

Review Article

Plant-Mediated Biosynthesis and Photocatalysis Activities of Zinc Oxide Nanoparticles: A Prospect towards Dyes Mineralization

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In recent years, nanoparticles synthesis by green synthesis has gained extensive attention as a facile, inexpensive, and environmentally friendly method compared with chemical and physical synthesis methods. This review covered the biosynthesis of zinc oxide nanoparticles (ZnO NPs), including the procedure and mechanism. Factors affecting the formation of ZnO NPs are discussed. The presence of active bioorganic molecules in plant extract played a vital role in the formation of ZnO NPs as a natural green medium in the metallic ion reduction processes. ZnO NPs exhibit attractive photocatalysis properties due to electrochemical stability, high electron mobility, and large surface area. In this review, the procedure and mechanism of the ZnO photocatalysis process are studied. The effects of dyes amount, catalysts, and light on photodegradation efficiency are also considered. This review provides useful information for researchers who are dealing with green synthesis of ZnO NPs. Moreover, it can provide investigators with different perceptions towards the efficiency of biosynthesized ZnO NPs on dyes degradation and its restrictions.

1. Introduction

Eliminating environmental pollution has been under scrutiny and gained extensive attention. Dyes are considered seriously hazardous for the environment because dyes are highly soluble in water [1, 2]. Commonly, methyl orange, methylene blue, rhodamine B, Congo red, and orange G are used in several industries including pharmaceutical, textile, and food [3, 4]. Numerous oxidants agents are used for degrading organic contaminants [5]. Among these oxidants, persulfate as an oxidizing agent is highly considered due to its high effectivity. Despite the efficiency, persulfate still has some drawbacks. The efficiency depends on generating a high amount of free radical [6] and high-cost operation [7]. Moreover, most of the dyes are resistant to AOPs and biological degradation [2]. Therefore, researchers work towards nanotechnology as a new promising manner for the degradation of dyes.

Nanoparticles (NPs) refer to nanoscale materials with sizes between 1 nm and 100 nm and show superior properties based on dimensions, distribution, morphology, and large surface-to-volume ratio [8]. These unique functional

properties make nanomaterials a capable candidate for various applications in medicine, food, and engineering [9–11]. In recent years, metal and metal oxide nanoparticles were very encouraging materials applied in several fields due to their numerous valuable properties, such as catalytic, optical, magnetic, and electrical properties. Metal and metal oxide nanoparticles were used in various fields such as light-emitting devices, biomedicine, soil stabilization, catalysis, water treatment, transparent, conductive contacts, piezoelectric transducers, laser deflectors, solar cells, biosensors, and gas sensors [12–14]. Among metal oxide nanoparticles, zinc oxide nanoparticles (ZnO NPs) seem to be the excellent candidates for antibacterial activity, green agrochemicals, photocatalytic degradation, antimicrobial activity, and photocatalysis for environmental remediation [15, 16]. Interestingly, many researchers emphasized that the zinc oxide nanoparticle has unique properties for mineralization of organic pollution through a photocatalytic reaction and other applications [17, 18]. These properties and applications have been covered in the following sections according to the present literature.

In this review, the biosynthesis method of ZnO NPs, the procedure, and the mechanism are studied. A special focus has been given on the biosynthesis method's factors, including temperature, pH, presource, and plant extract. In the subsequent sections, the activity of the photocatalyst of ZnO NPs and mechanism are discussed. The factors affecting the efficiency of the ZnO NPs photocatalysis process were also studied. Likewise, this study highlighted the efficiency of biosynthesized ZnO NPs on the removal of organic dyes. A combination of ZnO NPs and AOPs techniques to improve the degradation of organic dyes was proposed. Finally, a brief discussion was made of limitation of photocatalytic efficiency of ZnO NPs.

In this study, the Internet, mainly the Google Scholar database, was used to find and download manuscripts related to biosynthesis and photocatalysis activities of ZnO NPs using suitable keywords such as organic pollution, zinc oxide, nanoparticles, and photocatalysis. The basis of the collection of the articles was (i) biosynthesis, (ii) zinc oxide nanoparticles, (iii) photocatalysis, and (iv) dyes degradation.

2. Zinc Oxide Nanoparticles

Intensive scientific work has taken place in recent years on ZnO NPs synthesis and applications. ZnO is a semiconductor multifunctional substance of the II–VI group with a wide band gap and having a hexagonal wurtzite crystal structure [19, 20]. ZnO NPs have good absorption of UV light; therefore, it is used in sunscreens, coatings, and paints industries [21]. Present, ZnO NPs have been used in food packaging materials due to they are inactive with the food constituents offering preservative effects [22]. ZnO NPs are also being used as a component in antibacterial creams, ointments and lotions, self-cleaning glass, ceramics, and deodorants [23].

The study conducted by Song et al. [24] presented good results in the effect of ZnO NPs on the antimicrobial and antifungal potentials. Doping of ZnO NPs with other metals such as Ag and Au enhances the antimicrobial and antifungal activities [25, 26]. Interestingly, ZnO NPs meet the requirements of photocatalysis characteristics such as electrochemical stability, high electron mobility, and large surface area. These unique characteristics of ZnO NPs plus low-cost and nontoxic lead to extensive studies of ZnO NPs syntheses and applications [27–30]. Therefore, in this regard, this study deals with the biosynthesis and photocatalytic activities of ZnO NPs.

2.1. Biosynthesis and Formation of ZnO NPs. Generally, there are two main methods for nanoparticle synthesis, chemical methods (down to top) and physical methods (top to bottom) [31]. Despite their popularity, both methods have many disadvantages such as energy consumption, expensive, and harmful to the environment which limit their applications [32]. In contrast, biosynthesis is a promising method for nanoparticle synthesis due to it circumventing the usage of hazardous chemicals, massive consumption of energy, high-cost operations, long and complicated processes, poor yielding, strict conditions for reduction and stabilization, and generation of hazardous by-products [33]. There are

many sources used for biosynthesizing methods to synthesize ZnO NPs, such as plants, bacteria, yeast, fungi, and sea weeds [10, 16]. Production of nanoparticles using plants displays important advantages over other microorganisms (Table 1), for instance, the low cost, easiness, short production time, safety, and the ability to up production volumes [10, 34]. Thus, using a plant extract as a green synthesize of ZnO NPs was considered in this review.

