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Green synthesized zinc oxide nanostructures and their applications in dye-sensitized solar cells and photocatalysis: A review

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ABSTRACT

The demand for clean energy and sustainable technologies has led to extensive research in the field of nanomaterials, particularly zinc oxide (ZnO) nanostructures, due to their unique properties and promising applications. This review focuses on the green synthesis methods of ZnO nanostructures, emphasizing eco-friendly and sustainable approaches that minimize environmental impact. Various green synthesis techniques, such as plant-mediated, biological, and environmentally benign chemical methods, are discussed, highlighting their advantages over conventional processes. Moreover, the review explores the applications of ZnO nanostructures in dye-sensitized solar cells (DSSCs) and photocatalysis. DSSCs represent a prominent alternative to traditional solar cells, and the integration of ZnO nanostructures as photoanodes holds great potential for enhancing efficiency and performance. The underlying mechanisms of ZnO nanostructures in DSSCs are examined to provide a comprehensive understanding of their role in photovoltaic applications. Additionally, the photocatalytic properties of ZnO nanostructures are investigated, with a particular focus on their ability to degrade organic pollutants and their potential in wastewater treatment and air purification. The photocatalytic mechanisms and factors influencing the efficiency of ZnO nanostructures in these processes are critically analyzed. In conclusion, this review presents an overview of the state-of-the-art green synthesis methods for ZnO nanostructures and discusses their applications in DSSCs and photocatalysis. The environmentally friendly nature of these synthesis approaches and the multifunctional capabilities of ZnO nanostructures make them promising candidates for sustainable energy conversion and environmental remediation technologies. However, challenges and future prospects in the field are also highlighted to guide further research and development in this area.

1. Introduction

Zinc oxide (ZnO) is a chemical compound composed of zinc and oxygen atoms. It appears as a white, powdery solid in its natural form. ZnO is an essential semiconductor material widely used in various applications, such as electronics, optoelectronics, ceramics, and cosmetics [1,2]. It possesses unique properties like high transparency in the visible spectrum, excellent electrical conductivity, and remarkable photoconductivity. Due to its broad range of applications and relatively low cost, ZnO has gained significant attention in the fields of research [3,4]. Zinc oxide nanoparticles (ZnO-NPs) are tiny particles with dimensions typically ranging from 1 to 100 nm. Due to their nanoscale size, they exhibit unique properties and behaviors compared to bulk ZnO [5,6]. The properties of ZnO-NPs have a high surface-to-volume ratio, which means they have a large surface area relative to their volume. Their shape can vary from spherical to rod-like. ZnO-NPs exhibit interesting optical properties, including strong UV absorption and visible light emission (photoluminescence) [7,8]. These properties are useful in various optoelectronic applications. ZnO-NPs have been found to possess significant antibacterial and antifungal properties, making them attractive for use in medical applications, such as wound dressings and antimicrobial coatings. ZnO-NPs can act as catalysts, promoting various

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chemical reactions, which makes them valuable in industrial processes [9,10]. Bulk ZnO-NPs also display piezoelectric properties, generating electrical charges under mechanical stress. This characteristic is utilized in sensors and energy-harvesting devices. ZnO-NPs are semiconductors with a wide bandgap, making them suitable for electronic applications, including thin-film transistors and light-emitting diodes [2,11]. The large surface area of ZnO-NPs results in higher reactivity with other substances, leading to potential applications in sensors and adsorbents. In different device applications, metal oxide nanostructures such as zinc oxide (ZnO), tin oxide (SnO_2), titanium oxide (TiO_2), and iron oxide (Fe_2O_3) are very desirable and play an important role [12,13]. They are also commonly used in optics, biosensors, photonics, the medicinal field, photo-electrochemical devices, antimicrobial activity, drug delivery, bio-imaging, electronics, sensors, food preservations, and catalysis [1, 14]. Rock salt, hexagonal wurtzite, and cubic structures are the abundant forms of a ZnO semiconductor. The direct bandgap energy of wurtzite form is 3.40 eV. ZnO conductivity and luminescence improve the property of UV filtering and optical absorption, resulting in an advanced ZnO nanostructure bio-sensing capability with its unique physical and chemical properties, for example, high photostability, and high electrochemical coupling coefficient [15,16]. ZnO is a versatile material due to its high radiation absorption capacity and chemical stability [17,18]. Materials science classifies ZnO as a semiconductor in group II-VI with a covalence that is on the cusp of covalent and ionic semiconductors. Given its high bond energy of 60 eV, wide energy band of 3.37 eV, and great mechanical and thermal stability at ambient temperature, it is a desired material for usage in optoelectronics, electronics, and laser technologies in the future [19,20].

ZnO-NPs have a high surface-to-volume ratio and are non-toxic because of their unique chemical, electrical, and optical properties, they are employed in a variety of applications (photocatalytic degradation, solar cells, metal-insulator semiconductors, light-emitting diodes, and personal care products) [21,22]. As one of the widely accepted metal oxide NPs such as ZnO nanostructure are widely used in various fields due to their unique properties [23,24]. Among metal oxides, ZnO and TiO_2 are the most studied oxides for their application in DSSCs. The possibility of obtaining ZnO with characteristics such as small crystallite size, high surface area, and presence of crystalline light-absorbing defects enables to utilize of inexpensive ZnO materials in DSSCs [25,26].

Green synthesis of ZnO involves an eco-friendly and sustainable approach to produce these nanoparticles using natural or biologically derived reducing and stabilizing agents [27,28]. Natural sources like plant extracts, fungi, bacteria, or other bioactive materials are chosen as reducing and capping agents. These agents contain bioactive compounds capable of reducing metal ions. The selected green agents are collected, cleaned, and processed to obtain an extract rich in bioactive compounds. Various techniques like maceration, solvent extraction, or grinding may be employed. Zinc precursor salts, such as zinc nitrate or zinc chloride, are dissolved in a suitable solvent to create a precursor solution. The precursor solution is mixed with the green agent extract, initiating the reduction reaction. The bioactive compounds in the extract reduce the zinc ions, leading to the formation of ZnO-NPs [1,29]. The bioactive compounds also act as capping agents, preventing particle agglomeration and stabilizing the nanoparticles. The resulting ZnO-NPs are characterized using various techniques like scanning electron microscopy (SEM) [30], transmission electron microscopy (TEM) [31], X-ray diffraction (XRD) [32], and spectroscopic methods to analyze their size, shape, and crystalline structure. The green synthesis approach offers several advantages, such as reduced environmental impact, biocompatibility, and cost-effectiveness. It avoids the use of toxic chemicals and high-energy processes commonly associated with conventional nanoparticle synthesis methods. Additionally, the process can be optimized to control the size and shape of the nanoparticles by varying the green agents and experimental conditions, allowing tailoring of the nanoparticles for specific applications in various fields like electronics, medicine, and environmental remediation [33,34].

