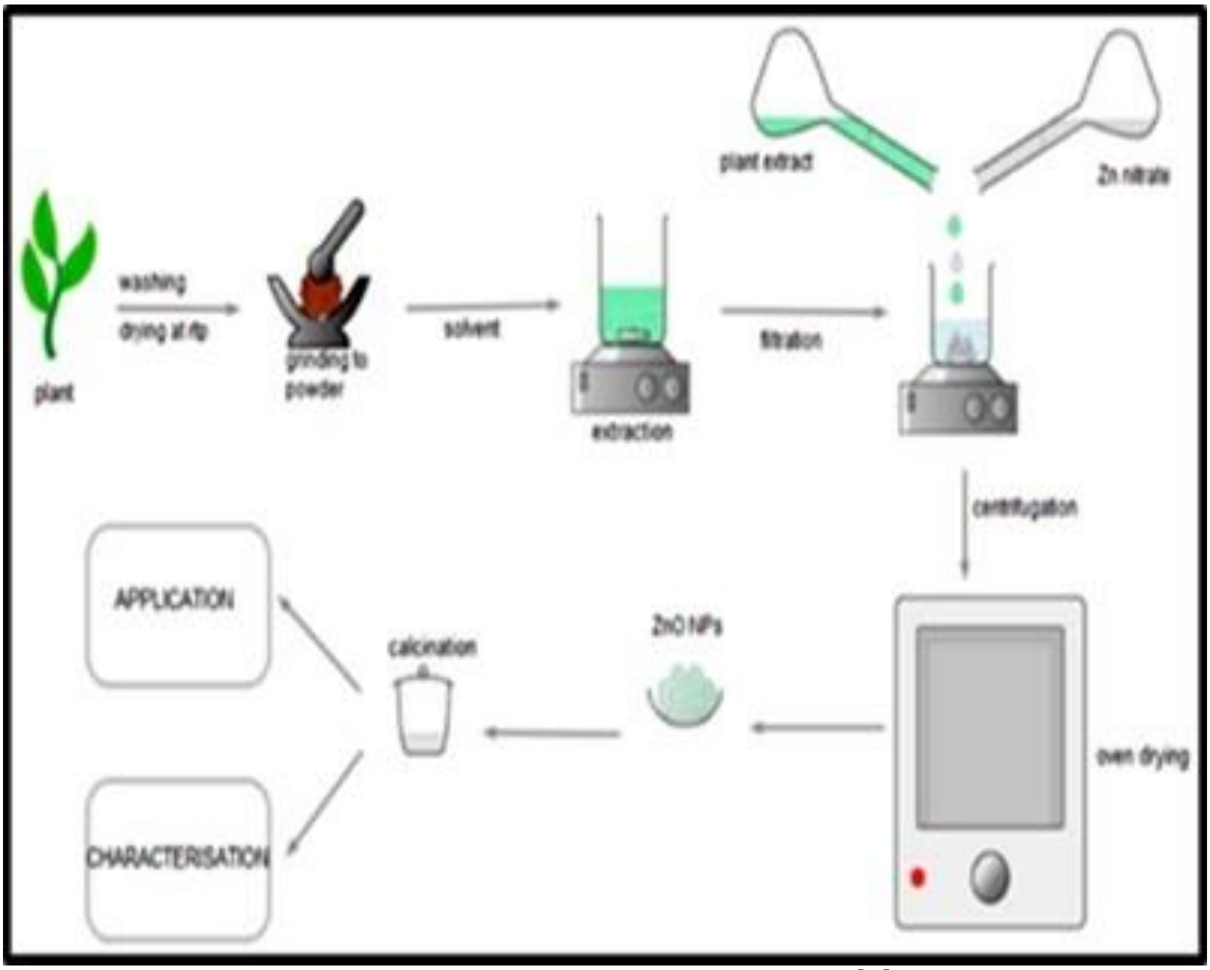

Figure 2. Green rout synthesis of ZnO-NPs [25]