Moreover, the presence of highly active bioorganic molecules in plant parts such as roots, leaves, bark, and fruits leads to increased demand for ZnO NPs and to develop such low-cost, secure, and simplistic syntheses approaches. According to Jafarirad et al. [35], the aqueous extract mostly contains biomolecules, which include amino, hydroxyl, and carboxyl biofunctional groups. These biomolecules would act as metallic reductants and as protective agents to form a stabilizing layer on the biosynthesized nanoparticles.

2.2. Procedure for Biosynthesis of ZnO NPs. Due to the advances in using the biosynthesis approach in synthesis of ZnO NPs, the researchers have used different types of plants and different parts. There is no role in choosing a specific plant to synthesize ZnO NPs. The most important issue should concern that every part of the plant contains natural compounds that act as reducing agents. Different parts of the plant were used for synthesizing ZnO NPs such as leaves, flowers, and fruits extract due to enrichment of reducing agents compounds [35–38] (Table 2).

Different growth morphologies of ZnO NPs can be synthesized, for example, nanorods, nanospheres, nanotubes, nanowires, nanoneedles, and nanorings by controlling the synthesis parameters [33, 56]. Figure 1 describes the steps of the most commonly applied method of green synthesis of ZnO NPs by using plant extract. Step one, collect plant parts of interest and thoroughly wash with tap water and then adopt with distilled water to remove impurities and other unwanted materials. Step two, keep plant parts at room temperature to dry in the absence of sunlight, followed by either using it directly to obtain an extract or crushing until it becomes a powder to obtain the extract. Step three, ultrapure water is added to the powder according to the wanted concentration. The mixture is heated below 60°C with continuous stirring using a magnetic stirrer. Step four, the mixture is filtered to obtain clear plant extract, and then, the extracted solution is then mixed with the desired concentration of Zn salts such as hydrated zinc nitrate, zinc oxide, and zinc sulfate as a metal precursor. Then, heated under optimal pH, temperature, and time, changing of mixture color to yellow is a visual confirmation of the synthesized NPs [57–61].

The addition of external chemicals as stabilizers is undesirable because various plant metabolites present in the extract play a significant role in the bioreduction of metal ions and stabilizing agents for yielding nanoparticles.

2.3. Factors Influencing the Biosynthesis of ZnO NPs. The parameters of temperature, pH, the concentration of presource, concentration of plant extract, and reaction time

TABLE 1: Comparison between the plant extract and another microorganisms system.

Biological source	Plant	Microorganisms
Preparation time	Short	Longer than plant due to incubation time
Components responsible for reducing metal salts into metal nanoparticles	Phytochemicals	Enzymes/proteins
Production yield	High	Lower than plant
Solvent	Water	Natural source/extract
The cost	Effective	Less effective
Procedure	One-pot synthesis process	More complicated
The location of nanoparticles formation	Intracellular (expensive) and extracellular	Intracellular synthesis and extracellular synthesis.

have great effects on the formation and characteristics of ZnO NPs biosynthesis (Figure 2). Among these parameters temperature, the concentration of presource and plant extract plays a dominant role in forming nanoparticles' biosynthesis. There are many modern techniques used to confirm the formation of ZnO NPs. Such techniques are useful in monitoring the formation of ZnO NPs and studying the effects of parameters as well. This study focuses on UV-Vis, FTIR, XRD, SEM, and TEM techniques due to the availability, and most of the researchers used such techniques.

2.3.1. Effect of Temperature. The UV-Vis, FTIR, SEM, TEM, and XRD have been used by many researchers to study and to monitor the effect of temperature on the biosynthesis of NPs [62]. In the study carried by Bala et al. [63], temperatures of 30°C, 60°C, and 100°C were used. The UV-Vis spectra showed that no characteristics absorption band was detected at 30°C, indicating the absence of ZnO NPs, while at 60°C and 100°C, showed sharp surface plasmon resonance (SPR) bands at 377 nm, confirming ZnO NPs [63]. A study of the influence of different temperatures (25, 60, and 90°C) on biosynthesis and characterization of ZnO NPs was conducted by Mohammadi and Ghasemi [64]. They concluded that 25°C is the optimum temperature for synthesized ZnO NPs which are absorbed at 378 nm. The UV spectra show SPR peak indicating the combined vibration of the nanoparticle's electron with the light wave [65]. Similar results were reported by Ghorbani et al. [66]. Additionally, Pal et al. and Fakharia et al. [39, 67] successfully synthesized ZnO NPs at moderate temperature. The absorption spectrum of ZnO NPs by UV-Vis was shown around 350 nm and 361 nm, respectively.

Many researchers in the biosynthesis method used FTIR analysis to facilitate identifying functional groups existing in the plant extract that play an important role in the mechanism of bonding with ZnO NPs. In this regard, Alamdari et al. [40] revealed that any alteration in the position and intensity of peaks in the plant extract spectrum via sample spectrum could be correlated with the interaction of the functional groups' bioactive substance with the ZnO NPs. Moreover, Stan et al. [68] at 80°C presented the FTIR spectrum of the green synthesized ZnO NPs. The result displayed a sharp and intense band at 546 cm⁻¹, indicating ZnO vibrations' existence.

Bala et al. [63] observed functional groups in ZnO NPs by the FTIR study. The FTIR spectra show a peak at 482 cm⁻¹ at different temperatures (60°C and 100°C) which was confirmed the presence of ZnO NPs, and this finding is in accordance with Das et al. [69]. The effects of temperature clearly appear in FTIR spectra when comparing bioactive compounds' absorption at 60°C and 100°C. For instance, at 60°C, many bioactive compounds were absorbed on ZnO particles' surface, such as aromatic compounds, asymmetric stretching of C=C-C, symmetric stretch -C-C=C, and bending vibration of the alcoholic -C-OH. Whereas in 100°C sample, these phytoactive components were either not existed or remained absorbed on ZnO NPs in a small amount. These variations were due to the rise in temperature as the bioactive components were lost at 100°C as they were calcinated at higher temperature [63].