NPs can be synthesized by physical, chemical, and green (biological) approaches. Apart from their toxic nature, chemical, and physical methods create several negative effects, including slow growth, inaccurate structures, and sluggish production rates of synthetic NPs [35, 36]. Green synthesis or biosynthesis is more affordable, safer, and more environmentally friendly. Two different grounds for synthesis are bottom-side-up and side-down procedures to obtain the desired NPs. Chemical and physical methods are mainly used to create NPs. Physical methods are thought to be costly, whereas chemical methods are considered to be detrimental to the atmosphere and living species [37, 38]. Because of many significant characteristics such as environmentally sustainable protocol through non-noxious by-products, mild reaction state criteria, safety, and usage of a green source of reducing and capping agents, biological methods have much more attention [39,40]. The NPs thus developed by green synthesis are relatively safer and stable, with far more variation in shape and size. The most commonly used biological/green technique for the manufacture of nanomaterial is the use of plant extract to formulate environmentally safe NPs, which avoids the use of harmful chemicals with poisonous effects [41,42]. Throughout this review, we will discourse the green synthesis method for ZnO nanostructures and its application in solar cells and photocatalysis.

2. Green chemistry approach

The biological or green synthesis process uses plant extrication and parts of plants (seeds, flowers, leaves, fruits), bacteria, yeast, fungi, and algae to have a Nano range of 10–100 nm of NPs. The plant extricates are applied as capping and reducing agents in the manufacturing of nanostructures [43]. Physical and chemical procedures to synthesize metal oxide nanostructures are time-consuming and sophisticated processes and use more energy while the green process is easy to process and cost-effective and pollution-free [12]. The green process has been successfully engaged in the development of ZnO nanostructures [26,44].

While greener methods would not replace all the current chemical procedures, with the onset of green chemistry carrying its twelve principles, less environmental damage may be done, researchers around the world are developing green methods for generating NPs. The green approach of nanomaterial preparation, involve the use of environmentally friendly material such as microbes and plant parts and does not include the usage of toxic chemicals [45,46]. The benefits of this green approach over the others are due to the easy availability of raw materials (which may be found in the surrounding) and large-scale production of impurity-free NPs. Using the green process, ZnO-NPs have been successfully developed. The use of natural materials in the green synthesis of ZnO-NPs is due to their phytochemical constituents which behave as capping (stabilizing) as well as reducing agents. The natural material reduces zinc (metal) to the zero-valance state and then oxide may be added to the zinc through calcination. Another very considerable route is that zinc ions in natural extract solution form complexation with phytochemicals and then through hydrolysis zinc hydroxide is formed. The complex decomposes after calcination, and eventually, the formation of ZnO-NPs takes place [26]. The use of microbes is another biological approach for the preparation of ZnO-NPs. However, this technique has certain limitations in contrast to the synthesis using plant extract. It is important to screen the microbes, which is a time-consuming procedure. Contamination is strictly prevented in this method, especially in the culture broth [47]. The entire procedure is not very lucrative compared to the synthesis in which plant extracts are involved shown in Fig. 1. The green synthesis has several advantages over other methods, Fig. 2.

2.1. Biosynthesis of ZnO-NPs using plant leaves

Melia azedarach and Cassia fistula are medicinal plants their leaf extract with zinc acetate dehydrate was employed to produce ZnO-NPs.

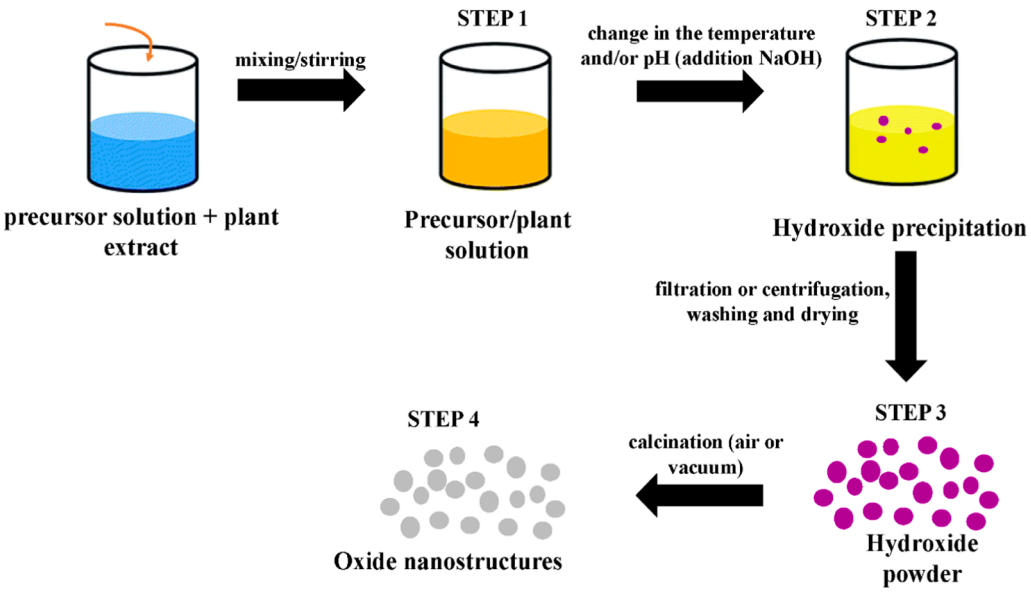

Fig. 1. Diagram explaining the step-wise general green synthesis procedure for developing nanostructures using plant extract.

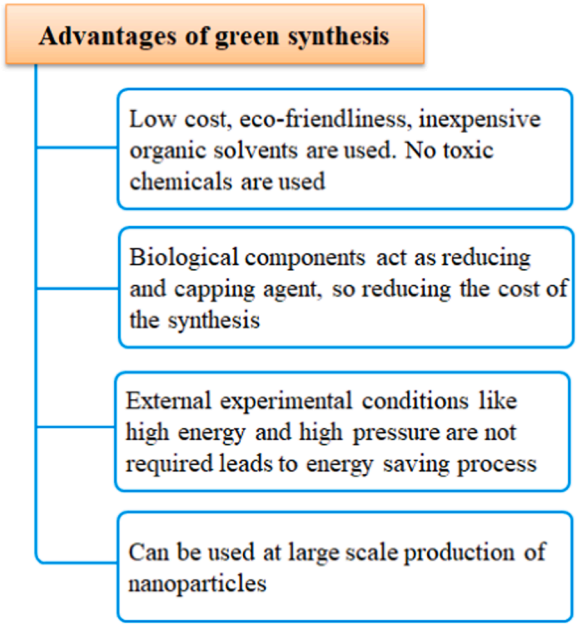

Fig. 2. The following chart indicates the advantages of the green synthesis approach.

The shape of the green prepared NP was revealed as spherical. The M. azedarach and C. fistula-mediated ZnO-NPs demonstrated good anti-microbial movement in contradiction of clinical pathogens. Indicating that plant-based green synthesis of NPs may be an outstanding strategy for producing medicinal products that are versatile and eco-friendly [48]. For the production of ZnO nanoscale particles 'L. Montana leaves' aqueous extracts were used. The antihyperglycemic and anti-oxidant properties of the prepared NPs were evaluated. By a normal particle size of 13.8 nm, the ZnO-NPs had a spherical and cubic morphology. The synthesized NPs showed outstanding antioxidant qualities. ZnO-NPs that were produced had a competitive mechanism of action on amylase, but a non-competitive one on glucosidase [49].