3. Capping agent

Generally, the effectiveness of nanoparticles has been widely investigated for various applications such as medicine, pharmaceuticals, cosmetics, electronics, etc. However, the physiochemical properties of these nanoparticles will extremely affect their interaction with cells causing an increase in potential hazards. As such there was the need to develop safer nanoparticles depending [24]. Zinc oxide nanoparticles accumulate over time during the production process, and the resulting particles may be environmentally damaging. Therefore, the stabilization of zinc nanoparticles is an important challenge that can occur through the use of external capping agents or surface modifiers. Certain requirements should be considered while choosing a capping agent, including non-toxicity, biodegradability, well-dispersibility, biosolvability, and biocompatibility. Furthermore, the high selective capping agent reduces the mass transfer and limits particle growth. Generally, the two main factors that affect mass transfer between the solution and the particles are (1) adsorption/desorption in the material and the extract; (2) bonding between the capping agent and the surface of the solid particle [26]. For example, sodium dodecyl sulfate (SDS), is a common synthetic capping agent that can reduce the toxicity of nanoparticles [27]. They are often used in colloidal dispersions to control nanoparticles, providing precise control over their growth, aggregation, and physicochemical properties. Because its molecules are amphiphilic without polarity hydrocarbon tail and a head that is polar group, they can regulate the formation of nanoparticles. Because of its amphiphilic nature—in which the polar head interacts with the metal atoms in the nanosystem while the non-polar tail interacts with the surrounding medium—the capping agent's functioning and compatibility with another phase are improved [28]. The usage of green covering substances, such as citric acid, fermentation enzymes, polyphenols, polymers that degrade, and extracts from plants, has increased noticeably, notably in the synthesis of nanoparticles, as the development of green technology and sustainability has grown in relevance. Water is the most often utilized solvent for extraction while making green ligands. Vegetation, vegetables, roots, leaves, and their peels that are fragrant can now be extracted to create bio capping using aqueous extracts of plants. The quantity and quality of the biochemical composition can be influenced by the operating parameters of the extraction process. The advantages of using these extracts were to reduce environmental pollution, successfully replace toxic chemicals, and be cost-effective [29]. Additionally, because the extract's structure contains living molecules, it may function as a reducing agent for the highly regulated assembly of molecules needed to create metallic nanoparticles. Furthermore, metabolites can function as capping agents and reducing agents. Examples of these include flavonoids, sapogenins, steroids, and carbohydrates. Table 2 and Table 3 describe various types of synthetic and green capping agents produced for the production of ZnO Nps.

. 4. Characterization of ZnO

Characterizing nanoparticles can be done in a number of ways. These nanoparticles were described in terms of their dimensions, form, accumulation, adsorbed possibility, zeta potential, size of particles, area of surface, porosity, solubility, and cyclic complexity [30]. Numerous morphologies of zinc oxide such as particles, strings of ribbons, electrical lines, tubes, plates, and flowers, have been produced and documented in published works[31]. Each result was involved special characteristics for the production of ZnO depending on the type of extract, preparation conditions and the source of Zno metal.[32] reported the successful synthesis of ZnO and C-doped ZnO from Cystoseira Clinite aqueous extract calcined at different calcination temperatures. Biomolecules contained in the plant extract and detected by FTIR play an important role in covering and stabilizing the newly formed nanoparticles. At the same time, the XRD results show sharp and intense peaks, indicating good crystallinity of the catalyst. An increase in crystal size due to carbon doping was observed. Moreover, an increase in crystal size was observed with increasing temperature. The spherical shape with agglomerations for ZnO was confirmed by SEM images. The tight interparticle space that results from the production of nanoparticles in water-based extracts and the high surface energy of zinc oxide nanoparticles may be the causes of these clusters. The successful carbon doping into the ZnO structure could reduce the energy gap from 3.09 EV for ZnO to 2.93 EV for 5% C-doped ZnO [33,34] supported the x-ray diffraction analysis and scanning electron microscope analysis's findings that zinc oxide hexagonal wurtzite structure had formed. According to calculations, ZnO nanorods have a band gap of 3.4 eV. The UV-vis spectra of the produced zinc oxide exhibit an absorption band edge in the ultraviolet spectrum at 373 nanometers [35].On the other hand, the effect of extract and metal concentration was stated by [36] for the synthesis of ZnO using Juglans regia L. leaf extract. The results of FE-SEM image for ZnO NPs confirmed the formation of spherical with aggregation range from 45 nm to 65 nm in size at zinc acetate concentration of 20 mM. However, at low concentrations i.e. 2 mM, a flower shape of ZnO was formed with size ranging from 95-150 nm. The author concluded that this difference in shape and size of ZnO could be refers to sequential core production ZnO NPS [37,23] reported a comparison study between the ZnO prepared chemically and ZnO prepared by the use of Vaccinium arctostaphylos leaf extract assisted ultrasonic radiation. According to the results, the ZnO prepared by the use of extract confirmed the formation spherical shape meanwhile a spindle-like shape was detected by the SEM and TEM analysis for the ZnO prepared chemically without the use of extract. The author noticed that the extract's bioactive components, which function as a capping agent to regulate the growth of the crystallites, caused the production of tiny, spherical particles [38].