Meanwhile, Fakhari et al. [67] synthesized ZnO NPs at room temperature. The FTIR spectra were detected at 1634 and (600, 450) cm⁻¹ related to Zn-O stretching and deformation vibration. The same results have been reported by Singh et al. [70].

XRD patterns give information about the material of interest such as chemical composition, crystallographic structure, and physical characteristics [63]. The XRD patterns of the ZnO sample were existent, and characteristics peaks were observed at 60°C and 100°C. However, the sample synthesized at 30°C was free of such characteristic peaks as it was shapeless. Above 30°C, XRD spectra showed that well-defined peaks of ZnO were formed, and the crystallinity of the sample improved with temperature rise. Alamdari et al. [40] reported that at 80°C, well-defined peaks in XRD showed that the prepared ZnO NPs particles were extremely crystallized, with a wurtzite crystal structure, and size was around 17 nm. While at room temperature, Pal et al. [39] successfully synthesized ZnO NPs, and the XRD patterns show hexagonal wurtzite structure of an average grain size of 52 nm.

The effects of temperature on nanoparticle morphology were also noticed by FESEM (field emission scanning electron microscopy) and EDX (energy dispersive X-ray). Biosynthesis of ZnO NPs at different temperatures (30, 60, and 100°C) showed significant diversity in their FESEM micrographs, at 30°C showed irregular surface morphology, and at 60°C displayed spherical structure. While at 100°C, it was more crystalline in nature and was forming a dumbbell-shaped structure. Additionally, EDX spectra clearly showed

TABLE 2: Examples of biosynthesized ZnO NPs and their photodegradation efficiency.

No.	Plant source	Part extracted	Presource of ZnO NPs	Size	Type of ZnO NPs (catalyst)	Light source	Type of dyes	Degradation efficiency % (time)	Reusability	References
1	<i>Coriandrum sativum</i>	Leaf	Zinc acetate	24 nm	ZnO NPs	Direct sun light irradiation	Yellow 186 dye	93.3–130 min	3 runs	[2]
2	Oak	Fruit hull	Zinc acetate dihydrate	34 nm	ZnO NPs	Visible light	Basic violet 3	93–2:30 h	—	[17]
3	<i>Garcinia mangostana</i>	Fruit	Zinc nitrate hexahydrate	21 nm	ZnO NPs	Solar light	Malachite green dye (MG)	99–180 min	—	[20]
4	<i>Mussaenda frondosa</i>	Leaf/stem	Zinc nitrate	5–20 nm	ZnO NPs	UV	MB	80–100 min 30–100 min	—	[33]
5	<i>Moringa oleifera</i> (drumstick)	Leaves	Zinc acetate dihydrate	52 nm	ZnO NPs	Visible light	Titan yellow dye	96–60 min	—	[39]
6	<i>Sambucus ebulus</i>	Leaf	Zinc acetate dihydrate	17 nm	ZnO NPs	UV	MB	80–200 min	—	[40]
7	<i>Vitex trifolia L</i>	Leaf	Zinc nitrate hexahydrate crystal	28 nm	ZnO NPs	UV	MB	92.13–90 min	—	[41]
8	<i>C. sinensis</i>	Peel	Zinc nitrate	22.6 nm	ZnO NPs	UV	MB	83–120 min	—	[42]
9	<i>Brassica oleracea L. var. italica</i>	Broccoli leaves	Zinc chloride ZnCl ₂ ·7H ₂ O	14–17 nm	ZnO NPs	UV	MB/phenol red (PR)	74–180 min 71–180 min	—	[43]
10	<i>Tabernaemontana divaricata</i>	Green leaf	Zinc nitrate	20–50 nm	ZnO NPs	Sunlight	MB	≈100–90 min	—	[44]
11	<i>Coriandrum sativum</i>	Leaf	Zinc acetate dihydrate	9–18 nm.	ZnO NPs	UV	Anthracene	89–240 min	—	[45]
12	<i>Dolichos lablab L.</i>	Leaf	Zinc acetate dihydrate	29 nm	ZnO NPs	Visible and near-UV	MB/rhodamine B (RhB), orange II (OII)	80–210 min 95–210 min 66–210 min	—	[46]
13	<i>Carica papaya</i>	Latex	Zinc nitrate	11–26 nm	ZnO NPs	UV	Alizarin red-S dye	99–120 min 100–70 min	4 runs	[47]
14	Sea buckthorn	Fruit	Zinc nitrate hexahydrate	17.15 nm	ZnO NPs	UV	Malachite green/Congo red/MB/eosin Y	99–80 min 99–70 min 100–90 min 96–80 min 38–80 min 94–80 min 35–80 min	5 runs	[48]
15	<i>Cannabis sativa</i>	Leaf	Zinc acetate	34 nm 38 nm	Ag-ZnO NPs/ ZnO NPs	Solar light	Congo red/methyl orange	—	—	[49]
16	<i>Ferulago angulata</i>	—	Zinc acetate dihydrate	32–36 nm	ZnO NPs	Visible light	Rhodamine B	93–3.5 h	—	[50]
17	<i>Chlorella</i>	—	Zinc nitrate	20 ± 2.2 nm	ZnO NPs	UV	Dibenzothioephene (DBT)	97–3 h	5 runs	[51]
18	<i>Euphorbia prolifera</i>	Leaf	Zinc chloride + ZnO NPs	5–17 nm	Cu/ZnO NPs	UV	MB/Congo red (CR)	100 ≈ 0 min/≈ 100–9 min	5 runs	[52]
19	<i>Azadirachta indica</i>	Leaves	Zinc nitrate	9–38	ZnO NPs	Sunlight/UV light	MB	92–120/ 85–120 min	—	[53]
20	Betel	Leaves	ZnO acetate	50 nm	ZnO NPs	Solar/UV	MB	90–210 min 96–210 min	—	[54]
21	Guava	Leaves	AgNO ₃	56.1 nm	Ag-ZnO NPs	Solar light	MB	98–60 min	6 runs	[55]

the effect of temperature in terms of carbon and nitrogen presence in 60°C and absence in 100°C, indicating bioactive compounds were adsorbed in 60°C [63]. Fakhari et al. [67] succeeded in synthesizing ZnO nanoparticles at room temperature with spherical shape and size of 21.49 and 25.26 nm. The high purity of synthesized nanoparticles is confirmed by EDX analyses.