Fresh leaf extract of Passiflora caerulea with zinc acetate was used for the green preparation of nanoscale ZnO-NPs. The spherical shape and crystalline nature of synthesized NPs were found and their size was in the range of 30–50 nm. The anti-bacterial movement of ZnO-NPs synthesized by using Passiflora caerulea leaf extract proved these NPs as effective anti-bacterial agent in urinary tract infections [50]. For the synthesis of ZnO nanostructures, the leaves of D. caffra have been used. With an element size of 25.3 nm, ZnO-NPs possessed a hexagonal wurtzite structure. The 2, 2-Diphenyl-1-Picryl-Hydrazyl-Hydrate (DPHH) was used to test the nanostructures of ZnO for antioxidant activity. ZnO-NPs made from D. caffra leaves had less antioxidant activity than the ascorbic acid control. When used against the Michigan Cancer Foundation-7 (MCF-7) breast cancer cell line, the prepared nanostructures were tested for cytotoxicity. When compared to the usual drugs, the nanostructures of ZnO displayed marginally less cytotoxicity activity [51].

The fresh leaf of Salvia officinalis samples was thoroughly washed and soaked in deionized water (DW) for 20 min to remove any dirt present on the skin. The leaf was then air-dried completely to remove moisture. Subsequently, the dried leaf was crushed using a mortar and pestle. For each sample, 10.0 g of the powder was boiled in 100 mL of DW for 30 min. The resulting extracts were filtered using Whatman No. 41 filter paper and stored at 4 °C for future use. Next, the synthesis of ZnO-NPs was performed by aqueous extract of Salvia officinatis shown in Fig. 3. Four 4.0 g of zinc acetate was dissolved in 50 mL of DW to obtain the final concentration of zinc acetate dehydrate. Then, 10 mL of Salvia officinalis leaf extract was slowly added to the zinc acetate dehydrate solution in separate beakers. Additionally, 10 mL of their mixture was added to another beaker containing the zinc acetate dehydrate solution in the same ratio. To control the pH of the solution at 12, 1 M of NaOH was added drop by drop [52]. The mixtures were heated and stirred on a magnetic stirrer for 2 h at 50 °C until a white precipitate was observed. Afterward, the pellets were centrifuged three times at 5000 rpm at 10 min intervals and then dried in an oven overnight. The dried samples were subsequently calcined in a muffle furnace at 450 °C for 2 h, and the resulting powders were collected for further characterization.

ZnO-NPs were synthesized by using a green, easy, and environmentally safe method. This method involves the use of Mentha spicata leaf aqueous extract as a capping and reducing agent. The results exposed that the form of eco-synthesized ZnO-NP was spherical with crystalline nature and their average size was 74.7 nm. ZnO-NPs antiviral activity against Tobacco Mosaic Virus (TMV) was studied. It was revealed that disease severity and virus accumulation level was reduced to 90.2% by the application of ZnO-NPs prepared by Mentha spicata leaf

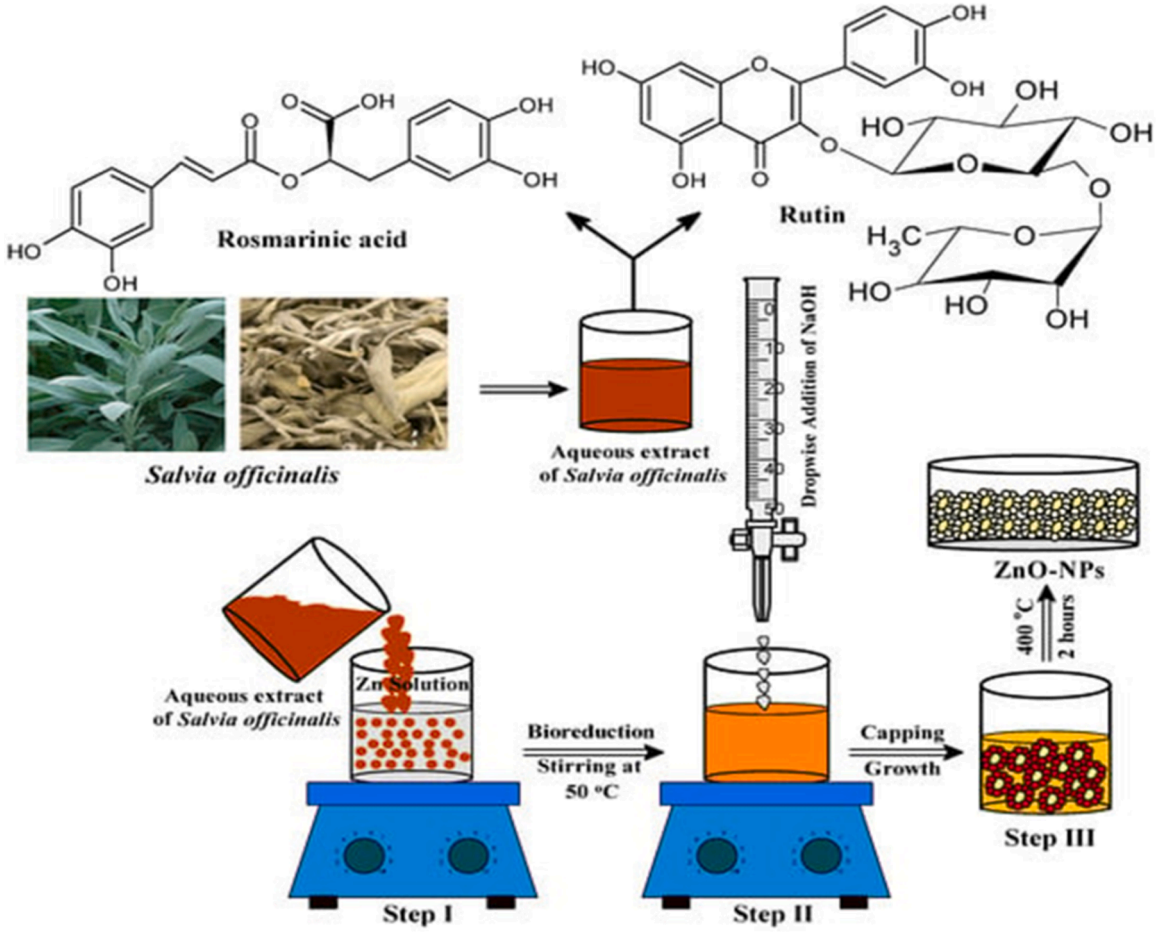

Fig. 3. The schematic representation of the synthesized ZnO-NPs using a mixture of Salvia officinalis extract.

extract. This study exposed the antiviral action of eco-synthesized NPs of ZnO and encourages the use of ZnO-NPs in the plant for the treatment of viral disease [53].

The plant Cayratia Pedata's leaf extract was employed to create ZnO nano-sized particles. Wet chemical synthesis was employed to create nanostructures of ZnO, having an average size of 52.2 nm. These NPs were then used to immobilize the enzyme glucose oxidase. The NPs showed 88.2% enzyme immobilization activity for glucose oxidase compared to native ZnO immobilization. Most of these NPs were found to have a horizontal shape, and the hexagonal nature of the NPs was discovered [54].