5. Applications of ZnO NPs

ZnO has a wide variety of chemical and physical properties. It can be used in many different sectors. Zinc oxide is important in many applications such as medicine to agriculture, paints to chemicals, and tyres to ceramics morphology and size.

Table 2. Various categories of synthetic capping agents affect the properties of the produced metals including

Type of capping	Capping agent	Morphology of ZnO	Size of ZnO NP (nm)	Reference
surfactants	Ethylene Diamine SDS Sodium dodecyl sulfonate SDS	Nanorods Plate-like & Nanorods Ultra-long nanowires Needle-like & Flower-like	110 ± 40 22–26 8 200	[39] [40] [41] [42]
Small Organic molecules	Histidine L-Lysine Sodium succinate hexahydrate Urea	Nanocrystals Hexagonal rod & Cubic-like Cluster whiskers & rod-like whiskers Hollow nanorods assembled into microflowers	4–7 400–650 100–800 10–50	[43] [44] [45] [46]
Polymers	PVP Poly acrylonitrile Triton X 100	Nano-rods Pineal-type, Flower-type & Sea-urchin-type Nanorods	30 NA NA	[47] [48] [49]
polysaccharide	Chitosan Xanthan Gum Honey	Nanorods Microstars Spherical	60–70 358 39	[50] [51] [52]

Table 3, Green capping agent utilized for the synthesis of ZnO nanoparticles

Plants Name	Plants parts	Shape/morphology	Size (nm)	Applications	Ref
Zizyphus jujube	Fruit	Spherical	29 nm	Photocatalytic activity.	[53]
Cydonia oblonga	Seeds	Flower	25 nm	Photocatalytic properties.	[54]
Musa acuminate	Peel	Triangular	30–80 nm	Photocatalytic properties.	[55]
Aloe Vera	Leaf	Spherical	63 nm	Antimicrobial and ¨Photocatalytic properties.	[56]
Banana	Peel	hexagonal wurtzite shape	128 nm	Photocatalytic activities.	[57]
Cassia Auriculata	leaves	Rod	20–30 nm	Antimicrobial properties.	[58]
Aloe Vera	Level	Flaky and rod	18 µm	Antimicrobial activities.	[59]
Ocimum Gratissimum	Leaf	Spherical	14 nm	Antimicrobial activity.	[60]

6. Mechanism of ZnO NPs as photocatalyst

Groundwater and the environment are rich in non-biodegradable dyes found in organic waste and wastewater from paints, textiles and many other industries. Researchers have developed various methods that are eco-friendly, cheap, non-toxic, simple and biodegradable. The photocatalytic activity is one of the most effective and interesting treatment methods where its catalytic activity is based on Shape, surface area, size and optical activity of the produced catalyst. Zinc oxide nanostructures fabricated using biological/eco-friendly approaches show better photocatalytic activity among various other materials [61]. Nanotechnology-reducing substances can be synthesized from phytonutrients and a wide range of biologically active compounds with several functional groups. These phytochemicals reduce metal ions to nanoforms through reduction mechanisms. The stability, size and shape controllability, and anti-aggregation of metal oxide nanoparticles are all highly influenced by phytochemicals. In the environmentally friendly synthesis of micron-sized particles, it is generally acknowledged that phytonutrients serve as capping and reduction agents. These compounds inhibit species exchange in the synthesis environment and preserve the firmness of the micron-sized particle interface. Stabilizers can affect the biological properties of nanoparticles. They can change the surface properties of nanoparticles to better communicate with target cells for therapeutic purposes. In this situation, the stabilizer must be non-toxic and biocompatible so that it can be taken up by the organism [62]. The mechanism of the Photocatalytic decolorization of dyes (See Fig.3) is thought to occur through the following steps: Electrons are moved from the band known as the valence band to the conductivity spectrum of the zinc oxide catalyst when it is subjected to ultraviolet light, producing pairs of electrons and holes

(Equation-1).Catalysts (ZnO) + hv → e_{cb} + h_{vb} --------------(1)

Where, in the electron band, h^+_{vb} represents the electron vacuum while, in the conduction band, e_{cb} denotes the number of electrons. These two entities have the ability to move towards the outermost layer of the catalyst, where they can engage in a redox mechanism with other surface-dwelling species.Equations 2 and 3 show that most of the time, h^+_{vb} will interact readily with surface-bound H_2O to form OH• radicals, but e $_{cb}$ can react with O2 to generate superoxide radical anion of oxygen.

$h^+ + H_2O \rightarrow H^+ + OH^\cdot$ ----- (2)
$e_{cb} + O_2 \rightarrow O_2^{-\cdot}$ ------ (3)

The holes and electrons that were created in the initial step cannot recombine thanks to this process. Equations 4 through 7 indicate that $OH^\cdot$ and $O_2^{-\cdot}$ can then react with the dye to generate other species, which is what causes the dye to become less colored.