Although many research studies have similarity of temperature for synthesis of ZnO NPs, there are still varieties in UV-Vis absorption, FTIR spectra, XRD, SEM, and TEM analyses. This, to some extent, might be due to the difference in methods and experiment conditions.

2.3.2. Effect of pH. Numerous investigations do not mention the pH effect in the preparation of nanoparticles. However, pH has a vital role in the formation and characteristics of nanoparticles. For those investigations, I assumed they used a neutral solution. Natural phytochemicals compounds existing in an extract are highly linked to the pH changes and consequently to a charge change in the solution, which might change their reducing and capping capability and then nanoparticle growth. Accordingly, this, in turn, may affect the morphology and yield of nanoparticles [71]. Mohammadi and Ghasemi [64] studied the effect of pH (4, 6, 7, 8, and 10) on the biosynthesis of ZnO NPs. It was detected that the color of the solution changed from light brown to colloidal brown as changing in pH from 4 to 10. Also, the effect of pH on reducing metal ions was considered by UV-Vis spectroscopy. It shows different absorption peaks ranging from 350 nm to 373 nm as pH changing from 4 to 10.

Nagarajan et al. [72] investigated the effects of pH (5, 6, 7, 8, 9, and 10) on the biosynthesis of ZnO NPs using seaweeds. No absorption peak was observed at pH in a range from 5 nm to 7 nm. Nevertheless, at pH 9 and 10, no absorption peaks were detected. Similarly, the detected absorption wavelength at 365 nm in pH 8 showed the total reduction of zinc nitrate to zinc nanoparticles [72].

Generally, nanoparticle synthesis's optimal pH depends on a substrate or biogenic molecule as a capping agent to synthesize nanoparticles. However, Mata et al. [73] indicated clearly that higher pH had favored higher reducing power.

2.3.3. Effect of Presource. The type and concentration of zinc salts affect significantly surface morphology and structure of synthesized ZnO NPs according to a study conducted by Fakhari et al. [67]. They studied the effect of precursor (zinc acetate and zinc nitrate) on structures, shape, and size of synthesized ZnO NPs [67]. The SEM results illustrated that using zinc acetate results in small spherical structures of zinc oxide. When using zinc nitrate, the spherical ZnO NPs are formed and accumulate to form flower-shaped bundles. Compared with the study conducted by Vijayakumar et al. [74], some differences were observed in terms of shape and size of synthesized ZnO NPs, though the same used presource (*L. nobilis* leaves and zinc acetate) for synthesis of ZnO NPs. This might be due to some difference in reaction conditions.

On the other hand, Mohammadi and Ghasemi [64] explored the effect of presource concentration. They used zinc nitrate as a presource with different amounts (0.005, 0.02, 0.05, and 0.3 M) for the biosynthesis of ZnO NPs. The SEM image shows a hexagonal structure of nanoparticles with an average size from 20.7 nm to 96.5 nm, convincing evidence of the zinc salt concentration on morphology of ZnO NPs [64].

2.3.4. Effect of Plant Extract. Plant extract plays a dual role in nanoparticle synthesis processes: reducing agents and other as stabilizing agents [17, 75]. Therefore, it is a very important factor in the biosynthesis process. Many of biological sources are used for ZnO biosynthesis such as *Carica papaya*, *Eucalyptus globulus*, *Corymbia citriodora*, *Nephelium lappaceum* L, and *Lycopersicon esculentum* (Table 2) [76–78]. The type of plant used has a limited effect on the formation and morphology of synthesized nanoparticles unless the extract is poor of bioactive compounds contributes to the metallic ion reduction processes [79]. Whereas the concentration of extract has an effect significantly on shape, homogeneity, and size of synthesized ZnO NPs.

Elumalai et al. [41] studied the influence of the concentration of *V. trifolia* extract on ZnO NPs morphology. The SEM image illustrates that ZnO NPs have different morphologies by increasing the *V. trifolia* extract volume from 10 ml to 40 ml. Using 10 ml of *V. trifolia* extract (capping agent) results in nearly spherical shapes of ZnO NPs. Increasing *V. trifolia* extract to 20 ml, the morphologies of ZnO NPs looked spherical. Still rising the volume of *V. trifolia* extract to 30–40 ml, ZnO NPs were extremely agglomerated because the smaller size nanoparticles bind together and formed secondary bulk nanoparticles [41]. Luque et al. [42] reported the effect of *Citrus sinensis* peel extract concentration (1%, 2%, and 4% by weight) on the surface morphology and size of the ZnO NPs. The HRTEM micrographs show different extract concentration influences in the size, shape, and surface morphology of the ZnO NPs [42].

2.3.5. Effect of Reaction Time. Reaction time is the time required for the completion of the reaction. Mainly, a plant extracted solution is mixed with the desired concentration of metal precursor and then heated under optimal conditions until changing of mixture color (step four). Time-consuming for the biosynthesis of ZnO NPs depends completely on temperature, pH, plant extract, and presources. Therefore, in this study, the effect of reaction time and centrifugation forces on ZnO NP's biosynthesis is not highly concerned.

In general, controlling all parameters including temperature, pH, presource, plant extract, and reaction time should be highly considered to form specific ZnO NPs for the desired application. However, controlling the constancy and aggregation of nanoparticles, adjusting crystal growth, morphology, size, size distribution, and separation of formed nanoparticles for more applications are the most challenging and problematic procedures and still in the development stage.

2.4. Mechanism of Formation of Biosynthesized ZnO NPs. Many investigations have been performed to understand the mechanisms of biosynthesis of nanoparticles. Moreover, they proposed several mechanisms. However, the mechanism of biosynthesis of nanoparticles is still not well understood and requires more studies.