In traditional Asian medicine, Sambucus ebulus, which has distinct usual properties, has been believed an auspicious material. S. ebulus leaf extract was used to synthesize ZnO nanostructures. The prepared nanoparticle structure was a hexagonal wurtzite crystal structure and the average size was 17 nm. These nanostructures prepared by using Sambucus ebulus showed possible methylene blue dye pollutants photocatalytic degradation and good antibacterial activity [55]. Using a green method, ZnO nanoscale particles were made using thyme leaf extract. The NPs were spherical with a size range of 39.4–51.8 nm [56]. Eucalyptus globulus Labill is an aromatic tree of the Myrtle family. Eucalyptus globulus Labill leaf extract was utilized to synthesize ZnO nanostructures from zinc hexahydrate salt [57]. Eucalyptus globulus Labill is utilized as a useful capping and chelating agent in the preparation of nanostructures. The experimental results showed the crystalline nature of nanosized ZnO particles. Their size was in the range of 27–35 nm with a spherical shape. The measured band gap was around 2.67 eV of ZnO [33,34,58]. Aqueous leaf extract of Aquilegia pubiflora was used to make ZnO-NPs. A mean size of 34.2 nm was observed for ZnO-NPs, which were spherical or elliptical. Good anti-oxidant and anti-Alzheimer properties have been demonstrated by the synthesized ZnO-NPs. Additionally, alpha-glucosidase and alpha-amylase enzymes are moderately inhibited by ZnO-NPs. At higher concentrations, these prepared NPs demonstrated effective anti-action Alzheimer's and displayed $>65\%$ inhibition against both AChE and BChE enzymes. ZnO-NPs also have decent anti-aging properties. ZnO-NPs produced by A. pubiflora have demonstrated effective anti-inflammatory properties by inhibiting inflammation-causing enzymes [59].

An aqueous extract of Solanum torvum fresh leaves was used to prepare ZnO-NPs from a zinc nitrate aqueous solution. The shape of the nanoparticle was found to be spherical and polydispersed with some non-spherical monoclinic particles. ZnO-NP size was 34–40 nm and the mean size was 38.0 ± 2 nm. This research revealed that dermatologically applied ZnO-NPs infused hydrogel can affect renal and hepatic performance in rats and that cumulative toxicological effects have been found at exposure time [60]. "Koseret" Lippia adoensis is an endemic medicinal plant. Its leaf extract with zinc acetate dehydrates was used in a green method to synthesize ZnO nanostructures. It was indicated that this prepared nanoscale ZnO particle's structure was hexagonal wurtzite with a spherical shape. Some Nano-rod shaped ZnO-NPs were also found. The size of the NPs 22.6 nm, 18.5 nm, and 26.8 nm sizes were found dependent on the ratios of Lippia adonis extract by volume. These biosynthesized NPs revealed stability above 400 °C. The findings indicated that green synthesized NPs of ZnO showed promising antibacterial activity [61]. Two unlike zinc salts (zinc nitrate and zinc acetate) and an aqueous extract of Laurus nobilis L. leaves were used in the green preparation of NPs of ZnO. The hexagonal wurtzite structure of NPs was confirmed by X-Ray Diffraction (XRD). The shape of ZnO NPs

synthesized by zinc nitrate originated to be flower-like with an average size of 25.2 nm and ZnO-NPs synthesized by zinc acetate were bullet-like in shape with an average size of 21.5 nm. It was concluded that L. nobilis has the capacity for eco-friendly, easy, and quick synthesis of ZnO-NPs [62].

Nanostructures of ZnO were developed by using zinc acetate dehydrate and Veronica multifida leaf extract. The morphology of NPs was found to be irregular to almost hexagonal and quasi-spherical. The size was found from 10 to 100 nm antibiofilm and the antimicrobial activity of ZnO-NPs was determined. The results explained that ZnO-NPs developed by lucrative and environmentally friendly techniques could be used as coating material and in a large number of applications in different fields [63]. Chamomile flower (Matricaria chamomilla L.), red tomato fruit (Lycopersicon esculentum M.) and olive leaf (Olea europaea) plant extract were utilized to generate ZnO Nanostructures. Scanning electron microscopy analysis has shown that the size of NPs produced by Lycopersicon esculentum, Olea europaea and Matricaria chamomilla was in the range of 65.6–133, 40.5–124 and 49.8–191 nm respectively. Overall, the prepared NPs were to be an important antimicrobial agent [64]. Ficus Hispida L belongs to the Moraceae family, it has the potential as an antidiarrheal, anti-inflammatory and antiulcer drug. Aqueous leaf extract of Ficus hispida L and zinc acetate were used to develop ZnO nanorods. The formation of phase pure NPs with a wurtzite shape was confirmed. The band gap of ZnO nanorods was 3.15 eV and the average size was 50 nm [65]. ZnO-NPs were prepared by a green approach using Gloriosa superba leaves extract and zinc sulfate heptahydrate. The investigational outcomes exposed that the shape of NPs was fibril with an average size of 28 nm with a 3.22 eV bandgap was found [66]. Aqueous pandanus odoriferous leaf extract was utilized in the green preparation of ZnO nanostructures. Zinc acetate dihydrate was the precursor. The obtained NPs were very pure with a spherical shape and a 90 nm size with a 3.10 eV bandgap was calculated. The NPs showed significant optical properties. These synthesized NPs could serve as antimicrobial and anticancer agents [67]. Summary of reported data on the biosynthesis of ZnO-NPs using various plant leaf extracts, along with their respective particle sizes and shapes shown in Table 1.

Ixora coccinea leaf extract and zinc acetate were used to synthesize ZnO-NPs with spherical shape and high stability [71]. The XRD confirmed the NPs with a hexagonal wurtzite phase. The size of the prepared nanoscale particles was 145.1 nm. NPs synthesized by this method have the potential to be used in different fields such as coatings, water remediation, biotechnology, optical devices, electronics and catalysis, etc [68]. Catharanthus roseus is a wealthy source of ajmalicine, vinblastine, alkaloids, vincristine, serpentine, phenolic compounds, etc which are important secondary metabolites. Catharanthus roseus (C. roseus) leaf extract and zinc acetate dehydrate have been used in the green preparation of ZnO nanostructures. Catharanthus roseus behaved both as a tumbling as well capping agent in the synthesis. The particle size was 5–92 nm with spherical and hexagonal wurtzite phases [72]. Zinc nitrate precursors and Rumex dentatus leaf extract were utilized to create ZnO-NPs, which were then positively used as an effective antibacterial agent [73]. Zinc nitrate and aloe Vera leaf extract solution were used to obtain nanostructures of ZnO. The particle obtained were primarily spherical and the particle size could be regulated by adjusting the leaf broth solution concentration. The prepared nanostructures of ZnO were polydisperse and the average size varied from 25 to 40 nm. The eco-friendly, highly effective NPs of ZnO organized from aloe Vera broth are projected to have extra compressive uses in the cosmetic and biomedical industries [70].

Table 1
Literature reported biosynthesized ZnO-NPs using plant leaf extracts.