$H_2O + O_2^{-\cdot} \rightarrow H_2O_2$ (4)
$H_2O_2 \rightarrow 2\ OH^\cdot$ (5)

$OH^{\cdot} + dye \rightarrow dye_{ox}$ (6) $Dye + e_{cb} \rightarrow dye_{red}$ (7)

Table 4 . Uses of zinc oxide nanoparticles produced from extracts of plants

Plants name	Plants part	Salt/precursor	Applications	Activities	Ref
Sweet cherry	Fruit	Zinc (acetate dihydrate)	Photocatalytic,	It demonstrates the excellent photocatalytic performance of 98 percent for the catalyst breakdown of methylene blue in sixty minutes when exposed to sunshine.	[63]
Indian Bay leaf	Leaf	Zinc dinitrate	Antibacterial properties.	They show possible action against *aspergillus niger*, *candida albicans*, *staphylococcus aureus*, and *escherichia coli*.	[64]
Spiral ginger	Leaf	Zinc (acetate dihydrate)	Antidiabetic, antibiofilm, and antioxidant properties.	Mammalian red blood cells with minimal hemolytic properties and zinc oxide nanoparticles demonstrated biological compatibility.	[65]
Japanese Barnyard Millet	Grain	Zinc (acetate dihydrate)	Cytotoxicity	When tested against the growth of escherichia coli AB 1157, zinc oxide particles showed a good cytotoxic effect.	[66]
Sodom apple	Leaf	Zinc dinitrate	Cytotoxicity	In the MDAMB-231 cell, biosynthesized zinc oxide nanoparticles may change the expression of apoptotic proteins and cause apoptosis.	[67]
Milk thistle	Seeds	Zinc nitrate hexahydrate	Applied to agriculture	The green zinc oxide-based Silybum marianum nanoparticles showed great promise for replacing the more dangerous pesticides.	[68]
cow dung extract	Leaf	Zinc nitrate hexahydrate	Application in agriculture	The initial stage of using zinc oxide nanoparticles derived from cow dung to help with seed priming and soil zinc deficiencies in a sustainable manner. Compared to commercial ZnO, the generated zinc oxide has lower toxicity.	[69]
Wild jack	Fruit	Zinc nitrate hexahydrate	Photocatalytic activity and antioxidant properties.	When exposed to ultraviolet and sunlight, the nanoparticles show potential photocatalytic activity towards the breakdown of the dye methylene blue (MB). It was shown that zinc oxide nanoparticles had significant antioxidant activity against free radicals known as 2,2-diphenyl-1-picrylhydrazyl.	[70]
H. sabdariffa	leaves	zinc acetate	anti-diabetic properties.	It was found that mice treated with STZ had significantly higher levels of glucose in their blood, which were thereafter restored by zinc oxide nanoparticle treatment. PZN60 and PZN100-treated mice had lower blood glucose levels than untreated mice by 59.5 and 48.27%, respectively.	[71]

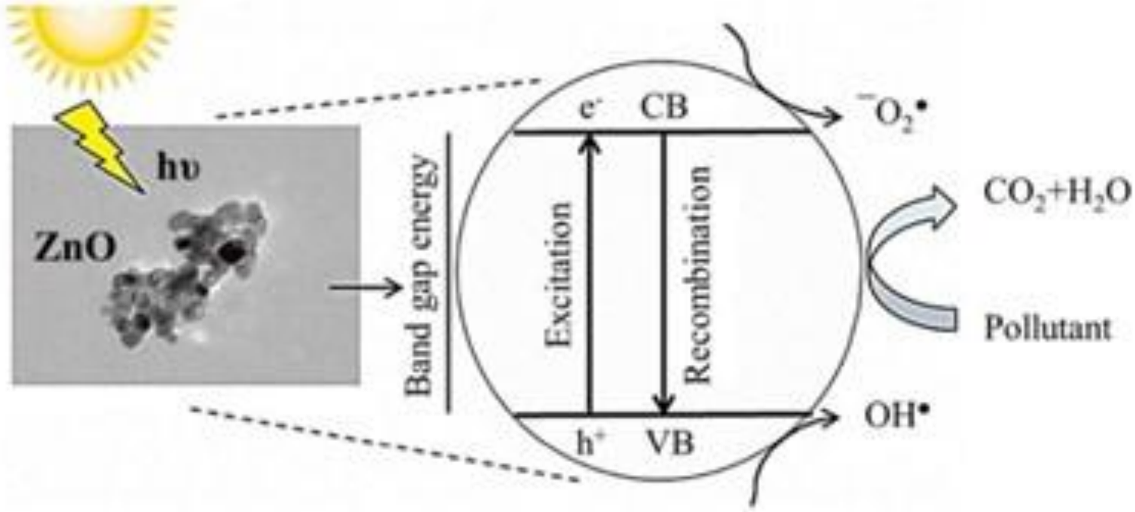

Figure 3. Degradation of dyes using the ZnONPs[72].