The most plausible general mechanism is concluded that the functional groups existing in the plant extract such as alcohol, polyphenols, flavonoids, aromatic, and aliphatic amines play an important role in the green synthesis of ZnO NPs [80–82]. The aqueous extract of the plant that contains a high amount of organic components contributes to the metallic ion reduction processes. Luque et al. [42] proposed organic extract where ligation occurs between the functional components of the extracts and the zinc precursor. Aromatic hydroxyl groups present in some of these components ligate with zinc ions and form zinc-ellagate structures that undergo direct decomposition when heated to form ZnO NPs. The FTIR analysis showed different bands corresponding to organic content within the samples such as C-H (aromatic) functional groups, C-C stretching of aromatic rings, and Zn-O, which confirm that the material is zinc oxide [42].

A corresponding mechanism was reported for starch-rich potato extract as green natural medium to form ZnO NPs and Fe₃O₄ NPs [83, 84]. Starch contains amylose and amylopectin with presences of extensive number of hydroxyl groups and aldehyde. Hydroxyl groups act to facilitating the complexation of Zn ions to the molecular matrix, and aldehyde terminals act as reduction of the Zn (II) ions to Zn (0) nanoparticles [83]. The FTIR results confirmed the existence of organic compounds along with new inorganic material (Zn-O) in the uncalcined ZnO powders, and the adsorption peaks of stretching vibrations of Zn-O were detected at 485 cm⁻¹ [83].

In general, the analysis FTIR spectrum leads to determine the potential functional groups of biomolecules that are responsible for the formation of ZnO NPs. Any shift or change in the position and intensity of FTIR peak in the sample can be correlated with bioreduction reactions between functional groups and biomolecules and between the interaction of a functional group of organic molecules with the ZnO NPs [20, 40, 83]. A most likely new sharp peak in the sample ranging from 400 cm⁻¹ to 600 cm⁻¹ correlated to the ZnO NPs [20, 40, 83].

2.5. Characterization of ZnO NPs. There are many different methods and techniques used to characterize nanostructure. In general, there are two important techniques: one is to characterize and confirm the morphology of the nanostructure and the other to characterize the chemical properties of the nanostructure. Most likely, the selected techniques depend on the nature of the study and desired applications. Different methods and techniques proved different characteristics of morphology and different chemicals forms of the nanostructure. Accordingly, the instruments class, sample pretreatment and experiment conditions were highly considered to get reliable results. Usually, UV-Vis, FTIR, XRD, SEM, TEM, FESEM, and EDX

techniques were frequently used in nanostructure investigations [85–87].

3. Photocatalysts Activity of ZnO NPs

Photocatalysis is composed of two well-known words, photo and catalysis. The photocatalyst is a term that means photon responded for producing catalytically active species. Hagen [88] defined photocatalysis as “a change in the rate of chemical reactions or their generation under the action of light in the presence of substances called photocatalyst that absorbs light quanta and is involved in the chemical transformations of the reactants.”

The photocatalysis is a unique method that can be used for several purposes such as antibacterial activity, degradation of various organic pollutants in wastewater (Table 2), production of hydrogen, purification of air, and drug delivery. Among these applications, recently, the degradation of organic pollutants in wastewater attains great attention. Compared to other methods, the photocatalytic process is getting more attention in wastewater treatment due to the complete removal of dyes in moderate conditions of temperature and pressure. Moreover, the photocatalytic process is more favorable than AOPs due to no sludge being produced and using visible light or near-UV light as irradiation source of cost-effectiveness.

Photocatalytic activity is a process of interaction between catalysis and visible or UV light to generate reactive species such as OH• and O₂•- that could interact with organic pollutants, resulting in the removal of organic pollutants [43]. Recently, nanoscale metal oxides' development has critically increased the catalytic activity. The very high surface to volume ratio of nanostructures makes them effective and promising for photocatalysis and other applications. The nanoparticle catalysts have superior photocatalytic effectiveness as compared to normal photocatalyst substances [75].

Many researchers have used TiO₂, ZnO, SnO₂, and CeO₂ as photocatalysts [89–91]. Among these semiconductors, TiO₂ and ZnO have close band gap energies which exhibit photocatalytic degradation and cost-effectiveness. Thus, they are considered an effective alternative for the mineralization of organic pollutant.

TiO₂ and ZnO have an excellent semiconductor photocatalyst behavior because they are (i) highly photocatalytic, (ii) able to utilize visible and/or near-UV light, (iii) having high physical and chemical stabilities, (iv) photostable (constancy toward photo corrosion), (v) low-priced, and (vi) nontoxic [92–94]. However, compared to TiO₂, ZnO NPs are favorable due to its cost-effectiveness because of their low cost, high photochemical reactivity, and nontoxic nature [44, 95].

3.1. The Mechanism of the ZnO NPs Photocatalysis Process.

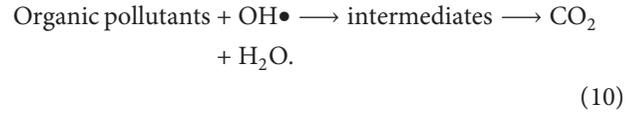
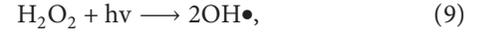
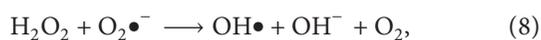
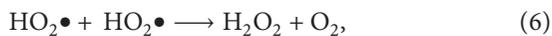
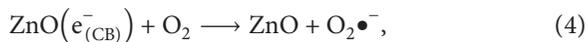
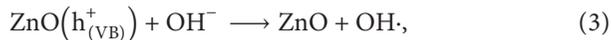
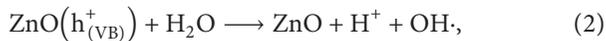
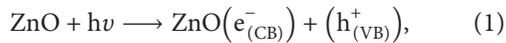
The photocatalytic process is a reaction that depends on the catalyst and wavelength of light energy (photon), whether the light from sunlight or artificial light energy. Many researchers have described the mechanism of ZnO NPs

photocatalytic reaction. Herrmann et al. [96] reported that ZnO NPs degrade organic pollutants as follows:

- (i) Organic contaminants diffuse from the liquid phase to the surface of ZnO NPs
- (ii) Adsorption of the organic contaminants on the surface of ZnO NPs and then oxidation and reduction reactions take place in the adsorbed phase
- (iii) Desorption of the products and consequent removal of the products from the interface region

Ong et al., Ibhaden et al., and Lam et al. [31, 97, 98] explored the mechanism photocatalytic reaction of ZnO NPs as follows:

- (1) Photocatalytic reactions are initiated when ZnO surface is exposed by a radiation of photonic energy (hv) equal to or greater than the ZnO band gap
- (2) The photonic energy responds to excitation of the electrons and then produces electron-hole (e^-/h^+) pairs, one is a positively charged hole in the valence band (VB) and the other is a negatively charged electron in the conduction band (CB) (equation (1))
- (3) The electron-hole pairs can transfer to the ZnO surface and be involved in redox reactions
- (4) The positive holes (H^+) created in the valence band reacts with absorbed water and hydroxide ions to produce powerful hydroxyl radicals. Then, hydroxyl radicals degrade the organic pollutants adsorbed on the surface of ZnO (equations (2) and (3)).
- (5) The conduction band electrons react with dissolved oxygen species to produce superoxide radical anions and then hydrogen peroxide (equations (4)–(6))
- (6) Hydrogen peroxide will then react with superoxide radicals to form hydroxy radicals which react with pollutants adsorbed on the surface of ZnO and then produce intermediate compounds that converted to green compounds such as CO_2 , H_2O , and mineral acids (equations (7)–(10))



The photocatalytic oxidation reaction depends on the generation and recombination of electrons and holes in the ZnO photocatalyst (Figure 3). Therefore, to ensure the photocatalytic process is accomplished, ZnO's quantum size, specific surface area of ZnO, and organic pollution concentration should be highly considered [99–103].

3.2. Procedure and Evaluation of the ZnO Photocatalysis Process. Evaluation of the effectiveness of the photocatalysis process (the percentage degradation of dyes) most likely was determined using the following equation [57]:

$$\text{Degradation \%} = \frac{C_i - C_f}{C_i} \times 100, \quad (11)$$

where C_i is the initial concentration of the dyes, and C_f is the final concentration of the dye. The dye degradation was monitored by PL spectroscopy and UV spectrum, and the spectra were taken at different times of irradiation [33].

3.3. Factors Affecting the Efficiency of the ZnO NPs Photocatalysis Process. Dyes, catalysts, and light are the main aspects that are contributing significantly on the photocatalysis process (Figure 4). Each factor plays a vital role in the photocatalysis process.

3.3.1. Influence of Dyes. The photocatalysis process is mainly a chemical reaction. Therefore, the pH, reactants concentration, and temperature are very important. An increase in reaction temperature mostly leads to an increase in reaction rate and accordingly photocatalytic activity. Hussien and Abass [104] approved that higher temperature helps decompose the pollutants in wastewater concerning the reaction time. However, Hassan et al. [45] showed that increasing temperature ranging 25–40°C causes a slight enhancement of photocatalytic degradation. Furthermore, Guettai and Amar [105] revealed that higher temperatures are responsible for removing oxygen from the reaction mixture, which is essential for the contaminant oxidation, and/or contaminant particles that might become more desorbed away from the catalyst surface lowers the reaction rate. The pH variation can also affect the reaction rate by changing the surface charge of reactants particles and shifts the potentials of catalytic reactions. Consequently, the dye's adsorption on the surface is changed and causes a change in the reaction rate [106].

The rate of photocatalytic degradation performance of a certain pollutant depends on significant factors that are governing the photocatalysis process. These factors can affect individually or/and together with the process. Kahsay et al. [46] reported that cationic dyes such as methylene blue and

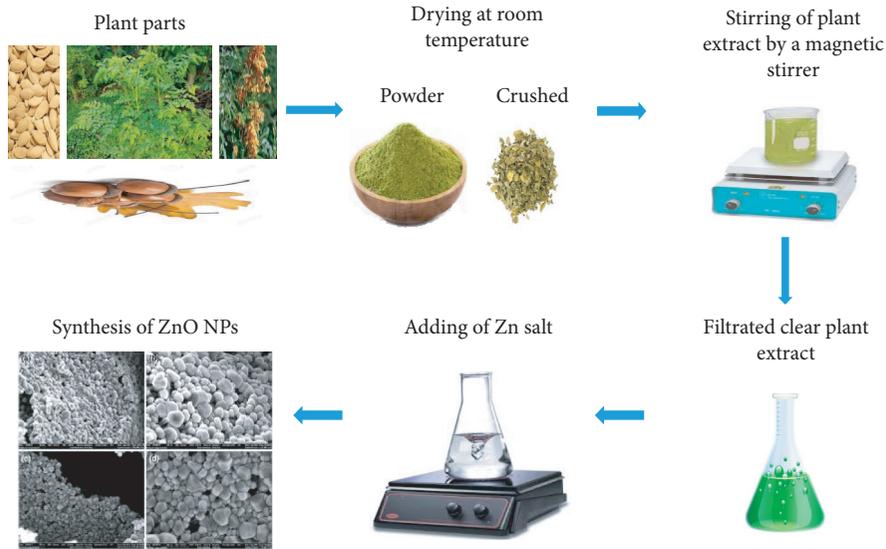


FIGURE 1: Proposed procedure for the biosynthesis of ZnO NPs.