Leaf extract	Particles size (nm)	Shape	References
Melia azedarach, Cassia fistula leaf	3.0–68	spherical	[48]
Passiflora caerulea leaf	30–50	spherical	[50]
Mentha spicata leaf	74.7	spherical	[53]
Sambucus ebulus	17.0		[55]
Eucalyptus globulus labill leaf	27–35	spherical	[57]
Solanum torvum fresh leaves	34–40	spherical and monoclimic	[60]
Koseret lippia adoensis leaf	22.6, 18.5, 26.8	spherical	[61]
Laurus nobilis L. leaves	25.26, 21.49	flower, bullet	[62]
Veronica multifida leaf	10–100	quasi-spherical	[63]
Chamomile flower (Matricaria chamomilla L.)	65.6–133	–	[64]
Red tomato fruit (Lycopersicon esculentum M.)	40.5–124	–	[64]
Olive leave (Olea europaea)	49.8–191	–	[64]
Ficus Hispida L	50.0	nano-rods	[65]
Gloriosa superba leaves	28.0	fibril-like	[66]
Pandanus odorifer leaf	90.0	spherical	[67]
Ixora coccinea	145	spherical	[68]
Catharanthus roseus leaf	5.0–92	spherical	[69]
Aloe Vera leaf	25–40	spherical	[70]

3. Benefits of ZnO-NPs materials in DSSCs

Sunlight provides us with a pure, inexpensive and sustainable supply of energy, while also acting as a key source of energy for other energy sources, such as bioenergy, fossil fuels, water and wind [74]. However, the consumption of fossil fuels has commanded to the current rise in the emission of carbon dioxide gas, as well as global warming. The implementation of many forms of green energy is one way to solve these problems. Solar radiation, which generates extremely high-temperature heat that can fuel a mechanical engine by transforming radiation into mechanical power and electricity to initiate a machine, is the most available natural source of energy. Solar power can also be transformed by a photovoltaic effect into electricity [75]. The primary cohort of photovoltaic technology is single-junction crystal solar cells or single and multi-crystalline silicon solar cells. A thin film has been introduced by the second-generation photovoltaic technology to minimize material costs. DSSC technologies are one example of the latest third-generation photovoltaic technologies that integrate nanotechnology, double junctions and triple junctions into the solar cells manufacturing process. It is also recognized as a photoactive electrode or photoelectrochemical cell based on nanostructure metal oxide film e.g. Fe_2O_3, Nb_2O_5, ZnO, CdS, SnO_2 and TiO_2 [76,77]. Global awareness has been posed by strong indications of the effects of climate change in recent years. It calls for fossil fuel usage to be minimized while highlighting the need to build a fully sustainable and renewable energy source. Significant consideration has been drawn toward cost-effective DSSCs [78]. Due to high electron affinity, high conductivity, excellent electron mobility, and high stability, ZnO semiconductor materials have been considered in solar cell applications [79]. The benefits of ZnO material for solar cell application are represented in Fig. 4.

3.1. Operating principle of dye-sensitized solar cell

Dye-sensitized solar cell operation is based on the transformation of solar energy into electrical energy [80]. The main components of DSSC are (I) a transparent conducting oxide that typically consists of indium-doped tin oxide or fluorine-doped tin oxide. For other different cost-effective materials used as transparent conducting oxide, however, reports are available. (II) A photoanode is made up of a mesoporous metal oxide layer. (III) Sensitizer (dye) embedded into photo-anode. (IV) An electrolyte that undergoes a redox reaction. (V) Counter electrodes, usually platinum coated on glass [81,82]. An illustration of a dye-sensitized solar cell is depicted in Fig. 5.

Fig. 4. The diagram illustrates the primary benefits and characteristics of ZnO semiconductor materials for solar cells and photo-catalysis applications.

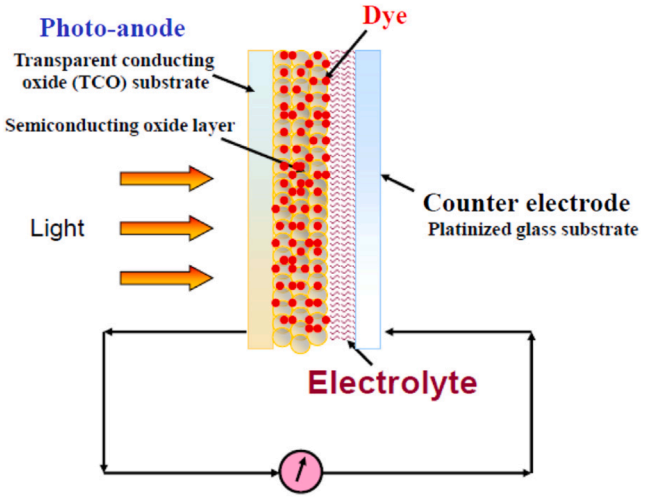

Fig. 5. The schematic diagram illustrates the main components and operating principles of a dye-sensitized solar cell.

3.2. Application of ZnO nanostructure on dye-sensitized solar cell

Zinc acetate dehydrates and Carica papaya leaf extract was used to synthesize green ZnO-NPs [83,84]. The experimental results revealed that hexagonal wurtzite NPs were obtained with ~50 nm size and spherical shape. The developed green ZnO-NPs have been used in DSSCs, as photo-anode [33,34]. By the doctor blade method, ZnO paste was coated over the FTO plate in the dye-sensitized solar cell. It demonstrates an energy conversion competence of 1.6% in DSSC with a present concentration of 8.1 mA cm^{-2} shown in Table 2. Tilia Tomentosa (Ihlamur) leaves and zinc nitrate hexahydrate was used in the biosynthesis of nanostructures of ZnO [85,86]. A single-phase hexagonal structure and spherical form of the NPs with small agglomeration was found in as-synthesized nanostructures. The average size of the above-mentioned nanoscale particles was 80 nm. ZnO nanostructure-based DSSC was manufactured successfully. The high efficacy of the processed DSSC was attributed to a major improvement in the absorption on the surface of ZnO nanostructures of the dye molecule with a 3.55 eV bandgap was calculated. This large bandgap is related to the semiconductor property that can be studied for applications. The DSSC parameters, such as short current density, efficiency, open circuit voltage and fill factor were 6.26 mA, 1.97%, 0.65 V and 48.5% respectively, at 100 mW/cm^2. The use of prepared ZnO nanosized particles in the manufacture of DSSC is therefore a promising and simple stratagem for our potential comfort [87].

Table 2
Summary of reported data on ZnO-NPs synthesized by plant extracts and their application in DSSCs.

Extract source	Power conversion efficiency (%)	Reference
Carica papaya leaf	1.60	[85]
Tilia Tomentosa (Ihlamur) leaves	1.97	[87]
Vernonia amygdalin (bitter leaf)	0.63	[88]
Amorphophallus konjac tubers	1.66	[89]
Ixora coccinea leaf	1.37	[71]
Hog plum	0.63	[90]

Vernonia amygdalin (bitter leaf) extract and zinc sulfate heptahydrate were used in the bio/green synthesis of nanostructures of ZnO. The spherical shape of developed NPs was found with granularity. The prepared nanostructures as the window layer were used in solar cells. A spin-coating machine was used to deposit ZnO-NPs on FTO-coated glass. As a source of natural dye, black cherry fruit was used [91]. Results revealed a 0.63% power conversion efficiency of NPs of ZnO [88]. An innovative technique to prepare rice-shaped ZnO nanosize particles using an extract of Amorphophallus konjac tubers with zinc acetate dehydrate has been introduced. Microwave-assisted drying method was used in nanoparticle synthesis. The 3.11 eV bandgap of NP was found through the doctor blade approach, the prepared ZnO nanostructures were coated on the FTO substrate. Using natural dyes isolated from leaves of Euphorbia pulcherrima, fruits of Terminalia catappa and flowers of Callistemon citrinus, the ZnO photoelectrodes were sensitized. It was determined that the highest efficiency was 1.66% for the solar cell manufactured using ZnO sensitized with Euphorbia pulcherrima [89]. By green synthesis, ZnO particles have been prepared using zinc acetate and Ixora Coccinea leaf extract [71]. Ixora Coccinea petal pigment and ZnO film on ITO substrate were used in DSSC with a 3.31 eV band gap of ZnO found. Results revealed that the synthesized particle size was 29 nm with a hexagonal structure. The DSSC parameters such as voltaic, fill factor and efficiency were found to be 0.461%, 0.276% and 1.367% respectively. Results demonstrated that the manufactured DSSC with green synthesized ZnO was found to target the ecologically welcoming and little price DSSC requirements [92].