Table 5. Mechanism of ZnO NPs as photocatalyst

ZnO-derived plant extract	Type of pollutant	Efficiency Of removal dye	Type of irradiation	Size of ZnO	Mechanism	Ref.
ZnO drived broccoli extract	Methylene blue(MB) and Phenol red	74% and 71%	UV irradiation	14 and 17 nm	The hydroxide complexes were formed by the phenolic component. Broccoli contains quercetin, one of the biologically active compounds of vegetables that are green. It has a phenolic component, which is abundant in functional molecules with groups of hydroxyl. Their involvement lies in the transformation of Zinc(II) chloride into zinc hydroxide.	[73]
ZnO-derived Neem leaf extract.	Methylene blue	68%	UV light	25.97 nm for green method 33.20 nm for sol-gel method	An analysis comparing the sol-gel technique to the process of biosynthesis for the manufacturing of zinc oxide. The breakdown of MB had a matching effectiveness of 68%, and the primary contributing component to the breakdown process was OH ·radicals. It is determined that the biosynthetic approach is more effective than the sol-gel method for the synthesis of nanoparticles made from zinc oxide based on a comparison of the attributes of the two types.	[74]

ZnO-derived Prosopis juliflora leaf extracts	methylene blue	99%	UV light	65 nm	Electrons bound by photo-induced oxygen shortages may have the advantage of capturing oxygen trapped on the outermost layer. Subsequently, the oxidation process of organic compounds was likely to be accelerated by the O2 reactive molecule.	[75]
ZnO-derived Laurus obilis plant extract	azo dye	98%	UV- light	20 and 30 nm	The organic pigments are broken down into the gases carbon dioxide and water by the extremely reactive ensuing anionic in nature radicals.	[76]
ZnO-derived pomegranate extract	MB dye	86%	Sunlight	28 nm	After sixty minutes of exposure to sunshine, the two catalysts acquired comparable activity levels. Initially, the decolorization effectiveness of zinc oxide with the addition of extract was higher than that of bare zinc oxide. The modest variation in their energy band gap was identified as the reason for the same catalytic activity for zinc oxide produced irrespective of the extract from pomegranate peels.	[20]

Reference [21] demonstrated the mechanism of antioxidants in Moringa oleifera leaves chelate zinc (II) ions forming zinc oxide after the calcination process. The present investigation employed Fourier transform infrared spectroscopy (FTIR) to examine extracts from plants and zinc oxide nanoparticles that were obtained at two distinct temperature modifications (one hundred degrees Celsius and five hundred degrees celsius). Plant extracts showed absorption bands typical of bioactive compounds, while ZnO synthesized at 100 °C showed hydroxyl (-OH) stretching bands, which may indicate the formation of antioxidant-zinc complexes during synthesis. When Fourier Transform Infrared Spectroscopy absorption bands typical of bioactive chemicals were found in green-synthesised zinc oxide nanoparticles this result is consistent with those of various research groups [77] that Three chemical reactions of dissolved Zn^{2+} ions take place with phytochemicals of Moringa oleifera, i.e H. Phenolic acids, flavonoids and vitamin-based compounds. Changes in the chemical behavior of L-ascorbic acid and zinc nitrate, possible oxidation of biological compounds i.e. H. Free radical-mediated L-ascorbic acid to L-dehydroascorbic acid, followed by electrostatic attraction between the free radical and precursor cations.

7. Conclusion

The large area of particular surfaces, spectral properties, and flexibility of ZnO nanoparticles demonstrate their promise for good photocatalytic usage, particularly in the breakdown of organic contaminants, self-purifying, and antibacterial activity. Selective factors, dopant type, synthesis pathways, and substrate kind all have a substantial impact on increased catalytic activity. Furthermore, the choice of capping agent affects zinc oxide's overall photo catalytic efficiency, stable dimension, measurement of particles, and crystal structure. The phytochemicals, with their numerous bioactive compounds, reduce metal ions to nanoforms through reduction mechanisms. Consequently, we can control the stability, size, shape, and anti-aggregation of metal oxide nanoparticles.

Conflict of Interest Statement: The authors declare that there is no conflict of interest.

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