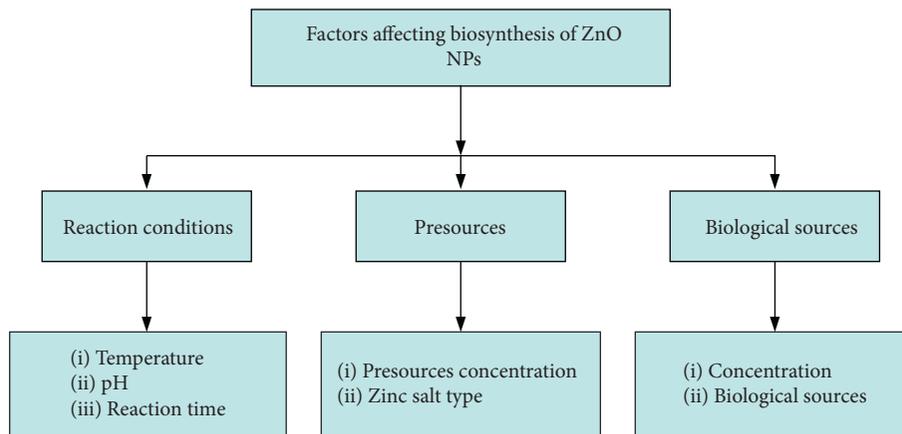


FIGURE 2: Factors' effect on the biosynthesis of ZnO NPs.

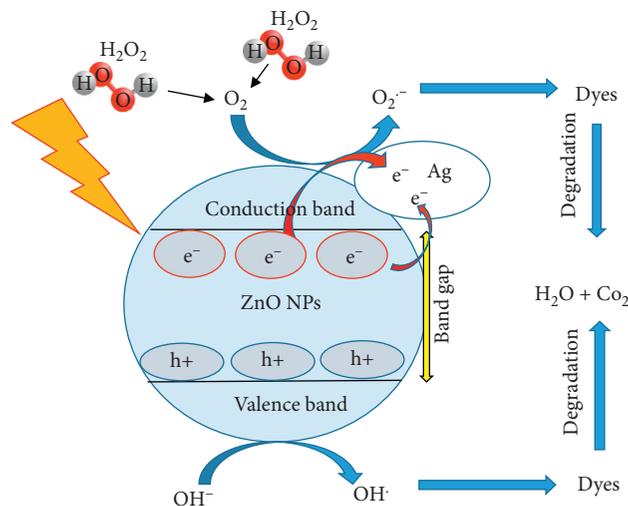


FIGURE 3: The proposed photocatalytic mechanism of ZnO nanostructures.

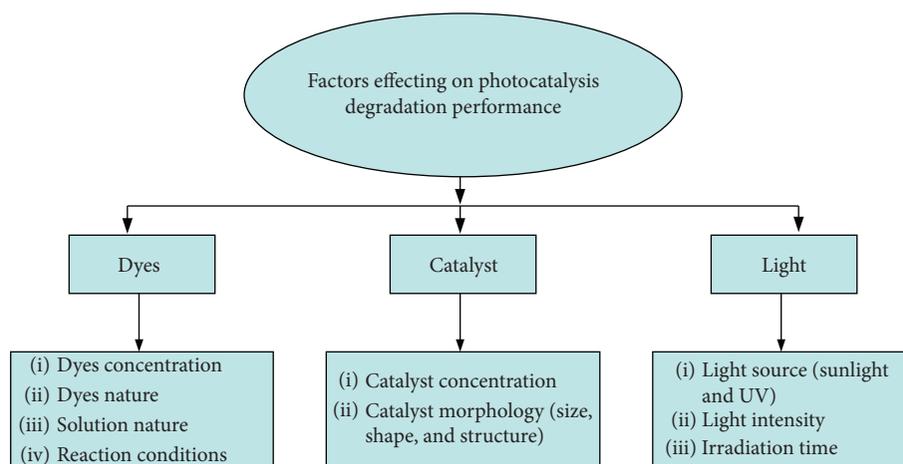


FIGURE 4: Factors' effect on photocatalysis processes of ZnO NPs.

rhodamine B increase the pH and lead to increase photocatalytic degradation, mostly due to the production of more hydroxyl ions on the surface of the photocatalyst. In contrast, increasing anionic dye's pH, such as orange II, decreased its photocatalytic degradation due to less hydroxyl radical formations on the surface of the photocatalyst. On the other hand, increasing the time of contact between the photocatalyst (ZnO NPs) and organic dye facilitated photodegradation of all organic dyes under visible and near-UV photoirradiation [46]. Generally, the matrix of polluted water (cationic, anionic dye) is different from each other. Thus, there are different degradation processes strategies. On the other hand, there is a reverse fit between dye concentration and the percentage degradation [107].

3.3.2. Influence of Catalysts. Catalyst is an important part of the photocatalysis process because of the oxidation or reduction reactions taking place on the catalyst surface. Therefore, many studies focus on the effects of the concentration, crystal structure, shape, size, and surface area of the catalyst on photocatalysis degradation [108, 109].

Based on Kahsay et al.' [46] study, the catalyst concentration has an effect significantly on the efficiency of photocatalytic degradation. Hassan et al. [45] also revealed that degradation efficiency increases by increasing the photocatalyst concentration of $1000 \mu\text{g L}^{-1}$. This is due to the increase in the number of active sites on the catalyst surface that lead to an increase in the number of absorbed photons and consequently, production of a large number of $\text{OH}\cdot$ radicals. While increasing the photocatalyst concentration greater than $1000 \mu\text{g L}^{-1}$, the solution becomes turbid, decreasing the effectiveness of the catalyst activation during the UV irradiation [45].

Moreover, the structure and morphology of the catalyst are critical factors in the photocatalytic process due to governing the surface area. When the surface to-volume ratio is increased, a massive number of atoms are gathered on the surface of a catalyst that enhance the number of active sites and interfacial charge carrier transfer rates, thereby achieving higher catalytic activities [110]. It has been

demonstrated that the structural and morphological characters, such as the crystalline form, shape, and size of semiconductors, are correlated to the photocatalytic activity [111].

Sharma [47] revealed that the photocatalytic activity of the ZnO NPs increased from 66% (prismatic tip) to the highest activity 99% (nanoflower), and nanobuds exhibited the second lowest activity (70%). In contrast, Saravanan et al. [108] reported that spherical-shaped ZnO shows better degradation efficiency than the spindle and rod-shaped ZnO due to its large surface. Also, Kenanakis et al. and Ma et al. [112, 113] studied the variation of photocatalytic performance of ZnO NPs based on the shape of ZnO NPs.