The generation of ZnO nanomaterial using hog plum extract as a fuel was done by quick and economical combustion method. The synthesized NPs were used for the production of N719 DSSCs [93,94]. The 18.3 nm size of nanostructures of ZnO was found with a quasi-spherical shape. The development of strongly crystalline ZnO-NPs was owing to the presence of ascorbic acid and sugar in biofuel. This method offers a fruitful and important increase in synthesis and eventual use of high-quality nanostructures of ZnO in DSSC as a photoanode. ZnO-NPs slurry was coated on FTO glass using the doctor blade method. The power conversion proficiency of DSSC fabricated with green synthesized ZnO-NPs was found to be 0.63%. Higher surface area by decreasing particle size and therefore improving dye loading capability demonstrate an enormous surge in power conversion efficacy. With a growing understanding of renewable and hygienic energy, the green route produced ZnO-NPs and their photovoltaic efficiency was initiated to be reliable for efficient, low cost and friendly to the environment energy conversion devices [90].

4. Photocatalytic activity of ZnO-NPs

Groundwater and the environment can be accumulated with non-biodegradable dyes which are present in organic waste and wastewater of dye, paints and textiles and many other industries [95]. Various environmentally friendly, cost-effective, non-toxic, easy and bio-degradable methods have been developed by researchers. Shape, surface, size and optical activity are the function of their photocatalytic activity. ZnO nanostructures created using a biological/green approach have better photocatalytic activity [96]. Solar radiations having high photonic energy, when irradiated on ZnO-NPs causes the excitation of electrons to the empty conduction band, electron-hole pair is produced, those drift to ZnO-NPs shells and oxidation/reduction reaction occurs in

which OH^- change into hydroxyl radicals and H^+ reacts with H_2O molecule. The electrons create superoxide free radical anions when reacting with oxygen, leading to the development of hydrogen peroxide that reacts additional with superoxide radicals to form hydroxyl radicals [97]. The resulting hydroxyl radicals are powerful oxidizing agents that react to ZnO surface-adsorbed inorganic and organic contaminants to create intermediate compounds and inorganic compounds [98]. The photocatalytic activity of ZnO-NPs is represented in Fig. 6. The photodegradation rate increases with the increase in ZnO-NPs concentration. The reason for it is the structure, size and large surface area of ZnO-NPs, Photo-catalyst ZnO and UV light, both are necessary for dye degradation [99].

4.1. Application of ZnO nanostructure in photocatalysis

ZnO nanostructures have shown remarkable potential as photocatalysts due to their unique properties, such as a wide bandgap and high surface area. When illuminated with light, ZnO nanostructures generate electron-hole pairs, which can participate in redox reactions with organic pollutants or water molecules, leading to the degradation of contaminants and the production of reactive oxygen species. This photocatalytic activity finds applications in various fields, including water purification, air pollution control, and self-cleaning surfaces. Garcinia mangostana (G. mangostana) fruit pericarp aqueous extract was utilized for the preparation of the nanomaterial of ZnO [100,101]. The findings revealed that high-purity ZnO-NPs were prepared, mainly spherical with a mean size of 21 nm. Under solar irradiation, the photocatalytic activity of green synthesized NP was measured by malachite green dye degradation. One of the main classes of harmful dyes is a malachite green dye, which is a cationic triphenylmethane dye. Within 180 min of irradiation, approximately 99% of the malachite green dye was depleted. Pseudo-first order was found in malachite green dye degradation. The results revealed that due to the chromosphere group, the malachite green dye has a strong absorption at 615 nm. Malachite green dye was effectively degraded by ZnO-NPs into less damaging products [102]. As a reducing agent, the root extract of Scutellaria baicalensis was used in the biosynthesis of ZnO-NPs using the one-pot method. Sphere-like NPs with 50 nm size were obtained. Under UV light irradiation, antimony ZnO-NPs were used in photocatalytic methylene blue degradation. The degradation rate constant of methylene blue with 0.05 mg mL^{-1} antimony ZnO-NPs (photocatalyst) was measured at 0.016 min^{-1} and 98.6% degradation was achieved in 210 min. Antimony ZnO-NPs are capable of being a photocatalysis reduction agent [103]. Literature reported ZnO nanostructure and their application for dye degradation was shown in Table 3.

The photocatalytic activity, antibacterial and antioxidant of ZnO-NPs made from cocos nucifera leaf extract have been effectively examined. With 16.6 nm as the average particle size, the NPs' structure was hexagonal wurtzite. ZnO-NPs photocatalytic behavior resulted in absorbance degradation at 640 nm and dye discoloration in methylene blue after 1.0 h, with a maximum degradation rate of 84.3% [117]. With the help of microwave-assisted extraction techniques, nanostructures of ZnO were prepared using C. edulis fruit extract. The average size of 50–55 nm, hexagonal wurtzite structure, and flower-like shape of as-synthesized ZnO-NPs was obtained. The photocatalytic degradation of Congo red dye was done with the help of a prepared ZnO nanostructure. The 15 min was starting time of photocatalytic degradation and 130 min was the optimum time for 97% removal of congo red dye was achieved. Degradation followed the first order with a rate constant of 0.497 [104]. Coriandrum sativum leaf extract with zinc acetate was used in the biosynthesis of ZnO-NPs 9–18 nm size was obtained. Degradation of anthracene was done using biosynthesized ZnO photocatalyst. Anthracene degradation of 96% was achieved in a 240 min irradiation period, pH 7.0 and loading catalyst dose of 1000 μg L^{-1}. The kinetic studies showed that photocatalytic degradation was pseudo-first order. It was suggested that the nanostructure of ZnO generated by corrigendum sativum plant extract are effective photocatalyst in anthracene degradation [105]. ZnO-NPs were obtained from Nephelium lappaceum (Rambutan) fruit peel extract. ZnO-NPs with a diameter of 25–40 nm, size of 25.7 nm and spherical shape were obtained in green synthesis. Photocatalytic activity of green synthesized ZnO-NPs for methyl orange was observed under sunlight. The decolorization efficiency was found to be 84.0% in 120 min [106].

Aqueous leaf extract of Dolichos Lab L was used in a one-pot method to produce NPs of ZnO. The XRD confirmed the hexagonal wurtzite structure of phase pure ZnO-NPs [118,119]. The average particle diameter of the as-synthesized ZnO-NP was 29 nm. The semiconducting behavior of the ZnO nanostructure was due to the 3.4 eV bandgap. In the photodegradation of orange II, methylene blue and rhodamine B under visible and near-UV irradiation, as-synthesized ZnO nanostructures have been used as a catalyst. The findings revealed that the maximum photodegradation efficiency of ZnO nanostructures at pH 5.0, 9.0 and 11.0 were 66.0% for orange II, 95.0% for rhodamine B and 80.0% for methylene blue, respectively. The highest competence was attained in 210 min [107,118]. Saffron leaf extract was used as stabilizing and reducing agent in the preparation of ZnO nanostructures. The investigational outcomes have revealed that synthesized ZnO-NPs were spherical with less than 50 nm diameter and had hexagonal wurtzite structure. The photocatalytic activity, for methylene blue dye degradation in an aqueous solution, of as-synthesized ZnO-NP was carried out. With an initial concentration of 12.0 mg mL^{-1} of ZnO-NPs under UV light at 200 min, the highest methylene blue removal was 64.0%. As a promising dye removal tool, the prepared ZnO nanomaterials can be used, particularly for wastes released by the cosmetics, textile, paper and pharmaceutical industries [108]. Using microalgae chlorella and zinc nitrate, the ZnO-NPs were synthesized easily. The results showed that ZnO-NPs have a hexagonal shape with an average particle size of 20.0 ± 2.20 nm. Under UV light irradiation, the as-synthesized green

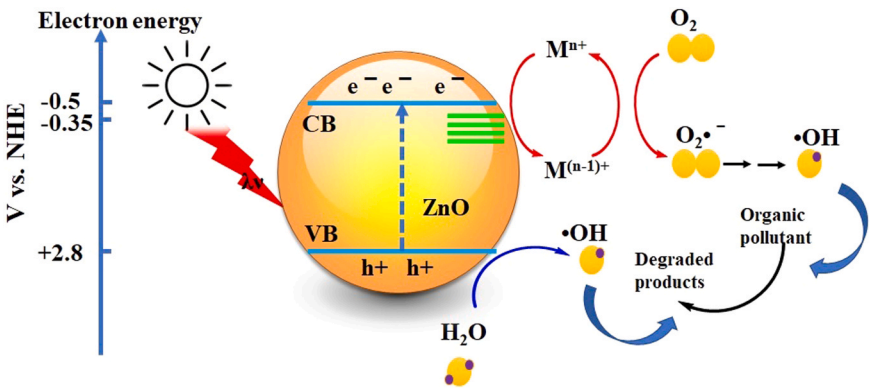

Fig. 6. Energy band mechanism for the photocatalytic activity of ZnO-NPs in an aqueous medium.

Table 3
Summarized data from various extract sources on ZnO nanostructure and their applications in photocatalysis.

Plant extract	Dye	Dye degradation (%)	Degradation time (min)	References
Garcinia mangos tana (G. mangos tana) fruit pericarp	malachite green dye	99.0	180	[102]
Scutellarin baicalinase root	methylene blue dye	98.6	210	[103]
9 C. edulis fruit	congo red dye	97.0	130	[104]
Coriandrum sativum leaf	anthracene	96.0	240	[105]
Nephilim lappaceum (Rambutan) fruit peel	methyl orange	84.0	120	[106]
Dolichos Lablab L leaf	orange II, methylene blue and rhodamine B	66.0, 80.0, 95.0	210	[107]
Saffron leaf extract	methylene blue	64.0	200	[108]
microalgae chlorella	Dibenzothiophene	97.0	180	[109]
C. au-rantifolia (lemon), L. esculentum (tomato) and C. sinensis (orange) peels	methylene eblue	96.0, 77.0, 97.0	180	[110]
Azadirachta indica (Neem) leaf	methylene blue	82.0	180	[111]
Tabernaemontana divaricata green leaf	methylene blue	100	90	[112]
A. carnosus leaf	methylene blue			[113]
Kalanchoe blossfeldiana	Eosin Y, methylene blue	94.0, 84.0		[114]
P. amboinicus leaf	methyl red dye	92.4	180	[96]
Eucalyptus globulus leaf	methyl orange, methylene blue	96.6	60	[115]
Broccoli	phenol red and methylene blue	100	180	[116]

ZnO-NPs were used in photocatalytic degradation of Dibenzothiophene contaminant at mild conditions and neutral pH. Within 3 h, a 97.0% maximum removal efficiency photocatalyst was achieved at an optimum concentration of 0.01 mg L^{-1} of ZnO-NPs [109].

Several phytochemicals are present in fruits that can assist in the preparation of NPs. Peels of C. au-rantifolia (lemon), L. esculentum (tomato) and C. sinensis (orange) were to prepare the extract for NPs synthesis [120,121]. The extract behaved as a capping and stabilizing agent in NPs synthesis. An average size equal to 19.7 nm with a polyhedral shape was found in the bio-inspired ZnO nanoparticle. Strong antibacterial activity and photocatalytic activity were demonstrated by biosynthesized ZnO-NPs. photocatalytic degradation of methylene blue with biosynthesized ZnO-NP was carried out under UV light. The findings demonstrated that after 180 min, NPs made with tomato extract exhibited a maximum of 97.0% degradation as compared to NPs prepared with orange and lemon extract which showed 77.0% and 96.0% degradation, respectively [110]. Using 25% (W/V) of Azadirachta indica (Neem) leaf extract, green preparation of ZnO-NPs was done. Pure NPs with sizes ranging from 9.60 to 25.5 nm and spherical shapes were obtained. The 3.87 eV bandgap and hexagonal wurtzite structure were found in ZnO-NPs. Photocatalytic activity under UV radiation has been shown by green synthesized ZnO-NPs to increase the photodegradation of methylene blue, which is considered to be a key aquatic contaminant produced by the textile industry. About 82.0% degradation of methylene blue was achieved by NPs after 180 min. It was concluded that NPs of ZnO may be used in environmental remediation as an important catalyst [111,122].

Aqueous Tabernaemontana divaricata green leaf extract was used to develop ZnO nanostructures. The outcomes revealed the spherical shape, hexagonal wurtzite structure and 20–50 nm size of as-synthesized NPs of ZnO. In the presence of solar radiation, methylene blue dye degradation was carried out using synthesized ZnO-NPs as a photocatalyst. Within 90 min complete degradation was achieved. Biosynthesized ZnO-NPs are very helpful for bacterial decontamination and removal of dye [112]. Zinc nitrate and leaf extract of A. carnosus were used to synthesize ZnO-NPs. The nanoparticle size was 30–40 nm with a quasi-spherical shape. The biosynthesized zinc NPs were used in the photodegradation of methylene blue dye degradation under UV-Vis light. Methylene blue dye color was transformed from blue to colorless due to photocatalytic degradation by ZnO-NPs [113].