On the other hand, along with the efficiency of degradation, the catalyst should be stable, chemically active, cost-effective, environmental friendly, and reusable as these properties are important tools for industrial applications [114]. The recyclability of biosynthesized ZnO NPs as photocatalyst has been studied by many researchers [2, 47]. Singha et al. [2] reported significant photostability and reusability of ZnO NPs for photocatalytic degradation of yellow 186 dye. The XRD analysis indicates that the structure of ZnO NPs remains same as before the photocatalytic process.

3.3.3. Influence of Light. Light is a crucial factor in the photocatalysis process. In fact, it is the skeleton of the photocatalysis process. Light source (sunlight or UV), light intensity, and irradiation time are very important to the photocatalysis process degradation. The degradation rate of photocatalytic reaction completely depends upon the light intensity, the light of irradiation response to exit the electron from the valence band to the conduction band or equal to band gap energy.

Rupa et al. [48] reported that the photodegradation of Congo red and methyl orange was highest at 80 min irradiation of visible light on the photocatalytic reaction. Other similar studies by Aminuzzaman et al., Pal et al., and Chauhan et al. [20, 39, 49] have also shown that the photodegradation efficiency increases with the increase of time

irradiation on the photocatalytic reaction. On the other hand, many researchers confirm that ZnO possesses high photocatalytic efficiency and has a high UV light response [33, 46]. Interestingly, ZnO NPs can absorb UV radiation and visible light, and this property enhanced photocatalytic efficiency and cost-effectiveness [50, 115].

In an optimum condition of photocatalytic processes such as temperature, pH values, and irradiation time, some researchers add a small amount of H_2O_2 to dye solution during the degradation process to provide more dissolved oxygen [46]. This strategy opens the door to using advanced oxidation processes (AOPs) with photocatalytic processes (ZnO NPs) to execute and enhance the degradation efficiency.

3.4. The Limitation of Biosynthesized ZnO NPs in the Photocatalytic Process. Despite the advantages of biosynthesis of nanoparticles in facile preparation, mild reaction condition, ecofriendly approach, and cost cost-effectiveness [116, 117], there are growing global concern on their capability to move to the large-scale production, i.e., production of nanoparticles in the pilot plant level using an ecofriendly and biocompatible process to be effective in industries and commercialization. Compared with other chemical and physical methods, still biosynthesis methods have some drawbacks that have to be negotiated such as difficulties to control all parameters to synthesize nanomaterials for actual/specific applications, as well as controlling the constancy and aggregation of nanoparticles. Further investigations should be performed to recognize controlled and optimized conditions for large-scale production of green NPs. However, Nagarajan et al. [72] proposed that using *S. myriocystum* as a biosource appears to be a potentially exciting tool for large-scale synthesis of ZnO NPs. Buazar et al. and Liu et al. [83, 118] reported a novel green one-step synthesis of ZnO NPs with the potential for large-scale production as an economical dye removal product for the industry. Abdelhakim et al. [117] succeeded to scale up the production of biosynthesized ZnO NPs for better exploration in the near future for many medical, agricultural, and industrial applications. Khalafi et al. [51] examined the performance of the catalytic activity of biosynthesized ZnO NPs in real polluted environment, and their results demonstrated high efficiency and durability of green ZnO NPs.

In general, the industrial and commercialization of nanomaterials for water and wastewater technology depend mainly on their impact on the aqueous environment [119]. Moreover, a number of issues should be considered such as collecting nanomaterials after treatment, preventing agglomeration of nanomaterials in the wastewater, and leaching of new contaminations from the nanomaterial composites to the wastewater sample. However, evaluation of nanoparticles stability in the environment can be achieved by estimating their propensity to aggregate or interact with the surrounding media.

The wide band gap of ZnO NPs plays a key role in the limitation of the photocatalytic efficiency under visible and near-UV photoirradiation [120, 121]. Indeed, the band gap

of ZnO restrains its photocatalytic activity within the UV light range. Consequently, it can consume a small amount of incident solar radiation [122]. Additionally, the photocatalytic activity of ZnO NPs is governed by its ability to create photogenerated electron-hole pairs, and the major disadvantage and constraint of ZnO NPs as a photocatalyst is rapid recombination of photo-excited electron-hole pairs [123]. To overcome this problem, many efforts have been made to adapt ZnO's properties, such as ion irradiation and metal doping which is a common strategy [124]. Several types of metal dopants have been experienced, including anionic dopants, cationic dopants, rare-earth dopants, and codopants such as Ag, Au, and Pt. Figure 3 shows the photocatalytic mechanism of ZnO NPs [125–127]. Furthermore, many investigations have shown that coupling with other semiconductors, such as CdO, CeO₂, SnO₂, TiO₂, graphene oxide (GO), and reduced graphene oxide (RGO), is a promising approach to enhance the photodegradation efficiency of ZnO NPs [128–130].

4. Conclusion

In this study, biosynthesis of ZnO NPs using plant extract has been exposed to be a potential candidate because of low-cost, secure, and simplistic syntheses approaches compared with other microorganisms. Extracted phytochemicals act as reducing agents and stabilizing agents during the formation of the ZnO NPs. Controlling synthesis parameters should be highly concerned to ensure the formation of ZnO NPs of interest. However, controlling the constancy and aggregation of nanoparticles, adjusting crystal growth, morphology, and separation of formed nanoparticles for more reusability are the most difficult parts and still in the development stage.

As can be seen from the survey of recent literature presented here, ZnO NPs have unique properties that make it a great candidate in many technologies such as dyes degradation, hydrogen production, antibacterial activity, and drug delivery. Using ZnO NPs as a catalyst is promising for dye degradation due to its ability to absorb a larger fraction of the solar spectrum and nontoxic behavior. The type and amount of dyes and catalysts are very important factors in photodegradation efficiency. According to the literature survey given here, the biosynthesized ZnO NPs can be considered as one of the most effective methods to remove dyes from water and wastewater.

Conflicts of Interest

The author declares that there are no conflicts of interest.

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