An aqueous extract of Kalanchoe blossfeldiana was used as a reducing agent in the green preparation of nanostructures of ZnO. The average size of obtained particles was 94.4 nm with a hexagonal wurtzite phase. As-synthesized ZnO-NPs were used as a photocatalyst in Eosin Y and methylene blue degradation. Under UV radiation, the maximum degradation of Eosin Y was 94.0% and methylene blue degradation was 84.0% which showed that green ZnO-NPs are efficient photocatalysts [114]. ZnO nanostructures were developed by using P. amboinicus leaf extract. The prepared nanoparticle shape was rod-like with 88 nm size and 3.07 eV bandgap. Methyl red dye degradation was done by using as-synthesized NPs. It was revealed that within 180 min, 92.4% degradation of methyl red was achieved these values showed that degradation followed first-order kinetics. Four cycles of photodegradation tested the reusability of the biosynthesized NPs of ZnO. In the 4th cycle of photodegradation, the ZnO-NPs photocatalyst (developed by the green approach) retains more than 85.0% efficiency, suggesting that the proposed photocatalyst offers excellent stability [96,123]. A green approach for the preparation of nanostructures of ZnO was executed by using Eucalyptus globulus leaf extract. The average size of NP was 11.6 nm these nanosized ZnO particles were used as a photocatalyst in methyl orange and methylene blue degradation. Underneath UV light, 30 mg doses of NPs showed 98.3% removal efficiency. Within 60 min 96.6%, of methyl orange degradation was achieved. Rate constant $k = 0.2997\ min^{-1}$ and $k = 0.264\ min^{-1}$ obtained values for methylene blue and methyl orange, respectively, followed first order [115].

Biosynthesis of ZnO structures was done by using broccoli extract aqueous solution [124,125]. The bandgap of NP was 4.09 of hexagonal phase revealed. By testing its photo-enhanced catalytic activity against phenol red and methylene blue under UV light, the photocatalytic activity of ZnO-NPs was assessed. Degradation of phenol red and methylene blue was achieved at 71.0% and 74.0% respectively by using green synthesized ZnO-NPs as photocatalysts [123,126]. Within 180 min, the change of dye colors also revealed the complete degradation of dyes. Pseudo-first order of methylene blue degradation was detected and the rate constant was found to be 0.0073. For the photodegradation of phenol red first order was observed with a rate constant of 0.006. The best absorption rate was demonstrated by correlation coefficients in both cases [116].

5. Challenges and their potential solutions

The main challenges are biosynthesis methods can yield varying sizes and shapes of ZnO nanostructures, leading to inconsistent optical and electronic properties. Achieving control over synthesis parameters is essential for reliable and reproducible performance in solar cells and photocatalysis. Biosynthesized ZnO nanostructures may have lower photocatalytic efficiency compared to chemically synthesized ones [127,128]. Improving efficiency through better synthesis techniques and structural optimization is critical for successful photocatalytic

applications. In DSSCs, achieving efficient charge transport and minimizing recombination losses is challenging with biosynthesized ZnO nanostructures [129,130]. Strategies like surface engineering and heterojunction formation can be explored to enhance charge carrier dynamics. The stability of biosynthesized ZnO nanostructures under prolonged exposure to light and harsh environments is crucial for their practical applications. Ensuring their long-term stability is essential for sustained efficiency in solar cells and photocatalysis. Biosynthesized ZnO nanostructures might present challenges in achieving efficient dye sensitization due to their specific surface properties. Enhancing dye adsorption and compatibility is vital for improving the performance of DSSCs. Before using biosynthesized ZnO nanostructures in biomedical applications or environmental photocatalysis, comprehensive toxicity assessments are necessary to ensure they are biocompatible and pose no harm to living organisms. The biosynthesis of nanostructures to meet industrial production demands while maintaining quality and cost-effectiveness is a significant challenge for their widespread application in solar cells and photocatalysis [36,131–134]. A comprehensive understanding of the underlying mechanisms of photocatalysis involving biosynthesized ZnO nanostructures is essential for targeted improvements in their photocatalytic performance. Integrating biosynthesized ZnO nanostructures with other components in DSSCs and photocatalytic systems, such as electrolytes and counter electrodes, requires careful optimization and compatibility studies. Assessing the environmental impact of using biosynthesized ZnO nanostructures and their potential fate in ecosystems is crucial for ensuring their sustainability in applications like photocatalysis for water treatment. Addressing these challenges will require interdisciplinary research efforts and collaboration between material scientists, biotechnologists, chemists, and engineers. By overcoming these obstacles, biosynthesized zinc oxide nanostructures can contribute to the development of efficient, environmentally friendly, and sustainable technologies for solar energy conversion and environmental remediation.

6. Conclusion and future perspectives

Zinc oxide nanoparticles (ZnO-NPs) synthesized through green approaches offer a harmless, simpler, and cost-effective alternative to chemical and physical methods. The use of plant extracts as reducing and stabilizing agents in the green synthesis of ZnO-NPs has gained significant attention, making them versatile and applicable in various fields. The photocatalytic ability and unique features of ZnO-NPs make them valuable for the degradation of harmful chemicals and dyes in water. The photocatalytic efficiency of ZnO-NPs can be further optimized for potential applications in the eradication of heavy metals and wastewater treatment. The review emphasized the potential of ZnO-NPs as photocatalysts and discussed related mechanisms. In DSSCs, ZnO nanomaterials developed through green approaches show promise as a superior technique. Although DSSCs currently exhibit lower efficiency compared to silicon solar cells, they are gaining attention due to their easy fabrication, cost-effectiveness, and efficiency. Further research on green synthesis techniques for ZnO nanoparticles is crucial to explore new ideas and enhance the power conversion efficiency of ZnO nanostructures in DSSCs.

Future Perspectives: Further advancements in green synthesis methods will lead to better control over ZnO-NPs' properties and functionalities, enabling tailor-made nanomaterials for specific applications. Continued research in nanostructure engineering will help enhance the photocatalytic efficiency and performance of ZnO-NPs, making them more effective in water purification and wastewater treatment. Investigating the biocompatibility and potential biomedical applications of green-synthesized ZnO-NPs will expand their utility in areas like drug delivery and medical imaging. Exploring the use of ZnO-NPs in environmental remediation such as air pollution control and soil decontamination, will contribute to sustainable solutions for environmental challenges. Investigating the integration of ZnO-NPs with other nanomaterials in hybrid nanocomposites can create multifunctional materials with improved properties for various applications. Research on the potential toxicity and environmental impact of ZnO-NPs will aid in establishing safety guidelines and regulations for their widespread use. Scaling up the green synthesis of ZnO-NPs and optimizing production processes will be essential for their commercial viability and industrial adoption. Increasing awareness of green nanotechnology and its benefits will foster public acceptance and support for the use of eco-friendly nanomaterials in various applications. Exploring the potential of ZnO-NPs in energy storage devices, such as supercapacitors and batteries, can contribute to the development of sustainable energy storage solutions. Encouraging collaboration between researchers, industries, and policymakers will accelerate the development and implementation of green-synthesized ZnO-NPs in real-world applications. In conclusion, green-synthesized ZnO-NPs hold great promise for sustainable and eco-friendly applications in various fields, including DSSCs and photocatalysis. Continued research and collaboration will drive innovation, leading to the widespread adoption of these green nanomaterials for a cleaner and more sustainable future.

CRediT authorship contribution statement

Jamshed Ali: Conceptualization, Methodology, Validation, Writing - original draft. **Safia Bibi:** Methodology, Investigation, Writing - original draft. **Wahid Bux Jatoi:** Conceptualization, Data curation, Validation. **Mustafa Tuzen:** Conceptualization, Data curation, Investigation, Supervision. **Mushtaque Ahmed Jakhrani:** Investigation, Visualization, Writing - review & editing. **Xinbin Feng:** Validation, Data curation, Resources, Supervision, Writing - review & editing. **Tawfik A. Saleh:** Resources, Supervision, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

No data was used for the research described in the article.

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